Emissions and characteristics of particulate matter from rainforest burning in the Southeast Asia

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ABSTRACT

The burning of tropical rainforests in the Southeast Asia emits considerable particulate matter (PM), which has significant effects on air quality and human health. Lacking of reliable local EFPM for rainforest burning in the Southeast Asia is one of the most important causes for uncertainty of the estimated pollutant emissions. In this study, 23 types of rainforest plants, including herbaceous, shrubs, evergreen trees and deciduous trees were burned to determine emission factors of PM and multiple chemical species, including organic carbon (OC), elemental carbon (EC), water soluble ions (WSIs), and elements using a custom-made dilution system. EFPM for the four vegetation types was relatively higher for burning of deciduous trees, followed by evergreen trees, shrubs and herbaceous. EFPM in the Southeast Asia was higher than those in the North America, South America and Africa, with biomass type and ambient temperature and humidity as the determinant factors. Organic matter was the dominant constituent of PM, accounting for 57%, followed by EC, WSIs and elements. Source profile of WSIs varied larger than those of OC, EC and elements for different biomass type and areas. For example, the fraction of K⁺, a typical biomass burning tracer, to WSIs was 30% and 28% in the Southeast Asia and North America, respectively, which was almost 2 times lower than those in the Europe. Finally, the temporal and spatial scales of PM, OC, and EC emissions from rainforest burning in the Southeast Asia in 2016 were estimated.

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1. Introduction

Tropical rainforest is one of the most important global ecosystems, which plays an important role in global climate change, air quality, and ecosystem material cycles (Burgess et al., 2017; Liu et al., 2017a, 2017b, 2017c). Forest fires occur each year due to anthropogenic and natural factors. Large amounts of particulate matter are emitted from these fires, with significant adverse impacts on human health and air quality (Jian and Fu, 2014). As estimated by Andreae and Merlet (2001), the global PM emissions from tropical rainforests fires were about 11.3 Tg per year, accounting for about 13.7% of the total emissions from biomass burning.

There are three tropical rainforest systems in the world, geologically distributed in the Africa, South America, and Southeast Asia. Among which, Southeast Asia has been recognized as one of the most active areas for forest fires detected by moderate resolution imaging spectro-radiometer (MODIS). Southeast Asia (e.g., Afghanistan, Bangladesh, Bhutan, India, Maldieves, Nepal, Pakistan, and Sri Lanka) covers an area of 1.9 × 10^6 km^2 of rainforest, which accounts for almost 42% of its total land area. Streets et al. (2003) reported that the burning of biomass (including agricultural waste burning and wildfires) during the non-monsoon season in the South Asia (Zhang et al., 2017). It was reported that more than 50% of the BC deposition on Tibetan glaciers can be influenced by biomass burning (including agricultural waste burning and wildfires) during the non-monsoon season in the Southeast Asia (Zhang et al., 2017).

Accurate pollutant emissions factors are imperative for estimating emissions inventories and conducting air quality assessments. Three methods are commonly used to measure EFs of air pollutants emitted from forest burning, including laboratory measurements, field measurements (sampling plume directly at biomass burning site), and plume tracking (monitoring at the downwind of the burning site) (Alves et al., 2011; Amaral et al., 2016; Hsieh et al., 2016). Considering the dangers of field measurements and the excessive costs of plume tracking, open burning simulation study is an optimal choice for investigating emission characteristics from burning different types of rainforest plant.

By combining satellite images with pollutant EFs, emission inventories of different pollutants from forest burning in the Southeast Asia have been compiled in many studies (Andreae and Merlet, 2001; Chang and Song, 2010; Chaïyo and Garivart, 2014; Shi et al., 2014; Shi and Yamaguchi, 2014; Streets et al., 2003). However, the accuracy and timeliness of the EFs in these studies should be updated. Currently, there are several types of satellite products for assessing the burned areas, including the global fire emission database (GFED), MODIS, and L3JRC. Among which, MODIS has been demonstrated to be more accurate than the other two products (Chang and Song, 2010). However, emission inventories of PM developed in previous studies in the Southeast Asia were usually based on EFs determined from other tropical regions, which would increase the uncertainty of PM emissions. For example, the PM EFs (EFPM) measured in other regions summarized by Andreae and Merlet (2001) are still used to estimate the emissions inventory in the Southeast Asia till now without consideration of regional differences in fuel types and meteorological/burning conditions (Duncan et al., 2003; Giglio et al., 2013). It is crucial to evaluate whether there are differences in emission.

