Response to Reviewer#1

We thank the reviewer for his or her helpful comments and insight. We respond to the general and to the 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.

<u>General Comment 1</u>: This study reports vertical profiles of aerosol number size distribution and black carbon (BC) concentrations from balloon measurements during a field campaign in Ny-Ålesund, Svalbard spring and summer 2011-2012. The authors divide the number size distribution into 3 modes and classify the vertical profiles in four shapes during spring. The authors also discuss secondary aerosol formation and emissions from shipping during summer.

This study is important because 1) the vertical distribution of aerosols affects its radiative forcing and 2) measurements of the vertical distribution of aerosols in the Arctic are particularly sparse. Since such measurements are highly needed and valuable, I think this study is relevant and within the scope of ACP. The Method section is clearly outlined and the different instruments used are sufficiently and well explained. The figures are nice and easy to follow. However, I think the overall presentation of the results should be improved before it can be published. The manuscript needs more work in terms of language and structure. If this can be achieved, I recommend the manuscript for publication.

<u>Answer to the General Comment 1 (AGC)1:</u> Thank you very much for your comment which underline the experimental efforts and the high relevance of the results presented in our paper. We agree with you that an improved organization of the manuscript and a better presentation of the results is necessary. For this reason, 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.

<u>General comment 2</u>. The quality of the English language in this manuscript is variable (some parts are good, but others less good), and I think it would benefit by a thoroughly review of the language (and a spell check!). I have added a few examples under minor comments.

<u>AGC2:</u> The manuscript was proofread and edited, to eliminate the typos and to improve the language as required. A particular attention was given to shorten the several long sentences present in the paper, as also required in your minor comment 4. Thank you for the suggestion.

<u>General comment 3</u>. The Results section would be easier to read if it was shortened a bit. Description of the methodology should always be under Methods, not Results. I have a few specific suggestions below.

<u>AGC3:</u> Thank you for this comment. We agree with you. The description of the methodology (presently in the result section) was moved to the method section. Moreover, sections 3.1 and 3.2 were shortened and merged together in the revised version of the paper.

<u>General comment 4</u>. I miss a broader implication of this study. Why did you separate the profiles into the four shapes? Comprehensive measurement studies like this can provide physical understanding for evaluation/improvement of the modeling of aerosol processes. Do you have any suggestions? I understand that you cannot add any modeling, but I would like to know more what we can learn from this study.

<u>AGC4:</u> Thank you for this question. Out answer is also related to that reported below for your specific comment SC22 (see our answer ASC22).

We separated the profiles in the four shapes because each shape is the result of an interplay of several processes: 1) transport events, 2) the planetary boundary layer dynamics and 3) the local formation of aerosol. The different combinations of these factors result in a specific profile class.

Figure 6 represents a good example in which the transport of polluted air masses from midlatitudes generated initially PG profiles that naturally evolved (due to the entrance into the PBL) into a NG profile.

Even though a modelling simulation is beyond the scope of the present paper, some indication can be obtained. One of this is related to your question about the validity of ground-based measurements with respect to the vertical aerosol distribution in modelling comparison (SC21). HO profile showed that ground measurements are fully representative of the vertical column (up until ~1 km, our vertical limit) while during NG and PG profiles the ground based measurements are representative for the column up to the PBL. DNG profiles show that ground-based measurements differ from the measurements performed within the column. However, the last case is influenced by secondary aerosol formation that can be easily detected by an SMPS.

Thus ground-based measurements (coupled with a proper PBL determination) are fundamental for model validation.

<u>General comment 5</u>. Measurements of vertical profiles in the Arctic are sparse, but there are a few, e.g.:

- two ARCTAS campaigns in the North American Arctic (Jacob et al. 2010) in April and June/July 2008

- the ARCPAC campaign conducted together with ARCTAS in spring 2008 (Brock et al. 2011) -the PAMARCMIP campaign in April 2009 (Stone et al. 2010) -the HIPPO campaign (Schwarz et al. 2010, 2013; Wofsy 2011) January and October 2009 + winter and autumn 2009

- the ARCTAS/ARCPAC campaign in spring 2008,

- the ARCTAS campaign in summer 2008

- the PAMARCMIP campaign in spring 2009.

On a general basis; How are those compared to your study? I suggest you also include more of these studies in the introduction.

Jacob, D.J., J.H. Crawford, H. Maring, A.D. Clarke, J.E. Dibb, L.K. Emmons, R.A. Fer- rare, C.A. Hostetler, P.B. Russell, H.B. Singh, A.M. Thompson, G.E. Shaw, E. McCauley, J.R. Pederson and J.A. Fisher, 2010. The Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) mission: design, execution, and first results. Atmospheric Chemistry and Physics, 10:5191-5212.

