- 1 Reply to Interactive Comment on "Atmospheric VOC measurements at a High Arctic site:
- 2 characteristics and source apportionment" from anonymous Referee # 2
- 3 This manuscript by Pernov et al. reports atmospheric non-methane volatile organic compounds
- 4 (VOCs) measurements at Villum Research Station at Station Nord, Green- land, from April to
- 5 October 2018. Given the scarcity of VOC measurements in the Arctic and the significance of
- 6 VOCs in the background atmosphere (formation of ozone, CO, and aerosols), this study will
- 7 make a valuable contribution to the body of literature. The manuscript is overall well written
- 8 and structured. My main concern is that the figures do not support the discussion and
- 9 conclusions (see comments below). Additionally, the introduction could be more succinct.
- 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
- 15 errors in the manuscript below with referee's comments numbered and the author's responses
- 16 in blue. New references are highlighted in yellow.
- Several of the reviewer's comment address the same concerns, where appropriate we havegrouped these comments together and responded to them all with one reply.
- 19 The referee suggested the Introduction could be more succinct. We have removed the following20 lines from the introduction in order to reduce the wordiness:
- 21 Lines 40–44
- 22 Lines 64–65
- 23 Lines 73–75
- 24 1) Diurnal variation
- 25 The authors say that certain compounds (e.g. DMS and OVOCs) follow a diurnal cycle. This
- is not shown in Figure 1 and I would like to see a Figure describing, for each compound ofinterest, the mean diurnal cycle per season.
- Line 217: "certain compounds (DMS and OVOCs) revealed a diurnal cycle that closely followsradiation". Please make a Figure to prove this.
- Line 219: "summer when a diurnal pattern following sunlight was observed". Same as above,please demonstrate this.
- Lines 225-227: "a clear diurnal variation was observed in the period July-August, with peak
- 33 mixing ratios occurring around midday (Fig. 1 a, c, d, e). The diurnal variation was less
- 34 pronounced in April-May and September-October, highlighting the dependence on sunlight".
- 35 None of this is shown in Figure 1.
- Line 255: "DMS showed a clear diurnal cycle during sea ice melt in the summer monthscorrelating with sunlight intensity". Prove/illustrate it.
- Lines 259-261: see comment above. As is, Figures S4-S6 do not do a good job at showing this.

- 39 Lines 286-287: "In addition to the previously mentioned dependence on the diurnal variations
- 40 of sunlight, providing strong evidence of a local photochemical source". Again, this has not
- 41 been demonstrated.
- Lines 383-384: "This factor shows an enhanced diurnal variation with a clear correlation to sunlight during the summer months (Fig. 5, Top)". Again, Figure 5 does not illustrate this.
- Lines 406-407: "Periods of high contributions and clear diurnal pattern by the Marine Cryosphere factors starts on June 23". I don't see the "clear diurnal pattern".
- 46 We agree with the referee's concern about a lack of the diurnal nature being illustrated properly.
- 47 We have therefore added two figures in the main text (Fig. 2 and 6) to display diurnal profile
- 48 for each of the relevant compounds and the four factors, respectively, during the summer
- 49 months, as well as the diurnal profiles for each season (as requested by the referee) in the
- 50 Supplement (Fig. S3, S4, and S5). We have amended the text throughout to reference these two
- 51 figures and removed references to a diurnal profile when they do not pertain to these figures.

