Inverse modelling of the global Carbonyl Sulfide (COS) budget

Jin Ma[1], Maarten Krol[1,2], Linda Kooijmans[2]

1. Institute for Marine and Atmospheric Research, Utrecht University

2. Meteorology and Air Quality, Wageningen University & Research

COS modelling meeting, Rotterdam, Jan 2020

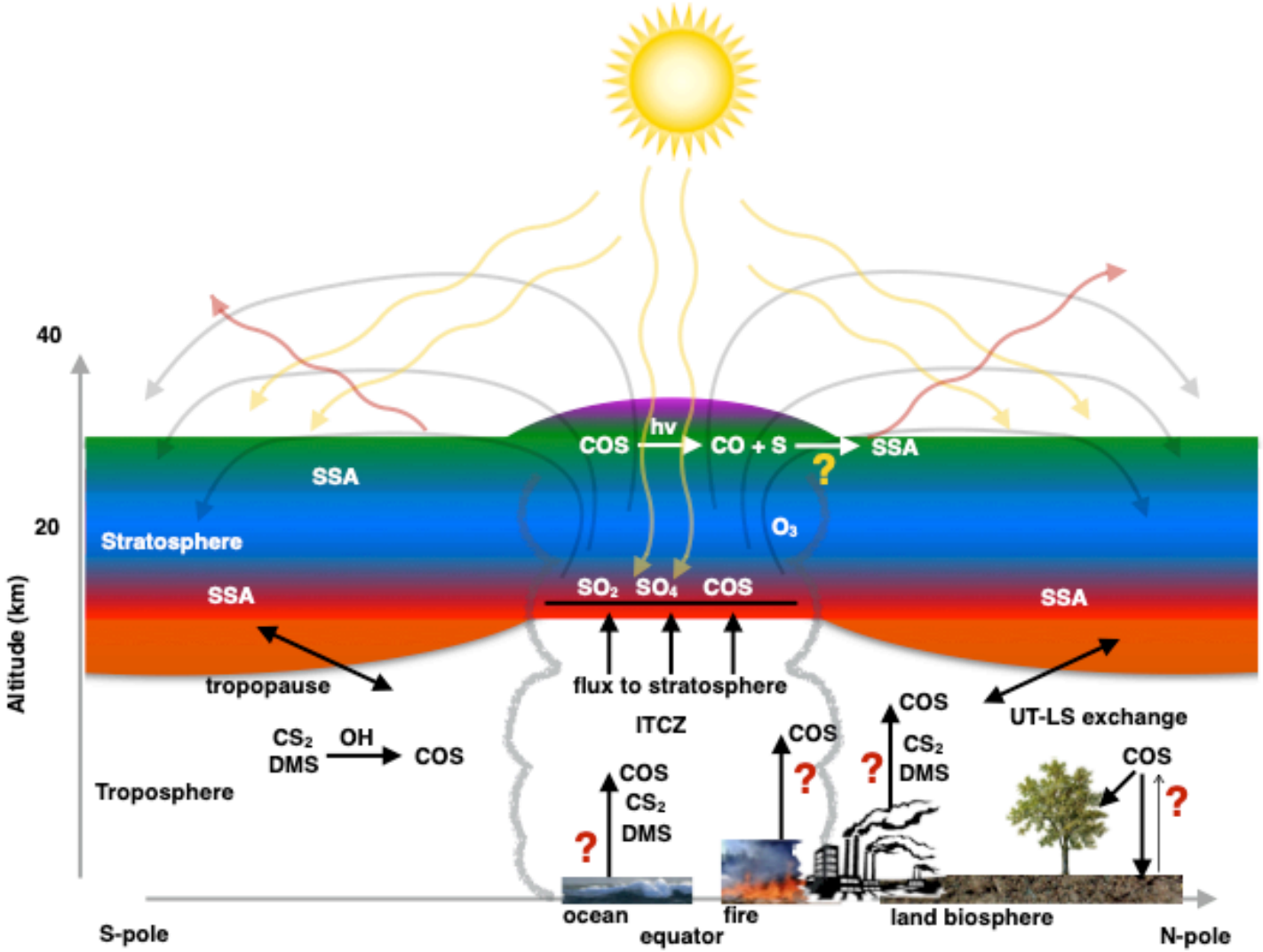
Acknowledgements to: NOAA flask and aircraft network, HIPPO aircraft data, TES Aura, MIPAS and ACE-FTS satellite product

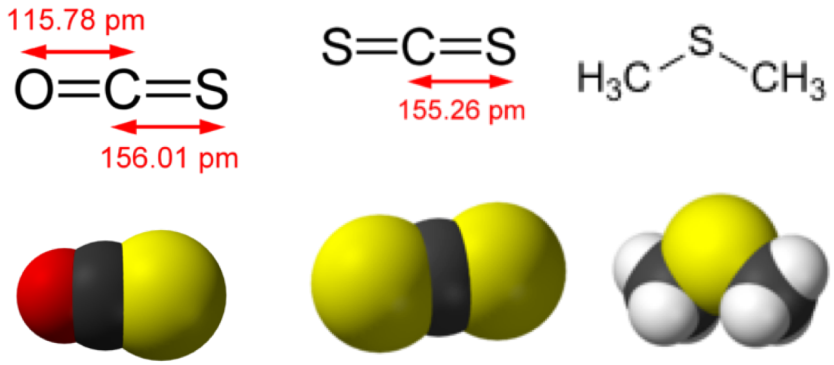
This project has received funding from the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation program under grant agreement No 742798

Objectives

- Implementation of inversion system of COS-CS_2-DMS
- Missing emissions in COS budgets: inversion based on unknown category
- Model validation with HIPPO and NOAA airborne profiles

COS, CS_2 and DMS in the atmosphere





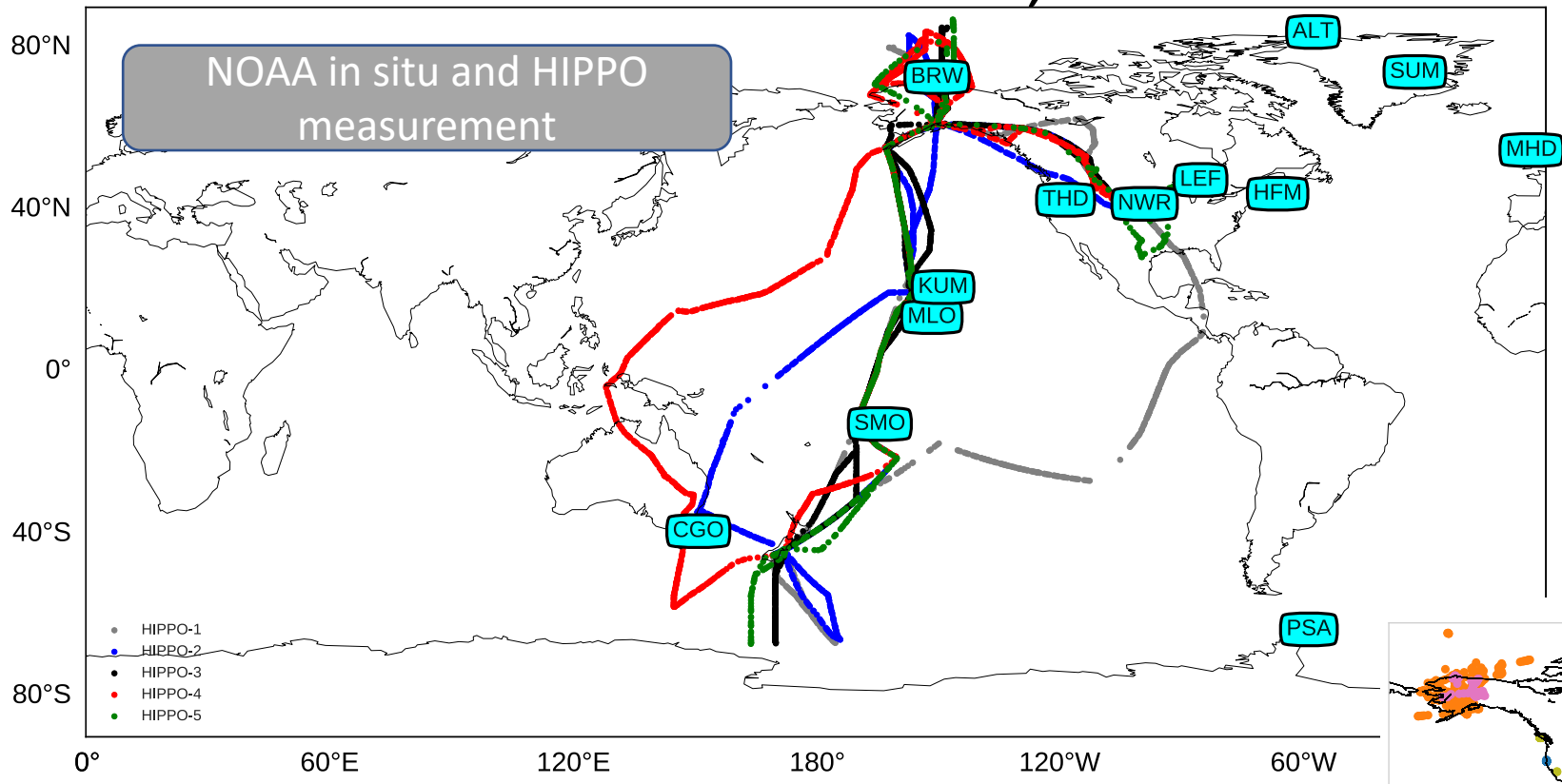
COS lifetime : ~2 years
CS_2 lifetime : ~15 days
DMS lifetime : ~1.2 days

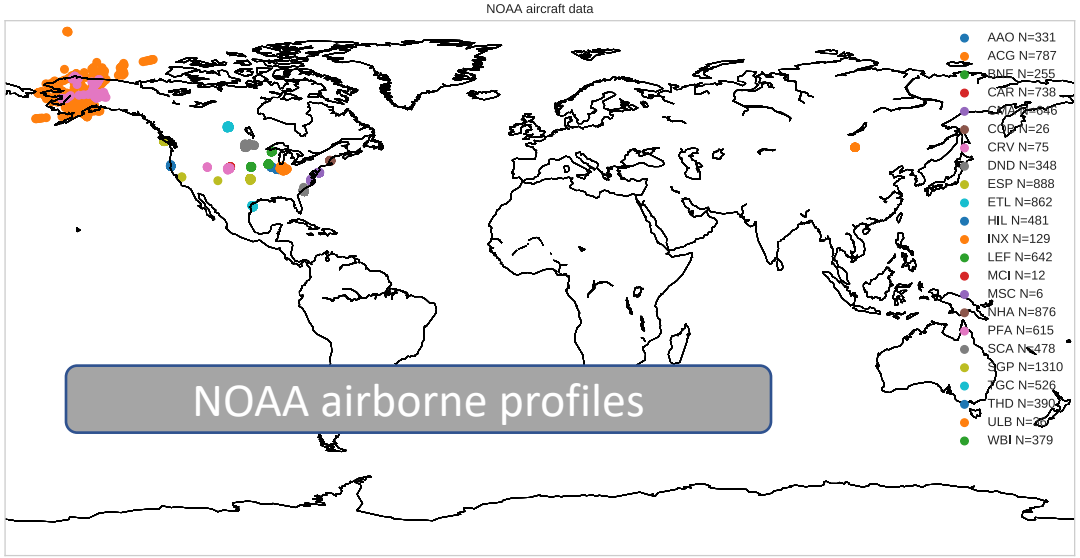
CS_2 and DMS can be oxidized quickly to contribute to COS formation in the atmosphere.

