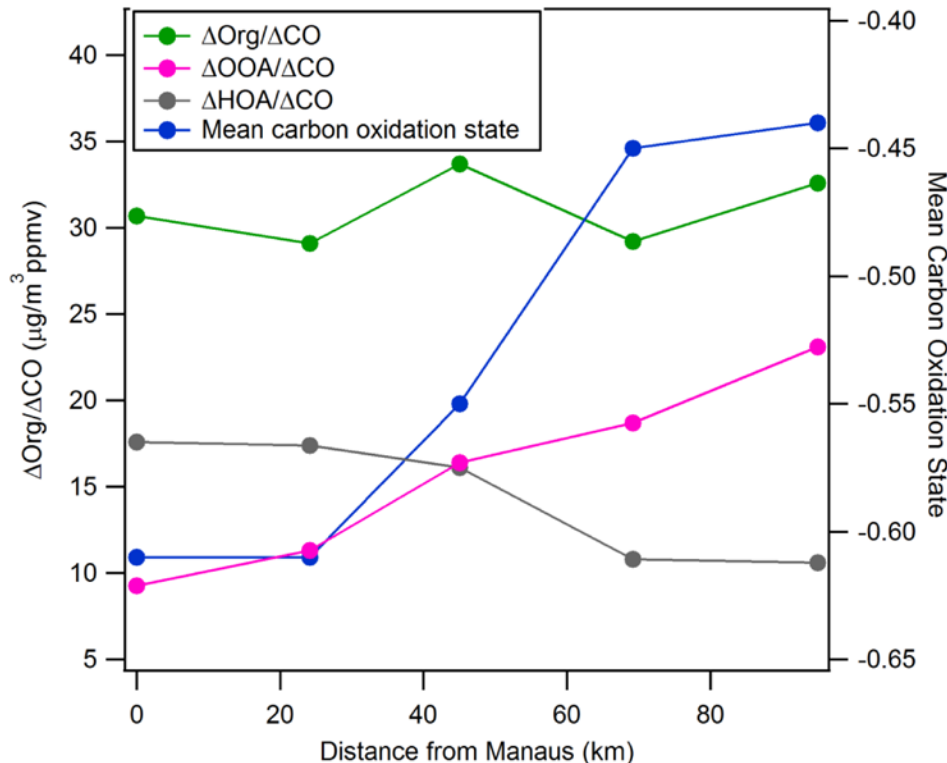


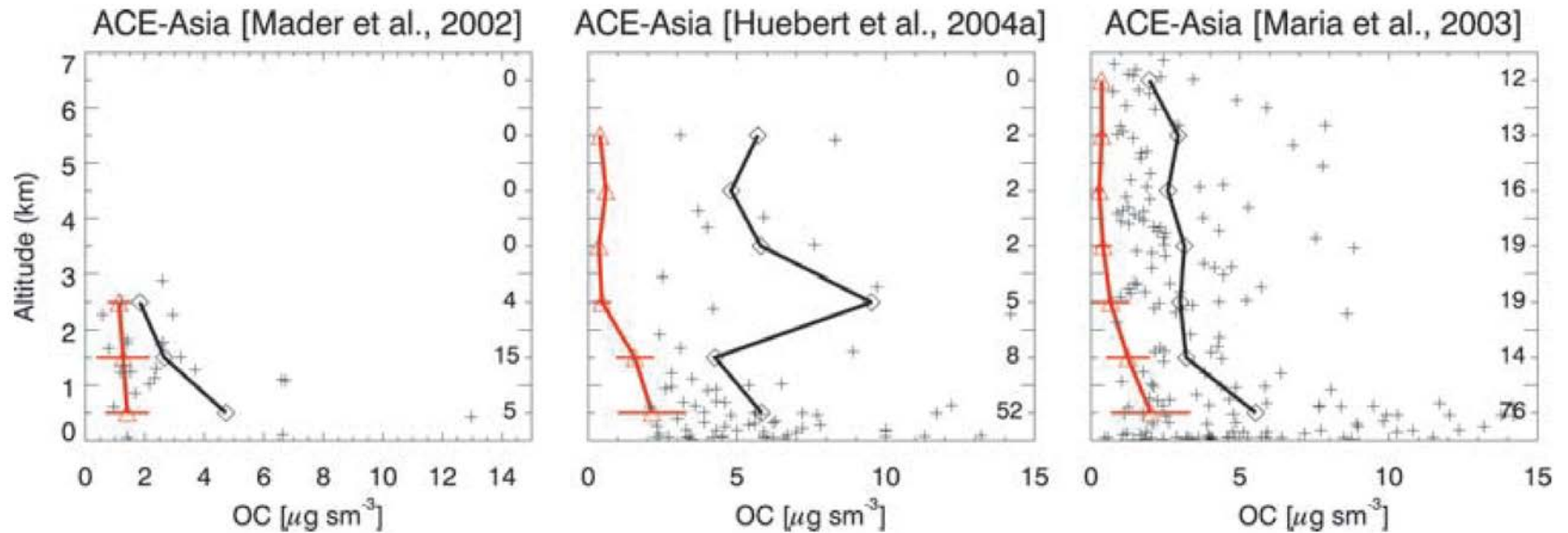
Aircraft Observations of Aerosol in the Manaus Urban Plume and Surrounding Tropical Forest during GoAmazon 2014/15

John E. Shilling¹, Mikhail S. Pekour¹, Edward C. Fortner², Paulo Artaxo³, Suzane de Sá⁴, John M. Hubbe¹, Karla M. Longo⁵, Luiz A.T. Machado⁶, Scot T. Martin^{4,7}, Stephen R. Springston⁸, Jason Tomlinson¹, Jian Wang⁸



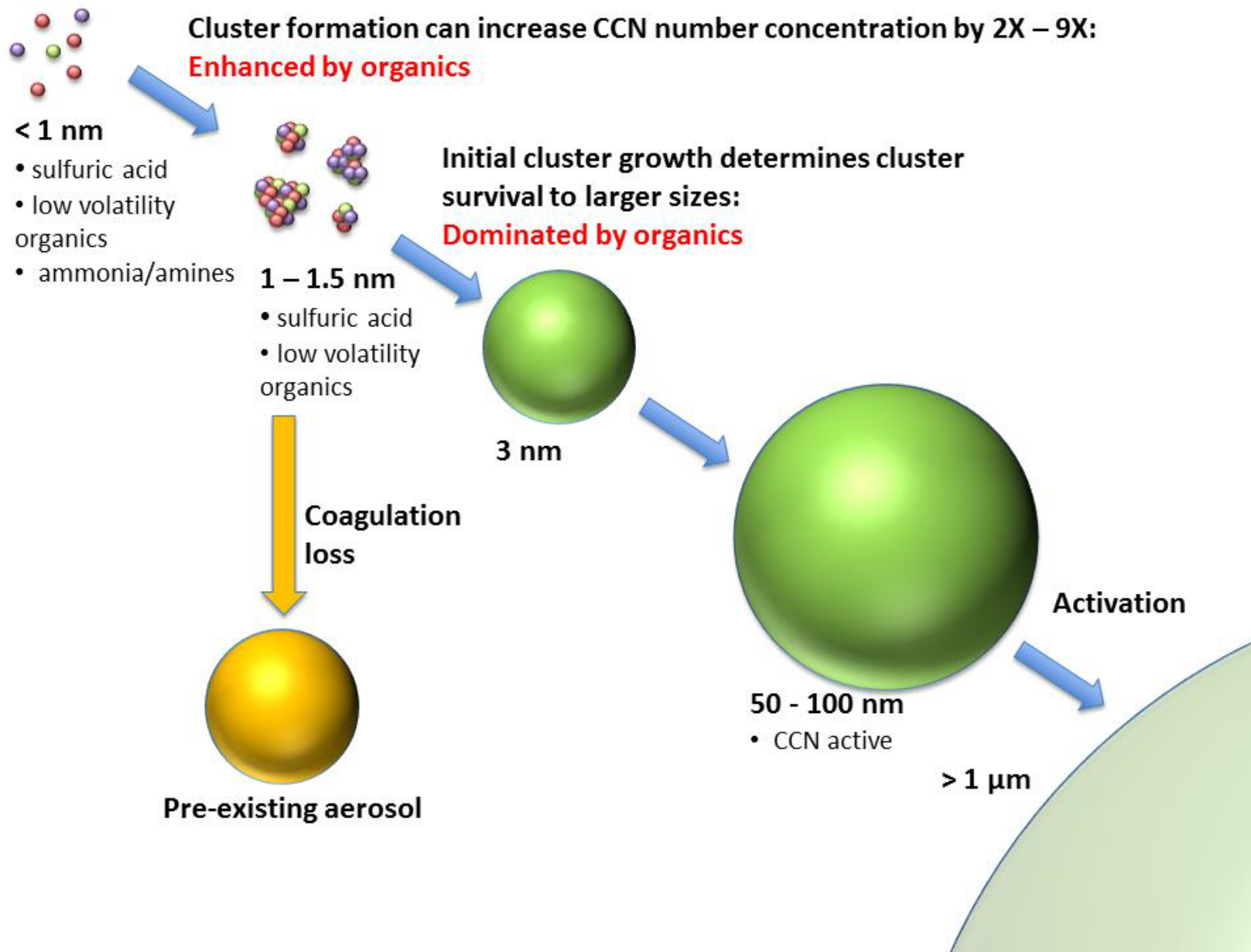
Key metrics describing the evolution of the Manaus urban plume on March 13. Each data point represents the average values for one pass through the plume. Mean wind speeds were 7 m/s on this flight, thus data capture approximately the first 4-5 hours of the plume aging. Calculations of $\Delta\text{Org}/\Delta\text{CO}$ values use method one with details on the calculations and methods provided in the SI. Spatial locations of each leg are labeled in Figure 1.

HEALD ET AL.: LARGE TROPOSPHERIC SOURCE OF OC AEROSOLS

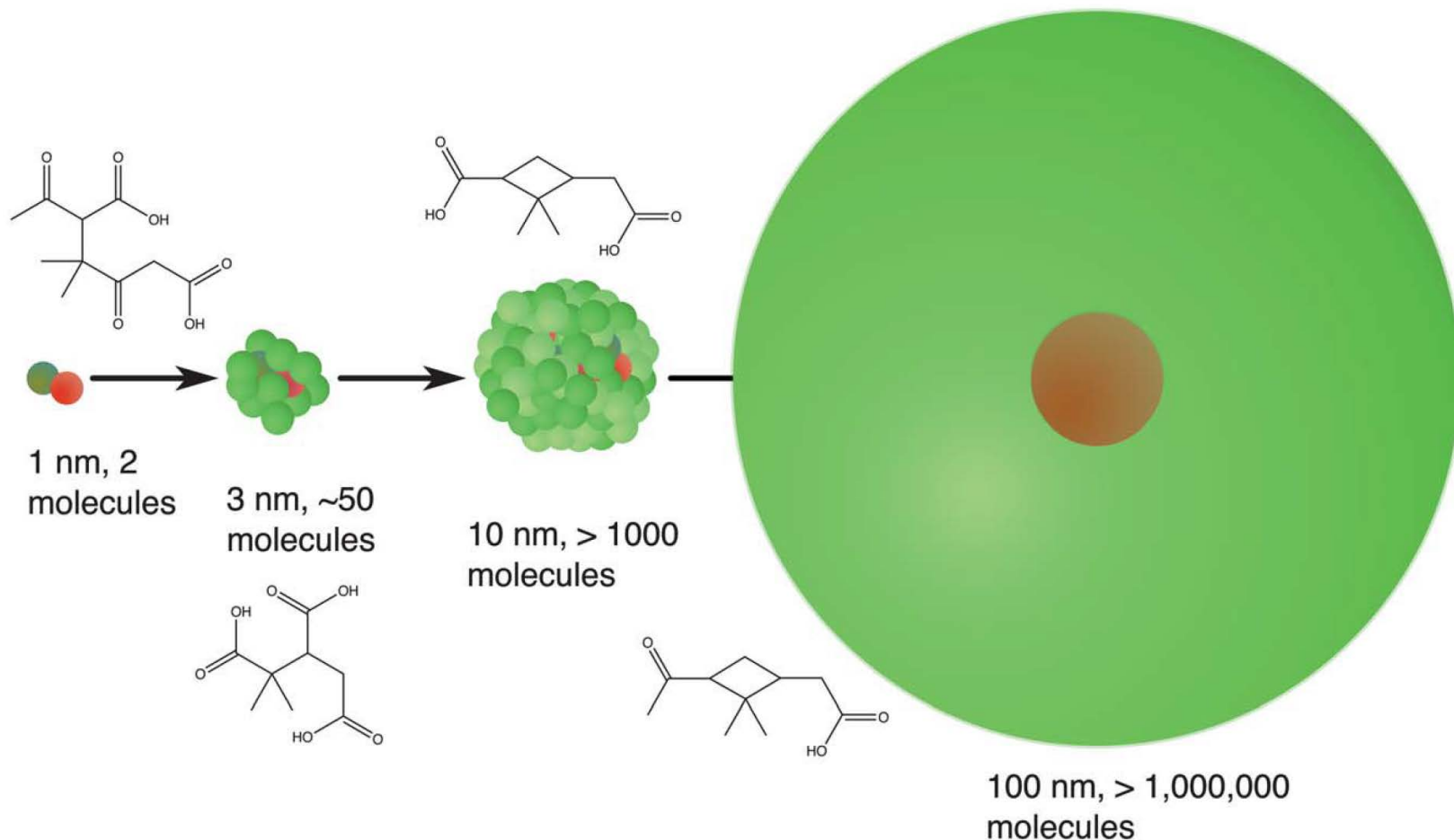


- 15 years ago: an unknown atmospheric component
- Today in 2018: **NOT included explicitly in any climate model**, even if it accounts for 50-85% of global aerosol loading: It is all parameterized..
- Even basic WRF-Chem do not include all SOA production mechanisms (urban or remote)

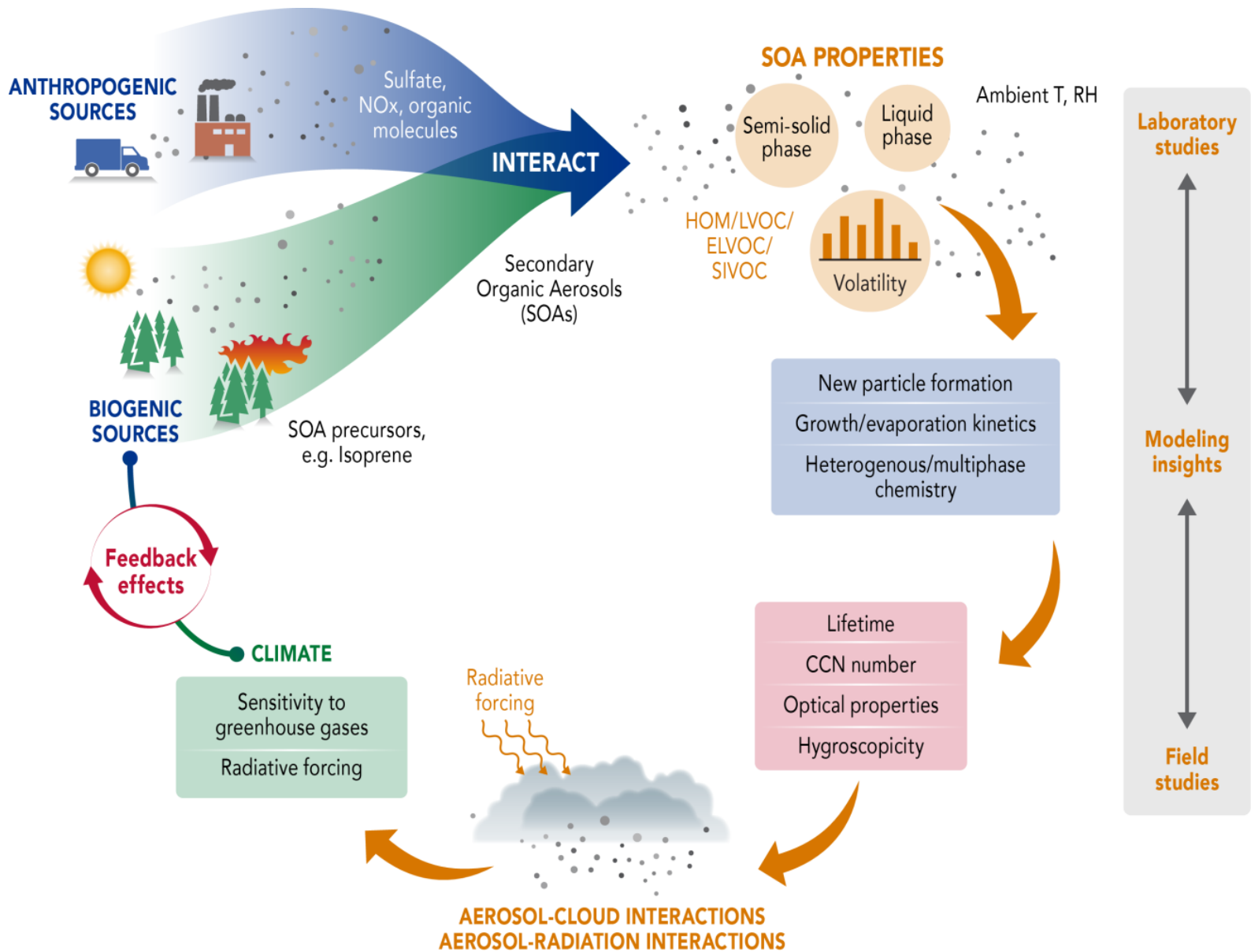
Schematic illustrating the participation of organic oxidation products as newly formed clusters either grow and survive to CCN-active sizes or are lost by coagulation with the pre-existing aerosol



Overall process of new-particle formation and growth involving association of oxidised organics and sulphuric acid. Initial stable cluster formation requires rare but highly oxidised, extremely low volatility organics. As clusters grow, progressively more common but also more volatile species can contribute to growth, until at some size near 10 nm the particle organic composition begins to resemble bulk organic-aerosol composition.

