Authors' Response to Reviewer Comments on

Long-term Trends in Aerosol and Precipitation Composition over the Western North Atlantic Ocean at Bermuda (ACP-2014-131-Revised)

By W. C. Keene et al.

We thank the two anonymous reviewers for helpful comments on the submitted version of the subject manuscript and respond to each point below. The reviewers' comments are presented in italics followed by our response to each. Unless otherwise noted, line numbers refer to those in the original manuscript.

Reviewer 1

The Bermuda rain and aerosol record represents one of the best long term records in relatively remote areas and I am very pleased to see this interrogation of the record particularly in the context of the relationship between trends in this record and emissions. I understand why the authors have worked hard to discern temporal signals in the aerosol and rain in Bermuda, but some of my comments below reflect my reading of their case as perhaps trying too hard to see this signal, when it may be too small to see against the inherent natural variability.

We respond to the Reviewer's specific concerns along these lines below.

This paper should certainly be published but I would recommend some changes prior to publication. Nearly all of these changes relate to the discussion and interpretation of the data, the methods are well established and validated. My only comment on the methods is that the oceanic and African flow regimes may actually be the related, it is simply the time that they were last in contact with land (p7036 line 1-3)

As described in detail by Moody et al. (2014), the transport classification scheme was based on the fractional residence times for FLEXPART retroplumes within prescribed footprint source regions. Primary transport patterns were defined as those for which the fractional footprint residence time within one source region represented 75% or more of the total footprint residence time. However, only a subset of days could be classified as primary based on this criterion. Consequently, influences of combined source regions as defined by Moody et al. were also considered. As a result, all data subsets classified based on this approach are influenced primarily but not exclusively by emissions within the specified source regions. Both the Oceanic and Africa regimes are associated with flow around the Bermuda high but, as reported by Moody et al., the retroplumes and the corresponding chemical and physical properties of aerosols for these two regimes are distinct.

P7039 line 10-15 Seasonality of emissions may also be relevant Section 3.2.1 and later. A major challenge with this necessarily complex and fragmented kind of time series is that estimating the slope is extremely sensitive to the starting year concentration, as is particularly evident in Figure 1 for sulphate. This problem decreases with length of record so I wondered if the authors had considered using the earlier Bermuda precipitation records they published in the early

1980s (e.g Jickells et al in the references or other work such as Church et al) to extend the record further. This may also apply to nitrate and ammonium.

We agree completely that longer-duration data records would afford greater resolution in evaluating temporal trends. However, for several reasons, we did not include the earlier data for Bermuda in this analysis. (1) The earlier time-series data are limited to precipitation that was sampled at different locations on Bermuda based on an event basis rather than daily protocol as employed for data reported in this manuscript. Consequently, the earlier precipitation data set is not directly comparable in all respects with that reported herein. (2) The earlier data record does not include measurements of corresponding aerosol composition. Interpretations reported in this manuscript are based in part on relative differences in trends for the composition of near-surface aerosols versus precipitation. (3) Resources were not available to calculate and classify FLEXPART retroplumes for the earlier period of record. Interpretations reporting in this manuscript are based primarily on data subsets corresponding to transport from distinct source regions. (4) The earlier data are interpreted in conjunction with the AEROCE precipitation data in another manuscript that is in an advance stage of preparation and will soon be submitted for peer review.

P7042 line 9-15. I accept the hypothesis that anthropogenic nss sulphate may travel at greater altitude, but it is a hypothesis, and yet in line 9 it becomes stated as a fact.

The text on lines 9 to 10 has been qualified as follows "… Consequently, oxidized S above the western NAO MBL, which available evidence suggests is primarily anthropogenic, should exhibit longer atmospheric lifetimes against removal …"

Then in lines 19-30 the authors seem to invoke the same mechanism as they use to explain the stronger trend in precipitation over aerosol sulphate in NEUS/SEUS flow to explain exactly the opposite pattern of rainwater/aerosol sulphate trends in the African flow. This seems illogical.

