The Chemical and Physical Nature of Particulate Matter Affecting Air, Water and Soil Quality. (NCR174)

Period covered: 01/01/2018 to 12/31/2018

Date of Report: 03/05/2019

Annual Meeting Dates: 01/07/2019, San Diego, CA

Participants:

Stephen Anderson	Missouri - University of Missouri (MO)
Daniel Strawn	Idaho- University of Idaho (ID)
Yucheng Feng	Alabama-Auburn University (AL)
Gediminas Mainelis	New Jersey - Rutgers University (NJ)
Ganga Hettiarachchi	Kansas - Kansas State university (KS)
Kang Xia	Virginia – Virginia Polytechnic Institute and State University (VA)
Don Sparks	Delaware- University of Delaware (DE)
Joseph Stucki	Illinois – University of Illinois (IL)

Brief Summary of Minutes of Annual Meeting:

1. Introductions: Meeting was started with introductions

2. Annual report requirements: Ganga Hettiarachchi shared a message from Jeff Jacobsen, Administrative Advisor, regarding the new report requirements and the formatting changes at the meeting. The report needs to be submitted within 60 days of the Annual Meeting. Ganga Hettiarachchi is to send out a notice to have everyone submit their reports to her by mid-February.

3. State research reports: each attending member gave a brief report on their research activities in 2018 as well as on-going and future research plans

4. Project renewal and new proposal writing (current project is active till 09/30/2020)-All agree that we need to start working on this soon after submitting the Annual report. Discussion was also focused on the new project title.

5. Discuss venue for 2019 Annual Meeting: The group decided to meet again at the 2019 ASA-CSSA-SSSA meeting, San Antonio, TX

6. Other business: potential collaborations and collaborative work
   a. The November–December 2018 issue of the *Journal of Environmental Quality* includes a special section "Soil Chemistry and One Health" (https://dl.sciencesocieties.org/publications/jeq/tocs/47/6) to bring attention to the role of soil as part of the One Health Initiative. Several group members were

involved in organizing the special section or contributing to the collection of papers.

7. The leadership for the group- the group agrees to continue as is.
    a. Chair – Ganga Hettiarachchi
    b. Vice Chair/Secretary- Wei Zhang

Major goals of the project:

1) Characterize the physical, chemical, biological and morphological properties of particulate matter and their environmental, health and economic impacts over a wide range of spatial and temporal scales, including their potential effects on ecological sustainability, food and energy production, climate change, and air, water and soil quality.
2) Upgrade the skills of project participants to do research in heterogeneous environmental systems at the micro- and nano-meter scale.
3) Integrate modern analytical instruments (e.g., synchrotron-based spectroscopy, diffraction and fluorescence, scanning force methods, conventional and laser-based spectroscopy, chemical analysis, and microtomography) and other techniques, including molecular to macroscopic modeling and measuring approaches to promote their use in the agricultural sciences and assist in the development and acquisition of equipment and expertise relevant to the agricultural science community

Impact Nugget

New facilities and equipment

In 2018, Michigan State University team purchased and installed a system of micro-sensors (UNISENSE A/S, Aarhus, Denmark) for laboratory measuring, monitoring, and profiling N_2O and O_2 fluxes in soil.

Time-resolved simultaneous measurement of organic matter and its volatile, intermediate volatility, and semi-volatile vapor phase counterparts at the molecular level enabled by the new instrument under development at University of California, Berkeley. Data collected by this instrument will provide valuable insights into the role of atmospheric aerosols in global climate and regional air pollution. Such data will aid in evaluating the role of biogenic emissions, in assessing importance of a wide variety of anthropogenic emissions in the urban environment, on understanding the global background, and in elucidating the processes transforming organic vapor emissions into particulate organic matter.

Unique project related findings

While the chemical composition of the plant residues and soil moisture and texture have been long known to influence N_2O production and emission, additional never before considered factors that affect the magnitude of N_2O production and emission from soil has been identified. These factors are soil pore structure and physical characteristics, e.g., porosity, of the plant residue (Kravchenko et al., 2018; Kutlu et al., 2018).

