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# Workshop to Identify Opportunities in Biological and Environmental Research Uniquely Enabled by the APS Upgrade

Argonne National Laboratory
August 28-29, 2018

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**On the cover:** Center image: Aerial view of the Advanced Photon Source (APS) at Argonne National Laboratory. Insets: Simulated x-ray beam source profiles, comparing the beam from the APS today (left) to the beam from an upgraded APS storage ring (right) with an APS Upgrade multi-bend achromat lattice.

# Workshop to Identify Opportunities in Biological and Environmental Research Uniquely Enabled by the APS Upgrade

August 28-29, 2018

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## Executive Summary

The development of synchrotron-based x-ray science has enabled tremendous scientific advances, driven both by discoveries of new fundamental ways to derive insight from the interaction of x-rays with matter and by the evolution of x-ray light sources themselves. Over the last 20-plus years, the U.S. Department of Energy's (DOE's) Advanced Photon Source (APS) at Argonne has yielded a multitude of transformational insights into science questions across numerous areas relevant to the biological, geological, geochemical, biogeochemical, and environmental sciences.

Recent developments in accelerator technology, particularly the multi-bend achromat lattice, have led to plans to dramatically upgrade the APS, allowing an upgraded APS to offer orders-of-magnitude improvements in key parameters such as coherence and brightness, while retaining high-energy capabilities. These enhanced parameters are directly relevant to almost all hard x-ray imaging, scattering, and spectroscopy experiments (Eriksson *et al.*, 2014). The APS Upgrade Project exceeds the capabilities of today's synchrotrons by 2 to 3 orders of magnitude in brightness and coherent flux in the hard x-ray range, enabling a transformational range of new probes for the structure, properties, and functionality of matter. With vastly improved coherent flux, nearly all techniques can become microscopies. For example, frontiers inaccessible today will be opened by x-ray instruments that provide three-dimensional (3-D) real-space resolution down to nanometers across, up to millimeters of field of view, providing the ability to follow in real time relevant processes and trace element sensitivity down to a few atoms. In addition to the unprecedented spatial resolution across a large field of view, and with appropriate time resolution, the ability for high-throughput, four-dimension/five-dimension data collection enables unprecedented statistical analysis of complex biological and environmental systems in changing environments.

A workshop to identify "Opportunities in Biological and Environmental Research Uniquely Enabled by the APS Upgrade" took place at Argonne on August 28-29, 2018. An emphasis of the workshop was an exploration of how imaging and microscopy experiments enabled by the APS-U will be most relevant for biological and environmental research. One of the goals of the workshop was to identify some of the opportunities for compelling biological and environmental research that will be enabled by an upgraded APS. Participants worked to identify synchrotron-based approaches at the APS and APS-U that would help them address their critical research questions. In addition, participants worked to identify *improved or new* synchrotron approaches provided by the APS-U that could help address their critical research questions. A final goal of the workshop was to articulate the role the upgraded APS can play in biological and environmental research within the context of other DOE user facilities and capabilities throughout the U.S. These include the Environmental Molecular Science Laboratory, the Spallation Neutron Source, numerous electron microscopy centers, other x-ray light sources, and DOE high-performance computation facilities that are already serving the biological and environmental research communities.

A sister workshop on "Biological Science Opportunities Provided by the APS Upgrade" was organized at Argonne to explore the biological science opportunities offered by the unique new capabilities of the APS-U and how the broad structural biology community can best exploit them and a workshop report (https://bit.ly/2xcYUrF) describes the conclusions from that workshop in great detail. The APS-U enables routine serial macromolecular crystallography as well as microbeam experiments ideally suited to study soluble proteins (Meents *et al.*, 2017) and membrane proteins (Martin-Garcia *et al.*, 2017), as well as study of protein dynamics, which today require a free-electron laser (Mizohata *et al.*, 2017). Finally, participants in both workshops recognized that the use of the APS-U to address macromolecular x-ray crystallography science provides part of the continuum of spatial and temporal scales needed to enable investigations of biological and environmental science questions from the sub-

nanometer to centimeter spatial scales and the sub-picosecond and longer temporal scales (see Table 1).

Table 1. Key complex system features, the spatial resolution required to observe them, and their importance to accelerating scientific discovery in the biological and environmental sciences.

| Resolution | Feature/Process | Utility |
|---|---|---|
| 0.1-500 nm | Mineral surface, mineral-microbe interface, mineral-root interface, aerosols, organo-mineral associations, C species, nutrient species, other elements, cellular and subcellular structure (cell walls, internal cell structure), cellular and subcellular biochemistry, pore chemistry, water | Reactive transport, thermodynamic models |
| 0.5-10 μm | Bacterium, hyphae, mycorrhizae, microsites, pore chemistry, biofilms, organo-mineral complexes, pore connections/throats, plant-microbe environments, water | Process-rich models, microbially-explicit models |
| 0.01 mm-2 mm | Colonies, roots, aggregate hierarchy, detritus, redox fronts, pore networks, water | Microbiome investigations, input to ecosystem models |
| 0.2 mm-5 cm | Plant physiology, wetting fronts, root networks, soil column features, redox fronts, water | Generalizable principles, input to local, regional models |

Attendees at this workshop identified many biological and environmental thematic research areas that would benefit from the projected 500-1000-times increase in x-ray brilliance of the APS-U. Attendees also identified specific key research questions in each of those thematic research areas that presently are not easily addressable, but could be addressed by using the high brilliance of the APS-U hard x-ray beams. Integrated and collaborative brainstorming sessions among workshop participants with biological science expertise, environmental science expertise, or x-ray physics expertise enabled identification of the specific roles different x-ray measurements could play in addressing these critical research questions.

Without exception, all workshop participants expressed great confidence and excitement in the role that the APS currently plays in their respective research areas and the role that the APS-U will play. Below is a short summary of the biological and environmental theme areas workshop participants identified as benefitting from the APS-U. More detailed discussions of each of these biological and environmental research areas, key questions that are addressed by the APS-U, and the specific means by which the APS-U will be used to advance the science are presented later in this report.

*Cell Structure*

Defining the relationship between structural organization and emergent properties is fundamental to understanding the function of complex biomolecular systems, critical for modifying or re-engineering these systems, and necessary for designing and synthesizing systems that exhibit novel properties. Lignocellulosic biomass is primed to play a major role in our future bio-economy by providing wood-based building materials and as a renewable resource for biorefineries producing chemicals, fuel, and energy. Challenges include characterization of the multi-scale organization of structural elements in lignocellulosic biomass and linking multi-scale organization to mechanical, chemical transport, thermal, moisture-induced swelling/shrinking, and acoustic properties. Synchrotron x-ray structural characterization techniques (*e.g.*, x-ray diffraction, ptychography, and transmission x-ray computed tomography) used in conjunction with *in situ* moisture chambers are already revealing new information

about the moisture swelling/shrinking in wood-based materials. Similarly, in biorefineries, the production of fuels and chemicals depends on diffusion of deconstruction chemicals into cell walls, in addition to transport of depolymerization products out of cell walls. Along with ptychographic and transmission tomography imaging, scanning x-ray microdiffraction is a promising new approach that has the potential to provide detailed information about the molecular-to-nanoscale architecture of these tissues.

*Cell Chemistry*

Although the first-row transition metals of the Periodic Table of Elements are critical in terms of enabling the chemistry of life, they are minor in terms of biomass. Understanding homeostatic control of trace-metal assimilation, utilization, and sequestration are required for new discoveries with respect to fundamental pathways of metal metabolism and are broadly applicable in biology. For example, the metalloproteins associated with microorganisms play an important role in the oxidation of methane gas, which is an important greenhouse gas with a 20-year global warming potential 84 times that of $CO_2$. Beyond microorganisms, the cellular chemistry of higher organisms, such as plants, is critical for understanding controls on biomass production for agricultural, chemical, and fuel production. Scanning-probe x-ray fluorescence microscopy and ptychography approaches in 3-D, coupled with x-ray absorption spectroscopy approaches with nanometer spatial resolution to understand the spatial organization of chemistries within cells, are critical for advancing our understanding of cell function. In addition, the higher throughput of these measurements enabled by the increased brilliance of the APS-U enables a statistical analysis of the cell-to-cell variations of chemistries within them. Finally, the visualization of larger fields of view enabled by the increased brilliance of the APS-U provides high spatial resolution (10 nm) investigation of the organizational structure within organelles without the need for sectioning the organelles typically required by other imaging modalities.

*Environmental Microbiology and Biogeochemistry*

Microorganisms inhabit nearly every environment on Earth, living in complex communities forming a network of integrated metabolisms that drive the biogeochemical cycles of major (hydrogen, carbon, nitrogen, and oxygen) and many minor elements (e.g., phosphorous, sulphur, manganese, and iron). These coupled processes play a critical role in major ecosystem processes including the assimilation of carbon, the mineralization of organic matter and accompanying release of $CO_2$, the formation and oxidation of $CH_4$, uptake and release of nutrients (*e.g.*, nitrogen and phosphorous), and the availability and transformations of contaminants. Development of a robust and predictive understanding of coupled biogeochemical processes and cycles requires a comprehensive knowledge of the chemical speciation and spatial organization of the elements of interest in a system and the processes controlling mass transfer. *Chemical speciation* information is critical for defining how biological behavior, abiotic-biotic interactions, and molecular transformation control the reactivity and mobility of contaminants (*e.g.*, uranium, technetium, and mercury), nutrients (*e.g.*, nitrogen, phosphorous, and carbon), and critical biogeochemical elements (*e.g.*, sulphur, iron, and manganese). The *spatial organization* and structure of heterogeneous microenvironments can greatly impact the biogeochemical properties within air, aquatic, and terrestrial systems. Given the ubiquity of these complex biotic and abiotic micro-environments, techniques are needed that can characterize the chemistry within them. By providing an orders-of-magnitude increase in coherence and brilliance, the APS-U provides the ability to image these microenvironments with larger fields of view, in 3-D, and with the higher spatial resolution than is possible today. In addition, the APS-U enables a statistical analysis of the variations of micro-environment function within critical environmental systems.

*Soil Structure and Chemistry*

Soil is a remarkably complex biomaterial that exists as a dynamic, structured, self-organized but highly heterogeneous mixture of mineral particles, organic matter, water, air, and countless living organisms. Soil organic matter exists as a continuum of decay products of varying size, composition, and position

within the soil matrix. The spatial organization of these components (i.e., soil structure) is dynamic and linked to environmental processes such as wetting/drying, soil mineralogy and chemical interactions, organic matter decomposition, and the biophysical actions of living plant roots and soil biota. The controls on soil hydrobiogeochemical cycles and their intimate coupling with plant productivity are at the core of Earth's life-support systems

Currently, micron- and pore-scale soil environmental properties affecting the stabilization, priming, and mineralization of soil organic matter are not explicitly considered in Earth system models. A critical but unresolved question is to what extent these small-scale phenomena must be considered to improve the predictive power of Earth system models as climate change occurs. For instance, relatively small changes in soil organic matter dynamics and storage could have a large impact on the global carbon cycle, and subsequent cycling of water and nutrients. Some of the critical questions in soil science that need to be addressed include: What is the spatial/temporal heterogeneity of soil organic matter composition and its relation to the soil mineral matrix and structure? What physical, chemical, biological, and environmental factors control the transformation of carbon and nutrients and water movement within soil?

Spatially resolved 3-D chemical and structural information of soil would be extraordinarily valuable for understanding nutrient cycling in soil, maintenance of soil fertility and water retention within soils, carbon storage in soils, and the dynamic co-control of biogeochemistry by microbial catalysis and diffusive/advective delivery of substrates within soils.

The orders-of-magnitude increase in coherence and brilliance provided by the APS-U provides greater fields of view (millimeter to centimeter) with increased spatial resolution (~10 nm) for 3-D imaging of the structure and chemical properties of soils. The increased brilliance of the APS-U also enables the development of x-ray Raman measurement approaches that can be useful for measuring carbon chemistries within bulk and hydrated soils. Combined use of all of these developments can provide critical information about 3-D pore size/shape-connectivity/soil organic matter-inclusions within soil, 3-D water flow paths and water location with soil, and 3-D organic carbon and microbial distribution within soils, particularly associated with plant roots and minerals.

*Atmospheric and Aerosol Research*

Atmospheric aerosol particles have significant impacts on air quality and visibility, public health, biogeochemistry, and climate change. A key challenge in understanding the uncertainties related to increasing greenhouse gases and aerosols is closely tied to their interactions with clouds. Presently, investigation of the nanometer-scale structure and chemical interactions of aerosols and their interactions with clouds is very difficult. In addition, statistical classification of aerosol samples is hampered by the reduced number of samples that can be analyzed. The development of a cloud chamber at the upgraded APS-U for imaging aerosols can enable the investigation of aerosol-cloud interactions at the nanoscale. Workshop participants recommended the exploration of the feasibility to develop a cloud chamber at the APS-U to investigate the structure and chemistries associated with aerosol-cloud droplet interactions.

