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Carbonyl sulfide, dimethyl sulfide and carbon disulfide in the Pearl River Delta of southern China: Impact of anthropogenic and biogenic sources

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ABSTRACT

Reduced sulfur compounds (RSCs) such as carbonyl sulfide (OCS), dimethyl sulfide (DMS) and carbon disulfide (CS_2) impact radiative forcing, ozone depletion, and acid rain. Although Asia is a large source of these compounds, until now a long-term study of their emission patterns has not been carried out. Here we analyze 16 months of RSC data measured at a polluted rural/coastal site in the greater Pearl River Delta (PRD) of southern China. A total of 188 canister air samples were collected from August 2001 to December 2002. The OCS and CS_2 mixing ratios within these samples were higher in autumn/winter and lower in summer due to the influence of Asian monsoon circulations. Comparatively low DMS values observed in this coastal region suggest a relatively low biological productivity during summer months. The springtime OCS levels in the study region (574 ± 40 pptv) were 25% higher than those on other East Asia coasts such Japan, whereas the springtime CS_2 and DMS mixing ratios in the PRD (47 ± 38 pptv and 22 ± 5 pptv, respectively) were 3–30 times lower than elevated values that have been measured elsewhere in East Asia (Japan and Korea) at this time of year. Poor correlations were found among the three RSCs in the whole group of 188 samples, suggesting their complex and variable sources in the region. By means of backward Lagrangian particle release simulations, air samples originating from the inner PRD, urban Hong Kong and South China Sea were identified. The mean mixing ratio of OCS in the inner PRD was significantly higher than that in Hong Kong urban air and South China Sea marine air ($p < 0.001$), whereas no statistical differences were found for DMS and CS_2 among the three regions ($p > 0.05$). Using a linear regression method based on correlations with the urban tracer CO, the estimated OCS emission in inner PRD (49.6 ± 4.7 Gg yr^{-1}) was much higher than that in Hong Kong (0.32 ± 0.05 Gg yr^{-1}), whereas the estimated CS_2 and DMS emissions in the study region accounted for a very few percentage of the total CS_2 and DMS emission in China. These findings lay the foundation for better understanding sulfur chemistry in the greater PRD region of southern China.

1. Introduction

The chemistry of reduced sulfur compounds (RSC) in the atmosphere has significant implications for global climate change, ozone layer depletion and the acidity of precipitation (Turco et al., 1980; Rodriguez et al., 1991; Solomon et al., 1996; Andreae and Crutzen, 1997). Carbonyl sulfide (OCS), dimethyl sulfide (DMS) and carbon disulfide (CS_2) are the main RSCs found in the marine and continental atmosphere (Chin and Davis, 1993; Kettle et al., 2002; Montzka et al., 2007). These RSCs are ultimately oxidized via sulfur dioxide to sulfuric acid in the troposphere and/or stratosphere (Johnson and Bates, 1993; Andreae and Crutzen, 1997). The formed sulfate particles alter the radiative properties of the Earth's atmosphere, potentially leading to the climate change, particularly due to augmented RSC emissions from anthropogenic sources (Seinfeld and Pandis, 2006). Further, these particles can affect the environment and ecosystem by acidification of precipitation (Andreae and Crutzen, 1997); and provide surfaces for heterogeneous reactions related to ozone destruction processes by chlorine and nitrogen radical chemistry (Rodriguez et al., 1991; Andreae and Crutzen, 1997). The atmospheric sources of RSCs include natural and biogenic emissions such as oceans (Leck and

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Persson, 1996; Kettle et al., 2002; Aranami and Tsunogai, 2004; Lee et al., 2010), volcanoes (Vasilakos et al., 2005), soil and marshes (Steinbacher et al., 2004; Liu et al., 2010), and vegetation (Geng and Mu, 2006; White et al., 2010); and anthropogenic emissions i.e. biomass burning (Meinardi et al., 2003), coal combustion (Chin and Davis, 1993), automobiles (Fried et al., 1992), aluminum production (Harnisch et al., 1995), and sulfur recovery (Chin and Davis, 1993).

OCS is the most abundant and stable RSC present in the troposphere and lower stratosphere due to its long lifetime (~ 16 years). Therefore OCS is the most likely source of stratospheric background sulfate aerosols due to the photochemical dissociation of OCS in the stratosphere to SO_2. Globally, about 14–32% of the atmospheric OCS is attributed to human activities (Chin and Davis, 1993; Kettle et al., 2002). It is reported that 20–39% of OCS is removed from the stratosphere and troposphere through the reaction with hydroxyl radicals (OH) whereas 52–71% is due to vegetation uptake (Chin and Davis, 1993; Watts, 2000; Kettle et al., 2002). In addition, the possible use of OCS to track gross plant CO_2 exchange increases the importance of understanding OCS sources and sinks (Stimler et al., 2010).

The major source of CS_2 (lifetime: ˜2–3 days) is chemical industrial emissions (52–58%), followed by the ocean contribution (27–34%). The rest is from soil and marsh, volcano and sulfur recovery (8–21%) (Chin and Davis, 1993; Watts, 2000). The atmospheric reaction of CS_2 with OH radicals is the dominant sink for CS_2 in the atmosphere as well as a source of OCS (Seinfeld and Pandis, 2006). Atmospheric CS_2 is therefore important in determining the stratospheric sulfur balance during non-volcanic periods.

DMS (lifetime: ~ 1 day) is mainly released from the oceans (~ 85–98% of total emissions) with about 2–15% from other sources i.e. vegetation, tropical forests, soil and wetlands (Watts, 2000; Gondwe et al., 2003). DMS is also emitted from bush fires (Meinardi et al., 2003) but the contribution of anthropogenic sources to ambient DMS levels is negligible (~ 0.5%) (Schäfer et al., 2010). The major fate of DMS is reaction with OH radicals during daytime and NO_3 radicals during night-time (Turnipseed and Ravishankara, 1993; Boucher et al., 2003).

