We thank Referee #1 for her/his comments and suggestions which helped to improve the manuscript. Our response is formatted as follows:

Reviewer's comments

Author's reply

Changes to the manuscript

All page, line, section and figure numbers in bold refer to the original manuscript, all others to the revised version.

> **This work in particular presents aircraft-based measurement results of particle-phase TMA in summertime Arctic aerosols, and analyzed its origin in combing with other supporting data. Overall, the paper is well written and clearly presented, in particular, I found the results results and data interpretation were convincing, overall I recommend its publication in ACP, while a number of comments need to be addressed first, as listed below**:

(1) **The paper in total analyzed 7412 particles. Is this number covering all the partciles sampled during the measurement? Or if you actually sampled more samples, then based on what principle, you chose this number of particles?**

7412 is the total number of particles analyzed by ALABAMA during out-of-cloud sampling for measurements between 4 and 12 July. The inlet we used for particle sampling is not suitable for in-cloud measurements. Therefore, aerosol particles analyzed inside clouds (in total 763) were discarded by using measurements of an under wing FSSP (Forward Scattering Spectrometer) probe which measured cloud droplet number concentration (Leaitch et al., 2016).

(2) **Also, as you have sampled particles during 4-12 July, any changes of the properties with the changes of meterological conditions of each day?**

Prevailing high pressure influence in the Resolute Bay region leads to comparable meteorological conditions each day (during 4 – 12 July 2014).

(3) **Any difference between daytime and nighttime if flight time allows?**

We conducted no flights during nighttime. The earliest flight take-off time was at 15 UTC (local time: -5 h) and the latest flight landing time was at 23 UTC. Nevertheless, we would not expect large differences due to the small diurnal changes in angle of solar light during this time of the year.

(4) TMA-, Na/Cl-, EC-, and levoglucosan-containg particles in total occupied a bit less than 50% of all particles. However, the rest particles (>50%) were not discussed, how about their properties?

We particularly focused on the presence of particulate TMA in the summertime Arctic, including origin, size and vertical distribution. For this reason, we discussed only other particle types which either could confirm the marine origin of TMA (Na/Cl-cont. particles) or which could be clearly put into contrast to TMA in terms of size, vertical dependence and/or origin (e.g., levoglucosan-cont. particles).

However, a large fraction (29 %) of the particles prior classified as "others" contain potassium and sulfate. In the revised manuscript such particles are summarized as "K/S-containing". This particle type is introduced in Sect. 3.2. We discussed this particle type by analyzing the associated size (Fig. 5) and vertical distributions (Fig. 6) as well as the mean spectrum (Fig. 4e and Tab. 2) in Sect. 3.2.1 as follows (p.8, l.17 – p.11, l.7):

Levoglucosan, EC and potassium are known to be primarily produced from fossil fuel and biomass combustion processes (e.g., Bond et al., 2007; Simoneit, 2002; Andreae and Merlet, 2001; Simoneit et al., 1999). In particular, levoglucosan is formed via the breakdown of cellulose during biomass burning processes. The size distributions of levoglucosan- and EC-containing particles are shifted towards larger diameters compared to other particle types (Fig. 5). This result suggests these particles were exposed to chemical aging during long-range transport from biomass burning sources. K/S-containing particles are more evenly distributed across the size distribution (280 - 970 nm). Mean mass spectra of EC-, levoglucosan- and K/S-containing particles (Fig. 4c-e and Tab. 2) indicate a concurrent presence of sulfate (m/z -97/99 (HSO_4^-)), MSA (m/z -95 ($CH_3SO_3^-$)) and organic nitrogen compounds (m/z -26 (CN^-), m/z -42 (CNO^-)). Further given that the K^+ ion signals (m/z +39/41) are dominant in mean cation spectra (Fig. 4c-e), we can likely attribute these particles to a biomass burning source (e.g., Silva et al., 1999; Hudson et al., 2004; Pratt and Prather, 2009; Pratt et al., 2011). Furthermore, Zauscher et al. (2013) assigned negative ion signals at m/z -73 ($C_3H_5O_2^-$) to glyoxylic acid, which is typically present in biomass burning related SPMS spectra. Pratt et al. (2011) analyzed biomass burning particles internally mixed with oxalic acid (m/z -89 ($C_2O_4H^-$)). Both peaks are present in EC and levoglucosan mean mass spectra (Fig. 4c,d and Tab. 2). The vertical dependence in EC-containing particles is not further analyzed here due to the low statistical significance of 138 particles detected over the entire study at all altitudes. From the vertical profile of levoglucosan- and K/S-containing particles given in Fig. 6, it can be seen that their fractions increase with increasing altitude. These observations correspond to enhanced CO mixing ratios and $N_{d>250nm}$ (Fig. 3) providing further evidence for biomass burning as the source of levoglucosan- and K/S-containing particles. Despite the potential for oxidation of levoglucosan during transport, it has been previously reported as associated with biomass burning aerosol in Arctic regions (Hu et al., 2013; Fu et al., 2013, 2009).

The abstract and conclusive parts of the revised manuscript were modified accordingly.

Furthermore, the non-classified number of particles in "others" is reduced. We further added the mean spectrum of "others" in the Supplement (Fig. S7), which is mentioned in the main manuscript as follows (p.8, l.8 – 10):

The mean spectrum of the remaining 2039 particles (28 % of mass spectra analyzed by the ALABAMA), which could not be classified into one of the five particle groups outlined above, is shown in Fig. S7. For the further analysis we summarize these remaining particles in "others".

(5) As analyses on 7412 particles in fact only covers a very small portion of ambient particles, and single particle analyses in principle is not a bulk analysis, I think such limitations should be mentioned clearly. On the other hand, why the results based on a small portion of particles are representative should be justified as well.

We included discussions considering limitations by the number of analyzed particles and representativeness in the conclusive part of the revised manuscript as follows:

p.20, l.6 – p.21, l.3

SPMS measurements do not provide bulk analysis of aerosol chemical composition, therefore we can not obtain TMA mass concentrations. Nevertheless, the number of particles analyzed by the ALABAMA (> 7000) is sufficient to conduct a statistical analysis. This allows us to draw conclusions about mixing state, vertical and size distributions as well as potential emission sources of particulate TMA in summertime Arctic regions.

p. 22, l.17 - 20

This is the first study demonstrating the incorporation of amines in Arctic aerosol from inner-Arctic sources. Based on spatial and temporal limitations of our measurements, it is difficult to assess how representative our findings are of the broader Arctic region. However, recent measurements confirm the presence of particulate amines and its marine-biogenic source at another Arctic site (Alert, 82.5 °N) (Leaitch et al., 2017).

(6) The analyses regarding the sources and processes of TMA-containing particles might be discussed in combination with data for other species. Is that possible?

Thank you for this suggestion.

We discussed the ALABAMA data compared to concurrent data of the ToF-AMS (MSA mass concentrations as well as H/C and O/C ratio, see Willis et al., 2016, 2017) as well as CO mixing ratios. In particular, the comparison of particulate TMA with H/C and O/C ratios shows that TMA particles within the Arctic BL are less oxidative aged. This analysis provides further evidence for an inner-Arctic source of particulate TMA. Furthermore, the comparison with MSA mass concentration gives further indications for a marine-biogenic influence on “Non-K,NH_4-containing” TMA particles. The comparison with CO mixing ratios shows that TMA particles containing potassium and/or levoglucosan (likely from above the local BL) likely originate from biomass burning sources. The last point picks up a discussion suggested from Referee#3.

The new figures (Fig. 8,12,14) and associated discussions are included in the revised manuscript as follows:

p.13, l.10-15

Comparison of HR-ToF-AMS estimated oxygen-to-carbon (O/C) and hydrogen-to-carbon (H/C) ratios withthe ALABAMA particulate TMA fraction gives an indication of the degree of particle oxidative aging (e.g., Jimenez et al., 2009; Heald et al., 2010; Ng et al., 2011;Willis et al., 2017). Less oxygenated organics measured with the HR-ToF-AMS were present when the fraction of TMA-containing particles was high (Fig. 8a, up to 75 % in the upper left corner). This suggests that a large fraction of particulate TMA, especially within the BL (indicated with green circles in Fig. 8b), had not been subject to extensive oxidative aging.

p.16, l.2-4

Comparison between CO mixing ratios and TMA sub-types abundance (Fig. 12) shows larger fractions of "K,NH_4,S-containing" and "levoglucosan-containing" TMA particle sub-types in higher CO environments compared to "Non-K,NH_4-containing" TMA particles.

p.17, l.10-15

Furthermore, Fig. 14 indicates a positive correlation between MSA mass concentrations measured with HR-ToF-AMS and the fraction of "Non-K,NH_4-containing" TMA particles. Given that MSA can be used as an indicator for marine influence on sub-micron aerosol, we can conclude that the existence of an inner-Arctic marine-biogenic source of TMA is likely. Moreover, "Non-K,NH_4-containing" TMA particles are most abundant at the lowest altitudes (Fig. 11) and are coincident with the presence of less aged particulate organic aerosol (Fig. 8).

The abstract and conclusive parts of the revised manuscript were modified accordingly.

We further compared our measurements on 8 July with data presented in a companion paper from Burkart et al. (2017) as follows (Sect. 3.4, p.19, l. 33-35):

In addition, high organic-to-sulfate and MSA-to-sulfate ratios measured with the HR-ToF-AMS during this flight leg (see Sect. 4.3 in Burkart et al. (2017)) indicate that particle growth was driven by ocean-derived precursor gases (dimethylsulfide and organic species).

(7) Do the authors look at other amines besides TMA in the aerosols? Although TMA might be the most abundant low molecular weight amine, other amines can be present in the aerosols as well.

We investigated the presence of other alkylamines (other than TMA) and amino acid in ambient single particles. We found that none of this previously identified SPMS marker ions of other alkylamines and amino acid distinctively appear in our ambient Arctic mass spectra besides TMA.

This investigation and the associated discussion are added to the supplementary part of the revised manuscript (Sect. 4). We further added the following comment in the main manuscript (p.8, l.5-6):

Other alkylamines (other than TMA) and amino acids could not be identified (Supplement Sect. 4).

In addition to changes suggested by the referees, we did some minor changes in the revised manuscript as follows:

(1) The vertically resolved fraction of different particle types in figures 6 and 11 are now cumulative presented. This improves readability and makes the comparison between different particle types easier.
(2) Particles summarized as “others” appear now in figures 6 and 11.
(3) We unified axis notation in figures 5, 6, 10, 11, 12, 14 and 15.
(4) The colored flight tracks in Fig. 2 were partly wrongly assigned. We changed this.
(5) As described in the response to reviewer #1 comment #1, the inlet we used for aerosol sampling is not suitable for in-cloud measurements. Therefore, aerosol measurements inside clouds had been discarded. In the revised version of the manuscript, this selection had been made up for the vertical profiles (median and interquartile ranges) of $N_{d>5nm}$ and $N_{d>250nm}$ (Fig. 3).

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We thank Referee #2 for her/his comments and suggestions which helped to improve the manuscript. Our response is formatted as follows:

Reviewer's comments

Author's reply

Changes to the manuscript

All page, line, section and figure numbers in bold refer to the original manuscript, all others to the revised version.

The manuscript presents a very interesting and comprehensive study, reporting the chemical composition of Arctic summertime aerosol, focusing particularly on TMA presence, defining particle origin, size and distribution. The chemical data are combined with important meteorological parameters and elaborated by proper methodologies. The discussion is extensive, clear and effective. For these reason I suggest its publication in ACP. I have only some minor questions:

(1) Do the number of collected particles was similar at the different altitudes? (Maybe I missed this information) I think you should have a comparable number of particles collected at different altitude level in order to assure representativeness.

The vertical profiles in **Fig. 7 and 8** show information of the total number of analyzed particles in each altitude on the left hand side. Relative fractions in **Fig. 11** refer to the total number already given in **Fig. 7**. However, **Fig. 11** had been changed anyway according to Referee#3. Now relative fractions in **Fig. 11** (Fig. 11) refer to the total number of TMA-containing particles in each altitude bin.

Numbers differ in different altitude levels between approximately 70 and 1200 **(Fig. 7),** 20 and 300 **(Fig. 8)** as well as 30 and 400 (Fig. 11). Nevertheless, the total particle number in each bin is sufficient to conduct statistical approaches.

(2) Although the results are very interesting, you analyzed only 7412 particles. . .how this small number could be considered representative for summertime Arctic aerosol?

We included discussions considering limitations by the number of analyzed particles and representativeness in the conclusive part of the revised manuscript as follows:

p.20, l.6 – p.21, l.3

SPMS measurements do not provide bulk analysis of aerosol chemical composition, therefore we can not obtain TMA mass concentrations. Nevertheless, the number of particles analyzed by the ALABAMA (> 7000) is sufficient to conduct a statistical analysis. This allows us to draw conclusions about mixing state, vertical and size distributions as well as potential emission sources of particulate TMA in summertime Arctic regions.

p. 22, l.17 – 20

This is the first study demonstrating the incorporation of amines in Arctic aerosol from inner-Arctic sources. Based on spatial and temporal limitations of our measurements, it is difficult to assess how representative our findings are of the broader Arctic region. However, recent measurements confirm the presence of particulate amines and its marine-biogenic source at another Arctic site (Alert, 82.5 °N) (Leaitch et al., 2017).

(3) You discussed TMA but there are other type of amines which were not investigated and which could help in source apportionment studies. . .why didn't you consider other compounds (eg. Amino acids)?

We investigated the presence of other alkylamines (other than TMA) and amino acid in ambient single particles. We found that none of this previously identified SPMS marker ions of other alkylamines and amino acid distinctively appear in our ambient Arctic mass spectra besides TMA.

This investigation and the associated discussion are added to the supplementary part of the revised manuscript (Sect. 4). We further added the following comment in the main manuscript (p.8, l.5-6):

Other alkylamines (other than TMA) and amino acids could not be identified (Supplement Sect. 4).

(4) Why did you analyze backward trajectories only on 4,7 and 8 July? It would be better to clarify these points in the respective sections.

This had been explained in the original manuscript (caption **Fig. 12/** Fig. 15) as follows:

Only time intervals with a total number of measured particles larger than 20 were considered. Measurements within the BL on 5, 10 and 12 July did not provide any 10-min time interval with more than 20 spectra.

In addition to changes suggested by the referees, we did some minor changes in the revised manuscript as follows:

(1) The vertically resolved fraction of different particle types in figures 6 and 11 are now cumulative presented. This improves readability and makes the comparison between different particle types easier.
(2) Particles summarized as "Others" appear now in figures 6 and 11.
(3) We unified axis notation in figures 5, 6, 10, 11, 12, 14 and 15.
(4) The colored flight tracks in Fig. 2 were partly wrongly assigned. We changed this.
(5) As described in the response to reviewer #1 comment #1, the inlet we used for aerosol sampling is not suitable for in-cloud measurements. Therefore, aerosol measurements inside clouds had been discarded. In the revised version of the manuscript, this selection had been made up for the vertical profiles (median and interquartile ranges) of $N_{d>5nm}$ and $N_{d>250nm}$ (Fig. 3).

Reference:

Leaitch, W. R., Russell, L. M., Liu, J., Kolonjari, F., Toom, D., Huang, L., Sharma, S., Chivulescu, A., Veber, D., and Zhang, W.: Organic Functional Groups in the Submicron Aerosol at 82.5◦ N from 2012 to 2014, Atmospheric Chemistry and Physics Discussions, 2017, 1–38, doi:10.5194/acp-2017-511, https://www.atmos-chem-phys-discuss.net/acp-2017-511/, 2017.

