

OS72D MCC: 104 Sunday 1330h

Impacts of Air/Sea Exchange on Biogeochemical Processes in the Ocean I (joint with A, B, GC)

Presiding: W M Landing, Florida State University; C Measures, University of Hawaii; G Cutter, Old Dominion University

OS72D-01 1330h

Atmospheric Transport of Natural and Anthropogenic Substances from East Asia Over NW Pacific During the IOC 2002 Cruise

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Aeolian dust and gaseous and particulate pollutants from the Asian continent are transported eastward over the North Pacific, especially in spring. The episodic atmospheric deposition of natural and anthropogenic aerosols containing iron and other essential trace elements may cause changes in primary productivity of phytoplankton, food web structure and chemical properties of marine atmosphere in the region.

During the 2002 IOC western Pacific cruise, we conducted atmospheric sampling of aerosol and gaseous components on board R/V Melville. Ambient aerosols segregated into two size fractions ($d < 2.5 \mu\text{m}$ and $d > 2.5 \mu\text{m}$) were collected for 24-48 hours on a PTFE fiber filter by using a high-volume dichotomous virtual impactor air sampler with a wind sector control. The filters were analyzed for major inorganic ions and trace metals. Simultaneously we conducted measurements of organic carbon (OC) and elemental carbon (EC) in aerosols ($d < 2.5 \mu\text{m}$) at every 4 hours and size-fractionated particle number densities counted by two optical particle counters ($d > 0.1 \mu\text{m}$ to $> 5 \mu\text{m}$).

Pronounced three atmospheric transport events were observed during the cruise. The first event was measured near the Japan main island ($30 - 35^\circ\text{N}$, $140 - 150^\circ\text{E}$) with the highest Al concentration of 1200 ng m^{-3} on 4-8 May. The second event was found at north of Stn KNOT ($44 - 50^\circ\text{N}$, $155 - 167^\circ\text{E}$) on 12-14 May. The third event was rather weak dust episode but associated with anthropogenic aerosols such as elemental carbon from 45°N to 33°N along the line of 171°E . Particulate Al in coarse mode was occupied over 80% of total Al during the first and second events, while the portion of fine mode Al was increased up to 45% in the third event. Mineral dust particles may have a chance to be modified by the interaction with anthropogenic substances during the transport, especially nitrogen compounds under dry condition.

OS72D-02 1345h

Continuous Measurement of Dust Size Distributions in a Marine Environment by Thermal Volatilization of Sea-Salt

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A newly designed miniature laser optical particle counter (OPC) was used to measure atmospheric dust size distributions on the IOC2002 cruise in the North Pacific in May-June 2002. The OPC was integrated with a 1000°C oven that preheated the aerosol to temperatures high enough to volatilize accumulation mode

pollution soot, sulfates, nitrates and coarse particle sea-salt. This left only crustal dust and fly ash (eg. silicates) as residual aerosol allowing the continuous assessment of dust concentrations in the marine boundary layer by optical techniques. Continuous ancillary measurements of total aerosol light scattering by nephelometry, light absorption by particle soot absorption photometer (PSAP) and total condensation nuclei (CN) counts were also made. These latter instruments all indicated several periods during the cruise when enhanced aerosol was evident and with back trajectories indicative of continental influences. The events were generally more intense near Japan and decreased as the ship tracked east and south toward Hawaii. Dust size distributions obtained from the OPC volatility measurements were also enhanced during these periods with most mass peaks in the diameter range of 2 to $10 \mu\text{m}$. Although elevated concentrations might last several days, variability in dust mass of a factor of 5 or more was often evident over several hours. Atmospheric dust concentrations north of 30°N and between Japan and 170°E were highest and varied generally between 1 and $10 \mu\text{g/m}^3$ with some higher values around May 18-20 (35°N , 170°E). After that period concentrations between $20-30^\circ\text{N}$ remained well below $1 \mu\text{g/m}^3$ for the rest of the cruise. Preliminary comparisons of these new thermal OPC dust measurements with bulk aerosol crustal indicators such as soluble Fe and Al are generally consistent, suggesting that this is a viable technique for making continuous dust size-distribution measurements in the marine boundary layer.

