Response to Reviewer#2

We thank the reviewer for his or her helpful comments and insight. We respond to the general and specific points below. All the comments are addressed in the revised manuscript. As requested, the whole text was proofread and edited, eliminate the typos and to improve the language.

General Comment 1: This manuscript describes vertical profiles of aerosol number density over Arctic during spring and summer and presents authentic and original scientific material that has relevant implications for atmospheric science (aerosol, clouds, CCN, and others). This study is based on very important aerosol data over Ny-Ålesund, Svalbard, although the tethered-balloon-borne aerosol measurements are restricted to the good weather (i.e., clear sky, calm winds etc.). On the whole, the topic of the manuscript is relevant and suitable for the scope of the "ACP". However, there are several points which require some careful revision and corrections before publication.

Answer to the General Comment 1 (AGC)1: Thank you very much for your comment which underline the originality and the high relevance of the results presented in our paper. Concerning your request of revision and correction we managed the paper accordingly to your suggestions (here below answered). The whole text was also proofread and edited, to eliminate the typos and to improve the language.

General comment 2. Quality of English I found many typo, miss-spell, and grammatical errors (e.g., location of ",").

AGC2: The manuscript was proofread and edited, to eliminate the typos and to improve the language as required.

General comment 3. Comparison with previous airborne aerosol measurements Several airborne aerosol measurements have carried out in Arctic area since 2000, for instances, ASTAR (2000, 2004, and 2007), ARCTAS (2008), ARCPAC (2008), and PAMARCMiP (2009 and 2011). Particularly, ASTAR campaigns were made around Svalbard. I suggest strongly that your data are compared to these previous results, and that these campaigns should be added into description of introduction.

*AGC3: Thank you for this comment. An important comparison was reported in the manuscript, at page 15 (lines 10-14), where "the columnar averages of both total aerosol number and BC concentrations [236.1±23.9 cm^{-3} (N_{14-260}), 21.1±1.3 cm^{-3} ($N_{260-1200}$), 0.2±4*10^{-2} cm^{-3} ($N_{>1200}$) and 52±8 ng m^{-3} (BC)]" were successfully compared with long-term data series collected over Ny-Ålesund at the Zeppelin observatory (Eleftheriadis et al., 2009; Tunved et al., 2013) during Spring. Moreover, at page 3 (lines 23-25) we cited some of the Arctic campaigns (i.e. Kupiszewski et al., 2013; Schwarz et al., 2010).*
However, we fully agree with you that a better contextualization of the measuring campaign with respect to the international is required. Thus, as also required by reviewer#1, we modified the introduction section adding and discussing the suggested campaigns. Moreover, as you suggested, we discussed the obtained results with respect to the same references.

General comment 4. Classification of aerosol type In this study, authors classified aerosol profiles into four groups. I agree with the classification of aerosol profiles. Unfortunately, typical weather/meteorological conditions and air mass origins in each type were not mentioned in the text. These information is very important to characterize vertical features of aerosols in Arctic region, and to be compared to aerosol data taken in the other project.

<u>AGC4:</u> Thank you for your comment, which supports the classification of aerosol profiles.
The weather and meteorological conditions for each profile class were addressed in Table 2, Figure 7 and discussed along section 3.3 and 3.4. However, they were mainly addressed to illustrate the differences between DNG profiles and the other profile classes. We modified the paper introducing the meteorological context for each profiles class.
The air mass origins are addressed below and we refer to the following explanation also for the answer to your specific comment 9.
We agree with you that air mass origin was shown and explained (in the manuscript) only for the case study reported in figure 6 and discussed in sections 3.3.2 and 3.3.3 for both PG and NG profiles.
However, before trying to find a relationship between the type of the vertical profile and air mass origin, it is necessary to consider that each profile shape is the result of an interplay among several processes: 1) transport events, 2) the planetary boundary layer dynamic and 3) the local formation of aerosol. Among this, only the transport event process is strongly related to the air mass origin (some precursors transported may also affect secondary aerosol formation), while the final profile shape is the result of the specific combination of the aforementioned processes.
Figure 6 represents a good example in which the same air mass origin that transported polluted air masses from mid-latitudes generated initially PG profiles that naturally evolved (due to the entrance into the PBL) into NG profiles.
Thus, the same air mass origin could be related to different profile classes.
Due to your question, even with the aforementioned limitations, we performed a cluster analysis of backtrajetories corresponding to each profile class obtained at using the Hysplit 4 (rev. 513). The result is attached here below (Figure AGC4.1).

