WORKSHOP #14

What is BEST for Atmospheric Sciences?

Allison McComiskey (BNL), Juergen Thieme (NSLS-II), Yang Yang (NSLS-II), and Daniel Knopf (SBU)

A principal challenge facing the Atmospheric Sciences is the understanding of fundamental processes on the small scale that manifest in global-scale patterns that drive temperature, water availability, air quality, weather extremes, and climate. This is especially true for aerosol science that underpins some of the greatest uncertainties in mitigating the negative impacts of climate change and air quality. Next-generation instrumentation such as the BEST STXM (Bio Environmental Scanning Transmission X-ray Microscope) would allow for compositional, structural, morphological and interfacial analyses of a high number of aerosols whereby critical elements can be mapped down to trace elemental concentrations levels such that the chemical state of these elements can be determined. These next-level studies are needed to fill gaps in the foundational understanding of aerosols and other atmospheric processes that are required to advance predictive models of the Earth system necessary for planning of a clean air future and informing policy. The lack of a STXM is a significant gap in the portfolio of instrumentation valuable for atmospheric sciences within the U.S. The National Synchrotron Light Source II (NSLS-II) at Brookhaven National Laboratory (BNL) is a state-of-the-art synchrotron radiation facility very well suited for X-ray microscopy and spectroscopy with high spatial resolution and elemental sensitivity. It provides the necessary brightness to meet the needs of a next-generation STXM. BEST would not only fill the gap in instrumentation, but it would advance the micro-spectroscopic sampling technique by increasing analyzed particle numbers via AI assistance. The outcome would be extremely valuable for providing the necessary data for air quality, cloud-resolving, and climate models. The goal of this workshop is to showcase the state of the art in the application of STXM analysis to atmospheric sciences and to discuss the benefits of BEST for this scientific community.

Start Time (ET)	Title	Speaker (Affiliation)
10:00	Welcome and Introduction	Juergen Thieme (BNL)
10:20	Soft X-ray Nanoprobe Beamline at NSLS-II – Design and Status	Wen Hu (BNL)
10:40	Chemical Imaging of Atmospheric Particles: Applications and Challenges	Alexander Laskin (Purdue U)
11:25	Multiscale measurements and modeling to tackle structural uncertainty in aerosol models. Or: What IS the aerosol state in the ambient atmosphere?	Nicole Riemer (U Illinois)
12:00	Lunch Break	
13:00	Paradigm Shifts in Atmospheric Aerosol Chemistry and Physics	Peter Alpert (PSI)
13:30	Integrated Spectroscopic Analyses of Atmospheric Aerosols to Probe Phase State in Extreme Environments	Andrew Ault (U Michigan)
13:50	Unraveling the mysteries of soil carbon using X-ray spectromicroscopy	Marco Keiluweit (U Mass)
14:10	Ambient Marine Aerosol Particles and the Potential for their Characterization by STXM-NEXAFS	Lynn Russell (UCSD)
14:55	BREAK	
15:15	Vertical profile of atmospheric particle composition	Swarup China (PNNL)
15:35	A Detailed View of the Chemical and Morphological Properties of Individual Aerosol Particles	Ryan Moffet (Sonoma Tech)
16:05	Discussion and Farewell	

Abstracts

Soft X-ray Nanoprobe Beamline at NSLS-II – Design and Status

- *Wen Hu (BNL)*

The Soft X-ray Nanoprobe (SXN) beamline is part of the DOE funded NEXT-II project to deliver 3 additional "best-in-class" imaging beamlines to NSLS-II. It will offer researchers state-of-the-art soft x-ray nano-imaging and spectroscopy tools with world-leading coherent high photon flux in the energy range from 250 eV – 2500 eV and full polarization control. It will provide element access from carbon (C) to sulfur (S) through K-edges and may other important elements through L- and M-edges. The primary endstation, nanoISM, will offer both a conventional scanning transmission x-ray microscopy (STXM) mode, for high throughput 2D/3D absorption imaging, and a coherent diffractive imaging (ptychography) mode, for extra high spatial resolution. In this talk, I will give an overview of the design and status of SXN beamline as well as its potential of nanoISM endstation and in future.

