Towards Understanding of the Changes in Aerosol Sources, Trends, and Composition in the Southeastern and Continental United States

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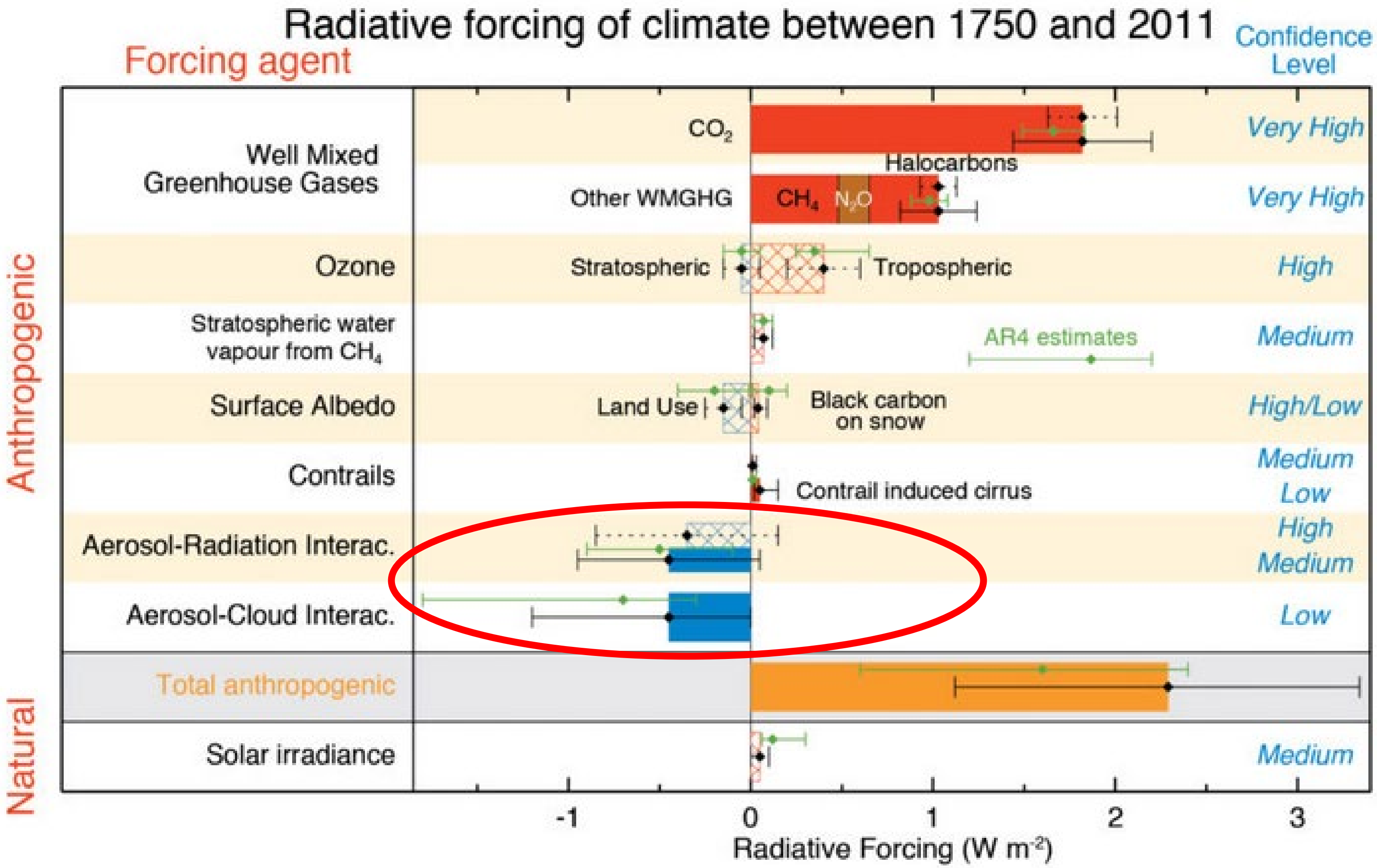
Air Quality Management Strategy Platform, Taiwan
February 24, 2025

Aerosols; Particulate Matter (PM)

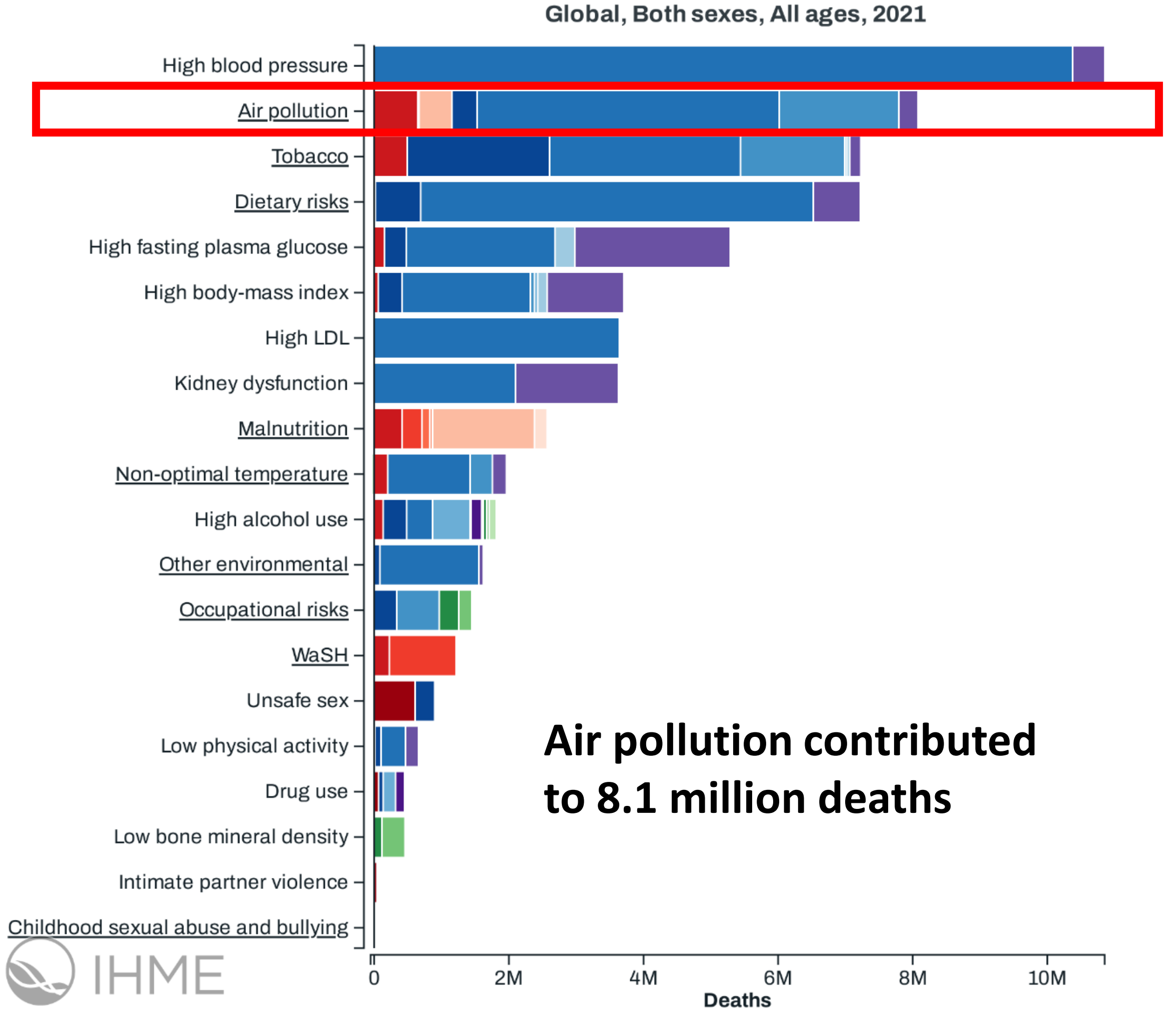
- Diverse sources, impacts on climate and human health

Effects of Aerosols on Climate

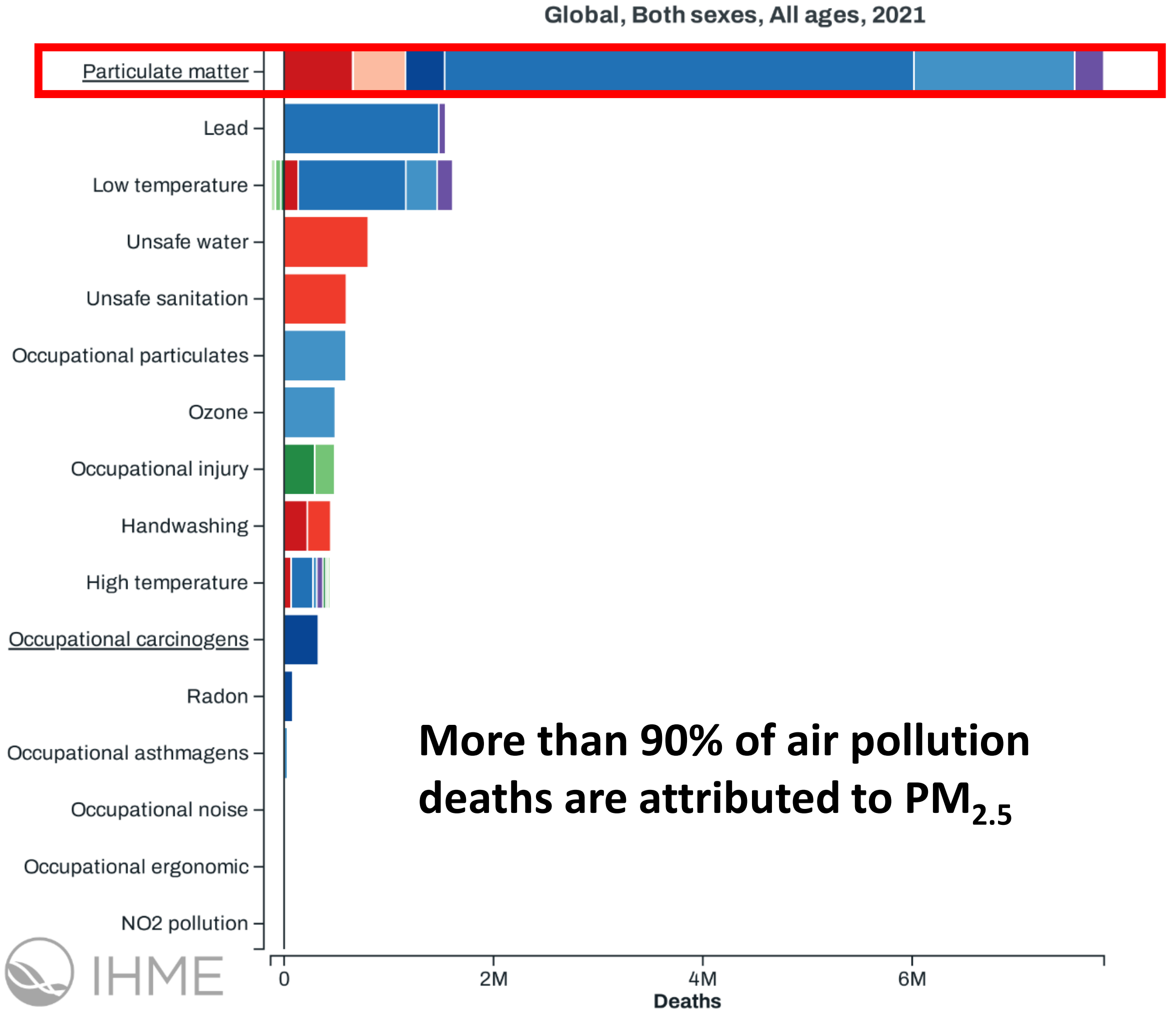
Intergovernmental Panel on Climate Change



Effects of Aerosols on Health



Air pollution contributed to 8.1 million deaths

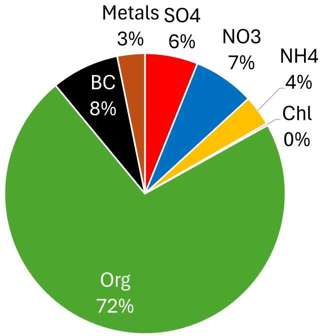


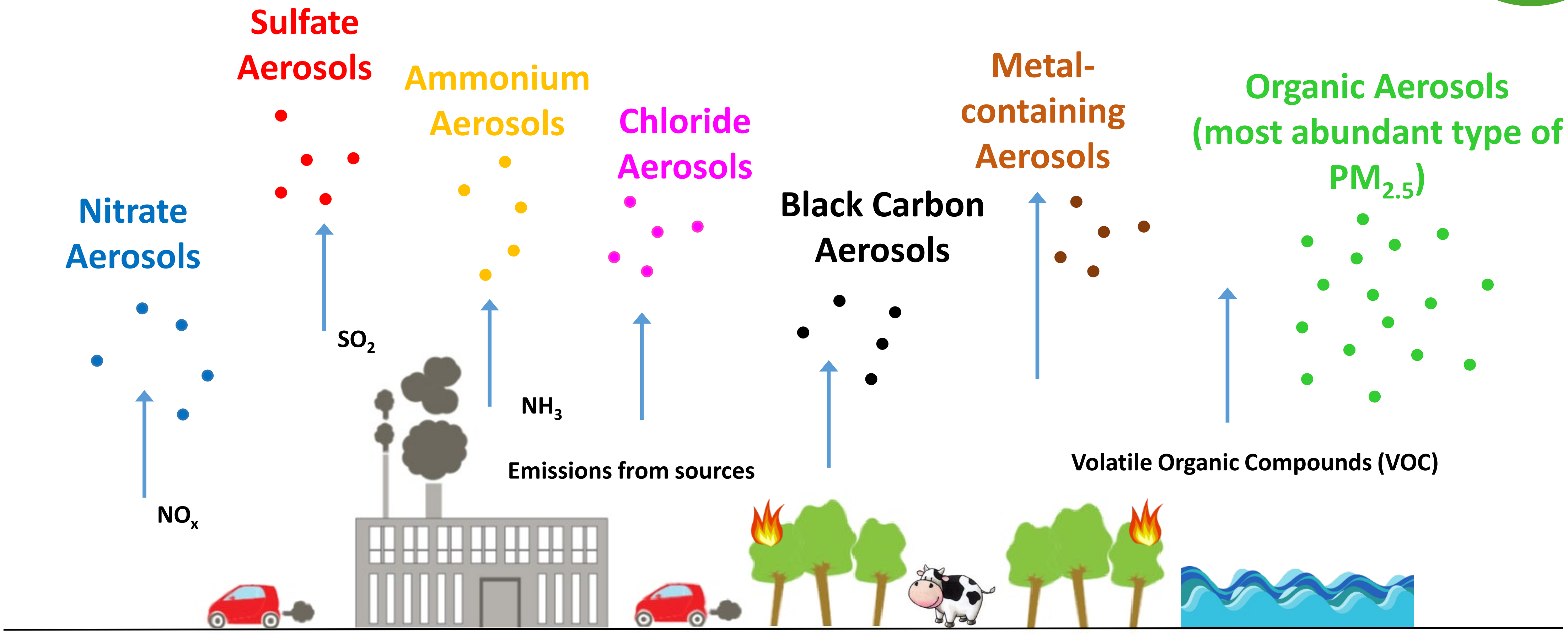
More than 90% of air pollution deaths are attributed to $PM_{2.5}$

