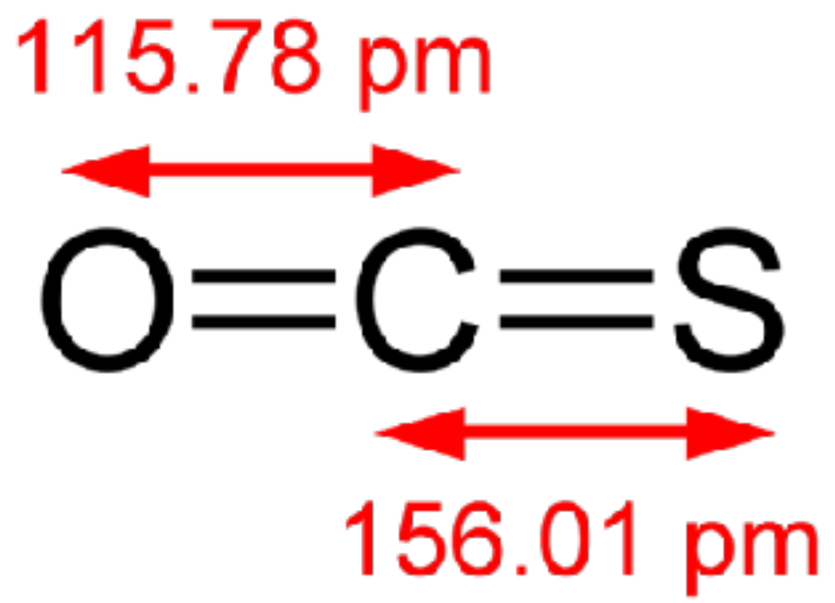
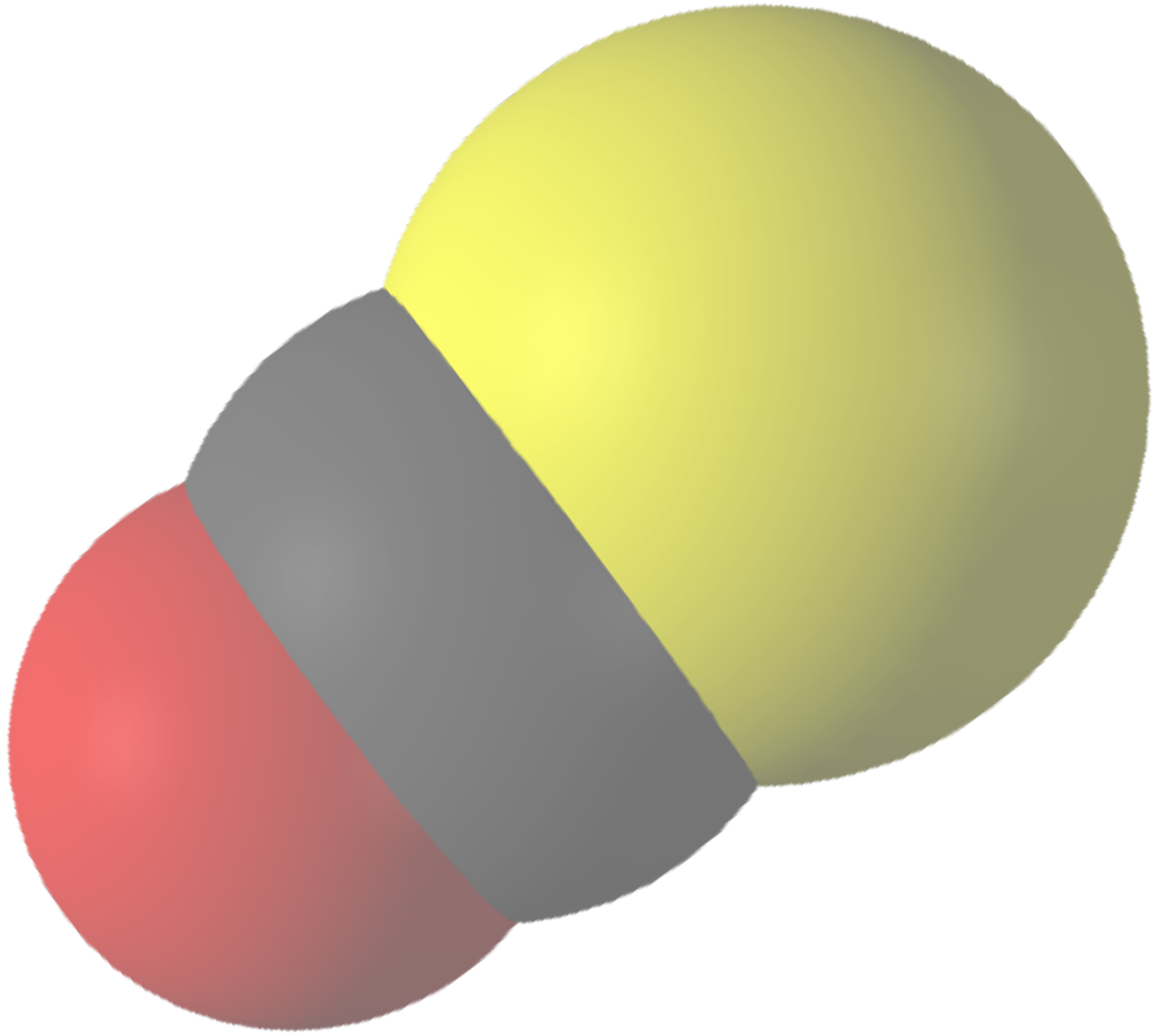
COS-OCS



The global budget of Carbonyl Sulfide, recent insights and remaining unknowns

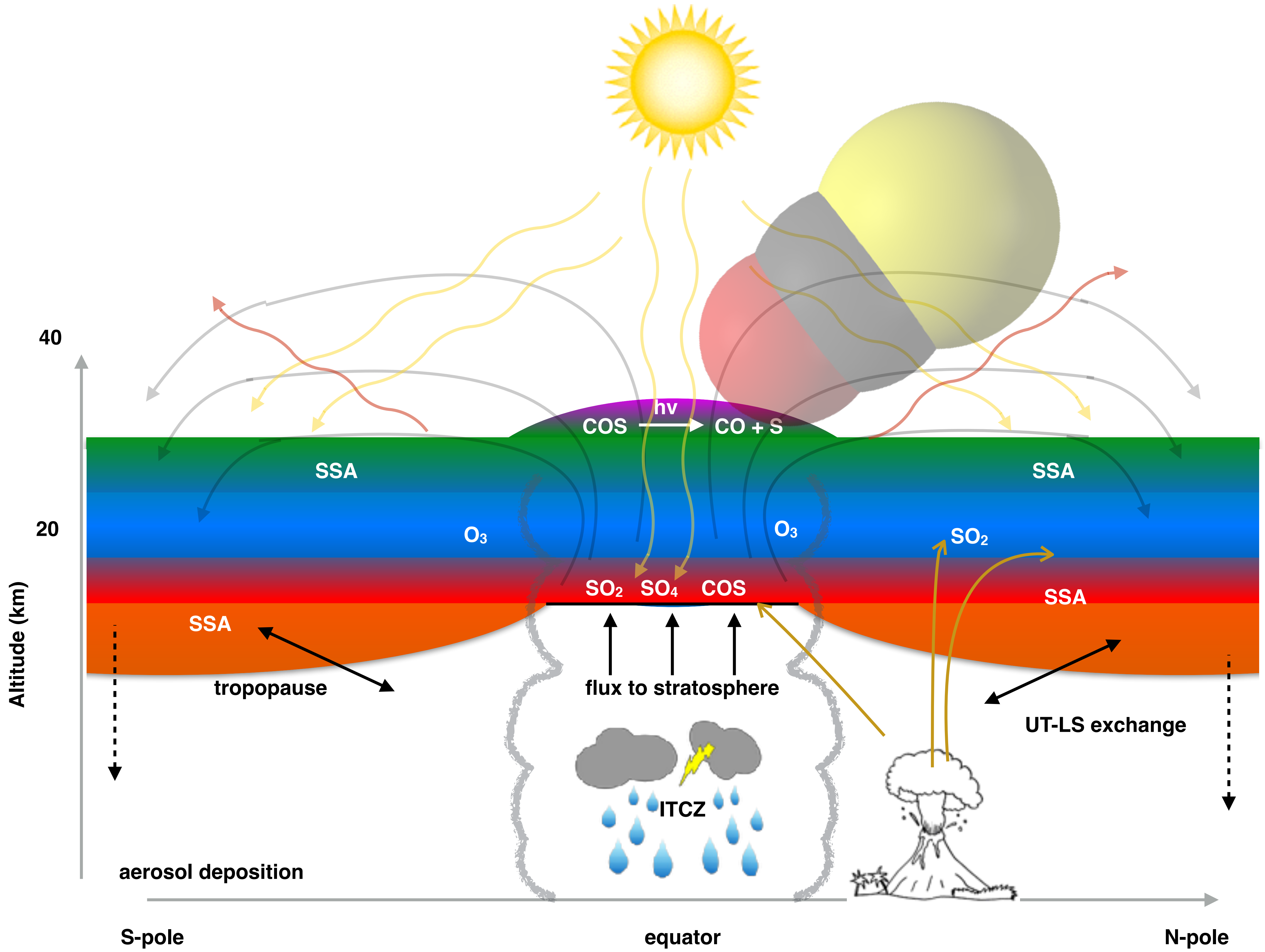
Maarten Krol, 2nd international COS workshop,
Obergurgl, Austria, 11-14 november, 2019



Thanks to: Linda Kooijmans, Jin Ma,
Peter Bosman, NOAA, TES-team,
SIB4-team, COS-OCS team

This project has received funding from the European Research Council (ERC) under the European Union's H2020 research and innovation programme under grant agreement No 742798

COS: New ways of Observing the Climate System



COS contributes to Stratospheric Sulfate Aerosol (SSA)

COS-OCS

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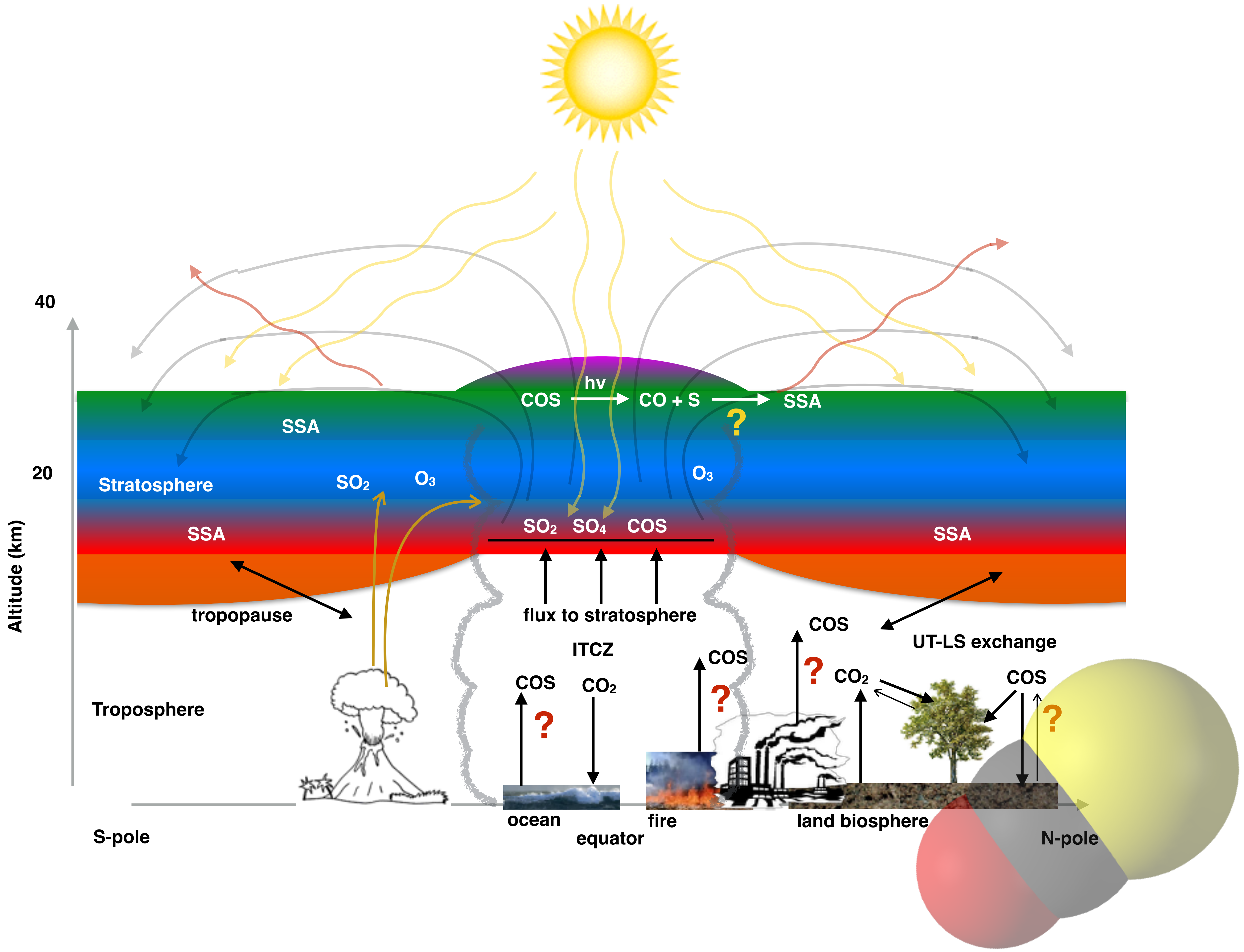
COS: New ways of Observing the Climate System



Large uncertainties in the global COS budget hamper use as GPP tracer

COS-OCS

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COS: New ways of Observing the Climate System

- Modelling COS & CO_2 (Linda, Peter, Jin)
- Air-core observations (Huilin)
- Measurements of COS isotopologues (Elena & Sophie)
- Modelling COS Isotopologues (Juhi)

COS-OCS

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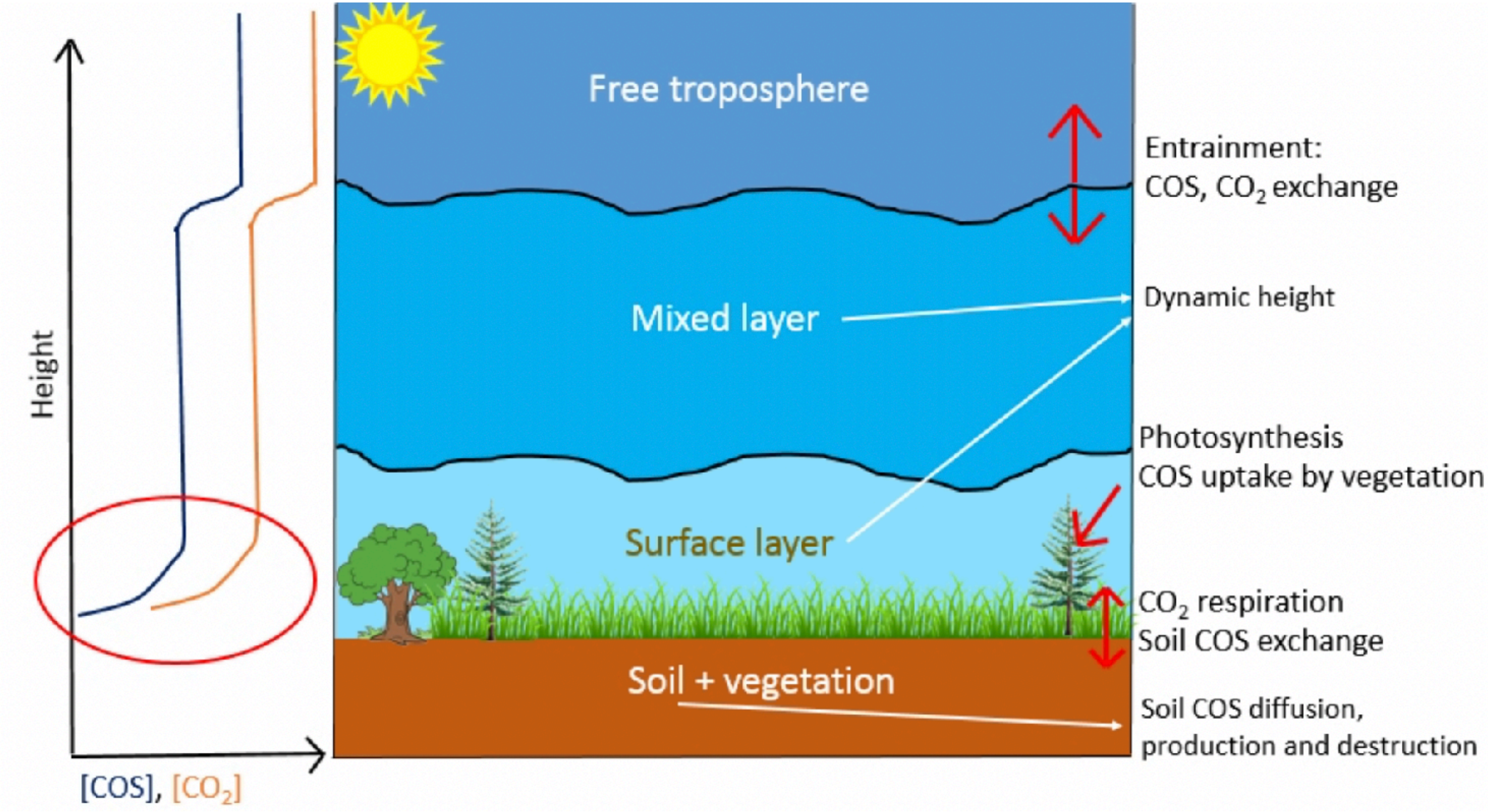
COS-OCS



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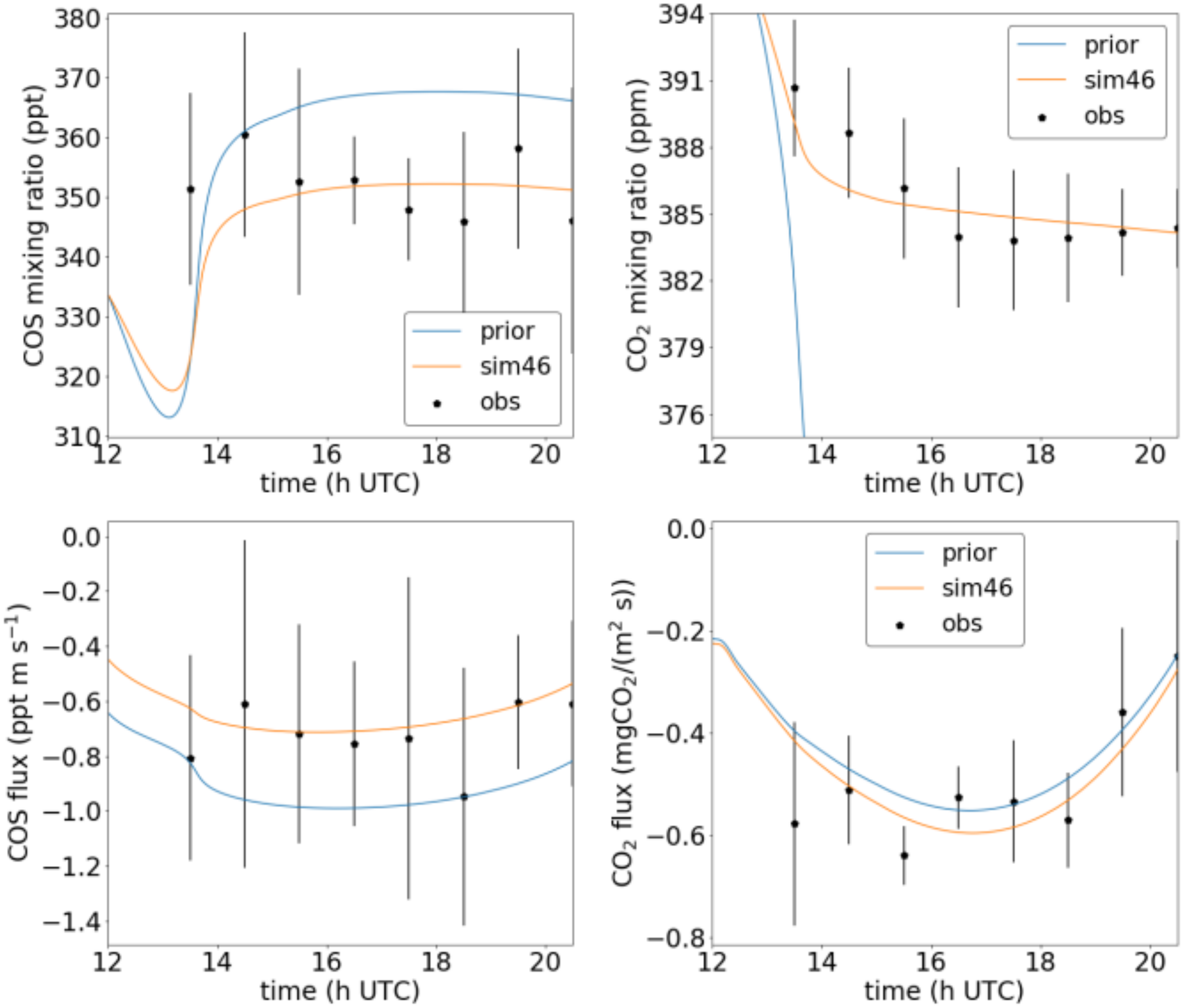
Proof of concept for Harvard Forest



Dataset from temperate forest in USA – Commane et al.

COS and CO_2 mixing ratios; eddy-covariance fluxes at 29 m above the surface +…

Averaged over 7 relatively sunny September days

Msc Thesis Peter Bosman

COS-OCS

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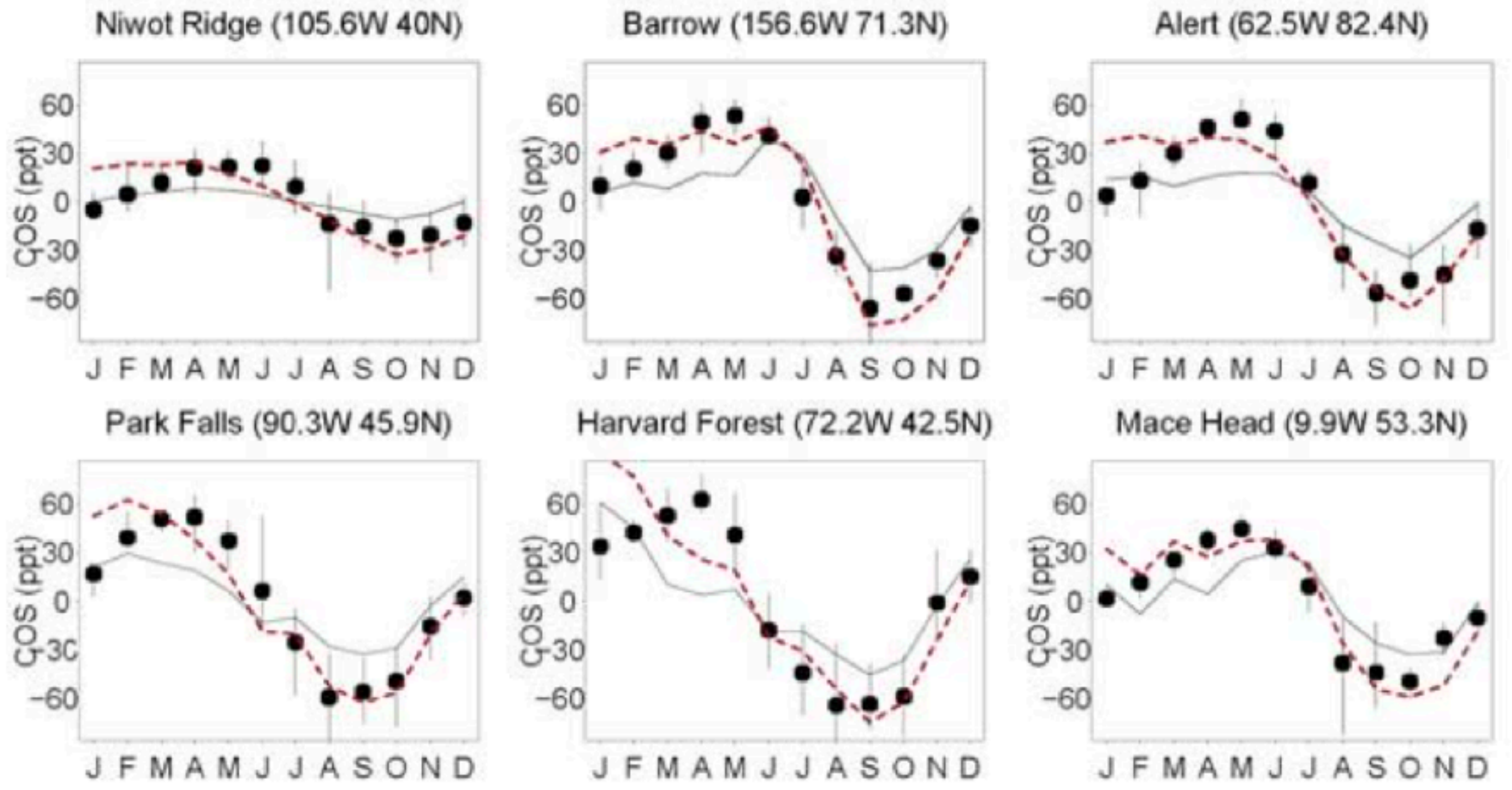
Global Inverse modelling

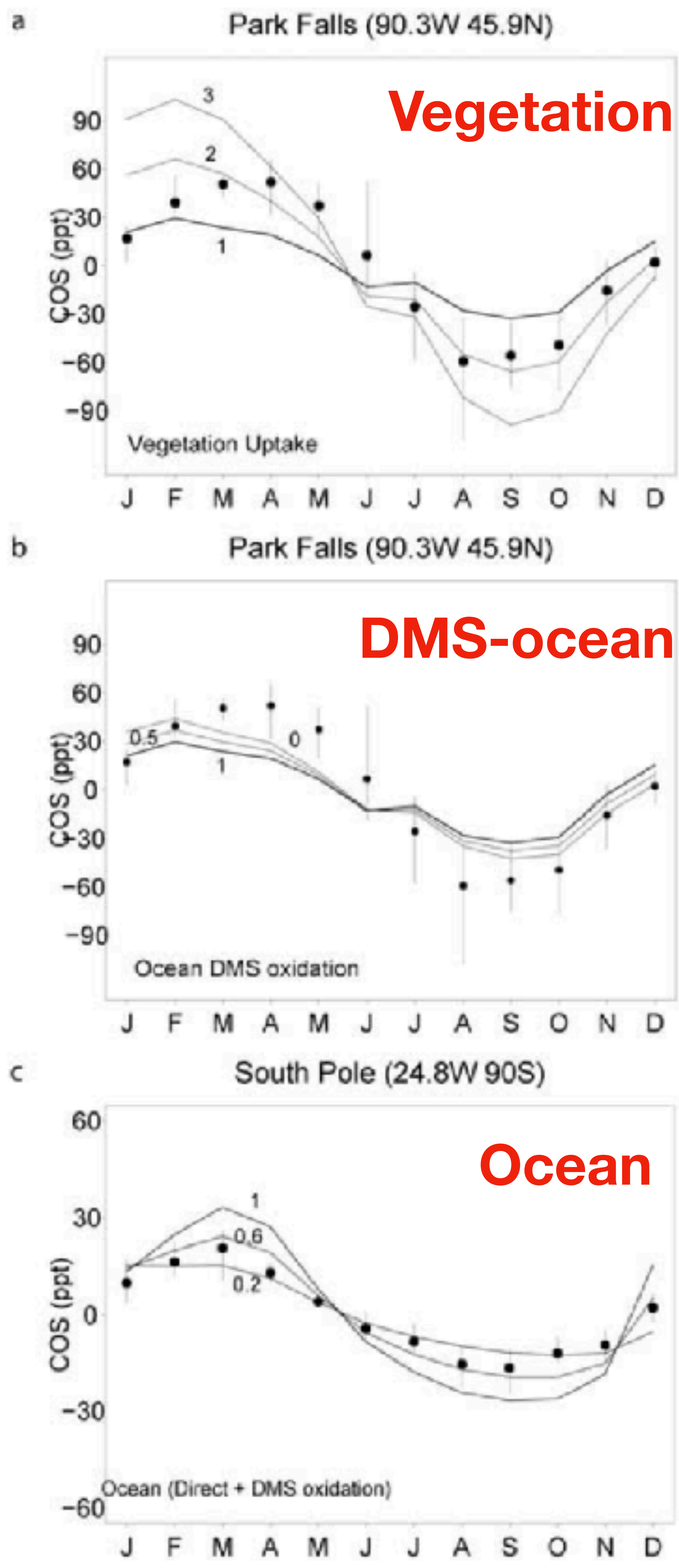
SUNTHARALINGAM ET AL.: SEASONAL CYCLE OF CARBONYL SULFIDE

2008



[*Montzka et al.*, 2007]. Simulations that increase the seasonally varying plant uptake by factors of 1.9 to 2.2 (to 460 – 530 Gg S y^{-1}) produce the lowest RMSEs and improved representation of the seasonal cycle. This top-

[18] Minimum RMSE values are obtained with a reduction of ~50 Gg S y^{-1} equivalent to ~60% of the southern extratropical ocean flux. This is within the uncertainty