*Integrated Use of Facilities*

Workshop attendees also discussed the opportunities that could be created by integrating the capabilities provided by other DOE user facilities and infrastructure with the strength of the APS-U into a holistic approach to biological and environmental science. Particular excitement was generated by the idea of integrating use of x-ray probes (provided by the APS-U) with neutron probes, electron probes, and other electromagnetic wavelengths to optimize the different types of contrast mechanisms and spatial and temporal resolutions for their research. The need for high-performance computing

capabilities to handle the transfer and analysis of the large datasets that will result from this future research was also recognized.

*Additional Needs for Maximizing the Value of the APS-U in Addressing Biological and Environmental Science*

Workshop participants felt strongly that the APS-U provides opportunities in creating many new types of 3-D x-ray microscopes, which will depend upon different types of interactions of x-rays with matter (absorption and scattering). To realize the scientific impact of the APS-U, workshop participants recommended as needs in several key areas:

1) availability of biological and environmental science lab infrastructure close to the APS-U for developing, characterizing, and/or manipulating time-sensitive samples;
2) development of standardized sample preparation protocols for the many different types of biological and environmental samples that might be measured at the APS-U;
3) development of standardized cryogenic sample preparation and cryogenic specimen imaging methods to mitigate radiation damage resulting from the brighter x-ray beams of the APS-U;
4) development of sample holders and manipulators so that a sample can be easily transferred among different synchrotron beamlines as well as multiple user facilities for multiple types of measurements;
5) "in-house" researchers at the APS-U (similar to the organizational model of the Environmental Molecular Sciences Laboratory) who understand the broad areas of biological and environmental science as well as the x-ray physics at the foundation of the synchrotron measurements and who facilitate access to the facility by the biological and environmental science researcher;
6) development of data analysis tools to help the biological and environmental scientist with x-ray data analysis; and
7) advancement of the combined use of electron, neutron, and visible, infrared, and x-ray wavelength imaging approaches, including correlative image analysis.

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## The Advanced Photon Source and the Advanced Photon Source Upgrade

X-ray imaging and microscopy has had tremendous impact on biological and environmental science questions over the past decade, addressing extremely broad and highly relevant scientific and industrial questions. It is particularly well suited to visualizing systems and materials across numerous length scales, in depth, and *in situ* or *operando*. Application areas are as broad as dynamic imaging of crack propagation in materials including rocks, mapping trace elements in biological and environmental systems to better understand their impact both in their natural occurrence as well as in man-made situations, and nanoscale imaging of melt formation. Hard x-rays are also extremely well suited to imaging systems where both hard and soft materials are combined (*e.g.*, microbes in soil or the rhizosphere), and where high spatial resolution is needed in the context of a “thick” sample.

The Advanced Photon Source Upgrade (APS-U) creates a synchrotron x-ray light source optimized to produce hard x-rays with a very high degree of spatial coherence. Components of the APS-U are being designed and fabricated, and first x-rays are scheduled to be delivered in the middle of calendar year 2023. The upgraded APS source exceeds the capabilities of today’s synchrotrons by 2-to-3 orders of magnitude in brightness and coherent flux in the hard x-ray range, enabling a transformational range of new probes of the structure, properties, and functionality of matter. A greatly increased flux of spatially coherent x-rays provides an unprecedented level of understanding of nanometer-scale heterogeneity and fluctuation dynamics in matter ranging from “soft” biomolecules to “hard” environmental materials (*e.g.*, minerals). The increase in brightness and coherence particularly benefits imaging techniques such as intensely focused x-ray beams for dramatically advanced nanoscale imaging, nanospectroscopies, and nanodiffraction; coherent diffractive imaging and ptychography reaching spatial resolutions approaching atomic length scales and permitting time-resolved studies; and projection microscopies that bridge the highest spatial resolution with high time resolution.

The importance of a multiscale understanding of systems has for a long time been recognized by biological and environmental scientists. Traditionally, studies across length scales were achieved by collaboration, employing different instruments and contrast mechanisms. The multi-bend achromat upgrade of the APS source enables, for the first time, effective visualization across all relevant length scales — atomic, mesoscale, and macroscopic — using a single experimental technique. X-ray wavelengths are ideally matched to atomic distances in solids, and structure determination can be “simply” a matter of placing a wide- or small-angle detector near the sample in the beam. The crucial enabling feature of a nanoprobe beamline on the upgraded APS is a mesoscale beam with high brightness and coherence. The focused coherent beam (*e.g.*, 20 nm in size) enables direct techniques (*e.g.*, x-ray fluorescence) to image individual mesoscale features that are broadly averaged by current techniques, while the high x-ray brightness enables ptychographic imaging in order to resolve features on the scale of 5 nm or below using hard x-rays. Finally, the improved beam combined with end-station upgrades supports sample scan rates up to 1000 times faster than currently possible, with unprecedented sensitivity. This allows for the rapid acquisition of statistics on a length scale of hundreds of micrometers and above. In this way, x-ray scanning probe techniques at the APS-U bridge the atomic, mesoscale, and macroscopic lengths scales in a single experiment, something that has not been possible in the past.

In this context, chief among the APS-U capabilities is nanoscale spatial resolution at high energies, which enables probes of structure, chemistry, and processes deep within heterogeneous systems. This enhanced spatial resolution brings a new capability: mapping trace metal distributions in “soft” materials down to length scales of ~10 nm. For radiation hard samples, the achievable spatial resolution can be pushed further, to 5 nm and below, with trace-metal sensitivity. The APS-U high brightness also significantly increases the three-dimensional (3-D) field of view of scanning microscopes (see Fig. 1), since source brightness determines the time needed to probe a volume of interest at the relevant spatial resolution. Crucially, this combination directly leverages the study of rare events, defects, and trace content in biological and environmental systems by increasing the sensitivity of the technique as well as by dramatically increasing the speed at which data can be acquired. This increased speed can be used to significantly improve the number of samples and number of different sample conditions that can be studied (and thus dramatically increases the statistical significance of findings), and makes it possible to follow the behavior of dynamic systems with trace-elemental sensitivity.

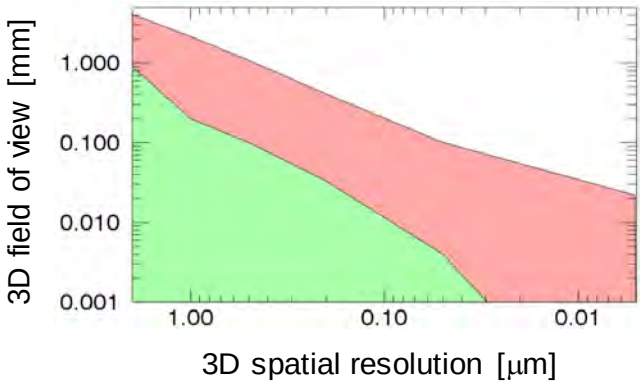


Fig. 1. Achievable 3-D spatial resolution (with trace element sensitivity) as a function of object size for the current APS (green parameter space) as well as the upgraded APS (red background). For example, the APS-U enables trace metal mapping down to 10-nm 3-D resolution for a ~100-μm$^3$-sized sample, whereas today only ~ 500-nm 3-D resolution can be achieved.

Micro- and nano-beams are also extensively utilized for applications of spatially-resolved x-ray spectroscopies, notably x-ray absorption near edge structure (XANES) and extended x-ray absorption fine structure spectroscopies. Numerous high-impact applications of these techniques cover a wide range of scientific disciplines. In the biological and environmental sciences, spatially-resolved XANES can be used to determine valence states for multivalent elements for determining the redox reactivity and transformation of iron-containing minerals and predict the mobilities of contaminants (*e.g.*, uranium, mercury, chromium) including speciation transformations instigated by bacterial cells and inorganic agents. Chemical states of trace metals in biological systems can also be interrogated to better understand redox reactions and biological transformations; spatially-resolved metal speciation determinations can provide the molecular basis for understanding a wide variety of biological and biochemical processes.

In addition to improving traditional nano- and micro-probe methods, the significantly increased coherence of the upgraded APS at hard x-ray energies makes it possible to employ lensless imaging approaches to visualize 3-D structure well beyond the resolution limitations imposed by traditional x-ray optics. Lensless imaging approaches such as ptychography have demonstrated the capability to resolve structures less than 10 nm in size and have already been used to visualize cryogenically preserved soft materials with better than 20-nm spatial resolution with hard x-rays. These techniques are limited by the available coherent flux and promise to be revolutionized by upgraded light sources such as the APS-U. In particular, the spatial resolution achievable by these approaches, with appropriate reconstruction techniques, is in principle only limited by the radiation hardness of the

sample and the available x-ray flux. The upgraded APS opens the possibility to eventually image samples as large as 1 $mm^3$ at a 3-D resolution of 10 nm, thereby carrying out studies that truly bridge numerous length scales.

Even comparatively conventional x-ray imaging techniques benefit tremendously from the upgraded APS. The increased coherence (in particular in the vertical direction) translates into significantly increased phase contrast in direct imaging, i.e., the improved ability to detect and resolve soft materials within hard/dense matrices. The high brightness also uniquely enables x-ray projection microscopies that allow a dynamic tradeoff between spatial resolution, field of view, and speed of data acquisition, opening new imaging regimes that are currently not accessible (see Fig. 2).

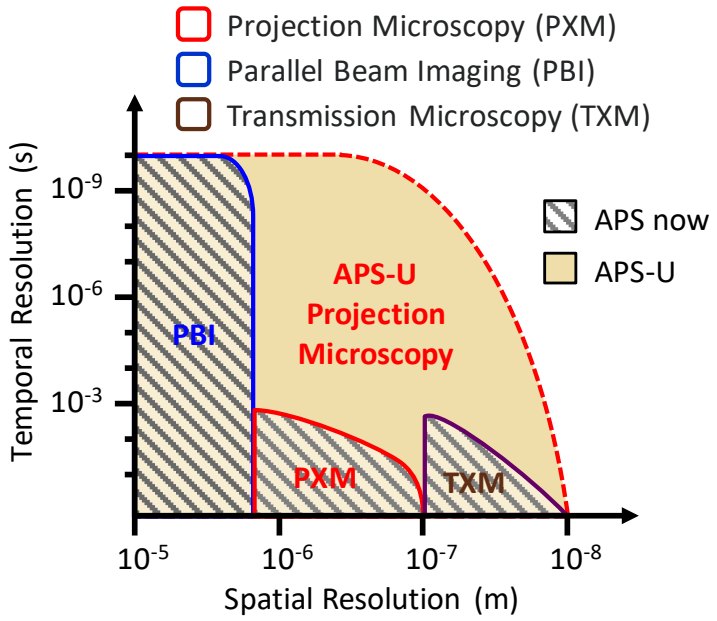


Fig. 2. The coupling between temporal resolution and achievable spatial resolution for hard x-ray imaging. Projection microscopy enabled by the APS-U extends the achievable parameter space for fast x-ray imaging at high spatial resolution. For example, imaging below the 1-µm level for most practical systems today is limited to approximately millisecond timeframes, whereas the APS-U will enable imaging of these systems down to microsecond-to-nanosecond levels.

## Grand Challenges in the Biological and Environmental Sciences

### Cell Structure

Many biological systems are *prima facie* proof-of-principle that hierarchical materials can exhibit properties unachievable in conventional materials. Complex materials — both naturally occurring and synthetic — that exhibit organization of structural elements on multiple length scales often display emergent properties with significant technological potential. Defining the relationship between structural organization and emergent properties is fundamental to our understanding of the function of complex biomolecular systems, critical for modifying or re-engineering these systems, and necessary for designing and synthesizing systems that exhibit novel properties. Comprehensive characterization of these systems represents a substantial technological challenge. For instance, lignocellulosic biomass represents one functionally and economically important example of a hierarchical material. Understanding its molecular architecture at the cellular to sub-cellular scales is a critical step in designing strategies for processing it for diverse economic and industrial applications.

Lignocellulosic biomass is primed to play a major role in our future bio-economy by providing wood-based building materials, in addition to being a renewable resource for bio-refineries producing chemicals, fuel, and energy. Examples of lignocellulosic biomass include wood, grasses, corn stover, bamboo, and other bioenergy crops. Wood and bamboo construction materials possess qualities that make them the choice for innumerable applications: they are readily available, renewable, and inexpensive, and possess exceptional strength-to-weight ratios even compared to engineered composites. They can be easily formed and shaped, and can afford unique aesthetic appeal. The lignocellulosic fraction of biomass also has great potential for use as a renewable feedstock to yield biofuels and biomaterials. Achieving this prospect can offset the diminishing availability of fossil fuels, and meet increasing consumer demand for green chemicals.

