TITLE:

Quantitative assessment of source contributions to PM_{2.5} on the west coast of Peninsular Malaysia to determine the burden of Indonesian peatland fire

AUTHOR(S):

Fujii, Yusuke; Tohno, Susumu; Amil, Norhaniza; Latif, Mohd Talib

CITATION:

Fujii, Yusuke ...[et al]. Quantitative assessment of source contributions to PM_{2.5} on the west coast of Peninsular Malaysia to determine the burden of Indonesian peatland fire. Atmospheric Environment 2017, 171: 111-117

ISSUE DATE:

2017-12

URL:

<http://hdl.handle.net/2433/235562>

RIGHT:

© 2017. This manuscript version is made available under the CC-BY-NC-ND 4.0 license <http://creativecommons.org/licenses/by-nc-nd/4.0/>; The full-text file will be made open to the public on 01 December 2019 in accordance with publisher's 'Terms and Conditions for Self-Archiving'; この論文は出版社版ではありません。引用の際には出版社版をご確認ください。; This is not the published version. Please cite only the published version.

Quantitative assessment of source contributions to PM_{2.5} on the west coast of Peninsular Malaysia to determine the burden of Indonesian peatland fire

Yusuke Fujii ^{a, *}, Susumu Tohno ^b, Norhaniza Amil ^{c, d}, and Mohd Talib Latif ^{d, e}

^a *Center for Environmental Science in Saitama, 914 Kamitanadare, Kazo, Saitama 347-0115, Japan.*

^b *Department of Socio-Environmental Energy Science, Graduate School of Energy Science, Kyoto University, Sakyo-ku, Kyoto 606-8501, Japan.*

^c *Environmental Technology Division, School of Industrial Technology, Universiti Sains Malaysia, 11800 USM, Penang, Malaysia.*

^d *School of Environmental and Natural Resource Sciences, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia.*

^e *Institute for Environment and Development (Lestari), Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia.*

* Corresponding author, phone (+81) 480-73-8352; fax (+81) 480-70-2031

Email: fujii.yusuke.am@pref.saitama.lg.jp

Abstract

Almost every dry season, peatland fires occur in Sumatra and Kalimantan Inlands. Dense smoke haze from Indonesian peatland fires (IPFs) causes impacts on health, visibility, transport and regional climate in Southeast Asian countries such as Indonesia, Malaysia, and Singapore. Quantitative knowledge of IPF source contribution to ambient aerosols in Southeast Asia (SEA) is so useful to make appropriate suggestions to policy makers to mitigate IPF-induced haze pollution. However, its quantitative contribution to ambient aerosols in SEA remains unclarified. In this study, the source contributions to $PM_{2.5}$ were determined by the Positive Matrix Factorization (PMF) model with annual comprehensive observation data at Petaling Jaya on the west coast of Peninsular Malaysia, which is downwind of the IPF areas in Sumatra Island, during the dry (southwest monsoon: June–September) season. The average $PM_{2.5}$ mass concentration during the whole sampling periods (Aug 2011–Jul 2012) based on the PMF and chemical mass closure models was determined as $20\text{--}21\ \mu\text{g m}^{-3}$. Throughout the sampling periods, IPF contributed (on average) $6.1\text{--}7.0\ \mu\text{g m}^{-3}$ to the $PM_{2.5}$, or $\sim 30\%$ of the retrieved $PM_{2.5}$ concentration. In particular, the $PM_{2.5}$ was dominantly sourced from IPF during the southwest monsoon season (51–55% of the total $PM_{2.5}$ concentration on average). Thus, reducing the IPF burden in the $PM_{2.5}$ levels would drastically improve the air quality (especially during the southwest monsoon season) around the west coast of Peninsular Malaysia.

Key words

Biomass burning; Peatland fire; PMF; Source apportionment; $PM_{2.5}$; Malaysia

1. Introduction

Peatland is a terrestrial wetland ecosystem in which the organic matter production exceeds its decomposition, resulting in net accumulation (Page et al., 2006). Indonesia has the largest tropical peatland area ($2.7 \times 10^5 \text{ km}^2$) in the world (Joosten, 2010) and almost every dry season, peatland fires occur in Sumatra and Kalimantan Inlands. As peatland fires are usually ignited underground and can travel large distances unseen before reappearing elsewhere, they are extremely difficult to extinguish (Fujii et al., 2015b). For this reason, peatland fires pose a more serious risk of uncontrollable burning than other sources of ignitable biomass.

Dense smoke haze from Indonesian peatland fires (IPFs) causes impacts on health, visibility, transport and regional climate in Southeast Asian countries such as Indonesia, Malaysia, and Singapore (Betha et al., 2014, 2013; Engling et al., 2014; Fujii et al., 2016a, 2015a, 2015b, 2014; Harrison et al., 2009; He et al., 2010; Page et al., 2002; See et al., 2007, 2006; Tacconi, 2003). $\text{PM}_{2.5}$ is the main constituent of the smoke haze that is chiefly responsible for adverse health and environmental effects (See et al., 2006). Chemical characterizations of IPF-induced smoke aerosols including $\text{PM}_{2.5}$ have been conducted to investigate the contribution of IPFs to the air quality in Southeast Asia (SEA) or identify the key indicator of IPF in transboundary pollution countries or near fire sources (Abas et al., 2004a, 2004b; Betha et al., 2013; Fang et al., 1999; Fujii et al., 2016a, 2015a, 2015b, 2014; Huboyo et al., 2016; Okuda et al., 2002; See et al., 2007). Most of these studies were based on intensive field observations and/or the chemical speciation of ordinary species (ions and metals) and organic species derived from IPFs have been seldom focused, despite the high proportion of organic carbon (OC) (~70% of the $\text{PM}_{2.5}$ mass) at the IPF source (Fujii et al., 2014). Based on long-term (annual) field observations, Fujii et al. (2015b)

