



Full length article

# Source emission contributions to particulate matter and ozone, and their health impacts in Southeast Asia

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## ARTICLE INFO

Editor: Dr. Xavier Querol

## Keywords:

Source apportionment  
Species tagging  
Concentration response function  
Premature mortalities  
Transboundary air pollution  
WRF-CMAQ

## ABSTRACT

Southeast Asia has been experiencing severe air pollution due to its substantial local emissions and transboundary air pollution (TAP), causing significant health impacts. While literature focused on air pollution episodes in Southeast Asia, we have yet to fully understand the contributions of local emission sectors and TAP to air quality in the region annually. Herein we employed air quality modeling with the species tagging method to first assess the contributions of source sectors and locations to fine particulate matter (PM<sub>2.5</sub>) and ozone (O<sub>3</sub>) in Southeast Asia and to hence quantify the resultant health impacts. Our results show that air pollutant exposure was associated with ~ 900 thousand premature mortalities in Southeast Asia every year. Of which, 77 % and 23 % were due to local emissions and TAP in the region, respectively. ~ 87 % of the premature mortalities due to local emissions were induced by PM<sub>2.5</sub> exposure, whereas the remaining were due to O<sub>3</sub> exposure. PM<sub>2.5</sub>-related health impacts were dominated by industrial (45 %) and residential (17 %) emissions, and O<sub>3</sub>-related impacts were mainly due to biogenic (40 %) and road transport (24 %) emissions. Furthermore, the health impacts of TAP were particularly adverse in Brunei, East Timor, Singapore, Laos, and border regions.

## 1. Introduction

Southeast Asia has experienced rapid industrialization and urbanization in recent decades, stimulating economic growth and development but coming with the substantial cost of environmental burdens (Vadrevu et al., 2017). To sustain the rising population, the fossil fuel consumption has been increased for transportation, energy generation, and residential uses. This caused an increase in the emission and concentration of airborne pollutants. The associated growing need for food has led to cropland expansion, resulting in the escalation of non-fire emissions from agricultural fertilizing and fire emissions during deforestation and waste burning (Bajželj et al., 2014; Ravindra et al., 2019). Excessive anthropogenic emissions in Southeast Asia continuously deteriorated air quality, disrupting climate and ecological systems, and imposing risks to human health and well-being, particularly for vulnerable groups (Afroz et al., 2003; Marlier et al., 2013; Taghizadeh-Hesary and Taghizadeh-Hesary, 2020). Long-term observations have identified the increasing trend of surface particulate matter (PM) and ozone (O<sub>3</sub>) concentration in Southeast Asia (Gaudel et al., 2018; Hammer et al., 2020). In contrast,

the neighboring countries in East Asia, including China, Japan, and South Korea, have taken actions and capped their PM and O<sub>3</sub> concentration levels (Kurokawa and Ohara, 2020; Zheng et al., 2018). This further highlights the severity of air pollution problems in Southeast Asia. It is therefore critical to formulate comprehensive emission restriction strategies to control air pollution in Southeast Asia.

In Southeast Asia, one dominant source of airborne pollutants is biomass burning, which includes waste burning, land clearing, and deforestation in the rural regions of mainland Southeast Asia, Sumatra, and Kalimantan (Aouizerats et al., 2015; Lee et al., 2017). As being associated with most of the peak pollution episodes (Adam et al., 2021), biomass burning was estimated to contribute up to 90 % of PM species in mainland Southeast Asia, which was responsible for 40 % to 60 % of haze events in the major cities (Lee et al., 2017; Pani et al., 2019). With the rapid industrialization and urbanization process, emissions from non-biomass burning sources tended to be enhanced. Vehicular emissions were mostly generated in urban road networks, particularly in Java (Li and Chang, 2019), including nitrogen oxides (NO<sub>x</sub>), carbon oxide (CO), and volatile organic compounds (VOCs). Previous study

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<https://doi.org/10.1016/j.envint.2024.108578>

Received 20 December 2023; Received in revised form 12 March 2024; Accepted 13 March 2024

Available online 18 March 2024

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estimated that vehicular emissions comprised 40 % of summertime planetary boundary layer O<sub>3</sub> in Southeast Asia (Niemeier et al., 2006). Meanwhile, as an important international shipping hub, Southeast Asia experienced substantial emissions of NO<sub>x</sub> and sulfur dioxide (SO<sub>2</sub>) along its major shipping routes (Streets et al., 1997), converting to 26 % of ambient PM<sub>2.5</sub> concentration in Singapore (Lee et al., 2019). Biogenic VOCs emissions originating from the vast-coverage forest in Southeast Asia were estimated to be the dominant VOC sources worldwide (Sindelarova et al., 2014b), enhancing the formation of tropospheric O<sub>3</sub> and secondary PM. Emissions from different sectors and regions indicated distinctive spatiotemporal variability in Southeast Asia (Roy et al., 2023). Therefore, determining major contributors is critical for underpinning legislative actions for local air pollution prevention. Past research emphasized the emission impacts from individual sectors and regions during a short-term episode. However, the short-term investigation is inadequate for the non-biomass burning sector with constant emissions production. Furthermore, secondary pollutants with precursors from different sources are not fully evaluated in individual sector studies. There is a critical demand for a systematic assessment of long-term source contributions to airborne pollutants in Southeast Asia.

Except for local sources, airborne pollutants emitted from remote regions can also substantially impact local air quality, under certain weather conditions such as favorable prevailing wind and no precipitation. One typical TAP event is biomass burning from Indonesia, which can cause significant increases in air pollutant concentration and deposition in Singapore, Malaysia, and Brunei, drawing substantial public and governmental concerns (Forsyth, 2014). Recent studies have also revealed the long-term TAP impacts of other emission sources, including industry, shipping, and power generation. The impacts of these emission sectors were higher than the TAP impact induced from biomass burning (Chen et al., 2020; Lyons, 2016). In addition, the longer-range TAP from outside Southeast Asia has also been identified as exerting potential threats to local air quality (Du et al., 2020; Lee et al., 2019; Nguyen et al., 2022). To address the TAP issue, Southeast Asian countries proposed collaborative measures, with one example of the ASEAN Agreement on Transboundary Haze Pollution (AATHP) (Jones, 2006). Nevertheless, their effect was marginal partly due to the imprecise provision of TAP controlling goals and enforcement. The failure is most likely due to the lack of understanding of the TAP and long-range TAP impacts among Southeast Asian countries. This calls for a comprehensive study to apportion the contributions of TAP to air quality in the region not only in transboundary haze events but also during non-haze periods.

Recent studies have evaluated the contributions of different emission sectors or regions to air pollutants concentration in Southeast Asia regions. Hansen et al. (2019) investigated the source regions of Singapore haze events over a 6-year period using the NAME model. The study has found significant variations of regional contributions to respirable particulate matter (PM<sub>10</sub>) concentration under different meteorological conditions. Khan et al. (2016) incorporated short-term PM<sub>2.5</sub> samples with the positive matrix factorization method to assess the sectoral emission contributions to PM<sub>2.5</sub> concentration at a Malaysia site. Biomass burning and vehicular emissions were identified as the main sources. Li et al. (2023) adapted the CESM model to quantify the effects of precursor emissions changes on the inter-annual growth of tropospheric O<sub>3</sub> concentration in Southeast Asia. The increasing emissions in East Asia and South Asia were found closely associated with the O<sub>3</sub> increasing in Southeast Asia. Ground transport was proved to account for most of this increasing trend. In addition to local studies, some global-scale studies also derived the source apportionment information for some Southeast Asian countries (Karagulian et al., 2015; McDuffie et al., 2021). Most of studies focused on the contributions from regions and emission sectors independently. The findings elucidated the main source of air pollution either from a particular location or an emission sector. In contrast, a joint analysis of both source regions and sectors, assessing the contributions of local emission sectors, other Southeast

Asian countries, and regions outside Southeast Asia would further enlighten in formulating the prescient emission control strategies, which is however still lacking.