If properly managed, lignocellulosic biomass resources can be used in sustainable, environmentally friendly ways, and wood itself can be the ultimate in green building material. However, the development of technologies to fully utilize lignocellulosic biomass resources is hindered by the lack of fundamental material structure-property-processing-performance relationships, especially at the small length scales such as individual cell wall layers.

Lignocellulosic biomass like wood is generally a cellular material consisting of cells that can be thought of as hollow tubes with polymeric multilayer walls. The three main polymers in wood are cellulose, hemicelluloses, and lignin. Cellulose is the most abundant biopolymer on Earth. It is a linear polysaccharide composed of repeating cellobiose units that coalesce through inter- and intra-molecular hydrogen bonding to form strong cellulose fibrils. Secondary cell wall layers can be thought of as nanofiber reinforced composites with cellulose fibrils embedded in a highly organized matrix of hemicelluloses and lignin. Individual cells are held together by the compound middle lamella, which is made of an open-cellular polysaccharide structure encrusted with lignin. Although many details have been proposed, the structure of the cell walls from the molecular scale up to about 100 nm remains unknown and thus an active area of research.

Some of the challenges related to cell structure that the APS-U is well-positioned to address include 1) characterization of the multiscale organization of structural elements in lignocellulosic biomass; 2) linking multiscale organization to mechanical, chemical transport, thermal, moisture induced swelling/shrinking, and acoustic properties; and 3) design of biomass predicted to have emergent properties of technological value. To make progress on these issues, fundamental questions about biomass structure and behavior must be addressed. These questions include: What is the nanostructure of secondary lignocellulosic cell walls? What are the mechanisms, rates, and moisture

effects on material transport through lignocellulosic biomass? What are the deformation mechanisms across length scales under mechanical loading or changes in moisture content?

The APS-U provides significant capabilities for addressing the characterization of these classes of materials. For instance, detailed structural studies are required to build and validate models for the interactions between liquids — such as water — and the biomass. Molecules of water are absorbed by all of the wood polymers, except between the highly ordered cellulose chains in the cellulose elementary fibrils. The wood moisture content (MC), defined as the mass of water in wood divided by the oven-dry mass of wood, depends on the ambient temperature and relative humidity. At low MC, all of the water in wood is bound by the wood polymers inside cell walls. As MC increases, the maximum capacity for bound water is reached at the fiber saturation point, which — depending on definition — is between 30% and 40% MC. Additional water added to wood above the fiber saturation point forms free water in empty spaces of the cellular structure. Exposure of wood-based engineered materials to different moisture conditions can lead to excessive dimensional changes that cause cracks (also known as “checks”) or failures in wood-adhesive bond lines. Synchrotron x-ray structural characterization techniques (*e.g.*, x-ray diffraction, ptychography, and x-ray computed tomography) used in conjunction with *in situ* moisture chambers are already yielding new information about the moisture swelling/shrinking in wood-based materials (see Fig. 3 and Fig. 4). (Jakes *et al.*, 2019) However, the measurements are typically “static” and two-dimensional. The APS-U allows researchers to better utilize these techniques to observe 3-D structural changes and capture the faster dynamic processes.

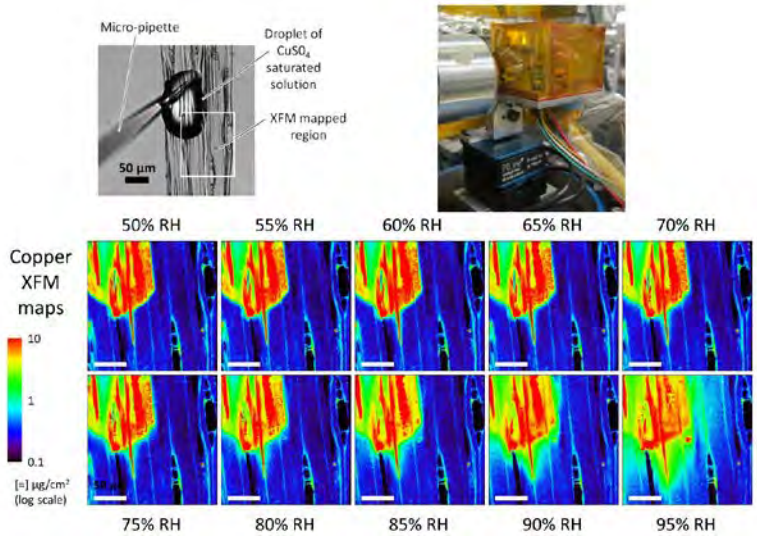


Fig. 3. Upper left: Implantation of copper ions into a 2-µm-thick longitudinal-tangential wood section of latewood loblolly pine. Upper right: Custom-built, relative humidity (RH) chamber for APS beamline 2-ID-E. Lower: A series of XRF microscopy copper maps conditioned at increasing RH. The maps revealed that ion diffusion only occurs above a threshold RH. The maps were collected by raster scanning a 0.5-µm-diameter x-ray probe with 0.3-µm step sizes and 5-ms dwell times. Each map took about 30 min to collect. With the APS-U much larger maps can be collected faster.

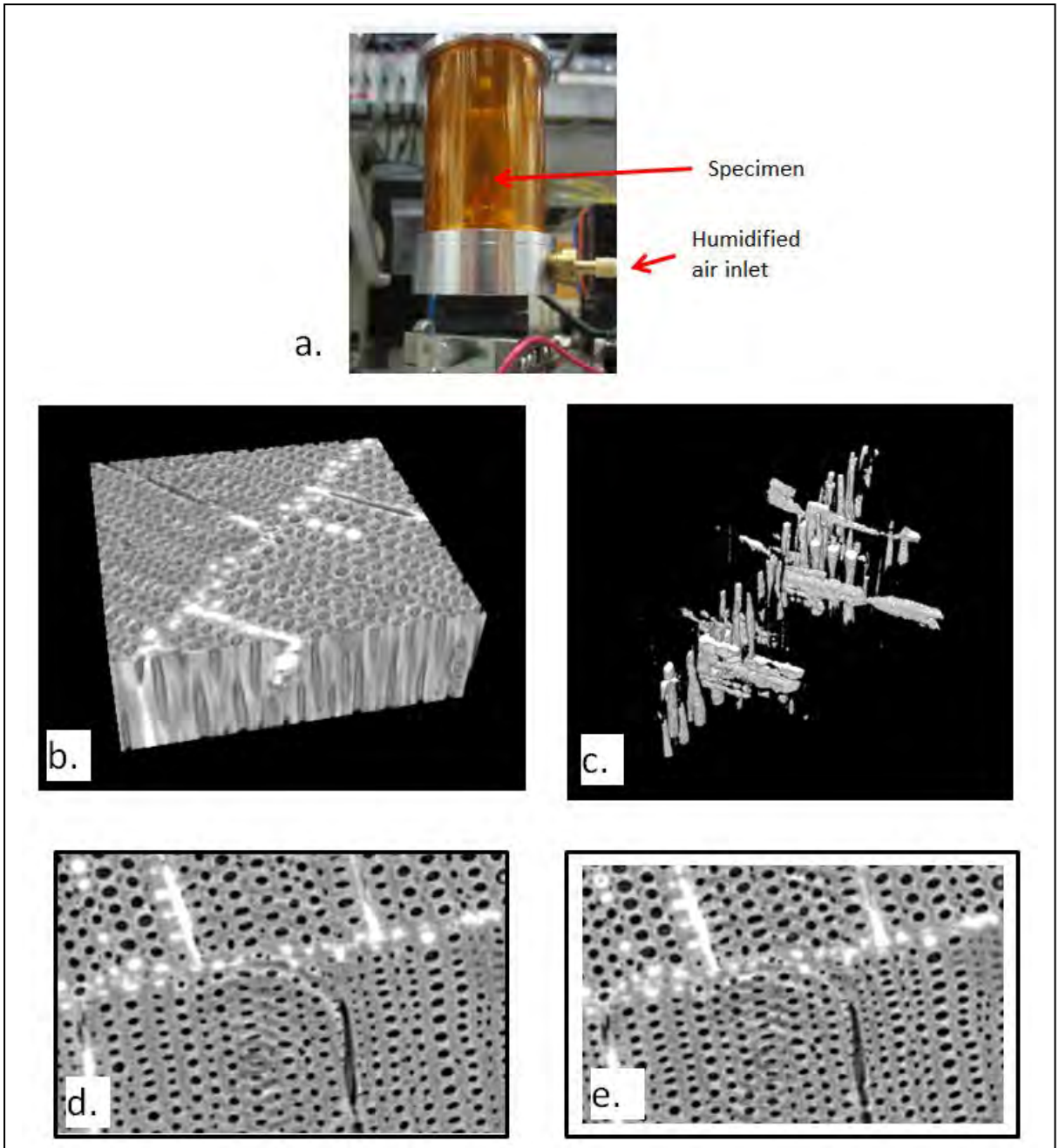


Fig. 4. (a) An *in situ* humidity chamber built at the USDA Forest Products Laboratory for APS beamline 2-BM-B in order to facilitate the study of moisture-induced swelling of wood structure. (b) Three-dimensional reconstruction of wood adhesive bond line and (c) adhesive penetration into wood of the same bond line shown in (b) with the wood cell walls computationally removed. (d) Tomographic slice of wood adhesive bond line conditioned at 100% RH, and (e) 0% RH. Roughly the same cells are shown in (d) and (e) and the black box in (e) shows how much the wood structure shrank during drying.

Analogous to multi-scale synchrotron x-ray structure studies of moisture-induced swelling/shrinking in lignocellulosic biomass, experiments studying mechanical deformations across length scales can be performed by incorporating *in situ* mechanical testing devices. The APS-U enables more 3-D and dynamic observations. Moisture control also facilitates studying the effects of MC on mechanical deformation mechanisms.

Diffusion through cell walls is critical to many of the technologies employed in the utilization of lignocellulosic biomass. In bio-refineries, the production of fuels and chemicals depends on diffusion of deconstruction chemicals into cell walls, in addition to transport of depolymerization products out of cell walls. Cell wall diffusion is also important in the manufacture of wood-based building materials, including treatments of wood with copper-based, waterborne wood preservatives; adhesive bonding of wood; and chemical modifications of wood. Additionally, wood-based materials are susceptible to damage mechanisms such as fungal decay and fastener corrosion, which also require diffusion through the cell wall. Despite the importance of diffusion, the mechanisms and rates remain poorly understood.

Employing current APS capabilities, experiments have been performed to measure ion diffusion constants through lignocellulosic cell walls at lower moisture contents (Zelinka *et al.*, 2015; Hunt *et al.*, 2018). However, at higher moisture contents, diffusion is too fast to capture. With the APS-U it is possible to do the experiments at higher moisture contents and even under water-saturated conditions to mimic industrial processes in bio-refineries. It is also possible to increase replicates and types of diffusive species. Additionally, it is possible to make diffusion observations in three dimensions.

Another example of an emerging characterization tool that will be greatly enhanced by the APS-U is scanning x-ray microdiffraction (see Fig. 5), a promising new approach that has the potential to provide detailed information about the molecular-to-nanoscale architecture of these tissues (Fratzl *et al.,* 1997; Liu *et al.*, 2013; 2016). An x-ray microbeam (1-5-μm in diameter or smaller) is used to collect thousands of diffraction patterns on a grid of comparable step size. Analysis of individual patterns is carried out to extract detailed information about the underlying molecular structures. The results of analyses of thousands of diffraction patterns make possible mapping of the variation in structural properties over the field of view scanned, providing information on structural organization over sub-cellular to cellular and even tissue-scale regions. Three-dimensional information about the variation of structural properties can be extracted via stacking of two-dimensional analyses of serial sections or by tomographic strategies (Liebi *et al.*, 2015; Schaff *et al.*, 2015).

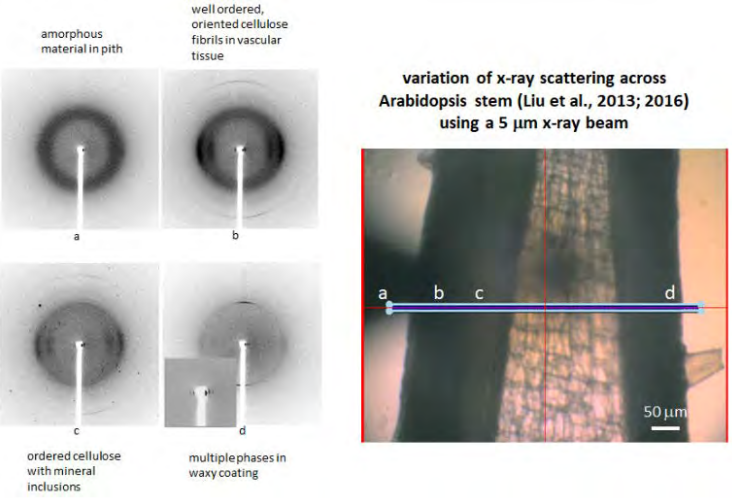


Fig. 5. Scanning microdiffraction of an *Arabidopsis* stem. A 5-μm x-ray beam was used to collect fiber diffraction patterns as a function of time across an *Arabidopsis* stem. A total of 480 patterns were collected in 5 scans of 160 patterns each across the stem. Four patterns are shown here, representing scattering in the stem.

