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Annual emissions of air toxics emitted from crop residue open burning in Southeast Asia over the period of 2010–2015

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ABSTRACT

Southeast Asia (SEA) has large agricultural crop production and huge amounts of crop residues generated annually are commonly burned in the field to quickly clear land for the next crop planting. This study developed annual emission inventory for crop residue open burning (CROB) covering 17 emission species/groups for 10 SEA countries during 2010–2015, illustrated with relative contributions by country and by crop type. The fractions of crop residue subjected to open burning (OB), a large source of uncertainty in the activity data, compiled from surveys in SEA were significantly higher than those suggested in international databases. Emission factors for rice and maize residue OB were obtained from field experiments conducted in Thailand. The best estimates of the annual emissions averaged over 6 years, of air toxics were: 32 Gg of polycyclic aromatic hydrocarbons, 0.03 Gg organo-chlorinated pesticides, 292 Gg total chlorines, and 94 g I-TEQ dioxins. Emissions of $PM_{2.5}$, BC and OC, in Tg yr^{-1}, were respectively 1.8, 0.08 and 0.8. The coefficients of variation of annual emissions during the period were relatively small (2.6–8.6% depending on species) but still showed an increasing trend that reflected the changes in production of major crops during the period. Regionally, CROB shared 10–43% of the total biomass open burning emissions but varying with country: by far dominant in Vietnam and Philippines, and much less dominant in Indonesia, Myanmar, and Thailand. Rice straw open burning was the most dominant (19–97%) in the total CROB emissions of the 8 considered crops. The spatial distributions of annual emissions (0.1° x 0.1°) showed higher emission intensity over the areas cultivated with rice and sugarcane, while higher monthly emissions coincided with major harvesting periods in the dry season. The obtained EI data can be further used for air quality modeling to assess effects of CROB emission and to promote non-OB alternatives in the region.

1. Introduction

Southeast Asia (SEA) is a dynamic region with fast growing economy and population. Most of the countries in SEA are agrarian with economic development based largely on the agricultural sector. The region also houses 3 countries among the world's top ten rice exporters in 2015 (WTE, 2016). To ensure the food security for more than 600 million people in the region and to meet the export demand, agricultural production has been intensified and that has been accompanied by the generation of a huge quantity of crop residues annually. Crop residue open burning (CROB) is viewed by regional farmers as the cheapest and fastest way to clear land for the next crop, and hence has been increasingly practiced for decades (Kanabkaew and Kim Oanh, 2011). Traditional slash and burn shifting cultivation (SBS) is still practiced in several countries in the region such as Indonesia (Ketterings et al., 1999; Murdiyarso et al., 2005) and some other SEA countries (Li et al., 2014). SBS is commonly believed to enrich the soil but heat from burning may also damage soil structure and remove the surface humic organic matter (Sanchez et al., 2005) while at the same time, huge quantities of agro-residue biomass are wasted.

Several alternative options to reuse the crop residues (i.e. mushroom growing, ploughing for on-site degradation, cooking fuel, animal feed, etc.) are available, but these are getting less practiced as farmers get wealthier. There are still practical problems to sustain business models that incorporate sufficient incentives to encourage farmers to stop open burning (Kim Oanh, 2012). Awareness raising focusing on the

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negative impacts of CROB, specifically the effects of smoke on human health, is necessary to encourage farmers to use non-open burning alternatives for the crop residue management. This approach in turn needs reliable information of the amount of toxic air pollutants released annually from this activity in every country of the region.

Presently, the CROB impacts are often overlooked in many SEA countries and normally gain less concern from the society as compared to the catastrophic SEA transboundary haze caused by forest fires. By nature, CROB is the low temperature combustion of the vegetation hence would release large quantities of products of incomplete combustion (PIC) which are toxic air pollutants, such as particulate matter (PM) with black carbon (BC) and organic carbon (OC) components, carbon monoxide (CO) and volatile organic compounds (VOCs). Certain amounts of nitrogen oxides (NO_x) and sulfur oxides (SOx) are also released along with key greenhouse gases (GHGs) like methane (CH_4), nitrous oxide (N_2O), and carbon dioxide (CO_2) although the released CO_2 is believed to be taken up by the next season's crop growth. In addition to the common emission species listed above, a range of semi-VOCs including the persistent organic pollutants, such as polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/PCDFs, here referred to as dioxins for short), polychlorinated biphenyls (PCBs), and polycyclic aromatic hydrocarbons (PAHs) are also emitted from the biomass burning (Estrellan and Iino, 2010). CROB also emits considerable amount of chlorine compounds with atmospheric lifetime varying from a day to several years that may catalytically affect tropospheric ozone concentrations (Burklin et al., 2002) and stratospheric ozone destruction (Khalil et al., 1999; Lobert et al., 1999). Therefore, it is important to comprehensively quantify the CROB emissions and thereby provide necessary scientific basis to promote the environment-friendly crop residue management practices in the SEA countries.

Previous emission inventory (EI) studies of CROB for SEA differ by the approaches used for the emission estimation (i.e. statistical data vs. satellite data), scale (i.e. country based, regional or global), and level of detail provided in the emissions database (i.e. species coverage, temporal and spatial information). The global databases so far provide only preliminary estimates of CROB emissions using the aggregate amount of crop residues burnt in the open field of the developing and developed countries, respectively (Seiler and Crutzen, 1980; Hao and Liu, 1994). Further refined EI databases were also prepared by using the available country specific information to some extent (Yevich and Logan, 2003; Andreae and Merlet, 2001; Akagi et al., 2011). More up-to-date global EI for CROB is provided by the Emission Database for Global Atmospheric Research (EDGAR) v4.3.1 for the base year of 2014 and with the spatial resolution of 0.5° and 0.1° using international crop statistics. Published regional EI databases, e.g. Streets et al. (2003) at 0.5° × 0.5° for all of Asia, and Shi and Yamaguchi (2014) at 0.01° × 0.01° for SEA, did not include dioxins and PAHs. The national CROB EI databases for Thailand and Indonesia developed using the national statistics and local survey results (Kanabkaew and Kim Oanh, 2011; Permadi and Kim Oanh, 2013) provided more detailed temporal (monthly) and spatial emissions (district/provincial) but also did not include these toxic semi-VOC. Gadde et al. (2009) estimated the national annual emissions from rice straw (RS) open burning for India, Thailand and Philippines, roughly included annual emissions of dioxins and PAHs but without spatial and temporal distributions. These semi-VOC emissions are included in a few existing global studies, such as dioxins in Brzuzy and Hites (1996) and UNEP (1999) or PAHs in Zhang and Tao (2008), but the emissions specifically for CROB have not been explicitly identified. None of these databases included emissions of reactive chlorine species from CROB. The global inventory of reactive chlorine compounds was only reported in the “Reactive Chlorine Emissions Inventory (RCEI)” within the “Global Emissions Inventory Activity” (Lobert et al., 1999). For urban ozone air quality, reactive chlorine compounds may be important as they can catalytically enhance the tropospheric ozone formation (Burklin et al., 2002).

Therefore, the SEA CROB emissions should be updated to more recent base years with consideration on the inter-annual variation and semi-VOC species. The EI should be based more on local survey data regarding crop practices and available emission factors (EFs) relevant for the region. In addition, the temporal variations of CROB emissions should be characterized as it is particularly important because of variations in local agricultural practices in SEA and the distinct dry and wet seasons. Local surveys would provide specific information to estimate the actual amounts of the residue from different crop types subjected to open burning (OB) and the common burning practices that strongly affect the EFs. For example, the use of satellite-derived fire counts in some existing EI databases for CROB emissions would not fully capture CROB emissions in SEA because the burning is done mainly in the late afternoon (Kanabkaew and Kim Oanh, 2011; Permadi and Kim Oanh, 2013) and occurs sporadically with short durations, i.e. about 1 h (Kim Oanh et al., 2011), while the Moderate Resolution Imaging Spectroradiometer (MODIS) of Terra and Aqua satellite provides only snapshots of the region at 10.30 a.m. and 13.30 p.m. local standard time (LST).

