Constraining Oceanic dust deposition using surface ocean dissolved Al

Qin Han, J. Keith Moore, Charles Zender, Chris Measures

Abstract

We use measurements of ocean surface dissolved Al, a global Biogeochemical Elemental Cycling (BEC) ocean model, and the global Dust Entrainment and Deposition (DEAD) model, to constrain dust deposition to the oceans. Our Al database contains all available measurements and with best coverage in the Atlantic. Vertical profiles and seasonal data exist in limited regions. Observations show that surface dissolved Al is distributed similarly to the dust deposition predicted by DEAD and other models. The Equatorial Atlantic Al maximum decreases toward higher latitudes. There are high Al concentrations in the Mediterranean Sea and the Arabian Sea and low concentrations in the Pacific and the Southern Ocean. The ocean basins maintain more distinct Al profiles than Fe profiles in the upper ocean, consistent with a weaker biological influence on Al than Fe.

The BEC-predicted surface dissolved Al compares well with observations. The Al distribution reflects the combined effects of Al input from dust and Al removal by scavenging and some biological uptake. Model-observed biases suggest a southward shift of maximum dust deposition compared to current dust model predictions. DEAD appears to overestimate deposition north of 30°N in the Pacific, and to underestimate deposition south of 30°N. Observed Al concentrations and the ocean model-predicted surface Al lifetime provide a semi-independent method to estimate oceanic dust deposition. This technique indicates that DEAD may underestimate total Atlantic dust deposition by about 30 percent, with large regional variations. Our results have implications for all dust-borne ocean nutrients including Fe and demonstrate the potential of marine geochemical data to constrain atmospheric aerosol deposition fields.

1 Introduction

Atmospheric aerosols deliver terrestrial elements essential to the ocean ecosystem and ocean biogeochemical cycles and thus have an important impact on the global carbon cycle. Aerosol-borne nutrients that can limit the growth of phytoplankton include nitrate, ammonium, phosphate, silicate and iron. Iron deficiency limits the primary production in High Nutrient Low Chlorophyll (HNLC) ocean areas [Martin and Fitzwater, 1988] and most iron in the remote ocean comes from mineral dust deposition [Fung et al., 2000].
Unfortunately aerosol deposition to the surface ocean is poorly quantified due to sparse measurements [Prospero, 1996]. Dissolved Al provides an independent estimate for atmospheric mineral dust aerosol deposition [Measures and Brown, 1996; Gehlen et al., 2003]. We use an augmented Al database which includes all relevant Al measurements to characterize, evaluate, and improve our understanding of ocean Al cycling as represented in a state-of-the-art ocean ecosystem model.

Direct measurements of oceanic Al or dust deposition are only available at a handful of remote islands for limited time periods [Duce et al., 1991; Prospero, 1996; Ginoux et al., 2001]. Estimates of ocean nutrient deposition are extrapolated from these observations or predicted by atmospheric models which have been evaluated against available concentration and optical depth data [Andersen et al., 1998; Mahowald et al., 1999; Ginoux et al., 2001; Zender et al., 2003]. The inter-model uncertainty in global deposition estimates is at least a factor of four [Zender et al., 2004]. Deposition uncertainty propagates into uncertainties in oceanic iron availability and in atmospheric aerosol direct and indirect radiative forcing. To better understand the role of aerosol deposition on ocean biogeochemistry and the global carbon cycle, dust input to the ocean must be better constrained.

The concentrations and distributions of trace metals in the ocean are controlled by processes including external input, removal, and internal cycling. It is likely that Al cycles in a manner similar to Fe in the oceans, with strong influence by biologically driven particle scavenging [Bruland and Lohan, 2003]. The most important source for iron and aluminum is partial dissolution from dust deposition. Al solubility estimates range from 0.5–86% with a mean about 5% [Prospero et al., 1987; Sato, 2003; Baker et al., 2006]. Removal processes include active biological uptake and passive scavenging. Most Al is removed by scavenging to particles on a time scale of hours to days and there is relatively little biological uptake. The removal rate is first-order dependent on the particle concentration [Moran et al., 1992]. There is some active uptake and incorporation into the frustules by diatoms [Gehlen et al., 2002]. According to the few deposition observations available, the dissolved Al distribution seems well correlated to the oceanic dust flux [Measures and Vink, 2000]. Typical surface Al concentrations range from 50 to 0.1 nM for high dust area and low dust area respectively.

