

- *L7 Orians and Bruland are not the primary reference for hydrolysis of dissolved Al. They do not show this to be true, they reference someone else*

→We have removed Orians and Bruland and just left Roberson and Hem.

- *L17 There are other papers that show a fluvial source of Al. There are two Brown and Bruland papers, and the Moran and Moore paper referenced here also shows a fluvial source. I think another one of your references or associated references shows a fluvial source as well. I suggest that you reference those papers for a fluvial Al source. Such referencing supports your Iberian margin surface Al data from the Tagus estuary.*

→We have added more references for these lines.

Page 2 line 10-14: Fluvial inputs were historically considered a dominant source of Al to the surface oceans (Stoffyn and Mackenzie, 1982). Whilst Al removal during estuarine scavenging processes appears to strongly reduce the riverine Al outflow (Hydes, 1989) a significant fluvial source of Al is still generally observed (Brown and Bruland, 2009; Brown et al., 2010; Grand et al., 2015).

- *L20 "Sediment resuspension processes at ocean margins with strong boundary currents (Jeandel et al., 2011) and in benthic nepheloid layers (BNLs) (Hesse et al., 1999; Middag et al., 2015b; Moran and Moore, 1991) represent important sources of Al to the deep ocean."*

Should be rewritten "Sediment resuspension represents an important source of Al to the deep ocean especially along ocean margins with strong boundary currents (Jeandel et al., 2011) and in areas with benthic nepheloid layers (BNLs) (Middag et al., 2015b; Moran and Moore, 1991).

→We have modified this part as suggested.

- *L27 Probably worth defining active and passive scavenging?*

→We have added a definition for active and passive scavenging.

Page 2, line 20-30: Removal of Al in oceanic waters occurs through particle scavenging with subsequent sinking of this particulate matter (Orians and Bruland, 1986). The removal occurs via both active and passive scavenging. Active scavenging occurs when dAl is actively incorporated into the atomic structure of opaline diatom frustules, a

process which has been demonstrated in laboratory experiments and is also supported by positive correlations between orthosilicic acid (Si(OH)_4) and Al in depth profiles upon the sinking and remineralization of diatomous material (Gehlen et al., 2002; Hydes et al., 1988; Hydes, 1989; Middag et al., 2009; Middag et al., 2015; Moran and Moore, 1988). Passive scavenging is defined as dAl being adsorbed onto any particle surface without being intrinsically incorporated into cellular structures. This is inclusive of adsorption onto biogenic particles. Evidence for the *post-mortem* incorporation (e.g. passive scavenging) of Al onto diatom frustules and concomitant removal from the dissolved phase is given in Koning et al. (2007) and Vrieling et al. (1999).

- *Note that Orians and Bruland say “The markedly different distributions of dissolved Al and the nutrient silicate in the open ocean, as well as their opposite inter-ocean fractionation trends support a passive adsorption removal mechanism.” Unless I don’t understand this paragraph, I don’t think that this is what you are saying.*

→ Yes. You are right. It is clear that in the Pacific Ocean, due to the scavenging nature of dAl versus the recycled nature of Si, both elements are decoupled and do not show a correlation. Dissolved aluminium will continuously be scavenged along the flow path of water masses. Thus, observations in the Pacific would, generally, conclude that passive adsorption is the main mechanism removing dAl. However, in our study region, with younger water masses, stronger phytoplankton blooms (including diatoms and coccolithophorids) and freshly exported organic material it seems that the cycle of dAl is much more closely related to primary productivity than is the case in the Pacific gyres.

- *Also, does this section preclude the removal of Al by particles that are not biogenic Si?*

→ No, not at all.

- *Can Al be scavenged by other particles? Must it be incorporated into the frustule or can it be on the organic matter?*

→ We have rewritten this paragraph in order to make clear that dAl is not necessarily removed by biogenic Si and that it also can be removed by other biogenic particles. See the above answer to comment “L27 Probably worth defining active and passive scavenging?”.

- *L5 do you mean high resolution sections as opposed to profiles?*

→Yes. We have rewritten this sentence.

