# Sedimentation and growth of volcanic aerosols within the Hunga plume: Insights from unique tropical in situ and satellite measurements

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**Abstract**

The Hunga eruption was an unprecedented event which influenced stratospheric aerosols. This study analyzes the unique dynamics of aerosol plumes using balloon measurements from the Brazil Volcano campaign in Bauru (22.36° S, 49.03 °W) and the Stratospheric Aerosol and Gas Experiment III/ISS along with theoretical calculations. They revealed consistent trends in particle sedimentation and spreading. Temporal analysis showed that smaller particles (<225 nm) experienced slower sedimentation rates, with a larger influence by the Brewer-Dobson circulation. Larger particles (>400 nm) exhibited faster sedimentation, descending approximately 3 km by the end of 2022. The descent of the plume into the Upper Troposphere and Lower Stratosphere (UTLS) by February-March 2023 is accompanied by an increase of effective radius from 400 nm to 450 nm due to hygroscopic growth from moist convection. These unique observations suggest that Hunga could have impacted cirrus cloud formation by re-entering the UTLS.

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## Key points

- Unique combination of in situ and satellite measurements to study Hunga plume microphysics
- Vertical evolution and spread explained by sedimentation and Brewer-Dobson circulation
- Evidence for aerosol growth near the tropopause and potential impact on cirrus cloud

## Abstract

The Hunga eruption was an unprecedented event which influenced stratospheric aerosols. This study analyzes the unique dynamics of aerosol plumes using balloon measurements from the Brazil Volcano campaign in Bauru (22.36° S, 49.03 °W) and the Stratospheric Aerosol and Gas Experiment III/ISS along with theoretical calculations. They revealed consistent trends in particle sedimentation and spreading. Temporal analysis showed that smaller particles (<225 nm) experienced slower sedimentation rates, with a larger influence by the Brewer-Dobson circulation. Larger particles (>400 nm) exhibited faster sedimentation, descending approximately 3 km by the end of 2022. The descent of the plume into the Upper Troposphere and Lower Stratosphere (UTLS) by February-March 2023 is accompanied by an increase of effective radius from 400 nm to 450 nm due to hygroscopic growth from moist convection. These unique observations suggest that Hunga could have impacted cirrus cloud formation by re-entering the UTLS.

## Plain Language Summary

The Hunga eruption was an unprecedented event which influenced stratospheric aerosols. This study analyzes balloon measurements from the Brazil Volcano campaign in Bauru (22.36° S, 49.03 °W) and remote sensing satellite observations along with theoretical calculations. We show that the vertical evolution of the aerosol plume is consistent with particle settling and the general stratospheric circulation. Temporal analysis showed that smaller particles are less affected by sedimentation and subject to uplift by vertical motion compared to large particles. The descent of the plume near the tropopause by February-March 2023 is accompanied by an aerosol growth due to increase of moisture. These unique observations suggest that Hunga could have impacted the formation of cirrus clouds near the tropopause.

# 1. Introduction

Stratospheric aerosols play a critical role in the Earth's climate, atmospheric chemistry and dynamics (Kremser et al. 2016). On January 15th, 2022, the eruption of the Hunga Tonga-Hunga Ha'apai (Hunga) injected a modest 0.42 Tg of $SO_2$ but reached mesospheric levels near 57 km (Proud et al., 2022; Carr et al., 2022). Additionally, this eruption injected approximately 150 Tg of water vapor into the stratosphere, which represents almost 10% of the global water vapor presence in the stratosphere (Millán et al., 2022; Zhu et al., 2022). The Hunga eruption produced a global Stratospheric Aerosol Optical Depth (SAOD) near ~0.016 close to the SAOD peak after the Raikoke eruption. However, the Raikoke event injected thrice the amount of $SO_2$ (Carn et al., 2022). The higher extinction efficiency of sulfate aerosol produced through coagulation due to the large water vapor injected and the longer residence time may explain this unusual SAOD (Asher et al., 2023). Initial $SO_2$ estimates might be largely underestimated (Carn et al. 2022, Hunga workshop Paris), given the rapid production of sulfate in the fresh plume which would have resulted in the rapid conversion of $SO_2$. Finally, the marine water injection (Khaykin et al., 2022) evident of HDO/H2O isotopic ratio (δD) by from ACE-FTS observations could have also led to the injection of marine aerosols.

Ground-based observations from the AErosol RObotic NETwork (AERONET, Holben et al., 1998) have shown that sulfate aerosols have experienced rapid growth to 300-500 nm particle diameters (Boichu et al., 2023). The increase in aerosol optical depth monitored from in situ and satellite observations was one of the most significant ever since the 1991 Mt. Pinatubo eruption (Asher et al., 2023).

