

Assessing the Contribution of Oceanic Fluxes to the Global Budget of Carbonyl Sulfide

P. Suntharalingam¹, Zhaohui Chen¹, Sinikka Lennartz², E. Buitenhuis¹

1: University of East Anglia, UK; 2: University of Oldenburg, Germany

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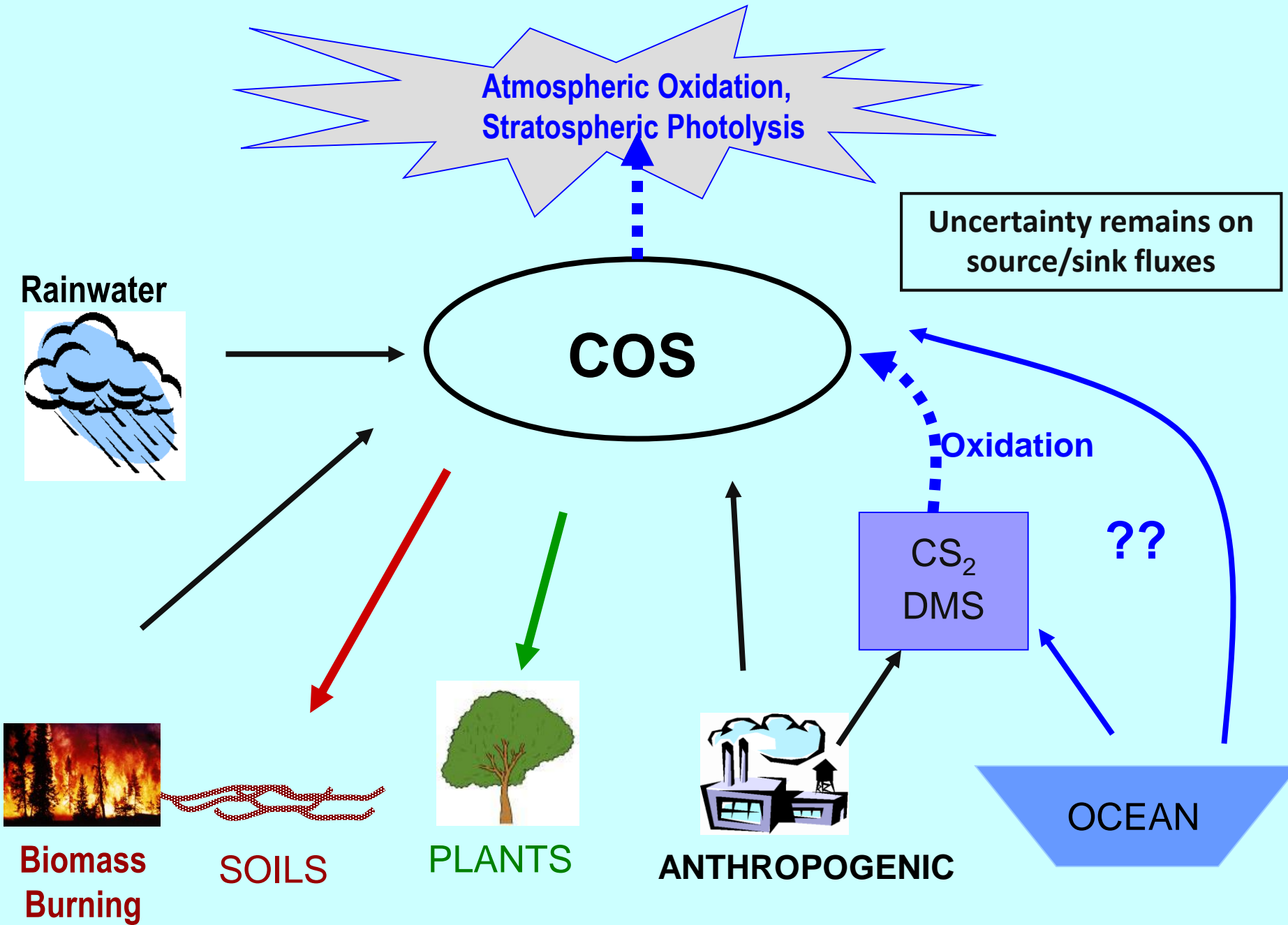
SUMMARY

