

Atmospheric polycyclic aromatic hydrocarbons observed over the North Pacific Ocean and the Arctic area: Spatial distribution and source identification

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Abstract

During the 2003 Chinese Arctic Research Expedition from the Bohai Sea to the high Arctic (37–80°N) aboard the icebreaker Xuelong (Snow Dragon), air samples were collected using a modified high-volume sampler that pulls air through a quartz filter and a polyurethane foam plug (PUF). These filters and PUFs were analyzed for particulate phase and gas phase polycyclic aromatic hydrocarbons (PAHs), respectively, in the North Pacific Ocean and adjacent Arctic region. The \sum PAHs (where \sum = 15 compounds) ranged from undetectable level to 4380 pg m^{-3} in the particulate phase and 928–92 600 pg m^{-3} in the gas phase, respectively. A decreasing latitudinal trend was observed for gas-phase PAHs, probably resulting from temperature effects, dilution and decomposition processes; particulate-phase PAHs, however, showed poor latitudinal trends, because the effects of temperature, dilution and photochemistry played different roles in different regions from middle-latitude source areas to the high latitudes. The ratios of PAH isomer pairs, either conservative or sensitive to degradation during long-range transport, were employed to interpret sources and chemical aging of PAHs in ocean air. In this present study the fluoranthene/pyrene and indeno[123-cd]pyrene/benzo[ghi]pyrene isomer pairs, whose ratios are conservative to photo-degradation, implies that biomass or coal burning might be the major sources of PAHs observed over the North Pacific Ocean and the Arctic region in the summer. The isomer ratios of 1,7/(1,7+2,6)-DMP (dimethylphenanthrene) and anthracene/phenanthrene, which are sensitive to aging of air masses, not only imply chemical evolving of PAHs over the North Pacific Ocean were different from those over the Arctic, but reveal that PAHs over the Arctic were mainly related to coal burning, and biomass burning might have a larger contribution to the PAHs over the North Pacific Ocean.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are formed by incomplete combustion or pyrolysis of materials containing carbon and hydrogen. They have both anthropogenic and natural sources. The former includes combustion processes of fossil fuels and biomass burning, and the release of uncombusted petroleum products; the latter includes natural fires and diagenetic processes (Simoneit, 1984). As a group of semi-volatile organic compounds (SVOCs), PAHs are ubiquitous and detected in a variety of environmental media, including air (Halsall et al., 1994; Buehler et al., 2001), soil (Motelay-Massei et al., 2004), sediment (Laflamme and Hites 1978; Mai et al., 2003) and biological samples (Page et al., 2004). Due to their lipophilicity and hydrophobicity, PAHs may partition and accumulate in organisms and thus have carcinogenic and mutagenic activity (Okona-Mensah et al., 2005; Perera et al., 2005).

The occurrence and sources of PAHs in the atmosphere have been widely studied. In most urban and rural areas, airborne PAHs originate mainly from automobile exhausts and/or combustion of fossil fuel (Mai et al., 2003; Ohura et al., 2004), while in remote areas, such as in open ocean air and the Arctic, both biomass burning and fossil fuel combustion may contribute to PAHs. Crimmins et al. (2004) analyzed particulate PAHs, organic carbon (OC), elemental carbon (EC), SO_4^{2-} , and K^+ in the atmosphere of Atlantic and Indian Ocean. They pointed out that fossil fuel combustion, rather than biomass burning, was the predominant source of PAHs in the Northern Hemisphere Indian Ocean atmosphere. However, few studies have been conducted in the North Pacific Ocean and the adjacent Arctic Ocean. In the Arctic, Daisey et al. (1981) measured particulate organic matter (POM), PAHs, and ^{210}Pb in the Arctic aerosol in Barrow, Alaska. They found that the principal source of PAHs was fossil fuel combustion in the mid-latitude regions in winter from the presence of fly ash and seasonal changes in organic species, ^{210}Pb and meteorology.

Seasonal changes in meteorology are key factors in determining the chemical composition of the Arctic atmosphere. In winter, the pollutants in Eurasia and North America can move into the Arctic region easily and rapidly by atmospheric transportation. In the Arctic haze, air mass back trajectories (BTs) analysis for February 1994

revealed long-range transport from Eurasia into the high Arctic (Halsall et al., 1997). In summer the meteorological barrier, however, will make the influence of northward transport from low latitudes weaker. Thus the variety of meteorology may lead to the seasonal change of sources of PAHs in the Arctic atmosphere. Slater et al. (2002) reported PAHs and radiocarbon (^{14}C) of EC in water-insoluble particles from a snow pit excavated at Summit, Greenland in 1996. They used the ratios of BghiP/BeP and Ret/BeP to quantify the relative contribution of fossil fuel and biomass combustion, and ^{14}C measurements to distinguish EC between the two sources. Results showed that fossil fuel combustion was dominant in spring, and biomass burning impacted deeply in fall to early winter, while both sources had influences in summer.

In July to September 2003, ship-board air samples were collected along a cruise from Bohai Sea to the North Pole Area (37–80°N). Detailed information about this expedition has been described elsewhere (Wang et al., 2005). In the present study, the purposes are to determine the spatial distribution of PAHs from mid-latitude source areas to the high Arctic, and to provide information about the long-range atmospheric transport (LRAT) and possible sources of PAHs in the North Pacific Ocean and adjacent Arctic Ocean during warm periods.

2. Experimental section

2.1. Field sampling

A total of 49 particle samples, 30 of which had vapor samples collected simultaneously, were taken aboard the expedition cruise from Bohai Sea (37.78°N, 123.12°E) to the Arctic (80.22°N, 146.75°W) between 11 July–21 September 2003. The sampling route is shown in Fig. 1. A modified high volume air sampler was placed windward on the uppermost front deck of the ship for simultaneous sampling of PAHs in particulate and gas phases. Air was drawn at a flow rate of $\sim 1.0\text{ m}^3\text{ min}^{-1}$ through a quartz fiber filter (QFF) (Whatman QM-A, 20.3 × 25.4 cm) to collect total suspended particles (TSP) from the air stream, followed by one-and-a-half polyurethane foam plug (PUF, 6.5 cm diameter × 8 cm height) contained in an aluminum cylinder to trap the gas-phase compounds. The sampled air volumes ranged from 1215 to 3030 m^3 (at 0°C and 1 atm). The back half PUF served as an indicator of breakthrough of

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