



Characterization of Particulate Matter Measured at Remote Forest Site in Relation to Local and Distant Contributing Sources

Nguyen Thi Kim Oanh^{*}, Nguyen Thanh Hang, Tipayarom Aungsiri[†], Thiansathit Worrarat, Tipayarom Danutawat[†]

Environmental Engineering and Management, School of Environment, Resources and Development, Asian Institute of Technology, Pathumthani, Thailand

ABSTRACT

This study characterized the particulate matter (PM) pollution at a forest site in the Khao Yai (KY) National Park of Thailand, 700 m above sea level and 120 km upwind to Bangkok during the dry season. Two dichotomous samplers simultaneously operated on each monitoring day to collect 2 samples (on 2 filter types: quartz and mix cellulose) of 24 h PM_{2.5} and 2 samples of 24 h PM_{10-2.5}. More focus was on the dry and more polluted season with a longer sampling period (43 days: Jan–Feb 2005) and less on wet season (10 days, Jun 2005). Samples were analyzed for mass, water soluble ions, elements, EC and OC (Sunset analyzer). PM_{2.5} levels obtained at KY in the dry period (47 $\mu\text{g m}^{-3}$) were comparable to those in several urban areas in Southeast Asia, but the wet season levels (7 $\mu\text{g m}^{-3}$) were significantly lower. In the dry season, levels of EC associated with PM_{2.5} at KY were remarkably lower than but OC were comparable to those found in urban areas which resulted in considerably low EC/TC ratios (0.08). The major mass groups of PM_{2.5} in the dry season were organic matter of biomass smoke origin (OM-smoke), secondary inorganic aerosol, organic matter of other origins than smoke (OM-others) and crustal. Similar contributors to PM_{2.5} were also found in the wet season but with only a small contribution from biomass burning smoke. Significant contributions from distant sources to PM_{2.5} levels measured at KY were also confirmed by the HYSPLIT backward trajectory analysis. The coarse fraction (PM_{10-2.5}) had major mass groups of OM-others, inorganic particles and crustal that were most likely related to local sources, and some amount of aged sea salt indicating a distant source origin.

Keywords: Fine particle; Composition; EC/TC ratio; Forest site; Thailand.

INTRODUCTION

The levels and nature of air pollution observed at a location are determined by a combination of processes, including the local source emission, atmospheric dispersion capacity, removal processes, physical and chemical transformation of pollutants, and the long range transported pollutants. Particulate matter (PM) air pollution in some urban areas in Asia has been studied rather intensively and the information on the PM levels, although still fragmented, has been made available (Kim Oanh *et al.*, 2006; Hopke *et al.*, 2008; Tao *et al.*, 2012). Studies show that high levels of PM in urban areas of Asian developing countries are mainly caused by local emission sources (Hopke *et al.*, 2008; Hai and Kim Oanh,

2013). Information on the PM pollution at non-urban sites is still largely insufficient. Co *et al.* (2014) reported considerably high PM_{2.5} levels (50 $\mu\text{g m}^{-3}$) measured at a rural mountainous area in Vietnam during the dry season which, nevertheless, were still well below those found in Hanoi (75 $\mu\text{g m}^{-3}$; Hai and Kim Oanh, 2013).

Remote sites, in principle are located far away from major emission sources. As a result, they are expected to be less influenced by local anthropogenic sources while contributions from the long range transport would be more pronounced. Accordingly, the PM pollution observed in the air at remote locations may have different compositions than that found in urban areas (Lim *et al.*, 2003; Bae *et al.*, 2006; Li *et al.*, 2013). Information on PM levels and composition at remote sites, therefore, can provide valuable input to understand important chemical and physical processes in the atmosphere, and to enable assessment of the contribution from the long range transport air pollution. Data on air pollution monitored at remote sites, especially in Southeast Asian (SEA) countries, is not commonly found in literature. This is because monitoring activities conducted at remote locations may be associated with several issues, such as accessibility,

[†] Now at Department of Environmental Science, Faculty of Science, Silpakorn University, Nakhon Pathom, Thailand.

^{*} Corresponding author.
E-mail address: kimoanh@ait.ac.th

complex logistic arrangement, and large requirement for resources.

The Asian regional air pollution research network (AIRPET) conducted monitoring activities and reported high levels in several Asian urban areas (Kim Oanh *et al.*, 2006). To fill up the data gap for remote areas, the monitoring activities were also extended to one remote/mountainous site in each participating country of China, India, Indonesia, Philippines, Thailand, and Vietnam (Kim Oanh *et al.*, 2012). The PM pollution studies at the remote sites of AIRPET aimed to provide information on contributing sources, both local and distant, to the measured pollution levels. Particulate pollution was the monitoring focus because of its important effects on human health (Ostro *et al.*, 1999; Fann *et al.*, 2012; Park *et al.*, 2013) and climate (UNEP-WMO, 2011; Bond *et al.*, 2013).

This paper reports the monitoring results of PM pollution at a remote forest site located in the Khao Yai National Park of Thailand. The mass and composition of coarse ($PM_{10-2.5}$) and fine particles ($PM_{2.5}$) were measured in the dry (Jan–Feb) and wet period (Jun) in 2005. The PM mass and composition, the reconstructed mass, and HYSPLIT air mass backward trajectories were analysed to reveal the major contributing factors.

METHODS

Study Area

Khao Yai is a National Park situated 120 km northeast of Bangkok, in the upwind direction of the Bangkok city during the dry season when the northeast monsoon is prevalent. The total area of Khao Yai is above 2,150 km² covering parts of 4 provinces (Saraburi, Nakhon Ratchasima, Prachinburi, and Nakhonnayok), as seen in Fig. S1, Supplementary Information (SI). A meteorological station named Khao Kheow, located at 1259 m above sea level, has been operated by the Thai Meteorological Department since 1976 (http://www.aws-observation.tmd.go.th/web/climate/climate_past.asp). The park is relatively humid all the year around with the monthly average relative humidity of 79–86%. The range of monthly average minimum temperatures is 21–24°C and that of the monthly average maximum temperature is 29–33°C. The monthly precipitation ranks from 70–80 mm (January, February and March) to above 300 mm (October and November), as shown in Table S1, SI. This rainfall pattern is remarkably different from that observed in other parts of Northern Thailand. For example, the dry season in Bangkok is from November to April when lower monthly precipitation is observed, i.e., 10–70 mm (Table S1, SI).

Since the Khao Yai Park is a preserved forest, it has less human activities that directly contribute to air pollution. Nevertheless, there are a few local sources, including personal vehicles (cars, motorcycles) used by the park management officers. During weekends, visitors are observed in the tourist areas of the park. Occasionally, some trucks carrying goods and tourist buses operate along fixed routes in the park. According to the Authority of Tourism in Thailand (2014), most of the tourist activities occur in the cooler period of year, from October to February. There are fire

events, such as forest fires and prescribed burnings, which happen more frequently during the drier months.

The sampling site was selected to be far away from public tourist areas but at a convenient distance to get electricity supply for the equipment. The site was located on an open hill in Khao Yai (KY) at 700 m above the sea level and at the coordinates of 14°25'48"N and 101°23' 24"E (Fig. S1, SI with an inserted photo of sampling equipment).

Sampling and Chemical Analysis

The detail of sampling and chemical analyses, as well as QA/QC applied in this study were the same as previously presented in Kim Oanh *et al.* (2009). Briefly, every step of monitoring including sampling preparation, sampler calibration, and analytical procedures were strictly done with QA/QC. Two collocated Sierra-Anderson dichotomous samplers (dichot) (US EPA, 1999) produced two samples of fine (24h $PM_{2.5}$) and coarse (24 h $PM_{10-2.5}$) PM fractions simultaneously on each monitoring day. One dichot used quartz fiber filters (prebaked at 550°C for about 5 hours before sampling) while the other used mix cellulose ester filters (MCE). The monitoring focused more on the dry season with daily samples taken from 12 Jan to 26 Feb 2005. For comparison, sampling during the wet season was also conducted over a short period in June (20–30 Jun 2005). Due to electricity failure and filter contamination, 4 pairs of samples in the dry season and 1 pair in the wet season were discarded, hence the total number of valid samples was 39 and 9 pairs (a fine & a coarse PM sample) for the dry and wet period, respectively. Difficulty in the sampling at this remote site prevented from collecting a larger number of PM samples, hence the short sampling period conducted in the wet season may not yet produce representative results for the PM pollution at Khao Yai.

One half of each sampled quartz filter was analyzed for water soluble ions by Ion Chromatography (IC), following the procedure described in Chow and Watson (1999). The other half was analyzed for elemental carbon (EC) and organic carbon (OC) by the thermal optical transmission (TOT) using a Sunset analyzer (NIOSH 5040 method) at the University of Illinois at Urbana-Champaign. A half of MCE filter was extracted in 4% HNO_3 solution for the subsequent elemental analysis by the inductively coupled plasma optical emission spectrometry (ICP-OES) following the US EPA Method IO-3.4 (US EPA, 1999). The ICP-OES (PerkinElmer, Optima 2100 DV) has the instrument detection limits of ≤ 1 ppb for most of the analyzed elements except for Si which has a detection limit of 10 ppb (<http://www.perkinelmer.com>). Because of a high uncertainty associated with Si measurements by the ICP method, the Si content in our samples was estimated using the average ratio (1.95) between the Si levels produced by PIXE (Proton Induced X-ray Emission) and those by ICP-OES for a set of 30 samples collected in Thailand by the AIRPET project. The standard curves used for qualification of the elements and ions, respectively, were constructed using five concentration points, and all had $R^2 > 0.99$.

