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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Hygroscopic growth study in the framework of EARLINET during the SLOPE I campaign: synergy of remote sensing and in-situ instrumentation" *by* Andrés E. Bedoya-Velásquez et al.

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Major comment:

In general, the paper presents an interesting study comparing direct measurements at different elevations with in-situ data at a nearby located mountain site. There is however a lack of information on the in-situ data.

We agree with the referee#2 that there is a lack of information on the in-situ instrumentation and its data treatment. Accordingly, we have extended the "Experimental

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site and instrumentation" section including a detailed description of the measurement set-up as follows:

p7, line 3 - 16 "At Sierra Nevada station (SNS, 37.09 N, 3.38° W, 2500 m a.s.l.), state-of-the-art in-situ instrumentation was operated to characterize aerosol properties. The inlet at SNS station is a whole air inlet located in the rooftop of a 3-story building. It is made up of stainless steel tube, with dimensions of 10 cm in diameter and 2.5 m in length. Inside the main tube there is a laminar flow of 100 litres per minute and there are several stainless steel pipes that drive the sampling air to the different instruments. Each one of the stainless steel pipes extracts the appropriate flow for each instrument. Different diameters of the pipes have been selected in order to optimize the efficiency of the system [Baron and Willeke, 2001]. The instrumentation used in this study includes an Aerodyne Aerosol Chemical Speciation Monitor (ACSM, Aerodyne Research Inc.), an Aethalometer (AE33 model, Magee Scientific, Aerosol d.o.o.), an Aerodynamic Particle Sizer (APS, TSI 3321) spectrometer and a Scanning Mobility Particle Sizer (SMPS, TSI 3938) spectrometer; all of them connected to the main inlet. The ACSM was used to measure on-line submicron inorganic (nitrate, sulphate and ammonium) and organic aerosol (OA) concentrations. Equivalent black carbon, eBC, mass concentration was obtained from measurements of the Aethalometer AE33 at 880 nm. A mass absorption cross section of 7.77 m2g-1 was used to convert the absorption coefficients at 880 nm in eBC mass concentrations (Drinovec et al., 2014). Particle number size distributions were retrieved by a combination of the measurements performed with the SMPS in the diameter range 13-600 nm and the APS for the range 0.6-20 μm."

Did the ACSM and Aethalometer have a specific inlet (PM10, PM1. . .)? Did they share an inlet? Yes, both instruments share the same inlet. It is a whole air inlet located in the rooftop of the building. For further details on the inlet and experimental setup see our comment above.

What was the mean or median size distribution during the selected case studies?

From the Fig. 1 R2 we can see the mean size distribution during the hygroscopic case investigated in this manuscript. The number size distribution has two main peaks at 35 and 115 nm.

Did the APS see particles above 2-3 μm, which is mentioned to be the upper limit of the Lidar?

From the mean size distribution (Figure 1 R2) during the hygroscopic case it can be observed that most of the aerosol is in the fine mode (< 1 μm). The upper limit of the lidar is not 2-3 μm, in the manuscript we stated that due to the lidar's wavelengths the instrument is more efficient at those wavelengths. Therefore, to avoid confusion, the following paragraph has been rephrased,

p14, line 7 - 9 "At SNS station, according to the mean size distribution calculated with SMPS during the hygroscopic growth case two main peaks were detected at 35 and 115 nm (from 20:00 to 21:00 UTC), therefore, it can be observed that most of the aerosol is in the fine mode (< 1 μm), but the lidar's wavelengths used are more efficient at those wavelengths."

Can dust particles be excluded during the case studies (which could lead to a lower backscatter coefficient but cannot be measured by either ACSM or Aethalometer)?

During this campaign, the chemical composition was measured with the ACSM and the Aethalometer. This set-up does not allow us to determine the mass concentration of dust particles neither to detect their presence. The air mass trajectories arriving at SNS during 16 June 2016 were coming from the North Atlantic Ocean and the atmospheric situation during this day and the previous days was characterized by clean conditions. Considering also additional data gathered during the campaign at SNS we can conclude that dust particles were not present during the case investigated here. In this sense, the scattering Angstrom Exponent (SAE) can be used as an indicator of the predominance of fine (SAE around 2) or coarse particles (SAE < 1). From the scattering coefficients measured with a nephelometer (TSI 3563) at SNS station, the

calculated SAE was > 1.2 during the hygroscopic case under study. In addition, AOD-derived Angstrom Exponent from AERONET Level 2.0 were above 1.4 during the whole day (16 June 2016). The fine mode fraction retrieved by AERONET was above 0.83 evidencing that coarse particles did not predominate.

