Model calculations of the contribution of SO_2 to the stratospheric aerosol layer

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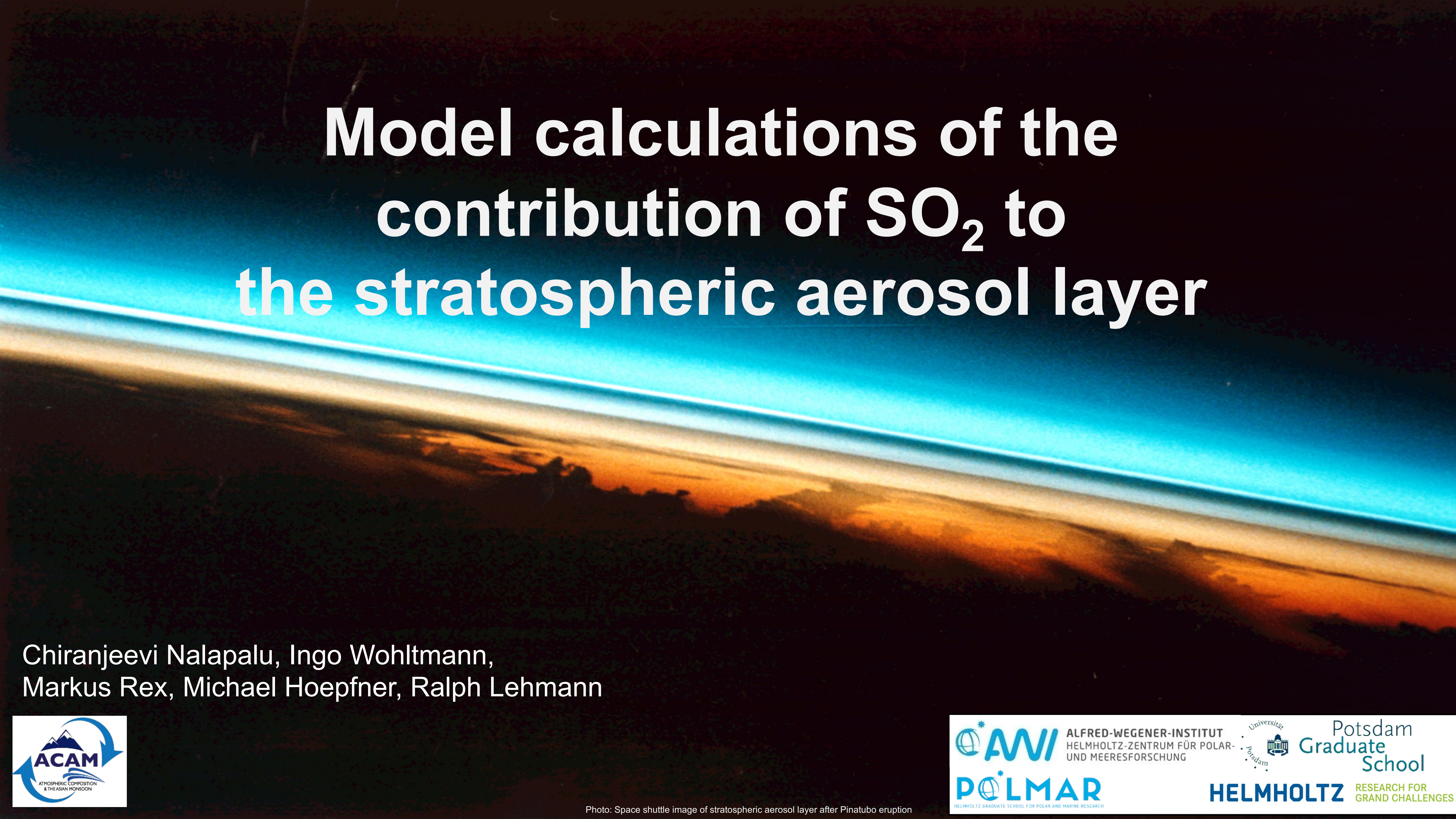

Photo: Space shuttle image of stratospheric aerosol layer after Pinatubo eruption

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Motivation

Stratospheric aerosol layer is important for

- Radiative balance of earth and climate change
- Stratospheric chemistry (“ozone hole”)
- Geo-Engineering

Many processes of the stratospheric aerosol layer are not well known

- What is the contribution of tropospheric species like SO_2 to stratospheric aerosol layer? Sources? Processes? Sensitivities?

