



Comparison of
vertical aerosol
extinction
coefficients

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

B. Rosati¹, E. Herrmann¹, S. Bucci², F. Fierli², F. Cairo², M. Gysel¹, R. Tillmann³, J. Größ⁴, G. P. Gobbi², L. Di Liberto², G. Di Donfrancesco⁵, A. Wiedensohler⁴, E. Weingartner^{1,a}, A. Virtanen⁶, T. F. Mentel³, and U. Baltensperger¹

¹Laboratory of Atmospheric Chemistry, Paul Scherrer Institute, 5232 Villigen PSI, Switzerland

²Institute of Atmospheric Sciences and Climate (ISAC-CNR), National Research Council, 00133 Rome, Italy

³Institute for Energy and Climate Research (IEK-8), Forschungszentrum Jülich, 52428 Jülich, Germany

⁴Leibniz Institute for Tropospheric Research, 04318 Leipzig, Germany

⁵Italian National Agency for new Technologies, Energy and Environment (ENEA), 00044 Frascati, Italy

⁶Department of Applied Physics, University of Eastern Finland, 70211 Kuopio, Finland

^anow at: Institute for Aerosol and Sensor Technology, University of Applied Science Northwestern Switzerland, 5210 Windisch, Switzerland

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Correspondence to: E. Herrmann (erik.herrmann@psi.ch)

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Abstract

Vertical profiles of aerosol optical properties were explored in a case study near the San Pietro Capofiume (SPC) ground station during the PEGASOS Po Valley campaign in the summer of 2012. A Zeppelin NT airship was employed to investigate the effect of the dynamics of the planetary boundary layer at altitudes between ~ 50–800 m above ground. Determined properties included the aerosol size distribution, the hygroscopic growth factor, the effective index of refraction and the light absorption coefficient. The first three parameters were used to retrieve the light scattering coefficient. Simultaneously, direct measurements of both the scattering and absorption coefficient were carried out at the SPC ground station. Additionally, a LIDAR system provided aerosol extinction coefficients for a vertically resolved comparison between in-situ and remote sensing results. First, the airborne results at low altitudes were validated with the ground measurements. Agreement within approximately ± 25 and ± 20 % was found for the dry scattering and absorption coefficient, respectively. The single scattering albedo, ranged between 0.83 to 0.95, indicating the importance of the absorbing particles in the Po Valley region. A clear layering of the atmosphere was observed during the beginning of the flight (until ~ 10 local time) before the mixed layer (ML) was fully developed. Highest extinction coefficients were found at low altitudes, in the new ML, while values in the residual layer, which could be probed at the beginning of the flight at elevated altitudes, were lower. At the end of the flight (after ~ 12 local time) the ML was fully developed, resulting in constant extinction coefficients at all altitudes measured on the Zeppelin NT. LIDAR results captured these dynamic features well and good agreement was found for the extinction coefficients compared to the in-situ results, using fixed LIDAR ratios (LR) between 30 and 70 sr for the altitudes probed with the Zeppelin. These LR are consistent with values for continental aerosol particles that can be expected in this region.

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1 Introduction

Atmospheric aerosol particles are known to interact directly with the incident solar radiation by either scattering or absorbing light. By doing so they can influence the Earth's radiative budget and therefore have an impact on climate (e.g. IPCC, 2013). Due to the layering of the atmosphere, a strong vertical gradient of particle concentration is found between the Earth's surface and the lowest kilometer of the troposphere. Most emissions are trapped in this so-called planetary boundary layer (PBL) which is part of the troposphere and the layer closest to the ground. The PBL is dynamic and strongly influenced by the solar radiation (Stull, 1988). One major point of interest is to know whether surface based measurements can be used to infer aerosol radiative properties at elevated altitudes. For this purpose remote sensing techniques, like light detection and ranging (LIDAR) instruments, are useful to monitor optical properties such as aerosol extinction and back-scattering coefficient over a large range of altitudes. This technique involves a pulsed laser beam to measure the back-scatter by gases and aerosol particles in the atmosphere (Klett, 1981). However, certain limitations exist for this method. One refers to the fact that accurate profiles can only be recorded above a certain threshold given by the altitude where the laser beam is completely within the field of view of the telescope (Sassen and Dodd, 1982). Below this altitude additional assumptions have to be made in order to retrieve the optical properties and different correction schemes have been proposed (see e.g. Biavati et al., 2011). In-situ measurements on aircrafts have been used to validate remote sensing data. However, these are often limited to low RH measurements, while RH typically varies with altitude. Changes in RH may lead to changes in size, shape and index of refraction if the particles are hygroscopic and take up water; hence the optical properties would be altered. Only few studies performed RH-dependent measurements at elevated altitudes (Morgan et al., 2010; Sheridan et al., 2012) and even fewer compared these to optical measurements from LIDARs (Zieger et al., 2011; Sheridan et al., 2012). Additionally, measurements on aircrafts commonly focus on altitudes of several km above ground

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therefore usually starting at the upper part or even above the PBL. Due to limited number of measurements focusing on changes in the PBL over the course of the day, the effects of PBL dynamics on aerosol properties remain poorly understood.

During the PEGASOS (Pan-European Gas–AeroSOls-climate interaction Study) project a Zeppelin NT airship was employed to study aerosols at altitudes between 50–800 m above ground. This offered a unique opportunity to compare in-situ measurements to low altitude LIDAR results which is known to be challenging (see e.g. Sheridan et al., 2012). Data from a flight on 20 June 2012 in the Po Valley (Italy) will be used. Rosati et al. (2015b) presented results of hygroscopicity measurements combined with chemical composition data for the vertical profiles on the same day. Here, this same data is used to calculate in-situ extinction coefficients and compare them to LIDAR observations. A consistency check for the airborne results is done by comparison to ground based data.

