Comment on "Stratospheric Aerosol Composition Observed by the Atmospheric Chemistry Experiment Following the 2019 Raikoke Eruption' by Boone et al.

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Abstract

This is a comment on the Boone et al. (2022) article. The authors analyzed spaceborne observations of stratospheric aerosol in 2019-2020 . They concluded, the dominating aerosol type was volcanic sulfate aerosol. They critisized Raman lidar observations of Ohneiser et al. (2021) and Ansmann et al. (2021). These authors classified the aerosol as wildfire smoke. Boone et al. (2022) stated that this classification is wrong. In this article, we clearly show that the dominant aerosol type was wildfire smoke.

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1 Introduction

Boone et al. (2022) and Ohneiser et al. (2021) studied the perturbation of the stratospheric aerosol layer over high northern latitudes in 2019-2020 and found strongly contradicting results regarding the dominating aerosol type in this layer. Ohneiser et al. (2021) concluded that Siberian wildfire smoke prevailed using ground-based multiwavelength Raman lidar observations. In contrast, Boone et al. (2022) identified sulfate aerosol originating from the eruption of the Raikoke volcano (48.3°N, 153.3°E) on the Kuril Islands in the western Pacific Ocean in June 2019 as the only aerosol component in the layer using spaceborne ACE-FTS (Atmospheric Chemistry Experiment Fourier Transform Spectrometer) observations. In particular, the authors did not find any indication for the presence of smoke. Guided by these findings Boone et al. (2022) concluded that Ohneiser et al. (2021) and Ansmann et al. (2021) erroneously classified the sulfate layer as wildfire smoke layer and that the aerosol typing scheme applied to spaceborne CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) observations correctly identified it as sulfate layer.

There are many aspects in the article of Boone et al. (2022) that need to be clarified, forcing us to write this commentary. Here we show again, that the major pollution source was rather smoke than sulfate. We begin with a short summary regarding the different instrumental and data analysis approaches of Boone et al. (2022) and Ohneiser et al. (2021) and their key findings in Sects. 2 and 3, respectively. Recent, updated Mie computations in support to the Raman lidar aerosol typing approach are presented in Sect. 4. We discuss and harmonize the different, apparently contradicting findings of Boone et al. (2022) and Ohneiser et al. (2021) in Sect. 5. Here, we include stratospheric aerosol observations performed with the spaceborne SAGE III/ISS instrument (Stratospheric Aerosol and Gas Experiment III aboard the International Space Station) (Knepp et al., 2022). In Sect. 6, we briefly discuss the potential and limits of the CALIOP aerosol typing scheme in situations with complex particle characteristics of externally and internally mixed aerosol particles.

2 Aerosol typing based on ACE-FTS, CALIOP, and SAGE III findings

First of all, we should emphasis that the three instruments, the spaceborne ACE-FTS and SAGE III/ISS instruments and the ground-based multiwavelength Raman lidar provided complementary information about the upper tropospheric lower stratospheric (UTLS) aerosol layer. Boone et al. (2022) presented ACE-FTS observations of strato-

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spheric aerosol over the Arctic from July 2019 to March 2020. The primary instrument on ACE is a high-resolution Fourier transform spectrometer that collects infrared spectra via the occultation technique with a vertical resolution of about 4 km. In their current study, quantitative analysis of ACE-FTS aerosol infrared spectra was used to evaluate stratospheric aerosols in the Northern Hemisphere following the 2019 Raikoke eruption. The Raikoke volcano erupted on 21 June 2019 and injected a plume of ash and SO_2 directly into the stratosphere, with cloud tops reaching at least 14 km and rising more than 6 km over a span of 4 days following the eruption.

Boone et al. (2022) claim that the ACE-FTS measurement can be used to "unambiguously determine the aerosol type within a stratospheric aerosol layer". They state that the ability to accurately reproduce the infrared aerosol spectra by using sulfate aerosol optical constants served as incontrovertible proof that the aerosol in all stratospheric aerosol layers over the Arctic from the summer 2019 to the spring of 2020 was sulfate aerosol (liquid droplets consisting of sulfuric acid and water) originating from the Raikoke volcanic eruption. In each case, the fitted sulfate spectrum reproduces the calibrated measurement extremely well, verifying the aerosol type as sulfate. The authors also stated that no evidence was found for stratospheric smoke in the Arctic during all observations from July 2019 to March 2020. Also, for latitudes north of 60°N, there were no stratospheric enhancements observed in biomass burning products (e.g., CO, HCN, C_2H_6), molecules that would have been transported into the stratosphere along with the smoke particles. The question regarding the ACE-FTS fitting and retrieval technique finally arises: Is it really possible to ultimately state that there was only sulfate and no smoke at all in the UTLS aerosol layer? Is the ACE-FTS data fitting procedure sensitive enough to resolve aerosol mixtures and to distinguish clearly pure smoke, pure sulfate, and smoke-sulfate mixture signatures?