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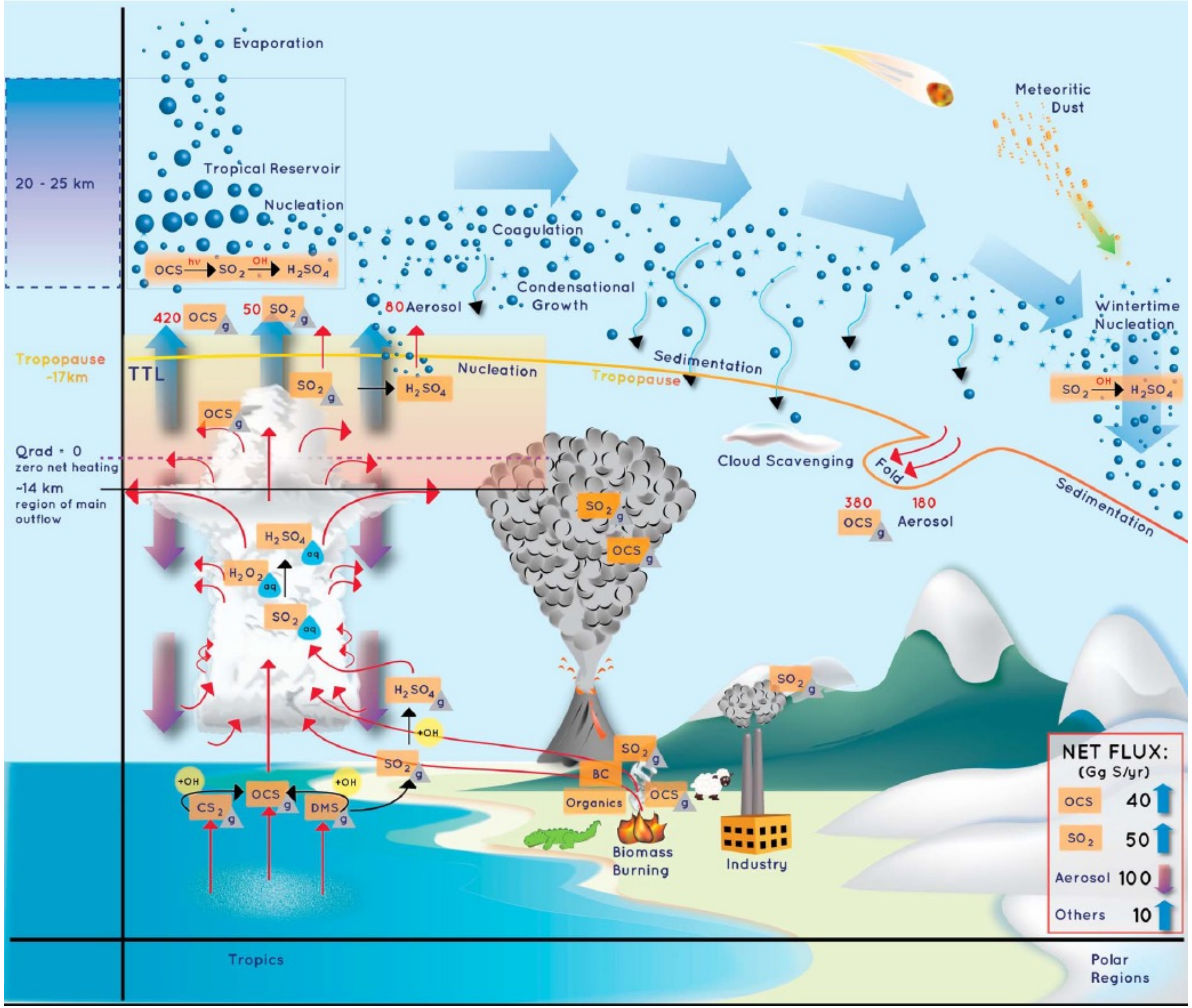

Kremser et al., Rev. Geophys., 2015, doi:10.1002/2015RG000511

Approach

- Examine chemistry of SO_2 and its transport to the stratosphere
- Chemical and microphysical “box” model on moving air parcel trajectories ending in the stratosphere
- Numerous sensitivity runs to assess range of uncertainty
- Focus on “background” values under volcanically quiescent conditions
- Don’t model the processes in the stratospheric aerosol layer, we are interested in the source of the SO_2 in the layer
- Initialization at start of trajectories from existing model data at top of boundary layer, boundary layer processes and SO_2 sources at surface are not our task

Our existing tools

- **Trajectory model** of the ATLAS Chemistry and Transport Model of our working group.
- **Convection model**: Trajectory model includes “statistical“ convection model on individual trajectories
- **Simplified chemical and microphysical model** focussing on the relevant SO_2 and sulfur chemistry and cloud interactions
- **CAMS data for initialization** of chemistry model at top of boundary layer

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Model

- Backward trajectories (≤ 4 months) starting at 400 K in [30N, 30S], ending at 80% ps (ps = surface pressure)
- Wind fields and cloud water content ← ERA 5 and ERA Interim
- Convection: trajectories are displaced vertically with a certain probability
- Chemistry is run forward in time on the trajectories

