

importance of these two sources changing from transect to transect. Coupled together, the bulk carbon and $\Delta^{13}C$ measurements show that the estuary is adding significant quantities of young DOC despite the existence of vast quantities of old marsh peat flanking the entire estuary. Furthermore, the DIC data indicate that $\Delta^{13}C$ -enriched modern material is also what is fueling the majority of heterotrophic respiration within the system.

B21C-04 0915h

Interactions Between Dissolved Organic Matter and Mercury in the Florida Everglades

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Interactions of mercury (Hg) with dissolved organic matter (DOM) play important roles in controlling reactivity, bioavailability and transport of Hg in aquatic systems. To better define the nature and magnitude of these interactions, experiments were designed using organic matter isolated from various surface waters in the Florida Everglades to determine Hg-DOM binding constants and to study the interactions between DOM and cinnabar (HgS). The isolates, obtained using XAD resins, exhibited a wide range of elemental compositions, aromatic carbon contents, reduced sulfur contents, and molecular weights. Chemical composition of the DOM, especially aromatic carbon and reduced sulfur functional group content, was found to be important in controlling DOM interactions with Hg(II).

Conditional distribution coefficients ($K_{DOM}^{DOM'}$), measured using an equilibrium dialysis ligand exchange method, were strongly affected by the Hg/DOM concentration ratio. Very strong interactions ($K_{DOM}^{DOM'} = 10^{23.2} \text{ L kg}^{-1}$ at pH = 7.0 and I = 0.1), indicative of Hg-thiol bonds, were observed at Hg/DOM ratios below approximately $1 \mu\text{g Hg per mg DOM}$. Hg/DOM ratios above approximately $10 \mu\text{g Hg per mg DOM}$ gave much lower $K_{DOM}^{DOM'}$ values ($10^{10.7} \text{ L kg}^{-1}$ at pH 4.9 to 5.6 and I = 0.1), consistent with Hg binding mainly to oxygen functional groups. These results suggest that the binding of Hg to DOM under natural conditions (very low Hg/DOM ratios) is controlled by a small fraction of DOM molecules containing reactive thiol functional groups.

DOM-Hg interactions were also studied by HgS (log $K_{sp} = -52.4$) dissolution and precipitation experiments. In the dissolution experiments, a significant amount of Hg was released from cinnabar in the presence of DOM, suggesting strong interactions. Conversely, precipitation of metacinnabar (black HgS) was strongly inhibited in the presence of low concentrations (<3 mg C/L) of DOM. In both the dissolution and precipitation experiments, organic matter rich in aromatic moieties was more reactive with HgS than less aromatic fractions and model compounds. These results suggest that DOM can influence the geochemistry and bioavailability of inorganic complexes of Hg in the Everglades, especially HgS, by strong Hg-DOM binding and colloidal stabilization.

B21C-05 0930h

Effects of Human Activities on the Composition of Organic Carbon in Estuaries: Insights Gained Through Lipid Biomarker Studies

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Applications of organic geochemistry have expanded in recent decades to include studies within fields as diverse as ecology and ecosystem science, biogeochemistry, and environmental geochemistry. In part, this expansion has occurred in response to increasing human impacts on the environment which are manifested to a large extent in freshwater and estuarine environments. Human influences to these regions include alterations in carbon and nutrient flow, changes in sedimentation and the introduction of industrial and petroleum products. Organic geochemists bring new tools and insights to these areas improving our ability to trace specific components of organic matter. These approaches have the capacity to improve our understanding of the role nearshore regions play in the global carbon cycle, better identification of the sources and mode of delivery

of natural and anthropogenic organic molecules, and an improved understanding of the fate of organic molecules including remineralization, transformation, or burial in sediments. Chesapeake Bay and San Francisco Bay are the largest estuaries in the continental U.S.A. and provide an interesting comparison for understanding processes influencing the composition of estuarine particulate organic matter (POM). Both estuaries have also been heavily impacted by human activities thus providing an opportunity to investigate how anthropogenic activities influence POM quantity and quality. Results from biomarker analysis of suspended POM and surficial sediments collected from each estuary will be presented and summarized in this talk. Stable isotopic signatures and lipid biomarker compounds reveal spatial variations in organic matter sources along the salinity continuum of each estuary with a stronger terrigenous signature at the freshwater end-member. In addition, temporal variations in freshwater inflow and phytoplankton production influenced the sources and reactivity of POM. This talk will demonstrate that results from biomarker studies provide insights more useful to the restoration and rehabilitation of estuaries than bulk measurements alone. I will also present examples of how biomarker applications can also be used to provide insights regarding changes in organic carbon inputs over historical and geological timescales. In the future, resource managers and policy makers should consider both the amount of carbon produced within each system as well as its composition and source. These parameters influence OM availability to heterotrophic organisms and determine whether a system is likely able to support production at higher trophic levels.

