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Key Points:

- Bottom-up estimate of carbonyl sulfide (OCS) emissions from biomass burning is insufficient to close the existing OCS budget gap
- Long-term observational records indicate that some previously reported OCS emission factors from peat are not globally representative
- Biome-specific emission factors imply a spatial redistribution of emissions relative to previous studies using fixed emission factors

Supporting Information:

- Supporting Information S1
- Table S1
- Table S2

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Biomass Burning Unlikely to Account for Missing Source of Carbonyl Sulfide

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Abstract Carbonyl sulfide (OCS) provides a proxy for measuring photosynthesis and is the primary background source of stratospheric aerosols. OCS emissions due to biomass burning are a variable and substantial (over 10%) part of the current OCS budget. OCS emission ratios from open burning fires, coupled with 1997–2016 data from the Global Fire Emissions Database (GFED4), yield OCS biomass burning emissions with a global average annual flux of 60 ± 37 Gg(S) year⁻¹. A global box model suggests these emissions are more consistent with observations from global atmospheric composition monitoring networks than fluxes derived from previous synthesis papers. Even after considering the uncertainty in emission factor observations for each category of emissions and the interannual variation in total burned dry matter, the total OCS emissions from open burning are insufficient to account for the large imbalance between current estimates of global OCS sources and sinks.

Plain Language Summary Carbonyl sulfide is a naturally occurring gas that can help us understand how much carbon dioxide plants take out of the atmosphere for photosynthesis. In this study we want to understand how much carbonyl sulfide comes from forest fires and other burning, as opposed to other sources. We estimate carbonyl sulfide emissions from fires around the world based on where fires occurred and what was burned then use a computer model to see whether our estimates line up with real-world measurements of carbonyl sulfide in the atmosphere. We find that fires are a smaller source of carbonyl sulfide than previously reported.

1. Background

Carbonyl sulfide (abbreviated OCS or COS) is the most abundant sulfur gas in the atmosphere, and is important both as a tracer for gross primary production and as a source of stratospheric aerosols. Current estimates of surface fluxes and atmospheric sinks of OCS do not fully account for all processes necessary to balance the OCS budget. For example, Berry et al. (2013) assume a deficit in the OCS source of approximately 600 Gg(S) year⁻¹ (annual mass of sulfur emitted as OCS), compared to a total sink of approximately 1,200 Gg(S) year⁻¹. While ocean chemistry has been hypothesized as a possible way to close this budget gap (Berry et al., 2013; Suntharalingam et al., 2008), other sources including biomass burning, anthropogenic emissions, and anoxic soils may be partially responsible.

Biomass burning has long been recognized as a source of OCS, particularly in smoldering fires (Chin & Davis, 1993; Crutzen et al., 1979, 1985; Crutzen & Andreae, 1990; Kettle et al., 2002; Khalil & Rasmussen, 1984). However, due to differing methodologies and study assumptions, estimates of global total OCS emissions vary widely in the literature (11 Gg(S) year⁻¹ in Kettle et al., 2002, 136 Gg(S) year⁻¹ in Berry et al., 2013, and 68–144 Gg(S) year⁻¹ in Montzka et al., 2007).

Emissions of OCS from biomass burning can be measured and reported in several ways (Akagi et al., 2011). Samples may be burned in a lab setting, allowing for simultaneous measurements of changes in mass and composition of the emitted plume (e.g., Yokelson et al., 1997). When the mass of fuel is known, measurements may be reported as emission factors (EFs), defined as the mass of a species emitted relative to the mass of dry matter (DM) burned (e.g., kg OCS/kg biomass). Samples may also be taken in the field where fires are

present, either at ground level (e.g., Meinardi et al., 2003; Nguyen et al., 1995) or by flying through a plume (e.g., Blake et al., 2004; Nguyen et al., 1995). In the field, it can be challenging to accurately measure the total mass burned, and hence, field measurements are frequently reported as emission ratios (ERs), that is, relative enhancements over background values (e.g., $[\text{mol OCS}_{\text{plume}} - \text{mol OCS}_{\text{background}}]/[\text{mol CO}_{\text{plume}} - \text{mol CO}_{\text{background}}]$, yielding units of mol OCS/mol CO).

The Global Fire Emissions Database (GFED version 4, <https://www.globalfiredata.org/>) provides spatially and temporally resolved maps of fire emissions in a variety of categories based on satellite observations (Giglio et al., 2013; Randerson et al., 2012; van der Werf et al., 2017), as well as a summary table of emission factors for a variety of species (not including OCS), primarily based on Andreae and Merlet (2001) and Akagi et al. (2011). We seek to expand these existing works with updated average emission ratios for OCS, suitable for use with burned totals from GFED.

