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SAGE III/ISS aerosol/cloud categorization and its impact on GloSSAC

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S1 Empirical model for TV13* method

In this section, we briefly describe how an empirical model fit to TV13* method works with the aid of Figure S1. Figure S1a depicts the 525 to 1020 nm extinction ratio as a function of 1020 nm extinction from SAGE III/ISS measurements for March during the period 2017 through 2021 at 17 km altitude. The vertical red line in Figure S1 shows $k_{0\mathrm{TV13}}$, which is defined as shown in box (c) of the flowchart (Figure 4 of the main paper). Here, we use the same empirical method as in TV13 but by using SAGE III/ISS data (Figure S1). To locate the aerosol centroid, we use median value as shown in Figure S1. In this case, the aerosol centroid is located at an extinction ratio (R_a) of 3.6 and extinction coefficient (k_a) of $1.9\text{x}10^{-4}$ km^{-1}. As we move away from the aerosol centroid of the data toward the tail in Figure S1, we notice a long arm of events that stretch from the centroid toward extinction values of about 10^{-2} km^{-1} (saturation limit of the instrument) and extinction ratios of 1. This long arm of data points are likely aerosol/cloud mixtures and for SAGE measurements (we interpret this data as mixtures of aerosol/cloud because of the varying path lengths through the atmosphere which may contain clouds, aerosols, or a mixture of aerosol and cloud). Following TV13, these observations can be modeled with an empirical formula:

$$R = \left(\frac{aR_c k_c + (1-a)R_a k_a}{ak_c + (1-a)k_a}\right) + \delta, a = \left(\frac{k - k_a}{k_c - k_a}\right) \tag{S1}$$

The empirical model in equation S1 returns a ratio of R, for any 1020 nm extinction coefficient k, between aerosol centroid extinction (k_a) and cloud centroid extinction (k_c). Based on the empirical model, we expect R to be close to 1.0 for higher extinctions. We, therefore use an artificial cloud centroid extinction (k_c= 10^{-1} km^{-1}) and an extinction ratio (R_c = 1). We use R_c = 1 as the extinction ratio for cloud centroid, because at this ratio it becomes difficult to distinguish aerosol and cloud. This is shown in Figure 3 of the main manuscript where we discuss how extinction ratio is related to average particle size based on Mie theory. However, for the observations, following TV13, we use an offset (δ) value of 0.4 to account for the spread we observe in aerosol cloud mixture tail in Figure S1a. The spread occurs due to increased measurement noise at higher extinctions in 525 nm extinction coefficient. Therefore, we use R_c = 1.4 for the analysis.

The vertical red line represents $k_{0\mathrm{TV13}}$, which separates standard aerosols (data to the left of $k_{0\mathrm{TV13}}$) and perturbed aerosols (data to the right of $k_{0\mathrm{TV13}}$). TV13 method also uses extinction ratio of 2.0, which roughly represents the wavelength range between 525 and 1020 nm (horizontal red line in Figure S1). Additionally, there is a wedge-shaped region between $k_{0\mathrm{TV13}}$ and the modeled curve, which is denoted as "W" in Figure S1b. As mentioned above, the extinction ratios that are close to 1.4 are likely aerosol/cloud mixtures. In the TV13* method, data that fall in this region were counted as aerosol/cloud mixtures and were removed from further analyses. This includes data below "W" region with extinction coefficients $> k_{0\mathrm{TV13}}$ as they are large particles with extinction ratios close to 1 and are flagged as aerosol/cloud mixtures. Figure S1b shows the data after filtering.

