

Biomass burning contributions to urban PM_{2.5} along the coastal lines of southeastern China

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ABSTRACT

Levoglucosan (LG), water soluble organic carbon (WSOC) and potassium (K^+), and the light absorption at 365 nm (Abs_{365}) of the extracted WSOC are measured in PM_{2.5} samples collected from November 2011 to July 2013 at four coastal urban sites in southeast China (Fuzhou, Putian, Quanzhou and Xiamen). These species are markers of biomass burning and used to determine the contributions of biomass burning to the PM_{2.5} burden in these locations. LG and WSOC concentrations exhibited a clear seasonal pattern, with a large enhancement in winter and spring and a minimum in summer, and annual means across all sites of $59.2 \pm 46.8 \text{ ng m}^{-3}$ and $2.69 \pm 1.21 \text{ } \mu\text{g C m}^{-3}$, respectively. The distinctive seasonal patterns of LG and WSOC are more explained by the East Asian monsoon than the upwind varying emission sources according to the HYSPLIT backward trajectories and MODIS fire spots. Observations produced significant correlation (at the $p < 0.01$ level) between LG and non-sea salt K^+ (nss- K^+) at each site, but the correlations exhibited no clear seasonal trend. The LG/nss- K^+ ratios ranged from 0.03 ± 0.01 to 0.24 ± 0.13 which lay within the limits for the crop residues and/or grass combustion smoke. Stronger correlations were found between WSOC or Abs_{365} and sulphate than between WSOC and LG. This observation is consistent with the fact that biomass burning is a less important contributor to WSOC and/or brown carbon than is secondary organic aerosol formation and oxidation. The average relative contributions of biomass burning to OC and WSOC in PM_{2.5} were 8.3 and 15.2 %, respectively, estimated by the measured LG to OC and WSOC (LG/OC and LG/WSOC) ratios in comparison to literature-derived LG/OC and LG/WSOC values for biomass burning smoke. Using the reported conversion factor of LG to PM_{2.5} for crop straw burning smoke, the LG-estimated PM_{2.5} contributions from biomass burning exhibited minimum values in summer and higher values in winter and spring. Positive Matrix Factorisation was used to analyse PM_{2.5} sources, and seven major source factors including biomass burning emissions were resolved. The biomass burning sources contributed 3.75–8.08 % to PM_{2.5} mass as an annual average while exhibiting a seasonal variability similar to and higher than those of LG-estimated contributions. These results demonstrate that the contribution from biomass burning smoke to PM_{2.5} – while small – is non-negligible, especially in the winter and spring sampling periods, that is, northeastern monsoon season.

Keywords: biomass burning, WSOC, levoglucosan, source apportionment, southeastern China

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1. Introduction

China is facing air pollution problems with $\text{PM}_{2.5}$ due to its rapid growth of economy and urbanisation, especially in regions such as Beijing-Tianjin-Hebei (BTH), the Yangtze River Delta (YRD) and the Pearl River Delta (PRD) (www.mep.gov.cn). Motor vehicles, fossil-fuel combustion, industrial processes, biomass burning and soil dusts are suggested to be the main sources of $\text{PM}_{2.5}$ in China (Song et al., 2006; Tao et al., 2013, 2014; Zhang et al., 2013a, 2013b). Meanwhile, biomass burning which is associated with field burning of biomass and agricultural wastes has drawn more concern due to the strengthening of vehicle emission standards and the use of power plant scrubbers as well as the increase of crop yield and less demand as fuel with the rapid economic development in rural areas (Yan et al., 2006; Lu et al., 2011; Wang et al., 2013). According to the inventory studies by Lu et al. (2011), biomass burning contributions accounted for about 40 and 70 % of elemental carbon (EC) and primary organic carbon (OC), respectively, during 2006–2010 in China. Moreover, biomass burning is suggested to be one of the major contributors to the uncertainty in air quality modelling (Tian et al., 2009). Studies to characterise the organic composition of biomass burning aerosols are also motivated by the climate change (Andreae and Rosenfeld, 2008), light absorption (Chen and Bond, 2010) and human health effects (Naeher et al., 2007) of this aerosol class. Therefore, estimation of biomass burning emissions is urgently needed for quantifying this contribution and improving air quality.

National-, regional- or urban-scale biomass burning emission inventories have been made in China (Streets et al., 2003; Yan et al., 2006; He et al., 2011; Zhang et al., 2013b). However, there still exist large uncertainties in biomass burning emission estimates due to the incomplete and incomprehensive collection of activity data and variable emission factors depending on moisture, type of vegetation and burning conditions (smouldering or flaming), wind direction and other meteorological factors (Streets et al., 2003; Bond et al., 2004; He et al., 2011; Oanh et al., 2011; Zhang et al., 2013a, 2013b). Since these difficulties exist in the establishment of a reliable emission inventory for biomass burning, individual source tracers including levoglucosan (1,6-anhydro- β -D-glucopyranose; LG) and water soluble K^+ and receptor modelling methods are often used to assess biomass burning contributions to $\text{PM}_{2.5}$ (Zhang et al., 2008, 2010; Sang et al., 2011; Giannoni et al., 2012; Reche et al., 2012; Mkoma et al., 2013; Ho et al., 2014; Zhu et al., 2015). LG, which is produced exclusively by the pyrolysis of cellulose and hemicellulose, is the most commonly used organic tracer for biomass burning aerosols (Puxbaum et al., 2007). This compound is relatively stable in the atmosphere with no decay over 10 d in very acidic conditions

(Fraser and Lakshmanan, 2000). Little loss of LG in biomass burning soot exposure to UV radiation and high concentrations of ozone was observed (Latif et al., 2011). In addition, the modelled and measured LG levels at a remote monitoring site in the Pacific Ocean were comparable, suggesting no considerable degradation during transport from the Asian continent within 4–5 days (Mochida et al., 2010). Although K^+ is also proposed as a conventional tracer for biomass burning, it is not a unique product of cellulose combustion as it can also be emitted from sea salt, soil dust, meat cooking, coal combustion and abrasion of leaf surfaces (Simoneit, 2002; Duvall et al., 2008).

However, chamber studies revealed that LG in aqueous solution can experience degradation in the presence of strong oxidants (e.g. OH radical) (Hennigan et al., 2010; Hoffmann et al., 2010; Lai et al., 2014). Nevertheless, LG is a preferred tracer for semi-quantitative or conservative quantitative assessment of biomass burning influence due to its specificity, high emission factor and high level in aerosols. Zhang et al. (2007) reported LG to organic carbon $(\text{LG}/\text{OC})_{\text{BB}}$ and $\text{PM}_{2.5}$ $(\text{LG}/\text{PM}_{2.5})_{\text{BB}}$ ratios ranging from 5.4 to 11.8 % (av. 8.2 %) and 2.98 to 6.47 % (av. 4.52 %), respectively, in $\text{PM}_{2.5}$ smoke from chamber combustion experiments of three major types of cereal straw (corn, wheat and rice) in China. The $(\text{LG}/\text{OC})_{\text{BB}}$ ratio is comparable to the $(\text{LG}/\text{OC})_{\text{BB}}$ ratios in the range of 8.0–8.2 % (av. 8.14 %) calculated from the emission factors for various types of biomass burning in Andreae and Merlet (2001) and the ratios of 0.5–9.97 % derived from rice and wheat burning smoke in the chamber (Sullivan et al., 2008), but much lower than the reported value (16.8 %) for field open burning $\text{PM}_{2.5}$ smoke of rice straw (Oanh et al., 2011). In addition, different removal rates by dry and wet deposition between LG and OC and secondary formation of OC during transport in the atmosphere can also influence the LG/OC ratio in ambient aerosol significantly (Zhang et al., 2012; Cheng et al., 2013). The contribution of biomass burning to OC using the simplified tracer-based method varied widely (6.5–52 %) depending on the biomass burning activities (Zhang et al., 2008, 2012; Sang et al., 2011; Mkoma et al., 2013; Ho et al., 2014; Zhu et al., 2015). Based on the Positive Matrix Factorisation (PMF) using concentrations of chemical species including LG and K^+ , biomass burning contributions accounted for 3–24 % of the annual $\text{PM}_{2.5}$ with lowest values in summer and higher in other seasons in Beijing (Cheng et al., 2013; Zhang et al., 2013c), Chengdu (Tao et al., 2014) and the southeastern US (Zhang et al., 2010). In addition, the impact of biomass burning could be assessed by using outdoor fire counts and HYSPLIT backward trajectories (Zhang et al., 2010, 2012; Sang et al., 2011; Mkoma et al., 2013; Ho et al., 2014; Zhu et al., 2015).