Model implementation within TM5-4DVAR

- 1. Emission implementation
- 2. Chemistry: troposphere OH and stratosphere removal
- 3. Data use: NOAA, HIPPO and satellite data
- 4. Inversion setup: correlation settings, and model validation

Observations: NOAA, HIPPO and airborne data





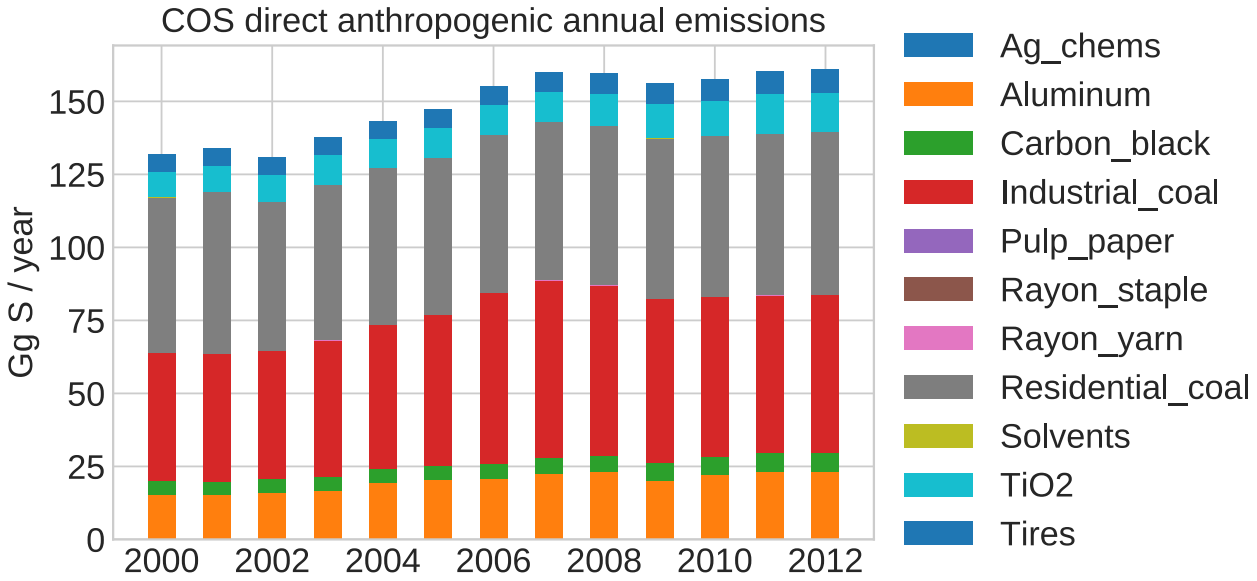
Global COS budgets: Berry2013

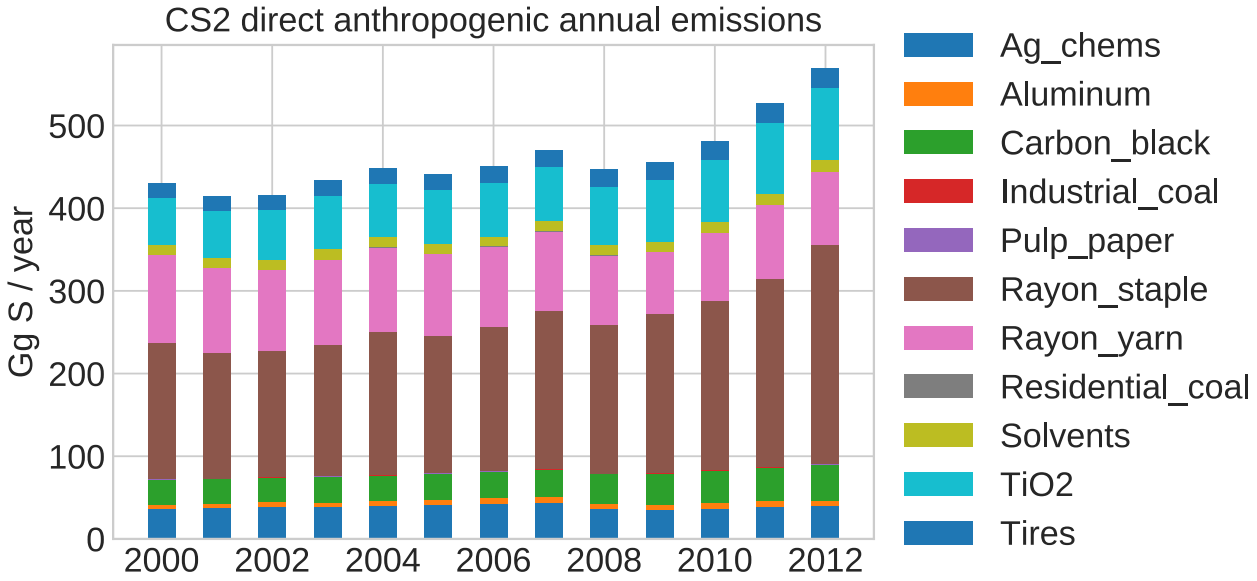
COS Global Budget (GgS/yr)	Berry2013	Prior of this study
Direct COS flux from oceans	39	40
Indirect COS flux as CS2 from oceans	81	81
Indirect COS flux as DMS from oceans	156	156
Direct anthropogenic flux	64	155
Indirect anthropogenic flux from CS2	116	188
Indirect anthropogenic flux from DMS	1	6
Biomass burning	136	136
Additional ocean flux	600	-
Anoxic soils and wetlands	-	-
Sources	1193	762
Destruction by OH	-101	-101
Destruction by O	-	-
Destruction by photolysis	-	-40
Uptake by plants	-738	-1053
Uptake by soil	-355	
Sinks	-1194	-1194
Net total	-2	-432

- Major updates on anthropogenic, SiB4 biosphere, and biomass emissions, and COS photolysis
- Net total prior is –432 GgS/yr, so add 432 GgS/yr uniformly as unknown emission
- Recent update of biosphere from Sib4 up to -1053 GgS/yr

(Berry et. al 2013, Zumkehr et. al 2018.)

Updates on $COS\&CS_2$ anthropogenic emissions

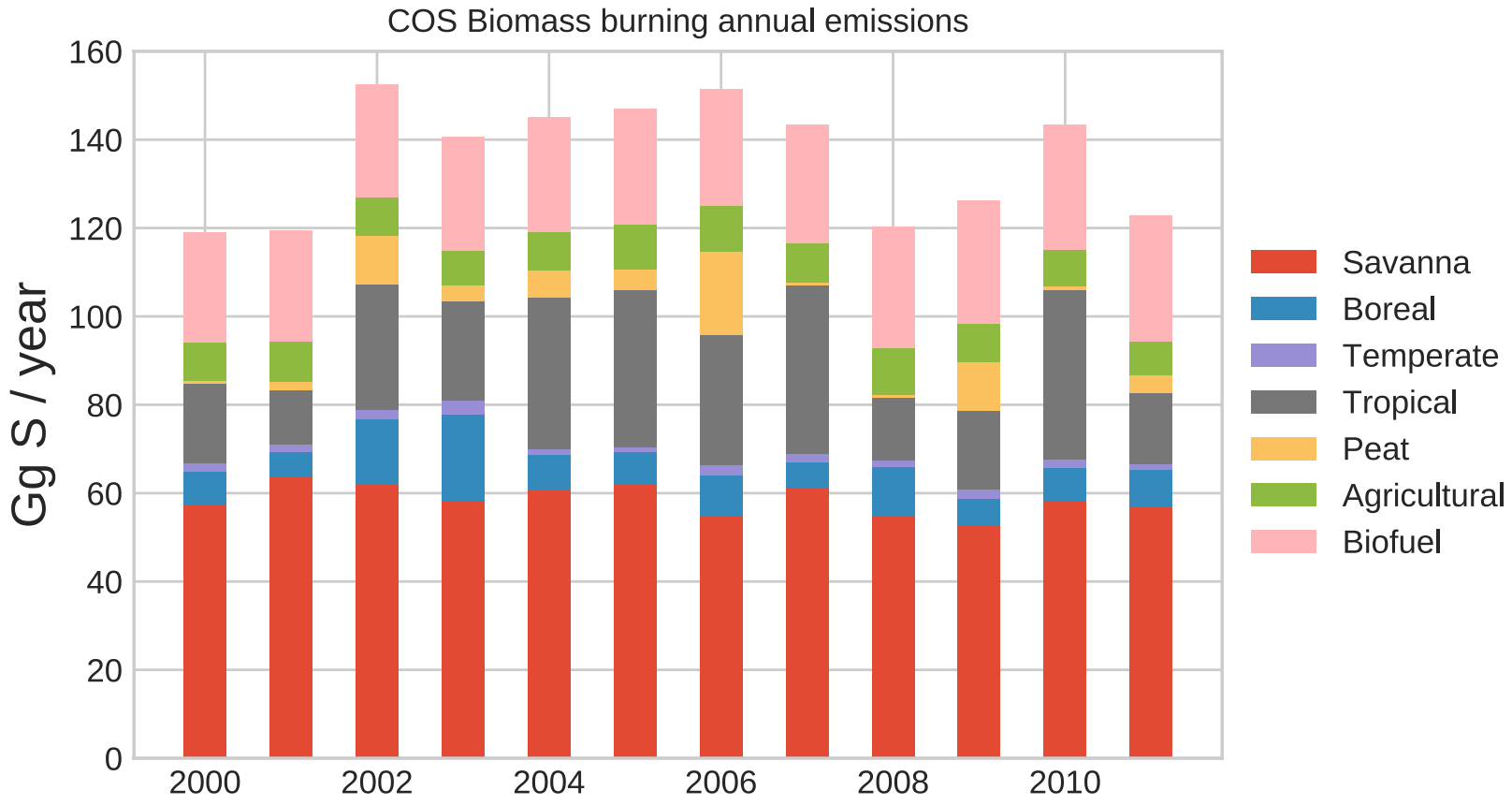




- Split total COS anthropogenic emissions to COS and CS_2 direct emissions

(Zumkehr et. al, 2018)

