Reply to *Interactive Comment* on "Atmospheric VOC measurements at a High Arctic site: characteristics and source apportionment" from anonymous Referee # 1

This manuscript reports on a long-term (spring through fall) Arctic VOC dataset observed at Villum Research Station at Station Nord in Greenland, and a PMF analysis performed on the dataset. The authors report the PTR-ToF-MS results for 10 detected ions, assigning 10 gas-phase molecular formulae and species/species groups to the observed ions in the PTR. A PMF analysis of the 10 species and species groups with a four-factor analysis is presented and discussed at length, including a Biomass Burning Factor, a combination Marine Cryosphere Factor, a Background Factor, and an Arctic Haze Factor. The authors give a very nice detailed analysis of the four factors, including the primary components, sources and influences and temporal variability.

We would like to thank referee # 1 for carefully reading the manuscript and for useful comments and feedback. We feel it improved the manuscript's readability and overall discussion of the results. As the first author is an early career scientist, they feel this exercise in the peer-review has tremendously helped them progress in critical thinking, manuscript writing, and the scientific method. We have addressed the referee's concerns and corrected errors in the manuscript below with referee's comments numbered and the author's responses in blue. New references are highlighted in yellow.

Several of the referee's concerns arose from the lack of explanation of the VOC specificity. We have group several of his comments into one and responded to them all with one reply.

1) One of my primary concerns with the paper, and with the majority of PTR- instrument papers, is that there is a lack of accounting or explanation of the VOC specificity (or lack thereof) of the PTR technique. The authors make no effort in this paper to discuss the interfering or additional species that may comprise each observed chemical formula that make up several of their measurements – e.g., propanal's contribution to the signal attributed to acetone, isobutanal's and butanal's contribution to the signal attributed to MEK – even to justify the omission of these species from the discussion with adequate explanation and literature references. As well, the authors' treatment of methyl acetate and propionic acid is to suggest that the contributions from each species (or other species that might contribute to the $C_3H_6O_2H^+$ signal) are un- known in Section 2.2, but then they attribute the signal to methyl acetate in the Biomass Burning Factor, and propionic acid in the Marine Cryosphere Factor, with no justification as to the reasons for the identification. The authors need to add commentary for the species identification, and justify the assumed VOCs under different conditions, or simply refer to the observations as a generic $C_3H_6O_2$ VOC group. Also, as detailed below, references to VOCs that comprise the $C_5H_8O$ observation should be clear that the measurement is not of an ion ($C_5H_8OH^+$ or $C_5H_8O^+$), but of the $C_5H_8O$ VOC group.

2) Lines 145-152 – the authors describe the method by which "compound names" are assigned to the nine protonated masses, including Pagonis et al. and references, which is reasonable, and a priori knowledge, which is not something that can be reference checked. I would argue that there is insufficient justification given to identifying the masses which ignore contributions from additional compounds that may be included in the concentrations measured. The authors write "Another compound ($C_4H_8OH^+$) was doubly assigned to

propionic acid and methyl acetate.", but they likely meant to write C3H6O2H+, which has m/z 75.058. However, they should explain here why they don't include ethyl formate or hydroxyacetone as possible compounds at this mass.

3) Line 337 – "it is a source of methyl acetate as well. . ." – the authors recognize that methyl acetate could be contributing to the C3H6O2H+ signal, but by labeling it "propionic acid" in Table 2 and Figures 1, 3, 5, 7, etc., the identity of the compound is muddied. If the authors truly believe that the species is primarily propionic acid, then the presence of methyl acetate would be unimportant. If they believe that it is indeed a mixture of the two (or more) species, then this should be made clear whenever it is being referred to.

We recognize that the points made by the referee are correct and have thus modified the manuscript, accordingly, adding a more detailed discussion of the possible and most likely assignments of the detected masses to chemical species:

Line 160: "The PTR-MS technique allows to observe species with a proton-affinity higher than water, this encompasses most VOCs found in the atmosphere with the important exception of alkanes. It does not allow for the distinction between isomers to be made. Compound names were assigned based on comparison with the libraries from the PTR-MS Viewer and Pagonis et al. (2019), and references therein. Inspection of the mass spectrum yielded ten protonated masses from which an empirical formula was calculated, and compound names were assigned for nine masses, as discussed in Sect. 3.1."

The following paragraph has been added at the beginning of Sect. 3.1:

Line 252: "The ten selected masses monitored by the PTR-TOF-MS and their assignments to species names are presented in Table 1. Assignments are made by choosing the most plausible contributions to an observed mass but each measured ion may have contributions from several different isomeric molecules. The assignment of masses in the table to protonated molecules of formaldehyde, acetonitrile, formic acid, acetic acid, and benzene appears to be unproblematic as no meaningful alternatives are found. For the remaining molecules, alternative assignments are possible. The mass assigned to acetone could be propanal as well, but propanal has a shorter atmospheric residence time and acetone is known to be one of the dominating VOCs observed in the atmosphere (Jacob et al., 2002), further, it has been found to have sources in the Arctic (Guimbaud et al., 2002). The mass assigned to DMS could be ethanethiol as well, but the large marine source of DMS makes it the most plausible assignment. Methyl ethyl ketone is isomeric with butenal, but being the second most abundant ketone in the atmosphere with, among others, apparently an oceanic source (Brewer et al., 2020) it appears to be the best assignment. $C_3H_6O_2$ may stem from propionic acid but also hydroxyacetone, methyl acetate, and ethyl formate. While it seems unlikely that ethyl formate could give a major contribution to this signal, the other three species are all plausible candidates: Low molecular weight organic acids are commonly found in the atmosphere (Lee et al., 2009), methyl acetate has been found in emissions from biomass burning (Andreae, 2019) and hydroxyacetone is known to be formed by the atmospheric degradation of isoprene (Karl et al., 2009). For what concerns the $C_5H_8OH^+$ ion we prefer not to make an assignment, possible isomers include, among others, pentenals and pentenones."

