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Transboundary atmospheric pollution in Southeast Asia: current methods, limitations and future developments

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ABSTRACT

Transboundary atmospheric pollution (TAP) has emerged over the last three decades as a major challenge to countries in Southeast Asia (SEA). A limited understanding of the link between pollution emissions and their presence and effects in receiving locations (or the source-receptor relationship), in such a dynamic region covering a huge geographical area, poses a major obstacle to resolving the multitude of challenges relating to TAP in SEA. Focusing on SEA, this paper reviews the approaches used in collecting atmospheric pollution data and determining TAP. Suggestions as to how the situation may be improved are provided. Future research directions are also highlighted.

KEYWORDS

Atmospheric pollution; long-range transported pollution; proportional contributions; source appointment; source-receptor relationship

Introduction

Atmospheric pollution in a given parcel of air at a particular point is a mixture of material originating from both local and more distant sources. This is because most of the atmospheric pollutants have considerable residence times in the atmosphere (Table 1), which enable their dispersion to locations potentially tens to thousands of kilometers away from their origins (Arya, 1999; Seinfeld, Pandis, and Noone, 1998). Pollutants form part of transboundary atmospheric pollution (TAP) on crossing jurisdictional boundaries, thereby potentially impacting the environment, economy and health of a receptor country (Beckers & Rinklebe, 2017; United Nations Economic Commission for Europe, 1979). TAP can be distinguished through differentiating between pollution from local and more distant sources. However, this can be highly challenging (Fenner et al., 2005; Scheringer et al., 2006).

Southeast Asia (SEA) is experiencing rapid economic growth. In 2016, the gross domestic product of the ten Association of Southeast Asian

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Table 1. Residence time in the atmosphere of short-lived pollutants.

Pollutants	Residence time	References
Heavy metals		
Lead (Pb)	10 days	(Abadin et al., 2007)
Mercury (Hg)	6 months–2 years	(Mason & Sheu, 2002; Schroeder & Munthe, 1998; Selin et al., 2007)
Arsenic (As)	4–5 days	(Wai, Wu, Li, Jaffe, and Perry, 2016)
Cadmium (Cd)	7 days	(Pacyna, 1987)
Chromium (Cr)	7–10 days	(Hanna, Briggs, and Hosker, 1982; Santonen, 2009)
Sulfur dioxide (SO_2)	10 days	(Anger, Dessens, Xi, Barker, and Wu, 2016; Railsback, 2006)
Nitric oxides (NO_x)	1–11 hours	(Lu et al., 2015; Romer et al., 2016)
Ozone (O_3)	Hours to 23 days	(Derwent et al., 2018; IPCC, 2013)
Carbon monoxide (CO)	60–90 days	(Derwent et al., 2018; Organization for Economic Co-operation and Development (OECD), 2008)
$PM_{2.5}$	2–50 days	(Jia & Jia, 2014; OECD, 2008)
PM_{10}	2–35 days	(Lin et al., 2005; Jia & Jia, 2014; OECD, 2008)
Dust	2 weeks	(North, Pyle, and Zhang, 2014)
Secondary Organic Aerosol (SOA)	5–15 days	(Hodzic et al., 2016; Tsigaridis et al., 2014)

Nations (ASEAN) countries, namely Indonesia, Malaysia, the Philippines, Singapore, Thailand, Brunei, Cambodia, Laos, Myanmar, and Vietnam, was 200 times that of the level in 1960 (World Bank & Organization for Economic Co-operation and Development, 2017). Industrialisation, the extensification and intensification of food production and urbanization, together with levels of consumption, have grown rapidly since the 1980s (Brahmasrene & Lee, 2017; Carter, Finley, Fry, Jackson, and Willis, 2007). The global demand for natural resources, such as oil palm and pulp and paper, sourced in SEA has also risen steeply during the same period, on occasion with devastating environmental consequences (Richards & Friess, 2016). However, although generating obvious benefits, economic and demographic transitions in the region have resulted in considerable increases in atmospheric pollution (Engels et al., 2018).

Developed countries are generally better equipped to tackle pollution issues and have achieved some progress on controlling their emissions. For example, Singapore has implemented the most stringent emissions' regulations in the region to minimize its vehicle and industrial pollution (National Environment Agency of Singapore (NEAS), 2016). By comparison, less economically developed countries in the region, reliant on relatively old technologies (Siong & Euston, 2018) and with less stringent atmospheric emission standards (International Council on Clean Transportation, 2018), produce more pollution per unit of activity, some of which is transported beyond their international boundaries. A good example is land conversion methods, involving the clearance of vegetation (or biomass) through burning, applied in many SEA countries (Figure 1). The burning of biomass in this way is associated with major and highly disruptive aerosol (haze) and greenhouse gas emissions, with the effects potentially spilling out across the entire Asian

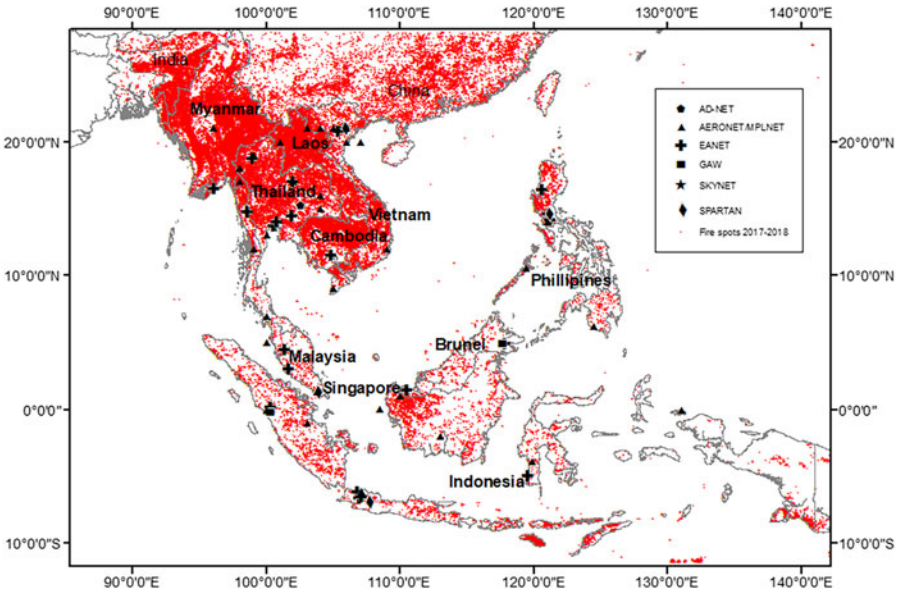
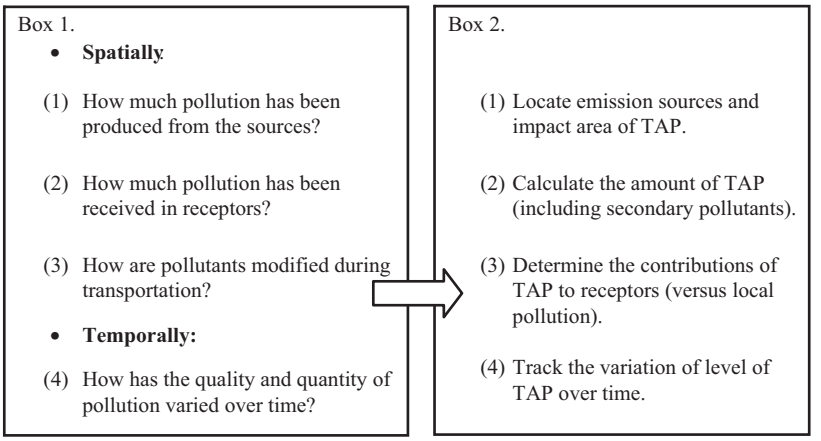


