NBS SPECIAL PUBLICATION 556

U.S. DEPARTMENT OF COMMERCE / National Bureau of Standards

Measurements and Standards for Recycled Oil - II

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Measurements and Standards for Recycled Oil - II

Proceedings of a conference held
at the National Bureau of Standards
Gaithersburg, Maryland
November 29 and 30, 1977

Donald A. Becker, Editor
Helen Anne Hurd, Associate Editor

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Foreword

The Recycled Oil Program of the Office of Recycled Materials, National Measurement Laboratory, National Bureau of Standards (NBS) was formed in 1976 in direct response to the Energy Policy and Conservation Act of 1975 [P.L.94-163, Section 383; 42 U.S.C. 6363(c)]. The goal of this program, as described in this legislation, is to develop test procedures for the determination of substantial equivalency of re-refined or otherwise processed used oil with new oil for various end uses. This legislative responsibility is appropriate for NBS because it involves both the development and evaluation of measurement methods and standards, and because it requires the independent third-party role for acceptance by all concerned parties (virgin oil refining industry, oil re-refining industry, user industries, and government regulators).

During the year since our first conference, there have been many new developments in the area of oil recycling. An important new development was passage of the Resource Conservation and Recovery Act (P.L. 94-580). The implications of this law for Federal government specifications and procurement of recycled oil are significant. Also, numerous state laws have been enacted on the subject of oil recycling. Inherent in these new developments, however, is the continued strong need for reliable test procedures and standards, to ensure adequate performance criteria and encourage consumer acceptance for recycled oil products.

This volume contains the proceedings of the second Conference on Measurements and Standards for Recycled Oil held at NBS on November 29 and 30, 1977, almost exactly one year after the first conference. As stated above, there were many new developments in oil recycling during this past year, and these proceedings contain information on most of them. In addition to the presentations, these Conferences provide the very valuable service of bringing together the many persons interested in recycled oil from different sectors, public and private, to discuss the new developments, the problems, and hopefully, some of the solutions. These proceedings should be of value to the many people and organizations who are interested and involved in encouraging oil recycling.

John D. Hoffman
Director
National Measurement Laboratory
National Bureau of Standards

Preface

On December 22, 1975, President Ford signed into law an act passed by the Congress of the United States entitled "The Energy Policy and Conservation Act of 1975" (Public Law 94-163). Section 383 of this act states that the National Bureau of Standards (NBS) shall develop test procedures "...for the determination of substantial equivalency of re-refined or otherwise processed used oil...with new oil for a particular end use." Further, NBS is to "...report such procedures to the [Federal Trade] Commission..." as soon as practicable. This NBS conference, held on November 29 and 30, 1977, is part of the continuing NBS response to this legislation.

This conference had three important objectives. The first objective was to report on the NBS efforts to date on Phase I of the NBS program, test procedures for used oil recycled as fuel. Associated with this objective were reports by others on non-lubricating end uses of recycled oil. The second objective of this conference was to discuss topics concerned with Phase II of the NBS Recycled Oil Program, which is test procedures for used oil recycled as engine oils. Presentations on this subject of engine oils included a panel discussion on the question of test procedures, for the "substantial equivalency" of re-refined motor oils to virgin motor oils. Finally, the third objective of this conference, and a very important one, is to bring together the many people involved with all the various aspects of recycled oil.

There was a total of 171 attendees at the meeting, with a breakdown as follows: 44 government representatives (Federal Trade Commission, Environmental Protection Agency, Department of Defense, Department of Energy, General Services Administration, ten State and Province governments, Department of Commerce, Tennessee Valley Authority, and NBS); 115 representatives from private industries (engine manufacturers--13, petroleum refiners and additive manufacturers--27, used oil re-refiners and processors--48, and other industry--8); and 17 attendees who were consultants or who represented consulting firms, testing laboratories, or non-profit organizations. Of the attendees described above, 12 were from foreign countries, primarily Canada, but also from France, Israel, and Columbia, S.A.

It was felt that the conference participants made a very important contribution to the available knowledge and understanding of the problems and opportunities in recycling. We plan to hold another conference on the testing of recycled oil in the fall of 1979.

Donald A. Becker
Manager, Recycled Oil Program

Abstract

This publication is a formal report of the second Conference on Measurements and Standards for Recycled Oil, held at the National Bureau of Standards on November 29 and 30, 1977. There were seven sessions on specific subject areas, with a total of 32 presentations. The subject areas were as follows: (I) The Existing Situation, (II) Environmental Considerations, (III) Nonlubricating End Uses, (IV) Legislation and Studies, (V) Evaluation of Re-refined Engine Oils, (VI) Additional Recycled Oil Activities, and (VII) A panel discussion on the topic: "What Data Base is Required to Establish the 'Substantial Equivalency' of Re-refined Motor Oils to Virgin Motor Oils." Included in this report are invited talks given and a summary of the discussion following session VII.

Key Words: Engine oil; fuel oil; hydraulic oil; industrial oil; lubricating oil; oil specifications; petroleum standards; petroleum test methods; recycled oil; re-refined oil; used oil; waste oil.

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WELCOMING REMARKS

E. Ambler

Director
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Good morning, and welcome to the second conference on recycled oil. Last year, at the first conference, we had 69 registrants; today there are over 170. To me, this growth is a strong indication of the increasing interest in recycled oil.

Recycled oil reminds me of those immortal words of Coleridge about water, water, everywhere. We have used oil everywhere--in factories, filling stations, and in the backyards of do-it-yourself auto mechanics. What we need is the wisdom and the will to make use of this ready resource.

This situation is like the young man who was complaining after his first day as a piano mover. The boss said, "When I told you you could have the job, I never said it was going to be easy." Well, we, too, have a job. As you know, the Energy Policy and Conservation Act of 1975 gave the National Bureau of Standards (NBS) the task of developing test procedures for evaluating re-refined oil. We never thought this job was going to be easy, and it isn't. There are some formidable technical problems involved, problems that are now being studied in our laboratories. As you will hear in detail later, we are making substantial progress and will forward test procedures for recycled oil used as burner fuel to the Federal Trade Commission next spring. This is just one aspect of our program, as there are many other fundamental questions to be answered.

One of the more pressing of these questions is that of the data base required to establish substantial equivalence of re-refined to virgin motor oil. This, like most of the questions concerning re-refined oil, is not one that the NBS can answer by itself. We have convened this meeting not only to let you know what we have accomplished during the year, but also to ask your assessment of our work and your help in determining what needs should be addressed.

We at the NBS take our work on recycled oil very seriously. As I said, it is a tough technical problem, and we do enjoy a challenge. The development of test procedures will pave the way for substantial reuse of oil, which will help preserve the environment, conserve a natural resource, and save money. No one can argue with those benefits.

But there is another less concrete benefit that will result from greater recycling of oil. And that benefit falls in the area of public attitude. We have been a nation of prodigal sons--and daughters--using resources with no thought for tomorrow. Turning this attitude around will be difficult, no question about it. But one way to do it is through programs such as recycled oil that have a sound technical basis and that permit widespread participation. Again, this is not something that the NBS can do alone, but it is an important outgrowth of our mutual efforts.

Thanks again for coming here today, and we look forward to the contributions you will make.

SESSION I. THE EXISTING SITUATION

National Bureau of Standards Special Publication 556. Proceedings of a Workshop on Measurements and Standards for Recycled Oil - II held at NBS, Gaithersburg, Maryland, November 29 and 30, 1977. (Issued September 1979)

CONFERENCE OBJECTIVES AND THE NBS RECYCLED OIL PROGRAM

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In my presentation this morning I would first like to describe the objectives of this second National Bureau of Standards (NBS) Conference on Measurements and Standards for Recycled Oil, and then I would like to briefly describe the NBS Recycled Oil Program and its efforts with regard to test procedures for recycled oil.

This conference has three important objectives. The first objective is to report on the NBS efforts to date. As most of you know, Phase I of the NBS program is on test procedures for used oil recycled as fuel. In addition to the NBS work, which will be discussed in detail later this morning, there are a number of people from other organizations who will discuss programs they are involved with on non-lubricating end uses for used oil, including using it as fuel. The second objective of this conference is to discuss topics and plans of interest for Phase II of the NBS Recycled Oil Program, which is test procedures for used oil recycled as engine oils. On this subject, the conference will contain presentations on recent legislation and studies, reports on programs to examine various aspects of the recycling processes and products, and a panel discussion on the topic of "substantial equivalency." Finally, a very important objective of this conference that should not be underestimated is to bring together in one place the people who are involved with all the various aspects of lubricating oil: the producers, the testers, the users, the recyclers, and the regulators. I feel these are very substantial objectives to expect from any meeting, and I hope that tomorrow afternoon at 1:00 p.m. all of you will feel that these objectives have at least partially been met.

I will now briefly discuss the NBS responsibilities in the area of oil recycling. I must apologize to those of you who are already familiar with what I am about to say. However, I feel that with the many new faces in front of me it is important that all of us fully understand the motivation and concern which the NBS has on this subject.

There are three areas of legislation which affect the NBS Recycled Oil Program. The initial NBS responsibilities were set forth in the Energy Policy and Conservation Act of 1975 [Public Law 94-163, Section 383(c); 42 U.S.C. 6363(c)]. The Resource Conservation and Recovery Act of 1976 (Public Law 94-580; 42 U.S.C. 6901 et seq.), in section 6002, established a deadline for the initiation of mandatory Federal procurement of recycled oil, and also mandated the revision of exclusionary clauses in U.S. Government procurement specifications. Finally, the Model (States) Used Oil Recycling Act, sponsored by the Federal Energy Administration (now the Department of Energy), has been introduced into a number of State legislatures. I will briefly discuss each of these areas of legislation and how they affect the NBS Recycled Oil Program, which is addressing the problem of test procedures and standards for recycled oil.

In the Energy Policy and Conservation Act, the purposes of the recycled oil section were stated as (1) to encourage recycling of used oil, (2) to promote the use of recycled oil, (3) to reduce the consumption of new oil, and (4) to reduce the environmental hazards from disposal of used oil. It is well known that there are serious environmental problems due to improper disposal practices with used oil, including widespread contamination by the high heavy-metal concentrations found in used oil, by the polynuclear aromatic content of used oil, and by the basic petroleum hydrocarbons themselves.

This act specifically directs the NBS to develop test procedures which can be used to establish the "substantial equivalency" of recycled oil products with virgin oil products. These tests are to be developed for all end uses, and each set of test procedures for a separate end-use category will be transmitted as a package to the Federal Trade Commission (FTC), which will utilize them for development of trade regulation rules.

In the Resource Conservation and Recovery Act, the stated purposes are essentially the same as for the first act. Section 6002 of this act will require government agencies to purchase recycled oils in preference to virgin oils, and to even possibly pay a higher price for it. This becomes effective on October 22, 1978, less than 11 months from now.

In addition, the review of government specifications for petroleum products must take place by April 22, 1978, approximately 5 months from now, and all exclusionary clauses must be revised to allow the use of recycled oils. For example, there is a clause in the present Federal procurement specification for burner fuel oil (VV-F-815C) which excludes fuel oil containing waste crankcase oil, even if it is processed to meet all other requirements. Also, military specifications for engine oils contain clauses excluding all lubricating oils made from recycled or previously used oils. These military specifications are used for essentially all Federal procurement of engine oils, and they often form the basis for procurement by industry and State and local governments.

The third area of legislation of interest is the Model (States) Used Oil Recycling Act. This model act was written by Mr. William Irwin (one of our speakers) for the Federal Energy Administration (now the Department of Energy), and was designed to assist individual States in establishing control over used oil disposal. It clearly encourages used oil recycling as a superior method of disposal. This model act makes use of the Energy Policy and Conservation Act-mandated NBS test procedures for establishing appropriate quality criteria for recycled oil products. A number of States have introduced such legislation, based almost word-for-word on the model act. California has just recently become the first State to pass such a law and put it into effect (California Senate Bill No. 68, signed September 1977).

The legislation just discussed will have four basic consequences in the real world. These are:

1. The FTC must change their labeling requirements for all recycled oils which are found to be "substantially equivalent" to virgin oils, using the NBS test procedures.

2. Federal agencies must review and revise specifications for petroleum products where necessary.

3. Federal agencies must purchase the highest percentage of recycled materials practicable.

4. Finally, in light of the above, it is a reasonable conclusion that the collection and recycling of used oil will be substantially increased.

In 1975 the Environmental Protection Agency estimated the amounts of recoverable used oil potentially available for recycling to total approximately 1.2 billion gallons, or over 28 million barrels. Of this amount, almost 55 percent was generated from automotive or vehicular uses. The fate of used oils in 1975 included approximately 47 percent burned as fuel--often without suitable emission controls--and apparently less than 10 percent was re-refined for reuse as lubricants. The fate of 35 percent of this used oil was stated to be unknown, and much of this is assumed to be disposed of by ecologically harmful methods such as dumping into ditches, landfills, and storm sewers by do-it-yourself mechanics.

In order for the NBS program to systematically consider the primary end uses of recycled oils, we have divided the work into four phases based on these end-use categories: Phase I--Recycled Oils Used as Fuel, Phase II--Recycled Oil Used as Engine Oils, Phase III--Recycled Oil Used as Industrial Oils, and Phase IV--Recycled Oil Used as Hydraulic Oils. Each type of oil has different specifications and performance requirements, based on a variety of test procedures. We recognize that many test procedures currently exist for virgin petroleum oils. However, we also know that at least some of these tests either are not valid with recycled oils, or that insufficient data exist to determine whether they are valid. Laboratory evaluation of these tests is required; and, in some cases, development of new or modified test procedures is necessary before a complete set of tests can be transmitted to the FTC.

We can briefly summarize our program plan as follows. For each end-use category we will

1. review existing (virgin oil) test procedures;

2. evaluate these tests for applicability and validity for recycled oil products;

3. develop new or revised test procedures when required;

4. transmit the set of approved test procedures to the FTC; and

5. provide technical support to the FTC as required.

The problems we have found with existing virgin oil tests and our technical efforts to date will be discussed in detail by Dr. J. J. Comeford later on this morning, so I will not go into them here.

Some of the progress we have made to date includes

1. setting up a petroleum testing laboratory;

2. holding the first NBS workshop in November 1976 (proceedings published as NBS Special Publication 488);

3. integrating cooperative interactions with the many committees and organizations interested in oil recycling; and

4. obtaining an Industrial Research Associate agreement with the Association of Petroleum Re-refiners (ARP). The Research Associate with the NBS Recycled Oil Program is Mr. Robert F. Pedall, Chief Chemist of the Motor Oils Refining Company in McCook, Illinois. The APR, in their October meeting, voted to continue their support of the Research Associate Program through the motor oils phase.

Finally, let me take several minutes to describe our priorities in the technical work in which the NBS is currently involved. As I mentioned earlier in my talk, we have initiated our program by considering the end use of burning used oil as fuel for energy recovery (Phase I). For this category, some examples of characteristics we are addressing for substantial equivalency are heat content (Btu); effects on the environment and health; burner erosion; tank, pipe, and burner corrosion (including effects on refractory materials); deposit formation; and so on. We expect to complete the technical portion of Phase I, Test Procedures for Recycled Oil Used as Fuel, early in 1978.

Once the fuel category is completed, the second end-use category to be addressed by the NBS program is engine oils (Phase II). As I am sure you realize, this second category contains the most difficult technical questions and problems of all the end uses. Some of these questions include the following:

1. For the straight mineral lubrication oil (API SA classification), which at the present has no performance tests or requirements--to what should recycled oils be equivalent?

2. How can the problem of obsolete engine sequence tests be resolved (API classifications SB, SC, SD, CA, CB, and CC)?

3. What is the variability, if any, of re-refined lube oil basestocks on a batch-to-batch basis, or as a function of time? What tests are capable of detecting that variability if it does occur?

4. What is the variability of petroleum oil products in the marketplace, both virgin and recycled?

5. Do recycled oils require additional or revised specifications compared to virgin oils?

As I stated before, we realize that there are some difficult technical problems which lie ahead of this NBS program. However, all of the information we have obtained, and that includes some of what you will be hearing this afternoon, indicates the following.

1. The basic hydrocarbon structure of petroleum lubricating oil is not significantly changed during use;

2. An adequately re-refined oil can be as good as a high-quality virgin lubricating oil; and finally

3. I think there is no question that oil recycling is here to stay.

I am looking forward to the papers to be presented here in the next 2 days, and I hope that the discussions and comments on the various topics will be both numerous and constructive. I also invite your comments and cooperation with the NBS program and with our efforts to help resolve these technical questions and problems.

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THE CURRENT STATUS OF THE RE-REFINING INDUSTRY

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It is a pleasure for me to speak to you this morning on behalf of the Association of Petroleum Re-refiners (APR), an organization which represents the re-refiners in the United States and Canada. The APR is privileged to place a research associate, Mr. Robert F. Pedall, at the Bureau of Standards, and this is an indication of our full support of the important effort being carried out at the Bureau on recycled oils.

In my remarks this morning I would like to take a brief look at the re-refining industry, and then share with you our views with respect to the key issues which surround the work being undertaken here at the National Bureau of Standards (NBS). I am sure most of the attendees at this session are aware of the decline of the re-refining industry. In 1965, there were an estimated 160 companies reported to be re-refiners, and today the number is about 25. From a high in the early 1960's of 300 million gallons of oil re-refined, the figure has dipped below 100 million gallons and continues at these low levels although we estimate production increased a bit in the past year.

It is hard to understand why this decline could occur at a time in our history when there has been a lube oil shortage due to an oil embargo and the word "recycle" has become a buzz word in our society. It happened, as I believe you know, through a combination of circumstances. Government action took its toll. Changes in tax policy hurt the re-refiners. Incredibly, the re-refiners pay a tax that producers of virgin oils do not pay. Labels are required to carry the onerous clause, "made from previously used oils." Market barriers were erected, particularly in the military. And, indeed, among the factors was also the performance of the industry--the fact that anyone could call himself a re-refiner and sell a product of any quality that he determined would fit his customers' needs.

I would like to state to the auto manufacturers here today, to the additive manufacturers, and to potential users of re-refined oil that we in the Association of Petroleum Re-refiners share with you a need to establish procedures to assure the quality of re-refined oils. We do not want a return to the pre-1965 days. We are calling for a system that will enable us to demonstrate the quality of re-refined oil and its equivalency with the original product. And we are asking also for the opportunity to compete equally with the producers of the original material.

Does it not make much more sense to recycle and use the product over and over again than to burn it and use it once?

It is a problem for the re-refiners that by far the largest disposal of used oil is burning. We estimate that three-fourths of all used oil is burned; that is about 10 times the amount re-refined. This compares to West Germany, where over half of all used oil is re-refined.

The lube fraction is the high-quality portion of the crude oil stream. It makes up roughly 2 percent of the crude barrel. From United States fields, it now averages 1.7 percent as the richer Pennsylvania and mid-continent fields play themselves out. Every authority I have talked to confirms that crude from the newer sources--that is the Alaskan North Slope and the North Sea and Mid-East crudes--are substantially lower in the lube fraction. A spokesman from a major lube producer told me, "There is considerable effort being devoted to the development of new technology in order to produce quality lube from lesser-quality crudes."

It is possible, of course, to manufacture lubes from the lower-quality portions of the crude stream, but it is more difficult and more costly, and also consumes more process energy.

One does not conserve energy by burning used oil. The process energy needed to refine virgin lube is double the process energy used to re-refine. Taking into account all the energy factors in burning versus re-refining, there is a net savings of 23,590 Btu's for every gallon of oil re-refined.

At the workshop held last year, I took some notes on statements from the following company representatives:

Chrysler:	"One rule we have is don't burn--then it's gone. We can make something of it."
Alcoa:	"We burn our oil only as a last resort. There are better uses."
Ford:	"We have a corporate mandate. We don't burn."

It was not too many years ago that there was a lube shortage. Lube oils were on allocation due to the import embargo. Have we forgotten this? With the political instability in the Middle East, we could awaken any morning and find that there is a war among those countries or that other political events have cut off our source of imported oil. Why does it not make sense to lessen our dependence on foreign sources of this very valuable fraction of the crude stream? And yet there are those who would say burn it--burn it and we will supply you with new.

In addition to the need to conserve a valuable resource, there are serious environmental considerations which favor re-refining over burning or dumping.

Used oil improperly disposed of is a considerable hazard to our environment. It contains high levels of contaminants, including the spent additive package; heavy metals such as lead, barium, zinc, and magnesium; and various complex organic compounds. Crankcase drainings contain an average of 0.8 percent weight lead; that is about 1,000 pounds of lead in every 1,000 gallons of drainings. Although the lead level in gasoline is declining, we do not find a comparable decrease in the lead in used oil. Lead will probably always be a significant part of the used oil contaminant. and, indeed, lead substitutes, the MMT's, are under investigation as potential carcinogens.

But let us look at lead. Lead buildup in the human body is cumulative, and it attacks the central nervous system, causing serious illness. Children are most affected, as they appear less able to pass the lead through their systems.

Used oil is burned as a fuel substitute primarily in populated areas where it is collected. Comparing this to auto emissions as a source of lead, there is particular concern for a source which is stationary, with stack emissions creating a steady fallour over the surrounding populace as compared to a moving source causing air movement close to the ground.

To date there has been a limited amount of testing of ambient lead levels surrounding the facilities burning used oil. It is good that the Environmental Protection Agency (EPA) is stepping up its activity in this area. One example is the Fairfax County school system in Virginia, which undertook to determine in 1974 the feasibility of burning used oil as a fuel supplement in the school system. Tests were conducted in conformance with the EPA specifications, and the findings were that, "...continual burning of waste oil in schools would expose students to unacceptable levels of contamination." This was in spite of the fact that the used oils in the test contained an unusually low amount of lead, only 0.2 percent, and the tests were run with as little as 25 percent used oil blend. The study further concluded that, "...it would be expected that the fallout of the contaminants would be in a concentrated area close to the building and hence would be expected to settle in the playground area and would be ingested into the school ventilating and fresh air system." Testing actual stack emissions, the schools concluded that, "...lead content would cause the level to increase as high as 6 to 8 micrograms per cubic meter, significantly higher than threshold levels quoted by EPA."

A recent Canadian study showed that atmospheric lead concentrations of less than 2 micrograms per cubic meter caused lead concentrations in the blood of exposed children higher than those of the average urban child. Blood levels exceed the amount considered hazardous. The American Petroleum Institute-conducted Hawaiian Electric tests showed atmospheric lead levels during blowout in excess of 2 micrograms per cubic meter.

The EPA stated in its 1974 Report to Congress that all airborn lead "...contributes to excessive lead exposure in...adults and children...[and] that it would be prudent to reduce preventable lead exposure."

I would ask you to reflect on the presentation before this workshop just a year ago by officials from the Aberdeen Proving Grounds. This was the basis for a directive to military and government facilities to burn used oil. The first oil tested was reported to contain jet fuel and was mixed with No. 2 fuel oil at a 1-percent level. It was found to burn without undue stack emissions. A more typical waste oil containing crankcase drainings was collected from the base, mixed at 50-percent levels with a No. 2 fuel, and was found to fail stack emission tests. And now for the conclusion. It is all right to burn at blend ratios up to 5 percent in No. 2 fuel! Who takes the responsibility for this?

At the risk of exaggerating the point, I would inform you that there is a panic among the people of northwest Washington; that is the affluent section of this community where many congressmen and government officials live. It has been found that children have dangerously high blood lead levels in this area and the cause is uncertain. Women are going from door to door in the Cleveland Park section in a campaign to have children's blood tested. We have no evidence that the burning of used oil has contributed to this, but we have no evidence that it has not.

An attempt in this area to determine where used oil is being burned is not productive because the collectors are not saying who is doing the burning. We are certain that most of the collected used oil in this area is burned and that it is burned untreated. The typical scavenger needs only a rental truck and he is in business, and it is difficult for the re-refiner, with his investment in plant and equipment, to compete for these feedstocks.

There are those who say let economics determine whether used oil is burned or re-refined. This is fine, but let the economics include the costs of pollution and the costs of a lost resource.

The APR opposes burning on conservation grounds. It believes that accountability for disposal will be nonexistent without government regulation and therefore supports the programs of the EPA and the Federal Trade Commission toward this end. In those cases where it is not practical to re-refine, burning may be conducted only after the used oil is processed to remove the contaminants and its equivalency to fuel demonstrated.

What about the claim that the re-refiners are too small to handle a surge of used oils diverted to this higher order? It is expected that regulations would have an implementation period to allow the industry to position itself to handle increased volumes. There is ample evidence that the industry is capable of this growth. Unused capacity would enable an immediate increase in volume to an estimated 150 million gallons a year. In recent years, corporations with larger capital resources have entered the field, including Esmark, Anchor Hocking, and most recently Phillips Petroleum Company. There is great interest in the future of re-refining. There can be no doubt that, given sufficient economic incentives, the re-refining industry will provide the capacity to handle the volumes of oil generated.

As to the technical problems that exist within the industry, we are very optimistic in this area also. A veteran re-refiner said to me just last week he has seen more process development and innovation in the past 2 years than in the entire 25-year history of our Association. Can we solve the acid/sludge problem? The answer is yes. Two new processes in pilot-plant stage, the Bartlesville Energy Research Center and Phillips processes claim to have solved the sludge problem. Two re-refiners, using the acid-clay process, report to me that they have process developments which have solved their sludge disposal problems. It is important that small members of the industry be allowed time to make adjustments so that they can expand in volume and still be capable of disposing of by-product materials. Process developments will have to be made available to these companies in an equitable way that will enable them to survive. The EPA must recognize the need for a growing re-refining industry and make allowances for interim sludge disposal at the local level.

Finally, I would like to comment on the question of the quality of re-refined oil. In the past year, two tests of considerable significance were run on re-refined oils. One was the City of San Diego fleet test. The city had used re-refined oil for 3 years in its vehicles. In all, 1,500 vehicles used 120,000 gallons of re-refined oil in the period. After 100,000 miles of service, typical engines were torn down and compared to another city whose vehicles used virgin oils. The report from an independent laboratory, conducted under the auspices of the EPA, concluded that the engines run on re-refined oil were "...as good as if not better than those using virgin oils."

In addition, Department of Defense/EPA-sponsored engine sequence tests were conducted on six samples of re-refined oil selected to be broadly representative of oils produced. Preliminary results of these tests are that four of the six passed all the tests for SE qualification and that overall pass/fail ratio for a one-time test was remarkable, comparing favorably to the best of the virgin oils.

The engine sequence test has been a problem for re-refiners. With costs up to $40,000 per oil, it is understandable that we cannot afford to conduct the tests. I recognize that for many of you the engine sequence tests are the product of many years development and that they have served you well. But there are things about this test that are worrisome besides its cost. First is the reproducibility of the test. I understand that there is a high fail rate of all oils, even the best that are subjected to the tests, and that it is the practice to run the test repeatedly until a pass is achieved and then to report only passes. I also understand that it is the practice to start with a low additive level and gradually increase the level until a pass is achieved. Is this a quality test for a base oil or for the additive package, or is it a test to show the minimum additive blend needed for those few companies who can afford repeated tests? Is it possible that any base oil with sufficient additive will pass an engine sequence test?

When all is said and done, what does the SE label mean? The purpose of any label is simply to inform the consumer. Not many consumers have any idea what the label on their motor oil means. Try your local service attendant and see how much he knows about the American Petroleum Institute (API) classification system. Add to this the fact that marketers of oils are apparently misrepresenting the SE label. This was shown in a recent survey by an automotive company showing that a low percentage of the oils sampled which were labeled SE actually met those quality levels.

These tests have been developed by a small number of large companies, and only a small number of large companies can afford to participate in the system. The problem is that the system, by excluding the re-refiner, has posed a barrier to recycling and the conservation of a valuable resource.

Now that the quality of re-refined oils is being demonstrated, the obstacle to broader acceptance seems to be questions as to consistency of re-refiner feedstock. Re-refiners have claimed that through proper segregation of incoming stocks they do have a consistent feed source. The Bartlesville Laboratory of the Department of Energy, in Part V of its Waste Lubricating Oil Research series entitled "A Comprehensive Characterization of Five Typical Re-refinery Feedstocks," reported that after careful analysis of samples of waste oil collected from 20 States to give geographic and seasonal variations, "The data show the various samples to be extremely similar, implying that an acceptable reclamation procedure should produce consistent re-refinery product with minimal effects from feedstock variations."

Is it possible that the re-refiners' feedstock would compare favorably with variations in crudes? There are changes in crude makeup when the field begins to run out. How easy is it to control the consistency of foreign crudes which make up more and more of domestic production? I am told this is very difficult and that foreign crudes are identified by their port of origin and these crudes can be a blend of oils from as many as three different fields. Most domestic plants are using a combination of domestic and foreign crudes. One plant I talked to has used 25 different foreign crudes in the past 5-year period. Half of the products on the Defense Department's qualified list are imported crudes.

At a time of national emergency, lube producers will probably have to take crude from wherever they can get it. What happens to the elaborate military specification qualification system then? It fails when it is needed most. Is it possible that the most consistent products at that time would be re-refined oils, the very products excluded by the current military specifications?

If consistency of feedstocks is indeed the major problem that the engine manufacturers and additive people have as to determining the consistent quality of re-refined oils, by all means let us now work together to develop the test methods that will enable us to satisfy you as to our feedstocks and then to show the equivalency of re-refined oils with the original materials. A re-refiner capable of producing a quality product should have the opportunity to qualify his product and then provide the ongoing assurances of quality based on the tests that will enable him to show that equivalency.

In conclusion, to preserve this valuable finite resource and abate pollution of our environment, the first step, in our view, is to manage and control the disposal of used oil. We ask your support for the need to demand accountability for the disposition of used oil and to stop the uncontrolled burning. We share the same goal in seeking an orderly development of the re-refining industry that will not be disruptive of fair procedures for establishing the quality of all such products. The re-refiners ask for the opportunity to compete by the same rules that apply to all and for the opportunity to demonstrate the equivalency of their products to the original for its intended use. And that is why we so heartily support the fine work at the National Bureau of Standards.

National Bureau of Standards Special Publication 556. Proceedings of a Workshop on Measurements and Standards for Recycled Oil - II held at NBS, Gaithersburg, Maryland, November 29 and 30, 1977. (Issued September 1979)

A LIMITED INTEGRATED ASSESSMENT OF THE WASTE OIL RE-REFINING INDUSTRY[1]

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In the United States, the Resource Conservation and Recovery Act of 1976 (RCRA, Public Law 94-580) will have profound impacts on the waste oil re-refining industry. The two broad goals of the RCRA are:

- to conserve natural resources both directly and through the management, reuse, and recovery of solid and hazardous wastes; and
- to assure that all solid and hazardous wastes are managed in a manner that will protect public health and the environment.

Few would argue with the individual merits of these goals. Unfortunately, there exist circumstances where the pursuit of one may jeopardize the other. Where national objectives work at cross purposes, administrative and regulatory agencies find themselves in the unenviable position of having to choose between the lesser of two evils. The waste oil re-refining industry's future may be determined, in part, by the relative priorities given these goals by Federal and State environmental agencies. Given the present technological profile of the re-refining industry, the re-refining of waste oil to promote the reuse of an exhaustible resource generates toxic by-products which present an environmental hazard. Strict regulation to prohibit the latter may result in diminishing the former.

In seeking to achieve resource conservation while assuring environmental protection, or at least to achieve one without undermining the other, the Environmental Protection Agency (EPA), in promulgating regulations under the RCRA, must be aware of the secondary impacts and tradeoffs involved.

In the case of waste oil re-refining, environmental regulation should bear in mind not only the reduction in solid waste, but also the secondary impact such a reduction might have on the cost of re-refining; the profitability of the industry; and, ultimately, the volume of waste oil re-refined. This requires an assessment of the industry and its likely response to different assumptions about future environmental regulations, the re-refined oil market, and the technologies available to re-refiners.

Supported by a grant from the National Science Foundation (Grant No. AEN-7517302), Teknekron's Resource Management Division conducted a Limited Integrated Assessment of the Waste Oil Re-refining Industry. Answers to the following questions comprise, in part, this analysis and permit such an assessment.

- Are there process technologies for re-refining currently available, or available in the near future, which will not produce a hazardous by-product while being capable of producing a quality product at a profit?
- What is the nature of the industry's present hazardous waste disposal problems?
- Can the present hazardous by-products be safely disposed of, either treated or untreated?
- Can economically justified investments be made to retrofit existing technologies to eliminate environmental harm?

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[1] This paper was not able to be presented at the Conference.

In short, what is the current health and what is likely to be the future health of the re-refining industry? How sensitive is its health to various assumptions about future technologies, environmental regulations, and the economy?

In order to conduct the assessment, a Limited Integrated Assessment Model (LIAM) of the re-refining industry was constructed. The LIAM permits us to look at a limited number of factors which can serve as measures of the re-refining industry's health and to determine the impact of different process technologies, environmental regulations, and growth rates on these industry factors. The factors for which we have conducted an impact analysis are:

- average cost of producing a gallon of re-refined base oil,
- volume of waste oil re-refined,
- before-tax profits,
- volume of solid waste generated, and
- volume of solid waste disposed.

Figure 1 is a conceptual framework for the model showing how we select a specific technology mix, environmental regulation, and growth rate to form a scenario. We then measure the likely impact of alternative scenarios on the industry's cost of production, profit picture, solid wastes generated, and volume of oil re-refined by 1983. These future impacts may be compared with the present status of the industry to determine the industry's sensitivity to the various scenarios and the extent to which the alternative scenarios achieve or jeopardize national objectives regarding solid waste management and resource recovery within the context of the re-refining industry.

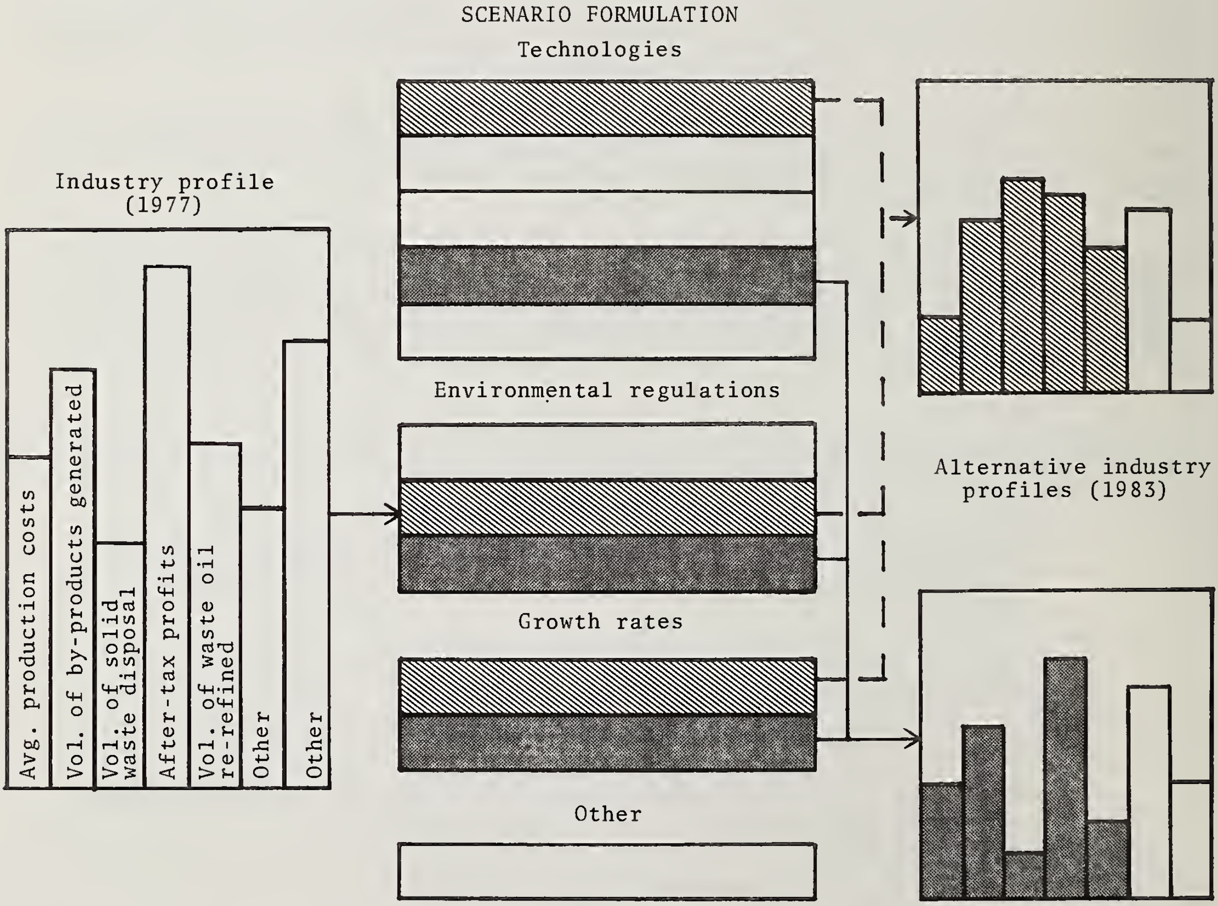

Figure 1. Limited integrated assessment model (LIAM) conceptual framework.

Figure 2 graphically depicts the steps that were taken to link the realities of the existing system with policy instruments intent on achieving national objectives as they relate to the specifics of investment in the re-refining industry. The LIAM was organized around three steps: (I) technological models, (II) financial models and analysis, and (III) scenario development and analysis.

The first step in the analysis was to understand the technologies capable of re-refining waste lube oils to high-quality re-refined lubes. Such understanding rests on both a quantitative and qualitative analysis of what is produced, how it it produced, what by-products are generated, what efficiencies are or are not utilized, and how any given technology compares to any of the others.

The second step was to understand the financial constraints under which the system operates. In order to do this, a model was constructed so that alternative investment schemes in the re-refining industry could be systematically analyzed. Investment schemes that were analyzed fell into two generic categories: new investment from outside the industry and reinvestment from within. In the first case, the task involved comparisons of net present values and internal rates of return under varying exogenous parameters. In the second case, two operating acid/clay re-refineries were closely modeled and then hypothetically retrofitted with distillation/clay processes. The investments required for retrofit were then analyzed for profitability.

The third step was to incorporate into the analysis all the real-time exogenous variables to which the system must respond. For example, such parameters as industry growth, changes in the mix of technologies that comprise the industry, potential future regulatory scenarios, plus the models that were developed in steps I and II were combined in such a way that estimates were made showing impacts and tradeoffs involved as any given scenario of future events played itself out.

An important instrument for decision making under uncertainty is a model of the system. The model is used to generate the future outcomes and impacts of hypothetical input conditions representing policy decisions. A suitable model simulates the actual system to a level of detail consistent with the needs of policy makers and makes it possible to explore the effects of alternative policies quickly and inexpensively. The model presented here is a representation of the re-refining industry systems: it accepts information and directions characterizing a pollution-control policy and provides output on that policy's possible economic and environmental impacts. The most salient findings are summarized here:

What is the present state of the re-refining industry?

- The acid/clay technology is decreasing in its share of total re-refined oil production. In 1970, nearly 90 percent of re-refined oil was produced by acid/clay technology; by 1974 this figure had declined to less than 80 percent, and by 1977 it had declined to 55.8 percent.

- Total re-refined oil production has increased for the first time since 1960.

- Nearly 75,000 tons of hazardous wastes were generated by the re-refining industry in 1977.

- Industry average production costs in 1977 were approximately $0.328 per gallon of product.

- In 1977, more than 125 million gallons of waste oil were re-refined.

What is the nature of the re-refining industry's present hazardous waste disposal problems?

- Using several criteria, it was determined that acid sludge is *intrinsically* hazardous.

- The *extrinsic* hazards of acid sludge disposal must be determined on a site-specific basis.

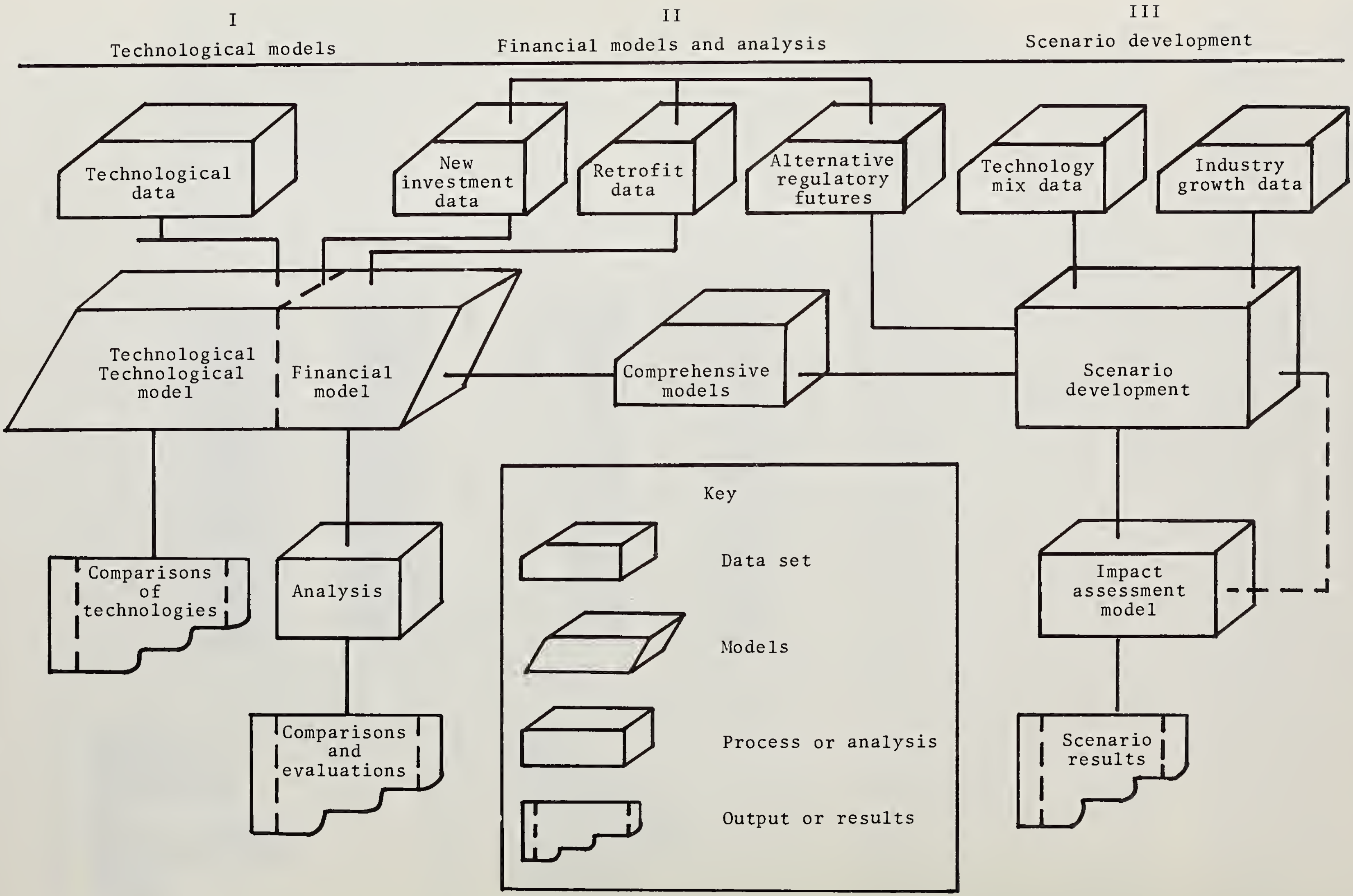

Figure 2. Conceptual framework.

- The acceptability of "neutralized" acid sludge as a non-hazardous waste will be determined not only by regulatory agencies, but also by landfill operators.

- Samples of "neutralized" acid sludge were tested and shown to be *intrinsically* hazardous, although their "neutralized" state may attenuate the percolation problems and, thus, may significantly affect the *extrinsic* hazards.

How will the EPA evaluate the disposal of acid sludge?

- There will be no equivocation regarding Federal and State judgments of acid sludge. According to every opinion we have heard in many discussions with the EPA and State health and environmental officials, acid sludge will be classified as a hazardous waste. This is because the Resource Conservation and Recovery Act (RCRA) of 1976 will determine *only the intrinsic* hazard of a given waste.

How will the RCRA affect the waste oil re-refining industry?

- Enforcement of the RCRA will result in an increase in average industry production costs of 7 to 28 percent.

- Potential regulations contained in the implementation phase of the RCRA (which, at this writing, will become effective in April 1978) will seriously affect small re-refiners who presently use the acid/clay technology. Large acid/clay re-refineries will be less affected, although their operating costs will increase.

- During a period of low economic growth, progressively more stringent RCRA policies will significantly decrease projected hazardous waste generation but will, in turn, reduce the amount of waste oil that is re-refined.

- The generation of hazardous wastes by the re-refining industry is extremely sensitive to combinations of the RCRA regulations and the industry's mix of technologies.

Can economically justified investments be made to retrofit existing technologies to eliminate environmental harm?

- Large acid/clay re-refiners have a profitable alternative to the regulations contained in the RCRA: if all technical aspects are resolved, they could retrofit their plants with a distillation-type technology; the resultant plant would run more efficiently and profitably than their present operation. Small acid/clay plants, if retrofitted, show unacceptable future cash flows, and, therefore, investment in retrofitting small acid/clay plants is not recommended.

Are process technologies for re-refining currently available, or available in the near future, which will not produce hazardous wastes while being capable of producing a quality product at a profit?

- The distillation-type technologies produce significantly greater quantities of marketable by-products than does the acid/clay technology. It is not known whether these products are hazardous; it is assumed that they are not.

- Several distillation-type re-refineries show high returns on investment, even when a worst-case scenario is considered.

Several conclusions may be drawn from these findings. First, most small acid/clay re-refineries will be highly affected by increasing waste oil costs and increasing acid sludge disposal costs. Second, since most acid/clay re-refineries have individual outputs of less than five million gallons per year, solid waste disposal policy makers should be aware that stringent solid waste disposal policies will negatively affect a large part of the industry. Third, in seeking to achieve resource conservation while assuring environmental protection (or at least to achieve one without undermining the other), the EPA, in promulgating and enforcing regulations under the RCRA, must be aware that progressively more stringent RCRA policies will significantly decrease hazardous waste generation but also may, in turn, reduce the amount of waste oil that is re-refined. Finally, financial analyses of distillation-type technologies (which generate nonhazardous wastes) show promising results for potential investors.

SESSION II. ENVIRONMENTAL CONSIDERATIONS

National Bureau of Standards Special Publication 556. Proceedings of a Workshop on Measurements and Standards for Recycled Oil - II held at NBS, Gaithersburg, Maryland, November 29 and 30, 1977. (Issued September 1979)

A SURVEY OF METALS IN OIL: OCCURRENCE AND SIGNIFICANCE FOR REUSE OF SPENT AUTOMOTIVE LUBRICATING OILS

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This paper summarizes the results of a literature review which was undertaken to (1) summarize and evaluate available data on the occurrence and on the chemical and physical forms of metals in used lubricating oils, and (2) evaluate the implications of the metal content of used lubricating oils to their use and disposition.

A considerable body of information is available concerning the levels of metals in used oils. The principal sources of metals in these materials are: (1) metal-containing additives incorporated in essentially metal-free basestocks, and (2) metals accumulated during use, primarily lead from gasoline additives and iron from wear processes. The most significant change in metal content during the service life of lubricating oils is the accumulation of lead, which increases in concentration by a factor of 10,000 to 1-1.4 percent (w/w). Additional metal inputs may occur during collection, handling, and storage of used oils.

The specific chemical and physical forms of metals in used oils are largely unknown. It appears, however, that much of the lead (in the range of 50 percent)--and possibly other metals--is present in the form of fine suspended particulates.

Primary applications of spent lubricating oils to which metal contents are relevant are: (1) re-refining, (2) combustion, and (3) uses for road oiling. A significant fraction is currently disposed of by landfilling.

Re-refining technology reduces metal levels to values substantially equivalent to those of virgin basestocks. While re-refining residues containing high metal concentrations present a disposal problem, metals per se do not appear to present a problem for the equivalency of re-refined oils to virgin materials for automotive use.

Metallic contaminants probably do not affect either combustion efficiency or short-term integrity of combustor components when used oil is burned, particularly if it is used diluted with fuel oil from virgin sources. There are well-defined chemical mechanisms by which metallic impurities can adversely affect refractories and metals in combustion applications, but such effects have not generally been observed in short-term pilot studies. Long-term studies are probably required, however.

The principal problems associated with the high levels of lead and metallic additives in spent lubricating oils are environmental, specifically emissions from combustion and uncontrolled release to the environment in road oiling and dispersion through landfill operations.

Better characterization of the metallic contaminants in spent oils is of potential benefit to reprocessing technology, as well as to assessing environmental consequences of waste oil use and disposition. Data are needed on the distribution of metals among water-soluble, oil-soluble, and particulate constituents; on size distribution and metal distribution in particulate fractions; and on the specific chemical forms in which metals occur. Acquisition of these data presents significant measurement problems in the chemical characterization of amorphous particulates and in the speciation of soluble and metal-organic compounds occurring in spent oils.

A copy of the complete survey is attached to this volume as Appendix I.

National Bureau of Standards Special Publication 556. Proceedings of a Workshop on Measurements and Standards for Recycled Oil - II held at NBS, Gaithersburg, Maryland, November 29 and 30, 1977. (Issued September 1979)

SPECIATION OF METALS IN USED OILS: RECENT PROGRESS AND ENVIRONMENTAL IMPLICATIONS OF MOLECULAR LEAD COMPOUNDS IN USED CRANKCASE OILS

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Introduction

The presence of many metals in used crankcase oils is well known to originate from wear processes in engines and from blow-by products in fuels consumed during engine operation. Additive "packages" introduced into new oils frequently contribute significant concentrations of several metals along with a variety of solubilizing ligands [1].[1]

End uses of spent crankcase oils include re-refining and combustion as fuel components. Probably more significant to environmental concerns, however, are additional uses in road oiling or unregulated dumping in landfills. Re-refined oils display considerable reductions in residual metal loadings, although refining sludges associated with commercial reprocessing can also constitute a hazardous disposal problem [2].

Since road oiling and landfill discard represent widespread direct imposition of metal components on environmental cycling through leaching into the aquatic system, there exists a pressing need to evaluate which metals in what forms represent the chemically and biologically important threats to the environment. One aspect of recent work at the National Bureau of Standards (NBS) has therefore focused on developing means for characterizing the principal toxic metal components in waste oils as typically collected and comparing these results with those for "new" oils. Annual uncontrolled disposal is responsible for over 200 million gallons of such waste oils finding their way into drained soils [1]. Consequently, our initial studies have been directed to an assessment of the typical amounts of a major waste oil component, lead, and the nature, or bioavailability, of lead chemical species presented by such a large and ubiquitous environmental stress.

Potential Environmental Impacts of Metals in Waste Oils

In studies dealing with hazards of technological materials or wastes to the biosphere, several fundamental considerations require priority. In table 1 are summarized such factors which underly the preliminary NBS studies on form and fate of toxic metals in waste oils or associated re-refining processes.

Bioactivity or bioavailability of toxic metals. Degradation of crude and waste petroleum products by soil or marine microorganisms has been long known [3]. More recently, it was shown that heavy metal-polluted oils in marine sediments were subject to active bacterial degradation, implying that biotransformations both of oil and metals were occuring [4]. In general, we can conclude that, while forms of toxic metals present in new or spent oils are undetermined, some degree of direct chemical solubilization can occur into ground waters by leaching or by biological degradation yielding leachable substrates.

A basic question remains concerning the mode(s) of dispersion of metal-containing materials by man and the chemical pathways by which these toxic substances are transported. Pertinent to environmental issues involving waste oils, for example, is the evaluation of automotive traffic as a primary dispersal mechanism.

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[1] Underlined numbers in brackets indicate the literature references at the end of this paper.

Table 1. Environmental Impacts of Waste Oils

- Bioactivity or bioavailability of toxic metals:
    - -- Metal dispersed in a form readily taken up by food chain
    - -- Metal dispersed in a chemical form permitting biotransformation
    - -- Metal dispersed in a chemical form which abiotically reacts to form toxic compounds of other metals already present
- Measurement problem:
    - -- Total concentration of toxic metals in dispersal system
    - -- Molecular characterization or speciation of bioactive molecules containing toxic metal(s)
    - -- Quantitation of species amounts and their rates of formation
    - -- Determine leaching rates of species across oil-water interfaces
- Approach:
    - -- Develop reliable means for speciation of toxic metal(s) in used/recycled oil or water matrices
    - -- Determine bioactivity of key metal contaminants

Concentrations of toxic metals in soils and vegetation have been positively correlated with proximity to roadways and traffic volume [5]. Elevated concentrations of lead and nickel were attributed to vehicular gasoline emissions, and cadmium and zinc both to tire debris and engine oil slicks. Significant in the Gish and Christensen study [5] was the demonstration that organisms in roadside soils accumulate metals dispersed by automotive traffic. Figure 1 illustrates data selected from that report treating bioaccumulation[2] of lead in roadside earthworms. Within 20-80 feet of a busy roadway near Washington, D.C., total lead concentrations in earthworms nearly mirror total lead concentrations in habitat soils, and both sets of values are considerably greater than background levels for lead in pristine soils. The exponential decay in lead uptake out from the roadway suggests that rain water runoff overrides a limited atmospheric transport mechanism.

The influence of water on bioavailability and bioaccumulation of the kinds of heavy metals associated [1] with vehicle operations is vividly demonstrated by the data summarized in table 2. Terrestrial earthworm bioaccumulations of lead and several other metals of interest are listed [5] as ranges for locations 20-80 feet distant from the roadway. In comparison, Patrick and Louitt examined a food chain consisting of lake sediment → bacteria → aquatic worm (tubificids) and found bioaccumulation for all the metals studied. Lead, for example, was ultimately accumulated in aquatic worms by a factor of 15 over sediment concentrations.

The chemical form in which metals are dispersed into environmental media--air, soils, waters--is crucial to biological uptake. The Environmental Protection Agency indicates that bioaccumulation of lead in man mainly results from automotive exhausts [6]. Originally, such atmospheric lead transport was thought to occur

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[2] For convenience, in this paper "bioaccumulation" (ratio) will be defined as the total metal concentration in an organism (or cell mass) divided by the total metal concentration determined in the supporting soil, sediment, or growth medium.

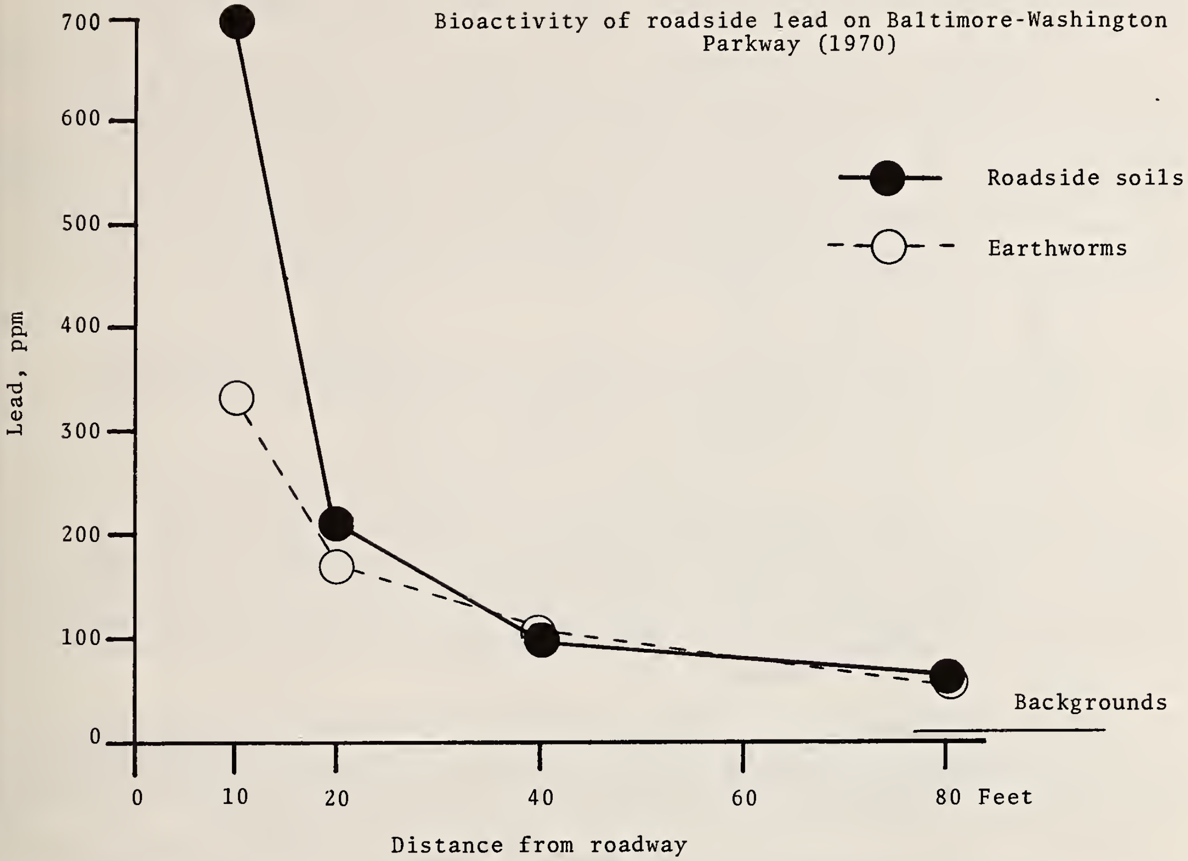

Figure 1. Bioactivity of lead emissions from highway traffic is indicated by uptake in nearby earthworms. Typical lead backgrounds in uncontaminated soils are shown on the right.

Table 2. Bioaccumulation of Metals

	Cd	Ni	Pb	Zn	Cr	Mn
Terrestrial						
Soil → earthworms	4.6-8.6	1.9-1.5	0.8-1.1	5.3-5.1	----	----
Aquatic						
Sediment extract medium → bacteria	---	---	4.8	8.9	17.6	25.7
Bacteria → worms (tubificids)	---	---	3.2	3.0	5.6	1.6
Overall accumulation			15	27	99	41

chiefly by particulate inorganic lead emissions, but more recent work suggests that gaseous "molecular" lead species may be significant transport components [7].

Participation of aerosol inorganic lead compounds in atmospheric transport seems likely in view of their appearance on urban foliage surfaces (8). It is interesting to note that lead associated with sulfur and phosphorous (as sulfated and pyrophosphate) was found on tree leaves rather than expected stable lead halides (1,8). Inorganic (ionic) lead compounds are shown to be highly absorbed in terrestrial soils where capacities for immobilization may be reasonably predicted by correlation between cation-exchange capacities and pH [9]. Significantly, the majority of bound lead in soils may be associated with organic matter [9] such as humic acids [10], and the fixation process exerts primary control over bioavailability to plants. Similar arguments for sequestration and chemical solubilization of other metals by humic materials have been put forth [10].

Biological mobilization of lead and other metals from soils, sediments, and sewage sludges is evident. Toxic metal concentrations in urban and industrial sewage sludges are high [11], and it is important to note that both aerobic and anaerobic incubation of these materials can strongly influence rates of release of metals, including lead, into overlying water [12].

Perhaps most significant to concerns for assessing biotransformations of lead-containing anthropogenic wastes was the nearly coincident discovery of lead biomethylation by fresh water bacteria isolated in Canada [13] and Germany [14]. Generally, methylation rates found for inorganic lead species were slower than those observed with organolead precursors, but tetramethyllead was found in all cases. Presumably, initial stepwise formation of relatively unstable mono- and di-methylead intermediates are rate-determining. This biogenesis of methylleads is a critical point to consider with respect to an overall biochemical cycle for lead involving oil → sediment → water → atmosphere.

Partition coefficients for aquated methylead ions, $(CH_3)_nPb^{(4-n)+}$, where $n \leq 3$, favor retention in solution, whereas volatile and hydrophobic tetramethyllead may readily escape to the atmosphere. This situation, which involves extreme range of volatility of trace bioactive organolead species, dominates the state of the art in current measurement techniques, as will be further discussed below.

The foregoing brief review assumes special meaning upon evaluating a recent environmental incident involving organometal contamination of food fish in the St. Clair River system, Michigan.

