Abstract: Aerosol samples were collected in the North Atlantic Ocean during the summer of 2003. Aerosols were fractionated into nine size classes ranging from >18 microns to <0.056 microns. Total element concentrations were measured by energy dispersive x-ray fluorescence to determine the size distribution of biogeochemically important elements. The solubility of Fe and other elements was measured in ultrapure deionized water to investigate the relationship between particle size and aerosol solubility. We found that the majority of soluble aerosol Fe was on particles of >1 μm aerodynamic diameter. Aerosol solubility was somewhat variable but in general, solubility increased with decreasing particle size from 18 microns to 1 microns but solubility decreased in the finest size fractions.
High Resolution Analysis of Particle Size and Aerosol Fe Solubility on Atlantic Aerosols

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Aerosol samples were collected in the North Atlantic Ocean during the summer of 2003. Aerosols were fractionated into nine size classes ranging from \(>18 \mu\text{m}\) to \(<0.056 \mu\text{m}\). Total element concentrations were measured by energy dispersive x-ray fluorescence to determine the size distribution of biogeochemically important elements. The solubility of Fe and other elements was measured in ultrapure deionized water to investigate the relationship between particle size and aerosol solubility. We found that the majority of soluble aerosol Fe was on particles of \(>1 \mu\text{m}\) aerodynamic diameter. Aerosol solubility was somewhat variable but in general, solubility increased with decreasing particle size from \(18 \mu\text{m}\) to \(1 \mu\text{m}\) but solubility decreased in the finest size fractions.

Key Words

Atlantic Ocean, mineral dust, aerosol iron solubility, size fractionated aerosols, mineral dust

1. Introduction

The recognition of the importance of aerosol Fe deposition to marine primary production has created great interest in quantifying both aerosol Fe flux to the oceans and its solubility. In the past decade, numerous studies have measured aerosol Fe solubility and attempted to determine the factors controlling dissolution. These studies have used a variety of techniques to resulting in a large range of solubilities (0.01% - 90%) (Mahowald et al., 2005). The lack of consistency in experimental design has made the
intercomparison of results from different researchers and regions of the world ocean difficult.

Laboratory studies and field experiments have found that soils are generally less soluble than atmospheric aerosols (Zhuang et al., 1990; Baker et al., 2006a; Bonnett and Guieu, 2004; Buck et al., 2006 & 2008) and these results have been interpreted to suggest that atmospheric transport and processing provides the mechanism by which aerosol Fe from mineral dust become increasingly soluble with time (Jickells and Spokes, 2001; Mahowald et al., 2005, Luo et al., 2005, Fan et al., 2006). Changing particle size and interactions between mineral dust and acidic aerosols have been suggested as the primary factors contributing to increased aerosol Fe solubility, though in general neither modeling nor field sampling have been able to confirm these effects (Baker et al., 2006b; Buck et al., 2006 & 2008; Hand et al., 2004; Luo et al., 2005). The size distribution of aerosol Fe is also important to efforts to model the marine Fe cycle as the deposition velocity for the dry deposition of aerosol particles, and in turn the flux of biogeochemically important nutrients, is dependent on particle size (Duce et al., 1991) and some dust aerosol models (Fung et al., 2000; Ginoux et al., 2001; Gao et al., 2003; Zender et al., 2003; Fan et al., 2006) include particle size in their estimations.

Previous investigations of aerosol Fe solubility, including both modeling studies and field experiments, have often divided aerosols into relatively low resolution fine and coarse size fractions using a range of size splits (1.0 – 3.0 μm) (i.e. Siefert et al., 1999; Johansen et al., 2000; Johansen and Hoffman, 2003; Chen and Siefert, 2004; Hand et al., 2004; Luo et al., 2005 & 2008). However, recent work has shown that the accuracy of mineral dust aerosol models may be improved by optimizing the number of particle size
bins and by varying the bin scheme based on particle size (Foret et al., 2006 and Menut et al., 2007) and therefore, high resolution aerosol size distribution data is valuable. We collected aerosol samples from 63°N to 3°S in the Atlantic Ocean that were segregated into nine size classes. These samples were analyzed using an ultrapure deionized water leach technique that allowed us to investigate the relationship between aerosol Fe solubility and particle size in greater detail than in previous studies.