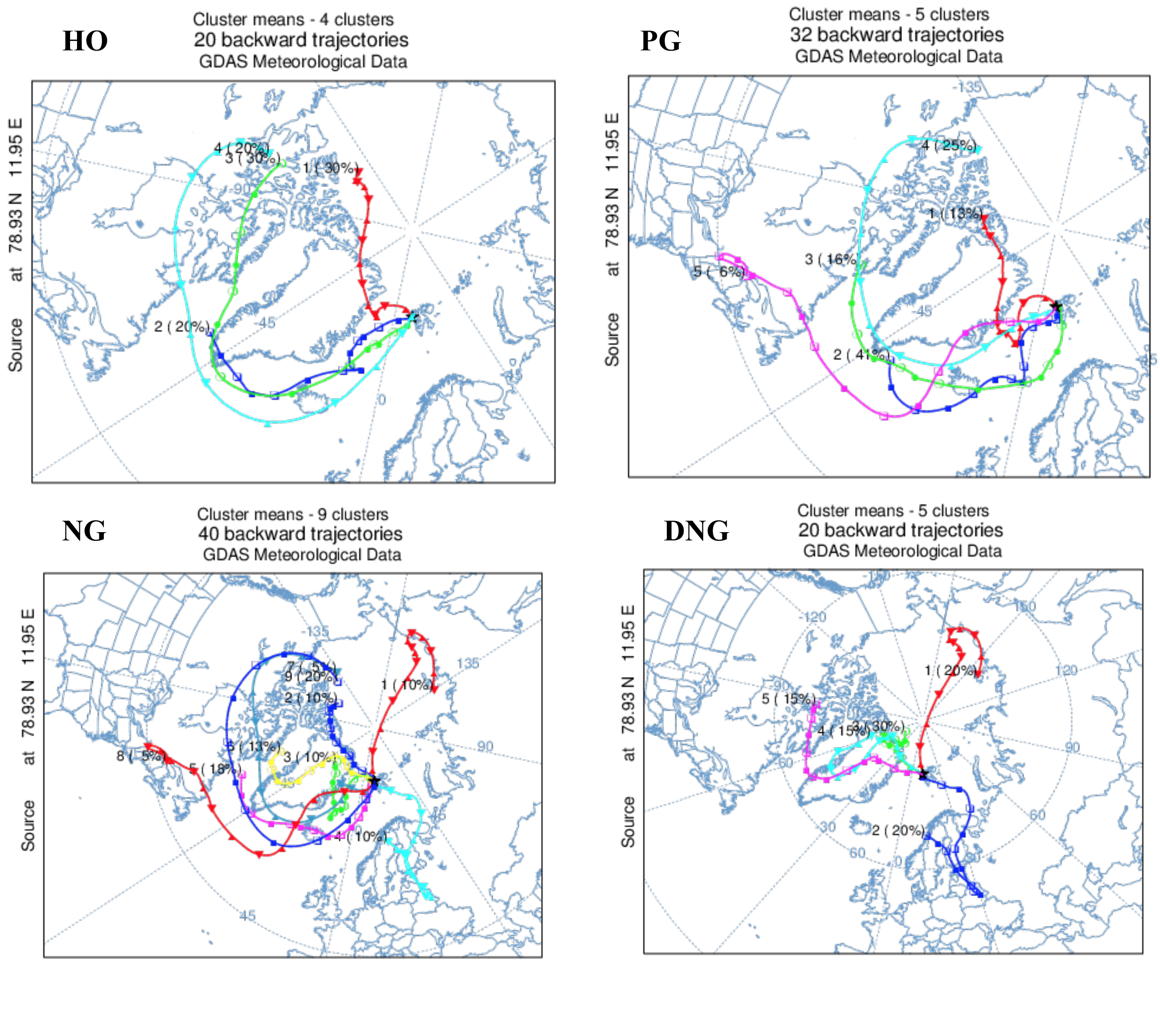

Figure AGC4.1. Cluster analysis of backtrajectories for each profile class.

From Figure AGC4.1 it is possible to observe that both PG and NG can be affected by transport from mid-latitudes. This can happen also for DNG profiles, which are special type of NG profiles. We added this figure to the supplemental material and we address the topic in the revised version of the manuscript.

General comment 5. Relation between aerosol vertical profiles and structure of boundary layer Vertical features of aerosols in the lower troposphere are associated with the structure of boundary layer (i.e., surface inversion and height of boundary layer). What is typical height of top of boundary layer in each type? Aerosol data should be compared to vertical structure of boundary layer (surface inversion and top of the layer).

AGC5: We fully agree with you that the relationship between the aerosol vertical profiles and structure of boundary layer is very important. In fact, sections 3.1-3.2 and figures 3-4 addressed this topic. Figure 3 illustrated the discussion of the case studies chosen as representative for each profile class. The relationship between θ inversions and AS_h was well documented in section 3.2. Figure 4 reports the frequency distribution with altitude of both AS_h and θ and RH gradient and section 3.2 is essentially dedicated to the discussion of figure 4.
What is missing, and we agree with on this point, is the average values of AS_h corresponding to H_s=0 for the profile classes reported in figures 5 and 12; they are: 417±266 m (PG), 506±212 m (NG), 585±90 m (DNG), 474±204 m (SP). We added these values to the sections in which each profile class was discussed.