Reactions in our Chemical Box Model

$$SO_2 + OH + M \rightarrow \text{Products}$$

$$DMS + OH \rightarrow \ldots \rightarrow SO_2 + \text{Products}$$

Gas Phase Reactions

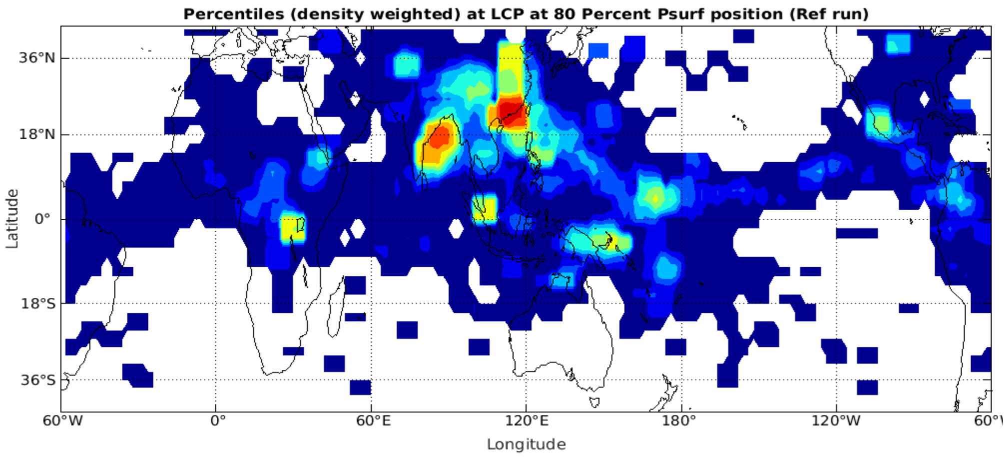
$$SO_2 \cdot H_2O + H_2O_2 \rightarrow \text{Products}$$

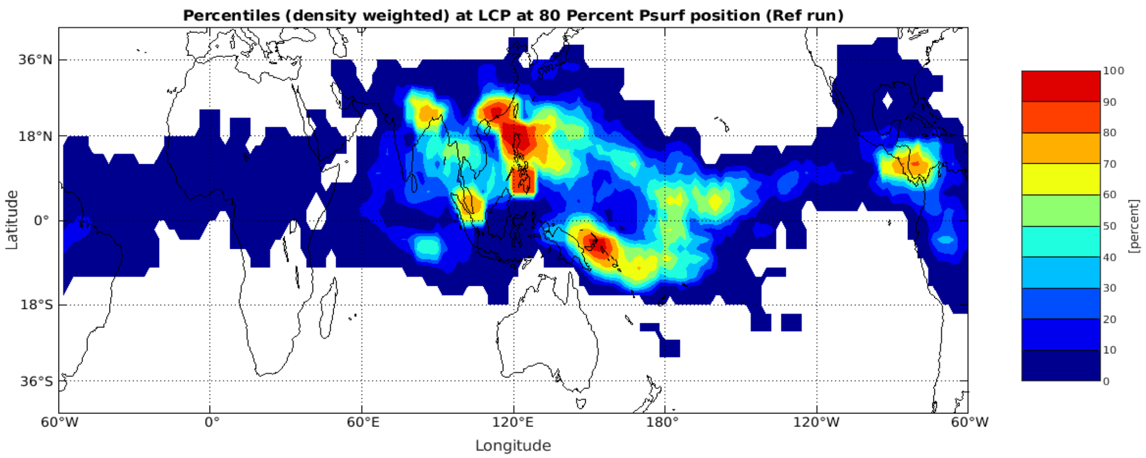
$$S(IV) + O_3 \rightarrow \text{Products}$$

Liquid Phase Reactions

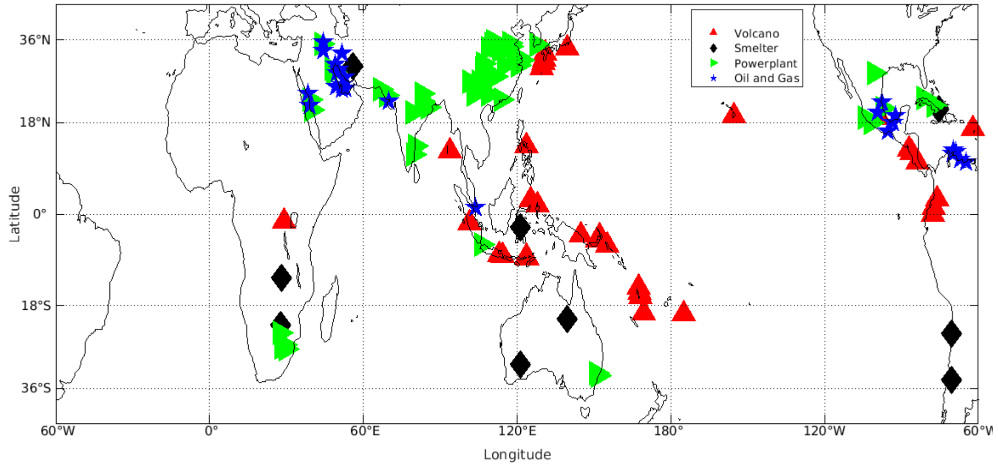
What are the main sources of SO2 in the troposphere?

