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EXECUTIVE SUMMARY

Since 1957, when measurements at Mauna Loa in Hawaii and the South Pole indicated increasing levels of CO_2 in our atmosphere, there have been concerns about the impact of greenhouse gas (GHG) emissions on the global climate. A 1979 **National Academies** Report stated:

> "*For more than a century, we have been aware that changes in the composition of the atmosphere could affect its ability to trap the sun's energy for our benefit. We now have incontrovertible evidence that the atmosphere is indeed changing and that we ourselves contribute to that change. Atmospheric concentrations of carbon dioxide are steadily increasing, and these changes are linked with man's use of fossil fuels and exploitation of the land. Since carbon dioxide plays a significant role in the heat budget of the atmosphere, it is reasonable to suppose that continued increases would affect climate.*"

Since its establishment in 1988, United Nation's **Intergovernmental Panel on Climate Change (IPCC)** has raised further concerns about climate change and stated that "*Most of the observed increase in global average temperatures since the mid-20th century is very likely due to the observed increase in anthropogenic GHG concentrations. It is likely that there has been significant anthropogenic warming over the past 50 years averaged over each continent (except Antarctica).*" Several international **Conferences of the Parties** (COPs) were held to plan nations' activities to counteract this trend and to develop national inventories of GHG emissions.

At COP 21 (Paris), there was general agreement that such programs will require an understanding of existing emissions as well as the development of programs to monitor, report, and verify emissions from a broad range of sources. It was also agreed that, since about 70% of GHG emissions originate from urban environments, where a majority of the world's population lives, such monitoring efforts should focus on urban environments. Also the contributions of the so-called sub-national actors (local governments and the private sector) were recognized as being important to emissions mitigation.

As the U.S. National Metrology Institute (NMI), NIST responded to this challenge with an aim to:

a) improve our capabilities to measure GHG emissions accurately;
b) demonstrate the capabilities of atmospheric urban monitoring networks ("top-down" approaches) to determine quantitatively GHG fluxes from industrial, residential, transportation, power generation and other activities;
c) complement such measurements with "bottom-up" approaches based on socio-economic data; and
d) demonstrate that the combination improves confidence in emission estimates while identifying areas of improvement.

NIST research programs were established to improve measurements of carbon dioxide (CO_2), carbon monoxide (CO), methane (CH_4), nitrous oxide (N_2O), isotopic components of CO_2 and O_2, aerosols, and GHG flux measurements from smokestacks of industrial and power generation facilities. New methods were developed to apply such technologies over large areas for long line-of-sight measurements. NIST's programs on gas standards were expanded to improve GHG calibration standards underpinning both measurement approaches, and to work more closely with the National Oceanic and Atmospheric

Administration (NOAA), WMO's (World Meteorological Organization) Central Calibration Laboratory for GHGs; these would ensure the accuracy and consistency of measurements via traceability to the International System of Units (SI), and strengthen the mutual recognition of atmospheric GHG emission measurements.

NIST also advanced computational tools, in collaboration with other institutions, to model spatially explicit emission sources, and biogenic emission and uptake processes, and to enable urban GHG source attribution from atmospheric measurements.

NIST established its first **Urban Testbed** in Indianapolis, Indiana (the **INFLUX** Project) in 2010 in collaboration with Purdue University, NOAA, and Penn State University (PSU). INFLUX was initially designed as a mass balance experiment to measure whole-city emissions. These measurements combine aircraft and surface observations, including $^{14}CO_2$ measurements to isolate fossil fuel CO_2 emissions, taken at two locations across the city along prevailing wind directions, and a newly developed urban, spatially explicit emissions model (**HESTIA**). NIST then expanded its efforts, working with Penn. State, to include atmospheric inversion analyses coupled with observations made with a spatially dense, surface-based network, including 15 towers. Extensive sets of data obtained as part of the INFLUX project have provided the framework for evaluation of different measurement methods and computational tools and their uncertainties, as well as the reliability of the results from bottom-up approaches such as the **HESTIA** model for cities. The INFLUX Project represented the first quantification of CO_2 emissions from a large U.S. city using a dense observation network and has served as a model for other efforts around the world to quantify anthropogenic CO_2 emissions from metropolitan areas. Results over a 3-year analysis period (2012-2015) demonstrated agreement between bottom-up and top-down emissions determinations of better than 3%. As a test of the combined approach, bottom-up emissions intentionally offset by +15% were corrected by the measurements (by -14.2%), indicating the capability of the measurement and analysis method to respond to observed data.

NIST's second Urban Testbed was established in California's South Coast Air Basin (**Los Angeles Mega City** Project). The Los Angeles region was selected because it is a much larger metropolitan area with more complex meteorology and emissions characteristics than those of Indianapolis; its proximity to the Pacific Ocean presents additional challenges. The NIST Testbed was operated in collaboration with NASA's Jet Propulsion Laboratory (JPL) and NOAA, who already had an ongoing effort in the South Coast Air Basin, as well as with the Scripps Institute of Oceanography (SIO), Caltech, and NIST's private-sector partner, Earth Networks Inc. Some earlier measurements had been made in the coastal Palos Verdes Peninsula and in inland Pasadena. Complexity of the LA topography, meteorology, and emission sources required a more extensive network of monitoring locations to acquire reliable information on GHG emissions from the LA Megacity. In collaboration with the California Air Resources Board (CARB) and universities located in the Basin, a network of 17 monitoring stations was established between 2013 and 2015, which included instruments located on communication towers as well as on building rooftops. NIST's collaborators at the Northern Arizona University provided results of bottom-up emissions modeling for the LA Mega City with the **HESTIA** model. Measurements made with the monitoring system were used to separate biogenic and fossil fuel contributions to CO_2 enhancements above the background levels entering Los Angeles. The observing network and inversion methods were also instrumental in detecting the impact of COVID-19 lockdowns on CO_2 emissions, which were reduced during this period as a result of reduced use of transportation, but also because of a mild winter. These results were extended to detect the impact of COVID-19 lockdowns in six North American cities (Los Angeles, Indianapolis, Salt Lake City, Boston, Washington, DC/Baltimore, MD and Toronto, Canada). Changes in urban GHG emissions resulting from the Aliso Canyon, CA blowout at a natural gas storage unit were also detected and quantified.

NIST's third Urban Testbed is focused on the U.S. **Northeast Corridor (NEC)**. It is the most comprehensive of the test bed projects in a geographical sense extending from the Washington, DC/Baltimore, MD area to Boston. The Northeast Corridor meteorology is somewhat similar to that of Indianapolis, but it features substantial coastline and topographic relief. The presence of the Chesapeake Bay also represents further complications, as demonstrated by earlier work of the University of Maryland and NOAA. The backbone of the NEC Project is a network of in-situ tower-based CO_2 and CH_4 observation stations, which are owned and operated by Earth Networks, Inc., with continuous high-accuracy mole fraction measurements of these two greenhouse gases. In addition, the University of Maryland, Stony Brook University, and Purdue University conduct multiple-week flight campaigns each year to estimate whole-city emissions. The design of the monitoring network for the NEC Project represented an unusual challenge because of the extent of the area to be covered. Several studies were undertaken to optimize the locations of the towers, given the presence of existing towers, economic constraints, and scientific requirements. The current network, which became operational in 2017, includes 28 towers, extending from North Carolina to New Hampshire; more tower-based observing nodes are expected to be added to the network in the near future. Detailed data has been obtained about GHG emissions in the Washington, DC area and in Baltimore, MD, including the impact of COVID-19 lockdowns. These compare well with the results of bottom-up estimates from the HESTIA model for the region. Aircraft flights have also been utilized to develop estimates of GHG emissions from New York City (NYC) in the absence of a monitoring network around NYC. Fugitive methane emissions along the Northeast Corridor are of significant interest; both tower and aircraft measurements have been used monitor CH_4 emissions and their seasonal variability.

Data collected from the NIST Urban Testbeds are of great interest to the GHG measurement community, and they are made widely available. The INFLUX data are maintained by Penn. State at https://sites.psu.edu/influx/. Data from the LA Mega City and the Northeast Corridor Projects are maintained by NIST and are available at https://data.nist.gov. Spatially and temporally resolved emissions model data are maintained by Northern Arizona University at https://hestia.rc.nau.edu/ .

These projects have been and continue to be very productive, resulting in more than 200 archival publications, which have been cited more than 5,000 times by researchers in over 90 countries. They have been successful because of the engagement of an outstanding team of U.S. researchers to address a global challenge to characterize the changes to our atmosphere. Each group brought complimentary skill sets to these challenging research projects. Internal NIST expertise has also been expanded in atmospheric inversion modeling with strong ties to technical programs of NIST Laboratories. Cooperative efforts have been initiated resulting in new internal NIST research efforts and strengthening of others. An excellent example is the FOREST project, a new effort taking advantage of NIST's radiometric measurements and standards expertise coupled with the ecological expertise of an academic partner, Boston University. The result is a testbed combining spectro-radiometric and ecological measurements of a forested portion of the NIST Gaithersburg campus to develop methods to assess the performance of emerging optical remote sensing methods to measure vegetative productivity. These NIST efforts with academic partners are also critical for the preparation of future researchers to address new challenges before the climate science community.

NIST has made a significant effort to engage the international community in this global challenge. NIST organized several workshops in Central and South America to promote discussion of ***Renewable Energy and Climate Science: Metrology and Technology Challenges.*** NIST also helped organize workshops in China and in Europe to bring together the metrology and meteorology communities (members of WMO and NMIs) to address this challenge of common interest. In a parallel effort, NIST helped catalyze the formation of the ***Integrated Global Greenhouse Gas Information System (IG3IS)*** at WMO. These efforts are expected to lead to the development of international documentary standards by ISO on GHG measurements. NIST efforts have also contributed to the development of a ***National Strategy to Advance an Integrated U.S. Greenhouse Gas Measurement, Monitoring, and Information System***. And NIST efforts to quantify GHG emissions are critical for implementation of the proposed guidance issued by the U.S. Commodity Futures Trading Commission (CFTC) regarding listing of ***Voluntary Carbon Credit Derivative Contracts*** in the Voluntary Carbon Market.

Key Words:

Greenhouse Gases; Urban Greenhouse Gas Measurement Testbeds; Urban Observing Networks; Emissions Models and Data Products; Urban Biogenic Sinks/Sources; Transport Models; Atmospheric Inversion Modelling; INFLUX Project; Los Angeles Mega City Project; Northeast Corridor Project; Conference of the Parties; Intergovernmental Panel On Climate Change.

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1. Introduction

Since 1957, when the Scripps Institution of Oceanography (SIO) established carbon dioxide monitoring stations at Mauna Loa (Hawaii) [1, 2] and the South Pole,[3] we have known that CO_2 concentrations have been higher in the Northern Hemisphere, primarily due to higher usage of fossil fuels in the north than in the south. These gradients, and increasing levels of CO_2, have been a subject of concern in the scientific community for more than a century.[4]

> "*For more than a century, we have been aware that changes in the composition of the atmosphere could affect its ability to trap the sun's energy for our benefit. We now have incontrovertible evidence that the atmosphere is indeed changing and that we ourselves contribute to that change. Atmospheric concentrations of carbon dioxide are steadily increasing, and these changes are linked with man's use of fossil fuels and exploitation of the land. Since carbon dioxide plays a significant role in the heat budget of the atmosphere, it is reasonable to suppose that continued increases would affect climate*".

This statement is from the foreword of a **1979 National Academies Report**;[5] the questions to be address are how much progress has been made since then, do we have the capabilities to accurately measure the changes in the atmosphere, and what direct evidence do we have for the progress claimed by many countries? This report summarizes NIST research efforts that have been implemented over the last decade to answer some of these questions.

Since the publication of the National Academies report, there has been general agreement in the scientific community that the Earth's climate is changing. The 2007 **Fourth Assessment Report** (FAR) by the UNFCCC's (United Nations Framework Convention on Climate Change) **Intergovernmental Panel on Climate Change** (IPCC),[6] representing the international scientific consensus on the Earth's climate system, announced several findings about climate change, key to the on-going discussion of the impact of human activity on the Earth's climate. These included the following:

- *Warming of the climate system is unequivocal, as is now evident from observations of increases in global average air and ocean temperatures, widespread melting of snow and ice and rising global average sea level.*
- *Global atmospheric concentrations of carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) have increased markedly as a result of human activities since 1750 and now far exceed pre-industrial values determined from ice cores spanning many thousands of years.*
- *Most of the observed increase in global average temperatures since the mid-20th century is very likely due to the observed increase in anthropogenic GHG [greenhouse gas] concentrations. It is likely that there has been significant anthropogenic warming over the past 50 years averaged over each continent (except Antarctica).*

At one of the earlier Conferences of the Parties (**COP 13**) held in Bali, Indonesia in 2007, as part of the Bali Action Plan, the concepts of "measurable, reportable, and verifiable (MRV) greenhouse gas mitigation strategies" were adopted.[7] At the 2009 **COP 15**, held in Copenhagen, Denmark, "Quantified economy-wide emissions targets for 2020" were the main topics of discussion.[8] But there was a general concern that developed countries would “oversee” the emission inventories of the developing countries. As a result, these topics continued to be the subjects of discussion at subsequent COPs.

A 2009 **GAO** Report stated:

> “*Elevated levels of greenhouse gases in the atmosphere and the resulting effects on the earth's climate could have significant impacts in the United States and internationally. Potential impacts include a change in sea levels, ecosystems, and ice cover. The United States Congress is considering proposals to limit greenhouse gas emissions using market-based mechanisms that would place a price on emissions. Such programs would create an economic incentive for regulated entities to limit their emissions. Limiting greenhouse gas emissions requires an understanding of existing emissions as well as the development of a program to monitor, report, and verify emissions from entities that might be affected by a future regulatory program. A greenhouse gas mitigation program also requires an understanding of the numerous emissions sources and methods for calculating emissions of six major greenhouse gases--carbon dioxide, methane, nitrous oxide, and several synthetic gases. This testimony focuses on (1) the importance of quality data on emissions in the context of a program intended to limit greenhouse gas emissions, and (2) key considerations in developing reliable data on greenhouse gas emissions*.”[9]

Prior to March 2009, NIST measurement science efforts related to climate-change were primarily supported by intermittent, project-specific funding from other federal agencies such as NASA, NOAA, and EPA. These resources have been critical to the development of NIST expertise in climate-related measurements and reference data needs of the atmospheric chemistry community, e.g., ozone depletion potential measurements and methodology development, and identification of chlorofluorocarbon (CFC) alternatives.[10] NIST’s internally funded activities, directly supporting industry needs resulting from regulatory requirements of EPA, resulted in advances in standards capabilities in air and water quality. Uncertainties and interruptions of other agency funding and the increasingly narrow focus of the agencies’ needs had made it challenging to develop a coherent and effective program and associated measurement infrastructure capable of providing measurement science and standards that were responsive to pressing national needs in climate change measurement science and GHG measurements of sufficient accuracy.

During this period of expanded discussions on climate change, several federal agencies, including NOAA, NASA and DOE, organized workshops to plan their activities to address climate change. An interagency workshop was held in May 2009 to assess needs and capabilities.[11] NIST also held several meetings with stakeholders to ensure that its measurement programs addressed the needs of the climate change community and filled the voids of the global research activities, especially related to quantifying GHG emissions. One of these meetings was held at the Scripps Institution of Oceanography (SIO) in La Jolla, CA, organized by a Steering Committee on Greenhouse Gas Emissions Quantification and Verification Strategies. [12]

The scientific results produced by the climate science community showing the impact of anthropogenic atmospheric emissions on the Earth's climate have largely been based on Earth system observations. Continued efforts by the climate science, emissions reduction, and regulatory communities concerned with atmospheric GHGs rely on advances in an array of measurement capabilities and the GHG concentration, or mole fraction, standards that are the basis of the accuracy and reliability needed.

Since climate change is a global issue that presents many global measurement challenges, climate data records must rely on measurement systems that are clearly linked to the International System of Units (SI).[13] Reliance on the SI for traceable measurements provides the sound basis for mutual recognition in the global climate science and atmospheric monitoring communities and have been highlighted, for example, in the strategic plan for the **U.S. Climate Change Science Program**:[14]

> *… Instrument calibration, characterization, and stability become paramount considerations. Instruments must be tied to national and international standards such as those provided by the National Institute of Standards and Technology (NIST).*

Despite the IPCC proclamation that the Earth is warming and that this warming is attributable to human activity, many questions remained. For example:

- At what rate is the Earth's climate changing?
- How do we accurately distinguish, quantify, and effectively reduce anthropogenic GHG sources?
- If GHG emissions mitigation efforts were to be put in place, what are the measures necessary to assess their performance, against what baseline data, and what levels of accuracy and reliability are needed to assess their progress, and what temporal and spatial granularity is needed both long- and short-term?
- If geo-engineering solutions were to be considered, how will both short- and long-term effects be quantified to evaluate the benefits or harm that may occur?

Our ability to judge the effectiveness of proposed emission reduction efforts will be significantly advanced by improvements to the measurements and standards upon which such information rests. Such efforts require more accurate measurements of greenhouse gases in the atmosphere and data analysis methods to identify and quantify specific sources of GHG emissions (source attribution). To address these challenges, NIST initiated a research program in 2010 to develop:

- new measurement methods to determine the concentration of GHGs in the atmosphere as well as aerosols that affect radiative transfer in the atmosphere;
- accurate measurement methods to determine GHG emissions from point sources such as stacks of power generation and industrial plants;
- improved emission measurement capabilities and standards to support their accuracy; and
- methods to quantify GHG fluxes combining atmospheric observation and analysis (top-down) approaches and process-specific (bottom-up) methods, based on socio-economic data, that are complementary.

Accurate and precise atmospheric measurements of greenhouse gas (GHG) concentrations have revealed the rapid and unceasing rise of global GHG concentrations due to human socioeconomic activity.[3] In

2010, a National Academies report [15] stated that "*independent verification of the self-reported fossil CO_2 emissions of individual countries will require additional atmospheric measurements and improved tracer-transport estimates of emissions. The density and coverage of measurements would be improved …by establishing new stations near cities and other large local sources and in sparsely sampled regions*…"

1.a NIST Greenhouse Gas Measurements Program

NIST programs were expanded in 2010 to address the need for improved measurement science to underpin advances in GHG emission measurement technologies and standards. The Greenhouse Gas Measurements Program assesses measurement gaps and projected future metrology needs of the climate science research community and efforts of the private sector as it strives to implement the many facets of climate change mitigation. Quantification of GHG emissions and uptake is central to the success of this large-scale effort to realize a set of technologies and innovations that can, in various ways, mitigate, reduce, or remove greenhouse gases and other sources of excess atmospheric heating. These joint governmental and private sector efforts represent a large nascent industry. Its foundation not only rests upon technologies and methods to reduce emissions or remove CO_2 from the atmosphere, but as importantly the ability to effectively measure and report GHG emissions and uptake at local scales where most mitigation actions are implemented; such efforts are fundamental for effective mitigation policy, setting reduction targets, and achievement of such goals. The primary objective of NIST's GHG Measurements Program is to advance the metrological foundation for such measurements so that companies and localities have the trusted information necessary to create markets and ecosystems. These efforts can ensure that the U.S. is a world leader in this new industry.

Greenhouse gas emission fluxes, flows to or from the atmosphere, are measurements made in gas mixtures; these originate from powerplant stacks, vehicle exhausts, residential or commercial buildings, or methane leaks. These measurements are made by direct sampling the atmosphere or by remote sensing. GHG concentrations and determination of their rate of emission and absorption are the fundamental parameters of emissions quantification. GHG concentration measurements results must be related to international standards for undisputed reference. References are usually national or international standards established through an unbroken chain of comparisons. Such comparisons establish the traceability of standards;[1] such standards range from field standards found in powerplants and atmospheric observing networks to remote sensing methods that also require sensitive and accurate spectroscopic data referenced to these standards.

NIST's GHG Measurements Program was initially composed of seven sub-programs and associated projects as shown in Table 1. As described in the remainder of this report, these research directions addressed a range of metrology and standards needs ranging from advances in GHG concentration standards and their provision to the development and demonstration of emissions quantification in cities and their surrounding regions. Program evolution has consolidated some of the initial efforts as additional

[1] Traceability is the property of a measurement result whereby the result can be related to a reference through a documented unbroken chain of calibrations, each contributing to the measurement uncertainty. www.bipm.org/documents/20126/54295284/VIM4_CD_210111c.pdf

assessments were made, particularly the Distributed Emission Source and GHG Inventory & Regional Emissions Profile sub-programs which were initially conceived as separate efforts but were merged into NIST's Urban GHG Measurements Testbed or Urban Dome Program focused on urban emissions.

Table 1. Initial GHG Measurements Program Sub-Programs and Projects

1. Point Source Emissions Metrology
    - Continuous Emission Monitoring Test Bed
2. Distributed Emission Source Metrology
    - Area Flux Measurement Tools
    - Emissions Dispersion Analysis
    - Field Reference Standards
3. GHG Inventory & Regional Emissions Profiles
    - Dense Observing Networks
4. GHG Measurement Standards & Reference Data
    - Gas Concentration Standards
    - Atmospheric Lifetimes
    - Spectroscopic Reference Data
    - Reference Data for Climate Models
    - Documentary Standards & Assessment Methods
5. Field-Deployable Measurement Technologies
    - Frequency Combs IR Spectrometry
    - Advanced Photoacoustic Spectroscopy
    - Advanced Non-Dispersive InfraRed (NDIR) Measurements
6. Satellite Instrument Calibration
    - Optical Reflectance & Transmittance Standards
    - Microwave Standards
    - Thermal Infrared Standards
    - Scene Generation Technologies
7. Aerosol Measurement Science
    - Black Carbon Optical & Bulk Properties
    - Volatile Organic Compound (VOC) Aerosol Formation

As noted above, GHG concentration and bulk flowrate measurements, and the standards rendering these traceable to national measurement standards, are fundamental to all emissions quantification technologies. NIST sub-program efforts in GHG Concentration Standards and Point Source Emissions Metrology address EPA's Clean Air Act Amendment regulations requiring determination of SO_2, NO_x, and CO_2 emission amounts via installation of Continuous Emissions Monitoring Systems (CEMs) in most U.S. power plants. CEMs require traceable measurement of both gas concentrations and bulk stack flow rate. NIST created the NIST Traceable Reference Materials (NTRM) program in collaboration with the specialty gas industry [16] to meet the demand from plants for traceable field standards supporting EPA's ±2 % concentration accuracy requirement.

The Measurement Standards and Reference Data and Field-Deployable Measurement Technologies subprograms utilize NIST measurements and standards expertise. This research addresses a range of advanced measurements and standards needs that support climate science and technologies. The Satellite Instrument Calibration subprogram continues to address remote sensing needs of Earth-observing satellites and surface-based remote sensing. In recent years this subprogram has begun addressing measurement issues associated with optical methods for determination of CO_2 uptake and respiration-based emissions of vegetation, a critical component of the climate measurements puzzle. The Aerosol Measurement Science_subprogram is focused on atmospheric aerosols, particularly black carbon (BC) aerosols, a short-lived climate forcer (See Figure 1 below, right panel). BC aerosols, with significant infrared absorption, act on the atmosphere as do GHGs, i.e., absorbing heat radiated from the Earth and transferring it to the atmosphere. NIST research has focused on characterization of BC aerosols, optical properties that were poorly characterized at program outset. This finding was reinforced by a NIST workshop on the topic held shortly after program start. [17]

The next phase of the NIST Greenhouse Gas Measurements Program was focused on emissions from urban environments. Cities contribute over 70% of energy-related CO_2 emissions and account for around two-thirds of global energy demand while occupying a small portion of Earth's landmass.[18, 19] If current trends continue, global urban primary energy use and CO_2 emissions would grow by about 70% and about 50% respectively between 2013 and 2050.[18] Cities, and regions surrounding them, also have a unique capacity to address the effects of climate change compared with national governments. It is widely argued that cities are both part of the problem and part of the solution to addressing climate change.[19] Cities are attracting increasingly larger proportions of the global population, wealth, and productivity. More than half of the world's population (54.5% in 2016) lives in cities, and the number is projected to reach 70% – 6.7 billion people – by 2050; [20] cities also generate more than 80% of global Gross Domestic Product (GDP).[21]

In 2009, NIST had received a proposal from Purdue University to develop measurements of urban emission fluxes of GHGs in Indianapolis (the **INFLUX Project**), i.e., to make whole-city emissions measurements applying both atmospheric and emissions modeling and emission measurements. Indianapolis was considered an appropriate location for such a study because of its flat topography and relatively consistent weather patterns; at that time Purdue University had also begun development of a spatially explicit urban emissions model (the **HESTIA** Model).[22] The proposal included researchers from Purdue University (Chemistry Department), NOAA (Global Monitoring Laboratory) and Pennsylvania State University (Department of Meteorology and Atmospheric Science). The plan was to establish monitoring stations on two communication towers to the southwest and northeast across the city to capture incoming and outgoing atmospheric GHG concentrations and use aircraft flights, with GHG-monitoring instruments, to also determine a mass balance of GHG emissions. The INFLUX project was initiated in 2010 with research funds from the American Reinvestment and Recovery Act (ARRA) administered by NIST.

Shortly after initiation of INFLUX under ARRA, NIST received a proposal from Penn State University to investigate the utility of dense observing networks and atmospheric inverse modeling in urban environments. Earlier GHG observing networks, such as NOAA's tall tower system, were directed at regional-to-continental-to-global data and analyses.[23] These networks were spatially sparse relative to the environs of cities. The INFLUX proposal involved establishing a spatially dense, 12 node surface-observing network having most nodes within or near to the city to provide 24/7 GHG concentration data. An atmospheric inversion analysis would transform these concentration data to emission fluxes with a spatial resolution consistent with the micro-meteorological models used at a city scale, i.e., 1 km to 3 km grid spacing, to represent and simulate meteorological dispersion and transport and land surface parameters. The spatially explicit emissions data provided by the HESTIA model represented a unique approach to emissions quantification and one of the early manifestations of combining the two approaches.

The Los Angeles (LA) Megacity Carbon Project, established in 2013, initiated NIST's Urban GHG Measurement Testbed System, joined INFLUX as the second NIST urban testbed. A third testbed was begun in 2014 to extend throughout the Northeast Corridor with an extensive surface network complemented by aircraft observations.

The purpose of the testbed system was to demonstrate and develop urban GHG emissions measurement approaches composed of the combination of spatially explicit emissions modeling and atmospheric GHG concentration measurements and models. Each testbed is characterized by differing meteorological conditions, city / regional typology, emissions and vegetative properties, and others. Indianapolis is far from compounding emission sources, surrounded by highly productive agriculture, located on a flat landscape far from surface topology that complicate application of the weather models required to add the velocity component to the atmospheric concentrations measured by surface and airborne methods. This combination provides the means to quantify surface emission fluxes. Potential future satellite observations should an overflight track come near it, would add another dimension to INFLUX and the other testbeds. The LA project adds complexity to all aspects of the measurement and analysis approach and include satellite overpasses. The Northeast Corridor Project contains four major metropolitan cities, extensive forested and agricultural areas, effects of oceans on atmospheric conditions, as does LA, and is sufficiently large to contain several OCO-2 and OCO-3 satellite tracks[2] as well as those of satellites of other nations. These multiple tracks and conditions necessitate high-fidelity modeling and measurement capabilities, and to some degree represent a microcosm of the United States.

1.b International Outreach

NIST developed a strategic approach to promote closer working relationships between the international metrology and climate science/atmospheric monitoring communities. Contacts were established with organizations within Europe, South America, and China. A workshop was held in Paris in 2010, organized by the World Meteorological Organization (WMO) and the International Bureau of Weights and Measures (BIPM), to bring together the metrology and meteorology communities to identify specific research needs and potential collaboration areas.[24] At that meeting, WMO and BIPM signed an agreement to further their collaborative efforts. A meeting was arranged in April 2012 in Paris at the laboratories of LNE, the French National Metrology Institute, to brief potential future collaborators about the NIST Program on GHG Measurements and the INFLUX Project.

A workshop was organized in May 2012 at the California Institute of Technology to discuss the preliminary findings from the INFLUX Project and to plan extension of this work to Los Angeles, a megacity of similar population to Paris, but with a much more challenging topography impacting meteorological conditions of the region, the South Coast Air Basin. Researchers from Arizona State, California Institute of Technology, California Air Resources Board (CARB), Earth Networks, JPL/NASA, LBNL, NIST, NOAA, Penn

[2] OCO-3 is located on the International Space Station as is the more recent EMIT instrument. https://www.jpl.nasa.gov/news/nasa-mission-excels-at-spotting-greenhouse-gas-emission-sources

State, Purdue, SIO, and UCLA participated in the workshop. Researchers from Laboratory of Climate and Environmental Science (LCSE) in France were also present and shared their Paris findings. This workshop was also utilized as an opportunity to raise interest in Brazil and China in GHG measurements. Invitations were extended to staff of INMETRO and NIM, NMIs of Brazil and China, respectively, to participate in the workshop; both NMIs were represented and expressed serious interest in initiating similar activities in Brazil and China.

NIM (China) staff were interested in developing a program on GHGs as well as aerosol measurements, since these represent important areas of concern for China. NIM staff started planning a workshop on GHGs and Aerosol Measurements at NIM; detailed information was provided to NIM about potential participants (from the USA as well as from within China) to assist them with organization of the workshop. These efforts resulted in a very successful workshop, which was held during the first week of September 2012 at NIM in Beijing; this workshop and subsequent visits by NIST staff were instrumental in establishing urban testbed projects in Zhengzhou and other locations in China.

NIST, in collaboration with Organization of American States (OAS), the Inter-American Metrology System (SIM, by its acronym in Spanish), and the U.S. State Department, organized five workshops in Central and South America, including Mexico, Guatemala, Uruguay, Colombia and Jamaica, to increase awareness of climate change issues and encourage monitoring of greenhouse gas emissions and development of renewable energy technologies.

1.c Continued Impact of UNFCCC Activities

In 2014, IPCC completed its **Fifth Assessment Report**,[25] which concluded:

- *Human influence on the climate system is clear, and recent anthropogenic emissions of greenhouse gases are the highest in history. Recent climate changes have had widespread impacts on human and natural systems.*
- *Warming of the climate system is unequivocal, and since the 1950s, many of the observed changes are unprecedented over decades to millennia. The atmosphere and ocean have warmed, the amounts of snow and ice have diminished, and sea level has risen.*
- *Anthropogenic greenhouse gas emissions have increased since the pre-industrial era, driven largely by economic and population growth, and are now higher than ever. This has led to atmospheric concentrations of carbon dioxide, methane and nitrous oxide that are unprecedented in at least the last 800,000 years. Their effects, together with those of other anthropogenic drivers, have been detected throughout the climate system and are extremely likely to have been the dominant cause of the observed warming since the mid-20th century.*

At **COP 20**, held in Lima, Peru in 2014, participants distanced themselves from the concept of MRV and started discussing the concept of **Nationally Determined Contributions** (NDCs), which seemed to be much more acceptable to developing countries. NIST and NASA gave a summary presentation of the initial results from the LA Megacity project, NIST's second Urban GHG Measurements Testbed, describing urban emissions research.

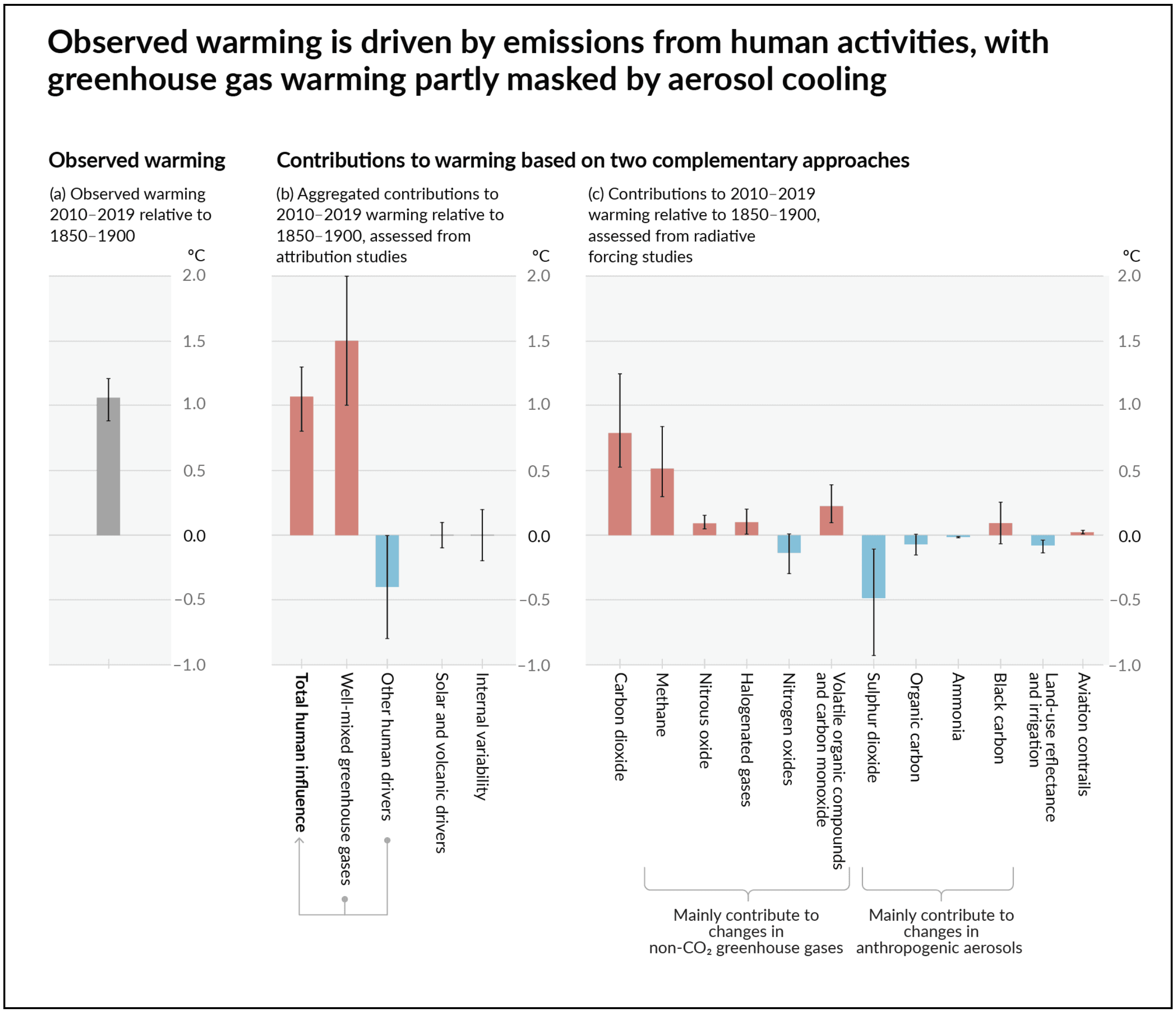


Figure 1 – The Role of Greenhouse Gases and Aerosols in Global Warming as Predicted by IPCC https://www.ipcc.ch/report/ar6/wg1/chapter/summary-for-policymakers/ (2021)

The following Conference of the Parties (**COP 21**) was held in Paris in 2015. The concept of Nationally Determined Contributions was agreed to and would become the heart of the Paris Agreement and the tool for achievement of its long-term goals. NDCs embody efforts by each country to reduce national emissions and adapt to the impacts of climate change. The Paris Agreement (Article 4, paragraph 2) required each Party to prepare, communicate, and maintain successive NDCs that it intended to achieve. Parties were expected to pursue domestic mitigation measures, with the aim of achieving the objectives of such contributions. The 2015 Paris Agreement concluded:

- *Recognizing that climate change represents an urgent and potentially irreversible threat to human societies and the planet and thus requires the widest possible cooperation by all countries, and their participation in an effective and appropriate international response, with a view to accelerating the reduction of global greenhouse gas emissions,*

- *Also* recognizing *that deep reductions in global emissions will be required in order to achieve the ultimate objective of the Convention and emphasizing the need for urgency in addressing climate change,*
- *Acknowledging that climate change is a common concern of humankind, Parties should, when taking action to address climate change, respect, promote and consider their respective obligations on human rights, the right to health, the rights of indigenous peoples, local communities, migrants, children, persons with disabilities and people in vulnerable situations and the right to development, as well as gender equality, empowerment of women and intergenerational equity.*

Article 6 of the Paris Agreement recognized the importance of efforts by sub-national actors, governmental organizations below the national level, and private and non-governmental organizations as a mechanism to promote mitigation and adaptation to climate change that, among other things, incentivize and facilitate participation in GHG emissions mitigation by public and private entities. It also created the basis for trading in GHG emission reductions (or "mitigation outcomes") across countries, thereby recognizing the potential benefit of carbon markets in mitigation efforts. In addition, Article 14 established the concept of a "global stock take" to evaluate implementation of the Agreement and to assess collective progress towards achieving its purpose and long-term goals.

