Response to Anonymous Referee #2 on

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

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We thank the referee for their thorough and pertinent review. We have addressed the comments as detailed here below and are grateful from the improvement this has made to the manuscript.

The description of how box modelling is done needs to be detailed better. In its current form it is really difficult to understand how to simulations are done. The reader is pointed to Kupc et al., 2020 for the description of the box model setup. However, in that paper both MAIA and TOMAS models are used, so it would be easier for the reader to understand the modelling part if it was briefly summarized in this paper.

We have expanded the description of MAIA at line 166 to read as follows:

"To more quantitatively assess the effects of thermodynamics on NPF in the LMS, box modeling is performed using the Model of Aerosols and Ions in the Atmosphere (MAIA), (Lovejoy et al., 2004;Kazil and Lovejoy, 2007;Kazil et al., 2007). MAIA describes the oxidation of SO_2 to gaseous H_2SO_4, the nucleation of neutral and negative H_2SO_4-H_2O clusters, aerosol growth by sulfuric acid condensation/evaporation, and particle coagulation. The production rate of H_2SO_4 is calculated assuming that the reaction of SO_2+OH is the rate limiting step of the oxidation of SO_2 to form H_2SO_4 (Lovejoy et al., 1996). Nucleation is described with laboratory thermochemical data for H_2SO_4 and H_2O uptake and loss by small neutral and negative clusters (Curtius et al., 2001;Lovejoy and Curtius, 2001;Froyd and Lovejoy, 2003;Hanson and Lovejoy, 2006). The thermochemical data for uptake and loss of H_2SO_4 and H_2O by large sulfuric acid aerosol ($\gg 5$ sulfuric acid molecules) are based on the liquid drop model and H_2SO_4 and H_2O vapor pressures over bulk solutions. These were calculated with a computer code (provided by S. L. Clegg, personal communication, 2007) which adopts experimental data from Giauque et al. (1960) and (Clegg et al., 1994). The thermochemical data for intermediate sized particles are a smooth interpolation of the data for small and large aerosol particles. The model uses 20 linear bins in which H_2SO_4 content increases by 1 molecule per bin, and 50 geometric bins in which H_2SO_4 content increases by a factor of 1.45 per bin, covering a dry (312.15 K, 10% RH) particle diameter range of ~0.5–800 nm.

MAIA operates along trajectories with changing pressure, temperature and relative humidity (Kazil and Lovejoy, 2007) in the temperature range 180-320 K and the relative humidity range 1-101 %, which includes upper troposphere conditions. MAIA parametrizes the OH diurnal cycle as a half-sine centered around noon with a prescribed noon OH concentration, while setting the nighttime OH concentration to 0. The length of the daytime period is calculated from the day of year and location. Atmospheric ionization rates due to galactic cosmic rays are calculated as a function of latitude, altitude, and solar cycle phase by a model of energetic particle transport in the Earth's atmosphere (O'Brien, 2005). The transformation between geographic and

geomagnetic coordinates is calculated with GEOPACK (http://geo.phys.spbu.ru/~tsyganenko/modeling.html) and the International Geomagnetic Reference Field 12 coefficients (https://www.ngdc.noaa.gov/IAGA/vmod/igrf.html).

Aerosol nucleation rates calculated from the experimental thermochemical data of neutral and charged H_2SO_4/H_2O cluster formation that are used in MAIA (Kazil and Lovejoy, 2007) compare well with neutral and charged H_2SO_4/H_2O 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 H_2SO_4/H_2O nucleation based on the CERN CLOUD chamber 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).

MAIA is run along back-trajectories, initiated at the aircraft location, which were calculated using the Traj3D trajectory model (Bowman, 1993)and the National Center for Environmental Prediction (NCEP) global forecast system (GFS) meteorology (2015). NCEP provides temperature, relative humidity, and pressure along the trajectories for the MAIA runs. The initial SO_2 concentration and the H_2SO_4 condensation sink of the initial aerosol size distribution were estimated from ATom observations at similar latitudes and altitudes. The geometric mean diameter (46 nm) and geometric standard deviation (2.8) of the initial aerosol size distribution were obtained by fitting a lognormal mode to the size distribution observed at the ATom measurement locations. The noon concentration of OH in the simulations was set to 3×10^6 molec. cm^{-3}. This estimate agrees well with aircraft-measured OH concentrations during ATom (Kupc et al., 2020)."

