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Passive Air Sampling of PCDD/Fs, PCBs, PAEs, DEHA, and PAHs from Informal Electronic Waste Recycling and Allied Sectors in Indian Megacities

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Abstract

Xenobiotic chemical emissions from the informal electronic waste recycling (EW) sector are emerging problem for developing countries, with scale and impacts that are yet to be evaluated.

We report an intensive polyurethane foam disk passive air sampling study in four megacities in India to investigate atmospheric organic pollutants along five transects viz., EW, information

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Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.est.1c01460>.

Sampling locations; Meteorological conditions during sampling; PMF model settings; descriptive statistics (PDF)

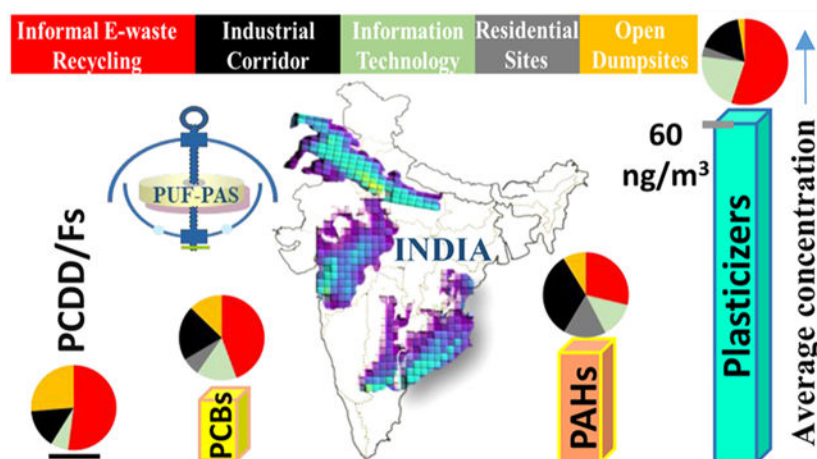
Table S1-1X (XLSX)

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technology (IT), industrial, residential, and dumpsites. Intraurban emission sources were estimated and attributed by trajectory modeling and positive matrix factorization (PMF). $_{17}\text{PCDD/Fs}$, $_{25}\text{PCBs}$, $_{7}\text{plasticizers}$, and $_{15}\text{PAHs}$ concentrations ranged from 3.1 to 26 pg/m^3 (14 ± 7 ; Avg \pm SD), 0.5–52 ng/m^3 (9 ± 12); 7.5–520 ng/m^3 , (63 ± 107) and 6–33 ng/m^3 (17 ± 6), respectively. EW contributed 45% of total PCB concentrations in this study and was evidenced as a major factor by PMF. The dominance of dioxin-like PCBs (dl-PCBs), particularly PCB-126, reflects combustion as the possible primary emission source. PCDD/Fs, PCBs and plasticizers were consistently highest at EW transect, while PAHs were maximum in industrial transect followed by EW. Concentrations of marker plasticizers (DnBP and DEHP) released during EW activities were significantly higher ($p < 0.05$) in Bangalore than in other cities. Toxic equivalents (TEQs) due to dl-PCBs was maximum in the EW transect and PCB-126 was the major contributor. For both youth and adult, the highest estimated inhalation risks for dl-PCBs and plasticizers were seen at the EW transect in Bangalore, followed by Chennai and New Delhi.

Graphical Abstract



Keywords

POPs; plasticizers; PAHs; PMF; FLEXPART

1. INTRODUCTION

Electronic waste (e-waste) recycling emerged as an impending problem due to vague accounts of emissions from misreported waste inflows.¹ Informal operations and disposal in developing nations² involves recovery of lethal yet expensive metals and other useful components.³ It is projected that by 2030, developing nations will exceed developed nations in obsolete personal computer waste generation by 100 million tonnes.⁴ In 2012, the transboundary movement of e-waste from rich nations was less than 1%, whereas close to 25% of total e-waste moved between the World Trade Organisation non-Annex VII countries.⁵ China doubled its e-waste generation between 2010 and 2015, producing 6 million tonnes, the highest in Asia.⁶ By 2014, India produced 2.7 million tonnes of e-waste and the compound annual growth rate of e-waste exceeded China for the years 2015–

2019.⁷ Hazardous recycling methods involved in processing of e-waste in the backyards of developing countries, present serious environmental and human health concerns. Due to informal e-waste recycling (EW), toxic emissions have been reported from China,⁸ Pakistan,⁹ Bangladesh,¹⁰ and Vietnam.¹¹ Emissions of persistent organic pollutants (POPs), plasticizers and polycyclic aromatic hydrocarbons (PAHs) due to crude dismantling, open burning of electronic coatings, acid stripping and other chemical processes involved in EW sector are well documented from India.^{12,13}

Polyurethane foam (PUF) disk passive air samplers (PAS) are one of the most improved techniques in the recent years for monitoring of POPs and widely recognized in studies across the globe.¹⁴ The efficacy of using PUF-PAS has been demonstrated among diverse sites along the urban, rural and latitudinal transects.¹⁵ The feasibility of coordinated sampling at the global/continental scale¹⁶ has been also demonstrated by studies in Europe,¹⁷ Asia¹⁸ and Latin America,¹⁹ where data from PAS surveys have confirmed their efficacy in defining known source/background areas of POPs²⁰ and PAHs²¹ contamination. At the national/regional scale in India, POPs,^{22,23} PAHs²⁴ and other particle bound POPs²⁵ have been studied using PAS resolving spatial and seasonal trends.²⁶ Sorbent impregnated passive samplers have been used for atmospheric monitoring of plasticizers;²⁷ however, PUF-PAS has been effectively used in India²⁸ and waste processing sites of Vietnam.²⁹