It is mentioned that it is difficult to assess the uncertainties in the in-situ data, but can any upper or lower limits be estimated?

The uncertainty in the f (RH) calculation is a function of the individual measurement uncertainties in the particle number size distribution and in the chemical composition. Furthermore, the different assumptions made to calculate the backscattering coefficients within the Mie model contribute to this uncertainty as well. These assumptions are: Since no experimental measurements of the growth factor, g (RH), were available during the campaign, the g (RH) was calculated based on g(RH) from the literature for the individual chemical components measured with the ACSM+Aethalometer. The change in the size distribution with changing RH is calculated based on a constant and size-independent g (RH). The change in refractive index with changing RH is calculated from a volume-weighting of the aerosol dry refractive index and the refractive index of water, using the mean g (RH) (Hale and Querry, 1973).

This procedure has been extensively used and it has been proven to provide successful results in closure studies (e.g. Fierz-Schmidhauser et al., 2010ACP; Zieger et al., 2013). Concerning the uncertainties in the retrieval, Adam et al. (2012) calculated f (RH) using Mie theory and performed a sensitivity study concluding that errors in the backscattering enhancement factor can vary from 10 up to 30% as the RH changes from <50% to 90%. This information has been included in the revised manuscript.

The following information is now included in the manuscript: p9, line 11 - 17 “Estimations of ãĂŰ fäĂŮ_(β) (λ,RH) uncertainty are very scarce because of their high complexity. Some studies (e. g. Adam et al., 2012; Zieger et al., 2013) provided estimations based on sensitivity analysis using Mie model calculations, reporting er-

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rors around 20% f_σ (λ,RH). where σ is the scattering coefficient. Titos et al. (2016) reported uncertainty estimations based on Monte-Carlo techniques, concluding that the more hygroscopic the aerosol, the higher is the uncertainty in f_σ (λ,RH) especially at high RH (RH > 80%). For moderate-hygroscopic aerosol, it was established a lower limit for the uncertainty in f_σ (λ,RH) of around 30-40% using nephelometry techniques."

Also, the size at which the hygroscopic growth factors listed in Table 1 were measured should be mentioned and a short comment on the mean/median size measured during the cases in this study should be added.

We have included the diameters in the caption of Table 1 and a short comment on the mean size distribution, as suggested by the reviewer.

p14, line 6 – 9 "At SNS station, according to the mean size distribution calculated with SMPS during the hygroscopic growth case two main peaks were detected at 35 and 115 nm (from 20:00 to 21:00 UTC), therefore, it can be observed that most of the aerosols are in fine mode (< 1 μm), but the lidar's wavelengths used are more efficient at those wavelengths."

Additionally, the introduction would profit from some information on the used method to retrieve in-situ hygroscopicity values (more comments on this are presented later).

We agree with the referee#2. A detail response is given in the next comment.

p2, line 24 "Several studies have been carried out over the past years in order to evaluate how water uptake affects aerosol properties. One parameter used to quantify these changes is the so-called aerosol hygroscopic enhancement factor, f (λ,RH), defined as the ratio between aerosol optical/microphysical properties at wet atmospheric conditions and the corresponding reference value at dry conditions (Hänel 1976; Ferrare et al. 1998; Feingold et al., 2003; Veselovskii et al., 2009; Granados-Muñoz et al., 2015; Titos et al., 2014, 2016, and references therein). Most of the previous stud-