The comparison between CEDS emission rates and observed aerosol concentrations in Figure 11 is problematic since it in now way takes into account the transport of SO2. As mentioned in the text, observations were made mostly outside of the flight corridors and there the number concentrations of ultrafine particles did not correlate with SO2 concentration. Can it be that new particle formation occurred near flight corridors and these particles were transported to the regions of aircraft observations?

Regarding Fig 11 and the comparison of measured SO2 flux with CEDS emissions, we have indeed omitted to explicitly address transport. We have noted in the manuscript the lifetime of SO_2 as around 1 month (line 394, and SM section 2) and zonal transport from flight corridors to the most distant observations from those corridors is on the order of half a day (line 386). Using the median observed windspeed of 10-30 m/s (line385) we can also see that the complete zonal mixing timescale is on the order for 1 month, approximately equal to the lifetime of SO2. Since the jet core with peak wind speeds of 40-60 m/s is typically located on the tropopause in the 30-50N region (Manney et al., 2014), and the peak in SO2 is also located in this region, the zonal mixing time could be as little as one to two weeks. We believe this justifies the presentation of zonally averaged SO_2 in Fig 11 parts b and c. We agree with the referee that this was not adequately addressed in the original manuscript and so propose to include the following explanation at line 386:

"Based on these measured windspeeds, zonal mixing is expected on timescales of around 1 month, which is approximately equal to the lifetime of SO_2. However, since the jet core with peak wind speeds of 40-60 m/s is typically located on the tropopause in the 30-50N region

(Manney et al., 2014), and the peak in SO2 is also located in this region, the zonal mixing time could be as little as one to two weeks. For this reason, we present zonally averaged SO2 concentrations in figure 11b and c."

The referee asks an interesting question about whether NPF could be occurring within flight corridors and then transported to the ATom flight paths where we observed them. It is important to note that SO2 lifetimes are estimated to be much larger than nucleation mode aerosol lifetimes (1 month (line 394) for SO2, compared with a few days (line 147)) for nucleation mode aerosols). We therefore do not see a mechanism for transporting newly formed particles from regions of higher SO2 concentrations to regions of lower SO2 concentrations, given the assumption that SO2 is driving nucleation. Where there is a lack of correlation between SO2 and nucleation mode aerosol, we believe this is more likely to be due a different chemical source for aerosol nucleation, lack of measurement precision or the different sinks and lifetimes of SO2 and aerosols.

Page 2, Lines 36-37: Solomon et al., 2011 does not discuss aerosol size distributions. Wouldn't Williamson et al., 2019 reference be more suitable reference here?

We thank the referee for catching our mistake with the Solomon et al 2011 on lines 36-37. We think the Williamson et al 2019 reference is not ideal here, because that paper concerns ATom observations in the tropics, which generally did not penetrate into the stratosphere. Instead, we have corrected this to:

"Aerosols in the lowermost stratosphere (LMS) are highly variable, even in the absence of major volcanic eruptions (Solomon et al., 2011), and models currently struggle to reproduce observed aerosol size distributions in this region of the atmosphere (Murphy et al., 2020)."

Page 2, Lines 126-130: Where is this information about condensation and coagulation rates used?

The condensation rates explained in the methods section line 126-130 are used to calculate the condensation sink for comparison with the MAIA box modeling presented in Figure 7 and discussed at lines 170 and 203-230. The coagulation rates are used in the supplementary material section S2 to estimate the lifetime of particles in the LMS (Fig S4), which is then referenced in the main manuscript at line 147 and subsequently. To clarify this in the text we have added the following sentence at line 132:

"Condensation and coagulation rates will be used in this analysis to relate our observations to theory and models, and to estimate particle lifetimes."