It has been evident for the last two decades that atmospheric polychlorinated biphenyls (PCBs) concentrations in Indian cities are consistent with and in some cases higher than other global megacities.^{22,26} Studies point to urban EW and open burning³⁰ rather than the in-use and stored closed sources accounted for in the official national PCB inventory.³¹ India's contribution of nearly one-fourth of the global PAH emission budget³² can be attributed to both urban (industrial and transport) and rural (biomass burning) sources, and recent studies observed no difference in the atmospheric PAHs levels along urban and rural sites.³³ Waste plastic burning such as wires/cables and plastic blocks were identified as primary sources for atmospheric PAHs and plasticizers.³⁴ About 64% of the total e-waste in India is generated by Mumbai (24%), New Delhi (21.2%), Bangalore (10.1), and Chennai (9%).³⁵ Since EW workshops are relatively new and growing sources for hazardous substances in the megacities of India,³⁶ we report ambient atmospheric concentrations of POPs, plasticizers and PAHs in the EW sector and other allied sectors using PUF-PAS. This study is aimed at realizing the atmospheric concentrations, sources, and risk associated with atmospheric polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzo-*p*-furans (PCDD/Fs), PCBs including dioxin like PCBs (dl-PCBs), plasticizers, and PAHs in four Indian megacities viz., Chennai, New Delhi, Bangalore, and Mumbai along EW sector, information technology corridor, open municipal dumpsites, industrial area, and residential sites.

2. MATERIALS AND METHODS

2.1. Sampling.

A total of 25 PUF-PAS were deployed in four major metropolitan cities of India: Chennai ($n = 10$), New Delhi ($n = 8$), Mumbai ($n = 2$), and Bangalore ($n = 5$). Sampling sites were established along five transects viz., EW sector, information technology corridor

(IT), industrial area (IN), open municipal dumpsites (DS), and residential sites (RS). Sampling details are given in Supporting Information (SI) Table S1. Each PUF-PAS was precleaned and deployment procedure can be obtained elsewhere²³ wherein authors calibrated PUF-PAS for organochlorine compounds against a high volume sampler in tropical and subtropical climate of Indian cities. Samplers were deployed for 28 days in January and February 2015. For each city one field blank sample was brought back to lab for further analysis. Details on wind speed, temperature and precipitation have been given in SI Table S2. For consistency sampling rate of 3.5m³/day used in other studies for POPs,^{19–22,26} PAHs²⁴ and plasticizers^{30,29} was used to obtain time integrated concentration of each analyte. Deployment-specific sampling rates were also estimated for each PUF-PAS deployment and for each study compound and PCB congeners using the PUF-PAS sampling rate model, simulated from 6-hourly meteorological reanalysis and the molecular weight, octanol-air partition coefficient, and internal energy of transfer for each compound.^{37,38} Details of the deployment-specific estimated sampling rates for each city have been given in the SI Table SI-1X.

2.2. Extraction and Analysis.

The PUF disks were extracted with 150 mL of toluene in the Soxhlet apparatus for 24 h maintaining an average six cycles per hour. The extract was concentrated in a Buchi (Switzerland) rotary evaporator and reduced to 2 mL. Each sample extract was divided into four parts and each quarter was subjected to compound specific cleanup. Prior to extraction, PCB-209, isotope labeled 12 coplanar PCBs, deuterated DnBP, DEHP and phenanthrene were added as surrogate recovery standards.

2.3. Column Clean Up and Instrumental Analysis.

2.3.1. PCDD/Fs.—The column cleanup for PCDD/Fs analysis was done as per United Stated Environmental Protection Agency (USEPA) method 1613 mentioned elsewhere.³⁹ Sample extracts were eluted in the extended multilayer column. The column was loaded from bottom with 0.9 g of anhydrous sodium sulfate, 3 g of silica gel impregnated by potassium hydroxide, 0.9 g of silica, 4.5 g of silica gel impregnated by 44% sulfuric acid, 6 g of silica gel impregnated by 22% sulfuric acid, 0.9 g of silica, 3 g of silica gel impregnated by 10% silver nitrate, 0.9 g of silica and finally added 6 g of sodium sulfate on the top. The sample extracts were loaded in the above-mentioned multilayer column and subsequently eluted with 150 mL of hexane followed by an active carbon column to separate PCB fraction using 40 mL of DCM/hexane (3:1) mixture. The dioxin fraction was eluted afterward with 60 mL of toluene by the reverse carbon column for complete separation of PCDD/Fs absorbed in the top layer. PCDD/Fs were analyzed in a high resolution gas chromatograph coupled with high resolution mass spectrometer, HRGC/HRMS (JEOL JMS-800D) with a DB-5MS fused silica capillary column (60m × 0.25 mm, film thickness = 0.25 μm) by splitless injection. Analysis parameters are given in Supporting Information and list of congeners analyzed are given in Table S3.

2.3.2. PCBs.—Multilayered silica column (150 mm × 15 mm) was packed from bottom to top with 3 cm alumina (3% deactivated), 3 cm silica gel (3% deactivated), 2 cm acidic silica (50% by sulfuric acid), and 1 cm anhydrous sodium sulfate (Merck, India, precleaned

and stored in desiccator). The column was prerinsed with 10 mL *n*-hexane (HPLC grade, Rankem, India) before sample was loaded. The elution of PCBs was subsequently carried out using 20 mL of hexane: dichloromethane (DCM) (1:1 v/v). The eluent was collected and reduced by a gentle stream of nitrogen gas. PCBs were analyzed in gas chromatograph coupled with mass spectrometer (Agilent 7890B Gas Chromatography and 5977A MS). Analysis parameters are given in Supporting Information and list of congeners analyzed are given in Table S3.

