Review on "Comparison of vertical aerosol extinction coefficients from in-situ and LIDAR measurements"

We would like to thank the referees for their detailed and constructive comments, which helped us to improve our manuscript. Our answers to the comments are given below in blue letters, while the referee comments are given in black italics. Additionally, we added the changes we made in the revised manuscript in **blue bold** letters.

Reply to referee #1:

Dear authors, After reading the supplement related to the description of the lidar data treatment, I still feel that the lidar analysis is inadequate. I have the following criticism and I will insist on that, especially because the title of the paper is on the "comparison of aerosol extinction from in-situ and lidar measurements": It is clear that the lidar does not operate a Raman channel in order to "measure" extinction. This is a backscatter/depolarization system, capable of providing aerosol backscatter coefficient and particle linear depolarization ratio. Thus, the title is inappropriate, since this is not an extinction measurement from the lidar point of view. The lidar researchers try to "estimate" the extinction coefficient, but this is a challenging task, especially for the height range between 50 and 800 m, where the Zeppelin was employed. This is because there are two unknowns for this region regarding the lidar inversion, namely the lidar ratio within the boundary layer (which may be different from free-tropospheric LR) and the incomplete overlap function. An iterative method for the estimation of the overlap function is used along with assumptions on the lidar ratio based on back-trajectories and a final consistency check against the AOD provided by a sunphotometer (including fine-tuning of the lidar ratio to match the total AOD). However, we will never know if the lidar ratio tuning Interactive actually corrects possible biases due to inadequate overlap correction in the boundary layer, or AOD differences in the free-troposphere. To my opinion, the authors have to use the in-situ measurements in the PBL and try to calculate the LR in ambient conditions using Mie scattering codes in order to minimize the assumptions for the lidar inversion. Otherwise, the authors should change the title and alter the language within the manuscript when it comes to the lidar-derived extinction (this is not a measurement but only estimation with lot of assumptions). I insist at this point, since it would be unfair to other closure studies employing lidar and in-situ, to show that this is a simple task that gives us a very good agreement.

We decided to change the title as follows "Comparison of aerosol extinction coefficients as retrieved from in-situ measurements and elastic back-scatter LIDAR". We specified the terms "retrieved" and "elastic back-scatter" since LIDAR retrievals of extinction coefficients are generally based upon assumptions. LIDARs do not measure extinction coefficients directly, instead they retrieve them by using assumptions which then cause a range of uncertainties. In principle, this uncertainty is lower for Raman LIDARs, but it strongly depends on system performance.

In order to make it clear, which LIDAR we used and that the LIDAR did not measure the extinction coefficient, we changed the text in the manuscript in several occasions:

Revised text: Page: 18611; line: 10-11:

Additionally, a single wavelength polarization diversity elastic LIDAR system provided estimates of aerosol extinction coefficients using the Klett method to accomplish the inversion of the signal, for a vertically resolved comparison between in-situ and remote sensing results.

Page: 18611; line: 22:

LIDAR estimates capture these dynamic features well…

Page: 18613; line: 6-7:

This offered a unique opportunity to compare in-situ measurements to low altitude LIDAR estimates, which is known to be challenging.

Page: 18631; line: 6-11:

At the same time vertically resolved extinction coefficients were retrieved with the remote sensing LIDAR system, which provides directly results for aerosols at ambient RH (for more detail see Sect. 2.2.5). As discussed in Sect. 2.2.5 the LIDAR applied in this study cannot directly measure the extinction coefficients, therefore a LR had to be assumed. Results were calculated for three separate fixed LR of 30, 50 and 70 sr, where the value of 50 sr is assumed to be the best-guess solution for the measurement location and the prevailing aerosol type. More discussion on the selection of the LR can be found in Sect. 2.2.5.

Page: 18632; line: 8-9:

The increased extinction coefficients retrieved by the LIDAR at approximately 300 m above ground during P1 and P2 could indicate an aerosol layer and/or an increased RH.

With regard to the request of the reviewer to use in-situ data combined with Mie-codes to perform calculations of the LR, we would like to stress the fact that the goal of this paper is to compare results from two independent methods. Therefore, we preferred to use LRs previously presented in the literature to obtain two separate data-sets to compare.