The references Jacob et al., 2002, Brewer et al., 2020, Lee et al., 2009 and Karl et al., 2009 are new and have been added to the list of references.

Line 271 and lines 282–284: Sentences have been deleted.

Line 286: ‘propionic acid’ has been replaced by “$C_3H_6O_2$”.

Lines 297–298: ‘an oxidation product of n-butane’ has been deleted.

Line 404–405: “one of the $C_3H_6O_2$ isomers” has been added to the sentence.

Line 462: ‘propionic acid’ has been replaced by “$C_3H_6O_2$”.

Line 469–471: “The $C_3H_6O_2$ is in this case assigned to propionic acid as the alternative isomers seem less probable, considering their typical origins (biomass burning for methyl acetate and isoprene oxidation for hydroxyacetone).”

Line 564: ‘propionic acid’ has been replaced by “$C_3H_6O_2$”.

The following sentence has been added:

Line 564–565: “$C_3H_6O_2$ may in this case result from all three of the isomers: propionic acid, methyl acetate, and hydroxyacetone.”

Throughout the manuscript, and specifically in Figure 1, 2, 4, 5, 8, and 10 as well as Table 2, S2, S3, S4 and S5, ‘propionic acid’ has been replaced by “$C_3H_6O_2$”.

4) My other primary concern is that the authors indicate that the data are available by contacting one of two author email addresses. I would strongly recommend that the paper not be published until the data are available in a publicly-available DOI.

The data for this manuscript including VOC mixing ratios and their associated uncertainty can be found in a publicly-available DOI. The Data Availability section has been amended to now read:

Line 673–675: All data used in this publication are available at https://doi.org/10.5281/zenodo.4299817 or by request to the corresponding authors Jakob Boyd Pernov (jbp@envs.au.dk) and Rossana Bossi (rbo@envs.au.dk).

Lines 155-157 – The authors should be specific about how the data were quality controlled using these parameters (PSND, WD, WS, etc.), and the resulting amount (total percentage, number of time periods, etc.) of data that had to be eliminated from the useful data set.

We have added Section 2 “Quality Control Procedure” in the Supplement which describes how local pollution was identified and removed (see text below). We have also a column in Table 1 which lists the total percentage of data removed due to QC (see an updated Table 1 below).

SI Line 35–52: **“Quality Control Procedure**

Data were quality controlled by analysis of PNSD, ozone, wind direction and speed, and internal activity logs. Local pollution at Villum can arise from activity around the measurement site (e.g., passenger vehicles, all-terrain vehicles, snowmobiles, and heavy machinery) as well as from activities from Station Nord (e.g., waste incineration, vehicular activity, and aircraft landing, idling, and take off). Internal activity logs of visits to the measurement building were used to highlight periods when human activity could affect the measurements, periods where VOC levels were elevated over background levels for the duration of the visit to the station were removed. Measurements of PNSD and ozone were analyzed, in tandem, for sharp and sudden increases in the ultrafine mode (< 100 nm) aerosol particles and concurrent sharp and sudden decreases in ozone, increases in ultrafine mode particles are indications of vehicular emissions while decreases in ozone results from its titration with nitrogen oxides. These periods were further inspected for wind direction and speed, with winds coming from due north at low speeds indicative of local pollution from Station Nord. All periods where local pollution was suspected of influencing the measurements were visually inspected by a panel of three persons, a consensus was required before data were removed. Data were also quality controlled for abnormal levels of instrumental parameters (i.e., E/N ratio, drift tube temperature, pressure, and voltage), periods with large deviations from nominal values were removed. Certain compounds (DMS, formic acid, and acetic acid) exhibited a slow return to nominal values after a blank than before, this issue was especially evident in the summer, these periods were removed.  All quality control was performed on VOCs at a 5 s time resolution, data was removed before averaging to 30-minute means.”

**Table 1.** Overview of measured protonated masses included in PMF analysis.  Mean refers to the arithmetic average of the mixing ratio for each compound. Mean, Mean LOD, and % < LOD were calculated after quality control of data influenced by local pollution. % QC represents the percentage of data removed due to the Quality Control Procedure (Sect. S2).