2. Methods

We collected all the published emission factors and emission ratios for carbonyl sulfide, along with corresponding measurements of CO and CO₂ emissions, as available (supporting information Table S1). Based on fuel type and sample location, we assigned samples to one of the six biomass burning categories used in GFED: savanna and grassland, boreal forests, temperate forests, tropical deforestation and degradation, peatland fires, and agricultural waste burning. Categorized samples are shown in supporting information (Table S2) with mean values for each category given in Table S3.

Crutzen and Andreae (1990) and Meinardi et al. (2003) show that while CO₂ peaks in the flaming stage, CO and OCS both peak during the later smoldering stage. Hence, in order to combine emission measurements between observational data sets, we compute all possible values as emission ratios relative to CO (mol OCS emitted/mol CO emitted). This has the benefit of including the many studies where the total mass burned was not available but does exclude several studies where CO was not measured. Where emission ratios are not given directly, we calculate them either as the ratio of OCS and CO emission factors or as an enhancement in OCS relative to CO as described above, depending on how data are presented in a given study (see supporting information Text S1).

To test the impact of these emission ratios on global OCS concentrations, we first compute total CO emissions for each category in the GFED4 inventory, using emission factors provided by GFED to convert from total burned dry matter to CO. We then apply the emission ratios described above to these CO totals to obtain global total emissions of OCS in each category.

The OCS emissions are used as an input in a one-box global model, run at monthly time steps, and driven by zero-order sources (biomass burning, biofuels, anthropogenic emissions and oceanic emissions), a zero-order soil sink, and first-order sinks for plant uptake and destruction of OCS by hydroxyl radicals. Zero-order fluxes vary independently of ambient atmospheric OCS concentrations, while first-order fluxes vary proportionally to ambient atmospheric OCS concentrations. A constant, supplemental source is added to maintain a balanced budget. Previous works (Berry et al., 2013; Campbell et al., 2017; Suntharalingam et al., 2008) ascribe this flux to a missing ocean source; however, more recent estimates by Lennartz et al. (2017, 2019) find that ocean emissions are insufficient to close the budget and other fluxes must be involved. The model assumes that the atmosphere is well mixed at every time step, which is reasonable because the atmospheric lifetime of OCS is approximately 2 years (Campbell et al., 2008; Khalil & Rasmussen, 1984; Montzka et al., 2007). Additional details are given in supporting information (Text S2).

We compare the outputs of this box model to NOAA ground-based flask samples taken at a variety of sites since approximately 2000 (Montzka et al., 2007) and annually averaged tropospheric mixing ratios derived from Fourier transform infrared (FTIR) solar spectra recorded at the Jungfraujoch station (Swiss Alps) since 1984 (Lejeune et al., 2017). The NOAA data sets provide a global perspective on OCS trends from 2000 onward, whereas the Jungfraujoch data set provides one of the only time series of OCS mixing ratios covering the entire GFED4 data period (1997–2016). Measurements at Jungfraujoch are specific to that site but are still influenced by changes in global concentrations like those predicted by the global box model because of the long atmospheric lifetime of OCS.

Figure 1. Comparison of emission ratio estimates from various literature sources. In order to span the wide range of values, the grey region of the top panel is shown with an expanded scale in the bottom panel. Black crosses are individual studies or measurements. Blue dots are the averages used in this paper. Red circles and triangles are average values from Andreae and Merlet (2001) and Akagi et al. (2011), respectively. Error bars represent the standard deviation of emission ratio observations included in the average value for each category. Categories are described in section 2 of the main text. Additional details and numerical data are given in the supporting information.

3. Results

Figure 1 shows the range of observed emission ratios for the six categories used in GFED. Emission ratios from both lab and field studies are shown as black crosses. Previous estimates from Andreae and Merlet (2001) and Akagi et al. (2011) use subsets of these data to estimate average emission factors by ecosystem type. As neither inventory covers all of the biome categories, we combine both inventories by using the reported value (if only one study provides a value) or averaging values from both studies, if available.

Akagi et al. (2011) present OCS and CO emission factors for boreal, temperate (extratropical), and peat ecosystems that are an order of magnitude larger than the other three categories (the three highest red triangles in Figure 1). This is due to the inclusion of an anomalously high observation of OCS emitted from a boreal peat sample in a chamber experiment (Yokelson et al., 1997—Table 1, Minnesota Peat). Compared to another peat sample from the same study (Yokelson et al., 1997—Table 1, Alaska Peat), this sample yielded over 25 times the OCS per dry matter burned and 35 times the OCS relative to CO. The emission ratio for this sample relative to CO is also more than 40 times larger than the emission ratio computed for a sample of Indonesian peat in Stockwell, Jayarathne, et al. (2016). Dixon's Q test (Dean & Dixon, 1951) identifies this largest peat sample as an outlier with 95% confidence, and we thus exclude it from our averages.

