Science of the Total Environment 954 (2024) 176252

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Impact of anthropogenic activities on atmospheric chlorinated paraffins in Ghana using polyurethane foam disk - passive air sampler

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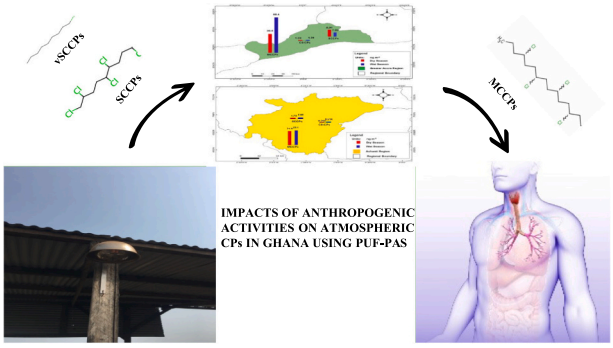
HIGHLIGHTS

- MCCPs dominated the atmospheric CPs in all the land use types of Ghana.
- E-waste was a significant source contributing to atmospheric CP pollution in Ghana.
- The non-cancer risks associated with CP exposure were within acceptable ranges in Ghana.
- Atmospheric MCCPs indicated a high potential cancer risk for the Ghanaian population.

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Paromita Chakraborty

Keywords:
Chlorinated paraffins
Electronic waste or e-waste
Risk assessment
Passive air sampler
Ghana

ABSTRACT

Chlorinated paraffins (CPs) are a global concern due to their high production, ubiquity in the environment and potential toxicity. In Ghana, there is a significant research gap on the concentration and sources of CPs in the air, as well as insufficient regular monitoring programs to track CP levels over time. This study utilized polyurethane foam-based passive air samplers (PUF-PAS) to examine the concentrations, sources and potential human health risks of CPs in the atmosphere surrounding e-waste sites, urban areas, commercial areas and control/background areas in Ghana. The medium-chain CPs (MCCPs) dominated with an average concentration of 26.0 ± 40.1 ng/m^3 and ranged from 1.78 to 240 ng/m^3. Short-chain CPs (SCCPs) ranged from 0.05 to 15.2 ng/m^3 and had an average concentration of 3.48 ± 3.99 ng/m^3. The very short-chain CPs (C9-CPs), had an average concentration of 0.544 ± 0.524 ng/m^3 and ranged from 0.091 to 2.14 ng/m^3. MCCPs exceeded SCCPs by a factor of 7.5 and C9-CPs by a factor of 48. $C_{14}Cl_8$ was the dominant congener in MCCPs and $C_{10}Cl_7$ was also the dominant congener in SCCPs. E-waste was the main contributor to SCCPs and MCCPs (>30 %) in Ghana. The assessed non-cancer risks associated with CP exposure were within acceptable ranges. For cancer risk, MCCPs indicated high potential health risk but C9-CPs and SCCPs showed low risk. To the best of our knowledge, this is the first study on CPs in

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https://doi.org/10.1016/j.scitotenv.2024.176252
Received 18 July 2024; Received in revised form 9 September 2024; Accepted 11 September 2024
Available online 13 September 2024

Ghana's atmosphere, and e-waste was identified as the country's main source of CPs. This study will help regulatory bodies create policies and procedures to control the use and disposal of chlorinated paraffins.

1. Introduction

Chlorinated paraffins (CPs), also called polychlorinated *n*-alkanes, are mixtures of *n*-alkane homologues with different degrees of chlorination (Huang et al., 2023b; Huang et al., 2023a; IARC, 1990). Based on their number of carbon atoms, CPs are divided into short-chain (SCCPs, $C_{10–13}$), medium-chain (MCCPs, $C_{14–17}$), and long-chain (LCCPs, $C_{>17}$) chlorinated paraffins. Very short-chain CPs (e.g., hexane- to nonane-CPs, C_6–C_9 CPs) have also been detected in the environment (Wang et al., 2020; Xia et al., 2019; Xia et al., 2021; Xia et al., 2021b; Yuan et al., 2021; Zhou et al., 2019), although no consistent definition exists for them. Since CPs are persistent, bioaccumulate, toxic and able to undergo long-range transport, they are categorized as persistent organic pollutants (POPs) (UNEP, 2015, 2021; USEPA, 2009, 2015). CPs are anthropogenic compounds and have been widely used as metalworking fluids, plasticizers, flame retardants, paints, sealants, adhesives, rubber, textiles and leather production, and other plastic products (Chen et al., 2022; Guida et al., 2020; van Mourik et al., 2016). CPs are emitted during production, storage, transportation, recycling, and disposal (Guida et al., 2020; van Mourik et al., 2016) and detected in soil, air, indoor dust, and water (Li et al., 2018; Wang et al., 2019a; Wong et al., 2017; Yuan et al., 2022).