In the past decades, the Asian continent has been recognized as a major source region of chemically active and radiatively important trace gases and aerosols, due to rapid urbanization and industrialization (Akimoto and Narita, 1994; Thornton et al., 1996; Streets et al., 2003; Blake et al., 2004). Both aircraft-borne and ground level measurements i.e. PEM-West A, TRACE-P and the Jeju Island study, have sought to understand the relative significance of biogenic and anthropogenic emissions for the local or regional sulfur cycles (Thornton et al., 1996; Kim et al., 2004; Blake et al., 2004; Inomata et al., 2006). These studies have provided valuable information about the sulfur chemistry in the Asian continent. However, each of these studies only covered limited time periods, usually 1–2 months during spring. In particular, the impact of biogenic and anthropogenic emissions on the sulfur budgets in China's premiere industrial region – the Pearl River Delta (PRD) area of southern China – remains unclear.

A 16-month sampling campaign was carried out at a strategically located rural/coastal site in the PRD from August 2001 to December 2002, in order to understand the physical and chemical properties of air masses in this region. Using this dataset, the causes of a multi-day ozone episode were analyzed (Wang and Kwok, 2003), the temporal variability and emission patterns of pollution plumes during October–December 2001 were characterized (Wang et al., 2003), and the seasonal profiles and atmospheric processes of trace gases (including NMHCs) were overviewed (Wang et al., 2005). C_1–C_5 alkyl-nitrates and their relation to their parent NMHCs and to ozone were also examined (Simpson et al., 2006), and the regional and local source contributions to NMHCs were quantified using a PCA/APCS receptor model (Guo et al., 2006) and by chemical ratios (Zhang et al., 2008). In addition, the source origins, profiles and apportionments of halocarbons in this region were studied (Guo et al., 2009a). In this study, our focus is the RSC data. The spatiotemporal variations of RSCs will be presented, and the source emissions of these RSCs will be estimated.

2. Experimental method

2.1. Sampling site

The sampling site, Tai O, was located in a rural/coastal area on the western coast of Lantau Island in southwest Hong Kong (22.25°N, 113.85°E), bounded to the south by the South China Sea and to the north by the rapidly developing greater PRD region (Fig. 1). The greater PRD is a home to around 40 million inhabitants and is also the most important industrial base in southern China. Meteorological conditions over this region and a large part of Asia are strongly influenced by Asian monsoon circulations. During the winter, prevailing winds are northerly and northeasterly. By contrast, southerly and southwesterly winds are predominant in the summer. Such meteorological conditions make the coastal region of southern China an ideal place to measure the chemical composition of polluted continental outflow in the winter and clean South China Sea air during summer. A detailed description of the site is provided in Wang et al. (2003). In brief, Tai O site is about 32 km to the west of the Hong Kong urban center and 32 km to the east of Macau. The three largest population centers in the PRD lie to the north of Tai O: Guangzhou (10 million), Hong Kong (6.9 million) and Shenzhen (4 million). Power plants, airports and seaports are mainly located along the two sides of the Pearl Estuary.

The study site was located on a hill 80 m above sea level, overlooking the Pearl Estuary to the west and north, the South China Sea to the south, and the Hong Kong urban center to the east. Local anthropogenic emissions are very small due to a sparse population and light traffic at Tai O. Many deciduous trees, sources of biogenic trace gases, immediately surround the site.

2.2. Sampling and chemical analysis

Whole air samples were collected into evacuated 2-L electropolished stainless steel canisters and pressurized to about 20 psi using a metal bellows pump. The canisters were cleaned and evacuated at the University of California, Irvine (UCI) before being shipped to Tai O. Details of the preparation and pre-conditioning of the canisters are described in Blake et al. (1994). Whenever a canister sample was taken, the canister valve was slightly opened, allowing about 1 min for the collection of the integrated whole air samples. The canisters were then shipped to UCI for chemical analysis. The samples were analyzed within two weeks. A 6-column multiple GC–MS system was used to identify and quantify volatile organic compounds. The combination that was used to quantify the sulfur gases was a DB-5ms (J & W Scientific) column (60 m; i.d., 0.25 mm; film, 0.5 μm) coupled to an HP-5973 quadrupole Mass Spectrometric Detector (MSD). The MSD was placed in the single ion monitoring (SIM) mode, choosing the most abundant ion of each compound without interference. The stability of these sulfur gases in the stainless steel canisters was tested for more than one week and no statistical difference was found between the first and last samples. The measurement precision was 5% and the measurement accuracy was 1–10% for these RSCs. The detection limits were 0.5 pptv for CS_2, 1 pptv for DMS, and 20 pptv for OCS. Detailed descriptions of the chemical analysis and relevant quality assurance/quality control for each species are given by Colman et al. (2001) and Blake et al. (2004).

