

How well do we understand oceanic emissions of carbonyl sulfide?

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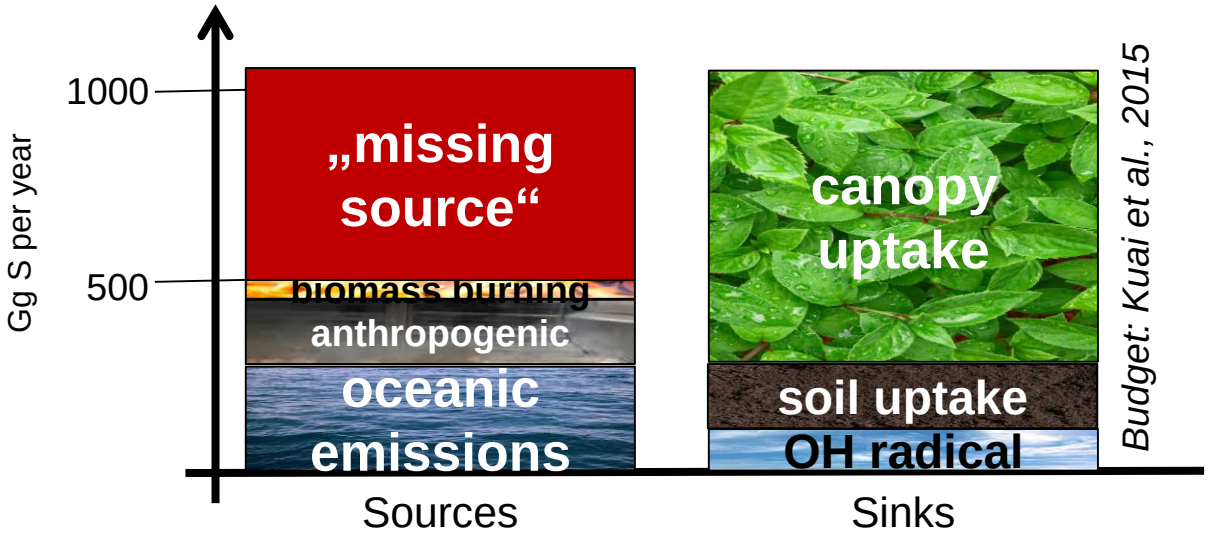
Why care about oceanic OCS emissions?

- OCS is **major precursor for stratospheric sulfur** in volcanically quiescent periods => tropospheric budget and dynamics need to be understood to quantify and predict entrainment into stratosphere

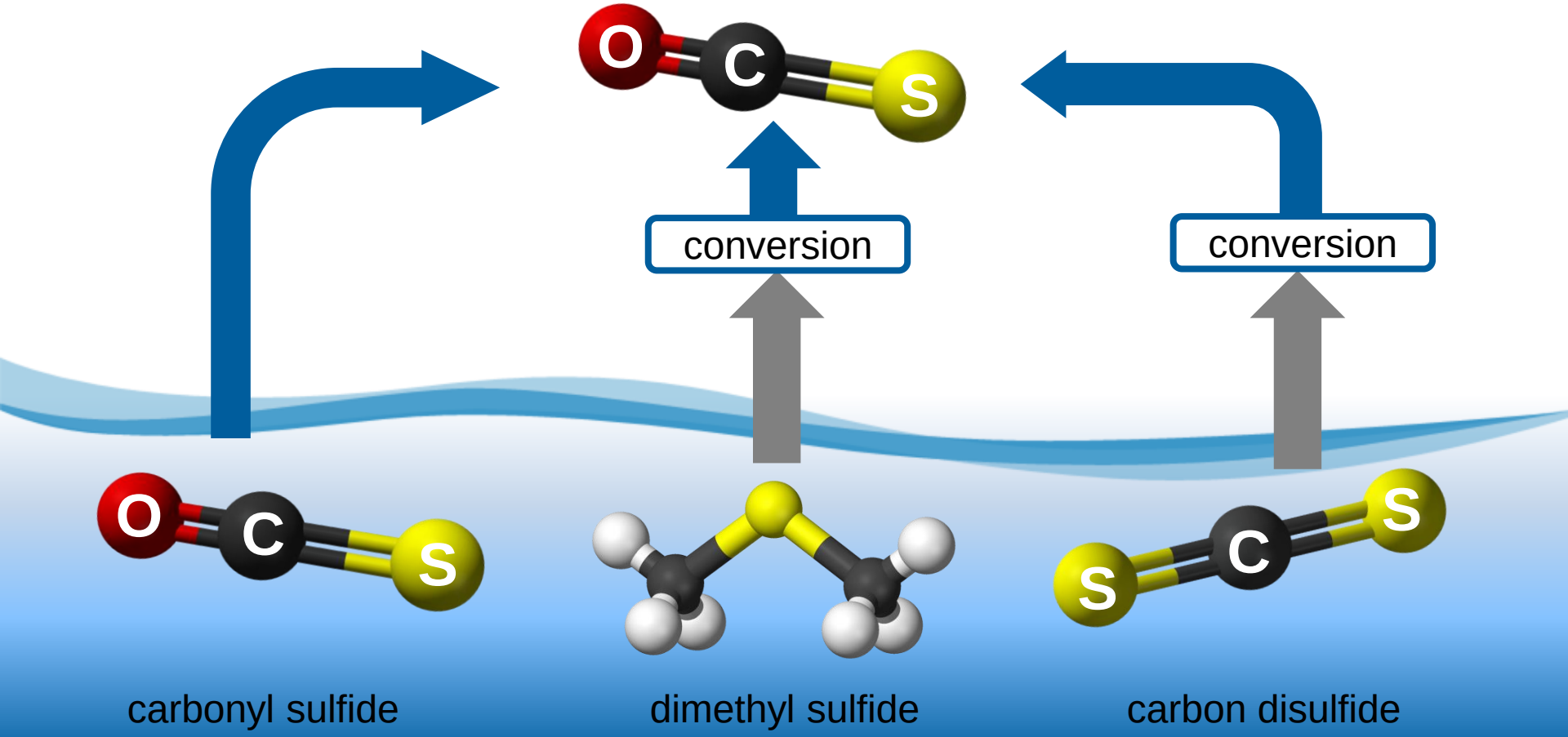
- The ocean is a major source of OCS to the atmosphere