Advanced spectromicroscopic data aided with macroscopic observations are useful in obtaining direct evidence of mechanisms of soil organic carbon stabilization within soil microaggregates. We obtained spectromicroscopic evidence in situ, in free soil microaggregates in their native state directly, while preserving the aggregate microstructure. We found preserved less recalcitrant organic carbon species within microaggregates, some stabilized with their original morphology. Strong organo-mineral associations and no-till promoted-stabilization of less recalcitrant organic carbon were also evident. This research provided direct evidence that the surrounding environment plays an important role in stabilizing organic carbon (survivability of organic carbon), stable or easily degradable, together with other factors (Pitumpe Arachchige et al., 2018).

Accomplishment summaries

Members of this project are applying a wide range of analytical tools to elucidate mechanisms of physical and chemical protection of carbon in soils, redox cycling of iron, cycling and reaction pathways of nutrients and contaminants in soils, micro-scale hot-spots of greenhouse gas production within soil pore structure, colloid transport through soil, removal and in situ stabilization of soil contaminants, effect of climate change on soil structure, storage and transport of soil water and contaminant mobility, detection of dense non-aqueous phase liquids (DNAPLs) in geomedia, testing a few different wastewater treatment processes that will supply unrestricted reuse water, will recycle nutrients, and will sequester carbon in soils to help mitigate greenhouse gas increases in the atmosphere, and time-resolved simultaneous measurement of atmospheric aerosols. More detailed description of research outcomes by the group is presented below.

Redox cycling of iron (Fe) in soils between Fe(II) and Fe(III) is linked to carbon and energy flow as well as the movement and bioavailability of most contaminants and nutrients. When soils flood with water and oxygen gets depleted, biogeochemical processes cause reductive dissolution of Fe(III) oxide solids into dissolved Fe(II) and neutralize pH. The fate of released Fe(II) is both complex and important to many paired processes and needs to be better understood. Model systems sorbing Fe(II) with synthetic, pure aluminum (Al) hydroxides show that above neutral pH, Al will partially dissolve and coprecipitate with Fe(II) to form Fe(II)-Al layered double hydroxides (LDH). This still remains to be seen in natural clay minerals, soil or the environment but would represent a missing sorbent and redox reactant in critically-important environments such as wetlands, riparian zones and reducing soils. Work done by Dr. Sparks' group at University of Delaware Our results have shown that Fe(II) precipitation on natural clay minerals is rapid (within 24 hours) and increases with pH. Below pH 7, Fe(II) will adsorb on natural clay minerals but above 7.0 it will begin to precipitate as mixed-metal hydroxides at the clay surface. Identification of the Fe precipitate showed a mixture of Fe(II), Fe(III) and Al in hydroxide minerals. Fe X-ray absorption spectra were best modeled as a combination of mixed Fe(II)-Fe(III) and Fe(II)-Al hydroxides (Fig. 1). This is the first evidence of Fe(II)-Al hydroxides precipitating on natural clay minerals. One important mechanism enhancing the precipitation of Fe(II) is sorption-induced oxidation at the surface of clay minerals. Fe(III) is much more insoluble than Fe(II) at neutral pH and precipitates quickly. To minimize the surface oxidation, a subset of clay minerals was treated with a chemical reductant to saturate the solid with electrons prior to Fe(II) addition. This treatment lowered the amount of Fe(II) sorbed and the precipitate contained more Fe(II) and Al than Fe(III), consistent with less surface oxidation of Fe(II). Our results have indicated that both electron transfer and clay mineral dissolution affect the precipitation of Fe(II) but most importantly, Fe-Al LDHs can form on natural clay minerals. These results are a promising step in proving their existence in reducing soil and their impact on contaminant and nutrient cycling in the environment.