2. Materials/Methods

Aerosol samples were collected during the CLIVAR/CO₂ Repeat Hydrography A16N section using the sector controlled, low volume sampling system described in Buck et al. (2008). Aerosol collection substrates (Osmonics 47 mm, 0.45 μm polypropylene filters) were deployed using a MSP Model #100 nine stage micro-orifice deposition impactor (MODI) for nine 24-hour periods (see Marple et al., 1991 for a more detailed MODI description). The MODI, as opposed to a micro-orifice uniform deposition impactor (MOUDI), was not equipped with a rotational motor resulting in the uneven distribution of aerosol particles on the impaction substrates. The sampler was mounted on an anodized aluminum tower approximately 13 meters above the sea surface and sampling filters for bulk aerosols were mounted approximately four meters higher on the same tower.

All sample handling was conducted inside the trace metals group’s clean van described in Measures et al. (2008). Soluble aerosol Fe was measured in ultrapure deionized water (18 MΩ) that was in gaseous equilibrium with the atmosphere (pH 5.6) following the procedure detailed in Buck et al. (2006). Briefly, this technique measures
the instantaneously soluble Fe fraction in marine aerosols by rapidly passing 100 mL of ultrapure deionized water (DI water) through the aerosol collection substrate. The DI water solution was immediately frozen and stored in the dark. Upon return to FSU, the sample was thawed and analyzed for soluble major anions by ion chromatography and then acidified to 0.024 M quartz distilled HNO$_3$ (Q-HNO$_3$). After several months, the samples were analyzed for DI water soluble elements, including Fe, by HR-ICP-MS.

Leached filters were refrozen and stored in the dark before being analyzed for insoluble aerosol Fe, Zn, Ni, and Cu by energy dispersive x-ray fluorescence (EDXRF). The data for Zn, Ni, and Cu were all at or near their respective analytical detection limits and are not included in this discussion. Because the aerosol particles were not evenly distributed on the substrates it was necessary to analyze each filter several times to produce data within acceptable statistical parameters. Each filter was originally analyzed four times with the filter rotated 90 degrees between each analysis. Minimum statistical requirements for inclusion were insoluble aerosol Fe concentrations of at least 5 ng/cm$^2$ and 20% relative standard deviation calculated from the integrated analytical signal. Samples that failed to meet these criteria were analyzed eight additional times with rotations of 45 degrees between analyses. Samples that remained outside of these statistical parameters are excluded from this discussion (7 of 81 substrates). Bulk samples were collected simultaneously for analysis of total aerosol Fe and soluble aerosol Fe (Buck et al., 2008).

Five and ten day air mass back-trajectories (AMBTs) were simulated using the NOAA Air Resources Laboratory Hybrid Single-Particle Lagrangian Integrated Trajectory Model (HYSPLIT, FNL data set) (Draxler and Rolph, 2003; Rolph, 2003).
The origin of each AMBT was set as the midpoint of the sampling time interval and the ship’s corresponding position for each 24-hour period. Although our samples were collected at a height of approximately 13 meters above the sea surface we selected trajectory arrival heights of 600, 1000, and 1400m; heights both within and above the marine boundary layer (MBL) as there is great uncertainty in the behavior of air masses within the MBL. The ensemble form of the model was also used to model air mass histories at eight locations, each trajectory offset 191 km from the origin in the X-Y plane to allow for better evaluation of the effects of small-scale meteorological conditions on the trajectories. The accuracy of modeled back-trajectories decreases as they move back in time however they are adequate to show general transport patterns on a basin-wide scale.

3. Results and Discussion

3.1 Atmospheric Regimes

Size fractionated aerosol samples were collected on nine sampling days during the summer of 2003 (Figure 1). Representative 5-day AMBTs show that four distinct air mass regimes were sampled; North Atlantic/European (Figure 2a), North Atlantic maritime (Figure 2b), Saharan (Figure 2c), and tropical Atlantic maritime (Figure 2d). These air masses and their respective bulk chemical composition are described in detail in Buck et al. (2008).