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ies investigating aerosol hygroscopicity are based on in-situ measurements. One of the most commonly used in-situ instruments for measuring aerosol hygroscopicity is the Humidified Tandem Differential Mobility Analyzer (HTDMA) (e.g. Swietlicki et al., 2008) that measures the hygroscopic growth factor g(RH) that quantifies the change in particle diameter due to water uptake. Humidified tandem nephelometers have been extensively used as well to quantify the effect of the hygroscopic growth in the aerosol optical properties, namely scattering, backscattering and extinction coefficients (e.g., Pilat and Charlson 1966; Titos et al., 2016). There are other in-situ instruments such as the white-light humidified optical particle spectrometer (WHOPS) (Rosatti et al., 2015) or the Differential Aerosol Sizing and Hygroscopicity Spectrometer Probe (DASH-SP), (Sorooshian et al., 2008) that have been used to determine the impact of enhance RH on the aerosol properties from airborne platforms. The effect of RH on the aerosol optical properties can be also determined with Mie model calculations (e.g. Adams et al., 2012; Fierz-Schmidhauser et al., 2010; Zieger et al., 2013) using the measured size distribution and chemical composition as inputs. For this calculation, the g (RH) is also needed as input. This factor can be determined experimentally (using HTDMA measurements for example) or it can be inferred from the individual growth factors of the different chemical compounds. The assumption of some aerosol properties such as the refractive index or the growth factor based on the chemical composition is the main drawback of this method. Generally speaking, one important limitation of most in-situ techniques is that they modify the ambient conditions and are also subject of particles losses in the sampling lines, therefore altering the real atmospheric aerosol properties."

General comments:

P2, line 30: Please make the section on commonly used hygroscopicity measurements clearer. The HTDMA has only been employed on the ground, whereas there are other instruments for airborne measurements (DASH-SP, WHOPS, AMS+Aethalometer...). Also add some information on the method used in this article and some pros and cons

as stated for the HTDMA.

According to the referee#2's suggestion we have clarified this. We have explicitly mentioned the different instruments used to investigate aerosol hygroscopicity from the ground and from airborne platforms but we have not extended the discussion much further since a review of techniques is not the focus of this manuscript. These lines have been replaced in the manuscript by. For detailed information see the comment above.

P6, line 6: Please add if any corrections were applied to the Aethalometer data and which MAC value was used to convert the absorption coefficient to the eBC concentrations (and reference).

No corrections were applied to the Aethalometer AE33 data and the MAC used was 7.77 m2g-1, as recommended by the manufacturer for this wavelength (Drinovec et al., 2014). This information has been added to the instrumentation section.

p7, line 13 – 16 "Equivalent black carbon, eBC, mass concentration was obtained from measurements of the Aethalometer AE33 at 880 nm. A mass absorption cross section of 7.77 m2g-1 was used to convert the absorption coefficients at 880 nm in eBC mass concentrations (Drinovec et al., 2015)"

Figures: Please use the descriptions of a) b) c) . . . in the text to refer to certain parts of the figures as this facilitates to follow the discussion. Also add legends to Figure 1 and mention what the γ values given in the legends are (the text states only "sold lines"). We agree with the referee#2. These changes have been performed in the manuscript.

Title of section 4.2.2: "measured and modelled f$\beta\lambda$(RH)" is a little misleading as no modelling (except HYSPLIT) was performed. Maybe rather use "measured f$\beta\lambda$(RH) and calculated using in-situ data and Mie theory" or something similar

Following the referee#2's suggestion, we have decided to modify the title as it is follow:

P13, line 29 "f_$\beta\hat{}\lambda$ (RH) measured and retrieved by combining in-situ data and Mie

theory”

Figure 5: The “measured” data exhibits some kind of jump at RH=95%. Can you comment on this and why, possibly, it is not seen in the Mie calculations with the in-situ data?

The jump seen on figure 5 on measured data, could be associated with the uncertainties on the RH calculation itself mostly above RH>95%, because of the combination of MWR temperature and calibrated WVMR signal. On this hand, the RH profile could have some data tendency to keeping constant meanwhile backscatter coefficient could remain increasing above RH=95 %, which may cause to the final calculation an increase of the uncertainty of f (RH).

Specific comments:

P3, line 26: change “on one hand” to “on the one hand” Done P4, line 5 and following: please rephrase the second part of the sentence with the case of RH>60%

We have rephrased it in the manuscript:

p5, line 6 - 8 “Navas-Guzmán et al. (2014) analyzed one year of measurements of RH profiles at Granada showing that RH values are low (below 60%) in the 75% of the cases studied within 1.0 and 2.0 km a.s.l. This study also showed that most of the cases with RH above 60% are found in spring and winter seasons.”

P4, line 23: please specify “incomplete overlap”

We have included the following sentence in the manuscript:

p5, line 29-30 “Atmospheric information retrieved from lower regions is limited by the full overlap height, which is reached above 1.3 km a.s.l due to the system configuration (Guerrero-Rascado et al., 2010; Navas-Guzmán et al., 2011)”

P6, line 6: change “werer” to “were” Done P7, line 18: which GDAS resolution (degrees) was used?

