Response to Anonymous Referee #1 on

"Large hemispheric difference in nucleation mode aerosol concentrations in the lowermost stratosphere at mid and high latitudes"

5 Christina J Williamson et al. 2021-03-16

We thank the referee for their insightful comments on the manuscript, which have improved the
 quality and clarity of the science presented. We have addressed the comments as detailed here
 below.

The size range 3-12 nm is called sometimes ultrafine particles, sometimes nucleation mode particles in the paper. I would strongly recommend avoiding the term ultrafine particles in this context, as the vast majority of air pollution scientists use this term for the whole <100 nm particle population. The authors have also applied the size ranges

15 scientists use this term for the whole <100 nm particle population. The authors have also applied the size ranges 3-12 nm and 12-60 nm for the nucleation and Aitken mode, respectively. In most studies conducted in the lower troposphere, the border between the nucleation and Aitken mode has been assumed to be somewhere in the range 20-30 nm, while the Aitken mode has been assumed to extend up to 90-100 nm. The lower size ranges for these modes applied here are acceptable because the whole particle population seems to be shifted to smaller sizes,

20 possibly due to lower concentration levels of aerosol precursors, compared with the lower troposphere. However, due to the somewhat unusual definitions of nucleation and Aitken mode size ranges, I suggest that the author add a couple of lines into the text to explain why they used such definitions for the nucleation and Aitken modes.

The referee is correct that our referral to 3-12 nm particles at "ultrafine" is out of step with the community at large, and revise the manuscript to adopt their recommendation of sticking with the defined term "nucleation mode". These changes are made in the following lines:

1 - title, "ultrafine aerosol concentrations" to "nucleation mode aerosol concentrations"

- 17 "mode of ultrafine aerosol" to "mode of aerosol smaller than 12 nm"
- 22 "ultrafine particles" to "nucleation mode particles" (both times)
 90, 92, 95, 102 "ultrafine" to "nucleation mode"
 257 "nucleation and/or growth of ultrafine aerosol" to "nucleation and/or growth of aerosol"
 276 "ultrafine" to "nucleation mode"

445 – "ultrafine particles lifetimes" to "nucleation mode particle lifetimes"

450 – "lifetime of ultrafine particles" to "lifetime of nucleation mode particles"
434, 439, 469, 473, 478, 480, 499 - "ultrafine" to "nucleation mode"

Regarding the boundary between Aitken and Nucleation mode being somewhat low compared to the lower troposphere standard, we agree with the referee that indeed the lower concentration of

- 40 precursors may well be a factor here. We also use this size range because the ATom dataset reports modes with these size limits, a conscious decision to allow direct comparison with observations by Anthony Clarke et al. of new particle formation and CCN in the remote marine atmosphere (Clarke et al., 1999;Clarke et al., 2013;Clarke and Kapustin, 2002;Clarke et al., 1998). This size range has subsequently been used in a number of publications discussion ATom
- 45 observations (Brock et al., 2019;Kupc, 2020;Williamson et al., 2019). We agree that it is worth adding further explanation of this in the manuscript and have done so at line 152 with the following statement:

"In this analysis we divide the aerosol size distribution into four modes: nucleation mode from 3-12 nm, Aitken mode from 12-60 nm, accumulation mode from 60-1000 nm, and coarse mode

- 50 above 1000 nm. We note that the 12 nm cut-off between nucleation and Aitken modes is defined at a smaller diameter than is often used for aerosol studies in the lower troposphere, and make this choice to allow for more direct comparison with previous datasets over the remote Pacific and Atlantic oceans (Clarke and Kapustin, 2010;Clarke et al., 1999;Clarke et al., 2013;Clarke and Kapustin, 2002;Clarke et al., 1998), and for consistency with the ATom data archive (Wofsy
- 55 et al., 2018) and other published works analyzing ATom size distributions (Brock et al., 2019;Kupc et al., 2020;Williamson et al., 2019)."

