

Intercomparison of atmospheric Carbonyl Sulfide (TransCom-COS; Part one): Evaluating the impact of transport and emissions on tropospheric variability using ground-based and aircraft data

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Abstract

We present a comparison of atmospheric transport model simulations for carbonyl sulfide (COS), within the framework of the ongoing atmospheric tracer transport model intercomparison project “TransCom”. Seven atmospheric transport models participated in the inter-comparison experiment and provided simulations of COS mixing ratios in the troposphere over a 9-year period (2010–2018), using prescribed state-of-the-art surface fluxes for various components of the atmospheric COS budget: biospheric sink, oceanic source, sources from fire and industry. Since the biosphere is the largest sink of COS, we tested sink estimates produced by two different biosphere models. The main goals of TransCom-COS are (a) to investigate the impact of the transport uncertainty and emission distribution in simulating the spatio-temporal variability of COS mixing ratios in the troposphere, and (b) to assess the sensitivity of simulated tropospheric COS mixing ratios to the seasonal and diurnal variability of the COS biosphere fluxes. To this end, a control case with state-of-the-art seasonal fluxes of COS was constructed. Models were run with the same fluxes and without chemistry to isolate transport differences. Further, two COS flux scenarios were compared: one using a biosphere flux with a monthly time resolution and the other using a biosphere flux with a three-hourly time resolution. In addition, we investigated the sensitivity of the simulated concentrations to different biosphere fluxes and to indirect oceanic emissions through dimethylsulfide (DMS) and carbon disulfide (CS₂). The modelled COS mixing ratios were assessed against in-situ observations from surface stations and aircraft.

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30 **Key Points:**

- 31 • The models-observation mismatch suggests there is a missing source in the tropics and a
32 missing sink in the high northern latitude in summer.
- 33 • The model spread reaches 80 ppt at northern latitude sites in summer
- 34 • The diurnal rectifier effect does not exceed 30 ppt.
- 35

37 Abstract

38 We present a comparison of atmospheric transport model simulations for carbonyl sulfide (COS),
39 within the framework of the ongoing atmospheric tracer transport model intercomparison project
40 “TransCom”. Seven atmospheric transport models participated in the inter-comparison experiment
41 and provided simulations of COS mixing ratios in the troposphere over a 9-year period (2010–
42 2018), using prescribed state-of-the-art surface fluxes for various components of the atmospheric
43 COS budget: biospheric sink, oceanic source, sources from fire and industry. Since the biosphere
44 is the largest sink of COS, we tested sink estimates produced by two different biosphere models.
45 The main goals of TransCom-COS are (a) to investigate the impact of the transport uncertainty
46 and emission distribution in simulating the spatio-temporal variability of COS mixing ratios in the
47 troposphere, and (b) to assess the sensitivity of simulated tropospheric COS mixing ratios to the
48 seasonal and diurnal variability of the COS biosphere fluxes. To this end, a control case with state-
49 of-the-art seasonal fluxes of COS was constructed. Models were run with the same fluxes and
50 without chemistry to isolate transport differences. Further, two COS flux scenarios were
51 compared: one using a biosphere flux with a monthly time resolution and the other using a
52 biosphere flux with a three-hourly time resolution. In addition, we investigated the sensitivity of
53 the simulated concentrations to different biosphere fluxes and to indirect oceanic emissions
54 through dimethylsulfide (DMS) and carbon disulfide (CS₂). The modelled COS mixing ratios were
55 assessed against in-situ observations from surface stations and aircraft.

56 The results indicate that all models fail to capture the observed latitudinal distribution of COS at
57 the surface. The COS mixing ratios are underestimated by at least 50 parts per trillion (ppt) in the
58 tropics, pointing to a missing tropical source. In contrast, in summer the mixing ratios are
59 overestimated by at least 50 ppt above 40°N, pointing to a likely missing sink in the high northern
60 latitudes during the summer. The seasonal variability and the latitudinal distribution of COS
61 surface mixing ratios are more sensitive to the transport model used than to a change in biosphere
62 fluxes. Regarding the seasonal mean latitudinal profiles, in the vicinity of anthropogenic sources,
63 the spread between models is greater than 60 ppt above 40°N in boreal summer. Regarding the
64 seasonal amplitude, the model spread reaches 50 ppt at 6 out of 15 sites, compared to an observed
65 seasonal amplitude of 100 ppt. All models simulated a too late minimum by at least 2 to 3 months
66 at two high northern-latitude sites, likely owing to errors in the seasonal cycle in the ocean
67 emissions. Finally, the temporal resolution of the biosphere fluxes (monthly versus three-hourly)
68 has a relatively small impact of less than 20 ppt (compared to model spread) on the mean seasonal
69 cycle at surface stations.

70

71 1 Introduction

72 Carbonyl sulfide (COS) is a rather long-lived sulphur-containing trace gas with a mean
73 atmospheric mixing ratio less than 500 parts per trillion (ppt). Due to its long lifetime (~2.5 years),
74 COS reaches the stratosphere, where its decay products contribute to the formation of Stratospheric
75 Sulfur Aerosol (SSA). COS is emitted directly and indirectly by the ocean and industrial activities,
76 directly by biomass burning and anoxic soils (Whelan et al., 2018). The main sink of COS is the
77 uptake by the biosphere (Campbell et al., 2008; Blake et al., 2008; Suntharalingam, P. et al., 2008;
78 Berry et al., 2013a), with minor sink contributions also from chemical break-down in the
79 troposphere and stratosphere (Whelan et al., 2018). COS is taken up in leaves through similar
80 pathways as carbon dioxide, but without significant respiration (Protoschill-Krebs et al., 1996;

81 Wohlfahrt et al., 2012). For this reason, COS has been proposed as a tracer that can be used to
82 infer Gross Primary Productivity (GPP) at large scale.

83 To infer GPP from COS, we need several pieces of information that are currently still highly
84 uncertain. First and for all, the current flux estimates do not lead to a closed COS budget that is in
85 line with the near-constant COS burden in the atmosphere from 2000 up to 2015 (Whelan et al.,
86 2018). Several studies suggest that sources are missing from the tropical oceans (Berry et al.,
87 2013a; Kuai et al., 2015; Glatthor et al., 2015; Launois et al., 2015b; Remaud et al., 2022), but
88 currently no hard evidence has been obtained from shipboard measurements (Lennartz et al., 2017;
89 Lennartz et al., 2020b; Lennartz et al., 2021a). Recent inverse modelling studies (Ma et al., 2021,
90 Remaud et al., 2022) confirm the need for a tropical source of COS (or a reduced tropical sink)
91 and more COS uptake at high Northern latitudes. Interestingly, while the results of Ma et al. (2021)
92 point to too low modelled COS mixing ratios in the free troposphere, Remaud et al. (2022) could
93 not confirm this finding. This discrepancy triggers the question how well atmospheric transport
94 models are able to simulate the global COS distribution. Since the source-sink distribution of COS
95 is distinctly different from that of CO₂, a COS model comparison may lead to additional
96 information relative to earlier comparisons that were conducted within the atmospheric tracer
97 transport model intercomparison project TransCom (Law et al., 1996; Gurney et al., 2002). For
98 instance, the one-way uptake of COS by the biosphere both during day and night (Kooijmans et
99 al., 2021; Maignan et al., 2021) differs from the CO₂ interaction with the biosphere with
100 respiration at night and uptake dominating during daytime. On larger scales, the seasonal cycle of
101 COS shows strong signs of biosphere uptake in the Northern Hemisphere (NH) summer and ocean
102 emissions in the Southern Hemisphere (SH) (Montzka et al., 2007). As a result, the gradient
103 between the NH and SH changes seasonally, and in a different way than the CO₂ gradient.

104 It is however important to realise that the various terms in the COS budget are currently still very
105 uncertain. It is therefore important to study the behaviour of various surface flux terms in a variety
106 of models, to investigate whether different models point to similar inconsistencies in the global
107 COS budget. The aim of this paper (and a complementary paper, part 2) is to analyse results from
108 a model intercomparison study that focuses on COS. Specifically, we address the questions:

109 1 - What are the comparative roles of uncertainties in transport versus emission distribution in
110 simulating the interhemispheric (IH) gradient, seasonal cycle and vertical profiles of COS?

111 2- How large is the model-to-model spread compared to the mismatch between model and
112 observations (i.e. how sure are we that there is something wrong with the fluxes?)

113 3 - What is the sensitivity of simulated tropospheric COS mixing ratios to the diurnal variability
114 in COS biosphere fluxes?

115 The latter question has been addressed before in TransCom for CO₂ simulations, and is commonly
116 referred to as a (diurnal) rectifier effect (Denning et al., 1995; Denning et al., 1999). Simply said,
117 the question addresses the issue whether the use of monthly mean biosphere fluxes is sufficient to
118 reliably simulate the COS tropospheric seasonal cycle, or should the biosphere be resolved on
119 higher time resolution?

120 To answer these questions the paper is structured as follows: Section 2 describes the modelling
121 protocol, the participating models, and the measurements that were used to evaluate the models,
122 Section 3 presents the results, and Section 4 ends with a discussion and conclusions. While Part

123 one of this study focuses on model simulations with prescribed bottom-up fluxes, Part two focuses
 124 on the two sets of fluxes that were optimized using atmospheric surface observations.

125

126 **2 Participating models and outputs**

127

128 2.1 Participating models and outputs

129 Seven atmospheric transport models participated in the inter-comparison of modelled COS mixing
 130 ratios. These models represent the diversity existing in the research community. The main features
 131 of each transport model, i.e. the horizontal and vertical resolution, meteorological drivers are given
 132 in Table 1. Almost all models use meteorological fields from atmospheric reanalysis (ERA5, ERA-
 133 interim, NCEP, and JRA-55), either by direct use, or by nudging toward fields of horizontal winds
 134 (e.g LMDz; MIROC4). The TOMCAT Atmospheric Transport Model (ATM) is forced toward
 135 fields of surface pressure, vorticity, and divergence from ERA-Interim. For this model and for
 136 TM5, the convective mass fluxes are taken from ERA-Interim and interpolated to the model grid,
 137 which has a coarser resolution than ERA-Interim. In terms of resolution, NICAM6 has the highest
 138 horizontal resolution ($\sim 1^\circ$) while the TM3 ATM has the coarsest resolution ($\sim 5^\circ \times 4^\circ$).

139 The vertical mixing in the convective boundary layer is represented with different
 140 parameterizations in the different models. For deep convective mixing, parameterizations rely on
 141 the mass-flux approach and are mainly adapted from three convective schemes: the Arakawa and
 142 Schubert (1974) scheme (MIROC4, NICAM 5&6), the Tiedtke (1989) scheme (TM3) and the
 143 Emmanuel et al. (1991) scheme (LMDz). The convective mass fluxes from ERA-Interim given to
 144 the TOMCAT and TM5 ATMs is based on a modified version of the Tiedtke (1989) scheme in
 145 the European Centre for Medium-Range Weather Forecasts (ECMWF)'s Integrated Forecasting
 146 System (Bechtold et al., 2014). The Arakawa and Schubert (1974) scheme spectrally represents
 147 multiple cloud types with different cloud base mass fluxes. The Tiedtke (1989) scheme is a single
 148 plume entraining-detraining model. The Emmanuel (1991) convective scheme, implemented in
 149 LMDz, represents an ensemble of cumulus by an undilute updraft and a spectrum of mixtures with
 150 the environmental air. The subgrid scale parameterization schemes are also referenced in Table 1,
 151 although most of them have been modified from their original formulations. In most of them, the
 152 sensitivity of the convective development to environmental humidity has been enhanced either by
 153 setting up a threshold based on relative humidity to prevent deep convection from triggering too
 154 often (MIROC4, Patra et al., 2018) or by increasing the entrainment of air from the environment
 155 in the mixtures (LMDz, Grandpeix et al., 2004) or in the plume (NICAM, Chikira and Sugiyama,
 156 2010) when the environment is too dry. In LMDz, the convective triggering is now based on sub-
 157 cloud scale processes and not on the Convective Available Potential Energy (CAPE) anymore,
 158 improving the diurnal cycle of convection (Rio and Hourdin, 2008).

159

Transport model	Meteorology	Horizontal and vertical resolutions	Reference	Convection scheme	PBL mixing scheme	Advection scheme

LMDz	Nudging towards horizontal winds from ERA-5	1.875°×3.75°, 39η	Remaud et al. (2018)	Emanuel (1991); Rochetin et al. (2013)	Small scale turbulence: Mellor and Yamada (1974) Shallow convection: Rio and Hourdin. (2008)	Leer (1997); Hourdin and Armengaud (1999)
TM5	Meteo-and surface fields from ERA-Interim	2°x2°, 25η	Krol et al. (2005)	Convective mass fluxes from ERA-Interim	Near surface mixing: Louis (1979); Free troposphere mixing: Holtslag and Moeng (1991)	Slopes advection scheme: Russell and Lerner (1981)
TM3	Meteo-and surface fields from NCEP	4°x5°, 19η	Heimann et al., 2003	Tiedtke (1989)	Louis (1979)	Slopes advection scheme: Russell and Lerner (1981)
TOMCAT	Forced with the surface pressure, vorticity, divergence from ERA-Interim	2.8° × 2.8°, 60η (surface to ~60 km)	Chipperfield (2006)	Convective mass fluxes from ERA-Interim	Louis (1979)	Prather (1986)
MIROC4	Nudging towards horizontal winds and temperature from JRA-55	T42 spectral truncation (~ 2.8° × 2.8°), 67η	Patra et al. (2018)	Arakawa and Schubert (1974), with updates	Mellor and Yamada (1982)	Lin and Rood (1996)
NICAM5	Nudging towards horizontal	~223 km (icosahedral grid), 40z*	Niwa et al. (2017)	Chikira and Sugiyama (2010)	MYNN (Mellor and Yamada, 1974;	Miura (2007) & Niwa et al. (2011)

	winds from JRA-55				Nakanishi and Niino, 2004) Level 2 scheme	
NICAM6	Nudging towards horizontal winds from JRA-55	~112 km (icosahedral grid), 40z*	Niwa et al. (2017)	Chikira and Sugiyama (2010)	MYNN (Mellor and Yamada, 1974; Nakanishi and Niino, 2004) Level 2 scheme	Miura (2007) & Niwa et al. (2011)

160 Table 1: Main characteristics (vertical resolution, horizontal resolution, meteorological drivers,
 161 transport and sub-grid parameterization schemes) of the TransCom models used in this
 162 experiment. η vertical coordinates are a hybrid sigma-pressure coordinate and z^* is the terrain-
 163 following vertical coordinate based on the geometric height.
 164

165 Simulations were performed using meteorology and surface emissions for the period from 2010 to
 166 2018. The two first years are considered as spin-ups and therefore not included in the analysis. As
 167 this study focuses on the spatio-temporal COS variability and the COS budget is currently not
 168 closed (see Table 3), we do not attempt to reproduce the observed mean COS values and the
 169 simulations started from a null initial state. For simplicity, oxidation within the troposphere,
 170 estimated as 100 GgS.yr⁻¹, photolysis in the stratosphere, estimated as 35-60 GgS.yr⁻¹, have not
 171 been considered, enabling to isolate the influence of transport processes on COS tropospheric
 172 variability.
 173

174 Model output was generated at each measurement time and location used in the analysis as an
 175 hourly average. Modelers chose the horizontal positions to report simulated concentrations; either
 176 from a nearest grid point value, or interpolated to the site location from values at surrounding grid
 177 points. Additionally, 3-D fields of monthly mean COS mixing ratios were stored and analyzed.
 178 Higher temporal resolutions were not considered since this study only looks at seasonal and longer
 179 timescales, and to prevent excessively large file sizes. A more complete description of each ATM
 180 is given in Annex A.
 181

182 2.2 Prescribed flux components

183 The prescribed COS flux components used as model inputs are presented in Table 2. Each
 184 participating group interpolated the emissions horizontally in space to their (coarser) model grid,
 185 while ensuring mass conservation. Subsequently, the fluxes provided lower boundary conditions
 186 of each atmospheric transport model, which then simulates the transport of COS by the
 187 atmospheric flow. Relying on the linearity of the atmospheric transport, each flux of each scenario
 188 was transported separately by all participating models, and the various concentration contributions
 189 of the individual fluxes were then added for different scenarios (i.e. combination of fluxes) as
 190 described in section 2.4.