2 Experimental

2.1 Site and flight description

During the PEGASOS measurement campaign 2012 vertical profiles were performed near the San Pietro Capofiume (SPC) ground station located in the Po Valley in Italy (see Fig. 1). The general set-up of the Zeppelin NT platform for aerosol measurements as well as meteorological data for the flight on 20 June 2012 were presented in Rosati et al. (2015b). The SPC station is suited to investigate air masses at a rural background area. Due to its vicinity to cities like Bologna (~ 40 km to the south-west) it also offers the possibility to study pollution from regional sources or aerosols transported over longer distances. Several campaigns have already taken place at this site focusing on variations in chemical composition as well as hygroscopic and optical properties (e.g. Mazzola et al., 2010; Saarikoski et al., 2012; Bialek et al., 2014; Decesari et al., 2014). The station is equipped with instruments comparable to those employed on the

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monodisperse aerosol is selected in a DMA. To obtain information on both the dry and humidified particles, they are once directly measured in a WELAS (Palas GmbH, Type 2300; see Heim et al., 2008 or Rosati et al., 2015a for more details) under dry conditions and once after exposure to high RH of typically 95 %. 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. The term “effective” is used because we assume perfectly spherical and internally mixed particles as well as a negligible imaginary part of the index of refraction. Relating the scattering cross section of the humidified particles to the mobility diameter provides the hygroscopic GF. Thereby a gradual decrease in the index of refraction of the grown particles towards the index of refraction of water $m_{\text{H}_2\text{O}} = 1.333$ is assumed. According to Rosati et al. (2015a) the absolute uncertainty in the effective index of refraction amounts to ± 0.04 while the relative uncertainty for $\text{GF} < 3$ is found to be approximately $\pm 10\%$.

The ground station in SPC was equipped with a hygroscopicity tandem differential mobility analyzer (HTDMA; see e.g. Swietlicki et al., 2008). Here GF for dry diameters of 200 nm were used (compare Rosati et al., 2015b). In the HTDMA two DMAs are operated in series and connected to a CPC. In the first DMA a dry monodisperse aerosol is selected which is then exposed to a defined elevated RH. The second DMA coupled to the CPC is used to measure the size distribution of the grown particles. The uncertainty for these HTDMA-GFs is expected to be approximately 5 %, if a 2 % uncertainty is assumed for the RH measurement.

The hygroscopic growth measured with the WHOPS and the HTDMA were used to convert scattering coefficients obtained from measurements of the dry aerosol to the corresponding value at ambient RH, as detailed in Sect. 2.2.3.

2.2.3 Aerosol scattering coefficient

At the SPC ground station the total light scattering coefficients were measured with an integrating nephelometer (TSI Inc., Model 3563) at three different wavelengths of $\lambda = 635, 525$ and 450 nm behind a PM_{10} inlet system and after drying to $\text{RH} < 40\%$.

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The truncation error correction introduced by Anderson et al. (1996) was applied. The uncertainty for these measurements is estimated to be $\pm 5\%$. As no direct measurement of the aerosol scattering coefficient was available aboard the Zeppelin NT airship, it was inferred using the particle size distributions, the effective index of refraction and Mie theory assuming spherical particles (Mie, 1908; Bohren and Huffman, 2007). First, scattering cross sections (σ_s) as a function of particle diameter (D) were calculated using the wavelength of $\lambda = 520$ nm and the range of indices of refraction (m) measured during the flight. This specific wavelength was chosen to compare the airborne data with results from the ground based and remote sensing measurements. The WHOPS retrieval yielded on average $m = 1.42 \pm 0.04$, while a comparison to the directly measured scattering coefficients from SPC showed that using $m = 1.42 \pm 0.02$ for the Mie calculations is enough to explain the variability of the nephelometer data. Second, the scattering coefficients (μ_s) were obtained by integrating the product of σ_s and the measured number size distributions ($\frac{dN}{dD}$) over the full diameter range:

$$\mu_{s,j}(\lambda, m, D) = \int_{D_{\min}}^{D_{\max}} \sigma_s(\lambda, m_j, D_j) \cdot \frac{dN}{dD_j} dD_j \quad (2)$$

The index j can be replaced by dry when calculating the dry scattering coefficients or by wet when the humidified coefficient is regarded. An uncertainty analysis showed that changes in the index of refraction caused the biggest errors in $\mu_{s,\text{dry}}$. Together with the size distribution uncertainty an overall uncertainty of approximately $\pm 18\%$ was obtained for the dry scattering coefficient. It is possible to directly compare ground based and airborne measurements with each other, as both were performed at dry conditions.

For comparison with the LIDAR remote sensing data, the Zeppelin measurements were corrected to ambient RH. The importance of this correction was previously studied by using a humidified nephelometer (Wet-Neph; Fierz-Schmidhauser et al., 2010a, b; Zieger et al., 2010, 2011, 2012, 2013). This instrument directly measures the scat-

tering enhancement due to elevated RH, which can be described by a wavelength (λ) dependent scattering enhancement factor $f(\text{RH}, \lambda)$:

$$f(\text{RH}, \lambda) = \frac{\mu_{\text{s, wet}}(\text{RH}, \lambda)}{\mu_{\text{s, dry}}(\lambda)} \quad (3)$$

As no such instrument was available during the PEGASOS campaign, the humidity correction was achieved by combining the GF results at 95% RH from Rosati et al. (2015b) with the ambient RH measurements to determine an ambient light scattering coefficient. This makes it possible to infer the effect of the hygroscopic growth on the light scattering coefficient through considering the effect on the size distribution. The GF was measured for a monodisperse aerosol of 500 nm but we assume it to be constant over the full size range. Small particles ($D < 200$ nm) could potentially have a different hygroscopic behavior since species like sea salt or mineral dust are predominantly found in the larger size ranges. Nevertheless, this assumption is deemed satisfactory since small particles have a minor contribution on light scattering compared to the effect of the larger sizes. In order to obtain an ambient GF (GF for $\text{RH}_{\text{ambient}}$) the results at $\text{RH} = 95\%$ were recalculated for $\text{RH}_{\text{ambient}}$ using the semi-empirical κ -Köhler theory introduced by Petters and Kreidenweis (2007):

$$\kappa = \frac{(\text{GF}(\text{RH})^3 - 1) \cdot (1 - a_w)}{a_w} \quad (4)$$

where a_w is the water activity which can be inferred from the RH and equilibrium droplet diameter (D_{wet}):

$$a_w = \frac{\text{RH}}{\exp\left(\frac{4\sigma_{\text{s/a}}M_w}{RT\rho_w D_{\text{wet}}}\right)} \quad (5)$$

Here $\sigma_{\text{s/a}}$ is the surface tension of the solution/air interface, M_w the molecular mass of water, R the ideal gas constant, T the absolute temperature and ρ_w the density of water.