State of Global Air, HEI, 2024

Chemical Diversity of $PM_{2.5}$

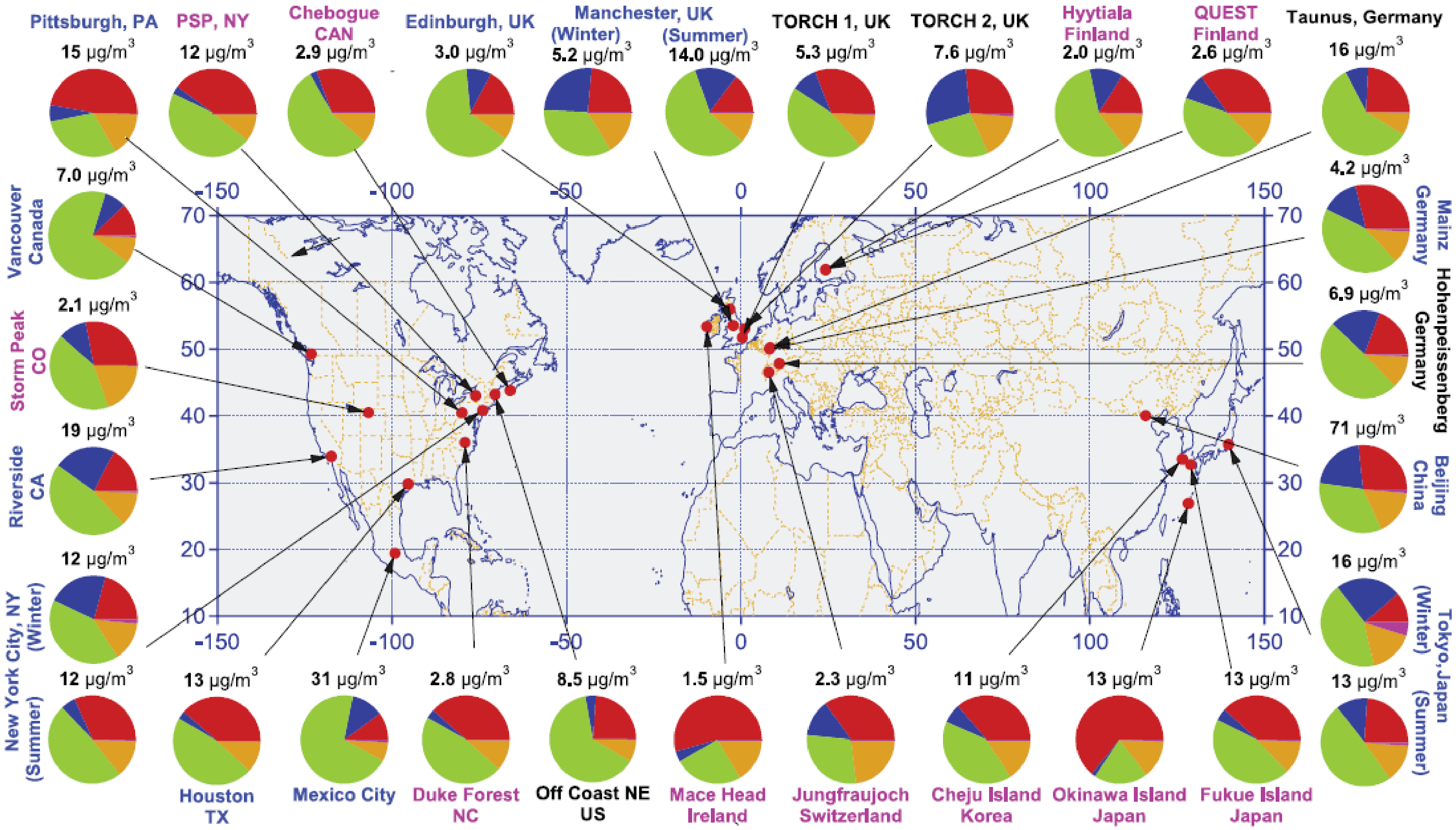
Data from Atlanta



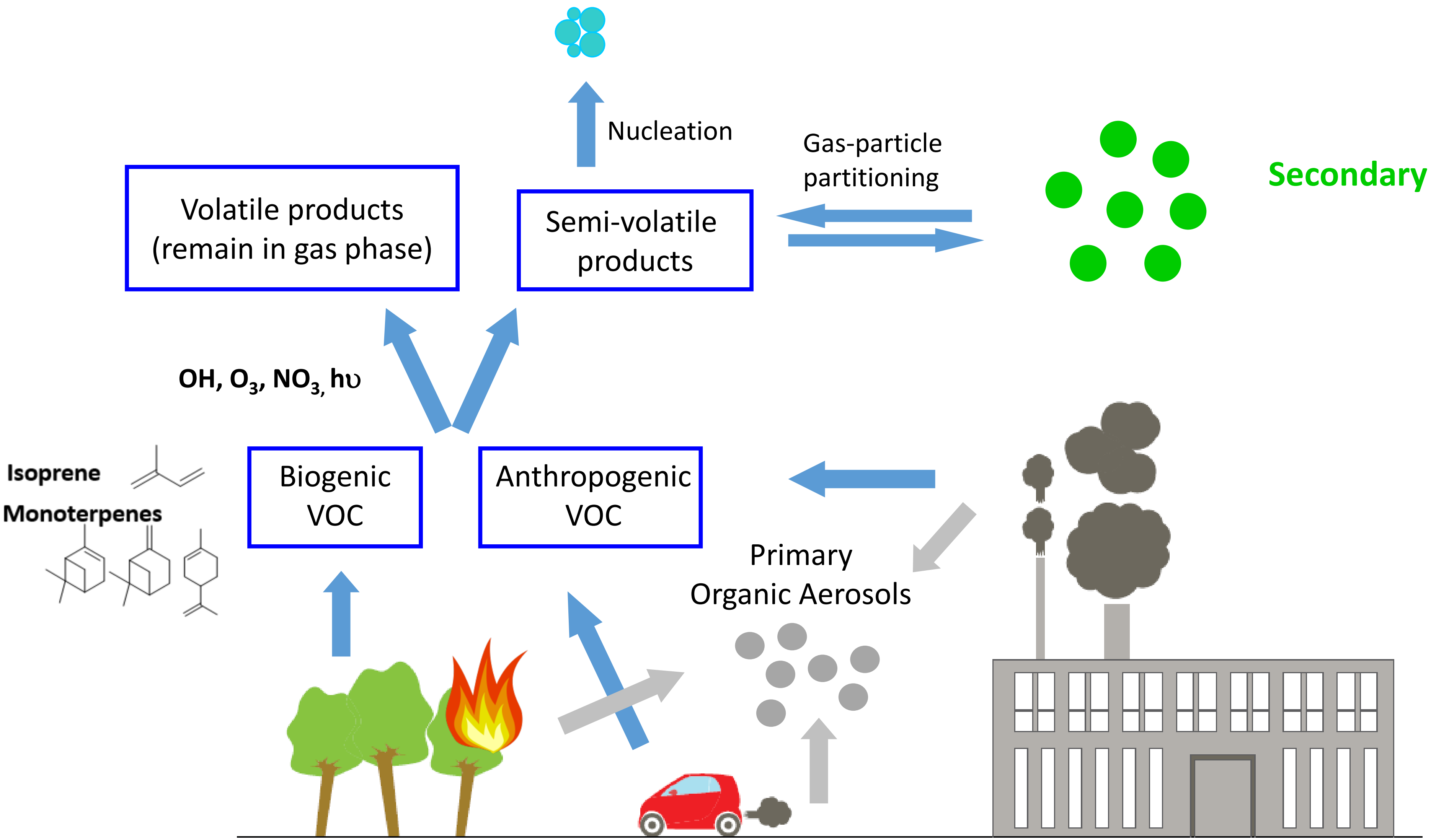


Organic and Inorganic Aerosols

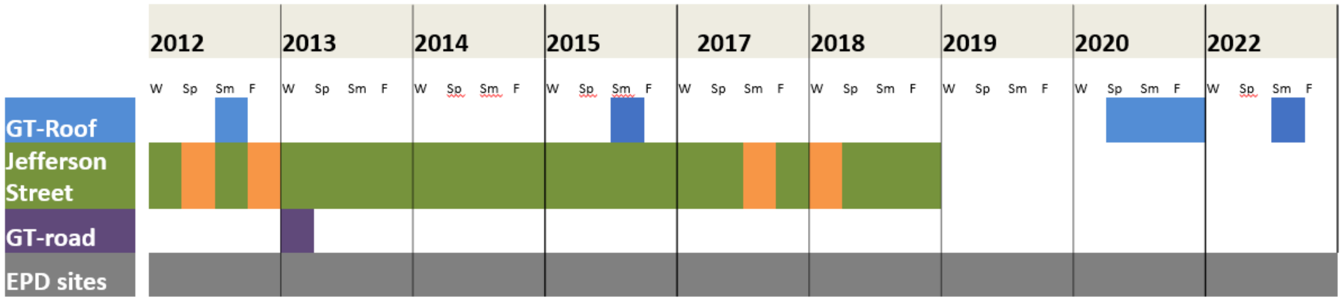
- Aerosol Mass Spectrometer (AMS): **Organics**, **Sulfate**, **Nitrate**, **Ammonium**, **Chloride**
- **Organics** account for ~50% of submicron non-refractory PM mass (Zhang et al., 2007)



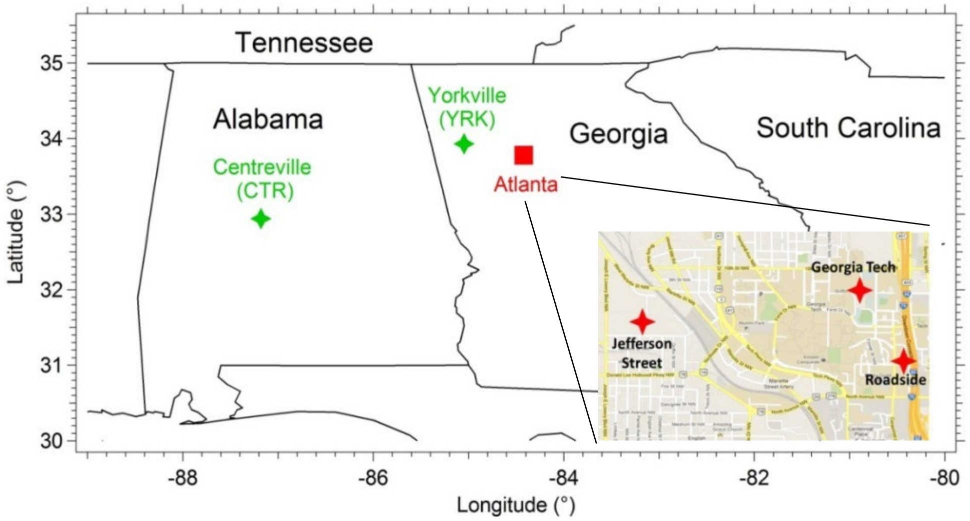
Organic Aerosols: Primary vs. Secondary



Decadal Aerosol Measurements in the southeastern U.S.



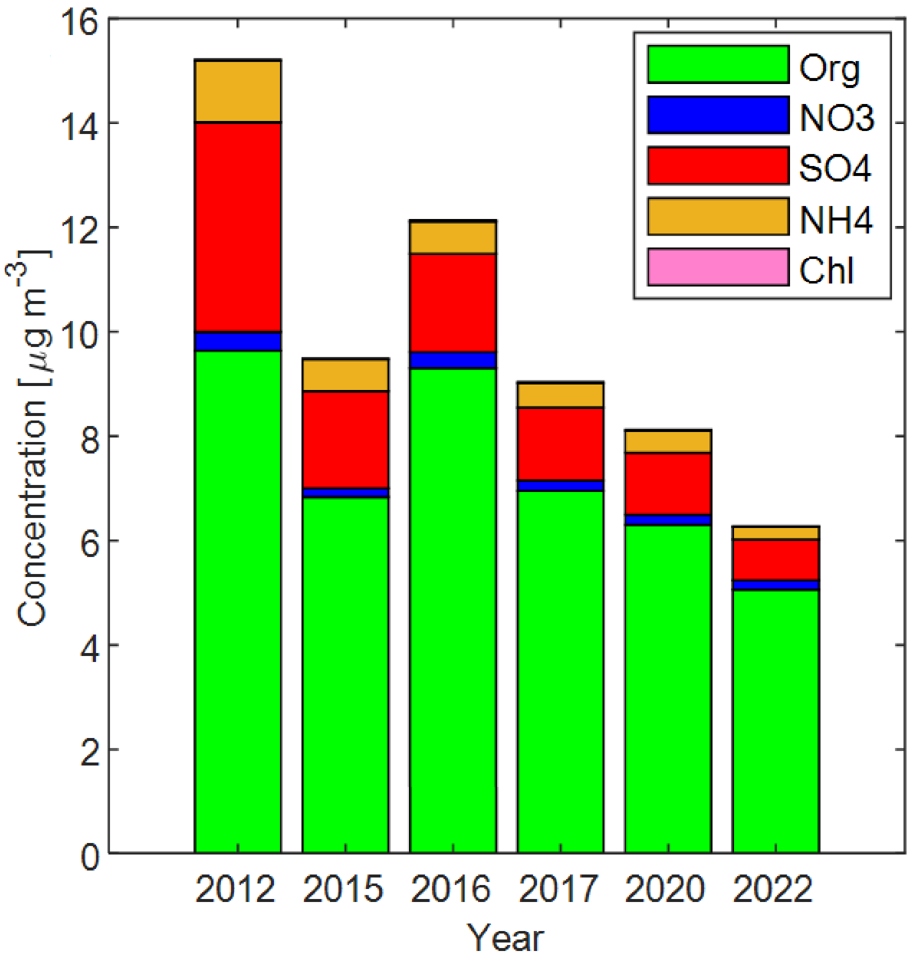
Xu et al., PNAS, 2015; Xu et al., ACP, 2015; Xu et al., ACP, 2018; Chen et al., ACP, 2020; Chen et al., ES&T, 2021; Joo et al., ACS ESC, 2021



