Polychlorinated biphenyls (PCBs) and hexachlorobenzene (HCB) in the equatorial Indian Ocean: Temporal trend, continental outflow and air–water exchange

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A R T I C L E   I N F O

A B S T R A C T

Nineteen pairs of air and seawater samples collected from the equatorial Indian Ocean onboard the Shiyan I from 4/2011 to 5/2011 were analyzed for PCBs and HCB. Gaseous concentrations of $\sum_{ICES}$PCBs (ICES: International Council for the Exploration of the Seas) and HCB were lower than previous data over the study area. Air samples collected near the coast had higher levels of PCBs relative to those collected in the open ocean, which may be influenced by proximity to source regions and air mass origins. Dissolved concentrations of $\sum_{ICES}$PCBs and HCB were 1.4–14 pg L^{-1} and 0.94–13 pg L^{-1}, with the highest concentrations in the sample collected from Strait of Malacca. Fugacity fractions suggest volatilization of PCBs and HCB from the seawater to air during the cruise, with fluxes of 0.45–34 ng m^{-2} d^{-1} and 0.36–18 ng m^{-2} d^{-1}, respectively.

1. Introduction

Polychlorinated Biphenyls (PCBs) are typical persistent organic pollutants (POPs) of worldwide concern because of their persistence, long-range transport ability, bioaccumulation, and negative effects on people and wildlife. PCBs were finally banned in the late 1970s/early 1980s (de Voogt and Brinkman, 1989) but are still ubiquitous in the environment (Gioia et al., 2011). Deep oceans are considered as a final sink for POPs, removing them from the "recyclable pool" in the environment (Dachs et al., 2002). Net deposition of PCBs has been observed in the North Atlantic and Arctic Oceans (Gioia et al., 2008a,b). However, volatilization of PCBs from the ocean to lower air has also been estimated in the open Pacific Ocean (Zhang and Lohmann, 2010) and tropical Atlantic Ocean (Lohmann et al., 2012), suggesting oceans could also act as a secondary source of atmospheric PCBs in the environment.

The equatorial Indian Ocean is surrounded by tropical developing countries, which have received large amounts of e-waste, like India, Pakistan, Malaysia, Vietnam and the Philippines (Robinson, 2009). India, Pakistan and Bangladesh continue to release PCBs to the Indian Ocean from the ship-breaking industry, with up to 0.25–0.8 metric tons of PCBs generated per scrapped merchant ship (Wurl et al., 2006). The air–water exchange process in the oceans is of great importance in controlling the environmental fate of POPs, and learning about sources of POPs in the adjacent countries (Iwata et al., 1993). The study of air–water exchange in the equatorial Indian Ocean could date back to Iwata et al. (1993), who suggested positive-to-negative range of fluxes for PCBs in the Bay of Bengal and Arabian Sea.

The aims of the present study are to: (1) update the data on the concentrations of PCBs and HCB in the air and surface water phase in the equatorial Indian Ocean, (2) discuss the spatial distribution and possible sources of PCBs, and (3) estimate the directions and fluxes of air–water exchange of PCBs and HCB in the target region. The study is based on 19 pairs of lower air and surface seawater samples that were collected from the equatorial Indian Ocean in 2011 onboard the Chinese research vessel Shiyan I.

2. Materials and methods

2.1. Sample collection

Onboard the Shiyan I of South China Sea Institute of Oceanology, 19 pairs of air and surface seawater samples were taken from the equatorial Indian Ocean (0–6°E, 80–98.5°N, Fig. 1, Table S1) from April 12th to May 4th, 2011. Air samples were taken via a

