Chlorinated Paraffins in Global Air: First Results from the GAPS and GAPS-Megacities Networks

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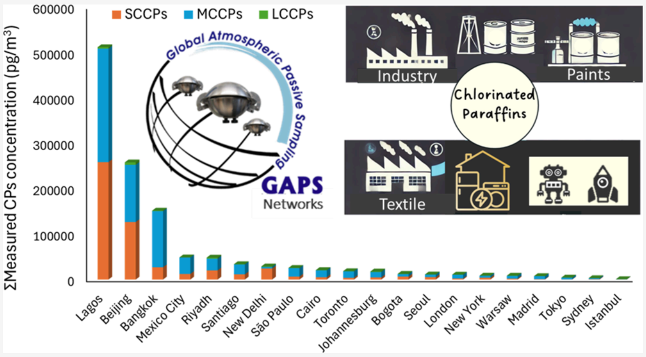
ABSTRACT: This study presents the first global data set of measured chlorinated paraffins (CPs), including short-chain (SCCPs), medium-chain (MCCPs), and long-chain chlorinated paraffins (LCCPs) in ambient air, derived from a single coordinated sampling network, i.e., the Global Atmospheric Passive Sampling (GAPS) network, using a passive sampling approach. Concentrations exhibited pronounced regional disparities, with the combined levels in two megacities—Lagos, Nigeria (512,000 pg/m^3) and Beijing, China (258,000 pg/m^3) exceeding by more than 1.5-fold the combined total levels observed across the rest of the world (~459,000 pg/m^3). Evidence of long-range atmospheric transport was observed at remote sites in western Canada (Little Fox Lake and Whistler), influenced by trans-Pacific air trajectories during the sampling period. These findings underscore the substantial global heterogeneity in the spatial distribution of CPs and the heavily disproportionate contributions of a few regions. Notably, the major producers/emitters, such as China, had several years of delay in ratifying the SCCP listing under the Stockholm Convention (Annex A, elimination since 2017 for congeners with > 48% chlorine content), and some countries have yet to ratify. Without the timely implementation of regulatory measures in these jurisdictions, global concentrations are expected to remain stagnant or even increase if emissions persist at current levels. These results further suggest that substantial time lags are likely before measurable declines in SCCP concentrations, and potentially in recently listed MCCPs, are observed even in regions where control measures are already in place. Hence, this global data set serves as a baseline for future assessments of temporal and spatial trends.



KEYWORDS: *chlorinated paraffins, GAPS network, GAPS-Megacities, passive air sampling, global occurrence, Stockholm Convention*

INTRODUCTION

Chlorinated paraffins (CPs) are a class of synthetic chemicals produced by the chlorination of *n*-alkanes with varying carbon chain lengths and degrees of chlorination. This combination of different carbon chain lengths, chlorination positions and uneven chlorine substitution leading to variable stereochemistry means that nearly half a million CP isomers could potentially exist.[1] CPs have been produced in high volumes and have been widely used in lubricants, plastics, rubbers, and metal-cutting fluids.[2] Due to their widespread production and applications, CPs have been detected in various environmental and biota samples, including outdoor air, soil, and even cetaceans collected from the Arctic, raising concerns about their potential impacts on ecosystems and human health.[3−6] *inter alia*

CPs are classified into three groups based on their carbon chain lengths: short-chain CPs (SCCPs, C_{10-13}), medium-chain CPs (MCCPs, C_{14-17}), and long-chain CPs (LCCPs, $C_{\geq 18}$). They are also classified based on chlorine content, with commercial mixtures such as CP-42, CP-52, and CP-70 referring to approximately 42%, 52%, and 70% chlorine by weight, respectively.[7,8] Due to the structural diversity and analytical complexity of CPs, Fernandes et al. recommended using the

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term "*measured polychlorinated alkanes (PCA)*" when reporting their environmental concentrations.[9] They also advocated reporting the sum of individual chain lengths, where quantifiable, rather than using the aggregated term "total CPs", to improve comparability across studies and better support toxicological evaluations and risk assessment processes.

Owing to their persistence in the environment, toxicity, bioaccumulation, and long-range transport potential, SCCPs with a chlorination content greater than 48% by weight were listed in Annex A (requiring elimination) of the Stockholm Convention on Persistent Organic Pollutants (POPs) since 2017.[10] As a result, MCCPs and LCCPs are increasingly used as substitutes, with growing evidence of their environmental occurrence.[11−13] Very recently, in the Conference of the Parties to the Stockholm Convention on POPs (SC COP-12), MCCPs were also listed as POPs under the Stockholm Convention in Annex A with specific exemptions, requiring Parties to eliminate their production and use.[14] However, the timelines of the listings for entering into force and implementation vary depending on each country's domestic processes, legal frameworks and ratification mechanisms. Under the Convention, new listings enter into force automatically for most Parties approximately 1.5 years after the COP decision (based on the opt-in mechanism chosen when they became Parties to the treaty), except for 18 Parties (10%) that enter into force only after ratification. For instance, SCCPs, listed in 2017, entered into force by December 2018 for 166 of the 186 Parties, whereas others, such as Canada and China, ratified the listing in 2022 and 2023, respectively.[15] Hence, the time lags in implementations of these regulations can result in the continued production, use, and export of listed chemicals during the interim, creating potential "loopholes" in global trade and undermining the effectiveness of collective elimination efforts. Nonetheless, the domestic regulations of Parties can mitigate these lags. For instance, Canada prohibited the manufacture, use, sale, and import of SCCPs and SCCP-containing products in 2013, through listing SCCPs to the Prohibition of Certain Toxic Substances Regulations, four years prior to the international listing, demonstrating that delayed ratification does not necessarily equate to delayed regulatory control in some cases.[16]