| Measured mass (*m/z*) | Empirical Formula | Assigned Compound | Mean (ppbv) | Mean LOD (ppbv) | % < LOD | Mean Relative Uncertainty (%) | % QC |
|---|---|---|---|---|---|---|---|
| 30.997 | $CH_2OH^+$ | Formaldehyde | 0.220 | 0.176 | 0.6 | 41 | 5 |
| 42.019 | $C_2H_3NH^+$ | Acetonitrile | 0.067 | 0.045 | 0 | 46 | 5 |
| 47.011 | $CH_2O_2H^+$ | Formic Acid | 0.454 | 0.250 | 17 | 37 | 7 |
| 59.062 | $C_3H_6OH^+$ | Acetone | 0.608 | 0.037 | 0 | 32 | 0 |
| 61.047 | $C_2H_4O_2H^+$ | Acetic Acid | 0.201 | 0.096 | 5 | 39 | 8 |
| 63.034 | $C_2H_6SH^+$ | Dimethyl Sulfide | 0.046 | 0.043 | 4 | 57 | 25 |
| 73.068 | $C_4H_8OH^+$ | Methyl Ethyl Ketone | 0.031 | 0.023 | 0.1 | 56 | 0 |
| 75.058 | $C_3H_6O_2H^+$ | Propionic Acid / Hydroxyacetone/ Methyl Acetate | 0.025 | 0.031 | 0.1 | 61 | 2 |
| 79.057 | $C_6H_6H^+$ | Benzene | 0.027 | 0.031 | 0.5 | 64 | 0 |
| 85.066 | $C_5H_8OH^+$ | N/A | 0.027 | 0.030 | 0.03 | 61 | 0 |

5) Lines 215, 212, 467, 506, etc. – Technically the authors did not observe ambient C5H8O+ ions (or C5H8OH+ ions), but rather a compilation of [some] gas-phase C5H8O species, which were protonated in order to be observed by the PTR system, similar to how they did not observe atmospheric ambient C6H6H+ ions, but rather gas- phase C6H6 (i.e., benzene). Thus, discussion of the species or group of species with the chemical formula

C5H8O should simply be “C5H8O species” or “C5H8O”, as in Figure 1, and should not imply the measurement of an atmospheric ion.

The group of species identified at m/z 85.066 is now referred to as simply “$C_5H_8O$” throughout the text.

6) Lines 245-252 – The comparisons presented against literature data from similar Arctic stations make sense, for the most part, but the comparison of wintertime benzene mixing ratios from Gautrois et al. (2003) to this study are not merited, as no winter time data is being presented here. As well, while I agree that it has been shown that benzene and acetonitrile are influenced by lower latitudes, the claim that acetonitrile is influenced by anthropogenic emissions is not backed up. Remote levels of acetonitrile are likely impacted by the significance of mid-latitude fire seasons, and are not expected to compare well from year to year.

The referee is correct no wintertime data was collected. We compared our spring period measurements to Gautrois et al. (2003) wintertime data. The authors agree this comparison could create some confusion as it was not indicated in the text that we compared springtime to wintertime data. The text had been amended to reflect only comparisons during summer.

Line 305–308: “Benzene has shown a seasonal pattern at Alert, NU with a higher mixing ratio in winter due to no or limited photochemistry and long-range transport from lower latitudes (Gautrois et al., 2003). They reported mean winter and summer mixing ratios of 0.200 and 0.034 ppbv, respectively; when compared to the present study there is good agreement during the summer.”

The authors interpreted the similar pattern as benzene during spring to be indicative of anthropogenic influence, although the referee is correct, year to year variability from fires could hinder the proper interpretation of this pattern. The text has been amended as follows:

Line 309–310: “Acetonitrile followed a similar pattern to benzene during the spring with decreasing values, as well as exhibiting minima in the summer and maxima during the autumn (Fig. 1b).”

We have added the following sentence:

Line 314: The main source of acetonitrile in the atmosphere has been found to be biomass burning (Singh et al., 2003).

The reference Singh et al., 2003 is new and has thus been added to the list of references.

The discussion of acetonitrile during spring in Sect. 3.3.1 has also been changed:

Lines 407-412: “The decrease in the spring is reflective of decreasing concentrations of benzene and acetonitrile; in the case of benzene this can be ascribed to anthropogenic emissions during this period as the polar dome is expanded during winter and spring allowing for emissions to be entrained from the mid-latitudes. In the case of acetonitrile, the reason is more uncertain, there are anthropogenic sources of acetonitrile, particularly wood burning for residential heating and solvent use (Languille et al., 2020), but they appear to be of very minor importance compared to forest fires (de Gouw et al., 2003).”

Languille et al., 2020 is a new reference that has been added to the list of references.

We have also added the following text in the Arctic Haze section when we compare our Arctic Haze Factor to other Haze factors from previous literature.

Line 617–620: "It is worth noting that the Arctic Haze Factor from this study is only for spring, while the other studies present data from the winter/spring, therefore any comparisons we make are from our spring Arctic Haze Factor to other Haze factors during winter and spring. While this is not a perfect comparison, it is one worth making, as Arctic Haze is the main source of anthropogenic pollution in the Arctic."

7) Line 308 – the authors state that species with S/N < 0.2 were excluded from the analysis, but all 10 species (or species groups) discussed in the paper are included in Table 2. Are there any other species that were measured but not included here?

The species listed in Table 1 and 2 were the compounds identified that could be reasonably identified with an empirical formula with a proton affinity greater than water, without interference from neighboring ions, and exhibited a meaningful temporal profile.

The PTR measures ions with a m/z ratio up to 430 Da, so there are hundreds of ions measured by the instrument, but the ions reported here are the only those the authors could be confident were real signals from ambient VOCs. To answer the referee's question, no there was not.

8) Line 445 – The back trajectories frequency map for the Marine Cryosphere Factor is interesting, but it would be more informative to highlight some of the brief periods where this factor is particularly elevated, rather than averaging over a three-month summer period. Given, as well, that all the species identified to contribute to the Marine Cryosphere Factor have atmospheric lifetimes < 5 days, it would be prudent to limit these back trajectories to 120 hours or less.