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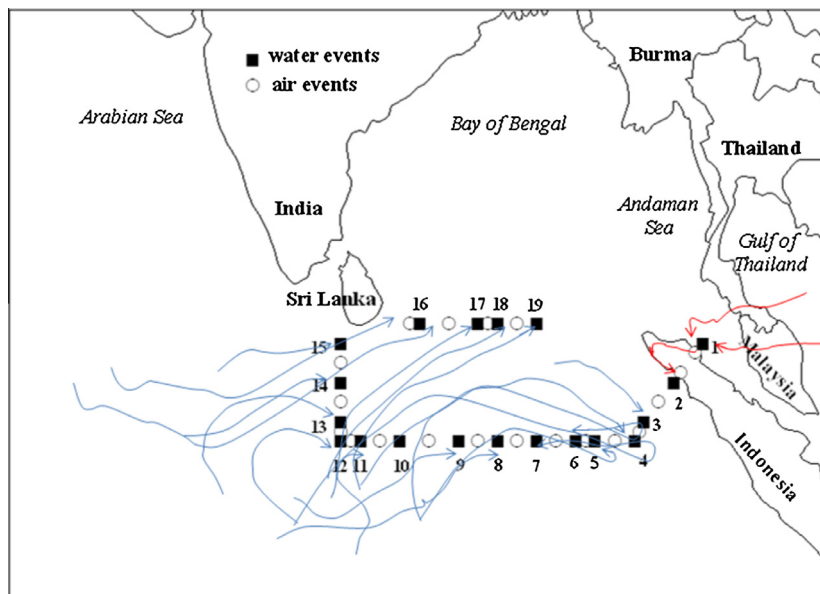E-mail address: junli@gig.ac.cn (J. Li).

Fig. 1. Sampling locations and air mass backward trajectories (BTs). Three BTs were continental from Malaysia and Indonesia (red lines), and the remaining BTs were oceanic from the Indian Ocean (blue lines). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

high-volume air sampler placed in the windward side on the fore-deck of the ship. About 200–500 m³ of air was drawn through a quartz fiber filter and subsequently through a polyurethane foam (PUF) plug. At each site, about 70–120 L of surface seawater was collected using a seawater-cleaned metal bucket and filtered through a precombusted (4 h at 450 °C) 150 mm diameter glass fiber filter (Gelman Type A/E, nominal pore size: 1 μm). Then the water sample was passed through a Teflon column (i.d.: 25 mm, length: 20 mm) packed with pretreated resin (XAD-2 and XAD-4, 1:1 v/v). All the samples (filters, PUFs and Teflon columns) were stored at −20 °C until analysis.

2.2. Sample preparation

The PUF samples spiked with recovery surrogates (TCmX, PCB209 and ^{13}C-PCB141) were Soxhlet extracted with dichloromethane (DCM) and further purified on a multilayer silica gel-alumina 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 Na_2SO_4 from bottom to top. 30 mL of DCM/hexane (v:v 1:1) were eluted to collect the PCB and HCB fraction. The XAD resin spiked with the above surrogates was ultrasonically extracted with methanol (3 × 50 mL) and DCM (3 × 50 mL). The extract was combined with 200 mL saturated NaCl solution, further extracted using DCM (3 × 50 mL), dried with 15 g anhydrous Na_2SO_4, concentrated and purified as the PUF samples. Concentrated fraction was solvent exchanged to hexane, further concentrated to a volume of about 50 μL under subtle nitrogen. ^{13}C-PCB138 was added as an internal standard for instrumental analysis. Before use, neutral silica gel and neutral alumina was each Soxhlet-extracted with DCM for 48 h, and baked for 12 h at 180 °C and 250 °C, respectively. Anhydrous Na_2SO_4 was baked at 450 °C for 12 h before use. Filter samples were not analyzed in this study.

