Replies to review comments

We thank the reviewers and the co-editor for the time and efforts to improve the manuscript. Please find the point-to-point reply to the comments below in blue color:

Anonymous Referee #2

The manuscript is much improved from its initial version. I my opinion, only a few relatively minor issues remain to be addressed before publication.

L51: I don't think it's true that "Most of the sulfate found in ice cores can be attributed to volcanic eruptions". Marine biogenic sources contribute a significant amount of sulfate to (some) ice cores, which is usually subtracted first wen estimating sulfate of volcanic origin. This sentence should be rephrased.

L52: Anyhow, the point to be made here I think is that ice cores contain sulfate from tropical eruptions.

We rephrased the sentence to "Part of the sulfate found in ice cores can be attributed to volcanic eruptions."

L61: The logical connection implied by "Thus" is not clear. We replace the word with "And".

L133: volcanic aerosol?

Sorry for the missing word. It is fixed in the texts.

L170: Can you provide justification (and references) for these chosen values of Dx and Dz? It's surprising to me that the value of Dx is 0 in the stratosphere but non-zero in the troposphere.

Sophisticated parametrization schemes for Dx and Dz are not available for the free troposphere and stratosphere by now. Depending on the atmospheric conditions, actual values of Dx and Dz may vary by several orders of magnitude [e.g., Legras et al., 2003, 2005; Pisso et al., 2009]. There is no "correct" value of diffusivities. In our simulations, we follow the approach of Stohl et al., (2005) and use $Dx = 50 \text{ m}^2/\text{s}$, $Dz = 0 \text{ m}^2/\text{s}$ in the troposphere and $Dx = 0 \text{ m}^2/\text{s}$, $Dz = 0.1 \text{ m}^2/\text{s}$ in the stratosphere as it is implemented in the FLEXPART model. The same values of diffusivities were also used in Hoffmann et al., (2017) for trajectory calculations in the Southern Hemisphere stratosphere (September 2010 to January 2011), and Wu et al., (2017) for trajectory calculations in the Northern Hemisphere upper troposphere and lower stratosphere (June to July 2009). We also conducted sensitivity tests using several combinations of horizontal and vertical diffusivities and

compared the model results with AIRS and MIPAS measurement. In fact, the choice of Dx in the stratosphere does not have large influence on the trajectories and our conclusions are not affected either.

Please note that the MPTRAC model (and also the FLEXPART model) applies additionally a parameterization of subgrid-scale wind perturbations, which is most relevant to the air parcel dispersion of long-range simulations.

L174: Can you provide justification for the value of 7-days? This seems rather fast compared to the usual 30-day timescale of SO2 decay inferred from satellite observations.

Sensitivity tests were made to test the appropriate SO_2 life time, and it turns out that when a relatively longer life time, e.g., two to four weeks is used, the simulated SO_2 density decay is far too slow comparing with the AIRS SO_2 measurement. The IASI and OMI SO_2 measurements also show a faster decay of the SO_2 released by the Merapi eruption. We attributed this to the high water vapor and OH concentrations in tropical troposphere and lower stratosphere region.

L221: Does this statement about the correlation between temperature and eddy heat flux come from your work, or prior studies? Perhaps reference should be made to works like that of Newman et al. (2001).

Newman, P. A., Nash, E. R., and Rosenfield, J. E.: What controls the temperature of the Arctic stratosphere during the spring?, J. Geophys. Res., 106, 19999, doi:10.1029/2000JD000061, 2001.

The statement is from prior studies (Edmon et al., 1980; Newman and Nash, 2000), which were referred to in the text. We also added the reference you suggested.

L286: There seems to be transport below the jet as well, why is the one above "most significant"?

We mainly focus on the transport in the upper troposphere and lower stratosphere. The fraction under the jet is generally out of the picture after one to two months. However, at least at the initial stage the fraction dispersed under the jets is also large, so it is not accurate to say the fraction above is most significant. The sentence is rephrased as "The significant pathways are above and under the core of the subtropical jet in the Southern Hemisphere".

L325: "confirming" seems too strong, "consistent with" would fit better. Fixed.

L396: The colorbars of Fig. 9 are confusing, since the same colors are used in panel a to show only positive values but also negative numbers in panel b. better would be to use a different color for negative values in panel b.

We used different color schemes for Fig. 9a and Fig. 9b. And we reproduced the Fig. 9b in bin size of 8 days and 4° latitudes to make the figure smooth.

L454: The Summary and conclusion section repeats a lot from the discussion. These two sections could be edited dow, or merged into a single section. We shortened the discussion section by removing some description of our conclusions that are also summarized in Sect. 5 (Summary).

Anonymous Referee #3

General comments

The manuscript contains now important improvements like the removal of the QBO-discussion and a more detailed description of the aerosol cloud index (ACI). However, it is still difficult to interpret the results because several figure captions are too short or inconsistent with the text, and because ACI depends on too many parameters if you try to find missing information in the references. It would be very helpful to include more information provided in the author's response into the manuscript, but also to include a frame with H2SO4-observations from Günther et al. (2018) in Figure 9. There are still distracting sentences in the text. The discussion of uncertainties is too short. We revised the manuscript to ensure consistency between text and figure captions with a special focus on the ACI.

Our changes with respect to the ACI were:

We added to Sect. 2.1:

"The ACI-based ice and ash cloud filter methods have been successfully used in studies of volcanic sulfate aerosol, e.g. Wu et al., (2017) and Günther et al., (2018)."

L313: "While individual aerosol detections can be shown using an ACI<7 (Fig. 3), we used zonal ACI averages to compare with the simulations."

Fig. 6 and Fig. 9 captions: "The change of MIPAS ACI values (Δ ACI) are derived by subtracting the biweekly median ACI between November 2010 and March 2011 from the biweekly median ACI of the reference period between November 2007 and March 2008."

And in Sect. 4, we added comparison between the MIPAS ACI and the sulfate aerosol retrievals by Günther et al. (2018) and comparison between our results (Fig. 5 and Fig. 9) with the Fig. 5 in Günther et al. (2018), which shows our simulations also agree with the poleward dispersion of sulfate aerosol in Günther et al. (2018). Some explanation in our last response is also added as

suggested, hoping it helps readers to better understand the correlation and difference between ACI and the recent sulfate aerosol retrievals by Günther et al. (2018).

Specific comments

In the abstract and at other places in the text it is always claimed that Merapi erupted in October, however the most important explosive eruption occurred in early November as in the references. This is misleading concerning a possible impact on the ozone hole of the same year.

Page 2, line 46: This should hold also for Merapi.

It is fixed in the abstract and the texts. "Merapi eruption in October 2010" is replaced by "Merapi eruption in 2010".

Page 2, line 48: Better write: strongly enhanced heterogeneous ozone depletion at the vortex edge. 'Penetration' is also in contradiction to the figures presented.

Fixed.

Page 3: Höpfner et al (2015) on MIPAS SO_2 should be cited here or in the AIRS-section.

Reference is added.

Section 2.2 and line 177: The SO_2 index indicates that a volcanic plume is there and how it moves, but provides only poor information on the amount of injected SO_2 and its altitude distribution. Here it would help also to include other satellite data or to derive a real L2 product.

The SO_2 index used in this study can be converted into SO_2 column density, using the correlation function described in Hoffmann et al. (2014), which was obtained by radiative transfer calculations. So it can not only indicate the presence and evolution of SO_2 but also show provide quantitative information on its concentration. We added a brief explanation in the text.

Although AIRS does not provide altitude information, we combine Lagrangian modeling with the AIRS measurements to estimate the vertical distribution of the SO₂ emission and its time evolution. Future work may combine multiple-source SO₂ measurements and reverse transport modelling to obtain more realistic altitude-resolved SO₂ emissions. A preliminary study regarding this purpose has been made by Heng et al. (2016).

Line 205: Misleading, in November/December the winter hemisphere is the Northern hemisphere. Section 3.1 contains some material not relevant for the study, please shorten.

We shortened this as suggested.

Line 240ff: This 'puff' in October is quite controversial. Maybe some CALIOP observations should be provided in a supplement.

Line 239-240 summarize the description in the literature we cited (Surono et al., 2010) of the Merapi eruption from 26 to 31 October 2010; it is: "the explosive eruption first occurred between 10:00 and 12:00 UTC on 26 October and this eruption generated an ash plume that reached 12 km altitude. A period of relatively small explosive eruptions continued from 26 October to 31 October."

