The Measured Stratospheric Sulfur Burden

Terry Deshler (University of Wyoming, Laramie)

Gas phase:

MIPAS (SO2, OCS) - Michael Höpfner, Anika Gunther, Norbert Glatthor (IMK-ASF, Karlsruhe)

ACE-FTS (OCS) - Marc von Hobbe, Corinna Kloss, (Forschungszentrum Juelich)

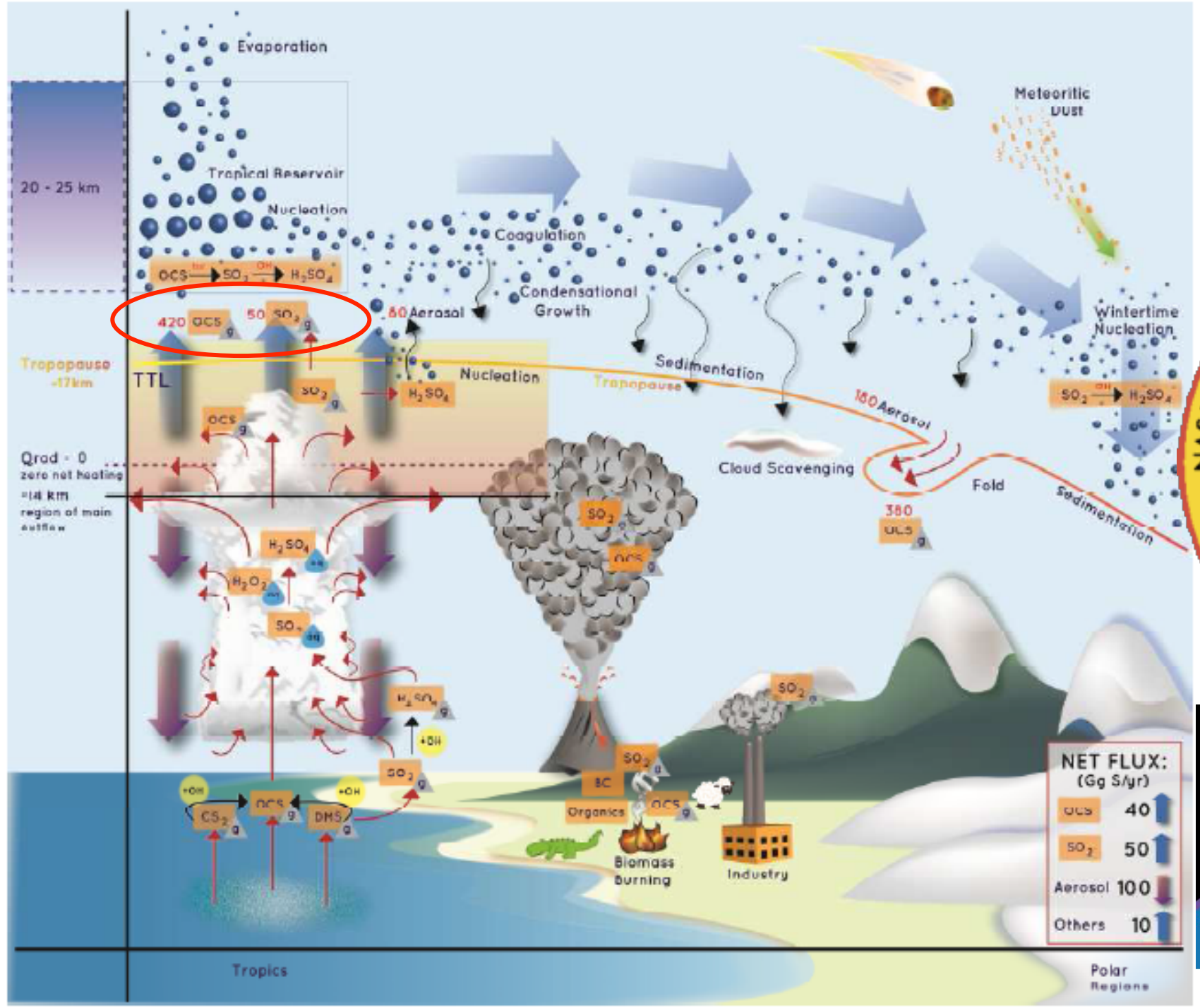
Ground based (OCS) - Stefanie Kremser, Nicholas Jones (Bodeker Scientific, Alexandra), Justus Notholt. (Univ. Bremen, Bremen)

Particle phase:

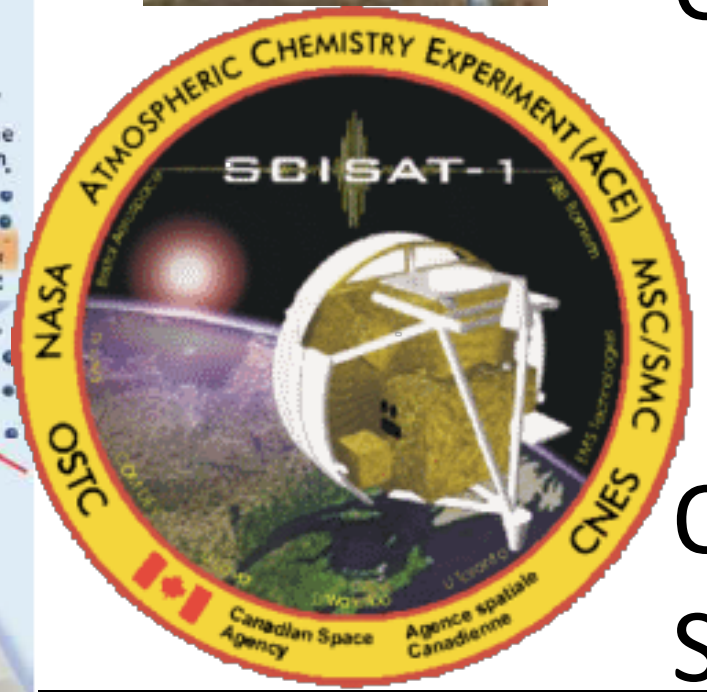
Satellite (Extinction): SAGE II - Larry Thomason (NASA-Langley, Hampton), OSIRIS - Adam Bourassa (Univ. Saskatchewan, Saskatoon)

Lidar (Backscatter) - Thomas Trickl (IMK-IFU, Garmisch-Partenkirchen), John Barnes (NOAA, Hilo)

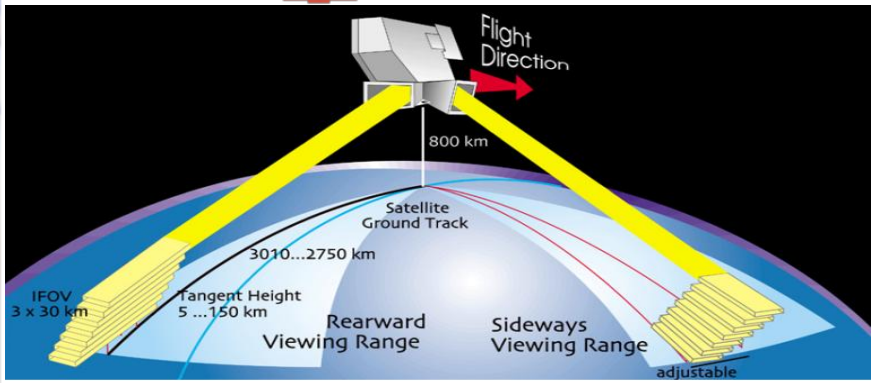
In situ (Size Distribution): Carribic - Markus Herman, (Leibniz Institute) Bengt Martinsson, (Lund University), FCAS - Chuck Wilson, Brian Meland, (Denver University), UWOPC - Terry Deshler (University of Wyoming)

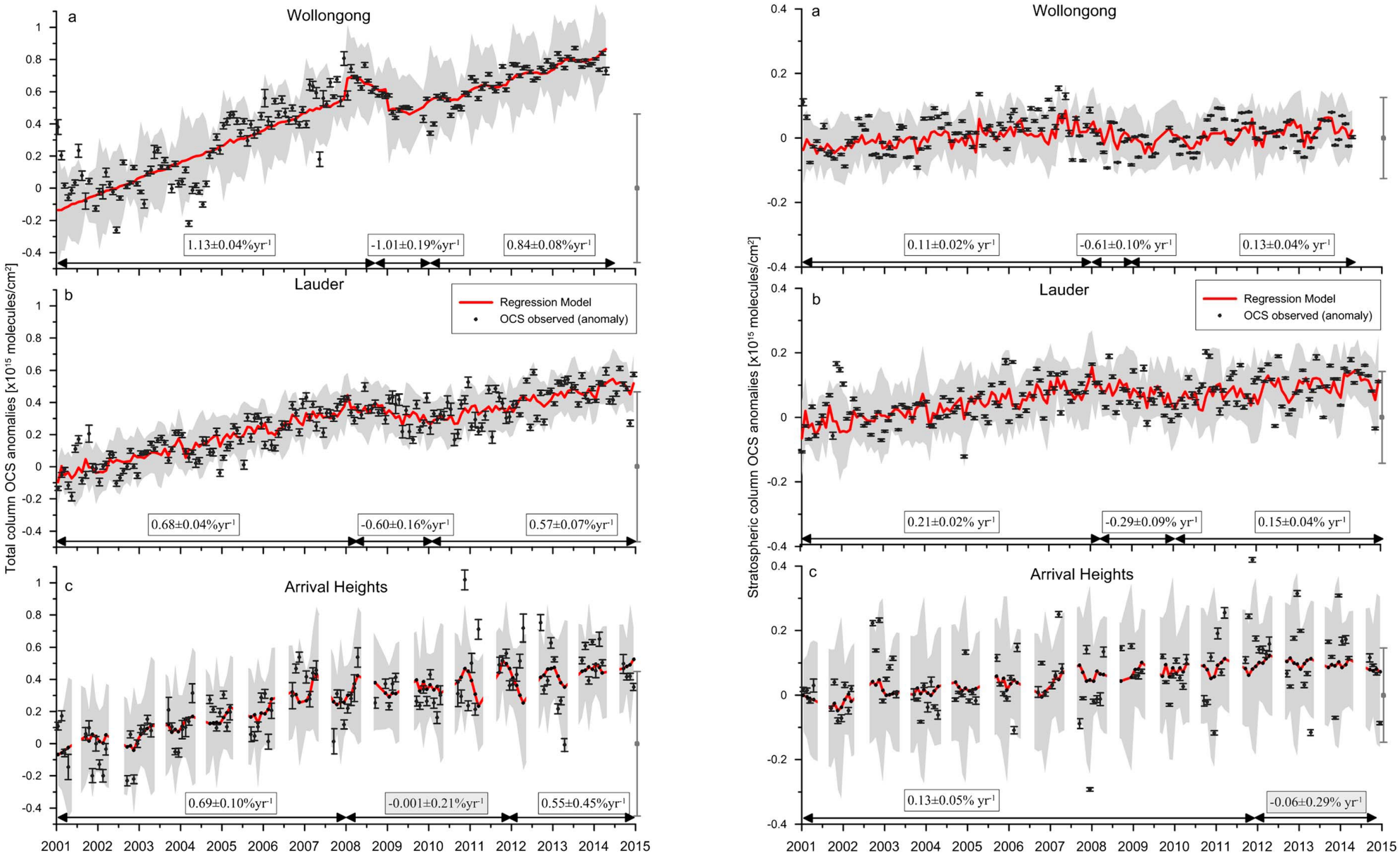

Evaporation
20 - 25 km
Tropical Reservoir
Nucleation
Coagulation
Condensational Growth
Meteoritic Dust
Wintertime Nucleation
Sedimentation
Tropopause
Nucleation
TTL
Cloud Scavenging
Fold
Qrad = 0
zero net heating
region of main outflow
Biomass Burning
Industry
Organics
NET FLUX:
(Gg S/yr)
OCS 40
SO2 50
Aerosol 100
Others 10
Tropics
Polar Regions

OCS

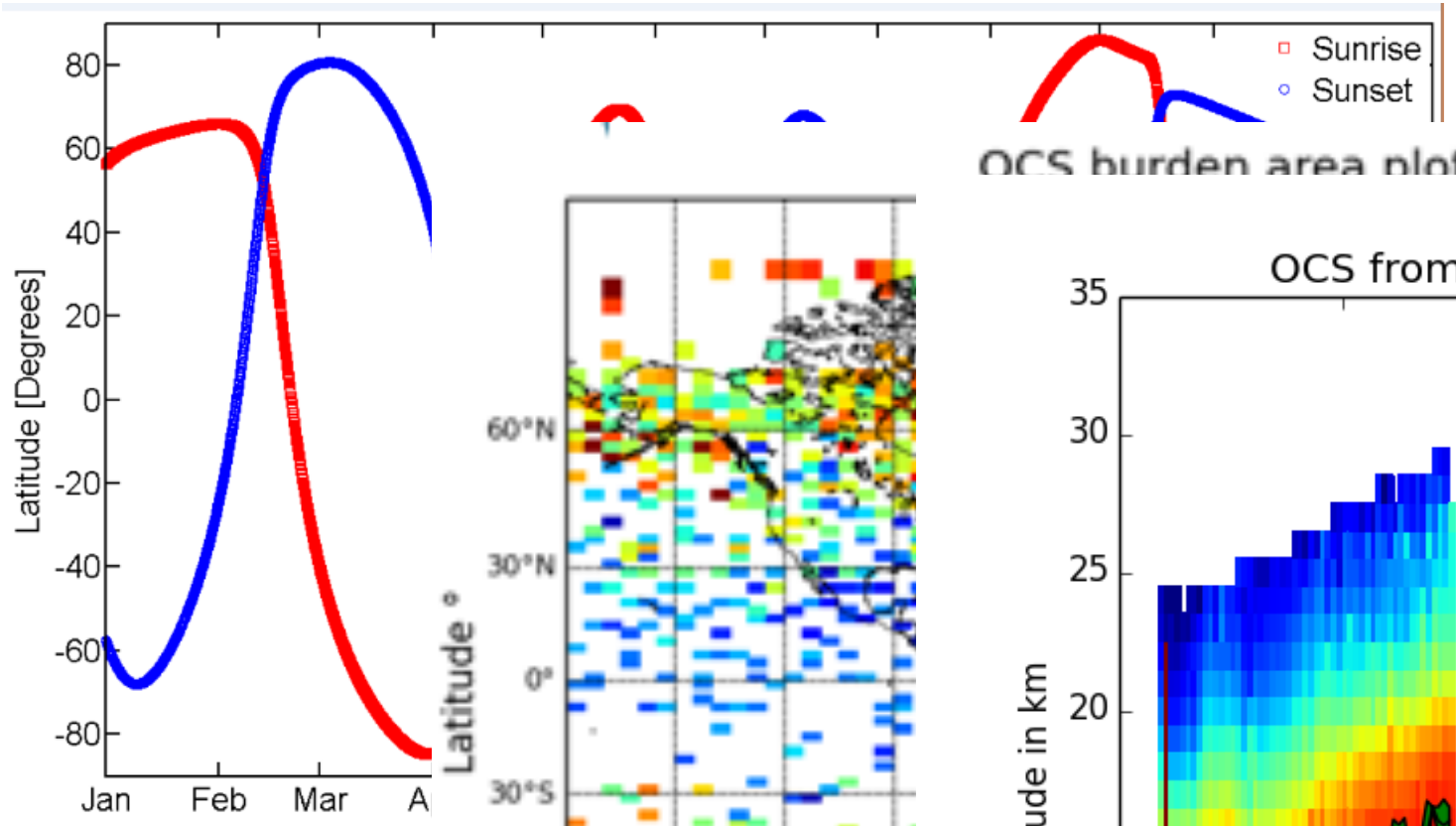

ATMOSPHERIC CHEMISTRY EXPERIMENT (ACE)
SCISAT-1
NASA
OSTC
MSC/SMC
CNES
Canadian Space Agency
Agence spatiale Canadienne

OCS
SO2

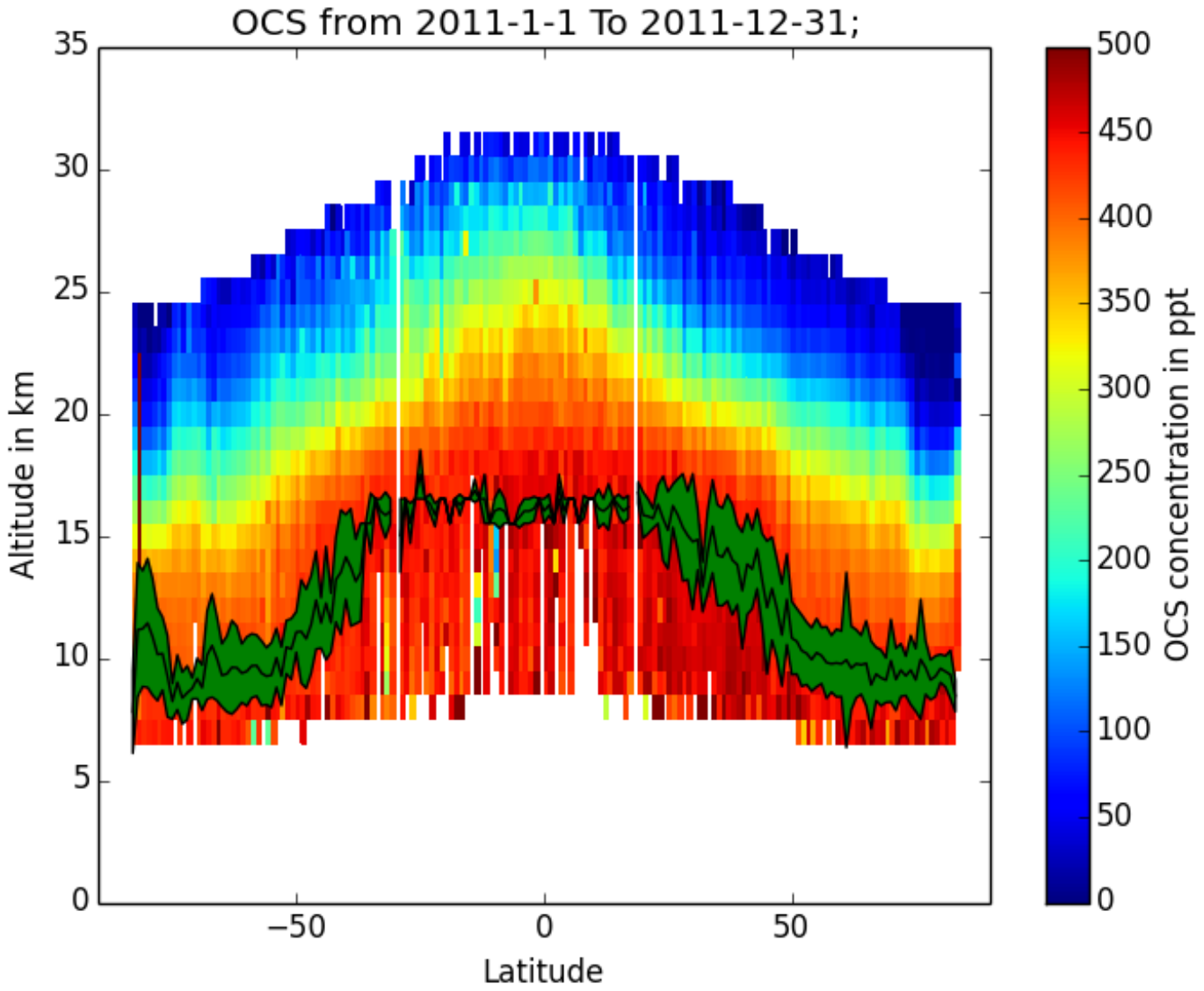

Flight Direction
800 km
Satellite Ground Track
3010...2750 km
IFOV 3 x 30 km
Tangent Height 5 ...150 km
Rearward Viewing Range
Sideways Viewing Range
adjustable