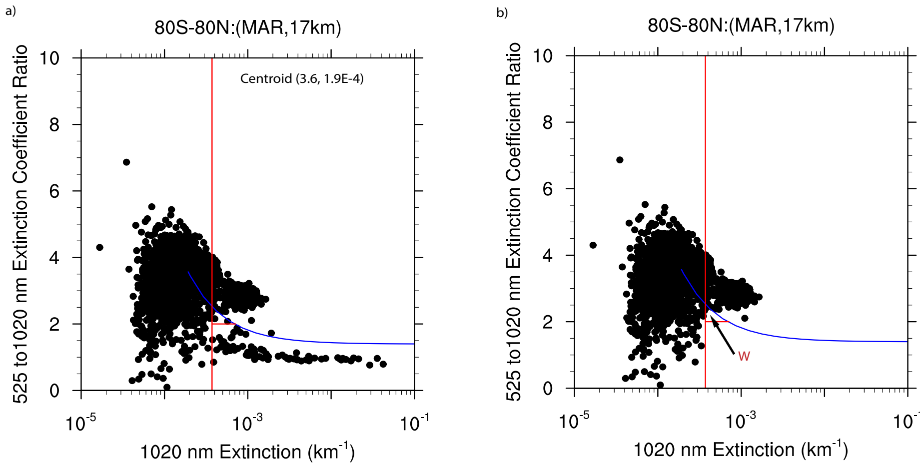

Figure S1. Scatter plots of 525 to 1020 nm extinction ratio as a function of 1020 nm extinction at 17 km altitude for March for the time period between 2017 and 2021, using TV13* Method. (a) before filtering and (b) after filtering. Vertical red line represents $k_{0\mathrm{TV13}}$ ($k_{0\mathrm{TV13}}$ = k_a + 3.0*MAD), whereas the horizontal red line represents extinction ratio of 2.0 that is roughly a representative of the range of wavelengths between 525 and 1020 nm.

S2 Empirical model for SOATCM

We use an empirical model fit to the observations same as in TV13* (supplementary section S1) but for a wavelength combination of 756 and 1544 for the reasons listed in section 3.3 of the main manuscript. The model is computed using formula given in section S1 that includes information on aerosol and cloud centroid co-ordinates. Here, we employed an unsupervised machine learning clustering algorithm called as “k-medoid clustering” (e.g. Kaufman and Rousseeuw, 1990; Park and Jun, 2009) to identify aerosol centroid. The K-medoid clustering algorithm is more robust in identifying noise and outliers as it picks one of the cluster members as the medoid (a medoid is a data point that has the shortest total distance to other members of the cluster). The cloud centroid co-ordinates are empirically determined as 1544 nm extinction and 756 to 1544 nm extinction ratio (r_{ext}) being set to $1.0X10^{-1}$ km $^{-1}$ and 1.4 respectively, similar to TV13* method. We then use equation S1 in to build the empirical model, which is shown in Figure S2 as blue solid curve. Figure S2 is same as Figure 8 of the main manuscript but the difference here is that we use an empirical model fit to the data same as in TV13* method (section S1). While the empirical model is similar to TV13* method, the difference in the SOATCM is that, we retain data that fall in the wedge shaped region (denoted as "W" in Figure S2) as they are likely aerosols from a perturbed event.

