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Atmospheric Chemistry and Physics Discussions



# *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 #2**

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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

Page 7026, Line 20: “Trends for precipitation”, this is confusing. Does it mean “Trends for precipitation amount” or “Trends for 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.

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

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.

Page 7040, Line 17, A ratio (18.8 $\pm$ 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.

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.

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

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.



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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.

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.

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

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

Page 7046, Line 14 – 15, the reference from (Leibensperger et al., 2012) indicated that "Between 1980 and 2009, wet deposition ïňĆuxes 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.

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

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

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

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.



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