Moreover, as also you requested in your specific comment 6 (see ASC6), we added averaged profiles of normalized θ and RH to Figure 5 in order to relate the mean vertical behavior of aerosol to that of meteorological parameters.

Specific Comment 1 (SC1): Abstract: Height range of aerosol measurements are added in the text of abstract.

Answer to the Specific Comment 1 (ASC)1: Thank you very much, we modified the abstract accordingly to your suggestion.

SC2: Page 5 Line 11, Unit of conductivity: M Ω cm-1 (not M Ω cm)

*ASC2: The measuring unit is right (*MΩ cm*) as it is the resistivity of the ultrapure (Milli-Q) water. We added to the text the specification: "resistivity".*

SC3: Page 9 – 10, Sensitivity (detection limit) of BC measurement. In general, high flow rate is required for BC measurements in regions with lower BC concentration. Flow rate for BC measurement was 2.5 X 10-6 m3 s-1 (0.15 L min-1) in this study. I understand that authors chose the largest flow rate of the micro-aethalometer. However, this flow rate might be not enough in lower BC concentrations. Although BC concentration increases during winter –spring in the Arctic regions, the BC level is lower than that in the mid-latitudes. What is the sensitivity (detection limit) of BC measurement in your measurement setting and analytical procedures?

ASC3: At page 10, lines 3-7, a first determination of the accuracy of equivalent BC (eBC) determination was reported by comparing the AE51 and the AE5x prototype measurements carried out simultaneously during spring 2011. The result of this intercomparison is reported in Figure 2c; as shown, eBC measurements obtained by the two micro-Aethalometers agreed very well

