This study is ambitious. The authors extend a well-established technique for separating the fine and coarse modes of dust particles, based on AERONET data and lidar polarization measurements. In this manuscript, the authors adapt this approach for lidar measurements from space. Estimating dust particle mass concentration from an elastic backscatter lidar involves numerous assumptions about region-dependent lidar ratios, conversion factors, etc., which the authors acknowledge and discuss thoroughly. It's worth noting that the authors are well-known experts in lidar measurements. The manuscript is well-written, includes detailed introduction, long reference list, and is definitely suitable for publication in AMT. I have just several technical comments.

The authors would like to thank the reviewer for his time, comments and suggestions. We did our best to incorporate the proposed changes and corrections in the revised manuscript, aiming at improving the presented paper. Following, you will find our responses, one by one to the comments addressed.

Kind regards,

Emmanouil Proestakis

 p.10 ln.19 "urban haze and biomass-burning smoke, with depolarizing effects of 1-4% at 532 nm". Actually, it can be much higher. Depolarization of smoke and urban particles can be up to 10%. (Veselovskii, I., Hu, Q., Goloub, P., Podvin, T., Barchunov, B., and Korenskii, M.: Combining Mie– Raman and fluorescence observations: a step forward in aerosol classification with lidar technology, Atmos. Meas. Tech., 15, 4881–4900, 2022. https://doi.org/10.5194/amt-15-4881-2022) and references therein. In upper troposphere depolarization of smoke at 532 nm can reach 15%. All this introduces uncertainties in separation of dust and smoke. Depolarization of pollen at 532 nm can reach 35%.

The authors agree with the reviewer, the section of the depolarization ratio of the aerosol subtype classes considered in the study could be enhanced by including the research outcomes reported by Veselovskii et al. (2022) and Bohlmann et al. (2021). The paragraph was re-written in order to provide more information. Below we provide the part of the section which was vastly rephrased and extended.

From:

"However, accurate implementation of the pure-dust decoupling methodology (Eq. (5)) requires, in addition to dust depolarization features (" δ_d "), proper consideration of the depolarization features of the non-dust aerosol subtypes composing the aerosol mixture (" δ_{nd} "). Broader aerosol subtype categories include sea salt, biomass-burning smoke, pollen, and volcanic ash. Regarding marine aerosol, the particle linear depolarization ratio increases from 2-3% at 532 nm for wet spherical sea salt particles in marine environment of high relative humidity to about 10 to 15% at 532 nm for dry cubic-like sea salt particles close to the Marine Boundary Layer (MBL) - Free Troposphere (FT) entrainment zone (Haaring et al., 2017). The presence of sea salt in the FT is considered negligible. Other aerosol subtypes frequently encountered both in the Planetary Boundary Layer (PBL) and the FT include urban haze and biomass-burning smoke, with depolarizing effects of 1-4% at 532 nm (Müller et al., 2007b; Nicolae et al., 2013). The pollen aerosol category relates to depolarization ratio in the range of 4–6% at 532 nm, although in extreme cases of significantly large particles (diameter \geq 50 µm) this effect may reach as high as 15% at 532 nm (Noh et al.; 2013). The presence of pollen is usually confined within the PBL and manifests high seasonality, with higher values evident during spring and during atmospheric convection conditions. Finally, a less frequently observed aerosol category is volcanic ash, with depolarization ratio effect ranging between 30 and 40% at 532 nm, as reported by EARLINET observational activities in the case of Eyjafjallajökull in 2010 (Ansmann et al., 2010; Groß et al., 2012). Here, and based on the above discussion, for the non-dust aerosol

subtypes category " δ_{nd} " equal to 0.05 ± 0.02 at 532 nm is assumed (Tesche et al. 2009, Mamouri and Ansmann, 2014; 2016; Marinou et al., 2017; Proestakis et al., 2018)."

