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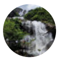

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Regional monitoring of biomass burning using passive air sampling technique reveals the importance of MODIS unresolved fires

Haoyu Jiang[a,b], Jun Li[a,b], Jiaqi Wang[a,b], Hongxing Jiang[a,b], Yangzhi Mo[a,b], Jiao Tang[a,b], Ruijie Zhang[c], Wanwisa Pansak[d], Guangcai Zhong[a,b], Shizhen Zhao[a,b], Jicai Ning[e], Chongguo Tian[e], Gan Zhang[a,b,*]

[a] *State Key Laboratory of Organic Geochemistry, Guangdong Key Laboratory of Environmental Protection and Resources Utilization, and Joint Laboratory of the Guangdong-Hong Kong-Macao Greater Bay Area for the Environment, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China*

[b] *CAS Centre for Excellence in Deep Earth Science, Guangzhou 510640, China*

[c] *School of Marine Sciences, Guangxi University, Nanning 530004, China*

[d] *Department of Agricultural Science, Naresuan University, Phitsanulok 65000, Thailand*

[e] *Key Laboratory of Coastal Environmental Processes and Ecological Remediation, Yantai Institute of Coastal Zone Research, Chinese Academy of Sciences, Yantai 264003, PR China*

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ABSTRACT

Field-based sampling can provide more accurate evaluation than MODIS in regional biomass burning (BB) emissions given the limitations of MODIS on unresolved fires. Polyurethane foam-based passive air samplers (PUF-PASs) are a promising tool for collecting atmospheric monosaccharides. Here, we deployed PUF-PASs to monitor monosaccharides and other BB-related biomarkers and presented a dataset of 31 atmospheric BB-related biomarkers in the Indo-China Peninsula (ICP) and Southwest China. The peak concentrations of monosaccharides in the ICP occurred before monsoon season. The highest concentrations were in the eastern Mekong plain, while the lowest were along the eastern coast. BB-related biomarkers displayed elevated concentrations after April, particularly in the monsoon season; however, fewer active fires were recorded by MODIS. This revealed the importance of MODIS unresolved fires (e.g., indoor biofuel combustion, small-scale BB incidents, and charcoal fires) to the regional atmosphere. The PAS derived levoglucosan concentrations indicated that, with the inclusion of MODIS unresolved fires, the estimated top-down emissions of PM (4194–4974 Gg/yr), OC (1234–1719 Gg/yr) and EC (52–384 Gg/yr) would be higher than previous bottom-up estimations in the ICP. Future studies on these MODIS unresolved fires and regional monitoring data of BB are vital for improving the modeling of regional BB emissions.

1. Introduction

Biomass burning (BB), particularly open biomass burning events such as land clearing, agricultural waste burning, prescribed burning, field burning, and wildfires (He et al., 2016), is of global concern as a primary source of pollutant emissions into the atmosphere, playing an important role in climate change (Andreae and Merlet, 2001; Crutzen and Andreae, 1990). With a high proportion of agriculture and limited energy access, the over-reliance on BB in developing and least developed countries hinders economic and environmental sustainability (Kaygusuz, 2012), posing a major threat to human health and ecosystems worldwide (Adam et al., 2020; Andreae and Merlet, 2001; Crutzen and Andreae, 1990; He et al., 2016).

The Indo-China Peninsula (ICP), including Cambodia, Laos, Myanmar, Thailand, and Vietnam, is one of the most active fire hotspots and an important source region for global air pollutant emissions (Chan et al., 2003; Gautam et al., 2013; Lin et al., 2009; Sinha et al., 2003). With high natural productivity of vegetation, and intensive agricultural activities, the regional air quality of the ICP is seriously affected by BB. According to the annual total carbon emissions data in Southeast Asia,

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* Corresponding author at: State Key Laboratory of Organic Geochemistry, Guangdong Key Laboratory of Environmental Protection and Resources Utilization, and Joint Laboratory of the Guangdong-Hong Kong-Macao Greater Bay Area for the Environment, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China.

E-mail address: zhanggan@gig.ac.cn (G. Zhang).

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BB contributed 91 TgC/yr from 1997 to 2004 and comprised 4.9 % of global carbon emissions, 3831 Gg of non-methane volatile organic compounds, and 324 Gg of black carbon during 2001–2010 (Giglio et al., 2013; Shi and Yamaguchi, 2014), and contributed to the formation of atmospheric brown clouds over Asia (Gustafsson et al., 2009). Severe air pollution in the form of smoke haze in the ICP occurs annually in the dry season owing to large-scale open fires,[13] resulting in the increased oxidative stress and elevated mortality risk from cardiovascular diseases to individuals when compared with urban air (Adam et al., 2020; Betha et al., 2014).

Previous studies on regional BB emissions mostly focused on official statistics and models based on remote sensing data, such as MODIS products (Moderate Resolution Imaging Spectroradiometer). Unfortunately, owing to the limitations of research scale and data availability or quality, there is still a gap between the predicted value and the real value (Xu et al., 2022). Hence field-based monitoring data is imperative for improving BB emission inventories and models (Du et al., 2018; Lü et al., 2006; Song et al., 2009), and accurately assessing the impacts of BB on air quality (Giglio et al., 2013; Lin et al., 2014; Luo et al., 2021; van der Werf et al., 2010). However, few studies have monitored BB at a regional scale. Field-based monitoring of BB is still confined to scattered individual cities or receptor sites (Liu et al., 2020; Phairuang et al., 2019; Popovicheva et al., 2017; Tao et al., 2020). The lack of spatiotemporal distribution of BB information hampers the accurate evaluation of regional BB and effective mitigation efforts for regional BB emissions. BB-induced aerosols are generally considered to be profoundly influenced by open BB in periphery rural areas and long-range transport (Cheng et al., 2021; Titos et al., 2017; Zhang et al., 2010); and therefore, current air pollution regulatory measures for BB are focused on open BB from the countryside (Bikkina et al., 2019). However, non-open fires and small-scale BB incidents, mostly unresolved by MODIS, may also have a significant impact on regional air quality and may likely have been neglected in emission regulations over a long term.

