



Optical properties of stratospheric aerosols from wildfires and volcanic eruptions constrained by satellite and ground-based lidars – Part II: Evolution of young plumes

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Abstract. Volcanic eruptions and intense wildfires can inject large amounts of aerosols and their precursors into the stratosphere, where they can persist from several months to years and affect atmospheric composition and radiative balance. The optical and microphysical properties of these aerosol populations evolve rapidly during the first weeks to months following injection as a result of transport, sedimentation, particle growth, aggregation and chemical ageing. Characterizing this early
15 evolution is essential for understanding the processes controlling the transformation and vertical evolution of stratospheric aerosol plumes.

Following the large-scale and long-term analysis of stratospheric aerosol perturbations presented in Part I, in this study, we investigate the early evolution of major volcanic and wildfire stratospheric aerosol perturbations using observations from the
20 Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) aboard Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) satellite. The CALIOP Level 2 5 km Aerosol Layer product is used to combine scattering ratio (SR), particulate linear depolarization ratio (PLDR), and color ratio (CR), providing complementary information on aerosol loading.

The analysis is extended beyond the CALIPSO observational period to the 2025 Panboreal Wildfire Outbreak (PWO) using
25 observations from the Atmospheric Lidar (ATLID) aboard EarthCARE. ATLID captures the progressive ascent of the stratospheric smoke layer and its evolving depolarization, providing an independent characterization of the self-lofting and microphysical evolution of a recent major wildfire perturbation. Together, CALIOP and ATLID observations demonstrate the value of combining aerosol loading, depolarization, size-sensitive optical properties and vertical transport to characterize the early evolution of stratospheric aerosol plumes.



30 1 Introduction

Part I of this study investigated the large-scale and long-term evolution of stratospheric aerosol perturbations associated with major volcanic eruptions and wildfire events using the CALIOP L1CC dataset, complemented by ATLID observations for the most recent events. The analysis showed substantial event-to-event variability in aerosol loading, vertical evolution and persistence, with distinct optical behaviours associated with volcanic and wildfire perturbations. In particular, the long-term
35 evolution was found to depend not only on the magnitude and altitude of the initial perturbation, but also on the subsequent dynamical and microphysical evolution of the aerosol layers.

Previous lidar studies have documented the optical and microphysical evolution of several major volcanic and wildfire stratospheric perturbations. For volcanic aerosols, CALIOP observations have shown substantial differences in particulate
40 depolarization and color ratio depending on plume composition. Prata et al. (2017) reported much higher particulate depolarization and color ratio for the ash-rich Puyehue plume than for the more sulfate-rich Kasatochi and Sarychev plumes. The latter, exhibited a rapid decrease in both depolarization and color ratio during the first days following injection. Similar persistence of ash in the lower stratosphere was documented after the 2014 Kelud eruption, for which CALIOP depolarization measurements revealed a significant contribution from non-spherical ash particles (Vernier et al., 2016). More recent
45 observations of the 2019 Raikoke eruption further showed that fine ash-containing aerosol can persist for extended periods and undergo diabatic self-lofting, demonstrating that volcanic plumes may exhibit complex coupled microphysical and dynamical evolution (Khaykin et al., 2022).

Stratospheric wildfire smoke also exhibits distinctive optical properties. Multiwavelength lidar observations of the 2017
50 Canadian smoke plume revealed substantial particle depolarization at 355 and 532 nm, indicating persistent non-sphericity of aged stratospheric smoke particles (Haarig et al., 2018). The same event demonstrated that absorbing carbonaceous aerosols can undergo radiatively driven self-lofting, whereby solar heating of black-carbon-containing smoke enables continued ascent after injection (Yu et al., 2019). This mechanism was particularly pronounced during the 2019–2020 Australian wildfires, when smoke-charged anticyclonic vortices transported the aerosol plume upward to approximately 35 km, while maintaining
55 a coherent and long-lived stratospheric structure (Khaykin et al., 2020; Kablick et al., 2020; Allen et al., 2020).

These event-specific studies demonstrate that depolarization, wavelength-dependent backscatter and vertical displacement provide complementary information on aerosol morphology, size-sensitive optical properties and plume dynamics. However, most previous studies have focused on individual perturbations or on a limited subset of optical parameters. A systematic
60 multi-event analysis combining scattering ratio, particulate linear depolarization ratio, color ratio, their statistical distributions, and their evolution in potential-temperature coordinates still lacking. Such an analysis is needed to identify recurrent optical



behaviours while also quantifying the substantial event-to-event variability among volcanic and wildfire stratospheric aerosol plumes.

- 65 The evolution of stratospheric aerosol plumes is controlled by interacting microphysical, chemical and dynamical processes, including sedimentation, aggregation, chemical conversion, mixing and radiative heating. These processes modify aerosol loading, particle morphology, size and vertical distribution during the first weeks to months following injection. Their effects can be investigated through the simultaneous evolution of several lidar-derived optical parameters.
- 70 The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), aboard CALIPSO, provided vertically resolved aerosol observations at 532 and 1064 nm between 2006 and 2023 (Winker et al., 2009). Its polarization-sensitive measurements at 532 nm provide information on particle non-sphericity, while the spectral dependence of aerosol backscatter provides information related to particle size. In this study, three complementary optical parameters are considered: the scattering ratio (SR), which characterizes the strength of aerosol scattering relative to the molecular background; the particulate linear depolarization ratio (PLDR), which is sensitive to particle non-sphericity; and the color ratio (CR), defined as the ratio of particulate backscatter at 1064 and 532 nm, which provides a size-sensitive optical indicator. Their combined analysis allows the evolution of aerosol loading, morphology and size-sensitive optical properties to be investigated simultaneously.

- Lidar observations have played a key role in documenting stratospheric aerosol (SA) injections and their subsequent evolution, owing to their ability to resolve the vertical structure and determine optical properties of aerosol layers. Since the beginning of the CALIPSO mission, spaceborne CALIOP observations, often complemented by ground-based lidar measurements, have been used to characterize numerous volcanic and wildfire perturbations of the stratospheric aerosol layer. Volcanic injections associated with Kasatochi (Hoffmann et al., 2010; Karagulian et al., 2010), Sarychev (Jégou et al., 2013; Prata et al., 2017), Nabro (Bourassa et al., 2012; Vernier et al., 2013), Puyehue–Cordón Caulle (Nakamae et al., 2014; Vernier et al., 2016), Kelud (Vernier et al., 2016; Kristiansen et al., 2015), Calbuco (Bignami et al., 2015; Lopes et al., 2019), Raikoke (Vaughan et al., 2021; Osborne et al., 2022), and Hunga (Legras et al., 2022; Sellitto et al., 2022) have been investigated using lidar observations, providing information on plume altitude, aerosol scattering, depolarization, and the evolution of ash- and sulfate-containing layers. More recently, Martinsson et al. (2026) used the complete CALIOP record to provide a global assessment of stratospheric aerosol perturbations during 2006–2023, identifying the influence of 15 volcanic eruptions and five wildfire events on the stratospheric aerosol layer.

- Lidar observations have also provided important constraints on stratospheric smoke injected by major pyroconvective events. Following the 2017 Canadian wildfires, satellite lidar observations from CALIOP documented the injection, large-scale transport, and vertical evolution of the stratospheric smoke plume (Khaykin et al., 2018; Torres et al., 2020), while ground-based lidar observations provided detailed measurements of its optical properties, depolarization, and persistence (Haarig et



al., 2018; Baars et al., 2019). The 2019–2020 Australian wildfires provided another major example, with satellite observations revealing the large-scale transport and pronounced vertical ascent of the smoke associated with radiatively driven self-lofting (Khaykin et al., 2020), while ground-based lidar measurements documented long-lived smoke layers and their distinctive optical and depolarization properties (Ohneiser et al., 2020). Together, satellite and ground-based lidar observations demonstrate their complementarity for characterizing both the large-scale evolution of stratospheric aerosol injections and changes in particle morphology and size-sensitive optical properties during plume evolution.

Part I of the current study investigates their large-scale and long-term evolution over the CALIPSO observational period, focusing on aerosol loading, vertical evolution, depolarization, and persistence. Here, in part Part II we focus on the early evolution of individual aerosol plumes and their optical properties during the first approximately three months following injection. Using the CALIOP Level-2 5 km Aerosol Layer product, the key optical parameters - SR, PLDR, and CR are analyzed through their temporal evolution, statistical distributions, and diabatic vertical evolution in potential temperature space. The analysis is extended beyond the CALIPSO period using observations from the Atmospheric Lidar (ATLID) aboard EarthCARE, launched in May 2024 (Wehr et al., 2023), to investigate the 2025 boreal Wildfire Outbreak. Together, the two companion papers connect the early plume-scale optical and dynamical evolution investigated here with the large-scale dispersion, persistence, and the long-term stratospheric impact document in Part I.

2 Data and Methods

2.1 CALIOP observations and Level 2 product

The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), aboard the CALIPSO satellite, provided vertically resolved observations of aerosols and clouds from 2006 to 2023 (Winker et al., 2009). CALIOP operated at 532 and 1064 nm, with polarization-sensitive measurements at 532 nm. The separation of the parallel and perpendicular components of the 532 nm backscatter signal provides information on particle depolarization, while the combination of the 532 and 1064 nm channels enables the wavelength dependence of aerosol backscatter to be investigated.

To study the early evolution of volcanic and wildfire plumes, we use the CALIOP Level 2 5 km Aerosol Layer product Version 4.51 (CAL_LID_L2_05kmALay-Standard-V4-51; Vaughan et al., 2005). Unlike the L1CC product used in Part I to characterize the large-scale and long-term evolution of stratospheric aerosol perturbations, the Level 2-layer product provides layer-resolved information on individual aerosol features, including their vertical boundaries and optical properties. This layer-based approach is therefore better suited to isolating recently injected plumes and characterizing their optical properties during the early stages of their evolution, before they become more spatially dispersed and mixed with the background stratospheric aerosol. Version 4.51 benefits from improvements in 532 nm calibration, aerosol classification, and lidar-ratio selection, leading to a more robust characterization of weak stratospheric aerosol layers (Vaughan et al., 2009).