Somewhat related to the previous comment, it is not a good practice to talk about small particles (e.g. title of section 3) or larger particles (lines 438 and 481) without specifying what is exact meant by small or larger here.

60 We agree that our use of unquantified "small" and "large" descriptors of particles was poor practice and have amended this throughout the manuscript to be more precise. Changes made, and the corresponding line numbers are as follows:

57, 74, 174, 183, 208, 221, 235, 262, 265, 290, 294, 314, 335, 426, 427, 440, 458, 459, 461, 1005 – "small" to "nucleation mode"

65 195, 197, 199, 338 – "small" to "nucleation and Aitken mode"
356 – "small particle emissions" to "particle emissions"
47 – "larger particles" to "particles without details of the nucleation mode"
196 – "larger" to "accumulation and coarse mode"
251 – "larger particles" to "particles with diameters between 350 and 600 nm"
70 312 – "larger particles of tropospheric origin" to "accumulation mode particles of tropospheric

70 512 – larger particles of tropospheric origin to accumulation mode particles of tropospheric origin"
 438, 439, 445,462, 463 – "larger" to "accumulation mode"

In several places of the text, the authors talk about correlation and even its character (significant, slight).
Technically, correlation is an exact statistical quantity, which should be used just based on visual the appearance on how two variables seem to be connected with each other. I recommend using some other term than correlation in the text or, alternatively, to calculate the actual correlation coefficient and its level of significance.

With the discussion of correlation as it relates to nucleation mode number concentrations and SO2 concentrations, the referee is correct. We have therefore modified Fig. 6 to show log-log fits between number concentrations and SO₂ with correlation evaluated using r-squared values.

- 80 between number concentrations and SO₂ with correlation evaluated using r-squared values. Please note, in order to remove the effect of instrumental noise on correlations, we have analyzed the SO₂ and nucleation mode data to 0.1 Hz (the original fig. 6 showed 1 Hz data) following the analysis of noise in Williamson et al. (2019).
- 85 The new fig. 6 and caption are as follows



Figure 1: Aerosol correlations with SO₂. Number concentration dependence on SO₂ at 0.1Hz time resolution to reduce the effect of atmospheric and instrumental noise following Williamson et al. (2019). Number concentration as a function of SO₂ mixing ratios in May (high resolution SO₂ data was not available in other seasons) for the Northern and Southern Hemispheres (red circles and blue dots respectively). Data are divided into four modes by size: (a) nucleation (3-12 nm); (b) Aitken (12-60 nm); (c). accumulation (60-1000 nm); and (d) coarse (1000-4500). Log-log fits are given by the sold lines. Panel (e) shows R² values for these fits. Observations where SO₂ is below the limit of detection (1 pptv) are not shown.

95 Line 220 has been changed to read

"Number concentrations of nucleation and Aitken mode particles show some correlation with SO₂, whereas number concentrations of accumulation and coarse mode particles show little to no correlation with SO₂ (Fig. 6)"

100 Regarding correlation with O3, we have changed how we describe the SO2 behaviour at line 322 to the following:

"We do not observe SO₂ increasing with O₃ in the NH LMS (Fig. 9a), and therefore conclude that the NH excess SO₂ is not from OCS oxidation. SO₂ does increase with O₃ in the SH LMS (Fig. 9c), suggesting OCS oxidation may be contributing to these lower, background levels of SO₂."

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We also note that we had mistakenly referred to Fig. 7 here, and have therefore corrected it to Fig. 9.

We also noticed some repetition of text between line 319 and 333. We have corrected this by deleted the repeated text from line 325-329.