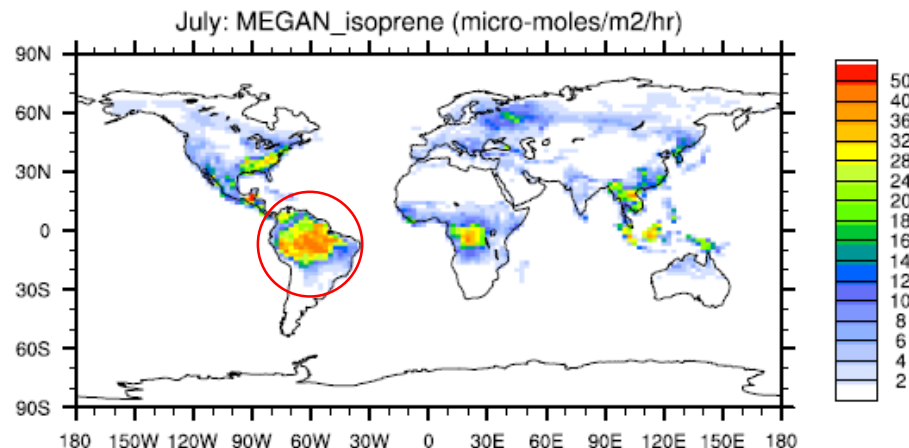


Processes governing the climatic importance of SOA

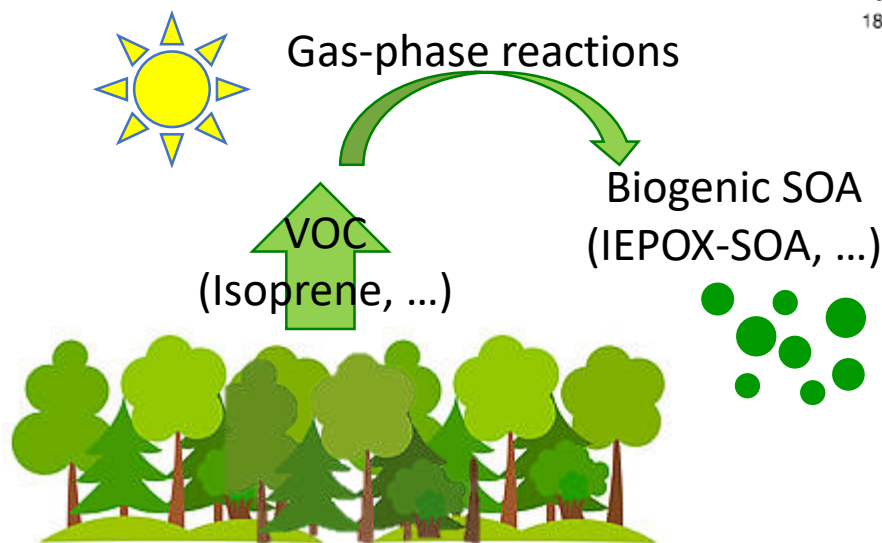


IEPOX-SOA

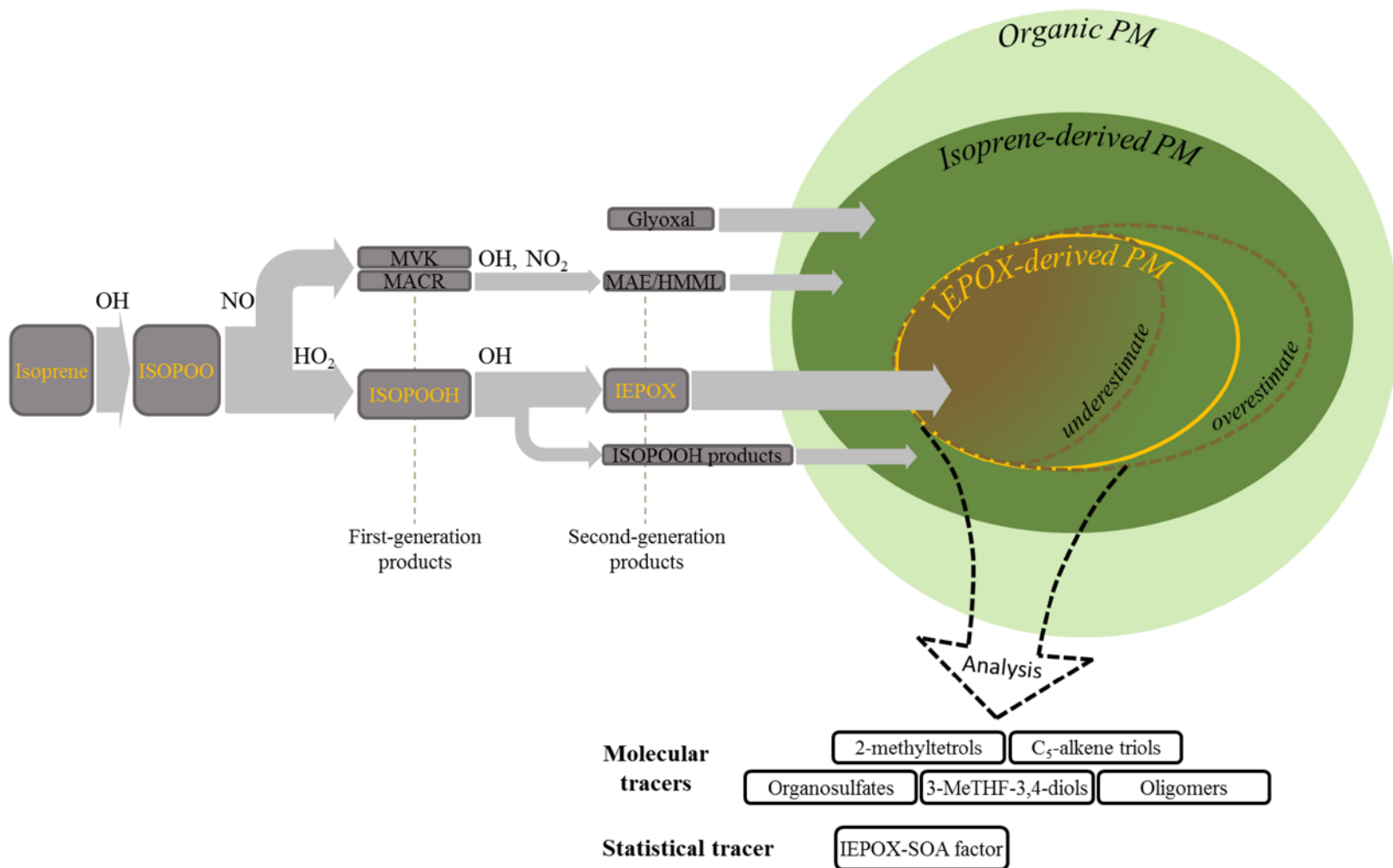
- Isoprene emitted by vegetation, gas-phase reactions to Isoprene EPOXYdiol (IEPOX)
- Conditions:
 - Low NO_x
 - Uptake by acidic aerosol → IEPOX-SOA aerosol



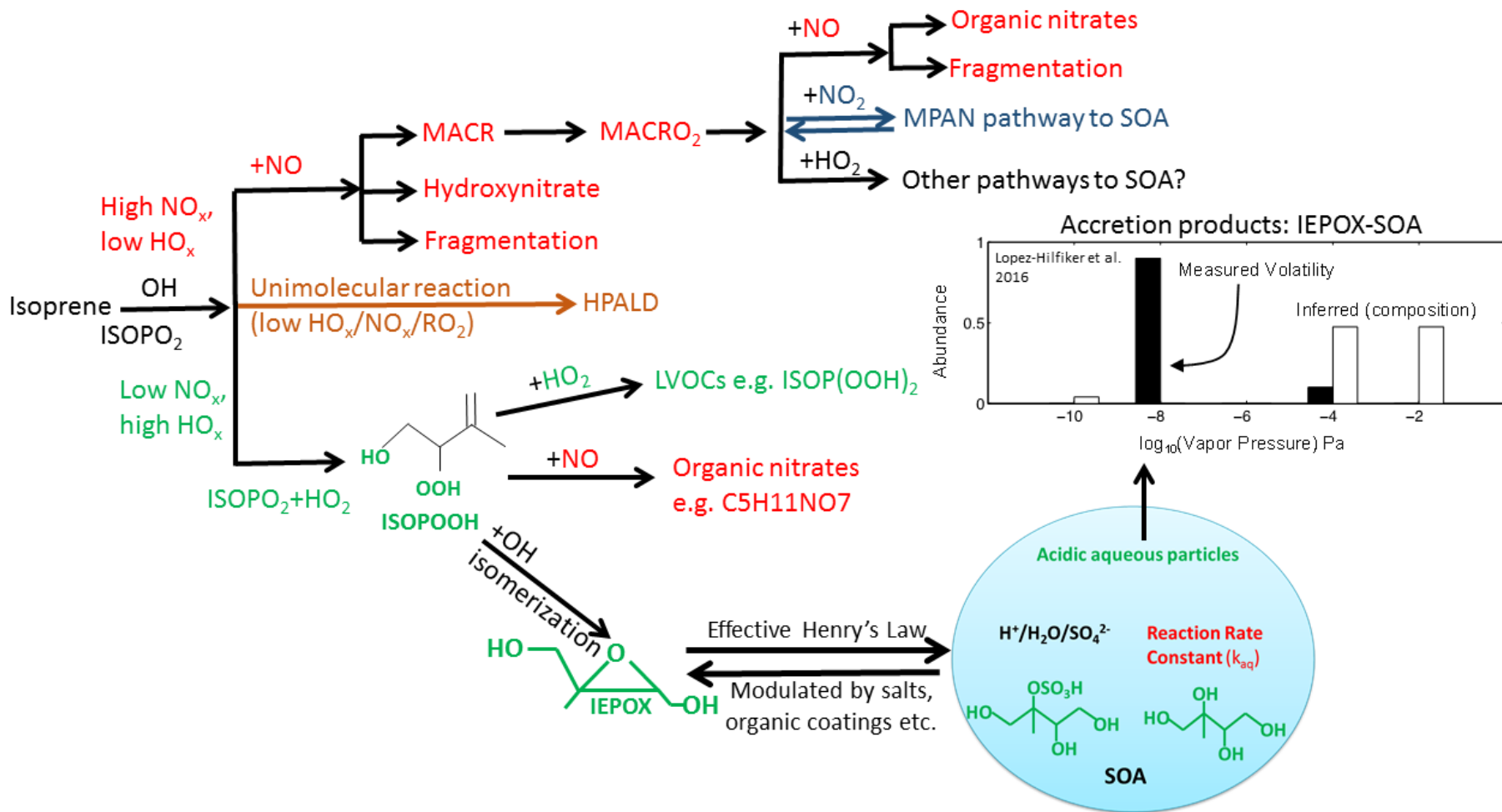
Guenther et al. 2012



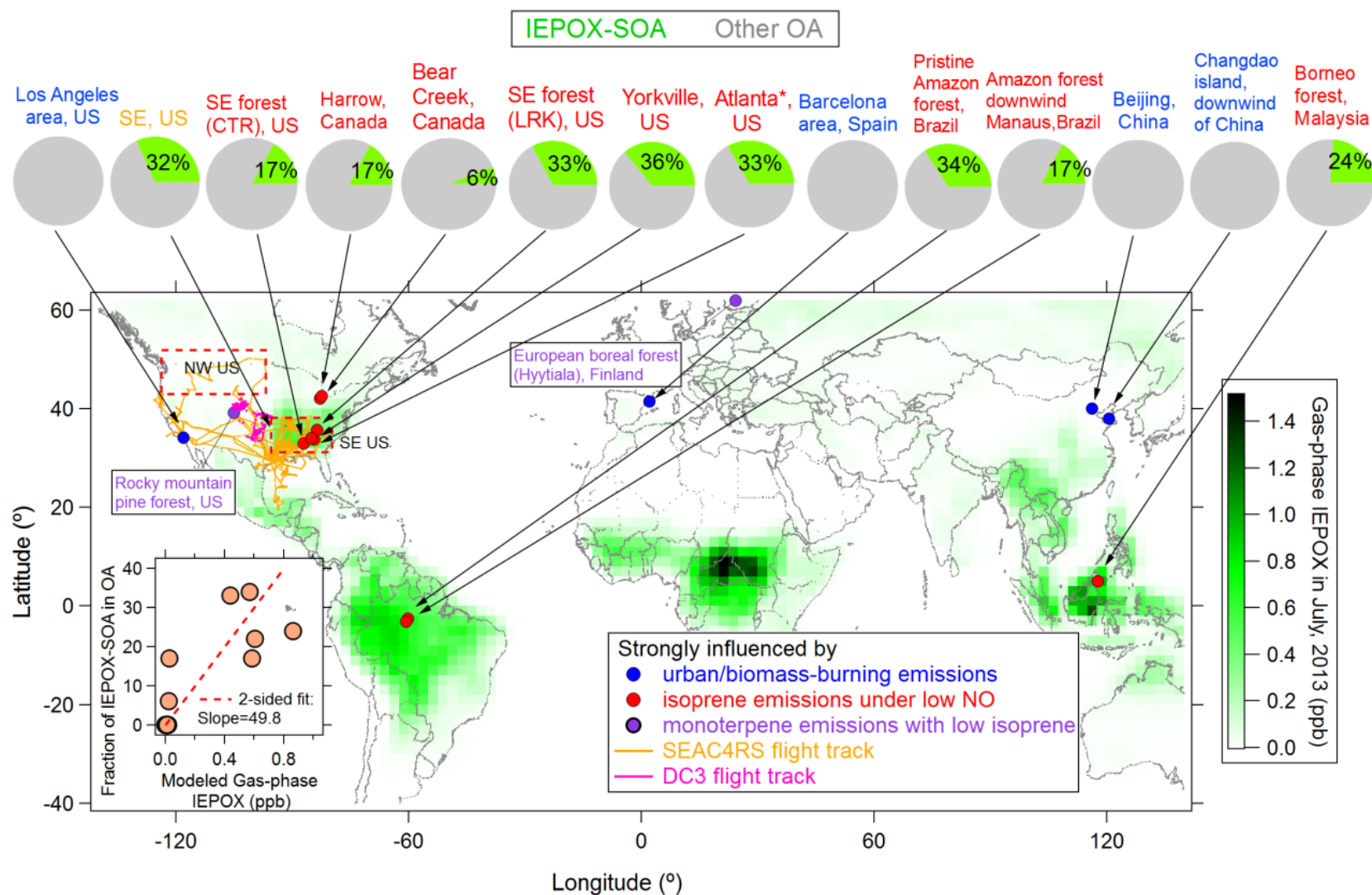
Production of IEPOX-derived PM from the photooxidation of isoprene



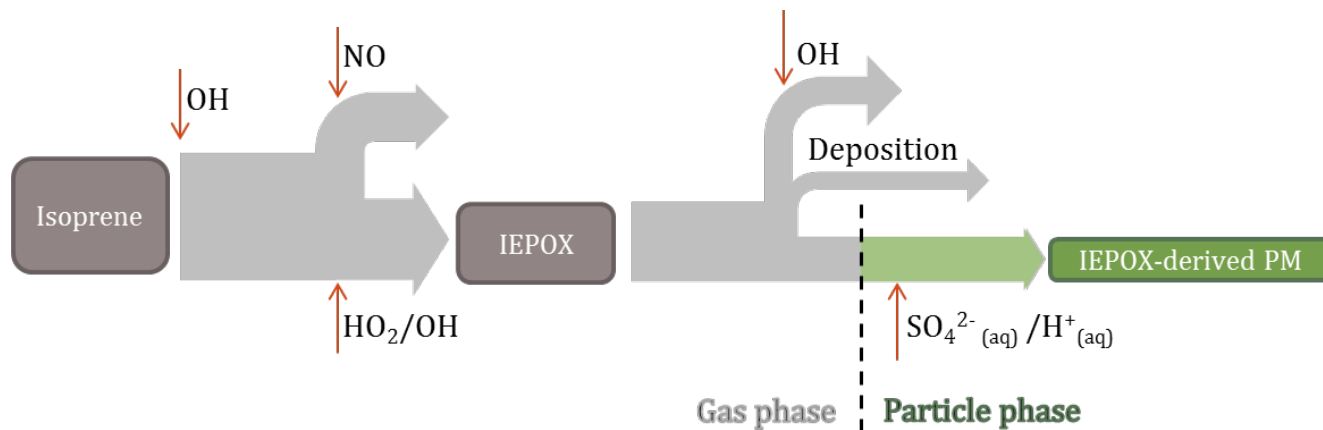
Fate of isoprene RO2 radicals (ISOPO2) depicting different chemical pathways that depend on the NOx, HOx, and RO2 regimes. Several pathways lead to SOA formation, but the particle loading/composition differs greatly between different regimes, and also depends on acidic/non-acidic seeds.