As shown in Fig. 9 in Surono et al. (2010), the initial SO₂ injection in October 2010 mainly occurred intermittently on 26, 29-30, and 31 October. But no altitude information is given except for the plume on 26 October at 12km. Our reconstructed SO₂ emission time series also shows some SO₂ on 26 October at ~12km but there is no matching CALIOP track found at or near the location of Mount Merapi on 26 October. Our reconstructed SO₂ time series also shows more SO₂ injection from 29 to 31 October, which generally agrees with the statement in Surono et al. (2010), but we can not find attitude information in Surono et al. (2010) for comparison.

On 29 October (see Fig. R1), a CALIOP curtain shows some dust and smoke on 1-9 km near Mount Merapi. Another matching CALIOP track on 30 October (see Fig. R2) shows dust and smoke at 1-7 km near Mount Merapi, which is similar to the SO₂ emission in Fig.2 which may indicate our results seem reasonable.

But in fact the altitude range of dust or smoke in the CALIOP curtain is doubtful because the signal under clouds is very limited, the retrieval confidence is low, and the dust signal was contaminated by dust appeared before the initial eruption of Merapi. Particularly we also have to bear in mind that CALIOP can not directly measure SO₂ (and no sulfate aerosol was detected yet in October 2010), and the injection of ash is not always accompanied by SO₂. So it has to be admitted that at least in October 2010 CALIOP did not provide perfect comparison for our case. So we decide not to add the CALIOP curtain image as a supplement.



Figure R1: (a) vertical feature mask and (b) aerosol subtype curtain on 29 October 2010 from CALIOP. The CALIOP track is overlaid on panel (a). The red triangle indicates the latitude and longitude of the Mount Merapi (elevation: 2930 m). The red boxes indicate the dust nearest to the location of the Mount Merapi, which is possibly associated with direct volcanic eruption. The blue box indicates dust contaminated by preexisting dust which is not supposed to be correlated with direct volcanic eruption.



Figure R2: Same as Fig. R1 but for 30 October 2010.

Line 261: This mass estimate is the upper end available in the literature. What is the stratospheric fraction?

The stratospheric fraction was not mentioned in the literature, unfortunately.

Line 364: Which transport barrier?

The "transport barrier" was explained in Line 353 to 365 in the marked version of the manuscript.

Section 4: Isn't it too late (December) for heterogeneous ozone depletion? Possible effect on next ozone hole season? Thank you for correcting this. We clarify it as "the ozone depletion in the

following year".

Captions of Figure 6 and Figure 9: Please include the odd definition of the difference in the text (lines 313 and 390) to avoid confusion on the sign of ACI

changes which is not following standard conventions. The definition has been added in the figure captions.

Figure 8 and page 13: Is this equivalent or geographic latitude? Please clarify. It is geographic latitude. It has been clarified in the figure caption.

References:

- Heng, Y., Hoffmann, L., Griessbach, S., Rößler, T., and Stein, O.: Inverse transport modeling of volcanic sulfur dioxide emissions using large-scale simulations, Geosci. Model Dev., 9, 1627-1645, doi: 10.5194/gmd-9-1627-2016, 2016
- Legras, B., Joseph, B., and Lefèvre, F.: Vertical diffusivity in the lower stratosphere from Lagrangian back-trajectory reconstructions of ozone profiles, Journal of Geophysical Research: Atmospheres, 108, doi:10.1029/2002JD003045, 2003.
- Legras, B., Pisso, I., Berthet, G., and Lefèvre, F.: Variability of the Lagrangian turbulent diffusion in the lower stratosphere, Atmos. Chem. Phys., 5, 1605-1622, doi: 10.5194/acp-5-1605-2005, 2005.
- Pisso, I., Real, E., Law, K. S., Legras, B., Bousserez, N., Attié, J. L., and Schlager, H.: Estimation of mixing in the troposphere from Lagrangian trace gas reconstructions during long-range pollution plume transport, Journal of Geophysical Research: Atmospheres, 114, doi:10.1029/2008JD011289, 2009.
- Wu, X., Griessbach, S., and Hoffmann, L.: Equatorward dispersion of a high-latitude volcanic plume and its relation to the Asian summer monsoon: a case study of the Sarychev eruption in 2009, Atmos. Chem. Phys., 17, 13439-13455, doi: 10.5194/acp-17-13439-2017, 2017.

Long-range transport of volcanic aerosol from the 2010 Merapi tropical eruption to Antarctica

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8 Abstract

Volcanic sulfate aerosol is an important source of sulfur for Antarctica where other local sources of 9 sulfur are rare. Mid- and high latitude volcanic eruptions can directly influence the aerosol budget of the 10 11 polar stratosphere. However, tropical eruptions can also enhance polar aerosol load following 12 long-range transport. In the present work, we analyze the volcanic plume of a tropical eruption, Mount Merapi in October 2010, and investigate the transport pathway of the volcanic aerosol from the tropical 13 tropopause layer (TTL) to the lower stratosphere over Antarctica. We use the Lagrangian particle 14 15 dispersion model Massive-Parallel Trajectory Calculations (MPTRAC) and Atmospheric Infrared Sounder (AIRS) SO₂ measurements to reconstruct the altitude-resolved SO₂ injection time series during 16 the explosive eruption period and simulate the transport of the volcanic plume using the MPTRAC 17 18 model. AIRS SO₂ and aerosol measurements, the aerosol-cloud-index values provided by Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) are used to verify and complement the 19 20 simulations. The Lagrangian transport simulation of the volcanic plume is compared with MIPAS 21 aerosol measurements and shows good agreement. Both the simulations and the observations presented in this study suggest that volcanic plumes from the Merapi eruption were transported to the south of 22 60 °S one month after the eruption and even further to Antarctica in the following months. This 23 relatively fast meridional transport of volcanic aerosol was mainly driven by quasi-horizontal mixing 24 from the TTL to the extratropical lower stratosphere and the most of the quasi-horizontal mixing 25 occurred between the isentropic surfaces of 360 to 430 K. When the plume went to Southern 26 Hemisphere high latitudes, the polar vortex was displaced from the South Pole, so that the volcanic 27 28 plume was carried to the South Pole without penetrating the polar vortex. Although only 4% of the sulfur injected by the Merapi eruption was transported into the lower stratosphere south of 60 S, the 29 Merapi eruption contributed up to 8800 tons of sulfur to the Antarctic lower stratosphere. This indicates 30

31 that the long-range transport under favorable meteorological conditions enables a moderate tropical

32 volcanic eruption to be an important remote source of sulfur for the Antarctic stratosphere.

33 **1 Introduction**

Over the past two decades, multiple volcanic eruptions injected sulfur into the upper troposphere and 34 lower stratosphere, which has been the dominant source of the stratospheric sulfate aerosol load 35 (Vernier et al., 2011), preventing the background level from other sources ever being seen (Solomon et 36 al., 2011). Stratospheric sulfate aerosol mainly reflects solar radiation and absorbs infrared radiation, 37 38 causing cooling of the troposphere and heating of the stratosphere. Stratospheric sulfate aerosol also has an impact on chemical processes in the lower stratosphere (Jäger and Wege, 1990; Solomon et al., 39 1993), in particular on polar ozone depletion (e.g. McCormick et al., 1982; Solomon et al., 1986, 1999, 40 2016; Portmann et al., 1996; Tilmes et al., 2008; Drdla and Müller, 2012). The presence of H₂SO₄ in the 41 polar stratosphere in combination with cold temperatures facilitates the formation of polar stratospheric 42 clouds (PSCs), which increase heterogeneous ozone depletion chemistry (Solomon et al., 1999; Zuev et 43 al., 2015). Recent healing of Antarctic ozone depletion was constantly disturbed by moderate volcanic 44 eruptions (Solomon et al., 2016). Mid- and high latitude explosive volcanic eruptions may directly 45 influence the polar stratosphere and may have an effect on ozone depletion in the next austral spring. 46 47 For example, the aerosol plume from the Calbuco eruption in 2015, including various volcanic gases, strongly enhanced heterogeneous ozone depletion at the vortex edgepenetrated the polar vortex and 48 caused an Antarctic ozone hole with the largest daily averaged size on record in October 2015 (Solomon 49 et al., 2016; Ivy et al., 2017; Stone et al., 2017). 50