Carbonyl sulfide (COS) is an agent of stratospheric ozone depletion. Measurements of its tropospheric variation also provide constraints on terrestrial primary productivity. The ocean is a major source of COS to the troposphere through direct emission, and potentially through emission of COS precursors such as carbon disulfide (CS₂). Recent estimates of the global COS budget, show large imbalances between known sources and sinks, with significant uncertainty in the magnitude of the oceanic flux (the largest natural COS source to the atmosphere).

Here we assess the role of oceanic fluxes in the global COS budget using top-down constraints from a global model analysis incorporating atmospheric COS measurements from a global network of surface sites (NOAA-HATS). Our initial results estimate oceanic COS fluxes in the range 98-191 Gg S/yr, and smaller than those of recent studies.

Our ongoing and planned work will address improvements to the methodology, including updates of prior flux inventories, and incorporation of additional COS measurements.

SOURCES AND SINKS OF ATMOSPHERIC CARBONYL SULFIDE



Recent Estimates of Ocean COS Fluxes (Gg S/yr)

Lennartz et al. 2017
Reported in Whelan et al. 2018

265 (\pm 210) Direct + from CS₂

Launois et al. 2015

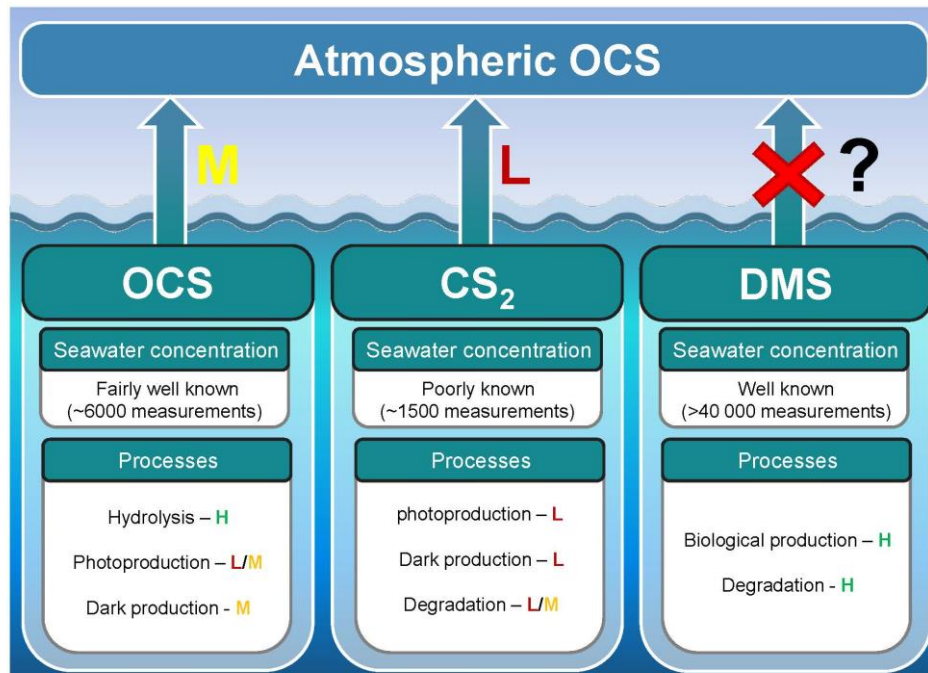
573-3997 Direct (Dark + photoproduction)

Berry et al. 2013

276 (prior) + **600** (photochemical ocean flux)

Suntharalingam et al. 2008

230 (Adjusted from Kettle et al. 2002)



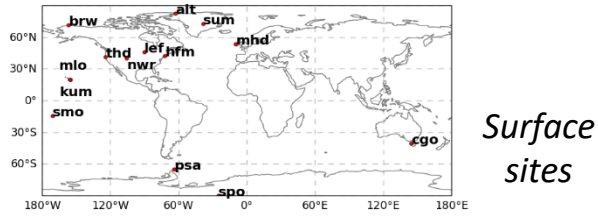
Level of understanding: H – high, M – medium, L – low