Despite global production and environmental release, relatively few studies have measured CP concentrations in air, especially for MCCPs and LCCPs, primarily due to analytical challenges in resolving such complex mixtures.[17] The levels, profiles, and atmospheric transport of CPs remain poorly understood, especially on a global scale. The Global Atmospheric Passive Sampling (GAPS) core network was established in 2005 to monitor POPs and chemicals of emerging concern in global air at over 100 sites, with 40+ sites contributing to long-term monitoring data.[18] The GAPS-Megacities (GAPS-MC) network, initiated in 2018, aims to assess chemical pollution in large urban centers.[19,20] Both networks use double-dome polyurethane foam-based passive air samplers (PUF−PAS), which have been successfully applied to monitor SCCPs in air in global regions including Canada,[21] China,[22−24] India,[25] Australia,[26] and Switzerland.[27] The applicability of PUF−PAS for monitoring MCCPs and LCCPs has also been previously demonstrated in a few studies.[4,21,24,28] Analytically, multiple technologies have been employed to quantify CPs, including gas chromatography coupled with mass spectrometry (GC-MS),[29,30] two-dimensional gas chromatography coupled with mass spectrometry (GC × GC-MS),[31,32] two-dimensional gas chromatography coupled with high-resolution mass spectrometry (GC × GC-HRMS),[33] and high-performance liquid chromatography coupled with high-resolution mass spectrometry (HPLC-HRMS).[34] Each technique offers different strengths, but HPLC-HRMS has shown particular promise in resolving and detecting longer chain CPs, particularly LCCPs.[6,34]

This study aims to assess the global distribution and chemical profiles of the full suite of CPs (SCCPs, MCCPs, and LCCPs) in ambient air. We developed and validated a high-throughput method using HPLC-HRMS for application to PUF−PAS samples from the GAPS networks. While previous studies have predominantly focused on SCCPs—with limited data on MCCPs—this is, to our knowledge, the first study to report global-scale atmospheric concentrations of all three CP groups using a harmonized passive sampling approach.

■ MATERIALS AND METHODS

Passive Air Sampling

Polyurethane foam (PUF) disks were deployed in double-dome type passive samplers as used previously under the GAPS network over decades.[35,36] This sampler design is able to capture both gas- and ambient particle-phase contaminants (up to about 5 mm in size) at similar sampling rates.[37,38] The samplers were deployed at 20 GAPS-MC sites and at the 10 Canadian sites under the core-GAPS network for 3 months (Quarter 3: July-September, with some exceptions) as a part of routine annual sampling campaigns over 4 quarters of the year. Further details of locations, sampling dates and sampling rates are given in Table S1, and the sites map is given in Figure S1 in the Supporting Information (SI).

Extraction and Processing

Samples were extracted using an accelerated solvent extraction system (ASE 350, Dionex Corporation, Sunnyvale, CA, USA). A combination of hexane: acetone (1:1, v/v) with the ASE method set at 70 °C, 1500 psi for two cycles (5 min static cycle with 100% flush and 240 s purge) was used for the extraction of target analytes. Post extraction, extract volumes were reduced to 1 mL using a Turbovap blowdown evaporator (Biotage, Sweden) under a gentle stream of nitrogen, followed by extract cleanup using BondElute Florisil normal phase columns (5 g, 20 mL; Agilent Technologies, USA). Prior to loading the concentrated raw extract onto the column, ∼ 1 g of sodium sulfate was added to the column, which was then preconditioned using 20 mL of hexane:acetone (1:1, v/v). The extract was then loaded onto the column, followed by the elution with 30 mL of hexane:acetone (1:1, v/v). The eluates were blown down to 0.5 mL using Turbovap and N-EVAP nitrogen evaporator equipment, followed by solvent exchange with methanol, before transferring the extracts to GC vials. 50 ng of $^{13}C_{10}$ *anti*-Dechlorane Plus (DP) standard was added as an internal standard. The extracts were push filtered using Whatman Mini-UniPrep syringeless 0.2 μm filters (Cytivia Life Science, USA) before instrumental analysis.

Instrumental Analysis

CPs were analyzed using an ultrahigh performance liquid chromatograph coupled with an Orbitrap Exploris 240 high resolution mass spectrometer (UHPLC-Orbitrap 240 HRMS) (Thermo Fisher Scientific, USA). An instrumental method was adapted from a previously published method by Kutarna et al.[39] and optimized using a Vanquish UHPLC system coupled with an Exploris 240 mass spectrometer. Separation of CPs was achieved with reverse phase chromatography using ZORBAX RRHD (Stable Bond CN, 1.8 μm, 2.1 × 100 mm) fitted with ZORBAX guard column (Agilent Technologies, USA). The injection volume was 5 μL with 0.1 mM ammonium chloride in ultrapure water (A) and 0.1 mM ammonium chloride in methanol (B) as mobile phases. Initially, 30% B was increased to 70% over 1.5 min, then increased to 100% at 2.5 min and held static for 2 min, followed by a decrease to the initial conditions of 30% B and held for 1 min to allow for equilibration. Flow rate was 0.4 mL/min. The column and sample compartment temperatures were maintained at 40 and 4 °C, respectively. Data were acquired in full scan MS mode. Parameters for full scan (100−1100 m/z) recorded at resolution R = 90 000 (at m/z 200) with a maximum of 3 × 106 ions collected within 200 ms. Mass spectrometric settings for negative electrospray ionization mode were as follows: spray voltage, 3.4 kV; sheath gas flow rate, 35 L/h; auxiliary gas flow rate, 7 L/h; capillary temperature, 275 °C; and probe heater temperature, 350 °C. Calibration curves were generated using a concentration series of 0.5, 1.0, 2.0, 3.9, 7.8, 15.6, 31.2, 62.5, 125, 250, and 500 ng/mL, which showed strong linearity ($r^2 > 0.99$). Further details on method optimization results are given in SI, Text S1 and shown in Figure S2.

Quantification Method: Automated Processing in R

Postacquisition processing and quantification of CPs were conducted using a custom-built R script. The script has three major components: construction of the screening database, peak picking/compound screening, and quantification. First, a screening database of CPs was constructed in R based on user input of desired carbon chain lengths. Each line of the database contained a formula with the general pattern $C_XH_{(2x+2-y)}Cl_y$, where y ranges from 1 to x, plus associated exact mass, isotopic abundance, mass-to-charge ratio for the detected $[M+Cl]^-$ ion, and two additional chlorine isotope masses for verification. Compound entries were also automatically grouped into categories based on carbon chain length (SCCP, MCCP, or LCCP).