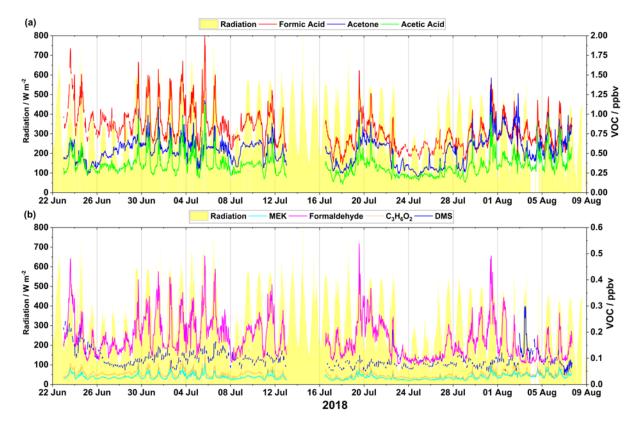
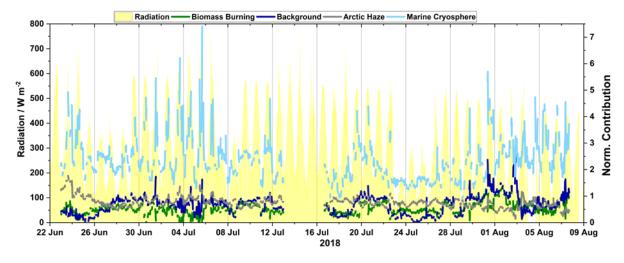


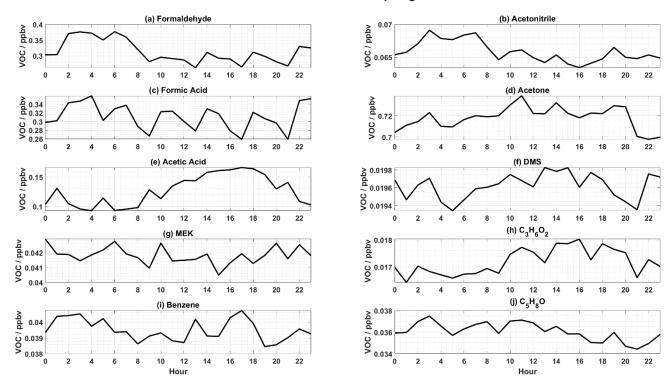


Fig. 2. Diurnal profile for (a) formic acid, acetone, acetic acid, and radiation and (b) MEK,
 formaldehyde, C₃H₆O₂, DMS, and radiation during the period 22 June–09 August.



55

Fig. 6. Time series of the four factors from 22 June–09 August displaying the diurnal profiletogether with radiation.



Diurnal Profile for Spring

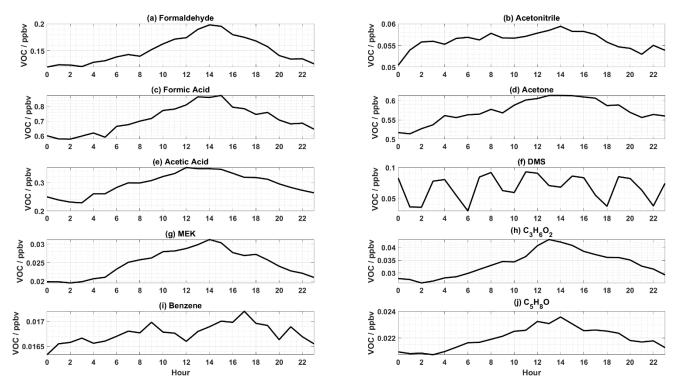
58

59 Fig. S3. Diurnal profile for the spring (April–May) of (a) formaldehyde, (b) acetonitrile, (c)

formic acid, (d) acetone, (e) acetic acid, (f) DMS, (g) MEK, (h) $C_3H_6O_2$, (i) benzene, (j) C_5H_8O .

61 Data were averaged to hourly medians.

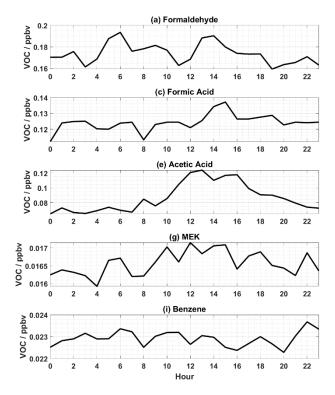
Diurnal Profile for Summer





63 Fig. S4. Diurnal profile for the summer (June–August) of (a) formaldehyde, (b) acetonitrile,

64 (c) formic acid, (d) acetone, (e) acetic acid, (f) DMS, (g) MEK, (h) C₃H₆O₂, (i) benzene, (j)
65 C₅H₈O. Data were averaged to hourly medians.