B21C-06 0945h

Carbon Dynamics Change Over Time in a Restored Freshwater Wetland in the Sacramento-San Joaquin Delta, California

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While gaseous carbon (C) losses from a restored freshwater wetland increased during the period from 1997 to 2002, mass loss of litter decreased over time in sequential litterbag decomposition studies. Previously farmed organic soils on Twitchell Island were flooded to two water depths to assess the use of continuous shallow flooding of peat soils to mitigate subsidence in the Sacramento-San Joaquin Delta, California. Compared to a neighboring agricultural field, flooding in the study field reduced total gaseous C losses by as much as one order of magnitude, though methane emissions were higher during. Increases in carbon dioxide emissions corresponded to increased colonization of emergent marsh plants. Decomposition of new litter inputs was examined using litterbags containing senesced shoots of two dominant freshwater marsh plants, cattails (Typha) and tules (Scirpus). Litterbag decomposition studies were initiated in November 1997, February 1999, and May 2000, and each continued for at least 2 years. These studies compared the effects of plant species, water depth, location in the water column on mass loss of litter, and C and nitrogen contents. While the treatment factors affected litter decomposition for each study, these factors did not affect the decreases in the extent of decomposition when consecutive studies were compared, regardless of individual treatments. Thus, total litter loss decreased as the wetland matured. Depending on plant species and litter location, mass loss of litter ranged from approximately 85 to 95% of inputs from November 1997 to January 2000; from approximately 60 to 85% of inputs from February 1999 to March 2001; and from approximately 60 to 75% of inputs from May 2000 to September 2002. These changes in mass loss of litter during marsh development could have a great effect on estimates of C storage potential of these wetlands in the Sacramento-San Joaquin Delta.

B21D MCC: 130 Tuesday 0830h

The Large-Scale Biosphere-Atmosphere Experiment in Amazonia (LBA) I (joint with A, H)

Presiding: C Nobre, Centro de Previsão de Tempo e Estudos Climáticos (CPTEC); M Keller, University of New Hampshire

B21D-01 0830h

Changes in CO₂, N₂O and NO Emissions in Response to Conventional Tillage and No-tillage Management Practices in the State of Rondonia, Brazil

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Land management in the Brazilian State of Rondonia is undergoing a new phase at the start of 21st century. In the 1970s and afterwards, vast tracts of tropical forest were cleared and planted to pasture for cattle grazing. With decades of use, the productivity of these pastures has declined. Now, in an effort to restore productivity, new land management regimes are being implemented that involve either tillage or no-tillage options combined with various combinations of fertilizer application, herbicide use and the planting of a cash crop prior to the planting of forage grasses.

We are studying a subset of these restoration practices in a large-scale (>3 ha), replicated field experiment in an area of degraded pasture at Fazenda Nova Vida, a 22,000 ha cattle ranch in central Rondonia. Here we report on the emissions of carbon dioxide (CO₂), nitrous oxide (N₂O) and nitric oxide (NO) from the initial phases (first six months) of three of the treatments. The treatments are - 1) control; 2) conventional tillage followed by planting of forage grass (Brachiaria brizantha) and fertilizer additions; 3) no-tillage/herbicide treatment followed by two plantings, the first being a cash crop of rice followed by forage grass. In treatment 3, the rice was fertilized.

Relative to the control, tillage increased CO₂ emission by 37% over the first two months, while the no-tillage/herbicide regime decreased CO₂ emissions by 7% over the same period. The cumulative N₂O emissions over the first two months from the tillage regime (0.94 kg N ha⁻¹) were much higher than the N₂O releases from either the no-tillage/herbicide regime (0.64 kg N ha⁻¹) or the control treatment (0.04 kg N ha⁻¹). The highest levels of N₂O fluxes from both management regimes were observed following nitrogen fertilizations. The cumulative NO releases over the first two months were largest in the tillage treatment (0.98 kg N ha⁻¹), intermediate in the no-tillage treatment (0.72 kg N ha⁻¹), and smallest in the control treatment (0.12 kg N ha⁻¹). For the first week following fertilization the percentage of fertilizer N lost as N₂O plus NO was 1.0% for the tillage treatment and 3.0% for the no-tillage treatment.