Next, raw mass spectrometry data files in mzXML format were imported into R. These were processed using functions from the 'xcms' package,[40] with these major steps: centroiding profile-mode data, smoothing, peak picking, peak grouping across samples, peak filling between samples, and retention time correction. The processed mass spectrometry data was then screened against the previously constructed CP database using exact mass matching with a default mass error of 5 ppm. Chlorine isotope peaks were used to verify peak identity, and a peak height cutoff of 10,000 was applied to reduce false positives.

Lastly, the screened CP results were quantified using a modified version of the method proposed by Reth et al.[41] For each technical mixture, peak areas of the most abundant isotope of each congener were collected and normalized by the peak area of the mass-labeled internal standard $^{13}C_{10}$*anti*-DP, which was added to every standard solution and spiked into every sample at the same concentration. Further details adopted from the Reth et al.[41] method are given in SI, Text S2 and Figure S3.

Quality Assurance and Quality Control (QA/QC)

Laboratory blanks were extracted and processed along with field blanks and samples to monitor any contamination occurring during the laboratory processes (Table S2). Recoveries were monitored by adding surrogate standards, i.e., 50 ng each of $^{13}C_{10}H_{16}Cl_7$ and $^{13}C_{12}H_{18}Cl_9$ to all blanks and samples before the extractions. The extraction method was validated by conducting spiked recovery tests. Clean PUF disks were spiked with a known amount of native MCCPs and mass-labeled SCCPs, and underwent the whole procedure as explained above. The recoveries of spiked standards ranged between 80−110%, confirming the validity of the extraction and processing procedures used for this study. The recoveries of surrogate standards added to samples and blanks were 107 ± 50%. The higher variability in recoveries of spiked surrogates to samples is attributed to larger PUF matrix interferences, which have been observed previously for other suites of compounds analyzed in PUFs in the GAPS networks.[42] The data was blank corrected using averages of laboratory and field blanks but not recovery corrected, hence the air concentrations reported here should be considered semiquantitative estimates of the real environmental levels of CPs. Method detection limits (MDLs) were estimated as the average of blanks ± 3 × standard deviation and are listed in SI, Table S2.

The GAPS template (given in Supporting Information) was used to measure the equivalent air volume (V_{eq}, m^3) sampled by PUF at each site to convert mass sampled by PUF to corresponding air concentrations.[43] The GAPS template corrects V_{eq} for the average temperature during the sampling period. Site-specific sampling rates were used for calculations that consider the site-specific meteorological data, such as wind speed (WS) based on the sampling dates and location coordinates (Table S1).[44] Further details on the calculation of sampling rates are provided by Schuster et al.[35] This template has been used in the past for previous reports under the GAPS network and other regional studies. The sampling rates used in this study were within the range as measured by He et al. in a field calibration study.[24]

RESULTS AND DISCUSSION

Spatial Profile

Chlorinated paraffins, hereafter, refer to measured polychlorinated alkanes with carbon chain length from C_{10} to C_{20} (i.e., $\Sigma PCAs\text{-}C_{10-20}$) with variable chlorine contents, as quantified under this study. The results have been reported as the sum of measured SCCPs, MCCPs and LCCPs that refer to $\Sigma PCAs\text{-}C_{10-13}$, $\Sigma PCAs\text{-}C_{14-17}$, and $\Sigma PCAs\text{-}C_{18-20}$, respectively, as well as sums of individual chain lengths, as recommended by Fernandes et al.[9] Results for descriptive statistics for GAPS-MC and core-GAPS networks are given in Table 1. Detailed results for each site and the sums of individual carbon chain lengths with variable chlorine content are given in Tables S3 and S4, respectively. Overall, across most of the sampling sites, CP profiles were dominated by MCCPs, followed by SCCPs, together accounting for up to 95% of total CPs. LCCPs typically contributed ≤ 5% of the total CP burden. Detailed results are given below and reported to 3 significant figures for ease of comparison.

Global—GAPS-Megacities Network:

Global air concentrations of total measured chlorinated paraffins ($\Sigma PCAs\text{-}C_{10-20}$) ranged from 3000 to 512,000 pg/m^3 (Figure 1 and Table S3). The highest levels were observed in Lagos, Nigeria, where SCCPs, MCCPs and LCCPs were measured at 259,000, 25,0000 and 3010 pg/m^3, respectively. Lagos hosts one of the world's largest dumpsites, which receives thousands of tons of unsorted waste, including substantial amounts of plastic and electronic waste.[45] Inadequate waste management practices, such as open burning, are known to release chlorinated contaminants into the atmosphere, consistent with previous

Table 1. Summary of Measured CP Concentrations (pg/m^3) in the Passive Air Samples under the Core-GAPS and GAPS-Megacities (GAPS-MC) Networks[a]

Network	Min−Max	Average	Median
	SCCPs (ΣPCAs-C$_{10-13}$)		
Core-GAPS (*n* = 10)	0.653*−3520	539	16.9
GAPS-MC (*n* = 20)	267*−259,000	26,200	5180
	MCCPs (ΣPCAs-C$_{14-17}$)		
Core-GAPS	56.4*−5890	1800	1090
GAPS-MC	428*−250,000	34,500	10,200
	LCCPs (ΣPCAs-C$_{18-20}$)		
Core-GAPS	6.97*−890	114	14.4
GAPS-MC	5.79*− 5450	824	333

[a] N represents the number of sites under each network monitored for this study. Asterisk (*) indicates the values below the method detection limit (given in Table S2) after blank correction and were included as is in average and median calculations. The values are reported to 3 significant figures.

