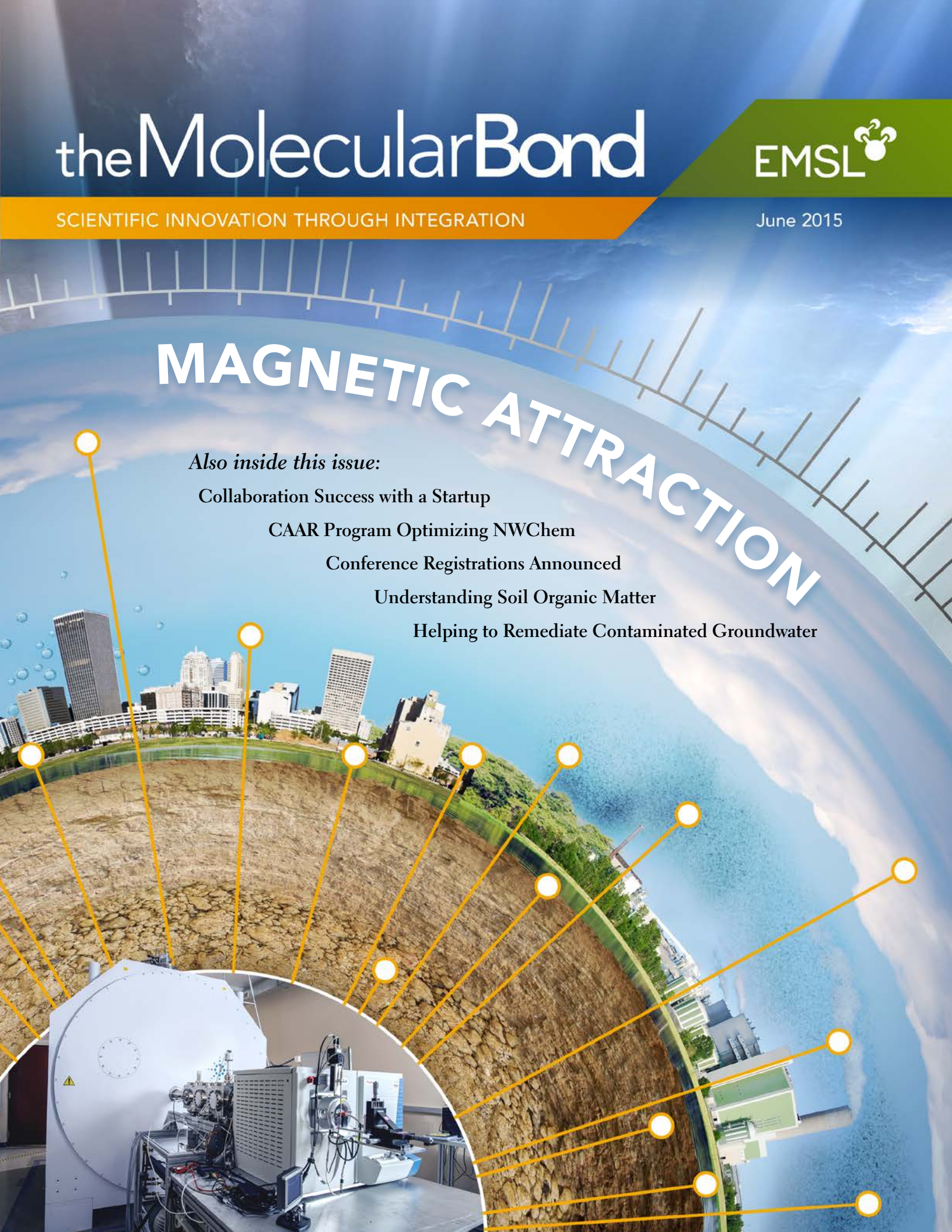
the Molecular Bond

EMSL

SCIENTIFIC INNOVATION THROUGH INTEGRATION

June 2015



MAGNETIC ATTRACTION

Also inside this issue:

Collaboration Success with a Startup

CAAR Program Optimizing NWChem

Conference Registrations Announced

Understanding Soil Organic Matter

Helping to Remediate Contaminated Groundwater

About the Cover

Scientists desiring a molecular-level understanding of complex systems to expand their studies of climate effects on carbon cycling in the soil, carbon transformations in the atmosphere and biofuel production processes will soon have access to EMSL's new 21 Tesla Fourier transform ion cyclotron resonance mass spectrometer. The 21T FTICR mass spectrometer's high field strength allows for higher resolution and higher mass accuracy measurements that ensure identification of molecular constituents in complex samples and materials.

About *The Molecular Bond*

The Molecular Bond is EMSL's bimonthly newsletter for users, potential users and other interested individuals. EMSL Communications oversees the production of *The Molecular Bond*.

EMSL, the Environmental Molecular Sciences Laboratory, is a national scientific user facility sponsored by the Department of Energy's Office of Biological and Environmental Research in the Office of Science. Located at Pacific Northwest National Laboratory in Richland, Wash., EMSL offers an open, collaborative environment for scientific discovery to researchers around the world.

Its integrated computational and experimental resources enable researchers to realize important scientific insights and create new technologies. More information about EMSL is available at: www.emsl.pnnl.gov

To request additional copies or to subscribe, contact: EMSL Communications at email: emslcom@pnnl.gov