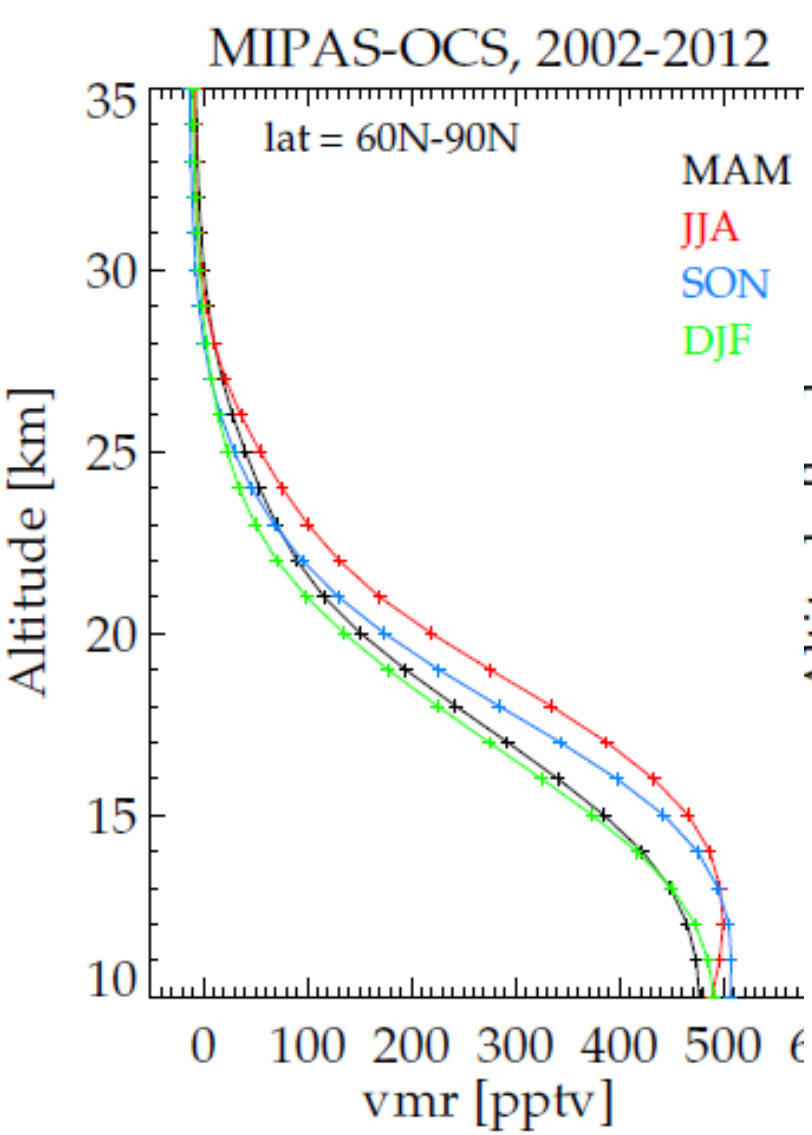


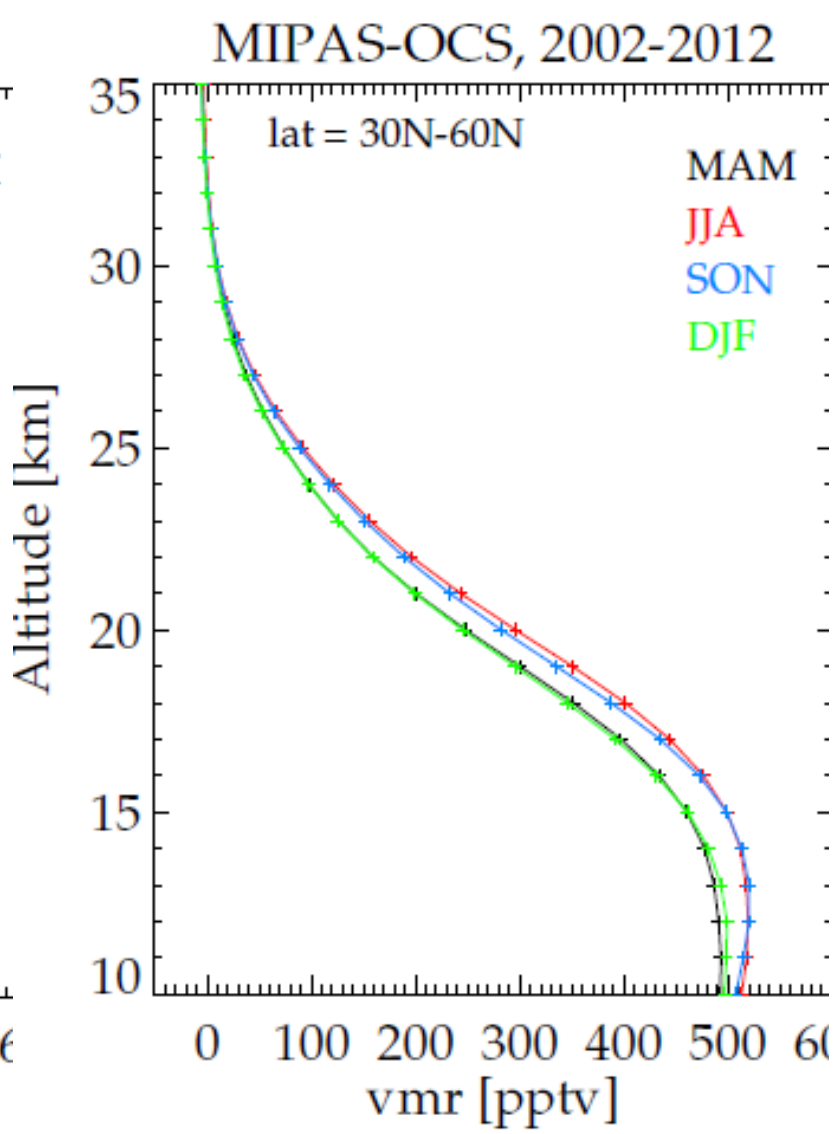
Kremser, S., N. B. Jones, M. Palm, B. Lejeune, Y. Wang, D. Smale, and N. M. Deutscher (2015), Positive trends in Southern Hemisphere carbonyl sulfide, Geophys. Res. Lett., 42

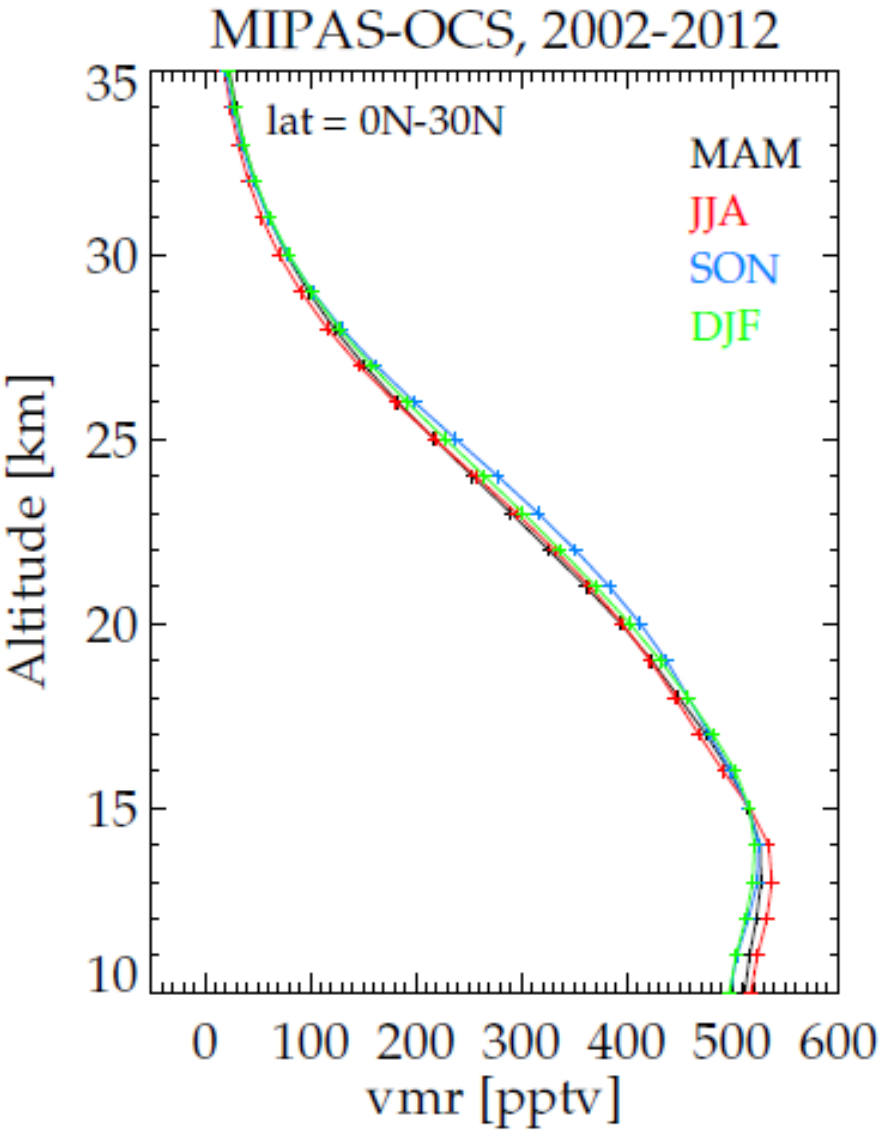


OCS burden area plot

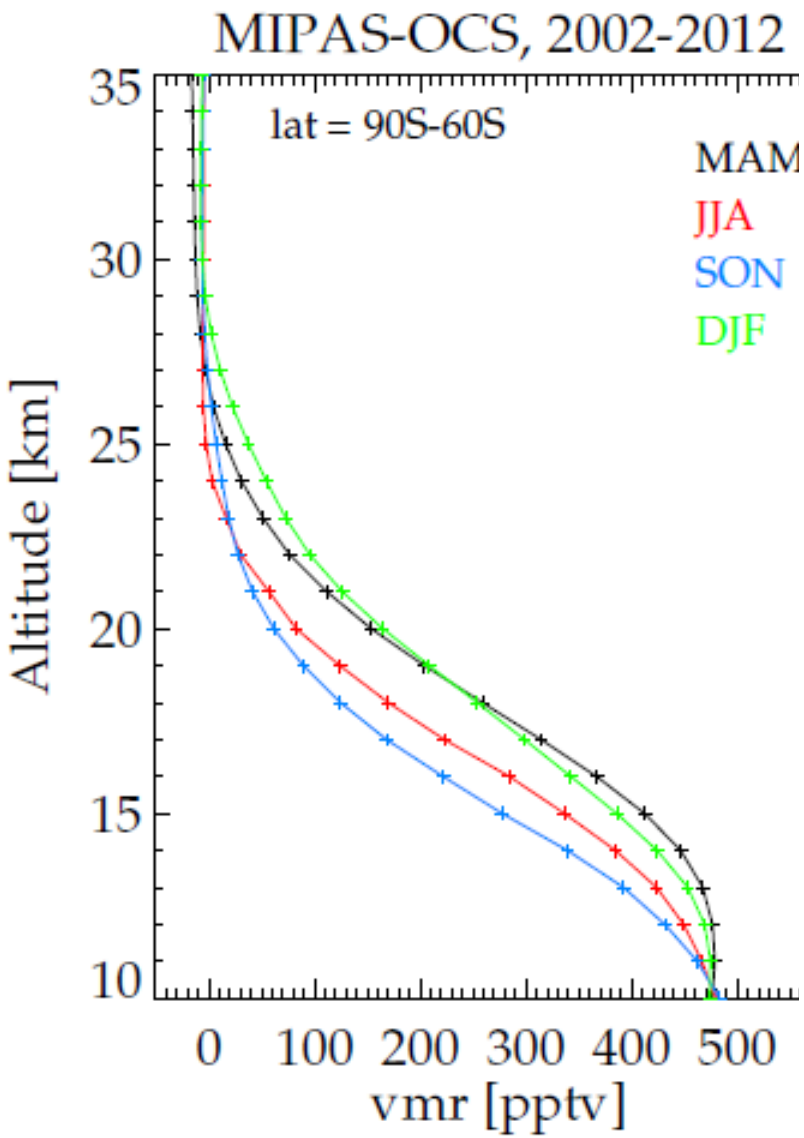


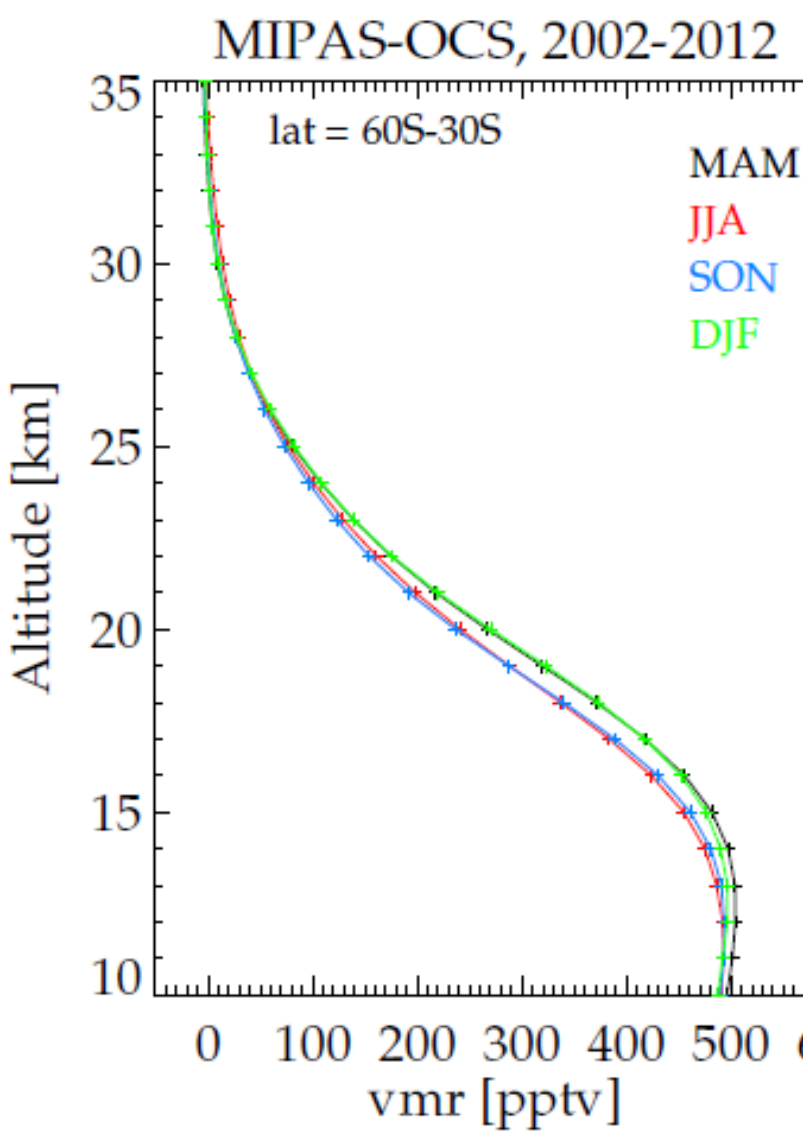


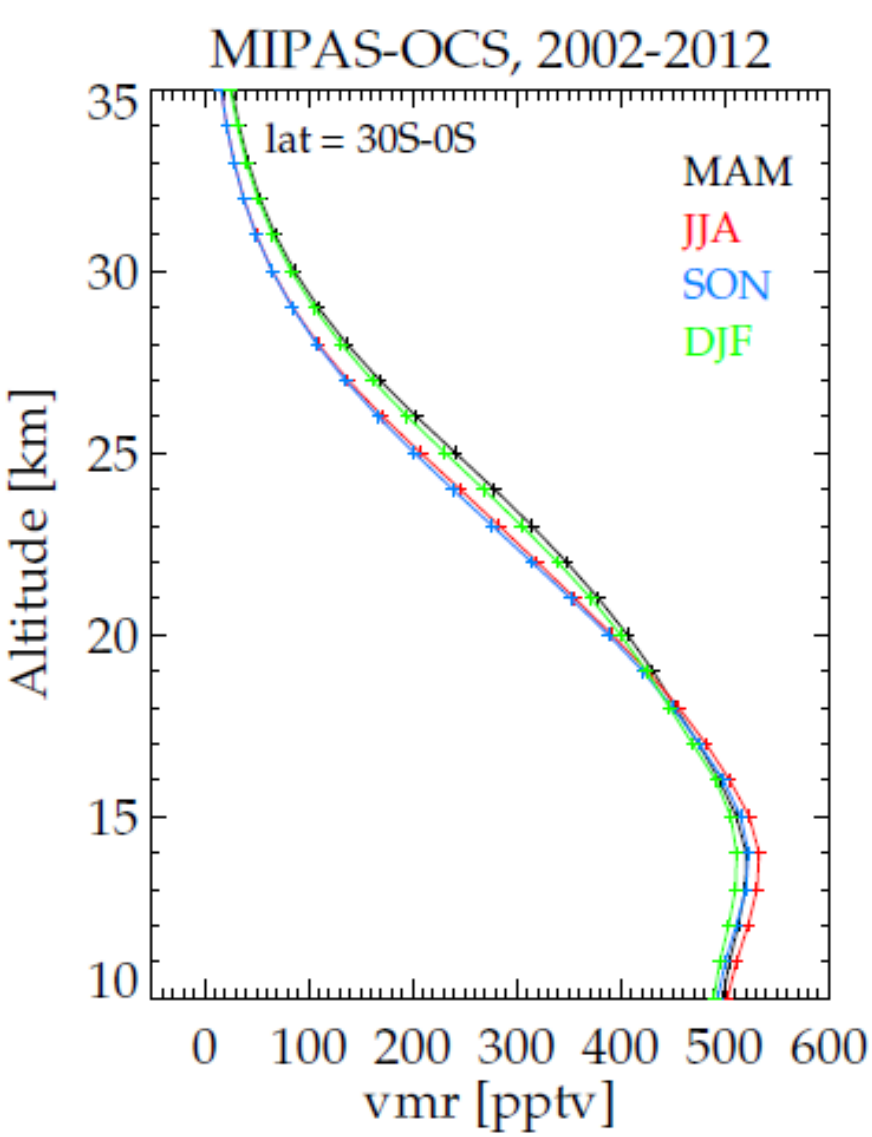




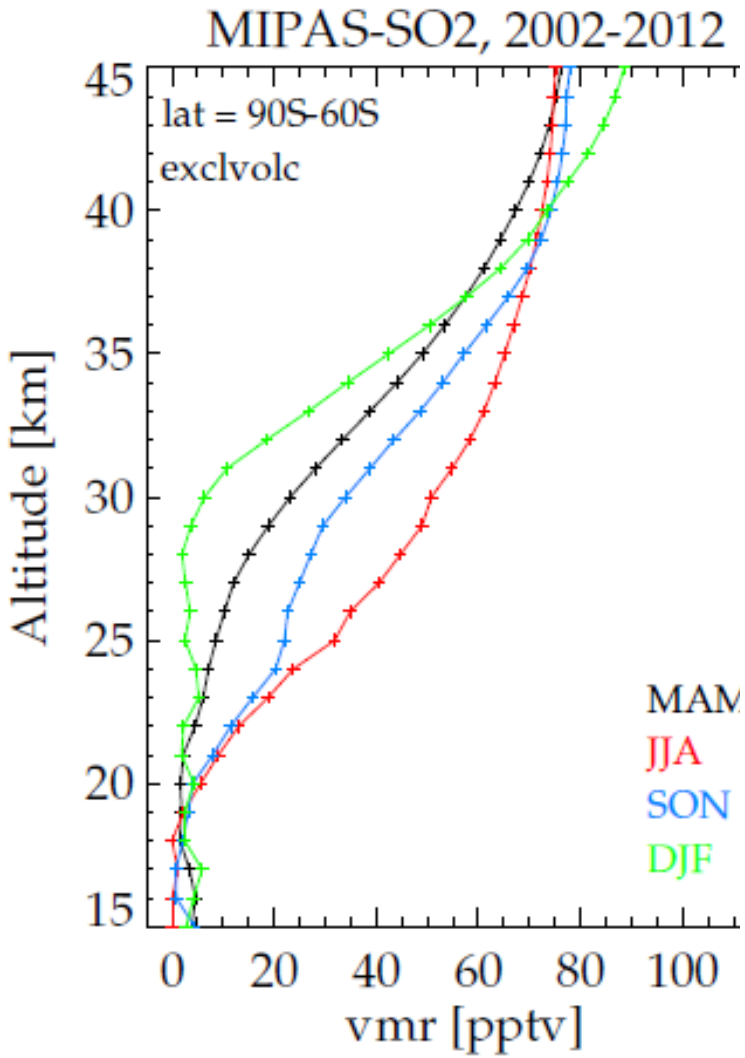
Höpfner et al., 2013, ACP.

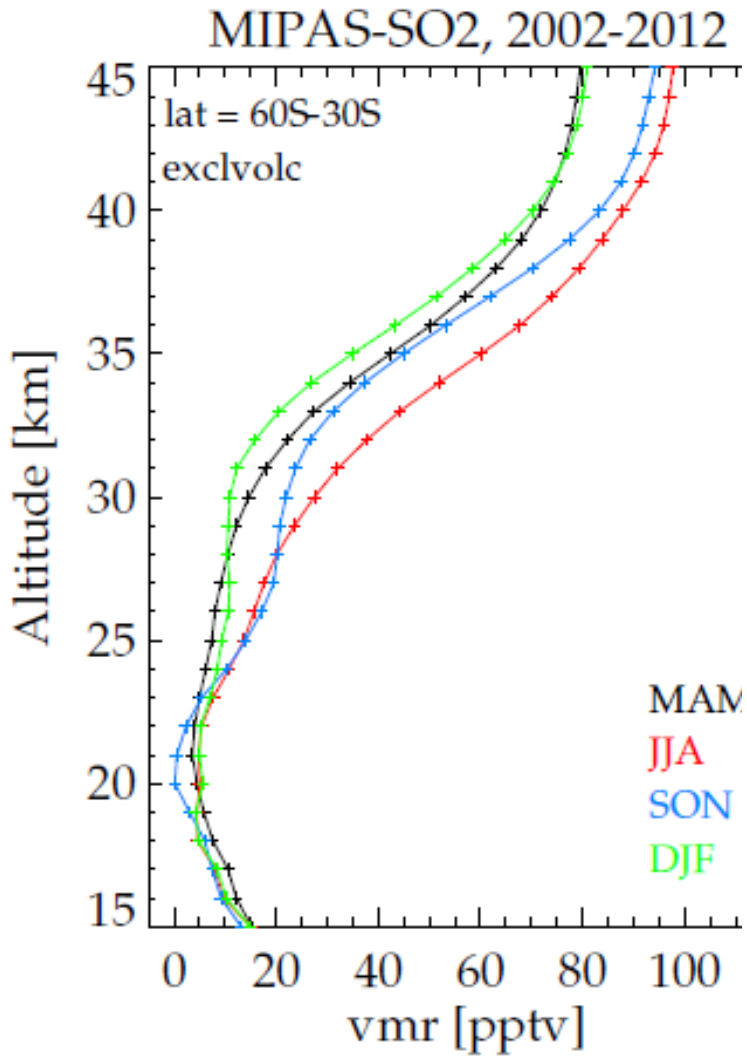






Höpfner et al., 2013, ACP.