- Large range in recent estimates of ocean COS fluxes
- Uncertainty on direct and indirect production mechanisms

Whelan et al. 2018

COS Flux Estimates from Atmospheric Inverse Analyses

Atmospheric COS Observations



Surface sites

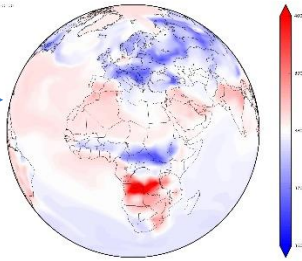
INVERSE MODEL

LETKF-GEOSChem

Local Ensemble Transform Kalman Filter

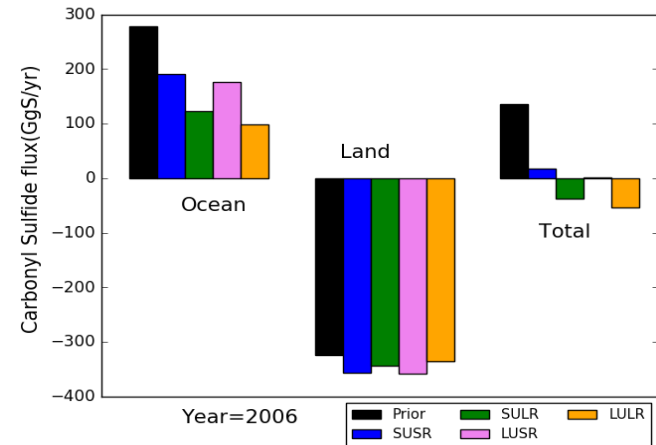
Model Concentrations

Prior Fluxes



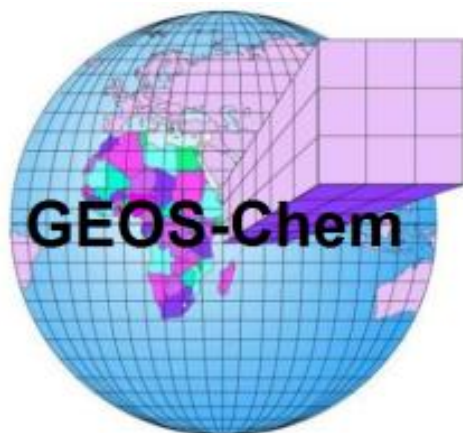
Atmospheric Transport Model
GEOS-Chem v11.01

Optimized Inverse Flux Estimates



Atmospheric COS measurements in conjunction with inverse analysis methodologies provide 'top-down' constraints on surface COS fluxes

COS Inverse Analysis Specifications



ATMOSPHERIC TRANSPORT MODEL : GEOS-Chem v11-01

Global 3-D model of atmospheric chemistry driven by meteorological input from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office.

Resolution : 2° (lat) x 2.5° (lon) ; 72 vertical levels

Meteorology : NASA GMAO GEOS-FP, GEOS-5, MERRA-2 (Available 1980-present)

COS Prior Fluxes : Wang et al. 2016; Suntharalingam et al. 2008; Kettle et al. 2002

INVERSE METHODOLOGY

Local Ensemble Transform Kalman Filter (LETKF)

(Hunt et al.2007, Miyoshi et al.2007, Chen et al., 2013)