Aluminum is an ideal tracer for quantifying the dust deposition to the surface ocean due to its geochemical characteristics. First, Al is a major and relatively invariant component of continental materials [Wedepohl, 1995]. Al is the third most common element in continental materials accounting for about 8% of crustal mass. Second, the residence time of Al in the surface ocean is relatively short (∼ 6.5yr) [Jickells et al., 1994] which impedes transfer from the coastal areas to the open oceans. That means the atmospheric input is the main source for the surface ocean Al concentration in the remote oceans. Third, ocean Al chemistry is simple compared to Fe because Al is not involved in complicated redox chemistry. Finally, Al removal is removed with relatively little direct active biological influence.

Measures and Vink [2000] used a simple model named MADCOW to invert observed Al concentrations to dust deposition fluxes. MADCOW assumes that dissolved Al is in steady state, that surface Al originates only from partial dissolution of deposited mineral dust, and
that the Al loss occurs solely from biological particle scavenging. With these assumptions, Al scavenging balances the partial dissolution of deposited Al. MADCOW-inferred dust deposition from surface Al concentrations agrees well with observations [Duce et al., 1991] over four orders of magnitude [Measures and Vink, 2000].

Gehlen et al. [2003] assembled a database for dissolved Al concentrations in oceanic water and used a geochemical ocean general circulation model coupled with the geochemical cycles of Al and Si to study the relationship between surface Al and total dust input. They provided an empirically corrected parameterization of the partition coefficient for Al removal by biogenic opal and they left the Al scavenging rate a free parameter. Their ocean model calculated Al concentration in the surface ocean from two different modeled dust deposition fields [Andersen et al., 1998; Mahowald et al., 1999]. They obtained the best fit between predicted and measured Al concentrations using an Al solubility of 1.5%–3.0%. Their work demonstrates how ocean observations may be used to evaluate atmospheric model estimates of mineral aerosol deposition. The many uncertainties involved in inferring dust deposition from Al measurements include the choice of scavenging parameters, Al solubility, and the surface ocean biology which itself through Fe addition may depend on dust deposition.

Our study extends understanding of ocean Al cycling in the following ways. First, we have assembled all known, relevant oceanic dissolved Al observations into a single database. The database includes eighteen more cruise tracks and stations and approximately three times as many data (10,460 points) as previous studies [Gehlen et al., 2003]. The newer data significantly improves the characterization of the North Pacific and Southern Ocean Al cycle. Second, we develop a more realistic and complete prognostic global ocean Al cycle model which agrees well overall with the measurements. The model provides new and interesting insights on the timescale, solubility, and basin distributions of Al that are most consistent with measurements.

## 2 Methods

### 2.1 Database of dissolved Al

The concentration of dissolved Al in the oceans has been measured during approximately forty cruises and stations since 1976. Our augmented database includes virtually all Al data published since 1979, as well as unpublished data from Measures and Hydes (Table 1). There are 10460 data points in all.

Gehlen et al. [2003] collated data from 12 cruises and stations. These and the 18 newer cruise tracks are plotted in Figure 1. More data will become available in the near future through samples collected on CLIVAR cruises. These data will come from the cruise tracks across the Pacific and the Indian Ocean and they will help to fill in the blanks in these ocean regions.

At the global ocean scale, most regions are still poorly observed. The Atlantic coverage is best, especially the equatorial Atlantic, where dust input is high due to dust storms from...
North African deserts. Most measurements are limited to the surface ocean and only 20% of data are beneath 100 m. There are several transects across the North Pacific. Some data are available in the HNLC region of the Southern ocean and around Southeast Asia. In limited regions, e.g. HOT, BATS, Arabian Sea, the data are sufficient to describe the seasonal variability.

The original batch fluorimetric method for measuring Al using the reagent Lumogallion was developed by Hydes and Liss [1976]. This method has a detection limit of 2 nM of aluminum and has been used in the relatively high concentration region of the Atlantic Ocean. The Flow Injection Analysis (FIA) version of this chemistry has a detection limit of ∼0.6 nM and has been used in various ocean basins. Methods that can be used on even lower values were applied successfully after that: the method using atomic absorption achieved the best detection limits of 0.1 nM [Orians and Bruland, 1986] and the gas chromatography method can be used at levels between 0.6 and 120 nM [Measures and Edmond, 1989]. There is no apparent discrepancy among these methods [Orians and Bruland, 1986; Measures et al., 1986].

