



# Radiative response of biomass-burning aerosols over an urban atmosphere in northern peninsular Southeast Asia

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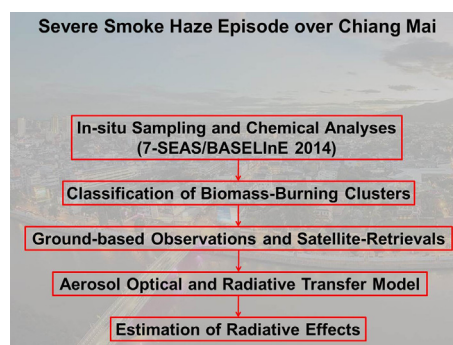
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## HIGHLIGHTS

- Impacts of BB on aerosol properties were investigated over Chiang Mai's urban atmosphere during 7-SEAS/BASE-LLnE 2014.
- Detailed radiation budget over BB sway urban site was quantified for the first time using in-situ datasets.
- Atmospheric heating rate was estimated as high as  $3.6 \text{ K d}^{-1}$ .
- Large surface cooling and atmosphere warming was due to enhanced atmospheric absorption.
- Severe haze episode linked to BB in northern PSEA can cause severe health impacts and modify the regional climate.

## GRAPHICAL ABSTRACT



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## ABSTRACT

A large concentration of finer particulate matter ( $\text{PM}_{2.5}$ ), the primary air-quality concern in northern peninsular Southeast Asia (PSEA), is believed to be closely related to large amounts of biomass burning (BB) particularly in the dry season. In order to quantitatively estimate the contributions of BB to aerosol radiative effects, we thoroughly investigated the physical, chemical, and optical properties of BB aerosols through the integration of ground-based measurements, satellite retrievals, and modelling tools during the Seven South East Asian Studies/Biomass-burning Aerosols & Stratocumulus Environment: Lifecycles & Interactions Experiment (7-SEAS/BASELLnE) campaign in 2014. Clusters were made on the basis of measured BB tracers (Levoglucosan,  $\text{nss-K}^+$ , and  $\text{NO}_3^-$ ) to classify the degree of influence from BB over an urban atmosphere, viz., Chiang Mai ( $18.795^\circ\text{N}$ ,  $98.957^\circ\text{E}$ , 354 m.s.l.), Thailand in northern PSEA. Cluster-wise contributions of BB to  $\text{PM}_{2.5}$ , organic carbon, and elemental carbon were found to be 54–79%, 42–79%, and 39–77%, respectively. Moreover, the cluster-wise aerosol optical index (aerosol optical depth at 500 nm  $\approx 0.98$ –2.45), absorption (single scattering albedo  $\approx 0.87$ –0.85; absorption aerosol optical depth  $\approx 0.15$ –0.38 at 440 nm; absorption Ångström exponent  $\approx 1.43$ –1.57), and radiative impacts (atmospheric heating rate  $\approx 1.4$ –3.6  $\text{K d}^{-1}$ ) displayed consistency with the degree of BB.  $\text{PM}_{2.5}$  during Extreme BB (EBB) was  $\approx 4$  times higher than during Low BB (LBB), whereas this factor was  $\approx 2.5$  for the magnitude of radiative effects. Severe haze (visibility  $\approx 4$  km) due to substantial BB loadings (BB to  $\text{PM}_{2.5} \approx 79\%$ ) with favorable meteorology can significantly impact the local-to-regional air quality and the,

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daily life of local inhabitants as well as become a respiratory health threat. Additionally, such enhancements in atmospheric heating could potentially influence the regional hydrological cycle and crop productivity over Chiang Mai in northern PSEA.

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## 1. Introduction

Particulate matter (PM) of either, natural and anthropogenic origin, is a significant worldwide environmental issue and well known to cause harmful effects on human health (Harrison and Yin, 2000; Metzger et al., 2004) and visibility (Tao et al., 2009). It also plays a crucial role in the local and regional air quality and represents a critical factor in solar radiation (Crutzen and Andreae, 1990; Andreae, 1993) and climate change (Novakov and Penner, 1993; Kanakidou et al., 2005). Many countries worldwide have adopted air-quality standards for ambient PM<sub>10</sub> (cut sizes  $\leq 10\ \mu\text{m}$ ) and PM<sub>2.5</sub> (cut sizes  $\leq 2.5\ \mu\text{m}$ ) and started monitoring projects to identify and quantify the sources in order to reduce their emissions.

Biomass burning (BB) is a widespread routine practice for land conversion and land clearing by deforestation and the burning of secondary forests and pastures (Tsay et al., 2013) in many regions. BB emits substantial amounts of trace gases and PM into the atmosphere (Jian and Fu, 2014), and its contribution to the carbonaceous aerosol at urban sites (Fine et al., 2001; Lanz et al., 2008) has been identified to be significant. Carbonaceous aerosol mainly consists of light-absorbing organic carbon (OC) and elemental carbon (EC) and has recently been of great concern because of its significant impact on regional-to-global climate (Cao et al., 2004; Kanakidou et al., 2009). The magnitude of the scattering and absorption of BB-derived carbonaceous aerosols, responsible for both cooling and warming effects on the climate primarily depends upon the fuel type, combustion phase, environmental conditions, and atmospheric aging. The radiative effects of these particles have remained poorly quantified due to their diverse optical and cloud-activating properties (Vakkari et al., 2014). Boucher et al. (2013) recently reported the radiative forcing of EC and organic aerosols associated with BB as  $+0.0$  ( $-0.2$  to  $+0.2$ )  $\text{W m}^{-2}$ .