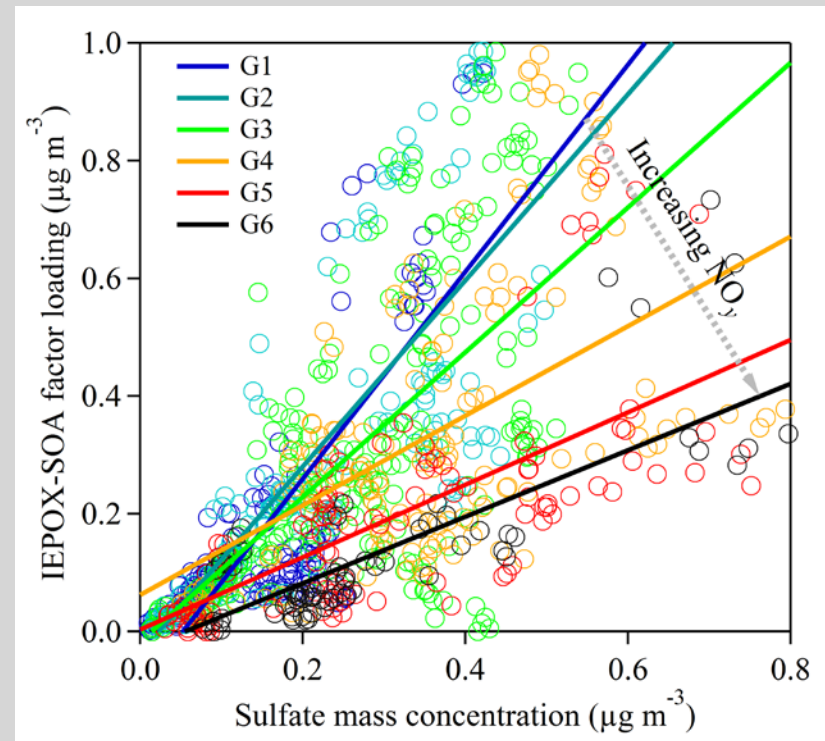
IEPOX-SOA fractions of OA in various studies around the world (Hu et al. [2015])



Influence of anthropogenic emissions on isoprene-derived particulate matter in central Amazonia



- Observational constraints of sulfate as a first order predictor and NO as a modulator of IEPOX-derived PM.
 - NO_y serves as indicator of integrated exposure of air mass to NO chemistry.
 - IEPOX-SOA factor obtained from PMF analysis of AMS data is a proxy for IEPOX-derived PM.
- Lower loadings of IEPOX-SOA factor observed for polluted compared to background conditions



BAMS March 2018

SYNTHESIS OF THE SOUTHEAST ATMOSPHERE STUDIES

Investigating Fundamental Atmospheric Chemistry Questions

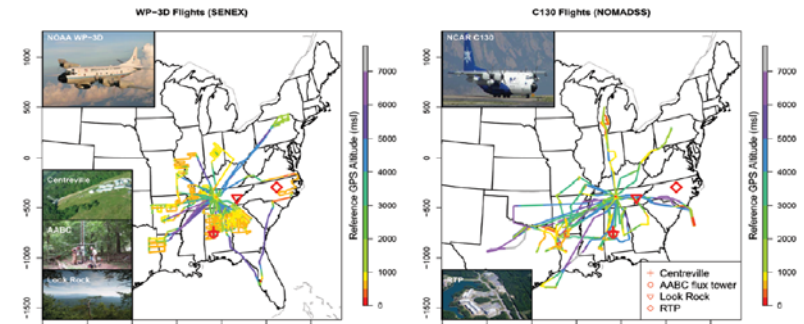
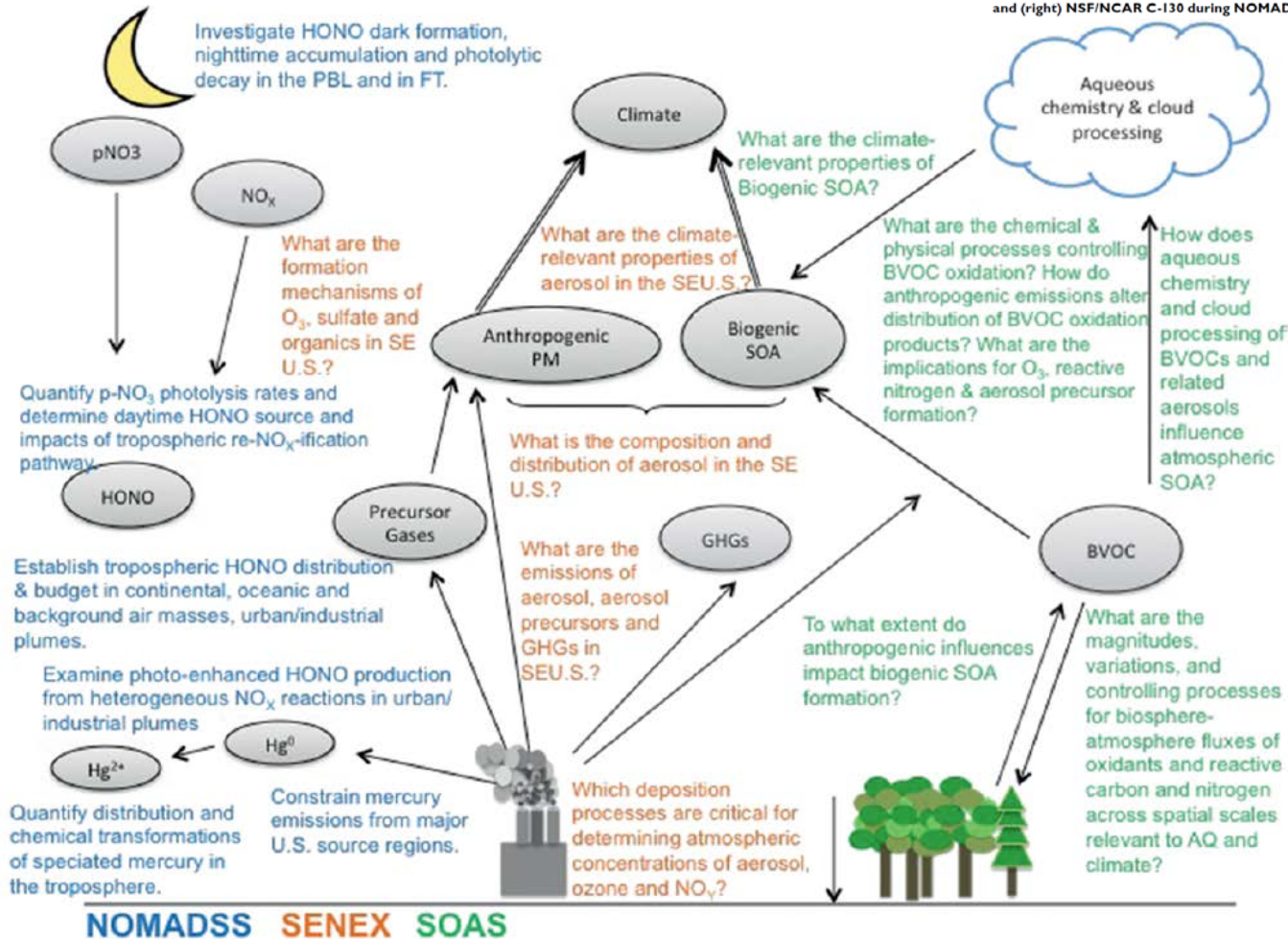


FIG. 1. SAS platforms and locations. Flight tracks are shown for (left) NOAA WP-3D aircraft during SENEX and (right) NSF/NCAR C-130 during NOMADSS.



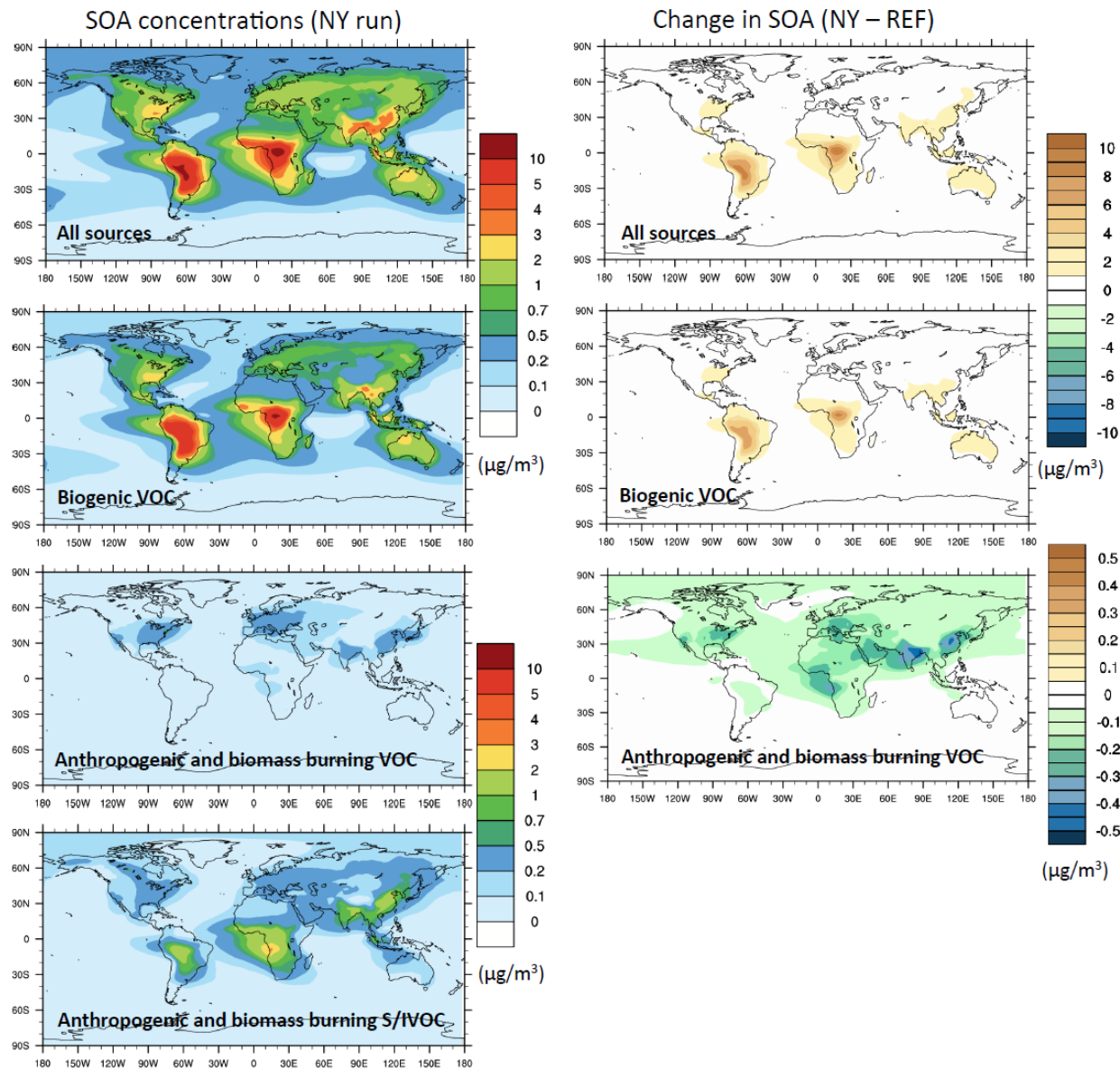
Biogenic SOA accounts for 30% of PM₁ in Southeast US

FIG. 2. Science questions and objectives of the coordinated SAS campaign.

This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

Rethinking the global secondary organic aerosol (SOA) budget: stronger production, faster removal, shorter lifetime

A. Hodzic¹, P. S. Kasibhatla², D. S. Jo³, C. Cappa⁴, J. L. Jimenez⁵,
S. Madronich¹, and R. J. Park²



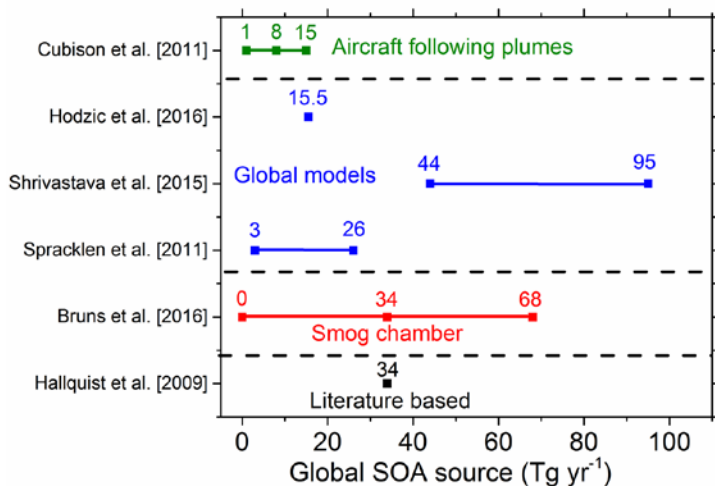
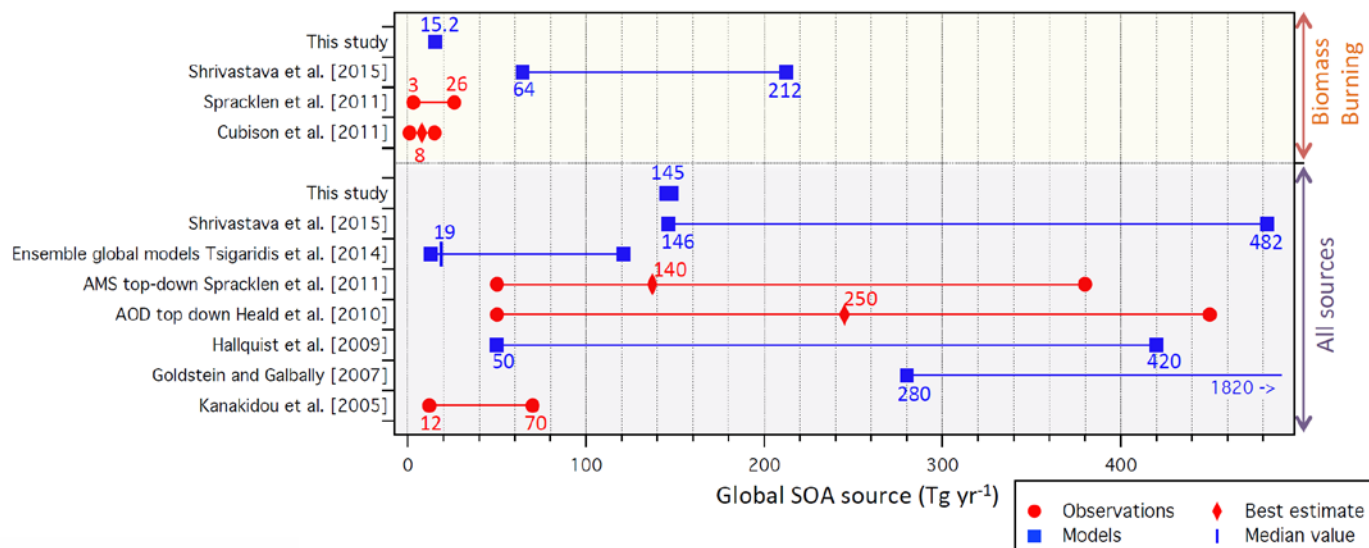
2005–2008 average concentrations for SOA and its constituents as predicted by the GEOS-Chem NY run in the lower troposphere (surface to 5 km; left column). Total SOA is separated into SOA from biogenic VOCs, anthropogenic and biomass burning traditional VOCs, and anthropogenic and biomass burning S/IVOC. The NY run is also compared with REF (right column).

