

Reviewer 1

We would like to thank the reviewer for taking the time to evaluate our manuscript and provide valuable feedback. We have modified the manuscript with the proposed changes along with step-by-step answers to the suggestions. Please note that changes have been highlighted in the manuscript and the corresponding answers to the reviewer by text below. The original comments are presented in black.

The manuscript presents an investigation into the particle linear depolarization ratio at 1565 nm for key aerosol types, namely extremely fresh smoke, aged smoke, and volcanic ash using Halo Doppler lidars. Three distinct case studies were conducted in Finland and South Africa, making possible to understand of how these particles behave at longer infrared wavelengths, where systematic observations have historically been scarce. Additionally, by using a combination of ancillary in situ instrumentation and modeling tools (including ground- and drone-based sensors, Mie scattering calculations, and air mass trajectory simulations) the authors provide a validation of their findings.

Providing new observational values at 1565 nm is a significant contribution to the field, especially given that depolarization research has traditionally focused on UV and visible spectra. The application of a consistent methodology across diverse environments and aerosol types provides a robust framework for investigating spectral dependence. Furthermore, the inclusion of a detailed review of existing linear particle depolarization values at this specific wavelength serves as a very useful reference for the community.

Given the scientific level of the work and its clear relevance to atmospheric aerosol characterization, I recommend the paper be accepted for publication after addressing some minor revisions to improve the work clarity:

-Line 21: “the ratio of cross- to co-polarized signal” is a poor definition of particle linear depolarization ratio, referred to the backscattered radiation. Please be more precise.

Thank you for your comment. We have revised the sentence to: ‘One such property is the ratio of cross- to co-polarized backscattered signal known as the particle linear depolarization ratio (δ), after removing the molecular contribution.’ in order to be more complete.

-The sentence in lines 48-50 (“They also found that ...”) may seem confusing, suggesting that δ increases with the total aerosol concentration. Do you actually mean that δ depends on the large-particle concentration and its relative contribution to the aerosol mixture? If so, please clarify.

Thank you for pointing this out. Yes, in our previous study, we found that δ increases and depends on the large-particle concentration and its relative contribution to the aerosol mixture. This is because the pollen concentration is rather low in the atmosphere and the effect in δ is non linear.

-Lines 55-64: this paragraph could be better structured (e.g. last sentence justifying one advantage of using 1565 nm), could you try to re-organize the main ideas?

We have restructured the paragraph as suggested. The updated paragraph reads as: ‘Multi-wavelength lidar observations, obtained from single or multiple instruments, enhance the reliability of aerosol characterization and typing. In this context, this study estimates the δ for smoke and volcanic ash particles using a Halo Doppler lidar operating at 1565 nm. Furthermore, we extend the applicability of such lidar systems by retrieving smoke aerosol mass concentration at this wavelength. Such estimates are particularly valuable, as they provide vertically resolved, model-ready information that improves the representation of aerosol loading, plume dynamics, and radiative effects in atmospheric models (Benedetti et al., 2009; Sekiyama et al., 2010; Zhang

et al., 2014). By addressing the current lack of δ observations at 1565 nm, this study supports the integration of this wavelength into operational aerosol monitoring networks, benefiting from the negligible contribution of molecular scattering and absorption, which simplifies signal processing and enhances retrieval robustness.’

-Lines 90-94: although the technical papers containing the main details of the methodology for calibrating and obtaining depolarization from Halo Doppler lidars are conveniently cited in the paper, some general ideas should also be included in the present manuscript for a better understanding. For example, a general definition of the bleed-through parameter and obtained values.

Thank you for your suggestion. We have added the definition of bleed-through to aid readers’ understanding. We are now providing more technical details for the lidar instrument itself and the post-processing procedure.

-Lines 100-101: “Similar assumption. . .” should be re-written to clarify the idea. What do you mean with “which represents the particle backscatter coefficient”? Is it equivalent? Proportional? Under which conditions?