Limited balloon-borne measurements of aerosol plumes provided insights into the microphysical, spatial, and temporal variations within the Hunga plume (Asher et al., 2023; Kloss et al., 2022; Deshler et al., 2024; Bian et al., 2023). These measurements indicate that the Hunga impact produced particles larger than 500 nm in diameter several months after the eruption (Deshler et al. 2024; Bian et al. 2023). Asher et al. (2023) underscored the rapid formation of aerosols following the Hunga Tonga eruption facilitated by the humidification of the

stratosphere. This increased humidity led to rapid and substantial ozone depletion likely from heterogeneous ozone chemistry due to injected sea salt (Evan et al., 2023; Zhu et al., 2023).

A comprehensive analysis (Li et al., 2024) of the microphysical processes in the Hunga plume between 0 and 30°S using the CESMI-CERMA aerosol model showed that the aerosol effective radius grew to peak at 400 nm in the early plume. The water vapor injected likely favored the coagulation growth by increasing the oxidation rate of $SO_2$, which led to an increase in the concentration of small particles that coagulated (Li et al. 2024). After 4 months, this study showed that the average effective radius between 20 and 25 km decreased due to sedimentation of large particles. The apparent sedimentation and growth of large particles near the tropopause was observed by SAGE III/ISS in February and March 2023 more than one year after the eruption (Knepp et al. 2024).

However, significant gaps remain in our understanding, due to the lack of continuous in-situ measurements in the tropics, which limits our ability to study the long-term evolution and behavior of the plume. Here we investigate the mechanisms responsible for the vertical evolution of Hunga plume and separate the effect of dynamics, sedimentation, and aerosol growth. We combine for the first time aerosol size distribution from balloon measurements conducted in Brazil and Particle Size Distribution (PSD) product from the Stratospheric Aerosol and Gas Experiment (SAGE) III/ISS.

The focus of this work is to: 1 - derive microphysical properties of the Hunga plume using optical particle counters; 2 - compare and associate OPC measurements with those from SAGE III/ISS observations; 3 - separate the role of sedimentation and dynamics in the vertical evolution of the Hunga plume; 4 - Estimate the hygroscopic growth of volcanic aerosols observed near the tropopause.

# 2. Methodology

## 2.1. BraVo Campaign

This study utilizes data collected during the Brazil Volcano project (BraVo) campaign, conducted at the Centro de Meteorologia de Bauru (IPMET-UNESP) in Bauru, Brazil (-22.28, -49.25), from May 24th, 2022, to August 13th, 2023. Thirteen balloon flights were carried out, deploying a suite of instruments including a Profiling Optical Counter (POPC), Compact Backscatter Aerosol Detector (COBALD), Cryogenic Frost Point Hygrometer (CFH), Electro-Chemical Counter (ECC), and an Aerosol Sampler. The POPC, which measures aerosol size distribution and concentration, is central to this analysis, providing crucial information for investigating sedimentation and aerosol growth processes within the plume. To complement the in-situ POPC measurements, we incorporate PSD product from SAGE III/ISS.

## 2.2. POPC

We employed the Profiling Optical Particle Counter (POPC, Dumelie et al., 2024; Li et al., 2023). Its sensitivity ranges from 0.3 µm to 10 µm with 30 particle diameter channels. The

calibration of the instrument is done with Polyethylene Spherical Particles (PSL). Since the scattering efficiency of PSL is higher than sulfate or sea salt aerosol, we applied a correction factor on the size threshold of the POPC data (see Figure S1).

The first balloon was launched 4 months after the eruption, 12 POPC occurred in total during 19 months with two intensive campaigns in May and August 2022. The balloon ascended to the stratosphere and burst at an altitude between 26 to 32 km above sea level, capturing the vertical extent of the plume. The system collects data at a 0.5 Hz frequency. POPC was used to study volcanic plumes after several volcanic eruptions (Li et al. 2023; Dumelié et al. 2024). It compared well with other counters such as Printed Optical Particle Spectrometer (POPS) and stratospheric aerosol extinction measurements from SAGE III/ISS.

This study provides multiple observations of the Hunga plume to investigate its vertical displacement and separate the contributions of the sedimentation, dynamics, and aerosol growth.