Page 5, Line 146: oder → order

We have corrected the spelling mistake on line 146 and thank the referee for spotting that.

Page 8, Lines 229-237: Here you discuss that the SO2 concentrations (40 pptv) were higher than the median observed values. Why was this value chosen? Was this level required to initiate nucleation? It is said that SO2 concentrations decreased to the observed concentrations. Was NPF still ongoing at these levels?

MAIA was run only at a few discrete levels SO2 because of constraints on computing time. The pertinent levels were 20 and 40 pptv, results of which are shown in Fig 7. As the referee points out, this is above the median observed values. NPF did occur at 20 pptv, but slower growth meant that, for higher condensation sinks, observable increases in number concentrations of particles larger than 2.65 nm were only seen on trajectories with higher RHw at these SO2 concentrations (Fig. 7f). At lower condensation sinks, increases in number concentrations of particles larger than 2.65 nm were clearly observable with SO2 at 20 pptv (Fig. 7h).

Section 6: The first three paragraphs explain how aerosol emissions are calculated. The motivation for this procedure is unclear to me. Is this done in order to obtain higher temporal resolution emission rates from CEDS monthly fields?

In section 6 the first paragraph explains why we are considering aircraft as sources for small particles and SO2. The second paragraph introduces the CEDS database and explains the relevant sources used therein. The third paragraph details how we calculate expected emissions of nucleation mode aerosol using the CEDS database, using the data on aircraft emissions therein, and literature values to convert from SO2 emissions to nucleation mode particle emissions, since the nucleation mode particle emissions were not, to our knowledge, included in the CEDS database. We would like to understand better the aspects of this that are unclear, or potentially duplicating CEDS fields in order to make any necessary corrections.

Page 13, Line 400: Can higher observed SO2 concentrations be due to transport from regions with higher emissions?

In the SH LMS, the higher observed SO2 concentrations relative to CEDS emissions referred to on line 400 may well be due to transport, or perhaps small, more local sources, but we do not have the data to make any firm conclusions regarding this.

Page 14, Line 430: "influence from fires may suppress NPF in the LMS, or that the additional surface area from biomass burning particles shortens the lifetime of newly formed particles". Aren't these two the same thing?

On line 430, we were attempting to reference the influence of biomass burning on two distinct processes – the nucleation of aerosols, and the subsequent growth of those particles. The referee has shown that this lacked clarity, so we have modified the sentence from the original "suggesting that the influence from fires may suppress NPF in the LMS, or that the additional surface area from biomass burning particles shortens the lifetime of newly formed particles" to "suggesting that the additional surface area from biomass burning particles may reduce nucleation mode number concentrations in the LMS through two mechanisms, suppressing the formation of particles by increasing the condensation sink, and shortening the lifetime of particles that do form by increasing the coagulation sink."

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 would also like to alert the referee to proposed corrections in our response to referee #1.

Extraneous comma removed line 57

For clearer reading, Line 62 change from
"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.

Line 123 "CH3Cl" corrected to "CH_3Cl"

Line 127
"using Fuchs expression for the coagulation rate coefficient(Seinfeld and Pandis, 2006)"
corrected to
"using the Fuchs expression for the coagulation rate coefficient (Seinfeld and Pandis, 2006)"

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.

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."

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 ~ days" changed to "Because the lifetime of these particles is on the order of days"

Line 341 "from" added to "through NPF resulting from SO_2 oxidation"

Line 402 "higher observed concentrations … are also likely" corrected from "is also likely"

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"

Line 451 "the only eruption to occur between the tropopause height: corrected to "the only eruption to reach between the tropopause height"

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."

We noted in the SM line 44 a forward slash had accidentally been used instead of a period, and have corrected this.