Although plant cell walls are composites of high molecular weight polysaccharides, highly glycosylated proteins, and lignins, the cellulosic constituents exhibit readily identifiable x-ray scattering, making it possible to separate their scattering from that of other constituents to allow mapping of their distribution,

orientation, and order within plant tissue (Liu *et al.*, 2013; 2016a). Cellulose fibrils usually constitute the bulk of the cell wall polysaccharide and their organization is a critical determinant of the mechanical properties of plant tissue. Consequently, detailed information on their spatial organization is critical to understanding the origins of the mechanical properties of plants. However, the organization of cellulose fibrils may vary across sub-cellular length scales, precluding study using conventional x-ray beams. Recent use of microbeams has opened the possibility of investigating the cellulose organization at micron-level resolutions to reveal patterns of structural variations within the tissues (Fratzl *et al.*, 1997; Riekel *et al.*, 1997; Riekel *et al.*, 2001; Lichtenegger *et al.*, 1999; Liu *et al.*, 2013; 2016a). The impact of directed changes in lignin biosynthesis on cellulose organization has been analyzed (Liu *et al.*, 2016a). Similar strategies have been used to study cellular-scale variation in molecular organization in nerve myelin (Inouye *et al.*, 2014), cornea (Daxer and Fratzl, 1997), amyloid deposits in the brain tissue of Alzheimer's patients (Liu *et al.*, 2016b), collagen organization in bone (Liebi *et al.*, 2015; Schaff *et al.*, 2015), and others (e.g., Lutz-Bueno *et al.*, 2016).

With current APS capabilities, the speed of data collection and processing are rate limiting, particularly when 3-D information is required, as it often is. It is currently difficult to scan fields of view larger than a few hundred microns with a 5-µm beam, whereas important functional information may require the use of micron to sub-micron beam sizes scanned over millimeter-scale regions of interest. With current APS beamline capabilities, utilization of micron to sub-micron beam sizes results in significant loss of flux, further slowing data collection. The APS-U generates 2 orders of magnitude greater flux in a micro-to-nano-sized beam than does the APS, thereby enabling scans of much larger fields of view in shorter lengths of time with far higher spatial resolution than are currently achievable. This allows investigators to generate statistically significant data sets with enough experimental replicates to validate observed differences, as well as to explore larger parameter spaces.

Environmental chambers to control humidity, temperature, pressure, and solvent environments are needed to fully utilize APS-U capabilities, as are strategies to mitigate, or at least identify, radiation damage. For instance, examination of materials under cryo conditions is predicted to greatly lower the amount of damage expected from x-ray exposure. The result is a new window into molecular-to-nano-to-macroscale structural organization of biological tissues and a far more complex experimental design aimed at establishing causal relationships between hierarchical structure and macroscale functional properties including mechanical, thermal, electrical, optical, diffusive, and acoustic.

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## Cell Chemistry

Although they are minor in terms of biomass, the first-row transition metals are critical for enabling the chemistry of life. These metals catalyze reactions that are not readily enabled by the functional groups found on amino acid side chains or even the specialized functional groups found in organic cofactors. Zinc, for instance, serves as an electrophile in many reactions, especially to activate water for chemistry; and copper, iron, and manganese enable oxidation-reduction reactions that are critical for extracting energy from oxidation of inorganic compounds for autotrophic growth or organic compounds for heterotrophic growth.

Studies from Merchant and coworkers (Blabby-Haas *et al.*, 2017; Kropat *et al.*, 2015) have established the experimental organism *Chlamydomonas reinhardtii* as a powerful reference system for understanding homeostatic control of trace metal assimilation, utilization, and sequestration. *Chlamydomonas* is in the green plant lineage, but also retains functions that were present in the common ancestor to both plants and animals (Merchant *et al.*, 2007). Their discoveries with respect to fundamental pathways of metal metabolism are therefore broadly applicable in biology. For study, there are several advantages offered by *Chlamydomonas*: 1) as a microbe, it is easy to manipulate nutrient supply; and 2) it can be grown either phototrophically or heterotrophically, thus allowing focus on either photosynthesis or respiration in the experimental design.

Another area in which metalloproteins associated with microorganisms play an important role is in methane oxidation. Methane is an important greenhouse gas with a 20-year global warming potential 84 times that of $CO_2$ (Myhre *et al.*, 2013). Sixty percent of methane produced each year is subsequently consumed by microorganisms, preventing teragrams of methane from reaching Earth's atmosphere (Knittel *et al.*, 2009). Anaerobic oxidation of methane is a globally important methane-consuming process that dominates in anoxic marine sediments in which methane oxidation is coupled to sulfate reduction by a syntrophic association of anaerobic methanotrophic archaea and sulfate-reducing bacteria. Both methane oxidation and sulfate reduction require metalloproteins for catalysis and for electron transfer, with cobalt, nickel, and iron-centered cofactors.

Beyond microorganisms, the cellular chemistry of higher organisms — such as plants — can be studied with the APS-U. The bodies of all multicellular organisms originate from undifferentiated stem cells, yet many steps in the journey from pluripotency to defined cell-fate remain unknown. Post-embryonic development mediated by maintenance of stem cell populations, a feature unique to plants, is ideal for studying the molecular regulation of cell identity. One such stem cell niche is located at the tip of the growing root and gives rise to all layers of mature tissue above it. Plant cells are immobile and thus facilitate easy visualization of cell lineages and developmental stages. Asymmetric divisions of the stem cells that generate the endodermis and adjacent cortex cell lineages are an ideal model to understand stem cell function. Key players controlling these divisions include the transcription factors SHORTROOT and SCARECROW, which regulate radial patterning and stem cell maintenance via positive feedback and feedforward loops (Drapek *et al.*, 2017). The Casparian strip, which is unique to endodermal cells, is a useful marker for endodermal identity (see Fig. 6). A more

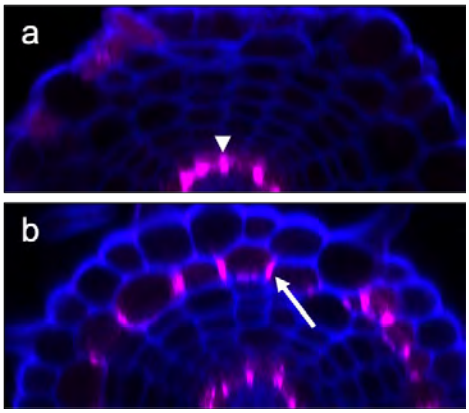

Fig. 6. CASP1, a Casparian strip-related protein, is revealed by red fluorescence in *Arabidopsis* root cross sections. CASP1 is natively found in the endodermal tissue layer (a, arrowhead) and is used as a marker for acquisition of endodermal identity in a non-native tissue type (b, arrow). Images courtesy of Colleen Drapek.

detailed visualization of changes in Casparian strip structure and development at a higher resolution will enhance our ability to study cell fate acquisition in mutant backgrounds.

### *Applications of the APS-U*

At the APS-U, the electron beam emittance approaches the diffraction limit in the hard x-ray energy range, with a much smaller emittance in the horizontal direction. The brightness is 2 orders of magnitude higher than that produced by the current APS. Coupled with developments in focusing optics and instrumentation, 10 nm or even smaller focus is realized with orders-of-magnitude increases in coherent flux. Such an upgrade enables scanning probes, including fluorescence microscopy and ptychography, to reach both length scales and time scales that are impossible to attain with present synchrotron techniques. Biological and environmental studies benefit greatly from the APS-U, specifically the investigation of metal pools and metalloproteins in cells. The higher throughput, higher spatial resolution for imaging, and higher spatial resolution for metal speciation measurements (e.g., microXANES) will be transformative for studies of cellular chemistry.

#### *High Throughput to Survey a Large Population of Samples in Both 2 and 3 Dimensions*

At present, a major limitation in studying elemental distributions and chemistries within cells is the small number of cells that can be studied in a given experiment. Because the measurements are extremely time-consuming, it is only possible to look at a very few cells. As a particular example, it can take several hours to image a single large cell like *Chlamydomonas* (8-10 µm) with 60-80-nm spatial resolution. By contrast, because of biological variability, biologists typically like to collect spectra from many cells, and they often want to compare treatments or genotypes. The higher-throughput elemental mapping measurements available with the APS-U  make it possible to study many more samples in two dimensions and to perform 3-D fluorescence tomography imaging, thus seeing and understanding cells in ways that were previously inaccessible due to a lack of spatial and dimension resolution, and statistical constraints.

#### *High Spatial Resolution*

The higher spatial resolution available with the APS-U permits the visualization of cellular features that are currently not resolvable. Subcellular structures, and elemental distribution and localization of small cells such as bacteria, will be well resolved using the <10-nm beam, which is not feasible with the current APS beam source. Nanospectroscopy will allow speciation to be differentiated at a few-nanometer scale. For example, the combination of the elemental mapping and redox speciation capabilities of the upgraded Bionanoprobe II (BNP-II) (Fig. 7 and Fig. 8) with complementary technologies for identifying cellular structure and activity, such as fluorescence microscopy, electron microscopy, and nanoscale secondary ion mass spectrometry (nanoSIMS), will be extremely powerful.

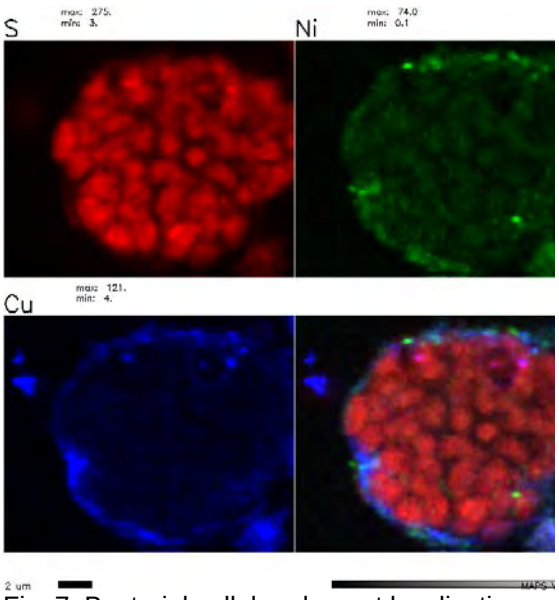


Fig. 7. Bacterial cellular element localization, as imaged with the current Bionanoprobe.

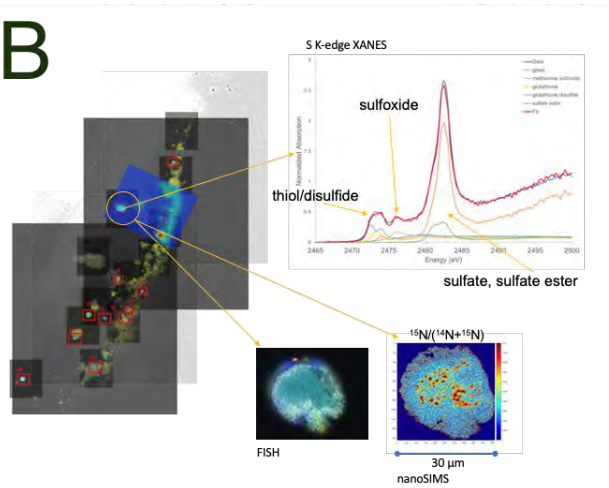


Fig. 8. Imaging with the BNP-II will be combined with complementary imaging by other methods, similar to current work on microbial aggregates at the Stanford Synchrotron Radiation Lightsource by Orphan and Magyar to correlate XRF imaging, XANES, and FISH-nanoSIMS. The upgraded BNP-II provides elemental speciation at similar spatial resolution to that of FISH-SIMS and electron microscopy, and makes studies such as these even more informative.