Table 1

| Categories            | Fuel name               | Moisture N | C     | H     | O     | C in ash |
|-----------------------|-------------------------|------------|-------|-------|-------|----------|
| Herbaceous            | Musa nana Lour.         | 9.33       | 1.92 ± 0.57 | 45.8 ± 1.76 | 5.71 ± 0.69 | 41.3 ± 7.94 | 16.0 ± 5.56 |
|                       | Rhynchelymus repens     | 8.73       | 0.97 ± 0.05  | 42.3 ± 0.17  | 5.96 ± 0.001 | 38.5 ± 0.26 | 12.0 ± 1.28 |
|                       | Eupatorium odoratum L.   | 12.42      | 0.95 ± 0.18  | 46.0 ± 0.12  | 5.4 ± 0.34  | 50.6 ± 0.05 | 52.8 ± 7.54 |
| Shrubs                | Lasiococa comberti Haines | 10.70     | 0.71 ± 0.18  | 44.0 ± 0.35  | 6.56 ± 0.15 | 47.6 ± 0.28 | 64.4 ± 10.6 |
|                       | Pseuderastachyum Plomphorm | 10.11     | 0.65 ± 0.02  | 48.7 ± 0.09  | 7.27 ± 0.05 | 47.5 ± 0.42 | 64.4 ± 10.6 |
|                       | Rauwolfieae verniculata  | 12.01      | 0.41 ± 0.01  | 48.6 ± 0.17  | 5.9 ± 0.02  | 49.3 ± 0.33 | 64.8 ± 10.1 |
| Evergreen trees       | Castanopsus Spach       | 10.78      | 0.78 ± 0.022 | 48.1 ± 0.12  | 6.19 ± 1.17 | 46.5 ± 0.18 | 35.7 ± 2.95 |
|                       | Antirias toxicaria Lesch | 13.75     | 1.5 ± 0.0     | 47.8 ± 0.21  | 6.28 ± 0.15 | 45.6 ± 0.28 | 66.9 ± 3.37 |
|                       | Toona ciliata Roem.     | 11.27      | 0.32 ± 0.01  | 49.4 ± 0.43  | 5.97 ± 0.14 | 45.5 ± 0.12 | 65.7 ± 13.0 |
|                       | Duabanga grandi          | 9.76       | 0.19 ± 0.27  | 48.4 ± 0.31  | 6.69 ± 0.27 | 44.5 ± 0.67 | 68.4 ± 9.39 |
|                       | Anthocephalus chinensis  | 9.49       | 0.33 ± 0.01  | 48.6 ± 0.11  | 5.86 ± 0.09 | 49.7 ± 0  | 81.7 ± 4.39 |
|                       | Morus alba               | 11.07      | 0.46 ± 0      | 44.4 ± 0.53  | 5.79 ± 1.22 | 44.5 ± 0.90 | 50.2 ± 7.17 |
|                       | Litchi chinensis Sonn.   | 12.46      | 0.66 ± 0.01  | 50.9 ± 0.35  | 5.68 ± 0.40 | 44.8 ± 0.10 | 86.0 ± 0.042 |
|                       | Pinales kevya var. lanhamenias   | 10.77     | 0.2 ± 0.28   | 47.9 ± 0.32  | 5.91 ± 0.40 | 47.7 ± 0.13 | 83.6 ± 0.163 |
|                       | Paramelia balliana       | 9.75       | 1.6 ± 0      | 45.3 ± 0.09  | 5.7 ± 0.01  | 43.9 ± 0.78 | 35.1 ± 7.19 |
|                       | PinnaspermunungahanieHue  | 13.62      | 0.21 ± 0.30  | 38.9 ± 0.04  | 5.47 ± 0.30 | 49.12 ± 0.09 | 27.2 ± 10.9 |
|                       | Cassia siamea           | 10.50      | 0.62 ± 0.12  | 45.7 ± 0.04  | 6.28 ± 1.37 | 43.8 ± 0.14 | 25.6 ± 3.77 |
|                       | Baccaraea ramiella Lour | 12.94      | 0.69 ± 0.02  | 47.3 ± 0.06  | 5.83 ± 1.20 | 48.1 ± 0.05 | 43.1 ± 6.73 |
|                       | Chatoecarpus castanocarpus | 22.62    | 0.86 ± 0.02  | 49.6 ± 0.12  | 6.87 ± 0.09 | 46.1 ± 0.23 | 77.9 ± 1.64 |
|                       | Citrus maxima            | 8.84       | 0.73 ± 0      | 49.6 ± 0.09  | 5.73 ± 0.01 | 23.7 ± 0.88 | 77.1 ± 1.72 |
| Deciduous trees       | Melia azedarach          | 13.12      | 0.00 ± 0.00  | 47.8 ± 0.14  | 6.12 ± 0.04 | 48.7 ± 0.3  | 64.8 ± 11.2 |
|                       | Bischofia polycarpa       | 10.34      | 0.49 ± 0      | 47.1 ± 0.08  | 5.53 ± 0.13 | 50.2 ± 0.95 | 50.6 ± 12.3 |
|                       | Brussonetia papyrifera    | 10.55      | 0.72 ± 0.02  | 45.7 ± 0.11  | 5.32 ± 0.02 | 55.8 ± 15.7 | 55.3 ± 28.6 |
factors and characteristics of PM as well as its constituents in emissions from different tropical rainforest regions worldwide and examine how much difference between them.

The objectives of this study are 1) to measure emission factors of PM and its constituents for various types of tropical rainforest vegetation in the Southeast Asia based on a real-world open burning experiment, 2) to compare emission factors of PM and its compositions from rainforest burning in the Southeast Asia and other tropical regions worldwide, and 3) to estimate emissions of PM, organic carbon, and elemental carbon from tropical rainforest burning in the Southeast Asia in 2016 on temporal and spatial scales.