Most of the data in Table 1 were obtained from filtered samples, some though were from unfiltered samples. However, the dissolution of particulate Al is negligible if the storage time is minimal and the samples are not acidified [Measures et al., 1986]. Also, the unfiltered data are consistent with those filtered [Bowie et al., 2002].

2.2 Global Biogeochemical Model

In this study, we incorporated ocean Al cycling in the Biogeochemical Elemental Cycling (BEC) ocean model [Moore et al., 2004]. The BEC ocean model is built by coupling the upper ocean ecosystem model [Moore et al., 2002] and an expanded version of the biogeochemistry module [Doney et al., 2003] with the ocean circulation component of the Community Climate System Model 3.0 [Collins et al., 2006]. This marine ecosystem model includes one zooplankton and four phytoplankton functional groups: coccolithophores, small phytoplankton, diatoms, and diazotrophs; five limiting nutrients: nitrate, ammonium, phosphate, iron, and silicate; sinking particulates and dissolved organic matter. In these components, the model tracks carbon, nitrogen, phosphorus, iron, silicon, oxygen and calcium carbonate and then reproduces the biomass, productivity, community structure, and carbon export in the ocean ecosystem.

This coupled model accurately predicts the HNLC regions in the Southern Ocean and subarctic Pacific Ocean except for a larger spatial extent of HNLC conditions in the equatorial Pacific region, which is a common problem in coarse resolution models [Moore et al., 2004]. It also reproduces known basin-scale patterns of primary production, export productions and macronutrient concentrations. The BEC model realistically simulates the biogenic silica production as well as the ocean circulation (e.g., mixed layer depth, element transport) which helps us determine dust deposition from dissolved Al. Sensitivity tests indicate that the model response to variations in atmospheric dust input is realistic compared to field experiments [Moore et al., 2004, 2006]. This indicates that this model is
appropriate for studying the relationship between atmospheric dust input and ocean Al concentration. The BEC model employed here includes modifications to the original version (Krishnamurthy et al., submitted, manuscript available at www.ess.uci.edu/ jkmoore/).

We incorporated a dissolved Al tracer in the BEC model largely following the parameterizations developed for iron (Moore and Braucher, in prep.). Partial dissolution of deposited dust provides Al to the ocean and particle scavenging removes it. Other processes simulated in the Al cycle include biological uptake, remineralization, and advection and mixing, due to ocean physics. Following the ballast mineral model which relates the ballast mineral flux to the POC flux [Armstrong et al., 2002], our model ties Al remineralization to the POC remineralization, which is a good proxy for the total sinking particle flux in the model. According to the ballast mineral model, there are two kinds of sinking particles: easily remineralized and hardly remineralized, which refer seperately to the soft and hard flux described by Armstrong et al. [2002]. The biological uptake of Al depends on the approximate relationship between Al and Si [Gehlen et al., 2002]. The full model details are given in the appendix.

The Dust Entrainment And Deposition (DEAD) model [Zender et al., 2003] provides the dust deposition input field for the BEC model. The DEAD model simulates size-dependent dust processes including mobilization, transport, and dry and wet deposition for particles size from 0.1–10 μm which includes most long-range transported dust. The predicted and observed climatological mean dust deposition flux are compared at 6 oceanic stations, including Oahu [Zender et al., 2003] which is near HOT. The predictions of DEAD for LGM, pre-industrial, current, and doubled CO₂ climates are evaluated against available observations in Mahowald et al. [2006]. Here we use the monthly mean dust deposition field obtained from driving DEAD with 1990s observed meteorology [Zender et al., 2003].

3 Results

3.1 Observed Al Climatology

3.1.1 Vertical profiles of dissolved Al

The observed Al concentrations show clear regional trends with depth at the basin scale (Figure 2a). The highest and next highest Al concentrations are in the Mediterranean Sea and Atlantic Ocean, respectively. Interestingly, the Al concentration near the bottom of the Mediterranean Sea decreases sharply. The Al concentration below the brine interface drops by an order of magnitude and is similar to the observations in the North Atlantic. These data are reported by Hydes et al. [1988] who think the clay mineral formation process may explain this low Al concentration. The Southern Ocean and the Pacific Ocean have very low Al concentrations.