This study considers the emissions from crop residue open/field burning in SEA with a focus on emissions of trace gases, PM, semi-VOC species of PAHs, dioxins and chlorinated pesticides (OCPs), reactive chlorines (5 compounds) and GHGs. The EI was done for 10 SEA countries for each year over a 6-year period (2010–2015), incorporating country specific activity data (local surveys) and available regional specific EFs. The EI results are presented as the low, high and best estimates to include the uncertainty range. Further, monthly emissions and the gridded emissions with a resolution of 0.1° x 0.1° were prepared which can be further used for three-dimensional regional air quality modeling to assess the impacts of current CROB emissions and emission reduction measures on the air quality, human health and environment. Our annual emission results for CROB in SEA (SEA CROB) are comparatively analyzed with the forest fires data taken from Global Fire Emission Database Version 3.1 (GFED3.1) (van der Werf et al., 2010) to reveal the relative importance of the sources.

2. Methodology

2.1. Emissions calculations

The EI was developed following the common approach for this source type (Shrestha et al., 2013; Hao and Liu, 1994). Accordingly, the annual emission from burning of crop residues was calculated using Equation (1).

$$E_{m,k} = M_k \times \eta_k \times EF_{m,k} \quad (1)$$

Where,

$E_{m,k}$ = Emission (mass amount yr^{-1}) of emission specie m and crop residue type k biomass;
M_k = Amount of biomass of crop residue type k that is subjected to OB per year (mass per year);
η_k = Burning efficiency of residue of crop type k (fraction, 0–1);
$EF_{m,k}$ = Emission factor of specie m from open burning of crop residue type k biomass (with consistent unit).

The main parameter of M_k in Equation (1) was estimated using Equation (2).

$$M_k = P_k \times S_k \times D_k \times B_k \quad (2)$$

Where, P_k = Crop production for crop type k (mass per year); S_k = Specific residue-to-production ratio for crop type k; D_k = Dry matter-to-crop residue ratio (fraction, 0–1); B_k = Fraction of dry matter of crop residue type k that is subjected to open burning (0–1).

To have good estimates for M_k, representative values of these parameters are required that in turn need to be generated by well-designed local surveys.

2.2. Activity data collection

In this study, the annual M_k amounts in SEA were estimated for 8 main crop types (rice, maize, soybean, potato, sweet potato, groundnut, sugarcane, and cassava) that commonly have residues subjected to OB. Wheat straw OB was not included as wheat production was not popular in SEA, e.g. only a small amount cultivated in northern Thailand for the beverage industry (FAO Statistics, 2015). The parameters required to calculate M_k (Equation (2)) were gathered from surveys conducted for several years by the air quality research group at the Asian Institute of Technology (AIT) supplemented by relevant published information (SUMERNET, 2017; Tipayarom and Kim Oanh, 2007).

Among the parameters included in Equation (2), the annual crop production information (P_k) was the most readily available. The P_k data for Thailand and Indonesia during the period from 2010 to 2015 were obtained from the national statistical reports for each of 33 provinces of Indonesia (BPS, 2016) and 76 provinces of Thailand (OAE, 2016). For other SEA countries, the national production data for the period of 2010–2014 were taken from the Food and Agriculture Organization (FAO) database (http://www.fao.org/faostat/en/#home). The 2015 data were extracted from the Association of Southeast Asian Nation (ASEAN) Food Security Information System (AFSIS): Agricultural Commodity Outlook 2015 (http://www.afsisnc.org/publications/archive/2016). Table S1, supplementary information (SI), presents the annual average production data over the study period for the selected crops in 10 SEA countries. Brunei was reported to have less types of crops (mainly rice, cassava, and sweet potato) while most other SEA countries have all 8 included crop types. The CROB emissions from Singapore were assumed to be negligible as the country has only a small agricultural production, and also due to the fact that open burning is strictly prohibited in the country (Environmental Protection and Management Act, Cap. 94A). As seen from Table S1, rice production dominated the annual average crop production in SEA during the period (204 Tg yr^{-1}) followed by sugarcane (162 Tg yr^{-1}), cassava (65 Tg yr^{-1}), and maize (38 Tg yr^{-1}). The major rice producers are Indonesia (70 Tg yr^{-1}) followed by Vietnam (43 Tg yr^{-1}), Thailand (31 Tg yr^{-1}) and Myanmar (28 Tg yr^{-1}). Except for the Philippines, SEA countries generally have rice production rates following the country population that primarily reflects the domestic consumption needs. The SEA total annual average production of 8 crops amounted at 486 Tg yr^{-1} and was mainly contributed by Indonesia (148 Tg yr^{-1}), Thailand (134 Tg yr^{-1}), Vietnam (77 Tg yr^{-1}) and the Philippines (55 Tg yr^{-1}).

It was more challenging to obtain country representative values for other parameters required for the calculation of M_k. These included the residue-to-production ratio (S_k), the dry matter to crop residue ratio (D_k), and the fraction of dry matter residue of different crops subjected to open burning (B_k). The latter is in fact the largest source of uncertainty in the M_k calculation because this B_k value depends mainly on the local specific agricultural practices and the harvesting time (in wet or dry periods) in a country. In some countries, such as Vietnam, the OB of rice crop residue happens both on-site in paddy field and off-site in villages. Thus, both should be included in the estimation of B_k. A wide range of values for this B_k parameter has been found in the global/regional EI reports. IPCC (2006) suggested a value of 0.25 to be used for developing countries and < 0.10 for developed countries, while Streets et al. (2003) suggested a value of 0.25 for South Asian countries and 0.17 for the remaining countries in the region. Information on B_k, specifically for RS, available from local surveys in some countries is included in Table 1 showing significantly higher values (0.17–0.90) than those suggested in the international databases. In addition, the B_k values relevant for SEA vary with crop cycles, i.e. higher in the dry and lower in the wet season (Table 1).

In our study, significant efforts were made to select the appropriate ranges to be used for the emission calculations (Tables S2 and SI). For example, for RS in Thailand, the B_k values were different for different parts of the country and strongly varied with season. In the central part of Thailand and for crops harvested in the dry season, we used the B_k value of 0.9 obtained from the AIT survey results by Tipayarom and Kim Oanh (2007) and SUMERNET (2017), the Sustainable Mekong Research Network rice straw co-benefits project (unpublished data as of 2018), while for crops harvested in the wet season, B_k value is typically 0.25 (SUMERNET, 2017). Our surveys (SUMERNET, 2017) were conducted in selected important agricultural areas in Thailand, Vietnam and Cambodia through a face-to-face interview with farmers. For other countries, the sources of information are presented in the footnote of Table 1.

Table 1
Values of B_k for rice straw in SEA countries compiled from available survey results.