COS-OCS

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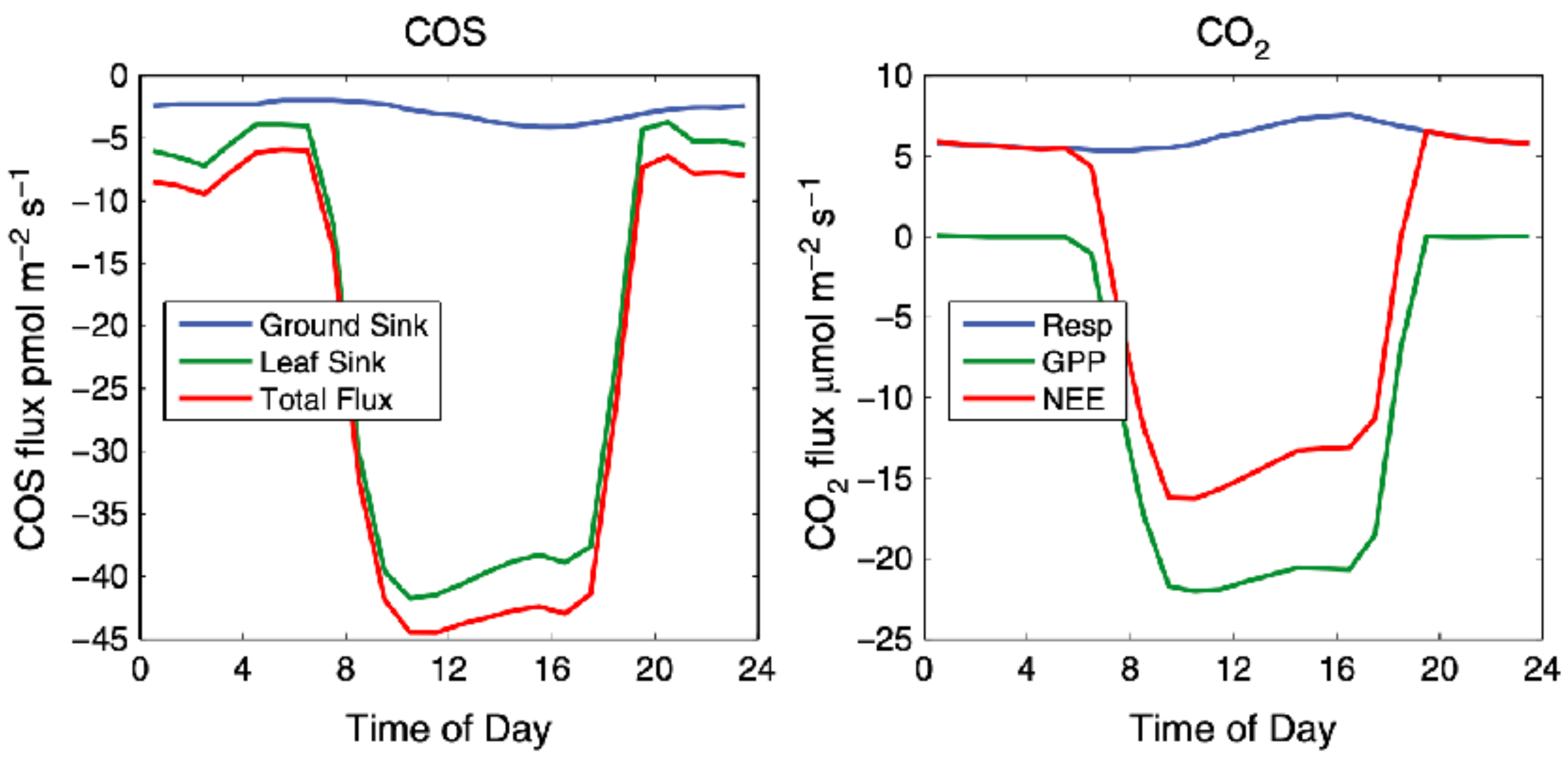
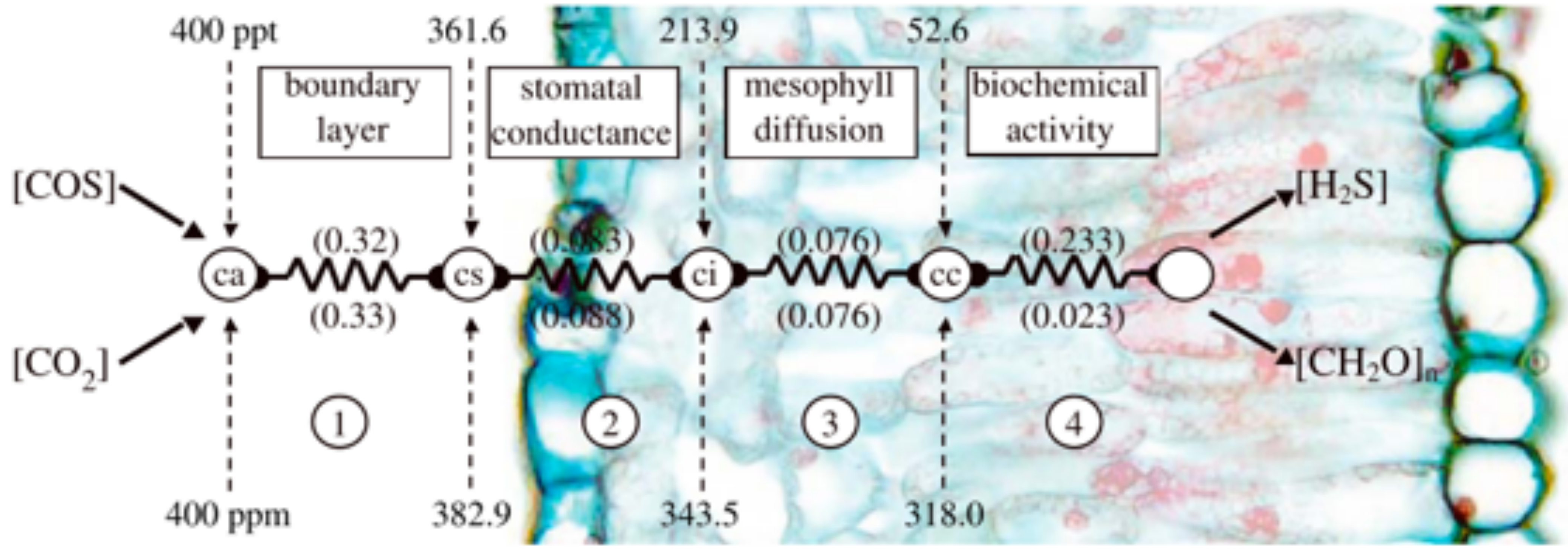
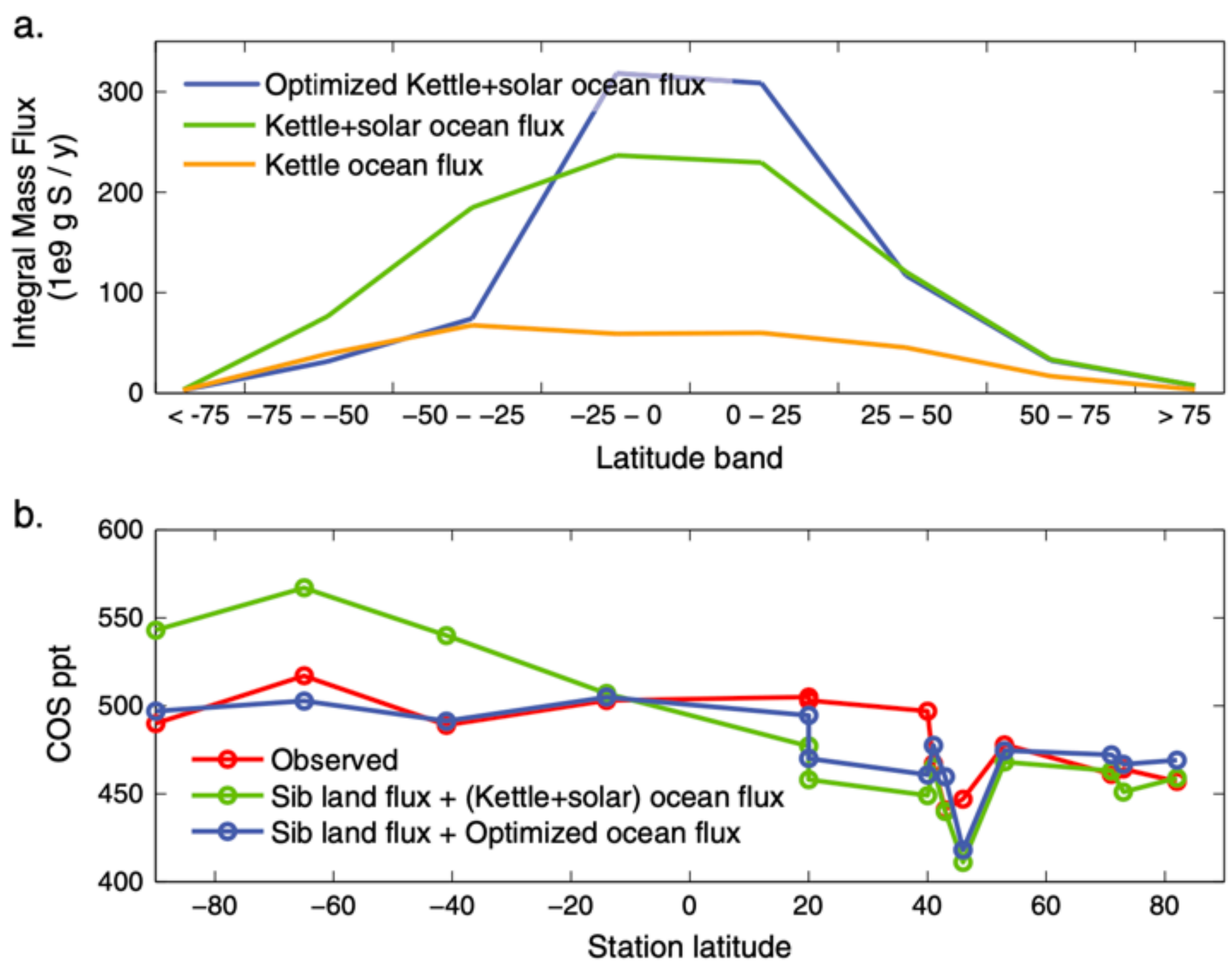
Inverse modelling

BERRY ET AL.: CARBONYL SULFIDE AS A GLOBAL CARBON CYCLE TRACER







(Berry et. al 2013)

COS-OCS

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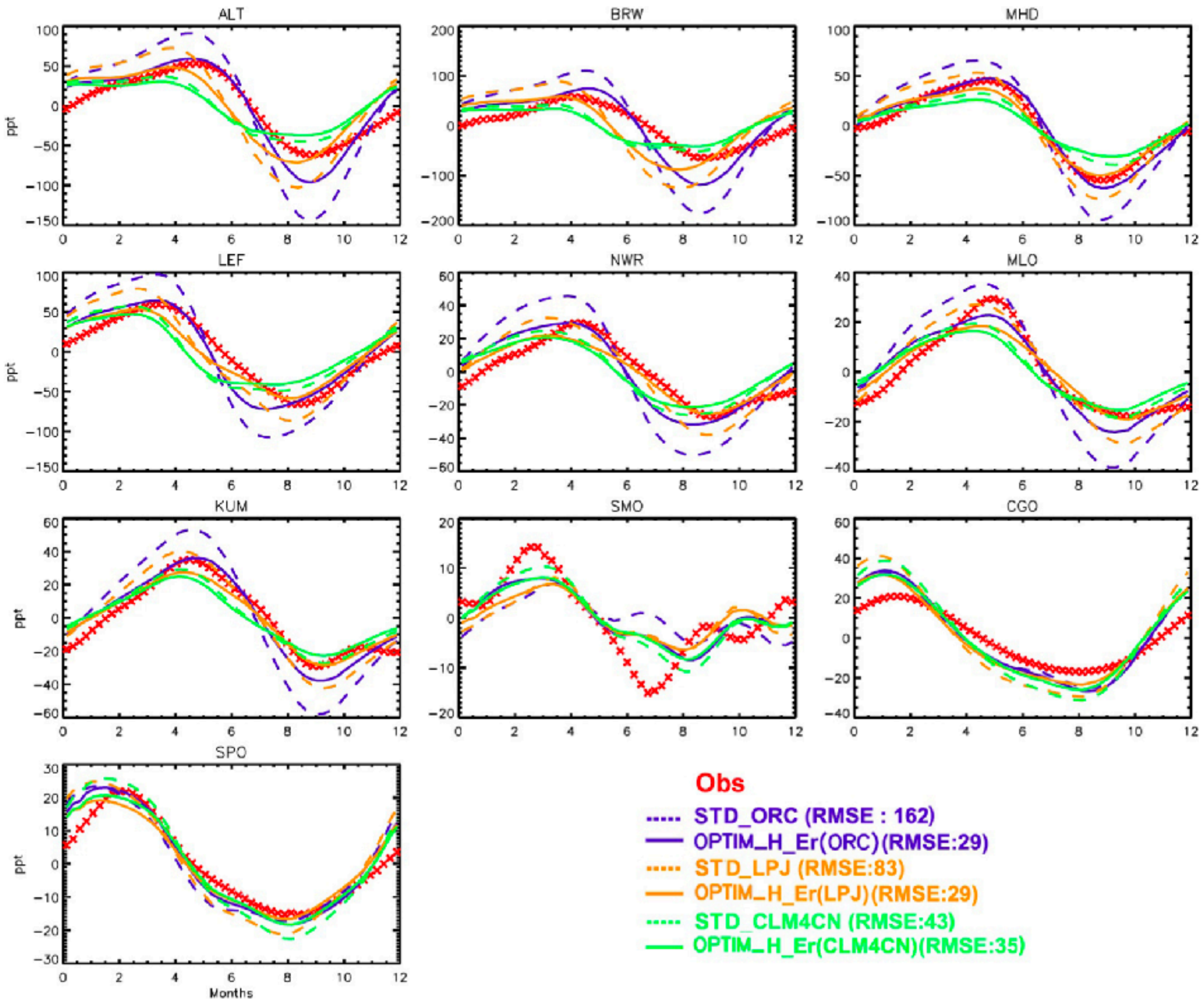
Inverse modelling



Launois et al, ACP 2015

Linearised Scaling Factors

when given a variation limit of 50 %. On average, vegetation and soil optimized uptakes are respectively converging around 714 and 396 Gg S yr^{-1} (Table 3). The atmospheric

1. the vegetation plays a determining role in the OCS global atmospheric budget
2. the leaf uptake of OCS is too large when using ORC, highlighting a too large global annual GPP flux.

Note: Very high prior Ocean flux

COS-OCS

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Inverse modelling

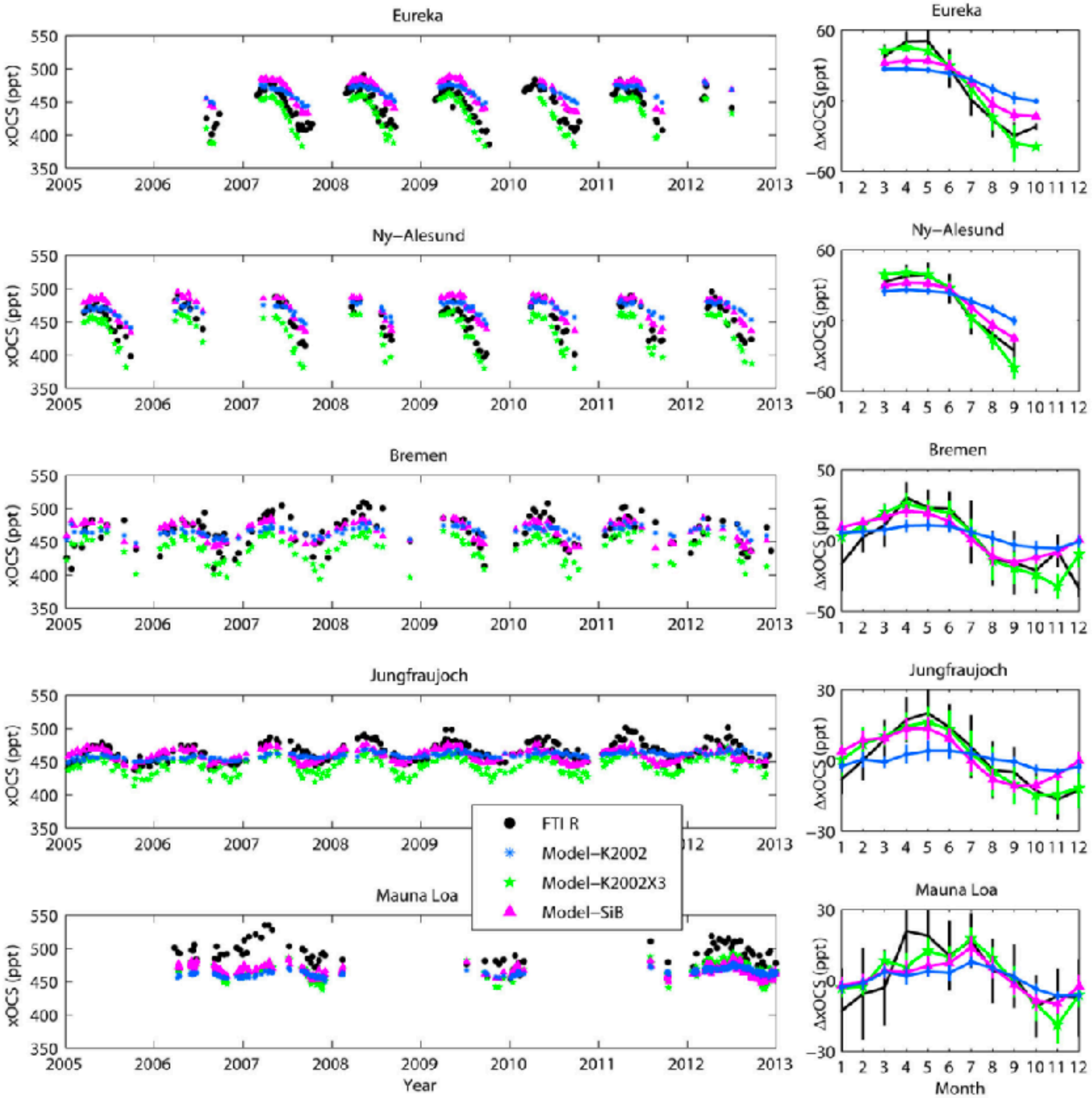
Y. Wang et al.: Towards understanding the variability in biospheric CO_2 fluxes

ACP, 2016

FTIR xOCS

Following Suntharalingam et al. (2008), we rescaled the fluxes in K2002, including increasing the plant uptake, increasing the ocean emissions in the tropics, and decreasing the ocean emissions in the Southern Ocean, to find a better match to the column measurements. Multiplying the plant uptake by a factor of 3 (K2002x3, Fig. 2 green stars) agrees with the measurements best.

lations with Kettle's fluxes. SiB did not capture the strong latitudinal gradient during growing season (HIPPO-5), indicating that the plant uptake of OCS in SiB in the boreal forest is too small, at least for the year (2011) in question.



COS-OCS

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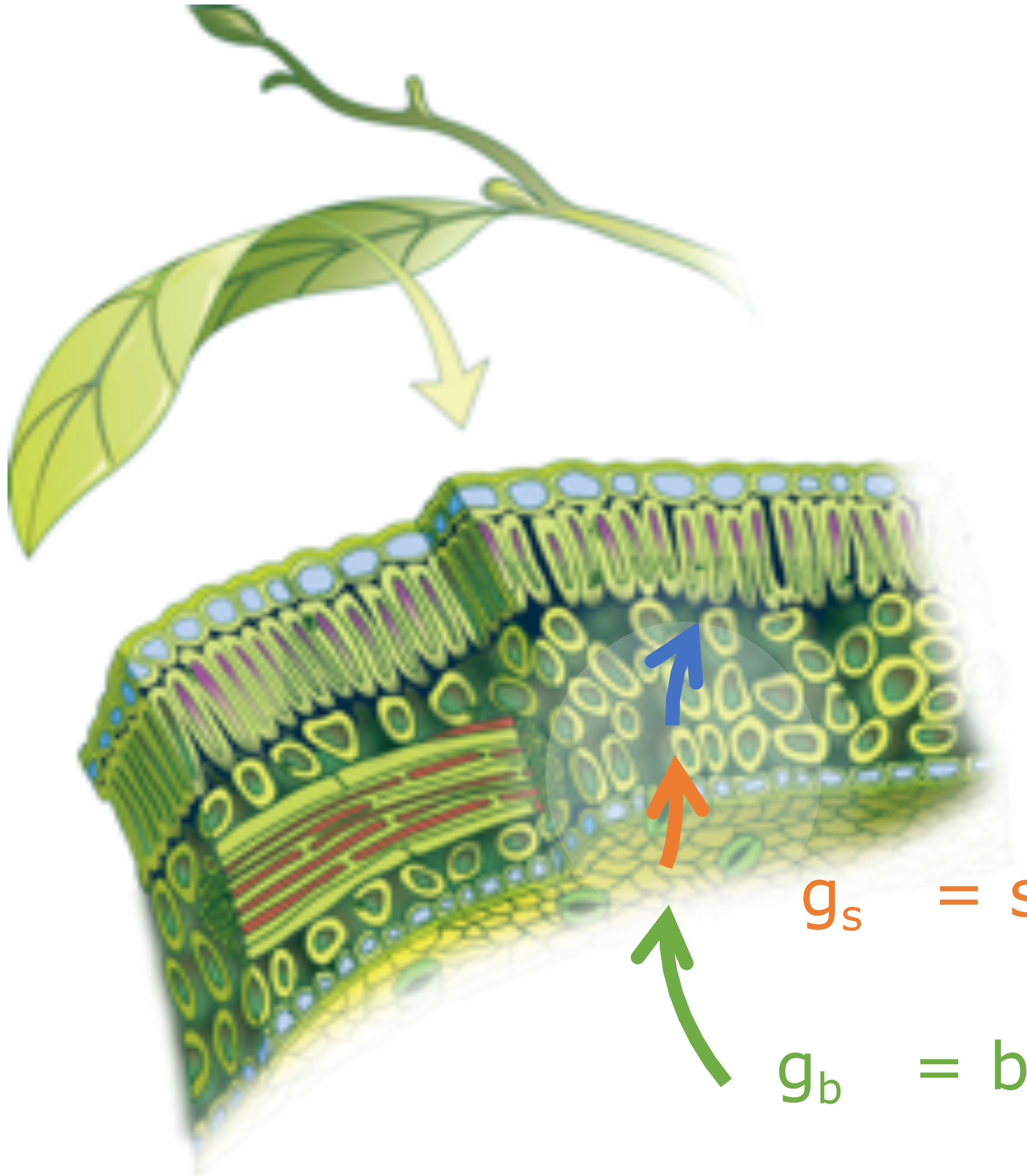
SIB4 Validation (Linda)



g_i = mesophyll diffusion + biochemical activity

g_s = stomatal conductance

g_b = boundary layer conductance

GPP

Ball-Berry

Vmax

$$Fcos = [COS]\left[\frac{1.56}{g_b} + \frac{1.94}{g_s} + \frac{1}{g_i}\right]^{-1}$$

Berry et al.: 2013

COS-OCS

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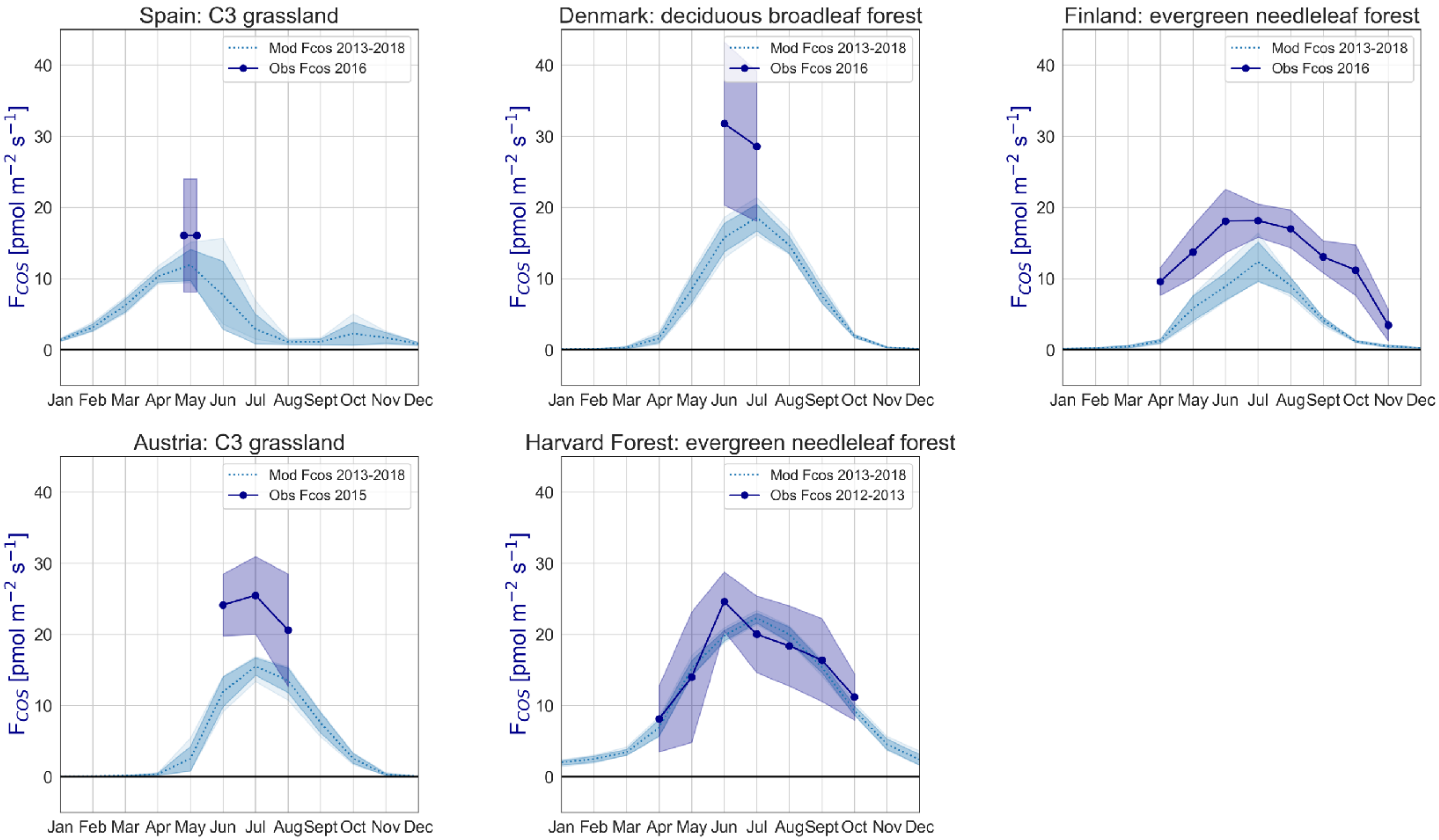

Data: Spielmann et al., 2019; Commane et al., 2015; Kooijmans et al., 2019/Vesala et al., submitted

COS-OCS

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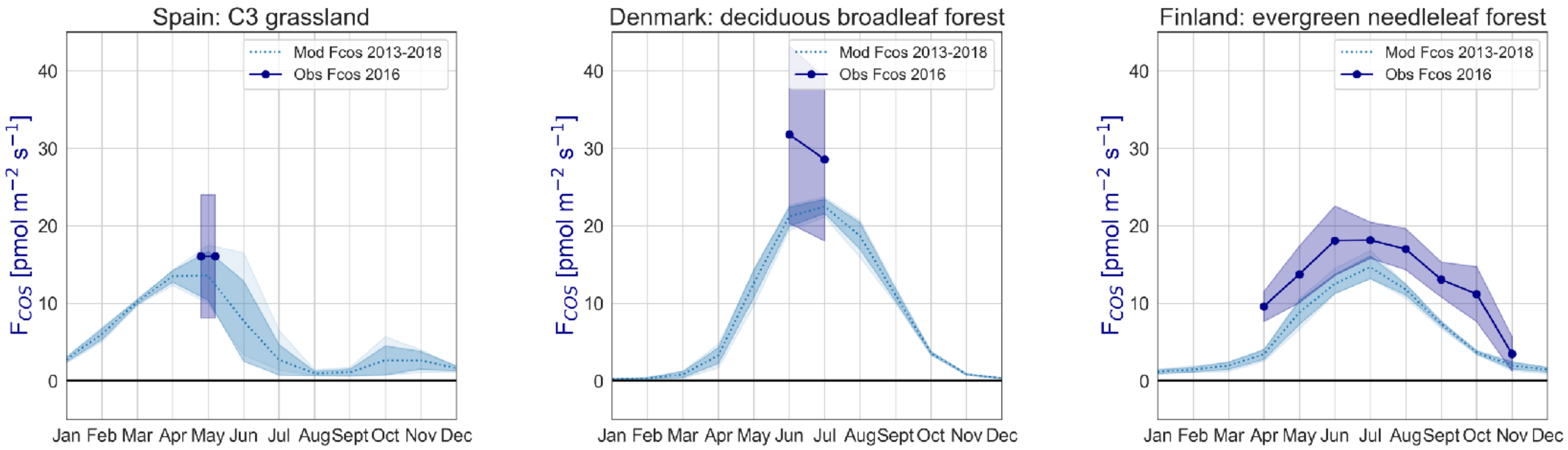
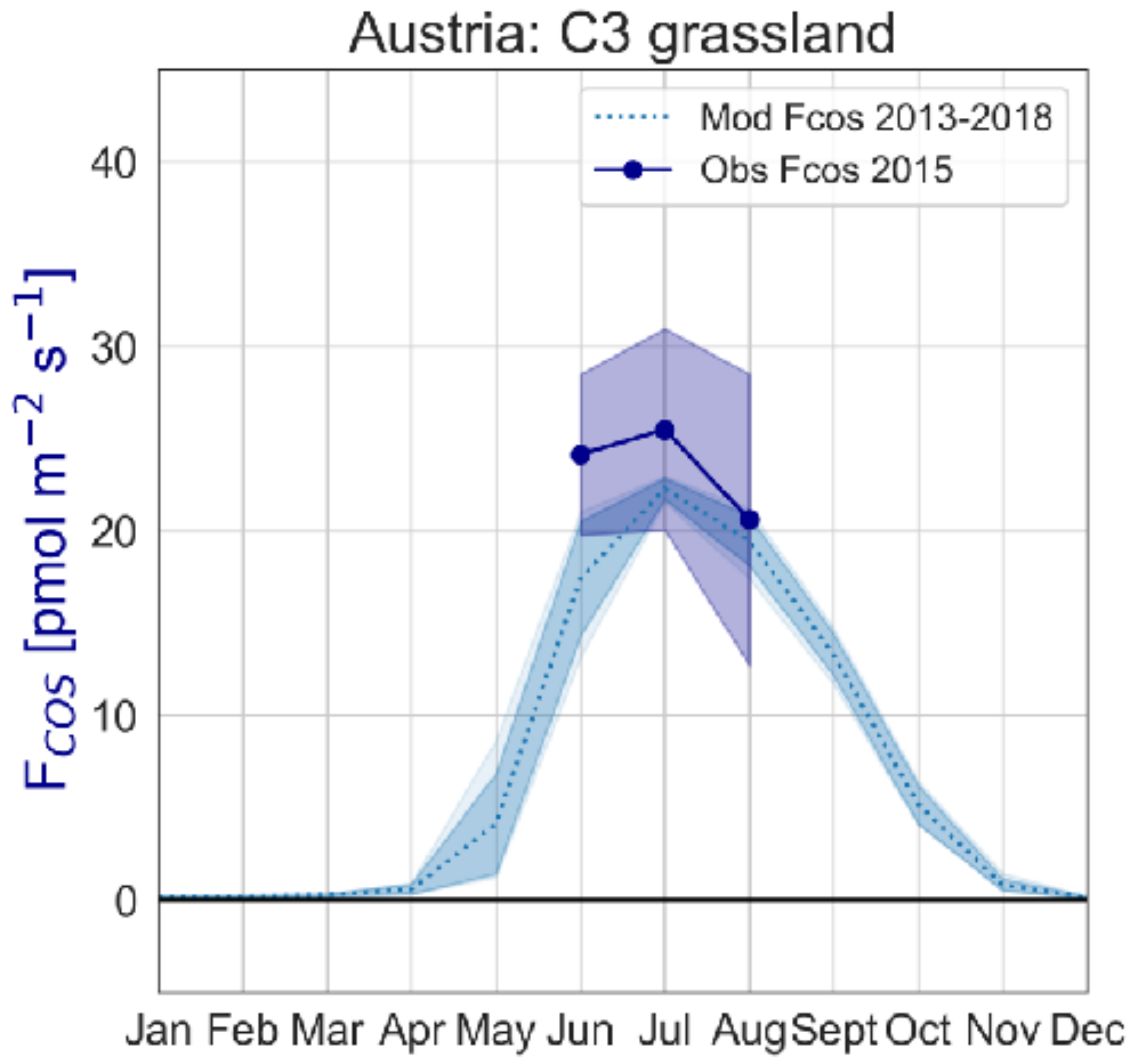
COS-OCS





Update in COS conductance calculations: extra temperature-dependence in g_i removed (has no effect on GPP)

Ian Baker, personal communication

Data: Spielmann et al., 2019; Commane et al., 2015; Kooijmans et al., 2019/Vesala et al., submitted

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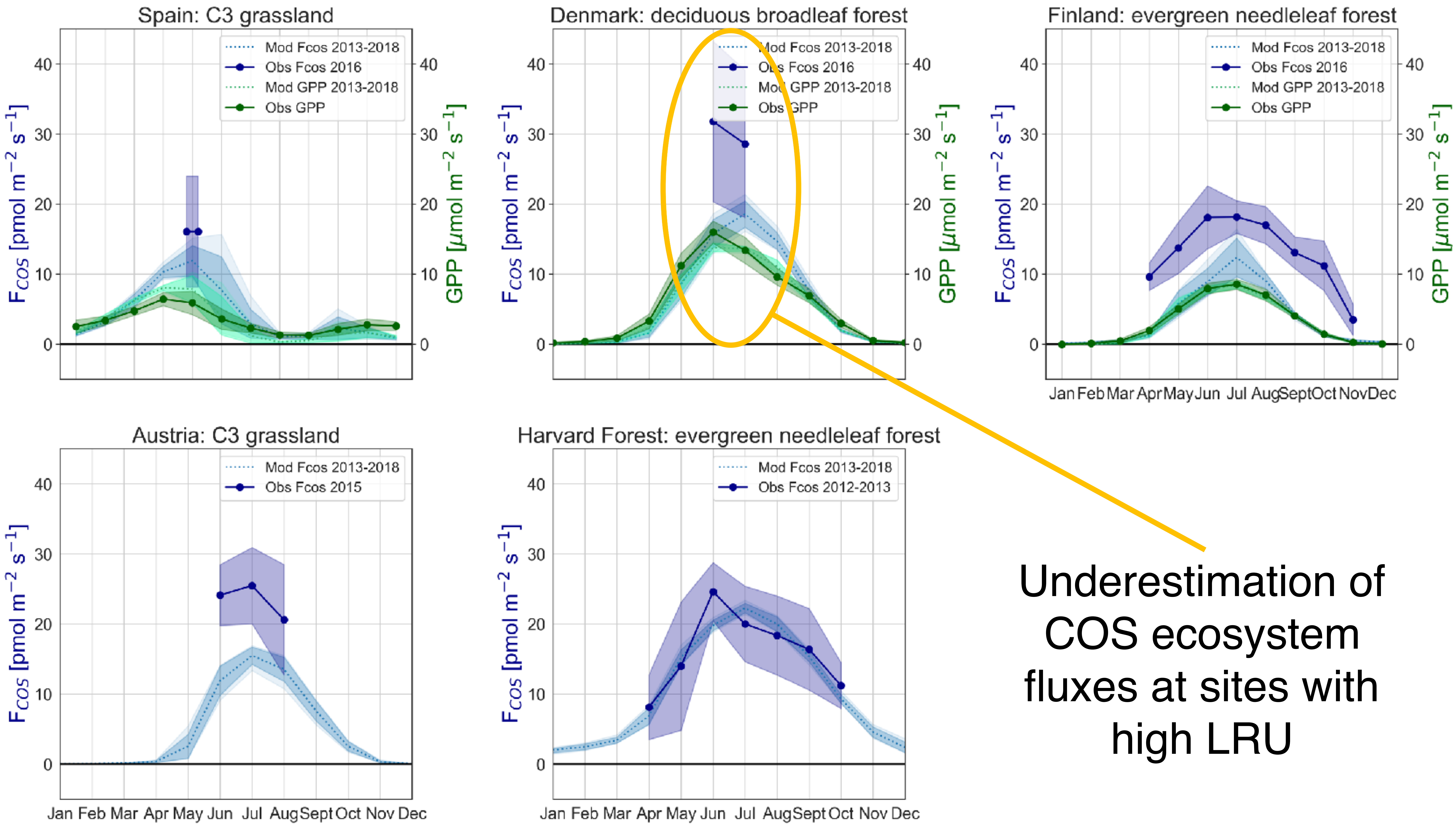
COS-OCS