51 Usually, Antarctica is relatively free of local aerosol sources, but aerosol from low latitudes can reach Antarctica through long-range transport (Sand et al., 2017). MostPart of the sulfate found in ice cores 52 can be attributed to tropical volcanic eruptions (Gao et al., 2007). Measurements of enhanced aerosol in 53 the lower Antarctic stratosphere right above the tropopause were made in October/November 1983, 54 1984 and 1985. These enhanced aerosol number concentrations were attributed to aerosol transported to 55 Antarctica from the eruption of the tropical volcano El Chichón in 1982 (Hofman and Rosen, 1985; 56 Hofmann et al., 1988). Model results indicated that numerous moderate eruptions affected ozone 57 distributions over Antarctica, including the Merapi tropical eruption in October 2010 (Solomon et al., 58 2016). However, due to the limit of spatial and temporal resolution of satellite data and in-situ 59 observations, it is difficult to investigate transport process as well as the influence of the location of the 60

61 eruption, the plume height and the background meteorological conditions. <u>Thus_And</u> the transport 62 mechanism is not well represented in present global climate models and the uncertainties of the 63 modeled AOD in polar regions are large (Sand et al., 2017).

Mount Merapi (7.5 S, 110.4 E, elevation: 2930 m) is an active stratovolcano located in Central Java, 64 Indonesia. Merapi has a long record of eruptive activities. The most recent large eruption with a 65 66 volcanic explosivity index of 4 occurred between 26 October and 7 November 2010 (Pallister et al., 67 2013), with SO₂ emission rates being a few orders of magnitude higher than previous eruptions. Following the Merapi eruption in 2010, evidence of poleward transport of sulfate aerosol towards the 68 Southern Hemisphere high latitudes was found in time series of aerosol measurements by the Michelson 69 Interferometer for Passive Atmospheric Sounding (MIPAS) (Günther et al., 2018) and Cloud-Aerosol 70 Lidar with Orthogonal Polarization (CALIOP) (Khaykin et al., 2017; Friberg et al., 2018). 71

There are three main ways for transport out of the tropical tropopause layer (TTL): the deep and shallow 72 branches of the Brewer-Dobson circulation (BDC) and horizontal mixing (Vogel et al., 2011). There is 73 considerable year-to-year seasonal variability in the amount of irreversible transport from the tropics to 74 75 high latitudes, which is related to the phase of the quasi-biennial oscillation (QBO) and the state of the polar vortex (Olsen et al., 2010). The BDC plays a large role in determining the distributions of many 76 constituents in the extratropical lower stratosphere. The faster quasi-horizontal transport between the 77 tropics and polar regions also significantly contributes to determining these distributions. The efficiency 78 of transporting constituents quasi-horizontally depends on wave breaking patterns and varies with the 79 time of the year (Toohey et al., 2011; Wu et al., 2017). Better knowledge of the transport pathways and 80 an accurate representation of volcanic sulfur injections into the upper troposphere and lower 81 stratosphere (UTLS) are key elements for estimating the global stratospheric aerosol budget, the cooling 82 83 effects and the ozone loss linked to volcanic activity.

84 The aim of the present study is to reveal the transport process and the influence of meteorological conditions by combining satellite observations with model simulations in a case study. We investigate 85 the quasi-horizontal transport by tracing the volcanic plume of the Merapi eruption from the tropics to 86 Antarctica and quantifying its contribution to the sulfur load in the Antarctic lower stratosphere. In Sect. 87 2, the new Atmospheric Infrared Sounder (AIRS) SO₂ measurements (Hoffmann et al., 2014), the new 88 MIPAS aerosol measurements (Höpfner et al., 2015; Griessbach et al., 2016) and the method for 89 reconstructing the SO₂ injection time series of the Merapi eruption are introduced. In Sect. 3 the results 90 91 are presented: first, the reconstructed time series of the Merapi eruption is discussed; second, the dispersion of the Merapi plume is investigated using long Lagrangian forward trajectories initialized 92

with the reconstructed SO_2 time series; third, the simulation results are compared with MIPAS aerosol measurements and the plume dispersion is investigated using MIPAS aerosol detections. In Sect.4 the results are discussed and the conclusions are given in Sect. 5.

96 2 Satellite data, model and method

97 **2.1 MIPAS aerosol measurements**

MIPAS (Fischer et al., 2008) is an infrared limb emission spectrometer aboard the European Space 98 Agency's (ESA's) Envisat, which provided nearly 10 years of measurements from July 2002 to April 99 100 2012. MIPAS spectral measurements cover the wavelength range from 4.15 to 14.6 microns. The vertical coverage of MIPAS nominal measurement mode during the optimized resolution phase from 101 January 2005 to April 2012 was 7-72 km. The field of view of MIPAS was about 3 km ×30 km 102 (vertically xhorizontally) at the tangent point. The extent of the measurement volume along the line of 103 104 sight was about 300 km, and the horizontal distance between two adjacent limb scans was about 500 km. On each day, ~14 orbits with ~90 profiles per orbit were measured. From January 2005 to April 2012, 105 the vertical sampling grid spacing between the tangent altitudes was 1.5 km in the UTLS and 3 km at 106 107 altitudes above. In 2010 and 2011, MIPAS measured for 4 days in nominal mode followed alternately 108 by one day in middle atmosphere mode or upper atmosphere mode. In this study, we focussed on 109 measurements in the nominal mode.

For the aerosol detection, we used the MIPAS altitude-resolved aerosol-cloud-index (ACI) as introduced by Griessbach et al. (2016) to compare with the model simulations and to analyze the poleward transport of the Merapi volcanic plume.

113 ACI is the maximum value of the cloud index (CI) and aerosol index (AI):

$$ACI = \max(CI; AI), \tag{1}$$

The CI is an established method to detect clouds and aerosol with MIPAS. The CI is the ratio between the mean radiances around the 792 cm⁻¹, where a CO_2 line is located and the atmospheric window region around 833 cm⁻¹ (Spang et al., 2001):

$$CI = \frac{\overline{I_1}([788.25, 796.25 \text{ cm}^{-1}])}{\overline{I_2}([832.31, 834.37 \text{ cm}^{-1}])},$$
(2)

where $\overline{I_1}$ and $\overline{I_2}$ are the mean radiances of each window. The AI is defined as the ratio between the mean radiance around the 792 cm⁻¹ CO₂ band and the atmospheric window region between 960 and 961 cm⁻¹:

AI =
$$\frac{\overline{I_1}([788.25,796.25 \text{ cm}^{-1}])}{\overline{I_3}([960.00,961.00 \text{ cm}^{-1}])},$$
 (3)

119 where $\overline{I_1}$ and $\overline{I_3}$ are the mean radiance of each window.

The ACI is a continuous unitless value. Small ACI values indicate a high cloud or aerosol particle load and large values indicate a smaller cloud or aerosol particle load. For the CI, Sembhi et al. (2012) defined a set of variable (latitude, altitude and season) thresholds to discriminate between clear and cloudy air. The most advanced set of altitude and latitude dependent thresholds allows for the detection of aerosol and clouds with infrared extinction coefficients larger than 10⁻⁵ km⁻¹. For the ACI, a comparable sensitivity is achieved when using a fixed threshold value of 7 (Griessbach et al. 2016). Variations in the background aerosol are also visible with larger ACI values.

