



Polychlorinated naphthalenes in the air over the equatorial Indian Ocean: Occurrence, potential sources, and toxicity



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ABSTRACT

Monitoring of marine polychlorinated naphthalenes (PCNs) is crucial, as they are considered persistent organic pollutants (POPs) by the Stockholm Convention. Data on PCNs in marine environment are scarce. In this study, 19 air samples were collected during a cruise in the equatorial Indian Ocean on board the Chinese research vessel *Shiyan I* from 4/2011 to 5/2011. PCN concentration of these air samples ranged from 0.033 to 2.56 $\mu\text{g m}^{-3}$, with an average of 0.518 $\mu\text{g m}^{-3}$, equal to or lower than the values reported for other oceans, seas, and lakes worldwide. Tri- and tetra-CNPs were the main homologues in most samples. Reemission of Halowax mixtures and incineration processes were the major sources of atmospheric PCNs in the study area. The PCN-corresponding toxic equivalency values ranged from 0 to 0.190 fg m^{-3} (average: 0.038 fg m^{-3}), falling in the low end of global range.

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1. Introduction

Polychlorinated naphthalenes (PCNs) are considered as persistent organic pollutants (POPs) by the Persistent Organic Pollutants Review Committee of United Nations, because of their persistence, long-range transportability, bioaccumulation, and adverse effects on organisms (SCPOP, 2011). PCN congeners are ubiquitous in various environmental media and biota (Bidleman et al., 2010). Technical PCN products had been widely used as insulators, lubricants, flame retardants, paint additives, and so on, until the 1970s/1980s (Falandysz, 1998; Hogarh et al., 2012). Evaporation from the historical residue has been a major pathway of PCNs in air (Harner and Bidleman, 1997). Recently, unintentional emissions of PCNs from combustion processes such as municipal waste incineration and metal smelting (including primary and secondary non-ferrous smelting, coking, iron ore sintering, and electric arc furnaces) have been considered more important than ever to atmospheric PCNs (Wang et al., 2012; Liu et al., 2014, 2015).

Oceans and seas are the final sinks of POPs, removing them from the “recyclable pool” in the environment (Dachs et al., 2002). However, oceans might also turn into a secondary source of certain POPs such as dichlorodiphenyltrichloroethane (Stemmler and Lammel, 2009),

alpha-hexachlorocyclohexane (Li and Bidleman, 2003), and polychlorinated biphenyls (PCBs, Zhang and Lohmann, 2010) long after those compounds were banned. Thus, it is important to monitor marine POP pollution status and estimate their sources. Several studies have been carried out to investigate PCN levels in air, sediments, or organisms of the arctic and subarctic oceans and seas (Falandysz and Rappe, 1996; Harner et al., 1998; Lundgren et al., 2002, 2003; Bidleman et al., 2010).

The equatorial Indian Ocean is surrounded by many South Asian countries, including India, Pakistan, Malaysia, Sri Lanka, Vietnam, and the Philippines. Recent studies in India and Pakistan have indicated that atmospheric PCN pollution in South Asia is comparable to or higher than other Asian regions, and is high worldwide (Mahmood et al., 2014; Xu et al., 2014). Our previous study suggested that atmospheric PCNs in the northern South China Sea were influenced by the proximity to source regions and air mass origins, and wood/coal burning and industrial processes were the two major combustion sources of the gaseous PCNs (Li et al., 2012). A similar condition may occur in the equatorial Indian Ocean. However, data on PCNs in the equatorial and adjacent oceans and seas are scarce.

A total of 19 air samples were taken from the equatorial Indian Ocean during the sampling campaign from Malaysia to the south of Bay of Bengal passing by Sri Lanka in 2011 on board the Chinese research vessel *Shiyan I*. In order to understand PCN pollution status in

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the equatorial Indian Ocean, PCN concentrations in the air samples were determined. The composition profiles, spatial distribution, potential sources, and toxicity of PCNs in the air are also analyzed.

2. Materials and methods

2.1. Sampling

On board the Chinese research vessel *Shiyan I* of South China Sea Institute of Oceanology, from 12 April to 4 May 2011, 19 air samples were taken from the equatorial Indian Ocean (0–6°E, 80–98.5°N, Fig. 1, Table S1). These samples were taken via a high-volume air sampler placed in the windward on the foredeck of the ship. Approximately 200–500 m³ of air was drawn through a quartz fiber filter (QFF), and subsequently through a polyurethane foam (PUF) plug (i.d.: 6.5 cm, length: 7.5 cm, density: 0.03 g m⁻³). The PUFs and QFFs were stored at -20 °C until analysis.

2.2. Sample preparation and instrumental analysis

The PUF and QFF samples spiked with ¹³C-PCB141 as recovery surrogate were Soxhlet-extracted with dichloromethane (DCM) for 36 h and further purified on a column packed with 3-cm neutral alumina (3% deactivated), 3-cm neutral silica gel (3% deactivated), 3-cm sulfuric acid silica gel (50% by mass), and 1-cm anhydrous Na₂SO₄ from bottom to top (Huang et al., 2013). The column was eluted with 30 mL of DCM/hexane (1:1) to collect the PCN fraction. The eluted solvent was concentrated and solvent-exchanged to hexane, and then concentrated to approximately 50 μL. PCB208 was added as internal standard before instrumental analysis.

