

Instrument Team Presentations: Organic Gases

Moderator: Jessica Gilman, NOAA CSL

September 28, 2022

14:00 to 15:00

AEROMMA

Organic Gases

Formaldehyde Measurements on Two Planes

AEROMMA



Jen Kaiser
GA Tech



Nidhi Desai
GA Tech

GOTHAAM



Reem Hannun
U. Pitt.



Abby Sebol
U. MD



Tim Canty
U. MD

The Help



Glenn Wolfe
NASA GSFC



Jason St. Clair
UMBC



Tom Hanisco
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Andrew Swanson
UMBC

Contact: glenn.m.wolfe@nasa.gov

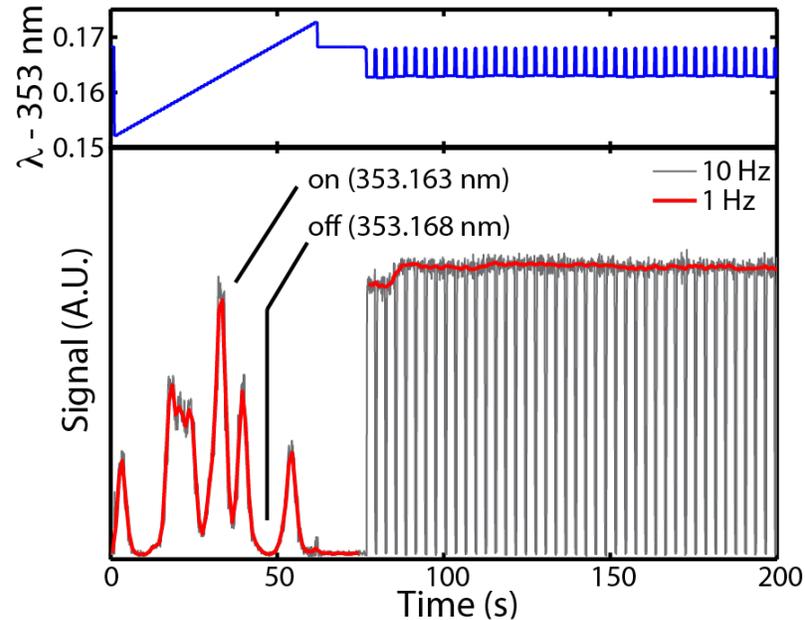
In Situ Airborne Formaldehyde (ISAF)

Technique: laser-induced fluorescence (LIF)

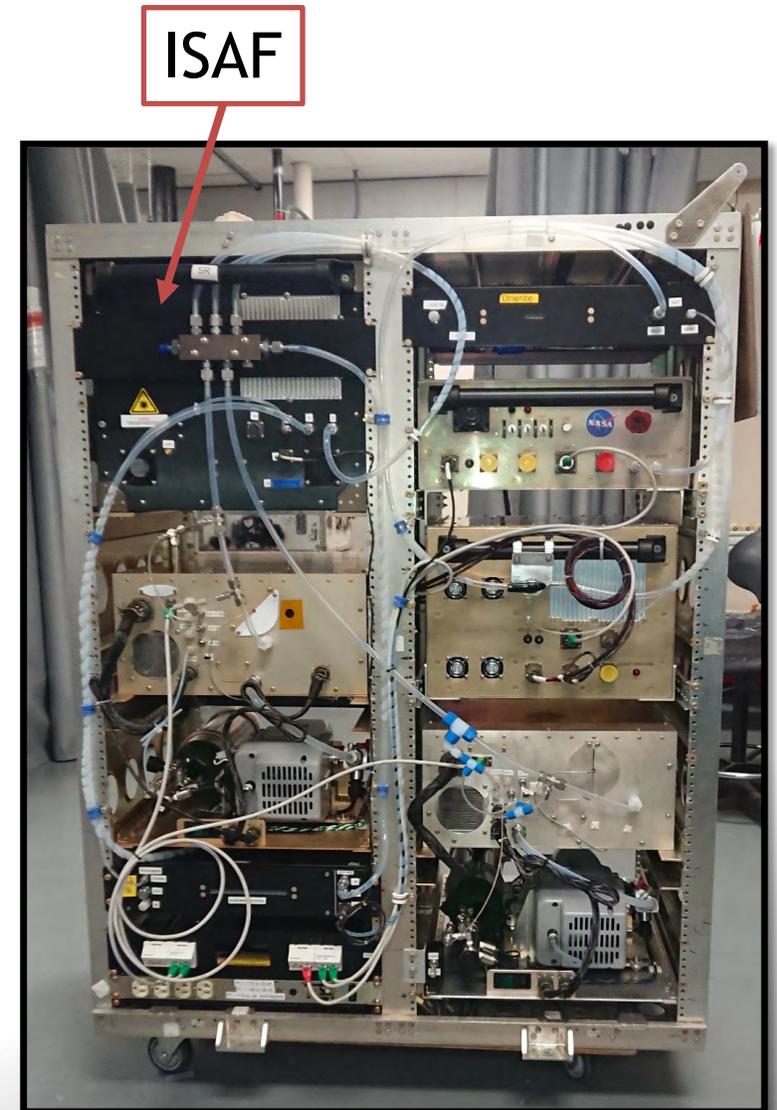
A laser is continuously tuned between a large formaldehyde (HCHO) rotational transition and a non-resonant wavelength.

The concentration of HCHO is proportional to the difference between the online and the offline signals.

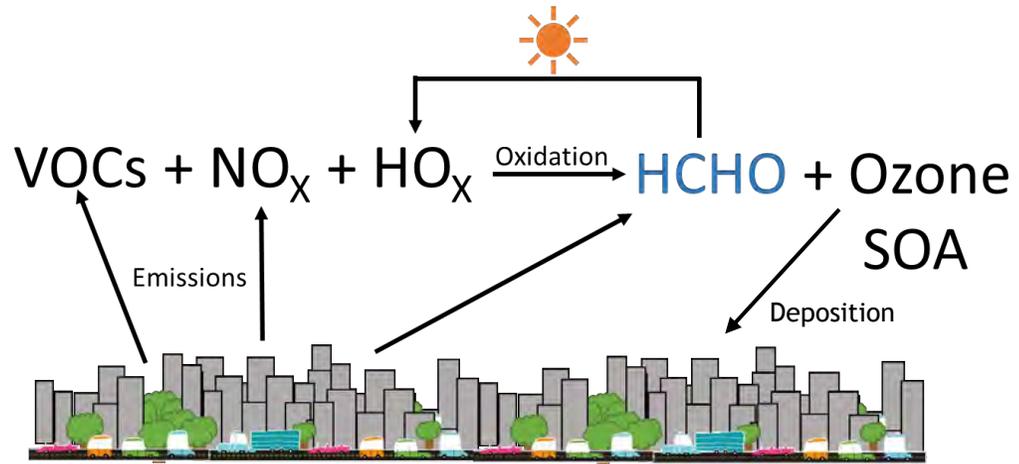
Reference: Cazorla et al., Atmos. Meas. Tech., 8(2), (2015).



Detection Limit: 35 pptv/s
Accuracy: 10%
Time Resolution: up to 5 Hz

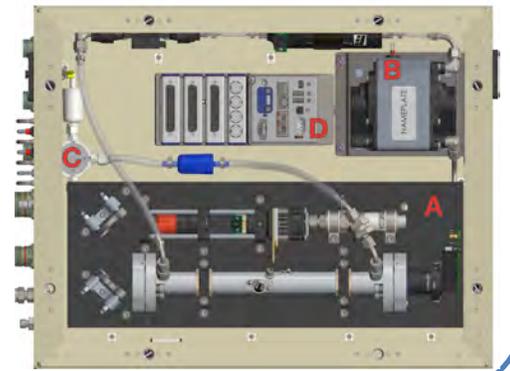


Fun Things to do with HCHO and Friends

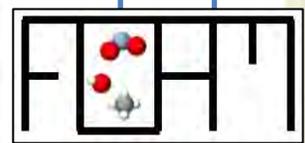
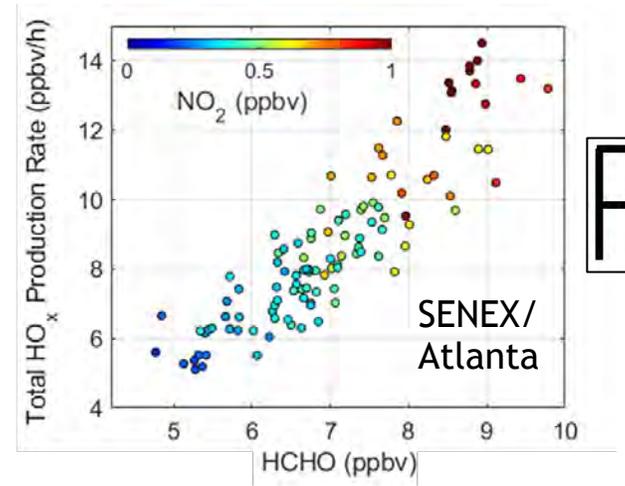


Ozone Deposition to Urban Surfaces

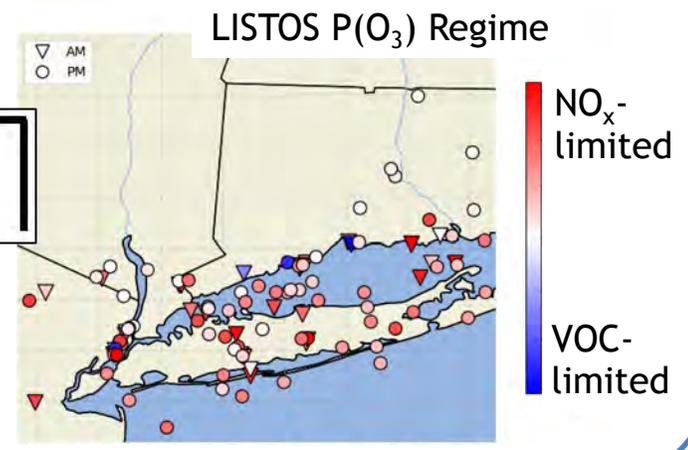
Rapid Ozone Experiment (ROZE)
 Hannun et al., AMT (2020)