However, we followed the referee's suggestion to evaluate our LR assumptions using the in-situ data but we would like to note that also the in-situ data is subject to many uncertainties. In this respect, we calculated the aerosol back-scatter cross sections using in-situ data and Mie codes and the ratio of these values to the in-situ aerosol extinction coefficients gives now LR from in-situ data.

To keep the discussion on the LR all together we shifted the paragraph on page 18633, lines 6-15 to section 2.2.5, page 18624, line 4.

Added and revised text: Page: 18624; line: 3:

Table 1 shows a list of LR used in our inversion, classified according to *R*, DR and altitude. The Po Valley aerosol is predominantly of continental origin and therefore LR values between 30 and 70 sr seem to fit best as found using the CALIPSO model by Omar et al. (2009) for clean and polluted continental aerosol particles, respectively, both at λ=532 nm. These values agree well with model results for continental aerosol presented by Barnaba and Gobbi (2004) who found LR values of 60 sr. Measurements performed in Southern Italy found LR values of approximately 50 sr below 2 km at a wavelength of 351 nm (Pisani, 2006; De Tomasi et al., 2006). Discrepancies in LR may arise from the selected method to retrieve it and from the exact location. For instance, Müller et al. (2007) showed that comparing elastic LIDAR against AERONET sun photometers generally yields higher LR compared to LR directly from Raman LIDARS. In order to evaluate the LR assumption from literature data, we performed Mie calculations for the back-scatter coefficients using the airborne in-situ data. By

applying Eq. (11) the LR was calculated and yielded values between 51-67 sr, with a mean value of 58 ± 4 sr. These results strongly support our LR selection from the literature.

We also compared aerosol optical depth (AOD) obtained from the column-integrated LIDAR extinction (at 532 nm) to the AOD from a sun sun-photometer (at 500 nm) at the same site (Campanelli et al., 2007) within the framework of the SKYrad NETwork (http://atmos2.cr.chiba-u.jp/skynet/). The comparison of the AOD variability during the time frame of the PEGASOS campaign showed good agreement between the two data sets. For this period LIDAR derived AOD, using a LR equal to 70 sr, yielded on average 7% higher values than those from the sun-photometer. A sensitivity study changing the value of LR to 50 and 30 sr resulted in underestimations of 5 and 25 %, respectively. Thus, in this range of LR values, LIDAR agrees with the sun-photometer in a column-integrated sense, within the reported limit. The supplement provides an in-depth discussion of LIDAR data treatment.

Reply to referee #2:

The manuscript presents an interesting study with a valuable data set of in-situ measurements retrieved in the lower part of the troposphere, along a period that allows searching the planetary boundary layer evolution. The instrumental set up allows the study of the aerosol absorption and scattering coefficients. Special care has been paid to the consideration of hygroscopic growth effects on the scattering coefficient. This in-situ data set has been used to test the evolution of the aerosol optical properties within the lowest 700 m of the troposphere. Similarities and differences between measurements at the Earth's surface and those at different levels have been used for discussing about the evolution of the atmospheric aerosol in the planetary boundary layer. The optical properties retrieved with the in-situ procedures have been combined for describing the vertical changes of the extinction coefficient. A final comparison among these extinction coefficients and the vertical profiles of extinction coefficient retrieved from lidar measurements is presented. The study is worthy to be published in Atmospheric Physics and Chemistry, but the manuscript requires substantial review before being acceptable for publication. In the following I include the general and particular comments that the authors must address Interactive for improving their manuscript.

General comments:

1) *In the first part of the manuscript the authors present the procedures to retrieve the aerosol optical properties from in-situ measurements. Procedures for direct measurements of the scattering coefficient (at the ground station) and the absorption coefficient (both on board the Zeppelin and at the ground station) are presented and discussed. Furthermore, explanations on the indirect procedure used to retrieve the scattering coefficient from the measured size distributions measured on board the Zeppelin are presented. Concerning this last procedure, the authors must improve the text, because it is a little bit confusing (see particular comments below).*

The answer to this comment can be found below where the reviewer presents the particular comments.