2.3.3. Plasticizers (PAEs and DEHA).—USEPA-listed six priority phthalic acid esters (PAEs) and bis-2-ethylhexyl adipate (DEHA) were cleaned and concentrated according to the method given elsewhere.¹² Briefly, the column was packed with neutral silica gel (6 cm), neutral alumina (3 cm), and anhydrous sodium sulfate (0.5 cm). Hexane was used for conditioning and 20 mL mixed solvent of acetone/*n*-hexane (2:8, v/v) was used for elution. PAEs and DEHA were analyzed in the aforementioned gas chromatograph coupled with mass spectrometer. Analysis parameters are given in Supporting Information and list of compounds analyzed are given in Table S3.

2.3.4. PAHs.—The column was packed from bottom to top with 3 cm silica gel and 1 cm anhydrous sodium sulfate. The column was conditioned with 10 mL of hexane. The elution was done with 20 mL of hexane: DCM (1:1 v/v). PAHs were analyzed in the aforementioned gas chromatograph coupled with mass spectrometer. Analysis parameters are given in Supporting Information and USEPA-listed 15 priority PAHs analyzed are given in Table S3.

2.4. Quality Assurance and Quality Control (QA/QC).

Spiked recovery of isotope labeled coplanar PCBs, PCDD/Fs and PCB-209 varied between 88% and 103% including samples and blanks, whereas seven plasticizers varied between 105% and 112%. Only DnBP and DEHP were observed in the procedural blanks with concentration above limit of quantification (LOQ) and were subtracted in samples from respective batches. Compounds were identified based on their retention time and quantified using internal standard procedure. Limit of detection (LOD) and LOQ for each congener was calculated from field and procedural blanks. LOD is average plus three times the standard deviation of all the blanks. LOD for congeners undetected in the blanks were calculated using the instrumental detection limit (signal-to-noise, 3:1). LOD for individual congeners are given in SI Table S4. Further details regarding field and procedural blanks are given in the Supporting Information.

2.5. Back Trajectory Analysis.

Flexible Particle Trajectory (FLEXPART) is a Lagrangian Dispersion Model that calculates atmospheric particle trajectories while accounting for subgrid scale processes such as convection, diffusion, turbulence, dry deposition, and wet scavenging.^{40–42} The backward mode output of the model provides back-trajectories and calculator source-receptor relationships on a three-dimensional grid.⁴³ We used National Centers for Environmental Prediction Final Analysis (NCEP FNL) Operational Model Global Tropospheric Analysis data (henceforth referred as FNL) to drive the model and calculated source-receptor

relationships for the four cities in a simulation of the Indian subcontinent. The model along with FNL data resolves transport pathways over south India.^{44,45} SI Table S5 summarizes FLEXPART input files and settings.

2.6. Positive Matrix Factorization.

The Positive Matrix Factorization (PMF) model developed by the USEPA was used for source apportionment of PCBs. PMF model V5.0⁴⁶ was used to identify the source types of PCBs by solving the negative factor loadings/scores by integrating non-negativity constrained factor analysis, making factor loadings and scores more interpretable to samples based on the fingerprints of the source types. Several studies have used PMF to identify sources for atmospheric pollution from PCBs, PAHs, and other study compounds.^{47,48} The factors were aligned through linear correlation such that each set of PCB congeners always referred to the same factor. The PMF configuration is reported in SI Table S6.

2.7. Risk Assessment.

The Toxic Equivalents (TEQs) for PCDD/Fs, dl-PCBs and carcinogenic PAHs were calculated by multiplying the detected concentration with the corresponding Toxic Equivalency Factors (TEFs) provided by the World Health Organization (WHO).⁴⁹ The calculations for TEQs and daily inhalation intake rate of PCDD/Fs, PCBs, plasticizers and PAHs are described in the SI.

2.8. Statistical Analysis.

SPSS version 22 (IBM SPSS Statistics for Windows, Version 22.0. Armonk, NY: IBM Corp) was used for statistical computations. Test of significance was performed using one-way ANOVA and/or independent *t* test. Statistical significance was defined as $p < 0.05$ and more confident observations were mentioned where relevant. Spearman's rank correlation was used for correlation analysis.

3. RESULTS AND DISCUSSION

3.1. Levels and Homologue Profiles.

Deployment-specific average PCB sampling rates varied from 3.39 m³/day in Mumbai to 4.30 m³/day in Bangalore (SI Table SI-1X), with the average congener-specific estimate across all samples not varying more than 5% from the previously reported calibrated standard value of 3.5 m³/day.^{2,24,19–24} Derived atmospheric concentrations of PCBs, PAEs, and PAHs from most widely used sampling rate of 3.5m³/day^{6,29,30} was correlated with modeled average sampling rate and compound specific sampling rates for each city. Strong statistically significant correlations ($p < 0.01$) were observed between the sum of concentrations of targeted group and subgroups of analytes obtained from both the previously reported and model estimated sampling rates (SI Figure S1a,b). Due to lack of significant differences between concentrations obtained from different sampling rates, we have used the sampling rate of 3.5 m³/day to maintain a consistency in the entire data set. Range of each target analyte has been given in Figure 1 and descriptive statistics in SI Table S7a–d.

3.1.1. PCDD/Fs.—PCDDs in major Indian cities ranged between 1.2 and 21.3 pg/m³, whereas PCDFs ranged between 1–5.8 pg/m³. The most abundant PCDD/Fs homologue was TeCDD (67%), followed by TeCDF (39%), OCDD (23%), and HxCDF (17%). Such dominance of TeCDF has been previously observed in Tianhe industrial district in China.⁵⁰ In Indian cities decrease in PCDFs concentrations with the increase in chlorination is in line with other studies from Latin America, Europe, U.S, Japan, and Australia.^{51,52} The metal recovery sites in New Delhi, Seelampur (EW-03), and Mustafabad (EW-04) were found with the highest PCDDs (21.3 pg/m³) and PCDFs (5.8 pg/m³) concentration in this study, respectively. These sites belong to primary EW area in India where one-fourth of e-waste generated in the country is recycled.⁵³ It is noteworthy that unlike other cities, PCDFs are associated with the extensive evidence of concurrent combustion and acid leaching processes during metal recovery in the EW sector of New Delhi. Hence, we suspect re-emission of PCDD/Fs from such contaminated soil in the EW recycling workshops of New Delhi.¹³ SI Table S7a shows the descriptive statistics for PCDD and PCDF homologue groups.