The sentence has been rephrased for clarity to: ‘In the absence of hydrometeors, the atmospheric transmittance is very close to unity therefore, the attenuated backscatter coefficient, hereafter ATB_{1565} , can be used as a proxy for the particle backscatter coefficient, hereafter β_{1565} . This assumption is valid provided that extinction within the sampled aerosol layer remains sufficiently small. Attenuation in dense aerosol layers can lead to an underestimation of the true β_{1565} . For the exceptionally fresh smoke case at Kiviniemi (FI), β_{1565} retrieved using the forward Klett method (Wiegner and Geiß, 2012) was used to estimate the smoke mass concentration, as this quantitative parameter requires an accurate measure of backscatter intensity and could therefore be biased by substantial departures of atmospheric transmittance from unity observed during periods of high plume density. A Lidar Ratio (LR) of 51 sr was used in Klett inversion according to Mie calculations (see Sect. 2.4)’.

Wiegner, M. and Geiß, A.: Aerosol profiling with the Jenoptik ceilometer CHM15kx, Atmos. Meas. Tech., 5, 1953–1964, <https://doi.org/10.5194/amt-5-1953-2012>, 2012.

-Line 141: what do you mean with “default” mass absorption cross-section? Please clarify and include a reference from where the value has been taken.

It is a “default” value because the MAAP instrument manufacturer uses it in the device. We have modified the paragraph to reflect this selection: “To gain a more detailed understanding of the aerosol composition, and particularly the black carbon (BC) and organic aerosol (OA) content during the burning event, ground-based multi-angle absorption photometer (MAAP, model 5012, Thermo Scientific, Petzold and Schönlinner (2004)) and aerosol mass spectrometer (AMS, HR-ToF-AMS, Aerodyne Research Inc., DeCarlo et al. (2006)) measurements from the container (Fig. 1c) were used to estimate the BC-to-organic aerosol ratio (BC/OA). MAAP measured dried aerosol particles from the station’s main inlet at a flow rate of 7.5 LPM at 1-min time resolution. The aerosol absorption determined by MAAP was converted to BC mass concentration using the instrument manufacturer set default mass absorption cross-section (MAC) of $6.6 \text{ m}^2 \text{ g}^{-1}$, which is a commonly accepted value for MAAP and broadly used in scientific literature (e.g. Ohata et al., 2021).”

Ohata, S., Mori, T., Kondo, Y., Sharma, S., Hyvärinen, A., Andrews, E., Tunved, P., Asmi, E., Backman, J., Servomaa, H., Veber, D., Eleftheriadis, K., Vratolis, S., Krejci, R., Zieger, P., Koike, M., Kanaya, Y., Yoshida, A., Moteki, N., Zhao, Y., Tobo, Y., Matsushita, J., and Oshima, N.: Estimates of mass absorption cross sections of black carbon for filter-based absorption photometers in the Arctic, Atmos. Meas. Tech., 14, 6723–6748, <https://doi.org/10.5194/amt->

14-6723-2021, 2021.

Petzold, A. and Schönlinner, M.: Multi-angle absorption photometry – A new method for the measurement of aerosol light absorption and atmospheric black carbon, *J. Aerosol Sci.*, 35, 421–441, <https://doi.org/10.1016/j.jaerosci.2003.09.005>, 2004.

-Line 145: are there any references for the mentioned software tools?

We have updated the text to: 'AMS data analysis was performed using standard tools- SQUIRREL v1.65C and PIKA v1.25C, downloaded from the ToF-AMS-Resources web page (<https://cires1.colorado.edu/group/ToFAMSResources/>), in Igor Pro.'

-Line 147: when authors write “all in situ aerosol observations”, they mean the previous optical properties described, but in the next paragraph pollen concentration (which are also a type of in situ aerosol observations) is described. Please specify if you refer only to the previously mentioned ones.

Thank you for your comment. We have specified the in situ instruments which this applies in the manuscript.

-Line 150: what is the time resolution for pollen concentration measurements?