Large quantities of CP-containing products such as plastics from computers, polyvinyl chloride (PVC) coatings used as insulation of copper wires, and other plastics as flame retardants are discarded as e-waste (van Mourik et al., 2016). Agbogbloshie, located in central Accra, Ghana's capital, is described as Africa's largest electronic waste dumpsite and has drawn international attention. In urban areas, there is no proper management of waste disposal, and therefore waste containing CPs like furniture, toys, artificial leather, sports equipment, stickers, polyethylene terephthalate (PET), polyethylene (PE), and polypropylene (PP) is burned, which contaminates the atmosphere (Guida et al., 2020). Also in urban areas, CPs are released into households since they are found in most consumer goods and products during their use (Brandsma et al., 2019; Gallistl et al., 2017; Gallistl et al., 2018; Wang et al., 2019; Xu et al., 2019; Yuan et al., 2017). Typically, little information is available regarding pollutant emissions or concentrations at these sites, including the ambient and breathing zone concentrations necessary to understand exposure risks to workers and the community.

There have been reports about CPs in soils in Ghana (Asante et al., 2019; Mockel et al., 2020), but no studies have reported on the simultaneous profiling of C_9-CPs, SCCPs and MCCPs in the air from Ghana. Therefore, the aims of the study are: 1) to ascertain the levels of CPs and homologue patterns in Ghana, 2) to investigate the extent to which CPs are linked to current usage and disposal practices, and 3) to evaluate human health risk from exposure to CPs in the atmosphere. This study will contribute to nationwide data, inform risk management strategies, and support international cooperation and agreements on the use and regulation of CPs.

2. Materials and methods

2.1. Sample collection

Air samples were collected from Accra and Kumasi of Ghana (Fig. 1), the two largest cities in Ghana, which are known for their comparable business and economic development. The sampling sites were categorized into four types: e-waste, commercial, urban, and control/background areas. The e-waste sites included Agbogbloshie 1 through Agbogbloshie 5, K-Landfill, K-Airport, K-Abattoir (burning area), and K-Abattoir (gas area). These locations were chosen because five of them are e-waste sites, while the others engage in waste treatment activities such as open burning, dismantling, or compacting plastic products containing CPs. The commercial areas, Palladium and K-Asawase, contain stores that sell lubricants, adhesives, and hardware equipment. The urban areas—Spintex, CSIR, Korle-Bu, Adabraka, K-Atonsu, K-Zoomlion, and K-KNUST—are within the city but lack e-waste activities and the commercial sale of lubricants, adhesives, sealants, and paints

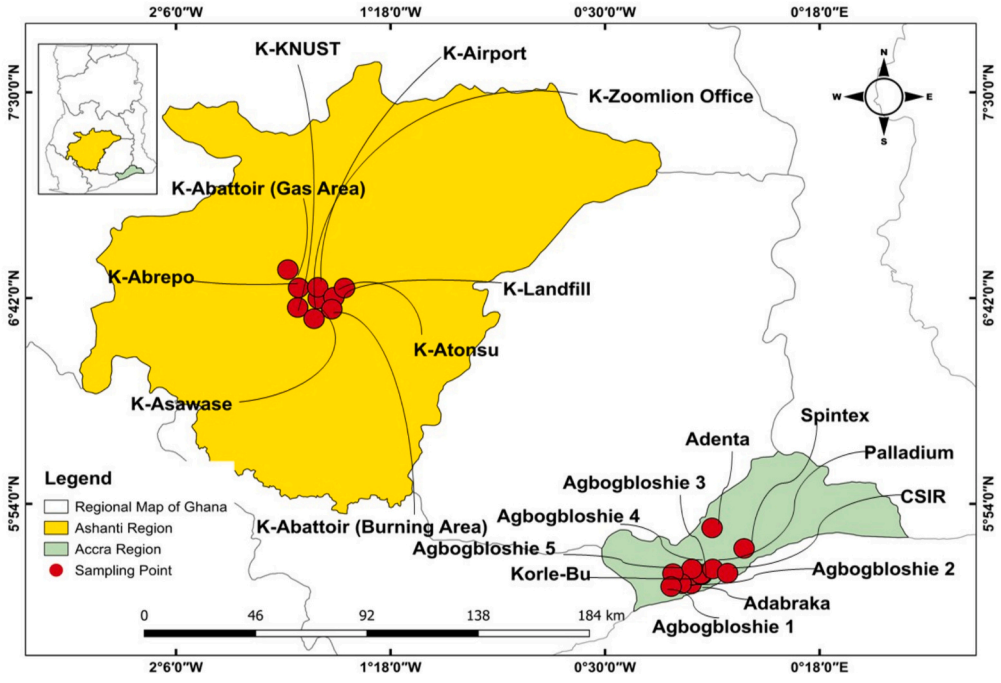

Fig. 1. Map of Ghana showing the sampling locations in the southern and middle belts.

containing CPs. The control/background sites, Adenta and K-Abrepo, are new settlements located far from any activities that could cause air pollution.