2.3. Instrumental analysis

Twenty-one types of PCBs (PCB18, 28, 44, 52, 66, 77, 81, 101, 105, 114, 118, 123, 126, 128, 138, 153, 156, 157, 167, 169 and 180) and HCB were measured on an Agilent 7890/7000 GC–MS/MS with a HP-5MS capillary column (30 m × 0.25 cm × 0.25 mm, Agilent, CA, USA). The mass spectrometer was operated with an EI source (−70 eV) in multiple reaction monitoring (MRM) mode. Specific MRM parameters for the target compounds are shown in Table S2. The temperatures of the transfer line, injector interface, and ion source were set at 280 °C, 250 °C, and 230 °C, respectively. The GC oven temperature was set at 80 °C for 0.5 min, then 20 °C min⁻¹ to 160 °C, 4 °C min⁻¹ to 240 °C, and finally 10 °C min⁻¹ to 295 °C, and hold for 10 min. 1 μL of sample was injected in splitless mode. Helium was used as carrier gas (1 mL min⁻¹). Collision induced dissociation (CID) gas and quench gas in collision cell were nitrogen and helium at the flow rate of 1.5 mL min⁻¹ and 2.25 mL min⁻¹, respectively. The scan time ranged from 0.234 s to 0.255 s for all the segments.

2.4. QA/QC

Three laboratory blanks, four PUF field blanks and two XAD field blanks were analyzed. For air samples, the recoveries were 97 ± 7.2%, 63 ± 13%, and 110 ± 11% for ^{13}C-PCB141, TCmX, and PCB 209, respectively. For water samples, the equivalent values were 97 ± 10%, 71 ± 9.5%, and 100 ± 18%, respectively. The method detection limits (MDLs) of the target PCBs and HCB ranged from 0.02 to 0.08 pg m⁻³ for the air samples, and from 0.05 to 0.25 pg L⁻¹ for the water samples. The target compounds were under the MDLs in the laboratory blanks. Trace amounts of PCB18, 28, 44, 52 and HCB were detected in the PUF field blanks (0.03–0.12 pg m⁻³), and 0.10–0.32 pg L⁻¹ of PCB18, 28, 77, 81, 118, and HCB were detected in the XAD field blanks. The reported PCB concentrations in the air and seawater samples in this study were corrected by deducting the average values in PUF and XAD field blanks, respectively, and were not recovery corrected.

2.5. Air mass back trajectories (BTs)

In order to assess the possible sources of PCBs and HCB in the lower air samples, air mass origins along the sampling segments were analyzed using the NOAA's HYSPLIT model (Draxler and Rolph, 2003). 5-day back trajectories were calculated at the beginning

and end of each sampling event in coordinated universal time (UTC) at 10 m above sea level. The results are shown in Fig. 1.

3. Results and discussion

3.1. Concentrations in the air and temporal trend

PCBs: Table 1 summarizes the results of air and seawater PCB concentrations. The sum of the 21 measured PCBs ($\sum_{21}$PCBs) in the air ranged from 2.0 to 29 pg m^{-3}. Tri- and tetra-PCBs dominated and constituted 64 ± 21% and 32 ± 22%, respectively, of the $\sum_{21}$PCBs. The concentrations of $\sum_{ICES}$PCBs (PCB 28, 52, 101, 118, 138, 153, and 180, identified by the International Council for the Exploration of the Seas (ICES) as markers of the degree of contamination) were 1.4–22 pg m^{-3} and contributed 64–77% to the total PCBs. PCB28 is the predominant congener in most samples, with concentrations ranging from 1.2 to 11 pg m^{-3}, except for air samples No. 2 and 3, in which PCB 52 was the most abundant congener with concentration of 22 pg m^{-3} and 15 pg m^{-3}, respectively. Concentrations of higher chlorinated PCB101, 118, 138 and 153 were below 1 pg m^{-3} in all samples. PCB138 was only detected in four samples. PCB180 and the nine dioxin-like PCB congeners (PCB105, 114, 123, 126, 128, 156, 157, 167 and 169) were under detection limit in all air samples.

The concentration range of $\sum_{ICES}$PCBs in this study was lower than the values reported over the North Pacific Ocean in 2008 (28–103 pg m^{-3}) (Zhang and Lohmann, 2010), and the open Atlantic in 2001 (4.5–120 pg m^{-3}) (Jaward et al., 2004) and in 2005 (3.7–220 pg m^{-3}) (Gioia et al., 2008a), and was similar to the measurements by Gioia et al. (2008b) over the Arctic (range: 0.76–43 pg m^{-3}, mean: 7 pg m^{-3}) and Zhang and Lohmann (2010) over the South Pacific Ocean (1.5–36 pg m^{-3}).