The mean sub-surface ocean (200 m to the bottom) and surface ocean (0–50 m) concentrations are tabulated by basin in Table 2. The Al profiles in each basin are very similar throughout the water column except in the Mediterranean (Figure 2b), and vary similarly to
the dust flux input.

### 3.1.2 Surface Al data and dust-Al conversion

The surface layer (0–50 m) Al data are averaged onto the global model grid (Figure 3a). Data are averaged unweighted where there are multiple points in one grid cell. The distribution of surface dissolved Al is very similar to the distribution of modeled dust deposition with a maximum in the equatorial Atlantic and decreasing to higher latitudes. There are high concentrations in the Mediterranean and Arabian Seas, and low concentrations in the Pacific and Southern Oceans.

We reversed the MADCOW model [Measures and Vink, 2000] to use the dust deposition field from the DEAD model [Zender et al., 2003] to predict the surface Al concentration (Figure 3b). Since, MADCOW relates the dust deposition flux to surface ocean Al concentration by assuming a steady state, a constant Al residence time, a fixed mixed layer depth, and no advection. We can invert MADCOW to estimate Al concentration from the model predicted dust deposition field. The estimated Al correlates well with the observations although there are significant regional biases (Figure 4 a). The inferred Al concentration is systematically too high in the Pacific and in the Arabian Sea, and too low in the South Atlantic. As shown below, our more realistic model with dynamic mixed layer depths and variable Al residence times, significantly reduces these biases.

### 3.2 Comparison to BEC Model

#### 3.2.1 Dissolved Al distributions

The BEC model predicts the dissolved Al distribution that results from the prescribed input Al (via dust) from the DEAD model removed by prognostic dynamics and scavenging. The resulting BEC-predicted surface Al distribution agrees fairly well with observations (Figure 5, Figures 4b) and captures the equatorial Atlantic maximum, with lower concentrations in the Pacific and Southern oceans. The BEC-predicted Al concentrations averaged by basins compare well with the observations at both the surface and the deep ocean (Table 2). In this paper, we will focus on surface Al and inferred dust budget rather than deep ocean Al.

The combined models produce large differences in some regions (Figure 5). The predicted maximum is too far north. The model tends to overestimate Al north of 30 °N in the Pacific and to underestimate south of it. These biases may arise several ways: (1) the BEC model itself, including the parameterization of production, remineralization and more importantly scavenging; (2) the input dust field, which we intend to constrain; (3) the geographic variation of Al concentration in the upper soil; (4) the Al solubility in the atmospheric aerosols. Sampling biases in the Al data cannot be ruled out. In most regions we are forced to compare climatological mean model predictions to Al data sampled at a particular location on a single day.
The BEC-DEAD model Al predictions are significantly improved relative to MADCOW-DEAD predictions (Figures 4a,b). Most BEC-DEAD predictions are within a factor of two of observations. The root mean square difference (rmsd, after logarithm transformation) is reduced by 38%. The regional biases in the Pacific Ocean, the Arabian Sea and South Atlantic Ocean have also been eliminated. Relative to MADCOW, the BEC model predicts longer residence times in the South Atlantic and shorter residence times in the Arabian Sea. This variable Al residence time explains the improved results in these regions. Remaining biases include overestimates in the north Atlantic and underestimations in the South Atlantic. These biases are most likely due to errors in either dust inputs or our assumed solubility.

3.2.2 Residence time

We define the residence time of surface ocean dissolved Al as the ratio of total surface layer Al to the rate of input or removal. BEC predicts surface Al residence time range from months to 73 years (Figure 6), consistent with previous estimates [Orians and Bruland, 1986; Maring and Duce, 1987; Moran et al., 1992; Gehlen et al., 2003]. Short residence times occur in high dust areas (e.g. the equatorial Atlantic) and high biological activity areas (e.g. the equatorial Pacific). Long residence times are associated with low dust input and low biological activity as occurs in polar regions and mid-latitude gyres. The long residence times at polar regions may be unrealistically high since the model includes no biological activity under the sea ice cover.

The wide variation in residence times indicates that the BEC model could improve Al predictions (e.g., from MADCOW) by using a variable residence time. The total Al residence time of the surface ocean (0–50 m) and the whole ocean columns are 2.2 and 53 years, respectively.