Only aerosol layers satisfying the quality-control criteria defined for this study were retained. The analysis was restricted to
130 layers classified as stratospheric aerosols (feature type = 4), with aerosol subtype values between 2 and 4, corresponding to
volcanic ash, sulfate, and elevated smoke, respectively, and with confident subtype classification (subtype QA = 1). Features'
aerosol optical depth (AOD) was restricted to values between 0.01 and 1.5, with the lower threshold excluding very weak
aerosol layers for which the retrieved optical properties are less robust and the upper threshold removing unusually optically
thick or potentially contaminated layers. The integrated volume depolarization ratio (VDR) was restricted to values between
135 0 and 0.4, excluding non-physical negative values and extreme retrievals, and was subsequently converted to particulate linear
depolarization ratio (PLDR), as described in Sect. 2.2. Color ratio (CR) values were restricted to 0–0.8 to remove extreme
values associated with poorly constrained spectral backscatter measurements. Layer-top altitudes were restricted to the
Tropopause–30 km range, consistent with the vertical coverage of the CALIOP Level 2 aerosol layer product. This upper limit
implies that aerosol layers extending above 30 km are not captured in the present analysis. In particular, the highest part of the
140 Hunga plume and the later stages of the Australian New Year (ANY) wildfire plume after its ascent above 30 km are therefore
not represented. Polar stratospheric cloud contamination was excluded by retaining only features classified as aerosols in the
CALIOP Vertical Feature Mask and rejecting features classified as clouds or polar stratospheric aerosols.

For each event, observations were referenced to the date of the initial injection and analyzed over the subsequent 90-day period.
145 This time window was selected as a compromise between capturing the main early-stage optical and microphysical evolution
of the aerosol plumes and limiting the influence of later large-scale transport, dilution, and mixing processes. A shorter period
would not fully capture the transition from freshly injected to aged aerosol populations, whereas a substantially longer period
would increasingly include the effects of hemispheric-scale transport and long-term plume dispersion addressed in Part I. For
the temporal evolution presented in Fig. 1, observations were aggregated into 6-day intervals to reduce short-term sampling
150 variability while preserving the evolution of the aerosol layers. Median values were calculated within each interval. The other
analyses presented in this study use the individual filtered layer observations without this temporal aggregation.

2.2 EarthCare – ATLID

The Atmospheric Lidar (ATLID) aboard the EarthCARE satellite is a high-spectral-resolution lidar (HSRL) specifically
designed to improve the characterization of aerosol–radiation interactions (Wher et al., 2023). ATLID operates at 355 nm and
155 exploits the spectral broadening difference between molecular (Rayleigh) and particulate (Mie) backscatter. A high-resolution
Fabry–Pérot interferometer is used to spectrally discriminate between these contributions, enabling direct separation of
molecular and particulate signals (Donovan et al., 2024).

In contrast to elastic backscatter lidars such as CALIOP, this HSRL technique allows the independent retrieval of aerosol
160 backscatter and extinction without prescribing an extinction-to-backscatter lidar ratio. This represents a major advantage for



the observation of compositionally complex stratospheric plumes, where the assumption of a constant lidar ratio can introduce significant uncertainties, particularly in the presence of mixed sulfate, ash, and carbonaceous smoke particles (Young et al., 2018).

165 This capability is particularly relevant for the analysis of wildfire-driven stratospheric aerosols, whose microphysical structure and optical properties can differ substantially from those of sulfate-dominated volcanic aerosols (Khaykin et al., 2020). In this study, we use the ATLID Level-2A EBD product (Baseline BA), which provides extinction coefficient, backscatter coefficient, and particulate linear depolarization ratio (PLDR) at 355 nm for aerosol layers. These parameters are used to characterize the optical and vertical evolution of the stratospheric smoke layers associated with the 2025 Panboreal Wildfire Outbreak.

170 2.3 Optical parameters

The early evolution of the aerosol layers was characterized using complementary optical parameters from both satellite lidars. For CALIOP, we use the scattering ratio (SR) and particulate linear depolarization ratio (PLDR) at 532 nm, together with the color ratio (CR) derived from the particulate backscatter at 1064 and 532 nm. For ATLID, the analysis relies on the scattering ratio (SR) and particulate linear depolarization ratio (PLDR) at 355 nm. Their combined analysis provides complementary
 175 information on aerosol loading, particle morphology and size-sensitive optical properties.

The scattering ratio (SR) characterizes the strength of aerosol scattering relative to the molecular background. Values close to unity indicate a weak aerosol contribution, whereas increasing SR values indicate enhanced aerosol scattering. SR is therefore used to characterize the intensity and vertical distribution of the aerosol perturbation:

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$$SR(\lambda, z) = \frac{\beta_{tot}(\lambda, z)}{\beta_{mol}(\lambda, z)} \quad (1)$$

where $\beta_{tot}(\lambda, z)$ and $\beta_{mol}(\lambda, z)$ are the total and molecular attenuated backscatter coefficients, respectively, at wavelength λ and altitude z .

The particulate linear depolarization ratio (PLDR) is sensitive to particle shape and is used to investigate changes in aerosol
 185 morphology. The CALIOP Level 2 product initially provides the volume depolarization ratio (VDR), which contains contributions from both molecules and aerosol particles. To isolate the particulate contribution, VDR was converted to PLDR using the corresponding scattering ratio (SR) and the molecular depolarization ratio δm , according to:

$$PLDR = \frac{VDR(1 + \delta m)(SR + 1) + (VDR - \delta m)}{(1 + \delta m)(SR - 1) - (VDR - \delta m)} \quad (2)$$

190 where $\delta m = 0.0037$ is the molecular depolarization at 532 nm. The conversion was applied for $SR > 1$, ensuring a positive particulate contribution to the backscatter signal. Low PLDR values are generally associated with more spherical particles,



whereas higher values indicate an increasing contribution from non-spherical particles. PLDR is therefore particularly useful for monitoring changes in particle morphology during the early evolution of volcanic and wildfire aerosol layers.

195 The color ratio (CR), defined as the ratio of particulate backscatter at 1064 nm to that at 532 nm, provides information on the wavelength dependence of aerosol backscatter and is sensitive to particle size. In general, CR increases with increasing particle size: small particles exhibit a stronger wavelength dependence of backscatter and therefore lower CR values, whereas larger particles exhibit weaker spectral dependence and CR values approaching unity in the geometric-optics regime (Vaughan et al., 2010). However, because CR also depends on particle composition, morphology, and refractive index, it is interpreted here as
 200 a size-sensitive optical indicator rather than as a direct measurement of particle size, and is analyzed together with PLDR and SR.

The combined evolution of SR, PLDR and CR is therefore used to characterize changes in aerosol loading, particle morphology and size-sensitive optical properties during the first three months following stratospheric injection.

205 **3 Results**

3.1 Temporal evolution of stratospheric plume's optical parameters

The CALIOP Level 2 Layer product identifies individual aerosol layers and provides layer-integrated optical properties. The volcanic and wildfire events investigated here were identified in Part I from their large-scale perturbation of the stratospheric aerosol layer over the CALIPSO observational period. Here, we focus on the first 90 days following these injections to examine
 210 the early evolution of their optical properties at the individual-layer scale.

Figure 1 shows the temporal evolution of the optical parameters averaged across the aerosol plumes for a selection of major volcanic eruptions and wildfire events. The events were selected based on the magnitude and clear detectability of their stratospheric aerosol perturbation, while also spanning the range of aerosol persistence identified in Part I. This selection
 215 includes volcanic and wildfire perturbations with contrasting optical properties and enables their early evolution to be compared during the first months following injection. Figure 1 highlights substantial differences among the investigated stratospheric perturbations.

A key distinction emerges when comparing the temporal evolution of the volume depolarization ratio (VDR; Fig. 1a) and the particulate linear depolarization ratio (PLDR; Fig. 1b). The VDR shows a systematic decrease for all events, reflecting not
 220 only particle evolution but also the increasing relative contribution of molecular backscatter as the plumes dilute. This convergence toward lower VDR values can therefore mask differences in the intrinsic depolarization properties of the aerosol particles.



In contrast, the PLDR isolates the particulate contribution and reveals distinct evolutionary behaviours (Fig. 1b). Volcanic
 225 plumes generally display a decrease in PLDR, consistent with the progressive removal of non-spherical particles and an
 increasing contribution from more spherical aerosol populations (Jégou et al., 2013; Prata et al., 2017). Wildfire plumes exhibit
 a different behaviour, with initially increasing PLDR values during the early stages of plume evolution (Haarig et al., 2018;
 Ohneiser et al., 2023). At later stages, PLDR gradually decreases. These contrasting behaviours highlight substantial
 differences in the evolution of volcanic and wildfire particle morphology among the investigated perturbations.

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The PLDR statistics (Fig. 1b) further allow the events to be grouped according to their dominant aerosol type. Ash-dominated
 eruptions such as Calbuco, Puyehue, and Raikoke exhibit the highest depolarization values associated with irregular ash
 particles and their gradual removal by sedimentation. Intermediate cases such as Kasatochi and Sarychev show mixed ash–
 sulfate signatures reflecting progressive coating processes. Hunga forms a distinct sulfate-dominated end-member,
 235 characterized by very low PLDR values indicative of nearly spherical liquid droplets. Wildfire events, represented here by
 PNE and ANY, fall outside this volcanic continuum and are characterized by broader PLDR distributions reflecting a large
 morphological diversity of pyroCb-emitted aerosols.