3.1.2. PCBs.—₂₅PCB concentrations varied between 0.5 and 52 ng/m³ (avg ± stdev: 9.4 ± 11.9). More than 45% of the measured ₂₅PCB were from EW sites (Figure 2). Average sum of indicator PCBs (I-PCBs) (5.2 ng/m³) from EW sites was 2-fold higher than other sites in the study. 4-Cl was the predominant homologue in the study at all transect types except RS. This observation is consistent with the metropolitan environment of Chennai in southern India⁴⁷ and non-metropolitan environment of Agra in northern India.⁵⁴ Between non-ortho and mono-ortho PCBs we found a strong correlation in both EW ($R^2 = 0.96$, $p < 0.01$) and IT sites ($R^2 = 0.99$, $p < 0.01$). It is noteworthy that unlike other cities, a strong correlation ($R^2 > 0.7$, $p < 0.05$) was found for ₂₅PCB concentrations among all sites in New Delhi, suggesting shared or similar sources across the city. SI Table S7b shows the descriptive statistics for individual PCB congeners. Nearly 40% of overall dl-PCBs in this study was found in the EW sites. The highest dl-PCB level (15 ng/m³) was observed at a dumpsite in Chennai (DS-01, Kodangaiyur) which is suspected to contain waste sludge discarded from a nearby EW site.¹³

We observed an increase in dl-PCB concentrations by a factor of 3–7 in all major cities since the last reported observations.^{22,30} Current dl-PCBs at the EW transect (Avg: 4 ± 3.5 ng/m³) were lower than e-waste recycling sites of Guiyu⁵⁵ and Taizhou⁵⁶ in China prior to the regulations in 2010, and comparable to those reported after the e-waste import ban and establishment of formal recycling zones in Zhejiang province.⁵⁷ E-waste related dl-PCBs are higher than e-waste levels reported from the Ivory Coast and Ghana.⁵⁸ Industrial emissions (8.1 ± 12 ng/m³) are comparable to metallurgical industrial belts of Shanghai (China)⁵⁹ and higher than Barcelona (Spain)⁶⁰ and Izmir (Turkey).⁶¹ Dumpsites in India are characterized by open burning of unsegregated municipal waste, another major contributor to dl-PCBs in India.^{13,47}

3.1.3. Plasticizers (PAEs and DEHA).—Total concentrations of six PAEs and DEHA (7 plasticizers) ranged between 7.5 and 520 ng/m³ (63 ± 107). The highest concentration was observed in a metal recovery site in Bangalore (EW-06, Goripalaya 520 ng/m³). Nearly 50% of 7 plasticizer concentrations were contributed by DEHP, followed by DEP (23%)

and DnBP (17%). These results are consistent with a past study from south India²⁸ and lower than waste processing sites of Vietnam.²⁹ Extensive use of DEHP representing 50% of total plasticizer production, with 95% being used in PVC products, can be attributed to the dominance of atmospheric DEHP in Indian cities. Higher DEP concentrations despite higher production and usage of DnBP is likely due to DEP's atmospheric half-life (1.8–18 days) being higher than DnBP (0.6–6 days).⁶² Average concentration of γ plasticizers in EW transect was approximately 5-fold higher than average concentrations at other transects. Similar dominance of PAEs in PM_{2.5} was observed at urban e-waste sites of China in summer and winter.³⁴ SI Table S7c shows the descriptive statistics for individual plasticizers.

3.1.4. PAHs.—Atmospheric sum concentrations of 15 priority PAHs enlisted in USEPA (15PAHs) varied between 6.2 and 33 ng/m³ (17 ± 6). By transect type, PAHs varied in the following order: IN (33%) > EW (29%) > RS (16%) > IT (14%) > DS (9%). Bangalore recorded the highest average PAHs (21 ng/m³) while Chennai recorded the highest average carcinogenic PAHs (7 ng/m³). This range for PAHs is lower than prior studies in Canada (3.5–61.4 ng/m³),²¹ Chile (30–230 ng/m³),⁶³ Europe (0.5–61.2 ng/m³),⁶⁴ Mexico (32–92 ng/m³), and Sweden (7.6–68 ng/m³); higher than England (6.5 ng/m³)⁶⁵ and Italy (0.9–4.7 ng/m³);⁶⁶ and comparable with Alaska.⁶⁷ Prior PAS campaigns reported slightly higher 15PAH concentrations (6.4–54.8 ng/m³) for Chennai, Mumbai, and Kolkata.²⁴ SI Table S7d shows the descriptive statistics for individual PAH compounds.

3.2. Source Receptor Modeling and Intercity Variation.

Atmospheric outflow of organic pollutants can be impacted by several mixed sources in Indian cities. Based on prime functional activities prevalent in different transects or site types, we found dominance of distinct compounds at different transects. For example, among all the transects, we found the maximum percentage of PCBs (44%) and plasticizers (55%) at EW transect, whereas PAHs were slightly higher in the IN (33%) over EW transect (29%) (Figure 2). Although India has never manufactured PCBs, recent studies evidenced maximum dl-PCBs in the e-waste workshops of the informal sector.^{13,68} In this study, we found 40% dl-PCBs and nearly 60% I-PCBs in the EW sites (Figure 2). Under tropical climate, emission and re-emission of PCBs are potential sources of atmospheric emission of PCBs in Indian cities.⁶⁹ Hence the PCB data set was further employed for source-receptor modeling by PMF. Each PCB congener could be affected by varied activities prevalent at different site types, hence source profile fingerprints were derived from observed concentrations at each site. Source profile fingerprints obtained from the PMF exercises are discussed along with the distribution patterns of other study compounds sampled (Figure 3). It is noteworthy that PMF profile did not vary significantly when we used PCB concentrations obtained from average estimated sampling rate for each city (SI Table: S1X; SI Figure S1). Surface-level source-receptor (SR) relationships derived from five-day FLEXPART back trajectories were used to assess sensitivity of concentrations at each site to the emissions from each grid cell.⁴³ Local air mass influence observed concentrations, with higher residence time for air parcels over inland cities such as New Delhi and Bangalore. This condition can lead to higher contributions from local sources in

inland cities over coastal cities (Chennai and Mumbai), with a mean factor of 2–3 order in the importance of local and intraurban emission sources (SI Figure S2).