### *Correlative Analysis*

For nanoSIMS, fixed cells typically are embedded in a resin and sectioned with an ultramicrotome (200-300-nm thickness) before analysis. The elements of interest are detected with 50-100-nm spatial resolution. Stable isotope probing experiments (using $^{2}H$, $^{13}C$, $^{15}N$, $^{18}O$, $^{34}S$, etc.) are also possible using nanoSIMS. The advantage of these approaches is the ability to image dozens of cells and multiple elements with isotopic resolution simultaneously, which yields conclusions about co-localization of elements of interest and about cellular activity. Electron microscopy images of the cells make it possible to relate the site of elemental focus to a sub-cellular structure. A key limitation at present, however, is the inability to see the organization structure within organelles. Coupling these methods to studies using BNP-II post-APS-U permits the imaging of cellular features and elemental compositions at substantially improved spatial resolutions (10 nm), which permits the visualization of organelle organizational structure. This high resolution also opens opportunities for the study of transition metal pools (e.g., copper, manganese, etc.) in smaller cells than can be investigated at present — such as marine cyanobacteria, which are only 1-2 µm in diameter. Studying these marine organisms, which lie at the base of the food chain and which account for >20% of global photosynthesis, will have major implications for our understanding of global carbon cycling. Studying transition metal localization and speciation at such small length scales is impractical at present but becomes routine with the increased capabilities of the APS-U.

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## Environmental Microbiology and Biogeochemistry

Microorganisms inhabit nearly every environment on Earth, living in complex communities that form a network of integrated metabolisms that drive the biogeochemical cycles of major (hydrogen [H], carbon [C], nitrogen [N], and oxygen [O]) and many minor elements (e.g., phosphorus [P], sulphur [S], manganese [Mn], and iron [Fe]), particularly in highly redox-active environments. Redox-dynamic environments are characterized by variations in redox gradients over temporal scales ranging from minutes to years and spatial scales from microns to meters, created by episodic or periodic changes in the abundance of electron donors and acceptors. At the most basic level, microorganisms survive by exploiting redox disequilibria, conserving energy by transferring electrons from electron donors to electron acceptors. In typical aquatic and terrestrial environments, reduced organic compounds (e.g., acetate, $CH_4$), Fe(II), and sulfide, along with $H_2$, ammonium ($NH_4^+$), and Mn(II), are potentially significant electron donors, while $CO_2$, Fe(III), and $SO_4^{2-}$, as well as $O_2$, $NO_x^-$, and Mn(IV), are dominant electron acceptors, providing a complex network of electron donor/acceptor couples linking the biogeochemical cycles of major and minor elements. Although much is known about the coupling of H, C, N, O, S, Mn, and Fe biogeochemistry in general, many fundamental aspects relevant to their redox dynamics in aquatic and terrestrial environments have yet to be elucidated, as illustrated by the recent identification of several cryptic processes that are a key to understanding energy flux, elemental cycling, and $CH_4$ oxidation in anoxic environments (Beulig *et al.*, 2019; Flynn *et al.*, 2014; Hansel *et al.*, 2015; Kappler and Bryce 2017). Furthermore, the interplay between physical and chemical heterogeneities and differential microbial activity leads to the formation of microenvironments, which complicates upscaling of processes at the molecular/cellular level to the ecosystem scale, thereby limiting the inclusion of many biogeochemical processes into Earth system models.

Dynamic redox conditions are ubiquitous in aquatic and terrestrial systems across all geographic zones (polar, temperate, and tropical) and are evident in a diverse range of environments including stratified lakes, lacustrine and marine sediments, floodplains and wetland environments, groundwater-surface water interaction zones, atmospheric aerosols, and many others. Moreover, the redox processes that couple the biogeochemical cycles of H, C, N, O, S, Mn, and Fe in redox-dynamic environments play a critical role in major ecosystem processes including the assimilation of C, the mineralization of organic matter and accompanying release of $CO_2$, the formation and oxidation of $CH_4$, uptake and release of nutrients (e.g., N and P), and the availability and transformations of contaminants. Development of a robust and predictive understanding of coupled biogeochemical processes and cycles requires a comprehensive knowledge of the *chemical speciation* and *spatial organization* of the elements of interest in a system and the processes controlling *mass transfer.* Synchrotron-based spectroscopic and imaging techniques have enabled major advances in our understanding of these processes in a broad range of environments and systems.

### *Chemical Speciation*

Determining chemical speciation and understanding how biogeochemical processes impact chemical speciation is critical for defining how biological behavior, abiotic-biotic interactions, and molecular transformations control the reactivity and mobility of contaminants (e.g., uranium [U], technetium [Tc], and mercury [Hg]), nutrients (e.g., N, P, and C), and critical biogeochemical elements (e.g., S, Fe, and Mn). For example, in aqueous sedimentary environments, U can exist as dissolved, adsorbed, and mineralized species in some combination of the +4 and +6 valence states (and occasionally in the +5 valence state). Hexavalent U is stable under oxic conditions as the uranyl cation ($UO_2^{2+}$), and its aqueous complexes are relatively soluble. Tetravalent U is stable under reducing conditions and is typically present as sparingly soluble minerals (e.g., uraninite [$UO_2$] and triuranium octoxide [$U_3O_8$]). Although the low solubility of U(IV) minerals tends to limit U migration, U(IV) can be mobilized via formation of soluble complexes with dissolved organic or inorganic C. Thus, the mobility of uranium in

the subsurface depends strongly on its oxidation state, with U(IV) being significantly less soluble than U(VI). However, solubility also depends on the molecular form of U, which can be affected by adsorption to environmental surfaces.

Using synchrotron x-ray absorption fine structure to determine the valence and molecular structure of U, researchers examined the ability of montmorillonite clay minerals to adsorb U(IV) resulting from the reduction of U(VI) and compared it to that of iron and titanium oxide surfaces (Boyanov *et al.*, 2017). At low clay-surface:U ratios, the reduction of U(VI) leads to the formation of the mineral $UO_2$. However, at high clay-surface:U ratios (more typical of environmental conditions), a significant fraction of the resulting U(IV) is present as adsorbed non-uraninite U(IV) species (up to 50% of total U) (Fig. 9). The threshold U(IV) surface coverage above which uraninite formation begins was determined to be significantly lower for montmorillonite than for iron or titanium oxides, suggesting that metal oxides may play a more important role than clay minerals in stabilizing the non-uraninite species observed in natural sediments. The solubility and reactivity (e.g., susceptibility to reoxidation) of non-uraninite U(IV) is substantially greater than that for $UO_2$, so determination of the speciation of U(IV) is critical for accurate prediction of uranium transport in subsurface environments.

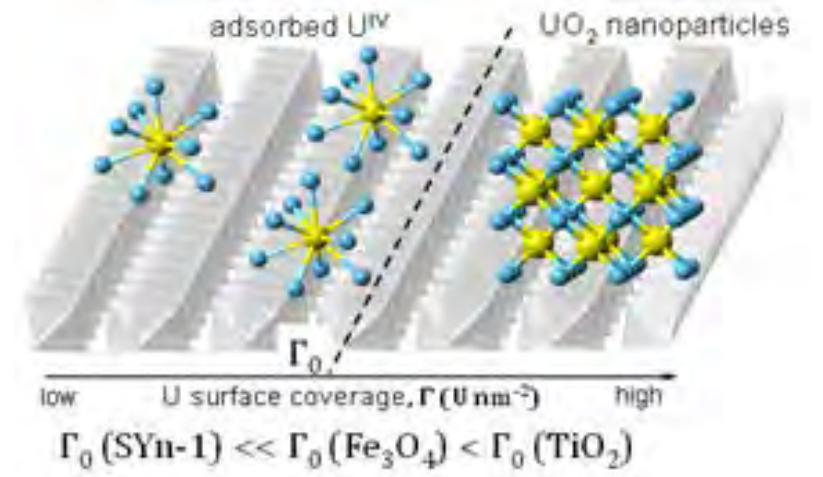


Fig. 9. U(IV) as uraninite ($UO_2$) dominates at high U loading, while non-uraninite U(IV) becomes significant with decreased U loading. From M. I. Boyanov; Latta, D. E.; Schere, M. M.; O'Loughlin, E. J.; Kemner, K. M.,, "Surface area effects on the reduction of $U^{VI}$ in the presence of synthetic montmorillonite" *Chemical Geology* **464**:110 (2017). Copyright © 2017 Elsevier B.V.

In addition to impacting contaminant fate and transport, chemical speciation affects the bioavailability of nutrients. For example, iron is a key micronutrient that is vital for all organisms. As such, the supply of bioavailable, soluble iron controls primary productivity in approximately 30% of the world's oceans. The significant contribution of atmospheric aerosols to the bioavailable iron budget in vast ocean regions underscores the need to understand the controls and transformations of aerosol iron solubility during atmospheric transport. Using a combination of iron near-edge x-ray absorption spectroscopy and wet chemical techniques, researchers examined the chemical speciation of iron in aerosol samples originating from Saharan dust plumes collected at three sites located in the Mediterranean, the Atlantic, and Bermuda representing different atmospheric transport lengths and time scales (Longo *et al.*, 2016). Iron(III) oxides were a component of aerosols at all sampling sites and dominated aerosol iron in Mediterranean samples. In Atlantic samples, Fe(II & III) sulfate, Fe(III) phosphate, and Fe(II) silicates were also contributors to aerosol composition. With increased atmospheric transport time, Fe(II) sulfates became more abundant, aerosol iron oxidation state became more reduced, and aerosol acidity increased. Atmospheric processing, including acidic reactions and photo-reduction, likely influence the form of iron minerals and the iron oxidation state in Saharan dust aerosols and contribute to increases in aerosol iron solubility (hence, bioavailability).

Overall, these findings, largely derived from synchrotron-based analyses, suggest that a combination of factors affects aerosol iron solubility during long-distance atmospheric transport and emphasize the need to consider reductive mechanisms as well as proton-induced solubilization of aerosol iron in modeling studies.

In a related study, researchers used a combination of sulfur near-edge scanning-probe x-ray fluorescence (XRF) spectroscopy and XRF microscopy to determine the chemical speciation of atmospheric sulfur in ambient aerosol samples collected from the greater Atlanta area (Longo *et al.*, 2016). The aerosol particles contained sulphur in two oxidation states: S(0) and S(VI) (Fig. 10). The finding of S(0) in micron-size aerosol particles was unique in aerosol studies and possible only through synchrotron-based approaches. S(0) in aerosols has important implications for identifying sources and the production of acidic species in aerosols during transport. In turn, such acidic reactions solubilize metals, such as iron, that control the production of highly reactive radical species in the atmosphere.

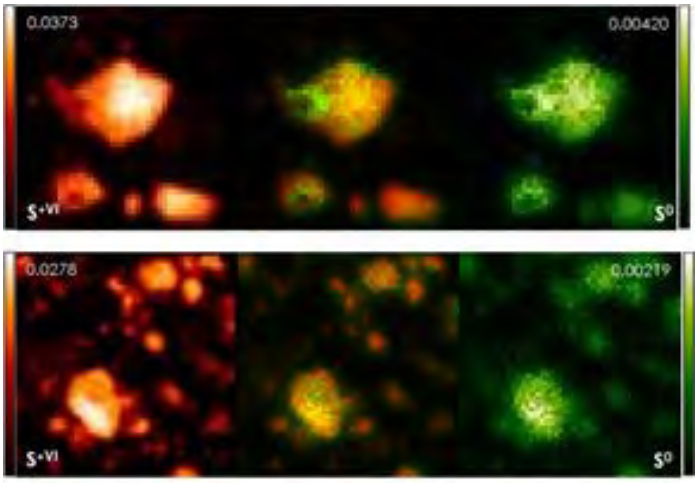


Fig. 10. X-ray fluorescence map of sulphur for two ambient aerosol particulate matter samples. The sulphur elemental density maps are scaled from black (lowest intensity) to white (highest intensity). Corresponding XRF intensity scale bars are shown on the sides of the elemental density maps. For both samples, S(VI) (red) is concentrated in the center of the particle, where the particle mass is highest. S(0) (green) has a more varied spatial distribution, but is generally concentrated in one specific area. An overlay of the S(0) and S(VI) elemental density is shown in the center. Particles are approximately 2 to 3 µm in diameter. From A. F. Longo; Vine, D. J.; King, L. E.; Oakes, M.; Weber, R. J.; Huey, L. G.; Russell, A. G.; Ingall, E. D., "Composition and oxidation state of sulfur in atmospheric particulate matter," *Atmospheric Chemistry and Physics* **16**:13389 (2016). https://www.atmos-chem-phys.net/16/13389/2016/

### *Spatial Organization/Structure*

Particle structure/texture, interfacial phenomena, and varying hydrodynamic dispersion can greatly impact biogeochemical and biogeophysical properties in air, aquatic, and terrestrial (soil/sediments) systems, thus forming heterogeneously dispersed micro-environments with distinct activities and conditions that likely contribute to "hot spots and hot moments" over time and space (McClain *et al.*, 2003). However, little is known about how biogeochemical and hydrogeophysical processes at relevant scales ultimately impact field-level system behavior and function, and few studies have determined the proper scale at which to delineate these relationships. To address this problem, it is necessary to distinguish the diversification, dispersal, selection, and drift of microbial consortia under field-relevant conditions from the interfacial to field scale and how these conditions dictate microbial activity in a predictable fashion. The relationship between biosystem complexity and ecosystem function rarely follow consistent patterns, and we do not have a complete understanding of the structure-function relationships across the gas, aqueous, and solid-matrix boundaries over time.