Reported data on PCB concentrations in the equatorial Indian Ocean are limited. The latest data showed that the concentration of $\sum_7$PCBs (sum of PCB31/28, 52, 90/101, 118, 153/132, 138, and 180) ranged from 5 to 73 pg m^{-3} with an average of 30 pg m^{-3} in the air over the Andaman Sea and Bay of Bengal in 2008 (Gioia et al., 2012), which were much higher than those measured in this study. Wurl et al. (2006) observed concentrations 6.8–16.1 pg m^{-3} for $\sum_{38}$PCBs in two air samples collected in the study area in 2005, with PCB 18, 44, 49, 52, 70, 74, 87, 95, 101 and 110 + 82 being the major congeners. Substantially higher concentrations of $\sum_{40}$PCBs (19–710 pg m^{-3}) were observed in the air over the Bay of Bengal and Arabian Sea in 1989–1990 (Iwata et al., 1993). Comparison with these data sets should be treated cautiously because different PCB congeners were measured in different studies. However, since low chlorinated PCBs are the dominant congeners in these studies, the comparison could indicate a significant decrease of PCB concentrations in the air over the equatorial Indian Ocean since

1990s. This may be related to the ban history of PCBs in the adjacent countries (UNEP, 2002). In addition, the variations of PCB concentrations measured in those studies over the same areas may be affected by the origin of air masses when the samples were collected. For example, continental air masses related to land outflow may give rise to the gaseous PCB levels in the Bay of Bengal and Arabian Sea in 1989 (Wurl et al., 2006), while the low gaseous PCB levels observed in 2005 may be related to the oceanic air masses (Wurl et al., 2006). Partly oceanic and partly continental air masses arrived when the air samples were collected in this study and in 2008 (Gioia et al., 2012).

HCB: HCB concentrations in the air were 2.2–17 pg m^{-3}, with an average value of 6.2 ± 3.9 pg m^{-3}. The concentration range is similar to those reported over the North Atlantic Ocean (3.6–17.6 pg m^{-3}) (Zhang et al., 2012) and the open Atlantic Ocean (0.8–21.9 pg m^{-3}) (Gioia et al., 2012), but is lower than those in Canadian Arctic Ocean (48–71 pg m^{-3}) (Wong et al., 2011), the eastern Arctic region (23–87 pg m^{-3}) (Lohmann et al., 2009), and the open Pacific (14–89 pg m^{-3}) (Zhang and Lohmann, 2010). Higher concentrations (4.6–38 pg m^{-3}, 24 ± 11 pg m^{-3}) were observed in the air over the Andaman Sea and Bay of Bengal in 2008 (Gioia et al., 2012).

3.2. Concentrations in the seawater

PCBs: $\sum_{21}$PCBs in the surface seawater ranged from 2.7 to 25 pg L^{-1}, with an average value of 7.5 ± 5.4 pg L^{-1}. Similar to the profile in the air samples, tri-and tetra-PCBs dominated, accounting for 58 ± 12% and 33 ± 11%, respectively, of total PCBs. Concentrations of $\sum_{ICES}$PCBs were 1.4–14 pg L^{-1} and its contribution to $\sum_{21}$PCBs (27–66%) is less than that in the air. PCB28 is the predominating congener, with concentrations varying from 0.89 to 10 pg L^{-1}. PCB101, 118, 138 and 153 displayed concentrations lower than 1 pg L^{-1}, and were detectable in 16–95% samples. PCB180 and the other dioxin-like PCBs (PCB 105, 126, 128, 156, 157, 167 and 169) were under detection limit in all samples.

Compared to those reported in other oceans, the $\sum_{ICES}$PCB concentrations in this study were similar to the recent finding in the North Pacific (0.2–15 pg L^{-1}), and higher than that in the South Pacific (0.3–7.8 pg L^{-1}) (Zhang and Lohmann, 2010). Much lower values were observed in the Atlantic (0.017–1.7 pg L^{-1}) (Gioia et al., 2008a) and the Arctic (<1 pg L^{-1}) (Gioia et al., 2008b). Iwata et al. (1993) reported concentrations of 13–46 pg L^{-1} for $\sum_{40}$PCBs in the Bay of Bengal and Arabian Sea in 1989–1990.