Factor 1: Informal E-Waste Recycling.—This factor was comprised of mostly penta-hepta PCB congeners, with PCB-77, -99, -105, -114, -118, -126, -138, -162, -167, -180 contributing 50–90% of the total factor profiles. SI Figure S3 compares the homologue patterns in this study with the major Aroclor formulations. Higher percentage of penta congeners, especially PCB-126 which is present in trace levels in all the Aroclor formulations (SI Figure S4), suggest the importance of combustion as the source and is consistent with soil profiles observed earlier.¹³ EW transect was found with 77% of PCB-118 and 55% of PCB-126 concentrations. Significant correlation ($R^2 = 0.85$, $p < 0.05$) was observed for PCB-118 and -126 at EW sites in New Delhi, and their levels were also comparable. On the other hand, PCB-126 (Avg: 3.7 ng/m³) was significantly higher than PCB-118 (Avg: 0.3 ng/m³) ($p < 0.01$) in Chennai and Bangalore, indicating the difference in processes observed in EW sites of these cities and consistent with the higher contribution from combustion. Penta and hexa homologue congeners grouped in this factor were previously observed in soil samples from informal e-waste metal recovery sites.¹³ The ratio of PCB-118 to -77 varied from 0.1 to 1.9 for all EW sites except a metal recovery site at New Delhi (EW-03, Seelampur,) and an e-waste processing center in Bangalore (EW-07, K.R. Market). In these two EW sites, the ratio was >3 indicating metal smelting processes.⁷⁰ Overall current findings supports the previous evidence of New Delhi as the metal recovery hotspot of India, while other auxiliary processes are prevalent in southern cities.^{12,13} PCB-77, -105, -118, and -138 were dominant in Bangalore, while PCB-167 was dominant in Chennai. The presence of a high hazard indexed landfill near Goripalaya (EW-06) in Bangalore might account for this signal. Chennai's PCB homologous profile is similar to previous accounts in dust⁶⁸ and air.⁴⁷

More than half of the γ -plasticizer concentrations stemmed from EW transect, with major contributions from DEHP (51%), DEP (28%) and DnBP (13%). DnBP and DEHP are the PAEs most commonly released during e-waste recycling activities by the informal sector.³⁴ Acid leaching and pyrolysis practiced in metal recovery sites are expected to be a strong source of DEHP in Indian soils.¹² Strong correlations were found between DEHP and PCBs in Factor 1 (EW) such as PCB-156, -169, -180, and -178 ($R^2 = 0.6–0.8$, $p < 0.05$). Further, DEHP showed significant correlation with carcinogenic PAHs ($R^2 = 0.967$, $p < 0.05$). High contributions of DEHP ($>50\%$) from e-waste can be attributed to its release during burning of wires/cables practiced predominantly in informal recycling of e-waste as observed at Luqiao district in China's Zhejiang province.³⁴ Nearly 30% of the higher molecular weight PAHs like DahA, BgP, and BkF were attributed to the e-waste sites. 5–6-ring PAHs are most likely produced due to incomplete combustion of e-waste.^{71,72} Flu/(Flu+Pyr) and Ant/(Ant+Phe) indicate that in Chennai and Bangalore the sources are mostly petroleum combustion, whereas in New Delhi they are mostly combustion products. Combustion using coal and/or wood is being carried out for e-waste recycling processes may contribute to higher concentrations of Pyr, Flu, Phe, and HMW PAHs at e-waste sites. Emission and re-emission from contaminated soil at the EW sites can be attributed for the dominance of DEHP and dl-PCBs in the EW sector.^{12,13}

3.2.2. Factor 2: Open Burning.—PCB-18, -37, -44, -52 were the major congeners loaded in this factor. These PCB congeners were observed in samples influenced by open burning in dumpsites. Dumpsites in India receive a concoction of waste from various sources without any source segregation. Further, in order to reduce the quantity of waste, open burning in the dumpsites is a common practice. These congeners accounted for over one-third of total PCB concentrations in this study. DS-01 and DS-02 are the sites located within 500m of two major dumpsites of Chennai city, Kodungaiyur and Perungudi, respectively. Around 2500 tonnes of unsegregated waste is dumped in Kodungaiyur dumpsite per day. A strong correlation was observed for tetra congeners between the aforementioned sites ($R^2 = 0.99$, $p < 0.05$). PCB-52 has been the predominant congener in this study, contributing 20% of total PCBs and can be associated with open burning of municipal waste in Indian cities.^{30,47,54} Open burning of waste is a recurrent and ubiquitous solid waste management issue in India that has resulted in its widespread. Re-emission of low molecular weight tetra congeners from such hotspots is a secondary source of PCB-52 in this region.⁶⁹ Nearly 25% of PCB-169 and loading of other dl-PCBs in this factor can be associated with incomplete combustion of dumped waste.