To:

"However, accurate implementation of the pure-dust decoupling methodology (Eq. (5)) requires, in addition to dust depolarization features (" δ_d "), proper consideration of the depolarization features of the non-dust aerosol subtypes composing the aerosol mixture (" δ_{nd} "). Broader aerosol subtype categories include sea salt, biomass-burning smoke, pollen, and volcanic ash. Regarding marine aerosol, the particle linear depolarization ratio increases from 2-3% at 532 nm for wet spherical sea salt particles in marine environment of high relative humidity to about 10 to 15% at 532 nm for dry cubic-like sea salt particles close to the Marine Boundary Layer (MBL) - Free Troposphere (FT) entrainment zone (Haaring et al., 2017). The presence of sea salt in the FT is considered negligible. Other aerosol subtypes frequently encountered both in the Planetary Boundary Layer (PBL) and the FT include urban haze and biomass-burning smoke, with depolarizing effects of 1-4% at 532 nm (Müller et al., 2007b; Nicolae et al., 2013). However, it should be noted that recent outcomes provided by Veselovskii et al. (2022), based on a combination of fluorescence and Mie-Raman lidar observations, further reveal the high variability of the "Pollen", "Urban", and "Smoke" aerosol subtype classes. More specifically, the authors report depolarization ratio between 2% and 10% for smoke and between 1% and 10% for urban aerosol subtypes. The pollen aerosol category relates to depolarization ratio in the range of 4–6% at 532 nm, although in extreme cases of significantly large particles (diameter \geq 50 µm) this effect may reach significant higher values (at 532nm ~15% - Noh et al.; 2013, ~30% at 532 nm - Veselovskii et al., 2022, ~38% - Bohlmann et al., 2021). The presence of pollen is usually confined within the PBL and manifests high seasonality, with higher values evident during spring and during atmospheric convection conditions. However, these values correspond to the upper and lower limits of the aerosol layer observations. Finally, a less frequently observed aerosol category is volcanic ash, with depolarization ratio effect ranging between 30 and 40% at 532 nm, as reported by EARLINET observational activities in the case of Eyjafjallajökull in 2010 (Ansmann et al., 2010; Groß et al., 2012). Here, and based on the above discussion, for the nondust aerosol subtypes category " δ_{nd} " equal to 0.05 ± 0.02 at 532 nm is assumed (Tesche et al. 2009, Mamouri and Ansmann, 2014; 2016; Marinou et al., 2017; Proestakis et al., 2018)."

 p.10 ln 22. "(Noh et al.; 2013)". More recent references about pollen depolarization should be added. Bohlmann, S., Shang, X., Giannakaki, E., Filioglou, M., Saarto, A., Romakkaniemi, S. and Komppula, M.: Detection and characterization of birch pollen in the atmosphere using multi-wavelength Raman lidar in Finland, Atmos. Chem. Phys. 19, 14559–14569, 2019. doi.org/10.5194/acp-19-14559-2019. Atmos. Chem. Phys., 21, 7083–7097, 2021 <u>https://doi.org/10.5194/acp-21-7083-2021</u>.

We agree with the reviewer that the manuscript would improve by including the recommended reference. Added in the manuscript.

3. p.11 ln.1. "In Eq. (6), "β_{λ,ncd}(z)" and "β_{λ,cd}(z)" correspond to the non-coarse-mode aerosol (i.e., non-dust and fine-mode dust)" I am confused. Because in p.11 ln.22 authors write "we assume mean linear depolarization effects of "δncd " and "δcd " equal to 0.16 ± 0.02 and 0.39 ± 0.03" Does it mean that smoke (it is non-dust) has depolarization 16%? These definitions should be clarified.

The two-step POLIPHON technique (Mamouri and Ansmann, 2014; 2017) here is applied to the CALIPSO dusty aerosol subtypes (Winker et al., 2010), hence to the "dust", "dusty-marine", and "polluted dust" (Omar et al., 2009; Kim et al., 2018). Hence, since the aerosol subtypes of interest are the dusty mixtures, it is assumed always to have a dust component and a non-dust aerosol component in the aerosol layer, present as external mixtures (Tesche et al., 2009; Ansmann et al., 2019). Here, as complement aerosol subtypes to the dust aerosol subtype (total, fine-mode, and coarse-mode dust) in the CALIPSO-based dusty mixtures are considered (1) the marine aerosol class in the "dusty-marine"