Fujian province is on China's southeastern coast and lies in-between the YRD and PRD (Fig. 1). Its climate is mainly dominated by the East Asian monsoon characterised with a southwest wind prevailing in summer and northeast wind prevailing in winter. The province is mostly mountainous and is traditionally described to be 'Eight parts mountain, one part water and one part farmland'. Thus, biomass burning activities such as crop residues in Fujian are not as common as those in northern and eastern China. According to our previous investigation, the common types of biomass burning in the mountainous rural areas of Fujian are rice straw and sugarcane leaves (in-field burning) and woody fuel (domestic burning), which are similar to those reported in PRD, China (Zhang et al., 2013b). However, emissions from wood-fuel boilers to provide warm water in prawn farming in cold season and plastic recycling to melt waste plastics, in-field grass burning in vegetable plots, and domestic joss paper and incense burning can influence carbonaceous aerosol inventory more significantly in coastal urban areas. Furthermore, Fujian's coastal area is a perfect region to capture the signals of biomass burning from the upwind sources during the northeast monsoon and southwest monsoon periods. The seasonal patterns of PM_{2.5} and its major components from the four sites at Xiamen, Quanzhou, Putian and Fuzhou cities along the coastal line coincided well with the monsoon cycles during a 2-yr monitoring period (Wu et al., 2015a). Organic matter ($= 1.6 \times \text{OC}$) was suggested to be the largest contributor, accounting for 30.2 % of PM_{2.5}, followed by SO_4^{2-} (19.8 %), NH_4^+ (10.6 %) and NO_3^- (9.7 %). However, the estimate of biomass burning contributions to OC was limited due to the lack of LG data in PM_{2.5}. Because the major part of OC from biomass burning was found to be water-soluble, water-soluble organic carbon (WSOC) could be used to evaluate the influence of biomass burning on carbonaceous aerosol. WSOC accounted for a large fraction (20–70 %) of aerosol OC (Feng et al., 2006; Park and Cho, 2011) and has been recognised as an important fraction of OC due to its potential influence on climate change (Ervens et al., 2005). Sources of WSOC have been shown to be complex including primary sources such as biomass burning (Zhang et al., 2010) and secondary sources such as atmospheric reactions (Hecobian et al., 2010). Brown carbon or humic-like substances (HULIS) in WSOC, which display relatively more absorbance in shorter wavelength regions and less in the longer wavelength regions, have been proved to be an important light-absorbing substance (Graber and Rudich, 2006). Because brown carbon typically absorbs at short wavelength, the light absorbance at 365 nm (Abs_{365}) was often chosen for evaluation of light absorbing properties and comparison with that of black carbon (Andreae and Gelencser, 2006; Kirillova et al., 2014; Du et al., 2014a).

HULIS, which has been shown to behave like surface active agents or surfactants, can reduce the surface tension in a droplet and enhance the formation of cloud condensation nuclei (CCN) in the atmosphere (Taraniuk et al., 2008). Thus, the spectroscopic characteristics of WSOC together with concentrations of chemical marker species can be used as PMF model input dataset (Zhang et al., 2010).

In order to attain a better understanding of the organic carbonaceous aerosols, we analysed LG as well as WSOC and the light absorption spectra at 365 nm (Abs_{365}) of WSOC and K^+ in PM_{2.5}. Moreover, the biomass burning contributions to OC and WSOC as well as PM_{2.5} were estimated using a molecular tracer technique. Finally, the PMF model in conjunction with HYSPLIT backward trajectory analysis and MODIS fire counts were employed to quantify average mass contributions from biomass burning and other sources of urban PM_{2.5} along the coastal lines of southeastern China.

2. Experimental methods

2.1. Sample location and collection

Four coastal urban sites along the coastal line in southeastern China were selected to collect PM_{2.5} in this region, namely (i) Hongwen Primary School in Xiamen (XM) (24°28'37" N, 118°09'07" E), (ii) Quanzhou City Environmental Protection Bureau in Quanzhou (QZ) (24°53'52" N, 118°35'50" E), (iii) Xiuyu District Campus in Putian (PT) (25°19'17" N, 119°06'05" E) and (iv) Fujian Normal University in Fuzhou (FZ) (26°04'31" N, 119°18'55" E; Fig. 1). The samples were collected in daytime (8:00–18:00) and nighttime (18:00–8:00) over a period of 15 d in November, January, April and June or July between November 2011 and July 2013. The sampling and major components analysis including element, water soluble ions and carbon were described in detail in Wu et al. (2015a). In this study, halves of the daytime and nighttime filters were merged into one sample for LG, WSOC and UV-Vis spectrum analysis. The values of precipitation for the eight sampling periods near the four sites were derived from Weather & Climate website (www.weatherandclimate.info).

2.2. Chemical characterisation

2.2.1. Levoglucosan. The analytical method for LG was modified from Wang et al. (2007) and Zhang et al. (2008). Half of the merged sample was cut and sonicated for 45 min in a 30-mL mixture of dichloromethane/methanol (2:1; v/v). After extraction, the solvents were concentrated to ~1 mL using a rotary evaporator. The extract was then

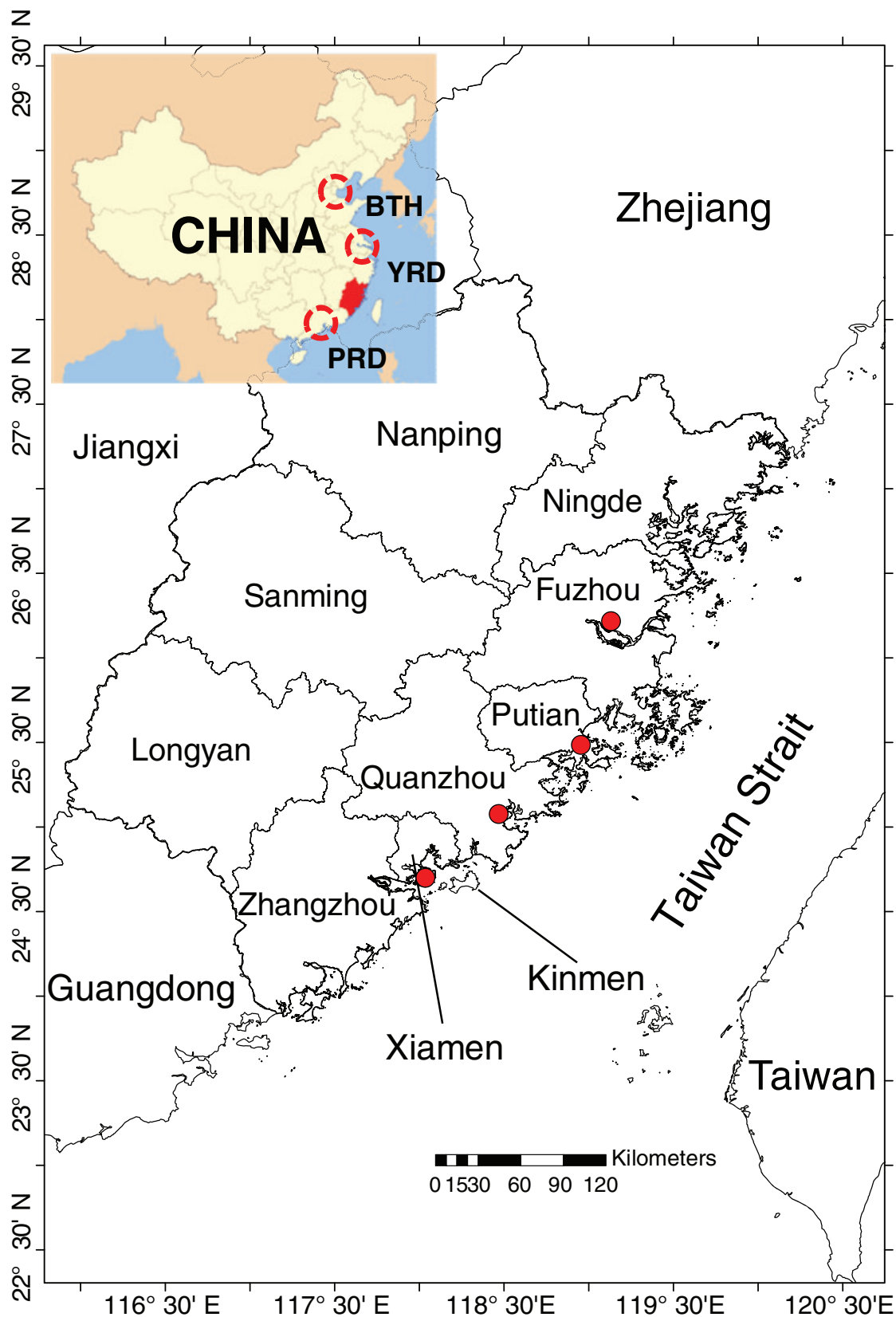


Fig. 1. Administrative division of Fujian province and location of the four sampling sites (solid point in Xiamen, Quanzhou, Putian and Fuzhou).

filtered through 0.45 μm PTFE Acrodisc syringe filter (13 mm, Pall Co., Ltd, Beijing, China) into a concentrator tube. Nitrogen blow down was used to remove most solvents and the remaining filtrate was pipetted into a 300- μL auto-sampler vial insert and concentrated to dryness under the stream of N_2 . Fifty microliters of *N,O*-bis-(trimethylsilyl) trifluoroacetamide (BSTFA) containing 1 % trimethylsilyl chloride (TMCS) and 10 μL pyridine were added to the vial insert, which was then rapidly capped and allowed to react for 3 h in an oven at 70 °C. Immediately before GC–MS analysis, the solution containing derivatised LG was evaporated to dryness under a continuous flow of N_2 and redissolved in 100 μL of hexane containing 10 ng internal standard ($\text{C}_{25}\text{D}_{50}$) for injection volume effects and variations in the GC–MS detector response.

An aliquot (1 μL) of silylated extract was injected into the GC–MS system in the splitless mode (Agilent 6890GC-5973 MSD). A HP-5 MS capillary column (30 m $\times$ 0.25 mm i.d. $\times$ 0.25 μm film) was used to separate these silylated products with an oven temperature program from 65 °C (hold for 2 min) to 300 °C (hold for 20 min) at a rate of 6 °C min^{-1} , and helium as carrier gas. Injector and GC–MS interface temperatures were 250 °C and 280 °C, respectively. LG was identified by comparison with mass spectrum (NIST 02) and retention time of standards and quantified using selective ion monitoring (SIM) with ionic fragment of $m/z = 204$. The calibration curves for LG were linear over a wide concentration range (25–2500 ng mL^{-1}) with the determination coefficient (R^2) greater than 0.99. The limit of detection (LOD) was 1.1 ng per sample calculated as three times the standard deviation of six blank measurements, which corresponded to 0.03 ng m^{-3} in the atmosphere. There were no significant differences between field blanks and procedure blanks and the contents of the target compound in blanks were much lower than those in the ambient samples, so blank correction caused no noticeable change in the data.

