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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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Author's response

We thank the anonymous reviewers for his/her comments and suggestions that have helped to improve the quality of the manuscript. According to the referees' reports, the following changes has been performed on the original manuscript and a point-by-point response is included below,

Answers to Referee#1:

Two major points to include in the manuscript: I. Take a modelled temperature profile, preferably from GDAS (Global Data Assimilation System), to compare your results obtained by the microwave radiometer (and the radiosonde). If it agrees well, this would extend tremendously the application of your method.

Following the reviewer#1's suggestion, Fig. 1R1 (a). presents a comparison among microwave radiometer (MWR), radiosounding (RS) and GDAS relative humidity data on 22th Jul 2013 at 20:00 UTC, selected as an example of one studied case in the manuscript. The following configuration is used in order to get a dataset comparable in time: a time-averaged profile from 20:00 to 21:00 UTC for MWR, GDAS output at 21:00 UTC, the RS launched at 20:00 UTC. This figure shows a negative bias for RHlidar+MWR within 0 to 10 % for almost all profile, instead the variability between RHlidar+GDAS is higher from -15 % to almost 20 % in the upper profile.

The disagreement between GDAS and RS profiles is mainly associated to two factors: (i) the complex terrain where the measurement station is located, surrounded by mountains of high elevation (up to more than 3000 m a.s.l in a very short horizontal distance of few tenths of kilometres) that makes more difficult for models to provide accurate thermodynamics profiles for this location; (ii) GDAS profiles have a lower temporal resolution (3 h) than the MWR one, which gives temperature profiles each 2 min and here is averaged up to 1 h. The combination of these two factors is the reason why we conducted our study in terms of MWR data, although we agree with the reviewer that the use of GDAS temperature profiles would extend the applicability of the methodology presented in this manuscript for locations with less complex orography.

Try to deliver extinction enhancement or even scattering enhancement factors as they are reported frequently in literature. Use all your information from the lidar and the in situ sampling to give at least an estimate. Just reporting the backscatter enhancement factor limits the outreach of your study.

We agree with the fact that the literature mostly reports values about scattering and extinction enhancement factors. It is due that most of hygroscopic studies have been addressed using in-situ instrumentation. The optical parameters that can be obtained from lidar measurements are backscatter (scatter at 180°) and extinction coefficients. Unfortunately, the extinction retrievals for the presented case are not available due to the low quality of the Raman-shifted lidar signals, and this is the reason why we have only focused on the backscatter coefficient. However, the outreach of our results is very important because most of the ground-based and satellite lidar observations provide only vertical information of the backscatter coefficient. Mayor comments

The key facts should be included in the abstract. At the moment, the abstract is too descriptive, and too few results are presented. For someone who only reads the abstract, the main findings should shortly be presented. Here or in the introduction, you should mention that the method is tested against the results of Granados-Muñoz, AMT 2015.

Following the recommendations of the referee#1, we have included in the abstract the following lines in blue colour:

"This study focuses on the study of aerosol hygroscopic growth during Sierra Nevada Lidar AerOsol Profiling Experiment (SLOPE I) campaign by using the synergy of active and passive remote sensors at Granada valley station and in-situ instrumentation at a mountain station (Sierra Nevada). To this end, a methodology based on the combination of calibrated water vapour mixing ratio (r) profiles, retrieved from an EARLINET multiwavelength Raman lidar (RL), and continuous temperature profiles from a microwave radiometer (MWR) for obtaining relative humidity (RH) profiles with high temporal resolution is used. This methodology is validated against an approach using radiosounding (RS) data, obtaining differences in hygroscopic growth parameter (γ) lower than 5% between the methodology based on RS and that based on remote sensing. During SLOPE I the remote sensing methodology used for aerosol hygroscopic growth studies has been checked against Mie calculations of hygroscopic growth using in-situ measurements of particle number size distribution measured at SNS. The hygroscopic case observed during SLOPE I showed an increase in particle backscat-

