

Specific comments:

- *Page 3 line 28 why were two different filters and pore sizes used and is there a difference between them and the Al fraction analysed?*

→ During some cruises we filtered the samples with capsule filters (Sartobran 300, Sartorius) and sometimes the samples were filtered with the filters used to collect the particulate trace metals (Supor®, Pall Gelman). No, there is no difference and this approach is common on a lot of the GEOTRACES cruises as water use is optimized to allow collection of filtered seawater and particulate metals. Both filtered fractions are considered as the dissolved fraction (www.geotraces.org/).

- *Page 4 line 22 Brown and Bruland used a 4 M buffer. Is there a reason for changing the concentration?*

→ Indeed, this is a slight modification of the method published by Brown and Bruland. We have added a sentence to clarify this. The important aspect was to reach an optimal reaction pH between 5.2 and 5.6 which was also reached with the 2M buffer. Another advantage is that you can prepare a single buffer for the Al analysis instead of having to prepare two different buffers.

Page 4, line 2-4: A slight modification of the method published by Brown and Bruland (2008) is the use of a 2 M ammonium acetate buffer (UpA, Romil), instead of 4 M, in the reaction stream.

- *Page 5 line 5 What is the effect on the sample pH when adding 3 times the required amount of buffer and the subsequent pre-concentration? I find the blank determination slightly odd. If I understand it correctly, 'regular' samples are buffered online, but for the blank determination a sample was buffered offline with varying amounts of buffer. Additionally an acidified MQ sample was analysed without buffer (so I assume the system had to be modified, i.e. the buffer line was removed?). Wouldn't it be far easier and more representative to analyse an acidified MQ sample as a regular sample? With the current approach, any blank resulting from the online buffering (if any) is not accounted for.*

→ Adding three times the required buffer will elevate the sample pH upon pre-concentration. However, in the improved method published (Brown and Bruland, 2008) it is shown (Fig. 2) that at a pH above 5 the effect of sample pH on column retention of Al is negligible. Indeed, the buffer line is "closed" for the blank determinations. The manifold blank was determined running an acidified seawater samples (not an acidified MQ sample) without buffering. Running an acidified MQ sample would be different in terms of sample matrix. Yes, it could be far easier to proceed as suggested, but overall it looks that the differences which arise from our procedure are relatively small or absent as the analysis were certified by the run of GEOTRACES reference seawater and internal standards. However, for future analysis we will bear in mind this recommendation.

- *Line 8 the average blank and standard deviation should be reported*

→ Done.

Page 4, line 24-25: This blank (Average (+/- standard deviation) blank = 0.23 nM ± 0.1 nM; n = 28) was subtracted from the results obtained.

- *Line 17 I would say similar analytical techniques in different labs and cite the paper (Rolison et al., 2015) for the other dataset. Here it should probably also be pointed out that one dataset was analysed shipboard, the other after storage for some time in a shore based lab and that there appears to be no difference between the approaches (At the GEOTRACES website there is a cautionary note accompanying the SAFe reference sample results warning there could be Al contamination from bottle caps during storage).*

→ We have changed this information and clarified the procedure.

Page 4, line 33-35 and page 5 lines 1-2: the GEOTRACES section GA04N (Station 1, 39.73 °N, 14.17 °W) (Rolison et al., 2015). Dissolved Al was analysed, based on the same method, using similar analytical techniques in different laboratories. It is noteworthy that dAl samples for GA04N were analysed on-board ship and for GA01 after prolonged storage at a later date. Samples analysed for GA01 were stored up right in order to minimize any contact and potential contamination arising from the polypropylene caps.

- *Page 7 line 25 room for clarification here; in the previous paragraph decreasing concentrations from eastern to western basins were described, so I guess the point here is that there is no significant difference? Also the decrease would not only be expected in the east-west direction, but also in the south north direction (away from Saharan dust source).*

→ Clarifications made. We have added a sentence clarifying that within errors no significant difference was observed. We have also added that atmospheric aerosol loading decreases in a westward and northward direction from the main aerosol source (e.g. Sahara).