Concerning the CALIOP classification efforts, Boone et al. (2022) stated that the results reported for the Raikoke eruption may not vindicate other instances of CALIPSO classification called into question by Ansmann et al. (2021). Because of the agreement between ACE-FTS and CALIPSO aerosol type identification they see no reason to criticize the CALIOP aerosol typing scheme as done by Ansmann et al. (2021). In particular, Ansmann et al. (2021) pointed out that the CALIOP aerosol typing scheme failed to identify the true aerosol type, namely wildfire smoke. In their article, Ansmann et al. (2021) compared aerosol observations of CALIOP with laser foot print close to Leipzig and ground-based lidar observations at Leipzig. The ground-based lidar observations clearly identified smoke as the dominating aerosol type in the stratosphere while the CALIOP misclassified the smoke aerosol as sulfate aerosol.

Knepp et al. (2022) used SAGE III/ISS aerosol extinction measurements (9 data points at near-infrared, visible, and ultraviolet wavelengths) and classified many stratospheric aerosol layers in 2019 as smoke. An example is shown in Fig. 19 in Knepp et al. (2022). Absorption by black carbon in smoke particles was proposed to explain self-lofting observed in the aerosol behavior taken as further proof of the presence of smoke. However, confronted with the ACE-FTS results, the authors were forced to assume that the sulfate aerosol particles were obviously larger than anticipated, and thus pushed the spectral response across the arbitrary threshold chosen to delineate smoke and sulfate aerosols. So, at the end they concluded that they erroneously classified the sulfate as smoke.

3 Aerosol typing based on multiwavelength polarization Raman lidar and main MOSAiC findings

Ohneiser et al. (2021) performed stratospheric aerosol observations with a state-of-the-art multiwavelength polarization Raman lidar aboard the icebreaker Polarstern at latitudes north of 85°N from end of September 2019 to September 2020. These observations were part of the MOSAiC (Multidisciplinary drifting Observatory for the Study of Arctic Climate) expedition, the largest Arctic research initiative in history. Engelmann et al. (2021) provides an introductory regarding MOSAiC remote sensing activities. In the multiwavelength polarization Raman lidar approach (a well-accepted and reliable aerosol typing scheme), the independently measured spectrally resolved particle backscatter (355 nm, 532 nm, 1064 nm) and particle extinction coefficients (355 nm, 532 nm) provide an excellent basis to distinguish main aerosol types (mineral dust, volcanic ash, volcanic sulfate, wildfire smoke, marine aerosol, urban haze) and especially to identify wildfire smoke (Haarig et al., 2018; Ohneiser et al., 2020; Ansmann et al., 2021). The rather aerosol-size sensitive wavelengths of 355 and 532 nm are used in the aerosol typing procedure. The independently measured 3 backscatter and 2 extinction coefficients also provide insight into the size distribution of the aerosol particles and their absorption and scattering properties (Veselovskii et al., 2002; Ohneiser et al., 2021). Based on 25 years of experience with this kind of lidar, since the first article on smoke by Wandinger et al. (2002), it can be concluded that the optical fingerprints of wildfire smoke are unique. Key fingerprint is a strong inverse spectral behavior of the lidar ratio (extinction-to-backscatter ratio), i.e., the lidar ratio at 355 nm is considerably lower by 20-30 sr than the lidar ratio at 532 nm. In addition, the lidar ratio at 532 nm is high ($\geq$70 sr), and indicates absorbing particles. No other aerosol type was found in lidar field observations so far that produces such a fingerprint. The main result as stated in Ohneiser et al. (2021) include that the particles in the UTLS regime over the High Arctic were small, much smaller than expected after a moderate volcanic eruption such as the Raikoke eruption and considerably smaller than in other cases with smoke in the stratosphere (Canadian wildfire smoke, Australian bushfire smoke) (Ohneiser et al., 2021).