The extraction efficiency of the LG target compound was determined by comparing the GC/MS response of the doped filters using LG standard solutions with those of the corresponding standard solutions that had not been spiked onto filters. The extraction recovery for LG was $94.6 \pm 5.3\%$ ($n = 7$). The ambient LG concentrations presented here were corrected by field blanks and extraction recovery.

2.2.2. WSOC and light absorption properties. The remaining half of the merged filter was extracted with 20 mL of Milli-Q water (Millipore Co., Ltd, Shanghai, China) via 45 min sonication. The liquid extract was then filtered into a 50 mL comparison tube using a 0.45 μm PTFE Acrodisc syringe filter (13 mm, Pall Co., Ltd, Beijing, China). WSOC in the extract (10 mL) was quantified using a Total Organic

Carbon Analyzer (TOC-VCPH, Shimadzu Co., Ltd, Beijing, China). The instrument was calibrated using a series of sucrose solutions every day through the analysis period. Another aliquot (10 mL) of the extract was used for the light absorbance measurement at 365 nm by a UV–Vis spectrophotometer (HP8453, Agilent Technologies Co., Ltd, Shanghai, China), using a wavelength range of 190–800 nm. In order to compare easily among samples, the absorption value was corrected by sampling volume and given as Abs_{365} .

2.3. MODIS fire counts and HYSPLIT backward trajectories

Fire counts detected by NASA satellites (Aqua and Terra) have been widely used to identify the geographical hot spots of biomass burning activities with a horizontal resolution of $1 \times 1 \text{ km}^2$ pixel resolution (Sang et al., 2011; Zhang et al., 2010, 2012, 2015; Liu et al., 2013; Ho et al., 2014). In this study, burning activities in Fujian were shown by fire spots, and the sum of fire counts were obtained from FIRMS Web Fire Mapper (<https://firms.modaps.eosdis.nasa.gov/firemap/>) operated by NASA of the United States for each sampling period (Supplementary Fig. 1). In order to investigate the influence of biomass burning in different parts around Fujian on LG measured at the four coastal urban sites in Fujian, 7-d backward air mass trajectories starting at 100 m above ground level at 8:00 local time were calculated every day in each sampling period using HYSPLIT4 downloaded from <http://ready.arl.noaa.gov/HYSPLIT.php>. Six-hourly archived Global Data Assimilation System ($1^\circ \times 1^\circ$) from the National Centers for Environmental Prediction were used as meteorological data. The meteorological data were downloaded from <ftp://arlftp.arl.noaa.gov/pub/archives/gdas1>. Cluster analysis was then applied to merge trajectories that are near each other into one cluster. The final cluster number ranged from 2 to 5 in order to obtain an appropriate solution (Supplementary Fig. 2).

2.4. Source apportionment analysis

LG is a better tracer of biomass burning than non-sea salt K^+ (nss- K^+) which has additional sources including soil dust, meat cooking and fireworks but has different emission factors depending on the type of vegetation and burning conditions (Wang et al., 2007; Zhang et al., 2010; Cheng et al., 2014). The ratio of LG to K^+ (LG/K^+)_{BB} in biomass burning smoke therefore ranged in an order of magnitude depending on the biomass burning types and modes (Zhang et al., 2013a). In this study, we used the ratios of LG to OC [(LG/OC)_{BB}, av. 8.2 %], WSOC [(LG/WSOC)_{BB}, av. 17.0 %] and PM_{2.5} [(LG/PM_{2.5})_{BB}, av. 4.5 %] as reference values in biomass burning smoke to estimate biomass burning source contributions (Zhang et al., 2007,

2008, 2010; Sang et al., 2011; Ding et al., 2013; Ho et al., 2014). In addition, source apportionment analysis was also carried out by EPA-PMF (v5.0) receptor model (Zhang et al., 2010). In our study, PMF runs were performed using a total of 21 parameters including 20 chemical species (Si, Al, Ca, Ti, Fe, Mn, V, Ni, Pb, Zn, Na⁺, Cl⁻, nss-K⁺, nss-SO₄²⁻ (non-sea salt SO₄²⁻), NO₃⁻, NH₄⁺, OC, EC, LG and WSOC) and light absorption at 365 nm (Abs₃₆₅) (Zhang et al., 2010; Reche et al., 2012). The EPA-PMF (v5.0) model can be found on US EPA website (<http://www.epa.gov/heasd/research/pmf.html>). The data of inorganic ions and elements, OC and EC have been already reported by Wu et al. (2015a). The nss-K⁺ and nss-SO₄²⁻ were calculated from the measured K⁺ and SO₄²⁻ minus the corresponding sea-salt fraction of K⁺ and SO₄²⁻, that is, $\text{nss-K}^+ = \text{K}^+ - 0.036 \times \text{Na}^+$, $\text{nss-SO}_4^{2-} = \text{SO}_4^{2-} - 0.252 \times \text{Na}^+$ (Seinfeld and Pandis, 2006). The number of base model runs was 20 and the base run with the minimum Q value (goodness-of-fit parameter) was retained. Numerous PMF runs were performed with 5–8 factors. Based on Q-values and physical interpretation of the solution, a seven-factor solution (including biomass burning, fuel oil, vehicular emission, secondary inorganic aerosol, crustal dust, metallurgical emission/mixed source of metallurgical emission and secondary sulphate aerosol and secondary aerosol/waste incineration emission) was selected as the most reasonable one.

3. Results and discussion

3.1. Seasonal variations of LG and nss-K⁺

Figure 2 presents the temporal variations in the concentrations of LG in PM_{2.5} and rainfall at the four coastal urban sites in Fujian province during the eight sampling periods. The LG concentrations ranged from 1.26 to 283 ng m⁻³, with minimum seasonal means in summer (21.5 ± 14.5 ng m⁻³) and maximum means in spring (83.5 ± 58.1 ng m⁻³) and annual mean of 59.2 ± 46.8 ng m⁻³ (Supplementary Table 1). In general, LG concentrations obtained in this study were much lower than those reported in megacities such as Guangzhou and Beijing and biomass burning impacted regions such as Chiang Mai in Thailand, Morogoro in Tanzania, and Florence in Italy and locations in the southeastern US (references in Supplementary Table 1). However, results of this study were considerably higher with respect to those measured on Okinawa Island in the northwestern Pacific and at a remote station on the Tibetan Plateau (Supplementary Table 1), suggesting minor impact from biomass burning in these regions. The seasonal variations of LG as seen from Fig. 2 are consistent with results from other studies dealing with LG measurements (Zhang et al., 2008, 2010; Giannoni et al., 2012; Zhu et al.,

2015) and similar to those of polycyclic aromatic hydrocarbons, dicarboxylic acids and other major water soluble ions associated with particulate matter observed in this region in different years (Hsu et al., 2010; Xu et al., 2012a; Li et al., 2013; Wu et al., 2014, 2015a, 2015b). As discussed previously, the seasonal pattern can be partly explained by the transport of air masses from upwind sources under the influence of the East Asian monsoons.

During the summer sampling periods, the prevailing wind direction is southeast–southwest (Supplementary Fig. 2) and the input of clean marine air masses transported over South China Sea could more readily dilute the PM_{2.5} along the coastal region (Wu et al., 2015a, 2015b). Moreover, fewer biomass burning activities were observed in the upwind regions of Philippines during the summer sampling periods, as seen from MODIS fire counts (Supplementary Fig. 1). As mentioned earlier, MODIS fire count data can be used as a measure of field biomass burning. As shown in Fig. 3, the fire counts variation in Fujian didn't match the patterns of LG and nss-K⁺. The post-harvest wheat or rape straw burning emissions in summer sampling period in southeastern China move northward driven by summer monsoon and have little influence on ambient LG measured at the coastal urban sites in Fujian. Consequently, local small scale combustion activities such as agricultural residues and joss paper/incense burning should be the major sources and lower levels of LG were expected in summer due to the favourable dilution, diffusion and photodegradation conditions.

Analogously, the impact of biomass burning on ambient atmospheric pollutants should be low in winter as the minimum fire spots were detected (Supplementary Fig. 1 and 3). However, much higher concentrations of LG were observed in winter in comparison with those in summer, which deviated too much from fire counts shown in Fig. 3. According to the LG concentrations obtained in previous studies listed in Supplementary Table 1, higher levels were generally found in the cold season due to the use of biofuels for domestic heating than those in the warm season in places like Beijing, southeastern US and Florence. Zhu et al. (2015) reported that the highest concentrations of LG in Okinawa aerosols collected from 28 December 2010 to 4 January 2011 were not consistent with fire spots detected in the upwind region of northeastern China and Russia, implying that biomass burning was likely in the form of indoor biofuel burning. In this study, however, the high levels of LG measured in winter should be more attributed to the stable atmospheric stratification resulting accumulation than to the burning activities in the upwind regions of northern and eastern China. Although the emission of LG from the indoor biofuel burning activities could significantly enhance for indoor heating in the upwind