OS72D-03 1400h

The Speciation and Solubility of Aerosol Iron and Aluminum in the Northwest Pacific Ocean: Results From the 2002 NSF/IOC Cruise

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As part of the 2002 NSF/IOC cruise in the north-west Pacific, we collected separate 10-hour aerosol samples during both the day and night aboard the R/V Melville leaving Osaka, Japan on May 1, 2002 and arriving in Honolulu, HI on June 5, 2002. The goal of this research was to measure the solubility and speciation of Fe and Al in Asian continental dust.

Four replicate samples were collected using an automatic sector-controlled aerosol sampling system that collected only when the wind was $\pm 90^\circ$ off the bow of the ship and exceeded 0.5 m/sec . The aerosols were collected on 47 mm PCTE and polypropylene filters for roughly 10-hour periods, filtering as much as 35 m^3 of air through each filter. The filters were changed twice each day for a total of 60 samples.

The filters were quickly leached with 100 mL of either freshly collected $0.2 \mu\text{m}$ filtered surface seawater at natural pH or 100 mL of unacidified ultrapure water. Seawater filtrates were analyzed for soluble Fe(II) using the FeLume chemiluminescent system. These samples were also analyzed for total soluble Fe using the ICP-MS isotope dilution method upon returning to FSU. The ultrapure water filtrate samples were frozen until they could be analyzed at FSU for major anions using ion chromatography. A replicate PCTE filter was analyzed for total Fe (and other elements) using energy-dispersive X-ray fluorescence at the NOAA/PMEL laboratory.

Soluble total aerosol Fe concentrations ranged from $8-130 \text{ pmol/m}^3$ of filtered air. The concentrations of soluble Fe(II) ranged from $0.35-95 \text{ pmol/m}^3$ and total soluble Al ranged from $20-600 \text{ pmol/m}^3$. We will also compare total Fe and Al solubilities in seawater and ultrapure water.

OS72D-04 1415h

Calculating Dust Deposition to the Surface Waters of the Western Pacific and Coming up Short

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The 2002 IOC western Pacific cruise was designed to examine the role that the massive annual outbreaks of dust, emanating from the desert regions of Asia, play in the addition of bioactive materials to the surface of the western Pacific Ocean. Additionally, the cruise was designed to investigate what synergistic role anthropogenic emissions of sulphur and nitrogen oxides from the large population centres in Asia might play in promoting the solubility of this atmospheric dust and delivering anthropogenic pollutants to the surface of the western Pacific Ocean. Initial shipboard results using surface water dissolved Al concentrations indicate that despite the massive offshore atmospheric transport of dust recorded by satellite imagery, actual dust deposition to the oceanic surface waters of this region was modest, in the range of $0.20-8 \text{ gm mineral dust m}^{-2} \text{ yr}^{-1}$. These values contrast sharply with computer deposition models for this region which predict at least $10 \text{ gm mineral dust m}^{-2} \text{ yr}^{-1}$. These lower measured deposition values, and the accompanying low dissolved surface water Fe values, are consistent with the High Nutrient Low Chlorophyll (HNLC) status of the waters north of the Kuroshio Current. The initial Al:Fe ratios also indicate that the surface waters of the NW Pacific region are similar to those of the Pacific section of the Southern Ocean around Antarctica, despite that fact that the NW Pacific underlies one of the most significant continent to ocean dust transfer paths.

OS72D-05 1430h

Dissolved Fe in the Central and Western North Pacific: Results from the 2002 IOC Cruise

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Dissolved Fe concentrations were determined in the North Pacific during the May-June 2002 IOC cruise aboard the R/V Melville. Samples were collected at nine vertical profile stations and continuous underway surface samples were collected using a towed fish during the transects between each station. Surface Fe concentrations varied significantly over the cruise track ranging from $0.1-1.2 \text{ nM Fe}$. The lowest concentrations of $0.1-0.2 \text{ nM}$ occurred around along a N-S transect near 50N , 176E , the tail-end of the subpolar N. Pacific HNLC region. In contrast, the highest surface Fe concentrations occurred along a W-E transect along 26N in the subtropical gyre. These results suggest that surface water Fe concentrations in these regions are due to variations in atmospheric depositions of mineral dust as well as to the hydrographic and biogeochemical boundaries associated with the subtropical and subarctic gyres. Dissolved Fe in the vertical profile samples showed lower concentrations of $0.2-0.8 \text{ nM}$ at or near the surface and increasing concentrations of up to $1.5-1.8 \text{ nM}$ at depth. These open-ocean profiles display the dominant input-output mechanisms to be eolian deposition and biological removal. Variations among select vertical profiles will be discussed as well.