Molecular markers or biomarkers with “chemical fingerprint”, such as cellulose and lignin degradation products (e.g., monosaccharides) released into the atmosphere, are typical indicators for tracking BB. High-volume active samplers are often used to collect particulate BB-related biomarkers (Hawthorne et al., 1989; Kuo et al., 2011). However, it is difficult to capture fire emission events and install multiple active samplers synchronously on a large-scale, because BB often occurs occasionally and/or in remote areas.

As the most popular passive sampling technology, polyurethane foam-based passive air samplers (PUF-PASs), have previously been demonstrated to be a cost-effective tool for collecting both the gaseous and particulate (<2 μm) phases of atmospheric monosaccharides in the Pearl River Delta of South China (Jiang et al., 2018). Degradation of monosaccharides in a PUF-PAS are likely to be negligible over a long-time deployment up to 92 days (Jiang et al., 2018). With the advantages of small size, low cost, portability, easy installation, power-free operation, and convenient maintenance, the PUF-PAS may be more applicable to large-scale, long-period sampling campaigns than active samplers. BB emissions from various sources could be captured by PUF-PASs via the time-weighted average (TWA) concentrations of monosaccharides derived by the PUF-PAS, thus reflecting the BB intensity of a given period (Jiang et al., 2018). In fact, not only monosaccharides, but also most BB-related organic tracers are semi-volatile (May et al., 2012; Simoneit, 2002), implying that PUF-PASs could be applied to sample more BB-related biomarkers.

Therefore, in this study, we conducted a regional monitoring campaign of BB in the ICP in 2016, using PUF-PASs to: (i) investigate the spatiotemporal distribution of 31 BB-related biomarkers including monosaccharides, lignin derivatives, saccharides, sterols, and polyols in the ICP; (ii) explore the applicability of PUF-PASs in collecting other BB-related biomarkers besides monosaccharides; (iii) determine the importance of MODIS unresolved fires by examining the discrepancy between BB-related biomarkers and MODIS active fires; and (iv) estimate the emissions of BB on atmospheric pollutants based on PAS derived levoglucosan concentrations. To the best of our knowledge, this is the first study to reveal the importance of MODIS unresolved fires by field-based monitoring of BB at a regional scale.

2. Materials and methods

2.1. Study area and sampling campaigns

A map of the sampling sites is shown in Fig. 1a. Southwest China (SWC) borders the ICP. A total of 42 locations including urban, suburban, and rural sites were selected. There were 3 sites in SWC and 39 sites in the ICP. Among the ICP sites, 6 sites were in Cambodia, 16 in Vietnam, 10 in Laos, and 7 in Thailand. The sampling campaign lasted from January to September 2016 in three deployment periods, each for 3 months (i.e., January–April, April–June, and June–September), assumed to be open BB season, pre-monsoon season, and monsoon season, respectively. Each sampling period (44–71 days) was within the 73-day linear exposure of levoglucosan according to our previous calibration exercise of PUF-PASs in the Pearl River Delta, China (Jiang et al., 2018). The air volumes of levoglucosan were expected to be in the range of 48–85 m^3. However, for logistical reasons, the samples in Thailand and Vietnam were successfully retrieved for only one period or two periods, respectively. Details of each sampling site and the sampling campaign are given in Tables S1-S2. Most of the sampling sites of the ICP are in a monsoon tropical climate zone, and those of the north ICP and SWC are in a humid subtropical climate zone, as is the Pearl River Delta, China. The average temperature (23–30 °C) and the average relative humidity (55 % – 86 %) during the sampling periods for each location were close to those of our previous calibration exercise period of July–October in the Pearl River Delta (25–31 °C, and 61 % – 85 % relative humidity). Wind during the deployment periods was generally calm with wind speeds in the range 2.4–4.1 m/s. Since this range is below the high wind speeds (>5 m/s) that might experience nonlinear aerodynamics (Herkert et al., 2018), we expect sampling rates to be fairly uniform across sites and over the duration of sampling.

The passive samplers used in this study consisted of a PUF disk (14.0 cm diameter, 1.35 cm thickness, 365 cm^2 surface area, 3.40 g mass, 207 cm^3 volume, and 0.0213 g/cm^3 density) housed in two stainless steel domed chambers (upper, 28 cm diameter and lower, 24 cm diameter) to dampen the effect of wind speed on the uptake rate and to protect the PUF disks from precipitation, direct particle deposition, and ultraviolet radiation (Wilford et al., 2004; Zhang et al., 2008). The PUF disks were precleaned by Soxhlet extraction with a mixture of methanol and dichloromethane (7:93, *v/v*) for 36 h. The chambers were precleaned and solvent-rinsed with methanol beforehand. The samplers were assembled at the deployment sites to avoid contamination during transit. After deployment, the PUF disks were retrieved, resealed by seal bags, and returned to Guangzhou where they were stored and kept at −20 °C until analysis.

To compare across different sites at a regional scale and to avoid the induced uncertainties, we chose to use ng/d to represent the masses of BB-related biomarkers collected in a single PUF disk over the deployment period as the unit of the PAS derived TWA concentrations of BB-related biomarkers. The concentrations in ng/m^3 derived from the sampling rate of 1.1 m^3/d for levoglucosan based on the previous calibration exercise of PUF-PAS (Jiang et al., 2018), were only necessary for emission estimations in this study.