observations for flame retardants.[18] The next-highest levels were observed at Beijing, China and Bangkok, Thailand, with total quantifiable CPs measuring at 258,000 and 152,000 pg/m^3, respectively. Elevated concentrations observed in Beijing align with China's status as the world's leading producer and consumer of CPs since the early 2000s, with a total CP production of up to 1.4 million metric tons in 2020.[46,47] Although SCCPs (> 48% chlorination by weight) were listed under the Stockholm Convention in 2017, China only ratified the listing recently in 2023, which implies that SCCPs were still being manufactured/used in the country.[48] Such delays in ratification and/or implementation can undermine the Convention's effectiveness, as a global decline in atmospheric SCCP concentrations would not be expected while emissions persist from the world's largest producer. Guida et al. also highlighted the challenges pertaining to effective implementations of regulations under the Stockholm Convention,[8] given that there are several other substantial producer countries, e.g., India, Russia,[49] that have not ratified the SCCPs listing as of August 2025. Consequently, the 2023 concentrations from GAPS networks reported here may represent near-maximum levels in the global atmosphere and therefore provide a valuable baseline for assessing future reductions in the levels due to the implementation of control measures by Parties to the Stockholm Convention. The concentration levels in the remaining mega/major cities were orders of magnitude lower than Lagos, Beijing and Bangkok, spanning from 48,400 pg/m^3 (Mexico City) to 3000 pg/m^3 (Sydney). Istanbul was the only GAPS-MC site where the sum of measured SCCPs, MCCPs and LCCPs (270, 430, and 20 pg/m^3, respectively) was below MDL after blank correction.

Air concentrations were also compared based on the five United Nations region groupings—Western Europe and Others Group (WEOG), Latin American and Caribbean Group (GRULAC), Eastern Europe (EE), Asia-Pacific, and Africa (Figure 2a). The median concentrations were highest and similar for the Africa, Asia-Pacific and GRULAC regions, while the WEOG and EE regions were about half and an order of magnitude lower, with a range of levels between 695−18,400 pg/m^3. A comparison was also made among sites located in the Northern and Southern Hemispheres (Figure S4). Lagos and Beijing were excluded from this analysis to avoid skewing the Northern Hemisphere distribution due to their exceptionally high concentrations. With a larger number of sites, the Northern Hemisphere (excluding Lagos and Beijing) displayed a wider spread and greater variability compared to the Southern Hemisphere. Nevertheless, the mean and median concentrations differed by less than a factor of 2, indicating that while hemispheric differences are evident, local sources and urban activities remain the dominant drivers of CP variability within each hemisphere.

The ΣMCCPs/ΣSCCPs ratios (M/S) for GAPS-MC sites are given in Table S3. Higher MCCPs/SCCPs ratios were observed for Tokyo (8.39), Madrid (7.49), Bangkok (4.61), Toronto (3.78), as well as in Cairo, Johannesburg, Warsaw, Mexico City, São Paulo and Sydney (all with ratios between 2−3), indicating a shift to the use of commercial formulations and/or products containing more MCCPs. Contrary, MCCPs/SCCPs ratio of ≤ 1 was observed for Lagos, Beijing, Seoul, Bogotá, New York and New Delhi, which reflects SCCPs' continuous emissions from

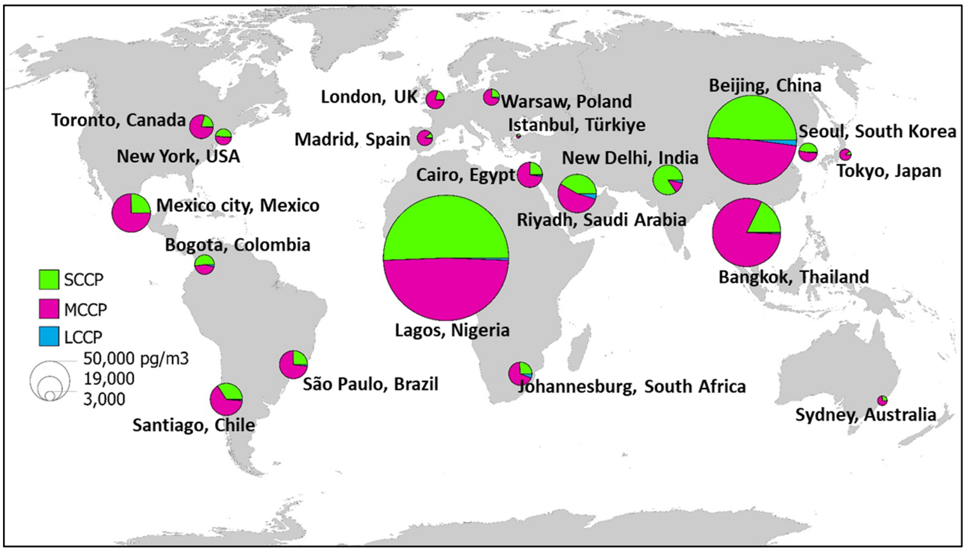

Figure 1. Pie chart representing the global air concentrations (pg/m^3) and contributions of the sum of measured SCCPs (ΣPCAs-C$_{10-13}$), MCCPs (ΣPCAs-C$_{14-17}$) and LCCPs (ΣPCAs-C$_{18-20}$) in samples from the year 2023. Map source: Esri, ArcGIS Online basemap.