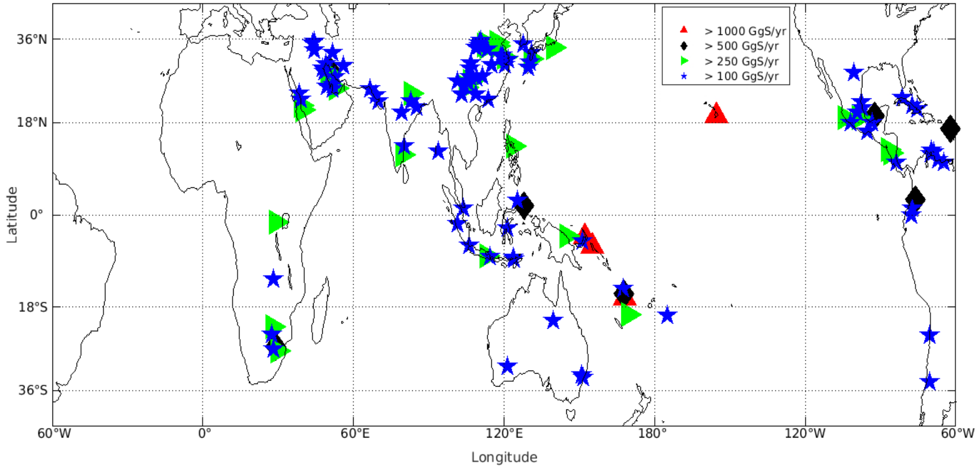
ERA5





ERA Interim





Fioletov et al., Atmos. Chem. Phys., 2016,
doi:10.5194/acp-16-11497-2016

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How much SO2 is transported to the Stratosphere?

No.	Convection	Year	ERA5/ERA_Interim	Z/Zeta	Updraft_Flag	Massflux(Gg/yr)
1	On	2009	ERA5	Zeta	RandConvAreaFrac	3.14
2	On	2009	ERA_Interim	Zeta	RandConvAreaFrac	9.76

Mass flux

Rollins et al. 2017 In Situ -> 3.6 Gg/Yr
Sheng et al. 2015 SOCAL-AERv1 Model -> 50 Gg/Yr

No.	Convection	Year	ERA5/ERA_Interim	Z/Zeta	Updraft_Flag	Mixing Ratio (pptv)
1	On	2009	ERA5	Zeta	RandConvAreaFrac	13.60
2	On	2009	ERA_Interim	Zeta	RandConvAreaFrac	42.75

Mixing ratio

Rollins et al. 2017,2018 In Situ -> 5-10 pptv
Höpfner et al. 2015 MIPAS Satellite -> 24 pptv
Doeringert et al. 2012 ACE-FTS Satellite -> 5-10 pptv
Feinberg et al. 2019 SOCOL-AERv1,v2 Model -> 20-30 pptv

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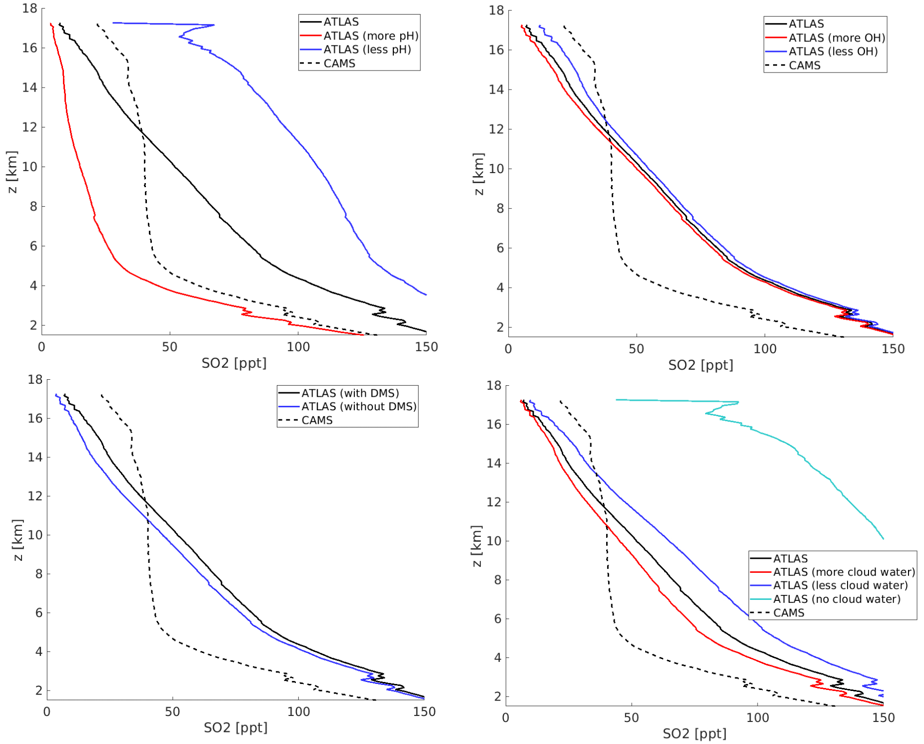
Sensitivities

