Understanding the global sources and sinks of atmospheric carbonyl sulfide in order to provide insights into carbon cycle processes.

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1.1 Introduction – Carbonyl Sulfide (OCS)

- OCS is the most abundant sulphur containing gas in the atmosphere.
- OCS can be used as a proxy for photosynthetic uptake of carbon dioxide (CO_2) by vegetation.
- OCS is the largest contributing gas to the stratospheric aerosol layer (Junge layer) in periods of low volcanic activity.
- OCS has a global average mixing ratio of ~500 pptv and a lifetime of 2.5 years in the troposphere and 64±21 years in the stratosphere.
- Negligible trend in atmospheric OCS Concentration (Montzka et al., 2007), therefore the budget is closed.
- Sources include, direct and indirect oceanic emission, anthropogenic and biomass burning.
- Sinks include photosynthetic uptake by plants, oxic soil drawdown, reactions with OH and O^1D, and Photolysis.

1.2 Introduction – Proxy for CO_2 photosynthetic uptake

- Carbonic Anhydrase (CA) breaks down OCS, via the **irreversible** hydrolysis reaction.
- CA consumes OCS preferentially to CO_2.
- All OCS taken into leaf stomata is consumed, but only one third of CO_2 is.
- OCS is not emitted through respiration processes.
- All of the above makes the OCS vegetative flux easier to measure than that of CO_2.
- Gross primary Productivity (GPP) can be calculated on a leaf-scale using a Leaf Relative Uptake (LRU) parameter, ~1.6 (Stimler et al, 2012).

$$\mathrm{OCS} + \mathrm{H_2O} \xrightarrow{\mathrm{CA}} \mathrm{CO_2} + \mathrm{H_2S}$$

$$\mathrm{GPP} = \mathrm{F_{OCS}} \frac{[\mathrm{OCS}]}{[\mathrm{CO_2}]} \frac{1}{\mathrm{LRU}}$$

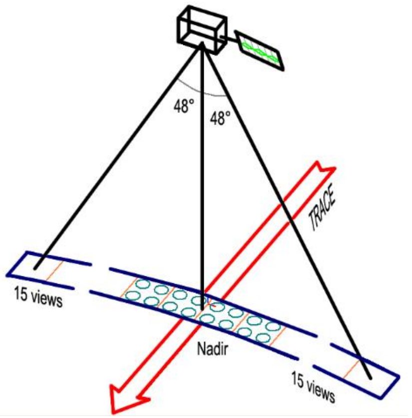
1.3 Introduction – Tools and Data

IASI-MetOp: 3 nadir viewing Fourier Transform Spectrometer instruments. Excellent spatial coverage.

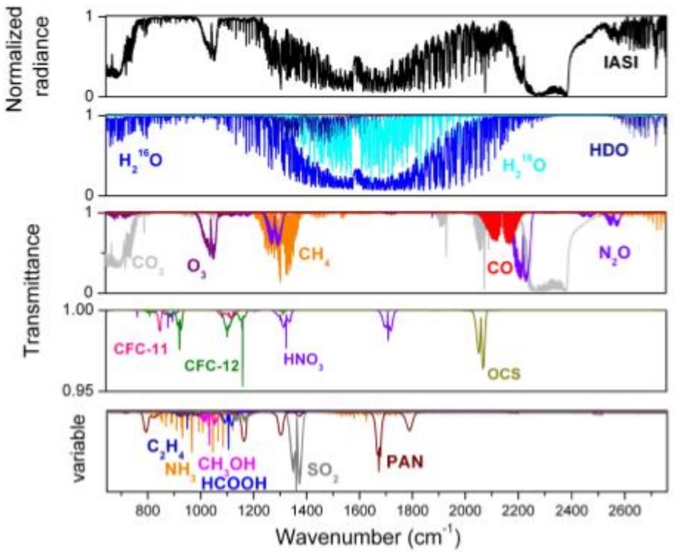
- Makes 14 orbits and 1.3 million spectral readings a day and covers a swath of 2200 km.
- University of Leicester IASI Retrieval Scheme (ULIRS) will be used to retrieve OCS total columns from IASI.

