Comment on amt-2021-333
Anonymous Referee #3

This paper presents a technique to use SAGE III/ISS solar occultation measurements of the spectrum of stratospheric aerosol extinction coefficient to classify enhanced aerosol cases as smoke or sulfuric acid. The main idea is to use the spectral slope of the extinction coefficient given that, under a set of assumptions, smoke generates a flatter spectral dependence than sulfuric acid. Four case study events are presented and then the technique is used to analyze measurements in the time period following the 2019 Raikoke volcanic eruption.

While this is an enticing proposition, the main concern already raised by Reviewer #2 (Mike Fromm) is valid. Figure 3, which plots the Mie theory dependencies of spectral slope on particle size, shows that BrC and aerosol in fact have very similar spectral slope characteristics. 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 uncertainly in particle size. So while, yes, the Raikoke spectra have flatter slope than background this is consistent with both larger particles and smoke. The authors are aware of this and state briefly regarding the Raikoke case: “Given the magnitude of this eruption, the spectra identified as smoke here may be the product of both ash and large particle formation.” The authors explain they are not suggesting the data be interpreted as either sulfuric acid or smoke but rather a mixture, but it is not clear how to untangle the effects of changing particle size. I appreciate the context of the case studies provided by the authors and the interesting consistency of the results with the hypothesis that the higher altitude plume is contaminated by smoke. 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).

Overall, I am not convinced that the results merit the main conclusion of the study that the method can discriminate between sulfuric acid aerosol and smoke even under the "applicable" scenarios in the SAGE III/ISS record. If a revision is considered, it should show the technique is valid given realistic uncertainty about changing particle size.