reported the chemical characteristics of carbonaceous PM_{2.5} in Malaysia for the first time. As some key findings, they demonstrated that IPFs significantly affect many carbonaceous species in PM_{2.5} (e.g., OP (Pyrolysis OC), *p*-hydroxybenzoic acid, and heptacosane), and the OP/OC₄ mass ratio is a useful indicator of transboundary haze pollution from IPFs at receptor sites even in light haze (the ratio during the haze periods were higher (> 4) than during the non-haze periods (< 2)). Note that OP and OC₄ were determined according to IMPROVE_A protocol in their study.

In addition to chemical characterization, quantitative knowledge of IPF source contribution to ambient aerosols in SEA provides valuable information to policy makers to mitigate IPF-induced haze pollution. However, it seems to be extremely difficult to extract IPF source contribution with accuracy, since SEA hosts one of the most complex aerosol systems in the world (Reid et al., 2013), especially with mixtures of a wide range of atmospheric pollutants (e.g., several types of biomass burning including IPF, industry, and mobile sources). See et al. (2007) and Engling et al. (2014) respectively reported that on hazy days, IPFs significantly contribute to the ambient aerosols in Indonesia (18 and 51% of PM_{2.5} mass) and Singapore (~76% of PM mass). Their analyses were conducted based on intensive field observations by chemical mass balance (CMB) modeling, incorporating typical chemical species such as inorganic ions and metals, and they mostly used source profiles in the US EPA data base (SPECIATE). Thus, the source profiles may not necessarily represent the actual local sources affecting the receptor sites, and the reliability is questionable. Besides, long-term observation data are necessary to determine quantitative source contribution in relation to PM_{2.5} ambient air quality standard. In Malaysia, the sources of ambient aerosols have been apportioned by the Positive Matrix Factorization (PMF) model, which requires no prior knowledge of the source profiles (Amil et al., 2016; Khan et al., 2016a, 2016b,

2015; Rahman et al., 2015). However, these studies applied the datasets of ordinary chemical species, and didn't focus on the IPF source. Thus, the quantitative contribution of IPFs to ambient aerosols in SEA (especially in Malaysia) remains unclarified. An approach based on the source indicators of IPF would effectively and efficiently determine the IPF burden at the receptor sites.

In this study, the source contributions to $PM_{2.5}$ based on the annual observation data at Petaling Jaya on the west coast of Peninsular Malaysia, which is downwind of the IPF areas in Sumatra Island, during the dry (southwest monsoon: June–September) season were determined by PMF modeling. Here, we strongly emphasize the following key point in this study: the highest priority was to extract the IPF source factor and quantitatively determine the contribution of IPF to the annual and seasonal average $PM_{2.5}$ levels based on the source indicator of IPF. Our study reveals the contribution of IPF source to $PM_{2.5}$ concentration throughout a year for the first time in Malaysia and can provide valuable information for $PM_{2.5}$ mitigation strategies in SEA.

2. Material and methods

2.1. Sampling location

The sampling location and methods have been detailed in our former reports (Amil et al., 2016; Fujii et al., 2015b). Briefly, the sampling was performed on the roof of the Malaysian Meteorological Department's main building (eight stories) located at Petaling Jaya in Malaysia ($3^{\circ} 06' 09''$ N, $101^{\circ} 38' 41''$ E) from August 2011 to July 2012. Classified as an urban-industrial area, Petaling Jaya is located approximately 10 km from Malaysia's capital (Kuala Lumpur), and is predominantly residential and industrial with high-density road traffic.

2.2. Sample collection and analysis

PM_{2.5} samples were collected by a Tisch high-volume air sampler (model: TE-3070V-2.5-BL) on a pre-heated quartz fiber filter (Whatman, 500 °C for 3 h) for 24 h at a flow rate of 1.13 m³ min⁻¹. The OC, elemental carbon (EC), water-soluble ions, trace metals, and solvent-extractable organic compounds in the samples were quantified. The carbonaceous content was quantified using a DRI model 2001 OC/EC carbon analyzer, which employs the thermal optical-reflectance method under the IMPROVE_A protocol. The detailed information of OC and EC is provided in our previous report (Fujii et al., 2015b). The target water-soluble ions (C₂O₄²⁻, Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, NH₄⁺, K⁺, and Ca²⁺) were analyzed in aliquots taken from the quartz fiber filters. Each filter was extracted by ultrasonic agitation for 20 min using 5 mL deionized water. The extract was filtered through a PTFE syringe filter (pore size 0.45 μm) and analyzed by ion chromatography (an ICS-2000 (Dionex) for C₂O₄²⁻; an HIC-10A (Shimadzu) for other ions). Metals were analyzed by inductively coupled plasma mass spectrometry (Elan 9000, PerkinElmer). Seven metals (Al, Fe, Pb, Zn, Cu, Ni, and V) were determined in this study. Detailed information of these metals is provided elsewhere (Amil et al., 2016).