Biofilms are ubiquitous in soils and sediments, which themselves are highly heterogeneous across multiple spatial scales and whose structural organization and attendant distribution of biogeochemical processes are similarly challenging to characterize, particularly in the context of hot spots and hot moments. The combination of XRF imaging and x-ray absorption fine structure is a powerful approach for probing the spatial distribution of elements and their chemical speciation in hydrated soils and

sediments. For example, this approach was used to determine the time-dependent spatial distribution of uranium concentration, valence state, and chemical speciation in a diffusion-limited, ethanol-amended column with sediment from the Oak Ridge National Laboratory Integrated Field Research Challenge site. As shown in Fig. 11, XRF measurement revealed that the distribution and speciation of uranium changed dramatically at the ~200-μm length scale, both over short times in the bio-stimulated diffusion layer (dark) and over longer times in the entire column. Reducing the beam size to 50 × 50 μm revealed localized areas (hot spots) of high concentrations of $U^{VI}$-phosphate in an otherwise mostly reduced area (as determined by a 500 × 500-μm beam). Although these smaller zones of elevated $U^{VI}$ concentration were a relatively small part of the total U balance, identification of their presence enables an improved understanding of the evolution of the non-uraninite U(IV) species in natural sediments.

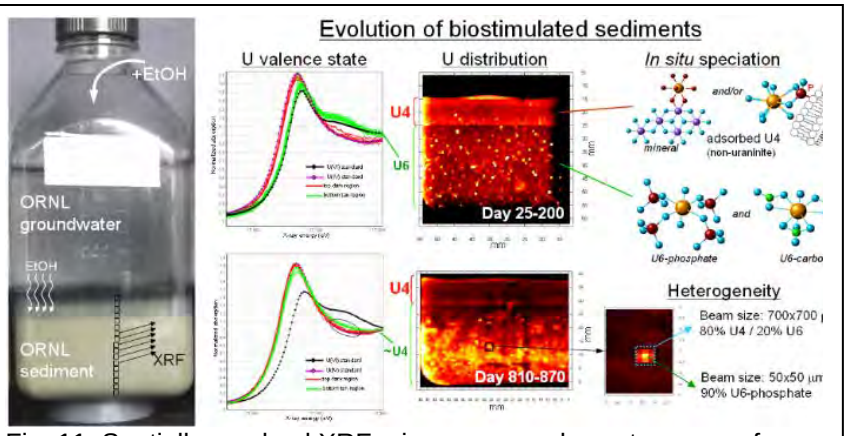

Fig. 11. Spatially resolved XRF microscopy and spectroscopy of ethanol-stimulated sediments from the Oak Ridge National Laboratory Integrated Field Research Consortium, demonstrating *in situ* molecular-level characterization over extended periods of incubation.

### *Mass Transfer*

The transformations and mobility of major/minor elements, nutrients, contaminants, and redox-active elements through terrestrial, aquatic, atmospheric, and subsurface environments are affected by their physical-chemical interactions with geomedia, such as sorption to the biogeochemical matrix, precipitation/incorporation into a solid phase, and colloidal transport. The occurrence and outcome of transformation processes are highly dependent on the chemical conditions at the location of the reactions (e.g., the concentration of dissolved reactants). Whether mass transfer is under diffusion-limited or advective-flow conditions, distinct transport properties for different reactants can be expected to result in spatial heterogeneity. Improved molecular-scale monitoring and understanding of the chemical interactions between reactants at the solution solid interface is required for better prediction of environmental processes.

Chemical speciation and distribution ultimately affects the mass transfer of substances and their potential impacts on environmental systems over a range of spatial scales. For example, at micron scales, mass transfer in bacterial biofilms can strongly affect corrosion of metal pipes and impact the efficiency of electrodes. At micron scales, aerosol particle surfaces are transformed by both acid-producing and photo-reduction reactions. At meter-to-kilometer scales in aquifers, oxidation or reduction of metals and organic substances through bacterially mediated processes can act to solubilize, immobilize, or decompose these substances. At the same scales in atmospheric systems, chemical transformations of aerosol particles during transport in urban environments can increase their tendency to form toxic radical species. At global scales, both aerosol transformations during long-range transport and the activities of marine microorganisms impact the solubility of key nutrient elements that can ultimately impact ocean productivity.

The application of synchrotron x-ray capabilities relevant to the study of mass transfer across scales is shown in a study by researchers tracing the movement of nutrient elements like iron through marine

systems (Ingall *et al.*, 2013). Ecologically significant uptake of iron by small microorganisms, called "diatoms," from the extremely low dissolved iron waters of the Southern ocean was quantified from synchrotron-based studies (Fig. 12). Synchrotron research resulted in a new potential pathway for iron removal from marine systems involving structural incorporation of reduced organic iron into biogenic silica. Export of iron incorporated into biogenic silica may represent a substantial unaccounted loss of iron from marine systems. For example, in the Ross Sea, burial of iron incorporated into biogenic silica is in the same range as the major bioavailable iron inputs to this region. As a major sink of bioavailable iron, incorporation of iron into biogenic silica may shift microbial population structure towards taxa with relatively lower iron requirements, and may reduce ecosystem productivity and associated carbon sequestration.

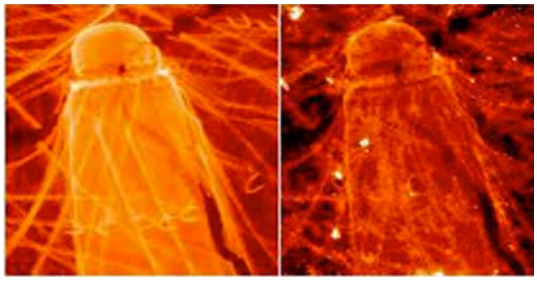

Fig. 12. X-ray fluorescence map of silicon (left) and iron (right) concentrations in an Antarctic diatom. Brighter colors represent higher concentrations. The diatom is approximately 80 x 40 µm. From E. D. Ingall; Diaz, J. M.; Longo, A. F.; Oakes, M.; Finney, L.; Vogt, S.; Lai, B.; Yager, P. L.; Twining, B. S.; Brandes, J. A., "Role of biogenic silica in the removal of iron from Antarctic Seas," *Nature Communications* **4**:1981 (2013). ©2013 Springer Nature Publishing AG

Transformations in well-mixed systems can be characterized with "bulk" observation methods. However, in many environmental systems, such as those described above, reactions occur at spatial and temporal scales that generate localized biogeochemical environments. Synchrotron-based methods, which allow tracing of chemical processes at the sub-micron scale, are especially vital for understanding such spatially heterogeneous systems.

### *Opportunities for the APS-U*

The spatial organization, chemistry, and structure of heterogeneous microenvironments can greatly impact the biogeochemical properties within air, aquatic, and terrestrial systems. Given the ubiquity of these complex biotic and abiotic microenvironments, techniques are needed that can characterize the chemistry within them. The APS-U revolutionizes investigation of biogeochemical processes in environmental systems by providing orders-of-magnitude improvement in key parameters, such as coherence and brightness, that are directly relevant to almost all hard x-ray imaging, scattering, and spectroscopy experiments, and provide 3-D resolution from angstroms to centimeters, time resolution from picoseconds to days, and the ability to perform ultrasensitive trace element analysis. Whether the challenge is microbial and biogeochemical interactions in soils/sediments or the environmental behavior of nanoparticles, the APS-U dramatically improves the ability to study highly relevant, smaller sample volumes with greater precision, spatial resolution, and throughput, and provides the detection sensitivity and spatial resolution required to investigate nutrients and contaminants at environmentally relevant concentrations in complex environmental matrices.

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## Soil Structure and Chemistry

Soil is a remarkably complex biomaterial that exists as a dynamic, structured, self-organized but highly heterogeneous mixture of mineral particles, organic matter, water, air and countless living organisms (Young and Crawford, 2004). Soil particle sizes span over 3 orders of magnitude, from clay (<0.002 mm), silt (0.002-0.05 mm), and sand (0.05-2 mm) to coarse fragments (>2 mm). Soil pore spaces occur over similar size ranges with varying degrees of connectivity and network tortuosity. Soil organic matter — derived from the products and residues of both living and dead organisms including plants, microbes, and soil fauna — exists as a continuum of decay products of varying size, composition, and position within the soil matrix. The spatial organization of these components (i.e., soil structure) is dynamic and linked to environmental processes such as wetting/drying, soil mineralogy and biogeochemical interactions, organic matter decomposition, and the biophysical actions of living plant roots and soil biota.

Soil processes at even the smallest scale exert strong controls over the fate of biologically important elements in ecosystems. The dynamic and hierarchical organization of soil structure exerts significant control over these processes (Jastrow *et al.*, 2007). For example, large pools of soil organic matter can persist for centuries or millennia via isolation in pores and associations with minerals at micron and submicron scales. If mobilized or otherwise made accessible, that organic matter may become more susceptible to microbial attack, affecting the fate of carbon, nutrients, and contaminants in terrestrial ecosystems.

The controls on soil hydrobiogeochemical cycles and their intimate coupling with plant productivity are at the core of Earth's life support systems (Fig. 13 and Fig. 14). Because those controls are intimately tied to fine-scale physical, chemical, and biological interactions and feedbacks in soils, the APS-U is game-changing for high-throughput, fine-scale observations critical to revealing hydrobiogeochemical processes and their control mechanisms, with replication, through time. Iteratively integrating measurements with analytical computation and process modeling will be particularly important. In collaboration with users, the APS-U team is positioned to couple high-performance computing with cutting-edge experimentation for iterative hypothesis testing and model improvement. The extent to which these small-scale phenomena must be considered to improve the predictive power of ecosystem, regional, and global models is a critical but unresolved question. Addressing this question demands new approaches that will permit real-time or near-real-time imaging, complementary spatial and chemical analyses, expanded scales of interrogation with preservation of high resolution, and high-throughput capacity for increased statistical rigor and replication. The APS-U provides all of these capabilities.

### *Linking Global Significance with Pore-Scale Mechanisms*

Globally, soils hold the largest terrestrial reservoir of organic carbon. Current estimates (2300 Pg to a depth of 2 m) amount to about four times the carbon stored in vegetation and 2¾ times that in the atmosphere (Cias *et al.*, 2013; Hugelius *et al.*, 2014; Batjes, 2016). The total soil reservoir could exceed 3000 Pg C when peat, wetland, and permafrost soils below a depth of 2 m are included in the estimate (Köchy *et al.*, 2015). Thus, relatively small changes in soil organic matter dynamics and storage could have a large impact on the global carbon cycle. Furthermore, soil organic matter cycling at local scales is a key factor influencing soil health, water and nutrient availability, and other soil functions, and thus a variety of ecosystem services (Koch *et al.*, 2013; Janzen, 2014).

The storage of organic carbon in soils is dynamic and depends on the balance (or imbalance) between the rates of (1) carbon inputs from the synthesis activities and turnover of plants, microbes, and fauna, and (2) carbon losses from soil organic matter due mostly to decomposition (mineralization to the

greenhouse gases carbon dioxide and/or methane). The persistence of soil organic matter is regulated largely by factors controlling decomposer access to organic substrates and their ability to break down, transform, and utilize those substrates. Although the chemical composition/complexity of soil organic matter can influence decomposition and greenhouse gas emission rates, even readily decomposable forms of organic matter can be protected from attack by decomposers through chemical associations with clay minerals, physical barriers that impede access, and micro-environmental constraints (such as oxygen, water, and nutrient availability) on decomposer metabolism and movement (Jastrow *et al.*, 2007; Kleber and Johnson, 2010; Janzen, 2014). For example, the structural organization of soil includes pore networks of varying size and connectivity (Fig. 13) that control the accessibility of soil organic matter to microbes and extracellular enzymes. Stabilized organic matter may become decomposable if changes in soil structure or the hydrologic connection of soil pores expose previously-isolated soil organic matter to microbial/enzymatic activities.

