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Occurrence and sources of selected organochlorine pesticides in the soil of seven major Indian cities: Assessment of air–soil exchange

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ABSTRACT

India is an agricultural country and organochlorine pesticides (OCPs) accounts for nearly three fourth of the annual pesticide consumption. Selected OCPs were therefore quantified in 81 soil samples along urban–suburban–rural transect from New Delhi and Agra in the north, Kolkata in the east, Mumbai and Goa in the west and Chennai and Bangalore in the southern part of India. ΣOCPs ranges from 2 to 410 ng/g dry weight (Mean, 35) with dominance of endosulfan sulfate in the rural sites. Urban centers and suburbs reflects OCP usage for vector control. Lower winter temperature in New Delhi favored site-specific deposition of most OCPs in soil. Volatilization of OCPs from soil occurred in the Indian cities having higher ambient temperature. Due to the compounded impact of past and ongoing usage of selected OCPs like DDT, a sporadic cycle of emission and re-emission from Indian soil is expected to continue for many more years to come.

1. Introduction

Nearly 70% of the total Indian workforce depends on agriculture for survival so application of pesticides to protect commercial crops is a basic practice. India is the fourth largest pesticide producer in the world after USA, Japan and China. But pesticide consumption in India (0.5 kg/ha) is nearly 6–10 times lower than developed nations in Europe or in USA (3 kg/ha) and considerably lower than Asian countries like Japan (12 kg/ha), Taiwan (17 kg/ha) and Korea (6.6 kg/ha) (Abhilash and Singh, 2009). Though pesticides have enormous beneficial effects in India for growth in agriculture however the unregulated and random application of pesticides has raised serious concerns about the ambient environment. Moreover the pesticide usage pattern in India is also different from that of the rest of the world. Almost 80% of pesticides used in India are insecticides whereas across the globe herbicides are the leading category followed by insecticides and fungicides. Organochlorine pesticides (OCPs) such as *p,p′*-dichlorodiphenyltrichloroethane (DDT), γ-hexachlorocyclohexane (HCH) and malathion accounts over 70% of the annual pesticide consumption (85,000 tonne per annum) in India (Gupta, 2004). Hence India is facing the adverse residual effects of extensively used organochlorine pesticides (OCPs) like HCHs, DDTs, endosulfan, etc. (Rekha et al., 2006; Agoramoorthy, 2008; Government, 2008) which are restricted or banned globally under the Stockholm Convention (Loganathan et al., 1994; Wang et al., 2009). Exposure to pesticides can lead to acute or chronic health problems. Nearly 10% of the total number of cancer patients are found to be exposed to pesticide poisoning (Sufen, 2005).

Recently elevated level of atmospheric OCPs has been reported from seven major Indian cities (Chakraborty et al., 2010). OCPs have the tendency to accumulate in the naturally occurring organic matter present in soil ultimately leading to bioaccumulation. Soil therefore plays an important role in the global distribution and fate of POPs because it does not act only as sink but also re-emit these pollutants into air thereby acting as a secondary source (Wild and Jones, 1995; Harner et al., 2001). To improve the accuracy in estimating the rate of emissions not only the amount of pesticide used at the time of application but also the residues in the soil has to be considered. There is paucity of data on residual concentrations of OCP from Indian cities. Hence the present study was undertaken to investigate the soil burden of selected OCPs from seven major Indian cities with the following objectives: i. their occurrence along urban–suburban and rural transect; ii. Source apportionment and iii. assessment of their air–soil exchange process by evaluating the

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fugacity fractions using our previously published air data by active and passive air sampling during the same time frame (Chakraborty et al., 2010).

2. Methods

2.1. Sampling sites

Eighty one soil samples has been collected from seven major Indian cities (Fig. 1) based on urban–suburban and rural transect. SI-X Table 1 in the Supporting Information has the details of the sampling sites. Most of the sampling sites are same where the passive samplers were deployed for the four major metropolitan cities viz., New Delhi, Kolkata, Mumbai and Chennai (Chakraborty et al., 2010). Each of the surface soil samples (0–20 cm depth) were comprised of 5 subsamples collected from along a 100 square meter grid, which is representative of each site. All soil samples were collected using a hand auger and then stored in polyethylene bags. End of each sample collection the auger was cleaned with distilled water followed by three times rinsing with acetone to avoid contamination. Soil samples were kept at −18 °C until analysis.

