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## Sulfur speciation and bioaccumulation in camphor tree leaves as atmospheric sulfur indicator analyzed by synchrotron radiation XRF and XANES

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### Abstract

Analyzing and understanding the effects of ambient pollution on plants is getting more and more attention as a topic of environmental biology. A method based on synchrotron radiation X-ray fluorescence and X-ray absorption near edge structure spectroscopy was established to analyze the sulfur concentration and speciation in mature camphor tree leaves (CTLs), which were sampled from 5 local fields in Shanghai, China. Annual SO<sub>2</sub> concentration, SO<sub>4</sub><sup>2-</sup> concentration in atmospheric particulate, SO<sub>4</sub><sup>2-</sup> and sulfur concentration in soil were also analyzed to explore the relationship between ambient sulfur sources and the sulfur nutrient cycling in CTLs. Total sulfur concentration in mature camphor tree leaves was 766–1704 mg/kg. The mainly detected sulfur states and their corresponding compounds were +6 (sulfate, include inorganic sulfate and organic sulfate), +5.2 (sulfonate), +2.2 (sulfoxides), +0.6 (thiols and thioethers), +0.2 (organic sulfides). Total sulfur concentration was strongly correlated with sulfate proportion with a linear correlation coefficient up to 0.977, which suggested that sulfur accumulated in CTLs as sulfate form. Reduced sulfur compounds (organic sulfides, thiols, thioethers, sulfoxide and sulfonate) assimilation was sufficed to meet the nutrient requirement for growth at a balanced level around 526 mg/kg. The sulfate accumulation mainly caused by atmospheric sulfur pollution such as SO<sub>2</sub> and airborne sulfate particulate instead of soil contamination. From urban to suburb place, sulfate in mature CTLs decreased as the atmospheric sulfur pollution reduced, but a dramatic increase presented near the seashore, where the marine sulfate emission and maritime activity pollution were significant. The sulfur concentration and speciation in mature CTLs effectively represented the long-term biological accumulation of atmospheric sulfur pollution in local environment.

**Key words:** synchrotron radiation; X-ray fluorescence; X-ray absorption near edge structure; sulfur speciation; sulfur bioaccumulation; camphor tree leaves

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### Introduction

Recent years, air pollution has become one of the top environmental concerns in China with a rapid economic development and huge amount of fossil energy consumption. Atmospheric sulfur pollution such as SO<sub>2</sub> and sulfur-rich airborne particulate matter (PM) in China generally remains on a high annual level in mega cities as coal is the dominant energy source which accounts for about 70% of the total primary energy consumption (Chan and Yao, 2008). The research revealed that SO<sub>2</sub> and sulfur-rich PM such as sulfate particles have profound influence on acid deposition, climate change and the balance of aquatic and terrestrial ecosystem (Boucher and Lonmann, 1995;

Last, 1982; Tsai et al., 2010; Ward, 2009). Compared to conventional physical and chemical monitoring, how to use the accumulative and indicative effects of biology to monitor and evaluate the extent of pollution on environment and its potential impact on biology health is receiving more and more interest (Szczepaniak and Biziuk, 2003; Zhang et al., 2009). Tree leaves can act as excellent biological filters because of large leaf areas and the physical properties of their surfaces, trapping and absorbing air pollutants (Beckett et al., 1998). On the other hand, long-time exposure to high level pollutants can affect the nutrient alimentation and metabolism of tree leaves. Higher sulfur concentration was reported in tree leaves from SO<sub>2</sub> and sulfate-rich particulates polluted areas than that from clean air areas (Bao et al., 2009; Cicek and

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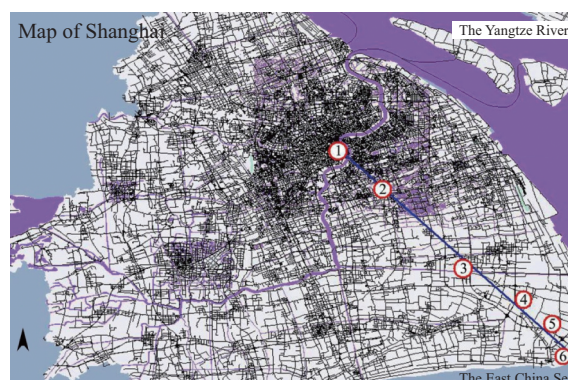
Koparal, 2004; Cowling et al., 1973). Sulfur is a nutrient element of plant with a proportion approximately 0.1% of dry matter (Leustek et al., 2000). It is essential for plants to form vital functions in most proteins and enzymes. Using tree leaves as indicators to monitor atmospheric sulfur pollution has some advantages such as their permanent and common occurrence in local environment, the ease on sampling and the accumulation of contamination in past period. It enables us to have a measure of integrated exposure in remote areas over an extended period of time without expensive technical equipments in sample collection. It also provides another way to understand the physiological hazard of atmospheric sulfur pollution on plants.