UNFCCC's IPCC has continued to emphasize the role of greenhouse gases and other atmospheric agents in global warming (See Figure 1) [26] Those responsible for implementing efforts to reduce greenhouse gases (GHG) need more timely and accurate emissions data to guide their activities. But measuring GHG emissions poses significant technical and scientific challenges. During the 2016 Conference of the Parties (COP21) of the UNFCC in Paris, 162 nations submitted emission reduction goals in the form of Nationally Determined Contributions (NDCs). The U.S. NDC states that, "*the United States intends to achieve an economywide target of reducing its greenhouse gas emissions by 26–28 percent below its 2005 level in 2025 and to make best efforts to reduce its emissions by 28 percent."* Other developed nations have put forth NDCs of similar magnitude. To judge progress toward these NDCs, and determine whether they are ultimately attained, will require measurement uncertainties consistent with target reduction amounts. For example, many NDCs translate to yearly reductions of 1% to 3%. Confidence in monitoring on-going reduction rates will require measurement uncertainties at or below those levels. Cities and metropolitan areas are major contributors to a nation's total GHG emissions inventory. Therefore, measurements that better quantify urban emissions can greatly increase the credibility of a nation's GHG emissions data. [27]

1.d Continued International Activities and Workshops

In March 2015, the monthly global average concentration of carbon dioxide in the atmosphere surpassed 400 micromoles/mole (ppm) for the first time since modern records began.[28] BIPM organized a **Workshop on Global to Urban Scale Carbon Measurements**.[29] The aims of the workshop were to identify requirements for advances in measurements, standards, and reference data; to present instrument comparisons for carbon measurement and related climate variables; and to advance the measurement science base underpinning a comprehensive, global greenhouse gas monitoring system for routine use as a method for validating greenhouse gas flux and emission data.

In 2016 NIST held a **Workshop on Urban Greenhouse Gas Measurements**.[30] The aim of the workshop was to bring together researchers engaged in urban GHG projects across the U.S. and to identify detection and quantification challenges, discuss the status of modelling efforts, identify currents issues of interest, and develop an understanding of challenges specific to each urban area.

To expand the GHG measurement related activities in the international community and promote coordination of these activities, NIST (along with NOAA and NASA) supported WMO efforts to establish an **Integrated Global Greenhouse Gas Information System** (IG^3IS). This complements WMO's **Global Atmosphere Watch** (GAW) Program, which had developed a coordinated monitoring system to enhance understanding of global atmospheric composition and its change. These measurements were made primarily in isolated areas away from urban centers. (NOAA's Mauna Loa observatory is an example sampling the atmosphere at an altitude of ≈ 3,400 m (≈ 11,000 ft) and was the first of these and has been emulated by several countries who participate in this WMO-led monitoring effort.) The aim of the IG^3IS Program was to promote measurements near urban environments where most of the GHG emissions originate and to emphasize the importance of atmospheric measurements to complement GHG inventories developed from socio-economic data.

IG^3IS intensified efforts of more than a decade to promote the use of atmospheric measurements and analyses to complement GHG inventory methods to enhance their reliability. The IPCC **Task Force on National Greenhouse Gas Inventories** (TFI) had been discussing this topic for some time. Finally, at the **COP 25** held in Madrid in 2019, the "2006 IPCC Guidelines"[31] were refined to include guidelines for atmospheric measurements as follows:

> ***Comparison of greenhouse gas emission estimates with atmospheric measurements***:
>
> *Guidance on comparison of greenhouse gas emission estimates with atmospheric measurement has been updated and elaborated to reflect the state of science for atmospheric measurements and their application to improving national greenhouse gas inventories. These approaches can be used to provide additional scientific verification of inputs and results for particular categories and gases, and therefore help countries to target areas of uncertainty. The most notable advances were achieved in the application of inverse models of atmospheric transport for emission estimates at the national scale. Thus, atmospheric measurements are being used to provide useful quality assurance of the national greenhouse gas emission estimates. The guidance highlights key components and steps that can be applied when using atmospheric measurements and inverse models for comparison with inventory emission estimates as part of a country's overall QA/QC and verification system."* [32]

On January 30-31, 2020, a workshop was held at NIST in Gaithersburg, MD to review recent developments in measurement capabilities and technologies addressing urban GHG emissions, to assess the state of the art and remaining scientific challenges, and to identify ways to serve the needs of the specific stakeholders, including large municipalities where mitigation actions are planned or underway. Plenary lectures provided an overview of current research activities and capabilities. Panel discussions were held on scientific and technical successes and knowledge gaps, on researchers' experiences with domestic and international stakeholders, and on ways to map technical capabilities to stakeholder needs. During the discussions over the two-day workshop, there were several themes that workshop participants emphasized about the scientific accomplishments of the community and the remaining technical challenges.[33]

COP 26, which was delayed because of the COVID pandemic, was held In November 2021 in Glasgow, UK. Countries reaffirmed the Paris Agreement goal of limiting the increase in the global average temperature to well below 2 °C above pre-industrial levels and pursuing efforts to limit it to 1.5 °C.[34] They stressed the urgency of action "in this critical decade," when carbon dioxide emissions must be reduced by 45 % to reach net zero around mid-century. But with present climate plans – the Nationally Determined Contributions — falling far short on ambition, the Glasgow Climate Pact called on all countries to present stronger national action plans the next year. Countries ultimately agreed to a provision calling for a phase-down of the use of coa-fired power plants and a phase-out of "inefficient" fossil fuel subsidies – two key issues that had never been explicitly mentioned in decisions of UN climate talks before, despite coal, oil and gas being the main drivers of global warming. Countries also reached agreement on the remaining issues of the so-called Paris rulebook, the operational details for the practical implementation of the Paris Agreement. Among them were the norms related to carbon markets, which will allow countries struggling to meet their emissions targets to purchase emissions reductions from other nations that have already exceeded their reduction targets.

To continue discussions of the international community on GHG measurements in urban environments, BIPM and WMO organized a joint workshop in 2022 in Paris. The aim of the workshop was to:

- Present progress and identify requirements for further development of advanced measurements, standards, reference data, comparisons, calibrations supporting the physical science basis for and adaptation to climate change, and
- Identify stakeholders' metrology needs, assess current metrological techniques, analyses, and modeling capabilities, and identify gaps in quantifying greenhouse gas emissions and uptake for effective actions on mitigating climate change and its impacts.

The output of the workshop provided a set of recommendations on key technical challenge areas for metrology over the next decade.[35]

On June 15-16, 2022, there was another workshop, organized by NIST and the Environmental Defense Fund, held in Washington, DC, to discuss "Quantifying methane emissions across natural gas infrastructure in urban environments". [36]

There has also been an increasing discussion about the need for international standards to validate measurements of GHG emissions and assessment of the impact of mitigation efforts, especially those taken up by the private sector, and to provide credibility for claims for carbon credits. NIST has initiated an effort to develop an ISO Standard through ISO TC 207 (Environmental Management)/ SC 7 (Greenhouse Gas and Climate Change Management and Related Activities). A New Work Item Proposal (NWIP) has been submitted on "Greenhouse Gases – Atmospheric Measurements of Greenhouse Gas Emissions in Urban Environments" to ISO TC 207/SC 7.

* * * * * *

Accurate GHG emissions quantification is fundamental to assessment of the impact of mitigation efforts. The emission determination process usually starts with an estimate of GHG emissions from socio-economic data (bottom-up approach) largely obtained at the emission or uptake process level; these include emissions from power-generation plants, industrial operations, commercial and residential buildings, landfills, and vehicles operating within a city's domain (See Figure 2). Atmospheric monitoring methods (top-down approach) are sensitive to all emissions and uptake. They use background concentration measurements of the incoming atmosphere to the analysis domain's "control volume", i.e., the urban environment and its immediate surrounding region, as defined by the atmospheric transport model used in the analysis scheme. Also, modeling of anthropogenic emissions and uptake, and emissions by the biogenic environment within and around the city, are coupled with computational tools to estimate GHG amounts exchanged with the atmosphere. Figure 2 illustrates the breadth of emissions and uptake source types from the facility scale of power generation and other processes to the global scale. Different measurement approaches are used to both measure GHG concentrations and to determine their transport in either individual processes or within the atmosphere.

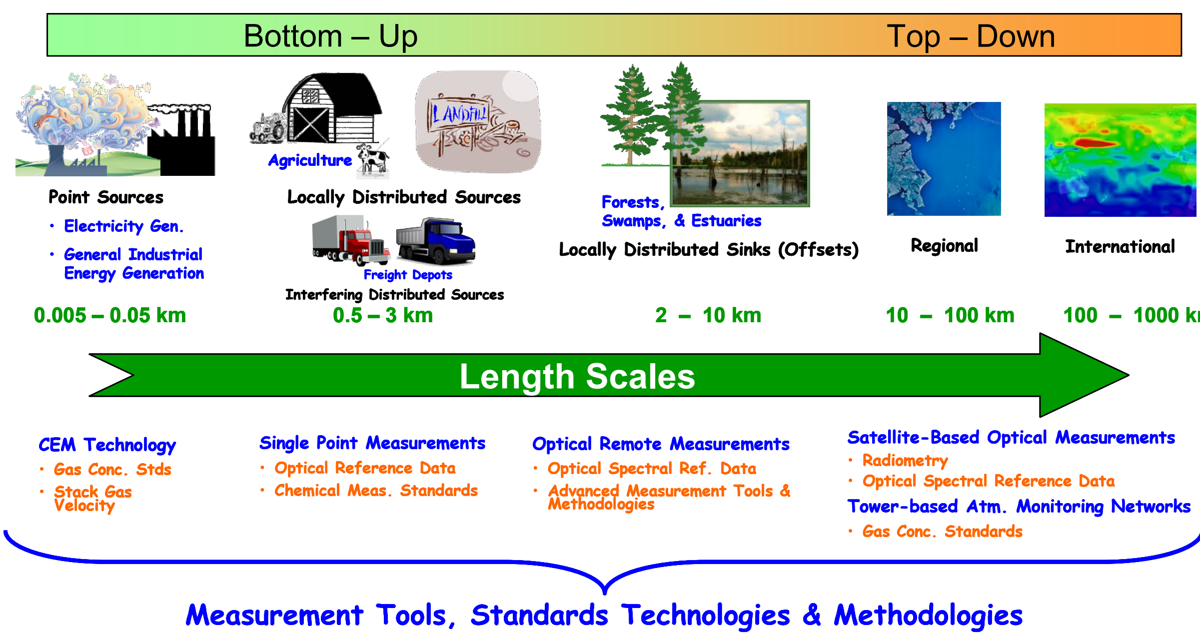


Figure 2. Emission models, ("**bottom-up**" approaches) based on spatially explicit socio-economic data, account for emissions from point sources as well as distributed sources. Atmospheric monitoring ("**top-down**" approaches) using satellite, aircraft, or tower-based measurements are utilized to characterize GHG fluxes into the atmosphere from urban or regional areas.

In the following pages, a summary is presented of the scientific developments achieved over the last decade aimed at:

a) enhancing GHG measurement capabilities;
b) advancing computational tools to estimate emissions from socio-economic activities, models to estimate biogenic emissions and uptake, and atmospheric inversion models;
c) advancing data analysis methods for urban GHG source attribution; and
d) developing Urban Testbeds to quantify GHG fluxes from cities.

These tools are being developed with a focus on studying city emissions via use of NIST's Urban GHG Measurement Testbeds established in Indianapolis, Los Angeles, and the Northeast Corridor of the U.S. Finally, a summary is presented of the collaborations established by NIST with national and international organizations to address the global challenge of climate change.

2. Enhanced NIST Capabilities for Measurement of GHGs and Aerosols

Greenhouse gases (GHGs), such as carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), the hydrofluorocarbon gases, sulfur hexafluoride (SF_6), and nitrogen trifluoride (NF_3), have the most impact for trapping heat in the earth's atmosphere and are the focus of emissions reduction policy discussions and proposed Congressional legislation aimed at mitigating the climate impact of GHGs from human activities. There is a growing acceptance that human activities have a strong influence on some climate effects, but there remains a continued and pressing need to improve the quality of long-term climate data records and the accuracy of pertinent measurement capabilities. Improving measurement accuracy will better illuminate processes in the atmosphere, on the land, and in the oceans. Better understanding of the mechanisms operating and controlling climate and its impact will also benefit climate model predictive capabilities. An excellent example of the need for improved understanding of atmospheric and land-based processes is the influence of atmospheric particulates on the radiative balance of the Earth. The IPCC Fourth Assessment Report identified aerosols as a significant source of atmospheric forcing for the first time and noted the large uncertainties stemming from significant deficiencies in our knowledge of the optical properties of particulates, particularly black carbon particulates, and their impact on atmospheric heating and cooling. Initial NIST research activities focused on enhancing our capabilities for measuring GHG and aerosol concentrations in the atmosphere.

Measurements of greenhouse gases in the atmosphere must be highly accurate to provide useful information about atmospheric emissions. GHGs emitted by sources get diluted in the atmosphere. Global mean values of the three highest concentration GHGs are shown in Table 2.[37] To discern GHG concentration changes, CO_2 concentrations must be quantified at the order of one part per million (ppm or µmole/mole); CH_4 and N_2O measurement accuracies have to be at the parts per billion level (ppb or nanomole/mole) or better. Furthermore, practical measurements with analyzers located on an aircraft or communication towers must utilize *in situ* measurement capabilities to provide measurements with high spatial and temporal resolutions. All these requirements have driven the scientific community to improve its measurement capabilities; NIST has been a leader in this effort, improving the sensitivity of GHG measurements and their accuracy and reliability and reflecting these accuracy levels in its primary gas concentration standards.

Table 2. Global Atmospheric Mean Abundance (Concentration) of GHGs (2023)

Carbon Dioxide (CO_2)	417.9 ± 0.02 ppm
Methane (CH_4)	1923 ± 2 ppb
Nitrous Oxide (N_2O)	335.9 ± 0.1 ppb

One of the early focus areas of the NIST program was improving the accuracy of GHG emission measurements from smokestacks of power generation plants and large industrial plants. Power generation plants account for 25% of U.S. GHG emissions; industrial plants account for 24%. [38] Commercial and residential buildings account for 13% of U.S. GHG emissions, agriculture for 11%, and transportation for 27%. Because more than half of GHG emissions come from distributed sources, i.e., sources other than power plants and industrial smokestacks, methods to measure emissions on an urban or regional basis are necessary to quantify such emissions and assess the results of mitigation efforts.

Most of these measurements require calibration standards to ensure the accuracy of GHG measurements. As the demand for such measurements and standards increases, manufacturing and distribution of internationally recognized calibration standards will represent a challenge. NIST is also developing absolute methods for measurement of GHG concentrations to obviate the need for GHG calibration standards.

2a. **Smokestack Measurements of GHGs**

Accurate measurement of GHGs emitted from the smokestack of a power plant or a large industrial facility may be regarded as a relatively easy task. However, the complexity of the flow fields in the smokestack makes it challenging, as illustrated in Figure 3. Flow velocities and direction can vary significantly, particularly near the stack inlet where high velocities and flow direction changes are strongly influenced by upstream structures that disturb flow properties. Their influence can continue to the top of the stack as illustrated in Figure 3, computational fluid dynamics modeling result of flow in a typical powerplant stack configuration. In fact, presently used methods to measure CO_2 and other emissions from smokestacks have errors of 20 % or more depending on the level of swirl in the flow.[39, 40] [41] The objective of this NIST project was to measure stack emissions with an uncertainty on the order of 1%

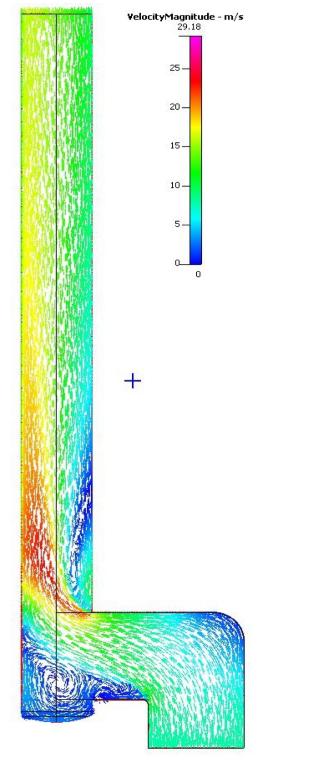


Figure 3. Axial flow properties modeled for a typical smokestack. (Courtesy of Elizabeth Moore)

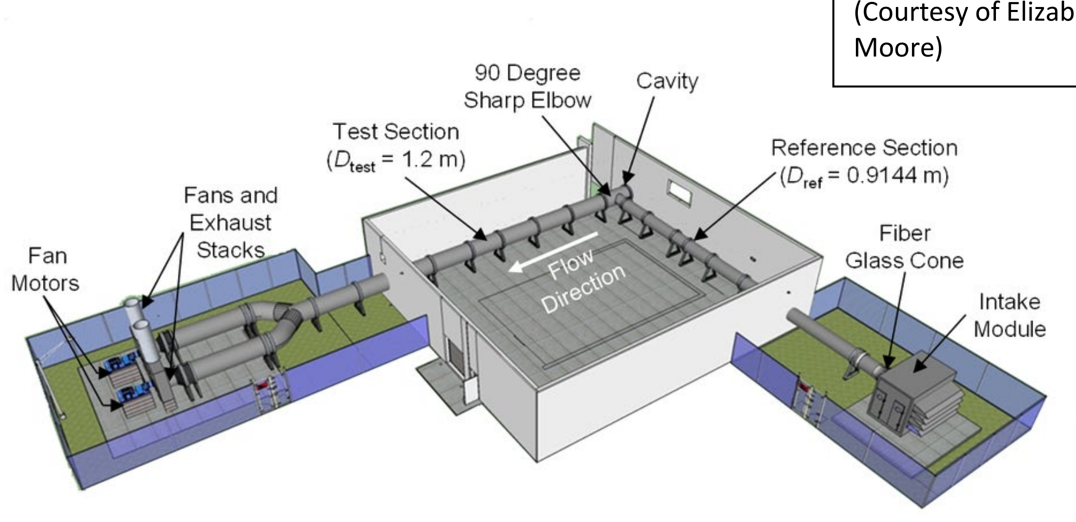


Figure 4. NIST's Smokestack Simulator. The test section has a length of 28 m and a diameter of 1.2 m (4 feet) which is approximately 1/10th the diameter of a power-plant stack. doi.org/10.6028/NIST.SP.1228

at a reasonable cost and to develop improved methods for calibration of *in situ* ultrasonic meters.

NIST and power industry partners, represented by the Electric Power Research Institute (EPRI), using the newly developed NIST smokestack simulator (NSSS) and field tests at power plant stacks, demonstrated that GHG emissions from smokestacks could be measured with an uncertainty of 2 %, regardless of the swirl level in the flow.[42]

The newly designed and built NIST 1/10th Scale-Model Stack Simulator, depicted in Figure 4, has an inlet cone and reference (upstream) section that draws in ambient (outside) air and generates a swirl-free, fully developed turbulent flow.[43] Two fans at the exit of the facility draw air into the intake module where it is accelerated through a bell mouth into the D_{ref} = 0.9144 m diameter reference section. The refence section is equipped with an 8-path ultrasonic flow meter (USM) that measures the volumetric flow (Q_{ref}) with an expanded uncertainty of 0.5 %. A sharp 90 ° elbow at end of the reference section generates asymmetric, swirling flow in the downstream test section with a diameter of D_{test} = 1.2 m. The average velocity in the test section ranges from 6 m/s to 26 m/s, which corresponds to the range of axial velocities in industrial-scale smokestacks. The flow rate was measured ten pipe diameters (10 D_{test}) downstream from the sharp corner 1) with an ultrasonic Continuous Emission Monitoring Systems (CEMS) flow meter and 2) by performing an EPA protocol known as the Relative Accuracy Test Audit (RATA) using four different pitot probes. The CEMS flow meter consisted of three individual paths including a vertical path and two horizontally oriented crossing paths (*i.e.*, X-pattern) all installed in the same meter body. Each path is oriented at 45° angle with the test section axis and comprises two transducers that both transmit and receive ultrasonic pulses.

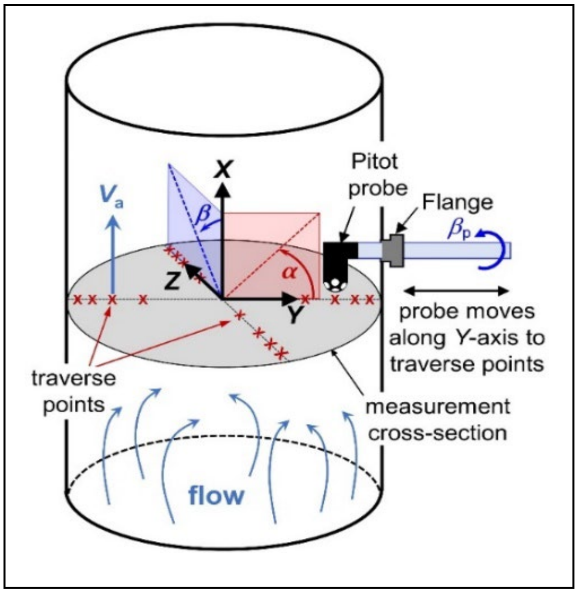


Figure 5. Schematic of the flow RATA where a pitot probe measures the axial velocity (V_a) at points along a traverse across the smoke stack. Flow angularity of the flue gas is depicted by the flow's pitch (*a*) and yaw (*b*) angles, and the rotation of the pitot probe about its axis (required for probes using the nulling method) is depicted by (b_p). doi.org/10.6028/NIST.SP.1228

Figure 4 shows a tee connecting the reference section to the downstream test section. (One leg of the tee is a small "dead" volume.) The tee generates counter-rotating vortices in the test section, thereby simulating the flow generated by a typical connection between a power plant's pollution control system and its stack. Obstacles (such as perforated plates) can be inserted in the test section to further complicate the flow. The exhaust fans generate flow velocities typical of power-plant stacks in the 28 m-long test section, which has a diameter of 1.2 m (4 ft).

The smokestack simulator generates well-characterized flows for evaluating flow meters installed in smokestacks for EPA-approved CEMS, usually transit-time ultrasonic flow meters. The facility was also utilized to evaluate profiling methods used to periodically calibrate the CEMS. The profiling methods position airspeed sensors at prescribed positions across the stack and integrate them to obtain the bulk flow. These RATAs follow EPA protocols and use differential pressure pitot sensors called S-probes or 3-D probes. (See Figure 5)

As part of this program, NIST and industrial collaborators developed new methods (e.g., the NIST non-nulling method) that are faster, more accurate, and easier to apply than those presently used. The NIST non-nulling method (NNN method) [44] and new probe designs have been tested in the NIST Smokestack Simulator, and in multiple field tests at power plants. The stack simulator is preferable for assessing the accuracy of the NNN method because it has a low uncertainty reference (0.65 %). Field tests are essential for demonstrating the practicality, robustness, and time efficiency of the NNN method. NIST researchers demonstrated that X-pattern ultrasonic CEMS provide excellent compensation for swirl. In the stack simulator the X-pattern reduced errors of a single path measurement from 17 % to 1 %. Load-dependent errors in the calibrated single path CEMS were reduced from 3% to less than 0.5 %. Similar trends were observed in field stack tests.

Field tests were used to demonstrate several capabilities. NIST researchers used a spherical probe (corrected for imperfect yaw-nulling) as the flow reference for field work, which is expected to show a less than 1 % uncertainty. The NIST Non-Nulling method was demonstrated to provide measurements that were four times faster than an S-probe and five times faster than a spherical probe. [45] [46] The increased speeds enable a denser sampling of stack cross-section velocities, which in turn, improves measurement accuracy. NIST has received a patent for development of the non-nulling hemispherical probe, which is a first of its kind for stack flow applications.[47] These capabilities for improved measurements of greenhouse gases emissions from smokestacks were shared with stakeholders at a NIST Workshop.[48]

A small business initiative research (SBIR) grant was awarded to a stack testing company in 2019. The stack testing company developed a rugged, stack-capable data acquisition system that supports the non-nulling measurement protocol. The company also developed, and field tested, an automated probe movement system to maximize the effectiveness and appeal of the non-nulling methodology to the stack testing community. The time-savings that are realized with the non-nulling methodology will be a significant benefit to industry, as plants can reduce the time required to maintain off-load conditions.

NIST is currently involved in writing ASME standard PTC 19.25 “Non-Nulling Gas Velocity Measurement” that describes: 1) non-nulling probe calibrations, 2) field application, and 3) uncertainty. Once promulgated, the non-nulling test standard will be available to plants and testing vendors that want to use it for official RATA test projects.

2b. **Measurements of Carbonaceous Aerosols**

Light absorption and scattering by aerosol particles can be one of the largest sources of atmospheric heating and cooling. Quantification of this effect is challenging due to the complexity and variability of aerosol species in the atmosphere; these lead to large uncertainties in their radiative forcing and hence on their climate impact. To address this complexity and reduce uncertainty, the Aerosol Optical Properties project at NIST has focused on developing methodologies to quantitatively characterize the chemical, physical and optical properties of atmospherically relevant single- and multi-component aerosols. NIST investigations were aimed at development of: a) quantitative measurements for aerosol absorption, extinction and scattering traceable to the SI; and b) aerosol materials with known spectroscopic properties for instrument calibration and intercomparison, to facilitate their applications to complex systems.

NIST efforts on quantitative measurements of aerosol absorption, extinction and scattering specifically relied on an aerosol particle mass analyzer (APM) so that the particles (and their optical properties) could be classified as a function of mass, an SI-traceable quantity.[49] This is in comparison to most other aerosol measurements where particles are sized by electrical mobility (as by a Differential Mobility Analyzer, DMA) or aerodynamic diameter (as by an Aerosol Aerodynamic Classifier, AAC). Beyond developing a routine method for mass-based optical classification, NIST also developed a method to measure "high-resolution" absorption spectra of aerosol particles utilizing a photoacoustic spectrometer with a supercontinuum laser spanning from visible to near-infrared (IR) (500 nm to 840 nm).[50] In addition to this manuscript, a book chapter on the absorption spectroscopy of black and brown carbon aerosol in the visible was published. [51]

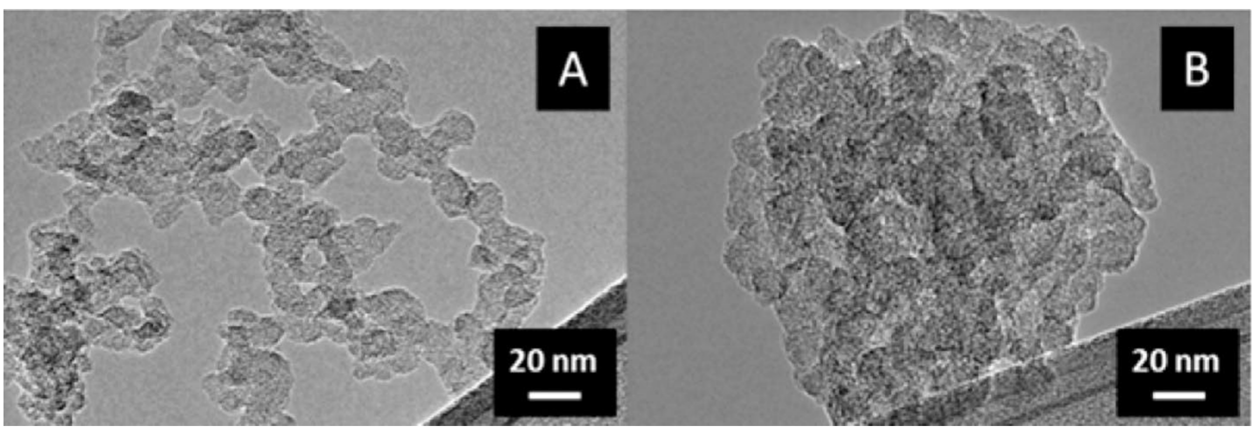


Figure 6 - TEM images of lacey (A) and compact (B) soot with a mobility diameter of 150 nm doi.org/10.1021/es4041804

Photoacoustic measurements are often biased low by humidity; particle adsorbed water preferentially evaporates *versus* being transformed into sound for detection by the microphone in the photoacoustic spectrometer. However, as the supercontinuum laser is quasi-continuous (78 MHz repetition rate), rather than a continuous-wave laser which is traditionally used, the measured absorption was higher than expected in the presence of humidity and was attributed to the fact that mass transfer is faster than energy transfer.[52] Additionally, limitations were identified to particle separations and classification by a tandem DMA-APM when the particles are small, have a low density (*e.g.*, some organics) or are highly non-spherical (e.g., soot aggregates).[53] It was also demonstrated that non-idealities in the collection efficiency of an aerosol impactor upstream of size and mass selection can modify the uncertainty of extinction measurements as a result of biases from multiply charged particles. [54]

Another output of this project was identification of a candidate material to act as a black carbon surrogate (Cab-O-Jet 200 from Cabot Corp.) for the calibration of aerosol absorption measurements. This material is referred to as "carbon black". It was demonstrated that carbon black could be used to calibrate photoacoustic spectrometers and filter-based measurements.[55] For photoacoustic spectrometers, carbon black allows performance of a calibration in a couple of hours *versus* what was previously a multi-day affair. [56] In addition to calibrations, carbon black was sent to twelve laboratories around the world for the first-ever aerosol photoacoustic spectrometer intercomparison study in which the instruments were not co-located. [57]

Beyond the development of a metrological method and identification of carbon black as a calibration material, NIST has applied its measurements to climate relevant systems. In one instance, the extinction and absorption properties of nascent lacey soot from a diffusion flame was compared to soot that had been collapsed through water condensation and evaporation. It was found that, for the immature soot measured here, collapse did not affect light absorption but did increase extinction as a result of increased scattering.[58] It was also demonstrated that the packing density of rigid, aggregated subunits that are compacted under low compression into larger structures (such as the collapsed soot case) was independent of particle size, remaining essentially constant, spanning the nanometer-sized soot to micrometer-sized pharmaceuticals.[59] Because of the low uncertainties of the optical measurements, it was possible to compare retrievals of aerosol refractive indices (a key aerosol input parameter for climate modeling) using size- and mass-selection to those performed when measuring the full distribution of particles; these methods correspond to lab-based and field-based scenarios, respectively.[60] It was shown that the full distribution retrievals yielded significant deviations that were a function of the particle type and size distribution measured and that these differences could lead to errors in the forward calculated optical properties of up to 26 %.

The broadband absorption spectra have been broadly applied as well. First, the wavelength dependent absorption enhancement of carbon black mixed with absorbing and non-absorbing materials was demonstrated.[61] Next, the quasi-material-dependent absorption spectra of particles generated from smoldering biomass burning of common woods was investigated. It was shown that oak, western redcedar and blue spruce exhibited unique absorption spectra while the spectra of hickory, mesquite and bald cypress were all similar.[62]

2c. **Advanced** M**easurements of Greenhouse Gas Concentrations**

Introduction of the concept of Cavity Ring-Down Spectroscopy (CRDS) in the 1980's by O'Keefe and Deacon, using pulsed laser sources, opened up a new horizon for spectroscopists.[63] The new technique provided the potential to significantly improve sensitivity of absorption measurements as well as the ability to make quantitative concentration measurements.[64] In CRDS, circulating laser power is stored in a high-finesse optical cavity containing the sample, and the passive decay of this power is monitored with the aid of a detector that measures the intensity of light transmitted through one of the mirrors. In an empty cavity formed by mirrors of 99.99% or higher reflectivity with a separation of 0.1 to 1 m, the light can be stored for microseconds during which time it makes thousands of round trips. The intensity of the circulating power decreases exponentially in time with a rate determined by the reflectivity of the mirrors and the length of the cavity. Therefore, by measuring the time constant for the intensity decay of the light stored in a cavity, referred to as the ring-down time τ, the reflectivity of the mirrors can be determined. When the cavity contains an absorbing gas, an additional optical loss occurs which causes a decrease in the ring-down time. By recording how the inverse of the ring-down time varies as a function of frequency of the incident light, the CRDS spectrum of the gas is obtained.