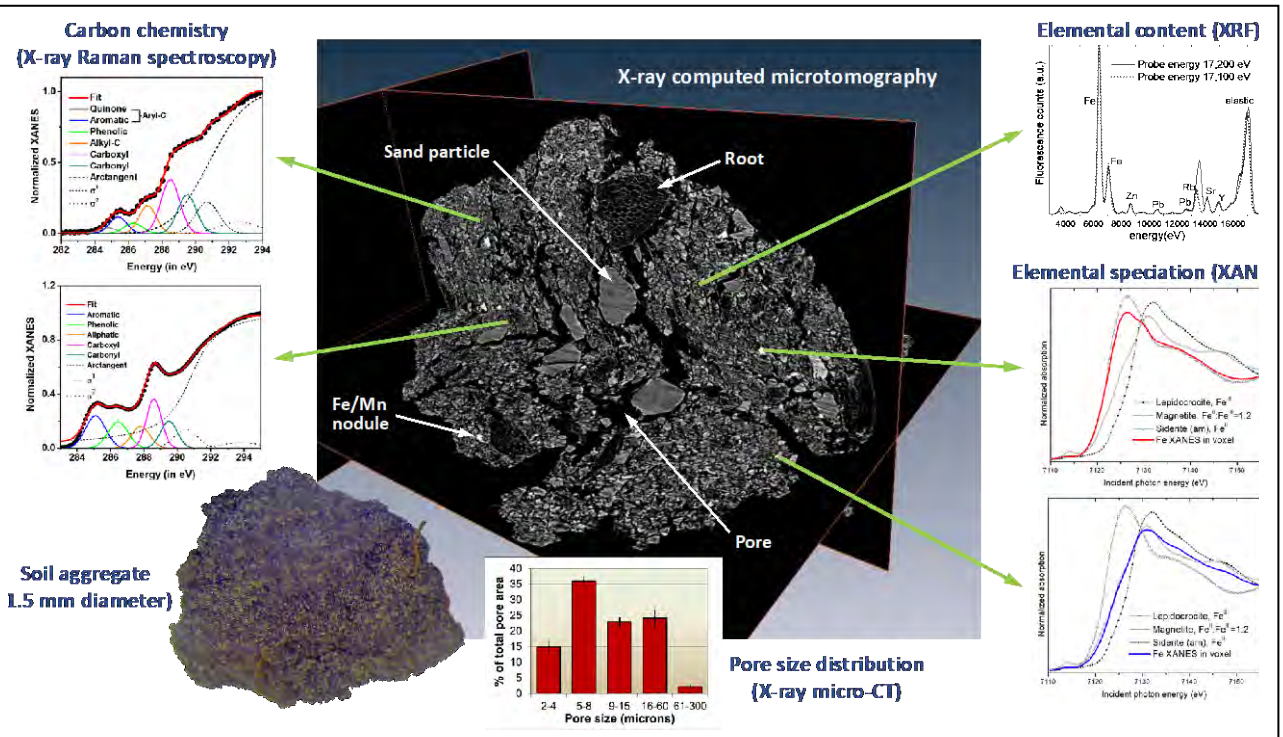


Fig. 13. Soil consists of a spatially heterogeneous mixture of mineral particles, organic matter in various stages of decay, and living organisms. Interactions among these components produce a structural organization of pore networks of varying size and connectivity that control water movement and retention, aeration, redox reactions, and the accessibility of soil organic matter to microbes and extracellular enzymes. With the APS-U, 3-D, spatially resolved information on soil organic matter chemistry can be coupled with variations in the elemental content and speciation of inorganic soil components at each voxel of interest within the context of intact soil structures. From “Technologies for Characterizing Molecular and Cellular Systems Relevant to Bioenergy and Environment,” DOE/SC-0189, U.S. Department of Energy Office of Science, science.energy.gov/ber/community-resources/.

Because plant productivity is often limited by soil nutrient availability, the interface between living roots and soils (the rhizosphere) is a central commodities exchange, where organic carbon exuded from roots fuels microbial and faunal decomposers that can, in turn, release nutrients stored in soil organic matter and make them available to those roots. The ongoing exchange of carbon and nutrients in the rhizosphere is operating in the context of oscillating water flow around active plant roots that is driven by diurnal and seasonal transpiration patterns constrained by water retention and flow through the soil’s

pore network (Fig. 14). The implications of these rhizosphere processes (e.g., the dynamics of decomposition of soil organic matter and soil carbon storage, protozoal grazing and associated trophically-driven mineral nitrogen release, competitive cation exchange over diel cycles, and root and microbial influence over critical associations of organic matter with minerals) remain largely unknown.

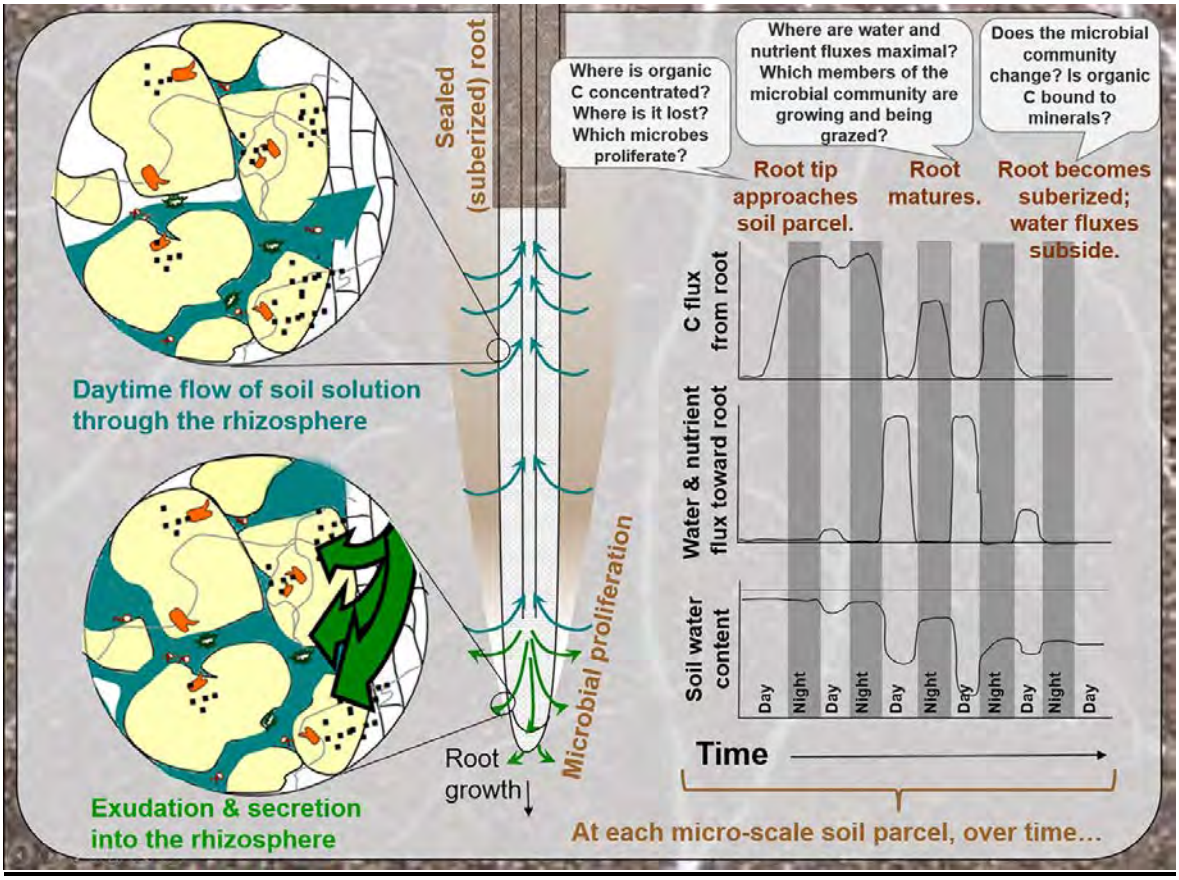


Fig. 14. Dynamic interactions among roots, microbes, minerals, and organic matter in the rhizosphere. The unique strength of the APS-U is the ability to probe extremely small-scale chemistry and structure with improved resolution, speed, and computational capacity, enabling observations to move beyond snapshots to capture dynamic processes.

The unique strength of the APS-U is the ability to probe extremely small-scale chemistry and structure with improved resolution, speed, and computational capacity, enabling observations to move beyond snapshots to capture dynamic process. In the case of soil, the composition of soil organic matter is typically studied through various fractionation approaches including physical, chemical, thermal, and spectroscopic techniques as well as combinations of various methods (von Lützow *et al.*, 2007). However, soil fractionation approaches often are limited because (1) most fractions do not represent a single homogenous pool; and (2) physical fractionations, chemical extractions, and even spectroscopic techniques destroy soil structures and ignore potentially crucial heterogeneity. Modern analytical tools (e.g., NanoSIMS, Fourier-transform ion cyclotron resonance mass spectrometry, synchrotron-based spectroscopic techniques; see Fig. 13) are providing new insights into the molecular-scale composition of organic materials and organo-mineral interactions in soils (Lehmann *et al.*, 2007, 2008; Tfaily *et al.*, 2015, 2017; Steffens *et al.*, 2017). In addition, synchrotron ultra-small-angle x-ray scattering of intact and combusted soil has been used to quantify the organic matter occurring within submicron pores (McCarthy *et al.*, 2008). Finally, synchrotron and bench-scale x-ray computed tomography can provide 3-D images of soil structure (Fig. 13), but clear separation of organic and mineral components (other

than roots and particulate organic matter) and microbial locations remains elusive (Bailey *et al.*, 2013; Kravchenko and Guber, 2017).

For soil specifically, both long- and short-term dynamics in soil organic matter pools are of current scientific interest and societal importance. Soils are complex, acting as long-term, large reservoirs of organic carbon; and as a participant in an active commodities exchange where plant roots, microbes, and soil minerals compete and cooperate, maintaining soil fertility.

Dynamic water retention and movement represents a significant knowledge gap limiting our understanding of the dynamics of soil organic matter processing and soil carbon sequestration, particularly at soil pore and smaller spatial scales. As climate change proceeds towards a future of more frequent and more extreme events that may be centered around drought, precipitation, and storms, the role of water movement through soils at all scales becomes an increasingly important factor to consider. In particular, changing hydrologic connectivity through increased water saturation or disturbance will impact soil aeration and mineralization of soil organic matter, and greenhouse gas fluxes to the atmosphere. Under conditions of partial-to-full water saturation, relatively labile compounds can desorb and diffuse from micropore domains to macropore networks to locations accessible to microorganisms. These transport processes are rarely explicit in mathematical models of soil organic matter decomposition and gas species flux, because the fundamental mechanisms have not been experimentally described. Mechanism-based models would allow us to integrate fundamental knowledge of soil organic matter transformations and the physical and biogeochemical factors controlling soil organic matter stabilization and decomposition, and also provide a mathematical basis for evaluating current models and for upscaling to new models at larger scales. Currently, micron- and pore-scale soil environmental properties affecting the stabilization, priming, and mineralization of soil organic matter are not explicitly considered in Earth system models, which operate at large scales and thus employ greatly simplified mathematical representations of coupled element and water cycles. A critical but unresolved question is to what extent these small-scale phenomena must be considered to improve the predictive power of Earth system models as climate change occurs.

Data from the APS-U, capturing details of soil structure and some chemistry (Fig. 13 and Fig. 14), and ideally also enabling insights into microbial colonization and activity, mineral-microbe-root interactions, and water content and flow, will be extraordinarily valuable for understanding recycling of nutrients in soil; maintenance of soil fertility and water retention through incorporation of organic materials; associated soil carbon storage and mechanisms of absorption to (and desorption from) minerals; and dynamic co-control of biogeochemistry by microbial catalysis and diffusive/advective delivery of substrates. All these processes are intimately connected to soil physico-chemical fine structure, diffusion, and advection.

Currently, we cannot assess the entirety of soil structure confidently at all biogeochemically relevant scales. Therefore, we lack the context for recognizing commonalities and heterogeneities at the heart of interactions among organisms and the soil itself. We also cannot resolve the location of water within the soil pore matrix, particularly in fine pores <20 μm in diameter, and we cannot observe how water moves through soil pores in all directions, *in real time*. Mechanistic understanding of dynamic soil-plant-microbe interactions will require technical and computational developments aimed at visualization and quantification at the micron spatial and temporal scales in which microbes, roots, and minerals interact. With such developments in hand, iterative experiments and modeling can be employed to decouple correlated dynamics to get at causal mechanisms.

