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PCBs in air, soil and milk in industrialized and urban areas of KwaZulu-Natal, South Africa

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Abstract

Information regarding polychlorinated biphenyls (PCBs) in environmental media in Africa is limited. This paper presents results of a monitoring program conducted in KwaZulu-Natal Province, South Africa designed to characterize levels, trends and sources of airborne PCBs. Particulate and vapor samples were sampled over the 2004–2005 period at three sites. The total PCB concentration averaged 128 ± 47 pg m^{-3}, and levels were highest in winter. Tri- through hexa-congeners predominated, and the vapor fraction was predominant. Several tetra- through hexa-chlorinated congeners had levels comparable to those at urban sites in the northern hemisphere, but hepta- through deca-congeners resembled levels at background sites. PCB source areas, deduced using spatial and temporal patterns, compositional information and trajectory analyses, likely included local, regional and global sources. Soils at three rural sites showed high PCB concentrations, and milk from a local dairy showed PCB concentrations comparable to USA levels in year 2000.

Keywords

Airborne contaminants; Africa; Organochlorine; PCBs; Transport; Vapor; Trajectory analysis; Soil; Milk

1. Introduction

Since production began in the 1930s, approximately 1.3 million tons of polychlorinated biphenyls (PCBs) have been manufactured (Breivik et al., 2007) and used in numerous applications, e.g., as coolants and insulating fluids for transformers and capacitors, stabilizing additives in PVC coatings, pesticide extenders, cutting oils, flame retardants, hydraulic fluids, sealants, adhesives, wood floor finishes, paints, and carbonless copy paper

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Airborne PCBs in KwaZulu-Natal, South Africa have similar concentrations but distinctive congener patterns compared to areas of historically high PCB use.

(Rudel et al., 2008). PCBs are semivolatile, inert, resistant to both alkali and acids, thermally stable, largely insoluble in water, lipophilic, resistant to degradation, highly persistent, bioaccumulative and toxic (ATSDR, 2000; WHO, 2003). Their widespread use and properties have led PCBs to become globally distributed. Their biphenyl structure, with two linked benzene rings and 1–10 chlorine atoms, supports 209 congeners although only 130 have been identified in commercial PCB products (ATSDR, 2000; WHO, 2003). In the atmosphere, PCBs are found in both vapor and particulate forms. Reaction with hydroxyl radicals is believed to be the dominant atmospheric transformation process, giving PCB half-lives from 10 days to 1.5 years, depending on the congener (WHO, 2003). Since PCBs bio-accumulate up the food chain, with fish, meats and dairy products containing the highest concentrations, food consumption has been and will continue to be the major exposure source in the general population (ATSDR, 2000). Mothers pass PCBs onto their unborn babies through blood, and to nursing infants through breastfeeding (WHO, 2003). PCB exposures have been linked to neuro-development effects in infants, cancer and immunotoxic effects (WHO, 2003). Despite restrictions or bans on PCB production and use in most countries, numerous environmental sources and reservoirs of these contaminants remain, including soils and sediments, for which atmospheric transport is a key transport and dispersal route (Huang et al., 2007). Global use and emission inventories of PCBs are very approximate, although a few estimates of emissions are available (Breivik et al., 2002, 2007).

In Africa, PCB measurements in any media are rare. PCBs were measured in water, sediment, and biota (e.g., vegetation, fish, shrimp, and humans) in the 1980s and 1990s in several countries (e.g., South Africa; UNEP, 2002); some more recent measurements in biota (Bouwman et al., 2008) and a few in air (Ngabe and Bidleman, 2002; Pozo et al., 2006) are also available. For the more chlorinated PCBs, recent emission inventories suggest that the single largest category contributor of airborne levels is disposal and uncontrolled (e.g., open) burning, while the less chlorinated and more volatile compounds are emitted primarily during the use phase (Breivik et al., 2002). Inappropriate disposal practices, including the dumping of waste from industrial nations (Loganathan and Kannan, 1994), represent another potentially important yet unquantified source in Africa. Finally, even as the use of PCBs in condensers and transformers was decreasing in the late 1970s, PCB use in the mining industries increased with the demand for fire-resistant hydraulic fluids (Fiedler, 2002). These circumstances suggest that measurements taken elsewhere have limited applicability in Africa. Given the prevalence of uncontrolled burning, the legacy of waste dumping, the significance of mining industries, and the potential health consequences of exposure, the lack of information on environmental levels of PCBs in Africa is significant and concerning.

Durban is the largest urban and industrial area in KwaZulu-Natal (KZN) Province in South Africa. Durban’s estimated (2007) population is 3.4 million, and KZN’s is 10.5 million. Located near the center of the metropolitan area, the Durban South Industrial Basin (DSIB) has one of the highest concentrations of industrial activity in Africa, containing two petroleum refineries, a paper mill, an international airport, large chemical tank farms, landfill sites, incinerators, processing and manufacturing industries, major trucking, harbor and rail facilities, and other industries. Residential, recreational and industrial areas are

intermingled. The metropolitan area also contains sizable concentrations of informal housing settlements, and cottage industries. KZN contains extensive agricultural lands, which extend into the metropolitan area.

Following a stakeholder process coordinated by the Durban health authority, a comprehensive air quality monitoring program was established in order to support health risk assessment and epidemiological studies. This program included very extensive measurements of both conventional and toxic pollutants. In this paper, we characterize levels and trends of airborne PCBs monitored at three locations in Durban, which include urban, industrial and residential sites. We also present results of preliminary investigations of PCB levels in KZN surface soils and locally produced milk. These data are compared to measurements elsewhere, and back-trajectory analyses and compositional information are used to identify potential source areas that contribute to airborne levels.

2. Materials and methods

2.1. Air sampling

Monitoring for PCBs in both gaseous and particulate phases was conducted at three sites (Fig. 1). The southernmost site, Nizam, was established at a primary school located in a small residential area amidst major industry, e.g., the Engen petroleum refinery (0.73 km to the NE), the Southern Works waste water treatment plant (0.81 km NW), the Mondi pulp paper plant (0.85 km W), and the Sapref refinery (0.88 km SW). This site was on a paved surface, close to a grassy bank, and ~10 m from the school building. The central Wentworth site, situated on a ~1.5 ha grassy knoll, was adjacent to a wooded area to the E, residential areas to the S and W, many smaller industries further (~0.5 km) to the W, a large hospital, and a cemetery to the N. Both the Nizam and Wentworth sites were south of Durban's central business district and port. The third site, Ferndale, was well to the north and about 20 km from the DSIB. This site was on a grassy patch at a primary school on a hilltop in moderately hilly terrain in a largely residential area that had a large fraction of open space. The closest industries were the new River Horse Valley and Phoenix (light) industrial parks (~4 km to the NE). Several low density informal settlements were located approximately 1 km to the N.

Identical equipment and protocols were used at each site. New high volume air samplers (TE-1000, Tisch Environmental, Cleves, Ohio, USA) were deployed to capture particulate matter on quartz microfiber filters (102 mm dia, QMA-4 filters, Whatman International Ltd, Florham Park, New Jersey, USA), and vapor phase pollutants on polyurethane foam plugs (75 mm dia, SKC, Ninety-four, Pennsylvania, USA). Sampling periods were 24 h, although a few longer samples were taken. The average sample volume was 300 m^3. All sensitive materials were sealed and checked for possible contaminations following shipping, and all blanks met QA/QC criteria. In the field, filters and PUF plugs were spiked with PCB-65 and 166 surrogates immediately after sampling to determine any matrix effects and recoveries. Collected samples were immediately shipped by air to our Michigan laboratory, and refrigerated (4 °C) until analysis.

We report on 15–20 sampling events at each of the three sites conducted over the period from August 2004 to September 2005.