Polyurethane foam-passive air samplers (PUF-PAS) were mounted in locations where they would be secured, safe, and not stolen so as not to disrupt the monitoring process. Air samples were collected continuously for three months during each sampling cycle. A total of 60 samples were collected for three sampling cycles in 2020, covering both the wet and dry seasons. All sampling sites were in an open space without any obstructions; samples were placed at an approximate height of 3 m. Details about sampling information are given in Table S1. For seasonal variation, samples from April to November constituted the rainy/wet season and from December to March, the dry season.

2.2. Chemicals and reagents

Commercial standards of mixtures (chlorine contents: 51.5 %, 55.5 %, and 63 %) and MCCPs mixtures (chlorine contents: 42 %, 52 %, and 57 %) were purchased from Ehrenstorfer GmbH (Augsburg, Germany). Recovery surrogate $^{13}C_{10}$-trans-chlordane was purchased from Cambridge Isotope Laboratories (Andover, USA). Internal standard ^{13}C-Mirex was purchased from Ehrenstorfer GmbH (Augsburg, Germany). All reagents for pretreatment, including dichloromethane (DCM), n-hexane (HEX), and acetone (ACE) were high-performance liquid chromatography grade and purchased from Oceanpak (Sweden) or Fisher Scientific (Hanover Park, USA). Silica gel (63–100 μm), alumina (150 mesh), and anhydrous sodium sulfate were purchased from Merck (Whitehouse Station, USA) and preheated overnight at 450 °C to remove organic matter. Neutral silica gel and alumina were deactivated with 3 % (*w*/w) ultrapure water, while acid silica was deactivated with 50 % (w/w) sulfuric acid.

2.3. Sample preparation

In brief, $^{13}C_{10}$-trans-chlordane was added to each PUF disk before Soxhlet extraction with DCM for 24 h. The extracts were concentrated via rotary evaporation and solvent-exchanged into hexane with a reduced volume of 0.5–1 mL. The extracts were cleaned by a multilayer acidified silica gel column constituted by neutral alumina (3 cm), acid silica gel (3 cm), and anhydrous sodium sulfate (1 cm) from bottom to top. The eluent solvent was then concentrated into 50 μL by a gentle nitrogen blow. Before instrumental analysis, 10 ng of ^{13}C-Mirex was added to each sample as an internal standard.

2.4. Instrumental analysis

Instrumental analysis was adopted from a previous study utilizing a comprehensive two-dimensional gas chromatography- tandem mass spectrometry (GC × GC–MS/MS). The GC × GC–MS/MS analyses were performed using an Agilent 8890 GC instrument (Agilent Technologies, Santa Clara, CA, USA) equipped with an SSM1810 solid state modulator (J&X Tech., Shanghai, CN), and linked to an Agilent 7000D mass spectrometer detector. A negative chemical ion source in selected ion monitoring mode was operated for the measurement of 24 SCCP congener groups and 24 MCCP congener groups listed in Table S2. The ion source and transfer line temperatures were set as 200 and 280 °C, respectively. A DB-5 ms column (30 m × 0.25 mm × 0.25 μm; Agilent Technologies) and a DB-17 ms column (1 m × 0.25 mm × 0.25 μm) were respectively selected as the first- and second-dimension column for GC separation. Helium was used as carrier gas with a constant flow rate of 1.2 mL/min. A 2 μL sample aliquot was injected in splitless mode with an injection port temperature of 280 °C. The GC oven temperature was set at 100 °C (held for 1 min), then increased at 30 °C/min to 160 °C (held for 5 min), then at 1.5 °C/min to 282 °C, and held for 2 min. The modulation period was 12 s. Canvas Panel v2.0 was used for data processing (J&X Tech., Shanghai, CN). The chromatogram of standards used in this study is presented in Fig. S4.

2.5. Quality control and quality assurance

All glassware was cleaned with ultrapure water and baked at 450° C for 6 h prior to use. PUF disks were precleaned by extraction with methanol, ACE, and DCM for a duration of 24 h each before use. Chambers were rinsed with methanol and sealed with clean foil. Seventeen field blanks were collected to investigate the potential contamination during operation, transport, and storage. For monitoring the potential contamination during pretreatment, procedural blanks (n = 7) were performed in each batch of 11 samples. The method detection limits (MDLs) for C9-CP, SCCPs, and MCCPs were 0.012, 0.110 and 0.146 ng/m^3, respectively. The recoveries for all the samples averaged at 127.4 %.