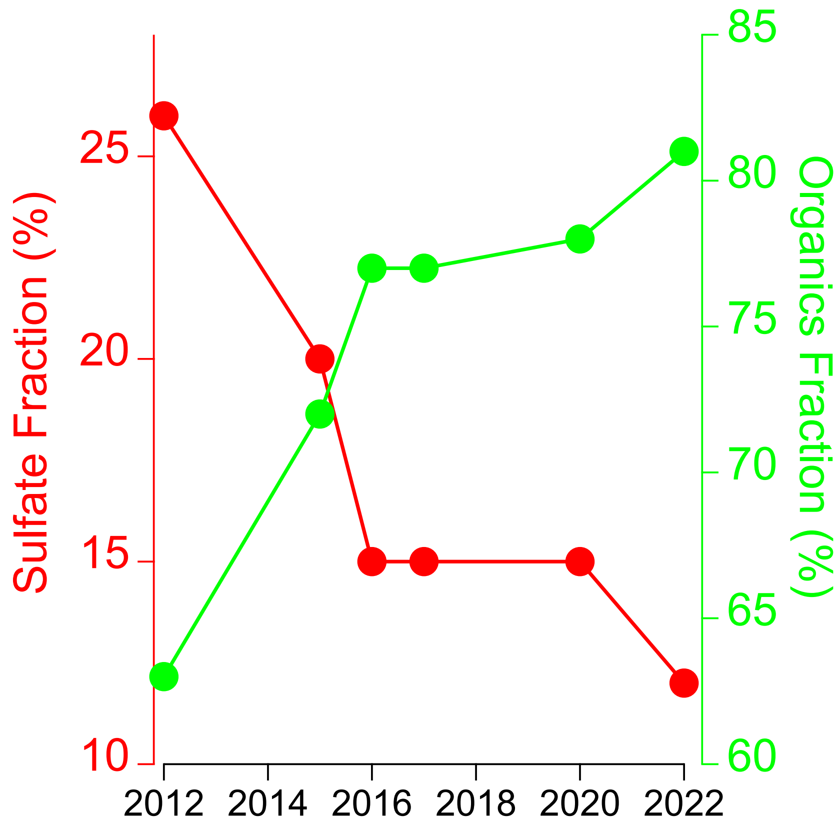
High Resolution Time-of-Flight Aerosol Mass Spectrometer (**HR-ToF-AMS**)

- Bulk aerosol chemical composition
- Non-refractory aerosols: Organics, Sulfate, Nitrate, Ammonium, Chloride

Increasing Dominance of Organic Aerosols

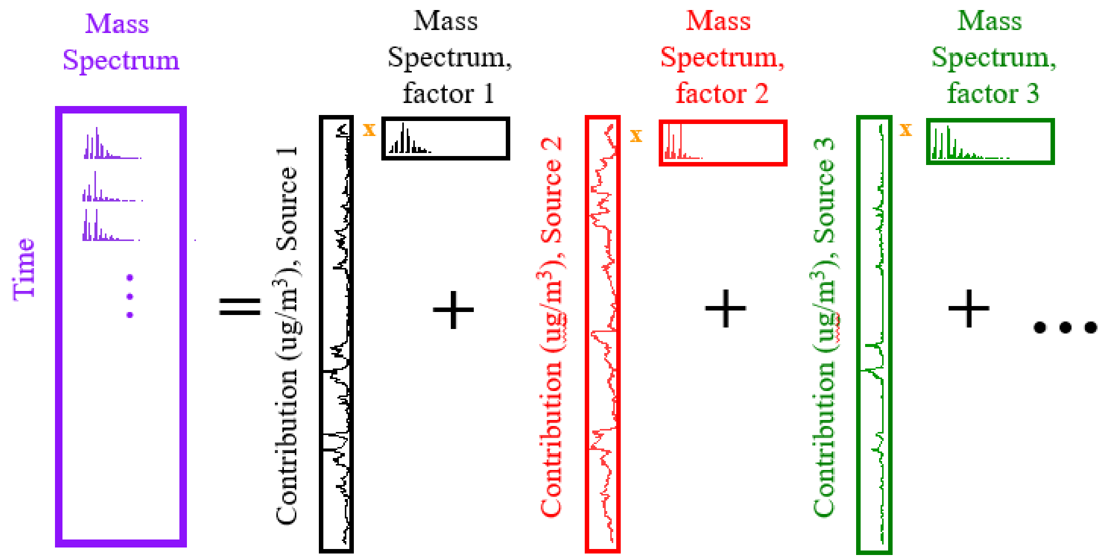


UNPUBLISHED DATA, Liang et al., in prep.



- Sulfate fraction decreased from 26% to 12%
- Organics fraction increased from 63% to 81%
- "Trading" sulfate for organics

Source Apportionment Analysis of Organic Aerosols



Unique mass spec features, diurnal trends, correlations with external tracers, etc.

Positive Matrix Factorization (PMF) is a bilinear mixing model in which a dataset matrix is assumed to be comprised of the linear combination of factors with constant profiles that have varying contributions across the dataset.

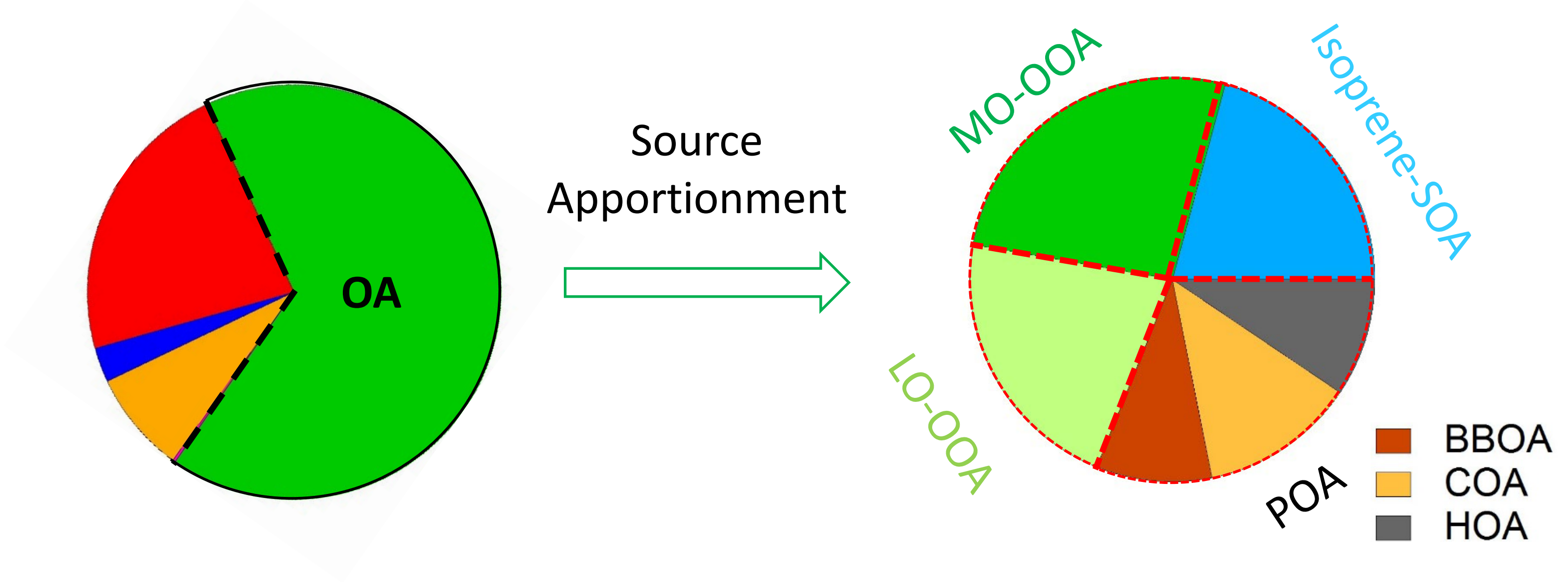
Ulbrich et al. 2009, ACP

Different Subtypes of Organic Aerosols

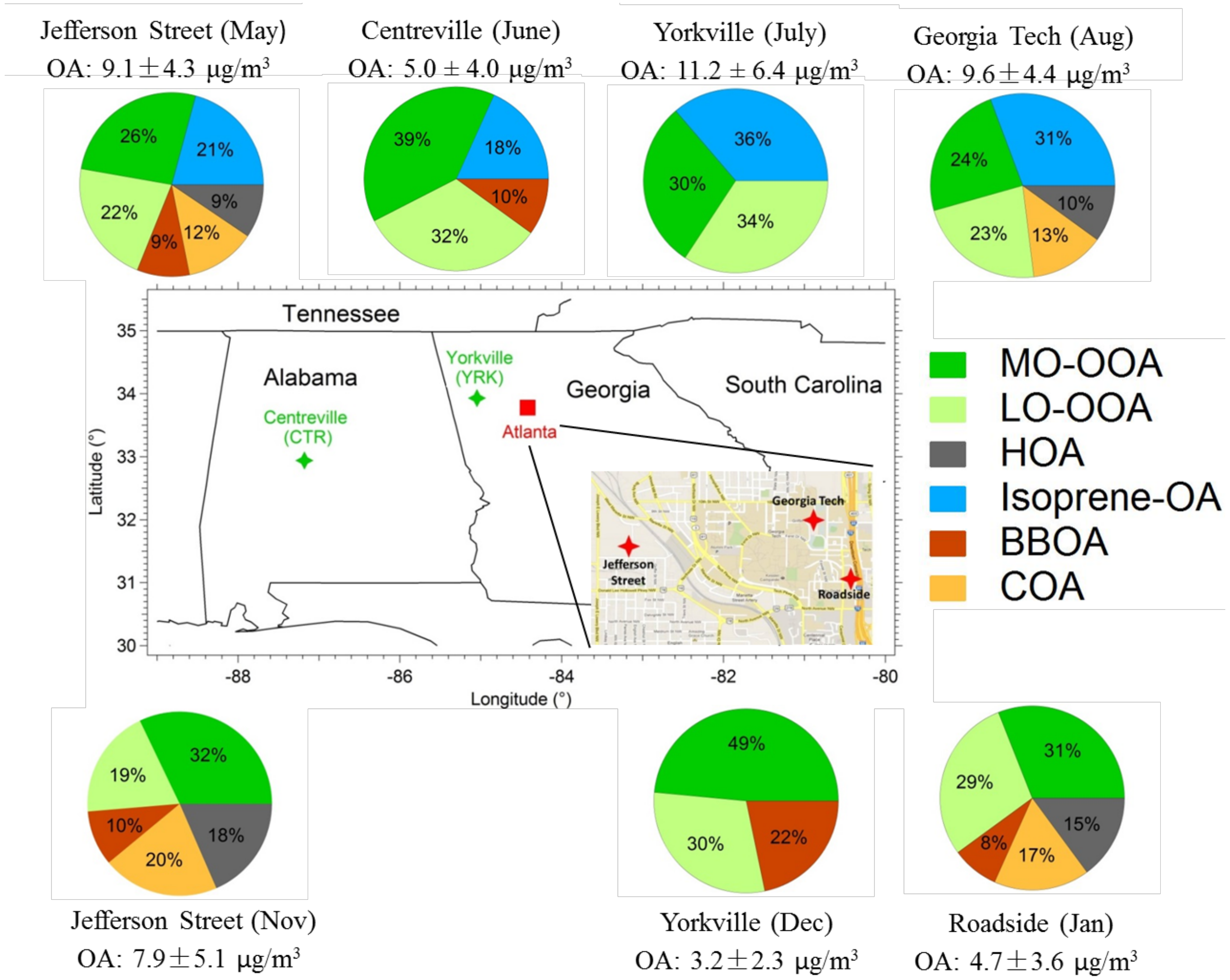
POA
- COA: Cooking OA
- HOA: Hydrocarbon-like OA

SOA
- LO-OOA: Less-oxidized oxygenated OA
- **MO-OOA: More-oxidized oxygenated OA**
- **Isop-SOA: Isoprene-SOA (biogenics)**

Source Apportionment Analysis of Organic Aerosols



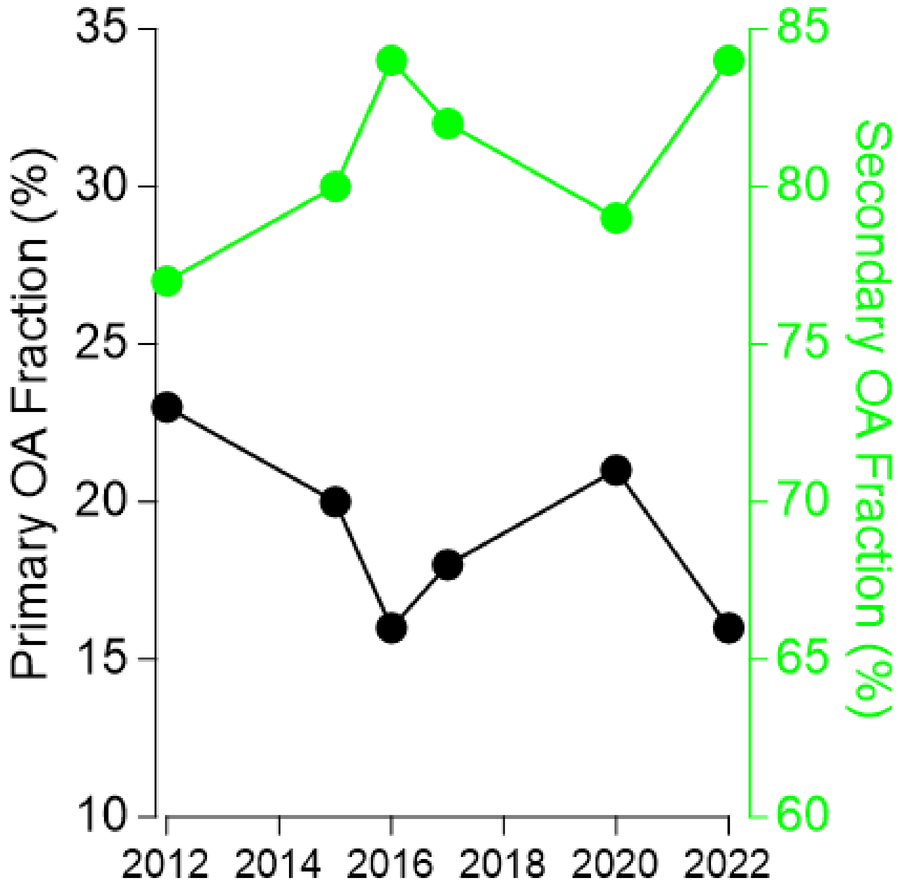
OA Source Apportionment in the southeastern U.S.



Xu et al., PNAS, 2015

- Small contributions of HOA (primary OA)
- Cooking OA present in urban areas
- BBOA present most of the time
- Majority of OA is SOA
- Isoprene-SOA only present in summer
- LO-OOA and MO-OOA
  - Present all year round
  - Large contributions

Changes of POA and SOA over the Last Decade



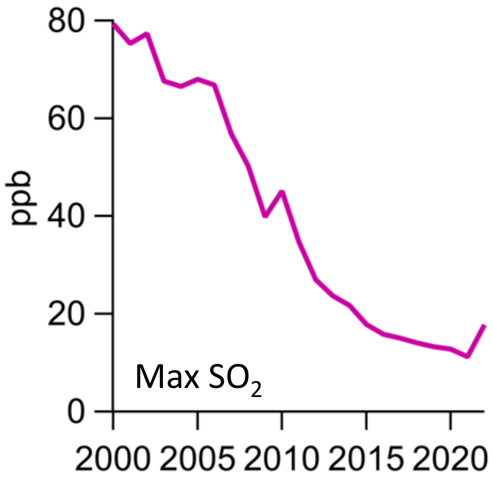
UNPUBLISHED DATA, Liang et al., in prep.