We thank Referee #3 for her/his comments and suggestions which helped to improve the manuscript. Our response is formatted as follows:

Reviewer's comments

Author's reply

Changes to the manuscript

All page, line, section and figure numbers in bold refer to the original manuscript, all others to the revised version.

Köllner et al present aircraft-based single-particle mass spectrometry (SPMS) results from flights in July 2014 in the Canadian High Arctic during NETCARE. The main individual particle types observed included trimethylamine and levoglucosan-containing particles. This is an important dataset that provides the first insights into amines in the changing Arctic and greatly improves our knowledge of Arctic aerosol mixing state. Concerns and suggestions (including major concerns about particle type assignments and associated conclusions) are described below.

(1) During revision, the authors should work to improve the writing, as there are some grammar mistakes throughout the manuscript, some of which are noted below. Also, there are over 10 "paragraphs" that consist of 1-3 sentences each and do not represent full paragraphs with fully developed thoughts; these sentences should be incorporated into longer paragraphs in the revision. There are two in preparation manuscripts that are referenced in the manuscript – Bozem et al 2017 and Molleker et al 2017 – yet neither the full reference or manuscript are provided for review purposes.

We corrected grammar mistakes which Referee #3 outlined below. Furthermore, the recent manuscript had been proofread with regard on spelling and grammar by English native speakers among the co-authors. We further incorporated several sentences into longer paragraphs. Together, such changes are highlighted in red (color refers to "own revisions") in the "tracked-changes" version.

We removed both references which are in preparation. If these manuscripts will be in review process before publication of our manuscript, we will add them.

Major comments:

(2) In particular, I have concerns about the low fraction of particles that appear to have been classified/identified. While over 7000 particles were chemically analyzed in the study, the percentages discussed in Section 3.2 and shown in Figure 5 indicate that only 46% of the particles were classified/identified in the data analysis. This is very low and seems concerning in that other particle types may be missing in the analysis.

We particularly focused on the presence of particulate TMA in the summertime Arctic, including origin, size and vertical distribution. For this reason, we discussed only other particle types which either could confirm the marine origin of TMA (Na/Cl-cont.

particles) or which could be clearly put into contrast to TMA in terms of size, vertical dependence and/or origin (e.g., levoglucosan-cont. particles).

However, a large fraction (29 %) of the particles prior classified as "others" contain potassium and sulfate. In the revised manuscript such particles are summarized as "K/S-containing". This particle type is introduced in Sect. 3.2. We discussed this particle type by analyzing the associated size and vertical distributions as well as the mean spectrum (Fig. 4e and Tab. 2) in Sect. 3.2.1 as follows (p.8, l.17 – p.11, l.7):

Levoglucosan, EC and potassium are known to be primarily produced from fossil fuel and biomass combustion processes (e.g., Bond et al., 2007; Simoneit, 2002; Andreae and Merlet, 2001; Simoneit et al., 1999). In particular, levoglucosan is formed via the breakdown of cellulose during biomass burning processes. The size distributions of levoglucosan- and EC-containing particles are shifted towards larger diameters compared to other particle types (Fig. 5). This result suggests these particles were exposed to chemical aging during long-range transport from biomass burning sources. K/S-containing particles are more evenly distributed across the size distribution (280 - 970 nm). Mean mass spectra of EC-, levoglucosan- and K/S-containing particles (Fig. 4c-e and Tab. 2) indicate a concurrent presence of sulfate (m/z -97/99 (HSO_4^-)), MSA (m/z -95 ($CH_3SO_3^-$)) and organic nitrogen compounds (m/z -26 (CN^-), m/z -42 (CNO^-)). Further given that the K^+ ion signals (m/z +39/41) are dominant in mean cation spectra (Fig. 4c-e), we can likely attribute these particles to a biomass burning source (e.g., Silva et al., 1999; Hudson et al., 2004; Pratt and Prather, 2009; Pratt et al., 2011). Furthermore, Zauscher et al. (2013) assigned negative ion signals at m/z -73 ($C_3H_5O_2^-$) to glyoxylic acid, which is typically present in biomass burning related SPMS spectra. Pratt et al. (2011) analyzed biomass burning particles internally mixed with oxalic acid (m/z -89 ($C_2O_4H^-$)). Both peaks are present in EC and levoglucosan mean mass spectra (Fig. 4c,d and Tab. 2). The vertical dependence in EC-containing particles is not further analyzed here due to the low statistical significance of 138 particles detected over the entire study at all altitudes. From the vertical profile of levoglucosan- and K/S-containing particles given in Fig. 6, it can be seen that their fractions increase with increasing altitude. These observations correspond to enhanced CO mixing ratios and $N_{d>250nm}$ (Fig. 3) providing further evidence for biomass burning as the source of levoglucosan- and K/S-containing particles. Despite the potential for oxidation of levoglucosan during transport, it has been previously reported as associated with biomass burning aerosol in Arctic regions (Hu et al., 2013; Fu et al., 2013, 2009).

The abstract and conclusive parts of the revised manuscript were modified accordingly.

Furthermore, the non-classified number of particles in "others" is reduced. We further added the mean spectrum of "others" in the Supplement (Fig. S7), which is mentioned in the main manuscript as follows (p.8, l.8 – 10):

The mean spectrum of the remaining 2039 particles (28 % of mass spectra analyzed by the ALABAMA), which could not be classified into one of the five particle groups outlined above, is shown in Fig. S7. For the further analysis we summarize these remaining particles in "others".

(3) To help better inform the reader, it would be useful in the methods section to state the number of particles chemically analyzed (mass spectra produced) by ALABAMA during the study, as well as the fraction with dual-polarity mass spectra and fraction classified by the CRISP software.

We replaced information about the number of particles chemically analyzed by ALABAMA from Sect. 3.2 to Sect. 2.4 (Methods) and added the fraction with dual-polarity mass spectra (p.5, l.26-27) as follows:

In total, 7412 particles were chemically analyzed (mass spectra produced) by the ALABAMA during the study. 94 % of these mass spectra include size information. 80 % of these mass spectra have dual-polarity.

The fraction of mass spectra classified/identified into five particle types is added (p.8, l. 2-3, Sect. 3.2.) as follows:

Applying the marker method (Sect. 2.4), we classified 5373 particle mass spectra (72 % of the mass spectra analyzed by ALABAMA (Sect. 2.4)) into five distinct particle types: TMA-, Na/Cl-, EC-, levoglucosan- and K/S-containing particles.

(4) In addition, the phrasing “Chemical classes” in Table 1 is confusing, as it appears in the text that these refer to marker species, rather than particle types; the authors might consider using the phrasing “ion markers” or similar to differentiate Tables 1 and 2, or the authors might consider combining Tables 1 and 2 for improved clarity

We re-formulated “Chemical class” by “Marker species” and “Marker conditions” by “Ion markers” in Tab. 1 and in several parts in the text. Thus, the caption of Tab. 1 had been changed.

(5) Page 1 Line 6, Figure 2, Section 2.5, and Page 19 Lines 4-5: These sentences, figure, and section detail the laboratory measurement of TMA. However, the mass spectrum of TMA has already been published, using 266 nm LDI single-particle mass spectrometry, by Angelino et al. (2001), and the other literature cited in Section 2.5 (Healy et al 2015, Rehbein et al 2011) simply cite Angelino et al. (2001) for this assignment, which isn’t clear as written. Therefore, unfortunately, this does not represent a new result. The authors are encouraged to move Figure 2 and Section 2.5 to the supplemental and to remove the sentences highlighting this result in the abstract and conclusions.

Section 2.5 and **Fig. 2** had been moved to the supplementary part (Sect. 2) and corresponding sentences in the abstract and conclusions had been deleted.

We further changed the part of citations as follows (Supplement Sect. 2):

These measurements confirm previous laboratory observations using 266 nm laser desorption/ionization single-particle mass spectrometry (Angelino et al., 2001). Based on the study of Angelino et al. (2001), particulate TMA have been previously detected in ambient air using SPMS (e.g. Roth et al., 2016; Healy et al., 2015; Rehbein et al., 2011).

We further added a sentence to this topic in the main manuscript (p.6, l.6-7):

The identification of ion markers m/z +59 and +58 for TMA by Angelino et al. (2001) was confirmed by additional laboratory measurements with the ALABAMA (Supplement Sect. 2).

(6) Table 1: It would be most useful to cite SPMS lab characterization studies when possible and include the citation (e.g. Angelino et al (2001)) in the table itself, as it appears there is room. Non-SPMS literature (e.g. #6, 10-12) is not appropriate to cite here for the assignment of ion markers. No SPMS literature is cited for EC, for example. When lab study literature is available, field study literature is less appropriate, as it often simply cites other lab studies (e.g., Healy et al 2015 and Rehbein et al 2011 simply cite Angelino et al 2001 for TMA ion markers, so all three references are not necessary; Corbin et al 2012 also cites Silva et al 1999 for the levoglucosan assignment), and including field + lab literature in a comprehensive manner has not been done here (nor would it be necessary).

We used non-SPMS literature for the assignment of certain substances to particles sources (e.g., levoglucosan to biomass burning) rather than assignment of SPMS ion markers to substances. However, to make this point more clear, we put the non-SPMS literature directly behind the related subjects. We further replaced the row including listed references for the assignment of ion markers to substances next to the list of ion markers.

Furthermore, we agree that lab studies are mandatory for ion marker assignment. Nevertheless, we think that conducted field studies with particle assignment based on those lab studies are necessary to mention in order to highlight the widely-spread acceptance in the SPMS community and that also other SPMS were able to detect those peaks. In order to make a clearer differentiation between lab and field studies, we separated those in the reference row. Subsequently, the caption of Tab. 1 had been changed as follows:

Marker species (with acronyms) and associated ion markers used in this study. Further given are references (SPMS lab and field studies) used for the assignment of ion markers as well as additional comments on marker species and ions.

We keep the citations as enumerated footnotes below the table. Otherwise, the table would be to full and would contain many repetitions. Furthermore, Schmidt et al. (2017) conducted lab studies with ALABAMA including measurements of elemental carbon.

(7) Silva et al (1999) is the only study cited that shows a SPMS mass spectrum of levoglucosan. In that work, m/z -73 does not appear to be discussed as a tracer, so it is not clear where this ion attribution is coming from (could m/z -73 correspond to glyoxylic acid?).

There is no consensus in the SPMS literature about the marker ions for levoglucosan. Laboratory mass spectra (Silva et al. (1999) and our own data) show -45, -59 and -71, while others (Moffet et al. (2008) and Corbin et al. (2012)) also used -73 as a marker ion. We conducted the levoglucosan analysis with and without -73 and found that the difference is only 2 %. Thus, the main fraction of levoglucosan-containing particle spectra includes m/z -73. According to Zauscher et al. (2013), ion signals at m/z -73 can be assigned to glyoxylic acid, which likely originate from biomass burning. We decided not using m/z -73 as ion marker for levoglucosan; therefore fractions of levoglucosan-containing particle types and tables 1 and 2 changed slightly. The discussion including ion signals at m/z -73 can be found in Sect. 3.2.1 (p.10, l.1 – p.11, l.2):

Furthermore, Zauscher et al. (2013) assigned negative ion signals at m/z -73 ($C_3H_5O_2^-$) to glyoxylic acid, which is typically present in biomass burning related SPMS spectra. Pratt et al. (2011) analyzed biomass burning particles internally mixed with oxalic acid (m/z -89 ($C_2O_4H^-$)). Both peaks are present in EC and levoglucosan mean mass spectra (Fig. 4c,d and Tab. 2).

(8) For greater clarity for the non-SPMS reader, the authors are encouraged to label the "Na/Cl" particles as "sea spray aerosol", after defining and explaining the corresponding mass spectrum and citing SPMS laboratory studies of sea spray aerosol (e.g. Gard et al 1998 (Science), Prather et al 2013 (PNAS), Guasco et al 2014 (ES&T)).

Our intention here was to name the particle types by the ion fragments used for the marker method rather than name them by their potential sources. The potential source of Na/Cl-containing particles is discussed in Sect 3.2.2. We want to be consistent in the manuscript and therefore keep the particle type notation also after discussing the source.

Literature, noted above, had been added to the manuscript.

(9) In the abstract and Section 3.2.2, the authors attribute peaks at m/z -45, -59, and -71/-73 to carbohydrates (stated as levoglucosan and cellulose), but no single-particle mass spectrometry sea spray aerosol literature is cited to support these assignments and attribution to organic carbon coating sea spray aerosol. Further Prather et al and Guasco et al (noted above here), show SPMS spectra of individual sea spray particles produced in the laboratory and internally mixed with organic carbon but do not mention these peaks. The authors are encouraged to view these papers to see if greater knowledge of the Arctic sea spray aerosol can be gained through comparison to these previously published laboratory studies of sea spray aerosol produced from biologically active waters. Also, the authors might look at Cahill et al 2015 (Analytical Chem.) and Pratt et al 2009 (Nat. Geosc.), which shows ATOFMS of primary biological particles and has some of these markers.

Thank you for catching up this point and suggesting different literature to view.
We added (amongst others) Prather et al. (2013), Guasco et al. (2014), Pratt et al. (2009a) in the revised discussion.

We further re-formulated this part as follows (Sect. 3.2.2, p.12, l.12 – p. 13, l.5):

Interestingly, some of the Na/Cl-containing particles are internally mixed with different inorganics (such as magnesium and calcium) as well as oxygen- and nitrogen-containing organic compounds, as indicated by the mean spectrum in Fig. 4b and Fig. 7. It is known from previous SPMS laboratory studies on sea spray particles produced from biologically active waters that magnesium, calcium and organic nitrogen species present on inorganic salts can be arise from biological activity (Prather et al., 2013; Guasco et al., 2014). In particular, organic nitrogen fragments together with calcium, sodium and phosphate have been linked to signatures of biological species (e.g., Pratt et al., 2009a; Schmidt et al., 2017). SPMS spectra of biological particles in Pratt et al. (2009a) further indicate the occurrence of oxygen-containing organic compounds at m/z -71 ($C_3H_3O_2^-$). Laboratory studies with the ALABAMA investigating biological species (such as bacteria and pollen) also showed the existence of negative ion signals at m/z -45 ($C_2H_5O^-$), m/z -59 ($C_3H_7O_2^-$/$C_3H_9N^-$) and m/z -71 ($C_3H_3O_2^-$/$C_4H_7O^-$) in addition to the presence of phosphate and organic nitrogen compounds (Schmidt et al., 2017). Anion signals at m/z -26 ($C_2H_2^-$) and m/z -42 ($C_2H_2O^-$/$C_3H_6^-$) can be further attributed to cellulose (Schmidt et al., 2017). Moreover, Trimborn et al. (2002) reported the concurrent presence of sodium, chloride and oxygen-containing organic compounds (m/z -73 ($C_3H_5O_2^-$) and m/z -59 ($C_2H_3O_2^-$)) in ambient SPMS spectra and attributed them to organic containing sea salt particles. Other Non-SPMS studies (e.g., X-ray microscopy methods) have reported the occurrence of organic-rich (e.g., carboxylate) sea spray particles originating from microorganisms and organic compounds enriched in the sea surface microlayer in mid-latitude oceans (e.g., Quinn et al., 2014; Blanchard and Woodcock, 1980) and in Arctic regions (e.g., Wilson et al., 2015; Frossard et al., 2014; Hawkins and Russell, 2010; Russell et al., 2010). Taken together, the presence of magnesium and calcium together with nitrogen- and oxygen-containing organic species in sea spray particles suggests that such organic fragments have a marine-biogenic origin.