This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

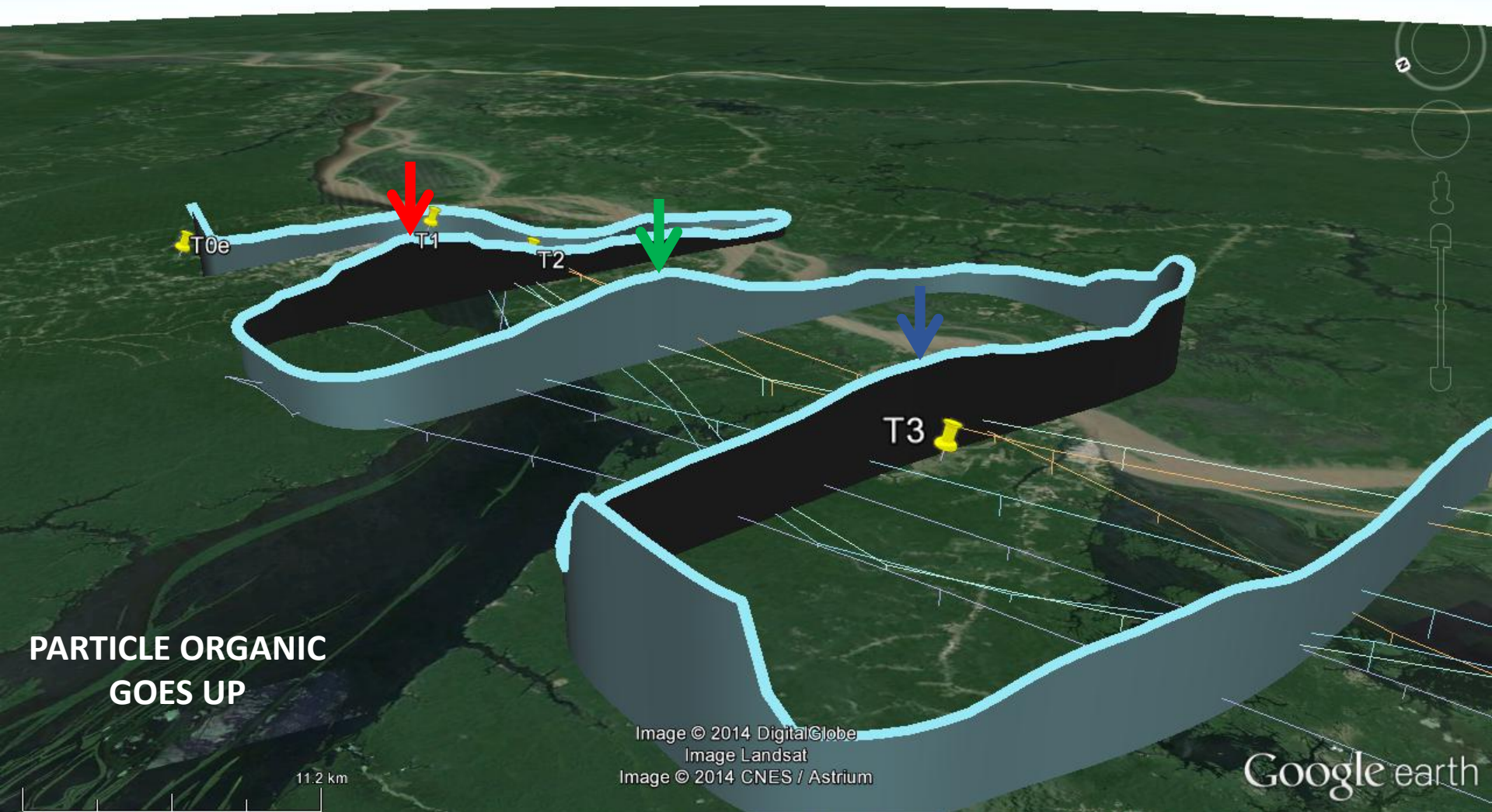
Rethinking the global secondary organic aerosol (SOA) budget: stronger production, faster removal, shorter lifetime

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Global SOA particle-phase source (Tg yr^{-1}) as predicted in this study (NY_DPH) and as reported by previous studies. SOA production from all sources (anthropogenic, biomass burning, biofuel and biogenic) as well as from biomass burning alone is shown



Global source of biomass-burning SOA (Tg yr^{-1}) as estimated by different studies: aircraft following plumes of biomass-burning fires (green), global models evaluated with OA measurements in regions influenced by biomass-burning emissions (blue), estimate based on measurements of biomass-burning SOA in smog chamber (red) and a literature estimate (black). Both measurements and modeling studies report a wide range of SOA produced from oxidation of biomass-burning emissions.



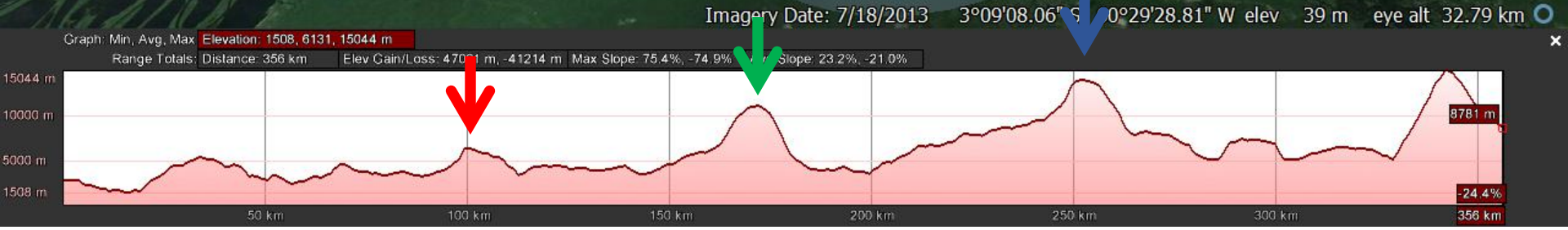
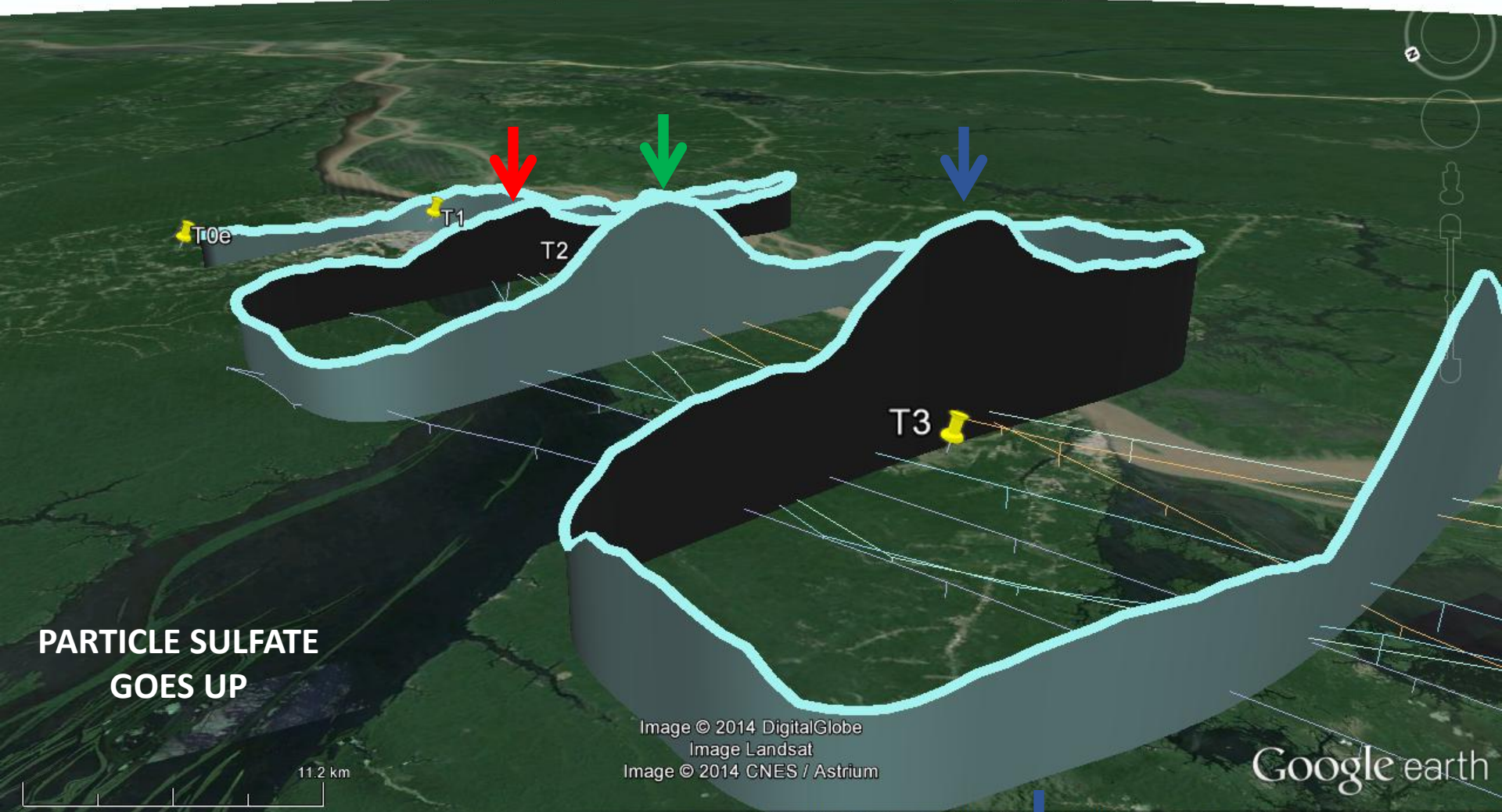
**PARTICLE ORGANIC
GOES UP**

Image © 2014 DigitalGlobe
Image Landsat
Image © 2014 CNES / Astrium

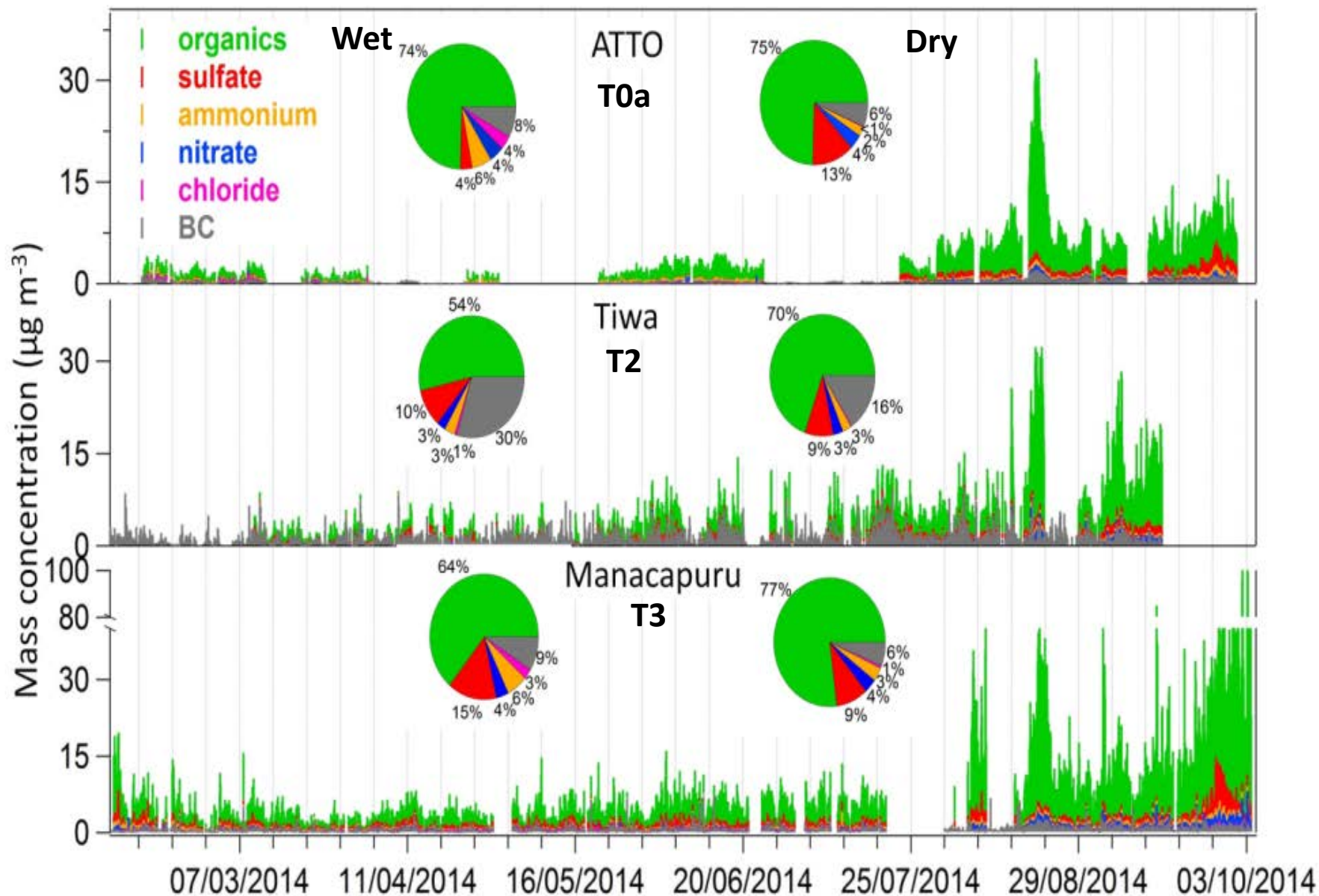
Google earth

Imagery Date: 7/18/2013 3°09'08.88" S 60°29'27.25" W elev 39 m eye alt 32.81 km



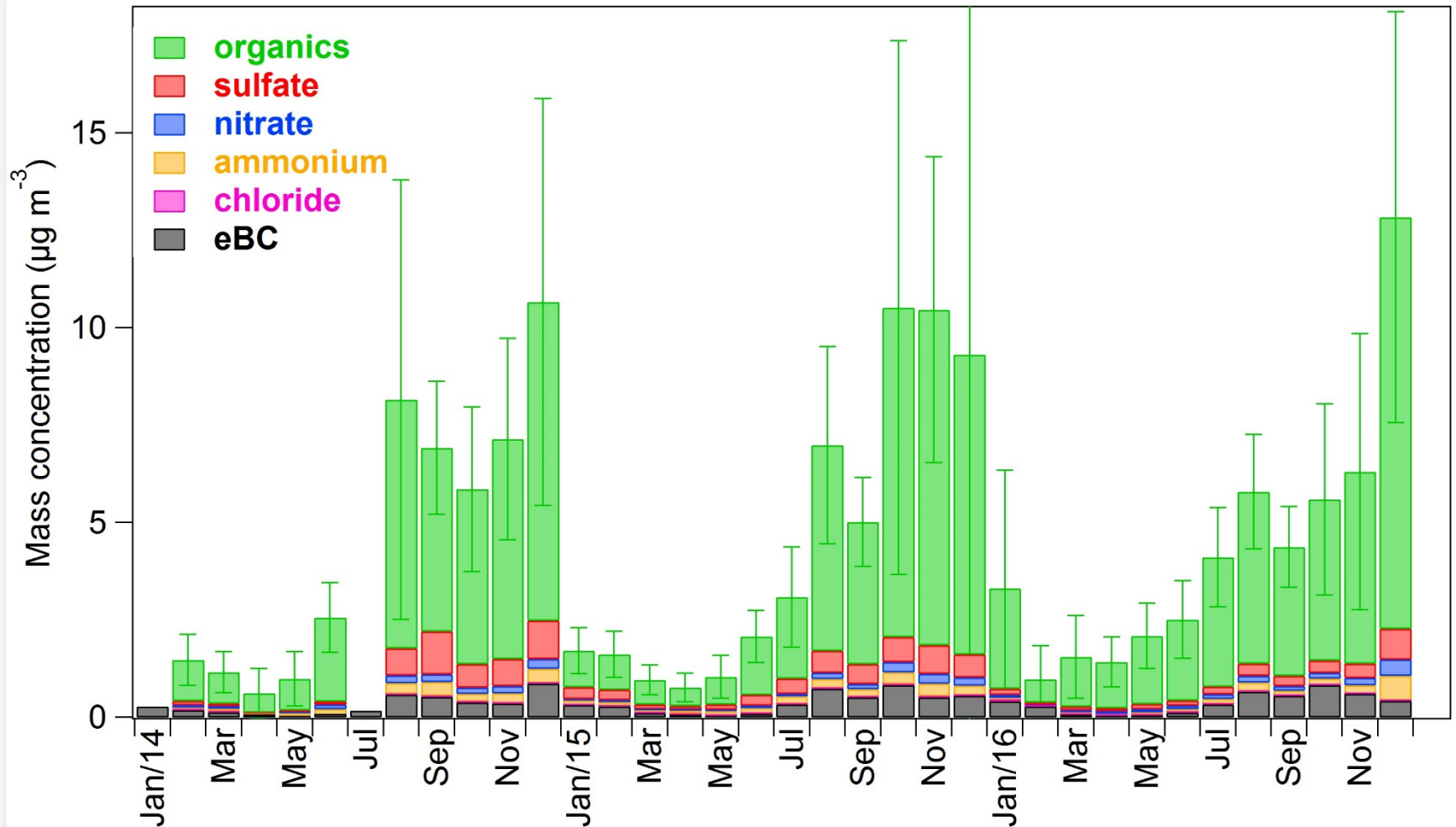


Organic aerosols from ATTO to Tiwa and Manacapuru (with BC)