Process	Name	Time resolution	Spatial resolution	Period	Reference	Total global flux (GgS.yr ⁻¹)
Vegetation + soil from SIB4	BIO_SIB4	monthly, interannual	1° x 1°	2010-2018: monthly fluxes	Kooijmans et al., 2021	-669 (vegetation) -91 (soil)
Vegetation + soil from SIB4	BIO_SIB4_Diurnal	3-hourly	1° x 1°	2015: 3-hourly fluxes	Kooijmans et al., 2021	-654 (vegetation) -92 (soil)
Vegetation + soil from ORCHIDEE	BIO_ORC	monthly, interannual, 3-hourly	0.5 x 0.5°	2010-2018: monthly fluxes, 2015: 3-hourly fluxes	Maignan et al., 2021 Abadie et al., 2022	-531 (vegetation) -264 (soil)
Biomass burning	BB	monthly, interannual	1° x 1°	2010-2016	Stinecpher et al., 2019	+53
Anthropogenic	ANT	monthly, interannual	1° x 1°	2010-2015	Zumkehr et al., 2018	+397
Direct oceanic emissions + indirect emissions from CS ₂	OCE	monthly, interannual	T42 grid (ca. 2.8x2.8°)	2010-2018	Lennartz et al., 2017; Lennartz et al., 2021	+203
Indirect oceanic emission via DMS	OCE_DMS	monthly, interannual	T42 grid (ca. 2.8x2.8°)	climatological	Lennartz et al., 2017; Lana et al., 2012	+70
Indirect oceanic emissions via DMS from NEMO PISCES	OCE_DMS_PISCES	monthly, climatological	91x144 (2°x2.5°)	climatological	Belviso et al., 2012	+119

192 Table 2: Prescribed COS surface fluxes used as model input. Mean magnitudes of the fluxes are
193 given in GgS.yr⁻¹ for the period 2010–2018.

194

195 The biosphere fluxes BIO_SIB4 and BIO_ORC, simulated by the SIB4 Land Surface Model
196 (LSM) (Kooijmans et al., 2021) and the ORCHIDEE LSM (Maignan et al., 2021, Abadie et al.,
197 2022), respectively, include the COS absorption by vegetation and the oxic soil fluxes. In both
198 LSMs, the absorption by plants is parameterized following the Berry et al. (2013) model, which
199 was rescaled with varying COS surface mixing ratios and slightly adapted to represent the COS
200 absorption at night that arises from incomplete stomatal closure (Kooijmans, et al., 2021, Maignan
201 et al., 2021). The spatially varying COS mixing ratios are from a monthly climatology that was
202 obtained by transporting the optimized COS fluxes by Ma et al. (2021) with the TM5 ATM. The
203 soil fluxes include the COS irreversible uptake via hydrolysis parameterized with the Ogee et al.
204 (2016) model and an abiotic production term. In the ORCHIDEE LSM, the abiotic term is
205 parameterized following the approach described in Whelan et al. (2016) while, in the SIB4 LSM,
206 it is based on Meredith et al. (2018). The emissions from anoxic soils are not considered in this
207 study because of the absence of reliable emission estimates at the beginning of this study.

208 The direct oceanic emissions of COS (including indirect emissions from CS₂) are derived from a
209 box model approach (Lennartz et al., 2021). The indirect oceanic emissions of COS through DMS,
210 OCE_DMS_PISCES and OCE_DMS, are based on two different approaches. OCE_DMS is a
211 monthly climatology produced from extrapolations of measurements in sea waters distributed
212 unevenly around the globe (Lana et al., 2011). OCE_DMS_PISCES is simulated by a mechanistic
213 model of DMS production implemented in the Ocean General Circulation Model NEMO-PISCES
214 (Belviso et al., 2012). It should be noted that the climatology of Lana et al., 2011 has been recently
215 updated using additional sea water measurements and a refined extrapolation method (Hulswar et
216 al., 2022). The spatial distribution of the new DMS fluxes is now closer to the mechanistic
217 representation of these fluxes from Belviso et al. (2012) with larger summer emissions in the
218 southern high latitudes.

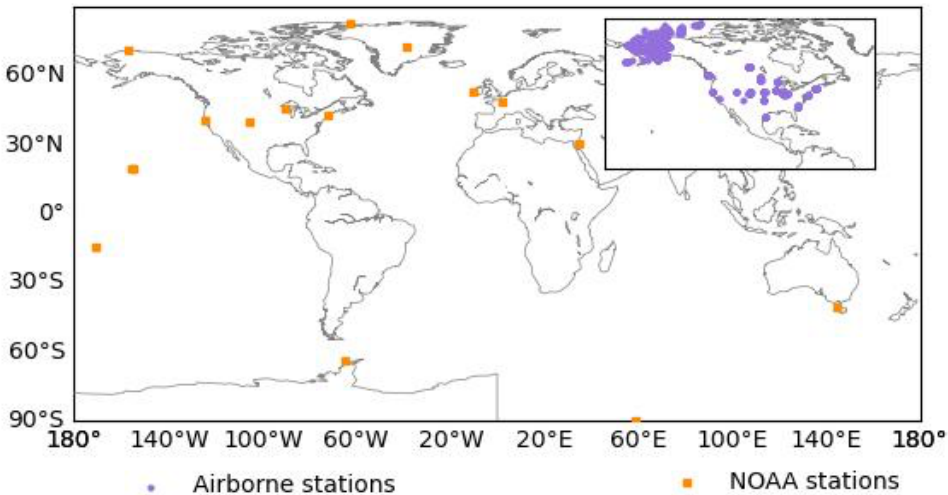
219 The open burning inventory emissions from Stinecipher et al. (2019), available for the period
220 1997–2016, include emissions from savanna and grassland, boreal forests, temperate forests,
221 tropical deforestation and degradation, peatland fires, and agricultural waste burning. The
222 inventory is obtained from CO emissions using the GFED Global Fire Emissions Database (GFED
223 version 4, <https://www.globalfiredata.org/>). Biomass burning sources from agricultural residues
224 and biofuels were not included in the absence of a global map although they were shown to be 3
225 times as large as open burning emissions over northern America (Campbell et al., 2015). In the
226 Supplement, the Stinecipher et al. (2019) inventory is compared with the GFED v4.1 and
227 Community Emissions Data System (CEDS) (Hoesly et al., 2018) (see Fig. S11), that includes
228 additional biofuel use, with a global total of 118–154 GgS.yr⁻¹ over the period 2010-2018
229 estimated in Ma et al. (2021). Choosing one inventory over the other is not expected to change the
230 findings of this study.

231 The Zumkher et al., 2018 inventory includes, in order of importance, anthropogenic emissions
232 from the rayon (staple and yarn) industry, residential coal, pigments, aluminum melting,
233 agricultural chemicals, and tyres. These emissions arise both from direct COS emissions and
234 indirect COS emissions through atmospheric oxidation of CS₂, that is supposed to be instantaneous

235 and to occur at the surface. The anthropogenic emissions are mainly located over China and
 236 Europe.

237

238 2.3 Measurements and data sampling



239

240 Figure 1. Geographical locations of the NOAA ground-based observations (orange squares) and
 241 the NOAA profile programme (inset).

242 We evaluated the simulations of COS mixing ratio against the NOAA/ESRL measurements
 243 between 2010 and 2018 at 15 sites: Cape Grim, Australia (CGO, 40.4°S, 144.6°W, 164 m above
 244 sea level, asl), American Samoa (SMO, 14.2°S, 170.6°W, 77 m asl), Mauna Loa, United States
 245 (MLO, 19.5°N, 155.6°W, 3397 m asl), Cape Kumukahi, United States (KUM, 19.5°N, 154.8°W,
 246 3 m asl), Niwot Ridge, United States (NWR, 40.0°N, 105.54°W, 3475 m asl), Wisconsin, United
 247 States (LEF, 45.9°N, 90.3°W, 868 m asl—inlet is 396 m above ground on a tall tower), Harvard
 248 Forest, United States (HFM, 42.5°N, 72.2°W, 340 10 m asl, inlet is 29 m aboveground), Barrow
 249 (named also Utqiagvik), United States (BRW, 71.3°N, 155.6°W, 8 m asl), Alert, Canada (ALT,
 250 82.5°N, 62.3°W, 195 m asl), Trinidad Head, United States (THD, 41.0°N, 124.1°W, 120 m asl),
 251 Mace Head, Ireland (MHD, 53.3°N, 9.9°W, 18 m asl), Weizmann Institute of Science at the Arava
 252 Institute, Ketura, Israel (WIS, 29.96°N, 35.06°E, 151 asl), Palmer Station, Antarctica, United
 253 States (PSA, 64.77°S, 64.05°W, 10.0 asl), South Pole, Antarctica, United States (SPO, 89.98°S,
 254 24.8°W, 2810.0 asl) and since mid-2004 at Summit, Greenland (SUM, 72.6°N, 38.4°W, 3200 m
 255 asl). The COS samples have been collected as paired flasks one to five times a month since 2000
 256 and have been analysed with gas chromatography and mass spectrometry detection. Most
 257 measurements have been performed in the afternoon between 11 and 17h local time when the
 258 boundary layer is well mixed, thereby optimizing the comparability with model simulations. Only
 259 COS measurements with a difference between the paired flasks of less than 6.3 ppt are considered.
 260 These data are an extension of the measurements first published in Montzka et al., 2007. In
 261 addition, we used the French sampling site, GIF (48.7°N - 2.1°E), located about 20 km to the
 262 south west of Paris where ground level COS has been monitored on an hourly basis since August
 263 2014 (Belviso et al., 2022).

264

265 To investigate the impact of transport errors on the vertical distribution of COS, we compared
 266 model results to 2012–2016 NOAA airborne data located at 11 sites over North America. The
 267 upper altitude that was typically reached by this sampling program is 8 km.

268

269 2.4 Emission scenarios: the different experiments

270 The bottom-up emission scenarios with their associated source and sink components of COS
 271 considered in this study are described in Table 3. The control (**Ctl**) scenario represents the state of
 272 the art in the COS global budget, as it combines the main known COS fluxes. Only volcano
 273 emissions, in the range 23–43 GgS.yr⁻¹, and emissions from anoxic soils, have not been considered
 274 (Whelan et al., 2018). Compared to previous studies (see Table 1 of Remaud et al., 2022), the
 275 budget for **Ctl** is almost closed with an imbalance of only -37 GgS.yr⁻¹ and leads to nearly stable
 276 atmospheric mixing ratios at surface sites (see Fig S1). However, we didn't take into account the
 277 chemical removal terms in this study: the photolysis loss of COS in the stratosphere amounting to
 278 around 50 GgS.yr⁻¹ and the oxidation loss of COS in the troposphere amounting to around 100
 279 GgS.yr⁻¹ (Whelan et al., 2016). If the chemical removal terms were included, the budget would
 280 be negatively unbalanced by 200 GgS.yr⁻¹, which deviates from the -37 GgS.yr⁻¹.

281 The **Diurnal** scenario differs from the **Ctl** scenario in that it uses biosphere fluxes (soil and
 282 vegetation) with a 3-hourly temporal resolution instead of a monthly resolution. Comparing
 283 scenarios **Ctl** and **Diurnal** addresses research question 3.

284 The two last emission scenarios, **Bio2** and **Ocean2**, aim to investigate the influence of a change in
 285 terrestrial and oceanic fluxes on atmospheric surface mixing ratios. The **Bio2** scenario differs from
 286 the **Ctl** scenario in that the biosphere fluxes are provided by the ORCHIDEE LSM instead of the
 287 SIB4 LSM. The **Ocean2** scenario differs from the **Ctl** scenario in that the DMS oceanic fluxes are
 288 provided by the NEMO-PISCES ocean model instead of the climatology of Lennartz et al. (2017).
 289

Name	Transported fluxes	Source-Sink Balance	ATMs
Ctl	ANT+BB+OCE+OCE_DMS+BIO_SIB4 (monthly)	-37 GgS.yr ⁻¹	All (see Table 1)
Diurnal	ANT+BB+OCE+OCE_DMS+BIO_SIB4_Diurnal	12 GgS.yr ⁻¹	LMDz, TM5, TM3, MIROC4, NICAM5, NICAM6
Bio 2	ANT+BB+OCE+OCE_DMS+BIO_ORC	-72 GgS.yr ⁻¹	LMDz
Ocean 2	ANT+BB+OCE+OCE_DMS_PISCES+BIO_ORC	11.7 GgS.yr ⁻¹	LMDz

290 Table 3: Description of the emission scenarios. Note that the budget does not include chemical
 291 removal terms of ~150 GgS.yr⁻¹ (Whelan et al., 2018).