## 2.3. SAGE III/ISS

The Stratospheric Aerosol and Gas Experiment III (SAGE III/ISS), mounted on the International Space Station (ISS) (Cisewski, et al., 2014), was utilized to analyze the stratospheric aerosol properties following the Hunga eruption (Knepp et al., 2024). SAGE III/ISS operates by scanning the limb of the Earth's atmosphere during solar and lunar occultations, offering a unique advantage point from the ISS's orbit. This method provides highly accurate vertical profiles of aerosols and gases by measuring the attenuation of solar or lunar light as it passes through the atmosphere (Chu et al., 1999).

We use profiles of the extinction coefficient (Level 2, v5.2) at 1020 nm, water vapor mixing ratio and the PSD product (Knepp et al., 2024) from the SAGE III/ISS data. The PSD product (V1.1) includes the “Number Density”, “Mode Radius”, “Standard Deviation”, and their associated uncertainty. It is based on finding the best solutions for number density, mode radius, and sigma to reproduce aerosol extinction spectra using Mie theory calculation. SAGE III/ISS PSD product variables are used to derive POPC-like observations (number concentration above the 30 sizes threshold).

The data collection was specifically timed to capture the dispersion of the volcanic plume as the ISS orbited over Bauru with the co-location criteria of +/- 10 degrees in longitude and latitude. During the campaign, three balloon flights were collocated with SAGE III/ISS observations within 100 hours of separation from the balloon measurements (see table S1 supplementary materials).

# 3. Results

## 3.1. Observational evidence for the vertical displacement of the Hunga Plume

The Hunga aerosol plume was observed up to ~40 km with OMPS-LP (Taha et al., 2022) and CALIOP (Sellito et al., 2022) within a few days after the eruption. Within 3 weeks, the plume descended to 24-26 km likely due to the radiative cooling from water vapor (Sellito et al., 2022; Legras et al., 2022). Figure 1 shows the zonal median aerosol extinction coefficient at 1020 nm and effective radius profiles from SAGE III/ISS between 0-30 °S from June 2021 to November 2023. The aerosol plume is observed with a maximum extinction coefficient at 1020 nm, descending from 25 km in January 2022 to near the tropopause at 16.5 km by November 2022. During the descent, the plume width increased by several kilometers, likely affected by both sedimentation and vertical uplift of the Brewer-Dobson circulation. This is a unique behavior for a volcanic plume in the tropics for the past 2 decades. The zonal mean scattering ratio profiles in Figure S1 show that all the plumes observed by CALIOP ascended in the stratosphere due to the upper branch of the Brewer-Dobson circulation (Vernier et al., 2009; Fairlie et al., 2014) but the Hunga plume had a different behavior. Knepp et al. (2024) proposed that the sedimentation was mostly responsible for the descent and vertical spread of the plume in the tropics. Another very interesting feature in Figure 2 is the aerosol growth from an effective radius of 400 nm in August 2022 to nearly 450 nm in February-March 2023 between 18-20 km. While Knepp et al. (2024) identified these features, this study delves deeper into the mechanisms driving the plume's vertical displacement. Specifically, we investigate the roles of sedimentation, the dynamics, and hygroscopic growth, aspects not addressed in previous work. We combined in situ measurements with SAGE III/ISS to further study the plume’s microphysical evolution.