(R^2=0.852; slope=0.976) with a RMSE of 2 ng m^{-3} (considering the average of the two measurements as the target value).
This result is important because the AE5x prototype operated at 265 ml/min ($4.42 \cdot 10^{-6}$ m^3 sec^{-1}) and the AE51 at 150 ml/min (AE51 commercial version).
We present below the analysis calculating the error (in percentage) of each eBC data considering the average of the two eBC measurements (AE51 and AE5x) as the target value (as reported at Page 10, line 5). Figure ASC3.1 reports the mean (± standard deviation) and the 90^{th} percentile of the absolute value of the error in percentage of the measured eBC as the function of eBC concentration using intervals of 5 ng m^{-3}. As it possible to observe at low concentration the error can exceptionally reach values of 90%. The error decreases with increasing eBC concentration, dropping below 20% for concentrations above 20 ng m^{-3}. Thus, it is possible to consider this value as the limit above which a single BC measurement point is not affected by instrumental noise.
Even though this limit for a single eBC measurement is close to the BC concentrations that have been previously measured in the Arctic (Eleftheriadis et al., 2009), the eBC profiles presented in the manuscript are an average of many measurements, hence the effect of the noise on the reported eBC concentrations is further reduced. The aim of this paper is to determine the seasonal phenomenology of the aerosol behavior along vertical profiles classifying the collected experimental data, according to their shape and averaging them for each season. This is very important as, even the error in percentage of each data point can reach high values (especially at low concentrations), the average of the data stabilizes the instrumental fluctuations. This effect is demonstrated by Figure ASC3.2 which reports the correlation between the eBC concentrations (AE51 and AE5x) averaged on the same intervals of 5 ng m^{-3} used in Figure ASC3.1 (R^2=0.986; slope=1.017).
The aforementioned results demonstrate the reliability of the seasonal phenomenology of the aerosol vertical profiles reported in Figure 5 and 12 and sections 3.3 and 3.4 along the manuscript for what concern BC concentrations.
Due to your question we added the aforementioned analysis to the revised version of the manuscript.

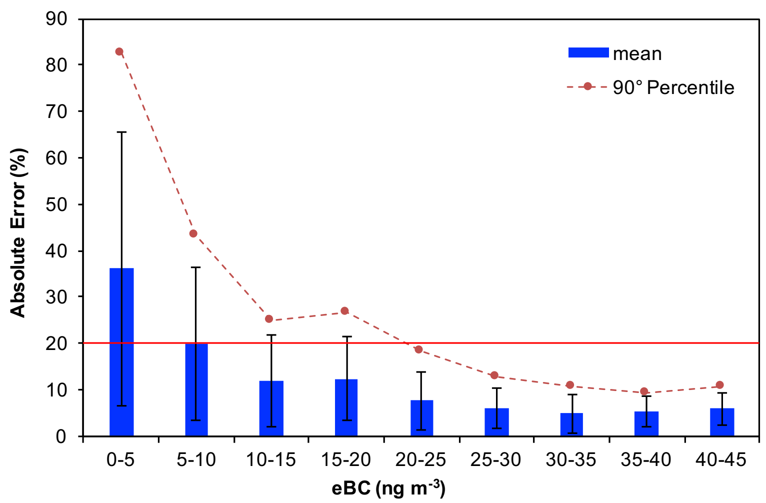

Figure ASC3.1. Absolute value of the error in percentage of the measured eBC in function of its concentration.

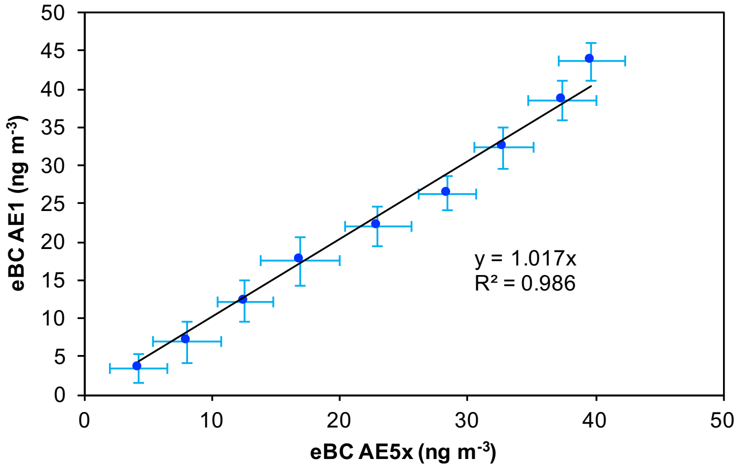

Figure ASC3.2. Absolute value of the error in percentage of the measured eBC in function of its concentration.