2.2. Chemical analysis

Three species of monosaccharides (levoglucosan, mannosan, and galactosan) were analyzed quantitatively as cellulose biomarkers by using calibration curves and internal standard methods. Sixteen lignin pyrolysis products including 3,5-dihydroxybenzoic acid, syringyl derivatives (2,6-dimethoxyphenol, 4-allyl-2,6-dimethoxyphenol, syringic

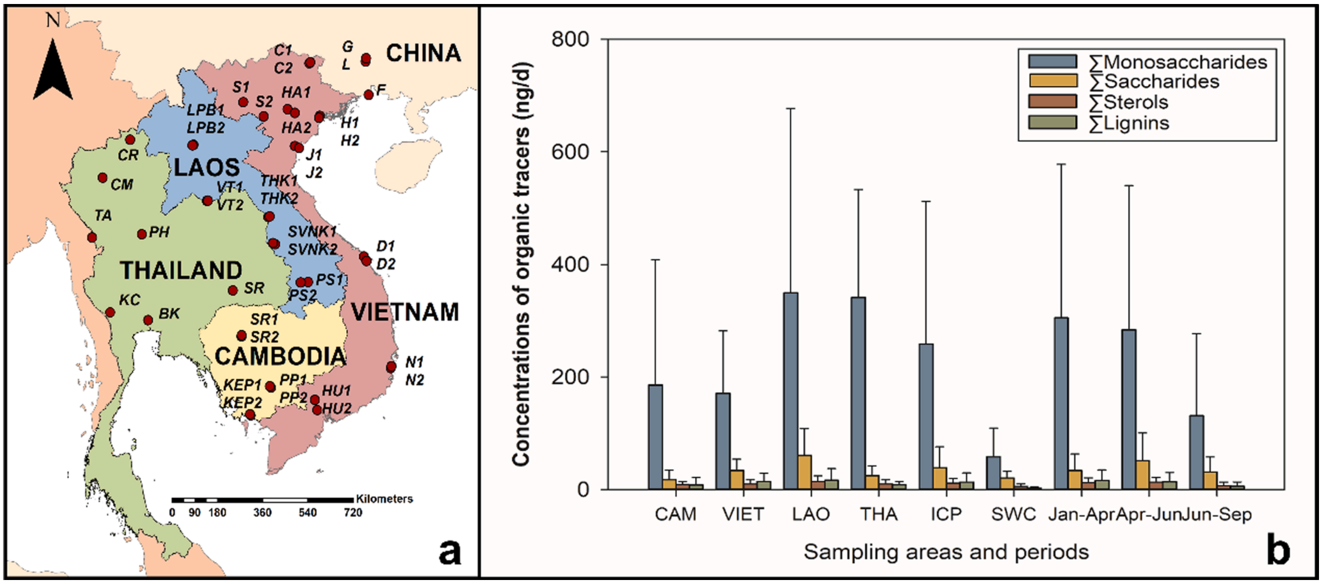

Fig. 1. a, Sampling sites in the Indo-China Peninsula (ICP) and Southwest China (SWC) in January–September of 2016. b, Comparison of BB-related biomarkers in different areas (CAM: Cambodia, VIET: Vietnam, LAO: Laos, THA: Thailand) and sampling periods. Background map was downloaded from https://www.arcgis.com/home/webmap/viewer.html. The details of sampling sites are listed in Table S1 of supplementary information. The unit of ng/d represents the masses of BB-related biomarkers collected in a single PUF disk over the deployment period.

acid, syringaldehyde, and acetosyringone), vanillyl derivatives (guaiacol, isoeugenol, vanillin, vanillic acid, acetovanillone), and coumaryl derivatives (p-coumaric acid, p-hydroxybenzaldehyde, p-hydroxyacetophenone, p-hydroxybenzoic acid, and ferulic acid) were analyzed to obtain information on the biomass species burned. In addition, seven other saccharides (saccharose, trehalose dihydrate, fructose, xylose, D-mannitol, *meso*-erythritol, and myo-inositol) were analyzed as biomarkers of hemicellulose, and five sterols or polyols (cholesterol, stigmasterol, glycerol, ergosterol, and coprostanol) were obtained from cooking sources, fungi, and animal feces, respectively. The gas chromatography-mass spectrometry (GC–MS) response factors of these target compounds were determined using authentic standards, and underwent relative quantitative analysis by the internal standard method. Chemical standards of monosaccharides were purchased from Toronto Research Chemicals. Chemical standards of other BB-related biomarkers were purchased from Dr. Ehrenstorfer GmbH, Germany and ANPEL, USA. The physical and chemical properties of these target compounds are shown in Table S3.

A surrogate standard of methyl-β-D-xylanopyranoside (MXP) was added to each of the samples (PUF) prior to Soxhlet extraction with a mixture of methanol and dichloromethane (7:93, *v*/*v*) for 36 h and treated as a quantification internal standard before injection. Aliquots for the extracts were dehydrated by a column (13 mm × 200 mm) containing coarse anhydrous Na_2SO_4. The eluent solvent was concentrated with a rotary evaporator and blown down to 0.5 mL with pure nitrogen gas. A 100 μL aliquot of the extract was dried with nitrogen and derivatized with a 50 μL mixture of N, O-bis (trimethylsilyl) trifluoroacetamide (BSTFA) with 1 % trimethylsilyl chloride and pyridine (2:1, *v*/*v*) at 70 °C for 1 h in a water bath. After derivatization, the derivatives were dried with nitrogen, cooled to room temperature, and then diluted to 0.5 mL by adding *n*-hexane before a determination by GC–MS.

The product analyses were carried out on a GC/MS-QP2010 (Shimadzu, Japan) system with a TG-5 ms (30 m × 0.25 mm × 0.25 μm) capillary column operating under full scan mode. The oven temperature began at 65 °C for 1 min and increased to 295 °C (20 min hold time) at a rate of 5 °C/min. A splitless injection of a 1 μL sample aliquot was performed with a 4-min solvent delay time. Spectra were obtained in a mass range of 50–550 *m*/*z*.

2.3. Estimation of BB emissions on atmospheric pollutants

Few studies have estimated the regional BB impacts on atmospheric pollutants based on field measurement data. Here we tried to estimate the emissions of particulate matter (PM), organic carbon (OC), and elemental carbon (EC) from BB based upon a simplified model of interactions between the earth's geospheres in which data on the average concentrations of levoglucosan derived from PUF-PAS and emission factors of biofuels in the ICP are used (Weiss et al., 1971).