Page 6, line 21-22: However, within analytical errors, no significant difference was observed between the basins.

Page 6, line 29-30: In the North Atlantic atmospheric aerosol loading declined in a westward and northward direction, i.e. with increasing distance from the major African dust source regions....

- *Section 3.2.2 What stations are used in the calculations? Probably the coastal stations should not be used here as particle concentrations and compositions here are influenced by continental and sediment sources as discussed in subsequent sections. Additionally, this section warrants some further discussion on the use of Al as a dust tracer, as this study implies the dissolved Al concentration is not only dependent on atmospheric deposition, but also the presence of diatoms.*

→The following point was addressed also by referee 1. We have added the stations numbers. See referee report 1. Yes, you are right. We are preparing a manuscript on the role of dAl as a dust tracer using surface mixed layer dAl concentrations from GEOVIDE, GA06, GA08, and GA10 cruise. Therefore we have not included a discussion on this topic.

- *Section 3.2.3 I'm not 100% convinced based on the current discussion the elevated Al is associated with river outflow; how can one be sure e.g. it is not all wet deposition or that sediment resuspension on the shelf doesn't play a role too (after all, a sediment resuspension source is argued as a significant source in section 3.4.2). Was there a correlation between dAl and salinity or other tracers of fluvial input (as for example observed in the Drake Passage for Al input associated with land run off (Middag et al., 2012))? The authors have the data to discuss this in more depth and to discuss the importance of fluvial input vs deposition and resuspension (the Al could be effectively removed but later re-suspended as suggested for the Californian shelf for Fe by Bruland and co workers).*

→We extended the discussion about this topic. Based on the data we have, the resuspension of shelf sediments is unlikely to be an important source to surface waters. We have added a new figure in the SI which shows that sediment resuspension is a source for bottom waters, but not a source for surface waters. Wet deposition is also unlikely to be the main source of dAl as the shape of the salinity profiles for stations 1 to 4 and for station 11 are different. Thus, we still consider that the river input is the main source for the elevated dAl concentrations observed.

Page 8, line 18-35. Possible sources which could explain the elevated surface dAl concentrations observed are shelf sediment resuspension, wet deposition, and riverine inputs. Shelf sediment resuspension is unlikely to be the source for the elevated dAl concentrations as deep profiles for stations 2 and 4 (Fig. S1) showed that the elevated levels of dAl observed in bottom waters were not a source for surface waters since minimum dAl values were observed between maximum surface and deep dAl values. Salinity profiles for GEOVIDE showed salinity minima (<35) in surface water for stations 1, 2, and 4 (Fig. S2), indicating a freshwater source. No evidence of freshwater input was observed at station 11, located just west of station of 1. Possible explanations for the observed distribution in salinity are therefore wet deposition and/or river inputs. Wet deposition events were registered in the region between stations 1 and 4 (Shelley et al., 2017a). Yet, the shape of the salinity profiles of stations 1 to 4 were unlikely caused solely by recent wet deposition as the differences in salinity, in comparison with station 11, were observed up to a depth of 45 m. GA01 ship's ADCP data (Fig. S3) showed that surface waters near the Iberian Peninsula flowed in a northward direction. Therefore, we suggest that the additional source of dAl to surface waters originated from the Tagus estuary, a polluted estuary which accordingly likely exhibits high dAl concentrations, (Cotté-Krief et al., 2000), located approximately 175 km south from stations 1, 2 and 4. Elevated concentrations of dissolved Fe in the Tagus outflow and strong correlations for salinity against dAl and dFe (Tonnard et al., 2018) observed during the GA01 cruise supports a riverine source of dAl.