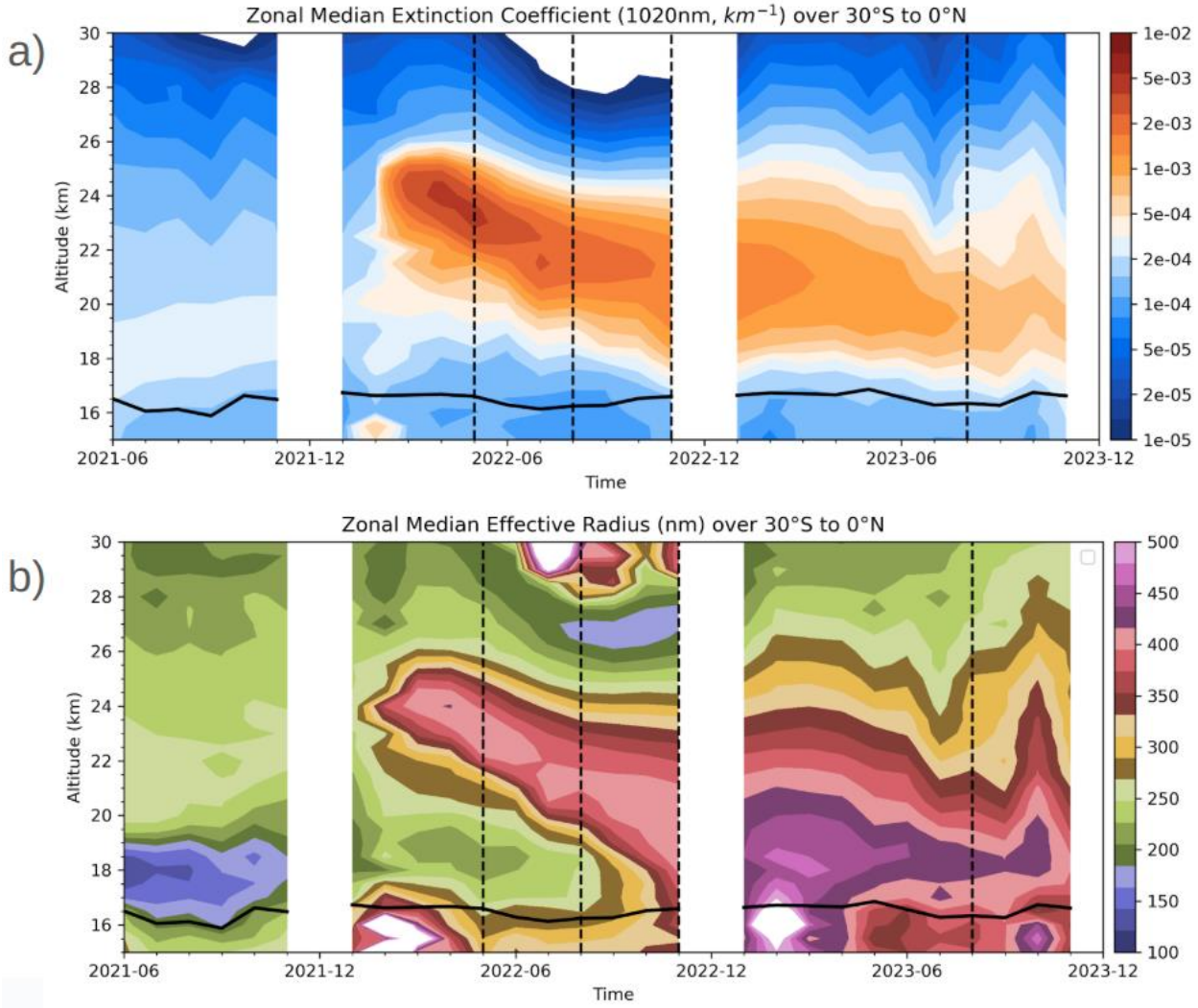


**Figure 1.** - *(a) Monthly zonal median profile evolution between 0-30°S of aerosol extinction at 1020 nm and effective radius. (b) from SAGE III/ISS from June 2021 to November 2023. The vertical dashed lines show the measurement periods of the BraVo campaigns in May 2022, August 2022, November 2023, and August 2023. The tropopause level is shown by the solid black line.*

Figure 2 shows POPC differential concentration profiles and effective radius to track the evolution of the Hunga plume. The concentration profiles across ten different particle sizes are displayed in the upper panel of Figure 2. The maximum concentrations, which occur at varying altitudes depending on the date, decrease as the plume descends over time, indicating gradual sedimentation. For instance, on May 24th, 2022, the highest concentration is observed around 21 km with values of 3 #/cm³ for particles near 200 nm while the peak for those near 300 nm was approximately 0.5#/cm³ near 23 km. The concentration values decrease in subsequent profiles, reflecting the plume's descent through the stratosphere and the diffusion of the plume through its transport in the southern hemisphere mid-latitudes. Measurements made on November 3rd, 2022, showed that the total number for r>250 nm peaked near 1-2 #/cm³, a value close to the one reported by Deshler et al. (2024) at 12 km over McMurdo station in

Antarctica a few months later in April 2023. The effective radius (reff) peaked near 21.5-24 km on May 24th, 2022, near 290 nm, progressively descending to 20-22 km on August 12th, 2022, spreading between 17-22 km by November 3rd, 2022. The reff maximum continues spreading to levels below the tropopause by August 2023. Reff maximum increased from 290 nm to 300 nm from May 2022 to August 2023. Relative to SAGE III/ISS, the peak reff observed by the POPC is lower by ~30% but still in the lower range of estimates from Boone et al. (2024) who found a reff between 280-400 nm. POPC counting efficiency near 50% for r>150 nm may also explain some of the discrepancies with SAGE III/ISS. Around 5 months after the eruption, Li et al. (2024) reported POPS measurement near Reunion Island [21S, 55.4E] located at a similar latitude as Bauru. They showed mean reff between 20-25 km around 330 nm, as well as profiles with a peak near 430 nm on March 31st, 2022, consistent with SAGE III/ISS.