Although PCB-105 has been segregated in factor 1 (EW sector) as one of the prevalent congeners, highest concentration was recorded at DS-01 (6 ng/m^3). Further PCB-105 was not observed in any other dumpsites. Dumping of waste sludge from the EW sites closer to DS-01, and chances of nonmunicipal hazardous waste fated to open burning can be reasoned with high concentrations of dl-PCBs in this dumpsite.¹³ It is important to note that this trend was not observed for DS-02, since Perungudi dumpsite is approaching defunct status and land reclamation has reduced the area available for dumping by 60%.⁷³

Like PCBs, the highest concentrations of plasticizers and PAHs such as Chr, BaA, Pyr were observed at DS-01. This observation can be attributed to the emission and re-emission from contaminated soil in open dumpsites.¹² DnBP contributed 36% of total plasticizers followed by DEHA (17%). DnBP is mostly used in retail packaging material and low density polyethylene products. Recent study in particulate matter from landfill sites in New Delhi reported concentration up to 379 ng/m^3 of DnBP can be released due to open burning of waste containing plastics.⁷⁴ Interestingly, major contribution of DEHA stemmed from this source factor. Previously, automobile shredder residue discarded in dumpsites was suggested to be a potential source of DEHA in dumpsite soil of India.¹²

3.3.3. Factor 3: Activities Related to IT Corridor.—PCB-28, -33, and -170 were the three major congeners in this factor. This factor contributed 10% of total PCBs. Maximum PCB-28 was recorded from Bangalore (IT-04) while highest PCB-170 was observed in Gurugram, the IT hub in the outskirts of New Delhi (IT-03). The presence of dismantling/shredding in the IT corridor is likely responsible for the dominance of the tri PCB congener, PCB-28. IT sites in major Indian cities generate over 65% of e-waste in the country, with the major metropolitan cities (current study locations) responsible for the largest share. Preliminary processes like e-waste dismantling and shredding are suspected to take place within the zone of generation, to reduce the load on transportation to e-waste metal recovery sites. While other suspected contributors of PCBs from in-use and stored materials have remained consistent at these sites, the proliferation of IT sector and allied

industries have emerged as significant waste generating sectors in India.⁷⁵ Similar to PCBs, DEHP and DnBP were dominant in IT transect but with relatively 2–3 fold lower levels than EW transect. Maximum levels of benzo compounds viz., BbF, BkF, BgP could be due to mixed anthropogenic activities in the growing IT corridor.

3.3.4. Factor 4: Industrial emission.—PCB-66, -70, -74, -101, -123 were dominant in this factor and represented the industrial transect. Over 70% of these congeners were recorded from industrial sites of Chennai. PCB-123 has been previously evidenced as the dominant congener associated with industrial emission in urban and suburban atmosphere of Chennai.⁴⁷ PCB-123 was at least three fold higher at Ambattur Industrial estate, Chennai (site IN-03) compared to other designated industrial zones. Diagnostic ratios viz., $123/(118 + 123)$ and $123/(114 + 123)$ resulted in a standard deviation of 0.17 and 0.45 respectively, which points to the combustion sources.⁷⁶ Common industrial processes that can emit PCB-123 are sintering,⁷⁷ kiln operations,⁷⁸ and wide array of thermal process.⁷⁹

Although DEHP contributed to 50% of the total plasticizers in the industrial area, 24% of the total BBP in the study came from industrial transect. Among PAHs, significant correlation was observed between BaP and Σ plasticizers in the IN sites ($R^2 = 0.828$, $p < 0.05$). Furthermore, significant correlation was observed between BBP and PCB-66 ($R^2 = 0.66$, $p < 0.05$) and PCB-70 ($R^2 = 0.894$, $p < 0.05$) in the IN transect. BBP may be released in the air through the combustion of refuse,⁸⁰ and it has also been detected in stack emissions from hazardous waste combustion facilities and from coal burning power plants.⁸¹ However, the contributions of Ant and BaP were visibly higher than other PAHs in industrial sites. Industrial transect contributed nearly 75% of total BaP and more than 50% of total Ant. BaP has been seen as a by-product of incomplete combustion and pyrolysis of carbon-containing fuels⁸² and Ant has been majorly observed in wood preservatives and manufacturing of dyes and pigments.⁸³ Chr and BaA are markers for coal combustion.⁸⁴ Use of coal for industrial purpose might have resulted in higher concentration of BaA and Chr in industrial sites. Furthermore, Flt/(Flt+Pyr) indicated mixed source while Ant/(Ant+Phe) indicate combustion sources.

3.3.5. Factor 5: Residential Emission.—Hexa and hepta congeners, PCB-156, -169, and -178 were majorly loaded in this factor. This factor accorded about 11% of total PCBs. PCB-156 and -169 have been related with vehicular emission in both metropolitan⁴⁷ and nonmetropolitan⁵⁴ atmosphere of India. Sampling locations are close to arterial roads of the city witnessing intense vehicular movement through the day. PCB-169 levels correlated with vehicular average numbers ($R^2 = 0.85$, $p < 0.05$) recorded in these Indian cities. The catalytic reduction process in internal combustion engines especially diesel engines are major sources of these congeners.⁸⁵ It is important to note that PCB-169 showed a strong correlation with PCB-126 at RS sites ($R^2 = 0.95$, $p < 0.05$) thereby indicating combustion sources from nearby hotspots. Significant correlation was observed between DEHP with Pyr ($R^2 = 0.85$, $p < 0.01$) and DahA ($R^2 = 0.9$, $p < 0.01$). Furthermore, Σ plasticizers showed significant correlation with Σ 15PAHs ($R^2 = 0.9$, $p < 0.05$) in RS sites. In RS sites, dominance of Nap and ratios of Flt/(Flt+Pyr) and Ant/(Ant+Phe) ratios >0.2 indicated petroleum combustion and vehicular emission as the prime sources for PAHs.