The evergreen *Cinnamomum camphora* (L.) Presl (commonly known as camphor tree) with a potential height as high as 30 m originates in Eastern Asia (China and Japan) and has infested to a lot of countries in Asia, Australia and North America (Starr et al., 2003). Camphor tree is a native common greening tree in Shanghai, China and widely used as amenity planting and ornamental forest which especially popular on roadsides, in parks and gardens. The leaves are broadly ovate, 7–10 cm long, 3–5 cm wide, have a glossy, waxy appearance and camphor smell. The old leaves fall and new leaves appear in spring, their exposure time (i.e., life time) in ambient environment is exactly one year. Mature leaves can be easily recognized and picked out as they are bottle green and have most expanded area compared to tender and old leaves on the same branch. Therefore, camphor tree leaves (CTLs) can potentially be used as effective indicators of atmospheric sulfur pollution in Shanghai. As sulfur is essential and ubiquitous with a large variety of species and valence states, from  $-2$  in inorganic sulfide to  $+6$  in sulfate, synchrotron radiation (SR) X-ray fluorescence (XRF) and X-ray absorption near edge structure (XANES) spectroscopy are combined to analyze both sulfur concentration and speciation in mature CTLs as a way to reveal the ambient sulfur bioaccumulation and transformation in local fields. Correlation analysis between sulfur accumulation in mature CTLs and sulfur content in ambient sources such as soil and atmospheric sulfur pollutants ( $\text{SO}_2$  and sulfur-rich airborne particulates) was also discussed.

## 1 Materials and methods

### 1.1 Sampling and preparation

Sampling sites were plotted in different local fields from the urban centre to southeast seashore in Shanghai, which were Bei-Cai (BC), Xuan-Qiao (XQ), Wan-Xiang (WX), Shu-Yuan (SY), Shanghai Maritime University (MU) and approximately 10 (52), 32 (30), 44 (18), 52 (10), 60 (2) km away from Shanghai urban centre (southeast seashore), respectively (Fig. 1, original map was from <http://www.cppblog.com>). These areas were selected as

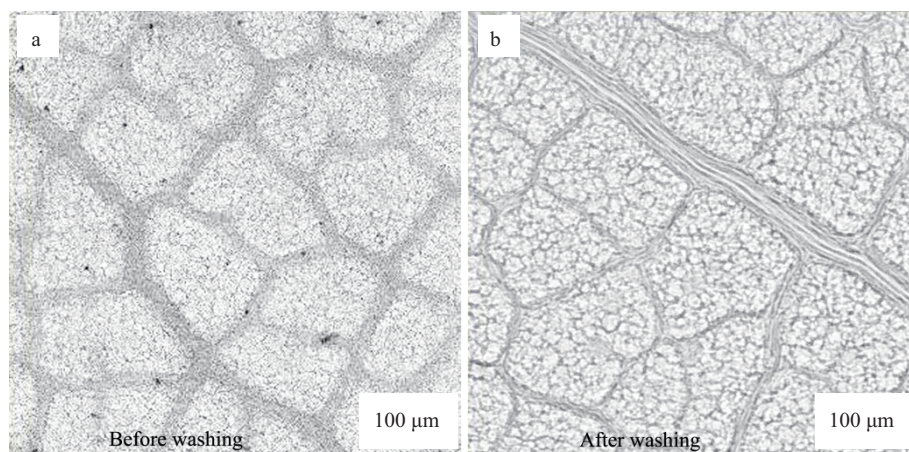


**Fig. 1** Sampling sites location. Five sites, which located in BC (Site 2), XQ (Site 3), WX (Site 4), SY (Site 5) and MU (Site 6), respectively, were assigned along a line from Shanghai urban centre (Site 1) to southeast seashore. The original map was from <http://www.cppblog.com>.

representatives of urban downtown (BC), urban-rural joint (XQ), suburb (WX), coastal suburb (SY) and coast (MU) respectively. The traffic road density relatively represented the burden of human activities on ambient environment in local fields. Except for routine residential activities, there are not any other large anthropogenic exhaust sources within 1 km around every sampling site.

The mature CTLs were sampled at a height of 3–6 m above ground on the sunny side from large camphor trees with trunk diameters over 20 cm at each site on 23 Nov. 2009. As Shanghai locates in the region of East Asian monsoon climate where the spring usually begins in early February, the sampled CTLs almost had grown for a life period and been exposure to the local environment nearly for one year. The process of sample preparation for XRF and XANES analysis are as the following. Fresh sampled leaves were washed in an ultrasonotor (KQ3200E, Kun Shan Ultrasonic Instruments Co., Ltd., China) using ultra-pure water (with a resistivity of 18 M $\Omega$ /cm, the same below) for 1 hr. To ensure the washing was effective, CTLs were imaged before and after washing using Synchrotron radiation X-ray phase contrast imaging technique. These phase contrast imaging experiments were carried on BL13W1 work station in Shanghai Synchrotron Radiation Facility. The electron energy in the storage ring was 3.5 GeV and X-ray of 12 keV was used. The black spots of contamination on leaves were obviously cleaned after washing as shown in Fig. 2. For each sampling site, over 30 pieces of washed mature leaves on the same branch were selected and dried in an oven at 65°C until constant weight was achieved. Both for XRF and XANES spectrum detection, dried leaves were pulverized into fine powder and then pressed into cylindrical pellets using a manual tablet machine. Each pellet had a diameter of 8 mm and a thickness of 4 mm.

To study the relevance between sulfur concentration in mature CTLs and the concentration of sulfur (or sulfur compounds) in local environment, ambient  $\text{SO}_2$  concentration,  $\text{SO}_4^{2-}$  concentration in atmospheric particulate,



**Fig. 2** Images of CTLs surface before and after washing. (a) before washing, the black spots of contamination remained on the leaf surface; (b) after washing treatment, contamination on the leaf was cleaned.