TOMCAT: 3D-CTM driven by ECMWF meteorological reanalyses, and by OCS fluxes from the literature

- Has a horizontal resolution of 1.2°x1.2° and a vertical range from surface to 10 hPa.
- Developed to simulate global OCS concentration.



IASI Viewing angle (Clerbaux et al., 2009).

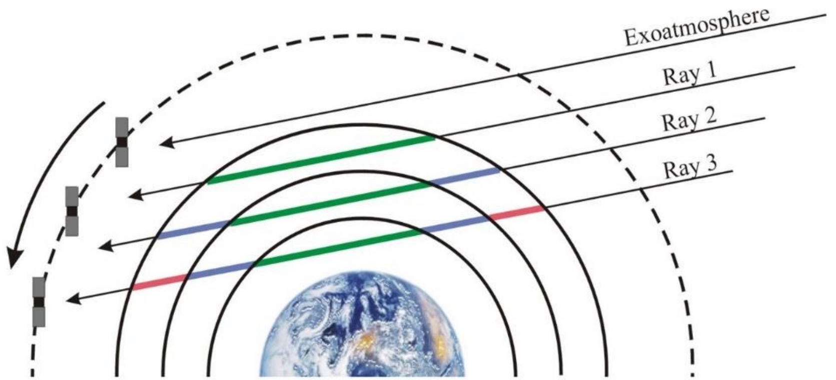


Full IASI Spectra (Clerbaux et al., 2009).

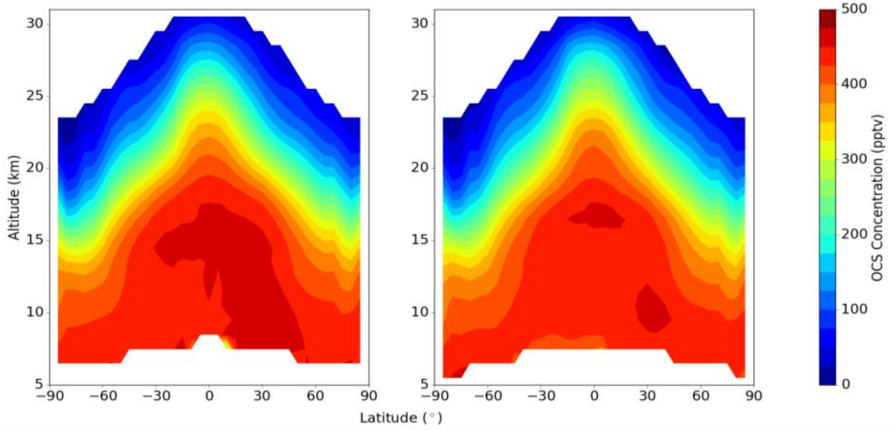
1.4 Introduction – Tools and Data

ACE-FTS: 1 limb-viewing Fourier Transform Spectrometer instrument.

- Makes a maximum of 30 measurements a day, due to its solar occultation measurement mode.
- Approximately 87000 measurements between 2004 and 2018.
- Retrieves with 3 km resolution and then interpolates on to a 1 km grid.
- Spectral resolution of 0.02 cm^{-1}.
- Version 4 of ACE-FTS OCS data (recently released) will be used in validation.
- Additional measurements added since version 3.6 and amendments to instrument line shape.