The second referee has asked for a potential source contribution function (PSCF) for source region analysis of the Marine Cryosphere Factor. The authors agree this would be the appropriate method for determining source regions for the Marine Cryosphere Factor. Therefore, the authors have become familiar with the programming language R and the R package Openair (Carslaw and Ropkins, 2012). Using this package, the authors were able to produce a (PSCF) for source region analysis of the Marine Cryosphere Factor. We have replaced the trajectory frequency map for the summer season with a PSCF map for the entire campaign. A PSCF for the summer period was also produced and compared to the entire campaign which produced similar results. Inclusion of the entire campaign data provides a more robust statistical calculation of the PSCF; therefore, we have chosen to perform the PSCF for the entire campaign.

We have replaced the trajectory frequency map in Fig. 7 (previously Fig. 6, we have added a figure showing the diurnal profile of the four factors in as the new Fig. 6 thus making this Fig. 7) with the PCSF as seen below, and updated the figure caption accordingly.

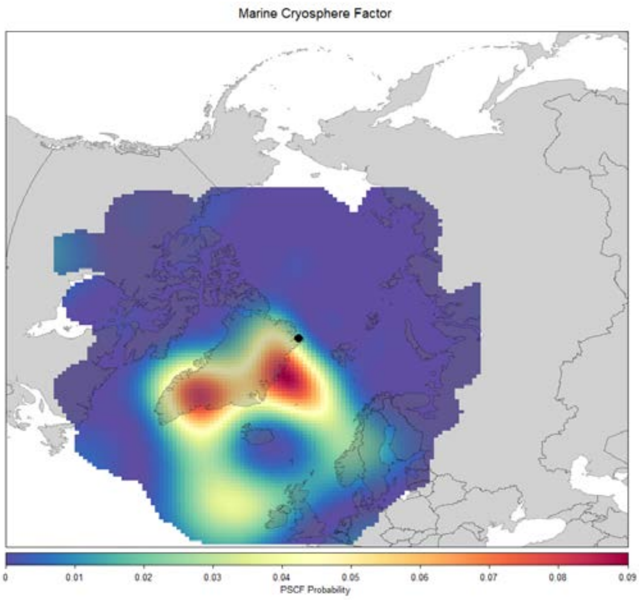


**Fig. 7.** PSCF for the Marine Cryosphere Factor and air mass back trajectories arriving at 100 m altitude, extending backward 120 hours in time. This plot and analysis method were produced in R and R Studio programs (R Foundation for Statistical Computing, Vienna, Austria, and R Studio Inc, MA, USA) and the OpenAir suite of analysis tools (Carslaw and Ropkins, 2012).

We have also updated Sect. 2.5 Back Trajectory Analysis to describe the PSCF:

Line 232–250: "To investigate source regions, the R package Openair (Carslaw and Ropkins, 2012) was utilized to produce a potential source contribution function (PSCF). Trajectories in Openair were calculated using the HYSPLIT model (Draxler and Hess, 1998; Rolph et al., 2017) at 100 m altitude and 120 hours backwards in time using Global NOAA-NCEP/NCAR reanalysis data archives on a 2.5° resolution. A PSCF, shown in Eq. (3), calculates the probability that an emission source is located in a grid cell of latitude *i* and longitude *j*, on the basis that emitted material in the gird cell *ij* can be transported along the trajectory and reach the receptor site.

$$PSCF = \frac{m_{ij}}{n_{ij}} \quad (3)$$

Where $n_{ij}$ is the number of times a trajectory has passed through grid cell *ij* and $m_{ij}$ is the number of times that a concentration was above a certain threshold value, in this case the 90th percentile. To account for uncertainty in cells with a small number of trajectories passing through, a weighting function was applied (Carslaw and Ropkins, 2012)."

We have also added the following text in the Marine Cryosphere Factor section discussing the results.

Line 527–541: "The spatial origin of the Marine Cryosphere Factor was investigated via a PSCF, calculated with the R package Openair (Carslaw and Ropkins, 2012). Figure 7 displays the PSCF for air masses arriving every hour during the measurement campaign, which provides increased statistical robustness to the results over calculating a PSCF just for the summer period. From Fig. 7, two areas with a high probability of being a source region for the Marine Cryosphere Factor can be discerned, the coast around Southeastern and Northeastern

Greenland. This analysis is supported by the CPF for the Marine Cryosphere Factor (Fig. S8b), which shows the dominant wind direction for this factor to be the south and south-south-east. Lee et al. (2020) used monthly chlorophyll-*a* derived from the MODIS satellite to demonstrate the coasts around Northeastern Greenland to contain high chlorophyll-*a* concentrations during June, which has been supported by previous studies (Degerlund and Eilertsen, 2010; Galí and Simó, 2010). Lee et al. (2020) also used a PSCF to determine this area to be the source regions for total particle number concentrations in the nucleation size range (3–25 nm). This area has been demonstrated to be a source region for MSA during the summer months (Heintzenberg et al., 2017). Thus, we propose the biologically active coasts around Eastern Greenland to be the source region for the Marine Cryosphere Factor."

The references Carslaw and Ropkins, 2012, Degerlund and Eilertsen, 2010, Galí and Simó, 2010, Lee et al. (2020), and Heintzenberg et al., 2017 are new and thus have been added to the reference list.

