Spatial trends of chlorinated paraffins and dechloranes in air and soil in a tropical urban, suburban, and rural environment☆

Maja Nipen [a,b,*], Rolf David Vogt [a], Pernilla Bohlin-Nizzetto [b], Katrine Borgå [c], Eliezer Brown Mwakalapa [d], Anders Røsrud Borgen [b], Susanne Jøntvedt Jørgensen [a], Samwel Moses Ntapanta [e], Aviti John Mmochi [f], Martin Schlabach [b], Knut Breivik [a,b]

[a] Centre for Biogeochemistry in the Anthropocene, Department of Chemistry, University of Oslo, P.O. Box 1033, 0315 Oslo, Norway
[b] NILU-Norwegian Institute for Air Research, P.O. Box 100, 2027 Kjeller, Norway
[c] Centre for Biogeochemistry in the Anthropocene, Department of Biosciences, University of Oslo, P.O. Box 1066, 0316 Oslo, Norway
[d] Department of Natural Sciences, Mbeya University of Science and Technology, Mbeya, Tanzania
[e] Department of Social Anthropology, University of Oslo, P.O. Box 1091, 0317 Oslo, Norway
[f] Institute for Marine Science, University of Dar Es Salaam, Zanzibar, Tanzania

ABSTRACT

There are large knowledge gaps concerning environmental levels and fate of many organic pollutants, particularly for chemicals of emerging concern in tropical regions of the Global South. In this study, we investigated the levels of chlorinated paraffins (CPs) and dechloranes in air and soil in rural, suburban, and urban regions in and around Dar es Salaam, Tanzania. Samples were also collected near the city's main municipal waste dumpsite and an electronic waste (e-waste) handling facility. In passive air samples, short chain CPs (SCCPs) dominated, with an average estimated concentration of 22 ng/m³, while medium chain CPs (MCCPs) had an average estimated concentration of 9 ng/m³. The average estimated air concentration of $\sum$dechloranes (Dechlorane Plus (DP) + Dechlorane 602 + Dechlorane 603) was three to four orders of magnitudes lower, 2 pg/m³. In soil samples, MCCPs dominated with an average concentration of 640 ng/g dw, followed by SCCPs with an average concentration of 330 ng/g dw, and $\sum$dechloranes with an average concentration of 0.9 ng/g dw. In both air and soil, DP was the dominating dechlorane compound. Urban pulses were observed for CPs and dechloranes in air and soil. CPs were in addition found in elevated levels at the municipal waste dumpsite and the e-waste handling facility, while DPs were found in elevated levels at the e-waste handling facility. This suggests that waste handling sites represent important emission sources for these pollutants. Investigations into seasonal trends and environmental fate of CPs and dechloranes showed that monsoonal rain patterns play a major role in governing air concentrations and mobility, particularly for the less volatile MCCPs and dechloranes. This study is the first to report levels of CPs in air from sub-Saharan Africa, and DP, Dechlorane 602, and Dechlorane 603 in soil from sub-Saharan Africa.

1. Introduction

A number of organic chemicals and chemical groups have been regulated as persistent organic pollutants (POPs) because they represent a risk for environmental and human health (UNEP, 2017). At the same time, there is considerable scientific and regulatory interest in substances which may have similar hazardous properties as the legacy POPs. Examples include chlorinated paraffins (CPs) and dechloranes.

Short chain CPs (SCCPs, C_{10}–C_{13}) are subject to international regulation under the Stockholm Convention on POPs and the Aarhus Protocol of the Convention on Long-Range Transboundary Air Pollution, while medium chained CPs (MCCPs, C_{14}–C_{17}) remain unregulated at the international level (Glüge et al., 2018; UNECE, 2010; UNEP, 2017). The latter also applies to Dechlorane Plus (DP) which is proposed for listing under the Stockholm Convention (UNEP, 2019). While legacy POPs have been extensively studied in terms of their environmental levels and fate, our

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understanding of the environmental occurrence and behaviour of CPs and dechloranes remains fragmented and incomplete. There is therefore considerable interest and need for further research into these two substance groups to support chemical management strategies.

CPs and dechloranes are added as flame retardants or plasticizers in a range of consumer products. More than a million tonnes of CPs are produced annually, mainly in China, and used in plastics, paints, sealants, rubber, and leather products, as well as in metalworking applications (Glüge et al., 2016). DP is used in electronics and plastic roofing materials (Hoh et al., 2006; UNEP, 2019). Production volumes of DP are lower compared to CPs, at 750–6000 tonnes annually (Hansen et al., 2020), while use categories and production volumes are less well described for Dechlorane 602 (Dec 602) and Dechlorane 603 (Dec 603).

Most studies on sources and fate of chemicals of emerging concern (CECs) like CPs and dechloranes have been conducted in developed economies of the Global North, where effective regulatory efforts to some extent have reduced production, use and trade of these chemicals. Countries in the Global South may on the other hand be experiencing increasing environmental burdens of CPs and dechloranes due to a range of socioeconomic factors. These factors include increased consumption and limited waste handling systems, limited capacity to enforce environmental regulations, as well as being on the receiving end of transboundary trade of used products and waste (Abbasi et al., 2019; Breivik et al., 2011; Li and Wania, 2016; Robinson, 2009). In addition, differences in climate between these regions will likely lead to differences in environmental fate of CPs and dechloranes. For example, higher temperatures in tropical regions increase the volatility of both substance groups (Wania and Mackay, 1996) and cause more rapid environmental degradation in various environmental compartments, including air and soil (Webster et al., 1998).