- Increasing dominance of SOA (84% in 2022)
- Contribution of POA decreased over time
  - Reductions in primary emissions

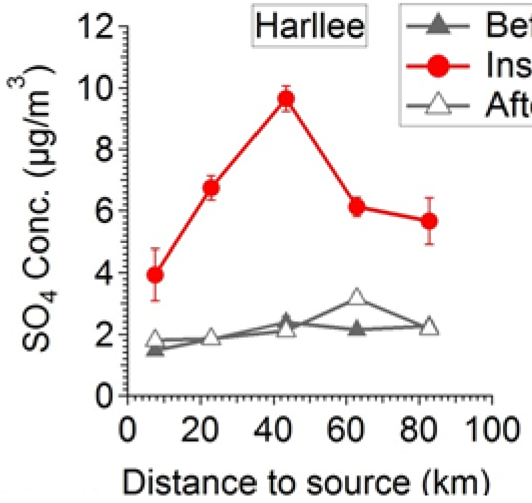
https://saportareport.com/atlantas-urban-tree-canopy-leads-nation-trees-not-protected/columnists/maria_saporta/

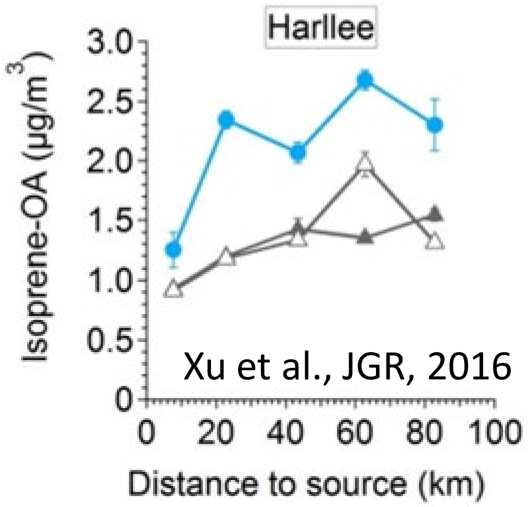
https://www.gpb.org/news/2022/06/21/three-georgia-power-plants-land-on-list-of-nations-dirtiest

A story of trees and power plant emissions



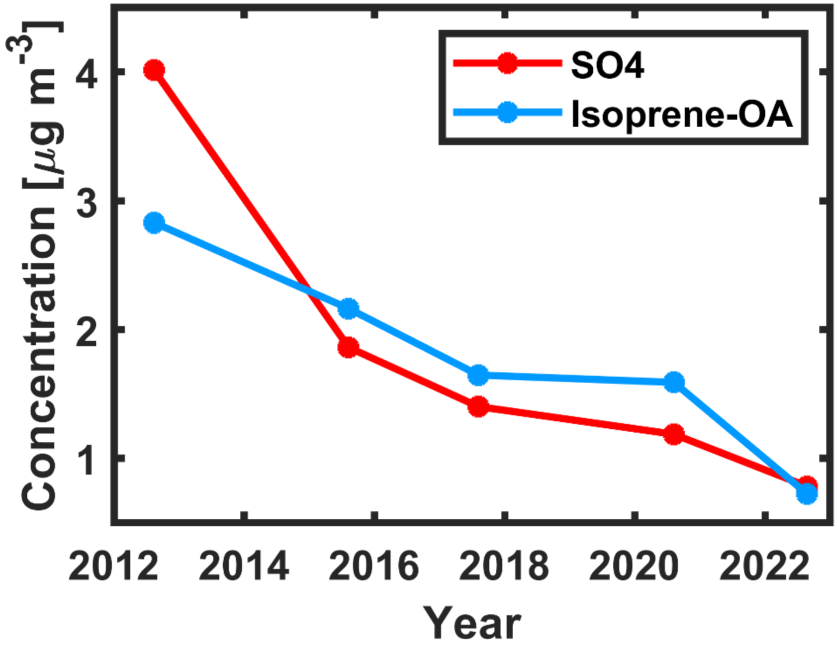
Isoprene-SOA Formation is Controlled by Sulfate





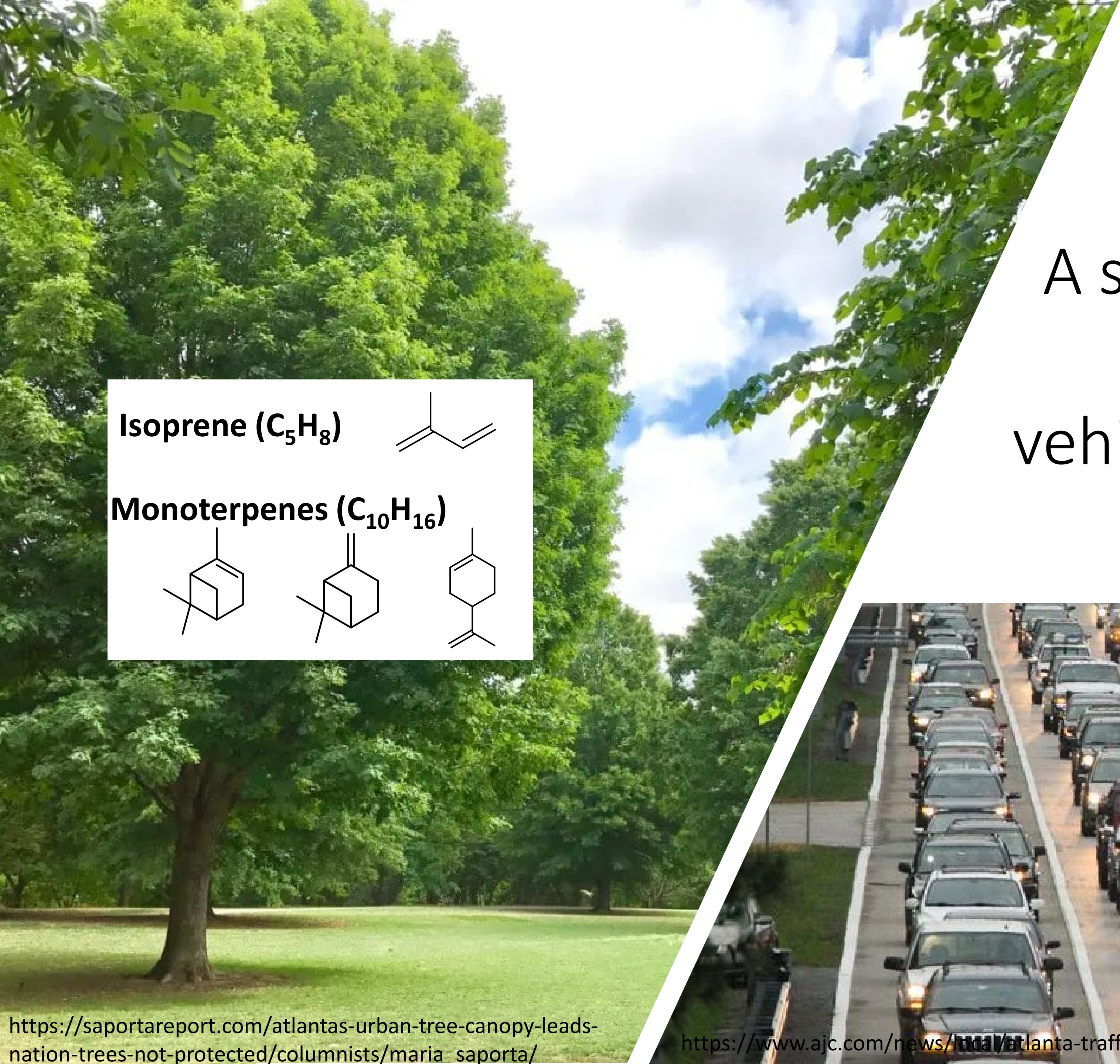
- Isoprene-SOA formation mechanism is controlled by sulfate (Xu et al., PNAS, 2015; Xu et al., JGR, 2016)

UNPUBLISHED DATA, Liang et al., in prep.

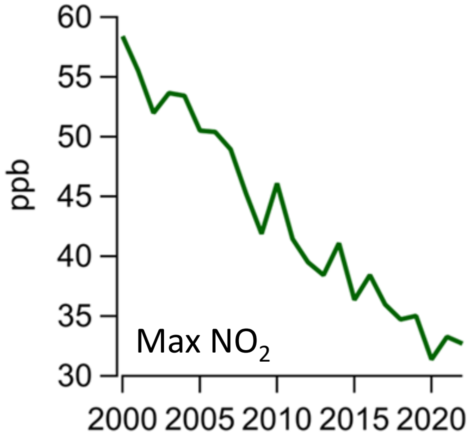


- Reducing SO_2 emission from power plants is an effective way to reduce Isoprene-SOA (tree emissions)
- 1 μg m^{-3} decrease of sulfate is accompanied by ~0.7 μg m^{-3} decrease of Isoprene-SOA

A story of trees and vehicle emissions

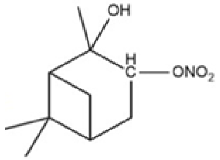


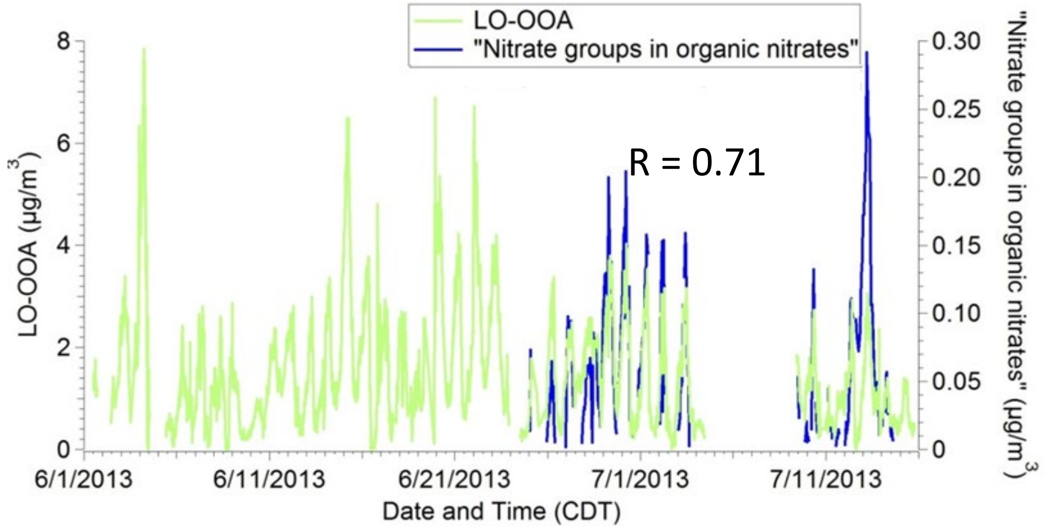
https://saportareport.com/atlantas-urban-tree-canopy-leads-nation-trees-not-protected/columnists/maria_saporta/



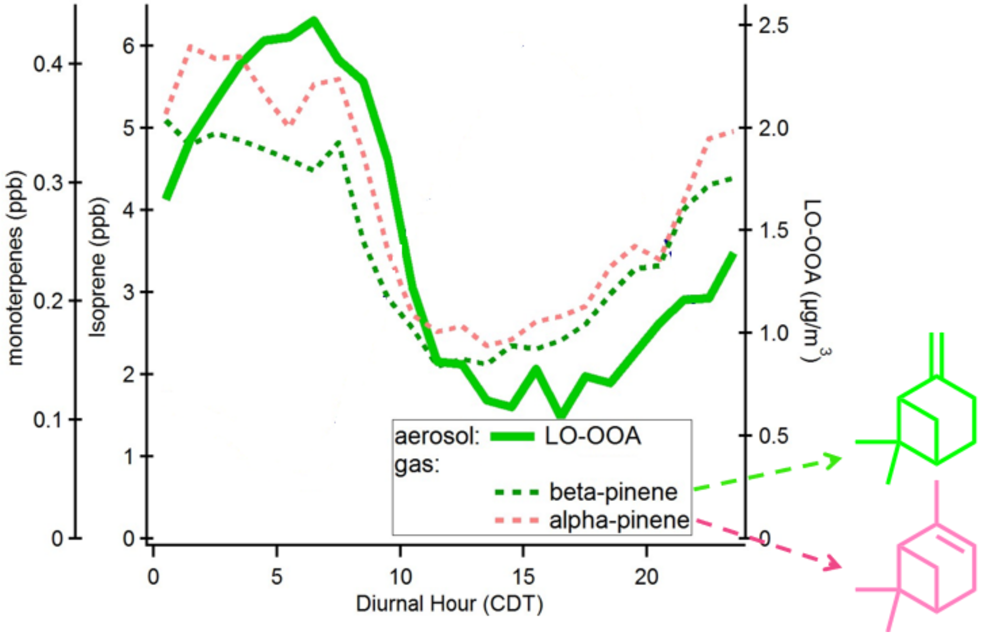
https://www.ajc.com/news/local/atlanta-traffic-among-worst-the-world-study-finds/C6JR110E1z9xZeGGmjJ2HM/

LO-OOA and Organic Nitrates





Xu et al., PNAS, 2015; Chen et al., ACP, 2020; ES&T, 2021



Monoterpenes are the largest source of summertime OA in the SE US (Xu et al., ACP, 2018)