Page 2, line 31-32: In the North Atlantic (40° N-65° N) vertical dAl profiles combined with high resolution sections were scarce prior to the GEOTRACES era.

- *L8 there must be more N Atlantic data from Hydes, Measures and others that you could reference here?*

→Indeed there are more studies which we could reference. We have tried (sections 3.2.3, 3.3, 3.4.1) to compare our data with the most recent GEOTRACES sections as all the datasets are going through the process of intercalibration.

P4

- NADR is used only once in the text (with two of the letters inverted.)

→Done

- SANARCT does not appear elsewhere in the text according to the search that I did. Please remove the parentheses and delete SANARCT

→It does appear at page 8 line 23. As such we have kept it as it is.

- ENACW does not appear elsewhere in the text according to the search that I did. Please remove the parentheses and delete ENACW

→Done

- *L26 change “present low levels of O2” to “is depleted in O2”*

→Changed

P7

- *The surface distribution in Al is hard to understand from the figures. Figure 2 needs to use a split axis so that values between 0-10 nM can be seen. I suppose that a log axis would work, but that makes it hard for the reader to gauge what the actual Al values are. Without split axis this figure is not very useful and the fresh water and Iberian input can be summarized in a couple of sentences.*

L5-12 If we could see the data in the figure then this section would not be so painful to read

→We have changed the plot. We also have added biogenic silica. The shelf stations have been removed from this plot as they are shown in figure 3 and 4.

- *L19 “in line” should be changed to “consistent with”*

→Modified.

P8

- *L7 NADR not NARD*

→Changed

- *L7 ARCT not defined anywhere*

→Indeed, this should be SANARCT which is defined in Page 5 line 24. We have modified it to SANARCT.

- *Tonnard et al should be made available. You list an A and a B, but don’t list the citations in the bibliography.*

→Tonnard et al., (b) has now been published in biogeosciences discussion. We have added the citations to the bibliography.

- *L10 Figure 6 called before figures 3, 4, and 5.*

→ We have changed the figure numbers.

- *L14 High correlation in euphotic zone between dAl and SiOH4. Why is this not plotted in an x-y plot? Depending on the time of year (pre-bloom, post-bloom, pre-dust, post-dust), this plot could be meaningless. What about correlations with P*S*i and pAl?*

→The sentence was not clear. We made the clarification suggested by referee 1. We meant strong correlation with depth and not in surface waters. We have now added BSi data into figure 2 and commented on this.

Page 7, line 17-27: Elevated bSi and pAl concentrations and low dAl concentrations coincided with diatom dominated phytoplankton communities for the ENAB, IrB, and LB (Fig. 2). In contrast, elevated dAl and low bSi and pAl concentrations coincided with coccolitophore dominated phytoplankton communities for the IcB (Fig. 2). The latter could suggest a preferential scavenging of dAl by diatoms rather than by coccolitophorids. This conclusion is supported by increased pAl to dAl ratios where surface waters were dominated by diatoms (Fig. 5), probably as a consequence of active dAl incorporation into siliceous shells (Gehlen et al., 2002) or scavenging of dAl onto biogenic opal (Moran and Moore, 1988a). One way ANOVA analysis was performed for the pAl to dAl ratio in the surface waters (>50 m) in each of the four basins which showed strong dAl to Si(OH)₄ correlations with depth (LB, IrB, IcB and ENAB, see section 3.3.1).