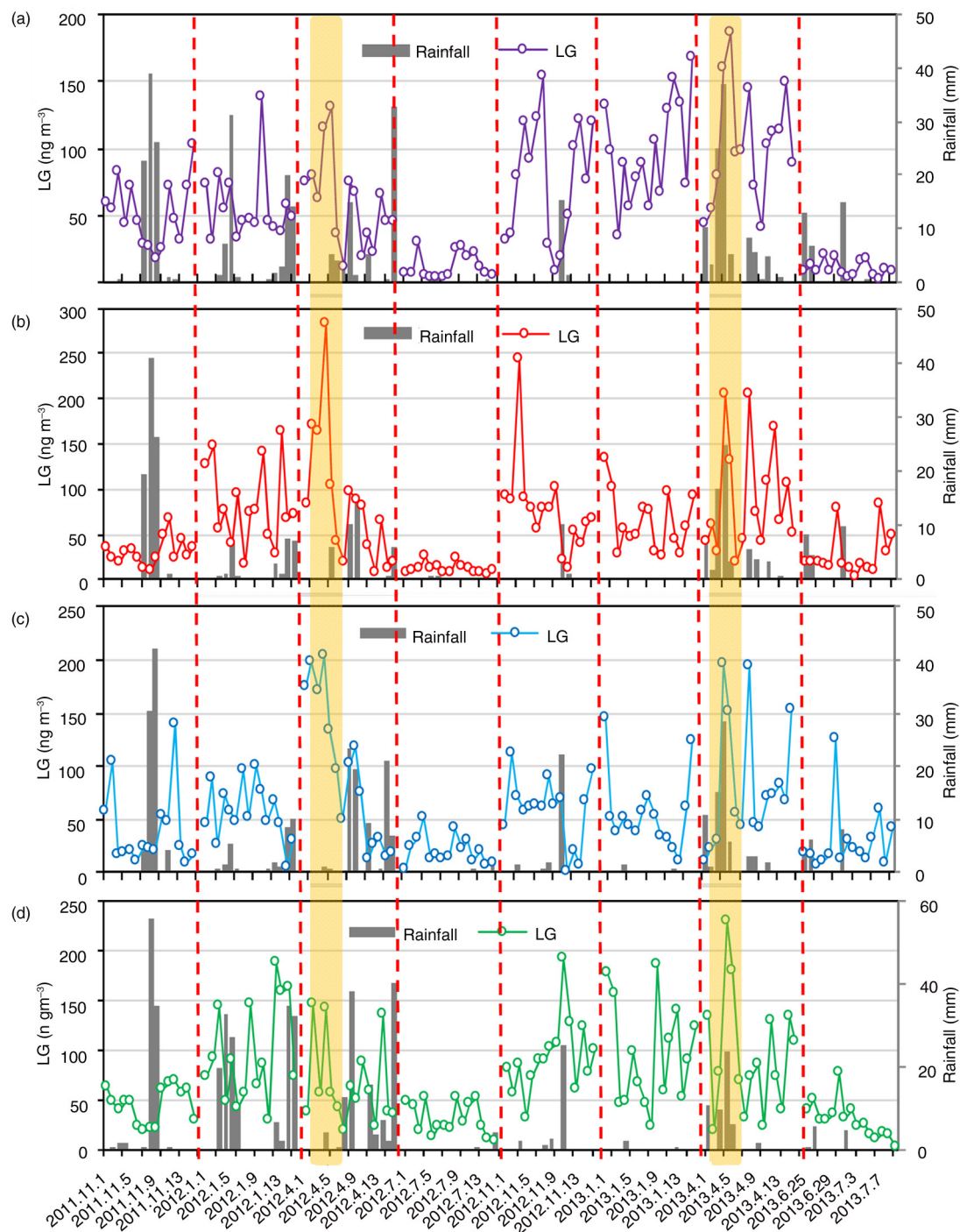


Fig. 2. Temporal variations of LG concentration and rainfall measured at (a) XM, (b) QZ, (c) PT and (d) FZ sites during the eight sampling periods. The two orange dashed bars represent high biomass burning activities around the traditional Ching-Ming festival.

regions, they might not be transported as far as plumes from stacks because the emission sources were close to the surface. Nevertheless, the air masses along the southeastern coastal region could carry small scale biomass burning smoke to the

sampling sites based on the backward trajectories in Supplementary Fig. 2. In addition, low temperature and solar radiation in winter might decrease the loss of LG due to the photodegradation and thus enhanced its ambient levels.

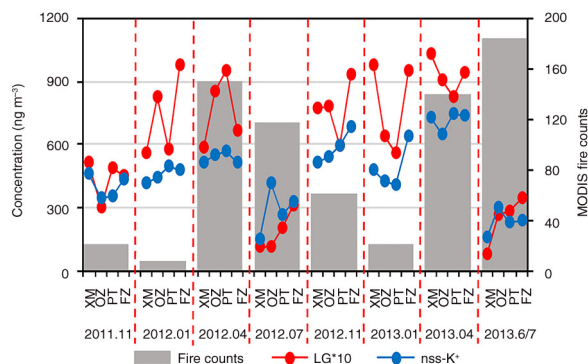


Fig. 3. Variations of LG and K^+ average concentrations and MODIS fire counts in Fujian province.

In April, intense biomass burning activities are apparent in southern, eastern and northern China (Supplementary Fig. 1), which are consistent with the common field burning of agricultural residues (e.g. crop stubble and wild grass) before spring ploughing. Owing to the elevated field burning of agricultural residues in Fujian (Fig. 3) and prevailing northeast winds (Supplementary Fig. 1) during the pre-monsoonal periods, concentrations of the LG were comparable to and even higher than those in winter (Figs. 2 and 3). The importance of local biomass burning was also observed at a background site in Ningbo where much higher levels of LG were found in summer with respect to other seasons due to the intensive biomass burning around the site (Liu et al., 2013). In November sampling periods, biomass burning activities such as post-harvest wheat and corn straw burning were much reduced (Supplementary Fig. 1) and the statistical fire counts in Fujian were also relatively low (Fig. 3). Therefore, less LG would be transported to the sampling sites by the air masses from the upwind regions (Supplementary Fig. 2) and less LG were emitted from local sources resulting low levels of LG (Figs. 2 and 3).

As shown in Fig. 2, there was a substantial day-to-day variation in the LG concentration at the same site in the same sampling period. Although there is some coherence between the time series of rainfall and low LG level, LG concentration didn't correlate well with rainfall, probably due to the ineffective wash-out of $PM_{2.5}$ by the light rain and/or a time lag between the reduction of $PM_{2.5}$ and the rain event (Wu et al., 2015a). In addition, occasional local biomass burning activities such as outdoor and indoor incense and joss paper burning as well as open burning of weeds, leaves and other plant materials on graveyards around the traditional Ching-Ming festival (the 4th of April in 2012 and 2013) to worship deceased ancestors might contribute to some short-lived enhancements of LG in this study (Tsai et al., 2010; Sang et al., 2011).

Nss- K^+ exhibited a similar seasonal trend to LG with minimum means in summer ($155\text{--}423\text{ ng m}^{-3}$) and higher values in other seasons ($349\text{--}744\text{ ng m}^{-3}$) (Supplementary Fig. 3). A comparison of LG and nss- K^+ shows different degrees of correlation among the four sites (Fig. 4). At the XM site on the Xiamen Island where road sweeping is more frequent, the temporal correlation between the two tracers was higher than at other sites ($r^2 = 0.5393$ vs $0.3467\text{--}0.3802$, $p < 0.01$). The lower correlation at the QZ, PT and FZ sites may be due to sources other than biomass burning for nss- K^+ , such as fugitive dusts, meat cooking and waste incinerators (Simoneit, 2002; Duvall et al., 2008). Although the correlations between LG and nss- K^+ are significant at $p < 0.05$ level at the four sites (Table 1), the distinct seasonal variations of LG to nss- K^+ ratio (LG/nss- K^+), with much lower values in summer sampling periods ($0.03\text{--}0.13$, Fig. 5b), appear to be evidence for enhanced loss of LG through photo-oxidation along the atmospheric transport of air masses and/or different biomass burning in summer sampling periods (Simoneit, 2002; Wang et al., 2007; Duvall et al., 2008; Hennigan et al., 2010; Cheng et al., 2013). Similar seasonal trends of LG/nss- K^+ ratio have also been reported in Beijing (Cheng et al., 2013; Wang et al., 2014), Guangzhou (Zhang et al., 2015), and southeastern US (Zhang et al., 2010) and variable emission sources and combustion conditions were suggested to be the main cause. Cheng et al. (2013) summarised the $(LG/nss-K^+)_{BB}$ in all kinds of source samples and concluded that the LG/nss- K^+ ratios are able to distinguish the wood burning $[(LG/K^+)_{BB} > 1.0]$ from crop residues $[(LG/K^+)_{BB} < 1.0]$. In this case, the LG/nss- K^+ ratios averaged from 0.03 ± 0.01 to 0.24 ± 0.13 , which were close to the corresponding source ratios of wheat straw (0.1) and corn straw (0.21) burning smoke (Cheng et al., 2013), slightly lower than the ratios for rice straw burning ($0.24\text{--}0.57$, Engling et al., 2009; 0.323 , Viana et al., 2008; $0.07\text{--}1.22$, Sullivan et al., 2008) and rape and wheat straw open burning ($0.10\text{--}1.0$, Tao et al., 2013), but much lower than those of wood burning smoke ($5.89\text{--}23.96$, Cheng et al., 2013; $1.45\text{--}6.40$, Andreae and Merlet, 2001) (Supplementary Table 2). However, the $(LG/K^+)_{BB}$ ratio of US grass burning varied over a wide range from 0.06 to 9.5 depending on the variety of grass and combustion conditions (dried or fresh) (Sullivan et al., 2008, 2014). Consequently, the measured LG in $PM_{2.5}$ at the four coastal urban sites in Fujian province was more likely associated with the combustion of crop residues and/or grass than wood based on the published $(LG/K^+)_{BB}$ ratios for different type of biomass (Supplementary Table 2), and the clear seasonal trend of LG/nss- K^+ was mainly due to the different loss of LG through atmospheric photochemical oxidation which depends on ambient atmospheric conditions in each