SC4: Page 10, Aerosol stratification height (ASh) Procedure for ASh estimation should be mentioned in the text.

ASC4: The procedure to estimate the AS_h is described in the manuscript at page 10 (lines 21-38) and page 11 (lines 1-2). It is based on the gradient method, which considers the minimum of the vertical derivative of the aerosol concentration as the AS_h. It is reported in the review of Seibert et al. (2000) and its application to vertical profile data has been widely described in previous works (Ferrero et al., 2012, 2011a, 2011b and 2007; Sangiorgi et al., 2011; Di Liberto et al., 2012). For this reason, we refer to the aforementioned publications in the manuscript.
However, due to your question, we explain the gradient method in section 2.2.3 in more detail (page 10, lines 21-38, and page 11, lines 1-2).

SC5: Sections 3.1 - 3.2 In the sections of 3.1 – 3.2, typical examples of vertical profiles of aerosols and meteorological parameters were mentioned. The description is slightly redundant. Some sentences can be simplified. In addition, general vertical structure of the boundary layer over Ny-Ålesund (i.e., thickness of surface inversion layer, and height of top of boundary layer) should be mentioned to understand characteristics of the vertical profiles. The vertical structure is associated closely with vertical features of aerosols.

ASC5: Thank you for this comment. We agree with you that description of the results in section 3.1 and 3.2 is slightly redundant. Thus, sections 3.1 and 3.2 were shortened and merged together in the revised version of the paper.
A comprehensive description of the general vertical structure of the boundary layer over Ny-Ålesund, is beyond the scope of the paper as the campaign was limited just to three months (April 2011, July 2011 and June-July 2012). We focus on the vertical structure of the boundary layer for the relevant periods.
With respect to the field campaign, θ and RH vertical behavior was described in sections 3.1-3.2 and figures 3-4. Figure 3 aimed to the discussion of the case studies chosen as representative for each profile class and Figure 4 reported the frequency distribution with altitude of both AS_h and θ and RH gradient.
With respect to the general behaviour of the PBL over Ny-Ålesund, we refer to the comprehensive work of Vihma et al. (2011) that describes the characteristics of temperature and humidity inversions and low-level jets over Svalbard fjords in spring.

The long-term upper-air observations by daily radiosondes provide an overview of the atmospheric vertical structure above Ny-Alesund, inlcuding the PBL altitude range (Maturilli and Kayser, 2016). In this climatological approach, Maturilli and Kayser (2016) identify the frequent occurrence of a temperature inversion layer in the shear zone above the mountain ridges that is typically present throughout the year, leading to a decoupling of the lowermost kilometer of the atmosphere from the free troposphere above. In between the mountains, the atmosphere is characterized by wind channeling along the fjord axis, disturbed by e.g. glacier outflow or land-sea breeze. Stable atmospheric conditions with suppressed vertical exchange occur frequently during polar night conditions, when a radiative surface-based inversion develops. Once the snow-melt leads to considerable sensible and latent heat fluxes at the surface, atmospheric stratification gets neutral or instable, allowing convection and vertical mixing.
We added these references to the introduction and we discussed the results reported in sections 3.1 and 3.2 (Figures 3-4) with respect to these references.

<u>SC6:</u> Figure 5 In addition to four groups, general vertical profiles (all data) should be shown in the Figure. The general profiles can be useful, when authors want to know general (average) vertical profiles. Because the vertical profiles of aerosols related to profiles of meteorological parameter (potential temperature and relative humidity), mean profiles of meteorological parameter (or normalized meteorological parameter) should be shown together with those of aerosols.

