Environmental Pollution 235 (2018) 907–917

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Historical atmospheric pollution trends in Southeast Asia inferred from lake sediment records☆

S. Engels [a, b, *], L.S.R.Z. Fong [c], Q. Chen [c], M.J. Leng [a, d], S. McGowan [a, e], M. Idris [f], N.L. Rose [g], M.S. Ruslan [f], D. Taylor [c], H. Yang [g]

[a] Centre for Environmental Geochemistry, School of Geography, University of Nottingham, Nottingham, NG7 2RD, UK
[b] School of Geography, Birkbeck University of London, Malet Street, London, WC1E 7HX, UK
[c] Department of Geography, National University of Singapore, Singapore, 117570, Singapore
[d] NERC Isotope Geosciences Facilities, British Geological Survey, Nottingham, NG12 5GG, UK
[e] School of Environmental and Geographical Sciences, University of Nottingham Malaysia Campus, Jalan Broga, 43500 Semenyih, Selangor Darul Ehsan, Malaysia
[f] Tasik Chini Research Centre, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600, Malaysia
[g] Environmental Change Research Centre, Department of Geography, University College London, London, WC1E 6BT, UK

ARTICLE INFO

Article history:
Received 12 October 2017
Received in revised form
2 January 2018
Accepted 3 January 2018

Keywords:
Fossil fuel combustion
Emission trends
Fly ash particles
Mercury
Southeast Asia

ABSTRACT

Fossil fuel combustion leads to increased levels of air pollution, which negatively affects human health as well as the environment. Documented data for Southeast Asia (SEA) show a strong increase in fossil fuel consumption since 1980, but information on coal and oil combustion before 1980 is not widely available. Spheroidal carbonaceous particles (SCPs) and heavy metals, such as mercury (Hg), are emitted as by-products of fossil fuel combustion and may accumulate in sediments following atmospheric fallout. Here we use sediment SCP and Hg records from several freshwater lentic ecosystems in SEA (Malaysia, Philippines, Singapore) to reconstruct long-term, region-wide variations in levels of these two key atmospheric pollution indicators. The age-depth models of Philippine sediment cores do not reach back far enough to date first SCP presence, but single SCP occurrences are first observed between 1925 and 1950 for a Malaysian site. Increasing SCP flux is observed at our sites from 1960 onward, although individual sites show minor differences in trends. SCP fluxes show a general decline after 2000 at each of our study sites. While the records show broadly similar temporal trends across SEA, absolute SCP fluxes differ between sites, with a record from Malaysia showing SCP fluxes that are two orders of magnitude lower than records from the Philippines. Similar trends in records from China and Japan represent the emergence of atmospheric pollution as a broadly-based inter-region environmental problem during the 20th century. Hg fluxes were relatively stable from the second half of the 20th century onward. As catchment soils are also contaminated with atmospheric Hg, future soil erosion can be expected to lead to enhanced Hg flux into surface waters.

1. Introduction

Asia has undergone strong economic growth over the last few decades leading to a doubling in regional energy consumption between 1980 and 2003 (Richter et al., 2005; Ohara et al., 2007), with a continuous growth of energy consumption being observed since 2003 (Kurokawa et al., 2013; EANET, 2015). Fossil fuel combustion emits atmospheric pollutants, particularly SO_2, NO_x, CO, non-methane volatile organic compounds, organic carbon, black carbon and trace metals. Although pollutant emissions, particularly from the burning of coal, have been declining in Europe and North America over the last two decades, this has not been the case for much of Asia (Amann et al., 2013; Klimont et al., 2013; Kurokawa et al., 2013). While emissions of SO_2 and particulate matter ($PM_{2.5}$) decreased by 12–15% in East Asia between 2005 and 2010, emissions of NO_x and non-methane volatile organic compounds increased by 15–25% (Wang et al., 2014a). Electricity demand in Southeast Asia (SEA), one of the world's fastest developing regions,

☆ This paper has been recommended for acceptance by Eddy Y. Zeng.

* Corresponding author. School of Geography, Birkbeck University of London, Malet Street, London, WC1E 7HX, UK.
E-mail address: s.engels@bbk.ac.uk (S. Engels).

https://doi.org/10.1016/j.envpol.2018.01.007
0269-7491/

is projected to be 83% higher in 2035 than in 2011 (International Energy Agency, 2013), with coal providing much of this increased energy demand (Lai et al., 2016).

Increased atmospheric pollution has major implications for society and the environment. An estimated 6.5 million deaths globally each year are attributed to poor air quality (International Energy Agency, 2016; World Health Organisation Press, 2016). Koplitz et al. (2017) suggest that the current estimate of around 20,000 ($11.4–28.4 \times 10^3$) excess deaths per year due to emissions from burning coal in SEA will increase to around 70,000 ($40.1–126.7 \times 10^3$) by 2030. Perhaps contrary to common perception (Lai et al., 2016), around 9000 of these excess deaths as a result of increased coal use in SEA are anticipated to occur in China; rising coal emissions in SEA could thus become an increasingly transboundary pollution issue (Koplitz et al., 2017).