Chemical Involvement of Other Metals in Biological Transport of Toxic Metals. In 1970, dangerously high mercury concentrations were found in fish taken from the St. Clair River locale. Patterns of mercury contamination in fish and various sediment locations implicated an industrial source [15]. When fish and sediment samples were analyzed by a gas chromatographic method (*vide infra*), permitting separation and quantitation of methylmercury moiety, an additional new organometal component was isolated and subsequently shown to be a triethyllead species. Multiple regression analyses of fish and sediment samples in terms of location, relative abundances of CH_3Hg^+ and $(C_2H_5)_3Pb^+$, and possible discharge sources revealed the situation summarized in figure 2.

A producer of organolead compounds for gasoline antiknock additives was found to have ethyllead species in its sewage effluent entering the St. Clair River. Similarly, at another river site a previously identified manufacturer released mercury into the same system. Though microbial action was expected to mobilize the inorganic mercury from sediments as toxic methylmercury [16], relative concentration patterns for total Hg and CH_3Hg^+ did not conform to those previously seen for such bioactivity [15-17]. Jernelöv *et al.* [15] showed that direct treatment of sewage effluent from the organolead producer's plant with $HgCl_2$ yielded those organometals seen in fish and sediment, namely,

$$(CH_3)_n(C_2H_5)_{4-n}Pb + Hg^{2+} \rightarrow (C_2H_5)_3Pb^+ + CH_3Hg^+$$

$$\rightarrow (CH_3)_n(C_2H_5)_{3-n}Pb^+ + C_2H_5Hg^+$$

Evidently, abnormal sediment and fish uptake of these abiotically formed organometals subsequently occurred.

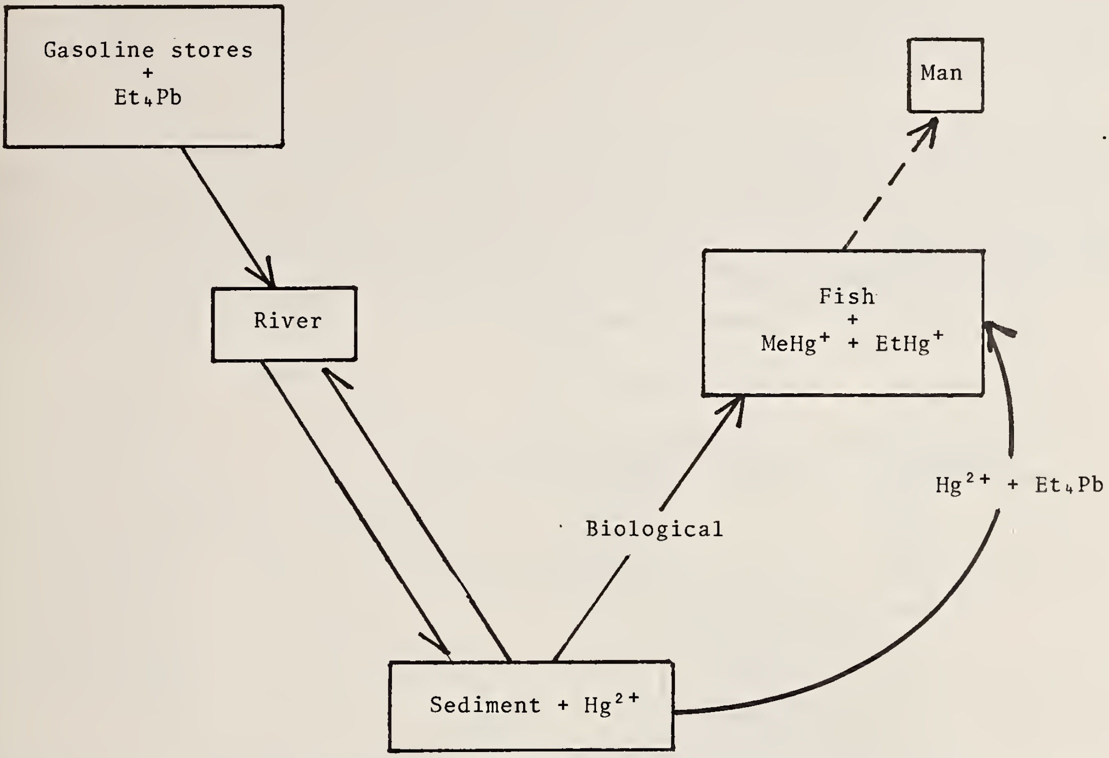

Figure 2. St. Clair River study shows environmental pathway through which organolead and mercury industrial wastes accumulate in aquatic food chain [15].

Prior to the above study, little was understood concerning the extent and rate of such aqueous nonbiological transalkylation reactions between metal ions. As a part of an overall program under the auspices of the Office of Air and Water Measurement, NBS scientists also began a study of these reactions for the purpose of acquiring kinetic data and mechanistic pathways, namely,

$$(CH_3)_3Pb^+ + Hg^{2+} \rightarrow (CH_3)_2Pb^{2+} + CH_3Hg^+.$$

Evaluations of important environmental parameters which control these chemical systems, e.g., salinity, pH, PO_2, light, ionic strength, etc., were major additional undertakings [18] in order to build a basis for environmental modeling [17] and trace characterization chemistry of bioactive organometals.

NBS Approach to Speciation of Trace Metals in Waste Oils

The total concentration of many elements, particularly lead, can be readily and accurately determined in spent or refined oil matrices by several methods [19]. For many years, commercial and NBS Standard Reference Materials providing certified concentrations of metallo-organic compounds[3] in lubrication oil solutions have been available for sale [20]. On the other hand, speciation, or molecular characterization, of dissolved metal-containing compounds in natural fluids, either aquatic or oily, is not so readily accomplished.

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[3] The term "metallo-organic" makes an important and formal structural distinction between two classes of metal-containing compounds frequently displaying bioactivity. For example, organolead molecules are compounds possessing lead-carbon bonds, namely, triethyllead cation or tetramethyllead. Metallo-lead compounds may or may not bear lead-carbon bonds, but are molecules possessing covalent or dative-bonded heteroatom-lead bonds. Examples are dimethyllead(IV) acetylacetonate or lead(II) diphenate. Similar considerations apply to other metals as well, as suggested in table 3.

Two major obstacles inhibit this relatively new field of environmental characterization. First, separation/concentration of the organometal analyte must be accomplished without structural alteration or decomposition. Second, compound- or element-specific detection at requisite trace levels must be attained reliably even in the presence of tremendous excesses of complex organic interferences.

Speciation of Volatile Metal Analytes in Hydrocarbons. Prior comments regarding the extreme range of polarity (or ionicity) and volatility of solvated organometals pointed to options available for separation schemes. Thus, for gaseous organometals, as have other workers, we developed gas chromatographic (GC) procedures which permit molecular separation compatible with element-specific detection at nanogram levels. One approach, schematically depicted in figure 3, indicates our novel use of a flameless graphite furnace atomizer (GFAA) for atomic absorption spectrophotometric (AA) detection of trace metal species eluted from the GC column.

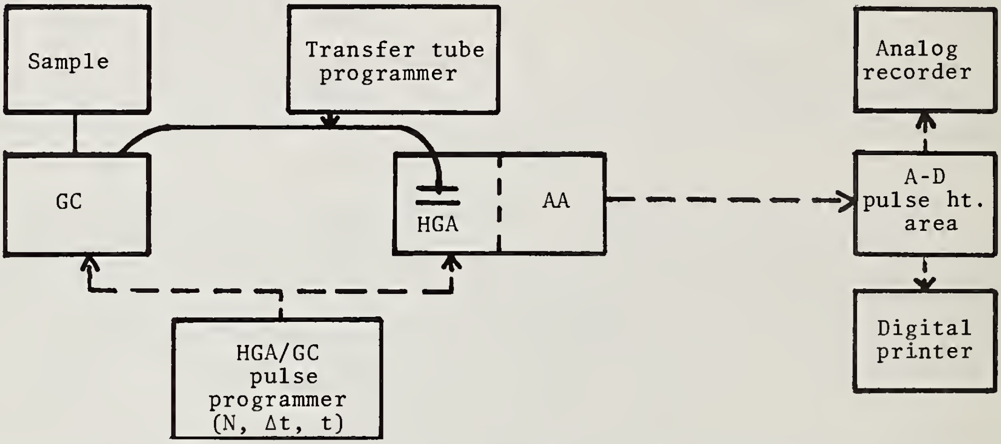


Figure 3. Schematic of gas chromatograph-flameless atomic abosrption, GC-GFAA system (see reference 21).

For most elements, continuous high-temperature operation of the furnace is not feasible. Consequently, periodic atomization or pulse analysis of the GC eluant gas stream entering the GFAA detector is performed to yield chromatograms of the type shown in figure 4.

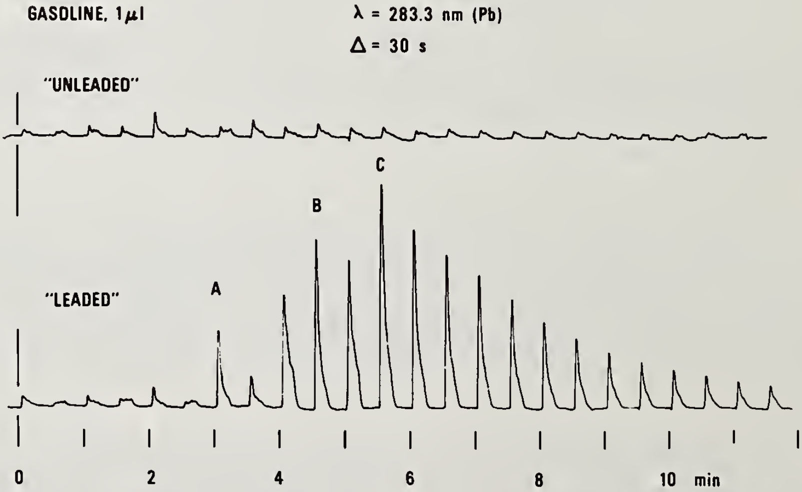


Figure 4. Gas chromatograms of commercial gasoline types obtained by GC-GFAA with lead analyses of effluent automatically performed at 30-second intervals (see reference 21).

Here, a qualitative comparison of microliter quantities of two commercial gasoline samples are compared for organolead content. The so-called "unleaded" fuel at the top appears to be just that; the familiar "leaded" gasoline apparently contains three recognizable organolead species tentatively identified as methylethylead compounds (A and B) and tetraethyllead (C), based on similar retention times reported for related GC conditions [22].

Similar application of GC-AA speciation to weathered petroleum samples containing organometals can give useful results if thermal conditions and peak resolution permit. Using a variation of the AA which employs a "cold" flameless Hg-specific detector, we examined oil samples extracted from polluted Chesapeake Bay sediments. Figure 5 demonstrates that such degraded oils contain organomercury or mercury-organic complexes in both very low and very high molecular weight fractions. Clearly, for more specific or highly resolved characterization of such metal-containing natural products, a better separation scheme was required.

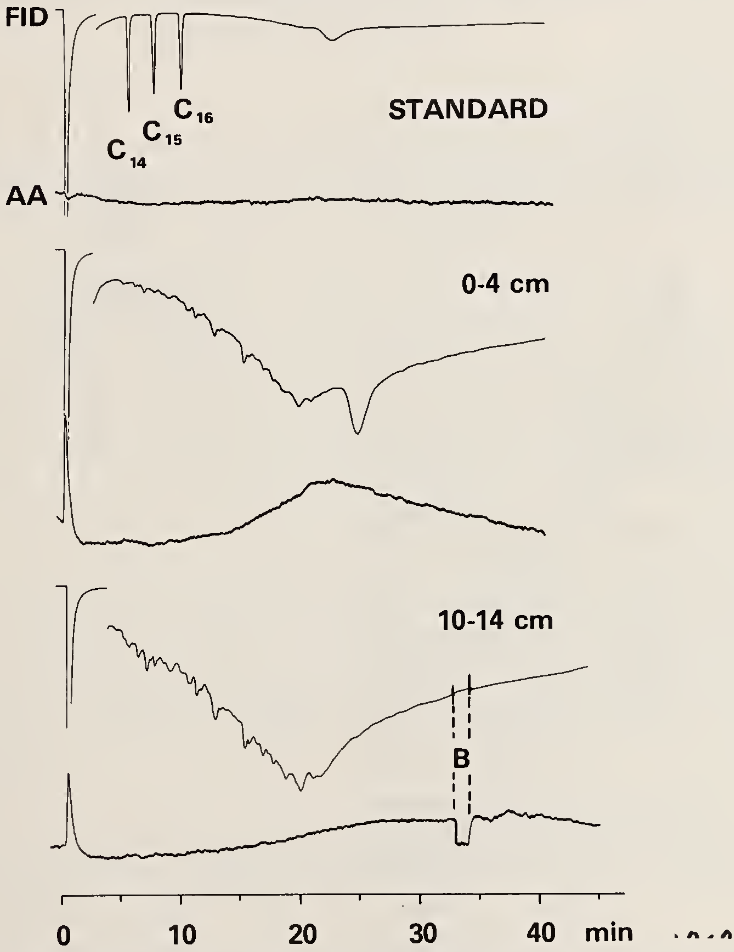


Figure 5. GC-AA chromatograms of sediment oils extracted at different core depths from Chesapeake Bay sample (see reference 17). The high MW Hg-containing "envelope" comprises about 98 percent total Hg in sample, with light mecurials such as Hg^0 and methylmercury appearing as the early small peak at 1 minute.

Speciation of Involatile Metal Compounds in Hydrocarbons. During the past decade, liquid chromatography has become a preferred method for separation of thermally sensitive or reactive molecules in both aqueous and organic media. Much of this advance results from considerable improvement in solvent delivery pumps and specially treated, fully porous column substrates possessing high surface activities and efficient mass transport properties [24].

We developed a high pressure liquid chromatograph (HPLC) automatically coupled to the element-specific AA detector. A schematic provided in figure 6 also shows that optical and other supportive detectors can be incorporated into the solvent flow system [25].

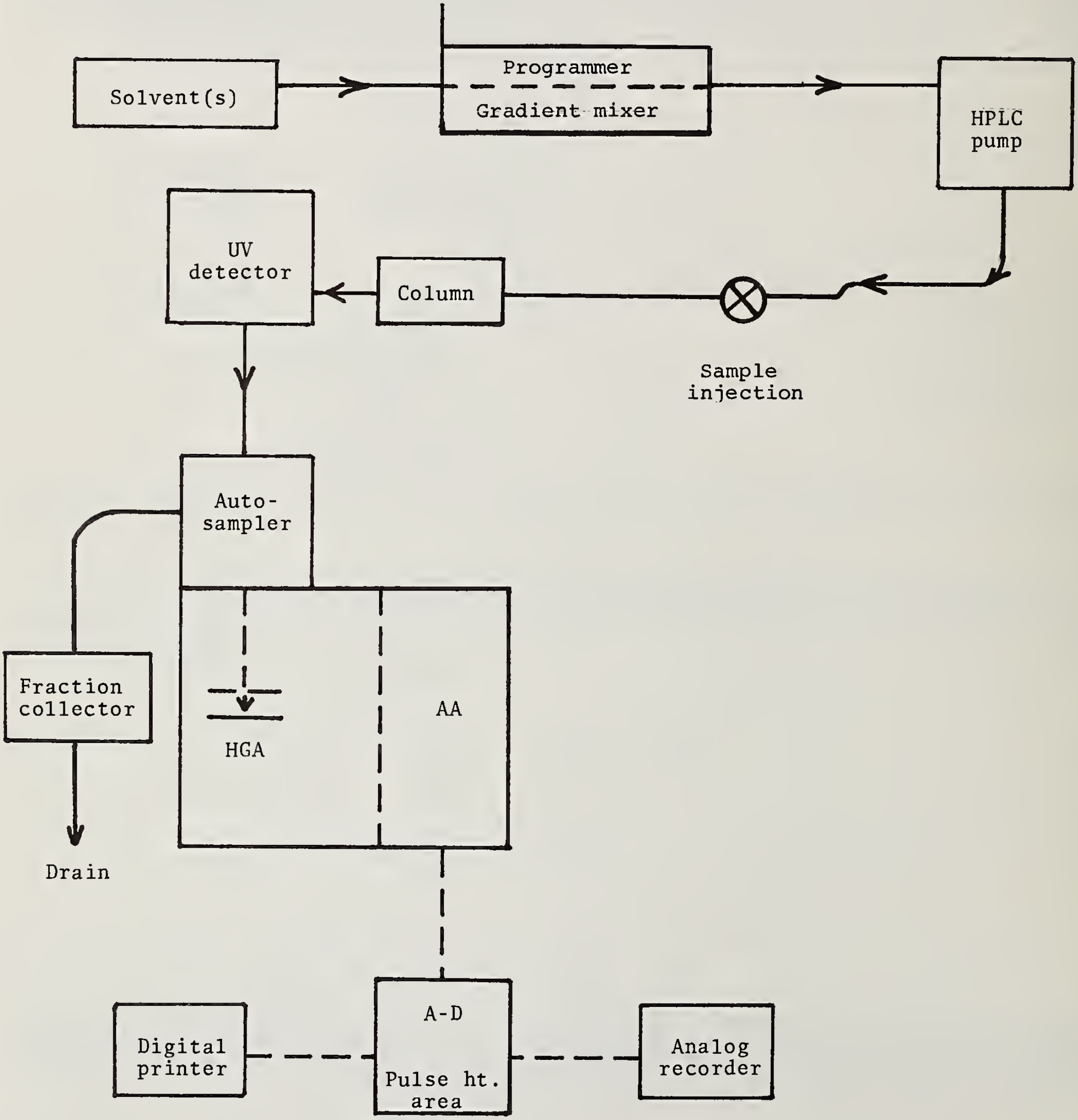


Figure 6. Schematic of high-pressure liquid chromatograph-flameless atomic absorption detector, HPLC-GFAA, system showing ancilliary UV detector (see reference 25).

The HPLC-GFAA system has already proven to be a valuable tool for speciation of trace organometals in organic and aqueous media. Absorption, reverse bonded-phase, ion-exchange, and size exclusion (gel permeation) type columns all perform efficiently with the element-specific automatic AA detection scheme. Of importance, the procedure is nearly nondestructive, requiring only 3 to 10 percent of total effluent stream for the pulsed AA analysis. This feature holds promise for recovery of separated metal "tagged" eluants in suitable solvents for additional off-line molecular characterization by sophisticated optical, mass, or magnetic resonance spectrometry. An example of our preliminary application of HPLC-GFAA to speciation of lead in waste oil is described below.

Another separation technique, that of thin-layer chromatography (TLC), has recently been applied to detection of trace lead in oils [26] and offers general utility for other metals also. Separated spots on TLC plates can be taken up for quantitation by element-specific or compound-specific characterization methods [27]. Though not currently employed in the NBS metals speciation work, such procedures are of interest as inexpensive, supportive characterization protocols.

Preliminary Results on the Speciation of Lead in Used Oil - Future Directions

A sample of used automotive oil was provided by the NBS Recycled Oil Program Office. As anticipated from commercial re-refining data [2], one batch of waste oil containing 1.4 percent as total lead showed a reduction to 0.7 percent total lead content following ultracentrifugation in 1:1 benzene solution. Optical microscopic examination of the supernatant oil revealed a stable suspension of uniform, spherical opaque particles of less than 0.3 micron size.

Samples of the centrifuged oil were further diluted and injected into the HPLC-GFAA system for lead-specific analysis. Figure 7 illustrates the simple experimental protocol employed. A number of batches of supernatant oil were examined, employing both isochratic and solvent gradient flow programs designed to establish optimal solvent strengths (polarities) consistent with efficient column performance [24]. Earlier we demonstrated [25] that solvent pairs of low strength (hexane: methylene chloride = 95:5) served to provide useful retention volumes (V_R) on a 10 micron silica gel column for a high molecular weight, nonpolar organolead, hexaphenyldilead. In the present survey, solvent pairs of much higher strength, such as methylene chloride-acetonitrile, were applied in order to insure elution of both higher molecular weight and polar lead-containing components.

Preliminary results show that the dissolved lead species can be eluted as two or more distinct peaks, depending upon the solvent program used and the column loading incurred by sample amount. Figure 8 depicts a HPLC-GFAA chromatogram for one methylene chloride-acetonitrile step-gradient program run. It is noteworthy that in this experiment, as with all the others, no column blockage arose as a result of the presence of the submicron suspension previously noted. Neither did frequent lead "spikes", nor high lead background, appear between separated peaks of lead-containing eluants. We conclude that the fine particle suspension moves through the HPLC column with little difficulty and that very little lead content is associated with the particles themselves.

The two lead-containing peaks shown in figure 8 and those obtained similarly in other runs cannot yet be identified but are estimated to comprise about 65 percent of the residual dissolved lead not removed by ultracentrifugation. Inasmuch as no collection of crankcase oil samples has yet been speciated for lead, we cannot yet infer that this result is typical.

Clearly, several molecular lead species, possibly organoleads, are present in the centrifuged samples obtained from one sample of crankcase oil. Compared with authentic organolead compounds (e.g., hexaphenyldilead or triphenyllead chloride), the chromatographic peak shapes found support this view. However, the much larger retention volumes (>20 ml) obtained here with stronger solvents suggest that these eluants must be of high molecular weight and/or polar character as oil-soluble complexes.

The concurrent use of an ultraviolet detector [(UV) operating at 254 nm] with the AA detector (figure 8) indicates that significant chromophores may be associated to one of the lead-containing peaks (V_R = 27 mL). Recalling that most automotive lubricating oils are treated with performance "additive packages" containing a variety of metal-solubilizing ligands of the type [1,2] listed in table 3, it is apparent that additional work characterizing ligands associated with soluble lead in waste oils will be complicated but feasible. In particular, sufficient phenyl-,

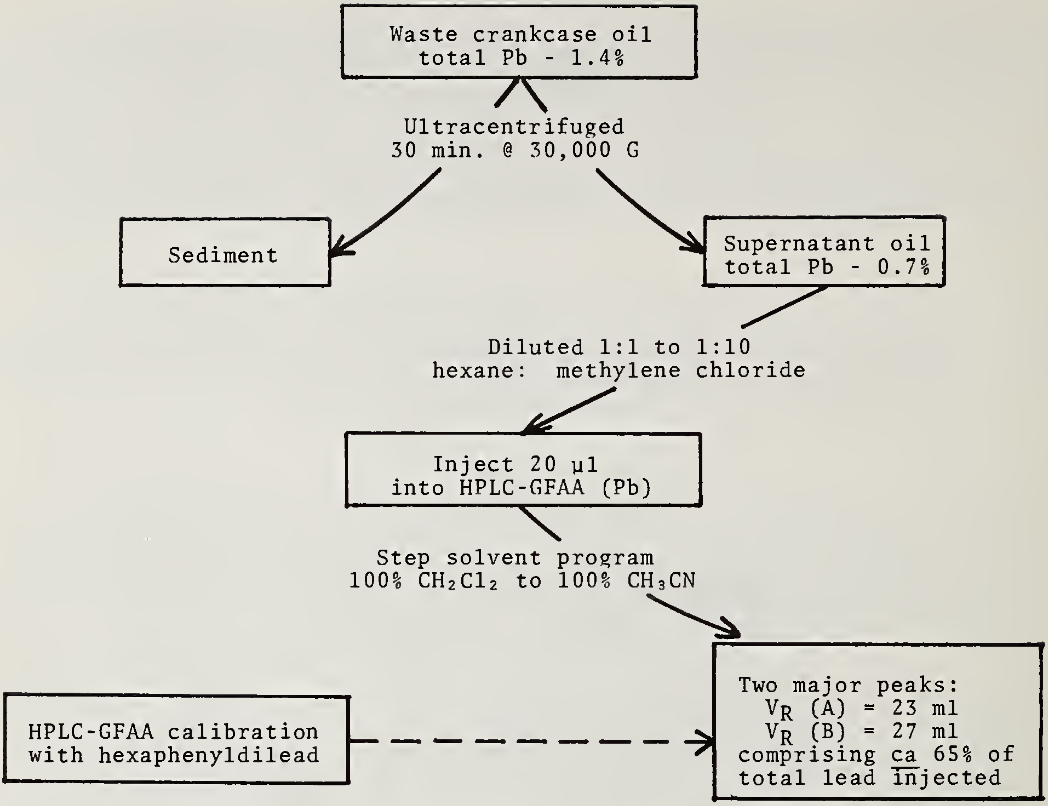


Figure 7. Flow chart indicates experimental protocol for HPLC-GFAA speciation of dissolved lead compounds in waste crankcase oil.

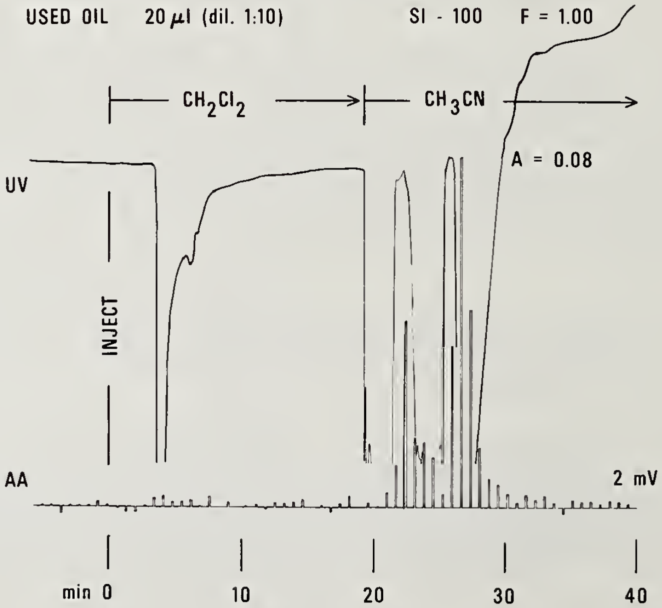
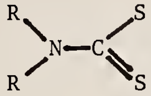
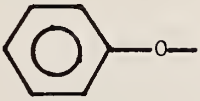
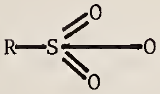
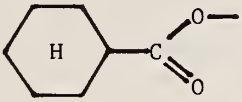


Figure 8. HPLC-GFAA chromatogram of used oil sample indicates elution of two major lead-containing components (AA detector) and several strong UV absorbing eluants, one of which may contain lead.

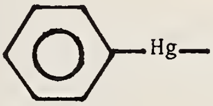
Table 3. Ligands Associated With Metal Additives

LUBRICANTS

	Sb, Mo, Zn (Ex: R = butyl)
	Ba, Zn
	Ba (Ex: R = alkyl)
	Pb
Phosphates Thiophosphates Organodithiophosphates	Ba, Cd, Mo, Zn (Ex: Alkyl)

Organometals:

(Butyl)$_3$Sn—



FUELS

Organometals:

(Ethyl)$_4$Pb

(Methyl)$_n$Pb(ethyl)$_{4-n}$

phenoxy-, dithiocarbamate, arylsulfonate-, phenate-, or cyclopentadienyl-, etc. groups may survive engine service temperatures to yield discrete organometal or metallo-organic complexes. Many of these, though highly polar, would form hydrocarbon-soluble molecules possessing strongly UV-absorbing moieties [28].

Among immediate objectives for future work should be improvements in the HPLC-GFAA speciation scheme, coupled with additional supportive compound-specific detectors operating coincidently with the GFAA detector. Of special benefit to identification of complexing ligands, particularly as a basis for comparison between new and re-refined oils, is addition of a spectrofluorimetric detector operating in either continuous or stopped-flow modes. This approach will accelerate our progress. The potential for precise characterization of many classes of chromophoric organic structures is well documented in another presentation at this conference [29]. Applications of spectrofluorimetry to speciation of trace biogenic organometallic species was shown to be highly successful at the NBS [30] in an earlier study.

Future work must also exploit another feature of the HPLC-GFAA speciation method. The availability of lead-containing molecules in oils for leaching into fresh or salt water media can be assessed by use of reverse-bonded-phase HPLC columns operated with acetonitrile-water or methanol-water solvent-gradient programs [31]. By coordinating these studies with the oil-based ligand characterization studies above, a partition chemistry relating forms and rates of interchange of lead-containing bioactive molecules from oils to waters can be established.

Finally, studies aimed to evaluate bioavailability of lead-containing (or other metal-containing) species in waste, re-refined, or "virgin" oils are required to provide comparisons for environmental impact/assessment from these different technological sources. In particular, the biotransformations of characterized lead-containing molecules leached into ground waters from such oils must form the focus of such research. Thereby, presence or absence of certain desirable "telltale" chemical features can be established or standardized and the results applied to redesigning additive packages or refining processes supporting production of such necessary commodities.

Acknowledgements

The authors gratefully acknowledge partial financial support from the NBS Office of Air and Water Measurement and the NBS Recycled Oil Program Office. We thank Mr. D. A. Becker and his associates for ultracentrifuged samples of oils; the assistance of Dr. W. P. Iverson was invaluable for microscopic characterization of the ultracentrifuged oils. We thank Mr. E. J. Parks for pointing out several valuable separation procedures for trace metal ions in oils.

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National Bureau of Standards Special Publication 556. Proceedings of a Workshop on Measurements and Standards for Recycled Oil - II held at NBS, Gaithersburg, Maryland, November 29 and 30, 1977. (Issued September 1979)

THE ANALYSIS OF SOME RESIDUAL FUEL AND WASTE LUBRICATING OILS BY A HIGH-PERFORMANCE LIQUID CHROMATOGRAPHIC PROCEDURE

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Introduction

In recent years there has been increasing concern about the possible harmful effects caused by some of the organic compounds that are found in the environment. The polynuclear aromatic hydrocarbons (PAH) are a class of pollutants that are of great concern because some of them, such as benzo(a)pyrene, benz(a)anthracene, and dibenz(ah)anthracene, have been shown to possess carcinogenic [1-3][1] and mutagenic [4,5] properties. PAH have been found in air, food, and water from such sources as engine exhaust; the burning of fossil fuels; tobacco smoke; and natural sources, such as petroleum-based oils.

Most schemes for the analysis of PAH involve an extraction, separation (sample cleanup using either open-column or thin-layer chromatography), and qualitative and quantitative analyses steps. Most early investigators used ultraviolet absorption (UV) or fluorescence spectroscopy for quantitative analysis of such extracts [6]. Later, gas chromatography (GC) became the analytical technique of choice because it allowed further isolation of the analyte and gave some qualitative information in the form of the chromatographic retention time. Recently, the use of capillary GC has become very popular and has been used in the analysis of PAH in engine exhausts [17], anthracene oil [8], cigarette smoke [9], soot samples [10], and sediments [11]. Janini, et al. [12] have discussed some of the difficulties associated with separating complex mixtures of PAH by GC and suggested the use of a nematic liquid crystal column for such analyses. Such columns have high bleed rates and relatively short lifetimes. The high bleed rates also make their use in gas chromatography-mass spectrometry (GC-MS) impractical.

GC-MS is generally recognized as being the most powerful tool available to the analyst for identifying trace components in complex mixtures. GC-MS has been used in the analysis of complex PAH mixtures [13-15] but does have certain limitations. Isomeric PAH are not readily distinguishable from their mass spectra alone and are often not resolved chromatographically. For example, 1,2-benzanthracene, chrysene, and naphthacene give essentially the same mass spectrum and are difficult to separate by GC.

High-performance liquid chromatography (HPLC) offers the analyst another powerful tool for use in the analysis of PAH. Larger sample sizes can be accommodated, and less sample pretreatment is necessary. Columns are available that provide rapid and efficient resolution of complex mixtures of PAH. However, the sensitivity and selectivity of the detection systems that may be used in conjunction with HPLC are the major advantages afforded by the technique. Christensen and May [16] have recently shown that ultraviolet and fluorescence detectors are comparably sensitive and more selective for PAH than the mass spectrometer.

In this presentation we will describe a HPLC method that we have developed for the analysis of benzo(a)pyrene in some waste lubricating and residual fuel oils.

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[1] Underlined numbers in brackets indicate the literature references at the end of this paper.

Experimental

Instrumentation. The liquid chromatographic unit used in this work consisted of two Model 6000A pumps and a solvent programmer (Waters Associates, Milford, MA); a Model 7120 sample injector (Rheodyne, Berkeley, CA); a Model 440, 254 nm filter photometric detector (Waters Associates, Milford, MA); and a Mark I spectrofluorometer, equipped with a 10 µL flow cell (Farrand Optical Company, Inc., Valhalla, NY). The columns used were µBondapa, NH_2 and µBondapak C_{18}. Data were recorded using a strip chart recorder.

Methodology. The HPLC method described here utilizes two chromatographic columns for the isolation and separation of individual PAH. The data presented in table 1 show that chromatography on µBondapak NH_2 provides a separation of PAH that is based on ring size. Alkylation has little effect on retention. On the other hand, chromatography on µBondapak C_{18} provides a separation that is related to the aqueous solubilities of the respective PAH. Parent PAH can be separated from both their alkylated homologs and their structural isomers. Efficient isolation of individual PAH from complex mixtures may be achieved by using both of these columns in an integrated procedure.

The concentration of benzo(a)pyrene in some residual fuel and recycled oil samples was measured using the procedure described below.

1. The density of each sample was determined by weighing 10 mL of oil on a Mettler balance. The volume was measured in a class "A" volumetric flask.

2. Dilution and filtration of the samples were accomplished by filling a 1.33 mL stainless steel sample loop with the oil to be measured, and pumping the contents of the loop through a 2 µm filter into a 10 mL volumetric flask.

3. Isolation of the five condensed ring PAH fraction was effected by injecting 200 µL of the diluted filtrate onto a 300 x 12 mm µBondapak NH_2 column. The benzo(a)pyrene fraction was collected in a volume of ∿20 mL (see figure 1).

4. This 20 mL was reduced to a volume of between 200 and 500 µL using N_2 purge at room temperature. The volume measurement was made by means of a Hamilton syringe, calibrated to the nearest 5 µL.

5. Separation of benzo(a)pyrene from other five condensed ring PAH and quantification was done by injecting 23 µL of the fractionated oil on a 300 x 4 mm µBondapak C_{18} column. Further selectivity was obtained by monitoring the chromatographic effluent fluorimetrically, with excitation and emission wavelengths optimized for detection of benzo(a)pyrene (ex-295nm, em-400nm). Benzo(a)pyrene was identified in each oil by chromatographic retention volume and fluorescence emission spectrus, as shown in figure 2.

Results. The concentration of benzo(a)pyrene in each sample was calculated using the following equation:

$$\text{Concentration of Benzo(a)pyrene in oil} = \frac{(\text{Peak ht.})\ (K_1)(K_2)}{(K_3)(K_4)(V)(\rho)}$$

Peak Ht. = Height of benzo(a)pyrene chromatographic peak in mm.

V = 1.33 mL, volume of oil analyzed

ρ = Density of oil in g/mL

K_1 = Unfractionated dilution factor; the ratio of the volume to which the filtered oil was diluted to the volume which was injected for fractionation (10/.2).

K_2 = Fractionated dilution factor; the ratio of the volume to which the five condensed ring PAH fraction was concentrated to the volume of the sample injected for analysis (x/.023).

K_3 = Fluorescence response factor for benzo(a)pyrene in mm/µg.

Table 1. Comparison of Retention Indices (I) of Aromatic Hydrocarbons on Several Liquid Chromatographic Packing Materials [17]

	Logarithm of the retention index (log I)	
	μBondapak NH_2 (hexane)	μBondapak C_{18} (acetonitrile-water)
One-ring aromatics		
Benzene	1.00	1.00
Toluene	1.00	1.51
m-Xylene	1.00	2.16
1,2,4-Trimethylbenzene	1.00	2.65
n-Pentylbenzene	0.71	3.80
Nonadecylbenzene	0.50	----
Two-ring aromatics		
Naphthalene	2.00	2.00
2-Methylnaphthalene	1.96	2.59
2,3-Dimethylnaphthalene	2.08	3.47
2,6-Dimethylnaphthalene	1.94	3.15
1,5-Dimethylnaphthalene	1.93	3.00
2,3,6-Trimethylnaphthalene	2.00	3.54
1,4,6,7-Tetramethylnaphthalene	2.05	3.86
2-Ethylnaphthalene	1.92	3.14
Biphenyl	2.25	2.57
3-Ethylbiphenyl	2.20	3.46
Acenaphthalene	2.00	2.73
Three-ring aromatics		
Fluorene	2.61	2.78
Dibenzothiophene	2.75	2.98
Anthracene	2.95	3.02
2-Methylanthracene	2.92	3.63
9,10-Dimethylanthracene	2.95	3.82
Phenanthrene	3.00	3.00
1-Methylphenanthrene	2.98	3.43
p-Terphenyl	3.40	3.94
Four-ring aromatics		
Fluoranthene	3.39	3.42
Benzo[*a*]fluorene	3.46	3.74
Benzo[*b*]fluorene	3.53	3.78
Pyrene	3.26	3.51
Naphthacene	3.93	----
Benzo[*a*]anthracene	4.00	4.00
Triphenylene	4.00	3.82
Chrysene	4.03	3.94
Five-ring and larger aromatics		
Benzo[*a*]pyrene	4.30	4.57
Perylene	4.47	4.43
Benzo [*g,h,i*]perylene	4.61	>5
Indeno[*1,2,3-c,d*]pyrene	4.72	>5
Dibenzo[*a,c*]anthracene	4.93	4.84
Dibenzo[*a,h*]anthracene	4.93	>5
Picene	>5	4.93
Benzo[*b*]chrysene	5.00	5.00

Figure 1. Prefractionation of an oil on μBondapak NH_2. Mobile phase: 5 percent methylene chloride in hexane.

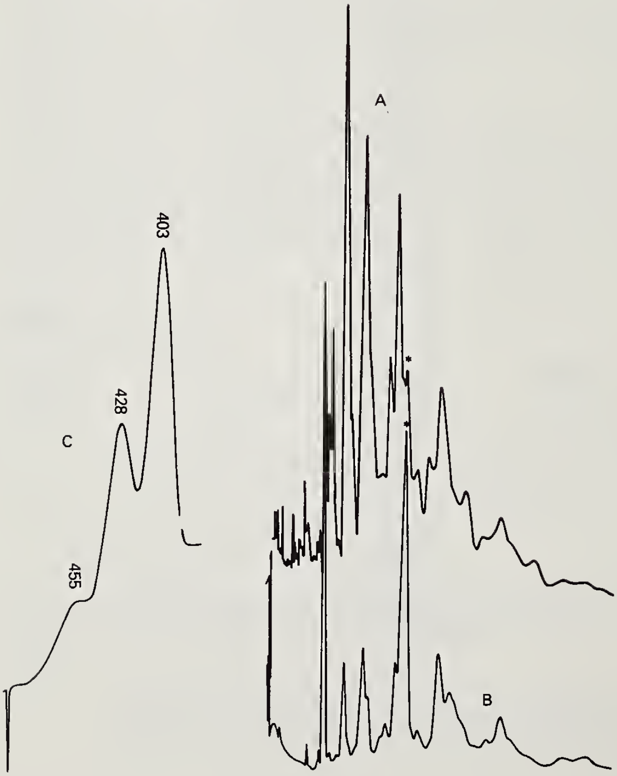

Figure 2. The analysis of a prefractionated oil on μBondapak C_{18}. A - ultraviolet elution profile at 254 nm; B - fluorescence elution profile with excitation at 290 nm and the emission monitored at 403 nm; C - Fluorescence emission spectrum of benzo(a)pyrene. Mobile phase: 30 percent water in acetonitrile.

K_4 = Attenuation factor; correction for the difference in detector attenuation used when calculating K_3 and that used for the analysis.

Data on the concentration of benzo(a)pyrene in several oils are given in table 2.

Table 2. Data on Benzo(a)pyrene Concentrations in Some Fuel and Motor Oils

Sample number	Description of oil	Benzo(a)pyrene conc. (µg/g)
201	No. 6 residual fuel oil (Bunker C, source A)	27 ± 3
203	Used motor oil (composite)	12 ± 3
212	Service station waste, source A	3.2 ± 0.6
213	No. 6 virgin residual fuel oil, source B	35 ± 2
214	No. 5 virgin residual fuel oil, source B	2.8 ± 0.1
220	No. 4 virgin residual fuel oil, source A	2.1 ± 0.3
221	No. 6 virgin residual fuel oil, source B	34 ± 1
222	Re-refiner's feedstock waste oil, period A	12 ± 2
223	Used synthetic motor oil (23,000 miles)	16 ± 1
224	Used diesel motor oil	<0.15
225	No. 5 recycled fuel oil, source A	8.4 ± 0.8
226	Re-refiner's feedstock waste oil, period B	8.8 ± 1.2
227	No. 5 recycled fuel oil, source B	3.7 ± 0.4
228	Unused re-refined motor oil	2.1 ± 1.2
229	No. 5 virgin residual fuel oil (duplicate of sample number 214)	3.3 ± 0.6
230	Used motor oil	14 ± 2
231	Service station waste oil, source A	5.2 ± 0.4

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SESSION III. NONLUBRICATING END USES

National Bureau of Standards Special Publication 556. Proceedings of a Workshop on Measurements and Standards for Recycled Oil - II held at NBS, Gaithersburg, Maryland, November 29 and 30, 1977. (Issued September 1979)

CURRENT EPA VIEWS ON AMBIENT AIR LEAD STANDARDS

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Because the author's written presentation was not received by the specified publication deadline, it will not be included in these proceedings.

National Bureau of Standards Special Publication 556. Proceedings of a Workshop on Measurements and Standards for Recycled Oil - II held at NBS, Gaithersburg, Maryland, November 29 and 30, 1977. (Issued September 1979)

AN UPDATED LOOK AT THE FUEL POTENTIAL OF WASTE AUTOMOTIVE OIL

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Abstract

Under an Environmental Protection Agency contract, GCA/Technology Division evaluated the technical, economic, and environmental feasibility of reusing automotive waste oil as a fuel [1] [1] The study found that the total quantity of automotive waste oil generated annually represents slightly less than 0.5 percent of total U.S. fossil fuel production. Certain precautions are necessary to prevent potential problems with boiler operation and increased levels of stack emissions, notably particulates and lead. Blending small quantities of untreated waste oil with virgin stocks appears feasible for large utility and industrial boilers, while some pretreatment of oil was found desirable for smaller boilers. The first portion of the paper discusses major findings of this study.

The second portion of the paper addresses methods which can be used to evaluate local or Statewide barriers and problems which hinder effective reuse of waste oil. Such a program is an essential step towards establishing an effective local or regional policy for reuse of waste oil.

Potential Supply and Marketability of Waste Oil as Fuel

The quantity of waste lubricating oil potentially available for reuse as fuel was estimated by GCA based on regional per capita consumption figures [2]. Table 1 indicates that nationwide approximately 715 million gallons of automotive and 330 million gallons of industrial oils are potentially available for reuse. Industrial lubricants, roughly 30 percent of potential waste oil supply, were found to vary widely in physical properties and are not as readily reprocessed to fuel as automotive waste oil. For this reason, GCA focused on reuse of waste automotive lubricants as fuel.

Table 1. National Waste Oil Supply Projections by U.S. Census Regions, 1975

Census region	Estimated waste oil supply	
	Vehicle x 10^6 gal.	Industrial x 10^6 gal.
New England	29.2	11.8
Middle Atlantic	86.0	62.0
South Atlantic	84.7	34.8
East South Central	50.8	19.6
East North Central	149.0	95.5
West North Central	78.7	15.3
West South Central	89.2	56.0
Mountain	42.6	8.6
Pacific	105.0	27.6
U.S. Total	715	331

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[1] Underlined numbers in brackets indicate the literature references at the end of this paper.

The quantity of waste automotive oil available for reuse represents less than 0.5 percent of domestic fossil fuel production, and about 5 percent of total oil consumed by domestic steam-electric power plants. Although the waste oil supply is concentrated in high-density population centers, the regularity and quality of this supply is variable. Consequently, it will be most effective to reuse waste oil as supplemental or reserve fuel until longer-term systems are developed to promote supply regularity through improved collection, storage, and distribution.

To assess the demand for waste oil as fuel, questionnaires were distributed to potential electric utility and industrial users. These data were supplemented by field site visits and personal interviews. Electric power generation consumes about 25 percent of domestic energy production and represents a significant potential market for waste oil fuels. Several power plants were found to be combusting waste oil routinely or testing waste oils for routine use. Eighty-six percent of the 68 electric utilities responding to the GCA survey felt some form of waste oil could be used at their facilities. Response to GCA's survey by industry was less positive towards waste oil reuse as fuel. Industrial sources are diverse in fuel requirements and requested a more detailed analysis of fuel composition and potential adverse affects on a particular process.

In most regions of the country, a network of private waste oil collection firms is currently collecting waste oils from automotive service stations. A recent GCA study of hazardous waste generation [3] found that roughly 50 percent of waste oil collected in Massachusetts is reused as fuel, while 23 percent is re-refined as lube oil. Waste oil collectors are currently paying service stations $0.02 to $0.07 per gallon for the oil and selling untreated or minimally treated oil to commercial boiler facilities for about $0.20 to $0.30 per gallon. This price structure compares locally to $0.35 to $0.40 for distillate and $0.30 to $0.35 for residual fuel oils.

Institutional/Legal Factors Influencing Waste Oil Reuse

Institutional and legal factors also influence reuse of waste oil as supplemental fuel. State waste oil management programs may either aid or hinder reuse as fuel. For example, Nebraska stipulates a maximum water and solids content and prohibits presence of highly flammable aromatic hydrocarbons in any waste oil reused as fuel. Local and Federal air pollution standards may limit quantity of waste oil combusted. Relatively high ash content of untreated waste oil can increase particulate emissions. Blending waste oil with residual oil can result in decreased sulfur oxide emissions since waste oil is relatively free of sulfur. Most power plants are restricted in percent sulfur allowed in fuel, and blending with waste oil could help meet sulfur regulations.

Waste Oil Composition and Technical Feasibility of Reuse as Fuel

Waste automotive oils also contain small amounts of transmission fluids, differential gear lubricants, hydraulic oils, and solvents. In addition, some or all of the following substances are commonly present in the waste oil mixture: (1) functional oil additives; (2) iron "fines" from engine fretting and wear; (3) gasoline components, oxidized materials, atmospheric dust, and combustion products transferred to the oil via piston "blowby;" (4) sedimentary materials which were formerly internal engine deposits; and (5) water and other contaminants introduced to storage tanks.

GCA summarized physical and chemical properties of waste oil and waste oil/virgin oil blends, based on published and in-house data. Tables 2 and 3 summarize physical and chemical properties of untreated waste oil, virgin distillate and residual oils, and various blends of waste and virgin oils. Technical impacts of using blended oils as a boiler fuel were assessed based on these data, results of the survey questionnaire, and interviews with potential utility and industrial users of waste oil products.

Viscosity of a 10-percent waste oil/distillate oil blend was found to be three times that of virgin distillate oil. However, centrifugal pumps normally used for distillate oil are capable of handling the 340 centistoke viscosity of a 10-percent waste oil/distillate oil blend. Waste oil/residual oil blends of 10 percent are somewhat less viscous than virgin residual oil.

Pour point of 10-percent waste oil/distillate oil and waste oil/residual oil blends is slightly less than pour points of the respective virgin fuels. Waste oils contain pour point depressants to prevent solidification at cold temperatures.

Table 2. Properties of Waste Oil/Distillate Oil Blends[a]

Property	Distillate oil	5 percent waste oil	25 percent waste oil	100 percent waste oil
Gravity, °API @ 60° F	37.8	37.1	34.4	24.0
Viscosity, centistokes	3.0	4.0	11.0	99.0
Pour point, °F	-12.5	-13.6	-18.1	-35
Flash point, °F	165	171.5	197.5	295
Heating value, Btu/lb.	19,020	18,891	18,374	16,436
BS&W, vol. percent	0.05	0.60	2.79	11.0
Sulfur, wt. percent	0.310	0.316	0.340	0.430
Ash, wt. percent	0.0025	0.098	0.479	1.91

[a] Median values, obtained from literature review.

Table 3. Properties of Waste Oil/Residual Oil Blends[a]

Property	Residual oil	5 percent waste oil	25 percent waste oil	100 percent waste oil
Gravity, °API @ 60° F	13.2	13.7	15.9	24.0
Viscosity, centistokes	379	345	288	99.0
Pour point, °F	52.5	48.1	30.6	-35
Flash point, °F	210	214	231	295
Heating values, Btu/lb.	18,945	18,820	18,318	16,436
BS&W, vol. percent	1.00	1.50	3.50	11.0
Sulfur, wt. percent	2.15	2.06	1.72	0.43
Ash, wt. percent	0.25	0.33	0.67	1.91
Calcium, ppm	47.9	138.0	498	1,850
Sodium, ppm	240.5	236	220	158.0
Iron, ppm	120.3	166	346	1,025
Magnesium, ppm	14.2	41.4	150.4	559
Lead, ppm	2.9	303	1,502	6,000
Vanadium, ppm	190.5	182	148.1	21

[a] Median values, obtained from a review of current literature.

Flash point of 100-percent waste oil is higher than either distillate or residual oils, indicating that no additional safety requirements are necessary for fuel handling. Heating value of waste oil varies slightly due to presence of bottom sludge and water (BS&W), but waste oil blends up to 25 percent were found to vary insignificantly in heating value from virgin distillate and residual oils.

Sediment and water content of waste oil/distillate residual oil blends is noticeably greater than virgin fuels, even at low blending ratios. Presence of water and sediment can cause certain operational problems such as strainer clogging, excessive wear of positive displacement pumps, erosion of burner nozzle tips, and burner flameout. These potential problems have been avoided by wide orifice nozzles with steam atomizing, flame sustaining torches, and minor redesign of fuel straining and fuel lines. As an alternative, waste oil can be pretreated to reduce or remove water and sediment.

Sulfur content of waste oil is low, and blending with high-sulfur residual oils can appreciably reduce both emissions of sulfur oxides and corrosion of internal boiler surfaces.

Ash content of waste oil/distillate-residual oil blends increased significantly above blending ratios of 5 percent for waste oil/distillate oil and 10 percent for waste oil/residual oil blends. Sediment in waste oils contain ash-forming materials which contribute to increased deposition on internal boiler surfaces and increased particulate emissions. Soot blowing, water washing, and manual cleaning are used to effectively remove internal ash deposits; particulate emissions are regulated by local and Federal agencies.

Lead in waste oils was found to be 0.1 to 1.0 percent by weight and, consequently, lead content of waste oil/virgin fuel blends increases appreciably with blending ratio. This is an area of concern as lead has been identified as a hazardous air pollutant and will be discussed further under environmental effects. Lead also contributes to deposition on internal boiler surfaces through fouling of heat exchange surfaces.

Other trace elements are present in waste oil in higher concentrations than in virgin fuels. Blending waste oil with virgin fuels may somewhat increase deposition on boiler surfaces of sodium, calcium, barium, zinc, and phosphorus, depending on blending ratios. Small increases in vanadium and iron could also cause increased corrosion of internal boiler surfaces. However, negative impacts of these elements in waste oil blends of 5 percent are considered minimal.

Environmental Impacts of Untreated Waste Oil Fuel Combustion

Blending waste oil with residual oil will reduce air emissions of sulfur, silicon, sodium, vanadium, and nickel. Emissions of other trace metals will be slightly increased: magnesium, calcium, iron, copper, barium, zinc, phosphorus, silver, tin, chromium, and lead. Increased emissions of trace elements were determined to be minimal with small blending ratios of waste oil (less than 5 percent), except for lead. Lead has been identified by the Environmental Protection Agency as a hazardous pollutant, and ambient air standards for lead will soon be promulgated.

Lead content of untreated waste oil varies widely from about 0.1 to 1.0 percent by weight, a result of leaded automotive fuels. Increased use of unleaded gasoline is expected to reduce lead content of waste oils, but no appreciable decrease has been measured to date.

To evaluate the potential lead emission problem, ground-level lead concentrations were calculated for a 560 megawatt power plant firing a blend of 5 percent waste oil and 95 percent No. 6 fuel oil. The "worst case" situation was assessed by computer diffusion modeling based on a lead content of 1.0 percent and the highest percent of lead emissions reported in the literature: 50 percent of lead present in the fuel. Isopleths of average ground level lead concentrations for typical December wind patterns at a New England power plant site are shown in figure 1. Maximum calculated lead concentrations are 0.15 $\mu g/m^3$, an order of magnitude less than 1.5 $\mu g/m^3$ lead standard under consideration by the Environmental Protection Agency. However, the safe level of lead in ambient air is unknown, and the 1.5 $\mu g/m^3$ level has only been proposed as a possible safe level. The diffusion modeling does indicate that lead emissions are not a substantial problem with low blending ratios.

Ground-level lead concentrations shown in figure 1 were calculated for a power plant with no air pollution control devices. Lead particles in flue gas are submicron in size, and a very efficient particulate control device would be necessary to significantly reduce lead emissions. High-efficiency electrostatic precipitators or fabric filters can provide high particle removals necessary to control lead emissions. These control devices are not commonly required for power plants burning oil, but are normally installed on newer, coal-fired boilers.

Reducing Technical and Environmental Impacts

Adverse technical impacts of combusting waste oil can be reduced by either modifying boiler fuel handling equipment or by improving waste oil quality by pretreatment. Potential environmental effects of increased particulate and lead emissions can also be reduced by pretreatment techniques or use of high-efficiency air pollution control devices. It is unlikely that any utility would install expensive pollution control devices solely to control emissions from waste oil combustion. However, when such control devices are already installed, environmental impacts of waste oil reuse as fuel will be substantially reduced.

Commercially available, high-level pretreatment processes--vacuum distillation or solvent extraction--can remove essentially all waste oil contaminants, including lead and other trace elements. Conventional acid-clay re-refining was not considered in this analysis of high-level treatment because of disposal problems associated with lead-containing waste products. Total costs for high-level vacuum distillation or solvent extraction pretreatment were estimated at $0.19 per gallon (updated to 1977)

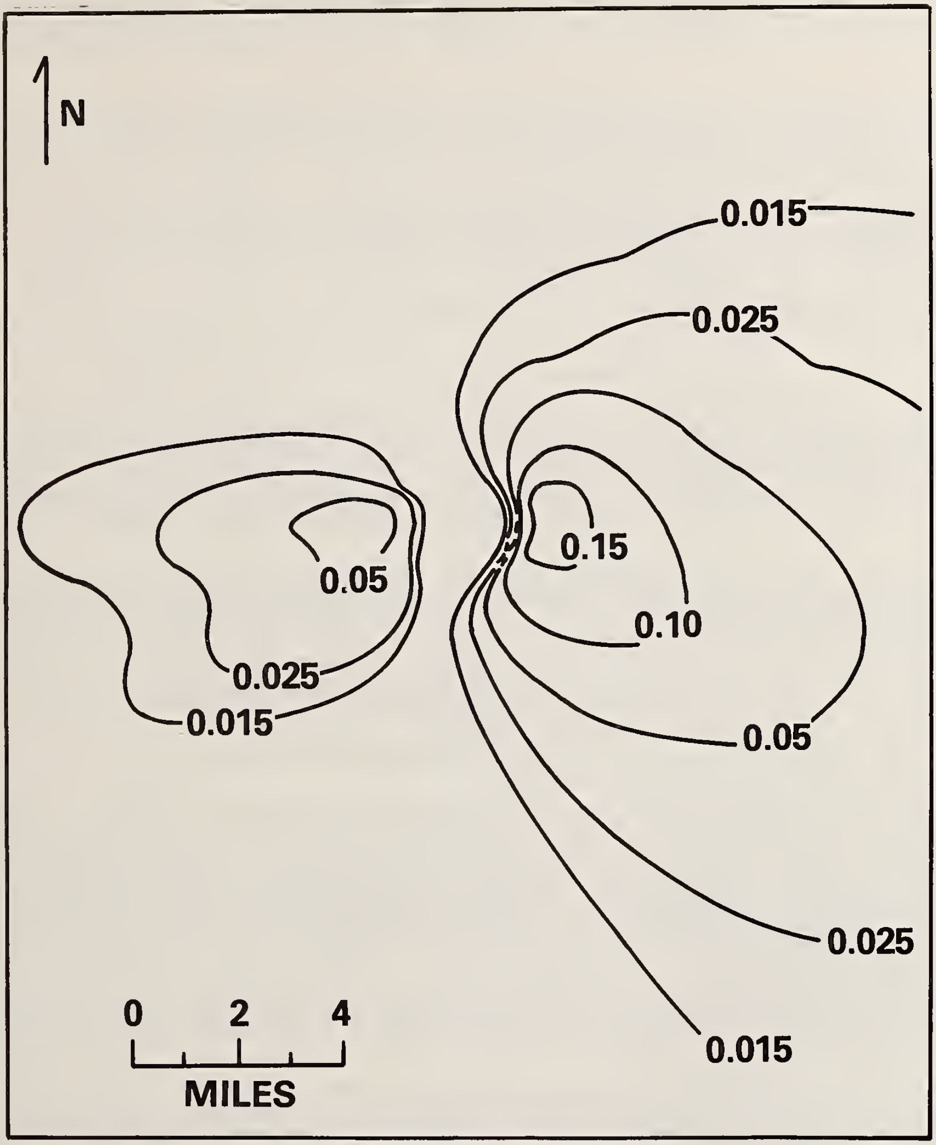

Figure 1. Isopleths of average ground-level concentration of lead at a large New England power plant burning a 5-percent waste oil/residual oil blend. Units are μg/m³.

by appropriate cost index). Since these processes are generally capable of restoring the quality of waste oil to the virgin state, they would be most likely employed for re-refining waste oil to lubricating oil, not fuel.

Low-level pretreatment techniques include gravity settling and centrifugation for reduction of unbound water and sediment. Trace metals, including lead, are present as very small particles which are virtually unaffected by gravity settling. Centrifugation, preferably preceded by addition of a demulsifier, can improve removal of sediment and water over gravity settling to about 1.5 percent from the typical 5 to 10 percent levels found in untreated waste oil. Total costs were estimated at $0.15 to $0.17 per gallon (updated to 1977) for either settling or centrifugation.

Study Conclusions

The GCA study identified three options for reuse of waste oil as fuel which would add to energy supplies while minimizing adverse environmental and technical impacts:

- Large users, especially utilities, could blend small quantities (5 percent) of either untreated or low-level treated waste oil with existing energy sources.
- Medium-sized users with existing high-efficiency emission control equipment could blend higher amounts of high-treated or low-treated waste oil with existing fuel sources.
- High-treated waste oil could be combusted by itself by small users.

Evaluating Reuse of Waste Oil on the Local Level

In recent years, a number of Federally sponsored research programs have identified and discussed reuse options for waste oil. Most of these programs, including the GCA study, are general feasibility studies which compare merits of each reuse option on a national basis. The next logical step is an evaluation at the local level since waste oil reuse policy is normally formulated and implemented on a Statewide or regional basis.

GCA is conducting a study for the State of Massachusetts to document and evaluate existing barriers to effective waste oil reuse specific to Massachusetts. Potential waste oil supply is under evaluation through an updating of earlier studies [3,4], evaluation of permitted waste oil collectors, contact with industry representatives, and analysis of current trends of waste automotive oil generation. Recovery and reuse mechanisms which are practicable in Massachusetts will be identified, focusing on existing facilities and distribution patterns. Specifically, re-refining as lube oil and reuse as fuel in several new municipal, steam-generating incinerators, appear to be viable alternatives in Massachusetts.

The major thrust of the ongoing program will identify technical, institutional, environmental, and economic barriers which currently restrict effective reuse of waste oil in Massachusetts. Technical barriers are analogous to problems identified in the general, nationwide study described in this paper, but the Massachusetts study will document the extent to which these problems have/will occur in local facilities burning waste oil. Through contacts made with waste oil haulers and users during the Massachusetts Hazardous Waste Survey, GCA will document the extent to which technical problems actually do occur in facilities burning or re-refining waste oil in Massachusetts. Waste oil haulers often mix oil from several sources. Efforts will be made to correlate the extent and types of process upset with various waste oil sources.

The technical applicability of waste oil reuse as fuel in existing municipal incinerators in the State will be investigated by contact and personal interviews at these facilities and the responsible engineering firms and/or manufacturers of combuston equipment. This option may be important in the future as the State proceeds with plans for regional resource recovery plants for municipal refuse.