2.6. Cancer and non-cancer risk assessment of CPs

To assess the cancer risk assessment of CPs in adults, the following equations were used which are from the (USEPA, 2005, 2015).

$$\text{Exposure Concentration (EC)} = \frac{CA \times ET \times ED}{AT} \quad (1)$$

$$\text{Cancer Risk} = EC \times IUR \quad (2)$$

where EC = exposure concentration (ng/m^3)

CA = concentration of pollutants in air (ng/m^3)

ET = exposure time, refers to the per capita outdoor exposure time of adults in Ghana, 8 h, 480 min/day

EF = exposure frequency, 365 day/year

ED = exposure time, refers to the per capita life expectancy in Ghana, 63 years

AT = averaging time, that is life expectancy, 552,000 h

IUR = inhalation unit risk for CPs, 2.5 E-05 (μg/m^3)$^{-1}$

The per capita life expectancy and per capita outdoor exposure were obtained from (Gamey, 2003; Mohammed et al., 2023; WHO, 2018).

The non-cancer risk assessment was assessed using the estimated daily intake (EDI, ng/kg/day), hazard quotient (HQ, unitless), and margin of exposure (MOE, unitless). The calculation of EDI, HQ, and MOE were performed individually for each sampling site according to Eq. (3), Eq. (4), and Eq. (5), respectively.

$$\text{EDI} = \frac{C \times IR \times T}{BW} \quad (3)$$

$$\text{HQ} = \frac{EDI}{TDI} \quad (4)$$

$$\text{MOE} = \frac{NOAEL}{EDI} \quad (5)$$

C is the concentration of CPs in air (ng/m^3). The inhalation rate (IR, m^3 /day) and body weight (BW, kg) were obtained from the USEPA guidelines (USEPA, 2005, 2015). The average exposure time per day (T, hour/day) was presumed to be 12 h. The total daily intakes (TDIs) of SCCPs and MCCPs, for non-neoplastic effects were all 100 μg/kg/day (UNEP, 1996). The non-observed adverse-effect level (NOAEL) of SCCPs and MCCPs were 10 and 23 mg/kg/day, respectively (UNEP, 1996; USEPA, 2005, 2015).

3. Results and discussion

3.1. Profiles of CPs in air

Table 1 provides descriptive statistics for CPs in the atmosphere of Ghana. With a mean concentration of 26.0 ± 40.1 ng/m^3, the concentration of MCCPs ranged from 1.78 to 240 ng/m^3. The concentration of

Table 1
Concentrations of CPs in PUF disks in the atmosphere of Ghana.

CPs	Concentration (ng/m^3)			
	Median	Mean	Standard deviation	Range
∑C9-CPs	0.350	0.544	0.524	0.091–2.14
∑SCCPs	1.93	3.48	3.99	0.05–15.2
∑MCCPs	12.0	26.0	40.1	1.78–240

SCCPs varied between 0.05 and 15.2 ng/m^3, with a mean of 3.48 ± 3.99 ng/m^3. The concentrations of C9-CP ranged from 0.091 to 2.14 ng/m^3, with a mean of 0.544 ± 0.524 ng/m^3. E-waste was an important contributor to CPs, responsible for 34.2 % and 32.2 % of the emissions related to SCCPs and MCCPs, respectively (Text S1 of supporting material).

Air concentrations of CPs in other studies are compiled in Table S3 for comparison. The concentrations of CPs in the air in the studied region were comparable to or even higher than CP concentrations in countries with a high degree of industrialization. For example, the concentration of SCCPs (0.05–15.2 ng/m^3) in this study was comparable to that of SCCP from residential areas in Canada (<MDL – 17.6 ng/m^3) (Niu et al., 2021). It was, however, lower than concentrations reported in studies from China (3.55–359 ng/m^3) (Li et al., 2021). The range of air concentrations of MCCPs (1.78–240 ng/m^3) in this study were far higher than MCCPs (<0.4–35 ng/m^3) in Dar es Salaam, Tanzania (Nipen et al., 2022) but lower than studies from China (1.95–59.8 ng/m^3) (Li et al., 2021). C_9-CPs from this study (0.091–2.14 ng/m^3) were lower than studies from Australia (<MDL – 30 ng/m^3) (van Mourik et al., 2020), China (14.0–127 ng/m^3) (Xia et al., 2019) and Norway (0.37–11 ng/m^3) (Yuan et al., 2021).