EMSL

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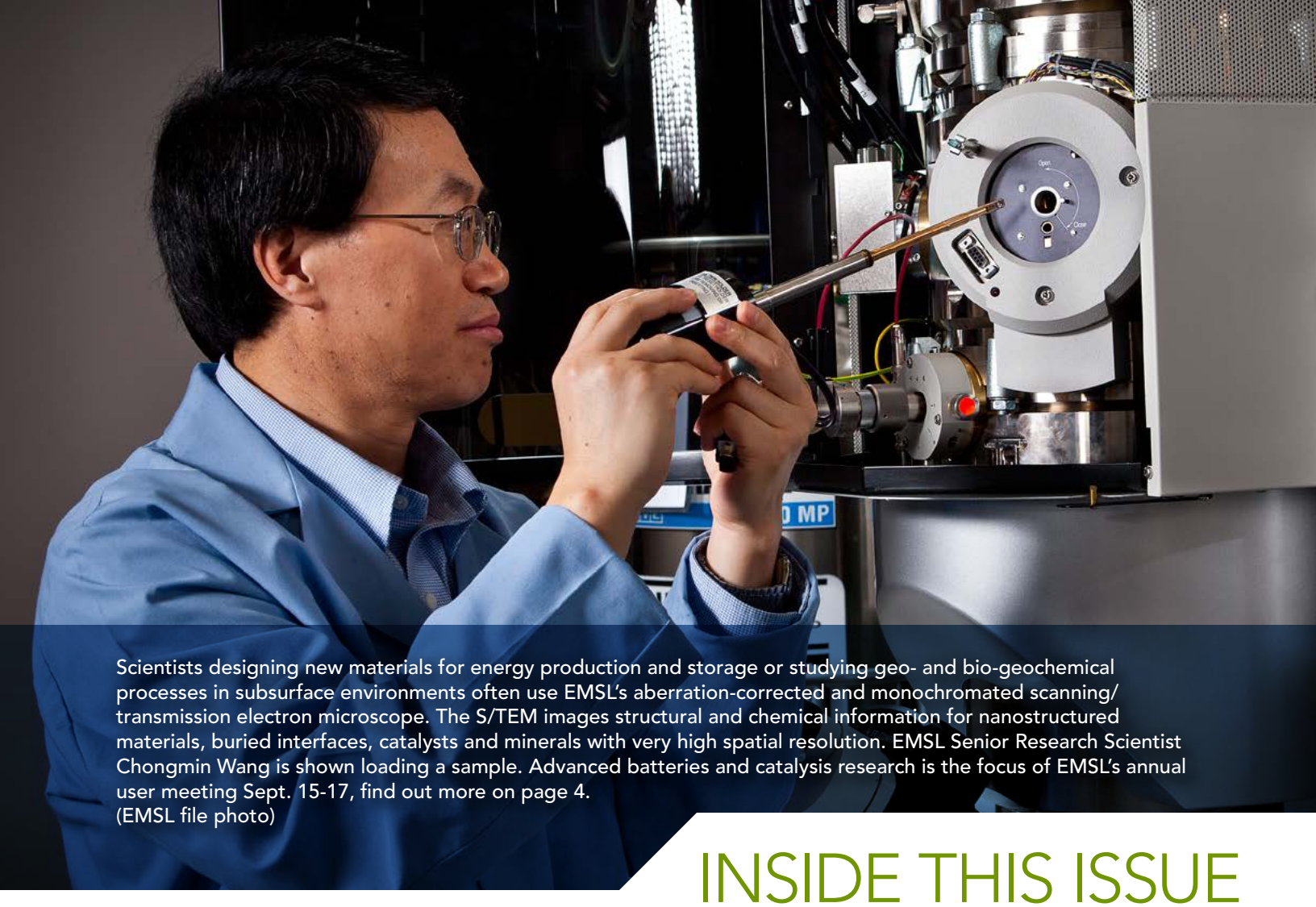

Scientists designing new materials for energy production and storage or studying geo- and bio-geochemical processes in subsurface environments often use EMSL's aberration-corrected and monochromated scanning/transmission electron microscope. The S/TEM images structural and chemical information for nanostructured materials, buried interfaces, catalysts and minerals with very high spatial resolution. EMSL Senior Research Scientist Chongmin Wang is shown loading a sample. Advanced batteries and catalysis research is the focus of EMSL's annual user meeting Sept. 15-17, find out more on page 4.
(EMSL file photo)

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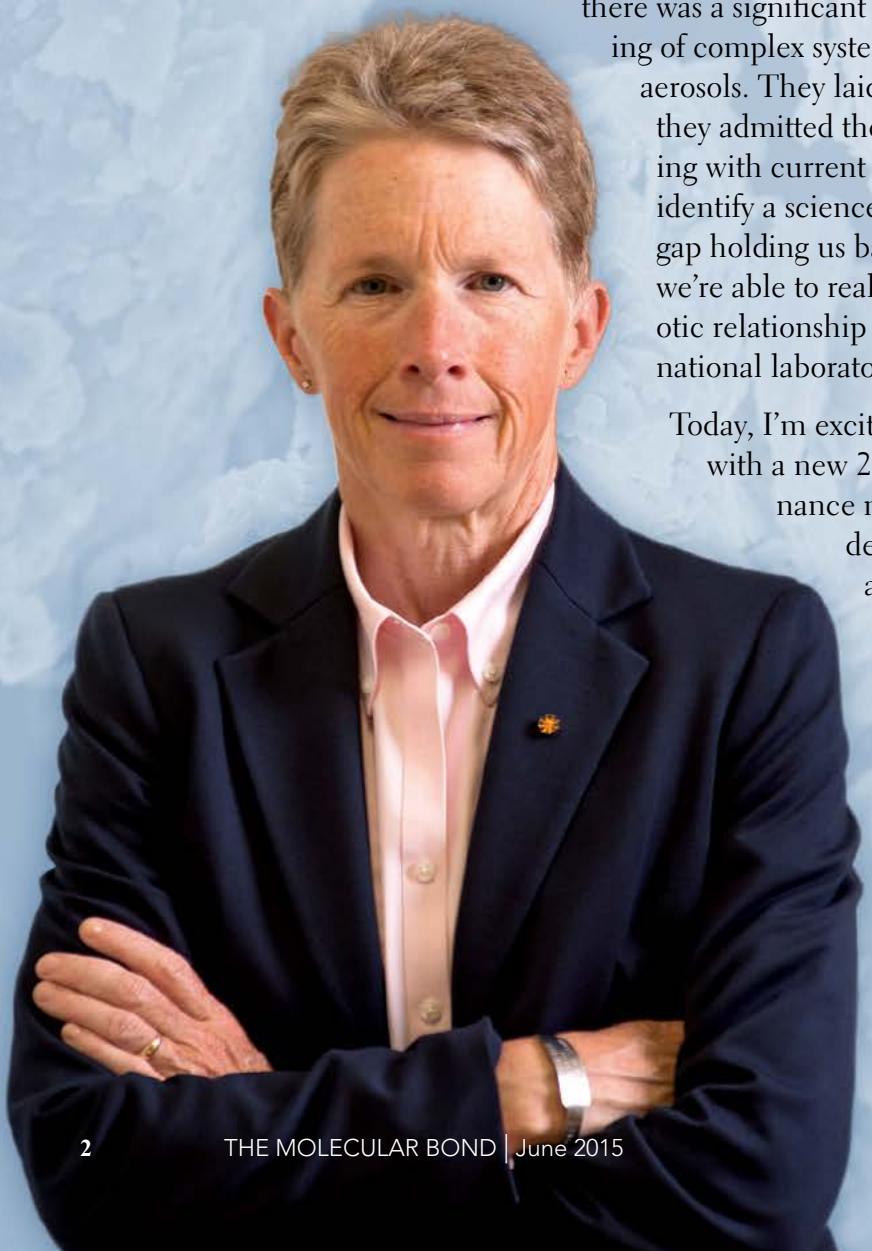
EMSL