292

293 2.5 Post-processing of the simulations and measurements

294 In Sect. 3, the features of interest (annual mean, monthly smoothed seasonal cycle) are derived
 295 from the surface mixing ratios using the CCGVU curve fitting procedure developed by Thoning

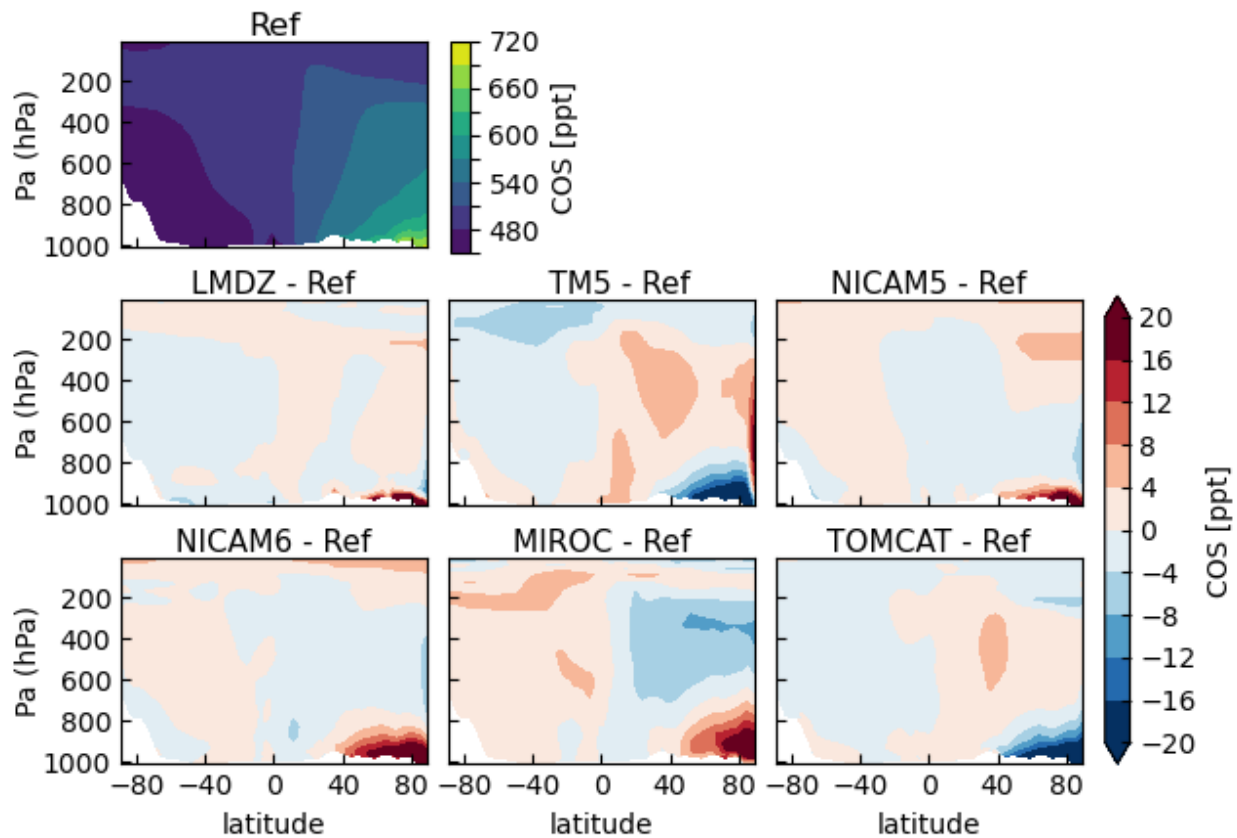
296 et al. (1989) (Carbon Cycle Group Earth System Research Laboratory (CCG/ESRL), NOAA,
 297 USA). The CCGVU procedure is fully described and freely available at
 298 <http://www.esrl.noaa.gov/gmd/ccgg/mbl/crvfit/crvfit.html>. The procedure estimates a smooth
 299 function by fitting the time series to a first-order polynomial equation for the growth rate combined
 300 with a two-harmonic function for the annual cycle and with the residuals that are filtered with a
 301 low-pass filter using 80 and 667 days as short-term and long-term cutoff values, respectively. The
 302 seasonal cycle and annual gradient are extracted from the smooth function. In addition, outliers
 303 are discarded if their values exceed 3 times the standard deviation of the residual time series.

304 3 Results

305 3.1 Impact of different transport models: using one flux scenario

306

307 3.1.1. General behavior: zonal mean structure



308
 309 Figure 2. Zonal mean mole fraction of COS (ppt) for the reference for the **Ctl** scenario (top row).
 310 The reference is the average of COS over all transport models, calculated for the summer months
 311 (June, July, August) in 2012–2018. The resulting COS abundances have been shifted by +396
 312 ppt, which brings the reference close to the observed concentrations averaged over all surface
 313 sites for January averaged over the years 2012-2018. Second and third rows: Zonal mean mole
 314 fraction difference between each individual transport model and the reference.
 315

316 We first study the zonal mean structure of the COS simulations. We focus on the boreal summer
317 (June-July-August - JJA) as convection is more active over the continents in the NH, which
318 causes the spread among the models to be the largest. Moreover, the use of COS as a
319 photosynthesis tracer requires quantifying the transport errors during boreal summer, when
320 photosynthesis is more active.

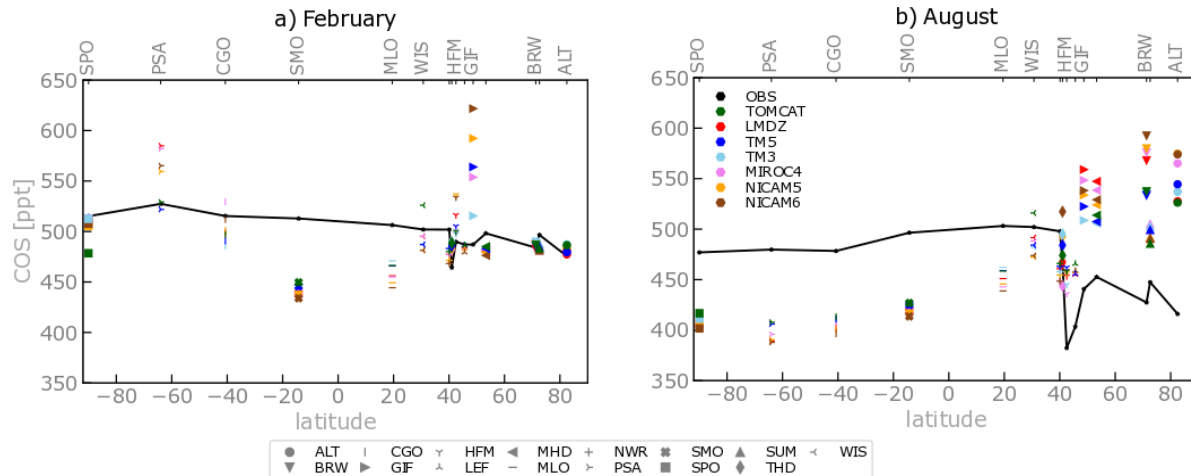
321 With the **Ctl** scenario, Figure 2 shows that the zonal mean distribution of COS averaged over the
322 transport models exhibits a strong meridional gradient in boreal summer with higher COS values
323 in the NH. The COS source outweighs the terrestrial sink, leading to a net accumulation of COS
324 mole fractions at the surface during winter. The reader is referred to Section 3.2 for a
325 decomposition of the COS total surface mole fractions into the signals caused by the different COS
326 budget components. In addition, Figure 2 depicts the zonal mean distribution of the difference of
327 COS mole fractions between each model and the multi-model average. The effect of the transport
328 differences is the largest below 800 hPa and exceeds 50 ppt above 40°N, where surface fluxes are
329 the largest. The spread of COS mole fractions at the surface reflects different strengths of vertical
330 mixing within the tropospheric column. Indeed, a positive anomaly of surface COS mole fractions
331 at the surface compared to the multi-model average is often associated with a negative anomaly in
332 the mid-troposphere. In particular, higher surface mole fractions of COS in the NICAM, MIROC4
333 and LMDz ATMs suggests that there is, on average, less convection penetrating into the upper
334 troposphere in these models compared to the TM5 and TOMCAT ATMs. The comparison between
335 the NICAM5 and NICAM6 ATMs indicates a modest contribution of the model horizontal
336 resolution to the model spread, as observed by Lin et al. (2018). This is also in agreement with
337 Remaud et al. (2018) who showed that the convective and the planetary boundary layer
338 parameterization schemes have larger impact on the CO₂ mole fractions in the low and mid
339 troposphere relative to the impact of horizontal and vertical resolutions.

340 In the three models exhibiting less vertical mixing, two of them, the TOMCAT and TM5 ATMs
341 use the convective masses fluxes from the ERA-Interim reanalysis extrapolated to their lower
342 resolution model grid. The TM3 ATM is based on the Tiedtke et al. (1989) which has been
343 recognized to trigger convection too often (Hirons et al., 2013). In the models exhibiting less
344 vertical mixing, the original formulation of the convective schemes has been modified to depart
345 from the convective quasi-equilibrium assumption proposed by Arakawa and Schubert (1974) and
346 to prevent deep convective clouds from developing too often, especially in a too dry environment.
347 In the LMDz ATM, the original closure based on the CAPE of the Emanuel (1991) scheme was
348 replaced by a closure based on sub-cloud processes that enables deep convection to be delayed
349 later in the afternoon and reduced in intensity (Rio and Hourdin, 2008). The entrainment function
350 in the mixtures has also been modified to be more sensitive to relative humidity of the environment
351 (Grandpeix et al., 2004). In the MIROC4 ATM, a threshold as a function of relative humidity has
352 been implemented in the Arakawa and Schubert (1974) scheme to prevent convection from
353 triggering when the relative humidity is too low. In the NICAM ATM, the Chikira and [Sugiyama](#)
354 (2010) scheme models the entrainment rate to vary vertically, depending on the humidity and
355 temperature profiles. These implementations generally lead to a more realistic tropical variability
356 (Lin et al., 2006) and could explain why the vertical mixing is weaker in MIROC 4, LMDz and
357 NICAM.

358 It should be noted that, in consistency with previous studies (Patra et al., 2011a, Saito et al., 2013),
359 the meridional gradient of COS reflects the intensity of the inter-hemispheric exchanges and seems
360 to be controlled by the vertical gradient in the northern hemisphere. Indeed, in the middle

361 troposphere, a negative anomaly of COS mixing ratio in the northern hemisphere is combined with
 362 a positive anomaly of COS mixing ratio in the southern hemisphere in most models exhibiting less
 363 vertical mixing (MIROC, NICAM 5 and 6). On the contrary, in models exhibiting stronger vertical
 364 mixing, a positive anomaly of COS in the northern hemisphere is associated with a negative
 365 anomaly of COS in the southern hemisphere.

366
 367 3.1.2. Latitudinal gradient at surface stations



368
 369 Figure 3: Comparison of the latitudinal variations of the COS abundance simulated by several
 370 transport models using the **Ctl** surface flux dataset (colored dots) with the observations (black line)
 371 for February (left), August (right) over the years 2012-2018. The simulated COS abundances have
 372 been shifted such that the means are the same as the mean of the observations (~500 ppt). The time
 373 series of COS mixing ratio have been detrended and filtered to remove the synoptic variability
 374 beforehand. In August, the value at site GIF simulated by the TOMCAT ATM was removed as it
 375 was an outlier (value above 800 ppt). For the same reason, the COS values at site GIF simulated
 376 by TOMCAT (800 ppt) and LMDz (around 700 ppt) have been removed in February. We removed
 377 the site KUM, which is co-located in longitude and latitude with site MLO, for the sake of
 378 simplicity.

379
 380 The latitudinal gradient of COS mole fractions reflects the latitudinal surface flux distribution and
 381 the intensity of the interhemispheric exchange (Denning et al., 1999). Figure 3 shows the observed
 382 and simulated mixing ratios at the surface stations as a function of the latitude in February and
 383 August. The simulated COS mixing ratios are averaged over time at each surface station. Since
 384 the simulations start from a null initial state, the simulated COS mole fractions have been shifted
 385 by 500 ppt to match the annual mean COS observations. In February, the distribution of the
 386 observed surface mixing ratios is relatively flat over all latitudes. In contrast, all models exhibit a
 387 COS mole fraction which is 50 ppt lower in the tropics than elsewhere. This suggests that all the
 388 models agree on a missing source or a too strong biosphere sink over the tropics. Given the oceanic
 389 footprint of the tropical sites MLO and SMO, previous top-down studies of Remaud et al. (2022),
 390 Ma et al. (2021), and Berry et al. (2013) increased the oceanic source over the tropics to decrease
 391 the model-observation mismatch. The ATMs are unable to represent the negative gradient of 15
 392 ppt from MHD to GIF (see also Fig. 11 of Remaud et al., 2022) and instead overestimate the
 393 mixing ratio at site GIF by up to 300 ppt. GIF is located in the vicinity of a misplaced hotspot of
 394 anthropogenic emissions in the Zumkher inventory (Remaud et al., 2022; Belviso et al., 2020)

395 while in reality, GIF is comparable to a background station relatively far from major anthropogenic
 396 sources (Belviso et al., 2020). Overall, the model spread does not exceed 50 ppt at all sites except
 397 at sites PSA and GIF. The spread at PSA arises from a combination of strong oceanic emissions
 398 in austral summer and variation in vertical mixing. The spread at GIF is caused both by the ATM
 399 resolutions and the sub-grid scale parameterizations. Indeed, the ATMs with the highest vertical
 400 resolution, TOMCAT, NICAM 6 simulate mole fractions up to 300 ppt higher than the ATM with
 401 the lowest resolution, the TM3 ATM. It is well known that, as the model resolution increases, the
 402 simulated mixing ratios become more sensitive to the detailed distribution of sources that are
 403 defined with finer resolution. Likewise, the sensitivity to model errors is enhanced near emission
 404 hotspots. Errors in horizontal winds or errors in the vertical mixing can have a large impact with
 405 emissions from hotspots being nearby atmospheric stations, possibly creating biases. For instance,
 406 errors in horizontal winds can produce peaks which are not present in the observations (Locatelli
 407 et al., 2015a). Therefore, extra care should be taken when assimilating stations like GIF to optimize
 408 the COS surface fluxes in an atmospheric inverse framework (Remaud et al., 2022).