- **But:** Currently large gap in tropospheric budget (~500-800 Gg S yr^{-1})
  ⇒ Several top-down studies suggest missing source from ocean

Direct & indirect OCS emissions

Total uncertainty = uncertainty OCS + uncertainty DMS + uncertainty CS_2



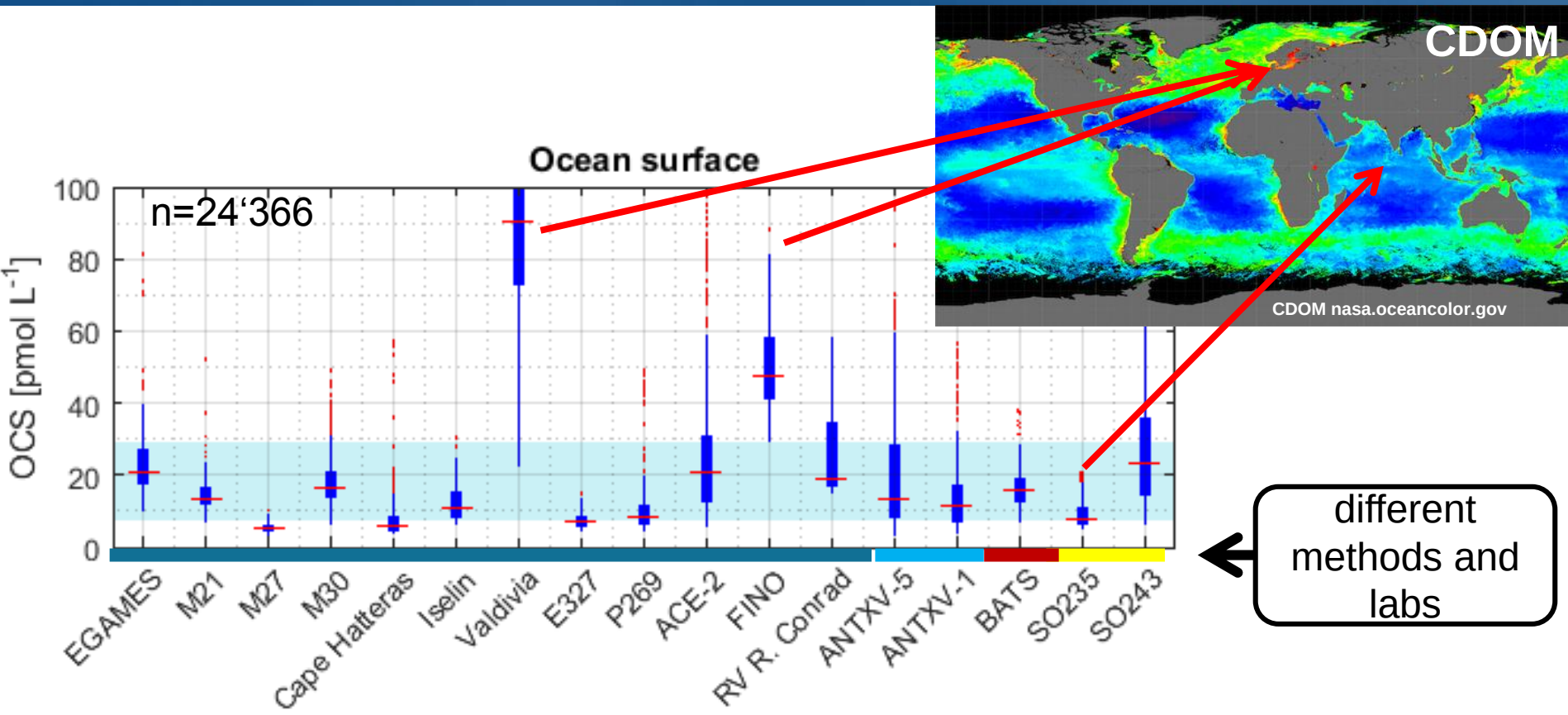
How to assess uncertainty?