2. Methodology

2.1. Sampling

Pollutant emissions from forest burning, as a special form of combustion, are difficult and dangerous to measure in the field. Thus, an open burning experiment was designed to carefully simulate the burning of tropical rainforest in the field. The open burning experiments were carried out in 2016 during summer in Yunnan province, China where ambient conditions were identical with those in the Southeast Asia. Twenty-three Southeast Asian tropical rainforest plants were collected, and the detailed information of these plants was given in Table 1. Generally, these 23 tropical rainforest plants were classified into four types including herbaceous plants, shrubs, evergreen trees, and deciduous trees. Raw materials were air-dried for several days, and the size of plants was approximately 20 × 3 × 2 cm³ to fit in the combustion installation.

First, homologous wood was used for ignition in a 40-cm-diameter stainless-steel bowl, which was composed with sufficient air supply. Then well-prepared fuel log collected directly from rainforest in the Southeast Asia was added to the combustion stainless steel plate. Finally, the smoke was collected through a self-designed dilution system. The sampling system consisted of a dilution tunnel, a residence time chamber, three PM samplers, an Andersen 8-stage sampler, and several temperature and humidity sensors (Fig. 1). Every plant type was burned three times, approximately 1–2 kg of fuel per burn. Fuel and ash were weighed before and after combustion. Quartz filters for PM sampling were prebaked at 450 °C for 4.5 h before sampling and were then stored in a refrigerator at −20 °C after sampling prior to analysis. Dilution ratios of each experimental process were calculated using the CO₂ concentrations before and after dilution.

2.2. Chemical analysis

Before and after sampling, the filters were conditioned at 25 °C and 40% relative humidity for 24 h before weighing. Then filters were weighed using a Mettler–Toledo electronic analytical balance with a precision of 10⁻⁴ g. The PM concentration was determined gravimetrically. Next, filters were used for the analysis of various chemical components, including OC, EC, water soluble ions, and multiple elements. The detailed analytical protocols have been described previously (Zong et al., 2015, Cui et al., 2016, 2017). Briefly, a punch with 0.544 cm² quartz filter was cut to analyse OC and EC using the Desert Research Institute (DRI) Model 2001 thermal/optical reflectance (TOR) carbon analyser (Atmoslytic Inc., Calabasas, CA, USA) with the Inter-agency Monitoring of Protected Visual Environment protocol. The samples were first heated at increasing temperatures under pure helium atmosphere, at 140 °C, 280 °C, 480 °C, and 580 °C (for produce OC1, OC2, OC3, and OC4, respectively); and then at 580 °C, 740 °C, and 840 °C to determine EC1, EC2, and EC3 under a 2% O₂/98% He atmosphere. WSIs on the filters were extracted ultrasonically in 8 mL of ultrapure water, and the process was repeated four times. The extraction solutions were then combined while ensuring a constant volume of approximately 35 mL using ultrapure water. Finally, the WSIs were detected using ion chromatography (Dionex ICS3000, Dionex Ltd., Sunnyvale, California, USA). The WSIs included in this study were NH₄⁺, K⁺, Cl⁻, NO₂⁻, NO₃⁻, SO₄²⁻, PO₄³⁻, F⁻, and Br⁻. For elements, filters were first digested with purified nitric acid for 10 h at 120 °C, then, measured using inductively coupled plasma mass spectrometry (ICP-MS; ELANDRC II, PerkinElmer Ltd., Hong Kong). The target elements include Na, Mg, K, Ca, Ti, V, Cr, Mn, Fe, Cu, Zn, and Pb.

Quality assurance and control measures include filter duplicate and blank samples were examined for quality control. For OC and EC analysis, the duplicate samples were analysed after a batch of 10 samples to ensure that the error was within 5%. The filter blanks were subtracted from samples for all water soluble ions and elements data detected in this study.

Fig. 1. Custom-made particulate matter sampling system.
2.3. Calculation

2.3.1. EFs

The EFs of pollutants emitted from the burning of tropical rainforest vegetation were calculated by the carbon balance methods.

\[
C_{\text{fuel}} - C_{\text{ash}} = R_{\text{FG}} \times (c(C_{\text{CO}_2}) + c(C_{\text{CO}}) + c(C_{\text{OC}}) + c(C_{\text{EC}}))
\]

where \( C_{\text{fuel}} \) and \( C_{\text{ash}} \) represent the amount of carbon (g C kg\(^{-1}\) in dry fuel and ash, respectively; \( R_{\text{FG}} \) is the flue gas emission rate (m\(^3\) kg\(^{-1}\) fuel); and \( C_{\text{CO}_2}, C_{\text{CO}}, C_{\text{OC}}, \) and \( C_{\text{EC}} \) represent the mass concentrations of carbon as CO\(_2\), CO, OC, and EC (g C m\(^{-3}\)) in the flue gas, respectively. It should be noted that the concentration of CO was unavailable in this study because the CO sensor did not work well in the field work. In order to avoid overestimating CO emissions from forest burning, modified combustion efficiency (MCE) values reported by many other studies were reviewed and listed in Table S1. MCE value was calculated by CO\(_2\) concentration divided by sum concentrations of CO\(_2\) and CO (MCE = \( c(CO_2)/(c(CO_2)+c(CO)) \)). As shown in Table S1, the MCE values reported by other studies ranged from 0.7 to 1, with an average of 0.91 ± 0.07. Therefore, the average MCE (0.91 ± 0.07) value was used in this study to calculate the CO concentration. According to the equation of MCE, 10% of CO\(_2\) concentration was adopted for CO concentration finally.