Institutional barriers to effective resource recovery from waste oils in Massachusetts will be identified and evaluated. For example, marketability and public acceptance of re-refined lube oil have been hampered in the past by Federal labeling requirements. Safety codes and Occupational Safety and Health Administration (OSHA) regulations may restrict use of unprocessed waste oils as fuel, and this possibility will be investigated through contact with the OSHA, operators of boilers, and consulting local city safety ordinances or codes. GCA will determine whether insurance policies covering either capital equipment or employee safety would be violated by use of unprocessed waste oil by contact and/or interviews with the Massachusetts Insurance Commissioner's Office, insurance companies, and the responsible personnel at several institutions which are using, or may use, waste oil as fuel. Also to be assessed is the effectiveness of public automotive waste oil receiving stations. The establishment of these stations has been specified by State law as a way of providing the public with a means of recycling the waste oil from "do-it-yourself" automotive oil changes. The program has maintained a low profile within the State, and its adequacy has not been formally evaluated.

Re-refining waste oils to form lube oils and pretreatment prior to use as fuel will generate an oily sludge which requires proper disposal. Of greater environmental importance is burning of untreated waste oil in combination with distillate oil; unprocessed waste oil is relatively dirty and, thus, particulate emissions will be somewhat increased. This increase must be assessed with respect to current air pollution regulations and ash content of local waste oil.

A major problem in Massachusetts is a strong economic incentive for waste oil reuse as fuel. Waste haulers pay service stations $0.03 to $0.07 per gallon for waste oil and sell collected oil locally as fuel for $0.20 to $0.30 per gallon. Unfortunately, little regard is given to the aforementioned problems with burning untreated waste oil: safety, burner maintenance and operation, and increased air pollution. A nearby facility is capable of restoring waste oil to virgin quality, but can only pay waste collectors about $0.10 per gallon. In the long run, it may be more desirable to re-refine waste oil instead of burning as fuel, but existing pricing structure is currently limiting re-refining. The GCA study will investigate mechanisms for promoting re-refining through a program of interviews with waste collectors and re-refiners and through a study of incentives which could be enacted by the State to promote the favored use option.

The final portion of this study will formulate recommendations to improve reuse of waste oil based on information obtained through the research program. Examples of possible action by the State include:

- require State vehicles and buildings to use recycled petroleum oils to set an example and stimulate the market;
- stimulate reuse by State tax incentives and alteration of any laws or regulations currently restricting reuse as fuel or re-refining as lube oil; and
- implement a program for improved public education and awareness concerning benefits of recycling lube and fuel oil.

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THE BURNING OF USED OIL AS A FUEL IN CEMENT MANUFACTURE

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Introduction

Approximately 200 million imperial gallons of used lubricating oils are sold in Canada each year [1,2].[1] This includes both petroleum oils used for lubricating purposes and other oils having special properties other than lubrication alone, such as hydraulic oils and industrial cutting oils. It has been estimated that at least 42 percent of this volume, about 84 million gallons per year, is potentially recoverable [3]. Presently, approximately 5 to 6 million gallons of used oil are being re-refined annually, and about 21 million gallons are used for road oiling. The remaining 57 to 58 million gallons are used or disposed of in various ways, ranging from being burned as fuel to being incinerated or dumped in sewers and on the ground.

Both the lubricating and energy properties of lubricating oil are retained in the used oil until its disposal, and so it can be recycled as many times as possible without any loss in value. Every gallon of used oil used again for recovery of either property causes a corresponding reduction in demand for an equivalent amount of "new" oil, thereby helping to extend the existence of our crude oil reserves. In terms of the energy value, a figure arrived at by comparing the average energy value of used oil to that of residual fuel oil (presently selling for about $0.30 per imperial gallon), the 84 million gallons of potentially recoverable used oil are worth about $20 million annually. A recent study done for the Environmental Protection Agency has shown that additional energy savings of over 25,000 British thermal units per imperial gallon of oil can be realized by re-refining used oil instead of using it as fuel [4]. This would mean an extra annual saving of the equivalent of about 12 million gallons of fuel oil and bring the total value of the potentially recyclable used oil to about $23 million per year, if it were all recycled once before being used as a fuel. These rough calculations put the value of used oil as a resource into perspective and need to be considered during the formation of programs to deal with the problem of used oil recovery and disposal.

As well as resulting in the loss of a valuable, nonrenewable resource, uncontrolled disposal of used oil can pose a serious threat to both the environment and public health. Used automotive crankcase oil, which makes up the largest part of used oil presently collected, contains both portions of the original additives and impurities acquired during service. In particular, they may contain significant concentrations of heavy metals (particularly lead), bromine, and polynuclear aromatic hydrocarbons. Indiscriminant disposal of these and other oils on land can harm vegetation and contribute to water pollution either by runoff or leaching processes. Burning or incineration without adequate controls can cause the emission of various toxic or potentially carcinogenic gaseous components which may endanger the health of people and animals in contact with them.

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[1] Underlined numbers in brackets indicate the literature references at the end of this paper.

One of the objectives of the Resource Conservation Division of Environment Canada is to promote the recovery and recycling or reuse of used oil in order to conserve our petroleum resources and to minimize the effects of its disposal on the environment. We hope to realize this objective by

(a) assisting other governments and generators of used oil in the development of recovery systems that will remove as much oil as possible from the waste stream (i.e., recovery of oil that is not presently recovered due to such factors as economics, contamination, etc.), and

(b) by assisting other governments in the development of procedures that will ensure the recycling or disposal of used oil in ways that are environmentally acceptable. This involves the assessment and evaluation of the environmental acceptability of end uses, as well as assisting in the improvement of existing technology and the development of new technology and methods for recycling and reusing used oil.

One method of reuse that Environment Canada has investigated and found to be both an environmentally acceptable method of disposal of used oil and one that recovers the energy value of the oil is its use as a fuel in the manufacture of cement. It has been known for some time that cement production, which requires a high heat input, can also act as a sink for many fuel components such as metals and sulfur, etc., by absorption in the cement clinker. In 1974, an experimental program to study the burning of used automotive crankcase oil in a dry process cement kiln was carried out jointly by Environment Canada, the St. Lawrence Cement Company, the Ontario Ministry of the Environment, and the Ontario Research Foundation [5]. The initial study burned 330,000 imperial gallons of used lubricating oil at the St. Lawrence Cement Company plant in Mississauga, Ontario; and, based on the results of this study, the plant has installed the capacity required to burn over 5 million gallons of used oil per year and is presently consuming about 3 million gallons annually.

I would now like to introduce one of the chief investigators of the study, Mr. Larry MacDonald of the St. Lawrence Cement Company, to describe the study and its results to you in more detail.

Cement Manufacture

General Principles

While a variety of raw materials may be used in cement manufacture, materials containing calcium, silicon, aluminum, and iron without an excess of certain other elements are required. These materials are ground to a find powder called raw meal, the chemical composition of which is carefully controlled by proper blending of the various materials. Normally, blending is achieved by grinding all the raw materials together (intergrinding). Raw meals required for wet and dry processes are similar except that the raw meal for the wet process is in the form of a slurry containing approximately 35 percent water, while raw meal for the dry process contains less than 0.5 percent water.

The raw meal is fed into the kiln and is burned to produce an intermediate product called clinker.

The kiln slopes towards the burning zone and rotates slowly, causing the raw material to gradually move into the burning zone. Reactions which occur during gradual heating in the kiln are: evaporation of free water; evolution of combined water; evolution of carbon dioxide from carbonates; and combination of lime with silica, alumina, and iron to form the desired compounds in the intermediate product clinker. These reactions require a final material temperature of 2,650° F.

After cooling, the clinker is ground with gypsum to a fine powder. The final product, called Portland cement, is the basic ingredient of concrete.

The operation of a suspension preheater is illustrated schematically in figure 1. Raw meal is introduced into the duct between the first and second stage cyclones. It is swept with the hot exhaust gas into the uppermost (stage I) cyclones, where gas and material are separated. The raw feed material from the stage I cyclone drops into the duct between the second and third stage cyclones and is again suspended and separated. This procedure is repeated in stages III and IV before the partially

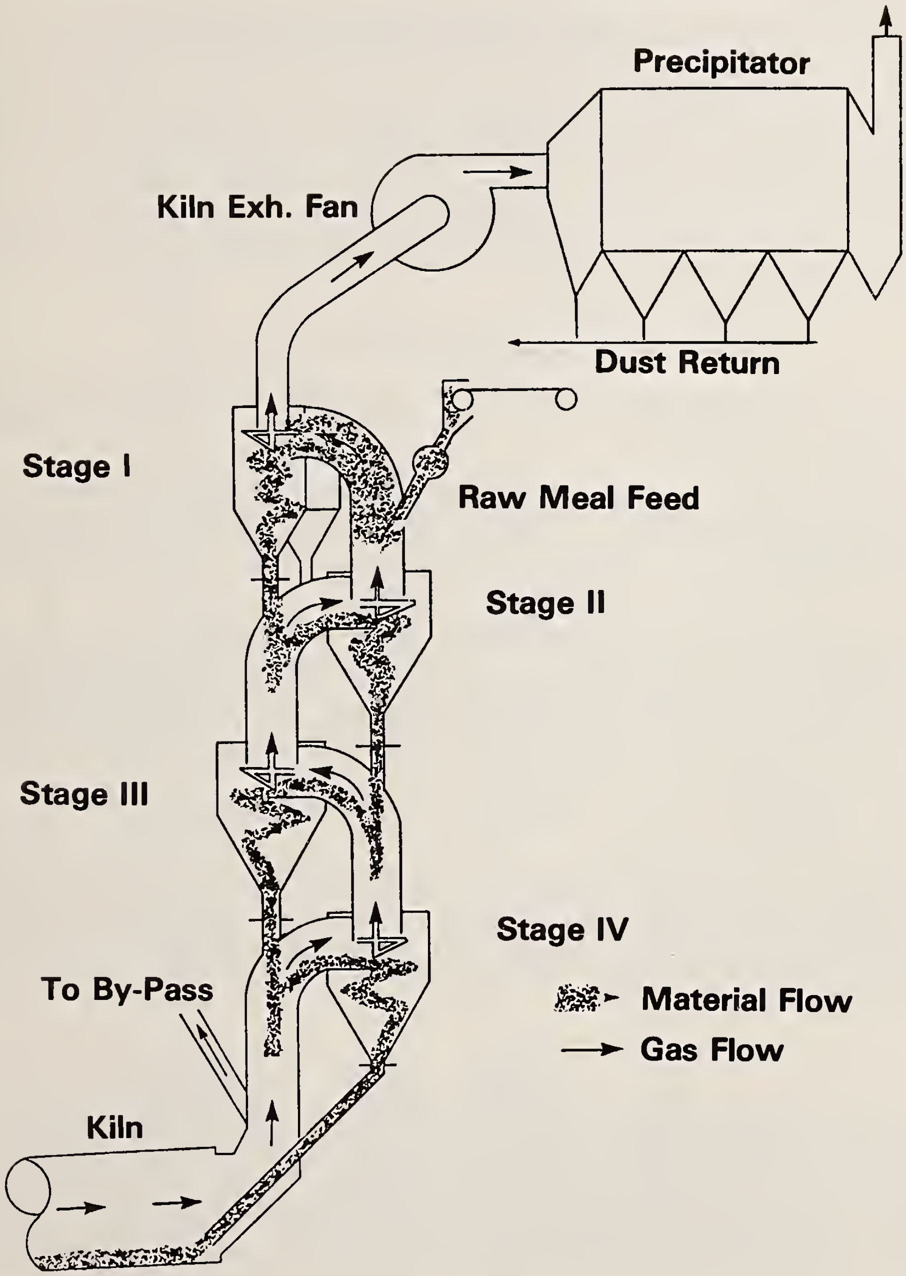

Figure 1. Principle of Fuller-Humboldt suspension preheater.

calcined feed enters the kiln. In the process of passing through these four stages, the raw meal is heated from about 150° F to about 1,450° F.

The lime formed by calcination of the raw meal in the kiln effectively absorbs various elements from the combustion and reaction gases. As with any solid gas-scrubbing procedure, the effectiveness of the removal is a function of the degree of mixing between the solids and the gases.

Volatile constituents (for example, potassium chloride and sodium chloride) in the raw materials and fuels are removed from the solid phases in the hottest zones of the kiln. In a straight kiln, these alkali chlorides condense in the cooler regions as minute particles and are removed from the gas stream by precipitators. Due to the intimate mixing of gases and raw material in the preheater, most of the alkali chlorides condense on incoming raw meal particles and are therefore trapped between the flame and the preheater. This circulation within the gas stream concentrates the alkali chlorides in the gas phase which can cause the preheater to plug. To reduce the alkali chloride buildup, a by-pass is included in the system. The by-pass acts as a vent to bleed off some of the kiln gases rich in alkali chloride. These are passed through a conditioning tower which cools and humidifies the gases to the optimum for precipitator operation.

Experimental

The purpose of this experimental burn was to determine whether used oil could be employed as a fuel in the cement kiln without the adverse environmental effects posed by other disposal methods.

While many contaminants are present in used oil, it was realized that it would be futile to attempt to determine the contribution to the process by elements present in large quantities in cement raw materials or by elements present in used oils at very low concentrations. The elements remaining after such consideration were, therefore, lead, bromine, zinc, and phosphorus.

The emissions from the three stacks were of prime concern. These stacks exhaust the by-pass precipitator gases and the gases from the precipitators for the two preheaters. Particulate material was collected by the Ontario Research Foundation in accordance with both Environmental Protection Service [6] and Ontario Ministry of the Environment source testing codes [7]. After determining the quantities of particulate collected, samples were analyzed for the four elements under study.

As a backup to the emission analyses, a mass balance over the process was made for the same four elements.

The experiment was carried out under full normal operation conditions; the only change in procedure was the introduction of waste oil as a fuel. Normally, three separate oil burners, each capable of supplying 14 IGPM of No. 6 oil, are used to fuel the kiln. For the experiment, one of these burners was converted to supply used oil. Through viscosity and pressure changes, it was possible to input better than 15 IGPM of used oil.

Results

The mean emission rates given in table 1 indicate that the only significant increase in emissions during used oil burning was with bromine, presumably as alkali bromide. A slight increase in concentration of lead in the emission particulate was noted. This is not apparent in the reporting of total emissions since the actual particulate emissions decreased during this period, possibly because of water in the used oil improving the precipitator efficiencies. No effect of used oil burning on zinc and phosphorus emissions (except as related to total particulate decrease) was found.

In the accumulated mass balance (table 2), it can be seen that practically all the phosphorus and zinc remains with the clinker. The lead stays essentially with the clinker, although a small amount goes to the by-pass precipitator. A large portion of bromine goes to the precipitator dust. This is analogous to the reactions of chlorine described earlier in this paper. Alkali chlorides and bromides are volatile at kiln temperatures and are removed mainly through the by-pass. This volatility also explains the slightly higher emissions encountered during used oil burning.

Table 1. Summary of Emissions

Stack	period	Fuel	Mean flow (DSCFM)	Mean loading (mg/m^3)					Mean emission rate (lb./hr.)				
				Particulate	Lead	Zinc	Bromine	Phosphorus	Particulate	Lead	Zinc	Bromine	Phosphorus
NW	11/3/74		61000	320.4	.0456	.0279	.0784	.1169	73.5	.0104	.0062	.0180	.0278
NE	to	No. 6	63000	162.0	.0145	.0175	.0374	.0519	38.7	.0034	.0042	.0090	.0125
SE	15/3/74		9500	89.0	.0305	0	.5458	----	3.42	.0011	0	.0210	----
Total									115.6	.0149	.0104	.0480	.0403
NW	22/4/74	No. 6	60000	251.8	.0297	.0267	.2496	.0792	56.6	.0067	.0060	.0561	.0178
NE	to	+ used	61300	103.0	.0140	.0116	.0866	.0307	25.0	.0032	.0027	.0199	.0071
SE	2/5/74	oil	10500	86.6	.0901	.0166	.6017	----	1.72	.0032	.0007	.0243	----
Total									83.3	.0131	.0094	.1003	.0249
NW	28/5/74		57200	87.9	.0221	.0100	.0411	.0223	18.8	.0048	.0021	.0087	.0048
NE	to	No. 6	62500	547.6	.0399	.0580	.1669	.1678	128.3	.0118	.0134	.0391	.0392
SE	30/5/74		15200	264.3	.1230	.0019	1.1732	----	14.4	.0068	.0001	.0639	----
Total									161.5[a]	.0234	.0156	.1117	.0440

[a] Note precipitator difficulties on third test period.

Table 2. Accumulated Mass Balances

Accumulation period	Fuel	Element	Accumulated input (lb.) (T_I)	Percent of T_I derived from waste oil	Accumulated retention (lb.) (T_R)	Percent of T_I retained in:		
						Clinker	BP dust	Total
9/3 to 20/3	No. 6	Lead	502	0	627	117.1	7.8	124.9
		Bromine	590	0	564	55.4	40.2	95.6
		Zinc	5,125	0	5,909	114.9	0.3	115.2
		Phosphorus	40,260	0	39,500	98.0	0.1	98.1
17/4 to 7/5	No. 6 + used oil	Lead	13,731	96.6	12,246	85.0	4.2	89.2
+		Bromine	6,187	52.3	4,466	35.3	36.9	72.2
8/5 to 30/5	No. 6	Zinc	22,168	10.6	24,526	110.6	0.2	110.8
		Phosphorus	150,135	1.7	147,833	98.2	0.1	98.3

The results of the mass balance go a long way in confirming the emissions testing. We have confirmed that zinc and phosphorus are almost exclusively retained in the clinker. Lead is retained to a large extent in the clinker, and bromine is partly retained in the clinker and partly collected in the by-pass dust. It should be realized that in this study we were dealing with low concentrations of elements in a large quantity of material. The raw meal, for example, is fed to the system at approximately 5,000 TPD, and an error of 0.0001 percent in the analysis yields an error of 10 pounds per day for that element. Thus, no attempt should be made to equate small material balance losses with true losses from the process. An idea of the overall retention and disposition of each element can, however, be gained from the mass balance.

Discussion

What is important in the present study is a comparison with other available methods of disposal of used oil. Methods such as disposal into sewers or road oiling do nothing to eliminate the hazard of potential pollutants. Acid-clay treatment leaves a sludge which is itself a disposal problem. Burning in conventional oil burners releases much of the lead into the atmosphere during normal operation; the remainder is released during soot blowing.

One study [8] has been reported of burning used oil in coal-fired boilers. It was found that much of the lead was retained in the fly ash during burning with coal, although not as high a proportion as in cement clinker.

In all of these studies, only lead emissions were recorded. A comparison of the relative proportions of lead emitted for the St. Lawrence study, two studies on oil-fired boilers (Humble [3] and Shell [9]), and a study on a coal-fired boiler is given in table 3.

Table 3. Comparison of Lead Emissions During Waste Oil Burning at St. Lawrence Cement with Other Types of Combustion

Test location	Sample identification	Lead (lb./hr.)		Out/In (%)
		Total in	Total out	
St. Lawrence Cement	22/4	45	0.014	.031
	23/4	42	0.013	.031
	30/4(1)	32	0.009	.029
	30/4(2)	32	0.012	.037
	1/5	32	0.017	.052
Humble, Baltimore terminal	15/8/72(1)	0.235	.104	44.8
	15/8/72(2)	0.235	.156	66.4
	18/9/72	0.227	.160	70.5
	22/9/72	1.17	.504	45.1
	23/9/72	1.17	.389	33.2
	6/9/72	1.14	.400	35.1
	8/9/72	0.914	.424	46.4
	11/9/72	0.933	.480	51.4
	13/9/72	1.05	.496	47.2
	11/10/72	3.77	.806	21.4
	17/10/72	3.11	.746	24.0
	18/10/72	3.57	.849	23.8
Shell Oil	1	22.6	7.75	34
	2	22.6	7.50	33
	3	22.6	6.26	28
	4	17.0	4.35	26
	5	17.0	3.40	20
	6	17.0	4.02	24
	7	17.0	3.67	22
	8	9.2	4.50	49
	9	9.2	3.83	42
	10	9.2	9.73	102[a]
Northern States Power Company (coal-fired)	12/6/73	4.19	<.007	0.167

[a] Included 3-minute soot-blower operation.

The St. Lawrence Cement Company has found the burning of used oil to be economically viable. It is necessary to have adequate storage capacity to accept used oil during kiln shutdowns. Large storage capacity is also desirable so the quality of oil fed to the kiln is more uniform. Since the study, the company has installed 2 x 250,000 storage tanks with the necessary pumps and piping to burn up to 21,000 gallons per day of used oil.

Conclusions

Since all other studies were concerned only with lead release, this can be the only basis of comparison for environmental aspects. The burning of used oil in a cement kiln removes from the environment a higher proportion of the lead than other means of disposal presently available.

Since cement production requires high heat input, the use of used oil in cement kilns is attractive from an energy conservation standpoint.

Also, there is a cement plant near practically every major center, and these are, of course, the areas where used oil is concentrated.

Our study was on a dry process preheater kiln. In respect to removal of volatile components from kiln gases, the preheater is a more efficient scrubber than a straight kiln. It is anticipated that similar results will be obtained with other kiln systems, as long as the dust collection efficiency is adequate.

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INDUSTRIAL OILS USED AS FUEL

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U.S. industry uses more industrial oil than automotive and aviation lubricants. Use in 1975 was 1.58 billion gallons of industrial oils and 1.27 billion gallons of automotive and aviation oils [1].[1] In addition, the gap becomes larger each year and, with samller automobiles and longer drain intervals, will probably continue to increase.

One estimated but incomplete breakdown of industrial oil sales for 1975 (but probably applicable to 1977) shows [2]:

	(Gallons x 10^6)
Hydraulic and circulating system oils	314
Metalworking oils	145
Railroad engine oils	58
Aviation and other oils	145
Gas engine oils	58
Process oils	340
Electrical oils	62
Refrigeration oils	11
	1,133

A recent estimate of industrial oil use in 1977 and projected use in 1981 is shown in table 1 [3].

It is estimated that 481 million gallons is recoverable. This estimate is based on recoverability factors used in the Environmental Protection Agency (EPA) Waste Oil Study, Report to the Congress, April 1974.

The National Petroleum News Fact Book (1977) has published the following estimates [4]:

		Barrels 10^6		
Year	Total	Automotive oils	Industrial lubes	Process oils
1985	61.8	27.8	22.0	12.0
1980	57.6	26.7	20.9	10.0
1978	56.7	26.7	20.7	9.3
1977	56.0	26.5	20.5	9.0
1976	55.3	26.2	20.5	8.6
1975	50.1	23.5	18.9	7.7
1974	56.7	25.9	21.9	8.9
1973	59.0	26.9	23.1	9.0

There is a trend toward use of synthetic fluids for metalworking and hydraulic and circulating systems, and I expect this to continue, as well as the use of dual-purpose hydraulic and metalworking fluids. A larger fraction of the hydraulic oils used is lost through leakage before it is unsuitable for hydraulic systems. Therefore, industry will try to minimize this loss which would reduce the disposal problem.

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[1] Underlined numbers in brackets indicate the literature references at the end of this paper.

Table 1. Preliminary Estimate of U.S. Market for Formulated Industrial Oils and Lubricants by Type - 1976 and 1981 [3]

	Million gallons		Million dollars	
	1976	1981	1976	1981
Machine oils				
Hydraulic fluids	240	290	230	275
Engine oils (railroad, airline, and other large engines)	150	170	150	170
Gear oils	75	90	105	130
Compressor oils, way oils, spindle oils, and all others	105	130	100	125
Subtotal	570	680	585	700
Metalworking oils				
Cutting oils[a]	140	160	130	150
Rolling oils	45	50	55	60
Rust-preventive oils	20	23	20	23
Quenching oils	20	23	20	23
All others	20	24	20	24
Subtotal	245	280	245	280
Process oils[b]	200	240	160	190
Electrical insulating oils				
Transformer oils	80	100	45	55
Switch oils	20	27	10	13
Cable oils	10	13	5	7
Subtotal	110	140	60	75
Greases	95	115	165	195
Total	1,220	1,455	1,215	1,440

[a] Includes straight, soluble, and chemical cutting oils.

[b] Excludes aromatic by-products.

There has been an increasing recycling of used industrial oils, both by in-plant reprocessing and by use of outside contractors. These reprocess the used oils and return them to the generating plant for reuse. The increasing price of petroleum oils and the environmental problems associated with disposal have been strong factors in industry's interest in recycling.

Unfortunately, we have no accurate numbers for the amount of used industrial oil collected or recycled. Large industry in general recycles used oil for reuse as metalworking fluids, hydraulic oil, specialty lubricants, and fuel. However, there is a strong move to recycle oils for uses other than fuel.

For this discussion, industrial oils do not include railroad, gasoline engine, or aviation lubricants since they belong more properly in the automotive oil category. The largest component of this class of lubricants is diesel engine oil. These are similar to used automotive oils in many respects, although they have a much lower lead content, a higher carbon content, and metals indicative of the additive package. Process oils, such as those used in ink, rubber, and plastic manufacturing, also are not included since little of this is recoverable. Included in this category are metalworking fluids, hydraulic oils, gear oils, rolling and forming lubricants, rust-proofing oils, air compressor oils, quenching oils, and many other specialty oils and fluids. The composition of industrial used oil streams can vary widely, reflecting the type of manufacturing and the lubricants and fluids used.

Let us take two industry examples. Steel and automobile manufacturing operations are the two larger users of lubricants and fluids. We can include manufacturers of farm equipment, home appliances, compressors, and others in the automotive sector since the oils and fluids used are the same as, or similar to, those used in automobile manufacturing, i.e., metalworking.

Steel mills have two major sources of used oil generation, rolling and lubrication. The former oil ranges from highly compounded petroleum oils to fatty oils such as No. 1 tallow. The latter is comprised of hydraulic oils, gear and bearing oils, and greases. Some steel mills use the services of in-plant and out-plant reprocessing contractors. The rolling oils can be reprocessed and returned to the roll stands. A mixture of all oils used in the mill is collected at a waste-water terminal treatment facility. Table 2 shows some analytical data on three samples of used oil from a steel mill waste-water terminal treatment facility. This used oil, when substantially dehydrated, includes 8 to 12 percent solids (mostly ferrous), as high as 60 percent fatty oil, soaps, emulsifiers, and original bearing and hydraulic oil additives. Ash runs from 3.0 to 6.0 percent and may be higher. Sulfur contents of 0.6 to 1.0 percent have been analyzed. The viscosity of these oils puts them in the No. 5 heavy fuel classification. Bottom settlings (BS&W), flash point, and even the 1.0 percent sulfur also place these oils in the high-sulfur, heavy-residual oil classification. The ash is far too high, and water can be borderline at 1.0 percent. The Btu [British thermal unit] content is low for the gravities shown. Virgin fuels contrasted with these oils would have the following heat content:

API gravity, used oil	Used oil (Btu/lb.)	Virgin oil (Btu/lb.)
21.9	17,990	19,105
19.3	17,747	18,998
15.6	17,225	18,820

We have no data on the effect of the ash components on burners or boiler and furnace surfaces. Currently, some oils such as these are reprocessed by contractors to reduce ash and water content and, when mixed with virgin fuels, are used in the mill. Some oils are used as is (no reprocessing) and are, again, diluted with virgin fuels.

If the oils are not reprocessed, one would expect excessive pump and burner nozzle wear and higher deposits in boilers and furnaces. The effects due to the burning of fatty oils containing fatty acids may also need to be addressed.

Industrial metalworking operations, such as those used in automobile manufacturing, generate used oils reflecting a composite of the type and quantity of oils used in the operations. Production of components, such as automatic and manual transmissions, engines, and axles, which requires extensive metal cutting and griding generates used oils which are far different from those generated by a steel mill, metal stamping plant, or a forging plant.

The greatest oil use in industry is that of hydraulic oils, followed by metalworking oils; quenching oils; gear oils and greases; and such specialty oils as compressor, air conditioner, and transformer oils.

Most of the hydraulic oil used is the petroleum-base type, although synthetic nonflammable ester, water-glycol, and water-in-oil emulsion products are used in special operations.

Metalworking fluids range from low viscosity oils (even high flash point kerosine) for honing and lapping, to high viscosity oils with fatty oils, sulfur, chlorine, and

Table 2. Analytical Data - Used Industrial Oil

	Analyses		
	1	2	3
Flash point, °F	345	355	315
Fire	395	385	375
Gravity, API	21.9	19.3	15.6
Specific gravity	.9224	.9383	.9619
Lb./gal.	7.681	7.814	8.011
Btu/lb.	17,990	17,747	17,255
Btu/gal.	138,181	138,652	138,230
Sulfur, %	0.57	1.03	1.02
Viscosity			
122° F, SSU	310	300	430
210° F, SSU	66	68	74
Ash, %	3.22	3.36	5.9
Water, %	<1.0	0.1	>0.1
Fatty oils, %[a]	40.0 - 60.0	40.0 - 60.0	40.0 - 60.0

[a] Author's estimate.

phosphorus additives, among others. Emulsifiable petroleum oils, often with extreme pressure and antiweld additives, are widely used. A wide range of synthetic products is also used for metalworking. These contain as many materials as a researcher's mind can conceive, including soaps, emulsifiers, glycols, borates, and esters.

Used industrial oils will also often contain lower flash solvents such as kerosine and even those with even lower flash points, although the Occupational Safety and Health Administration (OSHA) and insurance underwriters frown on this. Chlorinated solvents such as trichlorethylene, may also be present.

With few exceptions, in larger plants most of the used oil generated is collected at waste-water treatment facilities. Unfortunately, very few industrial plants segregate used oil at the source, which would simplify recycling. Most remove a high percentage of the water and solids which separate by gravity. In some cases, centrifugation or filtration is employed to remove water and solids. Used oils are also reprocessed by vacuum distillation, clay contact, and filtration. Oils used as in-plant fuel also may be centrifuged after gravity dewatering. Surfactants to aid water and solids separation may also be used.

Table 3 shows analytical data on two used oils from an automatic transmission plant. One is the mixture of oils recovered at the waste-water treatment facility. The second is used oil skimmed from a coolant system. It is a petroleum-base hydraulic oil which has picked up some emulsifiable oil components and metals from the operation.

The viscosities are fairly low, 143 and 167 at 100° F SSU, but there are wide variations in other areas, reflecting the used oil sources. No. 1, the mixture of all plant used oils which are collected at the waste-water treatment facility, shows 0.83 percent versus less than 0.1 percent chlorine; 0.71 percent sulfur versus 0.54 percent; total acid numbers of 5.2 versus 0.96; and barium, calcium, phosphorus, and zinc present in percentages from 0.1 to 1.0. Other metals such as aluminum, copper, iron, lead, silicon, and sodium are present in parts per million (ppm), ranging from less than 1 to more than 200. The larger quantities, as one would expect for this operation, are iron and aluminum.

Table 3. Analytical Data - Used Industrial Oils

	1	2
Viscosity		
100° F SSU	143.1	167.2
210° F SSU	42.6	43.2
Viscosity index	97.0	72.0
Water, %	0.3	4.6
Oil, %	99.5	94.4
Sludge, %	0.2	1.0
Gravity, API	25.7	26.2
Flash point	340	405
Color (diluted)	<6.5	<4.5
Chlorine, %	0.83	<0.1
Sulfur, %	0.71	0.54
Total acid number	5.2	0.96
Metals, additives, %		
Barium	<0.01	<1.0
Calcium	<0.01	<0.02
Phosphorus	<0.05	<1.0
Zinc	<0.01	<1.0
Trace metals, ppm		
Aluminum	<100	>100
Boron	2.0	1.0
Chromium	5.0	2.0
Copper	10.0	5.0
Iron	>200.0	34.0
Lead	38.0	>100.0
Silicon	24.0	7.0
Silver	<1.0	<1.0
Sodium	10.0	7.0
Tin	9.0	<1.0

The use of used industrial oils of the types discussed as fuel raises several "substantial equivalency" questions. The fact that most used oils (but not all) are mixed with virgin fuels should not be a factor in determining test methods for substantial equivalency. The American Society for Testing and Materials (ASTM) D 396 fuel oil standard is not overly definitive. Used oils do not fall neatly into any grade, but usually straddle No. 2 to No. 6. Unwritten industry standards for vanadium, sodium, and other elements must be considered since these and other operational factors are to be considered. These include the effects of the fuels on burners, combustion chamber surfaces, and flues and stack surfaces. Stack emissions also cannot be ignored. OSHA and EPA tank storage regulations will also have to be considered.

Industrial oils seem to present fewer problems in fuel use than used automotive oils. For example, they have a lower lead content. However, there are other constituents which may also be undesirable from an operational standpoint, such as sulfur, chlorine, and fatty oils; and additives such as metal sulfonates - calcium, barium, and sodium. On the other hand, there is some evidence that used oils, when mixed with virgin residual fuels, may be beneficial. It is possible that some of the calcium, barium, zinc, and even lead compounds may provide the beneficial effects of commercial fuel oil additives.

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RUNOFF OF OILS FROM RURAL ROADS TREATED TO SUPPRESS DUST

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Abstract

Two rural roads in Readington Township, Hunterdon County, New Jersey, which are treated with waste crankcase oils, were examined to determine whether or not the oil leaves the road. Analyses indicated that roughly 1 percent of the total oil estimated to have been applied remains in the top inch of road surface material, that oil penetration below the top inch of road was minimal, and that lead was concentrated (≈ 200 mg/kg) in the top inch of road material.

Laboratory weathering experiments indicate an estimated maximum weathering loss of oil from a road would be approximately 18 percent. Rain runoff studies on simulated rural road surfaces indicated two mechanisms by which oil is transported from the road: leaching of the oil by flotation, and flotation of oil-wet soil particles. The greatest oil transport is during the first few rains after oil application, with continuous, low-level leaching during each subsequent rain.

Analyses of soil samples taken from a field subjected to runoff from an oiled road showed significantly higher lead content than soil taken from a field 150 feet from the road.

Five photographs clearly showing oil leaching from the test road surface and entering drainage ditches after a rain are presented.

Introduction

An estimated 200 million gallons of waste crankcase oils and an unknown quantity of other waste oils are applied yearly to rural roads in this country for the purpose of dust control [1].[1] The waste crankcase oil contains approximately 1 percent by weight of lead compounds, which amounts to 14.0 million pounds of lead applied yearly to roads.

In Readington Township, Hunterdon County, New Jersey, dirt roads are oiled twice yearly, in June and August, to suppress dust in front of houses along the roads. Two of the roads in Readington Township were examined to determine if the waste oils were leaving the road surfaces. The problem was approached by attempting a mass balance to determine the amount of oil applied to the road surface, the amount remaining in the road surface, and the amounts leaving the surface by various mechanisms. Additionally, analyses were made to determine if the lead component of the oil were also leaving the road.

The top inch of road surface material extending the width of the road between drainage ditches was considered the object of the mass balance. Oil could leave the road surface by several mechanisms: volatilization; runoff mechanisms; adhesion to vehicles passing (with possible redeposition at other areas); adhesion to dust particles with wind transport from the road surface; or penetration into the road material below the surface, possibly through capillary action. Additionally, the oil remaining in the road surface could be biodegraded.

Some of the variables affecting the above movements are oil application history, frequency, quantity per application; oil type, source, viscosity, density, volatility, presence of surface active agents; road conditions: crown, slope, compaction, density

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[1] Underlined numbers in brackets indicate the literature references at the end of this report.

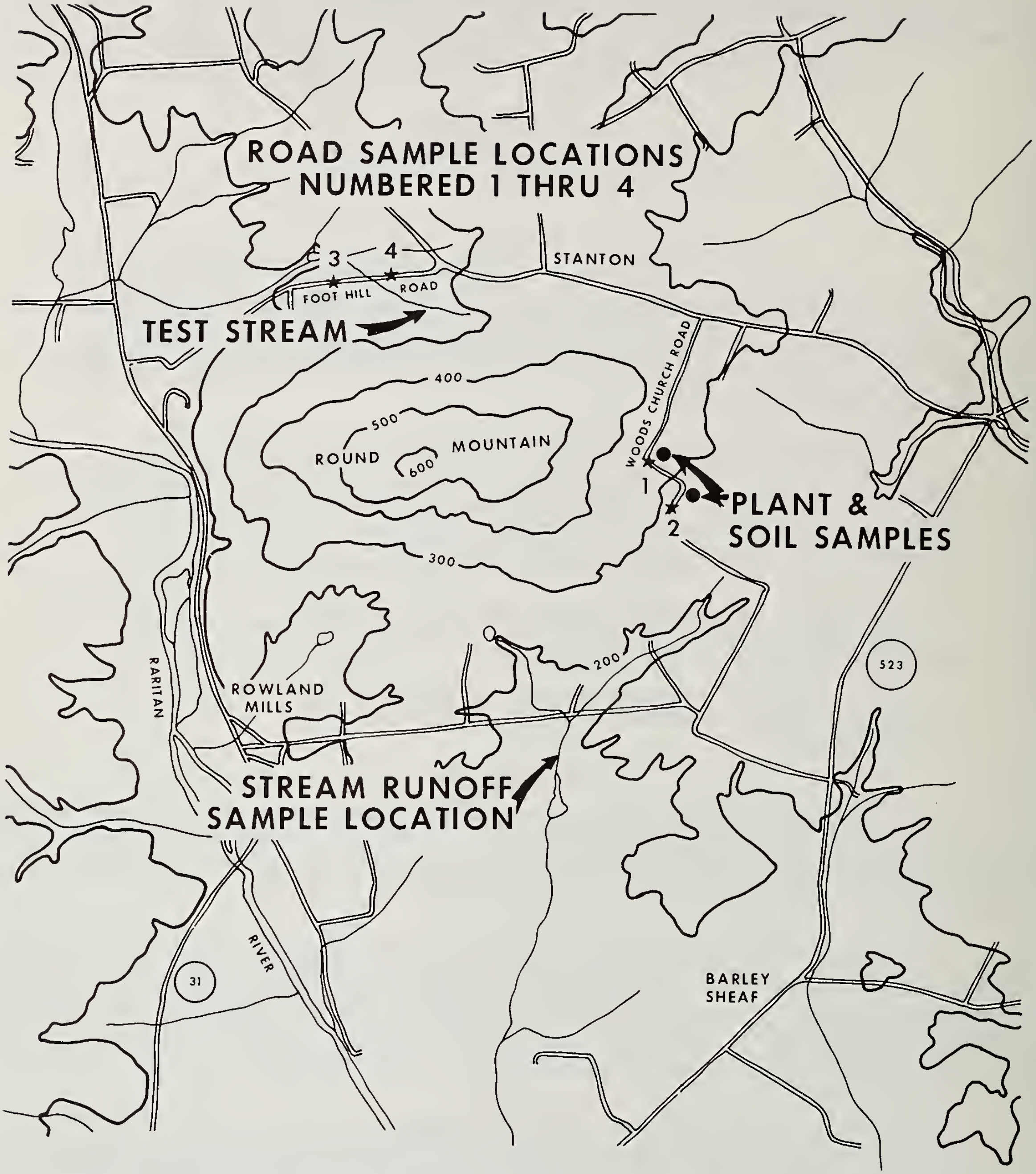

Figure 1. Test locations map adapted from the Flemington, New Jersey, U.S.G.S. quadrangle.

and porosity of soil, soil grain size distribution; traffic conditions on the road; weather conditions: time to the first rain after oil application, total yearly rainfall, temperature affecting volatility of oil, wind affecting dust transport; and factors affecting biodegradation: presence of oil-consuming microorganisms, road surface moisture content, and presence of nitrogen and phosphorus nutrients.

With such a range of variables and oil movement mechanisms, precise quantitation of oil movement applicable to numerous roads is beyond the scope of this study. However, with information available, estimates may be made of some of the more significant movements of oil.

Experimental

Oiling History

No records are available of the true oiling history of the test roads. The Readington Township Clerk indicates that the roads have been oiled twice yearly for at least 12 years. The waste oil collector who has done the oiling indicates the roads have been oiled by him for 16 years (since 1955) and by others before him for an unknown number of years. Road oiling to lay dust is usually done in other townships in response to complaints from persons living along the roads; thus, during dry years the roads may be oiled twice, and in wet years once or not at all. A conservative estimate of the total number of oilings on the test roads is 24, considering 2 oilings per year for 12 years, with the understanding noted above.

Oil Type and Application Rate

The oil type and application rate is highly variable. Conversations with the waste oil collector who oils the Readington roads indicate that he uses a mixture of Nos. 4, 5, or 6 fuel oil sludge obtained from tank cleaning operations, and waste crankcase oils collected from service stations in the Trenton, New Jersey, area. The ratio of the mixture depends upon the availability of the fuel oil sludge, and ranges from 15 to 30 percent sludge, the balance being waste crankcase oils.

A waste oil collector from the Boston area who oils roads in New England and upstate New York indicates that he uses waste water-soluble cutting oils, waste crankcase oils, terminal waste oils (from the oil/water separator on a water effluent), and oils from tank-cleaning operations. The oil applied to a given road depends on the availability of oils at the time of the oiling and follows no particular pattern.

Application rate is apparently related to the qualitative judgment of the operator of the truck doing the oiling and ranges from 0.025 to 0.05 gallon per square foot of road surface, with the 0.05 figure being used on the test roads.

Oil in the Road Surface: Concentration Versus Depth

The two test roads were sampled in four oiled locations and in two unoiled locations to determine concentration of hydrocarbons versus depth. In all oiled locations the top inch of road surface showed the greatest concentration of oil, an average of 0.74 percent weight, with trace quantities of oils at lower depths (which may be contamination attributable to the sampling technique). The road surface material was made up predominately of clay, with some sand. No attempt was made to characterize the material in more detail. Complete sampling data are presented in table 1.

Table 1. Penetration of Oil into Road Surface

Station	Hole	Depth (inches	Hydrocarbons, mg/kg
1	1	Surface	6,313.17
		4	18.04
		6	18.53
	2	Surface	12,572.70
		4	26.42
		6	52.62
	3	Surface	8,254.50
		4	88.72
		6	7.67
	4	Surface	5,880.24
		4	70.71
		6	7.65
	5	Surface	13,441.25
		4	39.95
		6	67.63
	6	Surface	2,555.91
		4	59.87
		8	9.35
		10	12.15
2 Control	1	Surface	347.76
		4	0
		6	0
	2	Surface	131.04
		4	0
	3	Surface	211.83
		4	0
3	1	Surface	1,586.22
		6	354.40
	2	Surface	9,437.94
		4	805.74
		6	198.35
	3	Surface	6,222.52
		4	276.21
4 Control	1	Surface	142.72
		6	10.04

Estimate of Oil Remaining in Test Road

Oil Applied

0.05 gal/ft^2 per application

2 applications/yr x 12 yrs = 24 applications

0.05 x 24 = 1.2 gal/ft^2 total applied oil

1.2 gal/ft^2 x 9 ft^2/yd^2 - 10.8 gal/yd^2

Oil penetrates 1 inch; consider unit of road surface to be 1 yd^3 and be of dimensions 1 yd wide, 36 yds long, and 1 inch deep

10.8 gal/yd^2 x 36 yd^2/yd^3 (for depth of 1 inch) - 388.8 gal/yd^3 road surface

Oil weight:

Oil specific gravity: 0.904

Water weighs 8.337 lb/gal

Oil weighs 0.904 x 8.337 = 7.537 lb/gal

Weight of oil applied - $388.8 \frac{\text{gal}}{\text{yd}^3} \times 7.537 \frac{\text{lb}}{\text{gal}}$

For 12 yrs of application = 2930.25 lb oil/yd^3 road surface

For 20 yrs of application - 4883.9 lb oil/yd^3 road surface

Oil Remaining in Road

Weight of road surface:

Average soil specific gravity = 2.75

$$2.75 \times 62.4 \text{ lb/ft}^3 \times 27 \frac{\text{ft}^3}{\text{yd}^3} = 4{,}644 \frac{\text{lb}}{\text{yd}^3}$$

Oil in road surface: average concentration = 0.74 percent weight (neglecting weight of water in soil

$$0.0074 \frac{\text{lb oil}}{\text{lb road surface}} \times 4{,}644 \frac{\text{lb road surface}}{\text{yd}^3 \text{ road surface}}$$

$$= 34.37 \frac{\text{lb oil remaining}}{\text{yd}^3 \text{ road surface}}$$

Percent of Oil Applied Which Remains

$= \frac{34.37}{2930} \times 100 = 1.17$ percent assuming 12 yrs of oiling

$= \frac{34.37}{4884} \times 100 = 0.7$ percent assuming 20 years of oiling

Oil Volatility

To estimate the level of volatility of waste oil applied to Readington Township roads, a laboratory weathering experiment was performed. Waste oil obtained from the waste oil collector and being of a "typical" composition of approximately 20-percent No. 6 sludge and 80-percent waste crankcase oil was placed in shallow 11-inch by 14-inch laboratory pans under infrared lamps and in the draft of a fan. The surface temperature of the oil was held at 100° F for a duration of 288 hours to achieve a high, though arbitrary, weathering stress. At the end of the weathering period, a 16- to 18-percent change in weight of oil was noted.

Waste oil obtained from a local service station was weathered under the same conditions for a period of 360 hours and changed weight by 7 to 9 percent. Exact weathering figures are presented in table 2.

Table 2. Weathering Data

Test number	Oil type	Temperature (°F)	Time (hours)	Weight loss (%)
1	1[a]	90	72	5.97
2	1[a]	100	360	9.05
3	1[a]	100	360	7.29
4	2[b]	100	288	18.15
5	2[b]	100	288	16.43
6	2[b] (applied to clay)	100	354	16.13
7	2[b] (applied to clay)	100	354	17.07

[a] Local service station.

[b] Waste oil collector.

Runoff Mechanisms

To observe runoff mechanisms of oiled roads, a laboratory tank with two compartments was filled with compacted clay material and sand material, respectively, in the two compartments. No attempt was made to characterize the materials in the tank more precisely than "sand" and "clay". The tank was so arranged that for each compartment there was a "road surface" of dimensions 13 inches by 35 inches and 18 inches deep, with a false bottom in the tank arranged to divert any penetrating fluids to a collection bottle below the tank. The surface of the material in the tank was sloped such that runoff of waters and oils could be contained in a large bottle adjacent to the tank.

Each "road surface" was roughened to simulate a lightly scarified condition as observed on the test roads. Oil was applied to each tank compartment at a rate of 0.05 gal/ft^2. The tank was then allowed to stand indoors for a period of 3 days. "Rain" was applied, corresponding to the June and July rainfall averages for the Flemington area (see table 3) on each of 2 days to the oiled sand and clay compartments. "Rain" and oil were applied through appropriate spray nozzles from pressurized containers. It is recognized that application rates of oils to sand roads may differ from those of clay roads and that rainfall averages for sandy soil areas may be slightly different. However, the intent of the experiment was observation of mechanisms rather than quantification. Oil was again applied at the above rate and "rains" corresponding to the August and September rainfalls were applied and collected. Complete data are presented in table 4.

The principal observations derived from this experiment were:

1. The oil penetrated the clay to a depth of about 1 mm below the scarified level.

2. Even though the surface was scarified, the oil ran off the clay during application and "puddled" in the lowest portion of the road surface.

3. Application of "rain" to the clay road washed oil from the puddled area and leached oil from the clay surface. Additionally, some oil-wet particles floated with the rain runoff and were carried from the road surface.

4. There was no penetration of oil or water entirely through the clay column.

Table 3. Rainfall Data - Flemington, New Jersey

	1931 - 1960	1951 - 1960 Averages	
Month	Average (inches)	No. rains > 0.1 inch	No. rains > 0.5 inches
January	3.32	6	2
February	2.78	7	2
March	3.99	8	4
April	3.80	9	3
May	4.01	6	3
June	3.80	6	2
July	4.52	6	3
August	5.02	7	3
September	3.59	5	2
October	3.32	6	3
November	3.74	7	3
December	3.42	6	3
Total	45.31		

Table 4. Laboratory Runoff Study

			Runoff			
			Sand		Clay	
Time (days)	Oil applied (ml)	"Rain" applied (l)	Water (l)	Oil (ml)	Water (l)	Oil (ml)
0	600	---	---	---	---	---
3	---	28.3	20.5	80.2	26.0	37.5
4	---	33.7	20.9	101.2	32.5	15.8
5	600	---	---	---	---	---
6	---	37.4	31.5	77.1	37.0	89.7
7	---	26.7	19.5	29.5	23.4	5.3

Total water penetration through the sand column was 10.0 l of water containing 12.2 ml of hydrocarbons. No penetration was observed through the clay column.

5. The oil penetrated the sand "road surface" to a depth of a few grain diameters and was evenly distributed over the sand with no puddling.

6. Application of "rain" caused the flotation of oil-wet sand particles which were then carried with the rain runoff into the collection container.

7. Some leaching of oil from the sand was observed; however, the predominant mechanism of oil transport from the sand road surface was by flotation of oil-wet sand particles.

8. After application of 2 simulated monthly rainfalls (June and July), approximately 20 percent of the total quantity of oil-wet sand particles had been transported by runoff from the road surface.

9. Ten liters of water penetrated the 18-inch deep column of sand. The water contained 12.2 ml of hydrocarbon material.

Biodegradation

The rate of biodegradation of the oils in the surface of the road material will depend principally upon the presence of hydrocarbon-consuming microorganisms, the moisture content of the soil, the presence of the nutrient materials (nitrogen and phosphorus), and ambient temperature. No attempt was made to estimate the amount of total applied oil which had been biodegraded in the road surface. It is our opinion that biodegradation will account for a small amount of the oil loss, in relation to other physical oil movement mechanisms such as volatilization and runoff. It should be noted that the road surface moisture content is low when the oil is applied and that when rain occurs physical mechanisms predominate.

Adhesion to Vehicles and Dust Transport

It is recognized that oil is transported along the test road by adhesion to tires and underbodies of passing vehicles and is redeposited in some manner. However, no satisfactory method has been defined to account for such transport. Similarly, it is felt that some oil may adhere to dust particles and be transported by wind from the road surface (see below), but no quantitation of this mechanism was attempted.

Analysis of Trace Quantities of Crankcase Oil in Runoff Waters

A stream which receives runoff waters from one of the test roads, Woodschurch Road, was examined to determine if crankcase oils were present. Two oil patches, each of several square inches in area and having "rainbow" colors, were noted in shallow pools on the banks of the small stream. Presumably, the oils were deposited in their noted locations during a period of high stream flow. Sampling and analysis of the oil patch indicated that the sample hydrocarbons bore great similarity to the hydrocarbons extracted from the surface material of Woodschurch Road. In addition, the sample hydrocarbons were similar to waste crankcase oil, but were different from 10 other common oils tested, including Nos. 2 and 6 fuel oils. Analysis was made by comparing the fluorescent spectral properties of the known materials with those of the sample.

Lead in Soils and Plants

Waste crankcase oils have been reported to contain up to 1-percent lead by weight [2] (see table 5). If ecological damage were to result from the application of waste oils to roads, the high lead content of the oils could have significant bearing on that damage. Therefore, tests were made to determine if the lead were leaving the road. Samples were taken from oiled and unoiled stretches of road, from drainage ditches at the sides of the roads, from a field 150 feet away from the oiled road, and at the outfall of a pipe carrying runoff water from several hundred feet of oiled road. Lead analyses of plant and soil samples from these areas are presented in table 6. Some of the more significant results are:

Table 5. Typical Composition of Waste Crankcase Oils [2]

Port Mobil Test Fuel Inspections

	Waste oil (weighted average of four deliveries)	No. 6 fuel
Gravity, °API	24.6	15.7
Viscosity @ 100° F, CS	53.3	915.7[a]
Viscosity @ 100° F, SUS	248	4239
Viscosity @ 210° F, CS	9.18	33.5
Viscosity @ 210° F, SUS	56.4	158
Flash point, °F	215[b]	192[c]
Water (by distillation), % volume	4.4	
BS&W, % volume	0.6[d]	0.7
Sulfur, % weight	0.34	1.9
Ash, sulfated, % weight	1.81	0.08
Lead	1.11 % wt.	59 ppm
Calcium, % weight	0.17	--
Zinc, % weight	0.08	--
Phosphorus, % weight	0.09	--
Barium, ppm	568	--
Iron, ppm	356	--
Vanadium, ppm	<5	440

[a] Viscosity, Furol @ 122° F 188.

[b] C.O.C. flash.

[c] P.M. flash.

[d] Poor separation on waste oil.

1. The average lead concentration in the oiled sections of road was 209.25 mg/kg, as compared to an average lead concentration in the unoiled sections of 39.0 mg/kg.

2. The soil samples taken in the (wheat) field 150 feet from the road showed an average value of 24.0 mg/kg, which is somewhat higher than the expected background for the soil near a rural road [3].

3. The plant samples taken in the same field as item 2 above showed an average lead concentration of 33.5 mg/kg, which is higher than the expected background level [3].

4. A possible explanation of items 2 and 3 above is that there may be a transport mechanism in effect which associates lead (and/or oil) with dust particles which are blown from the road surface by wind. The field is higher in the overall drainage pattern that the road.

Table 6. Lead Analysis Results

Sample type	Source	Sample number	Lead content, mg/kg
Oil road surface	Woodschurch Road	1	184.0
		2	223.0
	Foot Hill Road	3	172.0
		4	258.0
			Avg. = 209.25
Unoiled road surface	Foot Hill Road	5	36.0
		6	42.0
			Avg. = 39.0
Bottom soil (rocks, hard soil)	Drainage ditch, Woodschurch Road	7	42.0
		8	59.0
			Avg. = 50.5
Plants	Drainage ditch	9	42.0
		10	16.0
			Avg. = 29.0
Bottom soil (soft soil)	Drainage ditch, Foot Hill Road	11	122.0
		12	150.0
			Avg. = 136.0
Soil	Field 50 yds. from Woodschurch Road	13	25.0
		14	23.0
			Avg. = 24.0
Plants (wheat stubble)	Woodschurch Road	15	32.0
		16	35.0
			Avg. = 33.5
Soil	Field at end of drainage culvert (carrying runoff)	17	96.0
		18	78.0
			Avg. = 87.0
Waste oil[a]	Waste oil collector	19	350 ppm[b]
Waste oil	Local service station	20	525 ppm[b]

[a] Waste oil = 20% No. 5 fuel oil sludge and 80% waste crankcase oil.

[b] Average of three applications.

5. The soil samples taken at the outfall of the drain pipe carrying runoff water from the road showed an average lead concentration of 87.0 mg/kg.

6. The lead level in the oil sample provided by the waste oil collector averaged 350 ppm for 3 replicates.

7. The lead level in the waste oil from the local service station was 525 ppm average for 3 replicates (results by atomic absorption spectrophotometer).

Biological Assessment of Lead in Test Aquatic Organisms

Aquatic organisms from a stream passing beneath one of the test roads (Foot Hill Road) were analyzed for lead content (see table 7). A small stream passed perpendicular to the oiled road, and drainage from the road entered the stream at the crossing. The stream uphill of the test road did not receive any other runoff and was therefore designated "control." The stream section downhill from the test road received the runoff from approximately 1,000 feet of 12-foot wide road of which roughly 500 feet had been oiled. This stream section was designated "contaminated." A variety of aquatic organisms with varying feeding habits was collected, classified, and analyzed for lead content from each of the stream sections. Collections were made approximately 100 yards upstream from the road on the control side and approximately 200 yards downstream on the "contaminated side."

The results of the lead analysis of tissues of the sample aquatic invertebrates are, at best, sketchy. These data are not consistent, with some of the control taxa exhibiting higher lead concentrations than the same taxa from the exposed section of the stream.

The herbivorous mayflies from the "contaminated" area tend to have more lead in their tissues than do the control organisms (see pairs 1 and 2), but the predatory dipteran, *Tipula*, and the stoneflies which contain the carnivor, *Acroneuria*, show a reverse trend (see pairs 3 and 4). The filter-feeding blackflies exhibit more lead from the exposed area, possibly indicating a transport of lead in suspension; but the web-building caddis fly, *Cheumatopsyche*, shows a slight reversal of this tendency (see pairs 5 and 6). The omnivorous crayfish show no difference in lead content (see pair 7).

Estimate of Lead Remaining in Test Road

Total Lead Applied

Total oil applied: $2930 \ \frac{\text{lb oil}}{\text{yd}^3 \ \text{road surface}}$

Lead = 0.7 percent oil by weight.

$$0.007 \times 2930 = 20.51 \ \frac{\text{lb lead}}{\text{yd}^3 \ \text{road surface}}$$

Lead Remaining in Road

Lead in oiled surface, average concentration: 290.25 mg/kg

Lead in unoiled surface, average concentration: 39.0 mg/kg

Net lead contributed by oil: 170.25 mg/kg

$$0.00017 \ \frac{\text{lb lead}}{\text{lb road}} \times 4{,}644 \ \frac{\text{lb road}}{\text{yd}^3 \ \text{road}} = 0.789 \ \frac{\text{lb road}}{\text{yd}^3 \ \text{road}}$$

Table 7. Analysis of Aquatic Organisms for Lead

Pair no.	Taxa	Control Dry wt., gm	Control No. in sample	Control Lead, ppm dry wt.	Contaminated Dry wt., gm	Contaminated No. in sample	Contaminated Lead, ppm dry wt.
1	*Stonenema* herbivorous mayfly nymphs	.107	10	14.22	.064	6	46.36
	Ephemerella herbivorous mayfly nymphs	.104	904		.087	382	
2	*Ameletus* herbivorous mayfly nymphs	.122	151	20.74	.095	68	38.14
	Paraleptophlebia herbivorous, mayfly nymphs	.065	75		.023	28	
3	*Acroneuria* carnivorous stonefly nymphs	.073	8	54.8	.156	17	41.28
	Allocapnia herbivorous stonefly nymphs	.116	165		.062	97	
4	*Tipula* carnivorous cranefly larvae	.159	3	44.03	2.066	14	17.42
5	Simulidae filter-feeding blackfly larvae	.701	2075	14.26	.063	207	71.43
6	*Cheumatopsyche* web-feeding caddis fly larvae	.059	76	50.80	.088	60	45.45
7	*Cambarus bartoni* omnivorous crayfish adult	2.771	4	19.84	1.546	1	18.11

Percentage of Lead Applied Which Remains

$$\frac{0.789}{20.51}(100) = 3.85\%$$

$$\frac{0.789}{1.465}(100) = 53.8\% \text{ remains if 500 ppm lead in oil.}$$

$$\frac{0.789}{29.3}(100) = 2.6\% \text{ remains if 1\% lead in oil.}$$

Sampling Procedures

Road surface - The test roads were sampled for concentration of hydrocarbons at various depths. Six-inch diameter holes were bored in the road with a portable gasogasoline-powered post hole auger. After scraping the sides of the hole to remove extraneous material, 50- to 100-gram samples were removed from the sides of the hole at depths of interest. The "surface" sample was made up of a composite of surface material from the top inch of road. Other samples were taken from approximately one-inch thick layers of road material, with the center of the layer being at the depth of interest. The holes were bored as deep as was possible with the equipment used; the road was apparently underlain with a very hard material which was impenetravel to the borer.

Samples for lead were taken from the road surface only. All road samples were placed in plastic bags for transportation to the laboratory.

Soil - The soil samples in the drainage ditches and at the discharge end of the drainage culvert were taken by dislodging a small amount of (frozen) soil with a pick and placing the sample in a plastic bag. The soil samples from the field 150 feet from the road were taken from cultivated soil by the same technique. Samples were placed in plastic bags for transportation to the laboratory.

Plants - Plant samples collected in the field 150 feet away from the road were from the previous year's growth of wheat and were taken by breaking off the wheat stalk at the ground and placing the sample in a plastic bag. Grass samples from the drainage ditches were taken by breaking off the grass at the soil line by hand and placing the samples in plastic bags.

Aquatic organisms - Benthic organisms were sampled by disturbing the stream bottom material and allowing the organisms to be collected in a 500 μ mesh sampling net. The samples were preserved at the sampling site in 70-percent ethanol.

Analysis Techniques

Hydrocarbons - Hydrocarbon material was extracted from samples with carbon tetrachloride (4 extractions at 25 cc each for a 1 liter sample), and the infrared spectral peak at 2930 cm [1] compared to the peak of known concentrations of similar oils in similar solvent.

Lead - Samples were prepared in accordance with American Society for Testing and Materials' designation D 1548-63 (for vanadium, adapted by the Edison Laboratory for lead). The detection method was atomic absorption spectrophotometry.