The solvent-extractable organic compounds were determined by the gas chromatography/mass spectrometry technique. The target compounds were levoglucosan, mannosan, galactosan, *p*-hydroxybenzoic acid, vanillic acid, syringic acid, and *n*-alkanes (C₂₂ (docosane)–C₃₃ (tritriacontane)). Detailed information of these compounds is provided in our previous report (Fujii et al., 2015b).

2.3. Source apportionment

The PM_{2.5} sources at the fixed receptor site were identified and characterized by the source apportionment tool PMF5.0 (Norris et al., 2014). PMF is a factor analysis model that solves the chemical mass balance equations by a weighted least-squares algorithm and with imposing non-negativity constraints on the factors (Hasheminassab et al., 2014; Reff et al., 2007). The PMF model is described in detail elsewhere (Norris et al., 2014). The main research objective of the present study is extracting the IPF source factor.

The PMF calculation requires the sample species concentrations and their associated uncertainties as inputs. In this study, the chemical species in the PMF calculation were OC–OP (= OC1 + OC2 + OC3 + OC4), OP, C₂O₄²⁻, Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, NH₄⁺, K⁺, Ca²⁺, levoglucosan, mannosan, galactosan, *p*-hydroxybenzoic acid, vanillic acid, syringic acid, *n*-alkanes (C₂₂ (docosane)–C₃₃ (tritriacontane)), Al, Fe, Pb, Zn, Cu, Ni, and V. Referring to previous studies, the indicators of the IPF source factor were selected as OP, *p*-hydroxybenzoic acid, and C₂₅–C₃₃ (Fujii et al., 2016a, 2015a, 2015b). Two input data sets were constructed as follows. First, the concentrations of species below the detection limit (DL: Limit of Detection in this study) were set to DL/2. For data points at or below the DL, the uncertainty (σ) was set to 5/6 DL; above the DL, the uncertainty was calculated as $((\text{percentage uncertainty} \times \text{concentration})^2 + \text{DL}^2)^{1/2}$. The percentage uncertainty is sourced from the analytical uncertainty introduced through chemical analysis methods. The errors associated with the modeling assumptions, such as variations in the source profiles and chemical transformations in the atmosphere, were incorporated by adding a modeling uncertainty of 5% to each species (Yli-Tuomi et al., 2015).

To categorize the species, all variables were analyzed under the signal-to-noise (S/N) criterion,

in which variables with $S/N > 2$ and $0.2 < S/N < 2$ were categorized as strong and weak, respectively. The weak variables were then down-weighted by tripling the user-provided uncertainty. Although previous studies regarded variables with $S/N < 0.2$ as bad variables and excluded them from the PMF calculation (Achad et al., 2014; Richard et al., 2011), there were no bad variables in this study. Instead, vanillic acid and syringic acid were categorized as weak variables, and the others were categorized as strong variables.

Following Xie et al. (2012), the factor number (p) was determined by two criteria: the interpretability of the resultant PMF factor profiles and the success rate of factor matching in the bootstrap runs. One-hundred replicate data sets were generated from the original data set by resampling blocks of samples (where the block size was chosen algorithmically) with replacement using a stationary block bootstrap technique (Xie et al., 2012). Each dataset was independently analyzed by PMF calculations. The minimum correlation value, seed set, and block size in the bootstrapping procedure were set to 0.6, 20, and 5, respectively.

3. Results and discussion

3.1. Exploration of PMF solution

The optimal solution of the PMF model was determined by varying the number of factors from three to six. The simulation statistics of all data sets in the PMF calculations are summarized in Table 1. The three- and four-factor solutions yielded factor matching rates above 80%, while the five- and six-factor solutions showed low matching rates. Besides a high factor-matching rates ($> 80\%$), the four-factor solution attained the most physically interpretable results. The three-factor

solution resulted in a mixing of more different sources in one factor compared to the four-factor solution. Thus, we regarded the four-factor solution as the optimal solution.

3.2. Factor identification

The relative contributions of the chemical species in the extracted four factors are shown in Fig.

1. Factor 1 is heavily loaded with Cl^- , NO_3^- , and Na^+ , suggesting mixed sources of sea salt and nitrate (SS + Nitrate). Factor 2 is dominated by OP, $\text{C}_2\text{O}_4^{2-}$, NH_4^+ , levoglucosan, mannosan, galactosan, *p*-hydroxybenzoic acid, and $\text{C}_{25}\text{--C}_{33}$, implying an IPF source. Factor 3 contains large amounts of $\text{C}_2\text{O}_4^{2-}$, SO_4^{2-} , NH_4^+ , Ni, and V, suggesting a heavy oil combustion source. Factor 4 is heavily loaded with multiple chemical species (e.g., EC, Ca^{2+} , *n*-alkanes, Al, Fe, Pb, Zn, and Cu) and we assigned it to Unknown mixed sources. Based on the report by Kahn et al. (2016) (their sampling site is Bangi, which is located ~30 km southeast away from Petaling Jaya), factor 4 may include the sources such as vehicle related emission, soil dust, and coal combustion sources. To separately extract these sources, it is needed to prepare a dataset of additional individual source indicators.