OS72D-06 1445h

Organic Complexation of Fe in the Western Pacific During the 2002 IOC Expedition

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As part of a multi-investigator project, we participated in the 2002 IOC expedition from Osaka, Japan to Honolulu Hawaii. We collected 60 discrete samples from a clean towed fish (depth less than 1 m) for the determination of organic Fe complexing ligands and their associated conditional stability constants. The cruise track was designed to sample many distinct oceanographic regimes. These included, coastal, high nutrient low chlorophyll (HNLC), high atmospheric deposition, high Pb-210, and oligotrophic. We will present data

from competitive ligand equilibration cathodic stripping voltammetry analyses that were conducted both at sea on fresh samples and back at the laboratory on frozen samples. We will contrast the various regimes with respect to concentrations of organic Fe complexing ligands, conditional stability constants, inorganic Fe and organic Fe concentrations. Preliminary results indicate significant differences between the higher latitude NNLC region and the more tropical waters near Hawaii with a greater excess and higher stability constant present in the HNLC region.

OS72D-07 1520h

Organic Fe Complexation in the Gulf of Mexico

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The SWISS (Surface Water Iron Speciation Study) project conducted 3 cruises to the open Gulf of Mexico in 2001 and 2002 (one during the low dust spring season and two during the high dust late summer). The goal of these cruises was to further our understanding of the abiotic cycling of Fe as it relates to atmospheric deposition, redox, photochemistry and organic complexation. Surface (less than 5 m) samples were collected using a clean towed wing. The sample was pumped into a clean work area and filtered. Samples were either frozen for total ligand analysis or ultrafiltered (1 kDa) and frozen for determination of colloidal ligand concentrations. Some subsamples (totals and low molecular weight) were incubated in deck mounted constant temperature (surface seawater) baths in order to determine the importance of photochemistry for the degradation of organic Fe complexing ligands. We will present results from both cruises of 2001. Preliminary data support the hypothesis of photochemical degradation controlling the ligand concentration with the largest percentage of the ligand below 1 kDa.

OS72D-08 1535h

The Unusual Behavior of Metalloid Elements in North Pacific Surface Waters

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The metalloid elements have diverse behaviors in the ocean because of multiple sources (atmosphere, upwelling, rivers) and different chemical forms (speciation) affecting their chemistries and biochemistries. The atmosphere-ocean coupling of metalloids in the North Pacific has not received as much attention, although low productivity HNLC waters in the subarctic Pacific and dust plumes from Asia (that may have enriched metalloid concentrations due to coal combustion) offer ample opportunities to study this coupling. In May-June 2002 surface and deep waters were sampled on a transect from Osaka, Japan northward to the subarctic Pacific (50° N, 168° E), then south into the oligotrophic North Pacific central gyre (24° N, 170° E), and finally to waters northeast of Hawaii (27° N, 150° W). Arsenic and antimony speciation were determined on board ship, while selenium was determined in stored samples, both using selective hydride generation methods.

Surface water As(V) is relatively constant at 15±3 nM, and As(III) and methyl As were elevated in low phosphate waters (lowest in the HNLC region). In contrast, surface water Sb(V) was highest off NE Japan (2 nM) and declined into the HNLC region; thereafter it was ca. 1.2 nM for the remainder of the transect. Methyl Sb and Sb(III) were also highest near the coast of Japan. Selenium displayed the most variations of the 3 metalloids, with the highest total Se, Se(VI), and Se(IV) in the HNLC waters (up to 1.6, 1.0, and 0.2 nM, respectively). In oligotrophic waters it was quite low (0.4 nM) and organic selenide was the dominant form, but northeast of Hawaii the concentrations rose again (to 0.7 nM) in the region of Trichodesmium blooms. These data suggest a strong atmospheric input of Se, and to a lesser extent Sb, superimposed upon biotic processing in surface waters.