MIPAS-SO2, 2002-2012
lat = 90S-60S
exclvolc
MAM
JJA
SON
DJF
Altitude [km]
vmr [pptv]
15
20
25
30
35
40
45
0
20
40
60
80
100


MIPAS-SO2, 2002-2012
lat = 60S-30S
exclvolc
MAM
JJA
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Altitude [km]
vmr [pptv]
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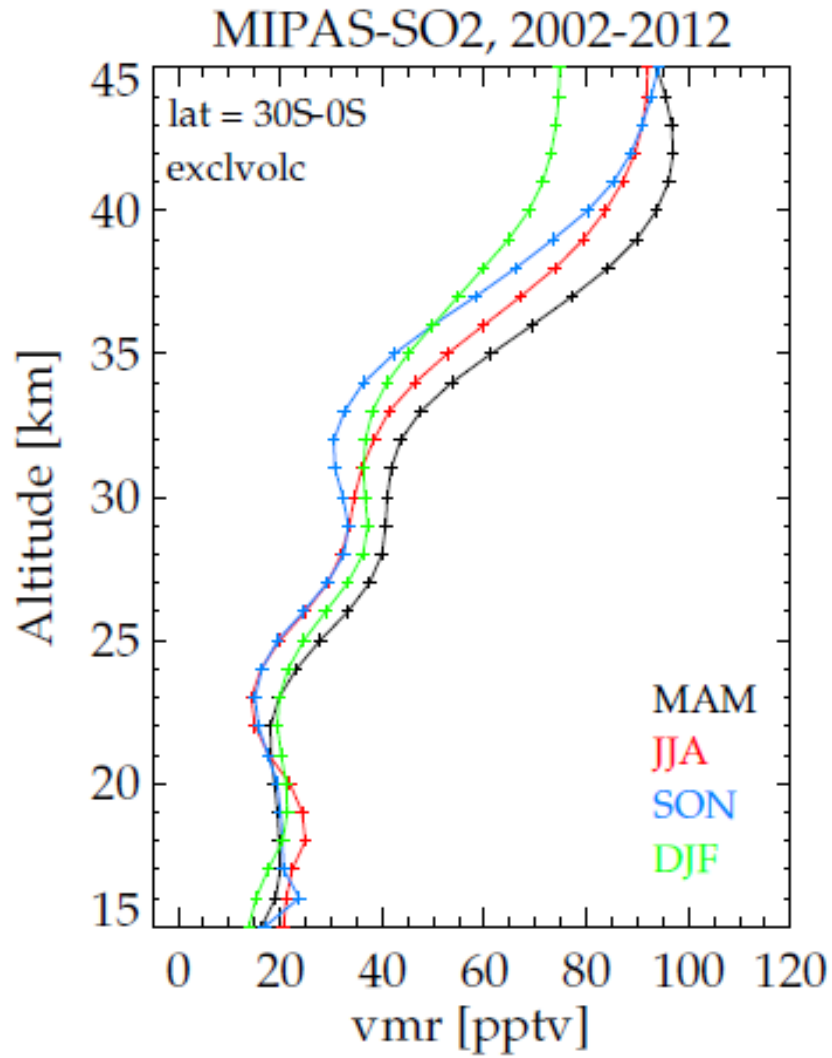
MIPAS-SO2, 2002-2012
lat = 30S-0S
exclvolc
MAM
JJA
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DJF
Altitude [km]
vmr [pptv]
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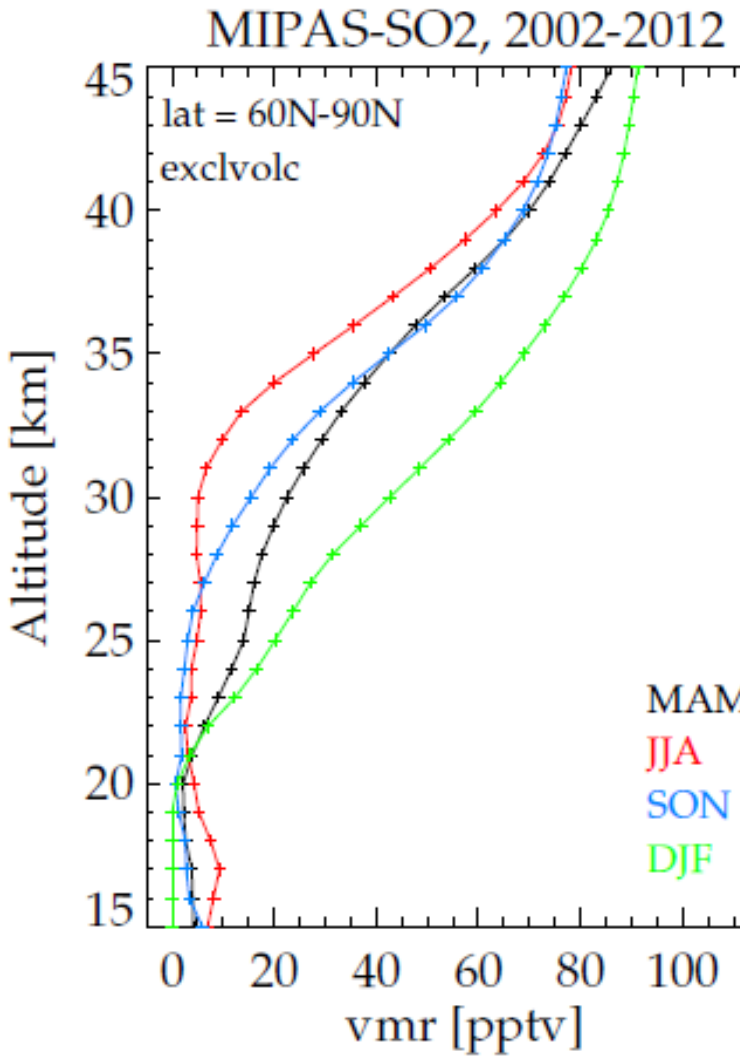
MIPAS-SO2, 2002-2012
lat = 60N-90N
exclvolc
MAM
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Altitude [km]
vmr [pptv]
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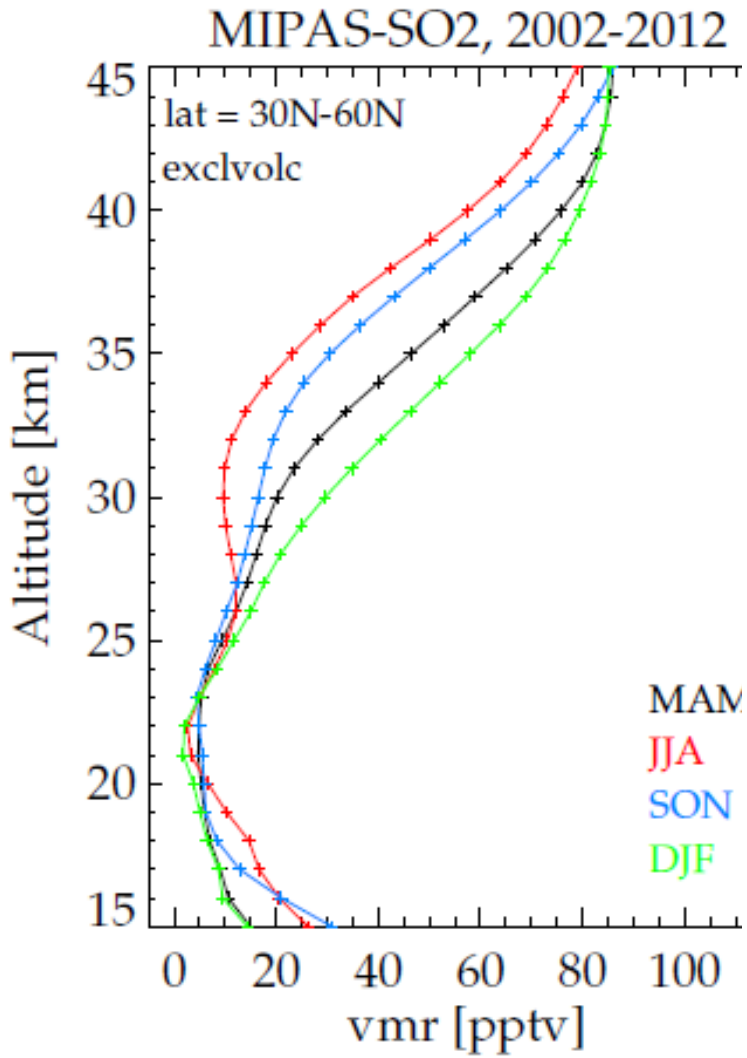
MIPAS-SO2, 2002-2012
lat = 30N-60N
exclvolc
MAM
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Altitude [km]
vmr [pptv]
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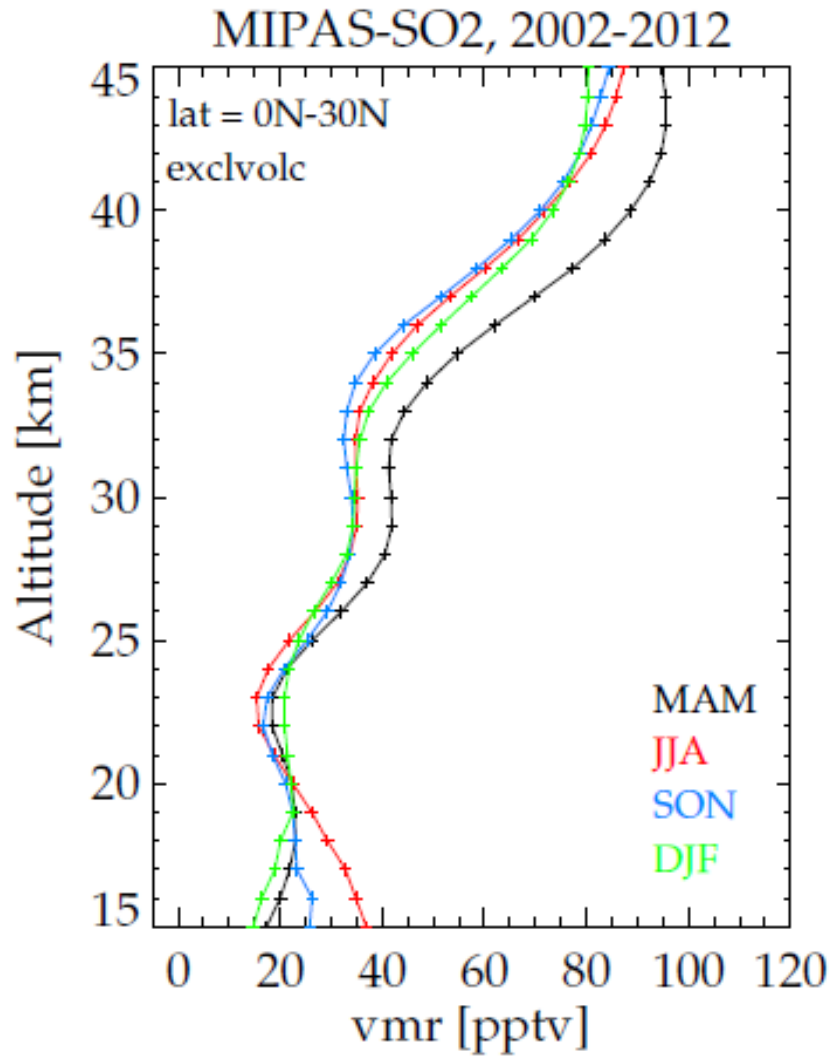
MIPAS-SO2, 2002-2012
lat = 0N-30N
exclvolc
MAM
JJA
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DJF
Altitude [km]
vmr [pptv]
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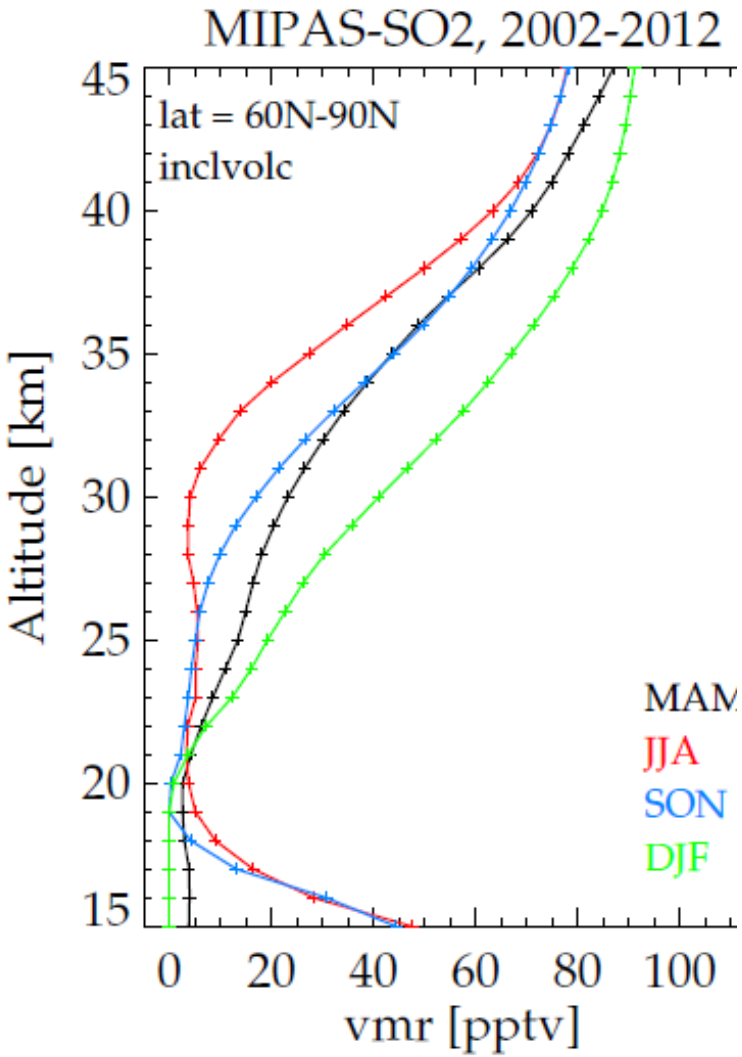
MIPAS-SO2, 2002-2012
lat = 60N-90N
inclvolc
MAM
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Altitude [km]
vmr [pptv]
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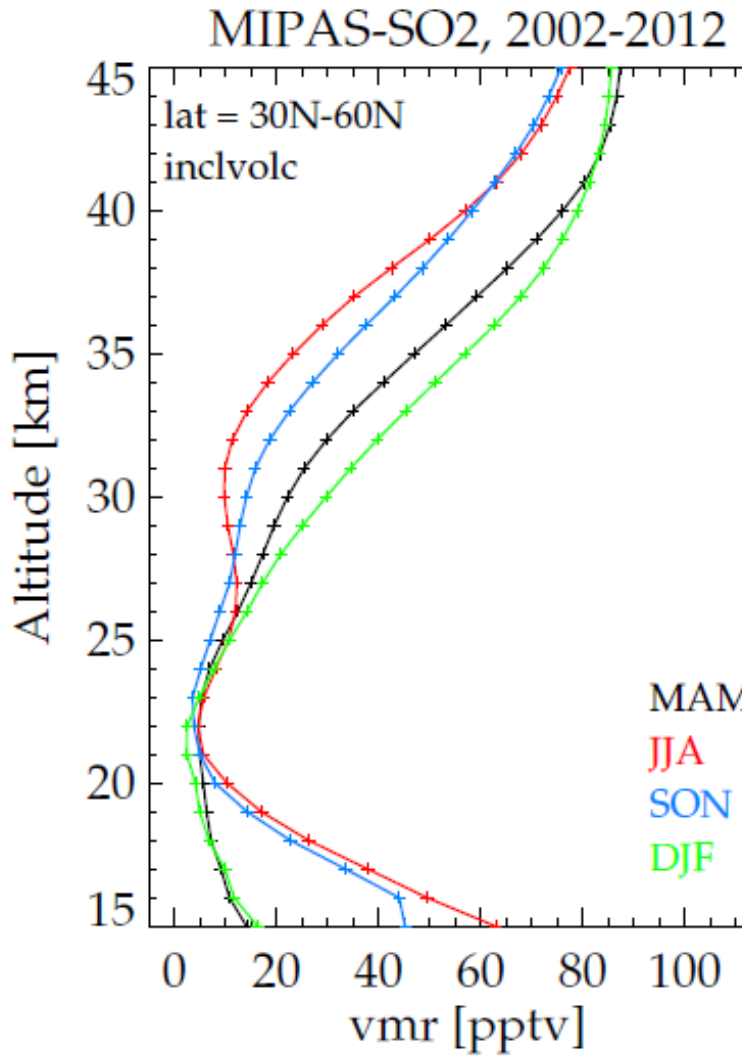
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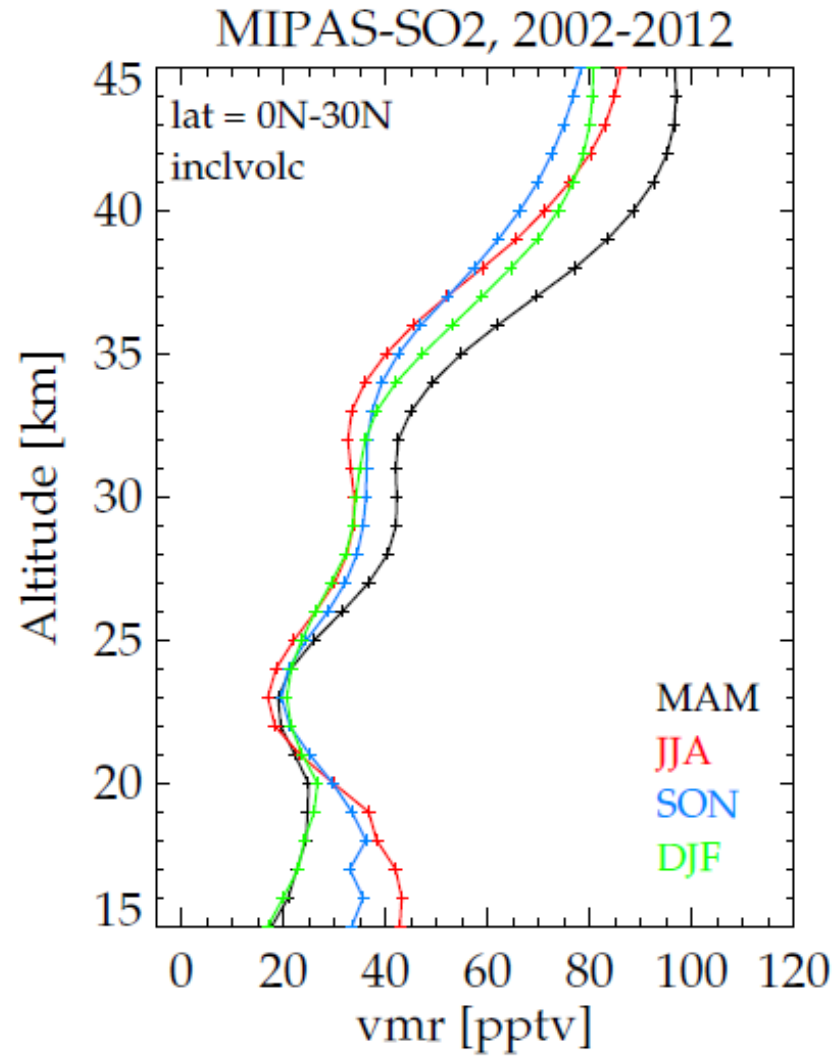
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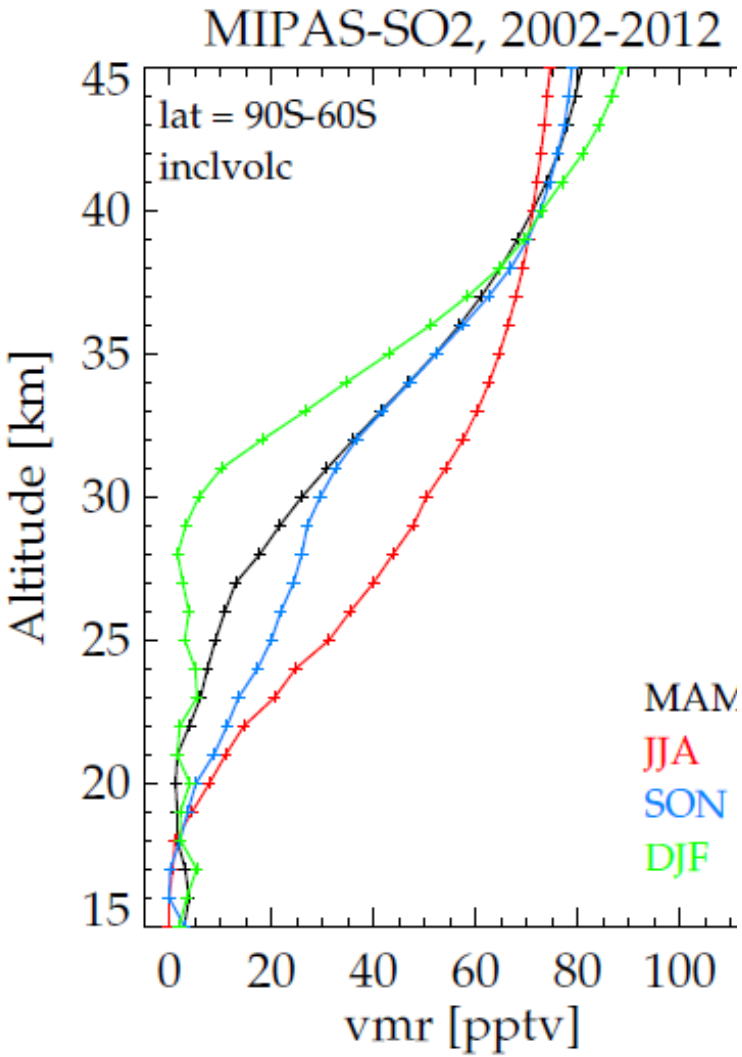
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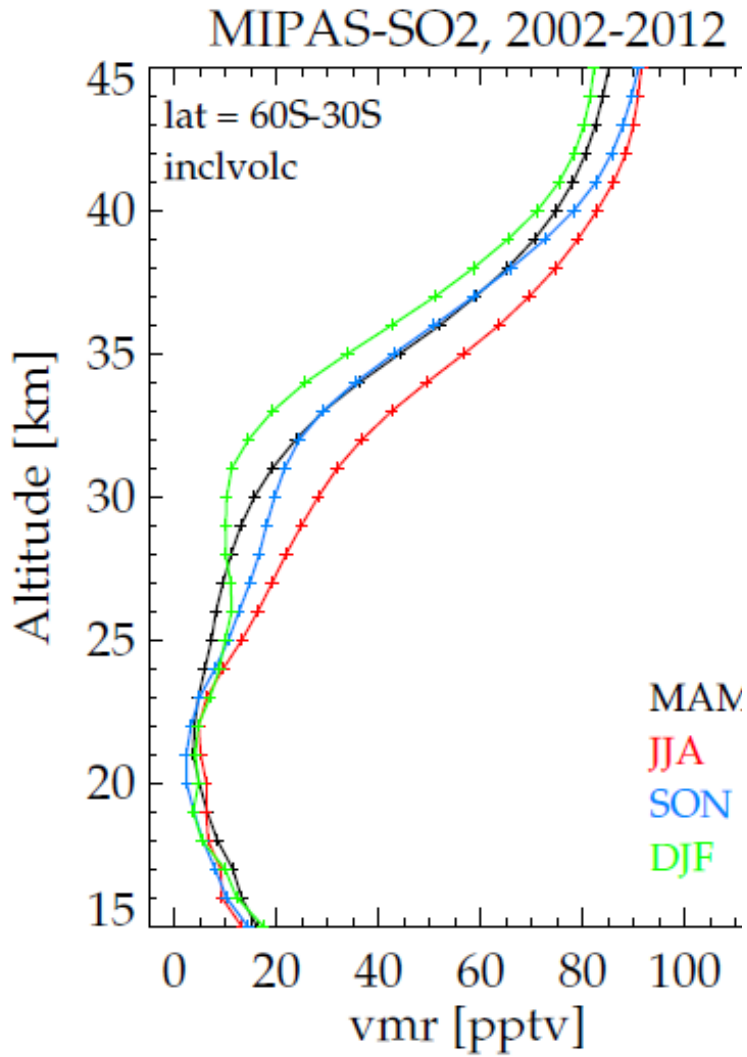
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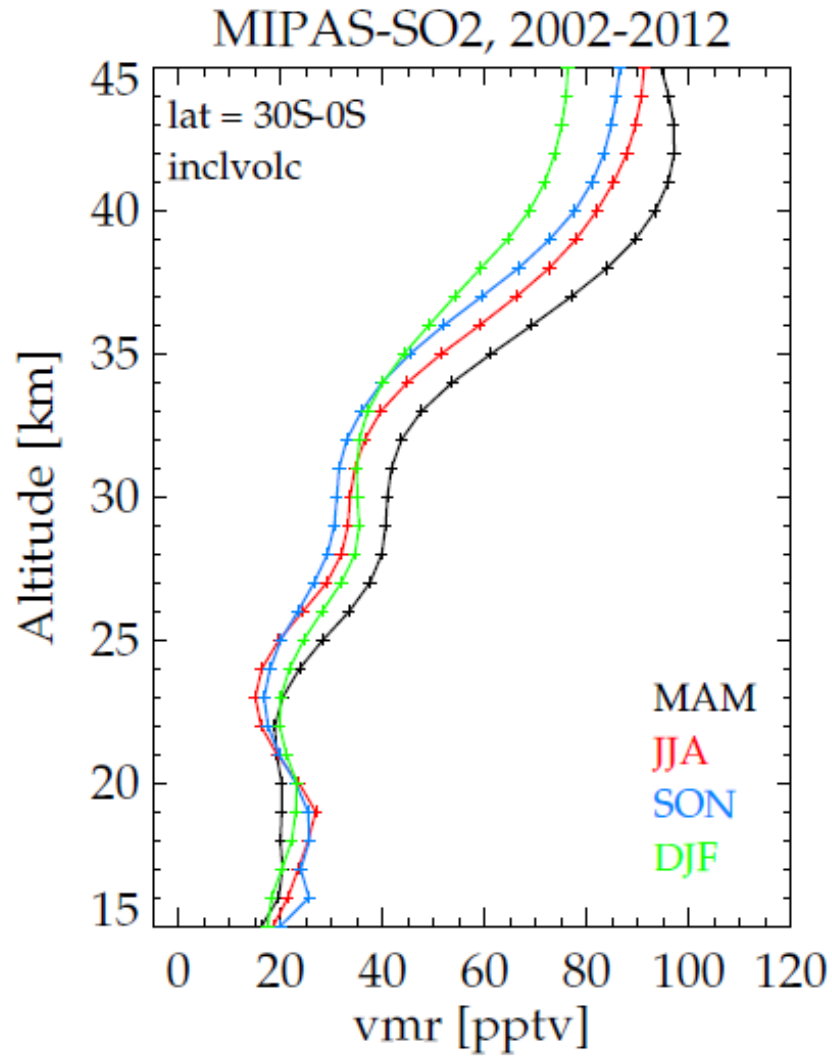
MIPAS-SO2, 2002-2012
lat = 30S-0S
inclvolc
MAM
JJA
SON
DJF
Altitude [km]
vmr [pptv]
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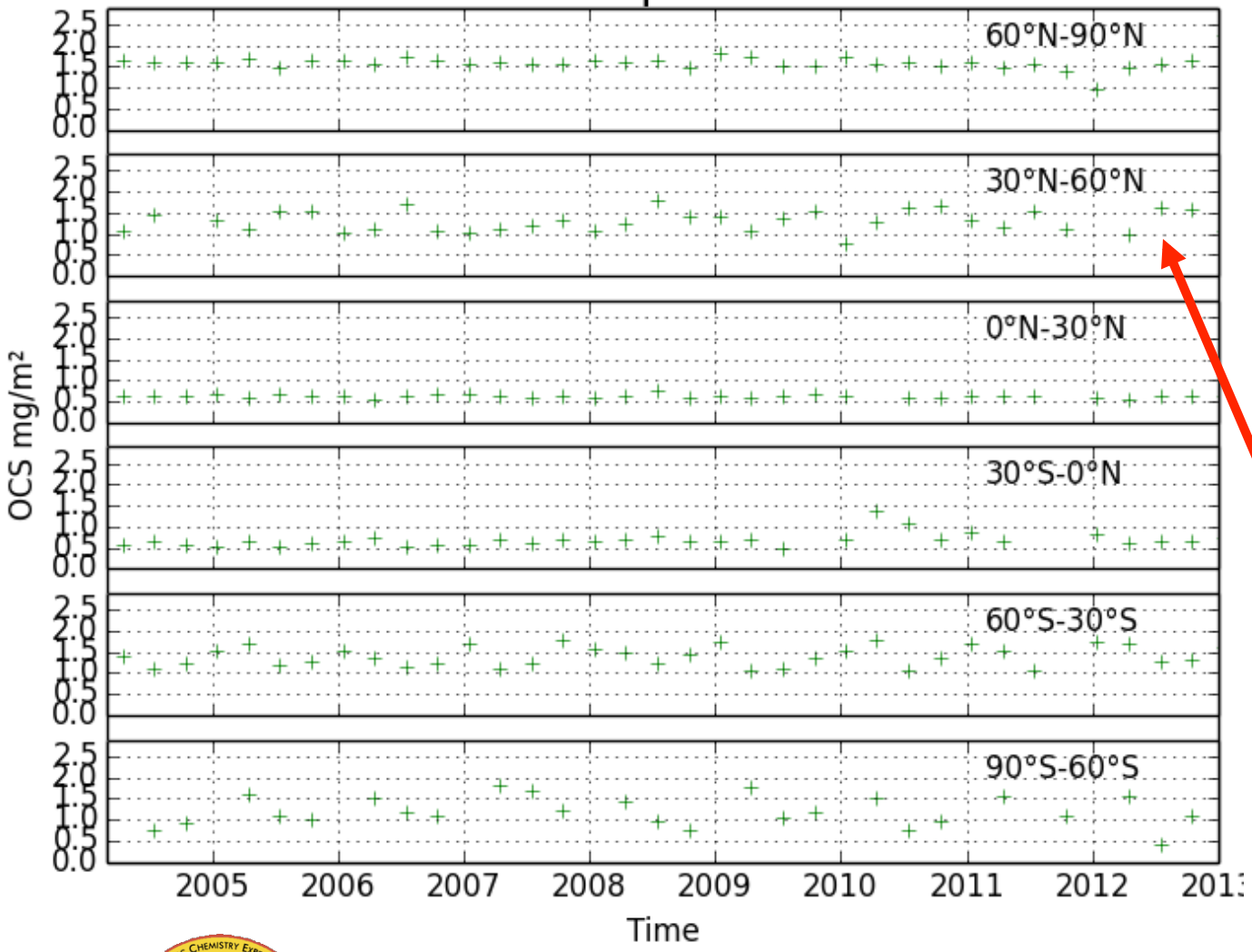
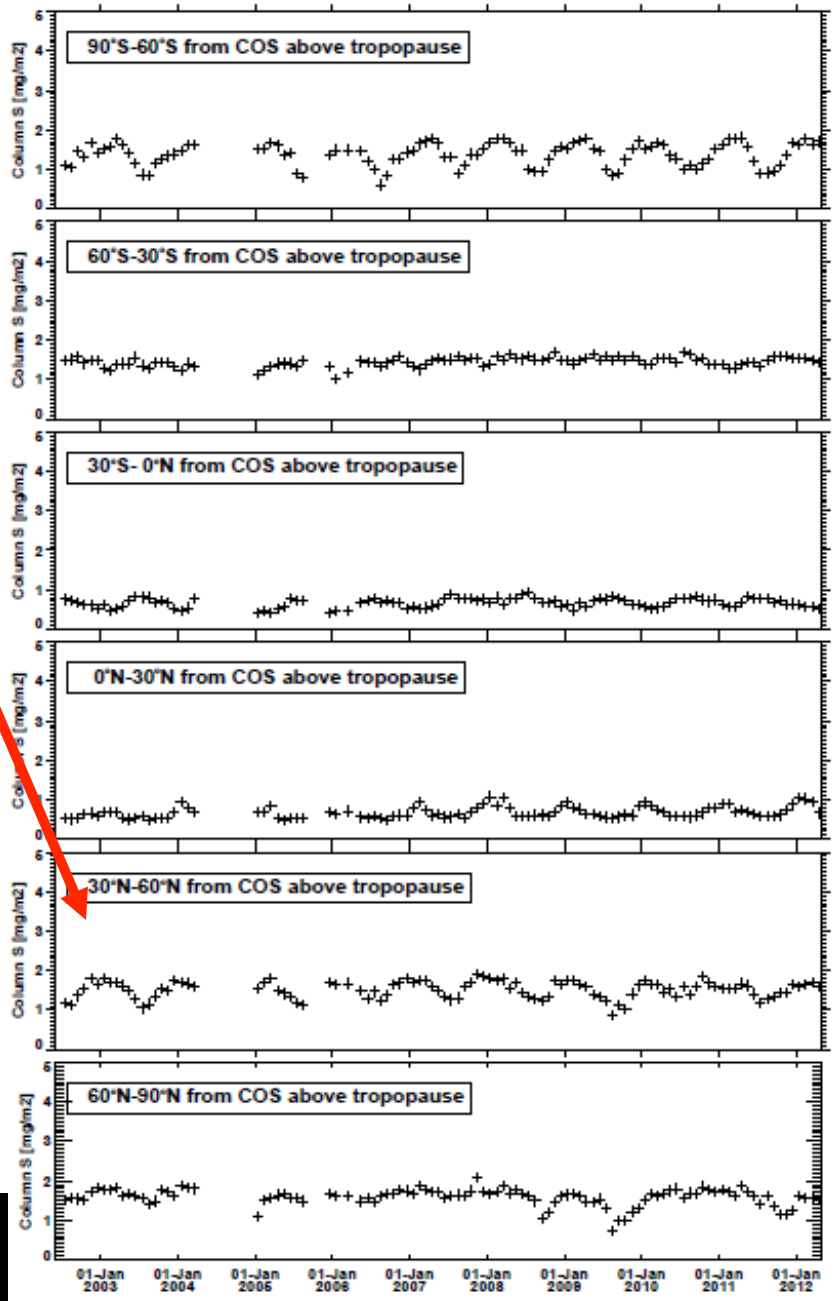
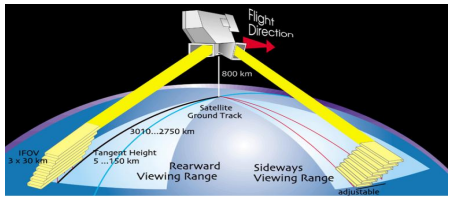
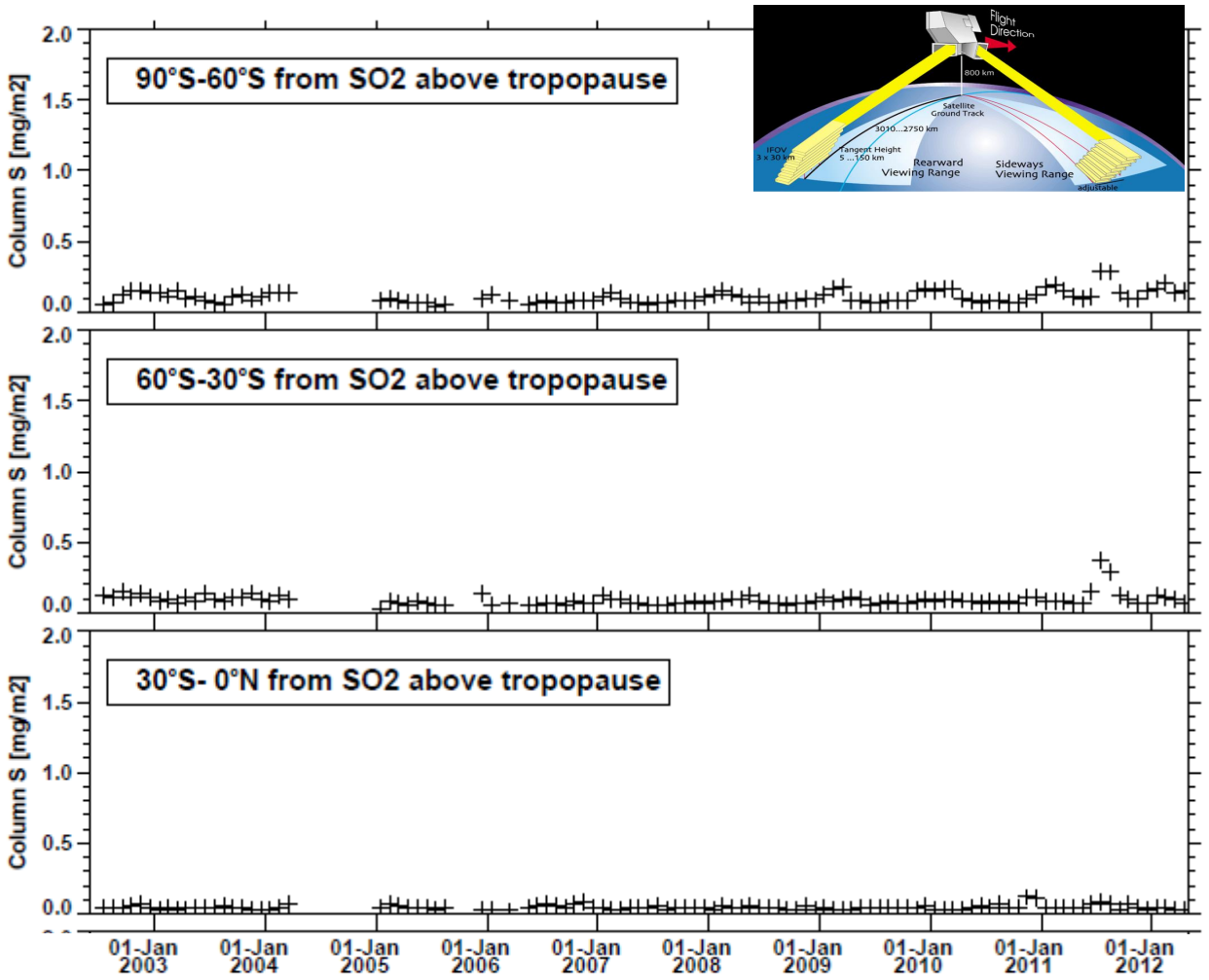
Seasonal Stratospheric OCS Burden