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We assume κ to be the same at all RH although this might introduce some bias as former studies found changes of κ with RH at elevated organic fractions (e.g. Pajunoja et al., 2015). However, the potential deviation due to this simplification is small in our case since the GF is anyways small at the moderate RH encountered in this study.

The recalculated GF for $\text{RH}_{\text{ambient}}$ were further used to retrieve humidified aerosol size distributions from the measured dry size distributions. The water uptake has also an influence on the index of refraction which is taken into account for the Mie calculations by applying a volume weighting mixing rule to determine the index of refraction of the grown particles. Finally a humid scattering coefficient ($\mu_{s, \text{wet}}$) was calculated according to Eq. (2). By propagating the uncertainties of the single parameters in Eq. (2), a mean uncertainty of approximately 18% was found for $\mu_{s, \text{wet}}$. The ratio of $\mu_{s, \text{wet}}$ to $\mu_{s, \text{dry}}$ was finally used to calculate $f(\text{RH})$ according to Eq. (3). The uncertainty in $f(\text{RH})$ amounted on average to 25%. Also the ground based data set was corrected for changes due to elevated RH by using hygroscopicity results from the HTDMA (see Sect. 2.2.2 for more details). In this case the size distributions from the SPC-SMPS were recalculated including adjustments for the $\text{RH}_{\text{ambient}}$ in SPC. Then the $f(\text{RH})$ was obtained with Eq. (3) using the dry and humidified size distributions measured and retrieved in SPC and finally it was applied on the directly measured scattering coefficients obtained from the nephelometer to get $\mu_{s, \text{wet}}$.

2.2.4 Aerosol absorption coefficient

A portable aethalometer (AE42, MAGEE Scientific; Berkeley, USA) was mounted in the Zeppelin NT for a continuous measurement of the aerosol light absorbing properties at seven wavelengths. This instrument monitors the attenuation of light through a quartz fiber filter. The signal was then corrected as proposed by Weingartner et al. (2003) for multiple scattering in the filter matrix (“ C value”) and the so called shadowing effect (“ f value”). A C value of 4.75 for $\lambda = 520\text{nm}$ was used according to personal communications with J. P. Putaud who performed a comparison between an aethalometer (model AE31) and a MAAP in summer 2012 in Ispra, Italy. The f value amounted

2.2.5 Aerosol extinction coefficient

The extinction coefficient (μ_e) can be calculated as the sum of the absorption and scattering coefficient.

$$\mu_e(\text{RH}) = \mu_a + \mu_s(\text{RH}) \quad (8)$$

5 For the airborne as well as ground based measurements the dry ($\text{RH} < 30\text{--}40\%$ as recommended by WMO/GAW, 2003) and wet (ambient RH) extinction coefficients were retrieved. μ_a was assumed not to vary substantially with ambient RH. The scattering coefficient, on the other hand, is strongly dependent on RH and was therefore corrected by measurements of the particles' hygroscopic growth (see Sect. 2.2.3). The propa-
10 gated measurement uncertainties for $\mu_{e, \text{dry}}$ and $\mu_{e, \text{wet}}$ amount to $\sim 6\%$ and $\sim 24\%$, respectively, at the SPC ground site, while an uncertainty of approximately 18% is found for both on the aircraft. Please note, that for the airborne data set the relatively large uncertainty in $f(\text{RH})$ is not propagated into the extinction results since the directly calculated $\mu_{s, \text{wet}}$ are used for Eq. (8). The ambient ground based results, however, are
15 influenced by this uncertainty because $f(\text{RH})$ is applied on the directly measured dry scattering coefficients from SPC.

Moreover a single wavelength polarization diversity elastic LIDAR system was deployed at the SPC ground station. This instrument uses a 532 nm pulsed Nd-YAG laser source, with a pulse duration of 1 ns , energy of $400\text{ }\mu\text{J}$ and a repetition rate of
20 1 kHz . The LIDAR system collects the radiation elastically back-scattered from the atmosphere (Rayleigh scattering) by separately detecting its parallel and cross polarization components with respect to the polarization of the laser. Additional technical details of the systems are presented in Cairo et al. (2012). The overlap of the laser beam within the field of view (FOV) of the detector begins at few tens of meters from
25 the system, and is complete at few hundred meters (50 and 300 m , respectively, in the simple approximation of a conical laser beam and telescope FOV). A nitrogen Raman scattering channel at 608 nm is also present, however these data are available only