As described in more detail below in response to a related concern raised by Reviewer 2 regarding interpretation of results for the Africa regime, possible explanations for differences in trends in nss SO_4^{2-} under NEUS/SEUS versus Africa flow have been added to the last paragraph of section 3.1.1.

P7041 line 5-13 I think that isotopic sulphate analysis in the North Atlantic aerosol does not suggest all of it is from DMS (Patris et al., 2000, JGR 105, 14457; Lin et al., 2012 Atm Environ 62, 615). Clearly the Moody analysis reaches a different conclusion, but this difference needs to be acknowledged.

Our data indicate that much of the nss S associated with aerosol over the North Atlantic Ocean does not originate from DMS, which is consistent with results from these other studies so it is unclear why the reviewer feels that these studies reach "different conclusions." Only 97 or 5% of the 1898 aerosol samples reported in our analysis were classified as Oceanic. Based on reported yields of $CH_3SO_3^-$ versus nss SO_4^{2-}, we infer that virtually all particulate nss SO_4^{2-} in this small subset of all samples originated from DMS oxidation. Lin et al. characterized sources of SO_4^{2-} in 33 samples of near-surface aerosols collected along a transect across the North

Atlantic at 36^o N. Although the samples were not classified based on transport, isotopic compositions suggest that virtually all (~95%) nss SO_4^{2-} in 1 of the 33 samples (3% of all samples) originated from DMS oxidation. This frequency of occurrence for samples dominated by S from DMS in the Lin et al. data set is consistent with expectations based on the frequency of and primary source for S in Oceanic samples characterized based on retroplume footprints and product yields in our analysis. We have added a citation to Lin et al. in the introduction. Patris et al. report date for only a handful of samples collected in the North Atlantic, which given the low frequency of occurrence of Oceanic transport patterns, constrains the reliability of direct comparisons with our results.

P7043 line 10-13. In some places in the text the authors do begin to discuss the US deposition records, so I think they need to be carefully to clearly state that here for instance they are back to discussing the Bermuda record

The text has been revised as recommended.

P7043 line 23-27. It's not really obvious what the authors mean by "damping" of the nitrate signal. I think they are arguing that nitrate is not as efficiently long range transported as sulphate which I accept, but that does not damp but rather attenuates the signal.

The word "dampen" has been changed to "attenuate" as recommended.

If the differential efficiency of transport is the issue here, the authors should also not the work done by Fowler and his colleagues (e.g. Matejko et al., 2009 Environ Sci Policy 12, 882 and references therein) on the transport efficiency of these components over Europe. Also p7045 line 23-29.

The differential efficiency of transport over land as investigated by Matejko et al. (2009) is not directly relevant to conditions over the ocean. Because of chemical interactions involving marine aerosol, NO_x and total NO_3 (HNO_3 + particulate NO_3^-), in particular, have much shorter atmospheric lifetimes against oxidation and deposition, respectively, over the ocean than over land (Russell et al., 2003; Keene et al., 2009; Long et al., 2014, *ACP*).

P7044 line 1-5 This argument would suggest that you would see the signal of decreasing nitrate in response to decreasing emissions in rainwater even if you don't see it in aerosol.

As described below in a related comment by Reviewer 2, we have added text to the first paragraph of Section 3.1.2 to point out this perplexing inconsistency.

P7045 line 2 If changes in rainfall at the US east coast is a potential cause of variability, this should be a testable idea using coastal rainfall records I would have thought.

Scavenging of soluble atmospheric constituents by precipitation can occur anywhere along the transport path; wet deposition is not limited to coastal regions. Given (1) the complexity and variability of retroplume footprints, (2) variability in emissions and oxidation of precursors as well as the dry deposition of precursors and reaction products within footprints, and (3) variability in the timing and scavenging efficiencies of storm systems traversing footprints, reliable characterization of variability in wet removal upwind of the site would be problematic at best and, regardless, is well beyond the scope of this investigation.

P7047 line 19 I think the argument about changing pH affecting ammonia speciation is an interesting one. The authors say here "as noted above" but they introduce rather than test this idea earlier in the paper so far as I can see.

