

This is a well written manuscript that reports on a beautiful data set from the Geovide expedition. The section of ocean covered by this work is important to understanding the Al budget in the Atlantic Ocean. It is clear that this manuscript should be published with a moderate amount of work. However, I would say that my reading of the manuscript reveals no new major thoughts about the biogeochemical cycle of Al in the ocean. The concepts that I thought to be the most original were mentioned in passing. The concepts that are important for the cycle of Al were not addressed in a new way, but instead repeated what has been said by others. In the literature on the Al cycle there are many inferences about the role of scavenging of dAl and its release with depth. Most of these publications fail to consider both dissolved and particulate phases for silica and aluminum and are thus left to inferring processes that may or may not be taking place. It would appear that full data sets for particulate Al and Si are available from Geovide. Co-author Gourain should allow his full pAl data set be used in this publication. I assume that pSi was measured and including both P_{Si} and P_{Al} that would be an incredible contribution to understanding the role of diatoms in scavenging dAl in this region. The small bits pAl that are reported are intriguing, but ultimately their absence hurts the full interpretation of dAl. In addition the Gourain et al, manuscripts is not available for me to look at. This is also true for the Tonnard et al manuscripts (A and B); the LeMaitre Ba manuscript contained some of the insight that I would have wanted from Tonnard et al.. This manuscript references Lemaitre et al for bSi, but the Lemaitre manuscript associated with this special issue does not show any biogenic Si. Particulate Si and Al would really make this a unique contribution to the literature on the Al cycle in the oceans. Despite the number of data sets about dAl in the literature only a few of them make use of particulate data to bolster the arguments about scavenging, removal, and remineralization.

Overall I am comfortable with the general concepts presented in the paper. I think they have a unique data set, however I think that their data set should allow them to say more than they do. This manuscript represents a great opportunity if pAl and pSi are considered. As written, it is a missed opportunity and does not appear to say anything different than what has been said before. It is still publishable and is still a nice contribution. Another opportunity for this paper, is to address the role (if any) of coccolithophores in scavenging Al. The LeMaitre paper documents the abundance coccolithophores and diatoms along this section. Do you have pCa data as well? If you could discount or demonstrate the role of coccoliths in the scavenging of Al from the surface ocean, that would be a novel finding. It seems that large quantities of particles produced during phytoplankton blooms should scavenge Al independent of the type of phytoplankton. However, perhaps my thinking is incorrect. Can these data address this question? A combination of pSi, pCa, pAl and dissolved Al would advance understanding of the marine Al cycle. (Do pSi and pCa data exist?). Barrett et al 2013 has bSi that supports these findings and Barrett et al 2014 has pCa. I think Barrett et al concentrated on dust more than the Al cycle.

In the end, it is important that this paper is not be published until the pAl and pSi (or biogenic silica) data are available on line. Preferably this manuscript would present the pAl and pSi.

I have a long list of details below. Most are pretty minor. Overall I find that there is a lot of incorrectly cited work, the occasional typo, and some places where clarity can be improved.

P2

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

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.

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).

Hesse et al only talk about the process of forming nepheloid layers and do not discuss Al, so it seems like a strange reference to me.

L27 Probably worth defining active and passive scavenging?

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.

Also, does this section preclude the removal of Al by particles that are not biogenic Si? Can Al be scavenged by other particles? Must it be incorporated into the frustule or can it be on the organic matter?

P3

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

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

P4

Do these bottles have polypropylene caps? This should be pointed out. I admit that most of us know the 125mL bottles from Nalgene have PP caps, but because PP caps probably contaminate for Al, this information should be included in your methods.

L26 Emission and Excitation wavelengths are backwards and the emission wavelength seems very low.

P5

L1 what is “low trace metal sea water”? perhaps you can define in terms of Al concentrations.

L10 “was” should be “were”

P6

L5 I am curious if you truly did a winkler titration or if you used an electrode and automated titration? Just fine either way, but few people are doing Winklers these days.

NAST is used once in the text and once in a figure; consider just using North Atlantic Subtropical in the text for second usage.

Define NADR in figure 2 as is done in figure 1.

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

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

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

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

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

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

P8

L7 NADR not NARD

L7 ARCT not defined anywhere

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

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

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 pSi and pAl?

P9

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

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? 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.

L7 should you reference figure 2 here as well?

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.”

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.

P10

L3 “were” not “was”

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?

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

L10 give depth range of “sub-surface”

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).

P12

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

L3 Moore and Millward not in bibliography

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.

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. 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.

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.

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?

L27 sentence about figure 7 does not make sense.

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.

L19 Sherrell and Boyle do not discuss dissolved Al.

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.

L14 might consider using beam attenuation instead.

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

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?

L26 should be “general increase in Al”

P15

L11 use “were” not “was”

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.

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.

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.

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

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.

F6c label needed on color bar with units

Figure 8 Need pAl on this plot or on a parallel panel.