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The Southeast Atmosphere Studies (SAS): coordinated investigation and discovery to answer critical questions about fundamental atmospheric processes
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| Abstract: | <p>The Southeast Atmosphere Studies (SAS), encompassing the Southern Oxidant and Aerosol Study (SOAS), the Southeast Nexus of Air Quality and Climate (SENEX) study, the Nitrogen, Oxidants, Mercury and Aerosols: Distributions, Sources and Sinks (NOMADSS) study deployed in the field from June 1 - July 15, 2013 in the central and eastern United States. SAS investigated atmospheric chemistry and the associated air quality and climate implications. Coordinated measurements from six ground sites, four aircraft, tall towers, balloon-borne sondes, existing surface networks, and satellites provide in situ and remotely sensed data on trace-gas composition, aerosol physicochemical properties, and local and synoptic meteorology. Selected SAS findings indicate that (1) dramatically reduced NO_x concentrations have altered ozone production regimes in this region and (2) indicators of 'biogenic' secondary organic aerosol (SOA), once considered part of the natural background, were positively correlated with one or more indicators of anthropogenic pollution, including ozone, sulfate and NO_x. These findings suggest that atmosphere-biosphere interactions modulate ambient pollutant concentrations, and show that the mechanisms and feedbacks are not yet adequately captured in atmospheric models. The SAS data set, now publicly available, is a powerful constraint on our understanding of the response in atmospheric composition to changes in emissions, chemistry and meteorology.</p> |
| Author Comments: | <p>Dear Editor,</p> <p>We upload figure files separately and inside the main text file (for ease of review).</p> <p>Best, -Annmarie (and on behalf of all co-authors)</p> |
| Suggested Reviewers: | <p>Daniel Jacob djacob@fas.harvard.edu Dr. Jacobs has led many field campaigns, in particular when atmospheric chemistry modeling was a strong component.</p> <p>Scot Martin smartin@seas.harvard.edu Dr. Martin led the Go-Amazon campaign which complements and contrasts the studies in the Southeast U.S.</p> <p>Christine Wiedinmyer christin@ucar.edu Dr. Wiedinmyer was involved initial planning for SOAS, a SAS component. She is well qualified to comment on whether science goals were followed and/or achieved.</p> |



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Dear Editor

We are submitting our manuscript concerning the major field campaigns for the summer of 2013. The field data is now public and available to anyone. Data mining by the entire atmospheric science community, in particular, not the chemists, has the power to transform our understanding of atmospheric processes and improve atmospheric models. We intentionally wanted to submit this manuscript to BAMS in order to reach out to the broader, non-chemistry, community. I helped organize a session at the AMS meeting last year as part of my own effort to engage more broadly

We include one paper that is 'in press' and one paper that is 'in review'. The first paper by MacDonald et al. is central to NOAA's SENEX campaign. I understand this is not ideal and will remove the paper and change the manuscript text if it is not sufficiently along during the review process. The second paper by Gratz et al. has been reviewed and the authors have already provided a revised manuscript and response. We suspect that they will hear back a favorable response any day now, and had actually expected it by now, but it is summer.

Sincerely,



Annmarie G. Carlton, Ph.D., P.E.
Associate Professor

The Southeast Atmosphere Studies (SAS): Coordinated investigation and discovery to answer critical questions about fundamental atmospheric processes

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ABSTRACT

The Southeast Atmosphere Studies (SAS), encompassing the Southern Oxidant and Aerosol Study (SOAS), the Southeast Nexus of Air Quality and Climate (SENEX) study, the Nitrogen, Oxidants, Mercury and Aerosols: Distributions, Sources and Sinks (NOMADSS) study deployed in the field from June 1 - July 15, 2013 in the central and

eastern United States. SAS investigated atmospheric chemistry and the associated air quality and climate implications. Coordinated measurements from six ground sites, four aircraft, tall towers, balloon-borne sondes, existing surface networks, and satellites provide *in situ* and remotely sensed data on trace-gas composition, aerosol physicochemical properties, and local and synoptic meteorology. Selected SAS findings indicate that (1) dramatically reduced NO_x concentrations have altered ozone production regimes in this region and (2) indicators of 'biogenic' secondary organic aerosol (SOA), once considered part of the natural background, were positively correlated with one or more indicators of anthropogenic pollution, including ozone, sulfate and NO_x. These findings suggest that atmosphere-biosphere interactions modulate ambient pollutant concentrations, and show that the mechanisms and feedbacks are not yet adequately captured in atmospheric models. The SAS data set, now publicly available, is a powerful constraint on our understanding of the response in atmospheric composition to changes in emissions, chemistry and meteorology.

INTRODUCTION

Reports by the World Health Organization (WHO) (WHO 2014), United Nations (UN) (IRAC 2013) and Intergovernmental Panel on Climate Change (IPCC) (IPCC 2014) indicate that the global population is negatively impacted by climate change and degraded air quality. Development of successful strategies to alleviate the most severe environmental outcomes is mired with uncertainty, in part, due to complex atmospheric processes that are responsible for and respond to pollution. The SAS science

topics were chosen to contribute to elucidating these processes and the feedbacks among them: 1) atmosphere-biosphere interactions, 2) nitrogen chemistry during the day and night, 3) anthropogenic emissions and the related trends in ambient concentrations, 4) atmospheric mercury, 5) gas-phase and multi-phase chemical mechanisms, 6) the organic aerosol budget, and 7) climate-relevant properties of aerosol. Resolving key scientific questions in these topic areas on a time scale relevant to addressing environmental and human-health impacts requires coordination among diverse researchers at different institutions working toward common goals and freely sharing information. Understanding, and recreating in models, the response in ambient concentrations from changes in emissions, chemistry, and meteorology is essential to provide guidance to policymakers who can then develop effective strategies to manage air resources, especially in a changing global climate (Gilliland et al., 2008; Jacob; Winner 2009). Accurate and reliable projections for the future atmosphere require that models not only accurately describe current atmospheric concentrations, but do so for the right reasons. Only through incorporation of the correct mechanisms evaluated against observations can future projections of the impacts from policy, energy, and climate scenarios be considered robust. This overview focuses on the scientific rationale behind the SAS studies and provides a broad description of experimental platforms (Figure 1) with an emphasis on deployment strategy, key findings to date, and strategic goals for coordinated future directions including modeling studies from local to global scales and data mining of the thousands of measurements.

The Southeast United States (SEUS) is an ideal location to investigate the fundamental atmospheric processes that determine climate and air quality. The SEUS has not warmed over the past 100 years contrary to the trend observed in most locations globally (IPCC 2013). The regional anomaly is referred to as the “warming hole” (Meehl et al. 2012; Portmann et al. 2009), though in the past decade the region has appeared to warm (Yu et al. 2014). Atmospheric scientists have discussed a variety of local and large-scale explanations to explain the regional phenomenon, but consensus remains elusive. Changes in soil moisture feedbacks (Pan et al. 2004), circulation modes (e.g., Southern Oscillation) (Portmann et al. 2009), cumulus clouds (Liang et al. 2006), sea surface temperature (Robinson et al. 2002), internal dynamic variability (Kunkel et al. 2006), moisture convergence patterns (Meehl et al. 2012) and land-surface processes (Pan et al. 2013) have been proposed to contribute to the SEUS anomaly. Another hypothesis is that short-lived climate forcers, in particular secondary aerosols (particles that form *in situ* and not directly emitted) impact regional climate of the humid and photochemically active SEUS (Goldstein et al. 2009; Leibensperger et al. 2012). Temporal trends of anthropogenic sulfur-dioxide emissions and the resulting ambient formation of sulfate aerosol are consistent with regional temperature trends (Leibensperger et al. 2012). SOA formed aloft from a combination of anthropogenic and biogenic emissions in the SEUS has also been hypothesized to contribute to summertime aerosol optical thickness in the region (Goldstein et al. 2009; Ford and Heald 2013), potentially influencing temperature (Goldstein et al. 2009; Portmann et al. 2009). The high rates of anthropogenic and biogenic emissions in the SEUS, where conditions favor

rapid gas- and multi-phase photochemistry, facilitates formation of ozone (Chameides et al. 1988) and SOA (Weber et al. 2007).