Brock, C.A., J. Cozic, R. Bahreini, K.D. Froyd, A.M. Middlebrook, A.McComiskey, J. Brioude, O.R. Cooper, A. Stohl, K.C. Aikin, J.A. De Gouw, D.W. Fahey, R.A. Ferrare, R.-S. Gao, W. Gore, J. Holloway, G. Hubler, A.Jefferson, D.A. Lack, S. Lance, R.H. Moore, D.M. Murphy, A. Nenes, P.C. Novelli, J.B. Nowak, J.A. Ogren, J. Peischl, R.B. Pierce, P. Pilewskie, P.K. Quinn, T.B. Ryerson, K.S. Schmidt, J.P. Schwarz, H. Sode- mann, J.R.Spackman, H. Stark, D.S. Thomson, T. Thornberry, P. Veres, L.A. Watts, C.Warneke and A.G. Wollny, 2011. Characteristics, sources, and transport of aerosols measured in spring 2008 during the aerosol, radiation, and cloud processes affecting Arctic climate (ARCPAC) project. Atmospheric Chemistry and Physics, 11:2423-2453.

Wofsy, S.C., 2011. HIAPER Pole-to-Pole Observations (HIPPO): finegrained,global- scale measurements of climatically important atmospheric gases and aerosols. Philo- sophical Transactions of the Royal Society, 369:2073-2086.

Stone, R.S., A. Herber, V. Vitale, M. Mazzola, A. Lupi, R.C. Schnell, E.G.Dutton, P.S.K. Liu, S.M. Li, K. Dethloff, A. Lampert, C. Ritter, M. Stock, R. Neuber and M. Maturilli, 2010. A threedimensional characterization of Arctic aerosols from airborne Sun pho- tometer observations: PAMARCMIP,April 2009. Journal of Geophysical Research: Atmospheres,115:doi 10.1029/2009jd013605.

Schwarz, J.P., J.R. Spackman, R.S. Gao, L. Watts, P. Stier, M. Schulz, S.M. Davis, S.C. Wofsy and D.W. Fahey, 2010. Global-scale black carbon profiles observed in the remote atmosphere and compared to model. Geophysical Research Letters, 37:L18812,doi:10.1029/2010gl044372.

Schwarz, J.P., B.H. Samset, A.E. Perring, J.R. Spackman, R.S. Gao, P. Stier, M.G. Schultz, F.L. Moore, E.A. Ray and D.W. Fahey, 2013b. Global-scale seasonally re- solved black carbon vertical profiles over the Pacific.Geophysical Research Letters, 40:5542-5547.

AGC5: Thanks for this comment.

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.

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 is required.

Thus, we modified the introduction section, adding and discussing the suggested campaigns (and related references). Moreover, we discussed the obtained results with respect to the same references.

Specific Comment 1 (SC1): Abstract: You should mention in the abstract that these were balloon measurements up to 1200 meters height.

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

<u>SC2:</u> Page 2, L 17: 'to influence with semi-direct effects the atmospheric properties'. Could this be rewritten and explained further, maybe by 1 or 2 examples?

<u>ASC2:</u> The sentence was rewritten as follows: "Many of these effects can be altered by the aerosol due to its ability to absorb and scatter solar radiation (direct effect) or to seed and modify the cloud properties (indirect effects). In addition, light absorption by BC can alter the atmospheric thermal structure within, below, or above clouds, consequently affecting cloud distributions (IPCC, 2013; Bond et al., 2013; Ramanathan and Feng, 2009; Koren et al. 2008; Koren et al., 2004; Kaufman et al., 2002)".

<u>SC3:</u> Page 2, L 34: You mention Arctic Haze here without explaining it. Since this is an important part of your study, I think you should briefly explain the phenomena with a few references (e.g. Stohl 2006). Stohl, A. (2006), Characteristics of atmospheric transport into the Arctic troposphere, J. Geophys. Res., 111, D11306, doi:10.1029/2005JD006888.

<u>ASC3:</u> Thanks for raising this point, we agree with you that a better description of the Arctic Haze could help the reader. For this reason, we added to the introduction the following description: "The Arctic Haze is an effect, where an inflow of pollution (aerosol and gases) from northern midlatitudes during winter-spring, result in a reduction in visibility (Jacob et al., 2010; Sthol et al., 2006; Radke et al., 1984; Barrie and Hoff, 1985; Brock et al., 1989; Shaw, 1995). The Arctic Haze manifests itself during special meteorological conditions (thermally stable stratifications with frequent and persistent occurrences of surface-based inversions) during which the air pollution can be transported into the Arctic at low-level (followed by ascent in the Arctic or low-level alone) or with an uplift outside the Arctic, followed by descent in the Arctic itself (Sthol, 2006)."

<u>SC4:</u> Page 3, L 25: 'These reports may well highlight opposing forms of behavior '. I am not quite sure what this means?

<u>ASC4:</u> The intention was to underline the differences in the vertical aerosol behavior found during the reported field campaigns. However, to avoid any confusion, we delated this sentence and replaced it with your suggestion in the following specific comment.

<u>SC5:</u> Page 3, L 25: One reason for this difference between the observations could be the strong influence of biomass burning during spring 2008 (Warneke et al. 2010).

Warneke, C., K.D. Froyd, J. Brioude, R. Bahreini, C.A. Brock, J. Cozic, J.A.de Gouw, D.W. Fahey, R. Ferrare, J.S. Holloway, A.M. Middlebrook, L.Miller, S. Montzka, J.P. Schwarz, H. Sodemann, J.R. Spackman and A.Stohl, 2010. An important contribution to springtime Arctic aerosol from biomass burning in Russia. Geophysical Research Letters, 37:L01801, doi:10.1029/2009GL041816.

<u>ASC5:</u> Thank you very much for the suggestion, we added the reference to the introduction section.