P9

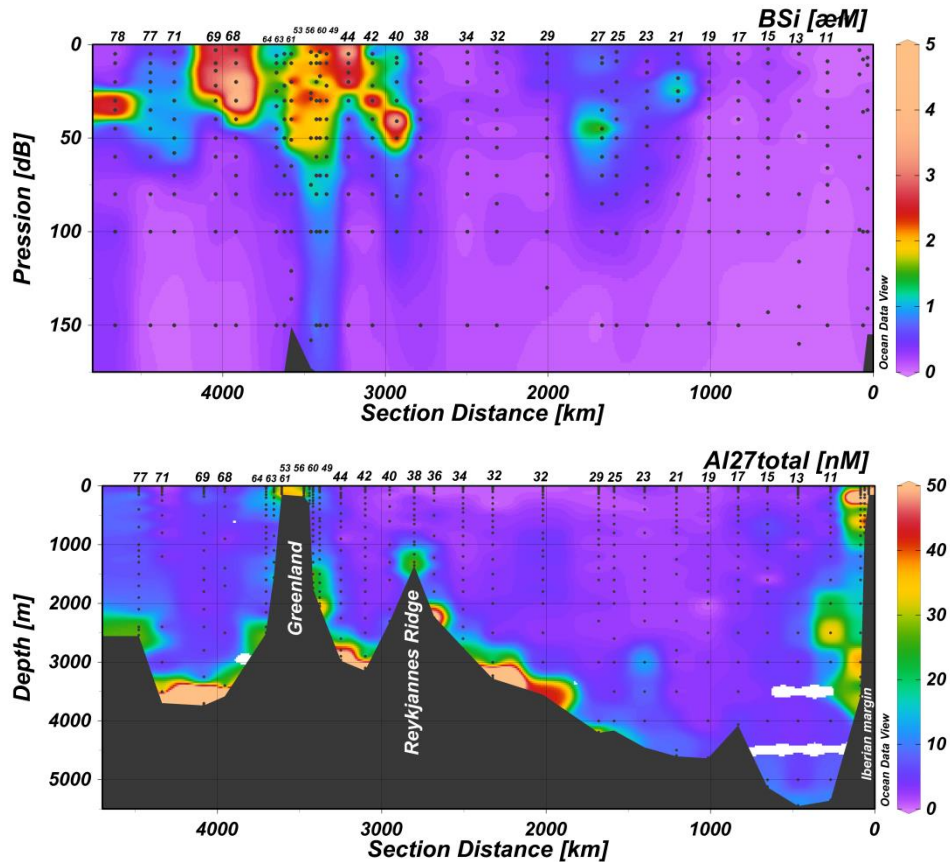
- “we conclude that the observed differences in pAl to dAl ratios were related to diatom abundance.”

→ We have modified the sentence as we are not able to say with 100% of certainty that the differences in pAl to dAl ratios are solely a function of diatom abundance.

Page 8 line 5-7: Considering the low aerosol deposition, reduced fluvial inputs into the basins (ENAB, IcB, IrB and LB), and elevated levels of bSi, we conclude that the observed differences in pAl to dAl ratios were mainly related to the abundance of diatoms.

- You don't present any biogenic silica data. You don't document that Al is not scavenged by other particles. As a result this appears to be mostly unsupported inference. With a combination of pSi, pCa, pAl and dissolved Al can we make an advance in our understanding of the marine Al cycle? Compared to many studies the combined use of particulate and dissolved chemistry can be a powerful tool.

→ Yes, the combination of dissolved and particulate datasets is one of the goals of large research programmes such as GEOTRACES and will help us to understand from an holistic point of view key questions regarding the Al biogeochemistry. However, it is sometimes impractical to present, interpret and combine all these datasets in one publication. We do not present BSi and pAl in this manuscript as there are going to be published by other authors within the GEOVIDE special issue. pCa data is not available for this section. However, we do have permission from the data owners to show the section plots of pAl and bSi (see below). The pAl and BSi sections are being published in the GEOVIDE special issue by Gourain et al., and Sarthou et al.,.



It is to notice that BSi was analysed from samples collected from the regular CTD and not from the trace metal clean CTD. Thus, the sampling depths do not always coincided between both CTDs.

- Can you say anything about the role of coccolithophores? The LeMaitre paper documents their abundance along your section. If you could discount their role in the scavenging of Al from the surface ocean, that would be a very novel finding. Is there a novel or transformative result here? So far your manuscript repeats what others have found and the results seem mostly as expected. I would think that other classes of particles would scavenge Al, especially large quantities of particles produced during phytoplankton blooms independent of the type of phytoplankton. However, perhaps my thinking is incorrect. Can your data address these questions?