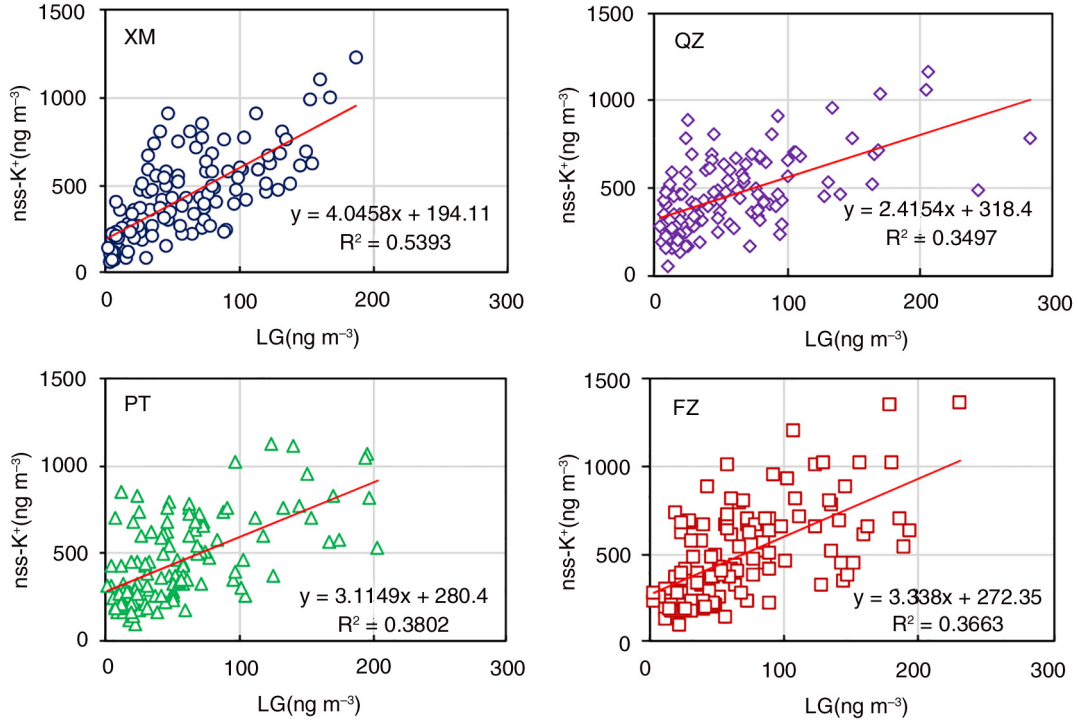


Fig. 4. Correlations between LG and nss-K⁺ for all samples at each site.

sampling periods (Hennigan et al., 2010; Cheng et al., 2013; Wu et al., 2015b).

3.2. Relationship between LG and carbon species

Like LG and nss-K⁺, WSOC also exhibited clear seasonal variations with minimum concentrations observed in summer season (Fig. 5a), which showed a similar trend as those reported in PRD (Ho et al., 2014), largely due to the Asian monsoon influence. The seasonal mean of WSOC among the four sites varied from $1.31 \pm 0.51 \mu\text{g C m}^{-3}$ to $4.94 \pm 1.55 \mu\text{g C m}^{-3}$, with an annual average of $2.69 \pm 1.21 \mu\text{g C m}^{-3}$, which were comparable to the levels of

WSOC in PM_{2.5} at four sites in the PRD region ($2.1 \pm 1.6 \mu\text{g C m}^{-3}$, Ho et al., 2014) and a rural site at Changdao Island ($2.10\text{--}5.89 \mu\text{g C m}^{-3}$, Feng et al., 2007) but much lower than those reported in Beijing ($7.2 \pm 5.5 \mu\text{g C m}^{-3}$, Du et al., 2014b). As listed in Table 1, strong correlations were found between WSOC and OC ($p < 0.001$) with relatively higher correlations in summer and fall compared to those in spring and winter ($r = 0.715\text{--}0.886$ vs $0.650\text{--}0.713$), which looks like the trend of correlation between WSOC and SOC (secondary OC estimated by the EC-tracer method and the details of SOC estimation can be found in Wu et al., 2015a). Furthermore, both the contribution of WSOC to OC (WSOC/OC) and SOC to OC (SOC/OC) exhibited a

Table 1. Correlation coefficient between species ($n = 60$)

Sampling period	LG vs Abs ₃₆₅	LG vs OC	LG vs WSOC	LG vs EC	LG vs nss-K ⁺	WSOC vs Abs	WSOC vs OC	WSOC vs nss-SO ₄ ²⁻	WSOC vs NO ₃ ⁻	WSOC vs SOC
2011.11	0.432**	0.564**	0.432**	0.529**	0.420*	0.889**	0.792**	0.757**	0.573**	0.670**
2012.1	0.283*	0.322**	0.307**	0.491**	0.254*	0.693**	0.650**	0.676**	0.604**	0.595**
2012.4	0.482**	0.475**	0.380**	0.227*	0.358**	0.690**	0.713**	0.548**	0.488**	0.552**
2012.7	0.418**	0.635**	0.253*	0.486**	0.358**	0.829**	0.886**	0.478**	0.401**	0.605**
2012.11	0.403**	0.348**	0.276*	0.266*	0.353**	0.838**	0.715**	0.726**	0.422**	0.697**
2013.1	0.485**	0.750**	0.596**	0.677**	0.384**	0.700**	0.663**	0.682**	0.527**	0.686**
2013.4	0.477**	0.389**	0.269**	0.435**	0.570**	0.706**	0.659**	0.584**	0.634**	0.620**
2013.6/7	0.312*	0.444**	0.453**	0.598**	0.575**	0.763**	0.746**	0.485**	0.346**	0.682**

**Correlation is significant at the 0.01 level (one-tailed). *Correlation is significant at the 0.05 level (one-tailed).

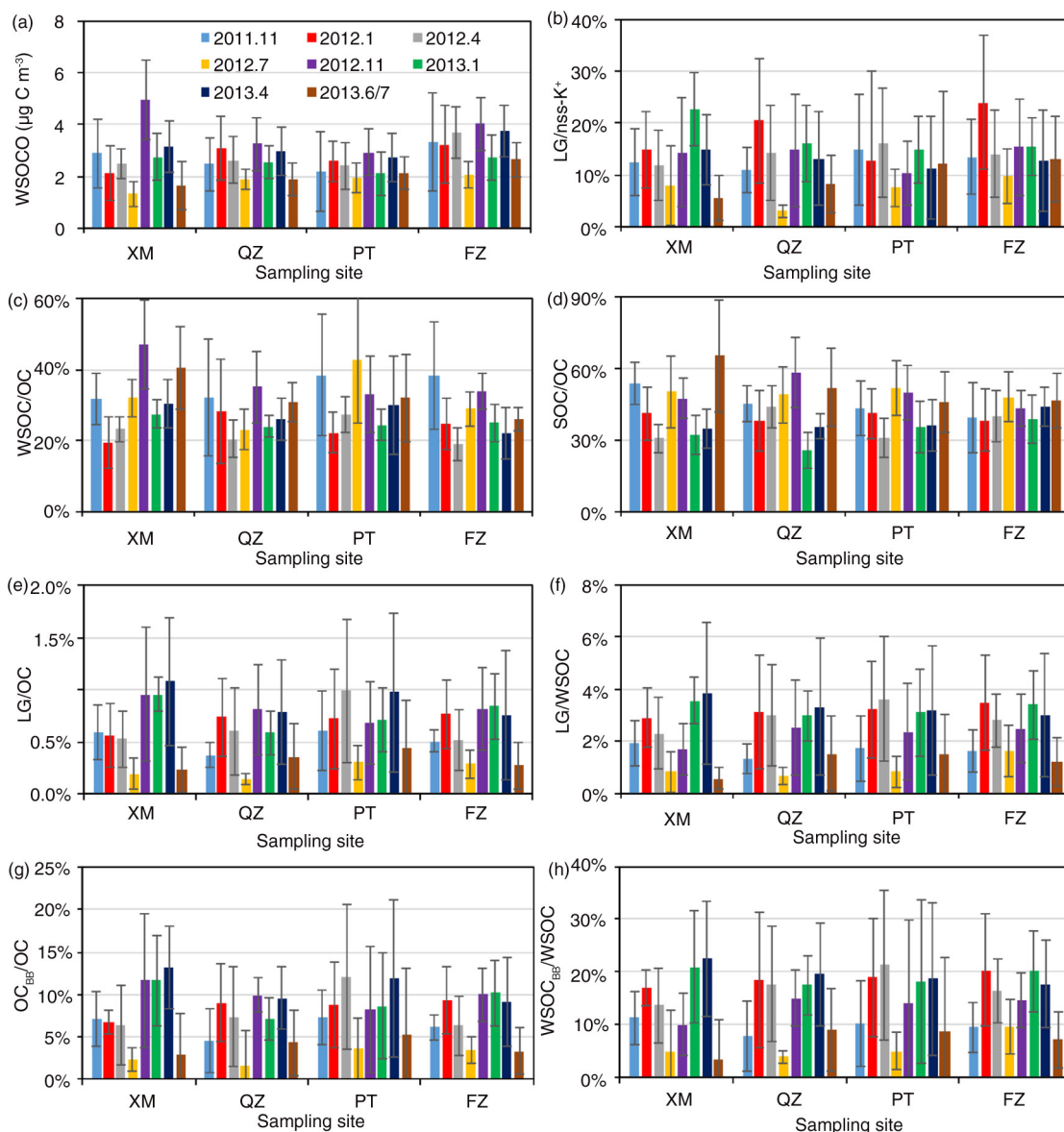


Fig. 5. (a) Seasonal variations of WSOC concentration, and ratios of (b) LG/nss- K^+ , (c) WSOC/OC, (d) SOC/OC, (e) LG/OC, (f) LG/WSOC, (g) OC_{BB}/OC and (h) WSOC_{BB}/WSOC in $PM_{2.5}$ at the four urban sites.

similar seasonal variation with higher values in fall and summer and lower values in spring and winter, as illustrated in Fig. 5c and d. According to Weber et al. (2007) and Ding et al. (2008b), the increase of WSOC/OC can be related to the enhancement of secondary organic aerosol (SOA) formation and/or biomass burning activities. The WSOC/OC ratios were on average lower than those measured in biomass burning smoke (48 %, Guo et al., 2014). In summer and fall when the levels of LG were relatively low, both the stronger correlations between WSOC and OC and the higher WSOC/OC ratios could be attributed to the favourable ambient conditions such as the higher

concentrations of O_3 , OH and $NO_3\cdot$ radicals as well as higher temperature and UV intensity for photochemical processes (Sullivan et al., 2004; Seinfeld and Pandis, 2006; Feng et al., 2007; Wu et al., 2015a, 2015b). The WSOC/OC ratios obtained in this study (18.9–46.7 %) were comparable with those reported in fall-winter in PRD (38.8–57.9%, Ding et al., 2013) but lower than those in Beijing (38.8–62.6% annual, Du et al., 2014b), southeastern US (44–72 % annual, Ding et al., 2008b) and Changdao Island, China (40–59%, Feng et al., 2007). It is worth noting that the SOC/OC ratio was on average 1.5 ± 0.6 times of WSOC/OC; that is, the amount of SOC was higher than that of WSOC.