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for nighttime conditions. This channel, which collects a signal proportional to the atmospheric molecular density, is used for the correction of the Rayleigh signal coming from the region of partial superposition between laser and FOV, the Partial Overlap Region (POR) where the back-scattered signal is partially lost. This correction is done by comparing the Raman signal received from the POR with the molecular density profile obtained by collocated simultaneous pressure and temperature balloon measurements, and thus retrieving an overlap correction function, from the ratio of the Raman signal to the molecular density. Uncertainties in the determination of the overlap function arise mainly from the pressure and temperature uncertainties, and from the Raman signal counting statistics and are reflected in inaccuracies in the reconstructed signal of around 10 % at 100 m, rapidly decreasing upward. The system provides a profile of back-scatter ratio (R) and Volume Depolarization Ratio (DR) every 5 min for an elevation of up to 15 km, where R and DR are defined as:

$$R = \frac{(\beta_a + \beta_m)}{\beta_m} \quad (9)$$

$$DR = \frac{(\beta_a + \beta_m)^c}{(\beta_a + \beta_m)^p} \quad (10)$$

Herein β_a and β_m are the aerosol and molecular back-scattering coefficient, respectively, and the superscripts p and c refer to their contribution in the parallel and cross polarized back-scattering. R and DR assume the value of 1 and 1.4 % respectively, in regions supposed to be free of aerosol at a normalization altitude z_0 , usually above 7 km. This normalization procedure introduces an additional possible inaccuracy in the data produced, as the derived back-scatter and extinction coefficients at any height z below z_0 , depends not only on the signal at z , but also on the extinction between z and z_0 , on the ratio of the signal at z to the one at z_0 and on the assumed values of R and DR at z_0 Russell et al. (1979). For the here presented data, we have performed a sensitivity test by varying the normalization height z_0 subjectively chosen in the region we supposed to be free of aerosol. This resulted in a dispersion of the extinction data at

these two layers could potentially be significantly different since the RL is mainly dependent on the PBL from the day before, while the new ML is affected by direct emissions from the same day. Then the strong increase in estimated mixing layer height points towards a fully developed ML throughout the probed altitudes after $\sim 12:00$ LT. Aerosol particles are expected to be homogeneously distributed in this layer and therefore their properties should be comparable at all altitudes. One possible exception is the RH which may be dependent on altitude and could thus induce height dependent humidity related effects.

3.1 Aerosol size distributions

Figure 3 illustrates the dry surface area size distributions at different altitudes and times. This kind of distribution was chosen since the optical properties are directly dependent on the surface area of the particles. The colored lines indicate the Zeppelin NT results, where each line represents the distribution in a different layer. Since two separate instruments were combined, the contributions by each instrument as well as the region in between are marked by different symbols. At the beginning of the flight the new ML was probed close to the ground (~ 100 m above ground) with the mode of the distribution at a diameter of ~ 290 nm. In addition, during the first part of the flight the RL was measured at ~ 700 m above ground. The blue line depicts the surface area size distribution in the RL with a mode at ~ 270 nm and only small contribution by particles with diameters larger than ~ 500 nm. In addition, the green and pink lines show two distributions at altitudes below 200 and above 500 m above ground, respectively, for the fully developed ML which is present after approximately 12:00 LT. The results in the fully developed ML are very similar to the ones from the RL with the mode of the distribution at ~ 260 nm. The comparison between surface area distributions in different layers indicates that no major changes occurred throughout the flight except for a slightly larger contribution of particles above approximately 300 nm in the new ML compared to the other layers. Moreover, the surface area size distributions from the SMPS at the SPC ground station before 10:00 LT (morning; black, dashed line) and

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this purpose GF measurements along with ambient RH measurements were used for the calculations described in Sect. 2.2.3. Figure 6 displays the ambient RH (blue line) present at different altitudes during the flight with the Zeppelin NT. The ambient temperature during the flight ranged between 24 and 33 °C. Clear differences with altitude were observed when flying in the fully developed ML while comparable temperatures were found in the new ML and the RL at the beginning of the flight. In addition, also the airborne GF values, recalculated for the ambient RH (see Sect. 2.2.3), are shown in Fig. 6 as well as the scattering enhancement factor $f(\text{RH})$. During the first flight hours the ambient RH was highest reaching values of approximately 60 % at low altitudes, resulting in $f(\text{RH})$ values up to ~ 1.7 in the new ML. These high $f(\text{RH})$ values are not only a function of RH but also of the chemical composition and especially of the fraction of inorganic species present in the particles. As described already in the previous section (Sect. 3.2) an enhanced fraction of nitrates was found in the new ML, which explains the enhanced hygroscopic growth at elevated RH. At the same time but higher altitude (~ 700 m; RL), RH was below ~ 40 % with $f(\text{RH})$ between 1.1 and 1.2. The RL was characterized by lower RH as well as a smaller fraction of inorganics compared to the new ML and the combination of both led to smaller $f(\text{RH})$ values. The second part of the flight (after $\sim 11:00$ LT) was dominated by a low RH in the range of 25–50 % together with a low inorganic fraction, which is reflected in low $f(\text{RH})$ values of 1.1 to 1.2.

The ground data recorded in SPC was also corrected for scattering enhancement effects by utilizing the GF values from the HTDMA and the size distribution measurements reported in Fig. 3 (see Sect. 2.2.3). The ambient RH in SPC varied from a maximum of ~ 50 % at the beginning of the flight to ~ 25 % at the end. The $f(\text{RH})$ ranged between approximately 1.3 and 1.1 during the whole flight and were applied to correct the dry scattering coefficients.

Zieger et al. (2013) previously published values from European sites with continental and background influence. At low RH of ≈ 40 %, $f(\text{RH})$ values between 1 and ~ 1.2

were recorded while at $\approx 60\%$ f (RH) between 1 and ~ 1.8 were found. When comparing these data sets good agreement is found between the two studies.