The congener group abundance profiles in the air for SCCPs and MCCPs are represented in Fig. S1 and Fig. S2, respectively. For the SCCPs, the dominant alkane-chain length group was C_{10} (ranging from 16.2 % to 55.1 %, mean 34.9 %), followed by C_{11} (ranging from 13 % to 30.1 %, mean 22.9 %). The next was C_{12} (ranging from 10.4 % to 45.3 %, mean 21.6 %), and finally the least was C_{13} (ranging from 10.1 % to 37.8 %, mean 20.3 %). This pattern was similar to those found in particulate matter (Brits et al., 2020; Huang et al., 2017; Liu et al., 2020), but different from those reported in sewage sludge, particulate matter, and commercial mixtures (Brandsma et al., 2017; Huang et al., 2023a; Li et al., 2018). Shorter carbon chain CPs with lesser degrees of chlorination tend to partition to the gas phase, while those congeners with larger K_{OA} and lower V_P values tend to partition to particle (South et al., 2022). For MCCPs, the dominant alkane-chain length group was C_{14} (ranging from 23.7 % to 58.2 %, mean 42.9 %), C_{15} (ranging from 15.9 % to 31.5 %, mean 22.3 %), C_{17} (ranging from 10.7 % to 36.0 %, mean 20.9 %) and lastly C_{16} (ranging from 8.47 % to 20.9 %, mean 13.7 %). This was like air particle samples from other studies (Huang et al., 2017; Huang et al., 2023a; Li et al., 2021; Li et al., 2018; Liu et al., 2020; Zhuo et al., 2019). A study conducted in China found that C_{10} was the dominant alkane-chain length in SCCPs and C_{14} was the dominant alkane-chain length for MCCPs, which was similar to that of this study (He et al., 2023).

With regard to the chlorine pattern for SCCPs, the order was Cl_7 > Cl_8 > Cl_6 > Cl_{10} > Cl_9 > Cl_5. The dominant congener was Cl_7 (ranging from 9.62 % to 52.3 %, mean 32.4 %), followed by Cl_8 (8.33 % - 48.7 %, mean 20.1 %), Cl_6 (6.32 % - 40.1 %, mean 17.2 %), Cl_{10} (3.52 % - 35.1 %, mean 11.2 %), Cl_9 (4.95 % - 17.2 %, mean 9.72 %) and Cl_5 (1.25 % - 20.5 %, mean 9.30 %). For the MCCPs, the order was Cl_8 > Cl_9 > Cl_7 > Cl_{10} > Cl_6 > Cl_5. The most dominant chlorine was Cl_8 (ranging from 14.4 % - 39.9 %, mean 25.1 %), followed by Cl_9 (14.1 % - 28.0 %, mean 20.7 %), Cl_7 (12.7 % - 25.4 %, mean 19.6 %), Cl_{10} (4.01 % - 27.2 %, mean 14.8 %), Cl_6 (6.16 % - 25.5 %, mean 12.8 %) and Cl_5 (2.73 % - 14.8 %, mean 6.98 %). The observation of a Cl_{6-9} dominant chlorine pattern in this study was consistent with the composition profiles of commercial mixtures CP-42 and CP-52 (Li et al., 2018).

3.2. Seasonal variation of CPs in the air

In the dry season in Accra, the mean concentrations of ∑C9-CPs, ∑SCCPs, and ∑MCCPs were 1.03 ± 0.686, 8.01 ± 4.72, and 36.6 ± 49.9 ng/m^3, respectively, while in the wet season they were 1.28 ± 0.854, 7.85 ± 7.06, and 68.4 ± 64.9 ng/m^3. During the dry season in Kumasi, the mean concentrations of ∑C9-CPs, ∑SCCPs, and ∑MCCPs were 0.294 ± 0.115, 1.75 ± 0.673, and 21.9 ± 24.2 ng/m^3, respectively; during the wet season, they were 0.514 ± 0.263, 2.08 ± 1.39, and 23.1 ± 19.7 ng/m^3, as shown in Fig. 2. In this study, the ∑SCCP concentration during the dry season ranged from 1.09 to 15.2 ng/m^3, with an average of 5.19 ± 4.71 ng/m^3. During the wet season, concentrations ranged from 0.55 to 23.9 ng/m^3, with a mean of 5.25 ± 5.98 ng/m^3. These levels were significantly lower than those reported in Jinan, China, where concentrations ranged from 21.1 to 69.9 ng/m^3 with an average of 37.7 ng/m^3 in spring, 9.80 to 45.3 ng/m^3 with a mean of 29.7 ng/m^3 in summer, 10.1 to 46.4 ng/m^3 with a mean of 32.8 ng/m^3 in autumn, and 27.0 to 105 ng/m^3 with an average of 54.8 ng/m^3 in winter (Li et al., 2019). This seasonal pattern – winter > autumn > spring > summer – was observed in studies conducted in Dalian, China (Zhu et al., 2017).