$\text{SO}_4^{2-}$  and sulfur concentration in soil were also analyzed. Daily and annual  $\text{SO}_2$  were monitored by Shanghai Environmental Monitoring Center and issued on the website (<http://www.semc.com.cn>). Annual  $\text{SO}_2$  concentration data in 2009 were gathered for analysis. Atmospheric particulate matter with aerodynamical diameter less than  $2.5 \mu\text{m}$  ( $\text{PM}_{2.5}$ ) at each site during the period of leaves sampling was collected using personal environmental monitors (PEM10-2.5, MSP Corporation, USA) with PTFE filters at a flow rate 10 L/min uninterruptedly for 24 hr (23–24 Nov. 2009). Part (16.53%) of each sampled filter was selected through perforation and extracted ultrasonically for 40 min by 5 mL ultrapure water. Soil samples were collected from each site both at surface layer (0–10 cm in depth) and deep layer (30–40 cm in depth). A part of the soil was dried at  $65^\circ\text{C}$  in an oven until constant weight achieved, then pulverized into fine powder and pressed into cylindrical pellets with a diameter of 8 mm and a thickness of 2 mm for XRF spectrum detection. Another part of soil was dissolved in ultrapure water for  $\text{SO}_4^{2-}$  analysis. Water soluble inorganic  $\text{SO}_4^{2-}$  in the atmospheric particulate and the soil were detected using an ion chromatography (ICS2000, Dionex Corporation, USA) with a separation column (Dionex AS19). The limits of detection for  $\text{SO}_4^{2-}$  was less than  $0.04 \mu\text{g}/\text{mL}$  and the relative standard deviation was less than 5% for reproducibility test.

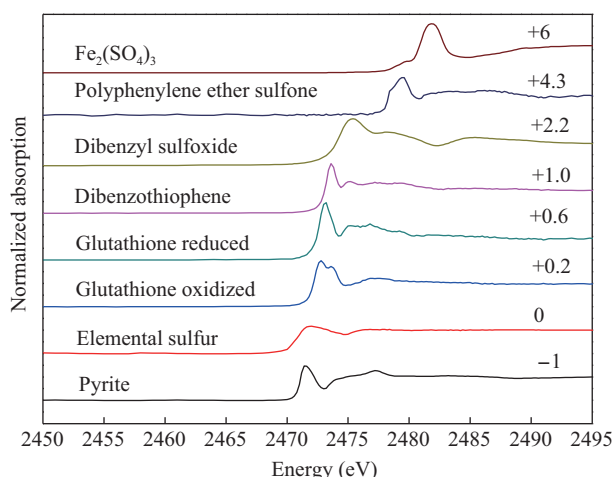
## 1.2 Analysis of XRF and sulfur K-edge XANES spectra

With characterization of high intensity, tunable energy, polarization and small divergence, synchrotron radiation has recently been introduced as a substitute to conventional X-ray and widely used in X-ray based measurement techniques. SR-XRF is a multi-elemental, rapid and sensitive method on detecting elements concentration and has been widely used for the analysis of environmental samples (Butler et al., 2009, 2011). XANES based on SR is particularly suitable for the characterization of sulfur chemical speciation in plants without digestion and separation of sulfur species from samples before analysis. It has

been successfully used to identify the characteristic sulfur species in various samples (Frank et al., 1987; Jalilehvand, 2006; Sandstrom et al., 2002). The combination of XRF and XANES provides a physical way with little risk of converting the forms of sulfur through redox reactions compared to conventional biological and chemical techniques on the determination of sulfur concentration and speciation.

The XRF and XANES spectra of mature CTLs together with the XRF spectra of soil were all detected in fluorescence mode using a Si(Li) detector in high vacuum environment on beamline 4B7A of the Beijing Synchronization Radiation Facility. Electron of 2.5 GeV and beam current of 80–180 mA in the storage ring were introduced during our experiments. A Si(111) double-crystal monochromator was used to vary the X-ray energy from 2420 to 2570 eV with an energy resolution 0.3–0.6 eV. For XRF spectra detection, X-ray energy was set at 3.2 keV and external reference method was used for the determination of sulfur concentration. Standard reference materials of plant (GBW07603) and soil (GBW07405) from the National Research Centre for Certified Reference Materials, Beijing, China, were used as external reference for quantitative analysis. The reference material was also prepared as pellet for XRF analysis. Experimental XRF spectrum was fitted and calculated using PyMca software specially developed for XRF analysis by the European Synchrotron Radiation Facility.

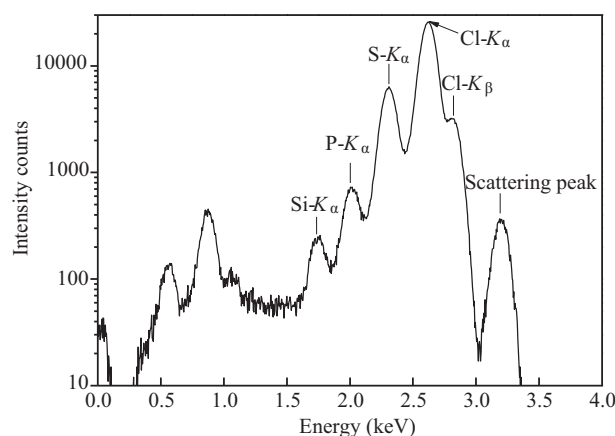
Here, the least-square fitting method developed by Huffman et al. (1991) was used to calculate the sulfur states distribution quantitatively in mature CTLs. This method fits the XANES to a series of peaks representing  $s \rightarrow p$  transitions and scattering resonances, and to several arctangent step functions representing the transition of ejected photoelectrons to the continuum. It bases on that a high degree of linear correlation has been observed between sulfur oxidation state and the K-edge absorption energy of main peak (the white line) (Frank et al., 1987; Waldo et al., 1991). Higher oxidation state shifts its main