Underestimation of COS ecosystem fluxes at sites with high LRU

Data: Spielmann et al., 2019; Commane et al., 2015; Kooijmans et al., 2019/Vesala et al., submitted

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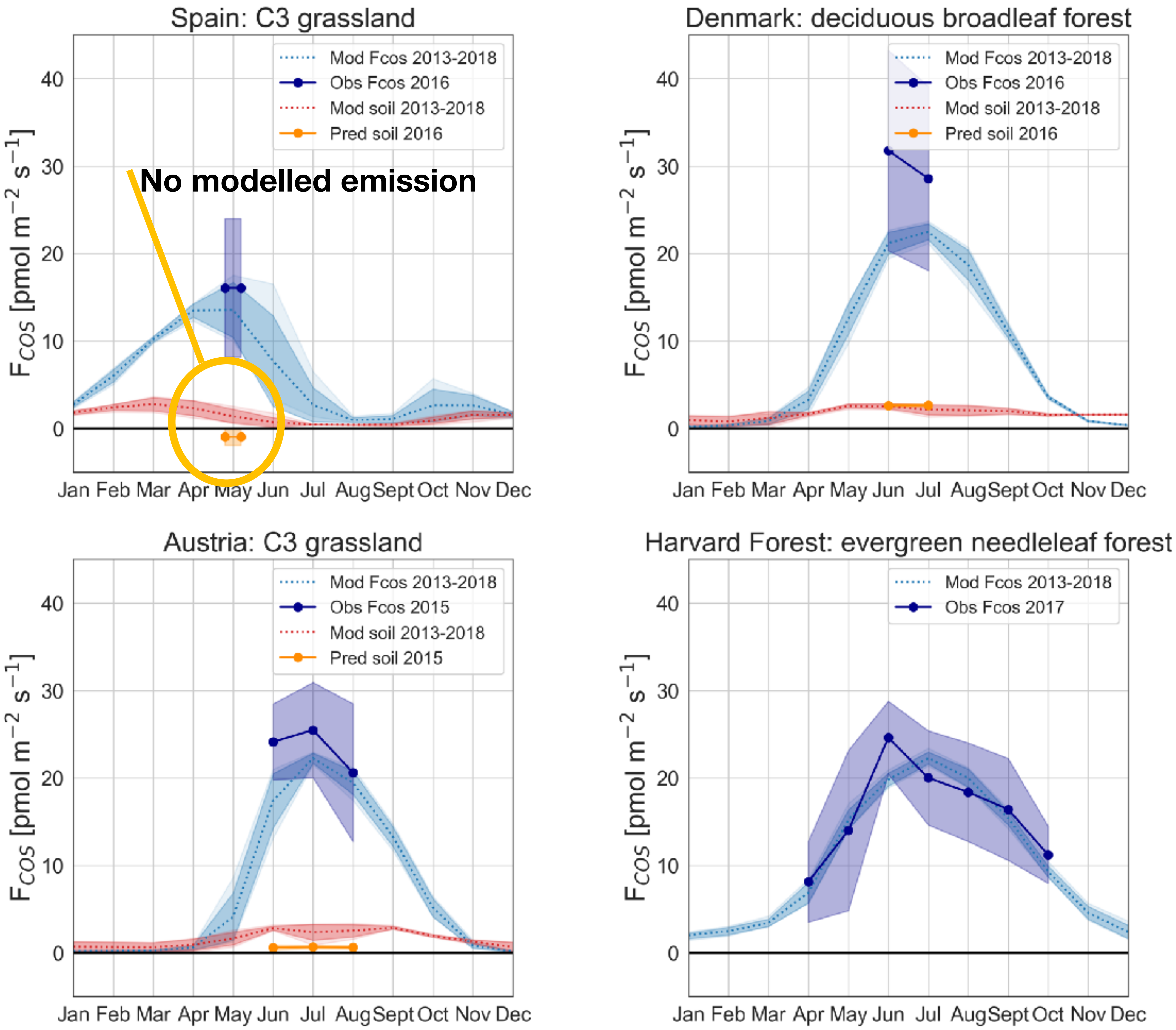

Implementation of mechanistic soil models (Sun et al., 2015, Ogee et al., 2016)
or empirical soil models (Mary Whelan)

Data: Spielmann et al., 2019; Commane et al., 2015; Kooijmans et al., 2019/Vesala et al., submitted

COS-OCS

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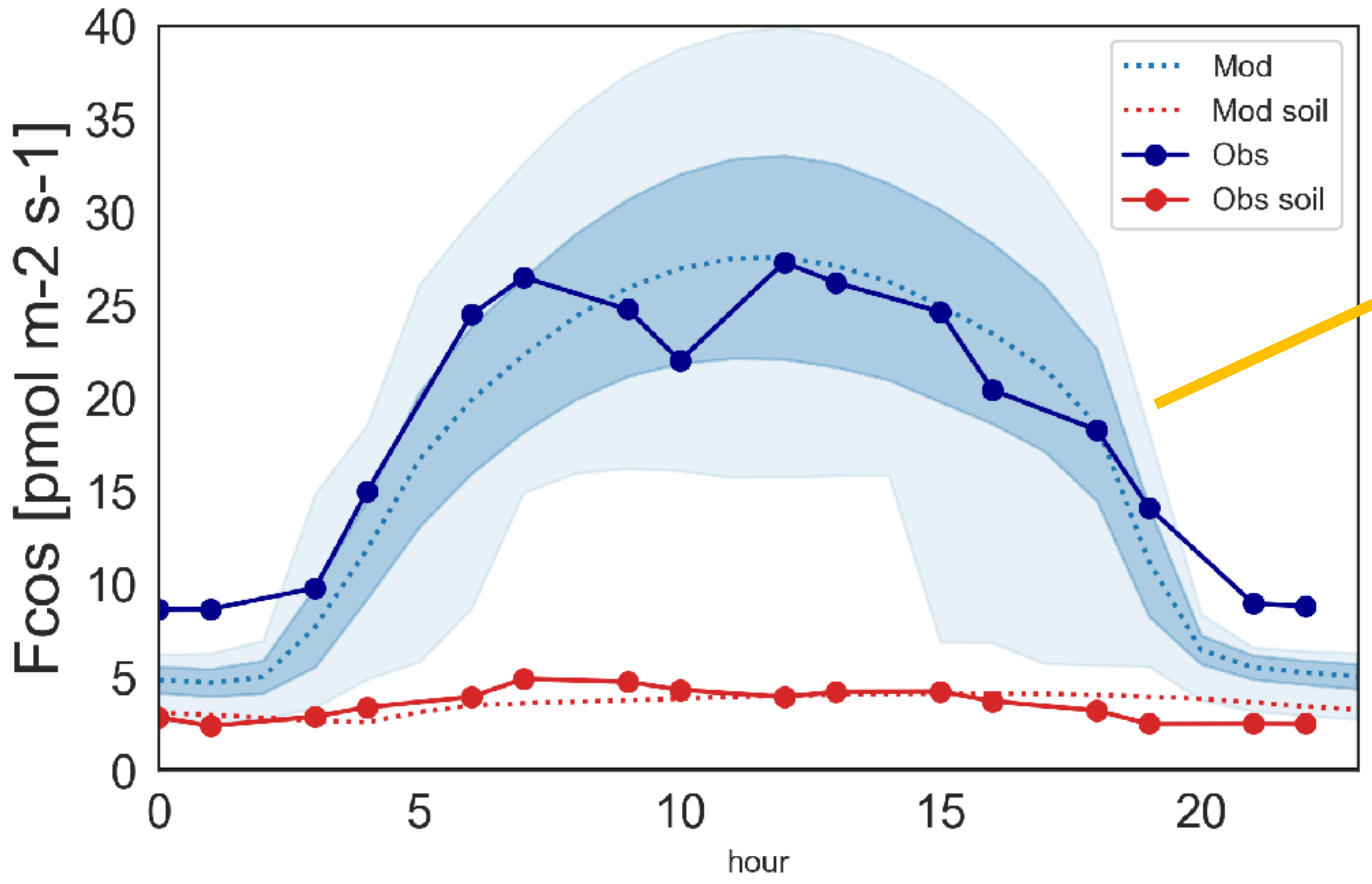
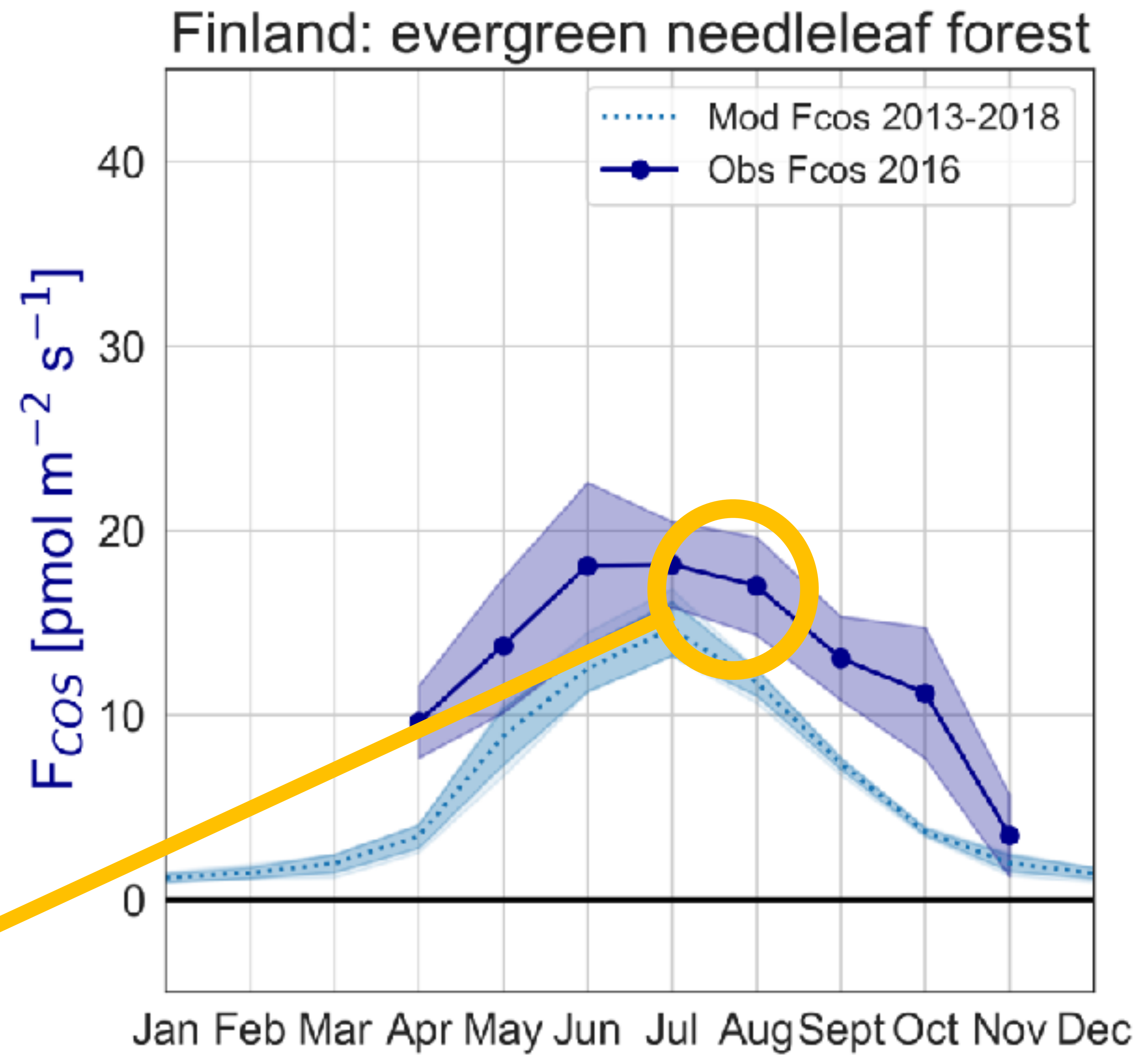
Underestimated SiB4 COS ecosystem flux partly due to underestimated nighttime uptake (= stomatal conductance)



Diurnal cycle of simulated and observed ecosystem and soil COS flux



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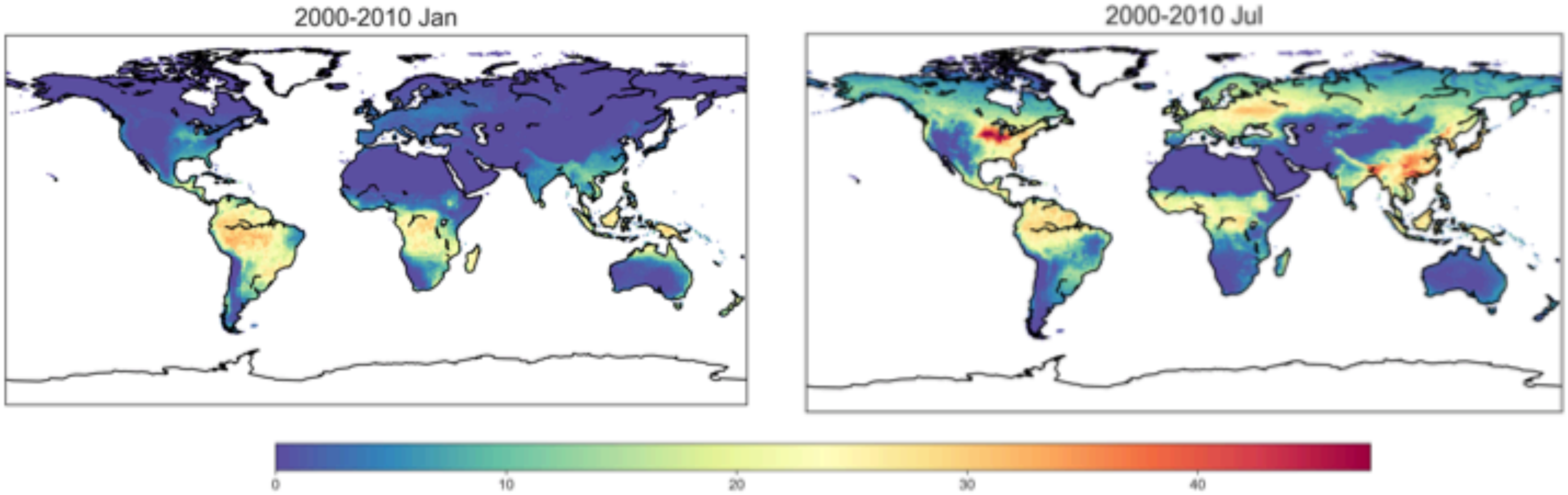
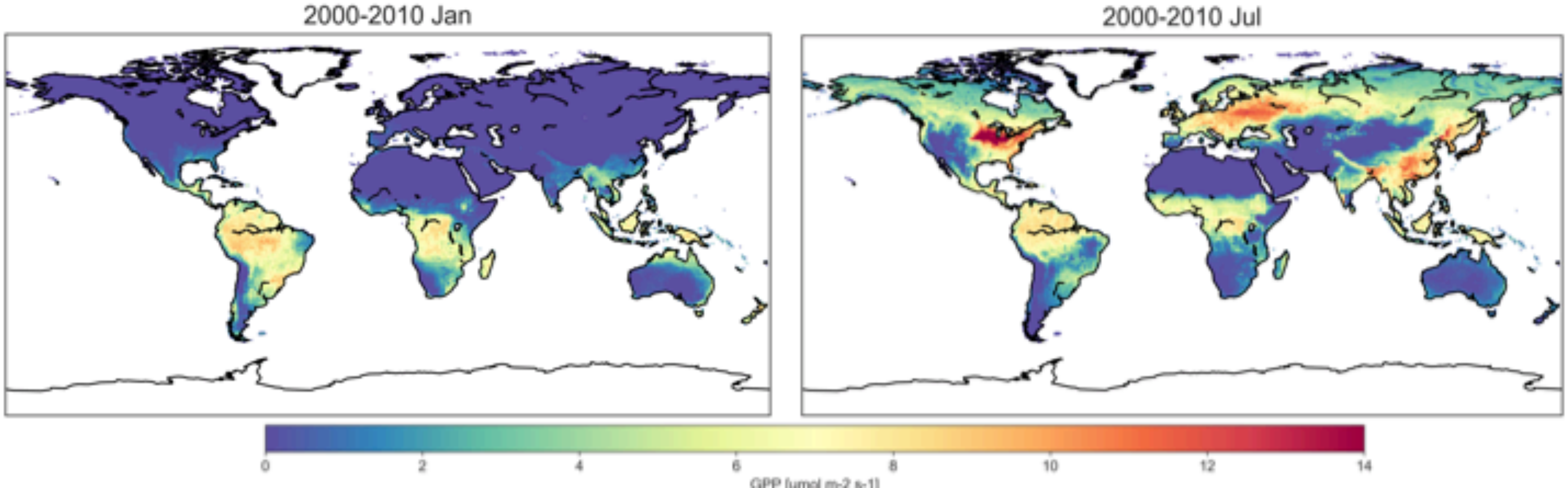
COS biosphere uptake (vegetation & soil)

1022 ± 12 GgS yr^{-1} (std over 2000-2010)



GPP



COS-OCS

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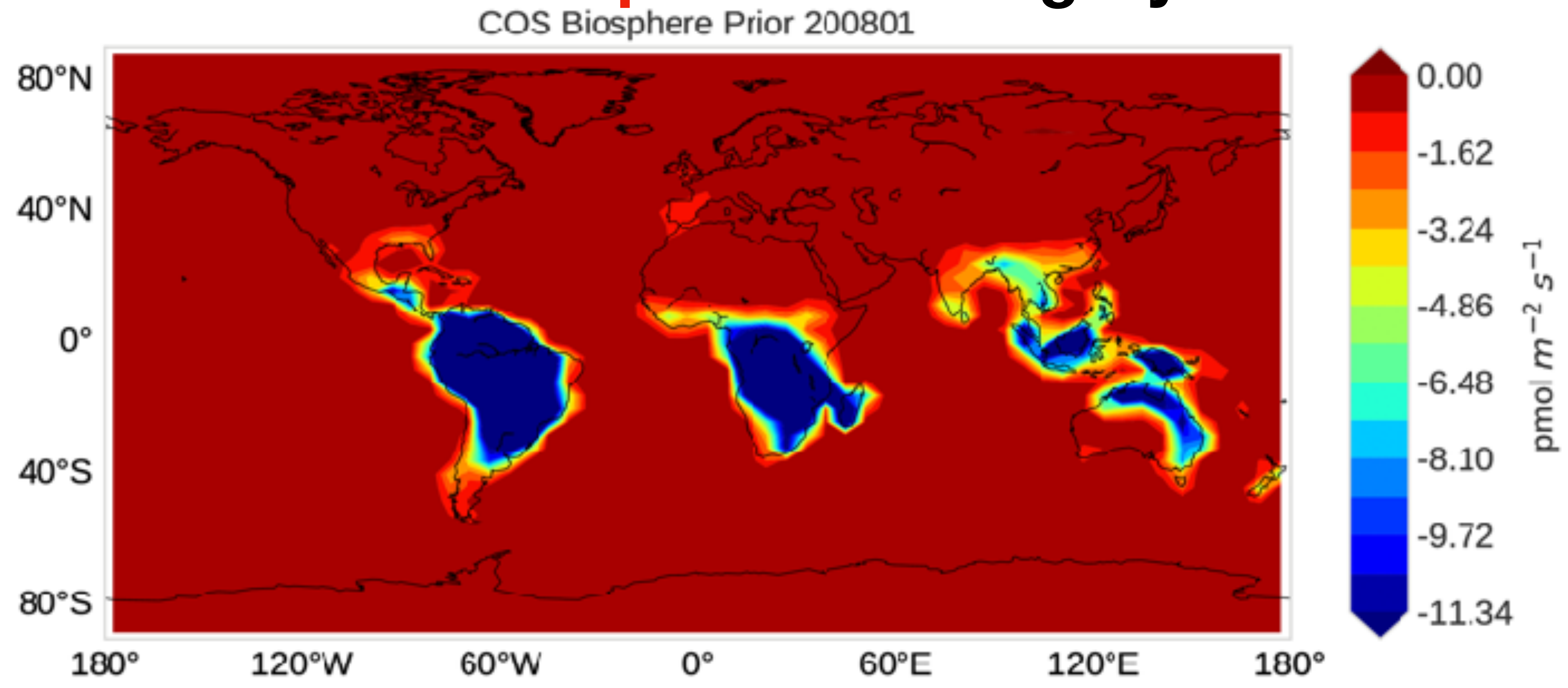
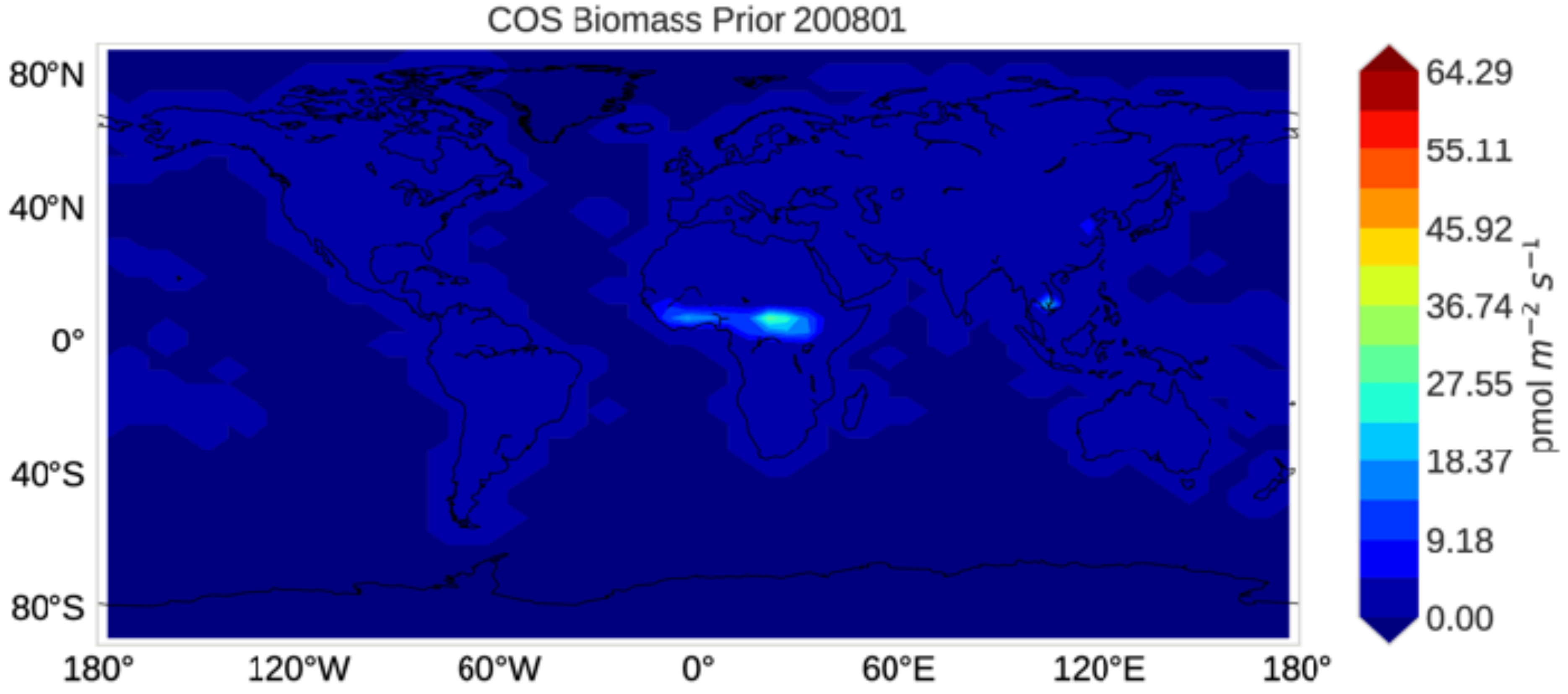
COS-OCS