It is 2h. We have added this information in the manuscript.

-Line 183: it would be helpful to briefly define the “potential emission sensitivity” and their units $[s^{-1}]$ according to Figures 8 and 11, even if you cited the original paper describing this model.

We have added the following text: 'FLEXPART was run in backward simulation mode, with potential emission sensitivity (PES, s^{-1}), describing the sensitivity of the sampled air mass to surface emissions along its transport history, output at 1 h temporal resolution.'

-Authors refer to Vakkari et al. (2021) as the methodology used for deriving particle backscatter coefficient and linear depolarization ratio from Doppler lidar measurements. In that paper, there is also a description of the uncertainty estimation for the depolarization ratio. In the current manuscript, that uncertainty is used e.g. in Figures 9 and 12 as color scale, so you should mention about its calculation in the methodology section. Additionally, there seems to be a filter applied in Figures 2, 6 and 10 to discard noisy values (appearing as white pixels in the plot), is this filter related with the depolarization uncertainty, with the SNR level or something else? Please include it explicitly.

Thank you for pointing this out. We have added the uncertainty calculation in the Sect 2.1. For each case study, an SNR threshold was identified from instrumental noise in aerosol- and cloud-free observations. For Kiviniemi (Fig. 2) and Welgegund (Fig. 10), a co-polar SNR threshold of 0.0005 was applied. For Kumpula (Fig. 6), a co-polar SNR threshold of 0.0006 was applied. We have added the related information in the captions.

-How do you define the boundaries of the layer of interest in each case? This should be explained in the methodology section.

The boundaries of the smoke layers were determined separately for each case. We have added the following text in Sect. 2.1 describing the approach in each one: 'To determine the upper boundary of the smoke layer in the lidar observations at Kiviniemi, we first identified the maximum of the ATB_{1565} profile within the lowest 500 m. The layer top was then defined as the first altitude above this maximum at which the δ_{1565} exceeded 0.1. If no such threshold was reached, the layer top was instead assigned to the altitude corresponding to the maximum of the ATB_{1565} profile. In a similar way, for the Welgegund case, the maximum of the δ_{1565} profile was first located between 2 and 4 km and then the base and top boundaries were determined

using the second derivative of ATB_{1565} . For the Kumpula case, ATB_{1565} observations with values lower than $0.2 \text{ Mm}^{-1} \text{ sr}^{-1}$ or higher than $2 \text{ Mm}^{-1} \text{ sr}^{-1}$ were excluded. The remaining observations were subsequently used to define the boundaries of the smoke layer. In all cases, cloud-free layers were considered only.'

-Lines 221-232: The idea of this paragraph is more or less understandable, but I think it should be re-written for better structure and clarity. For example, lines 221-222 seems unconnected with the rest, and the rest of the sentences seem connected to the idea that the higher values of linear particle depolarization ratio are due to mixing with pollen, but the main ideas should be clearly stated and then explained with the mentioned observations.

We have re-written the paragraph for clarity.

-Lines 252-265: here it is also a trend for a bit unstructured writing, so I also propose to re-write so that the main idea is highlighted and then the explanation is elaborated with the values, uncertainties and literature mentioned.

We have re-written the paragraph accordingly.

-Section 3.2: In the previous case, the explanation of higher beta related to lower depolarization was that higher fresh smoke concentration was present in the boundary layer mix, making the total depolarization ratio lower. How do you explain that relationship in this second, free-tropospheric case? In any case, I don't think there is a clear trend like this in Figure 9, but probably the higher the uncertainty, the more scattered are the values, but the general trend is a constant depolarization with respect to beta.

To be more precise in the first smoke event, for sufficiently high ATB_{1565} , δ_{1565} remains effectively constant. This behavior indicates that the signal is primarily governed by a single aerosol type, i.e. fresh smoke, with relatively uniform microphysical properties. Under these conditions, further increases in particle concentration enhance ATB_{1565} but do not affect δ_{1565} , which is governed by particle shape rather than concentration. We have included this in the manuscript.