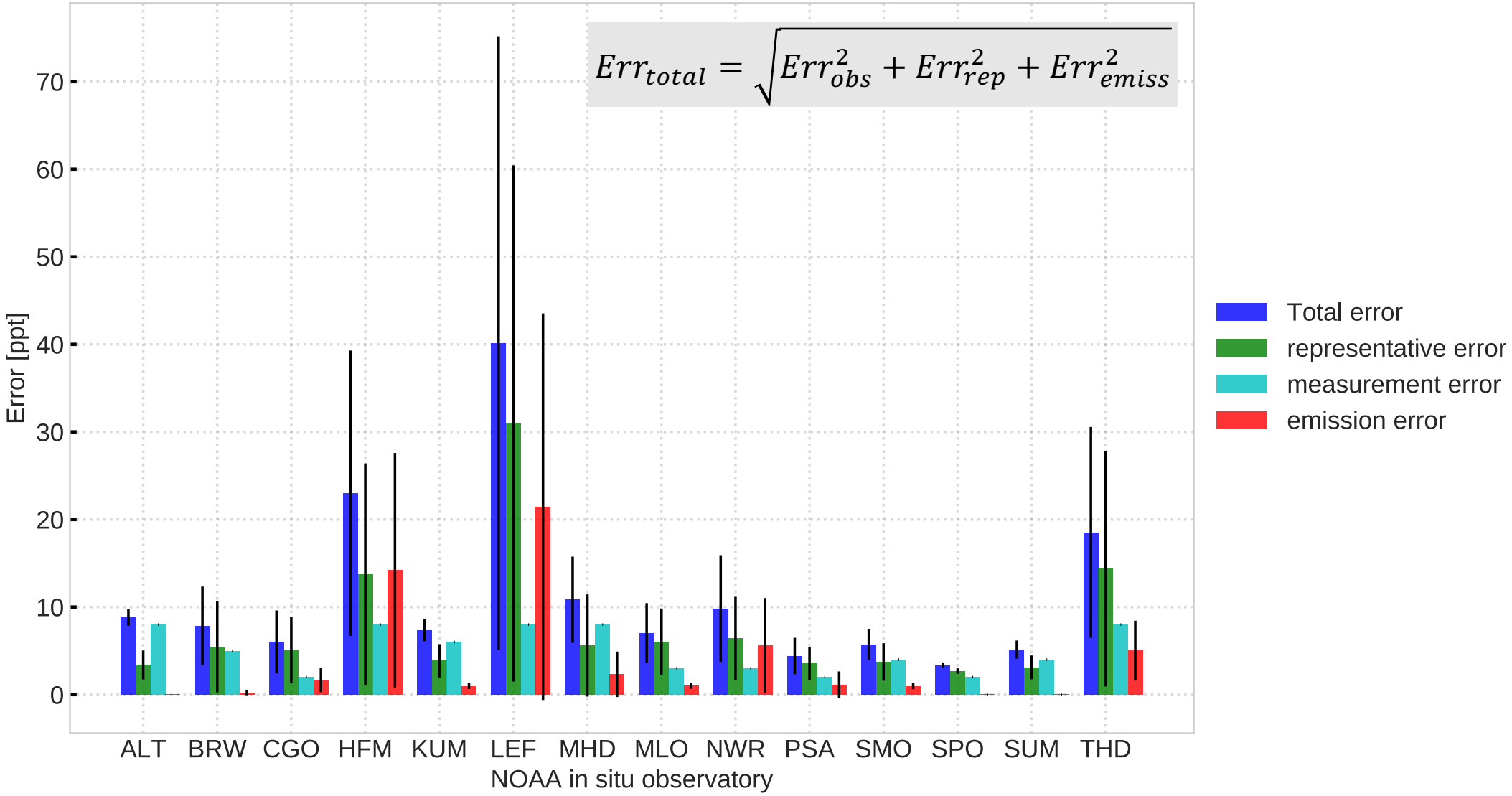
Updates on COS biomass emissions



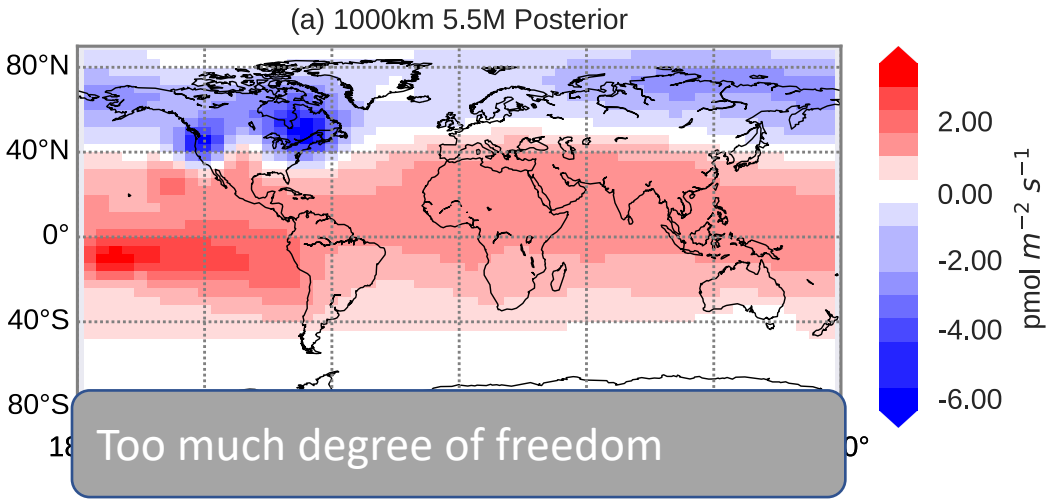
- Updated Emission factors for GFED 4.1 inventory
- Biofuel update in South Asia for CEDS inventory

(Andreae 2019, Fernandes et.al, 2007)

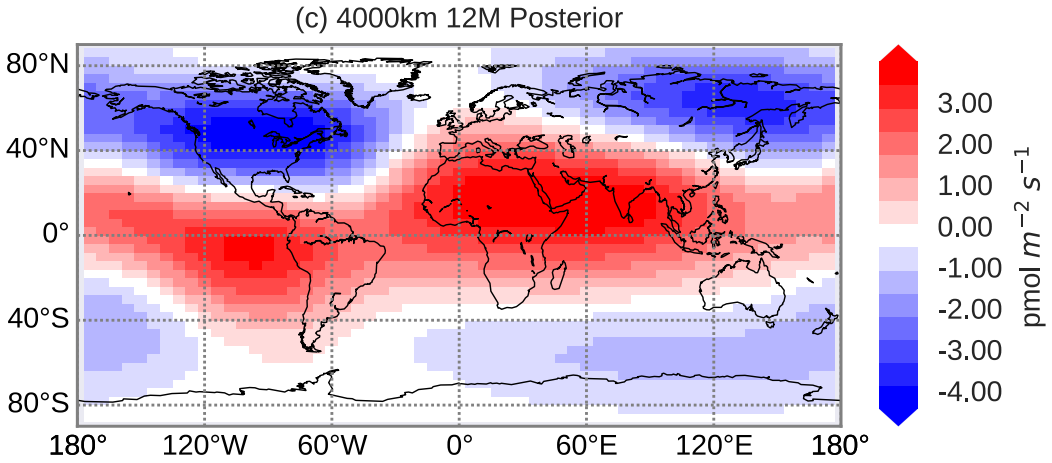
Error settings in inversions at NOAA stations



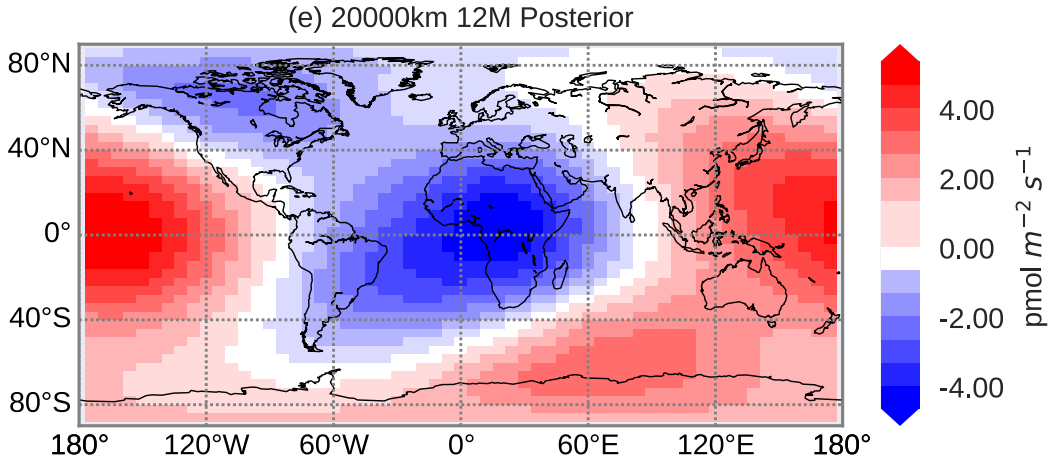
Unknown emission with different correlation settings