We have corrected the reference to no correlation between nucleation mode number concentrations and ozone from lines 330-332 to the following:

- "NPF can take place in descending air in the polar vortex in winter, and quasi-horizontal
 transport is a potential pathway for these particles to then enter the LMS. However, similar to SO₂, we do not observe nucleation mode particle concentrations increasing with O3 is (Fig. 9), suggesting they do not originate deeper in the stratosphere."
- Where we previously talked about lack off correlation, and slight anti-correlation between
 nucleation mode and biomass burning particles at line 427, we have corrected this to read:
 "Nucleation mode and biomass burning particles in the NH LMS did not follow the same trends with potential temperature (Fig. 10), which suggests that pyro-convection is not the major source of the nucleation mode particles we observed. In October, there is some indication in the vertical structure of nucleation mode number concentrations being suppressed at higher biomass burning
 particle mass concentrations, and enhanced at lower biomass burning particle mass
- concentration."

Line 361: The lifetime of nucleation mode particles depends both on the mean size of these particles and on the properties of the pre-existing particle population at sizes larger than the nucleation mode. Considering that both of these quantities are probably quite variable in the LMS, and especially quite different between NH and SH (as discussed in the supplementary material and illustrated in Figure S3), the authors should better justify the use of a single lifetime of 2 days for nucleation mode particles in their calculations.

We are grateful to the referee for drawing attention to this problem. On further examination, we concluded that calculating an overall lifetime of nucleation mode aerosol in the SH LMS, taking into account the orders of magnitude difference expected between plume (days) and non-plume (weeks-months) lifetimes, is not possible. For illustrative purposes we decided that potential concentrations calculated using a 2-day lifetime, and thus representing a lower-limit, were useful, and rewrote the paragraph beginning at line 360 as follows:

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"While particles lifetimes in non-plume LMS conditions are expected to be longer in the SH than the NH, nucleation mode particle lifetimes within aircraft plumes have been shown to be around 2 days (Schroder et al., 2000). The plume and non-plume lifetimes in the NH LMS are similar enough to justify the use of a 2-day lifetime in this region (Supplemental Materials S2). In the

145 SH LMS, there is a large difference between in-plume and non-plume lifetimes (from days to months). We use a 2-day lifetime here in the SH for illustrative purposes, but note that this is an over-estimate, and thus the resulting concentration will represent an underestimate. Calculating the volume of the region defined above as the LMS to be 3.12 x 10¹⁷ m³ and 3.65 x 10¹⁷ m³ for the NH and SH respectively, we get ambient concentrations of nucleation particle emitted by

aircraft in the LMS of 110 cm⁻³ in the NH and a lower limit of 0.26 cm⁻³ in the SH. Taking an average temperature and pressure of 220 K and 300 hPa for the LMS regions, and converting to standard temperature and pressure (STP), this gives maximum concentrations of 300 std. cm⁻³ for the NH and a lower limit of 0.71 std. cm⁻³ for the SH. Minimum aircraft emissions of nucleation mode aerosol reported by Brock et al. (2000) are a factor of 10 lower than the maximum we have used here, leading to concentrations of 30 and 0.07 std. cm⁻³ for the NH and SH respectively."

I am basically fine with the way this paper discusses and speculates about the particle origin in the study regions, including the mechanism of new particle formation. The only thing that could be improved the analysis of probability of (ion-induced) water-sulfuric acid nucleation is evaluated here based on the model framework developed by Kazil, Lovejoy and co-workers 10-20 years ago. Since then, detailed laboratory data on the same nucleation mechanisms has been obtained e.g. in CLOUD experiments, and these data have also been included in nucleation parameterizations. I am not saying that the authors should redo their calculations, but they could shortly discuss whether their conclusions are also consistent with this most up-to-date information on atmospheric water-sulfuric acid nucleation at cold temperatures.