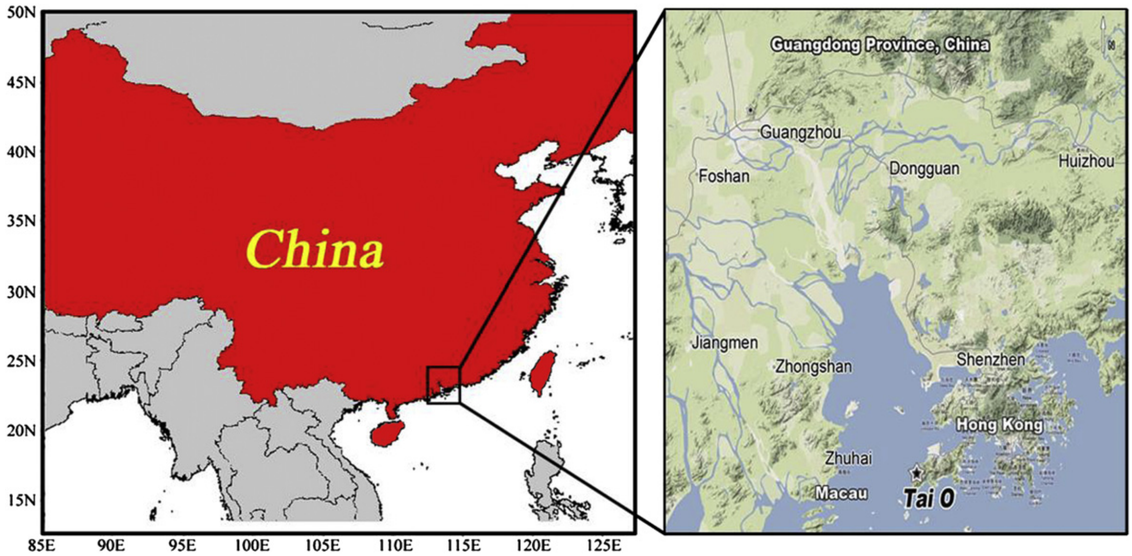

Fig. 1. Map showing the study site and Pearl River Delta of China (right). The greater PRD region mainly consists of nine cities within Guangdong Province, i.e. Guangzhou, Shenzhen, Zhuhai, Dongguan, Zhongshan, Foshan, Jiangmen, Huizhou and Zhaoqing, plus Hong Kong and Macau Special Administrative Regions.

Between August 2001 and December 2002, a total of 188 canister samples were taken. In general, at least one sample per week was taken during the sampling period to avoid sampling bias in the dataset. In addition, more samples were acquired between August and December to better understand the O_3 pollution in the region (Wang et al., 2003, 2005). All samples were collected in the daytime, when factories were under normal operation, with 75% of the samples taken in the afternoon and 25% in the mid-morning.

Additionally, O_3, CO, SO_2, NO and NO_y were continuously measured during the study period. Several meteorological parameters were simultaneously monitored at Tai O, including wind speed and direction, temperature, relative humidity, total ultraviolet radiation (320–400 nm), and global solar radiation.

2.3. Lagrangian particle release simulation and air masses classification

To help understand the air mass history, we conducted backward Lagrangian particle release simulations for all samples using the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT4, Version 4.8), which was developed by the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory (http://www.arl.noaa.gov/ready/hysplit4.html). The model was run in a 7-day backward mode with a total of 2000 particles released at 200 m above the site. For the calculation of particle dispersion, a 3-D method was adopted. The meteorological data that drove the model were GDAS dataset (3 hourly, global and 1° in longitude and latitude, and 23 pressure levels) (see http://www.arl.noaa.gov/ss/transport/gdas1.html for details). A detailed description of the simulation is provided in Guo et al. (2009a).

Different from the traditional trajectory method, the backward particle release simulation – including the dispersion processes related to turbulence mixing – provides more accurate information about the history of air masses. Essentially, a large number of particles is released at the receptor and transported backward in time. Then the residence time of all particles, normalized by the total number of released particles, is determined on a uniform grid. The residence time in a particular grid cell is proportional to the contribution a source with unit strength in this cell would make to the mixing ratio at the receptor. By multiplying the residence time with a tracer's actual source strength (in ppb s^{-1}) in the respective grid cell (assuming instantaneous mixing of the emitted tracer within that cell), we obtain the actual contribution to the mixing ratio at the receptor (in ppb) from this grid cell. Summing up the contributions from all grid cells finally gives the total mixing ratio at the receptor. Here we concentrate on a shallow layer adjacent to the ground i.e. 100 m. The lowest 100 m thickness is considered as the "footprint" retroplume layer, which is easily affected by surface emissions (Stohl et al., 2003; Ding et al., 2009; Zhang et al., 2009). With the sum of footprint air mass concentrations, all 188 samples are classified into four major categories: Hong Kong local air; inner PRD (consists of nine cities within Guangdong Province, see Fig. 1) – air masses from the PRD except Hong Kong and Macau; regional air from continental China, which is not discussed here; and marine air masses from the South China Sea or the Pacific. For the inner PRD and Hong Kong categories, we summed the air mass concentrations within each geographical area and identified each individual sample with a criterion that the total concentration was higher than the mean plus one standard deviation. Using this method, 27 and 23 samples were classified into Categories Hong Kong and inner PRD. Since previous studies showed that the air masses arriving in Hong Kong mostly come from the eastern and southeast China mainland (e.g. Wang et al., 2005; Guo et al., 2009b), we identified these air masses with a criterion that the total land residence time was higher than the median value, i.e. the highest 50% (95 samples) were selected. It should be noted that using these methods, some samples were duplicately classified into the three categories. The total number for the three categories of air masses with anthropogenic influences was 112. For the marine air category, we selected 19 samples (i.e. about 10% of total samples) based on the value of summed marine residence time. The residual 57 samples, which were relatively well-mixed with various air masses, were not included in the statistics in this paper.