Discussion

When attempting to quantify the oil runoff from the test road, it is obvious that only a small fraction of the oil applied to the road can be accounted for as remaining in the road and volatilizing. Unfortunately, no satisfactory scheme can be found to account for oil transported from the road by adhesion to vehicles and dust transport, nor can the effect of biodegradation be easily quantified. It is therefore impossible to assign a numerical value to the amount of oil which runs off the road by subtracting the sum of other effects from the amount of oil applied.

However, figures 2 through 5 clearly indicate that oil moves from the surface of the test road with rain runoff, and analytical results presented above indicated that oil found on the banks of a stream receiving runoff from the test road is similar in composition to oil extracted from the test road surface.

It is the opinion of this investigator that volatilization, adhesion to vehicles, and biodegradation taken all together probably account for only 25 to 30 percent of the oil applied to the road surface. The 70 to 75 percent of the oil applied which leaves the roads by dust transport and runoff could have significant ecological effects as a result of the oil or its accompanying heavy metals, or both.

It seems significant to relate that the rural roads in question are oiled twice yearly to suppress dust. The oilings are performed in late June and late August, which indicates that the oil has lost its capability to suppress dust during July and August and from August to June. It seems fair to assume that the predominant mechanisms of oil loss from the road in the summer are volatilization and possibly dust transport. During the other months, rain runoff is most likely the predominant mechanism since roughly 80 percent of the yearly rainfall occurs during the months of September through June (see table 2). In leaving the road through the various mechanisms described, the oil does not perform its intended function of dust suppression; however, it does pose a significant threat of ecological damage.

Conclusions and Recommendations

Roughly 1 percent of the total oil conservatively estimated to have been applied to the test roads remains in the top inch of road surface. Penetration of oil below this top inch is minimal.

Oil leaves the road during wet weather by flotation from wet road surface material and by flotation of oil-wet road surface particles.

Lead, which is contained in the waste crankcase oil, also leaves the road surface with runoff.

The road surface contains a considerable amount (≃ 200 mg/kg average) of lead which, if associated with dust particles on the road, could be carried by winds to contaminate fields and crops adjacent to the oiled road. This could be a problem where roadside crops are for direct human consumption (such as lettuce, cabbage, etc.) and/or where lead from the oiled road adds to the lead fallout on these crops from vehicular traffic.

Precise quantitation of oil movements was beyond the scope of this study; only estimates of major oil movements were made. The estimates apply most specifically to the roads tested and will vary somewhat with other roads and oiling conditions. Therefore, extrapolation of these data to other roads must be made with care.

Further study should be undertaken to determine the ecological consequences of runoff oil and its accompanying heavy metals, including lead. If preliminary evidence from such a study indicates ecological harm, then studies should be undertaken to define economically attractive alternatives to the application of waste oils for dust control and means for implementation of the alternatives.

Acknowledgments

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The cooperation of Howard Oldenburg, Mayor, Harry W. Haver, Clerk, and the Township Committee of Readington Township, New Jersey, is also gratefully acknowledged.

Figure 2. Woodschurch Road looking north from our sampling station. Tint on road surface is due to oil. Photo taken after a light rain and 6 months after the last oiling in August 1971.

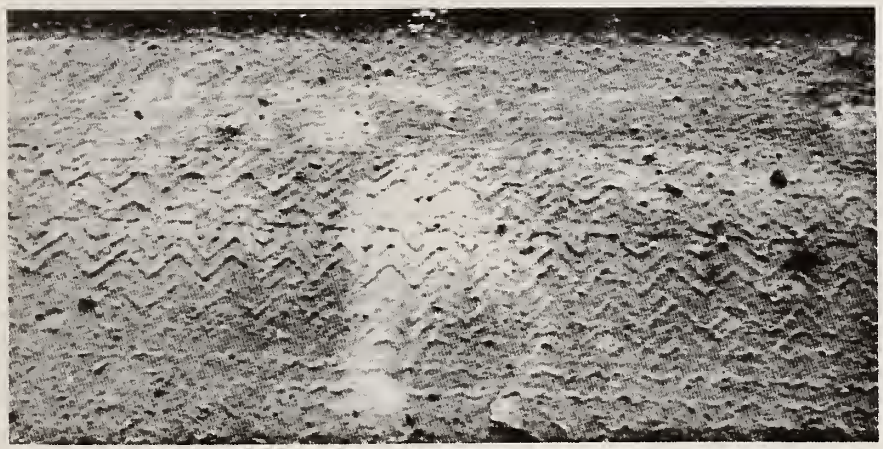

Figure 3. Oil patch has been squeezed from soft road surface material by compression of vehicle tires. When walking on oiled sections of this road, one's footprints become tinted bluish.

Figure 4. Oil on puddle surface, Woodschurch Road.

Figure 5. Oil which has been squeezed from road surface material moves with water runoff toward drainage ditch at the side of the road.

Figure 6. Oil on the surface of a drainage ditch at the side of the road. This oily water will be transported from this ditch into streams during periods of rain.

References

[1] U.S. Attorney's Office, Estimate based upon Study of Waste Oil Disposal Practices in Massachusetts (A. D. Little, Inc., Jan. 1969) and other sources.

[2] American Petroleum Institute, Final Report on the Task Force on Used Oil Disposal.

[3] Motto, H. L., et al., Lead in soils and plants: its relationship to traffic volume and proximity to highways, Environmental Science and Technology 4, 231-237 (Mar. 1970).

National Bureau of Standards Special Publication 556. Proceedings of a Workshop on Measurements and Standards for Recycled Oil - II held at NBS, Gaithersburg, Maryland, November 29 and 30, 1977. (Issued September 1979)

TEST PROCEDURES FOR RECYCLED OIL USED AS BURNER FUEL

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Before moving into my discussion of test procedures, I would like to briefly review some recent history relating to fuel oils. Although the development of the first successful oil well in northwestern Pennsylvania in the summer of 1859 predates the formation of the National Bureau of Standards (NBS) in 1901 by several years, the NBS became involved in the fuel oil specifications and tests at an early date [1].[1] The first general set of fuel oil classifications in the United States was issued by the NBS in 1929. These standards were the first industrywide attempt to classify fuels according to significant properties. These standards, based on a joint conference of consumers, burner manufacturers, and general interest groups held on January 9, 1929, were issued as Commercial Standard (CS) CS 12-29. This standard was revised several times, with the final revision issued in 1948 as CS 12-48 [2], and it remained in effect until 1966, at which time it was withdrawn.

As shown in table 1, these specifications are similar to the currently widely used specifications VV-F-815C or to the American Society for Testing and Materials (ASTM) D 396-76. Changes have occurred since 1948 in distillation temperature, flash point, bottom settlings and water (BS&W), gravity, viscosity, and sulfur. Grade No. 5 has been divided into light and heavy. Carbon residue, pour point, and ash have remained the same.

The NBS resumed activity in this area soon after the passage of Public Law 94-163, the Energy Policy and Conservation Act, in December 1975. This law is directed toward encouraging the recycling of used oils and reducing the environmental hazards associated with improper disposal of used oils. In section 383, the NBS is directed to develop test procedures which will establish the "substantial equivalency" of recycled oil to virgin oil for all end uses. What we believe is meant by substantial equivalency is that the performance is essentially the same as a virgin oil when measured by a set of identified relevant test parameters. The initial phase of the program is directed at test procedures for recycled oil for use as fuel, grade Nos. 1 through 6.

The NBS utilizes its own technical expertise to review existing test procedures. These reviews include consultation and experimental work by the staff of various NBS divisions, as well as the establishment of a special petroleum testing laboratory.

For each fuel oil grade we planned the following approach.

- Review existing virgin oil test procedures.
- Evaluate test applicability to used oils, recycled oils, and used oil/virgin oil blends (laboratory program).
- Develop new or revised test procedures where needed (laboratory program).
- Transmit set of approved test procedures for recycled fuel oils to the Federal Trade Commission (FTC).
- Provide technical support to the FTC.

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[1] Underlined numbers in brackets indicate the literature references at the end of this report.

Table 1. Commercial Standard CS12-48

Fuel oil grade	Flash point °F	Pour point °F	Water and sediment %	Carbon residue on 10% residuum %	Ash %	Distillation temps, °F			Viscosity Kinematic centistokes at				Gravity ° API	Sulfur %
						10% point	90% point	End point	100° F		122° F			
	Min	Max	Max	Max	Max	Max	Max	Max	Max	Min	Max	Min	Min	Max
1	100 or legal	0	Trace	0.15	----	420	---	625	2.2	1.4	---	----	35	0.5
2	100 or legal	20	0.10	0.35	----		675	---	4.3	---	---	----	26	1.0
4	130 or legal	20	.50	----	0.10	---	---	---	26.4	5.8	---	----	---	No limit
5	130 or legal	---	1.00	----	0.10	---	---	---	---	32.1	81	----	---	No limit
6	150 or legal	---	2.00	----	----	---	---	---	---	---	638	92	---	No limit

The law requires NBS to develop test procedures to demonstrate the substantial equivalency of recycled oils to virgin oils. From a practical point of view, this requires test procedures to demonstrate equivalent performance for each fuel oil grade.

Fuel oils fall into two main categories, distillate and residual. The intended general usage of each fuel oil grade is indicated below.

Grade No. 1:	Distillate oil intended for vaporizing pot-type burners.
Grade No. 2:	Distillate oil for general purpose heating use.
Grade No. 4:	Light residual or heavy distillate with preheating not required for handling or burning.
Grade No. 5: (Light)	Residual oil which may require preheating, depending on climate.
Grade No. 5: (Heavy)	Residual oil which may require preheating for burning and handling.
Grade No. 6:	Residual oil requiring preheating for burning and handling.

Grade No. 3 was not generally accepted and was dropped in 1948 [3].

The consumption by grade under Federal specifications [4] is indicated in table 2 and the total U.S. consumption for 1975 [5] is indicated in table 3.

Table 2. Fuel Oil Consumption Under Federal Specifications[a]

Grade No. 2	427 million gallons
Grade No. 4	28 million gallons
Grade No. 5	58 million gallons
Grade No. 6	355 million gallons

Table 3. Total U. S. Consumption of Heating Oils

Distillate	1,043 million barrels
Residual	895 million barrels

Distillate oils are commonly used, for example, in residential heating; the residuals are used in power plants or industrial-type installations. It is interesting to note that the average price for No. 2 fuel oil in current dollars increased from $0.15 per gallon in 1955 to $0.41 per gallon in 1976 [5].

The currently accepted specifications for virgin fuel oils are listed below.

VV-F-815C	Federal Specification, Fuel Oil, Burner
ASTM D 396-76	Standard Specification for Fuel Oils
MIL-F-16884G	Military Specification, Fuel Oil, Diesel, Marine
MIL-F-859E	Military Specification, Fuel, Burner

Most fuel oil purchased for the U.S. Government is under VV-F-815C, which is similar in many respects to ASTM D 396-76. These specifications are summarized in table 4. The numbers in parentheses refer to Celsius temperatures. The Saybolt viscosities have been dropped to simplify the table. Note that "legal" refers to the maximum set by local or regional authorities.

Our investigations to date indicate that relatively few changes in the test procedures called for in these specifications should be required in order to accommodate the use of recycled oils as fuel.

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[a] Consumption figures for Grade No. 1 were not available.

Table 4. Current Specifications for D 396-76 and VV-F-815C

Test procedures	Fuel oil grades					
	No. 1	No. 2	No. 4	No. 5 (Light)	No. 5 (Heavy)	No. 6 (Bunker C)
Flash point, °F (°C) [Min]	100 (38) or legal	100 (38) or legal	130 (55) or legal	130 (55) or legal	130 (55) or legal	150 (65)
Pour point, °F (°C) [Max]	0 (-18)	20 (-7)	20 (-7)	--	--	--
Water and sediment (V%) [Max]	0.05	0.05	0.50	1.00	1.00	2.00
Carbon residue on 10% bottoms [Max] %	0.15	0.35	--	--	--	--
Ash weight, % [Max]	--	--	0.10	0.10	0.10	--
Distillation Temp., °F (°C)						
10% point (Max)	420 (215)	--	--	--	--	--
90% point (Min)	--	540 (282)	--	--	--	--
(Max)	550 (288)	640 (338)	--	--	--	--
Kinematic viscosity, cSt						
at 100° F (38° C)						
Min	1.4	2.0	5.8	>26.4	>65	--
Max	2.2	3.6	26.4	65	194	--
at 122° F (50° C)						
Min	--	--	--	--	(42)	>92
Max	--	--	--	--	(81)	638
° API gravity (Min)	35	30	--	--	--	--
Copper strip corrosion (Max) (ASTM , 50°C, FED. SPEC. 100°C)	No. 3	No. 3	--	--	--	--
Sulfur, % (Max)	0.5 or legal	0.5 or legal	legal	legal	legal	legal
Aniline point, °F (°C) (Fed. spec. only)	Report	Report				

After our initial review of the test procedures required in these specifications, it was apparent that some test methods might be more affected by the presence of used oil or recycled oils than others. I will limit my discussion to a few illustrative examples. It should be noted that the tests described are applicable to both recycled crankcase oils and industrial oils.

In general, test procedures for fuel oils have been evaluated on the basis of (a) applicability to recycled oils and recycled oil/fuel oil blends; (b) if applicable, the extent to which the presence of recycled oil affects the precision of each procedure set forth in the test method; and (c) the special properties of recycled oils affecting equivalent performance which may require additional tests not necessary for "new" oils. For our laboratory evaluations of the test procedures, we used a variety of composite automotive crankcase drainings and recycled fuel oils.

I would now like to consider some specific test procedures.

Sampling of Fuel Oils

Fuel oil specifications and standards vary considerably in their requirements for sampling, ranging from no requirements to fairly detailed requirements. The presence of waste oils has two potential effects on sampling: a less homogeneous mixture due to separation on standing and a lowered stability of the fuel oil. Since a sampling procedure may be required, we are considering one based on MIL-F-859E. For bulk lots (where a bulk lot is defined as an indefinite quantity of hydrocarbon oil of one grade), upper, middle, and lower samples will be taken using thief or bottle samples as described in ASTM method D 270. Samples may be composited to a single sample if homogeneous. Further, the procedure states that homogeneity will be determined on the basis of density, ASTM method D 1298. Lots will be considered homogeneous if the specific gravity determinations for the upper, middle, and lower samples do not vary from the average by more than 0.0012 density units or 0.3° API (American Petroleum Institute) gravity.

Carbon Residue

Carbon residue is normally specified only for distillate fuel oils, grade Nos. 1 and 2, and is measured on the 10-percent residue after distillation. The carbon residue test appears to give excellent results in predicting the performance of distillate fuels heavier than grade No. 1. The simpler Ramsbottom method, ASTM D 524, is generally used in preference to the Conradson method, ASTM D 189. High carbon residue would indicate a tendency to form excessive carbon on burning, greatly reducing the efficiency of vaporizing pot-type burners or fouling nozzles in small residential-type burners. The carbon residue test is used primarily to control the carbon-forming tendency of No. 2 fuel. No. 1 normally would be so low in carbon residue as to be of little significance. The test values are often affected by the presence of used or recycled oils. This is due to the fact that the test is run on 10-percent residuum remaining after distillation and, thus, concentrates the high boiling fractions in the residuum. The repeatability of the method falls within the stated limits for the method.

Ashing Procedures

Two general methods are used for determination of ash in petroleum products, ASTM D 482 and D 874. Regular ash, D 482, is generally called for with the residual grades. However, according to the procedure it is not intended for waste crankcase oils or additive-containing oils where D 874 is specified. D 874, sulfated ash, has been used as an indication of additive level in oils, but again, is not recommended for used automotive oils according to the procedure.

For the residual fuels, ash results from a variety of inorganic compounds present in the crude oil, as well as contaminants acquired during transportation or storage. Used automotive oils added to fuel oils would be expected to contribute to the ash primarily due to high lead content, plus various additive metals such as calcium, barium, zinc, and magnesium. Ash in residuals is important as it can cause high-temperature corrosion of boilers, refractory erosion, or excessive deposits.

During our investigation of these ashing methods, we found that the regular and sulfated ashes give approximately the same values for the virgin oils. The Bartlesville Energy Research Center (BERC) annual heating oil survey showed a typical maximum of 0.5 percent ash for virgin No. 6 residual oil [6]. Since most used oils have considerably higher ash contents than 0.5 percent, it could be expected that ash-forming constituents would be of concern with regard to the substantial equivalency of recycled fuel oils to virgin fuel oils.

To determine if there is singificant metal loss during ashing using the sulfated as procedure, the metal content of used automotive oils was determined and compared with th metal content of the ash determined using atomic absorption spectroscopy. Although we have not completed analysis of the ashes, the data to date, based on a materials balance for lead, calcium, zinc and magnesium, indicate D 874 is satisfactory for use with used and recycled oils.

Lead Analysis

At the present levels of consumption of leaded gasoline, significant concentrations of lead may be anticipated for some time in used automotive crankcase oils. It is well documented that lead is an environmental hazard. Lead content, negligible in virgin fuel oils, occurs in concentrations of 0.5 to 1.5 percent in typical crankcase drain oils. Considerable effort by the EPA has gone into studies of the environmental impact of burning fuels containing lead and the feasibility of lead removal where emission controls are not available. Obviously, if it is determined by the FTC and/or EPA that lead is important in recycled fuel oils, a test procedure for lead will be required.

Various analytical methods have been evaluated in our Analytical Chemistry Division to establish their applicability to recycled oils. Determination of lead in recycled oils is difficult because the lead is present both in the solid phase and, presumably, also in the form of oil-soluble lead compounds. Sampling to obtain a representative and homogeneous sample requires considerable care.

At the present time, we have found a number of problems with existing analytical methods for lead in used crankcase oils, and are in the process of searching for a method with acceptable accuracy and precision.

Summary

In the limited time available, I have been able to mention only a few of the test procedures we have been working on. Others include water and sediment, copper corrosion and a test procedure for heating value. Several test methods require additional researc For example, polynuclear aromatic hydrocarbons (PAH's), or polycyclic organic matter, have been of interest due to the toxicity of some of these compounds. Work on development of a test procedure for PAH is discussed in another presentation at this conference.

We are also investigating chlorine and bromine test procedures. It is at present unclear whether these halogens are important for equivalence with virgin fuel oils. The complete report of our efforts in this phase of the NBS Recycled Oil Program will soon be available.

In conclusion, I would like to acknowledge our student help in the laboratory, Messrs. Richard Doitteau and Steve Serkiz, and to express our appreciation to the Association of Petroleum Re-refiners for their support of a research associate at the NBS. The very numerous contributions from the NBS staff in assisting with the program are appreciated, as well as the cooperation of various agencies and organizations outside the NBS.

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SESSION IV. LEGISLATION AND STUDIES

National Bureau of Standards Special Publication 556. Proceedings of a Workshop on Measurements and Standards for Recycled Oil - II held at NBS, Gaithersburg, Maryland, November 29 and 30, 1977. (Issued September 1979)

RECENT FEDERAL AND STATE LEGISLATIVE DEVELOPMENTS IN USED OIL RECYCLING

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United States legislation, both Federal and State, governing collection, recycling, and disposal of used oil has developed dramatically in the last 2 years. This has occurred in response to increased awareness of the needs to conserve a valuable resource and to protect environmental and public health from risks posed from improper reuse or disposal.

This article summarizes existing Federal laws under which used oil might be regulated; bills currently pending in the Senate and the House of Representatives; the model used oil recycling act; and, finally, State legislative initiatives--some enacted, some not--for controlling used oil collection, recycling, or disposal.

Existing Federal Laws

Various issues raised by recycling used oil are addressed by recent Federal laws. It is perhaps not likely, but certain oil products might be regulated as new or existing chemical substances or mixtures under the Toxic Substances Control Act, passed in October 1976. The operative standard throughout that statute is whether a substance poses an unreasonable risk of injury to health or the environment. Notorious substances like PCB's [polychlorinated biphenyls] come to mind in this connection, but there is no reason to think that products or derivatives made from oil--in new or used form--are beyond its scope.

If the Administrator of the Environmental Protection Agency (EPA) finds that the manufacture, distribution in commerce, processing, use, <u>or disposal</u> of a chemical substance or mixture <u>may present</u> an unreasonable risk of <u>injury to health</u> or to the environment, he <u>may subject it</u> to testing under section 4 or to regulation under section 6 of the Toxic Substances Control Act.

It is more likely that used oil would be regulated under the 1976 amendments to the Solid Waste Disposal Act, amended as the Resource Conservation and Recovery Act (RCRA). Flammability and toxicity are two of the criteria listed in section 3001 of the RCRA as providing the basis for regulations to be issued by the EPA which will identify characteristics of hazardous wastes and will list particular hazardous wastes or processes which produce them. Since used oil, or contaminated used oil, is sometimes flammable or toxic (or both), it has been considered a candidate for control under the hazardous waste provisions of the RCRA. While it would be justifiable to regulate used oil which <u>is</u> flammable or toxic as a hazardous waste, it would not be accurate to put <u>all</u> used <u>oil</u> on a list of hazardous wastes. Section 1004(5) defines hazardous wastes <u>as</u> substances that "...pose a substantial present or potential hazard to human health or the environment when improperly treated, stored, transported, or disposed of, or otherwise managed," or that "...cause, or significantly contribute to an increase in mortality or an increase in serious irreversible, or incapacitating reversible, illness." If a substance does not meet these tests or the section 3001 criteria, then by definition it is not a hazardous waste, and no amount of administrative convenience or discretion can make it legal to treat it as such.

As a practical matter, the RCRA's controls over generators, transporters, and disposers of hazardous wastes are admirably thorough (see sections 3002-3005). If the EPA determines that used oil qualifies as such a waste, then all sources and handlers of used oil would have to be regulated in hazardous waste management programs called for by subchapters C and D. For this reason, it is unlikely that any State would be eager to manage or that the Federal Government would be able to manage such programs if they were to include used oil as a hazardous waste. One possible means of limiting the scope of responsibility for used oil management is suggested by the EPA's draft RCRA regulations establishing standards for generators, transporters, and disposers of hazardous waste. The Administrator is only required by sections 3002-3004 to promulgate such standards "...as may be necessary to protect human health and the environment." Depending on its characteristics, it is possible that it would not be necessary to subject less than certain minimum amounts of used oil to such regulation.

As this indicates, the question is not simply whether or not used oil is put on an EPA list of hazardous wastes. Rather, it is whether any given batch of used oil, contaminated or not, meets the characteristics set forth in agency regulations for identifying a hazardous waste. If it does, it should be so treated by any person generating, transporting, or disposing of it. And if any such person should fail to do so, he would be liable to a citizen suit under section 7002. The issue of the relationship between the criteria identifying the characteristics of hazardous substances and the lists of the substances themselves has not yet been resolved, but the agency views the RCRA as controlling substances which meet the characteristics, whether or not they are on the list. The following quote from the cover letter of the most recent draft regulations on hazardous waste criteria, characteristics, and lists indicates the nature of this issue:

> "(5) Waste listings - The draft regulation does not include any lists of hazardous wastes. The Agency working group is still evaluating the use of lists with the criteria. Several options for lists include waste types, process types, industry categories, substances, or a combination. In addition, whether the list is an example of wastes that are potentially hazardous, a definitive enforceable list of those defined to be hazardous, or some other approach is still under discussion. The relationship of the criteria to these list types is also under review. Your comments on this issue would...be appreciated" [1].[1]

Further attention should be paid to developing the test procedure for determining toxicity of a potentially hazardous waste. The flammability test method proposed in the draft RCRA section 3001 regulations is the American Society for Testing and Materials standard D 93-73, using a Pensky-Martens Closed Cup Tester, which seems readily applicable to used oil. But the toxicity procedure may be inappropriate for small sources or batches of used oil. Another possible test procedure may come from work being done in Germany on means for collectors to determine the percentage of foreign matter in used oil.

A Federal law that deals specifically with used oil is the 1975 Energy Policy and Conservation Act (EPCA). Section 383 of the EPCA requires, among other things, that the Federal Trade Commission (FTC) prescribe labeling standards for containers of recycled oil "...indicating any particular end use for which a determination of substantial equivalency has been made." The FTC standards will preempt all other labeling rules addressing comparative characteristics of recycled oil. Under this provision, the EPA must also issue labeling standards "...applicable to containers of new oil, used oil, and recycled oil relating to the proper disposal of such oils after use." There is a possibility that the FTC and EPA rules may be combined into one set of requirements. The EPA has a good deal of leeway in setting its regulations: their purpose is "...to reduce, to the maximum extent practicable, environmental hazards and wasteful practices associated with the disposal of such oils after use." It would be possible--perhaps even advisable--for the EPA to develop three sets of labeling standards: one each for new, used, and recycled oil. That would be one way to make the regulations specific to the kinds of hazards and wasteful practices associated with each category of oil. There are, of course, subcategories of each of these, too, as discussed above in connection with the RCRA: some kinds of used oil pose severe environmental hazards and some do not, depending on what is done with it and where. There are also questions of how to label used oil containers: it is doubtful that the regulations would extend to the milk cartons frequently used by do-it-yourself oil changers, but that suggestion illustrates the problem.

Used oil can be burned as a fuel supplement. In some circumstances, e.g., where use of re-refining capacity is economically efficient, that might be a wasteful

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[1] Underlined numbers in brackets indicate the literature references at the end of this paper.

practice. Burning might also pose environmental hazards, depending on the environmental circumstances and the nature of the oil being burned. One approach to this question is to ask whether the burning will violate ambient air quality standards. These standards currently exist for particulates, and a lead standard was proposed in December 1977. The EPA is studying certain unregulated pollutants, including polynuclear aromatic hydrocarbons (which are sometimes found in used oil), to see whether they should be regulated via ambient air quality standards, hazardous pollutant emission standards, or new source performance standards.

When all the provisions of the Federal law summarized above are fully implemented, controls over used oil will be significantly increased, if not comprehensive.

Congressional Bills

Currently there are three different National Oil Recycling Acts introduced in various congressional committees. Two of them would remove the biggest economic block in the way of successful used oil recycling: the Internal Revenue Service (IRS) ruling that denies to re-refiners and to consumers the $0.06-per-gallon tax credit on virgin oil not used for highway vehicles. This can only be relieved by Congress, as the IRS seems unresponsive to the energy and environmental aspects of the problem. As part of the Energy Production and Conservation Tax Incentive Act (H.R. 5263), the House passed an exemption from the excise tax for new oil used in products that contain 25 percent or more re-refined oil and 55 percent or less new oil. Chances are good for passage of this bill by the Senate. The used oil bill introduced by Representative Donald Mitchell of New York in March 1977 (H.R. 5350) would similarly amend the tax code. Senator Pete Domenici's bill, introduced in May 1977 (S. 1482) not only allows the exemption, but also provides a $0.03-per-gallon tax credit for lubricating oil used in re-refining used oil. The outlook for these two bills is gloomy, though, because they are close to the bottom of Congress' list of priorities.

A third bill, introduced in the House by Representative Vanik in August 1977 (H.R. 8909), makes no mention of the tax problem. However, like the other two, Rep. Vanik's bill requires use of recycled oil by all Federal agencies, as well as by Federal contractors, and it sets up a consumer education program. All three bills provide for Federal grants to States for waste oil management plans, which, in order to qualify for funds, must issue permits to collectors and recyclers. The Mitchell and Vanik bills go farther here than does Senator Domenici's. The House bills compel State plans to require sellers of automotive oil to maintain collection facilities on their premises, and State plans must prohibit burning of used oil as fuel and use for road oiling unless the oil meets Federal and State pollution control standards. Both House bills also require that automotive oil sold for self-service or carry-away by consumers be packaged in resealable containers with a $0.30 deposit on each container, refundable upon return (either empty or full). A similar provision in Maryland's bill (S. 936) caused the governor to veto it.

Model Used Oil Recycling Act

In 1976 a model used oil recycling act was prepared for the Federal Energy Administration (see Congressional Record, September 30, 1976, pp. S17433-S17447).[2] The model bill states as its policy that, "Used oil shall be collected and recycled to the maximum extent possible, by means which are economically feasible and environmentally sound...." It defines "recycle" as "...to prepare... for reuse as a petroleum product...or to use...in a manner that substitutes for a petroleum product made from new oil...." The bill would require a government agency to issue rules which "...prescribe means for the provision of safe and conveniently located collection facilities for the deposit of used oil by persons possessing not more than 5 gallons at one time at no cost...." Rules would also require sellers of lubricating oils to post signs telling customers where such collection facilities are. The agency would also have to conduct a public education program and a licensing program. The former would involve assigning a person to inform people about the law and where to dispose of used oil and to encourage and assist the development of voluntary local recycling programs. The licensing program is designed to direct the flow of used oil via approved collectors only to approved recyclers or disposers. (A person who disposes of more than a certain amount of used oil by means other than recycling, e.g., road oiling, incineration, or landfilling, would have to obtain a special permit

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[2] The following description of the model bill is quoted from reference 2 with the permission of the author.

instead of a license.) In deciding whether or not to grant an application for a license or special permit, the agency would have to determine "...that the proposed means for collection, transport, transfer, storage, recycling, use, or disposal is operationally safe, environmentally sound and consistent with the policy of this Act..." and would have to "...impose terms...necessary to insure continuous compliance with existing laws and regulations." Licensed collectors and recyclers would leave receipts and keep records of their transactions and submit annual reports to the agency in order that the agency would be able to monitor and eventually manage used oil flows. The agency would also provide an annual report to the legislature on the law's effectiveness. The model bill would authorize recycled oil products to be represented as substantially equivalent to new oil products if they meet new oil product specifications (or have been determined to do so under FTC rules) and would require government officials to encourage the purchase of substantially equivalent recycled oil products. Finally, the bill prohibits disposal of used oil by discharge to sewers, surface or ground waters, or marine waters, and by incineration or land deposit unless in accordance with a special permit.

Legislation Enacted in California, Minnesota, and Utah [3]

The California Used Oil Recycling Act (Cal. Pub. Res. Code. SS 3460-3473), patterned after the model act, was passed by the legislature and signed by the governor on September 30, 1977. The final version of the act contains substantially the same legislative findings and policy statements as are provided in the model act, and it requires a similar public education program. Although the California act establishes a program for collecting and recycling used oil that is similar to that in the model act, several important deviations from the model act limit its coverage to regulation of recycling, rather than disposal, methods.

California's coverage of used oil disposal methods fails to include the model act's requirement for special permits for used oil disposers. California expressly permits application of used oil for purposes of road maintenance, agricultural dust control, or weed abatement. The model act's specific prohibitions on disposal "...by discharge into sewers, drainage systems, surface or ground waters, watercourses, or marine waters; or by incineration or deposit on land..." are retained, but road maintenance, dust control, weed abatement, and burning for fuel are excluded from the definition of "disposal." The deletion of the special permit requirement may not seriously hamper the effectiveness of the prohibitions; but in characterizing certain uses as not constituting "disposal," the California version of the model act leaves open the possibility that environmental and health hazards will continue to be created through unregulated methods of application for the permitted purposes.

The California act requires used oil collectors and recyclers to be registered rather than licensed, and it modifies the model act's definition of "collector" to exclude persons "...collecting solely from sources owned and operated..." by themselves. The change in language from "licensed" to "registered" does not appear overly significant since the California Solid Waste Management Board is required to register collectors and recyclers only if it "...determines that the [applicant's] proposed means for collection, transport, transfer, storage, recycling, use, or disposal is operationally safe, environmentally sound, and consistent with the provisions of [the Act]." The Board is also given the authority to impose restrictions or other terms "...requiring the registration holder to install or effect controls, processes, or practices necessary to insure continuous compliance with existing laws and regulations...", and registration can be revoked for noncompliance. The decision not to require registration by persons "collecting" their own oil, while it may result in permitting unregulated collection and transport of used oil by large commercial enterprises with geographically dispersed operations, does not undermine the basic thrust of the registration requirements. Although California requires only a one-time registration (rather than annual renewals), it does retain, both for recyclers and for collectors, the same record-keeping requirements that are provided in the model act.

The provision of collection facilities for the use of do-it-yourselfers and others who generate small quantities of used oil will be handled differently in California than under the model act. Persons selling oil to consumers need only post notices informing customers of the location of collection facilities. Rather than authorizing a requirement that sellers provide collection facilities, the California act delegates authority to the Solid Waste Management Board to adopt rules prescribing "...means for the provision of safe and conveniently located collection facilities for the deposit of used oil by persons possessing not more than five gallons at one time at no cost to those persons." Since earlier language requiring the Board to mandate that sellers create and maintain such facilities was dropped, the rulemaking authority of the Board probably would be construed as limited to regulation of voluntary private facilities, but it would include authority to adopt rules requiring maintenance of such facilities by State agencies and local governments.

The definition of used oil limits the California used oil program's coverage to oil that has been sold to consumers and is recyclable. By omitting nonrecyclable used oil from the definition, the California act allows continued unregulated disposal of oils which may nevertheless pose environmental hazards if disposed of improperly. Further, the addition of the phrase, "which, after sale to a consumer," could also be construed to exempt most industrial oils from coverage, although these may pose the same kinds of hazards as oil that has been sold to a consumer. Such an interpretation of the act should be avoided.

The California act, despite its drawbacks, is the most comprehensive effort by a State to control disposal and assist in recycling and collecting used oil. It still closely resembles the model act, and it demonstrates the usefulness of such model legislation in the legislative process. Undoubtedly, as other States continue to consider used oil legislation, the model act and its California incarnation will exert considerable influence.

Neither the Minnesota nor the Utah statute is as comprehensive as the California law or the model act. Instead, legislators in these States have focused on specific problems associated with collection or re-refining of used oil. Their legislation, although less complicated than the model act, fails to regulate the final disposition of used oil. Nevertheless, these statutes represent the first efforts to confront the used oil problem at the State level and should be effective in fulfilling their limited purposes.

Minnesota's statute (Minn. Stat. SS 325.814-816) focuses on the need to provide convenient collection facilities for oil used in motor vehicles. Rather than provide a general definition of used oil as in the model act, Minnesota has defined "used motor oil" to include only "...petroleum based oil used as a lubricant in a motor vehicle..." which "...through use, storage or handling has become unsuitable for its original purpose...." All retail sellers of motor oil are required either to provide collection facilities which are "accessible to the public" or to post notices informing customers of the location of the nearest collection facility. The Minnesota statute thus solves the critical problem (addressed in section 7 of the model act) of ensuring that do-it-yourselfers will know where they can dispose of their oil. The statute provides only a partial solution, since it neither requires users to deposit the oil at collection facilities, nor addresses the issue of how to ensure safe disposal once the oil has been collected.

The Utah Oil Rerefinement Act (1977 Utah Laws, ch. 55) is designed primarily to encourage used oil re-refining. After stating that re-refining is desirable and that used oil collection systems need to be better coordinated in order to channel more oil to re-refiners, the statute proceeds to regulate who may re-refine used oil. Re-refiners are required to obtain permits, and they must provide information on proposed operations--including methods of disposing of re-refining wastes--before permits can be granted. Furthermore, "...every automobile service station, boat marina, industrial operation, airport, trucking terminal or state or local government facility which generates at least 500 gallons of used oil annually..." is required to dispose of used oil, regardless of quantity, through either a "...used oil dealer or rerefiner."

The principal reason for the latter requirement is to channel quantities of oil that would otherwise be improperly disposed of or wasted into the hands of used oil dealers who can then sell the oil to re-refiners. However, the Utah statute leaves to used oil dealers the decision whether oil should be sold to re-refiners or otherwise disposed of. Further, it does not regulate uses or disposal methods other than re-refining, nor does it impose substantive waste disposal standards on re-refiners. These omissions occurred despite a legislative finding that used oil constitutes an environmental and a human health hazard.

Proposed State Legislation

Eleven of the 15 State used oil bills that we have studied provide for collection of used oil by retailers of lubricating or other oils, by service stations and mechanics, or, as in Maryland (in S.B. 936), by designated State inspection facilities. New York's bill (N.Y.A. 6289) goes no further than to require service stations and repair shops that sell motor oil to provide a collection receptacle for anyone wishing to use it. The Illinois bill (H.B. 358) includes a requirement that retailers provide collection facilities; exempted from it are those retail outlets that do not have oil-change facilities but that do have a written contract with a nearby storage facility for disposal.

Only 5 of the 15 bills mention getting rid of the oil once it has been collected. Pennsylvania's bill (H.B. 724) simply requires that the oil be removed periodically; it does not say how this should be done, and the bill puts no restrictions on disposal or use except that used oil burned for fuel must meet standards set by Federal and State air pollution control laws. Wisconsin's bill (S.B. 301) requires storage sites to be serviced only "...where economically and geographically feasible." Nevada's bill (A.C.R. 37) requires that, if possible, regulated establishments shall contract for its removal by used oil dealers for sale to re-refiners. The Maryland bill dictates that the oil be removed and disposed of legally; it also prohibits disposal other than by recycling or by other means permitted by the administering agency. Wyoming's bill (H. 319) and Florida's bills (S. 310; H. 1763) are identical to the model bill and provide for collection facilities for free use by persons with five gallons or less of used oil. Transporters of more than 500 gallons of used oil annually, storage facilities receiving more than 10,000 gallons of used oil annually, and recyclers of 5,000 gallons or more per year must be licensed and are subject to record-keeping and annual reporting requirements. Eight of the 15 bills included some kind of license, permit, or registration requirement for transporters and/or recyclers.

Disposal by means other than recycling without a permit is allowed by Pennsylvania's bill, which includes only the air pollution control standard mentioned earlier; by Illinois' bill, which makes no mention of any regulation of disposal or use; and by Wisconsin's bill, which allows use of unlimited quantities of used oil for dust control. Wisconsin, however, prohibits disposal into sewers and water and by incineration. Maryland's, Florida's, and Wyoming's bills allow for the issuance of permits for disposal other than recycling; Maryland would require a permit for such disposal of more than 55 gallons annually; and the Florida and Wyoming bills reflect the model bill by not setting a minimum amount for which a permit is needed.

Conclusion

The bills in the U.S. Congress, as well as those in various stages of consideration by State legislatures, have not progressed further in part because of a lack of widespread appreciation of the opportunities used oil recycling presents for combined measures for environmental protection and resource conservation. These measures need not be either draconian or administratively burdensome to public or private institutions. Modest recycling programs at the State and local level, tailored to the needs of the area, can result in significant cumulative gains in addition to those which may result from the implementation of Federal legislation.

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National Bureau of Standards Special Publication 556. Proceedings of a Workshop on Measurements and Standards for Recycled Oil - II held at NBS, Gaithersburg, Maryland, November 29 and 30, 1977. (Issued September 1979)

EPA ACTIVITIES IN WASTE OIL MANAGEMENT

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Resource recovery of waste products in general has been receiving greater attention because of the inherent resource and energy conservation benefits as well as the positive environmental effects. Each year our Nation produces, consumes, and throws away an increasing amount of waste. Multiple packaging, built-in obsolescence, and the convenience of disposable consumer items contribute to enormous resource losses. With only 7 percent of the world's population, we consume almost one-half of the earth's industrial raw material, yet we are recycling a lower percentage of our resources than ever before in history. Studies conducted by the Environmental Protection Agency (EPA) indicate that the failure to control the amounts of wastes produced and to recover waste resources have far-reaching environmental consequences. When two production systems are compared--one using virgin materials and the other secondary materials--the system using wastes typically causes less air and water pollution, generates less solid waste, and consumes less energy. Thus, resource recovery of any waste product, including used oil, is an important environmental concern.

The conservation and recovery of petroleum products is not a new phenomenon in this country. In fact, waste oil reclamation in the United States dates back to about 1915. The lubricating oils in use at that time were recycled by first heating waste oil, then adding a coagulant to accelerate settling, and finally centrifuging. Following this treatment, the renewed oils were then ready to resume their lubricating functions. The Armed Forces made use of such systems to conserve lube oils at many Army depots during the first world war, and the military's use of recycled lube oils continued after that war.

In 1932, commercial airlines first instituted similar oil recycling systems by segregating their oils and contracting with the re-refining industry to custom treat aircraft oils. These programs resulted in savings of 20 percent in lube oil costs to the airline industry and served to stimulate the use of re-refined oils by other industries. By 1939, the re-refining industry was processing 11 billion gallons per year of waste oil.

During the second world war the need to conserve limited supplies of petroleum, reduce costs, and simplify logistics led to the widescale use of re-refined lube oil products. Approximately 29 million hours of aircraft engine time were logged using re-refined lube oil without any evidence of deleterious effects on engine life. In fact, during that period average engine life was increased by almost 50 percent. Following the second world war, the Air Force continued to use re-refined products on an unrestricted basis. By 1949 almost one-fourth of all aircraft engine oil purchased by the Air Force was re-refined. However, with the advent of jet engines requiring synthetic-based lubricants, the need for re-refined oil products by the Air Force began to decline. The supply of waste oil feedstocks dwindled, and eventually the closed-loop arrangements between the Air Force and the re-refining industry were discontinued.

However, other markets for re-refined lube oil products continued to grow steadily. Re-refiners, using plants constructed prior to and during World War II, were able to compete favorably on an economic basis with virgin lube oil suppliers. These products went to both motorists and industrial users. In 1960, the industry consisted of 150 companies producing over 300 million gallons of re-refined lube oils. Although this represented almost 18 percent of the Nation's lube oil consumption, there were already forces at work which were to reverse the dramatic growth of the industry up to this time.

Increases in the number of motorists who change their own oil, longer periods between engine oil changes, and the increase in waste oil use in road oiling and as fuel have all served to reduce the flow of waste oil feedstock to the re-refining industry. Competition for waste oil suppliers resulted in increases in the cost of waste oil to the re-refiner. At the same time, low prices of competitive virgin lube oils prevented re-refiners from passing through increased re-refining costs to customers. This further limited the ability of re-refiners to outbid other users for limited waste oil supplies. This cost/price squeeze further reduced feedstocks available for re-refining, resulting in even more inefficient operation of plants. This condition continues to this day, when industry production averages from 30 to 50 percent of installed capacity.

During this same period, lube oil formulation became more complex in order to satisfy specifications for higher-performance engines. The need for detergents, multiviscosity oils, and longer change intervals has made lubricating formulation increasingly stringent. As a result, the job of removing the new additive compounds became more difficult, requiring higher levels of pretreatment and careful supervision of the re-refining process. Users began to perceive re-refined products as inferior, a stigma that continues to complicate the industry's marketing problems today.

Concern has also grown over residuals produced from the re-refining process that make investments in costly pollution control equipment necessary. Those residuals are in the form of highly acidic oily sludges, which have traditionally been disposed of either by dumping or lagooning. Many re-refiners have been unable to meet new disposal requirements of State and local governments and either shifted to the less-demanding production of fuel oils or ceased operations entirely.

Because of Congress's concern over these specific problems, the EPA was required under Public Law 92-500 to conduct a study of waste oil disposal problems and recycling opportunities. This report was completed and transmitted to the Congress in 1974.

The report found that the major market for waste oil collectors consists of users of fuel. This market can be divided into two segments. The first includes fuel processing, which removes many of the contaminants present in waste oil before marketing as a fuel. Most waste oil, however, is distributed directly to users who blend the waste oil with conventional fuel without first removing contaminants. These contaminants will vary, depending on the source of the waste oil, and include lead, barium, copper, zinc, phosphorus, tin, and chromium.

The most serious contaminant noted to date appears to be lead from automobile crankcase drainings. The lead, which averages 1 percent by weight in crankcase drainings, originates from lead-based gasoline additives. The uncontrolled combustion of undiluted automobile crankcase drainings can result in the emission of unacceptable levels of lead into the air. Although dilution of waste crankcase drainings can reduce lead concentrations, the uncontrolled burning of this waste is not a desirable disposal option. It is unlikely, however, that facilities not already equipped with pollution control devices will make the substantial investments required to limit contaminants from the relatively small quantities of waste oil consumed. The electric power industry, for example, which is the largest consumer of fuel oils in this country, does not generally employ pollution control devices on oil-fired units which could remove the lead particles.

The report also found that there was presently very little control over the use or final disposition of waste oil. Although a few States have enacted legislation regulating waste oil collection and disposal, efforts at enforcement have been limited. In the absence of such control, there was little incentive for fuel processors to remove the contaminants from waste oil prior to sale for use as a fuel.

The report found that another use of waste oil was in road repair, either in asphalt manufacture or applied directly as a dust suppressant. Combined, these applications accounted for 200 million gallons of waste oil use in 1972. Tests of use on roads have indicated that as much as 70 percent of waste oil applied to road surfaces could migrate to the neighboring lands either as rainwater runoff or as airborne particles.

In 1972, approximately 2.2 billion gallons of lubricating oil were sold in the United States. Approximately 50 percent of this lube oil was consumed in use, discarded with filter cartridges, or lost through leakages or in other ways, resulting in the generation of an estimated 1.1 billion gallons of waste lubricating oil. The major sources of waste oil are automotive operations, industrial and aviation operations, and other sources such as government uses and industrial process oils.

The report also concluded that the major re-refining process in use was acid/clay treatment. Another process identified and in limited use was vacuum distillation followed by clay treatment, which can include a caustic pretreatment. The acid/clay process does produce a high-quality lube stock; however, the residual sludge produced by this process presents a real disposal problem. Recent work in Canada, however, has demonstrated that these residuals can be burned in a properly operated cement kiln with virtually no adverse environmental effects.

Although solvent extraction is old in the art of lube oil manufacturing, it has not been used by waste oil reprocessors. This is because high solvent-to-oil ratios require high operating costs and continuous operation by highly skilled operating personnel. Further, all solvent extraction processes still require some acid and clay treatment, resulting in the generation of residual sludges.

The EPA has noted that new technologies for re-refining waste oil have been developed both in laboratories and in small commercial plants both in the United States and in Europe. These technologies include, but are not limited to, the following: (1) a laboratory plant at the Bartlesville Energy Research Center of the Department of Energy, (2) the Rhone-Poulenc plant in France, and (3) the Phillips Petroleum Company process. All of these plants, as well as other new technologies, appear to have higher yields of clean oil and do not generate as much waste as the existing acid/clay process used almost universally around the world today.

Since the EPA's 1974 report to the Congress was completed, the EPA has continued to study the issue of waste oil resource recovery, particularly the environmental, energy, and resource conservation aspects of the problem. A recent EPA study showed that as much as 50,000 barrels of oil per day could be saved if waste oil were re-refined to its original use as a lubricant and approximately 25,000 barrels per day could be saved if the waste oil were reprocessed and burned as a fuel.

As a result of this continued work we have found that many countries in the developed world and many States now have more active programs to promote the recycling of used oil than those of the Federal Government. We have also continued to find that present methods for managing used oils in the United States are unsatisfactory from an environmental and resource conservation viewpoint. The present methods do not reflect the inherent economic advantages that could be derived from proper resource recovery on a national scale.

Before proceeding to a discussion of the EPA's program related to the recycling of waste oil, it seems useful to review an impediment to recycling which Federal regulatory authority does not affect. This impediment is the tax advantage which virgin oil enjoys over recycled oil. The tax law changes in the mid-1960's had a discouraging effect on resource recovery of lubricating oils. Before 1965, re-refining lubricating oil had a tax advantage which frequently made it more attractive as a substitute for virgin oil. This resource recovery incentive was particularly important in sales of re-refined oil to large users of industrial lubricants.

The Federal Excise Tax Reduction Act of 1965 and a subsequent Treasury Department ruling rescind most of this tax advantage by creating a competitive tax exemption for virgin oil if bought by off-highway (e.g., industrial) users. Specifically, it provides for a Treasury refund of $0.06 per gallon to the ultimate purchaser of virgin oil carrying the $0.06 excise tax. In effect, the payment is a tax refund to off-highway users. In contrast, the act maintains the tax on lube oil used for automotive purposes and dedicates its revenues to the highway trust fund. Thus, virgin lube oil used for off-highway use now has the same tax-exempt status previously enjoyed only by recycled oil.

The Treasury Department ruling also concluded that re-refiners do not qualify as off-highway users and therefore are ineligible for a tax rebate on the virgin oil they purchase for blending. Therefore, the oil re-refining industry not only lost its tax advantage, but also gained a tax disincentive because all opportunities to receive an excise tax refund were lost.

Since 1974, the Congress has passed and the President has signed two important pieces of legislation that will affect waste oil recycling in the United States by providing the EPA and other agencies of the government with important mandates in the area of used oil re-refining.

The first is the Energy Policy and Conservation Act, Public Law 94-163, which requires the EPA to promulgate labeling standards applicable to containers of new, used, and recycled oil to reduce to the maximum extent practicable environmental hazards and wasteful practices associated with the disposal of such oils after use. This act also requires the Federal Trade Commission (FTC) to promulgate labeling standards based on the quality of oil. Presently, the EPA is planning to promulgate one labeling standard for lubricating oil jointly with the FTC. As you know, one of the major impediments to the re-refining of used oils has been the question of relative performance quality of re-refined and virgin lubricating oils, and it will be our intention in this joint activity with the FTC to provide consumer protection as well as environmental protection and resource conservation. At this point I would like to add that the EPA has financially supported an analysis of the quality of re-refined lubricating oil. The results of this analysis and comparative testing with virgin lubricating oils have shown that re-refined oils can be as good as or better than virgin oil as an automotive lubricant.

The second important piece of legislation affecting waste oil recycling in the United States is the Resource Conservation and Recovery Act (RCRA), Public Law 94-580. The RCRA requires the EPA to promulgate standards and regulations for hazardous waste management and land disposal.

Under the authorities of subtitle C of the RCRA, the EPA is required to develop and promulgate regulations that will establish criteria for what is a hazardous waste; will require generators and transporters of hazardous wastes to report on the character of wastes generated and the manner in which they are transported from the point of generation to the next step in their management; and will develop and promulgate regulations for the storage, treatment, and disposal of hazardous wastes. Subtitle D requires the EPA to issue criteria for acceptable land disposal of all those wastes that do not fall within the control of subtitle C. Hence, whether waste oils are hazardous or nonhazardous, these regulations, when promulgated sometime in 1978, may not only impact on generators of waste oils, but also on all who have any role in the management of this vital resource, including its ultimate disposal on the land.

There is one additional regulatory authority the EPA has which may affect the waste oil recycling industry. Next year the EPA will promulgate ambient air standards for lead. These standards will be required for use by the States in the mid-1980's to develop State air pollution control plans. Because lead is one of the major contaminants in waste oil and because the burning of raw waste oil is the largest end use of this product, we project a possible decrease in the amount of raw waste oil burned in certain localities to comply with these ambient standards.

In conclusion, we believe the EPA's regulations, particularly the hazardous waste and land disposal provisions of the RCRA, will provide new opportunities for the oil industry, resulting in the recovery and reuse of more waste oil in the United States, and may stimulate new technologies to meet these opportunities.

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THE ROLE OF THE FTC IN RECYCLED OIL LABELING

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I would like to take this opportunity to state briefly what I see as the role of the Federal Trade Commission (FTC) in recycled oil labeling. I am sure you realize that staff remarks such as mine are not necessarily binding on the Commission.

As you are aware, section 383 of the Energy Policy and Conservation Act directs the National Bureau of Standards (NBS) to develop test procedures for determining the substantial equivalency of recycled oil to new oil. When these data are compiled by the NBS and are submitted to the FTC, we are given 90 days within which to prescribe by rule (1) test procedures for the determination of substantial equivalency of recycled oil with new oil for a particular end use, and (2) labeling standards applicable to containers of recycled oil.

The act itself raises certain unanswered questions. For example, the act is silent as to the establishment of standard specifications and classifications for the various types of oils. In our discussions with Mr. Becker and his staff, we have agreed that is is important for specifications to be developed in order that the Commission may require proper labeling for recycled oil as to a particular end use. In this connection, we are hopeful that the NBS, working with the American Society for Testing and Materials specifications committee and the Federal specifications people, will be successful in developing standard specifications and classifications for recycled oil so as to enable the Commission to more effectively deal with the labeling problem.

The act also is silent as to whether recycled oil, which meets the substantial equivalency criterion, should be labeled "recycled" or whether no disclosure should be required. This is a problem we hope to resolve when we issue our proposed rule for comment.

We understand from Mr. Becker that his staff expects to have the necessary test procedures and data for fuel oils ready for submission to the Commission by the spring of 1978. A threshold problem of labeling immediately emerges. How does one label fuel oil which is not sold in the customary retail container, but rather is delivered to the consumer in bulk? At this point, we believe that this potential problem can be solved by requiring labeling information on the invoice at the point of sale and in bids prior to sale.

We intend to work closely with the National Bureau of Standards and the Environmental Protection Agency (EPA) in carrying out our responsibilities under the act. We must, of necessity, rely heavily on the NBS and the EPA for their scientific and technical expertise.

I would also like to state that we have had a hard and tough road with the Commission's used oil rule. While we recognize that this rule may have tended to discourage the recycling of waste oil, in the absence of trustworthy scientifically based data as to the quality and performance of recycled oil we have had no viable alternative but to require that recycled oil be labeled "previously used." We will welcome test data and procedures from the NBS which will demonstrate that recycled oil can be equated with new oil from the standpoint of both quality and performance. Armed with such data, recycled oil should, indeed, gain a positive image which should result in an increase in the demand for such a product. This, in turn, should encourage the recycling of waste oil.

Finally, when we reach the enforcement stages of our program, we will rely heavily on Mr. Becker's staff for the necessary technical expertise in resolving any issues which have to do with specifications, testing procedures, and other data.

I have intentionally kept this presentation on a general plane because our actions in this matter will depend, in large measure, upon both the data which the NBS furnishes and the comments we receive pursuant to our notice of proposed rulemaking. We encourage your participation in both the informal preliminary stages of our program and any formal hearings which may be scheduled.

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USED OIL RECYCLING PROGRAM ISSUES: THE DEPARTMENT OF ENERGY

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Among the objectives of the former Federal Energy Administration's (FEA) Office of Energy Conservation and Environment were: (1) to reduce the energy demand growth rate in the United States, (2) to develop and implement programs that increase the efficient use of energy; and (3) to ensure that environmental concerns are balanced with national energy goals.

We therefore sought to enlist participation by broad segments of the Nation's communities in programs that would effectively encourage

- the efficient use of energy,
- energy conservation activities, and
- energy usage in an environmentally satisfactory manner.

As used oil recovery was deemed an important opportunity for energy conservation, implementation of a program to increase such recovery was developed. The Department of Energy's Office of Conservation and Solar Application's (as it is currently named) Used Oil Program is designed to increase the collection and utilization of used oil through State and local efforts, to create useful market demand for used oil in areas of the country where there has been little demand, to assist in identifying issues relevant to the re-refining industry, and to increase awareness of the potential for recycling used oil through publicity and industrial education programs.

From its very onset--which for all intents and purposes, was back in June 1975 when it was nationally endorsed and released by the FEA administrator--the used oil recycling program possessed tremendous potential and was highly considered as an idea whose time had come. (Of course, for many renewable energy resource advocates it was well overdue.)

The program is national in scope in terms of serving the Nation's physical and psychological aims of energy conservation:

- physical by virtue of the creation and/or maintenance of an administrative/management mechanism to affect the maximum recovery/reuse of used oil produced in the United States, and

- psychological in that it contributes significantly to the maintenance and expansion of the national energy/environment conservation/preservation citizen ethic as a characteristic of our national lifestyle.

Orchestration of action is the essential key to accomplishment of these objectives.

I will now cite specific Department of Energy Used Oil Recycling Program planned actions.

- Continue/increase catalytic action to centrally acquire technical assistance information concerning recycling programs and projects across the Nation. Process this information and share it with appropriate contacts in the recycling network, including State and local governments, oil companies, business, industry, national citizen organizations, other Federal agencies, public interest groups, and citizens in general.

- Continue to inspire and promote contact within the three key elements of the recycling equation: governments, companies, and citizens. Promote creation of recycling projects and the publicizing thereof on a nationwide basis.

- Continue service and support contact with State energy offices to assure optimum consideration of used oil recycling as one of the prime voluntary programs if (as) indicated in the State energy conservation plan.

- Reinforce the potential for additional recycling funding to State governments.

- Continue cooperation with other Federal agencies interested in or affected by used oil recycling.

- Provide assistance and information to State and local governments in the adoption of legislation governing the controlled recovery/reuse of used oil based on the concepts contained in the Model Used Oil Recycling Act (distributed and prepared by the FEA).

- Encourage nationwide specific sponsorship of national used oil recycling programs by nationally organized citizen organizations such as the U.S. Chamber of Commerce, the U.S. Jaycees, the League of Women Voters, and the American Association of University Women, to name just a few.

- Coordinate support services by DOE field and regional offices to State and local elements of the recycling spectrum.

- Continue to encourage and respond to congressional interest in national used oil recycling issues, including matters of excise tax equalization and constituents' concerns relative to recycling.

- Continue contact and cooperation with national associations active in the recycling spectrum, i.e., the American Petroleum Institute, the Association of Petroleum Re-refiners, the National Petroleum Council, and the American Society for Testing and Materials.

- Monitor State and local activities aimed at used oil reclamation, reprocessing, and reuse.

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UTILIZATION OF USED OIL: ENERGY CONSERVATION ASPECTS

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A year ago I had the opportunity to speak before you at the first Conference on Measurements and Standards for Recycled Oil. At that time, we were into the initial phase of a study on recycling lube oil, and I was only able to give you the outline and objectives of the study. This time I would like to present some results obtained from the study relative to energy conservation aspects of re-refining used oil, which is the main thrust of the study.

This presentation is based on a study by The Aerospace Corporation, initiated under the auspices of the Energy Research and Development Administration and carried over to the newly formed Department of Energy (DOE). The study is being performed for the Industrial Energy Conservation Division under the guidance of Dr. Jerome Collins, although it was initiated for the Non-Highway Energy Conservation Division under Mr. Eugene Eckland. The study was started in late September 1976, and a draft of the final report was submitted to the DOE for review and comments. Final review comments were received a few weeks ago, and we are in the process of revising the report to reflect these comments. The scheduled publication date is January 1978. As I stated in my introductory remarks, the main objective of the study is to assess the energy conservation aspects of re-refining used oil. The analysis makes use of two scenarios to bring out the full potential of re-refining, i.e., all used oil goes either to re-refining or to burning. For each case, the alternative lube or fuel requirements are satisfied from virgin sources.

There are five parameters to be considered in the analyses, of which three are related to lube oil processing and two are associated with used oil burning. The parameters related to lube oil manufacture are process energy to produce virgin and re-refined oils and yield of the re-refining process. Parameters affecting burning are process energy and yield of the pretreatment process utilized prior to burning. These two aspects are treated independently for convenience and also because the need (or requirement) for pretreating used oil prior to burning has not been established.

An important parameter in assessing energy conservation aspects of re-refining used oil is the amount of energy required to produce the alternate product: virgin lube oil. To establish this baseline, we have defined a typical refining process that consists of the steps shown in table 1 and results in a process energy requirement of 2.16 million British thermal units (Btu's) per barrel, or 51,400 Btu's per gallon. This value differs from those calculated or reported by others and is discussed in the final report. For simplicity and clarity, those other values will not be considered in this presentation.

For this presentation, we consider two re-refining processes. The first is a typical acid-clay process, and the second is an example of an advanced process--that which may be obtained through solvent extraction. Note that the solvent extraction process has a higher energy requirement, as well as a higher yield. Also note that both processes include a fractionation step to produce a range of viscosities, even though this is not a common practice today with the acid-clay process.

The two scenarios considered in the study, either re-refining or burning of all used oil, and the separate analysis of pretreatment requirements prior to burning can be reduced to equation format (table 2). The first equation shows predicted energy savings due to re-refining in lieu of burning when used oil is burned without pretreatment. The second equation shows incremental energy savings due to re-refining if pretreatment is required.

Table 1. Lube Oil Production Processes

VIRGIN LUBE OIL

- Process steps
  - -- Vacuum distillation
  - -- Propane deasphalting
  - -- Solvent refining using furfural
  - -- Solvent dewaxing using MEK
  - -- Hydrotreat finish
- Process energy: 2.16×10^6 Btu's per barrel product

RE-REFINED LUBE OIL

- Current primary process: acid-clay
  - -- Acid treatment
  - -- Clay treatment
  - -- Fractionation
  - -- Process energy: 0.34×10^6 Btu's per barrel feedstock
  - -- Yield: 65 percent (based on feedstock containing 7 percent water and 4.2 percent light ends)
- Advanced potential process: solvent extraction
  - -- Solvent extraction
  - -- Fractionation
  - -- Clay treatment
- Process energy: 0.62×10^6 Btu's per barrel feedstock
- Yield: 76 percent (based on feedstock containing 7 percent water and 4.2 percent light ends)

Table 2. Analytical Relationships for Energy Savings

Energy conserved due to re-refining in lieu of burning used oil without pretreatment.