Too much degree of freedom



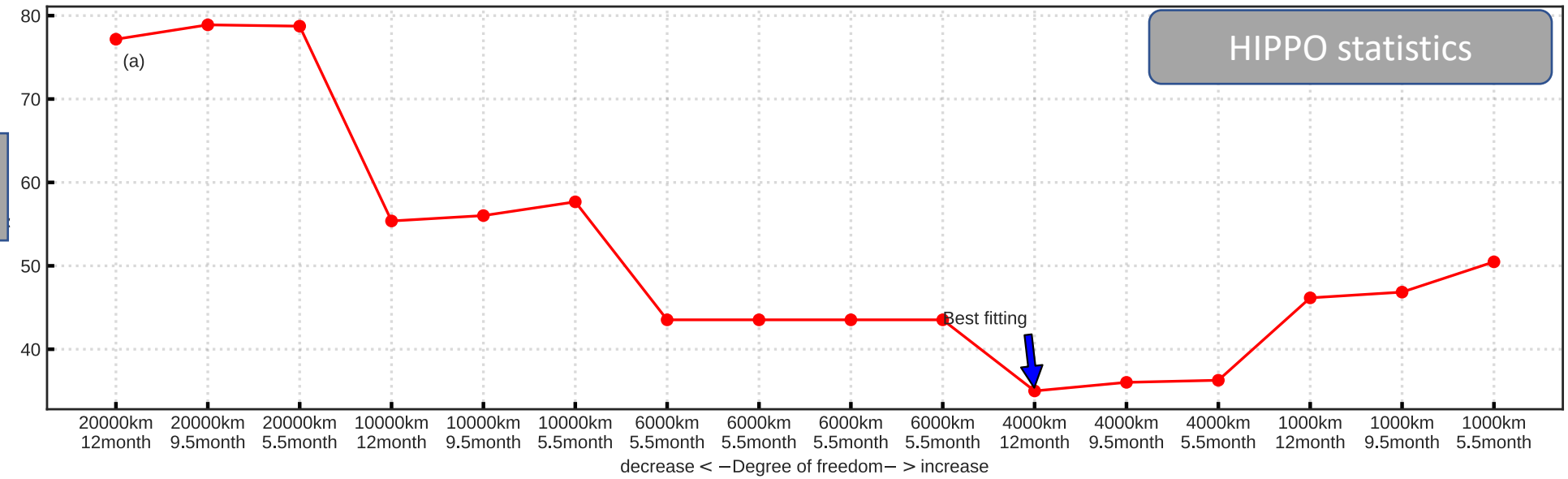
good-fitting correlation: 4000 km and 12 months



