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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Vertical distribution of aerosol optical properties in the Po Valley during the 2012 summer campaigns" *by* Silvia Bucci et al.

Silvia Bucci et al.

s.bucci@isac.cnr.it

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We would like to thanks the reviewers for the useful comments and insights that allowed us to better redefine the methods and presenting the results of this study.
First reviewer:

General comments

" *[. . .]in the analysis of the effect of RH on the aerosol linear depolarization ratio (δ_a), a correlation analysis between the RH at surface level and the values of δ_a at the lowest altitudes would offer more information about this process"*

Following the suggestions of the reviewer, we adopted a more quantitative approach to describe the near ground hygroscopic growth events. A correlation analysis between the ground RH and the δ_a along the time would not be meaningful, as the variability of aerosol depolarization near the ground is not affected solely by RH. We decided instead to better show, along the whole campaign period, the effect of different values of RH along the vertical profile of δ_a for dust and dust-free days. Details can be found in the answer to point "10. Issues regarding section 7"

" *According to the authors, "The main objective of this paper is to investigate the transport of desert dust in the middle troposphere and its intrusion into the planetary boundary layer (page 1, line 4)". In order to fulfil this objective it would be desirable a more detailed analysis of the events where the aerosol concentration increases at surface level and how the dust layer interacts with the PBL. However, the whole 21-day period is presented in a single figure (figure 2, lidar; figure 4, in situ). "*

To facilitate the individuation of dust intrusion in the PBL and its penetration down to the ground, a closer view is added (see Fig 5 of the revised manuscript). Here we show LiDAR aerosol particle types profiles, and the corresponding APSS volume size distribution of aerosol particles at the ground, during the dust events. The interaction of dust with the PBL and the direct effect of the dust layer intrusion on the particles concentration at the ground are more easily identifiable.

" *A great part of this work is based on measurements of a lidar system. However, the lidar signals have not been properly processed. The calculations regarding the lidar measurements must be revised (see specific comments). "*

Reviewer comments highlighted the need to revise the section on the adopted LiDAR signal processing. We rewrote this section to remove any ambiguities in the parameters definition. See also answers to points 2, 4 and 5.

" *Finally, the authors must carry on a thorough revision of the language.* "

We revised the manuscript to improve the language.

Specific comments

" *1. Page 4, line 20: "In the following discussion, we will make use of backscattering ratio (R)". Why do the authors use R, which depends on the molecular terms, rather than β_a, which only depend on aerosols?* "

The use of the total backscattering ratio R, instead of the aerosol backscatter β_a, has the advantage to be bereft of the noise that is introduced by the signal inversion for β_a estimation. This helps to have a refined aerosol classification. In the paper we therefore based the aerosol classification on the parameter 1-1/R, varying from 0 to 1 accordingly to the presence of an aerosol layer, instead of relying directly on R values. To emphasize the meaning of such parameter we changed its expression as a function of the aerosol backscattering ratio (Ra=R-1= β_a(r)/ β_m(r)) instead of R. Classification is now a function of the values of Ra/(Ra+1), still varying from 0, when β_a(r)=0, to 1, when in presence of dense aerosol layer (high β_a(r) values).

" *2. [. . .] I could not get to understand how the authors use δ_a to determine the lidar ratio prior to the signal inversion. However, in page 4, line 29 the authors claim: ". . .desert dust (identified by δ_a (r) > 10%) is characterized by L equal to 50 sr (Müller et al., 2007). . .". This is the same criterion used by Rosety [2016], although Rosety [2016] used δ instead of δ_a. Because of this, the most likely explanation is that the authors have mixed up both parameters (δ and δ_a). Another example of this is found in page 6, line 29 [. . .] Nevertheless, the authors barely find aerosols with δ_a > 20 % (e.g., Fig. 1). This is another indication that the authors are actually using δ instead of δ_a. Page 7, line 3: [. . .] Again, I think that these differences are because*

the authors are comparting two different parameters (δ and δ_a). The authors should revise their calculations to see if this is the source of disagreement. If, after this, the calibration stills play an important role in this disagreement, the authors should describe its effect in a more detailed way. In the corrected version the authors must indicate, unambiguously, which depolarization parameter are using. The volume depolarization ratio, δ, can still be used as a rough indicator of the presence of dust in the determination of the lidar ratio values. However, for aerosol classification in section 3 I strongly recommend using the aerosol linear depolarization ratio (δ_a), since it is an intrinsic property of aerosols and does not depend on the molecular terms. "