In other words, considerable WSOC was likely from secondary formation instead of primary biomass burning.

Since WSOC is mainly derived from biomass burning and secondary photochemical reaction of volatile organic compounds (VOCs) (Weber et al., 2007), Ding et al. (2013) calculated secondary organic carbon (SOC) contribution by the difference between measured WSOC (WSOC_{measured}) and WSOC from biomass burning (WSOC_{BB}, estimated using LG-tracer method); that is, $SOC = WSOC_{measured} - WSOC_{BB}$. They found much higher concentrations of WSOC (7.63–11.5 $\mu\text{g C m}^{-3}$) than those of SOC (4.18–5.79 $\mu\text{g C m}^{-3}$) in PRD rural and urban areas. Using EC-tracer method, however, Kunwar and Kawamura (2014) found lower SOC to WSOC ratios in winter-spring (0.49–0.78) but higher ratios in summer-fall (1.53–1.65) on Okinawa Island. Ding et al. (2012) has pointed out the significant overestimation of SOC by EC-tracer method, which could result in the higher amount of SOC with respect to WSOC throughout the sampling campaigns in this study. Nevertheless, the factors of LG in WSOC from BB sources used for the estimation of WSOC_{BB} depend strongly on fire conditions (fuel type and fire flaming/smouldering ratios) (Ding et al., 2008a; Sullivan et al., 2014; Yan et al., 2015). Therefore, particular caution is needed to investigate SOC because it is estimated using indirect approaches and much influenced by the selected reference OC/EC and LG/WSOC ratios in source emissions.

As also can be seen in Table 1, the correlations between WSOC and sulphate were positively significant as well ($r = 0.497\text{--}0.743$, $p < 0.001$, $n = 120$) throughout the eight sampling periods and much higher than those between WSOC and LG ($r = 0.253\text{--}0.596$, $p < 0.05$, $n = 120$) in the same sampling period. The correlation between WSOC and nitrate was similar with that between WSOC and sulphate in different sampling periods. In Beijing and Nanjing, strong correlations between WSOC with sulphate and nitrate were also observed throughout a year except summer, which possibly indicate their similar sources or formation pathways (Du et al., 2014b). The stronger correlations between WSOC and nss-SO_4^{2-} compared to that between WSOC and LG further indicate that WSOC was more closely related to secondary formation pathways (Seinfeld and Pandis, 2006; Du et al., 2014b). In previous studies carried out in winter in Beijing and Tokyo, considerable WSOC was suggested to be from photochemical production in spite of the high correlations existed between WSOC and biomass burning (Du et al., 2014b). In addition, no clear seasonal trend for the correlation between WSOC and LG was observed in this study, further indicating that the influences of biomass burning emissions on WSOC are not very important in Fujian, even in winter and spring due to the relatively low concentrations of LG compared to those measured in Beijing (Cheng et al., 2013; Du et al.,

2014b; Wang et al., 2014), Guangzhou (Ho et al., 2014; Zhang et al., 2015) and Chiang Mai (Tsai et al., 2013). It is noteworthy that the correlations between WSOC and nss-SO_4^{2-} or NO_3^- were relatively low in summer sampling seasons with respect to other seasons (Table 1). In summer, WSOC was more influenced by strong solar radiation and biogenic source than direct biomass burning activities. As for nss-SO_4^{2-} and NO_3^- , low concentrations were measured due to their decreased precursors such as SO_2 and NO_x under the influence of southwest monsoon as well as the enhanced loss of NH_4NO_3 at high temperature (Wu et al., 2015a). Despite the similar influencing factor in summer, WSOC and inorganic ions behaved differently resulting in lower correlations with respect to other seasons.

As mentioned in the introduction section, the absorption at 365 nm (Abs_{365}) can be used as an indicator of the light absorption properties of carbonaceous species or brown carbon. It is well accepted that biomass burning is one important sources of brown carbon in the atmosphere, especially in regions heavily affected by biomass burning emissions (Zhang et al., 2010; Du et al., 2014a). Other potential sources of brown carbon include tar materials from coal combustion, humic-like organics from soil, atmospheric aging of organics, and organic matter from ocean surface through bubble-bursting (Hecobian et al., 2010). The correlations between Abs_{365} and WSOC ($r = 0.690\text{--}0.889$, $p < 0.001$) were much stronger than those between Abs_{365} and LG ($r = 0.283\text{--}0.415$, $p < 0.05$) during the eight sampling periods, implying that secondary formation pathways might be more important than primary biomass burning emissions for brown carbon in PM_{2.5}. Although the temporal correlation between LG and nss-K^+ was significant ($r = 0.353\text{--}0.575$, $p < 0.001$) in each sampling period, no clear seasonal trend for the correlation was observed (Table 1), probably related to the interruption of K^+ from non-biomass burning emissions. Moderate to high correlation of LG with OC and EC were also obtained but without clear seasonal trends. These significant, but not very high, correlations of LG with other components (Abs_{365} , OC, WSOC, EC and K^+) documented that the temporal variation of LG was characterised by the biomass burning-related components to some extent.

3.3. Contributions of biomass burning based on tracer method and PMF analysis

While LG is expected to be potentially oxidised by atmospheric oxidants (Hennigan et al., 2010; Hoffmann et al., 2010), the significant correlations between LG and nss-K^+ shown in Fig. 4 and Table 1 indicate that LG still can be considered as a good tracer to estimate biomass burning contribution to carbonaceous aerosol. The $(\text{LG}/\text{OC})_{\text{BB}}$ ratio has been used to estimate the contribution of biomass

burning emissions to carbonaceous aerosols (Zhang et al., 2008, 2010; Mkoma et al., 2013; Ho et al., 2014). In this study, we used a widely used $(\text{LG}/\text{OC})_{\text{BB}}$ value of $\sim 8.2\%$ in the estimation of biomass burning contributions to OC (OC_{BB}) (Zhang et al., 2008, 2012; Sang et al., 2011; Mkoma et al., 2013; Ho et al., 2014; Zhu et al., 2015). Besides the $(\text{LG}/\text{OC})_{\text{BB}}$ ratio, the LG to WSOC ratio $(\text{LG}/\text{WSOC})_{\text{BB}}$ has also been used to estimate biomass burning derived WSOC (WSOC_{BB}) (Ding et al., 2013; Feng et al., 2013; Yan et al., 2015). The $(\text{LG}/\text{WSOC})_{\text{BB}}$ ratio was reported as 9.94 % and 8.06 % for forest fire plume and rice straw residues open burning, respectively (Ding et al., 2008b; Viana et al., 2008). Tao et al. (2013) reported higher values of $(\text{LG}/\text{WSOC})_{\text{BB}}$ from wheat straw burning (23.8–24.2 %) than those from rape straw burning (9.1 %). A value of 17 % from the test burns of rice and wheat straws was used to estimate WSOC_{BB} for summer aerosols in Beijing (Yan et al., 2015). The measured values for $(\text{LG}/\text{WSOC})_{\text{BB}}$ from controlled laboratory burns ranged from 1.7 % for marsh grass to 14.9 % for grass (Sullivan et al., 2014) (Supplementary Table 2). According to Yan et al. (2015), the $(\text{LG}/\text{WSOC})_{\text{BB}}$ ratio of 17 % from Chinese rice and wheat straws burning was applied to estimate WSOC_{BB} in this study.

As can be seen in Fig. 5e and f, both the LG/OC and LG/WSOC values in the ambient $\text{PM}_{2.5}$ were much lower than the reference value for biomass burning smoke (8.2 % for $(\text{LG}/\text{OC})_{\text{BB}}$ and 17 % for $(\text{LG}/\text{WSOC})_{\text{BB}}$) but exhibited very similar seasonal trends with minimum values in summer largely due to the enhanced loss of LG through photochemical oxidation processes. The LG/OC values ranged from 0.14 % to 0.44 % during summer sampling campaigns, which was 0.24 to 0.65 times of the values in other seasons, while the LG/WSOC value was 0.65–1.63 %, which was 0.42 times on average the values in other seasons. The seasonal trend of LG/OC ratio is consistent with those reported for Changdao Island located between Bohai Sea and Yellow Sea in north China (0.12 % vs 0.54–1.0 %, Feng et al., 2007), Okinawa (0.03 % vs 0.15–0.46 %) and Chichi-jima (0.062 % vs 0.11–0.38 %) over the western North Pacific (Mochida et al., 2010; Zhu et al., 2015), four cities in PRD in southern China (0.6 % vs 1.0–1.9 %, Ho et al., 2014) and Beijing urban area (1.4 % vs 1.5–3.3 %, Du et al., 2014b). In Beijing, however, the maximum values of LG/OC were observed in fall instead of winter due to the intensive post-harvest crop residues burning in the neighbouring regions (Du et al., 2014b). Similarly, minimum LG/WSOC values were also reported in summer in four cities in PRD (2.6 % vs 2.8–5.9 %, Ho et al., 2014), Beijing urban area (2.2 % vs 3.3–7.8 %, Du et al., 2014b), Changdao Island (0.2 % vs 0.96–2.51 %, Feng et al., 2007) and Okinawa (0.09 % vs 0.17–0.22 %, Zhu et al., 2015).