2.2. Soil sampling

Soil samples were obtained from three additional sites, all inland, between the small towns of Ixopo and Richmond, and approximately 85 km W of Durban. The only known and likely source of PCBs is a commercial medical waste incinerator in Ixopo. Soils were sampled in spring (22 November 2005); the previous 3-month period had had moderate temperatures (average 16.8 °C) and little precipitation (total of 10.8 cm). Three sites were sampled: two residential areas about 400 km apart, and an agricultural area about 3 km distant. Both surface (0–0.5 cm depth) and shallow subsurface (1–2 cm depth) soils were collected using composite samples (five subsites, the corners and center of a 5 × 5 m square). 2 g samples were collected from a 10 × 10 cm area and placed into a polyethylene bag with a zip lock closure. The collection spoon was rinsed with hexane and acetone prior to each use. Soil samples and two unused bags (as blanks) were shipped to Michigan where they were frozen (−30 °C) until analysis. Soils were quite dry (moisture content <20%), and the texture was a sandy loam.

2.3. Sample analysis

Analysis of the air samples followed US EPA (1999). Filters and PUF plugs were Soxhlet extracted for 36 h each using benzene/hexane (4:1). Extracts were reduced to 10 ml in a Kuderna-Danish evaporator, cleaned with 4.5 M sulfuric acid, and further reduced to 1.0 ml. Internal standards (PCBs-136 and 204) were added to each sample. Separate sample preparation and GC/MS runs were performed for each particulate and vapor phase sample. Including quality control samples (repeat analyses, surrogate recoveries, blanks, recovery checks, and checks on individual standards), we ran a total of 110 samples.

Analysis of soil samples followed Hengstmann et al. (1989) and Hagenmaier et al. (1992). Two 5 g subsets of each sample were spiked with a surrogate solution, soaked in acetone (25 ml) for 1 h, and Soxhlet extracted for 36 h again using benzene/hexane (4:1). Extracts were reduced to 10 ml, and cleaned with concentrated fuming sulfuric acid. Since most samples were highly colored, each sample was cleaned 2–5 times (depending on the intensity of color). Cleaned elutes were evaporated to 1.0 ml and then treated as described for the air samples. Including the samples and quality control analyses, we ran a total of 40 samples. Concentrations were expressed as ng g^{-1} wet weight.

Quantitation of 82 PCB congeners or congener groups was accomplished using a GC/MS (Model 6890/5973, Agilent Industries, Palo Alto, CA, USA), fused silica DB-5 column (30 m length, 0.25 mm id, film thickness 0.25 μm, J&W Scientific, Folsom CA, USA), selected ion monitoring (SIM) mode, negative chemical ionization mode, and methane as the reagent gas (Chernyak et al., 2005). Method detection limits (MDLs) determined using low concentration spiked samples (Keith, 1991) were estimated to range from 0.03 to 6 pg m^{-3}, depending on the congener, sample volume, degree of chlorination, and other factors.

2.4. Milk sampling and analysis

Milk samples for PCB analysis were obtained from Fairfield Dairy, a supplier to a "high-end" food store, located approximately 75 km NW of Durban in a largely rural and agricultural area near the town of Howick (population of 32,000, 1015 m elevation) and 20 km NW of Pietermaritzburg (population of 846,000). Two processed milk samples (150 ml each), along with a blank, were collected in April 2007 and analyzed for 31 PCB congeners by GC/MS by the Norwegian Institute for Air Research (NILU) laboratory in Kjeller, Norway. For these samples, MDLs ranged from 10 to 80 pg l^{-1}, depending on the congener, while recoveries ranged from 44% to 78%.

2.5. Quality assurance

Quality assurance activities for air samples included: routine use of blanks; spike and surrogate recovery determinations; duplicates and co-location studies; and inter-laboratory comparisons of split samples and standards. Blanks were run with each sample batch (separately for filters, PUFs, and soil samples). All solvents and other materials contacting samples were clean, as confirmed using blanks. Spike recovery tests were performed periodically during analyses. All of these showed acceptable recovery (80–101%). Surrogate recovery tests, performed on each sample using unlabeled standards (PCB-65 and 166), gave acceptable results (75–110%). Co-location tests using two air samplers at the Wentworth site showed good agreement, e.g., precisions averaging 7% and 21% in vapor and particulate phases, respectively. Individual congeners showed wider variation, but never above 50% (for trichlorohomologues). Several of the blanks showed a few PCB congeners (i.e., PCB-74, 84,149, 118) at low concentrations, close to MDLs. Field and laboratory blanks for the soil and milk samples showed acceptable recoveries on surrogate and spike tests.

2.6. Data analysis

For each contaminant, we calculated descriptive statistics by site and phase (vapor or particulate). Seasonal trends for the sum of the 82 measured PCB congeners, denoted Σ_{82}PCB, were depicted using an exponential smoother (smoothing parameter $\alpha = 0.01$ day^{-1}) for the 3-site average of total (vapor plus particulate) concentration, followed by polynomial curve-fitting.

To compare levels in Durban to measurements taken elsewhere, we selected 11 recent studies (since 2000) that took comprehensive airborne measurements (at least a season's average) in Europe, Asia, South America, North America, and Antarctica. No comparable African studies were located. (Data collected at De Aar, South Africa, Ghana, and Botswana in the Global Atmospheric Passive Sampling project (GAPS; Pozo et al., 2006) are noteworthy, but the passive sampling technique may not be comparable, and only one season was sampled, thus these data were not compared.) When necessary, we calculated annual means, summed vapor and particulate fractions if reported separately, and limited the comparison to congeners reported in at least half of the studies and this study. For soils, we selected seven recent studies measuring PCB levels in Europe, Asia, South America and elsewhere, including a few samples from South Africa. These studies are further described in the Supplemental material: supplemental Tables S1 and S2 give a general overview of air

and soil data, respectively. Supplemental Tables S3 and S4 describe key results for airborne and soil measurements, respectively.

Back-trajectory analyses used the Hysplit model and the FNL meteorological dataset to simulate 3-day back-trajectories every 4 h during each of the 24-h sampling periods, i.e., six back-trajectories were computed for each 24-h sampling event (Draxler and Rolph, 2003). Based on the trajectories, the number of hours that each trajectory was over five large continental areas (N KZN, S KZN, N interior Africa, S interior Africa) and three ocean areas (N and S Indian Ocean, S Atlantic) was determined and then used as weights to obtain area-weighted concentrations from these eight possible source area. Differences in the area-weighted concentrations can help to identify emissions that are local or regional in origin. The Nizam site, which had the most measurements, was used for this analysis, thus avoiding any site-to-site differences. (Other sites gave comparable results.)

3. Results and discussion

3.1. Levels in air

Levels of total (particulate and vapor phase) PCB congeners at the three sites are shown in Table 1. All congeners were predominantly in the vapor phase with the exception of PCB-49, which was found at very low levels and which can be difficult to quantitate. Considering the 82 PCB congeners quantified, an average of 94% of the concentration was in the vapor phase; similar results have been shown by Mandalakis and Stephanou (2002), Cindoruk and Tasdemir (2007), Sundqvist et al. (2004), and others. The total PCB (Σ_{82}PCB for vapor + particulate) concentration averaged 128 ± 47 pg m^{-3}, and the predominant compounds were PCB-33 (tri, 12 ±10 pg m^{-3}), PCB-118 (penta, 12 ± 8 pg m^{3}), PCB-138 + 163 (hexa, 8 ± 3 pg m^{-3}), and PCB-105 (penta 7 ±3 pg m^{-3}). Hepta- through deca-congeners also were detected, though concentrations were much lower, generally below 0.2 pg m^{-3}.