Tanzania is a sub-Saharan African country with a tropical climate, and an emerging economy with an annual average increase in gross domestic product of 6 % since the turn of the century (WorldBank, 2021). With a population of 6.4 million, Tanzania's largest city Dar es Salaam suffers from insufficient waste handling systems, notably including electronic waste (e-waste) (Mahenge et al., 2018; Yhdego, 2017). Dar es Salaam is not known as a major recipient in the illegal e-waste trade, but the amount of e-waste generated is growing (NBS, 2019). These factors combined suggest that the Tanzanian environment, particularly urban Dar es Salaam, may be experiencing increasing burdens of CPs and dechloranes. But to our knowledge, there are no previous studies reporting concentrations of CPs and dechloranes in the Tanzanian environment in peer reviewed scientific literature. However, the NGO network IPEN recently reported high concentrations of SCCPs in free-range chicken eggs collected near a municipal waste dumpsite in Dar es Salaam (Petrlik et al., 2020). This shows that these CECs are present in consumer products and waste in the region. Elsewhere, studies have shown elevated levels of CPs and DP near e-waste recycling sites (Chen et al., 2011; Möckel et al., 2020; Someya et al., 2016; Xu et al., 2019), and CPs near a municipal waste site (Möckel et al., 2020). In addition, elevated levels of both CPs and dechloranes have been found in urban environments (Barber et al., 2005; Saini et al., 2020).

The main objective of this study was to determine concentrations of CPs and dechloranes in the physical environment within and around Dar es Salaam, representing an urban tropical environment. We selected ambient air and soil as sampling media as atmospheric inputs represents the key pathway of environmental contaminants into terrestrial ecosystems, including the terrestrial part of the human food-chain (McLachlan, 1995). By choosing air and soil we wanted to gain insights into contemporary emissions and historical accumulation of these two substance groups in the physical environment, respectively. Our aim was furthermore to study to what extent environmental burdens of CPs and dechloranes are associated with contemporary use and/or disposal, as well as to evaluate potential dispersion of these chemicals from assumed source areas. Additional motivations for this study were to rationalize the main factors governing the spatial distribution, air-soil exchange,

and environmental fate of CPs and dechloranes in a tropical urban region.

2. Materials and methods

2.1. Sample collection

2.1.1. Sampling locations

Dar es Salaam has an average temperature of $26°C$, with only minor temperature fluctuations throughout the year, February being the warmest month (average $29°C$) and July the coldest (average $24°C$). The annual precipitation of more than 1000 mm is monsoonal, with most rainfall in April (~250 mm/month) and least in June, July, August, and September (~30 mm/month).

Air and soil samples were collected from 19 locations along a transect covering 40 km southeast of Dar es Salaam, the city of Dar es Salaam, and 60 km northwest of the city (Fig. 1). This transect comprised different settlement regions, including densely populated and industrialized urban areas (U), suburban areas (S), and rural areas (R) with little anthropogenic activity. Air and soil samples were also collected from 9 locations along a shorter (12 km), waste-source transect (W) targeting specifically the city's main municipal waste dumpsite and an e-waste handling facility on the outskirts of Dar es Salaam. The e-waste facility is by Tanzanian standards a large-scale facility, mainly handling electronics from domestic use. Being part of the formalized sector, crude recycling techniques (e.g., cable burning, acid leaching) commonly practiced in some East Asian and West African countries (Alabi et al., 2012; Wong et al., 2007) are not known to be carried out at this facility.

2.1.2. Air samples

Air samples were collected using polyurethane foam (PUF) based passive samplers (Shoeib and Harner, 2002) of the type used in the MONET monitoring network (Klánová et al., 2009; Klánová et al., 2006). PUF disks and sampler metal components were pre-cleaned and stored dust-free before deployment, and PUF disks were also kept refrigerated. Air samplers were deployed at least 2 m above ground level for approximately two months, between February and April 2019.

In addition to investigating spatial trends, seasonal fluctuations in levels of CPs and dechloranes in air were also monitored for more than a year (January 2018 to April 2019) at location S-13. Samplers were deployed for two-to three-month periods, with a total of six samples being collected.

2.1.3. Soil samples

Achieving representability and comparability of soils sampled in areas affected by local anthropogenic activities is challenging. To promote representability, samples from each site were composed of three pooled subsamples, collected within a range of three to five square metres using a stainless-steel handheld corer to a depth of approximately 5 cm. Comparability was optimized by collecting samples below the canopies of mature trees. This minimizes the risk for collecting displaced soils, although it might lead to overall higher soil concentrations of CPs and dechloranes compared to soil in open areas due to the forest filter effect and higher throughfall and litterfall deposition (Horstmann and Mclachlan, 1998). At the e-waste facility, soil was collected from the courtyard used for storing electrical components. At the municipal dumpsite, soil was collected within ~30 m of the waste piles. Soil samples were collected in February 2019.