The blanks (QC samples) included laboratory (lab) blanks, trip blanks and exposure blanks. A description of the QC

samples is given in Textbox S1 (SI) along with other QA/QC details used in this study. These QC samples were handled in the same way as the sample filters and were also analyzed for mass and compositions. The mass of passive exposure blanks for the MCE filters (mass gains) were only a few micrograms, from -1 to $+3$ μg per filter. The range of blank levels of quartz filters for water soluble ions was mostly 0.2 – 0.8 $\mu\text{g cm}^{-2}$ of the filter area, but higher for Cl^- and NO_3^- (1.2 – 1.3 $\mu\text{g cm}^{-2}$) and for Ca^{2+} and NH_4^+ (3 – 4 $\mu\text{g cm}^{-2}$). For elements, the average blank levels (of MCE) were all below 0.05 $\mu\text{g cm}^{-2}$ of the filter area except for Ca which was 0.4 $\mu\text{g cm}^{-2}$. The levels of ionic and elemental species did not vary significantly among types of blanks (lab, trip or passive field exposure blanks). However, for OC and EC levels, significant variations between the blank types were observed. The OC levels of passive field blanks were high (2.1 ± 0.3 $\mu\text{g cm}^{-2}$ for the wet and 1.5 ± 0.2 $\mu\text{g cm}^{-2}$ for dry sampling period) that may be caused by the absorption of volatile organic compounds (VOC) on the quartz filters. The blank levels of EC were about 0.2 ± 0.1 $\mu\text{g cm}^{-2}$ for the dry and zero for the wet sampling period. Note that dichots use filters of a 37 mm diameter hence the blank filter area is about 10.8 cm^2 . To correct the analytical results in a sampling period (dry or wet), we used levels in the passive exposure blank for EC and OC, and the average levels of all blank types for ions and elements obtained for the respective sampling period.

Additionally, PM_{10} concentrations measured (using Beta attenuation) by the Pollution Control Department (PCD) of Thailand in the surrounding towns of the Khao Yai Park were also collected to compare with our monitoring results. The meteorological data for the monitoring periods at the Khao Keaw meteorological station was used to cross check with the temperature and pressure measured at our sampling site. Due to the lower altitude of our sampling site, higher temperature (by about 3 – 5°C) and higher pressure (by about 40 mmHg: our site of 700 mmHg vs. Khao Keaw of 660 mmHg) were recorded. This paper reported the PM concentrations at the reference conditions of 760 mmHg and 25°C . Average daily relative humidity during Jan–Feb sampling period was 83 – 87% which was almost the same for the Jun period, 84 – 88% . Our sampling was conducted only on the days without heavy rain. There was heavy fog on some days in Jan–Feb, and alternating sunny and cloudy days were observed in the June sampling period.

Identification of Contributing Sources

Source identifications were conducted using the measured PM mass and composition which were used to calculate major mass groups (reconstructed mass). The air mass backward trajectories on the monitoring days were analysed to identify the potential long range transport pollution.

Reconstructed Mass

Major mass groups included the crustal [1.16 ($1.9\text{Al} + 2.15\text{Si} + 1.41\text{Ca} + 1.67\text{Ti} + 2.09\text{Fe}$)], soot (EC), organic matter of biomass smoke origin (OM-smoke) and OM of other origins (OM-others), inorganic particles, sea salt, and the trace (non-crustal) elements. The mass groups were

determined following the same approaches presented in Kim Oanh *et al.* (2006) and Arruti *et al.* (2011).

To estimate the OM-smoke group, first the potassium contribution by biomass smoke, K-smoke ($\text{Total K}^+ - 0.6 \times \text{Fe}$), in the samples was estimated (Chan *et al.*, 1997). The OC contributed by biomass smoke was determined using the ratio of OC to K^+ obtained from the measurement results for rice straw field burning smoke in Thailand of 6.6 for both PM fractions (Kim Oanh *et al.*, 2010). The OC-smoke was converted to OM-smoke using a factor of 2.0 which was the average value obtained for fresh fire smoke compiled by Turpin and Lim (2001). The remaining OC in $\text{PM}_{2.5}$ samples of both seasons were converted to OM-others using a factor of 1.8 assuming a significant contribution of secondary organic aerosol (SOA) at this remote site (Bae *et al.*, 2006). A factor of 1.4 was used to convert OC-others in the coarse $\text{PM}_{10-2.5}$ to OM-others in both seasons, assuming that contributions from biomass burning smoke and SOA would be less prominent in these samples while contributions from soil and tree debris may be significant.

The inorganic particles group was estimated as the sum of non-marine SO_4^{2-} , NH_4^+ and NO_3^- . The sea salt group was the sum of 2.54 Na^+ and the marine- SO_4^{2-} (Arruti *et al.*, 2011 and references therein). The marine- SO_4^{2-} was taken equal to 0.25Na^+ (Behera *et al.*, 2013 and references therein).

HYSPLIT: The 5-day backward trajectory of air masses arriving at KY on every sampling day was used to reveal possible contributions from long range transport pollution to PM at the study site. The HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory Model, <http://ready.arl.noaa.gov/HYSPLIT.php>) was run online using the default final analyzed meteorological data from the Global Data Assimilation System (GDAS). The backward trajectory was set to start at the KY site coordinates, at $0:00$ UTC ($7:00$ LST) on each monitoring day, from the height of 500 m above the ground level. The selection of the start height was made based on a preliminary analysis of the trajectories arriving at the site at three different heights (300 , 500 and 1000 m) which showed that the trajectories of these three start heights were quite similar. Therefore, the start height of 500 m was finally selected which was considered to be sufficiently low to represent the wind in the lower atmosphere and sufficiently high to minimize the friction effect from the Earth's surface. The 'model vertical velocity' option, i.e., to use the vertical velocity field that is provided in most meteorological data sources, was selected. Note that the similarity between trajectories at the 3 start heights was also found for the mountainous rural site in Vietnam (1000 m above sea level) as discussed in Co *et al.* (2014), and this may be linked to the elevated locations of the study areas. The backward trajectories were grouped based on their origins and pathways into different patterns and the association between PM pollution (mass and composition) with the trajectory patterns were analyzed.

RESULTS AND DISCUSSION

PM Mass

Fig. 1 presents the daily levels of PM during the Jan–

Feb period (39 valid samples) and Jun period (9 valid samples). In the wet sampling period, the 24h PM levels were low with PM_{2.5} varying from 4.0 to 11 $\mu\text{g m}^{-3}$, and PM₁₀ from 9.0 to 18 $\mu\text{g m}^{-3}$. Significantly higher PM_{2.5} levels were observed in the dry period, i.e., fluctuating from 22 to 78 $\mu\text{g m}^{-3}$ and exceeded the 24 h PM_{2.5} standard of Thailand (50 $\mu\text{g m}^{-3}$) in 14 out of 39 days (36%). The coarse fraction (24 h PM_{10-2.5}) in the dry period was, however, relatively low, varying from 5.0 to 18 $\mu\text{g m}^{-3}$. None of the 24 h PM₁₀ measurements exceeded the 24 h standard of Thailand (120 $\mu\text{g m}^{-3}$).

During Jan and Feb of 2005, forest fires happened quite often in the park with a total record of 11 events (6 in Jan plus 5 in Feb). Most of the fires were small, with a burned area of below 0.4 km², and located about 20 km upwind from the monitoring site. Specifically, during the 39 monitoring days (with valid samples) there were 3 days, highlighted in Fig. 1, with forest fires reported: 30 Jan with a large fire covering about 32 km² and the smoke was seen at the sampling site, 4 Feb (0.04 km², 18 km downwind) and 22 Feb (0.02 km², 8 km downwind). There was no fire event reported during the wet sampling period. Overall, the high frequency of fires (largely of prescribed burning) in Jan–Feb period suggested a potential influence of burning smoke on the measured PM levels. In particular, on 30 Jan when the forest fire smoke was seen around the site, a high level of PM_{2.5} (and OC) was observed (Fig. 1). However, high levels of PM_{2.5} were also observed on other days when no forest fires were reported. Other factors may contribute to the high PM pollution on a particular day, such as the formation of secondary particles and the transport of emission from distant sources. For example, the highest PM_{2.5} level recorded on 18 Jan (78 $\mu\text{g m}^{-3}$) may be linked to the long range transport pollution as discussed in the HYSPLIT trajectory analysis.