While the CERN CLOUD collaboration has, to the best of our knowledge, not published thermochemical data (enthalpy and entropy of formation) of neutral and charged H₂SO₄/H₂O clusters from their chamber experiments, which would allow a direct comparison with the
experimental thermochemical data used in MAIA (Curtius et al., 2001;Lovejoy and Curtius, 2001;Froyd and Lovejoy, 2003;Hanson and Lovejoy, 2006), two indirect comparisons exist in the literature. We have added the following passage to the description of MAIA at line 173:

Aerosol nucleation rates calculated from the experimental thermochemical data of neutral and
 charged H₂SO₄/H₂O cluster formation that are used in MAIA (Kazil and Lovejoy, 2007) compare well with neutral and charged H₂SO₄/H₂O nucleation rates measured in the European Organization for Nuclear Research (CERN) Cosmics Leaving Outdoor Droplets (CLOUD) chamber (Kirkby et al., 2011). Global model simulations, either using a parameterization of neutral and charged H₂SO₄/H₂O nucleation based on the CERN CLOUD chamber

- 180 measurements, or nucleation rates calculated from the experimental thermochemical data used in MAIA (Kazil et al., 2010) show a good agreement in the global mean profile of total (> 3 nm) aerosol concentration (Määttänen et al., 2018).
- 185 Line 83: The authors could add the study by Sipila et al. (2016, Nature 537, p. 532), because it is the very first study in which iodine compound have been measured in molecular clusters associated with new particle formation in a coastal atmosphere.

We thank the referee for this suggestion, and have added the reference accordingly.

- 190 Lines 193-194: Stating that SO2 concentration measurements are sensitive to <100 pptv does not really tell anything useful to the reader. What is the actual detection limit of the instrument under the operating conditions of this study, and reliable are the SO2 of just a few pptv reported in many of the figures? The referee makes a good point. The reference to concentrations <100 pptv was made to differentiate between measurements of SO2 made on ATom by the laser induced fluorescence</p>
- 195 (LIF) instrument, which have a detection limit of 1pptv, and are thus highly relevant to this discussion of NPF in very remote regions, and another SO2 measurement made on ATom, with detection limits > 100 pptv, which are more pertinent to analyses of the more polluted regions. This was poorly explained, and we have made the following changes to the text:

200 Line 115 in the method sections has been changed from "SO₂ observations sensitive at <100 parts per trillion by volume (pptv; nmol mol-1) were made on the fourth set of flights (May 2018) using laser-induced fluorescence techniques (Rollins et al., 2017)." to "SO₂ observations with a detection limit of 1 parts per trillion by volume (pptv; nmol mol⁻¹) were 205 made on the fourth set of flights (May 2018) using laser-induced fluorescence techniques (Rollins et al., 2017)" Lines 193-194 have been changed from "Measurements of SO2 mixing ratios with sensitivity to concentrations < 100 pptv were made on 210 the fourth ATom deployment in May 2018." to "Measurements of SO₂ mixing ratios with pptv sensitivity were made on the fourth ATom deployment in May 2018." 215 Line 243: It seems that something is missing from this text: ...it is conceivable the low...The same text is repeated on lines 319-323 and lines 325-329, except that they refer to a different figure. Thank you for spotting this! We have corrected it as noted above in the discussion of removing references to "correlation" where supporting evidence was lacking.

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225 Unsolicited corrections

We would like to alert the referee to some unsolicited corrections we would like to make to the manuscript that came up in the course of addressing referee comments. These are detailed here below. We also would like to alert the referee to proposed corrections in our response to referee #2.

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Extraneous comma removed line 57

For clearer reading, Line 62 change from

- 235 "Ammonia and amines have been shown to contribute to NPF"
 - to

"Ammonia and amines have been shown to be involved in NPF"

Line 82: "lowermost stratosphere" changed to "LMS" for consistency.