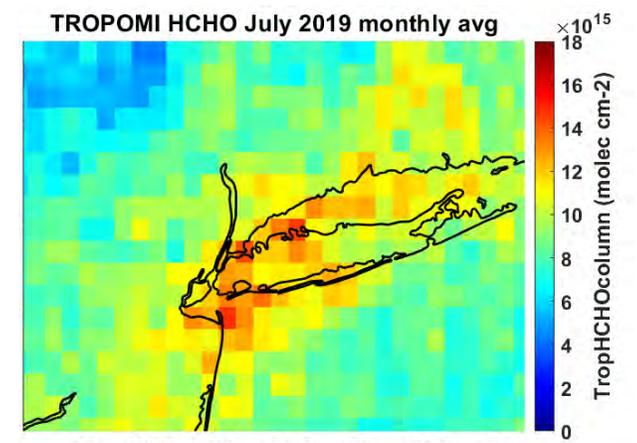
VOC Oxidation



Ozone Production



Satellite Retrieval Validation and Upscaling



ACES (Airborne Cavity Enhanced Spectrometer)

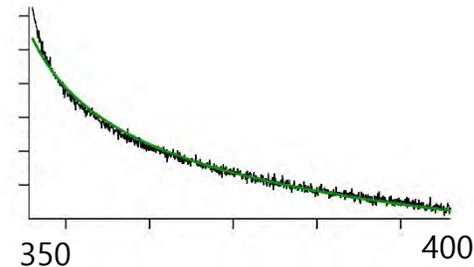
Technique: 2-channel UV-vis absorption spectrometer

Species measured: NO₂, glyoxal (CHOCHO), and total aerosol extinction (NEW)

2σ LOD (1 sec): 74 pptv (NO₂), 40 pptv (CHOCHO), 1.1 Mm⁻¹ at 365 nm (aerosol extinction)

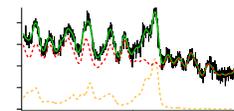


Channel 1:
UV aerosol



Measured spectrum
Fit, attributed to aerosol

Channel 2:
Vis gas-phase



Measured spectrum
Fit
0.5 ppbv NO₂
0.5 ppbv CHOCHO

435

480

NOAA / CIRES ACES team

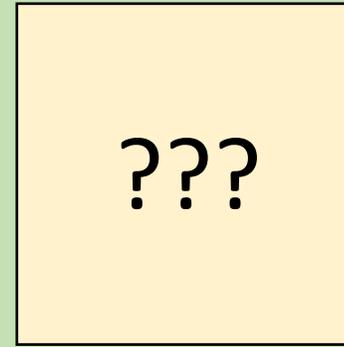


Carrie Womack
ACES PI

caroline.womack@noaa.gov



Wyndom Chace
NightNOx



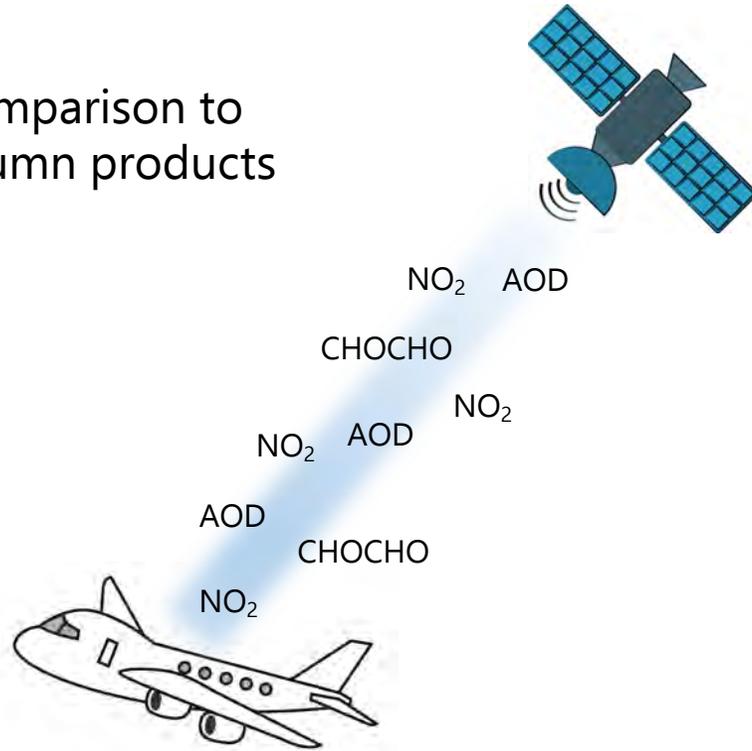
Additional
summer student?