Our goal is to compare the impact of different possible emission factors against observed OCS concentrations. Specifically, we compare the impact of prior emission estimates to an updated calculation of average emission ratios, which includes a wider set of observational data than in previous studies while excluding the anomalously high peat observation on the basis that it may not be representative of most locations. Our emission ratios for boreal, temperate, and peat ecosystems are an order of magnitude lower than estimates from Akagi et al. (2011), but within 25% of those presented in Andreae and Merlet (2001). Average emission ratios for savanna, tropical deforestation, and agricultural waste are within 40% of those presented in Akagi et al. (2011) and Andreae and Merlet (2001). Across categories, our emission ratios range from 1×10^{-4} mol OCS/mol CO (savanna and grassland) to 3×10^{-4} mol OCS/mol CO (agricultural waste burning).

We apply these emission ratios to global biomass burning inventories from GFED in order to estimate the corresponding OCS emissions. The GFED estimates of burned dry matter are dominated by savannas, followed by tropical deforestation and boreal forests (Figure 2a). The GFED estimates show particularly large interannual variability for peatlands, with nearly 10 times more peat burned in 1997 than the 1998–2016 average due to massive fires in Indonesia during a large El Niño event (Page et al., 2002). A similar, smaller spike occurs during the 2015 El Niño event.

Figure 2. Annual total GFED4 estimates of (a) burned dry matter (DM) and (b) associated emissions of CO based on GFED4 emission factors. OCS emissions based on (c) combined emission ratios from Andreae and Merlet (2001) and Akagi et al. (2011) and (d) this study. Dotted lines on panels (c) and (d) represent the average annual total across all years. Error bars in panel (d) are derived from the relative uncertainty in CO emission factors and OCS emission ratios, described in supporting information (Text S2). Note the change in y axis scaling between panels (c) and (d).

As mentioned above, we use CO as a reference species to compute OCS emissions. These CO emissions are computed from the emission factors provided by GFED and are shown in Figure 2b.

We combine these GFED CO emissions with (i) OCS emission ratio averages from Andreae and Merlet (2001) and Akagi et al. (2011) and (ii) the emission ratio averages we calculated to create a temporally and spatially varying OCS burning inventory (Figures 2c and 2d, respectively). Note that while savannas and grassland account for 52–78% of burned matter depending on the year (Figure 2a), they are a small source of OCS (3–26% of biomass burning emissions using emission ratios from previous synthesis papers and 21–51% of emissions using the present emission ratios) due to the relatively small emission ratio (Figures 2c and 2d). Conversely, peatland emissions are a small and variable part of the dry matter total (0.2–15%) but a major source of OCS, especially using emission ratios derived from previous synthesis papers (8–88% of emissions using previous emission ratios, 1–42% using present emission ratios).

Fluxes derived from previous emission ratio inventories are larger in magnitude and in interannual variability (mean 203 Gg(S) year⁻¹, standard deviation 161 Gg(S) year⁻¹ over all years) than those derived from our present estimates (mean 60 Gg(S) year⁻¹, standard deviation 15 Gg(S) year⁻¹ over all years). Additionally, while 1997 is an exceptional year for both sets of emission factors, the spike in emissions is more pronounced in the previous estimates: total 1997 OCS emissions are 4.6 times larger than the mean from 1998 to 2016 (compared to 1.9 times using our emission ratios). In the years following 1997, the previous emission ratios also yield emissions with coefficients of variation (standard deviation divided by mean) twice as large as those from our present emission ratios.

We examine the validity of these two alternative OCS emission scenarios through two runs of a global atmospheric box model, validated against atmospheric observations (Figure 3a). As in previous work, the simulation budget is balanced by adding a supplemental constant source (Berry et al., 2013; Campbell et al., 2017; Suntharalingam et al., 2008) to other sources and sinks described above (Figures 3b and 3c). Note that the supplemental source for the previous inventories is considerably smaller due to their larger average biomass burning contribution.