3.4 Vertical profiles of the single scattering albedo

The magnitude of the scattering coefficient exceeded the absorption coefficient on average by a factor of 7 when considering the dry values presented in Fig. 4. If also RH effects are taken into account (presented in Sect. 3.3 and Fig. 6) the ratio for the ambient coefficients reaches values of approximately 8. The relationship of the two coefficients is relevant for the sign of net aerosol radiation interactions and it is commonly described using the single scattering albedo (SSA; ω_0):

$$\omega_0 = \frac{\mu_s}{\mu_s + \mu_a} \quad (12)$$

Figure 7 illustrates the temporal evolution of both, the dry and ambient SSA for the airborne as well as ground data. Focusing first on the dry SSA (Fig. 7a), the airborne results at ~ 700 m above ground reveal a constant SSA throughout the flight. This is in agreement with expectations, as generally similar aerosol properties were observed at this altitude in the RL (morning) and fully developed ML (afternoon) in terms of size distributions (Fig. 3), composition (Rosati et al., 2015b) and optical properties (Fig. 4). Surprisingly, the dry SSA observed in the new ML before $\sim 10:00$ LT at 100 m above ground as well as at the ground site are similar to those in the RL above. Generally, SSA is rather expected to increase with air mass age due to the formation of non-absorbing secondary aerosol components. However, the unexpected similarity of the dry SSA in the new ML and RL above can be explained with peculiar differences in chemical composition (Sect. 3.2 and Rosati et al., 2015b): the aerosol in the new ML has a strongly increased nitrate mass fraction (non-absorbing) and also a slightly increased BC mass fraction (light absorbing), compared to the aerosol in the RL. This has compensating effects on the SSA. After, 10:00 LT the nitrate mass fraction decreases faster than the BC mass fraction in the evolving mixing layer, likely due to temperature and dilution

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related partitioning effects. This results in decreased dry SSA values at the ground site and 100 m above ground. In the afternoon, when the ML is fully developed, the dry SSA become again similar at all altitudes. A small vertical gradient is seen with lower values of the SSA at 100 m above ground as well as the ground station, which are likely related to the influence of local emissions. The dry SSA values observed in the three layers probed by the Zeppelin are 0.87 ± 0.01 (mean \pm 1SD), 0.89 ± 0.01 and 0.87 ± 0.02 for the new ML, the RL and the fully developed ML, respectively.

Figure 7b displays the ambient SSA where results from Fig. 6 are taken into account. The hygroscopic growth increases the SSA as it mostly affects the scattering coefficient. The ambient RH and thus the light scattering enhancement factor is highest in the new ML. Therefore, the largest increase of the ambient SSA compared to the dry SSA is observed at low altitude in the morning. The ambient SSA values observed in the three layers probed by the Zeppelin amount to 0.92 ± 0.01 (mean \pm 1SD), 0.90 ± 0.01 and 0.88 ± 0.02 for the new ML, the RL and the fully developed ML, respectively. The comparison of the SSA at dry and ambient conditions reveals that hygroscopic growth has a significant effect on the SSA even in cases with relatively low ambient RH such as this case study. Accounting for this effect is important to obtain the correct magnitude and sign of radiative forcing aerosol radiation interactions by anthropogenic aerosols.

Our results are in good agreement with previous studies performed with sun-photometers during the summer months in Ispra, located in the northern Po Valley, which found an average ambient SSA of 0.9 (Takemura et al., 2002). On the other hand, the study by Putaud et al. (2014) (also centered in Ispra) presented a long-term analysis of SSA measured with in-situ instrumentation comparable to the one employed in our case. Their mean SSA, valid for dry conditions ($< 40\%$), ranged between approximately 0.80 and 0.85 for the summer months with little variation between the years 2004–2011. These values appear smaller than those from our study for the fully developed ML when RH had only a small effect on the SSA. A possible reason for this difference might be the averaging over a whole month compared to our case study performed at one specific day but also variations due to slightly different locations.

developed ML ($P3$ – $P6$) are close to those in the RL (high altitudes in $P1$ and $P2$) and lower than those in the new ML (low altitudes in $P1$ and $P2$).

When comparing absolute extinction coefficient values of the in-situ and remote sensing results for the best-guess LR = 50 sr, the latter tends to overestimate μ_e at the higher elevations (> 250 m) in general, while agreement is better at lower altitudes (< 250 m). 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). This agrees well with model results for continental aerosol such as e.g. those presented by Barnaba and Gobbi (2004) who found LR values of 60 sr or from the CALIPSO model (Omar et al., 2009), an automated algorithm using satellite observations, finding LR values of 35 and 70 sr for clean and polluted continental aerosol particles, respectively, both at $\lambda = 532$ nm. 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 LIDARs against AERONET sun photometers generally yields higher LR compared to LR directly derived from Raman LIDARs.

In order to investigate the role of the selected LR on the agreement between in-situ and remote sensing data, a sensitivity study on the dependence of the LIDAR results on LR was carried out. In this respect LR values of 30 (orange line) and 70 sr (dark red line) in addition to the best-guess scenario were used, thereby covering the potential range of LR for continental aerosol reported in the literature (see Table 1). Largest discrepancies of up to 50 % between LIDAR retrievals and in-situ measurements are found for LRs of 50 and 70 sr above 600 m above ground. Choosing LR = 30 sr instead results in agreement within 5–20 % for these altitudes, depending on the actual flight profile. At low altitudes (< 300 m) the opposite is seen finding agreement within 15 % between in-situ and remote sensing data for a LR of 50–70 sr. This altitude dependence of the best-fit LR may on the one hand be related to true differences in aerosol properties at different altitudes, particularly in the morning, but on the other hand potential systematic biases in the overlap correction for low altitudes may also play a role.

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Acknowledgements. This work was supported by the EC project PEGASOS, funded by the European Commission under the Framework Programme 7 (FP7-ENV-2010-265148). We thank all PEGASOS participants for the excellent team work during the campaigns. Additionally we thank F. Holland for the analysis and maintenance of all meteorological and GPS data on the Zeppelin. We thank M. Zanatta for support concerning the nephelometers and the associated corrections. We also thank G. Bonafé for the meteorological data provided for the ground station and M. Campanelli for the sun-photometer data. M. Gysel was supported by the ERC under grant 615922-BLACARAT and E. Herrmann by MeteoSwiss in the framework of the Global Atmosphere Watch program. The LIDAR and ceilometer data have been acquired and made available in the framework of the project SUPERSITO of ARPA Emilia Romagna (DRG no. 428/10).

