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Global gridded anthropogenic emissions inventory of carbonyl sulfide

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ABSTRACT

Atmospheric carbonyl sulfide (COS or OCS) is the most abundant sulfur containing gas in the troposphere and is an atmospheric tracer for the carbon cycle. Gridded inventories of global anthropogenic COS are used for interpreting global COS measurements. However, previous gridded anthropogenic data are a climatological estimate based on input data that is over three decades old and are not representative of current conditions. Here we develop a new gridded data set of global anthropogenic COS sources that includes more source sectors than previously available and uses the most current emissions factors and industry activity data as input. Additionally, the inventory is provided as annually varying estimates from years 1980–2012 and employs a source specific spatial scaling procedure. We estimate a global source in year 2012 of 406 Gg S y^{-1} (range of 223–586 Gg S y^{-1}), which is highly concentrated in China and is twice as large as the previous gridded inventory. Our large upward revision in the bottom-up estimate of the source is consistent with a recent top-down estimate based on air-monitoring and Antarctic firn data. Furthermore, our inventory time trends, including a decline in the 1990's and growth after the year 2000, are qualitatively consistent with trends in atmospheric data. Finally, similarities between the spatial distribution in this inventory and remote sensing data suggest that the anthropogenic source could potentially play a role in explaining a missing source in the global COS budget.

1. Introduction

Measurement based estimates of regional-scale carbon fluxes are needed to improve our understanding of carbon-climate feedbacks (Cox et al., 2000; Friedlingstein et al., 2006; Field et al., 2007). While measurements of CO_2 over terrestrial vegetation are useful for quantifying the net land-atmosphere exchange of carbon (Gurney et al., 2002), they are not useful for partitioning the underlying photosynthesis (gross primary production = GPP) and respiration fluxes at regional scales. An emerging approach for solving this problem, is the use of regional and global COS observations to estimate the underlying GPP sink (Montzka et al., 2007; Campbell et al., 2008, 2017; Hilton et al., 2017].

The dominant COS sink at regional scales is uptake by terrestrial vegetation in a process that is closely related to GPP (Sandoval-Soto et al., 2005; Campbell et al., 2008; Berry et al., 2013). The COS plant sink is largely controlled by stomatal conductance which in turn is closely related to GPP, particularly for scales of analysis that are integrated in both space (regional to global) and time (monthly to annual) (Campbell et al., 2017; Hilton et al., 2017). While other ecosystem sources and sinks have been observed (Commane et al., 2015; Whelan et al., 2016), the COS plant sink is dominant at regional scales (Campbell et al., 2008). Existing observations of COS concentrations, from NOAA airborne and tower monitoring networks, as well as remote sensing platforms, can be utilized to drive the regional COS tracer approach (Carmichael, 2003; Montzka et al., 2007; Kuai et al., 2014, 2015; Glatthor et al., 2015; Wang et al., 2016).

The major source of atmospheric COS is derived from oceans (Berry et al., 2013; Kuai et al., 2014; Launois et al., 2014; Glatthor et al., 2015). The spatial separation of this source from the dominant sink of COS by terrestrial vegetation allows for continental regions to be well suited for the COS tracer approach. However, other continental sources and sinks of COS exist at certain locations and times. These other fluxes must be quantified for the COS tracer approach to be applied with confidence. Of these other fluxes that must be quantified, the largest continental source is emissions from anthropogenic activities (Campbell et al., 2015).

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The current gridded anthropogenic COS inventory suffers from multiple limitations that weaken its suitability for use in the COS tracer approach. This approach involves the use of gridded inventories as input to 3-D atmospheric transport models to constrain the influence of anthropogenic activities on COS observations (Berry et al., 2013). The only currently available global gridded anthropogenic inventory is from Kettle et al. (2002). However, recent work has demonstrated that the Kettle inventory underestimates the magnitude of this source, has a biased spatial distribution, and ignores large inter-annual variation in the source (Campbell et al., 2015). These limitations of the Kettle inventory are due to multiple factors including the use of industry activity data from over three decades ago, integration of a small subset of the available emission factor data, and spatial scaling by global SO_2 emissions rather than industry activity data. More recent inventories of global anthropogenic COS sources have corrected for many of these limitations (Campbell et al., 2015; Lee and Brimblecombe, 2016), but have not been extrapolated to spatially-explicit data sets that are the required input for atmospheric transport models. Additionally, high-resolution regional gridded data have been developed (Blake et al., 2004; Zumkehr et al., 2017), but these inventories do not provide temporally-explicit estimates and they are limited to the U.S. and Asia.