→ We suspect that dAl is scavenged by different types of particle and that the amount of scavenged dAl will increase as a function of the amount of each particle class present. As such, the North Atlantic Ocean represents a key area to investigate this process. However, it can be difficult to draw correct conclusions with just this study as the strong phytoplankton blooms in

the North Atlantic are dominated by diatoms and after diatom decay, by coccolithophorids. It would be really interesting; as such we have added a sentence in the conclusions section, to investigate the distributions of dAl and pAl along with bSi and pCa during the duration of a phytoplankton bloom.

Diatoms, potentially, scavenge more efficiently dAl from surface waters than other phytoplankton types (e.g. coccolithophorids). The reason is that dAl is adsorbed onto diatom frustules (passively) and/or incorporated Al into the structure of frustules (Whether this is an active or passive mechanism remains unclear).

CHEMTAX data are published by Manon Tonnard and co-authors in the special issue. Based on the CHEMTAX data, diatoms were the dominant phytoplankton community in the ENAB (40%), IrB (>60%), and the LB (>60%). In contrary, in the IcB coccolithophorids dominated the phytoplankton community (50%). From the CHEMTAX data it is clear that any attempt to disentangle the relationship that coccolithophorids and diatoms exert on the behaviour of dAl and pAl in surface waters would be challenging as the phytoplankton community composition was, in all the different basins, highly variable.

The enhanced levels of bSi in the SANARCT region (IrB and LB) indicate the presence of diatoms which is confirmed by the CHEMTAX data. Based on the ANOVA analysis, the ratio of pAl to dAl was significantly different for the IrB and LB compared to the Iceland and East North Atlantic Basins. In contrast, no significant difference in the ratio for pAl to dAl was found between the IcB and ENAB.

Page 11 lines 29-34: However, it should be noted that dAl only removal by diatom production could not be solely reason for the nutrient type distribution. Although we did not observed enhanced pAl concentrations were coccolithophorids were dominant (e.g. IcB; section 3.2.2) we assume that also other biogenic and non-biogenic particles (e.g. CaCO₃, organic carbon, lithogenic particles) as well as zooplankton fecal pellets will contribute to the vertical export of surface dAl into the deep ocean.

- *L7 should you reference figure 2 here as well?*

→We have also included figure 2.

- *L7-8 “which decreased westwards to 5.6 nM (station 11).” I would consider changing this to “which decreased westwards, reaching 5.6 nM at station 11.”*

→Changed.

- *L16 is there any relevance to the estuary being polluted? The more important point is that rivers can be large sources of dissolved Al (Brown and Bruland papers plus another one that you reference here.*

→We agree with you. We have removed the term polluted. We have added additional references.

Page 9 line 3-6: Our results indicate that a fraction of riverine dAl can be advected offshore, as observed previously in the Bay of Bengal (Grand et al., 2015), Gulf of Alaska (Brown et al., 2010), and coastal waters of Oregon and Washington (Brown and Bruland, 2009),....

- *P10 L3 “were” not “was”*

→The discussion in this paragraph has been rewritten according to comments from Rob Middag (Referee report 2).

P11

- *L7 -10 “Increased aerosol deposition was observed during GA03 (Shelley et al., 2015) in comparison with GEOVIDE (Shelley et al., 2017). Thus, enhanced dAl for GA03 could be related to enhanced removal of dAl in surface waters and concomitant remineralization of biogenic particles following the decline of the late summer-autumn bloom.” It seems that if you had more aerosol deposition during GA03 then you would expect higher dAl and remineralization is not required. Why do you make this point? What have I missed?*

→ We have rewritten these sentences for clarification. We argue that the accumulation of dAl in surface waters from atmospheric aerosol deposition during June, July, August, and September and the removal occurring during the bloom period (months) with dAl subsequently released at depth may have been the possible source for the enhanced concentrations observed. However, we still state at the end that this remains speculative.