ATTO ACSM Monthly averages

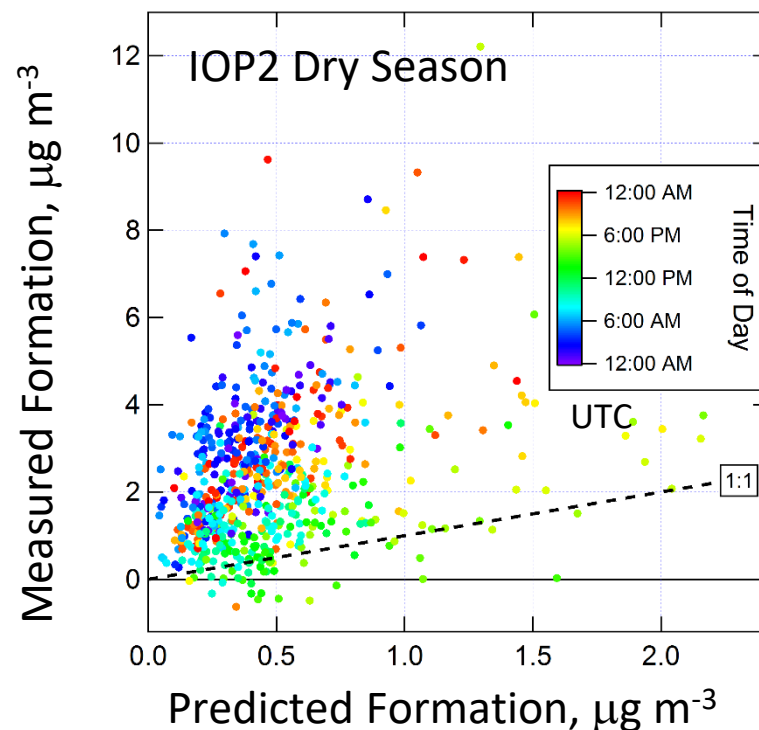
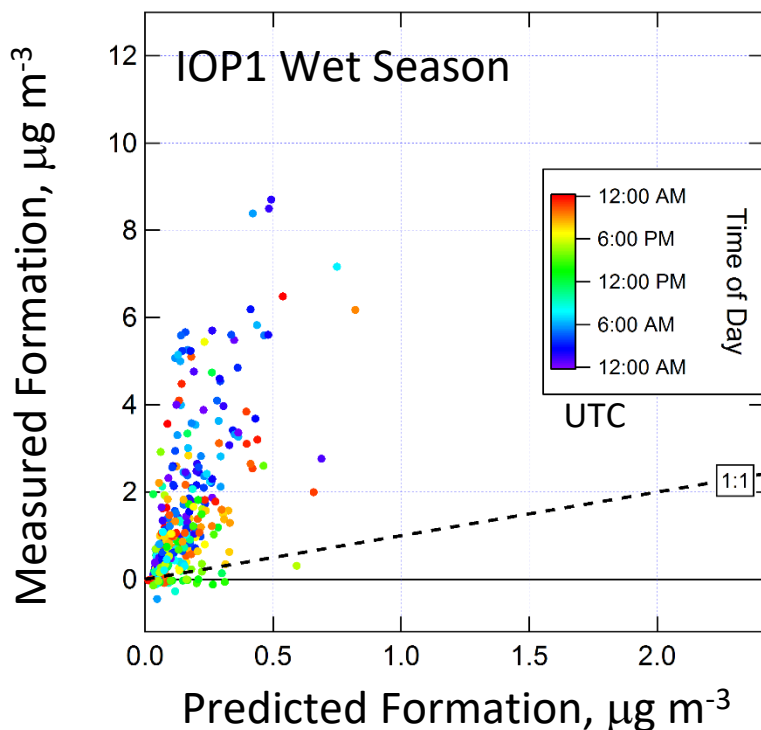
2014-2016



More Secondary Aerosol Potential than Expected

Objective:

Test through in situ data sets the potential for secondary aerosol production and compare to state of understanding with respect to known species and their reactive yields



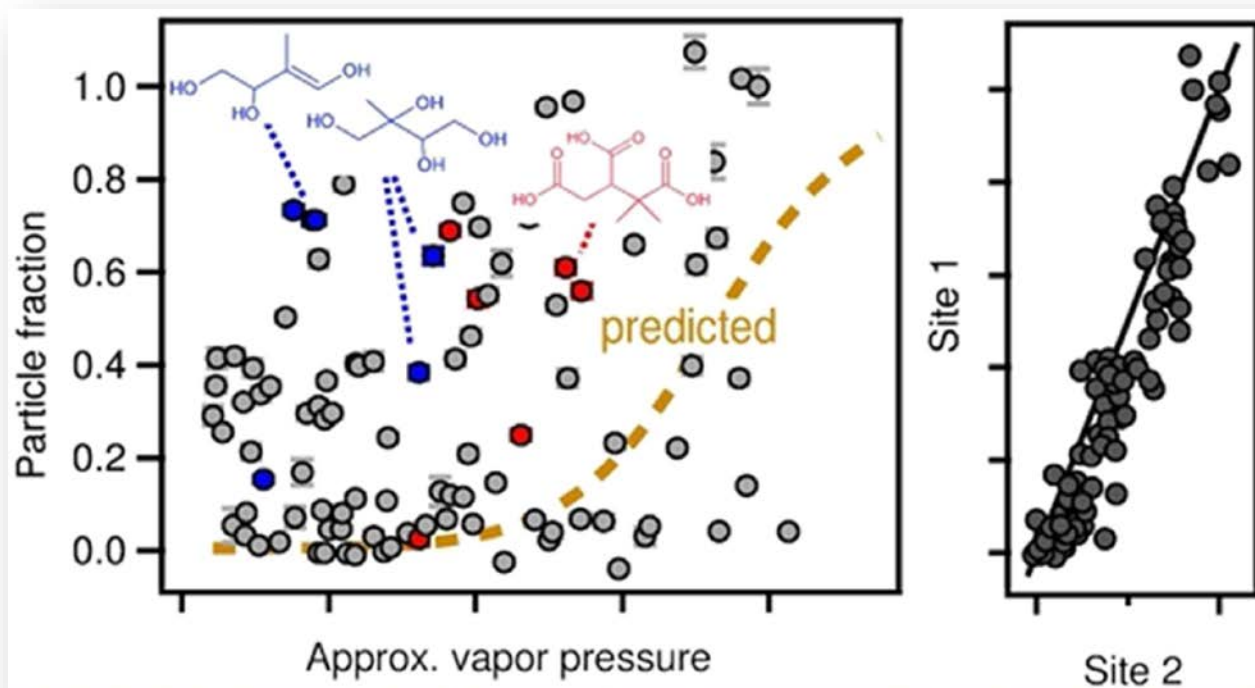
Results:

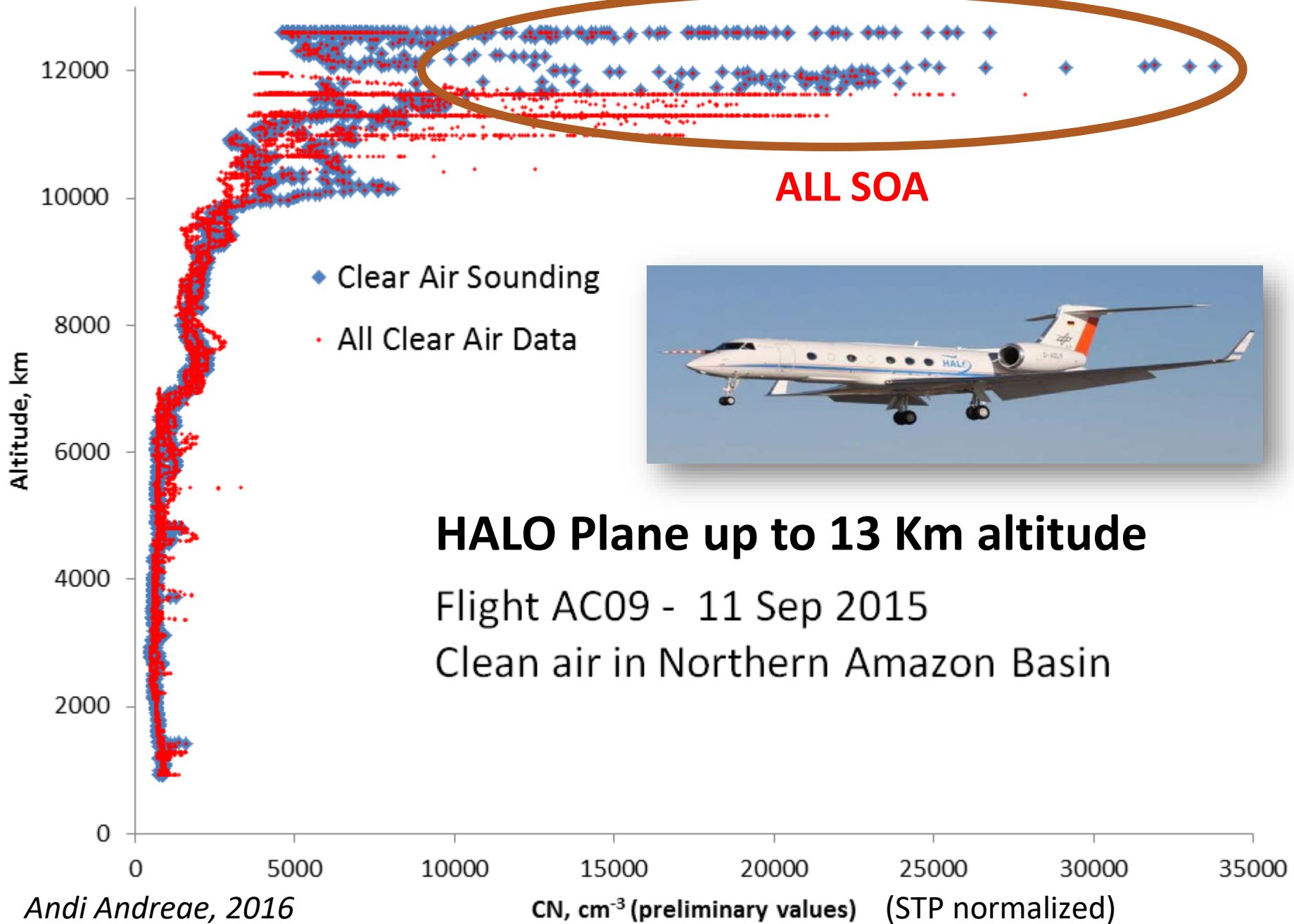
- Secondary aerosol produced from oxidation by OH of ambient air
- Production much greater than predicted from modeled yields of measured ambient precursors
- Suggests that production is dominated by unmeasured species



SV-TAG - semi-volatile thermal desorption aerosol gas chromatograph with in situ derivatization, which provides hourly resolved concentrations and gas-particle partitioning of most common BVOC oxidation tracers

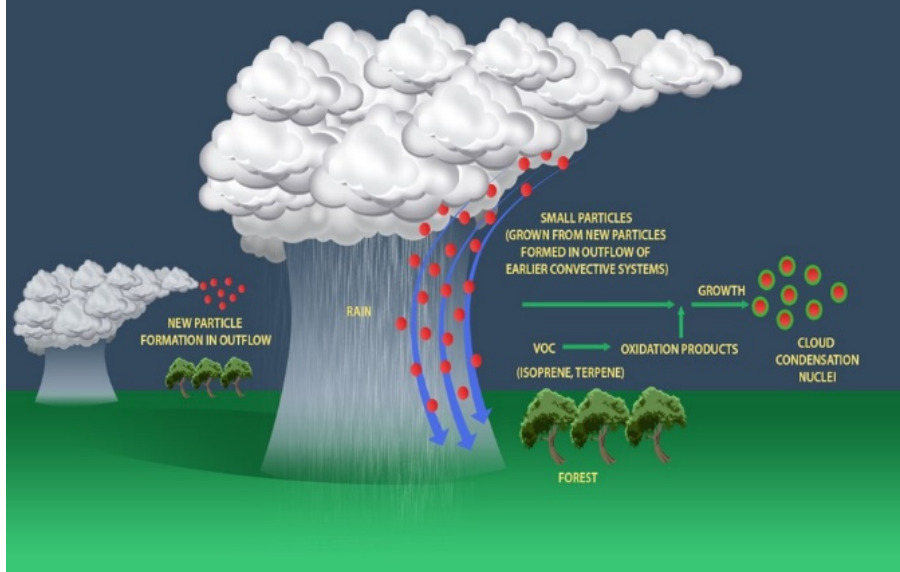
SOAS+GoAmazon: The results of the present study show that the gas-particle partitioning of approximately 100 known and newly observed oxidation products is not well explained by environmental factors (e.g., temperature). Compounds having high vapor pressures have higher particle fractions than expected from absorptive equilibrium partitioning models. Many commonly measured biogenic oxidation products may be bound in low-volatility mass that decomposes to individual compounds on analysis





Andi Andreae, 2016

Clouds as active aerosol processors in the atmosphere



Biogenic organic aerosol formation at low H_2SO_4 happens in the UT

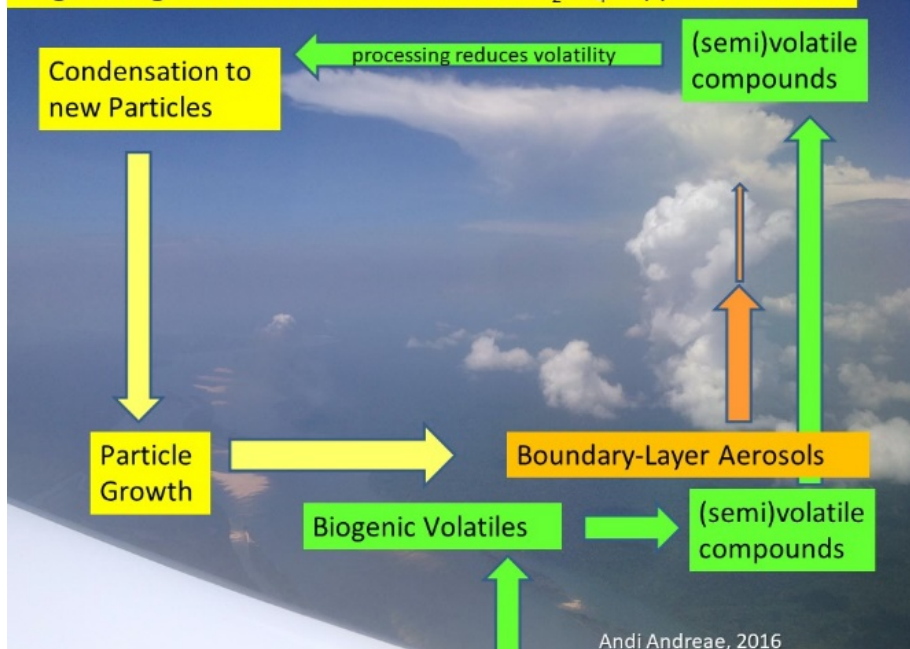
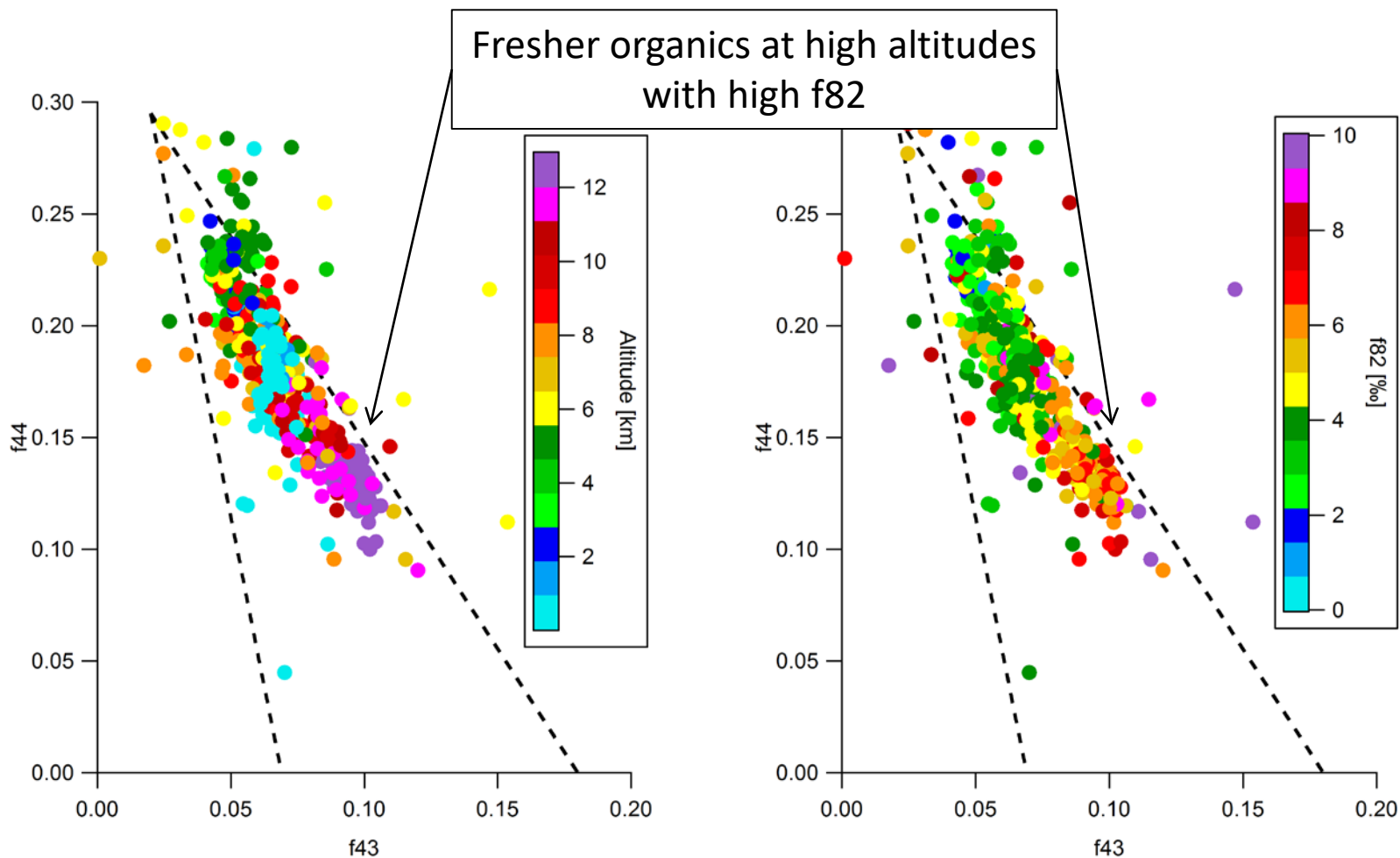


Figure 1.6.3 - On the left panel we can observe how clouds can be active aerosol processors in the atmosphere of Amazonia. The right panel shows how semi volatile compounds produced by the vegetation can be transported to the upper atmosphere by convection, where it condenses due to low temperature and is processed and oxidized to less volatile compounds and produce particles that are brought to the low atmosphere by precipitation.

