



Non-polar organic compounds in marine aerosols over the northern South China Sea: Influence of continental outflow



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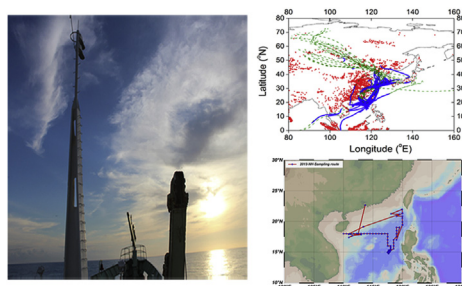
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HIGHLIGHTS

- A total of 115 NPOCs were detected in the marine aerosols over the northern South China Sea.
- Biomass burning in southern/southeastern China was the major source for NPOCs in marine aerosols in the open sea area.
- Fossil fuel combustion was the considerable source of NPOCs in marine aerosol samples with strong continental influence.
- Terrestrial plant contributed to the higher molecular weight *n*-alkanes in marine aerosols.

GRAPHICAL ABSTRACT



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ABSTRACT

Filter samples of total suspended particle (TSP) collected during a cruise campaign over the northern South China Sea (SCS) from September to October 2013 were analyzed for non-polar organic compounds (NPOCs) as well as organic carbon (OC), elemental carbon (EC) and water-soluble ions. A total of 115 NPOCs species in groups of *n*-alkanes, polycyclic aromatic hydrocarbons (PAHs), iso-/antiso-alkanes, hopanes, steranes, methylalkanes, branched alkanes, cycloalkanes, alkenes and phthalates were detected. The characteristics of NPOCs in marine TSP samples were investigated to understand the sources from the Asian continent and other regions. The concentrations of total NPOCs ranged from 19.8 to 288.2 ng/m³ with an average of 87.9 ng/m³, which accounted for 0.8–1.7% (average 1.0%) of organic matter (OM). *n*-Alkanes was the predominant group, accounting for 43.1–79.5%, followed by PAHs (5.5–44.4%) and hopanes (1.6–11.4%). We found that primary combustion (biomass burning/fossil fuel combustion) was the dominant source for the majority of NPOCs (89.1%). Biomass burning in southern/southeastern China via long-range transport was proposed to be a major contributor of NPOCs in marine aerosols over the

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Hopanes
Biomass burning
Long-range transport

northern SCS, suggested by the significant correlations between nss-K⁺ and NPOCs groups as well as the analysis of air mass back-trajectory and fire spots. For the samples with strong continental influence, the strong enhancement in concentrations of *n*-alkanes, PAHs, hopanes and steranes were attributed to fossil fuel (coal/petroleum) combustion. In addition, terrestrial plants waxes were another contributor to NPOCs.

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1. Introduction

Marine aerosol is one of the most important natural aerosol systems globally (O'Dowd and De Leeuw, 2007). It has been reported that marine aerosol can play an important role in global and regional climate directly by back scattering solar irradiation and indirectly via forming cloud condensation nuclei (CCN) (Meskhidze and Nenes, 2006). Generally, aerosol is not only the main route for marine species transferring to the continent but also a major channel for terrestrial species transporting to the ocean (Duce et al., 1991). Hawkins et al. (2010) highlighted that the aerosols in the marine boundary layer were mostly mixtures of local primary (e.g., sea salt and organic compounds from sea spray), local secondary (e.g., sulfate from dimethyl sulfide oxidation), and long-range transport products. In recent years, the strong influence of continental emissions (both natural and anthropogenic) in the atmospheres of coastal and open ocean areas has been observed (Ding et al., 2007, 2013; Fu et al., 2011; Kunwar and Kawamura, 2014). Measurements of organic tracers can support more accurate source apportionment. Non-polar organic compounds (NPOCs) such as *n*-alkanes, polycyclic aromatic hydrocarbons (PAHs) and hopanes are organic compounds of special interest because they are ubiquitous in the terrestrial atmosphere and emitted from well-known sources including anthropogenic (e.g., fossil fuel combustion, petroleum residues and biomass burning) and biogenic emissions (e.g., higher plant leaf waxes) (Simoneit, 1989, 2002; Simoneit et al., 1991). Therefore, the variation and composition of NPOCs could be used to trace the contribution of different sources and to understand the influence of how much continental emissions could influence marine aerosols.