Page 10 line 22-27: The region is known to receive enhanced atmospheric aerosol deposition (Mahowald et al., 2005). A possible explanation for this difference may therefore be that dAl accumulates in surface waters during June to September. Then, following the late summer-autumn bloom in the Iberian Basin, the excess of dAl accumulated in surface waters over summer may be scavenged by the increasing amount of biogenic particles produced by the bloom with release in sub-surface waters

as these particles are remineralized. However, this possible explanation remains speculative.

- L8 “in surface waters” should be “from the surface waters”

→ Changed.

- L10 give depth range of “sub-surface”

→ It was given some lines before in the same section. Page 10 line 7.

- Section 3.3.1 Need to mention scavenged type distribution in Pacific. Certainly Orians and Bruland and Brown and Bruland papers think about their data in terms of scavenging and not as nutrient-like. Also may need to take a closer look at the Barrett et al and Measures 2008 in terms of this question (not sure which of these two).

→ We have added that dAl in the Pacific Ocean shows a scavenged type distribution.

Page 11 line 8-9: A scavenged type distribution for dAl has also been described for the Pacific Ocean (Orians and Bruland, 1985).

P12

- L1 : “this vertical distribution coincides” should be “these distributions coincide”

→ Corrected.

- L3 Moore and Millward not in bibliography

→ Corrected.

- L3-5 it is not clear to me that Orians and Bruland nor Middag et al actually showed that dAl was removed onto particle surfaces. Both sets of authors surmised this from their data but did not demonstrate it. Middag et al 2009 summarized other people’s work that made some attempt to look at particulate phases, but Middag et al 2009 did actually not report on particulate chemistry and thus can only surmise that the dAl was moved onto particles. Thus you need to be careful in your references that you actually reference those papers that truly demonstrate your point. Although Middag et al 2009 has a nice summary of some of those papers, he is a secondary and not a primary source.

→ We have rephrased that part.

Page 11 line 14-17: Dissolved Al is considered to be removed from surface waters onto particle surfaces (Moore and Millward, 1984; Orians and Bruland, 1985), including diatom cells (Gehlen et al., 2002) and subsequently released at depth during the recycling of biogenic particles and desorption from non-biogenic particles.

- L7-8 you seem to imply that diatoms are the only surface that scavenges Al. Moran and Moore demonstrate that Diatoms are an important scavenger of dAl but do not address other substrates.

→ We have modified the sentence.

Page 11 line 17-20: In our study region, diatoms dominate the phytoplankton communities at the early stage of the spring bloom (Brown et al., 2003), and are an important producer of biogenic silica (bSiO₂) (Nelson et al., 1995) which is, along with other non-biogenic particles, a major carrier for scavenged Al (Moran and Moore, 1988b; Stoffyn, 1979).

- *Are “Lemaitre et al 2017” and “Lemaitre et al this issue” the same papers? IF so I can’t find the biogenic Silica in that manuscript. The full bSiO₂ data set would inform this manuscript.*

→ No. This was a mistake. They are two different manuscripts. The bSi is going to be presented in Lemaitre et al (b). It should be soon available on BGD as part of the special issue.

- *L16-17 “suggests that the net remineralization of dAl from particles was larger than the net removal of dAl from scavenging.” This implies that dAl should be building up over time or that as the water moves away new water is continually fortified by new remineralization.*

→ Correct.

- *The idea that remineralization is a dominant process is a key one in understanding Al biogeochemistry and it would be nice to have all of the data to properly demonstrate it. It really seems odd that the particulate Al and pSi are not a part of this manuscript. Are there changes in the ratio of pAl to dAl at different depths?*

→ The ratio between pAl and dAl decreased, generally, with depth. However, close to the seafloor and due to resuspension of sediments the pAl to dAl ratio increased again. This can be observed in Fig. 5 c.

- L27 sentence about figure 7 does not make sense.

→ We have rewritten the sentence and took out the word correlations.

Page 12 lines 14-17: Figure 7 displays dAl & pAl versus salinity in the MOW for the neutral density surface layer (σ_n) between 27.6 and 27.8 kg m⁻³, corresponding to a MOW core depth, based on highest salinity values, between 1000 and 1200 m.

P13

- L14 need a reference for Al in pore waters. In doing this review I came across Stoffyn-Egli 1982, but I would love to know if you are aware of anything else.