The potential for negative artifacts in particulate NH_4^+ based on aerosol sampled in bulk or in relatively coarse size fractions is discussed explicitly in the last paragraph of Section 2.3. In response to the Reviewer's comment, "… as noted above …" has been changed to " … as discussed in Section 2.3 …"

The ammonia/ammonium split is non-linearly related to pH and the key is pKa vs pH. So is the pH change in rainwater likely to be associated with aerosol pH crossing a key threshold?,

The significant decline in the acidity of rainwater clearly indicates that total concentrations of unneutralized acids in the multiphase gas-aerosol system at Bermuda decreased over the period of record. As discussed in the manuscript, this trend is consistent with expectations based on corresponding trends for nss SO_4^{2-}, NH_4^+, and NO_3^- in precipitation. Based on its thermodynamic properties, decreasing acidity will shift the phase partition of NH_3 towards the gas phase and, because the dry deposition velocity for NH_3 to the ocean surface is much greater than that of particulate NH_4^+, the associated importance of removal via wet versus dry deposition will decrease (e.g., Smith et al., 2007, cited in the manuscript). However, the pH of aerosol solutions cannot be reliably inferred from the corresponding pH of rainwater and, in contrast to the Reviewer's suggestion, there is no "key threshold" in this regard. See Smith et al. (2007) for a detailed evaluation of NH_3 phase partitioning as a function of aerosol solution pH. As stated in the original text,

"We recognize that trends reported herein are associated with large uncertainties, the corresponding changes in phase partition and deposition are nonlinear, and explicit evaluation of trends in aerosol solution pH and associated implications are not possible without paired measurements of particulate and gas phase species. However, these relationships provide relevant quantitative context for aspects of S and N cycling over the North Atlantic that are driven by changing emissions over surrounding continents."

is there evidence that significant amounts of ammonia gas are present in the atmosphere (if the fine mode aerosol is acidic it should take up available ammonia gas I would have thought)?,

To our knowledge, gaseous NH_3 has not been measured at Bermuda. However, NH_3 has been measured in parallel with size-resolved particulate NH_4^+ in polluted air along the east coast of the U.S. (Russell et al., 2003; Keene et al., 2004; Smith et al., 2007) and under both polluted and relatively cleaner conditions in the eastern Atlantic MBL (Keene et al., 2009). All of these papers are cited in the manuscript. As reported by those authors, NH_3 partitions with acidic aerosol based on its thermodynamic properties and finite gas-phase mixing ratios exist in marine air under virtually all conditions sampled.

does the aerosol sampling collect gaseous ammonia? – I would have thought Bill Keene was the best person to answer these questions.

To our knowledge, the sampling techniques employed for this study do not “collect gaseous ammonia.” As discussed in Section 2.3, available evidence suggests the possibility of negative artifacts in particulate NH_4^+ due to NH_3 volatilization from aerosol “sampled in bulk (or in relatively coarse size fractions).”

P7047 line 28. The authors argue that a pH shift could explain the increasing ammo nium/suphate ratios seen, this direction of change can be created by emission changes alone and given that the authors demonstrate that the changes between emissions and Bermuda aerosol/rain is not simple, I don’t think you can use a difference in Bermuda ammonium/sulphate ratios to emission ratios as evidence in support of a theory that pH induced ammonia speciation changes are responsible.

We appreciate the Reviewer’s point but he/she refers to only one aspect of the available evidence. As described in our response to the multiple questions embedded in Reviewer’s previous comment (regarding P7047 line 19), several lines of internally consistent evidence support the hypothesis that aerosol acidity has decreased over the period of record, which (based on thermodynamics) would have shifted the equilibrium partitioning of NH_3 towards the gas phase. This evidence includes (1) reductions in emissions of acid precursors SO_2 and NO_x but not NH_3 over the US, (2) the trend of decreasing precipitation acidity, and (3) larger decreases in nss SO_4^{2-} / $NH4^+$ ratios in precipitation relative to near-surface aerosols. As noted above, the text also includes are what we consider to be appropriate cautions regarding our interpretations of these results.