HCB: To our knowledge, HCB concentration is reported for the first time in the seawater of the equatorial Indian Ocean, with a range of 0.94–13 pg L^{-1}, which is higher than those in the open Pacific (0.4–1.6 pg L^{-1}) (Zhang and Lohmann, 2010), the South Atlantic (1.9–3.3 pg L^{-1}) (Booij et al., 2007) and the North Atlantic

Table 1
Summary of PCB and HCB concentrations in the air and seawater of the equatorial Indian Ocean.

Compound	Air concentration (pg m^{-3})				Water concentration (pg L^{-1})			
	Minimum	Maximum	Mean	SD	Minimum	Maximum	Mean	SD
PCB28	0.28	11	4.8	2.9	0.89	10	3.2	2.1
PCB52	0.22	22	2.9	5.6	n.d.	1.7	0.24	0.45
PCB101	n.d.	0.55	0.16	0.17	n.d.	0.74	0.28	0.17
PCB118	n.d.	0.20	0.09	0.06	n.d.	0.55	0.11	0.14
PCB153	n.d.	0.34	0.12	0.09	n.d.	0.52	0.10	0.14
PCB138	n.d.	0.21	0.03	0.06	n.d.	0.14	0.02	0.05
PCB180	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
$\sum_{ICES}$PCBs	1.4	22	8.1	5.1	1.4	14	4.0	2.6
$\sum_{21}$PCBs	2.0	29	12	6.9	2.7	25	7.5	5.4
HCB	2.2	17	6.2	3.9	0.94	13	4.0	3.0

n.d.: Not detected.

Ocean (0.1–0.8 pg L^{-1}) (Zhang et al., 2012). Similar concentrations were observed in the eastern Arctic (0.85–9.6 pg L^{-1}) (Lohmann et al., 2009) and a narrower range was observed in the Canadian Arctic regions (4.0–6.4 pg L^{-1}) (Wong et al., 2011).

3.3. Spatial variation and continental outflow

Fig. 2 shows the spatial distribution of $\sum_{ICES}$PCBs and HCB in the air and water phases along the sampling cruise.

Air samples with continental air masses from over the land of Indonesia and Malaysia showed higher levels of PCBs than those collected in the open ocean (Figs. 1 and 2), suggesting continental outflow of PCBs. Although the usage of PCBs has been banned in those countries (UNEP, 2002), the important sources such as older PCB-containing equipments, landfill, and incineration of e-waste are continuing to release PCBs to the environment (Breivik et al., 2007). A recent study suggested that large quantities of e-waste have been exported to Malaysia (Robinson, 2009). Furthermore, accidental release from fires may be a possible source, as PCBs previously deposited in the soil would volatilize when extensive biomass burning taking place in the continents (Eckhardt et al., 2007). High PCB concentration was also observed in the air sample No. 16 collected near Sri Lanka and India (Fig. 2), which may be related to continental outflow from India, where substantially high concentrations have been detected in the air (Pozo et al., 2011; Zhang et al., 2008). For example, $\sum_{ICES}$PCB concentrations higher by a factor of ~30 than this study were measured by Zhang et al. (2008) in Mumbai, Bangalore, and Kolkata with levels of 253, 243, and 239 pg m^{-3}, respectively. Emissions from the intensive ship-breaking activities and storage of obsolete equipment (Zhang et al., 2008), landfill, and incineration of e-waste were important potential sources for PCBs in India (Breivik et al., 2011).

In the water phase, the highest concentration of $\sum_{ICES}$PCBs and HCB was observed in site 1, which is located at the Strait of Malacca between Malaysia and Indonesia. This high concentration may be related to the riverine output of PCBs and HCB from Indonesia or Malaysia. In the remaining sites, small variations were observed, with a range of 1.4–5.5 pg L^{-1} for $\sum_{ICES}$PCBs and 0.94–9.0 pg L^{-1} for HCB, respectively.