**Fig. 3** Normalized sulfur K-edge XANES spectra of compounds with various sulfur oxidation states, in which the white line energy of elemental sulfur (0 state) was set at 2472.0 eV. They are pyrite (inorganic disulfide), elemental sulfur, glutathione oxidized (sulfide), glutathione reduced (thiol), dibenzothiophene (DBT, thiophene), dibenzyl sulfoxide, polyphenylene ether sulfone,  $\text{Fe}_2(\text{SO}_4)_3$  (sulfate). Indexes on the right refer to the sulfur states. All compounds were diluted (approximately to 0.3 wt.% sulfur) using boron nitride to reduce the self-absorption.

peak to higher energy. Compared to the white line energy of elemental sulfur, the oxidation index (and the main peak energy shift) of sulfur in different functional groups were reported as follows: pyrite  $-1$  ( $-0.5$  eV), organic sulfides  $+0.2$  ( $0.7$  eV), thiols and thioethers  $+0.6$  ( $1.1$  eV), thiophene  $+1.0$  ( $1.3$ – $1.8$  eV), sulfoxides  $+2.2$  ( $3.4$  eV), sulfone  $+4.3$  ( $7.5$  eV), sulfonate  $+5.2$  ( $8.7$  eV), sulfate  $+6.0$  ( $9.9$ – $10.1$  eV) (Huffman et al., 1991; Jalilvand, 2006; Vairavamurthy, 1998). XANES spectra of standard compounds with various sulfur oxidation states were detected in our experiments and these energy shifts were verified (Fig. 3). They also can be used to calibrate the sulfur white line energy of different sulfur compounds, in which the elemental sulfur white line energy was calibrated and set at 2472.0 eV in our experiments. The sulfur states (and the corresponding group types) in unknown samples can be identified and classified by the energy shifts of detected main peaks. In the actual experiments, peaks of close oxidation states were hard to separate. For example, the organic sulfides peak could not be separated both from the thiols peak and the thioethers peak. In that condition, the combined group types should best be exactly confirmed beforehand by other ways, or the fitted compounds are not necessarily the compounds actually present in the samples.

The IFEFFIT package primarily developed by Ravel and Newville (2005) was adopted to fit the peaks area in XANES spectra. Relative transition probabilities of different sulfur states were calibrated according the linear dependence of sulfur white line peak area on peak energy reported by Waldo et al. (1991). The overall estimated error for the fitting results produced by this method is typically within  $\pm 5\%$ – $10\%$  (Huffman et al., 1991).



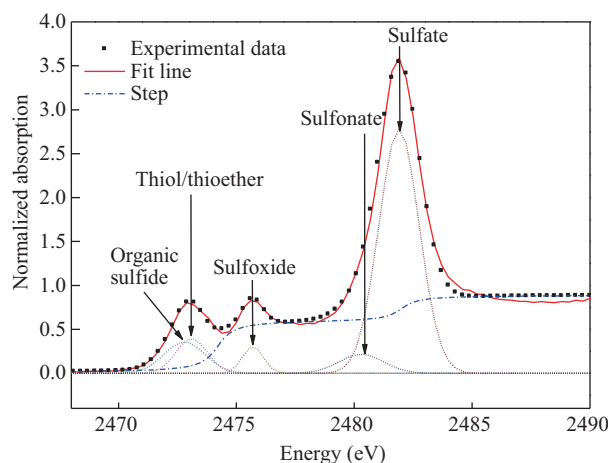
**Fig. 4** SR-XRF spectrum of mature CTLs sampled from BC (excitation energy was 3.2 keV).

## 2 Results and discussion

### 2.1 Sulfur concentration, speciation and accumulation in mature CTLs

Figure 4 shows a typical XRF spectrum of mature CTLs sampled from BC. The K-edge characteristic absorption peak of elements Si, P, S and Cl were clearly distinguished. Element concentration in samples was calculated by comparing with that in standard reference materials as external reference samples. The measurement indicated that sulfur concentration in mature CTLs sampled from different local fields varies dramatically from 766 to 1704 mg/kg, they are  $1704 \pm 141$ ,  $1647 \pm 136$ ,  $766 \pm 63$ ,  $899 \pm 75$ ,  $1215 \pm 100$  mg/kg, respectively for BC, XQ, WQ, SY and MU.

Figure 5 shows the least-square fitting of a normalized sulfur K-edge XANES spectrum of mature CTLs sampled from BC. Although each sulfur state should, in principle, have its own white line and arctangent step function, Huffman et al. (1991) found that the best results were obtained by using a two-step function approximation,



**Fig. 5** Least-square fitting of sulfur K-edge XANES spectrum of mature CTLs sampled from BC. The detecting step size were set at 0.3 eV between 2463 and 2500 eV while at 0.6 eV before 2463 eV and after 2500 eV during the characterization of all XANES spectra.

**Table 1** Sulfur speciation distribution in mature CTLs in diverse local fields (%)<sup>a</sup>

State	Functional group	BC	XQ	WX	SY	HS
+0.2	Organic sulfides	18	18	13	28	13
+0.6	Thiol/thioether	14	12	49	13	20
+2.2	Sulfoxides	6	10	2	8	8
+5.2	Sulfonate	5	7	8	9	3
+6.0	Sulfate <sup>b</sup>	57	54	17	42	56

<sup>a</sup> The estimated fitting error for organic sulfides, thiols/thioether and sulfoxide is about  $\pm(3-6)$  atom% S and for sulfonate and sulfate is about  $\pm(1-3)$  atom% S.