2) *After deriving "dry" properties with the in-situ measurements, the hygroscopic growth has been considered, including measurements of growth factor with an appropriate set up. In this way the ambient aerosol properties have been retrieved from the in-situ measurements. Getting the scattering and absorption coefficients at ambient conditions the authors combined them to get the extinction coefficient, both at the surface level and at different levels, in the new mixing layer, residual layer and developed mixing layer. At this point it is worthy to emphasize the large amount of assumptions used in some retrieval (e.g. the scattering coefficient on board the Zeppelin, the absorption coefficient at SPC based on assuming the Angström exponent retrieved on board the Zeppelin) and the impact on the retrieval of the extinction coefficient. In this sense, the authors must emphasize on the uncertainties associated to the quantities analyzed in this study.*

To emphasize our retrieval method and the associated uncertainties we added the following text to the manuscript:

<u>Revised text: Page: 18621; line: 7-9:</u>

In this respect, the calculation of the airborne scattering coefficient relies on the measured particle size distribution, the retrieved index of refraction of the dry particles and their hygroscopic growth. The most crucial parameter is the selection of the index of refraction, which leads to the largest uncertainties for the scattering coefficient. The absorption coefficient, on the other hand, is assumed

not to vary substantially with ambient RH and therefore no ambient correction was applied. However, the ground based absorption coefficient had to be recalculated for a different wavelength using Eq. (7). In order to do so, the Ångström exponent obtained from the airborne data-set was used.

3) *In the last part of the manuscript, the authors try to compare these values with the lidar retrievals. For this last purpose the authors rely on an elastic lidar (the Raman night time measurements are only used for overlap corrections of the system, as they stated). This is a weak point of the manuscript, because the only way they can derive the extinction coefficient with the elastic lidar is by assuming the so call aerosol lidar ratio, LR, as a constant value for the whole aerosol profile. This is not a single task, as Table 1 reveals, because the criterion used for selecting one lidar ration is clearly ambiguous (overlapping of the ranges assigned to different aerosol together with over- lap of the LRs). In this way the uncertainty of this retrieval is really large, this can lead to uncertainties around 30-50% in the retrieval of the backscattering coefficient or the scattering ratio (Figure 4 in Supplement) and to uncertainties larger than 100% in the extinction coefficient (Figure 4 in Supplement and Figure 8 in the manuscript). In this way, the comparison between remote sensing and in-situ retrievals of extinction coefficient has no sense. So, the authors must consider changing the tittle of the manuscript and reformulating the discussion on the comparison with the extinction coefficient profiles. It is evident that the large uncertainties of the extinction profiles retrieved from elastic lidars do not give reliability to the last part of the study.*

Indeed, the LIDAR used for this comparison is not the best type for such a task as the LR can vary strongly depending on the type of aerosol. However, literature data exists presenting LRs for similar locations and for a similar type of aerosol (in our case: continental aerosol). As this LIDAR system was the only one available in our campaign, we decided to explore if it is actually possible to retrieve comparable extinction coefficients from these two independent techniques despite having to rely on literature data.

We also decided to change the title as follows: "Comparison of aerosol extinction coefficients as retrieved from in-situ measurements and elastic back-scatter LIDAR".

As also mentioned in the last part of the answer to referee #1 we performed additional calculations using the airborne in-situ dataset combined with Mie codes to retrieve back-scatter values and finally derive in-situ LR. These calculations yielded LRs between 51 and 67 sr with a mean value of 58 ± 4 sr. As these values coincide with the LR range proposed by the literature we are confident that they are representative for the aerosol measured during this flight day.

To keep the discussion on the LR all together we decided to shift the paragraph on page 18633, lines 6-15 to section 2.2.5, page 18624, line 4.

<u>Added and revised text: Page: 18624; line: 3:</u>

Table 1 shows a list of LR used in our inversion, classified according to *R*, DR and altitude. The Po Valley aerosol is predominantly of continental origin and therefore LR values between 30 and 70 sr seem to fit best as found using the CALIPSO model by Omar et al. (2009) for clean and polluted continental aerosol particles, respectively, both at λ=532 nm. These values agree well with model results for continental aerosol presented by Barnaba and Gobbi (2004) who found LR values of 60 sr.