<u>SC6:</u> Page 3, L 30: Drop 'should', as this is written it seems like you tell the emissions to do so? In bracelets: we do not know for sure if these emissions will warm the surface and be deposited, but as you write above; studies show that there are higher probability for this to happen when the concentrations are located close to the surface.

<u>ASC6:</u> Thank you. We modified the sentence.

<u>SC7:</u> Page 4, L 23: Could you add just one sentence summarizing this table? E.g. 25 measurement days, balloons measured 2-14 profiles each day, altitude range?

<u>ASC7:</u> Thank you for the suggestion. The new sentence is as follows: "Table 1 lists the dates of the campaign (25 measurement days), the number of flights (197 measured profiles), the maximum altitudes (~700-1300 m) and the cloud base height (clouds present for 48% of the campaign)."

<u>SC8:</u> Page 5, L 5: Is there a reference for this instrument and the calculated uncertainty in mass concentrations?

<u>ASC8:</u> As stated at page 5, line 5, we used the 5-digit Sartorius ME235P microbalance. In this respect, 5% is the uncertainty related to the weighing procedure experimentally evaluated. The text is changed as follows: reproducibility error on Filter weighing was lower than 5% (experimentally evaluated).

<u>SC9:</u> Page 6; L 18: Could you briefly here explain what you mean when the atmosphere is 'stable' and does not encourage vertical mixing? (in terms of potential temperature)

<u>ASC9:</u> We modified the sentence as follows: "the atmosphere is stable for about 50% of the time along the year. The term stability refers to the propensity of air masses to move vertically: stable air resists to vertical motion, while unstable air masses are prone to vertical movements. A parcel of air results to be stable/unstable if the temperature lapse rate is lower/higher than the adiabatic

one, i.e. if the potential temperature is increasing/decreasing with height. Air within a stable layer is not turbulent and these conditions will cause pollutants to become trapped near ground level, as the vertical mixing of the aerosols is not encouraged".

<u>SC10:</u> Page 8, L 28: In this paragraph you define the 3 modes of particles, 'Aitken', 'Accumulation' and 'coarse' and say that you will also use these names for the rest of the discussion, but most of the time you use N14-260, N260-1200, and N>1200 anyway. I suggest you use the names Aitken etc. throughout the text once you have defined them, as this is easier to read.

<u>ASC10:</u> Thank you for addressing it. With the sentences reported at page 8, line 28 we wanted to explain the meaning of each investigated broadsize range. We consider your comment while preparing the revised version of the manuscript.

<u>SC11:</u> Page 8, L 36: Since there are many figures in this paper; I suggest removing fig 2 (or move to the supplementary).

ASC11: Figure 2 was moved to the supplemental material.

<u>SC12:</u> Page 9: There are various methods to measure BC concentrations, and they can disagree by a factor of seven or more (Petzold et al 2013). Since the (common) filter-based method like you have used is not a direct measurement of BC, it is recommended to report the resulting BC concentration (eBC) together with the assumed MAC value. Maybe you should change 'BC' to 'eBC' to make sure that we know that this is equivalent BC? I also think you should add a brief discussion on how your measurements depend on the assumed MAC number (you use 12.5 m2/g?) (or at least make a note about this).

Petzold, A., J.A. Ogren, M. Fiebig, P. Laj, S.M. Li, U. Baltensperger, T. Holzer-Popp, S. Kinne, G. Pappalardo, N. Sugimoto, C. Wehrli, A. Wiedensohler and X.Y. Zhang, 2013. Recommendations for reporting "black carbon" measurements. Atmospheric Chemistry and Physics, 13:8365-8379.

<u>ASC12:</u> Petzold et al. (2013), Andreae and Gelencser (2006) and other authors (Gilardoni et al., 2010; Sthol et al., 2013; Eckhardt et al., 2013), suggest reporting BC concentrations with the term eBC, arising from the need to report the method to determine BC and the parameters used in the method. When optical methods are used to measure light transmission through the filter loaded with BC, the mass equivalent concentration is determined using the mass attenuation cross-section (σ_{ATN}). For the case of our measurements, we report the σ_{ATN} value 12.5 m² g⁻¹ We note this value (Ferrero et al., 2011) in the method section, as the one used in the micro-Aethalometer AE51, page 9, line 23. This approach was also used by Eleftheriadis et al. (2009) when reported ten years of BC measurements in Ny-Ålesund.

Moreover, the σ_{ATN} value (12.5 m² g⁻¹) "was obtained by comparing the BC values measured with the microAeth® Model AE51, with an AE31 Aethalometer (880 nm wavelength) operating in a test chamber with different BC concentrations at low attenuation values. The comparison was then repeated using ambient air" (as reported in Ferrero et al., 2011).

This value is not far from the σ_{ATN} values of 15.2 $m^2 g^{-1}$ and 15.9 $m^2 g^{-1}$ reported in Eleftheriadis et al. (2009) and applied to the attenuation coefficient measured at the Zeppelin station (Ny-Ålesund) with the Aethalometers AE9 and AE31, respectively. The difference between these values results from the use of different filter materials to collect the sample in the different Aethalometers, which was quantified in Ferrero et al. (2011) and Drinovec et al (2015).