Country	Region	Compiled B_k values	
		Dry season	Wet season
Thailand	Central	0.90[a]	0.25[c]
	South	0.30[b]	0.25[c]
	Others	0.48[a]	0.25[c]
Vietnam	North	0.77[d]	0.54[f]
	Central	0.59[e]	0.54[f]
	South	0.70[f]	0.54[f]
Indonesia	Java Island	0.43[g]	0.31[i]
	Others	0.75[h]	0.50[i]
Philippines	Whole country	0.71[j]	0.57[j]
Cambodia	Whole country	0.17[k]	0.17[k]
Malaysia	Whole country	0.43[l]	0.31[l]
Myanmar[m]	Whole country	0.77	0.54
Lao PDR[m]	Whole country	0.17	0.17
East Timor[m]	Whole country	0.43	0.31
Brunei[m]	Whole country	0.43	0.31
SEA range and most probable value (in brackets)		0.17–0.9 (0.51)	

Remarks:
[a] Tipayarom and Kim Oanh (2007) and DEDE (2003).
[b] Cheewaphongphan and Savitri Garivait (2013).
[c] SUMERNET (2017), unpublished data.
[d] Dong (2013), local survey data in Northern Vietnam.
[e] SUMERNET (2017), unpublished survey data in central Vietnam.
[f] Tran et al. (2014), survey in Mekong Delta provinces of Vietnam.
[g] Sasongko et al. (2004), survey for rice straw in Java, Indonesia (used also for East Timor).
[h] Makarim and Sumanto (2007), survey in Sumatera, Kalimantan and Sulawesi, Indonesia (also used for Malaysia and Brunei).
[i] Hafidawati, personal communication regarding the survey results conducted in West Java during rainy season in 2017.
[j] Launio et al. (2013), surveys in the several areas in Philippines.
[k] SUMERNET (2017), unpublished survey data in Prey Veng Province, Cambodia.
[l] Rosmiza et al. (2014), estimated using the rice straw data in Kedah, Malaysia.
[m] No country specific data available hence data from neighboring countries were used.

Table S2 presents a summary of the values of the parameters used in the M_k calculation. The country specific values of S_k and D_k, mainly available for rice, maize, sugarcane, and potatoes, are those reported by the local surveys extracted from relevant studies in Thailand and other SEA countries. For other crop types, we relied on the values for Asian countries compiled by Koopmans and Koppejan (1998) and Yevich and Logan (2003). For the countries in SEA where no survey data were available, we considered the similarity in the cultivated crop variants and geographical location to assign appropriate S_k and D_k. For example, the values obtained for Indonesia were also used for Malaysia, Brunei, and East Timor while those obtained for Thailand were also used for other countries in the Mainland Southeast Asia. The information on the combustion efficiency of crop residues (η_k), i.e. the fraction oxidized per total amount of crop residue biomass subjected to open burning (biomass loading), was not readily available at the country level. For RS and maize residue, results of the field measurements conducted in Thailand by Kim Oanh et al. (2011) and Athiwat (2016), respectively, were used for all SEA countries. For other crop types, the relevant values reported in the international data sources were used following the same approach applied in previous studies (Permadi and Kim Oanh, 2013; Kanabkaew and Kim Oanh, 2011).

Table S2 provides the range and the most probable values of the parameters used for the M_k calculation. The "best estimates" of the inventory species were produced using the most probable values given in the brackets (if more than one value is presented) while the lower and upper values given in the range were used to calculate the low and high emissions estimates, respectively, as further detailed in Section 2.4. Note that, due to lack of information, we did not include the inter-annual variations for S_k, D_k, η_k and B_k during the study period. The most significant change with time is expected for B_k because it strongly depends on the local practice and, in principle, on the regulation enforcement, e.g. banning CROB. However, during the reported period, there were no significant regulations and/or technological intervention to substantially reduce the CROB activity in the region. There was no main infrastructure development that could induce a fast change in the agricultural waste management practice in SEA. For example, there was no drastic development to enhance off-site uses of rice straw and other crop residues to cut down the B_k values.

2.3. Emission factors

The available EFs were scrutinized to select the most relevant value for a specific crop for the calculation of the best emission estimates in a country. Accordingly, EFs produced by measurement in the country were used for the best estimates. If not available, corresponding values reported for other Asian countries were applied. The compilation of EFs for different species and crop types used in this study is given in Table S3, SI (data sources are included in Text box S1, SI). The upper and higher values in the EF range given in Table S3 were used to calculate the low and high emission estimate, respectively. When the EF data were not available, we used the corresponding values reported for combined crops provided in Andreae and Merlet (2001) that are specified under the column "others" in Table S3.

Kim Oanh et al. (2011) reported that depending on the harvesting method, RS field burning can be practiced as "spread burning" or "pile burning" and these two practices produce significantly different EFs. Therefore, information on local harvesting methods i.e. by traditional (manual) or mechanical (combine harvester), was gathered, so that the most appropriate EFs could be applied. In Thailand and Southern Vietnam, the mechanical harvesting was common. In northern Vietnam, manual harvesting was still prevalent, mainly due to the small size of paddy field. Likewise, in Indonesia, mechanical harvesting was common on the Java Island (6 provinces) whereas manual harvesting was practiced in smaller islands. In Cambodia, mechanical harvesting was also most common, i.e. > 90% for rice crops (SUMERNET, 2017). In Malaysia and Brunei, we assumed only mechanical harvesting method while for the Philippines, manual harvesting was considered for the hilly areas and mechanical for the rest (Javier, 2009). For other countries (Myanmar, Lao PDR and East Timor), it was assumed that manual harvesting was still common. The EFs were selected for the rice straw OB (RSOB) in the countries, depending on the harvesting method, i.e. the "spread burning" EFs were applied for mechanical harvesting and "pile burning" EFs were used for manual harvesting areas. Accordingly, the EFs of PM (PM_{10} and $PM_{2.5}$), trace gases (NO_2, SO_2, and CO), PAHs (16 US EPA priority compounds), OCPs (16 compounds), and GHGs (CO_2 and CH_4) obtained from the RS spread field burning measurements in Thailand (Kim Oanh et al., 2011, 2015) were used for the places with mechanical harvesting. The EFs of CO, CO_2, CH_4, NO_2, SO_2, $PM_{2.5}$ and BC for maize residue field burning also relied on the AIT field burning experiments in Thailand (Athiwat, 2016; Phitsuca, 2016). For Indonesia, the EFs were generated by RS field burning experiments in the country (Christian et al., 2003). The EFs for other crops were obtained from Asian and international studies (Textbox S1, SI) that have been compiled in the Atmospheric Brown Cloud Emission Inventory Manual (ABC EIM) (Shrestha et al., 2013).

Less EF data were available for PAHs hence the values obtained from RS open burning field experiments for 16 PAHs in both particulate and gaseous phases (Kim Oanh et al., 2011, 2015) were used for the best estimates while those from a RS burning study in China (Zhang et al., 2011a) were included in the EFs range. Jenkins et al. (1996) also provided EFs of 19 PAHs in the PM phase for several types of crop residues, and the estimated value for 16 PAHs is 80 mg kg^{-1} of RS, which is higher but still in the range with the EF for the PM PAHs used in this EI, 34 mg kg^{-1} of RS (Kim Oanh et al., 2011). For other types of crops, only limited information of EFs for PAHs, i.e. PM_{10}-bound PAHs for maize (Wiriya et al., 2015) was available for SEA. Hence, the EFs measured in China, both PM and gaseous phases (Zhang et al., 2011a) for maize, and the United States for sugarcane (Hall et al., 2012) were used. The EFs for combined agricultural residues provided by Andreae and Merlet (2001) were used for the rest of crops types. However, only a single EF value of PAHs (25 mg kg^{-1} of biomass) estimated based on the laboratory studies is provided in Andreae and Merlet (2001). This EF in fact may be low compared to the measured values, e.g. Jenkins et al. (1996) reported the EFs for corn stover of 270 mg kg^{-1} (estimated for 16 PAHs, only PM phase). Thus, the need for relevant local measurement data of EFs is further evident.