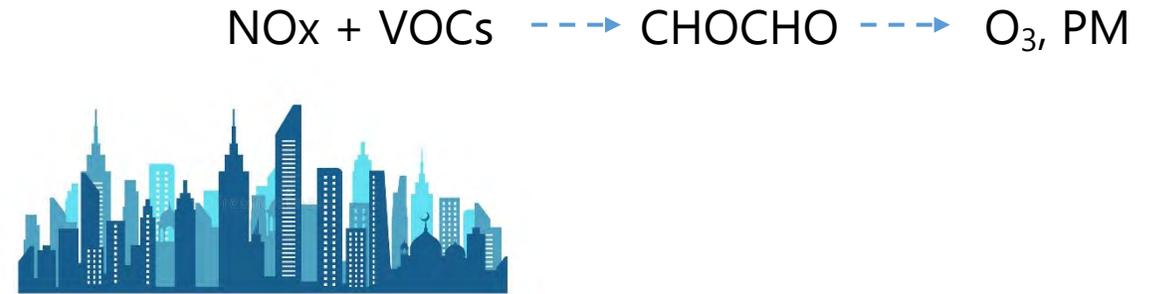
Steve Brown

ACES science goals during AEROMMA

1. Direct comparison to TEMPO column products

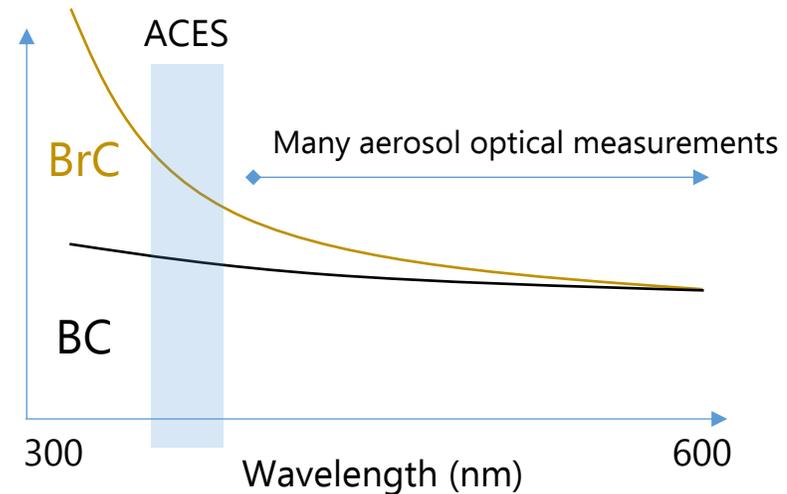


2. Role of NO_x and small VOCs (i.e. glyoxal) in ozone formation



3. Aerosol extinction in the UV \rightarrow role of BrC

Aerosol absorption

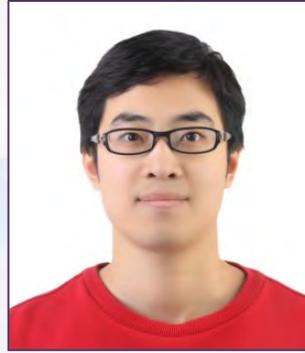




NOAA VOCUS PTR-ToF-MS on NASA DC8 measuring VOCs



Chelsea Stockwell
Instrument PI
CIRES/NOAA



Lu Xu
Instrument PI
CIRES/NOAA



Georgios Gkatzelis
Visiting Scientist
Julich



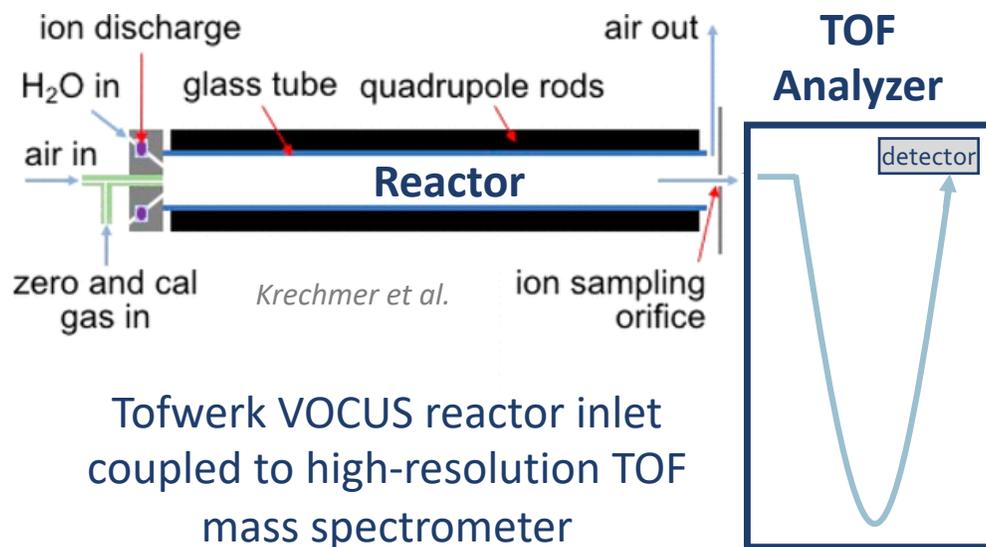
Matt Coggon
Instrument PI
NOAA CSL



Carsten Warneke
Team Leader
NOAA CSL

Contact: matthew.m.coggon@noaa.gov

NOAA VOCUS PTR-ToF-MS on NASA DC8 measuring VOCs



Targeted Species

Biogenic: isoprene, monoterpenes

Oxygenates : alcohols, ketones, aldehydes

Aromatics: benzene, toluene, C8-C10 aromatics

Furanoids: furan, methyl furan, furfural

VCPs: D5-siloxane, PCBTF, texanol

Instrument Performance

Time resolution: up to 10 Hz

Detection limits: <50 ppt for 1s

Precision: signal dependent

Accuracy: 30-100%

Mass resolution: up to 5000 m/ Δ m

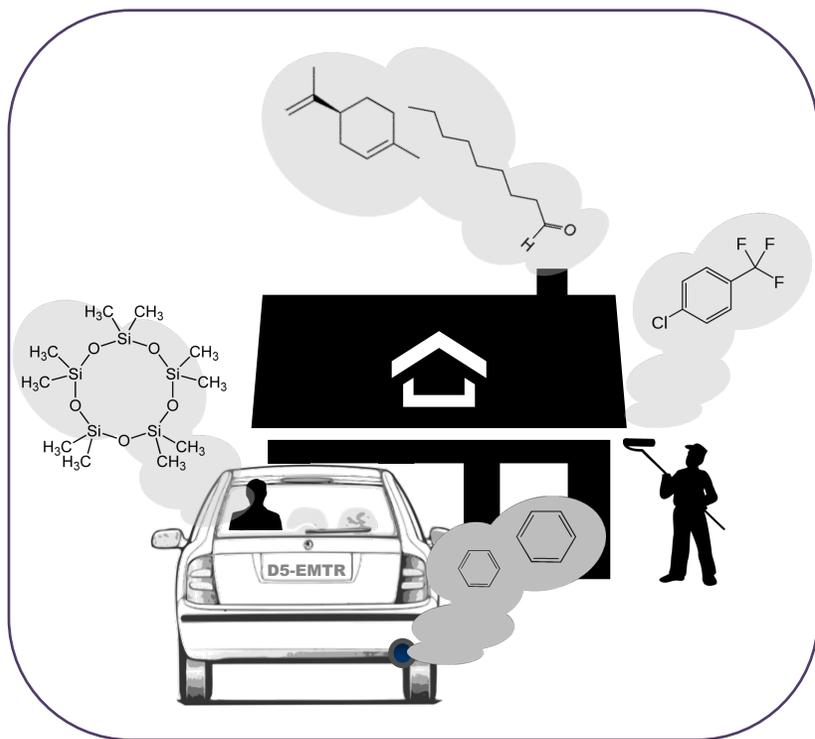
Key references for Instrument Capabilities

Krechmer et al. (2018), *An. Chem.*, DOI: [10.1021/acs.analchem.8b02641](https://doi.org/10.1021/acs.analchem.8b02641)

Gkatzelis et al. (2021), *ES&T*, DOI: [10.1021/acs.est.0c05467](https://doi.org/10.1021/acs.est.0c05467)

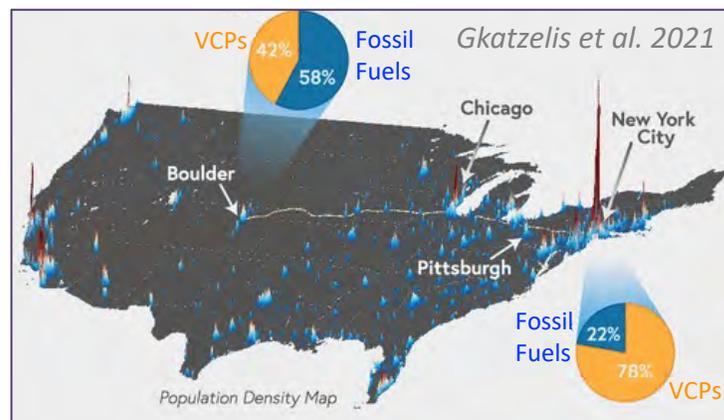


Science Goals and Foci

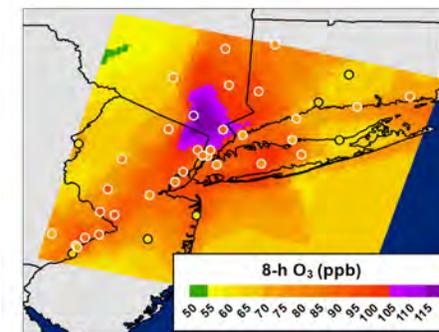


Use measurements to identify and quantify VOC source profiles

Work with modeling groups to evaluate VOC emission inventories across cities

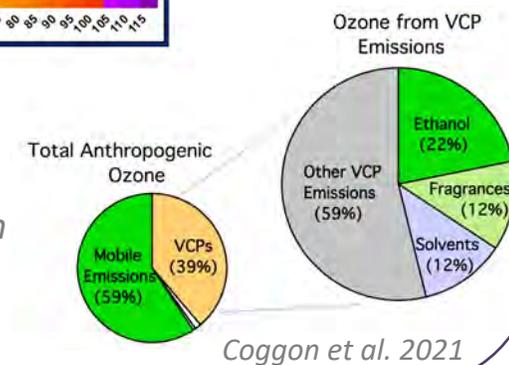


Work by Gkatzelis et al. show differences in VOC mixtures across cities of different population densities



Work with modeling groups to evaluate ozone production in major cities

Determine contribution of different emission sectors to ozone formation



Evaluate VOC chemistry and incorporate observations into chemically explicit models



NOAA Integrated Whole Air Sampling System (iWAS) for speciated VOC measurements aboard NASA DC-8



Jessica Gilman
Instrument PI
NOAA CSL



Victoria Treadaway
Instrument co-PI
CIRES/NOAA



Colby Francoeur
Graduate Student
CIRES/NOAA



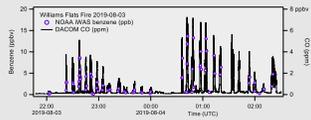
PTR-MS Team
Expert
"Can Openers"

Contact: Jessica.Gilman@noaa.gov

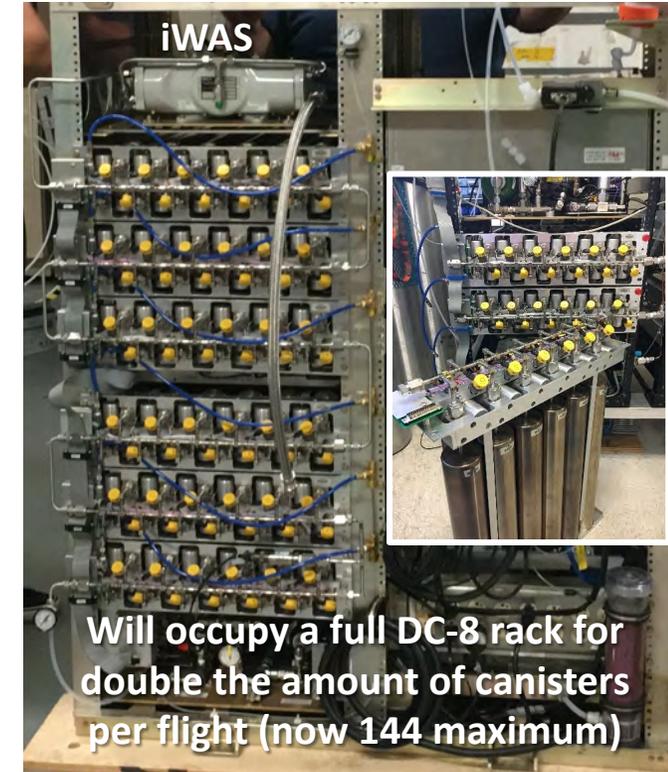
NOAA Integrated Whole Air Sampling System (iWAS) for speciated VOC measurements aboard NASA DC-8

1. Collect whole air samples

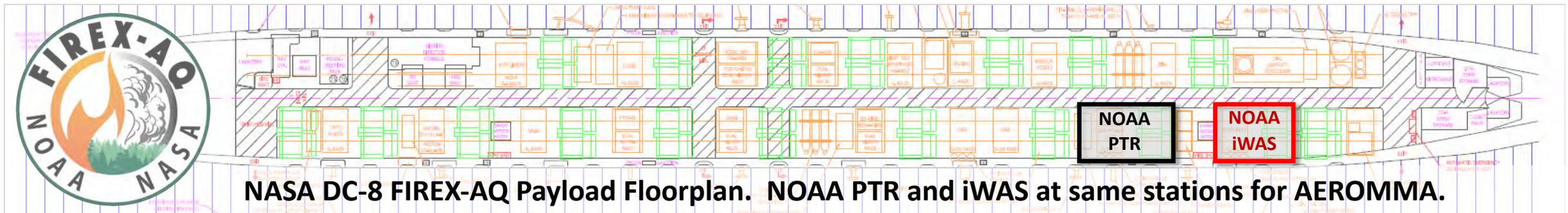
- Expanding to a maximum of **144 samples per flight!** Typical flight will collect 72 samples.
- **Fill time 3-10 seconds depending on altitude.** Filled to 50 psia via s.s. bellows compressor (>50 SLPM).
- **Computer controlled sampling:** automatic intervals or manual control for plume-specific analysis



Fast-fill, on demand sampling allowed for multiple samples to be collected across individual plume transects during FIREX-AQ. Can use real-time data from PTR and CO to determine when to collect iWAS samples.

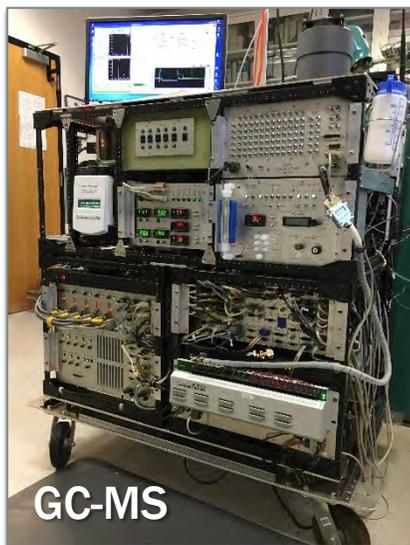


Will occupy a full DC-8 rack for double the amount of canisters per flight (now 144 maximum)



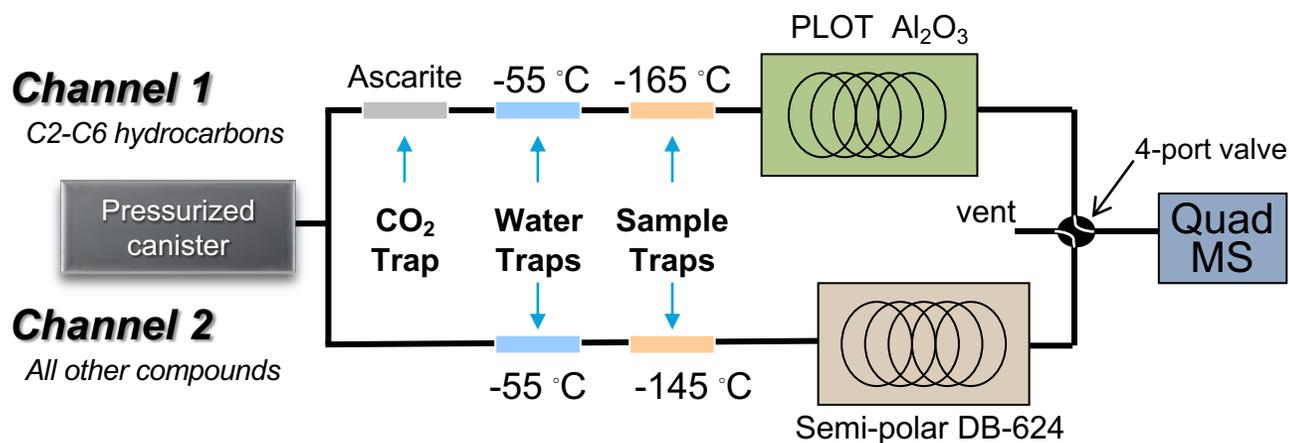
NOAA Integrated Whole Air Sampling System (iWAS) for speciated VOC measurements aboard NASA DC-8