3. Results and discussion

3.1. General characteristics of OCS, DMS and CS_2 in the greater PRD

3.1.1. Monthly variations

Fig. 2 illustrates the seasonal variations of the RSCs measured at Tai O for the period of September 2001–December 2002. It is the first time that the seasonal profiles of OCS, DMS and CS_2 are

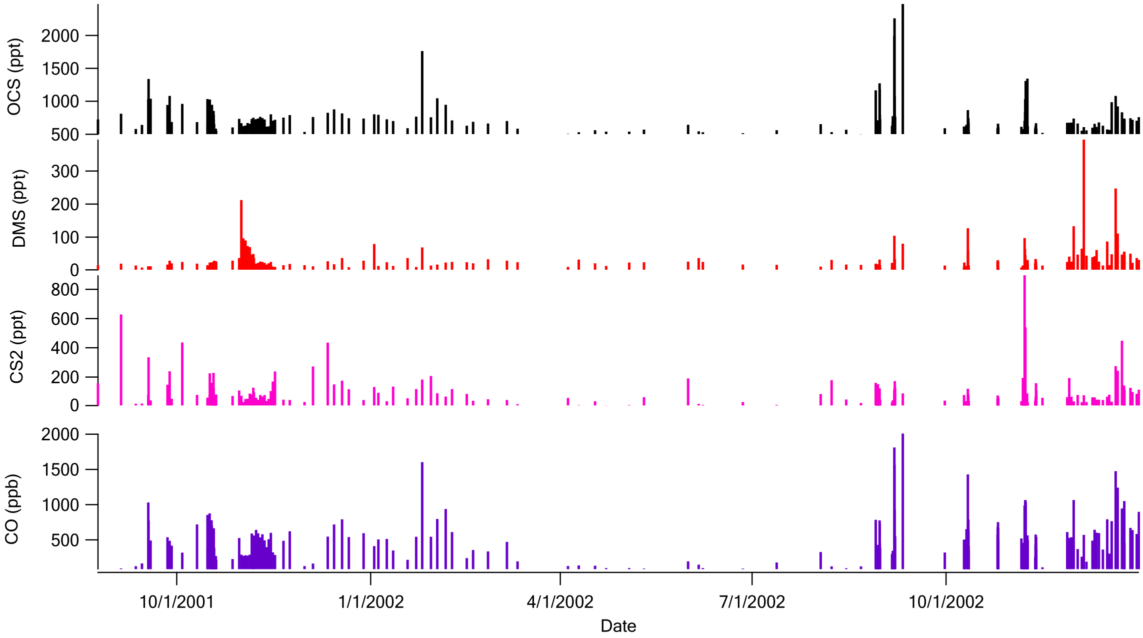

Fig. 2. Time series of observations of OCS, DMS, CS_2 and CO at Tai O from August 2001 to December 2002. The *X*-axis is date given in mm/dd/yyyy format. The *Y*-axis is mixing ratio in pptv, except for CO in ppbv. A total of 188 whole air samples were collected.

reported in the atmosphere of southern China. All three RSCs showed a strong seasonal dependence, with summer minima and winter maxima. The wintertime enhancements typically appeared as discrete episodes during which mixing ratios of all three compounds were enhanced together with the combustion tracer carbon monoxide (CO).

The OCS mixing ratios had a distinct seasonality, with maximum values (up to 2480 pptv) in autumn/winter and minimum values (505 pptv) in summer. This is different from the results obtained by Bandy et al. (1992) who examined a time series of OCS measurements between 1977 and 1991 in the northern hemisphere over a longitude range of 52° E–155° W and a latitude range of 10° N–85° N and found no seasonal variations. Griffith et al. (1998) and Deutscher et al. (2006) found a large peak-to-peak seasonal difference in 1996–1997 in the southern hemisphere, but in their case with a late summer peak and winter trough, due to a warm coastal ocean source. Furthermore, the monthly summertime OCS values (530 ± 17 pptv in June and 584 ± 61 pptv in May) observed in this study are consistent with the average northern hemisphere value (523 ± 63 pptv), confirming that background OCS is fairly evenly distributed throughout the northern hemisphere (Warneck, 2000). On the other hand, in this study the elevated levels found in autumn and winter suggest the presence of anthropogenic sources of OCS in the study region. In order to understand the possible sources of OCS in this region, correlation analyses of OCS with CO (combustion tracer), CH_3Cl (biomass/biofuel burning marker), CH_3I (oceanic tracer) and C_2Cl_4 (urban emission marker) were carried out for the whole 188 samples. A moderate correlation was found between OCS and CO ($R^2 = 0.57$), OCS and CH_3I ($R^2 = 0.55$), suggesting OCS emissions from both combustion sources and the ocean. Though poor correlation was observed between OCS and CH_3Cl ($R^2 = 0.15$) for the 188 samples, the scatter plot clearly showed two groups of data points and one group had moderate correlation ($R^2 = 0.63$), indicating part of the OCS was from biomass/biofuel burning in this region. Furthermore, poor correlation between OCS and C_2Cl_4, which is a purely anthropogenic compound associated with solvent use and dry cleaning (Simpson et al., 2004), implied that OCS was not emitted from urban sources.

Like OCS, CS_2 exhibited a maximum in winter (199 pptv in December 2001) and a minimum in summer (10 pptv in July 2002). The mixing ratio ranges observed in this coastal region are consistent with the CS_2 values in air influenced by anthropogenic activities and biomass burning (11–339 pptv), but are much higher than background free tropospheric and predominantly maritime measurements (2–6 pptv) (Bandy et al., 1993). Indeed, our previous studies found that predominant northeastern winds in autumn/winter in the study region often bring polluted air masses from inner PRD and southern China to Hong Kong (Wang et al., 2005; Guo et al., 2009b). Therefore the observed high autumn/winter levels of CS_2 are likely attributed to the transport of polluted air masses, weaker vertical mixing and slower chemical destruction rates. The low average summer values resulted from the transport of clean oceanic air masses from the tropics and unstable rainy weather. The Asian monsoon circulations, together with OH radical seasonality and boundary layer height, have been shown to similarly influence other anthropogenic gaseous pollutants and non-methane hydrocarbons in this region (Wang et al., 2005). Indeed, poor correlations of CS_2 with CO, CH_3I, CH_3Cl and C_2Cl_4 ($R^2 = 0.01–0.06$) were found among the 188 samples, indicating the diversity of CS_2 sources in the study region.