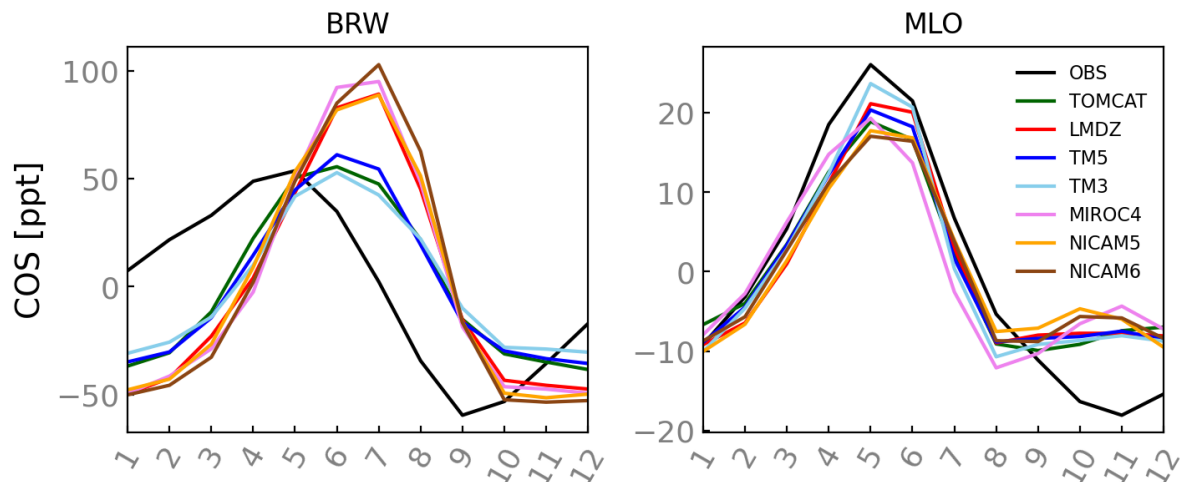
409

410 In August, the observed latitudinal distribution of COS was poorly captured by the ATMs. The
 411 observations exhibit a negative latitudinal gradient of almost 100 ppt between ALT and SPO. The
 412 lowest values of COS are located in the mid and high northern latitudes where the biosphere
 413 absorbs a substantial amount of COS from the atmosphere (Vesala et al., 2022, Maignan et al.,
 414 2021, Koojmans et al., 2021). In contrast, all ATMs simulate a positive interhemispheric gradient
 415 of 150 ppt between the northern and southern mid latitudes, with the highest values in the northern
 416 high latitudes. Overall, the deviation among models is much broader in August than in February,
 417 with a model spread exceeding 70 ppt in the northern high latitudes, for instance at BRW. This is
 418 due to the different intensities of the vertical mixing within the column (see Figure 1). The model
 419 spread does not exceed 15 ppt elsewhere but remains larger, by comparison, to the measurement
 420 uncertainty of 6 ppt.

421

422 3.1.3. Mean seasonal cycle at surface stations

423



424

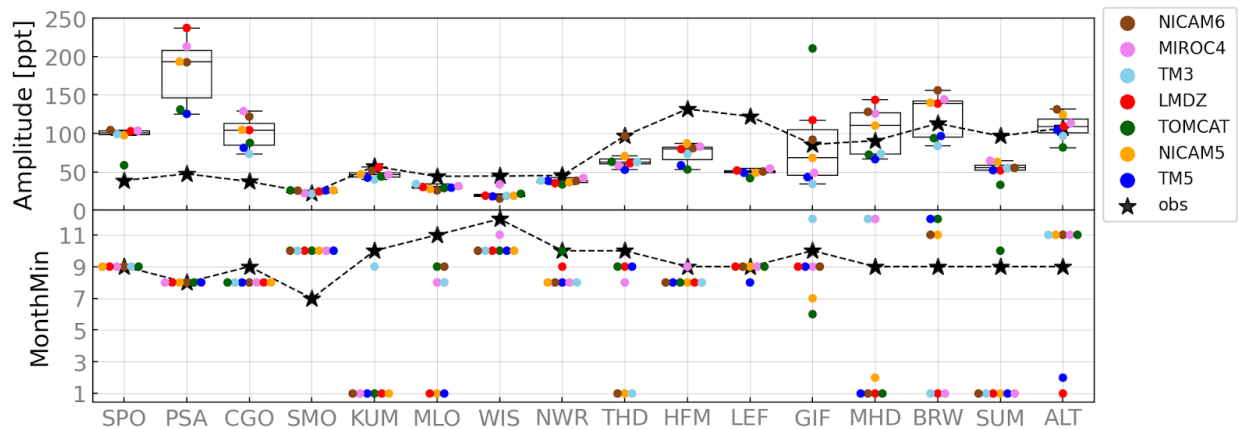
425

426 Figure 4 : Mean seasonal cycle of the observed (black) and simulated (color) COS mixing ratios
 427 at sites BRW and MLO. The curves have been detrended and filtered to remove the synoptic
 428 variability.

429

430 The impact of transport variability on the seasonal cycle is illustrated in Figure 4, which shows the
 431 mean seasonal cycle of COS given by all ATMs at sites BRW and MLO. BRW is a boreal station
 432 that samples mainly continental air masses coming from the mid and high latitudes (Parazoo et al.,
 433 2011). MLO is a background station with a strong maritime influence that samples air masses
 434 coming from the whole NH. Therefore, the seasonal amplitude at site BRW is twice as large as the
 435 seasonal cycle amplitude at site MLO. At site BRW, the simulated seasonal cycles lag that of the
 436 observations by 1 to 2 months in all transport models. In the observations, the mole fractions peak
 437 in May, whereas the modeled mole fractions peak in July. Focusing on the model spread, two
 438 groups of models can be distinguished: models with a large seasonal amplitude of 150 ppt and a
 439 weak vertical mixing (LMDz, NICAM5 & 6, MIROC4) and models with a small seasonal
 440 amplitude of 90 ppt and a strong vertical mixing (TOMCAT, TM3, TM5). It should be noted that
 441 the models with a large seasonal cycle amplitude have a steeper latitudinal gradient in August, as
 442 explained by Denning (1995). Compared to the site BRW, the models capture the phase of the
 443 seasonal cycle at site MLO relatively well, and their seasonal amplitudes diverge by not more than
 444 20 ppt. However, all models underestimate the seasonal amplitude by 20 ppt and do not represent
 445 the observed minimum in November.

446



447

448 Figure 5: Top: Mean seasonal amplitude (maximum minus minimum mole fraction) of the
 449 observed (black stars) and simulated (colored dots) COS mole fraction at 15 surface sites. Each
 450 color dot corresponds to the mean seasonal amplitude of COS mixing ratio simulated by a different
 451 atmospheric transport model for the Ctl scenario. Boxplots of the mean seasonal amplitude of
 452 simulated COS mole fractions are superimposed. Bottom: Month of the minimum of the mean
 453 seasonal cycle for the observations (black) and for the several transport models (color dots). For
 454 each site, the COS time series have been detrended and filtered to remove the synoptic variability.

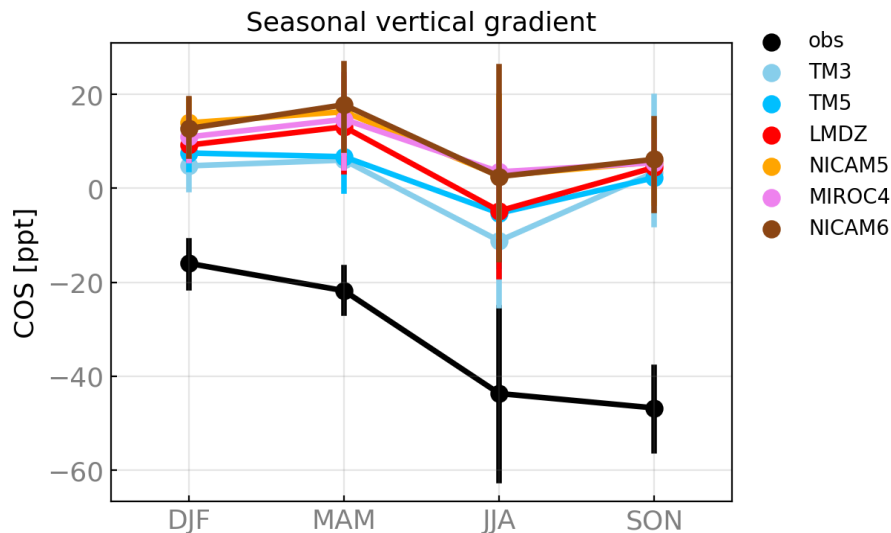
455

456 The performance of the transport models for the seasonal cycle amplitude is statistically evaluated
 457 for each surface station in the top panel of Figure 5. The complete mean seasonal cycle for each
 458 station and each model is shown on Figure S4 in the Supplement. The models capture the seasonal
 459 cycle amplitude well at the low latitude sites SMO, KUM, MLO, WIS, and at site NWR. These
 460 sites, representative of background air masses, exhibit a small seasonal amplitude of less than
 461 50 ppt. At the most southern sites (SPO, PSA, CGO), the models overestimate the seasonal amplitude
 462 by at least 50 ppt. Since these stations sample air masses mainly coming from the Southern Ocean

463 (Remaud et al., 2022, Montzka 2007), an overestimated amplitude is likely caused by too strong
 464 oceanic emissions in summer. In contrast, the seasonal amplitude is underestimated by 50 ppt at
 465 continental sites THD, HFM, and LEF. Since the seasonal amplitude to a great extent reflects the
 466 amount of COS absorbed by plants at these sites (Campbell 2008, Blake et al., 2008), a too small
 467 simulated seasonal cycle amplitude likely arises from a too weak photosynthesis sink during the
 468 growing season. Focusing on the transport errors, the spread is greater than 50 ppt at site PSA,
 469 located in the Southern Ocean, and sites MHD, GIF, BRW, ALT. As illustrated in Figure 4,
 470 differences in the strength of the vertical mixing within the column mainly contribute to the model
 471 spread. Only at station GIF the resolution is also crucial. It should be noted that the mean mole
 472 fractions showed the largest model spread also at these stations. To evaluate the simulated seasonal
 473 cycle phase, the bottom panel of Figure 5 focuses on the month of the minimum concentration of
 474 the mean seasonal cycle for each site. A striking feature is that, at mid and high latitudes sites
 475 MHD, SUM, BRW, ALT, the seasonal minimum occurs in September in the observations. In the
 476 models, this minimum occurs up to 6 months later between October and January, as illustrated in
 477 Figure 4. At sites LEF, NWR, THD, HFM over Northern America, the models tend to simulate an
 478 earlier minimum crossing of at least one month. This might be related to the too weak terrestrial
 479 sink.
 480

481 3.1.3. Mid-troposphere seasonal variations over Northern America

482



483 Figure 6. Seasonal mean observed and simulated COS gradient between 1 and 4 km (mole fractions
 484 at 1 km minus mole fractions at 4 km) averaged over airborne stations located over northern
 485 America for the **Ctl** scenario. For each subregion, the monthly COS gradients are calculated by
 486 averaging the differences in COS concentrations between 1 and 4 km over all the vertical profiles.
 487 For each season, the error bar represents the standard deviation of the seasonal COS gradient.
 488

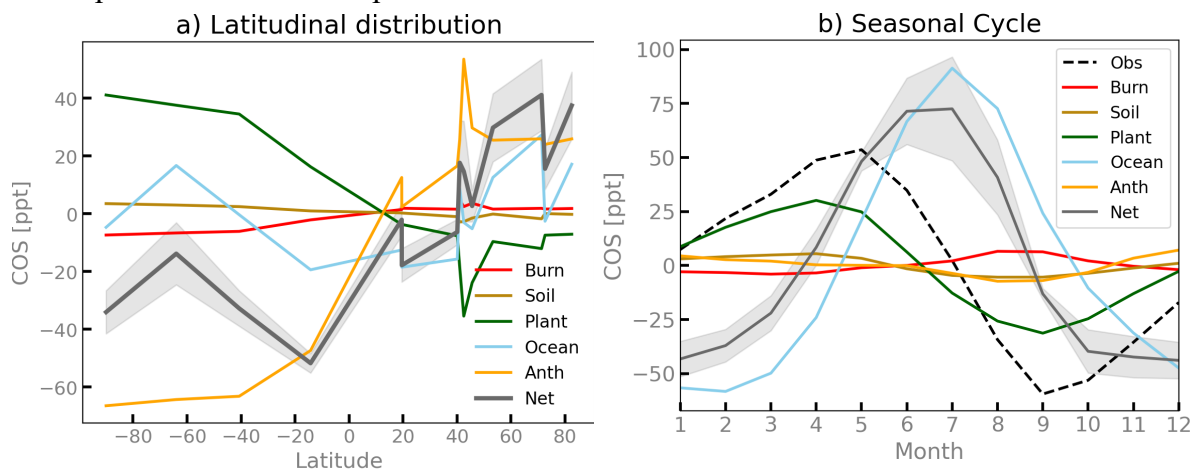
489

490 The vertical gradient between the boundary layer and the free troposphere reflects the effects of
 491 the surface fluxes and the atmospheric transport. Figure 6 shows the seasonal cycle of the vertical
 492 gradient of COS between altitudes of 1 km and 4 km averaged over the airborne sampling over
 493 Northern America (see Fig. 1). Since westerly winds prevail throughout the year in the entire free-

494 troposphere at each site (Sweeney et al., 2015), oceanic air masses from the Pacific Ocean move
 495 across the North American continent and mix with air that has been in contact with the biosphere
 496 and anthropogenic emissions. Thus, these sites sample both continental and oceanic air masses,
 497 with the proportion of oceanic air decreasing from the West to the East of America. The
 498 observations show a negative mean vertical gradient throughout the year, decreasing from -20 ppt
 499 in DJF (December, January, February) to -50 ppt in SON (September to November). This suggests
 500 that, on average, continental Northern America behaves as a COS sink. The strongest decrease of
 501 20 ppt occurs during the growing season in JJA. The large depletion of COS within the boundary
 502 layer seen in airborne profiles over Northern America has been reported previously to be
 503 concomitant to depletion of CO₂, indicating a strong and common biosphere sink during this
 504 season (Blake et al., 2008, Campbell et al., 2008, Parazoo et al., 2021).

505 Contrary to the observations, all ATMs show a mean positive vertical gradient all year round,
 506 except during JJA when gradients simulated by LMDz, TM3, and TM5 become slightly negative.
 507 The model spread is less than 10 ppt and reaches 15 ppt in JJA, much smaller than the mean bias
 508 of at least 30 ppt. The model spread and the observed and simulated standard deviation are higher
 509 in JJA. In summer, the weakening of the winds over the middle of the continent and over the east
 510 coast leads to less homogeneous vertical profiles in the free troposphere (Sweeney et al., 2015).
 511 Combined with enhanced convection, this effect might reinforce the model spread and the
 512 simulated standard deviation. Considering the model spread, the models underestimate the sharp
 513 decrease of vertical gradient in JJA by 50 % and do not prolong this decrease in SON. This model-
 514 observation mismatch is consistent with an underestimation of the mean seasonal cycle amplitude
 515 at sites HFM, LEF, THD. Kooijmans et al. (2021) showed that, on average, the SIB4 LSM using
 516 the Berry et al. (2013a) model for the plant uptake, combined to the Ogee et al. (2016) soil model
 517 with variable COS mole fractions, underestimates the COS terrestrial sink (soil and plant uptake)
 518 during the growing season over FLUXNET sites located in Europe and Northern America. Parazoo
 519 et al. (2021) came to the same conclusion by evaluating the SIB4 plant uptake against airborne
 520 measurements over three diverse regions in North America: the crop-dominated Midwest,
 521 evergreen-dominated South, and deciduous broadleaf-dominated Northeast. Photosynthesis was
 522 shown to peak later in the season over the humid temperate forest in the South compared to the
 523 SIB4 LSM (Parazoo et al., 2021), which is consistent here with an underestimated COS depletion
 524 in SON in Figure 5.

