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The annual emissions of sulfur gases from different tidal flats in the Yellow River Delta, China

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The annual emission of sulfur gases (H_2S and COS) from different tidal flats in the Yellow River Delta, China were studied from February to December 2013 by using static chamber-gas chromatography technique. The result showed that the annual emissions of H_2S and COS were featured with obvious seasonal variations. For the full year, the high tidal flats, middle tidal flats and low tidal flats were all the emission sources for H_2S, and the range was on the order of 0.14-7.31 $\mu g \cdot m^{-2} \cdot h^{-1}$, 0.22-6.38 $\mu g \cdot m^{-2} \cdot h^{-1}$ and 0.23-8.80 $\mu g \cdot m^{-2} \cdot h^{-1}$ respectively, and the means were 3.37 $\mu g \cdot m^{-2} \cdot h^{-1}$, 1.98 $\mu g \cdot m^{-2} \cdot h^{-1}$ and 3.29$\mu g \cdot m^{-2} \cdot h^{-1}$ respectively. For COS, the high tidal flats were also the emission sources, while the middle tidal flats and the low tidal flats were the sink, and the range was on the order of -1.16-2.52 $\mu g \cdot m^{-2} \cdot h^{-1}$, -3.32-1.25 $\mu g \cdot m^{-2} \cdot h^{-1}$ and -5.53-1.78$\mu g \cdot m^{-2} \cdot h^{-1}$ respectively, and with means of 0.68 $\mu g \cdot m^{-2} \cdot h^{-1}$, -0.09$\mu g \cdot m^{-2} \cdot h^{-1}$ and -0.17 $\mu g \cdot m^{-2} \cdot h^{-1}$ respectively. The annual emissions of H_2S and COS from the different tidal flats in the Yellow River Delta China were significantly affected by the annual seasonal variation, and the emissions of H_2S and absorptions of COS from the different tidal flats were mainly concentrated in the growing season of plants (from May to October). In the different tidal flats areas, the annual emission amounts of H_2S and COS were also different, among which H_2S was manifested as low tidal flats > high tidal flats > middle tidal flats, and COS as high tidal flats > low tidal flats > middle tidal flats.

***Key words*:** Yellow River Delta; Tidal flats; Sulfur gases; Annual emission; China

INTRODUCTION

Volatile sulfur gases were an important part of the sulfur cycle in nature, it had a great impact on the environment and was closely related to acid deposition, the greenhouse effect, aerosol formation and other processes [1, 2]. Sulfur gases emitted from the nature were one of the main sources of sulfur gases in the atmosphere. According to the estimates the sulfur gases from natural sources were equal to such gases from human activities [3, 4]. However, the greater spatial, and temporal variability, of natural sources of sulfur gases emission, coupled with limited monitoring data, has brought great uncertainty to the global sulfur budget [5, 6]. Wetlands were one of the most important natural sources of sulfur emissions, and sulfur gases emitted from wetlands were generally one, or several orders of magnitude higher than those emitted from inland due to their unique natural and ecological conditions [7]. Scholars had researched on the emission fluxes of sulfur gases, its effect factors and its emission mechanism in freshwater marshes, salt marshes, coastal waters and different types of wetlands [8-15].

The researches have shown that the emissions of sulfur gases from the wetlands had greater spatial and temporal variability, and the kind of sulfur gases emitted from the wetlands were mainly hydrogen sulfide (H_2S), carbonyl sulfide (COS), dimethyl sulfide (DMS), carbon disulfide (CS_2), mercaptanes (MeSH) and dimethyl disulfide (DMDS), and so on [8-11,13, 16]. Among them, H_2S was highlighted by a higher emission flux in the saline wetland along inshore areas and seashore [17], and was also closely related to carbon mineralization and methane emissions [18-21]; COS was a kind of reduced sulfur compound with the highest abundance in the atmosphere [22] and it was controversial in understanding about its source/sink [23]. Sulfur gases were released mainly from the decomposition of organic matter and sulfate reduction, and its emissions rate was affected by many factors, such as temperature, tides, redox potential, vegetation type, and so on

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[3,9,10,13,24].