- Energy saved = $-E_{R/R} + (Y \times (E_R + E_C)) - E_{UO} + (O_H \times E_{OH})$
- Where:

$E_{R/R}$ = Re-refining process energy for used oil, Btu per gallon feedstock.

Y = Re-refinery yield, gallon product per gallon feedstock.

E_R = Refinery process energy for virgin lube oil, Btu per gallon product.

E_C = Heat content of virgin crude, Btu per gallon.

E_{UO} = Heat content of used oil feedstock, Btu per gallon.

O_H = Overhead distillate yield from used oil, gallon per gallon feedstock.

E_{OH} = Heat content of overhead distillates, Btu per gallon.

Additional energy saved due to re-refining when pretreating used oil prior to burning is required.

- Additional energy saved = $E_{UO} - (O_H \times E_{OH}) - (Y_{P/T} \times E_{P/TO}) = E_{P/T}$
- Where:

E_{UO} = Heat content of used oil feedstock, Btu per gallon feedstock.

O_H = Overhead distillate yield from used oil, gallon per gallon feedstock.

E_{OH} = Heat content of overhead distillates, Btu per gallon.

$Y_{P/T}$ = Pretreatment yield of fuel from used oil, gallon product per gallon feedstock.

$E_{P/TO}$ = Heat content of pretreated used oil, Btu per gallon.

$E_{P/T}$ = Pretreating process energy, Btu per gallon feedstock.

Figure 1 shows a parametric presentation of the equation in table 2 for energy savings due to re-refining, where the variables shown are for refining and re-refining process energy. The third parameter of interest, process yield, is fixed at 65 percent, which is typical of the acid-clay process. Of course, there are other fixed parameters built into the graph, such as the heat content of the various petroleum products and the feedstock composition, but these are ancilliary to the analysis and are, in effect, assumed to be constants. Shown on the graph is the solution for the acid clay process, which results in a net energy loss due to re-refining. That is, with the acid-clay process it would be advantageous, from an energy standpoint, to burn used oil. But remember, this case assumes no pretreatment prior to burning. The energy loss is 3,700 Btu's per gallon of feedstock processed. This is equivalent to 890,000 barrels per year if all used oil generated were re-refined, which was about 1.4 billion gallons in 1975. It should be noted that, if energy requirements for producing virgin lube oil were higher than shown, we would get into a situation where the acid-clay process would save energy.

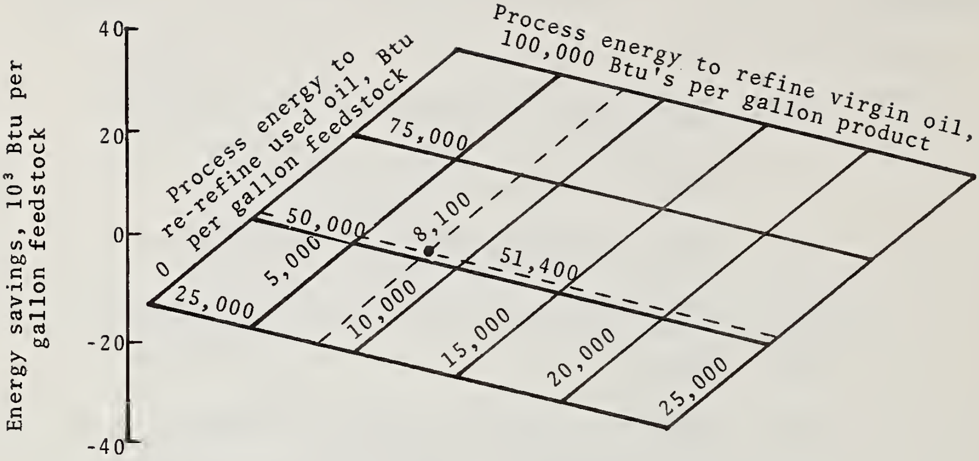

Figure 1. Analytical results--acid-clay process.

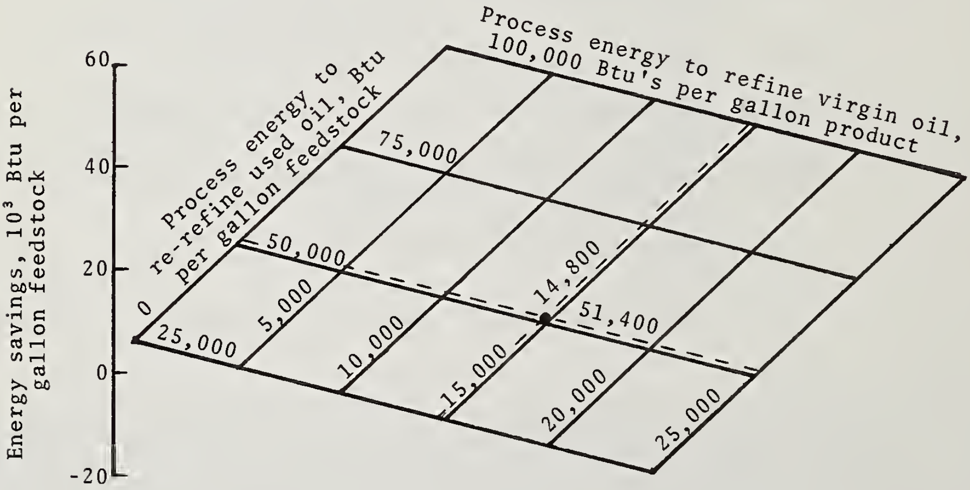

Figure 2. Analytical results--advanced process.

Figure 2 is similar to figure 1, except that the yield is now 76 percent, which is potentially obtainable by advanced re-refining processes such as solvent extraction. The example shown indicates energy savings of 11,900 Btu's per gallon of feedstock, or a potential of 2,820,000 barrels of petroleum per year. Although both this figure and the previous one show parametric data for fixed yields, it is possible to include yield as a variable in the graphical solution.

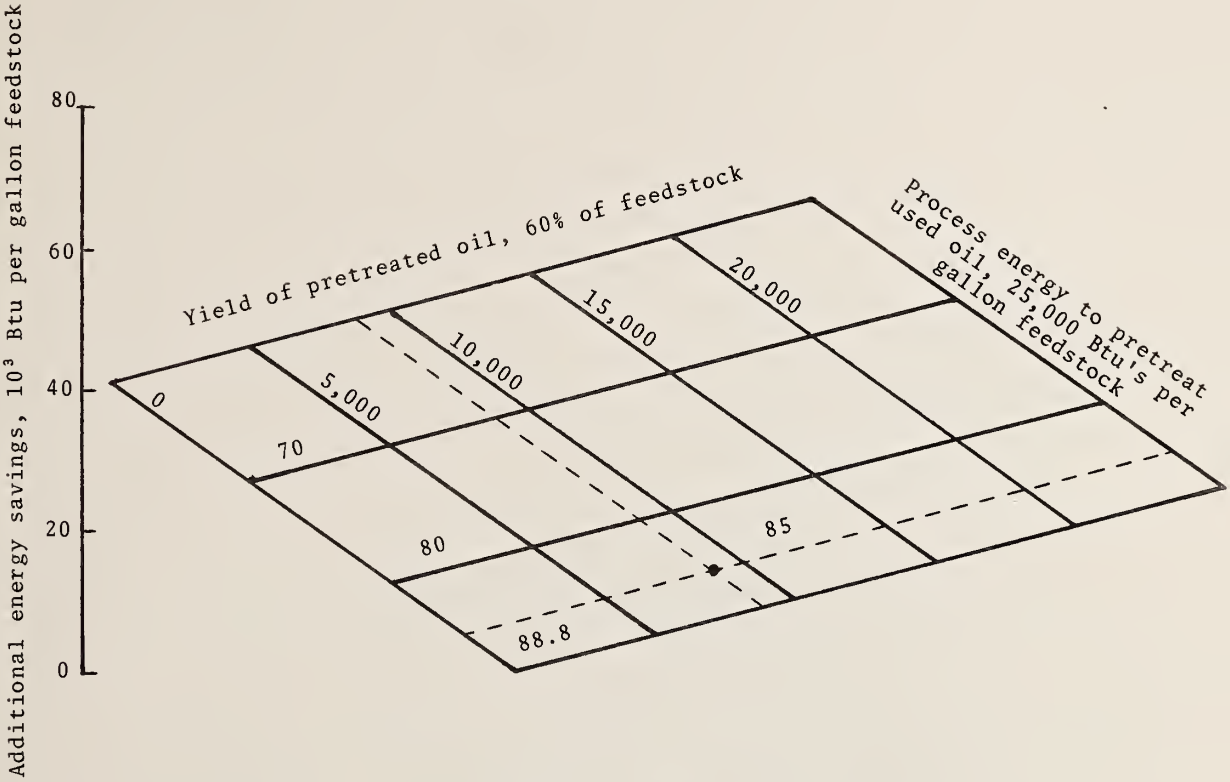

Figure 3. Additional energy savings due to re-refining.

Figure 3 illustrates the incremental savings added to that due to re-refining when used oil is pretreated prior to burning. As you will note, any pretreatment of used oil prior to burning enhances the energy savings due to re-refining. The zero point shown corresponds to no pretreatment. That is, process energy requirements are zero and the entire petroleum content of the used oil, which in this case is 88.8 percent (based on 4.2 percent overheads and 7 percent water), is recovered. An example is shown for what may be considered a high level of pretreatment (one that might produce a used oil product comparable to a virgin fuel). In this case, the incremental energy savings due to re-refining are 14,200 Btu's per gallon, or equivalent to 3,370,000 barrels per year. It should be noted that the savings are greater than that previously shown for an advanced process (11,900 Btu's per gallon). This analysis shows that the energy savings potential of re-refining is highly dependent on requirements for pretreatment prior to burning used oil.

To summarize the results of the two processes with and without pretreatment prior to burning, the acid-clay process results in a loss in energy of about 3,700 Btu's per gallon feedstock, while energy savings of 10,500 Btu's per gallon are realized if a high level of pretreatment is required prior to burning.

For the advanced process, energy savings are further enhanced with a pretreatment requirement, going up to 26,000 Btu's per gallon, or over 6 million barrels of petroleum per year, which represents a significant amount of oil.

SESSION V. EVALUATION OF RE-REFINED ENGINE OILS

National Bureau of Standards Special Publication 556. Proceedings of a Workshop on Measurements and Standards for Recycled Oil - II held at NBS, Gaithersburg, Maryland, November 29 and 30, 1977. (Issued September 1979)

THE EPA/DOD INVESTIGATION OF RE-REFINED ENGINE OIL

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In 1975, the Resource Recovery Division of the Environmental Protection Agency (EPA) became interested in a coordinated program to define the feasibility of using re-refined basestocks in MIL-L-46152 type lubricants, i.e., oils meeting performance classifications SE and CC. This interest lead to an interagency agreement with the Army, covering the program which will be discussed here today. The program has two objectives: (1) to demonstrate the performance capabilities of re-refined engine oil, and (2) to generate technical data which will increase the understanding of re-refined lubricants.

Accomplishment of these objectives has been approached in the following manner. First, re-refined stocks were obtained and analyzed using standard laboratory techniques. Based on the analyses, six stocks were selected, and each was formulated with additive to provide a finished engine oil. The finished lubricants were then subjected to the engine dynamometer tests used to define oil performance under the MIL-L-46152 specification. In addition to this effort, the field performance of a re-refined product was evaluated through engine teardown inspections.

This has given an overall look at the scope of the program. Now let us discuss each portion in a little more detail. The first task was obtaining re-refined basestocks. This was done through the Association of Petroleum Re-refiners, who contacted some 36 member and nonmember companies requesting assistance by providing production samples of their re-refined stocks. Of these re-refiners, 17 participated in the program, supplying some 19 samples of re-refined stocks or a finished product. After obtaining the samples, analyses were performed to determine physical and chemical characteristics of the stocks and to get a feel for the level of re-refining employed. With the assistance of the American Society for Testing and Materials B-I task force on re-refined oils, six of the stocks were selected for use in the engine test portion of the program. Rather than discuss all analyses data, I will limit my comments to these stocks.

As can be seen in table 1, the six stocks were similar in physical properties. From a viscosity standpoint, the stocks would be classified as SAE (Society of Automotive Engineers) 20W-20 oils and would be generally comparable with 250 to 350 neutral, high viscosity index virgin basestocks. Two dissimilarities, the low flash point of stock 6694 and the low pour point value of stock 6692, were noted and believed to be related to the level of re-refining used. Although similar in physical properties, the six stocks did show significant variation in their chemical makeup. As can be seen, there was a fair range between stocks in total acid number; two stocks (6690 and 6692) contain high sulfur levels, and significant difference in trace element content can be observed. The higher level of trace elements in sample 6692 tends to confirm the comment previously made concerning the low pour point observed for this stock. Also, we can see some variation in the boiling point distribution and characteristic group analyses. Most significant is the lower 1- and 5-percent point values for sample 6694 and the higher saturate/lower aromatic contents shown for samples 6694 and 6696. In the case of the low 1- and 5-percent points, these values indicate that some of the light ends may not have been removed in the re-refining process and tend to confirm the comment made concerning the low flash point of this sample. One last point that should be made is that data for several of the re-refined stocks were very similar to the values obtained for the virgin stock. In other words, it would not be possible using these data to distinguish several of the re-refined stocks from a virgin stock.

The second task was formulation of the selected stocks into finished engine lubricants. The major additive suppliers were contacted and asked for an additive system recommendation to meet MIL-L-46152 requirements. It was desired, but not required, that a single additive package be used for all six stocks. Also, the manufacturers were asked when making a recommendation to consider both the performance

Table 1. Summary of Analyses

Property	6690	6692	6694	6696	6697	6698
Kinematic viscosity						
cSt @ 210° F	8.02	8.81	7.86	8.54	9.23	6.84
cSt @ 100° F	64.7	70.9	59.0	67.1	76.9	50.6
Viscosity index	98	104	108	105	103	98
Gravity, °API	29.1	29.4	30.0	29.6	29.5	29.8
Flash point, °F	425	440	385	415	435	440
Pour point, °F	5	-25	15	10	10	15
Total acid number	0.16	0.11	0.05	0.10	0.08	0.04
Sulfur, %	0.31	0.34	0.14	0.12	0.15	0.16
Oxygen, %	0.70	0.76	0.65	0.41	0.55	0.59
Elemental, ppm						
Calcium	<5	111	<5	<5	44	<5
Zinc	<2	11	<2	<2	<2	<2
Magnesium	<1	3	<1	<1	2	<1
Lead	<1	47	<1	<1	<1	<1
Iron	<2	13	<2	<2	<2	<2
Total ash, %	<0.001	0.036	<0.001	0.002	0.009	0.001
Boiling point distribution, °C						
1%	346	298	240	276	314	337
5%	377	366	339	371	383	380
10%	394	390	377	390	400	395
50%	448	455	452	447	452	450
90%	540	537	551	550	532	518
Characteristic groups, %						
Saturates	77.3	71.2	84.4	80.7	74.9	75.1
Polar	2.6	3.0	2.6	2.7	2.5	2.5
Aromatic	20.1	25.8	12.9	16.6	22.6	22.4

and economics of the finished products. Responses from the additive manufacturers can be summarized as follows:

1. All manufacturers recommended a package currently used in qualified MIL-L-46152 oils.

2. With one exception, the additive system was recommended for use in all stocks at the same treatment level. One manufacturer, however, did recommend supplementing the basic system with additional inhibitors for three of the stocks.

3. Except where additional inhibitors were recommended, the treating cost for the re-refined oils was the same as for virgin-based products.

The additive recommendations were reviewed with the American Society for Testing and Materials (ASTM) task force, and one recommendation involving a single system for use in all stocks was selected.

Analyses conducted on the finished oils are shown in table 2. Four of the oils fall into a SAE 20W-20 classification, and two would be classified as a 20W-30. The additive concentration, indicated by the calcium, zinc, and phosphorus analyses, was consistent for all products.

Table 2. Finished Oil Analyses

Property	6690	6692	6694	6696	6697	6698
Kinematic viscosity						
cSt @ 210° F	9.3	10.2	9.2	9.8	10.6	8.2
cSt @ 100° F	79.1	86.3	72.5	80.8	92.6	62.7
cSt @ 0° F (ext.)	7084	7125	5136	6321	8159	4364
Viscosity index	103	107	111	109	106	107
Elemental, %[a]						
Calcium	0.22	0.22	0.21	0.22	0.22	0.22
Zinc	0.14	0.14	0.13	0.14	0.14	0.14
Phosphorus	0.12	0.12	0.12	0.12	0.12	0.12
Sulfated ash, %	0.95	0.98	0.95	0.97	0.96	0.96

[a] Analyses supplied by additive manufacturer.

The final task in this portion of the program involved the engine performance testing of the finished re-refined lubricants. Before reviewing these tests, let me say that the data being presented are single test results. The first results shown are the L-38 test (table 3). As can be seen, all oils performed satisfactorily in this test. The second set of data is Sequence IIC test results (table 4). Using reference oil test data, the reported results have been adjusted for severity trends within the testing laboratory. The adjustment value is shown at the bottom. Again, the data indicate the oils satisfactorily met specification requirements. The next results (table 5) are from the Sequence IIIC testing. Here the oils met the requirements in all areas except piston varnish. One oil (6692) failed to meet specification requirements, and three of the other oils were either at or just above the minimum acceptable value. Moving on to the Sequence VC results (table 6), we find a potential problem in the area of varnish deposits. Many of the varnish results are at or just above the minimum MIL-L-46152 level, and one oil (6690) failed to meet the average engine varnish requirement. In other areas--sludging, clogging, and ring sticking--the data show all oils to have performed in a satisfactory manner. Again, adjusted

Table 3. Summary of L-38 Performance Tests

	46152	6690	6692	6694	6696	6697	6698
Bearing weight loss, mg.	40 Max	28	22	15	5	8	21
Piston varnish	---	9.7	9.7	9.7	9.7	9.7	9.7

Table 4. Summary of Sequence IIC Performance Tests

	46152	6690	6692	6694	6696	6697	6698
Average rust	8.4 Min	8.6	8.8	8.8	8.9	8.6	8.8
Stuck lifters	None	None	None	None	None	None	None
Adjusted average rust		8.4	8.7	8.7	8.9	8.5	8.7

Table 5. Summary of Sequence IIIC Performance Tests

	46152	6690	6692	6694	6696	6697	6698
% Viscosity increase @ 40 hours	400 Max	105	111	126	88	69	62
Sludge	9.2 Min	9.8	9.8	9.6	9.7	9.7	9.7
Piston varnish	9.3 Min	9.3	9.2	9.5	9.4	9.5	9.4
Ring land deposits	6.0 Min	8.0	8.1	7.6	8.3	8.0	8.1
Ring sticking	None	None	None	None	None	None	None
Wear (cam and lifter)							
Scuffing	None	None	None	None	None	None	None
Avg., in. x 10^{-4}	10	2	2	3	4	3	3
Max., in. x 10^{-4}	20	6	6	6	7	6	5

Table 6. Summary of Sequence VC Performance Tests

	46152	6690	6692	6694	6696	6697	6698
Piston varnish	7.9 Min	7.9	8.0	7.9	8.2	8.2	7.9
Avg. engine varnish	8.0 Min	7.8	8.1	8.0	8.2	8.4	8.3
Avg. engine sludge[a]	8.7 Min (8.5) Min	9.5	(9.4)	9.5	9.5	9.5	(9.5)
Screen clogging, %	5 Max	0	0	0	0	0	0
Oil ring clogging, %	5 Max	0	0	0	0	0	0
Compression ring sticking	None	None	None	None	None	None	None
Adjusted piston varnish		7.9	8.0	7.9	8.2	8.1	7.9
Adjusted avg. varnish		7.9	8.3	8.2	8.3	8.5	8.4
Adjusted avg. sludge		9.6	9.6	9.6	9.6	9.6	9.6

[a] Bracketed value: Manual No. 10 rating.

Table 7. Summary of 1-H2 Performance Tests

	46152	6690	6692	6694	6696	6697	6698
Top groove filling, %	45 Max	9	1	24	2	12	10
Weighted total demerit	140 Max	80	74	176	68	188	155

results are shown at the bottom. The last results shown (table 7) are the 1-H2 diesel tests. All oils show acceptable performance in controlling top groove deposits. However, lacquer deposits resulted in three of the six oils (6694, 6697, and 6698) failing to meet the specification requirements.

In addition to the work just discussed, a portion of the program called for the field evaluation of a re-refined oil. Specifically, the evaluation involved the teardown inspection of engines from six vehicles operated by the city of San Diego, California, where re-refined oil had been used since February 1974. To complement the San Diego evaluations, engines from two vehicles using virgin-based engine oil and operated by the city of Hollywood Park, Texas, were also inspected. The data shown in table 8 summarize the vehicle fleets and operation. There are several differences (air conditioning, mileage and oil-change intervals) between the two groups of vehicles. These differences, in addition to the lubricants, likely influenced the deposit levels observed during the teardown inspections and are shown here.

Table 8. Description of Vehicle Fleets and Operation

	Re-refined oil	Virgin oil
Location	San Diego, CA	Hollywood Park, TX
Vehicles		
Make	1974 Ford Torino	1974 Ford Torino
Model	4-door sedan	4-door sedan
Engine	351 CID	351 CID
Transmission	Automatic	Automatic
Air conditioning	No	Yes
Operation		
Type service	Police cruiser	Police Cruiser
Avg. odometer miles	97,000	68,700
Oil/filter change, miles	4,000	6,000

To briefly summarize these data (table 9), the San Diego vehicle had moderate engine varnish, light sludge, and very light rust, compared to the heavy engine varnish and moderate-to-light sludge and rust deposits observed in the Hollywood Park vehicles.

Table 9. Summary of Engine Deposit Ratings

			Deposit level, merits[a]			
			Varnish		Sludge	Rust
Location	Number of units	Odometer miles, avg.	Piston	Engine	engine	Lifter
San Diego, CA	6	96,966	6.2	6.5	9.3	9.8
Hollywood Park, TX	2	68,668	6.3	4.4	8.6	8.9

[a] CRC merit rating scale (10 = clean).

Since there are some tests still in progress and there has been no real opportunity to make a detailed evaluation of the data generated during the program, I feel it is not proper at this time to attempt to draw firm conclusions. Therefore, I will conclude the presentation with a couple of general comments. From the positive side, the performance tests are most encouraging when compared to previous re-refined oil data generated by the Army. At the same time, these data have a negative aspect in that only one of the six oils appears to have met the desired performance level. In the case of the laboratory analyses, several of the re-refined stocks appeared equal to virgin basestocks. Again, this could be considered a positive factor in that it may indicate an acceptable level of re-refining. However, from the negative side, these data appear to provide no easy means for monitoring the consistency of the stocks in regard to engine performance.

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IOWA RE-REFINED OIL FLEET TEST

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Introduction

In 1975 the Iowa Department of Transportation (DOT) became interested in using re-refined oil in State vehicles. In order to properly assess the performance of re-refined oil, a road test, operating under normal fleet conditions, seemed desirable. The Bartlesville Energy Research Center (BERC), Department of Energy, was also interested in road tests of the oil from their recently developed process (1).[1] An agreement among the Iowa State University Engineering Research Institute (ISU), the Iowa State DOT, and the BERC was reached to test SE-CC service (10W30) oil from Motor Oils Refining Company (MORCO) and BERC on about 20 vehicles with a comparable number of vehicles operating on the normal DOT virgin stock under similar conditions.

In selecting the vehicles for the test, the primary consideration was that they operate in a service where they would accumulate sufficient mileage to allow meaningful comparisons in 2 years. The limited number of vehicles available of any one type required the use of station wagons, pickups, and trucks. One-year-old vehicles were also included to determine any possible effect from a change in oil. The engines were not premeasured because the cost of measuring all engines was prohibitive.

At the end of the 2-year period, 12 engines will be selected from those that complete the test, and deposit and wear ratings will be performed. Southwest Research Institute will assist in selecting the 12 engines and in performing the ratings. The cumulative oil analyses and the deposit and wear ratings are expected to provide, on the average, an adequate comparison of the oils. A lack of significant differences among the three oils would be a useful result and would permit the DOT and other State agencies to allow re-refined oil producers to bid for the State's business. It may also encourage environmentally superior use of waste oil in Iowa. A closed-loop recycle system might be the optimum result.

An evaluation of the potential for recycling oil in Iowa is another objective of this test. The cost of collecting and recycling oil from State fleets, urban areas, and rural areas is to be assessed along with the attitude of Iowans in general toward using re-refined oil. A survey of Iowa farmers has already been concluded.

Description of the Test Oil

The virgin oil was purchased on a bid basis from the Warren Oil Company to meet the performance requirements of an oil specified by the military specification MIL-L-46152 (i.e., American Petroleum Institute designation SE-CC service, 10W30). The MORCO re-refined oil was derived from the acid/clay process, while an experimental BERC solvent process produced the BERC re-refined oil. These oils were also formulated to SE-CC service (10W30).

Table 1 presents a detailed laboratory analysis, and Table 2 provides specific information regarding the various additive packages that were used to formulate each oil.

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[1] Underlined numbers in brackets indicate the literature references at the end of this paper.

Table 1. Test oil laboratory analysis.

Laboratory analyses	MORCO oil	BERC oil	Virgin oil
Viscosity, SSU @ 100° F	316	375	318
Viscosity, SSU @ 210° F	61.60	67.85	63.72
Viscosity index	136	137	141
Total acid number	2.15	2.35	1.63
Total base number	5.43	3.92	3.33
Ash (sulfated) (%)	1.00	1.30	.71
pH		7.4	
Benzene insolubles coagulated	Nil	Nil	Nil
Pentane insolubles coagulated	Nil	Nil	Nil
Sulfur (%)	.58	.5	.46
Flash, °F	405	425	405
Pour, °F	Below -50	Below -50	Below -50
Gravity	29.2	29.2	29.9
Color	4-½	3	3
Water	--	--	--
Fuel dilution	--	--	--
Spectrographic:ppm			
Iron	2	2	2
Zinc	980	830	820
Lead	0	0	0
Sodium	5	4	16
Calcium	1950	2600	1900
Silicon	4	2	5
Silver	0	0	0
Copper	0	0	0
Aluminum	0	0	0
Barium	24	25	35
Nickel	0	0	0
Chromium	0	0	0
Tin	1	1	1
Phosphorus	800	650	650
Boron	0	0	14
Magnesium	650	9	7
Vanadium	0	0	0
Molybdenum	0	0	0
Manganese	0	0	0
Cadmium	0	0	0
Titanium	0	0	0
3 hours @ 210° F copper corrosion	Class I	Class I	Class I

Table 2. Test oil additive packages.

Test oil	Viscosity index improver	Multipurpose additive
BERC oil	Lubrizol 3702 (Dispersant type) 2-10% (Vol.)	Lubrizol 4462 6.8% (Vol.) of basestock
MORCO oil	Lubrizol 3135 (Nondispersant polymethacrylate)	Hitec E-703 6.7% (Vol.) of basestock
Virgin oil[a]	Lubrizol 7013 (Poly Ethylene-Propylene) 8% (Vol.)	Lubrizol 4454-E 6.7% (Vol.) of basestock

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[a] i.e., Mid-continent crude.
= 70.8% high V.I. solvent extracted 100 neutral base.
= 14.8% 150 SUS @ 210° F of bright stock.

Test Design

The DOT test fleet (table 3) consists of 12 1976 Dodge pickups, 12 1976 Matador wagons, 10 1975 Chevy Malibus, and 10 1976 Ford 750 trucks with hydraulic systems. The vehicles are paired to provide maximum comparison of oils. New vehicles were operated 1,000 to 3,000 miles on "original equipment" oil before being switched to the test oil. The used vehicles (Chevy Malibus) had accumulated 13,000 to 38,000 miles before being switched to the test oil.

Table 3. Test fleet vehicle distribution.

Vehicle type	Engine size (in.^{3})	Fuel type	Oil drain interval	No. of BERC vehicles	No. of MORCO vehicles	No. of virgin vehicles
1976 Matador Wagons	360	Unleaded	8,000	2	5	5
1976 Dodge Pickups	225	Unleaded	8,000	---	4	3
	225	Unleaded	10,000	---	3	1
	360	Unleaded	10,000	---	---	2
1975 Chevy Malibus	350	Unleaded	4,000	---	5	5
1976 Ford 750 Trucks	361	Leaded	4,000	---	5	5

Conversion of the vehicles to the test oil required flushing the system by driving for 30 minutes on the test oil (with a new filter) and then changing oil and filter again. Appropriate labels were then placed on the vehicles to guard against the use of incorrect oil. A log book was placed in each vehicle to record gas consumption, unusual driving conditions, repairs, oil samples and changes, etc. The drivers and mechanics are keeping excellent records in these books. The log books are periodically inspected in the field.

Summary information is reported to Iowa State University at the time oil drain samples are taken: 4,000, 8,000, or 10,000 miles, depending on the specifications for that particular vehicle (table 3). The oil is drained into a clean pan used only for test oils and thoroughly mixed in the pan before two one-pint samples are obtained. One of these samples is labeled and stored at the site, and the other is labeled and mailed to Motor Oils Refining Company for analysis. The standard American Society for Testing and Materials analytical methods (table 4) are being used to determine the used oil's flash point, viscosity at 100° F and 210° F, viscosity index, benzene and pentane insolubles, total acid and base numbers, and fuel dilution. As indicated in table 4, 21 metals' concentrations are determined by standard spectrographic methods. Duplicate samples that were analyzed by Faber Laboratories of Chicago gave acceptable correlation with MORCO results.

Table 4. Used Oil Analyses

Laboratory analyses		
Flash, °F	Lead	Sodium
Viscosity @ 100° F	Copper	Calcium
Viscosity @ 210° F	Iron	Barium
Viscosity index	Aluminum	Zinc
Benzene insolubles	Chromium	Phosphorus
Pentane insolubles	Tin	Magnesium
Total acid number	Vanadium	Manganese
Total base number	Molybdenum	Cadmium
Fuel dilution	Nickel	Titanium
Antifreeze	Boron	Silver
Water (on selected samples)		Silicon

Dipstick samples were taken on the 1976 Dodge pickups (extended oil drain mileage vehicles) every 2,000 miles through the first oil drain sample to help protect against possible oil failure. These intermediate samples were obtained by means of a suction gun fitted with tubing that could be inserted through the dipstick hole. The length of tubing was premeasured and an end and a side hole in the tubing were provided in order to get a representative sample. The oil in the Matador wagons, two of which were using the BERC test oil, was originally changed and sampled every 4,000 miles. In an effort to further stress the three test oils and to conserve a limited quanitty of BERC oil, the drain interval was increased to 8,000 miles.

When the test terminates, 12 specially selected vehicle engines that used the various test oils will be subjected to deposit and wear ratings. A subcontract has been let to Southwest Research Institute to assist with the deposit and wear ratings and interpretation of the results.

Discussion of Preliminary Results

It should be emphasized that the vehicles involved in the fleet test are operated under a variety of conditions which may range from high speed/high load to stop and go with high idling time. No attempt has been made to segregate the used oil analysis data according to specific driving conditions. Instead, this paper will present the accumulated results from the fleet tested under normal (random) driving conditions. The results from the drain oil analyses for viscosity at 100° F (figure 1), lead concentration (figure 2), iron concentration (figure 3), and total base number (figure 4) are plotted for BERC, MORCO, and virgin oil against the oil drain interval (ODI).

Although the data for viscosity at 100° F is very scattered, there is a definite increase of about 25 percent for the BERC oil and 45 percent for the MORCO and virgin oils as the ODI is extended to 8,000 miles. Data scatter of this magnitude was expected, and the viscosity increase differences are not considered significant.

Lead concentrations vary from 35 to 55 ppm in the BERC oil, from 30 to 120 ppm (majority from 30 to 90 ppm) in the MORCO oil, and from 30 to 90 ppm in the virgin oil at a 4,000-mile ODI. At an 8,000-mile ODI, the concentrations were increased to around 150 ppm for the 3 test oils with some results on the virgin oil being slightly higher. The iron concentrations plotted in figure 3 show similar trends.

The total base number (TBN) results, plotted in figure 4, provide more discrimination among the three oils. The BERC and MORCO oil TBN usually lies between 1.0 and 2.0 at 4,000 miles and 8,000 miles, while about 80 percent of the virgin TBN's lie below 1.0 at 4,000 miles and all lie below 1.0 at 8,000 miles. One data point with TBN = 0 at an ODI of 9,700 miles was obtained. There is an obvious decrease in the TBN for each test oil as the ODI is extended.

The originally low TBN of the virgin oil compared to the modest BERC TBN and relatively high MORCO TBN would explain the lower virgin TBN at 4,000 and 8,000 miles. Assuming that an acceptable TBN should lie above 1.0 (see ref. 2), it would appear that an 8,000-mile ODI using the virgin oil is too long and that a 4,000-mile ODI is marginal.

On the average, fuel dilution ranged from 0.3 percent to 0.9 percent in the summer and from 0.8 percent to 2.0 percent in the winter. This reflects the importance of hot engine oil drain sampling in minimizing fuel dilution during the winter months.

Throughout the test, the pentane and benzene insolubles for all three test oils in the Matador wagons, Dodge pickups, and Chevy Malibus ranged from 0.01 percent to 0.3 percent. They were generally slightly higher at an 8,000-mile ODI than a 4,000-mile ODI.

To assess the effect of a cold weather test on the degree of water contamination of these oils, various duplicate oil drain samples were analyzed using the American Society for Testing and Materials method D-95 (water by distillation) technique. Figure 5 provides data on the percentage of water in the samples that were taken from September 1975 to April 1977. The data generally lie between 0.1 percent and 0.2 percent, regardless of the date or the type of oil. This is evidence that under the driving conditions experienced by these vehicles, water contamination is not a serious problem.

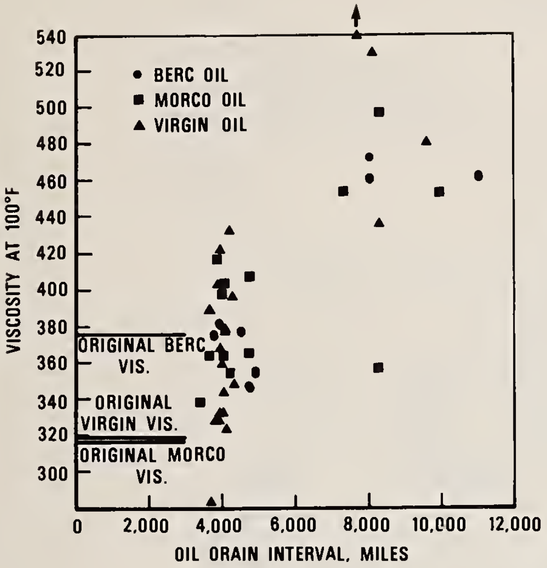

Figure 1. Viscosity data from Matador wagons.

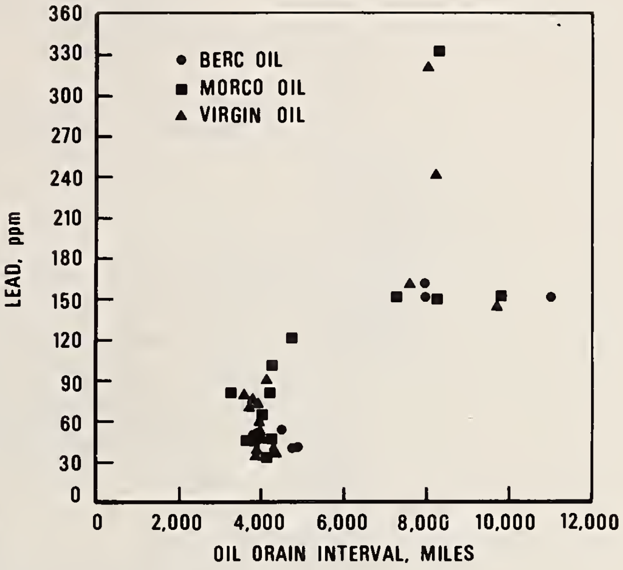

Figure 2. Lead concentrations from Matador wagons.

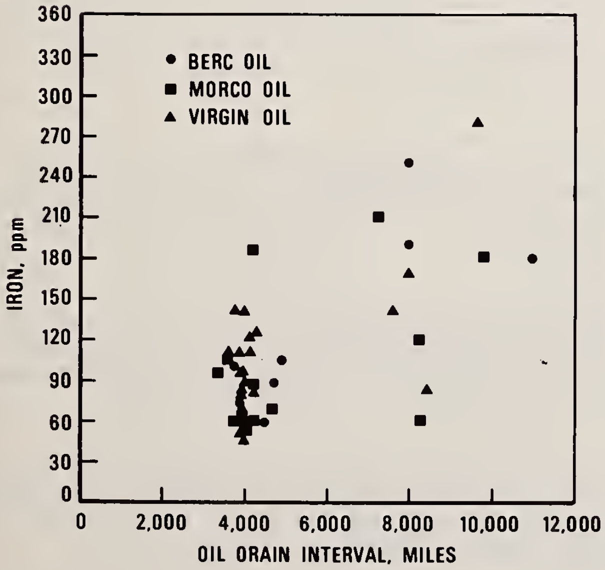

Figure 3. Iron concentrations from Matador wagons.

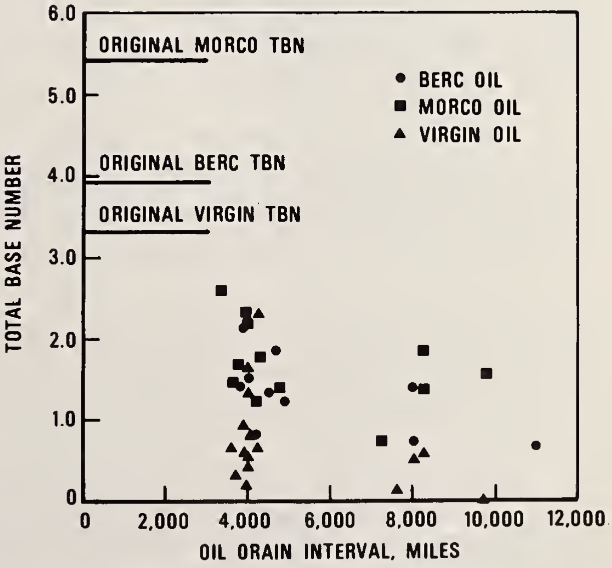

Figure 4. Total base number from Matador wagons.

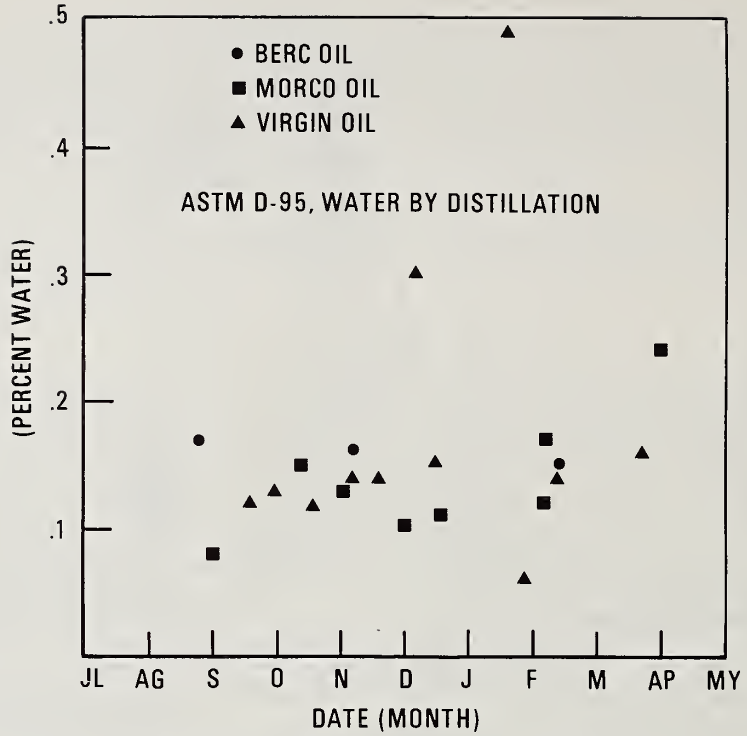

Figure 5. Water analysis of used oil.

Data obtained from the Ford 750 trucks (figures 6, 7, and 8) deviate radically from the norms established with the Matador wagons. This result was expected because of the different engine design, the type of operating service, and the use of leaded fuel. The viscosity at 100° F is plotted against the ODI (in hours) in figure 6. In this figure, the used oil viscosity is approximately 10 to 15 percent below the original value for most of the range. The large and consistent viscosity loss in these vehicles can partly be attributed to fuel dilution.

The pentane and benzene insolubles level is considerably higher in the Ford trucks than in the Matador wagons. In figure 7 the percentage of pentane insolubles (PI) is plotted against the ODI (in miles) and reflects an increasing trend in PI for the MORCO oil data from 4 percent to 6 percent as the ODI is extended from 140 to 280 running hours. The pentane insolubles are quite high in these vehicles but should not pose a serious problem in high dispersancy oils.

In figure 8, the total base number is plotted against the ODI (in hours). As in the Matador wagons, the virgin oil predominantly maintains a TBN below 1.0, while the MORCO oil usually ranges from 1.0 to 2.0. No other distinguishing trends are noted in this data. The concentrations of wear metals in the truck samples were much higher than in the Matador wagon samples. The use of leaded fuel is reflected in the "abnormally" high lead concentrations that range from 1 percent to 2.5 percent.

The rate of oil consumption for the Matador wagons and the Ford trucks is shown in figures 9 and 10. The oil consumption data are expressed as the number of quarts added between oil changes rather than an average consumption for each vehicle. The oil consumption levels are reasonable in all cases, although the trucks consume substantially more oil.

The rate of mileage accumulation for the wagons and trucks is shown in figure 11. A substantial number of the wagons will approach 50,000 miles by the conclusion of the test.

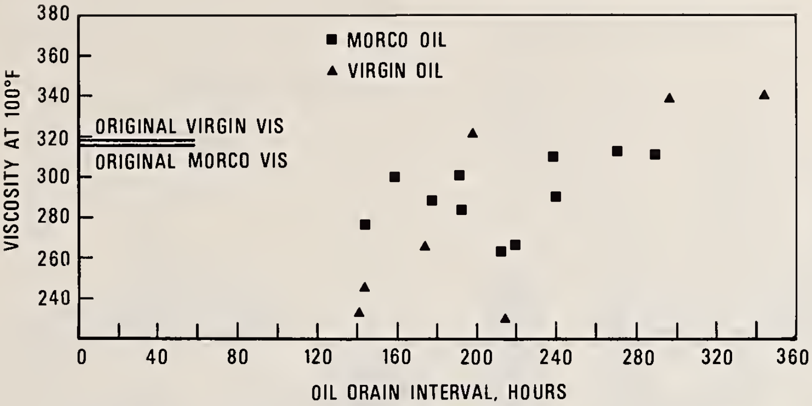

Figure 6. Viscosity data from Ford 750 trucks.

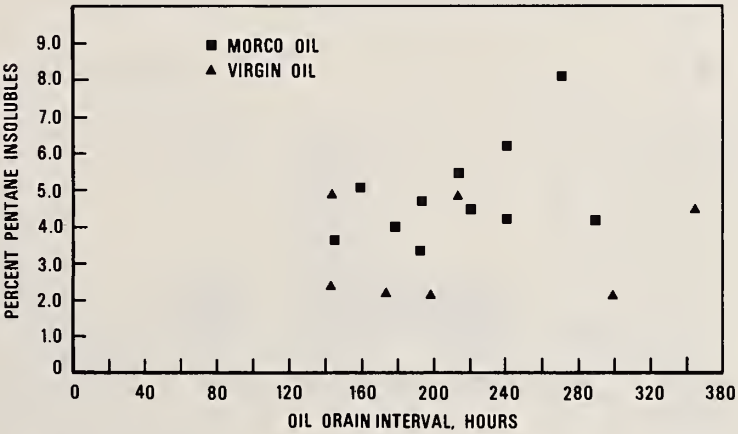

Figure 7. Pentane insolubles level from Ford 750 trucks.

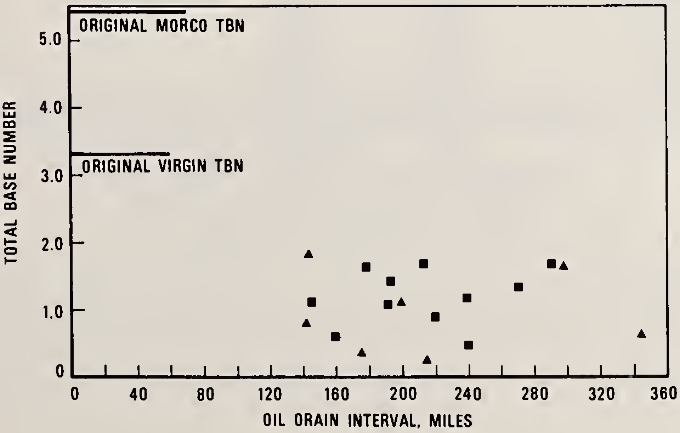

Figure 8. Total base number results from Ford 750 trucks

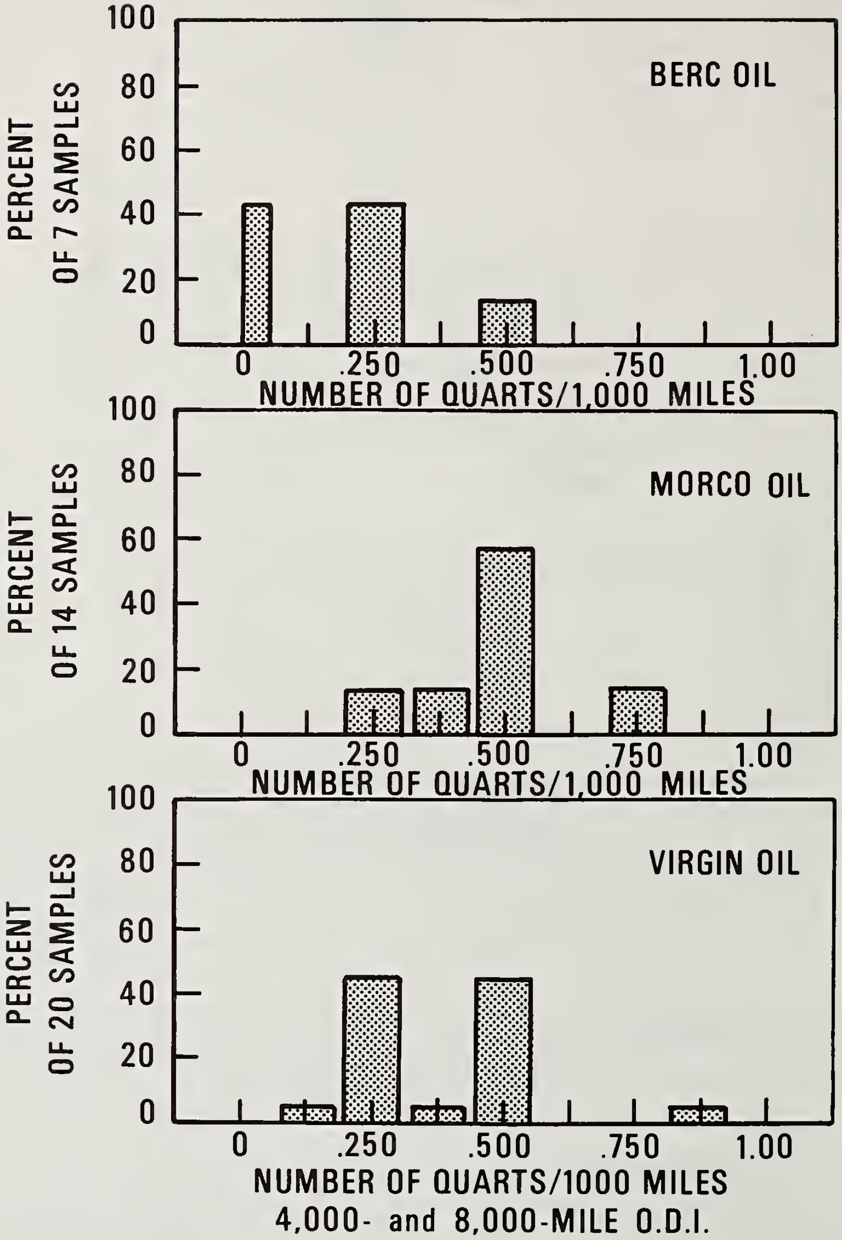

Figure 9. Oil consumption data from Matador wagons.

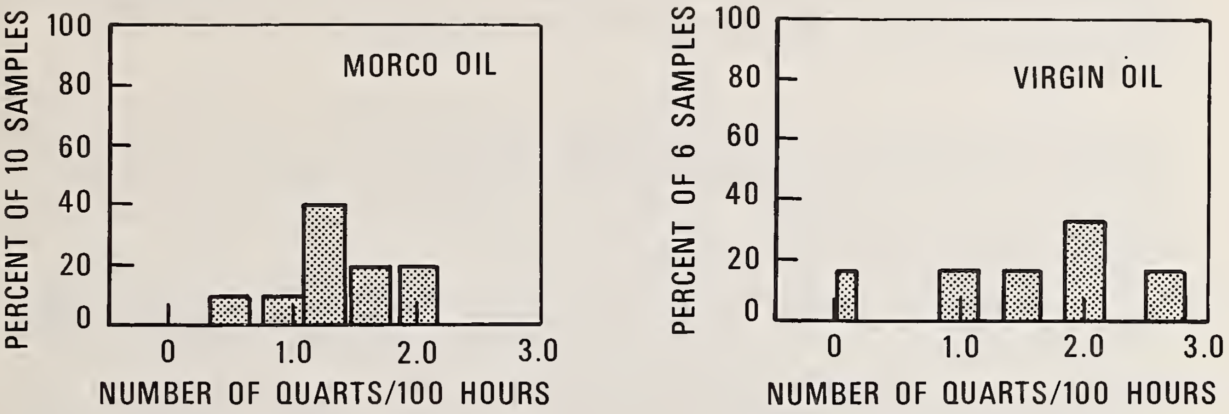

Figure 10. Oil consumption data from Ford 750 trucks.

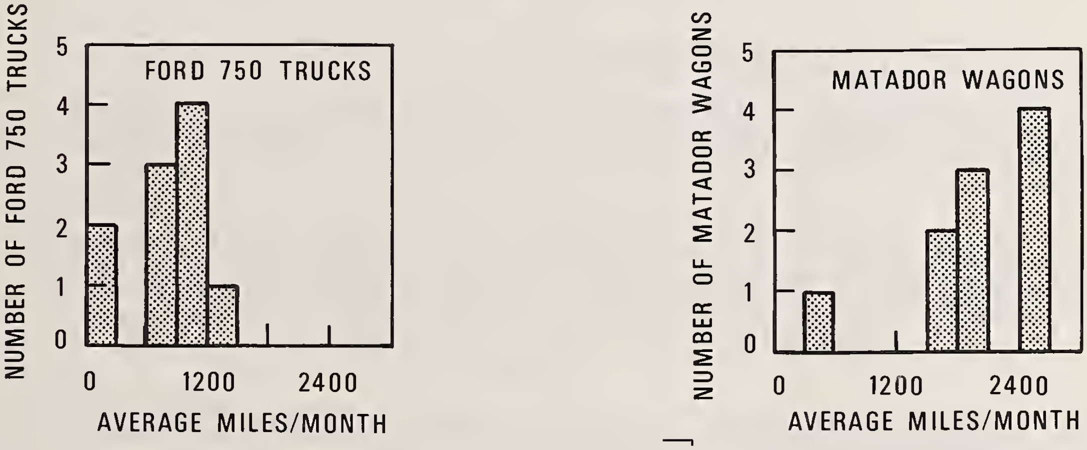

Figure 11. Rate of mileage accumulation.

Data similar to that presented above have also been accumulated on the Dodge pickups and used Chevrolet Malibus. The results of the oil analyses show no obvious differences from the wagons and trucks and are not presented in this preliminary report.

Iowa Farm Survey

One hundred farmers in each of four Iowa counties were selected at random and asked to complete a questionnaire. About 20 percent of the questionnaires were considered valid returns. Comparison with demographic and previous agricultural surveys showed very close agreement and indicated a representative sample.

The average oil consumption per farmer is 50 gallons per year, with a range of 5 to 200 gallons per year. The mode is 30 gallons per year. This translates to an average of one quart per acre of land per year.

The most common uses of the waste oil are prevention of rust, preservation of wood, and control of weeds and dust. A significantly large fraction (11 percent) of the farmers dump their oil on the ground in the farm yard area. The farmers who were collecting and using their waste oil for one of the above uses felt that they would continue to do so even if a recycle/collection service were established. Only two of the respondents felt that their method of disposal was harmful to the environment.

Although generalizations on the reasons for purchasing a particular oil are of uncertain validity, the reasons cited by the respondents were (in order of frequency of response): Society of Automotive Engineers' service designation, the brand their dealer handles, the equipment manufacturers' recommendation, and the brand name. There was a decided reluctance to indicate willingness to purchase re-refined oil because of the large equipment investment involved and because of doubt that waste oil could be restored to original quality.

The economics of collecting oil from farmers on a "milk route" type of operation are quite unfavorable. Assuming an operator could collect 20 gallons per location and could stop at another location every 10 minutes, the transportation cost alone would be $0.31 per gallon, based on standard trucking industry costs of $1.86 per mile [3]. Pickup by the local service station delivery truck would therefore seem to be the only viable method of collection. A maximum of about 1.3 million gallons per year would be available from farmers in Iowa by any collection method. This represents about 20 percent of their total consumption.

Conclusion

These results are preliminary, and final conclusions will not be drawn until deposit and wear ratings are completed.

The re-refined oils are now performing satisfactorily in extended oil drain operation. The total base number of the virgin oil is substantially lower than that of the re-refined products, but no operational difficulties have been observed as a result.

The most serious observation (but not a new observation) is that operating performance of an oil may not be adequately insured by any bid specifications in use today, whether the oil is from virgin or re-refined stock. Low-cost methods of testing for quality are needed for both virgin and re-refined products.

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[1] Whisman, M. L., et al., Waste Lubricating Oil Research, Some Innovative Approaches to Reclaiming Used Crankcase Oil, U.S. Bureau of Mines, RI:7925 (1974).

[2] Asseff, P. A., Used Engine Oil Analyses, SAE Paper No. 770642 (Review, Society of Automotive Engineers Fuels and Lubricants Meeting, Tulsa, Oklahoma, June 1977).

[3] Environmental Quality Systems, Inc., Waste Oil Recovery Practices, State of the Art, Report to the Environmental Protection Agency, PB-229-801 (1972).

National Bureau of Standards Special Publication 556. Proceedings of a Workshop on Measurements and Standards for Recycled Oil - II held at NBS, Gaithersburg, Maryland, November 29 and 30, 1977. (Issued September 1979)

PHILLIPS RECYCLED OIL PROGRAM

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Many of you are aware that Phillips has announced the commercialization of used automotive oil re-refining technology aimed at restoring these lubricants to their "virgin" oil quality. If you have noted the press and trade journal announcements of recent months, you also know that Phillips has a recycled oil plant under construction for 1978 delivery to the State of North Carolina.

Since we are here to exchange ideas and experiences on used oil recycling for the benefit and advancement of the National Bureau of Standards' (NBS) program under Mr. Donald Becker's charge, I have agreed to provide you with some background on the Phillips Re-refined Oil Process (PROP), to tell you why Phillips chose to become involved, to address the principal technical concerns that now impede progress in this recycling movement, and to present data including engine sequence test results on the recycled North Carolina waste oil as processed in our pilot plant at the Phillips Research Center.

Phillips' decision to commercialize the PROP process is based in part on (1) recognition of its potential contribution to resource and energy conservation, and (2) our development of advanced technology that provides for economical restoration of waste automotive oils to the high quality of their "virgin" oil counterparts. The re-refining processes involved are innovative, sufficiently sophisticated so as to avoid environmental and ecological pitfalls, provide for a high degree of oil recovery, and are economically attractive to major re-refining interests.

Phillips' management supports this program as being consistent with the resource conservation goals of our Federal Government as reflected in section 383 of the Energy Policy and Conservation Act of 1975, and as being supportive of the intent of the bills introduced in the 95th Congress, most often referred to as the "Mitchell Bill" (H.R. 5350), which require preferential emphasis on recycled oils in Federal procurement of lubricants beginning in 1978.

We expect that used lubricating oils will be classified as hazardous materials within the solid wastes definitions arising out of the Resource Conservation and Recovery Act. Certainly, this gives added impetus to the urgency of the NBS charge to develop test procedures for the determination of substantial new and recycled oil equivalency that would encourage further activity in the re-refining area. Used automotive oils deserve something better than ultimate disposal as hazardous materials. They become waste materials only if they are wasted--as they are when disposed of in municipal and industrial wastes eventually incinerated or included in sanitary landfills.

Today I want to deal with what I see as the three principal concerns that are inevitably expressed when discussing the acceptability of recycled oils. These concerns are not necessarily unlike those associated with the refining and blending of virgin basestocks with additives as premium-quality motor oils. The general areas to which I refer are (1) the constancy of acceptable raw material or charge stock for the refining process, (2) the sophistication and efficacy of the processing, and (3) the blending with additives and the testing of the end product to demonstrate the attainment of desired quality.

The Phillips PROP technology and our contractual approach to the recycling of used automotive oils impinges on all three concerns, and I want to comment on each.

The first is the matter of charge stock to the PROP process. In the manufacture of virgin base oils, we have long told ourselves that the constancy of the crude source as a charge stock to the refining process is a desirable ingredient. Certainly, we must all agree to this principle. Yet in recent times, many refiners have been forced to make minor to major adjustments in crude sources for continued operation.

These range from imported crude substitution to more subtle changes in production sites or producing zones of individual fields within the industry-recognized but broad geographical descriptions indicated on the application forms for formulation approvals. The degree to which these changes, as they affect lubricant quality, have been evaluated and revealed to those issuing approvals can only be answered by individual marketers.

I make these comments to suggest the possibility of a double standard in this matter of charge stocks to the refining or re-refining process. I hope you have noted in my earlier remarks that the PROP process is being offered initially for the recycling of drain oils from automotive equipment--and that this is a significant condition of our PROP performance assurances to those who contract for our technology with the intent of ultimately blending oils of premium quality. With American Petroleum Institute service SE and military specification quality in mind, the intent at this time is to purposely exclude the intrusion of substantial amounts of industrial and processing oils, metal-working oils, and similar materials that might substantially alter the high viscosity index, paraffinic nature of recycled automotive oils. Thus, with the used oil charge stock essentially restricted to automotive sources, we anticipate an acceptable constancy in charge stock composition to the PROP process.

Date shown in table 1 indicate the variability of the used oils charged to the many batches of oil processed in our pilot plant during the past several months. Major variations involved degree of water (and antifreeze) contamination, fuel dilution, lead levels, and viscosity. Keep these variations in mind. Processed oil data that I will show you later indicate that the PROP process can easily absorb these variations in charge stock without significant differences in recycled product quality.

Table 1. Waste Oil Charge Stock Variations

				North Carolina Only		
Oxygen, Wt. %	1.0	→	10[a]	1.0	→	2.75[a]
Nitrogen, Wt. %	0.03	→	0.54	0.03	→	0.54
Sulfur, Wt. %	0.13	→	0.47	0.13	→	0.47
Chlorine, Wt. %	0.03	→	0.49	0.03	→	0.11
Volatiles (to 600° F), Vol. %	5	→	36	5	→	36
Metals, ppm:						
Lead	1	→	11,000	1	→	11,000
Calcium	600	→	3,720	1,052	→	3,720
Zinc	560	→	1,550	560	→	1,550
Barium	2	→	1,631	2	→	1,631
Magnesium	3	→	500	3	→	500
Manganese	1	→	200	1	→	200
Iron	10	→	330	10	→	330
Phosphorus	600	→	1,410	650	→	1,410
Copper	1	→	120	1	→	67

[a] Includes water.