525 526 3.2 Impact of each flux components on COS surface mole fractions



528 Figure 7: a) Simulations of the interhemispheric gradient of tropospheric COS mole fractions at
529 NOAA surface stations. The net signal (gray line) is obtained from a multi-model average of global
530 simulations using the **Ctl** emissions, while the colored lines are obtained by running the global
531 atmospheric simulations with one component flux at a time. The shaded area represents the model
532 spread. b) Simulations of the seasonal cycle of tropospheric COS mixing ratios at the Barrow
533 Atmospheric Baseline Observatory (BRW) averaged over all transport models. The shaded area is
534 the standard deviation around the mean COS seasonal cycle associated with the different transport
535 models. The dotted black line represents the observed seasonal cycle.

536

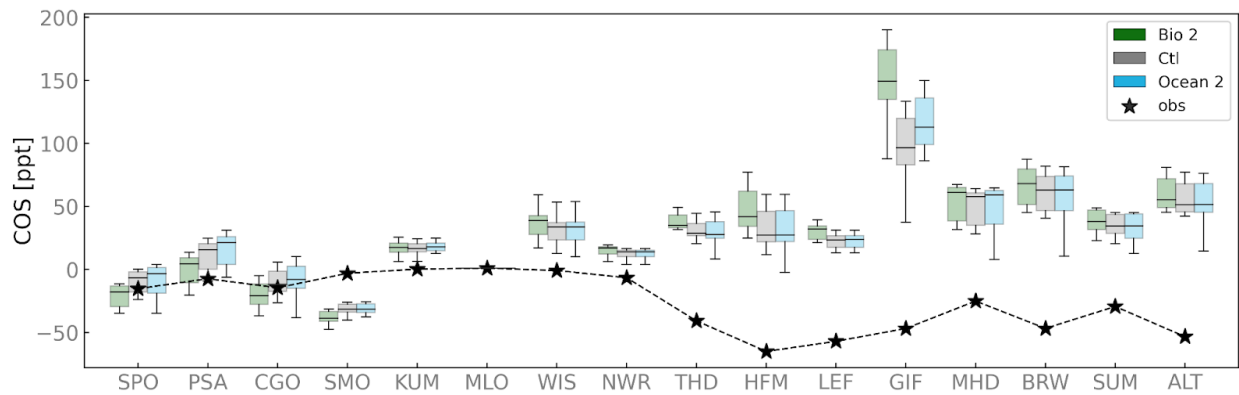
537 In order to interpret the model-observation comparison of figure 3, Figure 7a presents the
538 contributions of the COS budget components to the simulated interhemispheric gradient. Results
539 represent the average over all ATMs participating in this intercomparison experiment. The strongly
540 positive latitudinal gradient is driven by the anthropogenic component and to a lesser extent by the
541 ocean emissions. The oceanic component is characterized by two mole fraction peaks in the
542 Southern and Northern high latitudes and a minimum mole fraction in the tropics. The positive
543 mole fractions at high latitudes result from the direct oceanic emissions in summer (Lennartz et
544 al., 2017), the indirect emissions through DMS and CS₂ peaking in the tropics (Lennartz et al.,
545 2021, see also Figure 2. of Remaud et al., 2022). On an annual basis, the plant uptake is
546 characterized by a larger sink in the NH than in the SH. The resulting latitudinal gradient is
547 however not sufficient to compensate for the opposing gradients from the ocean and anthropogenic
548 emissions, leading to the overall mismatch observed in figure 3. The soil and the biomass burning
549 components have a relatively flat distribution and therefore play a minor role in the latitudinal
550 COS gradients.

551 Figure 7b shows the contributions of the COS budget components - oxic soils, ocean, plant uptake,
552 anthropogenic emissions, biomass burning - to the detrended mean seasonal cycle at site BRW.
553 The seasonality given by all ATMs is governed by the oceanic and plant uptake components. Since
554 the anthropogenic fluxes do not vary throughout the year, the anthropogenic component of the
555 simulated COS net concentrations is constant throughout the year, as expected. The weak
556 seasonality of the soil component arises from the COS soil emissions in warmer conditions in
557 summer that offsets the soil uptake in the Ogee et al. (2016) model that is implemented in the SIB4
558 model (See Fig 3. from Kooijmans et al., 2021). The one to two months lag between the observed
559 and simulated concentrations at BRW (see figure 4) is thus likely induced by too strong oceanic
560 direct emissions at high latitudes in summer or/and an underestimated plant sink in the boreal
561 ecosystems of the NH. An enhanced plant uptake or/and reduced oceanic emissions in the summer
562 high latitudes will also decrease the model observation-mismatch for the inter-hemispheric
563 gradient (Figure 3). Using an atmospheric inverse framework, Remaud et al. (2022) found that an
564 enhanced COS sink over the boreal regions associated with reduced oceanic summer emission in
565 the Atlantic enables the simulated COS mole fractions to be in better agreement with the airborne
566 measurements from the HIPPO campaign over the Pacific. From a bottom-up modeling
567 perspective, there are some indications that the direct oceanic COS emissions could be
568 overestimated and that the plant uptake is too low in boreal latitudes. For instance, the COS mole
569 fractions given by the ocean box model are higher than most of the measurements made in sea
570 waters sampled over different parts of the globe (Lennartz et al., 2017). In addition, Vesala et al.
571 (2022) showed that the biosphere sink in LSMs was too small at a forested boreal site, Hyytiälä,
572 in Finland. Scaled to all evergreen needleleaf forests over the whole boreal region, their empirical

573 model calibrated on observations at Hyytiälä produces a biosphere COS sink that is consistent with
 574 the missing COS sink identified by our analysis.
 575

576 3.3 Impact of different flux scenarios on COS surface concentrations: using the mean across
 577 transport models

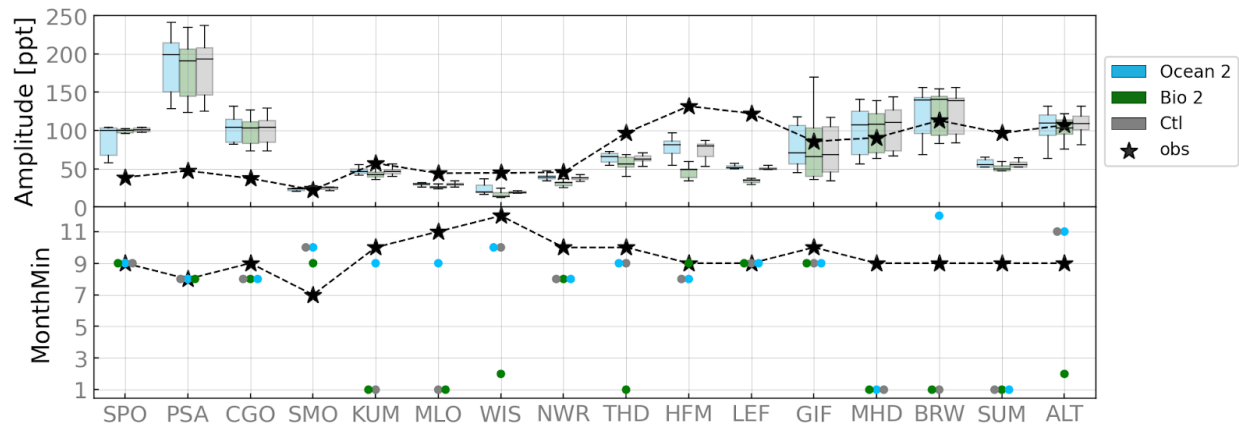
578 3.3.1. Changing model fluxes



579
 580

581 Figure 8: Box-plots of the simulated mole fraction gradient of COS between MLO and the other
 582 surface stations for the **Ctl** (gray), **Bio2** (green), **Ocean2** (blue) scenarios over the years 2012-
 583 2018 in August. The black stars denote the observed mean COS gradient between MLO and the
 584 other surface sites. The site codes are listed on the abscissa. For each site, the COS time series
 585 have been detrended and filtered to remove the synoptic variability.

586



587

588 Figure 9: Top: Box-plots of the peak-to-peak amplitude (maximum minus minimum mole fraction)
 589 of the mean COS seasonal cycle for the **Ctl** (gray), **Bio2** (green), **Ocean2** (blue) scenarios over the
 590 years 2012-2018. The black stars correspond to the mean seasonal amplitude for the observed COS
 591 mole fractions. The sites are listed on the abscissa. Bottom: Mean time of minimum crossing for

592 modelled (colored dots) and observed (black stars) COS for each scenario. For each site, the COS
593 time series have been detrended and filtered to remove the synoptic variability.

594

595 In this part, we assessed the sensitivity of the seasonal cycle and latitudinal distribution to a change
596 in biosphere fluxes and indirect COS emissions through DMS oxidation. For the biosphere part,
597 we consider two sets of biosphere fluxes produced by the ORCHIDEE LSM (**Bio 2** scenario) and
598 the SIB4 LSM (**Ctl** scenario). Compared to the SIB4 LSM, land uptake in the ORCHIDEE LSM
599 is 40 % lower over the tropical forests and over the eastern northern America (see Fig. S7). To
600 assess the sensitivity of the COS surface mole fractions to a change in the ocean component, we
601 compare the **Ctl** scenario against the **Ocean 2** scenario. The differences between the two fluxes is
602 noticeable mainly over the subtropical oligotrophic gyres and over southern high-latitude oceans
603 where the Belviso et al. (2012) DMS fluxes are 80% higher (see Fig. S8). In contrast, the latter are
604 50% weaker over the Western Pacific, which is not in line with the missing source location inferred
605 by top-down studies (Remaud et al., 2022, Glatthor et al., 2015, Kuai et al., 2015). The updated
606 version of the Lana et al. (2011) DMS climatology shows less DMS emissions over the Western
607 Pacific and over the Southern Indian ocean (Hulswar et al., 2022). The reader is referred to Section
608 2.2 for a description of these oceanic and biosphere fluxes.

609

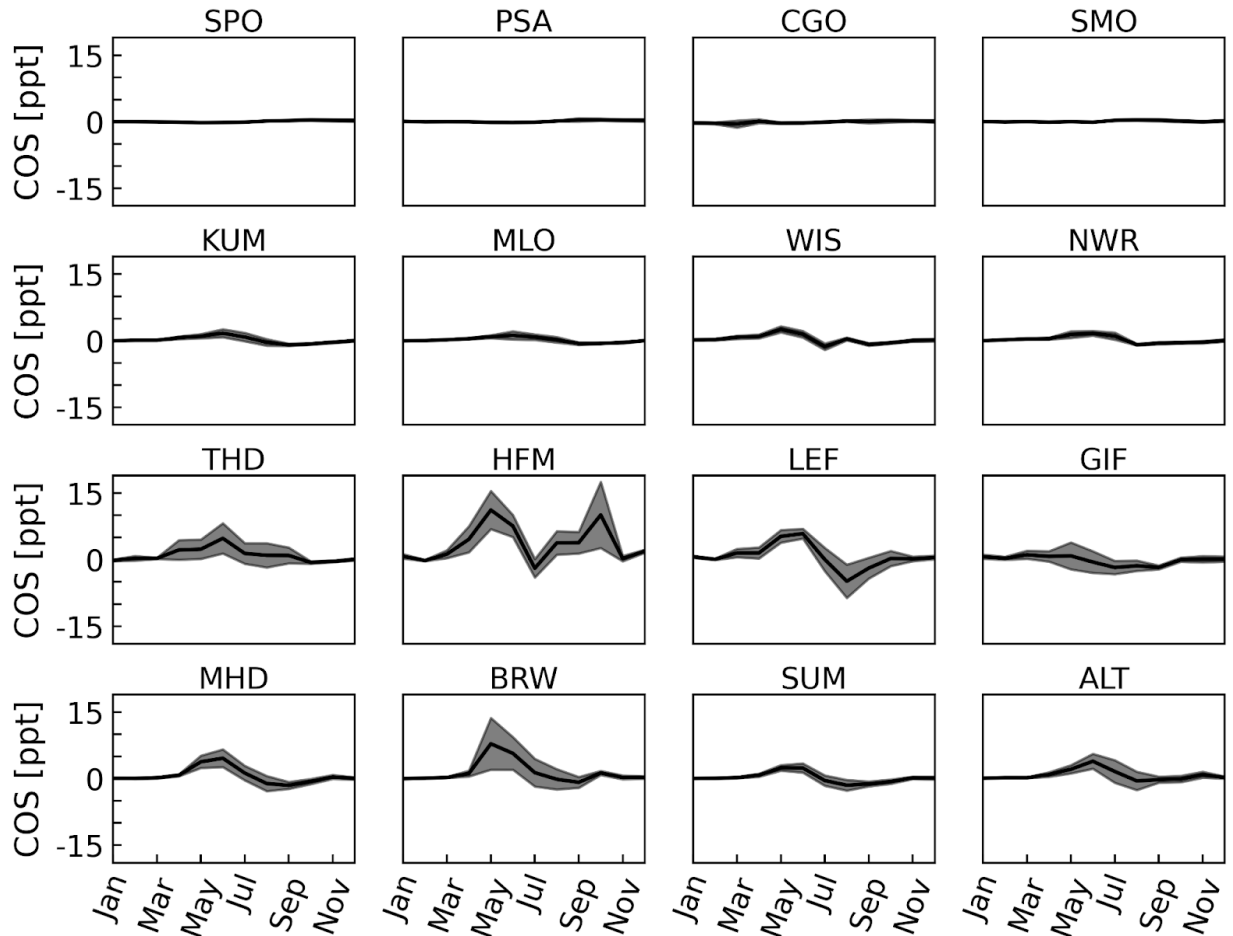
610 The annual gradient between a station and the MLO reference station relates to transport of
611 source/sinks within the regional footprint area of the station as well as to the background gradient
612 caused by remote sources. Figure 8 shows the boxplots of the mean annual gradients to MLO for
613 all stations for the observations and all ATMs. As the stations are ranked according to their
614 latitudes, Figure 8 enables us to compare the annual latitudinal repartition of COS simulated by all
615 ATMs using three scenarios, **Ocean 2**, **Bio 2** and **Ctl**. Except at site GIF, the change in either
616 biosphere fluxes or oceanic fluxes is translated into a change in mixing ratio that is smaller than
617 10 ppt and, also smaller than the model spread. The latter exceeds 50 ppt in the northern latitudes.
618 At sites GIF, HFM, LEF, the annual gradient to MLO is more sensitive to the biosphere fluxes as
619 the site is mainly influenced by continental air masses.

