



Aluminium in an ocean general circulation model compared with the West Atlantic Geotraces cruises [☆]

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ABSTRACT

A model of aluminium has been developed and implemented in an Ocean General Circulation Model (NEMO-PISCES). In the model, aluminium enters the ocean by means of dust deposition. The internal oceanic processes are described by advection, mixing and reversible scavenging. The model has been evaluated against a number of selected high-quality datasets covering much of the world ocean, especially those from the West Atlantic Geotraces cruises of 2010 and 2011. Generally, the model results are in fair agreement with the observations. However, the model does not describe well the vertical distribution of dissolved Al in the North Atlantic Ocean. The model may require changes in the physical forcing and the vertical dependence of the sinking velocity of biogenic silica to account for other discrepancies. To explore the model behaviour, sensitivity experiments have been performed, in which we changed the key parameters of the scavenging process as well as the input of aluminium into the ocean. This resulted in a better understanding of aluminium in the ocean, and it is now clear which parameter has what effect on the dissolved aluminium distribution and which processes might be missing in the model, among which boundary scavenging and biological incorporation of aluminium into diatoms.

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1. Introduction

The distribution and cycling of aluminium (Al) in the ocean has received attention for a variety of reasons. Firstly, if the Al cycle is understood well, aluminium surface concentrations can be used to constrain atmospheric dust deposition fields (Gehlen et al., 2003; Han, 2010; Han et al., 2008; Measures et al., 2005, 2010), which are used to predict aeolian iron addition to the euphotic zone. This is important, since iron is an essential trace-nutrient for phytoplankton; thus its availability has a direct consequence on primary production and air–sea CO₂ exchange (Boyd et al., 2007; de Baar et al., 2005; Martin, 1990).

Secondly, there is evidence that Al inhibits the solubility of sedimentary biogenic silica (Dixit et al., 2001; Emerson and Hedges, 2006; Lewin, 1961; Sarmiento and Gruber, 2006; van Bennekom et al., 1991). If less biogenic silica gets dissolved from sediments, eventually there will be less silicic acid available in the euphotic zone, which will reduce diatom production as silicon is an essential major nutrient for diatoms. Modified diatom productivity will impact

ocean food webs and the export of organic carbon to the ocean's interior. For advancement in both of these fields of interest a good understanding of the Al cycle is pertinent.

Currently it is assumed that the major source of Al to the ocean is via dust deposition (e.g. Kramer et al., 2004; Maring and Duce, 1987; Measures et al., 2005; Orsian and Bruland, 1986). When dust enters the ocean, a part of its aluminium content (1–15%) dissolves in the uppermost layer and is quickly distributed over the mixed layer by turbulent mixing. Most dust remains in the particulate phase and sinks to the bottom of the ocean, while a small fraction might dissolve in the water column. The dust that does not dissolve at all is buried in the sediment (Gehlen et al., 2003; Sarmiento and Gruber, 2006).

Arguments that dissolution occurs primarily in the upper layer of the ocean come from shipboard experiments and atmospheric moisture considerations. Maring and Duce (1987) and Measures et al. (2010) showed that within a day after deposition, most of the dissolvable Al will be dissolved. Assuming a sinking speed of dust of 30 m/day, most Al would then dissolve in the upper 30 m of the ocean. This depth is shallower than the mixed layer depth, which means that there is little dissolution below the mixed layer.

Even though some earlier studies showed that most deposition is dry (Jickells, 1995; Jickells et al., 1994), more recent work shows that dust deposition is mostly wet (Guerzoni et al., 1997; Vink and

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Measures, 2001). It has been argued that most dust passes through low pH environments in the atmosphere, which means that for wet deposition Al is already dissolved when it enters the ocean surface. Since the wetly deposited Al is most important, dissolution in the surface ocean is most relevant (as a lower bound, since dry deposition results in both surface and water column dissolution). Furthermore, based on the same low pH argument, dissolvable Al from dry deposition is likely to instantaneously dissolve in the surface ocean (Measures et al., 2010, and references herein), making the relative amount of surface dissolution even higher compared to water column dissolution.

Fluvial input can be thought to be important as well, since rivers carry large concentrations of Al, but in estuaries and coastal regions this Al is removed by scavenging of Al onto particles (Brown et al., 2010; Mackin, 1986; Mackin and Aller, 1986; Orrians and Bruland, 1986). There are also indications for Al input as a consequence of sediment remobilisation, as in the Arctic Ocean (Middag et al., 2009) and North Atlantic Ocean (Moran and Moore, 1991). However, the importance of sedimentary sources can vary by basin (e.g. in the Southern Ocean these are small as shown by Moran et al. (1992) and Middag et al. (2011)). Finally, hydrothermal vents are thought to only play a minor role (e.g. Hydes et al., 1986; Middag, 2010). In summary, the dominant external source of aluminium in the ocean is atmospheric dust deposition.

Dissolved aluminium (Al_{diss}) is removed mainly by particle scavenging (Bruland and Lohan, 2006; Moran and Moore, 1989; Orrians and Bruland, 1986; Stoffyn and Mackenzie, 1982). This is the combination of adsorption onto a solid surface, followed by sinking due to insufficient buoyancy of the particulates in the seawater (Bacon and Anderson, 1982; Bruland and Lohan, 2006; Goldberg, 1954). Typically, scavenging is deemed to be reversible, which means that during sinking release of the adsorbed, or particulate, aluminium (Al_{ads}) may occur. This happens both directly (by desorption) and indirectly (by dissolution of the biogenic carrier particles). As a consequence, Al_{diss} concentrations increase with depth (Anderson, 2006; Bacon and Anderson, 1982). In this way Al_{diss} is distributed over depth more efficiently than due to mixing and water mass transport. Aluminium is scavenged relatively efficiently and therefore has a relatively short residence time in the ocean (100–200 years) (Orrians and Bruland, 1985).