Over peninsular Southeast Asia (PSEA, here defined as Vietnam, Cambodia, Thailand, Laos, and Myanmar), large BB occurs annually in the dry season (February–April) due to slash-and-burn and land-clearing practices before the local growing season (Fox et al., 2009; Tsay et al., 2013; Gautam et al., 2013; Jian and Fu, 2014). This BB builds the regional haze generally known as “Asian Brown Cloud” (Ramanathan and Crutzen, 2003). Recently the 7-SEAS/BASELInE (Seven South East Asian Studies/Biomass-burning Aerosols & Stratocumulus Environment: Lifecycles & Interactions Experiment; Lin et al., 2014) campaign was conducted over northern PSEA during the dry seasons from 2013 till 2015 to explore numerous key atmospheric processes and the surface/atmosphere radiation budget associated with the regional BB (Lin et al., 2013; Tsay et al., 2016). Within this framework, some studies have emphasized the BB emissions from northern PSEA and explained their impact on the atmospheric composition, regional air quality, aerosol optics, and regional climate (e.g., Lin et al., 2013, 2014; Tsay et al., 2013, 2016). Prior to this campaign, Gautam et al. (2013) reported the aerosol characterization and satellite-based aerosol radiative impact over northern PSEA during the dry seasons of 2008 and 2009. The enhancement of free tropospheric warming by the transported BB plumes over northern South China Sea from northern PSEA was quantified during the 7-SEAS/Dongsha Experiment (Pani et al., 2016a). Wang et al. (2015) investigated the distribution of aerosol optical properties over northern PSEA during 7-SEAS/BASELInE 2014 and reported their distinct variability over different locations based on the meteorological conditions, fuel type, site elevation, and proximity to BB sources. Pani et al. (2016b) provided a detailed estimation of BB radiative effects for

near-source BB aerosols but was limited to a mountainous location in northern PSEA during 7-SEAS/BASELInE 2013. However, the BB radiative impact varies greatly between different source regions and even different locations in the same region due to their heterogeneity in mass loadings, the optical properties and their vertical distributions. The aerosol vertical distribution is uneven regionally (Toth et al., 2016) and can contribute to the uncertainty in the radiative-forcing estimations (Choi and Chung, 2014).

The focus site of this study is the city of Chiang Mai, a northern urban center located in Chiang Mai Province ( $17\text{--}21^\circ\text{N}$  and  $98\text{--}100^\circ\text{E}$ ) in northern PSEA. This province is the second largest in northern Thailand and covers approximately  $20,170\ \text{km}^2$  in area with a population of about 1,682,164 inhabitants as of June 2015 (<http://www.ChiangMai.go.th/>); it also attracts over 7 million visitors each year. Owing to its geographical features, this city faces serious air-quality degradation, especially during the dry season. Chiang Mai is typically influenced by medium-traffic vehicular emissions with mixed residential, commercial, and industrial emissions, and anthropogenic activities (Chantara et al., 2012; Tsai et al., 2013; Janta and Chantara, 2017). Moreover, a wide range of BB activities over the region, particularly in the dry season deteriorates the air quality of the city and coincides with the peak of the annual haze episode. This region is also tempered by a low latitude and moderate elevation, which makes the atmospheric boundary layer (ABL) more complex. Wang et al. (2015) reported the presence of widespread smoke haze over the region in 2014 when there was likely a significant contribution from BB to the carbonaceous-aerosol (mainly EC or black carbon [BC]) loading in the atmosphere, but the radiative effects have not yet been fully characterized. Hence, this study seeks to estimate the radiative effects over the urban atmosphere of Chiang Mai by using the data obtained during the 7-SEAS/BASELInE 2014 campaign.

In the present study, we use a synergistic approach to characterize the surface chemistry, optical properties, vertical distributions, and shortwave direct radiative effects of aerosols during the BB-dominated period over Chiang Mai's atmosphere by integrating ground-based measurements, satellite retrievals, and a radiative transfer model. Descriptions of the measurements made and methods used in the current analysis are provided in Sections 2 and 3, respectively. The analyses of the aerosol distribution with regard to the compositions, optical properties, vertical profiles, and radiative impacts are carefully investigated in Section 4; their implications for the regional air quality and climate are discussed in Section 5, and finally, a summary is given in Section 6.

## 2. Measurements and data

### 2.1. Aerosol sampling and chemical analysis

Aerosol sampling was carried out on the rooftop of a four-story building at Chiang Mai University (CMU;  $18.795^\circ\text{N}$ ,  $98.957^\circ\text{E}$ , 354 msl) in Chiang Mai, Thailand. The CMU site is located about 2 km west of the city of Chiang Mai. Details of the sampling procedures, analytical methods, quality control, and chemical analysis can be found in Khamkaew et al. (2016). Briefly, 24-h PM<sub>2.5</sub> samples were collected gravimetrically from 8 March till 7 April 2014 (number of samples,  $N = 31$ ) by using both Teflon (Whatman's, UK,  $2\ \mu\text{m}$ ,  $\emptyset = 46.2\ \text{mm}$ ) and quartz-fiber (Whatman's, UK,  $\emptyset = 47\ \text{mm}$ ) filters in mini-volume air samplers (MiniVol, Airmetrics, USA) at a flow rate of  $5\ \text{L min}^{-1}$ . PM<sub>2.5</sub> samples were analyzed for water soluble inorganic ions (WSIIs;  $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ , and  $\text{SO}_4^{2-}$ ) by ion chromatography

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