There was a remarkable difference in PM_{2.5} levels between the dry and wet sampling periods. The average PM_{2.5} level in the dry period was $47 \pm 10 \mu\text{g m}^{-3}$ (Table 1) which was about 7 times as much as the wet period level of $7.0 \pm 2.2 \mu\text{g m}^{-3}$. As for PM_{10-2.5}, the dry period level was $12 \pm 3.3 \mu\text{g m}^{-3}$, about twice as much as the wet period ($6.0 \pm 2.5 \mu\text{g m}^{-3}$). The seasonal variations of PM at KY were more significant than those found at urban sites in the Bangkok Metropolitan Region (BMR) of Thailand. Kim Oanh *et al.* (2006) reported the average PM_{2.5} measured at 4 sites in BMR during 2002–2004 of 50 $\mu\text{g m}^{-3}$ in the dry season (comparable to KY) and 18 $\mu\text{g m}^{-3}$ in the wet season (significantly higher than KY). Overall, the low PM levels recorded at KY in the wet sampling period may be explained by several factors, including less emission from local sources (less forest fires), more efficient wet removal (more rain), less formation of secondary particles and less long range transport pollution (as discussed in the HYSPLIT trajectory analysis). However, only 9 samples were taken over a short period of June hence the obtained results may not be representative for PM in the wet season at KY.

As compared to the daily PM₁₀ levels measured by PCD of Thailand in the surrounding towns, the levels at KY in both seasons, as expected, were the lowest (Fig. S2, SI).

Note that the hourly PM₁₀ data of PCD stations (by Beta ray attenuation) were averaged to get the 24 h levels (from 8 am to 8 am next day, i.e., the same period as our daily sampling). In fact, the daily levels in two surrounding towns, Nakhon Ratchasima and Saraburi, were fluctuating widely and were higher than those recorded at Pathumthani, a suburb area of Bangkok. The activities (traffic, cement industry) in these towns may contribute to the high PM₁₀ levels. No PM_{2.5} data was available from the PCD monitoring network in these towns for comparison.

The ratio between PM_{2.5} and PM₁₀ at KY, calculated for each sampling day, was high in the dry period (0.79 ± 0.06) but reduced to about 0.50 ± 0.17 in the wet sampling period. In the dry period, PM_{2.5} and PM₁₀ were linearly correlated with the coefficient of determination (R^2) of 0.92 (Table 1 and Fig. S3, SI). The high correlation between PM_{2.5} and PM₁₀ in the dry season was explained by the predominant portion of PM_{2.5} in PM₁₀. During the wet period, the correlation between PM_{2.5} and PM₁₀ was low with R^2 of 0.51. The weak correlation between PM_{2.5} and PM_{10-2.5} (Fig. S3, SI) suggested that the dominant contributing sources/processes (characterized by specific PM_{2.5}/PM₁₀ ratios) to the PM pollution at the site varied between the sampling days. The average PM_{2.5}/PM₁₀ ratio obtained for KY in the dry season was higher than those for 4 BMR sites (average of 0.64) and other Asian cities (Kim Oanh *et al.*, 2006). The dominance of PM_{2.5} in PM₁₀ observed at KY in the dry season in turn suggests high contributions from combustion, secondary particles and long range transport that contribute more to the fine fraction mass and less to the coarse fraction mass. For example, coarse PM may be found higher at urban areas due to dust resuspension and road dust emissions while in a preserved forest area as KY the contribution of these typical sources of dust could be lower.

PM Compositions

In this study EC, OC, 12 ions (F⁻, Cl⁻, Br⁻, NO₃⁻, PO₄³⁻, SO₄²⁻, Li⁺, NH₄⁺, Na⁺, K⁺, Mg²⁺ and Ca²⁺) and 18 elements (Al, Ca, Cd, Co, Cr, Cu, Fe, Li, Mg, Mn, Mo, Ni, Pb, Sr, Ti, V, Zn and Si) were analyzed for each PM_{2.5} and PM_{10-2.5} sample. The ion concentrations in these two size fractions were summed up to get the corresponding levels in PM₁₀ (Table 2). The levels of Li⁺ and Li were consistently found below the detection limit hence were not included in the further data analyses.

EC and OC Composition

The total OC was estimated as OC1 + OC2 + OC3 + OC4 + PyrOC (PyrOC is the pyrolyzed carbon fraction while the numbers 1, 2, etc. denote the corresponding OC peaks) and EC was calculated as EC1 + EC2 + EC3 + EC4 + EC5 – PyrOC, using the EC and OC results (NIOSH 5040 method). In the dry season, majority of OC and EC were present in PM_{2.5}, leaving only small fractions to PM_{10-2.5} (Table 2). The daily OC levels in PM_{2.5} showed significantly higher levels in the dry sampling period as compared to the wet period (Fig. 1), constituting about 22–23% of PM_{2.5} mass (Table 2). On average, PM_{2.5} in the dry season had OC of $11 \pm 3.9 \mu\text{g m}^{-3}$ and EC of $0.9 \pm 0.2 \mu\text{g m}^{-3}$ as compared to

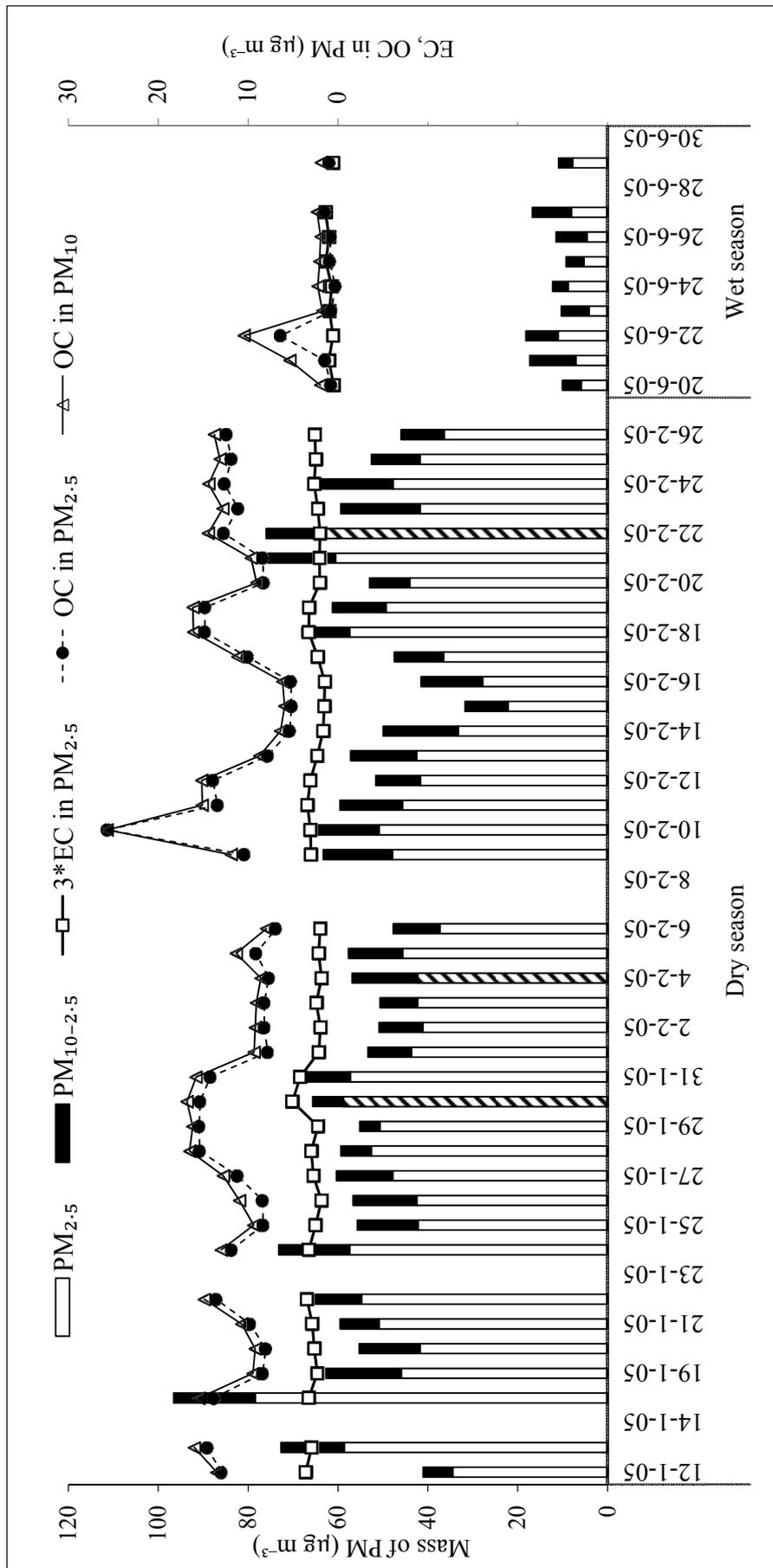


Fig. 1. Levels of PM and carbonaceous compositions during wet and dry season sampling periods at Khao Yai ($3 \times \text{EC}$ in $\text{PM}_{2.5}$ is presented). (Days with forest fire are highlighted with pattern: 30 Jan (32 km^2 burned area and smoke was observed at the site; 4 and 22 Feb with small fires, $0.02\text{--}0.04 \text{ km}^2$ located 18 and 8 km downwind from the site).