The Southern Oxidant Study (SOS), the largest previous SEUS intensive, spanned nearly a decade (1990s) involving several sites around Nashville, TN, Atlanta, GA and other locations, as well as the operation of several research aircraft (NCSU, 2016). Analyses of SOS observations led to many fundamental discoveries about mechanisms affecting atmospheric composition, and particularly the importance of biogenic isoprene in regional ozone formation (Chameides et al. 1998). Application of these discoveries redefined air-quality management for ozone in the eastern U.S. The Southeastern Aerosol and Visibility Study (SEAVS) in 1995 in the Great Smoky Mountains National Park (Andrews et al. 2000) contributed to our understanding of regional haze. Subsequent observations of long-term trends, in particular from multi-year monitoring stations (e.g., IMPROVE (IMPROVE, 2016), CSN (U.S.EPA, 2016), SEARCH (ARA, 2016)) have provided constraints on our understanding of atmospheric processes and the models used to describe them, and link sampling intensives to a broader context.

During the 20+ years since SOS, analytical instrumentation for laboratory and *in situ* measurements have vastly improved in temporal resolution, sensitivity, and spectrum of measurable compounds. It is now possible to explicitly quantify a variety of gas- and particle-phase species in near real time, including “sticky” compounds (e.g., glyoxal, and some multifunctional compounds), very short-lived species (e.g., radicals) and gas- and particle-phase oxidation products traceable to specific organic precursors.

Surface- and space-based measurements of aerosol optical depth are now routine and publically available (Holben et al. 1998, Martin et al., 2008), and these platforms have recorded changes in columnar loadings of trace species with geographic coverage not possible from other platforms. During previous SEUS intensive observation periods, the large confluence of such measurement techniques were not available, thus limiting potential discovery.

Thousands of physically and chemically diverse measurements from different surface sites and research aircraft (Table 1) collected during SAS are now available for use by anyone. The data provide a valuable opportunity to evaluate, diagnose and improve understanding of climate and air quality and the representation of specific processes in models of different temporal and spatial scales.

SAS PLATFORM DESIGN AND SCIENCE STRATEGY

SAS measurement platforms were optimized for coordinated investigation of open science questions spanning three component projects: the Southern Oxidant and Aerosol Study (SOAS), the Southeast Nexus (SENEX) campaign, and the Nitrogen, Oxidants, Mercury and Aerosols: Distributions, Sources and Sinks (NOMADSS) experiment (Figure 2). These campaigns were funded by the National Science Foundation, the National Oceanic and Atmospheric Administration, the Environmental Protection Agency, and the Electric Power Research Institute. Air quality modeling was employed to inform the individual ground-site locations based on the likelihood to experience a range of chemical conditions (Carlton et al. 2011), and chemical forecasting

was used to plan aircraft flight patterns. The SAS platforms included heavily instrumented ground sites in Centreville, AL; Birmingham, AL; Look Rock, TN; the Appalachian Atmospheric Interdisciplinary Research (AppalAIR) site near Boone, NC; and Research Triangle Park, NC, in addition to a flux tower in Alabama (Table 1). Four sampling-intensive periods, in which particle samples were collected with higher time resolution, were coordinated among the Centreville, Birmingham, and Look Rock sites. The Centreville supersite (“CTR”) in Brent, AL is the location of a routine monitoring network site, part of the SouthEastern Aerosol Research and Characterization (SEARCH) network. The site provides context to the SAS campaign with over a decade of meteorology, trace gas and particle species concentrations (Hidy et al. 2014). Additionally, four aircraft, NSF/NCAR’s C130, NOAA’s WP-3D (both operated out of Smyrna, TN), Purdue’s Duchess, and SUNY-Stony Brook’s Long EZ measured trace species aloft and helped characterize biogenic emissions, point source emissions, and evolution of chemical species from plumes to regional scales (Figure 2, Table 2). A critical component of the SAS campaign included long (up to 6 weeks) measurement intercomparisons and coordinated calibrations at the main SAS ground site, CTR. Coordinated quality assurance and quality control experiments under field conditions helped to connect past and future campaigns and enhances potential synergies among research groups. Another critical component of SAS was a coordinated post-campaign laboratory experiment (FIXCIT, Focused Isoprene Experiment at the California Institute of Technology) in a controlled environmental chamber to test field hypotheses and investigate measurement discrepancies (Nguyen et al. 2014a). SAS efforts were

complemented by NASA's Studies of Emissions and Atmospheric Composition, Clouds, and Climate Coupling Regional Surveys (SEAC⁴RS) later in the summer.

METEOROLOGICAL AND CHEMICAL CLIMATOLOGY AT THE CENTREVILLE SUPERSITE DURING SAS

We describe conditions in detail at the main ground site in Alabama, where the largest number of measurements were taken. SAS campaign meteorology (Figure 3) was characterized by cooler-than-average temperatures and higher-than-average total rainfall when compared to nearly 120 years of meteorological data for the same time of year. The anomaly for maximum daily temperature in the area was -1.7°C . It was the 11th wettest July since 1895 with a precipitation anomaly of $+8.5\text{cm}$. At CTR specifically, daytime temperatures (6am – 4pm) were uncharacteristically low, outside the 25th percentile median for the last decade (Figure 4a). The CTR site experienced many afternoon convective thunderstorms, and showers that at times were severe.

We describe synoptic meteorological conditions that impacted CTR during SAS (Table S-1). Convective thunderstorms were observed the first day of SAS (June 1, 2013) with nearly 2 cm of rain. Frontal passages and Tropical Storm Andrea to the east brought severe storms and approximately 7.8 cm of rain over a 3-day period (June 5-7). A synoptic quiet period followed, bringing high temperatures and little precipitation. June 13th was marked by a cold-front passage with moderate rain. Another cold-front passage began on June 18th and moved through the area, becoming occluded before moving out of Alabama June 20th. A final occluded front moved in June 29th and affected CTR into early July. A synoptic cold front occurred July 12th. The time period

between fronts was marked by convective storms and rain. Rain during this period was due to moisture from the Gulf of Mexico mixed with convective thunderstorms. July 2013 experienced the 2nd coolest maximum temperatures in the upper plains region of Alabama since 1895.

Four intensive sampling periods were optimized with chemical forecasting; during each, filter-based particle measurements were conducted at higher frequency (every ~3-hours during the day rather than typical 23-hour daily average sampling) in a coordinated fashion across all SAS ground sites. Wind-direction frequencies of each of the four SAS intensive sampling periods (Figure 3) at CTR are unique. West-Southwest (WSW) and South-Southwest (SSW) winds dominated the first intensive sampling period (June 10-12, 2013). The second intensive period (June 14-16, 2013) experienced variable winds from the North, North-Northwest (NNW), and East-Southeast (ESE) sharing near-equal frequency. The third intensive period (June 29 – July 1, 2013) experienced frequent 4-6 m s⁻¹ winds, mainly from the West and West-Northwest (WNW). The final and longest intensive period (July 9-14, 2013) had variable winds, with South and NNW directions exhibiting the highest frequency.

The atmosphere at CTR experienced a range of chemical conditions from clean “background” conditions to episodic spikes in pollutant concentrations from anthropogenic sources. In general, when averaged over the campaign time period, ambient mixing ratios of SO₂, NO_x and O₃ at CTR were lower than the 25th percentiles for the last decade (Figure 4b-d). These lower mixing ratios can be attributed to a general decreasing trend of SO₂ and NO_x emissions in the U.S. traceable to environmental

regulations (e.g. Blanchard et al. 2013), such as Title IV of the 1990 Clean Air Act Amendments (Hand et al. 2012), in addition to increasing energy efficiency and economic recession (U.S. EIA, 2016). During SAS, total daytime $PM_{2.5}$ mass was relatively constant (Figure 4f). Particle organic carbon (OC) concentrations were similar to median concentrations over the previous decade, which have been measured every three days at CTR. Trends in particle mass and OC are in sharp contrast to substantial decreases of SO_2 , NO_x , and O_3 during the previous decade at CTR.

FINDINGS IN SAS SCIENCE TOPICS

The scientific findings from SAS are described in seven different categories, which summarize the underlying science questions and motivating factors of the original campaigns. The topic areas are framed around answerable, open questions, to which substantial leaps in understanding are being made.

1. Atmosphere-Biosphere Interactions

SAS coordinated a comprehensive suite of ecosystem-atmosphere flux measurements on leaf, canopy, landscape, and regional scales. Instruments based on a mobile lift, above-canopy towers, and light aircraft platforms directly measured fluxes across leaf, canopy, and landscape scales at two representative forests (lowland and upland) near CTR. The processes investigated by these local-scale measurements were complemented by flux measurement systems on the NSF/NCAR C130 and NOAA WP-3D aircraft that, along with satellite observations, extend the SAS land-atmosphere exchange measurements to most of the major ecosystems across the SEUS.