According to Ohneiser et al. (2021) and Ansmann et al. (2022), the contribution of Raikoke sulfate aerosol was of the order of 10-20% to the measured overall aerosol optical thickness (AOT) at 532 nm. Knepp et al. (2022) concluded from the SAGE III observations a sulfate fraction of 10-30%. The multiwavelength MOSAiC lidar observations are in full agreement with the particle extinction spectra measured with the SAGE III instrument shown in Knepp et al. (2022). The aerosol was identified as smoke by both, the SAGE III and the ground-based lidar instrument. The SAGE III analysis scheme takes advantage of the different spectral properties of smoke and sulfuric acid aerosol, which is manifest in distinctly different spectral slopes in the SAGE III data. SAGE III is a solar and lunar occultation instrument. The standard products include profiles of the aerosol extinction coefficients at 385, 450, 520, 600, 675, 755, 870, 1020, and 1550 nm. Thus, based on 14 independently measured extinction and backscatter coefficients from SAGE III and the ground-based Raman lidar observations Ohneiser et al. (2021) and Knepp et al. (2022) unambiguously conclude that the dominating aerosol type was wildfire smoke. The particles were definitely not larger than expected after moderate volcanic eruptions, at least over the Arctic, and thus not too large for a proper SAGE III aerosol typing as hypothesized by Knepp et al. (2022).

4 Mie computations supporting Raman lidar aerosol typing: smoke is the major component

We updated our Mie computations for different aerosol types (wildfire smoke, volcanic sulfate aerosol) and performed simplified computations for sulfate-coated smoke particles as well (i.e., for an aerosol with a smoke particle size distribution but sulfate refractive index characteristics). Table 1 summarized the main findings.

The first two rows in Table 1 show two October 2019 MOSAiC observations (Ohneiser et al., 2021). We retrieved the effective radius given in Table 1 by using a lidar inversion method (Veselovskii et al., 2002). We adjusted a lognormal particle size distribution to the lidar-derived size spectra shown in Ohneiser et al. (2021), and the respective mode radius, median radius, and size distribution width are given in parentheses in the first row of Table 1. The lidar ratios in rows 1 and 2 represent well the MOSAiC cam-

Table 1. Measured (MOSAiC aerosol in the UTLS) and simulated optical properties of wildfire smoke (rows 3 and 4), volcanic sulfate aerosol (rows 5 and 6), and sulfate-coated smoke particles (rows 7 and 8). Particle extinction coefficients, σ in Mm^{-1}, backscatter coefficients, β in Mm^{-1} sr^{-1}, and lidar ratios, $S = \sigma/\beta$ in sr, are presented for 355, 532, and 1064 nm. In the Mie computations a monomodal lognormal size distribution defined by the mode radius, r_{mod}, and the size distribution width, s_{dev}, is assumed. Median and effective radius are given in addition. Numbers in paranthesis (in rows 1 and 2) indicate the values of the mode and median radius and size distibution width for a lognormal size distribution with effective radius as measured during MOSAiC. OC and BC particles are externally mixed (97.5% OC + 2.5% BC) in the case of the wildfire smoke computations. Values are normalized to σ_{532}=10 Mm^{-1}.

Aerosol	r_{mod}	r_{med}	r_{eff}	s_{dev}	σ_{355}	σ_{532}	σ_{1064}	β_{355}	β_{532}	β_{1064}	S_{355}	S_{532}	S_{1064}
MOSAiC	(0.150)	(0.165)	0.20	(1.3)	13.28	10.00	–	0.240	0.112	0.036	55	90	–
aerosol	(0.175)	(0.185)	0.22	(1.3)	13.56	10.00	–	0.280	0.143	0.048	48	70	–
Wildfire	0.150	0.165	0.20	1.3	14.56	10.00	2.09	0.280	0.130	0.065	52	77	32
smoke	0.175	0.185	0.22	1.3	12.17	10.00	2.61	0.330	0.151	0.056	37	66	47
Volcanic	0.200	0.245	0.37	1.5	10.02	10.00	4.54	0.338	0.173	0.061	30	58	74
sulfate	0.238	0.284	0.43	1.5	9.10	10.00	5.84	0.373	0.205	0.077	24	49	76
Smoke,	0.150	0.165	0.20	1.3	19.26	10.00	1.65	0.279	0.124	0.0612	69	81	27
sulf. coat.	0.175	0.185	0.22	1.3	16.31	10.00	1.96	0.272	0.124	0.050	60	80	39

paign mean values of 55 sr (355 nm) and 85 sr (532 nm). The shown (adjusted) mode radii are in good agreement with typical smoke mode radii of 125-180 nm as presented by Moore et al. (2021).