P7048 line 28-30. Having shown that the Bermuda record does not simply reflect US emissions, it does not seem logical to challenge the EPA emission estimates with a Bermuda record. This would be better done with a US mainland deposition record.

The statement in question is, “Our results suggest the possibility that EPA (2013) may underestimate increasing trends in NH_3 emissions over the US or, alternatively, …” We would not characterize the suggestion of a possible underestimate as a “challenge”. However, in response to the Reviewer’s comment, we have modified the text in question from

“Increasing concentrations and deposition fluxes of total NH_3 over the NAO based on all data and under the NEUS/SEUS flow regime imply that NH_3 emissions in source regions have increased over the period of record. However, EPA (2013) indicates that NH_3 emissions from the US did not vary significantly between 1990 and 2009. Our results suggest the possibility that EPA (2013) may underestimate increasing trends in NH_3 emissions over the US or, alternatively, …”

to

"Despite the larger interannual variability evident in the temporal trends, our measurements and associated interpretations suggest that concentrations and deposition fluxes of total NH_3 over the western NAO increased during the period of record (Fig. 2f, Table 2). However, EPA (2013) and Environment Canada (2014) indicate that NH_3 emissions in the US and Canada did not vary significantly between 1990 and 2009. If regionally representative, our results suggest the possibility that these inventories may underestimate increasing trends in NH_3 emissions over North America. Alternatively, …"

Note that here and elsewhere we have added and interpreted emissions from Canada in the analysis.

I think the subsequent discussion of long range ammonium transport form Europe is largely irrelevant. It seems inconceivable that such long range transport can be of a scale to significantly impact Bermuda deposition, and if it is the case then it should be much more evident in US west coast deposition records. That's not to say I question that long range transport occurs, just that it could be large enough to be significant over US emissions in the North Atlantic.

We presume that the Reviewer is referring to "the subsequent discussion of long-range transport" from Asia rather than Europe. Transport from Europe is not discussed in this section. The trend of increasing NH_4^+ in precipitation is significant. This implies that either (1) this increase is a statistical artifact or perhaps localized and not regionally representative or (2) upwind sources for NH_3 increased over the period of record. If the latter, available evidence suggests that either the EPA and Canadian inventories underestimates rising emissions in North America or that transport from other more distant sources are contributing to the trend. We cite measurements and model calculations that support the possibility of contributions from emissions in Asia. For completeness, we think it appropriate to address all possible explanations for the observed trend in NH_4^+.

P7049 line 16 and the rest of this paragraph. If the trend in sulphate aerosol concentrations is not significant, you cannot then use it to estimate a change in radiative forcing, the statistics say there is not a change.

We respectfully disagree with the reviewer on this point. If appropriate caveats are included, a marginally insignificant result should not preclude evaluation of potential implications. The original version clearly states that these results "provide insight regarding the *potential influence* of reductions in SO_2 emissions from the US on direct radiative forcing…" and that the analysis is based on the assumption "that the marginally insignificant trend in particulate nss SO_4^{2-} associated with NEUS/SEUS flow is reasonably representative of the actual long-term decline …" We also point out that "this approach is associated with large and poorly constrained uncertainties." Despite these uncertainties, we feel that the "insight" provided by this element of the analysis is relevant and appropriate.

Section 4. This seems unnecessarily long and repeats most of the earlier text.

We agree and have shortened this section accordingly.

Reviewer 2

The manuscript titled "Long-term trends in aerosol and precipitation composition over the western North Atlantic Ocean at Bermuda" analyzed the long-term trends of SO42-, NO3-, and NH4+ in both the particulate phase and precipitation at a remote observational site over the ocean. Two time frames were discussed, i.e. from 1989 to 1997 and from 2006 to 2009. Long-term trend analysis was performed based on four transport regimes. A major focus was the different behavior of long-term trend of the same species in aerosol and precipitation. Overall, the long-term observational data are precious and helpful indicators for tackling the trends of continental emissions from different regions. However, there are a number of issues needed to be addressed as follows:

Comments:

Page 7026, Line 14: define "VWA" when it first appears

The text has been revised accordingly.