Except for scavenging, there are strong suggestions from observations in certain regions that Al is biologically incorporated into the siliceous cell walls of diatoms (Gehlen et al., 2002; MacKenzie et al., 1978; Moran and Moore, 1988; Stoffyn, 1979). It seems that Al does not play an essential role for the diatoms, but it can be incorporated functioning as a replacement for silicon (Si), since it is similar in size. Therefore it is likely that the incorporation ratio Al:Si is close to that of the surrounding waters. These regions include the Arctic Ocean (Middag et al., 2009) and the Mediterranean Sea (Chou and Wollast, 1997; Hydes et al., 1988; MacKenzie et al., 1978). Given the ratio Al:Si of incorporation into the diatom in the photic zone, after remineralisation anywhere in the water column, the same dissolved Al:Si will be present, as long as this is the only source of Al and Si. When the dissolved Al and Si is then advected into the Atlantic Ocean by the North Atlantic Deep Water (NADW), this signal slowly disappears because of other sources of Al among which dust deposition and possibly sediment resuspension (Middag et al., 2011) and a source of Si from Antarctic Bottom Water.

Recent years have seen the development of models of the marine biogeochemical cycle of Al. Gehlen et al. (2003) implemented a basic scavenging model, while Han et al. (2008) also included a biological aluminium incorporation module.

Gehlen et al. (2003) had the objective of testing the sensitivity of modelled Al fields to dust input and thus to evaluate the possibility for constraining dust deposition via Al_{diss} . To this purpose they embedded an Al cycle in the HAMOCC2 biogeochemical model. The model consists of an equilibrium relation between, on the one hand, Al_{ads} and, on the other hand, Al_{diss} . In chemical equilibrium Al_{ads} is proportional to the biogenic silica ($bSiO_2$) concentration. In their work, as well as this

paper, the term biogenic silica or $bSiO_2$ refers to the detrital fractions which is fuelled by diatoms and other silicifying phytoplankton, which have no stable organic matter coating and sink. When $bSiO_2$ sinks to the seafloor (together with adsorbed Al), it is buried. The resulting concentration of modelled Al_{diss} was of the same order as the then published observations, but it suggested a significant overestimation of Saharan dust input (Gehlen et al., 2003) when the dust deposition field of Mahowald et al. (1999) was used.

The main goal of Han et al. (2008) was to better constrain the dust deposition field. For this purpose they used the Biogeochemical Elemental Cycling (BEC) model improved by Moore et al. (2008) as a starting point. They used all dissolved Al datasets used by Gehlen et al. (2003) and added more datasets. Except for scavenging Han et al. (2008) added a biological Al uptake module where the Al:Si uptake ratio is a function of the ambient Al and Si concentrations (Han et al., 2008). The surface residence time of Al for both modelling studies varies strongly between different locations (from less than 1 year to almost 80 years), consistent with other estimates (Orrians and Bruland, 1986; Maring and Duce, 1987; Moran et al., 1992).

Overall, there are a number of questions regarding the oceanic Al cycle that remain to be fully addressed. These touch on issues of ocean circulation, the specific sources and sinks of Al in different parts of the world ocean and what processes are needed to accurately simulate the oceanic distribution of Al.

Firstly, there is the question of the meridional (north to south) distribution of Al through the Atlantic Ocean. In the North Atlantic Ocean and northern seas, water sinks and forms NADW, which is then transported southward (e.g., Gary et al., 2011; Lozier, 2010). In the deep Atlantic Ocean dissolved silicon (Si) concentration increases from north to south (Ragueneau et al., 2000; Sarmiento and Gruber, 2006), while the concentration of Al_{diss} stays relatively constant until about 20°S and then decreases (Middag et al., in preparation; see also Section 4.5). Thus it has a generally opposite behaviour compared to Si. Since there are strong suggestions that the processes controlling the distribution of Si and Al are linked, the question is raised how this negative correlation is possible.

Secondly, there is the question about the observed profiles of Al_{diss} at different locations in the ocean. Generally, profiles of Al_{diss} have a reversible-scavenging profile (increasing with depth) and often with a minimum near 1 km depth and a maximum at the surface because of dust deposition. However, observations in the Mediterranean Sea (Chou and Wollast, 1997; Hydes et al., 1988) and IPY-Geotraces-NL observations in the eastern Arctic (Middag et al., 2009) show that there is a strong positive relation between aluminium and silicon. This supports the hypothesis of biological incorporation of aluminium into the cell wall of diatoms.

These issues can be analysed further by the use of numerical models. Since there is a strong spatial variation in aluminium concentration (and its relation to silicon), an ocean general circulation model should be used to simulate the distribution of Al_{diss} . Potentially crucial parameters and sources can be modified in the model to test its sensitivity to these changes. In this way a better understanding of the aluminium cycle can be reached.

In this paper the observed distribution of Al is modelled and the processes driving it are examined. Based on new observations and previous work on aluminium modelling (Gehlen et al., 2003; Han et al., 2008) a model of aluminium based on dust deposition and scavenging by biogenic silica is formulated. This model and the configuration of the simulations will be set out in the following section. Then the observations which are used to check and improve the model will be discussed. The results of the several experiments follow in Section 3, as well as a comparison with the observations. The discussion in Section 4 comprises of a comparison between our model results and Gehlen et al. (2003), a timescale analysis and based on that a discussion of our simulations. Our results are not compared with Han et al. (2008), since we have not performed simulations with biological aluminium

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