Monoterpene + OH/O_3 + NO_x (daytime) → LO-OOA + Organic nitrates

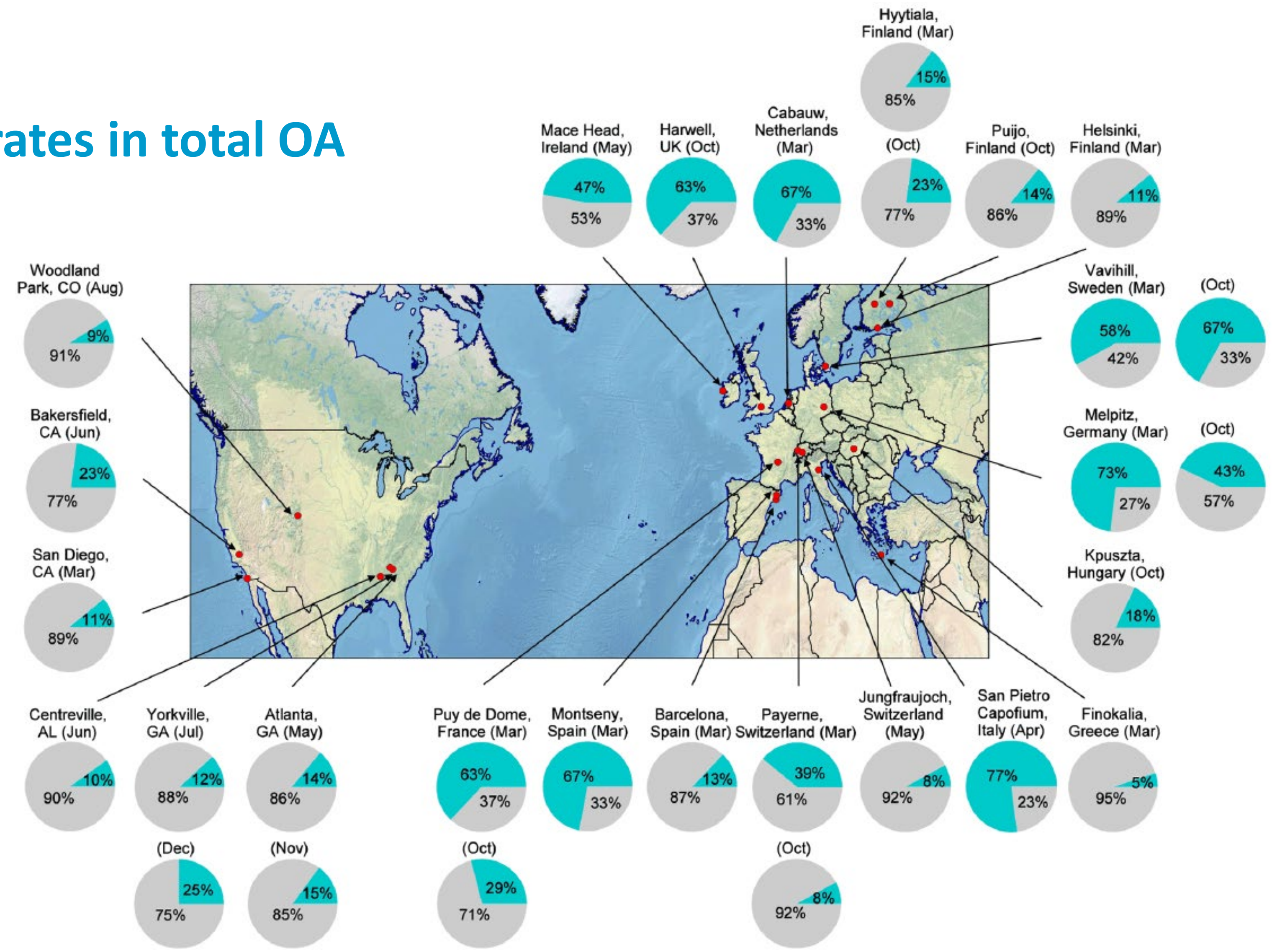
Monoterpene + NO_3 (nighttime) → LO-OOA + Organic nitrates

(biogenic) (anthropogenic)

Organic Nitrates

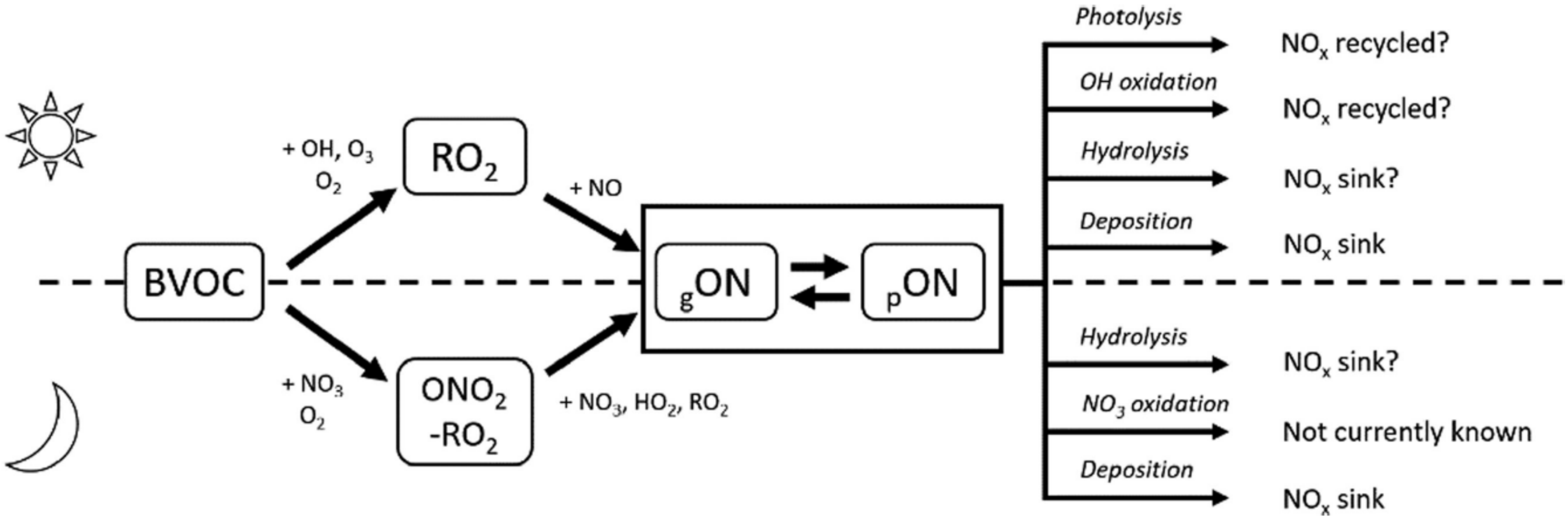
Ng et al., ACP, 2017

% of organic nitrates in total OA



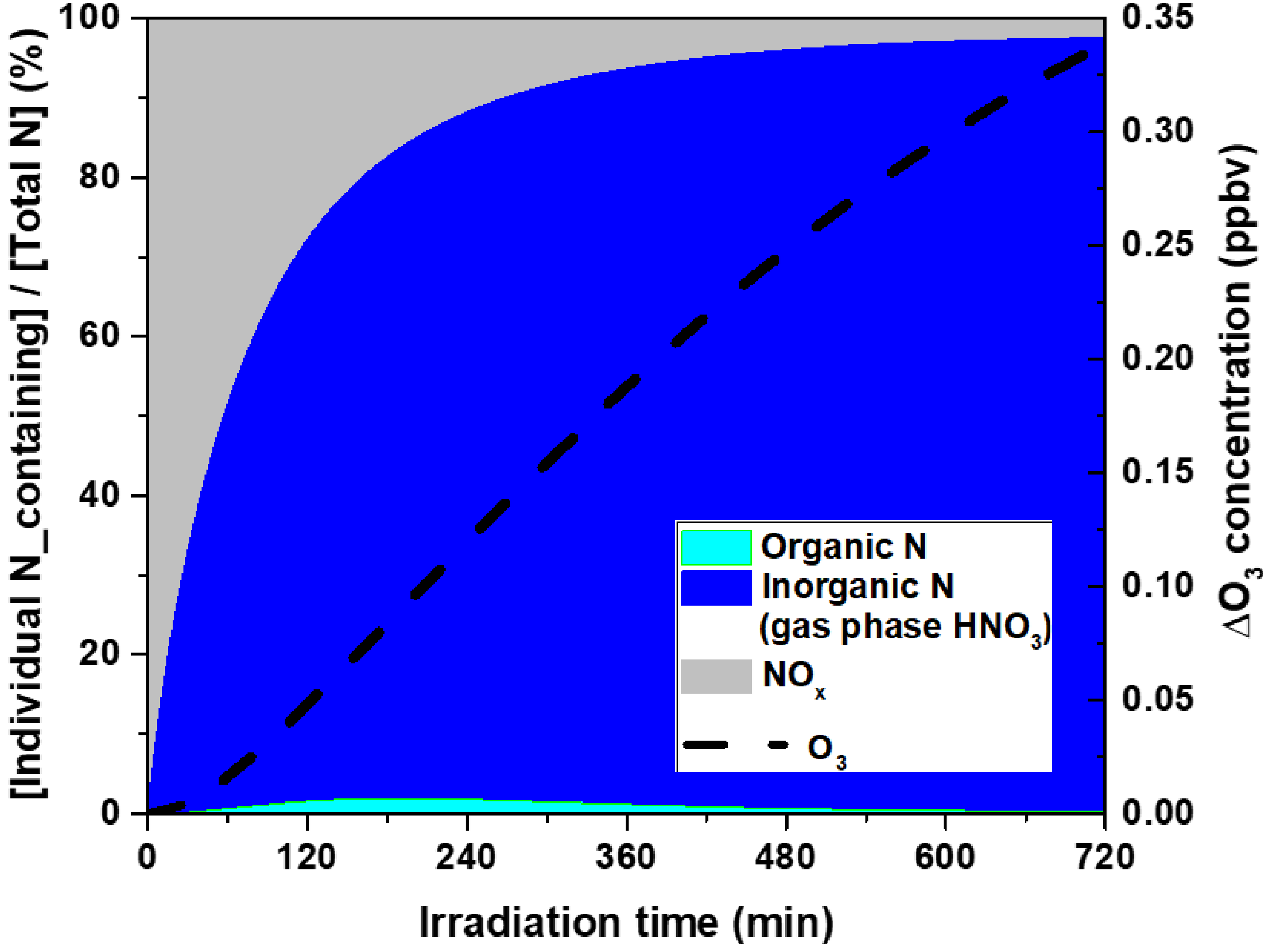
Organic Nitrates: Impact on NO_x recycling, Ozone, SOA

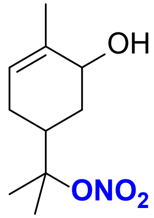
- Formation and fates of organic nitrates
- Main uncertainty in NO_x recycling comes from organic nitrates ($RONO_2$)



Takeuchi and Ng, ACS Book Chapter, 2018
Takeuchi and Ng, ACP, 2019
Wang et al. ES&T, 2021
Wang et al. J Phys Chem A, 2023
Takeuchi et al., AS&T, 2024

Organic Nitrate Photolysis and Ozone Formation





- Photolysis of organic nitrate
- Master Chemical Mechanism (MCM) and our proposed mechanism

- Organic nitrate was converted into:
  - NO_x (2.8% mass of reactive nitrogen)
  - Inorganic nitrate (gaseous HNO_3, 96.8%)
  - Small fraction (0.40%) remained in the form of organic nitrates

- **O_3 increased**

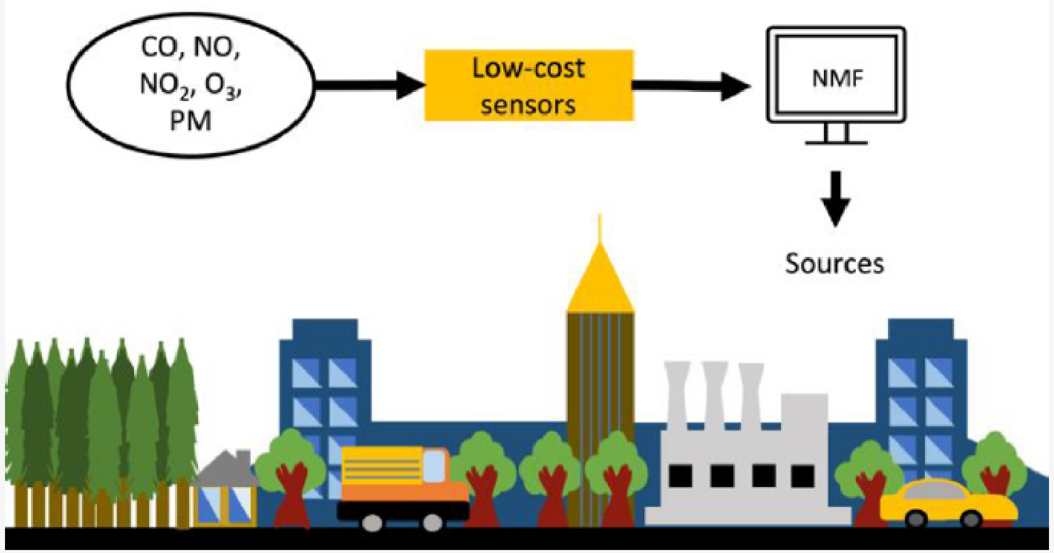
Wang et al. J Phys Chem A, 2023

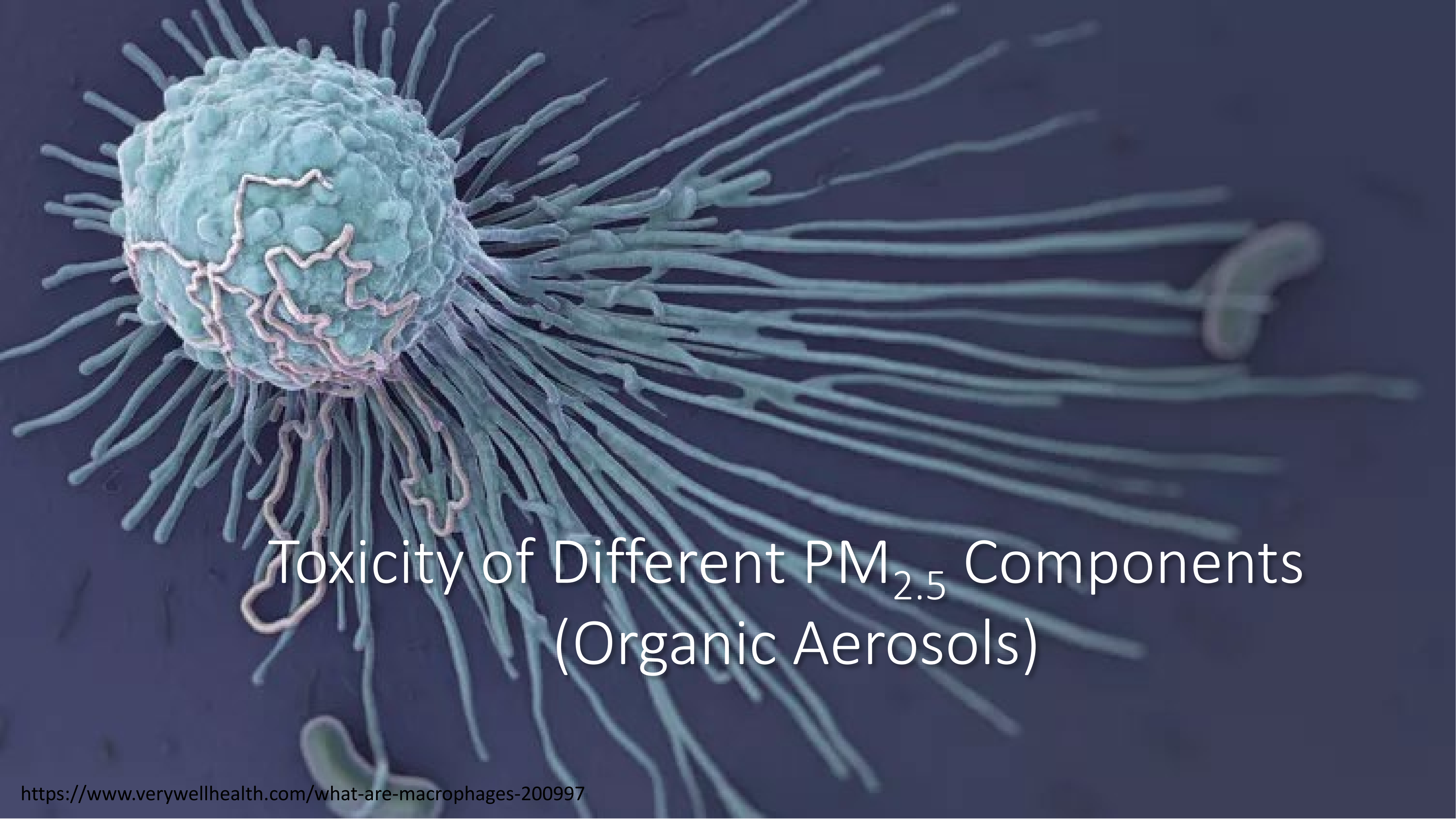
Investigating the Sources of Urban Air Pollution Using Low-Cost Air Quality Sensors at an Urban Atlanta Site