2. Post-flight analysis via automated GC-MS



- 2-channel GC with single quadrupole MSD
- “Cryomechanical” cold traps to preconcentrate VOCs
- **Fully automated analysis; 20 min per sample**
- Most canisters analyzed with 1-4 days of collection

- C2-C10 HCs, OVOCs, N-, S-, halogenated VOCs
- iWAS/GC-MS system is designed to be deployed at the airport to minimize sample aging and shipping hassles



3. Clean and condition WAS for reuse



- Evacuate/flush with N₂ three times while heating to 80°C
- Evacuate and fill with ~10 torr water vapor

Scientific goals and areas of interest



1) Characterize and compare VOC emissions downwind of urban centers

- Emissions, transport, and chemistry
- Compare to 2019 airborne and 2021 ground-based measurements in Los Angeles

2) Identify VOC source signatures for each city

- Mobile sources, VCPs, oil and gas emissions, cooking, biogenics, biomass burning, other industrial emissions

3) Investigate the potential O₃ and SOA formation from measured VOC emissions

- Reactivities to OH, O₃, NO₃ and SOA potential

Contact: Jessica.Gilman@NOAA.gov

ENVIRONMENT AND CLIMATE CHANGE CANADA (ECCC)

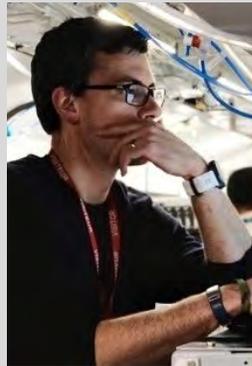
Air Quality Processes Research Section (ARQP)

NH₄⁺ and C₆H₆⁺ Vocus/AIM LToF-CIMS on NASA DC8: Measuring VOCs and OVOCs



John Liggio

Team Lead
ECCC/ARQP



Jeremy Wentzell

Instrument CO-PIs
ECCC/ARQP



Amy Leithead



Samar Moussa



Sumi Wren

Instrument operators & data analysis
ECCC/ARQP



Kathy Hayden

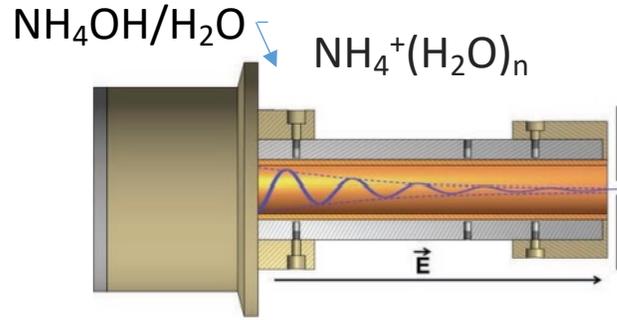


Vocus/AIM LToF-CIMS on NASA DC8

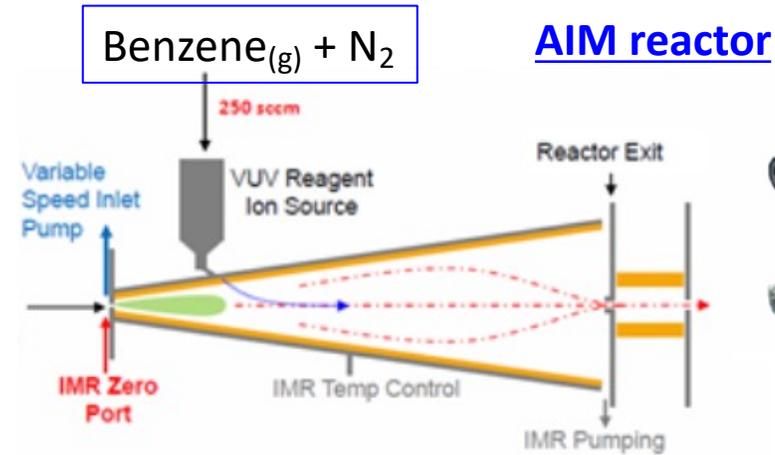
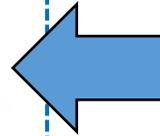
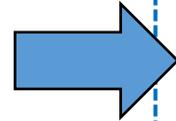


Environment and
Climate Change Canada

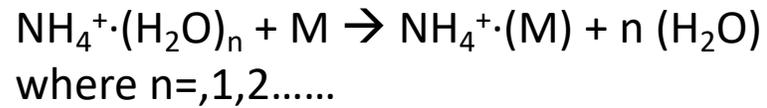
Environnement et
Changement climatique Canada



VOCUS Reactor
(www.Aerodyne.com)



(www.Aerodyne.com)



Target Species

Highly oxidized VOCs : Acids, alcohols, ketones, aldehydes, Others TBD

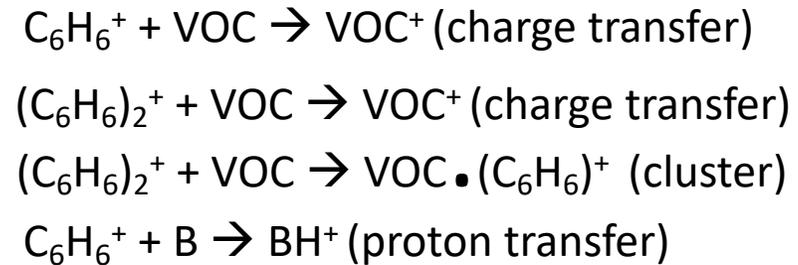
Instrument Performance

Time resolution: up to 10 Hz

Detection limits: <50 ppt for 1s (species dependent)

Accuracy: 30-100%

Mass resolution: up to 10000 m/Δm



Target Species

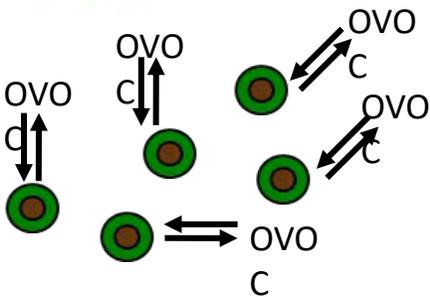
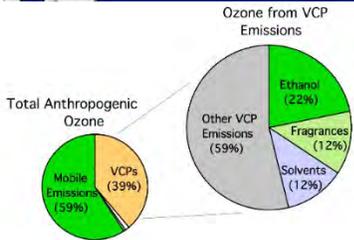
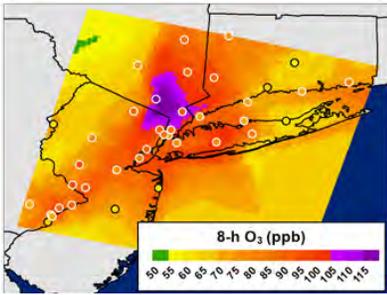
Unsaturated hydrocarbons: short & long chain olefins, mono & sesquiterpenes, aromatics, aromatic acids, others TBD

Sulfur containing compounds: DMS, Mercaptans, thiols...etc

Amines: DMA, TMA, MA, propyl amine, etc...others TBD

Currently working to unravel complex mass spectrum

Some Science objectives:



- Evaluate OVOC/VOC emissions/chemical formation and relation to ozone production
- Compare to Canadian emissions inventory and Canadian regional model (GEM-MACH) (**especially for Toronto Flights**)
- Compare emissions/chemistry with a following winter season study in Toronto (SWAPIT)
- Improve understanding of gas-particle partitioning by comparing with particle data from Vocus Inlet for Aerosols (VIA) also using NH_4^+
- Evaluate C_6H_6^+ ionization for real-time unsaturated hydrocarbon detection (compare with Whole air sample data (WAS))
- C_6H_6^+ ionization is especially sensitive for sulfur species, amines and sesquiterpenes; Urban sulfur species?? Relative importance of sesquiterpenes?? Urban reduced nitrogen & marine amine chemistry??

Caltech CIT-CIMS team on NASA DC-8 measuring inorganic acids and oVOC



Kat Ball
Graduate student

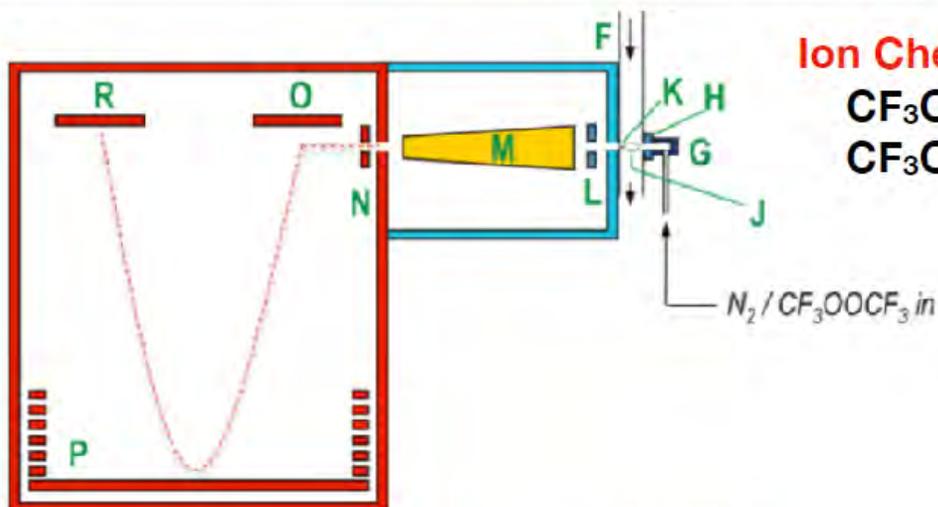


John Crouse
Research Scientist



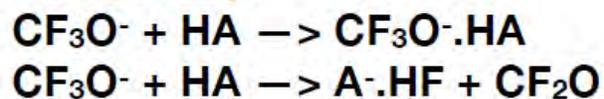
Paul Wennberg
PI

Caltech CIT-CIMS on DC-8



ToF

Ion Chemistry:

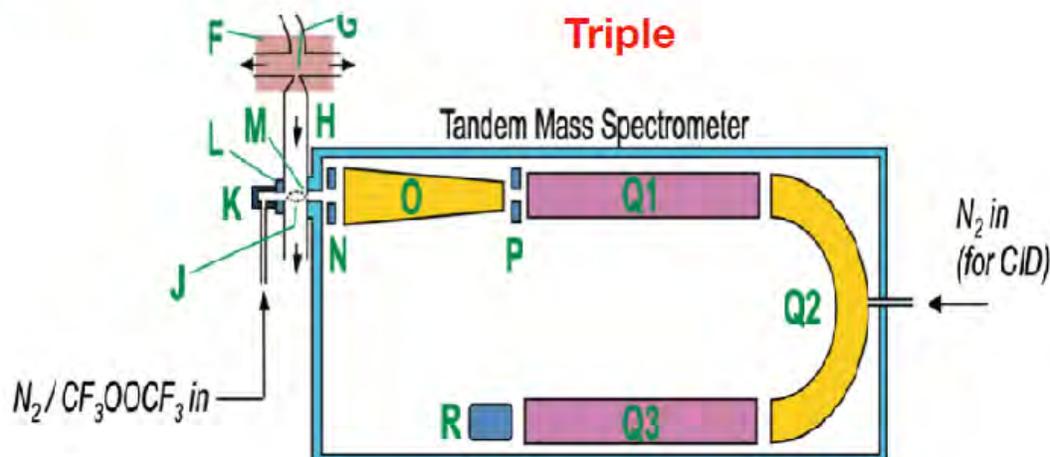


Key observations:

Inorganics: HCN, SO₂, HNO₃

Peroxides: H₂O₂, CH₃OOH, PAA

oVOC: hydroxyacetone, glycolaldehyde, isoprene epoxides, hydroxy nitrates and hydroxy peroxides from alkene oxidation, hydroperoxy carbonyls from alkane autoxidation.



Triple

ToF capabilities (upgrade to HToF):

Time resolution: 10 Hz

Mass resolution: ~3000 M/dM

Sensitivity: ~5-10 counts s⁻¹ pptv⁻¹ (most species)

Triple Quad capabilities:

Time resolution: ~1 sec every 10 sec

Mass resolution: unit

Speciation: MS/MS

Sensitivity: ~2-5 counts s⁻¹ pptv⁻¹ (most species)

References:

Crouse, et al, 2006: <https://doi.org/10.1021/ac0604235>

St. Clair, et al, 2010: <https://doi.org/10.1063/1.3480552>

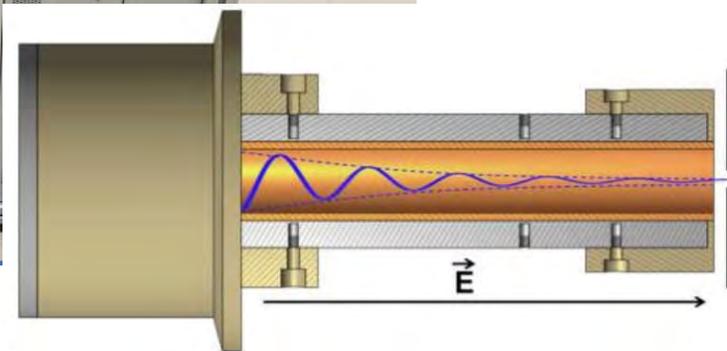
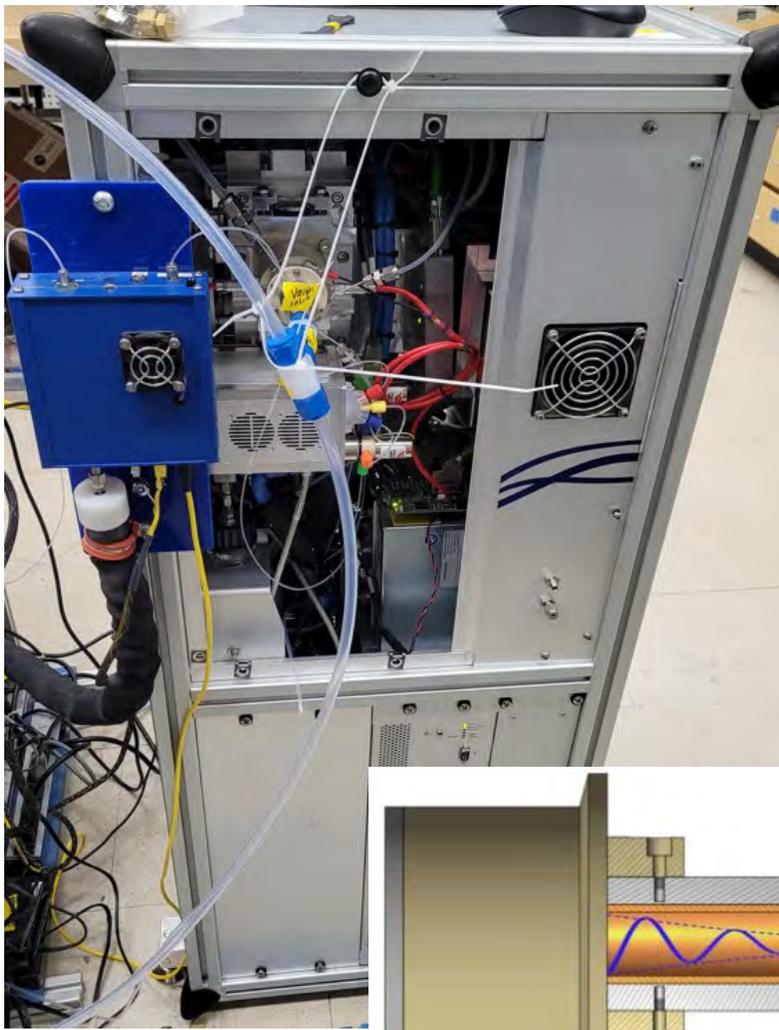
Science Goals

- Use observations and modeling to better understand the relative importance peroxy radical pathways and their contribution to formation of air pollutants across urban and marine environments**
- Use observations and modeling to understand the contributions of biogenic emissions to the formation of air pollutants across urban and marine environments**

GOTHAAM

Organic Gases

Vocus – Proton Transfer Reaction Time-of-Flight Mass Spectrometer



UW-Madison existing ground-based Vocus is currently being racked for deployment on the C-130.

Measurement:

Wide array of volatile organic compounds including (not limited to): ethanol, acetonitrile, acetone, isoprene, dimethyl sulfide, benzene, xylene, Σ monoterpenes, speciated siloxanes

Instrument Performance Metrics:

Mass resolving power: 5000 $m/\Delta m$ (HR-ToF)

Accuracy: Molecule dependent (20% for DMS)

Detection Limit: Molecule dependent (2.5 ppt for DMS @ $S/N = 3$, 10s averaging)

Time resolution: plan to operate at 5Hz

Vocus – Proton Transfer Reaction Time-of-Flight Mass Spectrometer



The owners



Subi Thakali
UW-Madison
Graduate Student

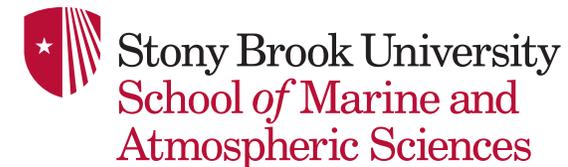


Kevin Wokosin
UW-Madison
Graduate Student



Tim Bertram
UW-Madison
Instrument PI

Operators (SBU)



John Mak

Cong Cao (PhD student)

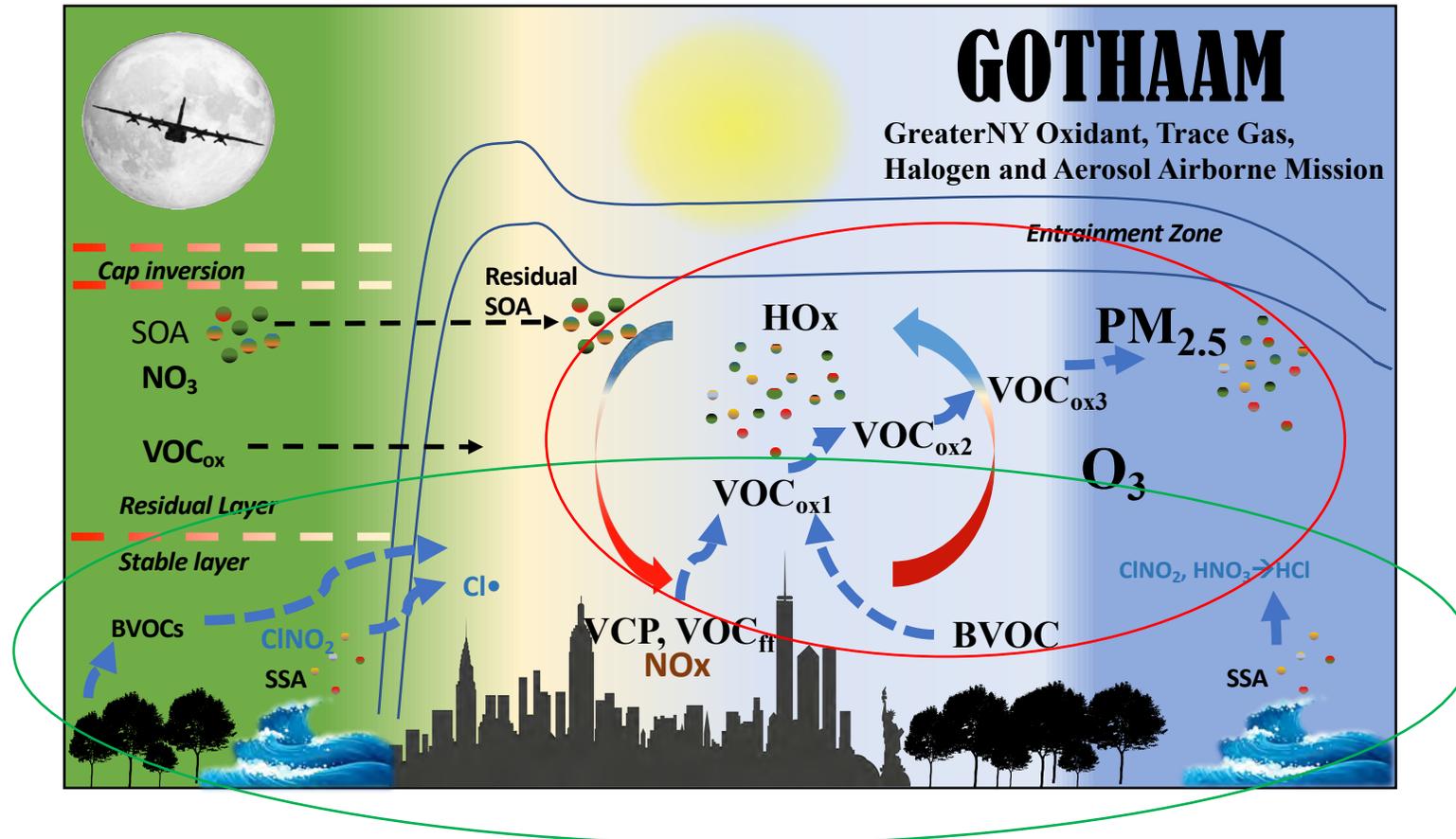
Julia Marcantonio (PhD student)