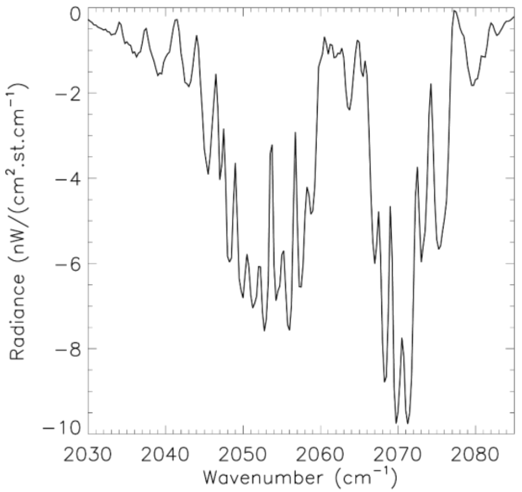
ACE-FTS viewing configuration.



Version 3.6 and Version 4 OCS concentration from ACE-FTS.

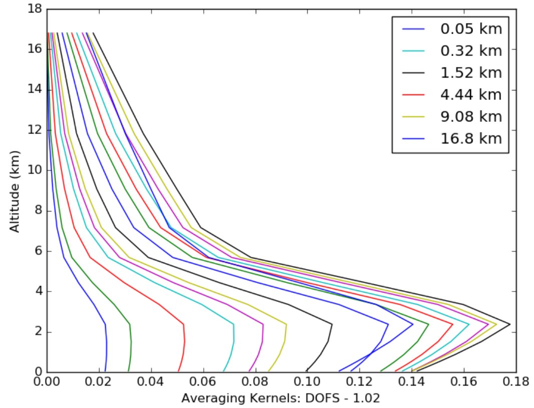
2.1 Satellite Retrievals

- Retrievals have been made from simulated IASI spectra, using the RFM as the forward model and ACE-FTS as prior and to calculate covariance.
- Retrieval micro-window is 2030 – 2070 cm^{-1}.
- Live retrievals have been made for SE USA.
- Further development is required and output to be compared to MORSE.

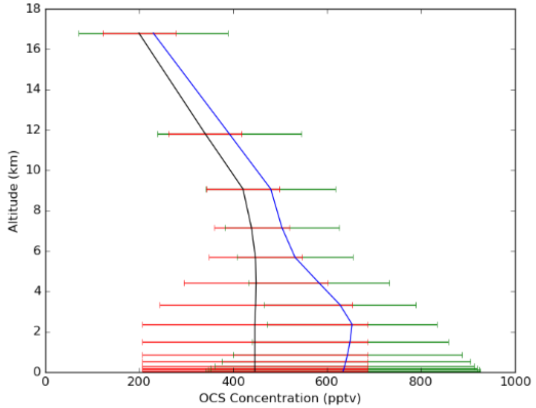


Difference in Radiance for simulated atmosphere with and without OCS ~ 18 km.

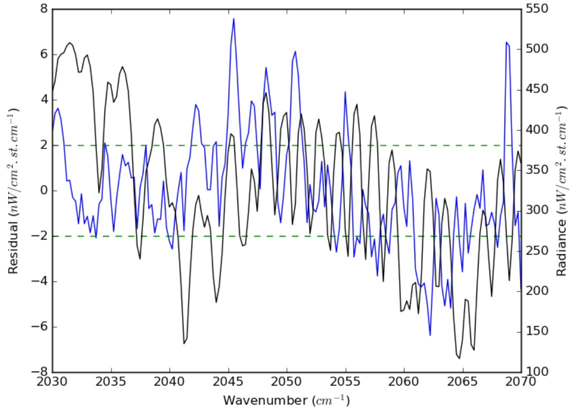
Residual spectra (blue) overlaying the final spectra (Black) and 2 nW noise range of IASI (green).



Averaging Kernels showing a DOFS of 1.02. Thermal contrast: 2.0°C.



A priori profile (black and red) and retrieved profile (blue and green).