### *Opportunities for Linking the APS-U with Other User Facility Strengths to Address Critical Questions and Knowledge Gaps in Soil Science*

An overall goal within soil science is to understand mechanisms underlying soil carbon sequestration (protection, absorption to minerals, and localization) and coupled biogeochemical processes providing essential ecosystem services to humanity. Particularly important is the partitioning of microbial and mineral vs. diffusive/advective control over diverse soil biogeochemistries, as affected by history and expected change (e.g., land use, drought or intensified rainfall, and climate warming). As major goals, tomography and spectroscopy at the APS-U (coupled with measurements at other user facilities) could ultimately provide:

- *3-D pore size/shape-connectivity/soil organic matter-inclusions.* The APS-U offers unprecedented fine-scale visualization, through tomography, of 3-D soil pore size, shape, connectivity, and low-density inclusions in a high-throughput manner. A goal must be to link across scales; for example, linking to tomographic systems at other user facilities (e.g., the Environmental Molecular Sciences Laboratory, EMSL) capable of scanning larger samples but at less-sharp resolution. These combined data will be invaluable for describing the diffusion of gases and solutes (both signals and resources) in soils, as well as potentially allowing investigation of pore-filling dynamics.

- *3-D water flow paths and water location.* While we have a significant and compelling understanding of water movement and chemistry at macroscales, we rely heavily on macroscale-derived theory to translate these processes to finer scales. Though water cannot be directly visualized using hard x-rays, it may be possible to gain insights into water flow and discontinuity in soil at extremely small scales using soil solution “doped” with solutes (e.g., potassium iodide) that can be detected via the high-energy x-ray beams of the APS-U. In combination with larger-scale direct imaging of water with neutrons at the Spallation Neutron Source (SNS) facility, the APS-U affords the ability to characterize soil water movement (including along preferential paths) and the structure of dynamic water pools (including disconnected pools in very dry soils) across a range of soil saturations.

Partnerships with the EMSL and the SNS may further advance water visualization capabilities in soils (see additional discussion in next section). Nuclear magnetic resonance technologies and neutron-based imaging approaches have had some success monitoring movement of water through the soil matrix and it may be possible to capture wetting in two dimensions at 20-μm resolution with 1-min time resolution.

- *3-D organic carbon and microbial distribution, particularly in association with plant roots and minerals.* The high-throughput and fine-scale resolution offered by the APS-U is a tremendous opportunity to probe in 3-D the microbe-root-mineral-soil organic matter interactions driving the biogeochemistry underlying Earth’s life support systems. Hard x-rays are not commonly used to view lighter elements such as carbon, nitrogen, and oxygen in organisms and organic matter, but improvements in avoidance of interference by iron (a significant elemental component of soils) may enable the chemical nature of these lighter elements to be obtained by x-ray Raman spectroscopy (Fig. 13). Perhaps more immediately tractable is the use of labeling techniques such a quantum dots or halogen *in situ* hybridization to tag specific microbes based on DNA sequence in phylogenetic and functional marker genes, which offer the possibility of observing time courses of microbial community development through sequential harvest, or the distributions of particular microbial functional gene capacities as a function of location in 3-D physical and chemical soil space. Ultimately — if techniques could be combined — dynamic, fine-spatial-scale observations could reveal the microbial and mineral “players” involved in

altered carbon:nitrogen ratios of organic materials, transformation of moieties, and the contribution and transformation of organic materials by plant roots over time.

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## Atmospheric Science

This session of the workshop discussed new and expanded opportunities with the APS-U that are of relevance to atmospheric science. Two of them are highlighted below.

### *Atmospheric Aerosols*

Atmospheric aerosol particles have significant impacts on a number of environmental fields, including air quality, visibility, public health, biogeochemistry (Session 5), and climate change. Effects of these particles are highly variable depending on their chemical and physical properties. However, characterization of atmospheric aerosols using conventional microspectroscopy techniques is challenging because of the sub-micrometer size and trace-level concentration of these particles (e.g., several 10s per $cm^3$ air). While U.S. Department of Energy (DOE) facilities such as the EMSL, the APS, and the Advanced Light Source have been used for characterizing atmospheric aerosols, these efforts have been limited by the number of samples analyzed and analysis techniques for atmospheric trace elements such as carbon, iron, phosphorous, copper, and other compounds in aerosol.

Iron-containing dust aerosols supply ~95% of the iron nutrients to the open-ocean biota, limiting the ocean's biologically driven carbon sequestration; but unknowns remain, largely about the mineral form, chemistry, size, and bioavailability of aerosol iron. In the past several years, research has demonstrated the capability of characterizing the iron mineralogy and oxidation state in atmospheric dust aerosols, using synchrotron-based XANES spectroscopy and micro-XRF measurements at the APS beamline 2-ID-D (Longo *et al.*, 2016; Ingall *et al.*, 2018). This capability has shown great promise in unraveling the role of dust particles in transporting bioavailable iron to remote oceans.

However, statistical classification of and advanced learning about the aerosol samples were hampered by the limited number of samples that could be analyzed and the quality of the aerosol sample images analyzed at the APS. The APS-U offers an opportunity to generate experimental data streams at extremely high rates (i.e., 2-to-3 orders of magnitude higher than today) via increases in source brightness, advances in x-ray optics, and the development of new detector technology. The improved spatial resolutions, down to 5 nm and better, will extend the current spatial resolution and chemical contrast to the nanometer level, making it possible to use nanoprobe techniques for three-dimensional/four-dimensional elemental and chemical imaging. Opportunities for extending the range of aerosols samples from mineral dust to organic aerosols and various forms of carbon aerosols could also become feasible as the ability to control the beam strengths at sampling ports is implemented. Developing atmospheric aerosol imaging and sample manipulation will be essential to fully leveraging this opportunity.

### *Cloud Chamber – Particle Interactions with Clouds*

A key challenge in understanding the uncertainties related to increasing greenhouse gases and aerosols is closely tied to their interactions with clouds. There are a number of unresolved scientific challenges related to clouds and cloud-aerosol interactions, including:

1) Increasing aerosol number concentrations is understood to lead to smaller cloud droplets and increasing cloud albedo (Twomey effect). How does this process operate at a molecular scale and what are the key water-vapor/water-droplet-aerosol interactions?
2) The inclusion of certain types of aerosols in cloud droplets increases the absorptivity of radiation by these particles. What physical and chemical forces control this inclusion process?

3) Ice nucleation under upper tropospheric environmental conditions is poorly understood. What are the roles of dust particles and microbial matter in the ice nucleation process?

A controlled environment for performing experiments that dwell in the atomic/molecular scale of the cloud-aerosol processes would be a big jump in our observing capability for these systems. Access to a controlled environment that is similar to a few cloud chambers that are in operation around the world (and that can be probed by APS-U beamlines) would be an enormous resource for the Earth system modeling and measurements community. The APS-U provides a unique opportunity to develop a cloud chamber that is focused on aerosols and aerosol-cloud droplet interactions in unprecedented molecular detail. Nanoprobe techniques for three-dimensional/four-dimensional elemental and chemical imaging available at the APS-U have the ability to image these aerosols in three-dimensional detail and on a time scale that would open an unprecedented view of the processes at the molecular scale.

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## DOE User Facilities as Models, and Collaboration

### *The Environmental Molecular Science Laboratory*

The Environmental Molecular Science Laboratory is a world-class environmental molecular science user facility that provides the scientific community with state-of-the-art capabilities and expertise for gaining a predictive understanding of complex biological, Earth, and environmental systems for energy and infrastructure security in support of DOE biological and environmental research missions. The EMSL capabilities are organized into six integrated research platforms that provide the framework to address and solve key research questions through the integration of experimental, analytic, and modeling approaches. They include: omics (proteomics, metabolomics, and transcriptomics); bioimaging and structural analysis; cell spatial and temporal dynamics; isotope and chemical analysis; plant, soil, and subsurface transport; and theory and simulation, data analytics, and visualization. In addition, experimental capabilities such as nuclear magnetic resonance, mass spectrometry, and advanced electron and super-resolution microscopy can be applied to a vast array of scientific challenges that support the DOE mission. Access to EMSL resources is provided through a peer-reviewed proposal process. Staff are available to assist researchers in identifying and making optimal use of the many capabilities that are available at the EMSL. For more details, please see https://www.emsl.pnl.gov/emslweb/ or contact Nancy Hess (nancy.hess@pnnl.gov), Deputy for User Science. (*Workshop participants felt that the EMSL user facility access model should be considered for employment at the APS for scientists focused on complex biological and environmental science*.)

### *The Spallation Neutron Source*

The Spallation Neutron Source is a DOE user facility located at Oak Ridge National Laboratory. The SNS produces neutrons via an accelerator-based system that delivers short (microsecond) proton pulses to a steel target filled with liquid mercury through a process called “spallation.” Those neutrons are then directed toward state-of-the-art instruments that provide a variety of capabilities to researchers across a broad range of disciplines including physics, chemistry, biology, and materials science.

Neutrons provide unique information on biological systems associated with multi-scale phenomena across the angstrom-to-micron range. Because they have no charge, neutrons cause no radiation damage and are highly penetrating, allowing measurements under native physiological conditions. Moreover, neutrons have energies similar to those associated with atomic and molecular motions, allowing studies of dynamic processes on time scales ranging from picoseconds to microseconds. Because photons and electrons interact with the atomic electric field, hydrogen is all but invisible to them. In contrast, neutrons interact with nuclei making it possible to observe the lighter elements such as hydrogen and deuterium, and distinguish these light elements next to heavy ones. Furthermore, the neutron scattering cross-sections of hydrogen and deuterium are very different, making it possible to selectively highlight different components within a complex system.

For mesoscale structural studies of environmental samples related to soils, microbes, and biogeochemistry, small-angle neutron scattering with $H_2O/D_2O$ contrast variation provides unique information about properties of these systems, including pore-size distribution and accessibility, and spatial organization (1- to 500-nm resolution). Ultra-low-angle neutron scattering extends accessible length scales to several microns. Neutron radiography and computed tomography image hierarchical and complex materials at a resolution of ~50 µm and can be used to obtain information about the kinetics of water movement in soils, wetting characteristics, and insight into biofilm formation. In addition, root-soil interactions can be studied in order to characterize root growth and water partitioning in intact systems.

Neutron scattering techniques are also ideal for studies of plant cell wall structure because they span the length scales (angstroms to micrometers) and time scales (picoseconds to minutes) relevant to lignocellulose characterization, while differentiating between components such as lignin and hemicellulose, as well as the crystalline and amorphous phases of cellulose. Moreover, because neutrons are non-destructive and highly penetrating, it is possible to study structural changes in insoluble biomass samples *in situ* and in real time.

Neutron scattering spectroscopy is a two-dimensional technique that provides information about atomic motions in both time and space. Inelastic and quasi-elastic neutron scattering provide information about vibrational modes, molecular motions, and diffusive properties of biomolecules and their hydration water on the picosecond-to-nanosecond time scale. These techniques are appropriate for the study of the dynamics of complex materials such as plant cell walls and soils, and the hydration water associated with these systems. Using neutron spin-echo spectroscopy it is possible to probe slower motions at micro- to milliseconds, such as motions associated with membranes in living cells and domain motions in proteins.

### *Multi-Facility Access — the Facilities Integrating Collaborations for User Science*

Many of the science challenges discussed at the workshop illustrated spatiotemporal complexity that requires multimodal analysis of samples for scientific advancements. Many of the remarkable achievements presented integrated capabilities and expertise across the DOE user facilities. Currently, these principal investigators must not only state that a certain capability could provide insight, they then have to apply for access to each facility independently, and integrate and interpret the resulting data. As a first step, user facilities can accelerate this process by contributing to a common Website or portal so that potential users can understand what is available at different user facilities, provide use cases, and describe how they can be used together. Moving forward, facilities can explore opportunities for joint facility access through a single proposal modeled after the Facilities Integrating Collaborations for User Science program between the EMSL and the Joint Genome Institute. The development of common data formats, metadata, and data servers so that data from different facilities can be more readily discovered, accessed, and integrated will also inspire more users to leverage multiple facilities. Finally, as more users utilize the array of capabilities, facilities can develop more versatile sample holders or specialize in sample preparation facilities that can be leveraged across the DOE user facility complex. These steps can help release the untapped potential for transformational science advances as users explore and define new approaches to solve these challenges.

### *Computation*

Development of mechanistic models for the biological and environmental phenomena listed above can benefit from close coupling between the measurements and models. The APS-U has the advantage of an exascale computing facility (Aurora at Argonne, to be deployed in 2021 [A21]) operating in close proximity, with a high-bandwidth network connectivity. Aurora will enable on-demand computation and analysis for APS-U workflows to accelerate scientific discovery. The development of deep learning software is a key target for A21. Thus, leveraging the computational resource of Aurora in terms of both the hardware and software to develop a newer breed of mechanistic models for biological and environmental research becomes viable. The APS-U will increase the amount of data collected by many orders of magnitude. The APS-U capability for producing images in 3-D will mean that, for a 1-$mm^3$ sample probed with 10-nm resolution and representing 1 voxel at 8 bytes, approximately 8 petabyte of data will be produced. Development of smart-sampling technologies that use computational models to inform and optimize sampling strategies will become relevant. Development of this workflow, from sampling at the APS-U to model development, will be an exciting opportunity.


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