3.4. Risk Assessment.

Concentrations at EW transect consistently yielded the highest overall TEQ followed by the dumpsite for PCBs and PCDD/Fs (SI Tables S7a and S8). Current PCB-TEQ values are comparable to e-waste processing zones of southeast China⁸⁶ and lower than transformer recycling sites of Taizhou, although PCB-126 is comparable.⁸⁷ Reported TEQs are higher for PCBs from waste processing sites in Africa⁵⁸ and Bangladesh.¹⁰ Average atmospheric PCB concentration at EW transect (14.8 ng/m³) slightly exceeded the USEPA recommended inhalation unit risk health guideline.⁸⁸ IT corridors yielded the highest PAH-TEQs, due to elevated concentration of three benzo PAHs (BbF, BkF and BgP) (SI Table S8). Maximum estimated daily intake of PCBs via inhalation was at least two fold higher in the EW transect over other transects (Table S9). Hence people working in the EW sector might be exposed to high inhalation risk.⁸⁹ Furthermore, in EW transect, the median value of the estimated WHO tolerable daily intake via inhalation expressed as TEQs for dioxin like compounds was the highest and, exceeded the maximum recommended limit (1–4 pg TEQ/kg bw-day)⁹⁰ for youth and adult. This was due to high dl-PCBs in the EW sites and PCB-126 was the major contributor. Estimated inhalation risk for youth and adult due to plasticizers was maximum in EW transect of Bangalore, followed by Chennai and New Delhi.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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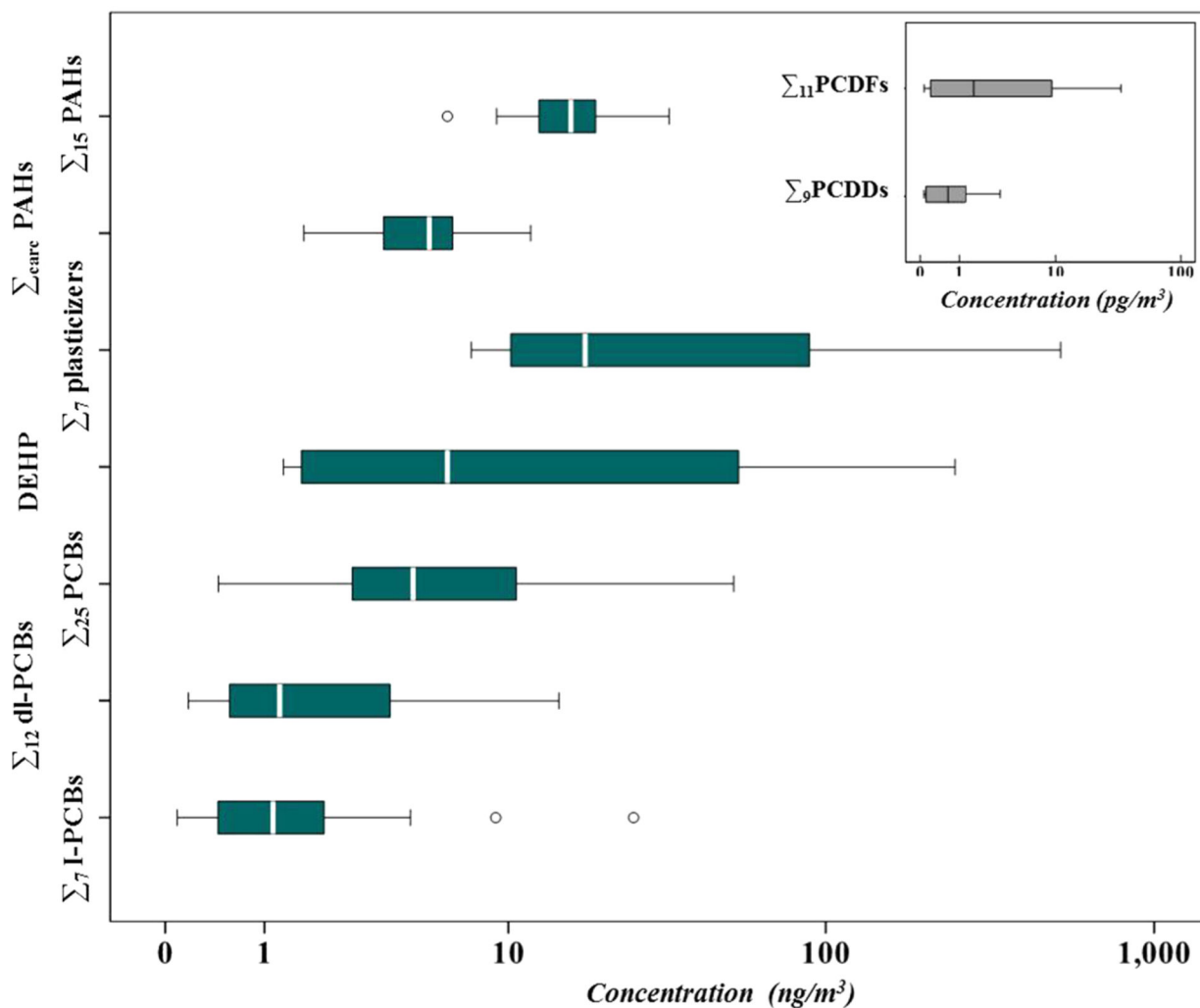


Figure 1.

Box whisker plots showing the range of polychlorinated dibenzo dioxin/furans (PCDD/Fs), polychlorinated biphenyls (PCBs) including indicator PCBs (I-PCBs), dioxin like PCBs (dl-PCBs), plasticizers, bis 2 ethylhexyl phthalate (DEHP), polycyclic aromatic hydrocarbons (PAHs), and carcinogenic PAHs (carc PAHs) in Indian mega cities. The central box represents the concentration values from the 25–75th percentile, and the bold white line represents the median. The horizontal line extends from the minimum to the maximum value, excluding the outliers (hollow circle) displaying the high/low concentrations values.