Fig. 2 depicts the spatial gradient in Durban, showing concentrations of homologues at the three sites. While levels at central Wentworth generally exceeded those at other sites for most of the homologues, the overlapping confidence intervals show only modest local gradients. The largest difference was a 2-fold reduction in the tri-congeners at the northern Ferndale site as compared to the central Wentworth site. Possibly some differences might have arisen due to missing data, but the gradient in Durban was small compared to Manchester, England where a 9-fold range in airborne PCB levels was found across a 79 km transect (Jamshidi et al., 2007), and Rome, Italy where a 5-fold range was found between central Rome site and a rural background site 60 km distant (Menichini et al., 2007). The lack of a strong gradient in Durban suggests that the three sites were affected by similar and relatively well dispersed local sources, and/or regional or global background sources. This inference, however, would be best confirmed by monitoring at a remote, truly background African site. While Pozo et al. (2006) reports (one season) total PCB levels of 35 pg m^{-3} in Ghana, <MDL in Botswana, and 252 pg m^{-3} in De Aar, S. Africa, these data are too sparse for the purpose of evaluating gradients.

The time trend of Σ_{82}PCB concentrations, shown in Fig. 3, shows increases of about 40% in winter (June through September) when levels reached 170 pg m^{-3} (3-site average), compared to 120 pg m^{-3} for the rest of the year. This trend was seen at each site, and also for the major congeners (data not shown). Σ_{82}PCB levels at Wentworth were higher, by about 50–100 pg m^{-3}, than levels at the other sites, a difference that was maintained over the year. Ideally, several years of data would be used confirm these trends. The similar trends at the three sites reinforce the argument that common sources of PCBs affect the three sites. Meteorological influences, specifically the greater frequency of inversions in winter and changes in the boundary layer depth, may also be responsible, as discussed next.

3.2. Meteorology and back-trajectories

The Durban area experiences the semi-permanent South Atlantic and South Indian high pressure cells that dominate circulation and cause lows with unstable conditions in the summer and persistent highs in the winter. The winter is characterized by light winds and inversions that are unfavorable to pollutant dispersion; however, the temperate control system produces frequent traveling low pressure systems that are associated with fronts, moderate winds and precipitation which tends to cleanse the air column. Local influences include: katabatic, valley and slope wind systems that develop about 150 km inland at the Drakensberg Mountains, the escarpment of the African plateau; nearby coastal ridges and valley systems that channel air flows; and the land/sea interface that causes diurnal shore and sea breezes. Concentrations of conventional pollutants in the DSIB, e.g., PM_{10}, SO_2 and NO_2, typically fall dramatically with a sea breeze or the passage of a front followed by precipitation.

Results of the trajectory analysis show that areas to the north and west of Durban are associated with higher concentrations of Σ_{82}PCB (and most congeners), while trajectories from the east (over the Indian Ocean) are associated with lower concentrations (Fig. 4). The highest concentrations were associated with trajectories that originated in the Atlantic Ocean, swept in the southeast direction across the Africa continent, and arrived in Durban. This might reflect emissions from PCBs used in mining in the interior portions of Africa, or possibly even longer range transport from the northern hemisphere. Such events occurred only in the winter, thus local KZN sources – amplified with inversion conditions (and possibly the widespread agricultural burns that occur in the fall) might be responsible. However, PM_{10} concentrations measured at five Durban locations for the same period (using TEOM instruments operated by the Durban Municipality and using the same analysis) do not show this pattern (Fig. 4). Instead, most of the PM_{10} arrives from local or regional sources in northern KZN province. Also, we would expect sharper spatial gradients from local sources (not seen in Fig. 2) under most conditions. Again, trajectories from the Indian Ocean were associated with lower PM_{10} levels. Similar analyses for persistent chlorinated pesticides, e.g., DDT and lindane, showed yet different patterns, e.g., potential, current and/or historical usage areas in Africa were associated with higher concentrations (Batterman et al., 2008).

There are limitations to the trajectory analyses. The various trajectories terminating in Durban at the monitoring site over each 24-h sampling period varied, and specific source

regions along each trajectory cannot be differentiated. Trajectories are complex and uncertain, in part due to the relatively coarse spatial resolution (191 km) of the meteorological data that drives the analyses, and each trajectory's limited duration. A larger number of samples over a longer period would help confirm results. If source inventories were known, modeling would complement and help to confirm the trajectory analyses. A recently completed 3-D global model of PCB transport and budgets (Huang et al., 2007) predicted that while the northern-to-southern hemispheric transport of PCBs is limited, the southern hemisphere gains PCBs (e.g., a net of 69 kg of PCB-28 and 8 kg of PCB-180), and that the southern hemisphere is a net sink for global PCBs. Unfortunately, emission inventories, budgets and modeling of PCBs on a local or regional scale, e.g., for KZN Province, are unavailable.

The trajectory analysis, as well as the modest spatial variation, high temporal correlation, and vapor phase dominance discussed earlier, all suggest the importance of distant or regional sources of PCBs. Compositional information, described next, provides another indication of those sources that contribute to the airborne concentrations.

3.3. PCB compositions and comparisons to airborne levels elsewhere

Because we could not locate previous measurements of airborne PCBs elsewhere in Africa, we compared the Durban measurements to results of 11 other recent studies. (The study designs are summarized in Table S1, and study results in Table S3.) Although the following distinctions are not fully exclusive, we grouped the sites in these studies into industrial, urban and remote categories, shown below, and then averaged results on a congener basis:

- *Industrial*. In this group, we included airborne measurements at a site in Izmir, Turkey located near steel, fuel oil combustion, vinyl chloride and petrochemical industries, averaged over summer and winter periods (Cetin et al., 2007); and results from a study in Santiago, Chile near waste disposal, incineration and other industrial sources (Mandalakis and Stephanou, 2002). In both studies, soil was suggested to be a main source of the airborne PCBs, although neither reported soil measurements.
- *Urban*. Here we include results from six studies in diverse areas: Rome, Italy (Menichini et al., 2007); Yokohama City, Japan (Kim and Masunaga, 2005); Zagreb, Croatia (Romanic and Krauthacker, 2007); Bursa, Turkey (Cindoruk and Tasdemir, 2007); the most urban site sampled in Manchester, England (Jamshidi et al., 2007); and urban sites near the Turkish industrial site mentioned above (Cetin et al., 2007).
- *Remote or background*. Seven studies provided background levels: a rural area in Italy (Menichini et al., 2007); airborne measurements over the Kattegat Sea near Sweden (Sundqvist et al., 2004); King George Island off Antarctica (Montone et al., 2003); the most rural location in the Manchester, England study (Jamshidi et al., 2007); measurements over the Atlantic Ocean (Jaward et al., 2004); the Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP) study using six background sites

in northern Europe and a three year period (Aas and Breivik, 2005); and nine remote sites in the US (Cleverly et al., 2004).

As shown in Fig. 5, airborne concentrations of PCBs in Durban mostly fell between levels found at urban and remote sites. Several of the tetra-, penta- and some hexa-congeners (e.g., PCBs-99, 105, 118, 138, 156) had levels in Durban that were very similar to the "urban" levels. The more chlorinated congeners usually remain closer to sources (Meijer et al., 2003), and suggest local (Durban area) emissions that affect the region. On the other hand, the tri-and other tetra-congeners found at the highest concentrations in both industrial and urban studies (e.g., PCBs-31, 33, 52, 49) had much lower concentrations (by 10–100 times) in Durban, and they approached levels observed at remote sites. These more volatile congeners have the capability for long-range atmospheric transport (Meijer et al., 2003). The lack of spatial gradient and similarity to background levels for the tri- and some tetra-congeners suggests relatively few open sources emitting these PCBs in Durban and the dominance of long-range transport.

The variability in the data, and the divergence from the literature congener pattern shown in Fig. 5, can result from a variety of sources. Not all studies measured the same congeners. Since not all sites were sampled each day, temporal variation may affect results. Some uncertainty is inherent, especially for low concentrations, highly contaminated samples, complex sample matrices, and congeners with similar elution times. In the present study, some of these issues apply to PCBs-49, 52, 91 and 200. These problems, however, did not apply to PCB-180, which has a unique retention time and no interferences. The depletion of PCB-180 from the Durban air samples, relative to urban levels elsewhere, appears to be a unique feature for South Africa.