- *Section 3.2.4 I find the argumentation in this section a bit shaky as detailed below. I do not disagree with the point reached, but the argumentation needs to be improved Line 7-10 it is probably worthwhile to mention these endmember estimates are conservative for dAl as they do not incorporate any Al scavenging or precipitation. Is that also the reason for pooling the dAl and pAl later in the paragraph when discussing endmembers?*

→ We have clarified that the endmember estimates for dAl are conservative. No, there was no specific reason.

Page 10 Line 10 I do not see how samples from a fjord and an iceberg are representative for glacial runoff and sea ice melt. Sea ice melt will be completely different (see also next comment) and the concentrations in the fjord will not only depend on run-off, but also the interaction with sediments and (biogenic) particles in the water column

→ Biogenic particles are not a major issue for low-salinity samples from inner-fjord environments, these environments are invariably light limited, low productivity environments (chlorophyll a in the region where these samples were collected was < 0.25 mg L⁻¹), but of course fine sediment is also a source of trace elements in these near-shore environments which is difficult to 'uncouple' from freshwater input. The interaction with sediments is a point we now address. We better distinguish between these sources in R1 and state the inevitable limitation that overlap between these sources makes it difficult to distinguish between them, but note that the high pAl signal we observed requires a terrestrially derived source.

Line 12-13 why would sea ice Al concentrations be more similar to ice berg than glacial run off? Ice bergs were once part of the glaciers, so wouldn't one expect that ice bergs and glaciers are quite similar and sea ice very different from those two?

→ Meltwater/runoff acquires, by marine standards, massive concentrations of dAl and pAl in pro-glacial streams or in shallow coastal areas around Greenland where sediment plumes are present throughout the year. Thus the pAl signal 'delivered' into the marine environment is much higher than that arising in ice melt itself. Hence we expect that the runoff derived signal will have a much higher pAl and dAl content than either ice-melt or sea-ice melt due to the greater sediment-water interaction, this is verified by considering the range of dAl and dFe data reported in prior work for these sources.

Line 14-15 the comparison between total dissolvable and dAl+pAl is not valid in my opinion, the total dissolvable is a 'gentle leach' at pH 1.8 whereas the pAl analysis is a complete destruction.

→ We now explicitly acknowledge in the text the difference between pAl and tdAl, but this does not change our finding that the relative contribution of different sources which is the basic point made.

Page 9 line 19-35 and page 10 line 1-7: Freshwater endmembers (salinity 0) for Al were determined from linear regressions between dAl, pAl and salinity for the eastern (stations 49, 53, 56 and 60; dAl 60.5 ± 9.9 and pAl 773.7 ± 125.6) and western (stations

61, 63 and 64; dAl 6.2 ± 1.2 and pAl 675.1 ± 124.7 nM) on the Greenland shelf (Table 1). These endmember estimates will be considered conservative as they do not incorporate Al scavenging processes. To gain some insight into what sources may have contributed most strongly to our high Al signals in surface waters off the Greenland shelf, we analysed a collection of fjord and iceberg samples from west Greenland. Mean total dissolvable Al (unfiltered) iceberg and fjord concentrations were 55 ± 2 nM and 12.8 ± 6 μ M, respectively (Table S1). Freshwater Al endmembers (dAl + pAl) derived from our shelf stations were an order of magnitude higher than the mean tdAl measured in iceberg samples. tdAl must by definition be < pAl due to the weaker leaching procedure applied. Yet given the large difference, the Al values off Greenland appear to be related to the input from terrestrial runoff enriched with glacially derived sediment with this enrichment occurring either downstream of glaciers in pro-glacial environments or in near-shore environments where sediment plumes can result in high trace element concentrations throughout the year (Hopwood et al., 2016). This is consistent with a similar elevated Fe signal on the Greenland shelf (BGD). Similar observations of elevated Al were made in Cumberland Bay, South Georgia (Schlosser et al., 2017), and attributed to suspended glacial flour as the main source for enhanced pAl concentrations. An alternative low salinity dAl signal could come from sea-ice, which contributes a total freshwater input to the EGC approximately equal to that of terrestrial runoff. Yet, whilst sea-ice dAl concentrations are not available in this study/region, Lannuzel et al. (2011) report median dAl and pAl concentrations in sea ice (pack ice) of 2.6 and 10.7 nmol L⁻¹. We therefore anticipate that local ice-melt (from sea-ice and icebergs) was a minor contributor to the shelf dAl signal compared to terrestrial runoff.