The third and fourth rows (wildfire smoke) contain Mie computations for this monomodal smoke size distribution. The refractive index characteristics were taken from Knepp et al. (2022). We assumed an external mixture of organic carbon (OC, 97.5%) and black carbon (2.5%). The results should be taken as rough estimation because BC refractive index values are not well known and it is also difficult to simulate internally mixed BC-OC particles. However, the shown results can almost be reproduced by assuming a 100% OC aerosol. As can be seen, good agreement with the MODSAiC observations in rows 1 and 2 is obtained regarding the spectrally resolved extinction and backscatter coeffients and lidar ratios for this simplified approach.

The fifth and sixth rows (volcanic sulfate) contain the Mie calculations for typical sulfate aerosol conditions a few months after a minor to moderate volcanic eruption. Typical mode radii and a typical size distribution width of monomodal lognormal distributions are selected (Deshler, 2008; Knepp et al., 2022). As shown the effective radii of 0.3-0.45 µm are considerably higher than the ones for smoke ensembles (0.2-0.22 μm). As can be seen, there is no match between the measured and computed spectrally resolved extinction coefficients and the volcanic sulfate lidar-ratio values are too low. These sulfate-related lidar ratios are in good agreement with Mie computations based on realistic, in-situ measured stratospheric sulfate size distribution, observed a few weeks after minor, moderate, and major volcanic eruptions and up to 5-10 years after volcanic eruptions (during quiescent background conditions) (Wandinger et al., 1995; Jäger & Deshler, 2003; Sakai et al., 2016). Based on the in situ measured size distributions, the lidar ratios accumulated between 15 and 30 sr at 355 nm and 25 and 50 sr at 532 nm for effective radii of 0.3-0.45 μm. Note that after moderate and major volcanic eruptions, the size distribution often shows a second mode of larger particles. Such an occurrence of a second mode leads to a further decrease of the lidar ratio. In the stratospheric lidar measurement praxis, most appropriate lidar ratios were always $<$50 sr at 532 nm (Jäger & Deshler, 2003; Mattis et al., 2010; Sakai et al., 2016).

We can conclude that typical volcanic sulfate size distributions (a few months after emission of the SO_2 plumes) cannot explain the observed extinction and backscatter spectra, and the related lidar-ratio values measured during the MOSAiC campaign. Finally, we performed Mie computations with a smoke size distribution (rows 3 and 4) and sulfate-related refractive index values (thus only scattering and no absorption features). Schill et al. (2020) reported that their airborne in situ observations indicate that aged smoke in the remote troposphere contain a significant amount of sulfate. They found a 20-80% mass contribution by sulfate (on average 40-50%) for aged smoke after several weeks of residence in the remote troposphere. These simplified computational results in rows 7 and 8 may give some hints regarding a scenario with an ensemble of sulfate-coated smoke particles. However, in reality, the smoke particles (with BC-containing core and OC-dominating shell structure) have now a shell containing a mixture of mainly organic substances, water, and sulfuric acid and the related refractive index characteristics for this internally-mixed aerosol is unkown.

The related extinction, backscatter and lidar-ratio values in Table 1 (rows 7-8) are in much better agreement with the observed ones in rows 1 and 2 than it was the case in the comparison between the observed and volcanic aerosol values (in rows 5-6). However, the lidar ratios at 355 nm are quite high. A mixture of sulfate-coated smoke particles and pure sulfate particles (with volcanic size distribution) may produce lidar ratios close to the ones observed in row 1 and 2. This scenario was already discussed by Ohneiser et al. (2021) with the conclusion that the Raikoke aerosol fraction was about 10-15%.