Figure 3. (a) Box model outputs and observations from NOAA air monitoring sites (Montzka et al., 2007) and the tropospheric FTIR data from Jungfraujoch, Switzerland (Lejeune et al., 2017). Previous emission factors are based on Andreae and Merlet (2001), Akagi et al. (2011), or an average of both. The shaded region represents the uncertainty due to the range in emission factors and emission ratios. (b and c) Annual fluxes of OCS by category as used in the “previous” and “current” models, with net fluxes indicated by the white dotted line. In both (b) and (c), a supplemental flux is added to close the budget, described in section 3. Additional details are given in the supporting information (Text S2)

Observed atmospheric surface mixing ratios from global and hemispheric averages of NOAA air monitoring sites (Montzka et al., 2007), as well as annually averaged tropospheric mixing ratios from Jungfraujoch (Lejeune et al., 2017), show a moderate decline in the 1990s followed by similarly moderate growth in the 2000s (both approximately $1\% \text{ year}^{-1}$), with relatively little interannual variation. These time trends have been attributed to trends in the anthropogenic inventory, driven by a decline in residential coal in the early 1990s and the decline and growth of the rayon industry from 1990 onward (Campbell et al., 2015; Du et al., 2016; Zumkehr et al., 2017, 2018).

A simulation using our new emission ratios yields concentrations and interannual variations that are broadly consistent with the observational records. In contrast, a simulation using emission ratios from the previous inventories is unable to capture these trends. In particular, the spike in concentrations following emissions in 1997 and the rapid growth from 2012 onward are accentuated in the model output but largely absent in the observations.

We quantitatively assess these differences in two ways. We first consider the spike in concentrations following the 1997 peat fires by taking the difference in concentrations between 1998 (the first full year after the event) and an average from 2000 to 2004 (after concentrations had stabilized). The Jungfraujoch data set shows a decrease of 18 ppt (4%), while the models based on previous studies and our present emission ratios show decreases of 98 (21%) and 25 ppt (5%), respectively. To compare slopes while minimizing bias due to the choice of time period, we perform linear regressions on the annual average concentrations in each model time series for every combination of years starting and ending between 2000 and 2016, inclusive. In each case, we compute root-mean-square errors against the corresponding slopes of the NOAA global average and the Jungfraujoch data. On average, our model using the present emission ratios yields root-mean-square error values that are half as large as the model based on previous synthesis papers (reducing from 6.29 to 2.69 ppt OCS year⁻¹ compared to NOAA and from 7.10 to 3.66 ppt OCS year⁻¹ compared to Jungfraujoch).

4. Discussion

The large discrepancy between the two box model simulations, with our new emission factors giving a much closer match to observations, suggests that the anomalous peat observation is not representative of peat emissions generally and its inclusion could bias global biomass burning inventories. Our estimates show major reductions in OCS emissions from boreal forests (from an average of 45 to 9 Gg(S) year⁻¹), temperate forests (from 10 to 1 Gg(S) year⁻¹), and peatlands (from 104 to 7 Gg(S) year⁻¹) due to the exclusion of the aforementioned sample. The observational constraints on the interannual variability in OCS emissions from biomass burning also constrain the annual average emissions. Unfortunately, even in years with larger-than-average OCS emissions, our estimated biomass burning flux is too small to fully close the existing OCS budget gap, so other fluxes must be involved.

Several sources of uncertainty exist in these estimates. First, although the current work includes more studies than in previous synthesis papers (Akagi et al., 2011; Andreae, 2019; Andreae & Merlet, 2001), observational studies measuring OCS emissions from fires are still few, making it difficult to gauge representativeness of the observations. We quantify the uncertainty using the spread in reported emission factors/ratios for each category, both for OCS (presented here) and for reference species (CO and CO₂) presented in the GFED documentation and Akagi et al. (2011). For each category, we add relative uncertainties (standard deviation divided by mean) for both emission factors/ratios (CO/DM and OCS/CO) in quadrature then apply the result to the total OCS emissions for that category. Relative uncertainties are largest for temperate forest (105%) and tropical deforestation (82%) categories, followed by savannas (69%). On an absolute basis, savanna uncertainty dominates due to the larger overall contribution. Total uncertainty across all categories is added linearly in order to more conservatively account for correlation in the underlying burned matter data set.

A second source of uncertainty is the choice of reference species in the emission ratios. OCS emissions derived from CO₂ tend to more closely follow the distribution of burned dry matter, as CO₂ emission factors (ranging from 1489 to 1703 g CO₂/kg DM in the GFED4 documentation) are less variable than CO (63 to 210 g CO/kg DM) across the six biomass burning categories. Estimates derived from CO₂ yield higher total OCS emissions than those derived from CO (70 ± 37 Gg(S) year⁻¹ versus 60 ± 37 Gg(S) year⁻¹, respectively), owing primarily to larger emissions from savanna and grasslands. Additional results obtained using CO₂ are presented in supporting information (Table S4).

