Environmental Science & Technology

Article

pubs.acs.org/est

High Concentrations of Organic Contaminants in Air from Ship Breaking Activities in Chittagong, Bangladesh

Therese H. Nøst,[*,†] Anne K. Halse,[‡] Scott Randall,[‡,§] Anders R. Borgen,[‡] Martin Schlabach,[‡] Alak Paul,[∥] Atiqur Rahman,[∥,⊥] and Knut Breivik[‡,#]

[†]FRAM Centre, NILU—Norwegian Institute for Air Research, P.O. Box 6606 Langnes, 9296 Tromsø, Norway

[‡]NILU—Norwegian Institute for Air Research, P.O. Box 100, 2027 Kjeller, Norway

[§]Division of Environment, Economics and Planning, COWI AS, P.O. Box 123, 1601 Fredrikstad, Norway

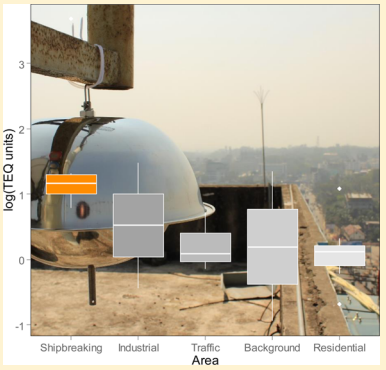
[∥]Department of Geography and Environmental Studies, University of Chittagong, 4331 Chittagong, Bangladesh

[⊥]Nanjing University of Information Science and Technology, School of Atmospheric Physics & Atmospheric Environment, 210044 Nanjing, China

[#]Department of Chemistry, University of Oslo, P.O. Box 1033, 0315 Oslo, Norway

Supporting Information

ABSTRACT: The beaches on the coast of Chittagong in Bangladesh are one of the most intense ship breaking areas in the world. The aim of the study was to measure the concentrations of organic contaminants in the air in the city of Chittagong, including the surrounding ship breaking areas using passive air samplers (N = 25). The compounds detected in the highest amounts were the polycyclic aromatic hydrocarbons (PAHs) and short-chain chlorinated paraffins (SCCPs), whereas dichlorodiphenyltrichloroethanes (DDTs), hexachlorobenzene (HCB), and polychlorinated biphenyls (PCBs) were several orders of magnitude lower in comparison. PCBs, PAHs, and HCB were highest at sites near the ship breaking activities, whereas DDTs and SCCPs were higher in the urban areas. Ship breaking activities likely act as atmospheric emission sources of PCBs, PAHs, and HCB, thus adding to the international emphasis on responsible recycling of ships. Concentrations of PAHs, PCBs, DDTs, HCB, and SCCPs in ambient air in Chittagong are high in comparison to those found in similar studies performed in other parts of Asia. Estimated toxic equivalent quotients indicate elevated human health risks caused by inhalation of PAHs at most sites.



INTRODUCTION

Environmental impacts of hazardous anthropogenic substances is a global concern which has motivated international regulatory initiatives such as the Stockholm[1] and Basel Conventions.[2] Persistent organic pollutants (POPs) and their potential harmful effects on humans and the environment have been researched extensively in industrialized countries, but far less is known about POPs emission sources and environmental concentrations in developing countries. Bangladesh is classified as a Least Developed Country according to the United Nations (UN). In addition, Bangladesh has ratified the Stockholm and Basel Conventions. Since the 1980s, national environmental regulations have existed in Bangladesh, such as the ban of many organochlorine pesticides and polychlorinated biphenyls (PCBs).[3] There are several suspected local emission sources of POPs in Bangladesh;[3−5] however, there are few studies monitoring concentrations of POPs in the environment in this country, and no studies have been conducted in Chittagong.

Chittagong is the second largest city in Bangladesh and has a population of approximately 4.5 million people.[6] The city hosts a large seaport and extensive industrial activities in and around the city. Ship breaking in Chittagong occurs when ships are dismantled directly on the tidal beaches. This activity began in the 1960s and has since represented a considerable share of the world market of ship breaking.[5,7−10] Out of a total of 1026 ships dismantled globally in 2014, 22% of the ships with known scrapping destinations were dismantled in the Chittagong area.[11] Concern has arisen regarding these activities and the subsequent emissions of pollutants to the environment.[12−14] The materials making up the scrapped ships contain heavy metals and industrial-use POPs like PCBs and polybrominated diphenyl ethers (PBDEs).[15] A large fraction of the volume of waste electric and electronic equipment (WEEE) in Bangladesh originates from the ship breaking activities.[5,16] Furthermore, there is no formal management and recycling of obsolete PCB-containing equipment, and this waste is simply deposited in landfills in many areas.[3,5] Burning of such waste could release POPs like PCBs to the air while also generating byproducts of

Received: June 25, 2015
Revised: August 28, 2015
Accepted: September 9, 2015
Published: September 9, 2015

DOI: 10.1021/acs.est.5b03073
Environ. Sci. Technol. 2015, 49, 11372−11380

combustion such as polycyclic aromatic hydrocarbons (PAHs).[17,18]

Other potential sources to contaminants in the ambient air in Chittagong area include release and/or generation of unintentionally produced PCBs and PAHs by industrial activities.[17−20] Also, there are suspected stockpiles and illegal use of pesticides in the area, especially of dichlorodiphenyltrichloroethane (DDT) which was produced in Chittagong until 1991.[3,4] Metal-working lubricants, sealants, plasticizers, and flame retardants are likely widely used in Bangladesh, which may contain short-chain chlorinated paraffins (SCCPs) that can also be considered a pollutant source.[21]

There is limited information on the environmental and human exposure to POPs as a result of ship breaking activities. The objective of this study was to monitor concentrations of PCBs, organochlorine pesticides, PBDEs, SCCPs, and PAHs in the air within the urban environment of Chittagong, including a focus on areas with ship breaking activities.

MATERIALS AND METHODS

Sample Collection. The sampling campaign was conducted in the Chittagong area using passive air samplers (PAS) equipped with polyurethane foam (PUF) disks.[22] PAS are regularly used for spatial mapping of pollutants in air monitoring studies on both a regional and global scale.[23−27] Detailed descriptions of this sampling method can be found in the literature.[22,25,26,28] In brief, the PUF disk has a high ability to sorb semivolatile organic compounds in air and thus accumulate such compounds on the disk during the exposure period. The theoretical amount sorbed on the PUF disk is equivalent to the rate of uptake minus the rate of loss. The initial rate of uptake is linear, but declines as the concentration in the PUF approaches equilibrium. The duration of the linear phase of uptake is dependent on the octanol−air partition coefficient (K_{OA}) of the individual compounds. Thus, compounds that are more volatile (low K_{OA}) will approach equilibrium faster than less volatile compounds (higher K_{OA}).

A total of 25 PAS were deployed at 23 different sites in Chittagong and near the ship breaking areas located northwest of the city center in late February 2013 (Supporting Information, Figure S1). The study was performed in parallel to a screening study for concentrations of sulfur dioxide (SO_2), nitrogen dioxide (NO_2), and ozone (O_3) in ambient air of Chittagong.[29] Deployment time for the PAS ranged from 7 to 9 days (see Supporting Information, Table S1, for details). A short deployment time was deliberately selected (i) because high frequencies of detection were anticipated for most analytes and (ii) to enhance the potential for sampling during the linear uptake phase, reflecting the high temperatures expected during the sampling period in combination with the volatility of the target compounds. The samplers were assembled, deployed, and disassembled by trained personnel from Chittagong University. The sampling campaign also included preparation of field and transport blanks (N = 4 each).

The research team selected sampling sites in residential, industrial, and background areas as well as in areas in close proximity to ship breaking activities in Chittagong. After the exposure period in the field, the samplers were disassembled, and the PUF disks were wrapped in two layers of aluminum foil, placed in sealed plastic bags, and stored in a freezer until analysis.