2.2. Extraction and analysis

Soil samples were thawed and freeze-dried for 24 h, pulverized and sieved through 1 mm stainless steel mesh. About 20 g of the subsamples were Soxhlet-extracted for 18 h with dichloromethane (DCM) (both DCM and hexane obtained from Merck and Co., Inc.). A mixture of surrogate standards of 2,4,5,6-tetrachloro-m-xylene (TCmX) and decachlorobiphenyl (PCB209) was added to each of the samples prior to extraction. Activated copper granules were added to the collection flask to remove elemental sulfur. The extract was concentrated and solvent-exchanged to hexane and purified on an 8 mm i.d. aluminum/silica column packed, from the bottom to top, with neutral aluminum (6 cm, 3% deactivated), neutral silica gel (10 cm, 3% deactivated), 50% (on a weight basis) sulfuric acid silica (10 cm), and anhydrous sodium sulfate. Before used, neutral aluminum, neutral silica gel, and anhydrous sodium sulfate were Soxhlet-extracted for 48 h with DCM, and then baked for 12 h at 250 °C, 180 °C, and 450 °C, respectively. The column was eluted with 50 mL of

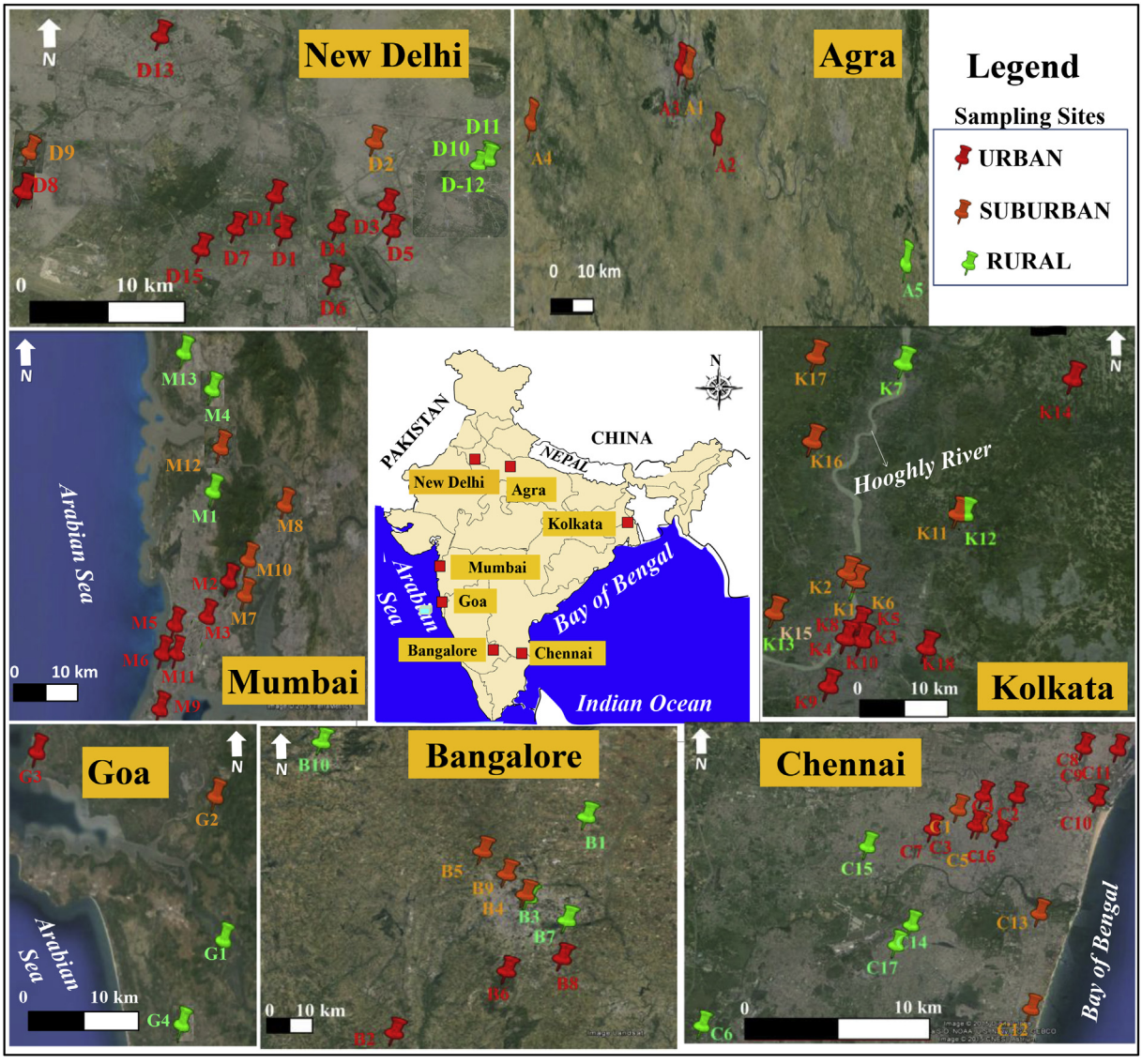

Fig. 1. Soil sampling sites along urban, suburban and rural transect for seven major Indian cities.