Certainly, we expect this will be true for our North Carolina venture. This 2,000,000 gallons-per-year plant will be operated initially by Prison Enterprises on a single-shift, 5-day-per-week basis, at an anticipated 500,000 gallons-per-year output. Its charge stock will be comprised of the oil drainings from the 97,000 pieces of State-owned equipment, ranging from passenger cars to dump trucks. North Carolina's used oil collection and recycled oil delivery procedures dove-tail into already established Statewide county delivery systems. However fortuitous this may be for our first PROP plant, this advantage is not necessarily unique to the State of North Carolina, nor is it essential to the economic viability of the PROP program.

The second area of concern is the matter of confidence in the re-refining process itself if you are to accept its products on a par with "virgin" oil quality. Many of us have long been scripted, to use transactional analysis terminology, to look with a jaundiced eye on re-refined oils. At this group's first conference last year, Mr. Hugh Kaufman had words of admonishment on this very point. Past experience suggests some re-refiners marketed products that were little more than oil drainings processed only to the extent necessary to restore some degree of color and clarity. This is an unfortunate legacy to which Phillips intends to make no contribution.

Ours is not a marginal process. PROP plants will provide for the elimination of contaminants and degradation products of prior use and for the neutralization and finishing so as to restore the inherent properties of the original base oils. These data (table 2) show the range of physical characteristics for the recycled base oil products of a dozen or more pilot-plant runs, most of which represent North Carolina-supplied oil drains, but which also include representative samples of waste oils currently being collected by several domestic re-refiners. A quick comparison to our virgin base oil suggests it is very difficult to distinguish between the properties of the re-refined oil and the base oil.

Table 2. Comparison to Virgin Lube Stocks

	Recycled	Virgin
Refractive index	1.4816 → 1.4831	1.4864
Density	0.8726 → 0.8759	0.8756
Molecular weight	384 → 440	449
Carbon, %	86.30 → 86.26	86.39
Hydrogen, %	13.69 → 13.61	13.46
Oxygen, %	<0.02 → <0.01	<0.03
Nitrogen, ppm	86 → 132	93
Sulfur, %	0.03 → 0.04	0.150
Chlorine, ppm	<10 → <10	<10
Sulfated ash, %	<0.01 → <0.01	<0.01
Viscosity index	104 → 100	97
Color	3 → 4.5	3
Viscosity @ 100° F, cSt	61 → 80	80
Viscosity @ 210° F, cSt	7.92 → 9.26	9.20

This is similarly indicated by comparing the infrared scan (figure 1) for our premium-quality motor oil basestock and a similar scan (figure 2) for a PROP-processed composite sample of the same oil after use, as collected by a local service station.

Solvent extraction and/or acid/clay processes are not incorporated in the PROP process, thus avoiding the environmental and ecological concerns that may be involved with such processes. Ninety-percent oil recovery is realized in this continuous process design. Solid wastes, low in volume, are suitable for conventional disposal in sanitary landfills; gaseous and liquid effluents are either recycled in the process or are dischargeable into municipal sewers.

The third area and one of greatest concern to many of you here is that of proof of performance quality.

Our alternative to divulging proprietary details of the re-refining process is assurance of plant performance in terms of demonstrated quality of the final blended product. In the case of the North Carolina project, this means American Petroleum Institute service SE/CC or perhaps SE/CD quality. At this point I wish I could tell you we also have the answer to Mr. Becker's prayer, namely, inexpensive, simple test procedures for the determination of substantial equivalency of re-refined oils with new oil for all end uses. We do not have this simple, inexpensive answer. The timetables of our process development program and our contractual commitments do not allow for our awaiting for this cooperative accomplishment yet to come from conferences such as this and the technical society involvements they engender.

But do not misunderstand me--to us, as well as to the NBS, the need is obvious. In lieu of such yet-to-be found tests, we simply proceeded with the evaluation of recycled-refortified oil formulations in the same manner that we develop motor oils blended with our "virgin" base oils.

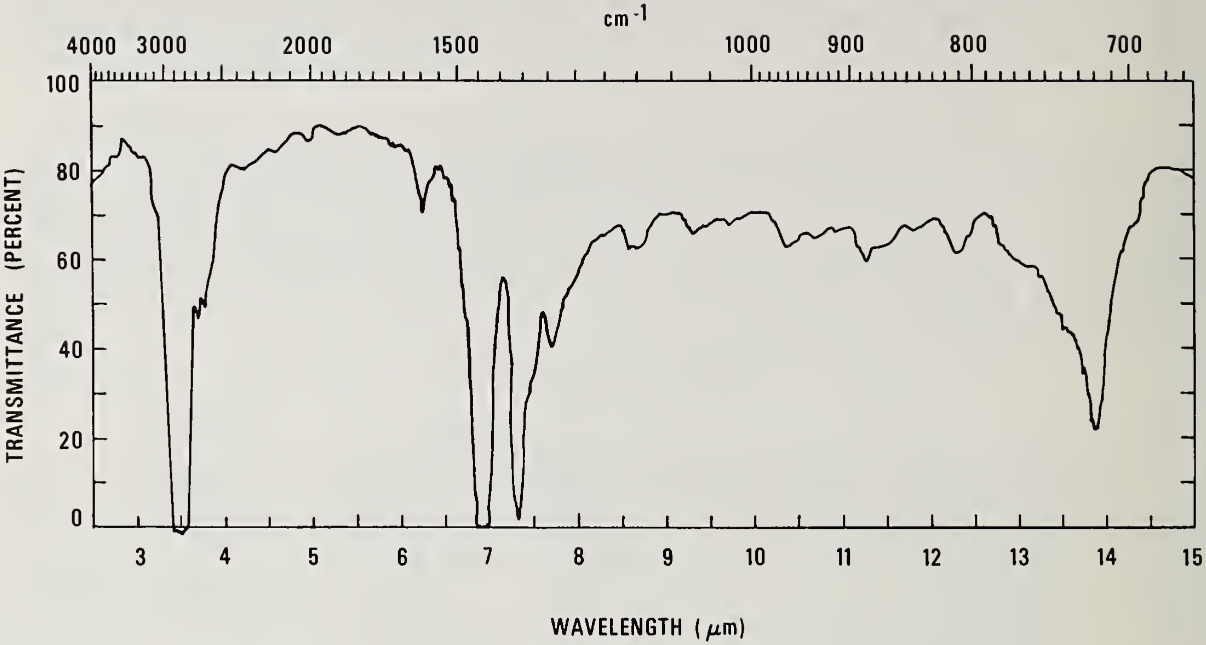

Figure 1. Infrared scan for Phillips' premium-quality motor oil basestock.

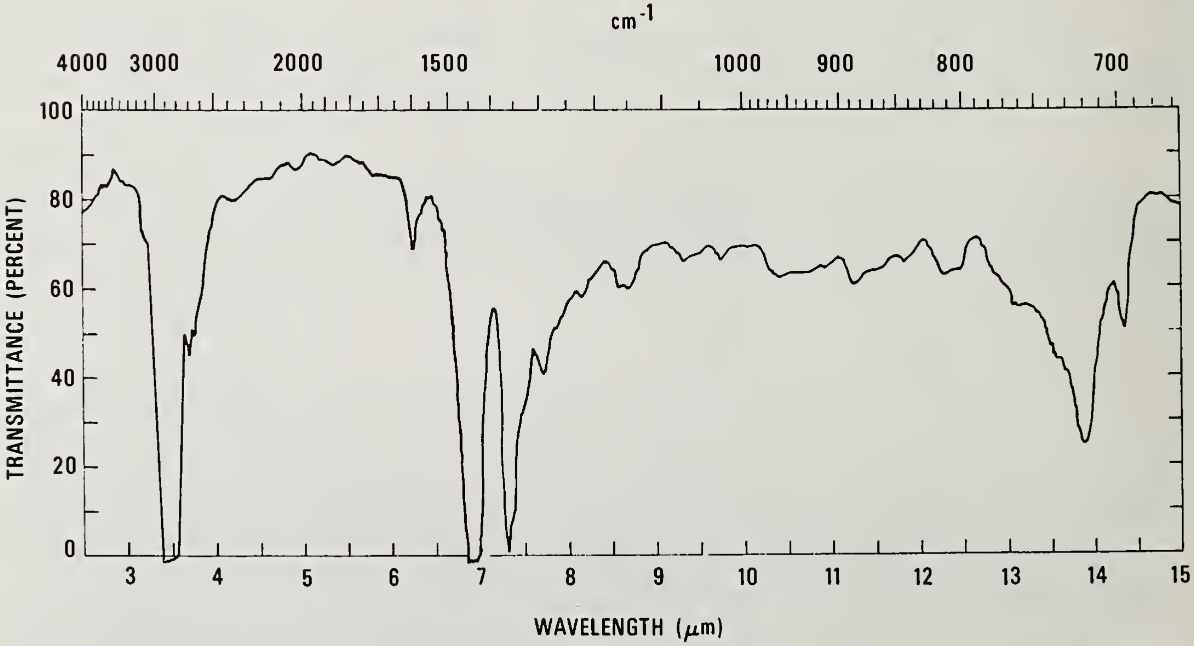

Figure 2. Infrared scan for a PROP-processed composite sample of the same oil after use as collected by a local service station.

Our North Carolina recycled oil prototype formulations are based on successful sequence IIC, IIIC, and VC engine testing. As indicated in tables 3 and 4, we have been able to achieve SE levels of performance with a Society of Automotive Engineers (SAE) 30 grade oil made from 100 percent recycled base oil treated with an SE/CD "universal" additive package. Diesel engine testing is planned, and substantial field experience is being gathered in company vehicles.

Table 3. Sequence IIC and IIIC Results
(SAE 30, SE/CD "Universal" Additive Treat)

		Recycled oil test results	Specification requirement
IIC	Rust rating	8.6	8.4 Min.
IIIC	Viscosity increase, %	119	400 Max.
	@ 40 hours	119	400 Max.
	Sludge rating	9.8	9.0 Min.
	Piston varnish	9.3	9.3 Min.
	Oil ring land varnish	7.6	6.0 Min.
	Ring sticking	None	None
	Lifter sticking	None	None
	Cam/lifter scuffing	None	None
	Cam/lifter wear		
	Average, inches x 10^{-3}	0.5	1.0
	Maximum, inches x 10^{-3}	1.0	2.0

Table 4. Sequence VC Results
(SAE 30, SE/CD "Universal" Additive Treat)

	Recycled oil test results	Specification requirement
Average engine sludge	9.6	8.5 Min.
Piston varnish	8.6	8.2 Min.
Overall varnish	8.0	8.0 Min.
Oil screen, % clogged	0	5.0 Max.
Oil rings, % clogged	0	5.0 Max.
Compression ring sticking	None	None

By and large, our engine testing to date, both laboratory and field, has been conducted with low- to mid-range SAE 30 grade formulations. This reflects the naturally occurring viscosity for the finished oil treatments resulting from processing of the North Carolina used oils. This does not preclude, however, a plant operator's use of virgin base oils for seasonal adjustments as needed of finished oil viscosities, nor the consideration of viscosity index-improved multigraded oil should North Carolina opt for an SAE 20W-40 grade, nor the ability to re-refine higher viscosity products, such as diesel oils.

In this regard, the recycled oil from the PROP plant will provide, volume-wise, some 50 to 60 percent of their annual automotive lubricant requirements. Hence, it is feasible to concentrate on the conversion of the available automotive oil drainings to high-quality oils only. Presumably, the future new oil purchases of the State will first fulfill their needs for lubricants other than crankcase oils, namely, automatic transmission fluids, hydraulic oils, and gear oils. However, I do not mean to imply that the PROP process is limited in its application only to motor oil blending; but for the North Carolina program, this is the practical approach to take.

We anticipate that the North Carolina plant will be operating by mid-1978. We plan to confirm our pilot plant product testing by additional sequence tests of finished formulations blended from production base oils upon plant startup. We are continuing to examine, by pilot plant processing, other sources of automotive waste oils representative of those being processed by or of specific interest to prospective customers of the PROP process. This promises to keep us very busy in the foreseeable future.

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THE SYSTEM OF USED OIL REGULATION IN THE FEDERAL REPUBLIC OF GERMANY

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Used oil represents a valuable raw material, even if the original user cannot himself put this used oil to further use as for example, a lubricant or fuel. The increasing realization of the value of used oil can be illustrated by the laws which have been passed in Germany on the reclamation of used oil. The first law concerning used oil collection and reclamation was passed around 1935, with the primary motive being conservation of lubricating oil, and the secondary motive being environmental protection. There exists in the current law a definite change compared to the various remedies in the past, in that the owners of used oil can obtain free storage of up to 200 kg of collected used oil. There is no storage costs for this used oil since the free storage of used oil has been made mandatory in the legislation. The Federal Department for Industrial Science, which has the responsibility for coordinating this program, has obtained a sufficient quantity of locations for this mandatory, free-of-charge storage.

A reclamation program is also required, for reasons of environmental protection, and includes through the Federal Department, an arrangement whereby the total costs of used oil collection, transportation, and reconditioning are taken into consideration. Proceeds to fund this program are obtained by means of a mineral oil tax which has been placed on all lubricating oil, gas oil which has lubrication uses, and lubricants with heavy oil constituents. The amount of re-refined oil sold comprises about 15% of the total lube oil sales in the market place. The actual costs of this recovery system are dependent upon the oil quantities, and adjusts itself according to the average costs experienced by the recyclers and disposers, including both re-refiners and burners.

The purpose of the used oil law is to encourage the recovery and reuse of petroleum, petroleum products, and similar types of waste from depots, storage, and transport facilities. It is realized that this type of used oil is often mixed with foreign matter in varying amounts. The foreign materials most frequently found in used oil are water, solvents (e.g., tri-chloroethylene, per-chloroethylene, acetone, organic ester compounds, and organic nitro compounds), as well as solid matter (e.g., dirt and metallic particles from abrasion). In order to encourage the recovery of segregated, clean used oil, the subsidy payments for the costs of mandatory free storage and recycling apply only to products with up to 10% foreign matter content. For products containing more than 10% foreign matter, the used oil owner must pay the costs of collection and reclamation. The amount of such costs are determined by the (Federal) Department according to a consignment price list, which is published for each type of recycling. The used oil owner is thereby given the opportunity to decide on which method of recovery would be most feasible for the least cost.

As long as this economic incentive can provide a motivation for recycling (for used oil with up to 10% foreign matter content), the program can be termed successful. Used oil containing more than 10% foreign matter, e.g., emulsions, does not fall within this category. For these materials, the law provides a requirement that used oil owners who generate more than 500kg of such used oil per year must provide information on the origin of the used oil, its storage location, and the cost for recycling or disposal.

The positive experiences with the (German) used oil law has also had an effect on the guidelines for subsidy rates of the European Community since June 6, 1975. These have resulted in the desired objective of a legal, unified regulation which is not only in the interest of environmental protection, but also guarantees a basis for fair market competition.

The economics of the used oil law should be convincing, with a cost of about 40 million Deutsch Marks per 350,000 (metric) tons of reclaimable used oil. An important consideration in such regulation is illustrated by the following:

- A metric ton of oil which has been removed from a wastewater treatment facility would have required the same biochemical reaction as the treatment of residential sewage of 40,000 people.

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TESTING AND USE OF RE-REFINED ENGINE OILS IN EUROPE

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An important requirement for fulfilling the National Bureau of Standards' (NBS) responsibilities on test procedures for recycled oil is to become totally familiar with all aspects of testing lubricating oils, both recycled and virgin. As is well known, there are a number of countries outside the United States which have also been involved with oil recycling for years. In particular, the system in West Germany has been held up as a model for oil recycling programs. It was felt important to determine the details of the West German program and of the quality-control testing which has been used in making this system work. This is one reason that Mr. Irouschek was invited to make a presentation at this meeting. Even though he was not able to attend, his paper provides considerable information on their work.

This past summer I therefore traveled to Europe to meet with persons involved in lubricating oil testing and recycling. This trip had a dual purpose. In addition to visiting many individuals in the lubricating oil field, I also attended a lubrication research conference held in London. This conference had over 100 delegates attending, with more than 60 representatives of lubrication research and testing. I visited with four re-refiners and several major oil companies, met with representatives at the Institute of Petroleum, and had discussions with engine manufacturers and a major additive manufacturer.

In order to guide our discussions during these visits I made up a list of over 30 questions on testing and specifications for recycled petroleum oils (primarily engine oils) and also had them translated into German for those visits I would make in West Germany. In the limited time I have available here I cannot possible report on all the discussions and information I received. What I will try to do is provide information on certain particularly interesting subjects, and then summarize some additional salient points. Let me also mention that I made a special effort to examine the retail lubricating oil containers, both in England and in West Germany, noting especially the specifications met by that particular oil.

In West Germany it was estimated that approximately 90 percent of the 350,000 tons of used oil is collected. There are approximately 9 major re-refiners in the country which recycle this used oil, of which about 20 percent is burned or incinerated, 40 percent re-refined into high-quality automotive lubricants, and the remaining 40 percent processed into industrial oils or asphalt production. The reprocessing for asphalt often apparently includes a vacuum distillation step, but no chemical treatment, and the product is called "black oil." It is unclear at present as to the concentrations of the various contaminants which remain in the oil with such processing.

The collection of waste oil in West Germany is made considerably simpler since it was estimated that only about 15 percent of the lube oil is currently being changed by do-it-yourselfers, compared to about 30 to 40 percent in the United Kingdom and almost 50 percent in the United States. There were reported to be increasing problems in the United Kingdom due to environmental pollution from indiscriminate dumping of used oil since increasing numbers of car owners are changing their own oil.

With regard to labeling, both West Germany and the United Kingdom do not require any indication on the containers that a lube oil is re-refined, and they also do not require performance characteristics or classifications to be displayed on the container. In West Germany, many of the virgin oil containers have no classifications or indications of tests passed (e.g., SE or SE/CC). However, the West German re-refined oils that I saw did have performance indications (such as SE/CC) and usually other statements about specifications met or tests passed. In the United Kingdom, most oils had performance specifications (SE/CC, automobile manufacturers, etc.). I was somewhat surprised that much of the automobile lube oil sold in Europe has a 20W-50 viscosity, and some of the largest companies even sell their oil with no viscosity labeling at all. Much of the motor oil sold retail in the United Kingdom is in five-liter cans at auto parts stores, discount stores, etc.

In my discussions with several major virgin oil refiners, they admitted that a good re-refined lube oil is an excellent quality basestock, but they also stated that the general public is still prejudiced against re-refined oil, even in West Germany. One refiner stated that if he used a re-refined oil in his top line and a competitor found out about it and started spreading the word, the image of that oil would be severely damaged. He went on further to add that the primary reason that many virgin oil refiners do not put performance characteristics or tests on cans in West Germany is to prevent comparisons between their oil and a less expensive brand. They spend large amounts of money in advertising to convince the public their oil is "better" than all others, and they apparently would prefer not to permit easy comparisons. West Germany also very strongly believes that the lack of speed limits on their autobahns requires the absolute best oil in order to prevent damage or premature wear to their vehicles.

The problem in the United States due to SA and SB oils (nondetergent oils or oils with only inhibitors added) apparently does not exist in Europe. The Germans claim no SA or SB oil is made for motor vehicle use; in the United Kingdom, apparently service stations may have it in the back, in bulk, but rarely use it except for very old cars or "clunkers." This is apparently so because almost all the lube oil in both countries is imported and is therefore controlled by the very large oil companies. The Germans claim that such a low-quality oil used on the autobahn (at 100 to 150 miles per hour) would severely damage any vehicle in which it was used.

The motor oil testing situation appears to be considerably different in West Germany than in the United States, primarily due to the engine manufacturers' efforts to control the lube oil quality in the marketplace. There are at least three engine manufacturers involved in this, to a greater or lesser extent. The major one is Daimler-Benz. These manufacturers test the oils and then provide a list of oils which can be used in their automobiles; these lists are continuously updated.

Daimler-Benz publishes a book which contains a listing of all the lubricating oils which are approved for service fill in their automobiles. In order to be placed on the list, an oil must be tested using the Daimler-Benz OM-616 engine sequence test. The refiner (or re-refiner) must pay for the engine test and subsequent evaluation (reported to be about $10,000), plus pay an additional substantial publication fee. This book contains a number of re-refined oils which have passed all the required tests.

Any oil which is tested by the engine manufacturer and fails to meet the requirements of that manufacturer is removed from the list, and a special letter to that effect is sent to all dealers and service shops. It was stated that such an action could destroy the marketability of even a major product line. In addition, apparently all of these companies have the option to sample oils from the marketplace at any time to compare them to their requirements and to the samples originally submitted to them. Just the threat of this action is a powerful deterrent to anyone who might want to lower their quality standards. In the United Kingdom, the testing system seems to be quite similar to that in the United States, with a government agency possibly reacting to specific complaints of problems.

In addition to the American Petroleum Institute classification (e.g., SE, SE/CC), which is widely used in Europe and all over the world, the Committee of Common Market Automobile Constructors has come out with its own set of lube oil test specifications (second issue, October 1976). These specifications include alternatives to some of the American Society for Testing and Materials (ASTM) engine tests (e.g., for low-temperature sludge--the ASTM VC test or the Fiat 600D test, for high-temperature oxidation--the ASTM IIIC or the Petter W1, for bearing corrosion--the L-38 or the Petter W1; in addition to the previous tests are added the ASTM IIC [rust], the OM 616 [wear], the Ford Cortina [high-temperature deposits], and the Fiat 124C [preignition]).

One important aspect of this trip was to determine whether or not the widely acclaimed success of the West German re-refined oil system was based on a thorough scientific and technical testing program and the resulting data. Unfortunately, it apparently is not. As in the United States, there appears to be no organized monitoring of the marketplace to see that any particular oil, virgin or re-refined, has even been submitted for testing anywhere. In the case of Daimler-Benz (or other automobile manufacturers), in order to become listed in their book (as previously mentioned) a re-refined oil must pass the OM 616 engine sequence test, plus the re-refiner must apparently supply a history of tests (chemical and physical) on individual batches of the re-refined oil for the past 1 to 2 years.

These chemical and physical tests are similar to ones normally run by the European re-refiners in order to monitor their own product quality, and include such things as neutralization number, flash point, viscosity, viscosity index, sulfated ash, pour point, carbon residue, and refractive index. Daimler-Benz also retains samples of the formulated oils which were engine tested and uses infrared spectroscopy to determine whether marketplace samples are similar to the original samples which were tested.

As mentioned previously, no one I talked to was aware of any substantial study which had been made to verify the capability of these or other tests (engine tests or monitoring tests) to assure the quality of re-refined oil, but apparently it works. I did find that two scientists involved in lubrication research and testing have recently approached the West German government to fund such a study. I discussed this project with them at the Imperial College meeting, and I hope to cooperate as much as possible in our respective areas of investigation. In addition, I understand the United Kingdom Ministry of Defense is interested in initiating a study to investigate the batch-to-batch variation of re-refiners in the United Kingdom who are interested in submitting their oil for bid on military contracts. The Ministry of Defense now accepts re-refined oils for military use, but requires complete tests for each batch (including engine sequence tests, where applicable).

The information obtained during these visits seemed to me to indicate the lack of a sound technical basis for adequate quality-control methodology for re-refined motor oils. There was a great deal of interest in the National Bureau of Standards' program and in current efforts in the United States, especially since the United States military specifications and the American Petroleum Institute/American Society for Testing and Materials/Society of Automotive Engineers lubricating oil classification system are both used throughout the world. One West German re-refiner stated he would be very happy to see the United States military specifications for lube oil revised to eliminate the exclusionary clauses since NATO [North Atlantic Treaty Organization] uses the United States specifications and the revisions would allow sales of re-refined lube oil to the West German army. This trip also provided the contacts for coordinating our efforts with the several proposed European efforts and should thereby help obtain the maximum amount of information and data possible in support of the NBS program.

In addition to the information obtained in Europe, correspondence with the South African Bureau of Standards (SABS) has provided much information on their oil recycling efforts and testing program. The SABS has prepared three national specifications for engine oils. These are SABS 704-A-1974 (similar to DEF-2101D and approximately the same as MIL-L-2104B); SABS 399-1972 (similar to MIL-L-46152 and SE/CC); and SABS 400-1974 (similar to MIL-L-2104C or CD). In these specifications, no distinction is made between virgin oils and re-refined oils. However, for re-refined oils the specifications have certain requirements for both control of the raw material feedstock and control of the re-refining process itself. When these conditions are approved by the SABS, a permit is issued to a re-refiner to use the "SABS mark."

Apparently, the experience in South Africa with re-refined oils has been satisfactory, since a letter from the Director General of the SABS states:

> "Field experience with the use of engine oils formulated with re-refined base oil produced under our SABS mark scheme over the past 25 years has proven that such oils are of equal performance and quality as engine oils produced from virgin base oils using the same additive formulations in the same concentrations. In addition to field performance internationally recognized engine performance tests e.g. Caterpillar, Petter, etc., were also carried out on re-refined oils in approved formulations both by our own laboratories as well as by such recognized organizations as South West Research Institute in the USA and Ricardo and Lubrizol in England."

As a part of the quality assurance program, there is a series of required tests on every batch of re-refined oil which is produced, and additional tests which are applied to every fifth batch of the re-refined basestock. There are also tests which are to be applied to the blended and formulated oils which are made using the approved re-refined base oils. The literature from the SABS identified these tests as follows.

Base Oil Tests on Every Batch

- "Free from suspended matter, sediment, water, and other impurities"
- Viscosity at 99° C
- Color
- Neutralization number
- Acid number
- Ash
- Light fractions (% volume)

Base Oil Tests on Every Fifth Batch

- Copper corrosion
- Carbon residue
- Flash point
- Viscosity index
- Saponification value

Blended and Formulated Oils - Tests on Every Batch

- Viscosity at 99° C
- Viscosity at 37.8° C
- Viscosity at -18° C (W grades only)
- Viscosity index
- Additive concentration by sulfated ash or other chemical or spectrochemical method

Blended and Formulated Oils - Tests on Every Fifth Batch

- Pour point
- Flash point
- Foaming properties
- Homogeneity

In addition to the tests indicated above, it was stated that the re-refiner must keep records of all quantities of oil and additives used and the test results which are obtained. Any new or different formulation or treat level cannot be used without specific approval by the SABS. All products which meet the requirements as stated above are marked with the special SABS standardization mark and are approved for sale within the country. The quality of the final product is assured and maintained not only by the refinery (or re-refinery) laboratories, but also by monitoring by the SABS laboratories through random sampling and testing of the lubricating oil on a routine basis. Samples are also often received from consumers for check-testing, especially when investigating a problem which may have occurred with use of an SABS mark-bearing product in the field.

The SABS efforts as described above are mainly concerned with engine lubricating oils, whether formulated with re-refined or virgin base oils. The SABS has also developed purchasing specifications for industrial oils; however, these latter specifications are not considered as national SABS specifications and the standardization mark scheme does not apply to these products. The onus in this case, therefore, is on the consumer to insure quality conformance.

The above information summarizes what I have learned about the use and quality control of re-refined lubricating oils in the various foreign countries. I think it is obvious that they have developed significant amounts of data which should be useful to the NBS in its Recycled Oil Program, and we will continue to look for additional information and data which will provide the necessary scientific and technical base for substantial equivalence.

SESSION VI. ADDITIONAL RECYCLED OIL ACTIVITIES

National Bureau of Standards Special Publication 556. Proceedings of a Workshop on Measurements and Standards for Recycled Oil - II held at NBS, Gaithersburg, Maryland, November 29 and 30, 1977. (Issued September 1979)

WASTE OIL RECYCLING UTILIZING SOLVENT PRETREATMENT

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Abstract

A research program was initiated at the Bartlesville Energy Research Center (BERC) in 1971 to pursue the more efficient utilization of used lubricating oil for the conservation of natural resources and abatement of environmental pollution attendant to the disposition of such material. After evaluating existing technology, the BERC scientists developed a new process which has been proven on a pilot-plant scale and is believed to be both technically and economically viable with less adverse environmental impact. Pilot-scale samples produced by the new technology have passed engine tests and are currently in the 15th month of fleet tests with satisfactory performance. Plans are cited for a 10-million gallon per year demonstration plant to enhance the transfer of technology from government to the public sector.

Introduction

As recently as 1971 there was little or no visible research, either in industry or government, being directed toward a more efficient utilization of used lubricating oils. This was true despite an obvious decline in the re-refining industry from nearly 150 re-refiners in 1960 to fewer than 40 in 1975.

The combination of factors causing this decline is well known to this group. The decline in re-refining, coupled with the increasing shortages of petroleum, have resulted in the burning of about 45 percent of the used automotive crankcase drainings, with only a few percent being recycled and far too much being indiscriminately dumped. It is estimated that the greatest single pollutant in the Atlantic ocean is used lubes that are dumped into sewers and streams and eventually find their way to the ocean.

Faced with a mission of conserving our natural resources and related environmental concerns, the Federal Government is in a unique position of providing unbiased research in the field of used oil reclamation that can be transferred to the public domain without proprietary encumbrances or conflicts--and such a program was initiated in 1971.

Experimental

The principal objective of this program of research was to develop new and improved technology for reclaiming used lubricating oil and to stimulate work by others in the field. The first step in this development was the evaluation of existing processes [1].[1] From information derived from patents and the scientific literature as well as personal contacts, our research staff reviewed many different approaches to reclaiming used automotive lubricating oil. Among these were the prevalent acid/clay process, caustic treatment, direct distillation/clay, propane extraction, treatment with fuming acid, and others. We were aware that quality lubricating oils could be and were being produced by the acid/clay process, but increasing environmental pressures related to acid sludge disposal led to an aggressive search for improved processes--preferably a process that would economically produce a quality oil and be more environmentally acceptable.

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[1] Underlined numbers in brackets indicate the literature references at the end of this paper.

A hydrocarbon compositional study [2-7] of many feedstocks by our staff showed only minimal regional and seasonal variations in feedstock composition. This composition characterization included typical re-refinery feedstocks from widely divergent locations across the continental United States. The contaminants were measured and reported for these feedstocks, and, more important, the hydrocarbon composition of each was determined by a combination of chromatographic and mass spectral techniques. The remarkable similarity of the composition of these feedstocks strongly suggests that consistent product quality can be achieved from a re-refinery for any season or geographical locale.

An early conclusion reached by our staff was the desirability and perhaps the necessity of a full vacuum distillation to restore used oil to original quality and provide fractions based upon boiling range and viscosity to broaden the spectrum and versatility of end products. Distillation of raw used crankcase oil proved to be impractical because of coking and fouling of processing equipment. Our investigations showed that cracking of unspent additives and other contaminants was evident at temperatures below 450° F. The resultant fouling required periodic shutdowns to clear lines and equipment of builtup coke that is detrimental to heat transfer in processing equipment.

To minimize fouling, a pretreatment that would substantially reduce coking precursors was needed. We have investigated dozens of pretreatment steps [8], and at this time we believe that a solvent system composed of one part isopropyl alcohol, two parts normal butyl alcohol, and one part methylethyl ketone represents a compromise between the most efficient solvent precipitation system and the most practical system. Three volumes of this solvent system are used with each volume of oil. We have found that combinations of alcohols are very effective but that solvent-to-oil ratios of 8 to 1 make such systems impractical. Propane is an effective pretreatment solvent, but the equipment required to maintain propane near its critical point makes propane less attractive than the three-component solvent system in readily available facilities.

The system that we have developed for the reclamation of used automotive lubricating oil consists of the following steps:

Step 1. Using distillation, the oil is first dehydrated to remove water and light ends. The light hydrocarbons are burned to recover their caloric content, and the water is treated for discharge into the sewer system.

Step 2. The dehydrated oil is mixed with the appropriate solvents and sent to settlers. It is believed that centrifugation, while more complex and expensive, might, in some cases, replace the settlers, but it is possible to operate continuously with a number of settlers. The sludge is allowed to settle for an appropriate period, after which the oil-solvent mixture is drawn off and sent to a solvent stripper while the sludge is drawn from the bottom of the settler and also stripped of solvent using heat. The solvents can be reused, and solvent losses are estimated realistically at about 0.5 percent.

Step 3. The solvent-stripped oil is sent to a fractionation tower where a fuel cut and several lube oil cuts are taken.

Step 4. These fractions are then clay-contacted individually, or they are blended prior to clay contacting. The clay-contacting step generally requires higher temperatures and more clay than do virgin-derived lube stocks. This is attributed to traces of metals and oxygenates left after solvent precipitation and distillation. Typically, about 0.4 pound of an activated bleaching clay such as Filtrol 20 is used per gallon of oil, and the temperature is elevated to near 425° F with steam sparge to produce an oil with a color of about 1-1/2 and a bland odor. Clay is removed by means of a filter press, and the oil is subsequently reformulated with appropriate additives for the designated service.

The process yields are typically 70 to 75 percent based upon the dry oil feedstock or near 90 percent on an oil-only basis. Since the sludges produced from both the solvent treatment and the distillation step are essentially neutral, they offer potential for use in roofing and road asphalt applications.

Oil produced by this process, for which two patents have been allowed [9,10], has been bench-tested exhaustively and has also been submitted for engine sequence testing [11,12]. Two oils re-refined by technology just described and one commercially re-refined automotive lubricating oil were subjected to engine test sequences IIC, IIIC, VC, and L-38. The commercially re-refined automotive oil successfully passed the IIC, IIIC, VC, and L-38 engine-test evaluations required by automobile manufacturers to meet the standards established for service SE. One oil processed by BERC-developed technology successfully passed IIIC and VC sequence tests and, with the addition of 1 percent corrosion inhibitor, passed the IIC evaluation. The second BERC-produced oil successfully passed the IIIC test, and this same oil with additional corrosion inhibitor passed the IIC evaluation after an initial fail. The L-38 test was not run on BERC-produced oils, but a bearing-corrosion bench test that is reported to correlate well with the full-scale L-38 was run on one of the two oils with a successful pass. These engine tests are believed to be the first documentation in the United States of successful passing of engine sequence tests by re-refined lubricating oils to meet standards established for service SE.

Further BERC-produced oils are satisfactorily performing in fleet tests in State of Iowa vehicles. This program is now in its 15th month, and at the end of 2 years a number of these vehicles will be disassembled and compared with like vehicles in similar service on commercially re-refined oil and on virgin oil. All test oils were performing satisfactorily at the 4,000-, 8,000-, and 10,000-mile drain intervals. Some data seem to indicate marginal superiority with regard to performance of the re-refined oils relative to new oil. Final comparisons among these oils will be based upon deposit and wear ratings to be made by an independent laboratory and on the accumulated analytical data from oil change samples.

Not all of the waste oil recycling program at the BERC is devoted to the development of new technology. Some areas of more basic research are in progress, with still other areas to be undertaken as time, personnel, and funds permit. Currently, our staff is conducting a closed-loop composition study designed to investigate basestock compositional changes resulting from repeated use and processing of a motor oil. The original oil for this study was blended within our facility from virgin-derived basestocks and fully formulated with additives. The composition of this finished motor oil was studied while aliquots of the oil were charged to staff-operated automobiles and driven in normal engine service. The used oil from this fleet was subsequently collected and re-refined using the BERC technology. Aliquots of both the used oil and the re-refined oil were submitted to composition studies. At least one more cycle of use and re-refining is scheduled for this oil. We hope to define the compositional changes accompanying use and re-refining, as well as to determine the feasibility of repeated processing.

We also are developing a test method for evaluating coking and fouling tendencies of feedstocks to re-refineries. We have had some very encouraging results from this study and believe that we can accurately estimate the coking tendencies of any re-refinery feedstock at any selected temperature up to the point of thermal cracking of the hydrocarbons. This analytical tool will be useful for evaluating pretreatment effectiveness to reduce coking and fouling precursors in heat exchangers and distillation apparatus.

A predesign cost estimate for a 10-million gallon per year re-refinery, based upon the BERC technology, was made by the engineering firm of Richard J. Bigda & Associates [13]. The preliminary conclusions from this study include an estimated capital cost of $3 million for the plant. Waste oil purchased at $0.15 per gallon could be re-refined into blendable lube oils for $0.39 per gallon. Considering a selling price of $0.47 per gallon in bulk, the return on investment before taxes would be about 45 percent. Inflation plays havoc with numbers such as these, even before they are published, but relative costs and profits are probably still valid. Bigda and Associates determined that the operation would be very sensitive to the cost of the used oil, but plant capacity could be doubled or tripled with only a modest increase in capital investment cost.

Currently we are directing the major thrust of our investigations toward engineering-type studies in pilot-scale operations that will aid in design of large-scale plants incorporating the BERC technology. Included in these studies are dehydration operating parameters, optimum solvent-treatment conditions, solvent stripping techniques, and fractional vacuum distillation.

Two recent economic studies have been made that involved comparisons of the BERC technology to the acid/clay reclaiming procedure. The first of these studies, made by Richard J. Bigda & Associates, indicated that capital investment would show a slight advantage for the acid/clay process, but total operating costs favor the BERC solvent process.

A significant conclusion of a more recent economic study was that the BERC method produces re-refined oil at a cost lower than that of the acid/clay process, and, as such, the BERC technology has a significant advantage over the acid/clay process both economically and ecologically.

With regard to other new processes that have appeared in the past and recently in the patent literature, we and other investigators have evaluated many of these for technical viability. We have found that many proposed processes are technically unsound. Therefore, we caution those seeking new processes for converting used automotive lubricating oil into a high-quality product to assess technical viability carefully before evaluating economic factors. Actually, the BERC process is the only new process for which published data are available to support its technical viability. No other new patented process has ever established, through published engine tests or fleet tests, that high-quality automotive lubricating oil can be produced using such methodology.

A member of the BERC staff recently visited France with Mr. Francois Audibert, the principal investigator of the French propane extraction method. Mr. Audibert reported in this interview that propane extraction cannot compete economically with acid/clay technology.

We were able to also visit Viscolube in Milan, Italy, which is one of only two plants that use the French propane process. Mr. Schieppati, manager of the Italian plant, reported that propane treatment currently is very marginal economically. However, Mr. Schieppati believes that with new legislation to control the competition for used oil in Italy by those who would burn it will improve the profit picture. Similar steps in the United States to discourage burning and increase availability of feedstock are needed.

Currently at BERC we are looking toward the selection of a partner from the private sector for the purposes of designing, constructing, and operating a 10-million gallon per year demonstration plant incorporating the BERC technology. Timing for startup of this plant at current funding levels is for January 1980; however, we believe that increased funding can advance the startup to April 1979. We hope to demonstrate both the technical and economic feasibility of the BERC process through the operation of a successful plant to bring about a timely transfer of our technology to private enterprise. If our plans succeed, we believe that our country will benefit from both the conservation of a valuable resource and from reduced environmental pollution.

Conclusions

A new and improved process for re-refining used automotive lubricating oil to original quality has been developed at the BERC. Engine and fleet tests have shown the process to be technically effective, and an independent engineering study indicates the new process has an economic advantage over conventional acid/clay treatment.

Supporting research of a more basic nature is currently being conducted at the BERC, including oil composition studies designed to estimate compositional changes related to engine use, as well as re-refining by the BERC process.

A test method is currently under development that appears to correlate with coking and fouling tendencies of re-refinery feedstocks. The method is believed to have application in the evaluation of particular feedstocks, as well as indicating the effectiveness of the pretreatment process in removal of coking and fouling precursors from used lubricating oil.

Engineering studies are in progress to provide the basis for design, construction, and operation of a 10-million gallon per year demonstration plant to evaluate technical and economic viability of the BERC process in full-scale application.

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API ACTIVITIES IN USED OIL

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In preparing for this meeting, I took the logical step of looking back at what I said at the first National Bureau of Standards workshop just about one year ago. And, quite frankly, I was amazed at how little things have changed since then: the pattern of used oil reuse probably has not varied substantially; the "do-it-yourself" problem is still with us; and, finally, many of the issues are unchanged.

One of my reasons for being here today is to discuss what we in the American Petroleum Institute (API) are trying to do in the do-it-yourself area, and the other reason is to consider in some detail the issue of used oil burning and why the petroleum industry is so vitally concerned about this matter.

First of all, let me say that I do not believe anyone has any truly accurate data on the volume of do-it-yourself drainings; I know that I do not. But, based on recent surveys, the number would appear to be in the vicinity of 150 million gallons per year. This is a large number in itself; but what is worse, it is dispersed throughout the U.S. in place and time and is available only in parcels of about one gallon.

Despite the difficulties, many attempts are being made to recover this oil and many people are deeply involved, including the API. The Used Oil Management Task Force, which I represent today, is hard at work on the do-it-yourself problem; but it is a tough one. Some of the ideas that we are looking at involve the following.

1. An advertising program to educate people through mass media communication.

2. Finding a way of getting greater participation of over 150,000 independent businessmen who operate service stations in the U.S. We want them to willingly accept drainings from the do-it-yourselfers, but we cannot coerce them.

3. An API-sponsored and -funded test program to evaluate some ideas we have to create incentives for the do-it-yourselfers to bring their drainings to a collection point.

Our task is extremely difficult, but we are hopeful of having some impact in this area.

My major points deal with the very serious concern that the API and its marketing members have regarding the future disposition of used oil, specifically used motor oil, commercial crankcase oil, and other used oils that are difficult to reclaim or re-refine. But, first, let me present a few statistics (or should I say educated guesses).

1. Total domestic purchases of lube oil are currently about 2.4 billion gallons per year.

2. About half of this oil eventually becomes used oil looking for a home; say 1.2 billion gallons.

3. About 25 percent of this used oil is being reclaimed or re-refined for reuse as lubes.

4. About 10 to 15 percent is generated by do-it-yourselfers and is essentially lost from recycling.

5. It is likely that another 10 percent of used oil is lost from recycling by illegal dumping and through junking of oil-containing equipment, such as filters.

6. The remaining 50 to 55 percent, something over 600 million gallons per year, is used predominantly as fuel or in various means of dust control, such as road oiling. Please keep the 600-million-gallon figure in mind, because I will refer to it shortly.

Several arguments against burning have been raised, and I would like to comment on each one to a limited extent.

1. The major objection to burning used motor oil is that it causes the emission of metals into the atmosphere, particularly lead. This is true, but only about 50 percent is so emitted, even in the absence of stack control equipment. Furthermore, very extensive testing has shown that concentrations of lead in air at ground level can be less than 2 micrograms per cubic meter if normal averaging periods and wind direction statistics are considered. Finally, to be even-handed, one must recognize that recycling to lubes also involves a lead problem, although in that case the concern is primarily real or potential water pollution.

2. The second argument is that burning used oil is inefficient in terms of energy conservation. I have personally spent many hours in studying this proposition, and I must say that analysis is extremely difficult because of the variability of input data on virgin lube processing and re-refining energy consumption. My best evaluation is that there may be a slight, almost insignificant, energy advantage for lube recycling over burning.

3. The third argument concerns the unfortunate conclusion that the U.S. is running out of potential lubricating oil feedstocks. This is just not true. If the demand existed and if refining capacity were available, feedstocks, or molecules, would be available to produce lubes at a rate several times what they are now. There is no doubt that they would cost more because the yields would be less. But they could be produced, and this situation will not change in the future. Just to put this matter in perspective, the U.S. consumes about 16 million barrels per day of crude for all purposes, but only 1 percent goes into lubes.

4. The fourth argument that I have heard mentioned is that used oils, particularly used motor oils, contain polynuclear aromatics (PNA's) and that they could be emitted to the atmosphere during burning. In looking into this matter, I found very few available data; in fact, only four for used motor oil. At drain intervals of 4,000 to 8,000 miles, analysis indicated the presence of active PNA's at concentrations varying from 30 to 65 ppm. By active PNA's, I am referring to compounds such as benz(a)anthracene and benzo(a)pyrene. However, a single data point available on a Bunker C fuel oil showed 330 ppm of active PNA's to be present. More data are being obtained, but the indication so far is that used motor oil is "cleaner" than heavy fuel oil. Therefore, substituting used oil for fuel actually appears to reduce the potential PNA problem.

You will recall that I suggested a figure of about 600 million gallons per year of used lube oil being consumed today in dust control applications and fuel. If we can ever solve the do-it-yourself problem and also recover the oil lost by illegal dumping and junking of oil-containing equipment, we will add another 250 million gallons to the used oil pipeline, making a total of about 850 million gallons per year to be recycled in some way.

Re-refining has been proposed as a viable and effective alternative to burning and, presumably, dust control uses. But according to a study recently completed for the Environmental Protection Agency, the current re-refining capacity is less than 100 million gallons per year of production. With an average yield of 65 percent, this capacity reflects a used oil containment of 150 million gallons per year. This is a relatively small number compared to the 850 million gallons that must be contained if burning and road oiling were prohibited.

Worse yet, this same study forecasts a 1983 re-refined lube production of 183 million gallons, if several factors will be conducive to greatly expanded re-refining capacity. One of the factors affecting the growth of re-refining is the matter of by-product disposal. In used oil burning, some metals are emitted with the flue gas; but in re-refining, the same metals are concentrated in acid or caustic sludge or in still bottoms. According to the recent Environmental Protection Agency study, about 80 percent of re-refiners dispose of sludge in landfills or on roads without any form of treatment. Although improved technology is being developed, recycling of these materials is not economically attractive at the present time. Perhaps this problem can be overcome. But, the projected optimistic volume of 183 million gallons relfects a used oil feed of 280 million gallons per year in 1983. This capability is a lot better than the 150 million today, but it is seriously deficient.

Burning used oil to recover the inherent fuel value is a very useful recycling method that has not yet been proven to be a significant environmental hazard and can substitute for the lack of re-refining capacity. No new investment is required now or in the future, and fuel-consuming equipment is available in every part of the country. It would be disastrous to stop burning; and yet we see an apparent desire on the part of many in government to do just that. As I have tried on numerous occasions to communicate, this matter is of the utmost concern to the API and its members. We see no significant environmental problem with burning, and no viable alternative seems to be in sight.

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VOLUNTARY TEST METHOD AND SPECIFICATION STANDARDS (ASTM) FOR PETROLEUM PRODUCTS FROM WASTE (USED) OIL

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The aim of this presentation is not to define the merits of voluntary standards. It is assumed that the merits are well known and recognized as pillars of our democratic systems. Neither is there an intent to extol the virtues of the American Society for Testing and Materials (ASTM), an organization pre-eminent in the field of voluntary standards. Rather, the aim is to describe what the ASTM Committee D-2, a technical committee dealing with petroleum products and lubricants, is doing in the field of standards for waste oils and recycled petroleum products and how it is planning to broaden these activities.

Many, indeed most, of the ASTM standards under the jurisdiction of D-2 developed for crude petroleum and virgin oils are already applicable to waste oils and recycled petroleum products. Many are in use for this purpose. When special needs have arisen, these have been handled by appropriate divisions in D-2. Three divisions are now involved: Technical Division B on Automotive Oils, Technical Division N on Hydraulic Fluids, and Research and Development Division IX on Oxidation.

Division B has a study group considering the equivalency of recycled engine oils to virgin engine oils. Largely it is monitoring the work of the Environmental Protection Agency/U.S. Army study, under which many re-refined base oils were evaluated and the best selected for blending with additives to make a number of experimental recycled engine oils for engine sequence tests in accordance with API [American Petroleum Institute] service classification SE. This part of the project is nearing completion.

Division N is seeking a short-term oxidation stability test for antiwear hydraulic fluids, including recycled fluids. Currently, the RBOT [rotating bomb oxidation] test, ASTM D 2272, is being evaluated, but other tests may also be tried.

Division IX is attempting to develop a short-term test for oxidation response of anti-oxidants in both virgin and recycled oils. The variety of anti-oxidants, in-house tests, test conditions, and oil types will tend to make progress slow. Even initial selection of a combination of additives, basestocks, and tests is difficult. Any advice in this area would be greatly appreciated.

None of these studies so far are leading to the elimination of engine sequence tests for engine oils or long-term oxidation tests for other oils.

Because of the very broad nature of waste oil and recycled oil product studies leading to standards, consideration is being given to establishing a new division under Committee D-2 to deal both with development and with coordination. Coordination will be very important because advances made in virgin oil technology will apply to recycled oil technology. In fact, those divisions in D-2 having responsibility over product types oriented towards application and having appropriate expertise, in all likelihood, will be asked to develop the specification standards so as to avoid duplication of effort and confusion.

A recent survey of members of the ASTM Committee D-2, petroleum re-refiners, government agencies, and others revealed a strong support for increased activity in Committee D-2 and the formation of a new division. There was some disagreement as to whether the new division should be technical (i.e., a product division) or solely coordinating. Committee D-2 will sort this out at its Dallas meeting next month (Appendix II). An interim summary of survey results has been prepared and a copy is provided (Appendix III).

In 1975, at the Second International Conference on Waste Oil Recovery and Reuse sponsored by the Association of Petroleum Re-refiners, a suggestion was made that a new division on recycled oils in Committee D-2, if formed, could be made up of five sections to deal with (a) properties of finished oils and basestocks, (b) properties of waste oils, (c) properties of fuel oils, (d) properties of road oils, and (e) test techniques [1].[1] This arrangement is believed workable; but, in the light of survey returns, some alterations would appear appropriate. Even these can be further altered before and even after implementation. The five sections could comprise (a) properties of recycled lubricants (largely a coordinating activity, as appropriate product divisions will probably be developing the standards which largely already exist based principally on virgin oils); (b) properties of recycled basestocks; (c) properties of waste oil feedstocks; (d) properties of fuels (coordinating probably, as Technical Division E has jurisdiction); and (e) properties of miscellaneous products.

Committee D-2 in the past has not concerned itself to any significant degree with railroad oils and aviation engine oils. These types of products, however, were covered in the survey. Recycled products such as railroad car journal box oils, locomotive diesel engine oils, and synthetic aviation turbine engine oils have been in use for many years, and it appears there is some interest in preparing voluntary standards for them.

Other categories, including possibly new types of recycled products, will also have to be handled in some manner. Road oils to suppress dust, cement kiln fuel oil, acid sludge and other re-refinery residues, waste oil fuel blends for diesel engines, recycled synthetic lubricants, waste synthetic engine oils, and solvents are a few such products.

Assuming that new standard test methods, possibly standard segregation, collection and handling practices, and product specifications and classifications will have to be developed, the new division can look forward to a long-term undertaking. Coordination not only within D-2, but with other outside organizations, can also be unwieldy and time consuming. Squeezing in additional meetings into the 200 regular technical meetings per week held during the semi-annual D-2 sessions will also be a strain.

However, if it is the wish for all to pull together, in all likelihood, the outcome will be crowned with some and possibly much success, provided the wish is sincere and is translated into participation.

References

[1] Strigner, P. L., ASTM Committee D-2 and the petroleum re-refiners industry (Proc., 2nd International Conf. on Waste Oil Recovery and Reuse, Feb. 24-26, 1975).

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[1] Underlined numbers in brackets indicate the literature references at the end of this paper.

SESSION VII. PANEL DISCUSSION

"WHAT TECHNICAL DATA BASE IS REQUIRED TO ESTABLISH THE 'SUBSTANTIAL EQUIVALENCY' OF RE-REFINED MOTOR OILS TO VIRGIN MOTOR OILS?"

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National Bureau of Standards Special Publication 556. Proceedings of a Workshop on Measurements and Standards for Recycled Oil - II held at NBS, Gaithersburg, Maryland, November 29 and 30, 1977. (Issued September 1979)

ESTABLISHING SUBSTANTIAL EQUIVALENCY FOR MOTOR OILS

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As the first speaker on the panel, I would like to discuss some of our concerns in the area of test procedures for gasoline and diesel engine lubricating oils. I would also like to ask some questions which we believe should be addressed by the industry (and on which we hope other panel members will comment), as well as direct our attention to areas of concern of industry.

First I will briefly review some basic nomenclature for motor oils. Most of this material has been touched on at some point during the conference, but I believe it would be useful to briefly review it at this point.

Table 1. Engine Service Classification System (Automotive)

Service classification	Type of service
SA	Straight mineral oil
SB	Inhibited oil
SC	1964 MS warranty approved
SD	1968 MS warranty approved
SE	1972 warranty approved

Table 1 shows the American Petroleum Institute (API), Society of Automotive Engineers (SAE), and American Society for Testing and Materials (ASTM) engine service classification system for gasoline engine oils. Note that oil meeting SA quality classification contains no additives and is unsuitable for current gasoline engines. The SE grade, the most highly formulated in terms of additive content, meets present engine manufacturers' warranty requirements. What is not apparent from this table but which should be kept in mind is the SE classification. For example, this classification implies that the oil, if subjected to a prescribed and costly series of tests on laboratory engines, will achieve certain minimum ratings in terms of performance. Some of the performance parameters include engine sludge rating, varnish rating, bearing weight loss, etc. These tests are described in the SAE J183a standard and also in the ASTM engine sequence tests.

Table 2. Engine Service Classification System (Diesel)

Commercial classification	Type of service
CA	MIL-L-2104A
CB	MIL-L-2104A, Supplement 1
CC	MIL-L-2104B; MIL-L-46152
CD	MIL-L-2104C

Table 2 lists the commercial and fleet classifications or diesel engine service. Some oils are labeled as meeting both services, such as SE/CC or SE/CC/CD. Note that CC oils meeting MIL-L-46152 also meet SE, and CD oils meeting MIL-L-2104C also meet service SC. The sequence tests contain many different single- and multi-cylinder engine tests. Some of the tests are no longer available in the classes other than SE/CC/CD.

Modern engine oils must be capable of dealing with a complex mixture of contamination products. In addition to reducing wear and dissipating heat, the oil must deal with these products, some of which are listed in table 3. These products cause a variety of problems for the engine and require carefully selected additive packages for the base oils to deal with them.

Table 3. Contaminant Species in Gasoline Engine Oil [1][1]

Contaminant type	Chemical species	Source
Nitrogen oxides	Nitric oxide (NO) Nitrogen dioxide (NO_2)	Combustion (atmospheric nitrogen)
Sulfur oxides	Sulfur dioxide (SO_2) Sulfur trioxide (SO_3)	Combustion (fuel sulfur)
Hydrocarbons	Olefins ($R_2C{=}CR_2$) Diolefins ($R_2C{=}CH{-}CH{=}CR_2$) Aromatics (ArR) Saturated hydrocarbons	Fuel and precombustion products from power section Fuel from tank
Carbonyl, carboxyl compounds	Formaldehyde (CH_2O) Higher aldehydes (RCHO) Ketones ($R_2C{=}O$) Acids (RCOOH)	Partial combustion
Peroxides	ROOH, ROOR	Partial combustion
Lead salts	Lead oxide (PbO) Lead chloride ($PbCl_2$) Lead bromide ($PbBr_2$) Lead sulfate ($PbSO_4$) Lead nitrate ($Pb(NO_3)_2$) Organolead halides	Tetraethyllead decomposition
Soot	Carbon	Partial combustion
Water	H_2O	Combustion
Carbon monoxide	CO	Combustion
Carbon dioxide	CO_2	Combustion

To evaluate basestocks and formulated oils, a variety of laboratory tests are used. At the present time, however, an accurate prediction of performance appears to require both engine sequence tests and field tests. The Environmental Protection Agency/Department of Defense project described earlier on re-refined oils using existing methodology is one approach to the problem. What correlations exist and how good they are pose questions that should be considered. In considering these questions, the following aspects may be considered:

- Can physical and chemical tests adequately monitor the re-refined basestock output of a re-refiner?
- Can laboratory "bench tests" adequately monitor performance in certain areas?
- What does "substantially equivalent" mean for SA oils? For SE oils?
- Is the "additive response" of a basestock of serious concern for re-refined oils, and can it be adequately monitored with other than engine tests?

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[1] Underlined numbers in brackets indicate the literature references at the end of this paper.

To summarize the various aspects of this problem, it would appear to me that we need to determine how best to measure substantial equivalency in terms of tests defining equivalency for re-refined and virgin oils and to correlate these tests with actual performance.

References

[1] Kreuz, K. L., Gasoline Engine Chemistry as Applied to Lubricant Problems, Lubrication, 55, 53 (1969).

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ACTIVITIES OF THE SAE FUEL AND LUBRICANTS TECHNICAL COMMITTEE

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I would like to report to you the latest activity of the Society of Automotive Engineers (SAE) Fuel and Lubricants Technical Committee, and, as I am the official liaison between the SAE and the National Bureau of Standards Recycled Oil Program, I will report on this meeting to the next SAE Fuel and Lubricants Technical Committee.

One item of interest to this group is that the Fuel and Lubricants Technical Committee will request the American Society of Testing and Materials (ASTM) to develop a test method which incorporates high-temperature, high-shear viscometrics, or other rheological characteristics which predict the performance of both single and multigrade (i.e., both Newtonian and viscosity index improved) engine oils in engine bearings and/or the ring and cylinder area.

On September 8, 1977, the SAE wrote a letter to the ASTM and the American Petroleum Institute (API) officially requesting that a new gasoline engine oil category be established to include SE performance, plus "long drain" capabilities and new antiwear requirements. Previously, the SAE had two active task forces working towards two possible new categories: (1) a long-drain oil, and (2) oils having improved antiwear. The two task forces have been combined and now recommend a single new category. The SAE requests that (1) the ASTM develop the performance criteria for the proposed category, and (2) the API develop the user language. Another area of active programs is the viscosity classification task force under Subcommittee 2 of the Fuel and Lubricants Technical Committee. Samples are being collected from oil companies, including re-refiners, to determine what viscosity ranges exist within a given W-grade engine oil at temperatures other than -18° C. The need for such a survey arose from an earlier survey of major engine builders throughout the world to determine their low-temperature starting and pumpability using a single commercial oil of unknown (to them) viscosity characteristics. Tests are then conducted at a near-the-minimum recommended use temperature for the particular viscosity grade, rather than at 0° F (-18° C) where engine oils are currently classified. Because of the probable difference among oils at temperatures other than 0° F, the present classification may be limited in meeting the needs of the engine builders and the users. From the estimated 1,000 samples, a random statistical sample of about 100 oils will be selected for analysis. Analysis will be conducted by an independent testing laboratory at a cost of approximately $15,000. These tests will include CCS, as well as other pumping tests now being studied by the ASTM.

For the immediate future, the Fuel and Lubricants Technical Committee has requested that the units of centiposes (cP) and centistokes (cSt) be used for the viscometric unit of viscosity by the SAE until further clarification was obtained.

The ASTM requested that the SAE take necessary action to reflect the tentative 1H2 and 1G2 test passing limits; Mr. J. A. MacLean has agreed to review these limits early in 1978 and to provide Subcommittee 2 with a recommendation for the revision of J-183b. In addition, a new task force within the Fuel and Lubricants Technical Committee would be formed to recommend that action be taken on the request from the ASTM to handle absolute categories and test procedures. The task force will also consider what means should be used to keep categories current.

A new task force was formed entitled "Low Temperature Viscosity of Used Engine Oils." The report is due in March 1978.

A new task force on Fuel Saving Engine Oils has been formed; there is no report at this date.

At the last Fuel and Lubricants Technical Committee meeting, considerable discussion was generated about the Engine Oil Review Board. At the request of the Army, the SAE entered into a contract to host the next meeting of the board. A nonprofit organization under the SAE was established, named "Lubricant Review Institute." Basically, the engine builders have supported this activity and wish the review of new oils by a board for approval and to insure they meet the classification on military specifications. This matter is still to be resolved because the military needs may be different from those of the commercial market

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SOME VIEWS ON SUBSTANTIAL EQUIVALENCY

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I appreciate this opportunity to participate in the panel discussion and to present some views on the subject of substantial equivalency specifically related to the entire test program which was recently completed on six selected re-refined oils.

The purpose of the test program was to examine possible variations in re-refined oil basestocks and to estimate the effect of these variations on the ability of the blended product to qualify under existing U.S. military specifications. To accomplish this objective, six oils were chosen by a subgroup of the American Society for Testing and Materials (ASTM) Re-refined Oil Task Force and representatives from the Environmental Protection Agency and the U.S. Army. Six different re-refined lubricants were selected to represent quality levels ranging from "best" to "low."

These oils represented products of differing chemical and physical properties, and the purpose of selecting various quality levels was to determine the effect on overall engine performance. Two lubricants were represented as being of the so-called "best" quality, three in the middle category, and one representing the poorest or low level. Recommendations were then received relating to additive treatment. All six lubricants were formulated with a single additive package at a specific treating level and subjected to engine sequence and Caterpillar 1-H2 tests.