NIST researchers appreciated the potential utility of CRDS for gas concentration measurements and initiated a research program. The technique was initially applied to measurements of diatomic oxygen.[65] It was soon realized that finite laser bandwidth effects tended to excite multiple optical cavity modes making the characteristic light intensity decay non-exponentially, resulting in an underestimation of the absorptive losses.[66] Single-mode CRDS was also shown to mitigate laser bandwidth complications thus providing high accuracy gas density and line intensity measurements.[67] Cavity-enhanced spectroscopies have since found many applications to make measurements in the atmosphere, in reacting and non-reacting flow systems.[68]

Recently, climate scientists have been developing a new generation of spectrometers for NASA, NOAA and other agencies to be installed on satellites (such as OCO-2/3, GOSAT, MIPAS, CarbonSat, ASCENDS, TROPOMI, etc.) that are aimed at measuring atmospheric greenhouse gases, most notably carbon dioxide and methane. Similarly, international ground-based networks such as the Total Carbon Column Observing Network (TCCON), the Network for the Detection of Atmospheric Composition Change (NDACC), and WMO's Global Atmosphere Watch (WMO-GAW) program, and more recently established Global Greenhouse Gas Watch (WMO-GGGW) program, are also dedicated to monitoring atmospheric CO_2 and methane. This measurement capability is critical to improving our understanding of atmospheric concentration variations, which arise from the various sources, sinks, and atmospheric circulation patterns. These "field" measurements are arguably the most demanding ever attempted using absorption spectroscopy. To ensure the reliability and long-term continuity of these measurements, spectrometer operations rely on reference data of the highest quality and traceability to the SI through national GHG concentration standards. These measurements will also be critical to the next generation of satellite-borne instruments, which may rely on laser technology, rather than reflected sunlight, to measure diurnal and seasonal variations in atmospheric greenhouse gases.

In the early 2000's, NIST established a program to develop new measurement tools to provide the spectroscopic parameters required for these spectrometers to operate at the targeted precision and accuracy. A major aim of this activity was to measure CO_2 concentrations at the parts per million (ppm) level with high accuracy. All CO_2 remote sensing activities, from both the ground and space, rely on monitoring CO_2 vibration-rotation spectra. They are, therefore, heavily dependent on laboratory spectroscopy for reliable parameters that are essential for interpreting the atmospheric spectroscopic data. These parameters are of three types: line center frequencies, line profiles, and line intensities, all of which require high-precision measurements for the most demanding applications. Spectroscopic parameters of high accuracy have been determined for carbon monoxide (CO),[3] nitrous oxide (N_2O), [69] methane (CH_4), and ethane (C_2H_6), [70] as well as for carbon dioxide (CO_2). [71] [72] [73]

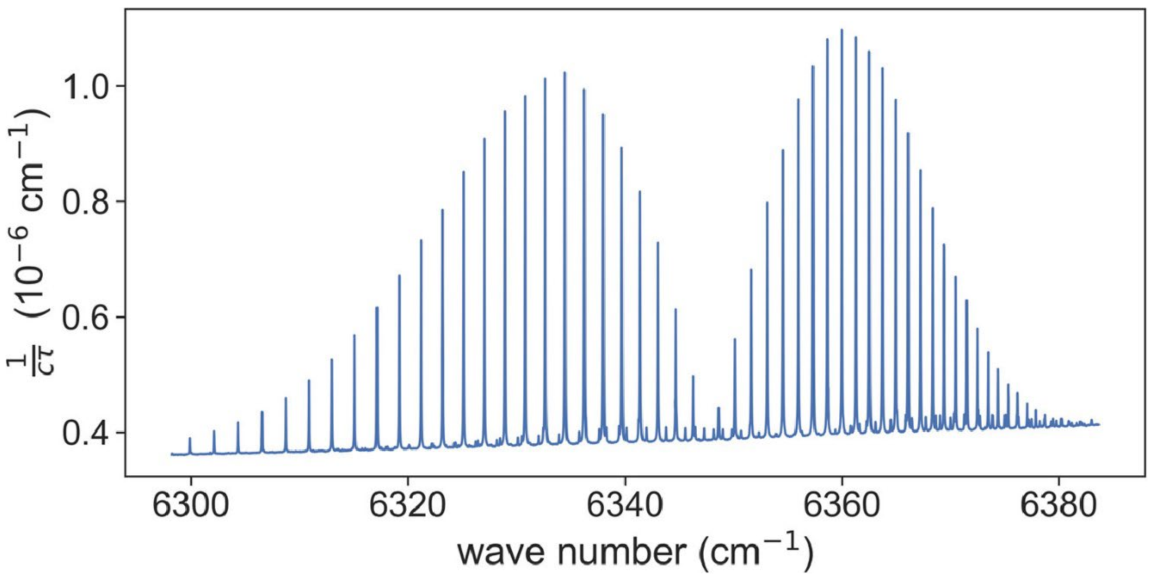


Figure 7 – Broadband CO_2-in-air FARS-CRDS spectrum of the (30012) ← (00001) band
doi.org/10.1016/j.jqsrt.2021.107669

NIST is also at the forefront of absolute CRDS measurements of molecular transition intensities needed for spectroscopic retrievals of amount of substance, and transition frequencies proportional to differences between quantized molecular energy levels. The experimental approach leverages the property that CRDS absorption spectra depend on quantifying the decay rate of the light intensity that probes the sample. Consequently, the results are pinned to highly precise and accurate measurements of frequency, which is the physical dimension that can be measured with the lowest uncertainty and highest precision. For the most accurate intensity determinations, sample purity, and analyte mole fractions need to be carefully assessed, necessitating well-characterized reference gas samples and

[3] Carbon monoxide concentration is a proxy for fossil fuel combustion due mainly to vehicle traffic and is used to constrain $FFCO_2$ estimates for that sector.

accurate measurements of sample temperature and pressure. To meet these requirements, NIST expertise on development of Standard Reference Materials (SRMs) for gas mixtures is utilized; these provide certified analyte mole fractions traceable to the SI.

In parallel, the experimentally measured intensities are compared to those based on *ab initio* quantum-chemical calculations of the corresponding molecular transition moments. These calculations are done by international collaborators using the most advanced computational methods. NIST researchers and their colleagues at the University College London, National Academy of Sciences in Russia, and Nicolaus Copernicus University in Poland have focused on the issue of determining accurate line intensities from experiment and theory. Twenty years ago, it was not possible to predict or measure most absolute intensities of atmospherically relevant molecules to better than several percent. But in the last fifteen years NIST's CRDS measurements have led to improvements in both experimental and theoretical capabilities, for assigning intensities, helping to bound uncertainties, and to identify where theory and experiment require additional effort. For many cases,[74] [75] [76] it has been shown that uncertainties in the theoretical and experimental intensities are of comparable magnitude and now differ by as little as 0.1%.[77] [78] In the case of molecular transition frequencies, NIST CRDS-based measurements substantially exceed the accuracies of traditional spectrometers, and are many orders of magnitude more accurate than state-of-the-art calculations. Furthermore, NIST transition frequency measurements serve as secondary frequency references, with direct traceability to the SI unit for frequency. [79] [80]

2d. Reference Data for Spectroscopic Measurements

Climate scientists are developing a new generation of spectrometers for the NASA-sponsored inventory of atmospheric greenhouse gases, most notably carbon dioxide and methane via their molecular absorptive properties. This inventory is critical to improving our understanding of the variations in the composition of our atmosphere, primarily due to anthropogenic sources of GHGs. To ensure the integrity and long-term continuity of these measurements, spectrometer operation relies on reference data of the highest metrological quality and traceability to the International System of Units (SI).

The study of molecular absorption and emission of radiation is of great importance for many scientific applications. Much of our knowledge of the geometrical and electronic structure of various molecules comes from optical absorption studies. Over the years, NIST has contributed much to this knowledge base on electronic and vibrational structure of molecules.[81] This information provides the necessary optical tools to design experimental probes of remote populations and processes such as remote sensing of the makeup of our atmosphere and the chemical processes occurring in hostile environments.

NIST is providing the spectroscopic parameters required for these spectrometers to operate at the targeted precision and accuracy. NIST is internationally known for its major contributions to producing some of the most accurate and precise spectroscopic data to date, including transition frequencies, intensities, cross-sections, and line-shape parameters. These high-quality data are suitable for stringent benchmarking of quantum-theory-based calculations used to populate spectroscopic databases. This strategy enables extension of the theory to a much broader range of conditions, wavelengths, etc. than can be achieved through measurements alone. This work involves significant interaction with academic groups, other NMIs, commercial entities, and government agencies throughout the world. Key collaborations and stakeholders include the NASA OCO-2/3 Science Team, the HITRAN Committee at the Harvard-Smithsonian Center for Astrophysics (CFA), University College London, Sorbonne (CNRS), Nicolas Copernicus University (Poland) and other committees including IUPAC, APS, Optica, and the Consultative Committee for Amount of Substance (CCQM) of BIPM.

Oxygen plays a key role in remote sensing measurements because its concentration in the atmosphere is accurately known and does not change with time. The O_2 A-band absorption spectrum in the wavelength vicinity of 760 nm is particularly important to many remote-sensing applications because it is largely free of spectral interferences. An important example includes measurements of CO_2 concentrations by orbiting spacecraft, [e.g., NASA's Orbiting Carbon Observatories, (OCO)] in which absorption path lengths and CO_2 mixing ratios are to be determined with an uncertainty of 0.3 %, using the O_2 A-band spectrum as a reference. These rigorous and quantitative measurements demand that the O_2 A-band line parameters (intensity, transition frequency, pressure-induced frequency shifts and broadening, lines shape and mixing, etc.) be measured with unprecedented precision and with accuracy established by traceability to the SI.

The Standard Reference Data® provided by this research enables new, high-precision satellite-based measurements of greenhouse gases in the Earth's atmosphere. Specifically, these reference data are required by the OCO-2 satellite mission, which maps column averaged atmospheric CO_2 over the Earth's surface, and by the Total Carbon Column Observatory Network (TCCON), an international network of ground-based high-resolution spectrometers that measure atmospheric trace gas concentrations across the infrared. These measurements are also critical to the following generation of satellite-borne instruments, which will rely on laser technology to measure diurnal and seasonal variations in the atmospheric greenhouse. Similarly, accurate measurements of the O_2 A-band will support meteorological observations and weather modeling and have the potential to determine global surface pressure in remote locations where current measurements are inadequate or non-existent.

In related studies, NIST researchers also carried out stringent experimental tests of advanced spectral models that go beyond the standard Voight profile.[82] As a result of these and other investigations, spectroscopic databases such as HITRAN can now incorporate more general line profiles required for high-accuracy spectral models that work over a wide range of temperature, pressure and composition. [83, 84]

2e. Measurement of Carbon and Oxygen Isotopes

Measurement of carbon isotopes has been used in “carbon dating” of ancient remains of plants, humans, and other organic materials. Carbon-14 (^{14}C), a radioactive isotope of carbon-12 (^{12}C), is formed in the upper layers of the atmosphere by cosmic rays and finds its way into all living things. ^{14}C is unstable, with a half-life of 5,730 years. When living things die, they stop incorporating carbon into their bodies, and their ^{14}C starts to decay. The remaining amount of carbon-14 in a dead body is measured to “date” the remains. But carbon-14 is extremely rare, and to use it for carbon dating, scientists need to be able to measure it at concentrations as low as 1 part in 10 trillion. These measurements are usually performed by accelerator mass spectrometry which is a costly process. Most of these methods also require standard samples to calibrate the instruments.

Carbon-14 measurements are also carried out with atmospheric samples to distinguish carbon dioxide emissions resulting from combustion of fossil fuels and those from biogenic sources. Fossil fuels are remains of plants and living species that died million years ago; their ^{14}C content has decayed and therefore they no longer contain any ^{14}C. Hence, the ratio of ^{14}C vs ^{12}C content of an atmospheric sample of carbon dioxide gives an indication of its source.[85] NIST researchers have developed a bench-scale instrument for absolute optical measurements of rare carbon and oxygen isotopes found in carbon dioxide.[86, 87] As shown in Figure 8, this approach leverages the linear absorption, species selectivity and high sensitivity properties of the CRDS technique resulting in an absolute ratio approach that does not require consumable and often unstable reference materials. To achieve the required sensitivity, NIST researchers enhanced existing CRDS technology by engineering a system that chills the cavity to a uniform - 55 °C and minimizes temperature fluctuations that allows their instrument to detect very faint signals of light absorption. This and other improvements boosted the instrument's sensitivity enough for accurate ^{14}C measurements.

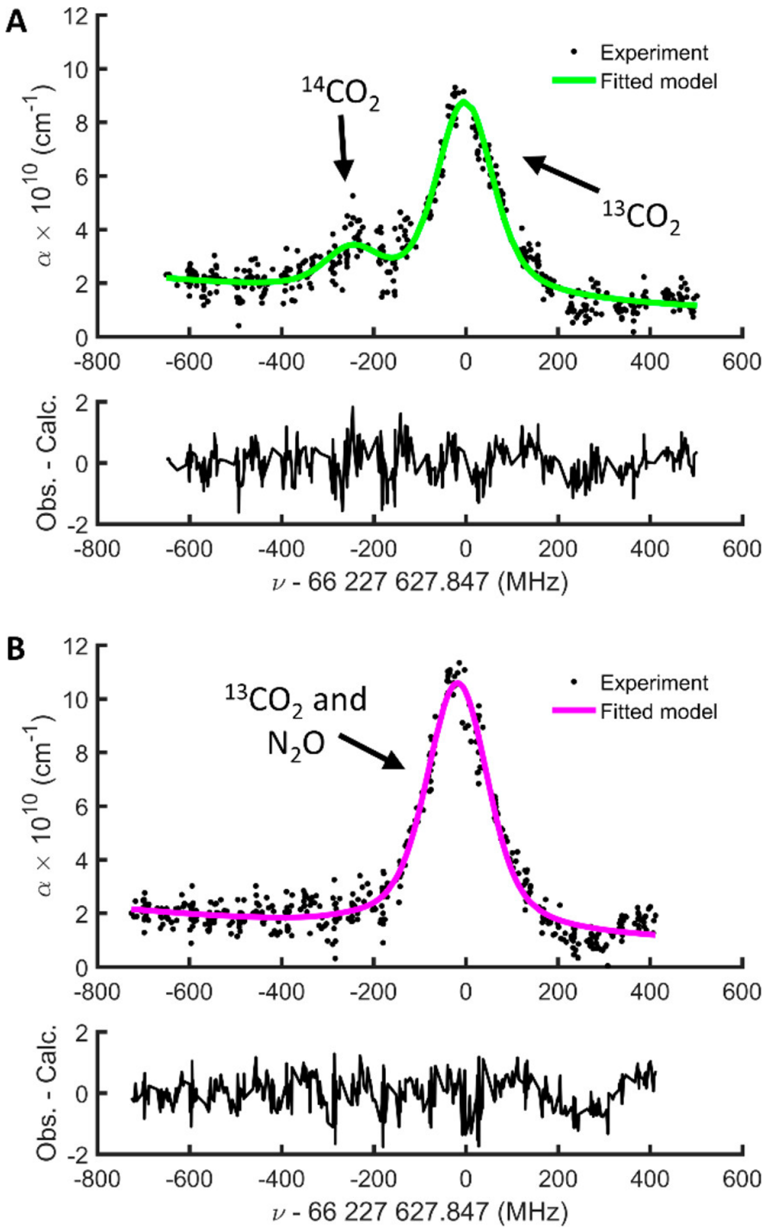


Figure 8 – Interleaved spectra of biogenic (A) and petrogenic (B) CO_2 recorded at T = 220 K and P = 0.9 kPa doi.org/10.1021/acs.jpclett.7b02105

Resulting studies included measurements of the radioactive isotope of carbon ^{14}C, and the remaining stable isotopes of carbon and oxygen, ^{13}C, ^{17}O, and ^{18}O in CO_2. Because fossil fuels are essentially free of ^{14}C, the quantification of radiocarbon dioxide in geophysical samples has numerous applications including dating, distinguishing between petrogenic and biogenic sources of carbon in fuel feedstocks, informing studies of carbon transport and the global carbon cycle, as well as enabling critical measurement needs for carbon trading. Key accomplishments of this project include a patented method providing the first linear absorption spectroscopy measurements of radiocarbon dioxide below natural abundance levels,[88] and the first accurate artifact-free measurements of $^{13}C/^{12}C$ isotope ratio in carbon dioxide.[87, 89] [90]

To estimate fossil fuel emissions in a geographic area, collected air samples across that area can be analyzed. Areas with high fossil fuel emissions, such as cities and industrial zones, will have below-normal concentrations of $^{14}CO_2$. Since fossil fuel emissions dilute the concentration of $^{14}CO_2$ in the air, accurate measurement of $^{14}CO_2$ concentration after it's been diluted will enable calculation of how much fossil fuel emissions are in those samples.

A report from the National Academy of Sciences [91] estimated that 10,000 samples a year, collected at carefully chosen locations around the United States, would be enough to estimate national fossil fuel emissions to within 10 % of the actual value. Such a system of measurements can increase the reliability of national emissions estimates. This would be especially useful in parts of the world where high-quality emissions data are not readily available.

This technology could also find applications in other industries such as biofuels and bioplastics. Analysis of the ^{14}C to ^{12}C ratio in a fuel mixture can determine what fraction of it is biofuel. In the airline industry, for example, this would be useful because some countries require aviation fuels to include a specific biofuel percentage. Such tests could also be used to verify that bioplastics, which sell for a premium, do not contain petroleum-derived compounds.

2f. Line-of-Sight Measurements with Dual-Comb Spectroscopy

Fully stabilized frequency combs provide a broadband spectral output that is comprised of a series of narrow spectral lines.[92] "Dual comb spectroscopy" (DCS) provides one method to harness these properties for broadband spectroscopy and remote sensing applications. It allows the user to read-out the spectrum of a gas mixture on a comb tooth-by-tooth basis. DCS combines many of the strengths of conventional broadband spectroscopy and tunable laser spectroscopy into a single platform. NIST conducted the first demonstration of truly coherent DCS by use of two phase-locked fiber frequency combs in 2008.[93] Since then, research in DCS has rapidly expanded.[94] DCS has several advantages over conventional spectrometers in the basic performance metrics of frequency resolution, accuracy, acquisition speed, and signal-to-noise ratio as well as in the potential for a compact, field-deployable system. In recent years, NIST research has focused on applying DCS to open-path detection of atmospheric trace gases, exploiting DCS for both multi-species gas detection and the detection of broadband absorbers. NIST has successfully developed several generations of broadband near-infrared dual-comb spectrometer (1.4 µm to 2 µm) for kilometer-scale open path atmospheric measurements and deployed them in proof-of-principle experiments. The dual-comb spectrometer can run continuously for weeks over open-air paths and accurately retrieve multiple atmospheric gases at sub-minute time resolution. Furthermore, it can operate over open paths to a retroreflector mounted on a flying quadcopter to enable spatial scanning or in a hub-and-spoke configuration for large-area monitoring. [95] In a parallel effort, the team is planning to extend this capability into the mid-infrared spectral region (3 µm to 5 µm) where the larger organic molecules have much stronger cross sections.[96] This work could lead to trace gas detection of broadband absorbers of interest to both the atmospheric science community and the defense community.

Given the number of potential applications of the DCS technology, it is not surprising to see the emergence of commercial applications.[97] In a recent demonstration, NIST's pair of laser frequency combs measured the simultaneous signatures of several greenhouse gases—including carbon dioxide, methane, and water vapor - along a 2-kilometer path between a NIST laboratory roof in Boulder, CO, and a nearby mesa. Researchers identified gases in the atmosphere by measuring the amount of comb light absorbed at different frequencies during its trip from the NIST lab roof to a mirror on the mesa and back to a detector in a lab. Because the optical frequencies are too high to be measured directly, the researchers borrowed a trick from early radio. They created two combs with slightly different spacing between the teeth. Mixing light from these dual frequency combs together creates a "beat" frequency shifted down to the radio band, low enough to be measured. This was the first demonstration of the technique over long distances outdoors.[98]

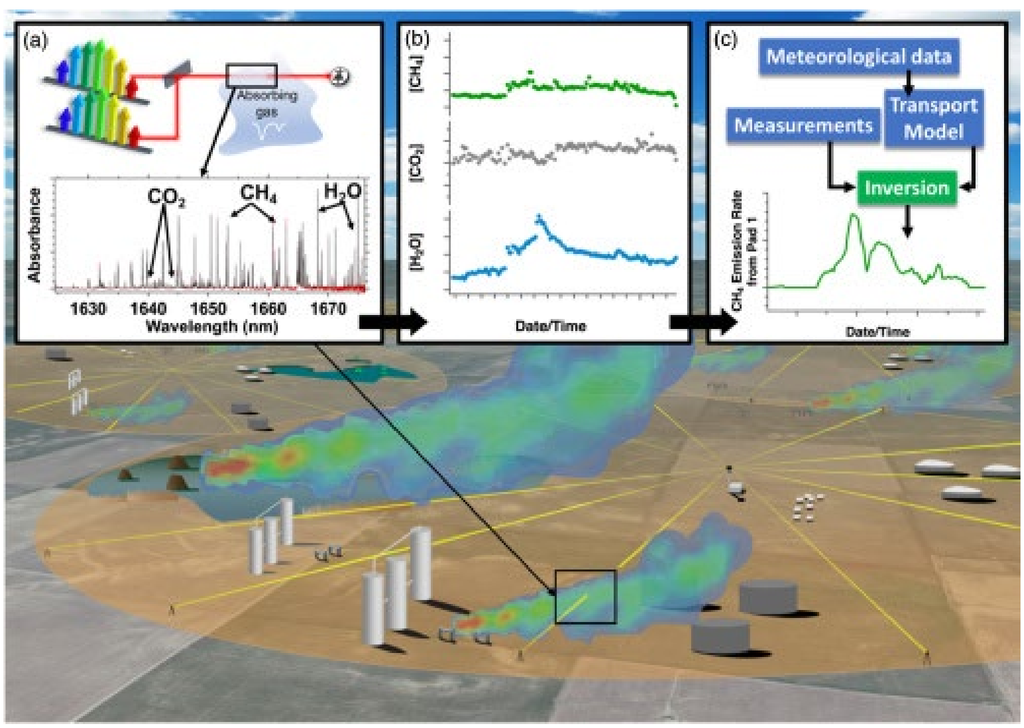


Figure 9 - Regional source monitoring with a centralized DCS. (a) The DCS measures trace gas absorption over an array of long-distance beam paths. (b) Time-resolved trace gas concentrations are determined from fits to the absorption spectra with ppb-km sensitivity and stability. (c) An atmospheric transport model and inversion determines source location and time-resolved emission rate. doi.org/10.1364/optica.5.000320

In the demonstration, the research team collected data continuously for three days under varied weather conditions. The results were comparable to data collected by a nearby point sensor under well-mixed atmospheric conditions. The comb measurements were also very precise—with uncertainty of less than 1 µmole/mole for carbon dioxide concentration, for example, obtained in five minutes. That is precise enough to ensure detection of small increases in trace gases due to large, distributed sources such as cities. Future systems should be able to achieve even better sensitivities over shorter timescales.

Accuracy in the measured atmospheric transmission is assured by the well-defined position of each frequency comb tooth. Because the technique makes repeated measurements rapidly over the same path, it is immune to signal distortions caused by atmospheric turbulence. And because the comb measurements can be averaged over the entire path length rather than relying on a few spot measurements, the comb method is better matched to the scale of atmospheric transport models.

Overall, the study results suggest that the dual comb technique is ideally suited to precise, reproducible sensing of trace gases in the atmosphere and can support the development of accurate models for use in global, satellite-based greenhouse gas monitoring. NIST researchers now plan to optimize the comb system by boosting power to improve sensitivity and expanding spectral coverage to identify additional gases. Portable frequency comb systems could eventually support regional gas monitoring at costs comparable to point sensors, but over the kilometer scales relevant to many transport models and to monitoring of distributed sources such as large cities. [89] [99] [100] In the last several years DCS technology has migrated to the commercial sector and is in use quantifying methane emissions in the oil and gas applications.

Multiple long-path measurements over an area of interest can also be applied, using tomographic reconstruction, to interrogate 2-D distribution of GHG concentrations. Such a system was developed commercially, called Greenhouse gas Laser Imaging Tomography Experiment (GreenLITE), which has the potential for real time monitoring of underground storage sites/sequestration sites, [101] as well as urban areas.[102] This system combines a set of laser transceivers based on an intensity modulated continuous wave approach with an array of retro-reflectors for 2-D sparse tomographic reconstruction mechanisms to compute a 2-D mapping of CO2 concentrations over the area of interest. This system was demonstrated in Paris for approximately a year in 2015 beginning around the time of the Paris COP.

2g. Development of Differential Absorption LIDAR (DIAL)

NIST is developing a spatially resolved, remote-sensing method for accurately quantifying greenhouse gas emissions from natural and anthropogenic sources and sinks to meet the requirements for local, national, and international mitigation efforts. This technology, known as differential absorption LIDAR (Light Detection and Ranging) or DIAL can provide species-specific concentrations of greenhouse gases with spatial information along the laser path. These spatially resolved measurements, coupled with wind velocity measurements, determine the flow of carbon dioxide and methane moving through the atmosphere. Used vertically, DIAL measurements can provide spatially resolved concentration measurements to near the top of the atmosphere.

The basic processes involved in elastic backscatter LIDAR are as follows. A laser source emits a pulse of light (typically a few nanoseconds), and as the pulse propagates, the photons interact with particles in the atmosphere. Some of these interactions, such as Mie and Rayleigh scattering, result in backscattered photons. The photons are collected by the detector and recorded as function of time. This time-of-flight data has a direct correspondence with the range (distance) at which the scattering event occurred.

Differential absorption LIDAR (DIAL) is based on the same principal, but operates at two wavelengths, one on-resonance and one off-resonance of the molecular absorption of the greenhouse gas of interest. Because the on-resonance wavelength is more strongly absorbed by the greenhouse gas, the difference between both signals is proportional to the number density of the GHG. Thus, this technology could provide regulators with quantity of greenhouse gases being released at a particular location and pinpoint their sources.

The DIAL system is being designed to span the near infrared spectral region where the greenhouse gases, carbon dioxide, methane, and nitrous oxide, absorb. DIAL is typically performed to measure the on-resonance backscatter signal for some time and then tune the laser to measure the off-resonance signal. However, the atmosphere may have changed between measurements. The new system is designed to perform on-resonance and off-resonance measurements simultaneously to provide precise measurements.

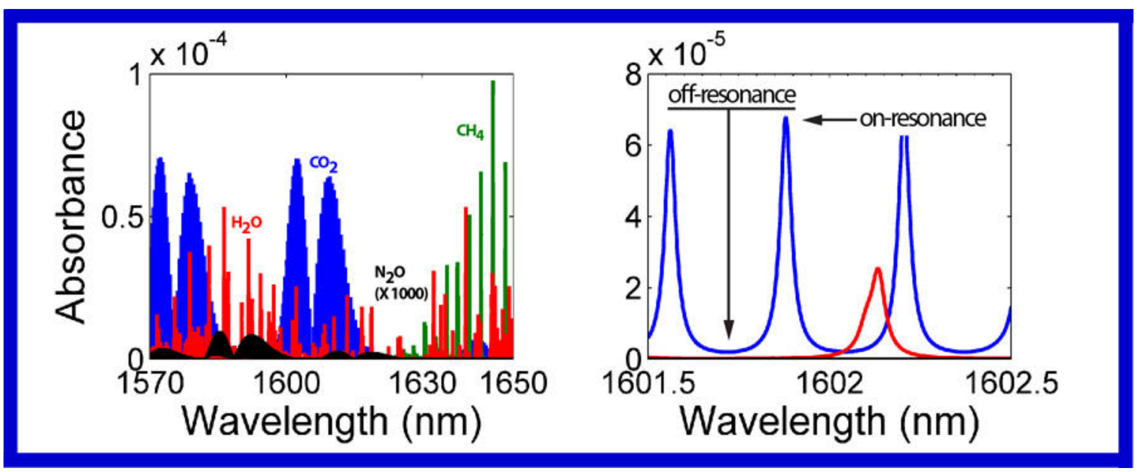


Figure 10. Absorption of photons at different wavelengths from 1.57 to 16.5 micrometers for water and each of the principal greenhouse gases. Right: Difference in absorption by carbon dioxide of photons on- and off-resonance.
www.nist.gov/news-events/news/2012/02/progress-and-promise-dial-lidar

All three of the principal GHGs have clearly defined absorption peaks at near-infrared wavelengths between 1.65 µm and 1.57 µm. That range turns out to be fortunate for atmospheric monitoring; water vapor is only weakly absorbing in that wavelength range. The 1.55 µm region is also one of the staple wavelengths used by the telecommunications industry because it travels with minimum dispersion in fiber-optic cable. As a result, many technologies have been developed for applications in this part of the spectrum, such as inexpensive near-infrared lasers that go from about 1.2 µm to almost 2 µm .

The NIST team is also taking advantage of another cutting-edge component to boost the output. The initial DIAL beam was generated by a diode laser, which alone did not have the power required to probe a distant volume of gas. In the new system, the laser beam is put into a device called an optical parametric oscillator, which increases the number of photons by approximately 10^9.

When these measurements are coupled to precise wind velocity measurements, the flux of the greenhouse gases, the important quantity for regulators, can be determined. An indoor testing facility is also being developed to rigorously test hardware components (optical receiver, laser control, etc.) and software (analysis algorithms, data throughput). [103] [104]

2h. Greenhouse Gas Concentration Measurement Standards

Measurements of atmospheric gases have been utilized in a wide range of industrial applications and implementation of environmental regulations such as the U.S. Clean Air Act,[105] National Ambient Air Quality Standards,[106] the Montreal Protocol on Substances that Deplete the Ozone Layer,[107] and many others. Protocols have been established for development of reference materials ("gold standards") by NMIs such as the Standard Reference Materials® (SRMs®) developed by NIST, [Ref] and similar certified reference materials (CRMs) issued by the NMIs of some other countries. These are traceable to the SI as a means of ensuring measurement accuracy. Quality management systems are established according to ISO 17025 and ISO 17034 to ensure consistency of these standards. Comparability of methods used to characterize reference materials among different countries is ensured through Key Comparisons, usually performed under the auspices of the International Bureau of Weights and Measures (BIPM) and its Consultative Committee on Chemistry and Biology (CCQM), Gas Analysis Working Group (GAWG).[108]

During measurements performed in the field, on towers, mobile units or on aircraft, measuring instruments are calibrated with secondary and tertiary standards that are traceable to the SI through national standards; they are provided by NMIs or by specialty gas companies that produce, for example, NIST Traceable Reference Materials® (NTRMs®),[16][16][16] [16] or other CRMs. Measurements are performed using analytical techniques such as Gas Chromatography/Flame Ionization/Thermal Conductivity Detectors (GC-FID/TCD), Fourier Transform InfraRed (FTIR) Spectroscopy, Non-Dispersive Infrared (NDIR) detectors, Non-Dispersive Ultra-Violet (NDUV) Spectroscopy, and more recently using Cavity Ring-Down Spectroscopy (CRDS). [109]

Historically, measurements performed as part of the WMO's Global Atmospheric Watch (GAW) network use standards provided by WMO Central Calibration Laboratories (CCLs). NOAA serves as the WMO-CCL for CO_2, CH_4, N_2O, CO, and several fluorinated gases. WMO uses a "scale" system, which allows the WMO-CCL to produce reference materials that have very small uncertainties in relationship to each other. This relationship is maintained in a database, and the value of any reference material can be traced back to the primary standard set defining the scale. A change in the "scale" can then be propagated to all WMO standards by the relationships contained in the database. Requirements for the CCLs and GAWG measurement laboratories are documented and periodically updated at the **WMO/IAEA Meetings on Carbon Dioxide, Other Greenhouse Gases and Related Measurement Techniques** (GGMT). The latest of these meetings was held at Jeju Island, South Korea in September 2019.[110] (Proceedings of GGMT 2022 Meeting in Netherlands are not yet available)

GGMT Reports state that "…WMO and IAEA strive for all monitoring systems to be formally traceable to Primary Reference Materials or Fundamental Constants (SI) through National Metrology Institutes (NMI) and the International Bureau of Weights and Measures (BIPM). This is an essential pre-requisite for an internationally recognized and homogeneous monitoring system of in situ chemical measurements and as such is a primary responsibility of the CCLs " In more recent times, WMO has required "CCLs that have signed a letter of agreement with WMO to act as designated institutes for WMO as a signatory to the CIPM (International Committee for Weights and Measures) Mutual Recognition Agreement should participate in Key Comparisons of the Consultative Committee for Amount of Substance – Metrology in Chemistry (CCQM)".[108] Recommended precision requirements to ensure WMO/GAW network compatibility are provided in several GGMT reports and are shown in Table 3.