PM<sub>2.5</sub> and O<sub>3</sub> exposure can increase the risk of cardiovascular and respiratory diseases and underlying premature mortality (Arden et al., 2011; Turner et al., 2016). Epidemiological studies described the exposure-risk relationship by a series of concentration–response models, which assisted in quantifying the public health impacts of local emission sources and TAP. Studies in Southeast Asia devoted to estimating the premature mortalities of PM<sub>2.5</sub> exposure (Shi et al., 2018; Yin, 2023), while limited attention was paid to the public health risks of excessive O<sub>3</sub> exposure. Meanwhile, ambient PM<sub>2.5</sub> and O<sub>3</sub> concentration exhibits distinctive spatiotemporal patterns, and so expected their health effects. Estimating the source contributions to PM<sub>2.5</sub> and O<sub>3</sub> in the region, and the consequential impact on premature mortality becomes essential.

Using the systematic source apportionment method incorporating WRF-CMAQ with Integrated Source Apportionment Method (ISAM), this study aimed to attribute ambient PM<sub>2.5</sub> and O<sub>3</sub> concentration in 2018 dynamically to local emissions and TAP sources in Southeast Asian countries and to hence estimate their resultant premature mortalities due to respiratory and cardiovascular diseases. Our results presented the contributions of local emission sector and nonlocal sources to PM<sub>2.5</sub> and O<sub>3</sub> concentration in different Southeast Asian countries. The spatial and seasonal ambient PM<sub>2.5</sub> and O<sub>3</sub> concentration was assessed to elucidate the forming mechanism.

## 2. Materials and methods

WRF-CMAQ, which is a widely used integration of meteorology and chemistry transport model system, was employed to apportion the spatiotemporal dynamics of ambient PM<sub>2.5</sub> and O<sub>3</sub> concentration to different emission source sectors and regions in Southeast Asia. WRFv3.8 is the weather model simulating the land–atmosphere exchanges of energy and water. The output meteorological information was then transferred to CMAQv5.3 model to drive the chemistry transport of air pollutants. ISAM is a source apportionment extension of WRF-CMAQ. The approach was recently updated with a new apportionment scheme that can attribute O<sub>3</sub> to emission sources appropriately in different chemical regimes (Shu et al., 2023). Finally, a series of concentration–response models (CRM) were introduced to lump the effects of different source emissions on PM<sub>2.5</sub> and O<sub>3</sub> concentration. The overall research flow is described in Fig. 1.

### 2.1. Meteorology and air quality modeling

CMAQv5.3 was adapted to simulate ambient PM<sub>2.5</sub> and O<sub>3</sub> concentration in Southeast Asia (Byun and Schere, 2006). The chemical speciation and transformation were performed by CB6 (carbon bond version 6) and AERO7 (aerosol module version 7) mechanisms. Photolysis rates were calculated by the inline radiative transfer module. The boundary and initial conditions were resampled from GEOS-Chem's outputs at a 5° × 4° spatial resolution (Bey et al., 2001). A comprehensive emission inventory that derived from multiple sources was recompiled to obtain accurate source apportionment results. Anthropogenic emissions were extracted from the HTAPv3 mosaic (Crippa et al., 2022). MEGAN v2.1 was applied to estimate biogenic emissions (Guenther et al., 2006). Fire emissions containing wildfire and waste burning were formatted from the FINNV2.5 inventory (Wiedinmyer et al., 2011). Volcanic erupted SO<sub>2</sub> emissions were derived from the CAMS inventory (Granier et al., 2019).

WRFv3.8 was integrated to provide the meteorological field driving the CMAQ simulations (Skamarock et al., 2008). The physical scheme parameterization is given in Table S1 in the supporting information. The default land-use and land cover data in the WRF model was updated to improve the representation of land–atmosphere interactions. Specifically, the land-use information was replaced with the 2018 500 m

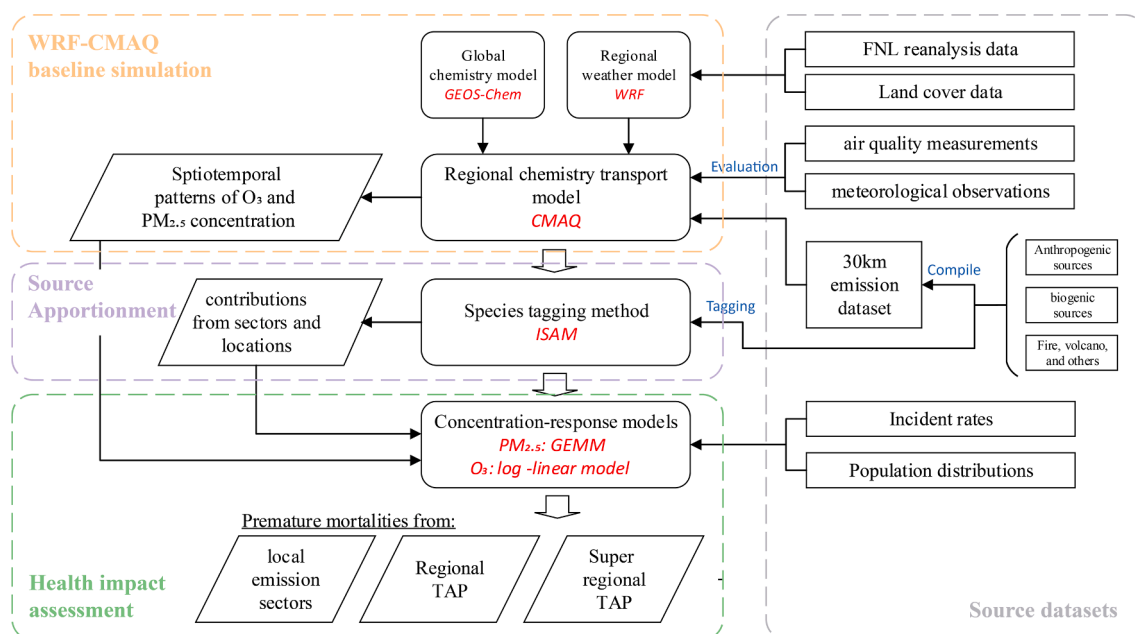


Fig. 1. Research flowchart adopted in this study.

MODIS land cover version 6 product (Sulla-Menashe and Friedl, 2018). Monthly-varied vegetation coverage data was updated by the 2018 300 m FCOVER version 1.1 product (Baret et al., 2016). Surface mosaic approach for eight categories was applied to represent the sub-grid land cover information. Initial and boundary conditions of WRF simulation were retrieved from the NCEP FNL Operational Global Analysis data at a  $1^\circ \times 1^\circ$  resolution (NCEP, 2000).

WRF-CMAQ model was configured to have a  $30 \text{ km} \times 30 \text{ km}$  domain, covering Southeast Asia and parts of South Asia, East Asia, and Oceania, as shown in Figure S1. 26 vertical layers were set in the model. To maximize computational efficiency while preserve the understanding of the annual and seasonal variation of SEA, WRF-CMAQ simulations were conducted for January, April, July, and October in 2018 to represent the air quality in the two monsoon seasons and their transitions in SEA. The simulated meteorological indicators and air pollutants concentration were compared with ground-level measurements to validate the performance of WRF-CMAQ. The meteorological observations from 120 stations in Southeast Asia were retrieved from Iowa Environmental Mesonet (<https://mesonet.agron.iastate.edu/>), including ambient temperature, wind vector, and relative humidity. In contrast,  $\text{PM}_{2.5}$  and  $\text{O}_3$  observations were less available. Measurements from 66 stations in Thailand, Indonesia, and Vietnam were collected from OpenAQ (<https://openaq.org/#/about>) platform. The model performance was evaluated by the statistic assessment of normalized mean bias (NMB), normalized mean error (NME), correlation coefficient ( $r$ ), and the index of agreement (IoA) between hourly observations and WRF-CMAQ outputs. The detailed evaluation of air pollutants concentration and meteorological indicators are described in section 1 of the results and section 2 of the supporting information, respectively.