3.4. Levels in soil

PCB concentrations in soils are listed in Table 2. We detected 38 congeners in all six samples, the most prevalent being PCBs-41 +71, 153 +132, 138 + 163, In the surface soils (0–0.5 cm depth), the average (3-site) concentrations for most of the congeners ranged from 1 to 10 ng g^{-1}, and site-to-site differences were large (factor of 10 from highest to lowest). Levels in the shallow soil (1–2 cm depth) were lower than those in the surface soils by 2.8 ± 1.4 times. Site-to-site differences were small, of the order of a factor of 3. Concentrations in surface and shallow soils were highly correlated (Pearson $r = 0.93$). The higher concentrations in the surface soils were expected given the variability of organic carbon and the possibility of recent and spatially varying atmospheric deposition. PCBs in surface and surface soils may be more accessible for uptake onto foods, dusts and crops that lead to human exposure. Levels in the agricultural soil seemed anomalously high, and thus this site was excluded from further analyses.

We compared the KZN soil measurements to results in seven recent studies (Fig. 6, Tables S2 and S4). These studies differed in the siting criteria, soil depths and congeners analyzed. Most reported relatively few congeners. These studies showed a wide range of concentrations that defied simple categorization, e.g., levels at background sites in some studies often exceeded levels at urban/industrial levels in other studies. Such differences can arise for many reasons, but the most important seems to be the study region's current and

historical use of PCBs, followed by siting and sampling differences. We grouped the literature results into the three categories based on concentration:

- *High concentration*. In the Moscow region, Russia, Wilcke et al. (2006) examined an urban-to-rural transect. Many congeners exceeded 1000 pg g^{-1}. We placed the two central Moscow sites, as well as four other nearby sites (within 40 km to the south of Moscow), in this category.
- *Medium concentration*. Several studies reported congener-specific concentrations from about 100 to 1000 pg g^{-1}. These include measurements at a background site approximately 100 km northwest of Moscow (Wilcke et al., 2006); a central site in Manchester, UK (Jamshidi et al., 2007); 101 rural sites across Great Britain (Heywood et al., 2006); 23 sites throughout Switzerland (Schmidt et al., 2005); and a global background estimate using 191 remote sites located mostly in the northern hemisphere (Meijer et al., 2003).
- *Low concentration*. A diverse set of studies report congener levels from about 10 to 100 pg g^{-1}. These include three sites in Brazil fairly close to industrial sources (Rissato et al., 2006); two open areas in national parks in NE South Africa reported in the global background study (Meijer et al., 2003); 52 urban, rural and remote sites throughout China (Ren et al., 2007); and nine sites outside of central Manchester (Jamshidi et al., 2007).

PCB levels in the shallow KZN soils generally resembled those in the medium concentration group (Fig. 6), and they far exceeded measurements at the two remote sites measured elsewhere in South Africa (Meijer et al., 2003). These results pertain to tri-through hepta-congeners (few of the literature studies reported more chlorinated congeners, thus no other comparisons are made). The similarity of levels in KZN to the medium concentration studies is surprising; the literature studies emphasized sites in the industrialized and northern hemisphere countries where PCB use has been high, while the rural KZN area was believed to be far from most PCB sources. In part, the relatively elevated concentrations resulted from our use of shallow soils (1–2 cm depth), while the literature studies examined 0–5, 0–10, or 0–20 cm depths. However, our comparison excluded surface soils (0–1 cm depth), a layer included in the other studies, which would likely and significantly increase the average concentration. Overall, the high concentrations found in the KZN soils suggest local sources.

While PCB concentrations in soils have not been as closely monitored as sediments (ATSDR, 2000), many factors are known to affect levels. Transect studies (e.g., Wilcke et al., 2006; Jamshidi et al., 2007) have shown the importance of proximity to urban and point sources. At the global scale, high levels at background sites are found between 30° and 60° N latitude, the region of historically high PCB use (Meijer et al., 2003). Soil organic matter plays a key role in PCB adsorption, and air-to-surface exchange and partitioning of PCBs is affected by climate, fraction of soil organic matter, and vegetation (Meijer et al., 2003). Several studies have shown that levels in aggregate surface soils exceed those of subsurface soils, e.g., differences of about 2-fold were seen in the Moscow region (comparing 0–1 cm versus 0–20 cm depths; Wilcke et al., 2006), and 5-fold differences in Bavarian forest soils (0–5 versus 15–20 cm depths; Krauss et al., 2000). Attenuation with depth is expected given

PCB's low solubility and the scavenging of the soil organic matter in topsoil and organic layers (Krauss et al., 2000), an effect that may be more pronounced for PCBs than for PAHs and heavy metals due to PCB's relative volatility and preferential sorption on aggregate surfaces (Wilcke et al., 2006). Since being banned in the late 1970s, PCB levels in soils (and sediments) have decreased across the USA, and the half-life in riverine sediments is estimated to be 9.5 ± 2.2 yrs (ATSDR, 2000).

Conclusions drawn from this pilot investigation must be limited since only three sites in a localized area in the large and diverse KZN Province were examined. A transect study spanning rural, urban and industrialized areas in KZN, and sampling surface as well as subsurface (0–5 cm depth) soils would be revealing.

3.5. Levels in milk

The milk samples showed 28 of the 36 target congeners at quantifiable levels, including dioxin-like PCB-77 (80 pg l^{-1}) and PCB-126 (30 pg l^{-1}), and a total PCB concentration of 22 ng l^{-1}. Levels of the predominant congeners in milk are shown in Fig. 7 and compared to average levels in Durban soils and air. The evaluation of bioaccumulation up the food chain and into milk is congener-specific and depends on many factors, e.g., plant uptake factors, cow diet and metabolism rates. Many of the congeners found at the highest levels in milk in Durban were found in surface soils in Durban; however, abundances of the airborne PCBs differed considerably.

For comparison of the milk results, during the period of high PCB use in the USA (1969–1976), 4.1% of milk samples showed detectable levels of total PCBs, and a (high) average concentration of 67 μg l^{-1} (ATSDR, 2000). (We note that measurements have considerably improved since these early studies, and comparisons of both detection frequency and concentrations from early studies require several caveats.) Over the 1990s, PCB concentrations in milk in the USA declined by about 50%, and a year 2000 national survey in the USA (Schaum et al., 2003) showed congener-specific concentrations that were roughly similar to the Durban measurements, e.g., PCBs-105, 118 and 156 were 77–80% lower in Durban, while PCB-77 was 12% higher. Because only one Durban area dairy was tested, we cannot draw broad conclusions regarding the presence of PCBs in milk in KZN. Additional work is needed to determine whether the South African diet provides significant exposure to PCBs. We suggest sampling of bottom feeding fish and especially human milk, both of which can contain significant levels of PCBs (ATSDR, 2000).

4. Conclusions and recommendations

Airborne sampling in Durban showed predominantly vapor phase PCB concentrations that were similar in magnitude to levels in urban and industrial areas where PCB use has been high historically. In comparison to some of the literature studies, however, the three sites showed only modest spatial gradients. The PCB composition suggests few open sources of PCBs, and the trajectory analyses and seasonal patterns suggest the importance of yet undetermined regional sources, as well as contributions through long-range atmospheric transport from global sources. Measurements at a remote African site would help to confirm contributing from distant sources. A preliminary study of surface and shallow soils outside

of the metropolitan area showed surprisingly high PCB concentrations, however, only a single region was examined and deeper soils were not tested. A province-wide transect study examining soils (and air) would help to identify local sources, sinks and reservoirs, and would increase our understanding of air-surface exchanges in arid regions with poor soil and low vegetation. Milk samples from one dairy tested showed PCB levels that were roughly comparable to year 2000 levels in the USA, however, this investigation was too limited to evaluate the significance of potential PCB exposure pathways. While diet (especially meat, dairy products and fish) is the main source of human exposure to PCBs for most individuals, the elevated concentrations found in air and soil suggest the need for further identification and characterization. Follow-up studies sufficient to characterize PCB levels in fish and human milk would help to answer questions regarding human exposure.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

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Appendix. Supplementary material

Supplementary data associated with this article can be found n the online version, at doi: 10.1016/j.envpol.2008.08.015.