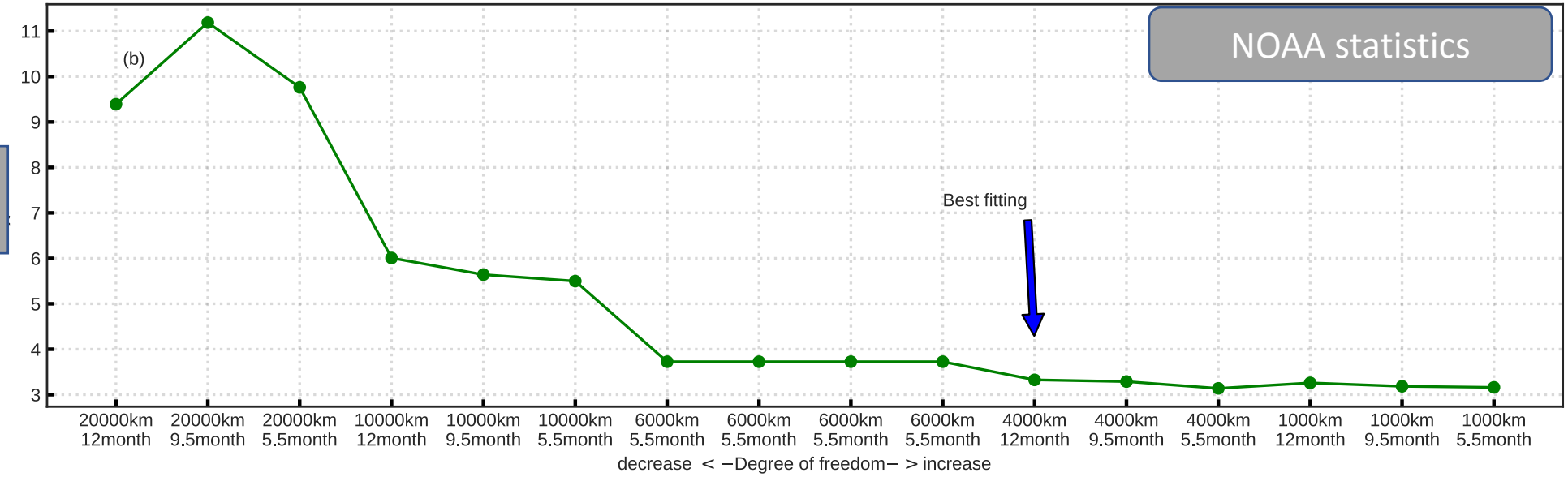
Totally different pattern

Spatial and temporal correlation test

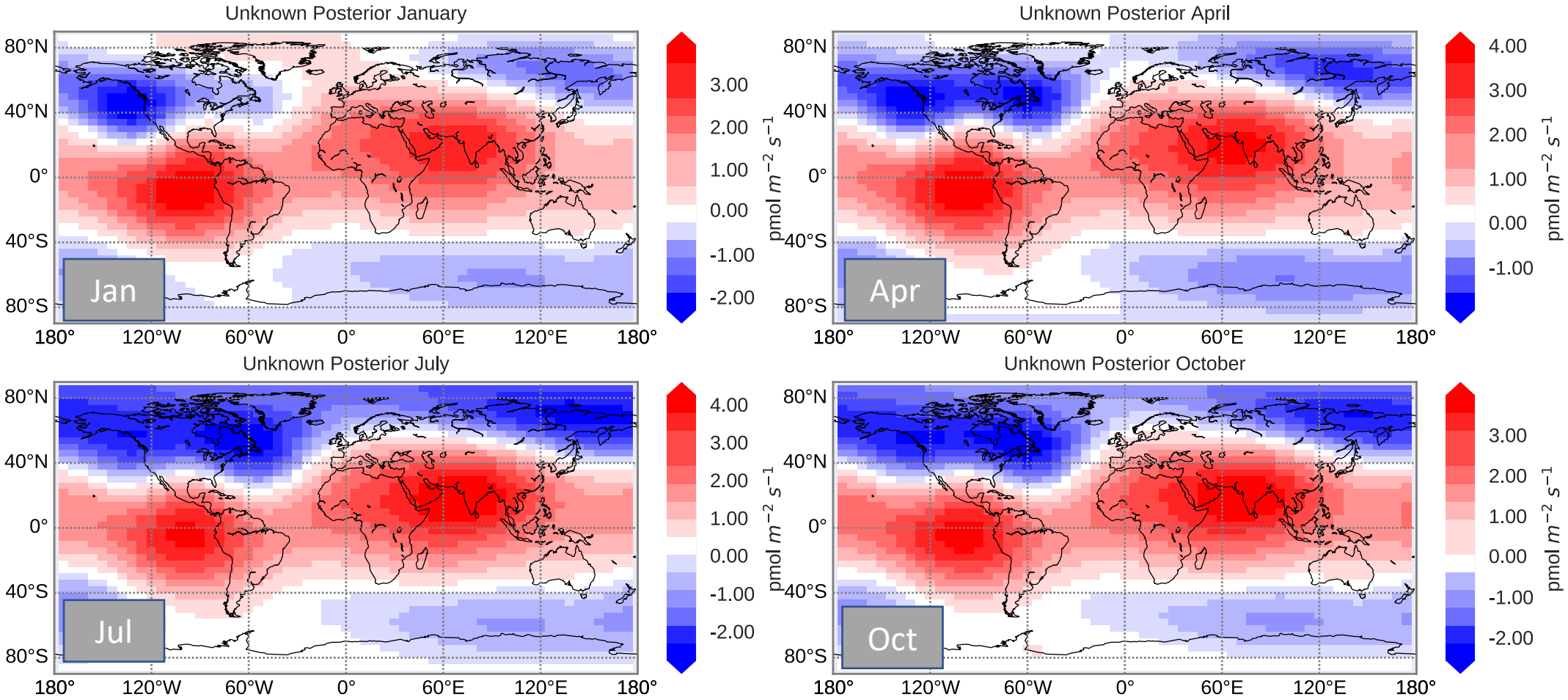
χ^2



χ^2



Optimized unknown emission: seasonal cycle

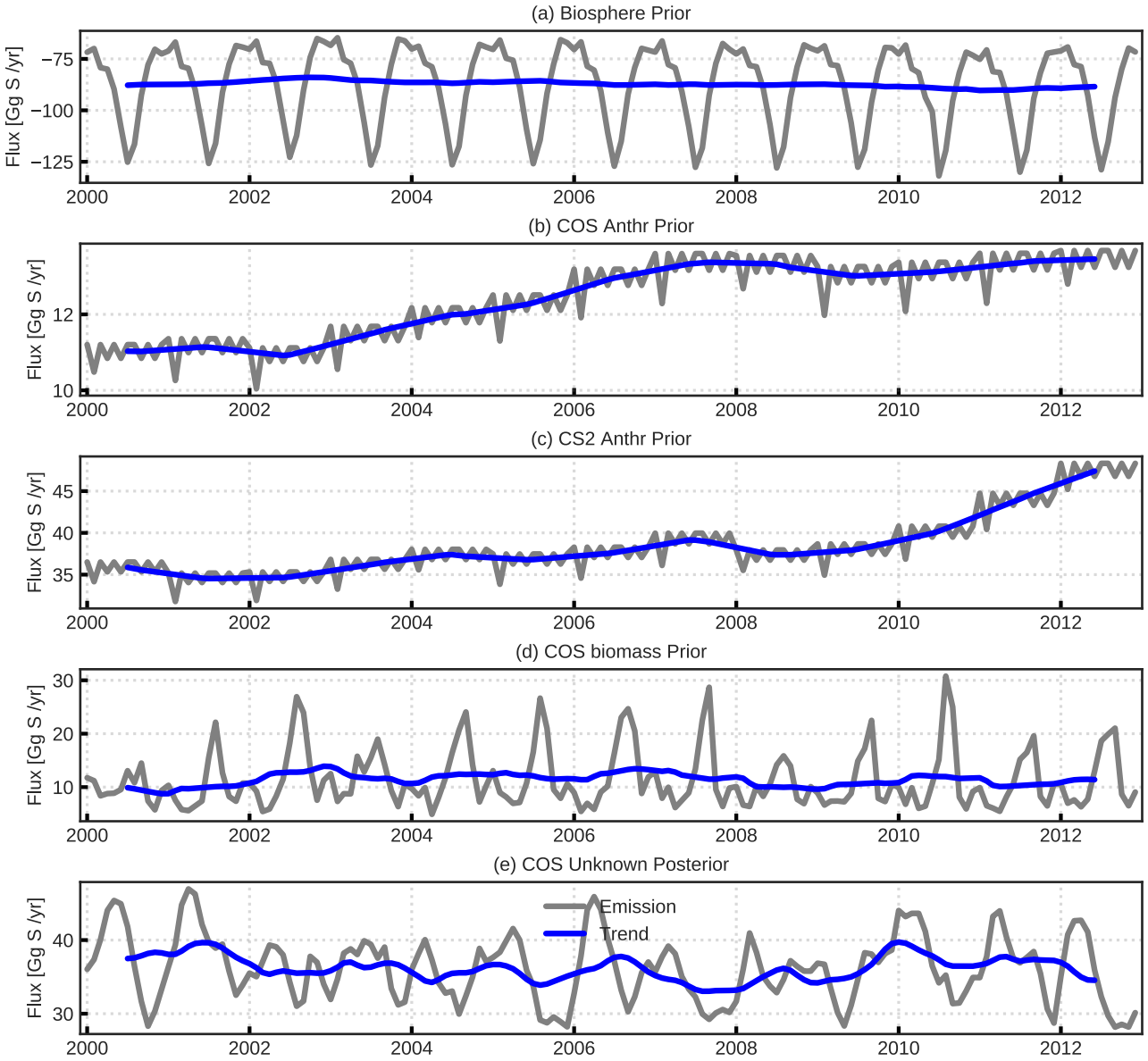


1: missing sources or less sinks in tropics

2: more sinks or less sources in high latitudes in North Hemisphere

3: July shows strong seasonal signal of unknown emissions

13 years emission trend: prior and optimized unknown

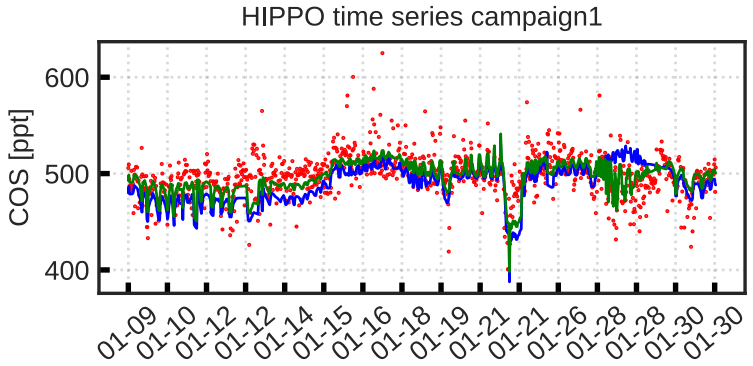


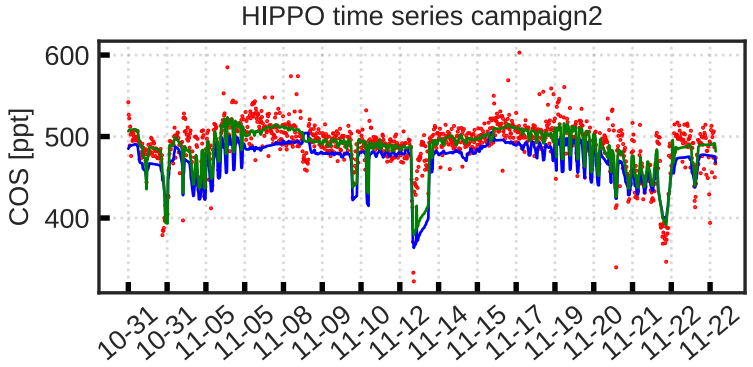
- $y_{emiss}(t) = y_{trend}(t) + y_{season}(t) + y_{noise}(t)$
- season = 365 days
- Trend of COS unknown Posterior is the result of balancing global budget