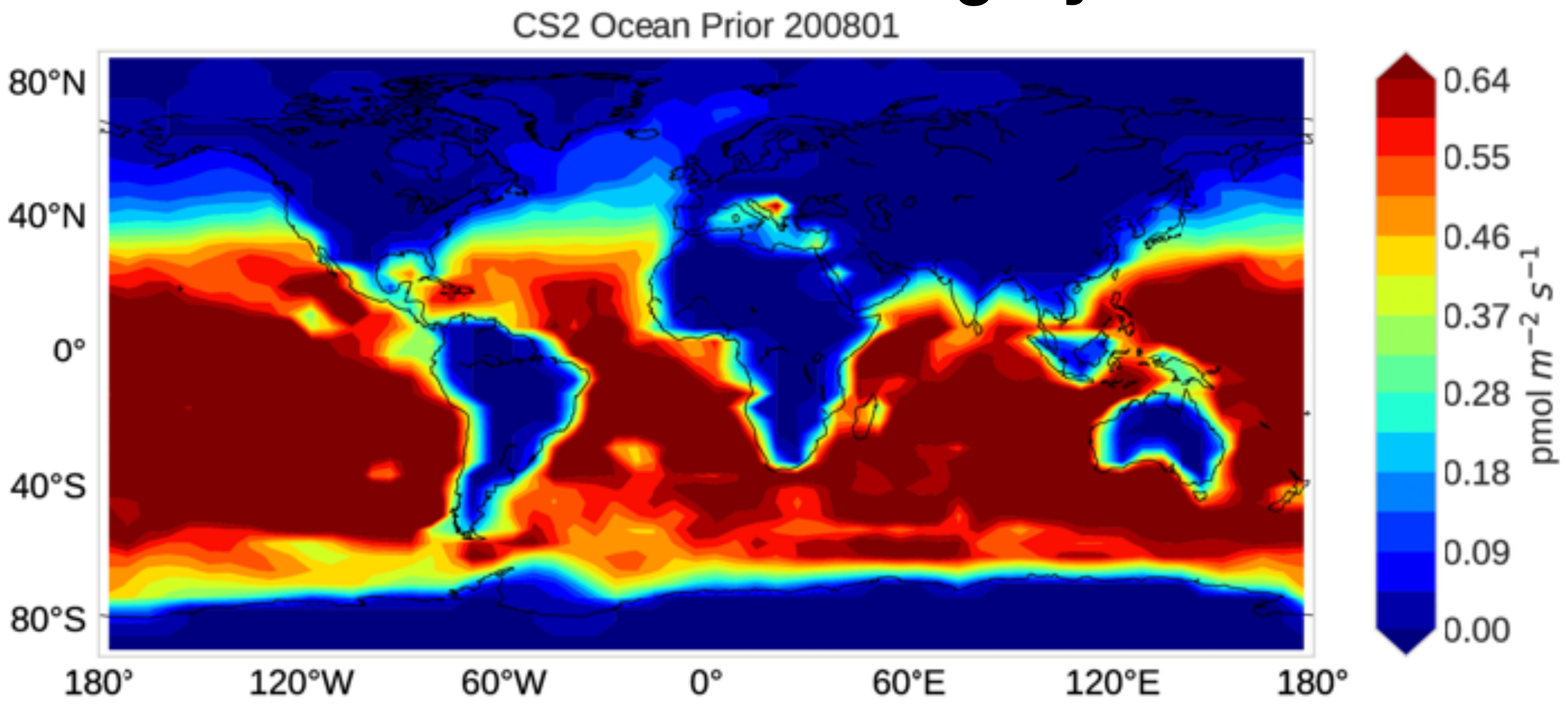
COS Biosphere: –898 TgS/yr “old” SIB4



COS Biomass burning: +136 TgS/yr GFED



CS2 Ocean: +81 TgS/yr Kettle 2002

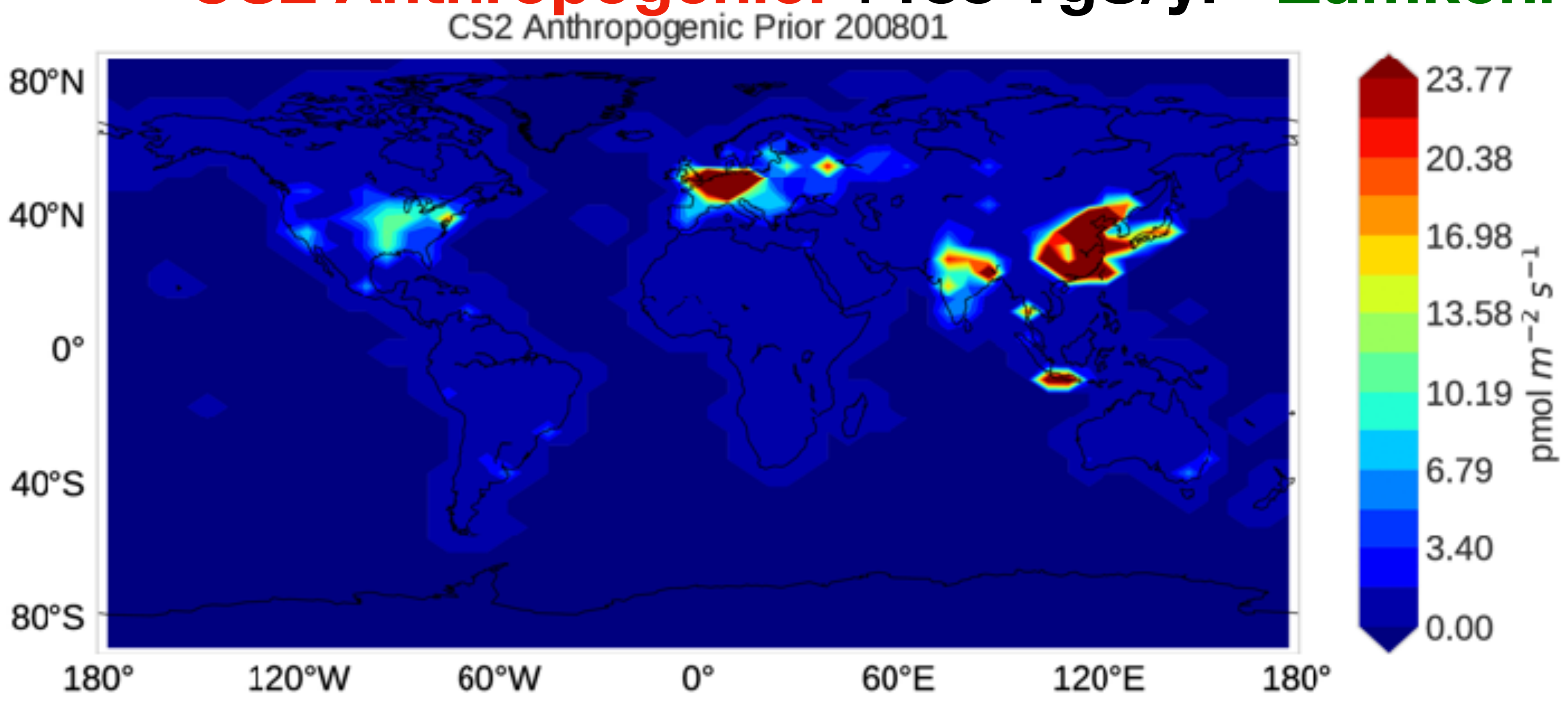


DMS Ocean+Land: +156 + 6 TgS/yr

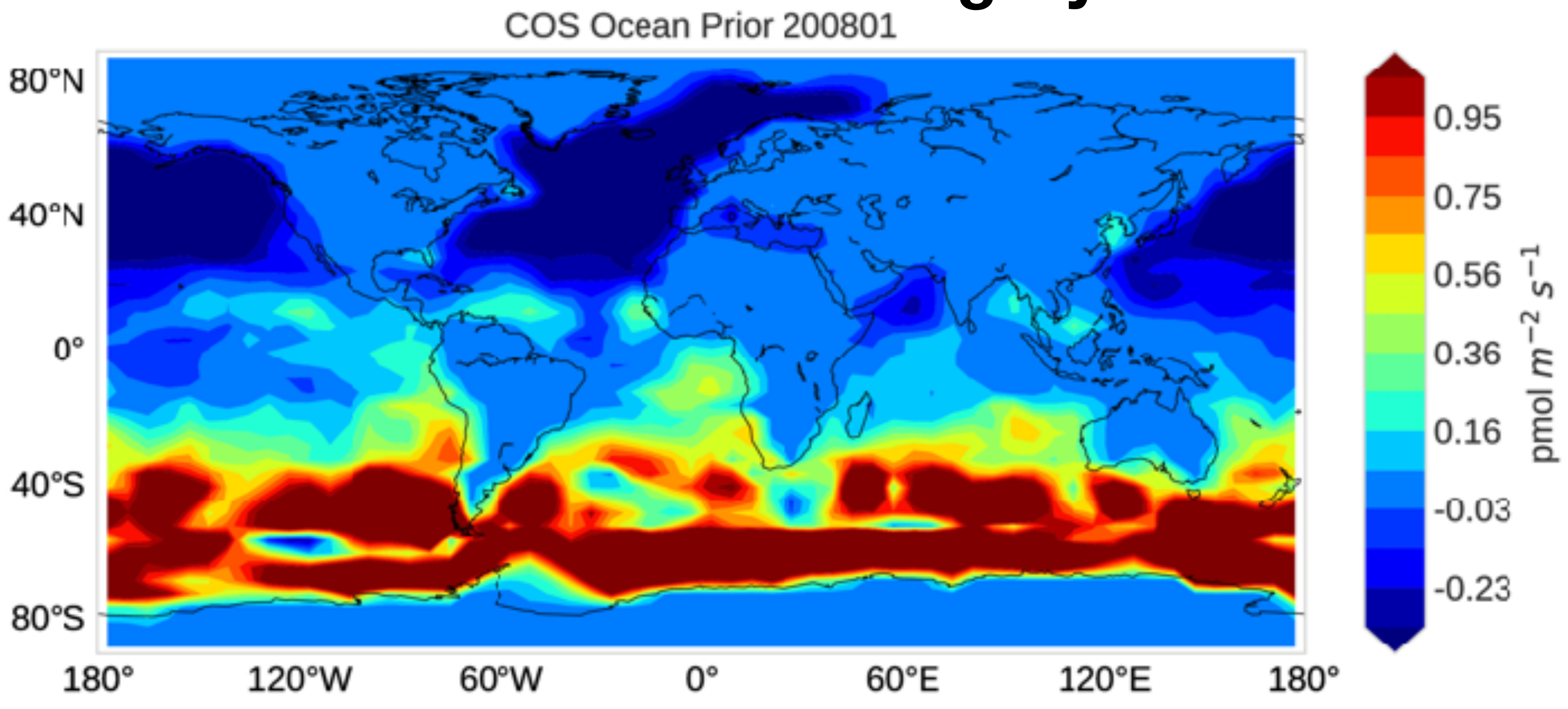
OH oxidation: –101 TgS/yr

Stratosphere: –40 TgS/yr

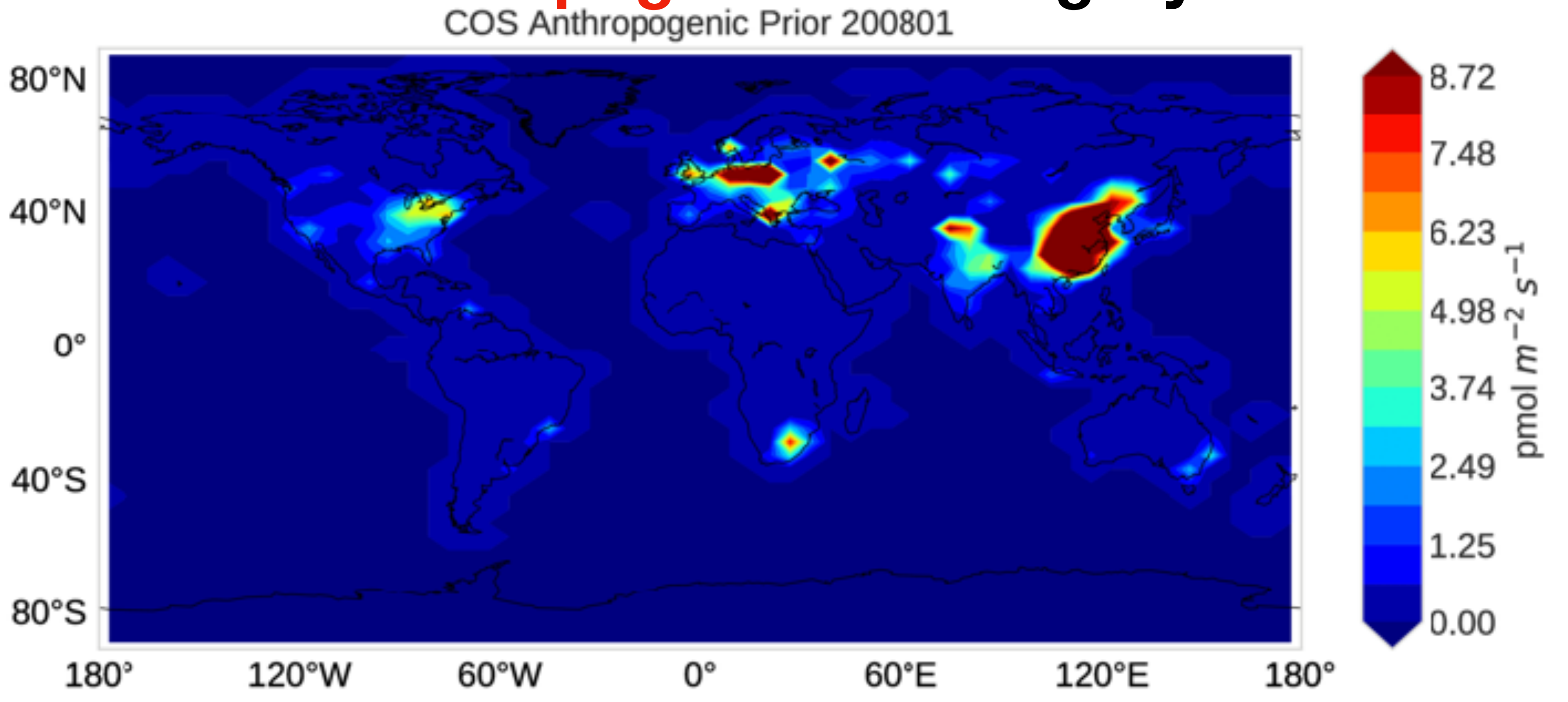
CS2 Anthropogenic: +188 TgS/yr Zumkehr



COS Ocean: +40 TgS/yr Kettle 2002



COS Anthropogenic: +155 TgS/yr Zumkehr



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COS-OCS

COS Global Budget (Gg S /year)	Berry2013	Prior of this study
Direct COS flux from oceans	39	40
Indirect COS flux as CS2 from oceans	81	81
Indirect COS flux as DMS from oceans	156	156
Direct anthropogenic flux	64	155
Indirect anthropogenic flux from CS2	116	188
Indirect anthropogenic flux from DMS	1	6
Biomass burning	136	136
Additional ocean flux	600	-
Anoxic soils and wetlands	-	-
Total Sources	1193	762
Destruction by OH	-101	-101[$]
Destruction by O	-	-
Stratospheric photolysis	-	-40[#]
Uptake by plants	-738	-898*
Uptake by soil	-355	
Total Sinks	-1194	-1039
Net total	-2	-277

[$]Using OH-climatology Spivakovsky (2000) multiplied by 0.92

[#]Independent from LUT J-values (TUV)

***Includes soil uptake based on scaled Rh (still uncorrected SIB4)**

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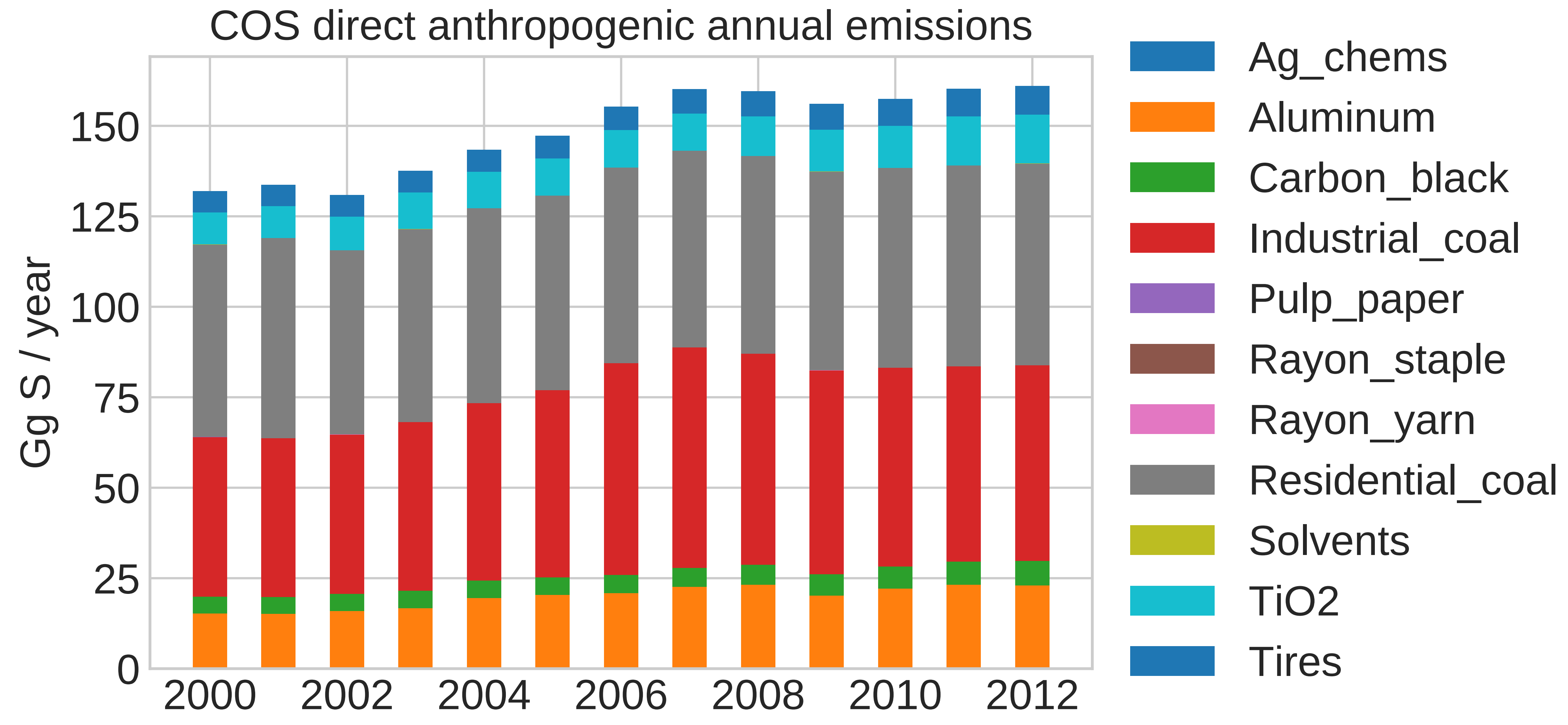
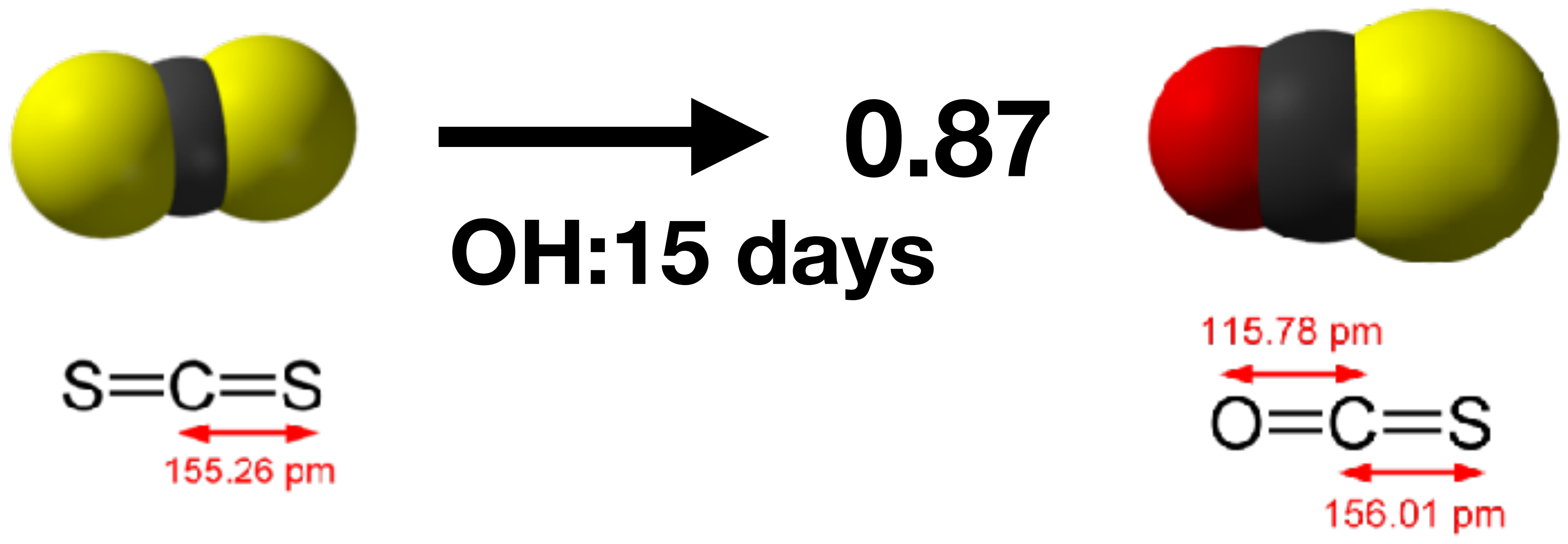
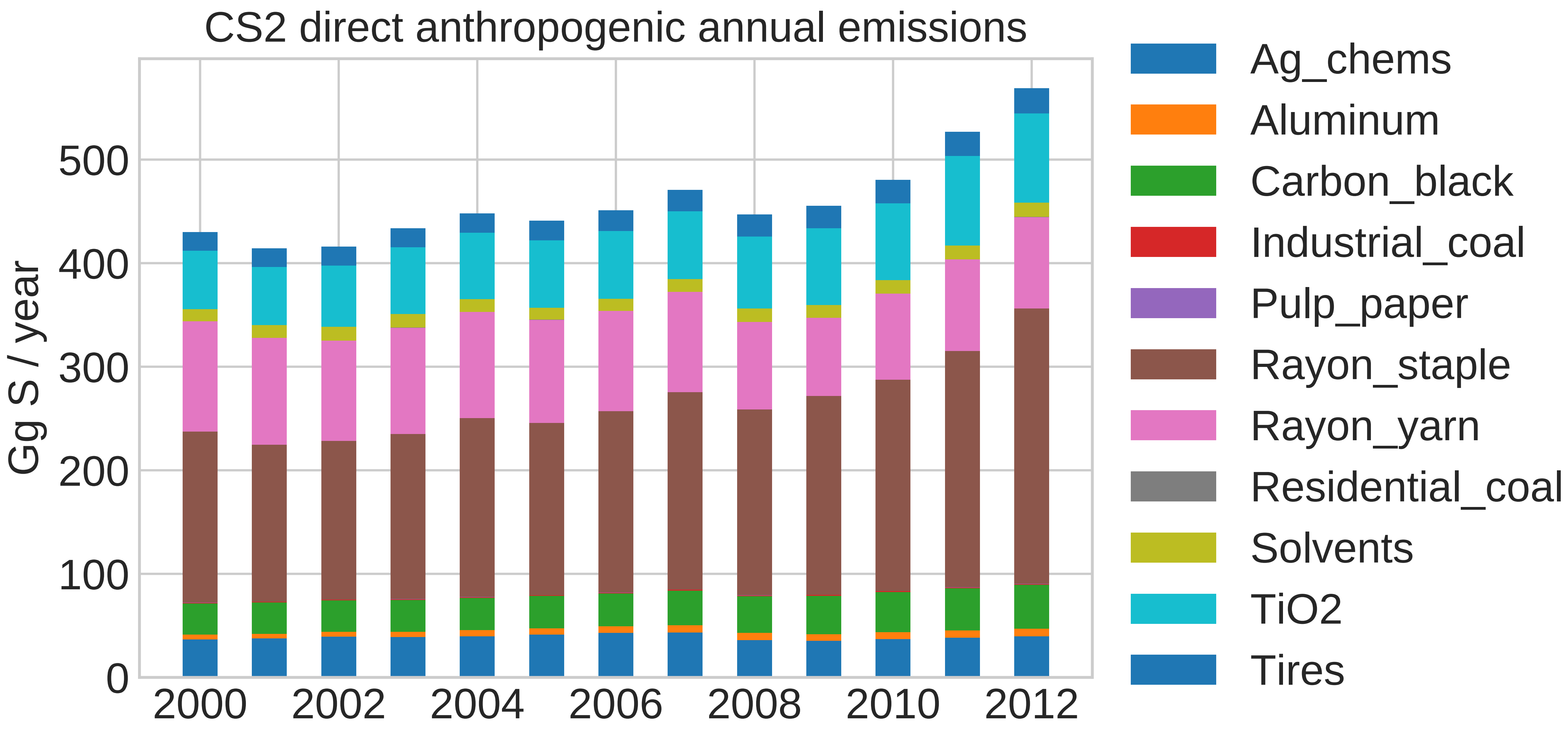
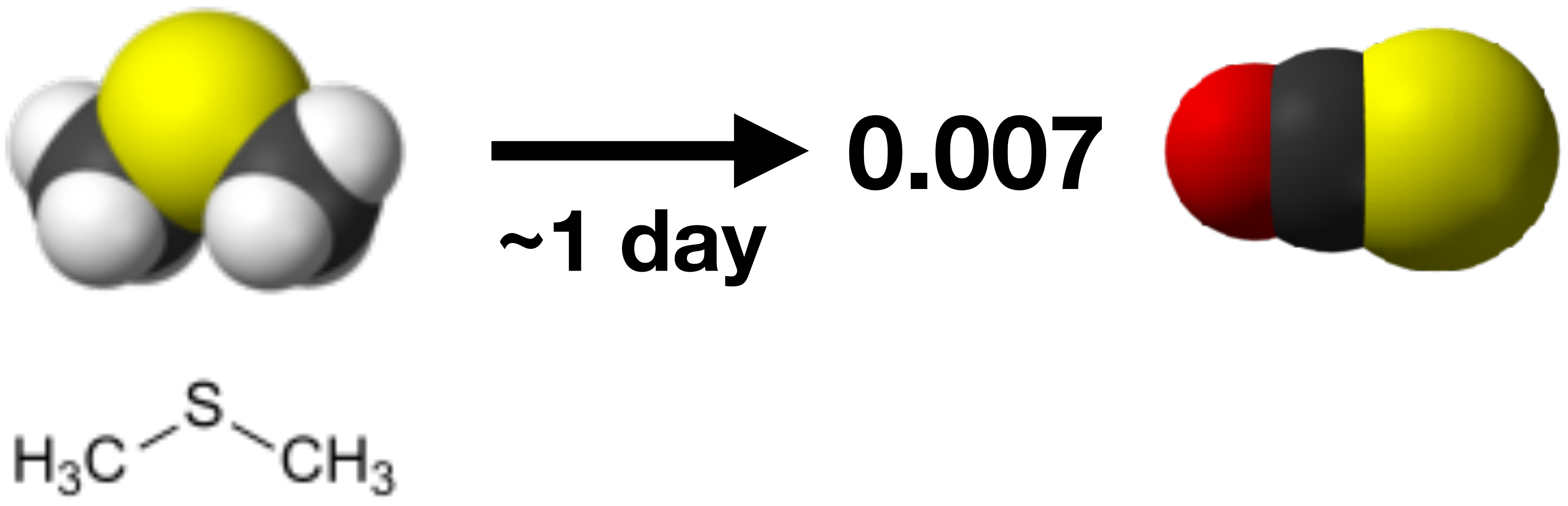
Anthropogenic Emissions COS/CS_2









Zumkehr et al., 2018

COS-OCS

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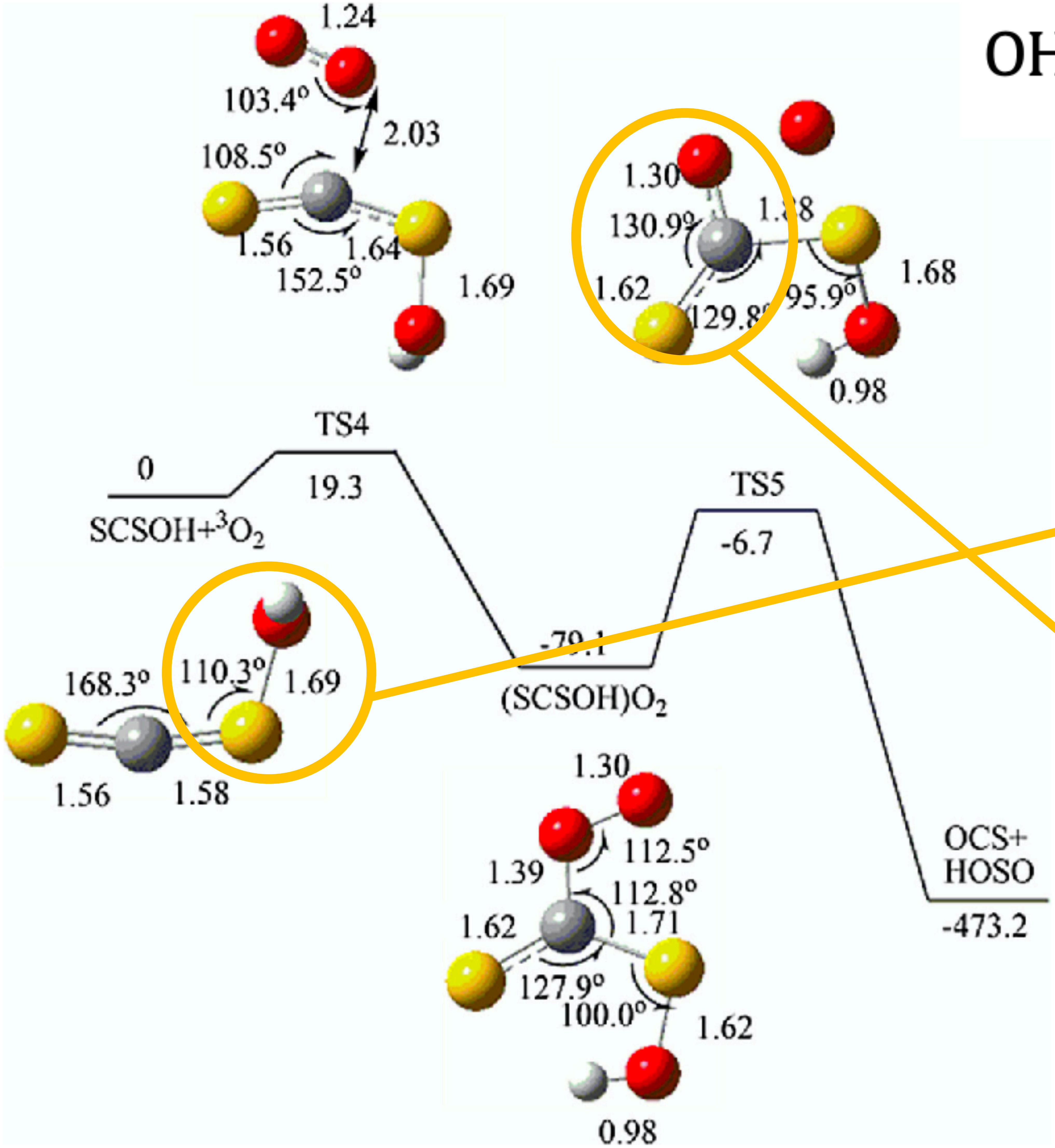
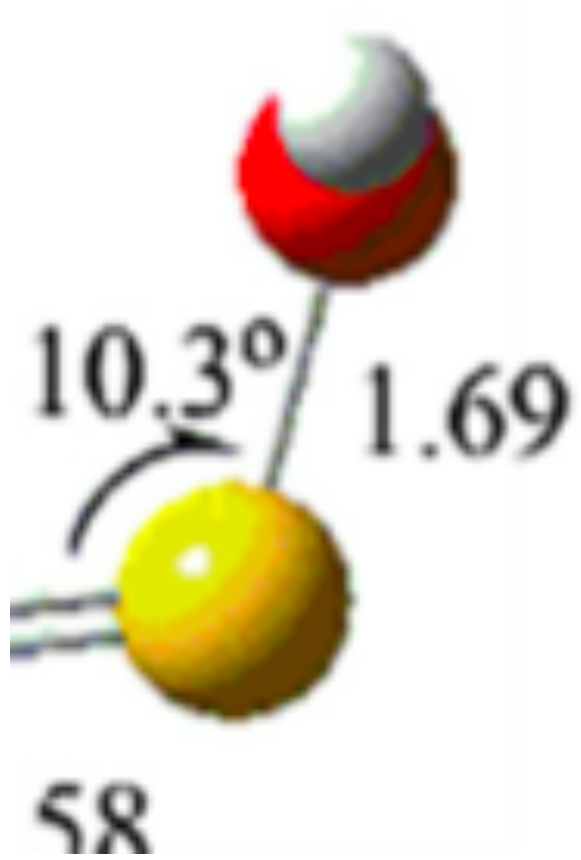
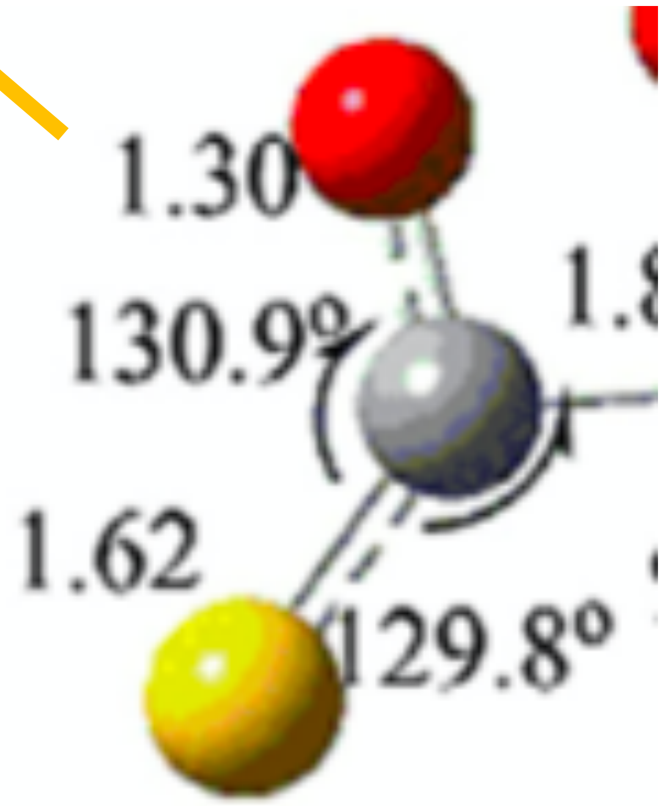
Note on fractionation ($^{34}S/^{32}S$)

$$OH + CS_2 \xrightarrow{k_1} OCS + SH$$





OH attaches easier on the ^{34}S



Which then does not end up in COS

Thus “34S”-depleted COS w.r.t CS2 might be expected

Zeng et al., Chemical Physics Letters 669 (2017) 43–48

COS-OCS

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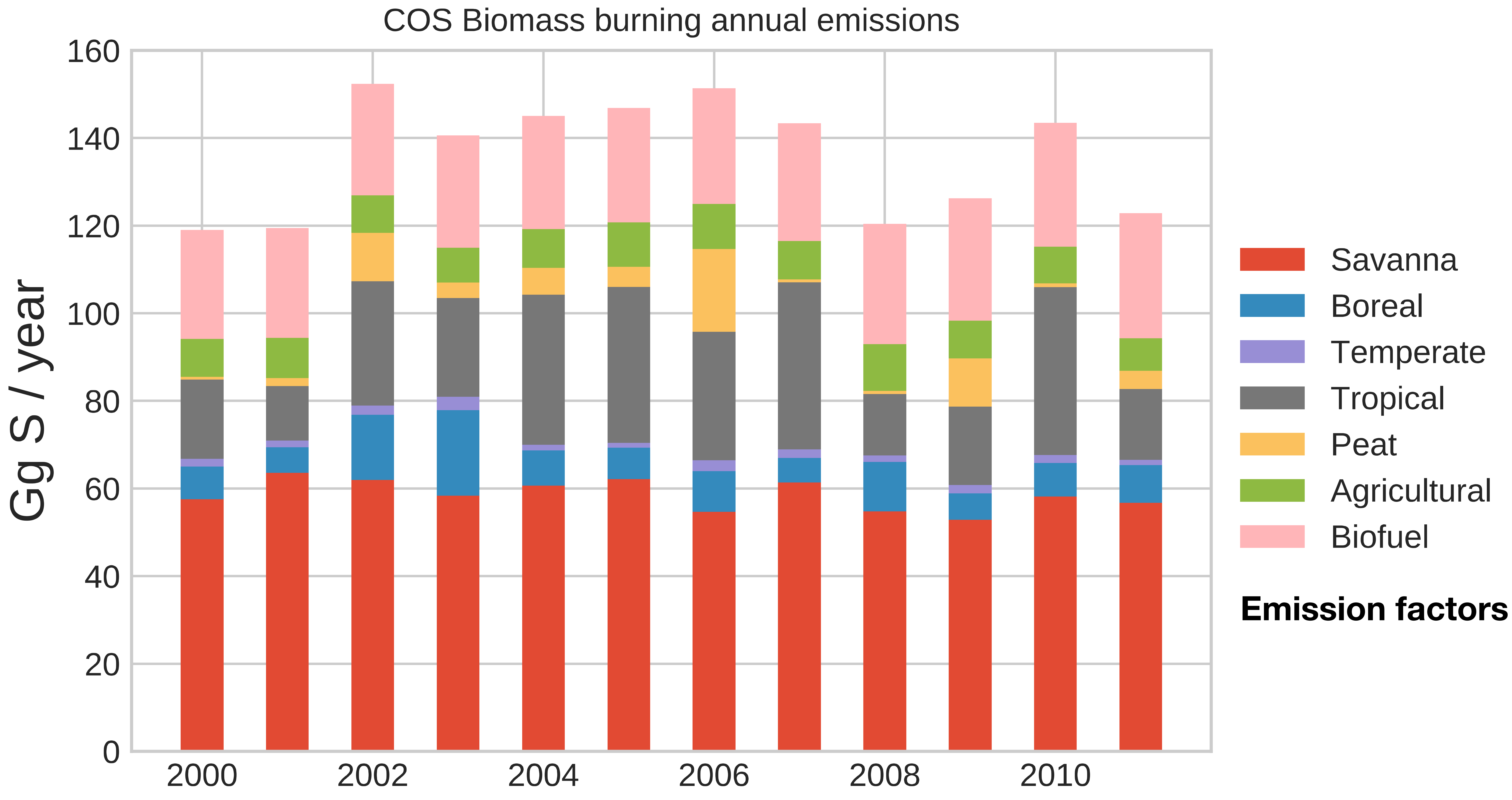
Biomass burning emissions



