

Reviewer no 1

Page 14814 Line 5 “the” not needed between “of” and “aerosols” Line 8 “the” not needed between “analyze” and three” Line 24-25 “climatic processes where our understanding is insufficient”

Reply:

All corrections have been made according to the suggestions.

Page 14815 Line 1-2 “to continue in the future” no “climates” Line 4 “chemistry of the high Arctic are still very limited due to its remote location and the small number of sites where measurements are made.”

Reply:

All corrections have been made according to the suggestions.

Page 14815, Line 8 needs a few references for albedo/aerosols.
Maybe Warren, Grenfell, or Perovich references?

Reply:

“Haywood and Boucher, 2000, Reviews of Geophys.” and “Jacobson, 2001, Nature”, has been added as references on aerosol radiation effect instead of the suggested references, since the paragraph does not refer to albedo effect and black carbon, but rather to direct aerosol effect like scattering in the atmosphere.

Page 14815, Line 22 “known as “Arctic haze””

Reply:

Correction has been made according to the suggestion.

Page 14815, Line 28 “in high Arctic air” References are needed here, too How about some snow references as well so the sentence reads: “Concentrations of aerosols in high Arctic air (refs) and snow (refs) are at their highest during late winter and early spring when anthropogenic pollution is most efficiently transported from lower latitudes (e.g. Heidam, et al. 2004).”

Reply

According to the suggestions we have inserted some new references and the sentence have been changed to: “Concentrations of aerosols in high Arctic air (Polissar et al., 2001) and snow (Douglas and Sturm, 2004) are at their highest during late winter and early spring when anthropogenic pollution is most efficiently transported from lower latitudes (e.g. Heidam, et al. 2004).

Page 14816 Line 2 “of the aerosols observed from” Line 5 “and” not needed after “atmosphere”

Reply:

Corrections have been made according to the suggestions

Page 14816: Lines 7- 8 “the composition of Arctic aerosols” Before “It is important” how

about “As a consequence, “

Reply:

Corrections have been made according to the suggestion.

Page 14816: Line 11 “inorganic anion components Line 16 “transformations, and transport patterns.” Line 19 “Nord for the first time” Lines 23-24 “the year, mainly during summer”

Reply:

Corrections have been made according to the suggestions.

Page 14817 Line 2 “exposed to Eurasian air masses coming from the northern and eastern directions” Line 4 “after polar sunrise” Line 10 no “s” needed at the end of “measurement” Line 19 “yet a comparison of wind”

Reply:

Corrections have been made according to the suggestions.

Page 14819 Line 25 “are associated with this size fraction.”

Reply:

Correction has been made according to the suggestion.

Page 14820 Line s 3-10 Is there a way to express any of this in terms of time or distance over ice versus snow versus nilas ice versus frost flowers? This gets to the heart of the paper. If you look at the papers a few years ago by Kaleschke and Simpson there was some interest in the “potential” location of frost flowers (ie new ice in dynamic locations where exposed ocean water was leading to frost flowers. This versus older ice where a snow cover may impede exchange. Can this paper address the trajectories over different types of ice?

Reply:

We have added the sea ice age and ice cover on Fig.7. so that the trajectories show transport over different type of ice. We have therefore excluded Fig 8 (since this also showed the ice cover). The data of the ice age is provided by the Jet Propulsion Laboratory (JPL) and Brigham Young University (BYU) and the method is now described in the paper on page 14820. On page 14820 in line 7 after Fig. 7) we have added following text: “For spatial reference the corresponding sea ice coverage and the continents are provided as background maps, showing first year-ice (FYI) and multi-year ice (MYI). Both background maps are retrieved from data provided by the Jet Propulsion Laboratory (JPL) and Brigham Young University (BYU). The data have been obtained by the SeaWinds instruments on board the QuikSCAT satellite. SeaWinds was an active microwave radar scatterometer launched in June 1999 and operative until November 2009. Apparent brightness temperature for each scatterometer pulse was modelled from the instrument noise measurements, providing information of open water and ice (JPL, 2006). The data quality is sufficient to distinguish between first year-ice (FYI) and multi-year ice (MYI). Sea ice classification and extraction of the continents was carried out in ArcGIS (ESRI, 2011) based on gridded polar-stereographic Level 3 Sigma-0 QuikSCAT data over the Arctic Polar region.”