Figure 1. Ground-based remote sensing and *in-situ* sampling and measurement observation network sites in ASEAN countries and fire spots from 1 Janurary 2017 to 1 Janurary 2018 (NASA, 2018). AD-NET: Asian Dust and Aerosol Lidar Observation Networks, observes dust and other aerosols; AERONET: Aerosol Robotic Network, observes aerosol optical depth (AOD); MPLNET: Micropulse Lidar Network, observes AOD; EANET: Acid Deposition Monitoring Network in East Asia, observes deposition of SO_2, HNO_3, HCl, NH_3, NO_x, O_3, $PM_{2.5}$; GAW: Global Atmosphere Watch, observes aerosols, GHGs, reactive gases, O3; SKYNET: Observation NETWORK Dedicated for Aerosol-Cloud-Radiation Interaction Researches, observes AOD; SPARTAN: Global Particulate Matter Network, observes $PM_{2.5}$, AOD, black carbon.

monsoon region (Li, Lau, et al., 2016; Sloan, Locatelli, Wooster, and Gaveau, 2017) and possibly contributing to global climate change (Yadav et al., 2017).

The problems of TAP are widely acknowledged among the countries in SEA (Jones, 2006), but this has not led to any long-lasting, effective responses. There is one legally binding regional agreement, the 'ASEAN Agreement on Transboundary Haze Pollution' (AATHP), aimed at tackling the haze problem in SEA (ASEAN Cooperation on Environment, 2015). However, the AATHP is widely seen as a failure (Forsyth, 2014; Nurhidayah, Lipman, and Alam, 2014). Uncertainties concerning the determination of geographic sources of pollutants have contributed to the difficulties in reaching a regional consensus concerning atmospheric pollution (Hook, Mason, and O'Shea, 2015; Nobuhiko, 2013). There are many determining methods for TAP, but none are widely adopted. For example, Singapore's *Transboundary Haze Pollution Act 2014* (THPA) relies on a rather subjective judgement in defining TAP. Thus, the THPA considers TAP to be a worsening of air quality in Singapore coincident with an increase of fire hotspots in neighboring parts of the region, based on satellite-borne observations, and the occurrence of on-shore (generally



Box. (1) Key questions regarding the source–receptor relationship.
(2) Key targets regarding TAP determination.

monsoonal) winds. In reality, air pollution may be a more chronic problem in Singapore, with poor air quality not simply restricted to highly visible haze pollution events. This is because local pollutant emissions, e.g. from vehicles (Zhang, Khlystov, Norford, Tan, and Balasubramanian, 2017), may be high at times and comprise sub-micron sized particles (PM_1 and below) that are particularly hazardous to health (Karthik, Baikie, Mohan Dass, Huang, and Guet, 2017), and less visible components, such as heavy metals (Chen, Boyle, Switzer, and Gouramanis, 2016). Some of the locally generated pollutants will also be dispersed into adjacent jurisdictions. Moreover, transboundary haze may not reach Singapore even if there are numerous fire hotspots in the vicinity and winds are favorable (Aouizerats, van der Werf, Balasubramanian, and Betha, 2014).

Understanding links between pollution emissions and the nature, quantities and effects of depositions in receiving areas, or the source-receptor relationship, is fundamental to TAP determination (Venkatram & Karamchandani, 1986). Difficulties in improving this understanding, in the context of SEA, are the main focus of this paper. The source-receptor relationship is broken down into four key questions (Box 1). The ways in which these four key questions may be addressed are provided in Box 2. This paper thus provides an up-to-date review of the methods used to determine TAP, focusing on the SEA region in particular. The limitations and problems associated with the determination of TAP in a region characterized by a relatively poor coverage of monitoring stations and pollution data, together with comparatively weak transboundary environmental governance, are discussed. Finally, suggestions for future TAP research are also provided.

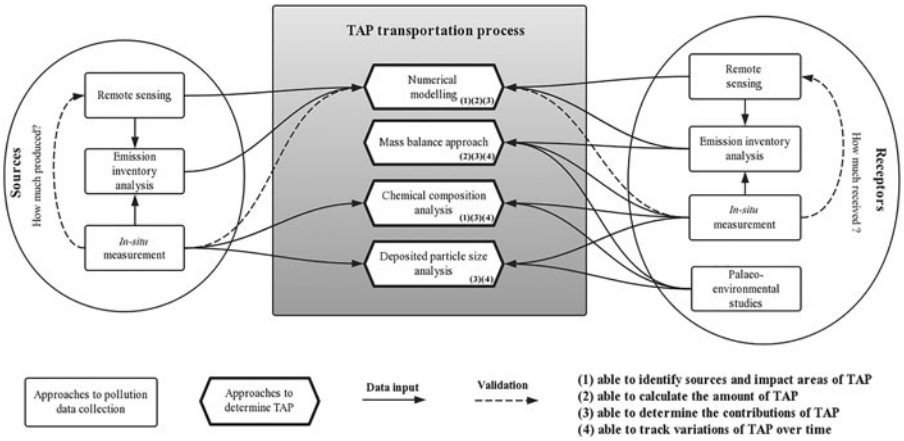


Figure 2. Methods of determining TAP according to the source-receptor conceptual framework. These methods start from techniques of pollution data collection in sources and receptors. The generated results then serve as data input to the techniques through which TAP can be determined.

