Reviewer 3:

Overview:

This manuscript presents results of the application of the MADCOW model for aerosol deposition to recent GEOTRACES data from the Atlantic. The authors expand on the original MADCOW model by varying previously fixed parameters through a combination of comparison with field data for fractional solubility and model date for residence times. While it is an interesting topic, much of the discussion reads like a summary of the earlier works and the manuscript would be better focused on providing new insights into the GEOTRACES datasets through examining how well the assumptions in the MADCOW model are adhered to. There are some question marks regarding the GA08 Al data set also as it the dissolved Al values appear to be overestimated possibly due to the lack of correction for CDOM fluorescence due to the methodology that was used during that expedition. Overall this paper does a good job in adding value to existing GEOTRACES datasets and could make a very useful contribution to this field if it is revised along the lines outlined below.

General Comments:

Atmospheric fluxes – wet and dry

While the paper does a reasonable job of explaining how the fluxes were calculated it does not get into a detailed comparison with atmospheric based fluxes for which there is also data from GEOTRACES and other programs. One aspect of the current work where the atmospheric data would help decipher things is in assigning how much of the surface Al comes from aerosol flux (dry deposition) and how much from wet deposition. In this regard making the link to the precipitation fluxes for each region (Liu et al., 2012) would be beneficial in examining if this is what determine the high inferred model solubility of the aerosols or not. As the assumption of the MADCOW model is that dry deposition is the only process occurring and that in areas where wet deposition is important a higher fractional solubility is assumed. There are data for aluminium solubility in marine rain (Heimburger et al., 2013; Losno et al., 1993), the Losno et al. (1993) paper includes several samples from the Atlantic. See also for example the impact of the Saharan air layer and the ITCZ on the relative humidity in the atmosphere (Braun, 2010). Addition of this type of analysis would greatly increase the impact of this work.

*We now have commented on the differentiation between dry and wet deposition influence on Al solubility. We are not really sure what you mean when saying “if this is what determine the high inferred model solubility of the aerosols or not”. In our model? If this is the case, the answer is no. The Al solubility values we use are all inferred from dry atmospheric deposition. However, it is true that the humidity present within different air masses coupled with large range transport of aerosols may play an important role affecting Al fractional solubility. The latter is valid also for dry deposition after a long transport within humid air mass layers.*

*We do not understand what you mean with atmospheric based fluxes. In case it is aerosol and rain concentration data, we do make comparisons of our calculated deposition fluxes against them. We also have tried to compare against other tracers as for example $^{7}$Be based deposition fluxes. However, the main goal was to compare our calculated fluxes against modelling fluxes since most of our data (mainly apart of cruise GA06) are from remote areas with few or any discrete deposition fluxes published. By comparing against Mahowald model we could extract the atmospheric flux for the same location as our calculated flux and therefore we are able to compare them. We now also compared our fluxes against DEAD model fluxes as suggested by reviewer 1.*

*We are not sure if you write about the parameters we used to constrain the MADCOW model in our study or if you refer to the actual original MADCOW model.*

Seasonality and residence time:

A critical weakness of simple box models like the MADCOW model is that areas with strong seasonality of inputs/outputs are inadequately described when using a single concentration term to fix the inventory. Previous work has indicated that the seasonal cycle (or interannual variability) off the west African coast is on the order of 60 nM for dissolved Al (Pohl et al., 2011) and presumably the residence time is then shorter than 2-5 years first postulated by Helmers and van der loeff (1993). Indeed comparison with Fe suggests that the residence time could be much less than a year or so (Croot et al., 2004; Dammshäuser, 2012; Dammshäuser and Croot, 2012) in these high dust impacted regions. At present there is little discussion regarding the assumptions inherent in a steady state model such as MADCOW, the focus in the paper is on the inventory size as determined by mixed layer depth and concentration and not on whether the fluxes are in balance over the time scales being investigated.

In this regard there are a number of studies that have looked at the seasonality of particle fluxes of Al in the North Atlantic (Chester, 1982; Hwang et al., 2010; Hwang et al., 2009; Jickells, 1999; Jickells et al., 1984; Kuss and Kremling, 1999a; Kuss et al., 2010). With regard to the seasonality in the Benguela region, there has been recent work looking at the fluxes from the Namib (Dansie et al., 2018; Dansie et al., 2017a; Dansie et al., 2017b) and their predominance during austral winter that is of relevance here to the question of inputs and residence times. The challenge that arises then is how to reconcile a snap shot residence time provided by a single concentration measurement within a very active seasonal cycle. For example most sampling is in summer which while likely to be the maximum sink for dissolved Al due to enhanced biological productivity and scavenging, but also could be a minimum in atmospheric deposition leading to a residence time of weeks. Contrastingly winter measurements may have higher deposition rates and minimal scavenging resulting in longer apparent residence times (though mixed layers may be deeper also). So understanding the drivers of the fluxes in each region is probably more important than a residence time calculated from a single surface measurement.