The overall atmosphere-biosphere interaction research question addressed by SAS is “what are the magnitudes, variations, and controlling processes for biosphere-atmosphere fluxes of oxidants and reactive carbon and nitrogen across spatial scales relevant for regional models?” Specific SAS scientific questions targeted oxidized VOC deposition, “unknown” biogenic volatile organic carbon (BVOC) emission, BVOC emission response to landcover change, and differences among BVOC emission estimation approaches.

Dry deposition is an important, but poorly quantified, sink for oxygenated VOCs and indirectly for SOA (e.g. Knote et al., 2015). Deposition of oxygenated VOCs, especially isoprene oxidation products, was measured by eddy covariance at both SAS flux tower sites using multiple types of chemical ionization mass spectrometry (CIMS) instruments. The measured deposition rates include compounds whose deposition had not previously been quantified (e.g., hydroxymethyl hydroperoxide, peroxyacetic acid, monoterpene nitrooxy hydroperoxide, and hydrogen cyanide). The results demonstrated that deposition formulations are incomplete in most models, resulting in rates for some compounds that are a factor of 2 or higher than typical values (Nguyen et al. 2015a). These observations also showed the importance of molecular diffusion for the atmosphere-biosphere exchange of water-soluble species.

Comprehensive measurements of leaf, canopy and landscape-level isoprene and monoterpene concentrations and emissions at the two SAS tower sites presented a unique opportunity to constrain the processes controlling BVOC emissions in this region. The earlier SOS studies included only leaf-level emission measurements that

represented just a small fraction of the ecosystem and were extrapolated to the canopy scale to predict fluxes into the above-canopy atmosphere (Guenther et al. 1996). The magnitude and diurnal variations of whole-canopy isoprene and monoterpene emissions measured during SAS compared well with both the extrapolation of leaf-level results and with vertical concentration profiles measured from the canopy to the top of the boundary layer (Su et al. 2016). The extensive eddy covariance isoprene and monoterpene flux measurements conducted during the NCAR C130 flights, and the complementary measurements from the NOAA WP-3D flights, extend these constraints to other SEUS ecosystems (e.g. Kaser et al., 2015).

In addition to reducing the uncertainties associated with deposition and emission rates, accurate VOC flux measurements at multiple heights in the daytime boundary layer were used to constrain oxidation rates through OH concentration estimates. Airborne isoprene flux measurements during SAS were used to estimate boundary layer OH concentrations of $2.8\text{-}6.6 \times 10^6$ molecules cm^{-3} that were within 16% of *in situ* aircraft CIMS measurements (Kaser et al. 2015). The reverse approach was used during the 1990s SOS campaigns to estimate isoprene and monoterpene emissions using measured isoprene and monoterpene concentrations combined with highly uncertain OH concentration estimates (Guenther et al. 1996). The comprehensive SAS emissions, chemistry and dynamics observations also directly showed, for the first time, that the surface heterogeneity of isoprene emissions leads to a physical separation of isoprene and OH resulting in an effective slowdown of isoprene loss rates by up to 30% (Kaser et al. 2015).

2.) Nitrogen Chemistry during Day and Night

NO_x regulates the rates and pathways of atmospheric oxidation, impacting ozone, aerosols and climate. The SAS campaign brought a wide range of established and new technologies and to study NO_x chemistry in the presence of high BVOC concentrations, and provided an opportunity to explore changing NO_x chemistry as a result of dramatic decreases of anthropogenic emissions in the SEUS during the past two decades. Most of the region, in particular rural areas, was often in a chemical regime where small variations in NO_x affected the fate of peroxy radicals, and thus ozone and aerosol production efficiency, providing an opportunity to investigate the oxidative chemical mechanisms in the presence of high isoprene. At the higher concentrations that were characteristic of the region at the time of SOS and SEAVS in the 1990s, peroxy radical chemistry was dominated by NO_x and thus insensitive to variations in its concentration. Observations from SAS provided fresh insight into NO_x lifetime as a result of better characterized production and sinks during day and night.

During SAS, daytime NO_x ranged from 1-5 ppb over cities and 0.015-0.1 ppb in rural areas. Rural concentrations were sufficiently low that less than half of the peroxy radicals formed from VOC oxidation reacted with NO_x. The fate of isoprene nitrates is not yet well understood but was a major research emphasis for SAS. A key issue is the extent to which isoprene (and monoterpene) nitrates resulted in rapid removal of oxidized N from the reactive pool vs. return to the atmosphere as NO_x, locally or downwind.

Novel mass spectrometric instruments at CTR measured speciated organic nitrogen in both the particle and gas phases (Lee et al. 2016). These data showed that organic nitrates are highly functionalized in the particle phase, that isoprene and monoterpene derived organic nitrates have distinct diurnal profiles consistent with their emission profiles, implying that particle-phase organic nitrates are, on average, short lived (2-4 hours). A separate study also concluded that organic nitrates had average lifetimes shorter than 2 hours, with their hydrolysis accounting for the largest source of HNO_3 . Production and loss of organic nitrates was the primary control for the lifetime of NO_x at CTR, estimated as 11 ± 5 hours at midday (Romer et al., 2016). A GEOS-Chem analysis of ground-based SOAS and aircraft SEACRS data estimated that isoprene nitrates account for 25-50% of total organic nitrates in surface air, and that aerosol uptake followed by hydrolysis to HNO_3 accounts for 60% of gas-phase organic nitrate loss in the boundary layer in the SEUS (Fisher et al., 2016). A separate study, however, found consistency between chamber measurements of the photochemical yield of isoprene nitrates (9%) and their observed ratio to other isoprene oxidation products at CTR with no hydrolysis loss (Xiong et al., 2015).

Nighttime BVOC oxidation through the nitrate radical, NO_3 , was a significant focus of SAS. The NOAA WP-3D executed three dedicated night flights as part of SENEX, and there were several analyses from the SOAS ground site of the role of NO_3 -monoterpene reactions as an SOA source. Analysis of aerosol composition data suggests that a third of the organic aerosol mass at CTR was attributable to NO_3 -monoterpene reactions (Xu et al. 2015a). A molar yield of 23-44% particle-phase

monoterpene nitrates from monoterpene oxidation by NO_3 was estimated, with little evidence for particle-phase isoprene nitrates (Ayres et al. 2015).

There were also several analyses of inorganic nitrogen chemistry. Particle-phase inorganic nitrate, for example, was found to be associated principally with sea salt and dust as a result of the high acidity of submicron aerosols that precluded the formation of ammonium nitrate (Allen et al. 2015; Guo et al., 2015). Speciated amines partitioned favorably to the particle phase (gas:particle amine mass ratio was ~ 0.01) and comprised approximately 2% of the sum of particle phase ammonium and amine mass (You et al., 2014). Based on aircraft data from the NSF C-130, an upper-limit yield of nitrous acid (HONO) was estimated to be 0.03 for the reaction between $\text{HO}_2 \bullet \text{H}_2\text{O}$ and NO_2 , too low for the reaction to be an important daytime HONO formation mechanism in the troposphere (Ye et al. 2015). Finally, comprehensive airborne observations over the North Atlantic Ocean provided evidence for rapid recycling of HNO_3 to HONO and NO_x in the marine boundary layer via particulate nitrate photolysis (Ye et al. 2016). The finding was further corroborated by laboratory experiments and a box model based on the Master Chemical Mechanism (Ye et al., 2016). Since oceans account for over 70% of Earth's surface, particulate nitrate photolysis could be a substantial tropospheric NO_x source. Recycling of NO_x in remote oceanic regions with minimal direct NO_x emissions could increase formation of tropospheric oxidants and SOA mass on a global scale.

Anthropogenic emissions and trends in ambient concentrations

Accurate knowledge of current emissions and trends for reactive trace gases, aerosol, and greenhouse gases in the SEUS is needed to understand atmospheric chemical transformations and predict trends in air quality and climate. Emissions of NO_x , SO_2 , Hg and VOCs from major US urban areas and power plants have decreased as evidenced by surface and satellite observations (Hand et al. 2013; Warneke et al. 2012) and these emission reductions are evident in the SEUS. During SAS, the NSF/NCAR C-130 and NOAA WP-3D research aircraft quantified emissions from a variety of sources in the SEUS, including urban areas; point sources (power plants, coal mines, animal husbandry, and biofuel refineries); oil and natural gas production; and agricultural burning. These measurements were interpreted in the context of previous intensives and long-term monitoring datasets to address key science questions, which were (1) What are the emissions from the main sources in the SEUS? (2) How well do current emission inventories represent these emissions? (3) How have anthropogenic emissions of gases and aerosols (SO_2 , NO_x , VOCs, NH_3 , PM, Hg etc.) recently changed? (4) How have the atmospheric concentrations of primary and secondary pollutants responded to these changes in emissions? (5) What are the implications for air quality and climate in the region? (6) What were the drivers of these emission changes (e.g. emissions changes driven by control programs, source activity changes, or economic shifts)?.