Subsequently, the EFs of CO\(_2\), PM, OC, EC, WSIs and elements were calculated by \( R_{\text{FG}} \). The details are available in Cui et al. (2017).

2.3.2. Emissions amount

\[
\text{Emission} = \sum_i EF_{i,t} \times B_{i,j} \times F_{i,j} \times CF_i
\]

where \( EF_{i,t} \) is the EF of pollutant \( t \) for vegetation \( i \) (g kg\(^{-1}\) fuel); \( B_{i,j} \) is the burned area of vegetation \( i \) in pixel \( j \) (km\(^2\)); \( F_{i,j} \) is the above ground biomass density for vegetation \( i \) in pixel \( j \) (kg m\(^{-2}\)); and \( CF_i \) is the combustion factor for vegetation \( i \).

The rainforest vegetation of interest in this study includes seven types: broadleaf evergreen forest, broadleaf deciduous forest, needle leaf evergreen forest, needle leaf deciduous forest, herbaceous (herbaceous and herbaceous with sparse trees), shrubs, and others (mixed forest, open trees, and sparse vegetation), according to the land cover data (version 3) from the Global Map database (https://globalmaps.github.io/). It should be noticed that the EFs used for vegetation designated as “others” were the average EFs for deciduous and evergreen forest plants in this study. Furthermore, the burned area data were from the MODIS active fire product (MCD45A1) with a resolution of 500 m (https://e4ftl01.cr.usgs.gov/MOTA/). The burned area for each plant \( i \) was determined by overlapping the MCD45A1 product with Global Map data. \( F_{i,j} \) was determined using data reported by Chang and Song (2010). The reported combustion factors of different types of vegetation can vary greatly (Shi and Yamaguchi, 2014). Therefore, types of vegetation have been considered in CF value calculation this study. The values were acquired by reviewing studies about PM emission inventories (Jain et al., 2006; Shi et al., 2014). In this study, the CF\(_i\) was 0.3 ± 0.09, 0.6 ± 0.18 and 0.9 ± 0.27 for coarse woodland, shrubs, and herbaceous vegetation, respectively.

2.3.3. Uncertainty of emission

The uncertainties of pollutant emissions were estimated based on the method of error transmission. Emission uncertainty is associated with EF, burned area, fuel load, and combustion factor, as described by Eq. (2).

\[
U_i^2 = \sum U_{i,j}^2
\]
\[(U_i/AVE_i)^2 = (U_{AV}/AVE_{AV})^2 + (U_{AV}/AVE_{EF})^2 + (U_{EF}/AVE_{EF})^2 + (U_{EF}/AVE_{CI})^2 \]  (4)

Where \(U_i\) is the emission uncertainty of pollutant \(i\), \(U_{AV}\) is the emission uncertainty of pollutant \(i\) for vegetation \(i\), \(U_{AV}/AV_{AV}\) is the ratio of standard deviation and average value of burned area for vegetation \(i\); \(U_{AV}/AV_{EF}\) is the ratio of standard deviation and average value of emission factor for vegetation \(i\); \(U_{EF}/AV_{EF}\) is the ratio of standard deviation and average value of combustion factor for vegetation \(i\).

2.4. Statistical analysis

All data statistical analysis was performed using Statistical Package for Social Science (version 16.0, SPSS, Inc.). The correlations between EFPM and humidity among evergreen trees, and between EFK and EFK were determined using Pearson correlation. And Spearman’s rank correlation was applied to investigate the correlations between fractions of WSIs for the 4 types of biomass. \(p < 0.05\) was considered as statistically significant.

3. Results and discussion

3.1. PM emission factors

The EFs of PM emitted from the open burning of 23 different tropical plants are shown in Fig. 2 and Table 2. The overall average EFPM for these 23 species was 21.3 ± 14.2 g kg\(^{-1}\) fuel, ranging from 3.23 ± 1.62 to 58.1 ± 22.2 g kg\(^{-1}\) fuel. The highest average EFPM for the four vegetation subtypes was deciduous trees (26.3 ± 16.1 g kg\(^{-1}\) fuel), followed by evergreen trees (21.1 ± 24.4 g kg\(^{-1}\) fuel), herbaceous (19.7 ± 12.6 g kg\(^{-1}\) fuel), and shrubs (18.8 ± 8.87 g kg\(^{-1}\) fuel), which was associated with the extent of lignin content of the vegetation and the burning conditions. It was reported that higher lignin contents could make plants more toughness and stoutness, which accounted for 7.4%–12.2% of lignocellulose for herbaceous and 21%–25% for deciduous and evergreen trees (Guo et al., 2008; Yang, 2016). A wide range of EFPM was found for evergreen trees, which was probably due to a large variation in humidity among the trees (Pearson correlation between EFPM and humidity: \(R = 0.68\), \(p < 0.01\)). The different EFPM of the four types of tropical forest vegetation in this study indicated that there was large uncertainty when only a single EFPM was used to represent all types of tropical forest vegetation. It has been reported that EFPM emitted from forest vegetation burning was 16 times higher than those from dambo grass (Aurell and Gullett, 2013; Sinha et al., 2003) and higher than those from other biomasses, such as extra-tropical forest and savanna grassland (Andreae and Merlet, 2001).