- Emissions calculated from concentration gradient between water and air
- Database from literature + own data + digitalized
  - **Number** of measurements available
  - **Locations/season** covered by measurements
  - **Consistency** of measurements across different methods
  - **Process** understanding
  - Indirect emissions: **conversion** to OCS

$\Rightarrow$ Uncertainty with respect to missing source

Database contributions from: M.O. Andreae, E. Atlas, H. Bingemer, G. Cutter, O. Flöck, M. von Hobe, G. Uher, V. Ulshöfer, X. Xu +digitalized from printouts (MBL) +own data

Direct emissions: OCS measurements

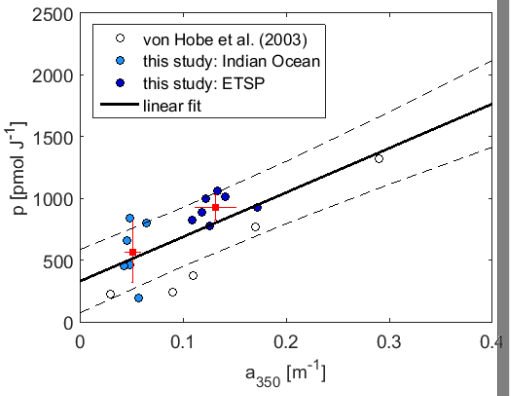


- Concentrations consistent across different labs/methods
- Locations cover large variability of major driver of marine production

Direct emissions: process parameterizations

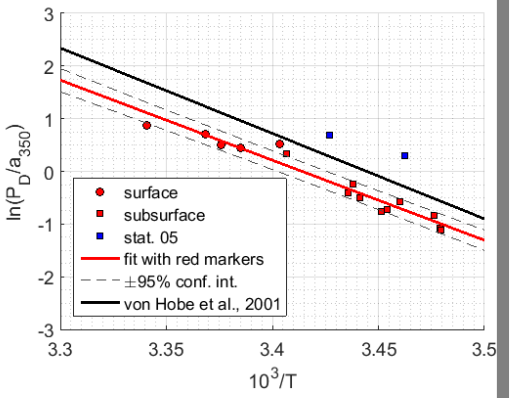
Important processes for OCS formation/degradation in seawater:

Photoproduction



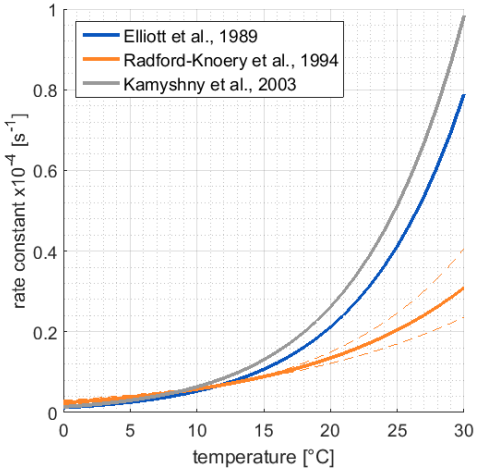
Lennartz et al., 2017 (mod.)

Dark production



Lennartz et al., in prep.

