Response to RC3

General Comments:

The paper "POLIPHON conversion factors for retrieving dust-related cloud condensation nuclei and ice-nucleating particle concentration profiles at oceanic sites" presents and discusses the dust-related conversion factors as extracted over remote oceanic/coast sites using the AERONET database around the globe. These different conversion parameters are of critical importance for the POLIPHON methodology in order to compute dust-related CCNC and INPC globally. The study falls within the scope of AMT. The authors have done a thorough job, the manuscript is well-written / structured, the presentation clear, the language fluent and the quality of the figures high. Furthermore, the authors give credit to related work and the results support the conclusions. However, in order to help improving the manuscript, I would kindly suggest the authors to take into account the following minor comments.

Response: We appreciate the reviewer's thoughtful review and constructive comments. All the comments have been addressed in the revised manuscript, and the responses to each comment are given below.

Specific comments:

Comments: Central component of the analysis provided is the AERONET-based depolarization ratio, which is established according to the model of randomly oriented spheroids. Thus, I would suggest to the authors to dive into significantly more discussion and details on the major building component of their approach, including methodology, approach, assumptions, and accuracy.

Response: Thank you for the reviewer's reminder. The spheroid shape model may indeed induce errors in particle linear depolarization ratio for mineral dust according to the results of the modeling study from Gasteiger et al. (2011). Even though, dust is the predominant particle to trigger significant depolarized signal, which can be well captured by AERONET spheroid model. We take a large threshold of particle depolarization ratio (0.30) to mitigate the error introduced by the spheroid model, in terms of the wrong characterization of dust. We have added some sentences to mention the spheroid model of AERONET retrieval and the benefit of using irregular particle shape for mineral dust in the modeling study by Gasteiger et al. (2011) in the introduction of the revised manuscript as follows

'…It should be noted that the particle linear depolarization ratio values in AERONET retrieval are calculated from the combination of the particle size distribution and complex refractive index based on a spheroid light scattering model (Dubovik et al., 2006). Based on a modeling study, Gasteiger et al. (2011) found that the lidar-measured particle linear depolarization ratio values for pure mineral dust can be well reproduced by using an irregular particle shape

compared with using the spheroid shape assumption. Nevertheless, we consider it adequate to adopt AERONET-derived particle linear depolarization ratio values to qualitatively identify the presence of dust in the atmospheric column (Noh et al., 2017).' (Please see Line 99-105)

Besides, in the methodology part (the third paragraph of section 2.2), we have already mentioned this issue. (Please see Line 211-218)

Comments: Since a significant number of PollyXT lidars operate at same time AERONET stations, my suggestion would be the extended intercomparison and evaluation of the AERONET-based depolarization ratio against the Polly lidar depolarization ratio, under events of dust, polluted dust, and non-dust, in order to strengthen the argument of the suitability of the AERONET-based depolarization ratio to extract CCNC and INPC conversion factors. This comparison over land should be a first stepping stone before attempting over ocean, where lidar systems are less frequently operated, and eventually before the claim of supporting 3D CCNC and INPC dust-related studies globally.

Response: Thank you for the suggestion. It would be better to fully confirm the validity of AERONET-derived PLDR with those measured by ground-based polarization lidar. As we mentioned in the manuscript, the related comparisons have been made and the results have been discussed in many previously published papers, such as Toledano et al. (2019), Shin et al. (2017), and Müller et al. (2010, 2012), especially for the comparisons in SALTRACE (Saharan Aerosol Long-range Transport and Aerosol–Cloud-Interaction Experiment) campaign at Barbados (Haarig et al., 2022), where is a great location to compare the PLDR values from AERONET and lidar measurements in transatlantic dust cases.

Comments: The authors should go into more details on the dependencies between the AERONET-based depolarization ratio to extract CCNC and INPC conversion factors around the globe and the discrepancies in dust microphysical properties of dust around the globe, for the main objective is to apply the conversion factors eventually in lidar observations through POLIPHON, possible at regions and conditions of dust transport significantly different than the observed at the specific stations of the present study. Moreover, the authors should discuss, possible through study cases, the change of the extracted and proposed CCNC and INPC conversion factors as a function of aeolian transport and distance, for aging and mixing with non-dust aerosol subtypes, even under the hypothesis of external mixing, alters the columnar observations of AERONET, thus affects the total conversion factors.