The associated parts in the abstract and conclusions had been removed.

(10) I would argue that the lack of negative ion mass spectra in high RH environments, particularly marine environments (e.g. Spencer et al 2008 (JGR)), is common. Therefore, I think that the authors can more strongly state that the lack of negatives is likely due to particle water suppression of negative ion formation. Recently, Guasco et al 2014 (ES&T) observed m/z 19 (H3O+) in sea spray aerosol particles that did not produce negative ions, further supporting the previous work of Neubauer et al.

We added the suggested literature in the supplementary discussion. We further added a brief comment on the lack of negative ions in the revised main manuscript (Sect. 2.4) as follows (p.5, l.27-30):

Considering the 20 % single-polarity spectra, potential reasons for the lack of negative ions are discussed in the Supplement Sect. 1. Briefly, it is likely that single-polarity spectra are produced in high relative humidity (RH) environments (Neubauer et al., 1998; Spencer et al.,2008), in particular marine environments (Guasco et al., 2014).

(11) Simoneit et al 1999 (Atmos. Environ.) showed that levoglucosan can be used as a tracer of biomass burning (BB), so it is confusing that the authors attribute the levoglucosan to pollution plumes (e.g., Page 11 Line 6) rather than BB.

This sentence had been anyway re-formulated with regard on your comment #42 (see reply to comment #42). However, biomass burning influenced air mass can be considered as polluted air within the clean summertime Arctic.

(12) Hu et al 2013 (Sci. Rep.) published a manuscript, using levoglucosan as a tracer of BB, and showing a significant impact of BB in the Arctic. The authors should be aware,

however, that levoglucosan is not a conservative tracer as it can degrade during transport (e.g. Hennigan et al 2010, GRL).

Thank you for catching this important aspect. According to Hennigan et al. (2010), the degradation of levoglucosan depends upon OH concentration. If levoglucosan is exposed to $1x10^6$ molec/ccm (OH global annual mean (Spivakovsky et al.,2000)) it has a mean lifetime of 1.1 days (range: 0.7-2.2 days) (Hennigan et al., 2010). We expect a longer lifetime for levoglucosan in sub-Arctic and high Arctic regions due to the lower OH concentration in these regions (June/July >65°N between 0.1 and $1x10^6$ molec/ccm (Spivakovsky et al.,2000 and Bahm and Khalil, 2004)).

However, we added the following sentence in the revised manuscript (Sect. 3.2.1, p. 11, l.7-8):

Despite the potential for oxidation of levoglucosan during transport, it has been previously reported as associated with biomass burning aerosol in Arctic regions (Hu et al., 2013; Fu et al., 2013, 2009).

(13) Further, the authors should consider comparing the Kdominant spectra to SPMS BB mass spectra (e.g. Silva et al 1999 (ES&T), Hudson et al 2004 (JGR), Pratt et al 2011 (ACP)), as the K-dominant particles are likely BB in origin (although they can have significant organic aerosol coating still – see Pratt and Prather 2009, ES&T).

We agree that dominant potassium ion signals most likely originate from biomass burning. This information had been added in Tab. 1 as follows:

K-dominant SPMS spectra associated (K) with BB particles

However, we identified particle types on basis of their signal intensities of certain compounds above an ion peak area threshold. This method makes it difficult to state whether a peak is dominant.

Nevertheless, mean spectra of levoglucosan-, EC- and K/S-containing particles present in all cases a dominant potassium signal. A further analysis showed that all levoglucosan particles contain potassium. We added this discussion in Sect. 3.2.1 (p.9, l.3 – p.14, l.1):

Further given that the K^+ ion signals (m/z +39/41) are dominant in mean cation spectra (Fig. 4c-e), we can likely attribute these particles to biomass burning soruce (e.g., Silva et al., 1999; Hudson et al., 2004; Pratt and Prather, 2009; Pratt et al., 2011).

Furthermore, the revised manuscript includes a broader discussion on TMA-containing particles additionally composed of potassium (see comment #17).

The abstract and conclusive parts in the revised manuscript were modified accordingly.

(14) Given the significant number of SPMS papers attributing K+ to BB, it is unclear why the authors state that "potassium in a single particle spectrum must be interpreted with caution" (Page 16, Line 11).

We deleted this sentence.

(15) It would be useful to conduct air mass trajectory analysis and look at satellite smoke/fire maps to investigate if transported BB plumes could be detected to support the observations discussed herein of the levoglucosan-containing and K-containing particles.

The focus of the paper is not long-range transport of biomass burning influenced air masses. These other particle types are added for reason of completeness and to show differences in size and vertical dependence of TMA-containing particles.

(16) I would encourage the authors to consider a broader particle class of BB particles.

We identified another particle type “K/S-containing” out of the prior “others” group (see comment #2).

We did not consider a more detailed classification of levoglucosan-containing particles with regard on potassium since: firstly, every spectrum contains potassium (see comment #13) and secondly, we think this approach is out of scope for this paper.

(17) Ge et al 2011 (Atmos Environ) shows that TMA is also emitted from BB. Given that the non-K-containing TMA particles are primarily found within the BL (so a marine source makes sense) and the K- and levoglucosan-containing TMA particles are found at higher altitudes, consideration of TMA from BB may be warranted. Currently this potential source of TMA is not mentioned

Thank you for pointing out this aspect. We discussed this additional source and explanation in the revised manuscript (Sect. 3.3) as follows (p.14, l.15 - p.16, l.6):

As can be seen in Fig. 9, a large fraction of TMA-containing particles (74 %) are additionally composed of biomass burning tracers such as potassium (67 %) and levoglucosan (7 %). According to Pöhlker et al. (2012), this internal mixture can be explained by potassium-containing particles acting as seeds for the condensation of organic material. Thus, the measured particulate TMA can be considered a secondary component that condensed on pre-existing primary particles. It is also conceivable that TMA particles containing potassium and levoglucosan are a result of biomass burning emissions (Schade and Crutzen, 1995; Ge et al., 2011a; Silva et al., 1999; Hudson et al., 2004; Pratt and Prather, 2009; Pratt et al., 2011). The size distribution of the TMA particles containing levoglucosan is shifted towards larger diameters compared to other TMA particle sub-types (Fig. 10). Moreover, Fig. 11 demonstrates that TMA particle sub-types including potassium and levoglucosan were more abundant above the BL in contrast to “Non-K,NH_4-containing” TMA particles. Comparison between CO mixing ratios and TMA sub-types abundance (Fig. 12) shows larger fractions of “K,NH_4,S-containing” and “levoglucosan-containing” TMA particle sub-types in higher CO environments compared to “Non-K,NH4-containing” TMA particles. Taken together, these results suggest that TMA particles containing levoglucosan and potassium likely originated from remote biomass burning emission sources and were transported to our measurement site.

The conclusions section and the abstract were modified accordingly.

(18) m/z -26 (CN-) and -42 (CNO-) are typically attributed to organic nitrogen fragments (e.g. Fergenson et al 2004 (Analytical Chem.) and Pratt et al 2009 (Nat. Geosc.)), rather than cyanide (Table 2 and text), in SPMS.

We replaced the attribution of m/z -26 and -42 to "cyanide" by "nitrogen-containing organics" in Tab. 2 and in the text in sections 3.2.1 and 3.2.2. We further added the following references for the assignment of m/z -26 and -42 to "nitrogen-containing organics" (Tab. 2): Pratt et al. (2009a), Silva et al. (1999), Fergenson et al. (2004), Prather et al. (2013) and Guasco et al. (2014).

(19) These are also common peaks in BB SPMS spectra (e.g. Silva et al 1999). On Page 11 Line 3, the authors cite Li et al 2000 for the assignment of cyanide, but that is not a SMPS paper.

We replaced the citation Li et al. (2000) by Silva et al. (1999).

(20) Page 11 Line 5: It is stated that there is no vertical dependence in the EC-containing particles, but do the authors have sufficient statistics to test this with only 138 EC particles detected over the entire study at all altitudes? How many at each altitude were detected?

We agree that the low number of EC-containing particles detected in different altitudes makes it difficult to interpret the data with regard on vertical dependence. We added the following comment in the revised manuscript (p.111, l.3-4):

The vertical dependence in EC-containing particles is not further analyzed here due to the low statistical significance of 138 particles detected over the entire study at all altitudes.

(21) Figure 9 and associated text: Given the issue of mostly positive-only mass spectra, the authors are encouraged to consider analysis of only the dual-polarity mass spectra here, as MSA as a negative ion would be similarly affected compared to sulfate. Without knowledge of the fraction of total mass spectra not having negative ions, this analysis is difficult to interpret.

We agree that a prior differentiation between spectra with dual- and single-polarity is necessary to discuss the presence of MSA (similar to sulfate). Thus, we added a query for the presence of dual-polarity mass spectra in the classification scheme of TMA internal mixing state (Fig. 9). Subsequently, fractions and notation of particle sub-types as well as the description of the classification scheme in Sect 3.3 changed. Furthermore, figures 10, 11 and 15 had to be adjusted.

(22) For further support of the presence of SOA, the authors might consider looking at m/z -43 (Qin et al 2012, Atmos. Environ.).

We found that approximately 51 % of TMA particle spectra include ion signals at m/z +43 and mz +27. Schmidt et al. (2017) reported that these peaks result from SOA formation from α-pinene ozonolysis in the laboratory. However, we are not sure to which extent α-pinene ozonolysis in the laboratory is comparable with Arctic marine SOA formation.

(23) It would also be useful to show the non-Kcontaining TMA average mass spectrum, as the marine source of these particles is a big highlight of this work – and yet Figure 4 shows particles with a large K peak, which may be from BB, as discussed above.

Thank you for this suggestion. We added a new figure (Fig. 13) presenting the mean spectrum of the "Non-K,NH_4-containing" TMA particles. Due to the new prior differentiation between dual- and single-polarity spectra, the TMA particle sub-type "Non-K,NH_4-containing" includes just single-polarity spectra (12 %). The missing 6 % are separately listed in dual-polarity. Since the latter group is below 7 %, we did not consider this group for the further analysis. This requires more explanation which had been done as follows (Sect. 3.3., p.16, l.8 – p.17, l.10):

This is consistent with results from particle size distributions of TMA sub-types in Fig. 10 illustrating that the fractional abundance of "Non-K,NH_4-containing" TMA particles is highest between 280 and 380 nm compared to other sub-types containing levoglucosan and/or potassium. In particular, positive ion mass spectra of the sub-type "Non-K,NH_4-containing" (12 % single-polarity (yellow box in Fig. 9) and 6 % dual-polarity (not colored in Fig. 9)) show ion signals only for carbon cluster ions and fragments of hydrocarbons (Fig. 13a,b). Due to a suppression of anion signals, likely in high RH environments (Supplement Sect. 1), we cannot state whether sulfate or MSA were present in these particles. However, the dual-polarity mean spectrum of the 6 % TMA-containing particles not including potassium and ammonium (Fig. 13b, not colored in Fig. 9) indicates the concurrent presence of sulfate or MSA. From the absence of ammonium in these TMA particles containing sulfate or MSA, we can further conclude that aminium salts were present. This result demonstrates that amines, in addition to ammonia, may take part in the neutralization of acidic aerosol. This is of particular interest considering the reduced sources of ammonia in the Arctic and the ocean as a net sink of NH3 in the summertime Canadian Arctic (Wentworth et al., 2016).

Minor Comments:

(24) Page 1 Lines 6-9: This sentence can be clarified, as the comparisons are incomplete as written.

We changed this sentence as follows:

Second, compared to particles observed aloft, TMA particles were smaller and less oxidized.

(25) Page 2 Lines 5-6: Fix sentence structure – particles are not a process.

We changed the sentence structure as follows:

Among the processes driving Arctic warming, direct and indirect radiative effects of aerosol particles play a key role.

(26) Page 2 Lines 9-10: Need references.

The listed literature at the end of the paragraph in the original manuscript refers to the last 3-4 sentences. However, this had been changed in the revised manuscript.

(27) Page 2 Line 9: Suggest changing "pollution sources within" to "pollution transport to", as the polar dome changes transport patterns, not the emissions themselves.

This sentence had been changed as follows:

First, pollution sources within the polar dome are reduced during summer, since the polar dome surface extent is smaller during summer compared to winter.

(28) Page 2 Line 10: Change "leading" to "leads".

This has been changed to "lead".

(29) Page 2 Lines 12-15: Not all of this literature corresponds to nucleation papers as implied, and many recent Arctic aerosol growth papers are also missing here. Also citations are not properly included in the sentence (formatting issue).

Thanks for pointing out the formatting issue. This has been corrected.

We agree that not all of the literature correspond to nucleation papers. The listed literature at the end of the paragraph refers to the last 3-4 sentences (see comment #26). This has been changed anyhow.

We added in the row several recent studies focusing on new particle formation and aerosol growth in Arctic region: Leaitch et al. (2013), Wentworth et al. (2015) and Croft et al. (2016b).

(30) Page 2 Lines 29-31: Sentence is missing reference.

The corresponding references can be found after the following sentence: Murphy et al., 2007.

(31) Page 3 Line 23: Fix grammar – change “parameters, several trace gases as well” to “parameters, and several trace gases, as well”

We changed this phrase as suggested.

(32) Figure 1 caption: Change “Image are” to “Image is”

This has been changed.

(33) Figure 1: Flight paths are difficult to view on small inset.

The revised manuscript includes a separate figure showing flight tracks (Fig. 2).

(34) Page 4 Line 2: Delete sentence as it is unnecessary and not incorporated in a paragraph.

This sentence has been removed.

(35) Page 5 Line 5: Give size range of PSLs used.

We added this information as follows:

By comparing these values with the velocity of manufactured monodisperse polystyrene latex particles in five sizes ranging from 190 to 800 nm, we can derive the particle vacuum aerodynamic diameter (d_{va}).

(36) Page 5 Line 8: Suggest adding the phrase “of individual particles” after “bipolar mass spectra” for clarity for the non-SPMS reader.

We added this phrase as suggested.

(37) Page 7, Lines 6-8: This statement seems to contradict the statement on page 5 line 25.

We re-formulated this sentence as follows (Supplement Sect. 2):

Despite the higher laser ablation energy deployed during NETCARE 2014 compared to lab measurements, the molecular ion of TMA (m/z +59 ($(CH_3)_3N$)) in combination with ions at m/z +58 (C_3H_8N) were produced during laser desorption/ionization (LDI) process.

(38) Page 8 Lines 6-7: Rather than “biogenic emissions”, the authors might consider the phrase “Arctic oceanic and terrestrial emissions” here, as the data is not presented to support the current statement (and the results show near equal contributions from TMA- and levoglucosan-containing particles).

This had been re-formulated as suggested.

(39) Page 8 Line 21: Word “respectively” is misplaced.

The word had been re-placed to the end of sentence.

(40) Figure 4 caption: Fix grammer – “spectrum” vs “spectra” – and repetition of this word.