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Line 123 "CH3Cl" corrected to "CH3Cl"

Line 127

"using Fuchs expression for the coagulation rate coefficient(Seinfeld and Pandis, 2006)"

245 corrected to

	"using the Fuchs expression for the coagulation rate coefficient (Seinfeld and Pandis, 2006)"
250	Line 141 changed from "in' to "by" in "This stratospheric definition is consistent with that used by Murphy et al. (2020)".
	We noted a missing parenthesis on line 195 and so added this.
255	Line 227, alteration made in order to read better from "20 of the 55 trajectories experience more humid air, indicative of UT or tropopause conditions." to "The other 20 trajectories experienced more humid air, indicative of UT or tropopause conditions."
260	The full description of Asian Summer Monsoon (ASM) has been placed on the first usage at line 253 instead of where it was mistakenly put on line 424.
	Line 309 "Because the lifetime of these particles is \sim days" changed to "Because the lifetime of these particles is on the order of days"
265	Line 341 "from" added to "through NPF resulting from SO2 oxidation"
	Line 402 "higher observed concentrations are also likely" corrected from "is also likely"
270	Line 405 "we get a total flux" change to "we determine a total flux"
	Line 435 "ASM-sources particles" corrected to "ASM-sourced particles" Line 465 "volcano emissions" changed to "volcanic emissions"
275	Line 451 "the only eruption to occur between the tropopause height: corrected to "the only eruption to reach between the tropopause height"
280	For clarity, line 495 has been rewritten from "How this would be achieved, and the potential consequences, both intended effects and side- effects are highly uncertain." to "How this would be achieved, and the potential consequences of such actions (both the intended effects and any unintended side-effects) are highly uncertain."
285	We noted in the SM line 44 a forward slash had accidentally been used instead of a period, and have corrected this.
200	Additional references have been added at line 53 and the order of references changed to "New particle formation (NPF) has been well documented in a variety of locations in the planetary boundary layer and free troposphere (Clarke et al., 1998;Clarke et al., 2013;Kulmala et

al., 2013;Williamson et al., 2019)."

Similarly, at line 284 which now reads New particles have previously been shown to ... form in the tropical UT (Clarke et al., 1998;Clarke and Kapustin, 2002;Clarke et al., 2013;Williamson et al., 2019)

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Figure 3: "level" removed from a,b y-axis and corrections made to caption text: "except for" changed to "except", "grew" changed to "grey"

Figure 4: y-axes for NH and SH changed to be the same range to make comparison easier

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Figure 8: greater than and less than symbols in legend corrected

Figure S3. We have added a more through explanation of the figure and how this was calculated from the data in the caption, and included an example mass spectrum to illustrate this. The new proposed figure and caption are as follows:



Figure S1: Aerosol acidity in the LMS. a) An example negative ion spectrum of an acidic sulphate particle. This spectrum is from a 0.39 μm diameter particle in the stratosphere at 12.2 km and 310 ppbv of O₃ on 20171009. Laboratory calibrations show that the H₂SO₄•HSO₄⁻ peak is very small or non-existent for particles composed of ammonium sulphate and the cluster ion peak increases with acidity until it is a large peak for nearly pure sulfuric acid. b) The bars for the NH (left) and SH (right), separated by season, show the average ratio of the size of the cluster peak at m/z 195 to the sum of the peaks at m/z 195 and 97. The averages are for particles when O₃ concentrations were 250 to 350 ppbv in the stratosphere. The averages are also for particles between 0.35 and 0.6 μm diameter because in the stratosphere most particles of that size originated in the stratosphere. Lab

- 315 calibrations of particles composed of (NH₄)_{0.25}H_{1.75}SO₄ had a negative ion ration m/z 195/(97+195) of 0.034, therefore we consider ratios higher than this (more acidic) to contain less than 0.25 mole fraction ammonium. Rather than analyzing possible differences in the acidity with season, here we emphasize that stratospheric particles in all seasons and both hemispheres are highly acidic. This sets limits on the possible concentration of gas phase ammonia. Calculations of uptake from the gas phase show that a continuous 1 pptv of gas phase ammonia could add 0.25 mol fraction ammonium to sulfuric acid particles in less
- 320 than a week.

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