Because pasture restoration will continue in Rondonia and other parts of the Amazon Basin, we can expect changes in CO₂, N₂O and NO emissions to the atmosphere from the region. For CO₂, both the sign and magnitude of these changes will depend on the restoration regime chosen. Nitrogen oxide emission will increase as the rate of pasture restoration increases with the magnitude of the increases being restoration-regime dependent.

B21D-02 0845h

Regionalization of Methane Emissions in the Amazon Basin with Multi-temporal Microwave Remote Sensing

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Remote sensing of the Amazon basin with passive and active microwave techniques were applied to determine the temporally varying extent of inundation and associated vegetation, and used in conjunction with field measurements to calculate regional rates of methane emission from wetlands to the atmosphere. Monthly inundation areas were derived from analysis of the 37-GHz polarization difference observed by the Scanning Multichannel Microwave Radiometer (1979 - 87) for the mainstem Amazon floodplain in Brazil, the Llanos de Moxos (Beni and Mamore rivers) in Bolivia, the Bananal Island (Araguaia River) and Roraima savannas. Data from the Japanese Earth Resources Satellite-1, L-band synthetic aperture radar were used to determine inundation and wetland vegetation for Amazon basin less than 500 m above sea level at high water (May-June 1996) and low water (October 1995). Although all the measurements of methane emission from aquatic habitats have been performed in the deeply inundated, central basin in open water, flooded forests or floating macrophytes, our basin-wide remote sensing has revealed large areas of seasonally flooded savannas. Therefore, improvements in basin-wide estimates of methane emission will require field studies in wetlands such as those in Bolivia, Roraima and the Bananal.

B21D-03 0900h INVITED

Aerosol and Trace Gases in Amazonia: Impacts on the Ecosystem Functioning and Climate

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Aerosols and trace gases were measured in several intensive campaigns and are being measured continuously in different sites in Amazonia as part of the LBA experiment. The effects of aerosol particles as CCN, as well as impacts on biogeochemical cycles of key forest nutrients such as phosphorus are being assessed. The natural biogenic aerosol is mostly in the coarse mode fraction, and a strong enhancement during nighttime is observed. The fine fraction is mostly organic compounds from the oxidation of monoterpenes. Sulfate and nitrate concentrations are very low. During the dry season, the high concentration of biomass burning aerosol is responsible for a significant absorption of solar radiation of approximately 100 to 250 watts/m², which has important effects on the ecosystem functioning, as well as carbon uptake. Also these aerosols carry significant amount of nutrients such as phosphorus. Ozone concentrations in the wet season averages at a very low 10 ppb at midday, increasing during biomass burning season to 80-100 ppb, in large regions far from the direct impact of biomass burning plumes. Continuous VOCs measurements are revealing interesting effects that could have impacts on secondary aerosol formation via oxidation of monoterpenes and consequently CCN production in the wet season.

B21D-04 0915h INVITED

Aerosols, Clouds, and Climate over the Amazon Basin

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Biogenic emissions dominate the aerosol population over the Amazon Basin under unpolluted conditions, which still exist over much of the basin during the wet season. Aerosol number concentrations and CCN concentrations are low, in the range typical of remote marine locations. A large fraction of particles is of primary biogenic origin, and consist of spores, pollen-related material, microbes, plant debris, etc. Secondary biogenic materials, including organics from VOC oxidation and biogenic sulfate account for much of the rest. Under these low-CCN conditions, cloud droplets can grow rapidly to the size where precipitation occurs and rain production by warm clouds is an important process. During the dry season, large-scale burning due to deforestation and clearing leads to a dramatic increase of aerosol and CCN concentrations. These smoke aerosols consist mostly of organic matter, include light-absorbing organic and near-elemental carbon species, and are efficient CCN. The result of the increased CCN abundance is a major shift towards clouds with high droplet number concentration, and thus increased colloidal stability of the cloud and a lower probability of rainfall from warm clouds. This favors rainfall mechanisms involving ice particles, which has substantial effects for the redistribution of energy and chemical species in the tropical atmosphere. First results from a campaign to study these relationships, using a combination of in-situ measurements, remote sensing, and modeling will be presented.