2.2. Sample preparation and analysis

Details on sample preparation and analysis are given in the SI and a summary is presented here. Soil samples (~7 g) were dried and homogenized with sodium sulphate in a mortar until a free-flowing powder was achieved. The air samples (PUF disks) and soil samples were
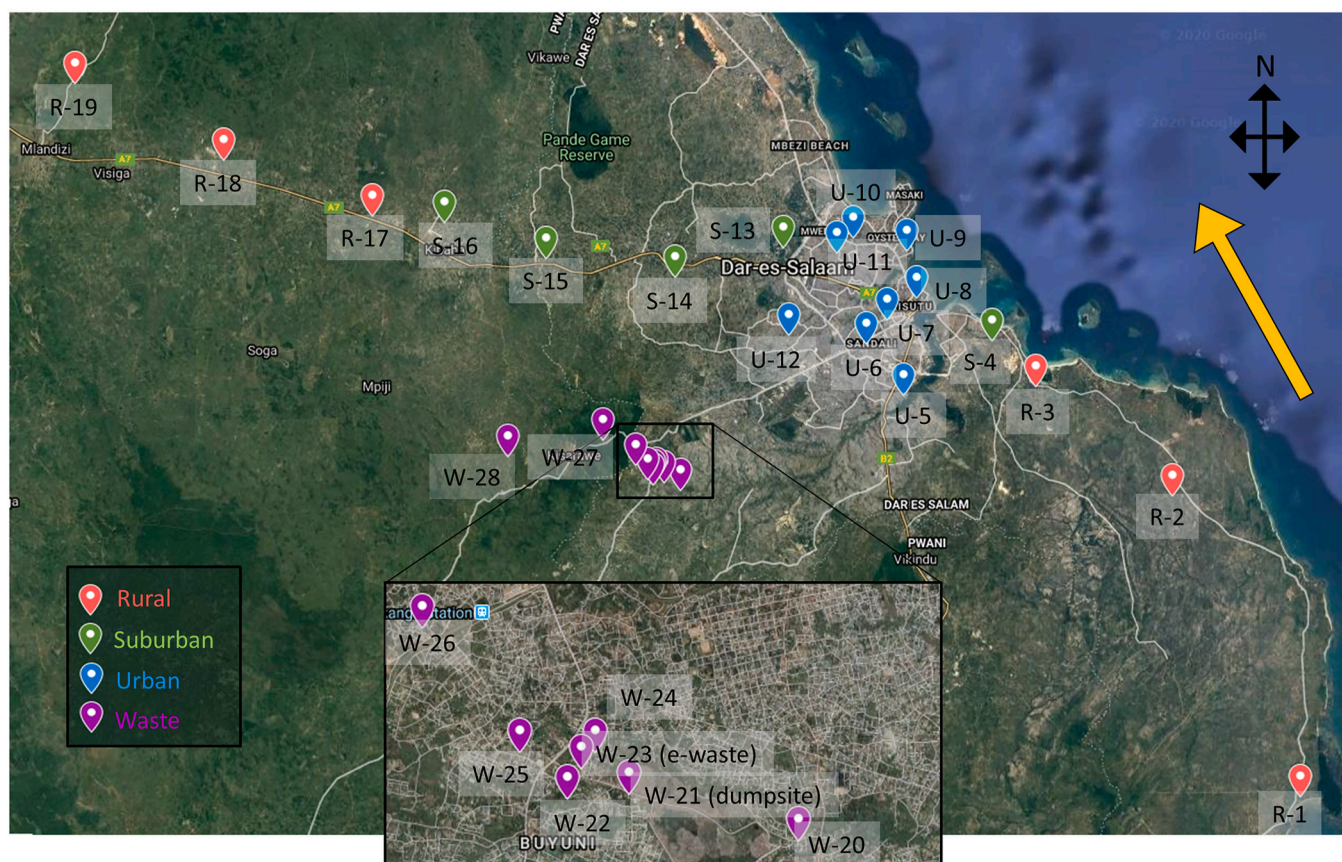
Fig. 1. Locations for air and soil sampling in Dar es Salaam and surrounding areas (R: Rural, S: Suburban, U: Urban, W: Waste). Predominant wind direction shown by arrow. Map from Google.com/maps.

extracted in acetone/n-hexane (1:1) using soxhlet extraction and accelerated solvent extraction, respectively, after the addition of internal standards (^{13}C labelled 1,5,5,6,6,10-hexachlorodecane, DP syn, and Dec 602). Extracts from both air and soil samples were volume reduced using a Turbovap system (Zymark) before being treated with concentrated sulphuric acid for the removal of lipids and polymeric impurities. Further, the extracts were cleaned using a dry packed column with 4 g activated silica covered by a 1 cm layer of sodium sulphate eluted with diethyl ether in n-hexane (1:9). Solvent was changed to isooctane, volume reduced to 100 μL, and 1,2,3,4-tetrachloronaphthalene was added as a recovery standard.

Extracts were analysed for SCCPs, MCCPs, DP (syn- and anti-isomers), Dec 602, and Dec 603 using gas chromatography quadrupole time-of-flight high resolution mass spectrometry (GC/Q-TOF) (Agilent, Santa Clara, USA) in electron capture negative ionization (ECNI) mode. A total of 34 SCCPs and 28 MCCPs congener groups (groups of CPs that share molecular formula) were identified and quantified in all samples. In addition, 14 very short chain CPs (vSCCPs, C_7–C_9) congener groups were identified in air samples, although due to lack of available quantification standards these were not quantified.

2.2.1. Quantification of CPs

CPs in industrial products and environmental samples potentially consist of thousands of structurally similar isomers, causing a level of complexity that makes quantification of CPs challenging (van Mourik et al., 2020a). In this study, a pattern deconvolution method developed by Bogdal et al. (2015) was used to quantify CPs. This method applies an algorithm to reconstruct the congener group patterns in the analysed samples using a linear combination of congener group patterns from multiple CPs quantification standards. SCCPs standards with 51 %, 55 %, and 63 % Cl (w/w), and single chain length standards of C_{10}, C_{11}, C_{12},

and C_{13} (50 % and 65 % Cl) were used, and for MCCPs, standards of 42 %, 52 %, and 57 % Cl, and single chain C_{14} with 52 % Cl were used. Close match between sample and quantification standard is important as instrument response factors are dependent on chlorination degree (Mézière et al., 2020). The deconvolution method accounts for this by including standards with different chlorination degrees. Match between sample pattern and reconstructed pattern was assessed using the coefficient of determination (R^2), where R^2 of above 0.5 was deemed acceptable. Average R^2 were 0.96 and 0.90 for SCCPs, and 0.99 and 0.76 for MCCPs for air and soil, respectively. Details are given in the SI.

2.2.2. Determining ambient air concentrations of CPs and dechloranes

Volume-based air concentrations of CPs were estimated using a sampling rate of 4.2 m^3/day (Li et al., 2012). For dechloranes, which to a large extent are sorbed to particles in air, a sampling rate of 2.3 m^3/day was applied (Drage et al., 2016). This reflects the lower uptake efficiency of particle-associated compounds compared to gas-phase compounds with the MONET type sampler (Markovic et al., 2015). We note that estimated volume-based air concentrations are associated with larger uncertainty compared to the sequestered amount per sample (Wania and Shunthirasingham, 2020). However, volume-based results are presented in this study to allow for comparisons with other studies.

2.3. Quality control/quality assurance

Method blanks (N = 12) and field blanks (N = 8) were analysed along with every batch of air and soil samples. The average blank levels compared to the levels in samples were 0.8, 3.6, 28, and 17 % for air samples, and 8.6, 2.7, 16, and 14 % in soil samples for SCCPs, MCCPs, DP syn, and DP anti respectively. All samples were blank corrected for CPs and DP content based on blanks prepared and analysed alongside each

batch of samples. For CPs, blank correction was performed using the integrated area for each individual congener group from the blank samples, which was subtracted from the corresponding integrated area for the samples. This procedure corrects the total CPs concentration but also acts to prevent the congener group patterns in samples from being skewed by blank contributions.