Assuming that levoglucosan was effectively removed mainly by rainfall from the atmosphere and with no atmospheric degradation, we take the density of the atmosphere to be 1.3×10^8 g/m^3, the weight of the atmosphere to be 5.1×10^{21} g, and a mean lifetime of 7.3 days for levoglucosan without atmospheric degradation (Li et al., 2021); thus the global annual emission of BB (E_{bb}, mass amount/yr) could be calculated as follows:

$$E_{bb} = C \times \frac{5.1 \times 10^{21}}{1.3 \times 10^8} \times \left(\frac{365}{7.3}\right) \times \frac{1}{R} \quad (1)$$

where C is the concentration of levoglucosan (ng/m^3), and R is the average days of rainfall frequency in a specific region. The average time between rains in the world is 10.1 days, while in the ICP, it is 2.8 days.

The annual emissions of environmental pollutants (PM, OC and EC) from BB ($E_{m,bb}$, mass amount/yr) in the ICP could be further roughly estimated as follows:

$$E_{m,bb} = E_{bb} \times \frac{EF_{i,t}}{EF_{i,lev}} \times \frac{A}{5.1 \times 10^8} \quad (2)$$

where $EF_{i,t}$ and $EF_{i,lev}$ are the emission factors of pollutant t and levoglucosan for vegetation i (g/kg fuel) in a specific region, A is the surface area of the ICP, and the global surface area is taken to be 5.1×10^8 km^2. In this study, the emission factors of PM, OC, EC and levoglucosan were acquired from particulate samples of controlled burning of typical biomass combustion in the ICP (Table S4) (Cui et al., 2018; Jin et al., 2018). The uncertainties of $E_{m,bb}$ were calculated from the relative standard deviations of levoglucosan sampling rate, levoglucosan concentrations in the ICP and combustion factors.

2.4. Quality assurance and quality control

Laboratory and field blank PUF disks were analyzed in the same manner as the samples. The analytes in the laboratory and the field blanks were significantly lower than the concentrations of the field samples. The analytes of all the blanks ranged of 0.03–11 ng/sample, which were typically less than 5 % of the sample amounts. The method detection limit (MDL) (0.23–12.7 ng/sample) was defined as the average of all blanks plus three times the standard deviation (Table S3). Field blank data were subtracted from all the samples. The sum of 3 monosaccharides, 7 other saccharides, 16 lignin pyrolysis products, 5 sterols or polyols, 5 syringyl, 5 vanillyl, and 5 coumaryl lignin derivatives were marked as ΣMonosaccharides, ΣSaccharides, ΣLignins, ΣSterols, ΣSyringyl, ΣVanillyl, and ΣCoumaryl, respectively. In this study, all the Spearman's coefficients were assessed to be significant at the 95 % or 99 % confidence limit by Student's *t* test.

2.5. Data acquisition

In this study, terra MODIS statistics of thermal anomalies / daily fire products (MODIS C6) were downloaded from NASA's Fire Information for Resource Management System. The data of GDP (PPP) per capita, urban population (%), and population density (/km^2) were taken from World Bank data of 2016 and a local government report of 2015. The average temperature, wind speed, and humidity were acquired from the website of the National Oceanic and Atmospheric Administration of U.S. (https://www.noaa.gov/web.html). All the maps for discussion were downloaded from https://www.arcgis.com/home/webmap/viewer.html.

3. Results and discussion

3.1. Spatiotemporal distribution of BB-related biomarkers

Generally, samples from the ICP countries had higher concentrations of ΣMonosaccharides (258 ng/d), ΣLignins (13.4 ng/d), ΣSaccharides (39.2 ng/d), and ΣSterols (11.5 ng/d) than those of samples from SWC (ΣMonosaccharides 58.4 ng/d; ΣLignins 2.02 ng/d; ∑Saccharides 20.4 ng/d; and ΣSterols 5.48 ng/d) (Fig. 1, Table S5-S7). The spatiotemporal concentrations of levoglucosan, mannosan, galactosan, ΣMonosaccharides, ΣSaccharides, ΣLignins and ΣSterols across the ICP and SWC regions are shown in Fig. 2a, Fig. S1-S5, and Supplementary Section 1. The highest concentrations of all types of BB-related biomarkers were observed in Laos, followed by the concentrations of ΣMonosaccharides in Thailand and ΣSaccharides and ΣLignins in Vietnam.

The concentrations of ΣMonosaccharides in January–April (ICP: 305 ng/d; SWC: 120 ng/d) and April–June (ICP: 284 ng/d; SWC: 19.5 ng/d) were generally higher than those in June–September (ICP: 131 ng/d; SWC: 35.9 ng/d) (Fig. S1, Table S5). A similar scenario was found for ΣLignin derivatives in the ICP, while the highest concentrations of ΣSaccharides in the ICP were observed in April–June and with no obvious differences of ΣSterols between January–April and April–June (Table S6). Nevertheless, at the onset of the summer monsoon starting in May, the significantly decreased concentrations of BB-related biomarkers in June–September were consistent with the variations in BB events captured by MODIS in Asia (Fig. S6).

Moreover, combining the background of the topographic map and the satellite map, the concentrations of levoglucosan were found to be the highest in the eastern Mekong plain (557 ng/d), surrounded by the west sides of the Annamite Mountains and Luang Prabang Mountains (red circle area of Fig. 2a), especially in April–June. On the east sides of the mountains, in contrast, the lowest concentrations of levoglucosan were found along the eastern coast of the ICP (194 ng/d). The concentrations of levoglucosan in hilly and mountainous topography with thick green forests was not as high as that in the river valleys and steep terrain of the mountainous landscape and the central plains of the ICP, where many cities are located.