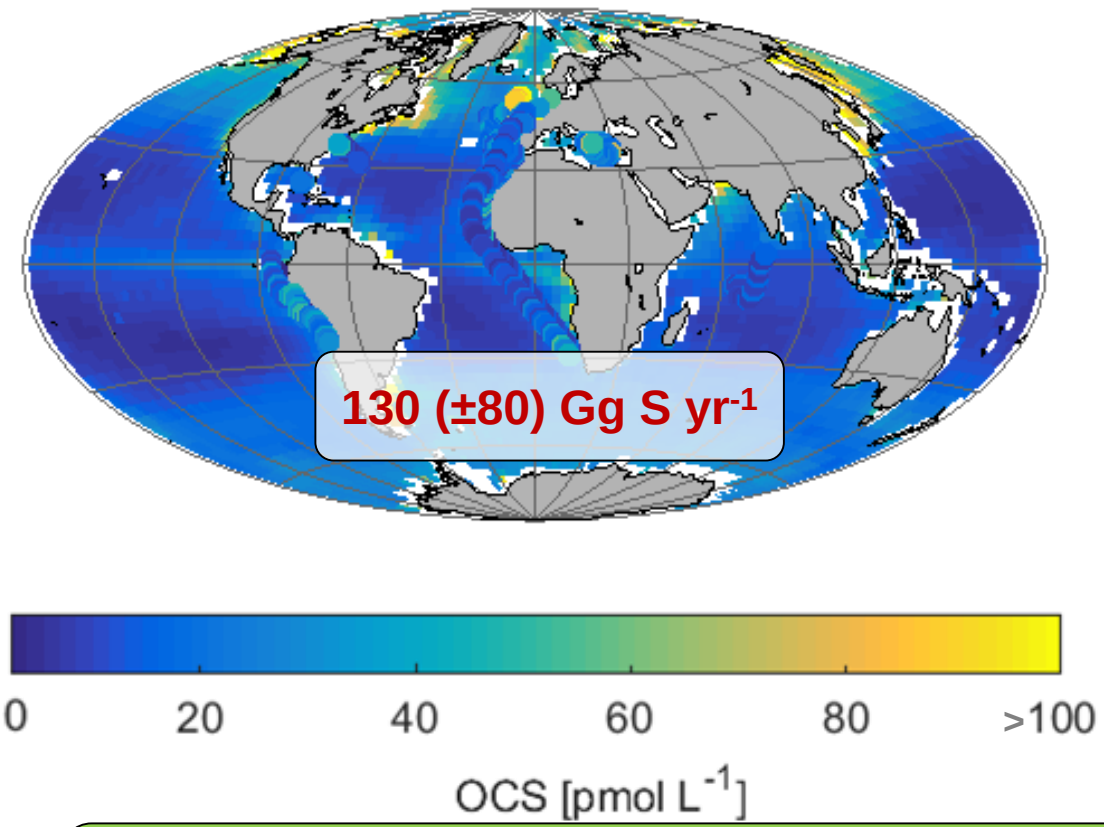
Hydrolysis

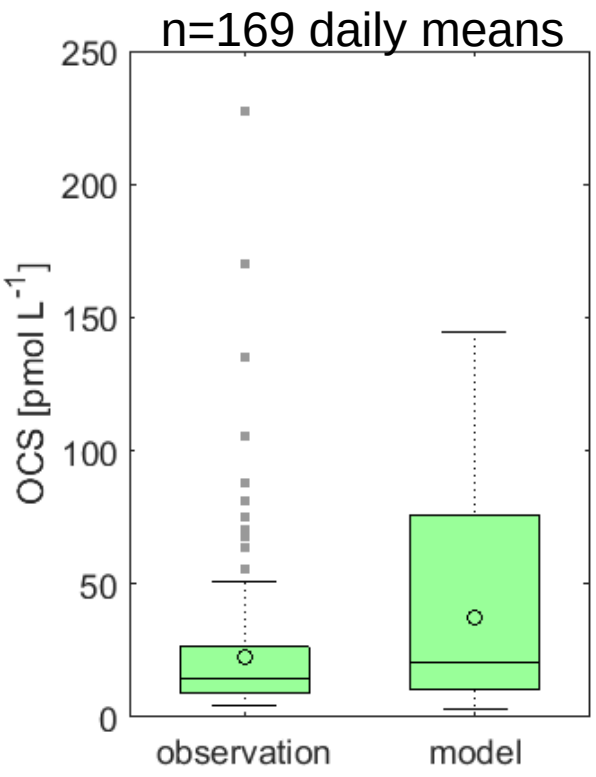


Whelan et al., 2017

Process parameterizations agree reasonably well across different oceanic regimes/laboratory studies.

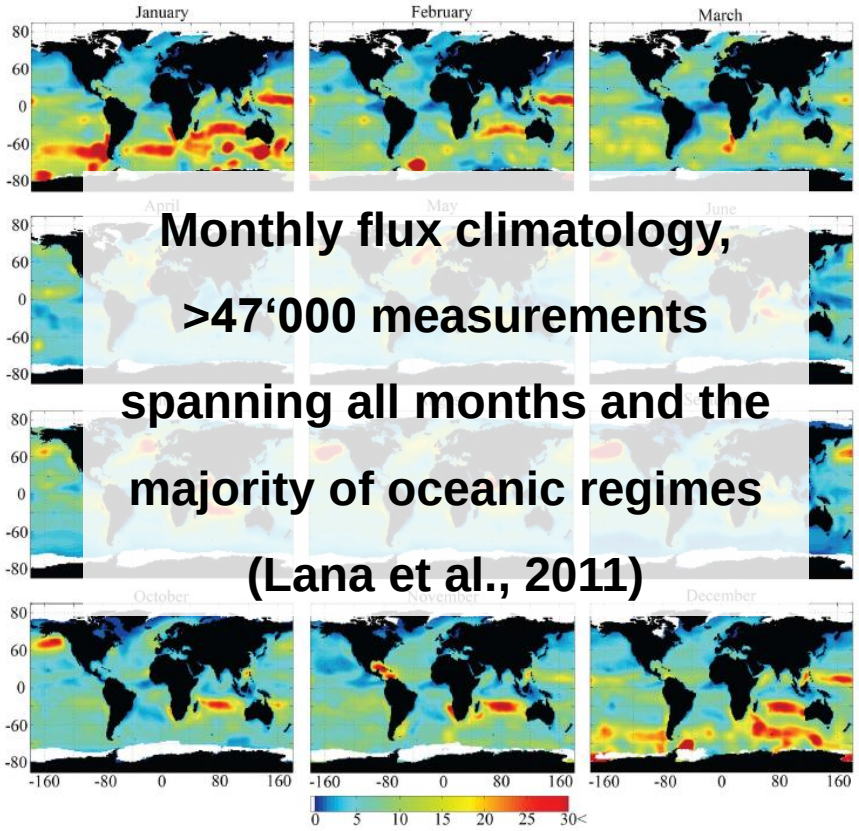
Direct emissions: model calculations





Process parameterizations yield simulated surface concentrations consistent with observations.

DMS: main uncertainty from conversion



Monthly flux climatology, >47‘000 measurements spanning all months and the majority of oceanic regimes (Lana et al., 2011)

Process understanding and available observations are sufficient for a DMS emission estimate.

28.1 (17.6-34.4) Tg S yr^{-1} DMS

OH oxidation (0.7%, Barnes et al., 1994)