3.1 TOMCAT Development

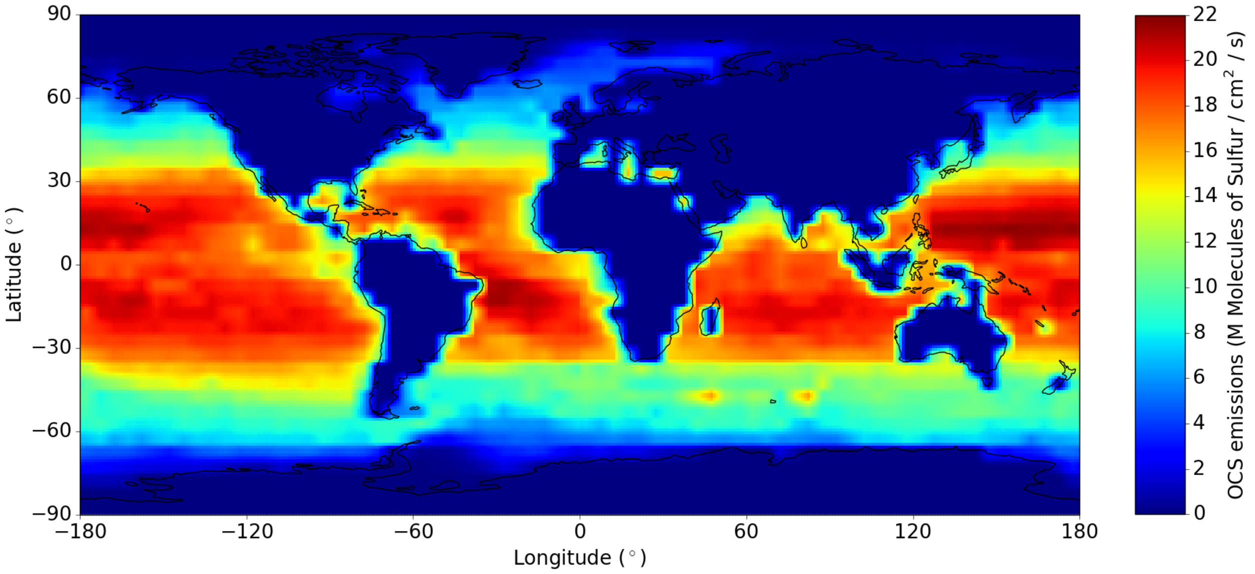
- The vegetative flux of OCS, F_{OCS}, has been calculated using the Net Primary Productivity (NPP) output from the JULES model:

$$F_{OCS} = \frac{[OCS]}{[CO_2]} \times NPP \times vd\_ratio.$$

- At each 6-hourly step F_{OCS} and [OCS] are updated.
- The emissions of CS_2 are scaled up to balance the increased vegetative sink.
- CS_2 is oxidized by OH to yield OCS:

$$CS_2 + OH \rightarrow OCS + SH$$

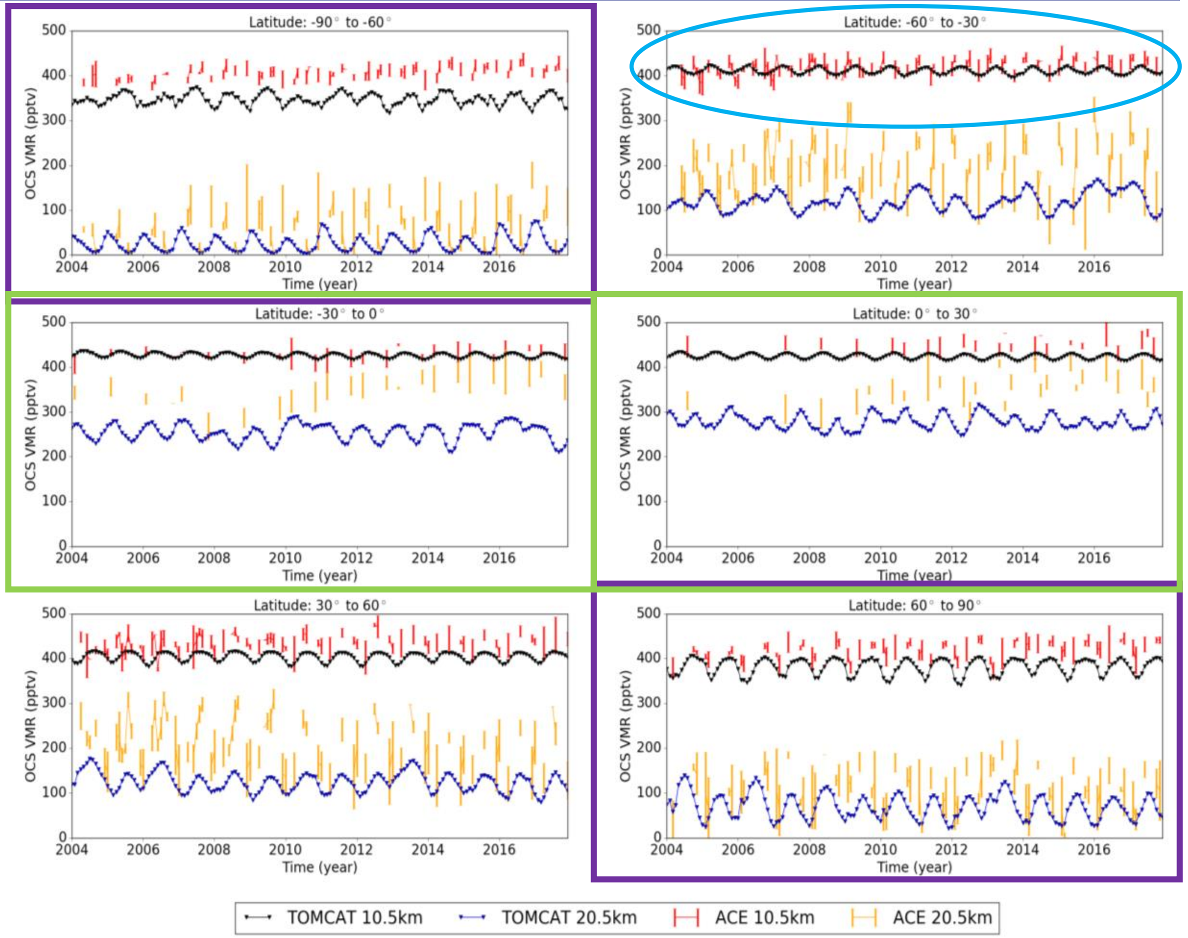
- These emissions are focused over the tropical ocean region – potential location of the 'missing source' (Glatthor et al., 2015).



Distribution of OCS emissions from oxidized ocean emitted CS_2.