A review of the test data obtained indicates several interpretations are possible (table 1). The first, and perhaps most obvious, approach is to look at the data on an overall basis to determine whether or not a specific lubricant would meet the requirements of the MIL-L-46152 specification. Another approach would be to evaluate the lubricants on a pass/fail basis relating to each individual test which was conducted. Still another possibility would be to look at the overall program based on assessment of the individual performance criterion involved--specifically, such items as top groove filling, bearing weight loss, engine rust, wear, viscosity increase, sludge deposits, etc. Also, it would be of interest to look at the data and relate the results to the quality levels of the basestock which were assigned to these lubricants by the ASTM Re-refined Task Force.

Table 2 shows the finished oil inspection data on these six lubricants for additive component levels and viscosity levels prior to engine performance testing.

The first approach in interpreting the data relating to the MIL-L-46152 specification would indicate that only one of the six lubricants could be considered as meeting the qualification requirements of the specification. Reviewing the data on the basis of the number of tests involved, a somewhat different picture is found; namely, that, of the 6 oils evaluated which were run on 30 different tests, 25 of the tests met the MIL-L-46152 requirements and 5 failed to meet the acceptance limits. The 30 tests involved 114 different numerical requirements which the lubricants must meet. Reviewing the data on this basis, the 6 lubricants met 109 numerical requirements and failed 5.

Another possible method of reviewing the test data on a pass/fail basis pertains to a comparison with the quality levels assigned to the basestocks by the ASTM Re-refined Task Force (table 3). Viewing this test program in this fashion shows that all of the oils passed the L-38 and IIC tests. The one lubricant, L-852, meeting MIL-L-46152 performance was considered to be in the middle category with respect to base oil quality. The two oils represented as being "best" failed the 1-H2 test.

A broad assessment of the test data points to the following potential strengths and weaknesses.

The strengths relate to IIIC deposit control, both from the standpoint of sludge and varnish as well as oxidation resistance as measured by the viscosity increase in

Table 1. Engine Test Data

Quality level: Re-refined base (CCL-):		Best L-853	Best L-863	Middle L-846	Middle L-850	Middle L-852	Low L-848
Test results	MIL-L-46152 Requirements						
Cat. 1-H2, % TGF	45 Max.	12	10	9	24	2	1
WTD	140 Max.	188.3	155.2	80.4	176.0	67.8	73.6
L-38, Mgs, Wt. loss	40 Max.	8.2	20.9	28.4	14.9	4.8	22.0
Piston varnish	9.0 Min.	9.7	9.7	9.7	9.7	9.7	9.7
IIC, average engine rust	8.4 Min.	8.8	8.8	8.6	8.7	8.9	8.5
Lifter sticking	0	0	0	0	0	0	0
IIIC, engine sludge	9.0 Min.	9.7	9.7	9.8	9.6	9.7	9.8
Piston varnish	9.3 Min.	9.5	9.4	9.3	9.5	9.4	9.2
Ring land varnish	6.0 Min.	8.1	8.1	8.0	7.6	8.3	8.1
Viscosity increase, 100° F, 40 hours	400% Max.	69	62	105	126	88	111
Viscosity increase, 100° F, 64 hours	No req't.	114	103	166	4,774	118	173
VC, engine sludge	8.5 Min.	9.5	9.5	9.5	9.5	9.5	9.4
Engine varnish	8.0 Min.	8.4	8.3	7.8	8.0	8.2	8.1
Piston varnish	7.9 Min.	8.2	7.9	7.9	7.9	8.2	8.0

Table 2. Finished Oil Inspection Data

Re-refined base CCL-	L-848	L-863	L-846	L-850	L-852	L-853
Calcium, % wt.	0.22	0.22	0.214	0.214	0.219	0.22
Zinc, % wt.	0.137	0.137	0.134	0.137	0.138	0.138
Phosphorus, % wt.	0.123	0.123	0.123	0.122	0.122	0.122
Viscosity at 100° F						
cSt	86.47	61.17	78.92	71.83	78.96	92.38
SUS	400	283.7	366	333	366	428
Viscosity at 210° F						
cSt	10.14	8.05	9.34	9.18	9.94	10.67
SUS	59.82	52.62	57.1	56.5	59.12	61.7

Table 3. Base Oil Quality Versus Performance

		Performance in:		
Base oil number	Quality level	1-H2	IIIC	VC
L-853	Best	Fail	Pass	Pass
L-863	Best	Fail	Pass	Pass
L-846	Middle	Pass	Pass	Fail
L-850	Middle	Fail	Pass	Pass
L-852	Middle	Pass	Pass	Pass
L-848	Low	Pass	Fail	Pass

All oils passed L-38 and IIC tests.

the IIIC test. The only possible exception here is with oil CCL-850, which gave a viscosity increase at 64 hours of 4,774 percent with an acceptable answer at 40 hours of 126 percent. This was the only lubricant which gave excessive viscosity increase even at 64 hours. The other oils ranged from 103 to 173 percent at 64 hours. All of the lubricants also exhibited acceptable valve train wear control as measured by cam and lifter wear in the IIIC test. Without exception, all of the lubricants exhibited satisfactory IIC antirust protection.

With respect to the L-38 test, although there was some variation in the bearing weight loss, the results indicate comparable performance from the standpoint of adequate copper-lead bearing corrosion protection.

The VC test results also showed satisfactory performance from the standpoint of overall sludge and varnish deposits.

The potential weaknesses of the formulations tested appear to be in the area of 1-H2 lacquer deposits where three of the six lubricants failed to meet the U.S. military MIL-L-46152 requirements. There also appears to be some indication of possible marginal performance with respect to piston varnish in the VC test.

The test program involving these six lubricants has provided a substantial amount of useful and significant information. However, in any program of this type there are obvious limitations. For example, it was felt necessary in this program to utilize a single additive system, as well as a given additive treatment. In addition, due to cost limitations, a single evaluation was made on each test procedure which does not allow assessment of test repeatability.

To summarize, we believe the test data developed demonstrate the capability of these six lubricants to meet the military specification requirements with a given additive system. As we have shown, there are several ways to interpret the data, any one of which may have equal validity. Also, the data suggest that re-refined oils do have satisfactory performance potential to be utilized in formulating lubricants to meet the U.S. military specification requirements. The test data obtained relate to a single additive type and treating level; therefore, the conclusions reached must be confined to a discussion of the re-refined oils within these parameters.

It is well known that to obtain satisfactory performance potential on re-refined oils it is necessary to establish adequate quality control of raw materials, as well as re-refining and blending processes. This can, however, be accomplished and has been satisfactorily demonstrated by the program established and operating in South Africa under the South African Bureau of Standards.

To achieve the maximum amount of information relating to the performance differences in re-refined stocks, the present program should be expanded to include other types of additive chemistry and variations in additive treating levels. This, we feel, is particularly important to adequately determine and define the performance of individual re-refined basestocks.

Significant progress has been made to achieve the original purpose of this program, which was to "examine variations in re-refined oil basestock and to estimate the effect of these variations on the ability of the blended product to qualify under existing military specifications." Additional work, however, needs to be done, and it is recommended that the responsible groups review the progress made to date for the purpose of setting up additional test programs to fully accomplish the program objectives.

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FIELD EXPERIENCES WITH RE-REFINED MOTOR OILS

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At the previous oil workshop held at the National Bureau of Standards (NBS) on November 22 and 23, 1976, Mr. Hugh Kaufman indicated he felt that only a few people wanted this country to recycle oil continuously [1][3], and this disturbed him since the Oil Producing and Exporting Countries (OPEC) were meeting on December 15, 1976, to determine how much to raise the price of oil. Today, in my opinion, very little has changed in the overall attitude toward recycling oil, and OPEC is meeting again to consider a price increase, while the current import price tag of oil to the U.S. is $45 billion per year.

In two papers given at the 1976 workshop, the several authors listed the primary barriers to oil recycling as (1) the Reduction Excise Tax, and (2) the Federal Trade Commission's labeling requirement (both instituted in 1965) [2,3]. The result of these was an enormous drop in the number of re-refiners' and the total volume of recycled oil output.

Recently, I made a telephone survey of 12 re-refiners, selecting those companies with a large portion of their production going into motor oils. The purpose was to obtain more recent information about barriers they face, and to also obtain information about their customers and thereby accumulate evidence that a number of re-refiners have been making high-quality motor oils for many years. From this survey I identified three barriers which these re-refiners feel constitute a severe constraint on their industry: (1) unfair taxation, (2) restrictive labeling requirements, and (3) limited customer acceptance. Seven of the companies contacted do the major part of their business competing with the lower-grade virgin oil products, where price is critical to moving the product. They market through wholesalers and jobbers and generally sell an API SA class product. This segment of the re-refining industry is currently very severely affected by these three barriers. First, in order to increase their product line by using virgin blending oils, they must face tax disadvantages. Second, these same re-refiners are unable to successfully compete in the premium-quality product market because their SE classification oils are subjected to the labeling requirement and must be labeled "made from previously used oil." This labeling requirement helps to bring about the third barrier, customer acceptance, because it gives customers the impression that the re-refined oil is an inferior product, regardless of actual quality. Now that you have heard the reports on the Environmental Protection Agency/ Department of Defense program and the Iowa State fleet test on re-refined engine oils [4,5], I feel that it is time these barriers be reconsidered. Although the labeling requirement may give the impression that these re-refined oils are inferior products, some of these same companies provided oils for inclusion in these evaluation programs, and the results have indicated that these oils are, in fact, quality products.

In addition to motor oil production, 3 of the 12 re-refining companies surveyed process railroad lube oils, recompounding their products to original specifications. They have been successful in this business for 25, 7, and 6 years, respectively. One of these companies has served 19 railroad systems, several of them for the entire 25 years.

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[1] Author was APR/NBS Industrial Research Associate, working full time at NBS for a year beginning June 1977.

[2] The opinions expressed in this paper are those of the author.

[3] Numbers in brackets are references, to be found at the end of the paper.

In order to minimize the customer acceptance problems, most re-refiners have generally operated by dealing directly with larger volume consumers, and they supply oils which are compounded as suggested by representatives of additive companies or as requested by customers. The following is a condensed listing of the information supplied by re-refiners pertaining to customers using their products:

- Company A has been supplying a 200 vehicle bus line for more than 6 years. It has also supplied oil for municipal vehicles for several years in a city with a population of approximately 700,000.
- Company B re-refines to produce quality products. For 5 years it has supplied oil to a large interstate trucking company with 110 tractor units. There have been no technical studies on the engines, nor any measurements made on parts from engines. However, their maintenance people indicated that the engines are visually cleaner using re-refined oils with a very noticeable improvement in the rocker box area. Another interstate trucking company with 25 tractors has been using this re-refined oil for 3 years, and 10 small trucking companies with 1 to 3 trucks each have been customers for the past 8 years. Company B has also supplied lube oil to a city cab company for 4 years.
- Company C supplies a company with a fleet of more than 3,000 mixed vehicles. For the past 19 years, this company has preferred to purchase and use re-refined oil. The fleet manager expressed his confidence in the re-refined lubricating oils his company uses. Engines opened after 100,000 to 125,000 miles show minimum sludge deposits and virtually no engine wear. Most units are being run on 10,000- to 12,000-mile oil changes. One of these units was opened up and critically examined. This engine still retained the honing marks from manufacturing after it had been in service for over 100,000 miles. All other engines examined visually were found to be clean. Company C also has 40 smaller customers with only trucks or mixed vehicles which have used re-refined oil for 2 to 15 years.
- Company D has supplied a large food distributor with oil for more than 14 years. The distributor's large diesels are run on an 18,000-mile oil change schedule, and they have gone more than 500,000 miles on this oil. The man in charge of this fleet is satisfied with this re-refined product. They do not have engine problems attributable to oil, and all opened engines are free of sludge and varnish deposits. Two large food chanins have also been using this re-refined oil in their trucks and car fleets for more than 5 years. Company D has supplied one cab company for over 12 years and a second very large cab company for more than 4 years. The delivery truck fleets of two large department stores have been using re-refined oil for over 4 years.
- Company E produces mainly motor oils. The following is a partial listing of its customers: a trucking company with about 22 tractors for the last 11 years, 3 trucking companies with 20 units for over 7 years, two school districts with 10 vehicles for over 4 years, a highway department with 30 mixed vehicles for over 3 years, an equipment company with 25 units for more than 4 years, 2 construction companies with 100 pieces of equipment for over 3 years, and a mining company with 50 units for more than 3 years.

All of the above companies supply fully formulated oil products to their customers. The products are formulated as required to meet the equipment manufacturers' recommendations or customers' specifications.

The survey data presented here give significant evidence of customer acceptance of re-refined oil. Although these companies produce quality products and deal directly with consumers, they remain subject to the FTC labeling requirement. In my opinion, it is due time that these current restrictive labeling requirements be reconsidered, and that the quality of the product to perform the intended function be the overriding consideration in any labeling rule.

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AN APPROACH TO QUALIFICATION OF RE-REFINED ENGINE OIL UNDER MILITARY SPECIFICATIONS

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The recent Energy Policy and Conservation Act of 1975 and Resource Conservation and Recovery Act of 1976 call for revised policies to encourage the procurement of recycled oil for military and nonmilitary Federal uses. This legislation required removing the current restrictions imposed on re-refined materials in military specifications. Therefore, a methodology must be developed which will permit the use of these materials while maintaining the high quality of products supplied to the Federal Government under the military specifications. The following will be a discussion of a possible approach which could be used to meet these objectives. As we will see, this approach is based on the current system of qualifying products and would revise the procedures used to define and control the basestock components employed in qualified products. I will first briefly review current procedures and then compare these with the proposed system.

Prior to qualification, military specifications place only general requirements on the materials to be used in formulating the lubricant and the properties to be exhibited by the finished product. As shown in table 1, basestocks are to consist of a petroleum, synthetic, or combination petroleum-synthetic material; are to be treated with functional additives such as detergents, dispersants, oxidation inhibitors, etc.: and no re-refined materials may be used. In respect to properties, limiting values are established for viscosity; viscosity index; and flash, pour, and stable pour points. At the same time, very stringent requirements are placed on the performance characteristics of the finished product. Requirements are established for stability and compatibility; foaming characteristics; and, of course, the performance characteristics defined by the engine dynamometer tests. After qualification, the latitude allowed in formulating the lubricant is removed. The manufacturer is restricted to the same basestocks and additives as those used for the qualification sample. In addition, manufacturing tolerances specific to the finished product are established. These tolerances (table 2) are more restrictive than the general requirements set prior to qualification and include limits for both physical and chemical properties of the lubricant.

Table 1. Materials Requirements Before Qualification

<u>Materials</u>

- Basestocks: petroleum, synthetic, or combination petroleum-synthetic
- Functional additives: detergents, dispersants, oxidation inhibitors, etc.
- No re-refined constituent materials

<u>Properties</u>

- Viscosity (at various temperatures)
- Viscosity index (some grades)
- Flash point
- Pour point
- Stable pour point (some grades)

<u>Performance</u>

- Stability and compatibility
- Foaming characteristics
- Engine performance testing

Table 2. Materials Requirements After Qualification

<u>Materials</u>

- Basestocks: same source and refining treatment as used in the qualification sample
- Additives: identical as used in the qualification sample

<u>Finished oil tolerances</u>

- Viscosity
- Viscosity index
- Pour point
- Flash point
- Gravity
- Carbon residue
- Foaming
- Elemental constituents
- Sulfated ash

Now that we have reviewed the current system, let us look at the proposed methodology which would permit the use of re-refined materials (table 3). Again, prior to the qualification, the same type of approach would be used to define the lubricant. The composition of the oil would be stated in the same general terms, except that the restriction on use of re-refined materials would be removed. Likewise, the same property and performance requirements as currently used would be incorporated into the proposed system (table 4). So far, the changes have been minimal and rightly so, because one is working with a specific batch or sample of oil which is being tested to determine its acceptability in respect to the specification requirements. The question arises, that after qualification, how can one control the consistency of the finished lubricant? Again, the current type of approach with more specific controls on basestock components could be used. As we see in table 5, the proposed system would require subsequent batches of oil to be manufactured from basestocks which are of the same type and/or source and had been subjected to the same refining, re-refining, or processing treatment as those used for the qualification sample. In addition, they would have to meet manufacturing tolerances established to define the physical and chemical characteristics of the basestock used in the formulation. Likewise, as with the current system, oils would have to be formulated with additives identical to those used in the qualification sample, and finished products would have to meet predetermined tolerance limits.

Table 3. Materials

Component	Current	Proposed
Basestock	Petroleum-synthetic combination	Same as current
Additive	Functional detergents, dispersants, inhibitors, etc.	Same as current
Re-refined	Not permitted to be used	Delete restriction

Table 4. Properties and Performance

Item	Current	Proposed
Property	Viscosity, viscosity index, flash point, pour point, stable pour point	Same as for current qualification
Performance	Stability and compatibility, foaming characteristics, engine performance	Same as for current qualification

Table 5. Materials and Finished Oil

Component	Current	Proposed
Basestock	Same source, same refining	Same type/source, same refining, re-refining, or processing, tolerances for physical and chemical properties
Additive	Identical as used in qualification sample	Same as current
Finished oil	Tolerances for: Viscosity Viscosity index Carbon residue Sulfated ash Elemental constituents Gravity Pour point Flash point Foaming	Same as current

It is realized that additional work must be undertaken to select the physical and chemical properties and tolerance criteria which will adequately define a basestock. However, it is also believed that this methodology is the only realistic means to meet the required objectives of allowing the use of re-refined materials and maintaining product quality.

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PERFORMANCE TESTING IS THE KEY TO ENGINE OIL QUALITY

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We are here today to discuss a very difficult question, "What technical data base is necessary to establish substantial equivalence of re-refined and virgin oils?"

There seems to be no argument among technically qualified people that a re-refined engine oil <u>can</u> be produced that is equivalent to a virgin engine oil. However, there may be disagreement as to how such equivalency can be demonstrated.

Evaluation of Engine Oils

One approach to answering the foregoing question is to first consider the evaluation of virgin engine oils. The evaluation procedures can be subdivided into those associated with physical properties and those associated with performance qualities. The physical properties of high- and low-temperature viscosity, pour point, foam inhibition, and miscibility, to name a few, are important requirements for an engine oil. They can be defined by straightforward physical analysis or bench testing.

Although several areas of engine oil performance qualities are covered in the current classification system, the system is not all inclusive. This system, which categorizes oils by performance, came into being in response to field problems which developed when oils were classified by words rather than by performance. The laboratory-engine tests used in this system were first introduced in 1958 and evolved to tests shown in table 1. These tests are used to define the SE/CC-quality level or MIL-L-46152 requirements.

Table 1. Engine Sequence Tests

Tests	Performance area tested
Sequence IIC	Low-temperature rusting and corrosion
Sequence IIIC	High-temperature oil thickening and deposits
Sequence VC	Low-temperature sludge and varnish formation, and screen clogging
L-38	Bearing corrosion
Caterpiller 1H	Light-duty diesel piston deposits and ring sticking

Each of these tests evaluates a specific performance area. The Sequence IIC test examines an oil's ability to provide adequate low-temperature rust and corrosion protection, while the Sequence IIIC measures resistance to high-temperature oil thickening and deposits. In the Sequence VC test, an oil's low-temperature screen clogging tendency, and sludge and varnish formation are measured. The single-cylinder L-38 engine test measures an oil's resistance to bearing corrosion. The Caterpiller 1H test evaluates a light-duty diesel engine oil's performance with regard to carbon and lacquer formation.

These tests were never intended to be "all-proof" tests or to replace field testing. They test only the specific performance areas outlined. In addition, correlation of these tests with field experience is based upon known engine oil chemistry. The use of new chemistry of basestocks may alter this correlation.

These laboratory engine tests can provide only an indication, not a guarantee, that an oil will give trouble-free consumer service. Depending entirely on engine-dynamometer tests is tantamount to driving a dynamometer down the road. As yet, nobody has figured out how to drive a dynamometer. The real test is the automobile! Therefore, it is necessary that oil and additive companies field test their products before they are marketed, and it is expected that they will continue to do so.

Field Problems With Oils That Pass Engine-Dynamometer Tests

Despite the development work and care taken in conducting engine-dynamometer tests, it is possible for an SE-quality oil which passes all the tests to give unacceptable field performance. Such undesirable results can occur if: (1) the oil involved has a quality shortcoming in a performance area not adequately evaluated in the engine-dynamometer tests, or (2) the oil is formulated with either additives or basestocks significantly different from those used in establishing the correlation between the engine-dynamometer test and field service.

One brief example will illustrate. In February of this year, Pless and Rodgers of General Motors (GM) Research Laboratories reported (1)[1] that excessive valve train wear occurred in both customer service fleets and in controlled fleet tests with some commercial SE-quality engine oils in severe type service. In four customer service fleets, the wear problems were reflected in the number of camshafts replaced by fleet operators (table 2). Three of these fleets involved heavy-duty type vehicles and one involved light-duty type vehicles. In addition, the authors reported results of six controlled fleet tests in general transportation, police, and taxi services which verified the lack of adequate wear protection with some oils, despite their satisfactory performance in laboratory tests. These tests provided the necessary field data upon which to base an engine-dynamometer wear test.

Table 2. Wear in Customer Service Fleets (1)

Vehicle duty type	Oil service classification	Number of engines	
		Total	Camshafts replaced
Heavy	SE/CC	11	10
	SE/CC	83	18
	SE/CD	193	41
Light	SE/CD	37	9

In response to this field problem, an American Society for Testing and Materials (ASTM) Ad Hoc Wear Panel was formed, and work was begun at the GM Research Laboratories on an engine test to determine the wear protection provided by engine oils. In June of this year, General Motors reported a new wear test procedure to the ASTM (2).

We hope that the wear test will eliminate one deficiency in the current system. But this situation clearly illustrates that laboratory tests are not the final determining factor in the performance of engine oils. Field tests and field experience are the ultimate standard of performance.

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[1] Underlined numbers in brackets indicate the literature references at the end of this paper.

Field Test Design

Since laboratory engine tests are not sufficient to completely define engine oil quality and field tests are necessary, the guidelines for field test evaluations should be defined. Just as no single laboratory test can examine all performance areas, neither can one field test provide an all-inclusive test of engine oil quality. Each test must be designed to examine specific performance parameters. The test must be severe enough to discriminate between good and bad performance. This testing is expensive and time consuming, but it is advisable with unproven lubricant technology to ensure that field problems do not occur.

The "Aunt Minnie Rust Test" is one type of field test upon which Sequence IIC [5] is based. The objective of this short-trip service rust test is to examine the ability of an oil to provide rust protection under severe operating conditions. An examination of the test design requirements as outlined by Pless [3,4] gives an idea of the amount of careful planning and control which is a part of field testing. The test site, make and model of car, reference oils of known short-trip performance, and the use of leaded fuel must all be considered. In addition, there are many details of the test which must be controlled. These include preparation of cars for the test, mileage accumulation, intermediate and final inspections, as well as performance criteria.

Obviously, there is a considerable amount of work involved in establishing the quality of oils in just one performance area. The information gained in ongoing field-test programs is valuable in two ways. First, it makes sure that oils perform satisfactorily in the field. Second, it allows engineers to revise or design sequence tests to provide the best possible correlation between engine tests and field experience.

Application of Virgin Oil Tests to Re-Refined Oils

The problems associated with testing virgin engine oils and classifying them according to performance are very difficult. But equipment manufacturers have confidence in the existing classification system because they know what performance range to expect from products in a given category. Re-refined engine oils represent an uncertain quality level. There is very little data on their performance, and they may represent a significantly different chemistry in terms of additive response. They should be examined in controlled field test situations, as well as in engine and bench tests before their widespread use in SE-quality engine oils is adopted. This testing will protect current and future engines, as well as ensure that re-refined engine oils do not get a bad reputation as a result of marketing an inferior product with too little testing.

Variations in Re-Refined Oil Quality

While there are general similarities in the processes by which used engine oils are re-refined, the need for holding this conference dramatizes the lack of uniform standards for re-refined oils. Each manufacturer is producing a product which may differ from others in its additive response and engine performance. As stated by the American Petroleum Institute in its explanation of the engine service classification system [6], it is the responsibility of the oil manufacturer to establish that the oil has the performance characteristics essential for the service classification for which the oil is recommended. This must apply to both virgin and re-refined oils.

There is an additional concern that batch-to-batch variations in the production re-refined basestocks may significantly affect performance. Some work has been done by Whisman, et al., on possible differences in the feedstock to re-refineries [7], but their survey did not examine possible performance differences which could be related to batch-to-batch variations in re-refined oils.

Today's performance-based classification system has been developed because of the need to assure that high-quality engine oils are available to the public. The current and future efforts of vehicle manufacturers to conserve our petroleum resources by extending the oil-change intervals, while maintaining engine durability, are dependent on the availability of even higher-performance engine oils. Until the validity of nonengine tests has been proven in the evaluation of re-refined engine oils, any attempt to replace the performance-based classification system by one based on bench or physical inspection tests would be a step backward.

Conclusion

In conclusion, the following observations can be made: (1) field testing provides the basic definition of lubricant quality, (2) field problems can occur in performance areas where testing has not been adequate, (3) re-refined oils should meet the same performance tests as virgin oils, and (4) added quality-control testing may be needed for re-refined oils.

A good start has been made toward establishing the technical data base necessary to establish substantial equivalence of re-refined and virgin engine oils. The work by the Bartlesville Energy Research Center, the National Bureau of Standards, and the joint Environmental Protection Agency/Department of Defense re-refined oil program are important first steps. However, more data are needed to establish equivalency. These data must not only be on oils from different sources, but also on oils from several different batches to demonstrate the ability to produce a uniform product of consistently high performance. The data obtained should include carefully controlled field testing under a variety of conditions, as well as laboratory engine test results and the results of other laboratory and bench tests.

General Motors encourages the recycling of used oils as an important means of conserving our vital resources and protecting our environment. Whether the most energy-efficient, cost-effective way of recycling used oil is to subject it to some mild reprocessing for use as fuel or industrial oils, or to completely re-refine it so that it is suitable for use as engine oil can be decided only after more data are obtained. In any recycling program, of course, the consumer must be protected by standards which measure the ability of the recycled product to perform its intended function.

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RE-REFINED OIL IN OUR FUTURE

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We have been invited here today to share our views on the technical basis that should be used to judge the adequacy of re-refined base oils. It is clear from recent developments that re-refined oils will become a significant factor in the U.S. domestic base oil supply. A principal driving force, of course, is a portion of the Energy Policy and Conservation Act. Our joint efforts have been focused on counseling the National Bureau of Standards in defining the technical basis for judging the "substantial equivalency" of re-refined and virgin base oils so that the law may be implemented. As a result, the long-standing barriers to the sensible use of re-refined oils in routine commercial application should finally be brought down.

Our company has been actively interested in the use of re-refined oils for a number of years. You may recall that last year Mr. Floyd Sam of our research organization reported on the rather extensive test work evaluating the use of re-refined oils in high-performance engine lubricants.

I would like to digress a bit from the technical aspects of the subject to comment on another important factor affecting the use of re-refined oil. The principal deterents to our company's proceeding with the program were the restraints of the required labeling and the product economics. The laws of many States (28, I believe) require that the use of re-refined oils in a product be clearly stated on the package. Marketing research studies, conducted for our company, have clearly shown that, in the marketplace, the stigma of "re-refined oil" demands a significant discounting of the product sales price to make the product acceptable to the customer. On the other hand, in our experience re-refined base oil cost approaches the cost of virgin base oil. When the evaluation costs are considered, finished product costs may exceed those using virgin oil. Under this set of conditions, economic incentive will be marginal at best.

An important provision of the new law is the stipulation that the Federal Trade Commission establish labeling standards which will preempt any State and local regulations which influence the acceptability of the product in the marketplace.

I had not intended to make more than passing reference to this labeling deterent. However, from the discussions yesterday and this morning one wonders if sufficient attention is being given to the marketability or customer acceptance of the products containing re-refined base oils. Despite all technical assurances as to equivalency, literally branding them as different will impose second-class status and inhibit their acceptance. This clarification of labeling is critical to the orderly, logical use of re-refined base oils in high-quality lubricants. While on the subject of deterents, it is hoped that the tax regulations which limit the use of re-refined oils in "off highway" and agricultural service by denying tax rebates will also be reviewed.

Our company is prepared to use re-refined oil in the manufacture of high-quality engine lubricants. Assuming that the restrictions mentioned are indeed removed, we see no barrier to the broad use of properly re-refined oil in automotive crankcase lubricants. In our case, we would initially concentrate on the use of re-refined base oils in this type of product. The rather specific and unique base-oil-related performance characteristics of many industrial oils would require much more extensive testing. This additional test cost burden probably cannot be justified as long as the amount of re-refined oil available can be accommodated in automotive crankcase lubricant production.

The intent and spirit of the law is to in some way provide the necessary quality assurances so that re-refined oils from any and all qualified re-refined oil systems may be used without concern as to finished oil quality. Perhaps what we have done here these two days will provide a basis for developing suitable, simple criteria. I am sure it is Mr. Becker's fond hope that it shall.

I would like to review what our company feels is an appropriately cautious approach to making the use of re-refined oils a routine, commercially viable operation. The following points are considered important.

1. Initially, we would limit our interest and study to re-refined oil from a refining group capable of supplying assured base oil volumes sufficient to justify the cost of the test program that we feel would be required to protect the quality image of our company's branded products in which the re-refined oils would be used.

2. Representative re-refined base oil samples will be subjected to conventional characterization tests. This would include physical tests, as well as proprietary bench tests. These would serve as reference for the future ongoing monitoring of the re-refined oil production. We will speak later on the re-refined base oil quality control.

3. A laboratory engine test program would be conducted to verify the suitability or to highlight significant weaknesses of the base oil as used in our branded product line. Some experts consider bench test procedures to be adequate for base oil evaluation. We have greater confidence in the engine test approach. We are always receptive to new evaluation procedures and will adopt them as they become proven.

A basic engine test program would be along these lines--using our normal additive systems in re-refined base oil formulations.

a. A CC/SE oil would go through

    (1) modified 1-G-2;
    (2) 1-H-2; and
    (3) sequences IIC, IIIC, and VC

b. A CC oil would be run in the modified 1-G-2 test.

c. An L-38 would be run on a gas engine oil formulation.

d. Counterpart current production products would be run as reference.

4. On successful completion of the laboratory engine test program, we would proceed to a field test of the CC/SE oil. This would probably be in a taxicab fleet test, again with appropriate reference.

When the adequate performance of the re-refined base oil is demonstrated in this test program, we would be prepared to use the oil in routine crankcase oil production.

As mentioned earlier, we would expect to deal with each re-refiner separately so that we may understand his situation and develop confidence in his operation as a routine base oil supply source. If problems arise which may relate to some aspect of the re-refining operation, we would expect to work jointly in an effort to control the matter. We have a worry, shared by others, that re-refined oil quality may vary considerably, both physically and as regards additive response. The potential variation in the feedstock used by a re-refiner is the principal basis for this concern. We have all seen data demonstrating unusual uniformity of re-refined oil production from a given facility, but can we depend upon it? We feel that a monitoring program providing physical checks on each shipment from the re-refiner should be established. Certain data obtained on the oil originally tested would be used as "specification" values, with realistic ranges applied. A periodic composite sample accrued from deliveries over a period would be evaluated for additive response in a suitable bench test.

As our experience and that of the industry with the application of re-refined oils is shared in meetings such as the one we are attending today, general confidence will develop and successful utilization of re-refined oils will be assured.

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SUMMARY OF DISCUSSION

SESSION VII. PANEL DISCUSSION ON THE TOPIC, "WHAT TECHNICAL DATA BASE IS REQUIRED TO ESTABLISH THE 'SUBSTANTIAL EQUIVALENCY' OF RE-REFINED MOTOR OILS TO VIRGIN MOTOR OILS?"

This panel was composed of seven invited participants. Each of the participants made a brief presentation on the above subject, and these presentations are included in these Proceedings, immediately prior to this summary of the discussion. [Note: An eighth invited participant, Mr. William Crouse (Suntech; also chairman of the ASTM Tech B Used Oil Task Force), was not able to attend the conference at the last minute due to illness.] The discussions included here were between individual panel members, as well as from the floor, and covered the primary topic of the panel discussion as well as a variety of related topics. A summary of the discussion follows.

The first discussion topic was on the quality level of virgin motor oils currently in the market place. Reference was made to analyses made by several organizations which suggested that some virgin oils labeled SE (or equivalent), may in fact not stand up to close examination. The point was made that the quality of motor oils is not being monitored in the marketplace, and very little is really known about the quality level of such oils actually in the marketplace. One person proposed that continuous surveillance of motor oils in the marketplace be initiated, and the results tabulated and published. However, it was not stated who should be doing this monitoring, government or industry. Finally, the comment was made that a double standard seems to exist at the present time -- there is much concern about the "quality" of re-refined oils, but very little being done about identified problems in the virgin oil field. This individual felt that any requirements placed on re-refined oils must be applicable to virgin oils also.

There was also considerable discussion on the topic of engine sequence tests and field testing of motor oils. Much of this discussion was in regard to the presentation made by the General Motors representative, as part of this panel. A question was asked whether field tests had been added to the requirements for the SE classification. The response was that the SE label <u>implies</u> that field tests have been made, but that requirement is not included specifically in the written description of the SE (or any other) classification. It was acknowledged by persons present that <u>not</u> all virgin oils in the marketplace have been field tested (many, especially from major oil companies, have been), and furthermore, it was also acknowledged that field problems have later developed with oils that had been field tested -- possibly due to changes in engine metallurgy, etc. Finally, it was again emphasized that any requirements placed on re-refined oils should apply as well to virgin oils.

The topic of the lower quality motor oils, the SA, SB and/or CA classifications, were brought up for discussion. These oils are non-detergent oils, have no performance requirements (or minimum requirements), yet represent a substantial portion of the motor oil market, estimated at 15-20%. NBS has been asking the question, to what should these oils be equivalent? Engine manufacturers state such oils should not be used for modern automobiles, except perhaps for "clunkers" that use large quantities of oil. The only response from the panel and from the floor was that SAE has a Task Force addressing this question, and that API is revising the whole motor oil classification system, with new recommendations for SA type oils, in 6-9 months.

There was much interest and discussion on the EPA/DOD program which put six re-refined oils through the engine sequence tests necessary for the MIL-L-46152 motor oil specification. One question was concerned with whether this EPA/DOD program had developed an adequate data base for the NBS "substantial equivalency." A number of responses indicated that it was very helpful, but the question of variability of re-refined oil had not been adequately addressed. One representative from an engine manufacturer stated that there still was not enough data to definitively state that engine sequence tests developed for virgin oils can be effectively used to evaluate re-refined oils.

Additional discussion on the EPA/DOD program questioned whether any bench oxidation tests were run on the re-refined oils (RBOT or other tests -- answers: No), and why didn't the "best" base stocks (as selected by the ASTM Tech B Task Force) show up as "best" in the engine sequence tests (answer: don't know)? It was suggested that the EPA/DOD testing program should be expanded to do additional testing with different additive levels and different additive chemistries, including multiviscosity additives to produce multigrade oils. Finally, the question was asked, how many times are the engine sequence tests normally run on virgin oils to obtain the required passes for the MIL specs. The response was that the military qualification board does not normally ask for this information, but is becoming more inclined to do so in the future. It was also stated that the entire motor oil system, especially the API/ASTM/SAE classification system but also the military system to some extent, is based "on the integrity of the people and the organizations participating in those systems...And it seems to work reasonably well."

There was some discussion on the capability of re-refiners to maintain adequate quality control of their products. Re-refiners were asked which quality control tests they normally ran. The response was that some re-refiners who normally sell in the quality motor oil market have extensive testing capabilities. Others, who sell parimarily in the SA motor oil market, have rather limited quality control facilities, but some of them expressed interest in upgrading their quality control should they be able to enter the quality oil marketplace. In addition, some testing of re-refined oil is done for the re-refiners by additive companies. A question to Phillips Petroleum about their use of infrared (IR) scans to monitor the constancy of their re-refined oil basestock, brought the response that IR is one technique used which, although not adequate by itself, does provide useful information.

The last area of discussion was on the NBS responsibilities, and how they were going to complete them. A particular concern expressed by a re-refiner was that costs of engine sequence tests were so high, they were looking for NBS to provide bench tests to replace the sequence tests. The NBS response was that total reliance on bench tests was unlikely, at least in the near future. There apparently are some bench tests which correlate well with an engine sequence test, but these are proprietary, and not available for evaluation at present. One possibility for the re-refined motor oils, is to establish reasonable lubricating oil basestock specifications, and then determine acceptable tolerances for the re-refined oil within these specifications. This would require extensive data on the variation of re-refined oil basestock, and detailed evaluation on which tests are important for characterization of re-refined lube oil basestock. This approach might provide an acceptable method for re-refined lubricating oil to be accepted into the quality motor oil market, while still assuring adequate performance to the consumer.

In conclusion, in spite of the extensive discussions held, there seemed to be no consensus on the specific data base required for "substantial equivalency." However, most participants seemed to feel that more information and data on re-refined oil was needed in order for it to be effectively utilized in the quality motor oil marketplace.

Acknowledgements

The editors wish to thank the authors for their assistance in bringing this useful and timely information to the attention of all the conference attendees. Also, we wish to thank all participants in the lively discussions, both formal and informal, for their sizeable contributions to the overall success of the meeting.

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APPENDIX I

METALS IN OIL: OCCURRENCE AND SIGNIFICANCE FOR REUSE OF SPENT AUTOMOTIVE LUBRICATING OILS

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1. Summary and Conclusions

The objectives of this study are:

1. to summarize and evaluate available data on the occurrence and on the chemical and physical forms of metals in used lubricating oils, and

2. to evaluate the implications of the metal content of used lubricating oils to their use and disposition.

A considerable body of information is available concerning the levels of metals in used oils. The principal sources of metals in these materials are: (1) metal-containing additives incorporated in essentially metal-free basestocks, and (2) metals accumulated during use, primarily lead from gasoline additives and iron from wear processes. The most significant change in metal content during the service life of lubricating oils is the accumulation of lead, which increases in concentration by a factor of 10,000 to 1-1.4 percent (w/w).

The specific chemical and physical forms of metals in used oils are largely unknown. It appears, however, that much of the lead (in the range of 50 percent)--and possibly other metals--is present in the form of fine suspended particulates.

Primary applications of spent lubricating oils are : (1) re-refining, (2) combustion, and (3) uses for road oiling. A significant fraction is currently disposed of by landfilling.

Re-refining technology reduces metal levels to values substantially equivalent to those of virgin basestocks. While re-refining residues containing high metal concentrations present a disposal problem, metals per se do not appear to present a problem for the equivalency for automobile use of re-refined oils to virgin materials.

Metallic contaminants probably do not affect either combustion efficiency or short-term integrity of combustor components when used oil is burned, particularly if it is used diluted with fuel oil from virgin sources. There are well-defined chemical mechanisms by which metallic impurities can adversely affect refractories and metals in combustion applications, but such effects have not generally been observed in pilot studies. Long-term studies are probably required, however.

The principal problems associated with the high levels of lead and metallic additives in spent lubricating oils are environmental, specifically emissions from combustion and uncontrolled release to the environment in road oiling and dispersion through landfill operations.

It is a primary conclusion of this study that research is needed to better characterize the nature of metallic contaminants in spent oils. Specifically, it is necessary to evaluate the distribution of metals among water-soluble, oil-soluble, and particulate constituents. Identification of specific chemical forms in which metals occur is required, as are data on size distribution and metal distribution in particulate fractions. These data are needed to support continuing development by the re-refining industry of effective and economical methods for removal of metals from used oils. Further, they can help provide a scientific basis for required labeling standards (Energy Policy and Conservation Act, P.L. 94-163, Section 383(d) (2)) designed to reduce environmental hazards associated with disposal of oils after use.

Acquisition of these data presents significant measurement problems in the chemical characterization of amorphous particulates and in the speciation of soluble and metal-organic compounds occurring in spent oils.

Needs for field data identified in this study include: (1) information on mass balances for toxic elements other than lead (barium, for example) in combustion of used oils, (2) more extensive field studies of toxic metal mobilization from oils dispersed from roads or landfills, and (3) evaluation of processes in which toxic metals are retained in refractory products.

2. Introduction

2.1 Resource potential of used oil.

Of the approximately 2.2 billion gallons of lubricating oil sold annually in the United States, about half is consumed in use, discarded with filter cartridges, or lost through leakage or in other ways. Of the remaining 1.2 billion gallons of potentially reusable waste oil, automotive crankcase oil represents 600 million gallons, and aviation and industrial lubricating oil make up 400 million gallons [23].[1] It is estimated that, of this total, 600 million gallons are burned as fuel, 200 million gallons are used as road oil or are incorporated into asphalt, and 110 million gallons are re-refined. The remaining 290 million gallons, otherwise unaccounted for, are presumably disposed of by dumping, pouring into sewers or landfills [1].

Although the total volume of used oil represents only about 1 percent of U.S. petroleum consumption, it is a potentially recoverable resource. Used oil production amounts to 78,000 barrels per day, about 1.5 percent of 1976 imports of crude oil. In the Energy Policy and Conservation Act of 1975, the Congress has specifically recognized the resource recovery potential of used oils and has assigned to the National Bureau of Standards (NBS) the task of developing equivalency test procedures to facilitate oil reuse.

Utilization of this resource, however, has significant environmental implications. Potential gaseous and particulate emissions from burning used oils must be considered in the context of present and prospective air quality standards. Direct dispersion of used oils as road oils or asphalt components provides a mechanism for uncontrolled distribution in the environment of constituents, particularly hydrocarbons and metals.

These interrelated problems are addressed in section 383(d) of the Energy Policy and Conservation Act, which calls for labeling standards designed to reduce environmental hazards and wasteful practices associated with disposal of oils after use. Thus, it has become important to identify specific environmental impacts potentially associated with reuse and disposition of spent oils.

2.2 Significance of metals in oils.

As described subsequently in this report, used oils may contain substantial quantities of metallic contaminants. The amounts and chemical forms of these metallic constituents are quite different from those in so-called virgin oils. To the extent that they influence the properties of the oil, the metals may affect equivalency of used and virgin oils in specific applications. Moreover, because of the well-known toxicity of heavy metals, their occurrence in used oils can directly affect the environmental impact of the disposal, reuse, and recycling of these materials.

Although oil itself is biodegradable, the metallic components in used oil may not be detoxified by biotransformation. Similar problems may be associated with disposal of re-refining process residues. While the status of used oil under the Toxic Substances Control Act is undetermined at present, potential risks to health or to the environment in some applications of used oil clearly need to be assessed.

2.3 Scope of this study.

The implications of metals in used oils have not been explicitly assessed previously. This study will first survey what is known concerning the amounts of chemical forms of metals in oils, including used oils. It will then consider the implications of the metal content of used oils for end-use applications in re-refining, combustion, and use as road oils. Significant environmental consequences of metals in used oils will be discussed. Where possible, significant gaps in present-day knowledge concerning metals in oils will be identified, and recommendations concerning research needs will be presented. Figure 1 summarizes the fate of metals in waste lubricating oil and in reprocessing and reuse.

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[1] Underlined numbers in brackets indicate the literature references at the end of this paper.

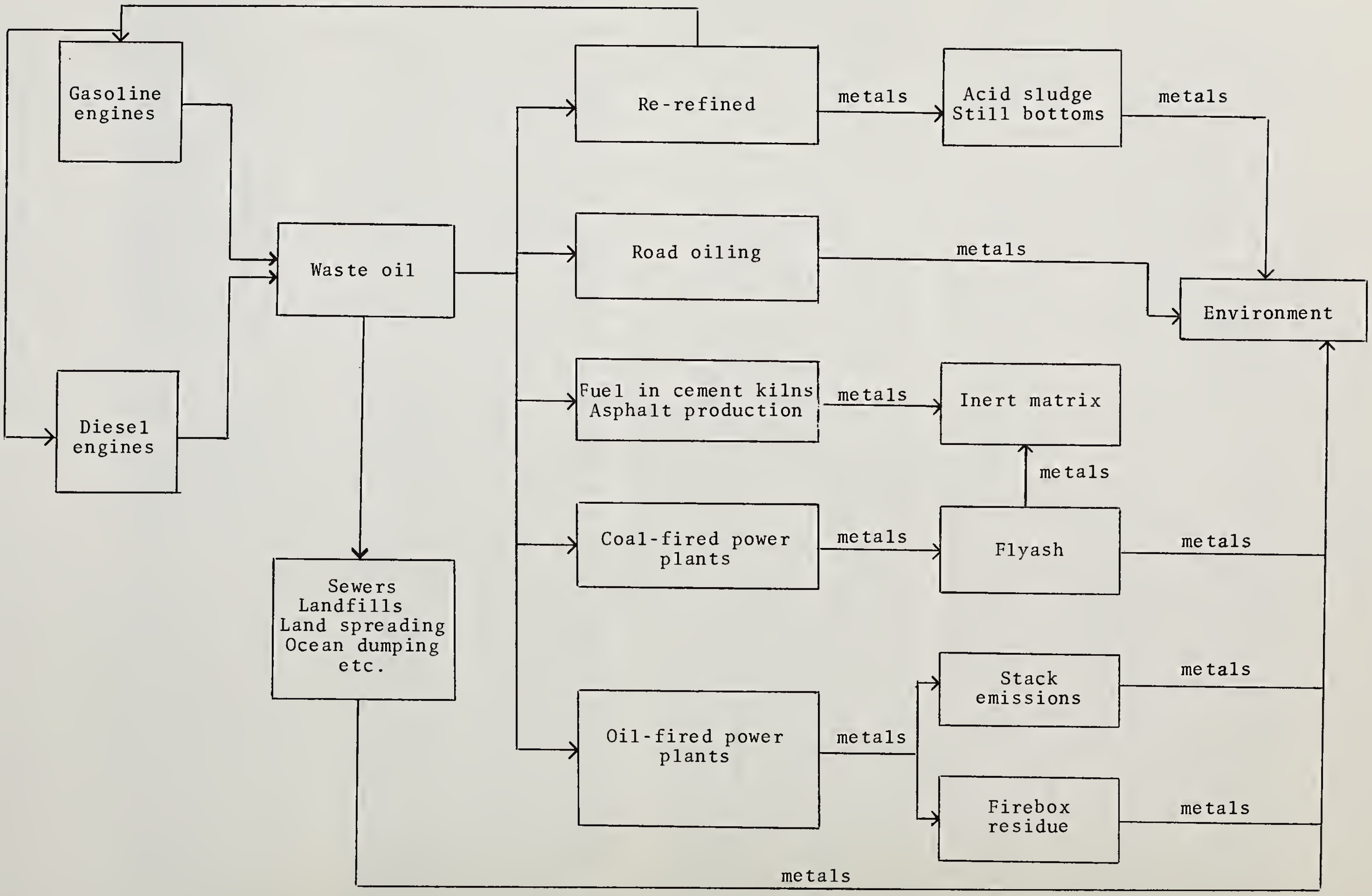

Figure 1. Fate of metals in waste lubricating oil.

3. Concentrations of Metals in Oil

Three general sources of metals in oil can be recognized: (1) naturally occurring metals, (2) metals incorporated in lubricant additives, or (3) metals introduced during use. This section surveys the quantitative data on the levels of metals in crude oil and virgin, in used oil, and in re-refined lubricating oil. In section 3, the literature on the chemical and physical forms in which these metals are found is discussed.

3.1 Concentration of metals in virgin and used oil.

Crude petroleum contains metals in low concentrations, principally zinc, iron, nickel, and vanadium. Typical concentrations are shown in table 1. The last three elements are present chiefly as very stable porphyrin complexes [2]. Since these metals can interfere with catalytic refining processes, they are removed during the refining operations. Consequently, in lubricating oil basestock, the product from which lubricating oils are formulated, almost all metals are present at negligible levels, i.e., 1 part-per-million (ppm) or less (see table 2).

Table 1. Metal Content of Crude Oils [2]

Element	Concentration range (μg/g)	Mean or median concentration(μg/g)
Antimony	0.03 - 0.1	0.055
Arsenic	0.056 - 1.11	0.26
Barium	Small amounts in Texas crudes	---
Cadmium	----	0.03
Chromium	0.0016 - 0.017	0.008
Cobalt	0.032 - 13.	1.7
Copper	0.13 - 6.3	1.38
Iron	3.4 - 12 .	41.
Lead	0.17 - 0.31	0.24
Manganese	0.63 - 2.54	1.28
Mercury	0.023 - 30	3.2
Molybdenum	0.008 - 0.053	0.031
Nickel	49.1 - 340.	166.
Selenium	0.03 - 1.4	0.53
Silver	Traces in many crudes	---
Tin	Identified in Mexican crudes	---
Vanadium	4.0 - 300.	89.
Zinc	3.6 - 86.	30.

In the formulation of lubricating oils from the basestocks, a number of metal-containing compounds are added to improve various lubricant characteristics. For example, a typical gasoline engine lubricating oil contains about 1 percent zinc dialkyldithiophosphates and 4 percent barium and calcium sulfonates. Table 3 shows a list of typical additives and the functions which they serve [3].

Table 4 [5] lists representative data on metal concentration in a commerical lubricating oil before and after automotive use. Relatively large amounts of barium, calcium, phosphorus, and zinc in the new oil reflect the presence of additives containing these elements. During use, the metal content of crankcase oil changes due to accumulation of wear debris (iron) and gasoline additives (lead). Breakdown of the lubricant additives, which change the chemical form of the metals, can also be expected both from tribochemical, i.e., friction-induced, processes and from reactions with water and acids in the oil. These changes are the result of dynamic processes. Factors such as intervals between drain times, extent of fuel leakage into the oil, and engine operating conditions may affect the overall metal profile.

Additional data from two separate references on the metal content of used oils are given in tables 5 [6] and 6 [7]. Differences between table 5, compiled in 1973, and table 6, unpublished product of a 1968 contract, may be due to increased interval between oil drains, decreased gasoline mileage, and changes in additive formulations in this period [6].

Table 2. Trace Metal Content[a] in Lubricating Oil Basestocks [3]

Element		245-type base oil[b]		ASTM no. 1 oil[d]
Name	Symbol	NBS[c]	analyses from supplier	
Aluminum	Al	1	0.05	<0.02
Barium	Ba	1	<0.08	0.36
Beryllium	Be	0.01	<0.02	<0.02
Boron	B	0.05	<0.04	<0.08
Cadmium	Cd	0.2	<0.08	<0.08
Chromium	Cr	0.05	<0.02	<0.02
Copper	Cu	0.05	<0.02	<0.09
Iron	Fe	1	0.10	0.12
Lead	Pb	0.1	0.10	<0.02
Magnesium	Mg	1	<0.02	0.06
Manganese	Mn	0.05	<0.08	0.25
Molybdenum	Mo	0.01	<0.05	<0.02
Nickel	Ni	0.01	<0.08	0.03
Silicon	Si	1	--	--
Silver	Ag	0.01	<0.02	<0.02
Sodium	Na	0.1	<0.08	<0.08
Tin	Sn	0.05	<0.05	<0.02
Titanium	Ti	1	<0.02	0.03
Vanadium	V	0.05	<0.08	<0.08
Zinc	Zn	5	<0.08	<0.02

[a] Micrograms element per gram of oil.

[b] Type 245 base oil supplied by Continental Oil Company, containing no additives.

[c] Actual concentrations are less than or equal to these values.

[d] Analyses done at NBS.

Table 3. Metal-Containing Lubricant Additives [3]

Metal	Representative compounds	Use
Antimony	Sb dialkyl dithiocarbamates	Antiwear, extreme pressure, and antioxidant additives in conventional and low-ash-type automotive crankcase oils, industrial and automotive gear oils, greases (amounts less than 1-3 percent)
Barium	Ba diorganodithiophosphates Ba petroleum sulfonates Ba phenolates Ba phosphonates or thiophosphonates	Corrosion inhibitors, detergents, rust inhibitor in automatic transmission fluids, greases
Boron	Borax, boric acid esters	Antiwear agents, antioxidant, deodorant, used in cutting oils, greases, brake fluid
Cadmium	Cd dithiophosphates	Additive in steam turbine oils
Chromium	Cr salts	Grease additive
Lead	Pb naphthenate	Extreme pressure additive in greases, gear oils
Mercury	Organic mercury compounds	Bactericide, e.g., cutting oil emulsions
Molybdenum	MoS_2-Mo dibutyl dithiocarbamate and phosphate	Extreme pressure additives, solid lubricants
Nickel	Cyclopentadienylnickel complexes	Antiwear agents, minimize carbon deposits, improve lubrication and combustion
Selenium	Selenides	Oxidation and bearing corrosion inhibitors
Tin	Organotin compounds	Antiscuffing additive, metal deactivators
Zinc	Zn diorganodithiophosphates Zn dithiocarbamates Zn phenolates	Antioxidant, corrosion inhibitors, antiwear additives, detergent, extreme pressure additives, in crankcase oils, hypoid gear lubricants used in greases, aircraft piston-engine oils, turbine oils, automatic transmission fluids, railroad diesel engine oils, differential and wet brake lubricants.

Currently, calcium- and magnesium-containing detergents are used as additives; lead and mercury derivatives are not commonly used in automobile lubricants.

Table 4. Metal Concentration in New and Used Lubricating Oil [5]

Element	Concentration (ppm) New oil[a]	Used oil[b]
Aluminum	1	14
Barium	162	150
Calcium	3,430	3,600
Chromium	0	10
Copper	<1	4
Iron	1	149
Lead	0	10,420
Magnesium	20	47
Manganese	<1	2
Nickel	0	2
Phosphorus	470	600
Potassium	7	7
Silicon	3	12
Sodium	5	18
Tin	0	10
Vanadium	0	0
Zinc	359	461

[a] SAE 10W30 service SE motor oil from a single manufacturer, purchased by Bartlesville Energy Research Center.

[b] Crankcase drainings collected during routine oil changes from automobiles using this same oil.

Table 5. Trace Element Content of Used Lubricating Oils [6]

Element	Parts per million Average	Range
Aluminum	16	10 - 30
Barium	269	10 - 900
Calcium	1,772	1,160 - 2,690
Chlorine[b]	1,350	
Copper	28	5 - 120
Iron	361	150 - 1,000[a]
Lead	15,240	9,600 - 20,700
Magnesium	155	10 - 420
Silicon	46	10 - 240
Sodium	64	20 - 110
Tin	30	
Zinc	1,111	560 - 1,610

[a] Given as 150 - 100 in reference 6; probably 150 - 1,000 is intended.

[b] From [27]

Table 6. Range of Trace Metal Content in Used Lubricating Oil[a] [7]

Element	Minimum	Maximum
Aluminum	10	800
Ash	0.03	3.78
Barium	10	2,000
Beryllium	6	6
Boron	3	20
Cadmium	4	4
Calcium	700	3,000
Chromium	8	50
Copper	5	348
Iron	50	2,000
Lead	800	21,700
Magnesium[b]	10	1,108
Magnesium[b]	5	10
Molybdenum	2	3
Nickel	3	30
Phosphorus	500	2,000
Silicon	10	875
Silver	1	1
Sodium	16	300
Strontium	10	30
Sulfur	0.21	0.65
Tin	5	112
Titanium	5	30
Vanadium	3	39
Zinc	300	3,000

[a] Sulfur and ash in % w/w, metals in ppm.

[b] Two sets of values for magnesium given in original reference. Lower values may refer to manganese.

Table 7. Metal Content of Acid Sludge (ppm)

Element	Anal. from ref. 7[a] Gasoline	Diesel	Anal. from ref. 6
Aluminum	140	40	560
Arsenic	---	---	45
Barium	1,300	400	740
Beryllium	---	---	0.1
Boron	50	40	18
Cadmium	---	---	9
Calcium	6,400	12,000	---
Chromium	50	190	28
Cobalt	---	---	0.8
Copper	40	40	190
Iron	1,100	500	2,200
Lead	20,000	1,000	19,000
Magnesium	1,000	70	---
Manganese	---	---	63
Molybdenum	---	---	18
Nickel	30	35	---
Phosphorus	4,300	1,000	1,700
Silicon	1,400	800	---
Silver	0	14	0.8
Sodium	4,000	200	---
Strontium	---	---	2.7
Tin	30	35	---
Vanadium	---	---	18
Zinc	2,100	200	2,100

("---" indicates data not available.)

[a] Source of the oils not specified in original reference.

Gasoline and diesel engine crankcase oils have quite different metal distributions. This is indicated in table 7 [6], which gives results of analyses for metals in acid sludge, the material precipitated from used oil by treatment with acid. The variation in lead content is noteworthy, for, in end uses such as combustion where the lead may be released into the environment, the diesel oils could be segregated. This may be relatively easy to do since diesel-powered vehicles often operate in fleets; however, the added cost of separate storage facilities might prove unacceptable.

In considering analytical results on oils, it should be borne in mind that used oil is a heterogeneous mixture with metal-containing particles held in suspension by detergent additives. In an oil heavily laden with sediment when the dispersing capacity of detergent is exceeded, these particles will tend to settle out. If they are not thoroughly redispersed, accurate analyses will be difficult to obtain [8]. Further, the viscosity of the oil can affect the results of metal analyses since it influences the amount of sample picked up on a rotating disc anode or nebulized into an atomic absorption spectrometer. In nearly all reports of analyses on used oil, the sampling procedures are not specified in sufficient detail to decide whether inaccuracies due to settling of particulare matter have been introduced.

In general terms, the major difference between new and used lubricating oils in terms of metal content is the accumulation of lead during use by a factor of up to 10,000 (except in diesel engine lubricants). Iron from engine wear debris also accumulates significantly. Smaller increases are found in chromium, aluminum, sodium, magnesium, and phosphorus.

3.2 Concentration of metals in re-refined oil.

Modern re-refining technology reprocesses waste oil by a combination of techniques, e.g., filtration, clay contacting, acid treatment, or distillation. Clay/acid treatment is the most widely used method at present. Table 8 presents unpublished analytical data on re-refined oils intended for use as basestock, obtained from 16 re-refiners [9]. These data indicate that the reprocessing techniques can be very effective in reducing metal content. Calcium, barium, zinc, lead, magnesium, copper, chromium, iron, sodium, silicon, and aluminum levels can be reduced to a few ppm's, comparable with commercial virgin basestock (see table 2). Vacuum distillation of used crankcase oil was reported in one case to yield a distillate of slightly higher lead content (13 ppm, see table 9) [10].

3.3 Summary

Metals in low concentrations, notably nickel, vanadium, iron, and zinc are found in crude oils. Their concentrations are greatly reduced during refining, and commercial basestocks contain only very small amounts of metals.

Compounds containing calcium, zinc, barium, and phosphorus are added to improve operating characteristics of engine lubricating oils. During use, lead from the fuel and iron from wear debris accumulate. Used oil, as presently found in commerce, therefore contains high levels of some metals (see tables 4, 5, and 6). Because of the toxicity hazard associated with lead, this element has justifiably received great attention in reuse applications of waste crankcase oil, as will be indicated in sections 4 and 5. Zinc and manganese (in high concentrations) and barium are, of course, also toxic, but the effect of these metals on reuse has received little attention. It has not been proven that procedures which reduce the emission or diffusion of lead into the environment are also effective with regard to these metals, however.

Available data on metal levels in re-refined oils indicate that re-refining processes reduce the metal concentrations to low levels, less than 1 ppm lead, for example. In this regard, the re-refined basestocks appear to be equivalent to virgin material for automotive use.

4. Chemical and Physical Nature of Metals in Oil

4.1 Overview and importance of the problem.

Information about the nature of metal-containing species in waste oil is inferential, primitive, and fragmentary. Most attention has been given to simply quantitating various elements, perhaps because a gross analysis is easy and speciation techniques were, until recently, very difficult. Information dealing only with

Table 8. Metal Content of Re-Refined Lubricating Oils [9]

Element,	Sample number[a]															
ppm	6688	6689	6690	6691	6692	6693	6694	6695	6696	6697	6698	6699	6700	6701	6702	6755
Aluminum	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50
Barium	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5
Calcium	10	10	< 5	10	111	10	< 5	15	< 5	44	< 5	16	< 5	75	< 5	< 5
Chromium	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5
Copper	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5
Iron	6	13	< 2	< 2	13	< 2	< 2	< 2	< 2	< 2	< 2	< 2	< 2	< 2	< 2	< 2
Lead	< 1	< 1	< 1	< 1	47	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
Magnesium	< 1	12	< 1	< 1	3	< 1	< 1	< 1	< 1	2	< 1	< 1	< 1	11	< 1	< 1
Silicon	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50	< 50
Sodium	< 2	< 2	< 2	< 2	9	< 2	< 2	< 2	< 2	< 2	4	5	5	5	< 2	< 2
Zinc	< 2	15	< 2	< 2	11	< 2	< 2	< 2	< 2	< 2	< 2	< 2	< 2	< 2	< 2	< 2

[a] These numbers identify separate samples but have no meaning in this review. Other data with lower detection limits on the same samples will be published elsewhere.