<u>*ASC6:*</u> *Thanks for the suggestion. We prepared the figure with all data for the revised version of the paper and we added this figure to the supplemental material (here below attached as Figure ASC6.1 for spring and ASC6.2 for summer). We also added the mean profile of normalized meteorological parameters (and related all data) to both Figures 5-12 and Figure ASC6.1 and ASC6.2 (here below). For normalized meteorological parameters we report absolute differences (Δ) of θ and RH along Hs with respect to the value assumed at Hs=0.*

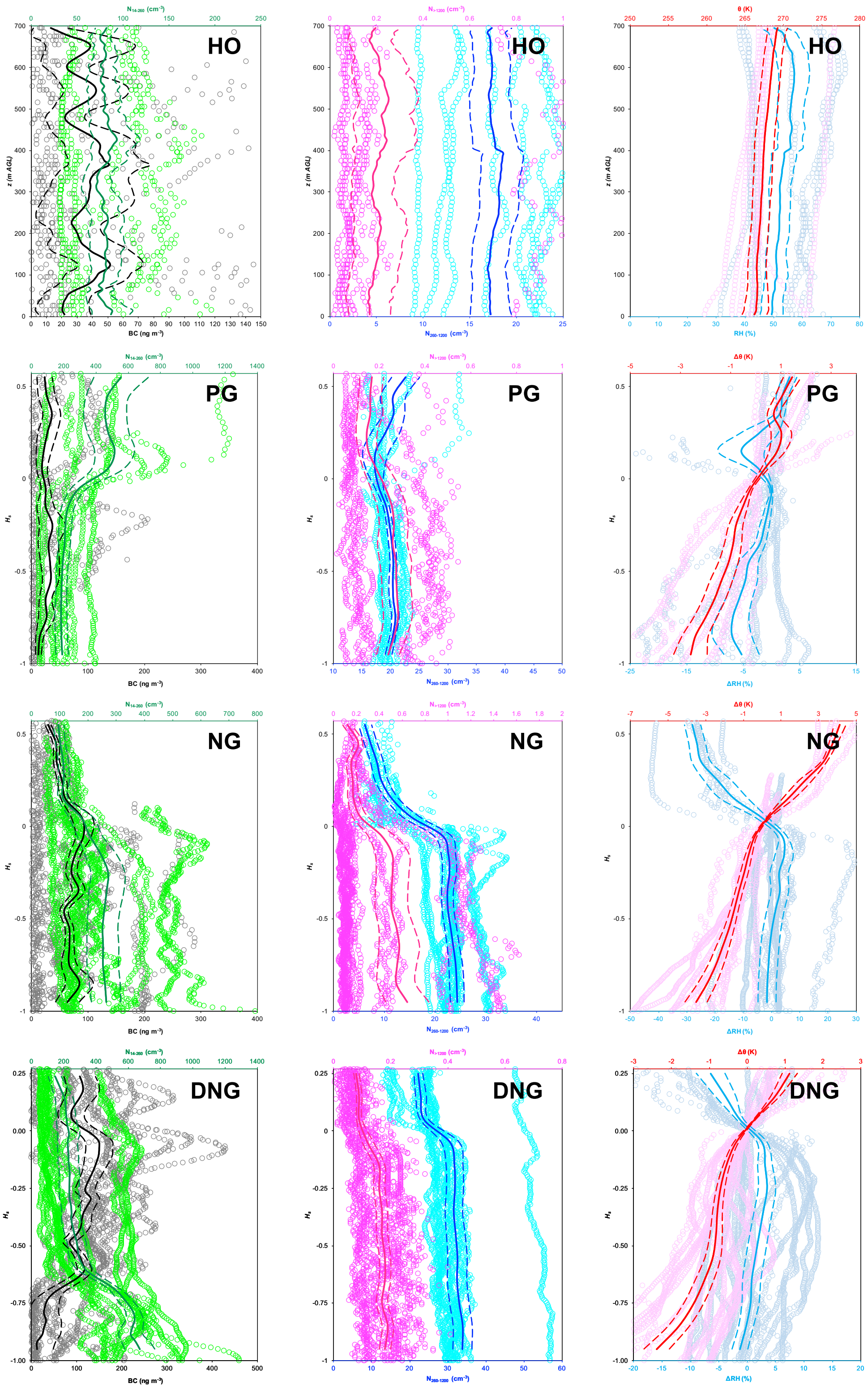

Figure ASC6.1. All data point of the collected vertical profiles during spring for each profile class. The average (solid line) and the mean standard deviation (dashed lines) are also reported.