## 2.2. Source apportionment approach

The source contributions of each sector and country/region to the concentration of  $\text{O}_3$  and PM species were simulated by a source apportionment tool, ISAM (Kwok et al., 2015, 2013). ISAM used species tagging algorithms that incorporate additional tracers to track the chemical transform, transport, and loss of focusing source species, including  $\text{O}_3$ , PM species, and their precursors. ISAM was performed in conjunction with the baseline WRF-CMAQ simulation, during which the same solver in each science processes (e.g., advection, emission,

chemical reaction, and etc.) can update multiple tracers' concentration at each CMAQ time step. Benefit from the ISAM framework design, the emission contributions of different source sectors and countries in Southeast Asia can be calculated in a single WRF-CMAQ-ISAM simulation. Meanwhile, the chemical transformation of a complex pollutant mixtures can be solved appropriately in ISAM tool, which avoids the nonlinearities of using the Brute Force method. The technical details of ISAM methods have been comprehensively presented in Kwok et al. (2013). In this study, nitrogen, sulfur, ammonia, black carbon, organic carbon, other PM, VOC, and  $\text{O}_3$  were marked as tracers. Emissions from different sectors and countries within Southeast Asia were tagged separately. Detailed information on countries and sectors is provided in Figure S1 and Table S2. The remaining emissions outside Southeast Asia were also tagged to investigate the long-range TAP (super-regional TAP) contributions to the local air quality in Southeast Asia. In addition, the contributions from initial conditions, boundary conditions, and other processes were also estimated. The boundary condition contribution was assigned to the super-regional TAP contribution, and the initial condition contributions were added up and accumulated proportionally to the sectoral contributions at the last hour of the previous day's results. Other processes included the precursors and processes not specified in the ISAM module with minor impacts on the secondary PM and  $\text{O}_3$  formation. Therefore, the value was added proportionally to the sectoral contribution.

## 2.3. Health impact assessment

Epidemiological studies have reported that ambient exposure to  $\text{PM}_{2.5}$  and  $\text{O}_3$  can trigger cardiovascular and respiratory diseases, increasing the risk of premature mortality (Arden et al., 2011; Turner et al., 2016). This study performed a health impact assessment of source contributions to premature mortalities per year using the CRM. Premature mortalities due to excessive  $\text{PM}_{2.5}$  exposure were estimated by the Global Exposure Mortality Model (GEMM) (Burnett et al., 2018), which specifies the risk to the causes of noncommunicable diseases (NCDs) and lower respiratory infections (LRI). All-cause mortalities due to  $\text{O}_3$  exposure were calculated based on the log-linear model (Turner et al., 2016). The CRM algorithm is expressed as follows:

$$E = \sum (RR - 1) / RR \times P \times f,$$

wherein P denotes the population above 25 years old; f means the baseline incident rates for the corresponding age range. The 2018 population and the age distribution in Southeast Asia were collected from the WorldPop dataset and GBD 2019 population estimates (Tatem, 2017; Vos et al., 2020). Incident rates by causes and ages were derived from GBD 2019 dataset. RR is the relative risk of various health endpoints when exposing excessive PM<sub>2.5</sub> and O<sub>3</sub> compared with baseline incidence rates, which are defined as follows:

GEMM:

$$RR_{pm2.5} = \exp\{\theta \log(C_1/\alpha + 1)/(1 + \exp\{-(C_1 - \mu)/\nu\})\},$$

$$C_1 = \begin{cases} 0 & (PM_{2.5} < 2.4\mu g/m^3), \\ PM_{2.5} - 2.4 & (PM_{2.5} > 2.4\mu g/m^3), \end{cases}$$

O<sub>3</sub>-CRM:

$$RR_{ozone} = \exp(\gamma C_2),$$

where C<sub>1</sub> and C<sub>2</sub> refer to the annual-averaged PM<sub>2.5</sub> and O<sub>3</sub> concentration at the ambient level, respectively. θ, α, μ, ν, and γ are the fitted coefficients derived from epidemiological studies (Burnett et al., 2018; Turner et al., 2016). Note that the CRM coefficients value of each pollutant specie was chosen from two-pollutant model that has adjusted the confounding effects of the other species.

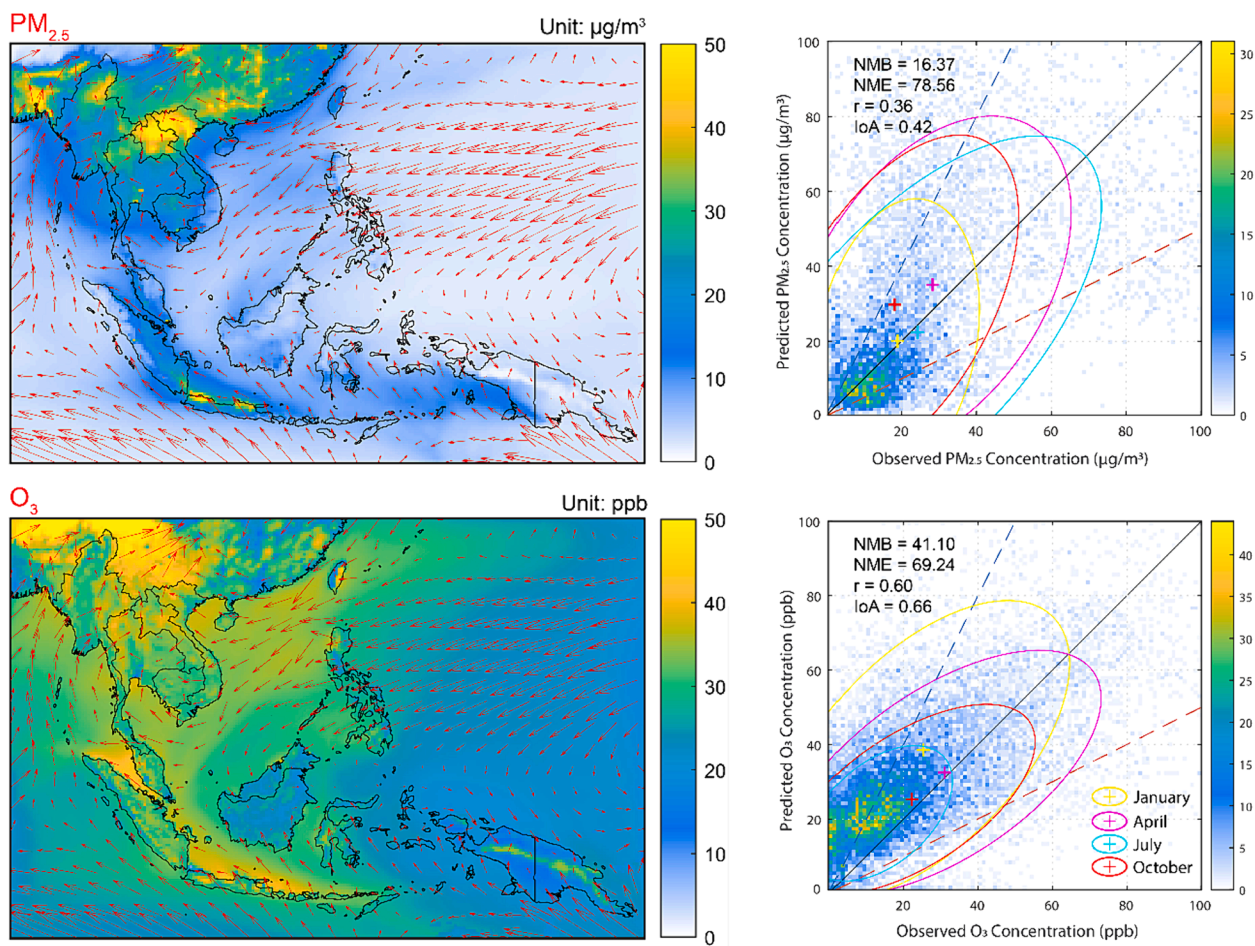
The uncertainties of WRF-CMAQ performance and CRM coefficients would be accumulated during the health impacts assessment. In this

study, the Monte-Carlo approach was employed to integrate the randomness of these variables, which was reconstructed by the median, 2.5 % CI, and 97.5 % CI values of CRM coefficients and the NMB of WRF-CMAQ simulations, assuming the triangular distributions. CRM was performed 2000 times, each randomly drawing unique CRM coefficients and NMB. The ultimate health impact estimates were collected in the form of the median value and its 95 % confidence interval of the outputs distributions of 2000 times CRM simulations.

