Atmospheric sulfur from the upper troposphere to the upper stratosphere: 10 years of MIPAS observations

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SO2 and OCS in the stratosphere: the aerosol layer

Assessment of Stratospheric Aerosol Properties, SPARC, 2006

The Persistently Variable "Background" Stratospheric Aerosol Layer and Global Climate Change
S. Solomon et al.
Science 333, 866 (2011);
DOI: 10.1126/science.1206027
Sulfur in the troposphere: OCS and the carbon cycle

- Limited understanding of CO$_2$ sinks and sources on land due to difficult separation between the CO$_2$ taken up by photosynthesis and that released by respiration
- OCS as proxy for photosynthetic CO$_2$ uptake
- OCS and CO$_2$ have the same diffusion pathway into leaves
- Hydration reaction is irreversible for OCS: OCS + H$_2$O -> H$_2$S + CO$_2$

115.78 pm

O=C=S

156.01 pm

O=C=O

116.3 pm

CO$_2$  OCS
The Michelson Interferometer for Passive Atmospheric Sounding on Envisat
MIPAS products retrieved at KIT

SO$_2$

- Retrieval from mean spectra: 13-45 km, monthly+zonal averages → Höpfner et al., ACP, 2013
- Retrieval from single limb-scans: 8-20 km, high temporal and horizontal resolution → Höpfner et al., ACPD, 2015

OCS

- Retrieval from single limb-scans: 8-35 km, high temporal and horizontal resolution → Glatthor et al., subm. 2015
SO$_2$ from mean MIPAS spectra
**Pre-MIPAS observations of SO₂ in the stratosphere**

**ATMOS: Rinsland et al., 1995**

- Observations of SO₂ in the upper troposphere and lower stratosphere and of H₂SO₄ and SO₂ in the middle and upper stratosphere would be extremely valuable to improve our modeling and predictive capabilities of stratospheric aerosol. Currently, there is a general scarcity of measurements of key sulfur-bearing gases during their transport from the upper troposphere into the upper stratosphere.
First global measurements of SO$_2$ throughout the stratosphere

Transport to high latitudes and downwelling inside the polar vortex

Evaporation of H$_2$SO$_4$ aerosols and production of SO$_2$ from H$_2$SO$_4$ photolysis

Production of SO$_2$ from OCS and conversion into aerosols

Uplift of SO$_2$ and OCS

CN-explosion: production of H$_2$SO$_4$ aerosols on availability of OH in polar spring

Höpfner et al., 2013
First global measurements of SO$_2$ throughout the stratosphere

Höpfner et al., 2013

Production of SO$_2$ from OCS and conversion into aerosols
The background aerosol layer: production from OCS at 25-30 km

Model SO2: Brühl et al., ACP, 2012

MIPAS observations
Evaporation of H$_2$SO$_4$ aerosols and production of SO$_2$ from H$_2$SO$_4$ photolysis

Upper stratospheric values can only be simulated by including near-IR photolysis of H$_2$SO$_4$ and an additional sulfur-sink (meteoritic dust?)

Brühl et al., JGR, 2015
Transport to high latitudes and downwelling inside the polar vortex

Unusual Behavior in the Condensation Nuclei Concentration at 30 km

J. M. Rosen and D. J. Hofmann
JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 88, NO. C6, PAGES 3725-3731, APRIL 20, 1983