Table 3. Recommended network compatibility of measurements within the scope of WMO/GAW

Component	Network compatibility goal[1]	Extended network compatibility goal[2]	Range in unpolluted troposphere (approx. range - 2019)	Range covered by the WMO scale
CO_2	0.1 ppm (NH) 0.05 ppm (SH)	0.2 ppm	380 - 450 ppm	250 – 520[3] ppm
CH_4	2 ppb	5 ppb	1750 – 2100 ppb	300 – 5900 ppb
CO	2 ppb	5 ppb	30 – 300 ppb	30 - 500 ppb
N_2O	0.1 ppb	0.3 ppb	325 – 335 ppb	260-370 ppb
SF_6	0.02 ppt	0.05 ppt	9 – 11 ppt	2.0 – 20 ppt

1 Scientifically desirable level of network compatibility for measurements of well-mixed background air. These represent the maximum bias that can generally be tolerated in measurements of well-mixed background air used in global models to infer regional fluxes. Some network compatibility goals may not be currently achievable within current measurement and/or scale transfer uncertainties. However, these network compatibility goals are targeted for application areas which require the smallest possible bias among different datasets or data providers, such as for the detection of small trends and gradients. Network compatibility goals are not direct metrics for instrument performance, however, instruments with significantly higher short-term imprecision make assessing network compatibility difficult.

2 Extended network compatibility goals are provided as a guideline for many other studies in which the smallest bias is not required, for example, a regionally focused study with large local fluxes, or services related to urban air quality.

3 The upper limit of the WMO CO_2 X2007 scale is 520 ppm. The upper limit is being increased to 800 ppm as part of the WMO CO_2 X2019 scale revision (expected in 2020) to meet the needs of atmospheric monitoring community engaged in urban measurements.

As a result of the *rapprochement* between WMO and BIPM, WMO-CCLs have recently participated in CCQM-GAWG organized key comparisons, along with a large number of NMIs. One comparison (CCQM-K120 Carbon dioxide at background and urban level) has been successful in demonstrating the degrees of equivalence of "CO_2 in whole air" standards amongst participating NMIs and CCLs at nominal mole fractions of 380 µmol/mol, 480 µmol/mol, and 800 µmol/mol, with the required measurement accuracies. [111] A subsequent publication has compared two systems: a) a stated reference system based on the International System of Units, and more specifically a realization of a primary standard with its value and uncertainty expressed in SI units, which offers many benefits such as long term stability and the ability to reproduce the standard within its stated uncertainty at any time; b) an alternative approach that uses measurements traceable to an empirical scale, an approach that has evolved for practical reasons to meet the needs of certain measurement communities. The article explored the benefits and drawbacks of these two measurement systems as they have evolved for greenhouse gas measurements at background levels and outlined the advantages of using elements from each to secure a more robust measurement infrastructure. In particular, this work examines the importance of compatibility, the accuracy of measurement results, and the benefits of comparisons with independent primary standards.[112] Another article on BIPM research explored the effect of isotopic composition of CO_2 on the measurements, especially when high accuracy measurements are required. [113]

The current WMO "scale" system has vulnerabilities. An expected increase in demand for standards due to the number of measurement sites for GHGs around the world expanding will require more sources for standards than currently exist. Developing multiple sources for standards would improve security and enable disaster recovery type plans to be readily implemented should there be a single point of failure in the scale maintained by the CCL.

An alternative system currently being evaluated within CCQM-GAWG is one that allows individual NMIs to maintain National and Regional GHG scales, using sets of equations relating these and their relationship to the WMO Scale while leaning on the CIPM MRA process and its key comparison program to establish and monitor consistency of these scales. The advantage of this system is that it could establish many sources for standards assigned on GHG scales, using facilities of NMIs that are already experts in gas handling and perhaps with linkage to the specialty gas supplier community. Such a system would also overcome the single point of failure issue, as the relationships between scales would be maintained with equations. As the scientific basis for such a system is just being investigated, demonstration of such an approach that includes contributions from WMO-CCL system and contributing NMIs is a necessary requirement for change.

Several NMIs have already expressed interest (NIST, NPL, NMIJ, KRISS, and the BIPM) to explore the measurement science fundamentals to set up such a system. Commercial gas suppliers have also expressed interest in participating in such an undertaking. The infrastructure for including commercial gas suppliers in the eventual production and dissemination of GHG standards currently exists within the NIST NTRM Program and the Environmental Protection Agency (EPA) Traceability Protocol for Assay and Certification of Gaseous Calibration Standards Program. [114] The EPA Protocol program was created to support the production of calibration gases for ambient and continuous emission monitors and to address concerns about commercial calibration gas accuracy and stability. The original protocol was prepared in 1978, and subsequently revised in 1997 and 2012, providing over 40 years of collaboration experience between NIST, the EPA, and the commercial gas suppliers.

Accurate measurements of GHG concentrations in the atmosphere is a field of technology that is quite mature and measurement standards and infrastructure are in place. Hence, the field is ready for expansion and development of international standards for measurement of CO_2, CO, CH_4, N_2O and HFCs in the atmosphere. International organizations such as NMIs, BIPM and WMO-CCLs are participating in activities towards achieving such an endeavor.

3. Advanced Computational Tools

Methods for quantifying GHG fluxes (from sources and sinks) are typically classified into two categories, often termed "bottom-up" and "top-down." **Bottom-up** methods utilize process-level understanding to estimate GHG fluxes while **top-down** methods use atmospheric measurements to estimate fluxes. **Bottom-up** anthropogenic emissions and biogenic flux models use process-level understanding to quantify emissions or uptake of GHGs; their development is independent of top-down methods. Anthropogenic emissions are derived from economic activity data and the associated emission factors. Initially developed by the academic community utilizing publicly available data, as a means of quantifying air pollutant levels locally and nationally with good spatial and temporal resolution, the method has also been applied for annual GHG emissions reporting to the UNFCCC. Gridded bottom-up estimates (i.e., flux maps) can also be used as initial guesses, or "priors", for top-down methods.

Flux estimation methods using atmospheric measurements utilize observed atmospheric GHG concentrations and their gradients coupled with atmospheric transport and statistical optimization models to estimate the emissions or uptake (i.e., flux) from a given region or an urban area. In any regional or urban top-down analysis, the aim is to isolate the "enhancements" in the GHG concentration caused by GHG fluxes originating in the region of interest. Thus, a "background" (or baseline) concentration must first be accounted for before determining the observed enhancement and the local GHG flux. [115] One top-down quantification method is **inverse modeling**;[116] other methods include **mass-balance** methods,[117] **tracer-tracer ratio** methods,[118] and more.

3a. Spatially Explicit Emissions Modeling (A Bottom-up Method)

Bottom-up approaches take form as Spatially Explicit Emissions and Biogenic Flux Models; their development is independent of top-down methods. The spatial sensitivity of these ranges from a few tens of meters to several kilometers in some applications. Spatially explicit models are maturing and are beginning to become commercially available.[119] These models have to account for anthropogenic sources, such as transportation, power generation, industry, commercial and residential buildings, landfills, etc. and they are typically used for traditional emissions reporting and accounting (nationally and locally). Their initial development has focused on fossil fuel emissions due to their significance as a large fraction of GHG emissions. As these GHG information sources become more prevalent, they are moving into the traditional emissions reporting and accounting (nationally and locally) regimes. Gridded bottom-up estimates (i.e., flux maps) can also be used as initial guesses, or priors, for top-down methods.

A separate research activity, the development of biogenic models, includes the effects of vegetation on atmospheric composition, both as a source and a sink of atmospheric GHGs. The carbon cycle processes of the biosphere are especially difficult to assess in urban environments where parks, woods, lawns, and other green areas are interspersed within residential and commercial areas. Properly accounting for photosynthesis uptake of CO_2 and respiration of CO_2 remains a complex task that will profit from continued research into these complicated fundamental processes. (See next section)

During the last decade, there have been major advances in development of emission models that vary in spatial and temporal scales, covering small areas such as cities and metropolitan areas relative to global models. The **Open-Data Inventory for Anthropogenic Carbon dioxide** (ODIAC) model is a high-spatial resolution global emission data product of CO_2 emissions from fossil fuel combustion.[120] In 2009, the first version of ODIAC was developed under the Greenhouse gases Observing SATellite (GOSAT) project at the National Institute for Environmental Studies (NIES) in Japan and then at NASA. ODIAC pioneered the combined use of space-based nighttime light data and individual power plant emission/location profiles to estimate the global spatial extent of fossil fuel CO_2 emissions. With the innovative emission modeling approach, ODIAC achieved a fine picture of global fossil fuel CO_2 ($FFCO_2$) emissions at a 1km x 1km resolution, possibly the most widely viewed global picture of man-made emissions. The **Emissions Database for Global Atmospheric Research** (EDGAR), developed by a European Union Joint Research Center, provides global past and present-day anthropogenic emissions of greenhouse gases and air pollutants by country and on a spatial grid.[121] The **Fossil Fuel Data Assimilation System** (FFDAS) was developed around 2010 as a multiyear, global gridded fossil fuel CO_2 emission data product.[122] Many of the global inventories of $FFCO_2$ emissions do not directly quantify emissions at local scales; instead, spatial proxies like population density, transportation statistics, nighttime lights, and power plant databases are used to downscale emissions from national or other totals publicly provided for specific geographic regions, e.g., census block or zip code areas. Suitability of these downscaled inventories for urban emissions monitoring suffer from lack of specificity in the input data, e.g., local activity patterns such as temporal variations of transportation activities.

More recently, high-resolution models have been developed in the research community to overcome some of these shortcomings and to provide tools that can be reliably used at city levels. One of these models is the **Anthropogenic Carbon Emissions System** (ACES) for $FFCO_2$, which provides high-resolution (hourly, 1 km^2) emissions data, based on local activity data for the year 2011 across the northeastern U.S. [123] The ACES model was compared with three widely used global inventories, finding significant differences at regional (20%) and city scales (50% – 250%). At a geospatial resolution of 0.1°, inventories were found to differ by over 100% for half of the grid cells in the domain, with the largest differences in urban areas and in oil- and gas-production regions. ACES has been updated to include data through 2017 and extended to the continental U.S.[124]

Another such model is **HESTIA** which quantifies fossil fuel CO_2 emissions on the scale of individual buildings, road segments, and industrial/electricity production facilities on an hourly basis for an entire urban landscape. With support from the NIST GHG Measurements Program, HESTIA has been developed for some large U.S. cities like Indianapolis, Salt Lake City, Los Angeles, Baltimore, etc. [125]

Inventories like **ACES** and **HESTIA** are crucial for mitigation efforts since much of the climate leadership has shifted to city and state governments. The development of a robust carbon monitoring system to track carbon fluxes is critical for emissions benchmarking and verification. These models are designed for easy updating, making them suitable for emissions monitoring under most city, regional, and state greenhouse gas mitigation initiatives. For the small- and medium-sized cities that lack the resources to regularly perform their own bottom-up emissions inventories, the sophistication of current emission modelling methods remains challenging.

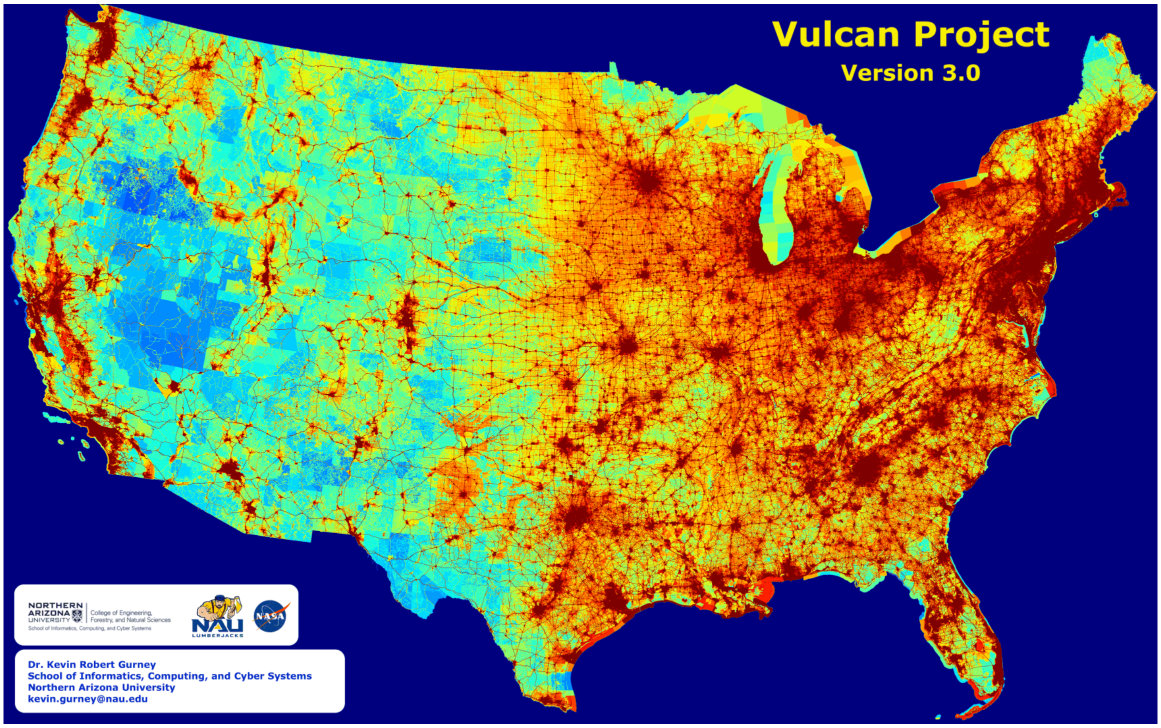


Figure 11. Typical output of the Vulcan Project, Version 3.0 https://vulcan.rc.nau.edu/

It should be kept in mind that these models rely on complicated data mining from a wide range of sources and diverse sectors of transportation, industry, power generation, commercial and residential buildings and landfills, types of fossil fuels utilized, leaks from natural gas distribution lines, and agricultural and other biogenic areas within a city's boundaries. To make them generally useful and reliable, and therefore usable by the private sector, metrics are needed. An international documentary standard, specifying performance metrics in terms of spatial and temporal resolution, and the type and quality of the data to be collected from each sector of potential emitters, would be a significant step towards their consistency and broad utilization.

Results of the **HESTIA** Project rely on the output of the **Vulcan Project**, initiated earlier, which quantifies all fossil fuel CO_2 emissions for the entire United States at high space- and time-resolution with details on economic sector, fuel, and combustion process.[126] The Vulcan Project is a multiagency (NASA, DOE, NOAA) funded effort spawned under the **North American Carbon Program** (NACP) to quantify North American fossil fuel carbon dioxide (FFCO2) emissions at space and time scales much finer than have been achieved in the past. The aim is to aid in quantification of the North American carbon budget, and to support inverse estimation of carbon sources and sinks. Vulcan provides data on each state in the U.S.;

recent versions have been refined to provide data on each county, and most recently on a 1 km x 1 km grid, and is being offered as a commercial product.[127]

As the threat of global warming grows—and with it, the specter of more extreme conditions such as wildfires, droughts, and tropical storms—cities across the U.S. are developing policies to reduce their emissions of greenhouse gases, mainly carbon dioxide (CO_2). Additionally, methane emissions from oil and gas production facilities, landfills, agriculture, and wetlands have gained significant national attention and focus as a Short-Lived Climate Forcer. (Methane's atmospheric half-life is estimated to be ≈ 9.1 years; its Global Warming Potential (GWP), based on a 20-year lifetime, is estimated to be ≈ 82 compared to CO_2)[128] Many local governments are committed to mitigation goals; however, the emissions data they must work with is often too general to provide a useful baseline, target the most effective policies, or track progress toward mitigation objectives.

Researchers at Northern Arizona University, who specialize in atmospheric science, ecology and public policy, have spent the past several years developing a standardized system, as part of the **HESTIA** Project. HESTIA quantifies and visualizes greenhouse gases emitted in urban regions down to each street and building, identifying problem areas and showing where public policy is—or is not—making a difference. Their goal is to provide cities with a means to strategically address problem areas instead of taking a sweeping and costly approach.

One of the more recent accomplishments of this team is mapping of the greenhouse gas emissions in the Los Angeles Megacity—the third largest metropolitan area in the world—occupying 12,560 km^2 (4,850 square miles) and housing more than 18.5 million people. In collaboration with scientists at Arizona State University, the University of Michigan, NIST and NASA's Jet Propulsion Laboratory, they developed the high-resolution, bottom-up emissions map that includes every building and road in the Megacity - for a staggering emissions total of 176 million tons of CO_2 per year.[129]

Mapping of emissions in California's South Coast Air Basin, including Los Angeles proper, was one of the latest efforts in a long-term project that involves collaborators at NASA, NOAA, NIST, the University of Colorado, Pennsylvania State University, SIO at the University of California, San Diego, the University of Utah, GNS Science in New Zealand and Purdue University. Other cities mapped to this degree are Indianapolis, Salt Lake City and Baltimore. However, the Los Angeles mapping is expected to have the most significant implications, especially considering the sheer size and scope of the area.

Although Indianapolis was the first city quantified at the building and roadway level, Los Angeles is the first megacity for which emissions have been quantified down to that scale. Nothing has been left out of an area that includes 80 cities and five counties. Given the fact that California has an aggressive policy when it comes to GHG emissions, [130] such data provides the tools to know what the best policy options are and whether those policies are working. This example illustrates the capability of providing such information to local governments, where immediate actions can be pursues. One of the strengths of the approach is the incorporation of atmospheric monitoring of CO_2 from ground-based and satellite instruments, as these become more widely available in the future. These increase credibility of the information. Most recently, the **HESTIA** Project has been extended to map GHG emissions in the U.S. Northeast Corridor, starting in the Washington, DC/Baltimore, MD area.[131] [132]

3b. Improved Biogenic Emission/Uptake Models

The signal of biological CO_2 flux can be many times larger than the signal of anthropogenic CO_2 flux from fossil fuel emissions during the growing season, especially in rural areas upwind of urban centers, but also in suburban and urban fringe locations. This complicates the ability to monitor urban fossil fuel emissions using the atmospheric data alone, which both globally and in urban areas reflects the integrated signal of both biological and anthropogenic CO_2 fluxes in near-field and upwind locations. During the afternoon hours when CO_2 measurements are typically used in inversions, the biological drawdown and fossil fuel emission signals can largely offset one another in suburban areas. Also, the specification of urban-region background conditions is complicated by upwind variability in biogenic CO_2 fluxes, particularly in the U.S. Northeast Corridor which is downwind of deciduous forests and Midwestern cropping areas.

To help address these concerns, NIST collaborates with the larger carbon cycle scientific research community in evaluating and improving biogenic models of CO_2 flux that can simulate diurnal variability, spatial patterns across diverse landscapes and the timing of the seasonal drawdown and release.[133] Biogenic models span the range from fully diagnostic models that rely on observed remote-sensing inputs to simulate fluxes with only simple representations of underlying processes, to more complex prognostic physically-based approaches that internally simulate quantities like the start of the growing season and photosynthetic parameters like Leaf Area Index and solar induced fluorescence, and track exchanges between a number of living and dead carbon pools. NIST and others in the research community are currently using the Vegetation Photosynthesis & Respiration Model (VPRM),[134] [135] a fully diagnostic model, to optimize parameters using GHG flux towers situated in the eastern US and Canada upwind of the Washington, DC/Baltimore region. NIST researchers are also collaborating with partners at other institutions who have been running an ensemble of CASA (Carnegie-Ames-Stanford Approach)[136] [137] model runs, another diagnostic model with monthly phenology and a more complex process-based approach for capturing plant and soil respiration fluxes compared to VPRM. Most recent results show that the new respiration model in VPRM helps to correct a growing season sink bias in the atmosphere associated with underestimated summertime respiration using the original model with annual parameters. The new VPRM also better explains fine-scale atmospheric CO_2 variability compared to other terrestrial biosphere models for non-urban fluxes.

To span the full range of model types, CO_2 fluxes from VPRM and CASA are also being compared with fluxes from SiB4 (the Simple Biosphere Model v4),[138] [139] a complex process-based model with fully prognostic phenology that simulates both the energy and carbon cycles. Fluxes from all three models have been compared in a regional domain spanning most of the eastern US and southeastern Canada, a domain which is important to characterize for accurate simulation of background conditions in the Northeastern Corridor, especially during the growing season. Biological CO_2 fluxes are also convolved with atmospheric footprints from atmospheric transport models and backward time Lagrangian models, e.g., NOAA's HYSPLIT model [140] and compared to observed CO_2 enhancements at a network of GHG observing towers measuring continuous atmospheric CO_2 mole fractions across the larger regional domain. In a recent NIST study, modeled biogenic CO_2 fluxes from VPRM [134] as well as other biogenic flux models like CASA (Carnegie-Ames-Stanford Approach) [136] and SiB4 (Simple Biosphere Model v4) [138] were inter-compared with atmospheric observational data and each other.[141] This study demonstrated that the respiration component of VPRM needed to be modified to better represent heterotrophic respiration in the US Northeast. These analyses are helping to identify relative strengths and weaknesses of each model and pinpoint specific deficiencies that will affect the simulation of biological CO_2 flux in urban domains and less populated and agricultural upwind areas, pointing the way towards needed model improvements. This research is expected to lead to best practices and the choice of models best suited for specific cities and regional areas.

A Standard for Remote Sensing of Vegetation

The NIST Headquarters in Gaithersburg, MD also provides a convenient testbed to evaluate the current state of knowledge relating optical remote sensing to biophysiological measurements of vegetative land cover. The mixed deciduous forest at NIST, with some tree specimens well over 100 years old, has a network of sensors deployed over a 1-hectare area within the larger 40-hectare forest. These sensors measure respiration, sap flow in trees, and meteorological parameters to provide ground truth to model simulations and optical observations. Optical remote sensing observations of vegetation can help constrain ecosystem models used to estimate carbon fluxes. Optical observations provide the advantage of sampling up to the global scale. However, linking a satellite scale footprint to the patch and leaf-level observations is an ongoing area of research.

The goals of the research are to:

- Provide a testbed (Forested Optical Reference for Evaluating Sensor Technology, FOREST) for eco-physiological and optical sensors with well-established reference points;
- Investigate ecosystem phenology that will allow for better estimates of carbon flux;
- Improve estimates of carbon flux of fragmented urban landscapes;
- Link leaf level to satellite footprint scale observations; and
- Facilitate traceability to NIST standards.

In a study conducted at the FOREST site located on the NIST Gaithersburg campus, a comprehensive CO_2 inventory of an urban temperate forest and unmanaged grassland was developed using a range of field instruments coupled with measurements of several ecological parameters. Figure 12 illustrates the FOREST research effort and the variety of observations involved. The green diagonal lines on the lefthand inset indicate surface tracks of NASA's OCO-2 satellite equipped with and instrument observing Surface Induced Fluorescence (SIF) signals. The annual storage of CO_2 by vegetation and soils was estimated and biogenic flux estimates were put in context of local fossil fuel CO_2 (FFCO2) emissions to determine when, where, and by how much biogenic fluxes alter net CO_2 flux dynamics. Hourly estimates of biogenic fluxes were compared to modeled estimates using a modified version VPRM in the Washington, DC/Baltimore area. During summer afternoons, the net flux was found to be dominated by the uptake of CO_2 by vegetation.[142] This work represents the most detailed carbon inventory of the NIST FOREST site and a link to satellite observations.

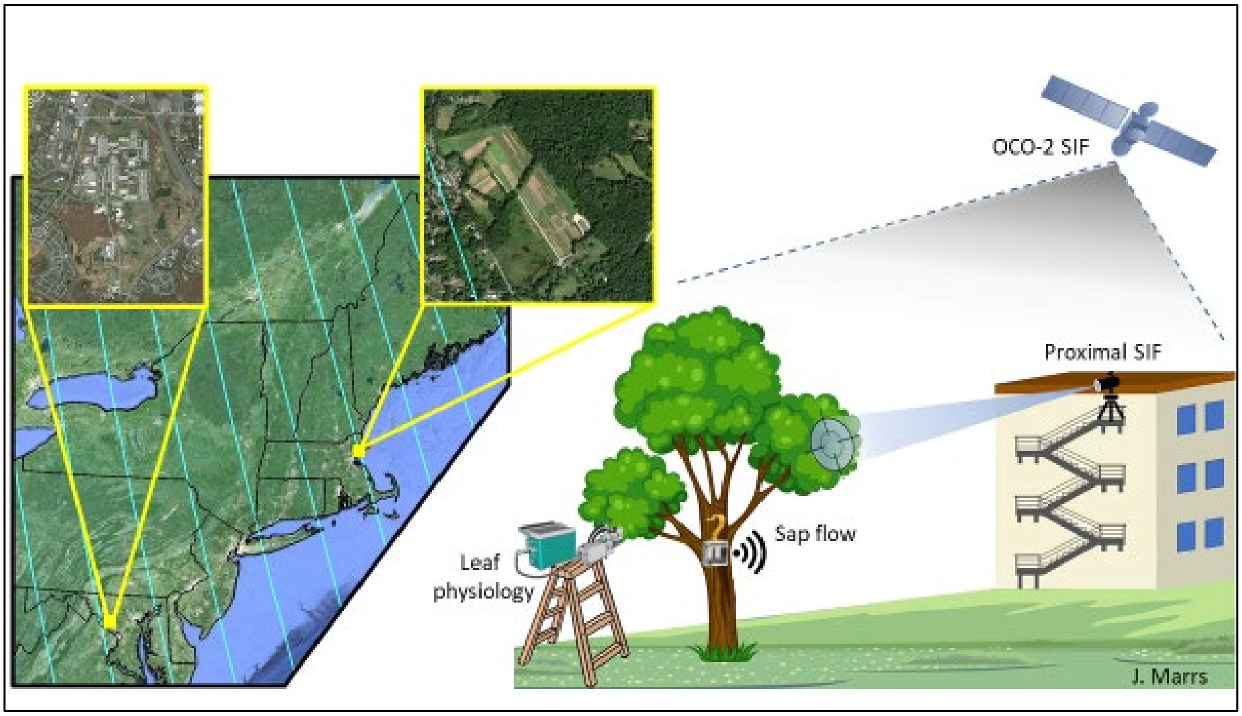


Figure 12. Optical remote sensing measurements are being performed in the NIST forest in Gaithersburg, MD and in the Harvard Forest in Petersham, MA. (Figure courtesy of Julia Marrs)

Solar-induced chlorophyll fluorescence (SIF) has been widely cited as a proxy for photosynthesis and is being incorporated as a common input in terrestrial primary productivity models. Though satellite-based SIF products show close relationships with terrestrial gross primary productivity (GPP), there is wide variability in the magnitude of published SIF retrievals made at intermediate scales. In a meta-analysis of the tower-based and airborne SIF literature, it was found that mean SIF retrievals from unstressed vegetation were quite wide. In order to quantify the importance of such methodological differences on final SIF retrievals made at a proximal scale, radiometric calibrations and corrections were performed for electronic processing issues and atmospheric O_2 absorbance on five field-deployed spectrometers. Dramatic changes were found in SIF retrieval magnitude before and after applying calibrations and corrections, as well as significant differences between instrument performance in the field and expected performance based on laboratory characterizations. Based on these tests, and on a Monte Carlo simulation of uncertainty estimates associated with each of these corrections, it is likely that calibration methodologies and hardware characterizations explain some of the observed variability in published SIF retrievals. This wide range in baseline SIF retrieval methodologies and resultant magnitudes severely limit researchers' ability to synthesize and advance the utility of SIF in modeling GPP across scales. Further, variability in calibration and correction methodology may explain the weak SIF-GPP relationship across studies at tower scales.[143] This paper highlighted the discrepancies in SIF publications and some instrument sensitivities that could account for some of the discrepancies found in the literature.

Since 2006, six satellites measuring solar-induced chlorophyll fluorescence (SIF) have been launched to better constrain terrestrial gross primary productivity (GPP). The promise of the SIF signal as a proxy for photosynthesis with a strong relationship to GPP has been widely cited in carbon cycling studies. However, a recent study found no change in SIF or steady-state PAM fluorescence, despite clear reductions in stomatal conductance, carbon assimilation, and light-use efficiency in treated leaves. These findings suggest that equating SIF and photosynthesis is an oversimplification that may undermine the utility of SIF as a biophysical parameter in GPP models.[144] This work showed that SIF does not always indicate photosynthetic rate at the leaf level, and it has been cited over 60 times since 2020.

Impact of Urban Forestation

Forest fragmentation impacts carbon uptake and storage; however, the magnitude and direction of fragmentation impacts on soil respiration remain poorly characterized. In a new study, soil respiration rates were quantified along edge-to-interior transects in two temperate broadleaf forests in the eastern U.S. that vary in climate, species composition, and soil type. Average soil respiration rates were observed to be 15% – 26% higher at the forest edge compared to the interior, corresponding to large gradients in soil temperature. No significant differences were found in the sensitivity of soil respiration to temperature between the forest edge and interior. Fragmentation and resultant shifts in microenvironment alter forest productivity and soil respiration near forest edges. Ecosystem models do not currently represent edge dynamics but given the prevalence of landscape fragmentation and its effect on carbon cycling along forest edges, this omission likely introduces an important source of uncertainty in our understanding of terrestrial carbon dynamics with a changing landscape and climate.[145] This paper showed that the ecological productivity of vegetation, primarily trees, varies significantly from the edge to the interior of forested areas. A 30 % difference in productivity can make a large difference in both the amount of CO_2 taken up by photosynthesis and that emitted by respiration in these fragmented urban forests or heavily vegetated areas. This work is likely to also impact non-urban environments where fragmentation is substantial.

3c. Advanced Techniques for Urban GHG Source Attribution from Atmospheric Measurements

Flux estimation methods using "top-down" techniques utilize atmospheric GHG concentration observations coupled with either simple or complex models to estimate the emissions or uptake fluxes from a given region. In any regional top-down analysis it is necessary to isolate the enhancement in the GHG concentration that is caused by fluxes in the region of interest. Thus, a background or baseline concentration, associated with the incoming air stream, must first be subtracted from the total observed concentration.[115]

One of the approaches used to quantify GHG fluxes in an urban environment is the **mass balance** method, which is typically applied to aircraft observations.[117] This approach utilizes direct measurements of the atmospheric boundary layer winds and depth from the airborne platform to approximate atmospheric transport across the city. Urban GHG emissions are calculated from an approximation of the atmospheric boundary layer GHG conservation equation.[146] The net mass flow of incremental GHG mole fractions above a measured background are integrated across an imaginary plane perpendicular to the wind direction, and across all elements of the outflow plume. This relies on precise mole fraction measurements inside and out of the plume, accurate wind speed measurements, and steady state wind conditions throughout the experiment.

Another approach is the **tracer-tracer ratio** method. In this method, simultaneous measurements of e.g., methane (CH_4), carbon dioxide (CO_2), carbon monoxide (CO) and ethane (C_2H_6) in the downwind plumes provide characteristic correlation slopes for each urban center that can be directly compared to current inventories. [118]

Another common approach is the **atmospheric inversion method,** used to infer GHG fluxes from airborne or tower-based in-situ measurements, or space-borne or ground-based remote sensing using a Lagrangian method. [147] Inverse problems are characterized as those where the chain of calculation or inference is in the opposite direction of real-world causality. "Tracer inversion" is defined as the process of deducing sources and sinks of emissions from measurements of concentrations in the atmosphere. The measured spatial distribution of CO_2 is used to infer the spatial distribution of surface fluxes. [148] Attempts to reconstruct the small-scale structure of the "cause" requires an amplification of attenuated details of the "effect"; this generally leads to amplification of errors. The atmospheric inverse problem is "ill-conditioned" because the diffusive nature of atmospheric transport means that relatively small variations or errors in observed or modeled atmospheric concentrations can correspond to relatively large differences or errors in the inferred fluxes. [149] Such "ill conditioning" is the basis for the definition of "inverse problems". Inverse techniques are therefore firmly based on the use of "prior" information; this is both an optimal use of available data and essential part of stabilizing an "ill-conditioned" problem. [150] The atmospheric inverse problem is often also "under-determined" because of the sparse set of observational data.

Atmospheric transport and dispersion models are fundamental tools for the inference of trace gas source magnitude and location. The model characterizes the transport of GHGs from an emission source, in both time and space, to where they are measured in the atmosphere using our understanding of atmospheric dynamics. Use of a numerical weather prediction (NWP) model, informed by atmospheric observations, creates a historical reanalysis of the state of the atmosphere over the time period of interest. [151] Additionally, large-scale atmospheric reanalysis products have found broad utility as an approach to interpolate data from limited atmospheric observational networks across space and time.[4] [152] [153]

One approach to account for atmospheric transport of trace gas species is to adapt the Weather Research and Forecast model with atmospheric chemistry (WRF-Chem) to simulate the transport of non-chemically reacting species like the greenhouse gases.[154] WRF-Chem can be formulated with many different domains, resolutions and parameterizations and, in historical mode, many different meteorological data sources can be used for assimilation to create a "reanalysis" product.[147] The accuracy and precision of

[4] Reanalysis data provide the most complete picture currently possible of past weather and climate. They are a blend of observations with past short-range weather forecasts rerun with modern weather forecasting models. They are globally complete and consistent in time and are sometime referred to as 'maps without gaps'. https://www.ecmwf.int/en/about/media-centre/focus/2020/fact-sheet-earth-system-data-assimilation

inference of urban GHG fluxes depends on the accuracy and precision of these atmospheric re-analyses. There are two approaches to quantifying and minimizing atmospheric transport model error. One approach is assimilation of atmospheric observations; data assimilation combines the strength of direct, local measurements of the atmospheric state with the representation of atmospheric governing equations contained in a numerical weather model. Meteorological observations such as surface and upper-air winds, temperature and moisture fields, and Doppler Lidar observed winds are assimilated into the models. Another approach to reducing transport model errors is testing and ultimately improving model structure and input data. These include investigation of the impact of updated land surface data and Atmospheric Boundary Layer (ABL) parameterizations on the accuracy and precision of the meteorological reanalysis.[155]

A Lagrangian Particle Dispersion Model (LPDM) may be used to associate atmospheric GHG observations to sources of GHG emissions that influence the GHG mole fractions at those observation points.[156] LPDM methods utilize the wind and turbulence fields produced by WRF-Chem or other atmospheric transport models to run backwards-in-time trajectories from observation points, mapping out the surface areas that contribute to any given atmospheric GHG observation. Particles are "released" from the observation points (e.g., tower sites) and tracked backwards in time. Times and places where the particles are within the atmospheric boundary and surface layers in this simulation are recorded, forming a function showing when the observations would be influenced by the surface (influence function, also referred to as a "footprint").[157] Convolution of the influence function with an estimate of surface fluxes yields the contribution of surface fluxes over the defined time period and within the chosen domain to the GHG mole fraction at the observation point. [151]

One type of atmospheric inversion method is a Bayesian inversion that incorporates atmospheric GHG mole fraction measurements, the atmospheric transport reanalysis products, the LPDM, and a "prior" estimate of urban GHG emissions as illustrated in Figure 13. [151] [153] One approach is to assume that the GHG boundary conditions for the city are represented by GHG concentration measurements made at an upwind tower(s) (background) and solve for GHG "enhancements" relative to this background measurement; these "enhancements" are then used to estimate GHG fluxes This method also requires quantitative estimates of the uncertainties in the prior fluxes, atmospheric observations, and atmospheric transport re-analyses.