Atmospheric greenhouse gas concentrations resulting from fossil fuel combustion are one of the key drivers of anthropogenic climate change (IPCC, 2014), and aerosols (particularly atmospheric black carbon) can significantly influence global radiative forcings (Jacobson, 2001; Streets et al., 2004). Fine aerosol particles can further influence regional climate via surface dimming (Ramanathan et al., 2005; Lau et al., 2006); Fu et al., 2017). Atmospherically deposited pollution places further pressure on anthropogenically impacted wetlands and lowland lakes in SEA, with existing impacts including eutrophication, intensified aquaculture, water abstraction, dam construction, biomass burning, as well as catchment disturbances such as agriculture (e.g. oil palm and other plantations), urbanisation and mining activities (Sharip et al., 2014). Those aquatic ecosystems that have not been severely degraded yield valuable services, such as food and water for local populations and the provision of livelihood opportunities such as eco-tourism (Shuhaimi-Othman et al., 2007; Stockholm International Water Institute, 2009; Sharip and Jusoh, 2010). These services may be difficult to replace. Moreover, human impacted aquatic ecosystems are likely to have significantly reduced biodiversity value (Kopf et al., 2015). Unfortunately, information on the current status of many of these ecosystems, and on the rates and directions of change in environmental conditions over recent decades, is generally lacking. In addition, detailed inventories of anthropogenic energy sources and emissions for Asia only span the last few decades (Kato and Akimoto, 1992; Akimoto, 2003; Streets et al., 2003; Kurokawa et al., 2013) and their number is comparatively low (Ohara et al., 2007; Rose, 2015).

Natural archives such as lake sediments have the potential to provide essential information on spatio-temporal variations in fossil fuel consumption in SEA, as spheroidal carbonaceous particles (SCPs), by-products of high-temperature industrial fossil fuel combustion, can be stored in these sediment records following atmospheric deposition, thus tracking changing influx with time (Rose, 2015). SCPs are fine carbonaceous aerosols, typically 2–50 μm across, formed from the incomplete high-temperature combustion of fossil fuels such as oil and coal (Rose et al., 1994; Rose, 2001; Chirinos et al., 2006). SCPs can be transported for thousands of kilometres through the atmosphere under favourable meteorological conditions (Rose et al., 1998; Yang et al., 2001; Inoue et al., 2014), and have been found in remote places such as the Falkland Islands and Antarctica, far removed from the nearest sources (Rose et al., 2012). SCPs are a component of the "black carbon continuum" (Rose, 2008, 2015), but whereas other components of black carbon can be the result of domestic emissions, road transport emissions, or biomass burning (e.g. Kurokawa et al., 2013), SCPs are only formed during high-temperature industrial fossil fuel combustion. As they have no natural sources, SCPs encountered in lake sediment records can be used as indicators of atmospheric deposition from industrial sources (Rose, 2001), especially as they are not susceptible to post-depositional alteration, movement in the sediment column (except by bioturbation), or degradation (Rose et al., 2003). Analysis of SCPs stored in sediments provides a means to reconstruct trends in emissions from the combustion of fossil fuels over time-periods that extend beyond the beginning of documentary and instrumental evidence, which commenced only in the second half of the 20th century and in some regions even more recently (Rose, 2001). Spatial patterns in SCP distribution have been shown to be closely linked to other pollutants, such as sulphur and polycyclic aromatic hydrocarbons (Rose and Juggins, 1994; Rose et al., 1998; Barst et al., 2017). A global collation of SCP records includes a disproportionately large number of records from Western Europe and none from SEA (Rose, 2015).

Anthropogenic emissions of Hg date back to pre-industrial times, but global Hg emission rates have tripled over the last 150 years, mainly due to increased coal burning (Hylander and Meil, 2003; Engstrom et al., 2014; Horowitz et al., 2014; Yang et al., 2016). Aside from industrial sources, there are many additional anthropogenic sources of atmospheric Hg emission, including waste incineration, sulphide ore processing, cement kilns and the production of various metals (Hylander and Meil, 2003). Another important potential source of atmospheric Hg is artisanal and small-scale gold mining (Mason and Pironne, 2009; Cordy et al., 2011). Hg is among the most toxic elements and poses serious threats to both human health and aquatic ecosystems due to its tendency to bioaccumulate and biomagnify through the food chain (Azimi and Moghaddam, 2013; Rice et al., 2014; Okelsrud et al., 2016). Mercury contamination in waterways, sediments and fishes in SEA are already threatening frigate birds as well as other species in the area depending on marine resources, including humans (Mott et al., 2017). With an atmospheric residence time of up to two years (Schroeder and Munthe, 1998), Hg can be released from the atmosphere through both wet and dry deposition. Hg can subsequently be stored in lake and wetland sediments, thereby providing the basis for reconstructions of past variations in pollution loads (e.g. Bindler et al., 2001; Fitzgerald et al., 2005; Yang et al., 2010; Shotyk et al., 2017).

Little information on past levels of industry-derived air pollution in SEA is available, especially for the period pre-dating 1980 (US Energy Information; www.eia.gov). This is despite the growing concerns of the effects of local and regional pollution in SEA (Koplitz et al., 2016). This paper addresses this information gap in long-term variations in air pollution deposition in SEA and uses sedimentary evidence as a basis for reconstructing changes in atmospheric pollution in SEA covering the period from the start of industrial fuel consumption to the present, including the time interval before 1980 where data on atmospheric pollution levels are otherwise scarce.

2. Materials and methods

2.1. Sites and sampling

Using logistical criteria such as site-accessibility and geographical spread, we selected sites from three countries in SEA in order to develop a regional reconstruction of atmospheric deposition pollution history: (1) sediment cores were obtained from three lakes in the Philippines (Yambo, Mohicap, Sampaloc) from the Seven Crater Lakes, a tight cluster of maar crater lakes located near San Pablo City, Laguna Province, on the island of Luzon (Fig. 1). The lakes are presumed to have formed through an explosive phreatic eruption from Mount Banahaw-San Cristobal (Brillo, 2016). The lakes are all moderately deep (>25 m water depth) and have surface areas ranging from 0.23 to 1.04 km^2 (Supplementary Table 1) (Aquino, 1983; Laguna Lake Development