Response to Reviewer#3

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, to eliminate the typos and to improve the language.

General Comment 1: This manuscript is based on vertical profiles of aerosol number density, eBC and ground measurements of the above at the Ny Alesund Arctic research station. This study is providing very useful data for the vertical structure of the aerosol column at a well studied area, where this type of data are still missing.
As a general outcome, the topic of the manuscript is relevant and suitable for the scope of "ACP". However, there are several points where the manuscript is failing to follow and deliver the methods and data quality needed for this study.
The description of the vertical structure of the atmosphere is well documented and useful and the classification of the different structures useful to relate to known aerosol properties based on the aerosol number size distributions from OPCs. There is also a good documentation of aerosol contamination events from harbour traffic of large boats.

Answer to the General Comment 1 (AGC)1: Thank you very much for your comment which underline the importance of the collected experimental data as well as the high relevance of the results presented in our paper. Here below we answer to the raised points.

General comment 2. The classification and discussion of results is not based on the understunding we can derive for the aerosol microphysics based the origin of aerosol during the study.

AGC2: The aim of the paper is to determine the seasonal phenomenology of the aerosol behavior along vertical profiles; this goal was chieved classifying the collected experimental data, according to their shape, both during spring and summer. Thus, attention was paid to the description of the vertical structure of the atmosphere and its relationship to the aerosol number size distribution.
We underline that, within the context of vertical profile classification, the aerosol microphysics and aerosol/air mass origin were investigated, when necessary, with the aim to deepen the understanding on aerosol vertical behavior for a specific vertical profile class.
In this respect, Figure 6 and related results reported in section 3.3.2 describe the influence of transport from mid-latitudes and the formation of PG profiles. Another example is related to the DNG profiles, which are related to the secondary aerosol formed close to the ground (section 3.3.4 and Figures 9, 10, 11).

General comment 3. There is no attempt to compare with data obtained by numerous studies in the area using aircraft or lidar techniques. Although several studies are mentioned no quantitative comparsion is given at least for the ground measuremnts or data published.

AGC3: Thank you for this comment as a comparison with previous studies is very important.
*An important comparison was reported in the manuscript, 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.*
This comparison underlined the accuracy of the collected data and, most importantly, suggested that all the investigated vertical profile classes may influence the background Arctic aerosol measured by Arctic observatories within GAW and EMEP observation programmes.

However, we fully agree with you that a better contextualization of the measuring campaign with respect to the international is required. Thus, we modified the introduction section referencing to previous airborne aerosol measurements carried out in Arctic area. Here below a brief resume.
The ARCTAS mission (Jacob et al., 2010 and reference therein) evidenced in spring a highly-layered air pollution transport from North America and East Asia characterized by anthropogenic aerosol below 2 km and by biomass burning above (2–4 km). Always in spring, the ARCPAC campign (Brock et al. 2011) allowed to describe and group the aerosol affecting the Arctic in four categories: a background troposphere (relatively diffuse, sulfate-rich aerosol), a region of depleted aerosol within the surface inversion layer over sea-ice, a layer of organic-rich biomass burning aerosol (above the top of the inversion layer) and a layer dominated by fossil fuel combustion.
During the trasnsition period from spring to summer in Svalbard, the ASTAR campaign (Engvall et al., 2008) found aitken and accumulation mode particles more concentrated in the free troposphere compared to boundary layer while, Kupiszewski et al. (2013), resumed results from the summer ASCOS campaign during which new particle formation events in the near-surface layer, possibly related to biological processes, were found.
For what concern the BC, the springtime results reported in the PAM-ARCMIP (Stone et al., 2010) and HIPPO (Schwarz et al., 2010) campaigns evidenced high BC concentrations close to the ground, below the thermal inversion but also dense pollution and BC at high altitudes over the Arctic (Wofsy et al., 2011). Interesting, the PAM-ARCMIP results evidenced a decrease of BC compared with respect to past measurements (i.e. AGASP, Hansen and Novakov, 1989). In addition, the HIPPO campaign revealed that in the lower troposphere the BC vertical gradient can change seasonally from positive to negative (Schwarz et al., 2013). In this respect, Spackman et al. (2010) and Koch et al. (2009) reported BC located mainly in the Arctic free troposphere with a positive gradient in the lower troposphere.
All the aforementioned findings appear very important from a long-term monitoring point of view as underline the need for continuous vertical profile campaign in the Arctic to improve the description of a seasonally resolved aerosol and BC vertical behaviour along time.
Thus, in order to discuss the results in the context of the aforementioned campaigns, we modified also the result sections including them.