The color ratio (Fig. 1c) shows a general decrease with time for all investigated events, with the strongest changes occurring
 240 during the first weeks following injection before values tend to stabilize. Hunga exhibits the highest initial CR, reaching
 approximately 0.43, followed by a rapid decrease during the first weeks. The other volcanic and wildfire perturbations show
 lower initial CR values, but follow a broadly similar decreasing evolution. Puyehue and PNE maintain comparatively high CR
 values during part of their evolution, whereas Calbuco exhibits one of the strongest decreases during the first month. After
 approximately one month, most events converge toward CR values of about 0.15–0.20, although some event-to-event
 245 variability persists. This decrease in CR is consistent with a reduction in particle size, since CR generally increases with
 increasing particle size. The progressive decrease therefore suggests an increasing contribution from smaller particles as the
 plume evolves.

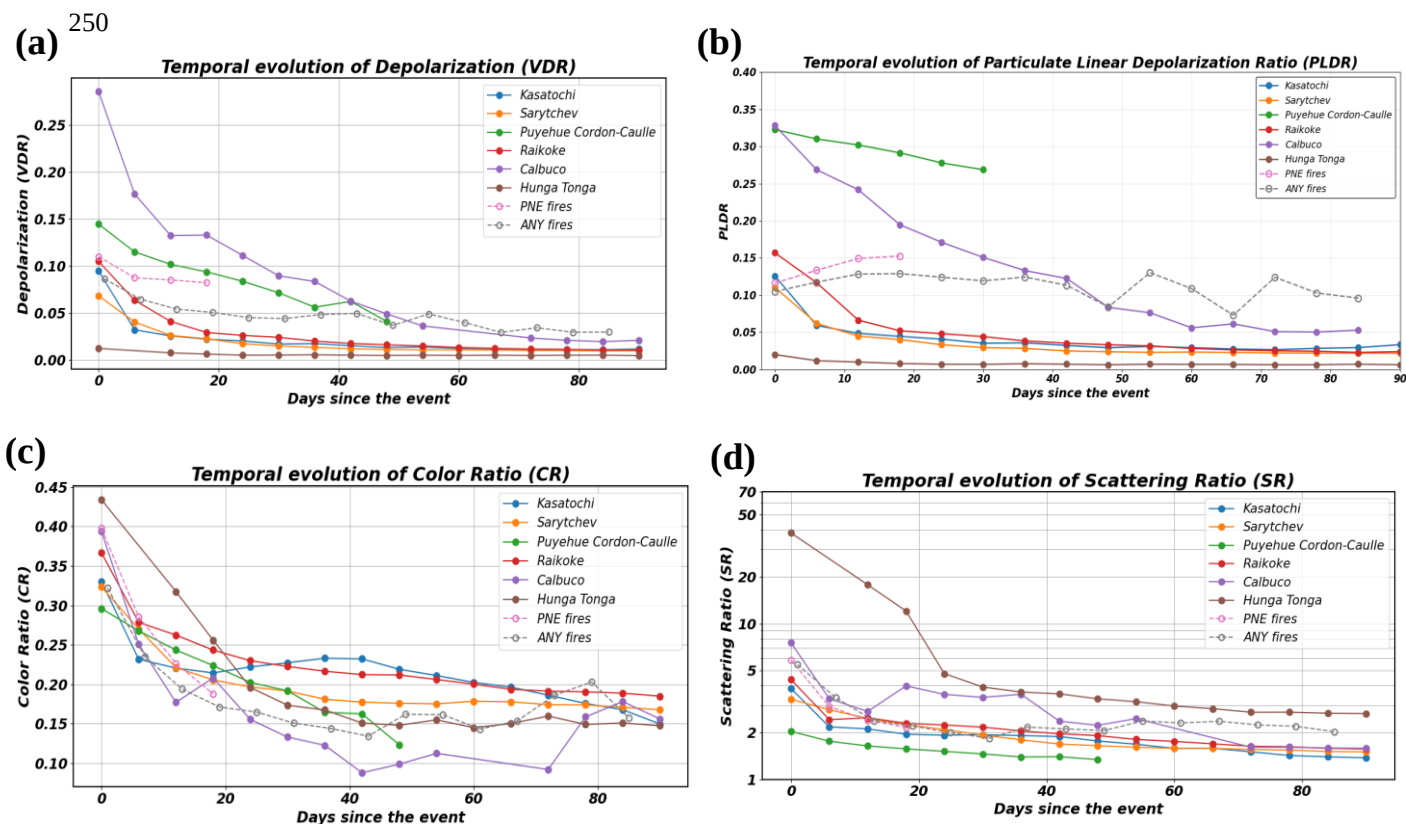


Figure 1: Temporal evolution of stratospheric aerosol optical properties following major volcanic eruptions and forest fires,

(a) Evolution of the volume depolarization ratio (VDR) during the first 90 days after eruption for volcanic eruptions and forest fires showing the type of particles. **(b)** Evolution of the particulate linear depolarization ratio (PLDR) during the first 90 days after eruption for volcanic eruptions and forest fires showing the type of particles. **(c)** Temporal evolution of the color ratio (CR) at 1064/532 nm for the same eruptions, indicating particle growth and presence of ash and sulfate aerosols. **(d)** Evolution of the scattering ratio (SR) at 532 nm highlighting the relative intensity and persistence of the stratospheric aerosol plumes from each event.

The scattering ratio (Fig. 1d) highlights differences in the aerosol concentration among the investigated stratospheric plumes. Hunga clearly stands out as the strongest concentration, exhibiting the highest SR values among all events. The wildfire events PNE and ANY also show strong scattering, particularly during the early stages of plume evolution. Kasatochi and Sarytchev exhibit intermediate SR values, whereas Puyehue shows comparatively weaker and shorter-lived enhancements. Raikoke displays an intermediate but relatively persistent SR signal. Overall, SR generally decreases with time as the aerosol plumes progressively dilute, although the magnitude and rate of this decrease vary substantially among events.

Overall, the combined evolution of PLDR, CR and SR reveals distinct and event-dependent changes in particle morphology, size-sensitive optical properties and aerosol scattering during the first months following stratospheric injection.



3.2 Statistical distribution of stratospheric aerosol plumes optical properties

285 The density distributions shown in Figure 2 provide additional insight into the variability of the optical properties across the stratospheric injections by highlighting how often specific values occur in the aerosol plumes. In several cases, the distributions exhibit bimodal structures, suggesting the transition between early injection conditions and the subsequent aged plume state. For the scattering ratio (Fig. 2d), Hunga displays the broadest distribution, extending toward the highest SR values and indicating a particularly large variability in aerosol scattering. Calbuco shows a narrower distribution centered on more moderate SR values, whereas the wildfire events exhibit broader intermediate distributions. These differences indicate substantial variability in both the magnitude and range of aerosol scattering among the investigated perturbations.

Ash-rich eruptions (Fig. 2b), such as Calbuco and Puyehue extend toward higher depolarization values, reflecting irregular mineral particles, whereas mixed ash–sulfate cases display broader distributions indicative of the diverse morphological properties and progressive coating. Wildfire plumes show wide PLDR variability associated with aggregation processes that generate irregular carbonaceous clusters before gradual relaxation as the plume ages. Hunga remains confined to very low PLDR values, consistent with the dominance of spherical sulfate droplets.

The CR (Fig. 2c) distributions further highlight differences in the size-sensitive optical property of the aerosol populations. Hunga extends toward the highest CR values among all investigated events, consistent with the particularly high values observed during the earliest stage of plume evolution (APARC, 2025, Boichu et al., 2023). Several events exhibit broad or bimodal CR distributions, particularly the mixed aerosol cases, suggesting the presence of two distinct particle populations, likely corresponding to larger ash particles and smaller sulfate aerosols. Puyehue also extends toward relatively high CR values, consistent with the substantial contribution of coarse ash particles. The wildfire events exhibit broad CR distributions, reflecting a large variability in smoke particle size and its evolution.

These statistical patterns confirm that the differences identified in the temporal analysis correspond to robust and recurrent optical regimes rather than isolated features, emphasizing the fundamentally different evolution pathways of ash, sulfate, and smoke aerosols in the stratosphere.