3.4. Air–water exchange of PCBs and HCB

3.4.1. Fugacity fractions

Fugacity fraction (ff) was used to assess the air–water exchange direction of PCBs and HCB. ff was calculated according to the following equations (Zhang et al., 2007):

$$f_a = C_a RT \tag{1}$$

$$f_w = C_w H \tag{2}$$

$$ff = f_w/(f_w + f_a) \tag{3}$$

where f_a and f_w are fugacity of chemical in the air and water, C_a and C_w are concentrations of chemical in the air and water (mol m^{-3}), R is the gas constant (8.314 Pa m^3 mol^{-1} K^{-1}), T is the absolute temperature (K), and H is the Henry's law constant (Pa m^3 mol^{-1}). ff = 0.5 indicates air–water equilibrium, ff < 0.5 indicates net deposition to the water, and ff > 0.5 indicates volatilization from the water (Zhang et al., 2007).

The ff values of PCB 18, 28, 44, 52, 66, 101, 118, 153, and HCB were estimated in this study, while ff of other PCB congeners were not calculated because of their low detection frequency. An average temperature value of 303 K in the surface seawater was used for the correction of H values. H values of PCB 28, 52, 101, 118 and 153 were derived from the equation for FAVs (Final Adjusted Values) compiled by Li et al. (2003) and H values of PCB 18, 44 and 66 were taken by Bamford et al. (2002). H of HCB was calculated according to the equation established by Jantunen and Bidleman (2006) and adjusted considering the salting out effect (Su et al., 2006). Uncertainty of 45% was assigned for the ff calculation, based on the error in the measurement of C_a, C_w, and H values (Zhang et al., 2007), which means that ff between 0.275 and 0.725 is considered as not significantly different from air–water equilibrium (Fig. 3).

As shown in Fig. 3, net volatilization of PCBs from the equatorial Indian Ocean was observed at most sampling sites, indicating that the water phase is a secondary source of PCBs in the lower air. Near equilibrium was found for PCB18 and 44 at several sites with ff ranging from 0.375 to 0.725. Net deposition was observed for PCB 52, 66, and 153 at some sampling sites when these compounds were under detection limits in the water phase, with ff value of zero. Volatilization of PCBs was also observed in the open Pacific Ocean during 2006–2007 (Zhang and Lohmann, 2010) and the tropical Atlantic in 2009 (Lohmann et al., 2012). Whereas, Gioia et al. (2008a,b) observed net deposition of PCBs dominated in the North Atlantic and Arctic, and equilibrium of PCBs in the South Atlantic.

With respect to HCB, net volatilization was observed at all sites in this study (Fig. 3), which may be in part due to the high H value (57.4 Pa m^3 mol^{-1}) of HCB used in the study area with high water temperatures. Net volatilization or close to equilibrium was deduced for HCB in the tropical Atlantic north of the equator (Lohmann et al., 2012). Air–water equilibrium or near equilibrium and deposition of HCB was observed in the oceans with much lower water temperatures, such as the North Atlantic (Lohmann et al., 2009; Zhang et al., 2012) and Arctic Ocean (Lohmann et al., 2009;
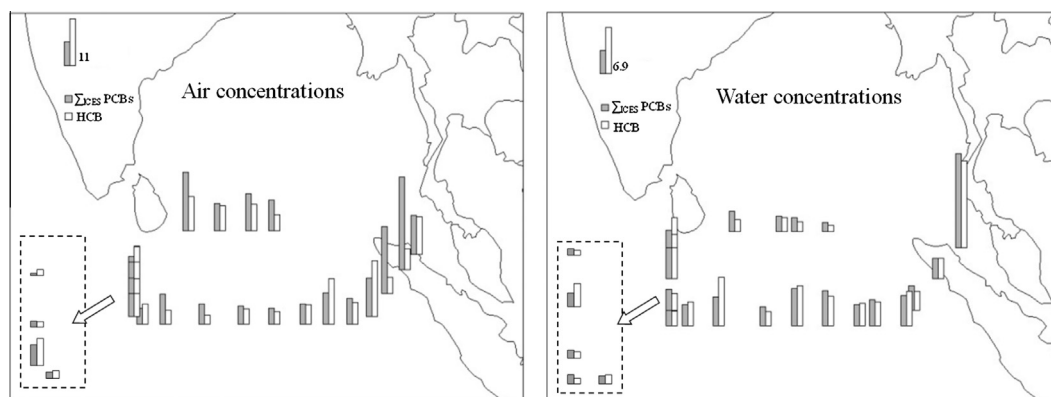