Table 1. Summary of PM_{2.5}/PM₁₀ ratio and ion balance in dry and wet season at Khao Yai.

Season	Parameters	Description	PM _{2.5}	PM _{10-2.5}	PM ₁₀
Dry	Mass	µg m ⁻³	47 ± 10	12 ± 3.3	59 ± 11
	PM _{2.5} /PM ₁₀	Ratio	0.79 ± 0.06		
		Equation	$PM_{2.5} = 0.87 PM_{10} - 4.3; R^2 = 0.92$		
	Ion balance: Anion/Cation	Ratio	1.1 ± 0.19	0.94 ± 0.32	1.06 ± 0.17
		Slope	1.06	0.98	1.20
		R ²	0.71	0.76	0.87
	EC/TC	Ratio	0.08 ± 0.01	0.15 ± 0.04	0.08 ± 0.01
Wet	Mass	µg m ⁻³	7.0 ± 2.2	6.0 ± 2.5	12 ± 3.2
	PM _{2.5} /PM ₁₀	Ratio	0.50 ± 0.17		
		Equation	$PM_{2.5} = 0.46 PM_{10} + 1.1; R^2 = 0.51$		
	Ion balance: Anion/Cation	Ratio	0.88 ± 0.08	0.51 ± 0.44	0.52 ± 0.44
		Slope	1.10	0.57	0.80
		R ²	0.96	0.89	0.92
	EC/TC	Ratio	0.24 ± 0.12	0.11 ± 0.06	0.16 ± 0.06

Table 2. Levels and compositions of PM in dry and wet season in Khao Yai (µg m⁻³ or ng m⁻³ as indicated, 25°C and 760 mmHg).

Parameters	PM _{2.5}		PM _{10-2.5}		PM ₁₀	
	Dry	Wet	Dry	Wet	Dry	Wet
Mass	47 ± 10	7.0 ± 2.2	12 ± 3.3	6.0 ± 2.5	59 ± 11	12 ± 3.2
F ⁻	0.03 ± 0.01	0.06 ± 0.18	0.01 ± 0.01	0 ± 0	0.03 ± 0.01	0.07 ± 0.18
Cl ⁻	0.2 ± 0.1	0.1 ± 0.2	0.3 ± 0.1	0.2 ± 0.2	0.5 ± 0.2	0.3 ± 0.3
Br ⁻	0.003 ± 0.003	0 ± 0	0.003 ± 0.002	0 ± 0	0.006 ± 0.005	0 ± 0
NO ₃ ⁻	1.2 ± 0.6	0.2 ± 0.1	1.4 ± 0.5	0.5 ± 0.2	2.5 ± 0.9	0.7 ± 0.3
PO ₄ ³⁻	0.03 ± 0.02	0.03 ± 0.01	0.01 ± 0.01	0.02 ± 0.01	0.05 ± 0.02	0.04 ± 0.01
SO ₄ ²⁻	7.8 ± 2.4	1.5 ± 0.4	1.2 ± 0.6	0.3 ± 0.2	9 ± 2.6	1.8 ± 0.6
Na ⁺	0.2 ± 0.1	0.04 ± 0.03	0.5 ± 0.2	0.3 ± 0.2	0.7 ± 0.3	0.3 ± 0.2
NH ₄ ⁺	2.2 ± 0.5	0.6 ± 0.1	0.3 ± 0.2	0.03 ± 0.02	2.5 ± 0.6	0.6 ± 0.1
K ⁺	1.0 ± 0.4	0.07 ± 0.03	0.09 ± 0.04	0.2 ± 0.2	1.1 ± 0.4	0.3 ± 0.2
Mg ²⁺	0.03 ± 0.01	0.02 ± 0.005	0.08 ± 0.03	0.05 ± 0.03	0.1 ± 0.04	0.06 ± 0.03
Ca ²⁺	0.1 ± 0.1	0.08 ± 0.01	0.2 ± 0.1	0.1 ± 0.1	0.3 ± 0.1	0.2 ± 0.1
EC	0.9 ± 0.2	0.3 ± 0.1	0.2 ± 0.1	0.2 ± 0.2	1.1 ± 0.3	0.5 ± 0.2
OC	11.1 ± 3.9	1.5 ± 1.9	1.3 ± 0.4	1.8 ± 1.3	12.4 ± 3.9	3.3 ± 2.9
Al	115 ± 63	46 ± 36	180 ± 104	58 ± 41	296 ± 140	104 ± 66
Ca	190 ± 199	59 ± 27	322 ± 283	151 ± 111	512 ± 436	210 ± 110
Cd	0 ± 1	0 ± 0	0 ± 0	3 ± 6	1 ± 1	3 ± 6
Co	1 ± 0	1 ± 0	1 ± 1	1 ± 0	2 ± 2	1 ± 0
Cr	9 ± 8	14 ± 10	9 ± 8	6 ± 7	19 ± 11	20 ± 12
Cu	5 ± 6	2 ± 1	3 ± 5	1 ± 0	8 ± 10	4 ± 1
Fe	232 ± 352	149 ± 279	228 ± 230	53 ± 41	460 ± 400	201 ± 260
Mg	80 ± 55	27 ± 20	169 ± 85	59 ± 64	249 ± 92	86 ± 73
Mn	6 ± 4	4 ± 2	15 ± 19	5 ± 1	20 ± 19	9 ± 2
Mo	4 ± 2	5 ± 0	3 ± 2	5 ± 0	7 ± 4	10 ± 0
Ni	3 ± 1	1 ± 1	2 ± 1	2 ± 2	5 ± 2	3 ± 2
Pb	7 ± 5	4 ± 2	5 ± 2	4 ± 2	12 ± 5	8 ± 3
Sr	1 ± 1	0 ± 0	2 ± 1	1 ± 1	3 ± 1	1 ± 1
Ti	2 ± 1	3 ± 2	2 ± 1	1 ± 2	4 ± 2	5 ± 3
V	5 ± 1	3 ± 2	4 ± 1	5 ± 0	9 ± 2	8 ± 2
Zn	52 ± 74	15 ± 5	25 ± 53	6 ± 6	77 ± 94	21 ± 8
Si ^a	373 ± 170	124 ± 56	692 ± 310	143 ± 87	1060 ± 480	266 ± 128
% mass explained ^b	55 ± 8	71 ± 59	59 ± 16	70 ± 23	57 ± 7	74 ± 27

Note: Li⁺ and Li were not detected in the samples.

^a Si levels were estimated using a ratio of Si by PIXE to Si by ICP-OES of 1.95 based on early AIRPET inter-comparison study.

^b The sum included original OC value (not converted to OM), and elemental (not ionic) levels of Ca and Mg.

the wet period with OC of $1.5 \pm 1.9 \mu\text{g m}^{-3}$ and EC of $0.3 \pm 0.1 \mu\text{g m}^{-3}$. In $\text{PM}_{10-2.5}$, OC and EC levels in the dry period were 1.3 ± 0.4 and $0.2 \pm 0.1 \mu\text{g m}^{-3}$, respectively, while the corresponding values for the wet period were 1.8 ± 1.3 and $0.2 \pm 0.2 \mu\text{g m}^{-3}$. The maximum OC of $26 \mu\text{g m}^{-3}$ was observed on Feb 10 which was mainly present in $\text{PM}_{2.5}$, constituting about 50% mass, and negligible in $\text{PM}_{10-2.5}$ (OC levels in $\text{PM}_{2.5}$ and PM_{10} were overlapped in Fig. 1). Despite of the low OC levels in the coarse PM fraction its relative contribution to the $\text{PM}_{10-2.5}$ mass was generally significant, 10% in the dry and 30% in the wet period.

As expected, due to less human activities around the monitoring site, EC levels in $\text{PM}_{2.5}$ at KY were significantly lower than those reported in other studies for urban areas in Southeast Asia, i.e., for a suburb site in BMR (dry season of $4.3 \mu\text{g m}^{-3}$ and wet season of $3.0 \mu\text{g m}^{-3}$ as reported by Sahu *et al.*, 2011), a mixed site in Hanoi (dry season, $2.7 \mu\text{g m}^{-3}$ by Hai and Kim Oanh, 2013) or an industrial site in Vietnam (about $2.0 \mu\text{g m}^{-3}$ in both seasons by Hang and Kim Oanh, 2014). As for OC, however, the levels measured in the dry season at KY were comparable with the quoted urban sites, namely at the suburb BMR site ($13 \mu\text{g m}^{-3}$), in Hanoi ($18 \mu\text{g m}^{-3}$) or at the industrial site ($11 \mu\text{g m}^{-3}$) which indicated a significant influence of biomass burning smoke (forest fires) and/or SOA in KY. In the wet sampling period, much lower OC levels were obtained for KY, i.e., 2–3 times lower than OC reported at the above mentioned sites, which was most probably due to less forest fires and also less intensive photochemistry (hence less SOA) in the wet season in Thailand (Zhang and Kim Oanh, 2002).