OS72D-09 1550h

Reactive Gaseous Mercury Formation Over The North Pacific Ocean: Influence Of Environmental Parameters On Elemental Mercury Oxidation In The Marine Boundary Layer

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Global mercury models have identified wet and dry particle deposition and evasion of dissolved gaseous mercury from the ocean and from land as key controls over global mercury cycling (1,2). Recent ocean studies (3,4) however, have indicated that estimated mercury evasion rates from the ocean substantially exceed estimated deposition. Oxidized reactive gaseous mercury species (RGHg) are now known to play a major role in the global mercury cycle (2,5). RGHg species are water-soluble, exhibit a much shorter atmospheric lifetime than elemental mercury, and contribute to a large extent to atmospheric mercury deposition (2,3,6). Although recent global mercury models have accounted for the dry deposition of RGHg derived from point source emissions (6,7), the formation and deposition of RGHg in remote areas have not been incorporated. We suggest that the oxidation of elemental mercury over the ocean, by gas phase or heterogeneous reactions, is an important part of the global mercury cycle. In agreement with previous studies (3,8,9) our recent data from atmospheric collections over the North Pacific Ocean support the notion of enhanced oxidation in the marine boundary layer. Our results show an inverse correlation between RGHg production and ozone, and a diurnal cycle with highest concentrations during periods of highest UV irradiation. In addition, the relationship between RGHg and other parameters measured during the cruise will be discussed. Our results clearly show that RGHg deposition to the ocean must be an important Hg source, and a crucial part of the global Hg cycle.

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OS72D-10 1605h

Rare Earth Elements in Surface Waters of the NW Pacific Ocean

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The IOC 2002 cruise of the R/V Melville was designed to measure the input of trace elements via atmospheric dust to the Northwest Pacific Ocean. The

expedition was conducted during the latter half of the peak Asian dust season, between May 1 and June 5. The cruise track was also designed in part to sample regions of the ocean where hydrography and biogeochemical gradients cause variations in the dominant processes acting upon trace element supply and remineralization. Departing from Osaka, Japan, the cruise initially followed a southeast track to 31N, 137E, then veered to the northeast to the HNLC region at 50 N, 167 E, during which three stations were occupied. Four stations were subsequently occupied along a southerly track to 24N, 170E. The cruise then veered east towards Hawaii, sampling two additional stations and ending at the Hawaii Ocean Time-series (HOT) station. Surface water samples were collected throughout the cruise by a towed surface sampling fish using Teflon lined tubing.

This presentation will discuss rare earth element (REE) concentrations measured in surface waters collected along the IOC 2002 cruise track. The REE were determined were by flow injection analysis-inductively coupled plasma-mass spectrometry (FIA-ICP-MS). Dissolved Fe and Al concentrations that were used to interpret our data were determined at sea by FIA fluorescence.

One of the main questions that we hope to answer from this work is how do variations in atmospheric transport and deposition of continental dust in surface ocean waters impact the REE content of seawater of the NW Pacific? We also hope to evaluate the solubility of REE that are associated with this dust through a comparison of our data with experiments conducted by Landing and colleagues as well as with dust deposition estimates made during this cruise based on surface water Al concentrations. Additionally, we hope to examine the roles that hydrography and biogeochemical processes play on the cycling of the REE in this part of the world ocean.

OS72D-11 1620h

The Effect of Air/Sea Exchange and Mixing on Organic Carbon Export Calculated from an Oxygen Mass Balance

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The production of organic carbon, and its export from the upper ocean, is a major control on the CO₂ concentration in the atmosphere and thus an important determinant of the earth's climate. The flux of organic carbon from the euphotic zone can be calculated from an upper ocean oxygen mass balance if the rates of physical processes that influence oxygen can be constrained. We present an intensive one-year dataset of oxygen, nitrogen, argon and neon measurements collected at the Hawaii Ocean Time-series (HOT) from July 2000 to June 2001. Oxygen is supersaturated in the surface waters during the entire year due to a combination of biological and physical effects, such as heating and bubble-mediated gas exchange. We use a one dimensional dynamic mixed layer (PWP) model, driven by local heat flux and wind speed estimates, to examine the processes that control gas concentrations. The observed inert gas measurements are used to constrain the rates of bubble-mediated gas exchange by different bubble mechanisms and vertical mixing within the model. Diffusive gas exchange is calculated from wind speed. The model-derived rates of the physical processes combined with the observed oxygen concentrations yield a net biological oxygen production of 1.6 ± 0.8 mol O₂/m²/yr (1.1 ± 0.6 mol C/m²/yr). Refinements to this provisional estimate and an analysis of its sensitivity to the rates of air/sea exchange and mixing will be presented at the meeting.