Chemical Imaging of Atmospheric Particles: Applications and Challenges

- *Alexander Laskin (Purdue U)*

This presentation will discuss recent advances in STXM chemical imaging of atmospheric particles collected in field and laboratory experiments. The unique advantage of STXM method is that it simultaneously provides two analytical measurements: imaging of particles to assess variability in their individual sizes and morphology, as well as particle specific speciation of carbon bonding and its spatial heterogeneity within individual particles. Obtained data provides unique experimental insights on the nature and sources of particles, understanding their physical properties, atmospheric reactivity, and transformations. We highlight applications of STXM chemical imaging in selected recent studies, discuss existing limitations, and forecast future research directions for this area.

Multiscale measurements and modeling to tackle structural uncertainty in aerosol models. Or: What IS the aerosol state in the ambient atmosphere?

- *Nicole Riemer (U Illinois)*

As the aerosol representation in global models and chemical transport models of different scales has become more sophisticated, the need for data to evaluate these models and to quantify their structural uncertainty has grown. In my presentation I will outline how a multiscale measurement/modeling strategy can help tackle this need. I will highlight the role of single-particle measurements in the hierarchy of measurement techniques and their potential to provide strong constraints on model accuracy.

Paradigm Shifts in Atmospheric Aerosol Chemistry and Physics

- *Peter Alpert (PSI)*

The majority of atmospheric aerosol particles have diameters between about 0.1-2.0 µm and are composed of mainly low Z atoms. It is no wonder then why STXM/NEXAFS has continued to be an unmatched single particle characterization tool in the field of atmospheric aerosol physics and chemistry. This talk will review the significant and recent advancements using STXM/ NEXAFS for atmospheric science investigations. First, in-vacuum studies will be reviewed that characterize particle mixing state and aerosol optical resonant effects relating to absorption enhancement. Then, a focus will be on in situ environmental cells used in studies that control water uptake on aerosol particles, liquid-liquid phase transitions, multiphase chemistry, aerosol photochemistry and ice nucleation. Here, there is a detailed need for increased particle statistics. This leads to a final discussion on open questions and the outlook of STXM/NEXAFS applied to future work in atmospheric chemistry and physics.

Integrated Spectroscopic Analyses of Atmospheric Aerosols to Probe Phase State in Extreme Environments

- *Andrew Ault (U Michigan)*

The phase (liquid, semi-solid, and solid) of atmospheric aerosols plays a key role in their ability to undergo reactions and nucleate cloud droplets or ice crystals. We used a combination of electron microscopy, atomic force microscopy, Raman microspectroscopy, and photothermal infrared spectroscopy (PTIR) to show that particles in the Arctic were unexpectedly solid at relative humidities (RHs) between the deliquescence (80 %) and efflorescence (40 %). Critical to understanding these particles was scanning transmission X-ray microspectroscopy with near edge X-ray absorption fine structure (STXM-NEXAFS) spectroscopy that showed thin organic coatings on the particles. These particles are likely increasing as sea ice decreases and may impact regional climate.