Determination methods of TAP and applications in SEA

The methods used to determine TAP can be grouped into two approaches, in accordance with the source-receptor relationship concept (Figure 2). The first focuses on pollution data collection in sources and receptors, and includes *in-situ* sampling and measurement, remote sensing, emission inventory analysis, and inferences from palaeoenvironmental results. They provide information on spatial and temporal variations in pollutant loads. The second is concerned with TAP determination, and covers numerical modeling, mass balance approach, and chemical composition and particle size analyses. Together they provide the means of determining source/impact area, and the quantity, composition, contribution and variation in TAP levels. Different methods come from different combinations of the two types of approaches, each of which can solve particular questions listed in Box 1, and therefore can achieve one or more of the targets presented in Box 2. The next section discusses in detail the approaches to data collection and TAP determination, as well as method applications in determining TAP contributions in the context of SEA.

Approaches to pollution data collection

In-situ direct sampling and measurement of atmospheric pollutants

In-situ direct sampling and measurement provides a means of determining the real-time concentrations of atmospheric pollutants at sampling locations (answering questions 1 and 2 in Box 1). One part of the measurements is routinely conducted by local government agencies through

monitoring programs. Their monitoring is based on *in-situ* automated or manual sampling and chemical analysis, and the production of high quality, high frequency and fine resolution data for Criteria air pollutants. These data are often in the public domain and increasingly made available via the Internet. Unfortunately, only seven of the ten ASEAN countries routinely make available data relating to the current surface concentrations of Criteria pollutants (Table 2).

This direct sampling and measurement is also conducted through regional ground-based *in-situ* measurement networks and sea- and air-based campaigns (see Tables S1 and S2 in Supplementary Material). The main, region-wide ground-based *in-situ* measurement networks in SEA are EANET, AD-NET, GAW and SPARTAN, which routinely measure near surface concentrations of pollutants such as SO_2 and particulate matter (PM) (Figure 1). Measurements are also conducted through balloon- (Witte et al., 2017) and aircraft- (Hewitt et al., 2010) campaigns. They can provide more detailed composition profiling of pollutants of interest, such as secondary pollutants, from near surface to lower troposphere, although their coverage is limited. Campaigns are also conducted that make use of international cargo shipping and have provided information on long-term horizontal variations in pollutants across stretches of ocean in SEA (Goni et al., 2010). Because of high stability and accuracy, these abovementioned *in-situ* sampled and measured data are widely used in validations and calibrations of numerical modeling results, and space-borne remotely sensed and global emission inventory data (Hertwig et al., 2015; Nara et al., 2011; Verma, Worden, Payra, Jourdain, and Shim, 2009).

In the past two decades, portable and miniaturized monitoring devices have also been developed for bicycles, automobiles, Unmanned Aerial Vehicles (UAVs, e.g. drones) and pedestrians to collect atmospheric pollution data in more flexible ways (Elen et al., 2013; Li, Wang, Lu, Peng, and Wang, 2018). However, such techniques are not yet sufficiently matured to be widely available or acceptable (Soysal et al., 2017).

In-direct measurement through remote sensing observations

Remote sensing is a technique that can be used to infer pollutant concentrations. Remote sensing instruments generate data based on received electromagnetic radiation signals from Earth's surface (passive systems) or changes in return signals from emitted energy (active systems) (Gupta, 2017). Researchers can extract near surface and vertical concentrations of pollutants through processing the raw remote sensing data (answering question 2 in Box 1). For instance, the total-column Aerosol Optical Depth (AOD) has been widely used to infer surface concentrations of $PM_{2.5}$,

Table 2. Ground-based *in-situ* measurements and ground-based remote sensing stations in ASEAN countries by March 2018.

		Regional measurement network projects and stations (including remote sensing and *in-situ* measurements)		Local government-maintained *in-situ* measurement stations								
ASEAN Country	Area (km^2)	Number of stations	**Projects***	**Number of stations$^{\Delta}$**	$PM_{2.5}$	PM_{10}	O_3	NO	NO_2	NO_x	SO_2	CO
Thailand	513,120	21	AERONET, AD-NET, EANET, MPLNET, SKYNET	56[a]	√	√	√	√	√	√	√	√
Malaysia	330,803	7	AERONET, EANET, GAW, MPLNET	65[b]		√	√		√		√	√
Philippines	300,000	7	AERONET, EANET, MPLNET, SPARTAN	28[c]	√	√	√			√	√	√
Indonesia	1,905,000	14	AERONET, EANET, GAW, MPLNET, SPARTAN	15[d]		√						
Singapore	720	3	AERONET, MPLNET, SPARTAN	23[e]	√	√	√		√		√	√
Vietnam	331,210	11	AERONET, EANET, MPLNET, SPARTAN	2[f]	√	√		√	√			
Brunei	5,765	0		5[g]		√						
Cambodia	181,035	1	EANET	0								
Myanmar	676,578	2	AERONET, MPLNET	0								
Laos	237,955	1	EANET	0								

*Projects using remote sensing measurement: AERONET, AD-NET, GAW, MPLNET, SKYNET, SPARTAN. Projects using *in-situ* measurement: EANET, GAW, SPARTAN.
$^{\Delta}$Noted not all stations measure all the checked criteria pollutants.
[a]source: http://aqmthai.com/index.php?lang=en.
[b]source: http://apims.doe.gov.my.
[c]source: (Philippine Environmental Management Bureau, 2018).
[d]source: www.bmkg.go.id.
[e]source: http://www.haze.gov.sg/haze-updates/pollutant-concentrations.
[f]source: http://cem.gov.vn/en-US/en/Home.aspx.
[g]source: http://env.gov.bn/Theme/Home.aspx.

PM_{10}, SO_2 and NO_x using Equation 1 (Brauer et al., 2016; Donkelaar et al., 2010; He & Huang, 2018).

$$\text{Surface pollutant concentrations} = \text{AOD} \times \eta \quad (1)$$

where η is the function of factors from AOD to surface pollutant concentrations (Van Donkelaar, Martin, and Park, 2006). Vertical concentrations of pollutants can also be inferred using active sensors, such as lidar. The calculation is based on the optical energy absorption abilities of pollutants in response to different wavelengths of energy emitted by sensors (Matějíček, Engst, and Jaňour, 2006).

Remote sensing sensors can be mounted on a range of platforms and are able to provide information over a range of spatial and temporal scales and resolutions. SKYNET, AERONET and MPLNET are the main ground-based remote sensing observation networks covering SEA (Table 1 and Figure 1). The satellites and onboard sensors that monitor aerosol optical and chemical properties over SEA are listed in Table S3. Miniaturized sensors can also be carried by UAVs and even cell phones (Cao & Thompson, 2014; Villa, Salimi, Morton, Morawska, and Gonzalez, 2016), although their deployment remains relatively limited (Matese et al., 2015).