Urban areas emit NO_x , CO, Hg and VOCs, due to high densities of vehicles and industries. The SAS aircraft measurements can be compared to previous campaigns, in particular the SOS flights in 1999. Mixing ratios of anthropogenic pollutants over urban areas such as Atlanta have been greatly reduced since then (Warneke et al., 2016). Analysis of

SEARCH measurements showed that mobile source emissions dropped by 59%, 49% and 51% for NO_x, VOC and PM_{2.5}, respectively, from 1999-2013 in a region that included Alabama, Georgia, Mississippi and Northwestern Florida (Blanchard et al. 2013b; Hidy et al. 2014). In addition, mobile source NO_x emissions were investigated using a fuel-based approach, where emissions were estimated using fuel use data and emission factors from *in situ* measurements. Compared to the NEI-2011, the fuel-based inventory decreased U.S. NO_x emissions by ~60% for on-road gasoline vehicles only, by ~30% for all mobile sources (on-road + off-road), and by ~20% for all anthropogenic sources (mobile + stationary) (McDonald et al., in prep.). Modeling O₃ in the SEUS using NEI-2011 during SAS resulted in large over-predictions, which were reduced with updated emission values (McDonald et al., in prep.). In addition, NO_x emissions were investigated using the GEOS-Chem chemical-transport model by comparing to observations made during the SEAC⁴RS campaign. It was found that the NEI-2011 NO_x emissions were too high and had to be reduced by 60% for all sources except for power plants to reconcile modeled NO_x and its oxidation species (Travis et al 2016). These overpredictions highlight a need for more thorough understanding of emissions models, and the design of field studies that isolate specific source sectors.

Power plant Environmental regulation and the large scale transition from coal-fired to natural gas power plants in recent years has resulted in large emission reductions of many species including SO₂ (de Gouw et al. 2014) and Hg across the SEUS. Since SOS, examples for the impacts of power sector emission changes can be seen in sulfate mass reductions (Blanchard et al. 2013a; Hand et al. 2012), decreased aerosol optical depth

(Attwood et al. 2014), and a potentially controlling effect on SOA mass as discussed below.

Oil and natural gas production emits CH₄, VOCs, and NO_x. CH₄ emissions determined by eddy covariance (Yuan et al. 2015) and a regional mass budget (Peischl et al. 2015) showed leak rates of 0.3%, 1.6%, and 1.9% for the Marcellus, Haynesville, Fayetteville shale gas production regions, respectively. These leak rates were substantially lower than earlier basin-wide estimates from Colorado and Utah (Petron et al., 2012, Karion et al., 2013).

Other point source emissions were quantified using aircraft, where for example de Gouw et al. (2015) examined ethanol and other trace gas emissions from a biofuel refinery. While emissions of SO₂ and NO_x agreed with emissions reported to the NEI-2011, emissions of several VOCs, including ethanol, formaldehyde, and acetaldehyde, were underestimated by an order of magnitude.

Biomass burning emissions, mainly from agricultural burning, is another important source for aerosol in the SEUS (Blanchard et al. 2016). Washenfelder et al. (2015) found that biomass burning aerosol dominates organic aerosol absorption (brown carbon; BrC) absorption in rural Alabama at the SOAS site in 2013, although BrC levels were very low.

Mercury Mercury (Hg) is a bioaccumulative neurotoxin and is globally distributed, and there are large uncertainties in the atmospheric processing and global budgets. Understanding the sources and chemistry of atmospheric mercury (Hg) was one of the goals of NOMADSS. The mercury-specific goals were: 1) constrain emissions of Hg from

major US source regions; 2) understand the origin of oxidized Hg in the free troposphere; 3) quantify the distribution of speciated Hg in the troposphere and compare with global model simulations; and 4) develop a calibration method using HgBr₂ for a commercial Hg instrument at the Birmingham, AL SEARCH site.

Mercury observations on the C-130 made using the Detector for Oxidized Hg Species (DOHGS) (Ambrose et al., 2015), measured both gaseous elemental mercury (Hg⁰), Gaseous Oxidized Mercury (GOM), plus a fraction of particle-bound oxidized Hg. GOM is believed to consist of Hg(II) compounds, such as HgCl₂, HgBr₂, etc. A key finding was that the NEI-2011 reflected Hg emission slightly better than the Toxic Release Inventory, but for some large coal-fired power plants (CFPP), total Hg emissions inferred from ambient measurements were higher than the NEI estimates (Ambrose et al 2015). Gratz et al. (2016) also evaluated Hg emission inventories, but for the Chicago-Gary metropolitan area. Their observations similarly suggested that the emission inventory is biased low, in part due to underestimated CFPP emissions, and also due to many small sources missing from the inventory.

Because of the importance of Hg⁰ oxidation in the global Hg cycle, a key NOMADSS goal was to understand the distribution and chemistry of GOM in the atmosphere. For this reason, NOMADSS flights sampled air in the mid-upper troposphere over the SEUS. In this region, prior studies and NOMADSS forecasts (Shah et al., 2016) suggested the presence of high GOM concentrations. On one NOMADSS flight, very high concentrations of GOM (0.266 ng m⁻³) were detected and,

simultaneously, BrO was significantly elevated (up to 1.9 pptv). Gratz et al. (2015) used these observations with a chemical box model and found strong support for the formation of HgBr₂ (and possibly other GOM compounds) by Br-initiated oxidation. Other mechanisms appeared unable to explain high GOM concentrations. Using the GEOS-Chem chemical transport model, Shah et al. (2016) provided further support for the bromine mechanism and demonstrate that BrO mixing ratios in the model are likely too low. They further showed that the highest GOM concentrations were predicted to occur in subsiding air in subtropical anticyclones, due to fast production and slow removal. Song et al. (2016) also used the NOMADSS observations with the GEOS-Chem model and suggested the northwestern Atlantic is a net source of Hg⁰, with large fluxes in summer, likely due to strong wet deposition of Hg in this region. In contrast, terrestrial ecosystems in the eastern U.S. during summer are likely a net sink of Hg⁰. These results are consistent with the constraints on surface-atmosphere cycling of mercury found using a global suite of observations and an inverse-modeling approach (Song et al., 2015).

Finally, ground-based observations of Hg provided new insights into our ability to measure Hg⁰ and GOM using commercial instrumentation. McClure et al. (2014) installed a novel system to measure total atmospheric mercury and a calibration system for GOM alongside a standard commercial instrument (Tekran Instruments Corp., Toronto, CA). The commercial system could measure GOM accurately in zero air, but suffered from significant interference at ambient ozone and humidity. This work

indicated the need for improved GOM calibration methods and further development of atmospheric Hg instrumentation.

Gas-phase and multi-phase chemical mechanisms

The SAS campaign brought together the latest instrumentation to address the following questions pertaining to gas and multiphase atmospheric chemistry: 1) What are the chemical and physical processes that control the oxidation of BVOC? 2) How do anthropogenic influences alter the distribution of the BVOC oxidation products, and what are the implications for the formation of ozone, reactive nitrogen and aerosol? 3) How does aqueous chemistry alter the fate of BVOC oxidation products and the formation of SOA?

Over the last decade, there have been major revisions in description of the chemical mechanisms that connect the release of alkenes from the biosphere with the formation of oxidants and aerosols. These include discovery of (1) chemically labile epoxide (Paulot et al., 2009a, Lin et al., 2013a) and lactone atmospheric intermediates (Nguyen et al., 2015b) and their heterogeneous reactions to form SOA (Surratt et al., 2010; Lin et al. 2012; Lin et al., 2013a; Nguyen et al., 2015b); (2) generation of low-volatility oxygenated hydrocarbons via autooxidation (Peeters et al. 2009; Crouse et al. 2013; Ehn et al. 2014); (3) more complete description of organic nitrate formation and loss (Xie et al. 2013 Lee et al. 2014); and (4) recognition of the impact of aqueous phase chemistry on oligomer, organosulfate, and secondary organic aerosol formation (Lin et al., 2014; Gaston et al. 2014; Nguyen et al. 2014b)--connecting back to earlier SEUS studies (Blando and Turpin 2000).