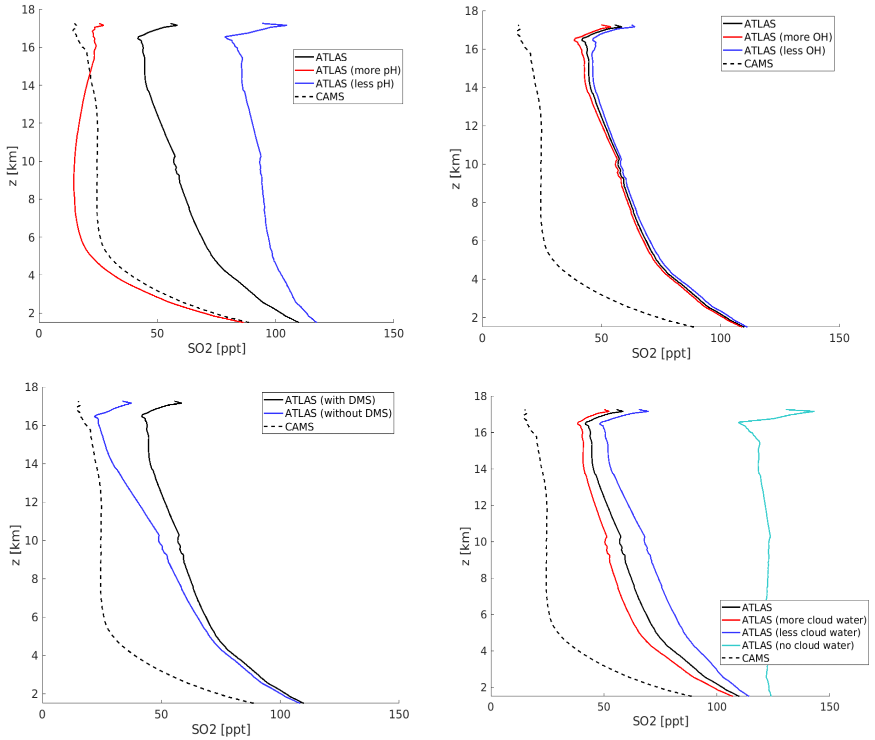
ERA5

	Massflux_LCP	Percentage change
REF	3.14	0.00
No DMS	1.73	-36.70
More Water	2.81	-12.44
Less Water	3.94	28.94
No Water	14.24	458.41
More H2O2	3.04	-3.65
Less H2O2	3.26	4.11
More OH	2.33	-21.78
Less OH	5.10	46.90
More pH	1.56	-57.03
Less pH	9.43	281.97
No NO3	3.18	1.06