We compare our results with those previously measured at Ny Ålesund in the manuscript, page 15, lines 4-8, where we state: "the columnar average of BC concentrations obtained by averaging the profile classes was 52 ± 8 ng m⁻³ (eBC)". This value "perfectly agrees with long-term data series collected over Ny-Ålesund at the Zeppelin observatory (Eleftheriadis et al., 2009) during Spring."

Finally, considering the relationship between the attenuation and absorption coefficients (page 9, equation 9), the apparent mass attenuation cross-section of 12.5 m² g⁻¹ corresponds to the mass absorption cross-section (MAC) of 6.1 m² g⁻¹ (using $C=2.05\pm0.03$). As suggested by Petzold et al. (2013), we report the wavelengths, the mass attenuation cross-section and the mass absorption cross-section used in the determination of the absorption coefficients, using the methodology reported in Weingartner et al. (2003). The MAC of 6.1 m² g⁻¹ is in agreement with the previously published range of values (see for example Petzold et al. (2013) and references therein).

<u>SC13:</u> Page 9: Filter-based methods are sensitive to absorbing and non-absorbing non-BC particles. Could you please add a few sentences about the uncertainties in your method as well?

<u>ASC13:</u> Considering the absorbing non-BC particles, different types of aerosol may in principle contribute to the signal in Aethalometers (i.e. Brown Carbon, dust). However, BrC is characterized by negligible absorption in the infrared (Andreae and Gelencsér, 2006), the wavelength range of the eBC measurements (micro-Aeth AE51 uses 880 nm). In this respect, Massabò et al. (2013) show the potential contribution of BrC to the determination of eBC to be below 10%.

To estimate the possible influence of BrC on eBC measurements carried out during the spring 2011 campaign, the few data collected with the micro-Aeth prototype AE5x at 370 and 880 nm were considered. Particularly, the Aethalometer model (Sandradewi et al., 2008) was applied to the apportionment of absorption due to both BC and BrC as reported in Massabò et al (2013) and in Shamjad et al. (2015) as follows:

$$\frac{b_{abs}(370 \ nm)_{BC}}{b_{abs}(880 \ nm)_{BC}} = \left(\frac{370}{880}\right)^{-\alpha_{BC}} \tag{1}$$

$$\frac{b_{abs}(370 nm)_{BrC}}{b_{abs}(880 nm)_{BrC}} = \left(\frac{370}{880}\right)^{-\alpha_{BrC}}$$
(2)

$$b_{abs}(\lambda) = b_{abs}(\lambda)_{BC} + b_{abs}(\lambda)_{BrC}$$
(3)

where α_{BC} and α_{BrC} represent the Absoprtion Angstrom Exponents of BC and BrC, respectively. The value for α_{BC} was taken to be 1 as suggested by Massabò et al (2013) and Sandradewi et al. (2008), while α_{BrC} was set at 3.5 (Yang et al., 2009), 3.95 (Massabò et al., 2013), 6.6 (Shamjad et al., 2015) and 9.0 (Bikkina and Sarin, 2013), respectively. With this inputs, the percentage of absorption at 880 nm due to BrC instead that BC was 8.5%, 5.8%, 0.5% and 0.1%, respectively. Thus, it is possible to estimate that the BrC positive artifact on eBC measurements was less than 10% during the campaign.

The effect of non-absorbing particles on the filter photometer measurements was also quantified for the Aethalometer AE33 Drinovec et al (2015), which uses the same filter material as the AE51, and was shown to be below 2.5%. To reduce the effect of the non-absorbing particles sampled onto the filter, the experimental protocol during the campaign followed that reported in Ferrero et al. (2011): "all vertical BC profiles were conducted by changing the filter ticket after each profile. As a result, ATN never achieved values higher than 20 during all profiles. The average ATN measured along vertical profiles was 5 ± 1 ". This means that the total amount of aerosol collected on each filter during the vertical profile was very low resulting in a negligible effect of non-absorbing particles.

We added the aforementioned considerations to the supplemental material and referenced them in the method section.

SC14: Page 11, L 15: 'Figure S1a shows a larger interannual springtime variability.' Of what?

<u>ASC14:</u> Thanks for addressing this point, with this sentence we meant that Figure S1a shows a larger interannual springtime variability of temperature measured close to the ground. Due also to your question here below (SC14) we rewrite the manuscript text at page 11, lines 13-20 with the following sentence: "The temperature measured in spring 2011 was within the standard deviation range of the long-term observations, while a 10-day period at the end of April 2011 was slightly warmer than the climatological mean (Figure S1a). The temperatures during the summer seasons 2011 and 2012 were mostly within the range of the long-term observations, so the vertical profile measurements can be considered to have been obtained under typical meteorological conditions representative for the Ny-Ålesund environment".

<u>SC15:</u> Page 11, L 13 - 20: Since you are referring to figures in the supplement, maybe you could rewrite this paragraph so this is easier to follow without the figures? Not 'Fig Sxx shows ..' but instead just briefly state that the spring season had surface temperatures close to the climatology, summer season had .. etc. and then mention that figures are in Supplementary (–OR- move the sup. figures to the paper, but then you already have many figures there).

<u>ASC15:</u> We modified the paper accordingly to your observation.