Diurnal Profile for Autumn

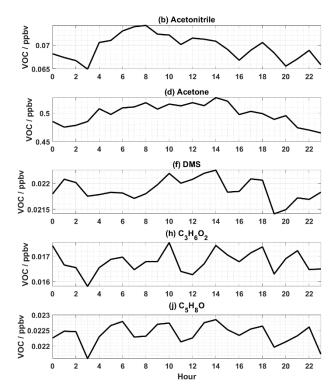


Fig. S5. Diurnal profile for the autumn (September–October) of (a) formaldehyde, (b)
acetonitrile, (c) formic acid, (d) acetone, (e) acetic acid, (f) DMS, (g) MEK, (h) C₃H₆O₂, (i)
benzene, (j) C₅H₈O. Data were averaged to hourly medians.

We have amended the text to mention the diurnal profiles for relative compound in the relativeseason:

Line 269–280: "For the ten selected VOCs, time series of mixing ratios during the entire 72 73 measurement period are displayed in Fig. 1a-f. During the spring (April-May), certain compounds (benzene and C₅H₈O) exhibited a maximum and thereafter a decreasing pattern, 74 75 similar to the timing and profile of the Arctic Haze phenomena. During the spring, compounds 76 did not display a diurnal profile except for acetic acid (Fig. S3) Whilst in summer (June-August), OVOCs revealed a diurnal cycle that closely follows radiation (Fig. 2 and S4). 77 Compounds of non-photochemical origin (benzene and acetonitrile) also displayed a slight 78 diurnal pattern, which could possibly be due to entrainment from aloft (Fig. S4). Interestingly, 79 80 several compounds (formaldehyde, formic acid, and acetone) peaked in the spring with decreasing levels until the summer when a diurnal pattern following sunlight was observed 81 (Fig. 1, 2, S4). During the autumn (September–October), all compounds were low except for 82 acetone and acetonitrile (Fig. 1) and only acetic acid displayed a diurnal profile (Fig. S5). The 83 levels, seasonal patterns, and comparison with other studies of these compounds will be 84 discussed below." 85

86 2) Springtime DMS

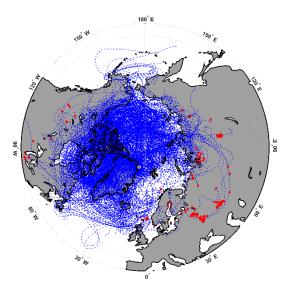
87 I have difficulties reading satellite images (Figures S4 and S5). The caption says that the 88 presence of open leads can be seen southwest of the station but I don't even know where the 89 station is. Then, according to the authors, "the back-trajectory calculations confirmed that, during the DMS emission episodes, the air masses (...) traversed over the open leads before 90 reaching the station". First of all, what is the meaning of the different colors? I do not 91 92 understand Figure S6. Then, this Figure does not show that air masses traversed over the open leads. If you want to show this, then please consider combining satellite images and back-93 trajectories on a single Figure. 94

95 Figures S4 and S5 have been removed from the manuscript. Both reviewers raised concerns 96 about the legibility of these two figures, therefore, we have removed them and directed the reader to the website where they were obtained (http://ocean.dmi.dk/arctic/nord.php). We feel 97 they add valuable information about the origin of the elevated DMS periods but displaying 98 them in a meaningful manner proved problematic. We have left the HYSPLIT back trajectories 99 in Fig. S6 in and updated the figure caption to indicate the meaning of the different trajectories, 100 the text now reads : 101 "HYSPLIT back trajectory analysis for (a) May 2nd- 6th (b) May 16th-20th arriving at 100 m 102

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)."