3.3. Source apportionment of major chemical species

This subsection assigns the major chemical species in $\text{PM}_{2.5}$ (OC, EC, and SO_4^{2-}). The ratios of predicted to measured OC, EC, and SO_4^{2-} concentrations were 1.0 ± 0.17 (average $\pm$ standard deviation), 1.0 ± 0.25 , and 1.0 ± 0.20 , respectively, confirming that the resolved sources effectively account for most of the variation in these chemical species (Fig. 2).

Figure 3 presents the time-series of the source apportionment of OC, EC, and SO_4^{2-} in the

analyzed PM_{2.5}. In addition, the statistical results of the source apportionment of these species for each season are shown in Table 2. During the southwest monsoon season from June to September, IPF contributes dominantly to OC (5.7 $\mu\text{gC m}^{-3}$ of the OC concentration on average, or 56% of the OC mass), followed by Unknown (1.9 $\mu\text{gC m}^{-3}$). During the northeast monsoon season from December to March, the OC concentration is contributed mostly by Unknown (3.0 $\mu\text{gC m}^{-3}$ on average), and secondly by heavy oil combustion (1.1 $\mu\text{gC m}^{-3}$). The OP/OC₄ mass ratio has been suggested as a useful indicator of IPF (Fujii et al., 2016a). The OC concentrations contributed by IPF were well correlated with the OP/OC₄ mass ratios during the sampling periods as shown in Fig. 4 (Pearson correlation coefficient = 0.93, p value < 0.001). This supports the assignment of the PMF factor as the IPF source in the present study. Regarding EC, Unknown is the dominant source of EC in both seasons (southwest monsoon season: 1.1 $\mu\text{gC m}^{-3}$, northeast monsoon season: 1.7 $\mu\text{gC m}^{-3}$). The SO₄²⁻ levels are dominated by heavy oil combustion in both seasons (southwest monsoon season: 2.4 $\mu\text{g m}^{-3}$, northeast monsoon season: 1.4 $\mu\text{g m}^{-3}$).

3.4. Source apportionment of PM_{2.5} mass

To estimate the contributions of the IPF and other sources identified in the PMF calculation to the PM_{2.5} mass, we applied a chemical mass closure model. Six categories were selected in this model: organic matter (OM), EC, NH₄⁺, SO₄²⁻ derived from non-sea salt (nss-SO₄²⁻), K⁺ derived from non-sea salt (nss-K⁺), and sea salt. The concentrations of each category in each source were calculated by the following equations.

$$[\text{Reconstructed PM}_{2.5}] = [\text{PM}_{2.5}]_P + [\text{PM}_{2.5}]_{\text{Others}} \quad (1)$$

$$[\text{PM}_{2.5}]_P = k[\text{OC}]_P + [\text{EC}]_P + [\text{SO}_4^{2-}]_P + [\text{NH}_4^+]_P + [\text{K}^+]_P \quad (2)$$

$$\begin{aligned} [\text{PM}_{2.5}]_{\text{Others}} = & 1.6([\text{OC}]_H + [\text{OC}]_{\text{SN}} + [\text{OC}]_U) + ([\text{EC}]_H + [\text{EC}]_{\text{SN}} + [\text{EC}]_U) \\ & + ([\text{SO}_4^{2-}]_H + ([\text{SO}_4^{2-}]_{\text{SN}} - 0.252[\text{Na}^+]_{\text{SN}}) + [\text{SO}_4^{2-}]_U) \\ & + ([\text{NH}_4^+]_H + [\text{NH}_4^+]_{\text{SN}} + [\text{NH}_4^+]_U) \\ & + ([\text{K}^+]_H + ([\text{K}^+]_{\text{SN}} - 0.037[\text{Na}^+]_{\text{SN}}) + [\text{K}^+]_U) \\ & + (1.47[\text{Na}^+]_{\text{SN}} + [\text{Cl}^-]_{\text{SN}}) \end{aligned} \quad (3)$$

where $[\text{Reconstructed PM}_{2.5}]$ denotes the $\text{PM}_{2.5}$ mass concentration calculated by the mass closure model based on the PMF result. $[X]_i$ is the concentration of X contributed by source i (the subscripts P, H, SN, and U refer to IPF, heavy oil combustion, SS + Nitrate, and Unknown, respectively). Turpin and Lim (2001) concluded that 1.6 ± 0.2 as an OM to OC mass conversion factor was a better estimate for urban areas, whereas 2.2–2.6 for aerosols originating from biomass burning (Yttri et al., 2007). In this study, to determine the amounts of OM in IPF and the other sources, the OCs in IPF and the other sources were multiplied by 2.2–2.6 ($= k$) and 1.6, respectively. As Petaling Jaya is urban-industrial area, we assume that amounts of OM not influenced by IPF source can be calculated by $1.6[\text{OC}]$. In Eq. (3), the terms $([\text{SO}_4^{2-}]_{\text{SN}} - 0.252[\text{Na}^+]_{\text{SN}})$, $([\text{K}^+]_{\text{SN}} - 0.037[\text{Na}^+]_{\text{SN}})$, and $(1.47[\text{Na}^+]_{\text{SN}} + [\text{Cl}^-]_{\text{SN}})$ compute the concentrations of nss-SO_4^{2-} , nss-K^+ in SN, and sea salt, respectively. Chemical mass closure models usually include a crustal matter category. However, the present study excludes the crustal matter because we lack sufficient data on the major components of crustal matter, such as Si and Ti. Thus, exclusion of this category would lead to underestimation of the $\text{PM}_{2.5}$ mass concentration in the present calculation. However, source contribution of crustal matter to $\text{PM}_{2.5}$ should be low, because

aerosols of crustal origin are predominantly in the coarse fraction (Seinfeld and Pandis, 2016). In fact, crustal matter in PM_{2.5} at Bangi, which is close to Petaling Jaya was estimated as ~4.0 % of PM_{2.5} mass (Fujii et al., 2016b). Thus, exclusion of crustal matter would lead to a several % underestimation in the present calculation.