FROM THE DIRECTOR

Allison Campbell

The Department of Energy national labs and user facilities play a unique role in the scientific community, serving as an enduring system focused on national needs. At EMSL, we regularly bring scientists together from academia, industry and government laboratories to discuss the current and emerging national needs and how we can address them. Nearly 10 years ago, EMSL brought together 100 scientists for such a workshop. The group determined there was a significant need to capture a molecular-level understanding of complex systems, from microbial communities to organic aerosols. They laid out a strong set of science challenges; yet they admitted there was no way of achieving this understanding with current technologies. As often happens when we identify a science challenge, we find there is a technological gap holding us back. As we build these new technologies, we're able to realize those scientific advances. This is a symbiotic relationship I see arise every day in the user facilities and national laboratories.



Today, I'm excited to share we have filled that technology gap with a new 21 Tesla Fourier transform ion cyclotron resonance mass spectrometer. We can achieve unprecedented mass resolution and accuracy that will allow us to characterize the molecular species in complex systems. In this issue of *The Molecular Bond*, you'll read about the first scientific studies that will be conducted on the system, starting this summer.

I'm proud of the EMSL scientists and engineers who designed, developed and now deployed a system we can use to realize the discoveries only imagined 10 years ago.

—*Allison*

Scientists researching ways to more efficiently produce biofuels and other bioproducts are using metabolomics, or the study of the fundamental metabolic workings of a cell in a changing environment. This petri dish contains colonies of *Yarrowia lipolytica*, an oil-producing yeast. Each colony is a different mutant strain of the fungus. Scientists use several EMSL capabilities to study the cellular metabolisms of these fungi. Find out more about metabolomics and other omics at the upcoming microbiomes conference co-sponsored by EMSL on page 4. (EMSL file photo)

NEWS & KUDOS

David Stahl

Stahl Named EMSL Chief Scientist

David Stahl has been selected as chief scientist for the EMSL user program. An environmental microbiologist at the University of Washington in Seattle, Stahl will hold a dual appointment and maintain his teaching and research position at the university.

As chief scientist, Stahl will lead efforts to build user communities in EMSL's Science Theme research areas, reach out and raise awareness of EMSL to the Department of Energy's Office of Biological and Environmental Research principal investigator community, and provide additional biology expertise at EMSL and Pacific Northwest National Laboratory.

Stahl has an international reputation in studying the application of molecular microbial ecology to environmental engineering. He's received funding from DOE BER and is a member of BER's Advisory Committee. In 2012, he was elected to the National Academy of Engineering.

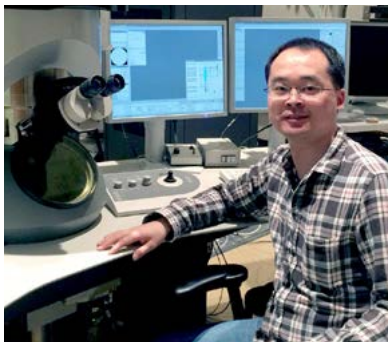
Before joining the University of Washington in 2000, Stahl held positions at the University of Illinois at Urbana-Champaign and at Northwestern University. He received his bachelor's degree in microbiology from the University of Washington and his master's and PhD in microbiology from the University of Illinois.

Yan Receives MSA Post-Doctoral Researcher Award

Congratulations to Pengfei Yan, an EMSL postdoctoral researcher, for receiving a 2015 M&M Post-Doctoral Researcher Award from the Microscopy Society of America. He earned the award for his paper entitled "Charge-Discharge Cycling Induced Structural and Chemical Evolution of Li_2MnO_3 Cathode for Li-ion Batteries."

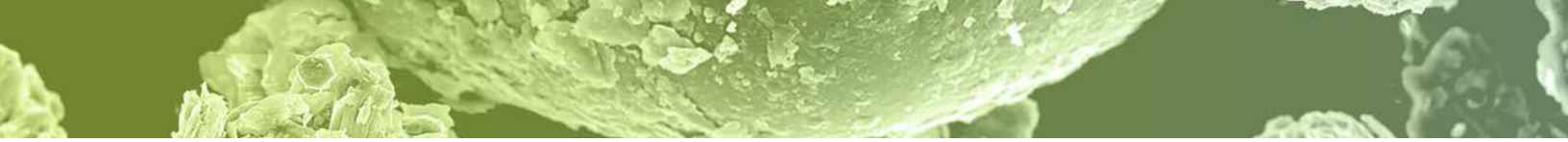
The application pool for 2015 M&M Meeting Awards was extremely competitive. More than 300 individuals submitted proceedings papers and only a few were selected for awards.

The 2015 award includes complimentary registration to the Microscopy & Microanalysis 2015 Annual Meeting Aug. 2-6 in Portland, Ore., and $1,000 to help cover travel expenses. Yan will be honored at the opening plenary session.



Pengfei Yan

Yan's research was funded by the Department of Energy's Office of Energy Efficiency and Renewable Energy. The work was performed at EMSL.



NWCHEM
HIGH-PERFORMANCE COMPUTATIONAL CHEMISTRY SOFTWARE

NWChem Named a CAAR Project

The Oak Ridge Leadership Computing Facility selected EMSL's NWChem computational chemistry software to be one of 13 partnership projects in its Center for Accelerated Application Readiness, or CAAR, program.

The CAAR program is a collaborative effort of application development teams from national laboratories and universities, and staff from the Oak Ridge Leadership Computing Facility Scientific Computing group. The program is focused on redesigning, porting and optimizing application codes for the hybrid central processing unit-graphics processing unit, or CPU–GPU, architecture that will be used in Oak Ridge National Laboratory's Summit next-generation supercomputer.

"It's a great honor NWChem is among the awarded projects," said Karol Kowalski, acting capability lead for EMSL's Molecular Science Computing Performance Software and a co-principal investigator for the project. "Being a part of the CAAR program will enable us to get NWChem ready for the hybrid architectures based on the CPU-GPU technology."

The other co-principal investigators on the NWChem development team include Edoardo Apra, at EMSL; Sriram Krishnamoorthy and Bruce Palmer, both at Pacific Northwest National Laboratory; and Julia Rice, at IBM Almaden Research Center.