URL: <http://www.mpch-mainz.mpg.de/smocc/>

B21D-05 0930h

A Large Eddy Simulation (LES) of the Boundary Layer Evolution Over a Deforested Region of Rondonia (Brazil)

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Large Eddy Simulation (LES) is used to study the atmospheric impacts of land use change over Rondonia (Brazil) during the wet season. Spatial distribution of land cover types for the region were evaluated from Landsat images and implemented into the model. Atmospheric variables recorded during the LBA Wet Campaign in January/February 1999 are used as initial conditions for the model and to evaluate its performance. Case studies for selected days having weak winds and strong solar radiation were chosen in order to capture possible landscape impacts on the atmosphere. The results show that the model is able to realistically represent the evolution of the boundary layer and the rainfall distribution. Also, the model shows that the fish-bone structure of the vegetation is able to organize local convection and impact the clouds and rainfall distribution.

B21D-06 0945h

South American Monsoon and the Land Surface Processes

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In this numerical modeling study, the NCEP GCM is applied to investigate the interactions between land surface processes and climate, particularly the effects of land processes on the South American monsoon system (SAMS). A model version with spectral triangular 42 truncation (T42) is used. The corresponding Gaussian grid for T42 is 128 by 64, which is roughly equivalent to 2.8 degrees in latitude and longitude.

Two land surface parameterizations are used. One is the Simplified Simple Biosphere Model (SSiB), which includes explicit vegetation representation. The other parameterization is a surface model with two-soil layers (SOIL) and no explicit vegetation scheme. Two 12-month long simulations were performed with the two parameterizations from initial conditions corresponding to May 1, 1987 and identical distributions of soil moisture and surface albedo. The simulations will be referred to as NCEP GCM/SOIL and NCEP GCM/SSiB. The simulations, therefore, differ in the land surface parameterizations and land cover conditions: one with vegetation and the other with only soil layers (but monthly mean vegetation albedo). This experiment aims to test the role of explicit description of vegetation process in the climate model and hence the role of vegetation in the South American hydrometeorology.

SAMS starts developing in Central America and then moves southeast towards the Amazons in South America. Afterwards, largest precipitation moves northward and eventually retreats northwest. NCEP GCM/SOIL and NCEP GCM/SSiB produce substantially different evolution and spatial distributions of SAMS. In the NCEP GCM/SOIL, the development of SAMS is too fast and too strong with no clear indication of the southward movement. Rainfall magnitudes are much stronger than in the observation. The NCEP/SSiB, on the other hand, correctly simulates SAMS evolution. To understand the mechanisms that contributed to the differences in the simulations, the surface energy and water balances are analyzed.

The NCEP/SSiB produces different spatial distributions of surface sensible and latent heat fluxes than the NCEP GCM/SOIL. These differences are consistent with changes in the circulation, the improvement in the simulation of SAMS. In the pre-monsoon stage, land surface processes mainly influence the flow divergence over Central America. In the mature monsoon stage, they also influence the simulations in the Bolivian high, South Atlantic Convergence Zone, and the southeasterly trade winds.

B21D-07 1030h INVITED

Carbon balance and response to seasonal and long-term forcing at Tapajós Forest old-growth site, Km 67

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We report > 2 years of data defining the carbon budget for this old-growth Equatorial evergreen forest, using comprehensive biometric methods and independent eddy covariance flux methods, and analyze the data in the context of extensive environmental ecological and demographic observations. Validation of the carbon budget is shown using independent ecosystem-scale data from continuous soil flux chambers (Crill and Keller) and continuous atmospheric Rn measurements (Martens and Shay) and comparisons are shown with eddy flux and biometric data from Km 83 control period (Goulden, de Rocha). Net Ecosystem Exchange shows uptake in the dry season and loss in the wet season, opposite to tree growth rates due to the dominance of decomposition processes. Trees are growing rapidly, but high mortality and decay of the large pool of CWD more than offsets growth. This result may be the legacy of ENSO events of the 1990's. Thus the Tapajós old growth forest is losing carbon to the atmosphere.