<u>SC16:</u> P11: 'Particularly, the maximum wind speeds registered at ground during balloon flights in spring 2011 and in summer 2011 2012 were 4.9 m s and 10.7 m s lower than the absolute wind speeds registered during the same periods: 27.9 m s and 16.3 m s.' I'm not sure if I understood this. The absolute winds measured by ..? With movement? How do you conclude that the measurement periods are representative for days with low winds?

<u>ASC16:</u> The wind speed was measured at the Amundsen-Nobile Climate Change Tower as described in section 2.1.2 (page 6, lines 13-16). The wind speed (average, max value) measured during balloon flights was lower than that during the whole period (April 2011, June and July 2011-2012) of the campaign. Thus, we reported that balloon flights have limitations with respect to its launch conditions, in particular they favor low wind conditions as it is very difficult to launch the balloons during high winds. We understand from your question that the sentence was not clear. Thus, we rephrased it as follows: "It should be reminded, however, that the tethered balloon has limitations with respect to its launch conditions (section 2.2). Particularly, balloon profiles are measured in low wind conditions, as it is very difficult to launch the balloons during high winds – this introduces a bias in respect to average meteorological conditions above the launch site. The maximum wind speed measured at the Amundsen-Nobile Climate Change Tower (section 2.1.2) during balloon flights was lower than that during the whole period of the campaign (April 2011, June and July 2011-2012): 4.9 m s⁻¹ and 10.7 m s⁻¹ (springtime and summertime balloon profiles) compared to 27.9 m s⁻¹ and 16.3 m s⁻¹ (full spring 2011 and summer 2011-2012)."

<u>SC17:</u> Page 11, line 32 - page 13, line 14: I think you spend too much time explaining figure 3. Parts of this can be moved to Methods, e.g. what type of information you can retrieve from the measurements. You can also move parts to Introduction as a way of motivating the study. When I read the 'Results'-chapter I want to read about the results right away. Could you also try to merge some of this information when you present the other results? I would skip everything between L31, p11 to L21,p12 and start on 'An example ...'. Is fig 3 needed at all? Why cannot the measured potential temperature and the RH for each group be plotted in fig 5 instead?

On the other hand, figure 7 is hardly mentioned. Can the wind roses be put in better context with the profiles described in 3.3.1-3.3.4? Also, this text is a bit hard to read, because of all the numbers listed. Do you need to list them all? Maybe put them in a table?

<u>ASC17:</u> We agree with your observation. Particularly, as we reply in AGC3 to your general comment, all the description of the methodology presently in the result section was moved under the method section. Sections 3.1 and 3.2 were shortened and merged together in the revised version of the paper. We considered your suggestion for Figure 3 and we added averaged meteorological parameters to figure 5.

Figure 7 was put in evidence in the revised version of the paper and a table resuming data of the campaign was prepared.

SC18: Page 17, L3: does this text and forward belong to 3.3.4 or should it have a separate heading?

<u>ASC18:</u> Page 17, line 3 and the following lines belong to section 3.3.4. Section 3.3.4 is quite long, but we shortened it following your suggestion (see AGC3) to move the methodology present here in the method section.

SC19: Page 18: anything that has to do with methodology should be under Methods, not Results.

<u>ASC19:</u> We agree with you and we modified the paper accordingly.

SC20: Page 19, L16: what is meant by a 'meaningful' impact of ship emissions?

<u>ASC20:</u> The intention was to point out that profiles were affected by a local, high, plume emitted from the ships. We agree that the sentence, as stated, was not clear and thus we rephrased it as follows: "Type 2, profiles characterized by the presence of shipping emissions (hereinafter addressed as SP), Figure 12c-d".

<u>SC21:</u> Page 20: It is interesting to see the impact of ship emissions. Could you remind us here how far the measurements were from the ships? This also relates to your final conclusion on page 21 (where you suggest that increased shipping could significantly increase BC concentrations during summer and enhance climate change in the Arctic). Currently, BC emissions from shipping in the Arctic comprise a small fraction of within- Arctic BC. Browse et al. 2012 found that even under a high-projection of shipping, by 2050 BC emissions from shipping would still contribute less than 1 % of total Arctic deposition. Do you suggest that current emission inventories are too low and that future emission projections should also be higher?

Browse, J., K.S. Carslaw, S. Arnold, K.J. Pringle and O. Boucher, 2012. The scavenging processes controlling the seasonal cycle in Arctic sulphate and black carbon aerosol. Atmospheric Chemistry and Physics, 12:6775-6798.

<u>ASC21:</u> Vertical profile measurements in summer were carried out from the German-French AWIPEV research base (78°55'24" N 11°55'15"E), 600 m far from the harbor (we added this information to section 2). Thus, the reported results (section 3.4.2, Figure 12c-d and Figure 13) refer to the local impact of ships.

The obtained results are in agreement with those reported in Eckhardt et al. (2013), who showed an enhancement of 72 and 45 % (up to 81 and 72 % in stagnant conditions) of eBC, when ships cruised in the Kongsfjord, compared to values when ships were not present.

From these results, yet Eckhardt et al. (2013) concluded that the eBC increase due to shipping emission can "be taken as a warning signal of future pan-Arctic conditions if Arctic shipping becomes more frequent and emission regulations are not strict enough".