Other sources of uncertainty are more difficult to quantify. As noted, relatively few studies report OCS emission data, especially for peat and agriculture categories. Further, studies vary in instrumentation, calibration, and approaches to calculating emission factors or ratios. Additionally, emissions vary over the life of the fire and measurements may yield different values based on sampling location (near a fire, downwind, or in a lab). Despite these factors, the difference in the variety of samples used in our averages is significantly smaller than the effect of including or excluding the previously identified peat observation.

Our best estimate of annual-mean global OCS emissions from biomass burning (60 ± 37 Gg(S) year⁻¹) is broadly consistent with existing estimates, which have typically been computed using global, fixed emission factors. Chin and Davis (1993) estimate the global biomass burning flux as 74.7 (21.3–138.7) Gg(S) year⁻¹, based primarily on tropical fires. Nguyen et al. (1995) report global totals of 70 (30–110) Gg(S) year⁻¹, later cited in Watts (2000) and Kettle et al. (2002). Andreae and Merlet (2001) present a global total of 144 Gg(S)

Figure 4. Maps showing (a) spatial distribution of average (1997–2016) annual OCS open burning emissions based on biome-specific emission ratios (relative to CO) presented in this work and (b) difference between the biome-specific OCS estimate and an estimate based on total burned dry matter, scaled up to the same global total (i.e., without specific scaling between biomes).

year⁻¹. This value includes 65 Gg(S) year⁻¹ in emissions from biofuel and charcoal, categories which are not considered in our estimate; subtracting these yields 79 Gg(S) year⁻¹. Montzka et al. (2007) calculate an estimate of 65 Gg(S) year⁻¹, based on CO emissions from Andreae and Merlet (2001) and Duncan (2003) and emission ratios from Nguyen et al. (1995). Berry et al. (2013) chose a value from the high end of the published range (136 Gg(S) year⁻¹) to study the impact of emissions from different sources. Campbell et al. (2015) estimate OCS emissions from open burning (33–96 Gg(S) year⁻¹) and agriculture waste burning (8–18 Gg(S) year⁻¹), totaling 41–114 Gg(S) year⁻¹. Finally, applying the emission factors computed in a similar review by Andreae (2019) to the GFED data set used here yields an average of 86 Gg(S) year⁻¹.

A further benefit of this work is the ability to derive spatially and temporally resolved OCS emission distributions using data sets such as GFED that provide biome-specific biomass burning totals. Applying the average emission ratios from this work to existing CO totals from GFED yields global maps of OCS emissions, such as the annually averaged fluxes shown in Figure 4a. While the present work has primarily relied on burned area data from GFED, the emission ratios presented are generally applicable to other data sets, which may be higher-resolution or more suitable for regional applications. For example, applying the present emission ratios to average burned matter data from the Fire INventory from NCAR (FINN; see Wiedinmyer et al., 2011) yields global biomass burning emissions of 84 Gg(S) year⁻¹, primarily due to increased tropical emissions (though FINN does not explicitly account for peat). Future studies comparing emissions derived from these data sets to in situ measurements could further constrain emission magnitudes and distributions.

Additionally, variation in emission ratios across biomes yields different spatial patterns and categorical totals than estimates that scale all fuel types evenly. To demonstrate this, we scale the total burned dry matter up to the same global annual OCS totals calculated above, disregarding differences in emission factors between biomes. On average, our biome-specific estimate predicts savanna and grassland emissions that are 17 Gg(S) year⁻¹ less than the upscaled dry matter estimate (21 versus 39 Gg(S) year⁻¹, respectively) with corresponding increases in emissions in other categories, especially peat (increase of 5 Gg(S) year⁻¹) and agriculture (increase of 6 Gg(S) year⁻¹) (Figure 4b). The categorical and spatial differences between burned matter and OCS emissions highlight the need for an emissions database that accounts for different sources of biomass burning.

5. Conclusion

Biomass burning represents an important component in global OCS emissions, and estimates can vary significantly in space and magnitude depending on which emission data are used. Our analysis shows that while biomass burning emissions are highly variable from year to year, the total OCS emitted is, on average, not sufficient to close the existing OCS budget gap (60 ± 37 Gg(S) year⁻¹, compared to a deficit of

approximately 200–600 Gg(S) year^{−1}). Further, previous emission factor averages yield unrealistic OCS concentration trends when implemented in a global box model, whereas inventories derived in this work better match observations.

This work provides a spatially resolved inventory of OCS biomass burning fluxes that can be implemented in atmospheric transport and chemistry modeling studies. While uncertainties remain in our present estimates, new observation campaigns (WE-CAN, FIREX, etc.) are already underway, and it is our hope that this work will spur additional campaigns to measure OCS emissions from biomass burning in the future.

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