The Yellow River is well known as a sediment-laden river, and the Yellow River Delta are China's best preserved, and largest warm temperate, nascent wetlands, and are also a typical estuarine wetlands. They are an important bird habitat, breeding ground, transit station, with a classic permittivity, fragility, rarity and are of international importance. The Yellow River on average delivers an annual input of 1.0×10^9t of sediment into the estuary, with about 45% of the deposition in the coastal zone. This provides the material basis for the development of tidal estuaries. The re-shaping caused by the frequent swings and marine dynamics of the Yellow River tail forms the Delta's broad tidal wetlands. With a total area of about 964.8km^2, accounting for 63.06% of the total area of the Yellow River Delta [25]. This area is one of the important ecological zones of the Yellow River Delta. Under the interactive influence of tides, overflow and other marine dynamics along with estuarine runoff, sediment and other land-based forces, the difference in intensity among these forces and the varying lengths of flooding time, give the Yellow River Delta coastal tidal area an obvious horizontal belt character, forming parallel high, middle and low tidal flats in a seaward direction [26]. This is accompanied by a succession of typical natural biomes. From the low tidal flats to medium tidal flats to high tidal flats, it is mainly *Suaeda salsa* communities, *Suaeda salsa-Tamarix chinensis* communities and *Suaeda salsa-Phragmites australis* communities that are successively encountered (Xing, et al. [27]). This parallel-shaped distribution of low mass and vegetation, pulsing the impact of land-sea interaction inevitably, cause differences in the biological substances circulating throughout the sub-tidal zone wetland ecosystem, thereby affecting the stability of the wetlands so that their appearance and structure are always in flux.

Current research on the biogenic elements cycle in the tidal flats of the Yellow River Delta China mainly focused on the accumulation and distribution features of plant elements (C, N, P, S and trace elements) [28,29], distribution of elements in soils [30,31] and greenhouse gas emissions [31-34]. To date, the studies on the emissions of sulfur gases from the tidal flats in Yellow River Delta china were still lacking, so the paper was to determine the annual emissions of sulfur gases from the different tidal wetlands in Yellow River Delta China by using static chamber-gas chromatography technique, so as to provide essential data for evaluating the influence of sulfur gases emission on atmospheric environment, understanding the relationship between the emission of H_2S and CH_4 and further studying on the sulfur cycle in the natural wetland of the Yellow River Delta, China .

1. MATERIALS AND METHODS

1.1 Site study

This study was conducted from February to December 2013 at the typically experimental plots located in the Nature Reserve of Yellow River Delta (37°35′N~38°12′N, 118°33′E~119°20′E) in Dongying City, Shandong Province, China. The nature reserve is of typical continental monsoon climate with distinctive seasons; summer is warm and rainy while winter is cold. The annual average temperature is 12.1°, the frost-free period is 196 days, and the effective accumulated temperature is about 4300 °. Annual evaporation is 1962 mm and annual precipitation is 551.6 mm, with about 70 percent of precipitation occurs between June and August. The soils in the study area are dominated by intrazonal tide soil and salt soil, and the main vegetations include *Phragmites australis*, *S. salsa*, *Triarrhena sacchariflora*, *Myriophyllum spicatum*, *chinensis* and *Limoninum sinense* [27].

The typical experiment area was selected in the coastal tidal wetland situated at the north of the mouth of the Yellow River. In the typical experiment area, the 3 typical sample points including high tidal flats ((37°46′7.25″N, 119°09′55.54″E), middle tidal flats (37°46′11.62″N, 119°09′56.09″E) and low tidal flats (37°46′15.95″N, 119°09′57.44″E) were selected, and it’s vegetation distribution was continuous, and the vegetation of high tidal flats, middle tidal flats and low tidal flats was *Suaeda Salsa-Phragmites australis,* the *Suaeda Salsa-Tamarix Chinensis* and *Suaeda Salsa* community respectively. Three repeated monitoring points were distributed in the each tidal flats, and the total monitoring points were 9, and the gas samples were collected every month or every two month, each sampling period was at 8:00-10:00am.