80 (65-110) Gg S yr^{-1} OCS

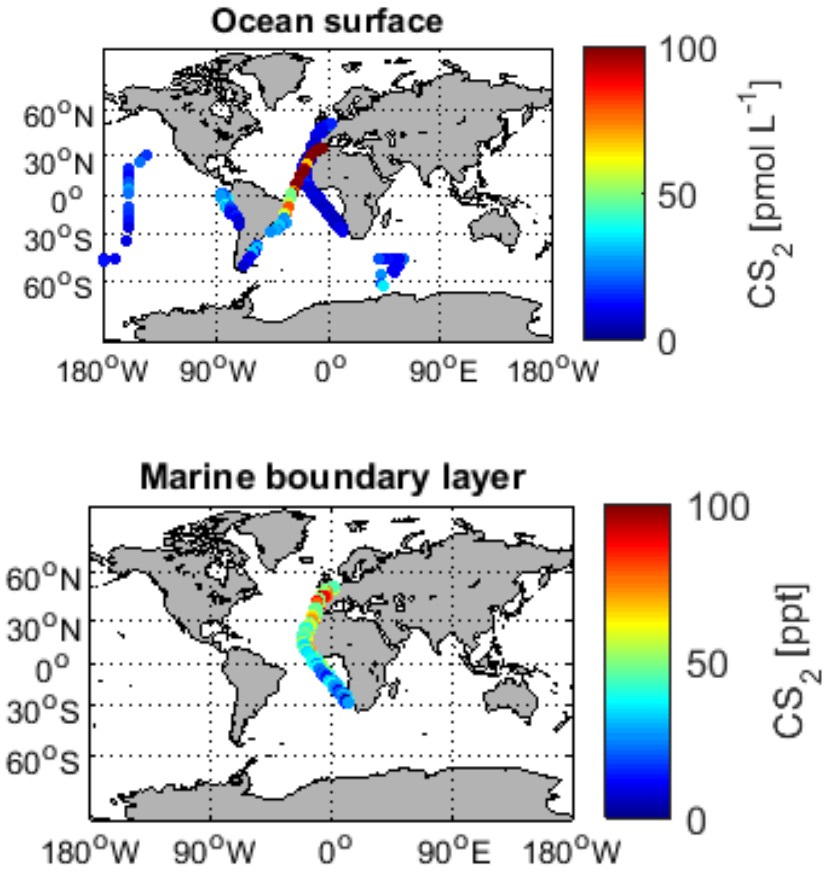
Uncertainty in conversion factor:

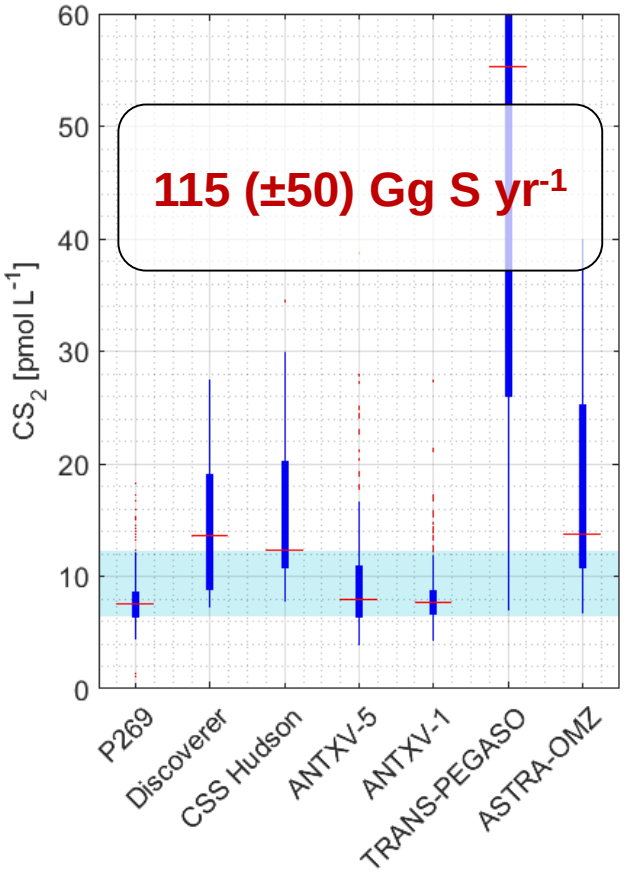
- No validation under environmental conditions
- likely smaller than 0.7% in presence of NOx

CS_2: main uncertainty from emission estimate

843 measurements, 1255 measurements, 7 cruises

3 cruises





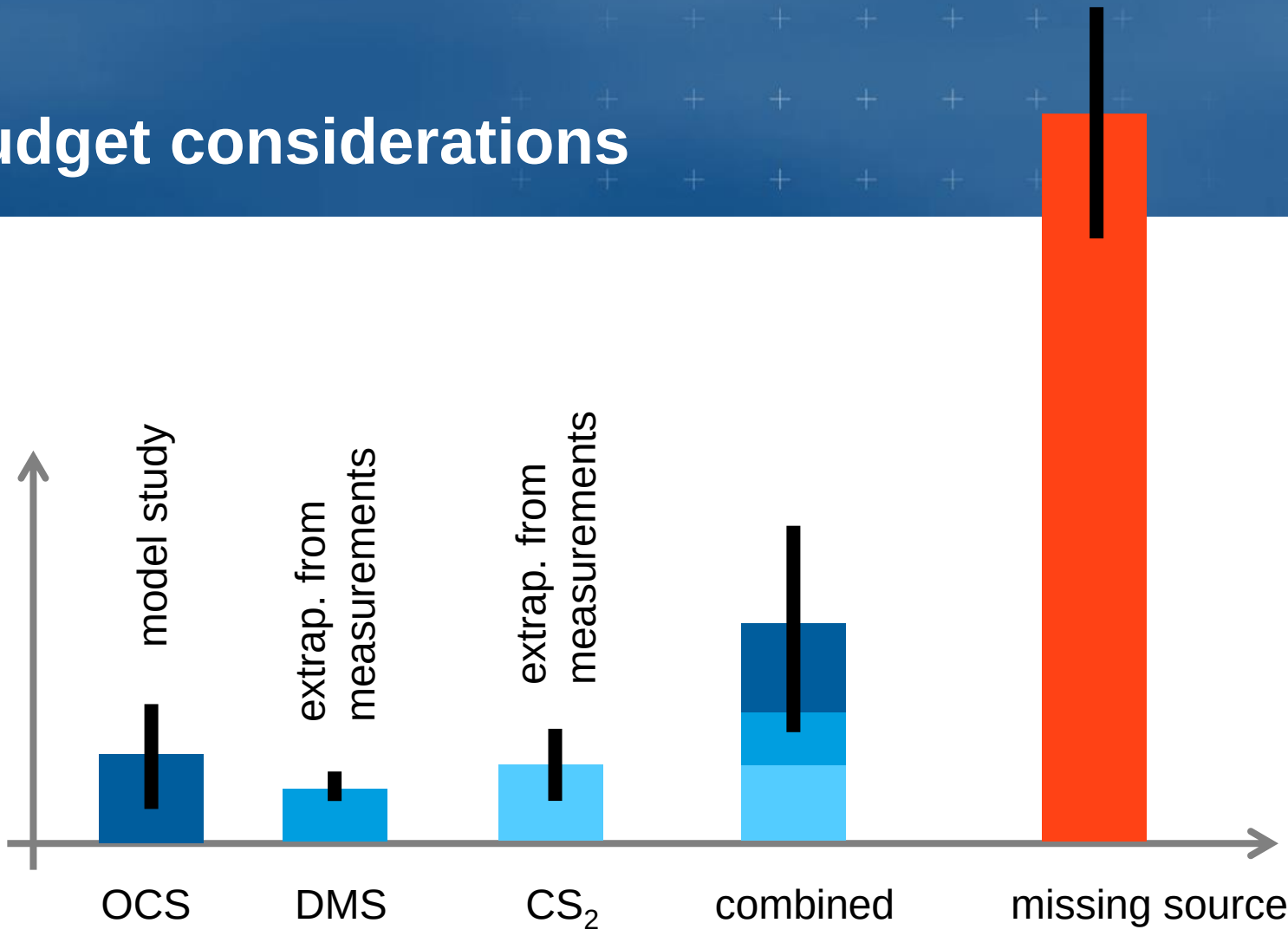
Comparably few measurements, but conversion factor has been tested several times (Chin 1992, Stickel et al., 1993).