During the wet season, the commercial site had the highest concentration of both C9-CPs and SCCPs (Table S4). The e-waste site had the highest concentration of MCCPs during the wet season (240 ng/m^3). The study found that the concentrations of SCCPs and MCCPs during the wet season followed a similar seasonal trend to previous research (Nipen et al., 2022; Niu et al., 2021), where SCCPs were highest in the colder and drier months. In contrast, concentrations were higher in the warm summer months in other investigations (Gillett et al., 2016; Huang et al., 2023a; Niu et al., 2020; Wang et al., 2012; Wang et al., 2013). A study also reported the mean concentration of ΣSCCPs from Beijing was 5.2 ng/m^3 in winter, whereas in summer the value was 11 ng/m^3 (Wang et al., 2012). Another study reported that the mean concentration of ∑MCCPs was 20.8 ng/m^3 in summer and 6.26 ng/m^3 in winter, which indicated that temperature affected the concentration of CPs. Temperature can be a factor driving seasonal variations. Higher temperatures in summer could enhance the volatilization of CPs from products and other environmental compartments to air leading to higher concentrations (Wang et al., 2012; Wania and Mackay, 1996). However, this was not the case in our study given the relatively low annual temperature fluctuations. Temperature and rainfall data were obtained from the Ghana Meteorological Agency (GMA, 2021). Precipitation showed no statistically significant relationship with CPs in the atmosphere of Ghana (Table S5). A further plot of CP concentration against rainfall also showed an inverse relationship. There was no correlation between CP concentrations and rainfall, as seen in Fig. S3.

3.3. Spatial variation of CPs in the air

Fig. 3 illustrates that the overall ∑MCCPs outnumbered the total ∑SCCPs and ∑C9-CPs across all site types. The e-waste sites had the highest concentrations of ∑C9-CPs and ∑MCCPs, while the commercial site had the highest ∑SCCPs (Table S4). C9-CPs at the e-waste site ranged from 0.14 to 2.08 ng/m^3, with an average of 0.99 ± 0.71 ng/m^3. SCCPs were between 1.09 and 15.9 ng/m^3, with a mean of 6.65 ± 4.99 ng/m^3, while MCCPs ranged from 2.34 to 240 ng/m^3, with an average of 59.1 ± 62.9 ng/m^3. At the commercial site, C9-CPs varied from 0.25 to 3.34 ng/m^3, with an average of 1.36 ± 1.39 ng/m^3. SCCPs ranged from 0.55 to 23.9 ng/m^3, with a mean of 10.4 ± 11.3 ng/m^3, and MCCPs ranged from 8.58 to 60.4 ng/m^3, with an average of 26.6 ± 23.5 ng/m^3.

In the urban area, C9-CPs ranged from 0.18 to 1.14 ng/m^3, with an average of 0.47 ± 0.25 ng/m^3. SCCPs varied from 0.72 to 5.75 ng/m^3, with a mean of 2.73 ± 1.63 ng/m^3, while MCCPs ranged from 6.50 to 85.5 ng/m^3, with an average of 20.1 ± 20.6 ng/m^3. At the control site, C9-CPs ranged from 0.48 to 0.98 ng/m^3, with an average of 0.67 ± 0.23 ng/m^3. SCCPs ranged from 1.86 to 2.86 ng/m^3, with a mean of 2.37 ±

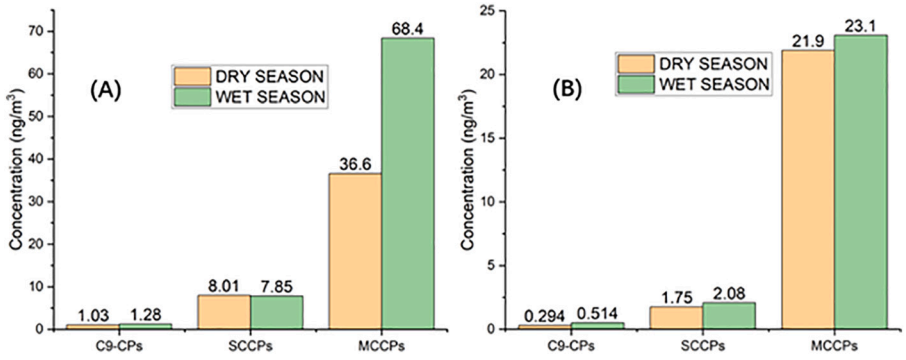

Fig. 2. Seasonal variation of mean CP levels in (A) Accra and (B) Kumasi of Ghana.