Page 7026, Line 20: "Trends for precipitation", this is confusing. Does it mean "Trends for precipitation amount" or "Trends for precipitation composition"?

The text has been clarified to indicate "Trends in precipitation composition."

Section 2.5: It is better to plot the ensemble clusters of air flows from the four regimes as defined in this study to give reader a better visualization.

We appreciate the reviewer's point. However, adding two 4-panel figures that depict ensemble clusters corresponding to the four flow regimes for aerosols and precipitation as he/she recommends would add considerably to the length what is already a rather long paper. A detailed explanation of the clustering approach along with examples of retroplumes for each flow regime are presented in the cited companion paper by Moody et al. [2014], which was published in *ACP* and is readily available on line. Rather than repeat this information here, we think it more appropriate to summarize major elements of the approach and refer readers to Moody et al. for a more detailed explanation. We had added text indicating that Moody et al. includes examples of retroplumes for the different flow regimes.

Section 3.2: How are the North and Oceanic regimes differentiated? Without a clear visualization, it is difficult to have a clear picture.

As described in Section 2.5, "primary flow regimes were characterized based on proportional footprint residence times within four prescribed source regions from which air was transported to Bermuda. See Moody et al., (2014) for detailed explanation of the procedure employed to identify primary air-mass source regions …" As indicated in our response to the reviewer's preceding comment, Moody et al. present figures depicting example retroplumes for these and other flow regimes.

Page 7039, Line 20-22, Even with adding the 1990 data, the annual trend of nss SO42- for the NEUS/SEUS was not expected to be statistically significant.

The reviewer does not explain his/her rationale for this expectation. In addition, we do not suggest that the trend would necessarily be significant if the 1990 data were added but rather "the absence of a significant trend in particulate nss SO_4^{2-} for the NEUS/SEUS flow was driven in part by the missing data for 1990." As indicated in the text, based on the trend line and the corresponding number of samples for all data, it seems reasonable to suggest that the absence of the 1990 data contributed to the marginal lack of significance in the NEUS/SEUS regression. In response to the reviewer's comment, we have added the corresponding p value (0.12) for the regression to provide quantitative context for our characterization of this trend line as marginally insignificant.

Page 7040, Line 17, A ratio (18.8 ±2.2) of particulate nss SO42 to CH3SO3- was used to estimate the yields of biogenic S, why the concentration of biogenic S was not proportional to that of particulate nss SO42 shown in Fig. 4.? Please clarify the methodology.

Particulate nss SO_4^{2-} that originated from DMS oxidation was calculated as 18.8 * $CH_3SO_3^-$. Nss SO_4^{2-} that originated from anthropogenic sources and $CH_3SO_3^-$ that originated from DMS oxidation varied independently and, consequently, estimated trends in anthropogenic and biogenic nss SO_4^{2-} depicted in Figure 4 were not proportional. The following text has been added to clarify the methodology:

"Briefly, in mass units, anthropogenic nss SO_4^{2-} was estimated from total nss SO_4^{2-} minus the product $CH_3SO_3^-$ * 18.8."

In section 3.2.1 of describing the sources of near surface aerosol and precipitation, the authors ascribed their sources to transport within and above the MBL. However, regarding the location of Bermuda from the continents, low level transport could be possibly not dominant for surface aerosol. In-cloud aqueous processing could still contribute to the surface aerosol via dry deposition.

We agree with the reviewer and did not intend to suggest that low-level transport was the dominant source for near-surface aerosol at Bermuda or that in-cloud aqueous processing was not important. The primary point is that the atmospheric lifetime of oxidized S within the MBL is shorter than that in the overlying FT, which contributes to differences in the observed trends for aerosols versus precipitation. This section has been revised to clarify the above.

Page 7040 – 7041 Line 4: what's the "the beginning of the record" and "the end of the record"? Please clarify it.