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)

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

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:

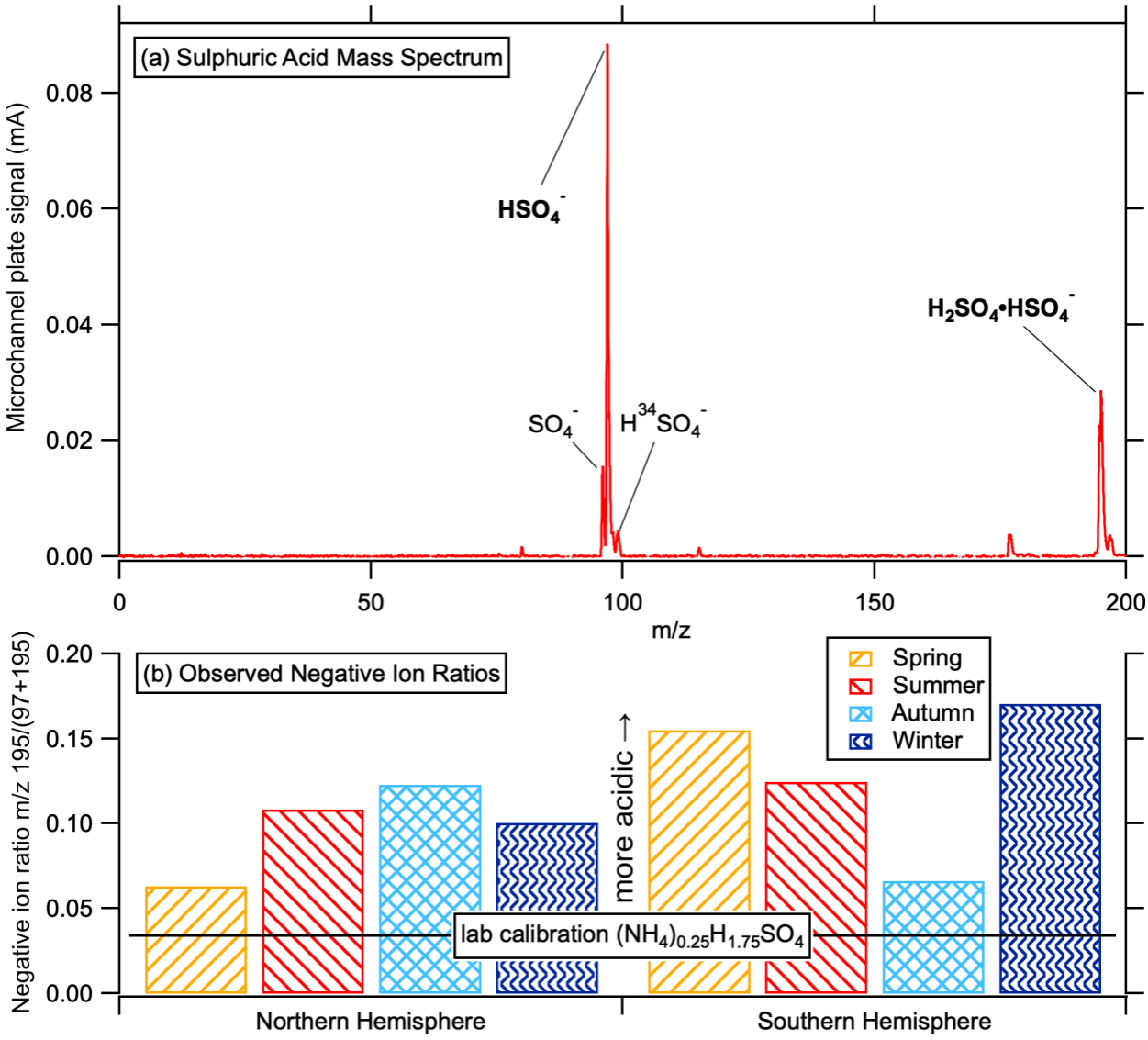

Figure S1: Aerosol acidity in the LMS. a) An example negative ion spectrum of an acidic sulphate particle. This spectrum is from a 0.39 µm diameter particle in the stratosphere at 12.2 km and 310 ppbv of O_3 on 20171009. Laboratory calibrations show that the $H_2SO_4 \bullet HSO_4^-$ peak is very small 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 O_3 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 calibrations of particles composed of $(NH_4)_{0.25}H_{1.75}SO_4$ 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 than a week.

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