Database S. Lennartz

Summary – level of understanding

	Oceanic emission	Conversion to OCS
OCS	medium	-
DMS	good	poor
CS_2	poor	good

Budget considerations

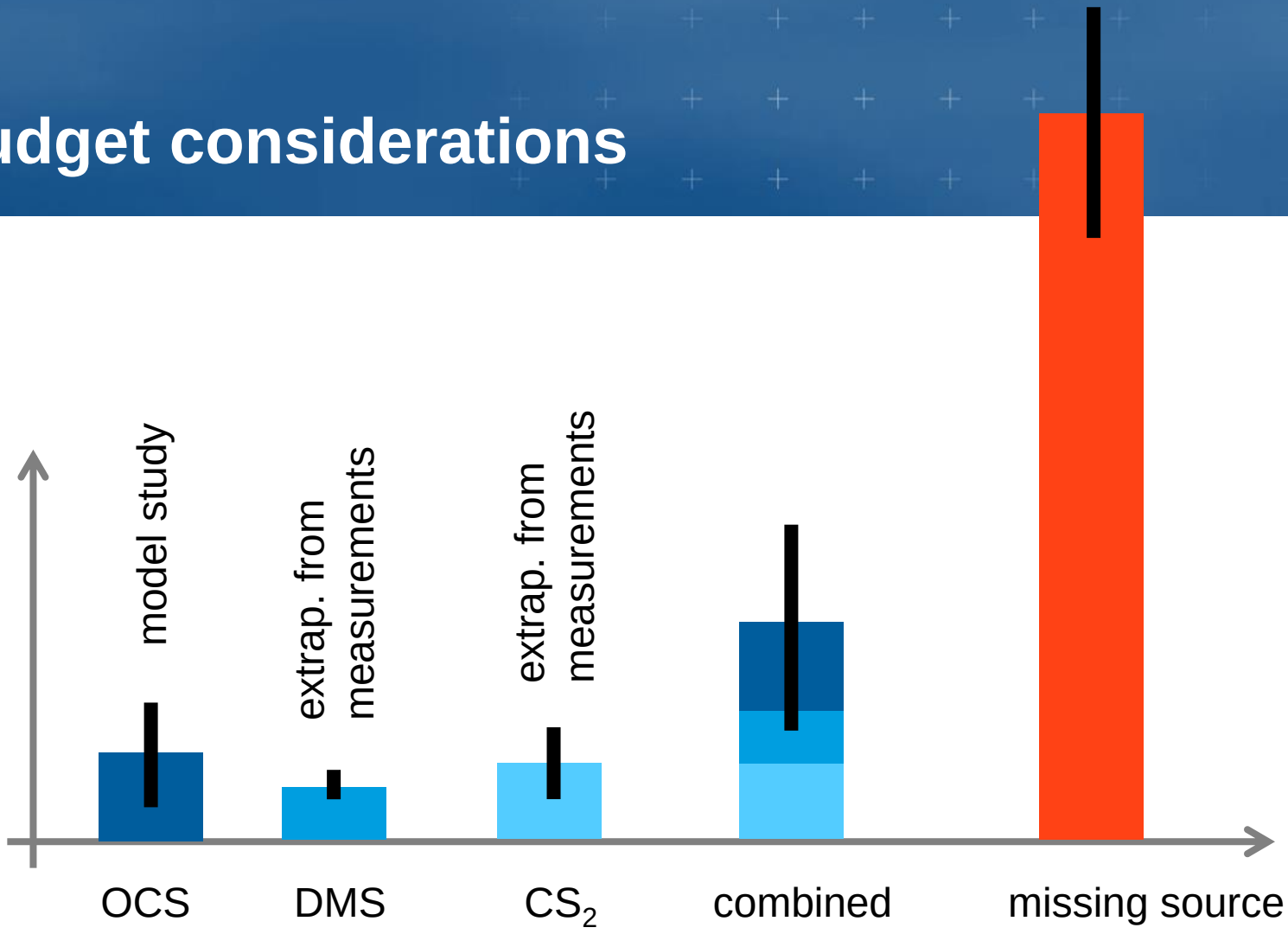


How well do we understand the ocean OCS flux?