<sup>b</sup> Including inorganic sulfate and organic sulfate.

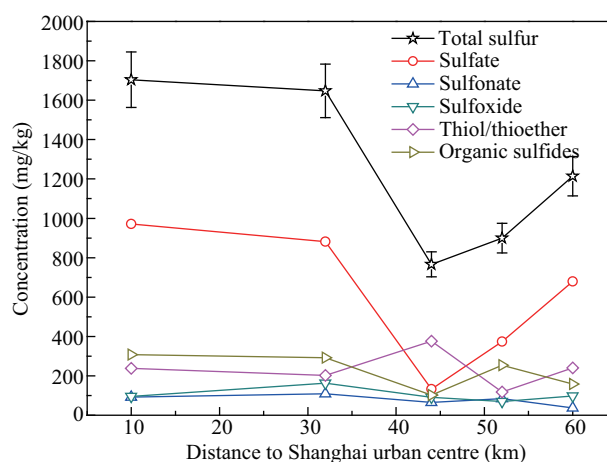
one step representing the transition to the continuum for unoxidized sulfur forms, the second representing the same feature for oxidized sulfur forms if significant. As reported by Leustek et al. (2000), organic sulfides, thiol, sulfoxide and sulfate were common sulfur compounds in plants. Jalilvand (2006) proposed that there were organic sulfides, thiol, thioether, sulfoxide, sulfonate and sulfate in plant leaf (magnolia leaf). According to these research and considering the peaks energy shifts, organic sulfides, thiols and thioethers (together as a thiol/thioether peak), sulfoxide, sulfonate and sulfate were proposed to fit the CTLs XANES spectra. The fitting results are shown in **Table 1**. Another fact is that  $\text{SO}_4^{2-}$  can be added to a hydroxyl group of an organic molecule especially when it is absorbed by plants (Leustek and Saito, 1999). Thereby, as suggested in a recent progress (Fors et al., 2011), the sulfate peak in plant samples can include both inorganic and organic sulfate such as ester sulfate.

Combined with the total sulfur (TS) concentration and the sulfur speciation distribution, concentration of each sulfur group in samples was calculated (**Fig. 6**). According to the oxidation index defined using the rule proposed by Vairavamurthy (1998), here, sulfur in plants is divided into reduced form (with a oxidation state  $< 6$ ) and oxidized form (with a oxidation state  $\geq 6$ ). Reduced sulfur (RS) mainly presents as organic sulfides, thiols and thioethers and is a major component of plant proteins. Oxidized sulfur (OS), i.e., sulfate sulfur, primarily as  $\text{SO}_4^{2-}$ , is mainly stored in vacuolar sap across the tonoplast or combined with organic molecules through sulfation (Cram, 1983; Leustek and Saito, 1999). Along the sampling routine from urban downtown (BC) to seashore (MU), the concentration of each kind of RS in CTLs seemed keeping nearly the same while OS varies dramatically. Correlations between TS and OS, RS and OS were analyzed and showed in **Fig. 7**. TS increases with OS increase following Eq. (1):

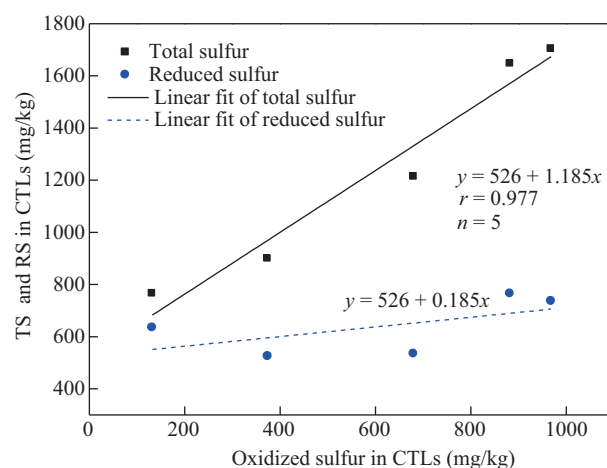
$$\text{TS} = 526 + 1.185\text{OS} \quad (r = 0.977) \quad (1)$$

where,  $r$  is the correlation coefficient.  
while RS follows Eq. (2)

$$\text{RS} = \text{TS} - \text{OS} = 526 + 0.185\text{OS} \quad (2)$$



**Fig. 6** Concentration of sulfur with different states in mature CTLs sampled from different fields.



**Fig. 7** Correlation between total sulfur (reduced sulfur) and oxidized sulfur in mature CTLs, in which  $r$  is the linear correlation coefficient and  $N$  is the number of data.