B21D-08 1045h

Terrain Drainage of CO₂ Enriched Air Might be the Explanation for High Estimates of Rainforest NEE Through Eddyflux Covariance

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Most eddyflux studies carried out in the Amazon on hilly terrain covered by pristine rainforest have found unreasonably high net sinks for atmospheric CO₂. If correctly measured, these CO₂ sinks should imply an increase of organic mass in the forest or carbon should be leaving the system through other pathways, like VOCs or dissolved carbon in the drainage waters. However, independent biometric or biogeochemical budgeting studies have not confirmed the large sinks measured by towers. On the other hand, the method of eddyflux covariance requires that a minimal turbulence exists in order for the system to resolve the signal from the noise in the fluxes of CO₂ across the forest-atmosphere interface. Different from most fluxnet sites, the rainforests of the Amazon have a very dense and closed upper canopy, associated with extremely calm wind conditions at night, which render eddyflux estimates for those periods unreliable. Because NEE is defined as the difference between all CO₂ absorbed by photosynthesis and all CO₂ released by respiration which can accumulate under the forest canopy during nighttime, any problem measuring emitted CO₂ will result in net sinks that are artificially larger. We have identified significant evidence that some emitted CO₂ will drain overland during the night to low lying valley areas, temporarily accumulating there. The venting to the atmosphere next morning, possibly outside the footprint of the tower, could thereby explain why towers positioned on the high lands would miss part of the emitted CO₂, therefore producing overestimates for NEE.

B21D-09 1100h INVITED

Measuring the Effects of Selective Logging on the CO₂ and Energy Exchange of a Tropical Forest

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We have been investigating the effects of selective logging on the exchanges of CO₂ and energy by an old-growth forest in the Tapajós National Forest (TNF), Par Brazil. Our study can be thought of as a large experimental manipulation, where we use a range of techniques, including eddy covariance, biometry and chamber gas exchange, to monitor forest physiology before, during, and after selective logging. Our measurements began in June 2000, and the forest was logged in September 2001. The 15 months of observations before September 2001 provide a measure of the exchanges of CO₂ and energy by intact forest, whereas the observations made after September 2001 provide a measure of the effects of logging. The logging operation, which was conducted by a local firm using reduced impact procedures, covered 400-ha of forest that extended ≈2 km upwind of the tower. The loggers removed only 6% of the biomass in large trees, but left another 18% of the biomass as slash, and eliminated 13% of the canopy on an area basis. Preliminary analysis indicates the forest lost 2 tC ha⁻¹ from September 2001 to May 2002 relative to the previous year, presumably as a result of the decomposition of logging residue and a reduction in leaf area. By May 2002, the forest was no longer losing carbon relative to the previous year, indicating that the canopy had largely recovered, presumably as a result of the rapid regrowth of vegetation in gaps.

B21D-10 1115h INVITED

Time lags in vegetation and the potential for interannual variation in the balance of photosynthesis and respiration in tropical forests

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Although decomposition rates are known to be rapid in Amazonian tropical forests, we measure values of ¹⁴C in soil respiration and soil CO₂ in these ecosystems that are consistently 10-40‰ higher than the ¹⁴C of atmospheric CO₂, the value expected for recent photosynthetic products. Given the rate of decline of ¹⁴C in the atmosphere, these values correspond to carbon fixed on average some 2-7 years ago. Estimates of time lags between photosynthesis and respiration become longer (4-9 years) if we consider only the heterotrophic component of respiration.

Comparison of the radiocarbon signature of CO₂ evolved in soil incubations with those of soil components show that the elevated ¹⁴C in respired CO₂ is largely derived from decomposing litter. Although leaves decompose rapidly on the forest floor, the radiocarbon signature of leaves falling in to litter collectors are equivalent to those of C fixed from the atmosphere 2-5 years previously. Carbon-13 measurements show that these ¹⁴C values are not caused by uptake of respired CO₂. Radiocarbon signatures of fine (<2mm diameter) roots in primary forests are consistent with an average elapsed time of 5-17 years since the fixation of C making up root structural tissues from the atmosphere, though measurements of ¹⁴C in newly grown fine roots indicate that some of that residence time takes place prior to root growth.

A simple model using periodic changes in production of leaf and root tissues, together with the lag between production, death and decomposition, suggests that periodic changes of 5% in NPP can lead to interannual differences in storage or release of 0.5 MgC ha⁻¹ yr⁻¹ of carbon in primary Amazonian tropical forests.