1.2 Collection and analysis of gas samples

Gas samples are collected with closed chamber method [24]. The chamber was made of polycarbonate with an internal height of 100cm, covered an area of 0.25m^2 (50cm in length and 50 cm in width) of test field. Each chamber, in the internal top had a small fan for mixing the air. To avoid disturbing the soil, the chamber were placed on a stake driven into the soil installed in December 10, 2011, meanwhile the boardwalk were installed for minimizing disturbance to the test field during sampling. Four gas samples of the chamber air were collected into a 1000ml Tedlar bag by a small sampling pump at a flow rate of 2000 ml.min^{-1} at 0, 20, 40 and 60 min after the chamber was set up. The sulfur fluxes were determined by measuring the temporal change of the concentration in the air inside the chamber. Therefore a positive values refers to the emission from the wetland to the atmosphere and negative values to the absorption into the plants and soil of wetlands.

The concentrations of H_2S and COS were determined as described in detail by Li et al. [24].

1.3 Statistical analysis

Data graphics are made available with Origin7.5 and statistical analysis is developed with SPSS13.0.

2. RESULTS AND DISCUSSION

2.1 Annual emission characteristics of sulfur gases from the different tidal wetlands in Yellow River Delta, China

2.1.1 Annual emission characteristics of sulfur gases from the high tidal flats. The high tidal flats are located between the high tide level and neap tide level, with a width in the range of 1-9 km range. It is mainly composed of *Suaeda salsa-Phragmites australis* communities. The annual emissions of H_2S and COS were shown in Fig.1, the annual emissions of H_2S and COS from the high tidal flats both showed significant seasonal variation (Fig.1), and the annual emission range of H_2S was 0.14-7.31μg·m^{-2}·h$^{-1,}$ with a mean of 3.29 μg·m^{-2}·h^{-1}, and a variation coefficient of 81.7%.

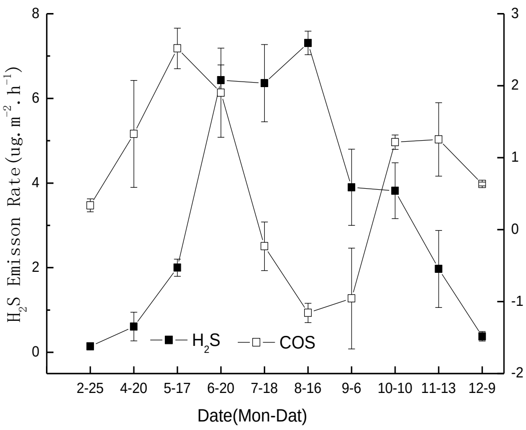

Fig.1 The annual emissions of H_2S and COS from the high tidal flats.

From February-August, the emission flux of H_2S increased, and peak emissions occurred in August, and from September to December, the emission amount of H_2S began to decrease. The annual emission of COS showed alternating emission-absorption characteristics, and the range of COS annual emission was -1.16-2.52μg·m^{-2}·h^{-1}, with a mean of 0.68 μg·m^{-2}·h^{-1}, and a variation coefficient of 175.0%. From February to May, the emission amount of COS increased, and afterwards gradually decreased. Absorption was manifested in July, the emergence of the absorption peak was seen in August with the value of -1.16μg·m^{-2}·h^{-1}. The weak absorption was shown in September, and from October to December, the emission flux of COS took over to release.

2.1.2 Annual emission characteristics of sulfur gases from the middle tidal flats

The middle tidal flats are located between the neap tide level and the neap tide low tide level with a width up to 1-4 km. It is mainly composed of *Suaeda salsa-Tamarix chinensis* communities.