Sea level rise (SLR) poses a great threat to coastal arsenic (As) contamination throughout the globe as its impeding effects will alter current soil biogeochemical conditions. Arsenic has a high affinity for iron (Fe) oxides, which will experience reductive dissolution when they are exposed to flooding conditions, which may lead to As release. However, it is not fully understood how As cycling will behave during flooding with seawater. To better understand As cycling in the face of SLR, laboratory-based experiments in model systems were implemented to simulate flooding conditions on As-sorbed Fe oxides, reflective of the contamination found along the Mid-Atlantic coast of the United States. Work has been shown that when As-contaminated soil was collected from coastal Delaware and reduced in either seawater or fresh river water, more arsenic was mobilized in the river water system. The reason for this difference in not entirely known. Possible formation of ternary complexes between arsenate, divalent cations in seawater, and the mineral surface may contribute to greater sequestration in the seawater system. Spark's group results show that these ternary complexes may be forming in a seawater system when arsenate [As(V)] is sorbed to goethite, a common iron oxide mineral, at a pH above 7. As seen in Fig. 1, more arsenic is sorbed to goethite at high pH in artificial seawater compared to artificial river water. Future experiments are planned to show direct spectroscopic evidence of ternary complexes in a seawater system, which seem to be lacking in the literature. To further assess the effects of SLR on coastal arsenic contamination, a model system consisting of a ferrihydrite-like Fe(III) oxide and arsenate co-precipitate was synthesized and reacted in microcosm vessels over redox and salinity gradients. The mineral was reacted with either artificial river water or artificial seawater and redox potential began at +200 and was abiotically reduced to -300 mV. As redox potential is decreased, results indicate solid mineral transformation from Fe(III) oxide to a mixed valence Fe(II/III) oxide resembling magnetite (Fig. 2). There is greater Fe(III) reduction to Fe(II) and mineral transformation seen in the fresh water system compared to the seawater. The newly formed mineral appears to retain As in its structure in both water environments. Evidence of solid phase As(V) reduction to As(III) was also apparent in both systems, but is more evident in the river water. Results from this study will help improve the current understanding of As mobility by tying together As and Fe cycles in this model system. Results will also contribute to management strategies and remediation plans in SLR impacted coastal soils throughout the world.

Dr. Kravchenko's team at Michigan State University studied the role of physical and biochemical interactions at micro-scale in governing soil's contribution to global cycling of carbon and nitrogen. Specifically, her team identified water absorption by decomposing residues to be one of the primary conditions defining occurrence and strength of micro-scale hot-spots of N_2O production (Kravchenko and Guber, 2017). While the chemical composition of the plant residues and soil moisture and texture have been long known to influence N_2O production and emission, Kravchenko's team identified additional never before considered factors that affect the magnitude of N_2O production and emission from such hot-spots. These factors are soil pore structure and physical characteristics, e.g., porosity, of the plant residue (Kravchenko et al., 2018; Kutlu et al., 2018). The findings will facilitate improving accuracy in modeling and predicting N_2O emissions from agricultural soils and will suggest new management strategies for reducing N_2O emissions.

Atmospheric aerosols are known to impact our climate, both through direct interaction with sunlight and through influence on the formation, lifetime, global extent and brightness of clouds. Secondary organic aerosol (SOA), formed through the atmospheric oxidation of volatile organic compounds (VOCs), comprise a substantial fraction of this atmospheric aerosol. Globally, the burden of secondary organic constituents is much larger than that of directly emitted primary organic particulate matter. This is observed in both rural and urban areas. Yet their formation pathways are only just now beginning to be understood. Time-resolved data at the molecular level for organic matter and its volatile, intermediate volatility, and semi-volatile vapor phase counterparts is critical for understanding the sources and chemistry of atmospheric aerosols. Time-resolved simultaneous measurement of these compounds at the molecular level enabled by the new instrument under development in this on-going project by Dr. Goldstein's Group at the University of California, Berkeley, will provide valuable insights into the role of atmospheric aerosols in global climate and regional air pollution. Such data will aid in evaluating the role of biogenic emissions, in assessing importance of a wide variety of anthropogenic emissions in the urban environment, on understanding the global background, and in elucidating the processes transforming organic vapor emissions into particulate organic matter. When linked to fundamental studies of hygroscopicity and nucleation activation, data at the compound level will be useful in predicting the effects of organic aerosols on clouds, and ultimately on the radiation balance. Such understanding of organic aerosol sources, atmospheric processes, and effects, is necessary for the development of effective pollution reduction strategies and for elucidating the role of aerosols in radiative forcing of earth's climate, thus providing the scientific basis necessary to support models of anthropogenic impacts on climate and sound environmental policy decisions.

Challenges with transport of DNAPLs in geomedia have been observed by scientists and engineers. Detection of residual amounts of dense non-aqueous phase liquids in porous earth materials is important to understand the extent of contamination at sites where these materials have been released. The objectives of the study conducted at University of Missouri by Dr. Anderson and his colleagues, were to develop and test x-ray computed tomography (CT) methods for detection of DNAPL mass in sand cores. Two methods were used to assess the mass balance of trichloroethane (TCA) injected into selected samples. These methods included the spatial distribution method (SDM) which determined the mass from the point of injection and the mass frequency distribution method (MFDM) which used noise correction algorithms. Results showed that these methods can be used to detect the distribution of DNAPL in sand cores. Methods to assess the effective energy of the CT scanner are critical in obtaining accurate information. This study indicates that the scanning methods generally agree with the mass of DNAPL injected.