Satellite-based remote sensing data are widely used in atmospheric pollution research. However, data quality is easily affected by a number of factors, including the frequency of satellite revisits, the observation abilities of particular sensors and, especially for passive sensors, the occurrence of cloud and smoke in the area of interest (Ford & Heald, 2016; Park et al., 2014; Zhao, Chan, and Heidinger, 2013). Ground-based observations often have higher temporal resolution (e.g. hourly or sub-hourly intervals) and better observation accuracy compared with satellite-based observations (Lin et al., 2014), and are often used to calibrate satellite-sensor products (Remer et al., 2005) and outputs from numerical models (Wang et al., 2014).

Emission inventory analysis

Emission inventory analysis (EIA) can be used to quantify local emissions of pollutants from sources in a given geographical area over a particular time span (answering question 1 in Box 1). Emission inventory involves compilation of a list of pollutant types, all possible emission sources and the corresponding source activity data, such as production (European Monitoring and Evaluation Programme & European Environment Agency, 2016). The source activity data and the corresponding locational information are obtained from either statistical documents, such as national statistical yearbooks, or from satellite-based remote sensing (Simpson et al., 2016; Wooster, Roberts, Perry, and Kaufman, 2005). Thus, the quantity of

a pollutant emitted from a given area is calculated by summing the emissions from every single source in that area (Equation 2).

$$\text{Local emissions} = \sum (\text{AD} \times \text{EF}) \quad (2)$$

where AD represents source activity data and EF represents the emission factor relating emission quantity of a pollutant and the source activity.

EIA, assisted by satellite-borne observations, has been used to examine pollutant emissions, both regionally (Kurokawa et al., 2013) and globally (Klimont, Smith, and Cofala, 2013). EIA can also accommodate time periods ranging from one year to hundreds of years (Liu et al., 2016), depending upon the availability of historical sources of evidence. SEA is well-served by global inventories (see Table S4). However, EIA may not always accurately represent actual emissions, especially in SEA. For example, significant differences occur when *in-situ* measurements of emissions from biomass burning in SEA are compared with global emission inventory data (Chuang et al., 2015). Localised inventories, such as national or city level EIA, and finely resolved inventories, which provide accurate data for analyzing local environmental problems, are generally lacking in SEA.

Palaeoenvironmental study

Palaeoenvironmental studies can potentially extend the temporal extent of monitoring records of past variations in total amounts of atmospheric pollution deposited in receiving areas (contributing to answering question 4 in Box 1). Palaeolimnology, for example, can be used to retrieve information on past depositions of atmospheric pollutants in lake catchments that stretch back from tens to thousands of years, and thus to the earliest pollutant releases and their impacts (Last & Smol, 2006; Smol, 2012). Long-term variations in the deposition flux of pollutants can be determined, based on concentrations of pollutants or their proxies in lake sediments with known sedimentation rates (Smol, 2012). Similar approaches are also used to quantify variations in pollutants and their effects preserved in marine sediments (Cai et al., 2017), coral reefs (Chen, Lee, Nurhati, Switzer, and Boyle, 2015), high altitude snow (Mochizuki, Kawamura, Aoki, and Sugimoto, 2016) and ice caps (Hertzberg, 2017). Past variations in air quality have also been successfully inferred from documentary evidence, such as airport visibility records (Field, Van Der Werf, and Shen, 2009) and bird species data (DuBay & Fuldner, 2017).

Sediment is one of the most easily accessible natural archives of pollutant variations (Sanchez-Cabeza & Druffel, 2009). Sediment-based, pollution-related studies have been conducted in reservoirs, lakes, rivers and coastal areas in most of the AESEAN countries (except Myanmar, Laos and Brunei) (Table S5). Although only around 50% of the sedimentary evidence

of past variations in pollution loading is reliably dated, the evidence is sufficient to reveal long-term variations in heavy metals, Persistent Organic Pollutants (POPs), and Spheroidal Carbonaceous Particles (SCPs)[1]. Sedimentary evidence that has not been reliably dated can also be used to highlight trends in pollution levels, however. Sediment-based studies can provide information on other, relevant factors, such as reconstructions of past changes in climate, including monsoonal activity (Wündsch et al., 2014), which can be critical to interpretation of sedimentary pollution data (Inness et al., 2015).

Approaches to TAP determination

Numerical modeling

Two types of numerical modeling are used in the determination of TAP: trajectory models and Chemical Transport Models (CTMs). These examine, respectively, the movement of air parcels, and the physiochemical transformation of pollutants during transportation between sources and receptors.

Trajectory models were first developed as early as the 1940s (Stein et al., 2015). They can help indicate TAP source areas by enabling reconstruction of the transportation routes of air parcels from a particular location and for a selected time span using regional meteorological data and lagrangian functions (Tang et al., 2007). They have been used to hindcast or forecast possible pollution sources or destinations with minimum computational cost. Such trajectory models include Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) (Draxler & Rolph, 2003) and FLEXible TRAjectory model (FLEXTRA) (Stohl et al., 2010).

Advances in computing power and knowledge of atmospheric physics have facilitated development of three-dimensional CTMs. They consider not only advection process, but also diffusion, deposition of pollutants, and, most importantly, chemical reactions and radioactive decay of pollutants in the atmosphere with time (answering question 2 and 3 in Box 1). For instance, HYSPLIT-Hg and FLEXPART, which are the CTM versions of the trajectory models HYSPLIT and FLEXTRA, respectively (Draxler & Rolph, 2003; Stohl et al., 2010). CTMs utilize eulerian grid models based on a fixed coordinate system with respect to the ground or/and lagrangian puff models based on a moving frame of reference. They are driven by meteorological and pollutant emissions inventory data together with retrieval products from remote sensing systems, utilizing a continuity

[1]SCPs are a byproduct of fossil fuel burning that are widely dispersed and preserved well under most sedimentary conditions. They are thus a useful proxy of anthropogenic activities leading to TAP (Rose, 2015).

equation (Equation 3) and embedded chemical reaction database to capture changes in pollutant concentrations and formations of secondary pollutants during transport in each computing grid and time step (Daly & Zannetti, 2007).

$$\frac{\partial q}{\partial t} = -\nabla \cdot q\vec{V} + E - R \tag{3}$$

where q represents the concentration of pollutants, t represents time step, $\vec{V}$ represents the vector of wind direction, E represents pollutant supply, R represents removal function, which includes deposition and transformation. The CTMs that have been applied in SEA are listed in Table S6. However, reported modeling results do not often correspond with *in-situ* measurements (Akimoto, 2017; Wang et al., 2014). This is mainly due to the quality of input data, and the challenge of accurately simulating physical and chemical processes in the atmosphere that lead to the transformation of pollutants (Reid et al., 2009).

