

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 particles 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 (Leitch et al., 2016).

- (2) Also, as you have sampled particles during 4-12 July, any changes of the properties with the changes of meteorological 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-containing 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 ($\text{C}_3\text{H}_5\text{O}_2^-$) 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 ($\text{C}_2\text{O}_4\text{H}^-$)). 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 $N_{d>250\text{nm}}$ (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, 1.6 – p.21, 1.3

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, 1.17 - 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) (Leaitech 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,NH₄-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, 1.10-15

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.

p.16, 1.2-4

Comparison between CO mixing ratios and TMA sub-types abundance (Fig. 12) shows larger fractions of “K,NH₄,S-containing” and “levoglucosan-containing” TMA particle sub-types in higher CO environments compared to “Non-K,NH₄-containing” TMA particles.

p.17, 1.10-15

Furthermore, Fig. 14 indicates a positive correlation between MSA mass concentrations measured with HR-ToF-AMS and the fraction of “Non-K,NH₄-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,NH₄-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, 1. 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, 1.5-6):

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).

References:

- Andreae, M. O. and Merlet, P.: Emission of trace gases and aerosols from biomass burning, *Global Biogeochemical Cycles*, 15, 955–966, doi:10.1029/2000GB001382, <http://dx.doi.org/10.1029/2000GB001382>, 2001.
- Bond, T. C., Bhardwaj, E., Dong, R., Jogani, R., Jung, S., Roden, C., Streets, D. G., and Trautmann, N. M.: Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850–2000, *Global Biogeochemical Cycles*, 21, n/a–n/a, doi:10.1029/2006GB002840, <http://dx.doi.org/10.1029/2006GB002840>, gB2018, 2007.
- Burkart, J., Willis, M. D., Bozem, H., Thomas, J. L., Law, K., Hoor, P., Aliabadi, A. A., Köllner, F., Schneider, J., Herber, A., Abbatt, J. P. D., and Leaitch, W. R.: Summertime observations of elevated levels of ultrafine particles in the high Arctic marine boundary layer, *Atmospheric Chemistry and Physics*, 17, 5515–5535, doi:10.5194/acp-17-5515-2017, <http://www.atmos-chem-phys.net/17/5515/2017/>, 2017.
- Fu, P., Kawamura, K., and Barrie, L. A.: Photochemical and Other Sources of Organic Compounds in the Canadian High Arctic Aerosol Pollution during Winter-Spring, *Environmental Science & Technology*, 43, 286–292, doi:10.1021/es803046q, <http://dx.doi.org/10.1021/es803046q>, pMID: 19238953, 2009.
- Fu, P. Q., Kawamura, K., Chen, J., Charrière, B., and Sempéré, R.: Organic molecular composition of marine aerosols over the Arctic Ocean in summer: contributions of primary emission and secondary aerosol formation, *Biogeosciences*, 10, 653–667, doi:10.5194/bg-10-653-2013, <http://www.biogeosciences.net/10/653/2013/>, 2013.
- Heald, C. L., Kroll, J. H., Jimenez, J. L., Docherty, K. S., DeCarlo, P. F., Aiken, A. C., Chen, Q., Martin, S. T., Farmer, D. K., and Artaxo, P.: A simplified description of the evolution of organic aerosol composition in the atmosphere, *Geophysical Research Letters*, 37, n/a–n/a, doi:10.1029/2010GL042737, <http://dx.doi.org/10.1029/2010GL042737>, l08803, 2010.

Jimenez, J. L., Canagaratna, M. R., Donahue, N. M., Prevot, A. S. H., Zhang, Q., Kroll, J. H., DeCarlo, P. F., Allan, J. D., Coe, H., Ng, N. L., Aiken, A. C., Docherty, K. S., Ulbrich, I. M., Grieshop, A. P., Robinson, A. L., Duplissy, J., Smith, J. D., Wilson, K. R., Lanz, V. A., Hueglin, C., Sun, Y. L., Tian, J., Laaksonen, A., Raatikainen, T., Rautiainen, J., Vaattovaara, P., Ehn, M., Kulmala, M., Tomlinson, J. M., Collins, D. R., Cubison, M. J., Dunlea, J., Huffman, J. A., Onasch, T. B., Alfarra, M. R., Williams, P. I., Bower, K., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Salcedo, D., Cottrell, L., Griffin, R., Takami, A., Miyoshi, T., Hatakeyama, S., Shimojo, A., Sun, J. Y., Zhang, Y. M., Dzepina, K., Kimmel, J. R., Sueper, D., Jayne, J. T., Herndon, S. C., Trimborn, A. M., Williams, L. R., Wood, E. C., Middlebrook, A. M., Kolb, C. E., Baltensperger, U., and Worsnop, D. R.: Evolution of Organic Aerosols in the Atmosphere, *Science*, 326, 1525–1529, doi:10.1126/science.1180353, <http://science.sciencemag.org/content/326/5959/1525>, 2009.