Scavenging and biological uptake are the only Al removal processes. In our model, surface Al scavenging dominates the removal processes over more than 50% of the surface ocean. In the North Atlantic the model predicted scavenging can be 1000 times more efficient than uptake. Biological uptake significantly exceeds scavenging where silica production is high and dust flux is low, e.g., the equatorial Pacific. Such regions account for < 10% of the surface ocean. In other regions, scavenging and biological uptake are of similar magnitude. Globally biological uptake accounts for 70% of dissolved Al removal from the upper 50m.

3.2.3 Time Series

The only sites where seasonal cycle data exist for multiple years are HOT (station ALOHA, 22°45′N, 158°W) and BATS (Hydrostation S, 32°10′N, 64°30′W). The predicted Al seasonal cycle amplitude is much weaker than observed (Figure 7), and almost non-existent at HOT, even though the dust deposition varies strongly with season at both sites. The modeled and observed Al concentrations at BATS show similar trends, and both lag the
dust deposition peak by about two months. There is a six-month lag between the dust deposition peak and the observed Al cycle at HOT.

Johnson et al. [2003] measured a surface Al concentration of 3nM lower than the Hawaii Ocean Time-series measurements and even these low end observations are much higher than the BEC predicted Al concentrations at HOT. This suggests a 10 times more dust deposition at HOT if we keep the same solubility. Air mass back-trajectories indicate that dust deposition is higher than predicted [Boyle et al., 2005]. The observed dust deposition flux at Oahu shows that DEAD underestimates dust deposition at only a factor of 2 [Zender et al., 2003]. The rest discrepancy between model and observations should be explained by Al solubility. However, the experiment solubility data suggest that the 5% average is fairly applicable at HOT [Sato, 2003]. Johnson et al. [2003] also found the inconsistency between the surface water aluminum and iron and the expected aerosol concentrations. They suggested a combined effect of higher solubility and higher aerosol scavenging rates than generally assumed. The measured Al solubility is about 4% near HOT on cruise track P16 and higher than model predicted dust deposition near HOT are also sometimes observed [McNaughton et al., 2006; Buck et al., 2006].

3.2.4 BEC model-inferred dust deposition field

If we assume dissolved Al is in steady state, then the observed Al concentration (Figure 3a), modeled mixed layer depth (not shown) and modeled surface Al residence time (Figure 6) define an inferred dust deposition (Figure 8a). This method applies the MADCOW method but removes the assumptions of constant mixed layer depth and residence time. The deposition inferred from observed Al is semi-independent of the modeled dust deposition. In all our experiments, the solubility and Al:dust weight ratio were assumed to be 5% and 8%, respectively.

The Atlantic is the only ocean basin with adequate Al measurements to estimate regional averages of the inferred fluxes (Figure 8a). Based on linear interpolation of missing data points the inferred dust deposition to the entire Atlantic is 300 Tg yr\(^{-1}\). This inferred flux is 30% more than the dust model fluxes [Zender et al., 2003] used to estimate the Al residence time.

One reason why the dust fluxes inferred from Al observations significantly exceed model predictions is the presence of large dust particles not adequately represented in the dust model [Zender et al., 2004]. Accurate but expensive-to-compute atmospheric advection processes result in significantly more mass transported long distances to the oceans [Ginoux, 2003; Grini et al., 2005].
4 Discussion and Conclusions

4.1 Spatial and temporal coverage

Though we have greatly expanded the Al database, the spatial coverage is still not enough
to fully understand the global Al climatology. Some regions, such as the eastern Atlantic,
have sufficient data for detailed comparisons with the model. The temporal coverage is
even poorer. We have only one or two months data at most measurement locations. These
few data determine the annual mean for purposes of model comparison. Since dust input to
the surface ocean is highly episodic, the lack of temporal coverage might bias the derived
annual mean values. At HOT and BATS, this bias can exceed 30% of the annual means
(Figure 7). Inter-annual dust deposition variability could also exceed 30%.

For regions with a longer Al residence time than HOT and BATS, this bias is accept-
able, while for regions with shorter Al residence time, the bias would be higher and would
probably cause a problem. Also, we do not know the atmospheric conditions when the ob-
servation sample were taken. If there were a big dust event just before the ship track went
by, the biases would be even bigger. However, observations display a regional coherence
with similar values, convincing us there was no huge biases.

