



Influence of continental organic aerosols to the marine atmosphere over the East China Sea: Insights from lipids, PAHs and phthalates



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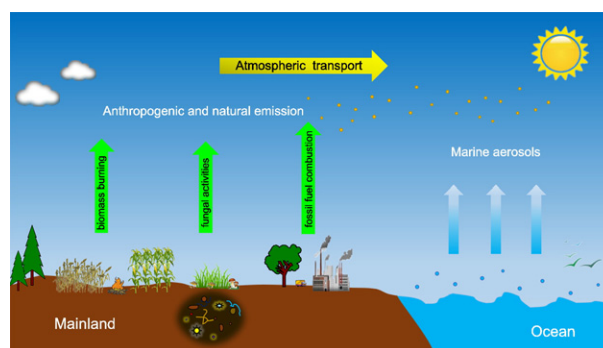
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HIGHLIGHTS

- Spatial distribution of lipids, PAHs and phthalates were measured in marine aerosols over the East China Sea.
- Higher abundance the target compounds was observed in the terrestrially affected aerosols.
- Factor analysis inferred the important contribution from terrestrial sources to the marine aerosols.

GRAPHICAL ABSTRACT



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ABSTRACT

Total suspended particle (TSP) samples were collected during a marine cruise in the East China Sea from May 18 to June 12, 2014. They were analyzed for solvent extractable organic compounds (lipid compounds, PAHs and phthalates) using gas chromatography/mass spectrometry (GC/MS) to better understand the sources and source apportionment of aerosol pollution in the western North Pacific. Higher concentrations were observed in the terrestrially influenced aerosol samples on the basis of five-day backward air mass trajectories, especially for aerosols collected near coastal areas. Phthalates were found to be the dominant species among these measured compound classes ($707 \pm 401 \text{ ng m}^{-3}$ for daytime and $313 \pm 155 \text{ ng m}^{-3}$ for nighttime), followed by fatty acids, fatty alcohols, *n*-alkanes and PAHs. In general, the daytime abundances for these compounds are higher than nighttime, possibly attributable to more intensive anthropogenic activities during the daytime. The factor analysis indicates that biomass burning, fungal activities and fossil fuel combustion maybe the main emission sources for organic aerosols over the East China Sea. This study demonstrates that the East Asian continent can be a natural emitter of biogenic and anthropogenic organics to the marine atmosphere through long-range transport, which controls the chemical composition and concentration of organic aerosols over the East China Sea.

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1. Introduction

Organic aerosols (OA) account for a large fraction of air particulate matter on a global scale, which are of great importance for global climate modeling (Kanakidou et al., 2005). They can alter the radiative balance of the atmosphere by directly scattering or absorbing solar radiation, and indirectly affect climate through modification of cloud properties by acting as cloud condensation nuclei (Albrecht, 1989; Lohmann and Feichter, 2005; Novakov and Penner, 1993; Ramanathan et al., 2001). Moreover, airborne particles contain substantial hazardous organic matters, biological organisms and pathogens (e.g., pollen, bacteria, spores, viruses), which can cause or enhance respiratory, cardiovascular, infectious, and allergic diseases (Pope et al., 2004; Pöschl, 2005). All these effects depend on the chemical compositions and abundances of atmospheric aerosols (Hinds, 2012; Kanakidou et al., 2005). Though studies on chemical characterization and behavior of OA in the atmosphere have been widely conducted at different locations so far (Fu et al., 2010; Pavuluri et al., 2015; Zhang et al., 2016), more attention has been paid to urban atmospheric aerosols owing to ever-increasing industrial activities and rapidly deteriorating living environment in urban area. In contrast, investigations about organic aerosols in marine atmosphere are relatively limited at present due to the difficulty and inconvenience of sampling marine aerosols as well as their diverse sources (Fu et al., 2011; Simoneit et al., 2004). However, it is widely known that oceans cover beyond 70% of the Earth's surface, which could exert a strong influence on the global climate change and biogeochemical cycling.

