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Contribution of aboveground plants, the rhizosphere and root-free-soils to total COS and DMS fluxes at three key growth stages in rice paddies

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ABSTRACT

Carbonyl sulfide (COS) and dimethyl sulfide (DMS) are trace sulfur gases that contribute substantially to sulfate aerosols or cloud condensation nuclei in the upper and lower atmosphere and therefore play great roles in the earth's radiative balance. COS and DMS fluxes in rice (*Oryza sativa* L.) have been measured previously in rice paddies, and this study was designed to further investigate the role of aboveground plants, the rhizosphere and the root-free-soils in the exchange of COS and DMS by comparing COS and DMS fluxes among planted plots (PP), non-planted plots (NPP), plots with aboveground plants just cut before test (plants-cut plots, PCP) at three key growth stages (tillering, jointing–booting and mature stage). The average COS fluxes in NPP, PCP, and PP were 10.5 ± 3.8, 6.1 ± 4.6, and -12.3 ± 5.0 pmol m^{-2} s^{-1}, suggesting that COS emission from root-free-soils (10.5 pmol m^{-2} s^{-1}) was surpassed by uptake of the rhizosphere (-4.4 pmol m^{-2} s^{-1}) and aboveground plants (-18.4 pmol m^{-2} s^{-1}). DMS emission rates were 60.5 ± 25.2, 20.2 ± 7.3 and 30.8 ± 10.8 pmol m^{-2} s^{-1} in PP, NPP and PCP, respectively. Calculated DMS emission from the aboveground plants, rhizosphere and the root-free-soils were 29.7, 10.6 and 20.2 pmol m^{-2} s^{-1}, accounting respectively for 49.0%, 17.6% and 33.4% of total DMS emission. Directly measured COS and DMS fluxes from aboveground plants averaged -14.2 ± 7.2 and 22.3 ± 10.6 pmol m^{-2} s^{-1}, approximating that of -18.4 and 29.7 pmol m^{-2} s^{-1} calculated by subtracting fluxes in PP and PCP, respectively. Preliminary diurnal flux observation at jointing–booting stage revealed significant linear correlation between COS uptake rates by above ground plants (MF_{COS}) and photosynthesis rates (Pn) with a MF_{COS}/Pn slope of 0.48 pmol μmol^{-1}. DMS emission from the aboveground plants was also found to be significantly increased with temperature.

1. Introduction

Carbonyl sulfide (COS), the most abundant volatile organic sulfur compound (VOSCs) in the atmosphere with an average global mixing ratio of about 500 ppt, is relatively inert with an atmospheric lifetime up to 2–6 years (Ulshöfer and Andreae, 1997). It can therefore transport and reach the stratosphere where it is oxidized, contributing to the stratospheric sulfate aerosols (SSA) that play a significant role in the radiative balance of the atmosphere (Charlson et al., 1987) and ozone depletion (Fahey et al., 1993; Solomon et al., 1996). SSA by COS oxidation has been reported to be the main source of stratospheric sulfate during non-volcanic periods (Engel and Schmidt, 1994), accounting for about 43% of the total background SSA (Pitari et al., 2002). DMS is the major VOSCs species that contributes to biogenic sulfate and is the principal precursor of sulfate aerosol and cloud condensation nuclei (CCN) in the troposphere, particularly over the seas and oceans, and hence it is also a very crucial trace gas that influences the earth's radiative balance (Charlson et al., 1987; Andreae and Crutzen, 1997; Andreae et al., 2001; Kesselmeier and Hubert, 2002). Due to the importance of these VOSCs in the chemistry of upper and lower atmosphere, as well as in climate change, their sources and sinks have long been vital issues in understanding their biogeochemical cycling (Andreae and Crutzen, 1997).

COS has both natural and anthropogenic sources (Chin and Davis, 1993; Weiss et al., 1995; Ulshöfer and Andreae, 1997; Watts, 2000; Wu et al., 2010), and its major sinks include vegetation (Kesselmeier and Merck, 1993; Protoschill-Krebs et al., 1995, 1996; Kuhn et al., 1999; Yonemura et al., 2005; Geng and Mu, 2006;

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Stimler et al., 2010) and soils (De Mello and Hines, 1994; Geng and Mu, 2004; Yi et al., 2007, 2008; Liu et al., 2010). Large source-sink imbalance was once a big problem for COS global budgets (Chin and Davis, 1993; Watts, 2000). Much more balanced budgets with similar magnitude of sinks and sources (Watts, 2000) came with a revision of the net influence of open and coastal oceans (Weiss et al., 1995; Ulshöfer and Andreae, 1997; Uher, 2006), and in particular with the new understanding of soil as a net sink for COS and a revised estimate of the COS sink by vegetation (Kesselmeier et al., 1999; Kuhn et al., 1999; Geng and Mu, 2004; Steinbacher et al., 2004). For DMS, quite a lot of flux measurements have been conducted on the oceans/coastal regions (Kettle et al., 2001; Huebert et al., 2004; Uher, 2006) and in terrestrial ecosystems (Staubes et al., 1989; Kanda et al., 1992; Yang et al., 1998; Geng and Mu, 2004, 2006; Yonemura et al., 2005; Yi et al., 2008; Yi and Wang, 2011). Since COS and DMS fluxes varied significantly in different ecosystems, or even in different compartments of the same ecosystem, uncertainties in COS and DMS fluxes in different ecosystems will be further narrowed with extensive field flux measurements and the understanding of roles played by different ecosystem compartments.