The reviewer correctly stresses a lack of clarity in the text, where in fact the parameters seem to be mixed. It is true that the aerosol depolarization can only be ascertained after the signal has been processed and then inverted in term of R, so that it cannot be used during the inversion. In our case the inversion of the LiDAR signal is accomplished with the Klett method using piecewise constant extinction to backscatter ratio (LR) values, chosen accordingly to the volume depolarization (DR, in Rosati et al., 2016) and not aerosol depolarization, as would be proper to correctly discriminate among different classes of aerosol. However, when the aerosol load is significant – and these are the cases where an improper choice of LR would have more effect - the value of the volume tends to that of the aerosol depolarization, so that the use of the volume as a proxy of the aerosol in the inversion of the LiDAR signal can be accepted. We do not think it is worthwhile to dwell too much on that in the text, but we need to remove the ambiguity in the text pointed out by the reviewer. To this aim we would then rewrite it as: (from 4,29 on) ". . . following the values reported in literature: using Volume depolarization as proxy of aerosol depolarization, highly depolarizing desert dust (δ (r) $<$10) is characterised by L. . .".

On the contrary, on page 6, line 34 we really meant Aerosol depolarization.

As the reviewer suggests, we expanded the discussion on inaccuracies in depolarization in (7,3...) as follows: “The reader should notice that the lower depolarization values that we observe respect what usually found in literature (especially for the dust layers) are likely linked to the calibration process, and in particular to the difficulty in individuating completely aerosol free layers in the vertical span of the adopted LiDAR system (from ground to 7 Km). In this work, depolarization has been calibrated following the "0° calibration" or the "atmospheric calibration" procedure, i.e. making use of a low aerosol height range in the lidar signal, where only the molecular contribution could be considered. There, the volume depolarization ratio has been forced to assume the well-known value of the air molecule linear depolarization ratio (Behrendt and Nakamura, 2002). We acknowledge that this calibration is unsatisfactory to produce quantitative results, as the possible residual presence of small amounts of highly depolarizing aerosol in the assumed clean range can easily compress the range of variability of the volume depolarization, and underestimate the final depolarization products (Freudenthaler et al., 2009; Freudenthaler, 2016). However, this possible source of inaccuracy does not compromise the purpose of this work.”

“ *3. Page 6, line 16. “For simplicity, dust on emissive areas is considered to be injected uniformly below 1000 m a.g.l., therefore only trajectories crossing this height are included in the footprint-emissions coupling”. Is this a common procedure? Is there any previous work that backs this procedure? ”*

The choice of 1000m as a level for mineral dust injection is indeed a sharp assumption. We decided therefore to estimate the level of injection as the PBL top height, extracted by FLEXPART itself from the GFS input meteorological field (Stohl et al. 2010), instead of adopting a fixed height. This does not significantly change the results of the analysis (see also answer to point 8)

“ *4. Page 7, line 5: “The LiDAR classification, based on the statistical distribution of the overall observed δ_a and R values, is also applied here to overcome such limitations.” However, in Fig. 1 it can be seen that the only parameter used for aerosol classification is δ. Because of this, we could get more information with a histogram of only δ values. Also, the figure should only show relevant δ values: more than half of the current figure corresponds to δ values (over 0.25) with almost no associated case. ”*

“ *5. Page 7, line 13: [...] The authors are comparing their results to other references although they have previously said that their results might not match them due to calibration issues (again, this is likely due to a confusion between δ and δ_a). If δ is finally used for the classification (although I strongly recommend δ_a), the threshold values should be derived from a statistical analysis of its value ”*

The distribution of δ_a and 1-1/R values (now Ra/(1+Ra)) is indeed what is really driving the aerosol classification. We agree with the review that was not clearly presented in the text and in Fig.1 of the manuscript. We corrected the text (from page 7, line 1: “Here we implement a three-types aerosol discrimination scheme to characterize the vertical and temporal aerosol variability over the region along the campaign period (15 June 2012 – 5 July 2012) based on the different statistical distribution of optical properties of each class (see Fig. 1). The reader should notice that [...]. The LiDAR classification, based on the statistical distribution of the overall observed δ_a and Ra values, is in fact applied here to overcome such limitations. The robustness of the results is then further supported by comparison with Lagrangian analysis and in-situ measurements.”) and modified the axes of Fig.1. to magnify the region on which the values are mostly concentrated. It’s easier now to identify the three different distributions of optical properties and the resulting classification. As is possible to see, each class shows a different range on the Ra/(1+Ra) parameter. We restricted therefore the threshold for

classification on such intervals.
From (7,12): "The different aerosol classes can be discerned in three distinct patterns:

1. $0.1 < Ra/(Ra+1) < 0.8$ and low values of δ_a (< 3 $\%$); such low values of aerosol linear depolarization ratio are indicative of spherical particles. These particles may be composed of anthropogenic pollution and, for higher values of Ra, by droplets, and are defined as non-depolarizing.
2. $0.3 < Ra/(Ra+1) < 0.7$ and high values of δ_a (> 10 $\%$); In this class we find the highest values of aerosol linear depolarization ratio (mainly ranging from 10$\%$ to 20$\%$) and this can be indicative of the presence of mineral dust particles. This class is defined as depolarizing.
3. $0.2 < Ra/(Ra+1) < 0.6$ and intermediate δ_a values (3 $\%$< δ_a <10 $\%$) which, based solely on Ra and δ_a, cannot be considered as indicative of a dominance of a defined aerosol type unless coupled to a more thorough correlation with additional observations. We will refer to this type as intermediate depolarizing.

"

As expected, the resulting aerosol mask (see Fig.2 of the revised manuscript) appears to be not meaningfully affected by such modifications.

" *6. Figure 3. The relevant data in fig. 3 covers less than half of the total area and is hard to visualize. The figure should be redesigned for better interpretation. Also, it could be improved if the surface temperature were plot in an independent panel. "*

As the purpose is to emphasize the evolution of meteorological conditions during the different phases of transport, we believe that it would be useful to represent the whole

period in the same figure. We increased nevertheless the plot dimensions, to facilitate the visualization of the main phases (as the dust transport events, that should be visible as variation in wind directions and increased wind intensity respect to the other days). Ground temperature is reported on a separate panel to improve the clarity of the figure.

" *7. Page 8, line 29 "Meteorological evolution is integrated with the aerosol optical variability from LiDAR (see Fig. 2) and with ground aerosol 30 number concentration and volume size distribution (estimated as the volume of a sphere of diameter corresponding to the volumeequivalent particle diameter) at SPC and CMN (see Fig. 4)" Meteorological parameters, lidar measurements, and in situ measurements are presented separately in different figures. They do not seem to be integrated. "*

What we meant here is that the results are derived from the addition and comparison of different information. To avoid confusion we changed the terminology from "Integrated" to "compared".

" *8. Page 10, line 18 "According to FLEXPART, the import of mineral dust persists until the morning of 23 June, when dust presence is not unambiguously inferable from observations but the aerosol mask still indicates the presence of intermediate depolarizing particles below 2000 m. The second desert dust event predicted by FLEXPART shows the same timing with respect to observations but, while the APSS and the OPSS indicate a similar dust burden for the two desert aerosol advection events, the dust load indicated by the model(between 3 and 5* µgm^{-3}*) appears lower respect to the previous events". In addition to limitations of the model, differences between estimations of FLEXPART over the SPC site at 1-2km and 3- 4km and surface-level measurements at SPC (different altitude level) or CMN (different location) might be partly caused by aerosol time-space variability. Have the authors considered a comparison against lidar-derived dust concentration? (These can be retrieved with the POLIPHON method by*

Mamouri [20142]). "

The use of FLEXPART trajectories in this paper is serving as a support for assessing the nature of the observed particle layers. It is out of the scope of the study to provide an accurate estimate of the amount of dust transported, that would require indeed to take into account several possible sources of variability. Moreover, the possible inaccuracies on the aerosol depolarization values retrieval presented above, make difficult to apply the POLIPHON method properly. We believe that this would lead to an additional work that would go far beyond the purpose of the paper. We decided therefore to report dust air mass fraction evolution instead of the mineral dust reconstructed concentration, to avoid additional uncertainties coming from the estimate of the aerosol load, as rightly suggested by the reviewer.