The screening study was performed during the dry season, and there was no precipitation recorded during the sampling period. Temperatures during the sampling period were consistently high; 25−30 °C during the day and around 15 °C during nighttime. The wind frequency distribution (wind rose) as measured at the Agrabad continuous air monitoring station in Chittagong is presented for the sampling period in Figure S2. The prevailing wind direction in this area was from the west−northwest (Bay of Bengal) with wind speeds between 1 and 4 m/s.

Sample Preparation, Cleanup, and Analysis. Sample preparation, cleanup, and analysis were performed as described in detail by Halse et al.;[27] only a brief summary along with minor adjustments is provided here. The PUF disks were precleaned using Soxhlet extraction (toluene for 24 h, acetone for 8 h, and toluene for additional 8 h) before deployment. The disks were wrapped in two layers of aluminum foil, placed in sealed plastic bags, and stored in a freezer until shipment. After the exposure period, PUF disks were spiked with internal standards and Soxhlet extracted using *n*-hexane/10% diethyl ether (8−10 h, 250 mL). An aliquot (10% volume of extracts) was stored for possible future analysis. The remaining extracts were divided into two identical aliquots for two separate cleanup procedures.

One aliquot targeted acid-resistant compounds: PCBs (congeners 28, 52, 101, 118, 138, 153, 180), PBDEs (congeners 47, 99), hexachlorocyclohexanes (α-, β-, and γ-HCH), dichlorodiphenyltrichloroethanes and one metabolite (DDTs; *p,p′*-DDT, *o,p′*-DDT, *p,p′*-DDE), hexachlorobenzene (HCB), chlordanes (*trans*-chlordane, *cis*-chlordane, *trans*-nonachlor), and SCCPs. This aliquot was treated with concentrated sulfuric acid (2 mL, 2−3 times) and further cleaned up using a silica column of activated silica (4 g, preheated 8 h at 550 °C) and eluted with *n*-hexane/10% diethyl ether (30 mL). The other aliquot targeted PAHs (fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[*a*]anthracene, chrysene/triphenylene, benzo[*a*]pyrene). As PAHs are acid-labile and difficult to elute from activated silica, a slightly different cleanup method was applied. The solvent was exchanged to cyclohexane before cleanup using a deactivated silica column (slurry of 5 g silica deactivated with 8% Milli-Q water, and 15 mL cyclohexane) and eluted with cyclohexane (100 mL). After cleanup procedures, the extract aliquots were reduced to ∼50 μL, and recovery standards were added.

The analyses of PCBs, PBDEs, HCHs, DDTs, and HCB were performed by gas chromatography coupled to a high-resolution mass spectrometer (GC-HRMS) in electron impact mode.[27] SCCPs and chlordanes were determined by GC-HRMS in an electron capture negative ionization mode and reported as bulk mixture.[30] PAHs were analyzed by gas chromatography coupled to a low-resolution mass spectrometer (GC-LRMS) in electron impact ionization mode.[27]

Quality Assurance and Quality Control. The analytical procedures were performed according to NS/EN ISO/IEC 17025 accredited routines. A calibration/quantification solution containing all target analytes was analyzed along with the samples. Also, analysis of standard reference materials (SRMs 1588 and 2206a, U.S. National Institute of Standards and Technology) assured low analytical uncertainties of PCBs, PBDEs, HCHs, DDTs, HCB, and chlordanes. Further, compounds with a retention time of more than 3 s later than corresponding isotope-labeled standards as well as compounds with isomer isotopic ratios deviating >20% from those in the standards were disregarded. Average recoveries for internal

DOI: 10.1021/acs.est.5b03073
Environ. Sci. Technol. 2015, 49, 11372−11380

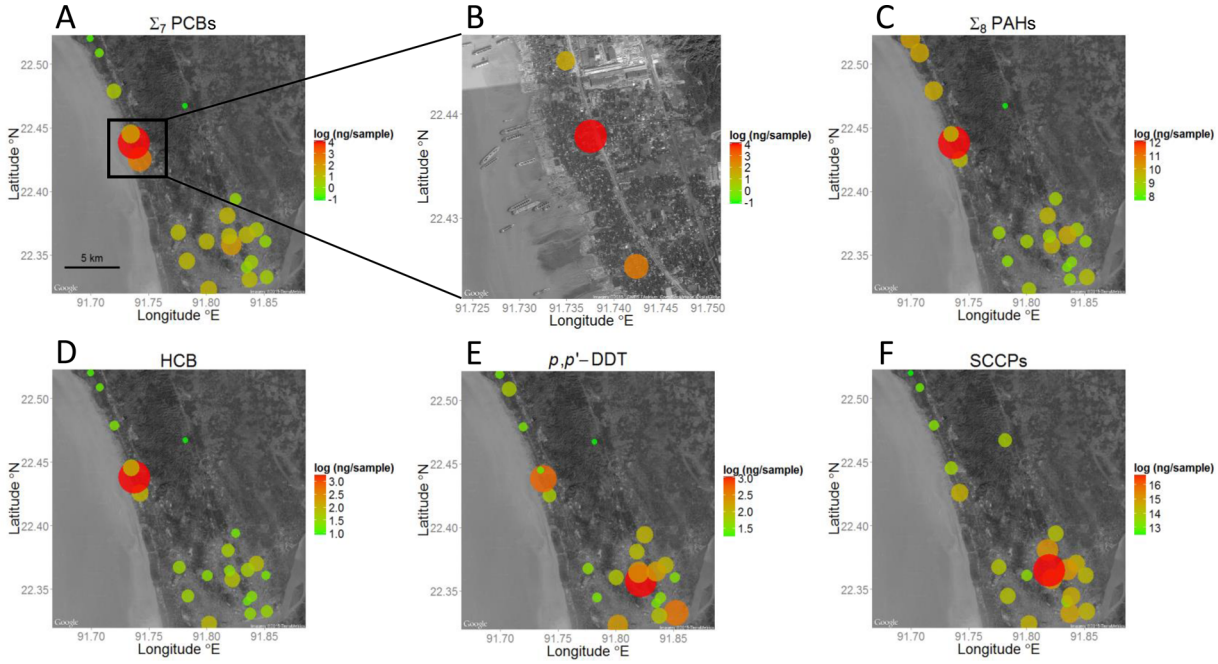


Figure 1. Sample amounts of (A) Σ_7PCBs (congeners 28, 52, 101, 118, 138, 153, and 180); (B) Σ_7PCBs in zoom image of sites with elevated amounts close to the ship breaking area; (C) Σ_8PAHs (fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[*a*]anthracene, chrysene/triphenylene, and benzo[*a*]pyrene); (D) HCB; (E) *p,p′*-DDT; and (F) SCCPs. Results are presented as ng/sample, and ranges differ between plots (given in legends).

standards ranged 48−113%, and their ranges for the samples and blanks are presented in Table S2.

Field (N = 4), transport (N = 4), and laboratory method samples (N = 3 blanks) were extracted and analyzed to reveal any contamination during handling, transport, and laboratory preparations. Field blanks are defined as PUFs that have been assembled in PAS in the field and then immediately disassembled; transport blanks are PUFs which were never exposed in the field; and laboratory method blanks are simply clean PUFs. The ranges of blank amounts are presented in Table S3. Method detection limits (MDLs) were determined for each compound from average blank amounts plus three times the standard deviation of these amounts. When the target compound was not identified in the blanks, an instrumental detection limit (signal/noise 3:1) was used to estimate a MDL value.

Parallel samplers were deployed at two sampling sites. Between parallels, the relative standard deviations were 0.1−27% for PCBs, 7.8−21% for PBDEs, 2.6−28% for PAHs, 3−39% for HCHs, 4−34 for *p,p′*-DDT, 4−28% for HCB, 6% for SCCPs, and 0−4% for chlordanes. The average value of the parallel samples represented sample results at these sites.