The South China Sea (SCS) is a marginal sea surrounded by fast-developing regions. Anthropogenic pollutants and natural products could be introduced into the atmosphere over the SCS through long-range transport (Zhang et al., 2007; Lai et al., 2010). Their chemical compositions including water-soluble ions, elements and organophosphate in airborne particles have been studied (Hsu et al., 2007; Zhang et al., 2007; Lai et al., 2015). However, with the exception of one study which reported the results of NPOCs in a sample collected in a round-the-world cruise passing through the SCS (Fu et al., 2011), very little research has been conducted to study NPOCs in marine aerosol particles in this region.

Here we present the results of NPOCs in total suspended particle (TSP) samples collected over the northern SCS during a cruise campaign. With this dataset we demonstrate that biomass burning in southern/southeastern China can influence marine aerosols over the SCS via long range transport in the open sea samples while other continental sources are also discussed.

2. Method

2.1. Sample collection

TSP samples ($n = 10$) were collected in the northern SCS during a 20-d cruise campaign from September to October 2013 (Fig. A1). A

high volume aerosol sampler ($\sim 1 \text{ m}^3/\text{min}$, Tisch, USA) was placed in the upper deck of the research vessel. One filed blank filter was obtained. All samples were collected onto pre-baked ($550 \text{ }^\circ\text{C}$) quartz fiber filters (8×10 inches, Pall MI, USA). After sampling, the filters were sealed with prebaked aluminum foil and stored in a freezer at $-20 \text{ }^\circ\text{C}$.

2.2. Chemical analysis

2.2.1. Non-polar organic compounds

An in-injection port thermal desorption-gas chromatography/mass spectrometry (TD-GC/MS) method was used to determine NPOCs including *n*-alkanes, iso-/antiso-alkanes, methylalkanes, branched alkanes, cycloalkanes, alkenes, PAHs (parent, alkylated- and oxy-PAHs), hopanes, steranes and phthalates. This method has less contamination from solvent impurities and higher sensitivity compared to solvent extraction (SE) followed by GC/MS. The detailed description of this method has been presented in previous publications (Ho et al., 2008, 2011). Briefly, for each sample, a filter aliquot (1.0 cm^2) was cut into pieces and inserted into the TD glass tubes for placement into the split/splitless injector liner of an Agilent 6890 GC/5975 MS detector (Santa Clara, CA, USA). The temperature of the injector port was set at $50 \text{ }^\circ\text{C}$ before analysis and then manually raised to $275 \text{ }^\circ\text{C}$ for desorption in a splitless mode. During this period, the temperature in GC oven was kept at $30 \text{ }^\circ\text{C}$. After the injector temperature reached the set point, the oven temperature program started. The limits of detection (LODs) for the TD-GC/MS method are listed in Table A1.

2.2.2. Carbonaceous fractions and water-soluble ions

Organic carbon (OC) and elemental carbon (EC) were analyzed (punch of 1.0 cm^2) using the thermo-optical transmittance (TOT) method (NIOSH protocol) (Sunset, OR, USA) (Birch and Cary, 1996).

Water-soluble ions were detected (punch of 4.0 cm^2) by ion chromatography (Dionex ICS3000, Thermo Fisher, MA, USA). Briefly, the cut portion was extracted in 3 mL ultra-pure water. A CS12 column ($4 \times 250 \text{ nm}$) with 20 mM methanesulfonic acid (MSA) eluent and AS11-HC column ($4 \times 250 \text{ nm}$) with 30 mM NaOH eluent were used for the determination of 5 cations (i.e. Na⁺, NH₄⁺, K⁺, Mg²⁺ and Ca²⁺) and 5 anions (i.e. MSA⁻, Cl⁻, SO₄²⁻, C₂O₄²⁻ and NO₃⁻), respectively. The LODs are shown in Table A2.

2.3. Quality assurance/quality control (QA/QC)

Contamination from the ship exhaust was prevented by manually shutting off the sampling pump when the ship stopped or was under unfavorable wind conditions. The contamination from the ship exhaust is confirmed to be negligible based on the previous publication (Lai et al., 2015). All the data reported here were corrected for the field blank.

2.4. Air mass back-trajectory and fire spot analysis

Air mass back-trajectory analysis was performed along the

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