The East China Sea lying over the broad shelf of the western North Pacific is adjacent to the Asian mainland, where continental-scale air pollution has drawn a strong attention in recent years due to the rapid increase of industrialization and urbanization in East Asia. It was reported that the westerly winds prevail during winter/spring in the western North Pacific (Fu et al., 2012; Kawamura et al., 2003). Therefore, the terrestrial organic matters from Asian continent can be carried to the western Pacific through long-range atmospheric transport, imposing a heavy load on the marine atmosphere when the westerly winds dominate in spring. Thus, surface waters of East China Sea are susceptible to air-sea transfer due to its proximity to intense natural and anthropogenic activities in Asia (Uematsu et al., 2010). Previous studies reported that continental air masses from Asia continent greatly affected the marine aerosols in western North Pacific region (Uematsu et al., 2010; Zhou et al., 1990), which may have further impacts on global radiation balance, atmospheric chemistry, and marine biogeochemistry. However, to date, little chemical information was documented on the concentrations and spatial distributions of polycyclic aromatic hydrocarbons (PAHs), phthalates and lipids in the marine aerosols over the western North Pacific Ocean. Lipid compounds of terrestrial biomarkers, such as *n*-alkanes, fatty acids and fatty alcohols, mostly are chemically stable and long-lived (Bush and McInerney, 2013). Thus, these terrestrial biomarkers can act as tracers to reconstruct the changes in the atmospheric transport and circulation patterns (Kawamura et al., 2003). PAHs are ubiquitous pollutants in ambient atmosphere, mainly from anthropogenic emissions, such as vehicle exhausts and other incomplete combustion sources (Masclat et al., 1987; Rogge et al., 1993a; Rogge et al., 1993b). Many PAHs have been identified as potential carcinogens and priority pollutants (Menzie et al., 1992). Once emitted into the atmosphere, most PAHs could enter into coastal aquatic systems and open oceans through atmospheric deposition, possibly contributing to the toxic burden of marine organisms (Mesquita et al., 2016). On the other hand, PAH diagnostic ratios are widely used to identify pollution emission sources (Yunker et al., 2002). Therefore, the measurement of composition and variation of PAHs could provide valuable information on the contribution of different sources to marine aerosol particles and the toxic potential of these organic constituents, helping understand the influence of continental organic matter on the marine atmosphere as well as the toxicity assessment of marine environment.

Phthalates or phthalic acid esters (PAEs), suspect environmental endocrine disruptors (Keith, 1998), are widely used in the manufacture and processing of plastic products as plasticizers. Because they are just physically rather than chemically bound to the polymeric matrix, PAEs can easily release into environment (Net et al., 2015). It has been reported that PAE exposure could affect human reproduction, including fertility problems, newborns' development and carcinogenic properties (Barbara et al., 2008; Bornehag and Nanberg, 2010; Fisher, 2004; Howdeshell et al., 2008; Swan et al., 2005). However, the data about composition, concentration and spatial distribution of PAEs in marine aerosols are poorly reported, especially their potential impacts on the marine ecosystems.

Therefore, the purpose of this study is to investigate the molecular composition, abundances and spatial distributions of PAHs, phthalates and lipid compounds collected at low- to mid-latitudes of the regions in the western North Pacific Ocean (i.e., the Yellow Sea, the East China Sea and the Eastern Taiwan waters). The emission sources of marine aerosols are identified and estimated using biomarkers, diagnostic parameters as well as factor analysis methods. The influence of long-range transport of continental aerosols on the marine atmosphere is also discussed. Our results provide useful information on better understanding the significant role of Asian continental outflow in the western North Pacific Ocean atmospheric aerosols.

2. Experiments and methods

2.1. Sample collection

Total suspended particles (TSP) were collected on pre-combusted (450 °C for 6 h) quartz fiber filters (20 cm × 25 cm, Pallflex) using a high-volume air sampler (Kimoto, Japan) at an airflow rate of 0.8 m³ min⁻¹ on board of the KEXUE-1 Research Vessel during a National Natural Science Foundation of China (NSFC) sharing cruise. Specific sampling site information is presented in Fig. 1. The sampler was mounted on the upper deck of the vessel with a wind speed/wind sector controller to avoid potential contamination from ship exhausts by shutting down the sampling pump during low wind speed (<5 m s⁻¹). The marine aerosol samples were collected from May 18 to June 12, 2014. On the whole, fifty-one daytime/nighttime aerosol samples were obtained during this campaign with the exception of blank ones. All the samples were stored at -20 °C before analysis.

2.2. Sample extraction and derivatization

Aliquots of the filter samples were ultrasonically extracted with dichloromethane/methanol mixture (2:1, v/v) for 10 min three times at room temperature. The solvent extracts were filtered through a Pasteur pipette packed with quartz wool, and concentrated using a rotary evaporator, finally dried in a pure nitrogen air stream. The dried extracts were reacted with 50 µL of *N,O*-bis-(trimethylsilyl)trifluoroacetamide (BSTFA) with 1% trimethylsilyl chloride containing 10 µL of pyridine at 70 °C for 3 h to make polar groups (e.g., COOH and OH) derivatized to the corresponding trimethylsilyl (TMS) esters and ethers, individually (Fu et al., 2008; Simoneit et al., 2004; Wang and Kawamura, 2005). After reaction, 140 µL internal standard (C₁₃ *n*-alkane, 1.43 ng µL⁻¹) was added to the derivatives prior to gas chromatography/mass spectrometry (GC/MS) injection. The field and laboratory blank filters were processed with the same method as the real samples for quality assurance.

2.3. Gas chromatography/mass spectrometry

In this study, five fractions (*n*-alkanes, fatty acids, fatty alcohols, PAHs and phthalates) were analyzed using an Agilent model 7890 GC coupled to an Agilent model 5975C mass-selective detector (MSD).

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