General comment 4. The use of micro aethalometers in this area can be only used for obtaining EBC concentrations at minimum concentrations, which the authors have yet to derive. They show in figure 2c) a good correlation between the two micro-aethalometers used. This also shows an uncertainty at a 100% level for concentrations below 30 ug/m3 The other serious flaw in the processing of these data is the calculation of the absorption coefficient using a well established methodology and an unrealistic "C" factor. They quote a study in Milan where the "C" factor was derived for urban concentration levels and mixture of urban aerosol species. The authors must remove all absorption coefficients calculated in this manner and reported in this manuscript.

AGC4: Your comment is divided in three parts. Here below we answer to each one:

1. *Terminology for BC: we agree with you with the need to report the BC concentrations with the term eBC as suggested by Petzold et al. (2013) and by Andreae and Gelencser (2006) and as widely reported in literature (Gilardoni et al., 2010; Sthol et al., 2013; Eckhardt et al., 2013). We specified it in the method section.*
2. *Figure 2c: data reported in Figure 2c showed a good correlation. This result is first 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). However, we agree with you that a single point can be affected by a high level of uncertainty and that a deeper analysis of this comparison is required. Moreover, your question allows us to better explain our approach. In this respect, considering the average of the two EBC measurements (AE51 and AE5x) as the target value (as reported at Page 10, line 5) the absolute error (in percentage) of each*

eBC data was calculated. Figure AGC4.1 reports the average (± standard deviation) and the 90° percentile of the Absolute value of the error in percentage of the measured eBC in function of eBC concentration using intervals of 5 ng m^{-3} each. As it possible to observe at low concentration the error can reach values of 90% and more. This error decrease with increasing eBC concentration and reach a reasonable value (less than 20% for both the average and the 90° Percentile) above 20 ng m^{-3}. However, we have to remind that the aim of this paper is to determine the seasonal phenomenology of the aerosol behaviour 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 stabilize the instrumental fluctuations. This effect is demonstrated by Figure AGC4.2 which reports the correlation between the eBC concentrations (AE51 and AE5x) averaged on the same intervals of 5 ng m^{-3} used in Figure AGC4.1 (R^2=0.986; slope=1.017). The 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 eBC concentrations. We added the aforementioned analysis to the revised version of the manuscript.

3. *"C" factor: the reviewer's statement does not consider the experimental conditions of the C determination reported in Ferrero et al. (2011). Ferrero et al. (2011) (page 2832) state that: "The experimental design of vertical profiles does not require any estimation of the aerosol loading factor R(ATN): all vertical BC profiles were conducted by changing the filter ticket after each profile. Every Aethalometer measurement cycle (ascent and descent of the balloon) took less than 40–50 min. As a result, ATN never achieved values higher than 20 during all profiles, meaning that the b_{ATN} measurements were not affected by the "shadowing" effect due to filter loading. The average ATN measured along vertical profiles was 5±1". The experimental conditions for the study in Milano (Ferrero et al., 2011) were intentionally selected not to be influenced by the accumulation of the sample on the filter. This means that the total amount of aerosol collected on each filter during the determination of the parameter C was negligible, making the determined C values a function of the "filter material and the instrument specification" as again stated at page 2832. It is also possible to estimate the total amount of aerosol collected on each filter (ascent and descent of the balloon) during the C determination using the data reported in the paper. Considering the average BC concentrations below and above the mixing layer; the AE51 flowrate, the sampling time and the percentage of BC in PM (both below and above the mixing layer), it is possible to determine that on each filter (changed for each balloon launch) the total PM collected was less than 400 ng. The influence of the type of PM on the C values is therefore negligible, and the C reflects the instrumental properties, which are dominated by the highly scattering filter material. Finally, it is also necessary to observe that the C value was determined using data collected not only below the mixing layer but also above it, in a cleaner atmosphere, along the vertical profiles. The reliability of the obtained C (2.05±0.03) was also demonstrated further in Ferrero et al. (2014), both below the mixing layer and in the free troposphere. For the all the aforementioned reasons, considering that also the detailed explanation of the measurement protocol using the Aethalometer AE51 on page 9 of the manuscript, which ends with: "In this study… the filter tickets were changed to always keep ATN lower than 20 as recommended by Weingartner et al. (2003)", we maintain the estimation of the absorption coefficient. However, as the value of the absorption coefficient is very important in the Arctic, due to your question we decided to add the reference of Ran et al. (2016, ACPD) to the paper. In fact, until now, the C (2.05±0.03) reported in Ferrero et al. (2011 and 2014) was the only one for the AE51. Recently, on ACPD Ran et al. (2016) proposed 2.52 as C determined at ground-level in China. This value of the parameter C was*

determined in a completely different environment and, unfortunately the authors have not (yet) reported its uncertainty.