ter coefficient at 355 and 532 nm with the relative humidity (with RH ranging between 78-98%), but also a decrease in backscatter-related Ångström exponent (AE) and particle linear depolarization ratio (PLDR) indicating that the particle became larger and more spherical due to hygroscopic processes.Vertical and horizontal wind analysis is performed by means of a co-located Doppler lidar system at IISTA-CEAMA station, in order to evaluate the horizontal and vertical dynamics of the air masses. Finally, the Hänel parameterization is applied to experimental data for both stations and we found good agreement on γ parameters measured (ãĂŰ γãĂŮˆ(532)=0.48±0.01 and ãĂŰγ ãĂŮˆ355=0.40±0.01) respect to calculated (ãĂŰ γ ãĂŮˆ532=0.53±0.02 and γˆ(355)=0.45±0.02), with relative differences between measured and calculated up to 9 % at 532 nm and 11 % at 355 nm."

The introduction is not well written. The main structure of an introduction should be clearer: Why is it important? What has been done in this field? What questions remain open? What is your contribution?

Discussion paper Furthermore, it is not clear, which aerosols are observed and how the publication is structured. Recent literature from the studied field should appear in the introduction. Some references are given later in the manuscript, but they could already appear in the introduction. Please revise carefully the literature.

Following the reviewer#1's suggestions 2 and 3, the introduction has been restructured as follows

"Atmospheric aerosol particles play a crucial role in the Earth′s climate, principally by means of the radiative effect due to aerosol-radiation and aerosol-cloud interactions, affecting the Earth-atmosphere energy balance and, hence, the Earth's climate. Furthermore, the aerosol might also modify optical and microphysical cloud properties, such as albedo and cloud droplet size distribution that influences cloud lifetime, since the particles could act as cloud condensation nuclei (CCN) and ice nuclei (IN) (Twomey, 1977; Albrecht, 1989; IPCC, 2013).

Water vapor plays a major role in the aerosol-radiation interaction due to the ability of some atmospheric aerosol particles to take up water from the environment. In this sense, hygroscopic growth is the process by which aerosol particles uptake water and increase their size under high relative humidity (RH) conditions (Hänel, 1976). Consequently, this process is also related to changes in the optical and microphysical properties of the aerosol particles and, hence, it becomes a crucial factor that modifies the role of aerosols in atmospheric processes and radiative forcing.

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 studies 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. In general terms, 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.

Remote sensing systems such as lidars have also been used in the last decades for aerosol hygroscopic growth studies performed with co-located RS measurements (e. g. Ferrare et al., 1998; Feingold et al., 2003; Veselovskii et al., 2009; Granados-Muñoz et al., 2015; Fernández et al., 2015; Lv et al., 2017). These systems have shown to be robust with high vertical and temporal resolution that allow for studying the aerosol hygroscopic growth under unmodified ambient conditions. This aim has also been studied by Zieger et at. (2011), showing the capability to combine Raman lidar, in-situ and MAX-DOAS instrumentation for study hygroscopic growth in ambient conditions extrapolating extinction coefficient from lidar to the ground studies. Also, some studies have been performed by using Automatic Lidar and Ceilometers (ALC) for hygroscopic and fog studies mostly for forecasting purposes of fog events, through the combination of attenuated backscatter with in-situ data from instrumented towers which reach almost 200 m above ground level (Haeffelin et al., 2016). In this work we are adding a comparison between ground city station to high mountain station in order to connect effects of the city over mountain and also avoid technical issues like lidar overlap.

Up to now, most hygroscopic growth using lidar systems combine the lidar measurements with RH data from RS. The main inconveniences are that RS measurements have low temporal sampling and they could be drifted away from the vertical atmosphere probed by the lidar systems. These inconveniences can be easily overcome by combining calibrated water vapor mixing ratio profiles, r (z), from Raman lidar (RL), with temperature profiles from ancillary instrumentation for obtaining RH and aerosol backscatter profiles, using them simultaneously for hygroscopic growth studies (e.g. Whiteman, 2003; Navas-Guzmán et al., 2014; Barrera-Verdejo et al., 2016). Navas-Guzmán et al. (2014) proposed a methodology for retrieving RH profiles by the combination of calibrated r (z) profiles from Raman lidar water vapor channel with temperature profiles obtained from microwave radiometer (MWR) measurements. RH profiles obtained using this approach and aerosol profiles from the lidar are used in this work to study aerosol hygroscopic growth. This methodology allows for obtaining a wider database for the analysis of the aerosol hygroscopic growth properties using remote sensors since some of the limitations associated to RS are overcome. Additionally, water vapour and aerosol measurements are performed with the same system and, thus, the same air volume is probed, avoiding the radiosonde drift and temporal mismatching sampling.