The DMS mixing ratios ranged from a minimum of 2 pptv in October to a maximum of 395 pptv in December 2002, with a mean value of 31 pptv (Fig. 2). Most of these values are on the low end of the range in the air over the open ocean (20–400 pptv) (Warneck, 2000). For example DMS mixing ratios of 300–400 pptv have been observed in productive oceanic regions of the equatorial Pacific (Thompson et al., 1993). The comparatively low values observed in this coastal region during summer suggest a relatively low biological productivity during summer months when oceanic air masses from the South China Sea are sampled. In general, distribution of DMS in surface seawater is determined not only by the total phytoplankton biomass, but also by other factors such as the distribution of special phytoplankton species, consumption by bacteria and photochemical oxidation, and rate of dispersal to the atmosphere (Leck and Persson, 1996). Yang et al. (2000, 2008) found that the sea-to-air DMS flux from the SCS (2.06–5.95 μmol m^{-2} d) fell into the lower end of the range (5.6–11.2 μmol m^{-2} d) of the coastal regions around the world, due to high sea surface temperature, low nutrient concentrations, low chlorophyll *a* levels, and low primary productivity in this subtropical region. The seasonal pattern found in this subtropical region is not consistent with that observed in temperate regions in the southern hemisphere, which presented a fairly strong seasonal cycle due to an enhanced production rate of DMS in summer (Putaud and Mihalopoulos, 1992).

During winter mixing ratios in excess of 300 pptv suggest the dominance of continental sources i.e. vegetation, tropical forests, soil, wetlands and bush fires, together with lower oxidation rates and a poor vertical exchange of air. The speculation on DMS sources is reasonable as data analyses showed poor correlations of DMS with CO ($R^2 = 0.17$), CH_3Cl ($R^2 = 0.06$), CH_3I ($R^2 = 0.04$) and C_2Cl_4 ($R^2 = 0.08$), suggesting that biomass/biofuel burning, oceanic release and urban emissions were not the major continental sources of DMS during winter.

3.1.2. Comparison with other studies

Table 1 compares the levels of the RSCs in the greater PRD region with other studies undertaken in eastern Asia. It is noteworthy that the levels of the RSCs in different locations are related to factors including sampling season, sampling duration, meteorological conditions, site topography, and sampling methods. Here, only spring data (March–May) were averaged in this study due to the fact that most studies were conducted in spring in East Asia. The average OCS and CS_2 mixing ratios in this study were consistent with those obtained in the TRACE-P study, which were measured using the same analytical system and calibration scale as this study (Blake et al., 2004). The OCS level measured from March–May (574 ± 40 pptv) was higher than that on the Japan Sea coast in April (440 ± 10 pptv), whereas the mean CS_2 mixing ratio measured here (47 ± 38 pptv) was significantly lower than on the Japan Sea coast (150 ± 50 pptv) (Inomata et al., 2006). This shows the relative strength of the OCS source in the Hong Kong region. By contrast, our spring CS_2 levels were much higher than that at Jeju Island, Korea (6.4 ± 1.0 pptv) (Kim et al., 2004), showing the remarkable spatial and temporal variations of this short-lived species. By comparison, the average DMS mixing ratio in this study was comparable to that at Jeju Island of Korea (18.7 ± 1.9 pptv) and on the Japan Sea coast (14 ± 2 pptv) but much lower than those measured at Gunsan and Donghae, Korea (Kim et al., 2004; Inomata et al., 2006; Song et al., 2007), showing the weaker marine and continental DMS sources at this study site.

3.2. Regional, local and marine RSCs in the greater PRD

Based on the results of Lagrangian Particle release simulation (Section 2.3), 27 of 188 samples originated from the Hong Kong local area, 23 samples were predominantly from the inner PRD region and 19 samples were identified as marine air in origin. The rest of the samples were affected by a mix of local, regional and marine air.

Table 2 shows the average mixing ratios of the RSCs for South China Sea marine samples, Hong Kong urban samples, and inner PRD regional samples. Interestingly, the mean mixing ratio of OCS in the inner PRD (1021 ± 221 pptv) was significantly higher than that in Hong Kong air (646 ± 48 pptv) and South China Sea air (662 ± 131 pptv) ($p < 0.001$). This indicates strong continental emissions of OCS in the inner PRD compared to Hong Kong and marine air above the South China Sea. The OCS levels in the inner PRD are consistent with that observed in Beijing (841 ± 50 pptv) (Mu et al., 2002). Nevertheless the OCS mixing ratio in the South China Sea air is much higher than that in the remote Pacific (495 ± 9 pptv), the northern North Pacific (452 ± 37 pptv), and the Indian Ocean (510 pptv) (Mihalopoulos et al., 1992; Thornton et al., 1996; Aranami and Tsunogai, 2004). The elevated marine OCS level in South China Sea air suggests potential impact of anthropogenic emissions from the greater PRD region. Indeed, correlation analyses on the South China Sea air showed strong association between OCS and the combustion tracer of CO ($R^2 = 0.88$), as well as between OCS and the marine tracer of CH_3I ($R^2 = 0.73$), implying that the marine OCS level over the South China Sea was determined by both oceanic emissions and anthropogenic sources in the region (Fig. 3a-1).

The DMS mixing ratios in the inner PRD and Hong Kong urban air are comparable to the continental levels measured over the North Atlantic (27 ± 4 pptv) and over the northern North Pacific (31 ± 6 pptv) (Cooper and Saltzman, 1993; Aranami and Tsunogai, 2004). In contrast, the DMS mixing ratio in South China Sea air is much lower than the marine values observed over the North Atlantic (118 ± 8 pptv) and North Pacific (471 ± 47 pptv), but consistent with the marine level in the South Atlantic (34 ± 3 pptv) (Cooper and Saltzman, 1993; Aranami and Tsunogai, 2004). This suggests that, like the South Atlantic, the South China Sea is not a region of strong marine DMS production. Statistical analysis found that correlation between DMS and CH_3I (oceanic tracer) in South China Sea air was moderate ($R^2 = 0.48$, Fig. 3a-2). In addition, there were no statistical differences in the DMS mixing ratio among the marine air, urban Hong Kong air and the inner PRD regional air ($p = 0.35–0.82$), confirming a weak oceanic source of DMS in the region. In addition to oceanic emission, DMS is also released from biomass burning, vegetation and soils (Watts, 2000; Meinardi et al.,

Table 1
Comparison of reduced sulfur compounds mixing ratios in different studies (unit: pptv).