9) Figures – all figures in the primary manuscript and supplement should be saved at a higher resolution. There is significant pixilation when zooming in on the plots. Some of the finer details are lost as a result, and some of the axis labels are rendered illegible.

All figures included in the manuscript have been saved at a resolution of 600 DPI. This is an excellent suggestion and in the future the authors will be more attentive to this matter.

10) Table 1 – The table title doesn't need to be so long. "Overview of measured protonated masses included in PMF analysis" would be sufficient. The rest is redundant with the table header, although Mean Mixing Ratio should be spelled out in the header or defined in a footnote. As well, it would be good to specify if the "Percentage below LOD" is the percentage of all data collected, or the percentage of only the data that was not removed due to the influence of local pollution. The same comment goes for the means reported.

The table title has been shortened with redundant information removed and the following text added:

Table 1: "Overview of measured protonated masses included in PMF analysis. Mean refers to the arithmetic average of the mixing ratio for each compound. Mean, Mean LOD, and % < LOD were calculated after quality control of data influenced by local pollution. % QC represents the percentage of data removed due to the Quality Control Procedure (Sect. S2)."

11) Line 27 – "rate" would be preferable to "speed".

Line 29: "Speed" has been replaced with "rate".

12) Line 33 – NOx should be defined.

Line 35 and 36: "$NO_x$" has been defined as "nitrogen oxides" and "VOCs" have been defined as "volatile organic compounds".

Line 52: "DMS" has been defined as "dimethyl sulfide".

13) Line 46 – there is a rogue hyphen/em dash that isn't needed.

Line 48: The rouge em dash has been removed.

14) Line 46 and others – Dall'Osto is missing an apostrophe both here and in the refer- ence list, where the reference is also missing several other diacritical marks, and the majority of C.D. O'Dowd's last name. The references should then be rearranged for this reference to come before the more recent Dall'Osto et al. references. Be wary of automatic reference management software – references should still be verified that they were transposed and recorded properly.

References – Please format all references properly: pay attention to things like consistent journal abbreviations, consistent DOI referencing, missing or n/a information (e.g., line 680), line wrapping (e.g., line 735), and capitalization of abbreviations and proper names (e.g., lines 632; 839; 842, etc.).

This is an excellent catch by the referee. The Dall'Osto reference has been corrected and the entire reference list has been checked for accuracy and updated where appropriate. This is excellent advice from the referee. We believe the problem arose from importing references from PDFs using the "Import" function in Endnote. We have now imported references either form Web of Science or the respective journal website. The authors were unaware of such pitfalls when working with automatic referencing software and will be more vigilant in the future.

15) Line 68 – "loss" rather than "reactions" would generate better agreement with the singular "sink".

Line 70: "reactions" has been replaced with "loss".

16) Lines 90-93 – "Furthermore, Boudries et al. (2002) observed emission from the snow- pack to the atmosphere of acetone, acetaldehyde, and formaldehyde, which were explained by photochemical production in the snowpack and depositional fluxes of methanol was also observed, which they postulated as a source of formaldehyde."- Consider making this two sentences: "Furthermore, Boudries et al. (2002) observed emission from the snowpack to the atmosphere of acetone, acetaldehyde, and formaldehyde, which were explained by photochemical production in the snowpack. "Depositional fluxes of methanol were also observed, which they postulated as a source of formaldehyde" Or at the very least, add a semi-colon and change "was" to "were" on line 92.

Line 94: This sentence has been made into two sentence following the referee's suggestions and "was" is now "were".

17) Line 94 – there should be a comma after "VOCs".

Line 96: A comma has been added after VOCs.

18) Line 103 – It would be good to mention that Barrow, AK is now Utqiagvik, AK.

Throughout the text "Barrow" has been replaced with "Utqiagvik".

On line 105, it is mentioned that Utqiagvik is formerly known as Barrow.

19) Line 104 and others – While "Alert, CA" is technically acceptable, "Alert, Canada,", or "Alert, NU," would be less ambiguous. Also, be consistent throughout. Greenland should probably be spelled out as well.

Throughout the text “Alert, CA” has been replaced with “Alert, NU”.

20) Line 122 – Use “s” instead of “sec” to adhere to SI units requirement.

Line 133: “sec” has been replaced with “s”. The entire manuscript and SI has also been checked for proper use of SI units were appropriate.

21) Line 122 – Use “southwest” instead of “south-west”.

Line 133: The hyphen has been removed.

22) Line 131 – “5 seconds scan rate” doesn’t describe a rate, which should be something per unit of time.

Line 142: “5 seconds scan rate” has been replaced with “5 second single spectra integration time” as specified in the PTR software.

23) Line 154 – “mixing ratios below LOD were set to”

Line 173: “was” has been replaced with “were”.

24) Line 155 – “the data were time-averaged to 30-minute means.”

Line 173: “was” has been replaced with “were” and “mean” was made plural.

25) Line 194 – “arriving from”

Line 222: “form” has been replaced with “from”.

26) Lines 199-200 – “Active fires during the period 15 August – 15 September 2018 were provided...” (you are defining the period here, not referring to it, so the commas are not needed.)

This sentence has been removed from this section and moved to the Biomass Burning section (Line 424), where the commas have been removed.

27) Line 259 – I recommend splitting this long sentence, “. . . frozen sea surface. Back trajectory. . .”

Line 324–325: The sentence has been split into two following the referee’s suggestion.

28) Line 267 – “strong negative correlation” is a little too generous for R = -0.68.