To remove ice clouds and volcanic ash from the MIPAS aerosol measurements, we first separated the 127 data into clear air (ACI>7) and cloudy air (ACI<=7). Then we applied the ice cloud filter (Griessbach et 128 129 al., 2016) and the volcanic ash and mineral dust filter (Griessbach et al., 2014) to the cloudy part and 130 removed all ice or ash detections. This ACI-based ice and ash cloud filter have been successfully used 131 in studies of volcanic sulfate aerosol, e.g. Wu et al. (2017) and Günther et al. (2018). However, since 132 the ice and ash cloud filters are not sensitive to non-ice PSCs, the resulting aerosol retrieval results still contain non-ice PSCs. We keep the non-ice PSCs in the MIPAS retrieval results in this study to show 133 the temporal and spatial extent of the PSCs, when and where the identification of volcanic aerosol is not 134 possible. 135

136 **2.2 AIRS**

137 AIRS (Aumann et al., 2003) is an infrared nadir sounder with across-track scanning capabilities aboard the National Aeronautics and Space Administration's (NASA's) Aqua satellite. Aqua was launched in 138 2002 and operates in a nearly polar Sun-synchronous orbit at about 710 km with a period of 98 min. 139 140 AIRS provides nearly continuous measurement coverage with 14.5 orbits per day and with a swath width of 1780 km it covers the globe almost twice a day. The AIRS footprint size is 13.5 km × 13.5 km at nadir 141 and 41 km \times 21.4 km for the outermost scan angles respectively. The along-track distance between two 142 adjacent scans is 18 km. The AIRS measurements provide good horizontal resolution and make it ideal 143 for observing the fine filamentary structures of volcanic SO₂ plumes. 144

In this study, we use an optimized SO_2 index (SI, unit: K) to estimate the amount of SO_2 injected into the atmosphere by the Merapi eruption in 2010. The SI is defined as the brightness temperature differences in the 7.3 μ m SO₂ waveband.

148
$$SI = BT (1412.87 cm^{-1}) - BT (1371.52 cm^{-1}),$$
 (4)

where BT is the brightness temperature measured at wavenumber ν . This SI is more sensitive to low 149 concentrations and performs better on suppressing background interfering signals than the SI provided in 150 the AIRS operational data products. It is an improvement of the SI definition given by Hoffmann et al. 151 (2014) by means of a better choice of the background channel (selecting 1412.87 cm⁻¹ rather than 152 1407.2 cm⁻¹). The SI increases with increasing SO₂ column density and it is most sensitive to SO₂ at 153 altitudes above 3-5 km. SO₂ injections into the lower troposphere are usually not detectable in the infrared 154 spectral region because the atmosphere gets opaque due to the water vapor continuum. A detection 155 156 threshold of 1 K was used in this study to identify the Merapi SO₂ injections. The SI was converted into 157 SO₂ column density using a correlation function described in Hoffmann et al. (2014), which was obtained by radiative transfer calculations. AIRS detected the Merapi SO₂ cloud from 3 November to 15 158 November 2010. 159

2.3 MPTRAC model and reconstruction of the volcanic SO₂ injection time series of the Merapi eruption

In this study, we use the highly scalable Massive-Parallel Trajectory Calculations (MPTRAC) to 162 investigate the volcanic eruption event. In the MPTRAC model, air parcel trajectories are calculated 163 164 based on numerical integration using wind fields from global meteorological reanalyses (Hoffmann et al., 2016; Rößler et al., 2018). The MPTRAC model can be driven by reanalyses, e.g., ERA-Interim, 165 Modern-Era Retrospective Analysis for Research and Applications (MERRA) and National Centers for 166 Environmental Prediction (NCEP)/National Center for Atmospheric Research (NCAR). Hoffmann et al. 167 (2016) showed that ERA-interim data provides the best trade-off between accuracy and computing time. 168 169 So in this study, our calculations are based on ERA-interim data.

Diffusion is modeled by uncorrelated Gaussian random displacements of the air parcels with zero mean 170 and standard deviations $\sigma_x = \sqrt{D_x \Delta t}$ (horizontally) and $\sigma_z = \sqrt{D_z \Delta t}$ (vertically). D_x and D_z are the 171 horizontal and vertical diffusivities respectively, and Δt is the time step for the trajectory calculations. 172 For the Merapi simulation, D_x and D_z were set to 50 m² s⁻¹ and 0 m² s⁻¹ in the troposphere and 0 m² s⁻¹ 173 and 0.1 m²s⁻¹ in the stratosphere, respectively. In addition, sub-grid scale wind fluctuations, which are 174 particularly important for long-range simulations, are simulated by a Markov model (Stohl et al., 2005; 175 Hoffmann et al., 2016). Loss processes of chemical species, SO₂ in our case, are simulated based on an 176 exponential decay of the mass assigned to each air parcel. In the stratosphere, a constant half lifetime of 177

7 days was assumed for SO₂. Considering that the Merapi eruption occurred in the humid tropics with a
high concentration of hydroxyl radical, a half lifetime of 2.5 days was assumed for the troposphere.

To estimate the time- and altitude-resolved SO₂ injections, we follow the approach of Hoffmann et al. 180 (2016) and Wu et al. (2017) and use backward trajectories calculated with the MPTRAC model together 181 with AIRS SO₂ measurements. Measurements from 3 to 7 November 2010 were used to estimate the 182 SO₂ injection during the explosive eruption. Since the AIRS measurements do not provide altitude 183 184 information, we established a column of air parcels at each AIRS SO₂ detection. The vertical range of the column was set to 0–25 km, covering the possible vertical dispersion range of the SO₂ plume in the 185 first few days. The AIRS footprint size varies between 14 and 41 km, hence in the horizontal direction, 186 we chose an average of 30 km as the full width at half maximum for the Gaussian scatter of the air 187 parcels. In our simulations, a fixed total number of 100,000 air parcels was assigned to all air columns 188 and the number of air parcels in each column was scaled linearly proportional to the SO₂ index. Then 189 backward trajectories were calculated for all air parcels, and trajectories that were at least 2 days but no 190 191 more than 5 days long and that passed the volcano domain were recorded as emissions of Merapi. The 192 volcano domain was defined by means of a search radius of 75 km around the location of the Merapi 193 and 0-20 km in the vertical direction, covering all possible injection heights. Sensitivity experiments have been conducted to optimize these pre-assigned parameters to obtain the best simulation results. 194 Our estimates of the Merapi SO_2 injection are shown in Sect. 3. 195

Starting with the reconstructed altitude-resolved SO₂ injection time series, the transport of the Merapi plume is simulated for 6 months. The trajectory calculations are driven by the ERA–Interim data (Dee et al., 2011) interpolated on a $1 \times 1^{\circ}$ horizontal grid on 60 model levels with the vertical range extending from the surface to 0.1 hPa. The ERA–Interim data are provided at 00, 06, 12 and 18 UTC. Outputs of model simulations are given every 3 hours at 00, 03, 06, 09, 12, 15, 18 and 21 UTC. The impact of different meteorological analyses on MPTRAC simulations was assessed by Hoffmann et al. (2016, 2017). In both studies the ERA–Interim data showed good performance.

203 **3 Results**

204 **3.1 Meteorological background conditions in Antarctica**

The Merapi eruption in October 2010 occurred during the seasonal transition from austral spring to summer when the polar vortex typically weakens and the ozone hole shrinks. The poleward transport from the tropics to the polar region is known to be modulated by the phase of the QBO and the state of the polar vortex. Just before the Merapi eruption in 2010, the QBO switched from the easterly phase to
the westerly phase. The westerly phase of the QBO promotes meridional transport from the tropics to
subtropics, especially into the winter hemisphere (O'Sullivan and Dunkerton, 1997; Shuckburgh et al.,
2001; Jäger, 2005). However, it also results in zonal wind acceleration at the high latitudes (Holton and
Tan, 1980; Watson and Gray, 2014) and a less dynamically disturbed polar vortex (Baldwin and
Dunkerton, 1999; Anstey and Shepherd, 2014), which will make it less possible for air parcels to
penetrate the polar vortex.