The experimentally derived EFs for dioxins were not available for RS in any SEA countries. It was thus necessary to use the value reported by Gullet and Touati (2003) for RS (0.54 ng International Toxic Equivalent, ng I-TEQ kg^{-1}), for the best emission estimate. Crop type specific EFs for dioxins measured outside SEA are also available for maize residue burning, i.e. 0.24 ng I-TEQ kg^{-1}, from a study in China (Zhang et al., 2011b) hence this value was used for our best emission estimate. Likewise, we used the dioxins EFs for sugarcane open burning of 1.6 ng I-TEQ kg^{-1} reported by Black et al. (2011). For other crop types, the aggregate EF provided in UNEP toolkit (UNEP Toolkit, 2005) for "Agricultural residue burning in the field" of 0.5 ng I-TEQ kg^{-1} was used for the best estimate.

EFs of the OCPs are available only from the RS field burning experiments in Thailand (Kim Oanh et al., 2015). The values were also used for rice straw OB in other SEA countries. The EFs for reactive chlorine compounds, including CH_3Cl, CH_2Cl_2, $CHCl_3$ and CH_3CCl_3, and particulate and inorganic chlorine for CROB were estimated from Burklin et al. (2002) using the provided emission ratios of the respective chlorinated compounds to the EFs of CO.

2.4. Range of the emission estimates

In this study, the range of the emission estimates for different species (also referred to as the emission uncertainty) was produced by incorporating the uncertainty of both activity data and of available emission factors. Determination of the "best", "low" and "high" values for the EFs and activity data followed the approach used previously in Kanabkaew and Kim Oanh (2011) and Permadi and Kim Oanh (2013). For the activity data, the ranges of the parameters used for M_k calculation and the range of burning efficiency η_k are given in Table S2 while those for the EFs are detailed in Table S3. For the crop production (P_k), this study assumed that the data taken from the various National Agencies or Ministerial Departments had an uncertainty level of 5%, while for those collected from the international agencies, e.g. the FAO statistics or other international agencies had a higher uncertainty, i.e. 10%, following the approach of IPCC (2006). Specifically, to calculate the "best estimate" for the emission of a specie, the most probable values of all parameters were used in Equations (1) and (2). The low and high emission estimates were calculated using the lower and upper values of the ranges of the activity data and EFs, respectively.

2.5. Spatial and temporal distribution

Mapping of the spatial distributions of the SEA CROB annual emissions of different species was done with the geographical information system (GIS). Crop cultivation land in SEA was based on the land cover product provided by the Moderate Resolution Imaging Spectroradiometer, available at https://lpdaac.usgs.gov/dataset_discovery/modis (Fig. S1, SI). The spatial distribution of the emissions was prepared with a grid resolution of 0.1° x 0.1° (~10 × 10 km^2) for the domain.

The monthly emissions for Indonesia, Thailand, and Vietnam were constructed using the monthly production data, estimated biomass amount subjected to open burning, and crop annual cycle for the year of 2015 following the same approach presented in the previous studies (Kanabkaew and Kim Oanh, 2011; Permadi and Kim Oanh, 2013; Dong, 2013). For other countries where no information on the monthly crop production was available, the daily MODIS MCD45A1 product (https://lpdaac.usgs.gov/dataset_discovery/modis/modis_products_table/mcd45a1) was used together with the land use/land cover map to identify agricultural fires from other fires, e.g. forest fires, and the monthly fractions of the agricultural fire hotspots out of the total annual agricultural hotspots number were used for the monthly emission segregation.

3. Results and discussion

3.1. Total residue biomass subjected to open burning

The annual average M_k of all 8 crop types in SEA during this period was 152 Tg yr^{-1} of which RS, sugarcane, maize, and "other residues" contributed 117, 15, 13, and 6.5 Tg yr^{-1}, respectively. The annual M_k values showed an increasing trend during the period of 2010–2015 with the highest amount in 2015 of 160 Tg (Fig. 1) and the lowest in 2010 of 139 Tg. Rice production in SEA increased during the period of 2010–2013 with an average growth of 5%, but dropped in 2014 and 2015, mainly caused by the reduction in the rice production in Thailand. There was a sharp increase of sugarcane production in Thailand in 2012 of almost 63% that increased the total sugarcane production in SEA. The total crop production showed an increasing trend during the period that was linearly correlated with the amount of M_k. Country-wise, Indonesia contributed most significantly to the amount of crop residue open burning annually, i.e. 36%, followed by Vietnam (21%), Myanmar (15%), Thailand (14.8%), and the Philippines (9%) while the rest "other countries" had only small shares (Fig. S2, SI).

3.2. Average annual emissions and inter-annual variations

3.2.1. Annual emissions

The annual emissions from CROB in SEA during 2010–2015 are presented in Table 2. The best estimates of the annual emissions averaged over 6-year period, from whole SEA CROB for different species in Tg yr^{-1} were 12.5 CO; 0.36 NOx; 0.03 SO_2; 1.0 NMVOC; 0.5 NH_3; 2.0 PM_{10}; 1.8 $PM_{2.5}$, 0.08 BC, 0.8 OC; 190 CO_2; 0.56 CH_4; and 0.015 N_2O. The best estimates of the annual average emission of PAHs was 32 Gg yr^{-1} while the benzo(a)pyrene (B*a*P) emission alone was 0.16 Gg yr^{-1}. The annual emission of dioxins was 94 g I-TEQ yr^{-1}, while that of the total chlorines was 292 Gg yr^{-1} and of OCPs (from RS burning alone) was 0.03 Gg yr^{-1}. The best estimates of the total SEA CROB emissions produced in this study for the base year of 2010 are compared with those presented in other global and regional databases (Table S4 and Fig. S3, SI), i.e. EDGAR (2012) for 2010, Shi and Yamaguchi (2014) as the average over 10 years (2001–2010), and GFED for 2010.

The EDGAR estimates were the highest of all 3 datasets for NOx, CO and PM (2–4 times higher than ours) and SO_2 (6 times higher than ours), as seen in Fig. S3, SI. However, our estimates were higher than EDGAR for NH_3 (1.7 times) and were similar for N_2O (Table S4, SI). One of the important reasons for the EI results difference may have been the inclusion of palm and other oil crops residue OB in the EDGAR database, but not in ours. Note that our extracted data (not shown in Table S1) showed that the annual oil crop production in SEA during 2010–2015 was 44 Tg yr^{-1} (26 Tg yr^{-1} in Indonesia and 13 Tg yr^{-1} in Malaysia alone) which in fact was relatively small compared to rice or sugarcane production (Table S1, SI). Further, the differences in the emission results between this study and other studies presented in Table S4 and S5, SI are also caused by the differences in the values of parameters used to calculate the amount of biomass burned and the EFs selected. The SEA CROB emission estimates provided by Shi and Yamaguchi (2014) produced consistently lower results than our study for all species despite their larger EI domain, i.e. including also southern China. The main factor causing lower EI results by Shi and Yamaguchi (2014) was perhaps the lower B_k values (0.10) for RS used in their study that substantially underestimated the M_k of RS for example, i.e. by 5 times on average (Table 1). The difference in the number of crops considered may also cause discrepancies in EI results, i.e. Shi and Yamaguchi (2014) focused only on 4 main crop types of wheat, rice, maize and sugarcane, as compared to our 8 crops (not including the wheat production as it was negligible in SEA) but the discrepancies may not be as substantial (as caused by B_k values) because the three top contributors (rice, maize and sugarcane) were considered in both studies. GFED had the lowest CROB emission rates of all species as compared to others presented in Table S4 and visually inFig. S3, SI and this may be due to their underestimation of M_k amount based on