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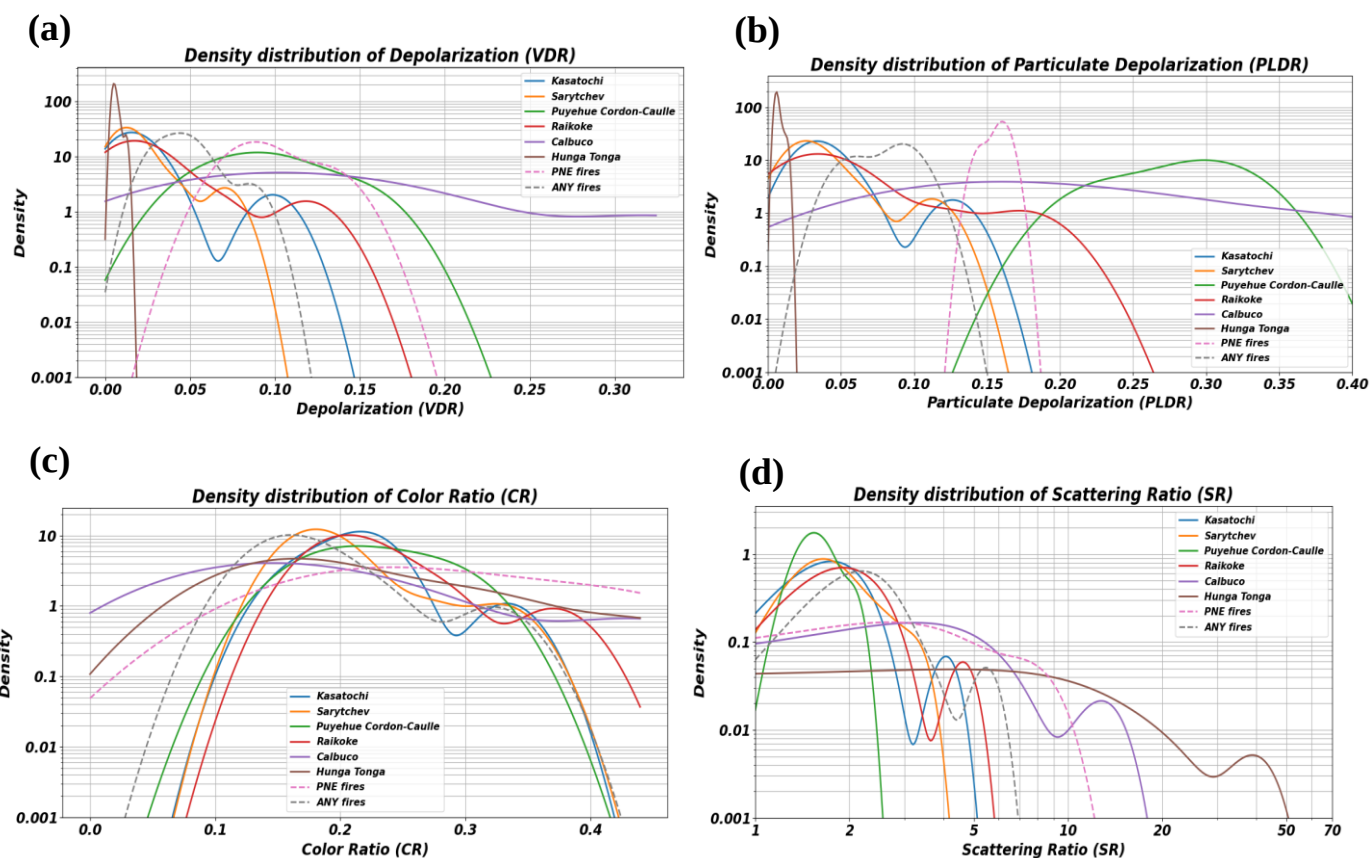


Figure 2: Statistical distributions of optical properties during the first 90 days following major volcanic eruptions and wildfire events. (a) Density distribution of the volume depolarization ratio (VDR) at 532 nm. (b) Density distribution of the particulate linear depolarization ratio (PLDR) at 532 nm. (c) Density distribution of the color ratio (CR), defined as the ratio of particulate backscatter at 1064 and 532 nm. (d) Density distribution of the scattering ratio (SR) at 532 nm. All parameters are derived from the CALIOP Level 2 aerosol layer product.

These statistical patterns confirm that the differences identified in the temporal analysis correspond to robust and recurrent optical regimes rather than isolated features, emphasizing the fundamentally different evolution pathways of ash, sulfate, and smoke aerosols in the stratosphere.

3.3 Vertical evolution and optical properties of volcanic and wildfire plumes

This section investigates the vertical (adiabatic) evolution of volcanic and wildfire aerosol plumes and their optical properties as a function of time and potential temperature. Potential temperature is used because it is conserved during adiabatic motion



in the stratosphere, allowing isentropic transport to be distinguished from diabatic vertical motions, such as the upward
350 displacement of wildfire smoke layers or the subsidence of volcanic aerosol layers.

The Level 2 data are treated using a binning method, which groups individual CALIOP data on aerosol layers into segments defined by time and potential temperature. In this work, a (1-day $\times$ 10 K) binning grid was applied. Each bin contains all valid measurements within the specified day and potential temperature interval, including all samples within event-specific latitude
355 bands selected from the large-scale time–latitude evolution of each stratospheric aerosol perturbation identified in Part I of the study. The selected latitude bands were chosen to encompass the main spatial extent of each plume while limiting contributions from unrelated aerosol perturbations. The latitude range used for each event is reported in Part I, Table 1. Potential temperature is calculated from the meteorological data provided in the CALIOP Level 2 product. This approach minimizes noise and enhances the signal-to-noise ratio for weak stratospheric signals.

360 Figures 3–5 present the evolution of three key lidar-derived parameters used to characterize the stratospheric aerosol plumes: SR, PLDR, and CR. Together, these complementary diagnostics provide information on aerosol scattering, particle morphology, and size-sensitive optical properties for the Calbuco (2015), Raikoke (2019), Australian New Year wildfires (ANY, 2019–2020), and Hunga (2022) events. These events were selected because they represent some of the strongest
365 stratospheric aerosol perturbations observed during the CALIPSO period and encompass volcanic and wildfire-driven injections with contrasting optical and vertical evolution.

3.3.1 Scattering ratio: concentration and vertical evolution

The scattering ratio primarily reflects the magnitude of aerosol backscatter relative to the molecular backscatter and therefore
370 provides information on the aerosol concentration in the plume.

Calbuco plume (Fig. 3a) shows a strong SR enhancement immediately after the eruption, followed by a progressive weakening and sedimentation the aerosol layer. This evolution is consistent with the progressive removal and sedimentation of volcanic ash particles observed following ash-rich eruptions (Prata et al., 2017). A more limited self-lofting behaviour is also observed
375 for the Calbuco plume. Following the initial injection, part of the aerosol layer shows a moderate increase in potential temperature before the subsequent evolution becomes dominated by sedimentation. Compared with the pronounced ascent observed for Raikoke eruption and especially the ANY smoke plume, the weaker vertical ascent of Calbuco plumes indicates that radiatively driven lofting can also occur in volcanic aerosol layers, although with substantially lower efficiency.

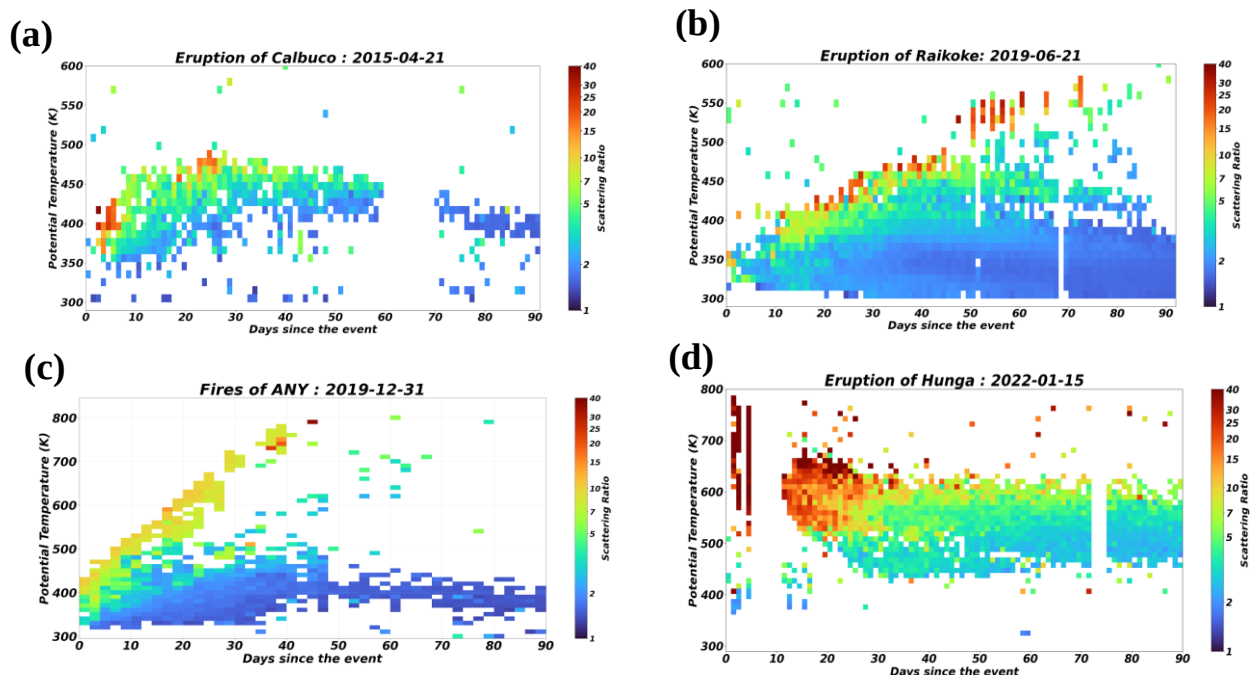


Figure 3: Temporal evolution of aerosol scattering ratio as a function of potential temperature during the first 90 days following selected volcanic eruptions and wildfire events. Scattering ratio (SR) at 532 nm is shown as a function of days since the event and potential temperature for (a) Calbuco (2015), (b) Raikoke (2019), (c) the Australian New Year wildfires (ANY; 2019–2020), and (d) Hunga (2022). The analysis is based on CALIOP Level 2 aerosol layer observations; the filtering criteria and latitude ranges used for each event are described in Sect. 2.1.

Raikoke plume (Fig. 3b) exhibits a different behaviour, with enhanced SR values persisting over a longer period and an upward progression of the most concentrated part of the aerosol plume. This behaviour is associated with the formation of a Vorticed Volcanic Plume (VVP), in which dynamical confinement maintained high aerosol concentrations and enhanced radiative heating. Raikoke is the only volcanic eruption during the CALIOP observation period for which such a VVP has been observed. Importantly, the self-lofting was restricted to this highly concentrated part of the plume, whereas the more diluted parts remained at lower altitudes, as can be inferred from Fig. 3b. The VVP underwent a pronounced diabatic ascent driven by radiative heating of volcanic ash, reaching approximately 27 km within three months (Muser et al., 2020; Gorkavyi et al., 2021; Vaughan et al., 2021; Khaykin et al., 2022).