Most of the urban sampling sites had higher BB intensity than the rural or suburban areas in the northeast ICP (red circle area of Fig. 2b), despite the existence of a negative relationships between BB-related biomarkers and population density ($r_s = 0.415–0.572$, $p < 0.05$, n = 25) (Table S8). Indeed, the average concentrations of ΣMonosaccharides in suburban or rural areas (293 ng/d) were higher than those in the urban areas (225 ng/d), but the seasonal variation was more notable in the urban area than the countryside during the sampling periods

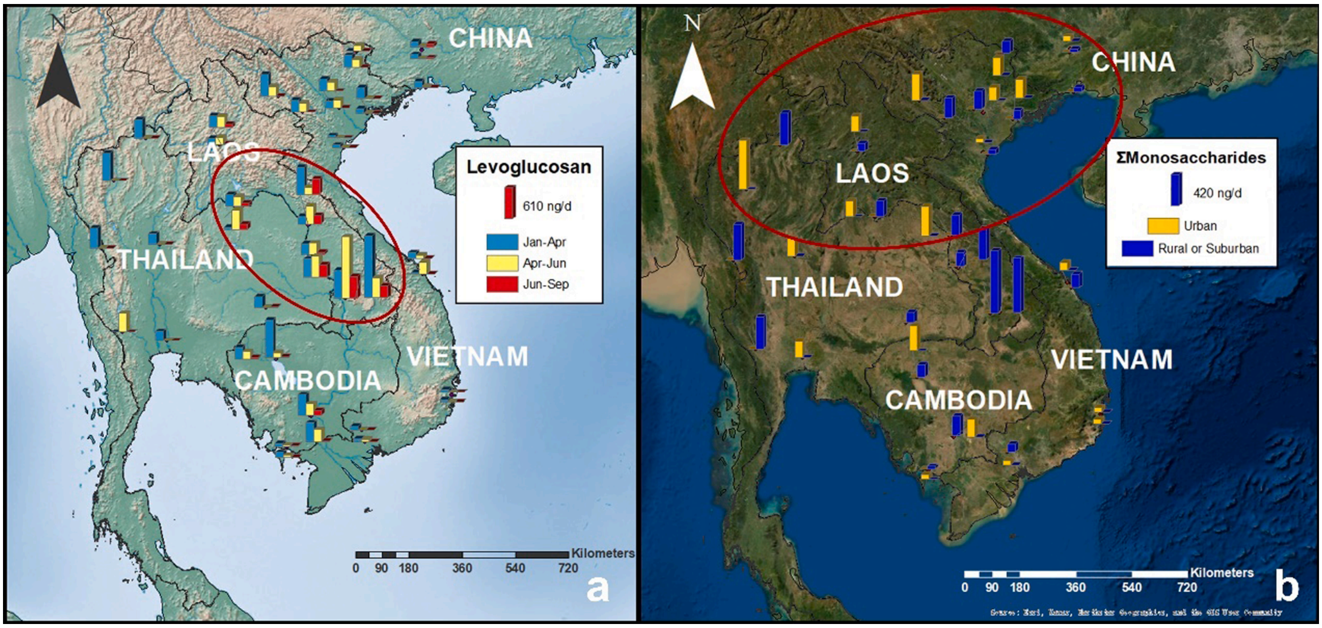

Fig. 2. a, Spatial and temporal distribution of levoglucosan in the Indo-China Peninsula and Southwest China in January–September of 2016. b, Spatial and temporal distribution of ΣMonosaccharides in the urban and rural or suburban areas of Indo-China Peninsula and Southwest China. The background topographic map and satellite map were downloaded from https://www.arcgis.com/home/webmap/viewer.html.

(Fig. 2b, Fig. S5, and Table S9), suggesting that the urban areas were influenced by the peripheral open BB.

3.2. Coupled BB-related biomarkers and source diagnostic ratios

The applicability of PUF-PAS in monitoring other BB-related biomarkers could be revealed by the positive Spearman and linear correlations between levoglucosan and other BB-related biomarkers besides monosaccharides ($\rho = 0.441–0.816$, $p < 0.01$, $n = 90$), especially for lignin derivatives (Table S8 and Fig. S7). This implied that their sources were identical and that the lignin markers and cellulose markers were not de-coupled by atmospheric degradation processes over the PAS deployment time, despite atmospheric saccharides and lignin pyrolysis products originating not only from BB but also from soil resuspension (Simoneit et al., 2004), biodegradation, and plant debris (Zangrando et al., 2016; Yan et al., 2019).

Additionally, except for abnormally high fire counts in CAM-SR and LAO-LPB, positive Spearman correlations were observed between several other BB-related biomarkers and open fire activities based on the MODIS fire data within a 50 km radius centered on the sampling sites which were counted by a previously developed numerical model (Table S10-S11) (Jiang et al., 2018). For example, positive relationships were observed for ΣSaccharides and lignin derivatives ($r_s = 0.352–0.397$, $p < 0.05$, $n = 38$). In particular, the average concentrations of ΣLignins, ΣSyringyl, ΣVanillyl, and ΣCoumaryl were all found to be positively correlated with the number of fire activities corresponding to each sampling site during January–April ($r_s = 0.364–0.483$, $p < 0.05$, $n = 41$). Suburban or rural areas had more relationships between the lignin derivatives and active fires than did urban areas during this open BB season. All these pieces of evidence demonstrated that most BB-related biomarkers besides monosaccharides derived from PUF-PAS could help reflect near-ground fire activities and could also track BB sources, especially in an open BB season.