- 107
- 108 3) Biomass burning

- Contrary to what the authors say, Figure 4 does not show "evidence of overlap between air 109 mass history and active fires during this period". Figure 4 shows all fires from mid-August to 110 mid-September and all back-trajectories. This does not prove that a given fire existed at the 111 time an air mass traveled in the area. I expect a more thorough statistical analysis here. In order 112 to link the fires up with back trajectories, you could for instance cross check the latitudes and 113 longitudes to a, let's say, 1-degree accuracy. If a longitude and latitude match exists between a 114 fire and a back-trajectory, then check if the time of the fire product and the back-trajectory 115 were within, let's say, 1 hour. Thus, a match is completely defined as a back-trajectory crossing 116
- 117 over a fire within 1 hour and within 1-degree difference.
- Lines 206-207: "The trajectory length was varied between 240 and 336 hours". Why did you use two different trajectory lengths? Additionally, I would recommend the use of shorter back trajectories (typically 5-7 days max) as uncertainties increase with time along the way (Stohl, 1998). I would also like to see a more critical discussion on back trajectories; they only give a general indication of source regions.
- Line 351: "336 hours backward in time". That's too long to my point of view. Use max 5-7days.
- We collocated back trajectory endpoints with active fires with 1° latitude/longitude and temporally within one hour as the reviewer requested. While there was evidence of active fires in North America and Eurasia occurring when an endpoint was near (see figure below), as the reviewer pointed out the uncertainty in individual trajectories at 336 hours is too great to draw meaningful conclusions from this analysis. Therefore, this figure has been removed from the manuscript.
- 131 We have included the figure in our response, this figure will not be included in the manuscript.
- 132 Individual trajectories are indicated in the dashed blue lines and active fires occurring within
- 133 1° lat/lon and within one hour of trajectory endpoints are indicated in red.



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136 We have amended the text for the Biomass Burning section.

Line 420–431: "To examine the geographical origin of this factor, air mass back trajectories 137 from the HYSPLIT model were calculated every hour during the peak of the Biomass Burning 138 Factor (15 August-15 September 2018) and extending 336 hours (two weeks) backward in 139 time. The trajectory length of two weeks was selected to account for the long lifetime of 140 acetonitrile. Active fires during the period 15 August–15 September 2018 were provided by 141 NASA's Fire Information for Resource Management System (FIRMS) (Schroeder et al., 2014). 142 Active fires occurring with one hour and one-degree latitude/longitude of a trajectory endpoint 143 was used to access the influence of active fires on the Biomass Burning Factor. While there 144 was evidence of active fires in North America and Eurasia occurring near a trajectory endpoint 145 within one hour, the uncertainty of a trajectory with a length of 336 hours is quite large (Stohl, 146 1998). Therefore, no meaningful conclusions can be drawn from this analysis other than the 147 transport time of emissions influencing the Biomass Burning Factor is greater than two weeks, 148 and that we are unable to capture these emissions with the current trajectory models with any 149 confidence." 150

151 4) Spatial origin of the Marine Cryosphere factor

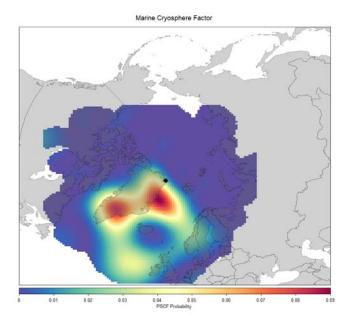
Figure 6 (trajectory frequency) shows more frequent air masses from coastal regions but does not show that these areas are responsible for enhanced marine cryosphere factor. It does not support this sentence in the conclusion "Back trajectory analysis yielded MIZs around the coasts of Greenland and the Arctic Ocean as source regions". I suggest a Potential Source Contribution Function (PSCF) analysis to determine probable locations of emission sources.

Lines 443-454: see comment above on Figure 6 and the fact that it does not show that coastal regions are responsible for enhanced marine cryosphere factor.