Method Limits of Detection (LODs) were set at three times the standard deviation of the blanks (normalized by the average sample amount/volume). No significant difference was seen between method blanks and field blanks ($p > 0.05$), so both were used in the determination of LODs. For air samples, the LODs were 0.2 and 0.4 ng/m^3 for SCCPs and MCCPs, and 0.4 and 0.6 pg/m^3 for DP syn and DP anti, respectively. The LODs for soil samples were 11 and 19 ng/g dw for SCCPs and MCCPs, and 0.01 and 0.02 ng/g dw for DP syn and DP anti, respectively. Dec 602 and Dec 603 were not detected in blanks, so instrument LODs were used (0.001 ng/g dw for soil samples, 0.007 pg/m^3 for air samples). LODs for CPs in air were high despite measures to prevent blank contamination, although this is not uncommon (van Mourik et al., 2020b). However, the high levels of CPs in exposed air samples in this study ensured that the high LODs only had a minor effect on the analytical data (Fig. S2). The much lower levels of DP in blanks for air had a larger effect on analytical data, as the DP levels in air were low (Fig. S4). Larger sample volumes would help address this issue. In addition, the use of controlled clean-room laboratories is recommendable for the analysis of CECs which may be present in indoor environments.

Air samplers were deployed in duplicate at three locations. These gave an average deviation in results of less than 20 % for CPs and dechloranes, while four soil samples analysed in duplicate gave an average deviation of less than 30 %. A spike recovery test using CPs standards was performed to assess differences in performance of the sample preparation method for individual congener groups. For total CPs concentrations, the test showed a recovery of 103 ± 3 % for SCCPs and 105 ± 10 % for MCCPs. Only minute differences could be seen in congener group profiles of the spiked samples having been through extraction and clean-up, and the congener group profiles of the applied CPs standards analysed directly ($R^2 = 0.998$). Recovery of CPs internal standards were 65 ± 14 and 73 ± 17 % for air and soil samples respectively, while recovery of Dechlorane internal standards were 67 ± 13 and 64 ± 13 % in air samples, and 61 ± 22 % in soil samples (Tables S5–S8). The quantification procedures for both CPs and dechloranes were based on ^{13}C labelled internal standards added before sample extraction and clean-up, and thus intrinsically corrects for recovery.

2.4. Mechanistic assessments of environmental processes and fate

Processes governing distribution and mobility of CPs and dechloranes in the environment were assessed using chemical partitioning space plots and mobility plots, as developed by Gouin et al. (2000) and Wong and Wania (2011). These plots visualize the degree to which individual CPs congener groups and dechloranes associate with the different soil compartments (pore air, pore water, and organic solids), and the relative importance of their mobility by volatilization, leaching, and erosion. The modelled distribution and mobility uses partitioning coefficients based on physicochemical property data from Glüge et al. (2013) for CPs, and Sverko et al. (2011) for dechloranes. CPs and dechloranes concentrations in air and soil were used to determine soil/air fugacity ratios (f_{soil}/f_{air}) to assess to which degree soils in the studied region serve as a temporary reservoir or secondary source. Fugacity calculations were done using equations from Mackay (2001), and parameters relevant for local conditions if available. Further details are provided in the SI.

2.5. Statistical analysis

Data treatment and statistical tests were performed using R studio 3.4.3 and Microsoft Excel. Summary statistics for compounds with detection between 40 % and 80 % were determined using the robust regression on order statistics method as described in Helsel (2011). The non-parametric Wilcoxon rank sum test was used for group comparisons, while correlation between compounds were determined using Spearman rank correlations. Principal component analysis (PCA) was used to investigate if there were systematic spatial differences in congener group profiles. Compounds with detection frequency <40 % were excluded from statistical analysis.

3. Results and discussion

3.1. CPs and dechloranes in air and soil

3.1.1. General overview

Descriptive statistics for CPs and dechloranes in air and soil are given in Table 1, while concentrations at individual locations are provided in Tables S5–S8. Median estimated concentrations of CPs and dechloranes in air had the following order: SCCPs > MCCPs ≫ DP > Dec 602. The MCCP/SCCP ratios in air ranged from 0.14 to 0.76, with a median value of 0.35. SCCPs were detected in all air samples collected, while MCCPs were detected in 93 % of air samples.

Although not quantified, vSCCPs were also detected in all air samples. C_7 CPs were only found in minor amounts, while C_8 and C_9 CPs combined comprised on average 9 % of total analysed CPs on an integrated signal basis. Their ubiquitous presence and relatively large contribution to total CPs underlines the importance of making vSCCPs quantification standards available, so their environmental presence can be quantified and reported along with other CPs.

In soil samples, median concentrations of CPs and dechloranes were ordered as follows: MCCPs > SCCPs ≫ DP > Dec 603 > Dec 602. Of the CPs, MCCPs dominated in soil, giving an MCCP/SCCP ratio above 1 (0.5–11) at most sites, with a median value of 3.1. Correlations between concentrations in air and soil were seen for SCCPs, MCCPs, DP, and Dec 602 (Spearman's rho: 0.45–0.71, $p < 0.05$) (Table S10).

Air and soil concentrations of CPs and dechloranes from other studies are compiled in Tables S11 and S12 for comparison. Our study is as far as we are aware the first to report CPs in air from sub-Saharan Africa. The concentrations of CPs in air and soil in the studied region were comparable or even higher than CPs concentrations in countries with a high degree of industrialization. For example, the concentrations of SCCPs in air at the highly industrialized Pearl River Delta in China (18 ng/m^3) were similar to our study (22 ng/m^3). The average soil concentrations of SCCPs and MCCPs (330 and 640 ng/g dw, respectively) were even an order of magnitude higher than the Chinese study (18 and 59 ng/g dw, respectively) (Wang et al., 2013). The range of air concentrations of SCCPs and MCCPs (0.3–63 and < 0.4–35 ng/m^3, respectively) were also comparable to urban areas in India and Pakistan (<LOD-47 and <LOD-38 ng/m^3, for SCCPs and MCCPs respectively) (Chaemfa et al., 2014). We caution that comparisons of CPs concentrations between studies are associated with significant uncertainties given analytical challenges and lack of certified reference materials (van Mourik et al., 2018).