We have included in the manuscript:

p8, line 22 - 23 “GDAS meteorological data used have a spatial resolution of 0.5°x0.5° available since 2010 with daily files every three hours on the native GFS hybrid sigma coordinate system.”

P13, line 6: missing space between “similar” and “γ” Done P13, line 8: change to “in one of their case studies” Done P13, line 24: what does the 4% refer to exactly?

It means that taking as the reference f_βˆ355 and f_βˆ532 from Mie calculations and making the relative error calculation with measured values for each wavelength, this relative error remains below 5 %. The relative error for f_βˆ355=2.7 % and relative error for f_βˆ532=4.3 %. P13, line 29: change to “associated with. . .” Done P13, line 31: change to “reported in..” Done P14, line 22: use past tense i.e. “were” instead of “are” Done P14, line 28: change “it is concluded” to “we concluded” Done P15, line 5: change “gamma parameter” to “γ parameter” Done P15, line 6: please add a more precise description of what the 13% and 10% describe We have commit an error in the percentages, to be precise, we found good agreement on γ parameters measured (ãĂŰ γãĂŮˆ532=0.48$\pm$0.01 and γˆ355=0.40$\pm$0.01) respect to Mie calculated ones (ãĂŰ γãĂŮˆ532=0.53$\pm$0.02 and γˆ355=0.45$\pm$0.02), with relative differences between measured and calculated of 9 % at 532 nm and 11 % at 355 nm, taking the calculated values as reference. P15, line 7: change “those” to “these” Done P15, line 7-8: explain what “favorable” means; change “no-advection” to “in absence of advected air masses” or something similar Favorable refers to the good atmospheric conditions needed to evaluate these phenomena like vertical homogeneity (good mixed layers), same origin of the aerosols within the analyzed layer, low horizontal velocity and the absence of advected air masses into the evaluated column. P15, line 11: change to “making it possible” Done P15, line 12: change “those” to “these” or “such” Done

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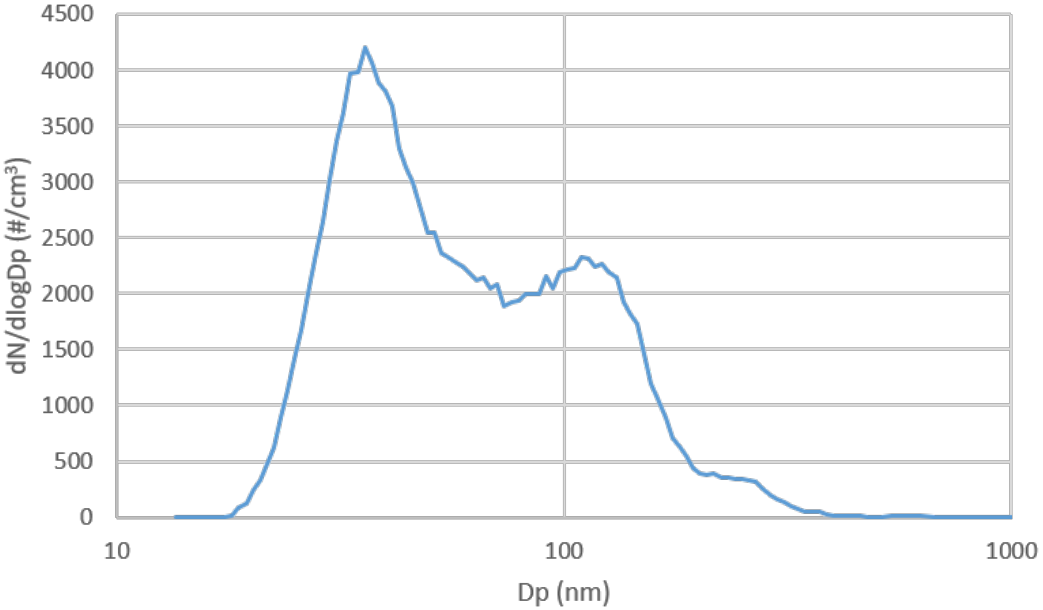

Fig. 1. Figure 1 R2. Mean particle number size distribution during the hygroscopic growth case on 16 June from 20:00 to 21:00 UTC.

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