The Bayesian method provides a rigorous mathematical framework to quantify uncertainties. However, to obtain reliable inversion-optimized emissions and corresponding uncertainties using Bayes requires a reliable characterization of the uncertainties of the "prior" fluxes, the observations, and the atmospheric transport and dispersion models. In the inverse modelling framework, the characterization of uncertainties represents one of the most fundamental steps to produce reliable inverse solutions and to quantify rigorously the uncertainties of the fluxes produced by the inversion.[146] At most spatial scales, from continental to urban, direct evaluation of the performance of inversions is difficult due to the absence of independent estimates of the fluxes at those scales. However, several methods have been suggested to assess the accuracy of the inversion results; these include: a) evaluation against unused atmospheric observations; b) evaluation at aggregated scales against large-scale scientific constraints; c) statistical diagnostics of inversion results; d) sensitivity tests and analysis of robustness; and e) synthetic data experiments. [149] Such approaches could become part of commonly agreed upon quality control procedures.

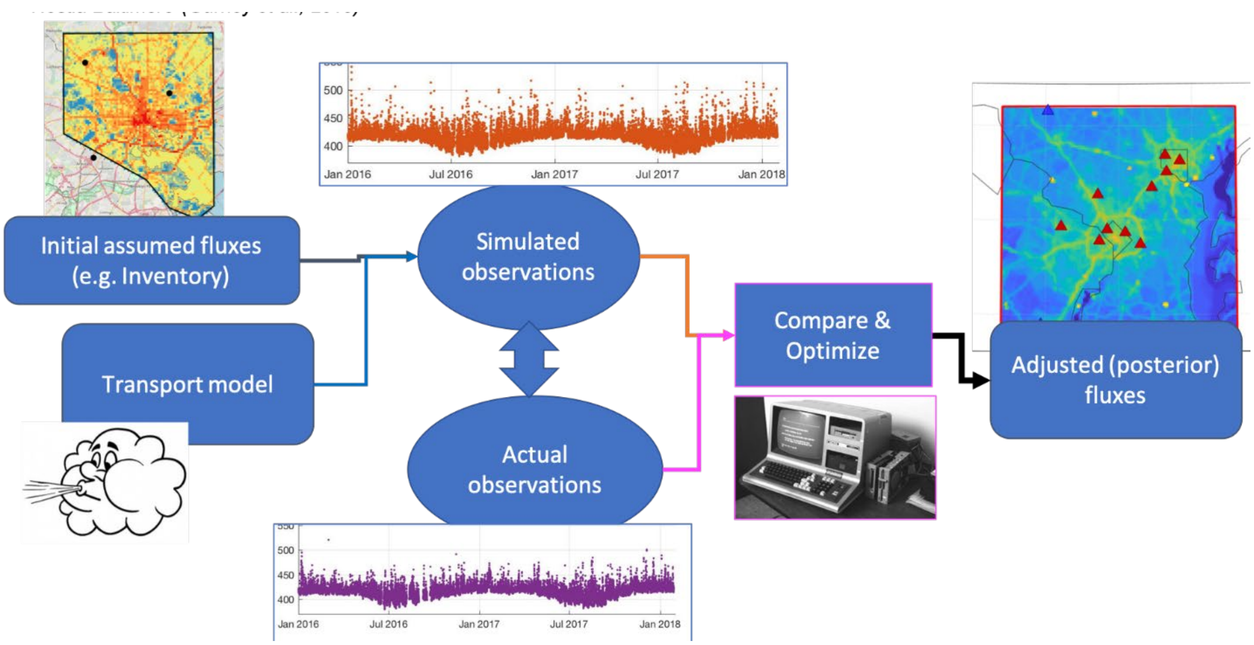


Figure 13 - A bird's-eye overview of the top-down flux estimation process. Components such as transport models, statistical optimization methods, and biogenic emissions modeling are areas of active research at NIST. (Figure courtesy of Anna Karion)

The inverse modelling community has also been relying on inter-comparison exercises to assess the robustness of flux estimates. Other approaches have involved the comparison of the optimized model with independent atmospheric measurements or to evaluate the sensitivity to specific components of inversion systems such as transport models and the "prior" information. These approaches have been used to quantify GHG emissions from metropolitan areas, [158] as well as agricultural areas.[159] At NIST, such Bayesian inverse modeling approaches are used and the impact of different model setups on estimated fluxes is a continuing area of research.

The ability of the transport model to simulate actual transport correctly or adequately will directly impact the quality of the estimated emissions and, although these models have come a long way and appreciable progress has been achieved, uncertainties remain. Advances in our limited understanding of and ability to represent the dispersion processes and turbulent mixing on the spatial and temporal scales involved are expected to significantly improve our capability to simulate these processes in transport models.

To better understand atmospheric transport, turbulent mixing, and dispersion, and to improve their representation in models, NIST, in collaboration with NOAA's Chemical Sciences and Air Resources Laboratories, is conducting research in mixing parametrizations, near-field dispersion, turbulence "grey-zone" and urban area impacts like the Urban Heat Island and the enhanced "roughness" induced by buildings.[160] In this research, a combination of measurements from aircraft and ground-based Doppler LIDARs are used with a suite of modeling tools such as Numerical Weather Prediction models, Large Eddy Simulations, and Lagrangian Particle Dispersion Models.

As discussed earlier, high-resolution meteorological modeling (using the Weather Research and Forecast (WRF) model) is often performed to generate the relevant meteorological field. This output is coupled to Lagrangian dispersion models such as the Stochastic Time-Inverted Lagrangian Transport (STILT) model [157] and the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model [140] to calculate a "footprint". The model is a tool to determine surface fluxes from atmospheric concentration data in the midst of distributed sources or sinks over land. STILT, based on the HYSPLIT model, is a complete system for computing simple air parcel trajectories, as well as complex transport, dispersion, chemical transformation, and deposition simulations have been merged and are available from NOAA/ARL. A common application is a back trajectory analysis to determine the origin of air masses and establish source-receptor relationships. HYSPLIT has also been used in a variety of simulations describing the atmospheric transport, dispersion, and deposition of pollutants and hazardous materials. The HYSPLIT model is used in a mode that allows it to emulate STILT and then compute the sensitivity of observations to surface fluxes or footprints.[161]

In a recent study, to assess the uncertainties introduced by transport models, HYSPLIT was driven with five different meteorological products: the High-Resolution Rapid Refresh (HRRR) NOAA operational forecast product and four configurations of Weather Research and Forecasting (WRF) model. [116] Within WRF, local and non-local Planetary Boundary Layer (PBL) schemes were used to represent the PBL dynamics; the North American Regional Re-analysis (NARR) product [162] was used to provide the initial and boundary conditions.

NIST researchers also take advantage of an extensive regional network of in situ atmospheric CO_2 measurement towers to model and evaluate fluxes. Atmospheric CO_2 data provides the opportunity to evaluate fluxes at regional scales (10's to 100's of km) seen by the atmosphere, and not just the small footprints (~ 1km^2) measured by the flux towers used in eddy covariance methods.

Observations of greenhouse gas concentrations in the atmosphere are the backbone of the analysis system. Emissions lead to elevated concentrations downwind of sources (and reductions downwind of sinks, such as trees, parks, or crops), which are measured directly using various instrumentation methods and platforms. These are used in various analyses to estimate emissions. Observations of meteorological parameters (wind speed or turbulence, for example) are also used either to evaluate transport models or directly in estimation of emissions.

4. Urban Testbeds for Monitoring GHG Fluxes

Urban environments are estimated to be responsible for about 70% of global GHG emissions. As a result, many GHG mitigation efforts are focused on cities and metropolitan areas. As illustrated in Figure 14, cities and metropolitan areas are complex, comprised of many types of human activities, with GHGs emitted from industrial sites, power generation units, transportation vehicles, commercial and residential buildings, landfills, etc. Accurate assessment of the impact of such mitigation efforts requires high-quality GHG emissions data due to the many different sources within a city, as well as data on the role biogenic areas play within and from surrounding areas.

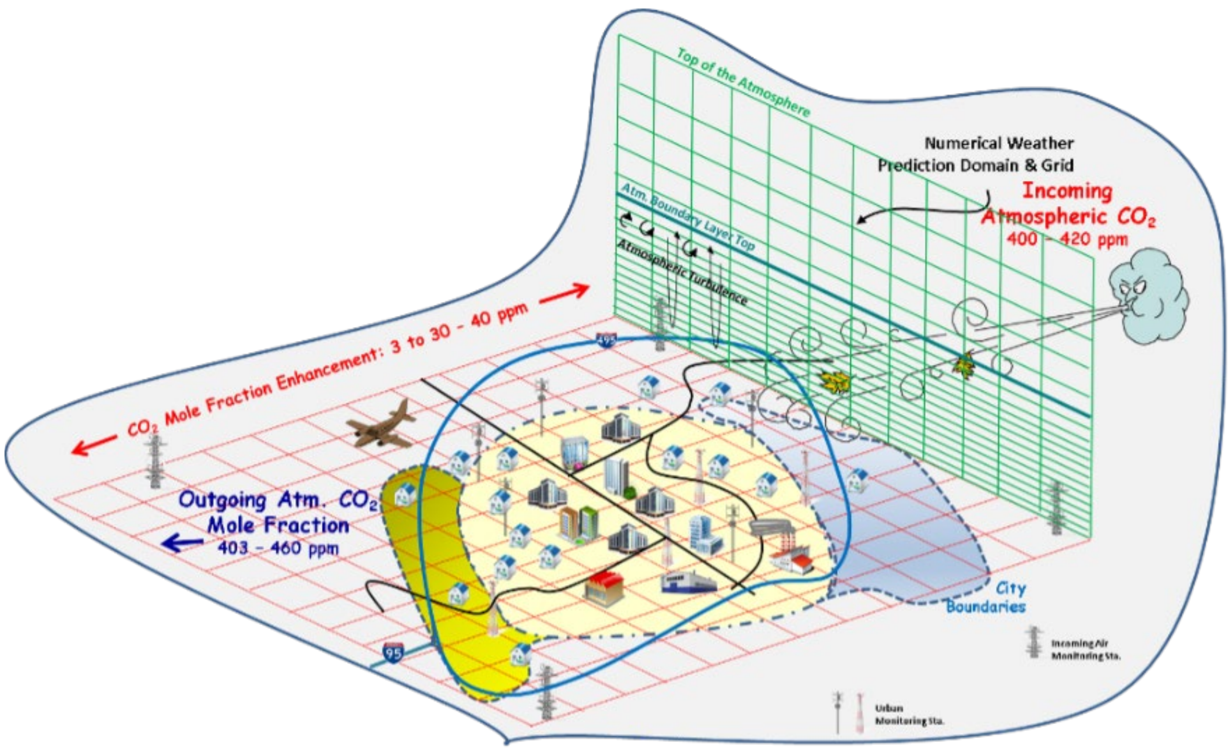


Figure 14. A schematic depicting the sources of GHG emissions in an urban environment and transport of GHGs into and out of the urban environment by atmospheric dynamics.

Instruments installed on aircrafts and surface observing networks, often located on communication towers to continuously sample as much of the planetary boundary layer as practical, measure atmospheric GHG concentrations across the city's domain. As discussed in previous sections, top-down concentration data analyses, coupled with meteorological simulations via Numerical Weather Prediction (NWP) models, translate these measurements to emission fluxes. If a spatially explicit emission model exists for a city, these bottom-up data are compared with top-down data, perhaps via Bayesian or other optimization methods. This comparison, at a minimum, has the effect of calibrating or verifying emissions modeling results, in addition to serving as a means of investigating the source(s) of discrepancies. Emission models such as **HESTIA** usually have spatial resolutions finer than the NWP models, which are used with grid resolutions of 1 km or 3 km in urban applications. Grid spacings are matched by upscaling emission model estimates. Comparison of the top-down and bottom-up determinations at grid cell levels can be a powerful diagnostic tool to identify potential improvements to either or both estimation methods.

Prior to 2010, most atmospheric greenhouse gas measurement capabilities were focused on continental and global scales.[27] Although the Urban Testbeds System, with its dense observations, is unique for monitoring GHGs in cities, many cities established air quality observing networks to meet regulatory requirements that have significantly improved urban air quality. Some of these activities were expanded to monitor GHG concentrations as well. One of the first urban test beds established in the U.S. was in Salt Lake City, UT. [163, 164], [165] [166] Some of the earliest work on monitoring of GHGs in urban environments was performed in Paris, France. [167] [168] Given the high degree of similarity in the analysis methods for both of these applications, NIST and NOAA have established an effort, described later in this work (Section 5), to explore the benefit of combining these in the Greenhouse Gas And Air Pollutant Emissions System (GRAAPES).

To provide high-quality data on the role that cities play in climate change, and to facilitate assessment of mitigation efforts, NIST initiated an Urban Testbed Program. The first testbed was established in Indianapolis, a second testbed in Los Angeles, and a third, and much more extensive, testbed along the Northeast Corridor of the U.S., starting in the Washington, DC/Baltimore area, with plans to extend it all the way to Boston, MA.

Combining high-quality, mole fraction measurements with well-characterized meteorology can provide flux estimates with stated uncertainties.[159] Use of these to assess satellite instrument performance, such as those on GOSAT, OCO-2, and future satellite missions, for example GeoCARB and OCO-3, contributes to improving the quality of both atmospheric observation-based estimates and traditional GHG inventory data and reports.

4a. **Indianapolis Flux Experiment (INFLUX)**

The INFLUX project was the first Urban Test Bed to be established by NIST. Initiated in 2010 with funding provided to NIST under the American Reinvestment and Recovery Act, it was subsequently expanded. INFLUX began as a collaborative effort between Purdue University, the University of Colorado, NOAA's Global Monitoring Laboratory, and the Pennsylvania State University (PSU).

The objective of INFLUX was to improve the quality of emission inventory data by investigating, assessing, and improving the performance of emissions measurement approaches with high spatial and temporal resolution.[147] Indianapolis was chosen because of its moderate size (about 1.8 million inhabitants in the city and surrounding area), relative isolation from other strong emissions sources, location in a landscape of flat terrain with minimal impact on its meteorology, and the existence of HESTIA in its early stages for the bottom-up component of the research. These features made the challenge of measuring GHG emission fluxes in Indianapolis tractable relative to other urban settings. In addition to being excellent test beds, urban regions with well-characterized GHG emission flux estimates can provide a useful information base for comparison to and assessment of remote sensing instrument performance whether these are satellites, airborne, or surface-based platforms.

Figure 15. Instrumented Beechcraft BE76 Duchess aircraft that serves as Purdue University's Airborne Laboratory Atmospheric Research (ALAR) www.science.purdue.edu/shepson/research/ALARGreenhouseGas/

The INFLUX project was funded wholly by NIST since its inception as a measurement science research project reflecting NIST GHG measurements program goals. The first goal was to advance urban GHG quantification capabilities based on traditional economic-sector and emission process-based methods (the bottom-up approach),[125] as well as those based on atmospheric observations (the top-down approach). [151] Most emissions inventories are based solely on bottom-up methods, and generally report on a yearly basis by economic sector. In contrast, top-down methods yield time- and location-specific data on scales of hours to weeks and several to tens of kilometers. The second, longer-term project goal was to develop methods for objectively comparing the results from these two method classes to move toward a comparative capability, or reference frame, a challenge for both method classes. Evaluating their relative performance provides critical insights into the strengths and deficiencies of both and identifies paths toward improvement. Finally, combining information from both approaches is likely to produce results that have greater accuracy, lower uncertainties, than either approach alone. The atmospheric observation methods utilized as part of INFLUX included aircraft mass balance methods designed to measure whole-city emission fluxes.[117] While these methods do not provide spatially resolved emissions information, they can be rapidly deployed to many environments and produce a quantitative snapshot of emission flux information. INFLUX was initially designed as a mass balance experiment, combining aircraft and surface network observations, including ^{14}C measurements to isolate fossil fuel CO_2 emissions, taken at two locations across the city along the prevailing wind direction.

Shortly after INFLUX began, NIST expanded its efforts to investigate the performance of a spatially dense, surface-based atmospheric observing network. These measurements coupled with atmospheric inversion methodologies provide capability to locate and quantify emission fluxes within an urban environment or similarly sized region. By 2012, the PSU element of the INFLUX team deployed mole fraction measurement devices at existing communication towers to sample the atmosphere at heights from 40 to 130 meters. This effort expanded to include 12 towers; one of them was located at a background inflow position; others were located throughout the city core and downwind. [169] (See Table 4) The surface observing network continuously measures the city's incremental concentrations of carbon dioxide (CO_2) and methane (CH_4).[170] In addition to real-time measurements, several observing points were fitted with flask sampling devices. Analyses of these flask samples included $^{14}CO_2$ and approximately 40 other atmospheric trace gases. To improve performance of atmospheric simulation, a Doppler LIDAR instrument was installed to provide real-time estimates of boundary layer heights and dynamics. With variable wind direction data and the use of Bayesian inversion methods, this network yields emission fluxes that are spatially resolved to approximately 1 km^2 – 2 km^2.

<u>Emissions Modeling for Indianapolis and Surrounding Counties</u>

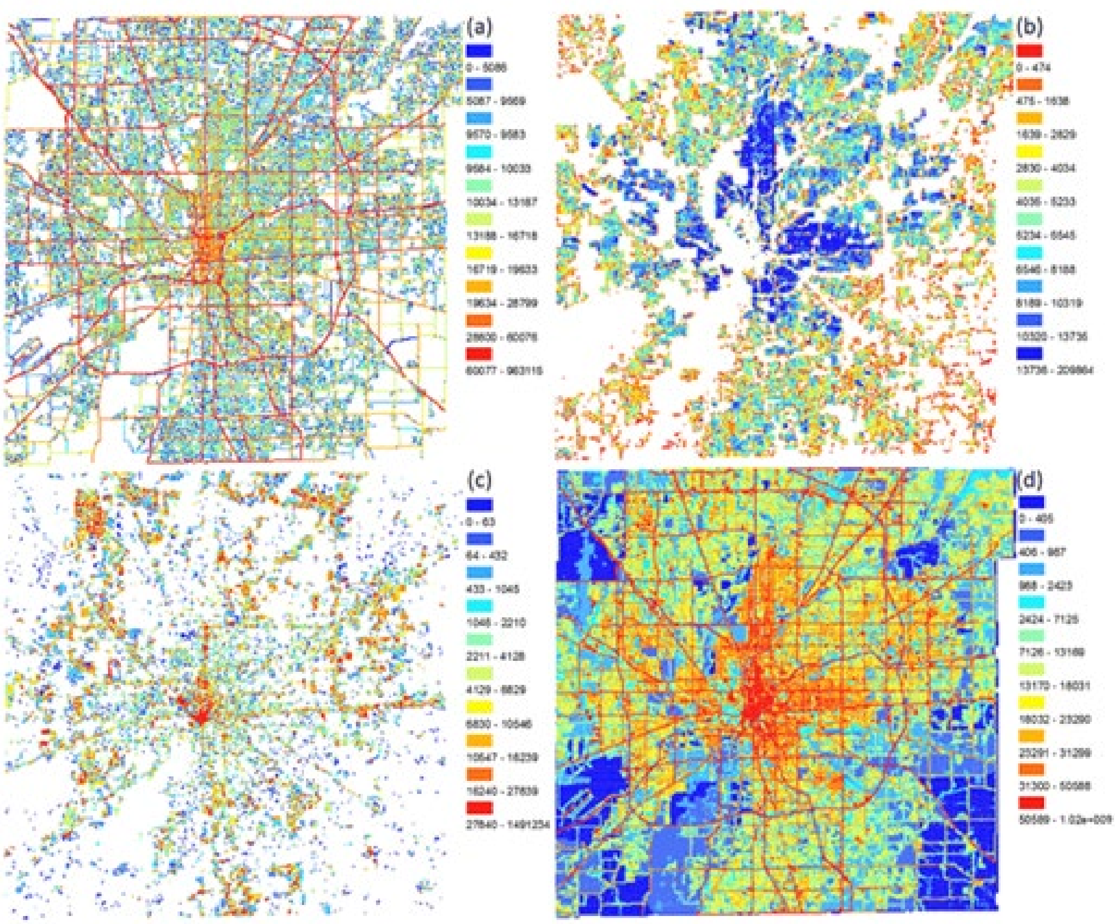


Figure 16. Map of gridded $FFCO_2$ emissions. Marion County, Indiana 2011 $FFCO_2$ emissions from the HESTIA version 3.0 gridded at 100 m × 100 m resolution for the a) on-road sector; b) residential sector; c) commercial sector; d) total emissions. Units: kgC/yr/grid cell. doi.org/10.1525/elementa.137

As mentioned before, when INFLUX was launched, an extensive and sophisticated emissions model, HESTIA, was under active development at Purdue University. **HESTIA** employs a bottom-up emissions modeling approach based on detailed data from local socioeconomic activities to produce fossil fuel CO_2 emissions flux estimates with temporal and spatial resolutions that equal or exceed those of the top-down approaches used in INFLUX. [171] Indeed, one goal of **HESTIA** was to provide local decision-makers with high-fidelity information useful in assessing and improving effectiveness of their CO_2 emissions reduction strategies. **HESTIA** is an important component of INFLUX, providing the means to compare bottom-up emission flux determinations from **HESTIA** with the top-down results of several atmospheric observing methods.[151] [172] Atmospheric inversion analyses for CO_2 have been a mainstay of INFLUX research. Such analyses use **HESTIA** data as an initial estimate for CO_2 emissions.

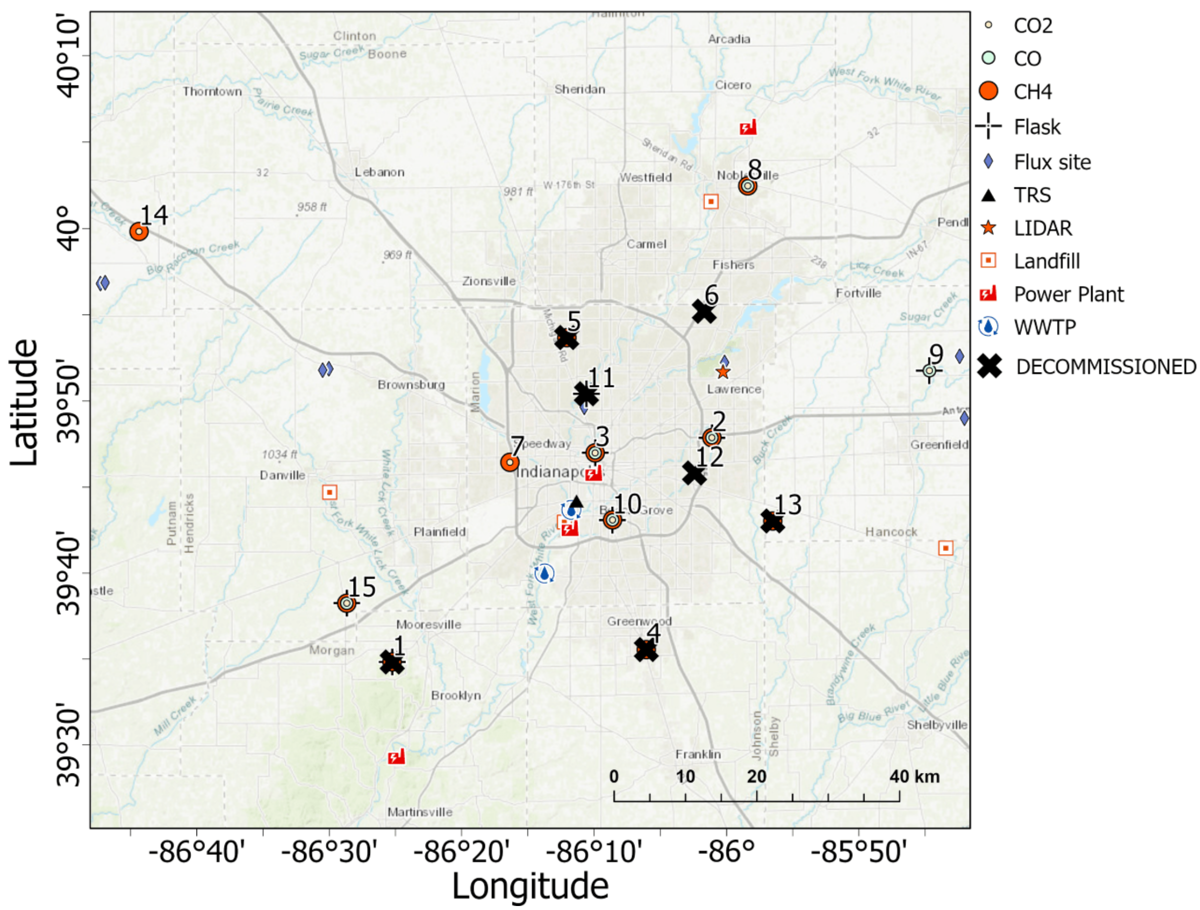


Figure 17. Historical tower network for the INFLUX testbed. Some sites have since been decommissioned, and new ones established. Locations of 14 towers are indicated along with the specific GHG measurements performed at those towers, as well as those towers where flask samples are collected. sites.psu.edu/influx/data/description-psu-in-situ/

Table 4. INFLUX Project Tower Sites, installation dates, latitude/longitude, and sampling heights at each tower in Indianapolis, IN, USA. (Decommissioned sites are marked by *)
https://sites.psu.edu/influx/site-information/

Tower Site	Install Date	Latitude (N)	Longitude (W)	Sampling Height(s)
				(m AGL)
Site 01 - Mooresville*	9/22/2010	39.5805	86.4207	10/40/121
Site 02 - E. 21st St.	9/24/2010	39.7978	86.0183	10/40/136
Site 03 - Downtown	6/1/2012	39.7833	86.1652	10/20/40/54
Site 04 - Greenwood*	8/8/2012	39.5925	86.1009	60
Site 05 - W. 79th St.*	3/30/2012	39.8947	86.2011	125
Site 06 - Lambert*	7/16/2013	39.9201	86.0280	39
Site 07 - Wayne Twsp Comm.	3/29/2012	39.7739	86.2724	21/58
Site 08 - Noblesville	5/28/2013	40.0411	85.9734	41
Site 09 - Greenfield	3/30/2012	39.8627	85.7448	130
Site 10 - Garfield Park	3/28/2012	39.7181	86.1436	40
Site 11 - Butler Univ.*	4/17/2013	39.8403	86.1763	130
Site 12 - 415 S. Shortridge Rd.*	8/9/2012	39.7637	86.0403	40
Site 13 - Pleasant View*	4/17/2013	39.7173	85.9417	87
Site 14 - Crawfordsville	4/26/2017	39.9971	86.7396	76
Site 15 - Clayton	6/4/2020	39.6376	86.4779	75

Although the **HESTIA** modeling approach is quite effective for fossil fuel CO_2 emissions, application to methane emissions is considerably less mature. However, progress is being made toward a similar emissions data product for methane.[173] Several ground-based and aircraft campaigns, most aimed at identifying and quantifying important point sources of methane emissions, have also been developed using Indianapolis as a testing location.[174, 175] [176] Progress on INFLUX, and similar urban studies begun after the start of INFLUX, indicate that the combination of top-down and bottom-up methods has excellent potential for providing urban CO_2 emission flux measurements sufficiently accurate to clearly assess progress toward Nationally Determined Contribution (NDC) targets agreed upon as part of the Paris Agreement. Indeed, the combination of methods strengthens the result of the comparison itself by identifying the strengths and weaknesses of each. For example, with bottom-up methods, unknown sources are not included in data products such as **HESTIA**, nor in U.S. GHG inventory reports. On the top-down approach, large discrepancies with **HESTIA** may reflect uncertainty, or they may point toward the possibility of an unknown source in an area of the city. For example, INFLUX demonstrated that atmospheric inversion could identify sources not included in the prior estimate obtained from HESTIA. In that case, further investigation, including potential sources of bias in the inversion method, revealed that a small power plant located on the outskirts of the city had not been included in the HESTIA bottom-up estimates because its emissions were below EPA reporting requirements.

The INFLUX Project had a running start. Some aircraft-based measurements of the carbon footprint of Indianapolis had already been performed [177], and some early work on development of the HESTIA model for Indianapolis had been initiated.[178] [125] This work represented the first quantification of CO_2 emissions from a large U.S. city utilizing a dense observing network. Some of the early measurements of GHGs were performed using an integrated flask sample collection system, which also included $^{14}CO_2$ measurements that help identify non-anthropogenic contributions.[179] Results of a more detailed study were published later which quantified CO_2 emissions from the city, accounting for the background levels, and identified some of the source sectors contributing to the fossil fuel CO_2 emissions.[180] Another study assessed the uncertainties of aircraft-based mass balance approach for quantifying urban GHG measurements.[146] Some of the first results of the high-resolution atmospheric inversions were published in 2016.[151] Methane emissions from Indianapolis were also quantified. [181] [182]

A special issue of the journal ***Elementa: Science of the Anthropocene*** featured 14 articles describing on-going measurement science research aimed at improving GHG emission flux quantification methods within Indianapolis and the surrounding region. The first article provided an overview of the INFLUX Project.[27] Another article provided a review of the INFLUX and provided a summary of recent accomplishments.[147] These included quantification of urban atmospheric GHG enhancements using aircraft and towers with measurements collected over multiple years; the project also provided estimates of whole city CO_2 and CH_4 emissions using measurements made with aircraft and on towers, and bottom-up inventory methods. Significant differences were identified between these methods and resolution of such differences continued to be the subject of research. Another topic of future work was distinguishing anthropogenic from biogenic CO_2 fluxes. Other papers in this series described efforts to reduce transport errors in inversion systems, [153] assessment of the biases and accuracies of simulated meteorological fields, and the impact of variables such as air temperature in the planetary boundary layer (PBL), the height of the PBL, wind speed and direction.[155] Another study tried to reconcile the differences between bottom-up and inverse-estimated CO_2 emissions. [183] Results were published of another study that assessed the precision of aircraft balance method for measurement of averaged urban greenhouse gas emission rates.[117] This was quite important since the aircraft mass balance is a practical method for rapid assessment of urban GHG emissions and any changes as a result of mitigation efforts. Two articles were published, involving multi-species and isotopic CO measurements, that could help source apportionment of CO_2 emissions. [184] [185] Evaluation of the impact of nearby sources, and sinks, on the urban atmospheric inversions is an important concern, and this topic was explored in a different publication.[186] Finally, another study assessed the sensitivity of inverse flux estimates to various measurement deployment strategies, biogenic CO2 fluxes and atmospheric transport errors.[187] Clearly, the extensive sets of data obtained as part of the INFLUX Project, including tower- and aircraft- based measurements and flask collections, have provided the framework for evaluation of different measurement methods and computational tools and measurement uncertainties. The INFLUX data was also utilized to evaluate the impact of the granularity of space-based CO_2 measurements on atmospheric inversions.[188]

Networks of tower-based CO_2 mole fraction sensors have been deployed by various groups in and around cities across the world to quantify anthropogenic CO_2 emissions from metropolitan areas. A critical aspect in these approaches is the separation of atmospheric signatures from distant sources and sinks (i.e., the background) from local emissions and biogenic fluxes. A study examined CO_2 enhancements compared to forested and agricultural background towers in Indianapolis, IN as a function of season and compared them to modeled results, as a part of the Indianapolis Flux (INFLUX) project. It was determined that growing season enhancements at urban sites relative to heavily vegetated background sites, on a monthly basis, were 2.6 times larger than those during the dormant season, pointing out the importance of proper accounting for biogenic fluxes and the determination of the background. [189]

INFLUX has continued to provide detailed information on urban GHG emissions and enabled comparison of different approaches to quantify GHG emissions. A more recent study compared results of CO_2 emissions estimation methods, including an inventory-based method and two different top-down atmospheric measurement approaches implemented in Indianapolis, IN urban area during the winter months. By accounting for differences in spatial and temporal coverage, as well as trace gas species measured, agreement was achieved among the wintertime whole-city fossil fuel CO_2 emission rate estimates within 7%. [158] This finding represented a major improvement over previous comparisons of urban-scale emissions, making urban CO_2 flux estimates from this study consistent with local and global emission mitigation strategy needs. A follow-up study demonstrated how advanced assimilation approaches could detect sub-city fluxes with sufficiently small uncertainty to track potential mitigation policies by associating atmospheric information with emissions from various data sources, including biogenic fluxes, on human activities. Results showed an agreement within 3% between atmospheric and bottom-up approaches on an annual basis. [190] It was also shown that, when a bias of 15% were introduced into the HESTIA prior, the atmospheric inversions produced a correction of -14.2%, removing nearly all of the introduced bias. (See Figure 18)

Significant scientific and technological challenges remain in achieving accurate determination of urban GHG fluxes. These include improving CO_2 flux measurement and quantification, which will help those faced with the challenge of more effectively managing emissions mitigation efforts. Another challenge is improving the quantification of urban methane emissions. Some advances have been made in this area; a recent paper described process-based estimation of CH_4 emissions from sources in Indianapolis, IN and compared these with atmospheric inferences of whole city emissions. [191] Emissions from the natural gas distribution system were estimated from measurements at metering and regulating stations and from pipeline leaks. Tracer methods and inverse plume modeling were used to estimate emissions from a major landfill and wastewater treatment plant. These direct source measurements informed the compilation of a methane emission inventory for the city. A follow up study evaluated the uncertainties in estimating urban methane fluxes as part of the INFLUX Project. [192] Some of the reasons for the discrepancies between various approaches were identified, including highly variable temporal and spatial distribution of the background CH_4 emissions, temporal variability of CH_4 emissions, and the fact that many CH_4 sources are not identified in public data sets, i.e., inventories. More progress on that front will likely occur in the next few years. Finally, the scientific community is improving its capabilities to distinguish between biogenic and combustion-based CO_2 emissions in urban settings. [180, 193] A recent study used ^{14}C measurements, to determine the ratio of CO to $FFCO_2$, and eddy-covariance measurements to decompose sources of CO_2 emissions. [194]

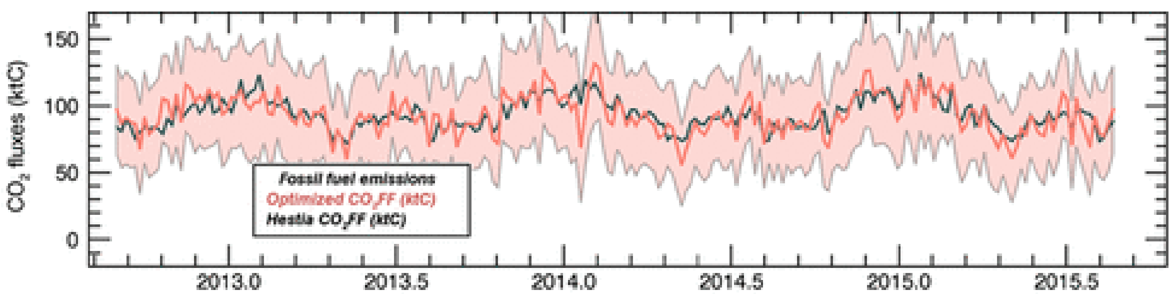


Figure 18. Whole-city 5 day averaged $FFCO_2$ emissions from the city of Indianapolis, IN over a three-year period (Sept. 2012 to Sept. 2015) https://doi.org/10.1021/acs.est.0c00343

4b. **The Los Angeles Megacity Carbon Project**

The LA Megacity Carbon Project,[195, 196] located in California's South Coast Air Basin, was the second location included in the NIST Urban Testbeds framework. This project was started in 2012. The Los Angeles region was chosen for several reasons; it is much larger than Indianapolis with more complex meteorology and emissions characteristics, and it is located directly on a coast where the ocean significantly impacts this meteorology, a characteristic shared by a significant number of megacities around the world. One of the goals of the project was to use atmospheric measurements of greenhouse gases (generally carbon dioxide and methane) to estimate emissions variability and trends in the South Coast Air Basin. A longer-term research objective of the Urban Testbeds system is the development of internationally recognized and broadly applicable measurement methodologies. INFLUX and LA Megacity represent at least two instances typical of cities.