CN-explosion: production of H$_2$SO$_4$ aerosols on availability of OH after sunrise in polar spring
SO$_2$ from single MIPAS limb-scans
Global measurements of vertically resolved volcanic plumes → Injection mass of SO₂ for climate-chemistry models
<table>
<thead>
<tr>
<th>Name</th>
<th>Eruption date</th>
<th>Location</th>
<th>(M(t_0)) [Gg] if present: (\tau) [d]</th>
<th>(M(t_0)) [Gg] from other sources</th>
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<tbody>
<tr>
<td>Ny</td>
<td>25 Jul 2002</td>
<td>-1.4/-29.2</td>
<td>22(1)/12(1)/3(0)/37(2) \textsuperscript{a}</td>
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<td>Ru</td>
<td>25 Sep 2002</td>
<td>2.3/-125.4</td>
<td>36(19)/39(9)/15(2)/90(21) \textsuperscript{b}</td>
<td>74 \textsuperscript{1}</td>
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<td>Rev</td>
<td>3 Nov 2002</td>
<td>-0.1/-77.7</td>
<td>54(47)/29(6)/12(2)/94(47) \textsuperscript{b}</td>
<td>65–84 \textsuperscript{1}; 100 \textsuperscript{2}</td>
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<td>So</td>
<td>12 Jul 2003</td>
<td>16.7/-62.2</td>
<td>68(19)/28(7)/2(1)/98(20) \textsuperscript{b}</td>
<td>100–128\textsuperscript{3}; 140\textsuperscript{4}</td>
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<tr>
<td>Manam</td>
<td>27 Jan 2005</td>
<td>-4.1/-145.0</td>
<td>79(15)/87(9)/39(3)/206(17) \textsuperscript{b}</td>
<td>180\textsuperscript{1}; 99 ± 13(&gt; 68.1 hPa)\textsuperscript{4}</td>
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<tr>
<td>An</td>
<td>6 Apr 2005</td>
<td>16.4/-145.7</td>
<td>34(11)/34(7)/0(0)/68(13) \textsuperscript{a}</td>
<td>165 \textsuperscript{1}</td>
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<tr>
<td>Tr</td>
<td>mid-Jul 2005</td>
<td>0.0/0.0</td>
<td>38(17)/21(5)/1(1)/60(18) \textsuperscript{a}</td>
<td></td>
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<tr>
<td>Tr</td>
<td>mid-Aug 2005</td>
<td>0.0/0.0</td>
<td>61(26)/23(5)/3(3)/88(27) \textsuperscript{a}</td>
<td></td>
</tr>
<tr>
<td>Manam</td>
<td>27 Feb 2006</td>
<td>-4.1/-145.0</td>
<td>21(4)/56(8)/1(0)/80(9) \textsuperscript{a}</td>
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<td>So</td>
<td>20 May 2006</td>
<td>16.7/-62.2</td>
<td>40(29)/38(4)/85(15)/162(33) \textsuperscript{a}</td>
<td>200\textsuperscript{1}; 123–233\textsuperscript{5}; 139 ± 24(&gt; 68.1 hPa)\textsuperscript{4}</td>
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<tr>
<td>Ra</td>
<td>7 Oct 2006</td>
<td>-4.3/-152.2</td>
<td>75(26)/118/34(12)/4(205/43) \textsuperscript{b}</td>
<td>125\textsuperscript{1}; 220\textsuperscript{2}; 190 ± 14(&gt; 100 hPa)\textsuperscript{4}</td>
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<tr>
<td>Ny</td>
<td>27 Nov 2006</td>
<td>-1.4/-29.2</td>
<td>49(6)/5(0)/54(6) \textsuperscript{a}</td>
<td>58–216\textsuperscript{1}</td>
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<td>Fo</td>
<td>4 Apr 2007</td>
<td>-21.2/-55.7</td>
<td>57(10)/12(1)/2(1)/71(10) \textsuperscript{a}</td>
<td>140(&gt; 7.5 km)\textsuperscript{6}</td>
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<td>Ta</td>
<td>30 Sep 2007</td>
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<td>26(11)/27(5)/3(1)/56(12) \textsuperscript{b}</td>
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<td>Ch</td>
<td>2 May 2008</td>
<td>-42.8/-72.7</td>
<td>26(7)/2(0)/2(0)/30(7) \textsuperscript{a}</td>
<td>10\textsuperscript{6}; 6\textsuperscript{9}</td>
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<td>Ok</td>
<td>12 Jul 2008</td>
<td>53.4/-168.1</td>
<td>110(41)/31(6)/2(0)/143(41) \textsuperscript{b}</td>
<td>200–300\textsuperscript{3}; 100–200\textsuperscript{10}</td>
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<td>Ka</td>
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<td>52.2/-175.5</td>
<td>645(127)/210(86)/43(8)/899(154) \textsuperscript{c}</td>
<td>900–2700\textsuperscript{11}; 2200\textsuperscript{12}; 1000(&gt; 10 km)\textsuperscript{13}</td>
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<td>Da</td>
<td>3 Nov 2008</td>
<td>13.8/-40.5</td>
<td>31(9)/47(1)/1(0)/79(13) \textsuperscript{b}</td>
<td>1200\textsuperscript{3}; 1700\textsuperscript{6}; 1600\textsuperscript{14}; 1350 ± 38(&gt; 215 hPa)\textsuperscript{4}</td>
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<tr>
<td>Re</td>
<td>23 Mar 2009</td>
<td>60.5/-152.7</td>
<td>182(10)/18(7)/20(0)/20(12) \textsuperscript{a}</td>
<td>100–200\textsuperscript{15}</td>
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<td>Fe</td>
<td>10 Apr 2009</td>
<td>-0.4/-91.6</td>
<td>14(2)/11(3)/2(0)/27(4) \textsuperscript{a}</td>
<td>225–325\textsuperscript{6}</td>
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<td>Sa</td>
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<td>48.1/-153.2</td>
<td>888(293)/542(60)/44(4)/1473(299) \textsuperscript{c}</td>
<td>1200\textsuperscript{17}; 900\textsuperscript{14}; 571 ± 42(&gt; 147 hPa)\textsuperscript{4}</td>
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<tr>
<td>Ny</td>
<td>2 Jan 2010</td>
<td>-1.4/-29.2</td>
<td>17(5)/3(1)/2(0)/22(6) \textsuperscript{b}</td>
<td>50\textsuperscript{18}</td>
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<td>So</td>
<td>11 Feb 2010</td>
<td>16.7/-62.2</td>
<td>11(3)/12(2)/5(1)/28(4) \textsuperscript{b}</td>
<td>20\textsuperscript{19}</td>
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<td>Pa</td>
<td>28 May 2010</td>
<td>14.4/-90.6</td>
<td>-10(2)/4(1)/14(2) \textsuperscript{b}</td>
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<td>Me</td>
<td>4 Nov 2010</td>
<td>-7.5/-110.4</td>
<td>-253(61)/23(7)/276(61) \textsuperscript{c}</td>
<td>440\textsuperscript{20}</td>
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<td>Sh</td>
<td>12 Dec 2010</td>
<td>56.7/-161.4</td>
<td>18(4)/10(0)/0(0)/20(4) \textsuperscript{c}</td>
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<td>Kar</td>
<td>1 Jan 2011</td>
<td>54.0/-159.4</td>
<td>-1/1(0)/1(0) \textsuperscript{b}</td>
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<td>Gr</td>
<td>21 May 2011</td>
<td>64.4/-17.3</td>
<td>273(101)/2(0)/--276(101) \textsuperscript{a}</td>
<td>350–400\textsuperscript{1}; 108 ± 11(&gt; 215 hPa)\textsuperscript{4}</td>
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<tr>
<td>Pu</td>
<td>4 Jun 2011</td>
<td>-40.6/-72.1</td>
<td>185(33)/-185(33) \textsuperscript{c}</td>
<td>250\textsuperscript{14}</td>
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<tr>
<td>No</td>
<td>12 Jun 2011</td>
<td>13.4/41.7</td>
<td>131(86)/343(79)/65(5)/539(117) \textsuperscript{c}</td>
<td>1500\textsuperscript{14}; 650(&gt; 10 km)\textsuperscript{21}</td>
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<td>543 ± 45(&gt; 147 hPa)\textsuperscript{4}</td>
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<td></td>
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<td>10 ± 2(&gt; 147 hPa)\textsuperscript{4}</td>
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</tbody>
</table>
SO$_2$-lifetime: differences between nadir and limb