AC 13

Aging of organic aerosol – Altitude & f82 dependence



1 Strong pollution-enhanced biogenic SOA over the Amazon rainforest

2 Manish Shrivastava^{1,*}, Meinrat O. Andreae^{2,3}, Paulo Artaxo⁴, Henrique Barbosa⁴, Larry K. Berg¹,
 3 Joel Brito⁵, Joseph Ching^{1,6}, Richard C. Easter¹, Jiwen Fan¹, Jerome D. Fast¹, Zhe Feng¹, Jose D.
 4 Fuentes⁷, Marianne Glasius⁸, Allen H. Goldstein⁹, Helber Gomes¹⁰, Dasa Gu¹¹, Alex Guenther^{1,11},
 5 Shantanu H. Jathar¹², Saewung Kim¹¹, Ying Liu¹, Sijia Lou¹, Scot T. Martin¹³, V. Faye McNeill¹⁴,
 6 Adan Medeiros¹⁵, Suzane S. de Sá¹³, John E. Shilling¹, Stephen R. Springston¹⁶, R.F. Souza¹⁷, Joel
 7 A. Thornton¹⁸, Gabriel Isaacman-VanWertz¹⁹, Lindsay Yee⁹, Rita Ynoue²⁰, Rahul A. Zaveri¹, Alla
 8 Zelenyuk¹, Chun Zhao^{1,21}

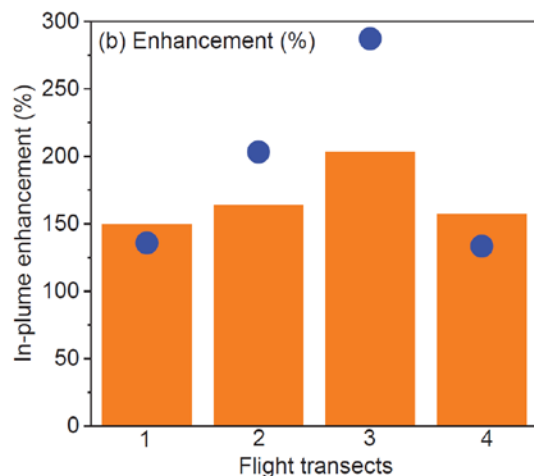
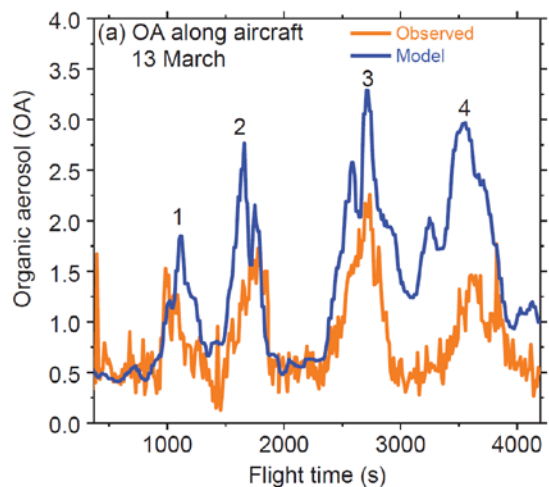
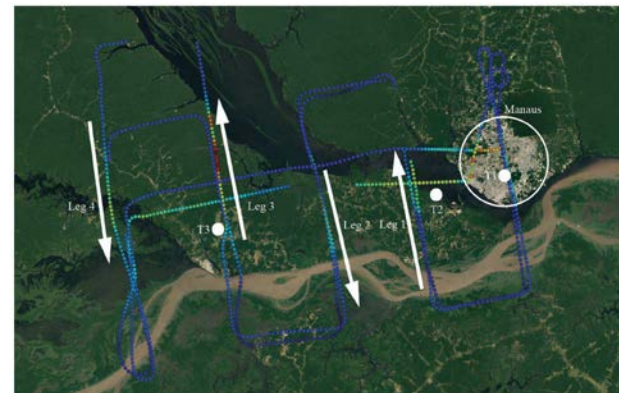
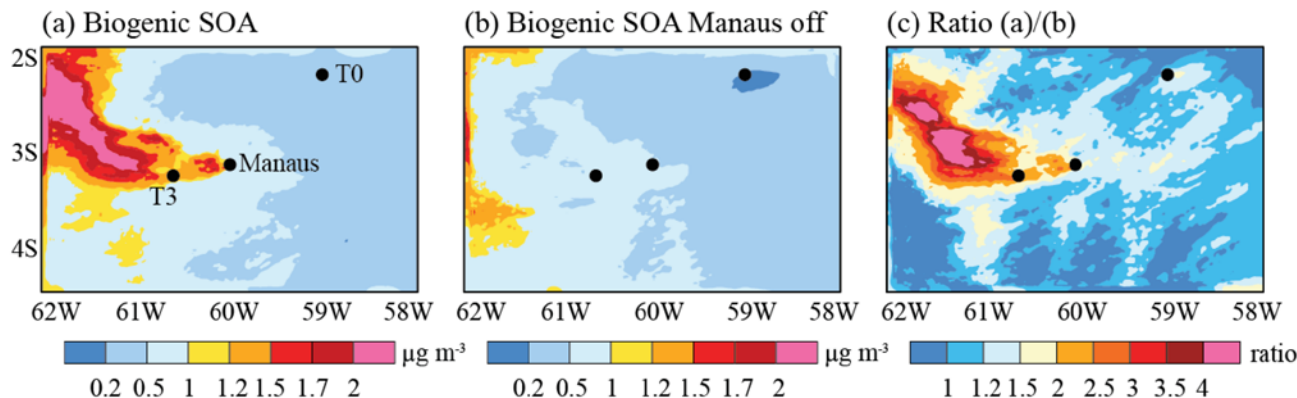


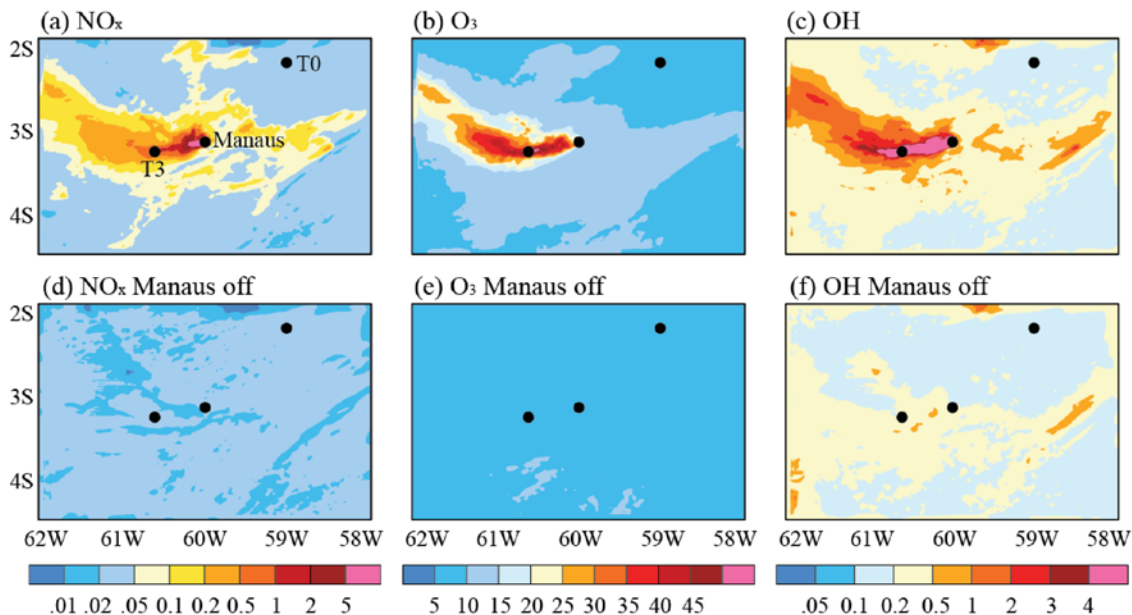
Fig.1. (a) OA measured along aircraft flight transects at 500-m altitude on March 13 (orange) and model-predicted OA, as described in the text. (b) Measured and model predicted average percent enhancement in plume compared to background organic aerosol on 4 different flight transects, as marked in (a). Bars represent measurements while symbols represent a model-predicted increase of OA within Manaus plume compared to background conditions



WRF-Chem simulations showing (a) Biogenic SOA when all emissions are on (b) Biogenic SOA when biogenic VOC emissions are on but Manaus emissions are turned off (c) Biogenic SOA enhancement factor calculated as the ratio of two simulations with Manaus emissions turned on/off i.e. (a)/(b). WRF-Chem predictions are at ~500 m altitude, averaged during the afternoon (16-20 UTC = 12-16 local time) of March 13, 2014.

2 Manish Shrivastava^{1,*}, Meinrat O. Andreae^{2,3}, Paulo Artaxo⁴, Henrique Barbosa⁴, Larry K. Berg⁵,
 3 Joel Brito⁶, Joseph Ching⁶, Richard C. Easter⁷, Jiwon Fan⁸, Jerome D. Fast⁹, Zhe Feng⁹, Jose D.
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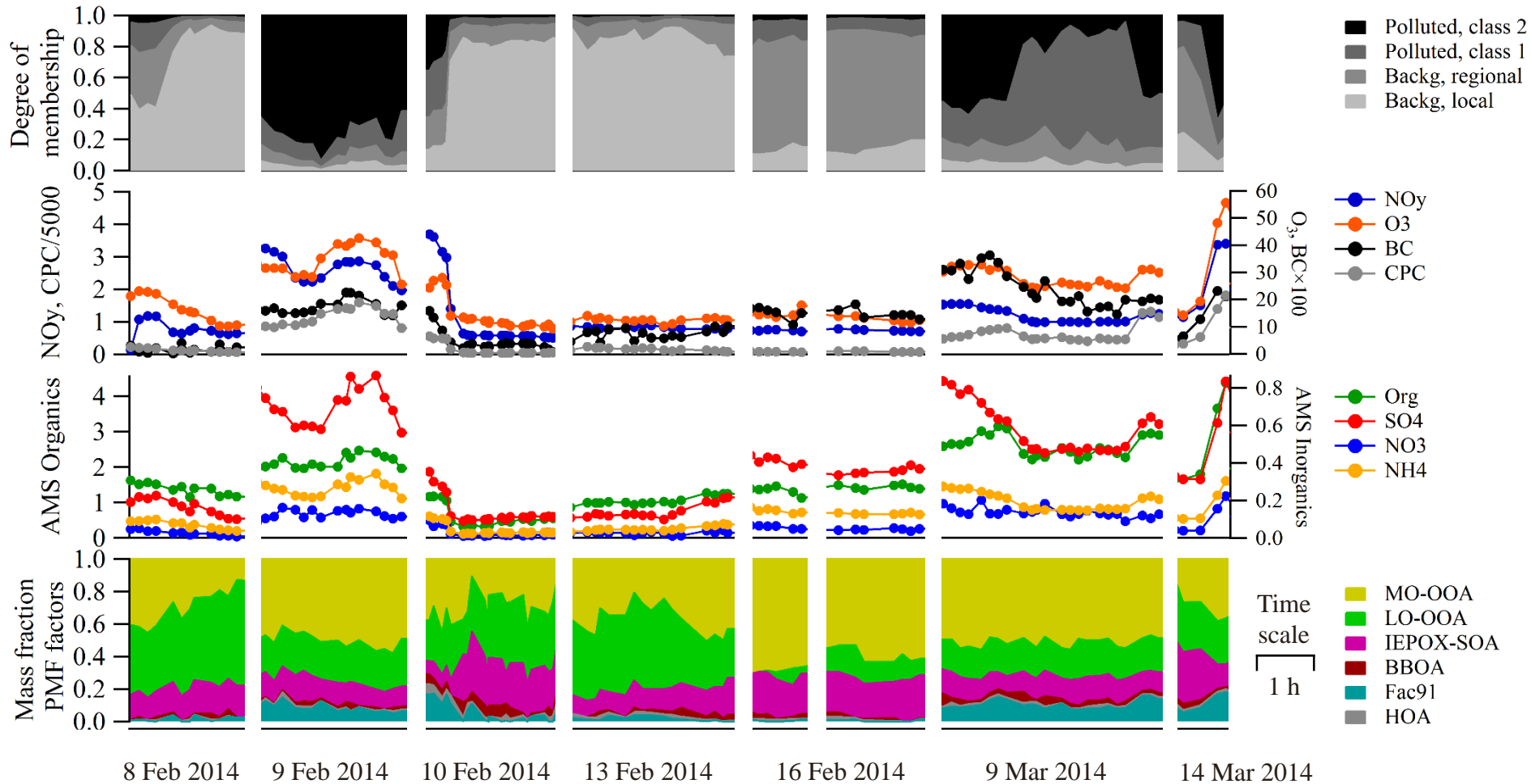
Fig. 3. WRF-Chem simulated concentrations of NO_x (ppbv), O₃ (ppbv) and OH (10⁶ molecule cm⁻³) for an altitude of ~500 m, averaged during the afternoon (16-20 UTC) of March 13, 2014. Top panels show simulations with all emissions on, while bottom panels show simulations with biogenic emissions on but Manaus emissions off. A comparison of top and bottom panels demonstrates how NO_x and oxidants are greatly enhanced by the Manaus plume within the otherwise pristine Amazon



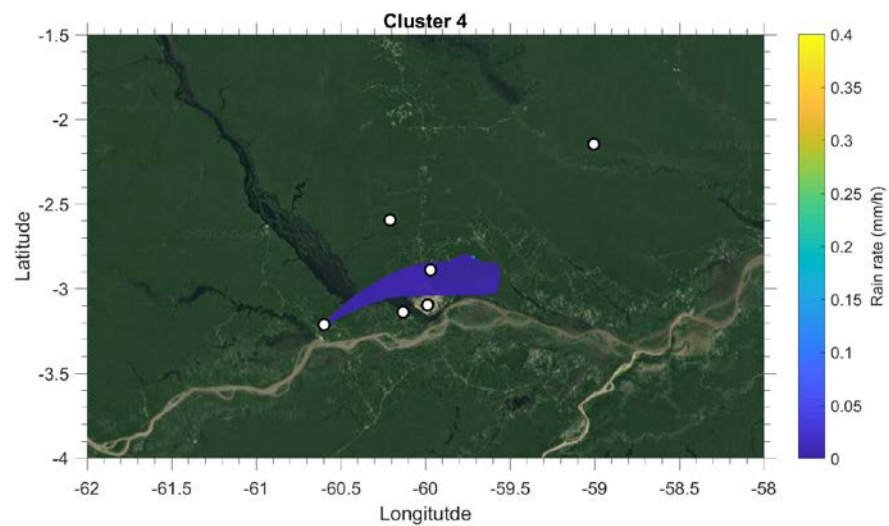
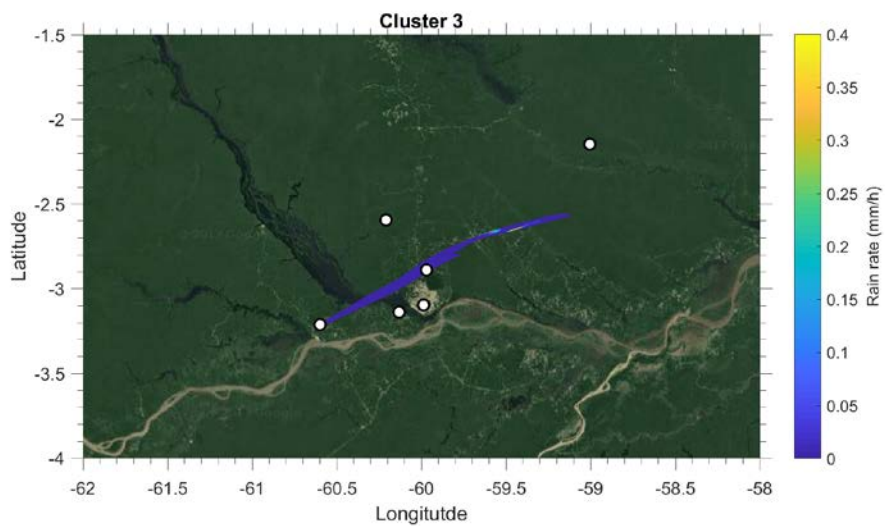
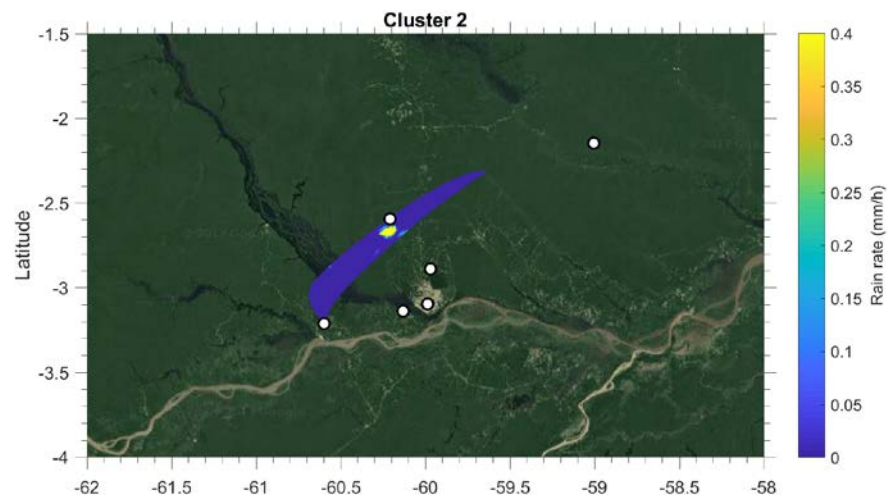
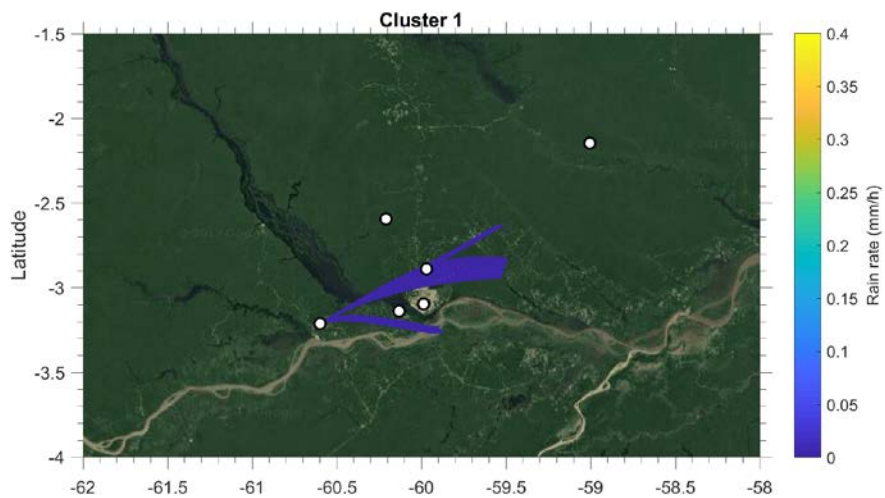
| OA | Measured OA (µg m ⁻³) | | Model OA (µg m ⁻³) | | Percent Enhancement (Plume-Background)/Background*100 | |
|---------------------------|-----------------------------------|------------|--------------------------------|------------|-------------------------------------------------------|-------|
| | In-plume | Background | In-plume | Background | Measured | Model |
| Flight Transects March 13 | | | | | | |
| 1 | 1.2 | 0.5 | 1.2 | 0.5 | 150 | 136 |
| 2 | 1.4 | 0.5 | 1.9 | 0.6 | 164 | 203 |
| 3 | 1.6 | 0.5 | 2.5 | 0.6 | 203 | 287 |
| 4 | 1.3 | 0.5 | 2.8 | 1.2 | 157 | 133 |
| Flight March 14 | 1.6 | 0.9 | 2.1 | 1.2 | 80.2 | 76.5 |
| Flight March 16 | 1.4 | 0.6 | 3.3 | 1.3 | 120.8 | 148.3 |