On the other hand, as you addressed, shipping contribution was less than 1% of BC emissions in the Arctic as reported in in the study of Stohl et al. (2013) where, conversely, gas flaring was estimated to contribute 42% to the annual mean BC surface concentrations in the Arctic dominating the estimated BC emissions north of 66° N.

However, even also Corbett et al. (2010) reported that "the magnitude of emissions from shipping on a mass basis may be modest compared to other anthropogenic sources, the proximity of activity to the Arctic may help explain regional effects important for global and regional climate change". The possible implications of shipping emissions were also addressed in the work of Sand et al. (2013), who underlined that the BC aerosols would be emitted by ships directly into the Arctic planetary boundary layer with a stronger interaction with the surface (both by deposition of BC on snow and ice and by radiative and sensible heat fluxes down to the surface).

Given the aforementioned considerations and the experimental conditions of vertical profile measurements, we rephrased the conclusion referring to the impact of shipping emission at a local and regional scale.

The emission inventories are beyond the scope of the present work. We report the increased number of ships (page 19, lines 37-40) and passengers (a proxy of ship dimensions) in summer 2012 (78 days with 138 ships) relative to summer 2011 (57 days with a total of 103 ships). The obtained results can be thus considered an important phenomenon which should remain under observation in the future due to the sensitivity of the Arctic environment.

<u>SC22:</u> Page 20, L22: 'forbidden' – by who/what? What is meant by: '. And the locally formed aerosol becomes in summer' ?

Do you find any (systematic) correlation between the different vertical profiles and the measurements at the ground? E.g. for special ground conditions, one can assume (with some certainty) a particular profile? Or that when using ground measurements (which are more abundant) when comparing to models, it is not such a bad assumption?

<u>ASC22:</u> Thank for addressing this point, the sentence was incomplete due to an erroneous application of tracking changes in the word processor. We rewrote the sentence as follows: "It is important to note that SP profiles were observed for the majority of cases in summer. In summer, the long-range transport of aerosol from mid-latitudes is minor (Browse et al., 2012; Quinn et al., 2008; Stohl et al., 2006) and the locally formed aerosol becomes dominant (Giardi et al., 2015; Tunved et al., 2013; Ström et al., 2009 and 2003)".

Figure 5 clearly shows that for HO profiles ground measurements are fully representative of the vertical column (up until ~ 1 km, our vertical limit); during NG and PG profiles the ground based measurements are representative for the column up to the planetary boundary layer. DNG profiles show that ground-based measurements differ from the measurements aloft. However, the last case is influenced by secondary aerosol formation that can be easily detected by an SMPS (or similar experimental devices).

Given the above considerations, in our opinion, ground-based measurements (coupled with a proper PBL determination) are fundamental and very useful for model validation. We added these considerations in the conclusion section.

<u>Minor Comment 1 (MC1)</u>: Line 28 page 2: write the Q as a full sentence, e.g. How does the aerosol (. . . .) vary by season?

Answer to the Minor Comment 1 (AMC)1: Done

<u>MC2:</u> Page 2, line 31: 'Very pronounced' \rightarrow drop 'very'

AMC2: Done

<u>MC3:</u> Page 3, line 1: know \rightarrow known

AMC3: Thanks, corrected.

<u>MC4:</u> There are several long sentences in this paper, which makes it a bit hard to read. E.g. Page 3. Line 2-7 is one sentence over 6 lines. Could this be split in 2? Also in this sentence: 'leads' \rightarrow 'could lead'.

<u>AMC4:</u> We agree with you. We modified the sentence at page 3 and we shortened the long sentences present in the paper. Thanks for the suggestion.

<u>MC5:</u> Page 4, line 28: form \rightarrow from

AMC5: Thanks, corrected.

MC6: Page 6, line 5 double ..

<u>AMC6:</u> Thanks, corrected.

<u>MC7:</u> Page 6, line 13: operates since $2009 \rightarrow$ 'have operated'

AMC7: Thanks, corrected.

MC8: Page 6, line 21: 'during snow covered or not periods' Please rewrite.

<u>AMC8:</u> We rephrased the sentence as: "This is clearly related to a different heating of the ground related to seasonal changes of the snow covering".

<u>MC9:</u> Page 8, line: closets \rightarrow closest

AMC9: Thanks, corrected.

<u>MC10:</u> Page 11, line 2: 'Aerosol and BC and vertical profile (. . .)' Please rewrite. By vertical profile do you mean the meteorological fields? And aerosol are the size distributions?

<u>AMC10:</u> We rephrased the sentence as: "Vertical profiles of aerosol number size distribution and BC concentrations were measured to assess changes in aerosol properties within the vertical column in the Arctic region". Here the intention was to draw the reader's towards the topic of this paper: the determination of the vertical behavior of the aerosol properties (number size distribution and BC concentration) in the Arctic.

MC11: Page 11, Line 10: 'Before to introduce' .. please change

<u>AMC11:</u> We rephrased the sentence as: "Here below, the ambient conditions under which the vertical profiles were measured are briefly described".

MC12: P 11, L 24: 'Moreover, quite all measurements were conducted' quite all? You mean 'all'?

<u>AMC12:</u> Table 1 summarizes the conditions during the measuring campaign. It can be observed that, during the majority of vertical profile measurements, clear sky conditions were present. Due to your question we rephrased the sentence to clarify this point.