Nadir sounding instruments seem to strongly underestimate the lifetime of SO$_2$ in the UTLS:

- Detection-limit of nadir sounders (global dilution of SO$_2$, Haywood et al., 2010)?
- Combination of lower SO$_2$-lifetime at lower altitudes and nadir averaging kernels?
Non-volcanic aerosol background

Strong annual cycle of SO2 in the upper troposphere? To be confirmed by in-situ observations
OCS from single MIPAS limb-scans

Glatthor et al., subm. 2015
First global distributions of OCS: tropical sink

MIPAS measurements

Model: Berry et al., 2013

Glatthor et al., subm. 2015
OCS-hole annual cycle: model comparison

Signal of biomass uptake over S-America: observations can only be reproduced by global models, when a much larger vegetation uptake and a corresponding increase in oceanic emissions than in earlier estimates is considered.

Glatthor et al., subm. 2015
Summary

MIPAS SO₂

- Stratospheric maximum @26-30 km
- Downwelling during polar winter and springtime depletion of SO₂ as explanation for the polar aerosol bursts
- Visible and near-IR photolysis of H₂SO₄ and irreversible sink of sulfur
- Height-resolved SO₂ masses and lifetimes for ~30 volcanic eruptions reaching stratospheric levels
- Nadir instruments seem to underestimate SO₂ lifetimes in the UTLS
- Strong seasonal cycle in the northern hemispheric UT
- Enhanced values in monsoon regions

MIPAS OCS

- First global distributions of OCS including observation of tropical sink: need of much larger vegetation uptake in models
- Strong source over W-Pacific in spring: not well captured by models
- Biomass burning cannot be identified as a strong source of OCS