Concentrations of dechloranes in air and soil were in the mid-to high range of concentrations reported in studies from elsewhere in the world (Table S11, Table S12). The Global Atmospheric Passive Sampling (GAPS) Megacities study (Saini et al., 2020) recently reported air concentrations of DP and Dec 602 in 19 large cities worldwide. In that study, Lagos, Nigeria, showed the highest DP air concentration of all megacities. The highest concentrations of DP and Dec 602 in Tanzanian air (20 and 0.2 pg/m^3, respectively) were comparable to the concentrations reported for Lagos (DP: 28 pg/m^3, Dec 602: 0.2 pg/m^3). Despite low detection frequency, the highest estimated concentrations of Dec 603 in air (0.2 pg/m^3) were an order of magnitude higher than in urban air in Spain (0.01 pg/m^3) (De la Torre et al., 2018). The highest Dec 603 concentration in soil (3 ng/g dw) were higher than any found in the

Table 1

Descriptive statistics and detection frequency of CPs and dechloranes in air and soil.

	CPs Air (ng/m³)		CPs Soil (ng/g dw)		Dechloranes air (pg/m³)			Dechloranes Soil (ng/g dw)		
	SCCPs	MCCPs	SCCPs	MCCPs	DP	Dec 602	Dec 603[a]	DP	Dec 602	Dec 603
Range	0.3–63	<0.4–35	<11–5300	<19–5100	<0.4–20	<0.007–0.2	<0.007–0.2	<0.01–6	<0.001-0.05	<0.001–3
Average	22	9	330	640	2	0.05	–	0.7	0.006	0.2
Median	13	5	51	170	0.7	0.03	–	0.2	0.003	0.009
Std. dev.	20	11	1000	1200	4	0.06	–	1	0.01	0.6
Det. freq.	100 %	93 %	89 %	86 %	57 %	46 %	14 %	89 %	71 %	61 %

[a] Detection frequency too low for determination of descriptive statistics.

literature. Concentrations of DP, Dec 602, and Dec 603 in soil has to our knowledge not previously been reported from sub-Saharan Africa. Neither CPs nor dechloranes are known to be produced in Tanzania (Glüge et al., 2016; Hansen et al., 2020). The levels of primary industrial use are expected to be low, although there are little data available to confirm this. The relatively high concentrations of CPs and dechloranes in air and soil in the study area despite lack of production may be linked to issues with the handling of municipal waste and e-waste.

### 3.1.2. Seasonal fluctuations of CPs and dechloranes in air

Monitoring of seasonal fluctuations of CPs and dechloranes in air at location S-13 showed that the highest concentrations occurred during colder, dry months (June–November). This contrasts with seasonal trends of CPs in air in temperate and sub-tropical regions of China and Australia, where concentrations were higher during warmer summer months (Gillett et al., 2017; Niu et al., 2020; Wang et al., 2012; Wang et al., 2013). This differs also from DP in air in temperate North America, which showed no seasonal trend (Shunthirasingham et al., 2018). While temperature may be a driver for seasonal trends of semi-volatile compounds in air (Wania and Mackay, 1996), this is not the case in our study given the relatively low annual temperature fluctuations and the inverse relationship with air concentrations of CPs and dechloranes. Amount of rainfall was investigated as an alternative explanatory factor. This was done by plotting air concentrations of SCCPs, MCCPs, and DP against cumulative rainfall during each sampling period. Dec 602 and Dec 603

were omitted as concentrations were near or below LOD. Rainfall data was gathered from the Tanzanian Meteorological Authority (TMA, 2021). Results showed negative liner association between SCCPs, MCCPs, and DP concentrations and amount of rainfall (Fig. S6), which was significant for MCCPs and DP ($p < 0.05$), but not for SCCPs ($p > 0.05$). This can be explained by a higher degree of washout of compounds associated with particles (e.g., DP and MCCPs) compared to gas-phase compounds (e.g., SCCPs) (Bidleman, 1988). A model and field-based study from India has shown comparable results for some legacy POPs, although in general, the effect of monsoonal rain patterns on both POPs and CECs in air remain understudied (Lammel et al., 2018). The observed seasonal fluctuations indicate that annual average levels of CPs and dechloranes in air may be higher than those reported in this study since the main sampling campaign for air samples (February to April) coincided with the rain-season. This is particularly relevant for the less volatile dechloranes, but potentially also for higher chlorinated, longer chain MCCPs congener groups, which to some extent may be sorbed to particles in air.