GFED4.1

CEDS

Emission factors from Andrea, ACP, 2019

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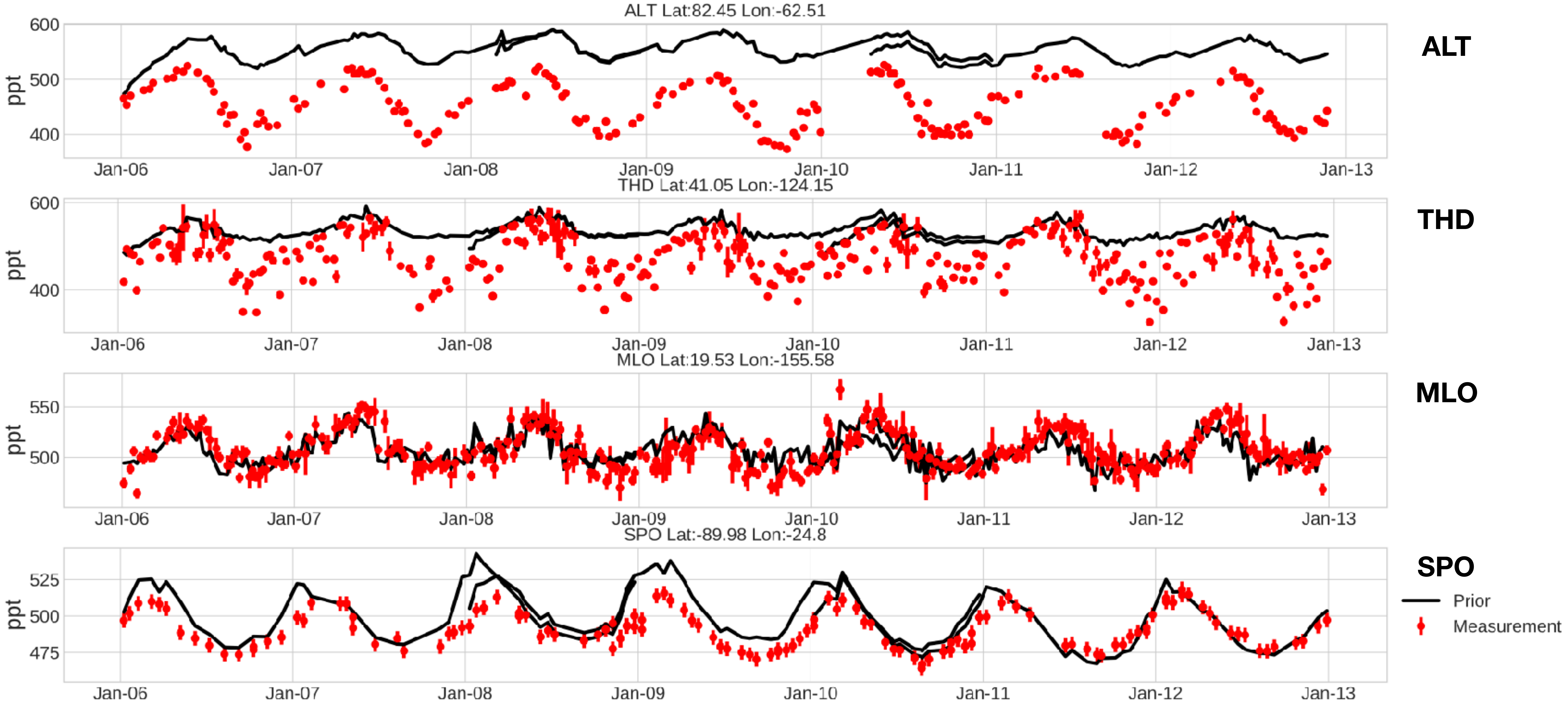
Added 277 Gg S/yr globally (land + ocean) to close the budget (3 overlapping simulations)



COS-OCS

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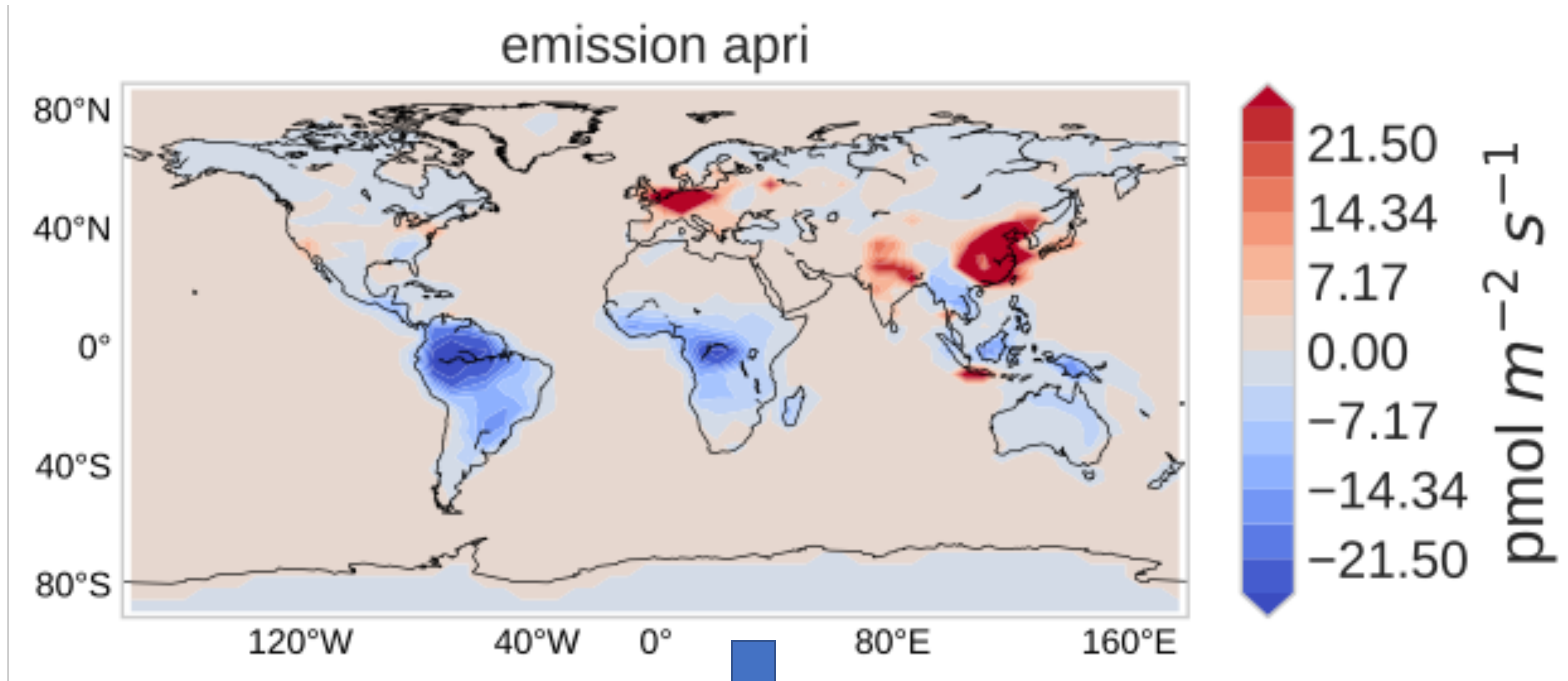
Mathematical framework to optimise emissions



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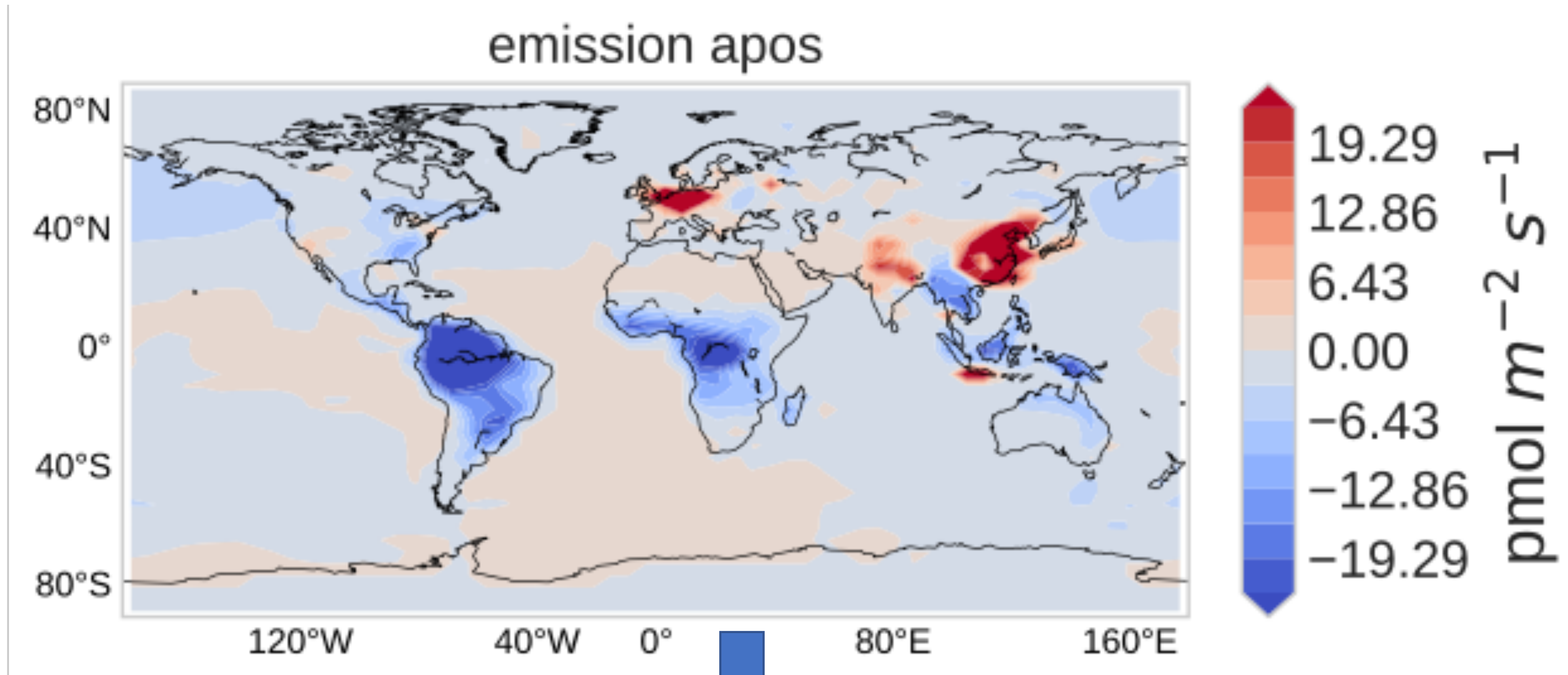
NOAA Observational constraints



+

HIPPO aircraft for validation

Input

Output

TM5-4DVAR

Cost function

$$\boldsymbol{J}(x) = \frac{1}{2}\left(x - x^b\right)^T \boldsymbol{B}^{-1}\left(x - x^b\right) + \frac{1}{2}\left(\boldsymbol{H}x - y\right)^T \boldsymbol{R}^{-1}\left(\boldsymbol{H}x - y\right)$$

Forward Model

$$y = \boldsymbol{H}(x)$$

Adjoint Model

$$x = \boldsymbol{H}^T(y)$$

Gradient of model

$$\nabla \boldsymbol{J}(x) = \boldsymbol{B}^{-1}\left(x - x^b\right) + \boldsymbol{H}^T \boldsymbol{R}^{-1}\left(\boldsymbol{H}x - y\right)$$

COS-OCS

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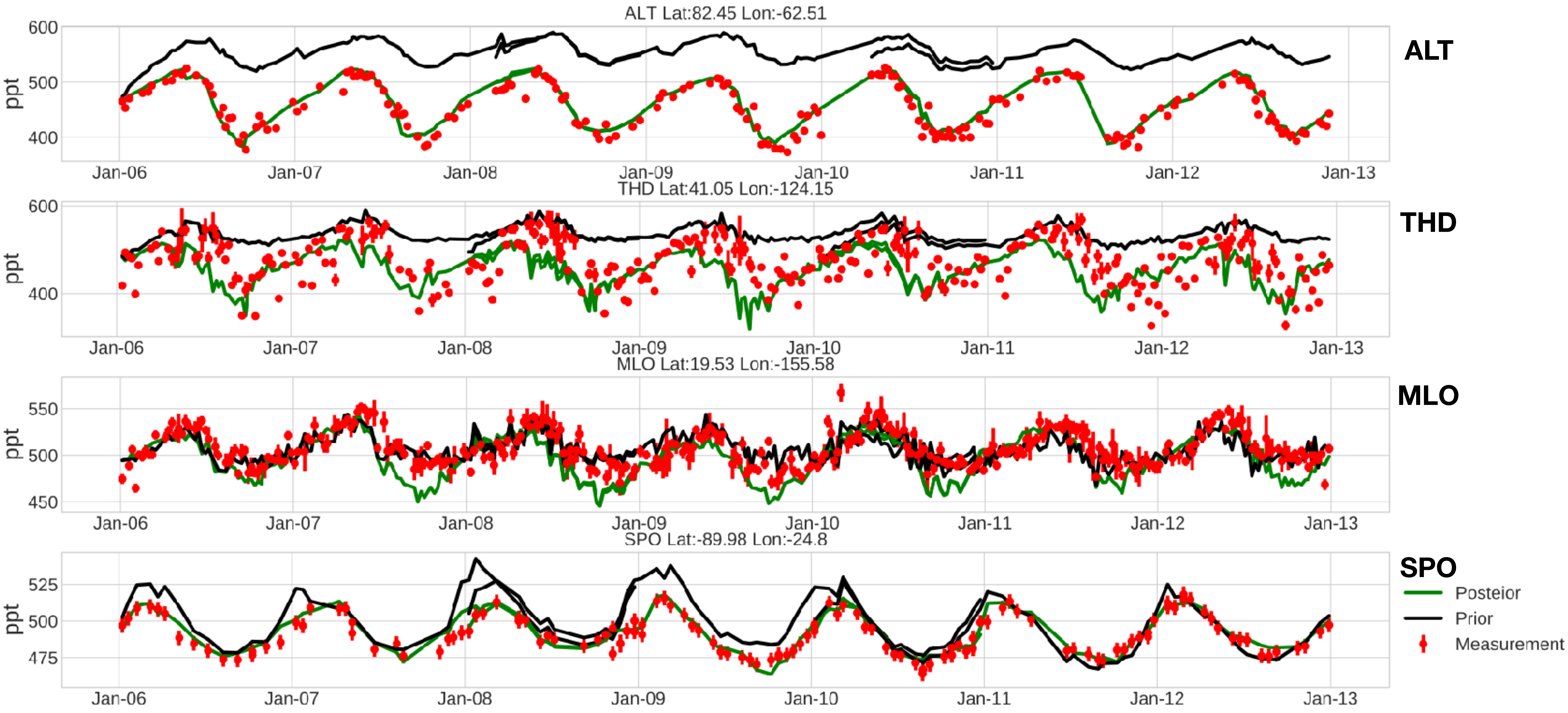
Depending on the settings, the fit with observations in mathematically improved



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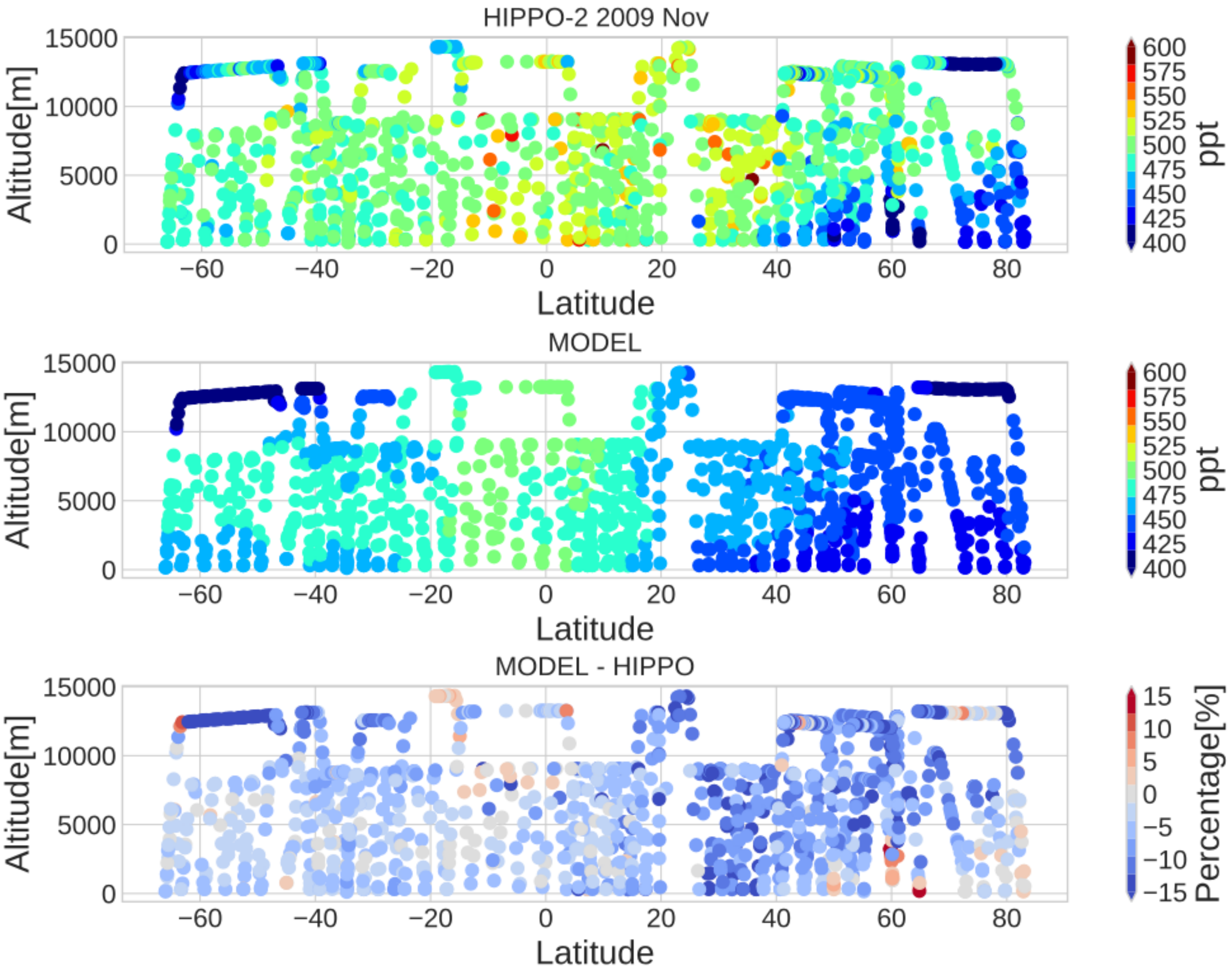

Better distribution

Model has low bias

COS-OCS

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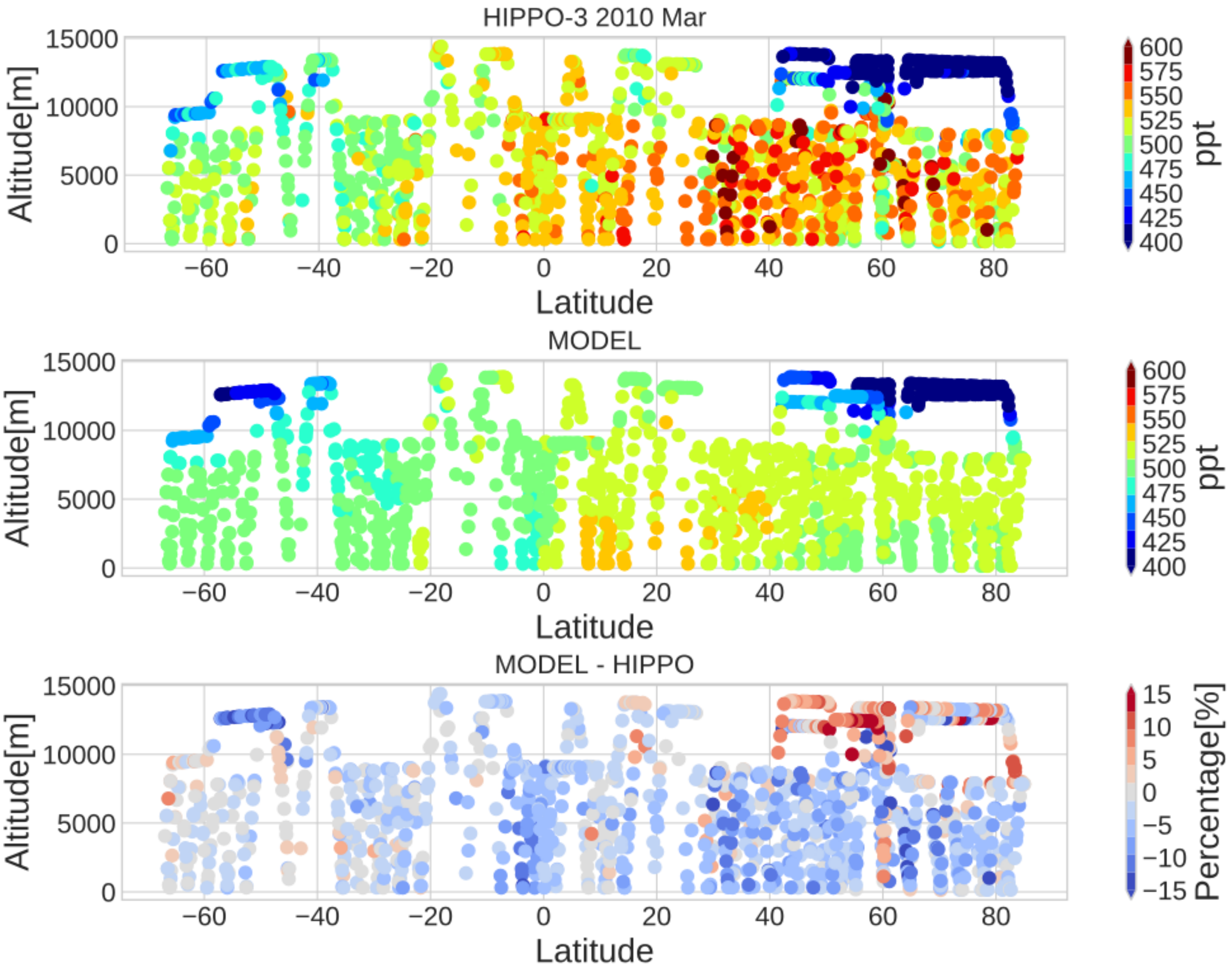
COS-OCS



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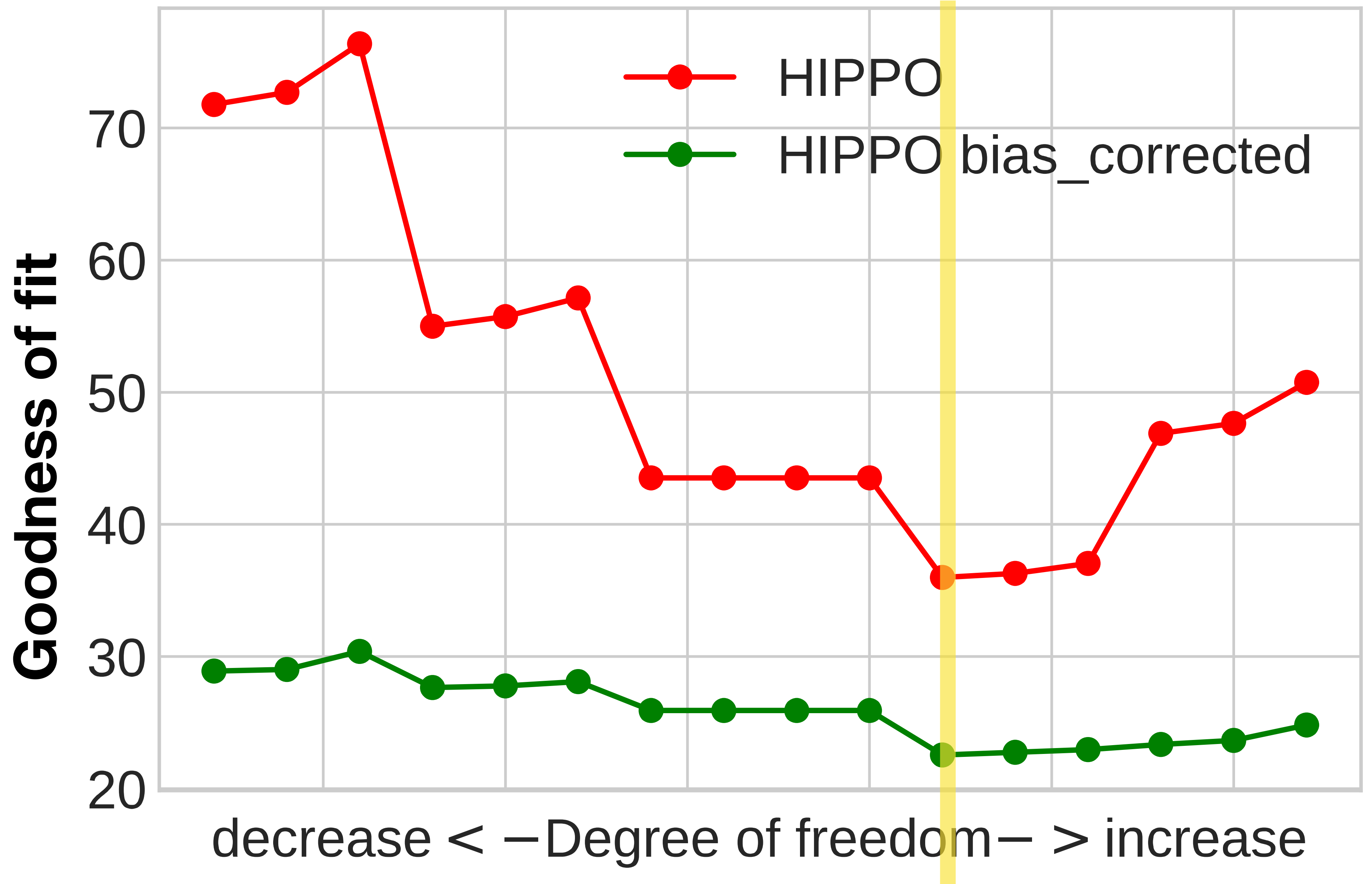
Determining the “optimal settings”

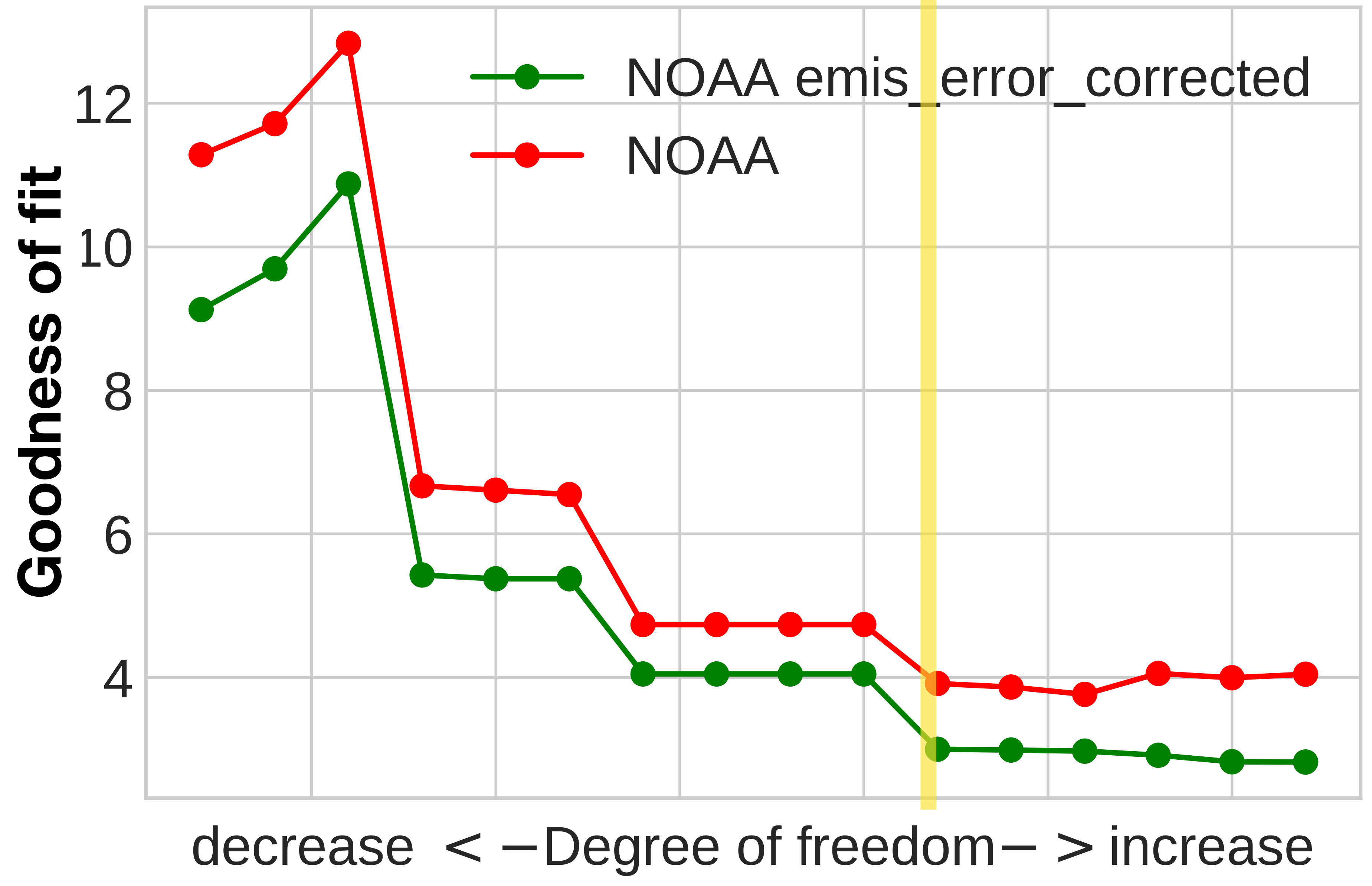
COS-OCS

Degrees of freedom

Degrees of freedom	Test inversions	spatial corr [km]	temporal corr [month]
High	1	1000	5.5
	2	1000	9.5
	3	1000	12
	4	4000	5.5
	5	4000	9.5
	6	4000	12
	7	6000	5.5
	8	6000	7
	9	6000	9.5
	10	6000	12
	11	10000	5.5
	12	10000	9.5
	13	10000	12
	14	20000	5.5
	15	20000	9.5
Low	16	20000	12