	This study[a]	Song et al. (2007)	Kim et al. (2004)	Blake et al. (2004)	Inomata et al. (2006)
Mean ± 95% Confidence Interval (Median)	OCS				
	574 ± 40 (560)	N/A[b]	N/A	580	440 ± 10
	DMS				
	22 ± 5 (24)	478 ± 34 (498)[c] 670 ± 425 (772)[d]	18.7 ± 1.9	N/A	14 ± 2
	CS_2				
	47 ± 38 (35)	N/A	6.4 ± 1.0	20	150 ± 50
Sampling period	March–May 2002	April 2004	April 2001	February–April 2001	April 1996
Location	Tai O, Hong Kong	Gunsan and Donghae, Korea	Gosan, Korea	Western Pacific	Wakasa Bay, Japan
Remarks	Rural/coastal areas	Residential/coastal areas East Sea coastal areas	Jeju Island	Below 2 km altitude	Japan Sea coast, below 0.5 km altitude

[a] The mixing ratios shown here are springtime values because the other studies were mainly carried out in spring.
[b] Not available.
[c] Mixing ratio at Gunsan, Korea.
[d] Mixing ratio at Donghae, Korea.

Table 2
Mean mixing ratios of RSCs in the inner PRD, urban Hong Kong and South China Sea (Unit: pptv).

RSCs	Lifetime	South China Sea air ($n = 19$)		Urban Hong Kong air ($n = 27$)		Inner PRD air ($n = 23$)	
		Mean (S.D.)	Median	Mean (S.D.)	Median	Mean (S.D.)	Median
OCS	16 yr	662 (131)	569	646(48)	592	1021 (221)	774
DMS	1–2 d	27 (7)	22	37(19)	23	29 (14)	14
CS_2	2–3 d	76 (63)	32	89(36)	57	72 (28)	51

S.D. = standard deviation with 95% confidence interval.

2003), though those sources also do not appear to be strong in the PRD region.

The CS_2 mixing ratios found in this study (5–896 pptv) are mostly much higher than those over the North Pacific (14–43 pptv) and the Atlantic (0.9–6.4 pptv) for both continental and marine air (Cooper and Saltzman, 1993; Aranami and Tsunogai, 2004), implying that the greater PRD is a relatively strong continental source of CS_2. The mean mixing ratio of CS_2 in the inner PRD (72 ± 28 pptv) was comparable to that in urban Hong Kong (89 ± 36 pptv) and the South China Sea air (76 ± 63 pptv) ($p = 0.07–0.22$), reflecting the presence of oceanic emissions of this RSC in the study region, in addition to continental emissions. This can be confirmed by the fact that CS_2 had moderate correlation with CH_3I ($R^2 = 0.48$) as well as CO ($R^2 = 0.67$) in the South China Sea air (Fig. 3a-3). It is reported that the ocean contributes about 27–34% to the total CS_2 in global scale (Chin and Davis, 1993; Watts, 2000), and the oceanic CS_2 is mainly released from the rotting of organic material in the oceans (Khalil and Rasmussen, 1984).

3.3. RSCs emission estimates for inner PRD and Hong Kong

Since sources of CO in the study region are relatively well known and CO often co-locates with other air pollutants including RSCs, we can use the slopes of CO to RSCs to roughly estimate the emissions of RSCs provided that there is a strong correlation between the measured CO and a particular RSC. To make quantitative estimates of emissions from the inner PRD and Hong Kong, the observed relationships between RSCs and CO were used to determine the corresponding RSC emissions. We removed the influence of the backgrounds of these pollutants on the RSC:CO