4.2 Al solubility

Based on numerical experiment with varying solubility to minimize model-measurement
discrepancies, Gehlen et al. [2003] estimate mean Al solubility is 1.5 to 3%. Our model
currently uses a global uniform solubility (5%) greater than Gehlen’s estimate. However, as
mentioned earlier, Al solubility ranges from 0.5% to 86% with a mean near 5% [Prospero
et al., 1987; Sato, 2003; Baker et al., 2006]. One recent study finds that the solubility of Al
from Saharan dust (median value 3.0%) is significantly lower than Al solubility from non-
Saharan source regions (median value 9.0%) [Baker et al., 2006]. The discrepancy between
model and observations at HOT also supports a regionally higher solubility. Measurements
and theoretical considerations suggest that solubility will vary regionally [Prospero et al.,
1987; Sato, 2003; Baker et al., 2006] and as a function of air mass history and chemical
transformation [Luo et al., 2005]. We plan to represent this in future research.

4.3 Comparison with previous research

Our ocean biogeochemistry ecosystem model and Al cycle are more complex than MAD-
COW [Measures and Vink, 2000] and the model of [Gehlen et al., 2002]. Those studies
couple Al to biogenic opal with a first order scavenging coefficient while our model repres-
sents this biological activity, scavenging, and remineralization which depend on the mass
of all sinking particles (mineral dust, organic matter, biogenic silica, and calcium carbon-
ate). This study and Gehlen et al. [2002] simulate realistic surface Al distributions using
different dust inputs. However the vertical profiles are very different. The observed vertical
profile has a surface maximum and subsurface minimum and gradually increases down to the ocean floor (Figure 2). Gehlen et al. [2002] simulate a profile with Al concentration increases with depth, which is opposite this observed trend. Our model agrees well with the observations at the surface and subsurface depths and mimics the observed deep ocean trend. Considering the good surface agreement, these inter-model differences are more likely caused by physical and biological processes than by differing dust inputs.

5 Conclusions

The observed Al concentrations show clear basin scale difference and indicate that Al is a good tracer for estimating dust deposition due to less involvement in biological cycles. We have improved the Al predictions and dust deposition estimations by using a global BEC model with dynamic mixed layer depth and Al residence time. Dust deposition inferred from the observed surface Al concentration with the improved MADCOW model suggests a higher deposition rate over Atlantic. The spatial variation of Al solubility is an important reason for the model-observation discrepancy and we plan to represent this in future research.

References


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Sato, T. (2003), The fractional solubility of aluminum from atmospheric aerosols.


**Appendices**

**A Al Model Description**

We added dissolved Al as a tracer to BEC following the parameterization for iron (Moore et al., in prep.). The only Al source to the ocean is partial dissolution from dust deposition and the only loss from the ocean is scavenging. For the whole ocean together:

\[
\frac{d[Al_{\text{diss}}]}{dt} = \text{DISS} - \text{SCAV} \times r
\]

where \([Al_{\text{diss}}]\) is dissolved Al concentration (mmol m\(^{-3}\)), DISS is partial dissolution of Al from dust deposition (mmol m\(^{-3}\) s\(^{-1}\)), SCAV is scavenging of Al (mmol m\(^{-3}\) s\(^{-1}\)), and \(r\) is fraction of scavenged Al assumed lost to ocean sediments, currently 20%.

Al reaches the deep ocean in sinking dust, by remineralization of production and previous scavenging, and by circulation and mixing. For each layer,

\[
\frac{d[Al_{\text{diss}}]}{dt} = \text{DISS} + \text{REMIN} + \text{CIRC} - \text{PROD} - \text{SCAV}
\]

where REMIN is Al released from remineralization from PROD and part of SCAV(80\%), CIRC is Al transported by circulation and mixing, and PROD is bio-uptake Al.
The dust deposition field can be from any observations or model predictions. Currently we use 1990s dust climatology from the DEAD model [Zender et al., 2003]. Deposited dust at the surface partially dissolves immediately into the mixed layer:

\[ \text{DISS} = S \times (r_{Al} \times F_{d0}) \]  

(3)

where \( S = 5\% \) is the solubility of dust at surface ocean, \( r_{Al} = 8\% \) is Al dust fraction by weight, and \( F_{d0} \) is the flux of dust at surface which comes from the dust deposition field.