The most noticeable feature in the $\text{PM}_{2.5}$ composition at KY in the dry season was the low EC coupled with high OC levels, hence resulting in low EC/TC ratios (TC = total carbon = OC + EC) of 0.08 ± 0.01 (Table 1), i.e., significantly lower than the ratio obtained at a suburb BMR site (0.23 by Sahu *et al.*, 2011) for example. This was expected as there were less local sources of EC at KY, e.g., less diesel traffic exhaust, while the contributions of OC from biomass burning and SOA may be comparable or even more significant than the urban and suburban sites. In the wet period, the ratio of EC/TC for $\text{PM}_{2.5}$ in KY was more comparable to that obtained for the SEA urban sites mentioned above and this may reflect a characteristic of the regional air masses affecting the site.

The linear regression between EC and OC in the dry season showed the R^2 value of 0.43 for $\text{PM}_{2.5}$ and 0.21 for $\text{PM}_{10-2.5}$ which indicated their weak correlations (Fig. S4, SI). Other SEA urban sites commonly reported higher R^2 (above 0.80) of the linear regression between EC and OC in $\text{PM}_{2.5}$ (Hang and Kim Oanh, 2014; Hai and Kim Oanh, 2013). In the wet sampling period, the correlation between EC and OC was also weak with R^2 varying from negligible for $\text{PM}_{2.5}$ to 0.47 for $\text{PM}_{10-2.5}$ (Fig. S4, SI). Overall, these relatively poor correlations between EC and OC suggested that the dominant contributing sources to PM at the site varied between sampling days and they had different EC/OC ratios. In particular, a large part of OC obtained at this site may be SOA and this may be linked to abundant

biogenic VOC precursors present around this forest site, but further studies are required to confirm the association.

In fact, the EC levels measured at KY appeared to be comparable to those reported for a rural upstate New York site of Pinnacle State Park (PSP) where the monthly levels (during 2002–2004) were around $0.2 \mu\text{g m}^{-3}$ (Bae *et al.*, 2006). However, the PSP site had lower OC than KY with the monthly level below $3 \mu\text{g m}^{-3}$. The EC and OC levels in KY were also significantly above the mean values, reported by Lim *et al.* (2003), at the background Pacific Ocean (0.09 and $0.21 \mu\text{g m}^{-3}$, respectively), Asian-influenced Pacific Ocean (0.29 and $0.70 \mu\text{g m}^{-3}$), offshore of Japan (0.27 and $1.00 \mu\text{g m}^{-3}$) and Sea of Japan (0.66 and $2.43 \mu\text{g m}^{-3}$). Our EC levels were somewhat higher than the equivalent black carbon (EBC, optical method) reported for a Western Tibetan Plateau site which had the daily levels varying from 33.6 to 330.2 ng m^{-3} (Zhu *et al.*, 2016) and indeed were much above the EBC levels measured at the South Pole (monthly EBC < 8 ng m^{-3}) reported in Sheridan *et al.* (2016). The EC and OC results thus showed that even the KY site was in a relatively remote forest area of Thailand the effects of human activities on pollution levels were still clearly shown as compared to other more “truly” remote areas in Asia reported by Lim *et al.* (2003) or the South Pole (Sheridan *et al.*, 2016).

Ionic Composition

Sulfates contributed the largest fraction among all analyzed ions and elements, i.e., 17–22% of $\text{PM}_{2.5}$ mass and 5–10% of $\text{PM}_{10-2.5}$ mass in both sampling periods, hence it was only second to OC among all the species presented in Table 2. Other ions that were found significantly in $\text{PM}_{2.5}$ included NH_4^+ ($2.2 \mu\text{g m}^{-3}$ in dry and $0.6 \mu\text{g m}^{-3}$ in wet sampling period) and K^+ ($1.0 \mu\text{g m}^{-3}$ in dry and $0.07 \mu\text{g m}^{-3}$ in wet period), which were well above those found in $\text{PM}_{10-2.5}$. Levels of NO_3^- were higher in $\text{PM}_{10-2.5}$ ($1.4 \mu\text{g m}^{-3}$ in dry and $0.5 \mu\text{g m}^{-3}$ in wet period) than in $\text{PM}_{2.5}$ (1.2 in the dry and $0.2 \mu\text{g m}^{-3}$ in the wet period). Noticeable levels of Na^+ , Cl^- , Mg^{2+} and Ca^{2+} in $\text{PM}_{10-2.5}$ (Table 2) resulted in their relatively high contributions (in %) to $\text{PM}_{10-2.5}$ mass. A summary of the ion levels in both PM fractions is presented in Fig. 2.

The ion balance was analyzed for $\text{PM}_{10-2.5}$ and $\text{PM}_{2.5}$ in the dry and wet sampling periods, respectively. The sum (in $\mu\text{Eq m}^{-3}$) of cations (NH_4^+ , Na^+ , K^+ , Mg^{2+} , Ca^{2+}) was compared with the sum of anions (NO_3^- , Cl^- , PO_4^{2-} , SO_4^{2-}) and the results are summarized in Table 1. In the dry period, the ratio between anions to cations was 1.1 ± 0.19 for $\text{PM}_{2.5}$, i.e., slightly acidic fine particles, and 0.94 ± 0.32 for $\text{PM}_{10-2.5}$, i.e., slightly basic coarse particles. In the wet period, the ratios were less than 1.0 for both size fractions hence suggesting that PM in the wet period was less acidic and this may be explained by more efficient wet removal of acidic particles. The scatter plots between cation and anion equivalents are presented in Fig. S5, SI which show good linearity with R^2 of 0.71–0.96 for $\text{PM}_{2.5}$ and 0.76–0.89 for $\text{PM}_{10-2.5}$.

The ammonium balance was checked and the estimated NH_4^+ in balance with its sulfate and nitrate salts ($\text{NH}_4^+ =$

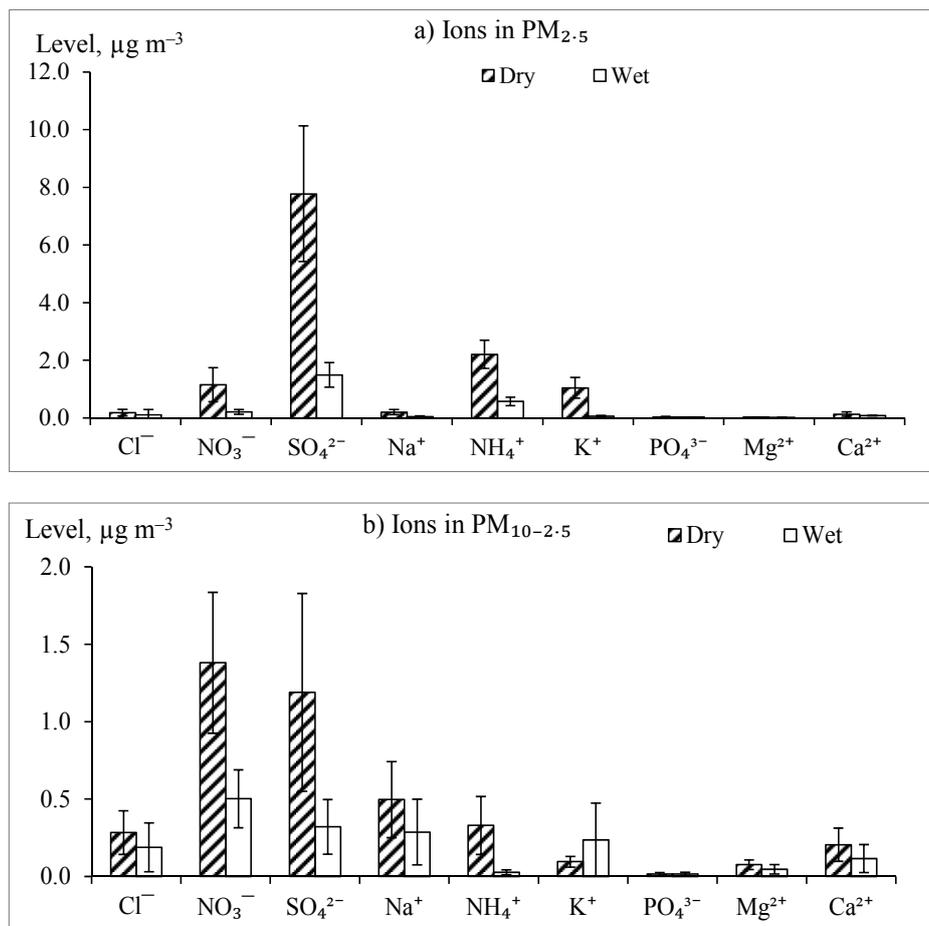


Fig. 2. Ionic composition in PM at Khao Yai during dry and wet sampling periods (note different scales of Y-axis).