The goal of the NWChem project is two-fold: to develop and disseminate an open-source database of accurate glycan conformational energies, and to define an improved force field for studying glycans of relevance to energy applications. Plant glycans such as cellulose, starch and chitin offer the promise for creating materials with new physical and chemical properties. The team's results promise to allow for the computational investigation of new carbohydrate-based materials from bulk polymers to nanoparticles with useful functionality, particularly in the areas of medicine, energy generation and advanced materials.

Developed at EMSL, NWChem is one of the Department of Energy's premier computational chemistry tools. Highly scalable and portable, NWChem runs on a wide range of platforms from desktop computers to supercomputers. The software became open source at the end of 2010 and has been downloaded more than 60,000 times. NWChem can be downloaded at http://www.nwchem-sw.org/index.php/Main_Page.

EMSL Sponsors Three Science Conferences

Registration is open for three EMSL-sponsored science meetings, one in July and two in September. The conferences include:

EMBEER Symposium – July 29-30

The "Electron Microscopy for Biological, Environmental, and Energy Research (EMBEER) Symposium," organized by the Chemical Imaging Initiative and EMSL at the Pacific Northwest National Laboratory, will be held July 29-30 at EMSL in Richland, Wash.

EMBEER will focus on recent developments in biological and environmental research, energy storage and conversion materials, and materials properties and processes utilizing microscopy, and recent developments in instrumentation and methods.

More information is available on the EMBEER website: http://1.usa.gov/1I9LIQo.

Microbiomes Conference – Sept 14-16

The "Multi-omics for Microbiomes Conference" will be held Sept. 14-16 at the Three Rivers Convention Center in Kennewick, Wash. Pacific Northwest National Laboratory and EMSL are co-sponsors.

This conference will highlight the latest developments and applications of molecular "omics," imaging and computational technologies to decipher functional processes and interactions carried out by complex microbial communities, including those residing in soil, the ocean and the human gut.

Visit the conference website for more information or to register: http://1.usa.gov/1LrN8uL.

EMSL's Annual User Meeting – Sept. 15-17

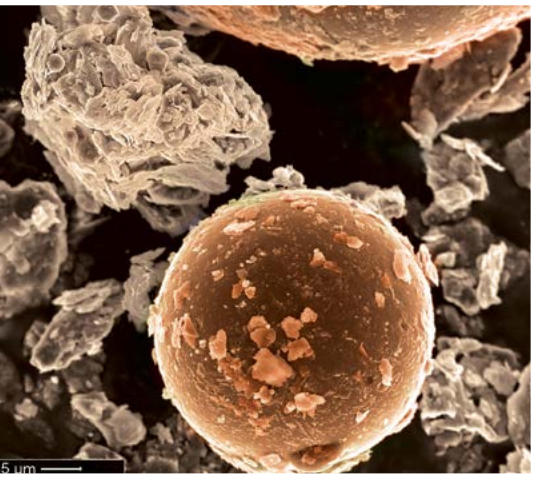
"EMSL Integration 2015: Energy Materials and Processes for Advanced Batteries and Catalysis" will be held Sept. 15-17 at EMSL.

This year's annual meeting focuses on advanced batteries and catalysis research at the molecular level. Researchers from around the world and from academia, industry and government research labs are encouraged to attend. The meeting is open to EMSL users and non-users.

More information is available on the meeting website: http://1.usa.gov/1BMTtPr.

A *Fruitful* Collaboration

EMSL expertise and capabilities propel a startup to success



This image is one in a series of scanning electron microscope images taken by Bruce Arey for GeoTree under the Technology Assistance Program.

Used to advance the scientific understanding of various phenomena, including interfacial behavior, biological transformations and geochemical processes is the helium ion microscope in EMSL's Quiet Wing. Bruce Arey, EMSL technologist, loads the sample holder into the sample exchange chamber. The helium ion microscope is another instrument Avery uses in addition to the scanning electron microscope. (EMSL file photo)

The image is of a sphere, suspended in a sea of black and grey, eerily perfect against its irregularly-shaped surroundings. To the untrained eye, it looks like it belongs in an asteroid field, but this sphere and all of the particles surrounding it are only visible at the nanometer scale.

The sphere is an aluminosilicate, one component of a cement-like geopolymer currently used by global manufacturing company Milliken to rehabilitate corroding sewer pipes. This geopolymer is a green technology that combines recycled industrial waste and mineral polymers to make a strong, long-lived solution for rehabilitating infrastructure.

Although the technology is widely accepted now, in 2010, when it was still in development, it was largely unproven. That was when Ben Cook, a consultant working with SGA Advanced Solutions and GeoTree (the latter being the company that sold the technology to Milliken), reached out to the Pacific Northwest National Laboratory. He applied for help through PNNL's Technology Assistance Program. It was through this program that Cook was able to access advanced imaging capabilities at EMSL, the Environmental Molecular Sciences Laboratory at PNNL, to visualize the components of the geopolymer.

The relationship with EMSL proved fruitful for Cook.

Bruce Arey is the technologist at EMSL who worked with Cook's samples. Arey has been with PNNL for 32 years, 10 of which have been with EMSL, making him exactly the kind of expert Cook needed to see the geopolymer's structure on the nanoscale. Arey, who specializes in microscopy, used an advanced scanning electron microscope (more specifically, the FEI Helios 600 Dual Beam FIB-SEM) to visualize the smallest of the spheres, which are less than 10 nanometers across or 7,500 times smaller than the width of a human hair.

One of the great strengths of SEM is that it is a relatively quick process in the world of microscopy; with imaging taking one to four hours depending on the level of detail a researcher wants to capture. Sample preparation is also straightforward, and within an hour of receiving a sample it can be in the microscope's sample holder. The sample prep for the geopolymer was routine, but for Arey the challenge was to get the best image.

"There's a certain recipe," to microscopy, said Arey and "each sample has to be tweaked to a different imaging recipe."

The images generated by Arey confirmed the science behind GeoTree's polymerization process and captured the imagination of partners who were able to take their product into the commercial market.

"When you're talking about nanotechnology, people in the general population just don't have a good feel for what that actually means," Cook said. This means that startup companies are often telling a story about the potential of an unproven technology to people who have heard a thousand similar stories. "Once they started seeing [the images] they believed the story, and that's a powerful thing."

A successful collaboration between PNNL and companies in TAP is not unique, although hearing a success story after so many years is a bit unusual. "We usually get updates at the end of the year," said Gary Spanner, who heads up TAP.