3.2 Total and soluble aerosol Fe concentrations
Bulk and size fractionated aerosol samples were collected simultaneously allowing us to compare results from each of the collection systems (Figure 3). The sum concentration of total aerosol Fe ($\Sigma$Fe$_T$; insoluble + soluble Fe) from the MODI samples was calculated by adding the measured concentrations on each of the particle size stages yielding a total concentration for the sampling day. The samples collected on 20 June and 6 July had the greatest variability between bulk and MODI aerosol $\Sigma$Fe$_T$ concentrations, with the concentrations in the MODI samples approximately an order of magnitude greater than in the corresponding bulk samples. Aerosol Fe$_T$ concentrations were also higher in the MODI samples during the five sampling days south of the Saharan dust plume though with less variability in general. The aerosol $\Sigma$Fe$_T$ concentrations are higher on the bulk filters for the samples collected within the plume, by approximately 25% on 24 July and nearly a factor of two on 26 July.

There was significant variability between the sum concentration of DI water soluble aerosol Fe ($\Sigma$Fe$_{DI}$) from the MODI samples and the bulk sample collected in tandem (Figure 4). In general, the concentration of aerosol $\Sigma$Fe$_{DI}$ was greater than the bulk aerosol Fe$_{DI}$ with the exception of 6 July and 4 August. As described in Buck et al. (2008), the 6 July sample was possibly contaminated as the calculated solubility percentage was 135% and therefore both the measured concentration of aerosol Fe$_T$ and aerosol Fe$_{DI}$ should be considered suspect. In evaluating the data in Figures 3 and 4, it is difficult to assess whether the measured concentrations were biased by either collection method as there was no clear pattern showing one method to be more efficient than the other.
Figures 5a-i show the concentration of aerosol Fe\textsubscript{T}, the aerosol Fe solubility percentage, and the cumulative fraction of aerosol Fe\textsubscript{DM} versus the stage aerodynamic diameter cut point for each of the nine sampling periods. Long term studies have shown that the transport of aerosol Fe in the atmosphere over the North Atlantic is dominated by the Saharan dust plume (Prospero, 1990) and samples collected within the dust plume (18.0°N – 11.7°N) showed the highest concentrations of aerosol Fe\textsubscript{T} and were the only samples to have the brown coloration associated with mineral aerosols. Figure 5c shows the aerosol Fe\textsubscript{T} size distribution for the sample collected on 24 July. The influence of mineral dust from the Saharan dust plume is evident in the aerosol Fe\textsubscript{T} concentration (ΣFe\textsubscript{T} = 3070 pmole/m\textsuperscript{3}) and the dominance of the size distribution by particles in the coarse size fraction. This characteristic has been recorded in previous studies in this region as well (Formenti et al., 2003). The aerosol ΣFe\textsubscript{T} concentration is likely to be underestimated as insoluble aerosol Fe data were not included for the 0.098 and 0.056 μm particle sizes because of poor analytical reproducibility. Data from the second Saharan sample, collected on 26 July, are shown in Figure 5d. The aerosol ΣFe\textsubscript{T} concentration is an order of magnitude greater than in the previous example (ΣFe\textsubscript{T} = 44,700 pmole/m\textsuperscript{3}) and the coarse fraction continues to dominate the aerosol Fe\textsubscript{T} load.

The concentrations of aerosol Fe\textsubscript{T} were significantly lower to both the north and south of the Saharan dust plume. The most northern sample (Figure 5a) was collected on 20 June beginning at 62.8°N and sampled the North Atlantic/European air mass. Aerosol Fe\textsubscript{T} concentrations ranged between 69 and 292 pmole/m\textsuperscript{3} (ΣFe\textsubscript{T} = 1050 pmole/m\textsuperscript{3}) with the highest concentration found on the 0.018 μm stage and a relatively high concentration on the 0.056 μm stage. Aerosol collection on 6 July sampled the North Atlantic maritime
air mass and concentrations of aerosol \( \text{Fe}_T \) ranged from 1 to 240 pmole/m\(^3\) (\( \Sigma \text{Fe}_T = 800 \) pmole/m\(^3\)). The concentrations of insoluble aerosol Fe were not reliably measurable on three stages; 18, 1.78, and 0.32 \( \mu \)m (Figure 5b). Anthropogenic aerosols have smaller aerodynamic diameters than mineral dust and the 0.18 \( \mu \)m and 0.056 \( \mu \)m stages had relatively high concentrations of aerosol \( \text{Fe}_T \) suggesting a possible anthropogenic source for both of these samples.