The lower panels of Figure 2 present histograms of particle size distributions at three distinct altitude ranges (18-20 km, 20-22 km, and 22-24 km), showing a shift towards larger particle sizes at lower altitudes. This trend is particularly evident in the progressive increase in the volume of larger particles, especially in the 500 to 1000 nm range, as shown in the histograms from higher to lower altitude bands.

SAGE III/ISS shows that the main plume contained particles with effective radii greater than 400 nm, and over time a partitioning between larger particles (> 400 nm) and smaller particles (< 300 nm) was observed, with the largest particles showing a rapid descent compared to the smaller ones (Knepp et al., 2024). By March/June 2023, some particles approached 500 nm near 18 km indicating significant growth and sedimentation over the period. This is consistent with observations from Boichu et al. (2023) who observed through column integrated AERONET data a peak radius progressively increasing from July 2022 to January 2023 near 500 nm. Further theoretical considerations are provided here to interpret these observations.

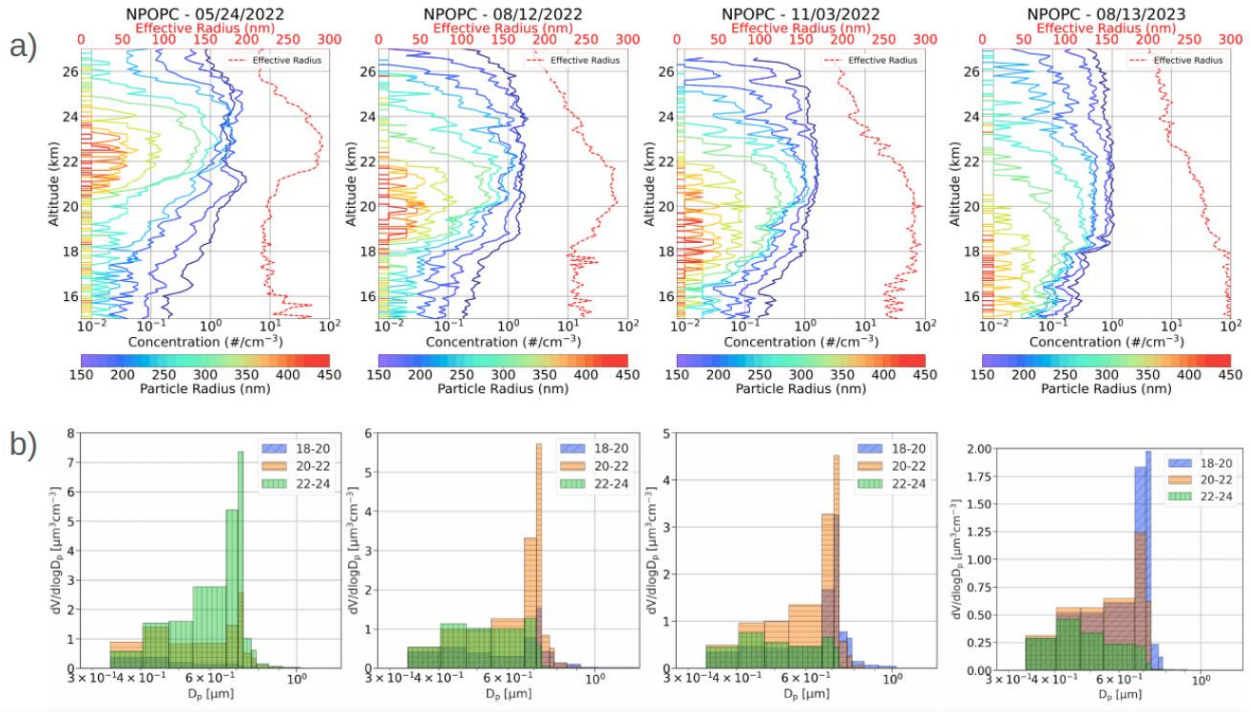


Figure 2 - The upper panels show the particle concentration profiles from POPC for 10 size ranges between 150 and 450 nm, different sizes are represented by the colors. The lower panels show the distribution of particle diameter for three different altitude ranges.