$0.29 \text{NO}_3^- + 0.38 \text{SO}_4^{2-}$; Chow *et al.*, 2006) was compared with the measured values (Fig. S6(a), SI) which showed that the estimated NH_4^+ in the dry season $\text{PM}_{2.5}$ was about 1.5 times of the measured. When NH_4^+ was estimated with the assumption of ammonium bisulfate and nitrate salts ($\text{NH}_4^+ = 0.29 \text{NO}_3^- + 0.192 \text{SO}_4^{2-}$; Chow *et al.*, 2006) the estimated NH_4^+ was only 0.83 of the measured in the dry season $\text{PM}_{2.5}$ (Fig. S6(b), SI). In both cases, high R^2 values of about 0.87 were obtained for the regression lines, as seen in Figs. S6(a) and S6(b), SI. This suggested that both ammonium sulfate and bisulfate salts may coexist in the dry season $\text{PM}_{2.5}$ along with other non-ammonium sulfate and nitrate compounds. For $\text{PM}_{2.5}$ of the wet period, the ratio between the estimated NH_4^+ in balance with sulfates was quite close to 1.0 (1.09) while that with bisulfates was only 0.61 (high R^2 of about 0.90 was obtained for both cases) indicating that the majority of ammonium may be in the form of sulfate compounds. For the coarse fraction ($\text{PM}_{10-2.5}$), in the dry season the ratios between estimated and measured NH_4^+ were high, i.e., 1.7 for bisulfates and 2.4 for the sulfates balance, while R^2 was low (0.36 and 0.54, respectively). In the wet period, there was a negligible correlation between measured and estimated NH_4^+ (with either assumption) for coarse PM, as shown in Figs. S6(a) and S6(b), SI.

There exists a wide range of inorganic ammonium

compounds in PM. The secondary inorganic aerosol (SIA) of ammonium that is formed in the atmosphere from the precursor gases normally constitutes a large portion of $\text{PM}_{2.5}$. These ammonium compounds may include NH_4HSO_4 , $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3 and NH_4Cl . Sulfuric acid (H_2SO_4) has a high affinity to NH_3 , hence would first react with the available ambient NH_3 to form ammonium sulfate and bisulfate. The remaining NH_3 would then react with HNO_3 and HCl to form the nitrate and chloride compounds (Behera *et al.*, 2013 and references therein). The sulfate and nitrate particles could also present in the forms of calcium and magnesium salts (Arruti *et al.*, 2011). Sea salt particles would mainly present in the coarse fraction (Hewitt and Jackson, 2005) hence NaNO_3 , an aging product of sea salt, would mainly be found in the coarse fraction. In fact, in our samples, levels of Na^+ , Ca^{2+} and Mg^{2+} were found more abundant in the coarse than the fine PM (Table 2, Fig. 2). Thus, there should expect discrepancies between the estimated and measured NH_4^+ in the PM samples, especially for the coarse PM, when assuming the balance of ammonium with only sulfate, bisulfate and nitrate ions.

Elemental Composition

Major elements found in $\text{PM}_{2.5}$ were Si, Fe, Ca, Al, Mg and Zn (Table 2 and Fig. 3) which had average levels in $\text{PM}_{2.5}$ of 100–400 ng m^{-3} in the dry season. In the wet season,

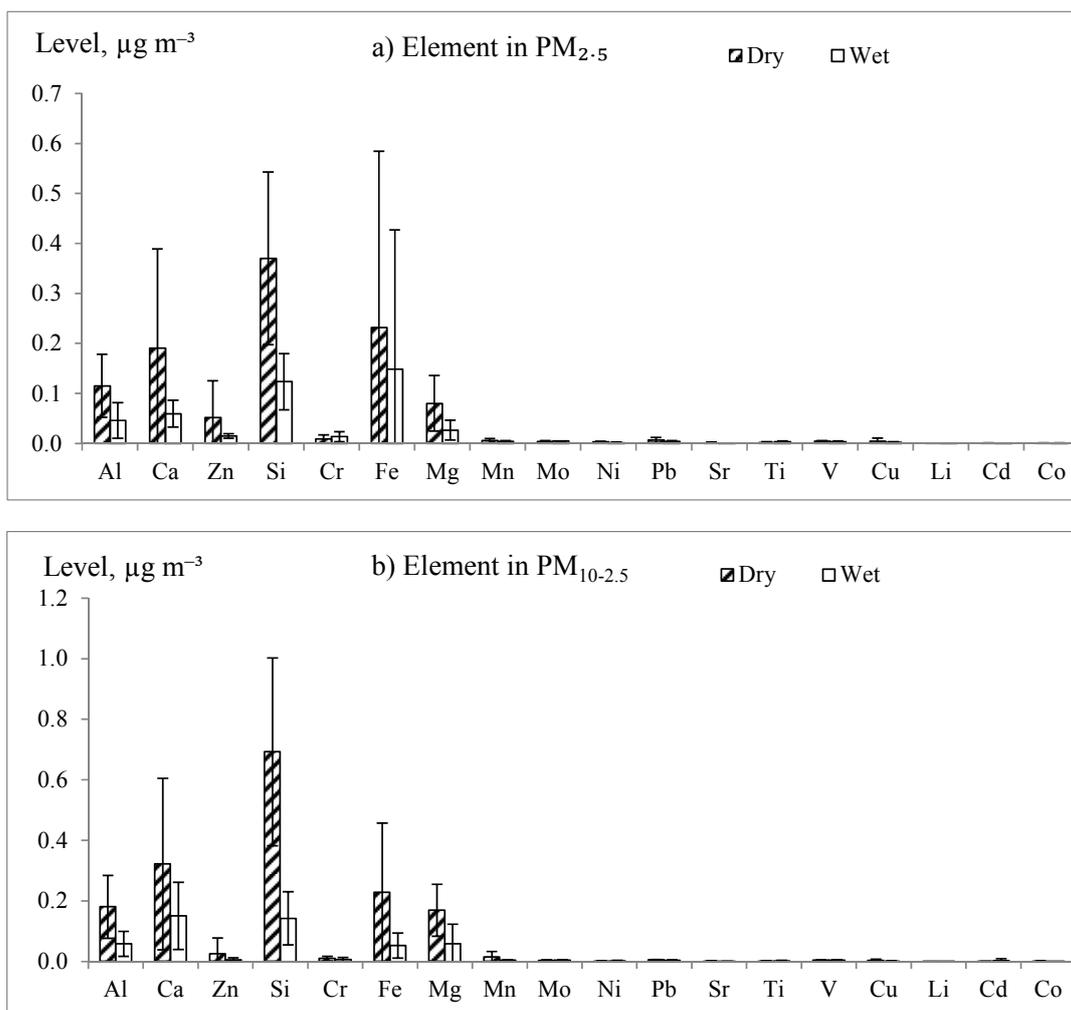


Fig. 3. Elemental compositions in PM at Khao Yai during dry and wet sampling periods.

the element levels were generally below 60 ng m^{-3} except for Fe which had a high level, averaged at 150 ng m^{-3} , but with a wide fluctuation range. Average levels of some major elements in $\text{PM}_{10-2.5}$ were $200\text{--}700 \text{ ng m}^{-3}$ in the dry season, i.e., higher as compared to the wet season levels of $60\text{--}150 \text{ ng m}^{-3}$. Other elements listed in Table 2 were found below 10 ng m^{-3} in both PM size fractions. In particular, Pb levels were about $4\text{--}7 \text{ ng m}^{-3}$, slightly higher in the dry season, that may be related to the combustion origin of distant industrial sources (in $\text{PM}_{2.5}$), and/or local soil/dust (in $\text{PM}_{10-2.5}$) because leaded gasoline has been phased out in Thailand since January 1996 (Wangwongwattana, 1999). The levels of crustal elements found at this forest site were lower than other urban areas in Asia reported in Kim Oanh *et al.* (2006) which were largely due to more rainfall in Khao Yai and less vehicular activities (less suspended soil/road dust) and less construction activities. Nevertheless, the relatively high levels of major crustal elements found in $\text{PM}_{10-2.5}$, especially in the dry season, indicated the presence of soil/road dust in the samples. Note that the Si levels determined by the ICP-OES method may be underestimated and the ratio between unadjusted ICP-OES Si to Al was low (1.8 ± 0.9). After adjusting the Si results using the average ratio (1.95)

between Si by PIXE and Si by ICP-OES, the Si content in our samples showed a reasonable ratio to Al, i.e., above 3.0 which is expected for this remote site (Kenneth, 1976). The adjusted Si levels, presented in Table 2 and Fig. 2, were used for the data analysis in this paper.