Estimation of Concentrations in Air. A sampling volume estimate is needed to calculate concentrations in air from the measured amounts of POPs in the PAS. Still, the most appropriate approach for back-calculation of concentrations in air from PAS sampling is currently debated, and sample volume estimations do include uncertainties.[31−34] In the current study, a range for the estimated sampling volume was obtained for each compound as proposed by Harner et al.[35] The estimation is based on duration of exposure, average air temperature, PUF characteristics, compound-specific K_{OA} values, and assumptions of sampling rates and particle fractions (see further details in the Table S5).

Data Treatment and Analysis. All 23 sites were included in the data analysis. Statistical analysis was executed using R, ver.3.1.1., and statistical significance was defined as $p < 0.05$. When summed amounts were calculated for POP groups, MDL/$\sqrt{2}$ was used for sample amounts that were lower than the MDL. Summary statistics for compounds with detection frequencies between 20% and 80% were calculated using the Kaplan−Meier method with the NADA package for R according to Helsel.[36] Correlations between components are presented as Spearman's rank correlations. Spatial presentation of the data was performed with the ggplot2 and ggmap packages in R, in which the background maps are provided by Google Inc. Due to uncertainties in back-calculated concentrations in air, spatial patterns are interpreted on the basis of measured amounts. Principal component analyses (PCA) were performed with the ggbiplot package in R on the compounds presented in Table S3 (SCCPs were excluded due to missing values).

Estimations of Human Health Risks from Inhalation Exposure. Toxic equivalent quotients (TEQs) were calculated based on toxic equivalency factors (TEFs) for PAHs[37] (fluorene = 0.001, phenanthrene = 0.001, anthracene = 0.01, fluoranthene = 0.001, pyrene = 0.001, benz[*a*]anthracene = 0.1, chrysene/triphenylene = 0.01, benzo[*a*]pyrene = 1), and PCBs[38] (PCB-118 = 0.000033), as well as the PUF-derived concentrations in air for each site.

RESULTS AND DISCUSSION

Results Summary. The measured amounts were highest for PAHs and SCCPs, whereas PCBs, DDTs, chlordanes, HCB, and PBDEs were several orders of magnitude lower in comparison. The correlations between compounds within each of the PAH and PCB groups were strong (most Spearman's rho >0.8), whereas correlations were weaker, or not significant, for PBDEs and pesticides (Table S4).

DOI: 10.1021/acs.est.5b03073
Environ. Sci. Technol. 2015, 49, 11372−11380

The levels for Σ_7PCBs were highest at three sampling sites located close to ship breaking activities, and especially at one site where measured amounts were 19 times higher than the median of Σ_7PCBs (Figure 1B). Still, the amounts of Σ_7PCBs at the other sampling sites were also considerable (Figure 1A). The lower-chlorinated PCB congeners contributed most to the Σ_7PCBs (34%, 27%, and 15% of Σ_7PCBs for PCB-28, -52, and -101, respectively; Table S3).

The levels of PBDEs were low, and results are presented only for PBDE-47 and -99 due to quality control measures and limitations of the sampling method. The highest amounts of the two PBDE congeners were found at sites in the city center.

The sites with the highest levels of Σ_8PAHs (Figure 1C) paralleled those of the PCBs, and there was moderate correlation of many PAHs with PCBs (Spearman's rho >0.3; Table S4). Phenanthrene (46%), fluoranthene (18%), fluorene (17%), pyrene (14%) contributed most to the median Σ_8PAHs.

The highest measured amounts of HCB was found at one site in the vicinity of ship breaking activities (Figure 1D) which was the same site that had the extreme values for Σ_7PCBs and Σ_8PAHs. However, HCB levels were high also at other sites in the ship breaking area and in the city.

The most abundant pesticides found in the study were DDT and DDE. The highest levels of *p,p′*-DDT (Figure 1E) and *o,p′*-DDT (data not shown) were found at sites in urban areas. The spatial trends for HCHs diverged between the three isomers measured (Figure S3). Levels of α-HCH were highest at the urban sites, whereas β-HCH levels were highest at one site near ship breaking activities as well as at numerous urban sites. Levels of γ-HCH were highest at sites close to ship breaking activities. Sample amounts of chlordanes were low, and spatial patterns (presented for *trans*-chlordane in Figure S3) resembled those for γ-HCH.

The highest levels of SCCPs were found in the urban area (Figure 1F), and there appeared to be a declining trend of these levels with increasing distance from the city center. SCCP results for the site closest to ship breaking activities that was highest for PCBs and PAHs were unfortunately not available, as this sample was damaged during laboratory analyses.

Interpretation of PAH Molecular Ratios. Molecular diagnostic ratios (MDRs) are ratios between certain PAHs which can indicate characteristics related to their emission sources.[39−41] Sampling performed in narrow time spans across limited geographical areas close to suspected sources has been suggested to facilitate interpretation of MDRs and renders the present study interesting in this context. Three MDRs were calculated on the basis of PAHs with high analytical detection (Figure 2). The ratios of anthracene/anthracene + phenanthrene (ANT/(ANT + PHE)) and benz[*a*]anthracene/benz[*a*]anthracene + chrysene (BaA/(BaA + CHR)) were significantly different between site classifications (five classes, see Table S1; Kruskal−Wallis test, $p = 0.004$ for both group tests). It should be noted that quantified amounts of chrysene include uncertainties due to analytical coelution with triphenylene. The third MDR representing fluoranthene/fluoranthene + pyrene (FLT/(FLT + PYR)) was not significantly different across classes (Kruskal−Wallis test, $p > 0.05$).

MDR values above given cutoff values indicate combustion of grass and wood rather than fossil fuel as sources to PAHs.[40,41] Ratios were above the respective cutoff values at four sites for ANT/(ANT + PHE) and at six sites for BaA/(BaA + CHR) in the ship breaking area. In addition, ratios were above the respective cutoff values at three sites for BaA/(BaA + CHR) that were classified as industrial or affected by traffic within the city. Together, MDR values were high at sites near ship breaking activities and at some industrial sites, which suggests that PAHs originate from combustion processes at these sites. The FLT/(FLT + PYR) ratio was above the cutoff value at all sites, which could be indicative of widespread combustion of wood and grass, etc. Compared to coastal, urban, and agricultural sites in southern India, ratio values for BaA/(BaA + CHR) were higher at the sites close to ship breaking activities in Chittagong, and FLT/(FLT + PYR) values were in the same range.[42]

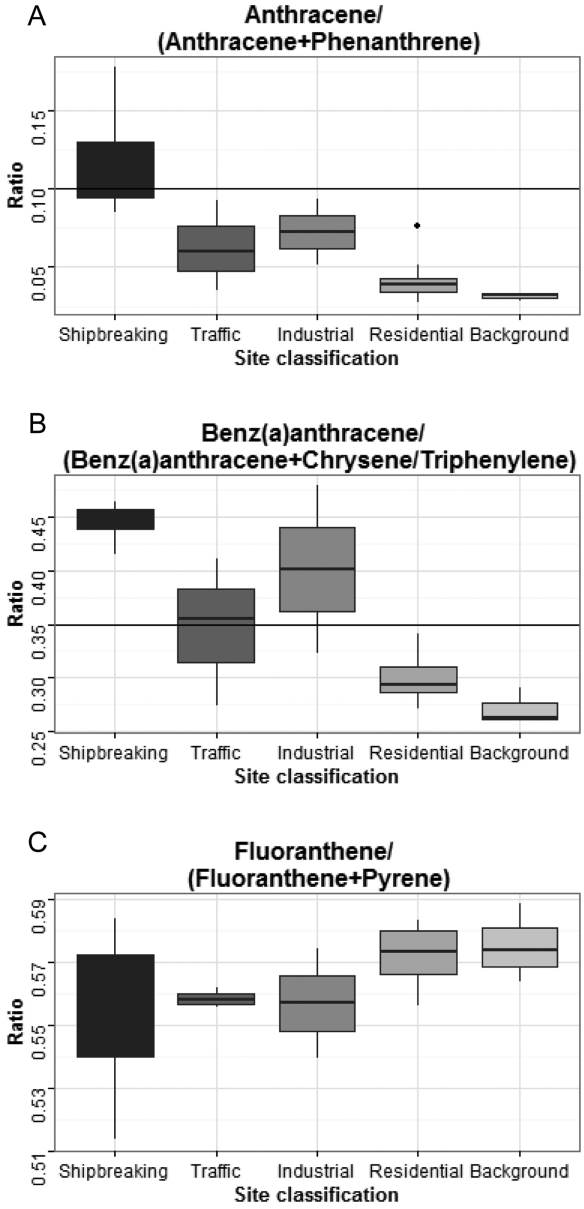


Figure 2. Molecular diagnostic ratios for three pairs of PAHs for five site classifications. The horizontal lines in plots A and B represent a cutoff value for pyrogenic sources,[40] and the differences across classes were significant for the ratios in these two plots, but not for those in plot C.