The period of record is specified in the methods section text, in Table 1, and in Figure 4 to which the text in question refers. In response to the reviewer's comment, the years have also been added to this section of the text.

Page 7042, Line 11 – 14, this discussion is contradictory to what has been indicated from Figure 4. The contribution of anthropogenic SO42- to its total in aerosol is evidently higher than in precipitation as shown in the figure.

The reviewer is correct. This statement was intended to refer to the relative differences in temporal change rather than absolute concentrations of nss SO_4^{2-}; the original language was not explicit in this regard. The text in question has been clarified as follows:

"We hypothesize that the differential vertical distributions and associated atmospheric lifetimes of oxidized S from anthropogenic versus biogenic sources coupled with reductions in anthropogenic emissions contributed to relatively greater proportionate decreases in anthropogenic contributions to nss SO_4^{2-} in precipitation versus near-surface aerosols over the period of record."

Page 7042, Line 19 – Page 7043, Line 2, It is stated that the Africa regime was related to emissions from Europe. This is confusing. Opposite to the NEUS/SEUS regime, the Africa regime showed stronger decrease of sulfate in aerosol than in precipitation. However, there are no discussions about the differences between the two regimes.

In response to the reviewer's comment, the text has been expanded as follows to address these differences:

"Particulate nss SO_4^{2-} associated with the Africa flow regime decreased significantly (34%) over the period of record (Fig. 2a, Table 2). Based on the SLR, corresponding VWA nss SO_4^{2-} concentrations in precipitation under Africa flow generally decreased, but unlike aerosols, this trend was not significant (Fig. 2b, Table 2). As was the case for the NEUS/SEUS regime, the temporal trends in biogenic nss SO_4^{2-} (inferred from $CH_3SO_3^-$ as described above) were statistically indistinguishable from 0.0 for both aerosols (SLR slope = -0.007 nmol m^{-3} yr^{-1}) and precipitation (SLR slope = -0.006 μmol L^{-1} yr^{-1}), which implies that reductions in anthropogenic contributions drove the decreasing trends in nss SO_4^{2-}. We hypothesize that large reductions in SO_2 emissions over Europe (Vestreng et al., 2007) coupled with transport over the subtropical NAO in association with the easterly trade winds (e.g., Savoie et al., 1989) contributed to the trend of decreasing particulate nss SO_4^{2-} for the Africa regime. However, unlike the differences in trends under NEUS/SEUS flow discussed above, proportionate decreases in nss SO_4^{2-} associated with aerosols and precipitation under Africa flow based on the SLR slopes (Fig. 2a,b) were of roughly similar magnitude. Several factors may have contributed to differences in temporal trends for aerosols versus precipitation based on available data for the two regimes. Anthropogenic contributions to nss SO_4^{2-} under Africa flow were significantly less than those under NEUS/SEUS flow (Moody et al., 2014). Based on the SLR for total particulate nss SO_4^{2-} under Africa flow (Fig. 2a, Table 2) and that for the corresponding anthropogenic component calculated as described above (not shown), anthropogenic contributions to particulate nss SO_4^{2-} decreased from 46% to 19% between 1989 and 2008-09. Based on the SLRs, the corresponding decrease for precipitation was of roughly similar magnitude (40% to 8%). Relative to the NEUS/SEUS, the lower absolute contributions of anthropogenic S to an interannually variable background of biogenic S afforded less resolution in evaluating trends for the Africa regime. In addition, available observations under Africa flow were relatively less frequent (Table 1), which further constrained resolution. Relatively longer transport times to Bermuda from Europe and Africa versus North America as well as differences in associated atmospheric dynamics for the two flow regimes may have also contributed to differences in trends. For example, during transport over the ocean, losses of biogenic S via deposition to the surface are continually replaced via oxidation of marine-derived $(CH_3)_2S$ whereas losses of anthropogenic S are not. Consequently, vertical gradients in the relative abundance of oxidized S from anthropogenic versus biogenic sources and the corresponding differences in ratios of anthropogenic to biogenic S scavenged from the column by precipitation versus those for near-surface aerosols would be expected to decrease with distance from anthropogenic source regions. Finally, to the extent that interactions with mineral aerosol influence the atmospheric lifetime of oxidized S against deposition, long-term temporal variability in mineral aerosol under transport from Africa to the western NAO (e.g., Evan et al., 2006) may have modulated the trend in associated particulate nss SO_4^{2-} relative to that for the NEUS/SEUS regime."