3.2 TOMCAT Results

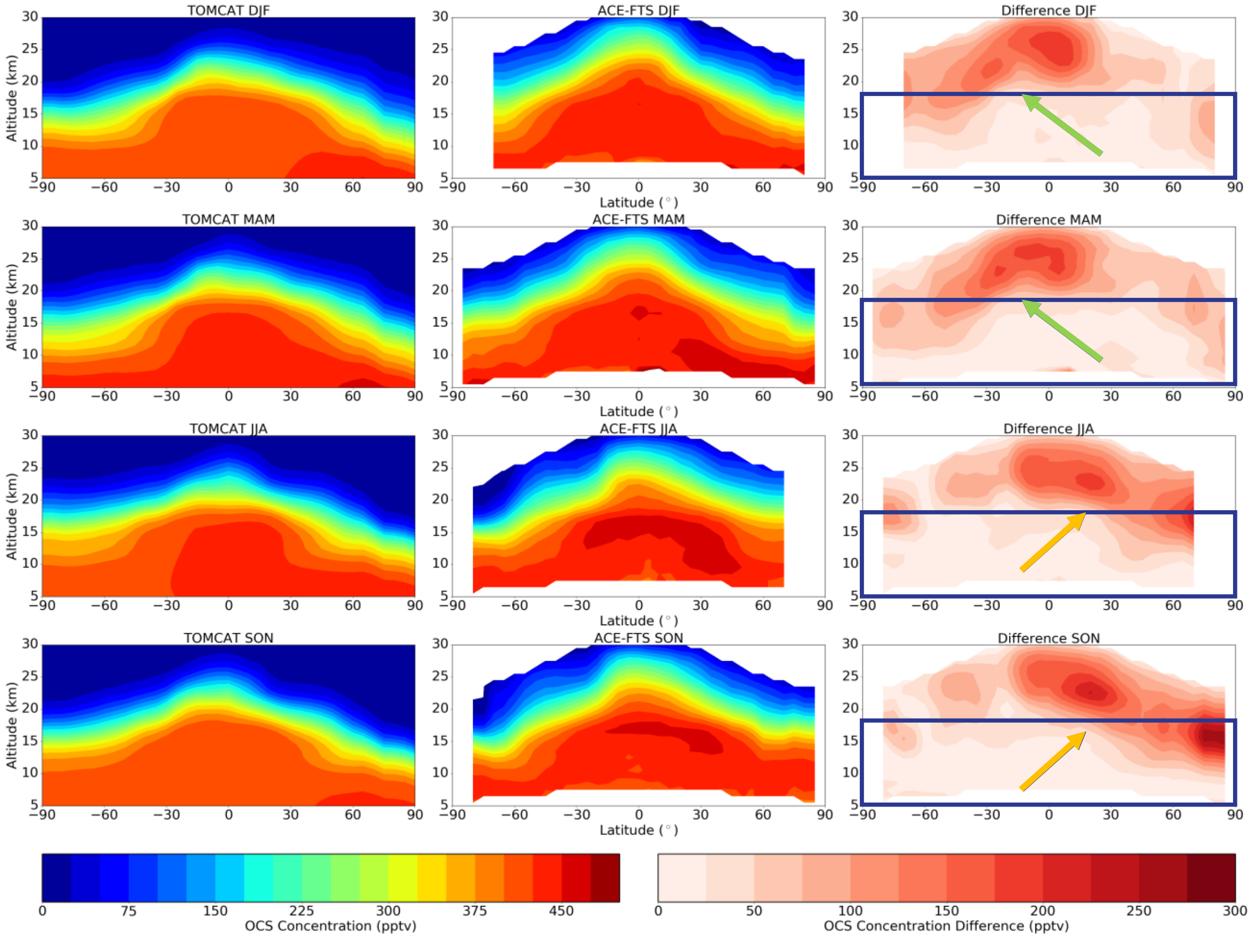
- SHML troposphere region shows the best agreement.
- Tropics show good performance in troposphere but underestimate UTLS region.
- High latitude regions underestimate the troposphere, but better in stratosphere.
- The model captures annual variability of OCS relatively well, with some inter-annual variability.



Inter-annual variability of OCS as modelled by TOMCAT and observed by ACE-FTS.