*This is a very interesting and good point. We now have added sentences explicitly addressing this issue. There is seasonality and this is now acknowledged in the text. In the tropical Atlantic seasonality plays a major role, especially through changes in the position of the ITCZ and the nature of episodic dust deposition events.*

*The point raised by the reviewer is quite tricky and more work is needed in the future to address this issues. Also, an important point made recently is the dual role of aerosols as a sink and a source of trace metals (Ye and Volker, 2017).*

*The study of Pohl et al., 2011 makes a great effort in examining the distribution of trace metals along a North to South transect and comparing it against a previous transect in 1990. However, in the latter study no dissolved Al samples were taken and only total Al was analysed. As such, the data (dissolved Al against total Al) are not directly comparable and any comparison would be merely speculative. We are not able to find the seasonal variation of 60 nM total (dissolved mentioned in the comment) Al in the manuscript mentioned in your comment.*

*We have added a sentence acknowledging the effort made by different colleagues on the limitations and assumptions of the MADCOW model. "The limitations of the MADCOW model and extended discussions on the inherent assumptions of the MADCOW model have been acknowledge in previous investigations (e.g Measures and Brown, 1996; Measures and Vink, 2000)..*

Numerous missing references to previous work in the Atlantic:

Not sure if there was some policy by the authors not to include pre-GEOTRACES work on Al in their discussion but there are several papers of direct relevance to this work that need to be included in the discussion as they directly address some of the questions the authors raised. In particular data on surface Al concentrations for dissolved (Gelado-Caballero et al., 1996; Helmers and van der loeff, 1993; Hydes, 1983; Kramer et al., 2004; Kremling, 1985; Kremling and Hydes, 1988; Moran and Moore, 1988; Moran and Moore, 1989; Sarthou et al., 2007) and particulate phases (Helmers, 1996; Kremling and Streu, 1993; Kuss and Kremling, 1999b; Moran and Moore, 1988; Moran and Moore, 1991; Moran and Moore, 1992; Wallace et al., 1981) along with data on the wet deposition of Al (Helmers and Schrems, 1995) and Al flux estimates from atmospheric concentrations (Jickells et al., 1994; Jickells, 1999; ). I am unaware of any analytical reason to exclude these data and the same analytical techniques are still used today.

*We acknowledge your comment. There was no policy to not include pre-GEOTRACES work as some pre-GEOTRACES work has been included (e.g. Vink and Measures., 2001, Van der Loeff et al., 1997, Bowie et al., 2002, Measures and Vink., 2000, Van Bennekom and Jager 1978). In our previous work (same issue) we nearly cite all the dAl works you mentioned above.* We now have added more references of dAl data within the text and *we have included the works for Al flux estimates from atmospheric concentration data (Jickells, 1999 was already included).*

*One of the main and powerful tools of GEOTRACES is the need of running reference material which was not done in the pre-GEOTRACES era. This does not mean that data before the GEOTRACES are not of high quality but they have not been (or only occasionally) cross check against reference seawater or inter-calibrated.*

Analytical quality of the GA08 aluminium data and river discharge:

The value of 784 nM that is reported from GA08 seems very doubtful unless some other information can be provided. Such a value is above the solubility limit for Al at seawater pH (May et al., 1979) and while it is close to undiluted river values for the Congo (Dupré et al., 1996; Meybeck, 1978; van Bennekom and Jager, 1978) most samples would be presumably located at least 12 miles offshore and thus significantly diluted. The linear range for most of the analytical systems is also not that large unless the sample is diluted prior to analysis. It raises questions then about the QA/QC applied to the data. If these samples were using the standard Lumogallion method (Hydes and Liss, 1976) as described in the methods section then they should have been corrected for the natural fluorescence of the samples as was pointed out previously for the Congo plume (van Bennekom and Jager, 1978). This correction should not be underestimated as the humic fluorescence at the excitation/emission used for Lumogallion can be considerable in humic rich waters. The methods that employ preconcentration schemes would not suffer from CDOM fluorescence. At present the fluxes calculated for GA08 all seem to be too high because of the influence of the river plume and potentially the lack of a correction for CDOM fluorescence. The role of river inputs of Al could be compared to estimates of the riverine influence on the Atlantic (Cotrim da Cunha et al., 2007; Cotrim da Cunha et al., 2009) along with Al contents for the major rivers; e.g. Zaire river (Dupré et al., 1996; Meybeck, 1978; van Bennekom and Jager, 1978), Amazon, Orinoco (Mora et al., 2017) and Niger.