These are *Suaeda salsa* communities with transition phases to *tamarisk* communities or *Phragmites australis* communities. In the middle tidal flats, the annual emissions of H_2S and COS also showed significant seasonal variation (Fig.2).

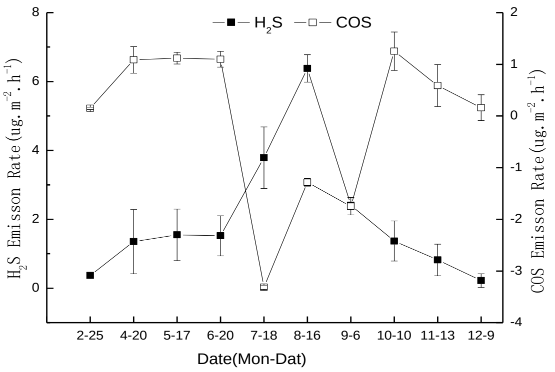

Fig.2. The annual emissions of H_2S and COS from the middle tidal flats.

The annual emission range of H_2S was 0.22-6.38 $\mu g\cdot m^{-2}\cdot h^{-1}$, with a mean of 1.98$\mu g\cdot m^{-2}\cdot h^{-1}$, and a variation coefficient of 93.8%, as an H_2S release source. From February to August, the emission amount of H_2S showed an increasing trend, and appeared the emission peak in August, then the emission amount of H_2S gradually decreased, and the lowest value appeared in December. The emission amount of COS was in the range of -3.32-1.25 $\mu g\cdot m^{-2}\cdot h^{-1}$, with a mean of -0.09$\mu g\cdot m^{-2}\cdot h^{-1}$, and a variation coefficient of 732.2% with large variability. From an annual point of view, the middle tidal flat was a weak sink for COS. The variation of COS emission showed fluctuations, among which, the emission amount of COS manifested from February to June, then volatile absorption was seen from July to September, and the absorption peak (-3.32 $\mu g\cdot m^{-2}\cdot h^{-1}$) appeared in July, and from October to December, COS emissions were seen again, while the emission amount of COS decreased.

2.1.3 Annual emission characteristics of sulfur gases from the low tidal flats. The low tidal flats are located between the neap tide low tide level and the low tide level. The low tidal flats are narrow with an average width of 0.5-2 km. They are mainly composed of *Suaeda salsa-Phragmites australis* communities which are succession pioneer communities in the Yellow River Delta wetlands. The annual emissions of H_2S and COS from the low tidal flats had a clear seasonal variation (Fig.3), in which the emission range of H_2S was from 0.23 $\mu g\cdot m^{-2}\cdot h^{-1}$ to 8.80 $\mu g\cdot m^{-2}\cdot h^{-1}$, with a mean of 3.37 $\mu g\cdot m^{-2}\cdot h^{-1}$, and a variation coefficient of 85.7%, as an H_2S release source.

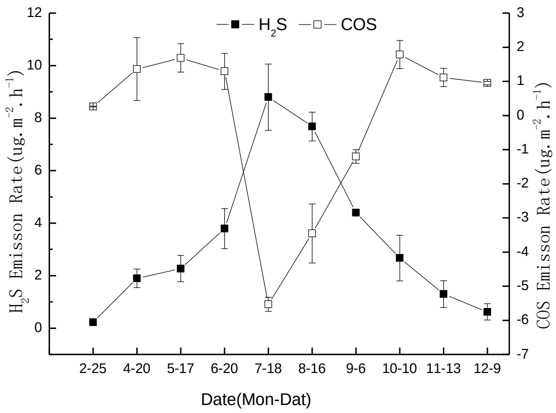

Fig.3. The annual emissions of H_2S and COS from the low tidal flats

The emission mode of H_2S was a single peak, and from February to July, the emissions amount of H_2S increased gradually until the peak discharge manifested in July, and from July to December, the emissions amount of H_2S gradually decreased. While the annual emission of COS showed alternating emission-absorption characteristics, and the range of the COS emission amount was -5.53-1.78 $\mu g\cdot m^{-2}\cdot h^{-1}$, with a mean of 0.17 $\mu g\cdot m^{-2}\cdot h^{-1}$, and a variation coefficient of 506.4% which showed a large amount of variability for COS emission. From February to June, there were emissions of the COS, but there was little change in the emission amounts. From July to September, absorption of COS was manifested, and the absorption peak occurred in July, as -5.53 $\mu g\cdot m^{-2}\cdot h^{-1}$, then weak emission was also manifested from October to December and the emission amount decreased.