OS72D-12 1635h

Marine Photochemistry of Hydrogen Peroxide in the Northwest Pacific Ocean

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A systematic study of hydrogen peroxide in seawater, rainwater, and marine air in the Northwest Pacific Ocean was conducted during a transect from Osaka, Japan, to Hawaii, USA, in May and June of 2002. During the transect, surface seawater samples were analyzed continuously for peroxide which showed the effects of photochemical production, wet deposition, and

terrestrial impact. In the surface waters, hydrogen peroxide decreased with latitude from a little over 25 nM in the north (50°N) to more than 150 nM in the south (22°N). This latitudinal variation of hydrogen peroxide followed a trend similar to shipboard measurement of ultraviolet radiation. Diel variations of surface hydrogen peroxide were observed at several locations, with surface water concentrations increasing during the day and decreasing at night. The concentration of surface water peroxide increased to over 200 nM following rain events. Higher concentrations of hydrogen peroxide (>150 nM) were also observed near Asia. The profiles of hydrogen peroxide were obtained at 10 stations that exhibited surface maxima of 24 to 120 nM. The rate constant of dark decay varied from 0.08 d⁻¹ to 0.22 d⁻¹. Rate of photo-production decreased from 10 nM hr⁻¹ at noon to 0 at night. The concentration of hydrogen peroxide varied from 16 μM to 526 μM in rainwater. The data set permits a systematical analysis and modeling of factors regulating the dynamics of hydrogen peroxide in marine environment.

OS72D-13 1650h

CO₂ supply from the North Sea and the Baltic Sea to the North Atlantic Ocean: evidence for the continental shelf pump

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Coastal and marginal seas are thought to act as a continental shelf pump transporting CO₂ from the atmosphere to the open oceans. The CO₂ uptake in coastal seas is triggered by high biological activity increasing the CO₂ concentrations of their waters which finally are transported to the open ocean. The North Sea and Baltic Sea located in north-west Europe are connected via the Skagerrak where the Baltic Sea water first enters the North Sea. The North Sea provides then link to the North Atlantic Ocean. Carbon budgets for the Baltic Sea and the North Sea will be presented in order to provide evidence that in both seas the transfer of CO₂, i.e. the continental shelf pump, acts in two different, but characteristic pattern. The Baltic Sea as a brackish water system collects river water and one might even call it as an estuarine system in a broader sense. Two major drainage areas provide the fresh water supply to the Baltic Sea: The Scandinavian shield supply CO₂ poor waters and the north-east European continent CO₂ rich waters. During the transport of Baltic Sea water to the North Sea the CO₂ concentrations increase continuously. Riverine inputs in part control primary production in the Baltic Sea, however the major control mechanism is the winter nutrient concentrations. These are established by an interaction of production, remineralisation, export and riverine and atmospheric inputs on decadal time scale because of the residence time of the Baltic Sea water of approximately 25years. The Baltic Sea thus acts as a continental shelf pump for atmospheric CO₂ which injects CO₂-rich water to the Atlantic Ocean (injection pump). In contrast the North Sea water is renewed once to twice per year most notably by water from the North Atlantic Ocean. The major control mechanism of the biological activity in the North Sea are thus the continuous (and - in comparison to the Baltic Sea - high) nutrient inputs for the North Atlantic Ocean. For the CO₂ export from the North Sea to the Atlantic Ocean this means that the water is enriched by CO₂ during its 6-12 month travel through the North Sea. Having in mind that almost no burial occurs in the North Sea, the North Sea acts as a continental shelf pump for atmospheric CO₂ by increasing the CO₂ concentrations in the Atlantic waters while they are bypassing through the North Sea (bypass-pump).