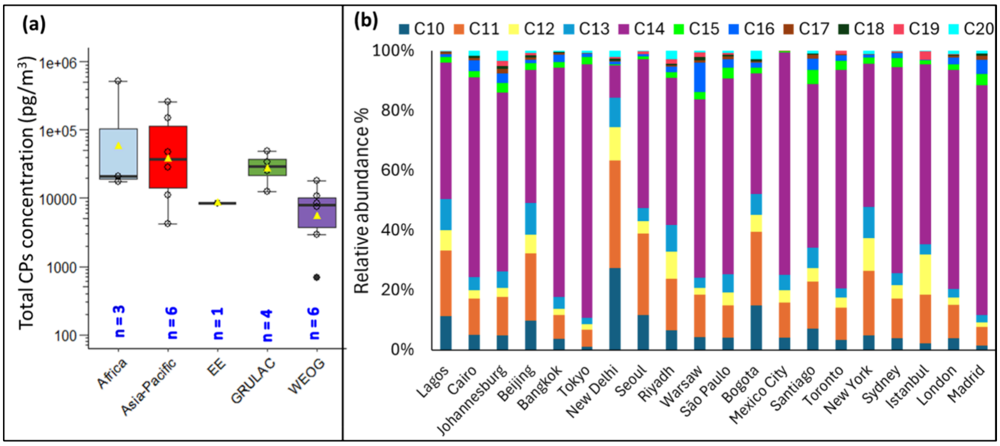

Figure 2. (a) Comparison of concentrations (during 2023) of total measured CPs (ΣPCAs-C_{10-20}) as shown in box and whisker plots across the five United Nations regional groups. Yellow triangle markers indicate the average; top, middle and bottom lines of the boxes show 75th percentiles, medians and 25th percentiles, respectively, and whiskers represent the minimum and maximum total concentration. N represents the number of sites sampled in the given region. UN regions: Africa; Asia-Pacific; EE (Eastern Europe), GRULAC (Group of Latin American and the Caribbean countries); WEOG (Western Europe and Other States Group). (b) Relative abundance of measured homologues based on carbon chain length for SCCPs (ΣPCAs-C_{10-13}), MCCPs (ΣPCAs-C_{14-17}) and LCCPs (ΣPCAs-C_{18-20}) as observed at GAPS-MC study sites. Sites are arranged in order of UN regions given in the left panel.

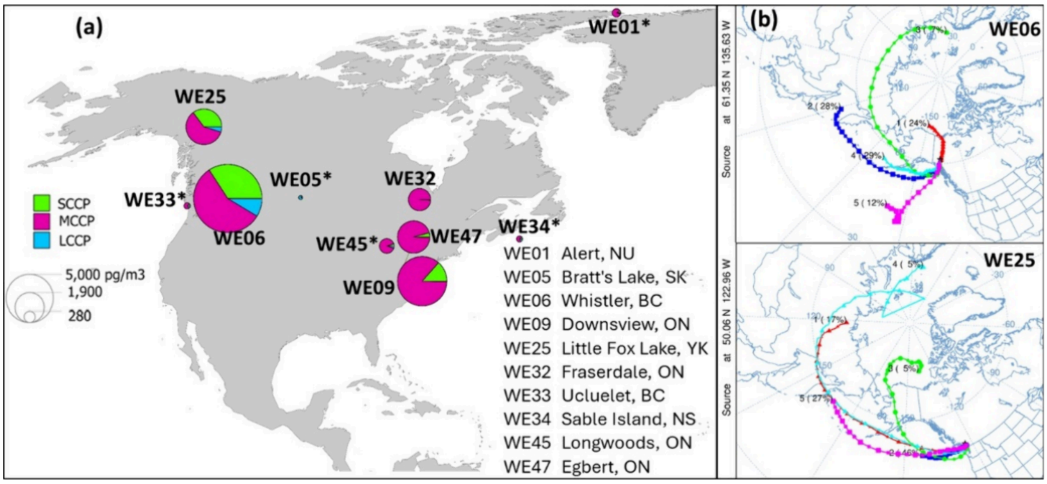

Figure 3. (a) Pie chart representing the Canadian air concentrations (pg/m^3) and contributions of the sum of measured SCCPs (ΣPCAs-C_{10-13}), MCCPs (ΣPCAs-C_{14-17}) and LCCPs (ΣPCAs-C_{18-20}) in samples from the year 2023. The sites with asterisks have data < MDLs. Map source: Esri, ArcGIS Online basemap. (b) Cluster analysis of air mass back trajectories arriving at Whistler (WE06) and Little Fox Lake (WE25) sites. Trajectories were computed using the HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model developed by NOAA's Air Resources Laboratory (https://www.ready.noaa.gov/hypub-bin/trajtype.pl). The model was initialized with GDAS one-degree archive meteorological data and run for 15-day back trajectories for every hour at a receptor height of 200 m above ground level (4400 trajectories over the sampling period per site). Each color represents a distinct cluster of similar trajectories based on their spatial paths, with clustering performed using Ward's hierarchical method.

production (e.g., in China and India) or release from in-use products, stockpiles and end-of-life products in the waste phase (e.g., in Lagos). Correlation analysis was performed between measured total CPs (ΣPCAs-C_{10-20}) and several demographic and socioeconomic indicators, including total population, population density, Gross Domestic Product (GDP), Industrial Production Index (IPI), and Human Development Index (HDI) (Table S1) of the cities in the current study. All variables showed statistically significant relationships but weak correlations (Figure S5). This pattern suggests that population-related and economically driven activities exert a modest influence on CP levels in air. The weak associations likely reflect the complexity in sources and environmental fate of CPs, which are shaped by multiple interacting factors such as industrial emissions, waste management practices, diffuse urban sources, and long-range atmospheric transport that are not fully captured or explained by these broad socioeconomic and demographic indicators. Besides, no correlation was observed among previously reported[18] organophosphate esters, flame retardants and CPs in the same mega/major cities, likely due to their distinct sources and fate in the atmosphere.

Canada—Core-GAPS Network

Total CP concentrations at ten Canadian sites under the core-GAPS network ranged from 35 (< MDL) to 10,300 pg/m^3 (Figure 3a). These sites represent a range of land-use types and locations, including five background, three rural, one semiurban, and one polar site (Table S1). The background site, Whistler in British Columbia, exhibited the highest levels of SCCPs, MCCPs and LCCPs measured at 3520, 5890, and 890 pg/m^3, respectively. Although Whistler is situated at a high elevation, it may still be influenced by local emissions resulting from tourism and recreational activities such as skiing. Another background site, Little Fox Lake (LFL) in Yukon, also exhibited noticeably high levels of CPs, with SCCPs, MCCPs, and LCCPs measured at 991, 1690, and 134 pg/m^3, respectively. Previous studies have identified the influence of long-range atmospheric transport (LRAT) in this region of western sub-Arctic Canada, highlighting the role of westerly winds from the Pacific and East Asia in transporting contaminants to remote sites, including LFL.[50,51] Backward air parcel trajectories were assessed for Whistler and LFL sites using the US National Oceanic and Atmospheric Administration (NOAA) Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model with Global Data Assimilation System (GDAS) one-degree archive meteorological data for the sampling period.[52] The model was run for 15-day back trajectories for every hour at a receptor height of 200 m above ground level, with a total of 4400 trajectories over the sampling period for each site. However, only one-third of trajectories were used for cluster analysis due to HYSPLIT's limitations in processing all trajectories simultaneously. Trajectory clustering was performed using the built-in clustering algorithm in HYSPLIT, which groups trajectories based on their geometric similarity using Ward's minimum variance method. This analysis showed the pattern of trans-Pacific movement of air mass with more than 60% of trajectories originating from East Asia/Russia that arrived at Whistler and LFL sites (Figure 3b). Hence, apart from possible influences of local sources at Whistler, these two sites have the impacts from trans-Pacific transport of contaminants, which is also supported by elevated levels seen at East Asian sites under the GAPS-MC network.