Hippo not assimilated





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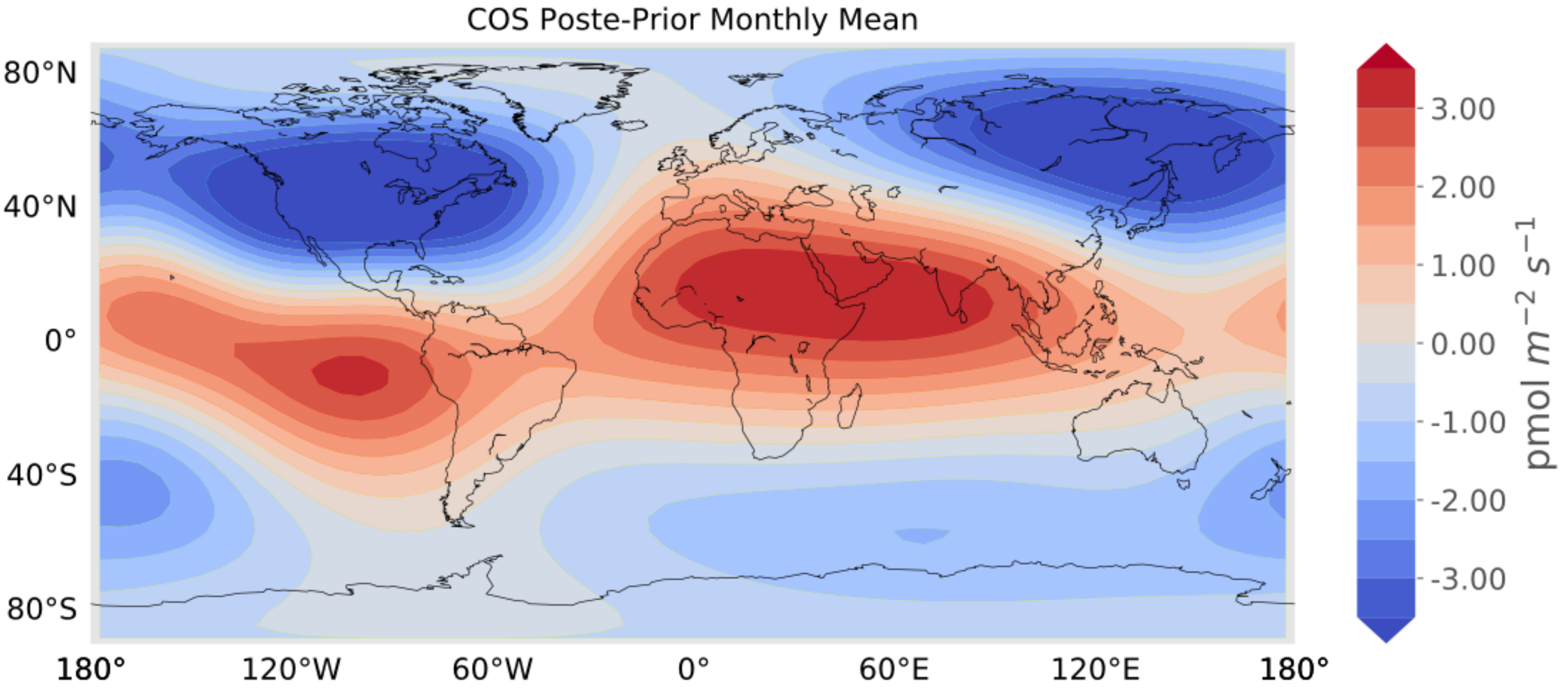
- We close the budget by adding 277 Gg S/year globally uniformly
- optimise this uniform monthly emission flux of 0.51 pmol m^{-2} s^{-1}
- Spatial correlation = 4000 km
- Temporal correlation = 9.5 months



COS-OCS

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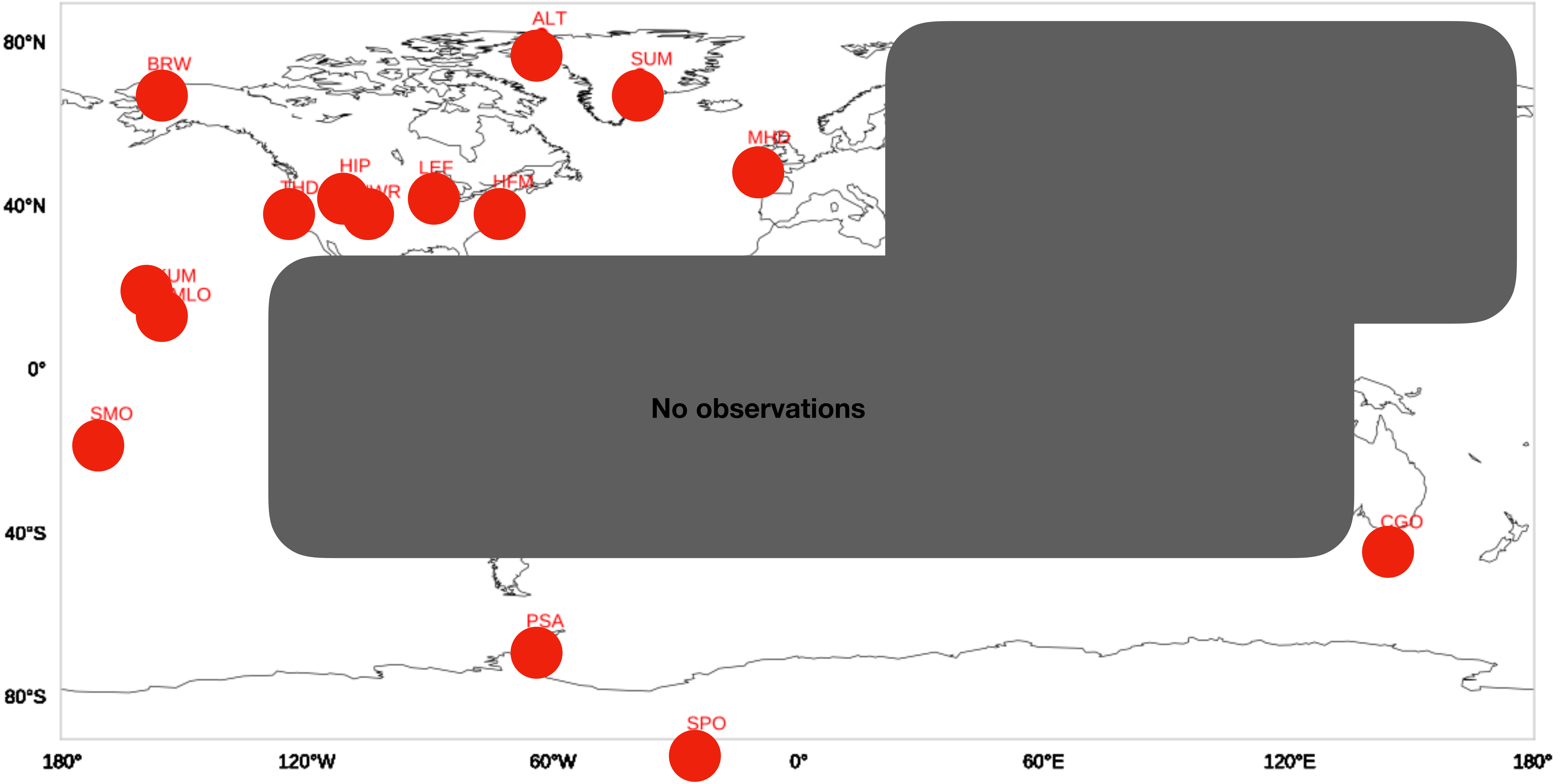
COS-OCS



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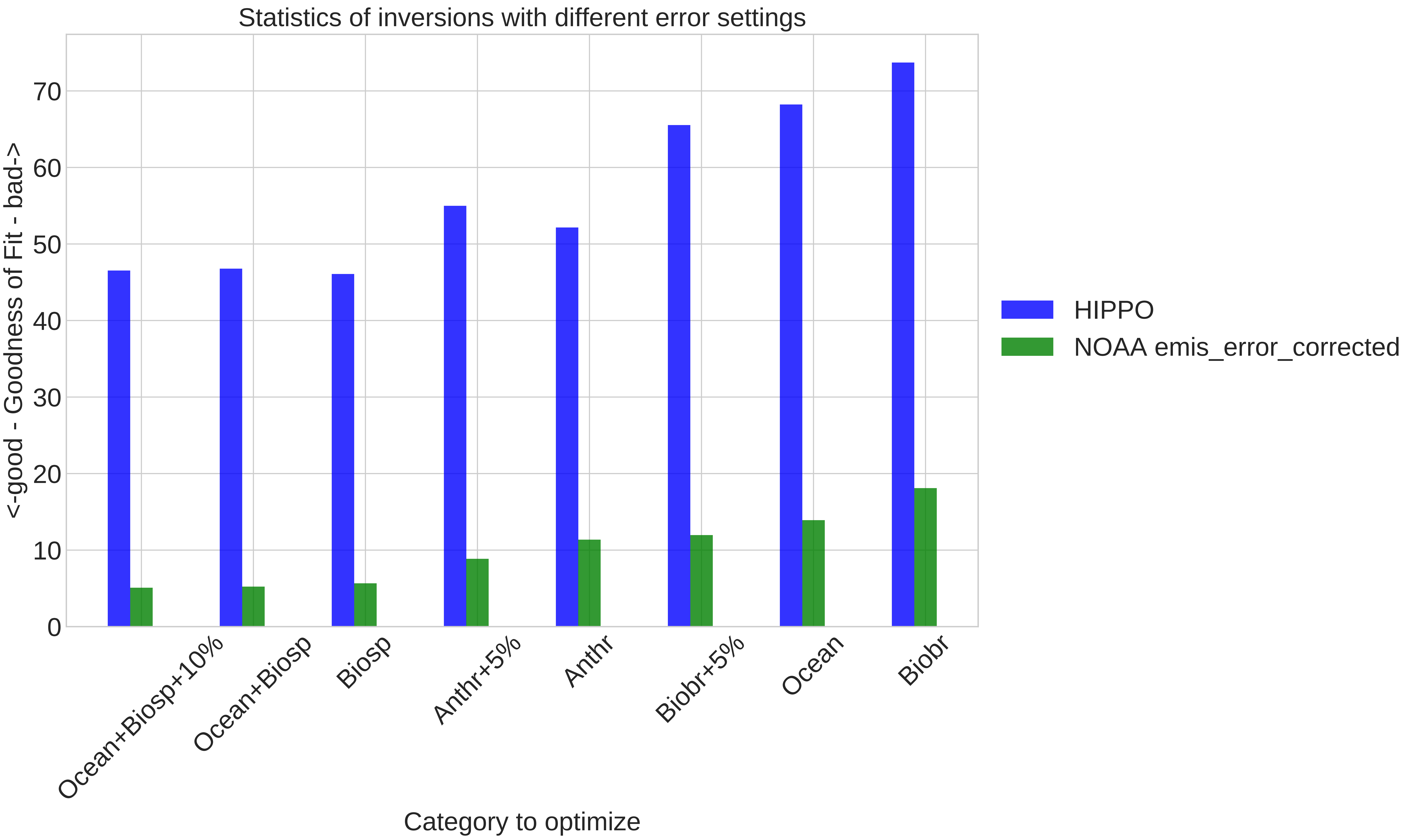

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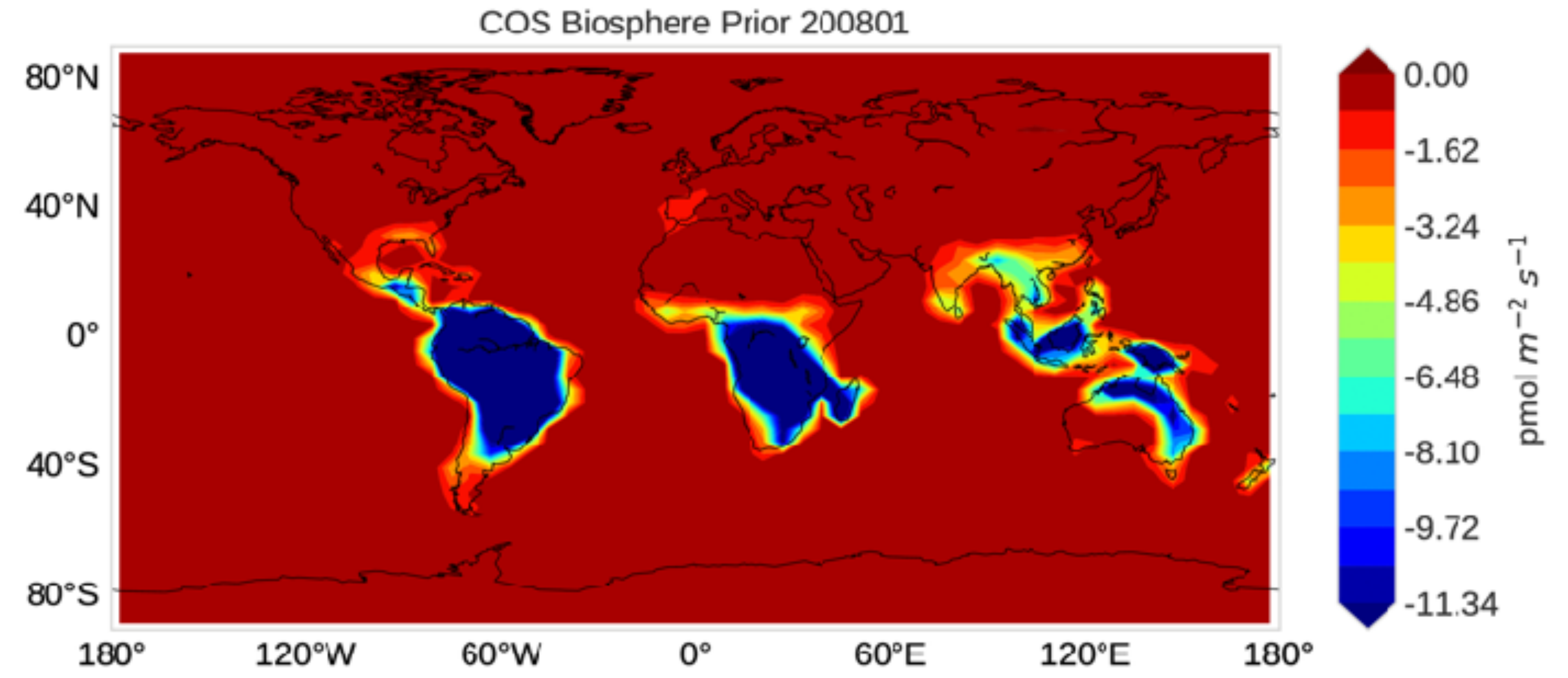
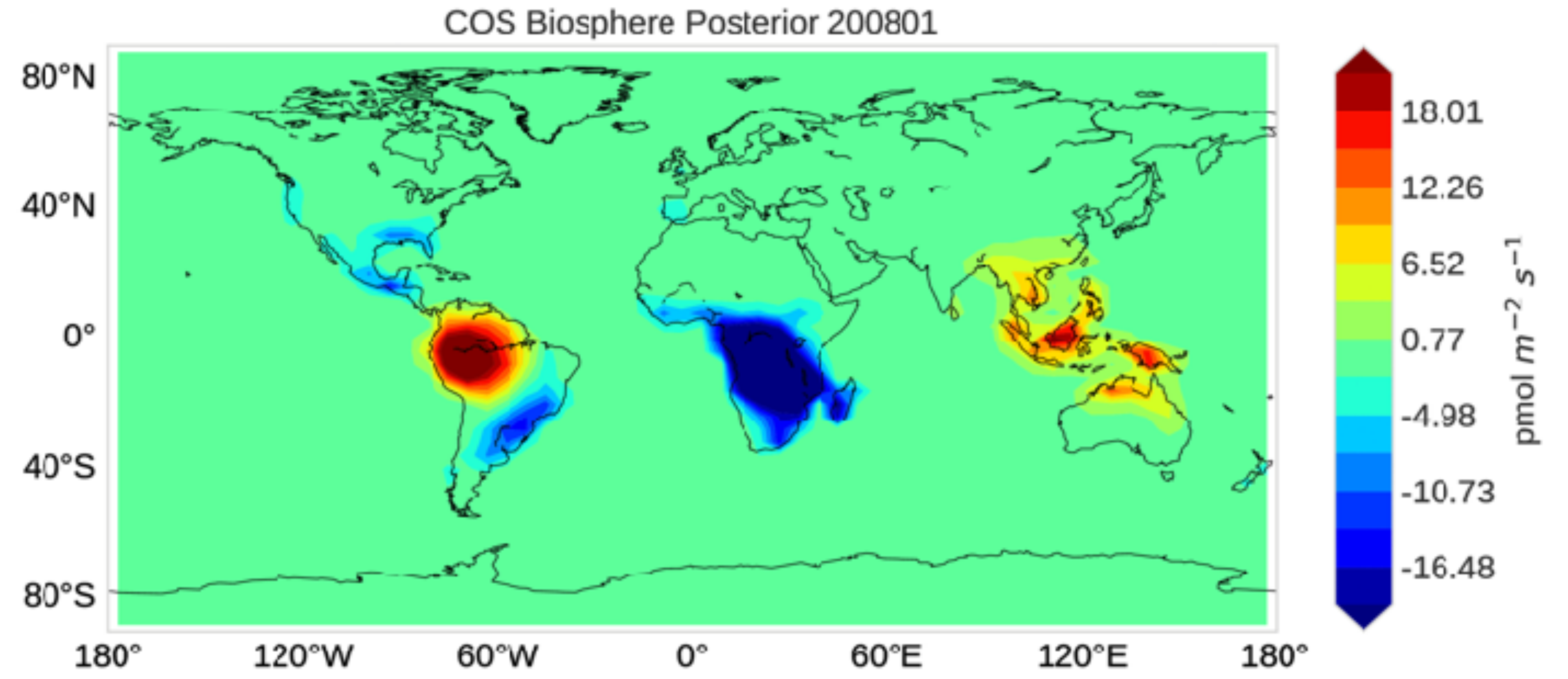
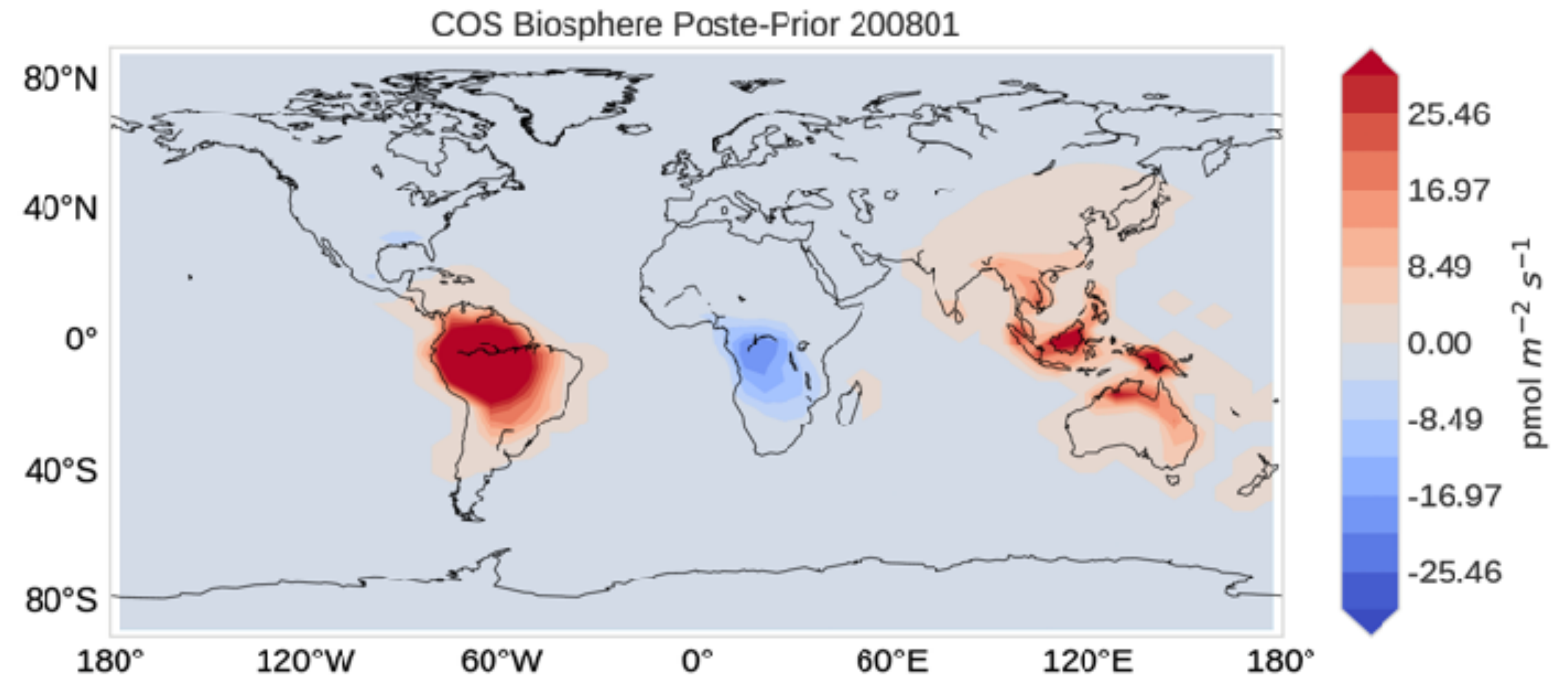


Prior **COS Biosphere:** –898 TgS/yr



Posterior **COS Biosphere:** –499 TgS/yr



Optimised Ocean + Biosphere + other categories

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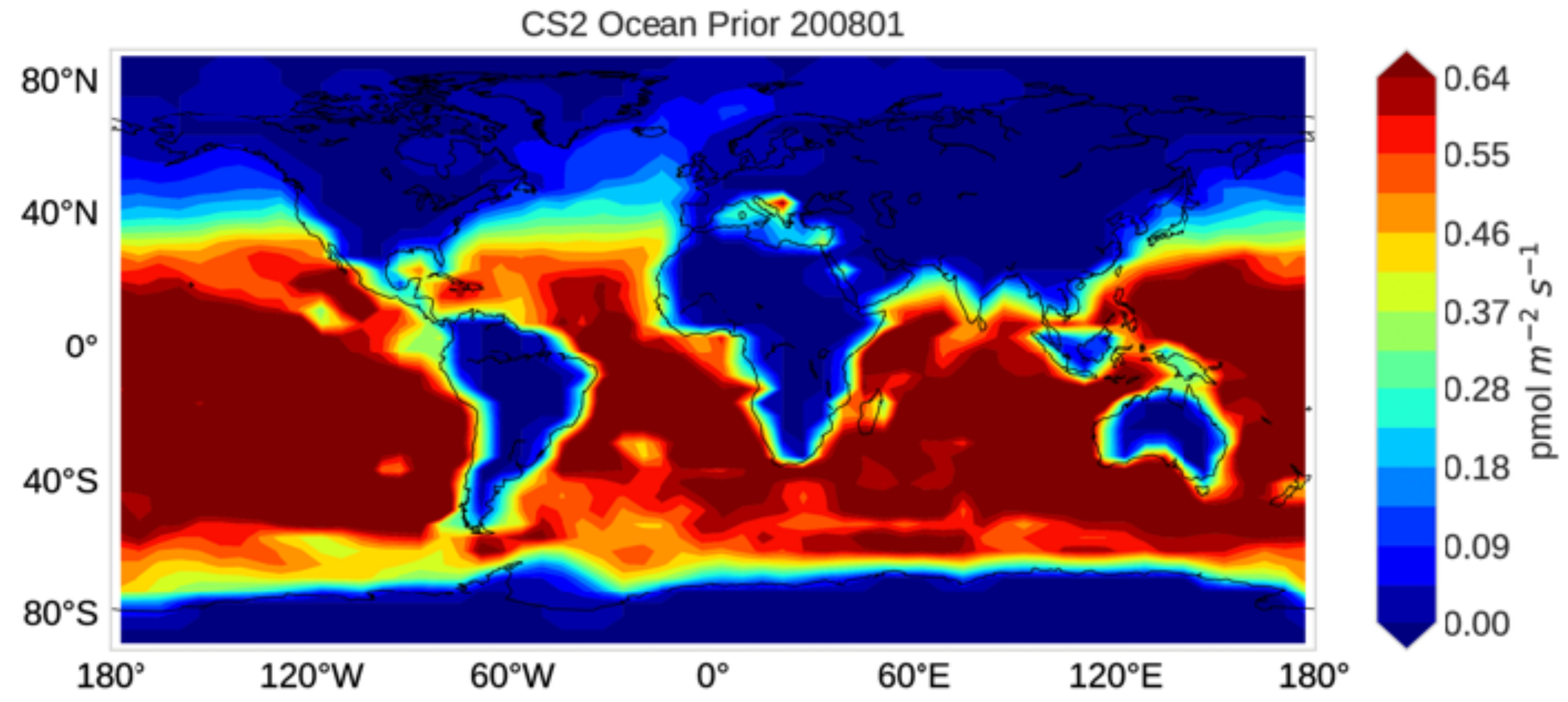
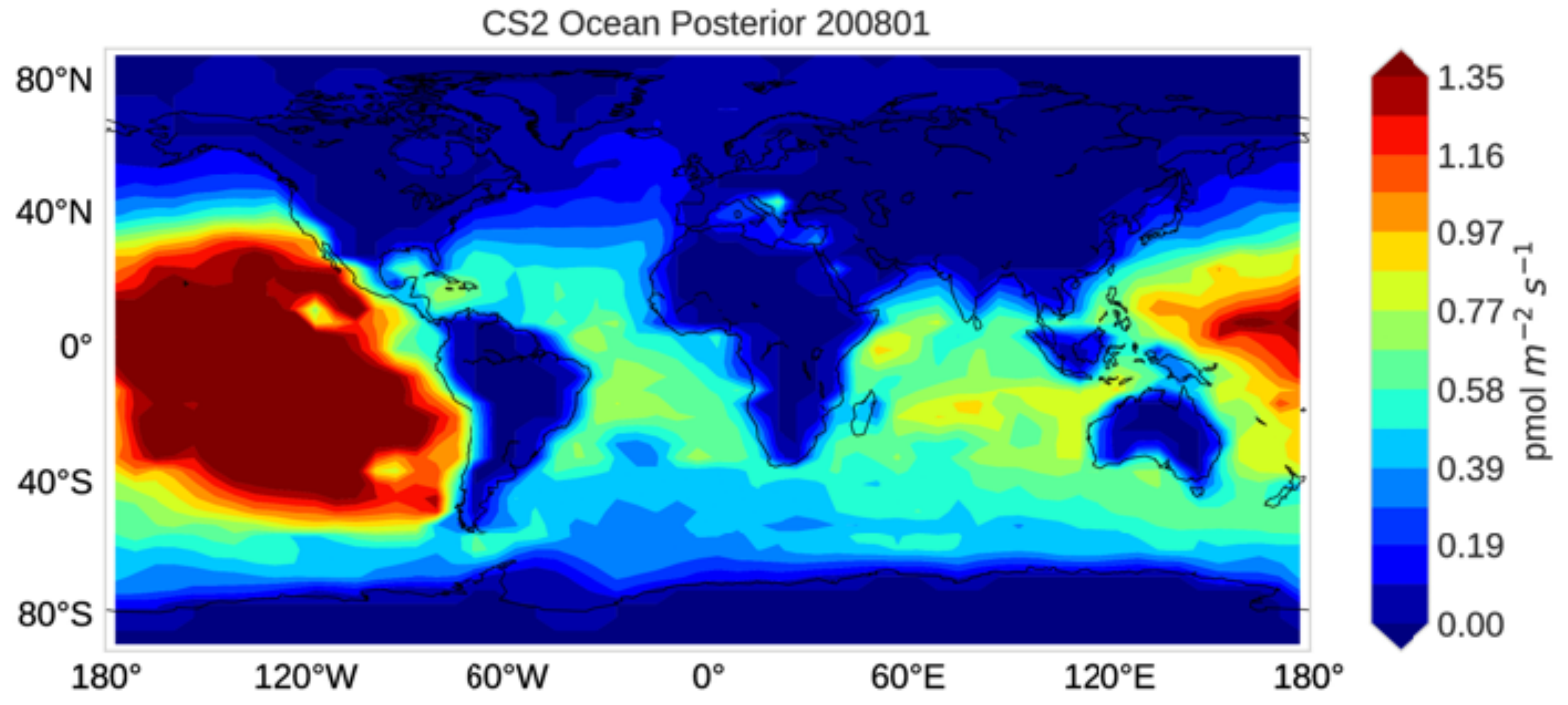
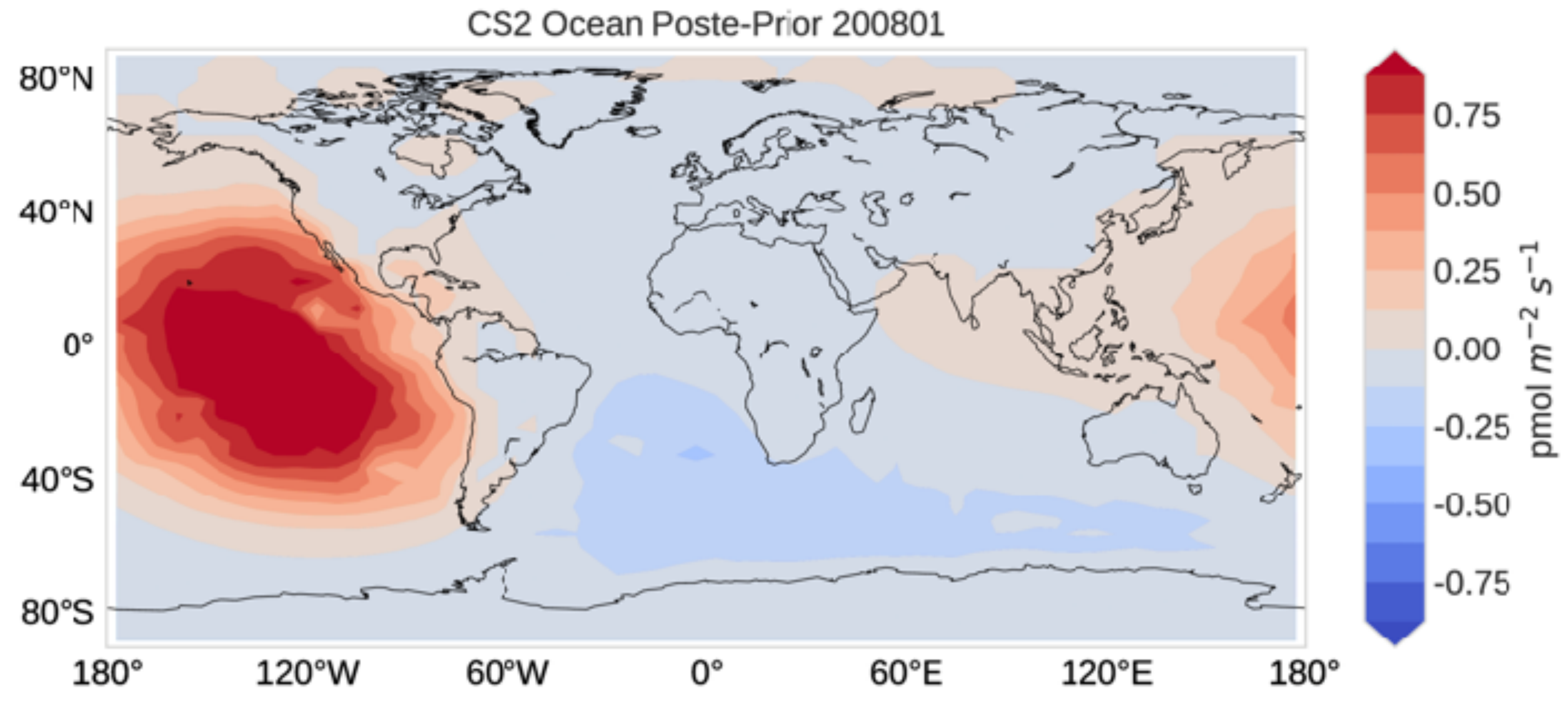
COS-OCS