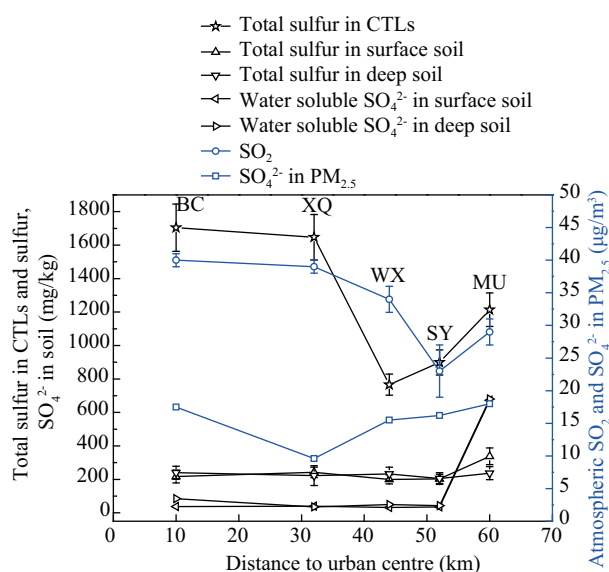
The growth factor in Eq. (1) of 1.185, which is close to 1, means that TS increases almost equivalently as OS increases. On the other hand, the growth factor in Eq. (2) of 0.185, which is close to 0, means that RS only increases slightly as OS increases. These suggest that in mature CTLs, only a little part of additional sulfur absorption is assimilated to RS form while most of it is accumulated as sulfate. The intercept (526) of the both formulas, which can be calculated by assigning zero to OS, means the sulfur concentration when there is no sulfate presented in the leaves. This may suggest that RS assimilation is sufficed to meet the requirement for growth at a balanced level of 526 mg/kg while sulfate is more or less accumulated at each site as the detected sulfur concentrations are all higher than this level. It also can be found that higher sulfur concentration, i.e., more sulfate accumulation occurred in urban downtown field (BC) and urban-rural joint (XQ), where have higher traffic road density than other fields (**Fig. 1**), indicating that higher pollution may lead to more sulfate accumulation in mature CTLs.

## 2.2 Correlation between sulfur concentration in mature CTLs and sulfur (or sulfur compounds) concentration in ambient sources at local fields

Generally, sulfur is mainly taken up actively by roots from soil to meet the plant nutrient requirement. However,  $\text{SO}_2$  and atmospheric particulates are also used by plant leaves from passive absorption as another sulfur source (Hell et al., 2010; Leustek and Saito, 1999).  $\text{SO}_2$  and atmospheric particulates with size less than  $1 \mu\text{m}$  can be absorbed and swallowed directly via stomas breathing (Uzu et al., 2010). Although particulates larger than  $1 \mu\text{m}$  can hardly pass the stomas directly, their soluble sulfate components can infiltrate into plant leaves when they deposit on the surface under moist condition. If sulfur supply such as the passive sulfur absorption by leaves is exceeding the metabolic demand for organic sulfur compounds synthesis, the sulfur absorption will result in accumulation as sulfate at the vacuole (Cram, 1983; Kaiser, 1989; Leustek et al., 2000).

**Table 2** shows the sulfur concentration in mature CTLs and the sulfur (or sulfur compounds) concentration in ambient sources from local environment along the sampling line. Their variation is illustrated in **Fig. 8**. Along the sampling routine, from urban centre to coast, sulfur concentration in CTLs decreased slightly from urban downtown (BC, 1704 mg/kg) to urban-rural joint (XQ 1647 mg/kg) and rapidly decreased to suburb (WX, 766 mg/kg), after that, it increased steadily to coastal suburb (SY, 899 mg/kg) and sped up to coast (MU, 1214 mg/kg). It presented a trend which was high in fields close to downtown and seashore but distinctly low in suburb.

Annual atmospheric  $\text{SO}_2$  concentration presented a similar variation and had a relatively strong correlation with the total sulfur concentration in mature CTLs. Actually, their coefficient of determination  $r^2$  in linear regression was up to 0.522, which also was the highest one among the coefficients of determination between the total sulfur concentration in mature CTLs and the sulfur (or sulfur compounds) concentration in ambient sources (the others were 0.096 for  $\text{SO}_4^{2-}$  in  $\text{PM}_{2.5}$ , 0.001 for  $\text{SO}_4^{2-}$  in surface soil, 0.0002 for  $\text{SO}_4^{2-}$  in deep soil, 0.036 for sulfur in surface soil and 0.038 for sulfur in deep soil, respectively). Although there was no obvious relativity between the concentration of  $\text{SO}_4^{2-}$  in collected atmospheric  $\text{PM}_{2.5}$  and the total sulfur concentration in mature CTLs on the whole



**Fig. 8** Variation of sulfur concentration in mature CTLs and sulfur-related sources concentration along the sampling line.

(as their coefficient of determination was only 0.038), they presented a simultaneous increase from WX to MU (**Fig. 8**), where the sampling sites were getting close to the seashore. On the contrast,  $\text{SO}_4^{2-}$  concentration both in deep and surface soil was almost constant except for an increase at the coast site (MU). Sulfur concentration both in deep and surface soil had a similar variation. These suggested that the dramatic sulfate accumulation in mature CTLs was mainly caused by atmospheric sources rather than soil and most probably dominated by  $\text{SO}_2$  emission. Although soil was the main source of sulfur for plants to meet their nutrient requirement, the atmospheric sulfur pollution had exceeded the capacity of CTLs and caused considerable passive accumulation. This could be harmful to plant growth. The brownish spots on some CTLs near the roads may reflect the deleterious effects of the redundant  $\text{SO}_2$  absorption.