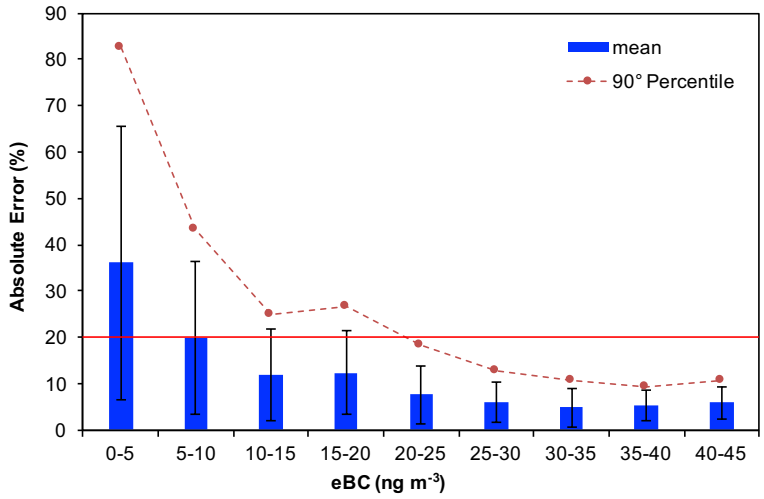

Figure AGC4.1. Absolute value of the error in percentage of the measured eBC in function of its concentration.

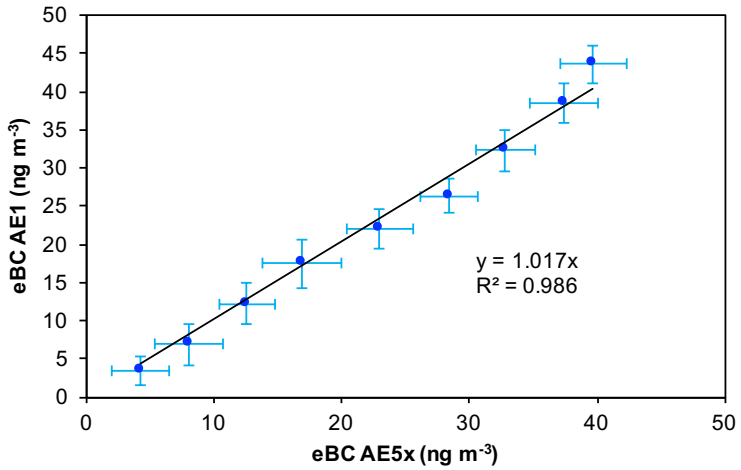

Figure AGC4.2. Absolute value of the error in percentage of the measured eBC in function of its concentration.

General comment 5. The chemical composition reported in figure 8 is given only in % of the total mass. How is the total mass derived and what are the actual mass concentrations of the different species reported in otherwise incredible detail where the non sea salt and non crustal fractions are calculated? These data do not appear realistic. For example in most cases the EC is found to 0.1 % of the aerosol mass. If one assumes that in the worst case eBC and EC mass concentrations can differ by a factor of 2 (+/- 100%) in the HO case where the eBC is found on average at 25 ng/m3 the aerosol mass concentration levels would range between1 to 50 ug/m3. This upper limit is totally unrealistic and even the 25 ug/m3 is extremely high. The other cases would produce even more grossly biased results.

This comparison puts in doubt the whole dataset of eBC and chemical data leaving the OPC and ground SMPS measurements as the only dataset worth considering for this manuscript.

AGC5: Thank you for this question which allowed us both to deepen the chemical characterization of the aerosol and to better describe the approach reported in the manuscript. Here below we answer to your questions following their order.

1- *The chemical composition reported in Figure 8 was given in percentage. This choice was done to put in evidence (section 3.3.4, page 16, lines 21-29) that "the total sulphate fraction in DNG profiles was double than that observed in the other profile classes" and that also "the nss-nc-SO_4^{2-} fraction (determined as reported in section 2.1.1) in DNG profiles was 2.4±0.4 times higher than that observed in the other profile classes while both the ss-SO_4^{2-} and the cr-SO_4^{2-} fractions remained quite constant". These observations, coupled with the lowering of eBC fraction in proximity of the ground (Figure 5g) and the SMPS data reported in Figure 9 show that the ground-based N_{14-260} concentration peak was due to secondary origin. Thus, the percentage of aerosol chemical composition was used in support of the other datasets to describe the origin of the aerosol found at ground in DNG profiles (see also AA2).*

   However, do to your question we are going to change figure 8 to report the absolute concentration values (ng m^{-3}) of each aerosol chemical component. We moved the previous figure 8, reporting relative contributions, to the supplemental material.