Figure 5e shows data from the first of five samples collected in a tropical Atlantic maritime air mass. These samples had aerosol \( \text{Fe}_T \) concentrations that are significantly lower than those measured in the Saharan dust plume. On 28 July, aerosol \( \text{Fe}_T \) concentrations ranged from 44 to 275 pmole/m\(^3\) (\( \Sigma \text{Fe}_T = 1200 \) pmole/m\(^3\)). Aerosol \( \text{Fe}_T \) from the sample collected on 31 July (Figure 5f) was evenly distributed across the size classes with the exception of a relatively high concentration on the 0.098 \( \mu \)m stage. The concentrations ranged from 33 to 255 pmole/m\(^3\) (\( \Sigma \text{Fe}_T = 750 \) pmole/m\(^3\)). The sample collected on 2 August (Figure 5g) had aerosol \( \text{Fe}_T \) concentrations ranging from 29 to 285 pmole/m\(^3\) (\( \Sigma \text{Fe}_T = 1036 \) pmole/m\(^3\)). Aerosol \( \text{Fe}_T \) was distributed relatively evenly across the size stages for the sample collected on 4 August (Figure 5h), though insoluble aerosol Fe data was omitted for the 0.18 \( \mu \)m stage. The concentrations ranged from 4 to 131 pmole/m\(^3\) (\( \Sigma \text{Fe}_T = 895 \) pmole/m\(^3\)). Aerosol \( \text{Fe}_T \) concentrations were lowest in the sole sample collected in the south Atlantic Ocean (Figure 5i). As in the previous sample, the aerosol Fe load was distributed relatively evenly across the size classes (39 – 80 pmole/m\(^3\); \( \Sigma \text{Fe}_T = 531 \) pmole/m\(^3\)).

Figure 6 plots the contribution of the coarse and fine fractions (1 \( \mu \)m size split) to the concentration of soluble aerosol \( \text{Fe}_{DI} \) for each of the MODI samples. The
concentrations were split evenly between the two size fractions for the samples collected on 20 June and 6 July. The coarse fraction begins to dominate in the dust plume and is most pronounced on 26 July. Sarthou et al. (2003) found a similar trend in the distribution of soluble aerosol Fe (pH = 4.7) collected in the eastern Atlantic Ocean. Samples collected to the south of the plume generally have relatively commensurate aerosol Fe\textsubscript{DI} distributions between coarse and fine fractions, however the sample collected on 2 August carried approximately 80% of the soluble aerosol Fe\textsubscript{DI} on fine particles.

3.3 Aerosol Fe solubility percentage estimates

Estimates of aerosol Fe solubility were calculated from the measured concentrations of soluble and insoluble aerosol Fe. Uncertainty in the solubility percentage was derived from the analytical uncertainty in the EDXRF and HR-ICP-MS analyses by standard methods of error propagation. The overall MODI solubility percentages (\(\Sigma\text{Fe}_{\text{DI}}\%\)) shown in Figure 7 were calculated by adding the insoluble aerosol Fe and aerosol Fe\textsubscript{DI} concentrations on each stage rather than averaging the aerosol Fe\textsubscript{DI} solubility percentages from each size class. In general, the solubility percentages were higher in the bulk samples than in the MODI samples. The two samples collected to the north of the Saharan dust plume, 20 June and 6 July, had overall MODI solubility percentages of 5.4% and 2.7%. On 20 June (Figure 5a), the aerosol Fe percentage solubilities ranged from 2% to 20% with the highest percentages found on the 0.97 and 0.56 \(\mu\text{m}\) stages. As a result, these two particle sizes account for 51% of the total amount of aerosol Fe\textsubscript{DI}. As stated previously, the 6 July sample (Figure 5b) may contain
anthropogenic aerosols given its AMBT and the relatively high concentrations of aerosol Fe$_T$ in the 0.18 and 0.056 \( \mu \text{m} \) size classes, however these samples did not have high solubility percentages (0.6% and 0.5% respectively) as might be expected if there was a large anthropogenic signal in those samples (Sedwick et al., 2007 and references therein).

The two samples collected within the Saharan dust plume, 24 and 26 July, had higher solubility percentages in the MODI samples than in the bulk samples. On 24 July (Figure 5c), the aerosol Fe$_{\text{DI}}$ solubility percentages in the MODI samples ranged from 2% to 25% with low solubilities in the largest two size fractions and the highest solubility on 0.32 \( \mu \text{m} \) particles. Because of its relatively high solubility percentage, this size fraction contributes a significant amount of aerosol Fe$_{\text{DI}}$ to the total amount (~14% of the total).