Although active fires occurred randomly and unevenly within a specific area, the PUF derived levels of BB-related biomarkers could provide additional information about BB such as biomass/biofuel types or combustion conditions. Intriguingly, in the monsoon season, the ratios of levoglucosan to ΣLignins rose slightly from approximately 20 to 22, while the ratios of levoglucosan to mannosan (L/M) decreased from approximately 10 to 7. The ratios of levoglucosan to ΣLignins suggested an increase in nonwoody biomass combustion during this season (Shakya et al., 2011). It was reported that for the biofuels from Texas, USA, higher emissions of levoglucosan were released during high-temperature flaming compared with low-temperature smoldering because the thermal lability of mannosan and galactosan is higher than that of levoglucosan (Kuo et al., 2011). Usually, indoor biofuel combustion is conducive to smoldering conditions because of the still air conditions and low air supply resulting from the overload of fuel in stoves (Maruf Hossain et al., 2012). Household biofuels usually include agricultural residues other than firewood (Du et al., 2018; Luo et al., 2021). Therefore, the changed ratios might reflect the fact that indoor biofuel combustion was more prevalent in the monsoon season than in the open BB season.

3.3. The importance of MODIS unresolvable fires

Basically, significant Spearman correlations and even linear correlations were found between the average concentrations of monosaccharides and the number of fire activities corresponding to each sampling site over the whole sampling period ($r_s = 0.647–0.728$, $p < 0.01$, $n = 38$) (Table S11 and Fig. S8). However, these significant correlations could probably be attributed to the much higher number of fire activities and concentrations in January–April and April–June, overriding those of June–September.

There were distinctions between the number of fire activities corresponding to each sampling site and the average concentrations of BB-related biomarkers in each sampling period. Generally, not only in the open BB season (January–April), but also in the pre-monsoon season (April–June), positive correlations were found between monosaccharides or lignin derivatives ($r_s = 0.364–0.800$, $p < 0.05$) and MODIS fire spot counts, indicating the extensive influence of large-scale open BB events. However, these correlations between monosaccharides and MODIS fire data in the pre-monsoon season ($r_s = 0.410–0.465$, $p < 0.05$, $n = 28$) were weaker than those in the open BB season ($r_s = 0.703–0.800$, $p < 0.01$, $n = 41$). In comparison, in June–September, no significant positive relationships were observed between the MODIS fire data and BB-related biomarkers.

We plotted the levoglucosan concentrations and corresponding MODIS fire counts of 42 sampling sites over the three deployments (Fig. 3). A well-fitting linear correlation between levoglucosan and fire activities during January–April ($r_s = 0.703$, $r^2 = 0.65$, $p < 0.01$, $n = 41$) is used as the reference line. This season of the ICP is also dry season and it is less likely to be affected by rainfall events. The scope within the 95 % confidence interval of this fitting plot could all be deemed to be mainly influenced by open BB, while the inconsistency distributed outside the 95 % confidence interval of the fitting plot could be divided into the following two scenarios.

Samples distributed in the grey area in Fig. 3 are characteristic of depleted BB-related biomarkers against the frequent open fire activities. This could generally be ascribed to meteorological influences, especially the unstable meteorological conditions during the pre-monsoon season and monsoon season. Usually, the lower boundary layer and stable meteorological conditions during January–April could favor a near-ground accumulation of BB-related biomarkers; however, frequent precipitation, high humidity, and strong solar radiation would facilitate the removal or/and degradation of BB-related biomarkers in the atmosphere (Jiang et al., 2018). In fact, most of the samples in this area were collected at sites with a higher humidity, wind speed, or temperature than the average levels (Table S12).

Samples distributed in the blue area in Fig. 3 show elevated concentrations of BB-related biomarkers but few MODIS fire counts. Since long-range air mass transport was considered to be a less influence factor compared to the local emissions in the ICP, this phenomenon indicates a significant contribution from BB emissions other than the MODIS-resolved open fires, such as indoor biofuel combustion, widespread small-scale BB incidents and charcoal fires.

With the arrival of the monsoons, indoor biofuel combustion becomes prominent, especially in suburban or rural areas of the ICP. Typical household biofuel-stoves have low efficiency and contribute high pollutant emissions to ambient and indoor air pollution (Du et al., 2020; Du et al., 2018; Shen et al., 2020). High indoor concentrations of polycyclic aromatic hydrocarbons and their nitro derivatives have been confirmed to mainly originate from indoor biofuel combustion during cooking in rural areas of Thailand (Orakij et al., 2017). Even in a low-density residential area, solid fuel burning can result in extreme air pollution (Lin et al., 2018).

Small-scale BB incidents and charcoal fires were also non-negligible. Charcoal fires used for food grilling and frying outdoors and indoors are widely popular in Laos and Cambodia, and barbecue stands are especially common in resorts. Compared with the small number of monosaccharides emitted from family meat cooking, these sources emit more levoglucosan into the atmosphere (Wu et al., 2021). Likewise, considering the local coal consumption and an economic level that is inferior to that of Southwest China (Wu et al., 2021), the contribution of these MODIS unresolved fires to urban levoglucosan in the ICP might be greater than those from other non-BB sources such as domestic and industrial coal burning (Yan et al., 2018), and open waste burning (Christian et al., 2010).

It is notable that the abnormally high fire counts of April–June occurred in the sampling sites located at two famous heritage cities, Siem Reap (CAM-SR) and Luang Prabang (LAO-LPB), although their concentrations of BB-related biomarkers did not peak synchronously.