We changed the caption of Fig. 4 as follows:

Bipolar mean spectra of the identified particle types: (a) TMA-containing (1688 particles = 23 %), (b) Na/Cl-containing (106 particles = 1 %), (c) EC-containing (138 particles = 2 %), (d) levoglucosan-containing (1312 particles = 18 %) and (e) K/S-containing (2129 particles = 29 %).

(41) Table 2: For greater clarity, align the text in the right-most column with the ion assignments in the third column.

This improves readability and has been changed as suggested.

(42) Page 12 Lines 1-2: This sentence is redundant – essentially the repeat of the previous sentence.

We changed this part as follows (p.11, l.3-8):

The vertical dependence in EC-containing particles is not further analyzed here due to the low statistical significance of 138 particles detected over the entire study at all altitudes. From the vertical profile of levoglucosan- and K/S-containing particles given in Fig. 6, it can be seen that their fractions increase with increasing altitude. These observations correspond to enhanced CO mixing ratios and $N_{d>250nm}$ (Fig. 3) providing further evidence for biomass burning as the source of levoglucosan- and K/S-containing particles. Despite the potential for oxidation of levoglucosan during transport, it has been previously reported as associated with biomass burning aerosol in Arctic regions (Hu et al., 2013; Fu et al., 2013, 2009).

(43) Page 12 Line 9: Sorensen et al 2005 is not appropriate here. The authors might also consider Gard et al 1998 (Science), which showed SPMS of sea spray aerosol aging.

We added this reference.

(44) Page 12 Lines 11 & 13: “NaCl2” should be “NaCl2-“ as these are ions

Thank you for catching this error. We changed this in the revised manuscript.

(45) Figure 7 seems redundant and unnecessary considering the spectrum in Figure 4.

The large ion signal of sodium in the positive mean spectrum makes it difficult to identify smaller peaks in the negative mean spectrum (Fig. 4b). Thus, we think it is necessary to present Fig. 7 to show small negative ion signals.

(46) Figure 8: This could be moved to the supplemental as it essentially repeats the information already in Figures 5 and 6, is more difficult to interpret, and has fewer particles included per bin to allow robust trends to be easily observed.

This figure had been moved to the Supplement (Fig. S8).

(47) Figure 10: To make the figure easier to read and interpret, the authors should consider showing as a fraction of TMA particles, rather than all particles, and also show the TMA particle numbers, rather than total particle numbers, as that information is already in Figure 5.

We agree that the readability of figures 10 and 11 improves by presenting data as fraction of TMA-containing particles and including the total number of TMA-containing particles. Therefore, we changed both figures as suggested.

(48) Page 16, Line 2: Do the authors have sufficient statistics in all size bins to state that these particles are only from 280-380 nm?

Every size bin includes a sufficient number of particles (>20) to conduct a statistical analysis. In terms of absolute number, “Non-K,NH_4-containing” TMA particles are most abundant between 280 and 420 nm which makes them distinguishable from other TMA particle sub-types. Considering relative fractions, the abundance of “Non-K,NH_4-containing” TMA particles is highest between 280 and 320 nm compared to the other types. We re-formulated this sentence as follows (p.16, l.8 – p.17, l.2):

This is consistent with results from particle size distributions of TMA sub-types in Fig. 10 illustrating that the fractional abundance of “Non-K,NH_4-containing“ TMA particles is highest between 280 and 380 nm compared to other sub-types containing levoglucosan and/or potassium.

(49) Page 17, Lines 4-5: If 8 July is a focus here, why did the authors choose not to show the wind data, even in the supplemental?

We added a figure illustrating the wind speed along the flight track on July 8 to the Supplement (Fig. S9).

(50) Figure 14: Please define "potential emission sensitivity" in the caption or text. The units of seconds are not clear.

We added an explanation on PES maps in Sect. 2.3 (p.5, l.20-22):

FLEXPART was operated in backward mode to provide potential emission sensitivity (PES) maps, which are the response functions to tracer releases from a receptor location. The value of the PES function is related to the particles' residence time in the output grid cell (for more details see Sect. 5 in Stohl et al. (2005) and Stohl (2006)).

Furthermore, the caption of Fig. 17 had been changed as follows:

The color-coded area presents values of the potential emission sensitivity (PES) function in a particular grid cell (Sect. 2.3).

In addition to changes suggested by the referees, we did some minor changes in the revised manuscript as follows:

(1) The vertically resolved fraction of different particle types in figures 6 and 11 are now cumulative presented. This improves readability and makes the comparison between different particle types easier.
(2) Particles summarized as "Others" appear now in figures 6 and 11.
(3) We unified axis notation in figures 5, 6, 10, 11, 12, 14 and 15.
(4) The colored flight tracks in Fig. 2 were partly wrongly assigned. We changed this.
(5) As described in the response to reviewer #1 comment #1, the inlet we used for aerosol sampling is not suitable for in-cloud measurements. Therefore, aerosol measurements inside clouds had been discarded. In the revised version of the manuscript, this selection had been made up for the vertical profiles (median and interquartile ranges) of $N_{d>5nm}$ and $N_{d>250nm}$ (Fig. 3).

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Changes/modifications according to referees #1, #2, #3 and our own revision are highlighted in yellow, green, blue and red, respectively.

Particulate trimethylamine in the summertime Canadian high Arctic lower troposphere

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Abstract. Size-resolved and vertical profile measurements of single particle chemical composition (sampling altitude range 50 - 3000 m) were conducted in July 2014 in the Canadian high Arctic during the aircraft-based measurement campaign NETCARE 2014. We deployed the single particle laser ablation aerosol mass spectrometer ALABAMA (vacuum aerodynamic diameter range approximately 200 - 1000 nm) to identify different particle types and their mixing states. On basis of the single particle analysis, we found that a significant fraction (23 %) of all analyzed particles (in total: 7412) contained trimethylamine (TMA). Two main pieces of evidence suggest that these TMA-containing particles originated from emissions within the Arctic boundary layer. First, the maximum fraction of particulate TMA occurred in the Arctic boundary layer. Second, compared to particles observed aloft, TMA particles were smaller and less oxidized. Further, air mass history analysis, associated wind data and comparison with measurements of methanesulfonic acid give evidence of a marine-biogenic influence on particulate TMA. Moreover, the external mixture of TMA-containing particles and sodium and chloride (“Na/Cl-”) containing particles, together with low wind speeds suggests particulate TMA results from secondary conversion of precursor gases released by the ocean. In contrast to TMA-containing particles originating from inner-Arctic sources, particles with biomass burning markers (such as levoglucosan and potassium) showed a higher fraction at higher altitudes, indicating long-range transport as their source. Our measurements highlight the importance of natural, marine inner-Arctic sources for composition and growth of summertime Arctic aerosol.

1 Introduction

A remarkable increase in Arctic near-surface air temperature (e.g., Chapman and Walsh, 1993; Serreze et al., 2009) has led to rather drastic changes of several climate parameters, in particular a decreasing sea ice extent of 3.5 % to 4.1 % per decade since 1979 (IPCC, 2014, with further evidence up to 2017 from the National Snow and Ice Data Center, Boulder, Colorado, https://nsidc.org). Among the processes driving Arctic warming, direct and indirect radiative effects of aerosol particles play a key role. The impact of aerosol particles on the radiation budget strongly depends on number concentration, size and chemical composition (e.g., Haywood and Boucher, 2000). Different measurements at Arctic sites show a strong annual cycle in these aerosol characteristics (e.g., Tunved et al., 2013; Leaitch et al., 2013; Engvall et al., 2008; Quinn et al., 2007; Rahn and McCaffrey, 1980). Three main processes drive the annual cycle in Arctic aerosol. First, pollution sources within the polar dome are reduced during summer, since the polar dome surface extent is smaller during summer compared to winter (e.g., Law and Stohl, 2007; Stohl, 2006; Klonecki et al., 2003; Barrie, 1986). Second, efficient wet removal processes in liquid clouds lead to a smaller condensation sink in the summertime Arctic in contrast to wintertime conditions (e.g., Croft et al., 2016a). Third, the substantial change in duration of daylight in Arctic summer leads to increased photochemical processes and increased biological activity, which further result in a higher nucleation potential (e.g., Burkart et al., 2017; Heintzenberg et al., 2017; Croft et al., 2016b; Wentworth et al., 2016; Leaitch et al., 2013; Kupiszewski et al., 2013; Kawamura et al., 2010, 1996).

To better understand the physical and chemical processes leading to a higher nucleation potential and the frequent appearance of clouds in the summertime Arctic, it is crucial to study emissions of the terrestrial and oceanic biosphere. So far, a few studies have discussed the importance of methanesulfonic acid (MSA), an oxidation product of dimethylsulfide emitted from ocean biomass, to take part in aerosol chemistry in the Arctic (e.g., Croft et al., 2016b; Leaitch et al., 2013; Sharma et al., 2012; Willis et al., 2016; Mungall et al., 2016). It is further known that marine biota also release certain gas-phase amines, such as trimethylamine (TMA), into the atmosphere (e.g., Ge et al., 2011a; Van Neste et al., 1987; Gibb et al., 1999; Facchini et al., 2008), which subsequently may contribute to aerosol chemistry. Numerous chamber, modeling and field studies at southern latitudes (e.g., Almeida et al., 2013; Kürten et al., 2017; You et al., 2014; Bergman et al., 2015; Müller et al., 2009) have focused on sources, emission rates and gas-to-particle partitioning processes of atmospheric amines. So far, this research has shown that amines may take part in aerosol chemistry in several ways. These include acid-base reactions to form aminium salts and dissolution in cloud droplets (owing to their high water-solubility) where subsequent acid-base reactions can occur in the aqueous phase (e.g., Glasoe et al., 2015; Dawson et al., 2012; Erupe et al., 2011; Ge et al., 2011b; Jen et al., 2016, 2014; Youn et al., 2015; Yu et al., 2012; Rehbein et al., 2011; Pankow, 2015). Amines compete with ammonia (NH_3) in neutralizing acidic aerosol. The base that is favoured by these reactions depends on several parameters, such as acidity of the aerosol, Henry's law coefficient and the concentration of both substances in the atmosphere (e.g., Pratt et al., 2009b; Barsanti et al., 2009). Amines further may take part in aerosol chemistry via gas-phase oxidation processes leading to the formation of species such as amides, nitramines and imines. The resulting lower volatility products can go on to form secondary organic aerosol (SOA) (e.g., Murphy et al., 2007; Ge et al., 2011b; Angelino et al., 2001).

Despite these considerable advances in studies of atmospheric amines, very little is known about their abundance in Arctic regions. Scalabrin et al. (2012) reported marine influence on amino acids in Arctic aerosol. Further particle measurements at Mace Head, Ireland have shown the presence of organic compounds, such as amines, in aerosol that originated in polar marine air masses (Dall'Osto et al., 2012). However, our knowledge about the influence of amines on Arctic aerosol number concentration, size and chemical composition remains incomplete. Based on chamber studies of enhanced sulfuric acid nucleation rates due to the presence of amines (Almeida et al., 2013), some studies have speculated that amines contribute to particle nucleation and growth in the summertime Arctic (Leaitch et al., 2013; Croft et al., 2016b). For this reason the main objective of this research is to investigate emission sources and aerosol chemistry processes of particulate TMA in the summertime Arctic. We used aircraft-based single particle chemical composition measurements conducted in the Arctic summer. In addition, we analyze concurrent data from further aerosol and trace gas instruments as well as Lagrangian modeling simulations from FLEXPART. This study provides an important opportunity to advance our understanding of the strong biological control over summertime Arctic aerosol.

2 Experimental and modeling section

2.1 Description of the sampling site and measurement platform

As one part of the NETCARE project (Network on Climate and Aerosols: Addressing Key Uncertainties in Remote Canadian Environments), aircraft-based measurements were deployed from Resolute Bay, Nunavut (Canada) during 4 - 21 July 2014. In this study, we focus on measurements made during 4 - 12 July 2014. The satellite image from 4 July 2014 shown in Fig. 1 presents sea ice and open water conditions around Resolute Bay, which can be regarded as typical during 4 - 12 July. Six research flights (around 20 flight hours) were performed during this time. Flight tracks covered altitudes from 50 to 3000 m above continental as well as marine (partly covered with sea ice) regions (Fig. 2). Three flights aimed to sample above two polynyas north of Resolute Bay. Notably, the sea ice south-east of Resolute Bay and close to the ice edge in Lancaster Sound was largely covered with melt ponds.

The instrument platform was the research aircraft Polar 6, a modified Basler BT-67 maintained by Kenn Borek and operated by the Alfred Wegener Institute for Polar and Marine Research (Herber et al., 2008). The aircraft was equipped with instruments to measure meteorological state parameters and several trace gases as well as aerosol particle number, size and chemical composition. In general, aerosol instruments were connected to a forward-facing near-isokinetic stainless steel inlet, which was followed by a 1 inch-stainless steel manifold inside the cabin. All instruments were connected to the common inlet line system with 1/4-inch stainless steel tubing. Reactive trace gases were measured via a second PTFA inlet line. Further detailed information on the inlet and sampling strategy can be found in Leaitch et al. (2016), Willis et al. (2016), Burkart et al. (2017) and Aliabadi et al. (2016b).

2.2 Instrumentation

Number concentrations of particles greater than 5 nm in diameter ($N_{d>5nm}$) were measured with a TSI 3787 water-based ultrafine condensation particle counter (UCPC). Particle size distributions of particles greater than 250 nm ($N_{d>250nm}$) were measured using an optical particle counter from GRIMM (model 1.129 Sky-OPC). Measurements of carbon monoxide (CO) were conducted with an Aerolaser ultra-fast CO monitor (model AL 5002). Sub-micron bulk aerosol composition was measured with an Aerodyne high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS). Operation of the HR-ToF-AMS aboard Polar 6 during NETCARE is described in Willis et al. (2016, 2017). State parameters and meteorological measurements

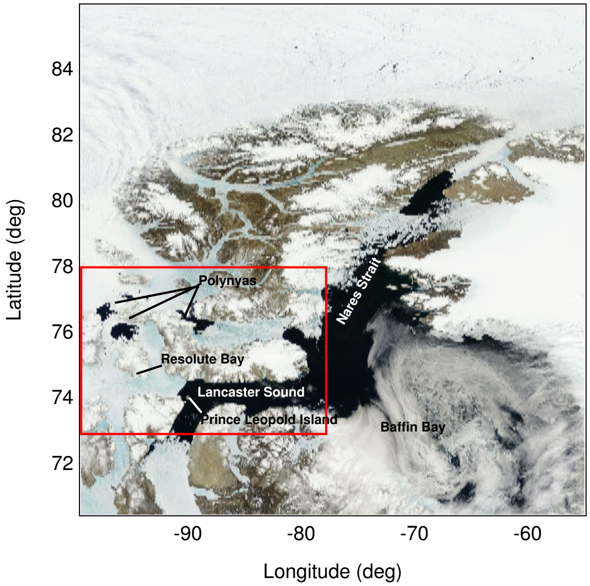

Figure 1. Satellite image (visible range from MODIS) from 4 July 2014 showing sea ice and open water conditions around Resolute Bay, in Lancaster Sound, Nares Strait and Baffin Bay. The red box indicates the region expanded with flight tracks in Fig. 2. Image is courtesy of NASA Worldview: https://worldview.earthdata.nasa.gov.