*We did correct for natural fluorescence as stated on the original method of Hydes and Liss. Buffered sample. We also diluted several times the samples within the Congo River plume. This high value of dAl is at a salinity of 24. In the same samples we have found values over 1 µM for Fe (pre-*

*concentrated onto Nobias resin and analysed via HR-ICPMS, Krisch et al., in prep). Yes, the fluxes are overestimated due to the influence of Al rich river waters. We have a manuscript in preparation regarding Al in the Congo River plume and in Congo River waters and comparing it with other major world rivers.*

Al composition of dust – the D term in the equation: The 8% value that Measures and Brown used in the original MADCOW was mentioned in the text but I could not find anywhere what value the authors decided to use (should be around 2.69 mmol/g-1 if 8% Al by weight and 26.981539 is the molecular weight for Al) and if they varied this according to region. If it is constant then the term could be incorporated into the S term to reduce the model variables. How valid is the assumption that it is constant? Could not some of the variability in the S term be related therefore to variation in the D term if other studies made the same assumption? At the very least the value used should be included somewhere in the text. Some explanation of how this was handled in the current work would be most illuminating!

*Sorry. Our mistake. We have used the same value as in the original MADCOW model (8.1%). We do not have varied this value. It is used as a constant value. Our S term varies. We think is better to keep the D term in the model equation.*

*It has been postulated that the content of Al in dust is nearly invariant. The minimal differences on this value would not introduce a significant error on the calculations. Some of the variability in the S term could be related to variations in D. However, the impact of those variations in the present work is negligible.*

Specific Comments:

P4 line 20: (sp) The chemical reagent is known as Lumogallion, not Lumogallium.

*Corrected*

P4 line 33. For consistency the dissolved Al concentrations should be in µmol m-3 .

*Corrected*

P5 line 1. There is no explanation of what value is used for the D term in the equation. The other terms are explained in sections 2.2.1 – 2.2.3 but not the D term. If it is constant it could be included then in the S term.

*Indeed it is a constant. We have used the same value as in the original MADCOW manuscript.*

P5 line 7. This is a very large value $\Delta\sigma\theta = 0.125$ kg m-3 to use for determining the mixed layer depth as more recent work have shown that using smaller constraints $\Delta\sigma\theta = 0.03$ coupled with $\Delta T = 0.2°$ C provides a better estimate (de Boyer Montégut et al., 2004), this is in fact the threshold that is used in the Argo mixed layer climatology as cited in Holte et al. (2017). Thus it would be beneficial if the same criteria was used for the observed mixed layer depths to have a consistent approach. The problem with using a value $\Delta\sigma\theta = 0.125$ kg m-3 is that can seriously overestimate the mixed layer depth in high latitude areas leading to an increased inventory and longer residence time.

*Indeed you are right. However, since we did not use our own dAl inventories to derive the residence times of dAl over the Atlantic Ocean we therefore feel that the differences of using one threshold or the other one is not important. In this study, we averaged the dAl values found within the mixed layer.*

P5 line 24. The authors should also be aware of work modelling the fractional solubility of aerosol Al (Han et al., 2012). It would therefore be prudent to include this work in the discussion and compare to the field data of Baker et al. (2013).

*We are aware of this study. However, we did prefer to use solubility estimates from field samples as there is a "good coverage" for the Atlantic Ocean. The estimates given by Han et al., 2012 also suffer from no measurements on the relation between the Al detachment rate or dissolution rate and pH. Also, Al solubility data used in the latter study are very scarce and only available from some cruises. We now mentioned the study of Han et al 2012 within the fractional solubility of Al section (2.2.2).*

P6 line 2. The residence time is a key variable in the version of MADCOW employed in this work and so it should be fairly well constrained. As the authors note the original version of MADCOW had the residence time fixed at 5 years along with the fractional solubility at 8% in order to simplify the calculations as changing one would impact the other. In the current approach it should be noted that the Han et al. (2008) work also includes many of the works that were not included in the citation list (see the general comments above) and these works were used to inform the residence times. It is also worth pointing out to the reader that Han et al. (2008) used a fixed mixed layer depth of 50 m and a constant solubility of 5% so this needs to be directly stated in the current manuscript with regard to how the values might compare.