… well enough to exclude that direct oceanic emissions account for the missing source

… indirect emissions very unlikely to account for the missing source

Budget considerations



Other possible sources:

- ~7% yield (=10fold) from DMS in tropical conditions?
- Anthropogenic sources (Lee & Brimblecombe 2015, Khan et al., 2017)?

How to assess uncertainty?

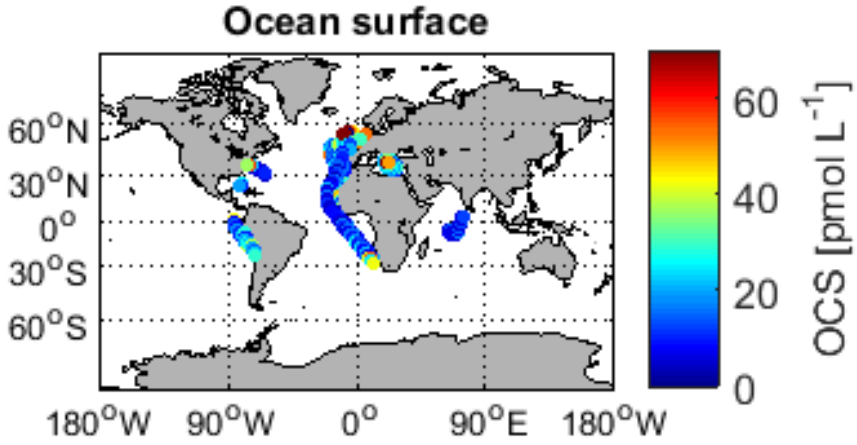
Each emission component is presented regarding:

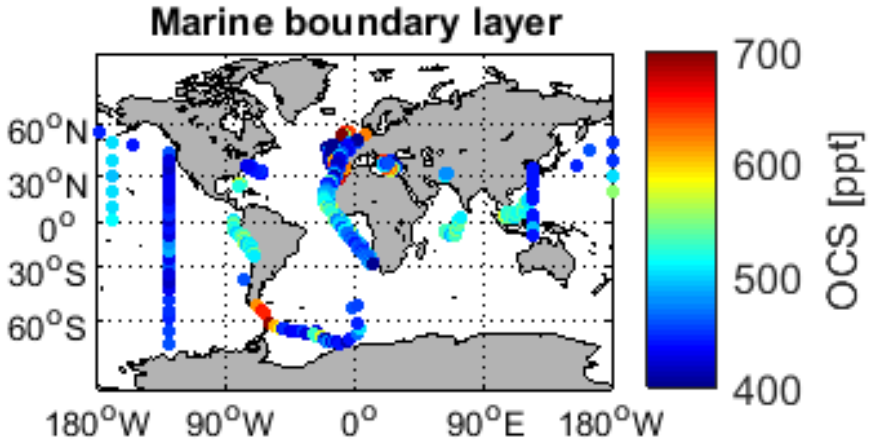
- **Number** of measurements available
- **Locations/season** covered by measurements
- **Consistency** of measurements across different methods
- **Process** understanding
- Indirect emissions: **conversion** to OCS

$\Rightarrow$ Uncertainty with respect to missing source

Direct emissions: OCS measurements

- Emissions calculated using water- and marine boundary layer concentration measurements