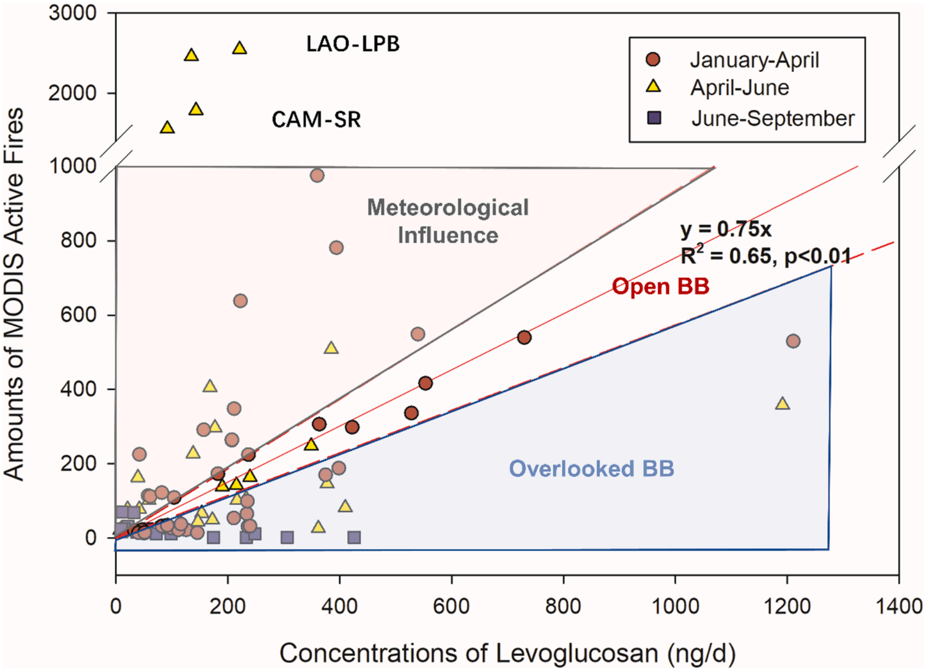

Fig. 3. Correlations between MODIS active fires within a 50 km radius centered at the sampling sites and concentrations of levoglucosan during different sampling periods in the Indo-China Peninsula and Southwest China in 2016. A fitting linear plot with a 95 % confidence interval was established between fire activities and levoglucosan during January–April. The sampling sites distributed in the grey and blue areas presented two contrary scenarios, respectively. The details of sampling sites are listed in Table S1 of supplementary information. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

These scenarios were probably not completely triggered by BB incidents. Siem Reap and Luang Prabang are Buddhist centers of Cambodia and Laos, respectively, where numerous temples are located. The number of vigorous active fires rose during the Songkran Festival in April, probably due to the frequent burning of fireworks and ritual items recorded by MODIS. It is reported that firework burning is one of the major contributors to non-BB sources of levoglucosan in China, accounting for 9.6 % of the total emission (Wu et al., 2021). However, due to the limited burning amounts of fireworks and ritual items compared to BB, the levoglucosan levels in these two sampling sites during this period (91–221 ng/d) were still slightly lower than those during the BB season (116–730 ng/d).

4. Implications for biomass burning emissions

The BB biomarker data could enable an observation-based evaluation of the impact of regional BB emissions on the atmosphere. With the inclusion of MODIS unresolved fires, the BB emissions of atmospheric pollutants in the ICP were estimated based on the measured levoglucosan concentrations, then compared with bottom-up estimations by remote sensing data or by integrating the emission factors and MODIS data (Chang and Song, 2010; Cui et al., 2018; Li et al., 2021; Oanh et al., 2018; Shi et al., 2015; Shi and Yamaguchi, 2014; Xu et al., 2022). Although there are anthropogenic, non-BB emissions of levoglucosan, most are minor and are likely still related to the pyrolysis of cellulose (Li et al., 2021; Wu et al., 2021; Yan et al., 2018). The concentrations of levoglucosan derived by the corresponding sampling rate of 1.1 m^3/d of PUF-PAS (Table S5), as adapted from our previous calibration exercise in the Pearl River Delta, could be comparable to those in the aerosols of Chiang Mai and Bangkok, Thailand (Tao et al., 2020; Thepnuan et al., 2019; Wang et al., 2020).

The annual emission of levoglucosan was estimated to be 3.83 Tg/yr in this study, using the average levoglucosan concentrations (197 ng/m^3) of January–September; this was comparable with the median of available scattered levoglucosan data (151 ng/m^3) in the world from several compiled studies of aerosols (Li et al., 2021). This estimated levoglucosan emission value was quite close to the calculated emissions of Li et al. (2021) (3.81 Tg/yr).

The estimated emissions from BB were 4584 Gg/yr for PM, 1476 Gg/yr for OC, and 218 Gg/yr for EC in the ICP. As shown in Fig. 4, these results were at the same magnitude, and consistent for OC and EC emissions or slightly higher for PM emissions when compared with those of bottom-up estimations in Southeast Asia from 1997 to 2016 (Table S13). The reported emission ranges of PM, OC and EC were 1780–4000 Gg/yr, 340–3318 Gg/yr and 32–383 Gg/yr, respectively (Chang and Song, 2010; Oanh et al., 2018; Randerson et al., 2017; Shi and Yamaguchi, 2014). The estimated EC emission of this study was slightly lower than the estimated total BC emission fluxes from BB in Southeast Asia which was measured by radiocarbon (~310 Gg/yr) (Liu et al., 2020). Moreover, all the emissions of this study were higher than those of previous reported emissions of PM (1527–2834 Gg/yr), OC (530–1100 Gg/yr) and EC (81–150 Gg/yr) in the mainland Southeast Asia study of Cui et al. (2018).

Considering only open BB was taken into consideration for all these bottom-up studies, the inclusion of MODIS unresolved fires by field-based monitoring data would give rise to higher estimated BB emissions. In fact, due to the limitations of MODIS data (e. g. persistent cloud cover, small fire, short duration, etc.) (Benali et al., 2016), different information such as emission factor, burned area, biomass density and combustion intensity would all influence the estimations of bottom-up emissions (Cui et al., 2018). Although the uneven BB distribution all over the world would also inevitably lead to the uncertainty of simple conversion based on surface area ratios, this simply calculated estimation could also be used as a reference. Further accurate estimations of BB emissions based on monitoring data are urgently needed in the future. Finally, the emission uncertainties were calculated based on the relative standard deviation of levoglucosan sampling rate, levoglucosan concentrations in the ICP, and combustion factors applied in this approach, which were assumed to be around 24 %, 100 % and 30 %, respectively (Jiang et al., 2018; Cui et al., 2018). Therefore, the estimated top-down emissions (minimum–maximum) were 4100–5070 Gg/yr for PM, 1180–1780 Gg/yr for OC, and 10–420 Gg/yr for EC, respectively.