Methodological Limitations. PUF-based PAS was selected as sampling material on the basis of its performance in spatial mapping on both a regional and global scale (see references in Materials and Methods section). However,

DOI: 10.1021/acs.est.5b03073
Environ. Sci. Technol. 2015, 49, 11372−11380

employing PUF-based PAS as sampling method of relatively volatile substances has both advantages and drawbacks.[32] In order to focus the discussion and avoid artifacts due to choice of sampling method, the presented results have been limited to compounds in a selected range of K_{OA} values (Table S3; ranging 6.21−11.38 at 24 °C, which was the average estimated temperature for the sampling period). An additional limitation is that the sampling could be influenced by high amounts of ambient particles as well as the fact that there was no precipitation to settle the particles during the sampling period.[29] In addition to particle impaction, the high temperatures found during the sampling period may have enhanced the uptake of both the gas phase and the particle-bound phase of POPs during deployment.[22] There is likely some variation in particular concentrations and temperatures within the study area that might have affected the contaminant uptake at some sampling sites, but the spatial comparisons are still considered valid. Overall, the sampling period provides a temporal snapshot in the area with regard to both seasonal variability related to changing intensities of ship breaking and industrial activities. There was no apparent trend with height of sampler position above sea level.

PUF-derived concentrations in air are presented even though they include uncertainties as exact normalization to measured sampling volumes was not possible. Therefore, a range of sampling volumes was estimated for each compound on the basis of ranges in temperatures and theoretical ranges in ambient particle fractions during the sampling according to Harner et al.[35] As the sampling rates vary by uptake phases, gas-particle partitioning, wind speed and temperature,[32] the estimates may appear crude, however, we have employed a compound-specific strategy to take account of the K_{OA}-specific variation in sampling volumes.

Local Emission Sources of POPs. This study presents ambient air data across an area that covers residential, industrial and ship breaking areas as well as sites that can be considered background sites. Poor air quality is an identified problem in several areas of Bangladesh, also in Chittagong.[29,43,44] However, to the extent of our knowledge no previous survey of POP concentrations in ambient air has been performed in Bangladesh. The results of the present study demonstrate high measured amounts of various POPs considering the short sampling period. Furthermore, the discrepancies in spatial patterns for PCBs, HCB, and PAHs as compared to DDTs and HCHs indicate different emission sources for the different compound classes. Specifically, the results indicate that ship breaking activities can be important potential local emission sources of PCBs, PAHs, and HCB. Furthermore, sites in urban areas appeared to be especially influenced by sources of DDTs and SCCPs. Together, these results add to the emphasis on challenges related to atmospheric pollution in Chittagong.

The PCA analyses demonstrated that the main variation in the sample set is related to the samples collected close to ship breaking activities and that the other site classes (industrial, traffic-influenced, residential, background) are distinguished from these samples (Figure S4). Overall, the PAHs, PCBs, and HCB were clustered together along the first factor away from *o,p′*-DDT and *α*-HCH, which agrees with the spatial trends of these compounds (displayed in Figures 1 and S3). The joint interpretation of spatial patterns in concentration trends, PAH MDRs, compound correlations and PCA patterns has provided indications of potential emission sources for the different contaminants. Overall, our results are in accordance with previously suggested potential emission sources such as release and/or production of contaminants related to ship breaking and industrial activities, as well as influence of stockpiles of pesticides or PCBs.[4]

Ship Breaking Activities. The levels of PCBs, PAHs, and HCB were high at sites close to the beach, and the spatial trends for these compounds indicate the role of ship structures and their dismantling processes as potential local emission sources of these compounds. Many registered activities in this area are designated as iron and steel manufacture, re-rolling mills, and simply as ship breaking. The spatial trends for PCBs, PAHs, and HCB and their inter-correlations indicate common emissions of these compounds which could be related to release and/or formation through combustion processes as burning of scrap materials is common in the ship breaking areas.[12] This is further supported by the high MDR ratios indicative of combustion of wood and grass[43] at sampling sites classified as influenced by ship breaking. Ships may contain PCBs in WEEE (e.g., transformers and capacitors) and paints.[3,15] Furthermore, PCBs, PAHs, and HCB could be generated and/or released during combustion through burning of scrap and organic materials from the ships.[17−19] The elevated levels of the same POPs at urban sites could also be related to ship breaking activities, as there is subsequent processing and treatment of materials from the ships in Chittagong city.[12,29] Based on prevailing winds, the urban areas are downwind from the ship breaking sites, and transport by air can also have contributed to the sample amounts at sampling sites in the urban areas.

Industrial Sites. The sampling campaign was conducted in the dry season, which is when the most intense emission sources are active in Bangladesh. The sources include industrial activities, brick kilns, wood burning, metal smelters, road dust, motor vehicles, soil dust, and sea salt—which have all been identified as important sources of particulate pollution.[29,43,44] As the sampling campaign was conducted during a period in which industrial activity and particle fractions in the air was generally high,[29] the sample amounts of PAHs, PCBs, and HCB may also reflect sampling of target analytes sorbed onto particles.[32] A study describing a sampling campaign conducted in parallel to the present study has reported concentrations of NO_2 and SO_2 in ambient air. Their spatial distributions were largely in accordance with recognized industrial activities and traffic, respectively, but did not display high concentrations at sites near ship breaking activities.[29] Spatial patterns within the urban areas for POP concentrations that could be related to industrial activities crudely correspond with those of NO_2 and SO_2 concentrations; this only adds support to the interpretation of the industrial activities as contributing emission sources to PAHs, PCBs, and HCB in ambient air in the city. Accordingly, contributions from various industrial activities have been shown to contribute to the ambient air loading of PCBs[20] and could contribute to the spatial patterns of PCBs in urban air in Chittagong (Figure 1A).

Waste Sites. Burning of waste at waste dump sites is common in Bangladesh,[3] and these combustion processes can release PAHs, PCBs, and HCB.[18,19] There are waste sites in the urban areas which also could be contributing to the high measurements at urban sample sites (Figure 1A−D). As mentioned, further processing of materials from ships could result in waste in urban areas[3,29] thereby also contribute to emissions to ambient air at those sites.