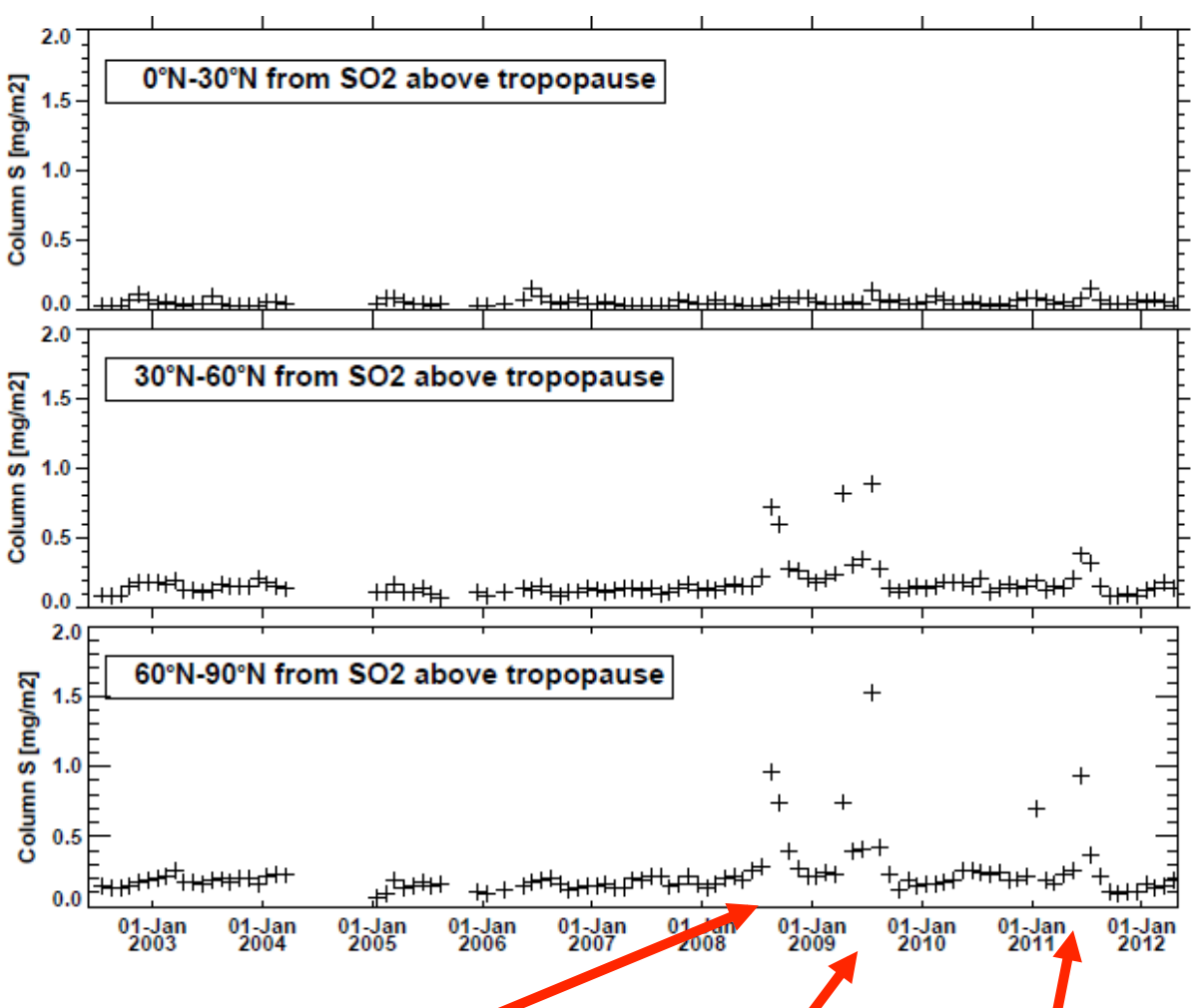


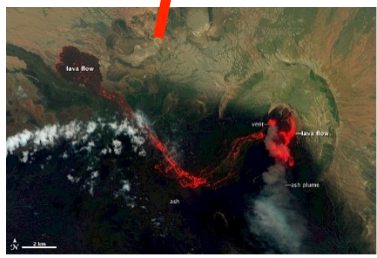


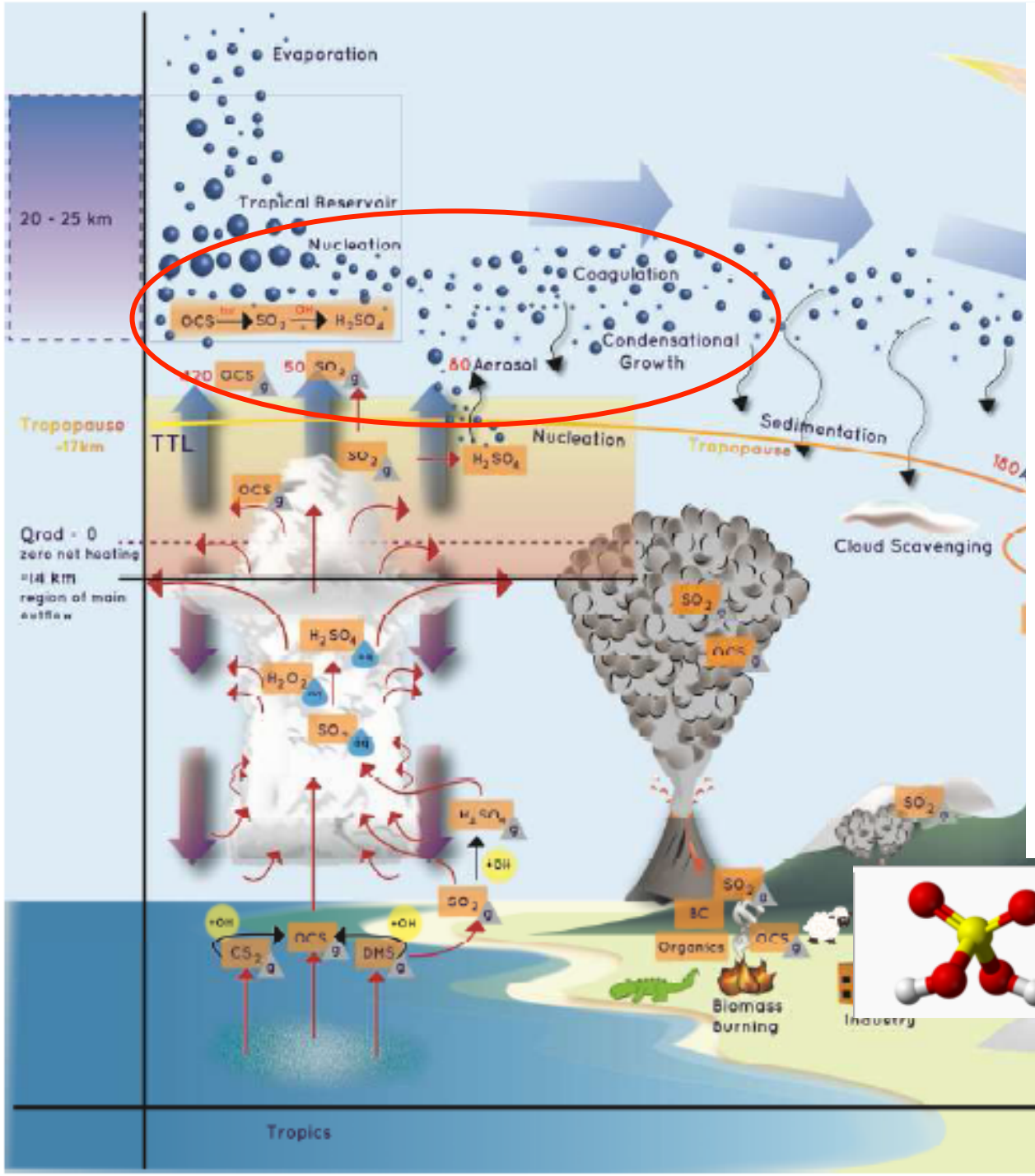


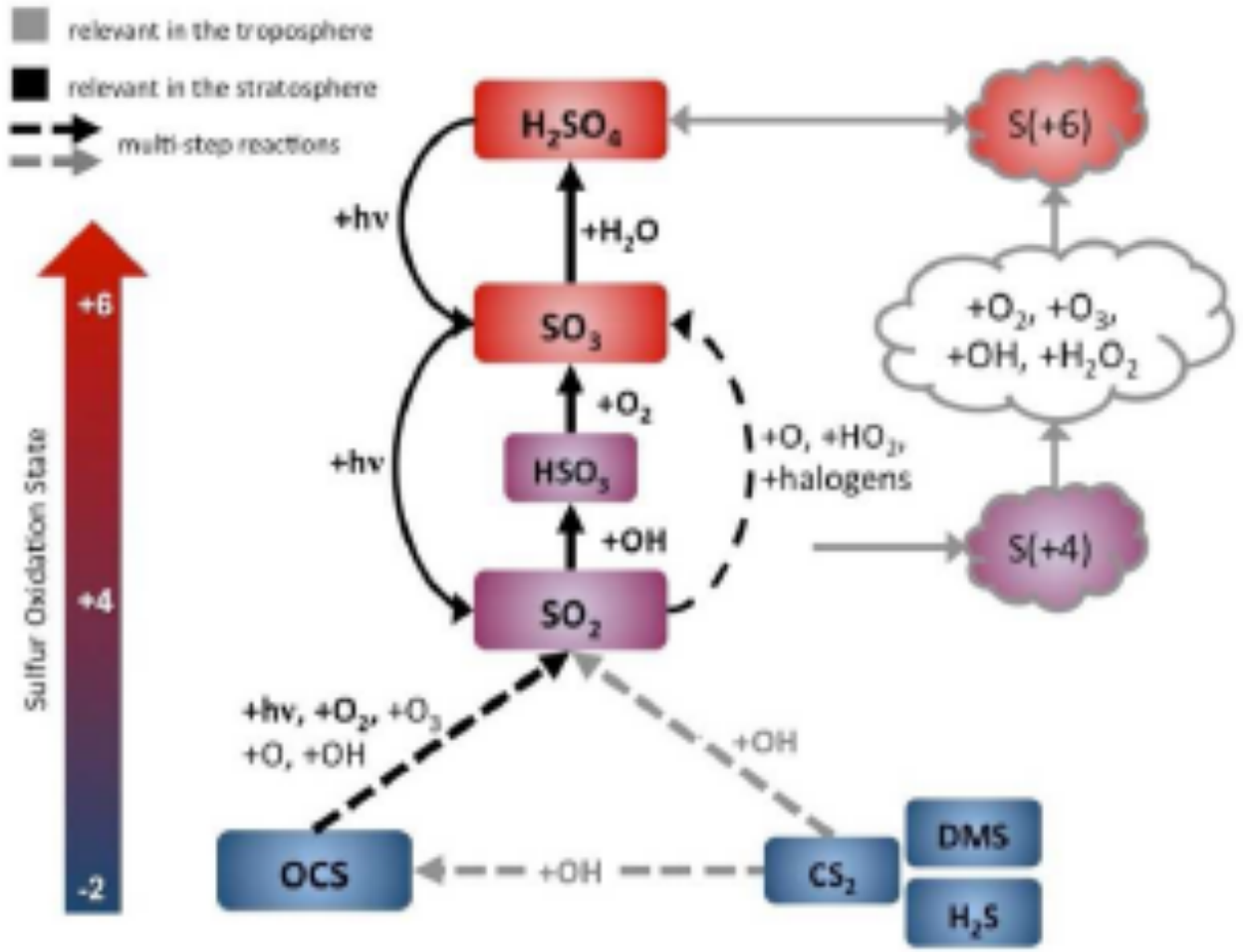

90°S-60°S from SO2 above tropopause
60°S-30°S from SO2 above tropopause
30°S- 0°N from SO2 above tropopause
Column S [mg/m2]
2.0
1.5
1.0
0.5
0.0
01-Jan 2003
01-Jan 2004
01-Jan 2005
01-Jan 2006
01-Jan 2007
01-Jan 2008
01-Jan 2009
01-Jan 2010
01-Jan 2011
01-Jan 2012