Fig. 2. Spatial distribution of $\sum_{ICES}$PCBs and HCB in the air (pg m^{-3}) and seawater (pg L^{-1}) of the equatorial Indian Ocean.
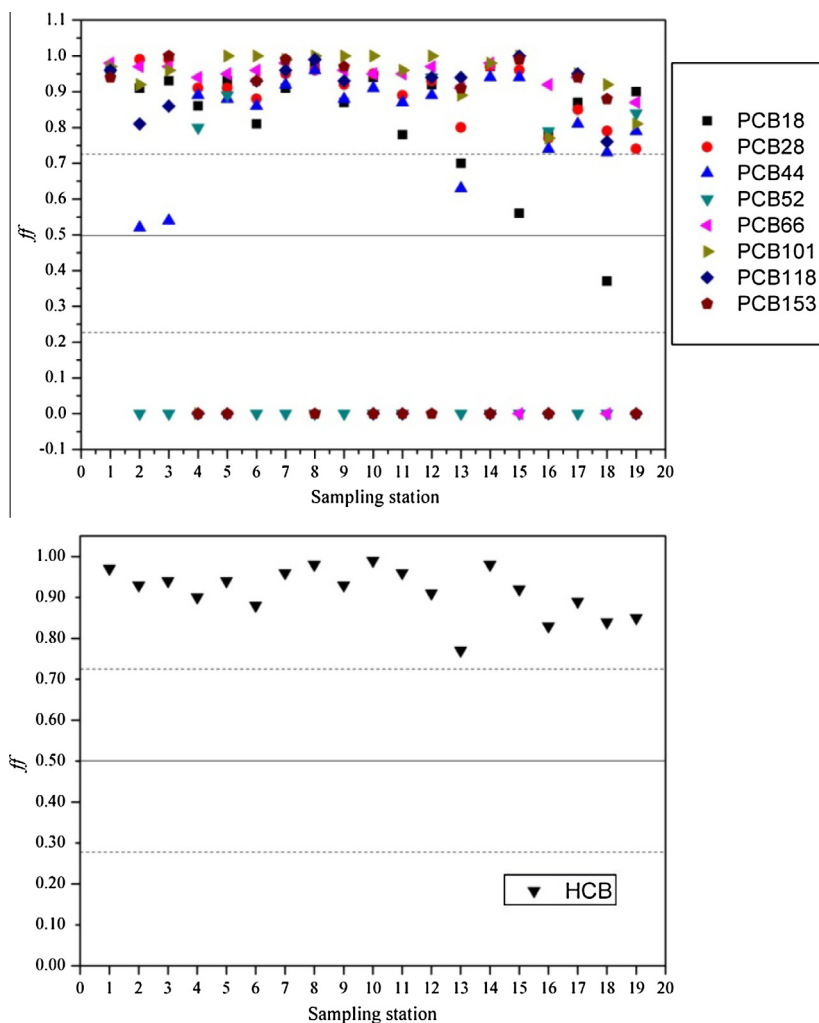
Fig. 3. Water–air fugacity fractions (*ff*) of selected PCBs and HCB along the sampling cruise. An uncertainty of 45% was used and marked as dashed lines in this figure.

Su et al., 2006; Wong et al., 2011). Equilibrium or near equilibrium of HCB was also indicated in the Pacific Ocean (Zhang and Lohmann, 2010).