Stockpiles and Use of Pesticides. The spatial trends for DDTs indicated emission sources in urban areas which could

DOI: 10.1021/acs.est.5b03073
Environ. Sci. Technol. 2015, 49, 11372−11380

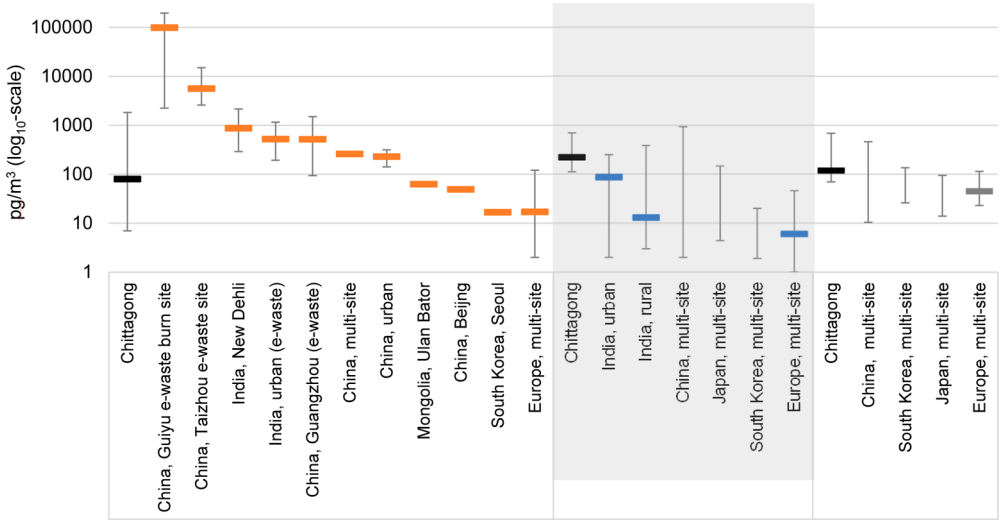


Figure 3. Concentrations in air (median and range) for Σ_7PCBs, *p,p′*-DDT, and HCB (from left or right) compared with results from other studies performed in Asia and Europe.[17,27,46,49,50,63−66,73] See text for further details.

be explained by historic production of DDT in the Chittagong area as well as stockpiles and continued illegal use.[3,4] The median *p,p′*-DDT/*p,p′*-DDE ratio was 1.92 (ranging 0.87−2.82), and the median *o,p′*-DDT/*p,p′*-DDT ratio was 0.48 (ranging 0.30−0.84). The high *p,p′*-DDT/*p,p′*-DDE ratios at some sites indicate that the emissions of DDT at these sites are recent.[45] Also, the ratio is higher at some urban sites in Chittagong compared to other contemporary studies in Asia[46,47] and Europe.[27] The low *o,p′*-DDT/*p,p′*-DDT ratios indicate that sources of DDT were technical DDT (ratio <1) and not dicofol (≫1)[48] usage which has been reported in other studies from Asia.[46,47,49,50] Together, these results support that there is recent use and/or emissions from stockpiles of DDT in the Chittagong area.

For the sum of HCHs, the median contribution from each isomer was 42%, 4%, and 54% for *α*-, *β*-, and *γ*-HCH, respectively. Similar contributions of both *α*- and *γ*- HCH to the sum HCH implies emissions of both technical HCH (*α*-HCH, 55−80%; *β*-HCH, 5−14%; *γ*-HCH, 8−15%) and lindane (>90% *γ*-HCH).[51] There were considerable amounts of *γ*-HCH found at the same sites that also contained the extremes for PCBs, PAHs, and HCBs at the sites near ship breaking activities. This could indicate their common release and/or signatures of continued or historic use of lindane at that site.

Sources of SCCPs. The spatial pattern of SCCPs shows elevated concentrations in the city (Figure 1F). This suggests that the main emission sources may be located within the urban areas. SCCPs are known to be found in metal-working lubricants, sealants, plasticizers, and flame retardants.[21] It is plausible that these usages may represent potential sources in Chittagong as well. SCCPs were in the same orders of magnitude as the PAHs, which shows that these compounds are abundant in air in the Chittagong area.

Human Health Risk Assessment. TEQ values based on PAHs and PCB-118 (ranging 0.39−40 ng TEQ/m^3; Table S5) were largely dominated by PAHs and above the carcinogenic risk limit (TEQ of >1 ng TEQ/m^3)[52] at 18 of the sites. This indicates that PAH exposure through inhalation cannot be considered safe at several sites and could involve elevated health risks. The highest value for TEQ was found at the site with the highest concentrations in the ship breaking area (Figure S5). It should be noted that the calculations (i) were based on a limited number of PAHs and only one PCB, and (ii) incorporate the uncertainties of the PUF-derived concentrations in air. Still, the relatively large exceedance of the threshold found at some sites indicate that further studies should address the health risks for humans in the areas with the highest concentrations of contaminants, with special consideration for the workers involved in the ship breaking activities. Possible exceedance of international standards and guideline values for health related to concentrations of SO_2 and NO_2 in ambient air has also been reported in this area.[29] The highest values in this study is higher than those reported in southern India where PAH-TEQs only exceeded the TEQ threshold in the summer months.[42] As this study was performed in the season during the most intense industrial activities, levels of PAHs and PCBs could be lower during the wet season, which also has less intense industrial activity. A follow-up study should therefore be performed to assess the seasonal variability of POPs in ambient air in Chittagong.

Ship breaking activities in Bangladesh employ over 100 000 workers, and an estimated 50 000 children are involved in informal e-waste recycling, of which 40% takes place in ship breaking yards.[5] Detrimental health effects have been emphasized following occupational exposure related to informal recycling processes of obsolete ships[12,53,54] and e-waste.[55−57] There are considerable health hazards for workers within ship breaking that are related to accidents in addition to the chemical exposures from asbestos, heavy metals, and POPs.[12,58] Furthermore, significant correlations between concentrations of various POPs in air (including PCBs, pesticides, and PBDEs) and human serum have been reported in China which emphasizes the relevance also of inhalation as exposure pathway in areas with considerable pollution.[59] Thus, the concentrations of POPs in ambient air in Chittagong could be detrimental to human health and add to the health hazards related to ship breaking activities.

Comparisons with Other Studies. Among the PAHs included in this study, phenanthrene was the most prevalent compound. This has also previously been observed in studies of background air performed in both Asia (India and South

DOI: 10.1021/acs.est.5b03073
Environ. Sci. Technol. 2015, 49, 11372−11380

Korea)[42,60] and Europe.[27] However, the estimated median and maximum concentrations of phenanthrene in this study were up to orders of magnitude higher than in the studies referred to above. Furthermore, the concentration of phenanthrene in this study was also higher than those in ambient air in Shanghai, China,[61] and at industrial sites in Taiwan.[62]

The estimated concentrations of Σ_7PCBs in air at sites close to ship breaking activities were within the range of other contemporary monitoring results in Southeast Asian areas influenced by informal e-waste treatment[17,63] or where e-waste treatment has been suggested to contribute to high concentrations[64,65] (Figure 3). The median concentration of Σ_7PCBs in air in Chittagong was comparable to those in large Asian cities[46,63,65,66] but higher than European background sites.[27] The PCB congener profile in air in Chittagong displayed less PCB-28 relative to the higher chlorinated congeners compared to that observed in multi-site studies in India and China[50,67] but was similar to that reported in multi-site studies in Europe.[23,27] In comparison to atmospheric PCB profiles resulting from industrial thermal processes, the observed contributions from PCB-28 were low, and can be considered similar to those for secondary copper or zinc smelting.[20] This result could be in agreement with the ship breaking and metal re-rolling activities in the area.

The current concentrations of DDT are within the ranges of those found in ambient air in India.[50] DDT is still actively used in India, which adds further support to the hypothesis of ongoing use of DDT in Chittagong. The concentrations of DDT were in the high range compared to other studies in China, Japan, South Korea, and Europe.[27,49]

Concentrations of HCB in Chittagong were also in the high range in comparison to concentrations found in the same studies mentioned above.[27,49]

Estimations of sampling volumes are not presented for SCCPs by Harner et al.,[35] but assuming a sampling rate of 4.2 m^3/day[68] for 8 days, the median concentration of SCCPs is 46 ng/m^3, which is several-fold higher than concentrations reported in India and Pakistan (sampled in 2006 and 2011, mean 8.11 ng/m^3, ranging from <LOD to 47.4 ng/m^3).[69] Although SCCPs is currently in production in China[21] the concentrations in Chittagong are within the range of those reported in China.[68] As there is no known production of SCCPs in Bangladesh, the concentrations observed in this study signify the probable widespread use of these compounds in Chittagong.