- *section 3.3 title of this section could be improved, section 3.2 has an informative title and deals with surface water, what is this section about and what distinguishes it from section 3.2 (as that section also deals with spatial distributions)*

→We have modified the title of section three. Section 3.3 deals with the vertical dAl distribution during the GEOVIDE cruise while section 3.2 deals with the surface dAl distribution.

Page 10 line 9: Overview of the water column distribution of dAl

- *Section 3.3.1 I think this section should be better linked to section 3.2.2. Is there anything that can be learned from (changes in) the particulate phase in deeper waters in this region? And an increase with depth is not only related to local (vertical) processes, but also processes during advection of deep water masses and mixing of water masses with different pre-formed concentrations. The latter should also be explored/discussed as a potential driving factor behind the observed correlations. Notably in the next section, sediment re-suspension is discussed which also lead to increased concentrations at depth so a better linking of section 3.3.1 with 3.4.2 is also warranted.*

→We have added some sentences linking sections 3.2.2 and 3.4.2 with section 3.3.1. We have added the potential factor of advection/mixing and sediment resuspension in controlling the dAl water column distribution. We are not sure about the point made with “anything that can be learned from the particulate phase in deeper waters in this region”. We have added a plot in the comments to reviewer 3 showing the distribution of the pAl to dAl ratio with depth. However, it is to note that the focus of this paper is dAl and not

the particulate phase. The particulate phase will be discussed in Gourain et al., this issue.

Page 11 line 35 and page 12 line 1-4: It is also relevant to notice that advection of water masses and mixing of water masses with different pre-formed dAl concentrations could also have an influence on the water column distribution of dAl. The latter is observed in regions of enhanced sediment resuspension (See section 3.4.2) and water masses with low dAl concentrations (e.g. DSOW) in comparison with overlying water masses with elevated dAl levels (e.g. ISOW; see section 3.4.2).

- *Section 3.4.2 Page 13 Line 4-5 how does a positive correlation between pAl and salinity indicate the salty MOW is depleted in pAl?*

→ I guess, you wanted to mention section 3.4.1. The point we wanted to make was that pAl concentrations seem depleted in comparison with dAl concentrations. We have rephrased the sentence to avoid confusion.

Page 12 line 19-21: The correlations shown in Fig. 7 and the dAl profile of station 11 (Fig. S4) indicate that the MOW is enriched in dAl and represents a major source of dAl to mid depth waters in the North Atlantic.

- *Line 9 Do we need another tracer for MOW? Conventional tracers such as salinity that are far easier to measure than Al seem to work well, so what is the benefit of the relatively expensive and challenging parameter Al?*

→ No, of course not. Salinity is an “easy” to measure parameter which perfectly traces the MOW. The reason behind measuring Al is not the use as a tracer of water masses (Although this has been acknowledged before (Measures and Edmond, 1990)), it is the importance of Al as an indicator for lithogenic inputs. As such, it is included as key trace metal in the GEOTRACES project.

- *Line 28 were there more stations on the shelf without elevated concentrations? Currently not clear.*

→ Yes. Station 56 over the Greenland shelf did not present any significant elevated dAl concentrations in the most bottom sample.

- *Page 14 line 3-8 not directly clear station 78 is on the shelf and how deep the seafloor is in relation to the mentioned 140 m. . And how does one explain the enhanced pAl levels in the absence of enhanced dAl? (discussed later I noticed, maybe move this discussion forward or mention it will be discussed later)*

→ We have clarified which station corresponds to the Newfoundland shelf and the depth at which the seafloor is found. We have added a sentence on the probable cause for not seeing elevated dAl when elevated pAl is found.