In the case of 26 July (Figure 5d, the aerosol Fe$_{\text{DI}}$ solubility percentage varied over a greater range, from 3% to 47%), and the MODI $\Sigma$Fe$_{\text{DI}}\%$ was three times that of the bulk sample (Figure 7). The 1.78 \( \mu \text{m} \) size class was the most soluble and such a high solubility for a coarse size fraction is surprising and not easily explained. This sample was enriched in soluble aerosol Fe$_{\text{DI}}$ relative to DI water soluble aerosol Ti (Ti$_{\text{DI}}$) but does not appear to be contaminated as this ratio ($\text{Fe}_{\text{DI}}$/Ti$_{\text{DI}} = 65$) is consistent with the those from the simultaneously collected bulk sample ($\text{Fe}_{\text{DI}}$/Ti$_{\text{DI}} = 61$, aerosol Fe$_{\text{DI}}\% = 3\%$) and the 1.78 \( \mu \text{m} \) sample collected on 24 July ($\text{Fe}_{\text{DI}}$/Ti$_{\text{DI}} = 41$, aerosol Fe$_{\text{DI}}\% = 15\%$) (Table 1). The remaining size fractions had generally high aerosol Fe$_{\text{DI}}$ solubility percentages as well, suggesting that this value was real and not the result of a sampling artifact.

The following two sampling days, 28 and 31 July, each contained particles sizes that had notably high Fe solubilities as well. The 0.97 \( \mu \text{m} \) particles from 28 July (Figure...
5e) had an aerosol Fe solubility of 31% while in general the coarse fraction was more soluble than the fine, however the solubility percentages of the finest particles (0.32 – 0.056 μm) were not significantly different than that of the 18 μm fraction. The 11% ΣFeDI% was significantly lower than the 17% measured in the bulk sample. On 31 July (Figure 5f), the 0.098 μm particles also had a relatively high aerosol FeDI solubility percentage (20%) though the highest solubility was found among the 18 μm diameter particles (31%). The solubility percentage was high on all of the stages as the aerosol ΣFeDI% was 16% (range: 5 – 31%), comparable to the bulk solubility of 18%.

ΣFeDI% was generally low (<10%) in the MODI samples collected in the tropical and equatorial Atlantic. On 2 August (Figure 5g), aerosol FeDI solubility percentages ranged over two orders of magnitude among the size fractions, 0.1% - 10%, and the ΣFeDI% was 4% (bulk solubility = 13%). Aerosol FeDI solubility showed an inverse trend with decreasing particle size. On 4 August (Figure 5h), aerosol FeDI solubility percentages ranged from 0.9% to 23% and ΣFeDI% was equal to 9% while the solubility of the bulk sample was 12%. The final MODI sample, collected on 6 August (Figure 5i), had relatively low aerosol FeDI solubility percentages (2% - 7%). The ΣFeDI% of the MODI samples was 4%, comparable to the 5% solubility measured in the bulk sample.

Figure 8 plots the aerosol FeDI solubility percentage versus particle aerodynamic diameter as well as the mean solubility percentages for each particle size. In general, there was a great deal of variability in aerosol FeDI solubility percentages. Aerosol FeDI solubility was higher in coarse mode particles (mean = 10.7%) than in the fine mode (mean = 6.9%). This is result is in contrast to previous studies that have shown particles in the fine fraction, size splits of 2.5 μm and 1 μm respectively, to be more soluble (Hand...
et al., 2004 and Baker et al., 2006a). Particles at the low end of the coarse mode (1.78
μm and 0.97 μm) were more soluble than the 18 μm and 3.16 μm stages, as would be
expected if particle dissolution was dominated by surface processes. If this were the sole
process involved in aerosol Fe dissolution, then the aerosol Fe_{DI} solubility percentage
would be expected to follow an inverse relationship with particle size. In these samples,
the mean solubility percentage decreased in particles smaller than 0.97 μm, with the
exception of the 0.098 μm stage. While the available data are too limited to draw firm
conclusions, there is evidence that the solubility of aerosol Fe is not a simple function of
particle size and surface area.