3.4.2. Air–water exchange flux

Air–water gas exchange fluxes ($F_{a/w}$, ng m^{-2} d^{-1}) were estimated using a modified version of Deacon's boundary layer model (Schwarzenbach et al., 2003). Water is chosen as the reference phase, and a positive $F_{a/w}$ value indicates a net flux from the water to the atmosphere:

$$F_{a/w} = v_{ia/w} \times [C_w - C_a RT/H] \qquad (4)$$

where C_w, C_a, R, T, and H are the same as defined above, $v_{ia/w}$ (m d^{-1}) is the overall air–water gas exchange mass transfer coefficient (MTC) for chemical i, and was calculated according to Schwarzenbach et al. (2003).

In this study, the flux of $\sum_8$PCBs (PCB18, 28, 44, 52, 66, 101, 118 and 153) ranged from 0.45 to 34 ng m^{-2} d^{-1}, with a mean value of 3.9 ng m^{-2} d^{-1}. This range is similar to the values over the Pacific Ocean, where the flux of $\sum_{15}$PCBs was from −0.4 to 30 ng m^{-2} d^{-1} (Zhang and Lohmann, 2010). Much lower fluxes were observed in the Atlantic (−7 to 0.02 ng m^{-2} d^{-1} for $\sum_5$PCBs) (Gioia et al., 2008a) and the North Atlantic and Arctic Ocean (−4 to 0.08 ng m^{-2} d^{-1} for $\sum_4$PCBs) (Gioia et al., 2008b), where deposition dominated. Tri- (PCB18 and 28) and tetra (PCB 44, 52 and 66) dominated the $\sum_8$PCBs fluxes in this study, accounting for

92% of the average value. This finding is similar to the observation in the Atlantic and Arctic Ocean, where tri- and tetra-PCBs contributed more than 70% to the total PCB fluxes (Gioia et al., 2008a,b). However, Zhang and Lohmann (2010) found that di- and tri-PCBs dominated the fluxes of $\sum_{15}$PCBs in the open Pacific, with abundance more than 80% at most sites.

The flux of HCB in this study ranged from 0.36 to 18 ng m^{-2} d^{-1}, with an average of 2.2 ng m^{-2} d^{-1}. Higher fluxes from the air to water were observed in the Canadian Arctic (−6.1 to −15 ng m^{-2} d^{-1}, mean = −9.6 ± 2.7 ng m^{-2} d^{-1}) (Wong et al., 2011) and North Atlantic (−30.33 to −3.45 ng m^{-2} d^{-1}, mean = −14.8 ± 7.5 ng m^{-2} d^{-1}) (Zhang et al., 2012).

Pearson correlation analysis suggested that fluxes were mostly determined by the concentration gradient (C_w–$C_a RT/H$), with correlation coefficients of 0.849 ($p < 0.001$) and 0.775 ($p < 0.001$), respectively, for $\sum_8$PCBs and HCB. Wind speed was also correlated with the fluxes ($R = 0.541$ and $p = 0.017$ for $\sum_8$PCBs; $R = 0.514$ and $p = 0.024$ for HCB). This is consistent with the result in the Pacific Ocean, where PCB fluxes were mostly driven by the concentration gradient and influenced by wind speed as well (Zhang and Lohmann, 2010).

4. Conclusion

In this study, a significant decline was observed in the concentrations of PCBs in the air over the equatorial Indian Ocean since

1990s. Compared with the levels reported over other oceans, the gaseous concentrations of PCBs and HCB fell in the low end of global range, while the levels in the seawater were higher or similar to those in other oceans. The air samples collected near the continents displayed higher PCB levels than those from the open equatorial Indian Ocean. This may be related to land-based emissions, such as storage of obsolete PCB-containing equipments, landfill, incineration of e-waste, and ship-breaking activities. Fugacity fractions suggest that both PCBs and HCB were undergoing net volatilization from the seawater to air in the equatorial Indian Ocean in most cases. The flux values were mostly driven by the concentration gradient and wind speed as well.

Acknowledgments

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.marpolbul.2014.01.007.

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