→ Added. Another reference is Van Beueskom et al. (1997)

- L19 Sherrell and Boyle do not discuss dissolved Al.

→ We have removed this citation.

P14

- L1. EGC is used 2 times in the text and once in the figures. Thus you should use “East Greenland Current (EGC)” each time. WGC is only once in the text and once in the figures. Also WGC is not defined when it is used in the text.

→ We have added the full name for the abbreviations mentioned.

- L14 might consider using beam attenuation instead.

→ Considered and changed as suggested.

- L20 light transmission is nice, but particulate Al would be much much better. Perhaps the PAI is from pumps and not from the same samples? Still adding pAl would be helpful.

→ A similar issue was raised by Dr. Middag’s review. We have now included a plot in the SI (Fig 3SI) showing the distribution of dAl and pAl with depth for station 2 and station 78 over the Iberian and Newfoundland shelf, respectively. The pAl data were analysed from samples collected using the trace metal clean CTD.

- L25 do you mean that dissolution/ remineralization from particles rather than partial dissolution of resuspended sediments? Do you mean a dominant process in supplying dAl to the mid and deep ocean?

→No, we meant that scavenging of dAl by resuspended particles may be dominating over Al dissolution from resuspended particles and that this may be the reason why no significant enhanced dAl concentrations are observed when enhanced pAl concentrations are present. We have rewritten this part and added a sentence for clarification

Page 14 line 3-5: These results suggest that occasionally scavenging of dAl onto resuspended particles may dominate over the partial dissolution of Al from resuspended sediments. However, the mechanisms controlling either a net dissolution or net scavenging of Al from resuspended particles remain unclear.

- L26 should be “general increase in Al”

→Changed.

P15

- L11 use “were” not “was”

→Corrected

- *Figure 1 NAST, NADR, ARCT should be placed within or above boxes instead of letters A, B, and C. All these acronyms are confusing enough, but then adding tertiary relationship makes it very painful to those of us who don't know the region. The reader should be able to look at the figure and understand what is being said. The ideal figure is one in which the reader only needs to read the figure caption once or not at all. It would be good for the basins to be listed as well. For example you list box B as NADR but often talk about the Iceland Basin. It would be helpful for your readers to look at the map and see where the Iceland and other basins are.*

→We have redone the figure.

- *Figure 2. the scale makes it so that the reader cannot see all of the data. Please use a split axis. There is a lot of discussion in the manuscript about these data and I cannot see the features. This is the only way for a reader to assess the meaning (and the quality??) of the data. IF you need to*

make a separate panel, then it is worth it. Need to show station 60 in this figure because it is shown in figure 5.

→ We have redone the figure. We now show all stations except the ones over the Iberian and Greenland shelf. We have included biogenic silica in this plot. The Iberian and Greenland shelf stations are shown in figures 3 and 4.

- *Figure 3. Very nice figure. Is there anyway to stretch the color bar to let us see how dAL at stations 11-17 changes. This can also be accomplished by making the suggested changes to figure 2.*

→ Thanks. It has been changed on figure 2.

- *Fig 6 need label on color bar. A separate section panel covering 0-500m is needed.*

→ Changes done.

- *Fig 6b I can't read any of the fonts on this figure. Increase y-axis font and decrease label interval from 5 to 10. Need to find a way to make x-axis readable as the font is tiny. Inside each of the profile boxes at the top of the figure are labels that could only be read with a zoom. In your figure, going left to right (the way we read) the literature data are in orange, green, and then blue. In your figure caption, these boxes need to be discussed in that order too.*

→ Changes done. Profile boxes are now figure 6c.

- *F6c label needed on color bar with units*

→ Figure 6c is now figure 5. Changes done.

- *Figure 8 Need pAl on this plot or on a parallel panel*

→ We have added a new plot for pAl. Figure S6.

References:

Brown, M. T. and Bruland, K. W.: Dissolved and particulate aluminum in the Columbia River and coastal waters of Oregon and Washington: Behavior in near-field and far-field plumes, *Estuarine, Coastal and Shelf Science*, 84, 171-185, 2009.