The annual estimated BB emissions of atmospheric pollutants of different countries in the ICP are summarized in Fig. S10 and Table S14, and were almost twofold higher than those estimated in 2003 (Streets et al., 2003). The highest annual emissions of PM, OC, and EC were found in Thailand, followed by Laos, Vietnam and Cambodia. The change in the order of BB emissions in Vietnam and Laos over the past

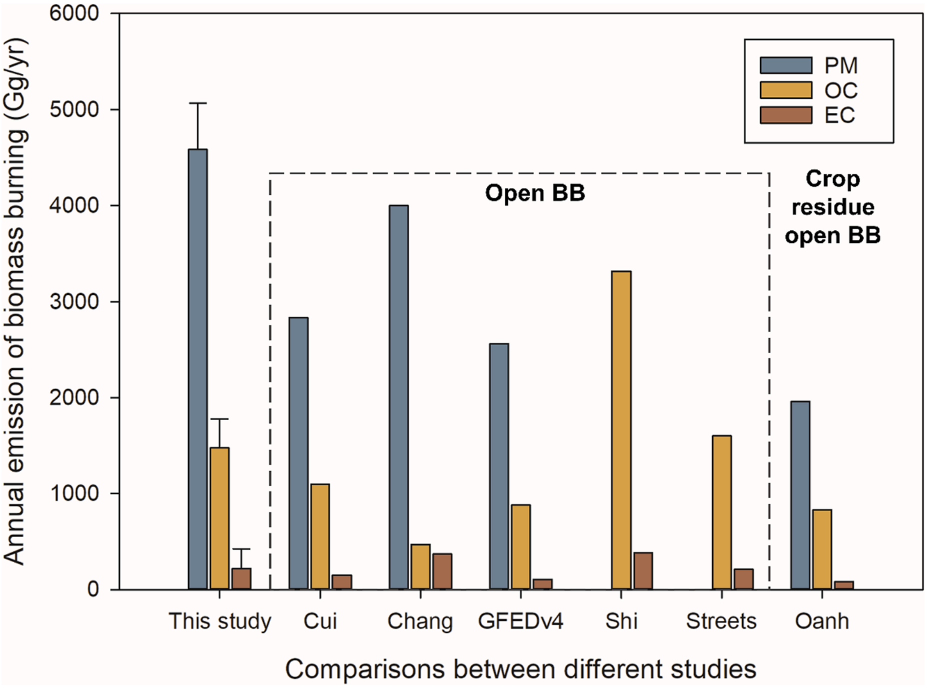

Fig. 4. Comparisons of annual emissions of biomass burning on atmospheric pollutants between the bottom-up emissions of Southeast Asia and estimated emissions of this study. Here the previously reported emissions were shown as the highest estimations. The data of Cui, Chang, Shi, Streets, Oanh and GEEDv4 were originated from the studies of Cui et al.(2018), Chang and Song (2010), Shi and Yamaguchi (2014), Streets et al.(2003), Oanh et al. (2018), and GFEDv4 dataset (Randerson et al., 2017), respectively.

13 years revealed the changed energy structure with the economic development in the ICP to a certain extent. As a representative developing region with an over-reliance on BB for agriculture and energy access, ICP countries recorded estimated BB emissions that were generally positively related to the GDP per capita and the urban population. However, of the countries with similar levels of GDP per capita, urban population, and urban population growth from 2003 to 2016, Laos had the highest GDP growth among the ICP countries, implying that BB might play an important role in the Laos economy as energy.

The inclusion of MODIS unresolved fires highlights the fact that these fires (indoor biofuel combustion / small-scale BB incidents / charcoal fires) have a non-negligible impact on the regional air quality, as evidenced in the estimated BB emission of atmospheric pollutants in the ICP. These MODIS unresolved fires accounted for a large deviation in the MODIS data in reflecting the near-ground BB intensity, as revealed by the discrepancy between the PAS derived levoglucosan and the number of MODIS active fires. Therefore, there is a need to recognize the significance of MODIS unresolved fire emissions and to evaluate their fugitive emission of air pollutants in order to improve the modeling of regional BB emissions. We therefore strongly suggest that the pollutant co-emissions of open BB and non-open fires should both be included in future efforts to mitigate BB, so as to strengthen the supervision of BB sources in both urban and rural regions, and to guide effective mitigation of BB emissions towards sustainable development. Last but not least, we call for accurate estimations of BB emissions based on more regional field measurements.

CRediT authorship contribution statement

Haoyu Jiang: Investigation, Data curation, Methodology, Formal analysis, Visualization, Writing - original draft. **Jun Li:** Conceptualization, Writing - review & editing. **Jiaqi Wang:** Methodology. **Hongxing Jiang:** Methodology. **Yangzhi Mo:** Visualization. **Jiao Tang:** Investigation. **Ruijie Zhang:** Investigation. **Wanwisa Pansak:** Investigation. **Guangcai Zhong:** Validation, Supervision. **Shizhen Zhao:** Validation, Supervision. **Jicai Ning:** Data curation. **Chongguo Tian:** Data curation, Formal analysis. **Gan Zhang:** Conceptualization, Supervision, Funding acquisition, Project administration, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary material

The data supporting the findings of this study are summarized in the supplementary information, and any other data analysed in the current study are available from the corresponding author upon request. Description of sampling campaign; physical-chemical properties and method detection limits of specific BB-related biomarkers; spatial and temporal plots of BB-related biomarkers; summary of BB-related biomarkers concentrations; number of fire activities based on the MODIS data within a 50 km radius centred at the sampling sites; correlations between fire activities and average concentrations of BB-related biomarkers; comparisons with bottom-up estimations; estimated BB emissions in different countries of the Indo-China Peninsula.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2022.107582.

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