The semiurban site in Toronto, Ontario (Downsview)—characterized by proximity to light industry, commercial areas, and residential buildings within a 5 km radius—recorded the second-highest concentrations of SCCPs and MCCPs (725 and 4650 pg/m^3, respectively) among core-GAPS sites, whereas LCCPs at this site were below MDL (15.1 pg/m^3). Fraserdale and Egbert, Ontario, had MCCPs measured at 1090 and 2200 pg/m^3, respectively, whereas SCCPs and LCCPs were below MDL at both sites (Table S3). In contrast, concentrations of all three CP classes were below MDL at the polar site in Alert, Nunavut, as well as at several rural and background sites, including Bratt's Lake (Saskatchewan), Ucluelet (British Columbia), Sable Island (Nova Scotia), and Longwoods (Ontario), their data are provided in Table S3 for reference.

Comparison to the Literature

Details of previous studies from 2005 to 2025, reporting on CP levels in air are given in Table S5. Overall, the concentrations measured at GAPS-MC sites were generally in agreement (within an order of magnitude) with previous studies reporting atmospheric CP concentrations in similar regions. In the current study, the highest atmospheric concentrations of both SCCPs and MCCPs were observed at an urban site in Lagos, Nigeria. However, to the best of our knowledge, there is no other study that has reported CP concentrations in Nigeria to date, and very limited monitoring studies have been conducted in a few other African countries. Arko et al. reported on CP levels in Ghana using PUF−PAS in source-sector based measurements, with SCCPs and MCCPs ranges of 50−15,200 and 1780−240,000 pg/m^3, respectively.[28] The elevated levels were observed at an e-waste site in Ghana, linking it to poor waste management, such as open burning practices. Nipen et al. also reported CPs measurements in Tanzanian air at rural, suburban, urban and dump/e-waste sites, with the waste-source transect and urban locations having the highest individual concentrations of CPs (ranges of SCCPs and MCCPs reported were: 300−63,000, < 400−35,000 pg/m^3, respectively).[4] In comparison to the levels observed at Lagos, Nigeria, in the current study, the atmospheric CP levels in Ghana, in particular MCCPs, are in close agreement, while the levels in Tanzania are more than an order of magnitude lower. Nevondo and Okonkwo[53] and Arko et al.[28] have highlighted the research gaps related to atmospheric studies on CPs in Africa as compared to other UN regions, which is linked to a lack of sampling resources and analytical capacity required for complex analyses of CPs.

China has the most abundant data reported so far for all three CP classes, with active air sampling being the widely used technique to collect the air samples. A wide range of CP concentrations has been reported in the Chinese atmosphere,[23,54−61] *inter alia* with a maximum ΣSCCPs level of 881,000 pg/m^3 in Beijing,[59] which is approximately 3.5 times higher than the levels reported in the current study. Niu et al. reported ΣSCCPs in a range of 6100 to 63,000 pg/m^3 in the Yangtze River Delta, China, comprising rural, suburban, and urban areas, with urban levels being comparable to the current study.[23] In terms of ΣMCCPs, the highest reported levels so far were 1,136,400 pg/m^3, measured near a CP production plant (location unknown).[60] The highest reported levels of LCCPs of 17,400 pg/m^3 were measured in the Pearl River Delta, China,[7] ~ 3 times higher than the current study. Dong et al. reported 22,000 and 1600 pg/m^3 of ΣSCCPs and ΣMCCPs, respectively, in rural environments of Beijing in 2022,[61] which were orders of magnitude lower than the current study, likely due to fewer emission sources as compared to urbanized environments. In the remaining parts of Asia, CP levels have been reported in India, Pakistan and Vietnam with a maximum ΣCPs of 85,600, 23,700 and 18,600 pg/m^3, respectively. These levels are in a similar range as observed in New Delhi, Seoul and Riyadh, but orders of magnitude higher than the levels measured in Tokyo (4200 pg/m^3) in the current study.

In Europe, the maximum levels of SCCPs and MCCPs reported in Switzerland were 3300 and 26,000 pg/m^3, respectively, measured in Zurich in 2012.[62] Balla et al. monitored CP levels in Athens, Greece, where the concentrations of measured SCCPs and MCCPs ranged from 290 to 25,500 pg/m^3 and from 1100 to 19,000 pg/m^3, respectively.[13] In comparison, the levels reported at European sites under the current study, including Madrid, Warsaw and London, were toward the lower end of these ranges (Table S5).

In terms of polar locations, in the Norwegian Arctic, Borgen et al. measured SCCPs at a background location of Mt. Zeppelin, Svalbard, ranging from 9.0 to 57 pg/m^3 in 1999,[63] whereas levels from 1800 to 10,600 pg/m^3 were measured in air at Bear Island, between Svalbard and mainland Norway.[64] In polar urban air, ranges of measured concentrations of SCCPs, MCCPs and LCCPs in three Nordic capitals of Finland, Norway and Iceland were 25−430, 63−1100 and 2−1370 pg/m^3, respectively.[65] In another study conducted in the urban location of Tromsø, only SCCPs were reported at the levels of 270 ± 100 pg/m^3.[66] In the Antarctic region, SCCP and MCCPs concentrations detected at the background sites were up to 4600 and 35 pg/m^3 for the years 2014−2018.[67] In comparison to these studies, the levels measured in the Canadian Arctic in the current study (although < MDLs) were 12.8, 137, and 6.97 pg/m^3 for SCCPs, MCCPs and LCCPs, respectively. However, the levels measured at remote locations of Whistler and Little Fox Lake in Canada (impacted by trans-Pacific LRAT from Asian sources) were ≥ 2 times higher than the Nordic urban air.