The main goal of this study is to apply the methodology proposed by Navas-Guzmán et al. (2014), based on the application of the synergy between RL and MWR for aerosol hygroscopic growth studies. First, this methodology for hygroscopic growth studies is compared with the approach presented in Granados-Muñoz et al. (2015) that uses RS and lidar data. Once the technique is validated, a study of the aerosol hygroscopic growth case observed during the SLOPE I (the Sierra Nevada Lidar AerOsol Profiling Experiment I) campaign is presented. The results obtained with remote sensing are compared with Mie simulations performed using in-situ measurements from a high-mountain station (up to 2500 m a.s.l).

This paper is organized as follows. Description of the experimental site and instrumentation are presented in Section 2. The methodology applied is introduced in Section 3. Section 4 presents the results and discussion of the combination of RL and MWR method for obtaining RH profiles and also the hygroscopic case analysed by combining measured and retrieved data from lidar and in-situ instrumentation, respectively. Finally, conclusions are given in Section 5."

In general, the reader is interested in which aerosols you are observing. Very late, in Table 1, you give an overview. Do you consider it as continental aerosol, urban pollution or biogenic particles? Please add a discussion about the aerosol type. In GranadosMuñoz, AMT 2015, sulfate, marine and mineral dust particles are discussed.

In order to improve the discussion about aerosol type we have added the following paragraph.

P14, line 14 -18 "Thus, the predominant aerosol studied during the event is a composition of smoke and urban polluted aerosol. This assumption about the aerosol type is supported by the relatively high sulphate concentration observed at SNS and the results discussed in section 4.2.1 (lidar properties, backward trajectories analyses). The backscatter-related AE values (close to 1.5) indicates a predominance of fine particles which is connected with the presence of smoke over that comes from North America as suggested by the backward trajectories analyses."

Section 2 is not well structured: first a description of the campaign (now Sect. 2.2), then a sub section to the valley station in Granada (now Sect. 2.1) and then a sub section to the hill station with the in-situ instrumentation, where you explain which instrument measures which quantity.

We have re-structured this section of the manuscript following the reviewer instructions, as follows: 2.1 SLOPE I field campaign 2.2 IISTA-CEAMA station 2.3 Sierra Nevada Station

A sketch of the location would be nice (maybe a vertical cut, showing the orographic profile with valley and hill station and the distance between them). And you mention several in-situ stations at different heights, an interesting fact, that is not used later on.

We agree with the reviewer and therefore we have added a new figure with the orographic profile showing the location of the IISTA-CEAMA station (located in the valley) and SNS station (located in the slope of Sierra Nevada, at 2500 m asl). During this campaign, only one in-situ station was operative so we have modified the manuscript accordingly.

It is great to have a station on a mountain, almost 2000 m above the lidar and several stations on the mountain slope. Somewhere you should mention hygroscopic studies which compared remote sensing measurements with (meteorological) tower based in situ instrumentation, which only reach up to approximately 200 m above ground or the use of horizontal pointing remote sensing instruments to ground-based in situ observations; and the advantage of having a mountain slope for performing such experiments.

We have added some information in the introduction about works related with aerosol hygroscopic growth and fog detection by using ceilometer attenuated backscatter data combined with instrumented in-situ tower. These works have centred their attention on forecasting, but also, they open new possibilities for low heights aerosol hygroscopicity growth studies. Also, some studies performed by synergy of Raman lidar, in-situ and MAX-DOAS instrumentation making possible a good extrapolation the extinction coefficient to the ground:

p3, line 17-23 "This aim has also been studied by Zieger et at. (2011), showing the capability to combine Raman lidar, in-situ and MAX-DOAS instrumentation for study hygroscopic growth in ambient conditions extrapolating extinction coefficient from lidar to the ground studies. Also, some studies have been performed by using Automatic Lidar and Ceilometers (ALC) for hygroscopic and fog studies mostly for forecasting purposes of fog events, through the combination of attenuated backscatter with in-situ data from instrumented towers which reach almost 200 m above ground level (Haeffelin et al., 2016). These works are focused in enhancing the possibility to study aerosol hygroscopicity growth at low levels in ambient conditions, but in this work, we are adding a comparison between ground city station to high mountain station in order to connect effects of the city over mountain and also avoid technical issues like lidar overlap."