3.2. Comparison of PM emission factors for different regions and burning methods

Through comparison of different EFPM value reported in references, there were large differences in EFPM values depending on areas (etc. meteorological) and ways that biomass was burned. As shown in Fig. 3, the average EFPM was highest in the Southeast Asia, followed by North America, South America and Africa. Different average EFPM values were mainly attributed to diversity of wood species, discrepancy of ambient temperature and humidity. EFPM in Africa was the lowest mainly because the main biomass species is grass which could burn more sufficiently compare with other wood types. For further discussion of the EFPM difference in different regions, the average ambient temperature and humidity in the world since 1981 were acquired (see Figs. S1 and S2 in supporting information). It is hotter and dryer than other

| Table 2 | Emission factors of PM and its constituents for 4 types of biomass (mg kg\(^{-1}\) fuel). |
|---------|---------------------------------------------------------------|
|          | Herbaceous | Brush       | Evergreen trees | Deciduous trees |
| Ave     | SD    | Ave    | SD    | Ave     | SD    | Ave    | SD    |
| PM\(^{a}\) | 19.7  | 12.6   | 18.8  | 8.87  | 21.1  | 24.4  | 26.3  | 16.1  |
| OC\(^{b}\) | 6.21  | 4.78   | 6.39  | 3.07  | 8.58  | 10.4  | 10.8  | 9.64  |
| EC\(^{c}\) | 1.00  | 0.38   | 1.13  | 0.64  | 1.13  | 0.90  | 1.27  | 0.99  |
| Na      | 4.21  | 7.29   | 11.2  | 15.8  | 10.1  | 21.2  | 9.10  | 15.8  |
| Mg      | 8.82  | 7.16   | 21.9  | 27.1  | 16.4  | 32.4  | 22.4  | 22.6  |
| K       | 412   | 474    | 119   | 90.6  | 92.0  | 103   | 177   | 149   |
| Ca      | 41.5  | 43.5   | 108   | 134   | 82.6  | 133   | 90.0  | 119   |
| Ti      | 0.51  | 0.89   | 1.75  | 2.15  | 2.07  | 4.31  | 2.04  | 2.98  |
| V       | 0.03  | 0.05   | 0.08  | 0.08  | 0.07  | 0.13  | 0.13  | 0.10  |
| Cr      | 1.26  | 1.87   | 0.35  | 0.31  | 0.37  | 0.49  | 1.15  | 0.84  |
| Mn      | 0.75  | 1.11   | 0.66  | 0.54  | 0.34  | 0.30  | 1.56  | 2.02  |
| Fe      | 15.4  | 20.4   | 40.3  | 47.3  | 23.1  | 49.8  | 24.2  | 14.5  |
| Cu      | 0.22  | 0.21   | 0.35  | 0.41  | 0.11  | 0.11  | 0.14  | 2.99  |
| Zn      | 2.51  | 3.97   | 22.0  | 37.5  | 6.07  | 13.3  | 11.8  | 9.58  |
| Ni\(^{d}\) | 7.84  | 13.6   | 0.00  | 0.00  | 0.88  | 3.29  | 253   | 438   |
| K\(^{+}\) | 356   | 392    | 99.8  | 76.1  | 96.1  | 89.1  | 238   | 239   |
| PO\(^{4}\) | 22.6  | 39.2   | 13.5  | 23.4  | 79.0  | 153   | 25.0  | 43.3  |
| NO\(^{2}\) | 20.8  | 32.3   | 3.77  | 6.54  | 1.92  | 5.69  | 0.00  | 0.00  |
| F\(^{−}\) | 107   | 175    | 10.9  | 11.0  | 11.9  | 13.0  | 6.23  | 4.46  |
| Cl\(^{−}\) | 198   | 125    | 393   | 556   | 39.6  | 31.7  | 701   | 1077  |
| Br\(^{−}\) | 59.1  | 81.7   | 10.6  | 12.0  | 18.4  | 16.3  | 26.8  | 27.7  |
| NO\(^{3}\) | 7.43  | 63.3   | 5.53  | 5.68  | 13.2  | 15.2  | 8.75  | 4.23  |
| SO\(^{2−}\) | 163   | 174    | 127   | 163   | 85.2  | 83.6  | 103   | 137   |

\(a\): units (g kg\(^{-1}\) fuel).
3.3. Characteristics of PM constituents from forest burning

Table 2 and Fig. S3 show the emission factors of PM constituents and PM mass balance for each type of biomass. The total amount of constituents analysed (organic matter (OM) = 1.6 \times OC, EC, WSIs, and elements) accounted for 66.5 \pm 19.2\% of the total PM. The proportion of average PM chemical constituents from burning of tropical forest species in this study was similar to that reported by Alves et al. (2011) (e.g., carbonaceous components: 52 \pm 20\%; WSIs: 2.6\%; elements: 1.23\%).