Considering the general decrease in POP environmental levels during the last decades,[70] the high concentrations found in this study call for future studies to assess if a temporal decline also extends to Chittagong.

Global Issue. These results add support to other studies indicating that ship breaking activities act as local emission sources of environmental contaminants and that there is considerable environmental and human health impacts associated with informal recycling activities.[8,13,71] It also adds to the growing body of literature which has documented elevated concentrations of legacy industrial-use POPs in developing regions, associated with recycling or disposal of imported wastes.[72] The results from this study emphasize the need for international engagement toward responsible management of end-of-life ships and cessation of illegal international transport of hazardous waste in the form of ships. As most WEEE in Bangladesh originates from ship breaking activities,[5] these issues may be intimately linked.

Closing Remarks. This study has provided data on PCBs, PBDEs, organochlorine pesticides, PAHs, and SCCPs found in ambient air in Chittagong using passive air samples. Ship breaking activities likely act as potential emission sources to the atmospheric burden of PCBs, PAHs, and HCB which adds to the international emphasis on responsible recycling of ships. Current or recent use of DDT stockpiles is likely contributing to the considerable ambient air burden of these compounds. Additionally, SCCPs were prevalent compounds found in this study that indicates their widespread use.

The concentrations of several compound groups in ambient air in Chittagong are high compared to contemporary studies in Asia, and elevated human health risks through inhalation exposure were indicated.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b03073.

Figures S1−S5 and Tables S1−S5, including details regarding sampling; sample recoveries; summary measures for measured amounts for POPs; additional spatial plots; compound correlations; compound-specific concentrations in air; PCA plot (PDF)

■ AUTHOR INFORMATION

Corresponding Author

*Phone: (+47) 77 75 03 86; fax: (+47) 77 75 03 76; e-mail: thn@nilu.no.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

The authors are indebted to the Chittagong University team which performed the field work in Chittagong. We also thank Mats Nordum for his contribution to this study, Dr. Nicholas Warner for reading the manuscript, and the Norwegian Research Council (FRIPRO 213577) for financial support.

■ REFERENCES

(1) Stockholm Convention. Stockholm convention on persistent organic pollutants. https://www.pops.int.

(2) Basel Convention. Basel convention on the control of transboundary movements of hazardous wastes and their disposal. http://www.basel.int.

(3) Environment and Social Development Organization (ESDO). Country situation report on persistent organic pollutants in Bangladesh; Dhaka, Bangladesh, 2005; http://ipen.org/sites/default/files/documents/1bgd_bangladesh_country_situation_report-en.pdf.

(4) Environment and Social Development Organization (ESDO). POPs hotspots in Bangladesh; Dhaka, Bangladesh, 2005; http://www.ipen.org/project-reports/pops-hotspots-bangladesh.

(5) Environment and Social Development Organization (ESDO). Study on E-waste: The Bangladesh situation; Dhaka, Bangladesh, 2010; http://www.esdo.org/images/Study%20on%20E-waste%20The%20Bangladesh%20Situation%202010.pdf.

(6) *The CIA world factbook*; https://www.cia.gov/library/publications/the-world-factbook/geos/bg.html.

(7) Puthucherril, T. G. *From shipbreaking to sustainable ship recycling: Evolution of a legal regime*. Koninklijke Brill NV: Leiden, The Netherlands, 2010.

(8) Hossain, M. M. M.; Islam, M. M. *Ship breaking activities and its impact on the coastal zone of Chittagong, Bangladesh: Towards*