It is difficult to conclusively determine what role aerosol acidity might have in
controlling aerosol Fe_{DI} solubility because of the low particle loads on the individual
MODI stages. The vast majority of the stages had concentrations of DI water soluble
NO$_3^-$ and non sea salt (nss) SO$_4^{2-}$ that were below analytical detection limits (Table 1) but
the available aerosol acidity data does not correlate with aerosol Fe$_{DI}$ solubility.
Simultaneously collected bulk aerosol samples also lacked any correlation between
aerosol Fe$_{DI}$ solubility and the concentration acidic aerosol species (Buck et al., 2008).
Johansen et al. (2000) found that soluble aerosol Fe(II) correlated with both nss-SO$_4^{2-}$ and
oxalate in the North Atlantic but neither Hand et al. (2004) nor Baker et al. (2006a)
observed such a relationship.

The concentration of mineral dust in the atmosphere can be estimated from the
concentration of aerosol Fe$_T$ by assuming that Fe is present in the aerosols in the same
proportion as in upper continental crust (3.5% by mass Fe, Taylor and McLennan, 1995).
One disadvantage to using aerosol Fe to predict mineral dust concentration is the
possibility that the concentration of mineral dust is being underestimated in samples containing large proportions of anthropogenically derived aerosols (presumably containing material with >3.5% Fe composition), for example those samples characterized by the AMBTs in Figures 2a and 2b. Figure 9 plots the aerosol Fe\textsubscript{DI} solubility percentage of each particle size versus the calculated concentration of mineral dust in the atmosphere. A significant amount of variability in aerosol Fe\textsubscript{DI} solubility exists but is not related to the calculated concentration of atmospheric mineral dust. Bulk aerosol samples from Buck et al. (2008) also failed to demonstrate a strong inverse relationship between aerosol Fe\textsubscript{DI} solubility percentages and mineral dust concentrations. Other studies of similar spatial range have shown an inverse relationship between mineral dust concentration and aerosol Fe solubility (Baker and Jickells, 2006) and have interpreted this to demonstrate that Fe solubility increases as larger particles are removed from the atmosphere.

4. Conclusion

The numerous efforts to model atmospheric dust and its role in the marine Fe cycle have incorporated a range of particle sizes into models simulating the input of aerosol Fe to the marine environment. Recent work has shown that the accuracy of these models can be improved by optimizing the number of particle size bins (Foret et al., 2006 and Menut et al., 2007) and therefore, highly resolved aerosol size distribution data is necessary to better constrain these models. We collected size fractionated aerosol samples from a variety of atmospheric regimes in the eastern North Atlantic and measured the solubility of aerosol Fe in DI water. The majority of soluble aerosol Fe\textsubscript{DI}
was in the coarse fraction (>1μm) for six of the nine sampling days and we observed that in Saharan samples, >80% of soluble aerosol Fe$_{DI}$ was in the coarse fraction. The aerosol Fe$_{DI}$ solubility percentages on each particle size showed significant variability. Within the coarse fraction, solubility tended to increase as particles decreased in size however in the fine fraction, the solubility tended to decrease with particle size. No simple explanation is apparent for this behavior but it suggests that aerosol Fe solubility is not a simple function of particle size and surface area.

Acknowledgements

We thank John Bullister and Nicky Gruber, chief scientists during this cruise. We also thank Peter McMurry for the use of the MODI deployed for this work. Nathan Buck and Michael Bizimis contributed greatly to the analytical work presented here. We also thank the Captain and crew of the RV Brown for their assistance. We are very grateful to Henry Fuelberg for his assistance in the use of air mass back-trajectories. The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and/or READY website (http://www.arl.noaa.gov/ready.html) used in this publication. We thank the United States National Science Foundation for their financial support of this research through grants OCE-0223378 and OCE-0550317. We would also like to kindly thank the reviewers for their insightful comments.
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- 2002 SWISS-III R.V. Pelican 21 days
- 2003 CLIVAR A16N NOAA Ronald H. Brown 65 days
- 2004 CLIVAR P2 R.V. Melville 72 days
- 2005 CLIVAR P16S R.V. Revelle 42 days
- 2006 CLIVAR P16N R.V. Thompson 42 days
- 2007 CLIVAR I8S R.V. Revelle 41 days

Refereed Publications:


International Meetings: (speaker underlined)


National Meetings: (speaker underlined)


The areas shown were sampled for size fractionated aerosols during CLIVAR/CO₂ A16N. The various atmospheric air regimes are reported; Blue = North Atlantic/European (20 June 2003), Pink = North Atlantic Maritime (6 July 2003), Red = Saharan (24 & 26 July 2003), Brown = Tropical Atlantic Maritime (31 July – 6 August 2003).