The ANY wildfire plume (Fig. 3c) exhibits the clearest self-lofting, with the region of enhanced SR progressively reaching higher potential temperatures while remaining detectable over an extended period. This sustained ascent results from



radiatively driven self-lofting of absorbing carbonaceous aerosols and is reinforced by the dynamical confinement of the smoke
415 within a persistent smoke-charged vortex (SCV; Khaykin et al., 2020; Sellitto et al., 2022; Podglajen et al., 2024). The SCV
limits the dilution and dispersion of the smoke plume, maintaining high aerosol concentrations within the confined air mass
during its ascent. Since SR increases with aerosol backscatter and therefore reflects the aerosol concentration within the plume,
this confinement explains the persistently high SR values observed during self-lofting. The combination of high aerosol
loading, enhanced solar absorption, radiative heating, and dynamical confinement therefore sustains the progressive ascent of
420 the ANY smoke plume.

In contrast, the Hunga plume (Fig. 3d) exhibits a pronounced settling of the aerosol layer in potential-temperature coordinates.
This behaviour differs from the sustained self-lofting observed for absorbing smoke plumes. The evolution of volcanic aerosol
layers is generally expected as particles undergo gravitational sedimentation. In the case of Hunga, however, this descent was
425 enhanced by the exceptional amount of water vapour injected into the stratosphere, which strongly affected the formation and
microphysical evolution of the sulfate aerosols. The enhanced water vapour promoted rapid sulfate formation and particle
growth, while radiative cooling associated with the moist plume further contributed to its early downward motion (Schoeberl
et al., 2022; Legras et al., 2022; APARC, 2025).

Overall, the SR distributions reveal contrasting dynamical behaviours among the four perturbations, ranging from subsidence
430 to sustained self-lofting. The strongest and most persistent self-lofting is observed for the ANY wildfire plume, whereas Hunga
exhibits marked subsidence, highlighting the close coupling between aerosol optical loading, radiative effects, and the vertical
evolution of stratospheric aerosol layers.

3.3.2 Particulate linear depolarization ratio: particle morphology and phase

In Figure 4a Calbuco plume displays high PLDR values during the early stage of the perturbation, indicating the presence of
435 strongly non-spherical ash particles. These high-depolarization layers progressively weaken as the plume evolves,
consistent with the preferential sedimentation of the largest ash particles and the progressive transition toward a more spherical
aerosol population (Prata et al., 2017).

Raikoke plume (Fig. 4b) also exhibits elevated PLDR values during the early stages of plume evolution, but the decrease is
440 more gradual than for Calbuco. The depolarizing aerosol layer remains detectable over an extended period and partly follows
the self-lofting identified in the SR distribution. This behaviour is consistent with the persistence and progressive
transformation of fine ash particles within the plume (Ruyneau de Saint Georges et al., 2026), as well as with the self-lofting
and dynamical confinement of the Raikoke aerosol layer reported by Khaykin et al. (2022).



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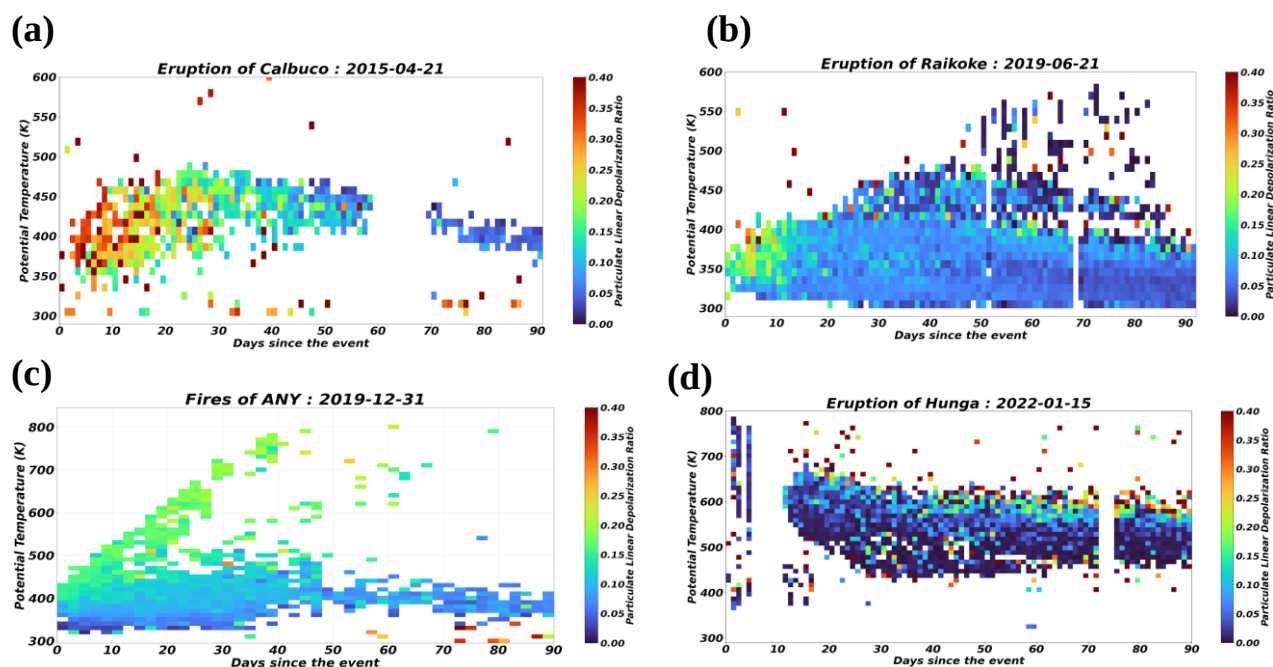


Figure 4: Temporal evolution of particulate linear depolarization ratio (PLDR) at 532 nm as a function of potential temperature during the first 90 days following selected volcanic eruptions and wildfire events. PLDR is shown as a function of days since the event and potential temperature for (a) Calbuco (2015), (b) Raikoke (2019), (c) the Australian New Year wildfires (ANY; 2019–2020), and (d) Hunga (2022). The analysis is based on CALIOP Level 2 aerosol layer observations; the filtering criteria and latitude ranges used for each event are described in Sect. 2.1.

The ANY wildfire plume (Fig. 4c) exhibits a different behaviour. Elevated PLDR values persist in the self-lofting part of the aerosol plume as it progressively reaches higher potential temperature levels. This highly concentrated part of the plume, characterized by enhanced SR, provides favourable conditions for particle aggregation, which can contribute to maintaining elevated PLDR values during the early stages of self-lofting. Previous lidar observations of the ANY plume reported similarly high PLDR values during its early stratospheric evolution, followed by a progressive decrease as the aerosol aged (Ohneiser et al., 2020, 2022).

In contrast, the Hunga plume (Fig. 4d) is characterized by very low PLDR values throughout most of the observed aerosol layer, indicating a predominantly spherical particle population. This behaviour is consistent with the progressive dominance of nearly spherical sulfate aerosols following the initial phase of the eruption (Legras et al., 2022). However, the uppermost part of the Hunga plume exhibits distinctly higher PLDR values, indicating the presence of a more strongly depolarizing particle population at higher potential temperatures. This upper layer likely reflects a different aerosol composition associated



with the early evolution of the Hunga plume, potentially including coated ash, in contrast to the predominantly sulfate-rich aerosol population observed below.

Overall, the PLDR distributions reveal contrasting morphological and vertical evolutions among the four events. Calbuco and Raikoke are characterized by initially depolarizing volcanic aerosol populations whose PLDR decreases during plume evolution, ANY maintains substantial depolarization during the self-lofting of the plume confined within the SCV, whereas Hunga is characterized by a weakly depolarizing and predominantly spherical aerosol population.

3.3.3 Color ratio: particle size and microphysical pathways

The CR is a size-sensitive optical parameter derived from the wavelength dependence of particulate backscatter and is therefore used here to investigate particle size evolution within the vertically evolving aerosol layers (Fig. 5).

For the Calbuco plume (Fig. 5a), the CR distribution shows a clear variability with potential temperature. Higher CR values are mainly associated with the aerosol observed at higher potential temperatures, indicating a greater contribution from relatively large particles, whereas lower CR values are found in the part of the plume extending toward lower potential temperatures.

Raikoke plume (Fig. 5b) exhibits a distinct evolution, with relatively high CR values persisting within part of the aerosol layer as it reaches higher potential temperatures. The persistence of these size-sensitive signatures within the self-lofting plume suggests that relatively large particles were maintained during its ascent. This behaviour is further supported by Ruynéau de Saint-George et al. (2026), who reported pronounced particle growth within the VVP, with fine-mode particle radii reaching 0.8–1 μm within three months after the eruption. This growth was attributed to sulfate formation and coating of submicronic ash and was substantially stronger within the highly concentrated VVP than in the dispersed plume. The persistence and growth of relatively large particles within the VVP may therefore contribute to the high CR values observed during the self-lofting phase.

The ANY wildfire plume (Fig. 5c) similarly maintains relatively high CR values within the ascending aerosol layer. Combined with the elevated PLDR observed in Fig. 4, this behaviour indicates the persistence of relatively large, non-spherical smoke particles during self-lofting. Such characteristics are consistent with particle aggregation within aged stratospheric wildfire smoke, while radiative heating and dynamical confinement allow the resulting particles to remain within the ascending smoke-charged vortex (Khaykin et al., 2020; Ohneiser et al., 2023).