Rice (*Oryza sativa* L.) is China's most important crop, grown on about 25% of the arable land. As an inland wetland ecosystem, rice paddy can absorb or emit VOSCs (Kanda et al., 1992; Yang et al., 1996, 1998; Nouchi et al., 1997; Redeker et al., 2003; Yi et al., 2008). In our previous study (Yi et al., 2008), we only measured COS and DMS fluxes at different growth stages of rice, and found elevated COS and DMS fluxes from rice cultivation, particularly at the jointing–booting stage. We also found that plots with rice plants acted as COS sink and non-planted plots as COS source, while DMS emission rates at plots with rice plants were significantly higher than those at non-planted plots. The results in our previous study suggested that rice plants (including aboveground plants and the underground rhizosphere) played great roles in COS and DMS fluxes. However, the roles of the aboveground plants and the underground rhizosphere remain unidentified. In the present study the fluxes were measured and compared among planted plots (PP), non-planted plots (NPP) and plants-cut plots (planted plots with plants cut just before flux measurement, PCP) at three key growth stages. The purpose was to further clarify the roles of the aboveground plants, the underground rhizosphere and the root-free-soils in the exchange of COS and DMS between the rice paddy and the atmosphere. The influence of photosynthesis rate (Pn), photosynthetically active radiation (PAR) and stomatal conductance (Gs) on their fluxes from aboveground rice plants were also investigated.

2. Materials and methods

2.1. Site description

Field measurements were performed in a rice paddy field (23°10′37″N, 113°20′10″E) located in the low-subtropical monsoon climate region. Soil properties at the experiment sites were listed in Table 1. The soil was waterlogged during our experiment periods except that the field was allowed to drain and remain dry for 5 days from 15th May. The fertilizer application was as follow: urea (150 kg ha^{-1}), calcium superphosphate (750 kg ha^{-1}) and potassium chloride (150 kg ha^{-1}) as base fertilizer before transplant, then urea (150 kg ha^{-1}), zinc sulfate (15 kg ha^{-1}) as tillering fertilizer 10 days after transplant, urea (90 kg ha^{-1}), potassium chloride (750 kg ha^{-1}) as flower-promoting fertilizer 35 days after transplant, and urea (90 kg ha^{-1}), potassium chloride (750 kg ha^{-1}) as flower-maintaining fertilizer 20 days before heading.

2.2. Field experiments

Three treatments were set up in rice paddies to measure COS and DMS fluxes: one was planted with rice plants under natural condition (planted plot or PP, Fig. 1A), the second was treated under the same condition except that the field had no rice plants (non-planted plot or NPP, Fig. 1C), and the third was treated the same as the planted plot except that aboveground plants were cut just before measuring the fluxes in this plot (plant-cut plot or PCP, Fig. 1B) at the same time when measuring fluxes in other plots. All the treatment plots were adjacent for the comparability, and each treatment plot had three replicates. Three campaigns were performed at tillering stage on 10th May, jointing–booting stage on 18th June, and mature stage on 6th July, respectively, and three measurements were conducted between 10:00 and 16:00 at each campaign.

With the measured fluxes of MF_{PP}, MF_{NPP} and MF_{PCP} respectively in PP, NPP and PCP, calculated fluxes of CF_{rfs}, CF_{ur} and CF_{ap} due respectively to the root-free-soils, the underground rhizosphere and aboveground plants can be roughly obtained as below:

$$CF_{rfs} = MF_{NPP} \quad (1)$$

$$CF_{ur} = MF_{PCP} - MF_{NPP} \quad (2)$$

$$CF_{ap} = MF_{PP} - MF_{PCP} \quad (3)$$

We can see that the sum of CF_{rfs}, CF_{ur} and CF_{ap} is MF_{PP}. COS and DMS fluxes from the aboveground plants were also measured directly with an enclosure method (Fig. 1D). Theoretically speaking, this directly measured flux from aboveground plants, MF_{ap}, should approximate the CF_{ap}, and we have:

$$CF_{ap} \approx MF_{ap} \quad (4)$$

MF_{PP}, MF_{NPP} and MF_{PCP} were measured by the static chamber method as reported in our previous study (Yi et al., 2008). Briefly, the chambers were 100 cm above the soil and each covered an area of 0.36 m^2 (60 cm × 60 cm). This area corresponded to about 9 stumps of rice plants and the distance between rice plants was about 20 cm. Air samples were collected 0, 5, 10 and 20 min after the chamber was covered on the soil. Fluxes from the aboveground plants (MF_{ap}) were measured using a small chamber (100 cm in height × 20 cm in length × 20 cm in width) to contain the above-water fraction of a stump of rice plants as illustrated in Fig. 1D. A fan and thermocouple (TES Electrical Electronic Corp., Taipei, Taiwan) were installed to mix the air inside and to measure the temperature inside the chamber. Four samples (about 500 ml each sample) were collected into 1 L Tedlar sampling bags (SKC Inc., USA) at 0, 5, 10 and 20 min after closing the chamber.

To find out the factors influencing COS and DMS fluxes from the aboveground plants, diurnal variation of the fluxes from the aboveground plants was measured at the jointing–booting stage on 18th June, and Pn, PAR and Gs were measured with Li-6400 (Li-cor Inc., USA) synchronously.

2.3. Laboratory analysis

COS and DMS in air samples were analyzed using an Entech 7100 Preconcentrator (Entech Instruments Inc., CA, USA) coupled to an Agilent 5973N gas chromatography–mass selective detector (GC–MSD, Agilent Technologies, USA). The m/z 60 and 62 was set as the target ion for COS and DMS. Detailed description of the method could be found in our previous studies (Yi et al., 2007, 2008).