Dr. Wei Zhang's team studied environmental processes and impacts of emerging contaminants (including pharmaceuticals such as antibiotics, engineered nanoparticles, and antibiotic resistance genes) in soil, water and plant systems. Specifically, his team examined the uptake of pharmaceuticals by vegetables and in-plant transformation of pharmaceuticals (Bhalsod et al., 2018; Chuang et al., 2018; Li et al., 2018). Overhead irrigation of lettuce with pharmaceuticals-containing water resulted in greater residue levels of pharmaceuticals in lettuce shoot (Bhalsod et al., 2018), suggesting that overhead irrigation of vegetables using reclaimed waters should be avoided. As pharmaceutical exposure changes bacterial communities in soils and lettuce, his team assessed whether pharmaceutical exposure through soils and irrigation water could influence the survival of human pathogens in vegetables, due to competition between native bacteria and invading pathogens. His group also investigated the role of stomata to the internalization of silver nanoparticles into plant leaves, using Arabidopsis thaliana as model plants. His team and collaborators investigated novel ZnO nanowires as catalysts for photodegradation of cephalexin, metal-organic frameworks materials for sorption of Se(IV) and Se(VI), and the effect of biochar amendment in soils on zinc uptake by wheat and rice. These findings will improve knowledge on fate and risks of contaminants in soil, water and food crops, and contribute to better strategies for protecting ecosystem and human health.

Biochar may be utilized as a soil-amendment to sequester carbon and enhance soil fertility. Before a widespread scale of biochar application, the potential negative impacts to the environment should be evaluated. Although soybean and lettuce plants were only able to assimilate small amounts of the total solvent-extractable PAHs, plant uptake due to biochar amendment may still pose a hazard to humans. Based on these findings, Dr. Feng's group at Auburn University recommends analyzing PAH concentrations in biochars prior to use as soil amendments, especially for vegetable crop production.

Phosphorus fertilizer use efficiency is poor (10-30% in the first growing season) in many acid and calcareous soils as a result of fixation reactions between the orthophosphate anion and various forms of Ca, Fe or Al that limit the nutrient's availability to plants. Similar issues do exist for micronutrient fertilizers as inclusion of micronutrients in commercial macronutrient fertilizers is a common practice throughout the world due to practical reasons. The cost of conventional phosphorus and micronutrient fertilizers as well as yield loss due to their inefficient utilization is considerable and therefore it is essential to find new application methods or fertilizer enhancement products that can increase the efficiency of P or micronutrient acquisition. Dr. Hettiarachchi and her group at Kansas State University continue to investigate the new fertilizer enhancement products that will enable nutrients to diffuse and/or furnish more plant available nutrients. The group used synchrotron-based x-ray techniques to obtain nutrient reaction products (speciation) to better understand P or micronutrient mobility and potential plant availability in soils. Improved understanding of the fundamental mechanisms responsible for the enhanced mobility or availability of different P and micronutrient fertilizers in carefully selected soil types will help to determine under which circumstances certain P and micronutrient fertilizers offer the potential to significantly increase agricultural productivity. Our work shows that the studied commercially available fertilizer enhancement products do not significantly improve the phosphorus or the micronutrient mobility or lability in soils. They do, however, change the phosphorus and micronutrient reaction pathways and reaction products in soils possibly through changing the Fe, Al or Ca chemistry right around the point of application.

One of the biggest challenges for improving agricultural productivity and environmental sustainability is accurate prediction of the amount of P that will be transported to surface waters in a particular system. The P loading problems to surface waters occur because nutrient management plans do not correctly account for P loading potential of soils and availability of P for offsite transport. The core of the problem is that the soil tests were designed to monitor P availability for plant nutrient management. Research has shown that on some soils, P availability tests values are correlated with the amount of dissolved reactive P (DRP) transported off-site, which is the phase that directly contributes to eutrophication. However, it is commonly observed that test P is not correlated to total offsite P loading to surface waters. Total P loading includes DRP, particulate and organic P. Although the latter two phases are not directly available for uptake by algae, the P associated with these phases can mineralize, desorb or dissolve, thereby releasing P to the solution phase and contributing to environmentally-available P. The extent to which this occurs is not known, and requires knowledge of P speciation of the organic and suspended particles. In the project Dr. Strawn's team at University of Idaho will investigate speciation of soil P, transport of soil solution and particulate P, and recovery of P for use as fertilizers for waste waters. Results will provide new knowledge that will lead to improved water quality and better nutrient use efficiency in agriculture.