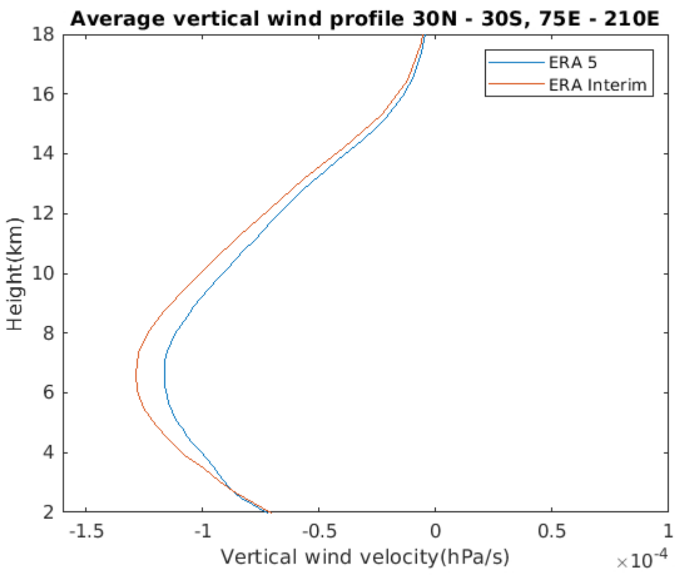


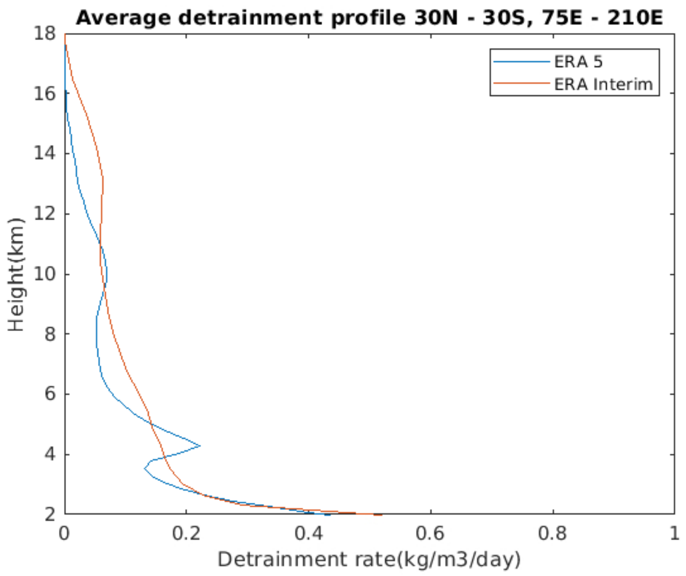
ERA Interim

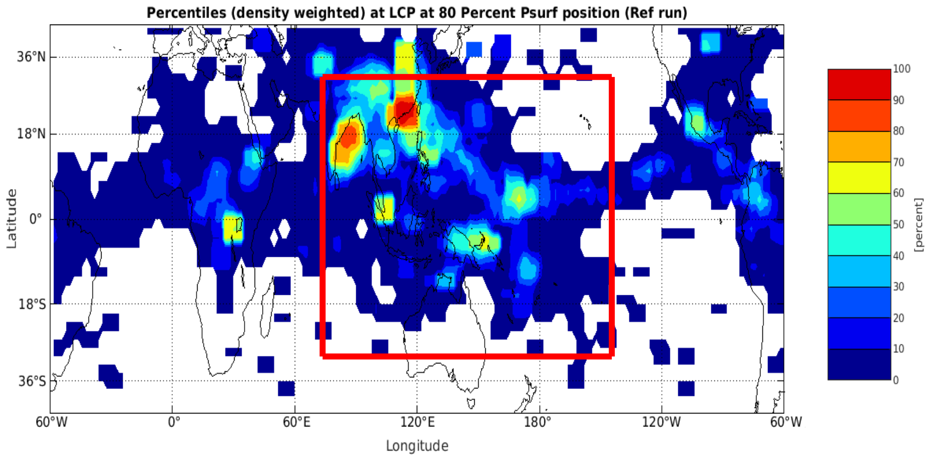
	Massflux_LCP	Percentage change
REF	9.76	0.00
No DMS	4.08	-58.20
More Water	9.25	-5.23
Less Water	10.81	10.76
No Water	23.24	138.11
More H2O2	9.57	-1.95
Less H2O2	9.97	2.15
More OH	8.83	-9.53
Less OH	11.91	22.03
More pH	6.65	-31.86
Less pH	17.26	76.84
No NO3	9.79	0.31



Process understanding:
Updraft and Detrainment rates







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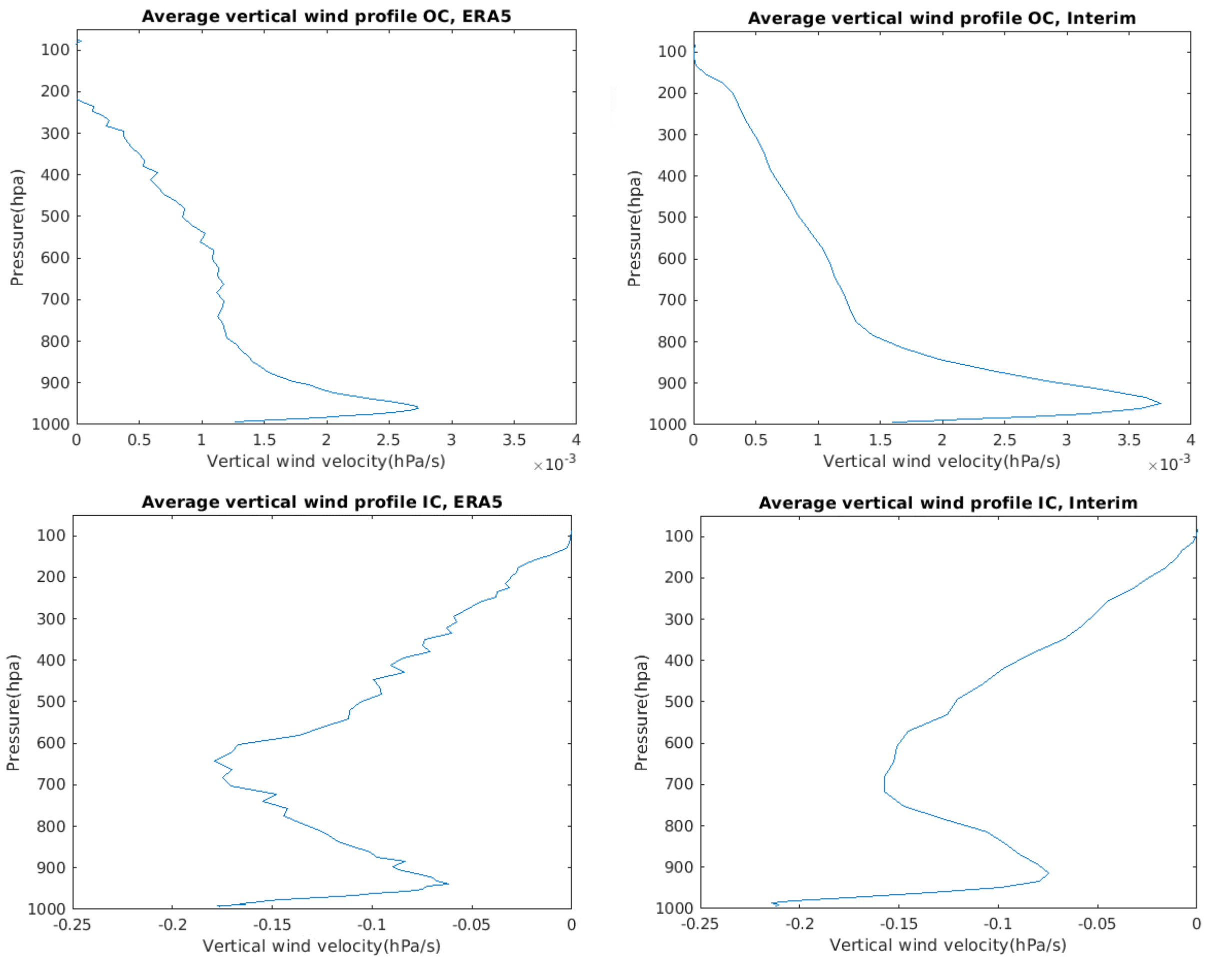
Updrafts outside and inside convection