Page 13 line 12-17: In contrast, on the Newfoundland shelf and margin (St. 78), no enhanced dAl concentrations were observed near the seafloor (Fig. S5). However, a large input of pAl was observed (station 78), and pAl concentrations increased from 94.6 nM at a depth of 140 m to 550 nM near the seafloor (377 m) (Gourain et al., this issue). Dissolved Al could be scavenged by resuspended particles, thus showing lower levels when elevated pAl levels were observed.

- *Section 3.4.3 this section is very brief. Perhaps some discussion as to why enhanced dAl has been observed at two hydrothermal locations (one very close by) but not at this location or numerous other active hydrothermal vent sites?*

→ Yes, you are right. This section is brief. The reason behind it is that we do not have any prove or complementary data to argue along this section. Thus, any more words would be just a merely speculation. A more detailed discussion on the possibly hydrothermal source of trace metals is given in Tonnard et al. (2018) and Gourain et al., special issue.

Technical corrections:

- *abstract last sentence of abstract is confusing, had to read it several times, please rephrase (possibly two sentences) for increased clarity*

→ We have split this long sentence into two separate sentences.

Page 1 line 28-31: This study clearly shows that the vertical and lateral distribution of dAl in the North Atlantic differs when compared to other regions of the Atlantic and global ocean. Responsible for the large inter- and intra-basin differences are the large spatial variability in the main source of Al, atmospheric deposition, and the main sink for Al, particle scavenging.

- *Page 2 line 6 from nanomolar to*

→ Corrected

- *12 into the ocean*

→ Corrected

- *26 want is meant by active scavenging*

→ We have now defined active scavenging.

Page 2 line 21-25: Removal of Al in oceanic waters occurs through particle scavenging with subsequent sinking of the particulate matter (Orians and Bruland, 1986). The removal is reported to be caused by active and passive scavenging. Active scavenging occurs through dAl being actively incorporated into the structure of opaline frustules of diatoms, as shown in laboratory experiments and supported by positive correlations between orthosilicic acid ($\text{Si}(\text{OH})_4$) and Al in depth profiles upon the sinking and remineralization of diatomous material

- *Page 5 Line 19 rephrase as readers are not all familiar with this test, e.g. 'A Fisher based test to compare vertical profiles as described by Middag et al. (2015) was used to: : : : This test calculates an integrated p-value as an objective metric to determine how far two profiles are consistent between each other within a given depth interval. This test determined: : :..*

→This point was addressed by referee 1. We have added the suggested sentence.

Page 5 line 4-8: The Fisher's exact test was used for comparison between profiles as both flow injection data sets were measured with replicates (Middag et al., 2015). This test calculates an integrated p-value as an objective metric to determine how consistent two profiles are within a given depth interval. The test determined no significant difference (i-p-value= 0.2-0.3) within analytical uncertainty comparing the two profiles.

- *Page 5 Line 28 mention were this will be explored*

→We are sorry not to understand the signification of the previous comment.

- *Page 8 line 3-5 citations needed*

→We have added citations.

Page 7, line 10-14: Biogenic opal production and biogenic particles play an important role in the removal of dAl in the surface ocean as a result of the high particle affinity of dAl (Moran and Moore, 1988b). Removal of dAl by biogenic particles therefore represents a mechanism which reduces dAl and increases pAl concentrations in surface waters (Moran and Moore, 1988a).

- *Line 12 how would scavenging of Al onto other particles explain the increased pAl/dAl ratio in regions where diatoms were dominant?*

→You are right. It does not. We have changed this sentence in order to make our point more clear.

Page 7 line 22-24: 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).