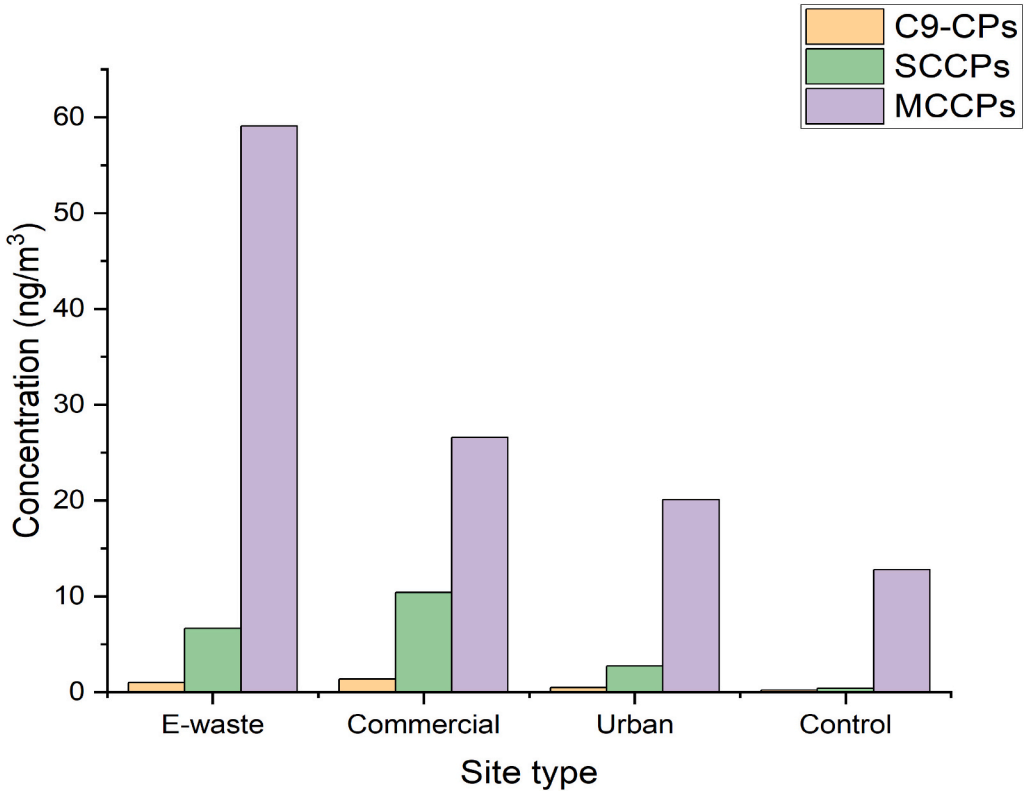

Fig. 3. Measured total ∑CP concentrations for 3 sampling cycles in Ghana.

0.41 ng/m^3, while MCCPs varied from 7.75 to 37.8 ng/m^3, with an average of 24.4 ± 12.8 ng/m^3. Notably, C9-CP and SCCP concentrations at the commercial sites were higher than those at the e-waste site. This might be because oil paints, adhesives, sealants, and metalworking fluids are sold at the commercial site, where CPs are released into the environment. At the commercial site, the C9-CPs and SCCPs were higher than those of the e-waste site.

A higher MCCPs/SCCPs ratio reflects an increase in the usage of MCCP products in the region (Zhuo et al., 2019). The ratio of MCCPs/SCCPs ranged from 2.17 to 17.4 with a mean of 8.86. The highest MCCPs/SCCPs ratio was at the e-waste site and the lowest at the commercial site. The ratios in this study were higher than those in Tanzania (Nipen et al., 2022), where the MCCPs/SCCPs ratio at the e-waste site was 0.6. In contrast, this study found a ratio of 18.1 at the e-waste site. At their dumpsite, the ratio was 0.55 while in our study it was 17.4 at our landfill site. The mean MCCPs/SCCPs ratio in our study (8.86) was far >1, indicating the usage of more MCCP products in this region. For example, China has recently been trying to reduce the content of short-chain components in industrial products (Chen et al., 2011). This is supported by the significant increase in the ratio of MCCPs/SCCPs in sediment, such as from Pearl River Estuary (Chen et al., 2011) and Laizhou Bay (Pan et al., 2018).