ERA5 | ERA Interim



Mean Updraft Velocity OC	ERA5	2.67E-04
Mean Updraft Velocity IC	ERA5	-0.09
Percentage of traj IC	ERA5	0.34

Mean Updraft Velocity OC	Interim	6.53E-04
Mean Updraft Velocity IC	Interim	-0.1
Percentage of traj IC	Interim	0.69

Vertical wind profiles from model runs measured in hPa/s

OC = Outside convection
IC = Inside convection

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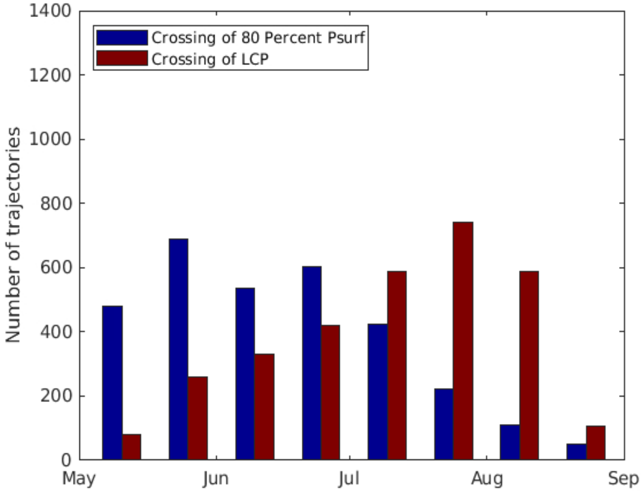
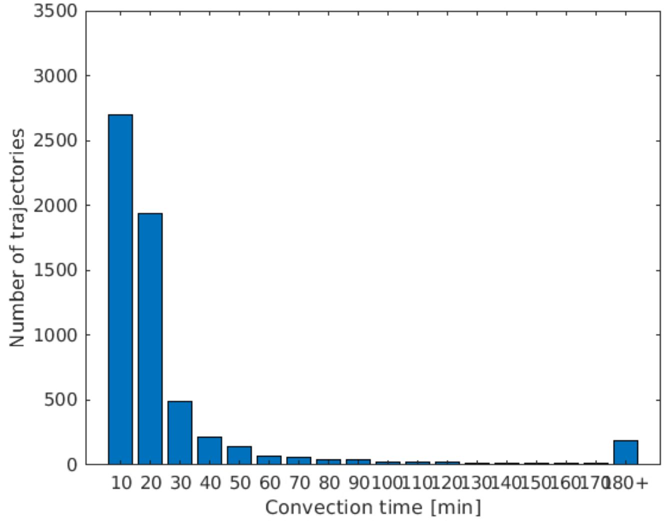
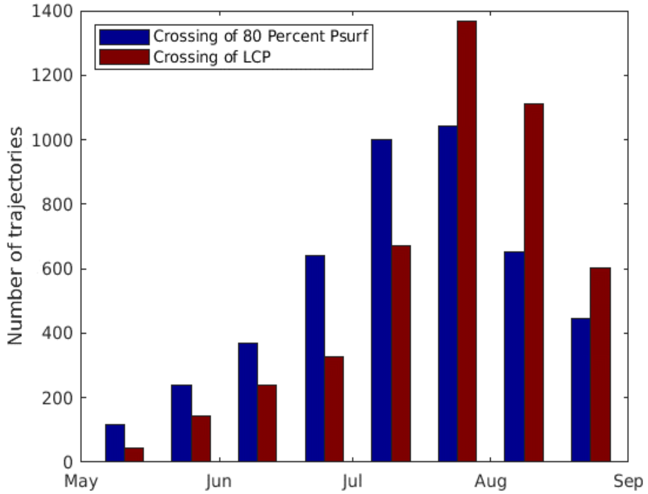
Crossing time and convection time