• *applications of numerical modeling in determining TAP contributions in SEA.* Indonesia is a major source of haze pollution in SEA, with the burning of biomass linked to plantation development and agriculture largely responsible. Reddington et al. (2014) combined a fire emission inventory database, a CTM and 5-day trajectory modeling to quantify the concentration and track the dispersal of $PM_{2.5}$ in the atmosphere over Singapore in 2004. Their results showed that fires in southern Sumatra had contributed 42% of the $PM_{2.5}$ in Singapore, with other substantial contributions from fires in central Sumatra (22%) and Indonesian Borneo (Kalimantan) (14%). The contribution from fires occurring in Peninsular Malaysia and Indochina was negligible (~2–3%). Similarly, Engling, He, Betha, and Balasubramanian, (2014) used HYSPLIT modeling to determine the main sources of haze in Singapore during biomass burning in 2006. Their results indicated that biomass fires in Indonesia contributed 75% of total suspended particles in the atmosphere above Singapore during hazy days.

Aouizerats et al. (2014) point out that local pollution may also make a substantial contribution to haze pollution. A haze event in Singapore in October 2006 was analyzed using a CTM to establish levels of local pollution at the time in Singapore. To do this, they compared overall pollution levels with overall pollution levels minus the estimated contribution from biomass fires in Indonesia. Results showed that of the 35 most heavily-polluted days, only 17 were associated with major outbreaks of burning in adjacent parts of Indonesia, while local pollution levels enhanced by stable meteorological conditions were the main reasons for poor air quality on the other 18 days. Their trajectory analysis also indicated that pollution from Kalimantan (Indonesia) had not reached Singapore during the haze

period. Lee, Bar-Or, and Wang, (2017) also emphasized the contribution of local pollution to air quality in major cities in SEA. Their results showed that biomass burning in SEA only contributed 39%, 36% and 34% of the low-visibility (<10 km) days in, respectively, Bangkok (Thailand), Kuala Lumpur (Malaysia) and Singapore from 2003 to 2014.

Mass balance approach

The mass balance approach can be used to quantify TAP and determine the total TAP contribution in a receptor area. The approach is based on the law of conservation of mass (Council, 1978), which states that in open systems the input of mass should be equal to the output and accumulation of mass (Goss & Petrucci, 2007). Pollution received at a receptor, which is an open system, comprises both TAP and local pollution. Estimating the contribution from local sources therefore provides the means to establish the level of TAP. To achieve this, a specific geographic area is defined as a receptor (e.g. an entire country, or a part of a country, such as an urban area or a lake basin and its catchment) (Paulson, Feely, Curl, Crecelius, and Romberg, 1988; Scudlark, Conko, and Church, 1994; Yang, Rose, Battarbee, and Boyle, 2002). Although the approach sounds simple, determining local pollution levels can be challenging (Heyvaert, Reuter, Slotton, and Goldman, 2000). A simpler approach involves the direct determination of TAP in receptor areas that are sufficiently far away from ambient local emission sources. Thus, the local emissions are presumed to be equal to zero and the TAP is directly represented by the total received pollution. These observation locations are normally chosen on offshore islands or inland places that are at least 100 km away from the nearest emission sources (Li et al., 2017; Vuthyrak et al, 2006). Also, the sedimentation records from remote lakes, where there are little or no local pollution inputs, can provide information on past levels of TAP (Greenwood, Mills, Vrana, Boer, and Van Bavel, 2009; Kallenborn, Hung, and Brorström-Lundén, 2015; Kuwae et al., 2013).

• *applications of mass balance approach in determining TAP contributions in SEA.* Pollutants generated by biomass fires burning on the Indochina peninsula, which comprises the SEA countries of Myanmar, Thailand, Laos, Cambodia and Vietnam, can impact air quality in Taiwan during the (Northern Hemisphere summer) south-westerly Asia monsoon. Li et al. (2017) conducted *in-situ* measurement and 3-day trajectory analysis of PM pollution in a remote offshore site in the Taiwan Strait between 2013 and 2015. Their results showed that the PM from the Indochina peninsula and Philippines account for 50% of background PM levels at the site. By comparison, Lai, Lee, and Huang, (2016) and Li, Yuan, et al. (2016) estimated the contribution of TAP to the background levels to be 24%, based on data

from the same site dated to 2007–2009. The differences between the two estimates (2013–2015 compared with 2007–2009) most likely reflect variations in either meteorological conditions or pollution emissions, or some combination of the two.

Chemical composition analysis

Changes in the chemical composition of pollutants in receptor areas before and after mixing with TAP can be used to determine levels of the latter. Chemical compositions commonly analyzed include the ratios of two isotopes (e.g., $^{206}Pb/^{207}Pb$) (Blais, 1996), ratios of two elements (e.g., Sb/Pb) (Kuwae et al., 2013) and ratios of two isomers (e.g., α/γ-Hexachlorocyclohexane) (Sanusi, Millet, Mirabel, and Wortham, 2000). Chemical compositions are unique in different geographical areas, and the method can also be used to fingerprint particular pollution sources (Inomata, Ohizumi, Take, Sato, and Nishikawa, 2016). Chemical composition of a pollutant also varies over time (Thiemens, 2006), which can be used to indicate the formation of secondary pollutants (Huang et al., 2014), and to reconstruct past variations of TAP loads when combined with palaeoenvironmental studies (Kuwae et al., 2013; Renberg, Bindler, Bradshaw, Emteryd, and McGowan, 2001). The proportional contribution of TAP is estimated by Equation 4 (Blais, 1996). Basically, the closer the sample ratio is to the source ratio, the larger the TAP contribution. However, the choice of source area largely determines the contribution (the denominator).

$$\text{TAP contribution} = \frac{\text{sample ratio} - \text{typical receptor ratio}}{\text{typical source ratio} - \text{typical receptor ratio}} \times 100\% \quad (4)$$

• *applications of chemical composition analysis in determining TAP contributions in SEA.* Chen et al. (2015) studied isotopic composition of Pb ($^{206}Pb/^{207}Pb$) records in marine coral samples from the Singapore Strait. The results revealed a decline in the local contribution to overall Pb deposition from 57% in the 1970s to 41% in the 2000s, presumably marking the effects on air quality of a tightening of restrictions on emissions in Singapore. Chen et al. (2016) used the same method to study variations in isotopic composition of Pb throughout a sediment core from a reservoir in Singapore. They concluded that Pb in the environment between the 1910s and the 1940s was from both natural and local sources, while in the 1990s the proportion of TAP increases, with rapidly industrializing and urbanizing parts of Malaysia, Thailand, Vietnam and Indonesia the most likely sources. This is consistent with the peak time of lead petroleum consumption in SEA (Chen et al., 2016).