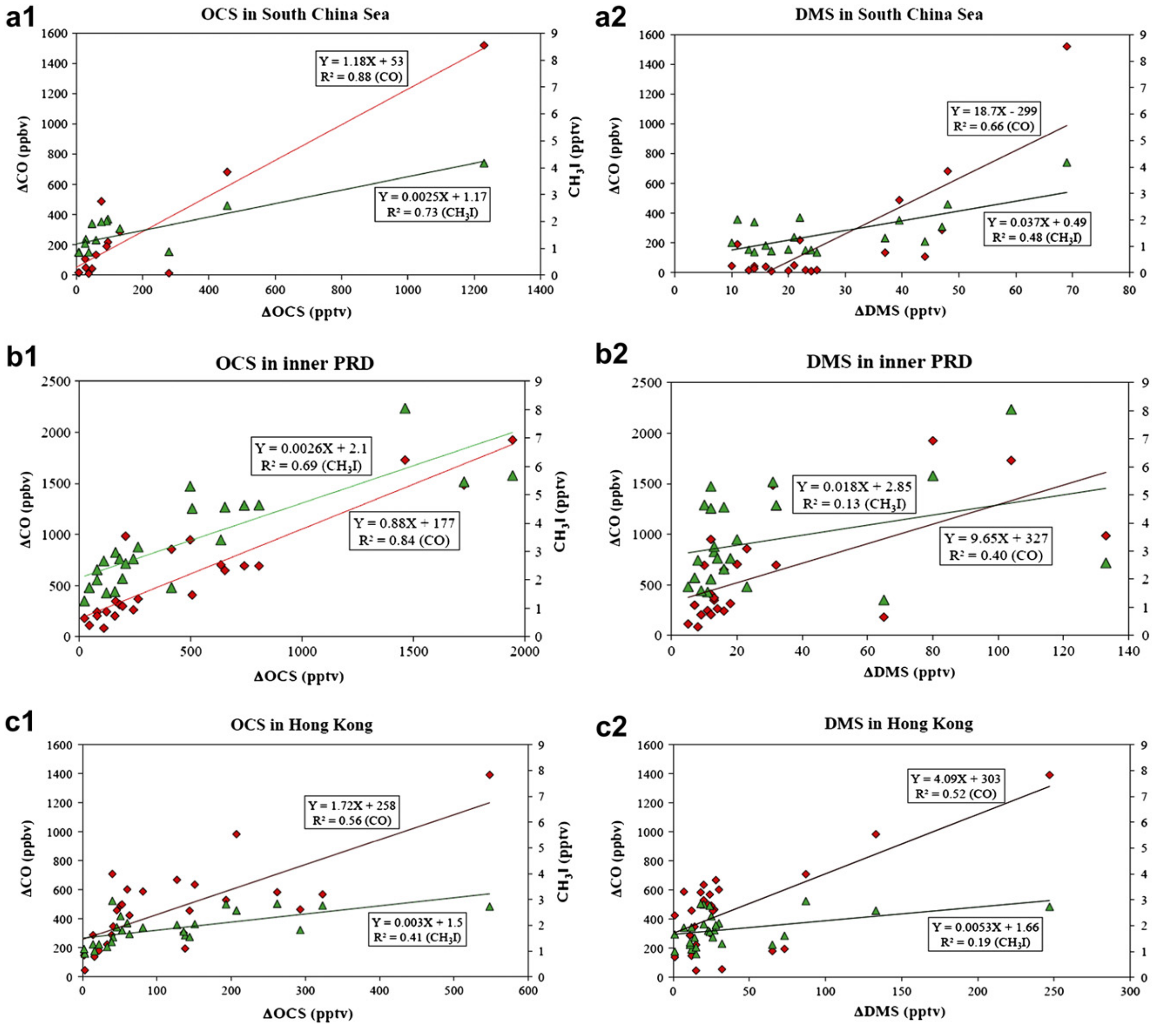

Fig. 3. Scatter plots of RSCs with combustion tracer CO and oceanic marker CH_3I (a) in South China Sea air; (b) in inner PRD; (c) in Hong Kong.

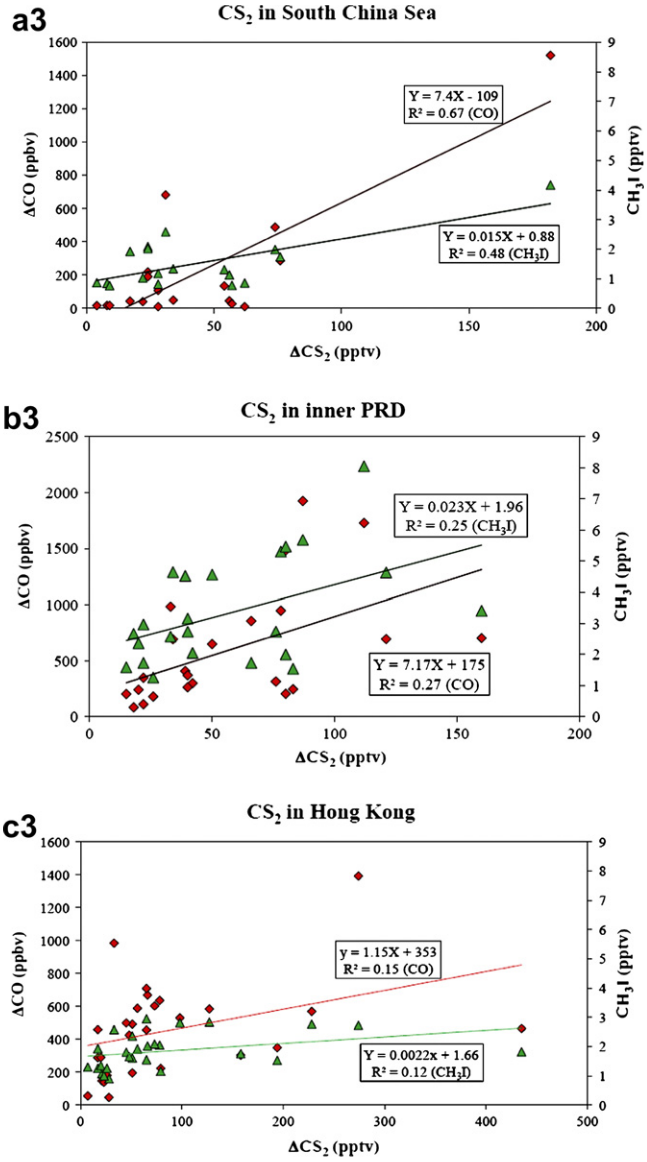

Fig. 3. (*continued*).

relationship by subtracting background values. Here the lowest 5 percentile OCS value of the 188 samples (532 pptv) and the global background level of CS$_2$ (1 pptv) are used as background values for the RSCs (Warneck, 2000). The OCS background value used here is higher than the remote marine background levels i.e. Pacific and Indian Ocean, but lower than the South China Sea background, suggesting it is appropriate to use the lowest OCS value as the background in the region. On the other hand, the global background of CS$_2$ is suitable for the background in the study region given that its lifetime is short. In addition, the lowest CO mixing

Table 3
Measured RSCs:CO relationships and estimated RSC emissions.[a]