B21D-11 1130h

Controls on Variations of Surface Energy, Water, and Carbon Budgets within Large-Scale Amazon Basin

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A key research focus of the LBA Research Program is understanding the space-time variations in interlinked surface energy, water, and carbon budgets, the controls on these variations, and the implications of these controls on the carbon sequestering capacity of the large scale forest-pasture system that dominates the Amazonia landscape. Quantification of these variations and controls are investigated by a combination of in situ measurements, remotely sensed measurements from space, and a realistically forced hydrometeorological model coupled to a carbon assimilation model, capable of simulating details within the surface energy and water budgets along with the principle processes of photosynthesis and respiration. Herein we describe the results of an investigation concerning the space-time controls of carbon sources and sinks distributed over the large scale Amazon basin. The results are derived from a carbon-water-energy budget retrieval system for the large scale Amazon basin, which uses a coupled carbon assimilation-hydrometeorological model as an integrating system, forced by both in situ meteorological measurements and remotely sensed radiation and precipitation fluxes obtained from a combination of GOES, SSM/I, TOMS, and TRMM satellite measurements.

Results include validation of (a) retrieved surface radiation and precipitation fluxes based on 30-min averaged surface measurements taken at Ji-Paraná in Rondônia and Manaus in Amazonas, and (b) modeled sensible, latent, and CO₂ fluxes based on tower measurements taken at Reserva Jaru, Manaus and Fazenda Nossa Senhora. The space-time controls on carbon sequestration are partitioned into sets of factors classified by: (1) above canopy meteorology, (2) incoming surface radiation, (3) precipitation interception, and (4) indigenous stomatal processes varied over the different land covers of pristine rainforest, partially, and fully logged rainforests, and pasture lands. These are the principle meteorological, thermodynamical, hydrological, and biophysical control paths which perturb net carbon fluxes and sequestration, produce time-space switching of carbon sources and sinks, undergo modulation through atmospheric boundary layer feedbacks, and respond to any discontinuous intervention on the landscape itself such as produced by human intervention in converting rainforest to pasture or conducting selective/clearcut logging operations. The results demonstrate how relative carbon sequestration capacity of the Amazonian ecosystem responds to these controls, and how interpretation of space-time heterogeneities in carbon sequestration depends on a fairly exact quantification of the interacting non-linear properties of photosynthesis in response to incoming solar flux, air-canopy temperatures, and leaf water interception – and soil respiration in response to upper layer soil temperature and water content. The results also show how the interpretation of the control processes is highly sensitive to the scales at which the surface fluxes are analyzed.

B21D-12 1145h

Fieldwork and Statistical Analyses for Enhanced Interpretation of Satellite Fire Data

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Data from satellites are very important for providing information on vegetation fires worldwide. Despite of the broad spatial and temporal coverage, there are several factors that complicate the interpretation of these data. Examples of these factors include fires occurring at times different than the satellite overpasses, the presence of clouds, fires occurring under plant canopies, small fires, and very reflective surfaces. In order to enhance the interpretation of satellite fire data, we are in the process of collecting ground-based data on fires, and relating these data to corresponding information from satellite fire products. Ground-based data are collected using a simple and passive method that allows for a large sample size. One method for data analysis is the construction of error matrices, which can provide statistics on inclusion (commission) and exclusion (omission) errors in satellite fire data. In this work we present results from fieldwork in Para and Mato Grosso, Brazil. These results include fires position, time, size, type of vegetation burned, cloud-cover, and statistics on inclusion and exclusion errors in related remote-sensing fire data. Preliminary analyses suggest that errors of omission are larger than errors of commission, and are dominated by satellite overpass times, cloud-coverage and fire size. Potential strategies to correct for these errors are discussed.

URL: <http://www.firetrip.sr.unh.edu/>

B21E MCC: 132 Tuesday 1015h

Microbial Activity on Mineral and Rock Surfaces II (joint with OS, V)

Presiding: G A Icopini,

Biogeochemical Research Initiative for Education (BRIE), Pennsylvania State University; **A R Zimmerman,** Pennsylvania State University

B21E-01 1015h

Pb(II) Reactivity in Laboratory Cultures of a *Pseudomonas putida* MnB1 - Mn(IV) Oxide Ensemble