*We have pointed out the reader the fixed mixed layer depth and the constant solubility*

P6 line 10. The modelled residence times will include advection and mixing to an extent, but the use of a fixed solubility and mixed layer depth will also induce some key differences for the regions examined in the present work. This likely explains why the residence times are longer in the Han et al. (2008) work than in others as for many locations, the underestimation of the solubility and the overestimation of the mixed layer will both work to increase the estimated residence time.

*You are right. In fact, some regions will show longer residence times and other regions shorter residence times in comparison with other studies. However, we needed to choose a benchmark in order to start our interpretations. We have mentioned the issues explicitly in the manuscript now. See end of section 2.2.3*

P7 line 9. See the general comment above regarding this extremely high value of dissolved Al.

*Indeed, this is a large value. We have answered to this comment within your general comment. However, it is averaged since more than one sample was taken within the mixed layer. We do have higher values up to 1.7 µmol of dAl (Menzel Barraqueta et al., in prep.). We also have measured dFe values over 1 µmol in the same waters (Krisch et al., in prep.).*

P8 line 12. There is a considerable amount of surface data for this region and compiling it all in one place may reveal more about the seasonal timings of the dust flux to this region and the aluminium response. See the general comment above regards other works that have data for this region.

*Indeed, there is a considerable amount of surface Al data for this region. However, many reported data are for unfiltered samples which does not match with our filtered dAl samples. We have added a number of additional references with comparisons of dAl data.*

P8 line 21. Not all of these studies attribute it to wet deposition, as the ITCZ acts partially as a barrier to the transport of the dust so the highest values are typically associated with direct dust deposition (Ravelo-Pérez et al., 2016; Tsamalis et al., 2013). Though precipitation is enhanced along the boundary between the ITCZ and the Saharan air layer (SAL) (Wilcox et al., 2010).

*We do not say that all authors attributed it to wet deposition. Our dAl maximum in the region coincide with minimum salinity values which is an indication of freshwater inputs.*

P9 line 2. From where does the Al rich upwelled waters come from? Al profiles normally decrease with depth (scavenged profile) so this needs to be explained further as it would have then be more likely to be resuspension of Al rich particles close to the shelf rather than a direct upwelling source.

*Certainly, the view that dAl concentrations normally decrease with depth is not uniformly correct. With all the new GEOTRACES data being published, it is clear that the dAl depth profiles are highly variable and that the distribution can resemble a scavenged type element but also a nutrient type element.*

*You are partially right. The sentence you point out comes from data presented in Bowie et al., 2002. There is not further discussion on type of source apart from coming from deeper waters. The same as upwelling of deep waters can induce phytoplankton blooms due to the supply of macronutrients and micronutrients it can also supply Al. We have not attributed this Al comes to either remineralization of biogenic particles or resuspension of sediments. However, both options could be correct and would have as a definite result the upwelling of Al rich waters.*

P9 line 3. Do you mean an increased number of particles or that they were enhanced in some other fashion? Larger? More sticky?

*Yes. Increased number of particles. We have reformulated the sentence to avoid confusion.*

P9 line 4. See the general comment on this above.

*Answered above*

P9 line 8. (sp) reported

*Corrected*

P10 line 2. A strong control of the fractional solubility is the relative humidity/hygroscopicity of the particle as this controls the pH, aerosol acidity (Keene et al., 2002).

*We have included this reference. "1) chemical processing during atmospheric transport which is influenced by the relative humidity of the particle (Keene et al., 2002), the balance of acid species (enhanced by anthropogenic sources e.g. fossil fuel combustion; (Ito, 2015; Sholkovitz et al., 2012) and the phase partitioning of $NH_3$ (Hennigan et al., 2015)*

P10 lines 22 and 24. This isn't a calculated result though, it is an estimate from a comparison with the work of Baker and colleagues.

*We have changed the word calculated for estimated*

P10 line 27. See the general comment about relating the fractional solubility to the precipitation or relative humidity levels in the atmosphere for these regions.

*We have replied to this issue above. We now explicit mentioned this issues in the text.*

P11 line 8. It would be useful to see a plot of the residence times (as a 2D map or property-property plot) to see how they look on spatial scales and in relation to primary productivity if it is the main loss term for Al in the mixed layer.