Figure 1 depicts the meteorological conditions at the polar lower stratosphere (150hPa, ~12km) after the 215 eruption. The minimum temperature south of 50 °S (Fig. 1a) was much lower than the climatological 216 mean during mid-November to mid-December but still higher than the low temperature necessary for 217 218 the existence of PSCs. The polar mean temperature in Fig. 1b, defined as the temperature averaged over latitudes south of 60 S, stayed lower than the climatological mean from November 2010 until February 219 2011. Corresponding to the low temperatures, the average zonal wind speed at 60 °S (Fig. 1c) was 220 221 significantly larger than the climatological mean value from November 2010 to mid-January 2011. The eddy heat flux in Fig. 1d is the product of meridional wind departures and temperature departures from 222 the respective zonal mean values. A more negative value of the eddy heat flux indicates that wave 223 224 systems are propagating into the stratosphere and are warming the polar region (Edmon et al., 1980; Newman and Nash, 2000; Newman et al., 2001). There is a strong anticorrelation between temperature 225 226 and the 45-day average of the eddy heat flux lagged prior to the temperature. Compared with the 227 climatological mean state, the polar vortex was more disturbed during mid-July to end of August, but from mid-October to late November, the heat flux was much smaller than the long-term average, which 228 229 meant a reduction in dynamical disturbances. Considering the temperature, the subpolar wind speed and 230 the heat flux, the polar vortex was colder and stronger in November and early December 2010 than it was at the same time in other years (see Fig. 1e). Consistent with the large wind speed and low 231 temperature, the polar vortex was stable after the Merapi eruption until early December 2010. 232 Afterwards, it shrunk abruptly and was destructed in by mid-January 2011. In accordance with the 233 234 strength of the polar vortex, in November and early December 2010 the ozone hole area in Fig. 1f, 235 defined as the region of ozone values below 220 Dobson Units (DU) located south of 40 %, was larger than the climatological mean. Meanwhile, the low polar mean temperature and stable polar vortex 236 resulted in a long-lasting ozone hole, which disappeared in the last week of December. The polar vortex 237 238 broke down by mid-January 2011 when the subpolar wind speed decreased below 15 m/s (Fig. 1c).

3.2 Merapi eruption and SO₂ injection time series

240 According to the chronology of the Merapi eruption that combined satellite observations from AIRS, the 241 Infrared Atmospheric Sounding Interferometer (IASI), the Ozone Monitoring Instrument (OMI) and a limited number of Ground-based ultra-violet Differential Optical Absorption Spectroscopy (DOAS) 242 243 measurements (Surono et al., 2012), the explosive eruption first occurred between 10:00 and 12:00 UTC 244 on 26 October and this eruption generated an ash plume that reached 12 km altitude. A period of 245 relatively small explosive eruptions continued from 26 October to 31 October. On 3 November, the 246 eruptive intensity increased again accompanied by much stronger degassing and a series of explosions. The intermittent explosive eruptions occurred during 4–5 November with the climactic eruption on 4 247 November, producing an ash column that reached up to 17 km altitude. From 6 November, explosive 248 249 activity decreased slowly and the degassing declined.

Figure 2 shows the time- and altitude-resolved SO₂ injections of the Merapi eruption retrieved using the 250 AIRS SO2 index data and the backward-trajectory approach. It agrees well with the chronology of the 251 252 Merapi eruption as outlined by Surono et al. (2012). SO₂ was injected into altitudes below 8 km during the initial explosive eruptions on 26-30 October. Starting from 31 October the plume reached up to 12 253 km. During 1-2 November the SO₂ injections into altitudes below 12 km continued but the mass was 254 less than the mass at the initial phase. On 3 November the intensity increased again and peaked on 4 255 256 November. Before 3 November the reconstruction indicates a minor fraction of SO₂ right above the 257 tropopause. The SO_2 above the tropopause is not reported in the study Surono et al. (2012), but is quite 258 robust in our simulations. Further, CALIOP profiles from show that some dust appeared at the height from about 14 to 18 km around Mount Merapi on 2, 3, 5 November 2010, and between 3 and 17 km on 259 6 November 2010. It could be a fraction of volcanic plume elevated by the updraft in the convection 260 associated with the tropical storm Anggrek. The center of the tropical storm Anggrek was on the Indian 261 ocean about 1000 km southwest of the Mount Merapi. The SO₂ mass above the tropopause is very small 262 263 compared with the total SO_2 mass.

To study the long-range transport of the Merapi plume, we initialized 100,000 air parcels as the SO_2 injection time series shown in Fig. 2. A total SO_2 mass of 0.44 Tg is assigned to these air parcels as provided in Surono et al. (2012). Then the trajectories are calculated forward for 6 months. Here, we only considered the plume in the upper troposphere and stratosphere where the lifetime of both SO_2 and sulfate aerosol is longer than their lifetime in the lower troposphere. Further, the SO_2 was converted into

sulfate aerosol within a few weeks (von Glasow et al., 2009; also confirmed by the AIRS SO₂ and 269 MIPAS aerosol data), and we assumed that the sulfate aerosol remained collocated with the SO₂ plume. 270 Figure 3 shows the evolution of the simulated Merapi plume and compares the plume altitudes to the 271 aerosol top altitudes measured by MIPAS between 7 and 23 November. Immediately after the eruption, 272 the majority of the plume moved towards the southwest and was entrained by the circulation of the 273 274 tropical storm Anggrek. After Anggrek weakened and dissipated, the majority of the plume parcels in 275 the upper troposphere moved eastward and those in the lower stratosphere moved westward. In general, the altitudes of the simulated plume agree with the MIPAS measurements. The remaining discrepancies 276 of air parcel altitudes being higher than the altitudes of MIPAS aerosol detections can be attributed to 277 278 the fact that the MIPAS tends to underestimate aerosol top cloud altitudes, which is about 0.9 km in 279 case of low extinction aerosol layers and can reach down to 4.5 km in case of broken cloud conditions 280 (Höpfner et al., 2009).

281 **3.3 Lagrangian simulation and satellite observation of the transport of the Merapi plume**

The early plume evolution until about one month after the initial eruption is shown on the maps in Fig. 3 together with MIPAS measurements of volcanic aerosol (only aerosol detections with ACI<7 are shown). Within about one month after the initial eruption, the plume is nearly entirely transported around the globe in the tropics, moving west at altitudes of about 17 km. The lower part of the plume, below about 17 km is transported south-eastward and reaches latitudes south of 30 °S by mid-November. The simulated long-term transport of the Merapi plume is illustrated in Fig. 4, showing the proportion of air parcels reaching a latitude-altitude bin every half a month.

289 The simulation results show that during the first month after the eruption (Fig. 4a–b), the majority of the 290 plume was transported southward roughly along the isentropic surfaces. The most significant pathways are is above and under the core of the subtropical jet in the Southern Hemisphere. However, because of 291 the transport barrier of the polar jet during austral spring, the plume was confined to the north of 60 S. 292 293 In December 2010 (Fig. 4c-d), a larger fraction of the plume was transported southward above the 294 subtropical jet core and deep into the polar region south of 60 % as the polar jet broke down. Until the 295 end of January 2011, the majority of the plume entered the mid- and high latitudes in the Southern Hemisphere. Substantial quasi-horizontal poleward transport from the TTL towards the extratropical 296 lowermost stratosphere (LMS) in Antarctica was found from November 2010 to February 2011 (Fig. 297 4a-h), approximately between 350 and 480 K (~10-20 km). In March 2011(Fig. 4i-j), the proportion of 298 the plume that went across 60 S stopped increasing and the maxima of the proportion descended below 299

300 380 K. Besides this transport towards Antarctica, a slow upward transport could also be seen. The top of 301 the simulated plume was below the 480 K isentropic surface at around 18 km in Fig. 4a and then the top 302 of the plume went up to 25 km five months later in Fig. 4j. This slow upward transport was mainly 303 located in the tropics and can be attributed to the tropical upwelling.

304 MIPAS ACI values are used to study the plume dispersion and compare with the simulations. Figure 5 305 displays the time-latitude section of the median value of the MIPAS ACI within the vertical range of 12 306 and 18 km from January 2005 to April 2012, covering the TTL and the LMS. Only ACI values from 4 to 8 are shown in Fig.5. The MIPAS data show all the key events that contributed to the aerosol load in 307 the UTLS, i.e., moderate volcanic eruptions from 2005 to 2012 and one large bushfire in 2009, as well 308 as the subsequent dispersion and change of aerosol load over time. The poleward transport of aerosol 309 310 from the Merapi eruption in 2010, marked by the red triangle in Fig. 5, caused the most long-lasting 311 aerosol signals in the Southern Hemisphere mid- and high latitudes. The small ACI values in the winter 312 months of both hemispheres are attributed to the non-ice PSCs.