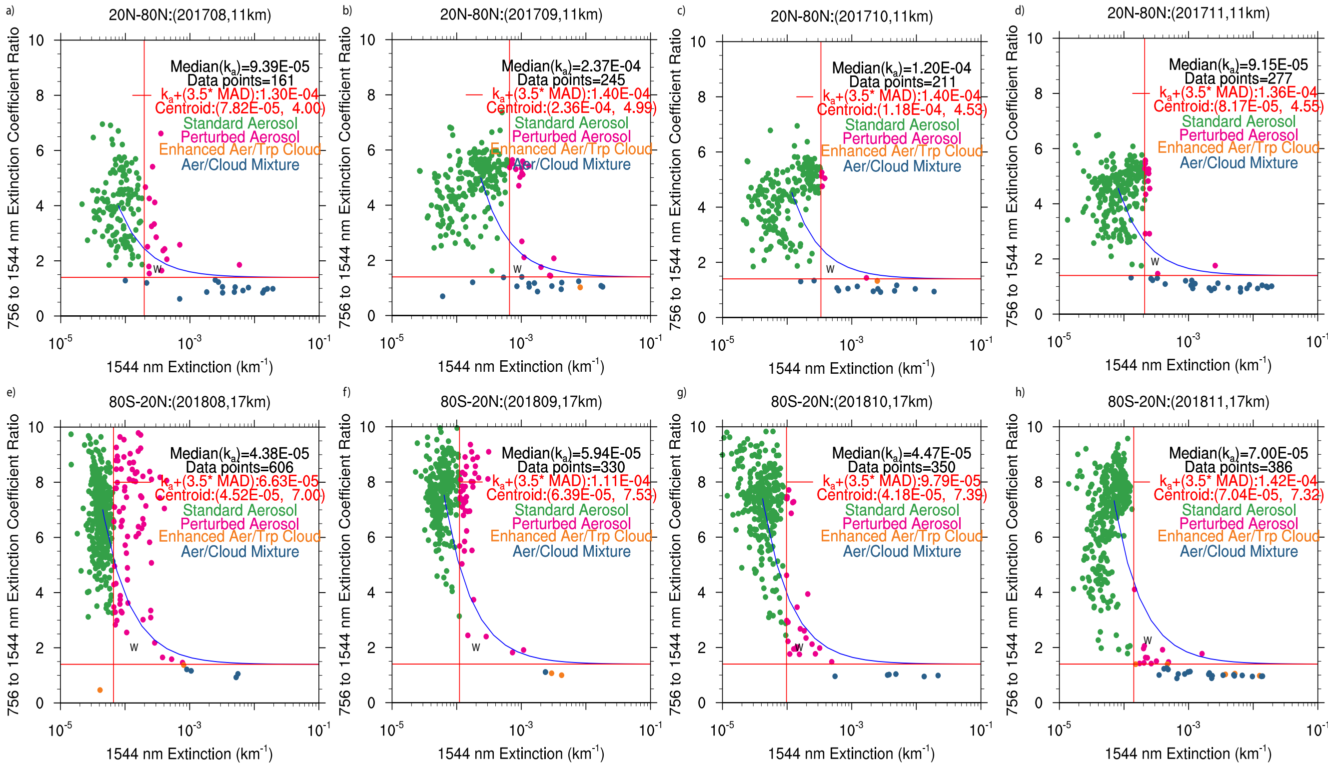

Figure S2. Scatter plots of 756 to 1544 nm extinction ratio versus 1544 nm extinction following Canadian Wildfire event in 201708 (a-d) and Ambae eruption in 201807 (e-h), after applying cloud/aerosol categorization. The vertical line represents k_0 ($k_0 = k_a + 3.5*\text{MAD}$), whereas the horizontal line shows the 756:1544 extinction ratio of 1.4. The blue curve shown in the plots are computed using empirical model and "W" in the plot represent wedge shaped region.

S3 Comparison between TV13* and SOATCM

In this section, we show additional profile comparison between TV13* and SOATCM for several other cases. Figure S3 shows extinction coefficient profile plots at 1544 nm for four cases following (a,e) Ulawun eruption, (b,f) California wildfires, (c,g) La Sourfriere, and (d,h) Tonga eruption. While GloSSAC version 2.2 released version does not contain data for the year 2022, we included a profile from Hunga Tonga eruption (erupted on January 15, 2022) for the comparison purpose only.

While there are differences in the perturbed aerosol category between TV13* and SOATCM, which is not surprising as they differ in statistics and the wavelength combination used as mentioned above. While the important difference between TV13* and the SOATCM is in categorizing "enhanced aerosols/tropopause cloud", the difference in wavelength combinations used for the analysis (525:1020 nm for TV13* and 756:1544 nm for the SOATCM) cause differences in identifying large aerosols. For TV13* method, 525 and 1020 nm extinction ratio approaches unity as the particle size approaches to $\approx 0.5\ \mu$m as per Mie theory, whereas for the SOATCM 756:1544 nm extinction ratio approaches unity when particle size is $\approx 0.8\ \mu$m. This in fact suggests that 756:1544 extinction ratio works better and more sensitive to larger particles. This difference can be seen in Figure S3b,d in particular, where enhanced extinction values are flagged as "aerosol/cloud mixture" in TV13* method. For Figure S3b, the enhanced layer between 14.5 and 16 km were flagged as "aerosol/cloud mixture", whereas the SOATCM flags them as "perturbed aerosol" in Figure S3f. The same is true for a profile following Hunga Tonga eruption (Figure S3d) as the enhanced layer between 22 and 24 km is flagged as "aerosol/cloud mixture" for TV13* method, while the same layer is flagged as "perturbed aerosol" in the SOATCM (Figure S3h).