Sources and Contributions

Reconstructed Mass

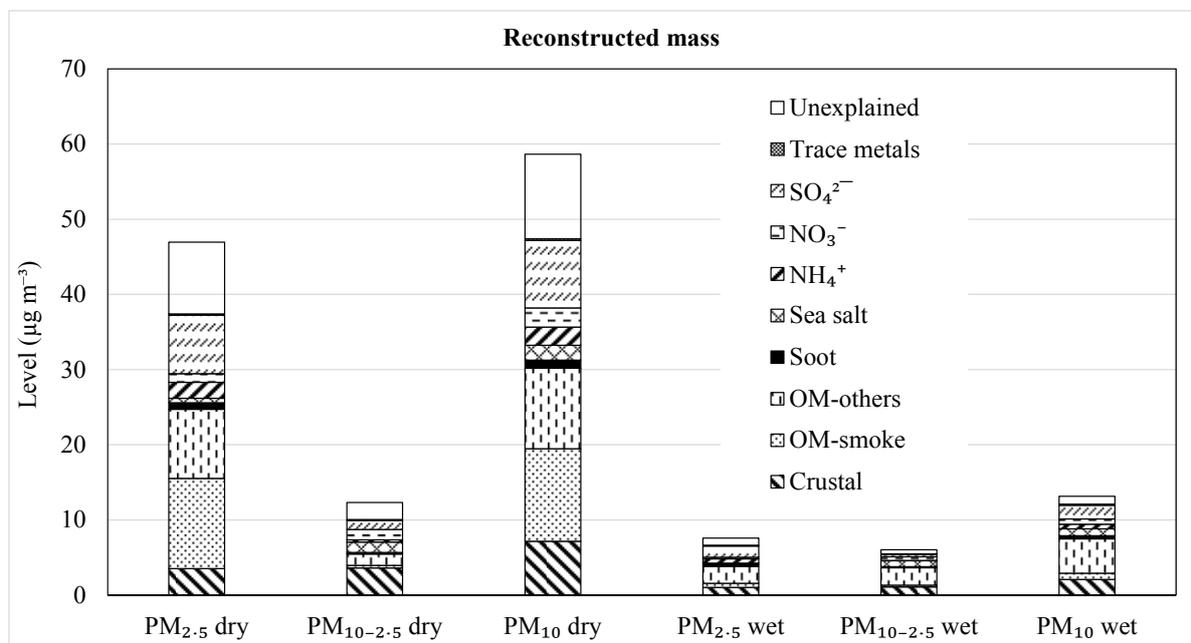
The sum of all analyzed species including OC (before converting to OM) accounted for about 55–71% of the $\text{PM}_{2.5}$ mass and 59–70% of the $\text{PM}_{10-2.5}$ mass (Table 2). After conversion of OC to OM, the portion of the total measured mass explained by the reconstructed mass substantially increased, i.e., to 80–87% for $\text{PM}_{2.5}$ and 80–91% for $\text{PM}_{10-2.5}$ (Table 3). The major mass groups are shown in Fig. 4 while Table 3 presents the combined group of inorganic particles. The additional indicators of K-smoke and marine- SO_4^{2-} (Table 3) were not included in the total mass explained to avoid double counting.

In the dry season, the largest contribution to $\text{PM}_{2.5}$ mass ($47 \text{ } \mu\text{g m}^{-3}$) was from the OM-smoke (26%), inorganic particles (24%) and OM-others (20%). The crustal group contributed about 7.5% while other groups had minor

Table 3. Major mass groups in $\mu\text{g m}^{-3}$ and share (%) of fine and coarse PM at Khao Yai.

Parameters	PM _{2.5} , dry season		PM _{10-2.5} , dry season		PM _{2.5} , wet season		PM _{10-2.5} , wet season	
	Mass	Share, %	Mass	Share, %	Mass	Share, %	Mass	Share, %
Mass	47 ± 10		12 ± 3.3		7.0 ± 2.2		6.0 ± 2.5	
<i>Major mass groups</i>								
Crustal	3.5 ± 1.7	7.5	3.6 ± 1.4	29.4	1.0 ± 0.7	13.3	1.1 ± 0.7	17.7
OM-smoke	12 ± 5.3	25.5	0.3 ± 0.5	2.7	0.6 ± 0.5	7.6	0.3 ± 0.2	4.1
OM-others	9.2 ± 6.6	19.6	1.5 ± 0.7	12.3	2.2 ± 3.1	29.4	2.3 ± 1.7	38.7
Soot	0.9 ± 0.2	1.9	0.2 ± 0.1	1.6	0.3 ± 0.1	3.9	0.1 ± 0.1	2.1
Sea salt	0.6 ± 0.2	1.2	1.4 ± 0.6	11.2	0.1 ± 0.1	1.6	0.8 ± 0.5	13.2
Inorganic particles	11 ± 2.5	23.6	2.7 ± 0.8	23.3	2.3 ± 0.5	30.0	0.8 ± 0.3	14.0
Trace metals	0.1 ± 0.1	0.3	0.1 ± 0.1	0.71	0.1 ± 0.01	0.8	0.05 ± 0.02	0.8
Mass explained		80 ± 10		80 ± 15		87 ± 21		91 ± 15
<i>Other indicators</i>								
K-smoke	0.9 ± 0.4		0.03 ± 0.04		0.04 ± 0.03		0.2 ± 0.2	
Marine-SO ₄ ²⁻	0.1 ± 0.02		0.1 ± 0.1		0.01 ± 0.01		0.1 ± 0.1	

Note: Inorganic particles amount is the sum of NH_4^+ , NO_3^- and non-marine- SO_4^{2-} .

**Fig. 4.** Major mass groups of PM at Khao Yai during dry and wet sampling periods.

contributions (soot: 2%, sea salt: 1%, trace metals: 0.3%). OM-others at this site is likely linked to SOA of local and distant sources origins while inorganic particles should represent SIA in the fine PM fraction. The results, thus, show a significant contribution of biomass burning to PM_{2.5} mass in the dry season and secondary particles, both inorganic (SIA) and organic (OM-others).

For the coarse fraction mass ($12 \mu\text{g m}^{-3}$) measured in the dry season, the largest share was from the crustal group (30%) hence indicating the contribution from local soil dust. OM-smoke, as expected, had only a minor contribution (3%) while OM-others contributed 12%. The OM-others in the coarse fraction may be largely associated with soil and plant debris (local sources). Among the inorganic particle group, SO_4^{2-} and NO_3^- had significant contributions (about 10% each) while NH_4^+ had only a minor contribution (2%).

The inorganic particle group of the coarse PM may be associated with the aged sea salt (NaNO_3) and sulfate particles (mostly non-ammonium) which is in line with the ammonium balance discussed above. The sea salt group contributed about 11% of the coarse particle mass. Note that the marine- SO_4^{2-} had a minor contribution to the PM mass, about $0.1 \mu\text{g m}^{-3}$ in both size fractions (Table 3).

In the wet season, the major mass groups of PM_{2.5} (total mass of $7 \mu\text{g m}^{-3}$) were inorganic particles (SIA, 30%), OM-others (SOA, 29%) and crustal (13%). The OM-smoke shared only 8% PM_{2.5} mass which indicated a small contribution from biomass burning smoke. For the coarse fraction mass ($6 \mu\text{g m}^{-3}$), the major groups were OM-others (39%, largely by soil and plant debris), crustal (18% soil/dust), inorganic particles (14%) and sea salt (13%). The inorganic particles were mainly consisted of NO_3^- ($0.5 \mu\text{g m}^{-3}$, 8% PM mass),

SO_4^{2-} ($0.3 \mu\text{g m}^{-3}$, 5% PM mass) while NH_4^+ had only minor contribution ($0.03 \mu\text{g m}^{-3}$, < 0.5% PM mass).

The SIA mass measured in fine PM at KY could be mainly related to the transport of pollution from distant sources, e.g., the precursors from industry located in the surrounding towns (regional) and other upwind sources (long-range, largely transboundary). Likewise, the presence of the crustal group in $\text{PM}_{2.5}$ in both seasons should indicate not only the local soil dust (inside the park) but also a potential contribution from cement factories located in the surrounding provinces (regional sources). Soot particles in $\text{PM}_{2.5}$ may also have both local and distant origins. Likewise, SOA in $\text{PM}_{2.5}$ may be linked to both the regional and long range transported SOA particles and their precursors as well as the SOA particles formed from local biogenic precursors.

The mass group results indicated the following major contributing factors to $\text{PM}_{2.5}$ in the dry season at KY: (1) biomass burning (largely local) indicated by OM-smoke ($12 \mu\text{g m}^{-3}$, i.e., 26% by OM-smoke alone), (2) SIA (24%, mainly from distant sources), (3) SOA (20%, both local and distant sources) and crustal (8%, both local soil dust and distant sources). In the wet season, the major contributions to $\text{PM}_{2.5}$ were: (1) SOA (29%), (2) SIA (30%) and (3) crustal (13%), while biomass burning had a relatively small contribution (OM-smoke: $0.6 \mu\text{g m}^{-3}$ or 8% of PM mass). Thus, the local contribution to $\text{PM}_{2.5}$ pollution at KY was more significant in the dry season which was mainly from biomass burning (forest fires). The transport from distant sources contributed substantially to $\text{PM}_{2.5}$ levels in both seasons, more significant in the wet season when the main local source of biomass burning was less pronounced.