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Table 1. List of LIDAR ratios (LR) selected as function of back-scatter ratio (R) and depolarization ratio (DR). For Polluted continental air masses, different values have been tentatively employed for a sensitivity study.

LR [sr]	R	DR [%]	Aerosol type
70 (30–50)	> 1.05; < 10	< 15	polluted continental (clean continental)
50	> 1.05; < 10	> 10	Saharan dust
60–70	> 1.05; < 10	< 15	biomass burning



Figure 1. Location of the ground station San Pietro Capofiume (SPC) in Italy (adapted from <http://www.italyworldclub.com/emilia/>). The region Emilia Romagna is highlighted showing the main cities in the area. In the lower left corner its position within Italy is indicated.

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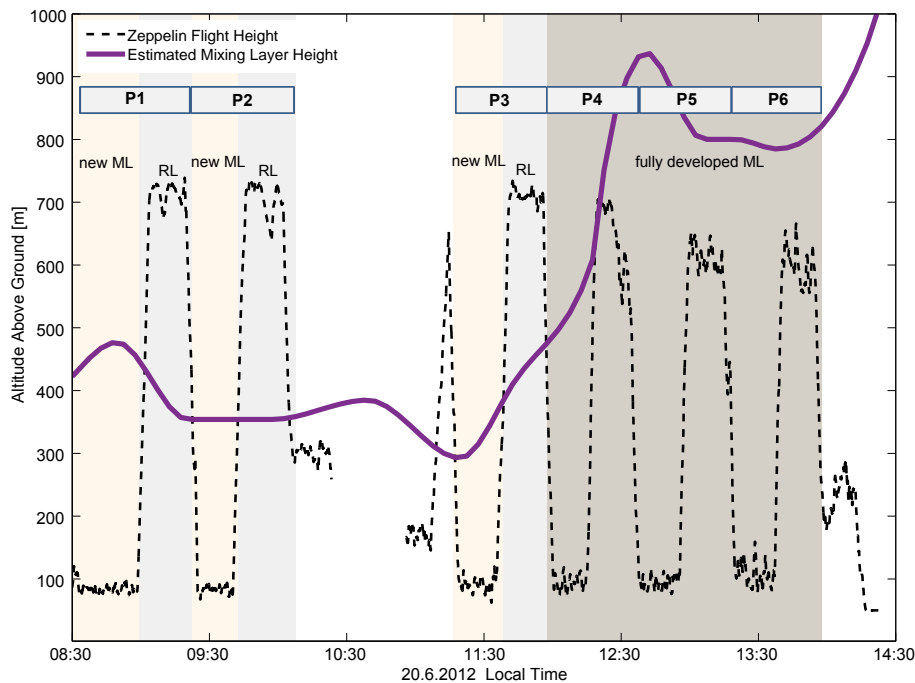


Figure 2. Overview of the flight on 20 June 2012 near San Pietro Capofiume (SPC). The black dashed line depicts the flight altitude of the Zeppelin NT airship, while the violet thick line illustrates the estimated mixing layer height. Additionally, the different layers which were probed are labeled and colored differently. Also, profiles *P1* to *P6* are marked.

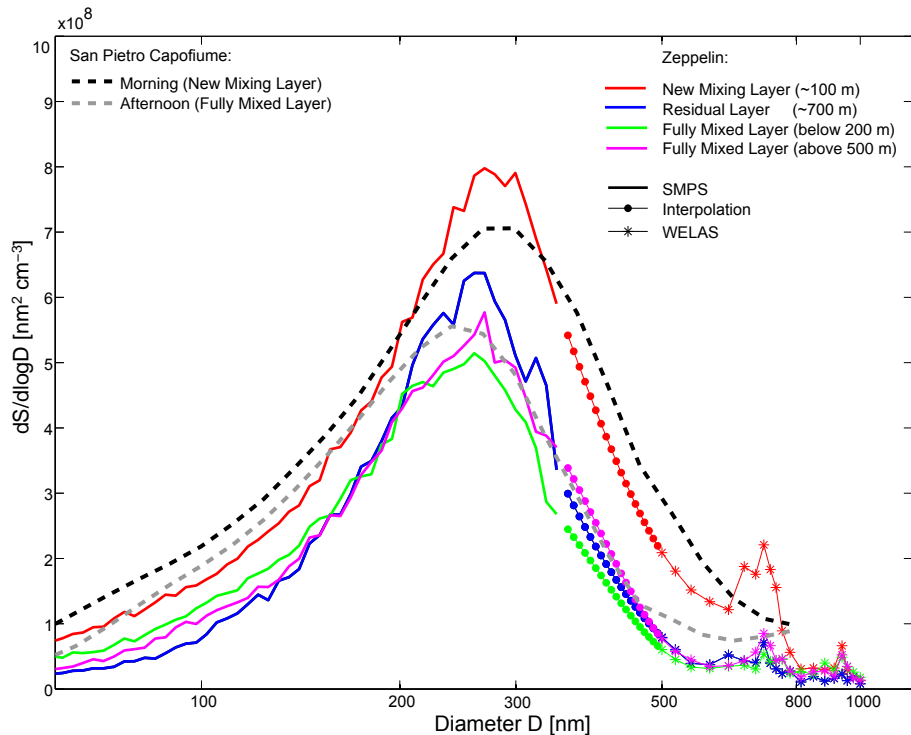


Figure 3. Dry surface area size distributions for different times and layers were probed during 20 June 2012. The colored lines denote the results measured on board the Zeppelin showing the contribution by the SMPS (straight line), WELAS (stars) and the interpolation in between (dotted line). During the early morning hours the new mixing layer (red line) and just above the residual layer (blue line) could be probed. Later the results are representative for the fully mixed layer (green and pink lines for different altitudes). The dashed lines were recorded at the SPC ground stations, where the black line shows the results for the morning hours (new mixing layer was probed) and the gray line for the early afternoon (fully developed mixed layer was probed).