3.4. Risk assessment of CPs

Since CPs in the atmosphere have the potential to cause cancer in people, the US EPA technique was used to assess the risk of cancer (USEPA, 2015). The lifetime carcinogenic risk (ILCRs) less than or equal to 10^{-6} are considered to be basically negligible lifetime cancer risk; ILCRs between 10^{-6} and 10^{-4} indicate low risk; ILCRs $>10^{-4}$ indicate high potential health risks (Ailijiang et al., 2022). The US EPA has guidelines that state that a cancer risk of 10^{-6}, or one in a million probability of developing another human cancer over the course of a 70-year lifetime, is tolerable or insignificant, and that a risk beyond 10^{-4} is deemed concerning and calls for taking action to lower the health hazards (Kang et al., 2019). The data presented in Table S6 indicates that

the cancer risk for C9-CPs, SCCPs, and MCCPs was 4.55×10^{-8} to 1.07×10^{-6}, 2.50×10^{-8} to 7.60×10^{-6}, and 8.90×10^{-7} to 1.20×10^{-4} respectively. The cancer risk for C9-CPs and SCCPs were found to be low risk, and MCCPs exceeded the acceptable risk range indicating high potential carcinogenic risk. We assessed the adult EDIs, HQs, and MOEs for non-cancer risk. Based on inhalation scenarios equivalent to 20 mg/day for adults, EDIs were computed with median concentrations for each homologous group (Harrad et al., 2016). Daily intakes were normalized to a body weight (bw) of 70 kg. EDIs for total CPs for adults were 56.7 ng/kg bw/day. The EDIs for $\sum$MCCPs were about six times that of the $\sum$SCCPs and thirty-four times higher than that of $\sum C_9$-CPs. The MOEs of SCCPs and MCCPs (Table S7) were all >1000 indicating that people in Accra and Kumasi may have no carcinogenic effect in terms of CPs. The hazard quotients (HQs) calculated were all <1 (Table S7), indicating no human health risk in association with CP inhalation.

In addition to inhalation, there are other exposure pathways that significantly contribute to the overall body burden of CPs. Consequently, it is essential to investigate these alternative exposure routes in the study area to gain a comprehensive understanding of the exposure risks. Human exposure to chlorinated paraffins is a complex and multifaceted issue with potential health implications. Continued research, regulatory efforts, and public awareness initiatives are essential for minimizing exposure risks and safeguarding human health.

3.5. Uncertainties and limitations

A common challenge with PUF-PAS is that it captures both particle and gas phases simultaneously (Herkert et al., 2018; Zhao et al., 2020), but this does not affect our findings in this study. Our samples were collected covering three sampling cycles: one dry season and two wet seasons. This imbalance could potentially make seasonal comparison skewed. In future studies, we aim to include an equal number of cycles for each season to enable more reliable comparisons.

4. Conclusions

Our study presented the levels, potential sources and human health risks of CPs in the atmosphere of Ghana. MCCPs were the predominant CP group, exhibiting values that were 7.5 times greater than those of SCCPs and 48 times greater than those of C9-CPs. MCCPs were dominant in both the dry and wet seasons. Temperature did not influence the concentration of CPs due to the relatively minor annual temperature variations in tropical regions. E-waste site was a significant source of CP emissions, responsible for 34.2 % and 32.2 % of the emissions related to SCCPs and MCCPs, respectively. Adults are not in danger of any health problems from CPs in Ghana's atmosphere, according to health risk assessment for non-cancer diseases. There is a low carcinogenic health risk for C9-CPs and SCCPs, but there was high potential carcinogenic risk for MCCPs. The prevalence of MCCPs may suggest that global SCCP usage has been somewhat reduced by international limits and UN POPs regulations.

Better waste management should be implemented at the regional level to address this issue. Since the problem is common to other developing countries, it should also be addressed in international regulatory activities relating to the trade of products, used products, and waste containing these substances. Because of the burning of e-waste, e-waste contributed the most to the overall CP pollution. The Ghana Ports and Harbor Authority (GPHA) and the Environmental Protection Agency (EPA) will use the findings of this study to develop policies and procedures to control the usage and disposal of e-waste across the nation. By ensuring adherence to health and environmental regulations, these measures will safeguard the general public as well as the environment. Additionally, this research will facillitate global collaboration and agreements regarding the use and regulation of chlorinated paraffins. The exchange of knowledge enables nations to work together in addressing global environmental and health challenges associated with these chemicals.

CRediT authorship contribution statement

William Ekow Arko: Writing – original draft, Investigation. **Shizhen Zhao:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization. **Jianchu Ma:** Methodology, Investigation. **Lele Tian:** Investigation. **Kwadwo Ansong Asante:** Writing – review & editing, Supervision. **Daniel Kwaku Amoah:** Investigation. **Shihua Qi:** Supervision, Resources. **Gan Zhang:** Supervision, Resources, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgements

This work is supported by the Alliance of International Science Organizations (ANSO-CR-KP-2021-05), the National Natural Science Foundation of China (42107120 and 42030715), the Guangdong Basic and Applied Basic Research Foundation (2023B1515020067 and 2023B0303000007), the Youth Innovation Promotion Association of CAS (2022359), the Tuguangchi Award for Excellent Young Scholar GIG (TGC202204), and the State Key Laboratory of Organic Geochemistry, GIGCAS (SKLOG2024-02).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.176252.

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