Based on above results it can deduce that the sulfur concentration and sulfate accumulation in CTLs were closely linked with the atmospheric sulfur pollution in local fields. Shanghai is a mega city with a population over 23 million by 2010 (issued by Shanghai Municipal Statistics Bureau, <http://tjj.sh.gov.cn>) and most people concentrated in the city urban loop. A prosperous residential activity and heavy daily traffic is proceeding in urban place all the

**Table 2** Sulfur concentration in mature CTLs and sulfur sources concentration in local environment

Sampling site	Total sulfur in mature CTLs (mg/kg)	Annual atmospheric $\text{SO}_2$ ( $\mu\text{g}/\text{m}^3$ )	$\text{SO}_4^{2-}$ in atmospheric $\text{PM}_{2.5}$ ( $\mu\text{g}/\text{m}^3$ )	Water soluble $\text{SO}_4^{2-}$ in soil (mg/kg)		Total sulfur in soil (mg/kg)	
				Surface	Deep	Surface	Deep
BC	1704 ± 141	40 ± 1	17.5	37	85	217 ± 38	230 ± 40
XQ	1647 ± 136	39 ± 1	9.6	39	35	242 ± 31	223 ± 60
WX	766 ± 63	34 ± 2	15.5	32	50	201 ± 27	233 ± 41
SY	899 ± 75	23 ± 4	16.2	36	43	204 ± 27	205 ± 35
MU	1214 ± 100	29 ± 2	18.0	672	681	338 ± 50	237 ± 39



year round. Compared to suburb place, anthropogenic air pollution such as vehicle exhaust and industrial emissions was much more serious and will lead passive sulfur absorption and bioaccumulation in plant leaves more easily. This was also approved by the annual  $\text{SO}_2$  concentration variation along the sampling line; it was much higher in the downtown near the urban center and decreased in the fields near suburb (Fig. 8). The increase of sulfur concentration in mature CTLs from suburb place to coast suggested the long-term influence of marine sulfate/ $\text{SO}_2$  emission such as sea salt particles and maritime vessel exhaust from harbors and coastal shipping routes. Shanghai is a typical city of East Asian monsoon climate; the prevailing wind direction is east/southeast in most months (from March to October). As the East China Sea borders on Shanghai both in the east and south, sulfur sources generated on the sea surfaces can be introduced to inland environment following the eastern and southern wind. Sea salt generated from ocean surface is an important source of atmospheric sulfate particulates as sulfate is a dominant ingredient of sea salt and accounts for about 7.7% in weight (Riley and Chester, 1978). During the inland transport, sea salt particles, especially the coarse ones ( $> 2.5 \mu\text{m}$ ), almost deposit in the first several kilometers near the seashore (Meira et al., 2008; Rossknecht et al., 1973). It was approved by the dramatic increase of water soluble  $\text{SO}_4^{2-}$  in both surface and deep at MU, where is only 2 km away the coastline. However, a part of sea salt particles, especially the fine ones ( $\leq 2.5 \mu\text{m}$ ) can penetrate and decay to tens of kilometers inland (Gustafsson and Franzen, 2000; Rossknecht et al., 1973). Actually this was approved by the increase of  $\text{SO}_4^{2-}$  in  $\text{PM}_{2.5}$  from WX to MU. The sampling sites in suburb (WX), coastal suburb (SY) and coast (MU) in our experiments are within 20 km away from the southeast seashore, the atmospheric environment in these local fields is inevitably more significantly affected by the inland transport of sea salt particles. It consequently provided more chance for the passive absorption of sulfate particulates by CTLs and could lead to more sulfate accumulation. Meanwhile, Shanghai is a world-class shipping center, it has the largest deep-water port of the world (Yangshan Deep Water Port), which locates in the East China Sea, about 30 km away from the southeast seashore of Shanghai. A mass of maritime vessel exhaust of  $\text{SO}_2$  surrounds the harbour and will spread to the fields near the seashore all over the year. This made the annual  $\text{SO}_2$  concentration in coast site (MU) higher than that in coastal suburb site (SY) obviously thus could lead to the increase of sulfur concentration in mature CTLs near the seashore to some extent.

### 3 Conclusions

Synchrotron radiation XRF and XANES are successfully used to study the sulfur concentration, speciation and

bioaccumulation in mature CTLs. An amount of 766–1704 mg/kg sulfur concentration was detected in mature CTLs sampled from five different local fields in Shanghai. The sulfur compounds detected were distinguished mainly as organic sulfides, thiol/thioether, sulfoxide, sulfonate and sulfates. A high degree of linear correlation was found between total sulfur concentration and sulfate concentration in CTLs. The increase of sulfate content directly led to increase of total sulfur concentration while other sulfur compounds relatively remained constant regardless of the sulfate increase. Sulfur was accumulated as sulfate in mature CTLs as the local sulfur pollution exceeding the metabolic demand for plant nutrient requirement. The sulfate accumulation was mainly caused by atmospheric sources such as  $\text{SO}_2$  and airborne sulfate particulates rather than soil. CTLs can be used as effective biomonitoring indicators of the long-term atmospheric sulfur pollution accumulation in local fields. Serious sulfate accumulation in downtown represented the traffic exhaust and industrial emissions while the sulfate accumulation near the seashore was probably due to the long-term deposition of sea salt particles on leaf surface and maritime vessel exhaust from harbours and coastal ships.

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### References

- Bao L M, Zhang G L, Zhang Y X, Li Y, Lin J, Liu W et al., 2009. Transfer characterization of sulfur from coal-burning emission to plant leaves by PIXE and XANES. *Chinese Physics C (HEP & NP)*, 33(11): 1010–1015.
- Beckett K P, Freer-Smith P H, Taylor G, 1998. Urban woodlands: their role in reducing the effects of particulate pollution. *Environmental Pollution*, 99(3): 347–360.
- Boucher O T, Lonmann U, 1995. The sulfate-CCN-cloud albedo effect. *Tellus B*, 47(3): 281–300.
- Butler O T, Cairns W, Cook J M, Davidson C M, 2011. Atomic spectrometry update. Environmental analysis. *Journal of Analytical Atomic Spectrometry*, 26(2): 250–286.
- Butler O T, Cook J M, Davidson C M, Harrington C F, Miles D L, 2009. Atomic spectrometry update. Environmental analysis. *Journal of Analytical Atomic Spectrometry*, 24(2): 131–177.
- Chan C K, Yao X H, 2008. Air pollution in mega cities in China. *Atmospheric Environment*, 42(1): 1–42.
- Cicek A, Koparal A S, 2004. Accumulation of sulfur and heavy metals in soil and tree leaves sampled from the surroundings of Tunçbilek Thermal Power Plant. *Chemosphere*, 57(8): 1031–1036.