Flight Direction
800 km
Satellite Ground Track
3010...2750 km
Tangent Height 5 ...150 km
IFOV 3 x 30 km
Rearward Viewing Range
Sideways Viewing Range
adjustable


0°N-30°N from SO2 above tropopause
30°N-60°N from SO2 above tropopause
60°N-90°N from SO2 above tropopause
Column S [mg/m2]
2.0
1.5
1.0
0.5
0.0
01-Jan 2003
01-Jan 2004
01-Jan 2005
01-Jan 2006
01-Jan 2007
01-Jan 2008
01-Jan 2009
01-Jan 2010
01-Jan 2011
01-Jan 2012

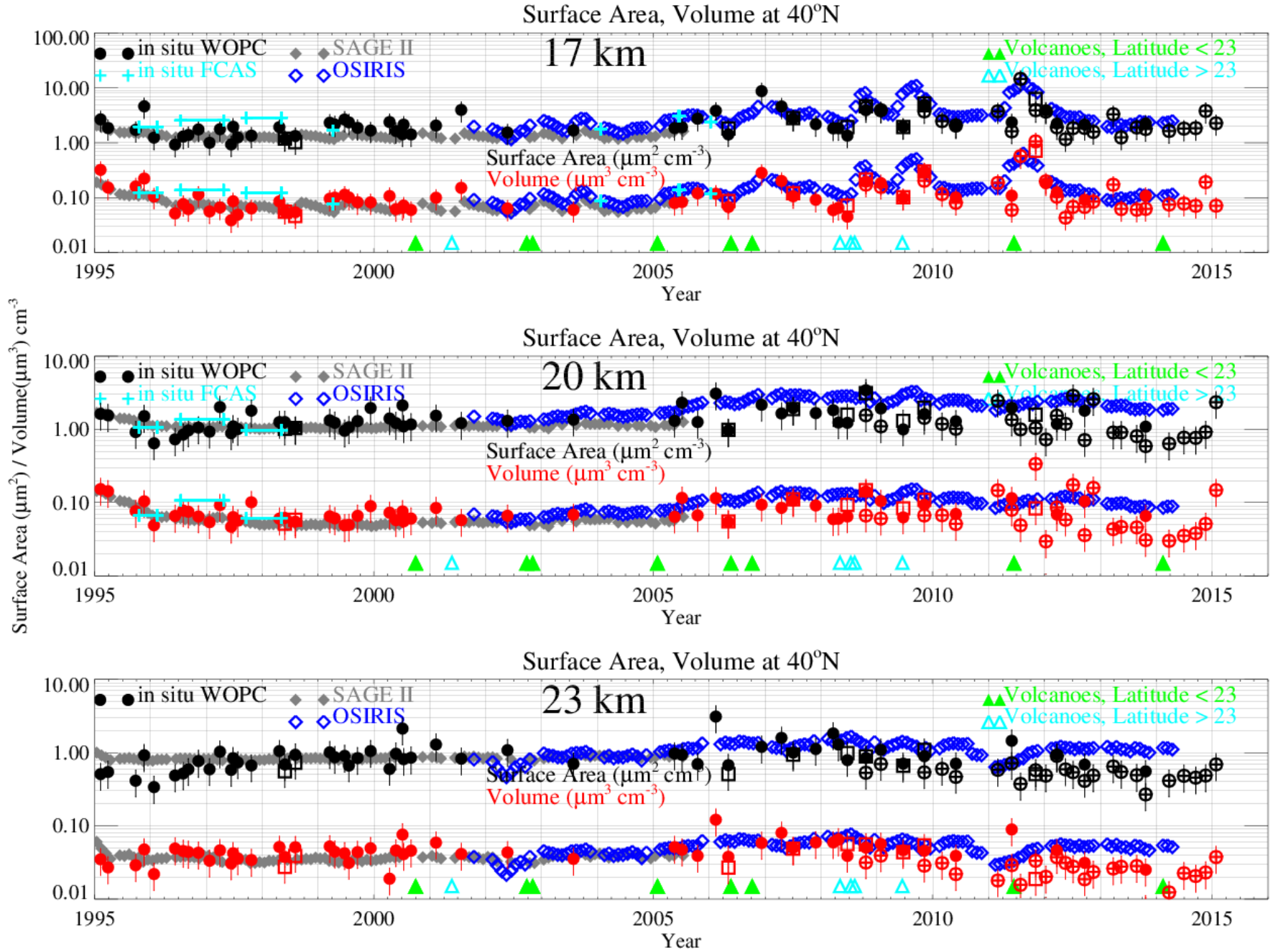


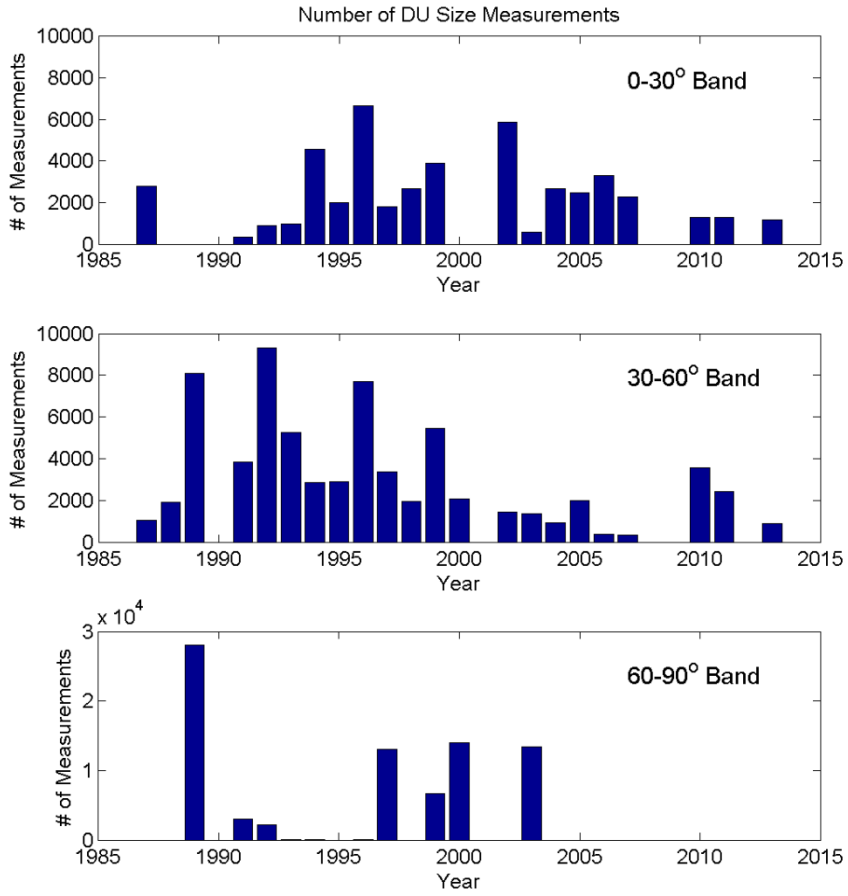


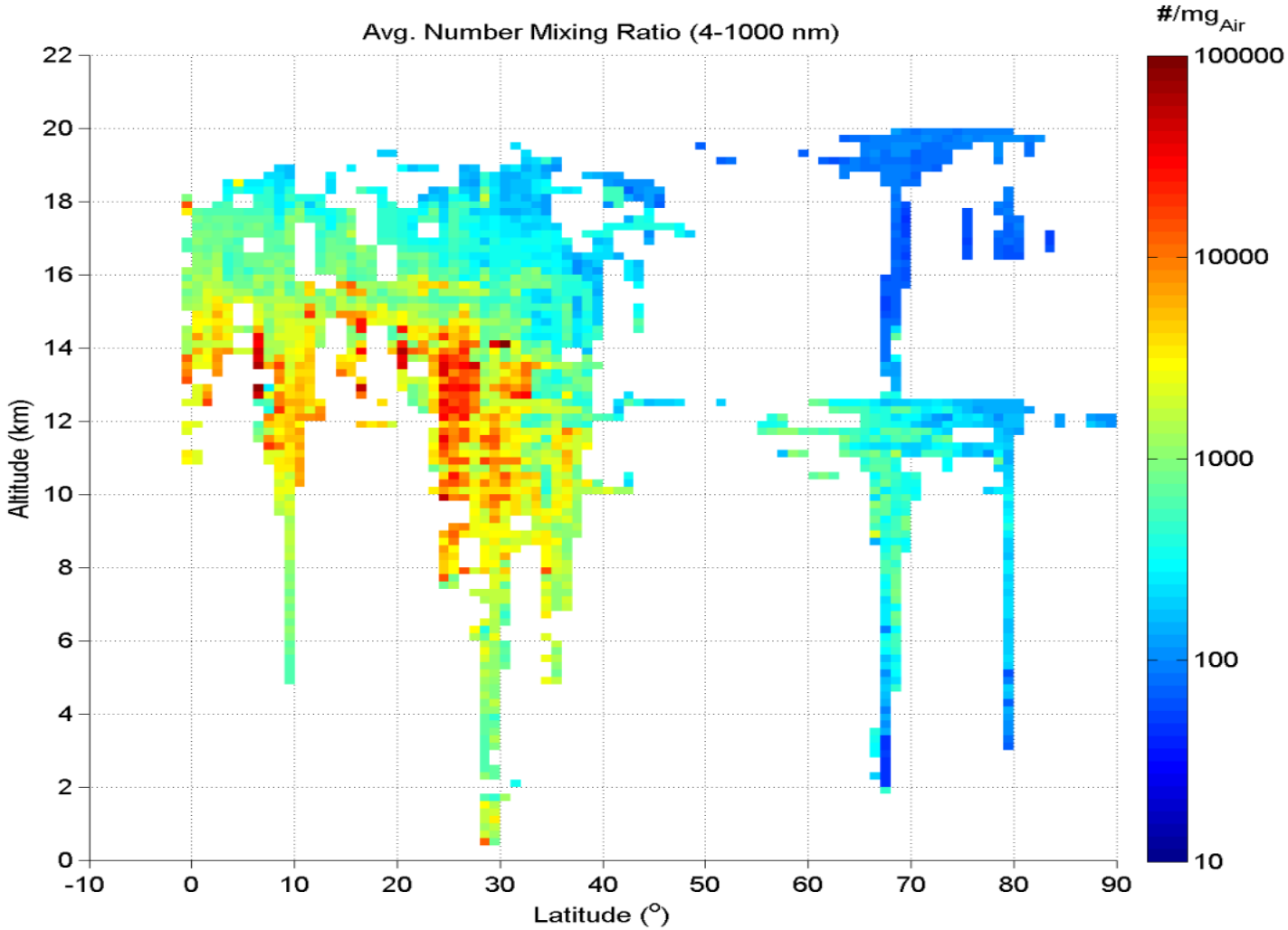


$$H_2SO_4 + H_2O \rightarrow H_3O^+ + HSO_4^-$$

$$HSO_4^- + H_2O \rightarrow H_3O^+ + SO_4^{2-}$$


Surface Area, Volume at 40°N
17 km
in situ WOPC
SAGE II
in situ FCAS
OSIRIS
Volcanoes, Latitude < 23
Volcanoes, Latitude > 23
Surface Area (μm² cm⁻³)
Volume (μm³ cm⁻³)
100.00
10.00
1.00
0.10
0.01
1995
2000
2005
2010
2015
Year
Surface Area, Volume at 40°N
20 km
Surface Area (μm²) / Volume(μm³) cm⁻³
Surface Area, Volume at 40°N
23 km





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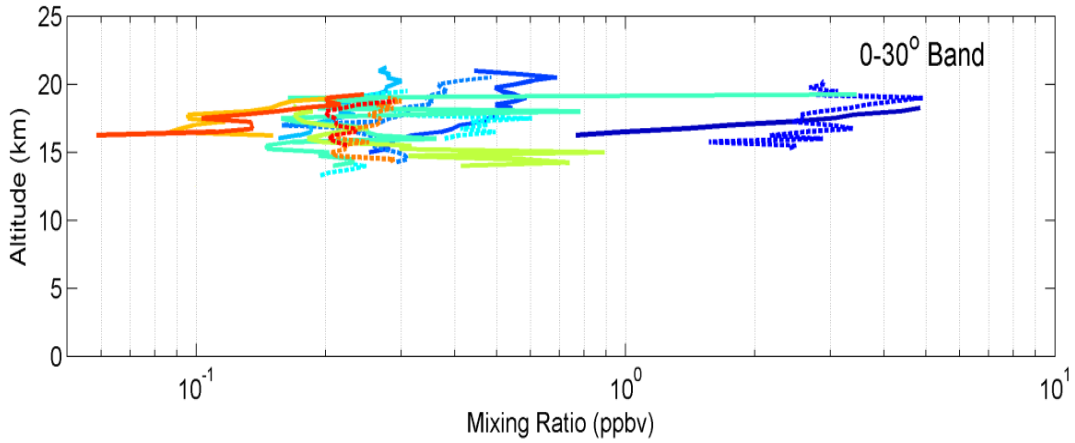
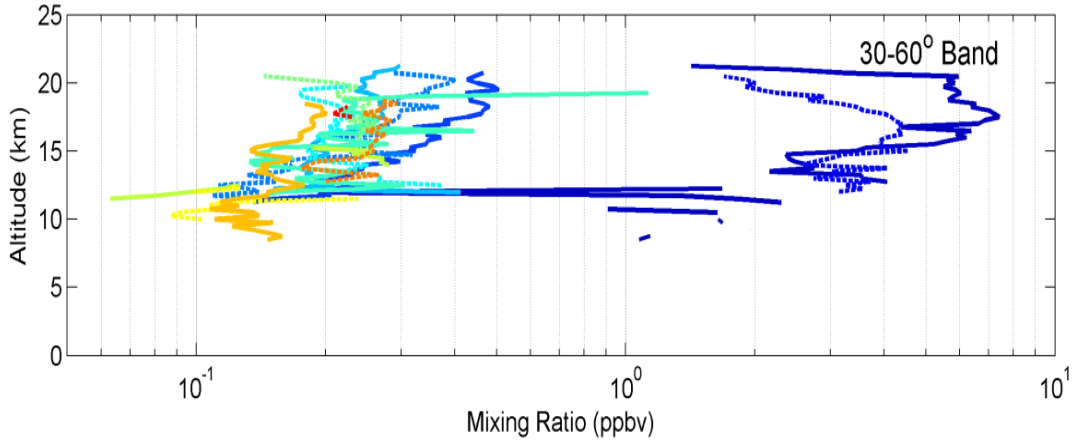
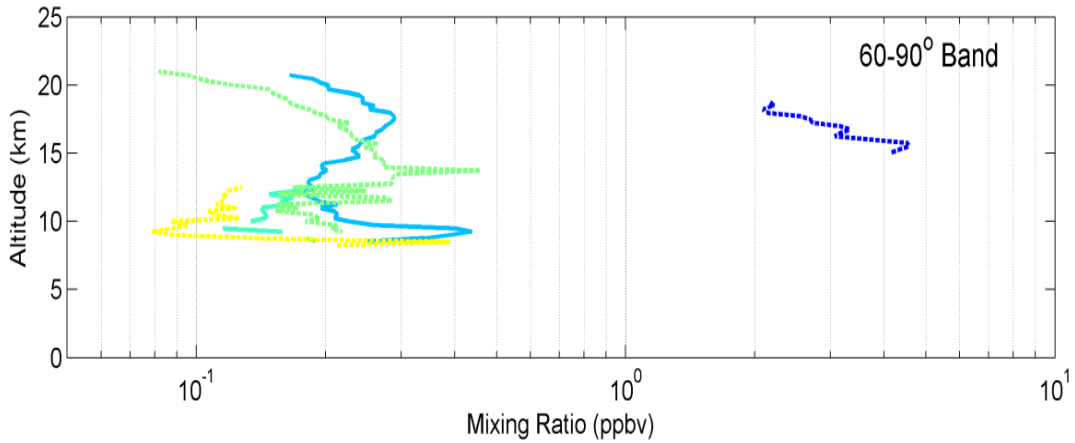
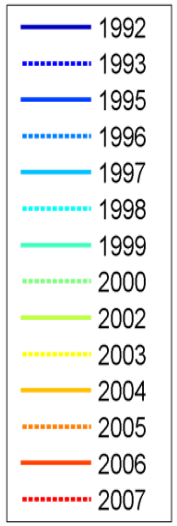
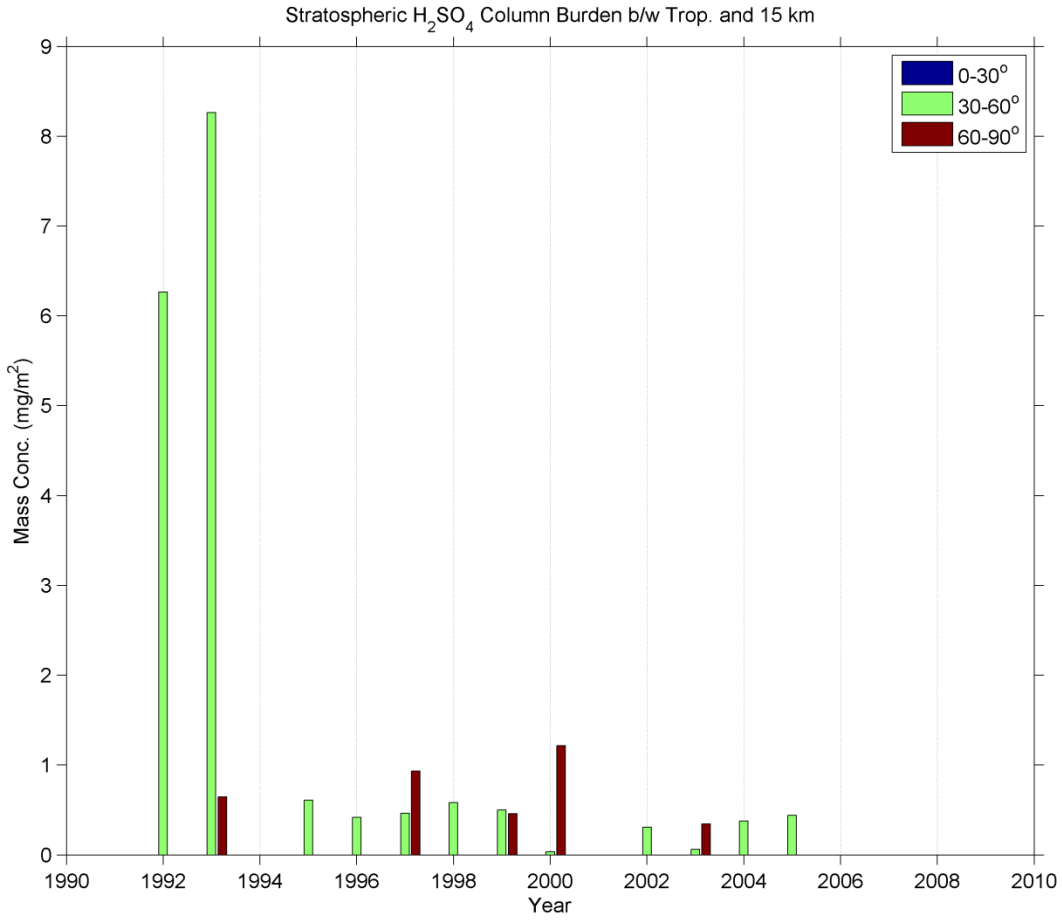
DANIEL FELIX RITCHIE SCHOOL OF ENGINEERING & COMPUTER SCIENCE