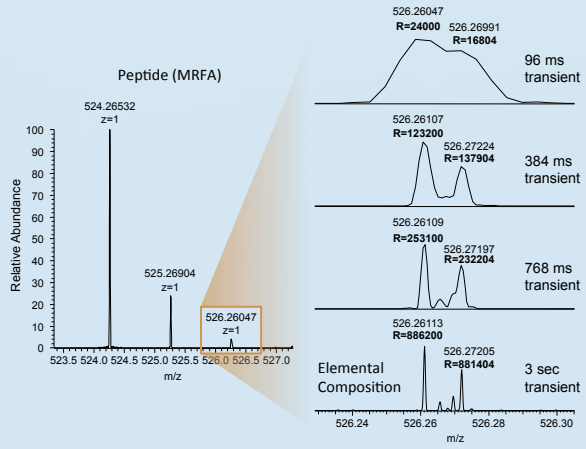
Cook said that his contact with Spanner was a nod to the long-lasting positive impact of EMSL's expertise and capabilities. Although GeoTree was sold to Milliken in 2012, Cook may want to call upon TAP resources as a consultant for his business, Green Building Systems, in the near future. "My questions are too sophisticated and too systems oriented," for any one local company in Seattle.

Apart from his appreciation of the SEM's ability to display chemical structure, Cook still is in awe of the images generated in 2010. Although he and his collaborators knew roughly what to expect in the structure of the geopolymer, they were caught off guard by one aspect of the material at the nanoscale.

"We didn't expect it to be so beautiful," he said, "It's really lovely to look at." ■

Amy Boaro is a post-doctoral researcher at Pacific Northwest National Laboratory.

On the forefront of understanding molecular-level science in the natural environment



Designed to characterize complex environmental and biological materials, EMSL's new 21 Tesla Fourier-Transform Ion Cyclotron Resonance mass spectrometer produced this first spectra.

The samples shipped to the mass spectrometry lab at EMSL bear labels from all over the world, from bacteria grown in U.S. laboratories to aerosol particles trapped in the Amazon basin. Although the contents vary greatly, they share two things: a complex molecular makeup and a place in the starting lineup of samples waiting for a spin in the new 21 Tesla Fourier transform ion cyclotron resonance, or 21T FTICR, mass spectrometer that was designed and built for research at EMSL.

Ten years ago, the studies of these samples wouldn't have been possible. That's when a group of 100 scientists from 40 institutions visited EMSL to discuss advancements in measurement science that would be needed in the following decades. One of the priorities identified was development of a 21T FTICR mass spectrometer. Subsequent meetings around the world confirmed the need for this kind of capability to enable "breakthrough discoveries in the understanding of complex systems," according to a 2008 workshop report on this subject. Scientists saw the potential for addressing what they called "analytical grand challenges" such as studying biochemical pathways, cellular communication and microbial communities at the molecular level.

"We recognized the opportunity to advance our understanding of complex systems across a broad spectrum of science, from characterizing organic aerosols to understanding microbial community dynamics to achieving high-resolution chemical imaging," says David Koppenaal, EMSL's chief

technology officer and one of the leaders of a 2008 workshop focused on identifying the science challenges for such a next-generation mass spectrometer.

These scientific questions drove development of the new 21T FTICR mass spectrometer, which is now available to scientists around the world for their research.

"We're poised to realize the possibilities of scientific studies we only imagined 10 years ago," says Koppenaal.

Now, scientists will be able to obtain molecular-level information to improve biofuel production efforts as well as our understanding of climate effects on carbon cycling in the soil and carbon transformations in the atmosphere.

"Scientists will be able to pinpoint the molecular makeup of complex chemical systems with near unequivocal certainty," says Koppenaal. "This instrument provides data with mass accuracy to within six decimal points. Conventional instrumentation today doesn't have that kind of resolution or mass accuracy."

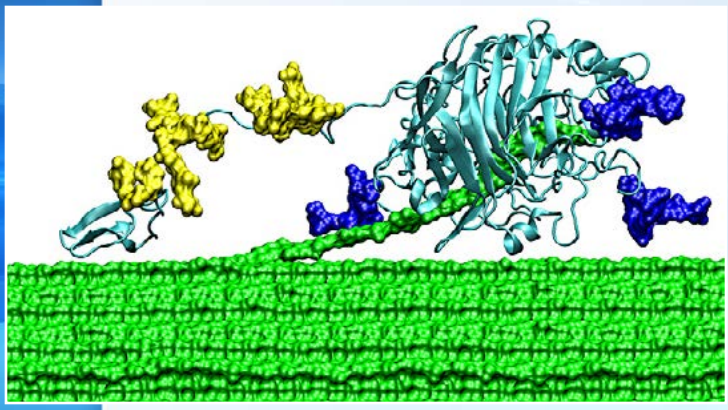
The system is expected to be fully operational by October. A Special Science Call went out last year for studies that incorporated EMSL's recently developed technical resources, including this new mass spectrometer. Researchers are keen to get results with the six inaugural studies selected through the call.

Center of Attraction

Lili Paša-Tolić, EMSL's lead scientist for mass spectrometry, said the "21T" has significantly ramped-up performance in terms of mass measurement accuracy and mass resolving capabilities. "At higher magnetic fields the accuracy, mass resolving power, sensitivity and dynamic range improve either linearly or quadratically."

Through a top down proteomics approach advanced by Paša-Tolić, the enhanced resolving power of the 21T will allow proteins up to 150 kDa (daltons) to be parsed in their entirety. With current resolution, researchers typically need to first pick apart big proteins and analyze them bit by bit – often missing key information in the process.

This top-down method is of particular interest to Jonathan Walton, a plant biologist at the Michigan State University-Department of Energy Plant Research Laboratory and member of the DOE Great Lakes Bioenergy Research Center, which is funding his study. His work with fungi that break down plant walls led him to study cellobiohydrolases, or CBH1. This group of enzymes is adept at degrading the tough cell walls of lignocellulose, an abundant biomass from which we could produce biofuels if the process operated more efficiently and economically. (Continued on page 8)



Jonathan Walton is researching the use of enzymes to degrade biomass for biofuels. One of the most dramatic "artist's conception" images of Walton's research is from the National Renewable Energy Laboratory. It shows a graphic of the enzyme his team is working with at EMSL, called cellobiohydrolase 1 from *T. reesei*. (Image provided by Jonathan Walton, Michigan State University-DOE Plant Research Laboratory)

Developed to advance carbon cycling, microbial biology and atmospheric aerosol characterization studies, EMSL's 21 Tesla Fourier-Transform Ion Cyclotron Resonance, or 21T FTICR, mass spectrometer will enable a variety of biomolecular and other natural organic compound studies. The 21T FTICR mass spectrometer's high field strength allows for higher resolution and higher mass accuracy measurements that ensure identification of molecular constituents in complex samples and materials. (EMSL file photo)

"These enzymes are particularly tricky to work with," says Walton. To express the cellobiohydrolases, he inserts their genome sequence into yeast that is more tractable than the enzyme's native fungal source. But Walton found that yeast alter CBH enzymes by adding sugars to them after they're biosynthesized. This "post-translational glycosylation" can change enzyme function.