Measurements performed in Southern Italy found LR values of approximately 50 sr below 2 km at a wavelength of 351 nm (Pisani, 2006; De Tomasi et al., 2006). Discrepancies in LR may arise from the selected method to retrieve it and from the exact location. For instance, Müller et al. (2007) showed that comparing elastic LIDAR against AERONET sun photometers generally yields higher LR compared to LR directly from Raman LIDARS. In order to evaluate the LR assumption from literature data, we performed Mie calculations for the back-scatter coefficients using the airborne in-situ data. By applying Eq. (11) the LR was calculated and yielded values between 51-67 sr, with a mean value of 58 ± 4 sr. These results strongly support our LR selection from the literature.

We also compared aerosol optical depth (AOD) obtained from the column-integrated LIDAR extinction (at 532 nm) to the AOD from a sun sun-photometer (at 500 nm) at the same site (Campanelli et al., 2007) within the framework of the SKYrad NETwork (http://atmos2.cr.chiba-u.jp/skynet/). The comparison of the AOD variability during the time frame of the PEGASOS campaign showed good agreement between the two data sets. For this period LIDAR derived AOD, using a LR equal to 70 sr, yielded on average 7% higher values than those from the sun-photometer. A sensitivity study changing the value of LR to 50 and 30 sr resulted in underestimations of 5 and 25 %, respectively. Thus, in this range of LR values, LIDAR agrees with the sun-photometer in a column-integrated sense, within the reported limit. The supplement provides an in-depth discussion of LIDAR data treatment.

4) *Particular comments*

This section includes some comments in different sections of the manuscript that require revision.

Page 18615 Line 10. The meaning of the acronym WHOPS must be presented the first time this acronym is presented.

The paragraph formerly presented on page 18615, line 10 is now shifted to the next section where the term WHOPS is specified.

Page 18615 Line 10. The authors must emphasize that they only retrieve the real part of the refractive index. In this sense, they must discus the limitation of this retrieval. Taking into account the single scattering albedo values retrieved in a later section, the assumption of negligible value for the imaginary part of the refractive index requires at least a comment. The retrieval of the refractive index is crucial for the retrieval of scattering coefficient on board of the Zeppelin that finally affect the quality of the retrieval of the extinction coefficient from the in-situ measurements.

The discussion on the effective index of refraction was split between two different passages. Therefore, we decided to present all limitations and assumptions together in Sect. 2.2.2.

Added text: Page: 18616; line: 6:

The term "effective" reflects the fact that several simplifying assumptions are made in the Mie calculations. The particles are assumed to be perfectly spherical and present a homogeneous internal mixture, and the imaginary part of the index of refraction is assumed to be zero. The latter approximation is justified by the fact that scattering coefficients exceed absorption coefficients by a factor of 8 (see Sect. 2.2.2 and Fig. 4). In this manner, an average effective index of refraction of 1.43 ± 0.04 (± 1σ uncertainty) was determined by Rosati et al. (2015b) for the particles probed during the flight in this study.

Page 18615 Line 21. The text defining the Growth Factor must be corrected to be coherent with equation 1.

Revised text: Page: 18615; line: 19-21:

The airborne platform was equipped with the white-light humidified optical particle spectrometer (WHOPS) to measure the hygroscopic growth factor (GF), defined as the ratio of the particle diameter at an elevated RH (D_{wet}) to the one at dry conditions (D_{dry}).

Page 18616. Lines 4-6. The following text is not clear: "By comparing the dry optical response (e.g. scattering cross section) with the initially selected diameter in the DMA the effective index of refraction can be inferred." Please, explain better the procedure for deriving the refractive index.

Revised text: Page: 18616; line: 1-6:

The scattering cross section ("optical size") of these particles is then alternately determined at dry conditions and at high RH by either leading the particles directly into the WELAS or by first exposing them to typically 95% RH before measurement in the WELAS. The dry responses from the two different techniques can then be compared to infer the index of refraction of the selected dry particles (details on the approach are presented in Rosati et al., 2015a). Assuming an index of refraction, the scattering cross section can be calculated from the dry diameter using Mie theory. The index of refraction that brings this theoretical scattering cross section into agreement with the measured one is defined as the effective index of refraction in the context of this work.

Page 18616. Lines 8-9. The following text is also unclear: "Relating the scattering cross section of the humidified particles to the mobility diameter provides the hygroscopic GF." Reformulate this section to help the reader to follow the procedures used.