The Fig 7 text now reads: “Air mass back trajectories from the model HYSPLIT. The back trajectories are calculated for arrival at 50 m and 500 m.a.s.l. every 12 h. The trajectories are shown for three typical cases: (a) west of Station Nord, (b) along the coast of North

Greenland and (c) north of Station Nord. For spatial reference the corresponding sea ice coverage and the continents are provided as background maps, showing first year-ice (FYI) and multi-year ice (MYI).”

Due to these changes we have added Thomas Becker, who did this analysis, as a co-author.

Page 14820: Lines 14-30 What about non-sea salt sulfate? I know you may not have the cations to quantify this but do others or have others? This is a likely player in the sulfate world.

Reply: Na⁻ and Cl⁻ are measured as weekly sampling at St. Nord, however we will need a higher sampling resolution for comparison to our data, because the air mass will originate from many different locations over the week. As we write in the text, we believe that the coarser fraction of sulfate originates from sea salt, but the small fraction seems to be associated with long range transportation.

Page 14821 Lines 22-26 Consider breaking this into a few sentences as it is long and rambling

Reply: The sentence:” It is likely that Cl⁻ and NO₃⁻ have more local origins, since the size distribution profiles differ from SO₄⁻ and are found in the larger fractions however splitting the sampling period into three major episodes, reveal size distribution profiles more comparable for all the ions in air masses originating from west, with less exposure to sea ice.” Is now changed to:

“The size distribution profiles of Cl⁻ and NO₃⁻ differ overall from SO₄⁻ and are generally found in the large particle fraction. This indicates that Cl⁻ and NO₃⁻ have more local origins. However, splitting the sampling period into three major episodes, reveals size distribution profiles more comparable for all the ions in air masses originating from west, with less exposure to FYI.”

Page 14822 Lines 4-5 I am not sure the Journal mandates this but here and elsewhere the study references should be presented in chronological order by year Line 17 no comma needed Line 18 no comma needed after “distribution”

Reply:

This is corrected according to the suggestion.

Page 14822: Line 19 Again do you have any sense from MODIS or other imagery as to whether there was in fact open water, then nilas, then frost flowers? Perhaps a map like the one as Figure 8 but of the area immediately near the Station could be used to show the ice situation with time during sampling? Maybe four MODIS images over the three week period (maybe a little before the sampling started to provide more info) or something like that?

Reply: Figure 8 is taken out and the new Figure 7 shows multi-year ice and first-year ice as described earlier.

Page 14822: Line 24 “and Domine et al. (2004)” Line 27 “On 20 March” or “on “March 20” Line 28 Here and elsewhere it should be “sea ice” with no hyphen. In some places there is no hyphen. Be consistent.

Reply:

This is now corrected according to suggestions.

Page 14823 Lines 1-8 I wonder if saltation of snow against frost flowers could kick up or break up the frost flower crystals? Also where you have frost flowers you have or recently had a lot of brine. Winds or saltating grains over and through the brine could be a source of halides without the frost flowers playing a role. In fact, with time since the frost flowers form the amount of brine goes down.

Reply: suggestion of surface brine is now added to the text

Page 14824 Line 15 “on the sea ice are a potential source of Cl-“

Reply:

This is corrected according to suggestions

Figures 3. Use the same x-axis time/scale/format for all Figures (ie 2, 3, and S1). Figure 2 has the easiest to comprehend x-axis scale and label.

Reply: X-axis in the figures are now changed.