2- *The total mass was measured by weighing the filters before and after the sampling, using a 5-digit microbalance (Sartorius ME235P) after they were conditioned for 48 hours (25°C and 50% relative humidity). The reproducibility error on filter weighing was lower than 5% (experimentally evaluated) as described in section 2.1 (page 5, lines 5-7).*

   The absolute concentration values (ng m^{-3}) of each aerosol chemical component were added in the new figure 8. Moreover the same concentration values (ng m^{-3}), together with the corresponding detection limit (ng m^{-3}) of each aerosol chemical component were reported in a new Table in the supplemental material. This table (Table AGC5.1) is attached here below. The detection limits refer to the sampling conditions and the analytical procedures. Particularly, as reported in section 2.1.1, for the ion chromatography, only half filter was used (extraction in 12 ml of ultrapure water), 55 m^3 of air were filtered for each sample (24 h sampling). For the EC/OC analysis, a 1.5 cm^2 punch was used and the volume of air filtered for each sample (96 h sampling) was 220 m^3.

   Table AGC5.1 (please see below) shows that all the analyzed chemical components were characterized by ambient concentrations largely above the detection limit.

   The non-sea salt and crustal fractions of sodium, calcium and sulphate were determined as well documented in section 2.1.1. This approach produce reliable results as documented in previous works (Udisti et al., 2016; Giardi et al., 2016; Becagli et al., 2012; Udisti et al., 2012) already referenced in the paper text.

   When comparing the eBC and EC several points has to be considered. First of all they are not the same quantity and, as reported by Petzold et al. (2013), they can differ by a factor of 7 and depend on the assumed MAC (for optical measurements) and the thermal protocol for EC/OC analysis. As EC is the only component, which is at the limit of detection, we decided to not report this component in the new figure 8 (EC is also not important in the discussion reported in section 3.3.4 in relation to DNG profiles).

   Most important, it has to be considered that vertical profile data are "quasi instantaneous" while the reported chemical composition was determined with the time resolution of 24 h for ionic species and 96 h for EC/OC. In this respect, the chemical speciation and in particular the sulphate content was only used along the paper to support the secondary origin of the ground-based concentration peak of N_{14-260} in DNG profiles. For this purpose,

the chemical composition was coupled with information coming from the vertical profiles, the SMPS data and the meteorological parameters.
These observations underline the reliability of the whole dataset used in the paper, and therefore we absolutely reject the statement that these data appear non-realistic.

Conc (ng m $^{-3}$)		Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_2^-	NO_3^-	SO_4^{2-}	Oxalates	F^-	Glycolate	Formate	MSA	EC	OC
HO	mean	410.85	66.33	21.73	54.21	39.45	495.82	22.51	59.92	504.71	4.79	0.18	1.16	2.15	2.28	<DL	534.36
	σ_m	252.44	13.85	8.43	27.28	7.67	332.03	10.00	12.96	93.15	0.98	0.08	0.16	0.70	0.58	<DL	39.52
PG	mean	655.24	74.61	28.81	79.34	43.28	871.20	36.00	68.66	584.69	5.46	0.21	1.29	2.92	4.47	<DL	522.87
	σ_m	293.67	12.22	9.50	31.70	7.07	392.54	15.66	12.22	72.69	0.54	0.13	0.29	0.68	1.07	<DL	52.69
NG	mean	590.94	85.04	27.34	73.35	38.88	745.67	31.43	51.64	779.09	4.99	0.07	1.05	2.99	3.57	<DL	517.13
	σ_m	192.18	15.89	5.98	20.50	4.71	267.28	11.98	9.59	204.18	0.50	0.09	0.14	0.47	0.81	<DL	43.96
DNG	mean	325.71	128.31	23.94	51.21	33.45	357.20	20.06	49.37	1441.91	6.97	<DL	1.27	2.78	1.84	31.85	689.94
	σ_m	84.23	29.21	2.40	13.16	3.17	173.05	2.33	15.90	354.09	1.12	<DL	0.19	0.38	0.36	0.20	19.59
Detection Limit		0.04	0.4	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.4	0.004	0.4	0.4	0.04	11	120

Table AGC5.1. Ambient concentrations (ng m^{-3}) of the aerosol chemical components (mean±mean standard deviation) and their analytical detection limits (DL).

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