Line 332–333: “strong” has been replaced with “moderate”.

29) Lines 271-273 – It would be informative to include wind direction in addition to wind speed in Figure 2.

Wind direction has been added to Figure 3, which is the old Figure 2 after addition of a figure showing the diurnal profile of certain VOCs during the summer as suggested by the second referee. Wind direction has also been added to Fig. S2. The effect of wind direction has been included in the text:

Line 338: These changes in mixing ratios are accompanied by a change in meteorological conditions, illustrated here by changes in wind speed and to a less extent wind direction (Fig. 3).

30) Line 275 – “with elevated acetone levels during ozone. . .” or something similar.

The text has been amended following the referee’s suggestion.

Line 339–341: Guimbaud et al. (2002) found a similar relationship between acetone and ozone during different field campaigns at Alert, Canada with elevated acetone levels during ozone depletion episodes accompanied by a concomitant decrease in the propane mixing ratios.

31) Line 279 – “gas-phase”

Line 342: A hyphen has been added to “gas-phase”.

32) Line 303 – “species with S/N. . .”

Line 369: “Signal-to-noise” has been removed.

33) Line 304 – “The uncertainties of ‘Weak’ species were tripled. . .”

Line 370: “Uncertainty” has been made plural to “uncertainties” and “was” replaced with “were”.

34) Line 314 – “VOCs devoid of episodic influence. . .”, and there is a period missing at the end of the sentence.

Line 381: “void” has been replaced with “devoid” and a period has been added to the end of this sentence.

35) Line 394 – The authors write “Estimated globally averaged atmospheric lifetimes against wet deposition for formic and acetic acid in the boundary layer is between 1 and 2 days respectively (Paulot et al., 2011).” – This is not clear. Are both of the estimated atmospheric lifetimes between 1 and 2 days? If so, “respectively” isn’t needed. Either way, though, it should state “are between”. . .

The text has been amended in the following manner:

Line 476–478: “Estimated globally averaged atmospheric lifetimes against deposition for both formic and acetic acid in the boundary layer are between 1 and 2 days (Paulot et al., 2011).”

36) Line 396 – “14C” (with a superscripted 14) or “carbon-14” (without a superscript).

Line 479: A superscripted 14 has been added to the front of C, the text now reads “$^{14}$C”.

37) Lines 405, 407, 410, 412, 443, 446, Figure 5, etc. etc. – sometimes “Factor” is capitalized in reference to one of the four factors, and sometimes it isn’t. This should be consistent throughout.

The text has been amended throughout, when referring to a specific factor, “Factor” is now capitalized.

38) Line 427 – “Factor”, not “Factors”. Also, there is an extra period in this sentence: “. . . speed (Fig. S2.).”

Line 510: “Factors” is now singular “Factor” and the extra period has been removed and reference to Fig. S2 has been removed and replaced with the correlation coefficient between the Marine Cryosphere Factor and wind speed (as requested by the second referee).

39) Lines 430-431 – Despite what the papers might claim, MSA is not measured in particle phase, but rather they measured the methanesulfonate ion, CH3SO2+. It would be better to simply indicate that the presence of gas-phase MSA has been indicated by the observation of methanesulfonate ion in particles.

The text has been amended in the following manner:

Line 513–515: "The presence of gas-phase MSA has been indicated by the observation of the methanesulfonate ion, which has been previously measured in the particle phase at Villum in February–May 2015 (Dall'Osto et al., 2018; Nielsen et al., 2019)."

40) Line 438 – ". . . Dibb and Arsenault (2002) measured levels. . ."

Line 522: The word "had" has been removed.

41) Line 440 – "matter, e.g.,"

Line 524: A comma has been added after "matter".

42) Line 444 – The sentence "These trajectories and trajectory frequency maps were calculated as described in Sect. 2.4." isn't necessary.

Line 529–530: This sentence has been removed.

43) Lines 460-461 – recommend: "One of the source areas identified in Fig. 6 is southeast of Villum, and a CPF analysis indicated high contributions (of what?) were observed when the winds were from south of Villum (Fig. S8a)." – this sentence needs a little clean-up for readability and clarity.

This sentence has been amended in the following manner:

Line 554–556: "One of the source areas identified in Fig. 7 is southeast of Villum, and a CPF analysis indicated high contributions of the Marine Cryosphere Factor were observed when the wind direction was south of Villum (Fig. S5b)."

44) Line 469 – Recommend changing "Most of its components, particularly acetone and formaldehyde, are known. . ." to simply "Acetone and formaldehyde are known. . ."

Line 565: The sentence has been amended following the referee's suggestion.

45) Lines 483, 484, 487, 508, 545 – do you mean "labile [organic] carbon"?

Throughout the text "liable carbon" has been replaced with "labile organic carbon".

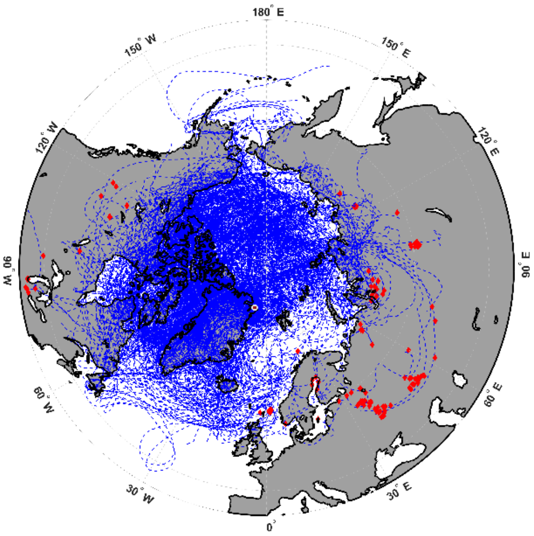
46) Line 531 – Circle should be capitalized.