Prior **CS_2 Ocean:** **81 TgS/yr**



Posterior CS_2 Ocean: **115 TgS/yr**



Optimised Ocean + Biosphere + other categories

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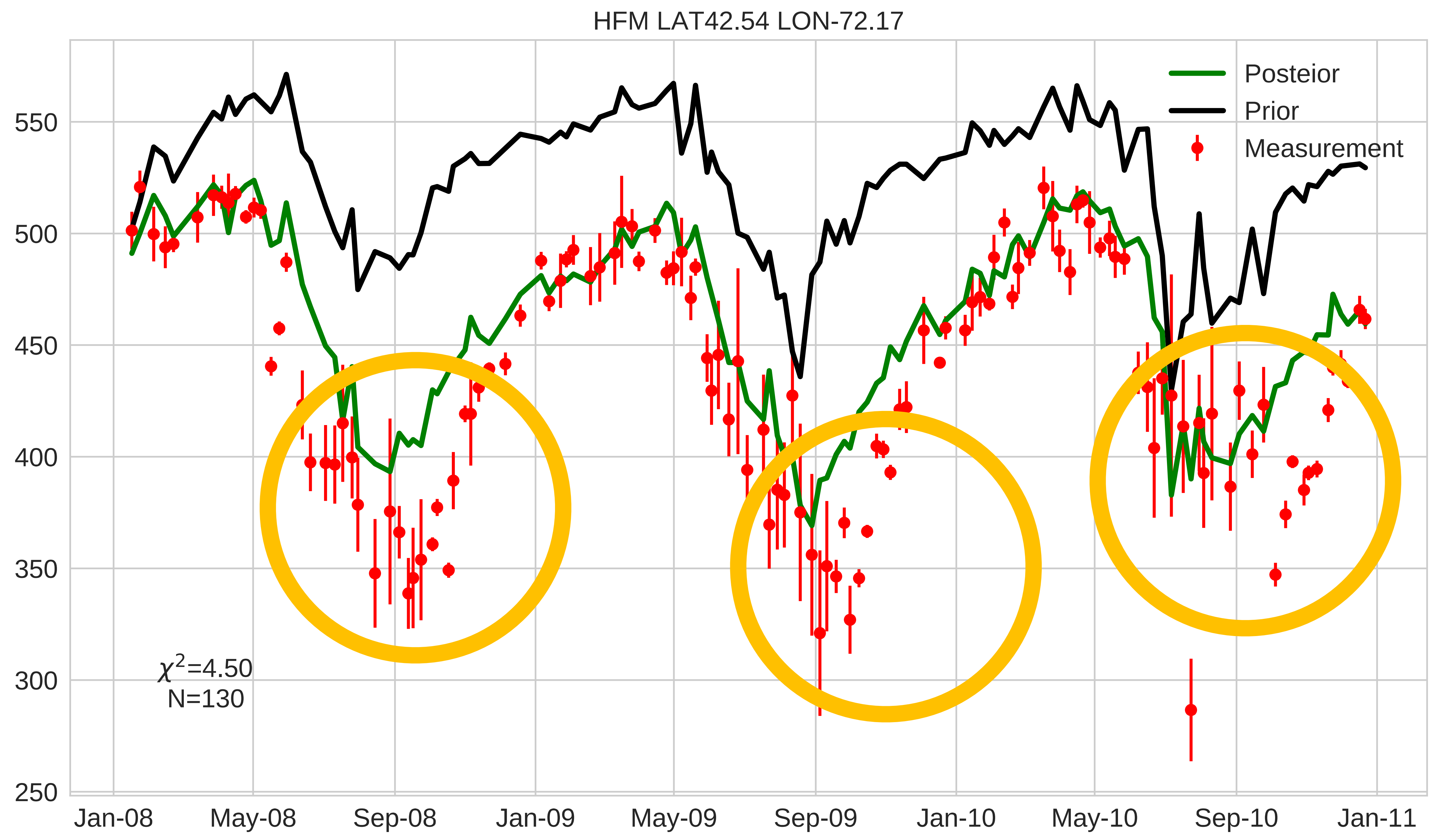
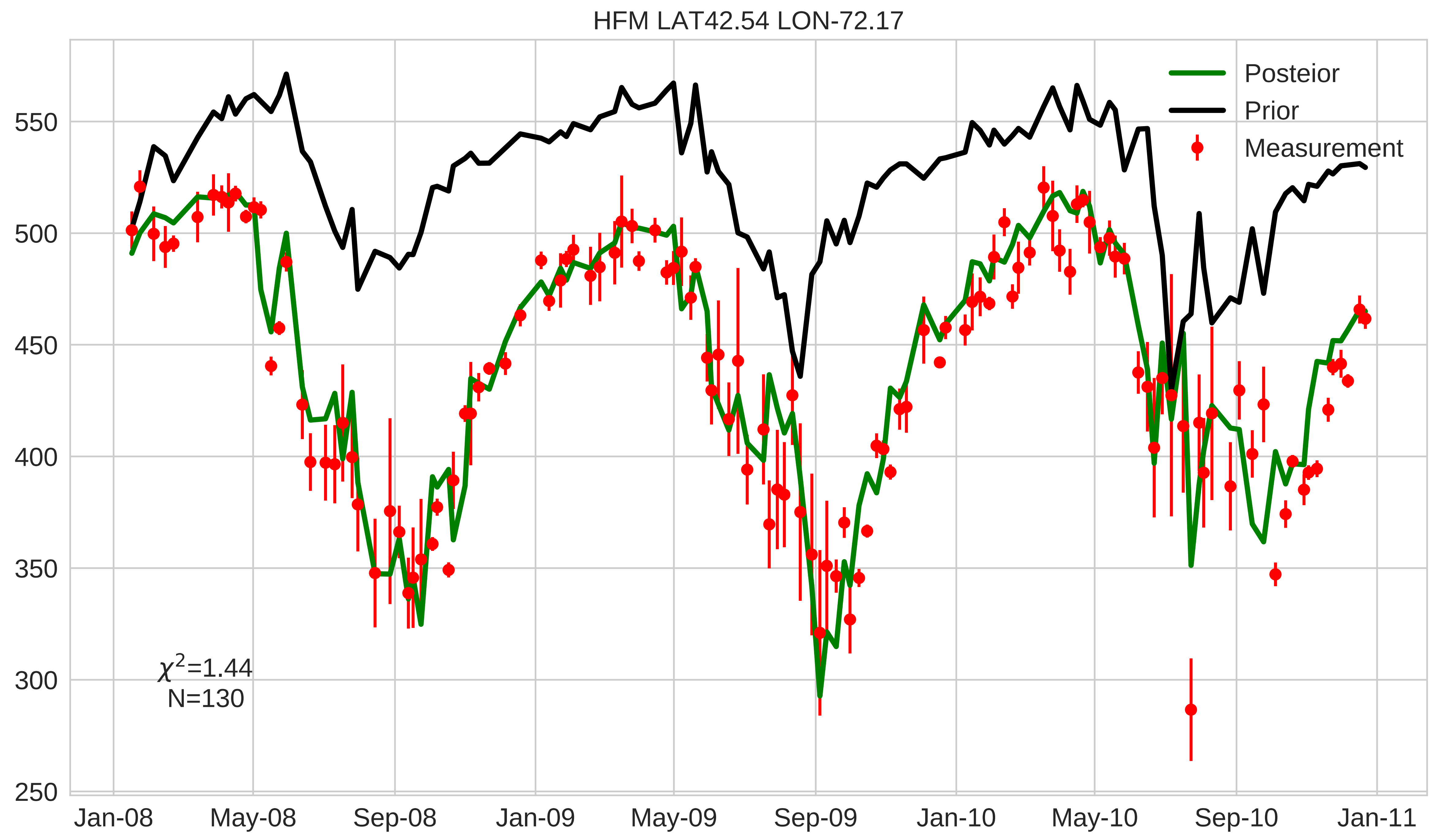
Posterior fit at Harvest Forest:



Optimised only ocean



Optimised only biosphere

COS-OCS

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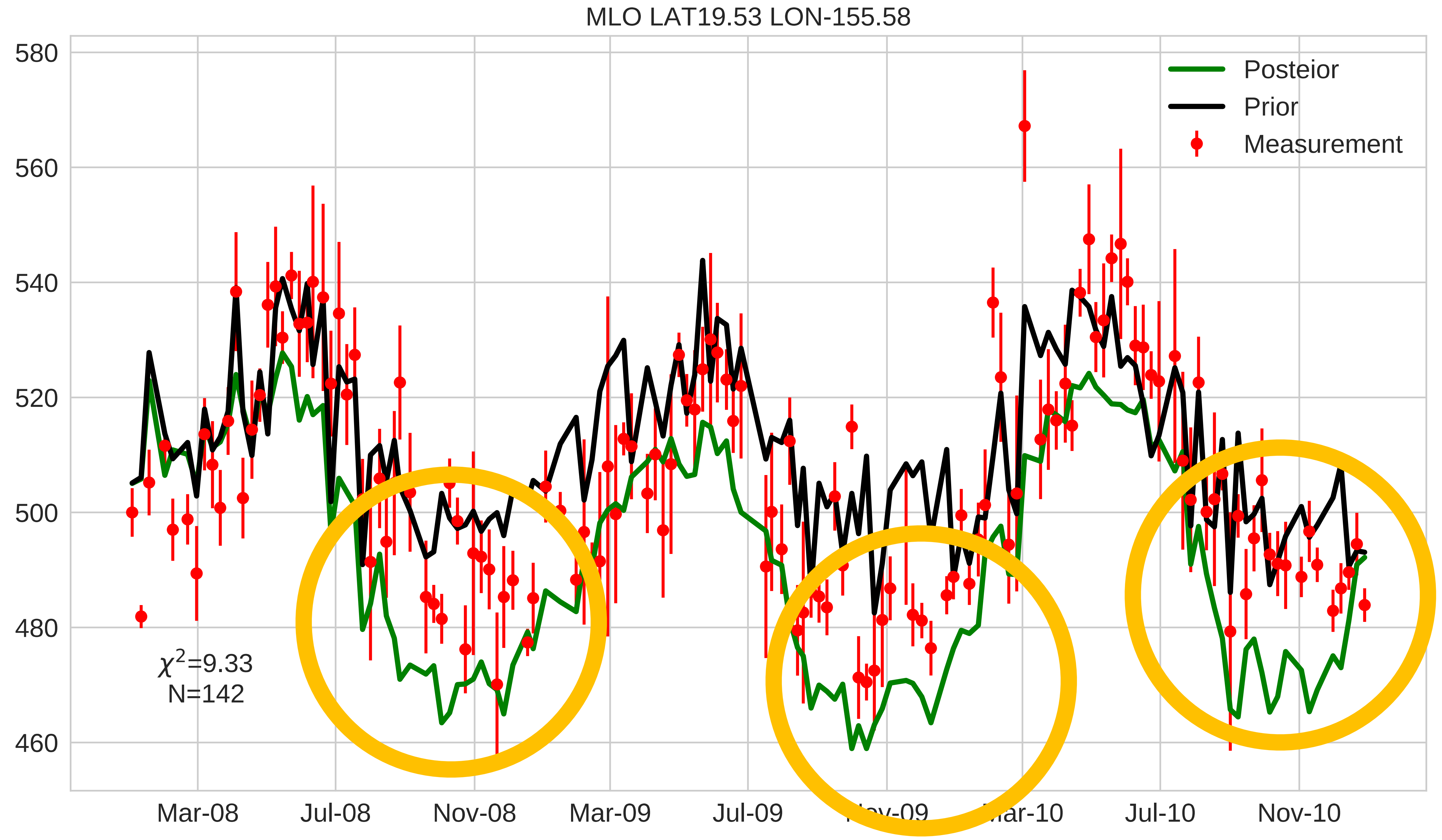
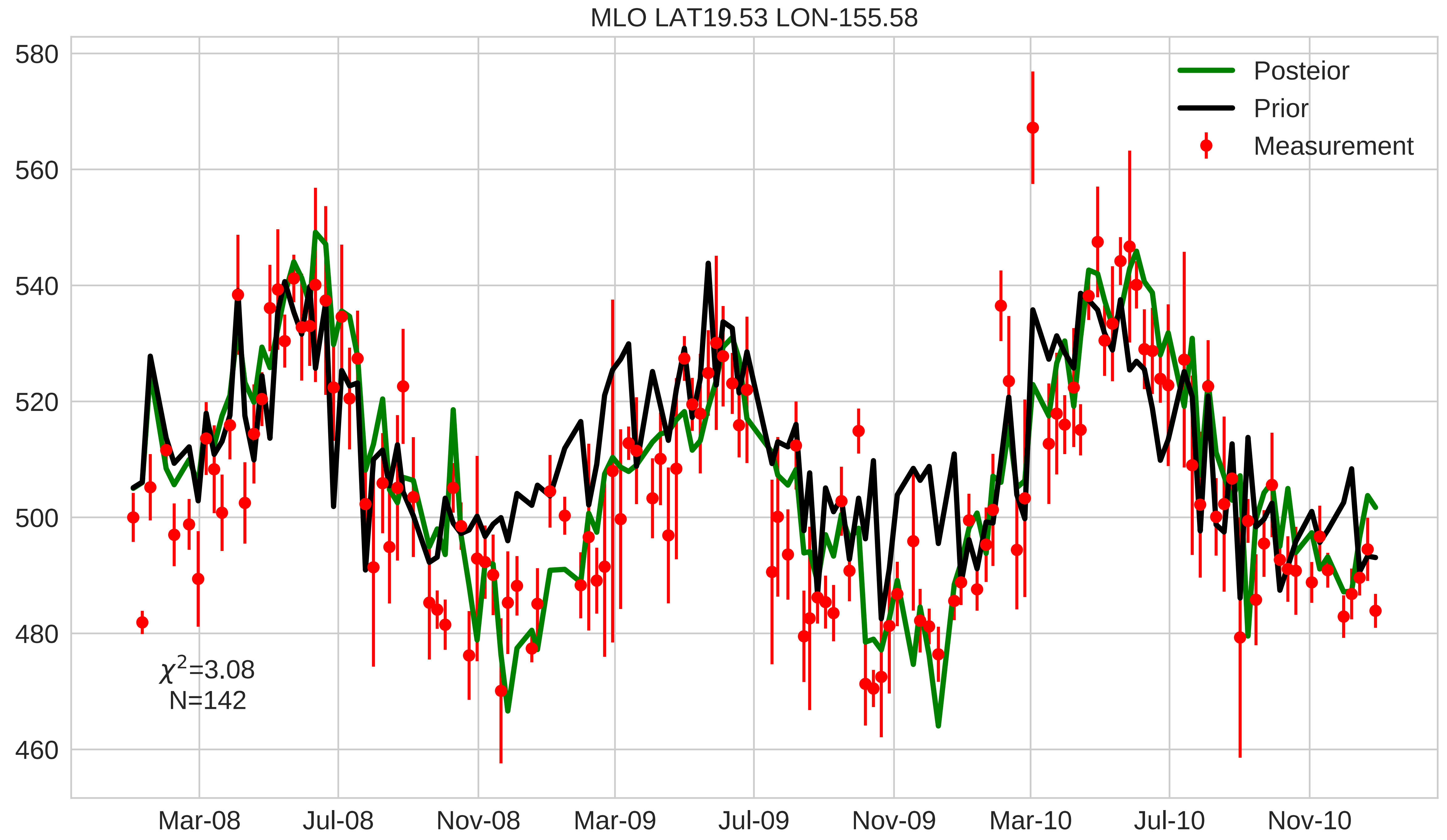
Posterior fit at Mauna Loa:



Optimised only ocean



Optimised only biosphere

COS-OCS

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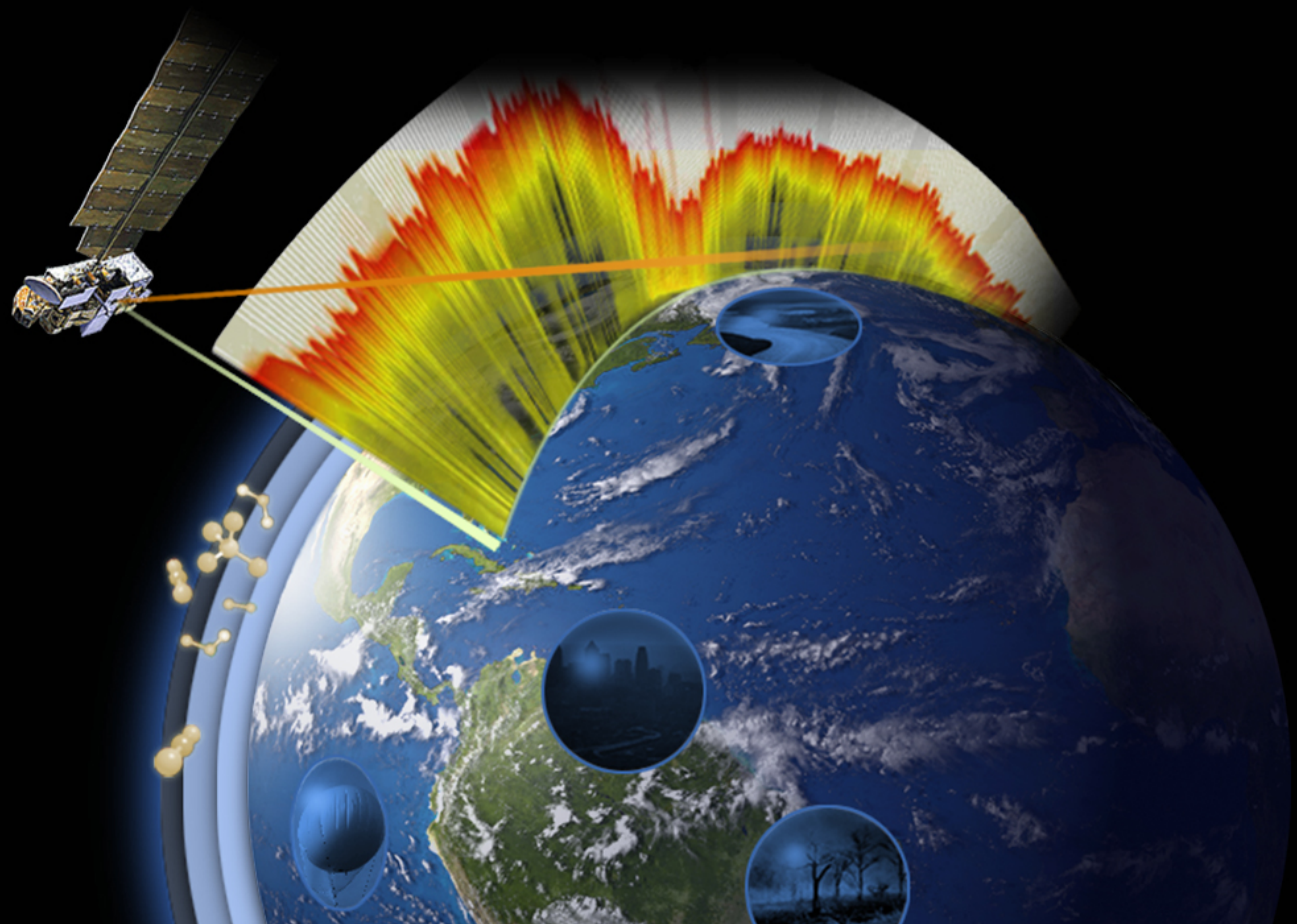
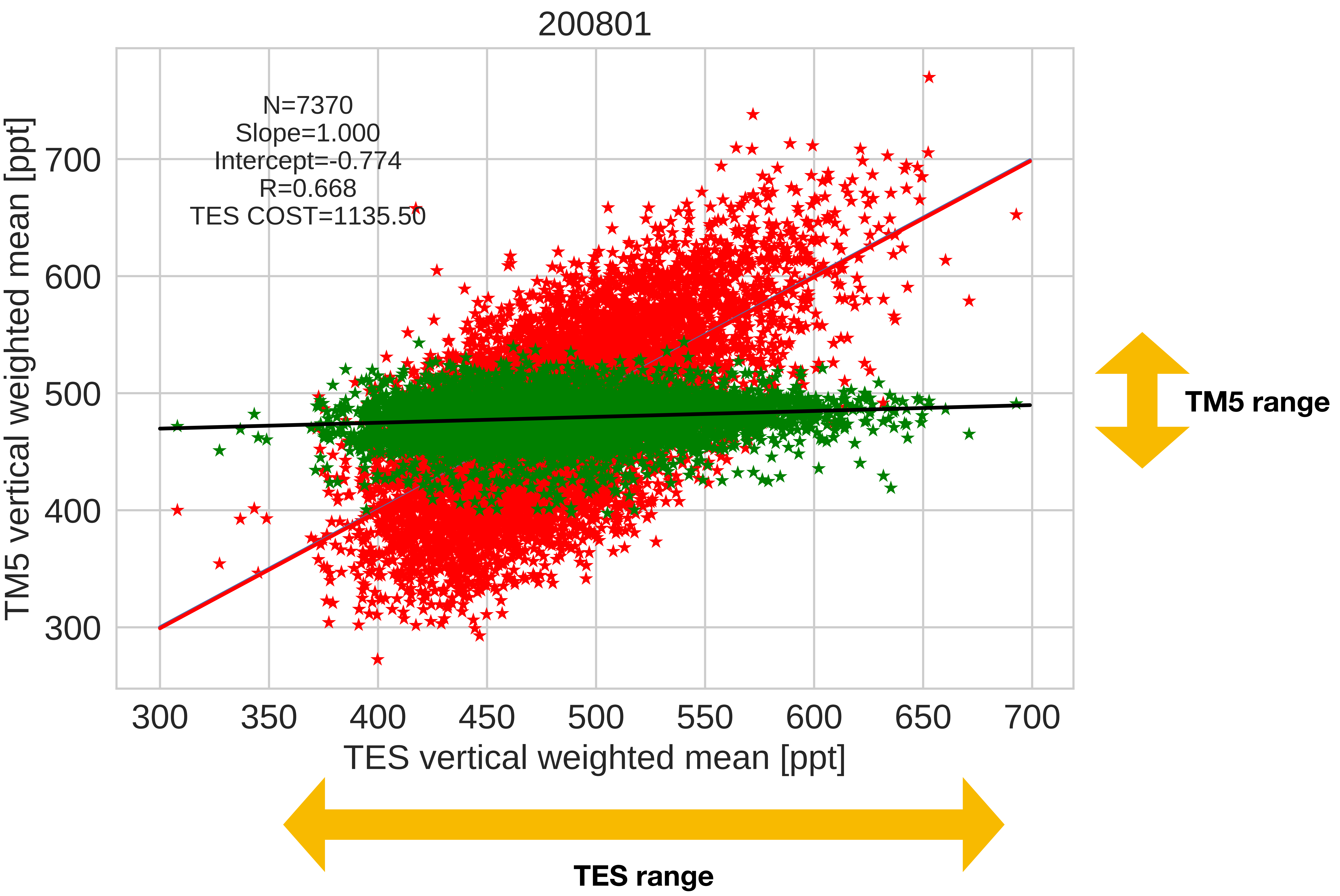
TES





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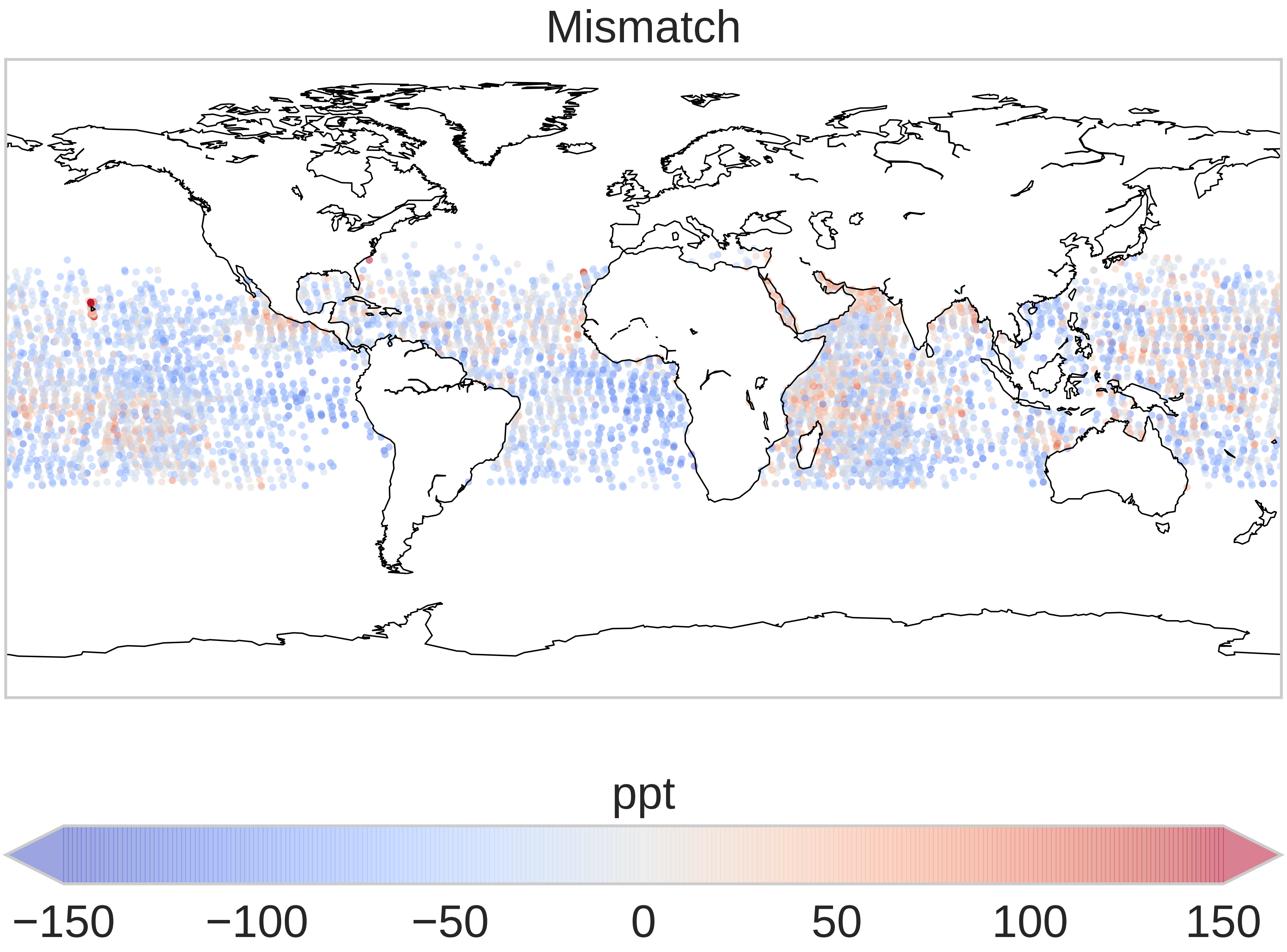
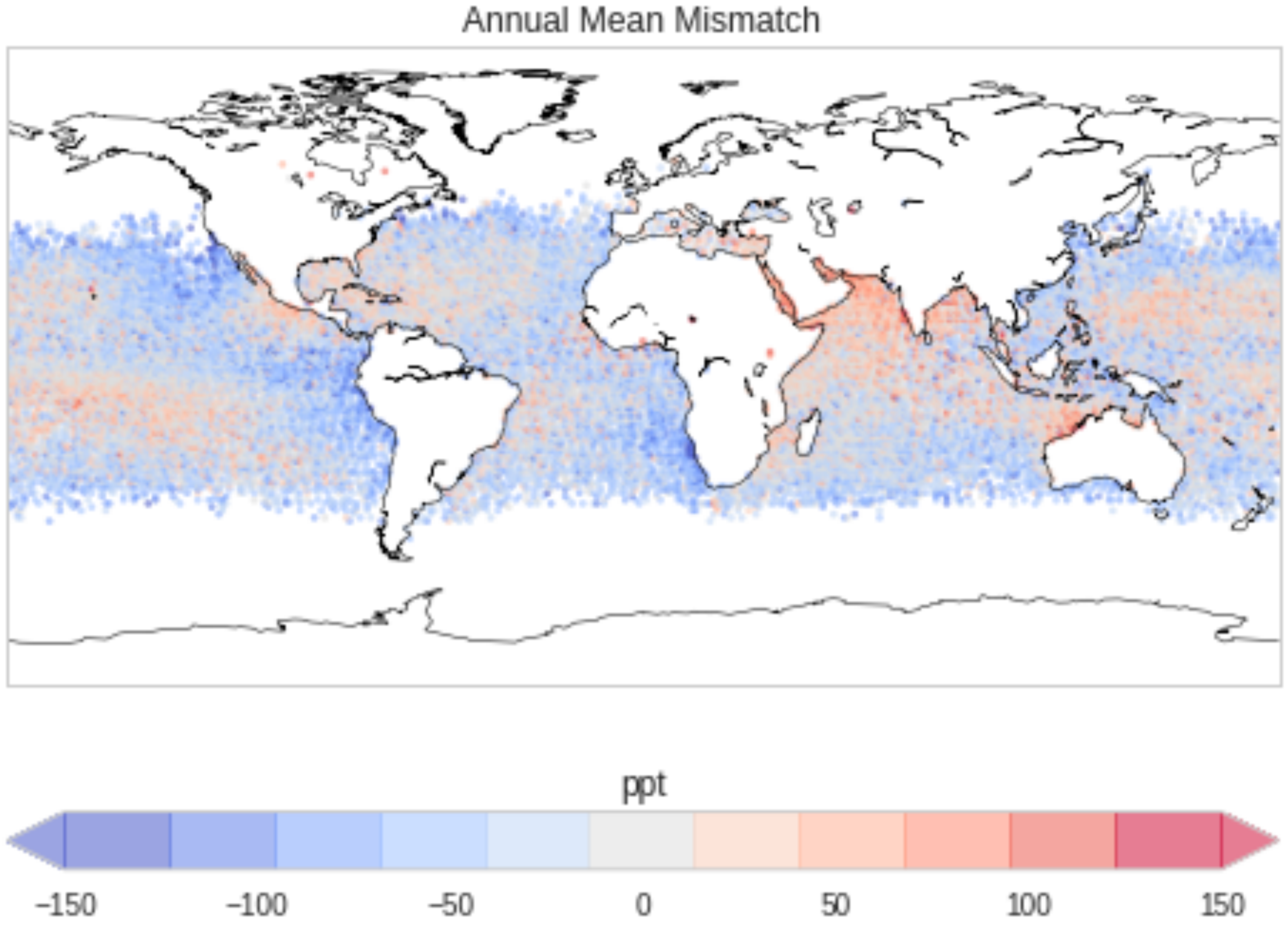
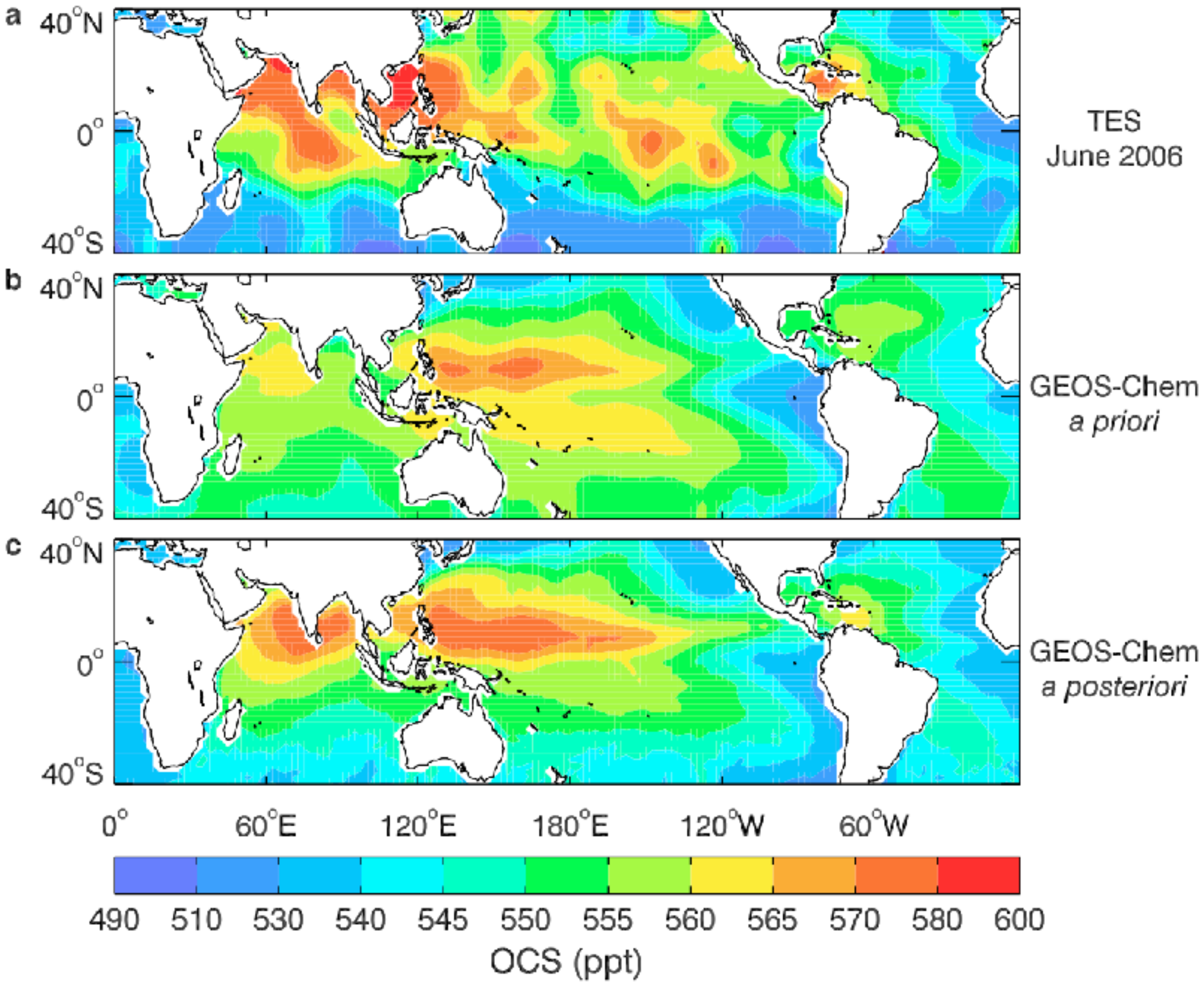
COS-OCS