ATMOSPHERIC COS MEASUREMENTS

HATS Flask Program *Montzka et al. 2004, 2007*

<https://www.esrl.noaa.gov/gmd/hats/gases/OCS.html>

Acknowledgements : S. Montzka, NOAA-GMD

Assessment at HATS Sites

Seasonal Variation : Year 2006

● Observations

— Prior Flux simulation

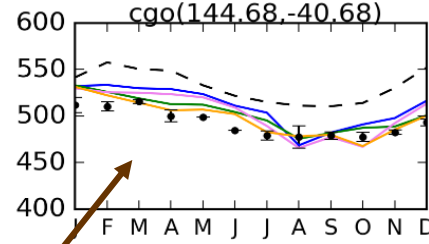
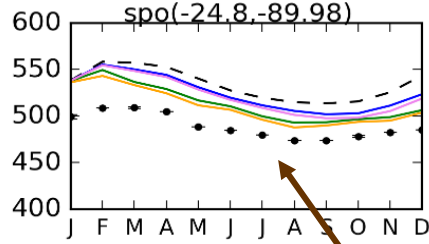
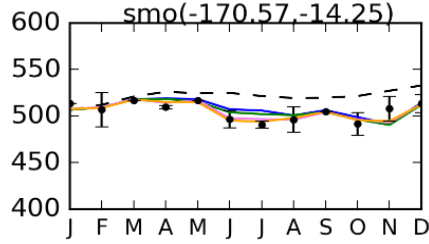
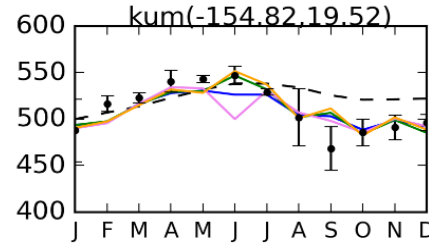
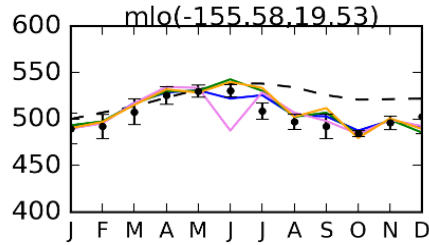
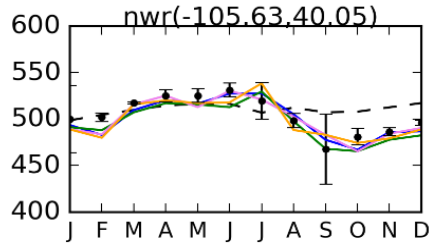
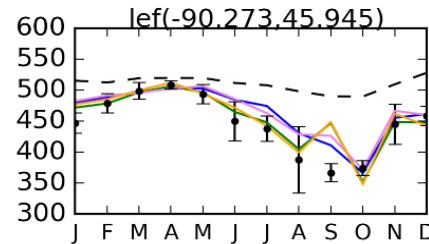
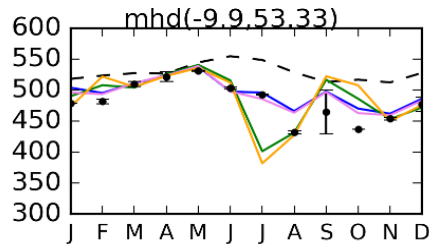
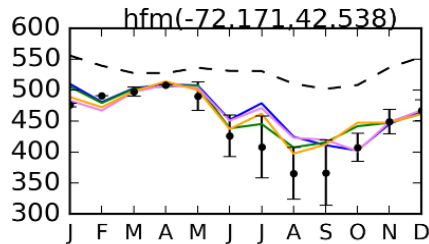
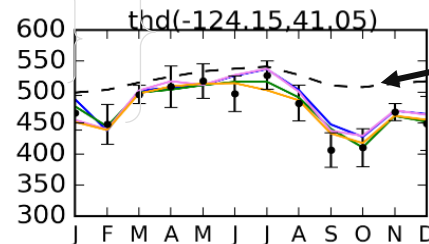
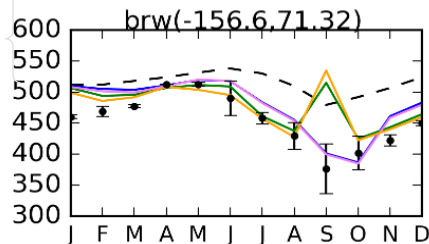
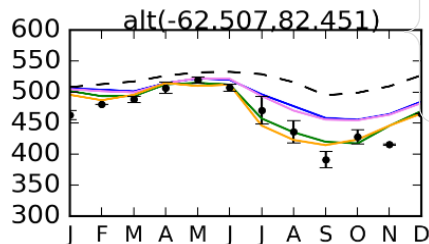
Posterior Estimates:
Sensitivity Cases