Postdoc TBD



GOTHAAM Scientific Objectives

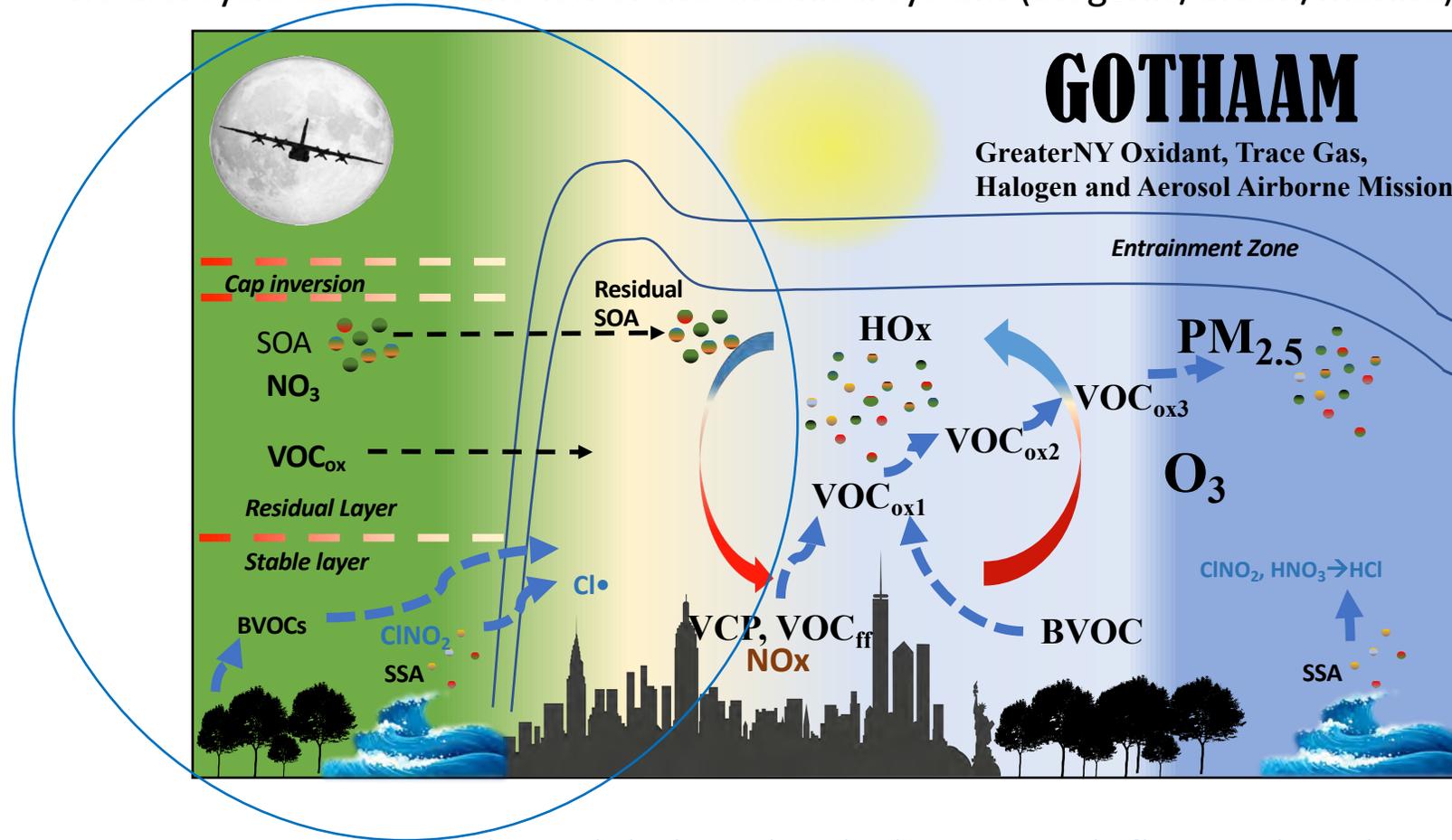
Objective 1. Quantify the relative contributions from the various volatile organic compound (VOC) sources (biogenic, fossil fuel, combustion, consumer products) and how they contribute to chemical reactivity.



Objective 2. Determine the relative potential contribution of each VOC class to secondary organic aerosol (SOA) as the anthropogenic plume evolves.

GOTHAAM Scientific Objectives

Objective 3. Quantify the relative importance of the various oxidation processes for both gas phase and aerosol species, and how the relative importance of these processes vary across the diel cycle and as a function of the chemical system (biogenic/urban/marine).



Objective 4. Investigate how nighttime chemical processes influence the subsequent day's initial chemical composition.

UW HR-ToF-CIMS on NSF C-130



Christopher Kenseth and Joel Thornton

Department of Atmospheric Sciences

W UNIVERSITY *of* WASHINGTON

September 28, 2022

ckenseth@uw.edu



UW HR-ToF-CIMS

- **NSF C-130** during **WINTER 2015** and **WE-CAN 2018**.

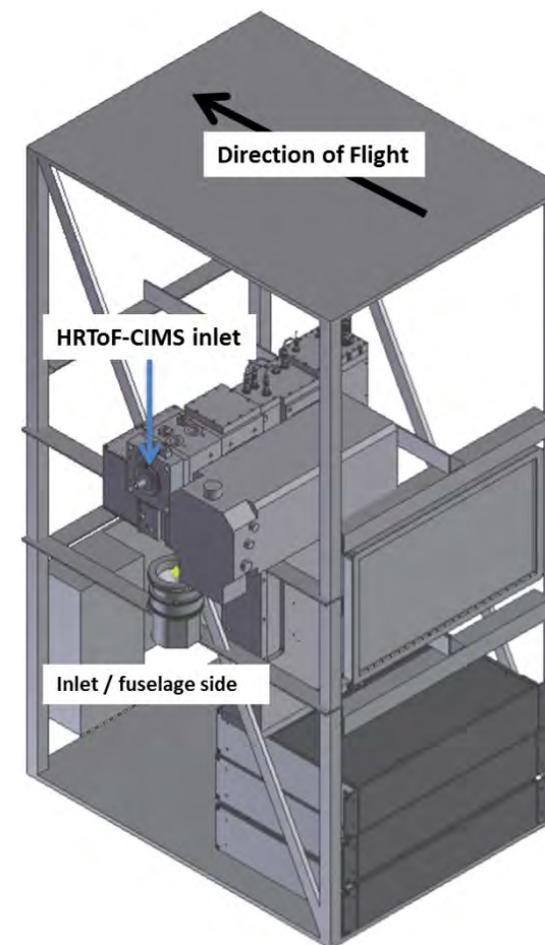
- Instrument Performance

- Time resolution: **>2 Hz**
- Mass resolution: **$m/\Delta m > 4500$**
- Mass accuracy: **<10 ppm**
- Detection limits: **<10 ppt**
- Uncertainty: **$\pm 30\text{--}50\%$**

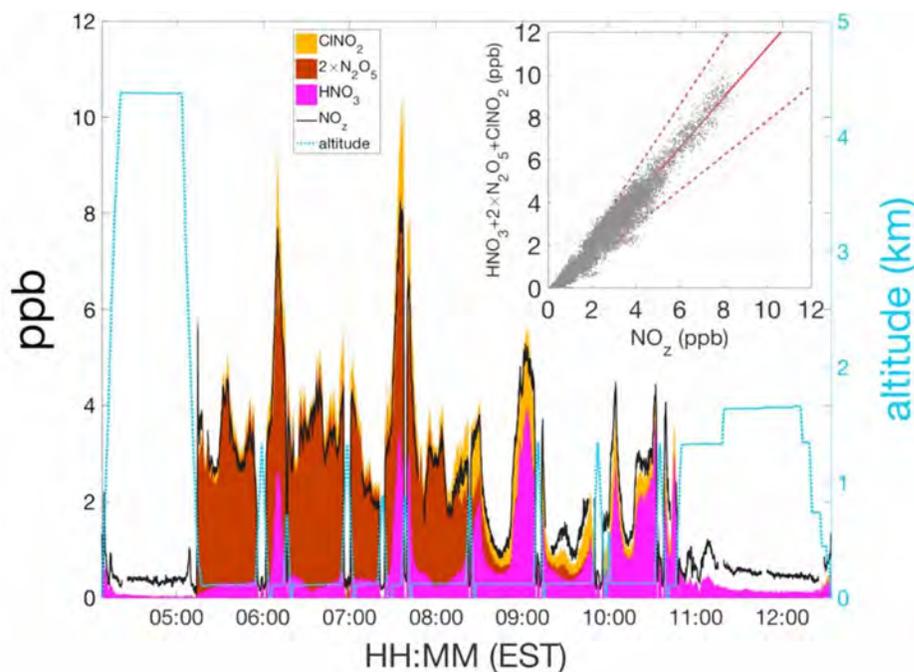
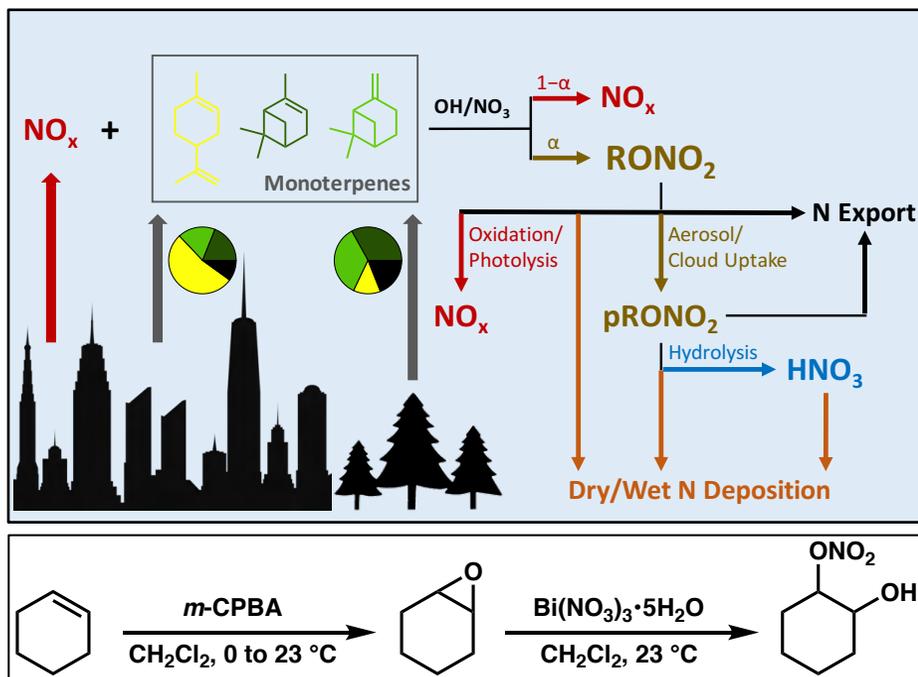
- Measured Species