## 3.2. Sedimentation and dynamical processes responsible for the vertical evolution of the Hunga plume

We derived further parameters and chose the lognormal fit of the concentration profiles to provide both the maximum and width of the plume (supp. mat.). Repeating the process for all POPC data collected, we derived the temporal evolution of these quantities. We collocated 3 balloon flights with SAGE III/ISS overpasses during the BraVo campaign. For comparisons with POPC, the 3 parameters of the lognormal distribution (mode radius, standard deviation, total concentration) from the SAGE III/ISS PSD product were used to obtain the concentration above a size threshold to mimic and fill the gaps in the POPC measurements. Fitted concentration profiles for r>150nm between are shown in Figure 3b, where we can see that SAGE III/ISS underestimate the concentration compared to POPC by about 40%. However, the altitude of detection of the plume is consistent between the two instruments. Figure 3a shows the evolution of the plume with both SAGE III and POPC data merged. Both instruments show the descent of the plume by ~3km from January 2022 to August 2022 before reaching a plateau until May 2023 and descend again afterward. The larger aerosols (r>300 nm) appear to descend faster than the smaller sizes. SAGE III/ISS PSD product shows the same sedimentation behavior in the first months after the eruption and also shows that the plume started to rise around October 2022, keeping a roughly constant altitude before descending again.

We used sedimentation calculations along with zonal mean vertical wind from MERRA-2 to try to reproduce these observations.

We apply Stokes' Law to estimate their settling velocity (W) under laminar flow conditions:

$$W = \frac{C\rho\frac{r}{2}2g}{18\eta} \quad (1)$$

where ***g*** is the acceleration due to gravity, ***r*** is the radius of the particle, ρ is the particle density, and ***η*** is the viscosity of air. **C** is written as environmental factors (e.g. temperature, pressure, and humidity) that can affect these variables, requiring adjustments to the calculation, especially for non-spherical particles or in turbulent flow conditions. Figure 3d shows the sedimentation for several values of radius in the same range as POPC channels.

The other factor affecting the plume's vertical evolution is the dynamics of the stratosphere and updraft from the tropical branch of the Brewer-Dobson circulation in the tropics. The vertical wind was calculated using MERRA-2 reanalysis dataset, which calculates vertical wind speeds by integrating observations with model forecasts, adjusting through physical laws and continuity equations, and ensuring consistency with observed atmospheric conditions (Gelaro et al., 2017). Figure 3c shows that the vertical wind was near 0 during the first part of the BraVo campaign in May 2022 becoming positive after August.

The theoretical calculations match the initial plume motion. However, the theoretical plume rise was 2 km higher than observed in November 2022. On the other hand, after May 2023, the plume altitude was underestimated compared to SAGE III/ISS. The particles show similar behavior with time, comparing the initial and final launch. The smaller particles seem to have slower sedimentation rate compared to the large ones, as expected from theory. However, the plume shows a different behavior in August 2023 compared with the theoretical curve. The plume rises above the predicted curve, indicating a more complex dynamics, reflecting some inhomogeneity in the plume dispersion.