Aircraft-measured and WRF-Chem simulated median values of absolute OA concentrations in-plume and at background locations. Calculations are shown for individual flight transects on 13 March 2014, since the aircraft clearly identified the plume evolution on this day, as described in the text. An Average across all flight transects is shown for March 14 and 16, and the modeled plume was determined by scanning all radial locations downwind T1 site

Shifts in PM with anthrop. influences



Shifts in aerosols with anthrop. influences



Shifts in aerosols with anthrop. influences

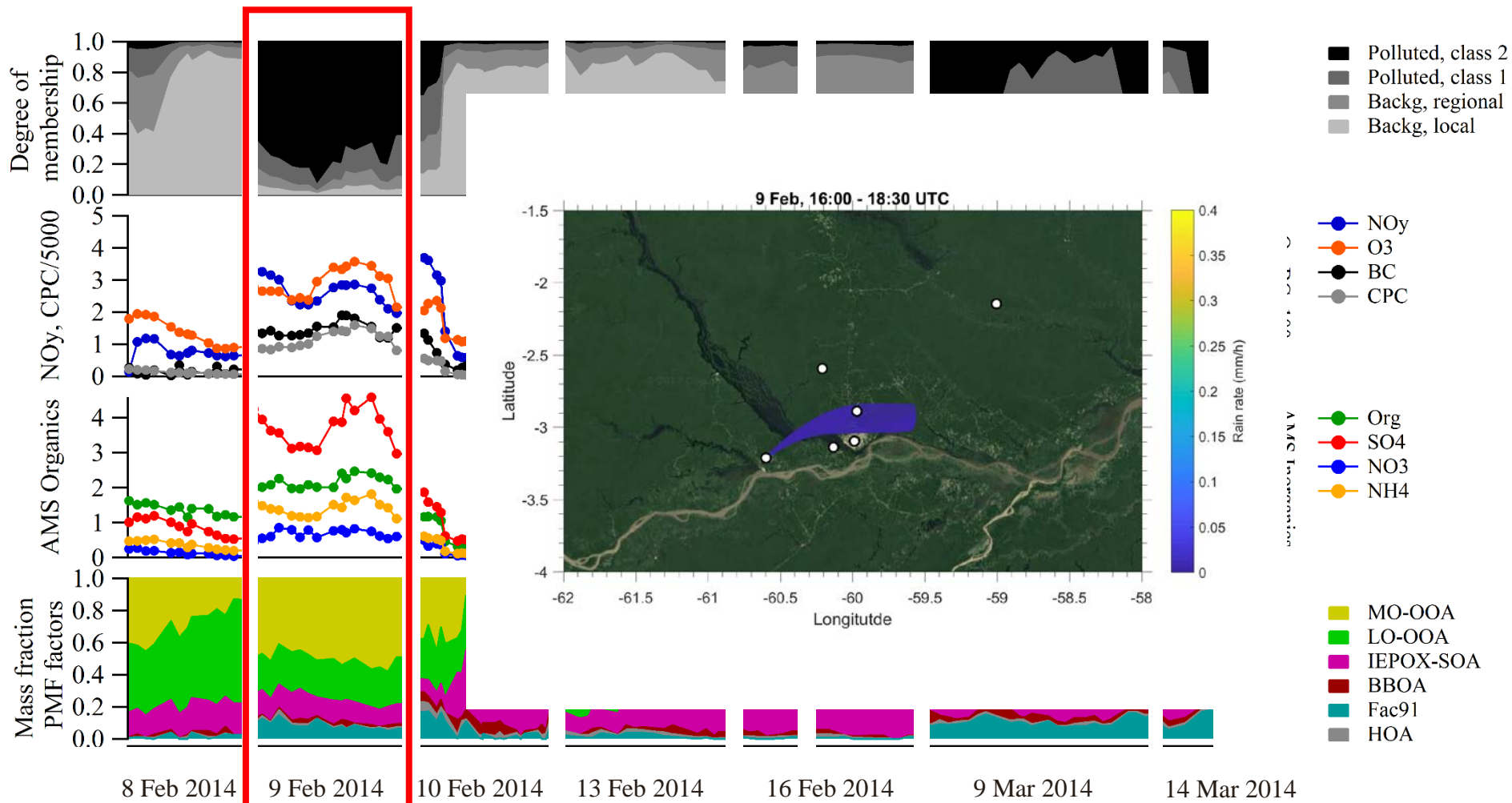


Figure 5

Shifts in aerosols with anthrop. influences

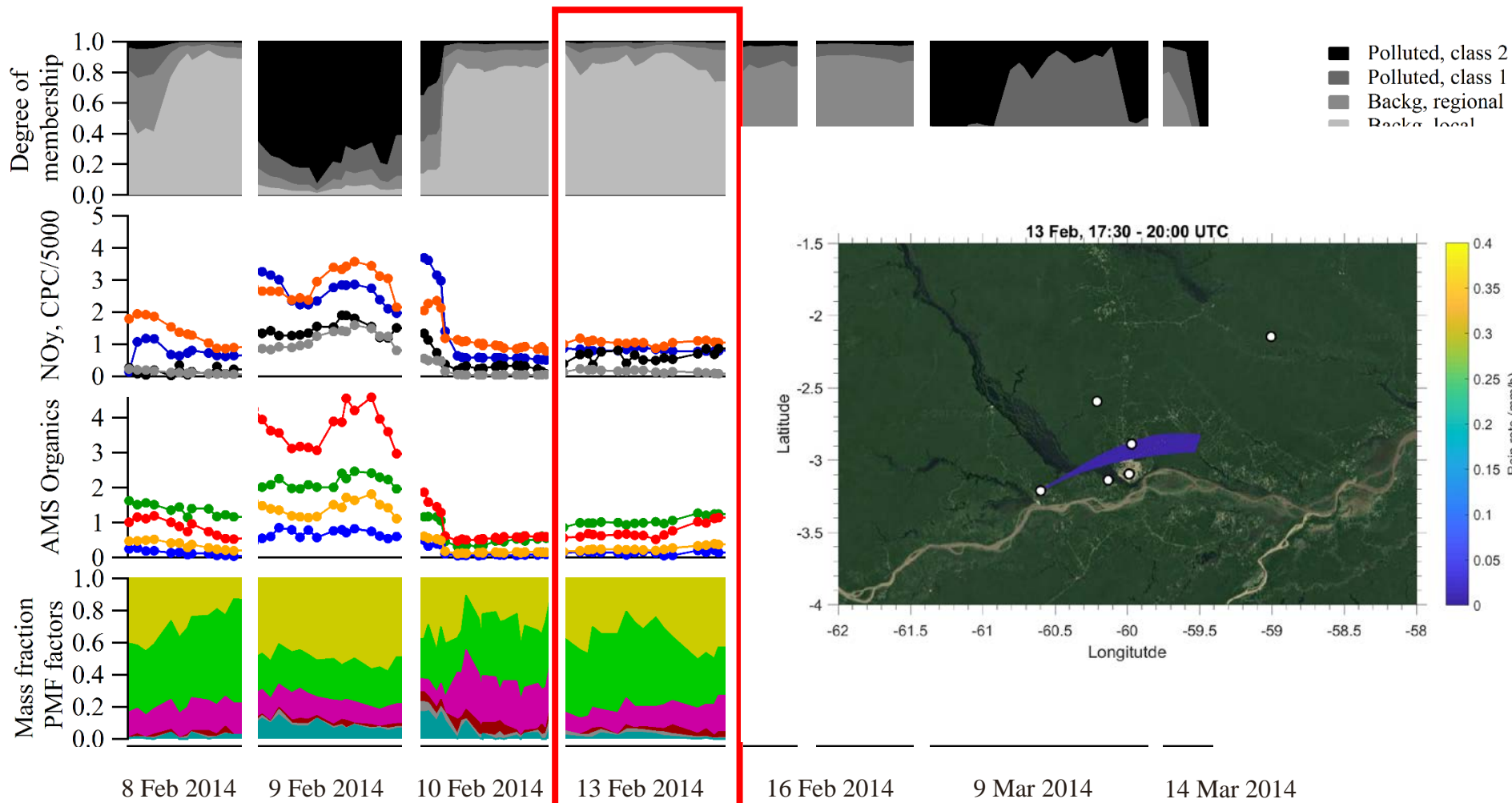
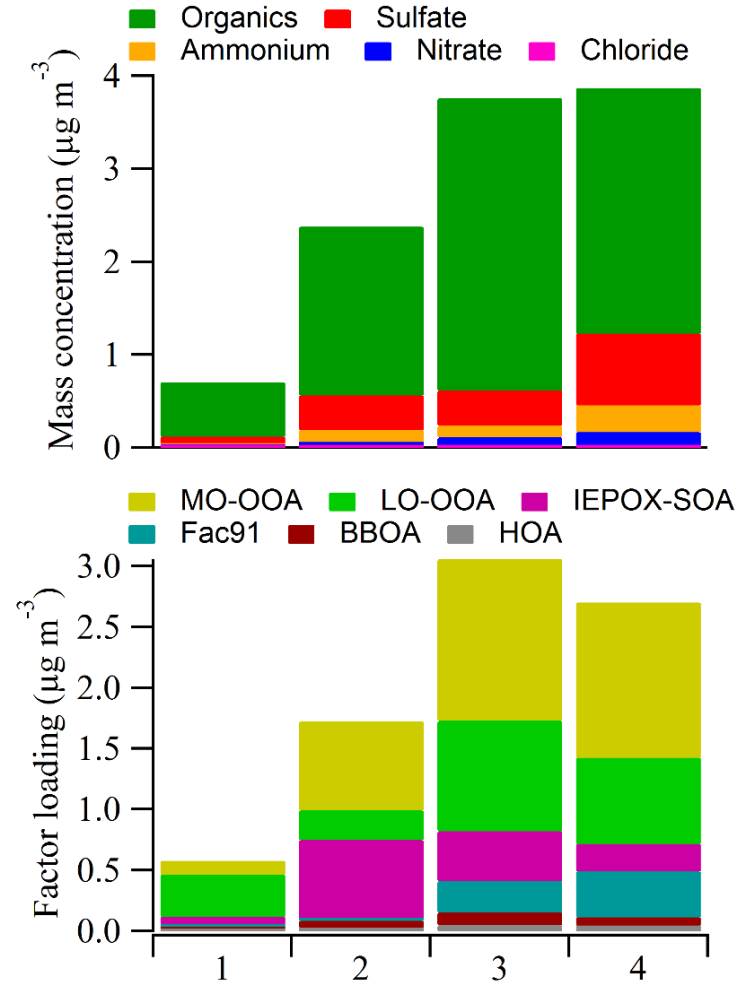
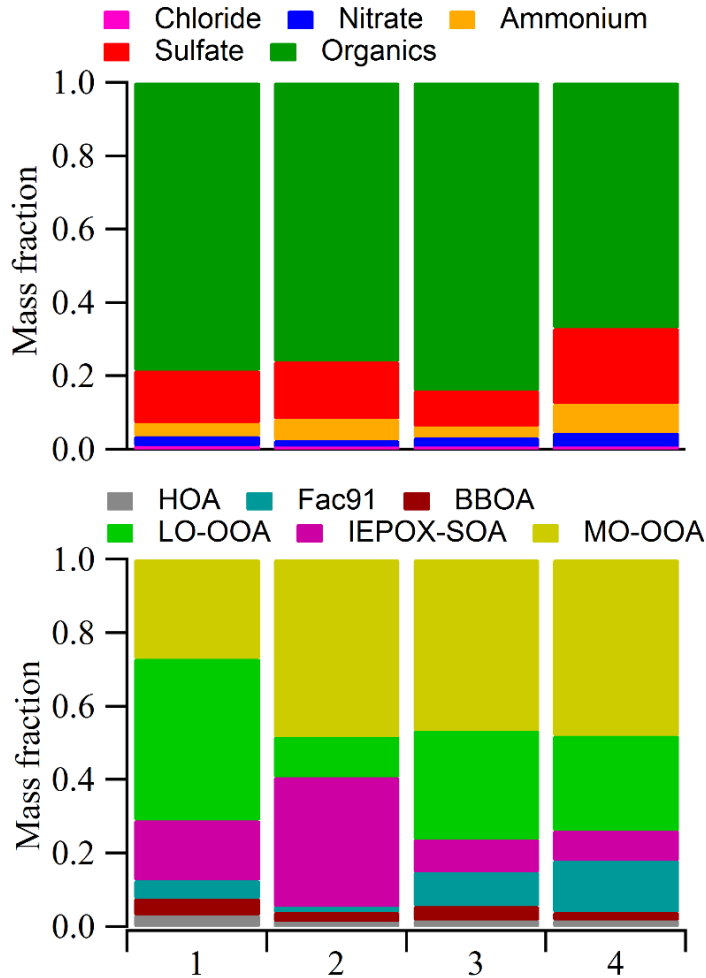


Figure 5

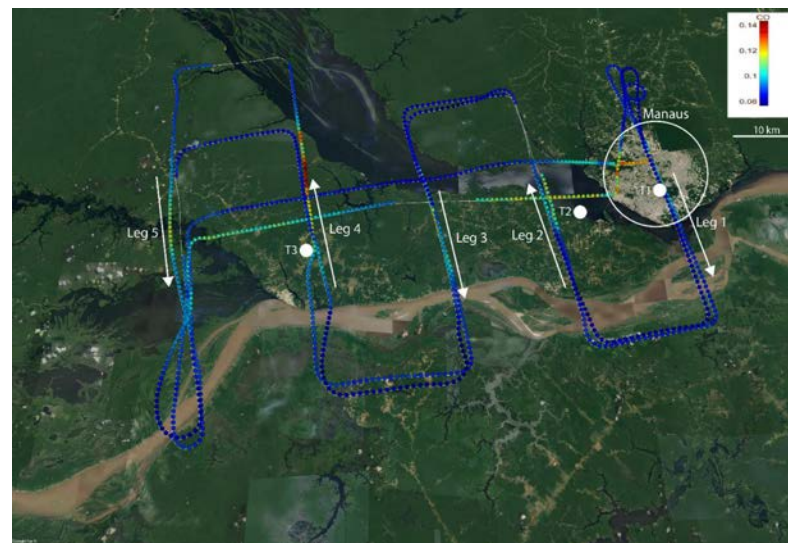
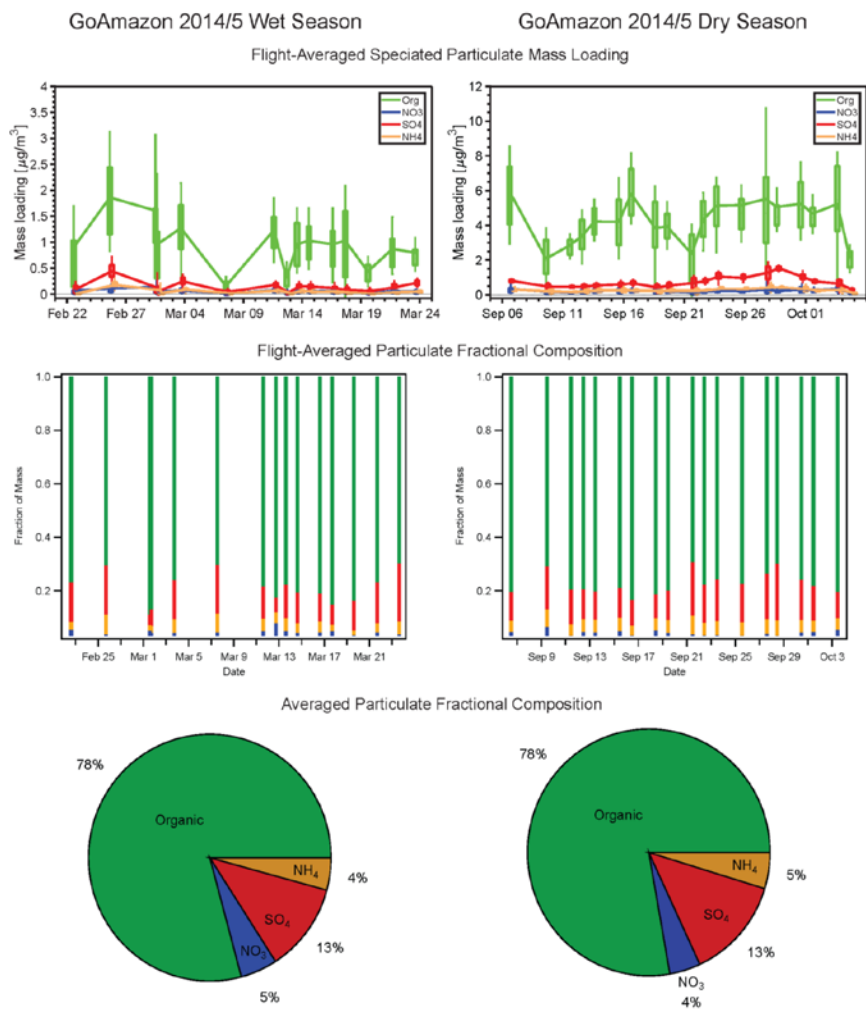
Shifts in aerosols with anthrop. influences