A central design goal of SAS was to make use of state-of-the-art analytical instrumentation to evaluate current chemical mechanisms for accuracy and completeness. Coincident deployment of multiple instruments provided for evaluation of these new techniques while deployment on many platforms including aircraft, towers, and ground installations allowed for regional characterization. In some cases, controlled experiments were conducted with ambient air.

The CTR and LRK sites experienced isoprene-dominated biogenic emissions, with high co-emission of monoterpenes, substantial aerosol liquid water concentrations (Nguyen et al, 2014), low aerosol pH (Guo et al., 2015), and a range of oxidation conditions implicated in the formation of biogenic aerosol. These surface sites also experienced varying degrees of anthropogenic pollution influence. Both sites are bordered by areas of high- and low-NO_x concentrations.

Many of the instruments deployed to CTR also participated in the FIXCIT post-campaign laboratory chamber study that simulated daytime and nighttime oxidation of biogenic alkenes under NO and HO₂ dominated regimes (Nguyen et al. 2014a). These experiments provided a key tie between tabulated mechanisms and the field observations. Experiments at FIXCIT also tested for consistency of various instrumentation (e.g. evaluated calibration and interferences for field instrumentation) (Rivera-Rios et al., 2014).

Preliminary results from SAS suggest several prominent types of interactions between biogenic organic and anthropogenic (SO₂ and NO_x) emissions including, for example, interactions leading to the formation of organonitrates (see previous section)

and organosulfates (Budisulistiorini et al. 2015; Boone et al. 2015). Oxidation in urban plumes was faster compared to the isoprene-dominated background (Kaiser et al. 2015). However, Kaiser et al., (2015) found that the glyoxal to formaldehyde ratio was not a reliable indicator of anthropogenic versus biogenic VOC mix, contrary to previous suggestions. Isoprene epoxydiol-derived SOA (IEPOX-SOA) was correlated with sulfate ($r^2=0.6$) (Budisulistiorini et al., 2015; Xu et al., 2015), and high IEPOX-SOA episodes were associated with westerly (anthropogenically influenced) flows in comparison to flows from the more rural and biogenic south and southeast (Budisulistiorini et al., 2015). Despite laboratory evidence that the rate of IEPOX-SOA formation is faster on acidic sulfate than ammonium sulfate (e.g., Riedel et al. 2015), IEPOX-SOA at LRK did not show a dependence on *local* pH. It has been postulated that this discrepancy may be because proton activity is not rate limiting in the SEUS or in the IEPOX reaction (Nguyen et al., 2014b; Xu et al., 2015). Or perhaps formation occurs regionally and thus is insensitive to *local* conditions (Lin et al., 2013b). Additionally, although some liquid water is needed for the formation of IEPOX-SOA, the SOA mass was shown to be weakly, and negatively, correlated with aerosol water consistent with the conflicting effects of higher IEPOX accommodation but lower inorganic ion activity as water increases (Gaston et al., 2014; Nguyen et al., 2014b). Ambient observations at SAS show poor correlation between IEPOX-SOA and aerosol water, possibly due to differences in the ionic activity of the ambient aerosol water sampled with diurnal and spatial variations. Future mechanism development will benefit from collaborative analyses of

the rich array of real-time measurements of intermediate and highly oxidized BVOC products, oxidant species and emission fluxes and shared scientific objectives.

Organic aerosol budget

A major fraction of fine PM mass worldwide and in the SEUS consists of OA (e.g. Zhang et al. 2007). Many sources contribute to ambient OA, their relative impacts are poorly understood, and predictions of OA mass by global models can disagree by more than an order-of-magnitude (Tsigaridis et al., 2014). OA sources are divided into primary (POA), which includes anthropogenic emissions (e.g. vehicles, cooking), biomass burning sources, and secondary (SOA). SOA forms in the atmosphere from anthropogenic, biogenic, and biomass burning precursors. The formation of SOA from BVOCs is influenced from anthropogenic pollutants, as discussed above. Globally and in the SEUS, BVOC emissions far outweigh anthropogenic sources, and most SOA is expected to form from biogenic VOCs (Lewis et al., 2004; Kim et al., 2015). SOA from biomass burning typically imparts a small net increment to OA mass (Cubison et al., 2011).

Many recent and rapid advances in OA- and precursor-measurement techniques were deployed during SAS (e.g. Lee et al., 2014; Isaacman et al., 2014; Krechmer et al., 2015, 2016; Lopez-Hilfiker et al., 2016; Martinez et al., 2016), and these studies conclude that SOA from biogenic VOCs indeed made substantial contributions to total OA during SAS. The isoprene epoxydiol pathway (IEPOX-SOA; Paulot et al., 2009b) contributed about 17 and 32% (on average) of the OA at CTR (Xu et al., 2015a; Hu et al., 2015) and LRK (Budisulistiorini et al., 2015), respectively. A larger contribution was

observed during warmer SAS periods, suggesting the unusually cool conditions during SAS led to lower IEPOX-SOA compared to typical summers (Marais et al., 2016; Budisulistiorini et al., 2016). Explicitly measured molecular tracers accounted for 85% of total IEPOX-SOA (Hu et al., 2015), which represents an unprecedented level of molecular closure. Many of these IEPOX-SOA tracers were observed to partition between the gas and particle phases, with a large fraction of those in the particle phase in the form of oligomers or accretion products that decompose during analysis in most instruments (Lin et al., 2014; Lopez-Hilfiker et al., 2016; Hu et al., 2016). Other pathways forming SOA from isoprene via low-NO oxidation were estimated to account for several percent of the OA at CTR and LRK (Krechmer et al., 2015; Marais et al., 2016; Riva et al., 2016). SOA from high-NO_x pathways (Lance et al., 2014; Nguyen et al., 2015b) is thought to be important (Kim et al., 2015); however, molecular tracers were less abundant (Rattanvaraha et al., 2016). Isoprene-derived SOA via glyoxal is also thought to contribute several percent to the total OA in the SEUS summer (Knote et al., 2014; Li et al., 2016). Marais et al. (2016) estimate 3.3% of reacted isoprene in the SEUS forms SOA through all the pathways.

SOA from monoterpene oxidation was also important during SAS, in particular, by oxidation of the NO₃ radical (Xu et al., 2015a; Ayres et al., 2015; Lee et al., 2015) and by other oxidants (Kim et al., 2015). Many of these oxidation productions were observed to be partitioned between the particle and gas phases (Isaacman-VanWertz., 2016). NO₃-monoterpene SOA is present during all seasons, in contrast to isoprene-derived SOA, which peaks in the summer (Xu et al., 2015b; Budisulistiorini et al, 2016). The

contribution of sesquiterpenes is thought to be smaller, but has not yet been directly quantified for SAS. SOA production in clouds (as opposed to in aqueous aerosols) was limited according to Wagner et al. (2015), who analyzed 74 aircraft vertical profiles in the region and concluded that differences in SOA mass concentrations above and below clouds were not statistically significant.

Past studies showed that SOA in the Southeast US correlates well with anthropogenic pollution tracers, suggesting formation of SOA from BVOCs may be controlled by anthropogenic emissions (Weber et al. 2007; Carlton et al. 2010). SAS results shed light on this issue. NO_x from anthropogenic sources (mainly vehicles and power plants) controls the rate of oxidation of BVOCs (de Gouw et al 2015b) and strongly modulates isoprene and monoterpene SOA as discussed above. Anthropogenic SO_2 emissions (mainly from power plants) control aerosol sulfate, acidity (Guo et al., 2015) and water uptake (Carlton and Turpin 2013; Nguyen et al. 2014c) and thus strongly influence IEPOX-SOA formation (Xu et al., 2015a; Budisulistiorini et al., 2015; Marais et al., 2016). Formation of organosulfates is an anthropogenic-biogenic interaction leading to SOA (Surratt et al., 2008), but contributed small fractions to the total OA and sulfate mass (Budisulistiorini et al., 2015; Hettiyadura et al., 2015; Liao et al., 2015; Rattanavaraha et al., 2016).

Overall SAS results are consistent with the hypothesis that anthropogenic emissions control a major fraction of OA in the SE US (Carlton et al., 2010; Xu et al., 2015a; Marais et al., 2016). The dependencies are complex and non-linear. Modeling

studies that incorporate SAS findings, e.g. Pye et al. (2015), estimate that a 25% reduction in NO_x emissions leads to a 9% reduction in OA in the SEUS. Marais et al. (2016) estimates that the EPA-estimated emission reductions by 2025 (34% for NO_x and 48 % for SO₂) will lead to a 36% reduction in total isoprene SOA, driven primarily by reductions in SO₂.