- *Line 14 what is meant by a 'high correlation'?*
→We wanted to say strong correlation. As such we have changed the word high for strong.
- *Page 9 line 9 here and throughout the ms, refer to transect (GA04N) rather than cruise 64PE370 to be consistent with references to other transects*
→We have changed all 64PE370 for GA04N
- *Page 10 line 1 rephrase e.g. 'had coefficients of determination above $R^2=0.89$ '*
→Changed
- *line 28 'concentrations was' singular plural mismatch*
→Changed
- *Page 11 line 10-11 the ranges in dAl indeed overlap, but the medians are quite different so quoting the medians here is confusing*
→That is right. We have removed the brackets with the median value.
- *Page 12 line 27 elsewhere in the ms salinity is used rather than S (I prefer salinity)*
→We have changed S for salinity throughout the whole ms.
- *Page 13 line 1 core between 1000 and 1200 does not correspond to depth of Al maxima mentioned just previously*
→Indeed, the maximum dAl concentration is found at station one at ca. 900 m depth. Once we follow the transect, the core of maximum concentrations was found between 1000 and 1200 meters depth (neutrally density surface layer $27.6-27.8 \text{ kg m}^{-3}$). This difference is possible due to a different branch of the MOW flowing northwards as the branch we plot which follows a westward direction. The core depth chosen is based on salinity which shows the highest values at this depth. We have rewritten the sentence.

Page 12, line 13-15: Figure 7 displays dAl & pAl versus salinity in the MOW for the neutrally density surface layer (γ_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.

- *Line 23 why z (not defined) rather than depth?*
→ We have modified z for depth. We also modified z for depth at page 13 line 28.
- *Fig1a what is the red square on Greenland?*
→ The red square represents the location where fjord and Iceberg samples were taken. This has been added to the caption.
- *Fig3 name the land masses on the map for those not familiar with this region*
→ We have added Iberian Peninsula to the plot.
- *Fig 4 caption is not clear, what is fig3 in the manuscript?*
→ We have modified the caption of figure 4 and clarify the issues mentioned.
- *Fig 5 caption, wouldn't 'around the southern tip of Greenland' be more appropriate than 'around SE and SW Greenland'?*
→ We have modified the caption and modified SE and SW Greenland for around the southern tip of Greenland.
- *Table 1 number of significant figures is inconsistent and seems inappropriate. Uncertainties on the slopes and intercepts should be reported (and hence for the endmember estimation)*
→ We have provided consistency for the significant figures. We have reported the uncertainty for the endmember estimation.
- *Fig 6 b Was this plot was made in ODV? Caption implies this*
→ No. Caption has been changed.
- *Fig 7 number of significant figures does not seem appropriate and why now GA01 rather than GEOVIDE? And why is the neutral density window reported in the legend for GA01 data and not the others, was it different? This figure could be made to look better if made in another programme (same for fig 8).*
→ We have changed the number of significant figures. We also have changed GA01 for GEOVIDE throughout the text. You are right. They are different. We have acknowledged the difference in the caption. For GA03 and GA04N only the highest dAl concentration

between a neutrally density surface layer (σ_n) between 27.6 and 27.8 kg m⁻³ is plotted. We have redone the figure.

- *Fig 8, why not station 78 as shelf station?*

→We have not added shelf stations because it would have been difficult to interpret the plot. However, we now have included a figure in the SI showing dAl and pAl for station 78 and 2 on the Newfoundland and Iberian shelf, respectively.

Page 12 line 3-6: On the Iberian shelf and margin enhanced dAl concentrations were observed near the seafloor (station 2: up to 21 nM at a depth = 140 m; station 4: up to 27 nM at a depth = 800 m) associated with enhanced pAl concentrations of up to 1.5 μ M at station 2 (Fig. S5).

Page 12 line 12-14 On the Newfoundland shelf and margin (St. 78), no enhanced dAl levels were observed near the seafloor (Fig. S5).

References:

- Brown, M. T. and Bruland, K. W.: An improved flow-injection analysis method for the determination of dissolved aluminum in seawater, *Limnol. Oceanogr. Methods*, 6, 87-95, 2008.
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