Increase in organic aerosols from <1 to $4 \mu\text{g/m}^3$

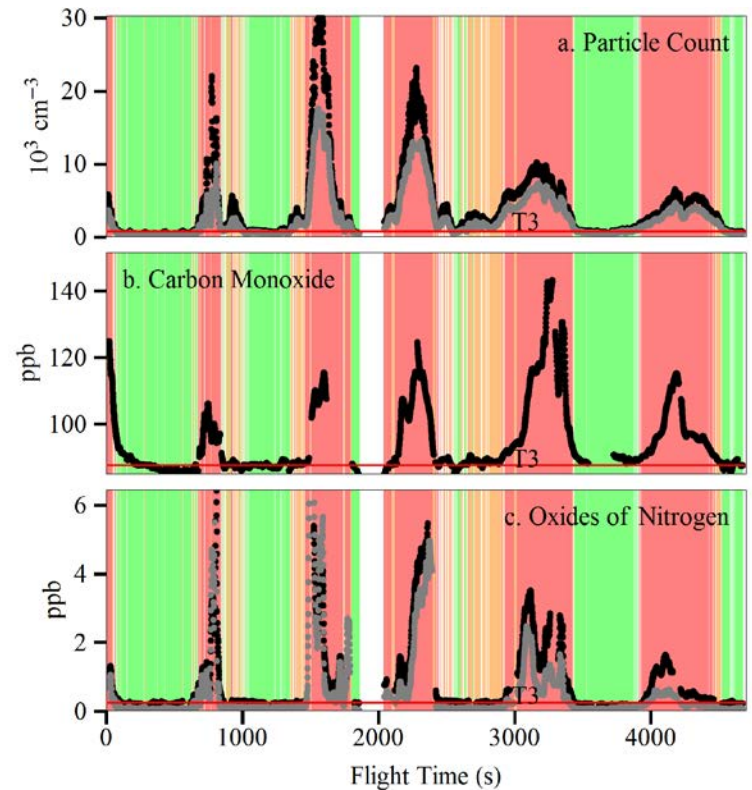
Aircraft Observations of Aerosol in the Manaus Urban Plume and Surrounding Tropical Forest during GoAmazon 2014/15

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Box and whisker representations of the G-1 AMS data for both the wet and dry seasons (top panel), average particle chemical composition for each flight (middle panel), and average aerosol chemical composition for all flights (bottom).

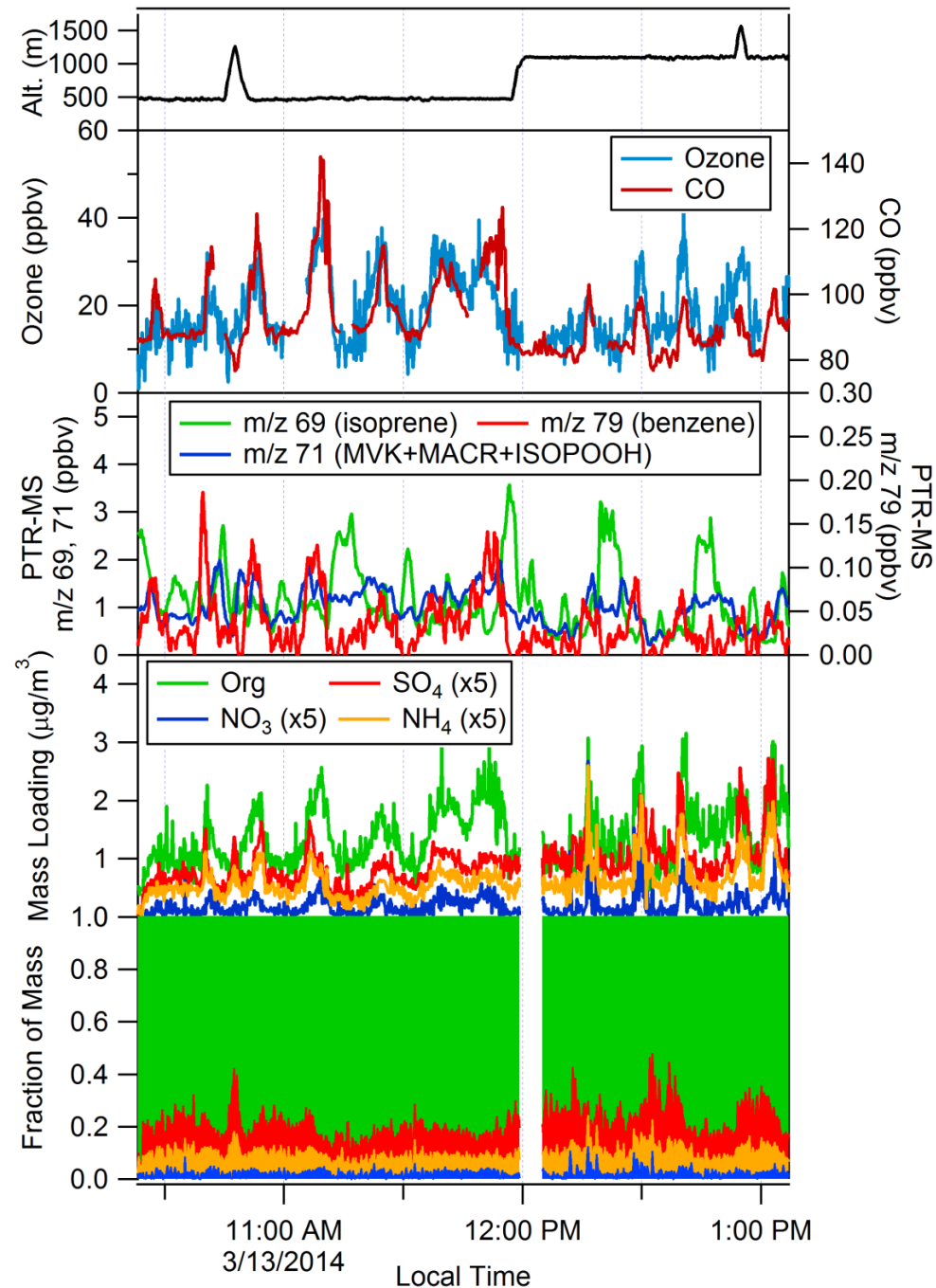
Transverse Transects of Urban Plume *500 m, 11 AM local, 13 March 2014*



Data Sources: Mei Fan, Stephen Springston, IARA Experiment, DOE AAF G1 Platform

Aircraft Observations of Aerosol in the Manaus Urban Plume and Surrounding Tropical Forest during GoAmazon 2014/15

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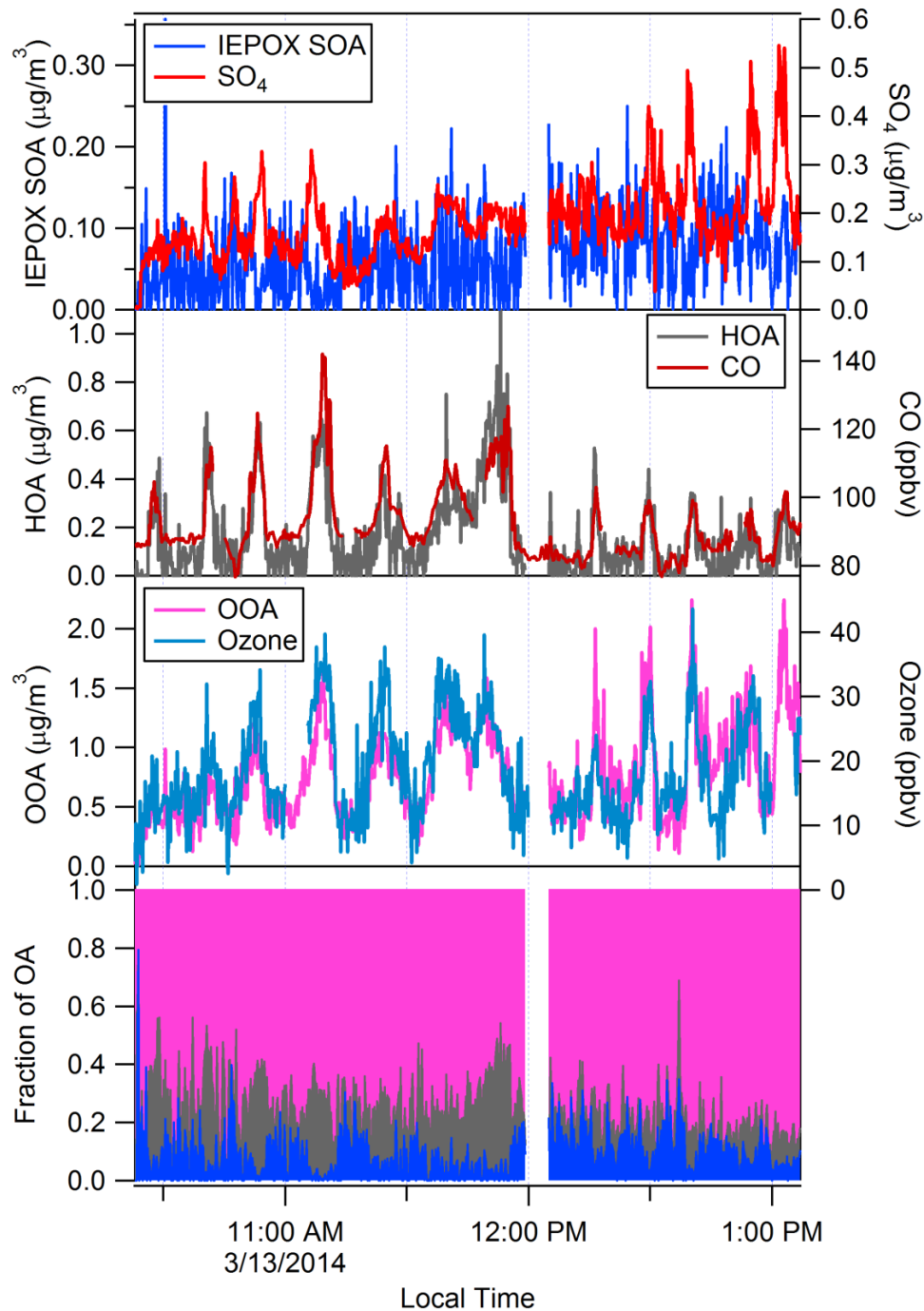


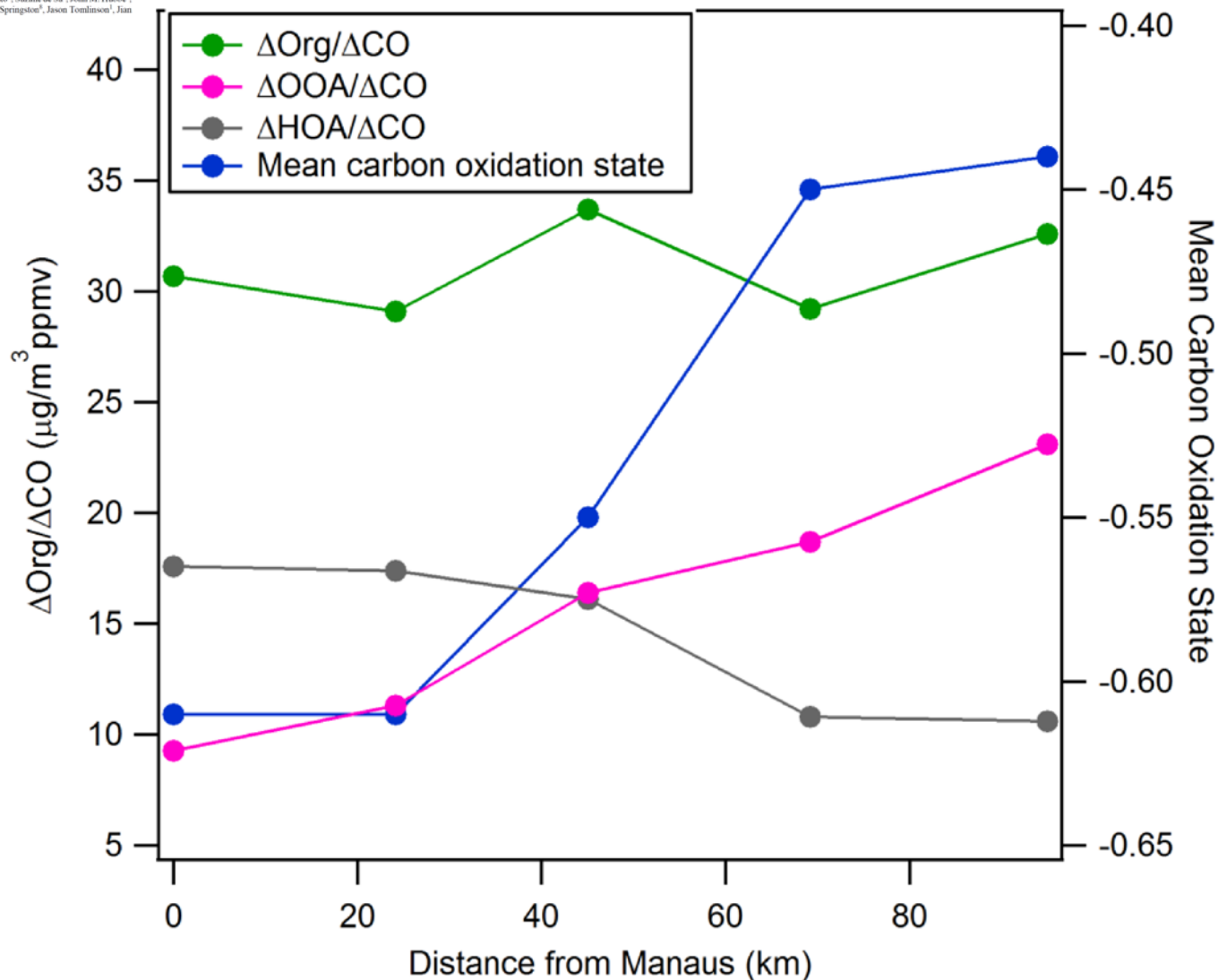
Time traces of relevant quantities on March 13th, 2014 flight. AMS data uses the HR-analysis routine described in the literature to minimize interferences (Aiken et al., 2007). Note the mass of inorganic species (SO₄, NO₃, NH₄) has been multiplied by a factor of five to improve the figure clarity. PTR-MS signal at m/z 69 corresponds to isoprene, m/z 71 corresponds to the sum of methylvinyl ketone (MVK), methacrolein (MACR), and isoprene hydroxyhydroperoxide (ISOPOOH), all oxidation products of isoprene, and m/z 79 corresponds to benzene.

Aircraft Observations of Aerosol in the Manaus Urban Plume and Surrounding Tropical Forest during GoAmazon 2014/15

John E. Shilling¹, Mikhail S. Pekour¹, Edward C. Fortner², Paulo Artaxo³, Suzane de Sá⁴, John M. Hubbe¹, Karla M. Longo⁵, Luiz A.T. Machado⁶, Scot T. Martin^{4,7}, Stephen R. Springston⁸, Jason Tomlinson¹, Jian Wang⁸

PMF analysis of the organic aerosol on the March 13th 2014 flight. The PMF analysis utilized the high resolution dataset. All data from the wet season were included in the analysis.





Key metrics describing the evolution of the Manaus urban plume on March 13. Each data point represents the average values for one pass through the plume. Mean wind speeds were 7 m/s on this flight, thus data capture approximately the first 4-5 hours of the plume aging. Calculations of $\Delta\text{Org}/\Delta\text{CO}$ values use method one with details on the calculations and methods provided in the SI. Spatial locations of each leg are labeled in Figure 1.