The information brought by such analysis, ancillary to the understanding of the nature of the layers observed by the LiDAR and in-situ measurements, remains mostly unchanged. The text on section 2.5 will be modified as follows: "To give an estimate of the variability in the mass of mineral dust advected over SPC we compute, for each release, the mass fraction of trajectories that encounter dust emissive regions respect to the total mass of the released cluster").

From (10,16): "Maximum mineral aerosol fraction from FLEXPART analysis occurs on June 20, both at the upper layer (9%) and at the bottom layer (9% also. According to FLEXPART, the import of mineral dust at the lower layer persists until the morning of 23 June, when dust presence is not unambiguously inferable from observations but the aerosol mask still indicates the presence of intermediate depolarizing particles below 2000 m. The second desert dust event predicted by FLEXPART again shows the same timing with respect to observation and also confirms the presence of a thick layer of dust that involves at the same time the 1000–2000 m and 3000–4000 m layers. The estimated mass fraction contribution on the contrary, especially in the lower layer (between 2 and 4 %), appears inferior respect to the previous events."

" *9. Figure 7. This figure does not seem to add any information to what can be seen in Fig. 6 and what is on the text. I think it can be removed. "*

We agree that this information may be redundant, and we removed it from the manuscript.

" *10 .Issues regarding section 7:*

Name of the section: "Effect of aerosol hygroscopic growth on aerosol particles light scattering and depolarization" -> Effect of aerosol hygroscopic growth scattering and depolarization "

Name of the section modified.

"*Page 12, line 19. "LiDAR data (Fig. 2) frequently show, during early morning hours, a shallow layer of non-depolarizing aerosol below 300 m height, more easily visible during days characterized by desert dust and mixed dust events". Below 300 m the overlap of the lidar is not complete. Despite the authors claim that "Experimental correction allows the reconstruction of the LiDAR backscattering profile down to around 100 m, with an acceptable uncertainty (4-17)", they also say that "The reader should notice that the lower depolarization values that we observe respect what usually found in literature (especially for the dust layers) are more likely linked to the calibration process, and in particular to the difficulty in individuating completely aerosol free 5 layers in the vertical span of the adopted LiDAR system (from ground to 7 Km) (7-5)" The question is: to what extent can we trust depolarization measurements below complete overlap?*"

For what concerns the uncertainties on the depolarization in the region of partial overlap, the Volume depolarization δ (being derived as a ratio of two signals both characterized by the same partial overlap loss) is not affected. Not so the δ_a, which is computed from δ and the total backscatter ratio (R), which can be impacted by systematic errors when retrieved in the partial overlap region. Errors on the R are discussed in Biavati et al. (2011), where the algorithm for partial overlap correction was presented; for the present work, the relative error on the reconstructed R remains below 10% in the altitude range where the correction has been applied.

The relative error on the δ_a in the region of partial overlap decrease with increasing R and increase for increasing δ . For the values of δ that we found in our work, the error on δ_a remain below 15% for R greater than 1.5 and below 30% for R greater than 1.25.

"*Figure 11 and Figure S2. For this kind of study it might be more appropriate to show the aerosol and humidity profiles at 5:00 UTC as regular plots (not color plots).*"

We modified the plots accordingly.

"*Page 12, line 29: [. . .]. In Fig. 11 and Fig. S2 we can see that in cases with presence of dust the depolarization decreases with RH. But this can be seen as the opposite: elevated dust layers (within dry hot airmasses from the Sahara) uncoupled from the PBL result in a decrease in the RH. In order to state that the dust depolarization properties are affected by the humidity, the authors should prove that a noticeable amount of dust actually reaches the lower altitudes where RH > 60 %. For instance, for the case used as example (30 June), at 5 UTC (time of the sounding) we do not see an especially high concentration of coarse particles at surface level compared to, for instance, 1 July (Fig. 4). Also, it would be interesting if the authors showed, in*

addition to the RH profile, the temperature profile for 30 June. This way we could see it a thermal inversion between the dust and the lower layers keeps them uncoupled. Finally, on 1 July we can see aerosols classified as dust reaching the surface (it is also the day with highest coarse-mode concentration at surface level). Although no soundings might be available at those times, it would be interesting to compare the depolarization values at the lowest altitudes available and the relative humidity at surface level. "

We used the 30 June as a case example as in this day the effect of RH variation on the vertical profile of δ_a was particularly easy to individuate. In figure 1 we compare the δ_a and RH profiles with the potential temperature (TH) profile: in particular, in contrast to the nearly isothermal layer in correspondence of the core of the dust layer, we can notice that the δ_a depletion, and the RH sharp increase observed below 500m, are associated to a notably more stable TH profile.