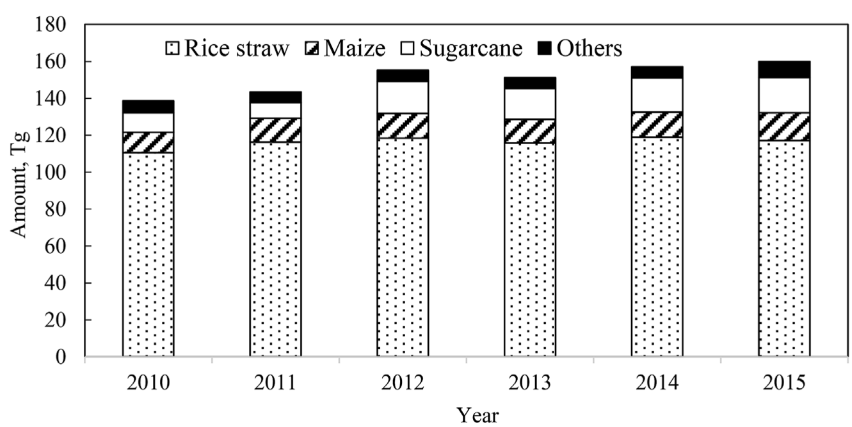

Fig. 1. SEA total amount of crop residues subjected to open burning annually by type of crop (M_k, Tg yr^{-1}).

Table 2
Annual SEA CROB emissions in the period of 2010–2015, Tg yr^{-1} (if not otherwise specified).

Species	Annual emissions						
	2010	2011	2012	2013	2014	2015	Average (CV,%)[b]
CO	11.65	12.18	12.73	12.79	12.81	12.94	12.5 ± 0.46 (3.6%)
NOx	0.33	0.34	0.37	0.37	0.38	0.38	0.36 ± 0.02 (5.6%)
SO_2	0.0279	0.0287	0.0327	0.0327	0.0333	0.0341	0.0315 ± 0.002 (7.5%)
NMVOC	0.95	1.00	1.04	1.05	1.05	1.07	1.03 ± 0.04 (3.8%)
NH_3	0.48	0.50	0.52	0.52	0.52	0.52	0.51 ± 0.02 (3.1%)
PM_{10}	1.87	1.91	1.97	1.99	2.00	2.02	1.96 ± 0.05 (2.6%)
$PM_{2.5}$	1.71	1.73	1.79	1.80	1.82	1.84	1.78 ± 0.05 (2.6%)
BC	0.074	0.076	0.084	0.084	0.085	0.087	0.082 ± 0.005 (5.8%)
OC	0.81	0.81	0.84	0.84	0.85	0.86	0.83 ± 0.02 (2.6%)
CO_2	172	178	195	196	198	202	190 ± 11 (5.8%)
CH_4	0.52	0.55	0.56	0.57	0.56	0.57	0.56 ± 0.02 (3.1%)
N_2O	0.0139	0.0144	0.0155	0.0156	0.0157	0.016	0.0152 ± 0.0008 (5.1%)
PAHs (Gg yr^{-1})	30.0	31.6	32.3	32.6	32.4	32.1	31.8 ± 0.09 (2.7%)
BaP (Gg yr^{-1})	0.143	0.159	0.166	0.167	0.168	0.184	0.164 ± 0.012 (7.6%)
Dioxin (g I-TEQ yr^{-1})	82	82	98	98	100	102	94 ± 8.1 (8.6%)
OCPs (Gg yr^{-1}), RSOB	0.025	0.0264	0.0269	0.0272	0.027	0.0266	0.027 ± 0.007 (2.6%)
Total chlorine (Gg yr^{-1})[a]	271	284	296	298	298	302	292 ± 10.6 (3.6%)

Note:
[a] Sum of five listed species in Table S3: CH_3Cl, CH_2Cl_2, $CHCl_3$, CH_3CCl_3, particulate and inorganic Cl.
[b] In brackets, CV is the coefficient of variation (SD/average), in %; OCPs are estimated for rice straw open burning only.

the satellite data of the fire hotspots. As mentioned above, agricultural fires in SEA are of small size, sporadic, and short duration and mainly taken place in the late afternoon (Tipayarom and Kim Oanh, 2007). Thus, these fires may not be adequately captured by the MODIS satellite, for example.

For the total chlorine emissions from SEA CROB, our result was 271 Gg yr^{-1} in 2010 for the 5 species. The result for 3 species of CH_3Cl, CH_2Cl_2, and $CHCl_3$ was 26 Gg yr^{-1} that is, as expected, only a small fraction of the emission result from the whole Asia domain, for the same species, reported by Lobert et al. (1999) of 307 Gg yr^{-1}. The SEA regional dioxins and PAHs emissions from CROB are generally included but not explicitly presented in the global EIs for dioxins (UNEP, 1999) and PAHs (Zhang and Tao, 2008) hence no relevant data are available for comparison. The results of RS open burning emissions for Thailand and the Philippines reported in Gadde et al. (2009) are not comparable to our results. For the Philippines specifically it was due to a much higher B_k value used in their study as discussed above. As a result, our M_k for this country of 8.6 Tg yr^{-1} for 2010 was 1.5 Tg yr^{-1} lower than the value of Gadde et al. (2009) for 2007 (Table S5). However, our EI results for PAHs were significantly higher (> 10 times), mainly because of the large difference in EFs, i.e. we used the EF of 264 mg kg^{-1} for 16 PAHs (both PM and gas phase) obtained from the field burning experiments in Thailand while a much lower EF of 18.6 mg kg^{-1} was used in Gadde et al. (2009).

Annual average SEA CROB emissions were compared with the results of the existing studies for China and India. Our emission results were generally higher by 1.1–2.8 times, varying with species, than those reported for China (Li et al., 2016) for 2012. SEA CROB emissions were also 1.3–1.5 times higher than the emissions estimated for India by Jain et al. (2014) for the year of 2008–2009. Given the larger population of China and India (above 1300 million in each country) as compared to SEA population (about 640 million) the per capita emission for SEA would be significantly higher than those for China and India. This further emphasizes the importance of the CROB emissions on the air quality in the SEA region and the urgent need for sustainable crop residue management.

3.2.2. Inter-annual variations of emissions

Annual SEA CROB emissions showed increasing trends for most species (Table 2). Coefficient of variations (CV) were computed (the ratio of the standard deviation and mean) that show a range of 2.6–8.6% depending on the species. This CV range represents relative weak inter-annual variations owning to the relatively stable crop production rates and the stable crop type composition hence less variation in the M_k values. These relatively small inter-annual variations of SEA CROB emissions suggested that the average of decadal historical emissions would still provide meaningful information for air quality management purpose, e.g. for the regional air quality modeling. However, the variations in B_k values should be incorporated, especially when major emission regulatory programs are being promulgated, e.g. banning of open burning, that lead to substantial changes in the annual Mk values.