MC13: P11, L31: drop 'now'

AMC13: Done

MC14: P11, L34: 'Several information can be derived' please rewrite

<u>AMC14:</u> We rephrased the sentence as: Figure 3a-d (and the whole ensemble of collected data), accurately describe the vertical distribution of the aerosol and its properties in the first kilometer above Ny-Ålesund.

MC15: P11, L34: P20, L24: reasing -> rising?

AMC15: Yes, rising. Thanks, corrected.

References:

Andreae, M.O and Gelencsér A.: Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols, Atmos. Chem. Phys., 6, 3131–3148, 2006.

Barrie, L. A., and Hoff, R. M: Five years of air chemistry observations in the Arctic, Atmos. Environ., 19, 1995–2010, 1985.

Bikkina, S., Sarin, M.M.: Light absorbing organic aerosols (brown carbon) over the tropical Indian Ocean: impact of biomass burning emissions, Environ. Res. Lett., 8 (4), 044042, 2013.

Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., Deangelo, B. J., Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y. and Quinn, P. K.: Bounding the role of black carbon in the climate system: A scientific assessment, J. Geophys. Res., 118, 1–173, doi:10.1002/jgrd.50171, 2013.

Brock, C. A., Radke, L.F., Lyons, J. H., and Hobbs, P. V. : Arctic hazes in summer over Greenland and the North American Arctic, I, Incidence and origins, J. Atmos. Chem. 9, 129–148, 1989.

Browse, J., Carslaw, K.S., Arnold, S., Pringle, K.J. and Boucher O..: The scavenging processes controlling the seasonal cycle in Arctic sulphate and black carbon aerosol, Atmos. Chem. Phys., 12, 6775-6798, 2012.

Corbett, J. J., Lack, D. a., Winebrake, J. J., Harder, S., Silberman, J. a. and Gold, M.: Arctic shipping emissions inventories and future scenarios, Atmospheric Chemistry and Physics, 10(19), 9689–9704, doi:10.5194/acp-10-9689-2010, 2010.

Drinovec, L., Močnik, G., Zotter, P., Prévôt, A. S. H., Ruckstuhl, C., Coz, E., Rupakheti, M., Sciare, J., Müller, T., Wiedensohler, A., and Hansen, A. D. A.: The "dual-spot" Aethalometer: an improved measurement of aerosol black carbon with real-time loading compensation, Atmos. Meas. Tech., 8, 1965-1979, doi: 10.5194/amt-8-1965-2015, 2015.

Eckhardt, S., Hermansen, O., Grythe, H., Fiebig, M., Stebel, K., Cassiani, M., Baecklund, a. and Stohl, a.: The influence of cruise ship emissions on air pollution in Svalbard – a harbinger of a more polluted Arctic?, Atmospheric Chemistry and Physics, 13(16), 8401–8409, doi:10.5194/acp-13-8401-2013, 2013.

Eleftheriadis, K., Vratolis, S. and Nyeki, S.: Aerosol black carbon in the European Arctic: Measurements at Zeppelin station, Ny-Ålesund, Svalbard from 1998-2007, Geophysical Research Letters, 36(2), doi:10.1029/2008GL035741, 2009.

Ferrero, L., Mocnik, G., Ferrini, B. S., Perrone, M. G., Sangiorgi, G. and Bolzacchini, E.: Vertical profiles of aerosol absorption coefficient from micro-Aethalometer data and Mie calculation over Milan., Sci. Total Environ., 409(14), 2824–37, doi:10.1016/j.scitotenv.2011.04.022, 2011.

Giardi, F., Becagli, S., Traversi, R., Frosini, D., Severi, M., Caiazzo, L., Ancillotti, C., Cappelletti, D., Moroni, B., Grotti, M., Bazzano, A., Lupi, A., Mazzola, M., Vitale, V., Malandrino, M., Ferrero, L., Bolzacchini, E., Viola, A., Udisti, R.: Size distribution and ion composition of aerosol collected

at Ny Ålesund in the spring-summer field campaign 2013. Rend. Lincei, DOI 10.1007/s12210-016-0529-3, 2016.

Gilardoni,S., Vignati, E., Wilson J.: Using measurements for evaluation of black carbon modeling, Atmos. Chem. Phys., 11, 439–455, 2011.

IPCC, 2013: Climate Change 2013: The Physical Science Basis. Cambridge University Press, Cambridge, United Kingdom and New York, USA, 2013.

Jacob, D.J., Crawford, J.H., Maring, H., Clarke, A.D., Dibb, J.E., Emmons, L.K., Ferrare, R.A., Hostetler, C.A., Russell, P.B., Singh, H.B., Thompson, A.M., Shaw, G.E., McCauley, E., Pederson, J.R. and J. A. Fisher.: The Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) mission: design, execution, and first results, Atmos. Chem. Phys., 10, 5191–5212, 2010.

Kaufman, Y.J., Tanré, D., Boucher, O.: A satellite view of aerosols in the climate system. Nature, 419, 215-223, 2002.

Koren, I., Kaufman, Y.J., Remer, L.A., Martins, J.V.: Measurments of the effect of amazon smoke on inhibition of cloud formation. Scienze, 303, 1342-1345, 2004.

Koren, I., Martins, J.V., Remer, L.A., Afargan, H.: Smoke invigoration versus inhibition of clouds over the amazon, Science, 321, 946-949, 2008.