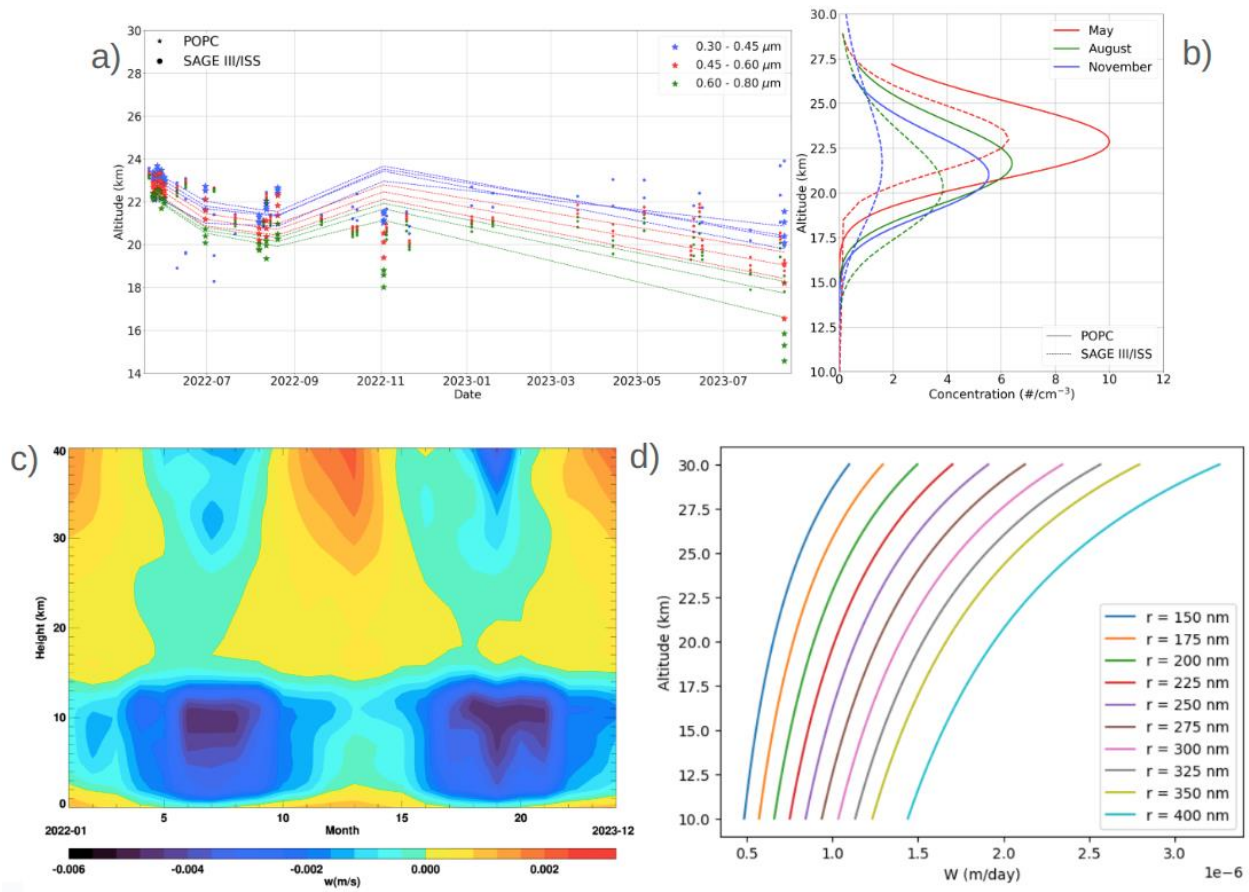


Figure 3. (a) The maximum of the lognormal fit corresponding to each size range shown by blue, red and green colored stars (for POPC) and dots (for SAGE III/ISS). Dotted lines represent the theoretical sedimentation velocity estimated for each POPC measurement. (b) Profiles of particle concentration for POPC and SAGE III/ISS during May (red), August (green) and November (blue) of 2022. (c) Zonally averaged (between 20°S - 30°S) vertical velocity derived from MERRA-2 reanalysis. (d) Sedimentation velocity for particles at different POPC size (radius) channels.

## 3.3. Hygroscopic Growth of Hunga Aerosols

After more than one year from the Hunga eruption, the sedimentation and transport of the volcanic plume lead to changes in its microphysical properties. The zonal mean $r_{eff}$ from SAGE III/ISS illustrated in Figure 1 shows that the plume descended more than 5km, from January 2022 to March 2023, and increased from ~ 400 nm at 24-26 km to 450 nm near 18 km. This indicates that aerosol within the plume underwent growth while reaching tropopause levels. The mechanism behind this growth is probably the increase in the relative humidity (RH) at lower altitudes, which might have favored the condensation of water droplets on volcanic aerosols. Figure 4a shows the zonal (0-30° S) mean RH profile from June 2021 to November 2023 derived from SAGE III/ISS water vapor mixing ratio and temperature from MERRA-2 (see method in supp. mat.). We overplotted on the RH contour the $r_{eff}$. At 18 km, the RH shows a

significant seasonal variation with maxima between January and March during the convective periods in the Southern hemisphere. During the descent, the RH within the plume goes from 2% to more than 40% which coincides with the increase in the $r_{eff}$. To model the particle growth, we took an effective radius ($r_{eff}$) profile from August 2022 and compared it with a profile on March 2023 in Figure 4b. We used hygroscopic growth with kappa-Köhler (Titos et al., 2016) theory that can be written as follows:

$$GF(RH) = \frac{r_{wet}}{r_{dry}} = \left(1 + K \times \frac{RH}{100 - RH}\right)^{1/3} \quad (2)$$

Where:

- $r_{wet}$ is the wet diameter of the aerosol particle.
- $r_{dry}$ is the dry diameter of the aerosol particle.
- $K$ is the hygroscopicity parameter known as Kappa
- RH is the relative humidity expressed as %

Using equation (2), we can show that growth factor (GF) can be expressed for hygroscopic growth from RH1 to RH2 by:

$$GF(RH_1, RH_2, K) = \left(\frac{100 - RH_1}{100 - RH_2} \times \frac{100 - RH_2(1 - K)}{100 - RH_1(1 - K)}\right)^{1/3} \quad (3)$$

Applying the theory to the profile in August 2022, assuming the presence of ammonium sulfate particles (Kappa=0.6) and an increase in the RH of 40%, we see that the $r_{eff}$ almost matches the values observed in March 2023. The corresponding growth assuming the presence of sea salts (Kappa=1.2) produces values above 450 nm. These calculations seem to indicate that the aerosol plume growth can be explained by hygroscopic processes.