Within Canada, there are very few reports on atmospheric levels of CPs. Tomy reported average concentrations of SCCPs in air samples ranging from 65 to 924 pg/m^3 at Egbert, Canada in 1990,[68] whereas, the concentration of ΣSCCPs in air over Lake Ontario was 249 pg/m^3.[69] In comparison, SCCPs levels measured at Egbert in the current study were 97.3 pg/m^3 (< MDL) while MCCPs were present at levels of 2200 pg/m^3. More recently, Niu et al. reported ΣSCCPs levels from 500 to 78,500 pg/m^3 at different source-sector based locations.[21] In the same study, average SCCP levels at the Toronto site (same location as under the current study) were ∼ 30,000 pg/m^3, which is approximately an order of magnitude higher than the levels reported under the current study. This difference might be due to the reduced/regulated use of SCCPs in Canada due to domestic as well as international regulations, and also attributed by differences in sampling durations (multiple periods in 2017/2018 versus one period in 2023) to a certain extent.

Homologues Composition and Profile

The composition of short-, medium- and long-chain CPs was variable among the cities (Figure 2b). Lagos, Beijing, Seoul, Riyadh, Bogotá and New York had about 40−50% of SCCPs, whereas the remaining cities except New Delhi, were 65−85% of MCCPs. In the New Delhi sample, SCCPs were 85% of the total composition. In each sample, the carbon chain length-based homologue profile was dominated by C_{14} followed by C_{11} at all sites, except New Delhi, India where C_{10} and C_{11} dominated the profile. Note that India has not ratified the SCCP listing yet under the Stockholm Convention.[15] There are several manufacturers in and around New Delhi (anecdotally via web search) that might still be using feedstocks and/or producing technical mixtures of different chain lengths, including SCCPs. Moreover, the existing consumer products containing SCCPs across these major cities also contribute to their continued emissions to the atmosphere. Guida et al. analyzed the homologue compositions of 36 technical mixtures from different global regions with their production dating back to the 1970s.[8] The study highlighted significant variability in the homologue composition and chlorine content, including those originating from the same manufacturer. The study[8] further emphasized that, due to the lack of transparent and accurate information regarding the contents of feedstocks and mixtures, producers or importer countries may unknowingly be incorporating substantial quantities of SCCPs into their products, which could have implications on the successful implementation of restrictions under the Stockholm Convention.

Chlorine-based homologue profiles of SCCPs and MCCPs were characterized by a predominance of congeners with lower chlorine content (Figure S6), which is consistent with previous findings.[13,24,28] The abundance of Cl_6 congeners was observed in C_{10}, C_{11}, and C_{14} carbon-chain lengths, whereas Cl_7 dominated C_{12}, C_{13}, C_{15} and C_{16} carbon-chain length congeners. In contrast, congeners with C_{17} and above carbon-chain length exhibited variable profiles, likely due to their relatively low contributions in the samples. Li et al. reported a similar dominance of lower chlorine length homologues in the composition profiles of commercial mixtures CP-42 and CP-52.[7]

The observed homologue patterns in air reflect both source composition and influence of physicochemical properties on atmospheric behavior. Lower-chlorinated, shorter-chain CPs exhibit higher vapor pressures and lower K_{OA}, favoring volatilisation from sources, gas-phase partitioning and long-range mobility,[17] while their faster photolytic and/or OH-driven degradation further reshapes the homologue pattern in atmosphere.[70] In contrast, higher-chlorinated congeners possess lower volatility and partition predominantly to ambient particles, resulting in greater persistence in air. While CP fate is complicated and data are still emerging, especially for MCCP and LCCPs, enrichment of lower-chlorinated CP homologues in air profiles typically reflects contributions from more volatile technical mixtures and diffuse urban sources. Elevated proportions of highly chlorinated CP homologues in air more strongly reflect local industrial emissions of less volatile, more chlorinated formulations, consistent with expected rise in MCCP/LCCP-rich formulations as SCCP substitutes.

MULTIVARIATE ANALYSIS

Principal component analysis and hierarchical clustering analysis were conducted on the log-transformed SCCPs, MCCPs and LCCPs concentrations to show how cities relate to each other in terms of their CP concentration and profiles. Principal component analysis was reduced to two principal components (PC1 and PC2) explaining the majority of the variance (likely associated with total CP burden) as well as capturing secondary patterns (e.g., variation in CP type proportions). Figure 4 displays the distribution of cities based on CP profiles, with superimposed loading vectors representing the contribution of SCCPs, MCCPs, and LCCPs. The sites with the highest total concentrations, i.e., Lagos, Beijing and Bangkok, were positioned together in the top-right quadrant far along the PC1 axis, whereas Istanbul, Tokyo and Sydney, with lower CP concentrations, were placed toward the left of the plot, indicating differing sources and/or composition profiles. The sites with concentrations above 75th percentile, i.e., Bangkok and Riyadh, were located in the middle of the right quadrants,