620

621 Figure 9 compares the mean seasonal cycle in terms of amplitude (top panel) and phase (bottom
622 panel) simulated by all ATMs using three scenarios, **Ocean 2**, **Bio 2** and **Ctl**. On the amplitude,
623 the effects of the biosphere and DMS fluxes are negligible compared to the model spread at most
624 sites. The seasonal amplitudes at sites HFM and LEF are more sensitive to the biosphere fluxes
625 than to the transport model and to the DMS fluxes as these sites sample continental air masses
626 coming primarily from areas covered by vegetation. The site HFM is located in a forest that
627 absorbs COS on average over the year (Commane et al., 2015). It should be noted that, at sites
628 HFM and LEF, the ORCHIDEE LSM simulates smaller seasonal cycle amplitudes than the SIB4
629 LSM. This is first because the ORCHIDEE LSM has a smaller plant absorption of COS than the
630 SIB4 LSM over northern America, also reflected by the global plant sink of COS (see Fig. S7) of
631 -514 GgS.yr^{-1} (ORCHIDEE) versus -669 GgS.yr^{-1} (SiB4). Secondly, at site HFM (Harvard
632 Forest), the soil fluxes of Abadie et al. (2022) have a smaller seasonal amplitude (Fig. 2 of Abadie
633 et al. (2022)) than the soil fluxes from Kooijmans et al. (2021) (Fig. 3 of Kooijmans et al. (2021)).
634 The absence of seasonal cycle in Abadie et al. (2022) is supported by the observations of soil fluxes
635 at Harvard Forest. The too low seasonal cycle amplitude compared to the observations suggests
636 again an underestimation of the COS plant uptake. Regarding the seasonal cycle phase on the

637 bottom panel of Figure 9, the change of biospheric and oceanic fluxes has a minor effect (by one
 638 month) on the minimum crossing. Only the seasonal phases at sites KUM and MLO are affected
 639 by several months by the change of DMS fluxes as these two stations are located in the Pacific
 640 Ocean. Note however that the two biosphere models do not represent the current diversity of global
 641 LSMs which have much larger variation in photosynthetic fluxes (see for instance Annav et al.,
 642 2013) and the selected ocean variants only differ by the indirect oceanic emissions of COS through
 643 DMS (and not by the direct emissions).

644
 645 3.3.2. Quantifying the diurnal rectifier effect on COS concentrations



646
 647 Figure 10: Monthly mean COS mole fractions obtained with the **Diurnal** scenario minus monthly
 648 mean COS mole fractions obtained with the **Ctl** scenario at each surface station for the year 2015.
 649 At each site, the solid line is the mean COS mole fraction across all models, and the shaded
 650 envelope represents the standard deviation around the mean.

651
 652 The simulated COS diurnal variation reflects the day–night contrast in both the prescribed fluxes
 653 and the PBL (planetary boundary layer) vertical mixing. The diurnal variability comes here from
 654 the plant fluxes, with minor contribution from the soil fluxes. The plants absorb more COS during
 655 the daytime when the stomatal opening enables the photosynthesis to happen. At night, plants
 656 continue to absorb COS as the carbonic anhydrase activity does not depend on light intensity
 657 (Protoschill-Krebs et al., 1996; Goldan et al., 1998) and the stomatal closure is incomplete.

658 Observed nighttime uptake was shown to be on average 25 % of the daytime uptake across several
659 sites located in Western Europe and Northern America between May–September (Kooijmans et
660 al., 2021). Another part of the diurnal variability is contributed by boundary layer processes: during
661 nighttime, COS accumulates near the surface within the shallower stable boundary layer, whereas
662 during daytime, the low COS concentration caused by the plant uptake is distributed over a deeper
663 convective PBL. Thus, the daily mean COS mixing ratio is expected to be greater than in the
664 absence of boundary layer processes and diurnal plant variability (Denning et al. 1995; Dennin et
665 al., 1999). This effect of the diurnal variability on longer time scales is called the diurnal rectifier
666 effect.

667 We quantify here the diurnal rectifier effect on the seasonal variability of COS surface mixing
668 ratios. To this end, Figure 10 shows the difference of monthly mean COS mixing ratio between
669 the **Ctl** scenario and the **Diurnal** scenario for the year 2015 at 16 surface stations. In the **Ctl**
670 scenario, the soil and plant fluxes are prescribed to the ATMs at monthly resolution whereas in the
671 **Diurnal** scenario, the soil and plant fluxes are prescribed at a three-hourly resolution. Averaged
672 over all ATMs, this effect is negligible and is less than the measurement uncertainties of 6 ppt at
673 11 stations out of 16. Even if the diurnal rectifier effect is more noticeable at sites HFM, LEF,
674 BRW, the difference between the **Ctl** and **Diurnal** scenario does not exceed 20 ppt. In summer,
675 the mainly positive difference in COS surface mixing ratios is induced by the temporal covariance
676 between strong vertical mixing and stronger COS uptake during the day. The difference of monthly
677 mean COS mixing ratio between the **Diurnal** scenario and the **Ctl** scenario results from the plant
678 absorption and not the soil fluxes (see Fig. S9). The soil fluxes have a small diurnal variability,
679 although on average, the soil flux becomes slightly less negative during the day when the abiotic
680 production term increases with growing temperature (Abadie et al., 2022). The use of the biosphere
681 fluxes from the ORCHIDEE LSM instead of the SIB 4 LSM leads to the same conclusion (see Fig.
682 S10).

683 To conclude, the diurnal rectifier effect for COS can be neglected when performing forward and
684 inverse modelling studies. This conclusion must be qualified considering the fact that the plant
685 uptake is underestimated in the two LSMs (Kooijmans et al., 2021, Maignan et al., 2021) and that
686 the long-term rectifier effect was not completely assessed. Because of the memory cost of saving
687 3 hourly fluxes, we only performed one year of the **Diurnal** scenario. Multi-year simulations
688 would allow an assessment of the effect of the rectifier effect on the mean latitudinal gradient
689 (Denning et al., 1995)

690 **4 Summary and conclusions**

691 With the participation of seven transport models, a control case has been constructed to evaluate
692 the state-of-the-art seasonal fluxes of COS while quantifying the transport errors, as another step
693 to better constrain the COS global budget. We analyzed the concentrations of COS simulated by
694 the atmospheric transport models at the location and time of surface and airborne campaign
695 measurements. Specifically, we focused the analysis both on the model-to-model and the model-
696 observations differences in:

- 697 1. Large-scale IH gradient, by comparing modeled and observed IH gradients of COS.
- 698 2. Simulated seasonal cycles, by comparing observed seasonal cycles at surface stations.
- 699 3. Vertical profiles of COS, by comparing modeled and observed vertical gradients of COS
- 700 between the PBL and the free troposphere.

701 In addition, we quantified the sensitivity of the seasonal cycle and the latitudinal distribution of
702 COS to a change in biosphere fluxes and to a change in oceanic fluxes. The diurnal rectifier effect

703 has also been quantified on the seasonal cycle of COS by comparing the COS mixing ratios given
704 by three-hourly fluxes and the COS mixing ratios given by monthly fluxes for the year 2015.

705

706 The main conclusions can be summarized as follows:

707 1. In regards to the mean seasonal cycle and the latitudinal distribution of COS mole fractions, the
708 model spread in COS simulations is mainly caused by the subgrid-scale parameterisation
709 (convective and boundary layer processes). However, in the vicinity of flux hot-spots, the model
710 resolution becomes crucial.

711 2. The model spread in COS surface mixing ratios is the largest in summer in the northern high
712 latitudes. The model spread at boreal sites can reach 70 ppt in summer, leading to divergences in
713 seasonal amplitude of more than 50 ppt. The transport errors can potentially lead to significant
714 uncertainties in the northern biosphere sink inferred through atmospheric inverse modeling using
715 COS observations.

716 3. Overall, the difference between the modeled and observed COS values is larger than the model
717 spread, pointing to incomplete knowledge of the COS budget, when using state-of-the-art
718 component fluxes.

719 4. In agreement with earlier studies, model-observation comparisons emphasize the need of a
720 missing tropical source, more biosphere uptake and likely smaller ocean emissions in the Northern
721 Hemisphere summer, especially at high latitudes.

722 5. Based on airborne measurements over Northern America, models predict a positive vertical
723 gradient between 1 and 4 km, while observations point to a negative gradient all year around, with
724 a stronger gradient in late summer. This again points to the need for stronger COS uptake over
725 Northern America.

726 6. Alternative flux combinations lead to similar conclusions. Indeed, the replacement of the
727 biosphere flux simulated by the SIB4 LSM (Kooijmans et al., 2021) by the biosphere fluxes
728 simulated by the ORCHIDEE LSM (Maignan et al., 2021; Abadie et al., 2022) in the **Ctl** scenario
729 leads to minor change in mean seasonal cycle and IH gradient. Likewise, the replacement of the
730 indirect ocean flux through DMS of Lana et al. (2011) by the ocean fluxes from Belviso et al.
731 (2012) in the **Ctl** scenario has a minor impact under the assumption of a global constant conversion
732 factor between DMS and COS (see discussion below).

733 7. The diurnal rectifier effect on the mean seasonal is negligible at most surface stations except at
734 a few continental stations over Northern America where the diurnal rectifier effect does not exceed
735 30 ppt. This implies that the use of monthly biosphere fluxes instead of three-hourly biosphere
736 fluxes is an acceptable simplification for COS budget studies. However, the assessment of the
737 diurnal rectifier effect on the latitudinal distribution would require to perform the same experiment
738 but over several years.

739 **5 Discussion and future work**

740 The atmospheric chemistry of COS was not included in the ATMs to isolate the transport errors.
741 However, the chemistry related to COS remains poorly resolved. The current notion of the
742 atmospheric chemistry of COS is that 100 GgS.yr⁻¹ is oxidized in the atmosphere and 50 GgS.yr⁻¹
743 is photolyzed in the stratosphere. Because of the small importance of these sources in the COS
744 budget, their introduction in the ATMs is not expected to modify the conclusions of this study. A
745 second assumption of this study is that the DMS emitted by the ocean is instantaneously oxidized
746 into COS with a yield from Barnes et al. (1996). Recently, a **stable** intermediate from DMS
747 oxidation, the hydroperoxymethyl thioformate (HPMTF), has been discovered to be the main

748 precursor of COS (Jernigan et al., 2022). The introduction of this chemical pathway in an
749 Atmospheric Chemistry Transport Model led to more COS emissions in the tropics but with a
750 magnitude three times lower than the DMS fluxes used in this study (Jernigan et al., 2022).
751 However, these results are still preliminary. If these reactions are confirmed by more chamber
752 studies and observations in the future, the full chemistry of DMS and COS needs to be taken into
753 account to accurately evaluate the state of the art COS fluxes.

754
755 This analysis, focused here on the mean seasonal cycle and the inter-hemispheric gradient, could
756 be extended in the future to the inter-annual variations and the long term trend of the COS mixing
757 ratios. The trend was not analysed because some inter-annual fluxes (e.g. anthropogenic) were not
758 always available. Moreover, the COS mixing ratios derived from the atmospheric inversion of Ma
759 et al. (2021) that was used to rescale the biosphere fluxes were climatological, which is not realistic
760 in regards to the current decreasing trend of COS mole fraction since 2014 and its implication on
761 biosphere fluxes (Belviso et al., 2022).

762
763 Finally, the sparse and uneven cover of the observations limits the evaluation of the COS fluxes to
764 the footprint area of these stations. A complementary paper will also evaluate the COS fluxes using
765 airborne measurements from the Atoms and HIAPER Pole-to-Pole Observations (HIPPO; Wofsy,
766 2011) campaigns. Although they are limited in time, these measurements will give additional
767 insight to the COS fluxes over the tropical Atlantic and Pacific Ocean. Satellites offer the
768 perspective of constraining the tropical areas over long periods of time (Glattor et al., 2015;
769 Stinecipher et al., 2022, Vincent and Dhunia, 2017), but the retrievals still entail large uncertainties
770 (Whelan et al., 2018; Serio et al., 2021). The complementary paper will evaluate the fluxes of COS
771 at several FTIR stations and will quantify the transport errors.

772

773 **Annexe: Additional transport model description**

774

775 LMDz

776 The LMDz ATM has a spatial resolution of $3.75^\circ \times 1.9^\circ$ (longitude times latitude) with 39 layers
777 in the vertical, based on the general circulation model developed at the Laboratoire de
778 Météorologie Dynamique, LMDz (Hourdin et al., 2020). LMDz6A is our reference version: it was
779 prepared for the 6th Climate Intercomparison Project (CMIP6) as part of the Institut Pierre-Simon
780 Laplace Earth system model. We use the offline version of the LMDz code, which was created by
781 Hourdin and Armengaud (1999) and adapted by Chevallier et al. (2005) for atmospheric inversion.
782 It is driven by air mass fluxes calculated by the complete general circulation model, run at the
783 same resolution and nudged here towards winds from the fifth generation of meteorological
784 analyses of the European Centre for Medium-Range Weather Forecasts (ECMWF) (ERA5). The
785 off-line model only solves the mass balance equation for tracers, which significantly reduces the
786 computation time. This LMDz version recently participated in the TRANSCOM experiment for
787 CO₂ weather (Zhang et al., 2022).

788

789 TM5

790 TM5 is the global chemistry Transport Model, version 5 (TM5) (Krol et al., 2005). It allows two-
791 way nested zooming and is specifically useful for multiple-resolution zooming modeling of trace
792 gases in troposphere and stratosphere. The earlier version of TM5 is the parent TM3 model, which
793 was originally developed by Heimann et al. (1988) and has been widely used in global atmospheric
794 chemistry studies. TM5 is designed for tracer models and it is used extensively in inversion studies
795 for various trace gases, e.g., CO, CO₂, CH₄ and COS. In this study, we used the forward-mode of
796 TM5-4DVAR for COS at a high resolution of 2° × 2° with vertically 25 layers.

797

798 MIROC4

799 MIROC4-ACTM is a new generation Model for Interdisciplinary Research on Climate (MIROC,
800 version 4.0) based atmospheric chemistry-transport model (Patra et al., 2018). The horizontal
801 triangular truncation at a total horizontal wave number of 42 (T42; latitude and longitude ~2.81 ×
802 2.81°) is used in the present study. MIROC4-ACTM has the fully resolved stratosphere and
803 mesosphere by implementing the hybrid vertical coordinate of pressure-sigma (surface to about
804 the tropopause) and pressure (about 300 hPa and above). The MIROC4-ACTM has a spectral
805 dynamical core and uses a flux-form semi-Lagrangian scheme for the tracer advection (Lin and
806 Rood 1996). The radiative transfer scheme considers 37 absorption bands, consisting of 23 in the
807 visible and ultraviolet regions enabling better representation of photolysis for chemical species
808 (Sekiguchi and Nakajima, 2008). The cumulus convection scheme is based on Arakawa and
809 Schubert (1974), in which cloud base mass flux is treated as a prognostic variable. The sub-grid
810 vertical mixing is parameterized based on the level 2 scheme of the turbulence closure (Mellor and
811 Yamada 1982). The model participated in various model intercomparison projects, e.g.,
812 TransCom-air of air (Krol et al., 2018), and flux inversions are performed for CO₂, CH₄ and N₂O
813 (Chandra et al., 2021a,b; Patra et al., 2022) which have contributed to various international
814 emission and removal budget assessments.