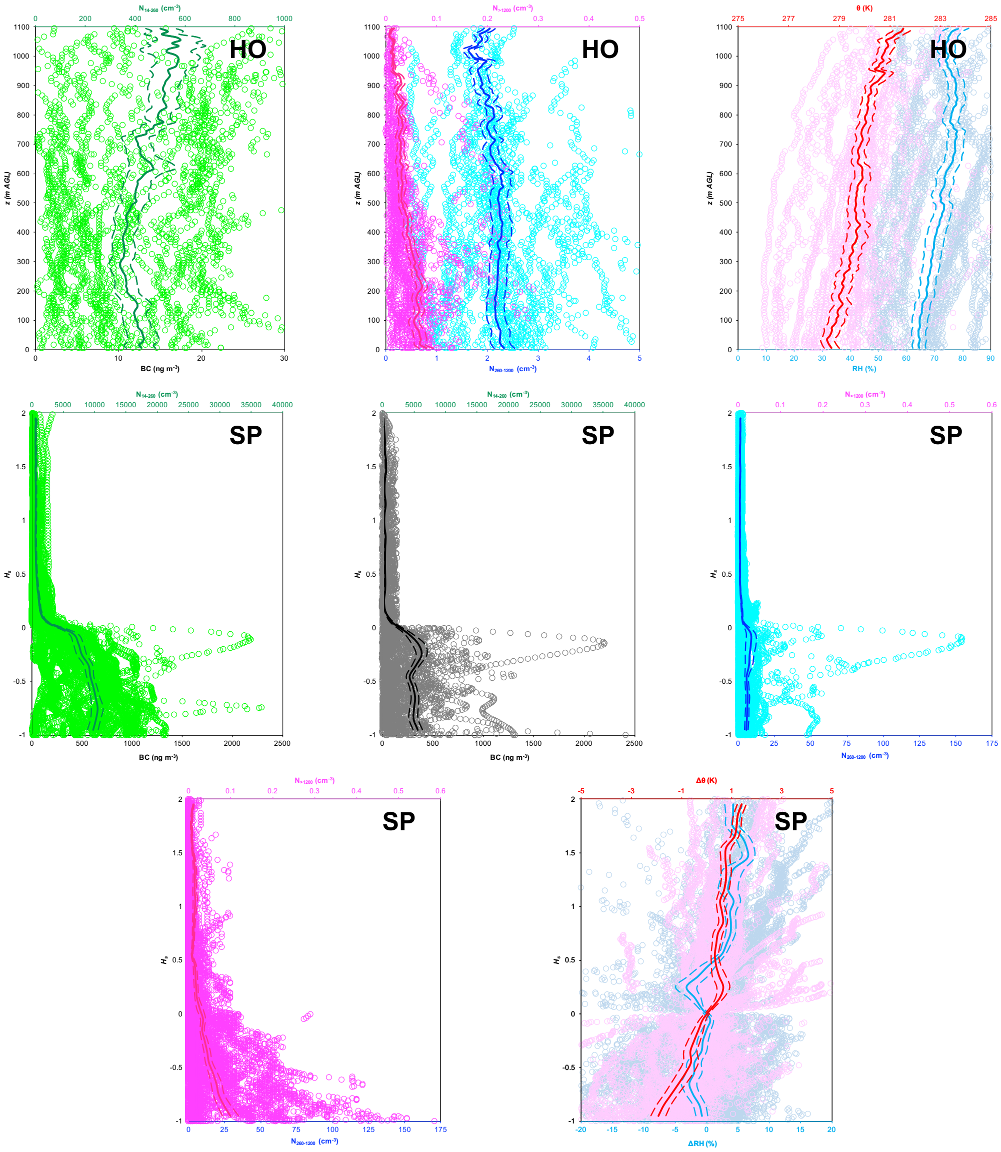

Figure ASC6.2. All data point of the collected vertical profiles during summer for each profile class. The average (solid line) and the mean standard deviation (dashed lines) are also reported.

<u>SC7:</u> Page 15 Line 8 Before statement about Figure 6, Figure 7 and explanation appeared here. Check or arrange figure number or description in the text.

<u>ASC7:</u> At page 15, lines 7-9 is stated: "Finally, the CCT wind data were used to compute wind rose graphs timely coincident with each profiles typology (Figure 7a-d). Results evidenced the absence of wind from north during the profiles, thus avoiding any influence from Ny-Ålesund". The sentence is right; Figure 7 reports the wind rose.

<u>SC8:</u> Page 15 Line 30 – 34 It is true that wet removal processes have an impact on aerosol number density, but dry deposition make an important contribution to the aerosol number density, especially in coarse particles.

<u>ASC8:</u> Thank you for this comment. We agree with you and we modified the sentence accordingly.

SC9: Page 15 Line 35-39 Air mass origins was shown and explained in only in "positive gradient profiles". However, transport pathway should be discussed together with aerosol source areas to show "plume" transport. In addition, general pattern and characteristics of transport processes in each type should be shown and discussed to understand relation between vertical profiles of aerosols and air mass origins.

ASC9: Please see our answer AGC4 to your general comment 4.

SC10: Page 15 Line 38- Some of particulate organics can be derived from secondary formation. However, there are matters to be discussed whether organics can play an important role in "new particle formation" or not. Actually, organics are condensable vapors to grow aerosol particles in ultrafine mode. So, authors should distinguish between new particle formation and secondary formation and mention them in the text.