These data were obtained by the U.S. Army Fuels and Lubricants Research Laboratory under a contract with the U.S. Army Mobility Equipment Research and Development Command, Energy and Water Resources Laboratory, Fort Belvoir, Virginia, as part of an Army/Environmental Protection Agency interagency project.

quantities of metals is helpful and, for the most part, adequate to predict the fate of the metals in applications where thermodynamic conditions obtain. Thus, if used oil is burned in an oil-fired power plant where oxidizing conditions and temperatures of 1,000-2,000° F exist, metals will be converted to their thermodynamically most stable form (oxides), regardless of the form in which they were introduced.

Knowledge of the chemical and physical forms in which metals occur in oil is important, however, in at least two ways. First, the cost and effectiveness of recycling processes in removing metals from used oils will depend to some extent on the specific form of the metals in the oil.

Second, the precise form of metals in used oil will be significant from the viewpoint of impacts on the environment, where both thermodynamic and kinetic conditions prevail. The rates and mechanisms of chemical processes in which metals are introduced into the environment, e.g., leaching and microbial transformation, are expected to be sensitive to the molecular form in which a metal is contained. In the case of particulate matter, the size of the particles will also be significant. It is reasonable to anticipate, therefore, that data on size distribution of metals in particulate matter and on the molecular identity of oil- and water-soluble metal-containing species in used lubricating oil will be of practical importance in determining how and how fast the metals are spread throughout the environment.

Both the accumulation of metal-containing species and their subsequent chemical transformation are time dependent. Longer intervals between oil drains will lead to higher lead levels. At the same time, the antiwear additives will have decomposed to a greater extent since their function is to provide, through decomposition reactions, low-friction films on moving parts. Rates of chemical reactions in the crankcase will depend on temperature and, therefore, on climate and the length of time a particular vehicle is operated after the engine has warmed up. Data on quantitation and speciation of metals are usually obtained on an average basis from pooled samples. Tables 5 and 6, for example, represent samples of oil obtained from several vehicles. Such data, however, are probably representative since nearly all potential end uses for used crankcase oil require pooling small volumes from single vehicles.

4.2 Survey and evaluation of the literature.

Because so very little is known about the chemical nature of metal-containing species in used oil, it is necessary to draw inferences from the fragmentary data that do exist.

At a recent workshop on recycled oil, Mr. Belton R. Williams [10] presented data from his re-refining operations which are useful, despite the lack of a mass balance, in partially characterizing some of the metallic species. Water (which arises as a product of fuel combustion, condensation, and seepage into storage tanks) is separated from the oil prior to filtration and distillation. Table 9 [10] shows that this aqueous phase contains large amounts of sodium, zinc, barium, calcium, iron, phosphorus, magnesium, boron, tin, and some lead. The water content of used lubricating oil ranges from 0 to 16.8 percent, with a typical oil containing 4.4 percent water by volume [6]. Assuming no significant introduction of metals from extraneous sources, these data suggest that the metals mentioned occur to a significant extent in water-soluble forms.

In another study dealing with the toxicity of waste oil to marine life, the oil was extracted with water (9:1 ratio for 18 hours). Table 10 indicates the metal content in the water phase, which contained large amounts of lead and zinc. Filtration studies indicated that more than 90 percent of the lead, cadmium, and zinc in the water passed through a filter with a pore size 0.05μm or less in diameter [11]. In this particular study, the waste oil was collected and stored in a barrel. Samples were withdrawn from time to time in an unstated manner. If sampling was done from the top, then the water and some of the particulate would have settled to the bottom, and this then corresponds to a second water extraction. If sampling was done from the bottom, then the sample would have been enriched in water-soluble metals and sediment. The significance of the results obtained is somewhat obscured because of ambiguities in the sampling procedure.

Appreciable amounts of water and sediment in oil can cause fouling of handling facilities: sediment can clog transfer pipes and burner nozzles when the oil is used as a fuel. Water may cause corrosion of tanks and pipes [12] and fuel line freezing. Consequently, water and sediment are usually discarded before waste oil is reused or processed further. In a review of disposal of recovery process residues, Mr. V. S. Kimball [13] indicated that the water layer is admixed with other waste water and is discharged into sewers while the sediment is, like sludge, disposed of by landfilling. This reference indicates that the principal concern in waste water quality is the content of phenols and other organic compounds: little attention has been given to the metals contained in waste water. However, the data in tables 9 and 10 suggest that toxic metals, particularly lead and zinc, may be discharged along with the water into the environment, although the quantities involved are not known.

Vacuum distillation of dehydrated crankcase drainings affords a product with a low metal content (table 9) [10]. The data indicate that some lead and phosphorus are present as volatile species and that the other metals occur in forms with little or no volatility. The lead in the distillate could either be entrained or present as tetraethyllead. The latter possibility might appear unlikely since tetraethyllead is decomposed by heating with aqueous acid, organic acids, and phenols [14]. Unpublished results indicate that entrainment is involved, for controlled laboratory vacuum distillations produce lead- and phosphorus-free distillates [45]. The nature of these lead species could probably be determined with modern chromatographic-atomic absorption instrumentation. Some phosphorus in the distillate may be present as volatile phosphate esters contained in the additives. The distillation residues are rich in metals (table 11) [10] and present a disposal problem.

Organic bromine compounds are added to gasoline to scavenge lead as lead bromide, and it has been suggested that some lead dibromide may be present in used oil. However, x-ray emission spectroscopic analysis of several samples of crankcase oil disclosed a lead/bromine ratio of 0.62-0.68:1 [15]. If all the lead were present as the dibromide, a 2:1 ratio would obtain. However, chlorides or mixed salts may be present.

It has been asserted that waste crankcase oil contains 1 to 3 percent particulate matter less than 2μm in diameter, which is principally carbon, calcium, lead, and phosphorus [16], although no data were shown to support this assertion.

Ultrafiltration studies in which used oil was passed through poly(acrylonitrile) membranes of unstated porosity have been reported [17]. The filtration process was found to result in substantial removal of metals and in a surprising reduction of ash content (table 12). Unfortunately, the nature of the pretreatment referred to in the table was not discussed in the text of reference 17. The use of physical techniques, settling, and centrifugation to reduce metal levels has been studied, and the results are summarized in table 13 (solvent extraction refers to diluting the used oil with hexane and 2-propanol, followed by centrifugation) [5]. The data indicate that this simple strategy achieves an order of magnitude reduction in lead content. The use of methylethyl ketone to precipitate and coagulate lead has been described in a recent patent [43].

The results of these gravity-assisted processes (i.e., filtration, settling, centrifugation) show that significant quantities of metals, and particularly about 50 percent of the lead, occur in particulates.

Analysis of oil samples taken from diesel-powered trains indicated that the "sludge" contained an average of 3 percent iron, mainly in the form of fine splinters from roller bearings [18].

The chelating agent EDTA (ethylenediaminetetraacetic acid) has been used to extract metals from waste oil [5]. In these experiments, one volume of oil was treated with eight volumes of 2:1 2-propanol-1-butanol to precipitate sludge which was removed by decantation and centrifugation. After distillation of the alcohol, the oil was diluted with toluene and 2-propanol and was extracted successively with aqueous disodium EDTA and water. While reduction in the levels of barium, calcium, magnesium, zinc, and lead were observed, a control experiment in which water alone was used was not reported. Thus, it is difficult to tell how much of the metallic components were removed by reaction with the EDTA and how much were removed by extraction with the water. The same workers [5] have described the use of acidic ion exchange resin columns to remove metals from used oil which was first treated with alcohol to precipitate sludge. No analyses of the oil after just the alcohol treatment were reported, and so it is difficult to judge the effectiveness of this treatment. Ordinarily, one would assume that the extraction procedure using a water-soluble chelating agent was simply effecting a phase transfer of an oil-soluble metallic compound into a water-soluble EDTA complex. This may be true for detergent additives containing calcium, magnesium, and barium. In the case of lead- and zinc-containing solids, the high interfacial tension at the surfaces of small particles may lead to simple dissolution of the particles in the presence of a solubilizing agent.

Table 9. Spectrographic Analysis of Used Dehydrated and Distilled Crankcase Oil (ppm) [10]

	Raw drainings	Dehydrated drainings	Vacuum distilled	Typical water
Aluminum	33	28	2	74
Barium	200	160	4	230
Boron	52	38	1	440
Cadmium	2	0	0	37
Calcium	1,000	1,100	0	2,200
Chromium	27	23	0	10
Copper	50	40	6	18
Iron	750	720	2	1,050
Lead	7,800	6,900	13	550
Magnesium	430	400	0	1,000
Manganese	42	39	3	65
Mercury (ppb)				0.88
Molybdenum	7	6	0	28
Nickel	3	2	0	26
Phosphorus	900	800	50	2,100
Silicon	47	40	4	100
Silver	0	0	0	4
Sodium	160	140	6	8,000
Tin	20	17	0	340
Titanium	5	1	0	45
Vanadium	5	4	3	20
Zinc	1,100	1,000	2	1,300

Table 10. Concentration of Metals in a Water Extract of Waste Lubricating Oil [11]

Metal	Concentration, µg/L
Arsenic	< 300
Cadmium	60 ± 2
Chromium	< 140
Cobalt	0.4 ± 0.4
Copper	< 100
Iron	< 100
Lead	4,400 ± 200
Manganese	< 100
Mercury	< 0.1
Molybdenum	< 200
Nickel	< 100
Selenium	< 100
Zinc	16,000 ± 1,000

Table 11. Analysis of Vacuum Still Bottoms From Crankcase Drainings [10]

Metal	Concentration, ppm
Aluminum	230
Barium	1,400
Boron	10
Cadmium	0
Calcium	7,000
Chromium	100
Copper	160
Iron	2,000
Lead	15,000
Magnesium	1,300
Manganese	70
Molybdenum	10
Nickel	10
Phosphorus	2,000
Silicon	550
Silver	0
Tin	140
Titanium	70
Vanadium	0
Zinc	3,500

Table 12. Effect of Ultrafiltration on Metal Content of Used Oil [17]

Metal content ppm	Used oil	Ultrafiltrate	Ultrafiltrate from pretreated oil
Ash, %	1.24	0.13	0.02
Barium	1,150	< 50	<10
Calcium	460	60	40
Copper	35	13	< 5
Iron	200	< 5	< 5
Lead	620	154	< 5
Magnesium	85	< 5	< 5
Phosphorus	520	385	<10
Silicon	35	23	36
Zinc	540	50	<10

Table 13. Effect of Physical Treatments on Ash, Sediment, and Lead in Waste Oil [5]

	Concentrations				
Treatment process	BS&W[a] % v/v	Water % v/v	Sediment % v/v	Ash[b] % w/w	Lead[b] % w/w
No treatment	10	8	5	3	1
Settling pretreatment	1	0	2.5	2.3	0.9
Centrifugation	1.5	1	1.7	1.5	0.75
Solvent extraction	0.3	0	0.3	0.3	0.1
Vacuum Distillation	0	0	0	0	0

[a] Bottom sediment and water.

[b] Referred to as v/v% in reference 15 and assumed to be w/w%.

Fractionation and infrared analysis of an SAE 30 additive-containing lubricating oil indicated that, after use in an automobile over 3,500 km, calcium dialkylnapthalene sulfonates were converted in part to inorganic calcium compounds and organic products. Zinc dialkyldithiophosphates were likewise degraded to inorganic zinc-containing substances [19]. This work demonstrates that metallic species in lubricating oil undergo changes in form and composition during use and underscores the need for more definitive speciation.

Both metallic iron and iron oxides are formed as wear debris from ferrous metals, and so it is likely that at least some of the iron in waste oil is present as Fe_2O_3 and Fe_3O_4. The level of knowledge about solid metallic wear debris is now quite sophisticated, and examination of such small particles is a valuable technique in evaluating rates and mechanisms of wear [20]. No data have been found, however, regarding soluble iron compounds in used oil.

4.3 Conclusions and recommendations.

The concentrations of metal-containing species in re-refined basestocks are very low. It seems unlikely that their detailed characterization would be significant in terms of impact on end uses.

Collectively, the literature shows large gaps in the knowledge of the chemical and physical nature of metals in oil. Figure 2 outlines the complex relationships among different physical and chemical forms of metals in such oils.

On the basis of the extraction studies mentioned above, we conclude that unknown fractions of sodium, zinc, barium, calcium, iron, phosphorus, magnesium, boron, tin, and lead are present in water-soluble forms, which implies that they exist in part as ionic or salt-like forms. This suggests that at least some of the metal content in used oils can be removed by aqueous extraction procedures.

The filtration and centrifugation studies cited earlier indicate that many metals, notably lead, zinc, calcium, barium, magnesium, and iron, are present in used oil in part as small-sized particles. Thus, gravity-assisted separations may serve to remove some of these metals from used oils. The literature provides no firm basis, however, for quantitative arguments, and the particle size distributions for the metallic species are not given.

Information on the chemical nature of metallic species in used crankcase oil is very scanty. Lead may be present as tetraalkyllead fuel additives and their hydrolysis product, lead oxide, as well as lead metal and lead bromide and chloride. Another obvious form in which metals may occur is simply that of the original additives. Zinc dialkyldithiophosphates are intended to form films of zinc sulfide on moving parts, and so it is reasonable to expect that zinc sulfide will occur in the oil. Alkali earth metal sulfonates may hydrolyze slowly to produce hydroxides of calcium, magnesium, and barium.

In our opinion, there is a definite need for better characterization of the physical and chemical forms in which metals occur in used oil. Careful measurements of distribution of metals among water-soluble, oil-soluble, and particulate fractions (fig. 2) and of the precise nature of the metal-containing chemical species in used oils will:

(a) provide essential data for establishing more economical and efficient processes for removing metals that may be deleterious in uses of spent lubricating oils, and

(b) provide a basis for evaluating potential environmental hazards associated with use or disposition of used oil by providing data from which to evaluate the possibility of mobilizing and transporting toxic metals into the environment.

The highest-priority research needs are, in our view, the following.

(a) Quantities and form(s) of metals in the particulate and oil-soluble compartments shown in figure 2 should be established. This information is needed to assess the toxic metal hazard associated with these phases.

Since the particulate phase has been shown to contain large quantities of toxic metals, initial focus on the solids is appropriate. Further, since the lead levels are orders of magnitude higher than for other metals of potential environmental

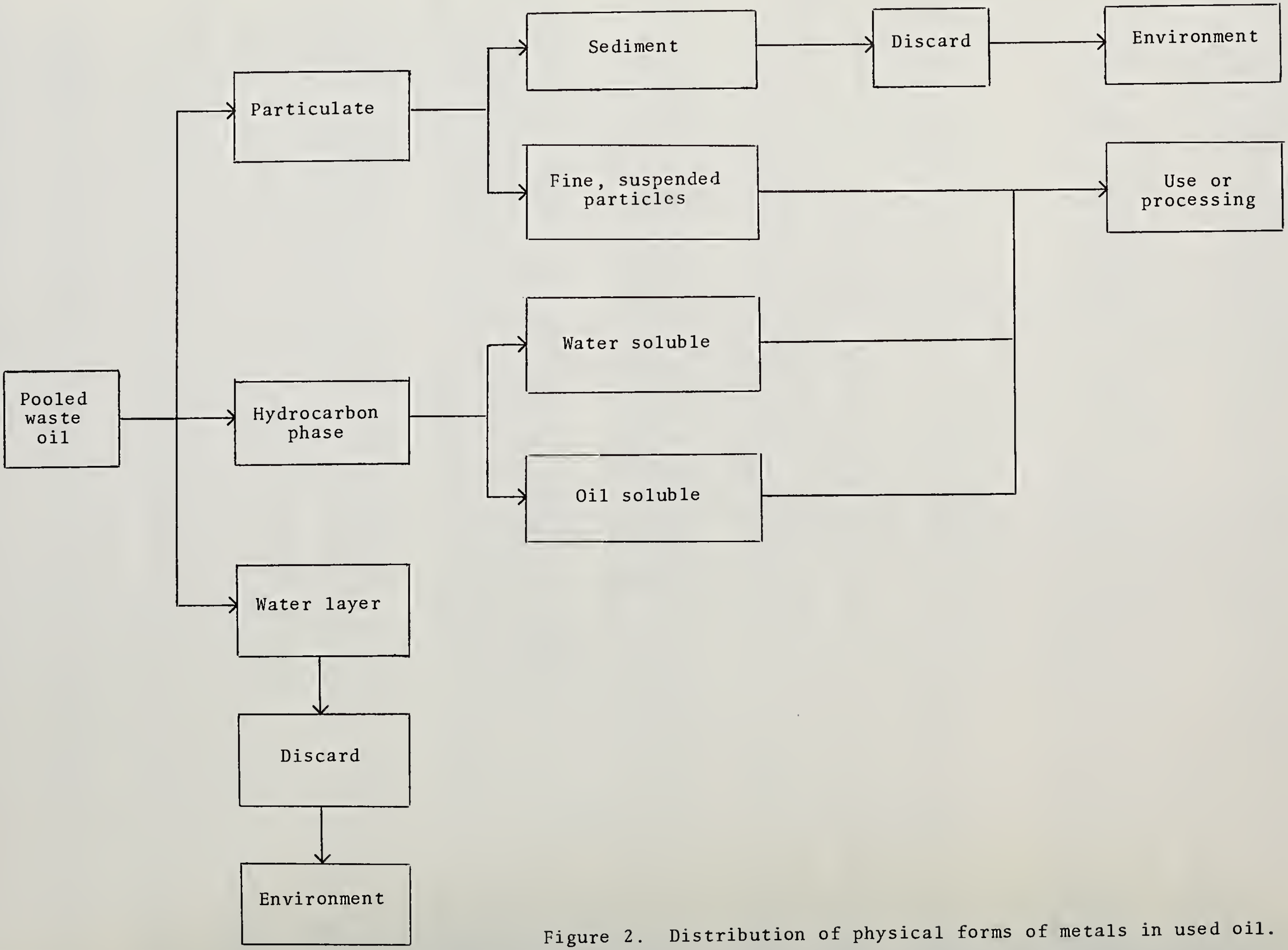

Figure 2. Distribution of physical forms of metals in used oil.

concern (e.g., zinc, barium, and chromium) immediate attention should be directed to lead. As has been amply demonstrated for mercury, the course and rates of biotransformations of heavy metals depend on the specific chemical form in which the metal is present in the environment. It is reasonable to expect that toxicity of lead, barium, chromium, and other metals found in used oils will be sensitive to chemical form. Thus, definitive speciation of particulate and oil-soluble metals, coupled with developing knowledge of environmental biological and abiotic transformations of metals, will contribute to legislative goals of minimizing environmental impact of use and disposition of spent oils.

(b) The size distribution of metal-containing particulates in waste oil should be established. Removal of metals by mechanical means (filtration and centrifugation) may be of value in end uses such as combustion (section 5.3) and road oiling (section 6.3) where the cost of transporting large volumes of used oil to a central "decontamination plant" would be excessive. Alternative re-refining methods which do not require disposal of acidic residues would be advantageous from an environmental standpoint.

Further, since the rates at which particulates are transported through the environment, solubilized, and metabolized by living organisms are expected to depend on the surface area of the particles involved, definitive characterization of the particulate fractions is of concern from the environmental viewpoint.

(c) The quantities of water-soluble metals in waste oil should be determined. Metals in water-soluble form are readily absorbed by plant and animal life [11,21], so toxicity is often directly related to water solubility. Information about water-soluble or water-suspended metals is needed to assess the degree of hazard associated with disposal of waste water from used oil and with leaching from landfills containing sludge and sediment, as well as to provide data from which to assess the effect of aqueous extraction procedures in spent oil processing.

Of subsidiary importance are the following.

(d) The dimensions of small iron particles in used oil should be confirmed by microscopic techniques. Small iron particles are expected to have an abrasive effect on pumps, valves, and burner nozzles. Data on size and concentration of these particles, if they are present, would be needed to design efficient magnetic removal techniques.

(e) When more data concerning the composition of metal-containing species and polycyclic aromatic hydrocarbons in used oil are available, this question should be addressed: Do any of these metallic compounds catalyze the oxidation of polycyclics to produce derivatives more carcinogenic than the aromatic precursors? *In vitro* catalysis of aromatic hydroxylation reactions by soluble transition metal salts, notably iron and copper, is well known, as are the analogous enzymatic processes [22]. Similar processes may well occur in situations where waste oil is dumped or applied to the ground. Although this consideration is only speculative at present, the potential hazards are sufficient to justify sampling and analysis for oxidized polycyclic aromatic hydrocarbons.

The proposed characterization studies present several significant measurement problems. Methods suitable for the characterization of small, amorphous particles, such as are expected to occur in used oil, are not well developed. New techniques such as ion and electron microprobe analysis and micro-Raman spectroscopy, offer substantial promise for characterization of oil-borne particulates, and the NBS expertise in these areas may offer opportunities for significant advances in the used oil characterization problem. Recent NBS accomplishments in the speciation of organometallic materials relevant to water pollution by chromatography/atomic absorption techniques suggest that the problem of determining the chemical nature of soluble species may also be tractable. These techniques could profitably be applied to the characterization of lead-containing species and to determining the fate of additives in lubricating oils.

5. Significance of Metals to Use of Waste Oil as a Fuel

5.1 Overview and magnitude of the problem.

Although the volume of waste oil available for use as a fuel is less than 1 percent of the total volume of fuel oil burned annually, the prospect of recovering in a useful form the energy content, estimated as 120,000-140,000 Btu per gallon, of waste oil is attractive from the resource conservation standpoint.

Potential problems associated with the occurrence of metals in waste oil used as fuels include (1) effects on overall combustion efficiency, (2) gaseous and particulate emissions, and (3) erosion and corrosion effects on combustion system components.

Studies of flame retardancy mechanisms have shown that metals exert an influence on flammability of fuels, smoke formation, and rates of radical reactions [44]. However, these effects are largely kinetic in nature and are most apparent in laboratory situations. They are not expected to be of great significance in efficient combustors which feature turbulent flow and long residence times for molecular species. These effects should be further minimized if waste oils diluted with large volumes of new oil are burned. Accordingly, the metal burden of spent oils used as an addition to other fuels is not expected to be significant.

The problems associated with lead emissions due to the high lead content of waste oil have received considerable attention. Combustion of the annual production of used automobile crankcase oil is calculated to produce 25,000,000 (2.5×10^7) pounds of lead. This estimate, which surely has large uncertainty limits, is based on the following assumptions [7,23].

(a) Only automotive lubricating oils contain lead levels high enough to be of concern.

(b) 6×10^8 gallons of such oil are produced annually. (Other estimates or extrapolations of estimates from regional data range from 4 to 7.3×10^8 gallons [6].

(c) Since about 40 percent of waste oil is burned as a fuel, $(0.4)(6 \times 10^8)$ or 2.4×10^8 gallons of automobile crankcase oil is burned.

(d) The used oil contains 1.37 percent (w/w) lead.

(e) The density is 7.6 pounds per gallon.

The lead output = $(2.4 \times 10^8$ gal. oil)(7.6 lb/gal)$(1.37 \times 10^{-2}$ lb. lead/lb. oil)= 2.5×10^7 lb. lead. Table 14 lists the calculated yields of metal oxides formed as combustion products from used crankcase oil [24].

Although Federal regulations concerning lead emissions have not yet been promulgated, limitations on lead output may be found in State ambient air quality standards: California, 1.5 μg/m^3 averaged over 30 days; Montana and Pennsylvania, 5 μg/m^3 averaged over 90 days.

The possibility that low volatility oxides might generate corrosive or erosive slags has not received much attention, and there is some controversy as to the seriousness of this problem. No serious problems have been described in reported studies, but detailed studies of the effects of metals in oil on refractories have apparently not been carried out. Corrosion of refractory linings may possibly be observed over long periods.

5.2 Survey and evaluation of the literature.

5.2.1 Combustion of mixtures of waste and virgin oil.

The lead content in waste crankcase oil has been previously discussed in section 2.2. An additional survey in 1973 found a lead:ash ratio of 35 to 40 percent for oil from service stations, and a lead content of 1.37 w/w percent versus 0.068 w/w percent in reference 23. Differences with respect to reference 5 were explained [25] on the basis of (1) increased interval between oil drains, (2) decreased gasoline mileage, and (3) changes in additive formulations. In any event, it seems clear that combustion

Table 14. Calculated Yields of Metal Oxides from Combustion of Lubricating Oil[a] [53]
(lb. oxide/10,000 gal. oil)

	Jackson, Miss.	Okla. City, Okla.	Washington, D.C.	Doraville, Ga.	San Carlos, Calif.	Dearborn, Mich.	St. Louis, Mo.	Houston, Tex.	Lyons, Ill.
Aluminum	4.6	5.1	2.6	5.1	4.4	4.8	4.5	4.4	6.3
Barium	43	25	57	20	31	9.3	33	45	38
Boron	3.6	3.6	2.8	4.3	3.6	5.9	3.6	3.8	5.9
Calcium	136	220	162	131	220	147	120	162	168
Chromium	2.6	2.9	4.8	1.5	3.8	2.6	2.2	2.9	1.2
Copper	1.1	.9	1.4	1.2	1.6	1.4	1.2	1.3	1.5
Iron	34	32	17	30	28	36	32	30	42
Lead	650	650	400	570	480	720	650	570	650
Magnesium	23	10	23	36	19	31	25	61	25
Nickel	.2	.3	.3	2.4	.5	.6	.3	.9	.7
Phosphorus	225	225	255	211	173	264	189	189	173
Silicon	29	22	13	24	24	27	64	19	24
Tin	0.6	0.6	0.6	1.0	0.9	1.0	0.9	1.3	0.9
Zinc	36	46	58	33	54	45	32	32	44
Total	1,188.7	1,243.4	997.5	1,070.5	1,043.8	1,295.6	1,157.7	1,122.6	1,180.5

[a] Calculations are based on analyses of pooled oil from the locations tested. Lead content averaged 0.68 percent (w/w). See discussion in section 4.2.1.

Source: EPA, based on tests conducted at various U.S. sites by members of the Association of Petroleum Re-refiners.

of used crankcase oil presents problems associated with emission of metallic species. Of these, lead has received the most attention in the literature (although other metallic elements also pose toxicity problems) because of its well-known physiological effects and high concentration in used oil.

Burning waste oil has been of interest both as a solution to the disposal problem and because of the prospect of recovering fuel value from the waste. A number of feasibility studies, discussed below, have been conducted, and the results are summarized in table 15 [7]. The lead levels in air at ground level calculated deal only with lead emitted at the site studied and could not be used to predict total ambient lead levels.

A comparison of metal levels in waste oil, residual oil, and coal is given in table 16.

Table 16. Metals in Waste Oil, Residual Oil, and Coal [6]

Element	Waste oil	Residual oil	Coal
	(concentration, ppm w/w)		
Barium	1,005	--	258
Calcium	1,850	48	7,768
Chromium	29	13.7	24
Copper	177	1	64
Iron	1,025	120	14,467
Lead	6,000	3	71
Magnesium	559	14	1,362
Phosphorus	1,250	--	30
Silver	1	0.3	1.7
Sulfur, %	0.43	2.15	2.75
Tin	58	--	225
Zinc	1,650	--	123

Studies at Esso Research and Engineering Company indicated that burning undiluted waste oil led to the formation of metal-containing deposits in parts of the boiler [26,27]. These studies also noted that lead is the most volatile element of the ash-forming metals in oil. Thus, as the boiler temperature was increased from 1,000° F to 1,600° F to 2,000° F, the lead content of the ash decreased from 31 w/w percent to 22 w/w percent to 2 w/w percent [27].

A study of the Shell Oil Company reported that, in burning mixtures of 36 to 89 v/v percent waste oil with No. 6 oil, stack emissions accounted for about 50 percent of the input lead. It was thought that since significant amounts of deposits in excess of equilibrium amounts were not observed most of the remaining lead was ejected during stack blowing. The powdery, flake-like residue in the firebox could be readily removed by this technique [28].

Results of a test carried out by the Humble Oil and Refinery Company in which undiluted waste oil was burned reported a ground lead level of 0.05 $\mu g/m^3$ during normal operations and 0.67 $\mu g/m^3$ while blowing out soot. The soot from the burner was a dark gray powder which was reported to contain 43 weight percent lead [28].

Data on emission characteristics are available from tests carried out at the Hawaiian Electric Company where 7.5 percent solutions of waste oil in No. 6 fuel oil were burned. Flakes of substance discharged from the stacks contained 36 percent iron, 4.3 percent lead, 1.7 percent calcium, 0.4 percent zinc, and 0.3 percent magnesium. The lead content of the total particulate averaged 18.4 percent, suggesting that the size distribution for lead-containing particulate emissions was nonuniform. Complications arose in sampling for particulate matter in the stack. The probe nozzles, which were inserted directly into the stack, were 0.2 inches in diameter, but some of the flakes described above were larger than that. Mass balance studies showed a throughput efficiency of 50 percent for lead, 60 percent for zinc, and 95 percent for sulfur [29].

Burning tests carried out by Esso using undiluted waste oil indicated that most of the lead and zinc are emitted as submicrometer-sized particles. Results for these elements, as well as calcium, phosphorus, iron, and barium, are shown in table 17 [26].

Table 15. Summary of Available Data on Used Lubricating Oil Burning Tests [7]

Test no.	Company and location	Burner		Virgin fuel	Fuel feed rate U.S. gal./hr.		Used oil in total fuel	Lead in used oil % w/w	Stack height	Soot blowing	Emission during normal operation	
		Type	Size		Total	Used oil					Pb out/Pb in % w/w	Ground level Pb conc. (μg/m³)
1	Mobil Oil Port Mobil, NY	Steam	18,000 lb./hr.	No. 6 F. O.	147	5	5% v/v	1	60	Yes	50	1
2	Shell Oil Ref. Wood River	Steam	60,000 lb./hr.	No. 6 F. O.	80-850	61-346	36-89 % v/v	0.5 0.8 1.0	130	No	42-106[a] 20-26 28-34	Not reported Not reported Not reported
3	Gulf R & D	Domestic home oil burner		No. 2 F. O.	3	0.8	25% v/v	1.1	15	No	28	<5
4	Humble Oil Sewell's Point	Steam	17,000 lb./hr.	None	100	100	100%	Not reported	35	Yes		<0.05 0.67[b]
5	Humble Oil Baltimore Terminal	Steam	75,000 lb./hr.	No. 6 F. O.	≈400	4 20 60	1% v/v 5% v/v 15% v/v	0.8	25	Yes	44-70 33-51 21-24	0.11 0.34 0.54
6	Aberdeen Pvg. Ground Edgewood Arsenal	Steam	5,000 lb./hr.	No. 2 F. O.	≈40		30% v/v	0.67		No	≈3	
7	Esso Research and Eng. Linden, NJ	Steam	1,000 lb./hr.	None	7.5	7.5	100%	0.4-0.6	--	No	<50	Not reported
8	Hawaiian Elec. Oahu, HI	Steam	≈20,000 lb./hr.	No. 6 F. O.	1,900	140	7.5% v/v	0.45	137	Yes	25-29	2.72
9	Northern States Power Co.	Steam	≈500,000 lb./hr.	Coal	≈30 ton/hr.		3.1% w/w	0.65	--	Yes	24-61[c]	
10	GCA Tech. Div. New Bedford, MA	Municipal incinerator		None	300	300	100%	1.0	100	No	Not reported	0.05

[a] A 15-minute sample during which 3 minutes were soot-blowing; sample results are low because sampling technique could not quantitatively pick up all lead loss during soot blowing.

[b] Based on ambient sample during soot blowing.

[c] Includes lead collected in precipator fly ash.

5.2.2 Boiler corrosion.

Metals in oils can contribute to corrosion of refractory materials in boilers through the formation of low melting phases and eutectics. Sodium in fuels, for example, can cause corrosion by the fluxing of low-melting sodium-vanadium compounds; the corrosiveness of these deposits is affected by the presence of other elements, metal tube temperatures, and boiler operating conditions [30]. However, sodium in waste oil is concentrated in the water layer in waste crankcase oil; remaining sodium could probably be removed, if desired, by water washing (see section 3.2). Vanadium levels in refined oils are very low.

The effects of other metals have been considered from the standpoint of boiler corrosion [23]. Magnesium in fuel oil forms a high-melting ash and prevents corrosion. It reduces a flyash strength and makes soot blowing easier. Calcium acts in a similar way, but it can also lead to the formation of calcium sulfate deposits which are hard to remove by water washing. Barium can also cause deposits on heat transfer surfaces.

Lead can also enter into eutectics. The $PbO-Al_2O_3$ phase diagram reveals a eutectic of m.p. 865° C when the PbO concentration exceeds 70 percent [31]. Such high levels of lead seem unlikely to be encountered in boilers vented to the atmosphere. High local lead concentrations could conceivably occur in cracks and behind firebricks, and the presence of other metals could reduce the quantity of lead required to yield a low melting phase.

While boiler corrosion is potentially a serious problem, the literature cited does not report difficulties due to attack on boiler tubes or linings during short-term trials in which mixtures of waste oil and new oil were burned. An 8-year study at Aberdeen Proving Ground led to similar conclusions [32]. The experience of the Snow White Cleaners and Launderers is instructive in that it appears to be a worst-case example in which undiluted waste oil was burned [7]:

> "This firm has burned waste oil for years with the following results. They have found no problems and deposits accumulated only on the firebox floor, not on tubes or refractory walls. These deposits are removed by brush about every three months. No excessive burner erosion has been experienced."

Reference 24, however, cites studies which indicate that difficulties due to scale accumulation were encountered when burning waste oil. It is possible that differences in combustion chamber design and operating conditions may account for some of this conflict. Careful observation of burner condition and performance over long periods of time in boilers using realistic new:used oil ratios is needed to clarify this situation. Accumulation of deposits on burner tubes will clearly tend to reduce boiler thermal efficiency and the economy of burning used oil.

5.2.3 Other combustion situations.

This section discusses combustion of waste oil under conditions where metals are retained in the combustion products or in materials heated with the combustion products, reducing emission to the atmosphere.

A study conducted at the Northern States Power Company employed a mixture of 3 percent waste oil and 97 percent coal as fuel. About half the input lead was emitted as particulate, but 98 percent of that was removed by the electrostatic precipitator [5]. The flyash contained 230 ppm lead, compared with 30 ppm when waste oil was not burned. Only 0.1 ppm of this lead was soluble in hot water, and it was concluded that the flyash would be suitable for use as aggregate in Portland cement, a normal market outlet for the ash [23]. It was suggested that the flyash acts as an adsorbant for metals in the feed [5], but it is not clear how the lead is incorporated into the flyash; the precipitator might remove particulate lead oxide even if it were not associated with ash.

Berry and McDonald have reported burning mixtures of used oil and fuel oil in a cement kiln. The cement clinker retained 70 to 80 percent of the total fuel lead; an additional 9 percent was retained in conditioning tower solids. Discarded, pelletized dust contained less than 4 percent lead [33]. Both phosphorus and zinc were found to be completely contained in the clinker. While it is known that addition of lead [34] and zinc [35] salts can alter the physical characteristics of cement, no significant changes in setting time or compressive strength were observed in these finished Portland cements which contained 0.0015 to 0.0058 percent lead by weight. No data were presented which indicate the rate at which lead could be leached from the cement, but given a large volume/area ratio, the rate might be expected to be slow.

A Japanese patent describes the production of charcoal by roasting waste oil sludge from petroleum refining. The product was used to remove heavy metals from waste water generated by electroplating plants [36]. The patent does not disclose how much lead is emitted in the roasting process or leached from the charcoal.

A preliminary study by the National Oil Recovery Corporation indicated that residues from the vacuum distillation of waste oil could be used as a fuel in secondary lead smelting. The lead in the fuel is evidently trapped by the emission control devices normally used in lead smelters [7].

The use of waste crankcase oil as a municipal incinerator fuel has been considered. Lead emissions were calculated on the assumption that the oil contained 1 percent (w/w) lead and that half of the metal was retained in the solid combustion products. The estimated maximum 3-month average ground level concentration of lead attributable to incinerator operations was about 0.05 $\mu g/m^3$. It was concluded that automotive waste oil was a suitably fuel for waste incineration combustion processes [13]. The Environmental Protection Agency waste oil study, prepared in 1973, does not indicate how much used oil is disposed of in this way [6].

5.3 Conclusions and recommendations.

In respect to suitability as a fuel in terms of heat content, new and waste oil appear substantially equivalent except for slight differences in density and provided that water and large particulates are moved. It does not appear that metals in used oil will significantly influence the combustion process or the short-term integrity of refractory linings.

The major problem associated with the metal content of spent oils used as fuels is that of emission of toxic metal-containing species, particularly lead compounds, into the environment.

Because of the high levels of metallic species in used oils (see table 5), removal of these metals, especially lead, from waste oil prior to combustion or from subsequent particulate emissions is required to reduce release of toxic metals to levels comparable to those resulting from combustion of new fuel oils. Combustion of used crankcase oil in conventional oil-fired power plants leads to an increase in emissions containing metals introduced in the used oil. Because of the toxicity and high concentration of lead, this metal has received the most attention. Other metals (e.g., barium, chromium, and zinc) have been discussed less. It is tacitly assumed that barium, chromium, and zinc are retained in solid combustion products along with lead. The throughput efficiencies for zinc and lead are comparable--about 50 percent; mass balances for barium and chromium were not reported.

The particle size distribution for metals found in one study of particulate emissions from an oil-fired plant is given in table 17. The preponderance of lead in the submicrometer particles is striking.

Table 17. Metal Content of Particulate Emissions from Burning of Waste Oil [26]

Metallic compounds	Wt. % of total particulate emissions		Wt. % of total emissions of metal in particles of indicated size range		
	Range	Average	1 Micron	1-10 Micron	10 Micron
Barium	1.2 - 2.6	1.9	3.3-51	40-79	8.9-18
Calcium	8 - 13	10	10 -19	71-74	10 -15
Iron	0.9 - 1.3	1.1	2.7-36	51-80	13 -18
Lead	14 - 19	16	76 -79	16-21	2.7- 4.4
Phosphorus	6.1 - 7.7	6.9	23 -42	49-66	8.9-10
Zinc	3.7 - 5.0	4.3	56 -73	23-39	3.4- 5.0

With the anticipated phasing out of lead in gasoline, the average lead content of waste oil is expected to drop. The Environmental Protection Agency estimated that by 1978, 53 percent of the pool of gasoline will be lead-free, 63 percent in 1979, and 72 percent in 1980 [23]. While the lead problem may attenuate in time, there is no basis to believe that it will disappear.

Organomanganese fuel additives will likely lead to increasing levels of this element in used engine lubricating oil. Use of (methylcyclopentadienyl)-manganese tricarbonyl (MMT) at a level of 0.125 gm manganese/gal. fuel has been suggested as a means of improving octane ratings, and MMT is contained in about one-half the unleaded gasoline now being sold [46]. The impact of current use of manganese additives is difficult to assess in detail, but some crude estimates are possible. Assuming that lead and manganese throughputs are about equal and that a fuel lead level of 2.5 gm/gal. [47] leads to a used oil level of about 1 weight percent and given that unleaded gasoline accounts for 25 percent of all automotive gasoline sales [48], then manganese in used oil concentrations on the order of 60 ppm are expected. If MMT were so used in all gasoline, this figure would increase to 480 ppm. This compares with manganese levels of 2 ppm in used oil as found in table 4.

The effect of such manganese levels on the environment is unclear. If it is assumed that toxicity hazard tracks threshold limit values (TLV's), then a comparison of TLV data is helpful. Sax [49] lists a TLV for lead of 0.5 mg/m^3, for manganese (as Mn) 5 mg/m^3, for barium 0.5 mg/m^3, and for CrO_3, 0.1 mg/m^3. This indicates that manganese additives present much less of a health hazard than lead because of lower toxicity and lower anticipated concentration in used engine oils. It is possible, however, that increased interest and research in manganese toxicity may lead to downward revision of the TLV.

The use of organomanganese compounds as fuel oil additives to reduce smoke levels has been described [50-52]. However, it seems unlikely that MMT from gasoline would be transported intact into crankcase oil, and manganese is more likely to be found there as an oxide.

Available data on the burning of waste oil in coal-fired rather than in oil-fired plants suggest a useful approach to the lead emission problem. Particulate emissions from oil-fired plants are below those mandated by Federal or State air quality standards, and so electrostatic precipitators are not normally installed in plants which burn only fuel oil [23]. The results obtained by Northern States Power Company indicate that the pollution control devices normally installed in coal-fired power plants are effective in removing lead from stack emissions. Both coal- and oil-fired operations leave about 50 percent of input lead behind, but much of this residual lead is vented to the atmosphere during soot blowing procedures in oil-fired plants. These data, taken with the conclusion of the Environmental Protection Agency Administrator that "...it would be prudent to reduce preventable lead exposure..." [23], make it appear likely that combustion of used crankcase oil will be of concern from the air quality standpoint because of an increase in total lead emissions from individual facilities, each of which meets current ambient air quality regulations. Removal of heavy metals, or the use of emission control equipment, are possible strategies which may be used to eliminate or reduce potential lead emission problems.

As discussed earlier, a better characterization of the physical and chemical forms of metals in oil can be expected to provide the underlying data from which to develop techniques for more effective and efficient removal of toxic elements prior to waste oil combustion. There is, however, a need for applied research on processes to achieve this removal. Additional research is probably needed to identify processes that can utilize waste oils under conditions where emissions are contained in the end product or combustion residues, including optimization of waste oil combustion in coal-fired plants.

In view of the almost total concern of previous studies with lead emissions, it appears highly important to develop better data on the emissions of other toxic elements, such as chromium, and barium from waste oil combustion.

6. Significance of Metals in Uses of Waste Oils for Road Oiling

6.1 Overview.

Application of waste oil to secondary roads in order to suppress dust is a principal (see section 1) use of waste oil generated in the United States [1,28]. The corresponding amount of lead applied to road surfaces is significant. If one assumes that half of the approximately 200 million gallons of used oil employed as a dust palliative and/or incorporated into asphalt is applied directly to road surfaces, then a calculation like that in section 5.1 shows that about 5 million pounds of lead are involved. Road oiling is essentially a "throw-away" or dispersive use and, as such, presents many of the same problems as dispersal by dumping of waste oil. Waste from re-refining operations are usually disposed of by landfilling. These residues can present special difficulties on account of their high lead (see table 7) and acid content. The literature does not indicate that metals in used oil have any adverse effect on properties relevant to use as a dust palliative. The problems associated with metals in road oil are chiefly environmental ones.

6.2 Survey and evaluation of the literature.

An Environmental Protection Agency study [38] disclosed that on one particular road 70 to 75 percent of the road oil was lost by runoff and transportation by dust. Twenty-five to 30 percent was lost by volatilization, adhesion to vehicles, and biodegradation. One percent of the total oil estimated to have been applied over the 12-year period stayed on the road. This study also showed that vegetation in adjacent fields was high in metallic elements contained in the oil. It is not known whether these results are typical.

Used crankcase oil contains detergents, dispersants, and emulsifiers. Their effect is to form water-oil emulsions and suspensions of oil and dust in runoff water. This may account for the high oil loss from the road surface. While some of these agents (e.g., detergents) contain alkaline earth metals, their action arises from physical phenomena associated with ligands rather than from the metals themselves. Nevertheless, the long-term effect is to disperse the heavy metal load in the used oil into the environment.

The literature on oil disposal and metal biotransformations offers some insight into the fate of metals discharged into the environment in road oiling. Disposal of used oil has been reviewed from the standpoint of biodegradation [39,40], a process which has implications in the case of road oiling applications. Bacteria are known to decompose oil to form cellular matter and humus. Such processes have the potential for freeing the particulate matter known to be present in used crankcase oil from an oily hydrocarbon matrix permitting a more rapid spread of the metal content of the particles through the environment. If lead is present as the sulfide (solubility product, $K_{sp} = 3.4 \times 10^{-28}$), it may be converted by ambient sulfur oxidizing thiobacili [41] to be the more soluble lead sulfate ($K_{sp} = 1 \times 10^{-8}$) [42]. Furthermore, it is also possible that microbial action will break down metal-containing additives in the oil. The importance of these processes is speculative and a proper assessment of their significance to environmental quality requires, as previously noted, a better understanding of the detailed chemical nature of the forms in which the metals are introduced.

A recent study indicates that plants are able to convert metals from a water-soluble form into solids. Pea plants and pine tree seedlings cultured in the presence of soluble zinc and lead released particles containing these elements from the plant surfaces [20]. This effect provides another mechanism for the diffusion of water-soluble metals contained in or derived from waste oil, particularly zinc, through the environment.

Finally, in a different road application of waste oil, an inexpensive road topping has been prepared by combining used oil with a medium-cured asphalt and coarse aggregate. It is thought that the rate of removal of metallic substances from this viscous, hydrophobic matrix will be extremely slow [7], but appropriate data appear not to exist.

6.3 Summary and conclusions.

Specifications for road oil are, at best, loose. The Province of Quebec has set standards for viscosity, flashpoint, percent water, and nonvolatile residue [7]. A waste oil which complied with these specifications could clearly contain large quantities of toxic metals. In the absence of performance standards, there is no objective basis for commentary on the equivalency of waste oil to commercial, unused oil which is applied to roads. The literature, however, suggests that used oil is less satisfactory because of the presence of detergent additives.

Current concern to reduce preventable lead exposure (cf. section 5.3) makes it appear unlikely that such use of untreated waste crankcase oil will be satisfactory over the long term.

In this area, again, research is needed at fundamental levels and in the field. Characterization and speciation of metals in used oils is essential to translate mobilization and transport studies into practical prediction of the environmental effects of metals dispersed in road oiling or simple landfill disposition.

Available field studies are extremely limited. There is a need for more careful evaluation of losses of road oil from unpaved roads to determine whether the few data available are representative. Process research to develop inexpensive methods for decontamination of small volumes of oil should also be encouraged.

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APPENDIX II

NEW ASTM COMMITTTE D-2 DIVISION

TITLE: Technical Division on Recycled Petroleum Products

SCOPE: The promotion of knowledge and development of specifications, classifications, recommended practices, nomenclature, and methods of sampling and testing for used oils and products derived from used oils. Much of the knowledge and technology, including standards, relating to virgin petroleum products and lubricants under the jurisdiction of other divisions will apply to used oils and recycled petroleum products. Accordingly, activities will be coordinated with these divisions in Committee D-2, with other committees in the ASTM, and with other organizations as appropriate.

STRUCTURE:
Section I - Automotive Lubricants
Section II - Used Oils and Basestocks
Section III - Fuel Oils
Section IV - Hydraulic Oils
Section V - Industrial Oils
Section VI - Miscellaneous Products
Section VII - Editorial

APPENDIX III

SUMMARY

OF

RESPONSES TO QUESTIONNAIRE ON NEED TO FORM IN ASTM COMMITTEE D-2 A NEW TECHNICAL DIVISION ON WASTE OIL AND RECYCLED PETROLEUM PRODUCTS

Number of copies of questionnaire circulated: 1,500

When circulated: Fall 1977

Questionnaire recipients: Members of ASTM Committee D-2 and the Association of Petroleum Re-refiners

Number of respondents: 212[1]

1. Does your organization have an interest in:

   (a) Recycled petroleum products? Yes 167

   (b) Waste oil? Yes 149

2. In what general ASTM category are you?

   Producer 70, Consumer 64, General interest 64

3. What types of recycled products or waste oil do you produce, use, or have an interest in?

(a)	Waste oil (engine, industrial, hydraulic, gear, transmission, and other)	57
(b)	Recycled engine oils	44
(c)	Fuel (waste oil, blends of waste oil and virgin fuels, fuel from waste oil)	35
(d)	All types of recycled lube oils	34
(e)	Recycled hydraulic oils	33
(f)	Recycled base lubricating oils	25
(g)	Recycled industrial lubricating oils	19
(h)	All types of recycled products	15
(i)	Slops, spills, and mixed products	11
(j)	Recycled gear oils	7
(k)	Greases from recycled oils and recycled base oil for greases	6

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[1] Some companies, organizations, and government agencies in some instances replied more than once owing to multiple memberships.

(l) Oils in metal manufacture, cutting, and forming:

Cutting oils	5
Machinery	4
Rolling mill oils	5
Quench oils	1
Way lubricants	1
Form oil	1
Soluble oils	2
Drawing compounds	2

(m)	Automatic transmission fluids	3
(n)	Additives for recycled lubricants	4
(o)	Transmission fluids	5
(p)	Turbine oils (gas and steam)	6
(q)	Recycled diesel engine oils (RR)	3
(r)	Recycled solvents and specialty products	3
(s)	Recycled oil for chemical pumps	1
(t)	Recycled car journal oils (RR)	2
(u)	Road and dust control oil	3
(v)	A/C engine oils (including synthetic)	4
(w)	Waste oil as turbine fuel	3
(x)	Fuels and lubricants supplements	1

4. Would you like to see ASTM develop standard specifications and classifications for recycled petroleum products?

Yes 160

If yes, are you interested in such standards for

(a)	Waste oil feedstocks?	Yes	84
(b)	Recycled lube oil basestocks?	Yes	128
(c)	Recycled engine oils?	Yes	117
(d)	Recycled industrial oils?	Yes	107
(e)	Recycled hydraulic oils?	Yes	113
(f)	Recycled turbine oils?	Yes	86
(g)	Waste oil as fuel (distilled, residual, blended)?	Yes	128

5. Would you like to see ASTM D-2 develop standard methods for waste oil and recycled petroleum products? List type of method needed and product.

By in large, most products, waste and recycled, listed under 3 were listed here. Many respondents stated that virgin product specifications and classifications tailored to end use already existed. These, along with existing standard methods defining product properties, it was stated, apply equally in most instances to recycled petroleum products and virgin products. Hence, very few new standard specifications, classifications, and methods, it was argued, would be needed.

Where a need was demonstrated, the appropriate technical division responsible for a product based on end use should be asked to develop the standard.

Notwithstanding, and in spite of what can be formidable undertakings, many standard methods, specifications, and practices were listed as needed and advice was given:

(a) Several requests for:

- Simple methods to replace expensive engine oil sequence tests.
- Standards for recycled fuel oils; little control over burning of untreated waste oil.
- Quality determination of waste feedstock; additional tests besides BS&W [bottom settlings and water] and light distillates.
- Dealing with recycled hydraulic and industrial oils rather than complex engine oils.
- Tests to assess adequate cleanup of recycled oils.
- Chemical characterization and oxidation stability of recycled lubricants.
- Standards for blends of waste oil and distillate fuel.
- Standard methods for testing and classifying waste oils and recycled basestocks.
- Methods for rust inhibition, oxidation resistance, demulsibility, extreme pressure (EP) and antiwear properties, etc. Many existing procedures should be applicable.
- Metallic contamination tests.
- Showing equivalency if recycled formulated oil is to be interchangeable with virgin-based oil.

(b) Fewer requests for:

- Modified grease tests for grease containing recycled oil.
- Lower standards for residual or heavy fuel oil (No. 6) from waste products.
- Methods to predict performance.
- Recycled oil for small vehicles.
- Less time-consuming oxidation test for hydraulic oils.
- Standard specification for recycled aircraft turbine engine oil.
- Methods for trace chemicals and hazardous materials in recycled oils.
- Empirical screening test for contaminants.
- A general classification system.
- Methods for assessing activity of inhibitor package (life expectancy) in lubes.
- Tests for cleanness, quality, and API gravity.
- Peroxide level of hydraulic oil, oxidation level of machining and hydraulic oils, organic additive level, contamination level, and flash point of all oils.
- Easily conducted physical evaluations for viscosity, distillation, residual additive content, and rust and corrosion.
- Data on filtration, blend ratios, and operational procedures when using used engine lube oil as a diesel engine fuel.

- Information on the collection and storage of waste products prior to recycling.

- Method for disseminating the processes used for recycling.

- Methods for viscosity, distillation, rust and corrosion, metals, and emulsification.

- Quick test for evaluating "street drainings."

- Accurate and reproducible test to predict coking tendencies of re-refinery feedstocks.

- Methods for BS&W, sediment, metals, bromine number, etc., to characterize spent and re-refined lube oil.

- Tests for lubricity, plate out, burn off, cleanability, sludge.

- Estimating heating value of derived fuel oils.

- Methods of classification for recycled lube basestocks by molecular type or physical properties.

- Methods to characterize and determine quality of waste oil as fuel or fuel blending component.

- Developing processes for re-refining all types of used lubricating oils.

- Statement on handling or treatment of waste oil prior to viscosity test.

- Ash analysis procedures for waste oils.

- Consistency of batch quality of recycled oils.

- Standardization of any after-marker automotive fluid.

- Defining criteria for acceptable recycled base oils by conventional physical and chemical lab tests. If additional standards are necessary, can consider (a) infrared, (b) analytical techniques for impurities in waste oil, and (c) standards associated with the re-refining techniques themselves.

- Supplementary tests in addition to those in existence to ensure quality, particularly for engine oils.

- Tests to show batch-to-batch variations in additive response and oil quality.

- Tests on basestocks: (a) storage, light and heat stability; (b) compatibility with virgin and other recycled stocks; (c) odor; and (d) clarity.

- Short-term screening tests of somewhat less precision and significance.

- Changing law requiring separate labeling for recycled petroleum products.

- Re-refining engine oil right on engine.

- Simple method for determining naphthenic oil content in lube quality feedstock.

- Tests for recycled lubricants:

    1. Residual particulates - petroleum ether insolubles-millipore, etc.

    2. Oxidation resistance - bench oxidation test - volatility, viscosity, sediment.

    3. Wear protection - Falex or pump test.

    4. Corrosion protection - humidity or ASTM D 665.

5. Specific tests for specific use applications.

6. Dissolved metals by AA, etc.

7. Foam tendency.

- Tests for:

  1. Fuel oil - water, ash, lead, halogens, sulfur, flash point, pour point, propane insolubles.

  2. Ester-based turbine oil - ash, foam, hydrocarbon contamination, oxidation stability (with additives), pour point, viscosity index, propane insolubles.

  3. Automotive lube oil - ash, lead, halogens, oxidation stability (with additives), pour point, viscosity index, propane insolubles, ester contamination.

- Characterizing waste oil by:

  1. Solids
  2. Water
  3. Rust number
  4. Sulfur
  5. Total chloride
  6. Total nitrogen
  7. Metals
  8. Ash
  9. ASTM D 116
  10. Gravity
  11. Viscosity
  12. Saponification number
  13. Color
  14. Asphaltenes
  15. Paraffin wax
  16. Degree of oxidation
  17. Flash point
  18. Light end diluents
  19. Heating value

6. Do you have standards or test methods to offer for consideration?

Seven respondents stated that they would have methods in the future. Eighteen respondents offered the following methods, many of which are obviously applicable to virgin oils as well. Some gave comments only.

- Many current standard methods should be applicable to waste oil feedstocks and re-refined basestock standards.

- Standard military and Federal specifications and qualification procedures.

- Coulometric method for sulfur, chlorine, and nitrogen.

- Some commercial airlines and some engine builders may have standards for recycled aircraft turbine engine oils.

- Laboratory oxidation stability test for fuels and lubricants.

- Percent ash and identification by emission spectroscopy.

- Coking and fouling prediction test for re-refinery feedstocks.

- Emulsion stability index test and sludge test.

- Quantitative and qualitative test for particulate contamination.

- Oxidation test.

- Heating value test similar to ASTM D 1405.

- Oxidation test, FTMS 5308.6.
- Engine and gear tests. Physical and chemical bench tests. Friction, rust, EP, wear tests, etc.
- Bench oxidation and wear.
- Different methods for the examination of waste oil.
- Company maintains extensive specifications for automotive factory-fill and service-fill products and for all sorts of manufacturing lubricants, cutting oils, hydraulic fluids, etc.
- Propane insolubles of formulated oil.

7. Are you interested in becoming a member of the division, if formed?

Yes 128

8. Will you attend meetings regularly?

Yes 130
Once a year 49
Twice a year 65

9. Will you participate actively in the work of some sections and/or study groups of the proposed new division? Yes 115

10. Comments:

Many comments were received. Herewith are a few that are representative. Some offer much encouragement.

B. W. Hutchings/F. W. Kroll, Exxon Company, U.S.A.

"In view of the growing interest in recycling and the work underway at the National Bureau of Standards and elsewhere, ASTM effort in this area is indicated to be necessary."

S. J. Lestz, U.S. Army, Fuel and Lubricants Research Laboratory, Southwest Research Institute

"Information exchange is required between major oil company refiners, re-refiners, end users, equipment builders, and military."

R. L. Riedel, SOHIO

"Believe ASTM should attempt to guide NBS in decisions and encourage reblenders to join our group."

G. A. Gettinger, Midwest Oil Refining and Recycling Company

"As a field representative, I would supply practical information and data as it accrues in the midwest section. I am glad to see ASTM take an interest in the waste oil problems in collection and reuse. It is vitally important to everyone in the nation for conserving energy and its vital national resources, which in turn through reuse will help protect our environment. This industry can use the help of Committee D-2 and ASTM as a whole to upgrade its image and its service to the committee."

W. W. Crouse, Suntech, Inc.

"I would like to point out that standards for a given product area (such as engine oils) should be handled by the appropriate technical committee (Tech B in this case) and that the new Technical Division concern itself with the feedstock-base stock consistency, rather than product standards. However, if there is a need for an additional standard for a product area, it should be turned over to the appropriate Technical Division, since that is where the expertise lies."

T. F. Sparks, Vacsol Corporation

"We are presently utilizing the ASTM Standards developed for virgin lube oils and are in most cases able to effectively use them. But in many areas associated with re-refining, there are no ASTM Standards applicable. For instance, in reference to re-refining feedstock, waste oil, it has been necessary for us to generate 'standards' within our own organization. These 'standards' are effective in determining what we feel are the important properties of waste oil, but they are, in no sense of the word, universally accepted. Accredited ASTM Standards in this particular area would be extremely helpful. There are several other specific areas, in re-refining, that ASTM Standards would also be helpful. We are commencing, right now, to prepare what we feel would be helpful synopsis on both the problem areas of re-refined product testing and also how we feel these areas should be viewed."

M. Willingham/D. J. Ellerbe, Department of Energy, Washington, D.C.

"The Agency is interested in the creation of an ASTM Technical Division on Recycled Petroleum Products to carry out indicated objectives. Given the accomplishment of its missions, the Division will enable and assist us in providing a means for satisfying the objectives of the law as prescribed [Energy Policy and Conservation Act of 1975 (EPCA) and Energy Conservation and Production Act of 1976 (ECPA)]."

Summary prepared by P. L. Strigner for Questionnaire Task Group:

D. A. Becker (NBS), B. R. Hall (API), P. L. Strigner (NRC Canada).

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16. ABSTRACT *(A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here.)*

This publication is a formal report of the second Conference on Measurements and Standards for Recycled Oil, held at the National Bureau of Standards on November 29 and 30, 1977. There were seven sessions on specific subject areas, with a total of 32 presentation The subject areas were as follows: (1) The Existing Situation, (II) Environmental Considerations, (III) Nonlubricating End Uses, (IV) Legislation and Studies, (V) Evaluation of Re-refined Engine Oils, (VI) Additional Recycled Oil Activities, and (VII) A panel discussion on the topic: "What Data Base is Required to Establish the 'Substantial Equivalency' on Re-refined Motor Oils to Virgin Motor Oils." Included in this report are invited talks given and a summary of the discussion following session VII.

17. KEY WORDS *(six to twelve entries; alphabetical order; capitalize only the first letter of the first key word unless a proper name; separated by semicolons)*

Engine oil; fuel oil; hydraulic oil; industrial oil; lubricating oil; oil specifications; petroleum standards; petroleum test methods; recycled oil; re-refined oil; used oil; waste oil.

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