- Cowling D W, Jones L H P, Lockyer D R, 1973. Increased yield through correction of sulfur deficiency in ryegrass exposed to sulphur dioxide. *Nature*, 243(5408): 479–480.
- Cram W J, 1983. Sulphate accumulation is regulated at the tonoplast. *Plant Science Letters*, 31(2-3): 329–338.
- Fors Y, Jalilehvand F, Sandstrom M, 2011. Analytical aspects of waterlogged wood in historical shipwrecks. *Analytical Sciences*, 27(8): 785.
- Frank P, Hedman B, Carlson R M K, Tyson T A, Roe A L, Hodgson K O, 1987. A large reservoir of sulfate and sulfonate residues within plasma cells from ascidia ceratodes, revealed by X-ray absorption near-edge structure spectroscopy. *Biochemistry*, 26(16): 4975–4979.
- Gustafsson M E R, Franzen L G, 2000. Inland transport of marine aerosols in southern Sweden. *Atmospheric Environment*, 34(2): 313–325.
- Hell R, Khan M, Wirtz M, 2010. Plant cell monographs. In: Cell Biology of Metals and Nutrients (Hell M, ed.). Springer Berlin, Heidelberg. 243–279.
- Huffman G P, Mitra S, Huggins F E, Shah N, Vaidya S, Lu F L, 1991. Quantitative analysis of all major forms of sulfur in coal by X-ray absorption fine structure spectroscopy. *Energy & Fuels*, 5(4): 574–581.
- Jalilehvand F, 2006. Sulfur: not a “silent” element any more. *Chemical Society Reviews*, 35(12): 1256–1268.
- Kaiser G, Martinoia E, Schröppel G, Heber U, 1989. Active transport of sulfate into the vacuole of plant cells provides halotolerance and can detoxify SO<sub>2</sub>. *Journal of Plant Physiology*, 133(6): 756–763.
- Last F T, 1982. Effects of atmospheric sulphur compounds on natural and man-made terrestrial and aquatic ecosystems. *Agriculture and Environment*, 7(3-4): 299–387.
- Leustek T, Martin M N, Bick J A, Davies J P, 2000. Pathways and regulation of sulfur metabolism revealed through molecular and genetic studies. *Annual Review of Plant Physiology and Plant Molecular Biology*, 51(1): 141–165.
- Leustek T, Saito K, 1999. Sulfate transport and assimilation in plants. *Plant Physiology*, 120(3): 637–643.
- Meira G R, Andrade C, Alonso C, Padaratz I J, Borba J C, 2008. Modelling sea-salt transport and deposition in marine atmosphere zone – A tool for corrosion studies. *Corrosion Science*, 50(9): 2724–2731.
- Ravel B, Newville M, 2005. ATHENA, ARTEMIS, HEPHAESTUS: data analysis for X-ray absorption spectroscopy using IFEFFIT. *Journal of Synchrotron Radiation*, 12(4): 537–541.
- Riley J P, Chester R, 1978. Chemical Oceanography (2nd ed.). Academic Press, London.
- Rossknecht G F, Elliott W P, Ramsey F L, 1973. The size distribution and inland penetration of sea-salt particles. *Journal of Applied Meteorology*, 12(5): 825–830.
- Sandström M, Jalilehvand F, Persson I, Gelius U, Frank P, Hall-Roth I, 2002. Deterioration of the seventeenth-century warship Vasa by internal formation of sulphuric acid. *Nature*, 415(6874): 893–897.
- Starr F, Starr K, Loope L, 2003. United States Geological Survey –Biological Resources Division Haleakala Field Station, Maui, Hawaii.
- Szczepaniak K, Biziuk M, 2003. Aspects of the biomonitoring studies using mosses and lichens as indicators of metal pollution. *Environmental Research*, 93(3): 221–230.
- Tsai I C, Chen J P, Lin P Y, Wang W C, Isaksen I S A, 2010. Sulfur cycle and sulfate radiative forcing simulated from a coupled global climate-chemistry model. *Atmospheric Chemistry and Physics*, 10(8): 3693–3709.
- Uzu G, Sobanska S, Sarret G, Muñoz M, Dumat C, 2010. Foliar lead uptake by lettuce exposed to atmospheric fallouts. *Environmental Science & Technology*, 44(3): 1036–1042.
- Vairavamurthy A, 1998. Using X-ray absorption to probe sulfur oxidation states in complex molecules. *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*, 54(12): 2009–2017.
- Waldo G S, Carlson R M K, Moldowan J M, Peters K E, Pennerhahn J E, 1991. Sulfur speciation in heavy petroleum: Information from X-ray absorption near-edge structure. *Geochimica et Cosmochimica Acta*, 55(3): 801–814.
- Ward P L, 2009. Sulfur dioxide initiates global climate change in four ways. *Thin Solid Films*, 517(11): 3188–3203.
- Zhang Y X, Cao T, Atsuo I, Cao Q C, Lou Y X, Zhang G L et al., 2009. Study of moss as air pollution monitor by SRXRF technique. *Chinese Science Bulletin*, 54(17): 2987–2990.

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