815

816 TOMCAT

817 TOMCAT/SLIMCAT is a global 3-D off-line chemical transport model (Chipperfield, 2006). It is
818 used to study a range of chemistry-aerosol-transport issues in the troposphere and stratosphere.
819 The model is usually forced by ECMWF meteorological (re)analyses, although GCM output can
820 also be used. When using ECMWF fields, as in the experiments described here, the model reads
821 in the 6-hourly fields of temperature, humidity, vorticity, divergence and surface pressure. The
822 resolved vertical motion is calculated online from the vorticity. Tracer advection is performed
823 using the conservation of second-order moments scheme of Prather (1986). For the experiments
824 described here the model was run at horizontal resolution of 2.8° × 2.8° with 60 hybrid σ -pressure
825 levels from the surface to ~60 km. The model was forced by ECMWF ERA-Interim reanalyses
826 (Dee et al., 2011). Convective mass fluxes were also taken from ERA-Interim reanalyses and
827 mixing in the boundary layer is based on the scheme of Louis (1979), as described in Stockwell
828 and Chipperfield (1999).

829 NICAM-TM

830 NICAM-TM is an atmospheric transport model based on Nonhydrostatic Icosahedral Atmospheric
831 Model (NICAM: Satoh et al. 2014), which has an icosahedral grid system. The mean grid interval

832 is 223 km and 112 km for “glevel-5” and “glevel-6”, respectively. Both the horizontal resolutions
833 have the same vertical layer, whose number is 40 and the model top is approximately 45 km.
834 Although NICAM-TM has on-line and off-line modes for atmospheric calculations, the off-line
835 mode (Niwa et al., 2017) is used in this study, whose meteorological data are derived from an on-
836 line NICAM-TM calculation with horizontal wind nudging. During both off-line and on-line
837 calculations, mass conservation is achieved without any numerical mass fixer (Niwa et al. 2011,
838 2017). Using NICAM-TM, several inverse analyses of greenhouse gases have been performed
839 (e.g., Niwa et al., 2021).

840
841

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852 collection of flask samples at WIS.
853

854 **Open Research**

855 The LMDz model is available from <http://svn.lmd.jussieu.fr/LMDZ/LMDZ6/> under the CeCILL
856 v2 free software license. The COS time series at station GIF from 2014 to 2019 are provided by
857 Sauveur Belviso and can be downloaded from [https://sharebox.lsce.ipsl.fr/index.
858 php/s/Yxbj6dZsrc6nsOZ?path=2FGIF-observations](https://sharebox.lsce.ipsl.fr/index.php/s/Yxbj6dZsrc6nsOZ?path=2FGIF-observations) (last access: 22 August 2022).
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Intercomparison of atmospheric Carbonyl Sulfide (TransCom-COS; Part one): Evaluating the impact of transport and emissions on tropospheric variability using ground-based and aircraft data

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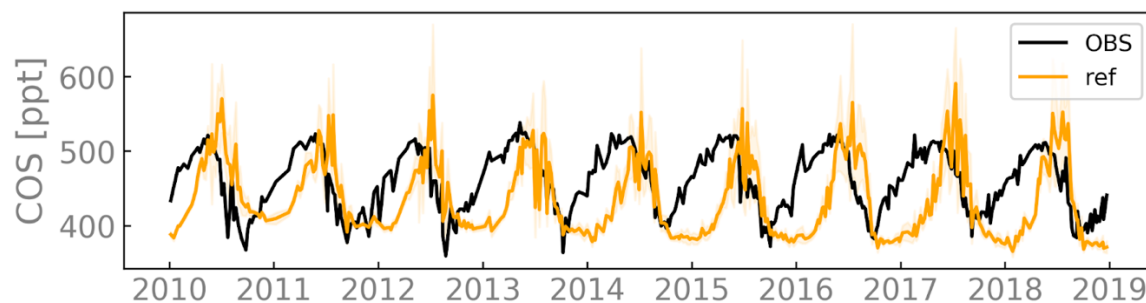


Figure S1. Temporal evolution of the surface COS mixing ratio at site BRW as observed (black) and simulated by several Atmospheric Transport Models (orange) using the **Ctl** scenario. The full line in orange is the averaged concentrations simulated by all transport models and the shaded area is the standard deviation at each time step of the simulated concentrations by all transport models. The simulated COS abundances have been shifted of 396 ppt, which is the observed concentrations averaged over all surface sites for January 2010.

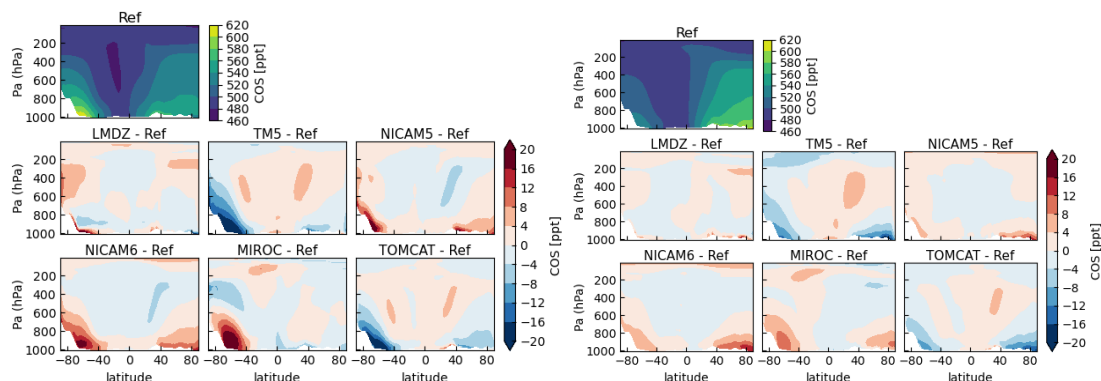


Figure S2: Zonal mean mole fraction of COS in ppt for the reference for the **Ctl** scenario (top row). The reference is the average of COS over all transport models. Second and third rows: Zonal mean mole fraction difference between each transport model and the reference. Left: The zonal mean is averaged in winter (DJF) from 2012 to 2018. Right: The zonal mean is calculated from 2012 to 2018 (annual mean).

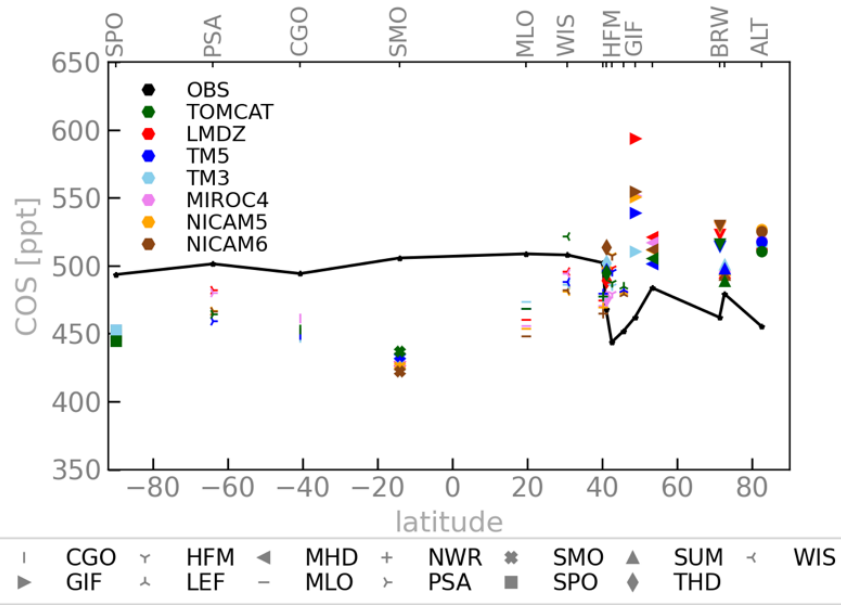


Figure S3: Comparison of the latitudinal variations of the COS abundance simulated by several transport models using the **Ctl** surface flux dataset (colored dots) with the observations (black line) averaged over the years 2012-2019. The simulated COS abundances have been shifted such that the means are the same as the mean of the observations (~ 500 ppt). The curves have been detrended and filtered to remove the synoptic variability. The value at site GIF simulated by the TOMCAT ATM was removed as it was an outlier (value above 755 ppt).

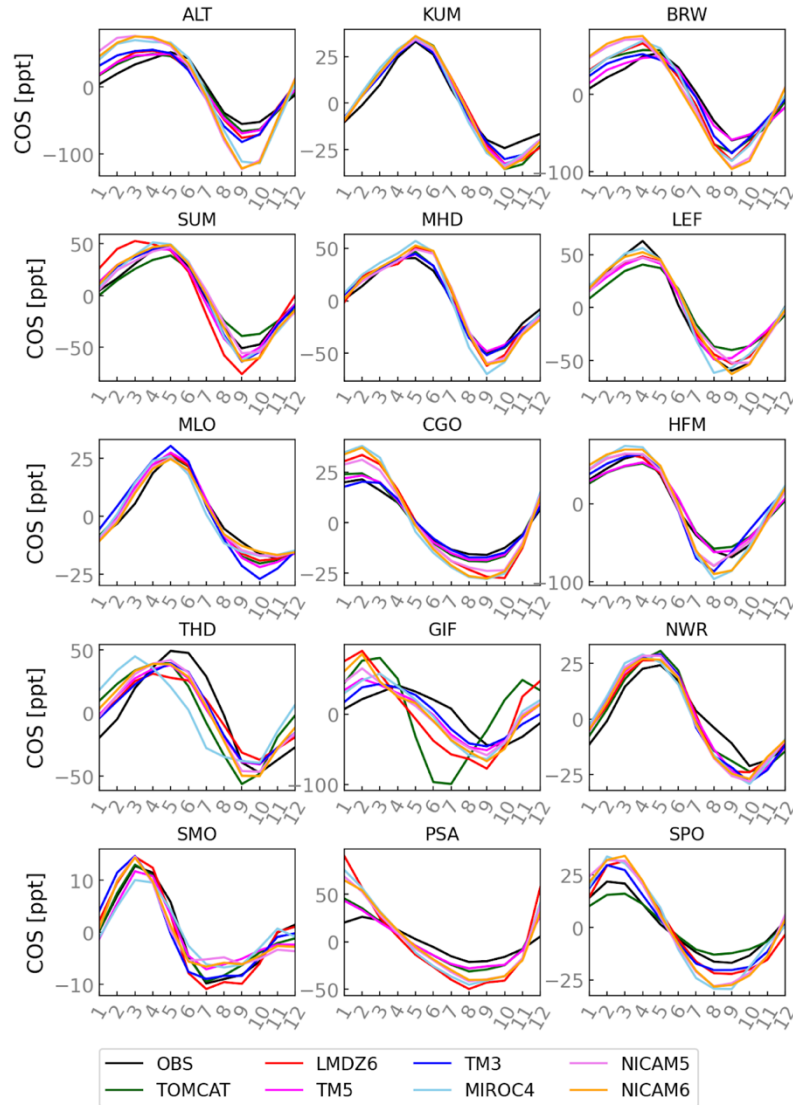


Figure S4 : Mean seasonal cycle of the observed (black) and simulated (color) COS mixing ratios at 15 surface sites for the Ctl scenario. The curves have been detrended and filtered to remove the synoptic variability.

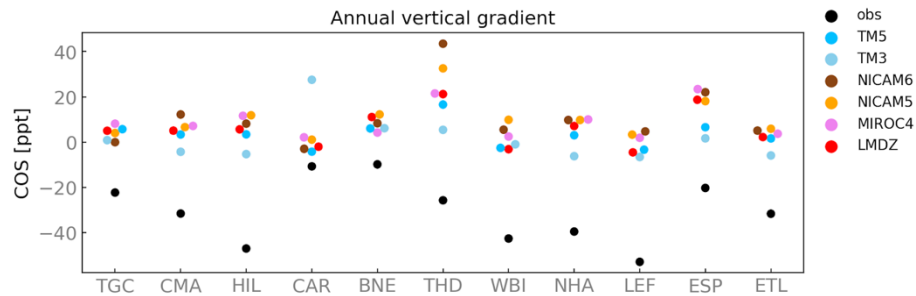


Figure S5. Annual mean observed and simulated COS gradient between 1 and 4 km at each airborne station for the Ctl scenario. For each subregion, the monthly COS gradients are calculated by averaging the differences in COS concentrations between 1 and 4 km over all the vertical profiles.

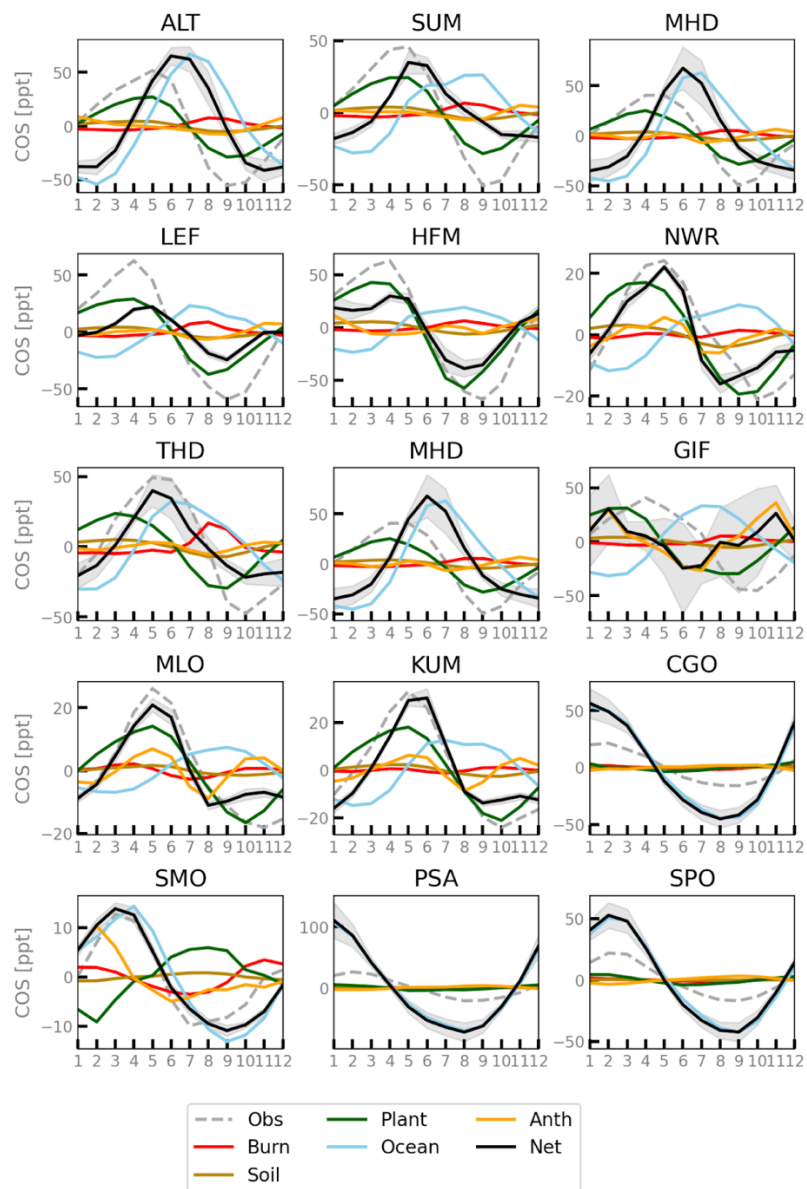


Figure S6: Simulations of the seasonal cycle of tropospheric COS mixing ratios at several surface stations averaged over all transport models. The shaded area is the standard deviation around the mean COS seasonal cycle associated with the different transport models. The dotted black line represents the observed seasonal cycle.