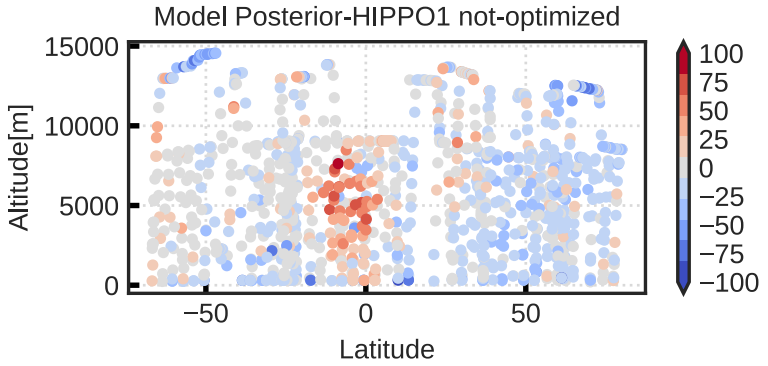
Emission in Gray

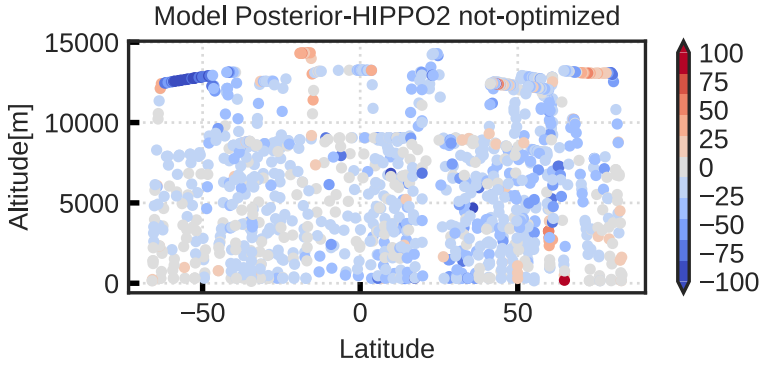
Trend in Blue

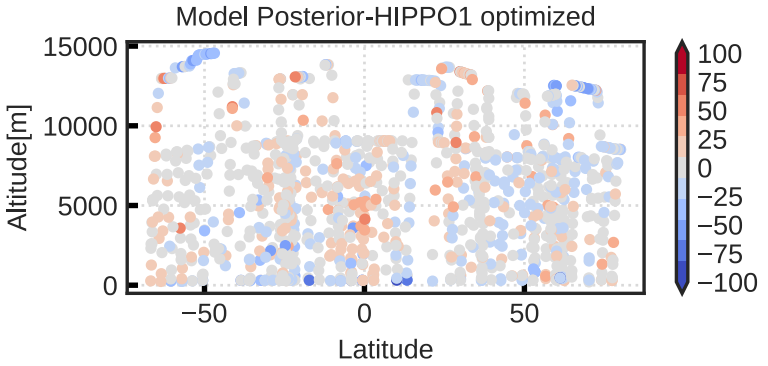
Hippo validation vs HIPPO-optimized

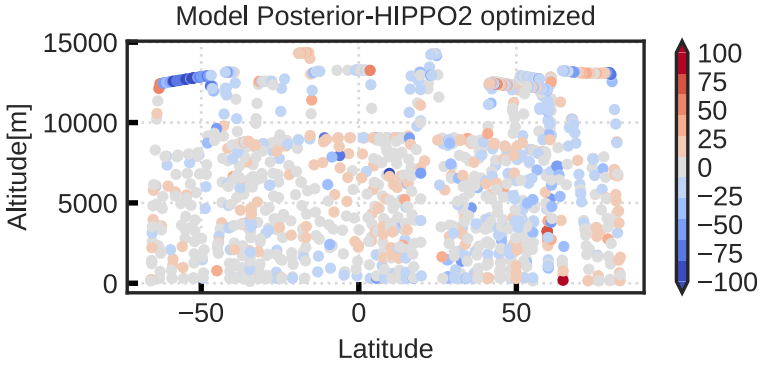












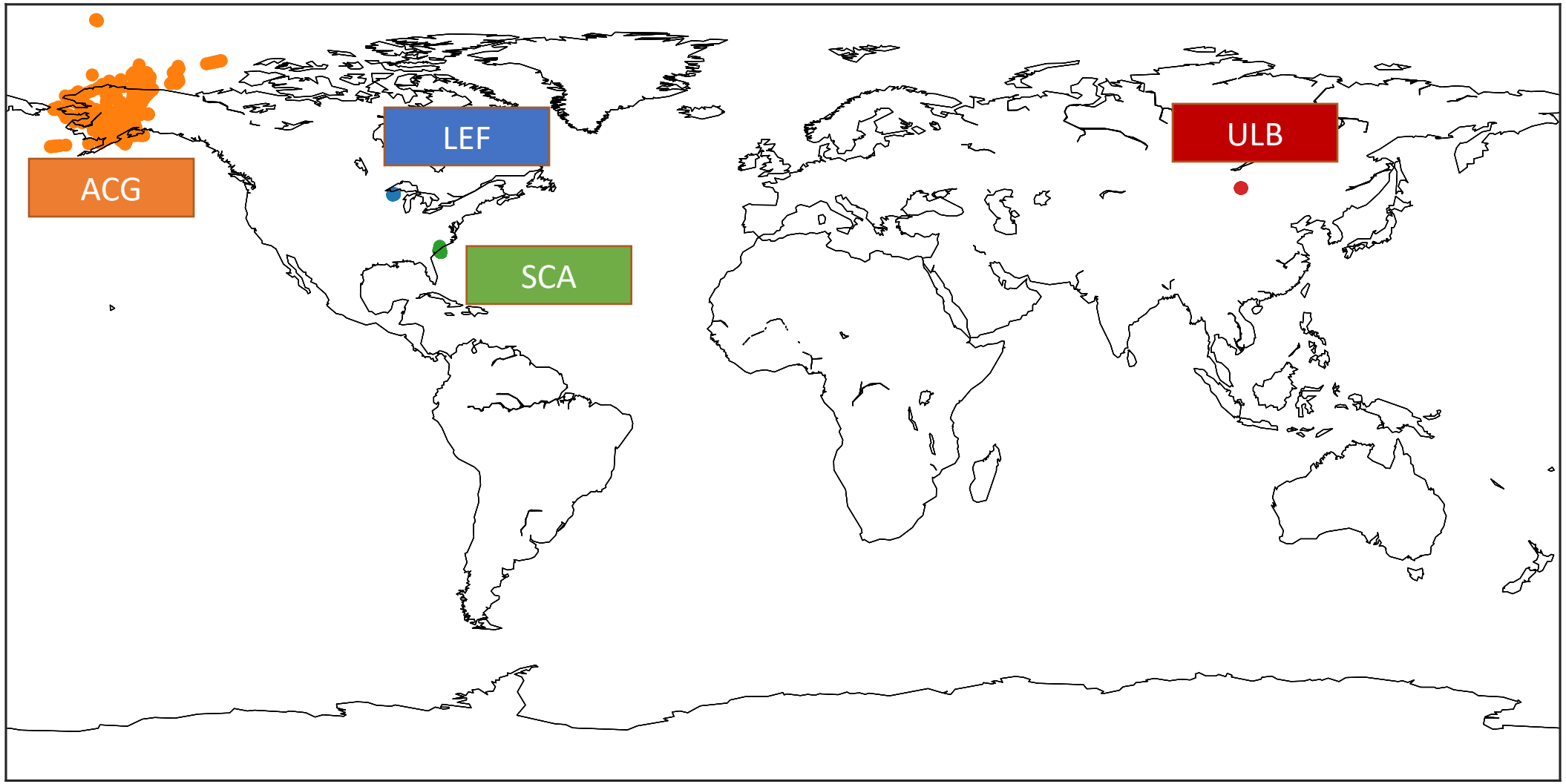
Green: HIPPO optimized
Blue: HIPPO only for validation

HIPPO not optimized

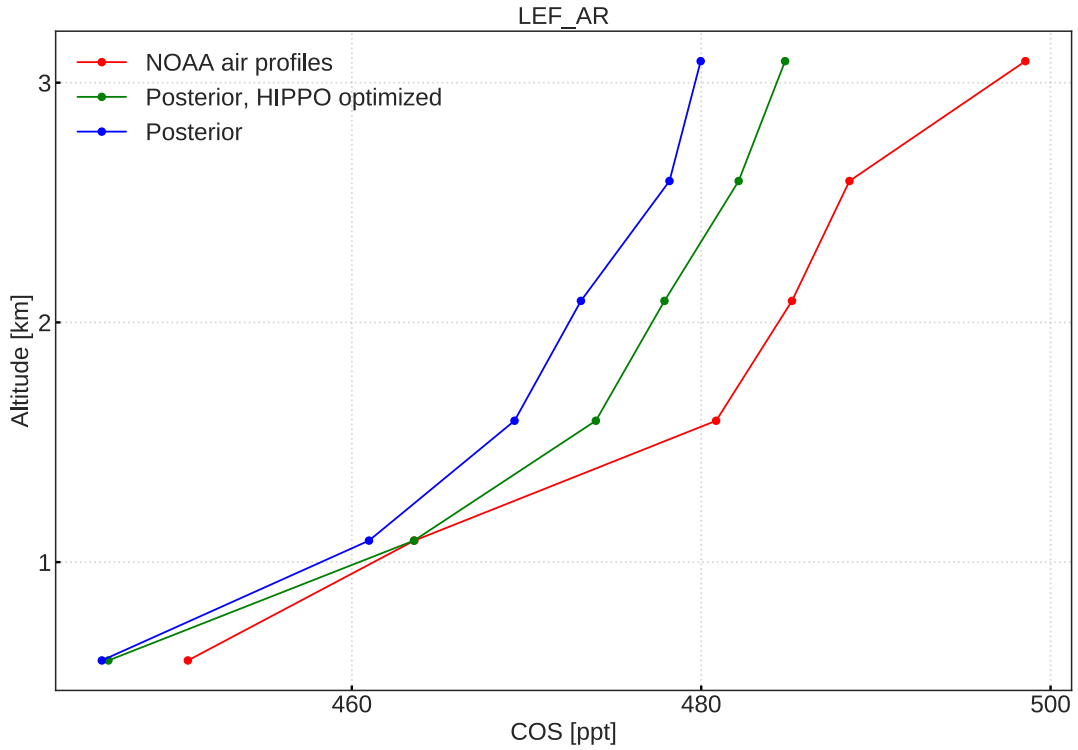
HIPPO optimized

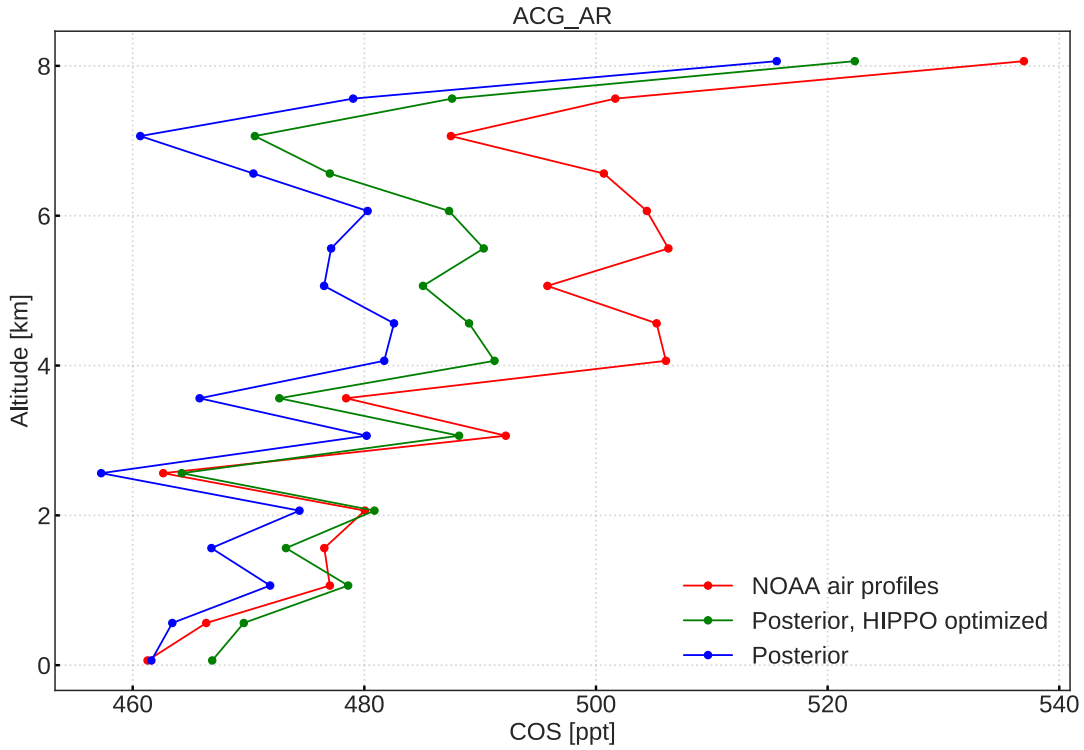
Anyway, HIPPO is not recommended to optimize emission fluxes due to short time range and limited spatial coverage.