Brown, M. T., Lippiatt, S. M., and Bruland, K. W.: Dissolved aluminum, particulate aluminum, and silicic acid in northern Gulf of Alaska coastal waters: Glacial/riverine inputs and extreme reactivity, *Marine Chemistry*, 122, 160-175, 2010.

Gehlen, M., Beck, L., Calas, G., Flank, A.-M., Van Bennekom, A., and Van Beusekom, J.: Unraveling the atomic structure of biogenic silica: evidence of the structural association of Al and Si in diatom frustules, *Geochimica et Cosmochimica Acta*, 66, 1601-1609, 2002.

Grand, M. M., Measures, C. I., Hatta, M., Hiscock, W. T., Landing, W. M., Morton, P. L., Buck, C. S., Barrett, P. M., and Resing, J. A.: Dissolved Fe and Al in the upper 1000 m of the eastern Indian Ocean: A high-resolution transect along 95°E from the Antarctic margin to the Bay of Bengal, *Global Biogeochemical Cycles*, 29, 375-396, 2015.

Hydes, D., De Lange, G., and De Baar, H.: Dissolved aluminium in the Mediterranean, *Geochimica et Cosmochimica Acta*, 52, 2107-2114, 1988.

Hydes, D. J.: Seasonal variation in dissolved aluminium concentrations in coastal waters and biological limitation of the export of the riverine input of aluminium to the deep sea, *Continental Shelf Research*, 9, 919-929, 1989.

Koning, E., Gehlen, M., Flank, A.-M., Calas, G., and Epping, E.: Rapid post-mortem incorporation of aluminum in diatom frustules: Evidence from chemical and structural analyses, *Marine Chemistry*, 106, 208-222, 2007.

Mahowald, N. M., Baker, A. R., Bergametti, G., Brooks, N., Duce, R. A., Jickells, T. D., Kubilay, N., Prospero, J. M., and Tegen, I.: Atmospheric global dust cycle and iron inputs to the ocean, *Global Biogeochemical Cycles*, 19, 2005.

Middag, R., de Baar, H. J. W., Laan, P., and Bakker, K.: Dissolved aluminium and the silicon cycle in the Arctic Ocean, *Marine Chemistry*, 115, 176-195, 2009.

Middag, R., Van Hulst, M., Van Aken, H., Rijkenberg, M., Gerringa, L., Laan, P., and De Baar, H.: Dissolved aluminium in the ocean conveyor of the West Atlantic Ocean: effects of the biological cycle, scavenging, sediment resuspension and hydrography, *Marine Chemistry*, 177, 69-86, 2015.

Moore, R. M. and Millward, G. E.: Dissolved-particulate interactions of aluminium in ocean waters, *Geochimica et Cosmochimica Acta*, 48, 235-241, 1984.

Moran, S. and Moore, R.: Temporal variations in dissolved and particulate aluminum during a spring bloom, *Estuarine, Coastal and Shelf Science*, 27, 205-215, 1988.

Orians, K. J. and Bruland, K. W.: The biogeochemistry of aluminum in the Pacific Ocean, *Earth and planetary science letters*, 78, 397-410, 1986.

Orians, K. J. and Bruland, K. W.: Dissolved aluminium in the central North Pacific, *Nature*, 316, 427-429, 1985.

Stoffyn, M. and Mackenzie, F. T.: Fate of dissolved aluminum in the oceans, *Marine chemistry*, 11, 105-127, 1982.

Van Beusekom, J., Van Bennekom, A., Tréguer, P., and Morvan, J.: Aluminium and silicic acid in water and sediments of the Enderby and Crozet Basins, *Deep Sea Research Part II: Topical Studies in Oceanography*, 44, 987-1003, 1997.

Vrieling, E. G., Poort, L., Beelen, T. P., and Gieskes, W. W.: Growth and silica content of the diatoms *Thalassiosira weissflogii* and *Navicula salinarum* at different salinities and enrichments with aluminium, *European journal of phycology*, 34, 307-316, 1999.