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Mn(II) oxidation is a widespread biological process in natural aqueous settings. The products are layered Mn(IV) oxide solids that are very reactive due to their high surface areas, negative charges, and reduction potentials. High affinities for heavy metal sorption have been reported for layer Mn(IV) oxides, and preferential associations in contaminated settings have been found between the Mn-oxide fraction and certain trace metals, including Pb(II). The present study investigates the nature of the interaction of Pb(II) with a layer Mn oxide produced by the soil and freshwater bacterial species *Pseudomonas putida* MnB1 in the laboratory. Wet chemical and X-ray absorption spectroscopy (XAS) techniques were used to elucidate the nature and structure of the sorbed complex in the biogenic Mn(IV) oxide and synthetic analogs. The oxide formed in laboratory culture media appears to be in close physical contact with bacterial cell/biofilm aggregates. In the absence of Mn oxides, Pb(II) was found to associate with organic groups in the biological system, but it preferentially binds to the oxide if present. However, under low ionic strength pretreatment of the ensemble, Pb(II) did not bind to the Mn oxide but was found associated to phosphate or to organic groups depending on equilibration conditions. The complex on the Mn oxide, regardless of loading, is an inner-sphere tridentate complex formed at interlayer positions over vacant sites, with average Pb-O distances of 2.3 Å, a Mn second shell with a Pb-Mn distance of 3.7 Å, and a detectable third shell with a Pb-Mn distance of 5.6 Å. Incorporation of Pb(II) at interlayer positions explains the extremely high sorption capacity of the biogenic oxide and its synthetic analogs. The efficiency of Pb(II) sequestration and the ubiquity of bacterial Mn(II)-oxidation suggest that this mode of sequestration could be a major sink/source for Pb(II) in soil solutions.

URL: <http://mnbiooxides.ucsd.edu/sciapps.shtml>

B21E-02 1030h INVITED

The Reduction of Mn and Fe Oxide Minerals Mediated by Bacteria

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Our work with dissimilatory metal reducing (DMR) bacteria has focused on reactions at the cell/mineral interface and the products that result subsequent to metal reduction. Studies of *Shewanella putrefaciens* have revealed mechanisms that help the bacteria to survive under the nutrient-limited conditions typical of the environments where they are found. These include the formation of cell flocs and the close association of cells with minerals, in which the mineral crystals can penetrate the outer cellular membrane. Furthermore, the interplay of cell- and mineral-related factors determines whether metal-mineral reduction will take place and the products that result. Under nutrient-limited conditions, we have observed that MnIV minerals (birnessite, pyrolusite) are reduced in preference to poorly crystalline ferrous hydroxide, and the crystalline Fe(hydr)oxides goethite and hematite are not reduced at all. The ability of bacteria to attack metal oxide minerals is determined by mineral reactivity and cell fitness. Cell fitness is controlled by the growth conditions of the bacteria and can be manipulated to affect reduction and the crystallization of new minerals. Fast metal reduction initially causes cultures to grow vigorously and minerals to break down rapidly; however, relatively slow rates of reduction extend cell viability and lead to more extensive mineral dissolution. Mineral reactivity has been postulated to be a function of surface area, surface energetics, redox potential and mineral solubility. The results of our work support that reduction potentials play a key role in the ability of *S. putrefaciens* to carry out dissimilatory metal reduction. It is apparent, however, that the processes and outcomes of metal reduction in subsurface environments where DRM bacteria are active depend on the intricate linkage of biotic and abiotic factors.

B21E-03 1045h

Biochemical Analyses of Dissimilatory Iron Reduction by *Shewanella oneidensis*

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Shewanella oneidensis demonstrates respiratory flexibility by the transfer of electrons to Fe (III) and Mn (IV) oxides under anaerobic conditions. Researchers postulate that the bacterium utilizes surface proteins to facilitate the respiratory mechanism for dissimilatory iron(III) reduction. Previous genetic and biochemical studies has shown that iron reduction is associated with the outer membrane of the cell. The identity of the terminal reductase is not yet known. *S. oneidensis* has been shown to use soluble extra-cellular compounds to facilitate iron(III) reduction as well as expression of novel proteins on the cell surface when interacting with iron(III) oxides. Our results show that the outer membrane fraction possess enzymatic activity for converting Fe(III) to Fe(II) as measured by ferrozine complexation. AQDS, extra-cellular organic extracts, and iron(III) both soluble and solid have been assayed for activity with outer membrane fractions. Zymograms of the membrane fractions separated by isoelectric focusing and native PAGE electrophoresis stained using ferrozine have implicated proteins that are directly involved in the Fe(III) reduction process. A proteomics analysis of outer membrane proteins has also been implemented to identify different expression patterns under Fe(III) reducing conditions. Proteins that are unique to Fe(III) reduction have been isolated and identified using N-terminal sequence analysis. We will also attempt to examine the effect of enzymatic iron(III) reduction on isotopic partitioning from in vitro assays.

