

We would like to thank the third anonymous reviewer (AR3) for providing feedback on this manuscript. AR3 provided little in terms of suggestions or criticisms, rather the reviewer suggested a general agreement with the comments posted by Dr. Mike Fromm (AR2). We kindly ask the reviewer to see our response to Dr. Fromm as a general response to AR3's general comments. However, we address 2 issues raised by AR3 below. Our responses are provided below (red) to AR3's comments (black).

A shift of only about 50 nm in mode radius brings the two curves on top of each other. (Not even accounting for changes in distribution width or multi-modal, gamma, etc., shaped distributions) This means that BrC is essentially indistinguishable from slightly larger sulfuric acid droplets in terms of spectral slope when allowing for uncertainty in particle size.

There has been much confusion over the interpretation and utility of Figure 3. As stated in the original manuscript, there are many assumptions that go into creating this figure; therefore, this figure should not be considered representative of actual atmospheric conditions during any of the events presented herein and is not presented as a predictive model. Rather, this figure presents a *very generalized guide* for how particles of differing composition may change our measured extinction spectrum. Using this figure we developed the hypothesis that we *might* be able to distinguish between smoke/sulfate using the slope method. This figure does not *prove* that this is possible; rather, the case study events speak to this. It is true no information comes out of this figure for use in subsequent analysis; it is just a stepping stone in our original thought process and is presented to help the reader understand the general behavior of smoke and sulfuric acid particles. Indeed, this figure could be removed from the manuscript and the four case-study events would provide ample support on their own.

The 2 smoke curves show a range of potential values that are dependent on the composition (or degree of "complete" combustion) of the smoke. The actual refractive index for smoke is highly variable as shown by Liu et al. 2015 (now reference in the revised manuscript), and the refractive indices we chose provide a reasonable representation of the BrC RI *lower boundaries* in Liu et al. 2015's Fig. 4. As stated above, wildfire burns result in a mixture of BrC and BC being released into the atmosphere and the BC/BrC ratio will be highly variable depending on burn conditions. Further, the composition of BrC determines its spectral properties (i.e., refractive index), which results in a wide range of possible refractive index values (as now shown in the revised manuscript). Of course, this is all complicated by the lack of in situ measurements of stratospheric smoke. Indeed, it would seem that there is a great measurement and modeling opportunity here that should be seized, but is outside the scope of this manuscript. Regardless, what the revised Fig. 3 now clearly demonstrates is that a smoke particle that contains just 10% BC and has a nominal radius of 120 nm is easily distinguished from background sulfuric acid aerosol.

We updated this section to make these points clear to the reader and we thank AR3 for pointing out this ambiguity.

However, figures 15 and 16 do not clearly show a sulfuric acid main (lower) peak and a smoke dominated secondary peak (unless I am missing something).

The reviewer is correct that there is not a clear partitioning of sulfuric acid and smoke as a function of latitude. We have updated our discussion of potential misclassifications within the

Raikoke data sets as well as added support for the identification of smoke from 25°N to 52°N, up to 20 km.