Response: We are appreciated for the reviewer's valuable suggestion. As mentioned in this manuscript, there will be a following-up work that focuses on the dust-related conversion factors at those polluted city sites with more complicated local aerosol emission conditions and then the global conversion factor dataset can be expected. AERONET sites are serried in some regions (especially the regions with large populations) and can be very sparse in those outlying regions. Thus, the retrieved conversion factors absolutely will be distributed unevenly. When selecting the AERONET sites for calculating the conversion factors, we try our best to ensure that there are at least regional-representative conversion factors available for most of the geographical locations around the world. A dust-related conversion factor at an isolated site can be applied to a large area around it. Also, geographical interpolation is another possible way to obtain the final global grid dataset of dust-related conversion factors. Nevertheless, the final processing for retrieving the dust-related global conversion factors will be determined only after finishing the following-up work (i.e., conversion factors at polluted city sites), which would be better discussed in detail in our next paper. Therefore, the final global dataset of dust-related conversion factors can reflect the regional characteristics of dust microphysical properties, such as for dust sources, places along dust transport pathways, downstream regions after long-range transport, or regions favoring dust aging and mixing with non-dust aerosols.

However, as the first step, this manuscript focuses on discussing the possibility of a dust case selection scheme employing the AERONET-derived PLDR and attempting the application to the retrieval of dust-related conversion factors at the clean oceanic sites (with simple background aerosol conditions). Besides, another attempt for retrieving the dust-related conversion factor (probably mixing dust situations with other aerosol types) at a polluted city site has been demonstrated by He et al. (2021). To concentrate on the main subject, we would like to mention the future work in the outlook part as seen in the last paragraph of the revised manuscript as below **'Once those conversion factors at polluted city sites are retrieved, a global dust-related conversion factor grid dataset will possibly be obtained by geographical interpolation.'** (Please see Line 397-398)

Comments: The aforementioned approach should be as robust as possible, for once the conversion factors are extracted and established for the dust CCNC and INPC over a region, the product output should consist a fingerprint of the dust related sources affecting the region as well, interconnecting the dust plumes over the oceanic sites with the dust sources. Towards this, I would suggest the authors to perform a cluster analysis of the dust sources affecting each oceanic site (i.e., backtrajectories).

Response: Thank you for the reviewer's suggestion. As mentioned in our response to the next comment (see below), we agree with the reviewer that different oceanic/coast sites in this study may be influenced by long-range transported dust aerosols from different dust sources over the world. However, the purpose of this study is to obtain the multi-year average characteristic of dust aerosols and associated dust-related conversion factors for the selected oceanic/coast AERONET sites. We do not intend to separate the respective contribution of different dust sources to a given site because it would be much more complicated to analyze the dust sources for different sites and regions, which has already been studied specifically in the existing literature (Bullard et al., 2016; Struve et al., 2020; Kok et al., 2021; Meinander et al., 2022).

In addition, it is believed that robustness can be guaranteed adequately. First, the data durations are long enough for the selected AERONET sites, which are at least 7 years (St_Helena) and can be up to 28 years (Mauna_Loa and Cape_Verde). Second, the linear Pearson correlation coefficients are generally >0.70 (most of them are >0.90), suggesting the INP-relevant properties for each site are well reflected. Third, the intercomparisons with the conversion factors in Ansmann et al. (2019) using a different dust identification scheme are conducted (in section 3.1). Last, the background atmospheric environment at oceanic sites is always clean, indicating that the identified dust cases are less influenced by other aerosol sources; this issue must be handled with more care when retrieving the dust-related conversion factors at other terrestrial sites in the future.

Comments: Please discuss the effect on the extracted AERONET-based depolarization ratios and accordingly on the CCNC and INPC conversion factors of different dust regions – with different dust properties (i.e. LR), affecting the same marine site.