Line 632: Circle is now capitalized.

47) Figure 4 – "red stars" – the resolution doesn't merit calling these stars. They're mostly just dots.

This figure has been removed from the manuscript. The second referee asked for a more statistical analysis of the back trajectories with the active fires. We collocated back trajectory endpoints with active fires with 1° latitude/longitude and temporally within one hour. While there was evidence of active fires in North America and Eurasia occurring when an endpoint was near, the uncertainty in individual trajectories at 336 hours is too great to draw meaningful conclusions from this analysis.

We have included the figure in our response, this figure will not be included in the manuscript. Individual trajectories are indicated in the dashed blue lines and active fires occurring within 1° lat/lon and within one hour of trajectory endpoints are indicated in red.



We have amended the text for the Biomass Burning section.

Line 413: To examine the geographical origin of this factor, air mass back trajectories from the HYSPLIT model were calculated every hour during the peak of the Biomass Burning Factor (15 August–15 September 2018) and extending 336 hours (two weeks) backward in time. The trajectory length of two weeks was selected to account for the long lifetime of acetonitrile. Active fires during the period 15 August–15 September 2018 was provided by NASA's Fire Information for Resource Management System (FIRMS) (Schroeder et al., 2014). Active fires occurring with one hour and one-degree latitude/longitude of a trajectory endpoint was used to access the influence of active fires on the Biomass Burning Factor. While there was evidence of active fires in North America and Eurasia occurring near a trajectory endpoint with one hour, the uncertainty of a trajectory with a length of 336 hours is quite large (Stohl, 1998). Therefore, no meaningful conclusions can be drawn from this analysis other than the transport time of emissions influencing the Biomass Burning Factor is greater than two weeks, and that we are unable to capture these emissions with the current trajectory models with any confidence.

Supplement

48) Line 26 – Either "(5 s)" or "(5 seconds)" would be acceptable SI units.

SI Line 27: "sec" has been changed to "s".

49) Table S1 - The way the authors divided up the seasons here seems oddly arbitrary. Why is "summer" only two months long, while autumn is three months? And changing seasons on the 7th of a month is oddly arbitrary. As well, it would be preferable to separate the measurement and units in the first column with a comma rather than a slash. Also, use either "autumn" or "fall" but not both in the table title and header. Lastly, the start and stop

dates in the title are not consistent with the dates given on Line 128 of the main text. Please make these consistent.

The authors admit this is an unusual set of dates for dividing seasons. This is because the data is split into three periods by interruptions (mainly due to power failure) as seen in Figure 1. Therefore, the authors divided the seasons according to these groups. “Fall” in the table has been changed to “Autumn”, the manuscript has also been checked throughout for consistency regarding this naming. The slashes between measurement and unit has been removed and replaced with a comma. The dates have been made consistent with the dates listed in the manuscript. See the updated Table 1 below.

**Table S1.** Statistics for meteorological parameters (mean ± s.d.) for all seasons, spring (April 4 – June 8), summer (June 9 – August 6), and autumn (August 7 – October 25). During the campaign, there were several large gaps in the data, most noticeably one in July and one in August, as seen in Fig. 1. The seasons are therefore divided based on the continuous collection of data uninterrupted by large missing gaps. The seasons roughly correspond to the conventional definition of seasons.

| | All Seasons | Spring | Summer | Autumn |
|---|---|---|---|---|
| Wind Direction, ° | 207.5 ± 89.0 | 202.4 ± 91.8 | 189.3 ± 2.6 | 223.8 ± 81.2 |
| Wind Speed, m $s^{-1}$ | 3.3 ± 2.6 | 3.1 ± 2.4 | 3.5 ± 2.4 | 3.4 ± 2.7 |
| Temperature, °C | -6.5 ± 9.6 | -13.8 ± 9.0 | 2.2 ± 4.1 | -7.0 ± 7.9 |
| RH, % | 77.4 ± 12.6 | 74.6 ± 10.6 | 78.0 ± 15.6 | 79.1 ± 11.4 |
| Radiation, W $m^{-2}$ | 174.9 ± 163.9 | 222.3 ± 146.3 | 295.9 ± 4.2 | 57.0 ± 97.4 |
| Pressure, hPa | 1010.6 ± 9.0 | 1014.8 ± 8.6 | 1007.5 ± 6.5 | 1009.6 ± 9.5 |
| Snow Depth, m | 0.9 ± 0.6 | 1.4 ± 0.1 | 1.1 ± 0.4 | 0.3 ± 0.4 |

50) Tables S2-S4 – It is unclear why June, July and September are included here, but not August and October. In the text, Villum Research Station is referred to as “Villum”, not VRS. It should be the same here, or spelled out in full. The vertical alignment of these tables is off, with the numbers right justified, and the headers left-justified, making it difficult to know which values go with which headers. As well, some of the compounds listed in the left-hand column blend together. Either increase the spacing, or shorten the names (i.e., MEK, DMS, etc.) to limit the amount of word-wrapping. Formic Acid across the head is also rather unfortunately split. Finally, the “All correlations, apart from . . .” in the titles should just be included as a footnote.