NOAA airborne profile validation



NOAA airborne profile validation



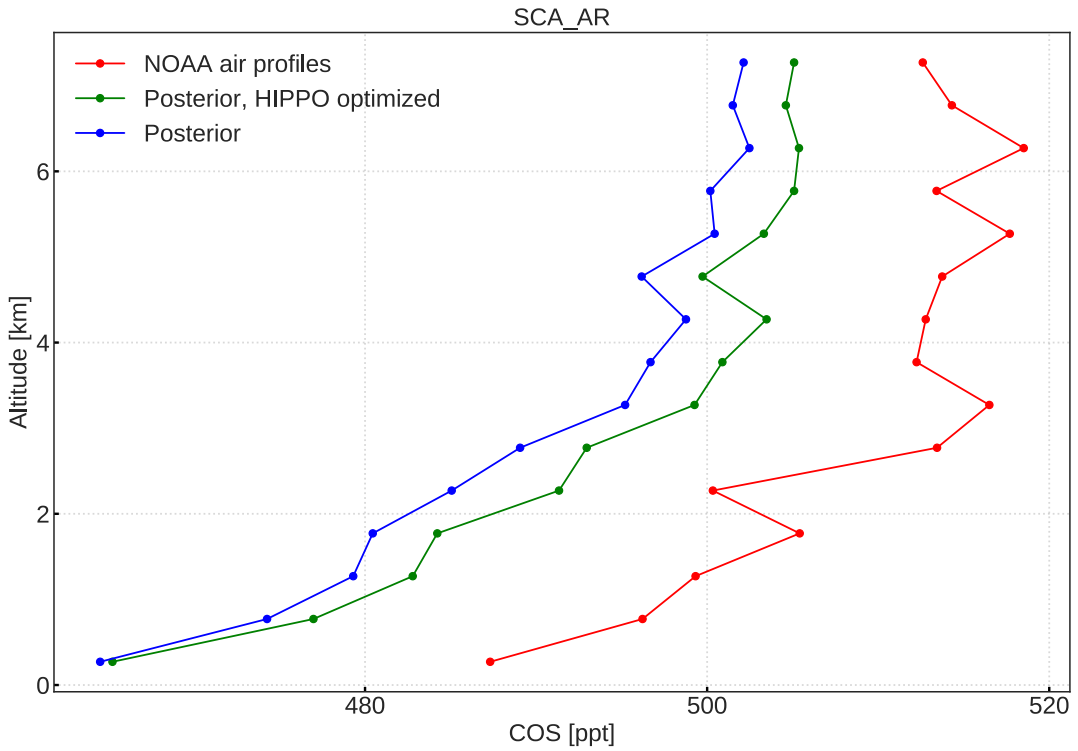


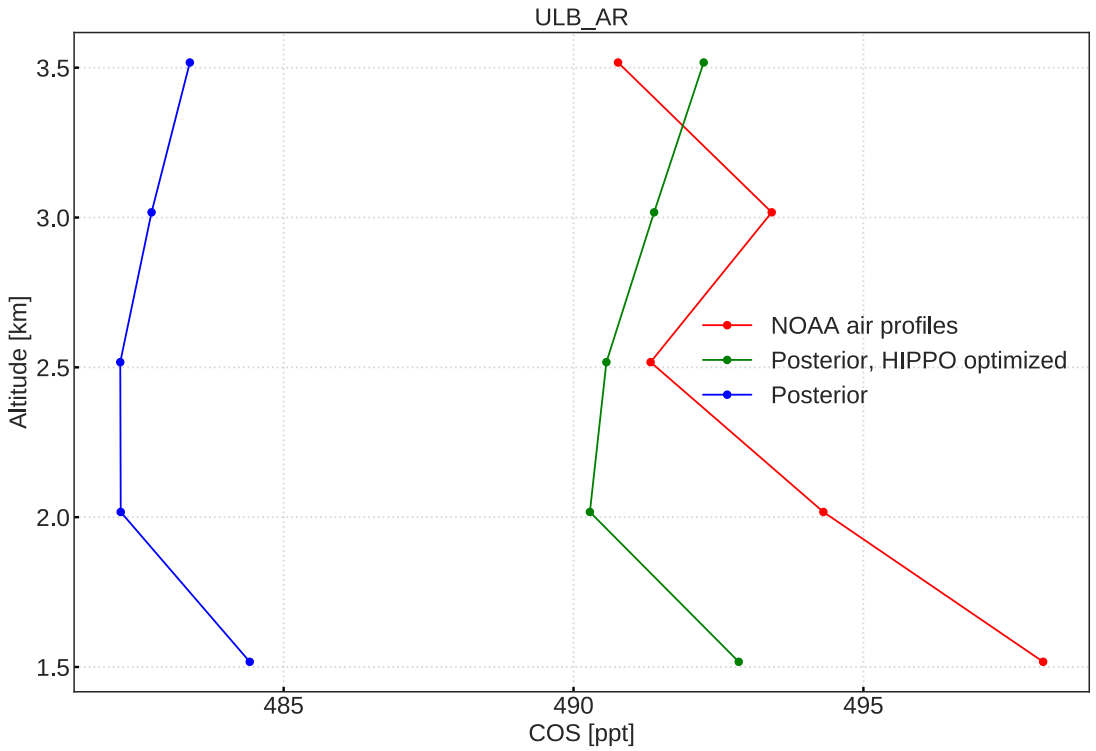
Central North America

Alaska

Profiles are averaged every 500 meters vertically.

NOAA airborne profile validation





Coastal North America

Mongolia in Asia

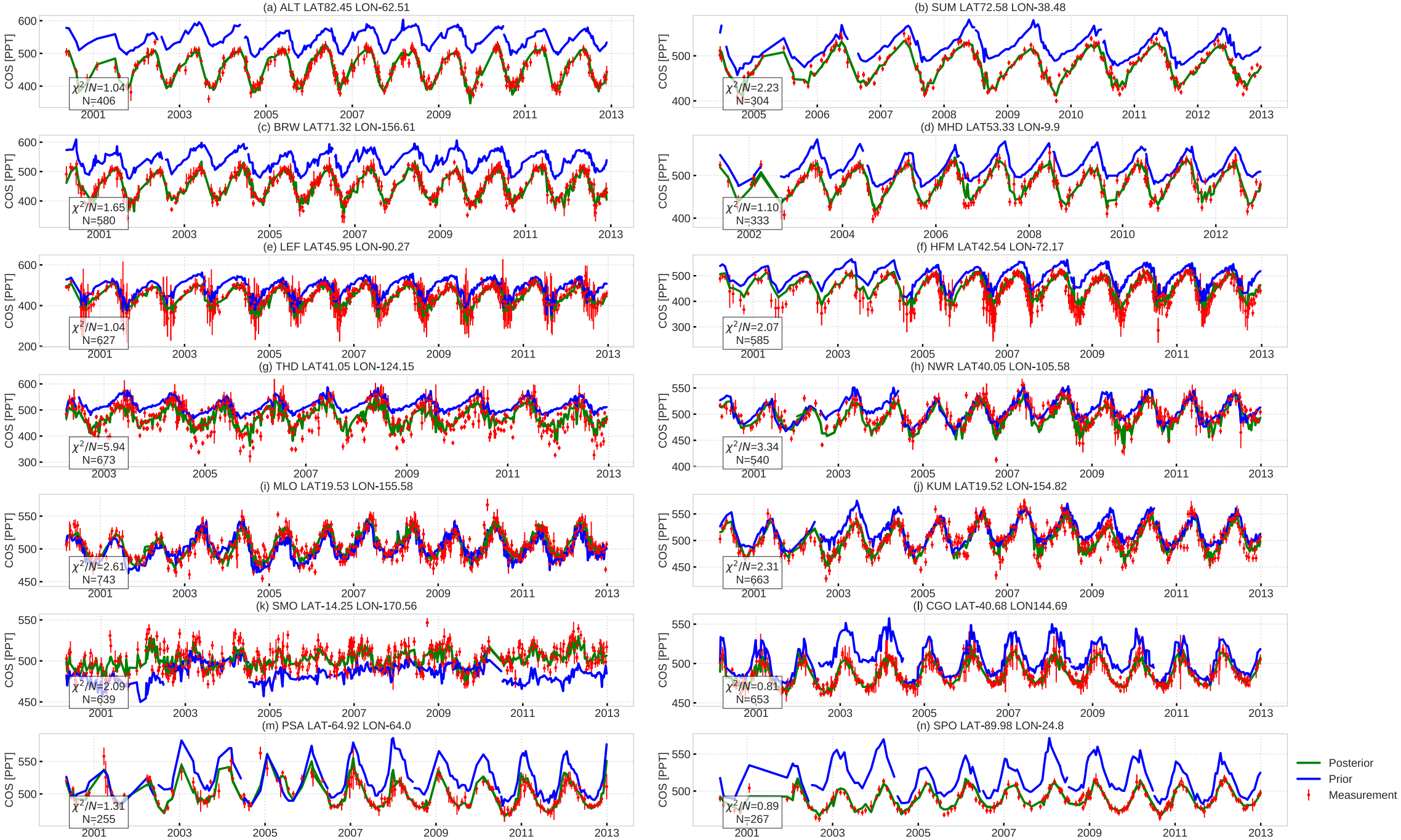
Profiles are averaged every 500 meters vertically.

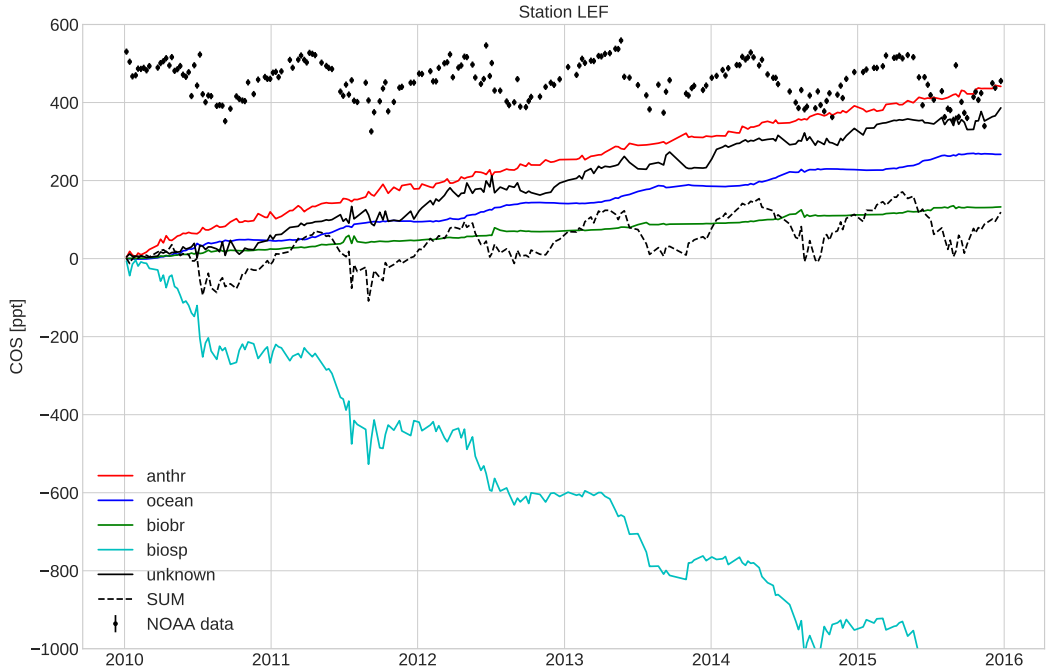
Conclusions

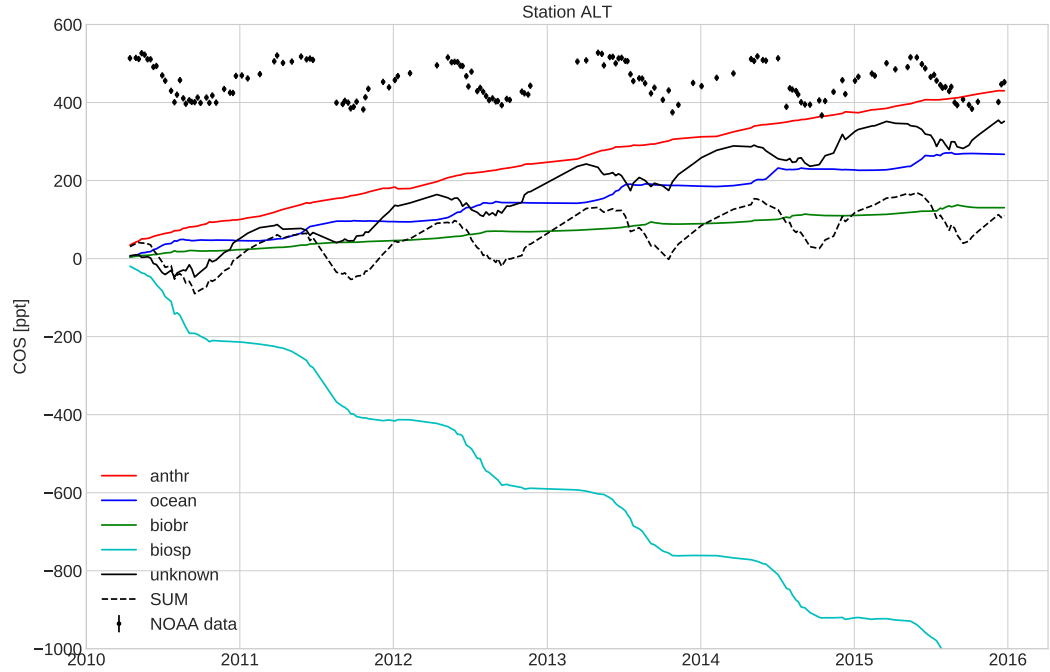
- Missing emissions from tropical regions, and more sinks in high latitude in Northern Hemisphere
- NOAA surface observations partially constrain emissions but we need more constraints in free troposphere
- Assimilating HIPPO improves fitting with independent data
- Future plan to assimilate satellite data