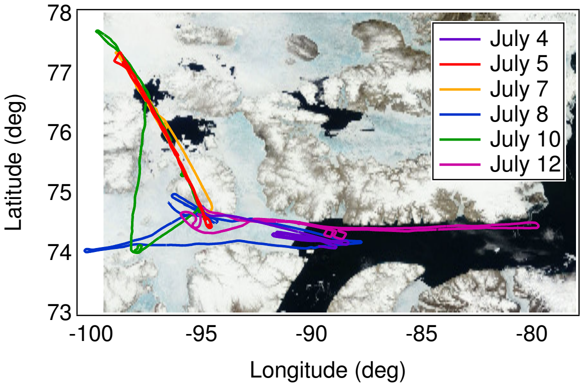

Figure 2. Satellite image (visible range from MODIS) from 4 July 2014 with a compilation of flight tracks conducted during 4 - 12 July 2014 (indicated with different colors).

were made using an AIMMS-20 from Aventech Research Inc. Detailed information on measurement principles and instrument calibrations are given in Leaitch et al. (2016) and Aliabadi et al. (2016b).

In order to provide information about the chemical composition of single aerosol particles, the ablation aerosol mass spectrometer ALABAMA (Aircraft-based Laser Ablation Aerosol Mass Spectrometer; Brands et al. (2011)) was deployed on the

Polar 6 during NETCARE 2014. The basic measurement principle of the ALABAMA is as follows: first, the particles enter the system through a constant-pressure inlet. While ambient pressure changes, this device (custom-made at the Max Planck Institute for Chemistry) maintains a constant pressure in the following aerodynamic lens by varying the volume flow rate into the instrument. A flexible orifice is either squeezed or relaxed, depending on atmospheric pressure, by a bottom and top plate that are connected to a rotor. After passing through the inlet, particles are focused into a narrow beam with the help of a Liu-type aerodynamic lens (Liu et al., 1995a, b). The focused particles are detected by two light scattering signals (using 405 nm laserdiodes and photo-multipliers) allowing the determination of the size-dependent particle velocity. By comparing these values with the velocity of manufactured monodisperse polystyrene latex particles in five sizes ranging from 190 to 800 nm, we can derive the particle vacuum aerodynamic diameter (d_{va}). Next, the particles enter the ablation and ionization region in the high-vacuum system. The particles are ablated and ionized by a single triggered laser shot (266 nm, frequency-quadrupled Nd:YAG laser). In the final step, cations and anions produced by laser ablation are guided into a bipolar Z-ToF (Z-shaped Time of Flight) mass spectrometer, which provides bipolar mass spectra of individual particles. Due to limitations of the aerodynamic lens transmission efficiency and the lower detection limit of the photo-multipliers, the ALABAMA covers a particle size range from approximately 200 to 1000 nm.

2.3 FLEXPART- Lagrangian particle dispersion model

FLEXPART (FLEXible PARTicle dispersion model (here: version 10.0)) is a Lagrangian particle dispersion model (e.g., Stohl et al., 2005). For our analysis, we used operational analysis data from the European Centre for Medium-Range Weather Forecast (ECMWF) with 0.125° spatial and three hour time resolution. FLEXPART was operated in backward mode to provide potential emission sensitivity (PES) maps, which are the response functions to tracer releases from a receptor location. The value of the PES function is related to the particles' residence time in the output grid cell (for more details see Sect. 5 in Stohl et al. (2005) and Stohl (2006)). We used such PES maps together with sea ice and open ocean coverage derived from the satellite image in Fig. 1 to determine the total residence time of the measured air mass above open water regions three days prior to sampling in altitudes up to 340 m. The model output frequency was set up to one hour and 0.125° spatial resolution.

2.4 Single particle spectra analysis

In total, 7412 particles were chemically analyzed (mass spectra produced) by the ALABAMA during the study. 94 % of these spectra include size information. 80 % of these spectra have dual-polarity. Considering the 20 % single-polarity spectra, potential reasons for the lack of negative ions are discussed in the Supplement Sect. 1. Briefly, it is likely that single-polarity spectra are produced in high relative humidity (RH) environments (Neubauer et al., 1998; Spencer et al., 2008), in particular marine environments (Guasco et al., 2014).

The software package CRISP (Concise Retrieval of Information from Single Particles) was used to perform m/z (mass to ion charge ratio) calibration of particle mass spectra and peak area integration as well as to classify particle mass spectra using ion markers for different species (Klimach, 2012). The marker method requires knowledge of certain ion markers belonging to a certain substance as well as knowledge of a certain marker threshold (ion peak area threshold). The typical fragmentation pattern of a substance due to laser ablation is crucial for defining the distinct ion markers. Fragmentation depends on laser wavelength and energy. Ion markers of many species are already well-known from laboratory measurements with the ALABAMA (Schmidt et al., 2017) and additionally from literature of other single particle mass spectrometers (SPMS) using the same ablation laser wavelength. Table 1 lists ion markers of substances used in this study to identify the external and internal mixing state of particles. The identification of ion markers m/z +59 and +58 for TMA by Angelino et al. (2001) was confirmed by additional laboratory measurements with the ALABAMA (Supplement Sect. 2). To decide whether an ion signal is present, we used an ion peak area threshold of 10 mV and 25 mV for positive and negative mass spectra, respectively. Both thresholds are chosen as a conservative measure on the basis of signal intensities of the non-occupied m/z values. Supplement Sect. 3 presents a detailed explanation of ion peak area threshold determination.

3 Results and Discussions

3.1 Meteorological conditions during the NETCARE 2014 campaign

The measurement period from 4 - 12 July 2014 was characterized by generally clear skies, calm wind speeds (Fig. 3) and occasional scattered to broken stratocumulus clouds (Leaitch et al., 2016) due to prevailing high pressure influence in the Resolute Bay region. Based on low CO mixing ratios, low aerosol number concentrations (Fig. 3) and backward trajectory analysis, air masses measured in this period experienced a weak mid-latitudinal influence and were mainly affected by local emission sources (also denoted as “Arctic air mass period” in Burkart et al. (2017)). As shown in Fig. 2, our measurements took place largely over remote areas, which are dominated by Arctic vegetation, open water regions (e.g., polynyas, Lancaster Sound) and sea ice coverage. Furthermore, seabird colonies were located close to the ice edge in Lancaster Sound and are likely a source of ammonia (Wentworth et al., 2016). Anthropogenic emissions might have affected our measurements, but are mainly related to the sparse Arctic settlements (Aliabadi et al., 2015) and can be ruled out by comparison with other tracers (e.g., CO). We can therefore expect that our observations from 4 - 12 July 2014 were mainly influenced by Arctic marine and terrestrial emissions.

As evident from vertical profiles of equivalent potential temperature (Theta_e) (Fig. 3), the mean upper boundary layer (BL) height for this measurement period was at around 340 ± 100 m. The vertical resolution of the profile in Fig. 3 (100 m) justifies the range of the mean BL height. The mean BL height within its range can be confirmed by results from an extensive study on BL height, mixing and stability during the NETCARE 2014 campaign (Aliabadi et al., 2016a). The capping temperature inversion above 390 m, inferred from values of Theta_e, represents a transport barrier for air masses between the BL and the free troposphere (FT). The BL, compared to the FT, was characterized by lower wind speeds, higher RH and enhanced $N_{d>5nm}$ in contrast to $N_{d>250nm}$, indicating an enhanced number of ultrafine particles due to nucleation in the Arctic BL. A detailed discussion on this topic is given in Burkart et al. (2017).

Table 1. Marker species (with acronyms) and associated ion markers used in this study. Further given are references (SPMS lab and field studies) used for the assignment of ion markers as well as additional comments on marker species and ions.

Marker species (Acronym)	Ion markers	References (lab/field studies)	Comments
Trimethylamine (TMA)	m/z +59 ($(CH_3)_3N^+$); +58 ($C_3H_8N^+$)	(1) / (2) (3) (4)	Additionally examined in laboratory measurements with ALABAMA (Supplement Sect. 2)
Sodium and chloride (Na/Cl)	m/z +23 (Na^+); (at least two of the following ions) +46 ($Na_2{}^+$), +62 (NaO^+), +63 ($NaOH^+$); (at least two of the following ions) +81/+83 (Na_2Cl^+), -35/-37 (Cl^-), -93/-95 ($NaCl_2{}^-$)	(5) (6) / (7) (2)	Sodium and chloride as indicators (5) (8) for sea spray particles Isobaric interference with MSA at m/z -95
Elemental carbon (EC)	(at least six of the following ions) m/z +36, +48, +60, ..., +144 ($C_{3-12}{}^+$) and/or (at least six of the following ions) m/z -36, -48, -60, ..., -144 ($C_{3-12}{}^-$)	(6) / (2) (9)	Except m/z -96 ($C_8{}^-$) due to the isobaric interference with $SO_4{}^-$
Levoglucosan	(at least two of the following ions) m/z -45 ($CHO_2{}^-$), -59 ($C_2H_3O_2{}^-$), -71 ($C_3H_3O_2{}^-$)	(10) / (11)	Levoglucosan as indicator for biomass burning (BB) particles (12)
Potassium (K)	m/z +39 (K^+)	(6) / (2) (9) (13) (14)	K-dominant SPMS spectra associated with BB particles(10) (13) (14) (15)
Ammonium (NH_4)	m/z +18 ($NH_4{}^+$)	(9) / (2) (6)	
Methanesulfonic acid (MSA)	m/z -95 ($CH_3O_3S^-$)	(16) / (17)	Isobaric interference with $PO_4{}^-$ can be excluded due to missing ion signal for $PO_3{}^-$ at m/z -79
Sulfate (S)	(at least one of the following ions) m/z -97 ($HSO_4{}^-$), -96 ($SO_4{}^-$)	(9) / (2) (6)	

The semicolon (;) used in the list of ion markers serve as "and". Given reference numbers are defined as follows: (1)Angelino et al. (2001), (2)Roth et al. (2016), (3)Healy et al. (2015), (4)Rehbein et al. (2011), (5)Prather et al. (2013), (6)Schmidt et al. (2017), (7)Sierau et al. (2014), (8)O'Dowd and de Leeuw (2007), (9)Brands et al. (2011), (10)Silva et al. (1999), (11)Corbin et al. (2012), (12)Simoneit et al. (1999), (13)Hudson et al. (2004), (14)Pratt et al. (2011), (15)Pratt and Prather (2009), (16)Silva and Prather (2000), (17) Gaston et al. (2010).

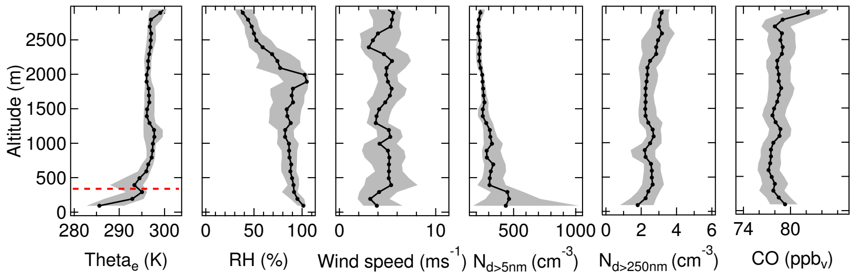

Figure 3. Vertically resolved median (black line) and interquartile ranges (gray shaded area) of the equivalent potential temperature (Theta$_e$), relative humidity (RH), wind speed, particle number concentration measured by the UCPC (N$_{d>5nm}$) and Sky-OPC (N$_{d>250nm}$) as well as CO mixing ratio (including all conducted flights from 4 - 12 July 2014). Measurements of N$_{d>250nm}$ started on 8 July due to prior technical issues. The red line depicts the derived mean upper height of the boundary layer during this measurement period (approximately 340m).

3.2 Size- and vertically resolved aerosol composition

Applying the marker method (Sect. 2.4), we classified 5373 particle mass spectra (72 % of the mass spectra analyzed by the ALABAMA (Sect. 2.4)) into five distinct particle types: TMA-, Na/Cl-, EC-, levoglucosan- and K/S-containing particles. TMA-, levoglucosan- and K/S-containing particles, with relative fractions of 23 %, 18 % and 29 %, respectively, appear to be the most prominent particle types. Other alkylamines (other than TMA) and amino acids could not be identified (Supplement Sect. 4). Furthermore, only 2 % and 1 % of all analyzed particles are assigned as EC- and Na/Cl-containing particles, respectively. The mean spectra in Fig. 4 combined with the additional ion signals listed in Tab. 2 provide an overview of the average chemical composition of each particle type. The mean spectrum of the remaining 2039 particles (28 % of mass spectra analyzed by the ALABAMA), which could not be classified into one of the five particle groups outlined above, is shown in Fig. S7. For the further analysis we summarize these remaining particles in “others”.

In order to describe the unique characteristics of TMA-containing particles compared to other particle groups, Fig. 5 and 6 depict the size and vertical distribution of each particle type, respectively. Both figures show the fractional abundance of each particle type per size and altitude bin, respectively. We show relative numbers of particles in order to eliminate the size-dependent transmission and detection efficiency of the ALABAMA (Fig. 5) and the dependence of the number of detected particles on sampling time in different altitudes (Fig. 6).

3.2.1 Levoglucosan-, EC- and K/S-containing particle types

Levoglucosan, EC and potassium are known to be primarily produced from fossil fuel and biomass combustion processes (e.g., Bond et al., 2007; Simoneit, 2002; Andreae and Merlet, 2001; Simoneit et al., 1999). In particular, levoglucosan is formed via the breakdown of cellulose during biomass burning processes. The size distributions of levoglucosan- and EC-containing particles are shifted towards larger diameters compared to other particle types (Fig. 5). This result suggests these particles were exposed to chemical aging during long-range transport from biomass burning sources. K/S-containing particles are more