Unraveling the mysteries of soil carbon using X-ray spectromicroscopy

- *Marco Keiluweit (U Mass)*

This presentation will focus on the role X-ray spectromicroscopy can play in disentangling the fundamental mechanisms that underly global feedbacks between soil carbon and the climate system. Soil carbon is a vast and dynamic terrestrial carbon pool. It responds to and drives climate change, so the question is how we can better predict the fate and impact of soil carbon on the environment. To improve predictions, a mechanistic understanding of the controls on carbon storage in soils is critical. X-ray spectromicroscopy using a STXM is the tool of choice to resolve the molecular microbe-mineral-organic matter interactions that control soil carbon storage. Using spatially resolved carbon NEXAFS spectroscopy, we show that plant roots and associated microbes can either stabilize or destabilize carbon attached to minerals, depending on the weathering stage of the respective soil. This consideration is critical for future predictions of the vulnerability of soil carbon to global change. For STXM measurements, the overall goal is to resolve microbe-mineral-organic matter interactions within microhabitats in structurally intact soils. Elements of interest here are carbon and oxygen for organic matter; nitrogen, phosphorus, and sulfur for nutrients; and calcium, iron, manganese, and aluminum for minerals. We will show how high spatial resolution and cryo capabilities would enable more nuanced investigations of microbial biofilms and mineral-organic interfaces in an aqueous environment with a beam damage impact of low to none. The ability for rapid screening of larger numbers of soil particles, very similar to screening processes in aerosol chemistry, would allow for the investigation of larger regions of interest and numbers of soil particles, putting results on a statistically more solid ground.

Ambient Marine Aerosol Particles and the Potential for their Characterization by STXM-NEXAFS

- *Lynn Russell (UCSD)*

The organic carbon fraction of the submicron marine aerosol has been identified as an important uncertainty for determining their contribution as cloud condensation nuclei (CCN). Very few measurements exist of the organic composition of aerosol particles over the world's oceans, even though oceans cover two-thirds of the planet. Airborne and shipboard samples of particles, cloud residuals, ice nucleating particles, seawater, and ocean microlayers have been collected to measure organic composition by scanning transmission X-ray microscope (STXM) with near edge X-ray absorption fine structure (NEXAFS). Organic functional groups from STXM-NEXAFS microscopy indicate that the majority of particles between 0.25 and 1.75 µm diameter with detectable organic components were consistent with marine organic signatures associated with sea spray particles. Specifically, 73 out of the 96 particles analyzed by STXM-NEXAFS over the Southern Ocean were consistent with sea spray organic components with and without a salt core. These particles were characterized by absorption in the alkyl, carbonyl, carboxylic carbonyl, carbonate, and potassium regions. Aerosol particles collected over the North Atlantic also showed a variety of marine particle types, with similarities to aerosol particles from both the Southern Ocean and the Eastern Pacific. However, quantifying the contributions of these different particle types to CCN requires increasing the number of particles analyzed and automating STXM strategies to reduce biases in the types of particles characterized. Strategies that can optimize "searching" for particles of designated sizes and sufficient signal for detection would also provide improved characterization of uncertainties. Objective sampling of increased numbers of particles could provide the basis for distinguishing sources and processes that control the evolution of the particle population, thus extending the scope of STXM-NEXAFS applications.

Vertical profile of atmospheric particle composition

- *Swarup China (PNNL)*

The unmanned aerial systems have gained significant interest in atmospheric science, which could minimize the error, risk and cost in environmental research. I will discuss about vertical profile of aerosol composition using a tethered balloon system deployed at the Atmospheric Radiation Measurement (ARM) sites. Multi-modal micro-spectroscopy of single particle analysis reveals vertical gradient of aerosol composition.

A Detailed View of the Chemical and Morphological Properties of Individual Aerosol Particles

- *Ryan Moffet (Sonoma Technology)*

Aerosols – small particles suspended in the earth's atmosphere – alter climate by directly interacting with solar radiation and by influencing cloud formation. The detailed physicochemical properties of individual particles have a controlling influence on aerosol optical and hygroscopic properties, which, in turn, influence radiative transfer and cloud formation. Synchrotron-based microscopic measurements of individual particles have allowed for the identification of important particle characteristics and processes. For marine sea spray aerosol, ocean biology can control aerosol composition and structure, resulting in particles that having differing cloud formation potential. Findings on the composition and morphology of complex soot particles have implications for the role that these particles play in atmospheric heating. Examples from field studies as far away as the Amazon basin or from as close as Sacramento will be surveyed and presented to illustrate how aerosols interact with the earth system.