Section 3.2.2. The trend of NO3- in precipitation was distinctly different from that of NOx emissions of US. However, there seems to be no convincing explanation about this.

We agree and have added text to the first paragraph of Section 3.1.2 to emphasize these perplexing relationships explicitly.

Page 7042, Line 8, "… in the MBL dry deposit directly to the surface ocean." This is confusing.

The text has been revised to state that these species "… in the MBL are depleted via dry deposition to the ocean surface."

Page 7045, Line 23 – 28, need to indicate which period was investigated.

The period has been added: "… during the entire study period."

Page 7046, Line 14 – 15, the reference from (Leibensperger et al., 2012) indicated that "Between 1980 and 2009, wet deposition of NO3- over the eastern US decreased 33 % while corresponding NOx emissions decreased 36 %.". This is not consistent with the NO3- trend in precipitation shown in Table 2 and Figure 2.

The NO_3^- trend for precipitation reported in Table 2 and Figure 2 corresponds to the period of record (1989 to 2009) and is interpreted in part based on the corresponding NO_x emissions reported by the EPA and Environment Canada (Figure. 3). While results reported by Leibensperger et al. provide useful context for our analysis, they correspond to an overlapping but distinct time period and, thus, our data cannot be interpreted directly based on their results.

Page 7047, Line 9 – 11, what's the source of the background NO3- and what accounts for its increasing trend?

We do not know and would prefer to not speculate.

Page 7047, Line 16 – 19, it is difficult to see that particulate NH4+ increased from Figure 1, but oppositely, had a slightly decreasing trend.

The text has been revised to indicate that "…NH_4^+ remained essentially unchanged or increased …" In addition, citation of Table 2 has been added.

Page 7048, Line 4, "… relative contributions from the scavenging of gaseous NH3 vs. particulate NH4+ to NH4+ …", this is confusing.

The text has been revised as follows: "…contributions to NH_4^+ in precipitation from the scavenging of gaseous NH_3 versus particulate NH_4^+ increased over the period."

Page 7049, Line 1 – 3, the transport of mineral aerosol from Asia to US was caused by the strong cold front originating from Mongolia, especially in spring. However, this doesn't necessarily mean that anthropogenic pollutants from Asia could be also transported to the east of the eastern US continent. The extent of transport from Asian anthropogenic emissions to the North Atlantic Ocean is questionable.

The text in question suggests that that the long-distance transport of pollutant emissions from Asia may have contributed to rising concentrations of NH_4^+ over the western North Atlantic. However, we do not argue that this is "necessarily" the case. Moody et al. (2014) is cited as an example of mineral aerosol transport from Asia to illustrate that atmospheric constituents can be transported in detectable amounts over such long distances. Dust is often intermixed with pollutants over source regions before lofting and advection over the ocean. For example, mineral aerosol transported across the Atlantic from North Africa is often intermixed with significant levels of pollutants from Europe (e.g., Savoie et al., 1989, among others). We also cite papers reporting that impacts of emission products from Asia on the eastern US are "small to negligible." From our perspective, additional work is required to reliably characterize the relative importance of different sources for NH_4^+ in the western North Atlantic atmosphere. However, in response to the reviewer's comment, we have modified the text from:

"… Alternatively, increasing transport of NH_3 and particulate NH_4^+ from more distant sources in Asia are contributing to rising atmospheric concentrations over the NAO."

to

"… Alternatively, increasing transport of NH_3 and particulate NH_4^+ from more distant sources in Asia may have contributed to rising atmospheric concentrations over the NAO."