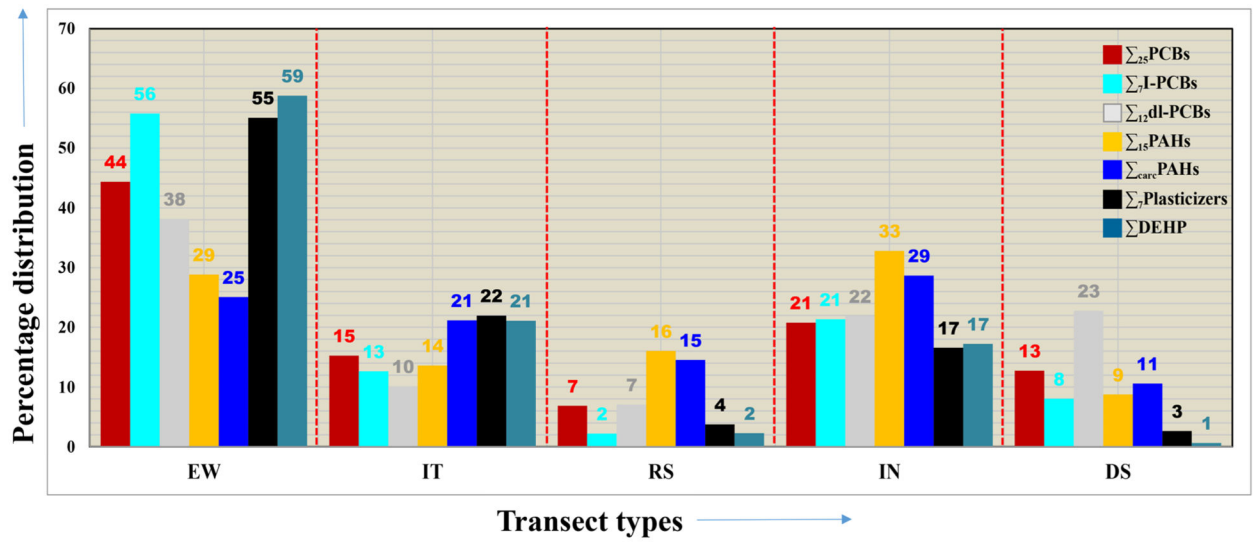


Figure 2. Percentage plot showing distribution of most abundant compounds (PCBs, plasticizers, and PAHs) across the five transects (EW: informal e-waste recycling sector; DS: open municipal dumpsites; IT: information technology corridor; IN: industrial area; RS: residential sites).

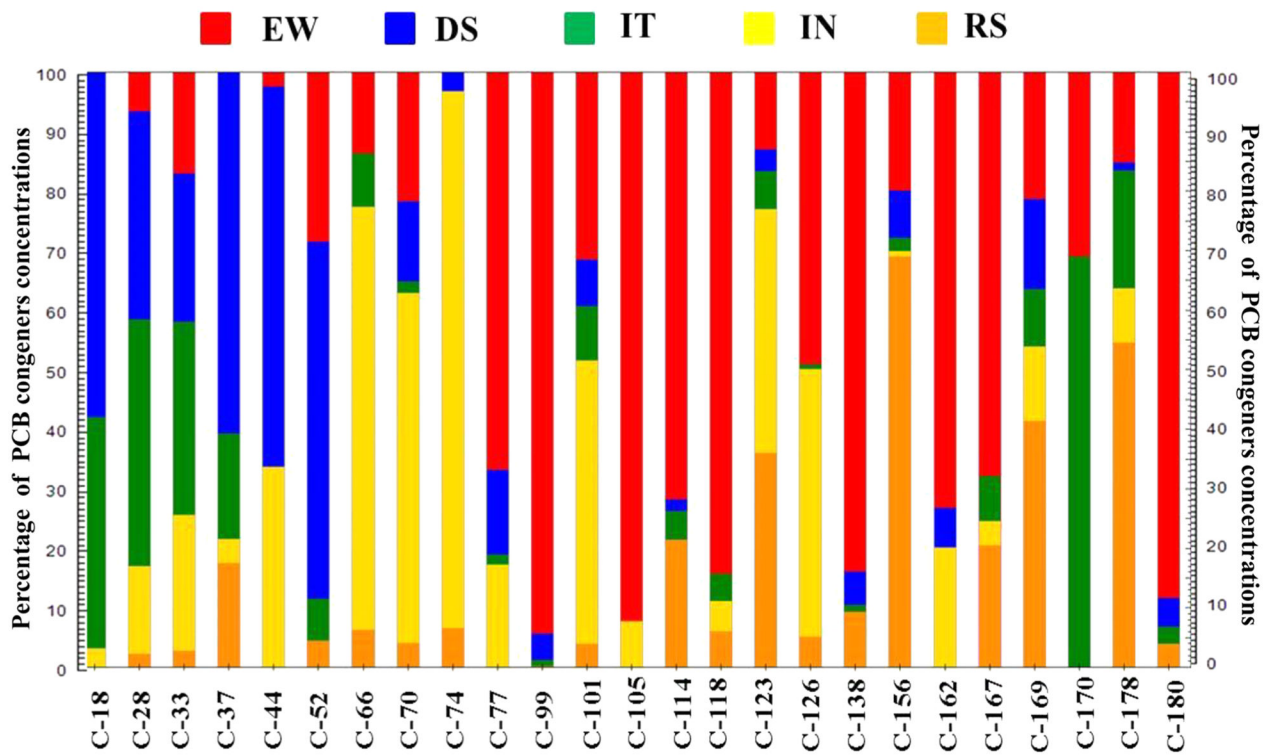


Figure 3. Positive matrix factorization fingerprints of each PCB congener for factor profiles representing five transects (EW: informal e-waste recycling sector; DS: open municipal dumpsites; IT: information technology corridor ; IN: industrial area; RS: residential sites).