3.2.3. Crop residue verse forest fires emissions

To roughly compare the magnitude of the SEA CROB emissions estimated in this study with forest fire emissions, forest fire data from SEA in 2010 were extracted from the GFED Version 3.1 (van der Werf et al., 2010) and presented in Table 3. The year 2010 was selected because it represents the normal climate without strong El Niño/La Nina effects. The GFED forest fire emissions included both above and below ground biomass (peat) burning. Overall, the CROB emissions (covering 8 crop types) contributed 10–43%, depending on species, of the total OB emissions (sum of CROB and forest fires) from the SEA region in 2010. The contributions from CROB were relatively more significant (> 30%) for toxic pollutants of $PM_{2.5}$, OC and NH_3 and less significant (14–21%) for GHGs and the least (10%) for SO_2 (Table 3). The relative contributions of CROB to the total biomass open burning varied widely between countries. The CROB contributed less in the countries having large forest reserve areas with frequent forest fire events, i.e. 4–31%, depending on species, in Indonesia; 16–36% in Thailand; and 8–38% in Myanmar. For other SEA countries, the contributions of CROB were higher than forest fires emissions, i.e. sharing 49–92% for Vietnam and 33–69% for the Philippines. Current concern is more on regional massive forest fires as they normally cause catastrophic haze transboundary events and are more visible on a scale that attract intensive international attention. Alternatively, CROB emissions mainly occur in populated areas and happen mostly in dry months when the air pollution levels in SEA countries are normally high. Therefore, they may cause significant local effects on health and the environment. Specifically, substantial amounts of dioxins, PAHs, and OCPs along with large quantities of toxic fine particles emitted annually from the crop residue field burning should be of concern and be the

Table 3
SEA CROB vs. forest fire emissions in 2010, in Tg yr^{-1}, in brackets are the percentage CROB contribution to the total biomass open burning.

Species	Indonesia		Vietnam		Thailand		Philippines		Myanmar		SEA region		
	CROB	Forest	CROB	Forest	CROB	Forest	CROB	Forest	CROB	Forest	CROB	Forest	Total
CO	4.5(8)	50.4	2.5(76)	0.8	0.94(29)	2.3	0.96(52)	0.90	2.3(23)	7.7	11.6(24)	37.6	49.2
NOx	0.13(15)	0.7	0.07(76)	0.022	0.029(33)	0.057	0.032(59)	0.022	0.06(24)	0.193	0.329(26)	0.93	1.26
SO_2	0.01(4)	0.223	0.0058(49)	0.006	0.003(16)	0.015	0.003(33)	0.006	0.005(8)	0.054	0.028(10)	0.263	0.291
NMVOC	0.37(15)	2.2	0.21(81)	0.05	0.07(29)	0.17	0.08(53)	0.07	0.182(26)	0.52	0.95(27)	2.6	3.5
NH_3	0.19(31)	0.42	0.103(81)	0.024	0.038(29)	0.093	0.039(57)	0.029	0.096(45)	0.118	0.48(43)	0.64	1.1
PM_{10}	0.77(17)	3.78	0.31(74)	0.11	0.09(25)	0.29	0.19(63)	0.11	0.47(33)	0.96	1.9(29)	4.7	6.5
$PM_{2.5}$	0.7(20)	2.9	0.28(78)	0.080	0.084(29)	0.21	0.17(68)	0.08	0.43(38)	0.7	1.7(33)	3.4	5.15
BC	0.028(14)	0.178	0.015(75)	0.005	0.0072(36)	0.013	0.007(59)	0.005	0.014(24)	0.045	0.074(25)	0.22	0.29
OC	0.34(19)	1.4	0.12(74)	0.040	0.031(22)	0.11	0.089(69)	0.040	0.22(37)	0.37	0.81(31)	1.8	2.6
CO_2	65.2(11)	522	36.8(72)	14	16(29)	39	16.5(52)	15	32.1(19)	135	172(21)	655	827
CH_4	0.2(31)	0.45	0.11(69)	0.05	0.038(20)	0.15	0.041(41)	0.06	0.104(18)	0.47	0.52(18)	2.3	2.8
N_2O	0.053(8)	0.063	0.003(92)	0.0018	0.0012(21)	0.0047	0.001(41)	0.0018	0.0026(13)	0.017	0.014(14)	0.082	0.096

Note: Forest – forest fire emission from GFED3.1, http://www.globalfiredata.org/data.html.

focal point of raising awareness to stop CROB activity.

3.3. Emission shares by country and by crop type

The emissions by country (Fig. 2) show that Indonesia was the dominant contributor to the total SEA CROB emissions of all species, i.e. 33–42%, followed by Vietnam (15–25%), Myanmar (11–23%), Thailand (7–21%), the Philippines (8–12%) and Cambodia (1–2%). The remaining 4 countries had relatively low emission shares, collectively of < 6%, namely 0.5–5% from Laos, 0.3–0.9% from Malaysia, and 0.06–0.2% from both East Timor and Brunei. The emission rates are explained by the country crop production rates that in turn are linked to population (domestic use), export quantity, and the amount of crop residues generated as well as residue management practices (B_k values). The high emissions in the top 4 countries (Indonesia, Vietnam, Myanmar and Thailand) were mainly due to the large crop production coupled with the high fractions of crop residues subjected to open burning. As noted earlier, this study did not cover palm oil plantation OB that is likely to be important in some countries, e.g. Indonesia and Malaysia being the world top two palm oil producers and to some extent Thailand (World Growth, 2011). Without consideration of palm oil production, the CROB emissions from Malaysia were only a small fraction in the SEA CROB emissions because the country had only a small production of the 8 included crops. Future studies should focus on the open burning emissions from various types of oil crops to provide a more complete view of CROB emissions in all SEA countries.

Contributions of different types of crops to the total CROB emissions of the considered species are presented in Fig. 3. Rice straw OB contributed dominantly to the total CROB emissions, sharing 19–97% (varying by species) followed by maize (2–78%), sugarcane (0.4–26%) while the rest of 5 crop types had only small shares (< 4.4%). Sugarcane residue open burning was high in Thailand due to the extensive plantations to provide raw material for the sugar industry in the country that is known as the world second largest sugar exporter (Sornpoon et al., 2014).

3.4. Range of the emission estimates

Figure 4 presents the best emission estimate for different species (shown as bars) for 2015 with the vertical lines indicating the high and low estimates, respectively. Note thatthe vertical lines shown in Fig. 4 do not show the standard deviation of the best estimate but the emission ranges which are linked to the relative difference between the low and the best estimates [(Low-Best)/Best], % negative) and that between the best and the high emission estimates (%, positive). Average uncertainty range of the emissions estimates for all species was −79 to +256%, with the lowest range for NMVOC (−68 to +235%) and the highest for BC (−72 to +323%). A wide range of EFs found in the literature for BC from CROB contributed to this large range of the emission estimates.

3.5. Spatial and temporal distributions of SEA CROB emissions

Using the MODIS land cover product for SEA (Fig. S1, SI), the spatial distributions of the annual emissions for year 2015 are mapped using GIS technique for all considered species at 0.1° x 0.1° (about

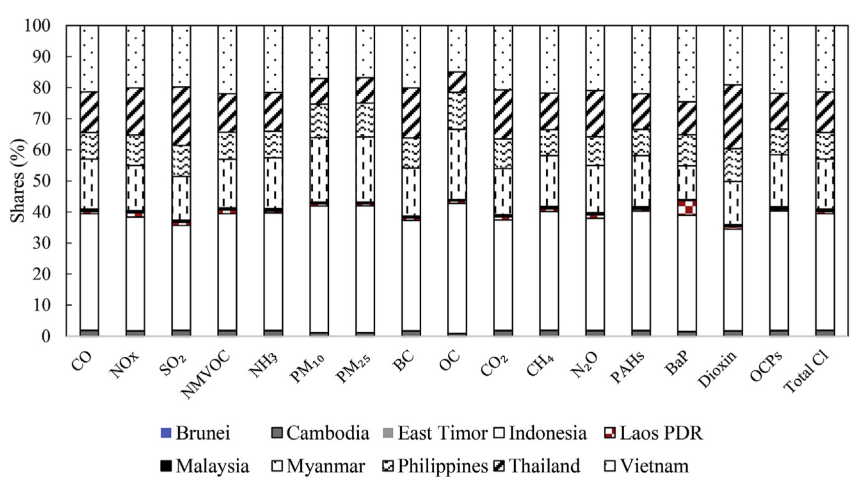

Fig. 2. Shares by country in the annual SEA CROB emissions of different species, averaged over 2010–2015 (refer to Table 2 for the total annual emissions).