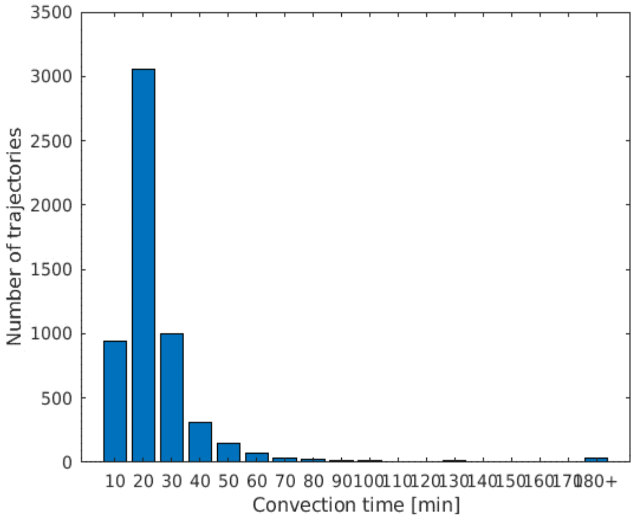
ERA5



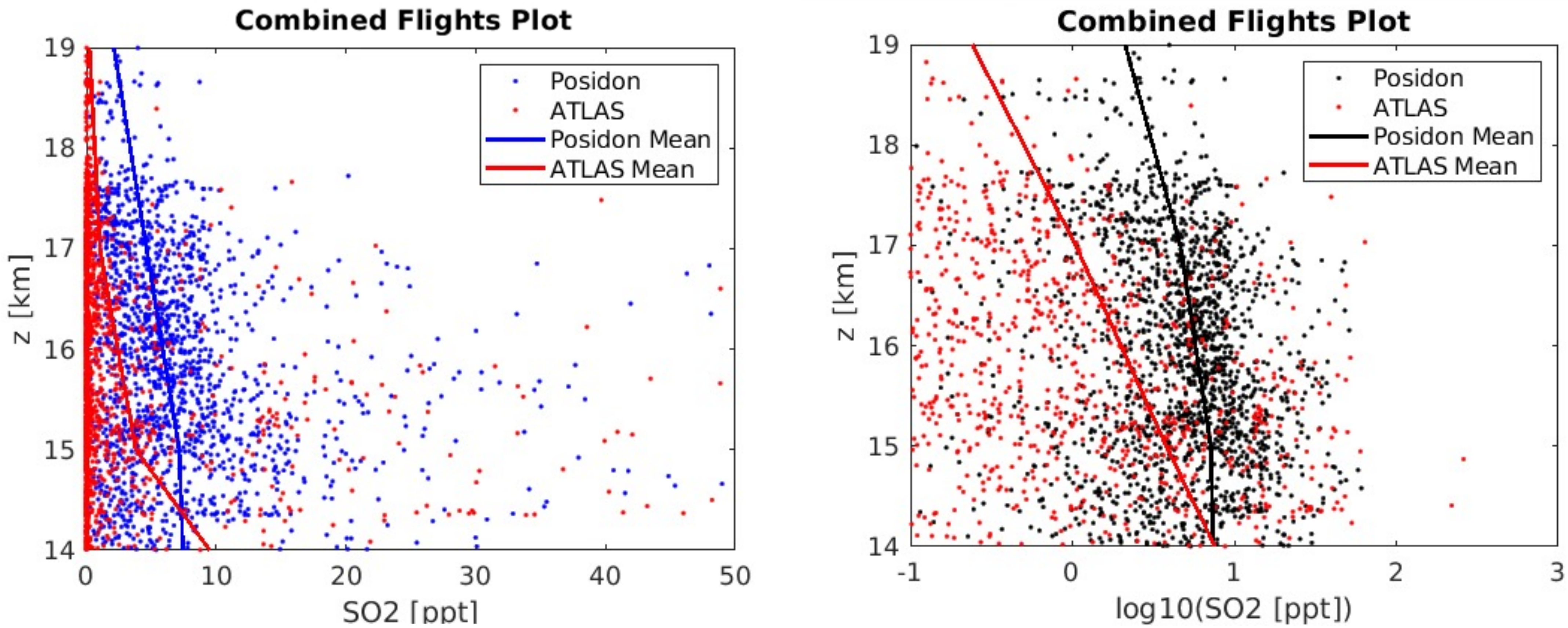


ERA Interim





Comparison with POSIDON data



Conclusion

- The main sources of SO_2 were found to be degassing volcanoes and anthropogenic sources
- The amount of SO_2 reaching the Aerosol layer is 3-10 GgS/yr
- The most sensitive parameter was pH(OH and DMS were also significant),
- The differences in ERA5 and ERA interim datasets were significantly large
- Our model underestimates SO_2 when compared to POSIDON data

Thank you

Questions?

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