Response: It would be difficult to comprehensively and quantificationally discuss this issue. Excluding some occasional extreme events (Uno et al., 2009), a given oceanic region is generally impacted by specific dust sources via typical dust transport pathways. In the middle- and low-latitude Atlantic, the primary dust transport pathway is from the Saharan desert in North Africa to the east coast regions of North America (Rittmeister et al, 2017; Yu et al., 2021). In the North Atlantic, it is reported that dust aerosols are mainly from Iceland (Baddock et al., 2017). Dust aerosols in the Arctic mainly come from the high-latitude dust sources in the North Hemisphere (e.g., Alaska, Canada, Denmark, Greenland, Iceland, Svalbard, Sweden, and Russia) (Bullard et al., 2016; Meinander et al., 2022), Arctic local sources (Shi et al., 2022), Asia (Zhao et al., 2022), and North Africa (Shi et al., 2022). As for the dust aerosols over the Pacific, they mainly originate from the Central and East Asia dust sources to North America (Guo et al., 2017; Hu et al., 2019). As for the remaining few oceanic sites in the South Hemisphere, dust aerosols can be related to Australia, New Zealand, Patagonia, and Southern Africa (Bullard et al., 2016; Struve et al., 2020; Kok et al., 2021; Meinander et al., 2022). Thus, as seen in Figure 6, the region-to-regions variations of conversion factors (i.e., $c_{\mathrm{v,d}}$, $c_{\mathrm{s,d}}$, and $c_{\mathrm{s,100,d}}$) can be attributed to the diverse contributions from different dust sources. In addition, as the downstream areas, the possible aging and mixing of dust with other aerosol types during long-range transport may also be responsible for the region-to-region variations of conversion factors (Kim and Park, 2012; Goel et al., 2020).

Therefore, we have added a new paragraph in section 3.2 to address the reviewer's concern. (Please see Line 313-327)

Comments: Table 2 provides the available number of data points for total, dust-dominated mixture, and pure dust, in AERONET inversion products. In specific cases the dataset is characterized by a very low number of cases. The authors should discuss on the fail-safes considered in order to guarantee the robustness of the conversion factors extracted, even in the low number of cases AERONET stations. Moreover, please provide at the table for each of the site (Table 2), with the basis AERONET products, for the Total Obs., DDM Obs., and PD Obs. (i.e., AOD+AOD_SD, AE+AE_SD, …). How does the low number of cases affect the uncertainties and confidence of the conversion factors? Please discuss providing additional input where necessary.

Response: In Ansmann et al. (2019b), the lowermost number of available data points for calculating the conversion factors is 17 at the Tuscon AERONET site. In this study, we provide INP-relevant ($c_{\text{v,d}}$, $c_{\text{250,d}}$, $c_{\text{s,d}}$, and $c_{\text{s,100,d}}$) conversion factors (see tables 3 and 4) with the number of available data points no less than 12 to ensure as many sites as possible can provide dust-related conversion factors with somewhat acceptable reliability. With data point number below 12, the data points may be diverging caused by occasional dust cases, causing very small linear Pearson correlation coefficients R. Here we provide R values for each dust-related INP-relevant conversion factor at each AERONET employed in this study as seen in the following table. It can be seen clearly that most of the conversion factors have corresponding R values exceeding 0.70, except for PD-derived $c_{\text{250,d}}$values at NR and AS (as marked in red in the table), which can guarantee the robustness of the retrieved conversion factors. Therefore, we have added some sentences to discuss this issue (in the second paragraph of section 3.2) as follows

‘We consider the conversion factors with the number of available PD data points $\geq$12 valid (provided in Table 4). Moreover, to guarantee robustness, only the retrieved conversion factors with the linear Pearson correlation coefficient *R* exceeding 0.70 are considered valid, except for PD-derived $c_{\text{250,d}}$ values at NR (R=0.32) and AS (R=0.50) which should especially be handled with care in scientific application.’ (Please see Line 289-292)

	Site	R for $C_{v,d}$		R for $C_{250,d}$		R for $C_{s,d}$		R for $C_{s,100,d}$	
		DDM+PD	PD	DDM+PD	PD	DDM+PD	PD	DDM+PD	PD
North Africa	CV	0.97	0.97	0.94	0.94	0.76	0.78	0.98	0.99
	DK	0.96	0.96	0.94	0.94	0.71	0.74	0.94	0.97
	IZ	0.98	0.98	0.92	0.92	0.77	0.77	0.99	0.99
Middle East	EI	0.95	0.97	0.91	0.92	0.56	0.82	0.56	0.99
	SV	0.97	0.98	0.96	0.96	0.76	0.78	0.91	0.98
	ME	0.96	0.98	0.93	0.96	0.76	0.82	0.79	0.97
Asia	DU	0.98	0.98	0.95	0.96	0.84	0.83	0.94	0.98