Particle size analysis

Particle size analysis can also help in determining TAP contributions to receptor areas. The technique is based on the physical properties of solid (particulate) pollutants. Due to gravity, larger and heavier particles have a comparatively shorter retention time in the atmosphere (Clark, 1988), and are therefore deposited relatively close to emission sources, while smaller sized particles are more buoyant and can therefore be transported over much longer distances and be deposited far from their origin (Larsen, 2000; Inoue et al., 2013; Vukić, Fott, Petrusek, and Šanda, 2006). Particle size can be used to determine whether a pollutant originated from local or more distant sources, with TAP generally being the material of long distance origin. The proportional contribution of TAP can be quantified using Equation 5.

$$\text{TAP contribution} = \frac{n_t}{N} \times 100\% \tag{5}$$

where n_t represents the count of particles that are smaller than the benchmark size of TAP; N represents the count of particles of all sizes.

Particle size analysis has been widely adopted in distinguishing the sources of solid pollutants, such as PM (Samara, 2017), charcoal (Clark, 1988) and SCPs (Inoue, Tomozawa, and Okudaira, 2013). It has also been used as part of sediment-based studies to reveal long-term variations in TAP (Larsen, 2003). However, there are no standard size criteria for differentiating local pollution and TAP. For instance, the threshold size of SCPs accumulating at a site from remote areas can vary from 5 µm to 20 µm (long axis dimension), owing to differences in meteorological conditions and emission sources (Hirakawa et al., 2011; Larsen, 2003). In addition, larger particles might not always be from local sources. Research shows that fine hygroscopic aerosols undergo chemical aging during transportation and particle size can, as a result, increase by a factor of seven due to particle coalescence (Liousse, Devaux, Dulac, and Cachier, 1995).

• *applications of particle size analysis in determining TAP contributions in SEA.* To the best of our knowledge, this approach has not been adopted in TAP research in SEA.

Discussion

The challenge of TAP studies in SEA

Insufficient in-situ sampling and measurement of atmospheric pollutants in SEA

In-situ sampling and measurements are the foundation of satellite remote sensing and numerical modeling approaches to establishing TAP. However,

the density and spread of monitoring stations throughout SEA is far from optimal. Only seven out of ten ASEAN countries have local government-maintained monitoring stations. Poor levels of maintenance of existing monitoring stations are also a problem in less economically-developed countries. For instance, one third of national monitoring stations in the Philippines were offline in March 2018 (Philippine Environmental Management Bureau, 2018). Further, biomass burning is one of the main atmospheric pollution sources in the Indochina peninsula (Yadav et al., 2017). Satellite-borne observations showed Myanmar, Cambodia and Laos have the highest fire occurrence in this region (Vadrevu & Justice, 2011). However, these three countries have no permanent local government-maintained monitoring stations, while they are locations for only four monitoring stations supported through international projects (Table 2).

The lack of a region-wide campaign aimed at measuring and monitoring TAP is another problem in SEA. There have been several field campaigns in SEA targeting atmospheric pollution (Table S2). However, only one campaign is currently ongoing and focused on TAP in SEA: Seven SouthEast Asian Studies (7-SEAS), which focuses on anthropogenic emissions, including those from biomass fires, and transport in SEA (NASA, 2010). The lack of regional networks and campaigns is in part due to political sensitivities within the region. For example, aircraft campaigns in SEA are normally conducted within one country (Hewitt et al., 2010), or over the oceans (Jacob et al., 2003). The first multinational aircraft campaign in SEA was proposed by NASA through the mission of Southeast Asia Composition, Cloud, Climate Coupling Regional Study (SEAC^{4}RS). However, it was canceled due to difficulties in obtaining consensus and approval among different governments in the region (Cole, 2012).

In order to address the problem of TAP and to mitigate potential rises in geo-political tensions in one of the world's fastest developing regions, SEA needs a monitoring network that is more comprehensive and that has a region-wide coverage compared with present. Such a network will also facilitate the quantification and interpretation of atmospheric pollution data (Koh & Teo, 2009). Governments and inter-governmental agencies in the region will also need to find the political will in order to deal, effectively, with the rising challenge of TAP.

Uncertainties in space-borne observation in SEA

Despite satellite-based observation data being fundamental to EIA and CTMs, they have limitations. The first is sampling bias. Satellite-based passive sensors can only generate high quality data when the surroundings are free of obstructions in the atmosphere, such as water vapor and aerosol particles (Zhao et al., 2013). However, cloud cover days in SEA may

account for as much as 60% of total (Feng & Christopher, 2013). In addition, high aerosol loading days are normally associated with cloud cover (Kaufman & Koren, 2006); thus, it is difficult for satellite-borne passive sensors to collect information during periods of heavy pollution. Therefore, the high cloud cover rate in SEA largely limits and causes bias in the utilization of satellite-based data (Feng & Christopher, 2013; Reid et al., 2013; Ford & Heald, 2016). Although satellite-borne remote sensing products from active sensors, such as Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), can provide a basis for the vertical profiling of pollutant concentrations irrespective of weather conditions, this comes at the cost of temporal resolution (Veefkind, 2001). For instance, the revisit time for CALIOP is 16 days (Winker, Hunt, and McGill, 2007). Therefore, one possible solution is to integrate other kinds of ground-based observation data that can provide day and night observation data despite the cloud and aerosol loading conditions (Husar, Husar, and Martin, 2000; Lee et al., 2017).

The second limitation is uncertainty in satellite AOD retrievals. AOD retrievals are used in many studies to estimate surface concentrations of targeted pollutants. However, the ambient environment can easily affect the accuracy of AOD-derived data. One factor is Relative Humidity (RH). Measurements of AOD may be exaggerated when RH is high and in the presence of abundant hygroscopic fine aerosols, which have high reflective ability (Ford & Heald, 2016). This is especially an issue in SEA countries, such as Singapore, where the 24-hr mean RH is 83% (NEAS, 2018). Therefore, validation and calibration with *in-situ* measurement data are needed in order to guarantee the data quality of satellite-based data.