Fig. 3. Comparison of EFPM for different sampling regions and methods reported in different studies.

Fig. 4. Relationships among EFPM, EF\text{OC}, EF\text{EC}, OC/TC, and EC/TC.
3.3.1. OC and EC

For the four types of tropical vegetation, the EF_{OC} increased from 6.21 g kg\(^{-1}\) fuel (herbaceous) to 10.8 g kg\(^{-1}\) fuel (deciduous trees), and EF_{EC} increased slightly from 0.998 g kg\(^{-1}\) fuel (herbaceous) to 1.27 g kg\(^{-1}\) fuel (deciduous trees). The trends of EF_{OC} and EF_{EC} from herbaceous to deciduous obtained in this study could be explained by the conclusion drawn by Alves et al. (2011), who found that EF_{OC} increased considerably when the combustion conditions changed from flaming to smouldering, whereas EF_{EC} seemed to stabilise between these two phases. Similarly, herbaceous vegetation is non-compat and easily combustible with O\(_2\), compared to compact deciduous trees, which could cause more frequently flame phase for herbaceous.

It was obvious that OM was the dominant constituent of PM from the 4 types of forest biomass burning, accounting for 56.9 ± 18.0% of the PM mass. The proportion of OM to PM from burning tropical forest species in this study was similar to that reported by other studies (Ferek et al., 1998; Schmidl et al., 2008; Alves et al., 2011). The range of OC/EC from the 23 tropical biomass species was from 2.41 to 24.0, and the variation of OC/EC ratios in this study was lower than that reported by Alves et al. (2011). Different pollutant EFs in this study might be attributed to thermodynamics, although the modified combustion efficiency (MCE) was not obtained. Generally, EF_{PM} and EF_{OC} increased with decreasing MCE values, while EC/TC values increased greatly at higher MCE values (Chen et al., 2007). As shown in Fig. 4, EF_{PM} and EF_{OC}, EF_{PM} and EF_{EC}, and EF_{PM} and OC/TC were significantly positively correlated, which indicated EF_{PM}, EF_{OC}, EF_{EC} and OC/TC were increased with MCE values. However, there was a significant negative correlation between EF_{PM} and EC/TC (p < 0.05). These relationships might indicate that the high EFs and variations in OC/EC ratios are dependent on combustion processes.

3.3.2. Water soluble ions

The range of total WSIs emission factors was 92–3558 mg kg\(^{-1}\) fuel, with average of 598 ± 820 mg kg\(^{-1}\) fuel, accounting for 0.49–9.0% of total PM. The average total EFs of WSIs in this study was obviously higher than the results reported by Sen et al. (2014) (383 ± 301 mg kg\(^{-1}\) fuel) and Alves et al. (2011) (453 ± 504 mg kg\(^{-1}\) fuel). However, Alves only detected five ions, K\(^+\), NH\(_4\)^+, Cl\(^-\), SO\(_4\)\(^2-\), and NO\(_3\)\(^-\), which was less than the quantity of ions detected in this study. When the other detected ions obtained in this study were added to calculate the amount of WSIs, the total EF_{WSI} in Alves’s research could reach to 560 mg kg\(^{-1}\) fuel. For individual species of WSIs, the average EFs of NH\(_4\)^+ for forest biomass burning in this study were in good agreement with those from Sen et al. (2014), and the average EFs of K\(^+\) and SO\(_4\)\(^2-\) were consistent with those from Alves et al. (2011) (see Fig. 5). The EF of Cl\(^-\) measured in this study was significantly higher than those reported by Alves et al. (2011) and Sen et al. (2014), while the EFs of NO\(_3\)\(^-\) and NO\(_2\)\(^-\) were lower than these references. Underestimated EF_{NO\(_3\)\(^-\)} in Alves et al. (2011) was caused by some extent from their low combustion temperature.

Fractions of water soluble ions to PM for 4 types of biomass are shown in Fig. 6. Cl\(^-\), K\(^+\), and SO\(_4\)\(^2-\) were the most abundant ions in most of the samples, whereas NH\(_4\)^+ level was the highest from deciduous trees burning. Except for deciduous trees, there were significant correlations of water soluble ions fractions between pairs of vegetation types, including herb, shrub and evergreen trees, indicating that the compositions of WSIs only varied slightly among the vegetation types. The different compositions of WSIs for deciduous trees from other rainfall forest biomass were mainly due to the habitat variation and the different abilities to absorb nutrients from the soil. K\(^+\) emitted from biomass burning exhibited highly significant positive correlations (p < 0.01) with most of the anions, including PO\(_4\)\(^3-\), F\(^-\), Cl\(^-\) Br\(^-\), and SO\(_4\)\(^2-\), which was consistent with results reported by Sen et al. (2014), who noted that K\(^+\) was significantly correlated with Cl\(^-\) and SO\(_4\)\(^2-\). Meanwhile, results by transmission electron microscopy demonstrated that crystal KCl and K\(_2\)SO\(_4\) could be clearly seen in individual particles from biomass burning (Liu et al., 2017a, 2017b, 2017c). It is noted that NH\(_4\)^+ and NO\(_3\)\(^-\) were not correlated with other ions, which might be attributed to complex combustion processes and detection limits for these two ions. The detection rates for NH\(_4\)^+ and NO\(_3\)\(^-\) were 13% and 96%, respectively. The detection limit for NH\(_4\)^+ in this study was 0.02 ppm, which caused several of NH\(_4\)^+ missing. However, the fraction of NH\(_4\)^+ was the highest for deciduous trees, different than those for other biomass. It was reported that less nitrogen accumulates as NH\(_4\)^+ since NH\(_4\)^+ are oxidized to NO\(_x\) at high temperature (Hegg et al., 1988; Alves et al., 2011).