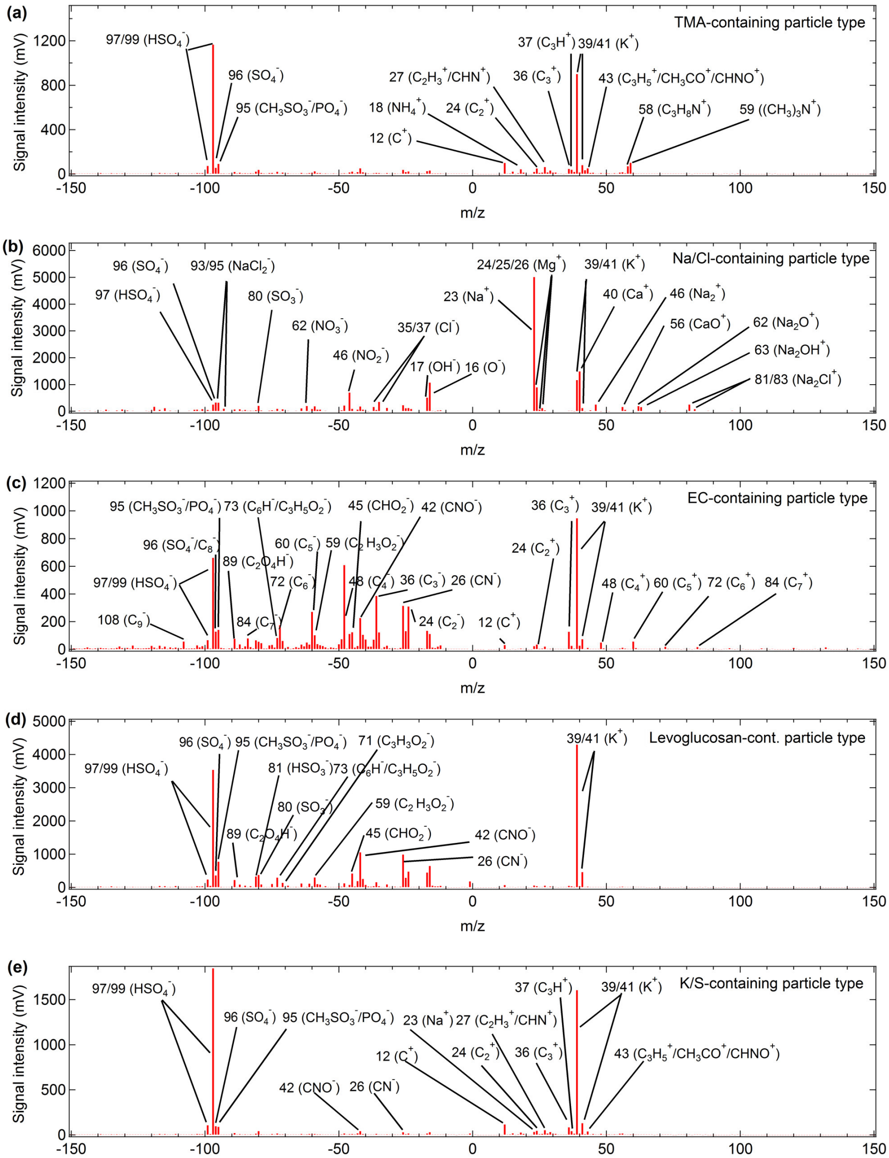

Figure 4. Bipolar mean spectra of the identified particle types: **(a)** TMA-containing (1688 particles ≘ 23 %), **(b)** Na/Cl-containing (106 particles ≘ 1 %), **(c)** EC-containing (138 particles ≘ 2 %), **(d)** levoglucosan-containing (1312 particles ≘ 18 %) and **(e)** K/S-containing (2129 particles ≘ 29 %).

evenly distributed across the size distribution (280 - 970 nm). Mean mass spectra of EC-, levoglucosan- and K/S-containing particles (Fig. 4c-e and Tab. 2) indicate a concurrent presence of sulfate (m/z -97/99 (HSO_4^-)), MSA (m/z -95 ($CH_3SO_3^-$)) and organic nitrogen compounds (m/z -26 (CN^-), m/z -42 (CNO^-)). Further given that the K^+ ion signals (m/z +39/41) are dominant in mean cation spectra (Fig. 4c-e), we can likely attribute these particles to a biomass burning source (e.g., Silva et al.,

Table 2. Overview of the obtained five particle types and their internal mixing state derived from the mean spectra in Fig. 4 and 7. Additional ion signals of sulfate (m/z -97/99 (HSO_4^-), -96 (SO_4^-)) and potassium (m/z +39/41 (K^+)) were present in every mean spectrum and have therefore not been listed here. Further given are references (SPMS lab and field studies) used for the assignment of the additional ion signals to the corresponding chemical species.

Particle type denotation	Characteristic ion signals in mean spectrum	Additional ion signals in mean spectrum	Corresponding chemical species
TMA-containing	m/z +59 ($N(CH_3)_3^+$), +58 ($NC_3H_8^+$)	m/z +18 (NH_4^+), C_{1-3}^+, +27 ($C_2H_3^+/CHN^+$), +37 (C_3H^+), +43 ($C_3H_5^+/CH_3CO^+/CHNO^+$) m/z -95 ($CH_3O_3S^-$)	ammonium carbon cluster ions hydrocarbons oxidized organics MSA
Na/Cl-containing	m/z +23 (Na^+), +46 (Na_2^+), +62 (NaO^+), +63 ($NaOH^+$), +81/+83 (Na_2Cl^+) m/z -35/-37 (Cl^-), -93/-95 ($NaCl_2^-$)	m/z +24/25/26 (Mg^+), +40 (Ca^+), +56 (CaO^+) m/z -26 (CN^-), -42 (CNO^-), -45 (CHO_2^-), -59 ($C_2H_3O_2^-$), -71 ($C_3H_3O_2^-$), -73 ($C_6H^-/C_3H_5O_2^-$), -46 (NO_2^-), -62 (NO_3^-)	magnesium(1) (2) calcium(1) nitrogen-cont. organics(1) (2) (3) (7) oxygen-cont. organics(3) (4) (5) nitrate(9)
EC-containing	C_{1-7}^+, C_{1-8}^-	m/z -26 (CN^-), -42 (CNO^-), -45 (CHO_2^-), -59 ($C_2H_3O_2^-$), -73 ($C_6H^-/C_3H_5O_2^-$), -89 ($C_2O_4H^-$), -95 ($CH_3O_3S^-$)	nitrogen-cont. organics(3) (6) (7) oxygen-cont. organics(8) (10) (11) MSA
Levoglucosan-containing	m/z -45 (CHO_2^-), -59 ($C_2H_3O_2^-$), -71 ($C_3H_3O_2^-$)	m/z -26 (CN^-), -42 (CNO^-), -73 ($C_6H^-/C_3H_5O_2^-$), -89 ($C_2O_4H^-$), -95 ($CH_3O_3S^-$)	nitrogen-cont. organics(3) (6) (7) oxygen-cont. organics(8) (10) (11) MSA
K/S-containing	m/z +39/+41 (K^+) m/z -97/99 (HSO_4^-), -96 (SO_4^-)	m/z +27 ($C_2H_3^+/CHN^+$), +37 (C_3H^+), +43 ($C_3H_5^+/CH_3CO^+/CHNO^+$) C_{1-3}^+ m/z -26 (CN^-), -42 (CNO^-), -95 ($CH_3O_3S^-$)	hydrocarbons oxidized organics carbon cluster ions nitrogen-cont. organics(3) (6) (7) MSA

Given reference numbers are defined as follows: (1)Prather et al. (2013), (2)Guasco et al. (2014), (3) Pratt et al. (2009a), (4) Schmidt et al. (2017), (5)Trimborn et al. (2002), (6)Silva et al. (1999), (7)Fergenson et al. (2004), (8)Zauscher et al. (2013), (9)Brands et al. (2011), (10)Pratt et al. (2011), (11)Sullivan and Prather (2007).

1999; Hudson et al., 2004; Pratt and Prather, 2009; Pratt et al., 2011). Furthermore, Zauscher et al. (2013) assigned negative ion signals at m/z -73 ($C_3H_5O_2^-$) to glyoxylic acid, which is typically present in biomass burning related SPMS spectra. Pratt

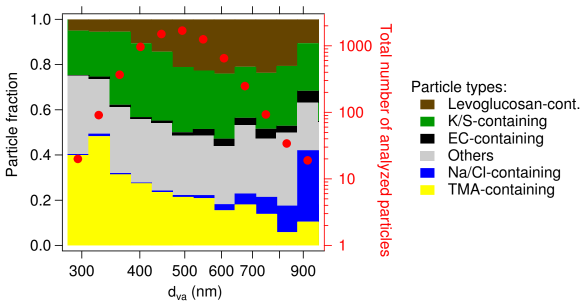

Figure 5. Cumulative size-resolved aerosol composition of the identified particle types (normalized to the total number of particles analyzed by the ALABAMA (indicated by red dots)): TMA-containing (yellow), Na/Cl-containing (blue), EC-containing (black), levoglucosan-containing (brown), K/S-containing (green) and “others” (gray).

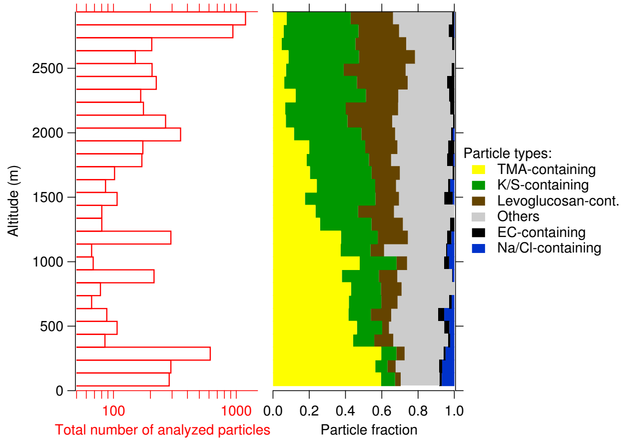

Figure 6. Cumulative vertically resolved aerosol composition of the identified particle types (normalized to the total number of particles analyzed by the ALABAMA (indicated by red bars)): TMA-containing (yellow), levoglucosan-containing (brown), Na/Cl-containing (blue), EC-containing (black), K/S-containing (green) and “others” (gray). There are in general two levels (below 340 m and above 2700 m) with enhanced number of particles analyzed by the ALABAMA, which is caused by a longer sampling time within these altitudes.

et al. (2011) analyzed biomass burning particles internally mixed with oxalic acid (m/z -89 ($C_2O_4H^-$)). Both peaks are present in EC and levoglucosan mean mass spectra (Fig. 4c,d and Tab. 2).

The vertical dependence in EC-containing particles is not further analyzed here due to the low statistical significance of 138 particles detected over the entire study at all altitudes. From the vertical profile of levoglucosan- and K/S-containing particles given in Fig. 6, it can be seen that their fractions increase with increasing altitude. These observations correspond to enhanced CO mixing ratios and $N_{d>250nm}$ (Fig. 3) providing further evidence for biomass burning as the source of levoglucosan- and K/S-containing particles. Despite the potential for oxidation of levoglucosan during transport, it has been previously reported as associated with biomass burning aerosol in Arctic regions (Hu et al., 2013; Fu et al., 2013, 2009).

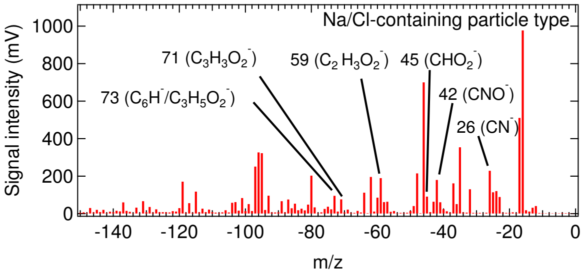

Figure 7. Expanded mean anion spectrum of 106 (1 %) Na/Cl-containing single particle spectra from Fig. 4b. Only the organics peaks are highlighted here.

3.2.2 Na/Cl-containing particle type

A number of studies have reported on the primary production of sea spray particles via bubble bursting at the sea surface (e.g., Blanchard and Woodcock, 1980; O'Dowd and de Leeuw, 2007). Na/Cl-containing particles observed in this study show particle diameters mainly larger than 600 nm and they primarily exist at lowest altitudes. Thus, the Na/Cl-containing particle type can be associated to locally emitted sea spray. The occurrence of sulfate and nitrate ion signals in the mean spectrum (Fig. 4b and Tab. 2) suggests that some particles have already been exposed to chemical aging via reactions with sulfuric and nitric acid forming nitrate and sulfate and releasing HCl to the gas phase (e.g., Gard et al., 1998; O'Dowd et al., 1999; Sorensen et al., 2005; Sierau et al., 2014). Internal mixing of Na/Cl-containing particles with MSA cannot be finally ruled out since $NaCl_2^-$ and MSA have an isobaric interference at m/z -95 (Tab. 1). However, due to the concurrent existence of other Na and Cl ion signals as well as signals at m/z -93 (isotope of $NaCl_2^-$), it is likely that ion signals at m/z -95 are largely produced by $NaCl_2^-$.

Interestingly, some of the Na/Cl-containing particles are internally mixed with different inorganics (such as magnesium and calcium) as well as oxygen- and nitrogen-containing organic compounds, as indicated by the mean spectrum in Fig. 4b and Fig. 7. It is known from previous SPMS laboratory studies on sea spray particles produced from biologically active waters that magnesium, calcium and organic nitrogen species present on inorganic salts arise from biological activity (Prather et al., 2013; Guasco et al., 2014). In particular, organic nitrogen fragments together with calcium, sodium and phosphate have been linked to signatures of biological species (e.g., Pratt et al., 2009a; Schmidt et al., 2017). SPMS spectra of biological particles presented in Pratt et al. (2009a) further indicate the occurrence of oxygen-containing organic compounds at m/z -71 ($C_3H_3O_2^-$). Laboratory studies with the ALABAMA investigating biological species (such as bacteria and pollen) also showed the existence of negative ion signals at m/z -45 ($C_2H_5O^-$), m/z -59 ($C_3H_7O_2^-/C_3H_9N^-$) and m/z -71 ($C_3H_3O_2^-/C_4H_7O^-$) in addition to the presence of phosphate and organic nitrogen compounds (Schmidt et al., 2017). Anion signals at m/z -26 ($C_2H_2^-$) and m/z -42 ($C_2H_2O^-/C_3H_6^-$) can be further attributed to cellulose (Schmidt et al., 2017). Moreover, Trimborn et al. (2002) reported the concurrent presence of sodium, chloride and oxygen-containing organic compounds (m/z -73 ($C_3H_5O_2^-$) and m/z -59 ($C_2H_3O_2^-$)) in ambient SPMS spectra and attributed them to organic containing sea salt particles. Other Non-SPMS studies (e.g., X-ray microscopy methods) have reported the occurrence of organic-rich (e.g., carboxylate) sea spray particles originating from microorganisms and organic compounds enriched in the sea surface microlayer in mid-latitude oceans (e.g., Quinn et al., 2014; Blanchard and Woodcock, 1980) and in Arctic regions (e.g., Wilson et al., 2015; Frossard et al., 2014; Hawkins and Russell, 2010; Russell et al., 2010). Taken together, the presence of magnesium and calcium together with nitrogen- and oxygen-containing organic species in sea spray particles suggests that such organic fragments have a marine-biogenic origin.

3.2.3 TMA-containing particle type

TMA-containing particles have several characteristics that are contrary to the other particles types. The size distribution of TMA-containing particles is shifted towards smaller diameters (Fig. 5) and the fractional abundance increases with decreasing altitude (Fig. 6). In addition, TMA-containing particles detected within the BL are smaller compared to particles observed aloft (Fig. S8). Comparison of HR-ToF-AMS estimated oxygen-to-carbon (O/C) and hydrogen-to-carbon (H/C) ratios with the ALABAMA particulate TMA fraction gives an indication of the degree of particle oxidative aging (e.g., Jimenez et al., 2009; Heald et al., 2010; Ng et al., 2011; Willis et al., 2017). Less oxygenated organics measured with the HR-ToF-AMS were present when the fraction of TMA-containing particles was high (Fig. 8a, up to 75 % in the upper left corner). This suggests that a large fraction of particulate TMA, especially within the BL (indicated with green circles in Fig. 8b), had not been subject to extensive oxidative aging. According to these results together with the existence of a stable stratified BL (Fig. 3), we can infer that particulate TMA present within the Arctic BL originated from inner-Arctic sources. Possible inner-Arctic sources of TMA, referring to Ge et al. (2011a), are oceanic phytoplankton biomass or other marine organisms and various human activities (e.g., waste incineration, vehicle exhaust, residential heating). Gaseous TMA emissions may then take part in aerosol chemistry in several ways including acid-base reactions, oxidation processes, dissolution in cloud droplets and nucleation (e.g., Ge et al., 2011a, b; Rehbein et al., 2011; Erupe et al., 2011; Murphy et al., 2007; Angelino et al., 2001).