As dust particles sink they keep dissolving and cause another \( \sim 6\% \) dissolution of Al throughout the ocean:

\[ \text{DISS} = r_{Al} \times [(F_{ds\_in} - F_{ds\_out}) + (F_{dh\_in} - F_{dh\_out})] \]  

(4)

where \( F_{ds\_in} \) and \( F_{dh\_in} \) are soft or hard dust flux coming into the layer, and \( F_{ds\_out} \) and \( F_{ds\_out} \) are soft or hard dust flux going out of the layer. They are derived by:

\[ F_{ds\_out} = e^{-dz/L_{ds}} \times F_{ds\_in} \]  

(5a)

\[ F_{dh\_out} = e^{-dz/L_{dh}} \times F_{dh\_in} \]  

(5b)

where \( dz \) is layer thickness, \( L_{ds} = 600 \text{ m} \) is the dissolution length scale for soft dust fractions, and \( L_{dh} = 120,000 \text{ m} \) is the dissolution length scale for hard dust fractions. In all layers except the surface \( F_{ds\_out} \) and \( F_{ds\_out} \) from above equal \( F_{ds\_in} \) and \( F_{dh\_in} \) to the next layer. At the surface:

\[ F_{ds\_in} = (1 - S) \times (1 - f_{hd}) \times F_{d0} \]  

(6a)

\[ F_{dh\_in} = (1 - S) \times f_{hd} \times F_{d0} \]  

(6b)

where \( f_{hd} = 97\% \) is the hard fraction of dust flux.

Remineralization of Al depends on POC remineralization:

\[ \text{REMIN} = \text{REMIN}_{\text{POC}} \times \frac{(F_{Al\_in} + F_{Al\_in})}{(F_{POC\_in} + F_{POC\_in})} \]  

(7)

where the notation and conventions for (6b) have been applied to soft and hard POC, and soft and hard Al, respectively. These fluxes are defined as:

\[ F_{Al\_out} = e^{-dz/L_{Al}} \times F_{Al\_in} \]  

(8a)

\[ F_{Al\_out} = e^{-dz/L_{Al}} \times F_{Al\_in} \]  

(8b)

where \( L_{Al} \) is the dissolve length for the soft fraction of produced Al. This length depends on temperature

\[ L_{Al} = \frac{22}{4((T - T_r)/10)} \]  

(9)

The dissolve length for hard fraction of produced Al is \( L_{Al} = 4000 \text{ m} \) is constant.
The fluxes $F_{ds\text{-}in}$ and $F_{dh\text{-}in}$ are zero at the surface, while beneath the surface

\begin{align*}
F_{Als\text{-}in} &= F_{Als\text{-}out} + (1 - f_d) \times (\text{PROD} + 80\% \times \text{SCAV}) \quad (10a) \\
F_{Alh\text{-}in} &= F_{Alh\text{-}out} + f_d \times (\text{PROD} + 80\% \times \text{SCAV}) \quad (10b)
\end{align*}

where $f_d = 37\%$ is the hard fraction of produced Al flux.

In the data Gehlen et al. [2002] reported, we found an approximate relation among bio-uptake Al, bio-uptake Si, dissolved Al and dissolved Si:

\begin{equation}
\frac{\text{PROD}}{P_{Si}} = C_1 \times \frac{[\text{Al}\text{diss}]}{[\text{Si}\text{diss}]} \quad (11)
\end{equation}

where $C_1 = 0.08845$ is a constant. To avoid overflow, we set $P_{Si} \geq 0.1\text{mmol m}^{-3}$.

Scavenging is the dominant process that removes dissolved Al. We model scavenging as proportional to the dissolved Al concentration

\begin{equation}
\text{SCAV} = r_{scav} \times [\text{Al}\text{diss}] \quad (12)
\end{equation}

where $r_{scav}$ is the scavenging rate and is determined by the surrounding particle concentration (including POC, CaCO$_3$, SiO$_2$ and dust)

\begin{equation}
r_{scav} = r_{scav\_base} \times (F_{POC} \times C_2 + F_{CaCO_3} \times C_3 + F_{SiO_2} \times C_4 + F_d \times C_5) \quad (13)
\end{equation}

where $r_{scav\_base} = 0.077\text{yr}^{-1}$ is the base rate, we choose this rate to make the model results best fit the observations; $C_2, C_3, C_4, C_5$, are scaling factors, $F_{POC}$ is POC flux, $F_{CaCO_3}$ is CaCO$_3$ flux, $F_{SiO_2}$ is SiO$_2$ flux, and $F_d$ is dust flux.