Other sources of OA were also important. The amount of anthropogenic POA and SOA were estimated to be 9% and 18% of the total OA (Kim et al., 2015). These results are consistent with the 18% fraction of fossil carbon at CTR, since only 50% of the anthropogenic POA and 70% of the anthropogenic SOA is fossil (Hayes et al., 2015). Kim et al. (2015) also reported a contribution of 11% for biomass-burning OA at CTR, consistent with a measurement-based estimate of 10% (Xu et al., 2015a). Budisulistiorini et al. (2016) did not detect a biomass-burning factor at LRK. Aging of OA in the atmosphere blurs the source-specific chemical signatures and makes it difficult to ascertain the sources of some OA (Jimenez et al. 2009; Xu et al. 2015b). Trends measured by long-term networks indicate decrease in OA mass in the SEUS and this provides a helpful constraint on sources and effects (Blanchard et al. 2013; Hand et al. 2013; Nguyen et al., 2015; Attwood et al., 2015).

Climate-relevant properties of aerosol

Particles in the atmosphere are an important component of the global climate system through direct interactions with incoming solar radiation and indirectly by acting as cloud condensation nuclei (CCN). CCN impact cloud droplet number and modulate cloud reflectivity, development, and lifetime. There is considerable uncertainty in the

magnitude of these climate impacts. The key climate-relevant characteristics of the aerosol--size, concentration, optical properties, and water uptake--are governed by gas- and particle-phase physicochemical processes that may occur after emission, including new particle formation (NPF). The number of CCN is affected primarily by primary aerosol emissions (and the associated size distributions of these primary particles), NPF, the growth of new and primary particles to climate-relevant sizes, and particle hygroscopic properties; these processes are tightly coupled to the production of condensable organic and inorganic gas-phase species from oxidation of biogenic and anthropogenic precursors. The spatial distribution of the key aerosol properties described above must be known to accurately--especially in the vertical-- calculate their radiative effects. Calculating these properties globally requires understanding of vertical transport, heterogeneous processing in clouds, variation in oxidative chemistry, and the spatial distribution of relative humidity.

SAS science plans and implementation were guided by overarching scientific questions related the interaction of aerosols and climate, including:

1) What are the extinction, absorption and cloud nucleating properties of aerosol produced from anthropogenic and biogenic sources in the SEUS? How do these properties vary over space and time, and relate to chemical composition, size distribution and RH?

2) What fraction of organic aerosol is natural versus controllable, and what are the impacts of anthropogenic sources on the regional radiation budget and on cloud properties and lifetime?

3) How do anthropogenic and biogenic emissions, impact NPF and the subsequent growth of ultrafine particles to CCN sizes? How does particle chemical composition affect CCN activity? What are the likely effects on cloud properties, extent, and lifetime?

4) Given that black carbon is co-emitted with other species, will controlling BC sources in the SEUS (e.g., diesel emissions, agricultural burning) have a net warming or cooling radiative effect?

5) How will these findings change in the future as a result of warming and changing anthropogenic emissions?

SAS provided a unique opportunity to study aerosol characteristics and processes related to their interaction with radiation and clouds. Instruments specifically for measuring aerosol optical and cloud-nucleating properties were operated at CTR, Look Rock, and on the NOAA WP-3D aircraft (Table 1). The ground sites provided detailed information on aerosol optical properties at multiple wavelengths, including spectrally resolved measurements in the near UV, measurements of BC concentration, mass, and coating thickness, and aerosol optical and physical characteristics following humidification and volatilization. The hygroscopic growth of the particles, aerosol liquid water (ALW) and size-dependent cloud-activation potential was determined, along with the water-soluble component of the aerosol. Aerosol volatility and its link to chemical composition and cloud activation potential was also measured. These observations were coupled with a number of detailed aerosol compositional measurements, which link the

optical and cloud-nucleating properties of the aerosol with biogenic and anthropogenic precursors and atmospheric gas- and aqueous-phase chemistry.

Aircraft measurements included aerosol optical properties and variation with humidity and thermal volatility. Variation in cloud-nucleating properties of particles as a function of water supersaturation, BC concentration, and the thickness and hygroscopicity of coatings on BC particles was studied. NOAA WP-3D measurements mapped the spatial variation of climate-relevant aerosol characteristics, especially in the vertical.

During SAS there was little evidence that particulate BrC from BVOC oxidation is a significant contributor to aerosol absorption in the SEUS. Absorption was associated almost entirely with BC from combustion; the small amounts of BrC were mostly associated with biomass burning. ALW played a crucial role in aerosol light scattering, and organic species contributed to ALW at CTR, particularly at night (Guo et al., 2014).

The vertical structure of the daytime planetary boundary layer and RH was found to be important for total AOD and radiation budget. Wagner et al. (2015) found that aerosol mass in the transition, or cloud layer, portion of the boundary layer, contributed significantly to AOD, but could not account for discrepancies in seasonal AOD cycle remotely sensed by satellites and *in situ* surface measurements (Ford and Heald 2013; Goldstein et al. 2009). AOD was sensitive to the RH profile and to the mean diameter and width of the size distribution, and less so to aerosol refractive index and hygroscopicity parameterization (Brock et al., 2016a,b). Diel cycle measurements at CTR showed that organic fraction hygroscopicity exhibited variations with particle diameter

and source origin. No positive correlation was found between organic hygroscopicity and oxygen-to-carbon ratio. Aerosol heating through a thermal denuder had little effect on hygroscopicity, even when 30% of the aerosol mass was volatilized (Cerully et al. 2014).

FUTURE DIRECTIONS

For a study this large in scope and ambition, it is crucial to collectively analyze findings and insure an integrative process among the entire atmospheric science community. Air quality trends and the SEUS warming hole provide excellent context for broad coordinated study among the different atmospheric communities, and SAS provides a high quality data set for sophisticated data mining techniques that can cost-effectively facilitate advances in science (Committee, 2016). The large SAS dataset, now open to the public and accessible through links listed in Table 1, provides valuable observational constraints for uncertain model parameters and processes. Like the field campaign, modeling experiments going forward should be coordinated around cohesive questions, such as: What are the relative roles of anthropogenic and uncontrollable near-term climate forcings and large-scale dynamics on temperature trends in the SEUS? Does the warming hole persist, is it influenced by human activity, is the region now warming consistent with global trends? Reconciling model discrepancies related to emissions, chemical pathways, local- and large-scale dynamics is necessary to move forward. Continued coordinated study through data analysis and model development

and application is key to determine the most-effective strategies to manage air quality, particularly in a changing global climate.

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Table 1. SAS Platform Locations and Descriptions

| SURFACE PLATFORMS | | | |
|---------------------------|------------|------------------|---|
| Latitude | Longitude | Elevation | Description |
| 32.90289 | -87.24968 | 126 | “CTR” Site chemical speciation, chemically-characterized gas and aerosol measurements, detailed meteorology Link to data: http://www.esrl.noaa.gov/csd/groups/csd7/measurements/2013senex/ |
| 32.69456 | -87.2488 | Multiple heights | Flux tower at Alabama Aquatic Biological Diversity Center. Fluxes, soil temperature, radiation Integrated Surface Flux Facility (ISFS): https://www.eol.ucar.edu/observing_facilities/isfs Link to data: http://www.eol.ucar.edu/node/654 |
| 32.892747 | -87.248515 | Multiple heights | meteorology at surface, soundings, LIDAR Integrated Sounding System (ISS); https://www.eol.ucar.edu/instrumentation/sounding/iss Link to data: http://www.eol.ucar.edu/node/654 |
| 35.63314 | -83.94185 | 802 | “Look Rock (LRK)” Surface chemical speciation Link to data: http://www.esrl.noaa.gov/csd/groups/csd7/measurements/2013senex/ |
| 35.889455 | -78.874694 | 325 | “RTP” Surface chemical speciation Link to data: http://www.esrl.noaa.gov/csd/groups/csd7/measurements/2013senex/ |
| 36.21 | -81.69 | 1000m | “Appal AIR” Surface chemical speciation |
| AIRCRAFT PLATFORMS | | | |

| | |
|----------------------------|--|
| NOAA WP-3D | Flight tracks in Figure 5a Link to data: http://www.esrl.noaa.gov/csd/groups/csd7/measurements/2013senex/ |
| NSF C130 | Flight tracks in Figure 5b Link to data: http://www.esrl.noaa.gov/csd/groups/csd7/measurements/2013senex/ |
| Purdue Duchess | Link to data: http://www.esrl.noaa.gov/csd/groups/csd7/measurements/2013senex/ |
| Stony Brook Long EZ | Link to data: http://www.esrl.noaa.gov/csd/groups/csd7/measurements/2013senex/ |