Kupiszewski, P., Leck, C., Tjernström, M., Sjogren, S., Sedlar, J., Graus, M., Müller, M., Brooks, B., Swietlicki, E., Norris, S. and Hansel, a.: Vertical profiling of aerosol particles and trace gases over the central Arctic Ocean during summer, Atmospheric Chemistry and Physics, 13(24), 12405–12431, doi:10.5194/acp-13-12405-2013, 2013.

Massabò, D., Caponi, L., Bernardoni, V., Bove, M.C., Brotto, P., Calzolai,, G., Cassola, F., Chiari, M., Fedi, M.E., Fermo, P., Giannoni, M., Lucarelli, F., Nava, S., Piazzalunga, A., Valli, G., Vecchi, R., Prati, P.: Multi-wavelength optical determination of black and brown carbon in atmospheric aerosols, Atmos. Environ., 108, 1-12, 2015.

Petzold, A., Ogren, J.A., Fiebig, M., Laj, P., Li, S.M., Baltensperger, U., Holzer-Popp, T., Kinne, S., Pappalardo, G., Sugimoto, N., Wehrli, C., Wiedensohler, A., Zhang, X.Y.: Recommendations for reporting "black carbon" measurements, Atmos. Chem. Phys., 13, 8365-8379, 2013.

Quinn, P. K., Bates, T. S., Baum, E., Doubleday, N., Fiore, a. M., Flanner, M., Fridlind, a., Garrett, T. J., Koch, D., Menon, S., Shindell, D., Stohl, a. and Warren, S. G.: Short-lived pollutants in the Arctic: their climate impact and possible mitigation strategies, Atmospheric Chemistry and Physics, 8(6), 1723–1735, doi:10.5194/acp-8-1723-2008, 2008.

Radke, L. F., Lyons, J. H., Hegg, D. A., Hobbs, P. V., and Bailey, I. H.: Airborne observations of Arctic aerosols, I, Characteristics of Arctic haze, Geophys. Res. Lett., 11, 393–396, 1984.

Ramanathan, V. and Feng, Y.: Air pollution, greenhouse gases and climate change: Global and regional perspectives, Atmos. Environ., 43(1), 37–50, doi:10.1016/j.atmosenv.2008.09.063, 2009.

Sandradewi, J., Prevot, A.H., Szidat, S., Perron, N., Rami Alfarra, M., Lanz, V., Weingartner, E., Baltensperger, U.: Using aerosol light absorption mea-surements for the quantitative determination of Wood burning and traffic emission contributions to particulate matter, Environ. Sci. Technol. 42, 3316-3323, 2008.

Schwarz, J. P., Spackman, J. R., Gao, R. S., Watts, L. a., Stier, P., Schulz, M., Davis, S. M., Wofsy, S. C. and Fahey, D. W.: Global-scale black carbon profiles observed in the remote atmosphere and compared to models, Geophysical Research Letters, 37(18), n/a–n/a, doi:10.1029/2010GL044372, 2010.

Shamjad, P.M., Tripathi, S.N., Pathak, R., Hallquist, M., Arola, A., and Bergin M.H.: Contribution of Brown Carbon to Direct Radiative Forcing over the Indo-Gangetic Plain, Environ. Sci. Technol., 49, 10474–10481, 2015.

Shaw, G. E: The Arctic haze phenomenon, BAMS, 2403–2413, 1995.

Stohl, A.: Characteristics of atmospheric transport into the Arctic troposphere, J. Geophys. Res., 111, D11306, doi:10.1029/2005JD006888, 2006.

Stohl, A., Klimont, Z., Eckhardt, S., Kupiainen, K., Shevchenko, V.P., Kopeikin, V.M. and Novigatsky, A.N.: Black carbon in the Arctic: the underestimated role of gas flaring and residential combustion emissions, Atmos. Chem. Phys., 13, 8833–8855, 2013.

Ström J., Umegard, J., Tørseth, K., Tunved, P., Hansson, H.C., Holmen, K., Wismann, V., Herber, A., König-Langlo, G.: One year of particle size distribution and aerosol chemical composition measurements at the Zeppelin Station, Svalbard, March 2000–March 2001, Phys. Chem. Earth, 28, 1181–1190, 2003.

Ström, J., Engvall, A.-C., Delbart, F., Krejci, R. and Treffeisen, R.: On small particles in the Arctic summer boundary layer: observations at two different heights near Ny-Ålesund, Svalbard, Tellus B, 61(2), 473–482, doi:10.1111/j.1600-0889.2008.00412.x, 2009.

Tunved, P., Ström, J. and Krejci, R.: Arctic aerosol life cycle: linking aerosol size distributions observed between 2000 and 2010 with air mass transport and precipitation at Zeppelin station, Ny-Ålesund, Svalbard, Atmospheric Chemistry and Physics, 13(7), 3643–3660, doi:10.5194/acp-13-3643-2013, 2013.

Weingartner, E., Saathoff, H., Schnaiter, M., Streit, N., Bitnar, B. and Baltensperger, U.: Absorption of light by soot particles: determination of the absorption coefficient by means of aethalometers, J. Aerosol Sci., 34(10), 1445–1463, doi:10.1016/S0021-8502(03)00359-8, 2003.