As shown in Fig. 7, relative abundance of water soluble ions between Southeast Asia, North America, South America, Africa and Europe are compared. In general, K\(^+\) and Cl\(^-\) were the most abundant species in different regions. It was obviously that fraction of K\(^+\) in Southeast Asia and North America was 30% and 28%, respectively, which was almost 2 times lower than those in Europe, while fractions of Cl\(^-\) in Southeast Asia and North America were higher than those in other regions. For SO\(_4\)\(^2-\), fractions in different regions were consistent, ranged from 15% to 23%. It should be noted that the fractions of NO\(_3\)\(^-\) in Southeast Asia and Europe were negligible (2.2% and 1.5%) compared with other regions.

3.3.3. Elements

Total EFs of elements for herbaceous plants, shrubs, evergreen trees, and deciduous trees ranged from 233 mg kg\(^{-1}\) to 487 mg kg\(^{-1}\), accounting for 1.1–2.5% of the total PM. The variation in total EFs of elements in the four types of vegetation was relatively small. As shown in Fig. 6, K, Ca, Fe, Na, and Mg were the most abundant elements for almost all of the combusted forest vegetation, and the average of the sum of these five elements accounted for 95% of the total elements. Ribeiro et al. (2017) reported similar result that Ca, Na, and Mg were the dominant elements in three types of residual forest biomass burning emission. In addition, Corsini et al. (2017) measured elements (e.g., Al, P, Ti, V, Mn, Fe, Co, Ni, Cu, Zn, As, Sr, Mo, Cd, Ba, and Pb) emitted from burning wood and found that Fe had the highest concentration. However, Ferek et al. (1998) reported that Mn, Cu, and Zn were the dominant elements emitted from savannah burning, which differed significantly from the results of this study. In addition, K emitted from rainforest burning in the present study was significantly correlated with K\(^+\) (R\(^2\) = 0.91, p < 0.01, n = 23), which was consistent with results reported by Chow et al. (2004).

The measurement of elemental emission from forest burning was very limited. Thus compositions of element in Fig. 7 were only available in three regions (i.e., Southeast Asia, Europe and South America). The results showed that Ca was the most abundant species in the Southeast.
Asia and Europe. Furthermore, fractions of Fe and Zn in Southeast Asia were higher than those in Europe. In the South America, Fe accounted for almost 58% of the total elements (Ca, Mg and Pb was not detected), which was consistent with fraction of Fe in Southeast Asia.

3.4. PM, OC, and EC emissions in the Southeast Asia

The burned area of Southeast Asian rainforest in 2016 was derived by overlaying the burned area from the MODIS burned area product (MCD45A1) and corresponding land cover data. As shown in Fig. 8, the most significantly extensive forest burning occurred in Myanmar, Cambodia, and Thailand, accounting for almost 95% of the total forest burned in the Southeast Asia. The main reason was that fire-related slash-and-burn agriculture and lands clearing by local farmers were dominant in these areas. Likewise, Shi et al. (2014) analysed biomass burned in the Southeast Asia from 2001 to 2010 using data from three frequent satellites and found that the more extensive burned areas in northern Southeast Asia were in Myanmar, northern Thailand, and eastern Cambodia, and the location of the most significant biomass burn in southern Southeast Asia was Indonesia. Indonesia has the largest peatland reserves in the world, and the extensive biomass burning was attributed to underground peatland burning, rather than to forest burning (Permadi and Nguyen Thi Kim, 2013; Shi et al., 2014; Shi and Yamaguchi, 2014). This may explain why no massive burned areas were detected in Indonesia in this study. As shown in Fig. 8, the annual emissions of PM, OC, and EC in 2016 in the Southeast Asia were 1780, 687, and 86.1 Gg, respectively. The regions with significant emissions included Myanmar, Cambodia, and Thailand. The spatial distribution of pollutant emissions in this study was slightly different from those of the burned areas. For example, the country with the greatest area of burned forest was Myanmar, whereas the most significant pollutant emissions were found in Cambodia. This might be due to the forests in the different countries had different densities (3.3 kg km\(^{-2}\) in Myanmar and 5.7 kg km\(^{-2}\) in Cambodia), which indicated that in addition to burned area, forest density was another important factor influencing pollutant emissions.