Figure 5 shows a time-series of the PM_{2.5} mass concentration during the study periods, reconstructed by the chemical mass closure model based on the PMF results. The PM_{2.5} was dominantly sourced from IPF during the southwest monsoon season (51–55% of the total PM_{2.5} concentration on average) and large contribution was clearly observed in the high PM_{2.5} mass concentration events. Particularly, PM_{2.5} for 11 September 2011 and 15 June 2012 are considered to be obviously affected by IPF source based on the backward air trajectory data by the Hybrid Single Particle Lagrangian Integrated model (Draxler and Hess, 2014). In contrast, the source contribution of IPF during the northeast monsoon season was negligible. The data of monthly hotspot counts in the Sumatra Island and backward air trajectories shown in our former report (Fujii et al., 2015b) support this conclusion.

From the PMF and chemical mass closure models, the average PM_{2.5} mass concentration was determined as 20–21 $\mu\text{g m}^{-3}$ during the whole sampling periods (Aug 2011–Jul 2012). In Kuala Lumpur (~10 km from the sampling site), the annual average PM_{2.5} mass concentration was 24 $\mu\text{g m}^{-3}$ in 2011 (Rahman et al., 2015), consistent with our result. During the whole sampling periods, IPF contributed (on average) 6.1–7.0 $\mu\text{g m}^{-3}$ to the PM_{2.5} mass concentration, or ~30% of the reconstructed PM_{2.5} concentration.

4. Conclusion

The source contributions to $PM_{2.5}$ based on the annual observation data at Petaling Jaya on the west coast of Peninsular Malaysia, which is downwind of the IPF areas in Sumatra Island, during the dry season were determined by PMF modeling. The following four factors were extracted in this study: (1) SS + Nitrate, (2) IPF, (3) Heavy oil, and (4) Unknown. During the southwest and northeast monsoon season, IPF ($5.7 \mu\text{gC m}^{-3}$) and Unknown ($3.0 \mu\text{gC m}^{-3}$) contributes dominantly to OC, respectively. Regarding EC, Unknown is the dominant source of EC in both seasons (southwest monsoon season: $1.1 \mu\text{gC m}^{-3}$, northeast monsoon season: $1.7 \mu\text{gC m}^{-3}$). The SO_4^{2-} levels are dominated by heavy oil combustion in both seasons (southwest monsoon season: $2.4 \mu\text{g m}^{-3}$, northeast monsoon season: $1.4 \mu\text{g m}^{-3}$). The average $PM_{2.5}$ mass concentration during the whole sampling periods (Aug 2011–Jul 2012) based on the PMF and chemical mass closure models was determined as $20\text{--}21 \mu\text{g m}^{-3}$. Throughout the sampling periods, IPF contributed an estimated $6.1\text{--}7.0 \mu\text{g m}^{-3}$ to the $PM_{2.5}$, or $\sim 30\%$ of the retrieved $PM_{2.5}$ concentration. In particular, the $PM_{2.5}$ was dominantly sourced from IPF during the southwest monsoon season (51–55% of the total $PM_{2.5}$ concentration on average). Thus, reducing the IPF burden in the $PM_{2.5}$ levels would drastically improve the air quality (especially during the southwest monsoon season) around the west coast of Peninsular Malaysia.

Acknowledgment

This study was supported by JSPS Kakenhi (grant numbers: 15H02589 and 15J08153) and UKM University Research Grant (grant number: DIP-2016-105).

References

- Abas, M.R.B., Oros, D.R., Simoneit, B.R.T., 2004a. Biomass burning as the main source of organic aerosol particulate matter in Malaysia during haze episodes. *Chemosphere* 55, 1089–1095. doi:10.1016/j.chemosphere.2004.02.002
- Abas, M.R.B., Rahman, N.A., Omar, N.Y.M.J., Maah, M.J., Samah, A.A., Oros, D.R., Otto, A., Simoneit, B.R.T., 2004b. Organic composition of aerosol particulate matter during a haze episode in Kuala Lumpur, Malaysia. *Atmos. Environ.* 38, 4223–4241. doi:10.1016/j.atmosenv.2004.01.048
- Achad, M., López, M.L., Ceppi, S., Palancar, G.G., Tirao, G., Toselli, B.M., 2014. Assessment of fine and sub-micrometer aerosols at an urban environment of Argentina. *Atmos. Environ.* 92, 522–532. doi:10.1016/j.atmosenv.2014.05.001
- Amil, N., Latif, M.T., Khan, M.F., Mohamad, M., 2016. Seasonal variability of PM_{2.5} composition and sources in the Klang Valley urban-industrial environment. *Atmos. Chem. Phys.* 16, 5357–5381. doi:10.5194/acp-16-5357-2016
- Betha, R., Behera, S.N., Balasubramanian, R., 2014. 2013 Southeast Asian smoke haze: Fractionation of particulate-bound elements and associated health risk. *Environ. Sci. Technol.* 48, 4327–4335. doi:10.1021/es405533d
- Betha, R., Pradani, M., Lestari, P., Joshi, U.M., Reid, J.S., Balasubramanian, R., 2013. Chemical speciation of trace metals emitted from Indonesian peat fires for health risk assessment. *Atmos. Res.* 122, 571–578. doi:10.1016/j.atmosres.2012.05.024
- Draxler, R.R., Hess, G.D., 2014. Description of the HYSPLIT_4 modeling system. NOAA Technical Memorandum ERL ARL–224.