Laura Hyesung Yang, David H. Hagan, Jean C. Rivera-Rios, Makoto M. Kelp, Eben S. Cross, Yuyang Peng, Jennifer Kaiser, Leah R. Williams, Philip L. Croteau, John T. Jayne, and Nga Lee Ng*

Cite This: *Environ. Sci. Technol.* 2022, 56, 7063–7073

ABSTRACT: Advances in low-cost sensors (LCS) for monitoring air quality have opened new opportunities to characterize air quality in finer spatial and temporal resolutions. In this study, we deployed LCS that measure both gas (CO, NO, NO_2, and O_3) and particle concentrations and co-located research-grade instruments in Atlanta, GA, to investigate the capability of LCS in resolving air pollutant sources using non-negative matrix factorization (NMF) in a moderately polluted urban area. We provide a comparison of applying the NMF technique to both normalized and non-normalized data sets. We identify four factors with different temporal trends and properties for both normalized and non-normalized data sets. Both normalized and non-normalized LCS data

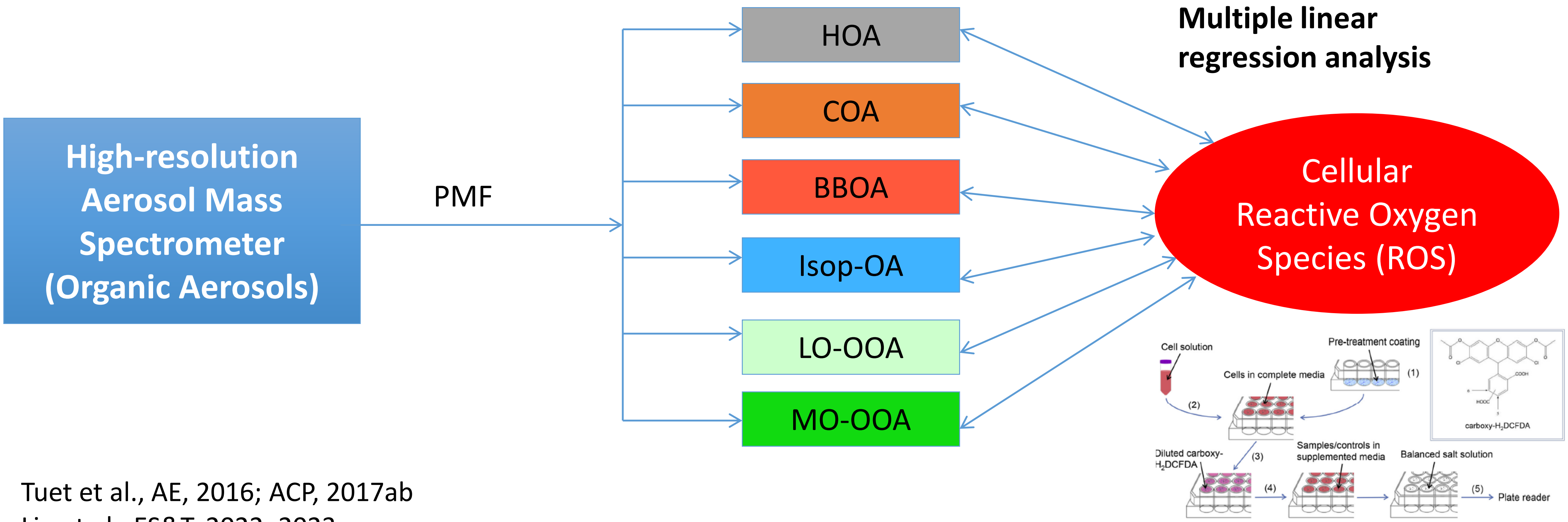




Toxicity of Different $PM_{2.5}$ Components (Organic Aerosols)

https://www.verywellhealth.com/what-are-macrophages-200997

Linking Organic Aerosol Subtypes to Toxicity



Tuet et al., AE, 2016; ACP, 2017ab
Liu et al., ES&T, 2022, 2023

HOA: Hydrocarbon-like OA; COA: Cooking OA; BBOA: Biomass burning OA; Isop-OA: Isoprene OA
LO-OOA: Less-oxidized oxygenated OA; MO-OOA: More-oxidized oxygenated OA → these are surrogates of SOA

Toxicity of Different Organic Aerosol Subtypes

Environmental Science & Technology

pubs.acs.org/est

Article

Oxidized and Unsaturated: Key Organic Aerosol Traits Associated with Cellular Reactive Oxygen Species Production in the Southeastern United States

Fobang Liu, Taekyu Joo, Jenna C. Ditto, Maria G. Saavedra, Masayuki Takeuchi, Alexandra J. Boris, Yuhan Yang, Rodney J. Weber, Ann M. Dillner, Drew R. Gentner, and Nga L. Ng*

Cite This: *Environ. Sci. Technol.* 2023, 57, 14150–14161

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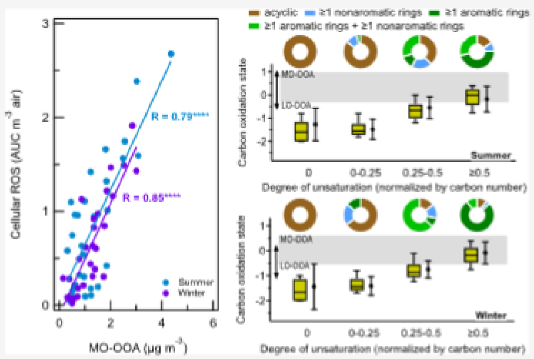
ABSTRACT: Exposure to ambient fine particulate matter ($PM_{2.5}$) is associated with millions of premature deaths annually. Oxidative stress through overproduction of reactive oxygen species (ROS) is a possible mechanism for $PM_{2.5}$-induced health effects. Organic aerosol (OA) is a dominant component of $PM_{2.5}$ worldwide, yet its role in $PM_{2.5}$ toxicity is poorly understood due to its chemical complexity. Here, through integrated cellular ROS measurements and detailed multi-instrument chemical characterization of PM in urban southeastern United States, we show that oxygenated OA (OOA), especially more-oxidized OOA, is the main OA type associated with cellular ROS production. We further reveal that highly unsaturated species containing carbon–oxygen double bonds and aromatic rings in OOA are major contributors to cellular ROS production. These results highlight the key chemical features of ambient OA driving its toxicity. As more-oxidized OOA is ubiquitous and abundant in the atmosphere, this emphasizes the need to understand its sources and chemical processing when formulating effective strategies to mitigate $PM_{2.5}$ health impacts.



KEYWORDS: *organic aerosol, secondary organic aerosol, oxygenated organic aerosol, reactive oxygen species, toxicity, aerosol health effects, aerosol mass spectrometer*

- MO-OOA (more-oxidized oxygenated OA) is the most toxic OA subtype
- Toxicity of OOA could arise from **aged** biomass burning aerosols / combustion-related aerosols

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https://ascent.research.gatech.edu

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Principal Investigator: Nga Lee (Sally) Ng, Georgia Institute of Technology

Co-PIs: Ann Dillner, University of California Davis; Roya Bahreini, University of California Riverside; Ted Russell, Georgia Institute of Technology

- First, long-term, ground-based, high time-resolution aerosol measurement network in the United States
- 12 Sites; urban, rural, remote sites
- Process level understanding of aerosols
- New opportunity in understanding aerosol sources, composition, properties, climate and health impacts across scales; model development; satellite validation

Mid-scale Research Infrastructure Program

NSF

ASCENT Site Map

Leverage Existing Sites/Infrastructure

NCore: National Core Network

- $PM_{2.5}$ mass and speciation; O_3, CO, SO_2, NO, and NO_y

PAMS: Photochemical Assessment Monitoring Stations, VOCs

IMPROVE: Interagency Monitoring of PROtected Visual Environment network

- $PM_{2.5}$ mass; gas-phase measurements

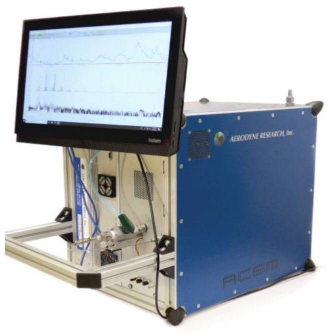
SCAQMD: South Coast Air Quality Management District

NEON: National Ecological Observatory Network

HNET: Houston Network of Environmental Towers

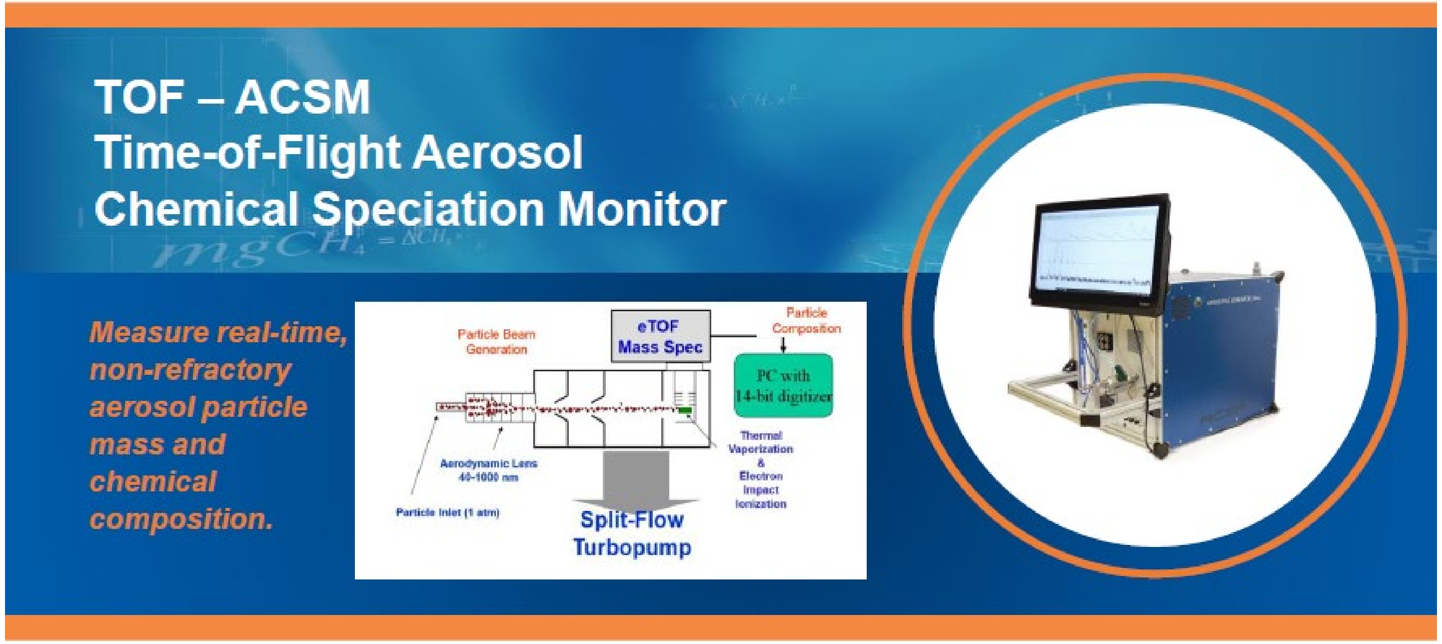
ASCENT Instrumentation

Instrument	Model and Manufacturer	Measurements
Aerosol Chemical Speciation Monitor (ACSM), $PM_{2.5}$	ToF-ACSM, Aerodyne Research	Organics, sulfate, nitrate, ammonium, chloride
Xact, $PM_{2.5}$	625i, Cooper Environmental	Trace metals: Sb, As, Ba, Cd, Ca Cr, Co, Cu, Fe, Pb, Hg, Mn, Ni, Se, Ag, Sn, Ti, Tl, V, Zn, more available
Aethalometer, $PM_{2.5}$	AE33, Magee Scientific	Wavelength-dependent absorption; black and brown carbon
Scanning Mobility Particle Sizer (SMPS), PM_1	3938W89, TSI	Particle number size distribution, number concentration



Aerosol Chemical Speciation Monitor (ACSM)

- Quantitative, continuous measurement of submicron non-refractory aerosols
  - Organics, Sulfate, Nitrate, Ammonium, Chloride
- Built upon the same technology as the widely used Aerodyne Aerosol Mass Spectrometer (AMS)
- Designed to be smaller, lower cost, and simpler to operate than the AMS
- Designed for long-term, autonomous, and stable field measurements of ambient aerosols



Aerosol Science and Technology, 45:780–794, 2011
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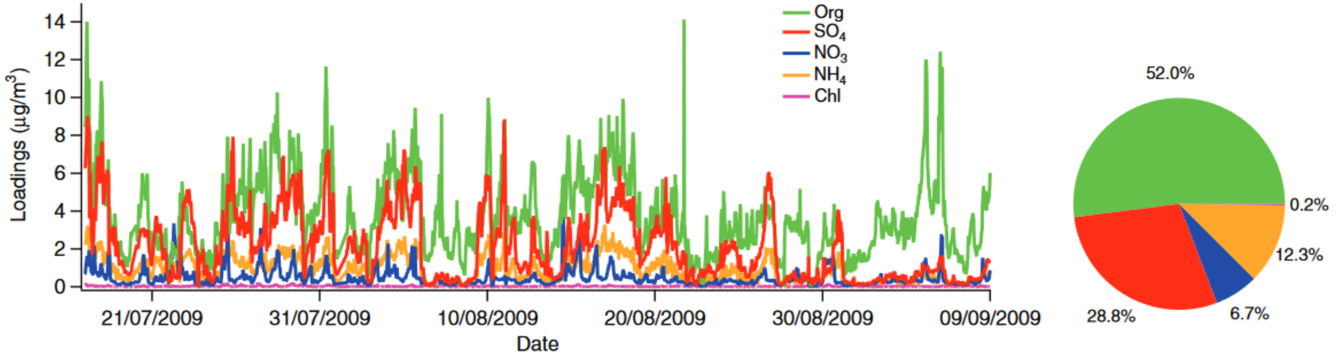
ISSN: 0278-6826 print / 1521-7388 online
DOI: 10.1080/02786826.2011.560211

An Aerosol Chemical Speciation Monitor (ACSM) for Routine Monitoring of the Composition and Mass Concentrations of Ambient Aerosol