Figures 5 and 6. This may be an artifact of my printer but the horizontal lines are not consistently present across the Figure groups. Are the lines needed at all?

Reply: We have now removed the lines

Figure 8. This is hard to read or identify subtleties. Perhaps have a second set of Figures from the same two times but for a “zoomed” in view of the sea ice around Station Nord? This could help identify ice types/frost flowers, open water, etc.

Reply: Figure 8 is now taken out.

Reviewer no 2

Introduction: Please summarise what has been concluded from earlier work (Heidam papers) at this station. I know that sulfate and nitrate at least over several years have been reported in these papers. In particular, please describe the typical concentrations encountered in March (so we can understand whether the values in the current paper are typical), and the context of the seasonality observed in previous data, plus any insights given in previous work about sources. This will all serve to set the scene for this paper.

Reply:

We have inserted following summary of work at St. Nord in the introduction: “Studies at Station Nord have reported SO_x (SO_x = SO₂ + SO₄²⁻) and total NO₃⁻ (total NO₃⁻ = NO₃⁻_{particle} + HNO₃_{gas}) with a typical Arctic haze pattern (Heidam et al., 1999; Heidam et al., 2004, Nguyen et al., 2012). Reported weekly concentrations of total NO₃⁻ during a study period from 1990-2001 range from a few ng m⁻³ to approximately 130 ng m⁻³ in March while reported weekly mean concentrations of SO_x for the same period were up to 750 ng m⁻³ (Heidam et al., 2004). A recent study by Nguyen et al. (2012) for the period from March 2008 - February 2010 has indicated the Siberian industries as the largest sources of total NO₃⁻ and SO_x while Cl⁻ was mostly attributed to marine source. Some SO_x was also attributed to long-

range transported anthropogenic pollution whereas some NO_3^- was associated with another anthropogenic Zn source (Nguyen et al., 2012).”

Page 14817, lines 10-20 and Fig 2. I am not sure why you include Fig 2, or such an extensive discussion of wind speeds, as they are not used in the later analysis. I would recommend removing Fig 2 and replacing it with a plot of the data (see under section 3).

Reply: We find that the wind speed is important in the context of aerosol transport. Especially it seems to be important for contribution from local sources. Good meteorological measurements are rare in high Arctic. We will therefore like to keep the figure. We have also added the figure suggested under section 3, and will remove the existing figure 8, so we will end up having 8 figures in total.

Page 14818, line 13. Is this the detection limit for each stage (if so, please say so)? But I need to question how these detection limits were reached. As an example, in the original version of this paper, the SD of the blank for nitrate analysis in the filter extracts was stated as 0.07 mg/L. With 6 mL of extraction water, that means the standard deviation for analysis of a filter is 0.42 μg . With a flow rate of 30 L/minute, 3 days gives 130 m^3 of sample. Therefore one would calculate an uncertainty of at least $0.42 \times 1000 / 130 = 3 \text{ ng/m}^3$ (9 ng/m^3 for 1 day samples). And yet you give an SD 10 times lower than that. This requires some explanation (similar for other anions), or else changes in the table.

Reply

This detection limit is for each stage, which is now pointed out more clearly in the text. There was a misleading sentence regarding uncertainty in the first submitted version of the manuscript, which was then corrected and clarified in the version published in ACPD. As we tried to explain at that point, those numbers could not be used to calculate limits of detection.

The limits of detection presented here, were determined as three times the standard deviation of 8 analyses of a standard (concentration within 10 times the expected limit of detection) divided by the slope of the calibration curve.

Page 14819, lines 10-11, please clarify that these concentrations are the totals for each stage.

Reply: The concentrations are the totals for each stage, which will be clarified in the text.

Page 14819, line 13: please explain what these standard deviations are. They are clearly not the SD of an anion across the 9 samples (for Cl, the mean value is 266 and the SD is 151 ng/m^3). So what are they (indeed I can't understand how there can be a range of SDs for each anion).