- Engling, G., He, J., Betha, R., Balasubramanian, R., 2014. Assessing the regional impact of Indonesian biomass burning emissions based on organic molecular tracers and chemical mass balance modeling. *Atmos. Chem. Phys.* 14, 8043–8054.
doi:10.5194/acp-14-8043-2014
- Fang, M., Zheng, M., Wang, F., To, K.L., Jaafar, A.B., Tong, S.L., 1999. The solvent-extractable organic compounds in the Indonesia biomass burning aerosols-characterization studies. *Atmos. Environ.* 33, 783–795. doi:atmos-chem-phys.net/14/8043/2014/acp-14-8043-2014
- Fujii, Y., Iriana, W., Oda, M., Puriwigati, A., Tohno, S., Lestari, P., Mizohata, A., Huboyo, H.S., 2015a. Characteristics of carbonaceous aerosols emitted from peatland fire in Riau, Sumatra, Indonesia (2): Identification of organic compounds. *Atmos. Environ.* 110, 1–7.
doi:10.1016/j.atmosenv.2015.03.042
- Fujii, Y., Iriana, W., Oda, M., Puriwigati, A., Tohno, S., Lestari, P., Mizohata, A., Huboyo, H.S., 2014. Characteristics of carbonaceous aerosols emitted from peatland fire in Riau, Sumatra, Indonesia. *Atmos. Environ.* 87, 164–169. doi:10.1016/j.atmosenv.2014.01.037
- Fujii, Y., Mahmud, M., Oda, M., Tohno, S., Matsumoto, J., Mizohata, A., 2016a. A key indicator of transboundary particulate matter pollution derived from Indonesian peatland fires in Malaysia. *Aerosol Air Qual. Res.* 16, 69–78. doi:10.4209/aaqr.2015.04.0215
- Fujii, Y., Mastura, M., Tohno, S., Tomoaki, O., Akira, M., 2016b. A Case Study of PM_{2.5} Characterization in Bangi, Selangor, Malaysia during the Southwest Monsoon Season. *Aerosol Air Qual. Res.* 16, 2685–2691. doi:10.4209/aaqr.2015.04.0277
- Fujii, Y., Tohno, S., Amil, N., Latif, M.T., Oda, M., Matsumoto, J., Mizohata, A., 2015b. Annual variations of carbonaceous PM_{2.5} in Malaysia: influence by Indonesian peatland fires. *Atmos.*

- Chem. Phys. 15, 13319–13329. doi:10.5194/acpd-15-22419-2015
- Harrison, M.E., Page, S.E., Limin, S.H., 2009. The global impact of Indonesian forest fires. *Biologist* 56, 156–163.
- Hasheminassab, S., Daher, N., Saffari, A., Wang, D., Ostro, B.D., Sioutas, C., 2014. Spatial and temporal variability of sources of ambient fine particulate matter (PM_{2.5}) in California. *Atmos. Chem. Phys.* 14, 12085–12097. doi:10.5194/acp-14-12085-2014
- He, J., Zielinska, B., Balasubramanian, R., 2010. Composition of semi-volatile organic compounds in the urban atmosphere of Singapore: influence of biomass burning. *Atmos. Chem. Phys.* 10, 11401–11413. doi:10.5194/acp-10-11401-2010
- Huboyo, H.S., Syafrudin, Fujii, Y., Tohno, S., 2016. Comparison of PM₁₀ pattern and PM_{2.5} carbonaceous fraction from episodic and non episodic period of peat land wildfire. *ARN J. Eng. Appl. Sci.* 11, 4793–4796.
- Joosten, H., 2010. The Global Peatland CO₂ Picture Peatland status and drainage related emissions in all countries of the world. Wetlands International, Ede, the Netherlands, p. 7.
- Khan, M.F., Latif, M.T., Lim, C.H., Amil, N., Jaafar, S.A., Dominick, D., Mohd Nadzir, M.S., Sahani, M., Tahir, N.M., 2015. Seasonal effect and source apportionment of polycyclic aromatic hydrocarbons in PM_{2.5}. *Atmos. Environ.* 106, 178–190.
doi:10.1016/j.atmosenv.2015.01.077
- Khan, M.F., Latif, M.T., Saw, W.H., Amil, N., Nadzir, M.S.M., Sahani, M., Tahir, N.M., Chung, J.X., 2016a. Fine particulate matter in the tropical environment : monsoonal effects , source apportionment , and health risk assessment. *Atmos. Chem. Phys.* 16, 597–617.
doi:10.5194/acp-16-597-2016