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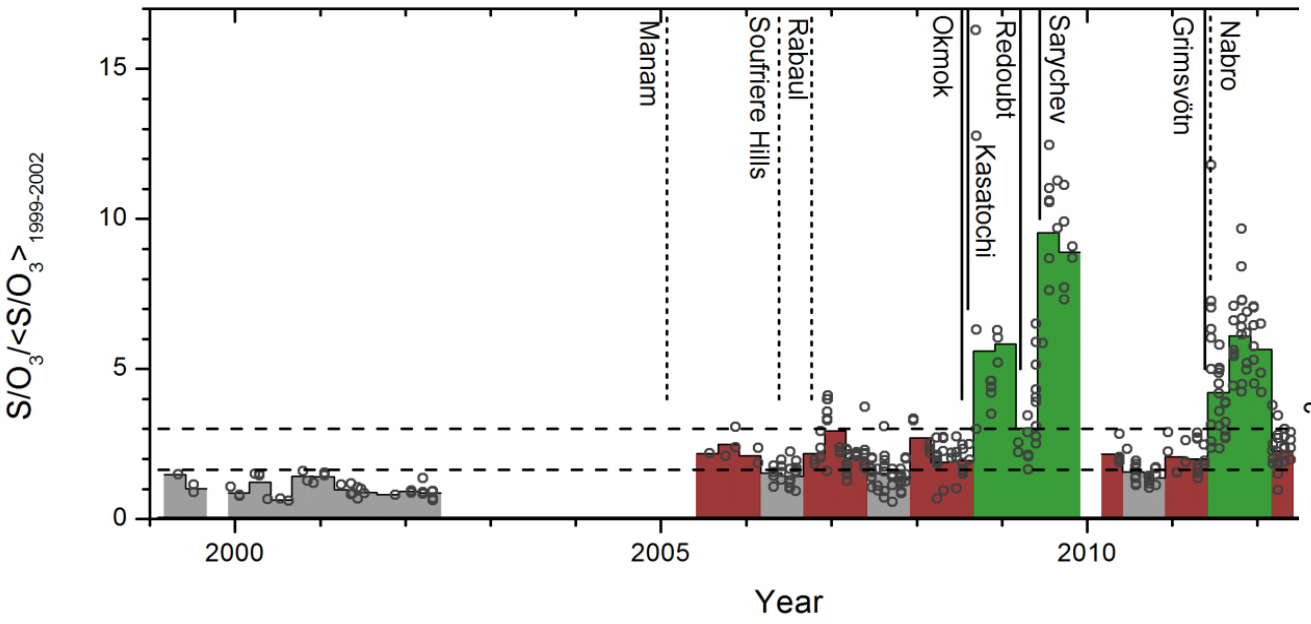
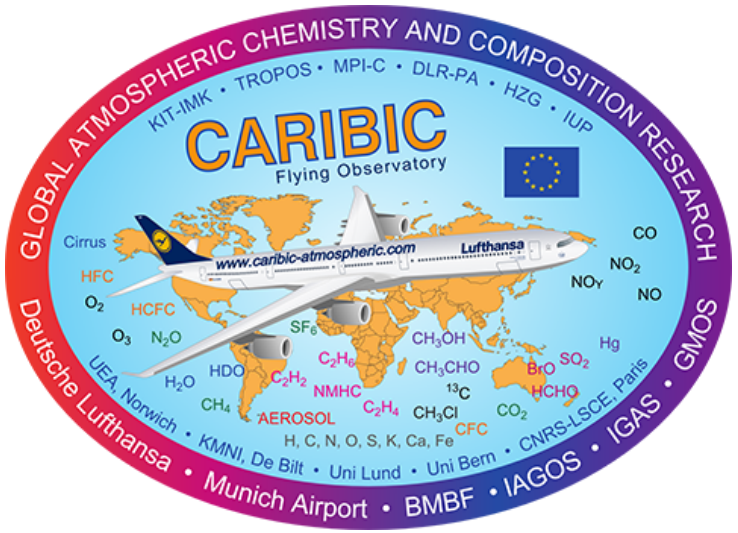
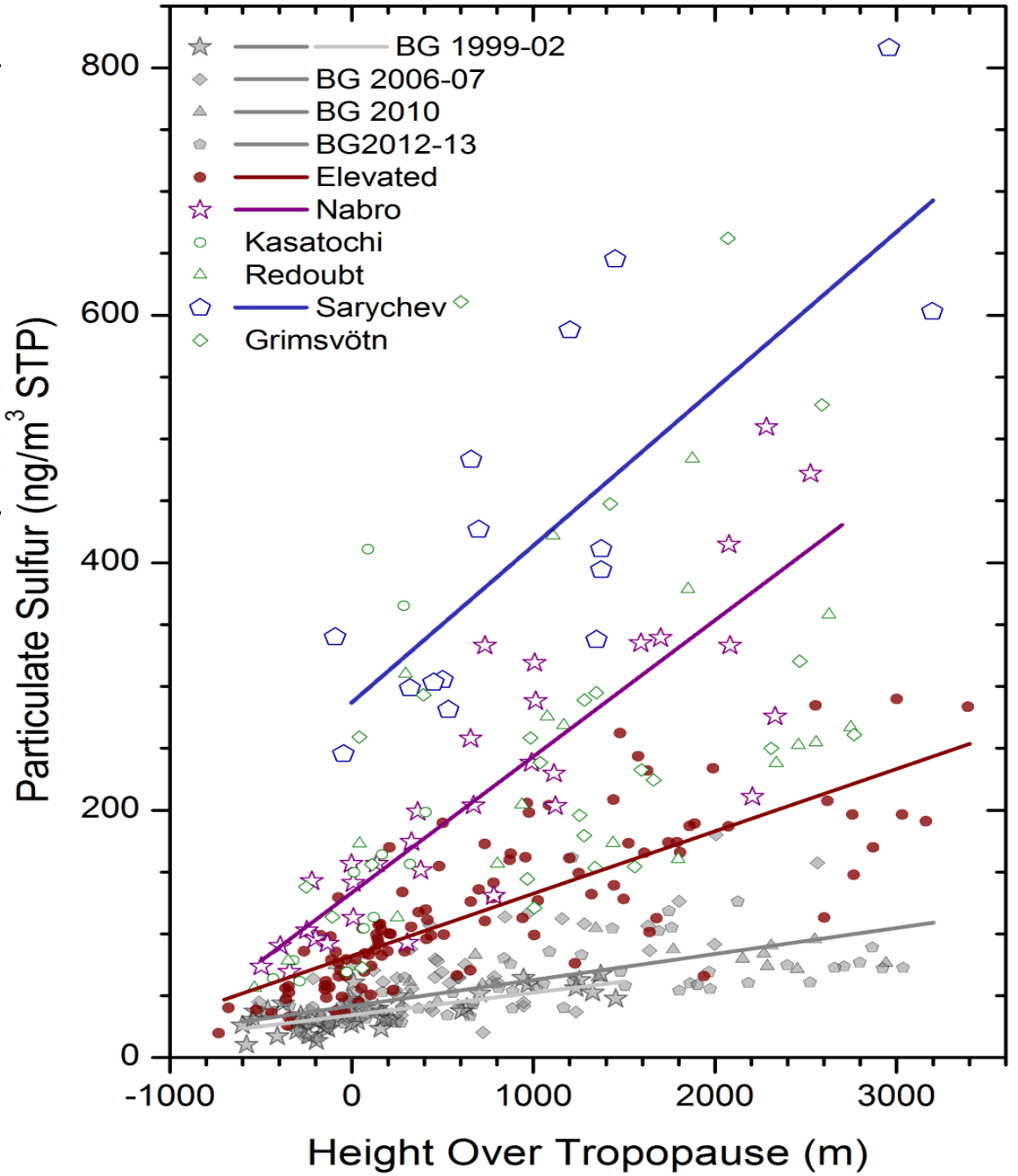
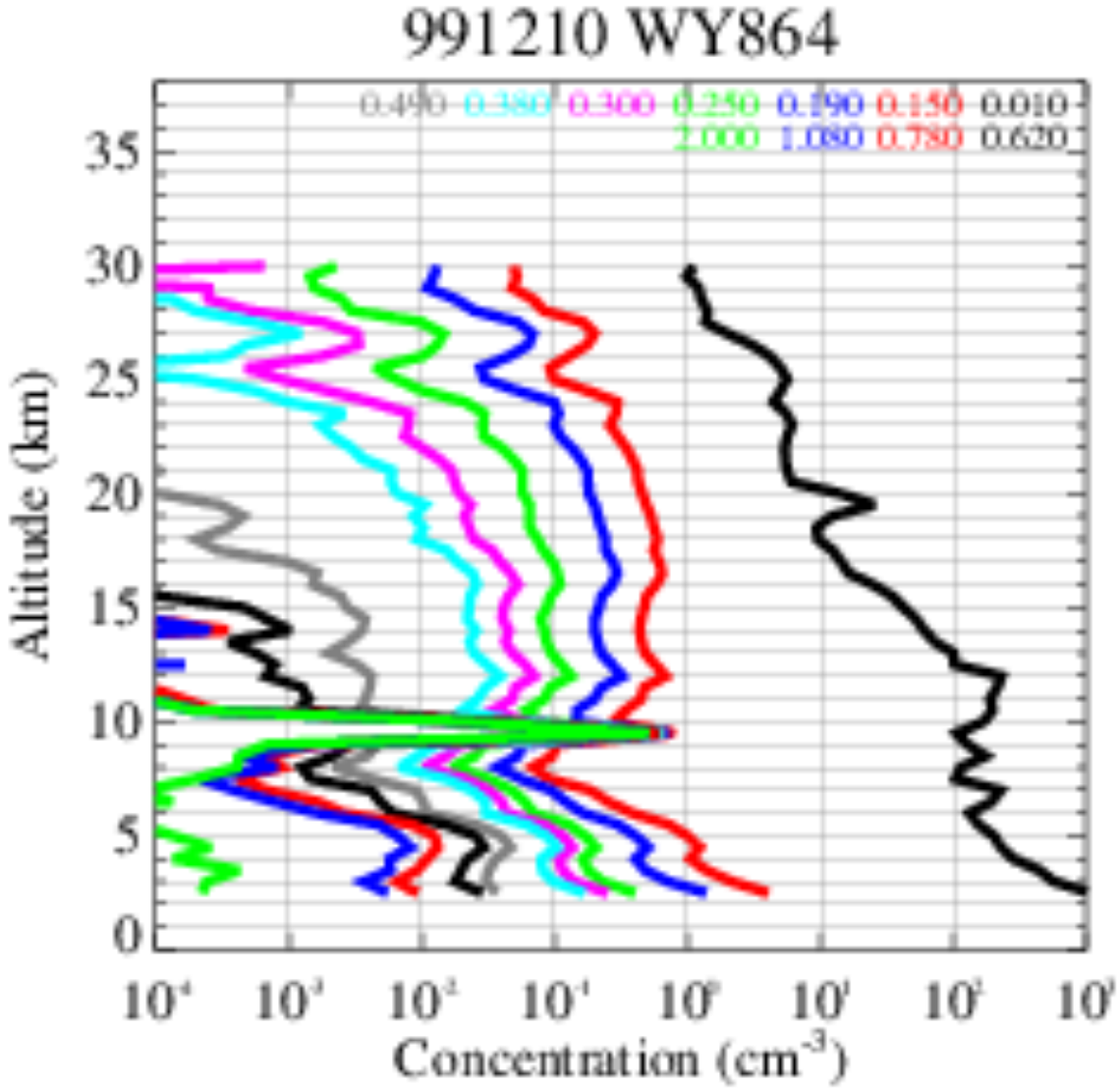
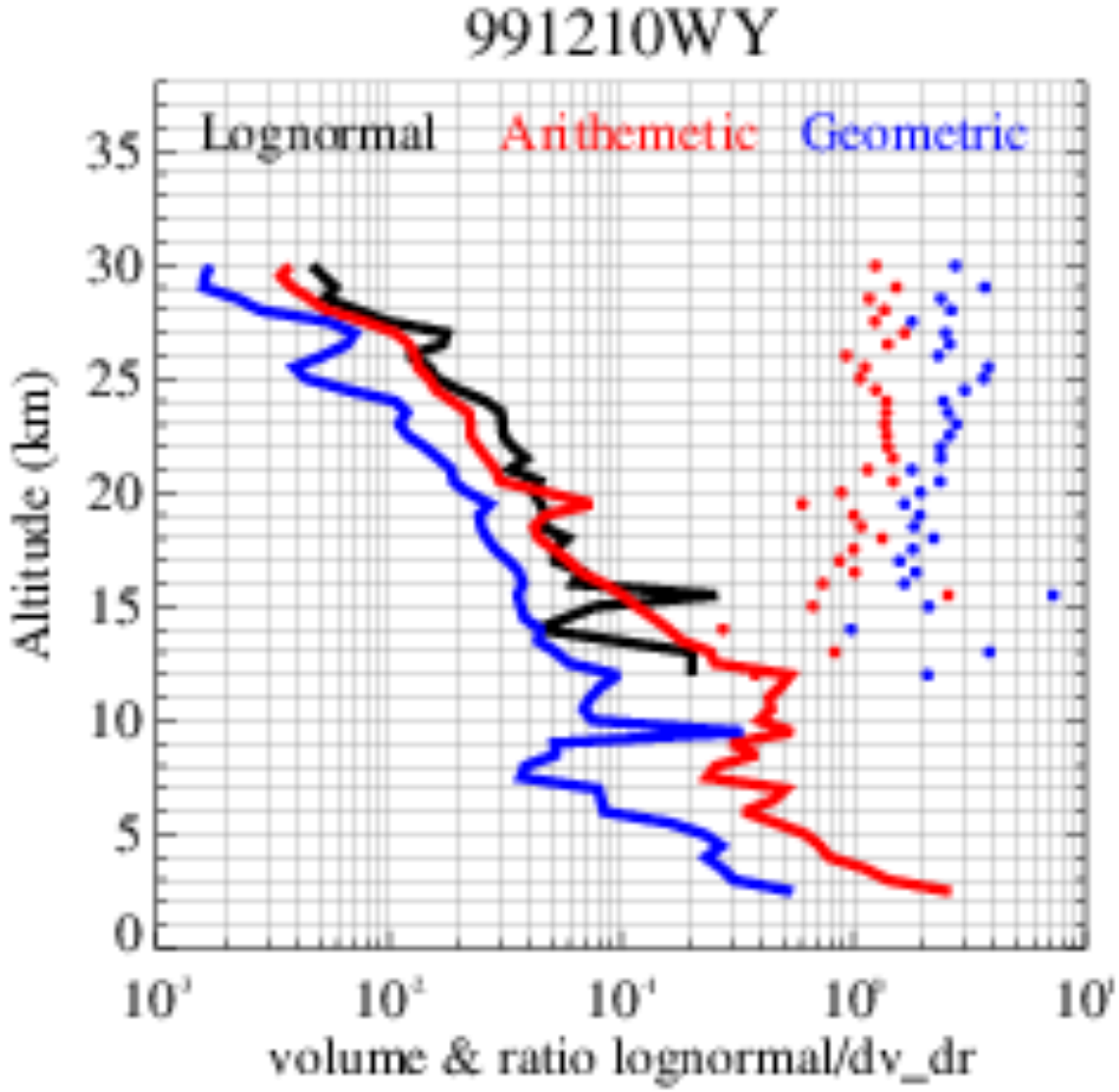

Manam
Soufriere Hills
Rabaul
Okmok
Kasatochi
Redoubt
Sarychev
Grimsvötn
Nabro
15
10
5
0
2000
2005
2010
Year
S/O3/<S/O3>1999-2002


GLOBAL ATMOSPHERIC CHEMISTRY AND COMPOSITION RESEARCH
CARIBIC
Flying Observatory
www.caribic-atmospheric.com
Lufthansa
KIT-IMK • TROPOS • MPI-C • DLR-PA • HZG • IUP
Deutsche Lufthansa • Munich Airport • BMBF • IAGOS • IGAS • GMOS
UEA, Norwich • KNMI, De Bilt • Uni Lund • Uni Bern • CNRS-LSCE, Paris
Cirrus
HFC
HCFC
O3
N2O
H2O
HDO
CH4
SF6
C2H6
C2H2
NMHC
C2H4
AEROSOL
CH3OH
CH3CHO
13C
CH3Cl
CFC
H, C, N, O, S, K, Ca, Fe
CO2
BrO
SO2
HCHO
Hg
CO
NO2
NOY
NO


BG 1999-02
BG 2006-07
BG 2010
BG2012-13
Elevated
Nabro
Kasatochi
Redoubt
Sarychev
Grimsvötn
800
600
400
200
0
-1000
0
1000
2000
3000
Particulate Sulfur (ng/m3 STP)
Height Over Tropopause (m)