### 3. Results

#### 3.1. Model evaluations

The validity of the simulated spatial distribution of PM<sub>2.5</sub> and O<sub>3</sub> has been proved by the model evaluation results shown in the right panel of Fig. 2. Most WRF-CMAQ estimates fell into the ±50 % range of PM<sub>2.5</sub> and O<sub>3</sub> observations, suggesting the model could accurately reproduce the gaseous and PM<sub>2.5</sub> species concentration in Southeast Asia (Amnuaylojaroen et al., 2014). Overall overestimations were identified with 16.4 % and 41.1 % NMB for PM<sub>2.5</sub> and O<sub>3</sub>, respectively, probably linking to the imprecise emission rate parameterization for O<sub>3</sub> and secondary PM<sub>2.5</sub> precursors, such as NO<sub>x</sub> and VOC. Previous study has found that most evergreen broadleaf forests in Southeast Asia have low isoprene emission capacities (Harper and Unger, 2018), while the biogenic emission model, MEGAN, did not specify the emission rates for detailed vegetation species, probably accounting for an overestimation



**Fig. 2.** The annual distributions of surface-level PM<sub>2.5</sub> and O<sub>3</sub> concentration in Southeast Asia (left panel). The comparison of ground-level measurements of hourly PM<sub>2.5</sub> and O<sub>3</sub> concentration with WRF-CMAQ predictions (right panel); Scaled colors describe overlaid evaluation samples; Colored ellipses correspond to the 95 % confidence interval of scatter points in each month with the centroid cross marking the median value. Solid and dashed lines denote the perfect and ± 50 % miss predictions, respectively. The value of statistical indicators represents the station-averaged evaluation performance.

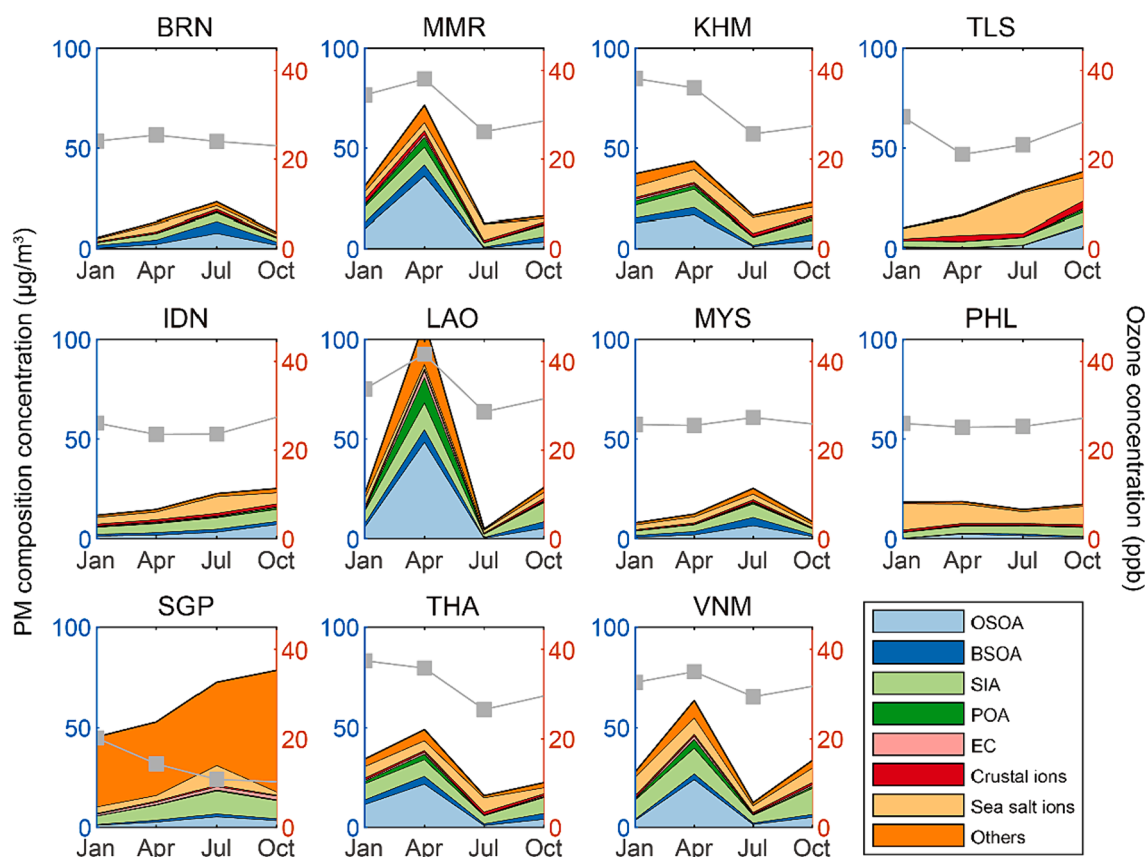
of biogenic VOC emissions and the consequential  $O_3$  concentration. Meanwhile, FINN fire emission inventory estimated higher  $PM_{2.5}$  emissions in Southeast Asia region than other fire emission inventories (Liu et al., 2020). The fire emission uncertainties may lead to the  $PM_{2.5}$  overestimation. Evaluation performance also shows a significant monthly variance. Centroids of confidence ellipses indicate that the consistent overestimations of  $PM_{2.5}$  and  $O_3$  occurred in January, April, and October. The higher estimates of VOC from biogenic emissions and  $NO_x$  from fire emissions in mainland Southeast Asia may account for this bias. In contrast, the underestimation of  $NO_x$  emission in July may account for the lower  $PM_{2.5}$  and higher  $O_3$  concentration. It is because underestimation of  $NO_x$  emissions may enhance  $O_3$  formation through weakening its titration process in the  $NO_x$ -saturated regime and may also reduce nitrate production with less available  $NO_x$ . Meanwhile, the  $[r, IoA]$  for  $PM_{2.5}$  and  $O_3$  were  $[0.36, 0.42]$  and  $[0.60, 0.66]$ , respectively, proving the WRF-CMAQ is capable of capturing the temporal fluctuation of  $PM_{2.5}$  and  $O_3$  concentration. The values were also compatible with other evaluation studies (Amnuaylojaroen et al., 2014; Nguyen et al., 2019; Xing et al., 2021).

### 3.2. Spatial distribution of $PM_{2.5}$ and $O_3$ impacts

WRF-CMAQ modeling system simulated the distribution of  $PM_{2.5}$  and  $O_3$  concentration in January, April, July, and October, which respectively represent northeast monsoon season, 1st inter-monsoon period, southwest monsoon season and 2nd inter-monsoon period. They were then averaged to represent annual mean concentration in Southeast Asia, as shown in the left panel of Fig. 2.  $PM_{2.5}$  concentration hotspots focused on the northern part of mainland Southeast Asia. Our

source apportionment results show that the high  $PM_{2.5}$  hotspots were contributed by fire emissions during the 1st inter-monsoon period. The annual mean  $PM_{2.5}$  concentration was the highest at ( $83.2 \mu\text{g}/\text{m}^3$ ) in Jakarta, which is a populous region with significant domestic anthropogenic emissions. We estimated that the local industrial production (52.8%), residential activities (23.6%), and road transportation (9.0%) were the main contributors to the  $PM_{2.5}$  in Jakarta. Similar  $PM_{2.5}$  peaks were also found in major metropolises, such as Singapore and Bangkok. Furthermore, the concentration pattern of  $PM_{2.5}$  extended to the distant region along with the prevailing wind direction, suggesting the underlying effects of TAP on air quality in the adjacent countries of Southeast Asia. Source apportionment results show that TAP contributed 14.3% to the  $PM_{2.5}$  pollution in the region. Specifically, high levels of  $PM_{2.5}$  in Java tended to be dispersed northwestward across Sumatra to Singapore and Malaysia, under the influence of the prevailing southeasterly wind.