As discussed in section 3.4 of the main manuscript, we use the influence of any perturbed event based on time series as shown in Figure 6 of the manuscript and use $r_{ext} \leq 1.4$ and $k_{(z,\lambda)} > k_0$ to identify "enhanced aerosols/tropopause cloud". This categorization is made only above tropopause altitude where enhanced extinction coefficient with $r_{ext} \leq 1.4$ could occur as a result of a perturbed event. This difference can be seen in Figure S3 e,f,g,h in comparison to TV13* method as we retain larger particles with $r_{ext} \leq 1.4$ above the tropopause, which is shown as orange filled circles. However, for TV13* method, these data points were flagged as "aerosol/cloud mixture" (blue filled circles in Figure S3 a,b,c,d). While the approach used in the SOATCM is a better step forward in retaining aerosols that may have been influenced by perturbed events, there could be a possibility of mixing up aerosols and clouds at these altitudes as the particles may be large enough so that it is hard to distinguish between enhanced aerosols and tropopause clouds. We therefore name the type as "Enhanced Aerosols/Tropopause Cloud".

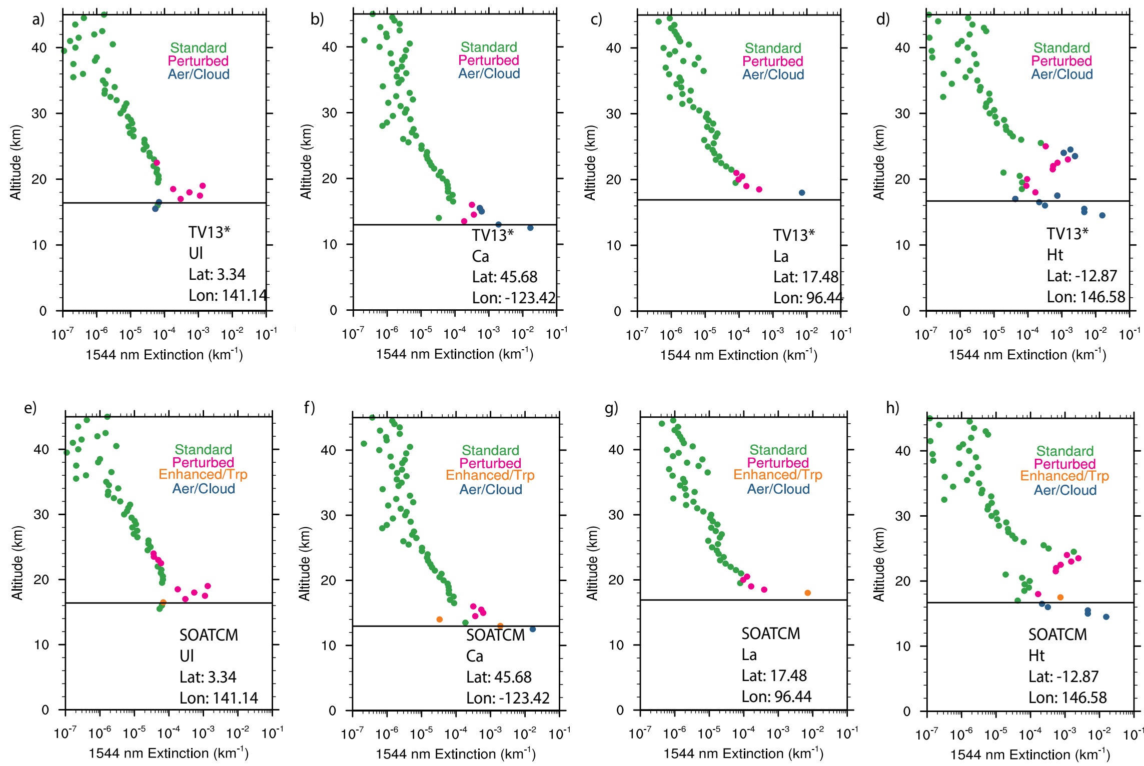

Figure S3. Aerosol/Cloud categorization of extinction profiles at 1544 nm for different volcanic/fire events. Upper panels show (a,b,c,d) extinction profiles for (a) Ulawun (Ul), (b) California Fires(Ca), (c) La Soufriere(La), and (d) Hunga Tonga eruption (Ht) after applying TV13* method, whereas the lower panel show the same profiles as upper panel but using the SOATCM. The location of each profile with latitude and longitude is also shown. Event date (id) for the profiles are (a,e) 20 August 2019 (2019082041SR), (b,f) 05 November 2020 (2020110520SR), (c,g) 09 June 2021 (2021060924SS), and (d,h) 19 March 2022 (2022031938SR).

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