With the 21T, Walton hopes to see precisely what's happening, and then correlate the glycosylation modifications to key enzyme properties such as thermostability and activity.

"From that we'll be able to design better enzymes," he says.

Although he's a first-time EMSL user, Walton plans to follow this work with more traditional "bottom up" proteomics and glycomics using the facility's mass spectrometry and nuclear magnetic resonance spectrometers to understand in greater detail how glycosylation occurs. And he's already submitted another proposal to look at another group of cellulases that degrade lignin.

"A lot comes down to understanding what happens when you express proteins from one organism into another," Walton says. "Strange things happen, and the new mass spectrometer at EMSL is the first step to understanding what's going on."

Another researcher working on the lignocellulose puzzle is Blake Simmons, the chief science and technology officer and vice president of the Deconstruction Division at the DOE Joint BioEnergy Institute, based in the San Francisco Bay area.

Simmons refers to lignin, one of the primary components in lignocellulosic biomass, as "the Gordian knot" of biofuels. To start cutting that knot, he hopes the 21T will help him map the metabolic pathways of organisms that break down lignin. Then he'll chart the way metabolism changes with varied environments, growth substrates and end products.

"Once we build that map, we can translate that knowledge into biorefining, and then deploy it to convert not only sugars to fuels, but also from lignin to fuels and chemicals," he says. His studies are funded by DOE's Office of Biological and Environmental Research.

Right now, Simmons and his team are working with *Klebsiella* and *Enterobacter*, two organisms he calls the "lab rats" of the bacterial world. During soil studies in the cloud forest of Puerto Rico, scientists discovered that both of these bacteria can degrade lignin, but there are differences in how the two use carbon and sugar resources. For his 21T samples, Simmons' team tracked the bacteria as they were grown in the lab and fed specific substrates, one at a time. At pre-set intervals, his team harvested the cells for 21T analysis of the cell fractions – the lipids, proteins, enzymes, metabolites and so on – to identify the molecules present during metabolic changes.

"We're trying to create an electrical circuit equivalent of metabolism in these organisms: finding out where those

fluxes are turned on, turned off, held constant," says Simmons. "Once we get that electrical wiring diagram of metabolism then we can start doing what we do best: fiddling with it, engineering it, and optimizing it for our specific purpose."

Carbon Cycling

Another set of samples headed for the 21T also started with a study in Puerto Rico's Luquillo Experimental Forest. There, microbiologist Jennifer Pett-Ridge, the lead scientist of Lawrence Livermore National Laboratory's Genomic Science Biofuels Scientific Focus Area, is using soil samples from various mountain elevations to study how climate change will affect the conversion of soil carbon into highly mobile dissolved organic matter.

Tropical forest soils are thought to store a huge amount of carbon, but changes in global weather patterns may make these terrestrial carbon dioxide sinks less reliable, notes Pett-Ridge. The tropics are expected to experience longer droughts punctuated by intensive rainfall, so they may become less consistently wet. Under those climate conditions, the carbon adsorbed onto soil mineral surfaces is dramatically affected when the soil goes from oxygenated to an anaerobic state.

"Carbon goes from tightly bound to mineral surfaces, to more amorphous loose association; this will affect the chemistry and quantity of dissolved organic carbon," says Pett-Ridge. Her studies are supported by a DOE Early Career Research Program award through the Office of Biological and Environmental Research.

With soil samples from forest sites that range from wet tropical to the colder cloud forest, Pett-Ridge hopes the 21T will help her understand how changes in soil oxygen and redox – "whether there are many, many changes or slow redox every week to two weeks"– affect the total amount of carbon released into the dissolved pool.

"I also want to know about the quality – the molecular composition – of the dissolved pool," says Pett-Ridge. "That's where the new mass spectrometer comes in."

Right now, she can detect elements in soil samples at the single element/isotope level with the NanoSIMS (nanoscale secondary ion mass spectrometry) at EMSL. If the 21T performs as anticipated, Pett-Ridge could identify molecular-level components in her samples, such as lipids, proteins and peptides. She and collaborator Aaron Thompson, a biogeochemist at The University of Georgia, started a pilot study on soil samples with the 15T mass spectrometer in use at EMSL now. Next, if the new instrument proves to be the right tool for her research questions, she hopes to expand her work with samples from multiple lab incubations of tropical soils.

"The long-range goal is to say something more globally about tropical soils and how their microbial communities and their carbon chemistry are responding to climate change," says Pett-Ridge. (Continued on page 10)

Jennifer Pett-Ridge is studying how climate change affects soil carbon in the Puerto Rican forest. The research site (insert photo) shows soil samples being gathered. (Image provided by Jennifer Pett-Ridge, Lawrence Livermore National Laboratory)

Jennifer Pett-Ridge's research project looking at climate change and soil carbon uses soil samples from Puerto Rico. Next to the soil samples is a soil gas chamber that will be buried to test if the soil goes anaerobic frequently. (Image provided by Jennifer Pett-Ridge, Lawrence Livermore National Laboratory)

Aerosol Approach

The 21T will also play a role in better understanding carbon chemistry changes in the atmosphere. Soon, the instrument should produce data sets with details on the composition of samples of aerosol particles collected by EMSL researcher Alexander Laskin from pristine Amazon areas, and from areas directly affected by emissions from Manaus, a city in Brazil. A team of aerosol scientists, including Laskin, Sergey Nizkorodov, a researcher at the University of California at Irvine, and their co-workers are anxiously awaiting the launch of the 21T instrument.

"The goal is to learn how natural aerosols are affected by emissions from a major city," says Nizkorodov. His studies are funded by the National Science Foundation and National Oceanic and Atmospheric Administration.

In areas of the Amazon with little manmade pollution, biogenic emissions get oxidized and become complex aerosol mixtures made primarily of carbon, oxygen, hydrogen and some nitrogen. With current tools, scientists have been able to sort through the composition of these aerosols. However, near a city, anthropogenic effects add more nitrogen, sulfur, silicon and other elements into the atmospheric mix.