Revised text: Page: 18616; line: 8-9:

The humidified mode aims at measuring the hygroscopic growth factors of the selected particles, following the approach in Rosati et al. (2015a). For this purpose, the measured scattering cross section of the humidified particles is converted to an optical diameter representing D_{wet}, such that the hygroscopic growth factor can be inferred with Eq. (1). In order to obtain meaningful D_{wet} and GF values, it is crucial to use the true index of refraction of the solution droplets in the Mie calculations.

Page 18620. Section 2.2.4. Please, considering the assumptions made in this section, offer an estimate of the uncertainty associated to the retrieval of the absorption coefficient at the ground station.

For the discussion of the absorption coefficient at the ground station we would like to add some information in the mentioned section, referring to the influence of the recalculations of the measurements for a different wavelength by using results from the airborne dataset.

Revised text: Page: 18620; line: 15:

During this flight the absorption Ångström exponent α_a amounted on average to 0.93 ± 0.15 (mean ± SD). Then Eq. (7) was applied to recalculate μ_a measured by the MAAP to the wavelength of interest (520 nm):

$$\mu_{a,MAAP}(520nm) = \mu_{a,MAAP}(637nm) \cdot \left(\frac{520nm}{637nm}\right)^{-\alpha_a} \quad (7)$$

This introduces an additional uncertainty of 3%, leading to a final uncertainty in $\mu_{a,MAAP}$(520nm) of ±13%.

Table 1, is a good example of the difficulties associated to the choice of the aerosol lidar ratio, and a reason to consider the large uncertainty associated to the extinction retrieval using only elastic lidar.

<u>Page 18624 Lines 8-14.</u> These statements are really broad and only suggest some confidence on the retrieval of the Backscattering coefficient using Klett-Fernald algorithm (see general comment). As the authors emphasize the local effect of the lidar ratio choice on the extinction coefficient could lead to really large uncertainties, a fact that is evidenced on Figure 8. This justifies the need for a change on the title and on the way the authors focus the comparison between in-situ and lidar retrieval.

As suggested we changed the title of the manuscript to: "Comparison of aerosol extinction coefficients as retrieved from in-situ measurements and elastic back-scatter LIDAR".

<u>Page 18624 Section 3.</u> Some indications on the uncertainty of the Planetary Boundary Layer Height retrieval are required.

<u>Added text: Page: 18624; line: 22:</u>

Retrieval of the estimated mixing layer height is performed by operating on a graphical interface presenting the maximum gradient points in the signal daily plot. In this way, the operator solves the ambiguities related to multiple relative maxima often present in the ceilometer signal (e.g., Angelini et al., 2009). The typical imprecision due to the operator's choice amounts to 3 pixels, i.e. ±45 m.

Figure captions for Fig 4 and 7 require more details on how the figures are built. They must include description on the meaning of the squares that are presented in the text. The figure caption in itself must help to understand the figure without reading the whole manuscript.

<u>Adapted Figure Caption: Figure 4:</u>

Time evolution of the dry scattering (a) and absorption (b) coefficients from the airborne and ground based platforms. The color scale represents the magnitude of the coefficients. The uncertainty of the dry scattering coefficients is estimated to be ±18%, while ±20% is estimated for the absorption coefficients. Additionally, the filled area in (a) and (b) denotes the estimated mixing layer height and each height profile (P1-P6) is marked.