Representative 5-day AMBTs created with the NOAA ARL HYSPLIT model. (a) 20 June 2003: North Atlantic Maritime/European air; (b) 6 July 2003: North Atlantic Maritime air; (c) 26 July 2003: Saharan air; (d) 4 August 2003: Tropical Atlantic Maritime air.

A comparison of the concentrations of total aerosol Fe (insoluble + soluble) for the sum of MODI stages (ΣFeₜ MODI) and bulk samples (Buck et al., 2008) collected in tandem.

A comparison of the concentrations of DI water soluble aerosol Fe for the MODI samples (ΣFeԢ MODI = the sum of the stage concentrations) and bulk samples (Buck et al., 2008) collected in tandem. The bulk sample collected on 6 July had an aberrantly high concentration of soluble aerosol FeԢ and is suspected of contamination.

Total aerosol Feₜ concentrations (bars), aerosol FeԢ solubility percentages (triangles), and cumulative fraction of aerosol FeԢ (line) versus the aerodynamic particle size. (a) 20 June, 62.8°N 20.0°W; (b) 6 July, 34.2°N 21.0°W; (c) 24 July, 18.0°N 29.0°W; (d) 26 July, 15.0°N 29.0°W; (e) 28 July, 11.7°N 29.0°W; (f) 31 July, 7.5°N 27.2°W; (g) 2 August, 4.0°N 25.5°W; (h) 4 August, 1.0°N 25.0°W; (i) 6 August, 2.0°S 25.0°W.

A comparison of the relative contributions of the fine (<1 μm) and coarse (>1 μm) aerosol fractions to the total measured concentration of soluble aerosol FeԢ.
Figure 7. Aerosol Fe$_{DI}$ solubility percentages in MODI samples ($\Sigma$Fe$_{DI}$%) and bulk samples (Buck et al., 2008) collected in tandem.

Figure 8. The aerosol Fe$_{DI}$ solubility percentages measured on 74 of 81 MODI stages (diamonds) and the mean solubility percentage of each size class (squares).

Figure 9. Aerosol Fe$_{DI}$ solubility percentages for MODI samples (diamonds) and bulk samples collected along the cruise track (crosses; Buck et al., 2008) versus the concentration of mineral dust in the atmosphere calculated from the concentration of aerosol Fe$_T$ (MODI) and aerosol Al$_T$ (bulk).

Table 1. Size fractionated aerosol concentrations and Fe solubilities along a north-south transect in Atlantic Ocean. Omit = samples did not meet analytical criteria, DL = samples below detection limit, NA = not available because of omitted data
NOAA HYSPLIT MODEL
Backward trajectories ending at 12 UTC 06 Jul 03
FNL Meteorological Data

Source ★ at 34.23 N 21.00 W
Figure 2c

NOAA HYSPLIT MODEL
Backward trajectories ending at 10 UTC 26 Jul 03
FNL Meteorological Data
NOAA HYSPLIT MODEL
Backward trajectories ending at 13 UTC 04 Aug 03
FNL Meteorological Data

Source ★ at 0.97 N 25.00 W

Meters AGL

Figure 2d
Figure 5b: Aerosol Fe$_T$ (pmole/m$^3$) concentration as a function of aerodynamic diameter. The graph shows a trend with increasing concentration as the diameter decreases. The bars represent the percentage (Aerosol Fe$_{DF}$% and Cumulative Fraction) for different diameter categories. The data indicates a higher concentration of Fe$_T$ in smaller diameter fractions.
<table>
<thead>
<tr>
<th>Date</th>
<th>Start Lat</th>
<th>Start Lon</th>
<th>Aerodynamic Diam.</th>
<th>Insoluble Aerosol</th>
<th>DI Water Solv. Aerosol</th>
<th>DI Water Fe</th>
<th>Bulk DI Water</th>
<th>DI Water Solv. Fe</th>
<th>% of Cumulative DI Water Fe</th>
<th>DI Water Solv. NO3^-</th>
<th>DI Water Solv. ss-SO4^-</th>
<th>DI Water Solv. Oxalate</th>
<th>Ti</th>
<th>Fe</th>
<th>pmol/m^3</th>
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2 Sigma Detection limit for 31 m3