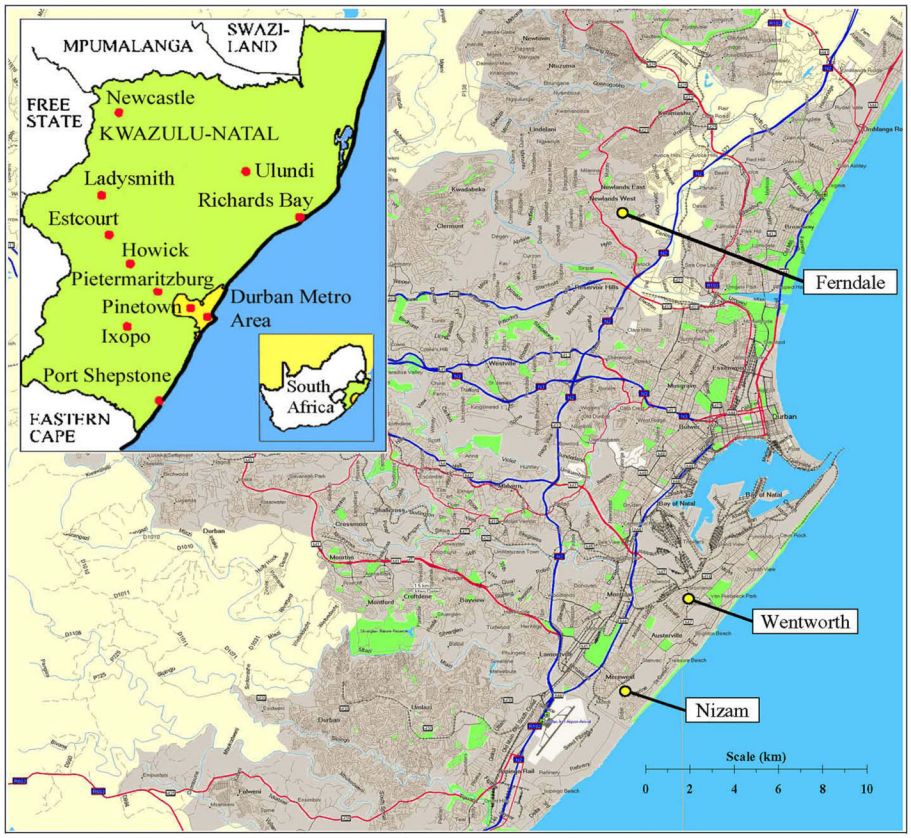

Fig. 1.

Locations of monitoring sites in greater Durban metropolitan area. The two southern monitoring sites lie in the DSIB. Inset maps show province of KwaZulu-Natal in South Africa, surrounding provinces, and locations of soil (Ixopo) and milk (Howick) sampling.

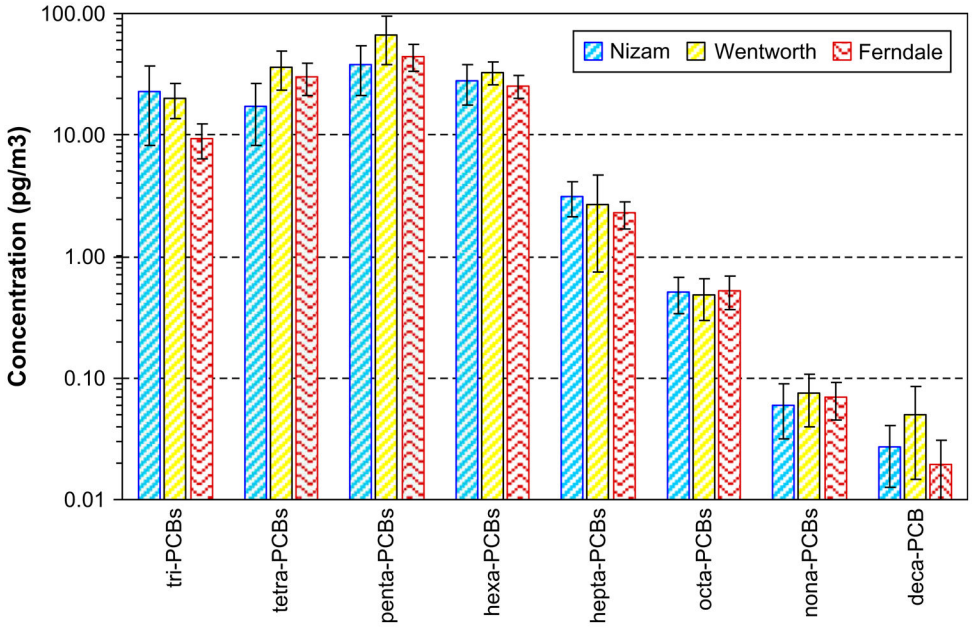

Fig. 2.

Average ambient total (particulate and vapor) PCB concentrations in homologue groups measured at three sites. Error bars show standard deviation. Sample sizes are 23, 22 and 13 at Nizam, Wentworth and Ferndale, respectively.

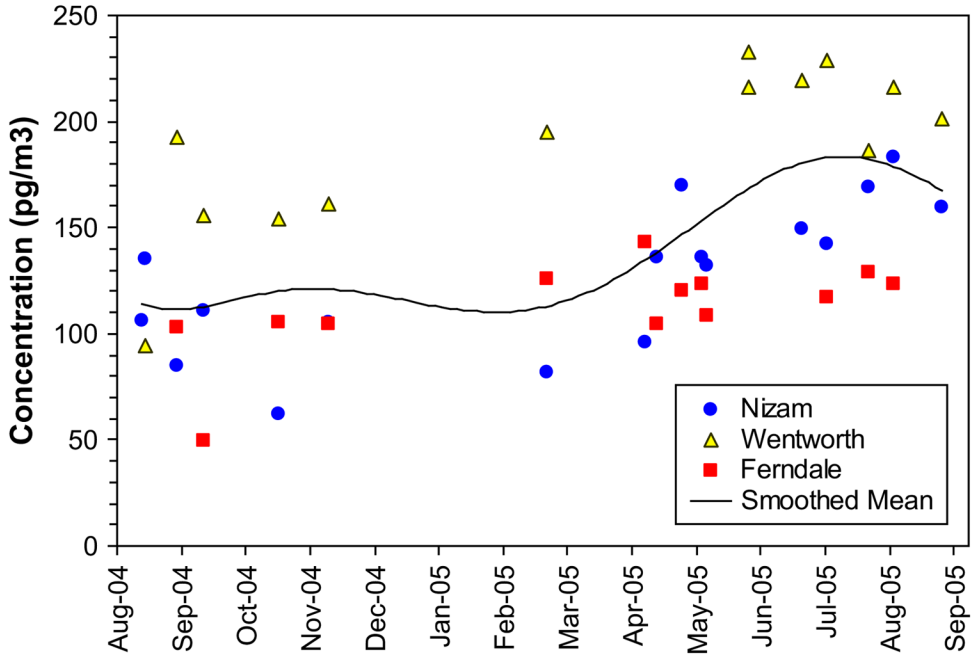

Fig. 3.

Total (particulate and vapor) Σ_{82}PCB concentrations at the three sites, and smoothed 3-site mean over the study period.