Hu, Q.-H., Xie, Z.-Q., Wang, X.-M., Kang, H., and Zhang, P.: Levoglucosan indicates high levels of biomass burning aerosols over oceans from the Arctic to Antarctic, *Scientific Reports*, 3, 3119, doi:10.1038/srep03119, <http://dx.doi.org/10.1038/srep03119>, 2013.

Hudson, P. K., Murphy, D. M., Cziczo, D. J., Thomson, D. S., de Gouw, J. A., Warneke, C., Holloway, J., Jost, H.-J., and Hübner, G.: Biomass burning particle measurements: Characteristic composition and chemical processing, *Journal of Geophysical Research: Atmospheres*, 109, n/a–n/a, doi:10.1029/2003JD004398, <http://dx.doi.org/10.1029/2003JD004398>, d23S27, 2004.

Leaitch, W. R., Korolev, A., Aliabadi, A. A., Burkart, J., Willis, M. D., Abbatt, J. P. D., Bozem, H., Hoor, P., Köllner, F., Schneider, J., Herber, A., Konrad, C., and Brauner, R.: Effects of 20–100 nm particles on liquid clouds in the clean summertime Arctic, *Atmospheric Chemistry and Physics*, 16, 11 107–11 124, doi:10.5194/acp-16-11107-2016, <http://www.atmos-chem-phys.net/16/11107/2016/>, 2016.

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.

Ng, N. L., Canagaratna, M. R., Jimenez, J. L., Chhabra, P. S., Seinfeld, J. H., and Worsnop, D. R.: Changes in organic aerosol composition with aging inferred from aerosol mass spectra, *Atmospheric Chemistry and Physics*, 11, 6465–6474, doi:10.5194/acp-11-6465-2011, <https://www.atmos-chem-phys.net/11/6465/2011/>, 2011.

Pratt, K. A. and Prather, K. A.: Real-Time, Single-Particle Volatility, Size, and Chemical Composition Measurements of Aged Urban Aerosols, *Environmental Science & Technology*, 43, 8276–8282, doi:10.1021/es902002t, <http://dx.doi.org/10.1021/es902002t>, PMID: 19924956, 2009.

Pratt, K. A., Murphy, S. M., Subramanian, R., DeMott, P. J., Kok, G. L., Campos, T., Rogers, D. C., Prenni, A. J., Heymsfield, A. J., Seinfeld, J. H., and Prather, K. A.: Flight-based chemical characterization of biomass burning aerosols within two prescribed burn smoke plumes, *Atmospheric Chemistry and Physics*, 11, 12 549–12 565, doi:10.5194/acp-11-12549-2011, <https://www.atmos-chem-phys.net/11/12549/2011/>, 2011.

Silva, P. J., Liu, D.-Y., Noble, C. A., and Prather, K. A.: Size and Chemical Characterization of Individual Particles Resulting from Biomass Burning of Local Southern California Species, *Environmental Science & Technology*, 33, 3068–3076, doi:10.1021/es980544p, <http://dx.doi.org/10.1021/es980544p>, 1999.

Simoneit, B., Schauer, J., Nolte, C., Oros, D., Elias, V., Fraser, M., Rogge, W., and Cass, G.: Levoglucosan, a tracer for cellulose in biomass burning and atmospheric particles, *Atmospheric Environment*, 33, 173–182, 1999.

Simoneit, B. R.: Biomass burning - review of organic tracers for smoke from incomplete combustion, *Applied Geochemistry*, 17, 129 – 162, doi:[http://dx.doi.org/10.1016/S0883-2927\(01\)00061-0](http://dx.doi.org/10.1016/S0883-2927(01)00061-0), <http://www.sciencedirect.com/science/article/pii/S0883292701000610>, 2002.

Willis, M. D., Burkart, J., Thomas, J. L., Köllner, F., Schneider, J., Bozem, H., Hoor, P. M., Aliabadi, A. A., Schulz, H., Herber, A. B., Leaitch, W. R., and Abbatt, J. P. D.: Growth of nucleation mode particles in the summertime Arctic: a case study, *Atmospheric Chemistry and Physics*, 16, 7663–7679, doi:10.5194/acp-16-7663-2016, <http://www.atmos-chem-phys.net/16/7663/2016/>, 2016.

Willis, M. D., Köllner, F., Burkart, J., Bozem, H., Thomas, J. L., Schneider, J., Aliabadi, A. A., Hoor, P. M., Schulz, H., Herber, A. B., Leaitch, W. R., and Abbatt, J. P. D.: Evidence for marine biogenic influence on summertime Arctic aerosol, *Geophysical Research Letters*, 44, 6460–6470, doi:10.1002/2017GL073359, <http://dx.doi.org/10.1002/2017GL073359>, 2017GL073359, 2017.

Zauscher, M. D., Wang, Y., Moore, M. J. K., Gaston, C. J., and Prather, K. A.: Air Quality Impact and Physicochemical Aging of Biomass Burning Aerosols during the 2007 San Diego Wildfires, *Environmental Science & Technology*, 47, 7633–7643, doi:10.1021/es4004137, <http://dx.doi.org/10.1021/es4004137>, PMID: 23750590, 2013.