

# **Impact of Biomass Burning on Ocean Water Quality in Southeast Asia through Atmospheric Deposition: Field Observations**

## **Supplementary Material**

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**Background information.** A significant and increasing source of nutrients to freshwater and marine ecosystems is atmospheric deposition (AD), either as “wet deposition” or as “dry deposition” of particles and “gaseous exchange” between the air and water (Fig. 1 in the present article). Three steps are involved in wet deposition: (i) the pollutants need to come into contact with condensed water in the atmosphere; (ii) the pollutants must be scavenged by the droplets; and (iii) it then has to start raining before the condensed water evaporates back into water vapor, thereby releasing the pollutants back into the air

(Seinfeld and Pandis, 2006). Dry deposition can be conceptualized as a three-step process: (1) the gas or particle is moved toward the surface by thermally or mechanically driven eddies; (2) it is transferred by diffusion across a thin layer close to the surface where turbulence is absent; and (3) the gas or particle is captured by the surface (Seinfeld and Pandis, 2006). Atmospheric deposition is a significant and potent source of nutrients that can accelerate eutrophication and its associated environmental consequences in freshwater, estuarine, and coastal ecosystems (Duce et al., 2008, Galloway et al., 2008). Both airborne N and P species are derived from natural and anthropogenic (point and non-point) sources and transported to surface water and their watersheds. Anthropogenic sources are agricultural, industrial and human activities, combustion of fossil fuels, transportations, and natural sources are gases and particles from mineral aerosols, primary biogenic particle and biomass burning emissions, and aerosols from the ocean. It has been reported that about 20-40% of new N inputs into coastal waters are of atmospheric origin (Duce, 1986; Paerl, 1995) and atmospheric deposition alone contributes from 300 to >1000 mg N/m<sup>2</sup>/yr in coastal waters (Duce et al, 1991). In the open sea, the contribution of atmospheric deposition of nitrogen is relatively large (40–50% of the total load, Cornell et al., 1995). Galloway and Cowling (2002) showed that anthropogenic nitrogen fixation will increase by ~60% by 2020 and most of the N increase will be in Asia (Galloway et al., 1995). AD of P typically occurs at concentrations less than a few percent those of N (Duce, 1986) which is especially true in regions where wet depositions exceed dry, since P is usually bound to particles such as dust and windblown soils. The nutrient composition in rainwater indicated a dominant anthropogenic source for N species and a continental, natural and anthropogenic source for P species (Herut et al, 1999). Overall, it appears that airborne P

typically accounts for 10 to 20 percent of total phosphorus loadings to water bodies from all sources (Swackhamer, 2004). The flux of P to the marine environment resulting from dissolution from eolian dust has been estimated to be  $3 \times 10^9$  mol P/yr which represents about 10% of river fluxes to the ocean (Delaney, 1998). From an ecological perspective, however, P may be of considerable importance since far less P than N is required for balanced plant growth (Redfield, 1958). The relative degrees of riverine inputs decreased with increasing distance from land, so atmospheric inputs of P played a major role on the biogeochemical processes of some oligotrophic surface seawater (Markaki et al., 2003). Markaki et al. (2003) reported that up to 38% of new production in the eastern Mediterranean was supported by the atmospheric inorganic phosphorus (DIP) dissolved from eolian dusts. Martin et al. (1994) provided strong evidence that the addition of iron to nitrogen and phosphorus rich water mass can result in marked increases in phytoplankton activity.

Biomass burning is a primary source of many trace substances that are important in atmospheric chemistry (Crutzen et al., 1979; 1985; Andreae et al., 1988; Crutzen and Andreae, 1990; Lobert et al., 1990). The smoke haze is caused by a high concentration of airborne particulate matter (PM), predominately of very fine particles with a diameter of less than 10  $\mu\text{m}$ , that is directly emitted from biomass burning together with those from other sources such as industries, on-road vehicles, road dust, as well as PM formed from gaseous pollutants in the atmosphere. These particles often grow in size as humidity increases, further impairing visibility. Haze can become “transboundary” pollution when it is dense at source thousands of miles away and extent great and remains at measurable levels after crossing into another country's air space at remote locations (Example see Fig.

4 in the present article). While coarse particles fall out from the atmosphere within several hours up to a day, fine particles have the longest residence time (up to weeks) in the atmosphere and travel extensive distances (hundreds to thousands of kilometres). It was first recognized in the late 1970s that tropical vegetation burning is a major global source of trace gases, such as CO<sub>2</sub>, CO, NO<sub>x</sub>, NH<sub>3</sub>, and aerosols with significant impacts on regional and global climate, atmospheric chemistry and hydrological cycles (Crutzen et al., 1979; 1985; Crutzen and Andreae, 1990; Lobert et al, 1990; Andreae, 1991; Crutzen and Carmichael, 1993; Yokelson et al., 1999). Ultimately, removal of trace gases from the atmosphere is mainly by oxidation processes.

