Authors Response to Reviewer 1 comments on

**Flow Climatology for Physicochemical Properties of Dichotomous Aerosol
over the Western North Atlantic Ocean at Bermuda**

By J. L. Moody et al.

We thank Reviewer 1 for helpful comments on the submitted version of the manuscript and respond to each point below. The reviewer's comments are in italics followed by our response to each. Unless otherwise noted, line numbers refer to those in the original manuscript.

Reviewer 1

This is a well-written paper summarizing 3 years of aerosol measurements at Bermuda. The data set is well documented and should be published so that the data are available to the scientific community. The data are compared with previous measurements in Bermuda and other data sets in the literature. The data are divided into 2 seasons and 5 transport sectors for comparison. There are no surprises. The data compare well with previous data sets with perhaps a decrease in SO4 from reduced emissions in the US. The only real issue I have with the paper is the calculation of a mass scattering efficiency based on total scattering and sulfate mass. The authors do define what they have done but this number is really meaningless. I fear someone will use the number as a real MSE without understanding how the authors have re-defined the term. Can you apportion the scattering to the major mass fractions and compare the different MSE for each component?

We appreciate the reviewer's point regarding our use of MSEs. However, most studies for which adequate analytical resolution is available to apportion MSEs among individual aerosol components correspond to short-duration intensive campaigns during which multiple measurement techniques and associated model calculations of water content were employed to generate comprehensive, high-resolution data for aerosol characteristics at specified relative humidities. As noted in the introduction (lines 121 to 125), compromises inherent in longer-term measurement programs such as ours yield less comprehensive data sets and, for some analytes, higher detection limits (DLs). As discussed further in Sections 3.1 and 3.7, analytical resolution varied among analytes and, compared to major ions, relatively fewer measurements of EC, OC, and mineral aerosol mass were greater than DLs. Consequently, only the upper portions of the concentration distributions for these later analytes were characterized. The small numbers of samples in each transport regime for which all analytes were greater than DLs were not sufficient to provide reliable statistics for MSEs apportioned by aerosol component.

We take exception with the reviewer's characterization of our reported mass scattering efficiencies based on total scattering and sulfate mass as "meaningless." These quantitative relationships are highly correlated, differ significantly among transport regimes, and thus provide reliable predictive tools for estimating scattering as a function of transport based on measured concentrations of nss SO_4^{2-}. These relationships also offer benchmarks against which model

simulations of scattering over the western North Atlantic Ocean as a function of source region can be evaluated.

The reviewer's concern regarding potential confusion and misuse of MSEs reported in the manuscript is well taken. To minimize the potential for such problems, we have clarified the text in paragraph 2 of Section 3.7 and also designated the modified mass scattering efficiency (mMSE) based on scattering versus nss SO_4^{2-} mass as follows:

"Aerosol components other than nss SO_4^{2-} (including other nss ionic constituents, primary marine aerosol, crustal dust, and organic matter) also scatter incident solar radiation (e.g., Quinn and Bates, 2005; Li et al., 2006). Scattering efficiencies are conventionally apportioned among the major aerosol components or calculated relative to total aerosol mass and reported in units of $m^2 gm^{-1}$. As discussed previously, many individual measurements of mineral aerosol and OC were less than DLs and, consequently, their associated contributions to scattering cannot be apportioned directly. However, the significant correlations evident in Figure 6 (molar units) and Table 4 (same data in mass units) indicate that the relative mixture of most light-scattering aerosol components within each transport regime during the warm season typically co-varied with nss SO_4^{2-} and, consequently, nss SO_4^{2-} represents a reasonable proxy and a useful predictive tool for scattering by the mixed aerosol populations within each flow regime. We adopted nss SO_4^{2-} as a proxy for all scattering aerosol components in each transport regime and define a modified mass scattering efficiency (mMSE) as the slope for the regression of scattering at 530 nm versus nss SO_4^{2-} mass. The lower limit for individual measurements of scattering coefficients associated with all transport regimes (about 5 Mm^{-1}, Figure 6) represents background scattering by particles such as mechanically produced marine aerosols that are uncorrelated with nss SO_4^{2-}."

Table 2. the super scripts 2 & 3 from table 1 carry over here.

Table 2 has been revised accordingly.

Table 5. Are 3 significant figures really appropriate here?

For consistency with data reported by Savoie et al. [2002], we think it appropriate to report mean concentrations to 3 significant figures. However, in response to the reviewer's comment, percent contributions from anthropogenic sources have been reduced to 2 significant figures.

Figure 6. See above issue with this plot but if you use it, what are the correlation coefficients.

As indicated in the text (lines 754 to 757), correlation coefficients for these regressions are reported in Table 4. In response to the reviewer's comment, a note referring readers to Table 4 for this information has been added to the figure caption.

Figure 7. Same argument.

We have now added regression lines on Figure 7, and we include correlation information in the caption. The relationship between aerosol composition and absorption, and the way it varies

with transport regime is a relevant result.   The last sentence in this section will be modified to read, "Nevertheless, viewed in aggregate, it is evident that physiochemical characteristics of aerosols (e.g., scattering versus nss SO_4^{2-} and absorption versus nss SO_4^{2-}, differ systematically as a function of source region."

Figure 10. I see no benefit from this figure. The correlation explains only 25% of the variance.

We believe this is an important result: Previously (Figure 6) it was shown that near-surface bulk scattering correlates significantly with bulk nss SO_4^{2-}. Figure 10 shows that column measurements of AOD also correlate significantly with near-surface bulk nss SO_4^{2-} and again, relationships for NEUS differ from those for AFR. To better illustrate these relationships, we have added regression lines for NEUS and AFR to the figure, with correlation information in the caption, and we have also added the following text:

 "One might expect paired near-surface observations of aerosol scattering and aerosol composition to be correlated. Results depicted in Figure 10 reveal there is also a statistically significant correspondence between total column extinction measured from space and corresponding concentrations of bulk nss SO_4^{2-} measured near the surface. These results are consistent with a paper by Aryal et al. (ACPD submitted, 2013) based on a subset of the Bermuda data reported here, using data from 2009 in parallel with Micro-pulse lidar vertical distributions of aerosol backscatter which found aerosol optical properties measured near the surface were often significantly correlated with those averaged over the column."