- **N_2O_5** , **ClNO_2** , **HNO_3** , **HNCO** , **HONO** , **HCN** , **ClNO_3**
- **Cl_2** , **HCl** , **HOCl**
- **Br_2** , **HBr** , **BrCl** , **HOBr**
- **HCOOH** , **SO_2**
- 100s of OVOCs [**ROH** , **RC(O)OH** , **RONO_2** , **ROOH**]

Bold = Directly Calibrated



1. Lee, B.H. et al. *Environ. Sci. Technol.* **2014**, *48*, 6309.
2. Lee, B.H. et al. *J. Geophys. Res. Atmos.* **2018**, *123*, 7670.
3. Palm, B.B. et al. *Atmos. Meas. Tech.* **2019**, *12*, 5829.



- Evaluate **monoterpene RONO_2** and impact on **NO_x , O_3 , and aerosol** using **GOTHAAM** measurements, **GOES-Chem** modeling, **lab** experiments, and **organic synthesis**.
- **Identify and quantify** species that comprise **NO_z** for **NO_y budget closure**.¹
- Assess (positive) **coupling** between **NO_x , HOM, and SOA**.²

1. Lee, B.H. et al. *J. Geophys. Res. Atmos.* **2018**, 123, 7670.

2. Pye, H.O.T. et al. *Proc. Natl. Acad. Sci. U.S.A.* **2019**, 116, 6641.

TOGA-TOF - the Trace Organic Gas Analyzer with a Time-of-Flight Mass Spectrometer

PI: Eric Apel, Alan Hills, Rebecca Hornbrook (NCAR/ACOM)



NMHCs	LLOD; ppt	OVOCs	LLOD; ppt	OVOCs	LLOD; ppt	Halogenated VOCs	LLOD; ppt
Alkanes		Aldehydes		Ethers/Furans (cont'd)		CH ₃ Cl	1
Propane	5	Formaldehyde (HCHO)	20	2-Methylfuran	0.5	CH ₂ Cl ₂	1
Isobutane	1	Acetaldehyde (CH ₃ CHO)	5	3-Methylfuran	0.5	CHCl ₃	1
<i>n</i> -Butane	1	Propanal	2	2,3-Dimethylfuran	TBD	CCl ₄	1
Isopentane	1	Isobutanal	TBD	2,4-Dimethylfuran	TBD	C ₂ Cl ₄	0.1
<i>n</i> -Pentane	1	Butanal	1	2,5-Dimethylfuran	TBD	1,2-Dichloroethane	0.5
2-Methylpentane	0.5	Acrolein (CH ₂ CHCHO)	1	2-Ethylfuran	TBD	CH ₃ CCl ₃	0.5
3-Methylpentane	0.5	Methacrolein	1	3-Ethylfuran	TBD	CH ₃ Br	1
<i>n</i> -Hexane	0.5	2-Butenal (Crotonaldehyde)	2	2-Vinylfuran	TBD	CH ₂ Br ₂	0.03
<i>n</i> -Heptane	1	Furfural	TBD	3-Vinylfuran	TBD	CHBr ₃	0.1
2,2,4-Trimethylpentane	0.5	3-Furaldehyde	TBD	1,3-Dioxolane	TBD	CH ₃ I	0.03
<i>n</i> -Octane	0.5	Ketones		Nitrogen-containing VOCs		CH ₂ I ₂	0.05
Alkenes		Acetone	5	Nitriles		C ₂ H ₅ I	0.5
Propene	5	MEK	0.5	HCN (Hydrogen Cyanide)	5	CH ₂ ICl	0.05
1-Butene/Isobutene	1	MVK	0.5	Acetonitrile (CH ₃ CN)	1	CHBrCl ₂	0.05
cis-2-Butene	1	2,3-Butanedione	TBD	Propanenitrile	1	CHBr ₂ Cl	0.03
trans-2-Butene	1	Alcohols		Acrylonitrile	1	CFC-11	5
Isoprene	1	Methanol (CH ₃ OH)	5	Methylacrylonitrile	2	CFC-12	1
α-Pinene	1	Ethanol (C ₂ H ₅ OH)	2	Nitrates		CFC-113	1
β-Pinene/Myrcene	1	2-Propanol	4	Methyl Nitrate	TBD	CFC-114	1
Camphene	1	Ethanol	TBD	Ethyl Nitrate	TBD	HCFC-22	1
Limonene/3-Carene	1	MBO (2-Methyl-3-Buten-2-ol)	1	Isopropyl Nitrate	2	HCFC-141a	1
Tricyclene	1	Esters		<i>n</i> -Propyl Nitrate	2	HCFC-142b	TBD
Aromatics		Methyl Formate	TBD	<i>t</i> -Butyl Nitrate	2	HFC-134a	1
Benzene	0.3	Methyl Acetate	TBD	2-Butyl Nitrate/Isobutyl Nitrate	2	Sulfur-containing VOCs	
Toluene	0.3	Methyl Propionate	TBD	<i>n</i> -Butyl Nitrate	2	OCS	1
Ethylbenzene	0.2	Ethyl Acetate	TBD	Other		CS ₂ (Carbon Disulfide)	0.2
<i>p</i> -/ <i>m</i> -Xylene	0.2	Ethers/Furans		Pyrrrole	TBD	CH ₃ SH (Methanethiol)	TBD
<i>o</i> -Xylene	0.2	MTBE (Methyl <i>t</i> -Butyl Ether)	0.3	Nitromethane (CH ₃ NO ₂)	TBD	DMS (Dimethyl Sulfide)	1
Styrene	0.1	Furan	1			C ₃ O ₂ (Carbon Suboxide)	TBD
Ethynylbenzene	TBD	THF (Tetrahydrofuran)	TBD				

In support of GOTHAAM science goals, we will make continuous *in-situ* 35-s observations of a large suite of C₁-C₁₀ VOCs, reported every ≈2 minutes

NYC-METS

Organic Gases

Online and offline measurements of VOCs-SVOCs at NYC-METS: Vocus PTR-ToF, I-CIMS, GC-ToF/MS



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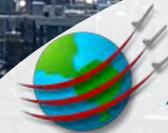


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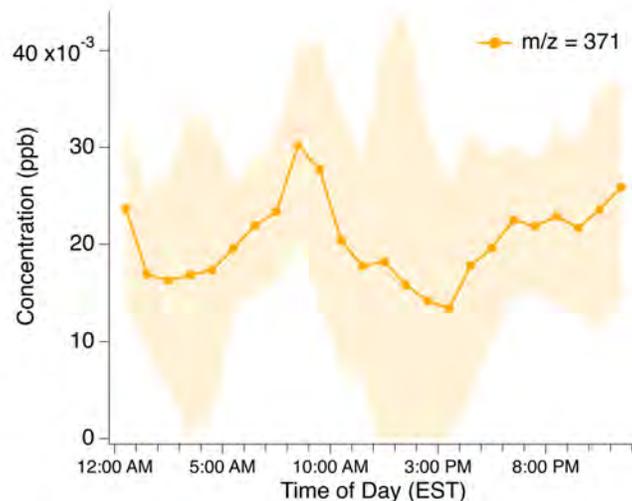
Aerodyne Research Yale ENVIRONMENTAL ENGINEERING



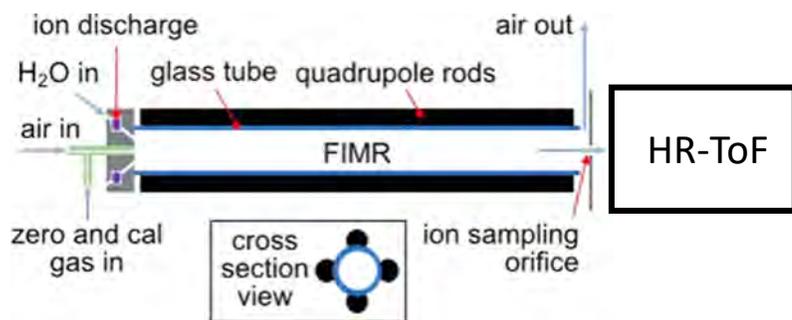
Online gas-phase measurements at NYC and coastal CT

VOCUS-PTR-ToF-MS (at Manhattan & coastal CT)

D5 Average Diurnal Profile
(NYC 2022 prelim data)