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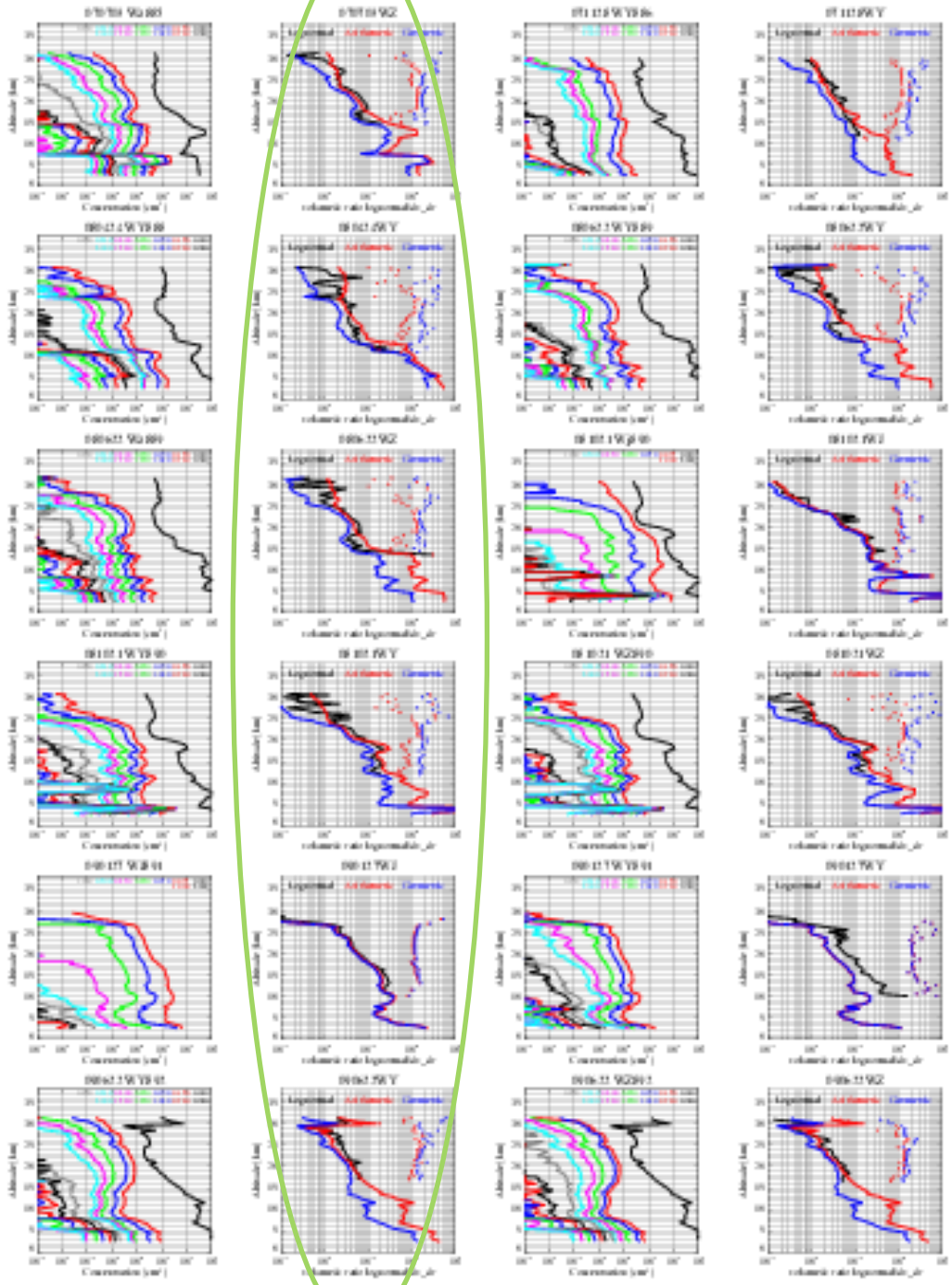
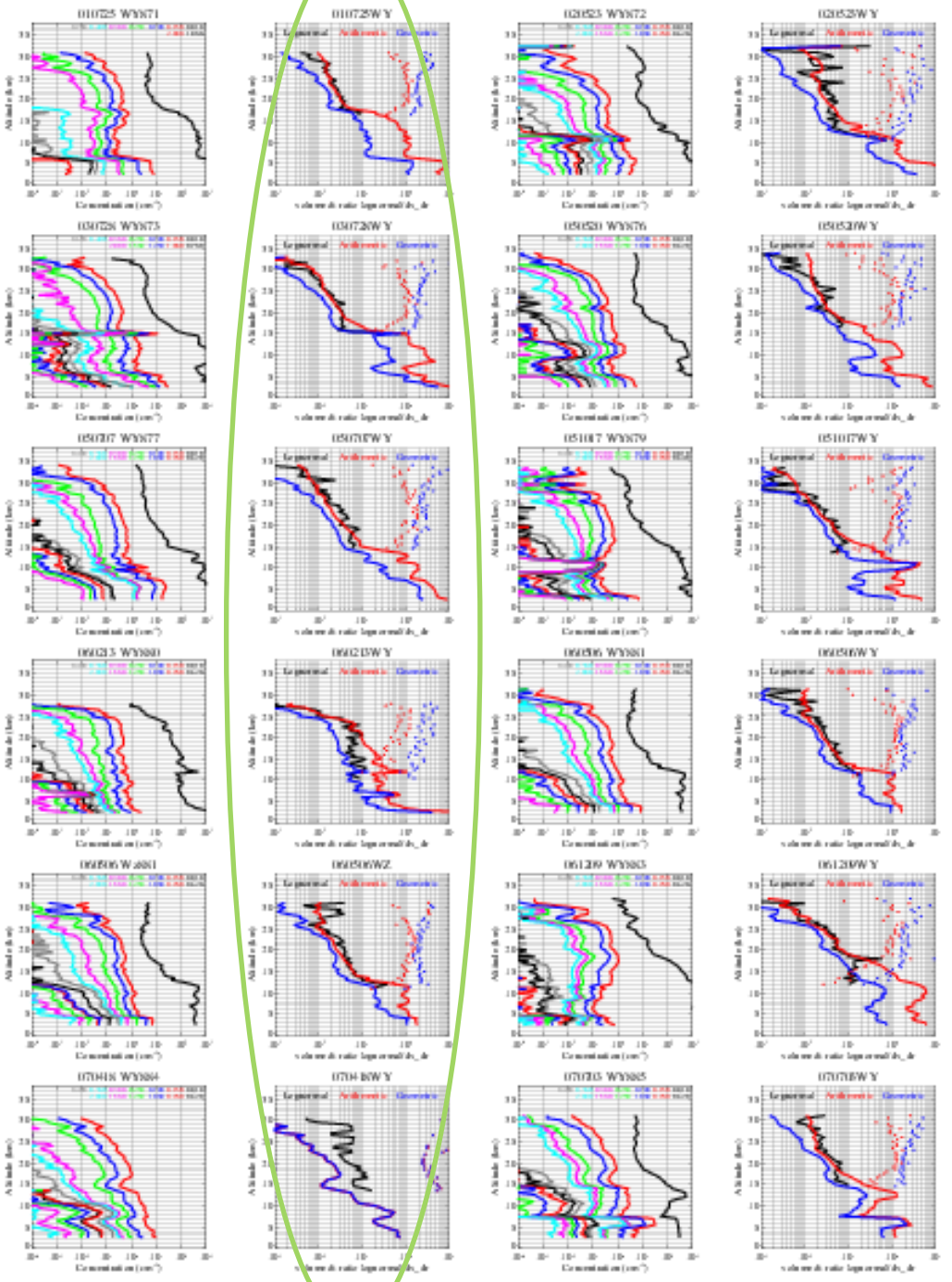
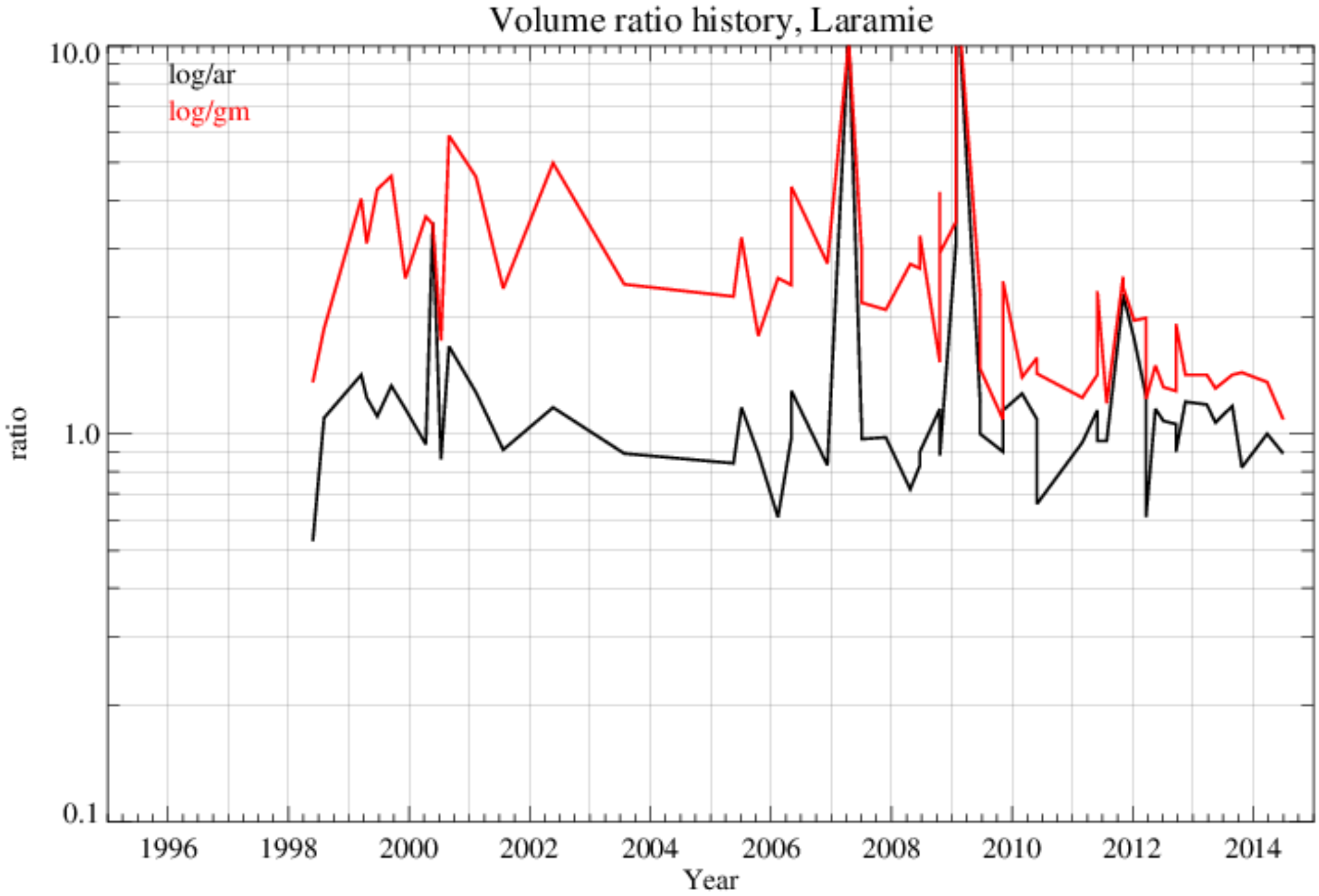
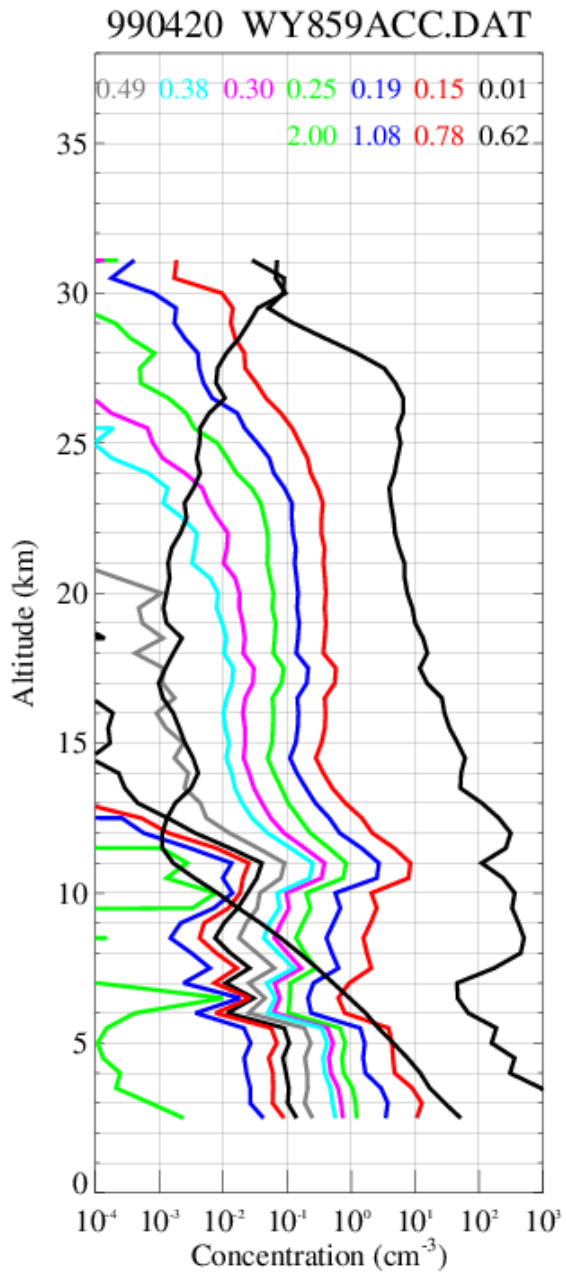
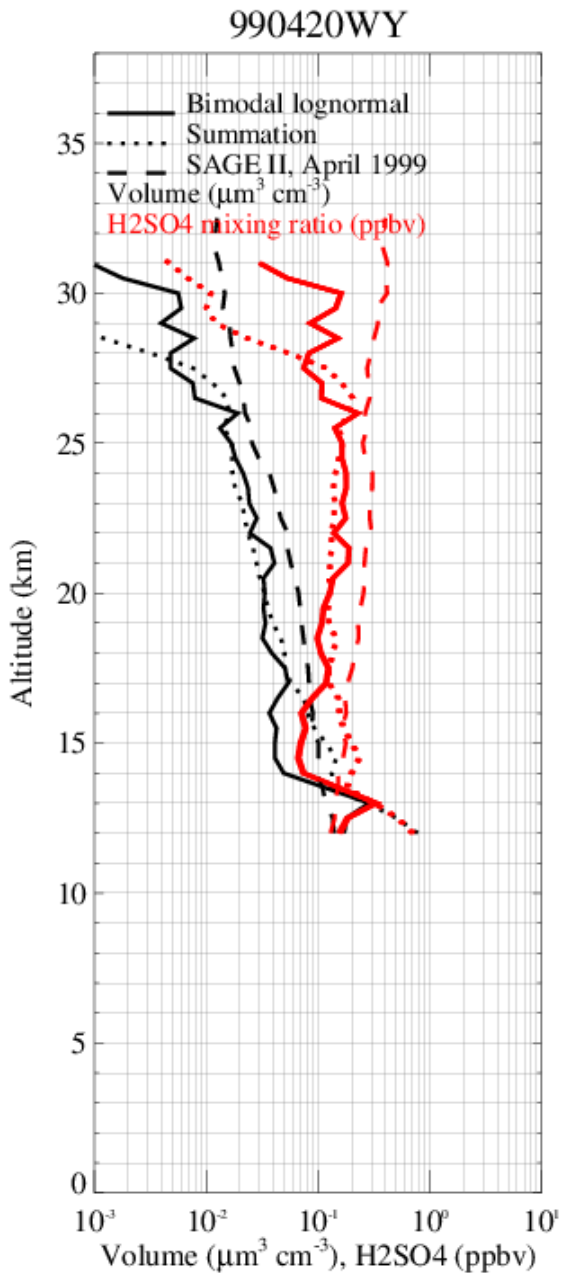
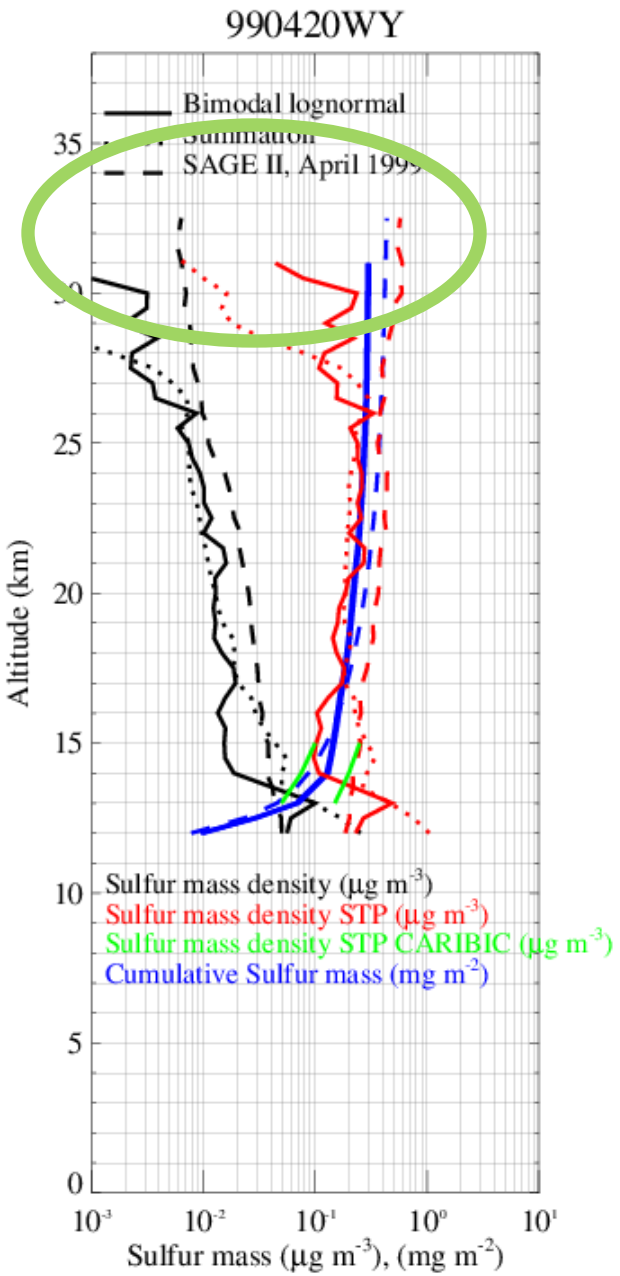


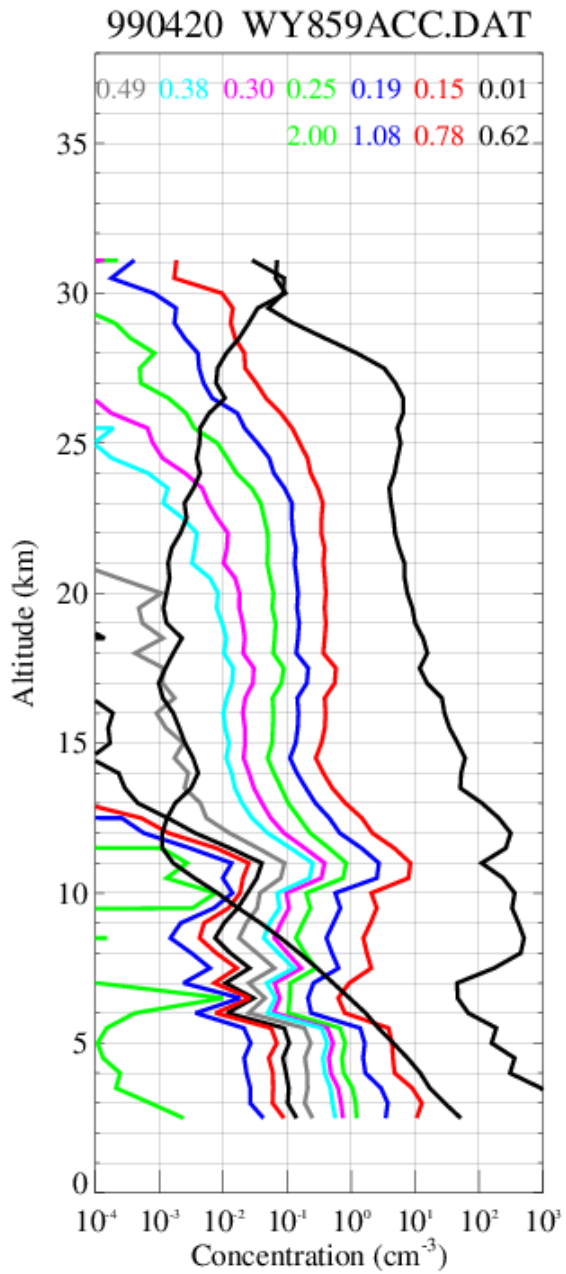


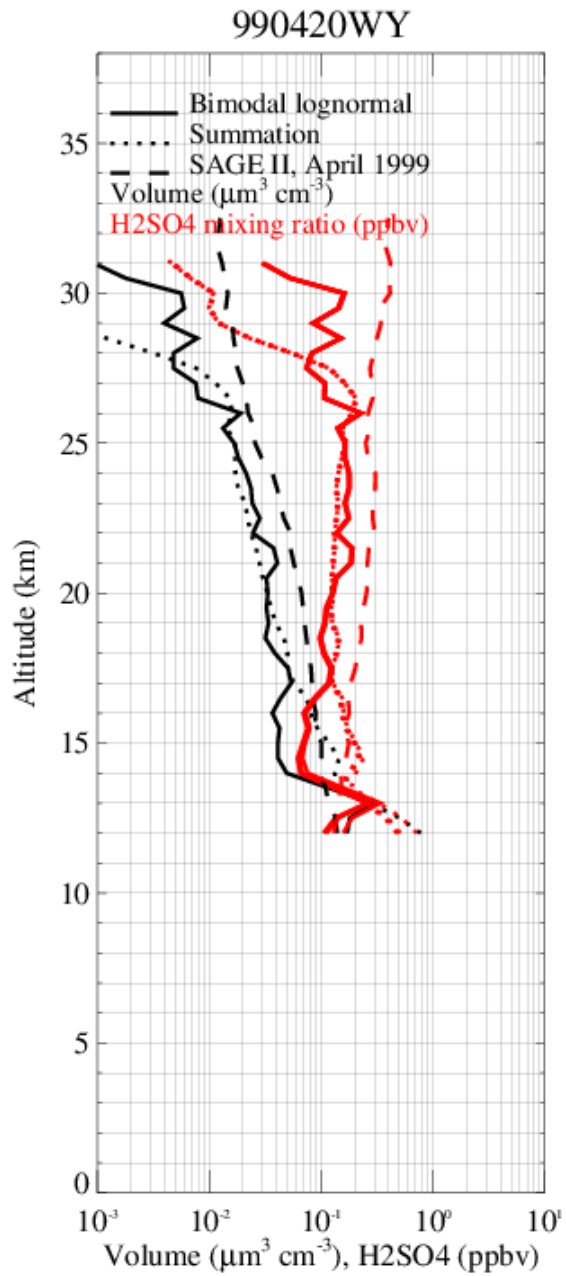

Volume ratio history, Laramie
log/ar
log/gm
10.0
1.0
0.1
ratio
1996
1998
2000
2002
2004
2006
2008
2010
2012
2014
Year

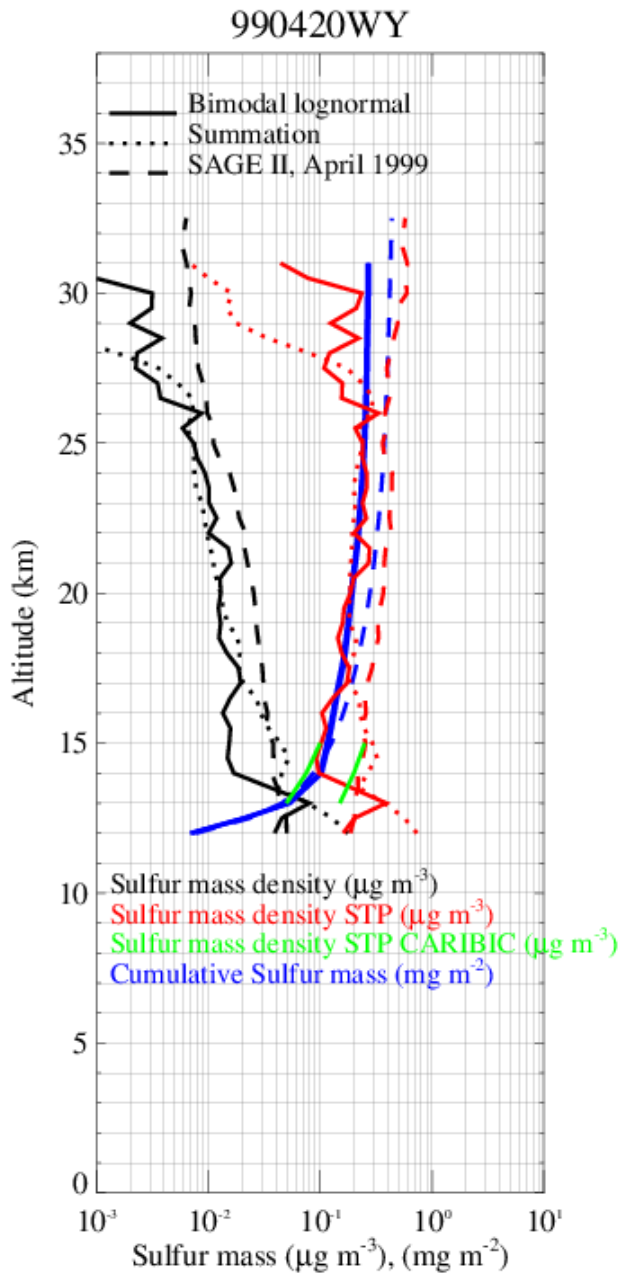

990420 WY859ACC.DAT
0.49 0.38 0.30 0.25 0.19 0.15 0.01
2.00 1.08 0.78 0.62
Altitude (km)
Concentration (cm-3)

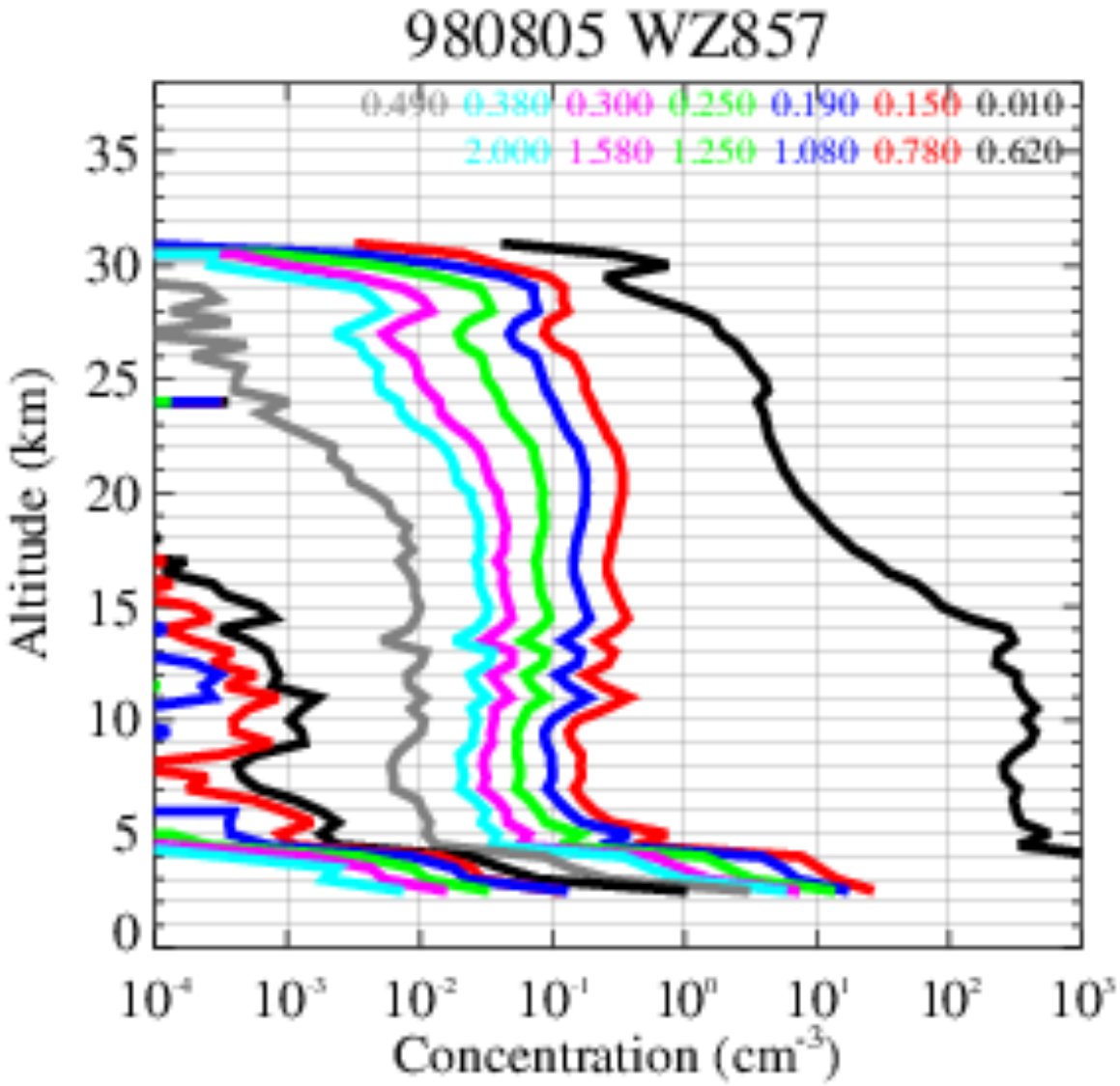

990420WY
Bimodal lognormal
Summation
SAGE II, April 1999
Volume (µm3 cm-3)
H2SO4 mixing ratio (ppbv)
Altitude (km)
Volume (µm3 cm-3), H2SO4 (ppbv)