Another key point, you retrieve backscatter enhancement factors and you state, that it

is difficult to compare them to values found in the literature. With your Raman signals, you can retrieve the extinction coefficient and determine the extinction enhancement factor or at least the lidar ratio. For the extinction enhancement factor, there are much more literature values to compare. Eventually, your in-situ measurements allow you to determine the single scattering albedo and then you can derive the scattering enhancement factor, which can be compared to results obtained by in situ observations of hygroscopic growth. These conversions would add a lot of value to the paper.

As we explain above, unfortunately the extinction retrievals for the presented case are not available due to the low quality of the Raman-shifted lidar signals, and this is the reason why we cannot perform the calculations proposed by referee#1, because of that we are not including the in-situ single scattering albedo to derive scattering enhancement factor. In this work we have only focused on the backscatter coefficient, but the outreach of our results is very important because most of the ground-based and satellite lidar observations provide only vertical information of the backscatter coefficient.

As I understand it right, one idea of the paper is to perform hygroscopic growth studies using a calibrated Raman lidar system without having a radiosonde available. The Raman lidar delivers the aerosol properties and water vapor mixing ratio (if the calibration constant is known). In order to derive the relative humidity, the temperature profile is needed, which you derive from the microwave radiometer. Another option would be to use the temperature profile of the GDAS model output, as there are all available radiosonde ascends are included. I would like to see, how this even easier method compares to your results.

As we shown on Fig. 1R1, we have compared MWR and GDAS temperature data profiles with RS temperature profiles, so the results were not good enough. The main argument is that Granada is located in a very complex terrain because of the Sierra Nevada, therefore GDAS meteorological data doesn't fit enough good and also the temporal resolution of the GDAS data (3h) is quite lower compared with MWR. The Fig. 3R1, shows the calculation of f (RH) by using temperature from MWR (lidar +

MWR) and temperature from GDAS (lidar + GDAS), in order to show as an example that relative error between γ_RS and γ_MWR is lower than 4 % instead γ_RS and γ_GDAS is lower than 30 %, taking γ_RS=0.99 as theoretical value.

10. p4, l6-12 What about marine aerosol in Granada?

We have added the following paragraph explaining the conditions with marine aerosols in Granada

p5, line 14 - 19 "The probability of marine particles to reach the city is low taking into account that Granada is far away from the coast about 50 km in straight line, the marine particles would have to overpass some mountains in the path from the sea to the city and the air masses monitored over Granada are really dry. Also, Titos et al. (2014) showed that the contribution of marine aerosols to PM10 mass concentration was almost negligible (<3%) at IISTA-CEAMA station during the period 2006-2010. In addition, this work also refers to the identification of fine (PM1) and coarse (PM10) particulate matter in an urban environment of Granada".

11. p6, l25 – p7, l3 At which temporal resolution do you derive the temperature profiles and the RH profiles?

We have added the following information on the manuscript:

p8, line 1 -3 'Temperature profiles from the MWR, which are continuously measured every 2 min, combined with 30 min- averaged r (z) profiles as proposed by Navas-Guzmán et al. (2014), are used to retrieve the RH profiles required for aerosol hygroscopic growth studies each 30 min. The following equation is used for retrieve the RH profiles'

12. p12, l6-21 and Fig. 4 Why the third moment of the vertical velocity and not the vertical wind velocity is shown? The vertical velocity would give valuable information about updrafts and downdrafts.

The third moment of the vertical velocity (skewness) provide us a detailed informa-

tion about the aerosols dynamic. It is directly associated with Turbulent Kinetic Energy equation, providing us information about the direction of convective movements, its intensity, and consequently if there is a predominance of updrafts or downdrafts (Moreira, et al., 2018a). Therefore, the observation of this moment together with the direction and speed of horizontal wind can provide us a detailed information about aerosol origin and density in the chosen period.