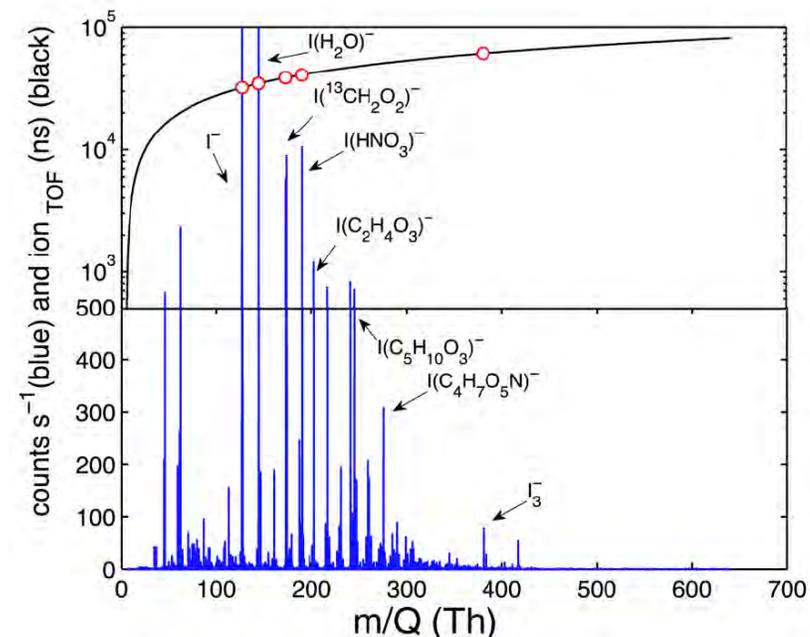


- Time resolution: **4-10 Hz**
- Detection limits: **<50 ppt (for 1 s)**
- Mass resolution: **up to 10,000 $m/\Delta m$**
- Volatility range: **VOCs - SVOCs**
- Inlet locations:
 - **NYC: 90 m ASL (60 m AGL)**
 - **Coastal CT: 10 m ASL**
- Targeted species, including primary emissions and oxidation products:
 - **Oxygenated VOCs - SVOCs**
 - **Aromatics (BTEX, etc.)**
 - **Terpenoids & oxy-terpenoids**
 - **VCP tracers (D4, D5, PCBTF)**
 - **Others (e.g., DMS)**



HR-ToF-I-CIMS (at Manhattan)

- Time Resolution: **1 Hz**
- Detection limits: **<10 pptv (for 1 s)**
- Mass resolution: **up to 6,000 $m/\Delta m$**
- Targeted Species: **Highly-functionalized VOCs-LVOCs, HONO, ClNO₂, Cl₂, HNO₃, HCOOH, oxidation products (e.g., C₅H₁₀O₃, C₁₀H₁₅NO₄, C₁₀H₁₆O_x)**



Example spectra from Lee *et al.* 2014, ES&T

Krechmer *et al.* 2018, *Anal. Chem.*

Offline VOC-SVOC measurements at NYC and coastal CT

Agilent 6550 Q-ToF

Mass accuracy: 1 ppm

Mass resolution: up to 40,000 $m/\Delta m$

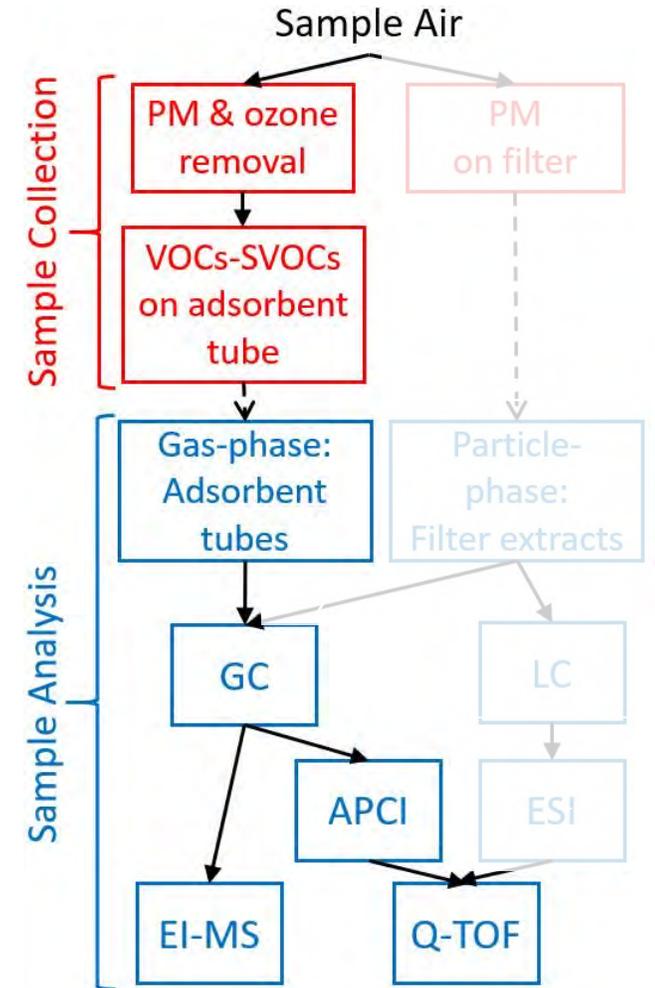
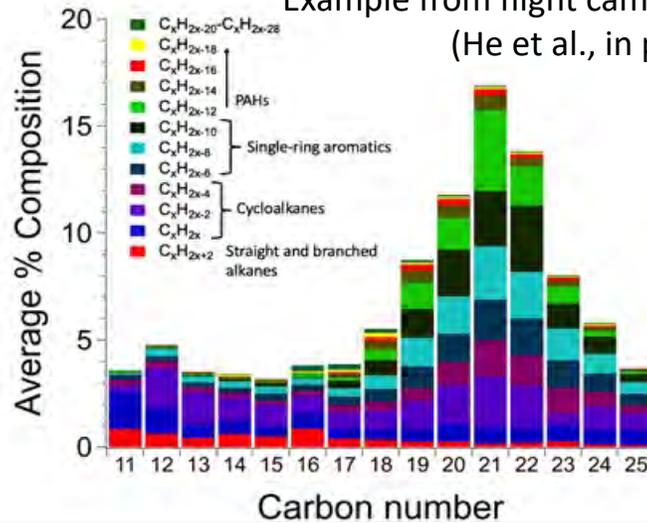
Configuration: with GC or LC



- Collection media: Custom adsorbent tubes
- Time resolution: Multi-hour offline samples
- Volatility range: IVOCs-SVOCs and larger VOCs
- Analysis approach: Thermal desorption (GERSTEL) with GC separation and column effluent split to:
 - (1) APCI-ToF (atmospheric pressure chemical ionization) for targeted complex mixture speciation and supplemental unknowns analysis
 - (2) EI-MS (electron ionization) for single compound measurements and supplemental identifications

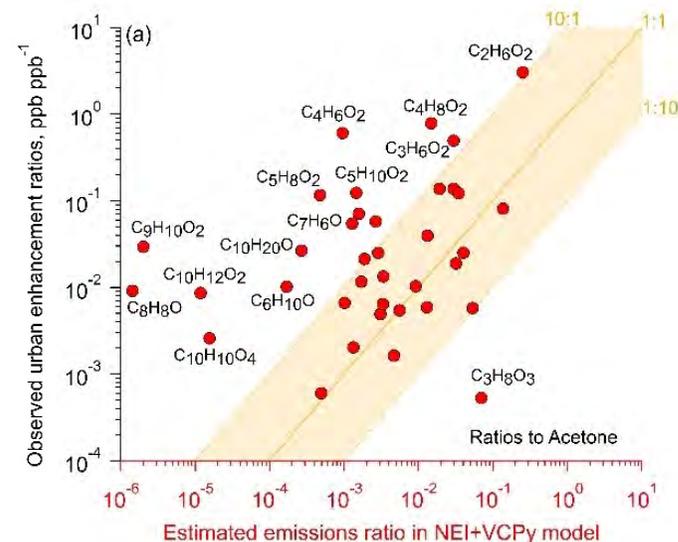
Complex Mixture Speciation via GC-APCI-ToF

Example from flight campaign
(He et al., in prep.)



Overview of science goals and foci

- Multi-site online and offline VOC-SVOC measurements spanning hydrocarbons to highly-functionalized organics
- Examining emissions and apportionment to traditional and emerging sources using VOC-SVOC and other measurements
- Comparisons to emissions inventories and models in collaboration with modelers
- Contributions to SOA, ozone, and OVOC formation affecting urban and downwind air quality, examined using gas and aerosol measurements
- Understanding present day and future AQ in NYC and downwind



Example of regionally-resolved NYC emission inventory comparison (Khare *et al.* 2022, *ACPD*)



Speciated measurements of VOCs in NYC by an *in situ* GC-TOF-MS for the characterization of primary emissions in urban air



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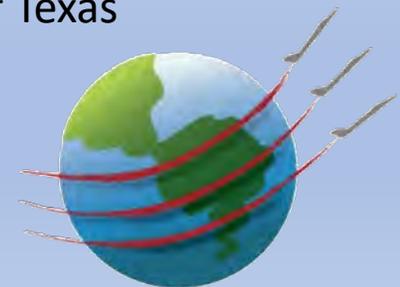


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AERODYNE RESEARCH, Inc.

2-Channel GC with EI-TOF-MS at NYC-METS ground site

Aerodyne 2-channel *in situ* gas chromatograph (GC) with thermal desorption preconcentration (TDPC) and electron ionization time-of-flight mass spectrometric (EI-TOF-MS) detection



- NYC-METS Measurement Intensives
 - 4 weeks each: Summer 2022, 2023
- Targeted Species
 - Alkanes, alkenes: $C_3 - C_{12}$
 - Biogenics: isoprene, monoterpenes
 - Aromatics: BTEX, C_9 aromatics
 - Oxygenates: alcohols, ketones, aldehydes, ethers, esters
 - Halocarbons, siloxanes, nitrogen containing species
- Instrument Performance
 - Time resolution: 30 min cycle
 - Each cycle contains 8.5 minute sample collection period
 - Typical detection limits: < 5 ppt (0.85 L precon. sample)
 - Mass resolution of EI-TOF-MS: $4000 m/\Delta m$

Science Goals

- VOC measurements
- Quantitative timeseries
- Emission ratios
- Photochemical clocks
- Source apportionment
- Impact of meteorological events (e.g. heat) on VOC composition
- Integrating our data with other VOC measurements at the ground site (e.g. PTR-MS)
- Summer 2022 data status: QA/QC and HR processing

First Look at GC-TOF data from Summer 2022 NYC-METS