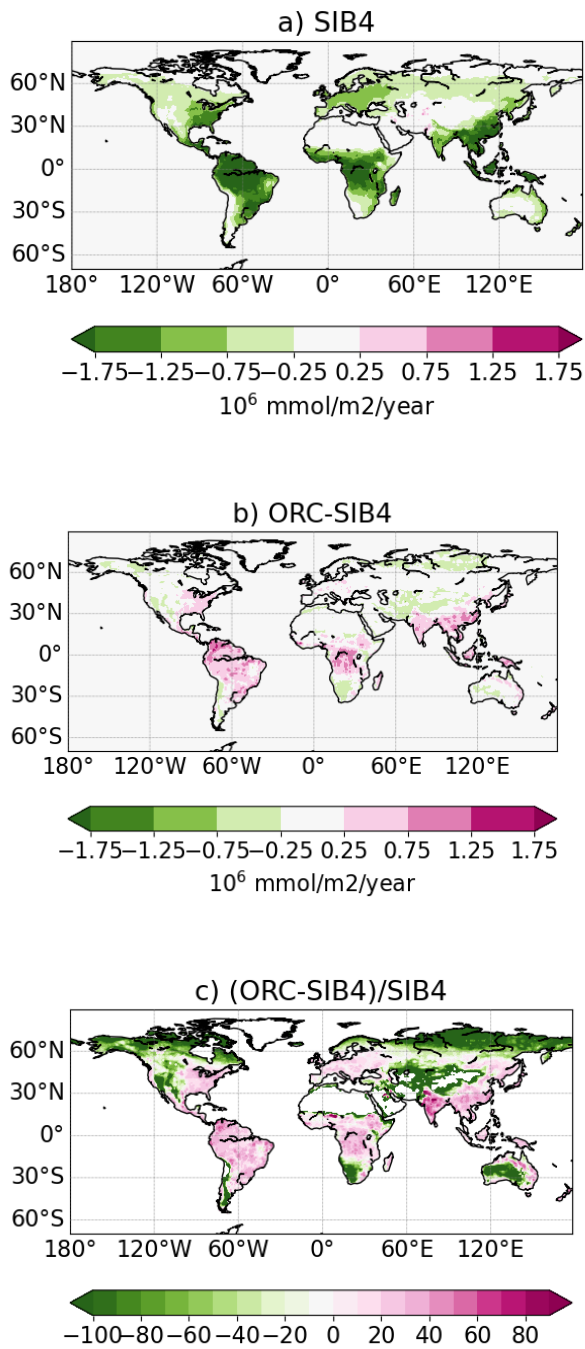


Figure S7: a) Climatology of the biosphere flux of COS (mmol/m²/yr) in the SIB 4 LSM, b) Climatology of the difference of the biosphere flux (mmol/m²/yr) between the ORCHIDEE LSM and the SIB 4 LSM. c) Same as b) but in terms of percentage.

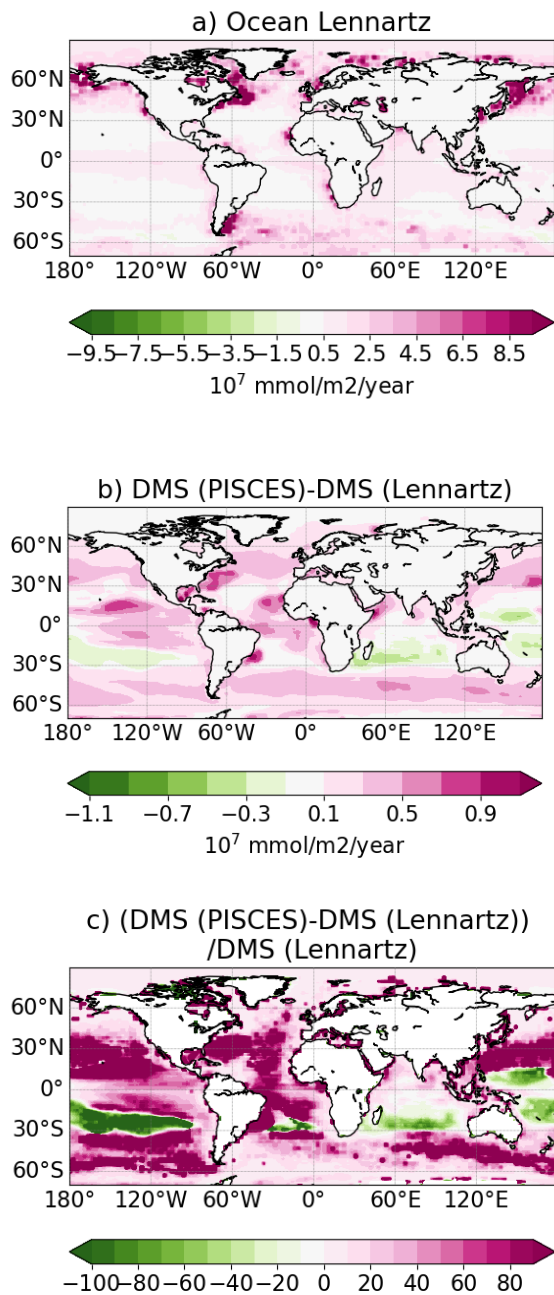


Figure S8 a) Climatology of the ocean flux ($\text{mmol}/\text{m}^2/\text{yr}$) using the Lennartz et al., 2017 DMS fluxes, b) Climatology of the difference of the ocean flux ($\text{mmol}/\text{m}^2/\text{yr}$) between the DMS fluxes of Lennartz et al., 2017 and the DMS fluxes simulated by the NEMO-PICSES Ocean Model. c) Same as b) but in terms of percentage.

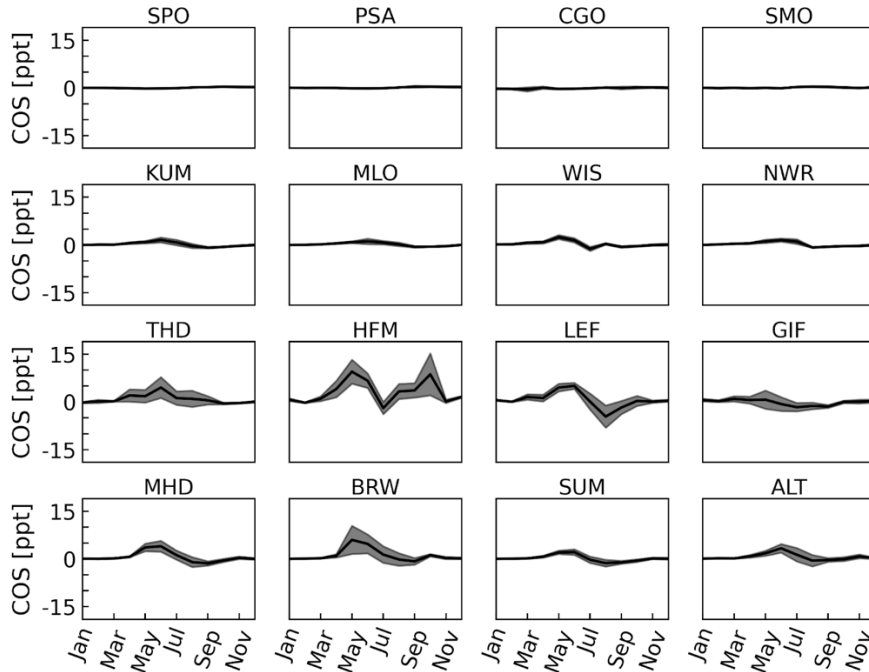


Figure S9 Difference of monthly mean COS mole fractions between the **Diurnal** scenario and the **Ctr** scenario (without the soil fluxes) at each surface station for the year 2015. At each site, the solid line is the mean COS mole fraction across all models, and the shaded envelope represents the standard deviation around the mean. Here, only the vegetation fluxes contribute to the difference of COS mole fractions.

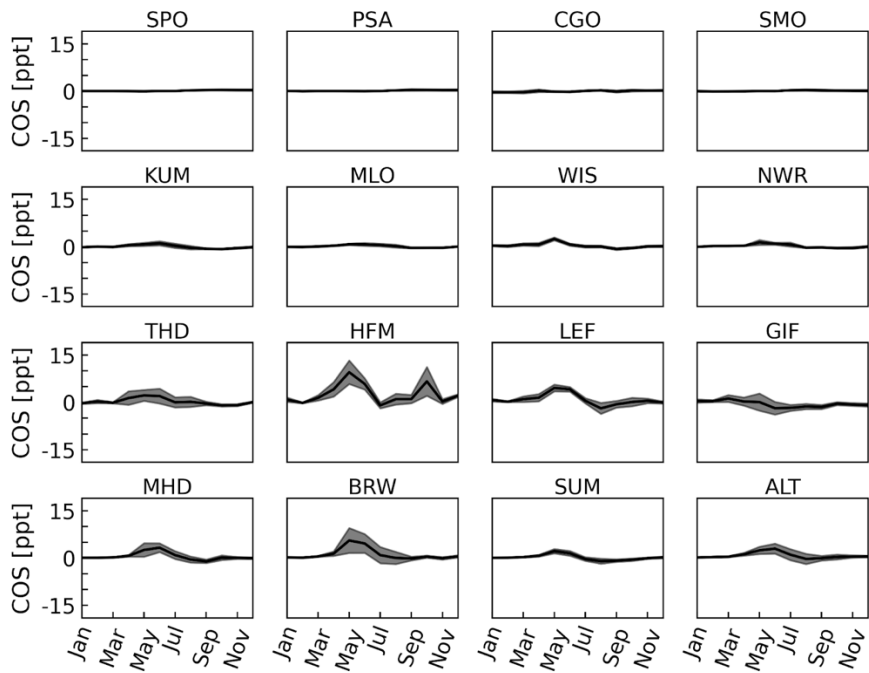


Figure S10 Difference of monthly mean COS mole fractions between the **Diurnal 2** scenario and the **Bio 2** scenario at each surface station for the year 2015 with the LSM

ORCHIDEE. At each site, the solid line is the mean COS mole fraction across all models, and the shaded envelope represents the standard deviation around the mean.

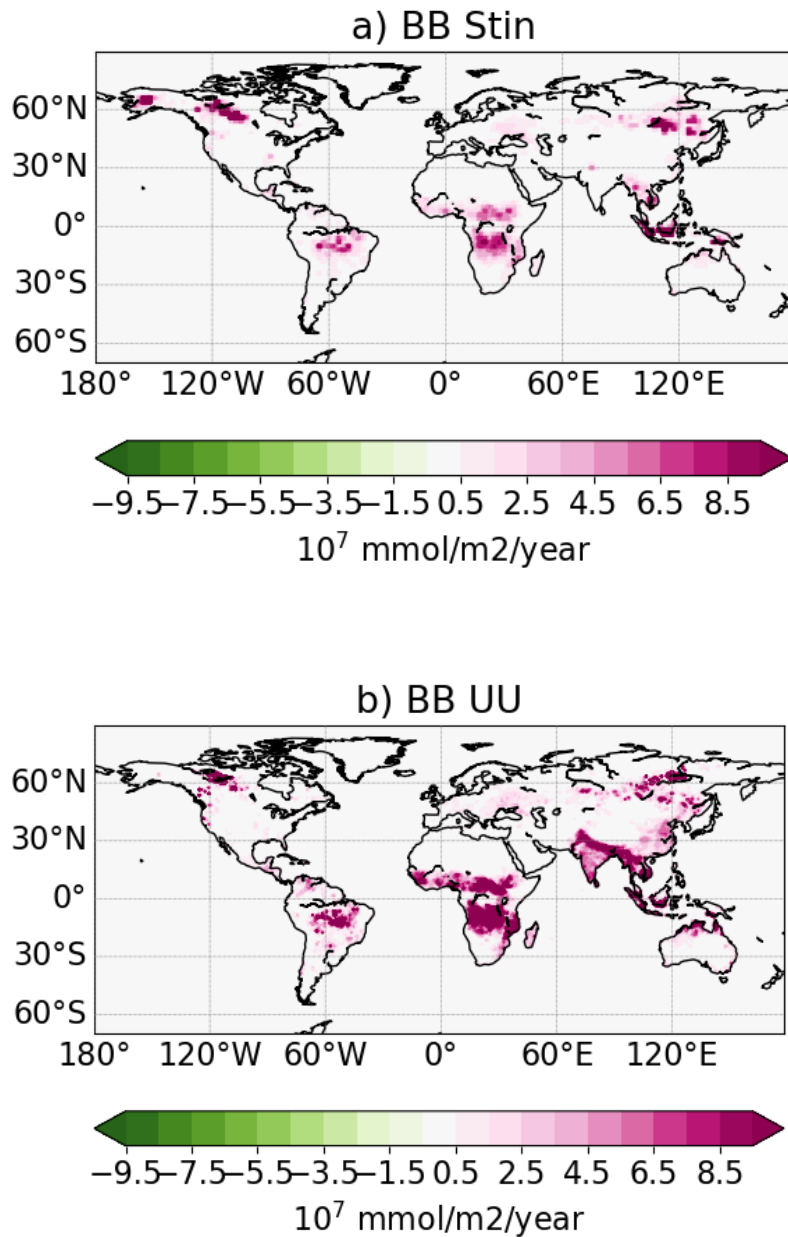


Figure S11: a) Climatology of the biomass burning flux (mmol/m²/yr) from Stinecipher et al., 2019. b) Climatology of the biomass burning flux (mmol/m²/yr) from Ma et al., 2021, which takes into account the biofuel use.