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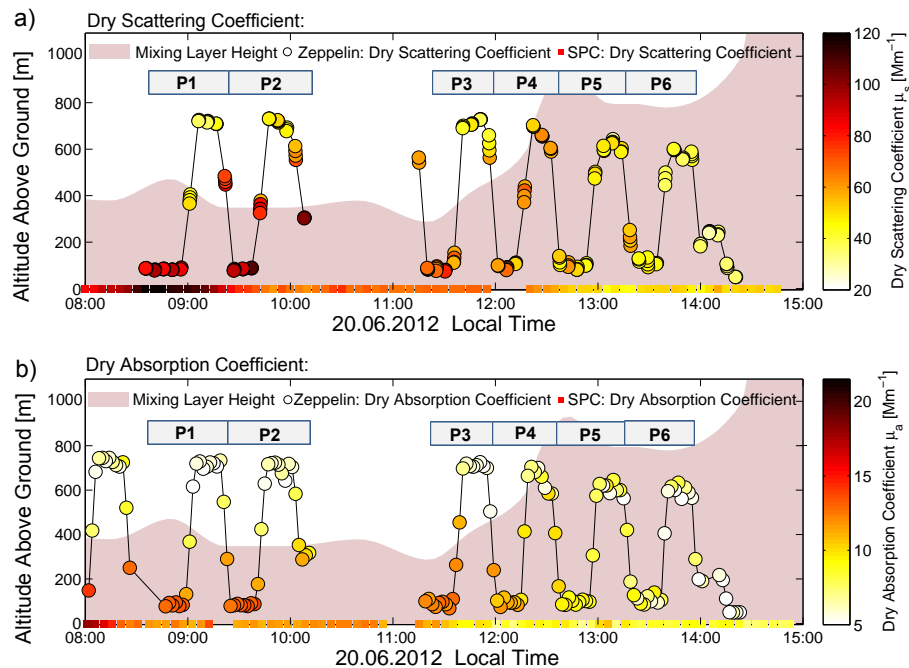


Figure 4. Time evolution of the dry scattering (a) and absorption (b) coefficients from the airborne and ground based platforms.

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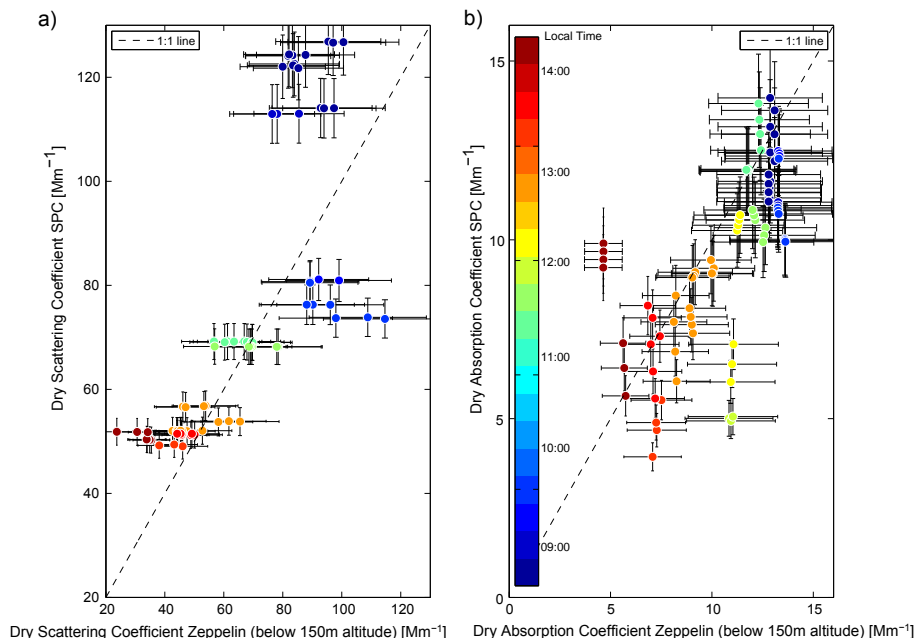


Figure 5. Comparison of dry scattering **(a)** and absorption **(b)** coefficients for ground-based and airborne measurements. The Zeppelin results were restricted to altitudes below 150 m in order to eliminate differences due to potential changes in atmospheric layers. The color of the symbols reflects the time of the day according to the color bar in **(b)**. **(a)** For the ground based data an uncertainty of 5 % is estimated while $\sim 18\%$ was found for the airborne calculations. **(b)** The uncertainty of the airborne absorption coefficients is estimated to be $\pm 20\%$, while $\pm 10\%$ is estimated for the ground based results.

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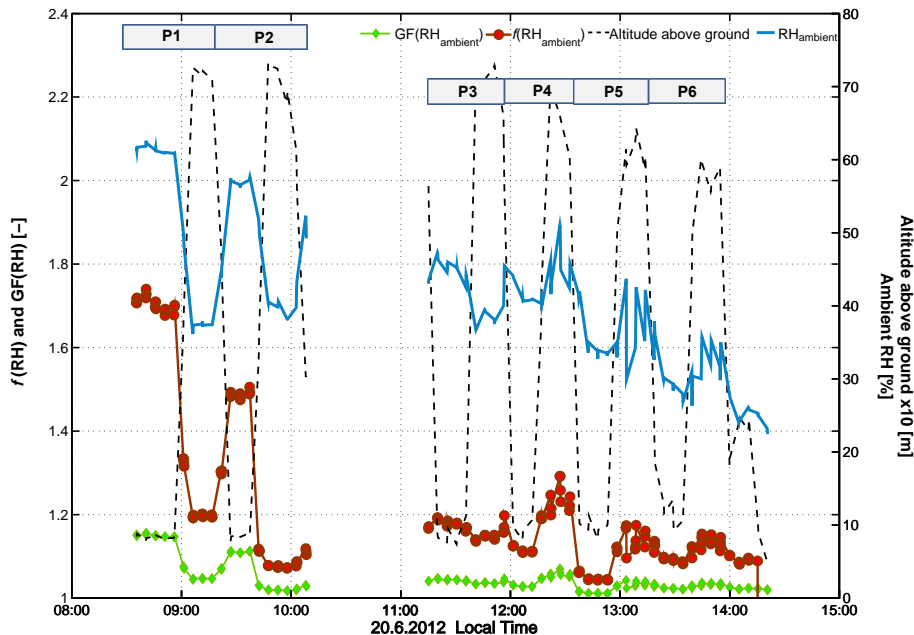