13. p14, l1-4 The statement is not convincing and needs more explanation. You can use a particle size distribution from the mountain station to show the influence of the large particles and how frequent they are. Furthermore, you can use the 1064 nm backscatter to be more sensitive for the large particles.

According to the mean size distribution (Figure 4 R1) during the hygroscopic case it can be observed that most of the aerosol particles are in the fine mode (< 1 μm), but we stated that due to the lidar's wavelengths the instrument is more efficient at those wavelengths. 14. Why don't you use the backscatter at 1064 nm? In Fig. 3 and 5 you can extend your study to include the near infrared. It would add value to your publication.

We completely agree with referee#1, however the 1064 nm channel was no operational during this phase of the campaign.

Minor comments:

1. Comma instead of dot in the list of affiliations (6 times)

Done 2. The space after a symbol or a bracket is often missing throughout the manuscript. Done

3. Indices should not be written in italics, except in the sum formula (Eq. (4))

Done

4. Units should not be written in italics.

Done

5. Maybe you should consider slightly reducing the number of abbreviations to make the paper easier to read.

Done

6. “upward wind”, better “upward wind velocity” throughout the manuscript.

Done

7. In recent years the term ice nucleating particles (INP) is used for the aerosols which nucleate ice, see Vali et al., ACP 2015 (just for your information). Vali, G., DeMott, P. J., Möhler, O., and Whale, T. F.: Technical Note: A proposal for ice nucleation terminology, Atmos. Chem. Phys., 15, 10263-10270, https://doi.org/10.5194/acp-15-10263-2015, 2015. 8. Done

9. p4, l16-19 The instrument description is confusing, better “It emits laser pulses at . . ., and it receives backscattered photons at . . . in . . . mode”

We have added on the manuscript: p5, line 23-26 “It emits laser pulses at 355 and 532 nm (parallel and perpendicular polarization channels) and 1064 nm and it receives backscattered photons at 355, 532 and 1064 nm in analog and photon counting modes. Also, it collects Raman backscattered photons at 607 and 387 nm for molecular nitrogen (N2) and at 408 nm for water vapor (H2O) in photon counting mode”

10. p4, l21-22 What is the approximate overlap height of the system?

We have added this information on 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)”

10. p9, Eq 4, What about rho? It is the density of which part? In Eq 4, rho is the total density of the aerosol, now included on the manuscript. 11. p9, l31 and Fig. 1 Where

does the lidar ratio of 65 sr come from? This lidar ratio was obtained in Granados-Muñoz et al., 2015, so we have used the same values retrieved in this work in order to be comparable. 12. p11, l26 “fine/coarse predominance” better “size” Done 13. p11, l28 “predominance of coarse particles”, better “predominance of larger particles” Done 14. p13, l26-27 Please indicate the uncertainty ranges for the 4 derived backscatter enhancement factors as it is done in the conclusion. Done 15. p14, l4 Please repeat the horizontal distance at this point. Done 16. Tab. 3 In the caption, be consistent with the date: 16th June 2016 Done 17. Fig. 1+3 units should not be written in italics, see beta (. . .) We have reviewed text in the Fig. 1+3, but we have write these symbols in latex format so beta looks like italic but it is the format itself. 18. Fig. 1 d+h It is difficult to separate points and lines. Done 19. Fig. 2 Height range up to 4 or 6 km is sufficient to show. Could you please state (in the caption) the time of sunset as additional information? Done 20. Fig. 3 in the caption: backscatter at 1064 nm is not shown but mentioned. Done 21. Fig. 4 It would be better to just show the same time interval as in Fig. 2 (1700-0000 UTC) to increase the number of details. Done 22. References: - Kotchenruther et al., 1999 (not 1998) - List, 1951, strange “f&” - p13,l 17 no Titos et al. (2014b) only Titos et al. (2014) Done

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Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-993, 2017.