Reply: This is now specified in the text. See the reply above.

Page 14819, same section. The paper needs a plot of the data. I suggest a bar chart to replace Fig 2, showing the dates of sampling, and for each sampling period, a bar showing the total concentration, either broken down into each size range as different colour steps on the bar, or as fine fraction ($< 1 \mu\text{m}$) and coarse ($> 1 \mu\text{m}$) or similar. Please also show with some symbols which type of trajectory each sampling period represents. (Fig 3 (NO_x) could easily be incorporated into the same figure, though I don't insist on that.) This will be much more

informative to the reader than the description of the data and sampling periods, which is very hard to retain in the reader's mind.

Reply: We have made a new figure, which is a bar chart, showing the dates of sampling, and for each sampling period, a bar showing the total concentration, broken down into fine fraction (< 1 μm) and coarse (> 1 μm).

Page 14820, line 21. Is Grube 2012 or submitted (also in ref list please state to what journal submitted)?

Reply: Grube et al., is submitted and this is now stated in the ref. list.

Page 14820, line 21-24. This sentence needs rewording: it is not sulfate that has a climate effect but the aerosol containing sulfate.

Reply: The sentence has been changed and reads: "SO₂—4 is by far the dominating anion during the collection period and is associated with particles having a Dg of approximately 0.32 μm . This size may have....."

Page 14821, line 24-27. I don't understand this part of the sentence, please rewrite it another way.

Reply: This sentence has now been changed. See reply to reviewer #1.

Page 14822. While frost flowers could explain the large particles, also consider the possible source from blowing snow (Yang, X., Pyle, J. A. and Cox, R. A.: Sea salt aerosol production and bromine release: Role of snow on sea ice, *Geophys. Res. Lett.*, 35, L16815, doi:10.1029/2008gl034536, 2008.) Presumably you have data on blowing snow occurrence at the station at least.

Reply: We have added this into the discussion and added Yang et al., 2008, as a reference.

Page 14823, line 5. While I agree the lab may not correctly mimic the field, remember that a similar result was found for some Arctic frost flowers (Obbard, R. W., Roscoe, H. K., Wolff, E. W. and Atkinson, H.: Frost flower surface area and chemistry as a function of salinity and temperature, *J. Geophys. Res.*, 114, D20305, doi:10.1029/2009JD012481, 2009.)

Reply: This is added to the discussion and the reference is added.

Page 14823. For nitrate there is plenty of evidence elsewhere that nitrate reacts with sea salt, and may exchange with chloride, and therefore end up partly in the size range of sea salt. For the Antarctic this is discussed in e.g. (Rankin, A. M. and Wolff, E. W.: A year-round record of size-segregated aerosol composition at Halley, Antarctica, *J. Geophys. Res.*, 108, 4775, doi:10.1029/2003JD003993, 2003.), and I am sure there must be some Arctic studies on this as well. Please consider this.

Reply: We have not been able to find this type of studies in the high Arctic, however we have added the findings of Rankin and Wolff to the discussion section. The findings of Rankin and Wolff could explain the high concentration of NO₃⁻ on the larger particles .

End of page 14823. While I have no doubt that snow photochemistry is an important source of NO_x, I don't understand the point being made here. The whole nitrate discussion is very messy and needs rethinking.

Reply: The discussion on NO_x has been changed and is now described more detailed in the text (see also last reply to review #3). We have now described the full reaction from NO_x over HNO₃ to NO₃⁻ in the aerosol.

References: Heidam 2004 has some non standard characters that have come out wrong.

Reply: This has been corrected

Table 1: What is DG? (do you mean DL?).

Reply: This is corrected to DL

Figure 8, please add a star or mark for the location of the station.

Reply: The figure is removed

Reviewer no 3

p.14815: Lines 13 – 15: Sentence needs to be edited for grammar.

