

Interactive comment on “Long-term trends in aerosol and precipitation composition over the western North Atlantic Ocean at Bermuda” by W. C. Keene et al.

Anonymous Referee #1

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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. 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

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and validated. My only comment on the methods is that the oceanic and African flow regimes may actually be related, it is simply the time that they were last in contact with land (p7036 line 1-3)

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.

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. 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.

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.

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

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

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signal. 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.

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.

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.

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

P7047 line 28. The authors argue that a pH shift could explain the increasing ammonium/sulphate 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.

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.

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I think the subsequent discussion of long range ammonium transport from 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.

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. Section 4.

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

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 7025, 2014.

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