While individual aerosol detections can be shown using an ACI<7 (Fig. 3), we used zonal ACI averages 313 314 to compare with the simulations. To show the change of aerosol load in the Southern Hemisphere due to the Merapi eruption, we first removed the seasonal cycle from the MIPAS aerosol data. Therefore, we 315 selected a time period from November 2007 to March 2008 with no major SO₂ emission in the Southern 316 Hemisphere UTLS (as shown in Fig. 5) as a "reference state". We calculated the biweekly median ACI 317 between November 2010 and March 2011 and subtracted it from the biweekly ACI median of the 318 "reference state". The results are shown in Fig. 6. Small MIPAS ACI values represent large aerosol load 319 and large ACI values represent small aerosol load, so in Fig. 6, the positive values indicate an increase 320 321 of the aerosol load and negative values indicate a decrease of the aerosol load. Reference time periods 322 from November 2003 to March 2004 and November 2005 to March 2006 were also tested and they all 323 showed qualitatively comparable results.

In the first half of November, the zonal median (Fig. 6a) does not show a signal of the Merapi eruption, 324 because during the initial time period, the plume was confined to longitudes around the volcano (see Fig. 325 326 3), and the MIPAS tracks did not always sample the maximum concentration, so the median ACI values 327 are large (low concentration or clear air). In the second half of November, the plume was transported zonally around the globe, and hence the largest aerosol increase appeared in the upper troposphere at the 328 329 latitude of the Mount Merapi (Fig. 6b) and then moved quasi-horizontally southward into the UTLS region at ~30-40 °S (Figs. 6c-d), confirming consistent with the simulation result in Fig. 4c-d. The 330 331 increase of the aerosol load south of 60 S started to become prominent after December 2010, and the

poleward movement is most obvious above the 350 K isentropic surface (Figs. 6e–h). The observations confirm the temporal and spatial characteristics of the poleward movement of the aerosol in the simulation in Fig. 4. Later in March 2011, the enhanced aerosol load in the tropics phased out and the aerosol load maxima descended to around the 350 K isentropic surface.

However, the background aerosol level in the tropical upper troposphere and stratosphere is constantly 336 337 disturbed by tropical and extratropical volcanic eruptions. The reference time period we chose 338 (November 2007 to March 2008) is relatively free of large aerosol sources in the Southern Hemisphere, but the aerosol load in the tropical stratosphere is already elevated by previous volcanic eruptions, e.g., 339 Soufriere Hills (May 2006), Tavurvur (October 2006), Piton de la Fournaise (April 2007), Jebel at Tair 340 (September 2007). So the change of the aerosol load in Fig.6 underestimates the increase of aerosol load 341 342 in the tropical stratosphere after the Merapi eruption. The slow upward transport in the tropics shown in Fig. 4 is about 7 km in five months. It is not visible in Fig. 6, but the time series of the MIPAS 343 measurements in the tropical stratosphere reveals a similar upward transport trend (see Fig. A1 in the 344 appendix). 345

346 **3.4 Quasi-horizontal transport from the tropics to Antarctica**

The MPTRAC simulations and the MIPAS measurements show the transport in the "surf zone" that 347 348 reaches from the subtropics to high latitudes (Holton et al., 1995), where air masses are affected by both 349 fast meridional transport and the slow BDC. The reconstructed emission time series in Fig. 2 and the MIPAS aerosol measurements in Figs. 3 and 6 show that the volcanic plume was injected into the TTL. 350 Hence, the main transport pathway towards Antarctica is the quasi-horizontal mixing in the lower 351 extratropical stratosphere between 350 and 480 K (see Fig. 4 and Fig. 6). Figure 7 illustrates how the 352 volcanic plume between 350 and 480 K approached Antarctica over time. Kunz et al. (2015) derived a 353 climatology of potential vorticity (PV) streamer boundaries on isentropic surfaces between 320 and 354 500K using ERA-Interim reanalyses for the time period from 1979 to 2011. This boundary is derived 355 from the maximum product of the meridional PV gradient and zonal wind speed on isentropic surfaces, 356 357 which identifies a PV contour that best represents the dynamical discontinuity on each isentropic surface. It can be used as an isentropic transport barrier and to determine the isentropic cross-barrier 358 transport related to Rossby wave breaking (Haynes and Shuckburgh, 2000; Kunz et al., 2011a,b). In Fig. 359 7, gray dashed lines mark PV boundaries on the 350 K isentropic surface. On isentropic surfaces below 360 380 K, the PV boundaries represent the dynamical discontinuity near the core of the subtropical jet 361 stream. Isentropic transport of air masses across these boundaries indicates exchange between the 362

tropics and extratropics due to Rossby wave breaking. On isentropic surfaces above 400 K, PV boundaries represent a transport barrier in the lower stratosphere, in particular, due to the polar vortices in winter (Kunz et al., 2015). For comparison, we also show the 220 DU contour lines of ozone column density (black isolines in Fig. 7), obtained from OMI on satellite Aura, indicating the boundary and size of the ozone hole. The PV boundary on 480 K is in most cases collocated with the area of the ozone hole, showing that both quantities provide a consistent representation of the area of the polar vortex.

The Merapi volcanic plume first reached the transport barrier on the 350 K isentropic surface in mid-November and went close to the transport barrier on the 480 K isentropic surface in December. The long-lasting polar vortex prevented the volcanic plume from crossing the transport barrier at 480 K in early December. But from mid-December, the polar vortex became more disturbed and displaced from the South Pole, resulting in a shrinking ozone hole. As mentioned in Sect.3.1, the ozone hole broke down at the end of December 2010 and the polar vortex broke down by mid-January 2011.

375 The fraction of the volcanic plume that crosses the individual transport barrier or the latitude of 60 °S on 376 each isentropic surface are shown in Fig. 8. In both cases, the proportion increased from November 377 2010 to January 2011. In November and December 2010 the largest plume transport across the transport barriers occurred between the 360 and 430 K isentropic surface (Fig. 8a), with a peak at 380–390 K. In 378 379 January and February 2011 the peak was slightly elevated to 390–400 K. In November 2010, the volcanic plume did not cross the 480 K transport barrier of the polar vortex at high altitudes, especially 380 381 above about 450 K. The high-latitude fraction increased from December 2010 to February 2011 as the weakening of the polar vortex made the transport barrier more permeable. In March and April 2011, the 382 total proportion decreased and the peak descended to 370 K in March and further to 360 K in April. The 383 proportion of the volcanic plume south of 60 % (Fig. 8b) increased slightly from November to 384 385 December 2010, and then increased significantly from December 2010 to January and February 2011 at 386 all altitudes as the polar vortex displaced and broke down. Finally, the transport to the south of 60 S 387 started to decrease in March 2011. From November 2010 to February 2011 the peak was around 370-388 400 K, but in March and April 2011 the peak resided around 350–370 K.

Figure 9a summarizes the simulated poleward transport of the volcanic plume between the isentropic surfaces of 350 and 480 K from November 2010 to March 2011 and the percentage of air parcels south of 60 °S. The percentage was calculated by dividing the number of SO_2 parcels between 350 and 480 K south of 60 °S by the total number of SO_2 parcels released for the forward trajectory simulation. Figure 9b demonstrates the change of aerosol load in the same vertical range for MIPAS aerosol detections. As in Sect. 3.3, we calculated the median ACI between November 2010 and March 2011 and subtracted it

395 from the ACI median of the "reference state" between November 2007 and March 2008. Positive values indicate an increase of the aerosol load and negative values indicate a decrease of the aerosol load. The 396 simulations show that the plume reached 60 °S in December 2010. Correspondingly, the aerosol load 397 south of 60 S was elevated. The percentage fluctuated, but increased until the end of February 2011, 398 with a maximum percentage of about 4%. A steep increase occurred from mid-December 2010 to end of 399 400 January 2011, following the displacement and breakdown of the polar vortex. The elevated aerosol load 401 south of 60 S decreased from March 2011, and part of the plume descended to altitudes below 350 K as shown by Fig. 8b. The MIPAS aerosol measurements in Fig. 9b where a positive ACI difference 402 indicates an increase in the aerosol load, confirm the simulated transport pattern in Fig. 9a. Overall, 403 simulations and observations indicate the largest increase of the aerosol load in the tropics and 404 405 mid-latitudes, but also show a significant enhancement over the south polar region after December 406 2010.