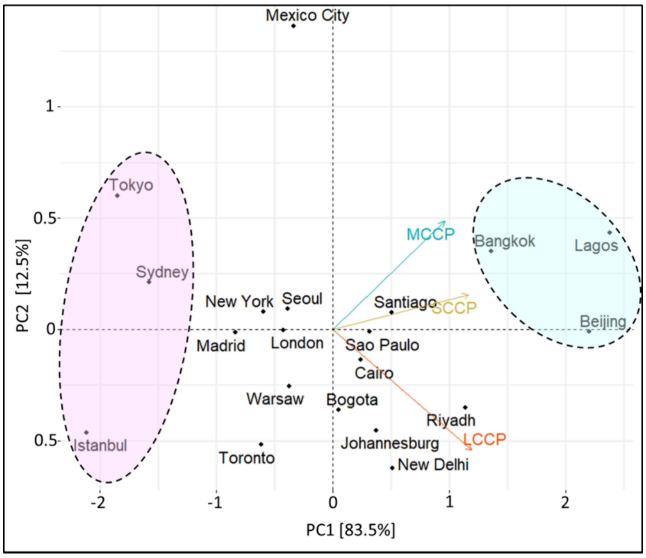

Figure 4. Principal component analysis biplot (score and loading plot) of the sum of measured concentrations (log-transformed) in air for SCCPs (ΣPCAs-C_{10-13}), MCCPs (ΣPCAs-C_{14-17}) and LCCPs (ΣPCAs-C_{18-20}) in 2023. Components 1 and 2 account for 96% variability in the data set. The blue and pink ovals show the sites with the highest and lowest total concentrations, respectively, clustering on the opposite ends of the score plot.

whereas the 13 sites with levels up to the 25th percentile clustered together around the origin. The remaining cities, except Mexico City, which was an outlier (perhaps due to substantially lower LCCP levels compared to SCCPs and MCCPs), were clustered in the center of the plot, indicating a similar origin, likely linked to emissions from their use in industrial processes and/or consumer products. Principal component analysis based on homologue profile data (Table S4) was also performed, using the log-transformed proportions, which provided complementary insight into compositional differences independent of total CP burden (Figure S7). In this analysis, PC1 explained 78.8% of the variability and separated cities according to the dominance of lower-chlorinated versus higher-chlorinated homologues. Beijing, Lagos, Riyadh, and New Delhi aligned strongly with the C_{10-C14} vectors in the upper-right quadrant, reflecting dominance of more volatile homologues. In contrast, Bangkok, Johannesburg, and Santiago showed stronger influence from MCCP (C_{15-17}) and some LCCP (C_{18} and C_{20}) homologues, consistent with less volatile, higher-chlorinated patterns. Cities such as London, Warsaw, São Paulo, and Bogotá clustered closer to the origin, suggesting intermediate or mixed CP signatures without strong dominance of any single homologue group. Hierarchical clustering analysis (Figure S8) also supported the principal component analysis findings, categorizing cities into three main groups: high, intermediate, and low CP burden. The horizontal lines represented clusters, with shorter lines meaning more similarity among sites. Together, these multivariate analyses highlight geographically distinct compositional patterns that may reflect differences in CP manufacturing, use patterns, regulatory controls, and atmospheric processing.

■ CONCLUSION

This study reports and compares CP concentrations in ambient air across 20 global cities, which provides a global baseline for future assessments of temporal and spatial trends. At the time of sampling in 2023, only SCCPs had been listed under the Stockholm Convention, with a few countries only recently ratifying the listing. The data reveal pronounced spatial variability in the distribution of SCCPs, MCCPs, and LCCPs, and indicate that cities such as Lagos, Beijing, and Bangkok exhibit substantially elevated levels of SCCPs and MCCPs, likely reflecting regional usage patterns, waste management practices, industrial activities, and regulatory differences. In contrast, cities including Tokyo, Sydney, and Istanbul show consistently low CP levels across all chain lengths. Principal component analysis and clustering approaches further demonstrated clear groupings of cities with similar CP profiles, underscoring distinct regional signatures and possibly differing sources or environmental fate and behaviors of these chemicals. The separation of LCCPs from SCCPs and MCCPs in multivariate space also highlights the necessity of chain-length-specific monitoring and reporting, especially when the industry is likely to shift toward LCCPs as a replacement after the recent listing of MCCPs under the Stockholm Convention. Overall, this global snapshot of atmospheric CPs underscores the need for harmonized international monitoring frameworks and targeted mitigation strategies, particularly in regions with high ambient concentrations and limited regulatory oversight. We recommend that, given the complexity of the analysis, GAPS networks samples be analyzed for CPs during every 6-year cycle of the Global Monitoring Plan, for the purposes of Effectiveness Evaluation, in which case the next analysis would be for samples collected in 2029. In the case of MCCPs, global baseline data from this study (2023) will be available for the period prior to listing of MCCPs in 2025. However, similar to SCCPs, the inherent time lag of about 1.5 to 2 years or more for the period between listing of a chemical/class and when most Parties ratify and begin implementing control measures, means that the 2029 data is expected to represent the period immediately following a sharp decrease in production/use of MCCPs.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsestair.5c00350.

Sites and sampling information for PUF−PAS deployed in Quarter 3 in 2023 (Table S1); averages (pg/m^3) of lab blanks (LB) and field blanks (FB) and method detection limit (MDL) (Table S2); concentrations (pg/m^3) as sum of measured SCCPs, MCCPs, and LCCPs at core-GAPS and GAPS-Megacities sites (Table S3); total of individual carbon chain lengths (C_{10}−C_{20}) with variable chlorine contents measured at each site given as ng/sample at core-GAPS and GAPS-Megacities sites (Table S4); summary of globally reported SCCP, MCCP, and LCCP concentrations in outdoor air in the studies reported since 2005 (Table S5); map showing the sampling sites monitored under this study (Figure S1); representative chromatograms of 1 μg/mL technical mixtures of chlorinated Paraffins before (a) and after (b) method optimization (Figure S2); response factor plots generated from technical standard mixtures data by R script as part of automated data processing (Figure S3); comparison of concentrations (during 2023) of total measured CPs as shown in box and whisker plots across the Northern Hemisphere (excluding Lagos and Beijing) and Southern Hemisphere (Figure S4); correlation plots of measured total CP levels (ΣPCAs-C_{10-20}) and demographic and socioeconomic indicators (Figure S5); chlorine-based homologue profiles of CPs assessed for a subset of sites in each UN region (Figure S6); principal component analysis biplot (score and loading plot) of the total of individual carbon chain lengths with variable chlorine chain length (log-transformed) in air (Figure S7); hierarchical clustering heatmap of log-transformed concentrations of SCCPs, MCCPs, and LCCPs across 20 global cities (Figure S8); GAPS Template: PUF Disk Effective Air Volume Calculation for Chlorinated Paraffins (XLSX)

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