Table 1: Sources of dissolved Al measurements

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<td>Sept. 22 – Oct. 25, 1996</td>
<td>Bowie et al. [2002]</td>
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<td>ANT VIII</td>
<td>May 1 – May 29, 1990</td>
<td>Helmers and van der Loeff [1993]</td>
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<th>Cruises or Locations</th>
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<td><em>Helmers and van der Loeff</em> [1993]</td>
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<td>Oct. 1977</td>
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<td>Apr. 1977</td>
<td><em>Stoffyn and Mackenzie</em> [1982]</td>
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<td>MC-80</td>
<td>Sept. – Oct., 1980</td>
<td><em>Orians and Bruland</em> [1985]</td>
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<td>VERTEX-4</td>
<td>Jul., 1983</td>
<td><em>Orians and Bruland</em> [1985]</td>
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<td>MBARI SOLAS</td>
<td>Mar.–May,2001</td>
<td><em>Johnson et al.</em> [2003]</td>
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<td>ORVSKC-34</td>
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<td><em>Narvekar and Singbal</em> [1993]</td>
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<td><strong>Mediterranean Sea</strong></td>
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<td><em>Caschetto and Wollast</em> [1979]</td>
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* The original data are not included in the published paper nor in our research.
Table 2: Mean Al concentrations by basin

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<th>Basin</th>
<th>dust deposition flux (g m(^{-2}) yr(^{-1}))</th>
<th>observed surface ocean (nM)</th>
<th>modeled surface ocean (nM)</th>
<th>observed sub-surface ocean (nM)</th>
<th>modeled sub-surface ocean (nM)</th>
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<tr>
<td>North Atlantic</td>
<td>3.88</td>
<td>23.55</td>
<td>33.56</td>
<td>14.5</td>
<td>19.64</td>
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<tr>
<td>South Atlantic</td>
<td>0.69</td>
<td>15.76</td>
<td>15.33</td>
<td>10.38</td>
<td>5.00</td>
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<tr>
<td>Pacific</td>
<td>0.25</td>
<td>3.07</td>
<td>2.07</td>
<td>2.90</td>
<td>2.69</td>
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<tr>
<td>Southern Ocean</td>
<td>0.29</td>
<td>2.23</td>
<td>1.44</td>
<td>2.04</td>
<td>1.71</td>
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<tr>
<td>Indian Ocean</td>
<td>0.83</td>
<td>15.03</td>
<td>12.82</td>
<td>N.A.</td>
<td>5.82</td>
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<tr>
<td>Mediterranean Sea</td>
<td>5.96</td>
<td>65.15</td>
<td>150.3</td>
<td>132.78</td>
<td>200.31</td>
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<td>tion and BEC (improved MADCOW). (b) Dust deposition predicted by DEAD using observed 1990s meteorology. (c) Relative difference: (Inferred—DEAD)/DEAD.</td>
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Figure 1: Individual cruise tracks. Compare to Gehlen et al. [2003] Figure 2(a).
Figure 2: Dissolved Al concentrations in different ocean basins. (a) 0–6000 m, (b) 0–500 m
Figure 3: Dissolved Al concentration in surface oceans (0–50 m) from (a) observations and (b) reversed MADCOW/DEAD model and dust deposition field.
Figure 4: Comparison of surface ocean (0–50 m) Al concentrations between observations and two models: (a) reversed MADCOW/DEAD, (b) BEC/DEAD. Rmsd means root mean square difference, after logarithm transformation
Figure 5: (a) Dissolved Al in surface ocean (0–50 m) from model results. (b) Dis- solved Al in surface ocean from observations. (c) Relative Error of model: (model- observation)/observation.
Figure 6: Modeled residence time of surface ocean (0–50 m) dissolved Al.
Figure 7: Observed (solid) and modeled (solid blue) Al seasonal cycle (0–50 m) at (a) HOT (unpublished data courtesy C. Measures), (b) BATS [Jickells, 1986] on left hand axis. Dust deposition (dashed) on right hand axis. Whiskers span maximum/minimum observations when multiple years of measurements are available. No whiskers indicates only one year of measurements.
Figure 8: (a) Dust deposition field inferred from observed surface ocean Al concentration and BEC (improved MADCOW). (b) Dust deposition predicted by DEAD using observed 1990s meteorology. (c) Relative difference: (Inferred−DEAD)/DEAD.