5 Contradicting ACE-FTS, SAGE III, and MOSAiC lidar observations: particle chemical and optical properties

It should be mentioned that an extremely unusual and unique stratospheric aerosol scenario developed during the summer of 2019. Severe, partly record-breaking wildfires at high northern latitudes (in Alaska, Canada, and Siberia) served as sources for the UTLS aerosol and at the same time, the largest volcanic eruption occurred (since the major Pinatubo eruption in the summer of 1991) and injected SO_2 plumes into the stratosphere from which sulfate aerosol particles formed. How can we harmonize the different, apparently contradicting observations, on the one-hand the ACE-FTS results, showing clear sulfate aerosol signatures in the infrared spectra, and on the other-hand, the SAGE III and MOSAiC lidar products that point out to the dominance of smoke in the UTLS aerosol layer? The only reasonable explanation is that the smoke particles were partly or completely coated with sulfate so that the optical properties (scattering and absorption) of smoke controlled the measurements from 355 nm to 1.5 μm and the sulfate infrared absorption features the transmission properties at wavenumbers from 750-3750 cm^{-1} (or wavelengths of 2.7-13.3 μm). As mentioned above, Schill et al. (2020) reported that aged smoke showed in most cases a sulfate fraction between 20-80% (on average 40-50%) after about two weeks of long-term travel in the troposphere. For their study, the authors determined the organic and sulfate mass fractions of individual biomass burning particles. As biomass burning particles age, they accumulate sulfate mass from condensation of gaseous sulfuric acid. This accumulation was most favorable in 2019, in the lower stratosphere after the Raikoe volcanic eruption. The Siberian smoke reached the lower stratosphere in the summer of 2019 when the conversion of SO_2 originating from the Raikoke eruption on 21 June 2019 into sulfuric acid was highest (from mid July to mid August 2019, about 4-6 weeks after the eruption) (Thomason et al., 2021).

In the same way as we checked the fractional contribution of the Raikoke sulfate aerosol to the observed 532 nm AOT over the polar region (Ohneiser et al., 2021; Ansmann et al., 2022), we analyzed the Raikoke-related AOT fraction by using the 1020 nm extinction profiles measured with a near infrared imager on the ACE satellite and presented by Boone et al. (2022). The imager collected four images per second of the Sun. A row of imager pixels co-aligned with the center of the ACE-FTS field of view was then used to retrieve atmospheric extinction at 1 μm as a function of altitude. Figure 5 in Boone et al. (2022) shows monthly average atmospheric extinction profiles at 1020 nm for the period following the Raikoke eruption, i.e., for July, September, and October 2019 and this for the latitudinal belts from 60-70°N and 70-85°N.

An excellent agreement between the ACE imager-based aerosol profiles and the respective MOSAiC Raman lidar profiles regarding the geometrical properties are found. Both systems detected the layer base at 7-8 km height, the maximum extinction coefficients around 10-11 km height, and the layer top at about 17-20 km. This agreement corroborates that the ACE-FTS and the MOSAiC lidar monitored the same aerosol layer. We integrated the 1020 nm extinction coefficients in Figs. 5c and d in Boone et al. (2022) from layer base to layer top and yielded respective AOT values of, e.g., 0.03 as September and October monthly mean values for the latitudinal belt from 70°-85°N. This is in agreement with the 532 nm AOT of about 0.08-0.1 measured with the MOSAiC lidar in October 2019 over the North Pole region taking a 532 nm-to-1020 nm extinction ratio of around 3 into account as suggested by Thomason et al. (2021) for the Raikoke aerosol, obtained from SAGE III observations. An extinction ratio around 3 holds also reasonably well for the smoke aerosol (Ohneiser et al., 2021). By comparing these actually measured AOTs and the expected Raikoke-related AOT for high northern latitudes, it is possible to check out to what extent the hypothesis of a pure sulfate aerosol layer is valid.

As discussed in Ansmann et al. (2022), sulfate aerosol originating from the Raikoke volcanic emission of 1.5-1.8 Tg SO_2 (Gorkavyi et al., 2021; Cai et al., 2022) point to maximum AOTs of around 0.025 at 500-550 nm at high northern latitudes in mid-August 2019 and around 0.008 at 1020 nm according to the well accepted relationship between SO_2 mass, sulfate mass (after completing the conversion of SO_2 into sulfuric acid), and the resulting maximum AOT (observable at mid to high northern or southern latitudes). Emissions of 10 Tg SO_2 lead to 500-550 nm AOTs of around 0.15. This clear relationship has been found, e.g., after the Sarychev eruption in 2009 (Haywood et al., 2010), the Chilean Calbuco eruptions in 2015 (Bègue et al., 2017), and even in the case of the Pinatubo eruption in 1991 (Ansmann et al., 1997). Taking an e-folding decay time of about 3-4 months for minor to moderate volcanic aerosol perturbation into account (Haywood et al., 2010) we should have observed 532 nm and 1020 nm AOTs of about 0.015 and around 0.005 in October 2019 at latitudes >60°N, respectively, if the Raikoke aerosol was exclusively responsible for the UTLS AOT. However, the actually measured AOTs of 0.08-0.1 at 532 nm and 0.03 at 1020 nm, mentioned above, are roughly a factor of 6 larger than the expected Raikoke-sulfate-related AOT. A similar result, i.e., a sulfate contribution to the overall 532 nm AOT of about 10-20% was found by Ohneiser et al. (2021) and Ansmann et al. (2021). So, it seems to be impossible that the aerosol typing result based on ACE-FTS observations is valid.