2.2. Comparison of the annual emissions of sulfur gases from the different tidal flats in the Yellow River Delta, China

Previous studies have indicated that the sulfur gases emissions from wetlands were significantly affected by annual and seasonal changes, as well as plant growth [10,11]. Looking at the full year, temperature was relatively high from May to October in the Yellow River Delta, china, and this was also growth season for plants. From November to the following April, the temperature was relatively low, and it was the season when plants are dying off, a non-growing season. We calculated the average emission amounts of H_2S and COS from the different tidal flats in the plant growth season and non-growth season according to this division (Table 1). From Table 1 we can see that the averaged emission amounts of H_2S from the high tidal flats, middle tidal flats and low tidal flats in the growing season were 6.4, 4.1, and 4.9 times higher than that in the non-growing season respectively. It can be seen that H_2S emissions were mainly concentrated in the growing season. For the COS, the averaged emission value in the high tidal flats showed emissions in both the growth and non-growth seasons, in the growing season, although there was periodic absorption in high tidal flats, its absorption capacity was less than the middle tidal flats and middle tidal flats, resulted in overall emission in the plant growing season. While the middle tidal flats and low tidal flats were different from the high tidal flats, and they showed absorption in the growth season and emission in the non-growth season which was well agreed with the results of Fall [36] and Whelan et al. [13].

From an annual perspective, the averaged emission amount of the H_2S from the low tidal flats was the highest (3.37 $\mu g \cdot m^{-2} \cdot h^{-1}$) which was related to the low tidal flats near the sea and effecting strongly by tidal, followed by the high tidal flats (3.29$\mu g \cdot m^{-2} \cdot h^{-1}$) and the middle tidal flats (1.98 $\mu g \cdot m^{-2} \cdot h^{-1}$). In terms of COS, the averaged emission amount from the high tidal flats was the highest (0.68$\mu g \cdot m^{-2} \cdot h^{-1}$), followed by the low tidal flats manifesting absorption, and that from the middle tidal flats was the lowest with weak absorption. The reasons for the different H_2S and COS amounts in each tidal flats may be related to differences in research samples in terms of vegetation type, soil matrix, hydrothermal interaction and salinity conditions, or other relevant conditions. There is a need to explore these areas in future studies.

3. CONCLUSIONS

The emissions of H_2S and COS from the different tidal flats in the Yellow River Delta China were all shown with obvious seasonal variations. For the full year, the high tidal flats, middle tidal flats and low tidal flats were emission sources for H_2S, and the range was on the order of 0.14-7.3$\mu g \cdot m^{-2} \cdot h^{-1}$, 0.22-6.38 $\mu g \cdot m^{-2} \cdot h^{-1}$ and 0.23-8.80 $\mu g \cdot m^{-2} \cdot h^{-1}$ respectively, and with means of 3.29 $\mu g \cdot m^{-2} \cdot h\text{-}^{1}$, 1.98$\mu g \cdot m^{-2} \cdot h^{-1}$ and 3.37$\mu g \cdot m^{-2} \cdot h^{-1}$ respectively. For COS, the high tidal flats manifested emissions, while the middle tidal flats and the low tidal flats manifested absorption, the range was on the order of-1.16-2.52 $\mu g \cdot m^{-2} \cdot h^{-1}$, -3.32-1.25 $\mu g \cdot m^{-2} \cdot h^{-1}$ and -5.53-1.78$\mu g \cdot m^{-2} \cdot h^{-1}$ respectively, and with means of 0.68 $\mu g \cdot m^{-2} \cdot h^{-1}$, -0.09$\mu g \cdot m^{-2} \cdot h^{-1}$ and 0.-0.17 $\mu g \cdot m^{-2} \cdot h^{-1}$ respectively.