- Khan, M.F., Sulong, N.A., Latif, M.T., Nadzir, M.S.M., Amil, N., Hussain, D.F.M., Lee, V., Hosaini, P.N., Shaharom, S., Yusoff, N.A.Y.M., Hoque, H.M.S., Chung, J.X., Sahani, M., Mohd Tahir, N., Juneng, L., Maulud, K.N.A., Abdullah, S.M.S., Fujii, Y., Tohno, S., Mizohata, A., 2016b. Comprehensive assessment of PM_{2.5} physicochemical properties during the Southeast Asia dry season (southwest monsoon). *J. Geophys. Res. Atmos.* 5, 1–23. doi:10.1002/2016JD025894
- Norris, G., Duvall, R., Brown, S., Bai, S., 2014. EPA Positive Matrix Factorization (PMF) 5.0 fundamentals and user guide, US Environmental Protection Agency. Washington, DC. National Research Laboratory, Research Triangle Park, USA.
- Okuda, T., Kumata, H., Zakaria, M.P., Naraoka, H., Ishiwatari, R., Takada, H., 2002. Source identification of Malaysian atmospheric polycyclic aromatic hydrocarbons nearby forest fires using molecular and isotopic compositions. *Atmos. Environ.* 36, 611–618. doi:10.1016/S1352-2310(01)00506-4
- Page, S., Rieley, J., Wüst, R., 2006. Lowland tropical peatlands of Southeast Asia, in: Martini, I.P., Martinez Cortizas, A., Chesworth, W. (Ed.), *Developments in Earth Surface Processes, Peatlands: Evolution and Records of Environmental and Climate Changes*. Elsevier, Amsterdam, the Netherlands, pp. 145–172.
- Page, S.E., Siegert, F., Rieley, J.O., Boehm, H.-D. V., Jaya, A., Limin, S., 2002. The amount of carbon released from peat and forest fires in Indonesia during 1997. *Nature* 420, 61–65. doi:10.1038/nature01131
- Rahman, S.A., Hamzah, M.S., Elias, M.S., Salim, N.A.A., Hashim, A., Shukor, S., Siong, W.B., Wood, A.K., 2015. A long term study on characterization and source apportionment of

particulate pollution in Klang Valley, Kuala Lumpur. *Aerosol Air Qual. Res.* 15, 2291–2304.

doi:10.4209/aaqr.2015.03.0188

Reff, A., Eberly, S.I., Bhawe, P. V, 2007. Receptor modeling of ambient particulate matter data using positive matrix factorization: review of existing methods. *J. Air Waste Manage. Assoc.* 57, 146–154. doi:10.1080/10473289.2007.10465319

Reid, J.S., Hyer, E.J., Johnson, R.S., Holben, B.N., Yokelson, R.J., Zhang, J., Campbell, J.R., Christopher, S.A., Di Girolamo, L., Giglio, L., Holz, R.E., Kearney, C., Miettinen, J., Reid, E.A., Turk, F.J., Wang, J., Xian, P., Zhao, G., Balasubramanian, R., Chew, B.N., Janjai, S., Lagrosas, N., Lestari, P., Lin, N.-H., Mahmud, M., Nguyen, A.X., Norris, B., Oanh, N.T.K., Oo, M., Salinas, S. V., Welton, E.J., Liew, S.C., 2013. Observing and understanding the Southeast Asian aerosol system by remote sensing: An initial review and analysis for the Seven Southeast Asian Studies (7SEAS) program. *Atmos. Res.* 122, 403–468. doi:10.1016/j.atmosres.2012.06.005

Richard, A., Gianini, M.F.D., Mohr, C., Furger, M., Bukowiecki, N., Minguillón, M.C., Lienemann, P., Flechsig, U., Appel, K., Decarlo, P.F., Heringa, M.F., Chirico, R., Baltensperger, U., Prévôt, A.S.H., 2011. Source apportionment of size and time resolved trace elements and organic aerosols from an urban courtyard site in Switzerland. *Atmos. Chem. Phys.* 11, 8945–8963. doi:10.5194/acp-11-8945-2011

See, S.W., Balasubramanian, R., Rianawati, E., Karthikeyan, S., Streets, D.G., 2007. Characterization and source apportionment of particulate matter $\leq 2.5 \mu\text{m}$ in Sumatra, Indonesia, during a recent peat fire episode. *Environ. Sci. Technol.* 41, 3488–3494. doi:10.1021/es061943k