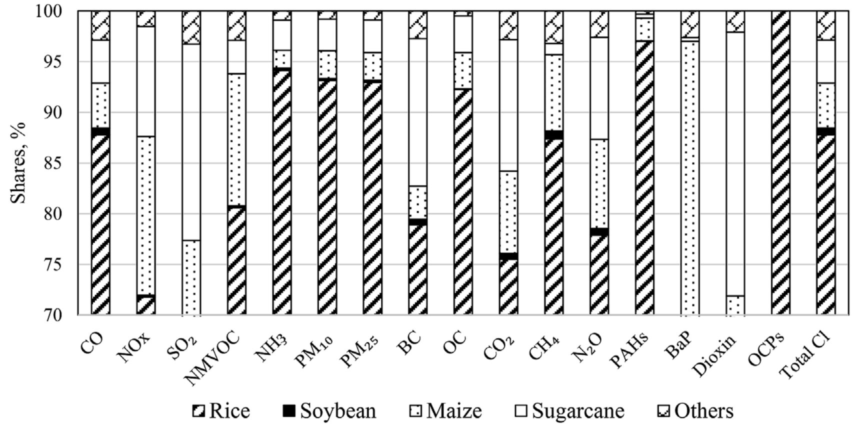

Fig. 3. Total annual emissions of different species and shares by crop types, averaged over 2010–2015 (refer to Table 2 for the total annual emissions). Note: OCPs were estimated only for RSOB.

$10 \times 10\,km^2$) resolution. The gridded emission maps for $PM_{2.5}$, BC, PAHs and dioxins are presented in Fig. 5, in black and white. The corresponding color figure is given in Fig. S4 while the maps for other species are presented in Fig. S5, SI. As expected, higher emissions are shown over the agricultural land areas. For example, in Indonesia, a higher emission intensity was seen over western and eastern Java that are known as the rice production hubs in the country and over the Sumatra Island where maize and paddy plantations are concentrated. In Thailand, emissions were higher in the central (rice) and northeastern regions (rice and sugarcane) (Chetthamrongchai et al., 2001). In Vietnam, higher emissions are seen in the Mekong River delta, Red River delta and the central coastal region, where agricultural crop production, especially rice, is intensive.

Monthly emission profiles are presented in Fig. 6 for different SEA countries. The monthly emission variations were constructed using information on the harvesting periods of different types of crops (related to *Bk* values in dry or wet season) in a year. In Indonesia, higher emissions were during August–October period which is in the dry season and follow the major rice harvesting time (August–September). For Thailand, the major crop residue burning emissions occurred also during the dry season (October–April) with peaks in November–December coinciding with the harvesting period of the rice crop. In Vietnam, emissions are clearly shown in three periods following the harvesting of 3 rice crops with the highest peaks in February–March followed by May–June and August–October. Higher emissions in February–March were mainly the effects of the dry season harvesting time in the central and southern Vietnam, while May–June peaks were mostly caused by the harvesting in the northern Vietnam. In Myanmar, similar to Thailand, the emissions peaked during the period of November–December coinciding with the rice crop harvesting time. The peak emission was found during August–October in Malaysia while that in the Philippines was during July–September. The total temporal variations were determined by the variations of different crop types, however due to its dominance in the emissions the rice straw open burning activity also dominated the monthly profiles of the SEA CROB emissions. As a way of example, the monthly BC emissions over SEA is given in Fig. S6, SI which closely follow the monthly emission fractions of the rice straw OB (Fig. S7, SI).

A good correlation between ambient air pollution levels and the CROB activities in SEA was reported in previous studies. Tipayarom and Kim Oanh (2007) showed higher levels of CO and PM_{10} during the RS burning period in Pathumthani, a sub-urban area of Bangkok, Thailand, and the linear regression between the monthly pollutant levels and the monthly MODIS hotspot counts over the area had R^2 of 0.56 for CO and 0.77 for PM_{10}. Klinmalee (2008) reported significantly higher levels of PAHs (in both PM and gas phases) in the RSOB area of Pathumthani during the dry season (400 $ng\,m^{-3}$ for 16 PAHs excluding naphthalene) as compared to the levels measured during non-RSOB period of 10–40 $ng\,m^{-3}$ while the levels found in a national park in both seasons were only 1–2 $ng\,m^{-3}$. The air quality measurements thus confirmed the seasonal effects of CROB emissions especially in the areas of intensive CROB.

The gridded SEA CROB emissions with the monthly distributions can be further used in the modeling studies to simulate the base case and various emission reduction scenarios, e.g. implementation of non-burning alternatives and/or a ban on CROB, to assess the impact on air quality and regional climate.

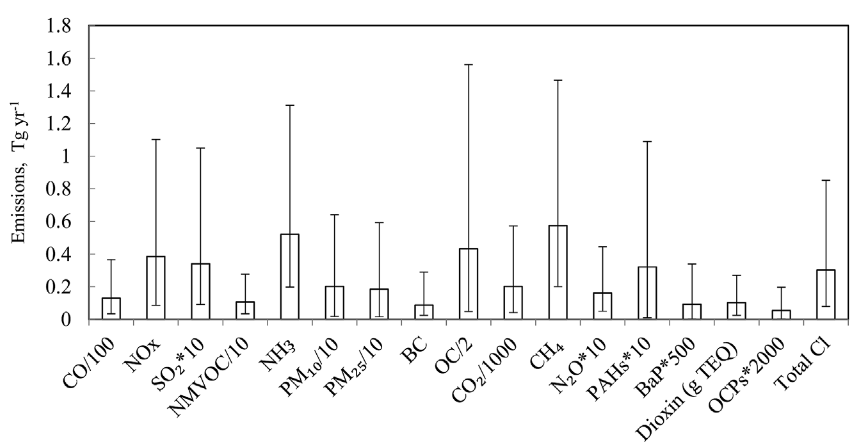

Fig. 4. SEA CROB emissions during 2015 showing with low, best and high emission estimates of different species.