3.3 TOMCAT Results

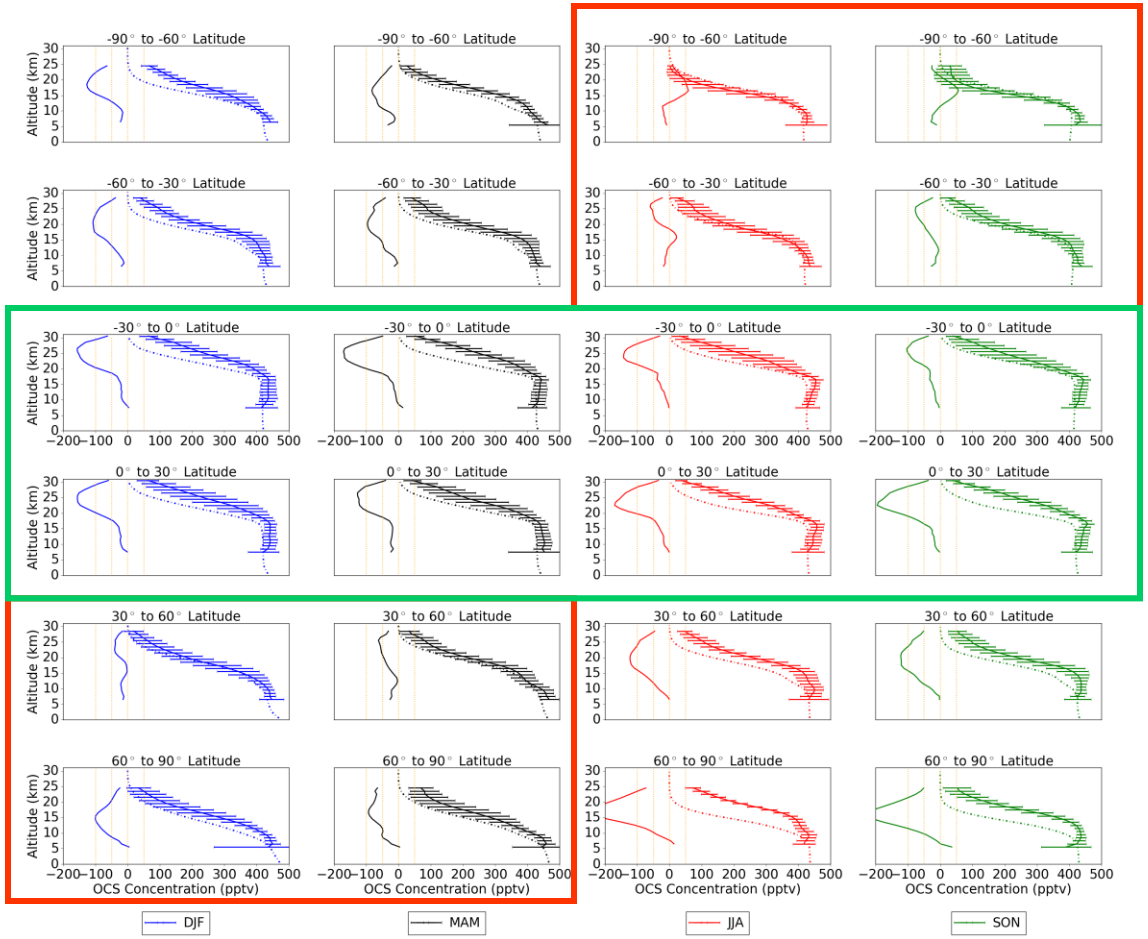
- 2004 – 2018 data from TOMCAT and ACE-FTS averaged.
- Troposphere region is generally within ±25 pptv globally.
- Tropopause region is where the most difference occurs between TOMCAT and ACE-FTS.
- Seasonal variation in the location of negative bias – DJF and MAM in SH and JJA and SON in NH.



Zonal contours of OCS from TOMCAT (left) and ACE-FTS (middle) for seasonal bins. The difference between the two is shown on the right.

3.4 TOMCAT Results

- Most of the profiles sit within or near the standard deviation of tropospheric ACE-FTS observations (up to ~15 km).
- Vertical gradient generally is good. Perhaps too strong in the tropics.
- Particularly good profiles include mid and high latitude regions in their respective winter/spring periods.



Profiles of OCS from TOMCAT (dot-dashed) and ACE (blocked), with the difference shown toward the left.

4 Summary and Future Work

Satellite Retrievals

- Using ULIRS, OCS can be remotely detected in the troposphere using the IASI instruments.
- Observing the real atmosphere yields far more complexity than simulating it.

Modelling

- TOMCAT has been amended to implement a new and stronger vegetative sink of OCS, as well as other fluxes from the literature.
- When compared to ACE-FTS, TOMCAT is particularly accurate in the troposphere.

Future

- Based on the success of various case studies, create a global satellite dataset of OCS using ULIRS and IASI.
- Distant future: use an inverse version of TOMCAT, in combination with satellite retrievals to yield a new flux inventory.

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