"Under those conditions, there are so many possible chemicals that, right now, no existing mass spectrometer can resolve all of them," says Nizkorodov. "This 21T has the capability of unambiguously resolving this very complicated mixture and teaching us something new about the atmospheric chemistry of aerosols."

With new information from the 21T, Nizkorodov hopes to learn whether mixing emissions from the city with those from the forest generates unique compounds. If there are new compounds, the next step will be determining what special properties they have: Do they absorb white light? Do they have toxic consequences?

Nizkorodov also plans to run model aerosol samples from a smog chamber. He's specifically interested in the effect of humidity on aerosol samples because most prior experiments were performed in dry chambers. With the information currently available, it's difficult to predict whether the humid environment in the Amazon will alter the predicted chemical transformations of aerosols. So, he plans targeted experiments in which he'll introduce nitric oxide, sulfur dioxide and dry air into a chamber loaded with a mixture of volatile organic compounds; then he'll use exactly the same mix in humid air and see whether this creates any difference at the molecular level, and on the properties of the aerosols.

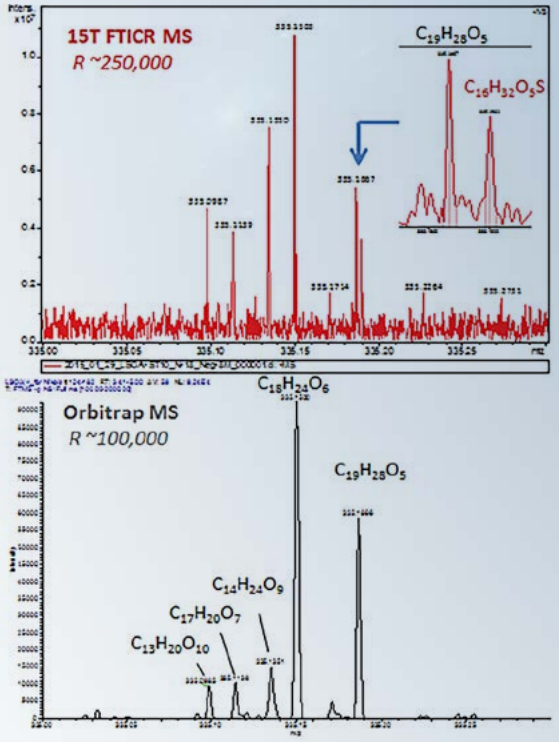
> "We're on the forefront of better understanding molecular-level science in the natural environment."

"The 21T will definitely take our studies to the next level," says Nizkorodov. "We'll be able to understand the aerosol chemistry so much better. I'm very excited to be one of the first users."

The new mass spectrometer is the single biggest investment that EMSL has made in an experimental instrument, notes Koppenaal.

"This instrument will really open the door to a whole new area of scientific inquiry," says Koppenaal. ■

Elizabeth Devitt is a science journalist and freelance writer.



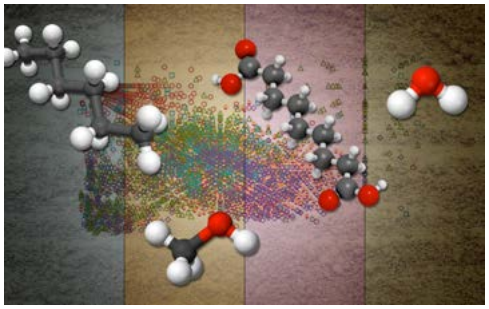
The project goal of Sergey Nizkorodov's and Alexander Laskin's research is to examine the effects of biogenic-anthropogenic interactions on the chemical composition of organic aerosol from the on-going GoAmazon field study. Shown are spectra of organic aerosol using EMSL's 15T FTICR mass spectrometer and Orbitrap mass spectrometer. The higher resolving power of the former makes it possible to resolve additional compounds. (Image provided by Alexander Laskin, EMSL)

SCIENCE HIGHLIGHTS

Soil Composition

First-time study sheds light on soil processes

Soils contain a vast amount of carbon and are responsible for most greenhouse gas emissions. However, current models do not predict effects of climate on specific soil carbon processes and vice versa, in part due to the lack of knowledge about the molecular composition of soil organic matter.



Researchers coupled different solvents with ultra-high resolution mass spectrometry to identify diverse classes of compounds from soil organic matter in a range of soil types. The researchers found different solvents extracted different types of compounds from soils, significantly expanding the ability to sensitively detect and identify the vast suite of diverse organic molecules that compose soil organic matter. The study could lead to more accurate predictions of the impacts of future climate scenarios modeled at novel regional and global scales.

Funding: This research was supported by the Department of Energy's Office of Biological and Environmental Research, Climate and Environmental Science Division, Terrestrial Ecosystem Science Program.

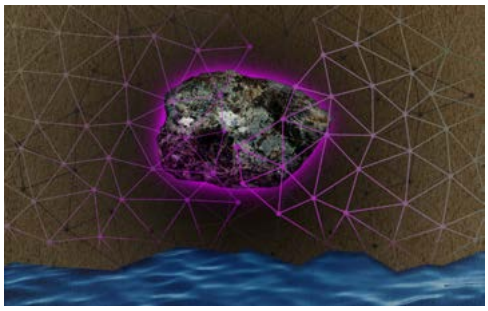
Publication: Tfaily M, RK Chu, N Tolić, KM Roscioli, CR Anderton, L Paša-Tolić, EW Robinson, and NJ Hess. 2015. "Advanced solvent based methods for molecular characterization of soil organic matter by high resolution mass spectrometry." *Analytical Chemistry*. doi:10.1021/acs.analchem.5b00116.

Participating Organizations: EMSL and Idaho National Laboratory.

New Hypothesis for Environmental Restoration

Carbonate minerals could remediate contaminated groundwater

Scientists found the incorporation of neptunium (V) (NpO2+, neptunyl) and uranium (VI) (UO22+, uranyl) in a variety of mineral structures were quite different despite similar molecular geometries. Specifically, dramatically different incorporation levels were found for U(VI) and Np(V) in carbonate minerals grown from similar aqueous solutions in ambient conditions.