Reply: This has now been edited

p. 14816, lines 3 – 7: Add more details to the description of the impact of increasing solar radiation on photochemistry in the atmosphere (e.g., formation of secondary aerosol such as SO₄) and in the snowpack (e.g., production of NO_x that can lead to production of aerosol NO₃). This discussion will then lead into results that are described later in the paper.

Reply: A description of the photochemistry in the snow including a reference (Grannas) has been added to the text.

P. 14816, line 19: change to “carried out at Station Nord FOR THE first time”

Reply: This has been changed according to the suggestion.

p. 14817, lines 4 – 20: The wind speed data is used in the discussion toward the end of the paper. I think the figure shown in the supplementary material should be put into the paper itself. Plus, the paper length is short enough to accommodate it.

Reply: We prefer to keep the wind data in the supplementary material, since one of the reviewers suggest that we don't show any wind data.

p. 14818, line 4: Was the laboratory at Greenland or were samples shipped somewhere?

Reply: The samples were shipped to Denmark. This is now stated in the text.

p. 14818, line 21: The SMPS size distributions should be shown averaged over the impactor periods. As indicated later in the paper, these size distributions were converted to mass and used to calculate total aerosol mass. It would add a great deal to the paper to show the SMPS number and derived mass size distributions in order to place the anion mass size distributions into context.

Reply: The mass calculated based on the SMPS data for the different impactor periods has now been added to the text.

p. 14819, line 4: change to “the concentrations of NO_x (NO₂ and NO) WERE estimated as : :”.

Reply: This has now been corrected

p. 14819, line 7: A time series of total (or submicron and supermicron) concentrations of each ion should be plotted and shown similar to the NO_x and NO data shown in Figure 3. Showing these time series will aid when comparing to the wind data in Figure 2 and in making connections between the measured aerosol NO₃ and the NO_x and NO concentrations.

Reply: A figure describing this has been added to the paper (se response to reviewer #1)

p. 14819, line 24: change to “are associated WITH this size fraction”.

Reply: This has now been corrected

p. 14820, lines 16 – 18: SO₄ size distributions are constant in terms of shape but not magnitude.

Reply: This has been corrected according to the suggestion

p. 14820, lines 25 – 27: Presumably the formation of SO₄ on coarse sea salt particles occurs via oxidation of SO₂. This process should be briefly described.

Reply: The process is briefly described and a reference is added

p. 14821, lines 3 – 4: The sentence starting with “Of the three analyzed anions: : :” is redundant as this has already been stated.

Reply: The sentence has been changed so it reads: “However it is still accounting for the highest mass.....”

p. 14822, lines 1 – 5: The process of chloride depletion from particles should be described in more detail including what is know about the dependence on particle size and how the process involves NO₃.

Reply: We have referred to several papers describing the process, and to be brief we have now added the gases (HNO₃ and H₂SO₄) reacting with sea spray and explained how Cl- occurs on the smaller fraction.

p. 14822, lines 6 – 7: It is stated that “the finest mode here (referring to Cl-) is found to be associated with the long-range transported SO₄”. What is the evidence for the association between Cl and SO₄? SO₄ peaks at 0.32 μm while Cl peaks at a larger diameter (0.56 μm). This difference indicates they are not internally mixed.

Reply: Looking over the data more careful we agree and the sentence have now been erased

p. 14822, line 9: I wouldn't call these Cl “profiles”. Stick with size distributions.

Reply: This is changed to size distribution

p. 14822, lines 16 – 19: explain the process of “re-fractionation of the aerosol size distributions”.

Reply: This has now been rephrased and explained (we meant redistribution)

p. 14822, line 20: What is meant by “more varying fractionation”?

Reply: We mean size distribution and this is now written instead

p. 14823, lines 26 – 27: This description of the relationship between NO_x in the snowpack and NO₃ in the aerosol should be expanded.

Reply: The description has now been expanded to describe how NO_x can oxidize to HNO₃, which will be taken up by Sea salt particles depleting Cl-.