3.2. Spatial pattern and potential sources

3.2.1. Urban pulse and waste related sources for CPs

An urban pulse was seen for CPs in air and soil. Concentrations of SCCPs and MCCPs in air and MCCPs in soil were significantly higher in urban compared to rural locations ($p < 0.05$) (Fig. 2). For SCCPs in soil,
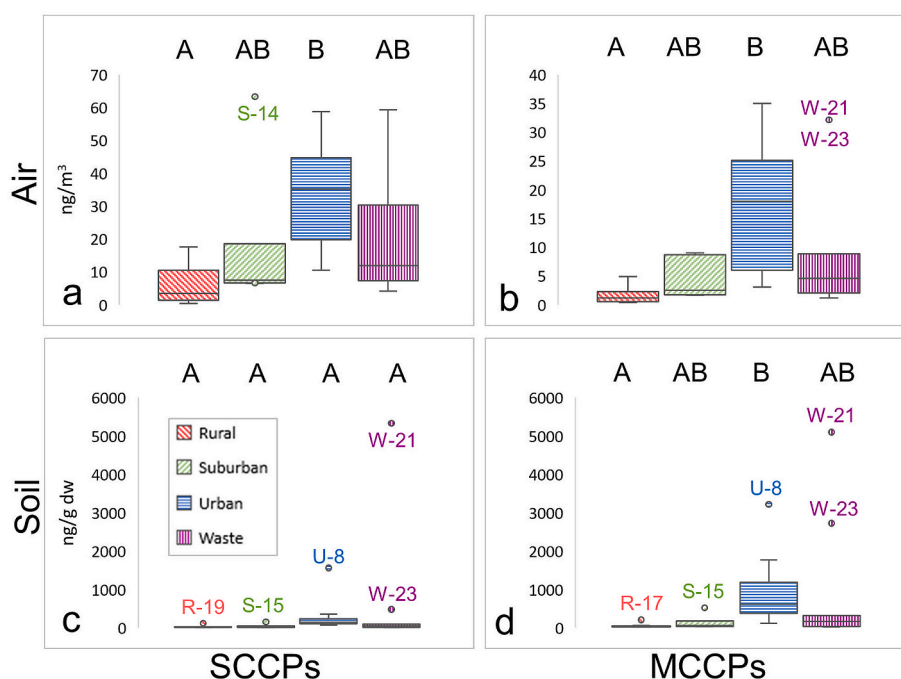
Fig. 2. Concentrations of SCCPs and MCCPs in air (top) and soil (lower) in different settlement categories. The median value is represented by the line inside the box, the upper and lower sections of the box show the upper and lower quartiles. Whiskers represent minimum and maximum values. Outliers are marked as points outside the whiskers. Groups not significantly different from each other (Wilcoxon rank sum test) share letter code (A, B).

there was no significant difference (p > 0.05), caused by one high concentration at a rural location (R-19, 120 ng/g). The corresponding air sample at R-19 did however not show elevated concentration of SCCPs (2 ng/m^3), indicating historic emissions at this location. The highest individual concentrations of CPs in air were found within the waste-source transect and at urban locations, in addition to one suburban location which had high concentration of SCCPs (S-14, 63 ng/m^3). In soil, the highest individual concentrations of CPs were found within the waste-source transect, followed by urban locations. The lowest concentrations of CPs in air and soil were found at the rural southeastern end of the transect, upwind from the city, where there is minor anthropogenic influence. SCCPs were strongly correlated with MCCPs in both air and soil (Spearman's rho: 0.95 and 0.91, respectively, p < 0.001).

Concentrations of CPs in air and soil within the waste-source transect showed large variability. Concentrations were generally low, except at the dumpsite (W-21, SCCPs: 59 ng/m^3 and 5300 ng/g dw, MCCPs: 33 ng/m^3 and 5100 ng/g dw) and the e-waste facility (W-23, SCCPs: 55 ng/m^3 and 480 ng/g dw, MCCPs: 32 ng/m^3 and 2700 ng/g dw). Considering the elevated concentrations of CPs in air and soil at these two locations, it appears clear that both municipal waste and e-waste represent important point sources for CPs. MCCP/SCCP ratios at the dumpsite (soil: 1.0, air: 0.55) and the e-waste facility (soil: 5.7, air: 0.60) imply that municipal waste is the dominant source for SCCPs, while both municipal waste and e-waste are important sources of MCCPs. This is likely related to differences in CPs content in products in the two waste streams.

Concentrations of CPs in African soils are previously reported for an e-waste site in Ghana (SCCPs: 150-28 000 ng/g dw, MCCPs: <LOD-1300 ng/g dw) and a municipal waste dumpsite in Sierra Leone (SCCPs: 69-1600 ng/g dw, MCCPs: <LOD-1400 ng/g dw) (Möckel et al., 2020). These MCCPs concentrations are two to four times lower than the concentrations found in soil at the dumpsite and e-waste facility in our study, while SCCPs concentrations are higher at the site in Ghana. Results also differ in two additional ways. Firstly, in the Möckel et al. (2020) study, concentrations of SCCPs were an order of magnitude

higher at the e-waste site than at the dumpsite, while in our study we found the opposite. Secondly, the MCCP/SCCP ratio in soil at the e-waste site was ≪1 in Ghana, compared to 5.7 at the e-waste facility in this study. The higher total CPs concentrations at the e-waste site in Ghana are likely related to i) Higher volume of e-waste processed, as Ghana is a major recipient in the global e-waste trade (Asante et al., 2019); and ii) Cruder processing methods in Ghana, including low temperature burning of cables and plastics. The higher MCCP/SCCP ratio in Tanzania is potentially also a consequence of socioeconomic factors, as Tanzania is more economically developed, and more modern electronics are therefore consumed. Modern electronics likely contain relatively more MCCPs due to recent production shifts from SCCPs towards MCCPs governed by international regulation (Glüge et al., 2018). This however also makes the year of sample collection a relevant factor (Ghana: 2015, this study: 2019).

Although both municipal waste and e-waste clearly act as important sources of CPs, also the urban environment, with its use of products containing CPs, contributes to local CPs pollution. However, widespread informal disposal of waste and small-scale informal e-waste processing are known to occur, making it difficult to separate the contributions of waste/e-waste compared to emissions from products in use based on CPs concentrations alone. Congener group profiles in soil (PCA, Fig. S11) does however suggest differences in sources between the e-waste facility, dumpsite, and urban areas, indicating that urban CPs contamination cannot be specifically or at least exclusively tied to waste and e-waste in the urban environment.