The annual bottom-up emissions of PM, OC, and EC in the Southeast Asia have been estimated in several studies (Chang and Song, 2010; Shi and Yamaguchi, 2014). The annual PM emissions in the Southeast Asia estimated in this study was 1780 Gg, which was considerably lower than the emissions reported by Chang and Song (2010)
Furthermore, the OC and EC emissions estimated in this study were within the range of data reported by other studies. The different pollutant emissions were related to several factors when accounting for various algorithms and data sources, such as differences in burned area, EFs, and biomass density. The necessary parameters to estimate pollutant emissions, including research area, research year, EFs, and burned area, are compiled in Table S2. It should be noted that the biomass densities and combustion factors are not shown in Table S2 as these data were not available in the studies we reviewed. Similar to PM emissions, we found that the research area and EFPM in this study were also lower than those used by Chang and Song (2010). Except for the countries we studied, India, Maldives, Sri Lanka, Nepal, Bhutan, and Bangladesh were assessed by Chang and Song (2010). As a result, the burned area in this study (14,657 km²) was smaller than that in Chang and Song (2010) (23,876 km²). Furthermore, the EFPM cited from Chang and Song (2010) was measured in 2001 by Andreae and Merlet (2001), and was three times higher than the values in our study in 2016, leading to the resultant higher annual PM emissions. The EFs of OC and EC used by Chang and Song (2010) were 6.8 and 0.66 g kg⁻¹ fuel, respectively. However, it is unlikely that the total EFs of OC and EC accounted for only 17.2% of the total PM. The annual emissions of PM, OC, and EC in GFEDv4.1 were higher than those reported by Chang and Song (2010), although the research area was similar in those two studies. These differences might be attributed to overestimation of burned area by GFEDv4.1. The MODIS 500-m burned area product is reportedly more accurate than the GFED3 burned area product, with 0.5° × 0.5° spatial resolution (Shi and Yamaguchi, 2014).

The intra-annual burned forest area, PM, OC, and EC emissions in Southeast Asia were shown in Table 3. The most extensive forest fires occurred during the four-month period from January to April, accounting for 99.5% of the total forest fires in 2016, which was consistent with results reported by Giglio et al. (2013) and Fu et al. (2012). Climate and agricultural activities were the two main reasons of temporal variations. January to March is the typical fire season. The monsoon season begins in May and lasts for at least four months, which can bring heavy rain. In addition, clearing land with fire before spring cultivation is an important activity for local farmers. Similar to spatial characteristics, trends of monthly variations in PM, OC and EC emissions differed slightly. For example, the area of burned forest and PM

Fig. 8. Pollutant emissions from burning rainforest in Southeast Asia in 2016.
emissions reached their maximums in March, whereas peak values for OC and EC occurred in January. The different temporal variations of PM, OC, and EC due to the different EFs of the four types of tropical rainforest vegetation examined in this study were another important factor affecting pollutant emissions.

3.5. Emission uncertainties

The uncertainty of satellite data might be resulted from the persistent cloud cover, small fire, and short duration and so on (Benali et al., 2016). In order to be more precise, we assumed that the total uncertainty from MODIS burned-area product was 20% which was reported by Hyer and Reid (2009). The standard deviations of mean values for EFPM, EFOC, and EFEC measured in this study were 66.7%, 89.8%, and 60.8% for forest burning, respectively. The standard deviations of mean values for EFPM, EFOC, and EFEC were 47.2%, 48.1%, and 57.5% for shrub. The standard deviations of mean values for EFPM, EFOC, and EFEC were 64.0%, 77.1%, and 37.6% for herbaceous. In addition, the combustion factor varied significantly by fuel types and moisture content (Goto and Suzuki, 2013). Thus, we assumed that the uncertainty of the combustion factor was about 30% by combining the combustion factors reported by Jain et al. (2006) and in this study. Finally, the estimated emissions (minimum-maximum) ranged from 1527 to 2834 Gg year\(^{-1}\) for PM, 529.9–1100 Gg year\(^{-1}\) for OC, and 81.02–150.4 Gg year\(^{-1}\) for EC.

4. Conclusions

In this study, EFs of PM, OC, EC, WSIs, and elements from rainforest burning in Southeast Asia were measured. Simultaneously, average EF\(_{PM}\) for different biomass types, sampling regions and burning methods were compared. The average EF\(_{PM}\) was the highest in Southeast Asia, followed by North America, South America and Africa, which was mainly attributed to the diversity of wood species, discrepancy of ambient temperature and humidity. The findings from this study indicate that using of EF\(_{PM}\) from other regions would lead to overestimations of PM emissions in Southeast Asia.

OM was the dominant constituent of PM from the biomass burning emissions in the Southeast Asia, accounting for 56.9 ± 18.0% of the total PM mass. K\(^+\), Cl\(^-\), and SO2-4 were the most abundant ions in most of the samples. Notably, K\(^+\) emitted from biomass burning exibited the most significant positive correlations (\(p < 0.01\)) with PO3-4, F\(^-\), Cl\(^-\), Br\(^-\), and SO2-4, consistent with levels reported in other studies. K, Ca, Fe, Na, and Mg were the most abundant elements in almost all emissions from the burned forest vegetation. Furthermore, source profiles of WSIs and elements varied largely in different areas.

The PM, OC, and EC emissions from rainforest burning in Southeast Asia in 2016 were estimated. PM emissions reached to its maximum in March, whereas OC and EC emissions levels peaked in January.
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