



Soil concentrations, occurrence, sources and estimation of air–soil exchange of polychlorinated biphenyls in Indian cities



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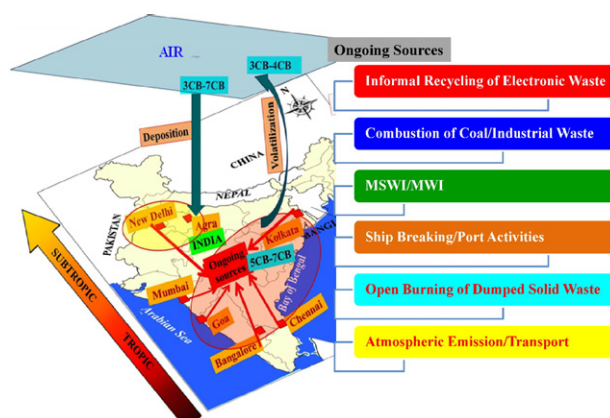
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HIGHLIGHTS

- Though the story of PCBs is fading in developed countries but high soil PCB concentrations have been observed in India.
- Except few hotspots, soil is acting as a sink for heavier congeners.
- Positive Matrix Factorization could identify six ongoing source types for PCBs in Indian cities.
- Occurrence of PCBs in sites away from the hotspots is likely due to atmospheric transport

GRAPHICAL ABSTRACT



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ABSTRACT

Past studies have shown potentially increasing levels of polychlorinated biphenyls (PCBs) in the Indian environment. This is the first attempt to investigate the occurrence of PCBs in surface soil and estimate diffusive air–soil exchange, both on a regional scale as well as at local level within the metropolitan environment of India. From the north, New Delhi and Agra, east, Kolkata, west, Mumbai and Goa and Chennai and Bangalore in the southern India were selected for this study. 33 PCB congeners were quantified in surface soil and possible sources were derived using positive matrix factorization-sub model. Net flux directions of PCBs were estimated in seven major metropolitan cities of India along urban-suburban-rural transects. Mean Σ_{33} PCBs concentration in soil (12 ng/g dry weight) was nearly twice the concentration found in global background soil, but in line with findings from Pakistan and urban sites of China. Higher abundance of the heavier congeners (6CB–8CB) was prevalent mostly in the urban centers. Cities like Chennai, Mumbai and Kolkata with evidence of ongoing PCB sources did not show significant correlation with soil organic carbon (SOC). This study provides evidence that soil is acting as sink for heavy weight PCB congeners and source for lighter congeners. Atmospheric transport is presumably a controlling factor for occurrence of PCBs in less polluted sites of India.

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1. Introduction

High levels of polychlorinated biphenyls (PCBs) have been occasionally reported in developing regions at lower latitudes, despite a relatively limited historical usage of these chemicals (Breivik et al., 2011). Urban centers are the primary source regions for PCBs (Harner et al., 2004; Iwata et al., 1993; Jaward et al., 2004). PCBs were never manufactured in India but recently high atmospheric PCBs have been observed in the urban environment along the coastal length of India (Zhang et al., 2008) particularly in the metropolitan cities (Chakraborty et al., 2013). Chakraborty et al. (2013) reported profound influence of primary sources associated with cumulative growth of informal electronic waste recycling among the informal scrap dealers, ship breaking activities and secondary sources like open burning of municipal solid waste or biomass burning on the atmospheric PCBs in Indian cities. Soil represents an interesting archive of PCBs as these are influenced by past atmospheric deposition. PCBs have a strong affinity for soil organic matter (SOM) (Mackay, 2001) and their global distribution is retarded by soil (Ockenden et al., 2003). Studies conducted in developed nations e.g., in the urban residential soil of Iowa USA (Martinez et al., 2012) and urban areas of European cities (Cachada et al., 2009) reported heterogeneity in soil PCB distribution. Reported soil PCB concentrations in the developing countries of Southeast Asia are also highly variable. Average PCB concentration in Chinese surface soil (Ren et al., 2007) is one tenth of the global background soil concentration (Meijer et al., 2003). However, urban locations and electronic waste (e-waste) recycling sites in China reported much higher PCB concentrations in soil (Jiang et al., 2011; Tang et al., 2010; Wang et al., 2011). Recent studies reported elevated levels of PCBs in soil from Pakistan (Syed et al., 2013). PCB burden in soil has been reported from the north eastern states of India (Devi et al., 2014) and agricultural sites of New Delhi (Kumar et al., 2011). Following ratification of the Stockholm Convention, Ren et al. (2007) encouraged the need for more national level studies to assess contemporary PCB burden and emission sources. Despite clear evidence of increasing atmospheric PCBs in India from early 1990s (Iwata et al., 1994) very limited information is available on the soil PCB concentration from the highly populated urban environment of India and on the possible role of soil as a secondary source to the atmosphere. The main objectives of this study were to: (i) investigate the surface soil PCB concentration along urban-suburban-rural transect from seven major metropolitan cities of India viz., New Delhi and Agra from North, Kolkata from East, Mumbai and Goa from West and Chennai and Bangalore from Southern part of India (ii) estimate the net air-soil exchange of PCBs using previously published atmospheric data from all these cities (Chakraborty et al., 2013) (iii) identify potential sources using multivariate pattern analysis technique and receptor model and (iv) assess the regional variability and identify potential hotspots at local level within each metropolitan city.