- See, S.W., Balasubramanian, R., Wang, W., 2006. A study of the physical, chemical, and optical properties of ambient aerosol particles in Southeast Asia during hazy and nonhazy days. *J. Geophys. Res.* 111, 1–12. doi:10.1029/2005JD006180
- Seinfeld, J.H., Pandis, S.N., 2016. Atmospheric chemistry and physics: from air pollution to climate change, 3rd ed. John Wiley & Sons, Hoboken, New Jersey.
- Tacconi, L., 2003. Fires in Indonesia: causes, costs and policy implications. *CIFOR Occas. Pap.* 38.
- Turpin, B.J., Lim, H.-J., 2001. Species contributions to PM_{2.5} mass concentrations: revisiting common assumptions for estimating organic mass. *Aerosol Sci. Technol.* 35, 602–610. doi:10.1080/02786820119445
- Xie, M., Hannigan, M., Dutton, S., 2012. Positive matrix factorization of PM_{2.5}: comparison and implications of using different speciation data sets. *Environ. Sci. Technol.* 46, 11962–11970. doi:10.1021/es302358g
- Yli-Tuomi, T., Siponen, T., Taimisto, R.P., Aurela, M., Teinilä, K., Hillamo, R., Pekkanen, J., Salonen, R.O., Lanki, T., 2015. Impact of Wood Combustion for Secondary Heating and Recreational Purposes on Particulate Air Pollution in a Suburb in Finland. *Environ. Sci. Technol.* 49, 4089–4096. doi:10.1021/es5053683
- Yttri, K.E., Aas, W., Bjerke, A., Cape, J.N., Cavalli, F., Ceburnis, D., Dye, C., Emblico, L., Facchini, M.C., Forster, C., Hanssen, J.E., Hansson, H.C., Jennings, S.G., Maenhaut, W., Putaud, J.P., Commission, E., Estate, B., 2007. Elemental and organic carbon in PM₁₀: a one year measurement campaign within the European Monitoring and Evaluation Programme EMEP. *Atmos. Chem. Phys.* 7, 5711–5725. doi:10.5194/acp-7-5711-2007

Table Captions

Table 1. Simulation statistics of all data sets in the PMF model.

Table 2. Source contributions of OC, EC, and SO_4^{2-} in $\text{PM}_{2.5}$ for each season (average $\pm$ standard deviation).

No. of factor	3	4	5	6
No. of samples	81	81	81	81
No. of species	41	41	41	41
No. of bootstrap replicate datasets	100	100	100	100
No. of data sets for which PMF did not converge to a solution	0	0	0	0
No. of data sets for unmatched factors*	1	16	48	34

*No. of bootstrapped cases in which the bootstrapped factors were poorly-reproducible and not uniquely matched to the base case factor.

Table 1.

		IPF ^c	Heavy oil	SS + Nitrate ^d	Unknown
OC [μgC m ⁻³]	SW ^a monsoon (Jun–Sep)	5.7 ± 6.9	1.8 ± 1.3	0.67 ± 0.63	1.9 ± 1.3
	Post-monsoon (Oct–Nov)	0.80 ± 1.2	1.1 ± 0.77	1.1 ± 0.62	2.2 ± 0.98
	NE ^b monsoon (Dec–Mar)	0.36 ± 0.38	1.1 ± 0.76	0.85 ± 0.43	3.0 ± 0.84
	Pre-monsoon (Apr–May)	0.35 ± 0.48	1.3 ± 1.4	1.3 ± 0.39	1.6 ± 0.84
EC [μgC m ⁻³]	SW monsoon (Jun–Sep)	0.47 ± 0.57	0.83 ± 0.59	0.61 ± 0.57	1.1 ± 0.73
	Post-monsoon (Oct–Nov)	0.066 ± 0.095	0.51 ± 0.36	0.96 ± 0.56	1.3 ± 0.55
	NE monsoon (Dec–Mar)	0.030 ± 0.031	0.50 ± 0.35	0.76 ± 0.39	1.7 ± 0.47
	Pre-monsoon (Apr–May)	0.029 ± 0.040	0.62 ± 0.65	1.2 ± 0.36	0.92 ± 0.48
SO ₄ ²⁻ [μg m ⁻³]	SW monsoon (Jun–Sep)	1.3 ± 1.5	2.4 ± 1.7	0.49 ± 0.46	0.14 ± 0.090
	Post-monsoon (Oct–Nov)	0.18 ± 0.26	1.5 ± 1.0	0.77 ± 0.45	0.16 ± 0.069
	NE monsoon (Dec–Mar)	0.081 ± 0.085	1.4 ± 1.0	0.62 ± 0.32	0.21 ± 0.059
	Pre-monsoon (Apr–May)	0.079 ± 0.11	1.8 ± 1.9	0.94 ± 0.29	0.11 ± 0.059

^aSW = southwest. ^bNE = northeast. ^cIPF = Indonesian peatland fire. ^dSS + Nitrate = mixed sources of sea salt and nitrate.

Table 2.

Figure Captions

Fig. 1. Relative contribution of each chemical species in the extracted PMF factors. LG = levoglucosan, MN = mannosan, GL = galactosan, pHBA = *p*-hydroxybenzoic acid, VA = vanillic acid, SA = syringic acid.

Fig. 2. Comparison between the predicted and measured PM_{2.5} concentrations of OC, EC, and SO₄²⁻.

Fig. 3. Time series of source apportionment of OC, EC, and SO₄²⁻ in PM_{2.5} during the sampling periods.

Fig. 4. Relationship between OC contributed by Indonesian peatland fire (IPF) and OP/OC4 mass ratios.

Fig. 5. Time series of source apportionment of PM_{2.5} during the sampling periods. Error bars represent estimates of uncertainty ranges, which derive from an uncertainty of OM to OC conversion factor for Indonesian peatland fire (IPF) source.

Fig. 6. Backward air trajectories during the sampling periods. The 3-days backward air trajectories every 3 hours with 500 m above ground level arriving at the sampling site in local time were calculated in (a) 12 September 2011 and (b) 15 June 2012 by the Hybrid Single Particle Lagrangian Integrated model (Draxler and Hess, 2004) based on meteorological data obtained from the Global Data Assimilation.

Fig. 1.

Fig. 2.

Fig. 3.

Fig. 4.

Fig. 6