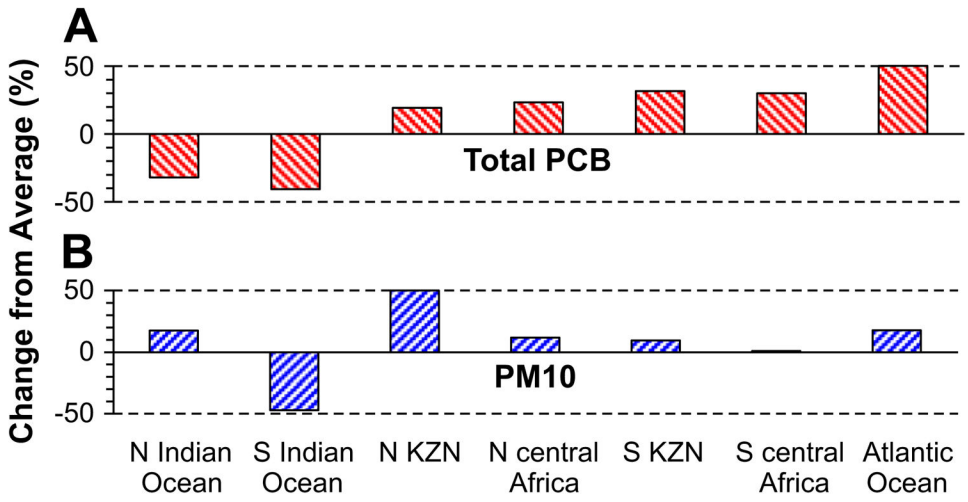

Fig. 4.

Relative change (percent from average) in concentrations for seven source areas for total Σ_{82}PCB and particulate matter (PM_{10}), for comparison, based on back-trajectory analyses.

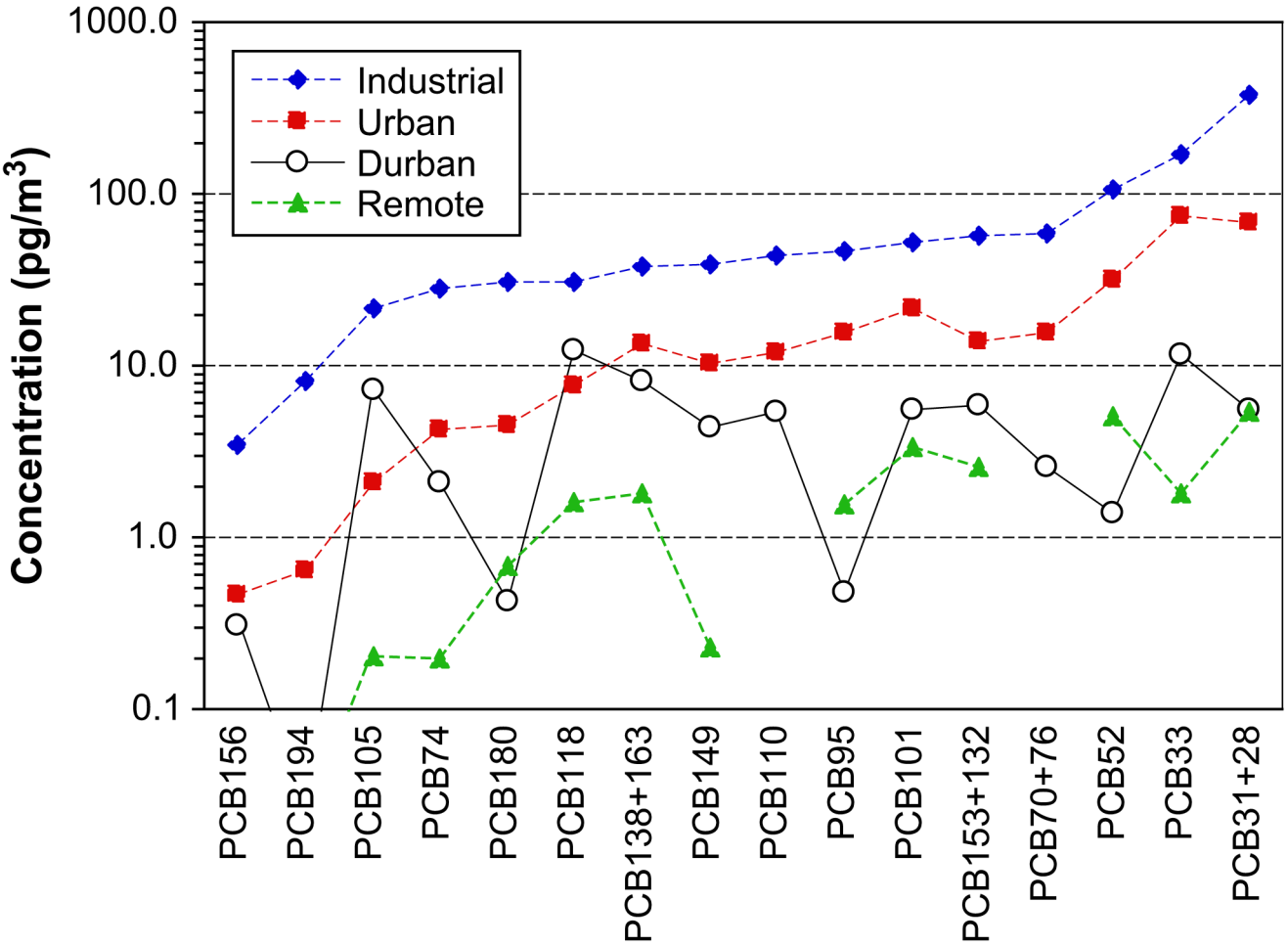

Fig. 5.

Comparison of average airborne PCB concentrations for selected congeners in Durban to literature averages for industrial, urban and remote sites. Congener sequence is ranked by industrial average.

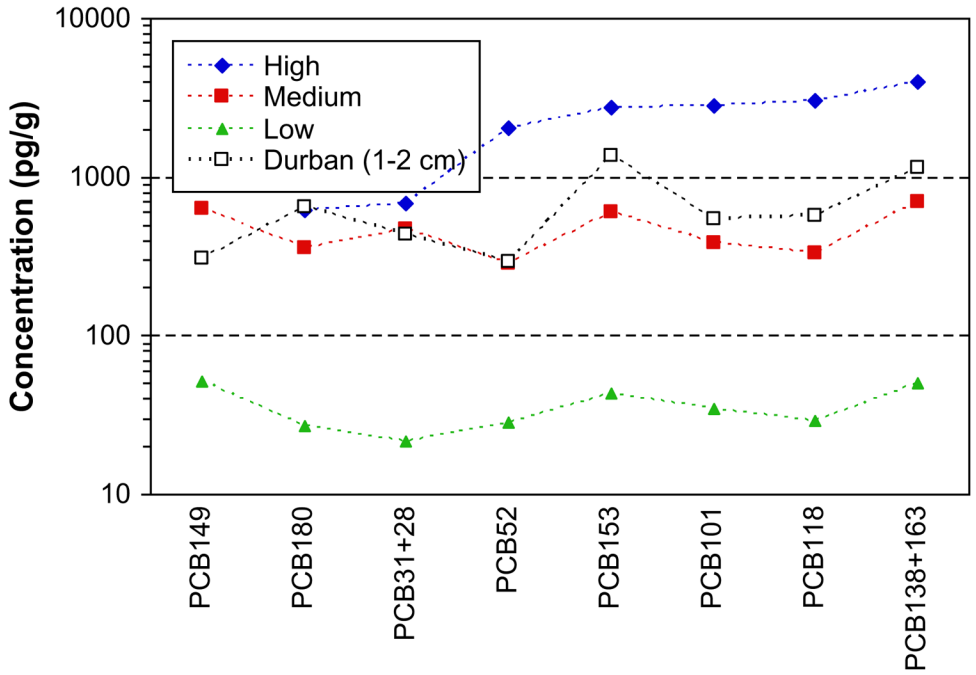

Fig. 6.

Comparison of PCB levels in Durban soils (surface and shallow; selected congeners) to literature averages for high, medium and low concentration sites. Congener sequence is ranked by urban/rural average.