407 4 Discussion

The results presented in Sect. 3 show that the main transport pathway for the poleward transport of the 408 409 Merapi volcanic plume to Antarctica was between the isentropic surfaces of 350 and 480 K (about 10 to 410 20 km), covering the TTL and the lower stratosphere at mid- and high latitudes. For this long-range 411 transport on timescales of a few months, fast isentropic transport associated with quasi-horizontal mixing played the main role in transporting the volcanic aerosol from the TTL to the Antarctic lower 412 413 stratosphere. The Merapi eruption occurred in austral spring and fast poleward transport was facilitated by the weakening of the subtropical jet and active Rossby wave breaking events. The polar vortex was 414 415 relatively stable when the Merapi erupted, but by the time the volcanic plume reached the south polar region, the polar vortex was displaced from the South Pole and distorted because more wave systems 416 propagated into the polar stratosphere and warmed the polar region, so that the volcanic plume was 417 418 transported from the tropics deep into Antarctica.

Our findings using the MPTRAC model and MIPAS ACI are supported by the sulfate aerosol volume
density retrievals by G ünther et al. (2018). The aerosol enhancement due to volcanic eruptions between
12 and 18 km revealed in Fig. 5 agrees with the sulfate aerosol enhancement shown in the Fig. 5 in
G ünther et al. (2018). The Fig. 5 in G ünther et al. (2018) shows that after the eruption of Merapi 2010,
clear poleward dispersion of sulfate aerosol can be seen between 12 and 18 km, and the most prominent
signal appears on 14 and 16 km. The sulfate aerosol mole fraction south of 60 % increased also from

- the end of December 2010 as shown in our Fig. 9, although the specific date is not clear because of the
 seasonal cycle in the data. The enhanced sulfate aerosol lasted until middle of March 2011 on 12 and 14
 km, and until early April on 16 km, which also generally agrees with our results.
- There are some differences in the MIPAS ACI data used in this study and the sulfate aerosol retrievals 428 in Günther et al. (2018), which may result in some minor discrepancies regarding the spatial distribution 429 430 of aerosols. The MIPAS ACI data in this study are given on 1.5 km vertical sampling grid in the UTLS 431 region, but the sulfate aerosol retrieval in Günther et al. (2018) had a vertical resolution of 3-4 km. And comparing with the MIPAS ACI, there are further steps in Günther et al. (2018) to deal with ice cloud 432 and filter out PSCs. Due to the vertical resolution of 3-4 km, once ice cloud is detected, Günther et al. 433 (2018) removed all retrieved data up to 4 km above the ice cloud. And they filtered out PSCs based on 434 latitude, time, and temperature criteria, i.e., if the temperature between 17–23 km dropped below the 435 NAT existence temperature (195 K) poleward of 40° in NH winter (15 November to 15 April) and SH 436 winter (1 April to 30 November) the retrieval results were discarded. Filtering out PSCs based on time, 437 latitude, and temperature criteria may lead to a loss of clear air profiles, e.g., when the synoptic temperature is 438 439 sufficiently low but no PSCs are present. Usually the PSCs signals are much stronger than the aerosol signals, so we keep the non-ice PSC data to give an impression on the time and region, when and where the 440 identification of volcanic aerosol is not possible. 441
- Based on the simulation results in Sect. 3.4, up to 4% of the volcanic plume air parcels was transported 442 from the TTL to the lower stratosphere in the south polar region till the end of February 2011. We 443 assigned a total mass of 0.44 Tg to all SO₂ parcel released, which means the Merapi eruption 444 contributed about 8800 tons of sulfur to the polar lower stratosphere within 4 months after the eruption, 445 assuming that the sulfate aerosol converted from the SO2 remained in the plume. Although the 446 447 MPTRAC model we used to simulate the three-dimensional movement of air parcels in the volcanic 448 plumes can estimate the conversion of SO₂ to sulfate aerosol during the transport process, however, it does not resolve the chemical processes of aerosol formation. Hence, the estimated 8800 tons of sulfur 449 450 are the maximum value since processes as e.g. wet deposition remove sulfur from the atmosphere. But 451 in the lower stratosphere, the atmosphere is relatively dry and clean compared with the lower troposphere, so the sulfate aerosol has a lower possibility to interact with clouds or to be washed out. In fact, in the 452 polar lower stratosphere usually sedimentation and downward transport by the BDC are the main removal 453 454 processes. Clouds and washout processes usually cannot be expected in the lower stratosphere. However, 455 the amount of sulfate aerosol in the plume could also be affected by other mechanisms that speed up the 456 loss of sulfur, for example, coagulation in the volcanic plume, or the absorption of sulfur onto fine ash

457 particles. But for the moderate eruption Merapi in 2010, sulfuric particle growth may not be as 458 significant as it is in a large volcanic eruption, so the scavenging efficiency of sulfur will be low. So 459 generally, our estimation may be larger than the actual value, but this number may be considered as the 460 upper limit of the contribution of the Merapi eruption.

Besides, a kinematic trajectory model like MPTRAC, in which reanalysis vertical wind is used as vertical velocity, typically shows higher vertical dispersion in the equatorial lower stratosphere compared with a diabatic trajectory model (Schoeberl et al., 2003; Wohltmann and Rex, 2008; Liu et al., 2010; Ploeger et al., 2010, 2011). However, the ERA–Interim reanalysis data used in this study to drive the model may constrain the vertical dispersion much better than older reanalyses (Liu et al., 2010; Hoffmann et al., 2017). The meridional transport in this study was mainly quasi-horizontal transport in the mid- and high latitude UTLS region, so the effect of the vertical speed scheme is limited.

The aerosol transported to the polar lower stratosphere will finally descend with the downward flow and 468 have a chance to become a nonlocal source of sulfur for Antarctica by dry and wet deposition, following 469 470 the general precipitation patterns. Quantifying the sulfur deposition flux onto Antarctica is beyond the 471 scope of this study, though. Model results of Solomon et al. (2016) suggest that the Merapi eruption made a small but significant contribution to the ozone depletion in the following year over Antarctica in 472 the vertical range of 100-200 hPa, roughly between 10 and 14 km. This altitude range is in agreement 473 with our results, where we found transport into the Antarctic stratosphere between 10 and 20 km. When 474 475 the volcanic plume was transported to Antarctica in December 2010, the polar synoptic temperature at these low height levels was already too high for the formation of PSCs. The additional ozone depletion 476 found by Solomon et al. (2016) together with the fact that sulfate aerosol was transported from the 477 Merapi into the Antarctic stratosphere between November and February where no PSCs are present 478 479 during polar summer, may support the study that suggested that significant ozone depletion can also occur on cold binary aerosol (Drdla and Müller, 2012). The Merapi eruption in 2010 could be an 480 interesting case study for more sophisticated geophysical models to study the aftermath of volcanic 481 482 eruptions on polar processes.

483 **5 Summary**

In this study, we analyzed the poleward transport of volcanic aerosol released by the Merapi eruption in 2010 from the tropics to the Antarctic lower stratosphere. The analysis was based on AIRS SO₂ measurements, MIPAS sulfate aerosol detections and MPTRAC transport simulations. First, we estimated altitude-resolved SO₂ injection time series during the explosive eruption period using AIRS data together with a backward trajectory approach. Second, the long-range transport of the volcanic plume from the initial eruption to April 2011 was simulated based on the derived SO₂ injection time series. Then the evolution and the poleward migration of the volcanic plume were analyzed using the forward trajectory simulations and MIPAS aerosol measurements. The simulations are compared with and verified by the MIPAS aerosol measurements.

493 Results of this study suggest that the volcanic plume from the Merapi eruption was transported from the tropics to the south of 60 °S within a time scale of one month. Later on, in the UTLS region a fraction of 494 the volcanic plume (\sim 4%) crossed 60 °S, even further to Antarctica until the end of February 2011. As a 495 result, the aerosol load in the Antarctic lower stratosphere was significantly elevated. This relatively fast 496 497 meridional transport of volcanic aerosol was mainly carried out by quasi-horizontal mixing from the TTL to the extratropical lower stratosphere. Based on the simulations, most of the quasi-horizontal 498 mixing occurred between the isentropic surfaces of 360 to 430 K. This transport was in turn facilitated 499 500 by the weakening of the subtropical jet and the breakdown of the polar vortex in the seasonal transition 501 from austral spring to summer. The polar vortex in late austral spring 2010 was relatively strong compared to the climatological mean state. However, in December 2010 the polar vortex was displaced 502 503 off the South Pole and later on broke down when the plume went to the high latitudes, so the volcanic plume did not penetrate the polar vortex but entered the South Pole with the breakdown of the polar 504 505 vortex.