The mean spectrum of TMA-containing particles (Fig. 4a) shows no indications that further N-containing compounds (such as amine oxidation products, e.g., amides, nitramines and imines) other than TMA (with specific ion signals at m/z +59 and +58) were present on these particles. Figure 4a and Table 2 further illustrate an internal mixing of sulfate and TMA, which indicates that aminium sulfate salts may be present (e.g., Murphy et al., 2007; Barsanti et al., 2009; Smith et al., 2010). We can therefore hypothesize that the formation of particulate TMA was accompanied with acid-base reactions including TMA, sulfuric and methanesulfonic acid (e.g., Facchini et al., 2008). Rehbein et al. (2011) reported enhanced gas-to-particle partitioning of TMA by dissolution in cloud/fog droplets and subsequent formation of aminium salts. Thus, it is further possible, due to the occasional presence of low-level clouds (Leaitch et al., 2016), that the formation of TMA-containing particles was favored by pre-existing wet and acidic particles.

3.3 Internal mixing state of TMA-containing particles

The internal mixing state of TMA-containing particles was further classified by applying the marker method introduced in Sect. 2.4 and Tab. 1 for compounds that are apparent in the mean spectrum (Fig. 4a and Tab. 2), such as potassium (K), ammonium (NH_4), MSA and sulfate (S). The diagram in Fig. 9 illustrates the classification algorithm as follows: an upper branch always

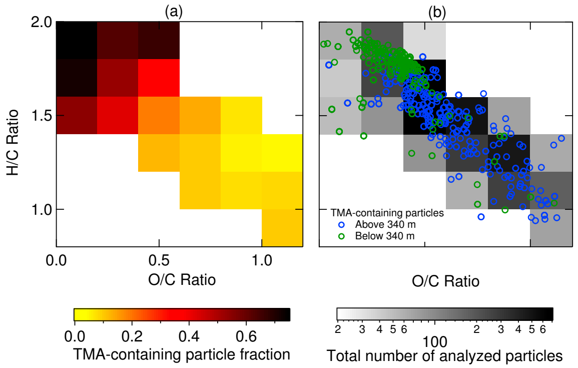

Figure 8. Comparison between the HR-ToF-AMS estimated oxygen-to-carbon (O/C) and hydrogen-to-carbon (H/C) ratios colored by: **(a)** TMA-containing particle fraction and **(b)** total number of analyzed particles (gray to black) as well as the presence of particulate TMA above (blue circles) and below (green circles) 340 m (mean upper BL height, Sect. 3.1).

refers to a positive response (“yes”) for whether different ion markers are present in spectra or not; a lower branch shows the opposite answer (“no”). Besides the substances that already appeared in the mean spectrum of TMA-containing particles here TMA-containing spectra are also viewed based on the concurrent existence of levoglucosan, Na/Cl and EC. We did not consider in detail the concurrent existence of carbon cluster ions (m/z +12, +24, ..), different hydrocarbons (m/z +27 and +37) and oxidized organics (m/z +43) since 90 % of all TMA-containing particles contain at least one of these ion signals. The classification of the TMA-containing particle type is further based on an initial differentiation between dual- and single-polarity mass spectra. As can be seen in Fig. 9, 28 % of TMA-containing particle spectra lack negative ions. Consequently, we cannot state if species producing anions (such as MSA and sulfate) were present on these particles. Potential reasons for the lack of negative ions are discussed in the Supplement Sect. 1. Particle sub-group notation is based on the existence or non-existence of different species in TMA-containing particles. For reasons of clarity, particle types with less than 7 % fractional abundance (corresponding to a total number of less than 118 particles) are not explicitly considered in this analysis, but are summarized as “others”. Following the categorization in Fig. 9, five groups of different internal mixing states arise: “K,NH_4,MSA,S-”, “K,NH_4,S-”, “K-”, “Non-K,NH_4-” and “levoglucosan-containing” particles. These five TMA particle sub-types will be divided into those containing biomass burning tracers (such as levoglucosan and potassium) and those not containing these tracers.

As can be seen in Fig. 9, a large fraction of TMA-containing particles (74 %) are additionally composed of biomass burning tracers such as potassium (67 %) and levoglucosan (7 %). According to Pöhlker et al. (2012), this internal mixture can be explained by potassium-containing particles acting as seeds for the condensation of organic material. Thus, the measured particulate TMA can be considered a secondary component that condensed on pre-existing primary particles. It is also conceivable that TMA particles containing potassium and levoglucosan are a result of biomass burning emissions (Schade and Crutzen, 1995; Ge et al., 2011a; Silva et al., 1999; Hudson et al., 2004; Pratt and Prather, 2009; Pratt et al., 2011). The size distribution of the TMA particles containing levoglucosan is shifted towards larger diameters compared to other TMA particle sub-types.

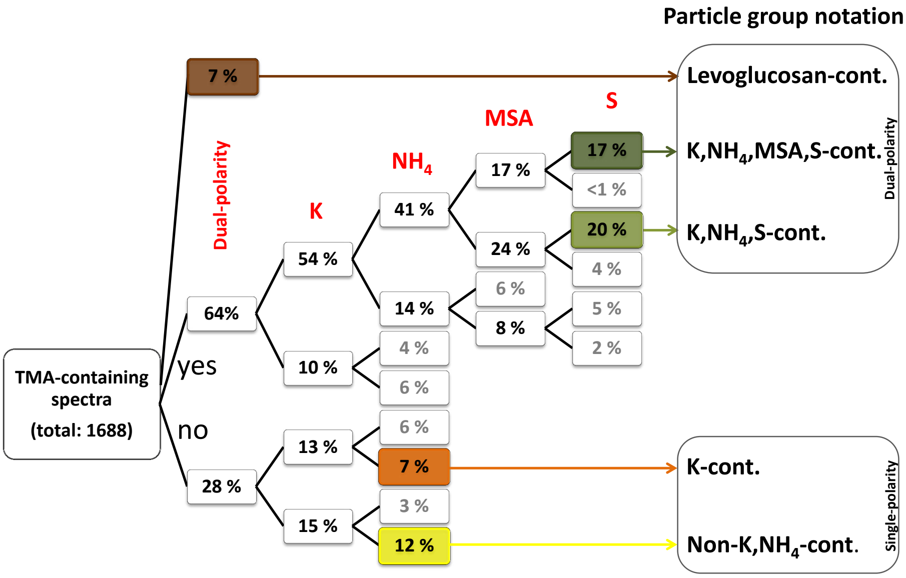

Figure 9. Classification of TMA-containing particles on basis of their different internal mixing states. Each branch describes the existence or non-existence of several substances (potassium (K), ammonium (NH_4), MSA and sulfate) on TMA-containing particles with relative abundances normalized to the occurrence of 1688 TMA-containing particles. An initial query regarding the existence of dual-polarity spectra is included. Based on this classification four TMA-containing particle sub-types arise (colored boxes with relative fractions): “K,NH_4,MSA,S-”, “K,NH_4,S-”, “K-” and “Non-K,NH_4-containing”. We further considered an internal mixing of particulate TMA with levoglucosan (7 %), Na/Cl and EC (not listed here), whereby the latter two types with relative fractions of less than 1 % are negligible for the further analysis. Gray-shaded numbers indicate groups with relative fractions of less than 7 % that are not further considered.

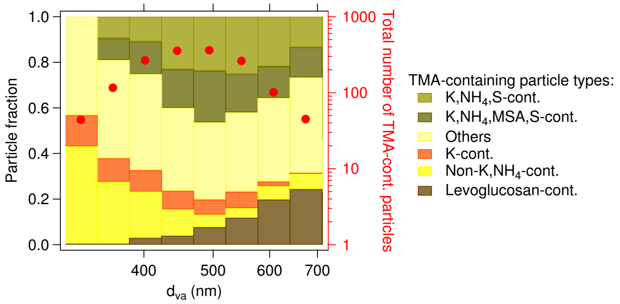

Figure 10. Cumulative size-resolved aerosol composition of TMA-containing particle sub-types (normalized to the total number of TMA-containing particles (indicated by red dots)): “K,NH_4,MSA,S-containing” (dark green), “K,NH_4,S-containing” (light green), “K-containing” (orange), “Non-K,NH_4-containing” (yellow), “levoglucosan-containing” (brown) and “others” (light yellow).

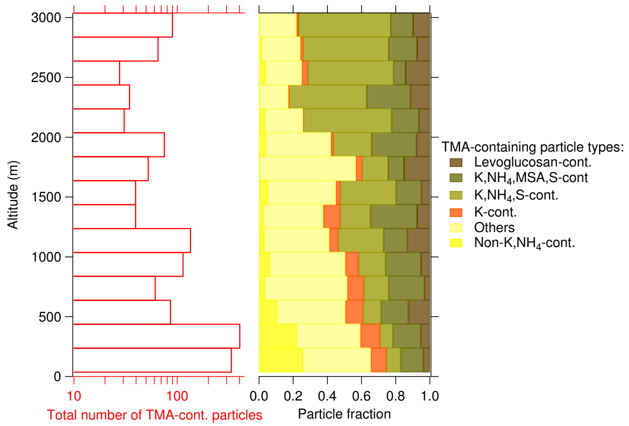

Figure 11. Cumulative vertically resolved aerosol composition of TMA-containing particle sub-types (normalized to the total number of TMA-containing particles (indicated by red bars)): "K,NH$_4$,MSA,S-containing" (dark green), "K,NH$_4$,S-containing" (light green), "K-containing" (orange), "Non-K,NH$_4$-containing" (yellow), "levoglucosan-containing" (brown) and "others" (light yellow).

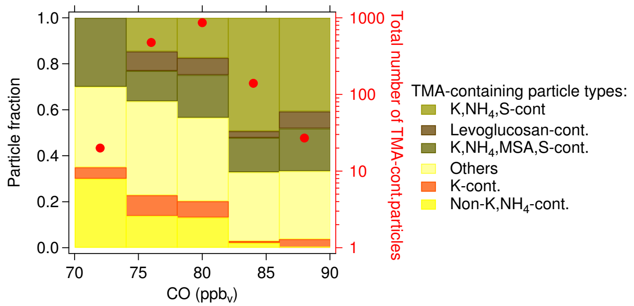

Figure 12. CO measurements compared with cumulative fraction of TMA-containing particle sub-types (normalized to all TMA-containing particles (indicated by red dots)): "K,NH$_4$,MSA,S-containing" (dark green), "K,NH$_4$,S-containing" (light green), "K-containing" (orange), "Non-K,NH$_4$-containing" (yellow), "levoglucosan-containing" (brown) and "others" (light yellow).

(Fig. 10). Moreover, Fig. 11 demonstrates that TMA particle sub-types including potassium and levoglucosan were more abundant above the BL in contrast to "Non-K,NH$_4$-containing" TMA particles. Comparison between CO mixing ratios and TMA sub-types abundance (Fig. 12) shows larger fractions of "K,NH$_4$,S-containing" and "levoglucosan-containing" TMA particle sub-types in higher CO environments compared to "Non-K,NH$_4$-containing" TMA particles. Taken together, these results suggest that TMA particles containing levoglucosan and potassium likely originated from remote biomass burning emission sources and were transported to our measurement site.

Another large fraction (25 %, Fig. 9) of particulate TMA is neither internally mixed with potassium nor with any other tracer of biomass burning. This result suggests that these TMA-containing particles resulted from SOA formation. This is consistent with results from particle size distributions of TMA sub-types in Fig. 10 illustrating that the fractional abundance of "Non-

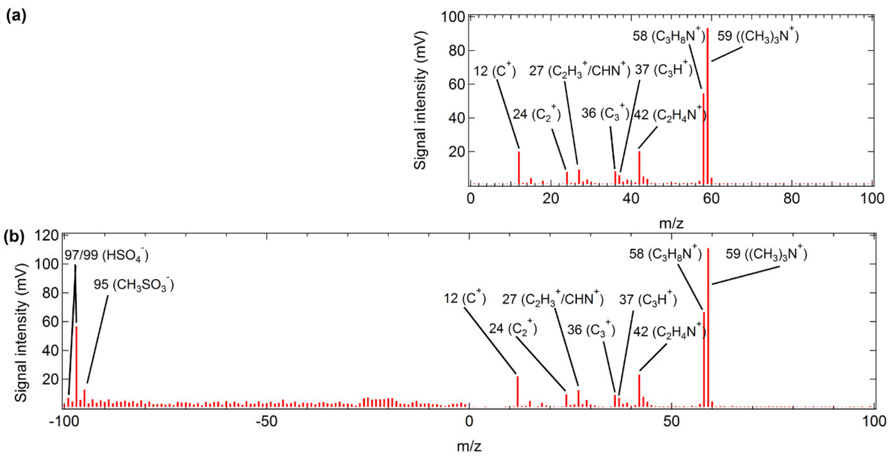

Figure 13. Mean spectra of “Non-K,NH_4-containing” TMA particle sub-type: **(a)** Single-polarity particle mass spectrum (12 %, yellow box in Fig. 9), **(b)** Dual-polarity particle mass spectrum (6 %, not colored in Fig. 9).

K,NH_4-containing” TMA particles is highest between 280 and 380 nm compared to other sub-types containing levoglucosan and/or potassium. In particular, positive ion mass spectra of the sub-type “Non-K,NH_4-containing” (12 % single-polarity (yellow box in Fig. 9) and 6 % dual-polarity (not colored in Fig. 9)) show ion signals only for carbon cluster ions and fragments of hydrocarbons (Fig. 13a,b). Due to a suppression of anion signals, likely in high RH environments (Supplement Sect. 1), we cannot state whether sulfate or MSA were present in these particles. However, the dual-polarity mean spectrum of the 6 % TMA-containing particles not including potassium and ammonium (Fig. 13b, not colored in Fig. 9) indicates the concurrent presence of sulfate or MSA. From the absence of ammonium in these TMA particles containing sulfate or MSA, we can further conclude that aminium salts were present. This result demonstrates that amines, in addition to ammonia, may take part in the neutralization of acidic aerosol. This is of particular interest considering the reduced sources of ammonia in the Arctic and the ocean as a net sink of NH_3 in the summertime Canadian Arctic (Wentworth et al., 2016). Furthermore, Fig. 14 indicates a positive correlation between MSA mass concentrations measured with HR-ToF-AMS and the fraction of “Non-K,NH_4-containing” TMA particles. Given that MSA can be used as an indicator for marine influence on sub-micron aerosol, we can conclude that the existence of an inner-Arctic marine-biogenic source of TMA is likely. Moreover, “Non-K,NH_4-containing” TMA particles are most abundant at the lowest altitudes (Fig. 11) and are coincident with the presence of less aged particulate organic aerosol (Fig. 8). Taken together, the characteristics of the “Non-K,NH_4-containing” TMA particle sub-type suggest that gaseous TMA

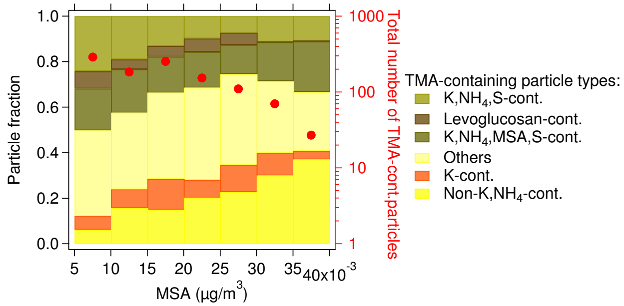

Figure 14. MSA concentrations measured with the HR-ToF-AMS compared with cumulative fraction of TMA-containing particle sub-types (normalized to all TMA-containing particles (indicated by red dots)): "K,NH4,MSA,S-containing" (dark green), "K,NH4,S-containing" (light green), "K-containing" (orange), "Non-K,NH4-containing" (yellow), "levoglucosan-containing" (brown) and "others" (light yellow).