Incomplete EIA in SEA

EIA provides local emissions information, which is the basis of TAP quantification. The global-scale, satellite-borne, observation-based emission inventories fully cover SEA. However, the application of global inventories, which have relatively coarse spatial resolution (from $0.1^\circ \times 0.1^\circ$ to $1^\circ \times 1^\circ$, see Table S4), across a region as diverse as SEA is problematic, because both the resolution and accuracy of the data are compromised (Granier, Artaxo, and Reeves, 2004; European Commission Joint Research Centre & Netherlands Environmental Assessment Agency, 2016). Another problem of global inventories is that they cannot track all kinds of pollutants, because of the limited range of retrieval products available from remote sensing systems. Most global inventories of emissions have not included heavy metals (except mercury) (European Monitoring and Evaluation Programme, 2018). However, heavy metals are emitted from both petrochemical processing

and biomass burning in SEA (Akagi et al., 2011), and in other parts of Asia, and are thus expected to feature relatively heavily in local pollution in, and TAP exported from and to, the region.

One solution is to use a bottom-up, statistical document-based national EIA as a supplement to global inventories (Stella & Keating, 2007). The country level inventory can provide comprehensive emission data for a variety of pollutants, including heavy metals (European Environment Agency (EEA), 2016). The data have a fine spatial resolution that can pinpoint individual major point emission sources, since they are summed from individual sources. All EU countries and the US have routinely conducted EIA for decades (EEA, 2016; United States Environmental Protection Agency, 2016). However, SEA countries do not routinely compile national EIA; only Singapore proposed establishing emission inventories for some major air pollutants, such as SO_2, CO and PM for Singapore and adjacent landmasses through a project of 14 months duration (Feng, 2015). However, no further research findings have been released to the public so far.

Suggestions and outlook of TAP studies

The following section discusses the uncertainties and inadequacies in current TAP studies and attempts to provide suggestions for future TAP studies in SEA, and also for wider applications.

Understanding the locations of receptors and sources

Many researchers adopt trajectory models to indicate the sources of TAP after determining the pollution situation in receiving areas, especially, despite its limitations, the online HYSPLIT trajectory model (Stein et al., 2015). However, it is significant to note that there are uncertainties attached to the use of trajectories of air mass parcels to identify pollution sources. First, many studies simply assigned the higher-than-usual portion of pollution to non-local sources (Fang, Kuo, and Zhuang, 2015; Hee, Khor, Lim, and Jafri, 2015), even though at least some of the excess may have originated from local sources (Aouizerats et al., 2014). Trajectory models cannot differentiate between pollution sources located different distances away. To reduce uncertainty, one possible solution is to conduct EIA in receptor areas and check the local meteorological conditions, to have a better understanding of local emissions and local weather conditions during the period of interest before interpreting the trajectory results.

Second, the objects of trajectory models are air parcels, not pollutants. During transportation, pollutants undergo dry/wet deposition, resuspension, and physicochemical changes (Chen et al., 2017; Konovalov, Beekmann, Berezin, Formenti, and Andreae, 2017). These changes all affect

the travel distances of pollutants, but trajectory models cannot simulate these processes. For better simulation results, researchers could couple dispersion functions with their trajectory calculations, by either choosing CTMs instead of trajectory models (Stein et al., 2015), or combining the trajectory results with the dispersion calculation results from CTMs (Chuang, Fu, et al., 2016; Godowitch & Draxler, 2006).

Third, chosen trajectory modeling times are normally less than five days, while the starting height of air parcels above the potential source is actually not near ground surface (the default starting height is 0.5 km in the HYSPLIT trajectory model) (Draxler & Rolph, 2003). However, proving that pollutants in air parcels 0.5 km above the assumed pollution source are actually from the source is difficult. Chances are that the potential pollution source is also a receptor, since many pollutants are able to suspend in the air for more than five days (Table 1). Therefore, caution is advised when interpreting trajectory results, and other source apportionment approaches should also be considered for validation (Kong et al., 2010). Simply using forward trajectory calculations, starting from the potential sources, may also be beneficial (Vogel et al., 2014).

Understanding pollutant behavior during transportation using CTMs

Pollutants can undergo chemical transformation during transportation, especially in tropical areas with relatively high levels of UV radiation and temperature (Chen et al., 2017; Konovalov et al., 2017; Radojevic, 2003). CTMs can simulate changes of pollutant concentrations during the entire transportation process. However, simulations of secondary pollutants and microphysical changes of aerosols are more problematic (Reid et al., 2009). This is especially the case in SEA, where relatively dense stands of forest can still be found in a region that is predominantly marine; the formation of Secondary Organic Aerosol (SOA) is harder to simulate accurately in CTMs owing to the presence of both biogenic and anthropogenic SOA precursors (sea salt, reactive nitrogen, CO, hydrocarbon, etc.) (Stone et al., 2011; Trivitayanurak et al., 2012). Problems in accurately simulating conditions also arise because of a shortage of accurate emission inventory input data and knowledge of chemical transformations of pollutants in the environment (Aouizerats et al., 2014; Fu et al., 2016; Yadav et al., 2017). Modelling results are also influenced by estimated injection heights of smoke plumes, which directly affects the simulation of pollutant mixing, chemical reactions, and transport distances (Jian & Fu, 2014; Paugam, Wooster, Freitas, and Val Martin, 2016). However, it is also hard to estimate correctly the smoke injection height in SEA due to different types of biomass burning (Jian & Fu, 2014) and ambient meteorological conditions (Labonne, Bréon, and Chevallier, 2007).

One possible solution to these problems is to conduct *in-situ* sampling and measurement campaigns. The aerosol compositions (Lee, Ram, et al., 2016; Xiao et al., 2017), microphysical changes (Chuang, Hsiao, Wang, Tsay, and Lin, 2016), and outflow of SOA over the West Pacific (Kondo et al., 2004) of fire plumes from biomass burning in the northern SEA (Indochina peninsula to Taiwan) have been extensively studied during several field, cruise and aircraft campaigns in 7-SEAS, BASE-ASIA[2] and TRACE-P[3]. These *in-situ* data were used as model input and for model validations. Aircraft-based campaigns provide an opportunity to collect samples directly from polluted air plumes along the transportation trajectory, and data from these can reveal the formation of SOA in the plumes (Stohl et al., 2007). However, such aircraft campaigns are rare in maritime SEA (Parker et al., 2016). The most recent campaign, 'Oxidant and particle photochemical processes above a South-East Asian tropical rainforest' (ACES/OP3), was carried out in Sabah (Malaysia) in July 2008. The samples collected from the aircraft were used to infer the formation of SOA (Stone et al., 2011). However, the amount of SOA as a proportion of total organic aerosol was low since the sampling was conducted during a non-hazy period (Stone et al., 2011; Trivitayanurak et al., 2012). Direct aircraft-based measurements of SOA during very hazy periods are still lacking. They are badly needed in SEA, in order to calibrate modeling results and satellite-based observations, and to observe the chemical transformation of pollutants during transport (Parker et al., 2016).