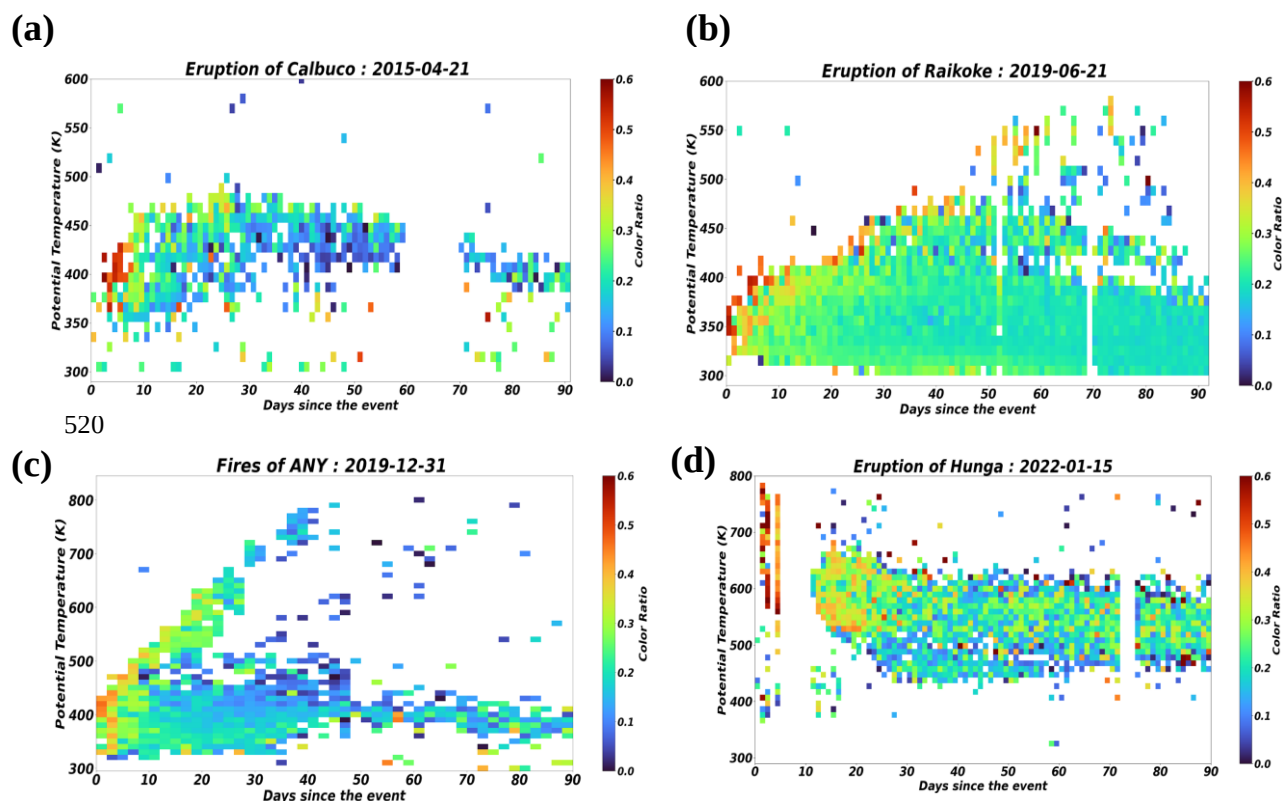


Figure 5: Temporal evolution of the color ratio (CR) as a function of potential temperature during the first 90 days following selected volcanic eruptions and wildfire events. The color ratio, defined as the ratio of particulate backscatter at 1064 nm to that at 532 nm, is shown as a function of days since the event and potential temperature for (a) Calbuco (2015), (b) Raikoke (2019), (c) the Australian New Year wildfires (ANY; 2019–2020), and (d) Hunga (2022). The analysis is based on CALIOP Level 2 aerosol layer observations; the filtering criteria and latitude ranges used for each event are described in Sect. 2.1.

Hunga plume (Fig. 5d) exhibits the highest CR values during the earliest stage of plume evolution, followed by a pronounced decrease with time. The initially high CR is consistent with the presence of relatively large ice and ash particles within the young plume following the exceptional injection of water vapour and volcanic material into the stratosphere (Khaykin et al., 2022; Legras et al., 2022). As these transient coarse components are progressively removed, CR decreases and the aerosol population evolves toward smaller, predominantly sulfate particles (Legras et al., 2022; APARC 2025).

Overall, CR generally decreases as the aerosol layers age, consistent with the preferential removal of larger particles. However, the persistence of relatively high CR values within the ascending Raikoke and ANY plumes indicates that comparatively large particles can remain associated with self-lofting aerosol layers, through coated ash in the volcanic case and aggregated smoke



545 particles in the wildfire case. Hunga represents a distinct initial regime, with particularly high CR associated with the presence of ice and ash in the young plume.

3.4 Panboreal wildfire outbreak observations by EarthCare/ATLID

Beginning in May 2025, intense boreal wildfires generated multiple pyroconvective events (pyroCbs) across the northern extratropics, with stratospheric smoke injections occurring over both Canada and Siberia. Because the resulting stratospheric
 550 perturbation originated from several distinct pyroCb injections rather than from a single fire event, it is referred to here as the Panboreal Wildfire Outbreak (PWO). Among these injections, the strongest stratospheric injection over Canada occurred on 29 May 2025 in Alberta and involved several pyroCb pulses, producing an exceptionally dense stratospheric smoke plume (Haarig et al., 2026; Vieille et al., 2026). Part of this plume subsequently became confined within a smoke-charged vortex (SCV), which underwent pronounced diabatic self-lofting during its transport across the Northern Hemisphere. The SCV
 555 persisted for approximately one month, travelled about 30,000 km and reached an altitude of 21 km (Vieille et al., 2026). These multiple injections and the subsequent self-lofting of the main Canadian plume provide a unique opportunity to investigate the early optical and vertical evolution of the PWO using observations from the recently launched ATLID instrument aboard the EarthCARE mission.

560 The Level 2 data are treated using a binning method similarly to CALIOP-based analysis. To restrict the analysis to well defined stratospheric aerosol layers, several selection criteria were applied to the ATLID L2A EBD product. Only layers classified as stratospheric aerosols with good retrieval quality were retained. Additional filters were applied to remove weak or poorly constrained detection: $AOD > 0.03$, $0 < PLDR < 0.4$, a layer base located at least 0.25 km above the tropopause, and a minimum layer thickness of 0.75 km. The ATLID BA baseline is affected by a known daytime bias in PLDR. To account for this effect,
 565 the daytime PLDR bias was estimated and subsequently subtracted from the daytime PLDR values. No corresponding correction was applied to nighttime observations. To further improve the robustness of the retrievals, only observations with $SR > 1.5$ during nighttime and $SR > 2$ during daytime were considered. These criteria ensure that the analysis is restricted to well defined stratospheric aerosols layers while minimizing contamination from tropospheric aerosols, clouds and noisy retrievals.

570 Figure 7 presents the evolution of SR and PLDR at 355 nm as a function of time and potential temperature, together with the temporal evolution of PLDR for the PNE, ANY and PWO wildfire events.

The ATLID scattering-ratio observations (Fig. 6a) reveal a pronounced self-lofting of the smoke plume during the weeks following injection. The region of maximum aerosol scattering progressively reaches higher potential temperatures during the first month, the PWO plume ascends from approximately 310 to 480 K, corresponding to a mean diabatic self-lofting rate of
 575 approximately $4.3 \pm 0.06 \text{ K.day}^{-1}$. This rate is substantially lower than that reported for the highly stable ANY smoke-charged vortex, which ascended at an approximately constant rate of $6.9 \pm 0.07 \text{ K.day}^{-1}$, and $3.4 \pm 0.04 \text{ K.day}^{-1}$ for PNE fires. The lower self-lofting rate observed for PWO is consistent with a less stable dynamical confinement of the smoke plume compared

with the exceptionally persistent ANY vortex. This behaviour is linked with radiatively driven self-lofting of absorbing smoke particles, previously documented for major pyroCb events (Yu et al., 2019; Khaykin et al., 2020). Independent ATLID observations of the same 2025 Canadian smoke event by Haarig et al. (2026) show that the top of the stratospheric smoke layer ascended from approximately 13.6 km over Canada to 17.4 km over Europe during transport, supporting the self-lofting identified here.

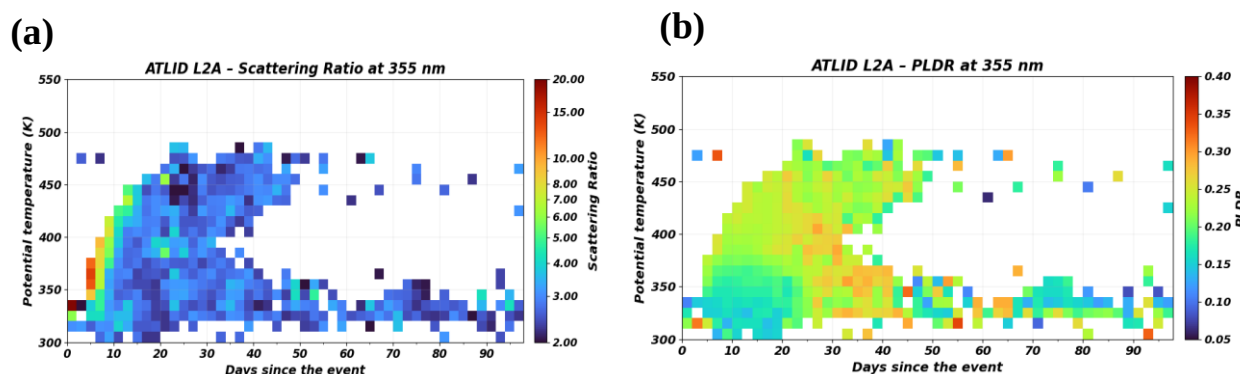


Figure 6: Optical and vertical evolution of stratospheric wildfire smoke observed during the 2025 Panboreal Wildfire Outbreak (PWO) (20°N–80°N). (a) ATLID scattering ratio (SR) at 355 nm as a function of potential temperature and time following the PWO injection, showing the vertical evolution of the smoke layer. **(b)** ATLID particulate linear depolarization ratio (PLDR) at 355 nm as a function of potential temperature and time.