aerosol subtype mixture and (2) the smoke or pollen or volcanic ash or urban or continental aerosol classes in the "polluted-dust" aerosol subtype mixture (Amiridis et al., 2013; Marinou et al., 2017; Proestakis et al., 2018). More specifically, the two-step POLIPHON technique assumes that the backscattered signal by an external aerosol mixture " $\beta_{\lambda,p}(z)$ " corresponds to the summation of the cross and parallel return signals from the (1) non-dust, (2) fine-mode dust, and (3) the coarse-mode dust aerosol components, as discussed above (Mamouri and Ansmann, 2014; 2017). Thus, with respect to the implementation of the first-step of the two-steps of the POLIPHON technique, towards determination of the atmospheric coarse-mode dust aerosol component, knowledge of the non-coarse-mode dust aerosol and coarse-mode dust light-depolarization characteristics, thus of " δ_{ncd} " and " δ_{cd} " respectively, is required. The " δ_{ncd} " corresponds to the particulate depolarization ratio of the non-coarse-mode dust component, thus for the aforementioned terms (#1) and (#2). The " δ_{cd} " term corresponds to the particulate depolarization ratio of the coarse-mode dust aerosol component, thus of the aforementioned term (#3).

Since " δ_{ncd} " includes broader aerosol non-dust aerosol subtype classes and since the CALIPSO aerosol subtype classification algorithm does not provide information on the complement-to-dust aerosol subtype class in the aerosol mixture, in order to apply the two-step POLIPHON technique a representative universal mean particulate depolarization ratio at 532 nm for the non-dust aerosol subtype and for the fine-mode dust particulate depolarization ratio at 532 nm have to be included. Following an increasing number of observations reporting on the particulate depolarization ratio at 532 nm of different aerosol subtype classes (i.e., DeLiAn database; Floutsi et al., 2023 and references therein) and laboratory experiments reporting on the particulate depolarization ratio at 532 nm of the fine- coarse- mode of dust (Sakai et al., 2010; Järvinen et al., 2016) assumptions are made on " δ_{ncd} " and " δ_{cd} ". According to the published outcomes of the studies, the term " δ_{ncd} ", the total effect on depolarization of the fine-mode of dust and the non-dust aerosol component (i.e., marine or smoke or pollen or volcanic ash or urban or continental) in the external aerosol mixture is assumed of ~0.16 ± 0.02 in the two-step POLIPHON technique applied to optical products of CALIPSO (Sect. 2.2.).

4. Fig.2. Depolarization 1.0 is confusing. Reader may have felt that dust depolarizations extend up to 1.0.

The authors have followed the official CALIPSO conversions in terms of colorbars, colormaps, and scale of CALIOP optical products. An indicative example is provided here-in-after, for the Godzilla dust transport event over the North Atlantic Ocean (June 2020), as provided by CALIPSO lidar browse images. In terms of depolarization ratio at 532 nm the official and suggested scale extends from lower than 0.0 to higher values than 1.0. This is the reason behind the lower limit of 0.0 and the upper limit of 1.0 in Fig.2.





Figure 01: CALIPSO nighttime granule (a), Total Attenuated Backscatter at 532 nm (b), and Depolarization Ratio at 532 nm (c) for the Godzilla dust transport event on the 18th of June 2020.

5. Table 4. The reference should be added: Atmos. Meas. Tech., 16, 1951–1970, 2023 https://doi.org/10.5194/amt-16-1951-2023.

We agree with the reviewer that the manuscript and the discussion would improve by including the recommended reference. Reference is added in the manuscript.

6. Fig.5b. Labels on right axes is difficult to read.

According to the reviewer's comment, the figure is changed as follows:





Figure 5: Major Saharan dust outbreak moving westwards over Dakar on the 20th of April, 2017 at ~14:43 UTC, including the CALIPSO overpass in the proximity of the AERONET-Dakar station (red line) and FLEXPART 6-day back-trajectories at 2 km at the area of interest (Lat: 14.39, Lon: -16.95) denoting the Saharan desert origin of the advected air masses (yellow line) (Fig.5a). CALIPSO L2 5 km backscatter coefficient at 532 nm cross section (Fig.5b). Column-integrated particle volume concentration as a function of particle radius observed with the AERONET sunphotometer over Dakar, Senegal (Fig.5c). Backscatter coefficient at 532 nm profiles of the coarse-mode pure-dust (red shaded area), fine-mode pure-dust (blue shaded area), and non-dust (gray shaded area) components of the total aerosol load (Fig.5d). Particulate depolarization ratio at 532 nm profile used for the decoupling of the coarse-mode pure-dust, fine-mode pure-dust, and non-dust components of the total aerosol load, as provided in Fig.5d (Fig.5e). Extinction coefficient at 532 nm profiles of the coarse-mode pure-dust (red shaded area) and fine-mode pure-dust (blue shaded area) components of the total aerosol load, as