In this case, nevertheless, we agree with the reviewer that the amount of dust at the ground was less meaningful than what observed during the 1 July, but indeed for this day we don't have the early morning radiosounding. We therefore report in figure 2 the temporal evolution of δ_a near ground ($\sim$150m) compared with RH and Temperature at the ground. It is possible to notice the clear decrease of δ_a during the early hours of the morning, associated to high values of RH (higher than 70%) and low values of temperature (less than 25°C). During late afternoon this effect is less evident both because of less extreme values of RH and T and because of the deposition of coarser and more depolarizing particles (see for example Fig. 2 and Fig.4 of the manuscript).

In the manuscript, for sake of completeness, we decided then to remove the single case study and discuss the vertical profiles of δ_a, TH and RH for dust and dust free days, where dust days corresponds to the days of increased coarse particles at the ground (from 20 to 22 June and from 30 June to 2 July). We discuss it on section 7, from (12,23) on.

Other comments, language errors and typos:
The manuscript was revised to fix language mistakes and typos.

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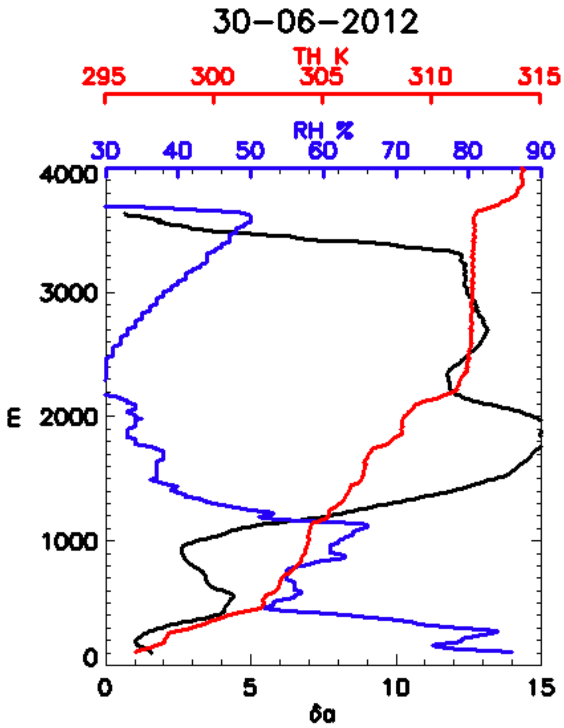

Fig. 1. Vertical profiles of delta_a (black), TH (red) and RH (blue) for 30 June, 05:00 UTC.

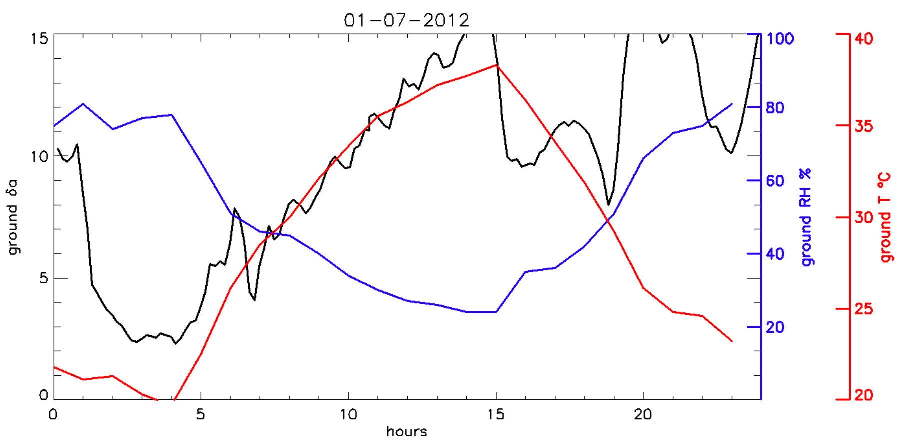

Fig. 2. Temporal evolution of delta_a (black) at 150m, and TH (red) and RH (blue) at the ground, 1 July case study