The emission production and characteristics from vegetation fires strongly depend on the combustion stage (basically flaming and smouldering combustion), the combustion efficiency and the physico-chemical properties of vegetation burnt (Lobert and Warnatz, 1993). The most significant emission of reactive nitrogen (NO<sub>x</sub>) is emitted during the flaming stage of fires while most other nitrogen containing compounds are emitted during smouldering stage of fires. Large diameter or densely packed necromass (such as logs, peats) and large diameter live vegetation (trunks) are usually partially consumed resulting in smouldering combustion (Stocks and Kaufman, 1997; Yokelson et al., 1997). Characteristically for low efficiency combustion processes, smouldering combustion emits larger amounts of incompletely oxidized compounds including CO, CH<sub>4</sub>, NH<sub>3</sub>, other nitrogen containing compounds and fine particles than flaming combustion per unit amount of biomass consumed by a fire. Biomass burning is found to be a major source of reactive nitrogen (Kondo et al., 2004; UNEP and WHRC, 2007) and phosphorous (Mahowald et al., 2005; Baker et al., 2006). The emission rates of NO<sub>x</sub> due to biomass burning were

responsible for ~9–20% of the estimated global rate of terrestrial nitrogen fixation (Lobert et al., 1990). Mahowald et al. (2005) reported that biomass burning emissions and human disturbance were responsible for ~23% of the phosphorus flux in the Amazon and the global source of atmospheric phosphorus (<10  $\mu\text{m}$ ) is 1.39 Tg P /yr, of which 4.8% is anthropogenic (Mahowald et al., 2008). Reid et al. (2005) reported that the fresh smoke particle mass has density around 1.8–2.2  $\text{g}/\text{cm}^3$ , the average density of dry smoke particles likely varies in the 1.20–1.4  $\text{g}/\text{cm}^3$  range; while lower in mass fraction than accumulation mode particles (10%), fires produce a variety of coarse mode particles (typically 2.5 $\mu\text{m}$  – 15 $\mu\text{m}$  diameter); in addition to coarse mode ash particles (2 $\mu\text{m}$ <dp<20 $\mu\text{m}$ ), giant ash particles having diameters of up to a millimeter or more can be generated by very intense fires (these have even been tracked by weather radar). Rising levels of atmospheric deposition lend urgency to understand the fate and impacts of ‘new’ N (Galloway et al., 2008). Nutrients increase due to biomass burning may thus strongly impact terrestrial and oceanic biogeochemistry.

Recurring incidence of air pollution phenomenon on an unprecedented scale due to land and forest fires has been a feature of SEA’s ecology since the Pleistocene Age. The forest fires in Indonesia were the main sources of extensive SEA haze events (Nichol, 1997; Radojevic, 1997, 1998, 2003; Fujiwara et al., 1999; Radojevic and Hassan, 1999). During the 1990s, six separate haze episodes (1983, 1990, 1991, 1994, 1997 and 1998) occurred in Southeast Asia (Radojevic and Hassan, 1999; Muraleedharan et al., 2000). The regional haze episode in SEA is usually associated with dry weather and droughts caused by the El-Niño Southern Oscillation (ENSO) phenomenon. El Niño is an ocean-atmosphere-climate phenomenon that is linked to the periodic warming of waters across the central and eastern

tropical Pacific Ocean. Despite the existing in-depth knowledge on fire and its underlying causes in SE Asian ecosystems (Goldammer et al. 1996), little literature and research exists on the implications of vegetation burning in this region on atmospheric chemistry, public health and aquatic ecosystem (Nichol, 1997, 1998; WHO, 1998; Balasubramanian et al., 1999). The fate of the initial fire emissions depends strongly on both their composition and the regional state of the atmosphere. Once airborne, the particles begin to grow slightly in size as they age through condensation and coagulation. In addition, new fine particles are created by nucleation of gaseous fire emissions such as the conversion of  $\text{NO}_x$  to nitrates (Jänike, 1993). Particles are removed from the atmosphere by gravitational settling, precipitation and cloud scavenging. Because gravitational settling velocity increases with particle diameter, larger particles (diameter  $> 10\mu\text{m}$ ) are lost from the plume faster than smaller ones. Wet removal thus dominates the atmospheric lifetime of the pyrogenic particles, which is therefore largely controlled by meteorology (Garstang et al., 1997).

The forest fires that hit the Association of Southeast Asian Nations (ASEAN) region in 1997-1998 caused the most environmental damage recorded in history (UNEP, 1999). The environmental effects of the more recent 2006 smoke haze episodes remain poorly understood. The United States Environmental Protection Agency (US EPA) developed PSI (Pollutant Standards Index) value, which is adopted by the National Environment Agency (NEA) in Singapore, gives an indication of the prevailing air quality and potential health effects. The PSI was developed to provide the public with information about daily pollution levels (i.e. good, moderate, unhealthy, very unhealthy and hazardous) and to enable authorities to decide on appropriate action to protect the public and to ameliorate the situation. The highest PSI value is reported after the concentrations of  $\text{CO}$ ,  $\text{SO}_2$ ,  $\text{NO}_2$ ,  $\text{O}_3$