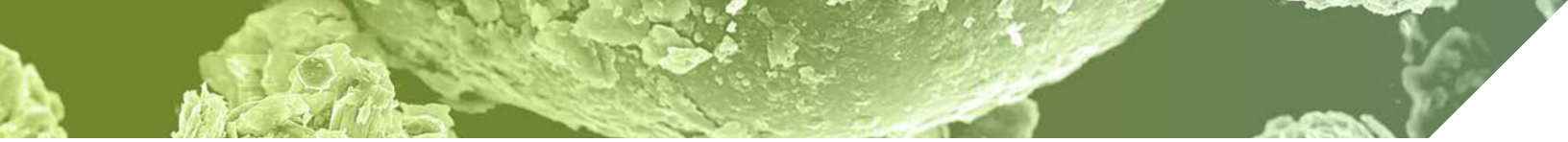


Scientific results clearly demonstrate crystal-chemical behaviors of Np(V) and U(VI) actinyl ions are sufficiently different than predicted. The new experimental results indicate that low-temperature mineral structures in the subsurface are highly selective for the incorporation of Np(V) or U(VI) actinyl ions, a mechanism that may limit their mobility and utilized as part of a groundwater remediation strategy.

Funding: This research was supported by Department of Energy's Office of Biological and Environmental Research, Subsurface Geochemical Research Program.

Publication: Balboni E, JM Morrison, Z Wang, MH Engelhard and PC Burns. 2015. "Incorporation of Np(V) and U(VI) in Carbonate and Sulfate Minerals Crystallized from Aqueous Solution." *Geochimica et Cosmochimica Acta* 151:133-149. DOI:10.1016/j.gca.2014.10.027.

Participating Organizations: University of Notre Dame, Pacific Northwest National Laboratory and EMSL.



Archael Tree of Life

First complete genomes reveal role in global carbon cycle

Archaea, a domain of single-celled microorganisms, represent a significant fraction of the earth's biodiversity, yet they remain much less understood than bacteria. One reason for this lack of knowledge is relatively poor genome sampling, which has limited accuracy of the Archaeal phylogenetic tree. In a recent study, researchers approximately doubled the genomic diversity sampled from this domain and reconstructed the first complete genomes for Archaea using cultivation-independent methods resulting in an extensive revision of the Archaeal tree of life.

The study revealed Archaea in the terrestrial subsurface contribute primarily to carbon and hydrogen cycling, suggesting these organisms may be involved in processing the sizeable reservoir of buried organic carbon. This finding can be immediately implemented within genome-resolved ecosystem models to more accurately reflect the key role played by Archaea in the global carbon cycle.

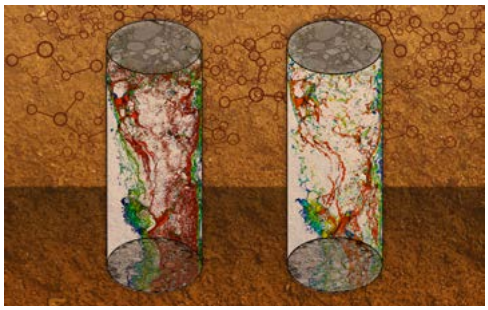
Funding: This research was supported by Department of Energy's Office of Biological and Environmental Research.

Publication: Castelle CJ, KC Wrighton, BC Thomas, LA Hug, CT Brown, MJ Wilkins, KR Frischkorn, SG Tringe, A Singh, LM Markillie, RC Taylor, KH Williams and JF Banfield. 2015. "Genomic Expansion of Domain Archaea Highlights Roles for Organisms from New Phyla in Anaerobic Carbon Cycling." *Current Biology*. DOI:10.1016/j.cub.2015.01.014.

Participating Organizations: University of California at Berkeley, The Ohio State University, Columbia University, Department of Energy Joint Genome Institute, EMSL, Pacific Northwest National Laboratory and Lawrence Berkeley National Laboratory.

Multiscale Simulations

Expanding the impact of pore-scale simulations



Pore-scale models are useful for studying relationships between fundamental processes at the scale of tens to hundreds of microns – the size of solid soil grains and pore spaces – and phenomena at larger scales that are relevant to real-world applications. However, conventional simulations of the flow of water and chemicals through soils at large scales typically neglect details of pore geometry due to computational demands associated with characterizing large 3-D volumes with pore-scale spatial resolution. This study introduces a method for overcoming this challenge and presents the first simulations of pore-scale flow and transport over a large, decimeter-scale volume.

Understanding how water and chemicals flow in soils is important for many practical problems, such as assessing risk of groundwater contamination, developing effective ways to clean up contaminated soils and aquifers, and designing subsurface repositories for waste materials. By bridging the gap between small and large spatial scales, the simulation method could shed light on the effect pore-scale processes have on these field-scale phenomena, paving the way for more accurate assessments of risk of groundwater contamination and development of more effective remediation strategies.

Funding: This research was supported by Department of Energy's Office of Biological and Environmental Research as part of its Subsurface Biogeochemistry Research, or SBR, Program. This contribution originates from the SBR Scientific Focus Area at PNNL.

Publication: Scheibe TD, WA Perkins, MC Richmond, MI McKinley, PDJ Romero-Gomez, M Oostrom, TW Wietsma, JA Serkowski and JM Zachara. 2015. "Pore-scale and Multiscale Numerical Simulation of Flow and Transport in a Laboratory-scale Column." *Water Resources Research*. DOI:10.1002/2014WR015959.

Participating Organizations: Pacific Northwest National Laboratory, EMSL, National Energy Research Supercomputing Center and University of Texas at Austin.

About this Art

The *Arabidopsis* plant's root structure, shown in a cross-section view, is helping to improve understanding of the rhizosphere, or plant root zone. Researchers believe learning more about the rhizosphere will help clarify environmental processes, and perhaps give rise to future climate and environmental solutions. Toward this goal, a rhizosphere research campaign was initiated at EMSL. The effort has focused on the *Arabidopsis* plant's management of carbon nutrients and subsequent impacts on the diversity of the rhizosphere. The campaign includes scientists from EMSL, Pacific Northwest National Laboratory, Department of Energy's Joint Genome Institute, Brookhaven National Laboratory, the University of Minnesota and the University of Missouri. The work is funded by DOE's Office of Biological and Environmental Research. The image was captured with the Helios Nanolab dual-beam focused ion beam/scanning electron microscope at EMSL.

Team members

Ljiljana Paša-Tolić, Alice Dohnalkova, Galya Orr, David Hoyt, Heather Brewer, Meng Markillie and Angela Norbeck, all of EMSL; Kevin Minard and Charles Ansong, both with PNNL; Susannah Green Tringe at DOE-JGI; Richard Ferrieri at BNL; Mike Sadowsky and Chanlan Chun, both of the University of Minnesota; and Gary Stacey, Yaya Cui and Lihui Song, all at the University of Missouri. The image was created by Alice Dohnalkova. (Image colorized.)