At the time of preparation of this manuscript, the authors did not possess the tools or 159 knowledge about how to perform a PSCF. The authors agree this would be the appropriate 160 method for determining source regions for the Marine Cryosphere Factor. Therefore, the 161 authors have become familiar with the programming language R and the R package Openair 162 (Carslaw and Ropkins, 2012). Using this package, the authors were able to produce a (PSCF) 163 for source region analysis of the Marine Cryosphere Factor. We have replaced the trajectory 164 frequency map for the summer season with a PSCF map using data from the entire campaign. 165 A PSCF for the summer period was also produced and compared to the entire campaign which 166 produced similar results. Inclusion of the entire campaign data provides a more robust 167 statistical calculation of the PSCF; therefore, we have chosen to perform the PSCF for the 168 entire campaign. 169

170 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.
- 172 7) with the PCSF as seen below and updated the figure caption accordingly.



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188

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).

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

Line 223–250: "To investigate source regions, the R package Openair (Carslaw and Ropkins, 180 181 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., 182 2017) at 100 m altitude and 120 hours backward in time using Global NOAA-NCEP/NCAR 183 reanalysis data archives on a 2.5° resolution. A PSCF, shown in Eq. (3), calculates the 184 probability that an emission source is located in a grid cell of latitude *i* and longitude *j*, on the 185 basis that emitted material in the gird cell *ij* can be transported along the trajectory and reach 186 187 the receptor site.

$$PSCF = \frac{m_{ij}}{n_{ij}}$$

 \mathbf{m} .

(3)

189 Where n_{ij} is the number of times a trajectory has passed through grid cell *ij* and m_{ij} is the number 190 of times that a concentration was above a certain threshold value, in this case, the 90th 191 percentile. To account for uncertainty in cells with a small number of trajectories passing 192 through, a weighting function was applied (Carslaw and Ropkins, 2012).

- We have amended the text in the Marine Cryosphere Factor section to reflect his new analysismethod:
- 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

- 201 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
- 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
- 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
- 208 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
- 210 source region for the Marine Cryosphere Factor."
- 211
- 212 Line-by-line comments:
- 5) Line 10: "we report a long-term dataset". The authors report measurement from April to
 October 2018, i.e., less than a year. This is not what I would call a "long-term dataset".
 Please edit this sentence.
- We have replaced "long-term" with "multi-season" throughout the text to better reflect theduration of the dataset.
- 218 6) Line 33: Define VOCs and NOx.
- Line 35 and 36: "NO_x" has been defined as "nitrogen oxides" and "VOCs" have been defined
 as "volatile organic compounds".
- 221 7) Line 49: Define DMS.
- Line 52: "DMS" has been defined as "dimethyl sulfide".
- 8) Line 108-109: "with low time resolution". Be more specific here. Gautrois et al. (2003)
 collected about one sample every 9 days. Additionally, the authors did not use a GC- MS,
 but a combination of GC-FID and GC-ECD.
- 226 The text has been amended to read:

Line 109–112: "Gautrois et al. (2003) reported long-term VOC concentrations for Alert, NU,
where a seven-year time-series of VOCs mixing ratios has been generated, although with a 9
day time resolution, using off-line techniques (GC coupled to flame ionization and electron
capture detectors)."

- 9) Line 110: You don't really explain why we need high time-resolved measurements of
 VOCs. Do you expect a high temporal variability? How about the global atmospheric watch
 reactive gases measurement network (Schultz et al., 2015) Aren't these measurements
 enough?
- 235

Line 111: Same comment as above. In the previous sentence, you highlight the need for long-term measurements of VOCs in the Arctic. While a substantial contribution to the literature, you "only" report several months of data. You could perhaps emphasize more the high temporal frequency of your measurements. The reviewer is correct in pointing out the lack of explanation for the need of high-time resolved measurement. We do expect a high temporal variability, especially in the summer when meteorological conditions can change rapidly. Schultz et al. (2015) is an important piece of literature, which highlights the need for high-time resolved measurements, therefore we have highlighted the need for high-time resolved measurements with the addition of the following paragraph:

Line 112–120: "High time resolution measurements are of vital importance for the study of 246 Arctic atmospheric chemistry. For instance, diurnal studies can only be accomplished with a 247 fast response instrument, as grab samples and time-integrated samples (i.e., adsorbent tubes) 248 will not capture the variability on short enough time scales (de Gouw and Warneke, 2007). 249 Understanding the effects of meteorological parameters on VOCs levels requires an instrument 250 response which is shorter than the transient event being observed. Also, flux measurements can 251 only be achieved through fast responding instrumentation (Müller et al., 2010). The study of 252 short-lived compounds, such as reactive halogen species, and their interactions with VOCs is 253 only possible on short timescales. Finally, global networks have highlighted the need for a 254 quick turnaround in the delivery of atmospheric species for the validation of global atmospheric 255 composition forecasting systems (Schultz et al., 2015)." 256

The reference de Gouw and Warneke, 2007, Müller et al., 2010, and Schultz et al., 2015 arenew and have been added to the reference list.

- 10) Line 123: Did you filter data for local contamination? If so, how?
- Line 157: How exactly did you remove the influence of local pollution. What criteria did you use for wind speed and direction?

We have added Section 2 "Quality Control Procedure" in the Supplement, which describeshow local pollution was identified and removed, as seen below.

264 SI Line 35– 52: "Quality Control Procedure

Data were quality controlled by analysis of PNSD, ozone, wind direction and speed, and 265 internal activity logs. Local pollution at Villum can arise from activity around the measurement 266 site (e.g., passenger vehicles, all-terrain vehicles, snowmobiles, and heavy machinery) as well 267 as from activities from Station Nord (e.g., waste incineration, vehicular activity, and aircraft 268 landing, idling, and take off). Internal activity logs of visits to the measurement building were 269 used to highlight periods when human activity could affect the measurements, periods where 270 VOC levels were elevated over background levels for the duration of the visit to the station 271 were removed. Measurements of PNSD and ozone were analyzed, in tandem, for sharp and 272 sudden increases in the ultrafine mode (< 100 nm) aerosol particles and concurrent sharp and 273 274 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 275 276 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 277 suspected of influencing the measurements were visually inspected by a panel of three persons, 278 a consensus was required before data were removed. Data were also quality controlled for 279 abnormal levels of instrumental parameters (i.e., E/N ratio, drift tube temperature, pressure, 280 and voltage), periods with large deviations from nominal values were removed. Certain 281 compounds (DMS, formic acid, and acetic acid) exhibited a slow return to nominal values after 282

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."

286

11) Line 125, Table S1: I have a hard time understanding how the seasons were defined. Skov et al. (2020) recently used a different (and more straightforward) definition: winter from December to February, spring from March to May, summer from June to August, and fall from September to November.

The authors admit this is an unusual set of dates for dividing seasons. This is because the data 291 is split into three periods by interruptions (mainly due to power failure) as seen in Figure 1. 292 Therefore, the authors divided the seasons according to these groups to include uninterrupted 293 294 collected data. Additionally, "Fall" in the table has been changed to "Autumn", the manuscript 295 has also been checked throughout for consistency regarding this naming. The slashes between 296 measurement and unit has been removed and replaced with a comma. The dates have been 297 made consistent with the dates listed in the manuscript. An updated Table 1 along with its 298 caption is presented 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.

304

	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	$1007.5\pm\ 6.5$	1009.6 ± 9.5
Snow Depth, m	0.9 ± 0.6	1.4 ± 0.1	1.1 ± 0.4	0.3 ± 0.4

305

12) Line 137-138: "measurements were interrupted for short periods ranging from days to
weeks". Could you please add a Table summarizing, for each month, the number of hours
of operation?

309 Table S2 has been added to the Supplement summarizing the number of hours the instrument310 was in operation for each compound for each month of the campaign. The following text has311 been added:

Line 151–152: "Table S2 summarizes the total number of operational hours for each compoundfor each month of the campaign." See Table 2 below.