Southeast Atmosphere Studies



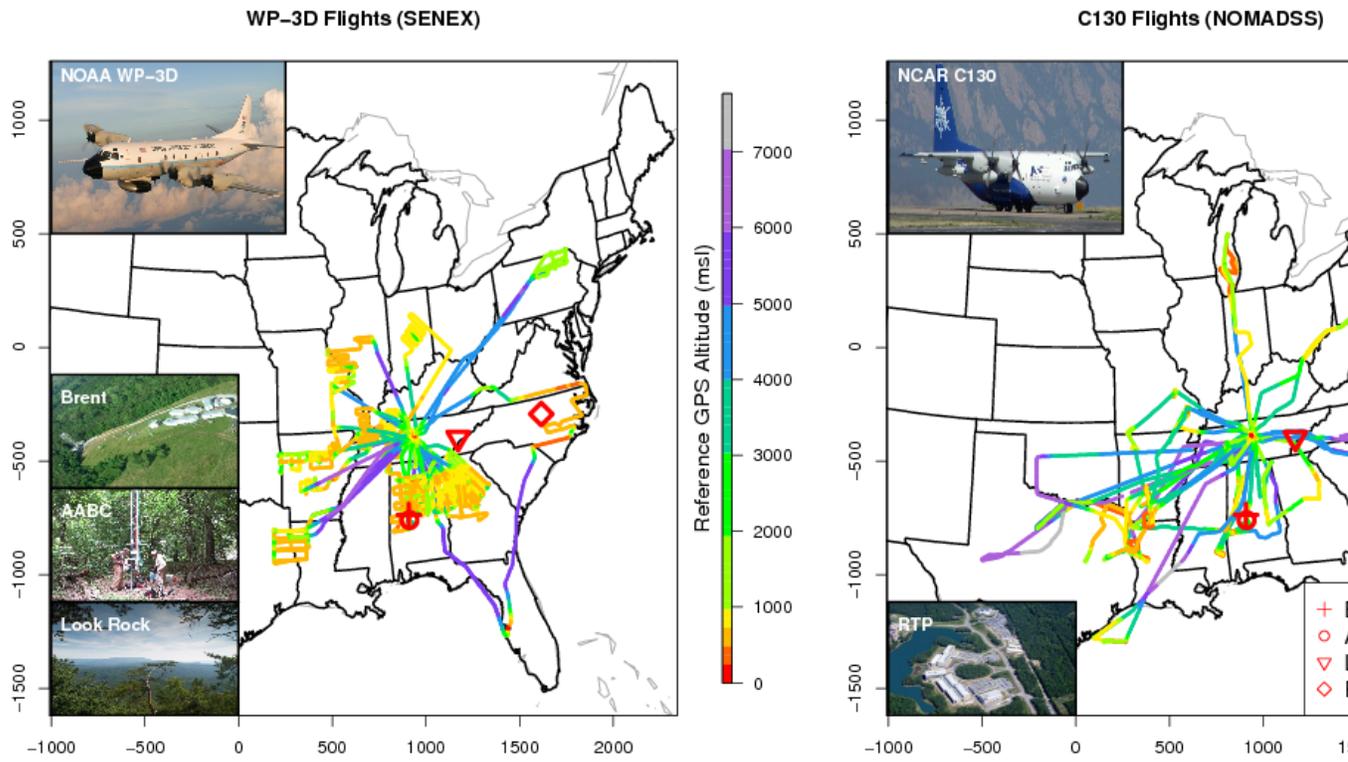


Figure 1. SAS Platforms and Locations. Flight tracks are shown for NOAA WP-3D aircraft (b) during SENEX and NSF/NCAR C130 during NOMADSS (c).

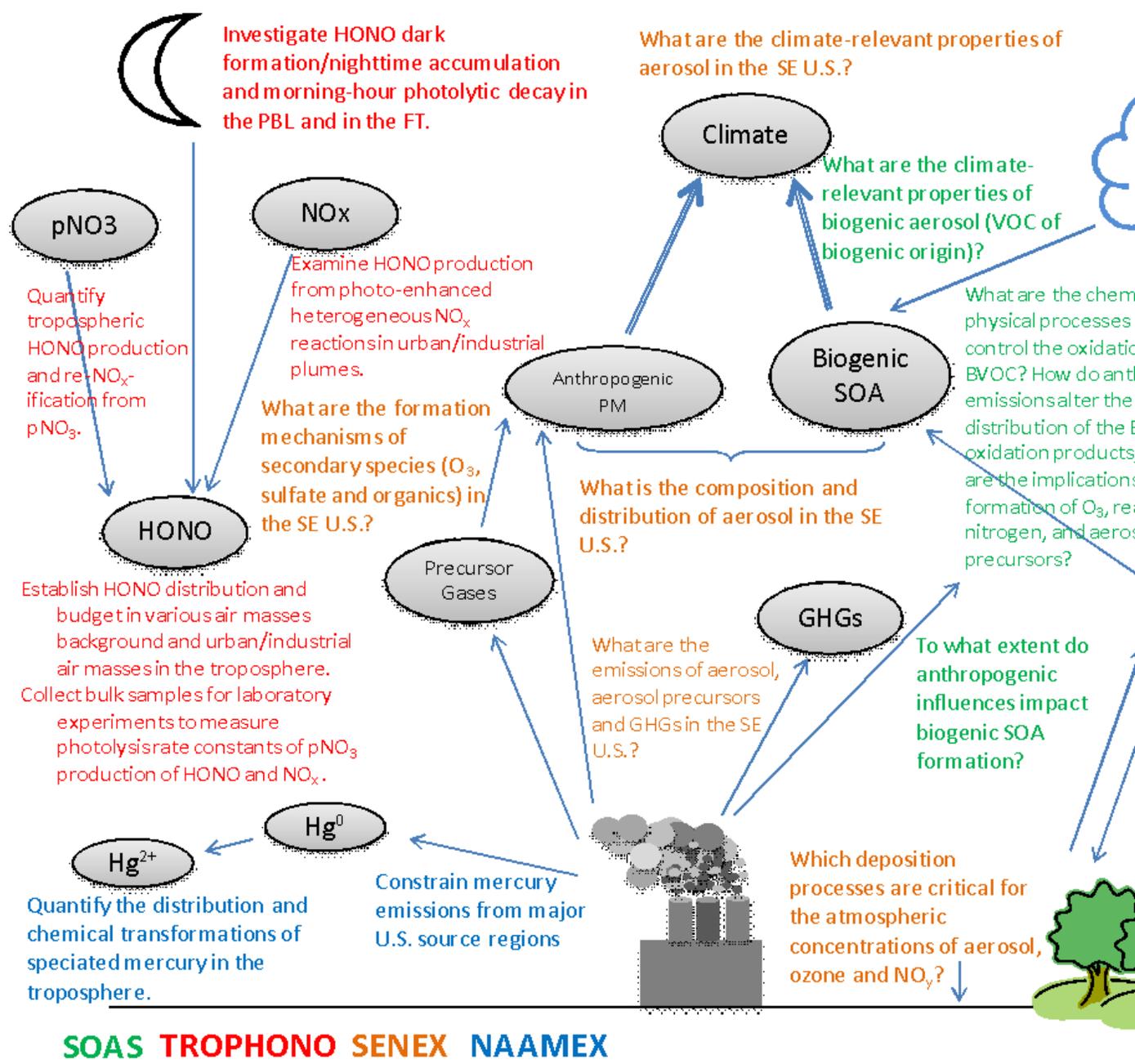
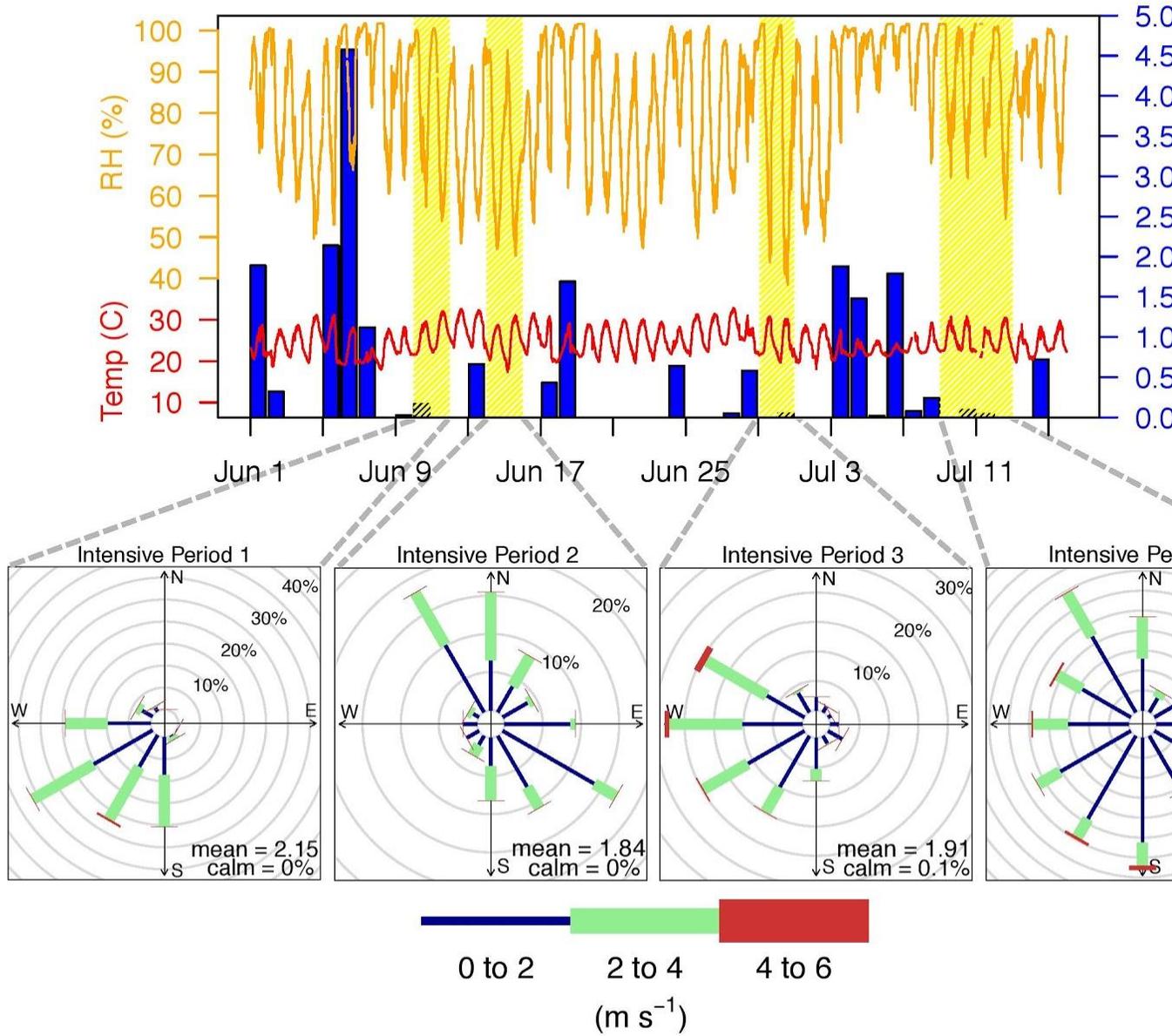