The annual mean  $O_3$  concentration was the highest (44.1 ppb) in the suburbs of Jakarta. Similar to  $PM_{2.5}$ , the  $O_3$  over there was dominated by the road transport (31.6%), biogenic (21.0%), and energy (16.1%) emissions. However, the surface-level  $O_3$  exhibited a different spatial pattern. Severe  $O_3$  pollution was distributed over the ocean following shipping routes. Emissions from international navigation were the main contributor, accounting for an average of 52.4% of  $O_3$  in these regions. Meanwhile,  $O_3$  concentration was prominent in the northern region of mainland Southeast Asia. Our results show that biogenic and road transport emissions were the main sources, contributing 48.5% and 16.4% of the  $O_3$  in the mainland Southeast Asia on average, respectively. Besides,  $O_3$  concentration depression was observed in major metropolises, including Singapore, Bangkok, and Kuala Lumpur. The  $O_3$  depression was due to  $O_3$  titration in urban areas with substantial  $NO_x$



**Fig. 3.** Seasonal variation of  $O_3$  and  $PM_{2.5}$  composition averaged in Southeast Asian countries. The area charts depict the concentration of  $PM_{2.5}$  compositions (left y-axis). The  $O_3$  level is described in the line charts (right y-axis).  $PM_{2.5}$  compositions were categorized as biogenic secondary organic aerosol (BSOA), other secondary organic aerosols (OSOA), secondary inorganic aerosol (SIA), primary organic aerosol (POA), element carbon (EC), crustal aerosol, sea-salt ions, and unspiciated PM (others). Country codes were determined according to the ISO 3166 international standard, as shown in Figure S1.

emissions under the VOC-limited regime (Gillani and Pleim, 1996).

### 3.3. Temporal variation of PM species and O<sub>3</sub>

The seasonal variation of detailed PM<sub>2.5</sub> species and O<sub>3</sub> concentration in each Southeast Asian country was assessed separately to elucidate the formation mechanisms of air pollution (Fig. 3). The results reveal that total PM<sub>2.5</sub> concentration in mainland Southeast Asian countries, including Myanmar, Cambodia, Laos, Thailand, and Vietnam, was significantly higher (up to two times) in the northeast monsoon season and the 1st inter-monsoon period, during which other secondary organic PM<sub>2.5</sub> compounds were the major PM<sub>2.5</sub> component in mainland Southeast Asian countries, contributing to 29.2 % and 43.9 % of total PM<sub>2.5</sub> concentration, respectively, followed by the influence of secondary inorganic PM<sub>2.5</sub> and unspiciated PM<sub>2.5</sub> (others). The results suggest that the drastic rise of PM<sub>2.5</sub> composition in these two seasons was associated with the larger fire emissions. Laos experienced the highest PM<sub>2.5</sub> concentration impact, up to 109.1 µg/m<sup>3</sup> in the 1st inter-monsoon period. During southwest monsoon season, except for reduced fire emissions, strengthened wet deposition by substantial precipitation (Figure S5) also led to the PM<sub>2.5</sub> concentration decreasing in mainland Southeast Asian countries, particularly for secondary inorganic PM<sub>2.5</sub>. The Philippines and East Timor are island countries with significant sea spray emissions from the nearby waters. In the two countries, sea-salt ions (Na<sup>+</sup>, Cl<sup>-</sup>, and K<sup>+</sup>) and crustal ions (Ca<sup>2+</sup>, Mg<sup>2+</sup>) continually dominated the PM<sub>2.5</sub> components, together making up 59.5 % and 60.7 % of the total PM<sub>2.5</sub> concentration, respectively. In East Timor, sea-salt ions concentration was found to peak in the southwest monsoon season, since the stronger prevailing wind [Fig. S4 (c)] enhanced sea salt emissions from the surf zone. Similarly, higher wind speed during the northeast monsoon season [Fig. S4(a)] and 1st inter-monsoon period [Fig. S4(b)] in the Philippines also induced the higher sea-salt ions concentration. In Malaysia and Indonesia, the PM<sub>2.5</sub> concentration peak occurred in the southwest monsoon season, which was largely contributed by the fire emissions, especially the peatland fire (Othman et al., 2022). Our estimation indicates that fire emissions accounted for 31.6 % and 15.4 % of the PM<sub>2.5</sub> in Malaysia and Indonesia, respectively. Secondary inorganic PM<sub>2.5</sub> (29.1 %), sea salt ions (25.2 %), and other secondary organic PM<sub>2.5</sub> (15.4 %) were the major compositions. Singapore shared a similar seasonal variation and compositions of PM<sub>2.5</sub> with Malaysia and Indonesia, implying the potential threat from TAP from these countries. Our analysis also shows the significant TAP contributions (51.5 %) to the PM<sub>2.5</sub> concentration in Singapore. Brunei experienced severe PM<sub>2.5</sub> pollution in the southwest monsoon season, which was associated with the extensive forest coverage. The oxidation products from biogenic emissions (BSOA) comprised 22.2 % of PM<sub>2.5</sub> concentration in this country, being the second-top contributor following other secondary organic PM<sub>2.5</sub>.

In mainland Southeast Asia, O<sub>3</sub> concentration in northeast monsoon season and 1st inter-monsoon period was higher, and had a larger variation than that in southwest monsoon season and 2nd inter-monsoon period. The higher O<sub>3</sub> concentration in the first two seasons was attributed to the substantial biogenic emissions, accounting for 43.6–49.5 % of the O<sub>3</sub> in the two seasons. In contrast, O<sub>3</sub> concentration in some maritime Southeast Asian countries, including Brunei, Indonesia, Malaysia, and the Philippines, indicates only a slight temporal variation, ranging between 23.0 ppb and 27.4 ppb, which may be attributed to stable surface air temperature (Figure S5) and the corresponding chemical reaction rate for O<sub>3</sub> production. The relatively low O<sub>3</sub> concentration in these countries may be associated with the vast forest coverage with the higher O<sub>3</sub> dry deposition rate. It was noted that the temporal trend of O<sub>3</sub> and PM<sub>2.5</sub> were opposite in Singapore. In the VOC-limited environment of the metropolis, excessive NO<sub>x</sub> in the southwest monsoon season and the 2nd inter-monsoon period enhanced the secondary PM<sub>2.5</sub> concentration, and at the same time NO<sub>x</sub> emissions intensified the titration effects, causing O<sub>3</sub> sinking in this period (Gillani

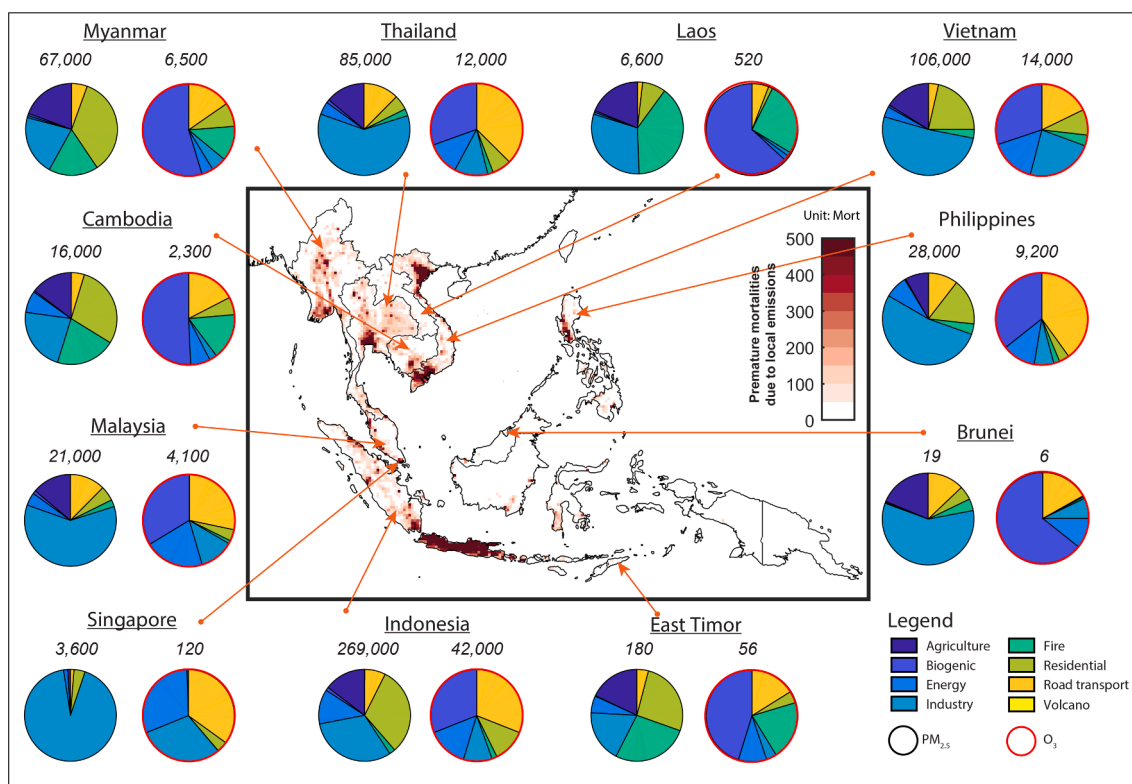
and Pleim, 1996).