N. L. Ng,[1] S. C. Herndon,[1] A. Trimborn,[1] M. R. Canagaratna,[1] P. L. Croteau,[1] T. B. Onasch,[1] D. Sueper,[1,2] D. R. Worsnop,[1] Q. Zhang,[3] Y. L. Sun,[3] and J. T. Jayne[1]
[1]*Aerodyne Research, Inc., Billerica, Massachusetts, USA*
[2]*CIRES, University of Colorado, Boulder, Colorado, USA*
[3]*Department of Environmental Toxicology, University of California, Davis, California, USA*

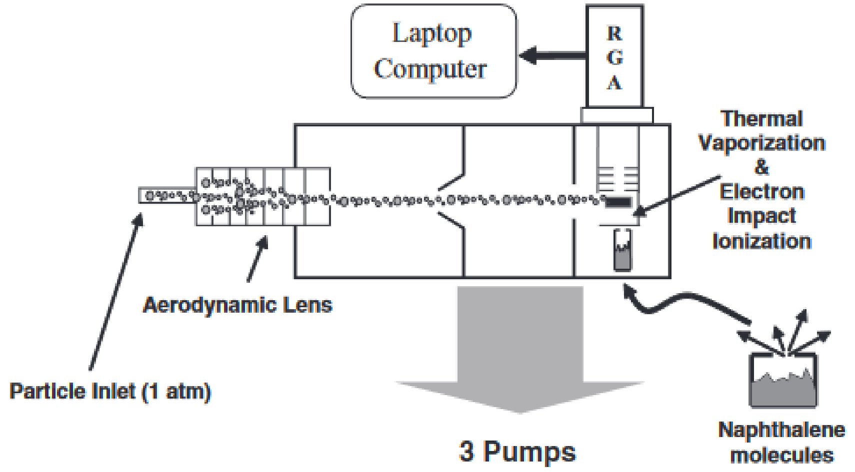


We present a new instrument, the Aerosol Chemical Speciation Monitor (ACSM), which routinely characterizes and monitors the mass and chemical composition of non-refractory submicron particulate matter in real time. Under ambient conditions, mass concentrations of particulate organics, sulfate, nitrate, ammonium, and chloride are obtained with a detection limit $<0.2\ \mu g/m^3$ for 30 min of signal averaging. The ACSM is built upon the same technology as the widely used Aerodyne Aerosol Mass Spectrometer (AMS), in which an aerodynamic particle focusing lens is combined with high vacuum thermal particle vaporization, electron impact ionization, and mass spectrometry. Modifications in the ACSM design, however, allow it to be smaller, lower cost, and simpler to operate than the AMS. The ACSM is also capable of routine stable operation for long periods of time (months). Results from a field measurement campaign in Queens, NY where the ACSM operated unattended and continuously for 8 weeks, are presented. ACSM data is analyzed with the same well-developed techniques that are used for the AMS. Trends in the ACSM mass concentrations observed during the Queens, NY study compare well with those from co-located instruments. Positive Matrix Factorization (PMF) of the ACSM organic aerosol spectra extracts two components: hydrocarbon-like organic aerosol (HOA) and oxygenated organic aerosol (OOA). The mass spectra and time trends of both components correlate well with PMF results obtained from a co-located high resolution time-of-flight AMS instrument.

1. INTRODUCTION

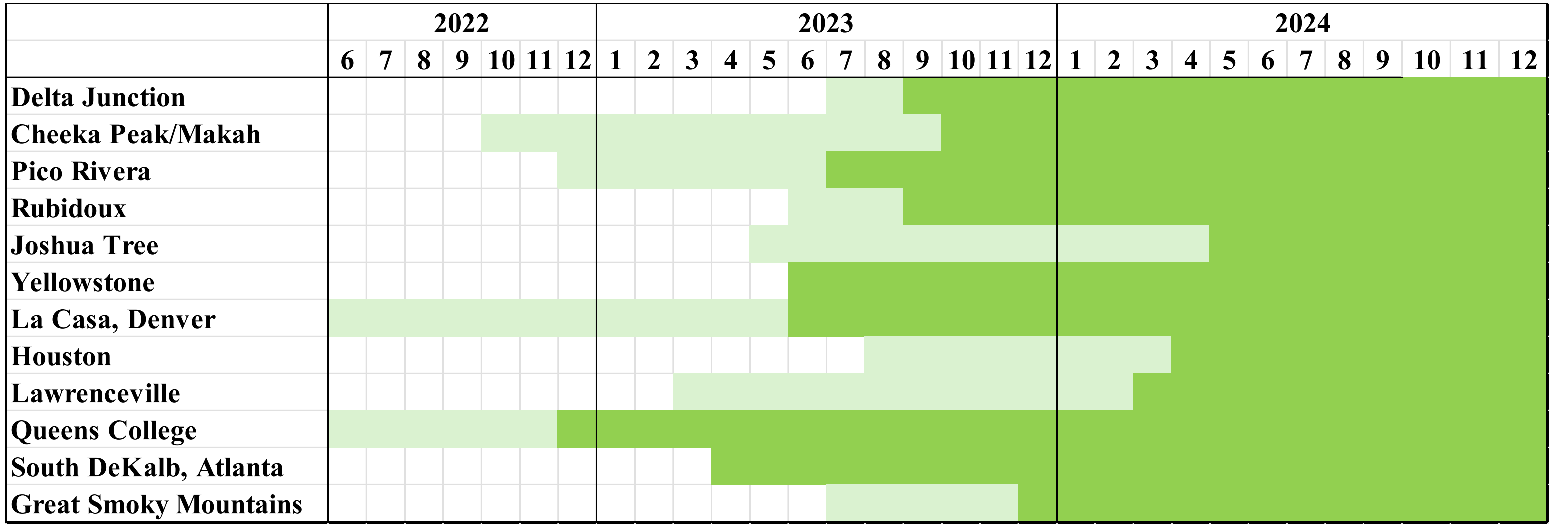
Aerosols play a significant role in altering the chemistry and the radiative balance of the Earth's atmosphere, in reducing visibility, and in adversely affecting human health (Pöschl 2005; Pope and Dockery 2006; IPCC 2007). In order to address aerosol effects on the environment and health, instrumentation capable of reporting the chemical and microphysical properties of ambient particles is needed. From an air quality monitoring standpoint, aerosol instrumentation that is simple to operate, capable of long-term, autonomous, and stable operation with real time results is also desired.

In recent years the Aerodyne Aerosol Mass Spectrometer (AMS) (Jayne et al. 2000; Canagaratna et al. 2007) equipped with quadrupole (Q-AMS) and time-of-flight (C-ToF-AMS and HR-ToF-AMS) (Drewnick et al. 2005; DeCarlo et al. 2006) mass spectrometers has been deployed in numerous field campaigns

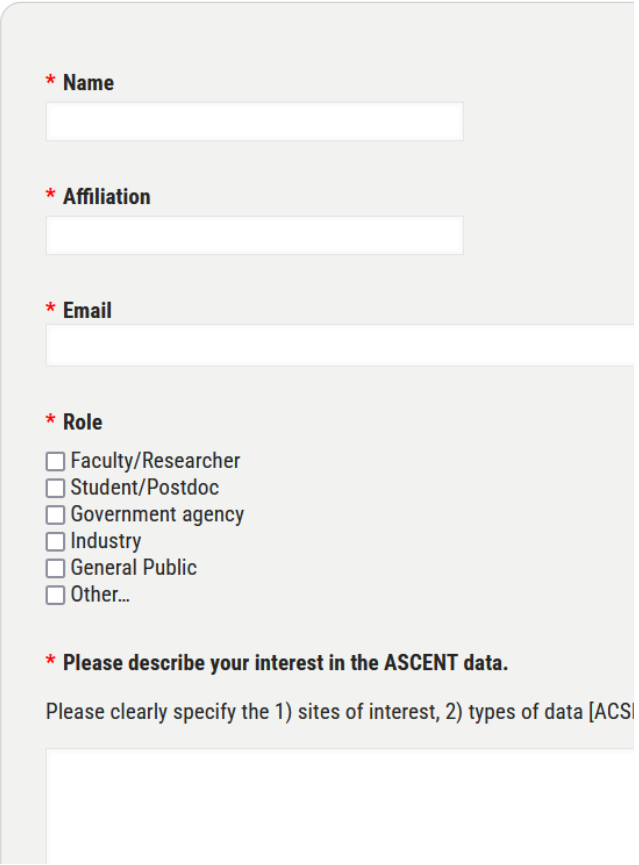


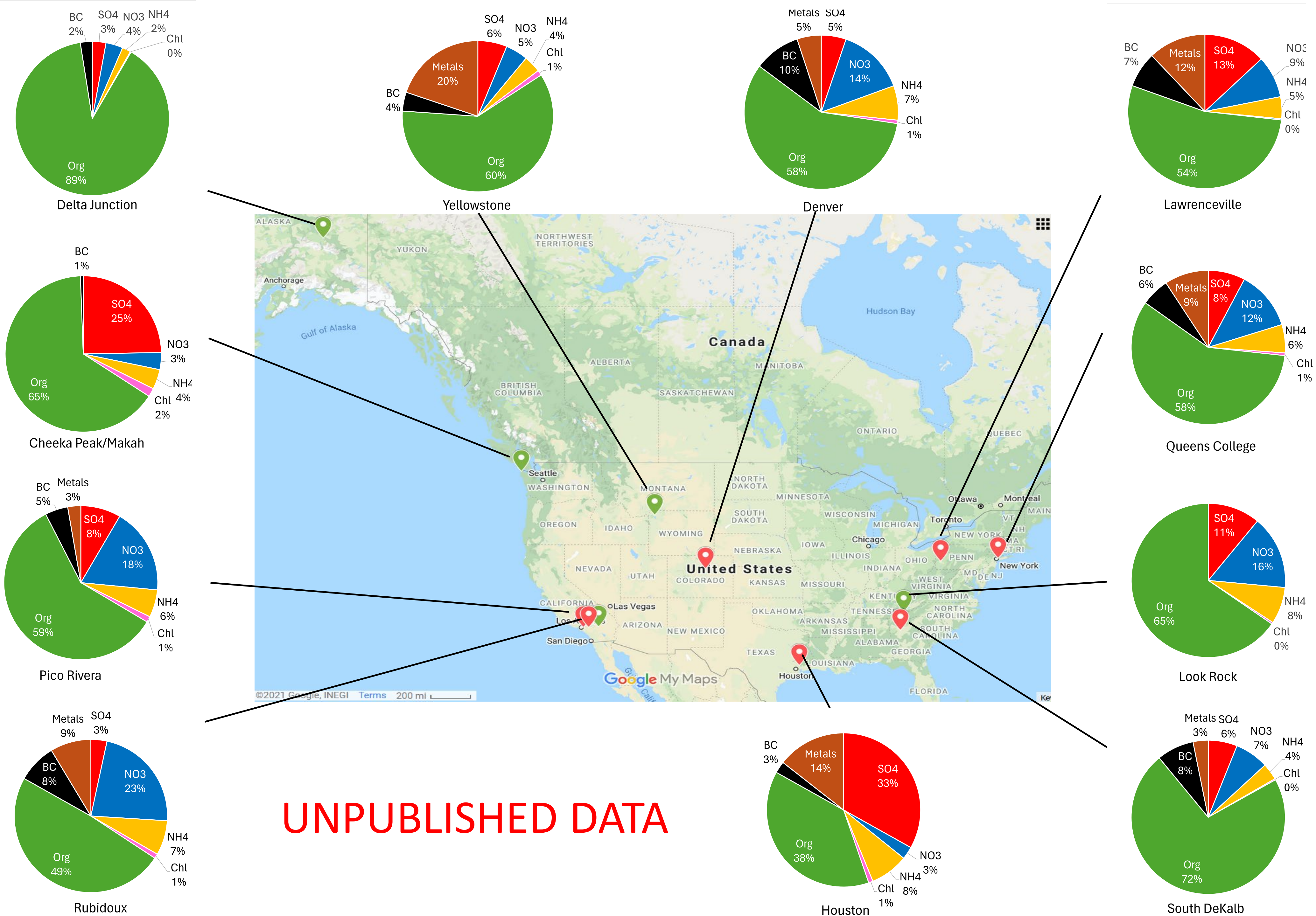
Timeline

- Project funded in October 2021
- All instruments delivered to universities in January 2023
- Sampling started at all sites in May 2024



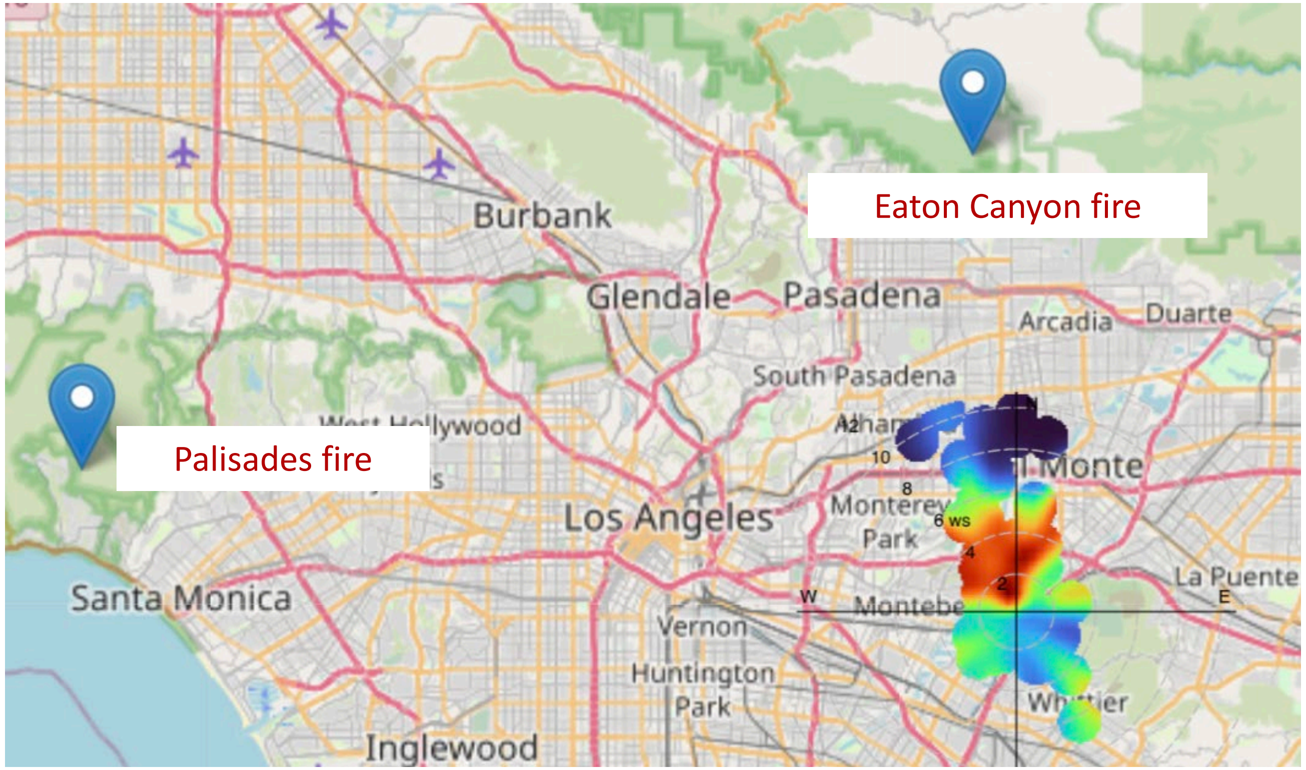
Request preliminary data
→ ASCENT website → Database





UNPUBLISHED DATA

Los Angeles Fires



ASCENT LA Site at Pico Rivera

~14 miles south of Eaton Fire

The New York Times

By Hiroko Tabuchi and Mira Rojanasakul

Jan. 20, 2025

Airborne Lead and Chlorine Levels Soared as L.A. Wildfires Raged

The findings give new insight into the dangers of urban wildfires that burn plastics and other chemicals in homes and property.