DOI: 10.1021/acs.est.5b03073
Environ. Sci. Technol. 2015, 49, 11372−11380

sustainable management. Advocacy & Publication Unit, Young Power in Social Action (YPSA): Chittagong, Bangladesh, 2006.
(9) Environment and Social Development Organization (ESDO). Illegal import and trade off of e-waste in Bangladesh; Dhaka, Bangladesh, 2011; http://ipen.org/sites/default/files/t/2012/09/Report-on-Illegal-import-and-trade-off-of-e-waste.pdf.
(10) International Federation for Human Rights (FIDH). Where do the 'floating dustbins' end up? Labour rights in shipbreaking yards in South Asia: The cases of Chittagong (Bangladesh) and Alang (India); Paris, France, 2002; https://www.fidh.org/IMG/pdf/bd1112a.pdf.
(11) NGO Shipbreaking Platform Website; http://www.shipbreakingplatform.org/ngo-platform-report-list-of-all-ships-dismantled-worldwide-in-2014-january-2015/.
(12) Hossain, M. S.; Chowdhury, S. R.; Jabbar, S. M. A.; Saifullah, S. M.; Rahman, M. A. Occupational health hazards of ship scrapping workers at Chittagong coastal zone, Bangladesh. *Chiang Mai J. Sci.* **2008**, *35* (2), 370−381.
(13) Hasan, A. B.; Kabir, S.; Selim Reza, A. H. M.; Nazim Zaman, M.; Ahsan, A.; Rashid, M. Enrichment factor and geo-accumulation index of trace metals in sediments of the ship breaking area of Sitakund Upazilla (Bhatiary−Kumira), Chittagong, Bangladesh. *J. Geochem. Explor.* **2013**, *125*, 130−137.
(14) Hossain, M. M. M. Marine pollution in the Bay of Bengal and sustainable management. In *Ecosystem Health and Management of Pollution in the Bay of Bengal*; Hoq, M. E., Haroon, A. K. Y., Hussain, M. G., Eds.; Bangladesh Fisheries Research Institute: Dhaka, Bangladesh, 2011; pp 9−22.
(15) Hess, R. W.; Rushworth, D.; Hynes, M. V.; Peters, J. E. *Disposal options for ships*; RAND: Santa Monica, CA, USA, 2001; http://www.rand.org/pubs/monograph_reports/MR1377.html.
(16) United Nations Environment Programme (UNEP). *Regionally based assessment of persistent toxic substances, Indian Ocean regional report*; UNEP Chemicals: Châtelaine, Switzerland, 2002; http://www.chem.unep.ch/pts/regreports/IndianOcean.pdf.
(17) Xing, G. H.; Chan, J. K. Y.; Leung, A. O. W.; Wu, S. C.; Wong, M. H. Environmental impact and human exposure to PCBs in Guiyu, an electronic waste recycling site in China. *Environ. Int.* **2009**, *35* (1), 76−82.
(18) Ruokojärvi, P.; Ruuskanen, J.; Ettala, M.; Rahkonen, P.; Tarhanen, J. Formation of polyaromatic hydrocarbons and polychlorinated organic compounds in municipal waste landfill fires. *Chemosphere* **1995**, *31* (8), 3899−3908.
(19) Bailey, R. E. Global hexachlorobenzene emissions. *Chemosphere* **2001**, *43* (2), 167−182.
(20) Liu, G.; Zheng, M.; Cai, M.; Nie, Z.; Zhang, B.; Liu, W.; Du, B.; Dong, S.; Hu, J.; Xiao, K. Atmospheric emission of polychlorinated biphenyls from multiple industrial thermal processes. *Chemosphere* **2013**, *90* (9), 2453−2460.
(21) Fiedler, H. Short-chain chlorinated paraffins: Production, use and international regulations. In *Chlorinated paraffins*; de Boer, J., Ed.; Springer Verlag: Berlin, Germany, 2010; pp 1−40.
(22) Shoeib, M.; Harner, T. Characterization and comparison of three passive air samplers for persistent organic pollutants. *Environ. Sci. Technol.* **2002**, *36* (19), 4142−4151.
(23) Jaward, F. M.; Farrar, N. J.; Harner, T.; Sweetman, A. J.; Jones, K. C. Passive air sampling of PCBs, PBDEs, and organochlorine pesticides across Europe. *Environ. Sci. Technol.* **2004**, *38* (1), 34−41.
(24) Gioia, R.; Eckhardt, S.; Breivik, K.; Jaward, F. M.; Prieto, A.; Nizzetto, L.; Jones, K. C. Evidence for major emissions of PCBs in the West African region. *Environ. Sci. Technol.* **2011**, *45*, 1349−1355.
(25) Pozo, K.; Harner, T.; Shoeib, M.; Urrutia, R.; Barra, R.; Parra, O.; Focardi, S. Passive-sampler derived air concentrations of persistent organic pollutants on a North-South transect in Chile. *Environ. Sci. Technol.* **2004**, *38* (24), 6529−6537.
(26) Harner, T.; Pozo, K.; Gouin, T.; Macdonald, A. M.; Hung, H.; Cainey, J.; Peters, A. Global pilot study for persistent organic pollutants (POPs) using PUF disk passive air samplers. *Environ. Pollut.* **2006**, *144* (2), 445−452.
(27) Halse, A. K.; Schlabach, M.; Eckhardt, S.; Sweetman, A.; Jones, K. C.; Breivik, K. Spatial variability of POPs in European background air. *Atmos. Chem. Phys.* **2011**, *11*, 1549−1564.
(28) Bartkow, M. E.; Booij, K.; Kennedy, K. E.; Müller, J. F.; Hawker, D. W. Passive air sampling theory for semivolatile organic compounds. *Chemosphere* **2005**, *60* (2), 170−176.
(29) Randall, S.; Sivertsen, B.; Schneider, P.; Paul, A.; Rahman, A.; Hasan, M. Ambient air pollution screening study in Dhaka and Chittagong, 16 February−01 March 2013. Report 68/2014; Norwegian Institute for Air Research: Kjeller, Norway, 2014; http://www.nilu.no/Default.aspx?tabid=62&ctl=PublicationDetails&mid=764&publicationid=28062.
(30) Halse, A. K.; Schlabach, M.; Schuster, J. K.; Jones, K. C.; Steinnes, E.; Breivik, K. Endosulfan, pentachlorobenzene and short-chain chlorinated paraffins in background soils from Western Europe. *Environ. Pollut.* **2015**, *196*, 21−28.
(31) Zhang, X.; Wania, F. Modeling the uptake of semivolatile organic compounds by passive air samplers: Importance of mass transfer processes within the porous sampling media. *Environ. Sci. Technol.* **2012**, *46* (17), 9563−9570.
(32) Melymuk, L.; Bohlin, P.; Sáňka, O. e.; Pozo, K.; Klánová, J. Current challenges in air sampling of semivolatile organic contaminants: Sampling artifacts and their influence on data comparability. *Environ. Sci. Technol.* **2014**, *48* (24), 14077−14091.
(33) Melymuk, L.; Robson, M.; Helm, P. A.; Diamond, M. L. Evaluation of passive air sampler calibrations: Selection of sampling rates and implications for the measurement of persistent organic pollutants in air. *Atmos. Environ.* **2011**, *45* (10), 1867−1875.
(34) Bohlin, P.; Audy, O.; Škrdlíková, L.; Kukučka, P.; Přibylová, P.; Prokeš, R.; Vojta, Š.; Klánová, J. Outdoor passive air monitoring of semi volatile organic compounds (SVOCs): A critical evaluation of performance and limitations of polyurethane foam (PUF) disks. *Environ. Sci.: Processes Impacts* **2014**, *16* (3), 433−444.
(35) Harner, T. 2014_GAPS Template for calculating PUF and SIP disk sample air volumes_November 6 2014. Website: www.researchgate.net/profile/Tom_Harner?ev=hdr_xprf (retrieved March 10, 2015).
(36) Helsel, D.*Nondetects and Data Analysis: Statistics for censored environmental data*; Wiley: Weinheim, 2005.
(37) Nisbet, I. C.; LaGoy, P. K. Toxic equivalency factors (TEFs) for polycyclic aromatic hydrocarbons (PAHs). *Regul. Toxicol. Pharmacol.* **1992**, *16* (3), 290−300.
(38) Van den Berg, M.; Birnbaum, L. S.; Denison, M.; De Vito, M.; Farland, W.; Feeley, M.; Fiedler, H.; Hakansson, H.; Hanberg, A.; Haws, L. The 2005 World Health Organization reevaluation of human and mammalian toxic equivalency factors for dioxins and dioxin-like compounds. *Toxicol. Sci.* **2006**, *93* (2), 223−241.
(39) Katsoyiannis, A.; Sweetman, A. J.; Jones, K. C. PAH molecular diagnostic ratios applied to atmospheric sources: A critical evaluation using two decades of source inventory and air concentration data from the UK. *Environ. Sci. Technol.* **2011**, *45* (20), 8897−8906.
(40) Yunker, M. B.; Macdonald, R. W.; Vingarzan, R.; Mitchell, R. H.; Goyette, D.; Sylvestre, S. PAHs in the Fraser river basin: A critical appraisal of PAH ratios as indicators of PAH source and composition. *Org. Geochem.* **2002**, *33* (4), 489−515.
(41) Brändli, R. C.; Bucheli, T. D.; Ammann, S.; Desaules, A.; Keller, A.; Blum, F.; Stahel, W. A. Critical evaluation of PAH source apportionment tools using data from the Swiss soil monitoring network. *J. Environ. Monit.* **2008**, *10* (11), 1278−1286.
(42) Sampath, S.; Shanmugam, G.; Selvaraj, K. K.; Ramaswamy, B. R. Spatio-temporal distribution of polycyclic aromatic hydrocarbons (PAHs) in atmospheric air of Tamil Nadu, India, and human health risk assessment. *Environ. Forensics* **2015**, *16* (1), 76−87.
(43) Begum, B. A.; Nasiruddin, M.; Randal, S.; Sivertsen, B.; Hopke, P. K. Identification and apportionment of sources from air particulate matter at urban environments in Bangladesh. *Br. J. Appl. Sci. Technol.* **2014**, *4* (27), 3930−3955.
(44) Government of Bangladesh, Department of the Environment. Air pollution reduction strategy for Bangladesh, final report; Dhaka,