The target PCN congeners were measured on an Agilent 7890A/5975C GC-MS with a DB-5 ms capillary column (30 m × 0.25 mm × 0.25 μm, Agilent, CA, USA). The mass spectrometer was operated with an electron capture negative-ion (ECNI) source in selected ion monitoring (SIM) mode. The initial temperature of the oven

was set at 80 °C for 0.5 min, then increased at a rate of 15 °C min⁻¹ to 160 °C, 2 °C min⁻¹ to 240 °C, and finally 6 °C min⁻¹ to 270 °C and held for 2.5 min. The temperatures of the injector interface, transfer line, and ion source were set at 265, 280, and 150 °C, respectively. Tri- to octa-CN congeners (tri-CN congeners: CN19, 24, 14, 15, 16, 17/25, 23; tetra-CN congeners: CN42, 33/34/37, 47, 36/45, 28/43, 27/30, 39, 32, 35, 38/40, 46; penta-CN congeners: CN52/60, 58, 61, 50, 51, 54, 57, 62, 53, 59, 49, 56; hexa-CN congeners: CN66/67, 64/68, 69, 71/72, 63, 65; hepta-CN congeners: CN73, 74; octa-CN congener: CN75) were quantified against the technical PCN mixture Halowax 1014 (Xu et al., 2014).

2.3. Quality assurance and quality control

Two laboratory blanks, two PUF field blanks, and three QFF field blanks were analyzed. The surrogate recoveries of ¹³C-PCB141 were 97 ± 7.2% and 78.1 ± 23.3% for PUF and QFF samples, respectively. The reported PCN concentrations in this study were not recovery-corrected. The instrumental detection limits (IDLs) were calculated from the lowest standard, extrapolating to the corresponding amount of analyte that would generate a signal-to-noise ratio of 3:1. The method detection limits (MDLs) were assigned as average values of blanks plus three times of the standard deviations of blank values. Because target PCN congeners were below the IDLs in the laboratory and field blanks analyzed, the MDLs of measured PCNs were assigned as three times of IDLs, with values ranging from <0.001 to 0.021 pg m⁻³.

Breakthrough of PUF samples was assessed by two consecutive PUF plugs, which were analyzed separately. Target PCN congeners were under MDLs from the subjacent PUF plug, suggesting that these compounds could be effectively collected by the upper PUF plug.

2.4. Air mass back trajectories

NOAA's (National Oceanic and Atmospheric Administration) HYSPLIT model was used to trace the air mass back trajectories of the air samples collected along the sampling cruise (Draxler and Rolph,

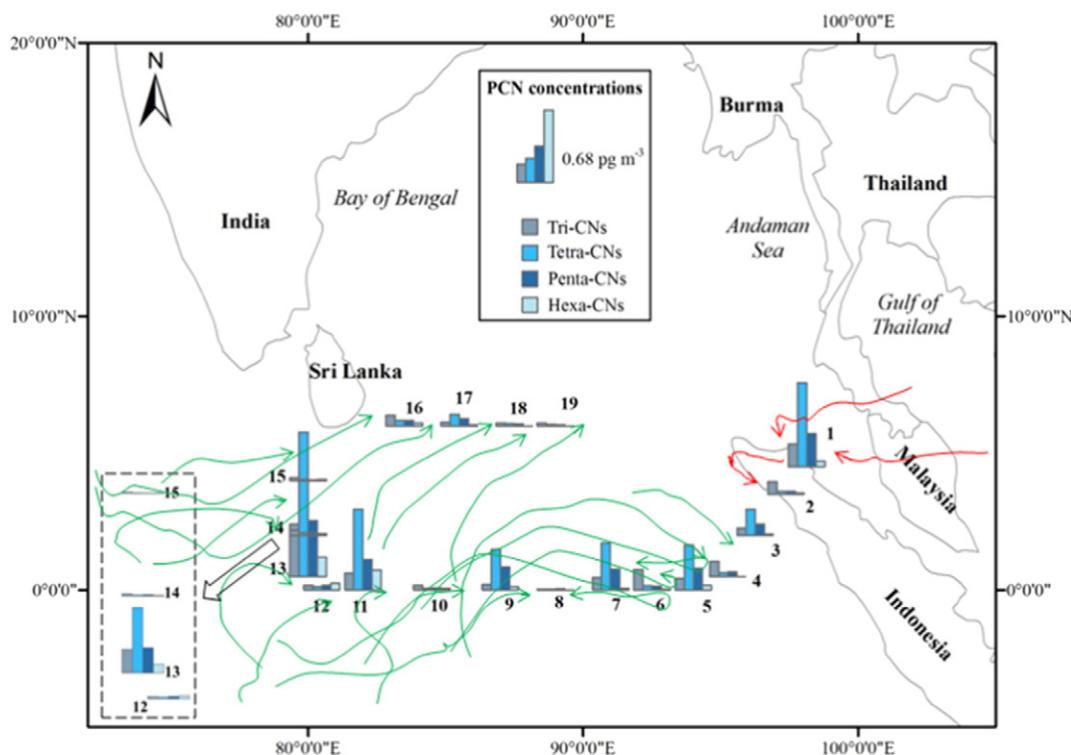


Fig. 1. Atmospheric concentrations of PCN homologues and the air mass back trajectories (BTs) during sampling campaign. Three BTs were continental passing over Malaysia and Indonesia (red lines), and the remaining BTs were oceanic passing over the Indian Ocean (green lines).

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