The major contributing factors to $\text{PM}_{10-2.5}$ were: (1) soil dust (> 50%, collectively by crustal group and OM-others groups in both seasons), (2) inorganic particles (23% in dry and 14% in wet season), and (3) sea salt (> 10%, both seasons). Most of the contributing factors to the coarse PM could be linked to the local sources except for the sea spray which should be linked to the transport from the sea (the nearest coastal line is more than 100 km away from the site).

HYSPLIT Patterns and Potential Long Range Transport Pollution

The HYSPLIT 5-day backward trajectories were grouped into patterns based on their origins and pathways. The results showed that the southwest trajectory patterns (SWM) were observed on all days (100%) during the wet sampling period which was consistent with the dominant southwest monsoon in the season for the central part of Thailand. This marine air mass type was originated from the Indian Ocean and had a long marine pathway before arriving to KY hence it had generally low PM pollution (Table 4). These air masses may bring aged sea salt particles to the site, which is also shown in the reconstructed mass results (Table 3).

In the dry sampling period, three (3) major HYSPLIT trajectory patterns were identified (Table 4 and Fig. S7, SI). The E-M-SW pattern, the most frequently observed (59% of 39 sampling days), had the origin from the Pacific Ocean, near the Philippines (E to KY), and had a marine pathway (M) before arriving to the site from the Gulf of Thailand following the south-southwest direction (SW). Its longest marine pathway, among all the 3 HYSPLIT patterns found in the dry season, induced the lowest PM levels, i.e., average $\text{PM}_{2.5}$ of $43 \mu\text{g m}^{-3}$ and $\text{PM}_{10-2.5}$ of $11 \mu\text{g m}^{-3}$ (Table 4). The NE-CMC pattern (Fig. S7, SI) originated from the continental China, NE of KY, and had a continental pathway (C) through China and a marine pathway (M) over the Bac Bo Gulf of Vietnam, and then a continental pathway (C) over Vietnam, Laos and Cambodia territories before arriving to KY. This pattern, with an occurrence frequency of 33%, had the second highest average PM levels (average $\text{PM}_{2.5}$ of $51 \mu\text{g m}^{-3}$ and $\text{PM}_{10-2.5}$ of $14 \mu\text{g m}^{-3}$). In particular, Jan 18, the day with the highest $\text{PM}_{2.5}$ measured at KY in the sampling period (Fig. 1), belonged to this pattern (shown in Fig. S7, SI). There was no forest fire reported in the park on that day. It was also a Tuesday hence may not be specifically affected by local tourist activities. The high $\text{PM}_{2.5}$ level measured on Jan 18 suggested that the contributions from distant sources may be significant.

The third pattern was named E-M-E to indicate its origin in the Pacific Ocean, off the coast of Southern Vietnam (E-

Table 4. Levels and composition of PM in different HYSPLIT patterns.

Parameter	In dry season			In wet season	
	E-M-SW (59%)	NE-CMC (33%)	E-M-E (8%)	SWM (100%)	
$\text{PM}_{2.5}$	Mass, $\mu\text{g m}^{-3}$	43.2 ± 9.0	51.2 ± 10.9	56.3 ± 9.2	5.6 ± 2.0
	SO_4^{2-} , $\mu\text{g m}^{-3}$	7.6 ± 2.2	7.8 ± 2.7	8.8 ± 2.9	1.5 ± 0.4
	EC, $\mu\text{g m}^{-3}$	0.8 ± 0.3	1.0 ± 0.1	0.8 ± 0.3	0.3 ± 0.1
	TC, $\mu\text{g m}^{-3}$	11.2 ± 3.8	13.3 ± 4.5	12.3 ± 2.9	1.8 ± 1.8
	EC/TC	0.08 ± 0.01	0.08 ± 0.02	0.07 ± 0.02	0.24 ± 0.12
	EC/ SO_4^{2-}	0.12 ± 0.06	0.16 ± 0.12	0.11 ± 0.08	0.21 ± 0.08
	$\text{SO}_4^{2-}/\text{OC}$	0.85 ± 0.41	0.72 ± 0.37	0.81 ± 0.35	1.68 ± 0.98
$\text{PM}_{10-2.5}$	Mass, $\mu\text{g m}^{-3}$	10.7 ± 2.9	14.1 ± 3.5	14.1 ± 0.9	5.8 ± 2.7
	SO_4^{2-} , $\mu\text{g m}^{-3}$	1.1 ± 0.5	1.4 ± 0.8	1.0 ± 0.4	0.3 ± 0.2
	EC, $\mu\text{g m}^{-3}$	0.21 ± 0.05	0.21 ± 0.03	0.2 ± 0.05	0.2 ± 0.2
	TC, $\mu\text{g m}^{-3}$	1.4 ± 0.50	1.5 ± 0.40	1.8 ± 0.30	2.0 ± 1.4
	EC/TC	0.14 ± 0.38	0.15 ± 0.04	0.11 ± 0.01	0.11 ± 0.06
	EC/ SO_4^{2-}	0.23 ± 0.12	0.21 ± 0.15	0.22 ± 0.04	0.82 ± 0.93
	$\text{SO}_4^{2-}/\text{OC}$	0.91 ± 0.44	1.0 ± 0.60	0.67 ± 0.29	0.28 ± 0.22

KY), passing along the coastal line of Southern Vietnam and over the Gulf of Thailand (M) before arriving to the site from the east (E). As compared to the E-M-SW pattern discussed above, it had a longer continental pathway and a slower movement above the Northern Thailand before arriving to the site. This pattern had the lowest occurrence frequency (8%) but the highest PM levels (average at $56 \mu\text{g m}^{-3}$ for $\text{PM}_{2.5}$ and $14 \mu\text{g m}^{-3}$ for $\text{PM}_{10-2.5}$) and the highest sulfates in $\text{PM}_{2.5}$ ($8.8 \mu\text{g m}^{-3}$).

As compared to other patterns in the dry season, the NE-CMC pattern had the highest EC and TC levels in $\text{PM}_{2.5}$, and the highest ratio between EC (indicator of primary PM) and SO_4^{2-} (indicator of SIA in $\text{PM}_{2.5}$), i.e., $\text{EC}/\text{SO}_4^{2-} = 0.16$. The continental origin and long continental pathway of the air masses belonging to the NE-CMC pattern may explain a more excessive presence of primary particles (including EC) which were transported to the site. The ratio between sulfates (SIA) and OC was the lowest for the NE-CMC pattern, indicating a more relative importance of SOA in this air mass type.

CONCLUSIONS

In the dry season, the levels of $\text{PM}_{2.5}$ at the remote forest site of KY were comparable with those reported for several urban locations in Southeast Asia while $\text{PM}_{10-2.5}$ levels were remarkably lower. During the wet sampling period, both fine and coarse PM at KY were much lower than urban sites. The PM composition in KY was also different from that obtained in urban areas. Significantly lower EC but comparable high OC (to urban areas) measured at KY yielded low EC/TC ratios, specifically for $\text{PM}_{2.5}$ in the dry season. Low EC levels suggested a low contribution from fossil fuel combustion, especially by diesel vehicles, around the site.

The contribution from local sources to $\text{PM}_{2.5}$ at KY was more substantial in the dry season. The major local source was biomass burning (forest fire) which shared above 25% of the PM mass, as indicated by the OM-smoke group alone. Secondary PM, both organic (local and distant sources) and inorganic (mainly from distant sources), contributed significantly to $\text{PM}_{2.5}$ in both seasons but relatively more important (higher percentage) in the wet season when local source of biomass burning was not pronounced. The transport of secondary particles and their gaseous precursors from distant sources to the site appeared to be more important contributing factors to $\text{PM}_{2.5}$ levels at KY than local sources in both seasons.

The major source factors of $\text{PM}_{10-2.5}$, identified based on the reconstructed mass, were soil dust, organic matter (OM-others), inorganic particles and aged sea salt. Most of the contributors to the coarse PM could be linked to local sources except for the sea spray which would be linked to a distant source origin.

The HYSPLIT backward trajectory patterns also confirmed the possibility of the long range transport pollution. The patterns having longer continental pathways were characterized by higher $\text{PM}_{2.5}$ levels measured at KY during the dry season. In the wet season period, when the

marine air mass type dominated, the levels of PM measured at the site were quite low. The sampling periods in this study were relatively short, especially for the wet season, hence further studies are required to collect systematic data on PM mass and composition which can be analyzed by receptor models to produce more quantitative source apportionment results.

ACKNOWLEDGMENTS

We would like to thank the Swedish International Development Agency for the generous financial support, without which the study would not be possible. The authors would like to thank the Khao Yai National Park administration for the kind support and assistance during the sampling period. The Thailand Pollution Control Department is acknowledged for providing air quality monitoring data for comparison. The Khao Kheow meteorological station is thanked for providing the meteorological data. Special thanks to Dr. Tami Bond at University of Illinois, Urbana-Champaign for her generous support in the EC/OC analysis. The air quality research group at AIT, especially Ms. Do Thanh Canh and Dr. Prapat Pongkiatkul, is thanked for their various contributions during the sampling and sample analyses.

SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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Received for review, December 25, 2015

Revised, April 9, 2016

Accepted, April 15, 2016