Additionally, the capabilities of both private sector, governmental, and academic organizations located in the basin were quite substantial, from the perspective of both greenhouse gas and air quality observing capabilities. The NIST testbed is operated in collaboration with other GHG efforts in this region, including research and measurements funded and/or operated by NASA [197] and NOAA (whole air sampling systems; periodic aircraft campaigns, etc.).[198] The primary institutions working with NIST in this testbed are the Scripps Institution of Oceanography, NASA's Jet Propulsion Laboratory, and NIST's private sector partner, Earth Networks, Inc.

There has been a long standing interest in characterizing GHG emissions of California. Most of these studies had focused on characterizing methane emissions for the entire state. [199] [200] [201] [202] [203] One of the first studies to focus on the emissions of Los Angeles megacity utilized measurements at two sites, one at the coastal Palos Verdes peninsula (as an indicator of background levels) and another one in inland Pasadena.[204] Measurements were made over the 2006-2013 period, and compared with results of HESTIA-LA fossil CO_2 emissions product.[122] Another study used these findings to evaluate a high-resolution land-atmosphere modeling system, where the WRF-Chem model was coupled to the Vulcan 2.2 and HESTIA-LA emissions products.[205] They evaluated multiple WRF configurations and the impact of the spatial resolution of emission products and the meteorological transport model on spatiotemporal variability in simulated atmospheric CO_2 concentrations. The high spatial resolution of the fossil fuel CO_2 emissions was found to be more effective than that of the atmospheric model to capture CO_2 concentration variability across the LA megacity. Another study used data from three different observing systems: Total Carbon Column Observing Network (TCCON), Orbiting Carbon Observatory-2 (OCO-2) and the Greenhouse gases Observing Satelite (GOSAT).[206] They found column CO_2 ($\boldsymbol{X}_{CO2}$) results to vary significantly due to atmospheric dynamics and nonuniform distribution of sources. They observed persistent significant differences, even between two TCCON sites that were located only 9 km apart. Over the 2012-2014 period, measurements were also made in the Caltech campus with a Mini-MicroPulse LIDAR (MiniMPL) system to characterize atmospheric mixing over the LA megacity.[207] Complexity of the LA topography, meteorology, and emission sources clearly demonstrated the need for a denser network of monitoring locations to acquire reliable information on GHG emissions from the South Coast Air Basin (SCAB) containing the LA Megacity.

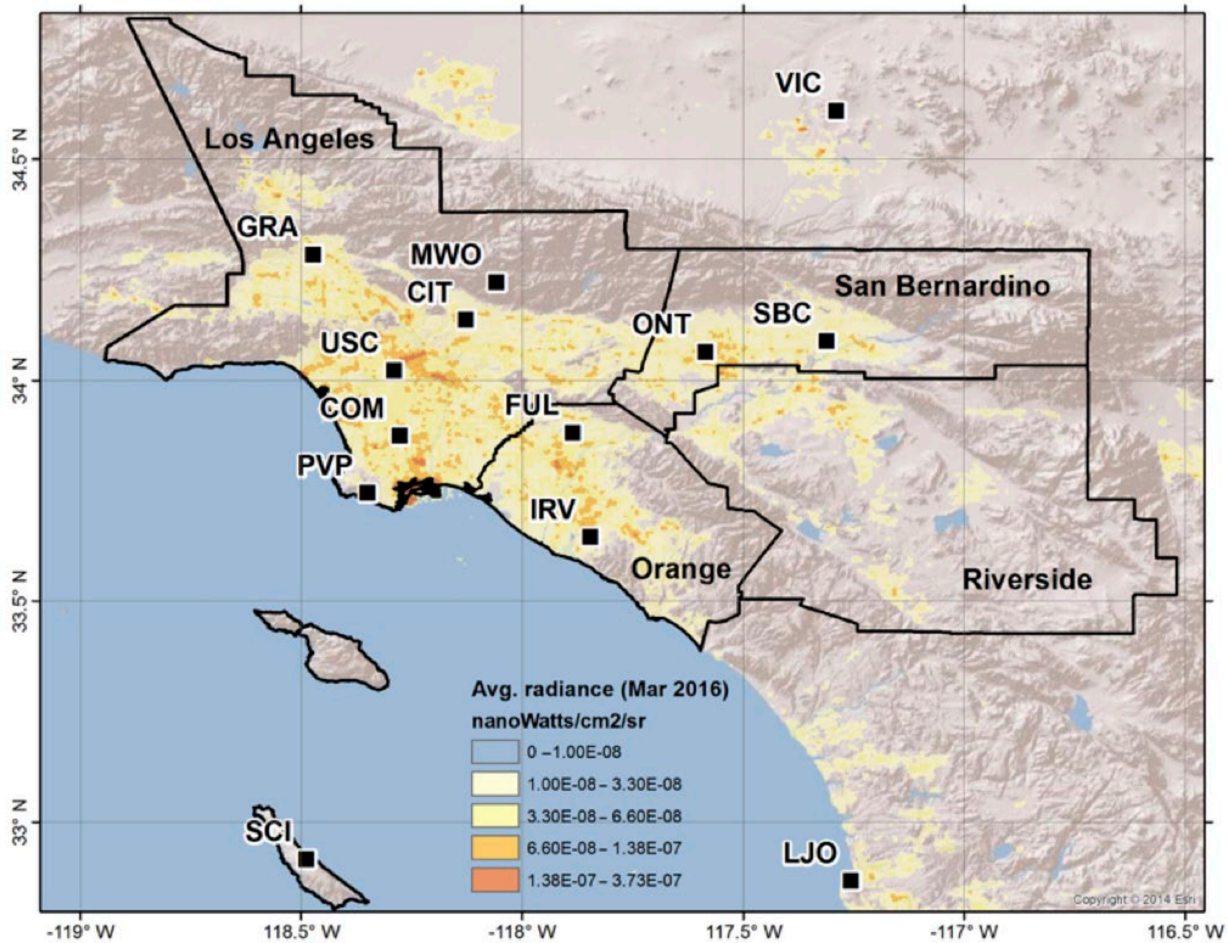


Figure 19. Map of the Los Angeles megacity and locations of the greenhouse gas monitoring network sites. The South Coast Air Basin is a geopolitical boundary including non-desert portions of the Los Angeles, Riverside and San Bernardino counties as well as all of Orange County. Network observing nodes are identified by location names given in Table 5.
doi.org/10.5194/acp-17-8313-2017

The motivation of this NIST project was to determine the emissions trends from a large metropolitan area with sufficient detail to enable characterization of GHG fluxes with some spatial and temporal resolution. The objective of this project was to:

1. develop and demonstrate scientifically robust capabilities to measure multiyear emission trends of carbon dioxide, methane, and carbon monoxide and to attribute these to selected major economic sectors, and
2. reduce uncertainty of emissions determination by jointly improving greenhouse gas emissions models and atmospheric observations of emissions.

Table 5. Site information for the Los Angeles Megacity Carbon Project surface network. Included are site codes, full site names, latitude/longitude of site location, site elevation, and inlet height(s). doi.org/10.5194/acp-17-8313-2017, 2017

Tower Site	Latitude (N)	Longitude (W)	Site Elevation (mASL)	Sampling Height(s) (m AGL)
VIC - Victorville[a]	34.61	117.29	1370	100/100/50
GRA - Granada Hills[a]	34.28	118.47	391	51/51/31
USC 1 - Downtown LA (Univ. So.Cal.)[b,c]	34.02	118.29	55	50
USC 2 - Downtown LA (Univ. So.Cal.)[b,c]	34.02	118.29	55	50
COM - Compton[a]	33.87	118.28	9	45/45/25
FUL - Fullerton (CSU Fullerton)[b,c]	33.88	117.88	75	50
IRV - Irvine (UC Irvine)[b,c]	33.64	117.84	10	20
SCI - San Clemente Island[a]	32.92	118.49	489	27
ONT - Ontario[a]	34.06	117.58	260	41/41/25
CNP - Canoga Park[b]	34.19	118.6	245	15
LJO - La Jolla (Scripps Pier)[b,c]	32.87	117.25	0	13
CIT-1 - Pasadena (Caltech Arms Lab.)[b,c]	34.14	118.13	230	10
CIT-2 - Pasadena (Caltech Hall)[b]			230	48
MWO - Mt. Wilson[b,c]	34.22	118.06	1670	3
PVP - Palos Verdes Peninsula[b,c]	33.74	118.35	320	3
SBC - San Bernardino[a,c]	34.09	117.31	300	27/58
RAN - Santa Monica[a]	34.01	118.49	17	24

The approach taken in the LA Megacity project was similar to the INFLUX work; they utilized a surface–based GHG observing network, coupled with state-of-the-art boundary layer measurements and atmospheric transport methodologies via NWPs. Tower and rooftop-based in-situ measurements of carbon dioxide (CO_2) and methane (CH_4) are the backbone of the LA Megacity test bed.[208] This surface-based component of the LA observing system includes a network of Greenhouse Gas (GHG) monitoring stations that is intended to encompass those portions of the South Coast Air Basin producing the most intense GHG emissions (the urbanized portions of Los Angeles, Orange, Riverside, and San Bernardino counties). Measurements began some years ago at several sites within the basin such as Caltech; sustained measurements have also been underway for many years from the Scripps Pier in La Jolla. Between 2013 and 2015 installation of a 14-node measurement network was completed within and around the LA basin. Most network sites are operated by Earth Networks, Inc., in collaboration with Scripps Institution of Oceanography, JPL, and NIST. [122]

Details of the Los Angeles Megacity Carbon Project network are provided in Table 5.[208] Network observing node sites (marked by (a)) include VIC, GRA, COM, SCI, ONT, and SBC; these stations (except for SBC) are operated by Earth Networks, Inc., with support from NIST. All other sites (marked by (b)) have rooftop configurations (USC, FUL, IRV, CIT, and CNP), with air inlets plumbed to the four corners of the building, sampling each corner every 15 min. These stations are also operated by Earth Networks and supported by NIST. For rooftop configurations, "upwind" hourly averages are computed using data from the upwind corner of the building determined based on the corner with the highest wind speed measurement. Rooftop inlet height indicates the total height above the surface (building + mast). Nodes marked (c) indicate the flask collection sites for ^{14}C observations. The remainder of the sites listed in Table 5 are supported and operated by other collaborators.

CO_2 flask observations began near the La Jolla (LJO) site in 1979. The Earth Networks configuration at LJO was implemented in January 2012 with an inlet that is located near Scripps Pier. At the time of this study, the USC site (marked by c) had two Picarro analyzers installed (model G2301 and G2401), referred to here as USC-1 and USC-2, respectively. Flask observations at the Caltech Arms Laboratory site (CIT-1) began in 1998. At Mt. Wilson site (MWO), flask data have been collected by NOAA/ESRL since 2010 and are included as part of the background analysis. There were three continuous analyzers installed at the California Laboratory for Atmospheric Remote Sensing (CLARS) facility near MWO, which are managed by the Air Resources Board.

Every site was equipped with at least one *in situ* gas analyzer providing high-accuracy, continuous measurements of carbon dioxide (CO_2) and methane (CH_4) mole fractions, and about half of the sites measure carbon monoxide (CO). Two of the sites were deployed in the high desert east of LA and one on San Clemente Island to help track the relatively clean "background" (incoming air) conditions and to track the outflow of polluted air from the basin. High-precision trace gas analyzers sample the air using pumps and air hose inlets located on towers or tall buildings. Every site is also equipped with one or more weather stations to help interpret the greenhouse gas measurements. As with the other two testbeds, tower-based continuous observations of CO_2, CH_4, and CO are calibrated to WMO scales and reported with time-varying uncertainties, calculated in the manner documented elsewhere.[208]First continuous surface observations of carbon dioxide (CO_2) and methane (CH_4) from the Los Angeles Megacity Carbon Project were performed during 2015 utilizing a large number of monitoring stations. Procedures were devised for calibration of instruments, methods for selection of background air masses, calculation of urban enhancements, and a detailed algorithm for estimating uncertainties in urban-scale CO_2 and CH_4 measurements.[208] These methods were essential for understanding carbon fluxes from the LA megacity and other complex urban environments globally. Background mole fractions entering LA were estimated using observations from four extra-urban sites including two marine sites located south of LA in La Jolla (LJO) and offshore on San Clemente Island (SCI), one continental site located in Victorville (VIC), in the high desert northeast of LA, and one continental/mid-troposphere site located on Mount Wilson (MWO) in the San Gabriel Mountains. A local marine background was established to within ~1 ppm CO_2 and ~10 ppb CH_4 using these local measurement sites. Overall, atmospheric carbon dioxide and methane levels were found to be highly variable across Los Angeles. Urban and suburban sites showed moderate to large CO_2 and CH_4 enhancements relative to a marine background estimate. The USC (University of Southern California) site near downtown LA exhibited median hourly enhancements of ~ 20 ppm CO_2 and ~150 ppb CH_4 during 2015 as well as ~15 ppm CO_2 and ~80 ppb CH_4 during mid-afternoon hours, which is the typical period of focus for flux inversions. The estimated measurement uncertainty is typically better than 0.1 ppm CO_2 and 1 ppb CH_4 based on the repeated standard gas measurements from the LA sites during the last 2 years, similar to [209]. The largest component of the

Figure 20. Air inlets and weather stations at the four corners of the roof of Caltech Hall on the Caltech campus in Pasadena, Calif. (Photo courtesy of Earth Networks, Inc.)

measurement uncertainty is due to the single-point calibration method; however, the uncertainty in the background mole fraction is much larger than the measurement uncertainty. The background uncertainty for the marine background estimate is ~10 % and ~15 % of the median mid-afternoon enhancement near downtown LA for CO_2 and CH_4, respectively. Overall, analytical and background uncertainties were estimated to be small relative to the local CO_2 and CH_4 enhancements; however, these results suggested that reducing the uncertainty to less than 5% of the median mid-afternoon enhancement would require detailed assessment of the impact of meteorology on background conditions.

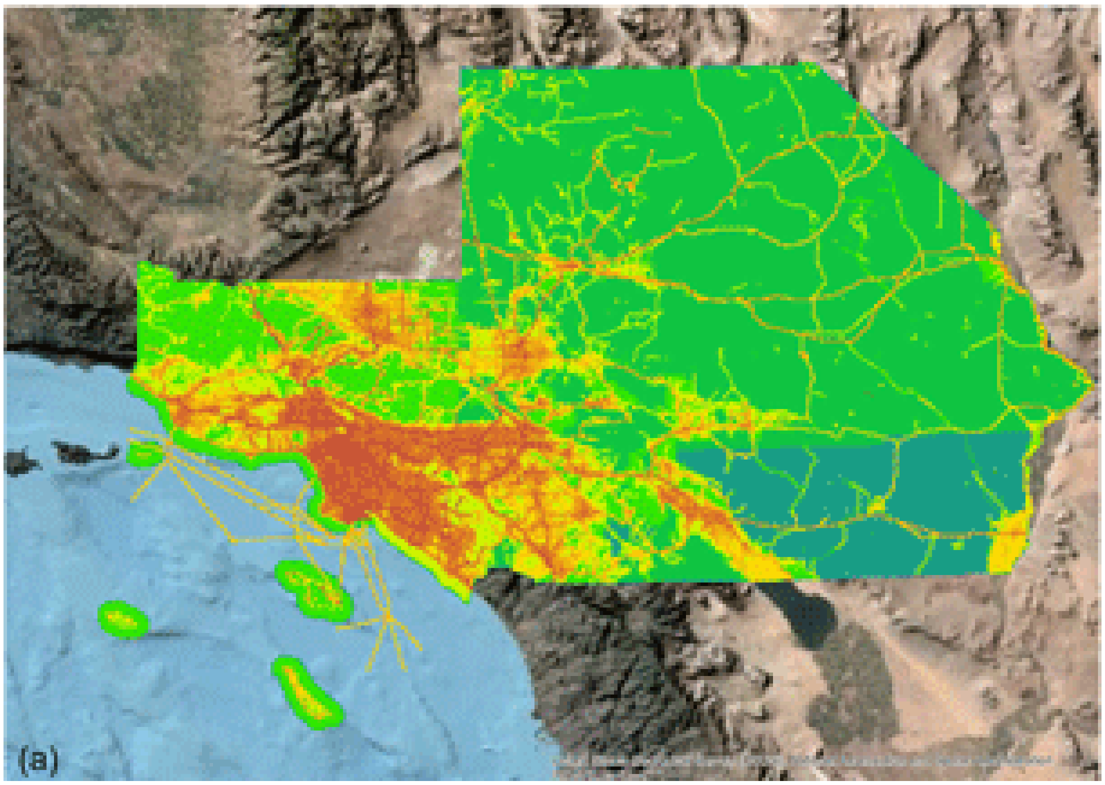


Figure 21. HESTIA-LA v.2.5 Total $FFCO_2$ emissions for the year 2011 represented on a 1 km x 1 km grid. Units: natural logarithm of kg C yr^{-1} doi.org/10.5194/essd-11-1309-2019

As mentioned earlier, methane leaks in California have been of concern for some time.[199] [201] [210] [211] [212] [213] [214] Methane emissions from oil and gas production have been of concern throughout the U.S.[215, 216] [217] [218] [219] These concerns were magnified after a blowout at an underground gas storage unit in 2015 in Aliso Canyon, CA, just north of Los Angeles.[220] The LA Megacity monitoring network and multiple airborne surveys were utilized to monitor the leaks and quantify the rate of emissions from this large source.[221] Near real-time capability of analysis, using aircraft-based observations, to detect the changes in urban emissions and the emissions from Aliso Canyon was demonstrated.[222] Another study combined observations from the network of atmospheric monitoring stations with inverse modeling to uniquely obtain spatiotemporal estimates of methane emissions from the Los Angeles megacity and the broader South Coast Air Basin for 2015-2016.[223] These inversions used customized and validated high-fidelity meteorological output from WRF to disentangle the spatiotemporal information content of observations as manifested through estimated fluxes. They were also able to track and constrain fluxes from the Aliso Canyon natural gas leak and detect closure of Puente Hills landfill, with no prior information of these events. The Aliso Canyon natural gas leak was also used a natural gas tracer experiment to assess errors in aircraft atmospheric inversion estimates of point-source emissions.[224]

Additional advances were made over the following years. An improved HESTIA-LA emissions data product was developed that provided higher spatial and temporal resolution.[129] Measurements made in 2015 of $\Delta^{14}C$ and CO_2 were used to separate biogenic and fossil contributions to CO_2 enhancements above background for LA megacity.[225] These results revealed high values of fossil CO_2 and a significant and seasonally varying contributions of CO_2 emission fluxes from the urban biosphere. The urban biosphere was found to be a source in winter and a sink during the summer months. The observing network and inversion methods were also instrumental in detecting the impact of COVID-19 on CO_2 emissions, which were reduced during this period as a result of reduced use of transportation, but also because of a mild winter.[226] These results were extended to detect the impact of COVID-19 lockdown in six North American cities (Los Angeles, Indianapolis, Salt Lake City, Boston, Northeast Corridor, and Toronto, Canada).[227] It is interesting to note that international colleagues have used the information generated by these studies to characterize the uncertainties in atmospheric inversions of fossil fuel CO_2 emissions in California.[228]

4c. **The Northeast Corridor Project**

The most recent testbed of NIST's Urban GHG Measurements Testbed System, the **Northeast Corridor (NEC) Project,** was established in 2014 as the third NIST urban testbed following those for Indianapolis and Los Angeles. The NEC project is currently quantifying GHG fluxes throughout the Washington and Baltimore urban regions. This surface network is being extended incrementally toward Philadelphia, New York, and ultimately Boston. The NEC utilizes monitoring stations operated by Earth Networks, Inc. whose data is contractually acquired by NIST and subsequently made public at regular intervals. The observing network was designed to have 12 to 16 observing nodes with 4 of these located near the corners of the perimeter encompassing the inner analysis domain.[229] These observing nodes are placed to better quantify GHG concentrations of the incoming atmosphere, thereby improving the observation and analysis system to quantify the emissions of each city itself. Several additional observing locations further removed from the area have been installed to aid in better understanding incoming atmospheric GHG burdens and in preparation for further instrumenting the NEC. Establishing the NEC Project coincided with expansion of NIST expertise into atmospheric inversion modeling and high accuracy field measurements of atmospheric concentrations; a close collaboration between NIST staff and Earth Networks Inc. was critical in achieving consistently high data quality.

The Northeast Corridor (NEC) Project is perhaps the most comprehensive of the testbed projects in a geographical sense extending from the Washington, DC/Baltimore, MD area to Boston. The region was selected because the meteorological complexity of the region was moderately more complex than that of Indianapolis, but significantly less so than the South Coast Air Basin of California. Utilization of these three regions for test beds provides a range of meteorological conditions that should allow excellent performance evaluation of emission measurement technologies developed in this program.

The Northeast Corridor is large geographically and composed of many governmental jurisdictions:

- States involved include Virginia, Maryland, Delaware, Pennsylvania, New Jersey, New York, Connecticut, Massachusetts, Rhode Island, New Hampshire and Maine, and the District of Columbia.
- Many of these states already participate in the Regional Greenhouse Gas Initiative (RGGI), the first market-based regulatory program in the United States to reduce GHG emissions. Outcomes of the testbed effort will contribute toward characterization of the RGGI initiative.
- Meteorology of the region is best described as plume oriented in that the northeast is absent significant air trapping areas such as those found in the LA basin, though it has complex terrain and coastal influences not found in Indianapolis.
- Within the region are various university, private and public sector organizations that possess pertinent expertise as well as existing infrastructure and ongoing initiatives that was expected to contribute to and be leveraged by this effort. Some of these organizations participated in the Indianapolis and Los Angeles testbeds.

While the NEC has similar meteorology to that of Indianapolis, it features substantial coastline and topographic relief. Although the NEC shares coastline features similar to those of Los Angeles, the latter has far more dramatic topography and the two have substantially different meteorology. For example, long-term air trapping is not prevalent in Indianapolis or the NE Corridor, although a frequent occurrence throughout the Los Angeles basin. Recent activities by researchers at the University of Maryland and NOAA on ozone transport in the Washington, DC/Baltimore, MD area have shown that to achieve reasonably good agreement between experimental and analytical results, it is critical to properly characterize the impacts of air flows associated with the Chesapeake Bay. As such, observational capabilities, both greenhouse gas and meteorological, that surround the NE Corridor and are placed within its urban domains will be required. To this end, the following capabilities were planned:

- Both real-time and flask GHG observations at tower locations in and around the area of interest.
- Enhancements to the Weather Research Forecasting (WRF) model,
  - Support higher levels of fidelity and sub-grid resolutions (e.g., the NIST high spatial resolution modeling to better investigate features below WRF grid scales of 1-2 km), and
  - Surface/atmosphere energy transport at appropriate scales for correct representation of regions ranging from rural (regional boundaries) to densely populated urban shall be evaluated for use in Indianapolis, LA Megacities, and the NE Corridor
- Meteorological observations in addition to those normally obtained from NWS and similar sources are needed to better constrain boundary layer transport,

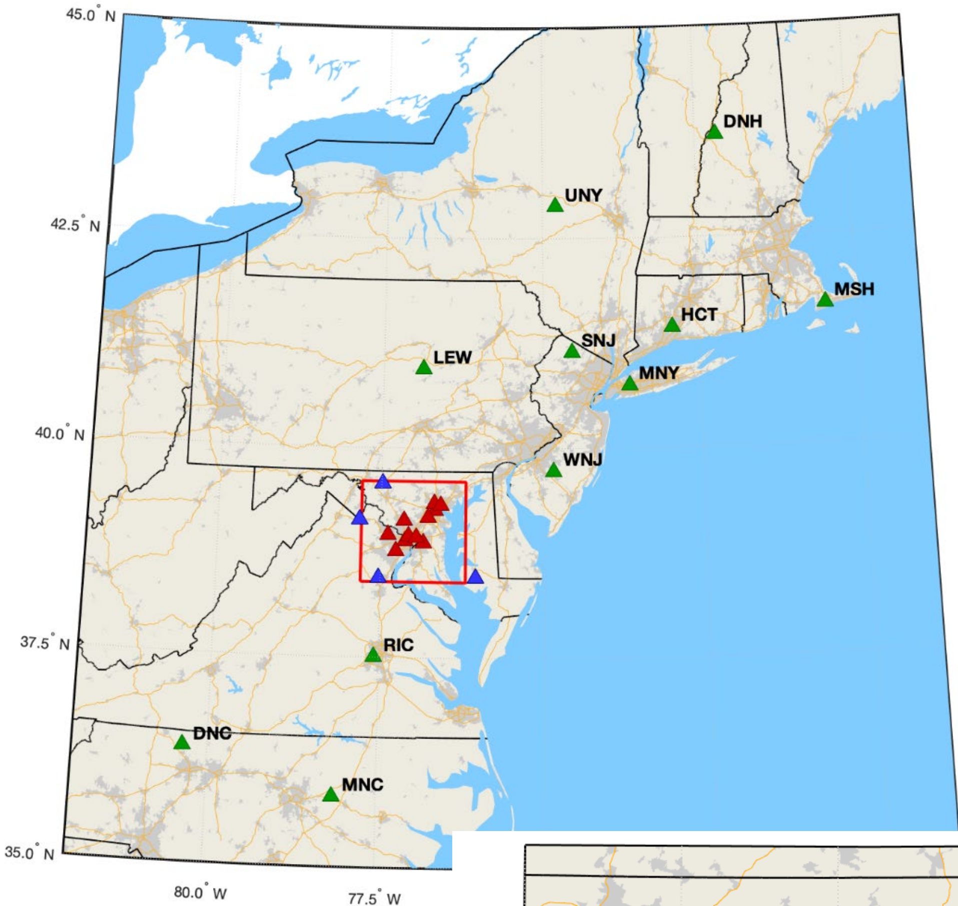
- High Resolution LIDAR – 3D winds and turbulence, planetary boundary layer (PBL) depth and information on the dynamics of the transition layer to the lower troposphere,
- Aerosol LIDAR – the transportability of these systems enables measurements both at single locations and by performing transects of specific locations for boundary layer depth and variability across selected regions, and
- Thermodynamic profiling – measure the vertical extent of temperature and moisture in PBL.

- Differential Absorption LIDAR (DIAL) and similar remote sensing approaches for CO_2, and perhaps methane, concentration profiling both vertically and horizontally,
- Aircraft cross sections – GHG vertical cross sections, both GHG concentration and meteorological parameters, and

The NEC project includes multiple measurement and analysis components. The backbone of the NEC project is a network of in-situ tower-based CO_2 and CH_4 observation stations with continuous high-accuracy mole fraction measurements of these two greenhouse gases. In addition, the University of Maryland, Stony Brook University, and Purdue University conduct multiple-week flight campaigns periodically each year.

Before the design and completion of the monitoring network around the Washington DC/Baltimore area, there were already some activities to monitor greenhouse gases in this area. Researchers at the University of Maryland had started testing the accuracy and stability of CO_2 measurements with non-dispersive infrared (NDIR) sensors and compared their performance to CRDS instruments.[230] It was demonstrated that such sensors could be useful for monitoring GHG emissions in wide ranging areas with proper calibration and corrections for environmental variables. Such inexpensive sensors of CO_2, CH_4 as well as air quality sensors could be incorporated into the network and provide complementary information. This group, in collaboration with NIST staff, also evaluated the predictability of simulated trace gas mole fractions in urban observing systems and investigated sources of variability and error in such systems.[231] Work on lower-cost CO_2 sensors has continued at NIST, with recent efforts focusing on developing a network and data infrastructure that can be used for any distributed sensor network. Such infrastructure would support a network of about 50 GHG measurement stations in the NEC and provide the ability for upgrades as sensor technology evolves and other measurements are needed. Work is also continuing on determining the best calibration strategy for these types of sensors given their susceptibility to meteorological variability and temporal drift. Multiple laboratory and field-based experiments have been conducted to understand how these sensors respond to varying meteorological conditions, and a preliminary correction technique has been established to correct measurements using lab-based algorithms and in-field reference standards.[232] Plans are in place to deploy 3 prototype GHG stations in the NEC in late 2023.

The design of a network system to monitor GHG concentrations and inferring GHG fluxes using inversion methods and atmospheric transport models is a complex problem. Best locations for collecting observations for constraining GHG emissions may be inaccessible or limited by economic and other non-scientific factors. Approaches have been developed for designing optimal GHG observing networks that consider performance and cost.[233] This was an especially difficult challenge for the NEC Testbed because of the extent of the area to be covered. Several studies were undertaken to design a tower based GHG measurement network for the NIST Northeast Corridor Testbed. An advanced Weather Research and Forecasting (WRF) model with the Stochastic Time-Inverted Lagrangian Transport (STILT) model were used to derive the sensitivity of hypothetical observations to surface GHG emissions (footprints).[229] An



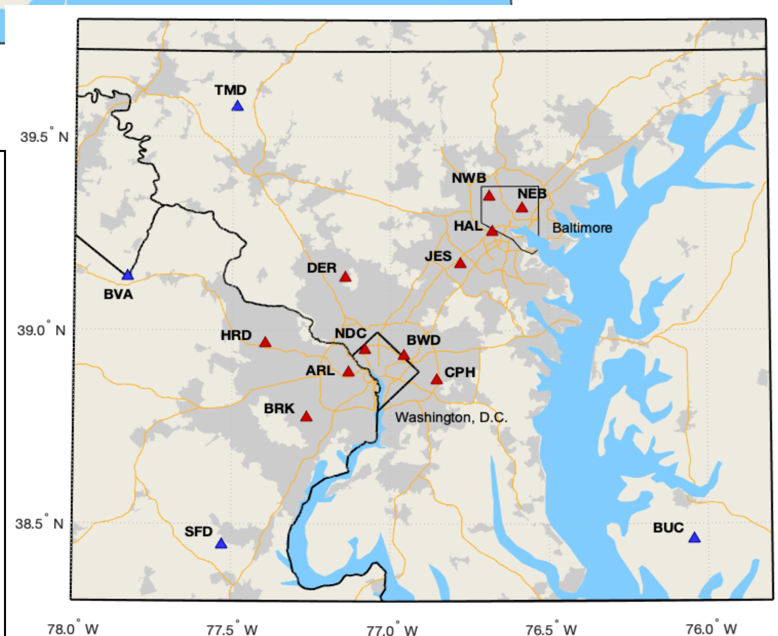


Figure 22. Tower sites sponsored by NIST, with continuous CO_2 and CH_4 observations publicly available. Top: larger regional network (site BRI in Bremen, Indiana excluded for clarity). Right: urban network located in Washington, DC/ Baltimore, MD. Blue triangles indicate background stations. Gray shading indicates urban areas. doi.org/10.5194/essd-12-699-2020

iterative selection algorithm was applied to minimize the similarities between the temporal response of each site and maximize sensitivity to the urban emissions contribution. Once a network was selected, a synthetic inversion Bayesian Kalman filter was used to evaluate observing system performance. Performance of various measurement networks configurations consisting of different number of towers and tower locations was then presented.

A follow-up study used a coupled Bayesian Information Criteria and a geostatistical regression approach to help site four background locations that best explain CO_2 variability due to extra-urban fluxes modeled at 12 urban towers.[234] This synthetic experiment used an atmospheric transport and dispersion model coupled with different flux inventories to create modeled observations and evaluated 15 candidate towers located along the urban domain for February and July 2013. The analysis showed that, during the summer months, the incoming air dominates the variability of synthetic enhancements at the urban towers. These results emphasized the importance of accurately accounting for background emissions.