Dr. Dan Strawn conducted research on a waste water treatment process that will supply unrestricted reuse water, will recycle nutrients, and will sequester carbon in soils to help mitigate greenhouse gas increases in the atmosphere. He conducted research on how soil factors affect cadmium uptake by wheat grown in the Inland Pacific Northwest wheat growing regions. He researched how dissolved organic matter in manure-amended soils affects Cu mobility, and used FTIR and UV/Vis spectroscopy to elucidate speciation of the dissolved organic matter. He conducted experiments on soil P availability and watershed export from a long-term agriculture watershed research site located on the R. J. Cook Agronomy Farm (CAF) on the Palouse landscape in the Northwest Wheat and Range Region. In this project, he speciated P as inorganic and organic phases. He conducted research to understand distribution, speciation, and availability of phosphorus in a forested landscape in Lake Tahoe National Forest in California.

Similar efforts are on-going at Kansas State University. Dr. Hettiarachchi and her colleagues from College of Engineering (Dr. Prathap Parameswaran and Stacy Hutchinson) working on testing the hypothesis that innovative wastewater treatment technologies can produce the right water from different sources (such as swine wastewater), while recovering nutrients and producing soil amendments for crop production and protecting the environment. Anaerobic membrane bioreactors and Microbial Reverse Electrodialysis Cells are new technologies that can operate sequentially and remove harmful substances from wastewater to produce clean water for reuse. These processes also produce nutrient-rich co-products, allowing balanced/tailored nitrogen and phosphorus applications. Inclusion of new resource/reuse technologies will make agriculture production more sustainable, economical, and environmentally-friendly by reducing food and water quality deterioration from land application of livestock wastewaters and increasing water availability. This transformation will have global impact as confined animal production wastewaters are increasingly used in water scarce areas in both developed and developing countries to meet food production and other demands.

Our group members contributed to organize two special sections in 2018: The November–December 2018 issue of the *Journal of Environmental Quality* included a special section "Soil Chemistry and One Health" (https://dl.sciencesocieties.org/publications/jeq/tocs/47/6) to bring attention to the role of soil as part of the One Health Initiative. Several group members were involved in organizing the special section or contributing to the collection of papers; and one other group member served as one of three guest editors for a special issue "Advancing soil physics for securing food, water, soil and ecosystem services" in Vadose Zone Journal.

In 2018, many of our group members were also focused on increasing communication of science through organizing symposiums at various scientific meetings, in general, the symposiums focused on improving our knowledge on fate and risks of nutrients or contaminants in soil, water and food crops, and contribute to better strategies for protecting ecosystem and human health. Examples are: Dr. Zhang also served as Secretary General for International Symposium on Agro-Environmental Quality, Nanjing, China, November 2-5, 2018. Dr. Hettiarachchi served as chair of International Union of Soil Science Division 4, Commission 4.2 - Soils, Food Security, and Human Health for 2014-2018. She organized and convened a symposium at the 21st World Congress of soil Science entitled "Soil quality to secure human and environmental health", which was held in Rio de Janeiro, Brazil in Aug. 2018.

Impact Statements

1. Our research provided fundamental knowledge to developing better management of soil, soil amendments including fertilizers and crop irrigation practices to maximize crop production while minimizing soil and water contamination, and their risks to food production.

2. Multiple doctoral students have gained significant experience with multiple synchrotron-based tools at national and international laboratories. They have coupled these techniques with other spectroscopic and microscopic approaches to glean insight into cycling of metal contaminants or nutrients under changing environmental conditions.

3. 1 Ph.D. student (Saranya Norkaew, University of Missouri) graduated during this report period.

4. The State of Missouri is investing more than $20 million per year in cost-share with farmers and ranchers to use cover crops to improve long-term soil health for enhanced crop production and to prevent soil erosion. Current research with tomographic imaging has shown increased levels of soil macropores and improved water transport with cover crop management compared to traditional methods.

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