The evolution of PLDR as a function of potential temperature (Fig. 6b) shows that elevated depolarization remains associated with the self-lofting smoke plume. Between approximately 20 and 60 days after the event, the aerosol layer progressively ascends from about 320–330 K to 450–480 K while maintaining PLDR values predominantly around 0.20–0.25. This indicates that strongly depolarizing, non-spherical smoke particles persist within the plume throughout its self-lofting phase, without a clear decrease in PLDR with increasing potential temperature. Recent ATLID observations of the Canadian plume similarly reported enhanced depolarization associated with non-spherical stratospheric smoke particles (Haarig et al., 2026).

The comparison of PLDR evolution for the PNE, ANY and PWO wildfire events (Fig. 7) reveals a common increase in particulate depolarization during the early stages of plume evolution. Differences in absolute PLDR values between PWO and the PNE and ANY events should, however, be interpreted with caution because PWO is observed by ATLID at 355 nm, whereas PNE and ANY are observed by CALIOP at 532 nm. Multiwavelength lidar observations of stratospheric wildfire smoke have shown a pronounced spectral dependence of PLDR, with systematically higher values at 355 nm than at 532 nm (Hu et al., 2018; Haarig et al., 2018). Despite this wavelength dependence and differences in the duration of elevated PLDR among the three events, the increase in PLDR indicates a progressive evolution toward more irregular particle morphologies

within the smoke layers. This common behaviour is consistent with aggregation processes during smoke ageing, whereby particle–particle interactions can produce increasingly complex and non-spherical structures (Haarig et al., 2018; Ohneiser et al., 2023). The persistence of elevated depolarization during vertical transport therefore appears to represent a recurrent optical characteristic of major stratospheric wildfire plumes.

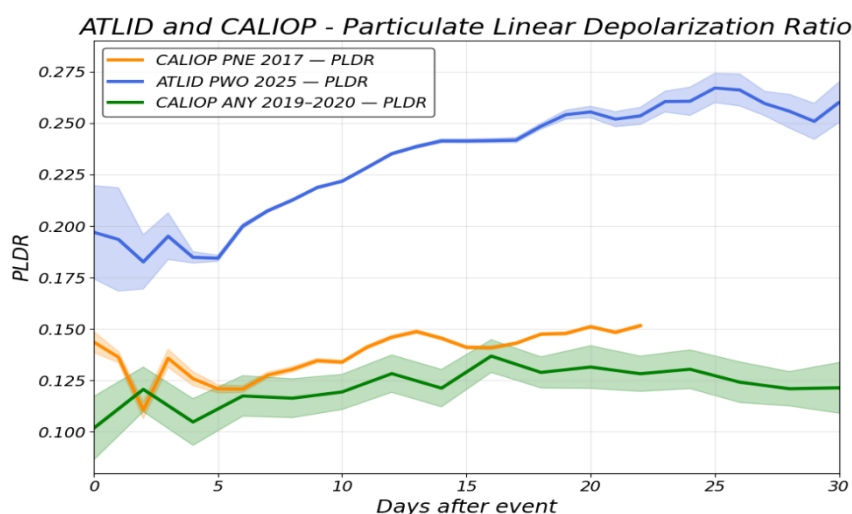


Figure 7: Temporal evolution of PLDR during the first 30 days following the PNE (2017), ANY (2019–2020), and PWO (2025) wildfire events, derived from CALIOP at 532 nm for PNE and ANY and from ATLID at 355 nm for PWO.

Overall, the ATLID observations show that the characteristic optical and vertical behaviour identified for previous major wildfire plumes with CALIOP is also observed during the 2025 Panboreal Wildfire Outbreak. The ATLID HSRL observations provide an independent characterization of stratospheric smoke at 355 nm, revealing pronounced self-lofting together with an evolving particulate depolarization signature. The similarities observed among PNE, ANY and PWO further suggest a common microphysical evolution of major pyroCb smoke plumes despite differences in their injection conditions and atmospheric transport.

4 Discussion and summary

4.1 Optical evolution and microphysical interpretation of volcanic and wildfire plumes

The analysis of young stratospheric aerosol plumes reveals markedly different optical evolutions during the first three months following injection. Previous studies have generally investigated these perturbations individually, documenting the evolution of specific volcanic eruptions or wildfire events. Here, the application of a common CALIOP-based framework to multiple



perturbations allows their early evolution to be directly compared through PLDR, CR, SR, their statistical distributions, and their vertical evolution in potential-temperature coordinates.

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The observed diversity indicates that the early evolution of stratospheric aerosol plumes cannot be described solely according to aerosol source, but depends strongly on the initial composition of the injected material and its subsequent microphysical and dynamical transformation. This is consistent with the results of Part I, which showed that the persistence of the resulting large-scale stratospheric aerosol perturbation, characterized by its return and decay times, cannot be explained solely by the amount of injected material or the initial injection altitude.

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The PLDR evolution illustrates these differences particularly clearly for volcanic perturbations. Ash-rich eruptions such as Puyehue, Calbuco, and Raikoke exhibit initially elevated depolarization followed by a progressive decrease during the following weeks. High depolarization is a well-established optical signature of non-spherical volcanic ash (Nakamae et al., 2014; Vernier et al., 2016; Prata et al., 2017), whereas its subsequent decrease reflects the progressive transformation of the aerosol population. Gravitational sedimentation preferentially removes the largest and most irregular ash particles because of their higher settling velocities, progressively reducing their contribution to the lidar signal. Simultaneously, oxidation of injected SO₂ produces H₂SO₄, followed by nucleation and condensation leading to the formation and growth of nearly spherical sulfate droplets. The increasing relative contribution of these weakly depolarizing particles further decreases PLDR as the plume ages (Jégou et al., 2013; Prata et al., 2017). The optical evolution of ash-containing volcanic plumes therefore reflects the combined effects of preferential ash sedimentation and the progressive development of a sulfate aerosol population. Similar transitions have been documented following Sarychev, where a persistent stratospheric sulfate aerosol layer developed after the initial volcanic injection (Jégou et al., 2013), and Kasatochi, where SO₂, ash, and H₂SO₄ were observed within the evolving volcanic plume (Karagulian et al., 2010).

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The bimodal PLDR distributions observed for Kasatochi and Sarychev further demonstrate that this transition does not necessarily correspond to a single continuously evolving aerosol population. Distinct depolarization modes indicate the coexistence of optically different particle populations, consistent with the simultaneous presence of volcanic ash and newly formed sulfate-containing aerosols following sulfur-rich eruptions (Karagulian et al., 2010; Jégou et al., 2013). The evolution from ash-rich toward sulfate-rich conditions should therefore be regarded as a progressive process during which different aerosol populations can coexist within the volcanic plume.

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Raikoke plume provides a particularly clear example of an additional microphysical pathway. Comparatively high CR values persist within part of the plume while PLDR progressively decreases, indicating that the particles remain comparatively large while becoming less depolarizing. This behaviour is consistent with sulfate coating of the remaining ash particles. Condensation of sulfuric acid and sulfate material onto irregular ash particles can increase their size while progressively

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smoothing their initially irregular surfaces, thereby maintaining or increasing their size-sensitive optical response while decreasing depolarization. This interpretation is supported by Ruynau de Saint Georges et al. (2026), who reported pronounced particle growth within the Raikoke VVP associated with sulfate formation and coating of submicronic ash. The coexistence of uncoated ash, progressively coated particles, and newly formed sulfate droplets may additionally contribute to the multiple optical populations observed during the early evolution of mixed ash–sulfate plumes. The dynamical confinement of the concentrated Raikoke aerosol population and its diabatic self-lofting further prolonged its stratospheric residence, providing additional time for these microphysical transformations to occur (Khaykin et al., 2022).

Hunga represents a markedly different volcanic end-member. Its exceptionally high initial CR, followed by a rapid decrease and persistently low PLDR throughout most of the aerosol layer, distinguishes it from the ash-rich volcanic cases. Previous studies have emphasized the exceptional nature of the Hunga eruption, particularly its massive injection of water vapour and the unusual composition of the young plume (Sellitto et al., 2022; Legras et al., 2022; Duchamp et al., 2025). A distinct layer characterized by higher PLDR values was nevertheless observed above the main low-depolarization aerosol plume during its early evolution. Our observations are consistent with an initially complex particle population containing relatively large particles, followed by rapid evolution toward a weakly depolarizing population dominated by nearly spherical particles. Its vertical evolution also differs from the self-lofting observed for strongly absorbing aerosol layers. The exceptionally large water-vapour injection produced a radiative environment fundamentally different from that of smoke or ash-rich plumes, with radiative cooling contributing to diabatic descent across isentropic surfaces.

Wildfire plumes exhibit a contrasting depolarization evolution. Unlike the predominantly decreasing PLDR observed for ash-containing volcanic populations, PNE, ANY, and PWO exhibit persistent and, during their early evolution, increasing PLDR. This behaviour is consistent with previous lidar observations of stratospheric wildfire smoke (Haarig et al., 2018; Hu et al., 2019; Baars et al., 2019; Ohneiser et al., 2020, 2022). For PNE, Haarig et al. (2018) measured particle depolarization ratios of approximately 0.22 at 355 nm and 0.18 at 532 nm in aged stratospheric smoke from the 2017 Canadian fires, while similarly enhanced depolarization was reported by Hu et al. (2019) and Baars et al. (2019). For ANY, multiwavelength lidar observations over Punta Arenas revealed similarly high PLDR values, reaching approximately 0.20–0.26 at 355 nm and 0.14–0.22 at 532 nm during January 2020 (Ohneiser et al., 2020), with subsequent observations documenting the evolution of the depolarization signature during plume ageing (Ohneiser et al., 2022). The recurrence of persistent or initially increasing PLDR across PNE, ANY, and PWO therefore suggests a common microphysical behaviour of major stratospheric pyroCb smoke perturbations. The joint evolution of PLDR and CR provides insight into the processes responsible for this behaviour. Following pyroconvective injection, carbonaceous particles undergo ageing, coagulation, and aggregation within the dense smoke layer. Particle–particle interactions can produce increasingly complex and irregular aggregates, enhancing particulate depolarization during the early stages of plume evolution (Haarig et al., 2018; Ohneiser et al., 2023). As these aggregates grow, however, the largest structures are preferentially removed by gravitational sedimentation because of their higher settling velocities. Their



progressive removal reduces the contribution of the largest and most irregular particles to the lidar signal and can therefore contribute to the subsequent decrease in PLDR. At the same time, preferential sedimentation of the largest aggregates shifts the remaining particle population toward smaller sizes, consistent with the general decrease in CR observed during plume ageing. The wildfire PLDR evolution can thus be interpreted as the result of competing processes: aggregation and morphological restructuring initially increase particle irregularity and depolarization, whereas the subsequent sedimentation of the largest aggregates progressively removes the most strongly depolarizing particles. Continued chemical ageing may further modify the morphology and internal structure of the remaining aerosol population. This sequence provides a coherent interpretation of the initial increase and subsequent evolution of PLDR observed across the wildfire cases.