While the design of the monitoring network was under way, advantage was taken of the availability of research aircraft from Purdue University and University of Maryland to conduct mass balance experiments, as part of the Wintertime Investigation of Transport, Emissions, and Reactivity (WINTER) campaign, to monitor NO_x, CO, and CO_2 emissions in the Washington, DC, and Baltimore area.[235] Results of these experiments were compared with EPA's National Emissions Inventory for 2011 and 2014.

Another aircraft campaign was undertaken as part of the University of Maryland Fluxes of Atmospheric Greenhouse Gases in Maryland (FLAGG-MD) program during winters of 2015 and 2016, again utilizing both Purdue and UMD aircrafts.[236] A typical flight experiment consists of both upwind and downwind transects at different altitudes to capture the urban enhancements of greenhouse gases; in this case the focus was on measurement of methane enhancements. Urban CO_2 [237] and CH_4 emissions [236] from the area were estimated from data collected by both aircraft using the mass balance approach. Back trajectories were used to illustrate the transport history associated with the sample air parcels and to identify CH_4 point sources such as landfills and power plants that influence the background; back trajectories were calculated using NOAA's HYSPLIT model. These results were compared with bottom-up estimates of CH_4 and CO_2 emissions.

Such measurements of fugitive methane emissions are of interest along the entire Northeast Corridor as well as other parts of the country. There have been several other studies to quantify the CH_4 emissions from urban centers using aircraft observations.[118] More detailed measurements of methane emissions have been performed in the Baltimore/Washington, DC area, using atmospheric observations from four tower stations and a Lagrangian particle dispersion model to simulate CH_4 concentrations at these stations. Observations spanning four seasons with an ensemble approach were used to consider multiple meteorological representations, emission inventories, and upwind CH_4 values to estimate localized emission rates.[238] Seasonal variabilities indicate about 40% lower methane usage during summer months.

Aircraft data have also been used to estimate urban emissions using an inverse modelling technique. [116] Previous aircraft work has indicated large daily variability in the results. In this study, measurements of CO_2, CH_4, and CO from aircraft over 5 days were used with an inverse model to estimate emissions from the Washington, DC/Baltimore region. Results showed good agreement with previous estimates in the area for all three gases. However, aliasing caused by irregular spatiotemporal sampling of emissions was shown to significantly impact both the emissions estimates and their variability. Extensive sensitivity tests allowed quantification of the contributions of different sources of variability and indicated that daily variability in posterior emission estimates was larger than the uncertainty attributed to the method itself (i.e., 17% for CO_2, 24% for CH_4, and 13% for CO). Analysis of hourly reported emissions from power plants and traffic counts showed that 97% of the daily variability in posterior emissions estimates was explained by accounting for the sampling in time and space of sources that have large hourly variability and, thus, caution must be taken in properly interpreting variability that is caused by irregular spatiotemporal sampling conditions.

Figure 23. University of Maryland experimental Cessna 402B research aircraft used for airborne mass balance experiments. www2.atmos.umd.edu/~flaggmd/

Accurate simulation of PBL height is key to greenhouse gas emission estimation, air quality prediction, and weather forecasting. A study was initiated to assess the performance of several Weather Research and Forecasting (WRF) Model configurations in which observations from ceilometers, surface stations, and a flux tower were used to study their ability to reproduce the PBL height and the impact that the urban heat island (UHI) has on the modeled PBL heights in the greater Washington, DC area.[239] In addition, CO_2 measurements at two urban towers were compared with tracer transport simulations. The ensemble of models used four PBL parameterizations, two sources of initial and boundary conditions, and one configuration including the building energy parameterization urban canopy model. Results have shown low biases over the whole domain and period for wind speed, wind direction, and temperature, with no drastic differences between meteorological drivers. It was found that PBL height errors are mostly positively correlated with sensible heat flux errors and that modeled positive UHI intensities are associated with deeper modeled PBLs over the urban areas.

HESTIA Model for the NEC Region

Subsequent efforts for estimating GHG emissions from the Washington, DC/Baltimore were facilitated with the arrival of higher resolution **HESTIA** fossil fuel CO_2 emissions model for the Baltimore area, as shown in Figure 24.[132] This model provided gridded annual and hourly emissions estimates for 2010 through 2015. These results were also compared with the overlapping Scope 1 $FFCO_2$ emissions in Baltimore's self-reported inventory for 2014.

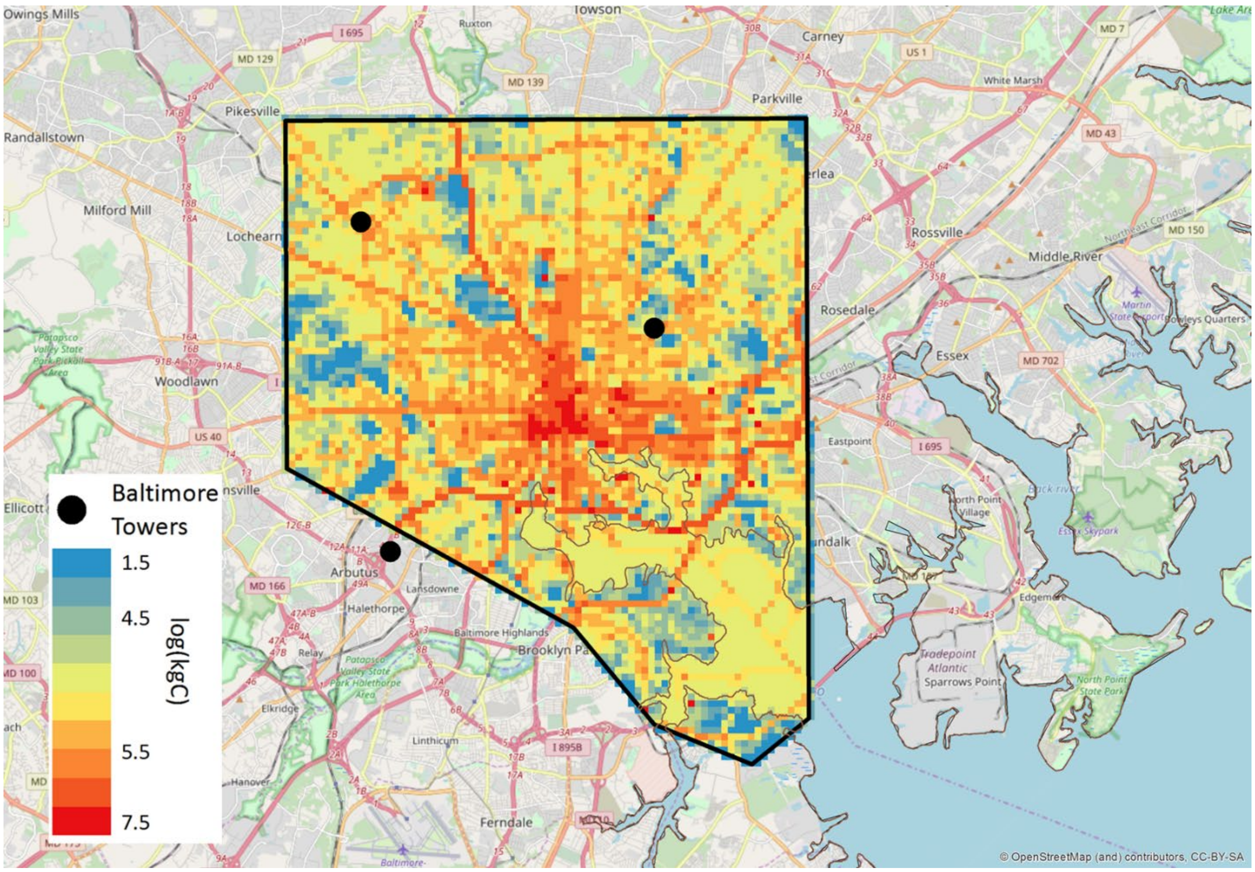


Figure 24 - HESTIA fossil fuel CO_2 emissions for Baltimore, MD. (Courtesy of Kimberly Mueller)

The first part of the Northeast Corridor tower network became fully operational in 2017, with the implementation of most of the surface network providing background and urban area measurements. This network included a high-density of observation stations around Washington, DC and Baltimore, MD. Additional stations were located throughout the northeastern U.S., established to complement other existing urban and regional networks toto begin a full network investigate emissions throughout this complex region with its high population density and multiple metropolitan areas. This is part of NIST's NEC strategy to instrument the entire NEC with its four major metropolitan areas including multiple current satellite overpass tracks. The NEC monitoring network was described in detail in a recent publication. [240]

Atmospheric Inversion Modeling for NEC

The NEC project includes an extensive modeling component. Atmospheric transport is simulated using high-resolution Weather Research and Forecast (WRF) model, with its output coupled to Lagrangian dispersion models such as HYSPLIT/STILT. These transport and dispersion models are used to interpret observations from both aircraft and tower stations and are used in atmospheric inverse analyses to estimate fluxes and their locations in urban areas as well as the larger region covering the entire US Northeast and its upwind influences. A biosphere model is also being developed for the project as well as the high-resolution fossil-fuel CO_2 inventory (**HESTIA**).

NIST partnered with Earth Networks, Inc. to deploy tower-based CO_2 and CH_4 measurements in the NEC. Initial efforts have been focused on a dense network around the Washington, DC and Baltimore, MD region, beginning in October 2015. (See Table 5) Additional stations are being established in neighboring states, with plans to expand to other cities. Tower-based continuous observations of CO_2 and CH_4 are calibrated to the WMO concentration scales and reported with time-varying uncertainties, calculated as documented in [208] and [240]. Whole air samples are collected in flasks at a subset of tower locations (MSH, NEB, NWB, BWD, and TMD) for analysis by NOAA/GML for hydrocarbon, halocarbon, and GHG species, as well as $^{13}CO2$ and radiocarbon.

Existence of other large urban areas near Washington, DC and Baltimore, MD adds complexity to determination of GHG enhancements from the local areas of interest. Hence, to attribute enhancements to a particular region, one must isolate the component of the observed concentration attributable to fluxes inside the region by removing the background, the component due to fluxes contained in the incoming atmosphere. In a recent study, methods were demonstrated to construct several versions of a background for the NIST carbon dioxide and methane observing network in the Washington, DC, and Baltimore, MD, metropolitan region. [115] Some of these versions relied on transport and flux models, while others were based on observations upwind of the domain. First, backgrounds were evaluated in a synthetic data framework, and then evaluated against real observations from the NIST urban network. It was found that backgrounds based on upwind observations capture the variability better than model-based backgrounds, although care must be taken to avoid bias from biogenic carbon dioxide fluxes near background stations in summer. Model-based backgrounds also performed well when upwind fluxes could be modeled accurately. This study evaluated different background methods and provided guidance in determining background methodology that can impact the design of urban monitoring networks.

Table 5. Atmospheric monitoring stations along the Northeast Corridor. Stations existing prior to the establishment of the NIST NEC Project are noted by *; data obtained prior to 1/2015 were not part of the NIST NEC Project. doi.org/10.5194/essd-12-699-2020

ID	Site Code	Place Name	Longitude	Latitude	Elevation (m)	Inlets
AWSHQ	AWS	Germantown, MD	-77.2631	39.20056	161	35m (roof)
GHG01	BUC	Bucktown, MD	-76.043	38.4597	3	46m, 75m
GHG08	BRI	Bremen, IN	-86.193501	41.457909	252	50m, 100m
GHG12	DNC	Danbury, NC*	-80.368892	36.376861	703	50m, 100m
GHG15	MNC	Middlesex, NC*	-78.145329	35.831252	74	50m, 243m
GHG18	RIC	Richmond, VA*	-77.5761	37.50878	104	50m, 95m
GHG19	SNJ	Stockholm, NJ*	-74.5387	41.14356	406	42m, 53m
GHG21	HCT	Hamden, CT*	-72.945224	41.433705	204	55m, 115m
GHG25	LEW	Lewisburg, PA*	-76.8789	40.94465	166	50m, 95m
GHG35	DNH	Durham, NH*	-72.154076	43.7088876	559	61m, 121m
GHG38	UNY	Utica, NY*	-74.785196	42.878973	483	60m, 68m
GHG47	MNY	Mineola, NY*	-73.6384	40.74951	34	50m, 62m
GHG48	HAL	Halethorpe, MD	-76.675278	39.255194	70	29m, 58m
GHG54	MSH	Mashpee, MA*	-70.4975	41.65669	32	25m, 46m
GHG55	ARL	Arlington, VA	-77.131667	38.891667	111	50m, 66m
GHG56	NDC	NW Washington, DC	-77.079639	38.949944	128	45m, 91m
GHG58	NWB	NW Baltimore, MD	-76.685071	39.344541	135	27m, 55m
GHG59	NEB	NE Baltimore, MD	-76.583000	39.315417	44	50m, 67m
GHG60	JES	Jessup, MD	-76.776500	39.172300	67	49m, 91m
GHG61	TMD	Thurmont, MD	-77.488056	39.576778	561	49m, 113m
GHG62	BVA	Bluemont, VA	-77.832600	39.137900	486	50m, 111m
GHG63	DER	Derwood, MD	-77.141900	39.134700	125	35m, 54m
GHG64	BWD	Brentwood, MD	-76.95555	38.934261	17	33m, 51m
GHG65	SFD	Stafford, VA	-77.529970	38.445930	77	50m, 100m, 152m
GHG66	CPH	Capitol Heights, MD	-76.853692	38.870731	50	25m, 44m
GHG67	HRD	Herndon, VA	-77.393472	38.966306	120	27m, 38m
GHG68	BRK	Burke, VA	-77.263107	38.774243	111	24m, 40m
GHG69	WNJ	Waterford Works, NJ	-74.844092	39.728802	33	43m, 98m, 151m, 201m, 241m
GHG73	NWK	Newark, NJ	-74.165613	40.734330	5	143m 130m

		SE Washington, DC	TBD	TBD	TBD	
		Delta, PA	TBD	TBD	TBD	
		Philadelphia, PA	TBD	TBD	TBD	
		Flax Pond, NY	TBD	TBD	TBD	
		Near Allentown, PA	TBD	TBD	TBD	

Summary of Results from the NEC Research

Airborne measurements of atmospheric CO concentration from 70 flights conducted over six years (2015–2020) were analyzed using an inverse model to quantify the CO emissions from the Washington, DC and Baltimore metropolitan areas. [241] It was found that CO emissions had been declining in the area at a rate of ≈−4.5 % per year since 2015 or ≈−3.1 % per year since 2016. It was also found that CO emissions showed a "Sunday" effect, with Sunday emissions on average being lower than for the rest of the week and that the seasonal cycle was no larger than 16 %. Results also showed that the trend derived from EPA's state level National Emissions Inventory (NEI) agreed well with the observed trend, but that NEI daytime-adjusted emissions are ≈50 % larger than the emissions estimated by the NEC observation and modeling methods. In 2020, measurements collected during the shutdown in activity related to the COVID-19 pandemic indicated a significant drop in CO emissions of 16 % relative to the expected emissions trend from the previous years, or 23 % relative to the mean of 2016 to February 2020. Results also indicated a larger reduction in April than in May. It was concluded that this reduction in CO emissions was driven mainly by a reduction in traffic.

An aircraft campaign was conducted in February 2015, as part of the Fluxes of Atmospheric Greenhouse-Gases in Maryland (FLAGG-MD) project, to study emissions of CO_2 in the Baltimore, MD-Washington, DC (Balt-Wash) area. During the campaign, elevated mole fractions of CO_2 were observed downwind of the urban center and local power plants. [237] Upwind flight data and HYSPLIT model analyses helped account for the impact of emissions outside the Balt-Wash area. The accuracy, precision, and sensitivity of CO_2 emissions estimates based on the mass balance approach were assessed for both power plants and cities. Estimates of CO_2 emissions from two local power plants agreed well with their Continuous Emissions Monitoring Systems (CEMS) records. For the 16 power plant plumes captured by the aircraft, the mean percentage difference of CO_2 emissions was −0.3%. For the Baltimore-Washington area as a whole, the 1 sigma CO_2 emission rate uncertainty for any individual aircraft-based mass balance approach experiment was ±38%. Treating the mass balance experiments, which were repeated seven times within 9 days, as individual quantifications of the Balt-Wash CO_2 emissions, the estimation uncertainty was ±16% (standard error of the mean at a confidence level of 95%). Aircraft-based estimates were compared to various bottom-up fossil fuel CO_2 ($FFCO_2$) emission inventories. Based on the FLAGG-MD aircraft observations, fossil fuel CO_2 emissions were estimated to be 1.9 ± 0.3 MtC from the Baltimore-Washington area during the month of February 2015. The mean estimate of $FFCO_2$ from the four bottom-up models was 2.2 ± 0.3 MtC.

It was mentioned earlier that the capabilities of urban greenhouse gas (GHG) measurement network were tested to detect abrupt changes in emissions, such as those caused by the roughly six week COVID-19 lockdown in March 2020 using hourly *in situ* GHG mole fraction measurements from six North American cities. [227] One of these urban areas was the Washington, DC-Baltimore, MD metropolitan area. Observed changes in CO_2, CO, and CH_4 for different mole fraction metrics (diurnal amplitude, vertical gradients, enhancements, within-hour variances, and multi-gas enhancement ratios) during 2020 were compared to previous years for three periods: pre-lockdown, lockdown, and ongoing recovery. The networks showed decreases in CO_2 and CO metrics during the lockdown period in all cities for all metrics, while changes in the CH_4 metrics were variable across cities and not statistically significant. Traffic decreases in 2020 were correlated with the changes in GHG metrics, whereas changes in meteorology and biology were not, implying that decreases in the CO_2 and CO metrics were related to reduced emissions from traffic and demonstrating the sensitivity of these tower networks to rapid changes in urban emissions. The enhancements showed signatures of the lockdowns more consistently than the three micrometeorological methods, possibly because the urban measurements are collected at relatively high altitudes to be sensitive to whole-city emissions. This suggests that urban observatories might benefit from a mixture of measurement altitudes to improve observational network sensitivity to both city-scale and more local fluxes.

Figure 25. The tower in Thurmont, MD, measuring CO_2 and CH_4 (Photo courtesy of Anna Karion)

Another study focused on the impact of COVID-19 on emissions in LA and Washington, DC/Baltimore metropolitan areas.[226] It was found that societal responses to COVID-19 had resulted in unintended reductions of city-scale carbon dioxide (CO_2) emissions. Decreases in CO_2 emissions in Los Angeles and Washington DC/Baltimore were detected during March and April 2020. Three lines of evidence were presented using methods that have increasing model dependency, including an inverse model to estimate relative emissions changes in 2020 compared to 2018 and 2019. The March decrease (25%) in Washington DC/Baltimore was largely supported by a drop in natural gas consumption associated with a warm spring whereas the decrease in April (33%) correlates with changes in gasoline fuel sales. In contrast, only a fraction of the March (17%) and April (34%) reduction in Los Angeles is explained by traffic declines. Methods and measurements used herein highlighted the advantages of atmospheric CO_2 observations for providing timely insights into rapidly changing emissions patterns that can empower cities to course-correct CO_2 reduction activities efficiently.

Another study summarized some of the recent advances that integrate bottom-up hourly, street level emission data with top-down GHG atmospheric observations that have begun to yield GHG estimates at annual and sub-annual timeframes with sufficient accuracy, which could replace the use of self-reported inventories by many cities to plan their mitigation efforts. This emerging approach that could help cities achieve their climate and sustainability goals was demonstrated for Indianapolis and the City of Baltimore. [242] More recently, a harmonized data set was presented synthesizing urban GHG observations from cities with monitoring networks across North America that will facilitate cross-city analyses and address scientific questions that are difficult to address in isolation. [243]

Presently, there is no well-established monitoring network around New York City; the NEC project currently includes one tower in NYC (Queens), one in Northern NJ, and a newly established tower observing node in the middle of Long Island. However, aircraft flights have been utilized to develop estimates of greenhouse gas emissions from the city. In a recent study, results were presented from 9 research flights measuring CO_2 and CH_4 around New York City during the nongrowing seasons of 2018–2020.[244] An ensemble of dispersion model runs in a Bayesian inverse modeling framework was used to derive campaign-average posterior CO_2 and CH_4 emission estimates for the New York–Newark, NJ, urban area of (125 ± 39) kmol CO_2 s^{-1} and (0.62 ± 0.19) kmol CH_4 s^{-1} (reported as mean ± 1σ variability across the nine flights). We also derived emission estimates of (45 ± 18) kmol CO_2 s^{-1} and (0.20 ± 0.07) kmol CH_4 s^{-1} for the five boroughs of New York City. These emission rates, among the first top-down estimates for New York City, are consistent with inventory estimates for CO_2 but are 2.4 times larger than the gridded EPA CH_4 inventory, consistent with previous work suggesting CH_4 emissions from cities throughout the northeast United States are currently underestimated. These results support findings of a previous study that used airborne measurements over several U.S. East Coast cities to also find higher CH4 emissions than reported in the EPA inventory. [118]

Another study introduced a scaling factor (SF) method that combines aircraft measurements and dispersion modeling to estimate urban emissions and apply it to nine nongrowing season research aircraft flights (same flights mentioned above) around New York City (NYC) in 2018–2020. This SF approach uses a weighting function to focus on an area of interest while still accounting for upwind emissions.[245] Carbon dioxide (CO_2) emissions from NYC and the Greater New York Area (GNA) were estimated and compared to nested inversion analyses of the same data. The average calculated CO_2 emission rates for NYC and the GNA, representative of daytime emissions for the flights, were (49 ± 16) kmol/s and (144 ± 44) kmol/s, respectively (uncertainties reported as ±1σ variability across the nine flights).[5] These

[5] 1 kmole/s of CO_2 corresponds to 44 kg/s

emissions are within ~15% of an inversion analysis and agree well with inventory estimates. By using an ensemble, the variability introduced by several sources was also investigated and it was found that day-to-day variability dominated the overall variability. This work demonstrated the capability of an SF method to quantify emissions specific to particular areas of interest.

* * * * * *

Data collected at the three NIST Testbeds is of great interest to the GHG measurement community, especially for validation of GHG inventories and assessment of mass balance and inverse models used for source apportionment. Hence, the data collected at these sites are made widely available. For the INFLUX project, there are multiple types of observational data sets being maintained and archived. Generally, information about the different data sets is kept up to date at the main INFLUX web page maintained by Pennsylvania State University (PSU), at https://sites.psu.edu/influx/. There are links to the data from the *in-situ* tower measurements by PSU, the eddy flux tower data by PSU, the aircraft measurements by Purdue University, and the measurements from whole air samples taken at the tower and from the aircraft by NOAA's Global Monitoring Laboratory (GML). Currently many of these data sets are archived at the PSU Data Commons.

For the LA Megacity Project, the *in-situ* tower observations were initially maintained and archived at NASA/JPL; they are currently being maintained in an archive by NIST at https://data.nist.gov and https://doi.org/10.18434/mds2-2388. This data is updated with new calendar year data regularly (generally annually, but sometimes less often). An update is expected to the methods for calibration of the carbon dioxide measurements by transferring to the updated WMO X2019 CO_2 scale sometime in the 2023 calendar year, and that update will have its own DOI.

The Northeast Corridor project *in-situ* tower data is being maintained by NIST similarly to the LA data, at https://data.nist.gov and the latest version is at https://doi.org/10.18434/mds2-3012 with an update to the WMO X2019 CO_2 scale sometime in the 2023 calendar year. Flask data collected at four NEC sites are available from NOAA/GML by request. Airborne data from UMD's flights related to the NEC project are maintained by UMD and available publicly at https://www2.atmos.umd.edu/~flaggmd/.

Spatially and temporally resolved emissions model data for the three testbeds (as well as Salt Lake City) are available at NIST at https://doi.org/10.18434/mds2-1902 and at the Northern Arizona University website https://hestia.rc.nau.edu/ .

Meteorological measurements for INFLUX and the NEC from the LIDAR deployed by NOAA's Chemical Sciences Laboratory (CSL) are available at https://csl.noaa.gov/projects/influx/ and https://csl.noaa.gov/groups/csl3/measurements/2021dcflux/.

5. National and International Collaborations and Program Impact Assessment

National Collaborations

These projects have been successful because of the engagement of an outstanding team of U.S. researchers to address a global challenge to characterize changes in our atmosphere and the resulting impacts on Earth's climate. Each group represented leading researchers in their fields while bringing complementary skill sets to these challenging research projects.

The teams from Purdue University (led by Prof. Paul Shepson, now Dean of the School of Marine and Atmospheric Sciences, Stony Brook University, and Prof. Kevin Gurney, now at Northern Arizona University), NOAA/University of Colorado (led by Drs. Jocelyn Turnbull and Colm Sweeney), and Pennsylvania State University (led by Prof. Kenneth Davis) were instrumental in establishment of the first U.S. Urban Testbed in Indianapolis, IN for monitoring GHG emissions. The Purdue team brought their expertise in atmospheric chemistry and unique aircraft-based atmospheric measurement capabilities to get the project to a running start. The Penn State team brought in critical observational and meteorological expertise. Along with their expertise in atmospheric transport and atmospheric inversion data analysis capabilities; they enabled accurate interpretation of data obtained from atmospheric sampling based on communication towers and resultant source apportionment. These efforts were complemented by the NOAA team who took responsibility for collecting flask samples of the atmosphere at some of the tower locations and analysis of samples for GHG isotopes. Contributions of the Northern Arizona University team (led by Prof. Kevin Gurney) in development of bottom-up estimates of GHG emissions with high spatial and temporal resolution, were critical in interpretation of atmospheric measurements. Over time, the Penn State team also took over the operation of all the INFLUX towers, with support from NIST.

The Los Angeles Megacity Project involved another team of outstanding experts. Before the beginning of the NIST Urban Testbed Project, several universities were already involved in monitoring the atmospheric composition in L.A. and its effect on climate change. These had started with the pioneering work of Prof. Charles D. Keeling in the 1950's and 1960's at the Scripps Institution of Oceanography of the University of California at San Diego. The Scripps team (led by Prof. Ralph Keeling and Prof. Ray Weiss) had continued this work into the present and became part of the LA Megacity Project. Researchers at Caltech (Prof. Sally Newman), University of California at Riverside (led by Prof. Francesca Hopkins), and NASA's Jet Propulsion Laboratory (led by Riley Duren and Charles Miller), along with participation by the California Air Resources Board, had well established credentials in monitoring atmospheric GHG emissions. NIST's involvement, along with its private sector partner Earth Networks, Inc., catalyzed the formation of a powerful team to add to existing monitoring stations in the Los Angeles area and establish a complete network of monitoring stations and acquire detailed information on GHG emissions in the Los Angeles South Coast Air Basin. Earth Networks, Inc. currently operates most of the continuous in-situ GHG monitoring stations in LA Megacity Testbed.

The Northeast Corridor Project, led by Dr. Anna Karion of NIST, represented a different challenge, given the scale of the territory to be covered. Participation of teams that had already demonstrated their skills, such as the Penn State team and the Purdue team (after Prof. Shepson's move to Stony Brook University in Long Island, NY) enabled this project to get a good start. The addition of the University of Maryland team (led by Prof. Russ Dickerson) who brought long standing experience in monitoring atmospheric pollutants with their aircraft-based instruments, that now combine GHG and air pollutant measurements, gave the team the unusual capability to perform simultaneous measurements with two different aircraft. It also enabled monitoring of emissions in areas where a tower monitoring network was not yet established, such as around New York City.

The contributions of the Northern Arizona University team, led by Prof. Kevin Gurney, were critical for the success of all three NIST Urban Testbed Projects.[6] Spatially Explicit Emission Models provided by this team for Indianapolis, Los Angeles and Washington, DC/Baltimore MD area provided the foundation for the analysis of the tower based atmospheric measurements and ensuring the resolution of an otherwise "ill defined" problem. A team from Boston University (led by Prof. Lucy Hutyra), working closely with Dr. David Allen oh NIST, brought new capabilities to improve our understanding of the role biogenic processes play in modifying GHG composition of the atmosphere and how urban processes differ from rural ones.

Equally important was the preparation of future scientists and engineers by these academic institutions who will be the future experts to address climate change related challenges. The NIST GHG Measurements Program has contributed to the development of more than 40 M.Sc. and Ph.D. level graduates of these universities.(See Appendix B) In addition, more than 30 postdoctoral fellows and research staff have been involved in these projects; several of these research scientists have moved on to other institutions and other countries to contribute to their scientific programs addressing climate change (T. Lauvaux of PSU to France, J. Pitt of Stony Brook and K. Wu of PSU to U.K., H. Lin of NIM to China, B. Nathan of PSU to New Zealand, etc.).

Outstanding contributions of NIST researchers to improve our measurement capabilities should not be forgotten. New capabilities for accurate measurement of smokestack emissions, spectroscopic reference data supporting all remote sensing measurements of greenhouse gases and isotopic measurements that facilitate identification of emission sources, long path measurements in the atmosphere, using dual-comb spectroscopy, improved Differential Absorption LIDAR (DIAL) technology, and improved gas mixture standards, the foundation of all GHG concentration measurements, all help improve the accuracy and reliability of GHG measurements. NIST researchers have also helped improve the atmospheric transport models and solution methods of inversion analyses, all critical for interpretation of atmospheric GHG measurements particularly important for source apportionment applications.

Program Impact

This has been a very productive effort, with a large number of publications (more than 200) over the last decade. NIST internal research efforts alone have resulted in more than 50 archival publications. The importance of these publications has been recognized by the research community and they have been heavily cited by other researchers with more than 5,000 citations, which provides an indication of the

[6] Prof. Gurney was one of the initiators of the INFLUX project while at Purdue University in the Department of Earth, Atmosphere and Planetary Science. He then moved his group to Arizona State University; they are now at Northern Arizona University.

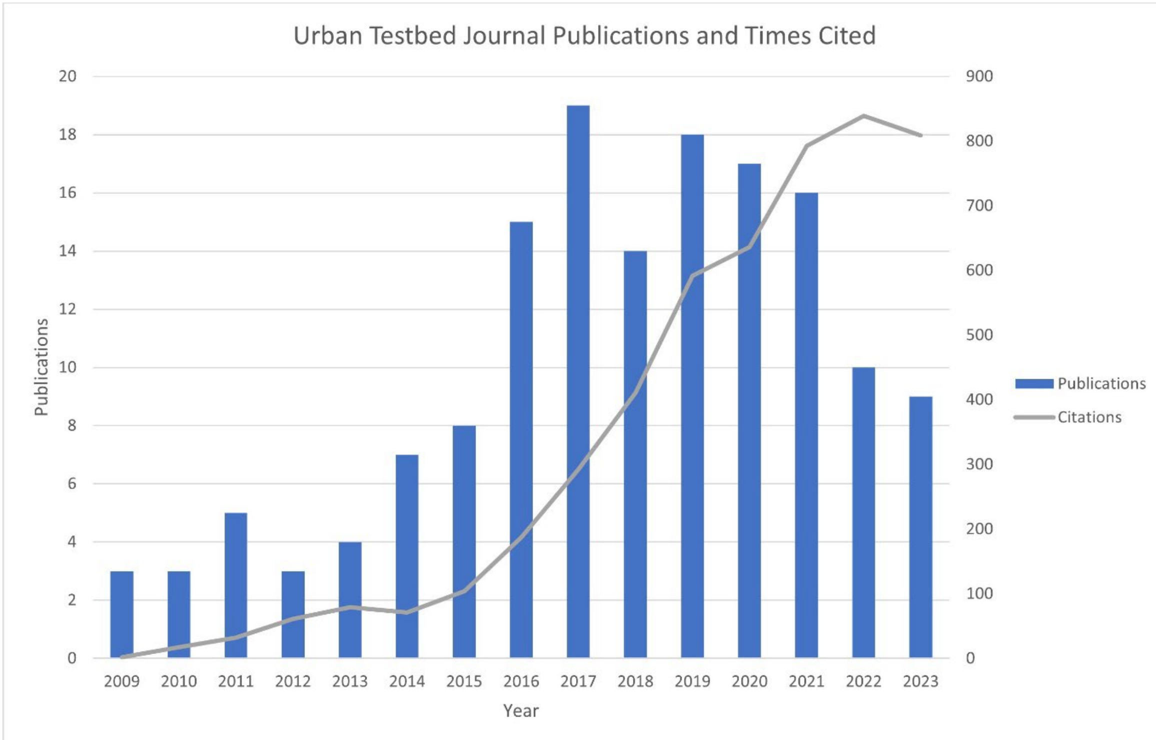


Figure 26. History of publications from the NIST Urban Testbeds Program over the last decade and their citations.

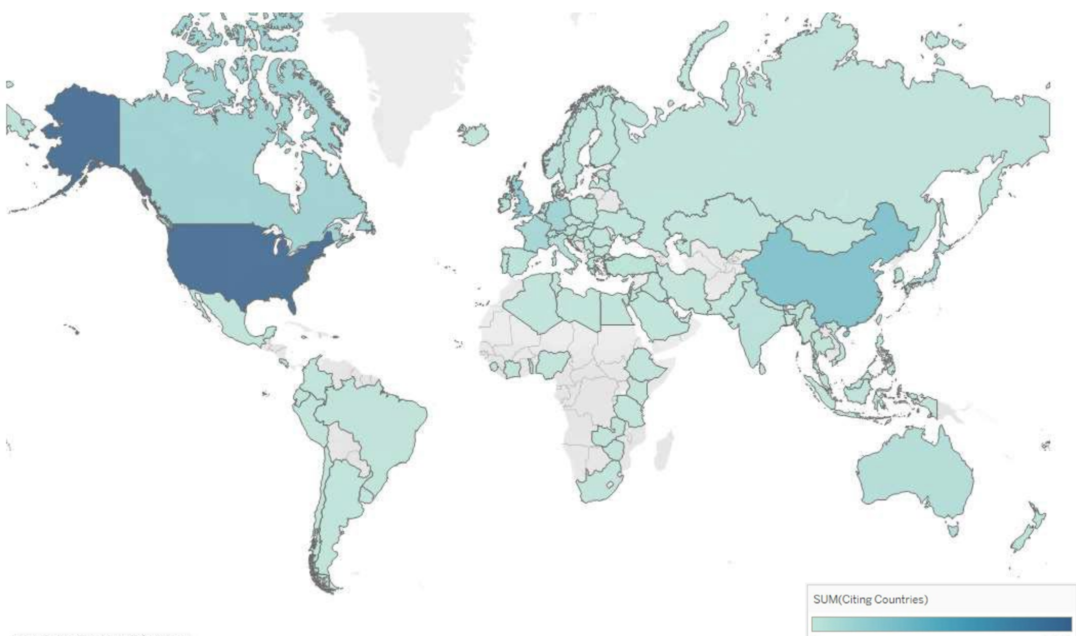


Figure 27. Geographic citation maps showing the country of origin for the citing papers (Source: Clarivate Web of Science)

impact of the NIST Greenhouse Gas Measurements Program. (See Figure 26)[246] It should also be noted that publications resulting from the NIST Urban Testbeds Program have been widely cited by researchers in the U.S. (1643 citations) and in more than 90 countries, primarily in developed countries such as China (539 citations), UK (327), Germany (275), Canada (269), France (227), Netherlands (186), Japan (135), Italy (118), Australia (111), Switzerland (95), etc., but also in developing countries indicated by the lightly shaded areas in Figure 27, again demonstrating the global impact of this program.