SUSR : Small Prior
Uncertainty; Small
Localization Radius

SULR : Small Prior
Uncertainty; Large
Localization Radius

LUSR : Large Prior
Uncertainty; Small
Localization Radius

LULR : Large Prior
Uncertainty; Large
Localization Radius

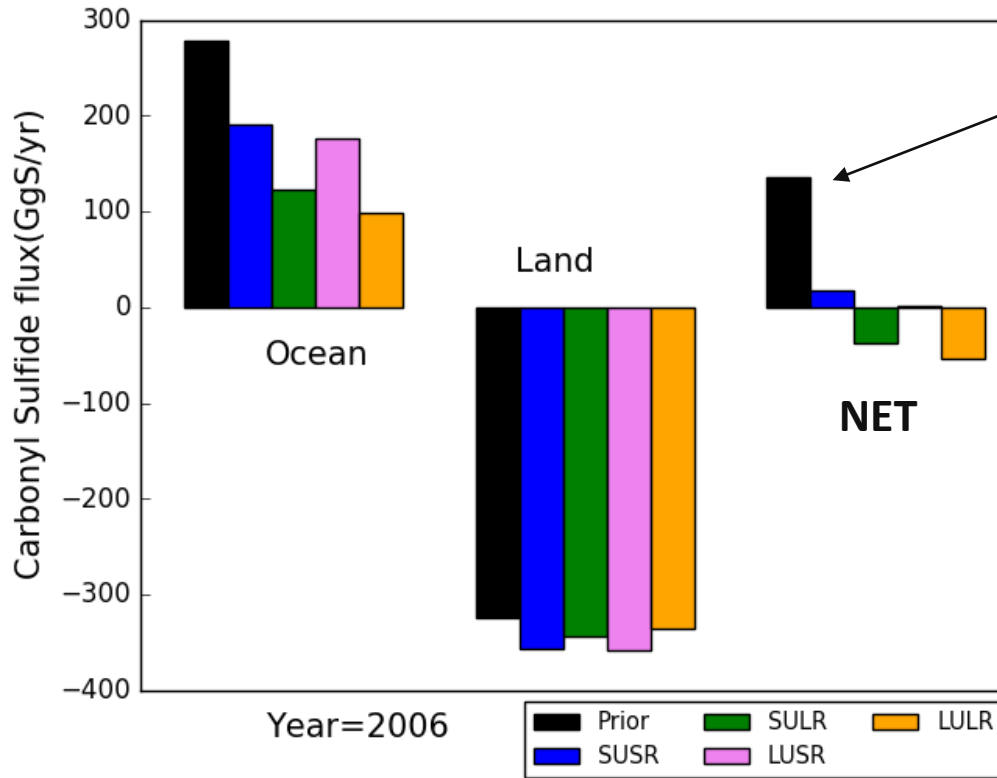


Simulations overestimate COS at Southern Hemisphere sites

Carbonyl Sulfide mixing ratio(ppt)

COS Flux Estimates

Global Totals for Year 2006



BLACK: PRIOR FLUXES

Posterior Estimates: Sensitivity Cases

SUSR : Small Prior Uncertainty; Small Localization Radius

SULR : Small Prior Uncertainty; Large Localization Radius

LUSR : Large Prior Uncertainty; Small Localization Radius

LULR: Large Prior Uncertainty; Large Localization Radius

GLOBAL FLUX TOTALS Year 2006 (Units (Gg S/yr))

	Prior	Posterior (range)
Ocean	280	98-191
Land	-325	-358 to -335
NET*	135	-54 to +18

OCEAN = Direct and Indirect Emissions

LAND = Uptake by Vegetation and Soils + Biomass Burning Emissions

*NET Flux also accounts for Anthropogenic emissions (kept constant at 180 Gg S/yr)