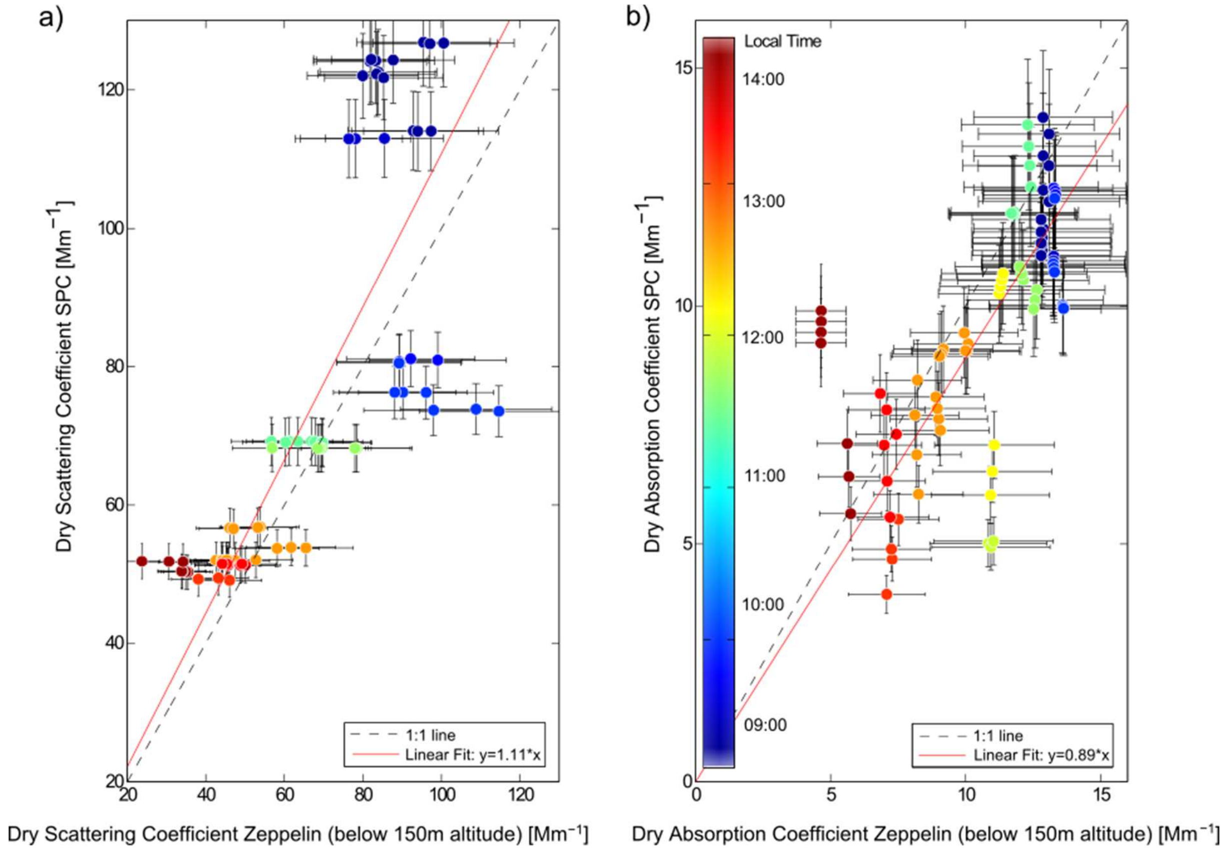
<u>Adapted Figure Caption: Figure 7:</u>

Time evolution of the dry (a) and ambient (b) single scattering albedo from the airborne and ground based platforms. The color scale represents the magnitude of the coefficients. The uncertainty of the dry single scattering albedo is estimated to be ±7% and ±26% for SPC and the Zeppelin, respectively. The uncertainty of the ambient single scattering albedo is estimated to be ±36% and ±26% for SPC and the airborne data, respectively. Additionally, the filled area in (a) and (b) denotes the estimated mixing layer height and each height profile (P1-P6) is marked.

Figure 5 includes appropriate information on uncertainties. But this figure requires additional information on the quantitative analyses of the linear fitting of the data. The Interactive comment, in Page 18627 Line 20, indicating a "very good correlation" is really ambiguous.

To show a quantitative analysis of the linear fitting of the data we included the regression curve and equation in Fig. 5 and in the text.

New Figure 5:



Adapted Figure Caption: Figure 5:

Comparison of dry scattering (a) and absorption (b) coefficients for ground-based and airborne measurements. The Zeppelin results were restricted to altitudes below 150 m in order to eliminate differences due to potential changes in atmospheric layers. The colors of the symbols reflect the time of the day according to the color bar in (b). Additionally, the regression curves and equations are shown. (a) For the ground based data an uncertainty of 5% is estimated while ~18% was found for the airborne calculations. (b) The uncertainty of the airborne absorption coefficient is estimated to be ±20%, while ±13% is estimated for the ground results.

Revised text: Page: 18627; line: 20-22:

By using a linear fit, a regression equation of y=1.11*x was found, where y denotes the dry scattering coefficient in SPC and x the dry scattering coefficient from the Zeppelin. Figure 5b illustrates the dry absorption coefficients. In this case the linear regression yields the equation: y=0.89*x, where y represents the dry absorption coefficient in SPC and x the dry absorption coefficient from the Zeppelin.