Using the $(\text{LG}/\text{OC})_{\text{BB}}$ ratio of 8.2 % and $(\text{LG}/\text{WSOC})_{\text{BB}}$ ratio of 17 % from the crop straw burning smoke, the calculated relative contributions from biomass burning to OC ($\text{OC}_{\text{BB}}/\text{OC} = \text{LG}/8.2\%/\text{OC}$) and WSOC ($\text{WSOC}_{\text{BB}}/\text{WSOC} = \text{LG}/17\%/\text{WSOC}$) in each sampling campaign varied in 3.3–13.1 % (av. 8.3 %) and 7.1–22.5 % (av. 15.2 %), respectively (Fig. 5g and h). Using the same $(\text{LG}/\text{OC})_{\text{BB}}$ ratio of $\sim 8.2\%$, the calculated biomass burning contributions to OC were 18–38 % in Beijing (Zhang et al., 2008), 6.5–11 % in PRD during spring time (Sang et al., 2011), 7.6–21.6 % in four cities in PRD (Ho et al., 2014), 1.9–7.9 % in wet season and 15.8–23.3 % in dry season in PRD region (Zhang et al., 2015), 26–68 % in Tanzania (Mkoma et al., 2013) and 0.4–5.6 % on Okinawa island (Zhu et al., 2015). Using a radiocarbon-based source apportionment method, the contribution of biomass burning emissions to OC in $\text{PM}_{2.5}$ was estimated to be 32–40 % at a regional background site on Hainan Island, South China (Zhang et al., 2014). During the 2-yr sampling period, the values of $\text{WSOC}_{\text{BB}}/\text{WSOC}$ were lower than those reported in Shanghai (11–54 %, Feng et al., 2013), PRD region during fall-winter period (42–47 %, Ding et al., 2013), and urban Beijing in winter and summer (23 % and 16 %, Yan et al., 2015). The underestimation of $\text{WSOC}_{\text{BB}}/\text{WSOC}$ in this study may be partly caused by the higher $(\text{LG}/\text{WSOC})_{\text{BB}}$ of 17 % than those used in Shanghai (12.5 %) and PRD region (9 %) (Ding et al., 2013; Feng et al., 2013). Radiocarbon measurements of WSOC in $\text{PM}_{2.5}$ at a background site in Yellow Sea in spring with outflow from northern China showed that the non-fossil source fraction (biogenic and biomass burning) to WSOC was estimated to be 30–50 % (Kirillova et al., 2014).

In addition, the impact of biomass burning on ambient $\text{PM}_{2.5}$ could be assessed by using the ratio of LG to $\text{PM}_{2.5}$ $(\text{LG}/\text{PM}_{2.5})_{\text{BB}}$ for typical biomass (Wang et al., 2007; Sang et al., 2011). The reported $(\text{LG}/\text{PM}_{2.5})_{\text{BB}}$ values for wood burning in fireplaces varied from 7.13 % to 22.6 % (Fine et al., 2001; Schauer et al., 2001), which were much higher than those reported for Chinese cereal straw burning with an average ratio of 4.52 % (Zhang et al., 2007). Since crop residues or grass burning are suggested to be the major type of biomass burning based on the measured LG/K^+ ratios, the $(\text{LG}/\text{PM}_{2.5})_{\text{BB}}$ ratio of 4.52 % is used to estimate the biomass burning contributions to ambient $\text{PM}_{2.5}$ at the four site along Fujian coastal line. The results showed that on average 0.67–1.99 % (av. 1.49 %) in summer and 1.44–3.91 % (av. 2.58 %) in other seasons of ambient $\text{PM}_{2.5}$ were due to biomass burning (Fig. 6), which were comparable to the biomass burning estimates (1.6–3.7 %) at the rural site in PRD during springtime (Sang et al., 2011), but were lower than the values (3.0–19 %) reported in Guangzhou in October (Wang et al., 2007) and in the range of 1.52–12.09 % using bottom-up methodology for biomass burning

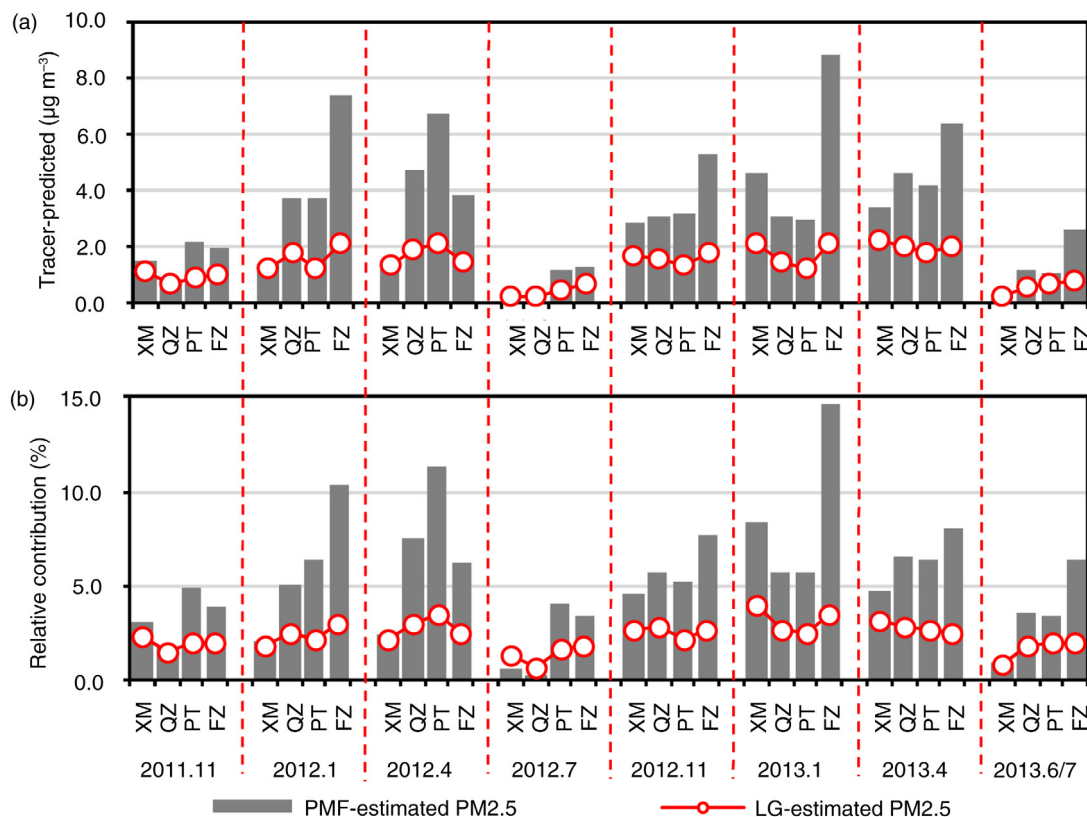


Fig. 6. (a) Mass contributions and (b) relative mass contributions of biomass burning on PM_{2.5} between LG-predicted and PMF-predicted analysis.

emission inventory in PRD (Supplementary Table 3). It is noted that the contributions of biomass burning based on the simplified tracer method could be underestimated due to the loss through degradation by hydroxyl radicals, dry/wet deposition and dispersion during the atmospheric transport (Hennigan et al., 2010; Zhang et al., 2012). It should also be noted that using fixed values of (LG/OC)_{BB}, (LG/WSOC)_{BB} and (LG/PM_{2.5})_{BB} in different seasons would lead to considerable uncertainty given the known strong dependence of LG and carbonaceous aerosol emissions on biomass burning conditions.

A further source apportionment of PM_{2.5} was carried out using USEPA PMF 5.0 software and a total of seven factors were resolved (Supplementary Figs. 3–6). The first factor was biomass burning, reflected in the high loading LG (varying from 62 % at FZ to 80 % at PT) but low K⁺ (varying from 6 % at XM to 18 % at QZ), which further confirmed that LG was a better biomass burning tracer with respect to K⁺ in this study. Moreover, much lower characteristic values of WSOC (9–17 %) and Abs₃₆₅ (4–18 %) were observed in this factor, again indicating that biomass burning was a minor contributor of WSOC and brown carbon in PM_{2.5}.

The second factor had the highest characteristic values of V, which is a good marker for fuel oil combustion (Chow and Watson, 2002). Huang et al. (2014a, 2014b) attributed the factor with high contribution of V to the residual oil or heavy oil combustion from ship emissions in the coastal cities of Hong Kong and Shenzhen in South China. Moreover, moderate contributions of Na⁺ were also found in this factor, suggesting that this factor can be determined to be fishing and/or cargo vessel emissions along the coastal line of Taiwan Straits.

The third factor, vehicular emission, was characterised by relatively higher EC with respect to OC and moderate Pb. Relatively higher EC compared to OC has also been reported in vehicular emissions factors resolved by PMF in Shenzhen and Hong Kong in China (Huang et al., 2014a, 2014b). Since unleaded fuel oil still contains lead derived from the crude oil, the vast fuel oil consumed by vehicles could be an important source of atmospheric lead in China. In this study, 24–38 % of annual PM_{2.5} associated Pb was attributed to vehicular emissions, which was comparable to the reported values in Xi'an (~30 %, Xu et al., 2012b) and Shanghai (~55 %, Li et al., 2009).