Overall, after the Merapi eruption, the largest increase of aerosol load occurred in the Southern Hemisphere midlatitudes and a relatively small but significant fraction of the volcanic plume (4%) was further transported to the Antarctic lower stratosphere within 4 months after the eruption. As a maximum estimation, it contributed up to 8800 tons of sulfur to the Antarctic stratosphere, which indicates that long-range transport under favorable meteorological conditions can make moderate tropical volcanic eruptions an important remote source of sulfur to Antarctica.

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513 *Code and data availability.* AIRS data are distributed by the NASA Goddard Earth Sciences Data 514 Information and Services Center. The SO₂ index data used in this study (Hoffmann et al., 2014) are 515 available for download at <u>https://datapub.fz-juelich.de/slcs/airs/volcanoes/</u> (last access: 19 October 516 2018). Envisat MIPAS Level-1B data are distributed by the European Space Agency. The ERA–Interim 517 reanalysis data (Dee et al., 2011) were obtained from the European Centre for Medium-Range Weather 518 Forecasts. The code of the Massive-Parallel Trajectory Calculations (MPTRAC) model is available under the terms and conditions of the GNU General Public License, Version 3 from the repository at
 https://github.com/slcs-jsc/mptrac (last access: 19 October 2018).

521

522 Appendix

Figure A1 shows the 9-day running median values of the MIPAS ACI between 10 N and 10 S. During the time period of the reference state (November 2007 to March 2008), the aerosol load in the tropical stratosphere from 20 to 25 km is elevated by a couple of previous volcanic eruptions. The aerosol load at this vertical range after the Merapi eruption in 2010 is apparently smaller compared with the reference state.

It should be noted that there are semiannual data oscillations in the MIPAS ACI aerosol detections. This 528 529 periodic pattern is caused by the aerosol index that uses the atmospheric window region between 960 and 961 cm⁻¹. Around this window region, there are CO₂ laser bands. Due to the semiannual 530 temperature changes at about 50 km (semiannual oscillation), the CO₂ radiance contribution to this 531 window region also oscillates. As this window is generally very clear of other trace gases, this 532 533 oscillation is not only visible at higher altitudes but also in the lower stratosphere, because the satellite line of sight looks through the whole layer (Wu et al., 2017). But even though with the semiannual data 534 oscillations, the upward transport of the aerosol from the Merapi eruption in the tropical stratosphere is 535 still visible and the vertical speed is estimated to about 7-8 km in five months (November 2010 to 536 March 2011). 537

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539 *Competing interests.* The authors declare that they have no conflict of interest.

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Figure 1: (a) Minimum temperature south of 50 °S at 150 hPa; (b) temperature averaged over the polar cap for latitudes south of 60 °S at 150 hPa; (c) zonal wind speed at 60 °S at 150 hPa; (d) eddy heat flux averaged between 45 °S and 75 °S for the 45-day period prior to the date indicated at 150 hPa; (e) the area of the polar vortex for 1 July–31 December 2010 on the 460 K isentropic surface; (f) ozone hole area for 1 July 2010–31 December 2010. Temperatures for PSC existence in (a) are determined by assuming a nitric acid concentration of 6 ppbv and a water vapor concentration of 4.5 ppmv. (a)–(f) are based on Modern-Era Retrospective analysis for Research and Applications reanalysis version 2 (MERRA2) data (Bosilovich et al., 2015). The ozone hole area in (f) is determined from OMI ozone satellite measurements (Levelt et al., 2006). The red triangles indicate the time of the Merapi eruption.



Figure 2: Merapi SO₂ emission time series (unit: kg m⁻¹ s⁻¹) derived from AIRS measurements using a backward trajectory approach (see text for details). The emission data are binned every 1 h and 0.2 km. The ticks for the horizontal axis mark 0 UTC on each day. Black dots denote the height of the thermal tropopause (based on the ERA–Interim reanalysis).



Figure 3: Distribution of the volcanic plume (showing only air parcels higher than 10 km, shading) from MPTRAC simulations (shown for 00:00UTC on selected days) and MIPAS aerosol detections (ACI < 7) within ± 6 h (color-filled circles). The altitudes of all

air parcels, regardless of their SO_2 values, are shown. The red triangle denotes the location of Mount Merapi.



Figure 4: Percentage (%) of air parcels in proportion to the total number of air parcels released in the Lagrangian forward simulation, overlaid with monthly mean zonal winds (black contours), the thermal tropopause (black dashed line), the 380 K potential temperature isoline (thick gray dashed line) and 350 and 480 K potential temperature isolines (thin gray dashed lines). Results are binned every 2° in latitude and 1 km in altitude. The red triangle denotes the latitude of the Mount Merapi. Please see title of each figure for the time period covered.



Figure 5: Median of MIPAS ACI values between 12–18 km (bin size 10 days and 2° in latitude) from January 2005 to April 2012. Only ACI values from 4 to 8 are shown. The red triangle indicates the eruption of Mount Merapi (No. 13). The black filled circles indicate 1 Manam, 2 Soufriere Hills, 3 Tavurvur (Rabaul), 4 Piton de la Fournaise, 5 Jebel at Tair, 6 Chait én, 7 Kasatochi, 8 Dalaffilla, 9 Australian bushfire, 10 Sarychev Peak, 11 Nyamuragira, 12 Pacaya, 14 Puyehue-Cord ón Caulle, 15 Nabro.



Figure 6: The change of MIPAS ACI values (Δ ACI) after the eruption of Merapi in 2010, overlaid with monthly mean zonal winds (black contours), the thermal tropopause (black dashed line), the 380 K potential temperature isoline (thick gray dashed line) and 350 and 480 K potential temperature isolines (thin gray dashed lines). Δ ACI are derived by subtracting the biweekly median ACI between November 2010 and March 2011 from the biweekly median ACI between November 2007 and March 2008. Positive values indicate an increase of the aerosol load and negative values indicate a decrease of the aerosol load. Please see the texts for details. Results are binned every 2° in latitude and 1 km in altitude. The red triangle denotes the latitude of the Mount Merapi. Please see title of each figure for the time period covered.



Figure 7: Percentage (%) of air parcels between the isentropic surfaces of 350 and 480 K in proportion to the total number of air parcels released in the Lagrangian forward simulation. All results are at 12:00 UTC on selected dates and binned every 2° in longitude and 1° in latitude. The black contours indicate the 220 DU contour lines of daily mean ozone column density provided by OMI. PV contours marked with gray dashed and solid lines show PV boundaries on the 350 and 480 K isentropic surfaces respectively (Kunz et al., 2015). The red triangle denotes the location of Mount Merapi.



Figure 8: (a) Proportion (%) of the air parcels poleward of the PV-based transport boundaries at the end of each month; (b) proportion (%) of the air parcels south of the geographic latitude of 60 S.



Figure 9: Time and latitude evolution of the (a) percentage (%) of air parcels between the isentropic surfaces of 350 and 480 K from the Lagrangian simulations (shading, only percentages larger than 0.05% are shown; bin size: 12h and 1° in latitude) and the proportion (%) of air parcels south of 60 S (black dots); (b) the change of MIPAS ACI values (Δ ACI) after the eruption of Merapi in 2010 between 350 and 480 K. Δ ACI are derived by subtracting the biweekly median ACI between November 2010 and March 2011 from the biweekly median ACI of the reference period between November 2007 and March 2008. Positive values indicate an increase and negative values indicate a decrease of the aerosol load (bin size: <u>8 days and 4° 24h and 2°</u> in latitude). Please see the texts for details. The red triangle denotes the time and latitude of the Merapi eruption.



Figure A1: MIPAS 9-day running median of ACI between 10 °N and 10 °S from May 2006 to December 2011. The dashed vertical lines indicate the reference state from November 2007 to March 2008, and the investigation period for the Merapi eruption from November 2010 to March 2011. The red triangle indicates the time of the Merapi eruption (No. 9). The black dots indicate 1 Soufriere Hills, 2 Tavurvur (Rabaul), 3 Piton de la Fournaise, 4 Jebel at Tair, 5 Kasatochi, 6 Dalaffilla, 7 Sarychev, 8 Pacaya, 10 Nabro.