We analyzed AIRS (Atmospheric Infra-Red Sounder) observations over northern Siberia and the adjacent Arctic regarding the carbon monoxide (CO) concentration in the lower stratosphere in August for the years from 2013-2022. Enhanced levels of CO are commonly used to identify air mass originated from wildfire regions and thus to identify smoke aerosol. We checked the satellite-based observations of the CO concentration for the area from 67°-143°E and 70°-87°N from July to October 2019 and found a clearly enhanced monthly mean CO concentration in the lower stratosphere (100-150 hPa) in August 2019 compared to the background years of 2013-2018, 2020, and 2022. The August 2019 mean CO concentration in the defined area at the 150 hPa level (13.5-14 km height) was 63.6 ppb, about 5 ppb larger than the long-term CO background mean value of 58.7 ppb. The CO background values varied within ±1 ppb in these selected 8 background years of 2013-2018, 2020, and 2022 for the defined Siberian and Arctic area. For the 100 hPa level (15-15.5 km height), the data analysis yielded August 2019 mean values of 44.7 ppb, 2.3 ppb higher than the August CO background mean value of 42.4 ppb with background variations of only ±0.5 ppb around the mean in the considered 8 background years. Also these observations are in line with our findings that smoke aerosol was definitely present in the UTLS layer.

As a final remark, we would like to add another (independent) aspect here that points to the clear presence of smoke. One of the main topics of the MOSAiC observations is the combined profiling of aerosols and clouds with the aerosol Raman lidar and a cloud Doppler radar to study, e.g., ice formation processes in cirrus clouds. And we found only indications for heterogeneous ice formation (indicated by a rather low numbers of < 5 ice crystals per liter falling out of the ice clouds), and heterogeneous ice nucleation requires aerosol particles with a solid (insoluble or glassy) particle fraction to initiate ice nucleation. If the aerosol in the UTLS regime would have consisted of pure liquid sulfate particles homogeneous freezing would dominate indicated by a large number of ice crystals of 100 per liter. But these high numbers of ice cyrstals were not observed.

6 Comment on the CALIOP aerosol typing scheme

As stated by Boone et al. (2022), observations by the spaceborne lidar CALIOP generally designated stratospheric aerosols during the second half of 2019 as sulfate, and Ohneiser et al. (2021) suggested the aerosols were smoke rather than sulfate, prompting a call for the revision of years' worth of sulfate identifications from the CALIPSO mission (Ansmann et al., 2021). Since Boone et al. (2022) did not find any evidence for stratospheric smoke in the Arctic in their observations, they consequently concluded that Ohneiser et al. (2021) and Ansmann et al. (2021) misclassified the sulfate as smoke and then erroneously claimed that the CALIOP aerosol typing scheme failed to identify the true aerosol type.

Boone et al. (2022) also claimed that the original SAGE III aerosol typing failed to identify sulfate as the true aerosol type because of the presence of unusually large particles prohibiting an unambiguous aerosol typing. However, as the size distributions presented by Ohneiser et al. (2021) show, the opposite was the case, at least over the Arctic. The particles were considerably smaller than expected after conversion SO_2 emitted by a volcano. There is no reason to assume that the volcanic particles were extraordinarily large and prohibited a successful aerosol typing. So, we think that the SAGE III aerosol typing scheme successfully identified the aerosol layers as smoke layers. The question arises: Why should 14 independent optical information pieces of backscattering and extinction properties measured with two independent, but well designed aerosol remote sensing instruments fail to correctly identify the dominating aerosol type in a stratospheric aerosol layer?

The discussion above may however indicate that the aerosol layer basically consisted of smoke particles but the aerosol smoke-sulfate mixture was rather complex. However, the question remains on how to classify a layer consisting of wildfire smoke particles (coated with sulfate) and pure sulfate particles originating from the Raikoke eruption. We probably need a more detailed aerosol typing schemes in future for spaceborne as well as ground-based lidar applications.

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