2. Materials and methods

2.1. Sampling

84 surface soil samples (0–20 cm) were collected from seven major cities of India along urban, suburban and rural transects (Fig. 1). Surface soil sampling was conducted during the same time frame of active air sampling in these cities (Chakraborty et al., 2013). Details of each site can be obtained from Supplementary Material. Five sub samples were collected from an approximated 100 square meter grid in zig-zag manner and mixed well to make one representative composite sample from each site. During the sample collection, gloves were worn, and surface soil samples collected were handled using acetone-rinsed stainless steel spatula. At the end of the collection, the soil samples were sealed in ziploc bags, and returned to the laboratory where they were stored in sealed, solvent-cleaned brown glass jars at -20°C until extraction.

2.2. Extraction and analysis

Soil samples were air dried, mixed and sieved through a 2 mm sieve. Prior to extraction a mixture of surrogate standards (2,4,5,6-tetrachloro-*m*-xylene (TCmX), decachlorobiphenyl (PCB209), $^{13}\text{C}_{12}$ -PCB138 and $^{13}\text{C}_{12}$ -PCB180) were added to each of the samples. Soil samples (20 g each) were Soxhlet extracted for 18 h with dichloromethane (DCM). Activated copper granules were added to the collection flask to remove potential elemental sulfur. The extract was rotary evaporated and transferred with hexane to a 15 ml amber vial. This was blown down under a gentle stream of nitrogen to about 0.5 ml and cleaned on a 8 mm i.d. chromatography column with 6 cm alumina (BDH neutral Alumina 3% deactivated), 10 cm of silica gel (Merck Silica 60 3% deactivated), 10 cm of 50% sulfuric acid silica and 1 cm of baked sodium sulfate (all baked at 450°C overnight) and the column was eluted with 10 ml mixture of 50:50 hexane: DCM (v/v). The samples were reduced to a final volume of 25 μl after adding 25 μl of dodecane as solvent keeper and a known quantity of PCB-54 was added as an internal standard prior to GC-MSD analysis. PCBs analysis was carried out on a Finigan-TRACE GC-MS system with a CP-Sil 8 CB capillary column (50 m, 0.25 mm, 0.25 μm), operating under single-ion monitoring (SIM) mode. Helium was used as the carrier gas at 1.2 ml min^{-1} under constant-flow mode. The oven temperature began at 60°C for 1 min and increased to 290°C (10 min hold time) at a rate of $4^{\circ}\text{C min}^{-1}$. Splitless injection of a 1 μl sample was performed with a 5 min solvent delay time. Injector temperature was at 250°C . A total of 33 PCB congeners were detected and quantified. Congener details have been given in Supplementary Material.

2.3. QA/QC

Chemical standards were purchased from Accustandard Co. US. Method blanks were analyzed for every batch of 9 samples to provide an indication of the overall precision of the laboratory method. Limit of Detection (LOD) were derived from the blanks and quantified as the mean plus three times the standard deviation of the concentration in the blanks, based on surface soil sampling. LOD values varied between 0.0019 and 0.0023 ng/g dry weight (dw). Procedural blanks were below the instrument quantification limit. Surrogate recoveries were 91–135% (mean 97%) for PCB209, 72–81% (mean 77%) for TCmX, 87–91% (mean 89%) for $^{13}\text{C}_{12}$ -PCB138 and 82–109% (mean 90%) for $^{13}\text{C}_{12}$ -PCB180. Concentrations of the samples were corrected based on the recovery ratios and blank values.

2.4. Soil organic carbon

1 g of soil sample was mixed with 3 ml of 10% HCl acid and kept for 8 h to remove inorganic carbon. Soil samples were then rinsed with doubled distilled water (three times) dried in oven at 45°C . Weight of the soil samples were recorded before analysis by Elemental Carbon-Hydrogen-Nitrogen Analyzer (Elementar VARIO EL III).

2.5. Fugacity fractions

Fugacity fractions (*ff*) were calculated for eight PCB congeners viz., PCB-28, 52, 101, 105, 118, 138, 153 and 180. The average concentration of atmospheric PCBs from each of the Indian cities measured by active air sampling during similar time frame (Chakraborty et al., 2013) were used to calculate the fugacity in the air (f_{AAS}). Measured soil concentrations from each site were used to calculate the fugacity in soil (f_s) for each site. The following equations were used:

$$ff = f_s / (f_s + f_{\text{AAS}}) \quad (1)$$

$$f_s = C_s RT / 0.411 \Phi_{\text{OM}} K_{\text{OA}} \quad (2)$$

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