and PM<sub>10</sub> are measured (<http://app.nea.gov.sg/psi/>). During haze episodes the PSI is invariably based on PM<sub>10</sub> measurements, because this pollutant far exceeds the concentrations of other pollutants. The role of atmospheric deposition is least understood in SEA and the recurring forest fires & abundant rainfall in SEA region might be an important contributor to “new” nutrient loading of coastal zones and marine ecosystems (Sundarambal et al., 2006, 2009). The atmospheric deposition of nutrients has been identified in SEA as a major unknown, and in order to fully assess the magnitude of the atmospheric sources of P and N species and their impacts of forest fires on air quality and surface water quality, their annual deposition rates to the SEA region need to be quantified. The nutrient compositions (N and P species) derived from total suspended particulate matter (TSP) and rainwater in Singapore are quantified and the results obtained are discussed in this present article (Sundarambal et al., 2010).

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## Appendix A

Deposition velocity ( $V_d$ ) calculation. Aerosol dry deposition velocity at a particular vertical height is calculated as:

$$V_d = \frac{1}{R_a + R_b + R_a R_b v_s} + v_s$$

Where  $v_s$  is the gravitational settling velocity,  $R_a$  is aerodynamic resistance, and  $R_b$  is quasi laminar layer resistance.

Aerodynamic Resistance $R_a$	$R_a$	$R_a = \left[ \frac{9}{u \sigma_0^2} \right]$
Quasi-laminar layer resistance	$R_b$	$R_b = \frac{1}{(1 - \alpha_{bb})(v_B - v_i) + \alpha_{bb}(v_a + v_w)}$
Depending on particle sizes, the viscous (quasi-laminar) layer resistance is largely controlled by processes of Brownian diffusion, interception and impaction. The terms, $v_B$ (Brownian diffusion velocity), $v_i$ (impaction velocity), and $v_w$ (washout velocity) were calculated using following equations:		
Friction velocity	$u^*$	$u^* = \left( \frac{u}{R_a} \right)^{0.5}$
Area of bursting bubbles	$\alpha_{bb}$	$\alpha_{bb} = 1.7 * 10^{-6} * u^{3.75}$
Brownian diffusion velocity	$v_B$	$v_B = \left[ \frac{u^*}{3(S_c R_e)^{0.5}} \right]$
Schmidt number	$S_c$	$S_c = \frac{v}{D}$
Kinematic viscosity of air ( $\text{cm}^2/\text{s}$ )	$v(T,1)$	$v(T,1) = v(273.15\text{K},1) * (T/273.15\text{K})^{1.81}$
Molecular diffusivity of specific molecule ( $\text{cm}^2/\text{s}$ )	$D(T,1)$	$D(T,1) = D(273.15\text{K},1) * (T/273.15\text{K})^{1.81}$
Hydraulic diameter in m	$z_0$	$z_0 = 5\text{cm}$ (assumed)
Reynolds number	$R_e$	$R_e = \frac{u z_0}{v}$
Stokes number	$St$	$St = \frac{v_{gi} (u^*)^2}{g v}$
Impaction velocity	$v_i$	$v_i = u^* 10^{\left( \frac{-3}{St} \right)}$

Average particle deposition velocity in particle size interval $i$ ( $v_{gi}$ ) (m/s)	$v_s$	Obtain from gravitational settling velocity, $v_s$
$v_a$	$v_a$	$v_a = 1/R_a$
Washout velocity	$v_w$	$v_w = 0.5(2\pi r_{sd}^2)(2z_d)F_{sd}$
Diameter of spray drop (m)	$r_{sd}$	$r_{sd}=50 \mu\text{m}$
The average height reached by the spray drops	$z_{sd}$	$z_{sd}=50 \text{ m}$
Flux of spray drops	$F_{sd}$	$F_{sd}=5 \times 10^6 \text{ (m}^{-2}\text{s}^{-1}\text{)}$
$V_s$ (gravitational settling velocity) is calculated based on particle diameter, density, and viscosity:		
Gravitational settling velocity (m/s)	$v_s$	$v_s = \frac{D_p^2 \rho_p g C_c}{18\mu}$
Particle diameter (m)	$D_p$	$D_p$
Particle density ( $\text{kg/m}^3$ )	$\rho_p$	$\rho_p=1 \text{ g/cm}^3$
Cunningham correction factor	$C_c$	$C_c = 1 + 2\lambda^* \left[ 1.257 + 0.4 \exp(-1.1D_p/(2\lambda)) \right] / D_p$
Mean free path of gas molecules in air	$\lambda$	$\lambda=0.065 \times 10^{-6} \text{ m}$
Absolute viscosity of air	$\mu$	$\mu=v_{\text{air}}\rho_{\text{air}}$
Density of air ( $\text{kg/m}^3$ )	$\rho_{\text{air}}$	$\rho_{\text{air}} = \text{Pressure}_{\text{air}} / \text{Temperature}_{\text{ambient}}$ * 287.05J/(kg.K)