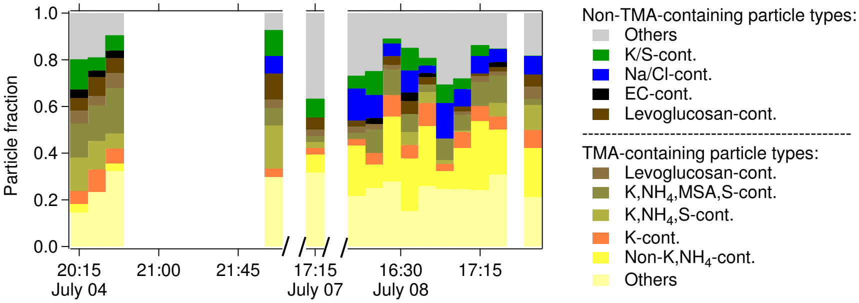

Figure 15. Temporally resolved aerosol composition of the identified non-TMA-containing particle types (normalized to the total number of particles analyzed by the ALABAMA): Na/Cl-cont. (blue), EC-cont. (black), levoglucosan-cont. (brown), K/S-cont. (green) and "others" (gray) as well as TMA-containing particle sub-types (normalized to the total number of particles analyzed by the ALABAMA): "levoglucosan-cont." (light brown), "K,NH4,MSA,S-cont." (dark green), "K,NH4,S-cont." (light green), "K-cont." (orange), "Non-K,NH4-cont." (yellow) and "others" (light yellow). Fractional abundances of the particle types were calculated for 10 minute time intervals. Only time intervals with at least 20 measured particles were considered. Measurements within the BL on 5, 10 and 12 July did not provide any 10 minute time interval with more than 20 spectra.

emissions from inner-Arctic sources (likely marine-biogenic) act as precursor for the formation of SOA within the summertime Arctic BL.

3.4 Source apportionment analysis of TMA-containing particles

This section will further explore potential emission sources of TMA in the Arctic BL. Thus, the following analysis was restricted to measurements below 340 m (mean upper BL height, Sect. 3.1). Figure 15 shows the temporal distribution of non-TMA-containing particles (such as Na/Cl-, EC- , levoglucosan- and K/S-containing) and TMA-containing sub-types. Figure 16 depicts the spatially resolved fraction of TMA-containing particles below 340 m (left panel) as well as the measured wind direction (right panel) for measurements on 4, 7 and 8 July. We further used three-day FLEXPART backward simulations (Sect. 2.3) for air mass history analysis of the three measurement legs (Fig. 17) to understand the source regions of TMA-containing particles.

Potential emission sensitivity (PES) maps combined with sea ice coverage (Fig. 1) show that air masses measured on 4 and 7 July spent less than one hour and around seven hours, respectively, in the previous three days in regions of open water (polynyas north of Resolute Bay and Nares Strait). On both days the air was mainly advected above sea ice and snow covered regions north of Resolute Bay (Fig. 17 compared with Fig. 1). The prevailing wind direction on 4 and 7 July along the flight tracks (Fig. 16) is from the north and east and therefore consistent with FLEXPART backward simulations (Fig. 17). From measurements on 4 and 7 July it is not possible to attribute TMA emissions to marine-biogenic or anthropogenic sources (e.g., vehicle exhaust, residential heating and waste incineration emissions in Resolute Bay). A more detailed air mass history analysis was carried out on observations from 8 July.

The case of 8 July provides further evidence for a marine-biogenic influence on TMA-containing particles through secondary processes. The prevailing wind direction along the presented flight leg is from the east (Fig. 16) with low wind speeds up to a maximum of 7 m/s (Fig. S9). The fraction of TMA-containing particles decreases with a shift to a more southerly wind direction (yellow to green colors, Fig. 16). The highest fractional abundance of particulate TMA was measured close to the ice edge (Fig. 16) at low wind speeds close to zero m/s (Fig. S9). Thus, the ice edge in the western section of Lancaster Sound where the highest surface phytoplankton production rate and chlorophyll *a* concentration were measured (M. Gosselin, personal communication) and large bird colonies at Prince Leopold Island (Fig. 1) (Wentworth et al., 2016) likely contribute to TMA emissions in the area. Consistent with these observations, previous aerosol chemical composition measurements on Bird Island in the South Atlantic (> 50°S) have reported the presence of amines and amino acids emitted from local fauna including seabirds, penguins and fur seals (Schmale et al., 2013). Further, air mass history predicted by FLEXPART three-day backward simulations (Fig. 17) illustrates that these air masses were advected at low levels above open water regions in Lancaster Sound, Baffin Bay and Nares Strait (compare with Fig. 1). Air masses measured during this flight leg on 8 July resided for more than 17 hours during the three days prior to sampling above regions of open water. Further, anthropogenic influences on amine emissions from nearby Resolute Bay are likely negligible since CO concentrations are very low. Another important finding is that primary sea spray particles (Na/Cl-containing) and TMA-containing particles measured on 8 July are externally mixed (Fig. 15) although both substances seems to be released from the ocean. This analysis solidifies the earlier hypothesis (Sect. 3.3) that particulate TMA presents secondary aerosol (Facchini et al., 2008). The higher abundance of the TMA-containing particle sub-type "Non-K,NH_4-containing" on 8 July (Fig. 15), compared to other days, further supports the hypothesis of SOA formation. It is further relevant to discuss that on 8 July from 15:50 UTC until 17:20 UTC (respective flight leg in Fig. 15) we flew low over sea ice in the vicinity of dissipating low-level clouds. These clouds had formed above the open water regions east of our flight leg (Burkart et al., 2017; Leaitch et al., 2016). We can therefore assume that cloud processing likely contributed to enhanced gas-to-particle partitioning of TMA as earlier reported in Rehbein et al. (2011). In addition, high organic-to-sulfate and MSA-to-sulfate ratios measured with the HR-ToF-AMS during this flight leg (see Sect. 4.3 in Burkart et al. (2017)) indicate that particle growth was driven by ocean-derived precursor gases (dimethylsulfide and organic species). Taken together,

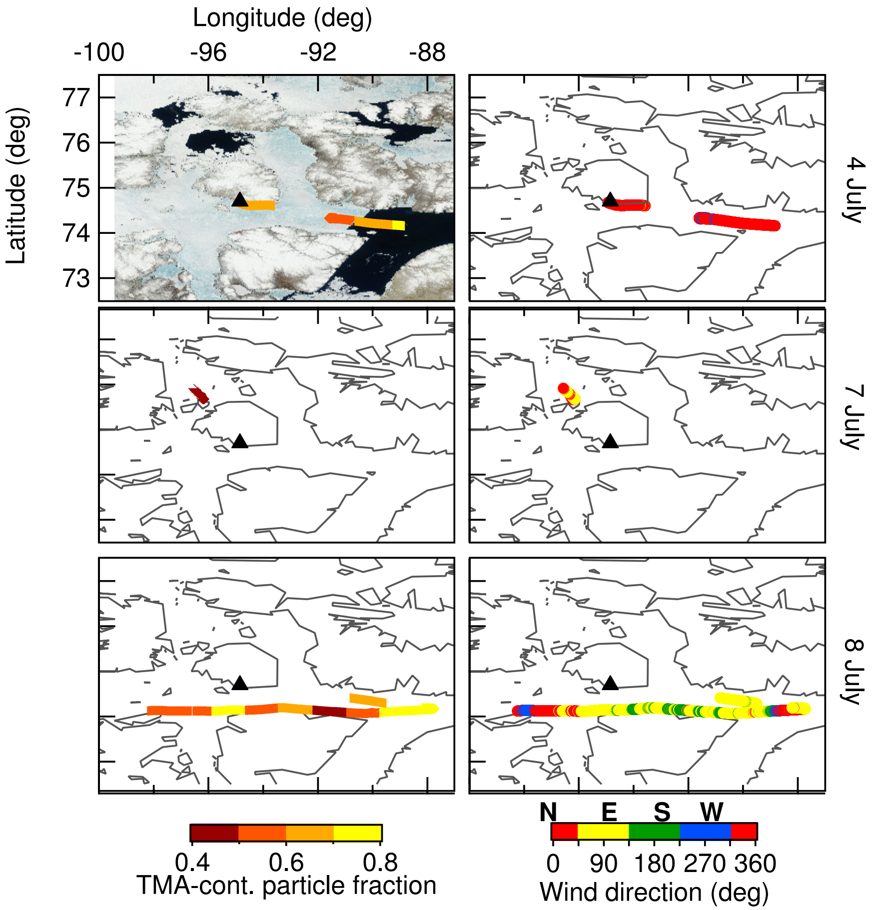

Figure 16. Spatially resolved fraction of TMA-containing particles (left column, color-coded) and wind direction (right column, color-coded) below 340 m. Different rows present different measurement days. The first graph additionally shows the satellite image on 4 July in the visible range. Further satellite images are not presented here due to negligible changes in sea ice coverage from 4 - 8 July. Abbreviations N, E, S and W refer to North, East, South and West. The black triangle presents the location of Resolute Bay on the map.

results from 8 July demonstrate secondary organic aerosol formation from marine-biogenic sources of gas-phase precursors, including TMA.

4 Conclusions

We presented results from aircraft-based single particle aerosol measurements in the summertime Canadian High Arctic. Our study has shown the presence of particulate TMA in the Arctic summer, comprising 23 % of all particles analyzed by the ALABAMA. SPMS measurements do not provide bulk analysis of aerosol chemical composition, therefore we can not obtain

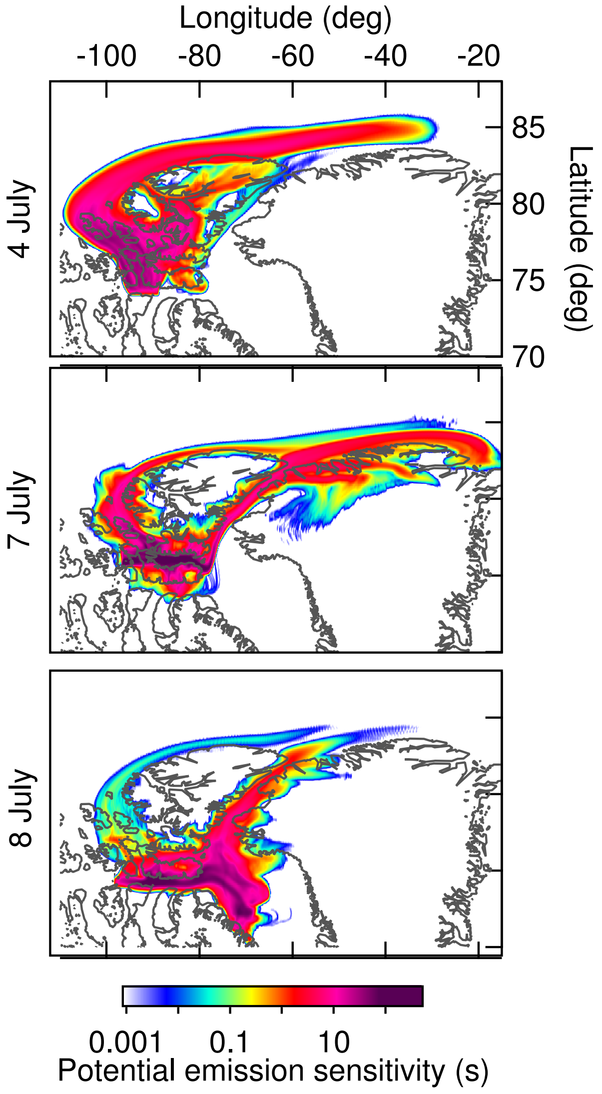

Figure 17. FLEXPART backward simulations of the considered measurement periods (Fig. 15) three days prior to sampling and at altitudes below 340 m. The color-coded area presents values of the potential emission sensitivity (PES) function in a particular grid cell (Sect. 2.3). Different rows depict different measurement days.

TMA mass concentrations. Nevertheless, the number of particles analyzed by the ALABAMA (> 7000) is sufficient to conduct a statistical analysis. This allows us to draw conclusions about mixing state, vertical and size distributions as well as potential emission sources of particulate TMA in summertime Arctic regions.

We present two main sources of particulate TMA in the summertime Arctic. First, we show the presence of inner-Arctic marine-biogenic sources resulting in secondary aerosol formation by TMA, sulfate, MSA, ammonia and other organics. Second, we have indications for long-range transport from biomass burning sources. We measured the maximum occurrence of particulate TMA (approximately 60 %) in a clean and stable stratified Arctic BL. In addition, TMA-containing particles present within the Arctic BL were smaller and were associated with less aged organic aerosol compared to aerosol observed aloft. High fractions of particulate TMA were measured at low wind speeds (near zero m/s) and close to the biologically active marginal ice zone. Further, BL air masses including high fractions of particulate TMA spent a long time (more than 17 hours) prior to sampling above Arctic open water regions. Moreover, the TMA particle sub-type containing MSA, sulfate and other organic species was more abundant when MSA mass concentrations (measured with HR-ToF-AMS) were high. Furthermore, the concurrent existence of sulfate, MSA and TMA in single particle spectra indicates the presence of aminium salts. This demonstrates that TMA may take part in neutralizing acidic aerosol along with ammonia. We additionally found that primary sea spray particles and TMA-containing particles are externally mixed although both substances are released by the ocean. It is further possible that gas-to-particle partitioning of TMA was enhanced in the vicinity of clouds and fog through dissolution of TMA in droplets and subsequent acid-base reactions (Rehbein et al., 2011). In contrast to the marine inner-Arctic sources, we have evidence for particulate TMA from long-rang transport of biomass burning aerosol. We demonstrate that levoglucosan and potassium (biomass burning tracers) are internally and externally mixed with particulate TMA. These particle types were more abundant above the Arctic BL as well as larger in particle sizes compared to particles not including these components.

Taken together, these findings contribute to our knowledge of marine-biogenic influences on secondary aerosol chemical composition and particle growth in the summertime Canadian Arctic. This is the first study demonstrating the incorporation of amines in Arctic aerosol from inner-Arctic sources. Based on spatial and temporal limitations of our measurements, it is difficult to assess how representative our findings are of the broader Arctic region. However, recent measurements confirm the presence of particulate amines and its marine-biogenic source at another Arctic site (Alert, 82.5 °N) (Leaitch et al., 2017). Future wide-spread and long-term Arctic measurements of atmospheric amines would help to extend our results to other regions.

Author contributions. Jon Abbatt, Richard Leaitch and Andreas Herber designed the research project. Franziska Köllner, Johannes Schneider, Heiko Bozem, Richard Leaitch, Megan Willis and Julia Burkart carried out the measurements. Amir Aliabadi processed the wind measurements. Thomas Klimach, Frank Helleis and Johannes Schneider re-designed and further developed the ALABAMA for aircraft-based measurements. Franziska Köllner analyzed the data with the help of Johannes Schneider, Peter Hoor, Thomas Klimach and Daniel Kunkel. Franziska Köllner wrote the manuscript. All co-authors commented on the manuscript.

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