*We acknowledge your comment. However, we are not able to track down the residence time files from Han et al., 2008 in order to extract the actual modelled residence time for each station. Therefore we used a fixed value for each biogeochemical province.*

P11 line 33. It should be pointed out that statistically there are no differences between the values estimated here and those by Mahowald et al. (2005). So speculation on why the Mahowald is over estimated is somewhat spurious.

*This is not fully correct. There are regions were differences are statistically different and regions were they are not. See table 1 (first version of the manuscript).*

P13 line 25. The more northerly flux values are likely underestimated as the residence time used is too long as it is likely in reality, days to weeks (see discussion about this above). This is an important point as the MADCOW model should work well where the Al fluxes and concentrations are the highest.

*Indeed, you are right. We have added a couple of sentences showing that re adjusting the residence time for this region would yield much higher atmospheric fluxes which would make MADCOW calculated fluxes fit within previously reported atmospheric fluxes for this region.*

*See page 15 lines 24 to 27*

P14 line 2. Most likely – it clearly overestimates the fluxes when most of the Al is from the river.

*We have removed "most likely"*

P14 line 12. See the general comment above about relating the seasonality of the dust fluxes.

*Yes. However, this is really difficult to disentangle. The highly productive waters of the BENG region probably have a lower residence time of dAl during the upwelling season and probably a higher residence time during non-upwelling season. It is clear that if we would have sampled during low productivity season we may have found higher dAl concentrations as we found at the time of sampling. The latter would have yielded higher calculated dust fluxes. However, it has been acknowledged before the difficulties of the MADCOW to calculate "accurate" atmospheric fluxes in near-coastal regions and in such highly dynamic regions. Several cruises sampling and catching the seasonal variability of dAl in these waters would provide a better estimation of the fluxes.*

*We now have explicit mentioned this issues in the text.*

P14 line 25. So how do samples collected in the Pacific and Indian oceans tell us anything about deposition to the South Atlantic? Please explain this sentence more clearly.

*Wagener and colleagues performed model simulations to revise atmospheric deposition to the Southern Ocean. Their aerosol samples were taken in the Pacific Ocean and South of the Kerguelen Islands (Indian Ocean). However, they modelled the atmospheric deposition also for the South Atlantic. Therefore, they acknowledge that their largest uncertainties corresponded to regions downwind South America. The latter uncertainties arise from not taking into account or not having samples affected by Patagonian dust.*

*To avoid confusion we have remove the words 'from this sector'.*

P15 line 12. …lack of an island site…

*Corrected. " ..lack of island sites"*

P15 line 16. It is great that Al is measured on GEOTRACES cruises but this does not make this approach using MADCOW any stronger as the majority of the development of this type of work was done pre-GEOTRACES.

*We have modified the sentence. "Dissolved Al is a key trace element of the GEOTRACES programme and as such it is measured on all the GEOTRACES cruises which implies a great chance to use the MADCOW model"*

P15 line 21. Which IDP 2014 or 2017 – both are citeable now.

*The new one. 2017. We have added the reference.*

P15 line 23. For the Atlantic there are a number of north-south transects for Al and so some sort of seasonal signal is probably already possible and should be examined in the current work.

*You are right. We have acknowledge this in the text and use some historical data to decipher the influence of seasonality.*

*However, for a correct interpretation on the seasonal variability samples would need to be taken for the same transects and same stations (assuming the parcel of water is the same which is not the case). In this regard, many of the north to south transects sample different locations (similar in terms of the biogeochemistry) and such the seasonal signal could be bias by different conditions occurring at different stations.*

Figure 2: Please state in the caption the climatology range used here, is it over an annual cycle?

*Yes, it is the annual cycle. Caption corrected*

Table S5: The residence times used in this study are significantly shorter than what has been used previously in the MADCOW model (see above) and they are now on the same time scale as seasonal phytoplankton turnover so does this mean the residence time for Al can be scaled to productivity rather than input fluxes?

*It could be probably done if we consider that productivity is a measured of removal flux. However, you would need to add a non-biogenic removal term too. This would fit into the definition of residence time being the ratio of the dAl inventory in the mixed layer to the rate of input or removal. However, this is beyond the scope of this manuscript.*

Figure S4: The figure and the legend for this figure don't match up and there is no explanation of what the circles represent. While it is easy enough to conclude that the circles may represent discrete measurements at stations, the contoured data isn't explained and clearly does not share the same colour scale as the circles as the lowest value on the colour scale is blue and there is no blue in the contoured data. This figure needs to be fixed and explained better prior to acceptance.

*Done*