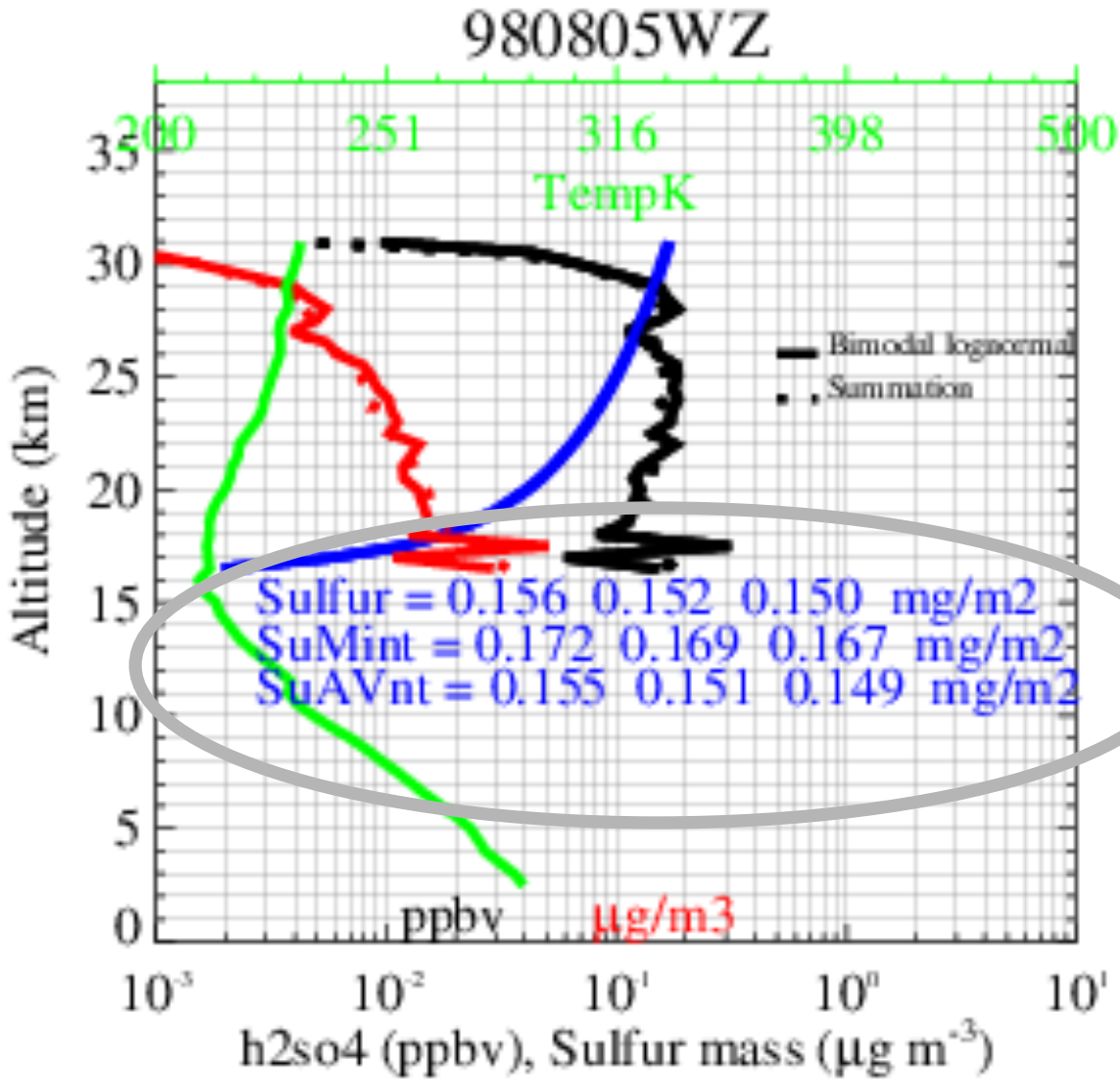

990420WY
Bimodal lognormal
SAGE II, April 1999
Sulfur mass density (µg m-3)
Sulfur mass density STP (µg m-3)
Sulfur mass density STP CARIBIC (µg m-3)
Cumulative Sulfur mass (mg m-2)
Altitude (km)
Sulfur mass (µg m-3), (mg m-2)

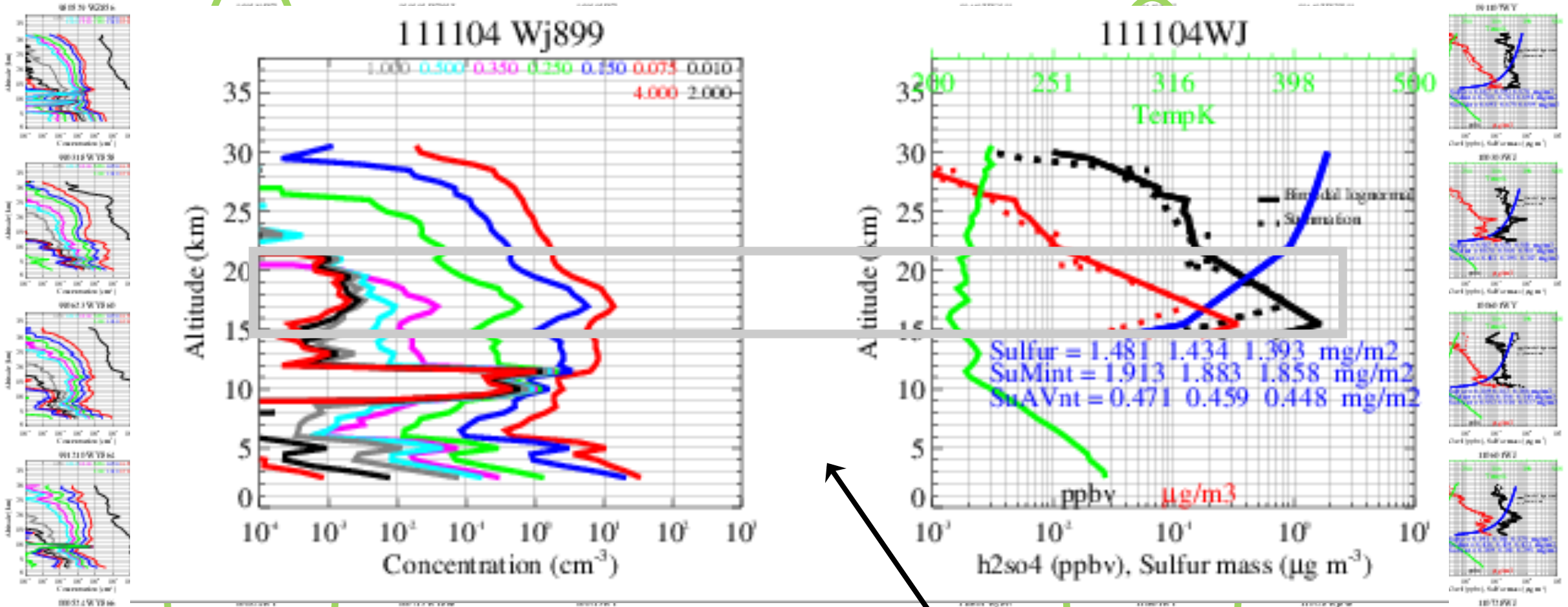

990420 WY859ACC.DAT
0.49 0.38 0.30 0.25 0.19 0.15 0.01
2.00 1.08 0.78 0.62
Altitude (km)
Concentration (cm-3)

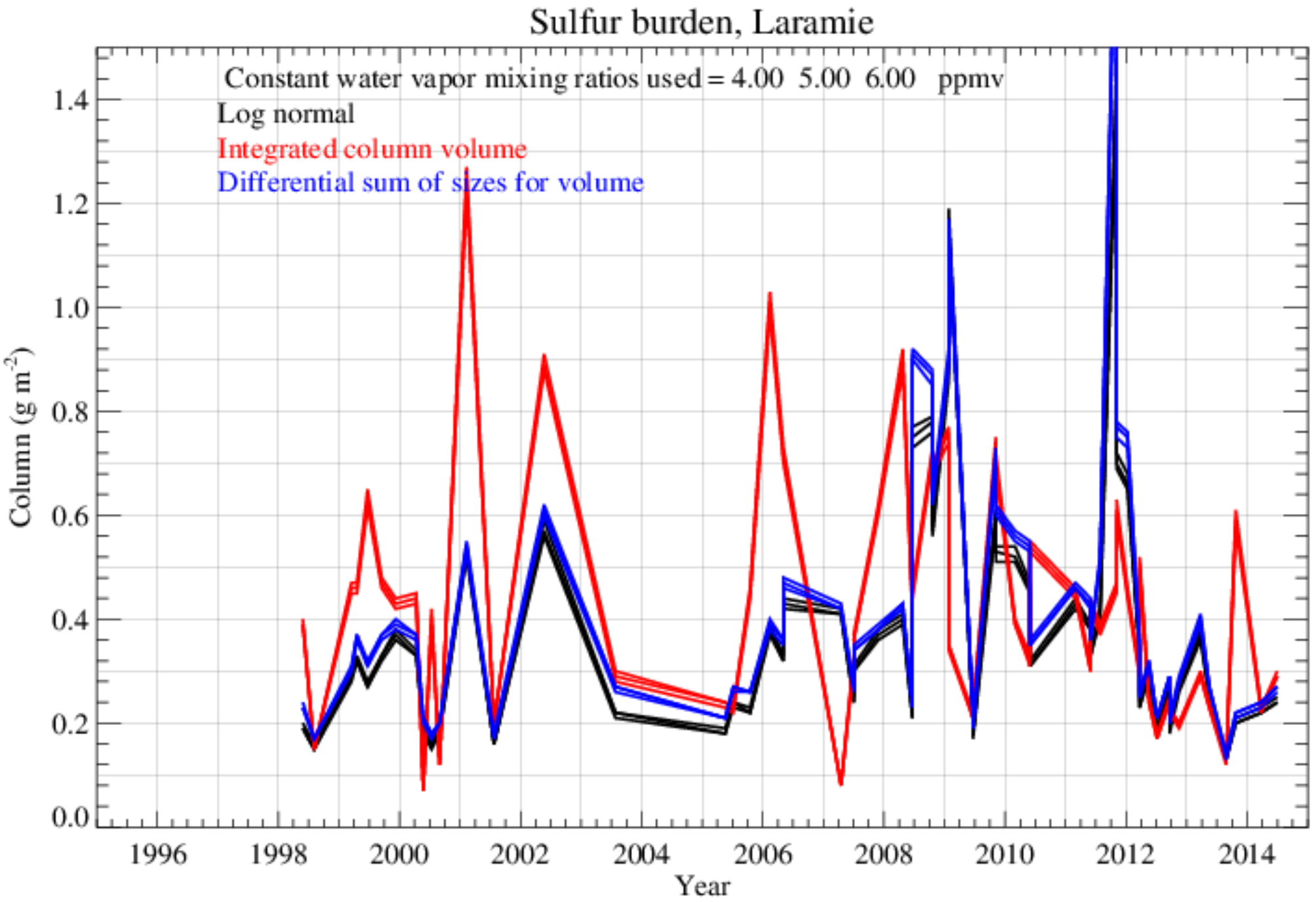

990420WY
Bimodal lognormal
Summation
SAGE II, April 1999
Volume (µm3 cm-3)
H2SO4 mixing ratio (ppbv)
Altitude (km)
Volume (µm3 cm-3), H2SO4 (ppbv)

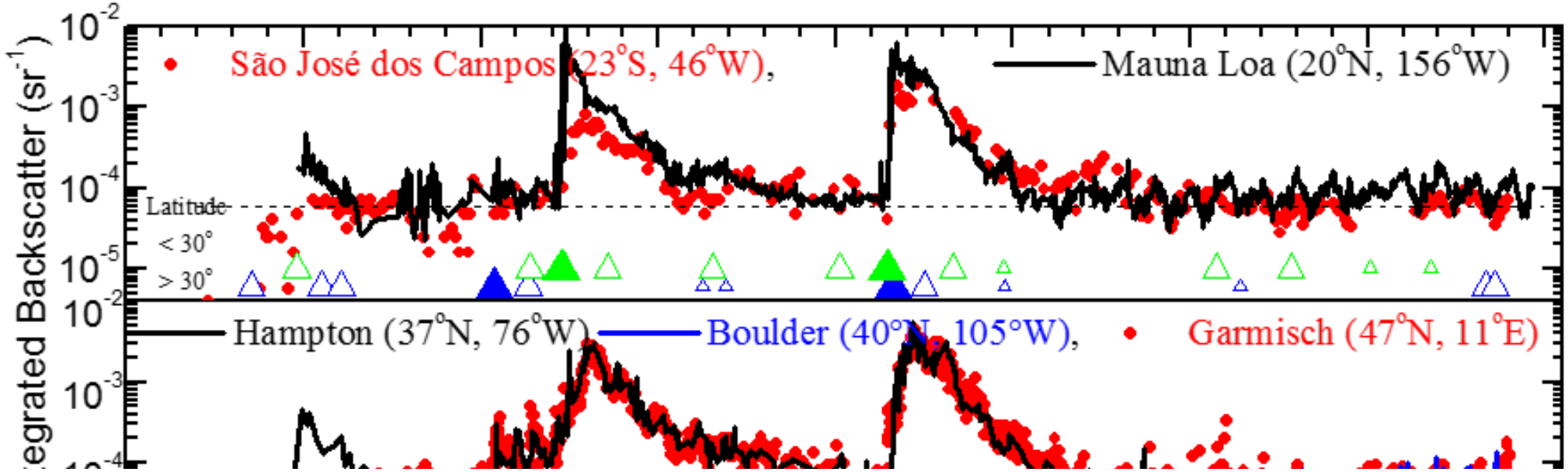

990420WY
Bimodal lognormal
Summation
SAGE II, April 1999
Sulfur mass density (µg m-3)
Sulfur mass density STP (µg m-3)
Sulfur mass density STP CARIBIC (µg m-3)
Cumulative Sulfur mass (mg m-2)
Altitude (km)
Sulfur mass (µg m-3), (mg m-2)


980805 WZ857
0.490 0.380 0.300 0.250 0.190 0.150 0.010
2.000 1.580 1.250 1.080 0.780 0.620
Altitude (km)
Concentration (cm-3)

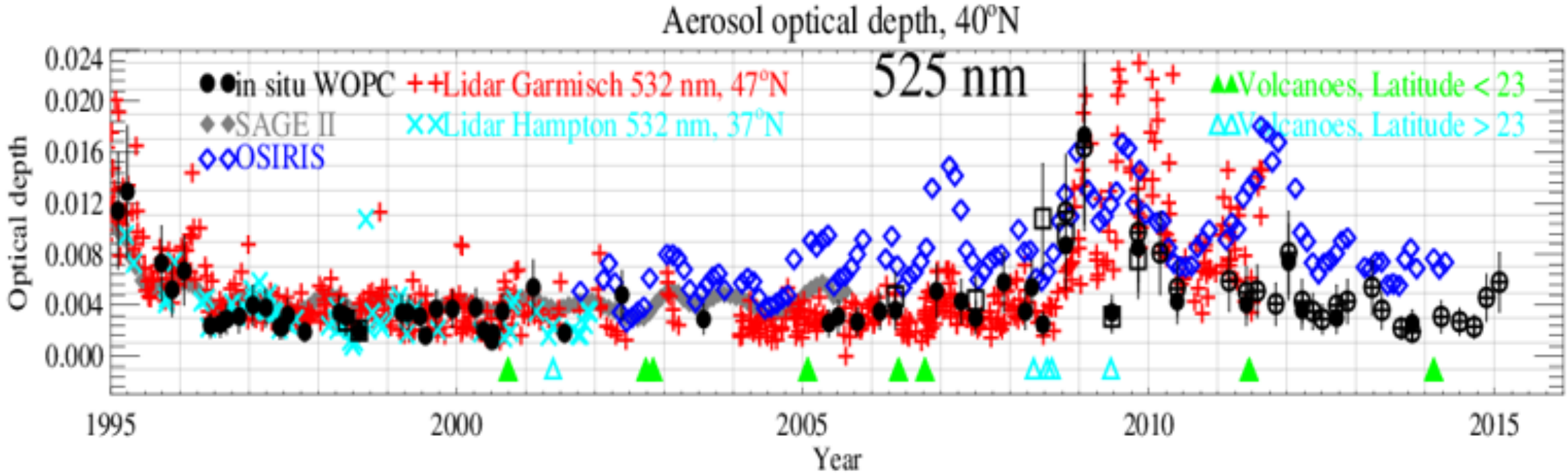

980805WZ
200 251 316 398 500
TempK
Bimodal logncemal
Summation
Sulfur = 0.156 0.152 0.150 mg/m2
SuMint = 0.172 0.169 0.167 mg/m2
SuAVnt = 0.155 0.151 0.149 mg/m2
ppbv
µg/m3
Altitude (km)
h2so4 (ppbv), Sulfur mass (µg m-3)

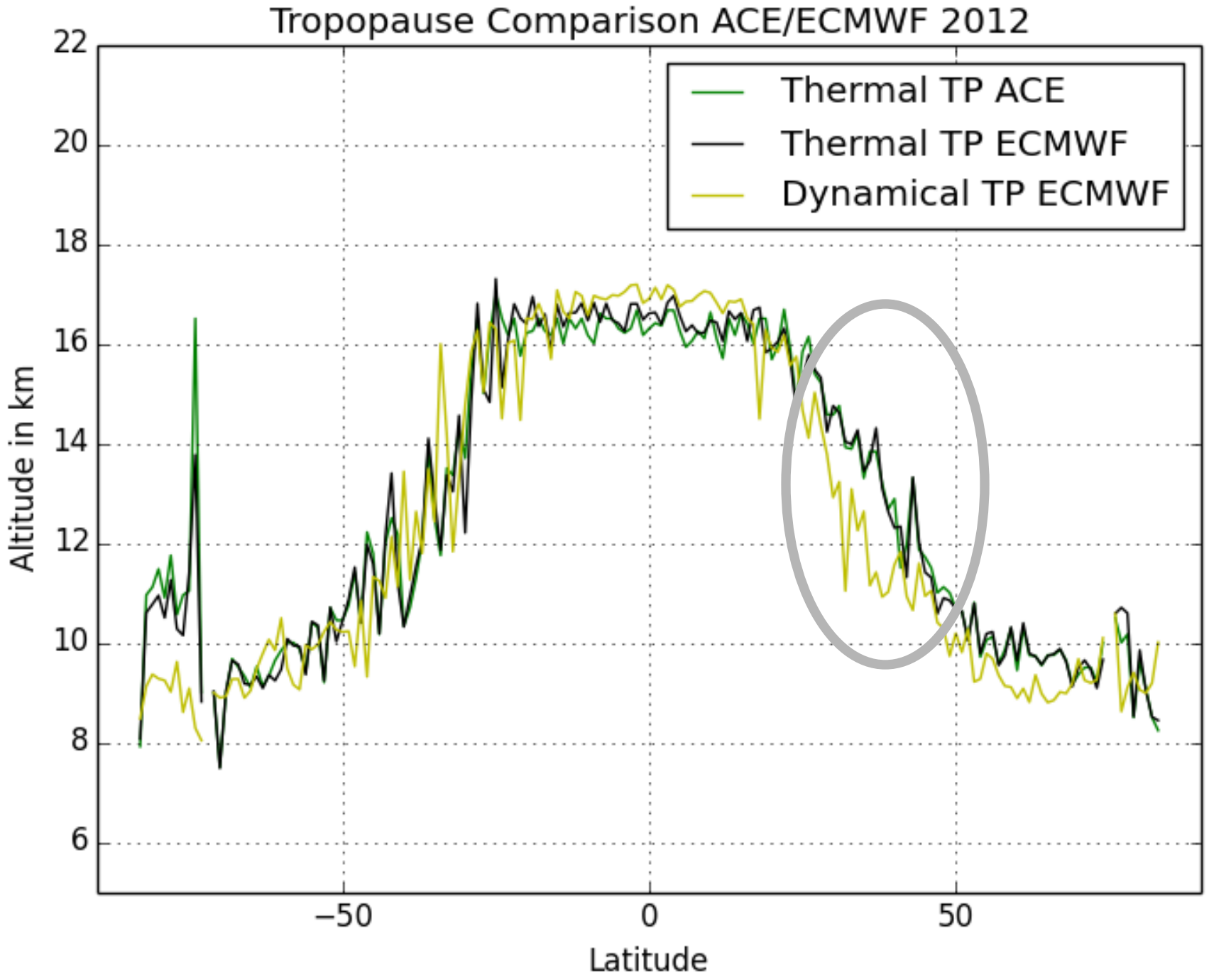

111104 Wj899
1.000 0.500 0.350 0.250 0.150 0.075 0.010
4.000 2.000
Altitude (km)
Concentration (cm-3)
111104WJ
200 251 316 398 500
TempK
Sulfur = 1.481 1.434 1.393 mg/m2
SuMint = 1.913 1.883 1.858 mg/m2
SuAVnt = 0.471 0.459 0.448 mg/m2
ppbv
µg/m3
h2so4 (ppbv), Sulfur mass (µg m-3)


Sulfur burden, Laramie
Constant water vapor mixing ratios used = 4.00 5.00 6.00 ppmv
Log normal
Integrated column volume
Differential sum of sizes for volume
Column (g m-2)
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1996
1998
2000
2002
2004
2006
2008
2010
2012
2014
Year



Kremser et al., *Rev Geophys*, 2016





Integrations to be done above dynamical tropopause, ~ 380 K

Conclusions / Plan

- **Provide a temporal history the gas and particle phase sulfur burden in:**
  - **30 degree latitude bands**
  - **Monthly to seasonal averages**
  - **Use measurements to create a global temporal / latitudinal burden**
- **Time period**
  - **2002-2012 – Envisat period primarily**
    - **OCS (ACE, MIPAS, Lauder)**
    - **SO2 (MIPAS)**
    - **Particles – SAGE II, OSIRIS, Lidars, In situ**
  - **Extend the time period backwards to 1984 with particle measurements and some relevant gas phase component.**
- **Call out to the many funding agencies involved in supporting the work on which this is built**

Future of In Situ Measurements

Lars Kalnajs