In addition to using gridded anthropogenic inventories to infer regional GPP, these inventories may also help resolve a missing source in the global COS budget. Comparisons of global atmospheric transport simulations and COS observations have identified a large missing source of atmospheric COS that is 230–800 Gg S yr^{-1}, and has generally been attributed to a missing tropical oceanic source (Suntharalingam et al., 2008; Berry et al., 2013; Glatthor et al., 2015; Kuai et al., 2015; Lennartz et al., 2016). However, ocean cruise data show under-saturation of ocean surface waters with respect to COS and low global annual direct emissions of COS from oceans, suggesting that the oceans alone may not be able to account for the missing COS source (Lennartz et al., 2016). While additional ocean cruise data are needed, revisiting the global simulation studies with updated anthropogenic inventories would be useful for testing the hypothesis that anthropogenic activities could contribute to the missing COS source.

Given the limitations of the Kettle gridded inventory and the recent advances in understanding of the anthropogenic source (Campbell et al., 2015; Lee and Brimblecombe, 2016), we have developed a new global gridded inventory of the primary emission sectors. The inventory has a 0.1° spatial resolution and uses the most current emission factors and industry activity data as input. The inventory is provided as annually varying estimates from years 1980–2012 and employs a source specific spatial scaling procedure.

2. Methods

The inventory developed here includes direct and indirect anthropogenic sources. Indirect sources of COS result from anthropogenic carbon disulfide (CS_2) emissions which are rapidly oxidized to COS in the atmosphere (Watts, 2000; Wang et al., 2001; Kettle et al., 2002). The atmospheric oxidation of CS_2 to COS has been estimated to have a molar conversion rate of 87% (Barnes et al., 1994). Source estimates reported in this study are the sum of direct and indirect sources. Uncertainty estimates are based on the range of emission factor data within each sector as noted below. For sectors with only a single reported emission factor, we apply an uncertainty of 50% as in Watts (2000).

The anthropogenic sources of COS considered in this study are: rayon production, aluminum smelting, carbon black production, industrial and residential coal consumption, agricultural chemicals, pulp & paper industries, industrial solvent applications, titanium dioxide production and tire wear (Watts, 2000; Campbell et al., 2015; Du et al., 2016; Lee and Brimblecombe, 2016). Industrial coal consumption includes manufacturing and electricity production. We did not include biomass burning (e.g. open burning, agriculture waste, biofuel) but we note that the most recent synthesis of source estimates and emission factors for biomass burning is reported by Campbell et al. (2015). Furthermore, soil emissions from managed lands are not included here as they are typically modeled separately using soil biogeochemistry models that account for concurrent soil sinks and sources (Whelan et al., 2016; Hilton et al., 2017).

We estimate emissions for the years 1980 through 2012 due to the limited availability of concurrent data for COS sources. Data past 2012 exists for some sources, but these data were not incorporated into our result for simplicity in presenting the dataset.

For each anthropogenic source, four parameters are required to estimate the magnitude of the source and then to distribute the source in space and time: industry activity, emissions factor, sub-country spatial scaling, and temporal scaling. Often the industry activity data set also provides the information for country-level spatial and temporal scaling. Sub-country spatial scaling is performed by distributing country-level estimates to a grid based on a gridded proxy flux. The IPCC code associated with the gridded proxy flux is used as the criteria for identifying the flux that most closely describes the specific anthropogenic estimate that needs to be distributed. Table 1 summarizes the input data used in the creation of each of the anthropogenic sources of COS calculated in this study. The following sections describe the specific methods and data used to create the estimates of direct or indirect emissions of anthropogenic COS for each source sector.

2.1. Coal

Coal combustion is traditionally considered the largest single direct industrial source of COS (Watts, 2000; Blake et al., 2008). We consider two classes of coal consumption in this study: industrial coal consumption and residential coal consumption for cooking and heating. Residential coal has not been included in previous global inventories, but recent work suggests potentially large emissions of COS from residential coal consumption in China (Du et al., 2016).

2.2. Industrial coal

Country-level industry activity data of industrial coal consumption is readily available (Smith et al., 2011). While stack and plume observations for COS emission factors from coal power plants in the United States have been made (Khalil and Rasmussen, 1984; Blake et al., 2008), varying combustion efficiencies, emissions controls, and the sulfur content of coal in other countries suggests that the U.S. emission factors may not be appropriate to apply globally without modification. To address this limitation, previous work has used the ratio of SO_2 emissions to coal consumption for each country to scale emissions factors (Campbell et al., 2015). This correction for estimating emissions factors is adopted in this study. For sub-country spatial scaling, industrial SO_2 emissions are used to proportionally distribute the estimated country-level COS estimates (Joint Research Centre, 2011). Uncertainty for our OCS source estimate from industrial coal combustion is derived from the uncertainty in the emissions factor (± 18%).

2.3. Residential coal

Emission factors for residential coal consumption have been made in laboratory coal stove experiments and household air-sampling in China (Du et al., 2016). The observed emission factors were 0.57 ± 0.10 and 1.43 ± 0.32 g COS emitted per kg of coal for the laboratory and farmhouse studies, respectively (approximately 50 times larger than power plant emission factors), which encompass the range of uncertainty in our estimate for COS from this source (Du et al., 2016). Here, an average of the laboratory and farmhouse emission factors is assumed for the baseline scenario and the range of the two emission factors is the estimated uncertainty. Country-level residential