Aside from the conventional method, using satellite-borne, active sensors that are difficult to calibrate (Labonne et al., 2007), smoke injection height can be determined, largely independently of cloud cover, through aircraft-, balloon- and potentially UAV-based observations. For example, the altitude of the aerosol mixing layer in Indonesia was estimated at 1.5–2.5 km (Labonne et al., 2007). Aircrafts, balloons and drones can all potentially reach this height. However, both aircraft and balloon campaigns are relatively rare in maritime SEA (Table S2). By comparison, with higher mobility and greater availability (and increasingly so in future), UAVs have been successfully used in *in-situ* sampling and measurement of emissions from biomass burning and industrial activities (Krüll, Tobera, Willms, Essen, and von Wahl, 2012; Ronkainen, 2016). They have also been used to sample atmospheric pollutants from a few hundreds of meters to a few kilometers above ground level (Villa, Gonzalez, Gonzalez, Miljievic, Ristovski, and Morawska, 2016). Therefore, the application of drones in *in-situ* smoke

[2]Biomass-burning Aerosols in South East Asia: Smoke Impact Assessment.

[3]Transport and Chemical Evolution over the Pacific.

injection height detection is theoretically feasible and worth exploring further in SEA.

Understanding the contribution of TAP

The results of TAP contributions from different geographical sources provide valuable information for policy-making. Because contribution apportionment requires the understanding of both local pollution and TAP, the possibility of overlooking or underestimating local pollution (or TAP), while exaggerating the impact of TAP (or local pollution) is reduced. For instance, local pollution in Singapore is reported to have decreased following implementation of tightened environmental regulation since 1970s (Chen et al., 2015). However, other studies report that local pollution still forms a major contribution to poor air quality (Aouizerats et al., 2014; Lee et al., 2017). This reveals the possibility of different interpretations under the same TAP context from different points of view: local pollution is decreasing, which implies TAP is the only reason for poor local air quality, or local pollution is still serious, which implies both local pollution and TAP are responsible.

This paper has reviewed four approaches that can be used to determine the contribution of TAP from different geographical sources. However, due to their added complexity, the number of contribution studies is far fewer than the number of simple source appointment, back-trajectory studies (there are almost 800 HYSPLIT trajectory model related papers, according to the search results from Web of Science in March 2018). The choice of methods depends on research purposes. If the research aims to provide suggestions for local environmental management or to assist regional cooperation on addressing TAP issues, future studies ought to consider local emission analysis and results of an examination of the contribution of TAP, thereby providing decision-makers with a basis for comprehensive, balanced and holistic understanding.

Understanding historical variations of TAP

Past variations in pollution, especially over the past 100 years during which conditions in SEA have changed dramatically, are invaluable to current understanding of TAP, because they can provide possible avenues for future pollution abatement (Last & Smol, 2006). Even over a period as short as six years, solid evidence of variations of TAP, as a result of regional development, exists, as shown in the example from the Taiwan Strait cited in the section of Mass Balance Approach. However, long-term TAP studies are not common in SEA; most studies have focused on short-term pollution episodes (Li et al., 2014). The earliest *in-situ* measurement network in SEA started only after the 1990s

(EANET since 1993, see Table S1). These short-term studies and observations do not provide an adequate foundation for examinations of long term pollution trends in the region (Kanniah, Lim, Kaskaoutis, and Cracknell, 2014).

Palaeoenvironmental studies, such as those utilizing lake (and reservoir) sediments, have the potential to generate information on variations in atmospheric pollution depositions and their effects that stretch over hundreds, even thousands, of years to the present. They can therefore be used to in-fill gaps and extend the total period of time covered by existing monitoring records, and to investigate the effects of attempts to mitigate pollution levels. This potential is already being realized in other parts of the world, for example in Ireland (O'Dwyer & Taylor, 2010). However, their application in SEA has to date been limited. This is largely due to the difficulties in separating atmospheric deposition inputs from other pollutant input paths, such as catchment natural runoff and anthropogenic sewage input (Yang, 2015). For SEA, the challenge also lies in data availability; there is an absence of data from several countries in the region, while a large proportion of the sediment records that are available are poorly resolved and insufficiently-well dated, or not dated at all (Table S5).

The utilization of sediment-based data in TAP studies remains promising (e.g. Engels et al., 2018). This promise is likely to be enhanced if future studies investigating long-term variations in atmospheric pollution loads and depositions: 1) include long-term EIA and adopt a mass balance approach to differentiating local pollution and TAP (Heyvaert et al., 2000); and/or 2) establish correlations between the preserved sedimentary proxies and contemporary observations of pollutant loads (via satellite or *in-situ* measurements). The latter provide a means of calibrating relationships between pollutants accumulating at the receptor location (sedimentary basin), and those released from potential sources (Field et al., 2009).

Conclusions

There currently exist serious challenges in SEA concerning TAP, which is increasingly becoming a major geopolitical issue. The situation is likely to get even worse in the future, as rapid economic growth and increases in the consumption of resources, including energy, continue. However, the conventional methods used in TAP determination, which depend on satellite-borne remote sensing data, *in-situ* measurement and CTMs simulation, have many limitations when applied in SEA. Researchers on air quality and related issues in the region are therefore encouraged, 1) to explore the utilization of state-of-the-art measuring equipment (e.g. UAVs) to collect data, 2) to make the most of the existing data and opportunities (e.g. the availability of proxy pollution-related data in sedimentary records) to cover

existing gaps in information and extend monitoring records, and 3) to experiment with new quantitative methods that are independent of space-based and ground-based data (e.g. particle size analysis and palaeolimnology with mass balance approach).

In the shorter-term, SEA countries ought to consider improved collection (monitoring) of pollution data. Improved data collection could involve deploying more ground-based air quality observation stations, conducting regional field campaigns that allow TAP to be distinguished in SEA, and making country-level EIAs routine. However, these actions cannot be done without cooperation within the region (and farther afield), and without the political will to implement measures aimed at responding to evidence of poor air quality, a proportion of which may be from sources located in a different jurisdiction (i.e. be transboundary). These factors make the problem-resolution as much a political as environmental challenge (Lee, Jaafar, et al., 2016). Responding to this challenge will require governments in the region to make long-term financial and political commitments to pollution monitoring and research, and to share information and respond effectively to evidence of persistently poor air quality.

For better environmental management and human well-being, this paper emphasized the importance of understanding local pollution in TAP issues. This is because that information has the potential to provide an unbiased understanding of pollution levels in receptor areas. Also, this paper highlights the importance of understanding pollution trends, and long-term variations in pollution concentrations and depositions. These long-term data have the potential to provide information that is crucial for environmental managers and policy-makers at local, national and regional scales.

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