<u>Federal Collaborations</u>

NIST also established excellent collaborations with other Federal Agencies such as NOAA's Global Monitoring and Chemical Sciences and Air Resources Laboratories, NASA's Jet Propulsion Laboratory, and the U.S. Environmental Protection Agency. Each of these agencies brought unique capabilities to address the global challenges we all face and to provide improved accuracy and reliability for the information acquired by these testbeds and the impact of this research on national and regional GHG emission inventories.

Figure 28. NOAA and NIST installed a Doppler LIDAR instrument on top of the Department of Commerce's Herbert Clark Hoover Building in Washington, D.C. on April 16, 2021, for a research project studying urban greenhouse gas emissions.

An outstanding illustration of the collaboration between NIST and NOAA's Chemical Sciences and Air Resources Laboratories was the installation of a Doppler LIDAR instrument on top of the Department of Commerce's Herbert Clark Hoover Building in Washington, D.C., in addition to an existing weather station, to measure wind flow and turbulence in the lowest part of the atmosphere as part of a research project to study greenhouse gas emissions in the Capitol area. (See Figure 28) LIDAR, which stands for Light Detection and Ranging, is a remote sensing method that uses a pulsed laser light that is reflected by atmospheric particulates to measure atmosphere transport, similar to how radar uses radio waves to gather information on thunderstorms. Scanning Doppler LIDARs can make high-resolution measurements of horizontal and vertical winds, and resolve turbulent processes in the atmosphere to generate data required by sophisticated atmospheric transport models.

The Doppler LIDAR, operated by NOAA, was added to an existing weather station called DCNet that was built by NOAA after September 11, 2001, to provide a highly localized ability to track the dispersion of pollutants or hazardous substances in the urban landscape. Installation of the LIDAR was completed on April 16, 2022, and the DCNet instrumentation was upgraded in 2023.

The D.C. installation will contribute data to NIST's Northeast Corridor Testbed Project, which measures anthropogenic greenhouse gas emissions and other pollutants in urban areas along the U.S. East Coast. The Air Resources Laboratory also uses the DCNet data to help the intelligence community understand how potentially hazardous gases and/or particles could be dispersed into urban areas.

NOAA's Chemical Science Laboratory (NOAA CSL), NIST's Greenhouse Gas Measurement Program (NIST GHG) and Statistical Engineering Division have initiated a new partnership. Known as the **GReenhouse gas And Air Pollutants Emissions System** (**GRAAPES**) project, the objective is to jointly develop and maintain a new capability for the Department of Commerce, to measure, model, and map emissions of GHGs and air pollutants across the U.S., and to qualify these products with evaluations of their associated uncertainties. Additional contributors to the research are anticipated and will be a welcome addition.

This new capability will utilize publicly accessible greenhouse gas emissions data that are in scope, from activities in multiple economic sectors, including energy production, manufacturing, transportation, and agriculture and land use, among others. This tool will rely on metrics of consumption and utilization in each sector, and on their current emission factors designated by the EPA or other authoritative source to convert such metrics into emission amounts. This corresponds to the so-called bottom-up estimation of emissions. At present, the estimation of emissions of GHGs and of air pollutants are largely independent exercises. However, their inter-dependence, especially at urban scales, is increasingly gaining attention because the reduction of GHGs in the atmosphere brings about synergistic benefits: improving air quality (AQ) and mitigating climate impacts. A reliable bottom-up estimation of emissions data of GHGs and of air pollutants will serve as a valuable complement to top-down estimates derived from the atmospheric observations (made using fixed and mobile land-based or ocean-based sensors, as well as aircraft and satellites) that are used in conjunction with atmospheric transport models.

This information will have a low latency and be publicly available at disaggregate spatial and temporal resolutions. Since bottom-up and top-down estimates ideally should be mutually consistent within their respective margins of uncertainty, they constrain each other and support a consensus assessment of the state of the atmosphere. Taken together, the top-down and bottom-up estimates of GHG emissions will improve on what either approach can deliver on its own.

International Collaborations

NIST's GHG Measurements Program and the Urban Testbeds Projects have promoted many international collaborations and resulted in expansion of international activities to monitor GHG emissions in urban environments, improved quantification of GHG fluxes and national and regional GHG emission inventories. NIST helped organize a WMO-BIPM Workshop to bring together the metrology and meteorology communities to address ***Measurement Challenges for Global Observation systems for Climate Change Monitoring: Traceability, Stability and Uncertainty***, on March 30-April 1, 2010, in Geneva, Switzerland.[24] There were two subsequent workshops in 2015 and 2022 to continue these efforts and establish measurement standards that are accurate, reliable, and internationally recognized. Recommendations for improving measurement capabilities are forthcoming.[29] [35]

A workshop was held as part of the Inter-American Metrology System (SIM) General Assembly at Queretaro, Mexico in October 2013 to increase awareness of climate change issues among South and Central American countries. The Workshop provided an excellent platform to identify measurement and standards needs of member countries, and to develop an ***Action Plan for the Americas*** for training and other capacity building activities. These activities attracted the attention of the U.S. State Department and the Organization of American States (OAS). A proposal was approved to engage all members of SIM (34 countries of the Americas) in a discussion of ***Renewable Energy and Climate Science: Metrology and Technology Challenges***. Efforts were made to include speakers from all leading NMIs from the Americas (Brazil, Canada, Mexico, and USA) as well as PTB (Germany). Subsequent workshops were held in Guatemala (May 2014), Uruguay (September 2014), Colombia (November 2014), and Jamaica (April 2015). Two subsequent training workshops were held, one at CENAM, Mexico on Measurements of Gas Concentration for Monitoring Air Quality and Climate Change (September 2015) and another at NIST on Building Energy Conservation.

NIM (National Institute of Metrology - China) staff were interested in developing a program on GHGs as well as aerosol measurements, since it represents an important area of concern for China. NIST invited NIM staff to participate in a workshop held at Caltech in 2012 and arranged for the NIM staff to meet with a Caltech faculty member who is a world recognized expert on aerosol measurements. NIM staff expressed interest in planning a workshop in early September 2012 on GHGs and Aerosol Measurements at NIM; detailed information was provided to NIM about potential participants (from the USA as well as from within China) to assist them with organization of the workshop. These efforts resulted in a very successful workshop, which was held during the first week of September 2012 in Beijing. Representatives from more than 15 organizations from within China participated in this Workshop, including Prof. Zhou Linxi from the Chinese Academy of Meteorological Sciences, who is China's representative to the UN IPCC. These efforts eventually resulted in development of an urban GHG and air quality monitoring network in the city of Zhengzhou, a major industrial city in central China. NIST provided training for several Chinese scientists in development of experimental and computational tools. [247] Another similar effort is now being developed in Beijing, China. NIST also assisted in development of a facility at NIM for flow rate measurements of smokestack GHG emissions, similar to the one at NIST. NIM has established the Zhengzhou Institute of Metrology (ZIM) in collaboration with the Zhengzhou city; this is a new division of NIM China, and its first director partially completed his post-doctoral work at NIST.

NIST has also established close working relationships with countries who have developed advanced capabilities for monitoring urban emissions. Researchers in France were one of the first to start monitoring GHG emissions in urban environments, specifically around the Paris environs. NIST has established collaborations with researchers at **LSCE** (Laboratoire des Sciences du Climat et de l'Environnement) who are some of the world's leading experts,[167] [248] [249] [250] [251] as well as private sector researchers at NEXQT, who have developed a **City Climate Intelligence** platform for urban stakeholders,[119] and are developing advances in emissions modeling. In the U.K., NIST has close working relationships with **NPL** (National Physical Laboratory) and researchers at the **UK Met Office**, who have established tower networks to monitor GHG emissions. [252] [253] [254] [255] [256] In Canada, **Environment and Climate Change Canada** has started efforts to monitor GHG emissions in Toronto and other cities.[257, 258] Researchers in New Zealand have developed unique capabilities for measurements of GHG isotopes, particularly radiocarbon, which are invaluable to differentiate fossil fuel emissions from biogenic emissions.[259, 260] In Europe, the **ICOS** (Integrated Carbon Observation System) Program has established a network which includes stations in 14 European countries. Each of the current 39 Atmospheric observing stations measures greenhouse gas concentrations (such as carbon dioxide and methane) in the atmosphere, as well as meteorological parameters. ICOS is now expanding its work to monitor GHG emissions in urban areas of Europe and utilizing information gained from U.S. urban testbeds as a model for its activities.

NIST, working with NOAA and NASA, also catalyzed the formation of **IG^{3}IS** (Integrated Global Greenhouse Gas Information System) at WMO that has increased the visibility of the community within WMO and the UNFCCC's IPCC. IG^{3}IS efforts have been instrumental in development of the most recent "refinement" of 2006 IPCC Guidelines to include atmospheric measurements to supplement emission inventory estimates. NIST also promoted the development of the IG^{3}IS **Urban Greenhouse Gas Emission Observation and Monitoring Best Research Practices**.[261] These practices will form the foundation of new efforts, promoted by NIST, to establish the first documentary technical standards developed to support existing ISO (International Standards Organization) management standards (ISO 14064 and 14065) for measurement of GHGs in the atmosphere.

6. Conclusion

The NIST Greenhouse Gas Measurements Program has had many critical accomplishments over the last decade. NIST capabilities in spectroscopic measurements of GHGs have improved drastically, enhancing the accuracy, precision, and reliability of these measurements; improved theoretical capabilities to predict spectral line shapes and intensities have complemented the experimental efforts. This reference data is fundamental to GHG concentration measurements made by spectroscopic remote sensing methods and has materially advanced the accuracy of satellite measurements of atmospheric CO_2 concentrations. Development of spectroscopic methods for measurement of carbon and oxygen isotopes have added new practical capabilities that have the potential to replace time consuming accelerator mass spectrometry, primarily of radiocarbon. Significant improvements have also been made in determining absorption and scattering characteristics of carbonaceous aerosols, improving our ability to predict their impact on climate change.

A new NIST capability, the Smokestack Simulator has enabled development of techniques for measurement of GHGs emitted from smokestacks of power plants and large industrial facilities with significantly improved accuracy (with less than 2% uncertainty) and speed. These capabilities were demonstrated in field tests in collaboration with the Electric Power Research Institute. A new NIST designed Non-Nulling probe, developed for this purpose, was patented and is being commercialized by the private sector. A new ASME standard is being developed for broad implementation of this technology.

The capability to monitor GHG emissions over a large area was demonstrated using Dual-Comb Spectroscopy and DIAL LIDAR, as well as laser imaging tomography, that add to our tool kit to identify local emissions by point sources or leaks from natural gas distribution systems.

With the expected increasing demand for calibration standards for GHG measurements, as global efforts expand to monitor GHGs especially in urban environments, a new effort has been initiated, in collaboration with BIPM, WMO CCLs (particularly NOAA GML) and National Metrology Institutes (NMIs), to establish a framework where NMIs and WMO CCLs maintain national and regional scales, participate in key comparison programs to establish, and monitor consistency of these scales. Such a system would overcome the possibility of a single point of failure and meet the need for increasing requirements for calibration standards.

There have also been significant improvements in computational tools that are a critical part of efforts to monitor atmospheric GHG emissions and for source attribution. Emission models that are used to predict emissions from urban environments have improved their spatial and temporal resolution and have found broad application in interpretation of GHG measurements in NIST's Urban Testbeds. Recently there have been efforts to commercialize some of these modeling capabilities for use of local governments. These inventories are commonly used as "priors" in inverse modeling approaches. Their improved spatial and temporal resolutions make them quite useful for urban planners involved in mitigation actions. They have also demonstrated some of the shortcomings of Self-Reported Inventories (SRIs) that are used by cities to plan their mitigation efforts.

Significant improvements have also been made in our understanding of biogenic fluxes and their impact in monitoring urban GHG emissions. A new testbed established at NIST's Gaithersburg campus, the Forested Optical Reference for Evaluating Sensor Technology (FOREST), is being used to investigate ecosystem phenomenology for a better understanding of biogenic carbon fluxes.

Finally, there have been significant advances made in techniques for GHG attribution in urban environments from atmospheric measurements, including mass balance and inversion methods. These techniques are being used for interpretation of aircraft-based as well as tower-based measurements to predict the distribution and intensity of GHG sources in a city and its surrounding areas. They have been applied to monitoring CO_2, CH_4, and other GHGs.

These improvements have been critical in our capability to monitor and interpret GHG emissions in urban environments. These capabilities have been tested in the three NIST Urban Testbeds established in Indianapolis, Los Angeles Megacity, and the Northeast Corridor. The network of monitoring stations in each testbed have provided valuable information on the intensity and distribution of emission sources, and determination of GHG fluxes.

Clearly there have been significant advances made to characterize GHG emission fluxes from urban environments, which account for more than 70% of global GHG emissions. Emerging estimation approaches are expected to help cities achieve their climate and sustainability goals. [242] Other studies have demonstrated the importance of accurate GHG emission inventories for cities, which are shown to be underestimated in many cases. [262] Studies have demonstrated good agreement between bottom-up and top-down methods, which is quite encouraging. [190]

Quantification of GHG emissions from urban environments provides a valuable tool to validate self-reported inventories of cities, point out deficiencies, identify the most beneficial mitigation policies and goals, and track their progress. These are also useful tools for assessing the impact of mitigation efforts that encourage such undertakings. Such tools have also encouraged the undertaking of similar efforts around the globe and development of "best practices" to monitor GHG emissions from urban environments.[261]

As noted throughout this report, accurate GHG emissions quantification is fundamental to targeting mitigation actions and assessing their impacts. Financial markets are critical enablers of these actions as implementation of these actions will mostly be done by private sector institutions involved in financial transactions. We see that voluntary carbon markets (VCMs) are growing rapidly, have been recognized by the recent COP 28,[263] and are becoming the focus of regulatory activities. For example, the U.S. Commodity Futures Trading Commission (CFTC) has issued proposed guidance regarding the listing of voluntary carbon credit derivative contracts. [264] This is the first guidance specifically targeting VCMs by a federal regulator. These concerns are driven by carbon offsetting that are denominated as carbon credits, 1 ton of CO_2 equivalent. Questions concerning the authenticity and accuracy of carbon offset credits are widespread.[265] Essentially, is "a ton a ton?" and the correct GHG species? As these markets continue their rapid growth, there are contentions that the current approximately $2 billion VCM offset credits are not accounting for the emissions reductions promised, thereby reducing market confidence. Although not the sole source of confusion, poor credit quantification represents a fundamental source of such assertions and can be expected to impede market growth and thereby, climate actions. The measurement capabilities developed and demonstrated by NIST's GHG Measurements Program are an important step in laying the foundation for improved confidence in credits, particularly those associated with urban settings. One may view these developments as being like those underpinning the U.S. weights and measures system, which gives the everyday consumer confidence that the commodities they buy and use in everyday life are the quantities stated. To make significant progress toward the mitigation efforts needed to attain the UNFCCC's goal of limiting global warming to 1.5° C, the financial structures supporting carbon credits must aim for a similar level of confidence and operational capabilities. NIST can play a similar role in these efforts as it does in the U.S. weights and measures system, that of providing measurement technologies and standards supporting quantification.

NIST research on Greenhouse Gas Measurements and development of Urban Testbeds have had a huge impact in the U.S. and globally on monitoring of urban GHG emissions, improving their accuracy and reliability, and encouraging mitigation efforts, especially by the private sector. NIST efforts have catalyzed global activities in many countries to address climate change, reduce the impact of anthropogenic emissions on our atmosphere, and minimize environmental impacts of climate change.

A recent National Academy report on ***Greenhouse Gas Emissions Information for Decision Making*** has emphasized the importance of usability and timelines, information transparency, evaluation and validation of GHG emission information, completeness and inclusivity of the information, and effective communication of this information to the user community.[266] NIST programs have already gone a long way to meet these needs.

These efforts have been recognized by the Department of Commerce and the White House Offices of Science and Technology Policy (OSTP) and Management and Budget (OMB). The newly developed ***National Strategy to Advance an Integrated U.S. Greenhouse Gas Measurement, Monitoring, and Information System*, [267]** prepared by the Greenhouse Gas Monitoring & Measurement Interagency Working Group (OSTP), includes many of the efforts already under way at NIST. NIST has made critical contributions to the development of this Strategic Plan which is expected to be implemented as an operational system for both the U.S. government and non-governmental entities.

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Appendix A

Acronyms

ABL	Atmospheric Boundary Layer
ACES	Anthropogenic Carbon Emissions System (Boston University)
AQ	Air Quality
ASCENDS	Active Sensing of CO2 Emissions over Nights, Days, & Seasons Program (NASA)
BIPM	International Bureau of Weights and Measures (in French)
C40	Cities Climate Leadership
CAP	Climate Action Plan
CARB	California Air Resources Board
CarbonSat	Carbon Monitoring Satellite (European Space Agency)
CCL	Central Calibration Laboratories (WMO)
CCQM	Consultative Committee on Amount of Substance (BIPM)
CDIAC	Carbon Dioxide Information Analysis Center (ORNL)
CIPM	International Committee on Weights and Measures (in French)
CO2 USA	CO_2 Urban Synthesis and Analysis workshops
COP	Conference of the Parties (UNFCCC)
CRDS	Cavity Ring-Down Spectroscopy
CRM	Certified Reference Material
EDF	Environmental Defense Fund
EDGAR	Emission Database for Global Atmospheric Research (European Joint Research Center)
EPA	Environmental Protection Agency (USA)
FF	Fossil Fuel related emissions
GAW	Global Atmosphere Watch (WMO)
GAWG	Gas Analysis Working Group (CCQM-BIPM)
GDP	Gross Domestic Product
GGMT	Greenhouse Gas Measurements and Technology conference
GHG	Greenhouse Gas
GGMI	Greenhouse Gas Management Institute
GOSAT	Greenhouse gases Observing SATellite (Japan)
GWP	Global Warming Potential

HESTIA	A project that quantifies all fossil fuel CO_2 emissions for individual cities to the building and street scale (Northern Arizona University)
HITRAN	HIgh-resolution TRANsmission molecular absorption database (US Air Force)
HRRR	High Resolution Rapid Refresh, a NOAA real-time 3-km resolution, hourly updated, cloud-resolving, convection-allowing atmospheric model
HYSPLIT	HYbrid Single Particle Lagrangian Integrated Trajectory Model (NOAA ARL)
ICLEI	Local Governments for Sustainability
ICOS	Integrated Carbon Observation System (Europe)
IG^3IS	Integrated Global Greenhouse Gas Information System (WMO)
INFLUX	Indianapolis Flux Experiment
IPCC	Intergovernmental Panel on Climate Change (UN)
IPCC TFI	IPCC Task Force on national greenhouse gas Inventories
KRISS	Korea Research Institute of Standards and Science
LSCE	Laboratoire des Sciences du Climat et de l'Environnement (France)
LNE	Laboratoire National de Metrologie et d'Essais (France)
LLNL	Lawrence Livermore National Laboratory (Dept. of Energy)
MIPAS	Michelson Interferometer for Passive Atmospheric Sounding (European Space Agency)
MOU	Memorandum of Understanding
NAM	North American Mesoscale Forecast System
NASA	National Aeronautics and Space Administration
NDACC	Network for the Detection of Atmospheric Composition Change (NOAA)
NDCs	Nationally Determined Contributions (UNFCCC)
NDIR	Non-Dispersive InfraRed instruments
NEI	National Emissions Inventory
NIM	National Metrology Institute (of China)
NIST	National Institute of Standards and Technology (Dept. of Commerce)
NMI	National Metrology Institute
NMIJ	National Metrology Institute of Japan
NOAA	National Oceanic and Atmospheric Administration (Dept. of Commerce)
NPL	National Physical Laboratory (UK)
NWP	National Weather Prediction models
OAS	Organization of American States
OCO	Orbiting Carbon Observatory (NASA)
ODIAC	Open-Data Inventory for Anthropogenic Carbon dioxide (NASA)

ORNL	Oak Ridge National Laboratory (Dept. of Energy)
OSSE	Observing Systems Simulation Experiments (Global Modelling & Assimilation Office, NASA)
OSTP	Office of Science and Technology Policy (The White House)
PBL	Planetary Boundary Layer
PSU	Pennsylvania State University
R&D	Research and Development
SBU	Stony Brook University
SIF	Solar Induced Fluorescence
SIM	Inter-American Metrology System
SIO	Scripps Institution of Oceanography
SOCCR	State of the Carbon Cycle Report (USGCRP)
SRM	Standard Reference Material (NIST)
STILT	Stochastic Time-Inverted Lagrangian Transport model
TCCON	Total Carbon Column Observing Network (An international collaboration)
TROPOMI	TROPOspheric Monitoring Instrument (on the Copernicus Sentinel-5 Precursor satellite)
UNDP	United Nations Development Program
UNEP	United Nations Environment Program
UNFCCC	United Nations Framework Convention on Climate Change
USGCRP	US Global Change Research Program
Vulcan	A project that quantifies all fossil fuel CO_2 emissions for the entire U.S. at high space- and time-resolution (Northern Arizona University)
WMO	World Meteorological Organization (UN)
WRI	World Resources Institute

Appendix B

Research Staff Involved in Greenhouse Gas Measurements Program

Purdue University: Prof. Paul B. Shepson (PI)

Graduate Students:	Degree Year	Research Topic (Current Position)
Dana R. Caulton	Ph.D. 2014	(Asst. Prof. at Univ. Wyoming, Atmospheric Science Dept.)
Tegan N. Lavoie	Ph.D. 2016	(U.S. Environmental Protection Agency)
Olivia E. Salmon	Ph.D. 2018	Urban Atmospheric Water Vapor and its Stable Isotopologues. (Wisconsin Dept. Natural Resources)
Kristian D. Hajny	Ph.D. 2020	(Senior Postdoctoral Fellow at Stony Brook Univ.)
Jay M. Tomlin	Ph.D. 2022	(NASA Ames)
Kelly L. Mays	M.S. 2009	(Eurofins Biopharma)
Chloe Gore	B.S. 2016	

Postdoctoral Researchers:

Maria O.L. Cambaliza	2010 – 2014	(Associate Professor and Chair of the Department of Physics of the Ateneo de Manila University, Philippines)
Alexie M. Heimburger	2014 – 2016	(Purity Laboratories/Ayumetrix, Hillsboro, OR)
Anna-Elodie M. Kerlo	2015 – 2016	
Rebecca M. Harvey	2016 – 2017	
Thilina Jayarathne	2017 – 2019	(Bristol Myers Squibb, NJ)

Stony Brook University: Prof. Paul B. Shepson (PI)

Graduate Student:	Degree Year	Research Topic (Current Position)
Renée P. Chabot	in progress	

Postdoctoral Researchers:

Joseph R. Pitt	2019 – 2021	(University of Bristol, UK)
Kris Hajny	2021 – Present	
Austin Hope	2021 – Present	

Senior Research Staff:

Israel Lopez-Coto	2022-Present

Pennsylvania State University: **Prof. Kenneth J. Davis (PI), Dr. Natasha L. Miles and Dr. Scott J. Richardson (Co-PIs)**

Graduate Students:	**Degree Year**	**Research Topic (Current Position)**
Laura McGowan	M.Sc. 2013	Simulation of Atmospheric Carbon Dioxide in the Indianapolis Region. (Climate Risk Modeling)
Daniel P. Sarmiento	Ph.D. 2017	Urban land-surface impacts on atmospheric processes. (NASA/Goddard Space Flight Center)
Kai Wu	Ph.D. 2020	Joint estimation of fossil fuel and biogenic CO_2 fluxes in an urban environment. (Institute of Atmospheric Physics, China)
Yue Pan	Ph.D. 2022	Evaluation of the impact of grid spacing and scale-aware PBL scheme to urban meteorological simulations using WRF-LES. (Private Sector)
Vanessa C. Monteiro	M.S. 2018	The Convective Boundary Layer in the Amazon rainforest.
Samantha Murphy	in progress	Flux measurements and modeling of rural CO_2 fluxes.
Helen Kenion	in progress	Determination of local fluxes using Monin-Obukhov Similarity Theory. (MOST)
Jason Horne	in progress	Understanding urban grass fluxes.
Eliott Foust	in progress	Modeling neighborhood scale near-surface climate.

Postdoctoral Researchers:

Nikolay Balashov	2016-2019	Background heterogeneity and other uncertainties in estimating urban methane flux. (NASA Goddard Space Flight Center)
Thomas Lauvaux	2011-2018	Urban Inversions. (Université de Reims Champagne-Ardenne, France)
Brian J. Nathan	2015-2017	Multi-gas Inverse methods. (SRON Netherlands Institute for Space Research))

Research Staff:

Vanessa C. Monteiro	2019-2022	Micrometeorological Analysis of Urban GHG Observations. (University of Auckland, NZ)
Aijun J. Deng	2014-2017	Urban Atmospheric Modelling and Data Assimilation. (Utopus Insights)
Brian J. Gaudet	2014-2018	Urban Atmospheric Modelling. (Pacific Northwest National Laboratory)
Douglas K. Martins	2014-2015	Urban GHG Observational Networks. (Teledyne FLIR)
Zachary R. Barkley	2016-Present	Atmospheric Inversions and Budget Analysis.
Benjamin Ahiswede	2023-Present	Urban Eddy Covariance Flux Measurements.

University of Maryland: Profs. Russell R. Dickerson, Ross Salawitch and Ning Zang (PIs)

Graduate Students:	Degree year	Research Topic (Current Position)
Cory R. Martin	Ph.D. 2019	Evaluation and environmental correction of ambient CO_2 measurements from a low-cost NDIR sensor. (NOAA)
Sarah E. Benish	Ph.D. 2020	Examination of Photochemistry and Meteorology of Atmospheric Pollutants from the North China Plain. (Research Scientist at EPA, North Carolina)
Courtney D. Grimes	Ph.D. 2021	Light Absorbing Aerosols: Calibration, Measurements, and Emissions from New York City. (Environmental Defense)
Doyeon Y. Ahn	Ph.D. 2021	Quantifying the Emissions of Carbon Dioxide (CO_2), Carbon Monoxide (CO), and Nitrogen Oxides (NO_x) from Human Activities: Top-Down and Bottom-Up Approaches. (Postdoc at George Washington Univ.)
Dolly L. Hall-Quinlin	Ph.D. 2022	Air Pollution Emissions from Highway Vehicles: Quantifying Impacts of Humidity, Ambient Temperature, and COVID-19–Related Travel Restrictions. (Traffic Planner, Durham, NC)
Shaun T. Howe	M.S. 2019	Model Validation and Estimation of the Complete Carbon Cycle in the Northeastern United States.
Hannah Daley	in progress	Aircraft Observations for Determination of GHG Flux.
Sayantan Sahu	in progress	Evaluation of GHG Measurements and Models in the Baltimore-Washington Area.
Phillip R. Stratton	in progress	Measurements of GHGs from Landfills in the Baltimore-Washington Area.
Hannah Zafar	in progress	
Bari N. Turpie	B.S. 2015	
Kristy J. Weber	B.S. 2016	(Dept. Geography, Univ. Colorado at Boulder)
Julia Cavallo	B.S. 2016	Large Scale Surface Wind's Influence on CO_2
Alexa Maines	B.S. 2016	Using NASA GMAO G5NR to compare CO_2 concentration to atmospheric variables.
Jonathan R. Hansford	B.S. 2017	Computer Science
Bo Chen	B.S. 2018	Human Impact on Wildfire Carbon Emissions.
Research Staff:		
Xinrong Ren	2014-2019	(NOAA ARL)
Hao He	2015-	(Assistant Research Professor)

Northern Arizona University: Prof. Kevin R. Gurney (PI)

Graduate Students:	Degree Year	Research Topic (Current Position)
Daniel Mendoza	Ph.D. 2012	On-road $FFCO_2$ emissions. (Research Professor, Univ. Utah)
Jianhua Huang	Ph.D. 2016	Hestia codebase. (Senior Scientist, Univ. Phoenix)
Yang Song	Ph.D. 2018	On-road $FFCO_2$ emissions analysis.
Darragh O'Keeffe	M.S. 2017	Hestia-LA socioeconomic analysis. (GIS professional, Ireland)
Kevin Coltin	M.S. 2013	Power plant $FFCO_2$ estimation.
Ryan Anderson	M.S. 2015	Hestia-Indy policy analysis. (Env. Professional)
Huilin Sun	in progress	Urban $FFCO_2$ emissions. (NAU)

Postdoctoral Researchers:

Salvi Asefi-Najafabady	2012-2015	(Research Professor, Univ. Virginia)
Jianming Liang	2015-2018	(Environmental Systems Research Institute, Redlands, CA)
Risa Patarasuk	2013-2016	(GIS Planner, Orange County, FL)
Yuyu Zhou	2008-2010	(Associate Professor, Iowa State Univ.)
Geoffrey S. Roest	2019-2022	(Senior Data Scientist, Crosswalk Lab.)
Igor Razlivanov	2011-2013	
Preeti Rao	2013-2016	(Research Scientist, Univ. Michigan)
Yang (Terry) Song	2019-2021	
Anna Kato	Current	

Boston University: Prof. Lucy R. Hutyra (PI)

Graduate Students:	Degree year	Research Topic (Current Position)
Lauren Cabrera	B.Sc. 2018	(Natural Resources Analyst, Ag Reserves Inc.)
Wiley Hundermark	B.Sc. 2020	(Field Research Assistant, BU)
Conor Gately	Ph.D. 2015	(Postdoc at BU and Harvard)
Julia Marrs	Ph.D. 2022	Evaluating solar-induced fluorescence across spatial and temporal scales to monitor primary productivity. (Postdoc at NIST)
Ian Smith	Ph.D. 2023	Evidence for edge enhancements of soil respiration in temperate forests. (Postdoc at BU and World Bank)
Hristiana Stoynava	in progress	
Leeza Moldavchuk	in progress	

Postdoctoral Researchers:

Conor Gately	2015-2019	(Senior Planner, Metropolitan Area Planning Council, MA)
Joy Winbourne	2017-2020	(Faculty at University of Massachusetts, Lowell)
Taylor Jones	2019-2023	(Research Faculty at BU; Faculty at Georgetown Univ.)
Andrew Reinmann	2014-2017	(Faculty at City University of New York)
Yo Rum Hwuang	2023-	

Scripps Institution of Oceanography: Prof. Ralph Keeling and Prof. Ray F. Weiss (PIs)

Graduate Students:	**Degree year**	**Research Topic (Current Position)**
Research Staff:		
Jooil Kim	Ph.D. 2011	

University of Michigan: **Prof. Eric A. Kort (PI)**

Graduate Students:	**Degree year**	**Research Topic (Current Position)**
John Ware	Ph.D. 2018	Factors Impacting Observation-Based Estimates of Urban Greenhouse Gas Emissions (Union Organizer at AFT Michigan)
Alexander Gvakharia	Ph.D. 2019	The Atmospheric Impact of Energy and Food Production (Senior Researcher at Systems & Technology Research)
Erica Whiting	in progress	
Postdoctoral Researchers:		
Genevieve Plant	2017-	(Assistant Research Scientist at Univ. Mich.)
Yaoxian Huang	2017-2019	(Assistant Professor at Wayne State University)

NOAA/Univ. Colorado Researchers

Jocelyn C. Turnbull		(GNS Science, NZ)
John Miller		
Colm Sweeney		
Ariel Stein		
Timothy Newberger		
R. Michael Hardesty		
Timothy A. Bonin		
W. Alan Brewer		
Isaac J. Vimont	Ph.D. 2017	(NOAA/ESRL)
Kathryn McKain		

NASA Researchers

Charles E. Miller		
Riley M. Duren		(Carbon Mapper and Arizona State Univ.)
Kristal R. Verhulst		
Vineet Yadav		
Preeti Rao		(Univ. Michigan)
Jonathan A. Wang		(Univ. Calif-Irvine)
Thomas Pongetti		
Claire Wong		
Francesca M. Hopkins		(Univ. California - Riverside)

Earth Networks, Inc.

Christopher D. Sloop
Steve Prinzivalli
Clayton Fain
William Callahan
Michael Stock
Elizabeth DiGangi

NIST Researchers

Anna Karion	
Israel Lopez-Coto	(Research Staff, Stony Brook University)
Kuldeep Prasad	
Sharon M. Gourdji	(Lead Agricultural Modeler, RMS)
Subhomoy Ghosh	(Research Staff, University of Notre Dame)
Kimberly L. Mueller	
Daniel V. Samarov	
Antonio Possolo	
Tamae M. Wong	
James R. Whetstone	

NIST Guest Researchers

Jin Tao	(NIM China - GHG Measurements)
Hong Lin	(NIM China - CRDS, Urban Testbeds)
Zhang Liang	(NIM China - Smokestack measurements and standards)
Liu Xiaomeng	(NIM China - Differential Absorption Lidar)
Hu Heming	(NIM China - Atmospheric transport modeling in cities)
Wu Hai	(NIM China - GHG concentration standards)