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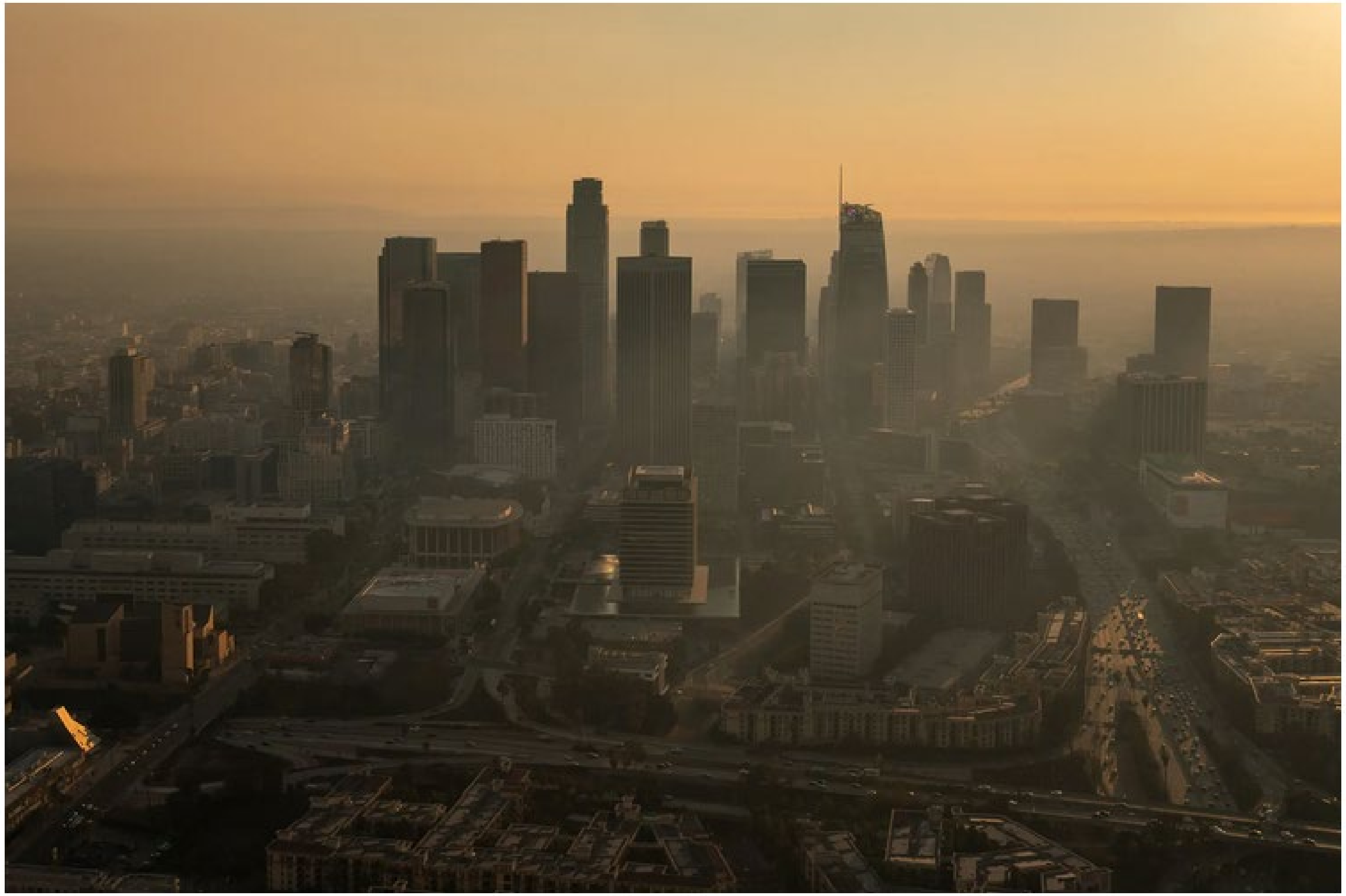

Researchers used a new network of sensors to track chemicals in the Los Angeles air in real time. Loren Elliott for The New York Times

Notes from the Field: Elevated Atmospheric Lead Levels During the Los Angeles Urban Fires — California, January 2025

Weekly / February 20, 2025 / 74(5);69–71

Print

Haroula D. Baliaka[1]; Ryan X. Ward[1]; Roya Bahreini[2]; Ann M. Dillner[3]; Armistead G. Russell[4]; John H. Seinfeld[1]; Richard C. Flagan[1]; Paul O. Wennberg[1]; Nga L. Ng[4] (VIEW AUTHOR AFFILIATIONS)

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Summary

What is already known about this topic?

Smoke is a complex mixture of gases and airborne particulate matter; urban fires and conventional wildfires emit different air pollutants. The Atmospheric Science and Chemistry mEasurement NeTwork (ASCENT), a new, advanced air quality measurement network, provides real-time measurements of the chemical constituents in fine particulate matter ($PM_{2.5}$).

What is added by this report?

During the January 2025 Los Angeles fires, ASCENT recorded an approximate 110-fold increase in $PM_{2.5}$ lead levels compared with values from the previous few days.

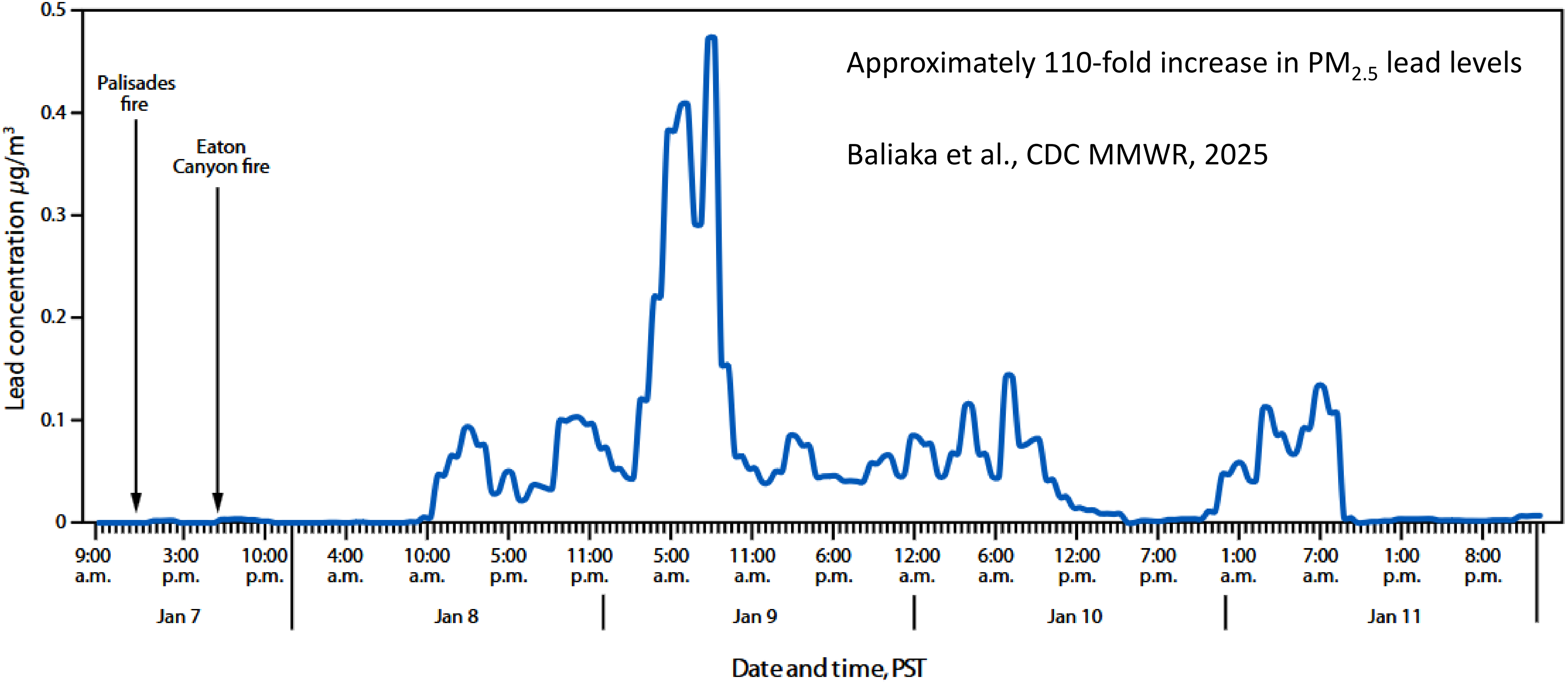
What are the implications for public health practice?

Urban fires emit air pollutants that pose risks different from those of conventional wildfires. It is important for epidemiologic studies to consider $PM_{2.5}$ composition when assessing the impacts of urban fire smoke exposure. Health officials should communicate protective measures to the public (monitor air quality forecasts and follow guidance by local emergency management officials).

Figure

Related Materials

FIGURE. Hourly lead concentrations*,† of particulate matter <2.5 μm in diameter at the Los Angeles Atmospheric Science and Chemistry mEasurement NeTwork site relative to the start of the Palisades and Eaton Canyon fires — Pico Rivera, California, January 7–12, 2025



Approximately 110-fold increase in $PM_{2.5}$ lead levels

Baliaka et al., CDC MMWR, 2025

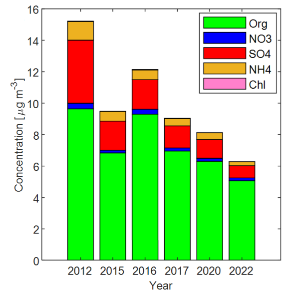
* μg/m3.
† The National Ambient Air Quality Standard for lead in total suspended particles over a 3-month rolling average is 0.15 μg/m3.

ASCENT Website → Database
Real-time lead, chlorine, bromine data from the three ASCENT sites in CA

Summary; Looking Forward

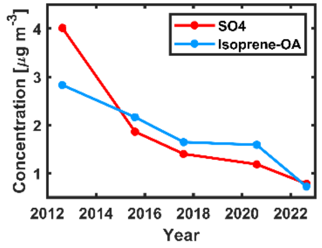
Dominance of Organic Aerosols in $PM_{2.5}$

- Increasing dominance of OA, especially SOA
- Critical to understand sources and properties of OA and particularly SOA to control $PM_{2.5}$



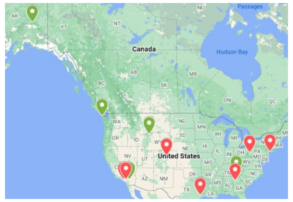
Biogenic/Anthropogenic interactions

- Reducing SO_2 and NO_2 are effective measures to control aerosol formation from biogenic emissions (natural tree emissions)



ASCENT
Atmospheric Science and Chemistry mEasurement NeTwork

- High-time resolution; process level understanding
- Air quality, climate; model development; satellite validation
- Health effects: composition, particle size



Thank you for your attention!

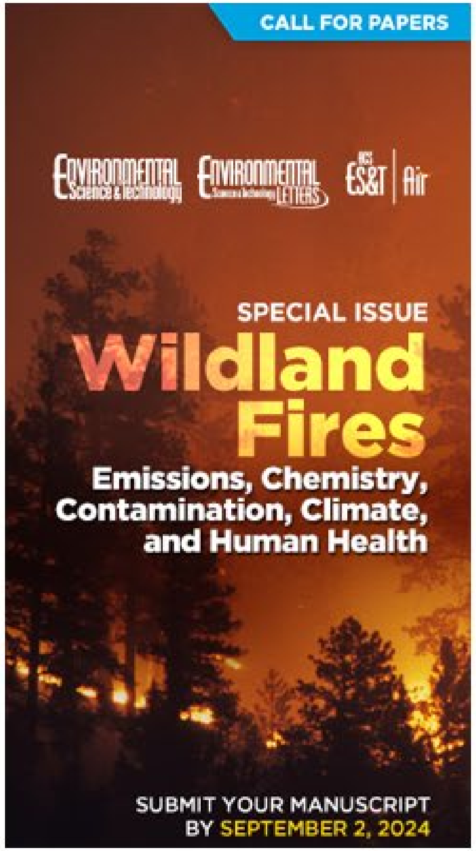
NSF

NATIONAL OCEANIC AND ATMOSPHERIC ADMINISTRATION
NOAA
U.S. DEPARTMENT OF COMMERCE

UNITED STATES ENVIRONMENTAL PROTECTION AGENCY

Special Issues

ACS ES&T | Air



(Just Published)

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Amy Sullivan
Amara Holder

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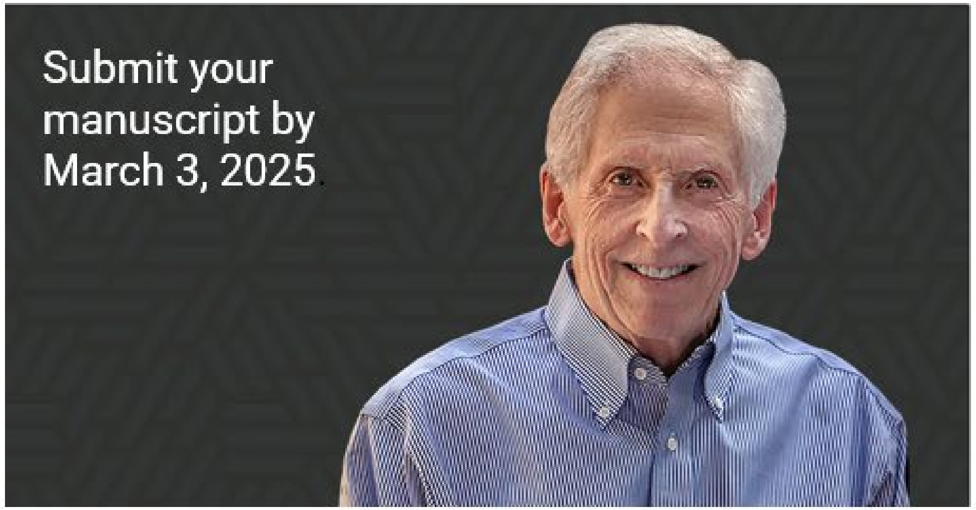

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