Table S2. Total hours of operation of the PTR-ToF-MS for each month of the campaign and
 for each compound. Periods removed through the QC procedure are not included.

316

April May June July August September October

Formaldehy de	374	601	288	661	417	443	403
Acetonitrile	229	601	288	661	417	443	403
Formic Acid	349	601	288	641	417	443	403
Acetone	376	601	288	661	417	443	403
Acetic Acid	375	577	288	661	417	411	359
DMS	300	577	169	391	357	443	377
MEK	376	601	288	661	417	443	403
$C_3H_6O_2$	327	601	288	661	417	443	403
Benzene	376	601	288	661	417	443	403
C_5H_8O	376	601	288	661	417	443	403

317

13) Line 144: "within the analytical uncertainties". Please refer to Table 1 here. Additionally,how often did you perform a calibration?

VOC mixing ratios were quantified using the kinetic rate reaction method (Supplement Sect.
1) and were validated against a certified reference standard at the beginning of the campaign.
We have added a reference to (Holzinger et al., 2019) in the sentence, which refers to the
quantification method and the reference standard. The phrase "using the reaction kinetics
quantification method." has been added to the Supplement on Line 2. Table 1 has been referred
to in the sentence. The main text was amended to:

Line 153: "Data generated by the PTR-ToF-MS instrument were processed with the PTR-MS Viewer software v. 3.2.12 (Ionicon Analytik). Mass calibrations and VOC mixing ratios were calculated by the PTR-MS Viewer, based on the reaction kinetics quantification method (Sect. S1). The instrument quantification was validated against an external gas-phase calibration standard (Apel-Riemer Environmental), a comparison between standard and instrument mixing ratios yielded percent errors that were within the analytical uncertainties (Table 1), therefore we are confident in the quantification method (Holzinger et al., 2019)."

14) Lines 179-181: Did you perform a sensitivity test? How does changing concentrationsbelow LOD and missing concentrations influence the PMF results?

The authors performed an innumerable amount of PMF runs, varying treatment of data below 335 the LOD, treatment of missing values (either removing the sample or replacing with median 336 for the dataset), treatment of the uncertainty matrix, number of species included in the model 337 (species were systematically removed and added), threshold values for species categorization, 338 and number of factors. While each model run, produced unique results, the overall shape of the 339 factor time series and species profile for each factor was consistent with the final reported 340 model setup. The optimal model solution (as configured in the study) was deemed robust to 341 these different variations of the dataset. 342

343 The text has been amended to include this description of model robustness.

Line 203–211: "Numerous sensitivity runs were performed to evaluate the validity of this data

345 preparation protocol including varying the treatment of data below the LOD (replacing with

half of the LOD or leaving as is), treatment of missing values (removing the sample or replacing

- 347 missing species with the median), treatment of the uncertainty matrix, number of species
- included in the model (species were systematically removed or added to observe their influence

- on the model solution), threshold values for species categorization, and the number of factors.
- Each variation of the input data, of course, produced a unique solution, however, the overall
- 351 shape of the time series and factor contributions profile was consistent with the solution present
- in this study. The optimal model solution, for the configuration present here, was therefore
- 353 deemed robust to these variations of the input data and provided acceptable diagnostics."
- 15) Line 191: "automatic weather station placed close to the measurement site". How close?Be more specific.
- 356 The text now states the distance of the automatic weather station from the measurement site.
- **357** Line 218–219: "Meteorological data including temperature, relative humidity, wind speed,
- 358 wind direction, pressure, radiation, and snow depth were generated by an automatic weather
- **359** station placed ~44 meters away from the measurement site."
- 16) Line 257: "Elevated DMS mixing ratios". What do you mean by "elevated"? Be morespecific.
- 362 The text has been amended to better describe the elevated mixing ratios of DMS during these363 periods. The main text now reads:
- Line 321: "Elevated DMS mixing ratios were observed for two short periods of a few days' duration in May (1–5 May and 16–19 May), where DMS mixing ratios increased an order of magnitude from ~0.02 to >0.2 ppbv (Fig. 3a and b)."
- 17) Line 273: "illustrated here by changes in wind speed". I would expect changes in wind direction to be a more useful tracer of change in meteorological conditions.
- The authors also expected wind direction to be more useful tracer of meteorological conditions, 369 however, for the two episodes of elevated DMS, changes in wind speed appear to be a better 370 indicator than wind direction. To reflect this, we have added wind direction to Fig. 3 in the 371 manuscript (see below). For the first episode, the wind direction is quite variable while 372 increased wind speeds are observed during depletions in acetone and elevations of DMS, and 373 it is unfortunate meteorological data are missing on 3rd and 4th of May. For the second episode, 374 the wind direction does change concurrently with an increase in wind speed, although 375 throughout the episode wind direction is also variable with contributions from the north and 376 the east. We have mentioned this in the text: 377
- Line 338–339: "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.
 380 3)."