>20000* measurements, 17 cruises

*including 2 cruises with minute resolution

>10000 measurements, 21 cruises

Database S. Lennartz

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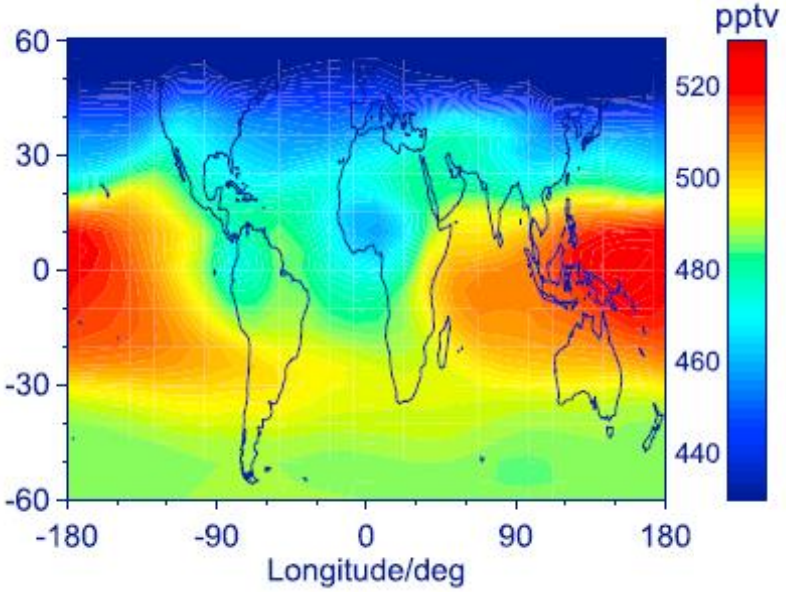
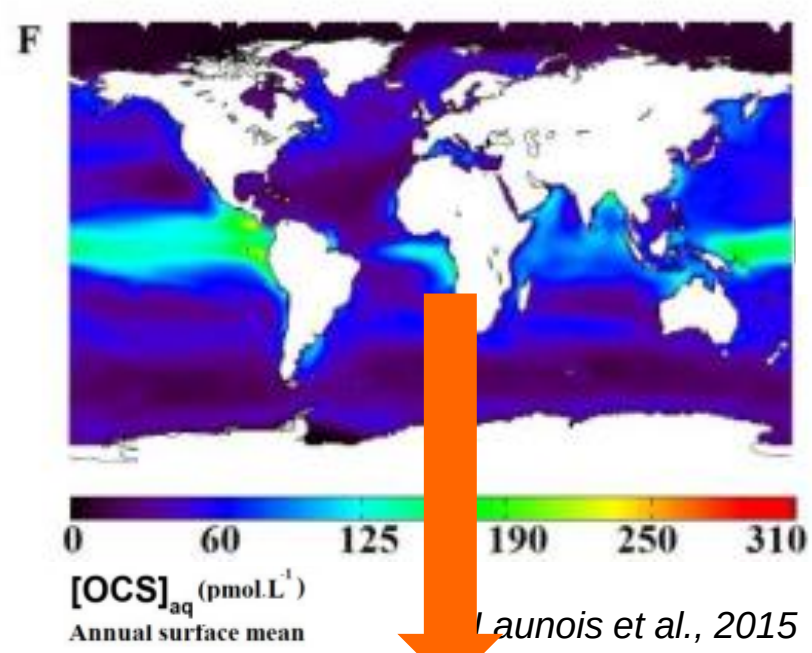
The missing source: from tropical oceans?

assumed source region



Glatthor et al., 2015

modelling approach



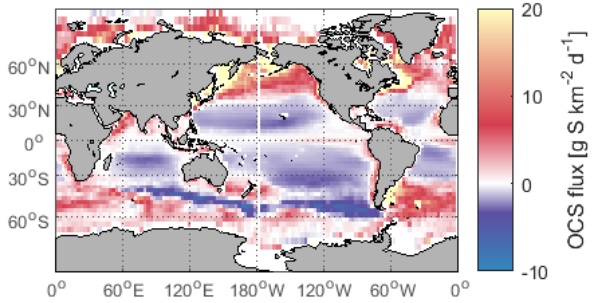
Launois et al., 2015

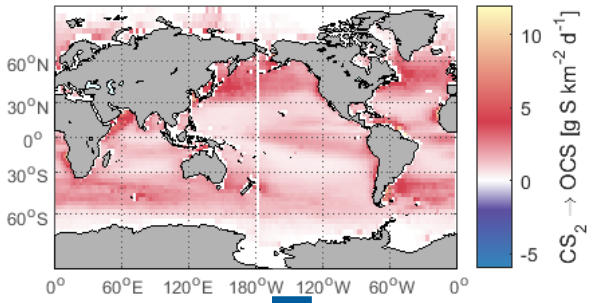
not validated:
⇒ observations from key regions are missing

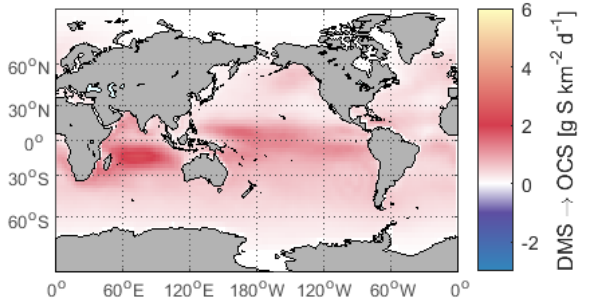
Emission climatologies

OCS + **CS_2** + **DMS**

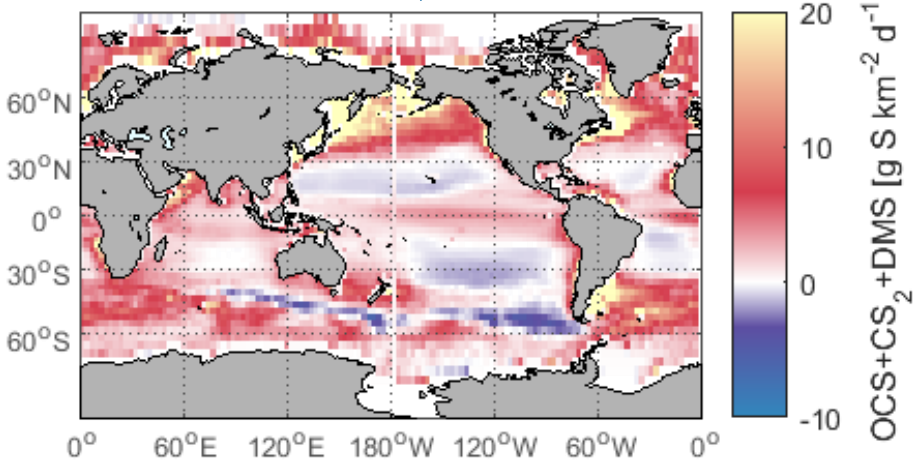
130 Gg S yr^{-1} | **180 Gg S yr^{-1}** | **80 Gg S yr^{-1}**







total



390 Gg S yr^{-1}