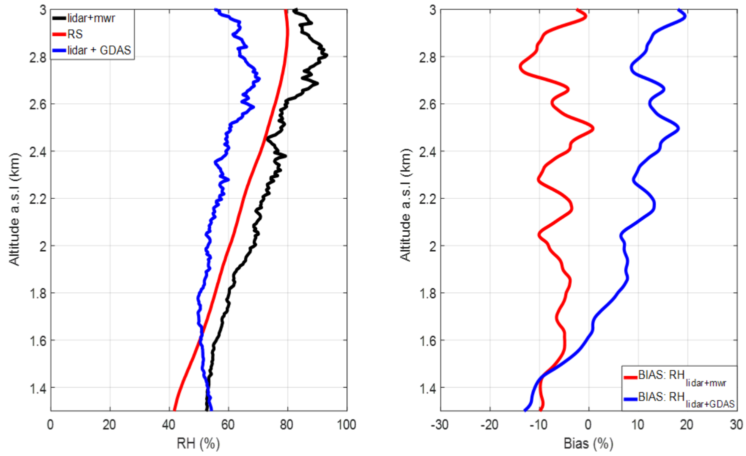

Fig. 1. Figure 1 R1. RH comparison for 22th Jul 2013 around 20:00-21_00 UTC. (a) RH profiles retrieved from combination of lidar+MWR (black line), lidar+GDAS (blue line) and RS (red line) and (b) Bias calcula

Fig. 2. Figure 2 R1. Vertical profile of SNS. The yellow star refers to IISTA-GRANADA station and green star refers to SNS.

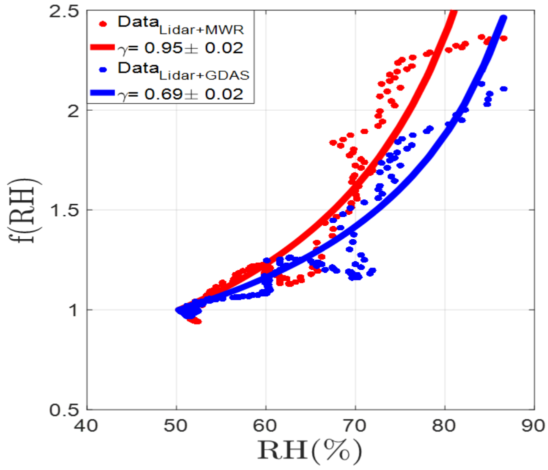

Fig. 3. Figure 3 R1. Backscatter enhancement factor retrieved for 22th July 2013. In red lines/dots is shown Lidar + MWR and blue lines/dots shown Lidar + GDAS calculations.

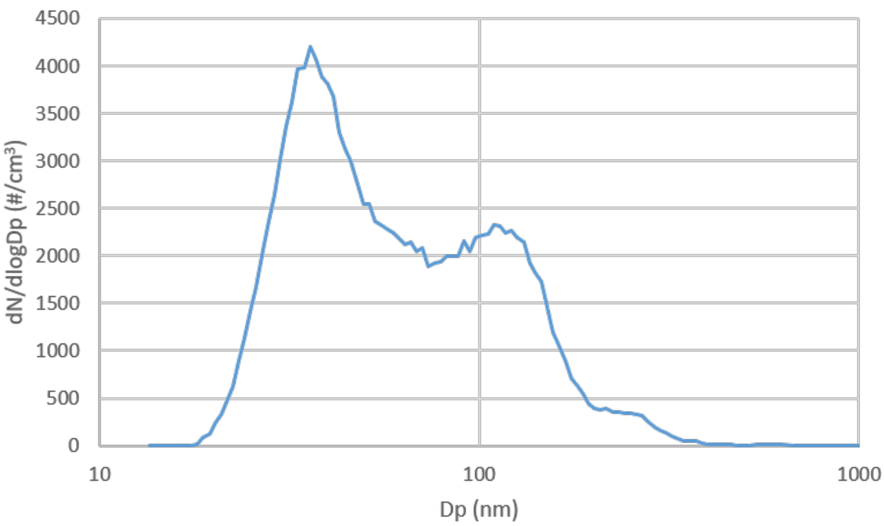

Fig. 4. Figure 4 R1. Mean particle number size distribution during the hygroscopic growth case on 16 June from 20:00 to 21:00 UTC.