	DA	0.90	0.77	0.76	0.83	0.41	0.93	0.67	0.70
	LA	0.96	0.97	0.86	0.93	0.76	0.73	0.95	0.99
Pacific	TA	0.88	-	0.89	-	0.87	-	0.94	-
	NR	0.93	0.80	0.90	0.32	0.91	0.79	0.96	0.76
	MI	0.92	0.91	0.96	0.97	0.95	0.97	0.97	0.98
	AS	0.89	0.91	0.84	0.50	0.86	0.75	0.92	0.81
	GA	0.80	-	0.86	-	0.89	-	0.93	-
	ML	0.88	0.92	0.92	0.95	0.90	0.95	0.95	0.99
Pacific Coast	HU	0.84	1.00	0.84	-	0.87	-	0.93	-
	OS	0.86	0.90	0.75	0.98	0.66	0.79	0.76	0.87
	SH	0.93	0.99	0.94	0.97	0.89	0.88	0.90	0.98
	SI	0.92	-	0.94	-	0.89	-	0.96	-
	TR	0.88	0.98	0.93	0.98	0.93	0.97	0.95	0.98
Atlantic	AG	0.98	0.99	0.98	0.98	0.95	0.97	0.99	0.98
	TH	0.93	0.98	0.92	0.97	0.92	0.97	0.96	0.99
	ST	0.91	-	0.91	-	0.95	-	0.98	-
Indian Ocean	MG	0.82	-	0.83	-	0.92	-	0.95	-
	AI	0.93	0.82	0.89	0.82	0.93	0.85	0.98	0.92
Arctic Ocean	NA	0.92	-	0.93	-	0.76	-	0.88	-
	TL	0.95	-	0.91	-	0.91	-	0.95	-
	OP	0.76	-	0.94	-	0.87	-	0.91	-
	IQ	0.74		0.72	-	0.84	-	0.88	-

Moreover, we have added the AOD (at 532 nm) and AE (between 440 nm and 870 nm) in the updated Table 2 as suggested by the reviewer. Note that table 2 is too crowded to give the corresponding standard deviations for AOD_{532} and $AE_{440-870}$.

Comments: In table 1 the authors provide the uncertainties of the approach. The uncertainties have been established on the basis of long-term ground-based observations (i.e., EARLINET, PollyNET). Since the objective of the study, as mentioned in the very beginning of the manuscript, is to "to characterize the 3-D distribution of dust-related Cloud Condensation Nuclei Concentration (CCNC) and Ice Nucleating Particle Concentration (INPC) globally", which can be achieved only based on satellite-lidar systems (i.e., CALIOP, CATS, ATLID), where the uncertainties of backscatter and particulate depolarization ratio are of the same order of magnitude as the backscatter and particulate depolarization ratio. In this case, as the higher uncertainties are used as input in the error propagation, the final uncertainties will be significantly higher in satellite-based lidar products than when ground based products are extracted. Please discuss.

Response: Thank you for pointing out this issue. We agree with the reviewer's point of view that the uncertainties in the retrieved aerosol extinction/backscatter coefficient from spaceborne lidar measurement may differ from those for ground-based lidar measurement. Hence, we have used global CALIOP level-2 aerosol profile data during the night on 1 January 2010 as an example, to examine the typical uncertainties in aerosol extinction and backscatter coefficient as well as particle linear depolarization ratio (only select data points with PDR≥0.05) at 532 nm (see the figures below).