	Inner PRD ($n = 23$) Uncertainty for ΔCO: 516 ppbv				Urban Hong Kong ($n = 27$) Uncertainty for ΔCO: 290 ppbv				South China[d]
	R^2	Uncertainty for ΔX (Standard deviation)	ΔX/ΔCO (pptv/ppbv)	Emission (Gg yr^{-1})[c]	R^2	Uncertainty for ΔX (Standard Deviation)	ΔX/ΔCO (pptv/ppbv)	Emission (Gg yr^{-1})	Emission (Gg yr^{-1})
OCS	0.84	541 pptv	0.96 ± 0.09[b]	49.6 ± 4.7	0.56	126 pptv	0.32 ± 0.06	0.32 ± 0.05	52 ± 4
CS$_2$	0.27	38 pptv	0.04 ± 0.01	0.92 ± 0.23	0.15	96 pptv	0.13 ± 0.06	0.05 ± 0.009	18 ± 3
DMS	0.40	34 pptv	0.04 ± 0.01	0.95 ± 0.24	0.52	51 pptv	0.13 ± 0.02	0.07 ± 0.009	–
Total				51.5 ± 4.7				0.44 ± 0.05	

[a] The parameter n is the number of samples; R^2 is the Pearson correlation coefficient of ΔX/ΔCO; ΔX/ΔCO is the reduced major axis regression slopes (pptv/ppbv).

[b] Slope uncertainties are calculated by assuming the linear model.

[c] The emission estimates are calculated using ΔCO:ΔRSC relationships measured in the study period and a bottom-up emission inventory for anthropogenic CO in inner PRD and Hong Kong is used (Streets et al., 2003, 2006; CGRER, 2007).

[d] Values based on estimated emissions from TRACE-P (2001) (Blake et al., 2004).

ratio (82 ppbv) in the South China Sea air is defined as the CO background value. Fig. 3b (1–3) and c (1–3) illustrate the correlations of ΔRSCs (i.e. OCS, CS_2 and DMS) with ΔCO and CH_3I in inner PRD and Hong Kong, respectively. Table 3 shows the measured ΔCO:ΔRSC correlations using reduced major axis (RMA) regression method and the estimated RSC emissions for inner PRD and Hong Kong. The RMA is more appropriate than standard ordinary least squares regression as both variables are subject to sampling and measurement error. The estimates of the uncertainty (i.e. standard deviation) for each variable (species) are also listed in Table 3. The background mixing ratio for each species was considered to be constant. The CO emissions for the inner PRD and Hong Kong that were used for this calculation are based on the TRACE-P emission inventory for the year 2000 (Streets et al., 2003; CGRER, 2007) and updated inventory for 2001 (Streets et al., 2006), namely 8557 Gg and 170 Gg, respectively. It should be noted that the relatively weak $\Delta CS_2/\Delta CO$ correlations ($R^2 = 0.27$ and 0.15, respectively) for air masses originating from the inner PRD and Hong Kong means that our corresponding emission estimates are highly uncertain. It is also noteworthy that uncertainties on the ΔRSC:ΔCO slopes (Table 3) are calculated by assuming a linear regression model, and there are additional errors associated with this assumption that we do not take into account.

Bearing these issues in mind, the total RSC emissions in the inner PRD (51.5 ± 4.7 Gg yr^{-1}) were much higher than those in Hong Kong (0.44 ± 0.05 Gg yr^{-1}) (Table 3), reflecting different source strengths between these two areas. For individual RSCs, the estimated emissions of OCS, CS_2 and DMS in the inner PRD were 155, 18 and 14 times those in Hong Kong, respectively, suggesting significant contributions to ambient RSCs from the inner PRD region. The estimated OCS emission (49.6 ± 4.7 Gg yr^{-1} for the inner PRD and 0.32 ± 0.05 Gg yr^{-1} for Hong Kong) in this study was about 96% of that in South China (52 ± 4 Gg yr^{-1}) estimated by Blake et al. (2004), and 92% (54 ± 28 Gg yr^{-1}) of the total emission in China in anthropogenic emission inventory developed by the University of Iowa (Blake et al., 2004), suggesting that the greater PRD region is a main contributor of OCS in South China and even in China. On the other hand, the estimated CS_2 emission in this study (0.92 ± 0.23 Gg yr^{-1} for inner PRD and 0.05 ± 0.009 Gg yr^{-1} for Hong Kong) accounted for only 5% of the total CS_2 emission (18 ± 3 Gg yr^{-1}) in South China, and 2% in China (43 ± 28 Gg yr^{-1}), implying that the predominant sources of CS_2 are not in the greater PRD region of southern China, or the conversion of CS_2 into OCS is fast in this subtropical region. Though the estimated DMS emission in the inner PRD (0.95 ± 0.24 Gg yr^{-1}) was much higher than that estimated in Hong Kong (0.07 ± 0.009 Gg yr^{-1}), the total DMS emission in the study region appeared not significant, compared to the global flux (24.5 ± 5.3 Tg yr^{-1}) (Watts, 2000).

4. Summary and conclusion

A comprehensive dataset of RSCs collected at a site on the South China coast was analyzed. The 188 air samples were classified into inner PRD, urban Hong Kong, and South China Sea air based on the backward Lagrangian particle release simulations. The ΔRSC:ΔCO ratios were applied to the classified samples to estimate emissions of three major RSCs in the inner PRD and urban Hong Kong. Results indicate that in the greater PRD, the OCS and CS_2 levels were higher in autumn/winter and lower in summer because of the impact of Asian monsoon circulations, while comparatively low DMS values observed in this coastal region during summer suggest a relatively low biological productivity during summer months when oceanic air masses from the South China Sea are sampled. In comparison, the mixing ratios of OCS in the study region were higher than those in other East Asia regions, but the CS_2 and DMS levels were low. On the other hand, the mean OCS level in the inner PRD was much higher than that in Hong Kong and South China Sea air, whereas the mixing ratios of CS_2 and DMS were comparable among the three locations. The estimated OCS emission in the greater PRD was about 96% of the total emission in South China, while the estimated CS_2 and DMS emissions accounted for a very small percentage of their total emissions in South China, suggesting the importance of OCS emission in the greater PRD region.

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