Kuai et al., JGR, 2015

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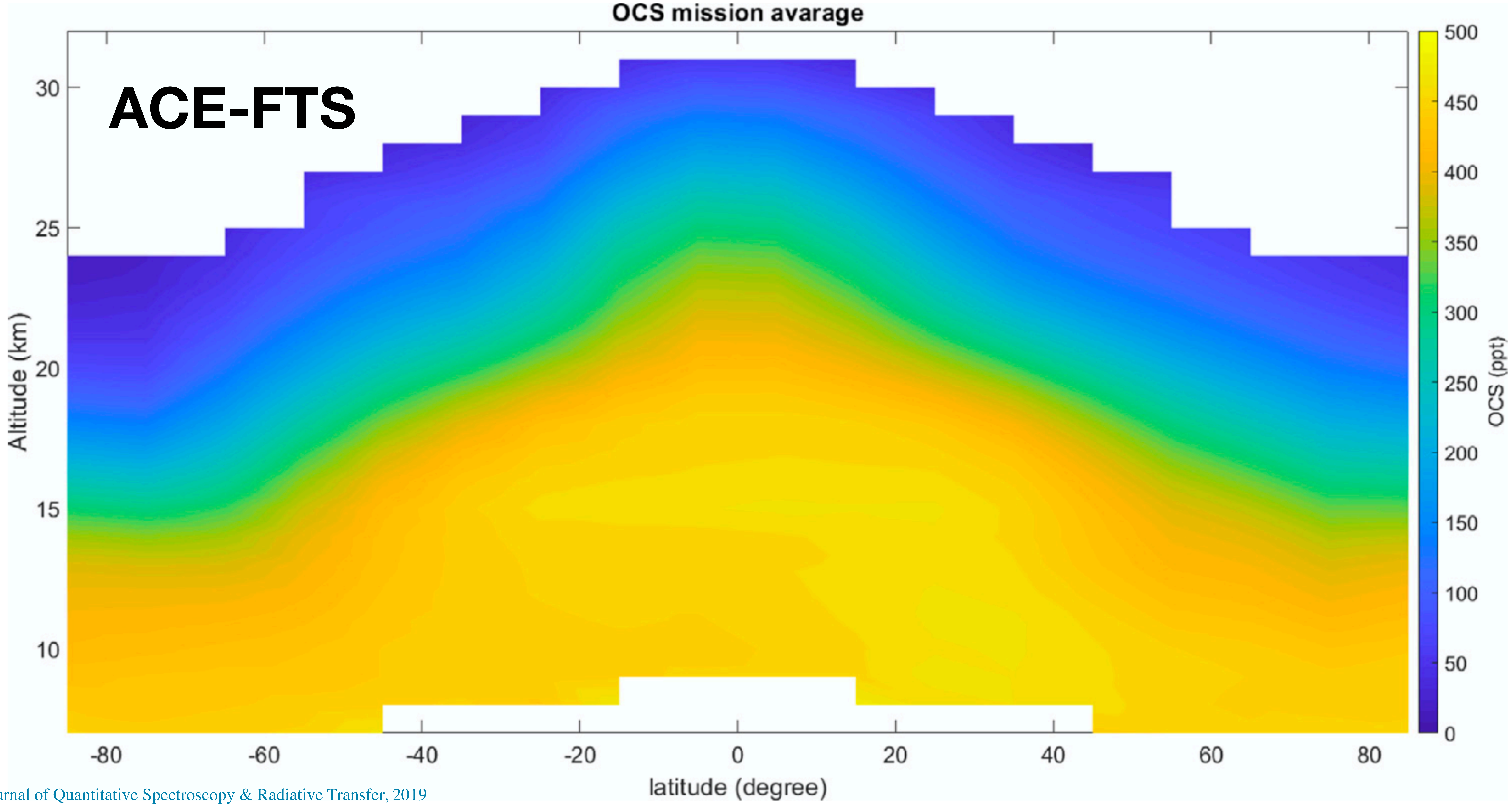
Global measurements of atmospheric carbonyl sulfide (OCS), $OC^{34}S$ and $O^{13}CS$

Mahdi Yousefi[a], Peter F Bernath[a,b,c,*], Chris D Boone[c], Geoffrey C Toon[d]



Journal of Quantitative Spectroscopy & Radiative Transfer, 2019

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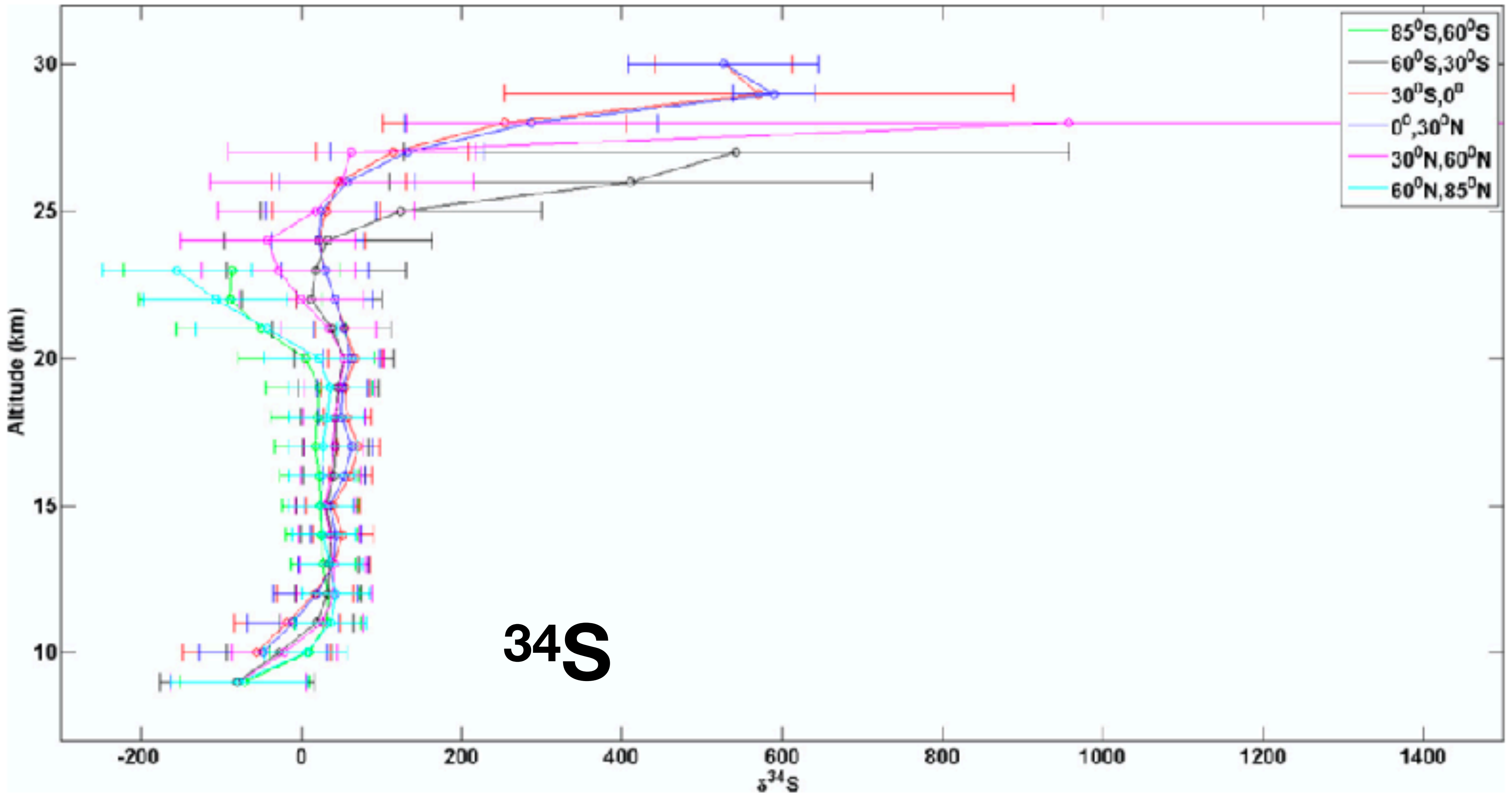
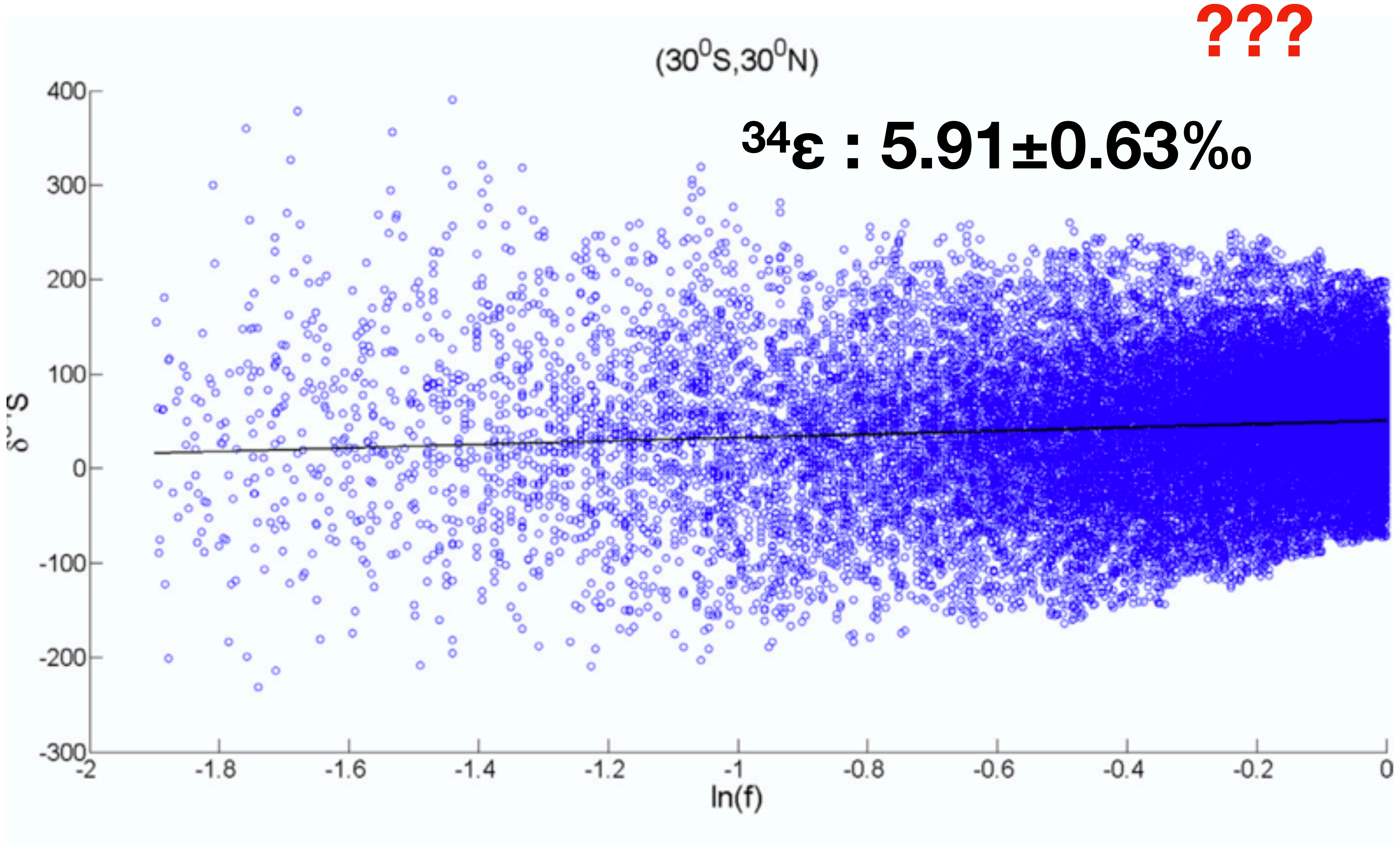
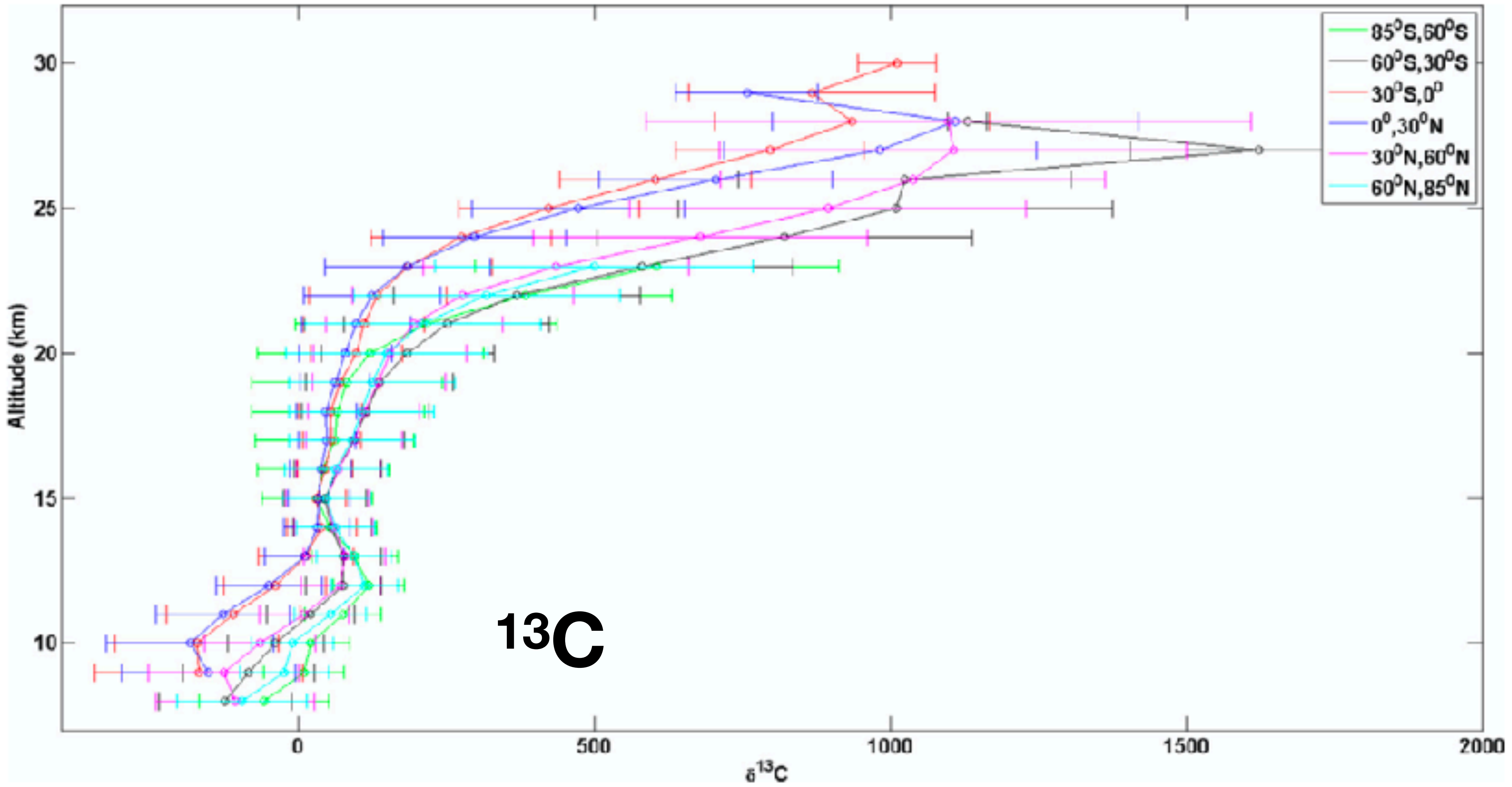
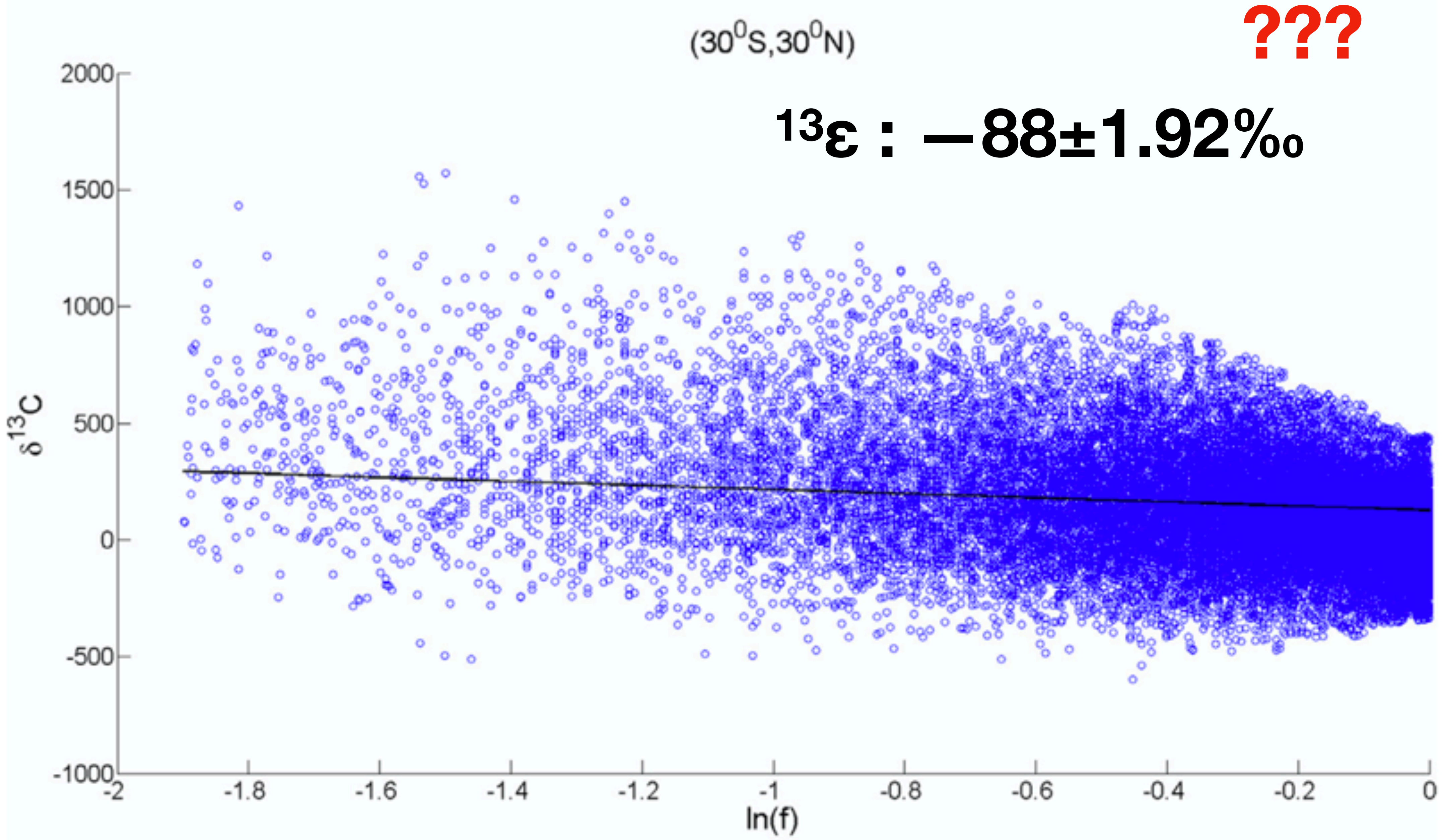
ACE-FTS









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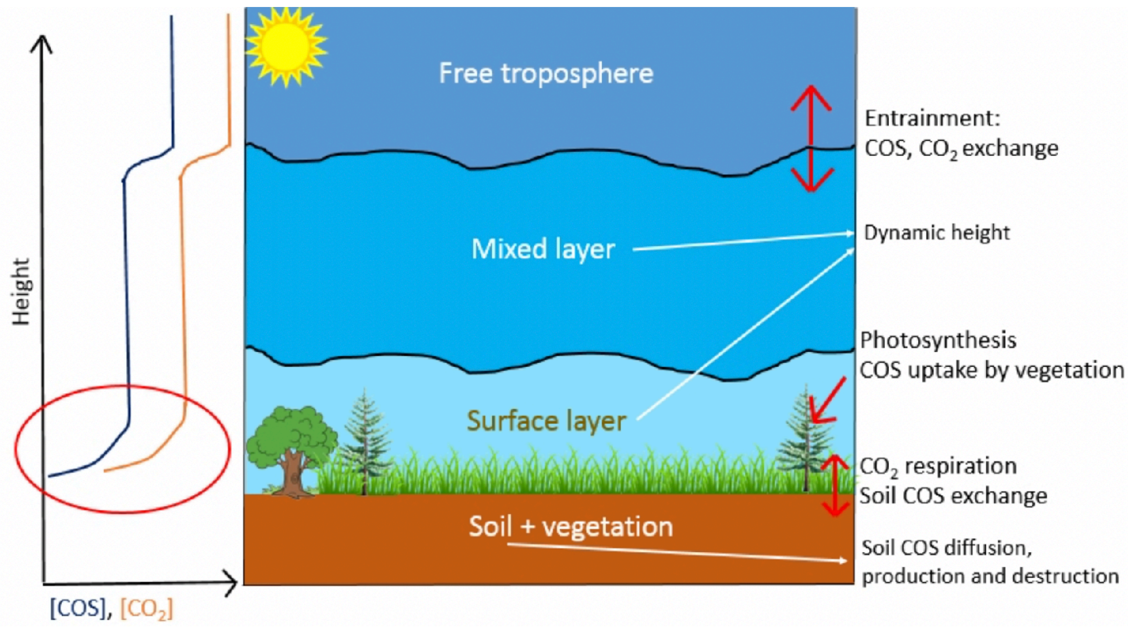
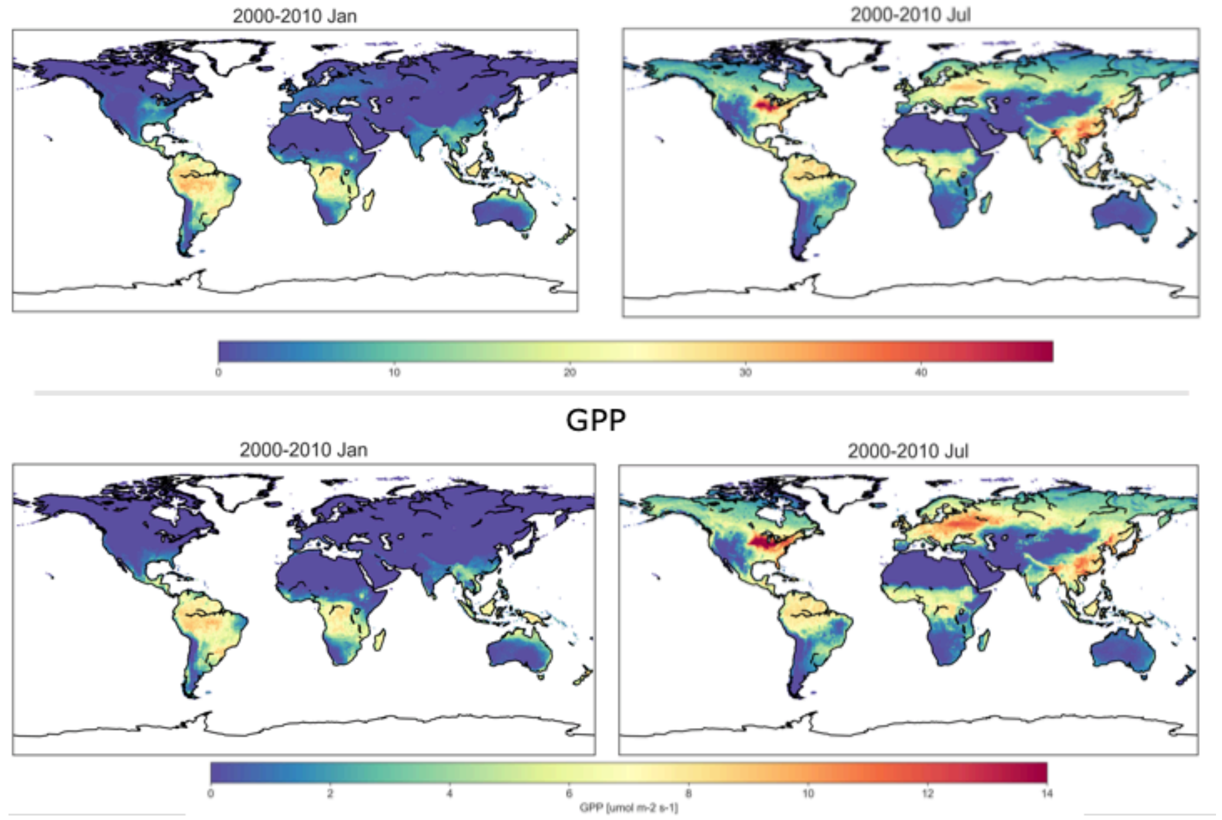
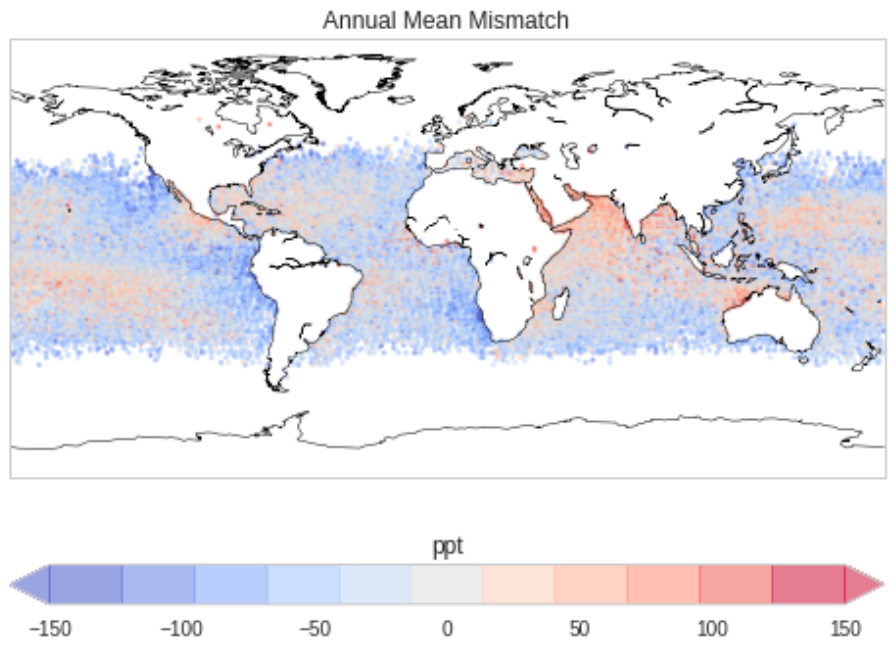
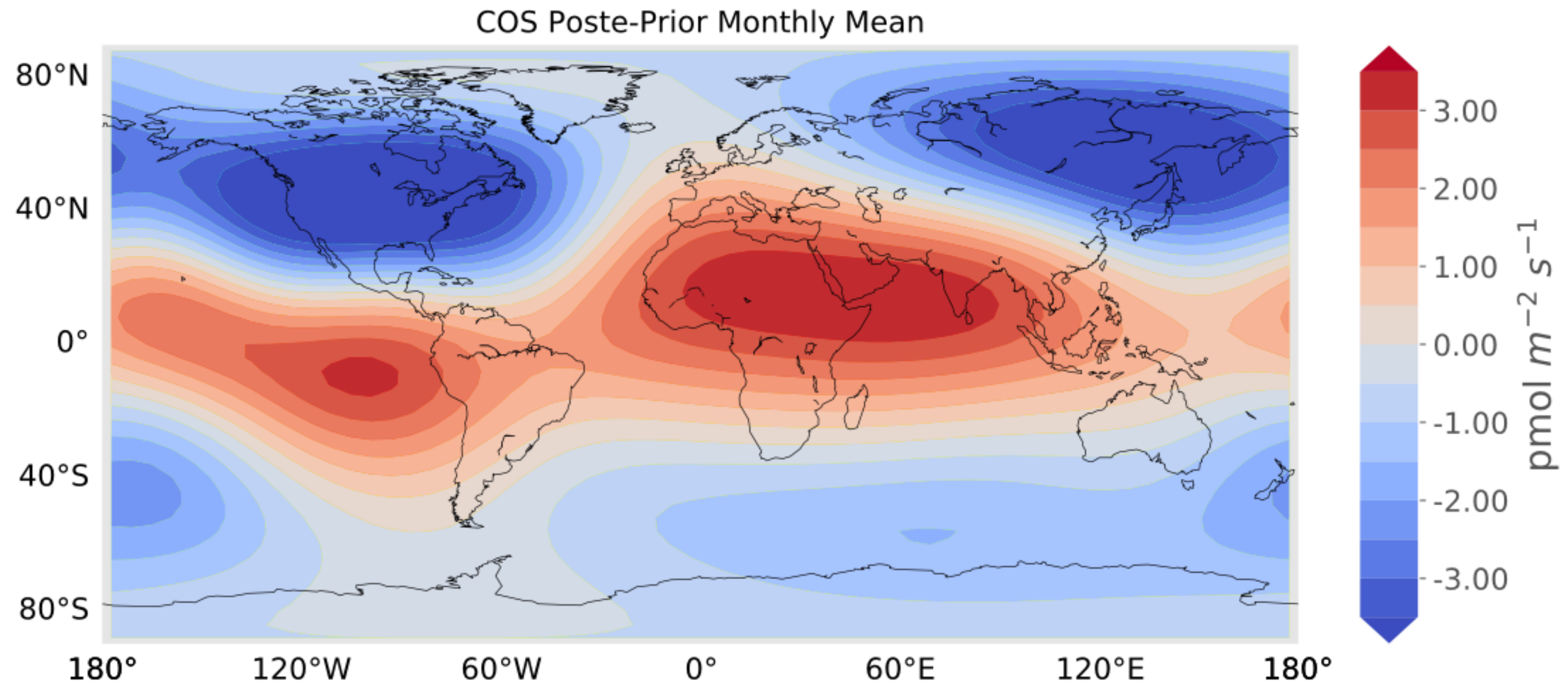
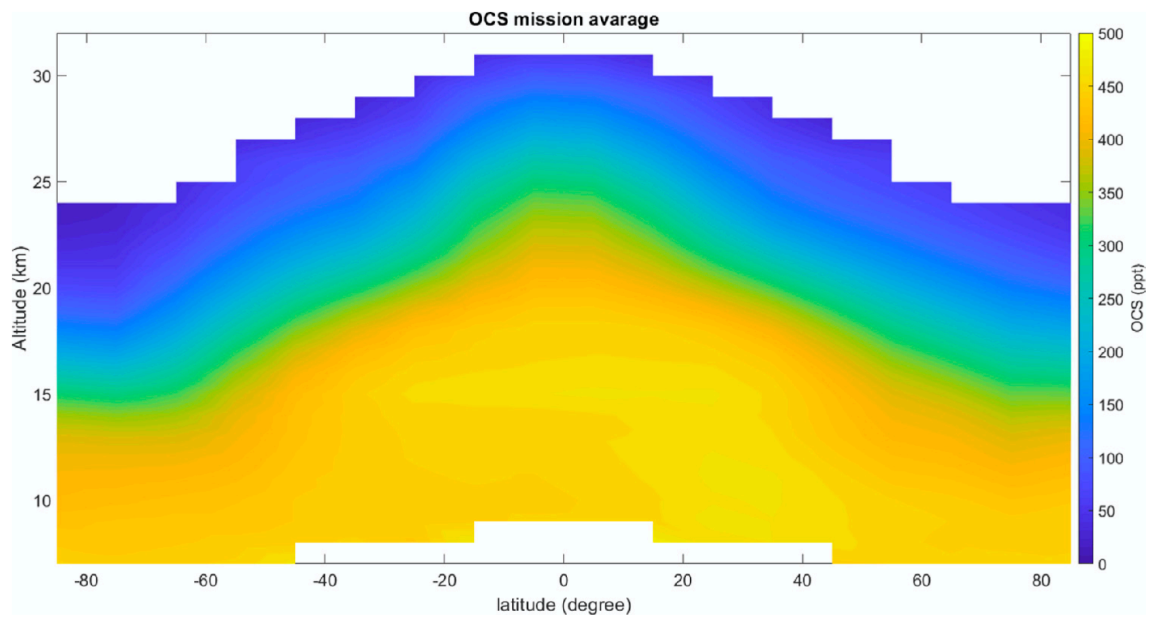
Take Home Messages











COS-OCS

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