The optical evolution of these aerosol populations is closely coupled to their vertical transport. Absorbing particles convert incoming solar radiation into heat, producing local diabatic warming and allowing sufficiently concentrated aerosol layers to ascend across isentropic surfaces. For wildfire smoke, absorbing carbonaceous material can drive persistent self-lofting, as demonstrated by modelling and observations of major pyroCb plumes (Yu et al., 2019; Khaykin et al., 2020; Podglajen et al., 2024). Under sufficiently strong conditions, radiative heating can become dynamically coupled to an anticyclonic circulation, producing a smoke-charged vortex capable of confining and transporting the aerosol layer over extended periods (Khaykin et al., 2020). The pronounced ascent observed for ANY is consistent with this mechanism and with the exceptionally stable smoke-charged vortex documented following the 2019–2020 Australian wildfires.

Radiatively driven ascent is not restricted to wildfire smoke. As mentioned previously, volcanic ash also absorbs solar radiation, and sufficiently concentrated ash-containing layers can therefore undergo diabatic heating and upward displacement. Raikoke provides an important example, with part of its volcanic plume undergoing self-lofting while confined within a long-lived anticyclonic structure (Khaykin et al., 2022). The comparable upward displacement of ANY and Raikoke establishes an important connection between otherwise distinct aerosol regimes: despite their different compositions and microphysical pathways, sufficiently absorbing and concentrated aerosol populations can produce similar dynamical responses. Conversely, Calbuco (Prata et al., 2017; Lopes et al., 2019) exhibits an initial ascent of the aerosol layer, with an estimated rate of approximately 3–4 K.day⁻¹ during the first weeks, suggesting limited self-lofting during the early stage of its evolution, followed by a progressive downward displacement. In contrast to Raikoke, this ascent was not associated with a VVP and was not sustained over time. It is conceivable that formation of a VVP associated with Calbuco ash-rich plume could have been hindered by a more perturbed stratospheric dynamical environment at the time of Calbuco eruption in April during Austral Fall, compared with a stable summertime stratosphere at the time of Raikoke eruption in June.

Hunga subsequently undergoes a more pronounced diabatic descent. These contrasting behaviours demonstrate that the vertical evolution of stratospheric aerosol layers cannot be inferred solely from aerosol source or initial injection altitude, but depends on the evolving composition, concentration, radiative properties, and dynamical confinement of the plume.



745 The coupling between microphysics and dynamics can in turn influence aerosol persistence and subsequent transformation. Self-lofting and dynamical confinement can maintain aerosol layers at high altitudes and increase the time available for coating, aggregation, chemical ageing, and other microphysical processes, whereas sedimentation and dilution progressively reduce aerosol concentration and modify the particle population. Microphysical evolution and vertical dynamics should therefore not be considered independently: changes in particle properties affect the radiative response of the plume, while the resulting
750 vertical transport and confinement determine the conditions and timescales over which further particle transformation can occur.

Taken together, these results demonstrate that the early evolution of stratospheric aerosol perturbations is controlled by coupled microphysical and dynamical processes rather than by aerosol source alone. Volcanic plumes span a continuum from ash-rich
755 to sulfate-dominated populations, in which preferential sedimentation, sulfate formation, and coating produce distinct and sometimes coexisting optical signatures. Wildfire plumes follow a different pathway, characterized by aggregation and morphological restructuring that initially enhance depolarization, followed by preferential sedimentation of the largest aggregates as the plume ages. At the same time, the potential-temperature analysis demonstrates that these microphysical transformations are closely coupled to plume dynamics: sufficiently absorbing and concentrated smoke or ash-containing
760 layers can undergo radiatively driven self-lofting, whereas Hunga illustrates a contrasting downward evolution associated with its exceptional composition. The combined interpretation of SR, PLDR, CR, and potential temperature therefore provides a coherent framework linking aerosol loading, particle morphology and size-sensitive optical properties to the vertical evolution of young stratospheric aerosol plumes.

4.2 Independent constraints from ATLID LiDAR observations

765 The inclusion of ATLID observations provides an important independent perspective on the optical properties and vertical evolution of stratospheric wildfire aerosols and extends the CALIOP-based analysis to the EarthCARE era. Unlike the elastic-backscatter measurements provided by CALIOP, ATLID operates as a high-spectral-resolution lidar (HSRL) at 355 nm, allowing molecular and particulate backscatter contributions to be separated spectrally (Wehr et al., 2023). The PWO 2025 event therefore provided an opportunity to assess whether the characteristic optical and dynamical signatures identified from
770 previous CALIOP observations are reproduced by an independent spaceborne lidar operating at a different wavelength.

The pronounced self-lofting observed during the first weeks following the 29 May 2025 Alberta injection confirms that the aerosol layer did not behave as a passive tracer. The plume reached approximately 21 km during its evolution, due to the radiatively driven self-lofting mechanism previously documented for stratospheric wildfire smoke, in which absorption of
775 solar radiation by carbonaceous particles induces diabatic heating (Yu et al., 2019; Ohneiser et al., 2020). The particularly high SR immediately following the Alberta outbreak of 29 May reflects the strong aerosol scattering associated with the young



plume. Its subsequent decrease while the layer continues to ascend indicates that weakening of the optical signal primarily accompanies plume dilution and dispersion rather than termination of the vertical transport.

780 The PLDR observations provide an additional constraint on the evolution of the smoke particle population. Comparison of
PNE (2017), ANY (2019–2020), and PWO (2025) over the first 30 days following injection reveals a common early increase
in particulate depolarization, despite substantial differences in absolute PLDR values among the events. Similar elevated
depolarization ratios have previously been reported for Canadian wildfire smoke observed at 355, 532, and 1064 nm,
demonstrating that stratospheric smoke can retain substantial non-sphericity for several weeks after injection (Haarig et al.,
785 2018).

However, the absolute PLDR values measured by ATLID and CALIOP should not be directly compared without considering
their different measurement wavelengths. Particle depolarization is wavelength dependent and is sensitive to particle size,
shape, and internal structure, as demonstrated by multiwavelength lidar observations of aerosol populations (Müller et al.,
790 2007; Haarig et al., 2018). ATLID measurements at 355 nm and CALIOP measurements at 532 nm therefore provide
complementary rather than quantitatively interchangeable constraints on smoke morphology. Consequently, the agreement
between the two records is most robustly interpreted in terms of their temporal evolution rather than their absolute PLDR
magnitude.

795 The differences in PLDR magnitude and evolution among PNE, ANY at 532nm and PWO at 355nm; additionally indicate that
stratospheric wildfire smoke cannot be characterized by a unique depolarization signature. Variability in combustion
conditions, fuel characteristics, injection intensity, particle composition, and subsequent atmospheric ageing may all contribute
to the event-to-event differences in particle morphology. Nevertheless, the common early increase in PLDR and the occurrence
of pronounced self-lofting demonstrate that major pyroCb perturbations share recurrent optical and dynamical characteristics
800 despite differences in their initial conditions.

Taken together, the ATLID observations extend aerosol plumes behaviours identified from the CALIOP wildfire cases to the
2025 PWO and provide an independent constraint on the early optical and dynamical evolution of stratospheric smoke. Despite
the different observing wavelength, the persistence of enhanced depolarization and the upward displacement of the aerosol
805 layer are consistent with the morphological evolution and radiatively driven self-lofting previously observed for major pyroCb
perturbations. The comparison with ANY further highlights the role of dynamical confinement in controlling the efficiency of
self-lofting, with the less stable PWO plume exhibiting a weaker ascent than the exceptionally persistent Australian smoke-
charged vortex. These early differences in plume dynamics provide a physical context for the contrasting persistence of wildfire
perturbations documented over longer timescales in Part I, where differences in stratospheric residence time were identified
810 among PNE, ANY, and PWO. ATLID therefore confirms that the early evolution of stratospheric wildfire plumes results from



the coupling between particle microphysical transformation and plume dynamics, while extending this comparative framework beyond the CALIPSO observational period.

Data availability

815 EarthCARE ATLID data are available at <https://earth.esa.int/eogateway/catalog/earthcare-l1-products> (last access: 5 June 2026); CALIPSO CALIOP data are available at <https://www.earthdata.nasa.gov/data/tools/asdc-calipso-search-subset-tool>

Author contributions

SK conceived the study. OS performed the analysis and wrote the manuscript. SK and SGB revised the manuscript. NK transformed CALIOP and ATLID L2 products. All authors contributed to the final manuscript.

820 Competing interests

The contact author has declared that none of the authors has any competing interests.

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Review statement

830 The review statement will be added by Copernicus Publications listing the handling editor as well as all contributing referees according to their status anonymous or identified.



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