Figure 6. Time series of hygroscopic properties and ambient RH. The blue line illustrates the ambient RH present during the flight (right y axis). The green diamonds reflect the growth factor (GF) for the ambient RH calculated from GF(RH=95 %) measurements for 500 nm particles with the WHOPS. The red line shows the scattering enhancement factor ($f(\text{RH})$) during the flight as derived from the ambient RH and the GF measurements. The uncertainty of the airborne $f(\text{RH})$ amounts to approximately 30 %, while the GF values are expected to have uncertainties < 10 %.

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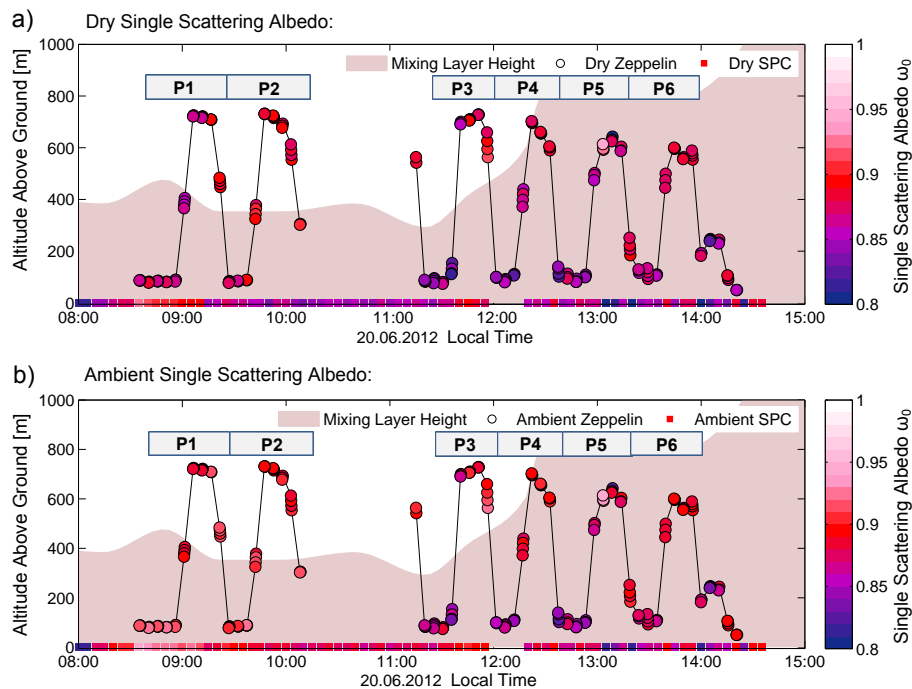


Figure 7. Time evolution of the dry (a) and ambient (b) single scattering albedo ω_0 from the airborne and ground based platforms.

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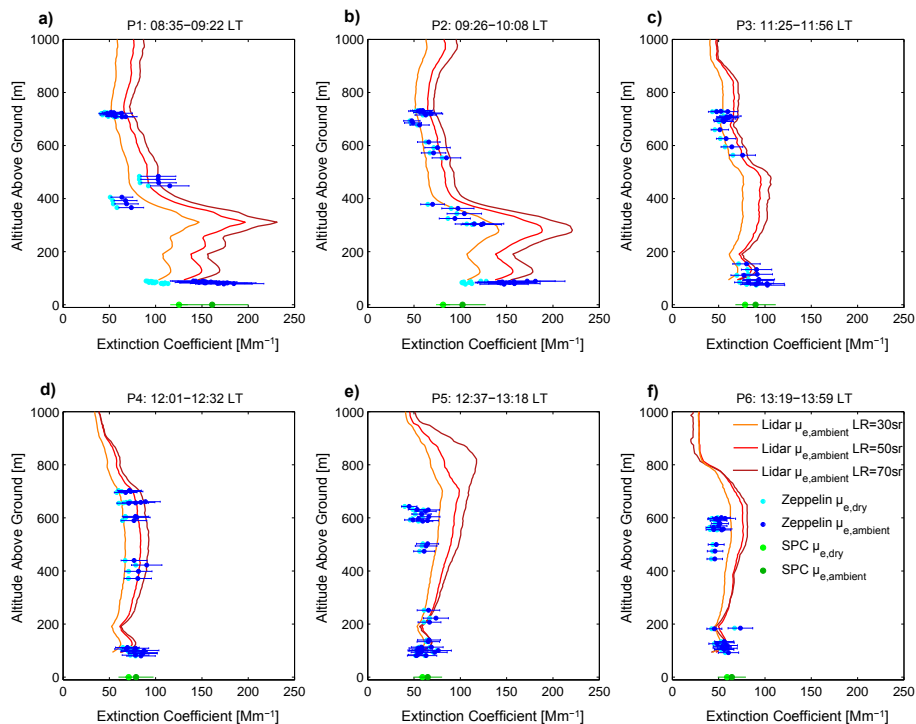


Figure 8. Extinction coefficients for profiles *P1–P6* at different times of day. The lines reflect LIDAR results for assumed LR of 30, 50 and 70 sr in orange, red and dark red, respectively. The dots describe in-situ results. In light and dark blue, dry and ambient airborne extinction coefficients are shown while the light and dark green dots represent the dry and ambient ground based results, respectively.