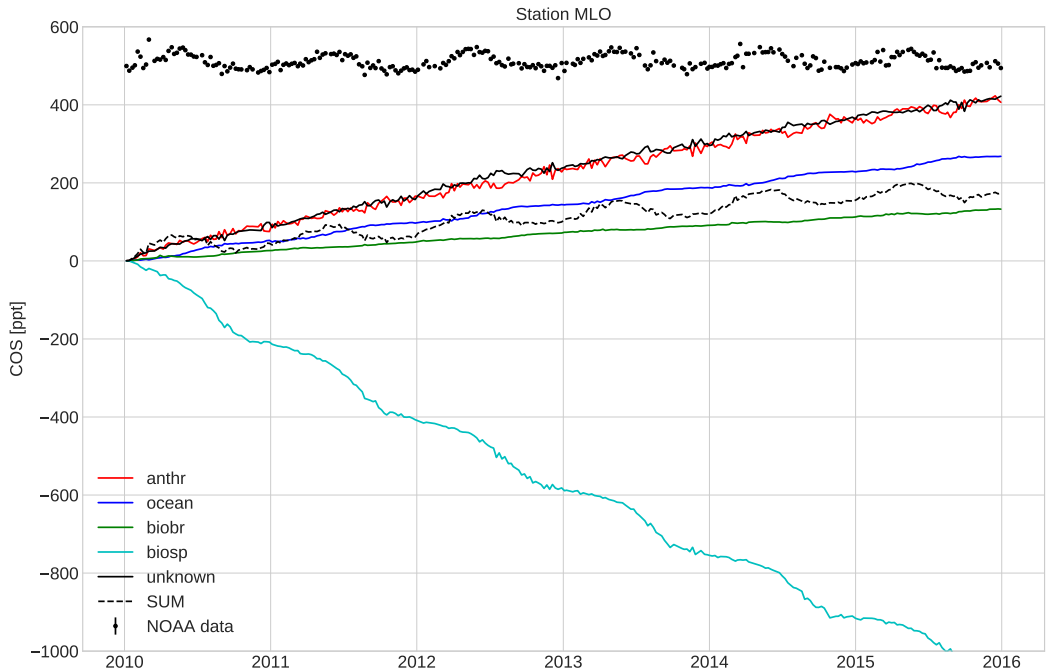
Thank you for your attention!

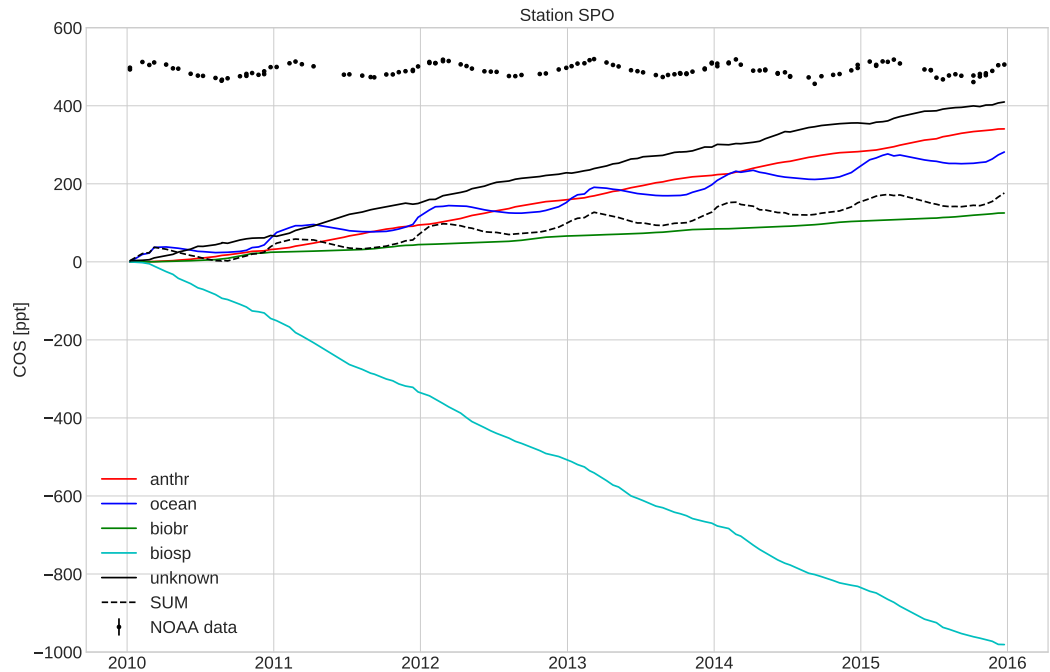
- Questions?


(a) ALT LAT82.45 LON-62.51
χ²/N=1.04
N=406
(b) SUM LAT72.58 LON-38.48
χ²/N=2.23
N=304
(c) BRW LAT71.32 LON-156.61
χ²/N=1.65
N=580
(d) MHD LAT53.33 LON-9.9
χ²/N=1.10
N=333
(e) LEF LAT45.95 LON-90.27
χ²/N=1.04
N=627
(f) HFM LAT42.54 LON-72.17
χ²/N=2.07
N=585
(g) THD LAT41.05 LON-124.15
χ²/N=5.94
N=673
(h) NWR LAT40.05 LON-105.58
χ²/N=3.34
N=540
(i) MLO LAT19.53 LON-155.58
χ²/N=2.61
N=743
(j) KUM LAT19.52 LON-154.82
χ²/N=2.31
N=663
(k) SMO LAT-14.25 LON-170.56
χ²/N=2.09
N=639
(l) CGO LAT-40.68 LON144.69
χ²/N=0.81
N=653
(m) PSA LAT-64.92 LON-64.0
χ²/N=1.31
N=255
(n) SPO LAT-89.98 LON-24.8
χ²/N=0.89
N=267
COS [PPT]
Posterior
Prior
Measurement

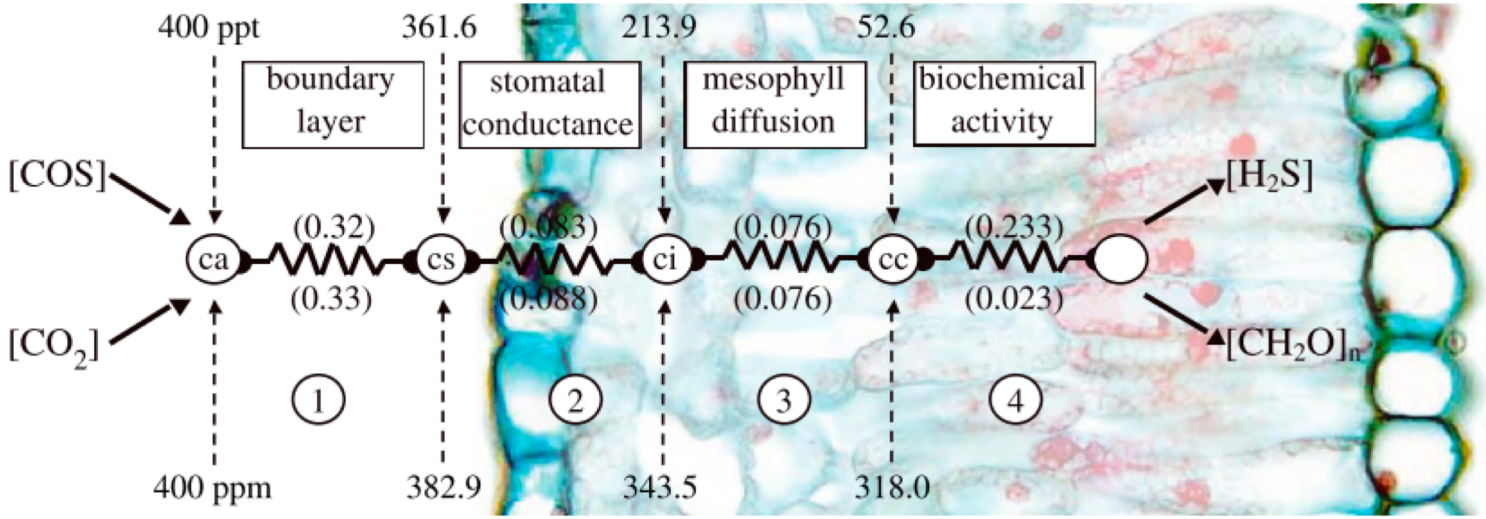

Station LEF
COS [ppt]
600
400
200
0
−200
−400
−600
−800
−1000
2010
2011
2012
2013
2014
2015
2016
anthr
ocean
biobr
biosp
unknown
SUM
NOAA data


Station ALT
COS [ppt]
600
400
200
0
−200
−400
−600
−800
−1000
2010
2011
2012
2013
2014
2015
2016
anthr
ocean
biobr
biosp
unknown
SUM
NOAA data


Station MLO
COS [ppt]
600
400
200
0
−200
−400
−600
−800
−1000
2010
2011
2012
2013
2014
2015
2016
anthr
ocean
biobr
biosp
unknown
SUM
NOAA data


Station SPO
COS [ppt]
600
400
200
0
−200
−400
−600
−800
−1000
2010
2011
2012
2013
2014
2015
2016
anthr
ocean
biobr
biosp
unknown
SUM
NOAA data

Main sink: COS uptake by a leaf



(Berry et. al 2013)

- CO_2 is taken up by a leaf via photosynthesis and emitted via respiration
- COS is only taken up by a leaf