B21E-04 1100h

Interactions of *Geobacter* with Insoluble Fe(III) and Mn(IV) Oxides

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Geobacter species are dominant members of the microbial community in subsurface environments in which Fe(III) reduction is an important process. Little is known about how these Fe(III)-respiring microorganisms locate and access their substrate, insoluble Fe(III) oxides, which are the predominant form of Fe(III) in sediments and subsurfaces. Previous studies have suggested that *Geobacter* must directly contact insoluble Fe(III) oxide in order to reduce it. When cultured using insoluble Fe(III) or Mn(IV) oxides as the terminal electron acceptor, *Geobacter* expressed flagella and became motile. Furthermore, motile cells were shown to be attracted to Fe(II) and Mn(II) as evidenced by the accumulation of cells around these compounds in chemotaxis assays. Cells also produced pili only when grown with insoluble Fe(III) or Mn(IV) oxides, suggesting that flagella and pili are necessary for the reduction of Fe(III). In order to evaluate the potential role of pili in Fe(III) oxide reduction in *Geobacter sulfurreducens*, deletion mutations were constructed in a gene encoding for piliin, the structural subunit of a piliin filament, and in a gene encoding for a protein involved in twitching motility, a form of surface translocation. Based upon our results, we propose that *Geobacter* senses when soluble electron acceptors are depleted and then synthesizes the appropriate appendages to permit it to search for and establish contact with insoluble Fe(III) or Mn(IV) oxides, and that this strategy may contribute to the predominance of *Geobacter* in Fe(III)-reducing environments.

URL: <http://www.geobacter.org>

B21E-05 1115h INVITED

Microbial Tungsten Extraction From Basalt Under Hydrothermal Conditions (100°C)

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Microbial leaching of trace metals has been shown to be an important process in many environments. Hydrothermal fluids are, however, often more concentrated in transition metals (e.g., Fe, Mn, Cu, Zn) that are scarce in oxic seawater. It is not presently known whether microbes living in hydrothermal fluids need to leach micronutrients from their basaltic environment. Our study has aimed to test this.

Several hyperthermophiles require tungsten (W) for growth including the facultative aerobic *Pyrobaculum aerophilum* (Afshar et al, 1998) and the strict anaerobe *Pyrococcus furiosus* (Roy and Adams, 2002). In contrast to Mo, W is present in about an order of magnitude higher concentration in hydrothermal fluids than it is in seawater (Gallant and Von Damm, 2001) making it a useful trace metal for enzymes that traditionally use Mo in their active sites. Basaltic rocks are, however, poor in both Mo and W. Due the scarcity of W in crustal rocks, we have tested whether W-dependent hyperthermophiles leach W from basalt. Our results have indicated that *Pyrobaculum aerophilum*, when grown under aerobic conditions in the absence of Fe and W and in the presence of basalt powder, enhances the release of W from basalt powder. Concentrations of about 3 ppb were obtained after 15 days, with an abiotic control experiment releasing only 0.1 ppb. The enhanced biological leaching here of W is not observed when growth is in the presence of powdered synthetic basaltic glass enriched in W where abiotic W concentrations were as high as 27 ppb at 15 days. Surprisingly, parallel experiments indicate that *Pyrobaculum aerophilum* does not leach W under anaerobic conditions, while the strict anaerobe *Pyrobaculum islandicum* does demonstrate moderate W-leaching with W levels reaching 9 ppb in 15 days, with an abiotic control experiment releasing 3 ppb. During all of these experiments, no significant microbial leaching above background was observed for Fe, Ni, or Cr suggesting a high degree of metal specificity in this process.

Experiments involving Thermococcales will be discussed, as well as the implications of the results for microbial-rock interaction in hydrothermal settings. Afshar S., Kim C., Monbouquette H.G., Schroder I. (1998) AEM 64, 3004-3008. Roy, R. and Adams, M. W. W. (2002) MBS 39, 673-698. Gallant, R.M. and Von Damm, K.L. (2000) RIDGE 2000 Conference on the Subsurface Biosphere at Mid-Ocean Ridges.