### 3.4. Health impact due to local emission sectors

Our estimations show that PM<sub>2.5</sub> and O<sub>3</sub> exposures in 2018 caused 899,000 [95 % Confidence Interval (CI): 397,000–1,511,000] premature mortalities in Southeast Asia, with 77 % contributed by local emissions. Fig. 4 depicts the premature mortality distribution and the country-level apportionment to eight major local emission sectors in Southeast Asia. PM<sub>2.5</sub> exposure was the major cause of the health impacts, leading to 601,000 (95 %CI: 246,000–1,012,000) premature mortalities every year. Of these, 53.4 % occurred in maritime Southeast Asian countries, predominantly in Indonesia and followed by Malaysia, the Philippines and Singapore, whereas PM<sub>2.5</sub>-related health impacts in East Timor and Brunei were less severe. The results show that significant local emission impacts were concentrated in urban areas with substantial populations and deteriorated air quality, especially in Java and some coastal metropolises, including Kuala Lumpur, Manila, and George Town.

The industry sector with extensive emissions was one major contributor, accounting for 31.2 % to 92.4 % of PM<sub>2.5</sub>-related premature mortalities in Southeast Asian countries, except in East Timor that has limited industrial emissions. The large contribution of industry sector was due to its proximity to urban regions with a dense population that led to high population exposure to PM<sub>2.5</sub>. Besides industry sector, agriculture sector emissions, particularly ammonia emissions from widespread pasture and farmland, also made substantial contribution in Indonesia, Brunei, and East Timor (Figure S3). The agriculture sector accounted for 14.6 % (Indonesia), 18.7 % (Brunei), and 18.0 % (East Timor) of the PM<sub>2.5</sub>-related health impacts in the countries. Residential, road transport, and energy sectors exerted more pronounced health impacts in the populous countries. For example, the three sectors respectively accounted for 31.2 %, 7.3 %, and 12.2 % in Indonesia, whereas 16.2 %, 10.5 %, and 8.0 % in the Philippines. Previous research has reported the significant impacts of fire emissions in maritime Southeast Asia in transboundary haze episodes. Nevertheless, our results reveal that fire emissions contributed to less than 4 % of the annual PM<sub>2.5</sub>-related premature mortalities in maritime countries. This finding highlights the great contributions of domestic anthropogenic emissions to PM<sub>2.5</sub>-related premature mortalities in non-episode years. Other sources, including biogenic emissions and volcanoes, were less important, together contributing to less than 2 % of the PM<sub>2.5</sub>-related premature mortalities across different countries.

Over mainland Southeast Asia, PM<sub>2.5</sub> exposure due to local emissions caused 280,000 (95 %CI: 118,000–469,000) premature mortalities annually, 91.4 % of which happened in Vietnam [106,000 (95 %CI: 45,000–173,000)], Thailand [85,000 (95 %CI: 35,000–146,000)], and Myanmar [67,000 (95 %CI: 28,000–112,000)]. Cambodia and Laos, with lower population densities, experienced fewer impacts. Industry emissions asserted dominance, accounting for 21.1 % to 60.3 % of PM<sub>2.5</sub>-related premature mortalities among mainland Southeast Asian countries, in particular Thailand and Vietnam, with the rapid expansion of the manufacturing industry and the corresponding waste emissions (Figure S3). Significant impacts ranging from 13.9 % to 19.3 % were associated with the agriculture sector, revealing the substantial environmental burden of the developed agricultural industry. Vietnam and Myanmar recorded significant contributions from residential sectors for 21.5 % and 35.0 %, respectively, owing to the larger population than other Southeast Asian countries. However, even Cambodia, with its smaller population, also received a substantial contribution from its residential sector at 29.1 %. The inconsistent impacts of residential sector indicate the poor techniques of residential energy consumption with high emission rates in this country. Fire emissions were determined as significant in Laos, Cambodia, and Myanmar, accounting for 39.4 %, 21.0 %, and 17.5 % of PM<sub>2.5</sub>-related premature mortalities, respectively. The significant contributions were associated with the massive PM<sub>2.5</sub>



**Fig. 4.** Annual-averaged premature mortalities due to the local sectoral emissions in Southeast Asian countries. Spatial figure in the middle depicts the number of premature mortalities in Southeast Asia. The surrounding pie charts are the fractional contributions from eight local emission sectors to the premature mortality due to (left) PM<sub>2.5</sub> and (right) O<sub>3</sub> in each country. Values at the top of pie charts represents the total premature mortalities due to (left) PM<sub>2.5</sub> and (right) O<sub>3</sub>.

pollutants from the agricultural waste burning in the northern part of the peninsula during wet seasons. The prominent contribution of the road transport sector was identified as 12.4 % and 9.8 % of the PM<sub>2.5</sub> in Thailand and Malaysia, which are the countries known for heavy traffic volume with higher vehicular emissions (Figure S3). Other sources, including biogenic emissions and volcanoes, were less important, together contributing to PM<sub>2.5</sub>-related premature mortalities < 1 % among different countries.