Figure 2. Coordinated SAS science questions

SOAS Weather Conditions – Centreville, AL



Frequency of counts by wind direction (%)

Figure 3. SAS campaign (June 1 – July 15, 2013) meteorology for the main ground site in Centreville, AL. Wind roses correspond to the meteorology during the four intensive sampling periods coordinated between the Centreville, AL and Look Rock, TN sites.

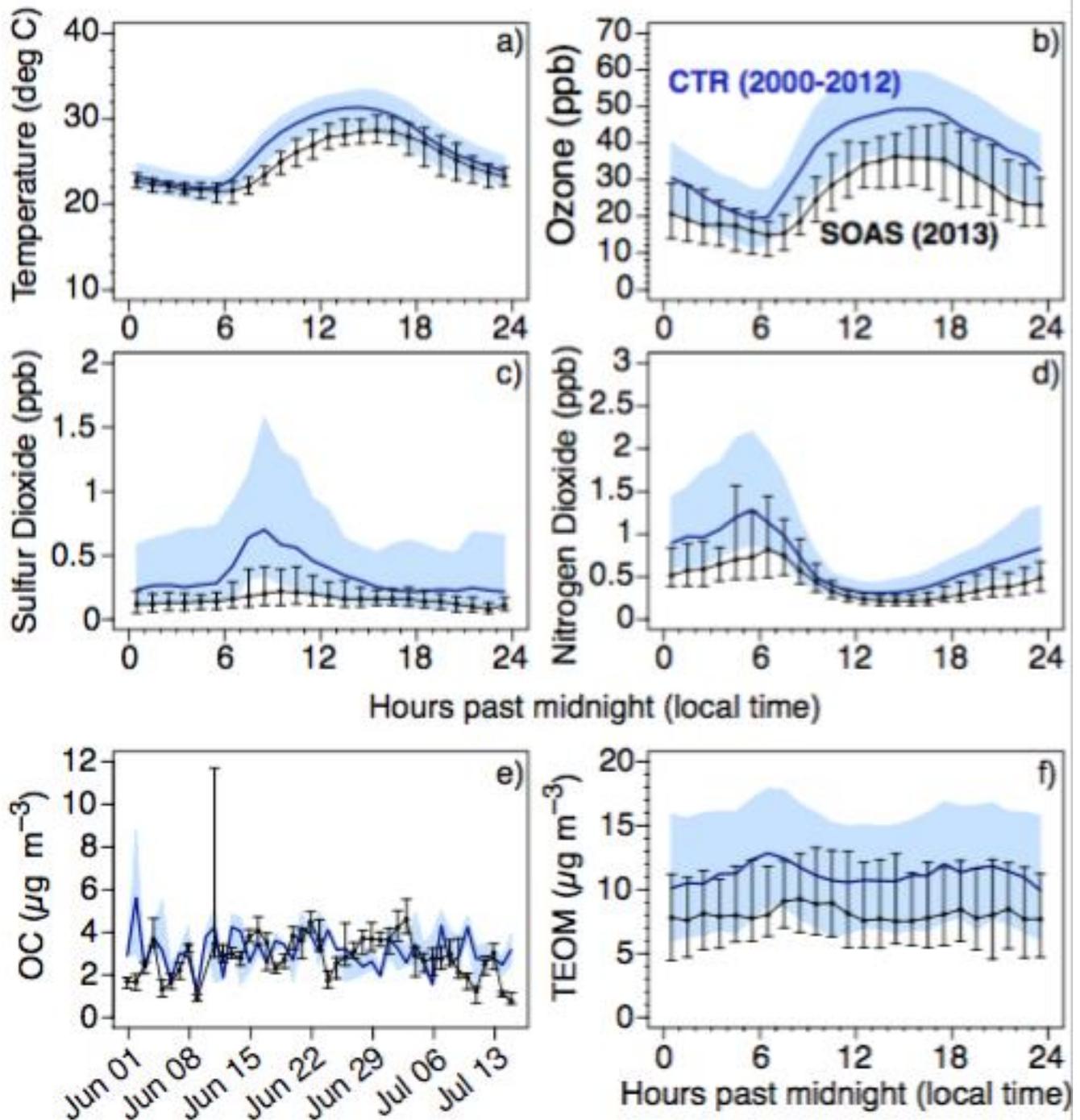
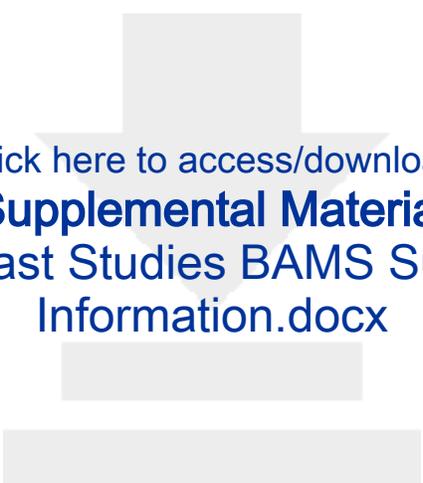


Figure 4. Median diurnal profiles at the Centreville SEARCH site for a) temperature, b) ozone, c) SO_2 , d) NO_2 , e) median 3-day averages in particulate OC and f) median diurnal $\text{PM}_{2.5}$ mass from 2000-2012 (blue: median and 25th-75th distribution). SAS campaign measurements at the main SAS surface site located at the SEARCH “CTR” ground site in black.



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Supplemental Material

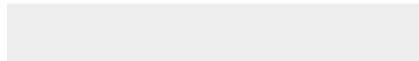
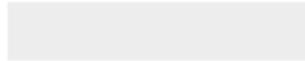
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