3.2.2. Urban pulse and waste related sources for dechloranes

Urban pulses were also observed for the dechloranes (Fig. 3). For concentrations of Dec 602 in air, and DP and Dec 603 in soil, differences between urban and rural locations were significant (p < 0.05). Although for Dec 603 in soil, the highest individual concentration by an order of magnitude was found in a suburban area (S-13, 3 ng/g dw). There were no significant differences between urban and rural locations for concentrations of DP in air and Dec 602 in soil (p > 0.05), however, detection frequencies were higher in urban areas.
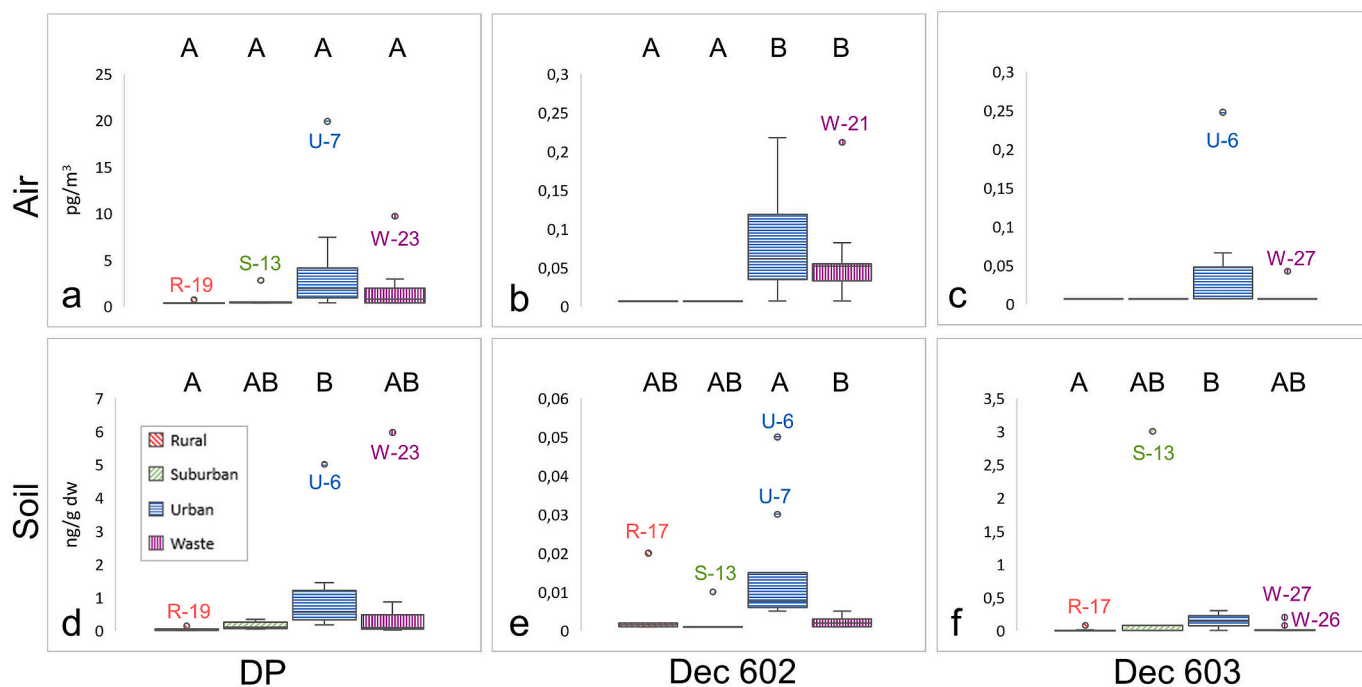

Fig. 3. Dechloranes concentrations in air (top) and soil (lower) in different settlement categories. The median value is represented by the line inside the box, the upper and lower sections of the box show the upper and lower quartiles. Whiskers represent minimum and maximum values. Outliers are marked as point outside the whiskers. Groups not significantly different from each other (Wilcoxon rank sum test) share letter code (A, B).

The concentration of DP in air and soil at the e-waste facility (9 pg/m^3 and 6 ng/g dw, respectively) was higher compared to the dumpsite (3 pg/m^3 and 0.9 ng/g dw). This suggests that unlike for CPs, the release of DP to the regional environment is greater from e-waste than municipal waste. Elevated levels of DP in the environment near e-waste recycling sites have also been documented in e.g., China and Vietnam (Chen et al., 2011; Someya et al., 2016). The concentration of DP in soil at the e-waste facility was comparable to the highest urban concentration (U-6, 5 ng/g dw), while the concentration of DP in air at the e-waste facility was a factor two lower compared to the highest urban location U-7 (20 pg/m^3). U-6 is located within ~500 m of an urban e-waste handler, and U-7 is located in the vicinity of a market where trade and repair of used electronics are taking place. It is thus possible that the high DP levels found at these locations are connected to electronics and e-waste. Any similar associations with electronics are not known for the other urban locations included in this study. Despite some apparent differences in primary sources, strong correlations were found between DP, SCCPs, and MCCPs in both air and soil (Spearman's rho: 0.66–0.88, p < 0.001).

Dec 603 generally showed weak correlations with the other analysed CECs (Table S10). This is also seen in a study from North America where the authors pointed to the occurrence of Dec 603 as an impurity in the historically used pesticides Aldrin and Dieldrin as the cause of these differences (Shen et al., 2010). The low concentrations and low detection frequency of Dec 603 in air samples compared to soil, and the lack of association with domestic waste or e-waste could comply with Dec 603 sources being of a legacy rather than contemporary nature. Combined, these factors suggest that the Dec 603 found in soils may be from past pesticide use. Spatial trends of Dec 602 did not allow any conclusions to be drawn regarding its sources.

3.3. Environmental processes and fate of CPs and dechloranes

3.3.1. Correlations of CPs and dechloranes with SOM

The SOM content was typically low in the studied region with a median value of 1.7 % (<0.01–10 %), and no significant correlations were found between SOM and soil concentrations of total SCCPs or MCCPs, nor individual congener groups (p > 0.05). This is consistent with other studies on CPs (Halse et al., 2015; Wang et al., 2014; Wang

et al., 2017). The correlation between SOM and soil concentration of DP was also not significant (p > 0.05). This suggests that proximity to sources is the main governing factor for the spatial variation in soil concentrations of CPs and DP in this study region. Correlations for SOM versus Dec 602 and Dec 603 in soil were significant (Spearman's rho: 0.47 for both, p < 0.05), further supporting legacy rather than current sources for Dec 603 in the region.

3.3.2. Distribution of CPs and dechloranes in soil and environmental fate

Soil consists of solids of minerogenic and organic nature, in addition to pore space filled with air and water. For organic compounds like CPs and dechloranes, distribution within the various soil compartments and associated loss/mobility processes are key factors in determining their environmental fate. These distributions and processes are governed by soil properties and the physicochemical properties of the compound, in addition to climatic conditions, and have here been assessed using a simple soil model (Fig. 4) (Gouin et al., 2000; Wong and Wania, 2011).