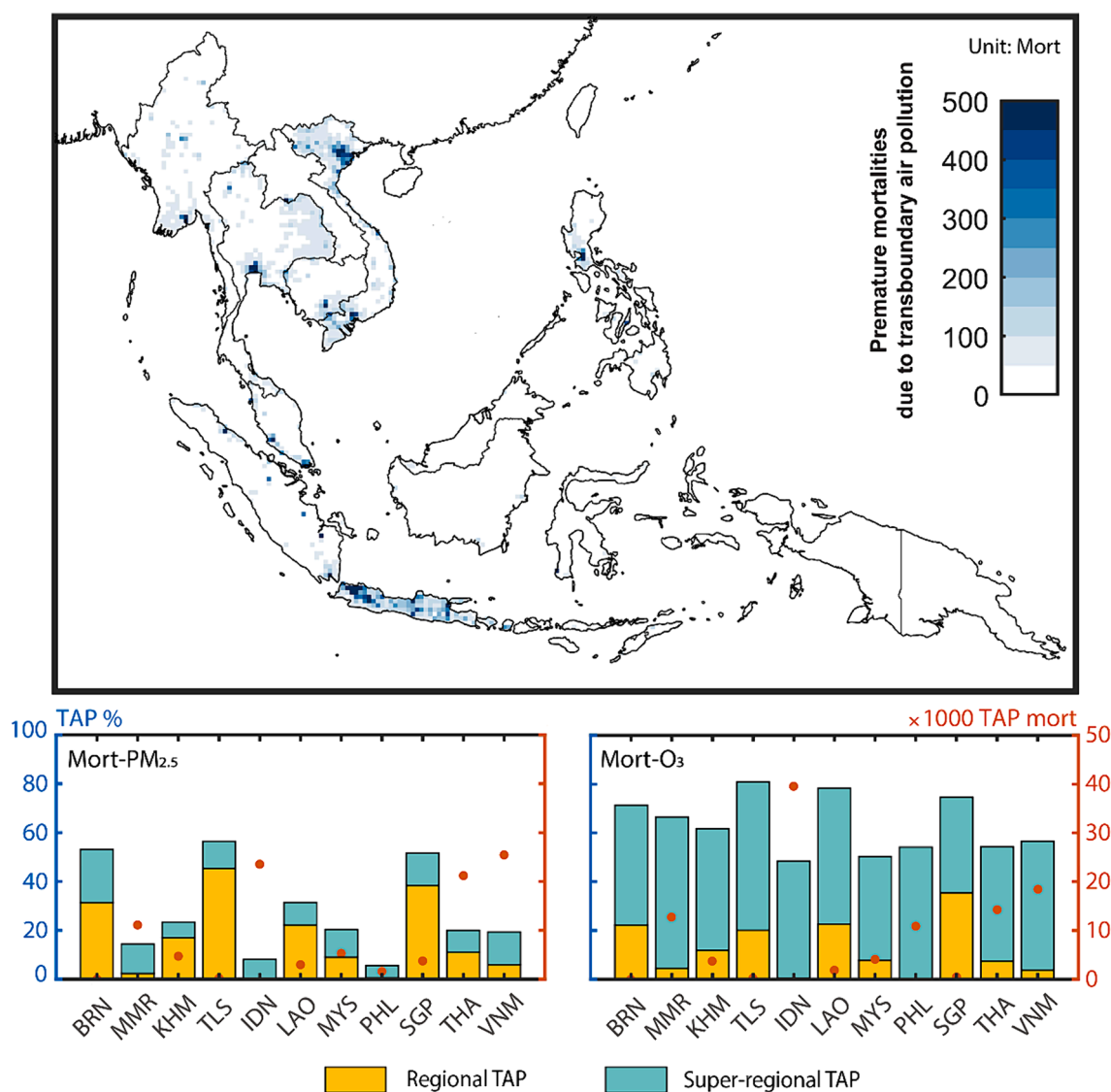
On the other hand, significant local emission impacts were also associated with the ambient O<sub>3</sub> exposure, which were estimated to induce 91,000 (95 %CI: 51,000–152,000) premature mortalities annually in Southeast Asia. Indonesia predominated the impacts for 42,000 [95 %CI: 24,000 – 70,000] mortalities, followed by Vietnam [14,000 (95 %CI: 7,900 – 24,000)], and Thailand [12,000 (95 %CI: 6,600 – 20,000)]. The overall impacts were less than those of PM<sub>2.5</sub> exposure, as the flattened risk curve of O<sub>3</sub> concentration–response relationship. Sectoral contributions to O<sub>3</sub>-related premature mortalities depend on their emissions of O<sub>3</sub> precursors, including NO<sub>x</sub> and VOCs. As the major VOC pool, biogenic sources, including forest and other nature vegetation, predominated in O<sub>3</sub>-related premature mortalities for 30.1 % to 64.4 % among Southeast Asian countries, except for Singapore with less nature forest coverage. NO<sub>x</sub> was primarily emitted from road transport, shipping, energy generation, and fire emission sectors. The road transport sector accounts for the dominance due to the combined effects of substantial emissions and its distribution with higher public exposure (Figure S3). Our estimates show that 5.9 % to 40.1 % of O<sub>3</sub>-related health impacts in Southeast Asian countries were attributed to their road transport emissions. In contrast, energy and fire emission sectors are always located in less populous regions and thus contributed less O<sub>3</sub>-related premature mortalities. Meanwhile, the emission plume of these sectors injected to the upper level, exerting less impact on the lower-level public health. Specifically, Singapore and Malaysia received comparatively larger contributions from the energy sector for 30.6 and

20.9 %, respectively, due to their high electricity demands. 20.9 %, 26.3 %, and 16.2 % of health impacts were respectively attributed to the fire emissions in East Timor, Laos, and Cambodia. Emissions from agriculture and volcanoes were not involved in the O<sub>3</sub> chemistry and thus contributed marginally to O<sub>3</sub> health impacts.

### 3.5. Public health impact due to transboundary air pollution

Premature mortalities due to transboundary PM<sub>2.5</sub> and O<sub>3</sub> exposure in Southeast Asian countries were illustrated in Fig. 5. The TAP impacts were further apportioned to the regional TAP sources and super-regional sources. The former consisted of emissions from other countries within Southeast Asia relative to the designated country, together with that from international shipping and aviation sectors, whereas the latter referred to the contributions from sources outside Southeast Asia. Our assessments show that regional and super-regional TAP together caused 206,000 (95 %CI: 100,000–347,000) premature mortalities in Southeast Asia, accounting for 23.0 % of the total health impacts. Significant TAP impacts occurred in the populated regions including Java and other urban areas with substantial air pollution exposure. Prominent TAP could also be identified along the border or coastal regions that closer to the remote emissions sources. With favorable meteorological conditions, the TAP impacts were further amplified. Significant health impacts were attributed to the TAP in surrounding Singapore, including the eastern coastline of Sumatra and southern Malaysia. Seasonal cycle of north-easterly and southwesterly wind promoted the TAP from fire and shipping emissions in this region. In Laos, fire smoke spread along the southeasterly prevailing wind to the remote regions, converting to the great contributions of regional TAP in the northwestern Vietnam and northeastern Thailand. In addition, perceivable TAP impacts were found in some outlying islands of the Philippines and Indonesia, which may be dominated by the super-regional TAP from East Asia and Oceania.

The public health impacts of PM<sub>2.5</sub> and O<sub>3</sub> exposure induced by TAP



**Fig. 5.** Annual-averaged premature mortalities due to the transboundary air pollution (TAP) in Southeast Asian countries. Spatial figure above depicts the premature mortalities due to TAP in Southeast Asia. The TAP impacts and contributions to the total premature mortalities in each country are summarized in bar (left y-axis) and scatter (right y-axis) charts below: left sub-figure is PM<sub>2.5</sub>, whereas right sub-figure is for O<sub>3</sub>.

show distinctive patterns among Southeast Asian countries. Specifically, 53.8 % of O<sub>3</sub>-related premature mortalities were attributed to TAP in Southeast Asia, which is larger than the contributions of local emissions. Super-regional TAP was particularly severe that accounted for 50.5 % of the health impacts. This was attributed to the long lifetime of O<sub>3</sub> and its precursors (e.g., PAN and CO). As the presence of descending gradients from the mid-latitude to the low-latitude, substantial transportation of O<sub>3</sub> and the precursors took place along the prevailing wind from the western Pacific and Indian Ocean to Southeast Asia region. In contrast, local emissions and regional TAP may have suppressed the domestic O<sub>3</sub> formation. In urban areas, excessive NO<sub>x</sub> from anthropogenic and fire emissions strengthened the titration effect and decreased the O<sub>3</sub> concentration under the VOC-limited regime. Excessive biogenic emissions from tropical forest also inhibited the O<sub>3</sub> production in the VOC-saturated regime, which has been observed in Southeast Asia from previous literature (Ashworth et al., 2012; Harper and Unger, 2018; Sillman, 1999). Meanwhile, most populated countries, including Indonesia, the Philippines, and Vietnam, are located at the border of Southeast Asia, which further amplified the population exposure to super-regional TAP of O<sub>3</sub>. Our estimates indicate that 47.9 %, 54.1 %, and 52.7 % of domestic O<sub>3</sub>-related premature mortalities were

attributed to super-regional TAP in Indonesia, the Philippines, and Vietnam, respectively. For countries at the center of Southeast Asia including Malaysia and Singapore, the contributions dropped to 42.5 % and 39.2 %, respectively. On the other hand, regional TAP merely made up 3.3 % of O<sub>3</sub>-related premature mortalities. The results were due to the fact that the populated countries, including Indonesia and Philippines, were at the upwind locations of major emission sources of Southeast Asia with regional TAP impacts less than 1 %. Still, regional TAP impacts in some countries could be significant when in close proximity to remote emission sources. Singapore and Brunei received 35.4 % and 22.2 % of their O<sub>3</sub>-related premature mortalities from the regional TAP, respectively, mostly associated with the shipping and fire emissions originated to Sumatra and Kalimantan. In Laos, substantial contributions (22.7 %) to the health impact were attributed to fire emissions in the neighboring countries.