The H_2S and COS emissions from the different tidal flats in the Yellow River Delta China were significantly affected by annual seasonal variation, which caused that H_2S emission and COS

Table 1. The average emission flux of H_2S and COS in plant growing season and non growing season from different tidal flats in the Yellow River Delta China.

Sulfur gases	Time	Average Emission Fluxes ($\mu g \cdot m^{-2} \cdot h^{-1}$)		
		High tidal flat	Middle tidal flat	Low tidal flat
H_2S	Growing Season (May-October)	4.97	2.84	4.93
	Non-growing Season (November-April)	0.78	0.69	1.01
	Annual Average	3.29	1.98	3.37
COS	Growing Season (May-October)	0.55	-0.48	-0.90
	Non-growing Season (November-April)	0.89	0.50	0.92
	Annual Average	0.68	-0.09	-0.17

absorption were mainly concentrated in the growing season. In different tidal wetland areas, H_2S and COS emission amounts were different, among which H_2S was manifested as low tidal flats > high tidal flats > middle tidal flats, and COS as high tidal flats > low tidal flats > middle tidal flats. The differences may be related to vegetation types, tidal action, the soil matrix, hydrothermal interaction, salinity conditions, or other relevant conditions. Future in-depth studies are required.

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ГОДИШНИ ЕМИСИИ ОТ СЯРА-СЪДЪРЖАЩИ ГАЗОВЕ В ПРИЛИВНИТЕ ПЛИТЧИНИ В ДЕЛТАТА НА ЖЪЛТАТА РЕКА, КИТАЙ

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(Резюме)

Изследвани са годишните емисии на сяра-съдържащи газове (сероводород и карбонил-сулфид) от различни приливни плитчини в делтата на Жълтата река за периода от февруари до декември, 2013 г. с помощта на статично пробонабиране и газ-хроматографска техника. Резултатите показват наблюдаеми сезонни колебания на изследваните вещества. За целогодишния период източник на сероводород са високите, средните и ниските плитчини. Интервалите от концентрации са от порядъка съответно 0.14-7.31 $\mu g \cdot m^{-2} \cdot h^{-1}$, 0.22-6.38 $\mu g \cdot m^{-2} \cdot h^{-1}$ и 0.23-8.80 $\mu g \cdot m^{-2} \cdot h^{-1}$, като средните стойности са 3.37 $\mu g \cdot m^{-2} \cdot h$-^{1}, 1.98$\mu g \cdot m^{-2} \cdot h^{-1}$ и 3.29$\mu g \cdot m^{-2} \cdot h^{-1}$. За карбонил-сулфида, високите плитчини са източник на замърсяване, докато средните и ниските са по-скоро консуматори, при кнцентрации от порядъка на -1.16-2.52 $\mu g \cdot m^{-2} \cdot h^{-1}$, -3.32-1.25 $\mu g \cdot m^{-2} \cdot h^{-1}$ и -5.53-1.78$\mu g \cdot m^{-2} \cdot h^{-1}$.Средните стойности на концентраците са съответно 0.68 $\mu g \cdot m^{-2} \cdot h^{-1}$, -0.09$\mu g \cdot m^{-2} \cdot h^{-1}$ и -0.17 $\mu g \cdot m^{-2} \cdot h^{-1}$. Годишните емисии на сероводород и карбонил-сулфид от различните приливни плитчини в делтата на Жълтата река се влияят значително от годишните сезонни колебания, като емисиите на сероводород и карбонил-сулфид са главно в периода на растителен растеж (от май до октомври). В различните плитчини годишните емисии на двата замърсителя са различни, като сероводородът се проявява най-много в ниските, после във високите и накрая в средните плитчини. Карбонил-сулфидът най-много се проявява във високите плитчини.