The correlation analysis was performed for one month from each season which had a good data coverage for the parameters being compared. We have added Table S2, which details the number of measurement hours for each compound for each month that displays this. VRS has been changed to Villum in the table headers. The columns of Tables S3, S4, and S5 are all now left aligned. DMS and MEK are now used in the left column and top row, which eliminated all word-wrapping. The text “All correlations…” has been made a footnote. Please see the updated tables in the SI, considering the length of the tables they are not included here.

51) Figure S1 – the text suggests that there were times when the wind speeds were < 2 m/s, but this is not included in the figure. Please either include these, or justify their omission. Also, the resolution on the figure does not allow for the reader to discern anything > 14-18 m/s

(blue). Either improve the resolution, or change the legend to eliminate the highest wind speed categories.

The figure has been remade to include all wind speeds and the intervals of the color bar have been changed to allow the relative wind speeds to be discerned. The figure has been expanded for individual wind roses for each season. The figure was also saved at a higher resolution (300 vs 600 DPI). See the updated Fig. S1 below.

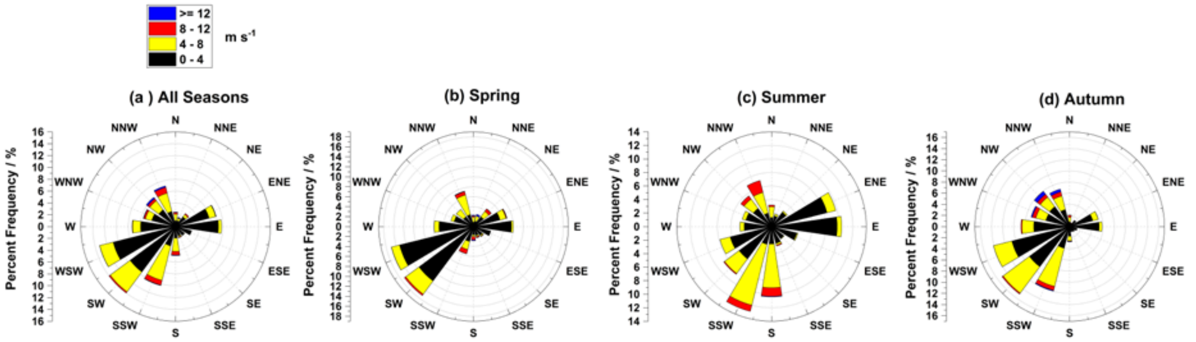


**Fig. S1.** Wind Rose for mean wind speed at 5 min time resolution for (a) all seasons, (b) spring, (c) summer, and (d) autumn. The y-axis represents the percent frequency of wind direction in percent and the colors indicate mean wind speed in m $s^{-1}$. The seasons follow the selection outlined in Table 1.

52) Figure S2 – “Time series of meteorological parameters. . .”; consider adding wind direction to this figure as well.

Wind direction has been added to this figure. The figure was also saved at a higher resolution (300 vs 600 DPI). Please see the updated Fig. S2 in the Supplement.

53) Figure S4 (and S5) – there is a lot of information shown that is repetitive and unneeded to the right of each satellite image, and as a result the majority of the important de- tails are illegible. Remove the unnecessary parts, and make higher res and/or larger versions of the plots, and label the leads and the station in the image(s). As well, the labels a-f should be moved to the top left, or top right, or could be included inside the images in white for clarity. Lastly, here and throughout the manuscript, re: the ACP style guide, dates should be in the form dd month yyyy (or simply dd month).

The old Figures S4 and S5 have been removed from the manuscript. Both referees raised concerns about the legibility of these two figures, therefore, we have removed them and directed the reader to the website where they were obtained (Line 324). We feel they add valuable information about the origin of the elevated DMS periods but displaying them in a meaningful manner proved problematic.

Throughout the manuscript, texts and figures have been amended to display the correct date format for ACP.

54) Figure S6 – caption “A new trajectory was [calculated/generated] every 24 hours.” The back trajectory trace colors in the plots should have a legend or be described.

The Fig. S6 caption now reads:

"**Fig. S6.** HYSPLIT back trajectory analysis for **(a)** May 2st – 6th **(b)** May 16th–20th arriving at 100 m above ground level extending 72 hours backward in time. The colored trajectories represent a new trajectory started every 24 hours after the last day of each period until the first day, in descending order the trajectories are red (last day), blue (fourth day), green (third day), light blue (second day), and purple (first day)."

The figure was also remade at a higher resolution and with panel labels ((a) and (b)) located at the top left of each panel.

55) Figure S7 – The caption should include the fact that this is from the PMF analysis.

The caption for Fig. S7 now reads:

"The ratio of $Q_{true}$ to $Q_{theo}$ versus the number of factors for the PMF analysis."

The caption for Fig. S8 now reads:

"Conditional probability function roses for **(a)** Biomass Burning Factor, **(b)** Marine Cryosphere Factor, **(c)** Background Factor, and **(d)** Arctic Haze Factor."

56) Figure S8 – plots (a) and (c) have the same size CPF scale, but different numbers of ticks and significant figures. They should be the same.

Figure S8 has been updated to include all factors. The Biomass Burning, Background, and Arctic Haze Factors now all have the same scale, and all panels now have the same number of significant figures for the scale.

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