PM<sub>2.5</sub>-related health impacts were largely determined by local emissions. TAP was estimated to account for 14.3 % of PM<sub>2.5</sub>-related premature mortalities in Southeast Asia. In particular, the lifetimes of PM<sub>2.5</sub> species, and their precursors (i.e., NO<sub>x</sub>, SO<sub>2</sub>, NH<sub>3</sub>, and VOC) were shorter than that of O<sub>3</sub>. The vast coverage of ocean between Southeast Asia and other regions also enhanced the deposition and weakened

PM<sub>2.5</sub> transport. Therefore, super-regional TAP only made small contributions of PM<sub>2.5</sub>-related premature mortalities in the region, ranging between 5 % and 21.7 % in different Southeast Asian countries. Conversely, regional TAP was more important in most of countries due to the short transport distance and the stationary positive contributions of transported PM<sub>2.5</sub> precursors. East Timor received the largest contributions of regional TAP, accounting for 45.4 % of PM<sub>2.5</sub>-related premature mortalities in the country. The health impact was contributed by the fire emissions from Indonesia. Comparable impacts also occurred in Singapore, accounting for 38.4 % of the PM<sub>2.5</sub>-related premature mortalities in the country. The substantial impacts were mainly attributed to the adjacent emissions from heavy shipping traffic and massive industrial sector in Johor, Malaysia. Indonesia and the Philippines were the exceptions that located at the Southeast Asian border and were hardly affected by downwind regional TAP of PM<sub>2.5</sub>. The contributions of regional TAP to domestic PM<sub>2.5</sub>-related premature mortalities in these countries were less than 1 %.

#### 4. Discussion and conclusion

Airborne PM<sub>2.5</sub> and O<sub>3</sub> pollution in Southeast Asia were estimated to cause 899,000 (95 %CI: 397,000–1,511,000) premature mortalities in 2018, 78.0 % of which were contributed by PM<sub>2.5</sub> exposure. Still, premature mortalities attributed to PM<sub>2.5</sub> and O<sub>3</sub> exposures were found to be comparable in the Philippines, East Timor, and Brunei, suggesting the need for comprehensive measures to minimize the health impacts of air pollution. Our source apportionment results show distinct sectoral contributions to ambient concentration and the resultant premature mortalities. In northern mainland Southeast Asia, PM<sub>2.5</sub> pollution was mainly fire emissions, however the PM<sub>2.5</sub>-related health impact due to was mostly from industrial and residential emissions, accounting for 45.4 % and 17.3 % of contributions, respectively. Meanwhile, shipping and fire emissions tended to dominate the O<sub>3</sub> formation in the region, however they became less important than biogenic sources (40.0 %) in terms of O<sub>3</sub>-related health impacts. Such inconsistency was due to different geographical locations of sources and thus population exposure. Since the smoke plumes of shipping and fire emissions are usually dispersed at the upper level in the rural or offshore regions, exerting less health impact than the local sources of populous districts. In particular, wildfires in Southeast Asia are short-term emission events. The wildfires intensities were highly dependent on climatic conditions, including El Niño-Southern Oscillation (ENSO) and Indian Ocean Dipole (IOD) (Fang et al., 2024; Huang et al., 2024; Zheng et al., 2023; Yim et al., 2024). Therefore, the long-term health effects of fire emissions were limited (Lee et al., 2018; Marlier et al., 2013). Our assessment highlights the significance of the domestic anthropogenic and biogenic emission sectors in Southeast Asia, which should become an obvious concern of mitigating the public health impacts of air pollution in the region.

TAP has been investigated to impose great contributions to the health impacts, accounting for 23.0 % of the premature mortalities due to PM<sub>2.5</sub> and O<sub>3</sub> exposure in Southeast Asia. The TAP effects were more significant in maritime Southeast Asian countries, especially in East Timor, Brunei, and Singapore. The TAP impacts in these countries were higher than local contributions for both PM<sub>2.5</sub> and O<sub>3</sub> exposure, dominantly originated to the industrial, shipping, and fire emissions in the surrounding regions. Meanwhile, residents living along the state borders or coastal regions were more vulnerable to the health impacts of TAP. In addition, super-regional TAP had remarkable impact on O<sub>3</sub> pollution and public health, accounting for 50.5 % of O<sub>3</sub>-related premature mortalities. In terms of premature mortalities of PM<sub>2.5</sub> exposure, the impacts of super-regional TAP were also higher than the regional TAP in Myanmar, Indonesia, the Philippines, and Vietnam. TAP has recently been recognized as a global environmental issue, especially severe for low-emission countries in Europe and North America (Zhang et al., 2017). Our analysis of TAP contribution to public health impact in Southeast Asia was about two times of the global average (12 %). These

findings underscore the significant impacts of TAP in Southeast Asia. A collaborative emission reduction framework among Southeast Asian countries, and by extension into adjacent regions, is crucial to jointly prevent air pollution and to thus reduce the resultant public health impact in the region.

This study has made substantial efforts to improve the quality of source apportionment and health impact assessment. Species tagging for PM<sub>2.5</sub> compositions and O<sub>3</sub> precursors has been adapted to avoid nonlinearities in the source attributions. During health impact assessment, advanced concentration–response models were employed with coefficients fitted based on Asian samples. Nevertheless, there are some limitations that have not yet been resolved in the uncertainty assessment. Firstly, single-year source apportionment may not fully address the interannual variability of natural emissions events, such as volcanic eruption and open field fire. This issue needs further research. Also, Some assumption made in the current tagging method may lead to uncertainties in the source attribution. For example, the tagged species concentration was updated assuming to be proportional to the bulk concentration changes during non-transport processes. The O<sub>3</sub> chemical regime in the tagging method was determined by the ratio of the instantaneous production rates of hydrogen peroxide to nitric acid (PH<sub>2</sub>O<sub>2</sub> / PHNO<sub>3</sub>) with a fixed transition value of 0.35. However, this value may vary at different sites. Last but not the least, available air quality measuring stations were mainly in Thailand, Indonesia, and Vietnam. The uncertainty assessment of model evaluations can be improved when more observations available in the future. Overall, our results provide a critical reference for future studies and policy formulation.

#### 5. Significance statement

While previous studies in Southeast Asia focused on air pollution episodes, this is the first source apportionment study to comprehensively assess the local and nonlocal sectoral contributions to long-term particulate matter and ozone concentration in the region, and to hence quantify the resultant health impact in terms of premature mortality. By apportioning the nonlinear attribution between different sources using the species tagging method, this study shows the remarkable human health impacts of emissions from industrial and residential sectors, together with biomass burning in Southeast Asia. Moreover, we found the significant health impacts of TAP even during non-haze periods.

#### CRedit authorship contribution statement

**Yefu Gu:** Writing – original draft, Visualization, Software, Methodology, Investigation, Formal analysis. **Tingting Fang:** Writing – review & editing, Formal analysis. **Steve Hung Lam Yim:** Conceptualization, Formal analysis, Resources, Writing - Review & Editing, Supervision, Project administration, Funding acquisition.

#### Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: [Steve H.L. Yim reports financial support was provided by Government of Singapore Ministry of Education. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.].

#### Data availability

Data will be made available on request.

## Acknowledgement

This research is jointly supported by the Ministry of Education, Singapore, under its MOE AcRF Tier 3 Award MOET32022-0006, the Start-up Grant (021452-00001) (LKC) and Start-up Grant (021384-00001) for Assoc. Prof. Yim (ASE), the MOE Academic Research Fund (AcRF) Tier 1 Project (award no: RT2/22) and EOS FY2022 funding (award no: EOS MOE RCE FY 2022). The authors would like to acknowledge the High Performance Computing Centre of Nanyang Technological University (NTU) Singapore, for providing the computing resources, facilities, and services that have contributed significantly to this work. This work comprises EOS contribution number 589.

## Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2024.108578>.

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