We agree with the reviewer that, for the second smoke event, higher uncertainties are reflected in increased scatter. Despite this, the overall trend indicates an approximately constant depolarization with respect to ATB. The scatter is likely influenced by contributions from the edges of the smoke layer.

-Line 307: it would be appropriate to explicitly indicate how were backscatter coefficient and depolarization ratio uncertainties accounted in the weighted average.

Thank you for your comment. We have included the weighted average equation below and in the manuscript.

$$\frac{\sum w \delta_{1565}}{\sum w} \quad \text{where} \quad w = \left(1 - \frac{\frac{\sigma_{\delta_{1565}}}{\delta_{1565}}}{\max\left(\frac{\sigma_{\delta_{1565}}}{\delta_{1565}}\right)} \right) \frac{ATB_{1565}}{\max(ATB_{1565})} \quad (1)$$

-Line 321: "The feature between 2.5 and 3.5 km" should be more concrete, e.g., "the increase of backscatter coefficient in a particular region..." or something similar. The same when you describe the observation of the layers in the previous cases.

Corrected as suggested.

-Section 3.3: the attribution of the detected layer as ash originating from the Copahue Volcano is reasonably justified with the calculated air trajectories and the already studied information about emission and propagation of the ashes by Paez et al. (2021). However, in light of the

trajectories in Figure 11, would it be possible the presence of other highly depolarizing aerosol type?

Thank you for pointing this out. There are drylands in the air mass trajectory path (Ganem et al., (2022)). According to the animation which shows the Dust Score from 04 to 18 December 2016 using the Aqua/AIRS product available both day and night available here, there could be dust particles in the atmosphere. At the same time, monthly SO₂ column mass from MERRA2 here show elevated SO₂. The SO₂ regions overlap with the regions in which higher dust score is given. Because the Dust Score alone cannot distinguish between silicate-rich volcanic ash and desert dust due to their overlapping spectral properties, a high Dust Score paired with co-located sulfur dioxide, most probably indicates volcanic ash. Given the very high PDR value which is remarkably higher from dessert dust aerosols as summarized in Table 2, we conclude that aerosol layers and therefore, the PDR value reported is rather due to volcanic ash than dessert dust particles.

Ganem, K.A.; Xue, Y.; Rodrigues, A.d.A.; Franca-Rocha, W.; Oliveira, M.T.d.; Carvalho, N.S.d.; Cayo, E.Y.T.; Rosa, M.R.; Dutra, A.C.; Shimabukuro, Y.E. Mapping South America's Drylands through Remote Sensing—A Review of the Methodological Trends and Current Challenges. *Remote Sens.* **2022**, *14*, 736. <https://doi.org/10.3390/rs14030736> .

-Table 1: due to the useful intention of including this table as a review of different observations of linear particle depolarization ratio at 1565 nm, you could also include the pollen values in Vakkari et al. (2021) or other types in Le et al., (2024).

We have added the suggested aerosol mixtures from the aforementioned studies.

Other corrections or typos:

-Line 49: “depended” should be “dependent”

Corrected as suggested.

-Line 105: the first “alpha” should also have the lambda dependence indicated in brackets.

Added as suggested.

-Lines 111, 243, 248: please check notation for the cv numeric value (it should be 10⁻¹² and not e⁻¹²)

Corrected as suggested.

-Line 208: “realized” should be “was carried out” or “was conducted” (or similar), and maybe the word “events” could be better replaced with “stages” or “phases”.

Corrected as suggested.

-Figures (2, 6, 10, A1, A2) including backscatter coefficient present logarithmic color scale, if I am not wrong. Please indicate it if it is like that.

The reviewer is correct.

-Line 214 and others: “seen by the lidar” may seem a bit colloquial for this context, please consider using more technical or accurate verbs as “recorded”, “measured” or “detected”.

Corrected as suggested.

-Line 217: “black filled” should be “red filled”

Corrected as suggested.