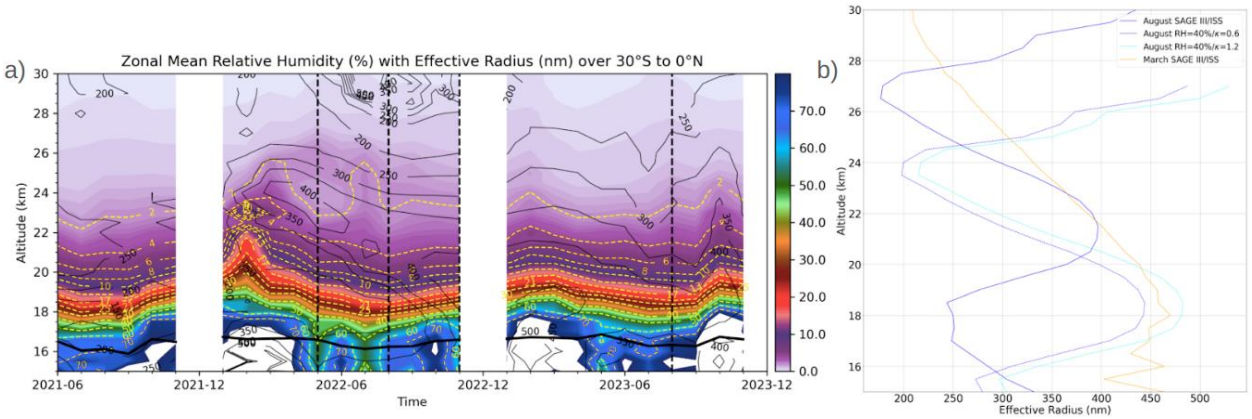

Figure 4. (a)- Filled color contours show the time-series (June 2021 - November 2023) of zonal mean relative humidity between 15 and 30 km altitude averaged over 0 - 30° S. Relative humidity is derived from SAGE III/ISS water vapor mixing ratio and MERRA-2 temperature profiles using Murphy and Koop (2005). Black contour lines superimposed on the color contour show the effective radius obtained from the SAGE III/ISS PSD product. (b) Comparison between August 2022 and March 2023 $r_{eff}$ profiles from SAGE III/ISS.

# 4. Conclusion

This study investigated the dynamics of aerosol plumes from the Hunga eruption using in-situ measurements and satellite observations from SAGE III/ISS during the BraVo campaign in 2022 and 2023. By applying lognormal fits to the aerosol concentration profiles, we derived key parameters such as plume width and center altitude, revealing insights into the plume's evolution.

The BraVo campaign included three balloon flights coordinated with the ISS passages, enabling a unique comparison between POPC and SAGE III/ISS observations. Both instruments detected the plume at similar altitudes, with SAGE III/ISS corroborating the POPC's findings of sedimentation and plume dispersion. However, the POPC recorded 40% - 50% higher concentrations than SAGE III/ISS within the plume (Figure 3), possibly due to differences in instrument sensitivities or measurement techniques.

Our analysis revealed distinct temporal changes in plume behavior. Smaller particles (< 250 nm) exhibited slower sedimentation and greater vertical spreading compared to larger particles. These smaller particles gradually descended by ~2 km before being lifted again by the Brewer-Dobson circulation, returning to their initial altitudes by late 2022. Conversely, larger particles (225-400 nm) experienced faster sedimentation, moderated by the Brewer-Dobson circulation, resulting in a ~3 km descent by late 2022.

Plume width also varied with particle size. Smaller particles showed limited lateral spreading, while larger particles, especially those > 300 nm, exhibited an increase in plume width of over 2 km.

Using equations for sedimentation and vertical wind from MERRA-2, the vertical displacement of the particles during the first one and a half year after the eruption was reproduced. Aerosol growth observed near 18 km observed from February to March 2023 can be explained by the increase of relative humidity during the convective season, which led to hygroscopic growth. This indicates that the sedimenting Hunga plume might have influenced the formation of cirrus clouds near the tropopause during that time, but this needs further investigation.

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# Open Research

The SAGE data are available at https://asdc.larc.nasa.gov/project/SAGE%20III-ISS. We used Python codes available online.

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