DOI: 10.1021/acs.est.5b03073
Environ. Sci. Technol. 2015, 49, 11372−11380

Bangladesh, 2012; http://www.doe.gov.bd/publication_images/60_air_pollution__reduction_strategy.pdf.
(45) Pozo, K.; Harner, T.; Wania, F.; Muir, D. C. G.; Jones, K. C.; Barrie, L. A. Toward a global network for persistent organic pollutants in air: Results from the GAPS study. *Environ. Sci. Technol.* **2006**, *40* (16), 4867−4873.
(46) Baek, S.-Y.; Jurng, J.; Chang, Y.-S. Spatial distribution of polychlorinated biphenyls, organochlorine pesticides, and dechlorane plus in Northeast Asia. *Atmos. Environ.* **2013**, *64*, 40−46.
(47) Zhang, L.; Dong, L.; Yang, W.; Zhou, L.; Shi, S.; Zhang, X.; Niu, S.; Li, L.; Wu, Z.; Huang, Y. Passive air sampling of organochlorine pesticides and polychlorinated biphenyls in the Yangtze river delta, China: Concentrations, distributions, and cancer risk assessment. *Environ. Pollut.* **2013**, *181*, 159−166.
(48) Liu, X.; Zhang, G.; Li, J.; Yu, L.-L.; Xu, Y.; Li, X.-D.; Kobara, Y.; Jones, K. C. Seasonal patterns and current sources of DDTs, chlordanes, hexachlorobenzene, and endosulfan in the atmosphere of 37 Chinese cities. *Environ. Sci. Technol.* **2009**, *43* (5), 1316−1321.
(49) Jaward, F. M.; Zhang, G.; Nam, J. J.; Sweetman, A. J.; Obbard, J. P.; Kobara, Y.; Jones, K. C. Passive air sampling of polychlorinated biphenyls, organochlorine compounds, and polybrominated diphenyl ethers across Asia. *Environ. Sci. Technol.* **2005**, *39* (22), 8638−8645.
(50) Zhang, G.; Chakraborty, P.; Li, J.; Sampathkumar, P.; Balasubramanian, T.; Kathiresan, K.; Takahashi, S.; Subramanian, A.; Tanabe, S.; Jones, K. C. Passive atmospheric sampling of organochlorine pesticides, polychlorinated biphenyls, and polybrominated diphenyl ethers in urban, rural, and wetland sites along the coastal length of India. *Environ. Sci. Technol.* **2008**, *42* (22), 8218−8223.
(51) Li, Y.-F.; McMillan, A.; Scholtz, M. T. Global HCH usage with 1° × 1° longitude/latitude resolution. *Environ. Sci. Technol.* **1996**, *30* (12), 3525−3533.
(52) World Health Organization. Air quality guidelines for Europe, 2nd ed.; WHO Regional Publications, European Series, No. 91, 2000; http://http://www.euro.who.int/__data/assets/pdf_file/0005/74732/E71922.pdf.
(53) Rahman, J.; Vajanapoom, N.; Van Der Putten, M.; Rahman, N. Impact of coastal pollution on childhood disabilities and adverse pregnancy outcomes: The case of Bangladesh. *Int. J. Med. Public Health* **2012**, *2* (3), 12−20.
(54) Wu, W.-T.; Lin, Y.-J.; Shiue, H.-S.; Li, C.-Y.; Tsai, P.-J.; Yang, C.-Y.; Liou, S.-H.; Wu, T.-N. Cancer incidence of Taiwanese shipbreaking workers who have been potentially exposed to asbestos. *Environ. Res.* **2014**, *132*, 370−378.
(55) Grant, K.; Goldizen, F. C.; Sly, P. D.; Brune, M.-N.; Neira, M.; van den Berg, M.; Norman, R. E. Health consequences of exposure to e-waste: A systematic review. *Lancet Global Health* **2013**, *1* (6), e350−e361.
(56) Xu, X.; Chiung, Y. M.; Lu, F.; Qiu, S.; Ji, M.; Huo, X. Associations of cadmium, bisphenol A and polychlorinated biphenyl co-exposure in utero with placental gene expression and neonatal outcomes. *Reprod. Toxicol.* **2015**, *52*, 62−70.
(57) Frazzoli, C.; Orisakwe, O. E.; Dragone, R.; Mantovani, A. Diagnostic health risk assessment of electronic waste on the general population in developing countries' scenarios. *Environ. Impact Asses.* **2010**, *30* (6), 388−399.
(58) Sujauddin, M.; Koide, R.; Komatsu, T.; Hossain, M. M.; Tokoro, C.; Murakami, S. Characterization of ship breaking industry in Bangladesh. *J. Mater. Cycles Waste Manage.* **2015**, *17* (1), 72−83.
(59) Wang, Y.; Xu, M.; Jin, J.; He, S.; Li, M.; Sun, Y. Concentrations and relationships between classes of persistent halogenated organic compounds in pooled human serum samples and air from Laizhou bay, China. *Sci. Total Environ.* **2014**, *482*−*483*, 276−282.
(60) Kim, J. Y.; Lee, J. Y.; Choi, S. D.; Kim, Y. P.; Ghim, Y. S. Gaseous and particulate polycyclic aromatic hydrocarbons at the Gosan background site in East Asia. *Atmos. Environ.* **2012**, *49*, 311−319.
(61) Wang, X. Y.; Li, Q. B.; Luo, Y. M.; Ding, Q.; Xi, L. M.; Ma, J. M.; Li, Y.; Liu, Y. P.; Cheng, C. L. Characteristics and sources of atmospheric polycyclic aromatic hydrocarbons (PAHs) in Shanghai, China. *Environ. Monit. Assess.* **2010**, *165* (1−4), 295−305.
(62) Fang, G.-C.; Chang, K.-F.; Lu, C.; Bai, H. Estimation of PAHs dry deposition and BaP toxic equivalency factors (TEFs) study at urban, industry park and rural sampling sites in central Taiwan, Taichung. *Chemosphere* **2004**, *55* (6), 787−796.
(63) Han, W.; Feng, J.; Gu, Z.; Wu, M.; Sheng, G.; Fu, J. Polychlorinated biphenyls in the atmosphere of Taizhou, a major e-waste dismantling area in China. *J. Environ. Sci.* **2010**, *22* (4), 589−597.
(64) Pozo, K.; Harner, T.; Lee, S. C.; Sinha, R. K.; Sengupta, B.; Loewen, M.; Geethalakshmi, V.; Kannan, K.; Volpi, V. Assessing seasonal and spatial trends of persistent organic pollutants (POPs) in Indian agricultural regions using PUF disk passive air samplers. *Environ. Pollut.* **2011**, *159* (2), 646−653.
(65) Chen, L.; Peng, X.; Huang, Y.; Xu, Z.; Mai, B.; Sheng, G.; Fu, J.; Wang, X. Polychlorinated biphenyls in the atmosphere of an urban city: Levels, distribution, and emissions. *Arch. Environ. Contam. Toxicol.* **2009**, *57* (3), 437−446.
(66) Chakraborty, P.; Zhang, G.; Eckhardt, S.; Li, J.; Breivik, K.; Lam, P. K. S.; Tanabe, S.; Jones, K. C. Atmospheric polychlorinated biphenyls in Indian cities: Levels, emission sources and toxicity equivalents. *Environ. Pollut.* **2013**, *182* (0), 283−290.
(67) Zhang, Z.; Liu, L.; Li, Y.-F.; Wang, D.; Jia, H.; Harner, T.; Sverko, E.; Wan, X.; Xu, D.; Ren, N.; Ma, J.; Pozo, K. Analysis of polychlorinated biphenyls in concurrently sampled Chinese air and surface soil. *Environ. Sci. Technol.* **2008**, *42* (17), 6514−6518.
(68) Li, Q.; Li, J.; Wang, Y.; Xu, Y.; Pan, X.; Zhang, G.; Luo, C.; Kobara, Y.; Nam, J.-J.; Jones, K. C. Atmospheric short-chain chlorinated paraffins in China, Japan, and South Korea. *Environ. Sci. Technol.* **2012**, *46* (21), 11948−11954.
(69) Chaemfa, C.; Xu, Y.; Li, J.; Chakraborty, P.; Hussain Syed, J.; Naseem Malik, R.; Wang, Y.; Tian, C.; Zhang, G.; Jones, K. C. Screening of atmospheric short- and medium-chain chlorinated paraffins in India and Pakistan using polyurethane foam based passive air sampler. *Environ. Sci. Technol.* **2014**, *48* (9), 4799−4808.
(70) Kong, D.; MacLeod, M.; Hung, H.; Cousins, I. T. Statistical analysis of long-term monitoring data for persistent organic pollutants in the atmosphere at 20 monitoring stations broadly indicates declining concentrations. *Environ. Sci. Technol.* **2014**, *48* (21), 12492−12499.
(71) Khan, M. A. A.; Khan, Y. S. A. Trace metals in littoral sediments from the North East coast of the Bay of Bengal along the ship breaking area, Chittagong, Bangladesh. *J. Biol. Sci.* **2003**, *3* (11), 1050−1057.
(72) Breivik, K.; Gioia, R.; Chakraborty, P.; Zhang, G.; Jones, K. C. Are reductions in industrial organic contaminants emissions in rich countries achieved partly by export of toxic wastes? *Environ. Sci. Technol.* **2011**, *45* (21), 9154−9160.
(73) Hogarh, J. N.; Seike, N.; Kobara, Y.; Habib, A.; Nam, J.-J.; Lee, J.-S.; Li, Q.; Liu, X.; Li, J.; Zhang, G.; Masunaga, S. Passive air monitoring of PCBs and PCNs across East Asia: A comprehensive congener evaluation for source characterization. *Chemosphere* **2012**, *86* (7), 718−726.

DOI: 10.1021/acs.est.5b03073
Environ. Sci. Technol. 2015, 49, 11372−11380