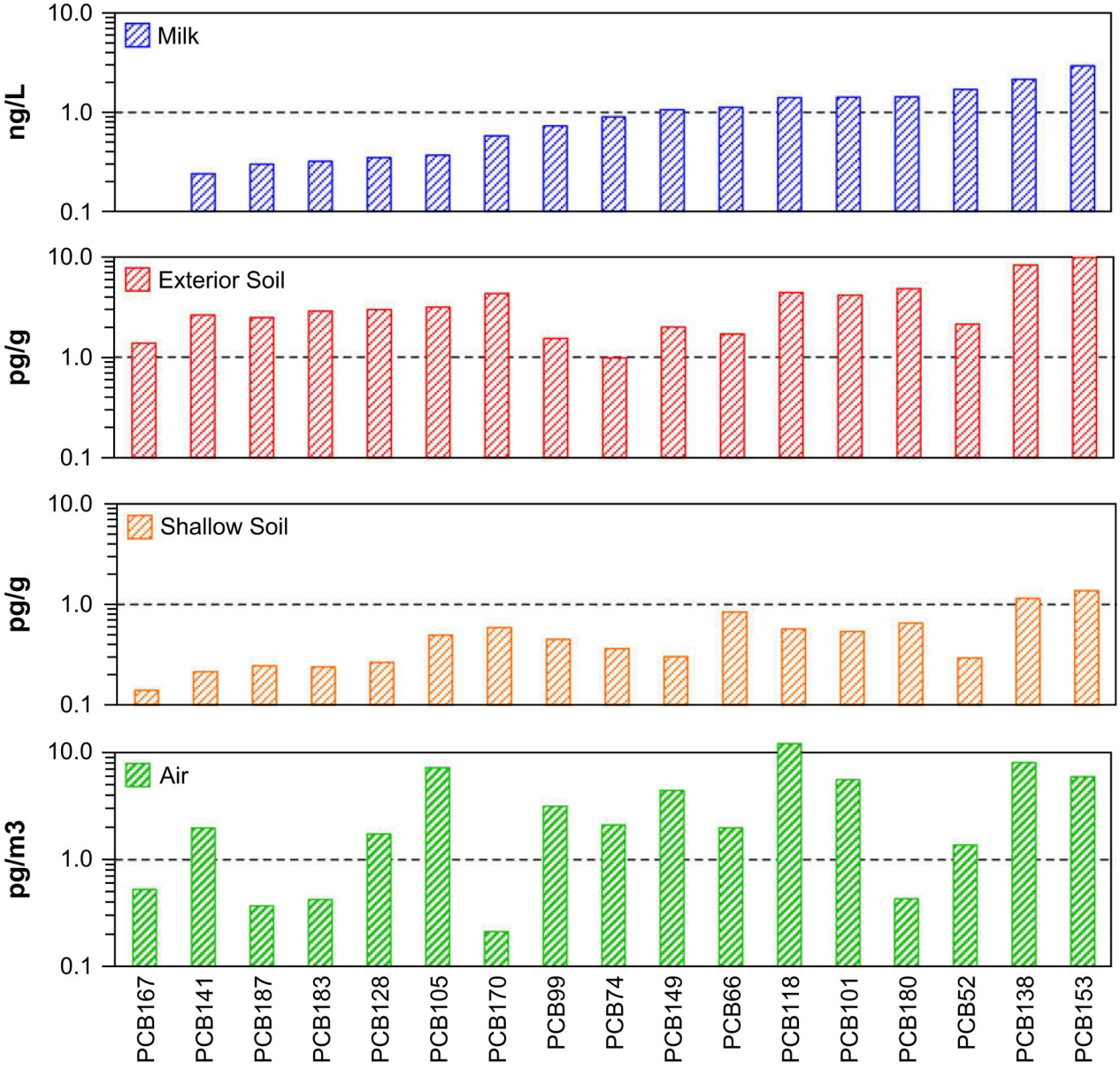

Fig. 7.

Comparison of PCB levels in milk, soils and air in Durban. Selected congeners, ranked by levels in milk level.

Table 1

Statistics of PCB concentrations across three sites

IUPAC no.	Positions	Homologue	Concentration (pg m^{-3})			Vapor fraction	*n*
			Average	St. dev.	Max		
31 + 28	2,4′,5 + 2,4,4′	Tri	5.46	2.78	19.23	100.0	58
33	2,3′,4′	Tri	11.71	10.05	41.06	99.5	56
22	2,3,4′	Tri	1.40	1.06	4.29	100.0	45
52	2,2′,5,5′	Tetra	1.37	1.82	6.11	97.1	27
49	2.2′4,5′	Tetra	0.10	0.52	2.92	2.7	8
47 + 48	2.2′4,4′ + 2,2′,4,5	Tetra	3.70	6.96	22.33	77.9	34
44	2,2′,3,5′	Tetra	0.72	1.59	6.31	83.0	14
42	2,2′,3,4′	Tetra	1.05	1.61	7.14	87.7	26
41 + 71	2,2′,3,4 + 2,3′,4′,6	Tetra	6.29	6.56	31.67	84.2	58
64	2,3,4′,6	Tetra	5.70	5.54	27.38	100.0	56
40	2,2′,3,3′	Tetra	0.07	0.16	0.76	98.5	14
63	2,3,4′,5	Tetra	0.22	0.23	0.93	99.6	42
74	2,4,4′,5	Tetra	2.11	2.08	9.57	100.9	58
70 + 76	2,3′,4′,5 + 2,3′,4′,5′	Tetra	2.54	1.73	8.23	98.8	58
66	2,3′,4,4′	Tetra	1.99	1.59	7.30	99.9	58
95	2,2′,3,5′,6	Penta	0.48	1.01	4.48	99.5	54
91	2,2′,3,4′,6	Penta	0.12	0.17	0.72	99.4	53
56 + 60	2,3,3′,4′ + 2,3,4,4′	Tetra	1.18	0.75	4.10	99.9	58
84 + 92 + 89	2,2′,3,3′,6 + 2,2′,3,5,5′ + 2,2′,3,4,6′	Penta	5.78	4.73	21.43	98.7	58
101	2,2′,4,5,5′	Penta	5.60	4.16	21.61	98.6	58
99	2,2′,4,4′,5	Penta	3.14	2.32	11.43	98.7	58
119	2,3′,4,4′,6	Penta	0.57	0.88	3.72	99.8	54
83	2,2′,3,3′,5	Penta	0.19	0.19	0.90	99.7	58
97	2,2′,3,4′,5′	Penta	1.19	0.69	3.21	97.9	58
81 + 87	3,4,4′,5 + 2,2′,3,4,5′	Penta	3.87	2.53	11.23	99.1	58
85	2,2′,3,4,4′	Penta	0.89	1.05	5.95	93.7	58
77	3,3′,4,4′	Tetra	0.31	0.20	0.99	96.8	58