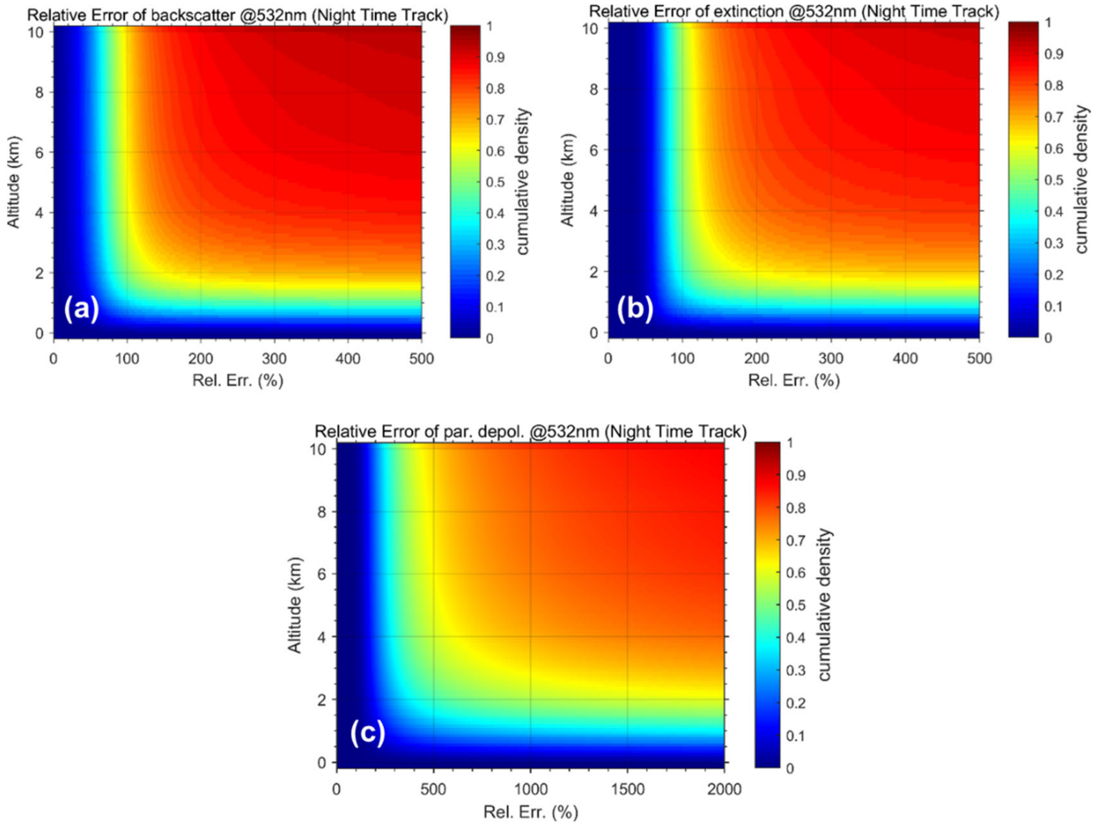

Figure 1R. The Cumulative density of relative error/uncertainty of different CALIOP level-2 aerosol profile products: (a) total backscatter coefficient at 532 nm; (b) extinction coefficient at 532 nm; (c) particle linear depolarization ratio at 532 nm. Both of these results were calculated based on CALIPSO night-time orbits on 1 January 2010.

CALIOP level-2 aerosol profile data with the 5-km horizontal resolution are used here, corresponding to 15 laser pulses. In this situation, the uncertainties in the aerosol extinction coefficient, aerosol backscatter coefficient, and are approximately <180% and <120%. As for the particle linear depolarization ratio, the uncertainty is larger; here, we considered it as <300%. Typically, we merge the raw data of ground-based lidar to obtain a time resolution of one minute (this time can be even larger as 15 min or 30 min are always used in INP retrieval with ground lidar observations), corresponding to 1200 laser pulses (if using a laser with a pulse repetition frequency of 20 Hz). Thus, from 15 laser pulses to 1200 laser pulses, the uncertainty will be declined by a factor of ~9. As a result, we would like to estimate the uncertainty in aerosol extinction coefficient, aerosol backscatter coefficient, and particle linear depolarization ratio to be approximately <20%, <13%, and <33%, respectively. Considering the uncertainty in the dust lidar ratio (30-60 sr) of 33% and the assumed non-dust depolarization ratio (0.05) of 30% (Burton et al., 2013), the uncertainties in the dust backscatter coefficient and dust extinction coefficient should be approximately <49% and <59%. Thus, we consider that the uncertainties in M_{d}, $n_{250,\mathrm{d}}$, s_{d}, and $s_{100,\mathrm{d}}$ are estimated to be approximately <60%. Similar to the original table 1, the final uncertainties in INPC and CCNC are still estimated to be <500% and <200%. In addition, it should also be noted that the uncertainty level of CALIOP-derived optical parameters can be further improved by integrating the data to decrease the spatio-temporal resolution. However, the largest uncertainty is contributed by the parameterization schemes for CCN and INP currently; hence, the improvement of lidar-derived optical parameters makes no sense for the moment at least.

Thus, we have updated table 1 based on the uncertainties of optical properties for space-borne lidar measurement accordingly.

<u>Reference:</u>

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