ASC10: In section 3.3.4, during the discussion of DNG profiles, we summarized that the experimental results show high acidic sulphate fraction, low BC fraction, low temperature, high relative humidity, low wind speed during (page 17, lines 33-34) at the same time of the presence of a huge Aitken mode (page 18, lines 4-7). All of the aforementioned results pointed towards the presence of a ground-based plume of locally formed secondary aerosol (page 18, lines 7-9).
With the last sentence, we meant newly formed particle. Thus, to clarify the sentence we modified the paper accordingly to your suggestion.
For what concern the organics, at page 17, lines 38-40 we reminded that non only binary H_2SO_4–H_2O system is important for new particle formation but also the presence of organics, as well demonstrated in the work of Riccobono et al. (2014; cited in the paper). This was done to complete the discussion as several organics that can participate to this process were measured in spring at the Gruvebadet site (the site of the vertical profile campaign) in spring as reported in Zangrando et al. (2013).
Thus, for this reason we modified section 3.3.4 in the revised version of the paper accordingly to your observation.

SC11: Section 3.4 Different condition of solar radiation between spring and summer can engender change of height of top of the boundary layer. This change is very important to vertical profiles of aerosols and meteorological parameters. Other comments about section 3.4 is similar to previous comments about section 3.3.

ASC11: Thank you very much for this comment. We added the radiation data in section 3.2 of the revised version of the paper. The new sentence is as follows: "Summer homogenous profiles were more than twice in percentage that those measured in spring, due to a synergy of the higher solar power density at disposal (186.4±71.2 W m^{-2} in summer and 109.2±35.9 W m^{-2} in spring), together with a lower albedo (0.15±0.01 in summer and 0.87±0.04 in spring) induced by the summer snowmelt in Svalbard".

References:

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