IUPAC no.	Positions	Homologue	Concentration (pg m^{-3})			Vapor fraction	*n*
			Average	St. dev.	Max		
110	2,3,3′,4′,6	Penta	5.32	3.56	15.56	99.2	58
82	2,2′,3,3′,4	Penta	1.81	1.32	5.83	98.4	58
151	2,2,′3,5,5′,6	Hexa	1.48	1.22	5.19	99.3	58
144	2,2′,3,4,5′,6	Hexa	1.19	0.72	3.44	98.6	58
107	2,3,3′,4′,5	Penta	0.61	0.37	1.79	97.5	58
123	2,3′,4,4′,5′	Penta	0.24	0.19	0.71	98.2	58
149	2,2′,3,4′,5′,6	Hexa	4.43	2.52	12.67	98.2	58
118	2,3′,4,4′,5	Penta	12.15	8.07	35.48	98.4	58
134	2,2′,3,3′,5,6	Hexa	0.54	0.25	1.11	85.6	58
114	2,3,4,4′,5	Penta	0.47	0.22	1.30	98.4	58
131	2,2′,3,3′,4,6	Hexa	0.21	0.20	0.82	98.1	58
146	2,2′,3,4′,5,5′	Hexa	1.24	0.83	3.58	99.1	58
153 + 132	2,2′,4,4′,5,5′ + 2,2′,3,3′,4,6′	Hexa	5.94	1.95	11.40	96.1	58
105	2,3,3′,4,4′	Penta	7.217	2.579	14.675	96.8	58
141	2,2′,3,4,5,5′	Hexa	1.966	0.850	4.148	98.2	58
176 + 137	2,2′,3,3′,4,6,6′ + 2,2′,3,4,4′,5	hp/hx	0.055	0.076	0.442	98.8	57
138 + 163	2,2′,3,4,4′,5′ + 2,3,3′,4′,5,6	Hexa	8.041	2.925	18.096	94.9	58
158	2,3,3′,4,4′,6	Hexa	1.388	0.703	3.350	96.9	58
129	2,2′,3,3′,4,5	Hexa	0.128	0.112	0.432	96.7	58
126	3,3′,4,4′,5	Penta	0.051	0.030	0.108	84.3	58
178	2,2′,3,3′,5,5′,6	Hepta	0.204	0.147	0.619	96.9	58
175	2,2′,3,3′,4,5′,6	Hepta	0.090	0.119	0.668	98.9	57
187 + 182	2,2′,3,4′,5,5′,6 + 2,2′,3,4,4′,5,6′	Hepta	0.368	0.221	0.870	91.0	57
183	2,2′,3,4,4′,5′,6	Hepta	0.424	0.291	1.661	87.7	58
128	2,2′3,3′4,4′	Hexa	1.738	0.854	4.738	97.5	58
167	2,3′,4,4′,5,5′	Hexa	0.526	0.184	1.150	97.0	58
185	2,2′,3,4,5,5′,6	Hepta	0.205	1.161	8.896	95.4	58
174	2,2′,3,3′,4,5,6′	Hepta	0.397	0.150	0.737	97.1	58
177	2,2′,3,3′,4,5′,6′	Hepta	0.356	0.0162	0.767	98.4	58
202	2,2′,3,3′,5,5′,6,6′	Octa	0.042	0.024	0.108	99.6	58

IUPAC no.	Positions	Homologue	Concentration (pg m^{-3})			Vapor fraction	*n*
			Average	St. dev.	Max		
171	2,2′,3,3′,4,4′,6	Hepta	0.046	0.026	0.147	99.3	58
156	2,3,3′,4,4′,5	Hexa	0.311	0.110	0.590	97.5	58
173	2,2′,3,3′,4,5,6	Hepta	0.020	0.017	0.060	95.9	44
157	2,3,3′,4,4′,5′	Hexa	0.103	0.092	0.403	99.2	52
200	2,2′,3,3′,4,5,6,6′	Octa	0.052	0.065	0.374	99.7	52
172	2,2′,3,3′,4,5,5′	Hepta	0.053	0.035	0.157	99.4	58
197	2,2′,3,3′,4,4′,6,6′	Octa	0.006	0.010	0.049	66.7	20
180	2,2′,3,4,4′,5,5′	Hepta	0.430	0.151	0.726	87.4	58
193	2,3,3′,4′,5,5′,6	Hepta	0.054	0.082	0.620	81.6	58
191	2,3,3′,4,4′,5′,6	Hepta	0.021	0.017	0.070	98.5	52
199	2,2′,3,3′,4′,5,5′,6′	Octa	0.018	0.012	0.069	98.3	52
170 + 190	2,2′,3,3′,4,4′,5 + 2,3,3′,4,4′,5,6	Hepta	0.213	0.121	0.482	97.3	58
198	2,2′,3,3′,4,5,5′,6	Octa	0.007	0.008	0.029	91.5	32
201	2,2′,3,3′,4,5′,6,6′	Octa	0.145	0.079	0.364	90.8	58
203 + 196	2,2′,3,4,4′,5,5′,6 + 2,2′,3,3′,4,4′,5,6′	Octa	0.176	0.078	0.364	89.4	58
89	2,2′,3,4,6′	Hepta	0.026	0.027	0.088	96.7	41
95	2,2′,3,5′,6	Octa	0.022	0.17	0.079	94.0	49
208	2,2′,3,3′,4,5,5′,6,6′	Nona	0.021	0.021	0.118	95.4	49
207	2,2′,3,3′,4,4′,5,6,6′	Nona	0.015	0.010	0.049	92.5	54
194	2,2′,3,3′,4,4′,5,5′	Octa	0.023	0.017	0.079	93.6	56
205	2,3,3′,4,4′,5,5′,6	Octa	0.011	0.016	0.088	86.5	32
206	2,2′,3,3′,4,4′,5,5′,6	Nona	0.032	0.016	0.079	93.8	57
209	2,2′,3,3′,4,4′,5,5′,6,6′	Deca	0.034	0.027	0.120	60.5	55
	Total PCB		128.4	46.8	232.8	96.6	58

IUPAC, chlorine positions and homologues shown. Fraction of concentration in vapor phase shown in percent. “*n*” is number of detections.

Table 2

PCB concentration (ng g^{-1}) in surface (0–0.5 cm depth) and shallow (1–2 cm depth) soils

Congener	Surface soils		Shallow soils		Ratio
	Average	St. dev.	Average	St. dev.	
31 + 28 Tri	2.40	2.58	0.44	0.76	2.6
52 Tetra	2.13	2.35	0.30	0.59	3.1
41 + 71 Tetra	9.22	5.61	2.62	3.5	2.4
74 Tetra	0.98	0.38	0.37	0.55	2.0
70 + 76 Tetra	1.22	0.44	0.50	0.67	1.9
66 Tetra	1.70	0.68	0.84	1.18	1.6
56 + 60 Tetra	0.85	0.34	0.29	0.49	2.1
84 + 92 + 89 Penta	2.85	1.48	0.90	1.45	2.3
101 Penta	4.15	4.72	0.54	1.32	2.8
99 Penta	1.54	10.76	0.45	1.01	2.1
81 + 87 Penta	2.98	3.62	0.38	0.61	3.8
85 Penta	1.97	2.07	0.30	0.57	3.0
110 Penta	5.56	5.85	0.19	1.77	2.6
151 Hexa	1.48	1.95	0.12	0.19	5.7
144 + 135 Hexa	1.70	2.22	0.17	0.20	6.9
107 Penta	1.70	2.16	0.16	0.22	6.0
149 Hexa	2.00	2.01	0.31	0.59	3.0
118 Penta	4.44	5.08	0.57	1.44	2.8
146 Hexa	2.61	3.16	0.27	0.49	4.2
153 + 132 Hexa	9.91	11.28	1.38	3.21	2.8
105 Penta	3.15	3.66	0.50	0.74	3.3
141 Hexa	2.64	3.40	0.22	0.34	6.0
138 + 163 Hexa	8.36	9.57	1.16	2.54	2.8
158 Hexa	1.64	2.15	0.15	0.27	4.4
178 Hepta	1.37	1.81	0.55	0.46	5.9
187 + 182 Hepta	2.48	3.02	0.25	0.47	4.2
183 Hepta	2.90	3.78	0.24	0.37	5.9

Congener	Surface soils		Shallow soils		Ratio
	Average	St. dev.	Average	St. dev.	
128 Hexa	2.99	3.91	0.27	0.42	5.5
167 Hexa	1.38	1.72	0.14	0.23	4.7
174 Hepta	1.44	1.70	0.13	0.21	5.7
177 Hepta	1.80	2.25	0.17	0.34	4.1
172 Hepta	1.43	2.02	0.62	0.52	5.3
180 Hepta	4.85	5.70	0.66	1.86	2.4
170 + 190 Hept	4.32	5.08	0.59	1.44	2.8
201 Octa	2.05	2.60	0.19	0.38	4.0
203 + 196 Octa	2.51	3.15	0.26	0.56	3.4
194 Octa	1.59	2.24	0.71	0.61	4.5
206 Nona	0.89	1.27	0.41	0.38	3.5
Total PCB	109.64	116.07	19.22	33.23	2.8

Based on 3 sites, composite samples. Indicated congeners found in all samples. Ratio of surface:shallow concentrarions in right hand columns.