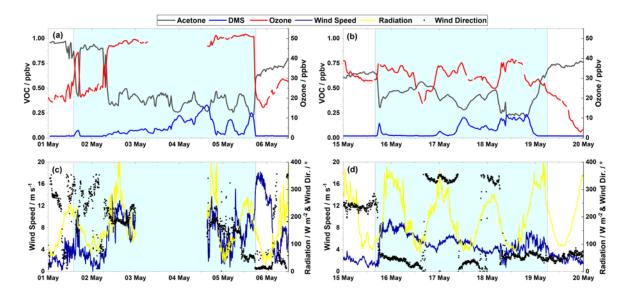


Fig. 3. Left: The first period of elevated DMS mixing ratios (May 1–5). Right: The second period of elevated DMS mixing ratios (May 15–19); (a) and (b) mixing ratios of acetone, DMS (left axis), and ozone (right axis); (c) and (d) wind speed (left axis) and radiation and wind direction (right axis). The shaded area represents episodes of elevated DMS mixing ratios.

18) Line 344: "a sink during the summer". A sink of what?

- Line 414: "Increased areas of open water in the Arctic also act as a sink for acetonitrile duringthe summer (de Gouw et al., 2003)."
- 19) Lines 426-427: "Although, the variation of the Marine Cryosphere Factors seems not to be
 driven mainly by the dependence on horizontal wind speed (Fig. S2)". Figure S2 does not
 illustrate this. What is the correlation coefficient between the Marine Cryosphere Factor
 and wind speed?
- The reference to Fig. S2 has been removed and replaced with the correlation coefficient
 between the Marine Cryosphere Factor and wind speed (R=-0.04) to better illustrate the lack
 of dependence between the two.
- Line 509–510: "Although, the variation of the Marine Cryosphere Factor seems not to be driven
- mainly by the dependence on horizontal wind speed (R=-0.04)."
- 20) Lines 428-429: "given the distance of the measuring site from open water". What is thedistance between the station and open water?
- 400 The fjord immediate to the station is located ~1.7 km away, during the summer this is mostly
- 401 ice free, although is prone to freeze-ups when the temperature drops below zero for several
- 402 hours. The station is located on a peninsula which is surrounded by sea ice throughout the year,
- 403 taking this sea ice into account, open water is ~25 km away. The following text has been added:
- 404 Line 510–513: "Marine microorganisms produce DMS (Stefels et al., 2007; Levasseur, 2013),
- 405 and given the distance of the measuring site from open water (taking sea ice into account the,
- 406 station is approx. 25 km distance from open water), it is proposed that the majority of DMS
- 407 produced is already oxidized to MSA and other products when reaching the station."

- 408 21) Section on Arctic Haze: please mention/discuss more clearly that you do not have data in
 409 wintertime, when Arctic Haze is expected to be at its maximum.
- 410 It has been made clear to the reader that our Arctic Haze Factor is only from spring and other411 studies present data from winter and spring. The following text has been added.
- 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
- 414 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

416 anthropogenic pollution in the Arctic."

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