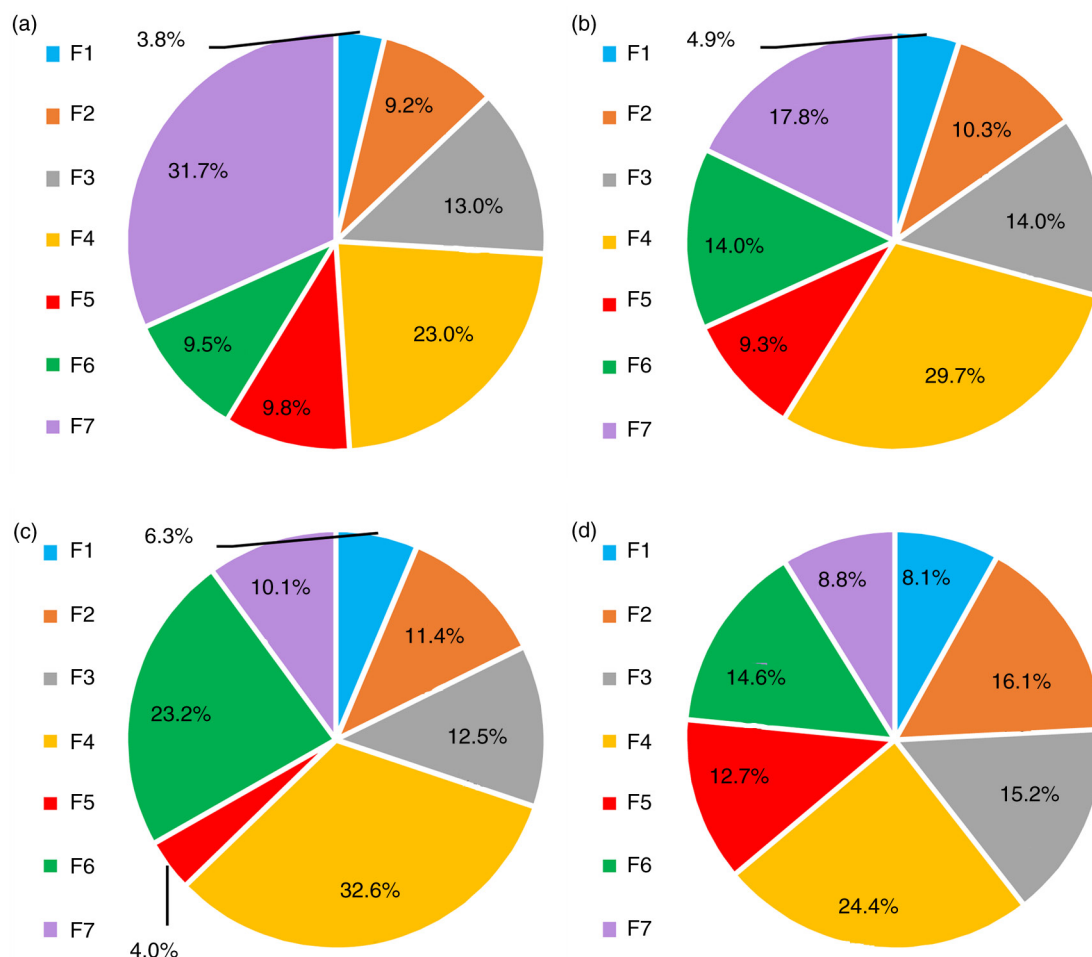


Fig. 7. Pie-charts showing the source apportionment of $PM_{2.5}$ at (a) XM, (b) QZ, (c) PT and (d) FZ. The sources include biomass burning (F1), fuel oil (F2), vehicular emission (F3), secondary nitrate and sulphate aerosol (F4), crustal dust (F5), metallurgical emission at XM and QZ or mixed source of metallurgical emission and secondary sulphate aerosol at PT and FZ (F6), secondary sulphate and organic aerosol at XM and QZ or waste incineration emission at PT and FZ (F7).

The fourth factor was distinguished by the high characteristic value of NO_3^- followed by NH_4^+ and SO_4^{2-} with a strong seasonal variation peaking in spring or winter and is suggested to be associated with the formation of secondary nitrate and sulphate aerosol. The fifth factor had high characteristic values of crustal elements such as Si, Al, Ca and Ti, so this factor is considered to be soil and road dust.

For the sixth factor, Zn had the prominent characteristic values (52–64 %) probably due to the emission of metallurgical processes (Kulshrestha et al., 2009). In addition, garbage burning is another important potential source of atmospheric Zn because more and more municipal wastes are treated by incineration system instead of traditional landfill in this region. Industrial activities and waste incineration have been suggested to be the main sources of Zn particles in Mexico City and Beijing (Moffet et al., 2008;

Li and Shao, 2009). At the PT and FZ sites, relatively high characteristic values of SO_4^{2-} , WSOC and Abs_{365} were also observed in this factor documenting the mixed source of secondary aerosol formation and Zn-related sources. At the XM and QZ sites, however, relatively high characteristic values of SO_4^{2-} , WSOC and Abs_{365} were observed in the seventh factor. The seventh factor at the PT and FZ sites was characterised by moderate Zn, Fe and Mn, suggesting that this factor was associated with metallurgical industrial emissions.

Annually, the highest contribution of biomass burning to $PM_{2.5}$ resolved by PMF was 8.08 % at the FZ, followed by 6.32 % at the PT, 4.94 % at the QZ and 3.75 % at the XM (Fig. 7 and Supplementary Table 3). The PMF-estimated annual contributions were lower than those PMF results using total K^+ in urban Beijing (11 %, Song et al., 2006; 7.0–16.4 %, Song et al., 2007; 24 %, Cheng et al., 2013;

13 %, Zhang et al., 2013c), urban Chengdu (11 %, Tao et al., 2014), suburban Hong Kong (19.5 %, Huang et al., 2014b) and urban Shenzhen (9.8 %, Huang et al., 2014a), and PMF results using LG in the southeastern US (13 %, Zhang et al., 2010) (Supplementary Table 3).

As shown in Fig. 7, the average values of LG-estimated PM_{2.5} were generally lower than those of PMF-estimated, especially in winter and spring sampling periods. The seasonal patterns of biomass burning contributions were consistent with the trends of LG and backward trajectories of air masses or wind direction. Both PMF- and LG-estimated results suggest that biomass burning might not significantly affect the ambient PM_{2.5} along the coastal line. However, the estimated biomass burning contribution could be underestimated due to the loss of LG during the atmospheric transport (Hennigan et al., 2010; Zhang et al., 2012). Nevertheless, the influence of WSOC derived from biomass burning can enhance the hygroscopic property of particles in the atmosphere and serve as cloud condensation nuclei. Further studies at more locations on SOA tracers together with LG and ¹⁴C measurements are needed to get better understanding of the contributing sources of carbonaceous species in PM_{2.5} (Ding et al., 2013; Feng et al., 2013; Liu et al., 2013; Kirillova et al., 2014; Zhang et al., 2014).

4. Conclusions

In this study, the concentrations of LG and WSOC as well as the light absorption at 365 nm of WSOC in PM_{2.5} were measured during the eight sampling campaigns at four urban sites along the coastal line of southeast China. LG showed large seasonal variations with summer minima and winter/spring maxima. The summer low levels may be largely due to the joint effects of fewer biomass burning activities in the upwind regions of Philippines, enhanced degradation of LG and enhanced thermal convection associated with summer monsoon. On the contrary, biomass burning emissions either from crop residues open field burning or domestic cooking using biofuel in Fujian and Zhejiang coastal regions in winter and spring affect the levels of LG at the four coastal urban sites via northeastern winds. Similar to PM_{2.5}, the day-to-day variations of LG were much influenced by rainfall scavenging. In addition, abrupt local biomass burning activities during traditional Ching-Ming festival could enhance the levels of LG at the urban sites. Although LG correlated well with nss-K⁺ in each sampling periods, the lack of clear seasonal pattern for the correlations between LG and nss-K⁺ suggest multiple nss-K⁺ sources in addition to biomass burning sources. Nevertheless, the minimum LG and nss-K⁺ concentrations as well as LG/nss-K⁺ ratios observed in summer appeared

to be more influenced by enhanced photochemistry when there was little biomass burning than different types of biomass burning. Higher WSOC/OC and SOC/OC ratios and stronger correlations between WSOC and OC and SOC were obtained in fall and summer when the ambient conditions were favourable for photochemical processes, indicating that WSOC and SOC should have similar formation pathways (i.e. atmospheric photochemical oxidation). The stronger relationship between WSOC and sulphate with respect to that between WSOC and LG seemed to further suggest the importance of secondary formation for WSOC during all the sampling periods. Clear seasonal trends of the contributions of LG to WSOC and OC were obtained with summer minima and spring/winter maxima.

Using the characteristic ratios of (LG/OC)_{BB} and (LG/WSOC)_{BB} in the biomass burning smoke from previous studies, it was estimated that on average 13.6 % of OC and 26 % of WSOC at the four coastal urban sites may be contributed from biomass burning activities. Through the PMF source apportionment model, seven major potential source factors were identified. The factor, characterised by high values of LG (62–80 %), was identified as biomass burning sources. However, much lower characteristic values of WSOC (9–17 %) and Abs₃₆₅ (4–18 %) associated with this factor suggest that biomass burning contribution to WSOC or brown carbon is less of importance. The biomass burning sources contributed 3.75–8.08 % to the PM_{2.5} annually, with minimum values in summer and maximum values in spring/winter. Generally, the levels of LG-estimated PM_{2.5} were lower than those of PMF-estimated PM_{2.5}, especially in spring and winter. Considering the potential loss of LG in ambient PM_{2.5} as a specific marker for biomass burning and the significant difference between PMF- and LG-estimated results, PMF method is expected to give a reliable estimate of biomass burning contribution. This study also suggests that the influence of biomass burning activities on PM_{2.5} cannot be ignored along the coastal line of southeast China, especially during the northeastern monsoon season. Owing to the hygroscopic nature of WSOC and the relative high contribution of biomass burning (av. 15.2 %), it is expected that the addition of biomass burning smoke could affect cloud condensation nuclei activity.

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