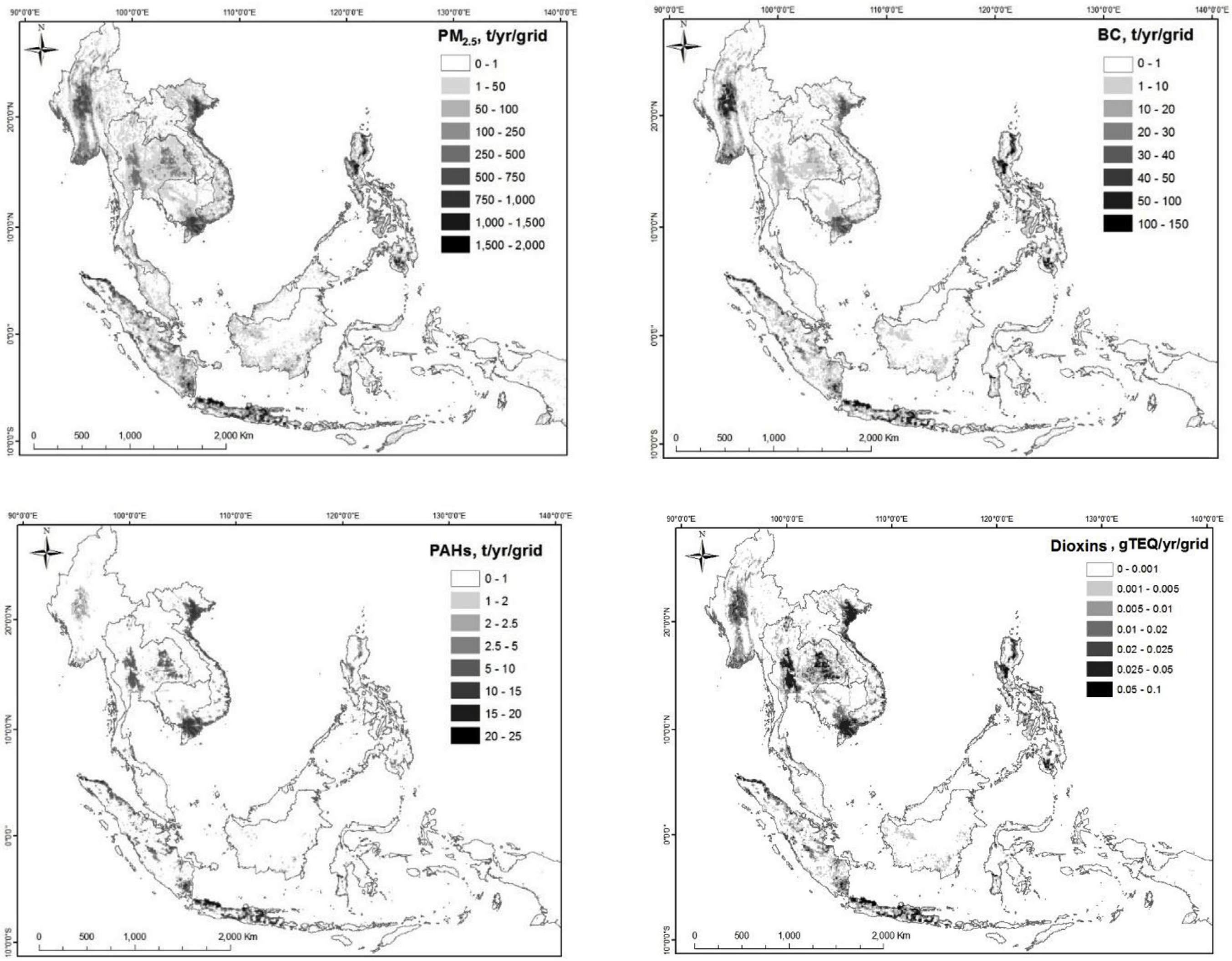

Fig. 5. Gridded SEA CROB emissions in t yr^{-1} grid^{-1}, if not otherwise specified, with grid resolution of 0.1° x 0.1°, in 2015 for selected species (colorful version is in Fig. S4, SI).

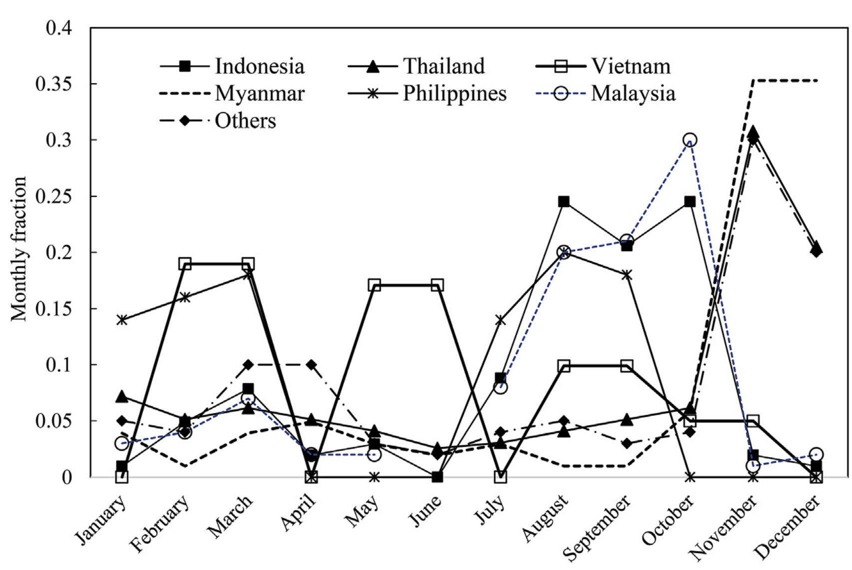

Fig. 6. Monthly fractions of CROB emissions from different countries in SEA, 2015. Note, this figure was constructed based on monthly crop production and harvesting time compiled for the countries. The sum of the monthly fractions over the year (January–December) is 1.0 for each country.

4. Conclusions

The annual average quantity of crop residue biomass subjected to open burning in SEA during 2010–2015 was 152 Tg yr^{-1}. An increasing trend in the annual emissions was observed during the 2010–2015 period but the inter-annual variations were relatively small, 2.6%–8.6% varying with species. The best estimates of annual average SEA CROB emissions over the period in Tg were: 12.5 CO; 0.36 NOx; 0.03 SO_2; 1.0 NMVOC; 0.5 NH_3; 2.0 PM_{10}; 1.8 $PM_{2.5}$, 0.08 BC, 0.8 OC; 190 CO_2; 0.56 CH_4; and 0.015 N_2O. The annual average emissions of toxic pollutants, in Gg yr^{-1}, were 32 PAHs (0.16 for B*a*P alone), 292 total chlorines and 0.03 OCPs. The annual emission of dioxins was estimated at 94 g I-TEQ yr^{-1}. On average, the lower and higher emission estimates were between −79% and 256% of the respective species best estimates. BC had the highest uncertainty range due to a wide range of the available EFs reported in literature. The EI data for CROB in SEA available in literature showed significant discrepancies for some species that could be explained by the difference in the methodology (top-down or bottom up), inclusion of the local specific detail (e.g. B_k values and harvesting methods), and EF selection.

Indonesia was the top contributor to the total SEA CROB emissions (33–42%) followed by Vietnam (15–25%), Myanmar (11–23%), Thailand (7–21%), and the Philippines (8–12%). Rice straw open burning contributed dominantly to the total SEA CROB emissions (19–97%) followed by maize (2–78%), sugarcane (0.4–26%) while the rest five crops had small shares (< 4.4%). The top three contributing crops should therefore be of priority for implementation of alternative non-burning measures.

The CROB emissions contributed 10–43% of the total OB emissions (sum of CROB and forest fires) in SEA in 2010 but varied widely with country. Forest fire emissions dominated in Indonesia (69–96%), Thailand (64–84%) and Myanmar (62–92%) but CROB emissions dominated in Vietnam (49–92%) and the Philippines (33–69%). The adverse local effects of the CROB emissions on human health and the environment should not be overlooked as these emissions occur in populated areas and mainly in the dry season when the air pollution levels in SEA are significantly higher than other time of the year. The spatial distributions of the SEA CROB emissions showed higher intensity over the agricultural areas where rice and sugarcane were mainly cultivated. Monthly emissions profiles varied by country and were affected by the local agricultural practices (harvesting times for different crop types) and seasons (dry or wet).

The emission data produced in this study, with spatial and temporal distributions, could be used for air quality modeling studies to assess the effects of current and future emissions on the ambient air quality. It is suggested that multi-year average of past emissions can be used for air quality modeling input data due to relatively small inter-annual variations of CROB emissions. Future studies should also include other types of oil crops that are commonly cultivated in some countries of the region. Locally measured EFs are required to improve the SEA CROB emission inventory results. Health effects of the CROB emissions should be quantified to provide a driving force for elimination of this open burning activity and to promote non-open burning alternatives for the sustainable agricultural waste management.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2018.05.061.

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