The studied CPs congener groups and dechloranes are predicted to be mainly associated with organic solids, rather than pore air or pore water (Fig. 4, left). This is also predicted to be the case when the soils are very dry (as during dry months) and very wet (as during rain-season) (Fig. S12), and within most (>90 %) of the range of SOM found in the studied soil. In dry soils with very low SOM content the area of mineral surfaces, thus the content of clay minerals (not included in the model), likely play a significant role in sorption (Goss et al., 2004).

CPs and dechloranes in soil are subject to diffusive and advective loss processes, mainly by erosion due to surface runoff, evaporation, and leaching to groundwater (Wong and Wania, 2011). Most of the studied CECs, particularly the dechloranes and longer chain, highly chlorinated CPs, are predicted to be mobilized mainly by erosion (Fig. 4, right). Evaporation may be an important loss process for more volatile short chained and low chlorinated congener groups. These predictions correspond well with congener group profiles of CPs found in air and soil. Air samples from all location categories were dominated by the congener groups shown by the soil model to be more mobile via evaporation (Fig. S8 and Fig. S9), while congener groups in soil showed an urban fractionation pattern (Fig. S10). Congener groups with relatively low volatility that are emitted in source areas appear to be deposited in soil locally. Congener groups in rural soil on the other hand are less
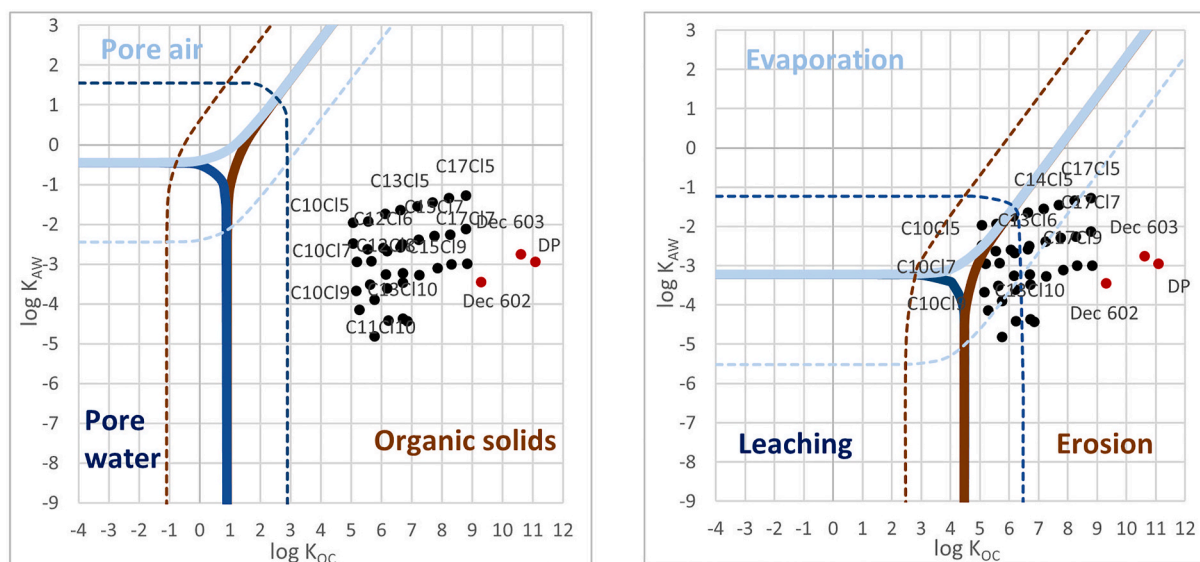

Fig. 4. Chemical partitioning space plot and mobility plot for CPs and dechloranes in soil with characteristics typical of the study area. CPs in black, dechloranes in red. Left: Predicted distribution of selected CPs congener groups and dechloranes in soil organic solids, soil pore air and soil pore water. Thick and dotted lines represent 50 % and 1 % in the phase, respectively. Right: Predicted relative importance of mobility processes (evaporation, leaching, and erosion). Thick and dotted lines represent 50 % and 1 % relevance of process, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

chlorinated and have shorter chain lengths (i.e., are more volatile), and are likely transported there via air from source areas. The model also corresponds well with spatial patterns for dechloranes in air and soil and fugacity calculations (see SI Text S3). Strong air to soil flux of dechloranes indicate that soil functions as a temporary storage reservoir for these compounds. This is a consequence of their physicochemical properties (low K_{AW}, high K_{OC}), suggesting that once in soil, dechloranes will not return to the gas phase. The model further predicts that leaching from soil to groundwater may be a relevant loss process for short chained CPs congener groups, while chlorination degree is not predicted to affect potential for leaching. This differs from a study on soil profiles in China, where both chain length and chlorination degree of CPs were seen to affect potential for leaching through soil (Zeng et al., 2011). Degradation processes may however have influenced these results. The soil moisture content, and by extension the monsoonal rain patterns, are predicted to affect the relative importance of mobility processes (Fig. S13). Less evaporation from soil due to soil pore-space being occupied by water could contribute to lower air concentrations of the more volatile SCCPs during periods of heavy rain. This has been observed in Australia (Gillett et al., 2017), and could help explain the seasonal trend seen for SCCPs. Also, the magnitude of mobility of the studied CECs will be affected by rainfall, particularly mobility via erosion which will increase with increased rainfall. Overall, the monsoonal rain patterns in the region seem to have significant effects on the environmental fate of CPs and dechloranes.

4. Conclusions

The levels of CPs and dechloranes in air and soil in the Dar es Salaam region presented in this study adds to the knowledge on CECs in tropical environments and in the Global South. Urban pulses and spatial association with general waste and e-waste indicate that consumption and waste handling practices contribute to local pollution of CPs and dechloranes. This should be addressed at a regional level through improved waste management. It should also be addressed in international regulatory efforts relating to international trade of products, used products, and waste containing these compounds, as the situation is likely to be similar in other countries with comparable socioeconomic conditions.

The relatively high levels of CPs and dechloranes reported in this study underlines the importance of expanding environmental monitoring of CECs in economically developing tropical regions, particularly in urban areas where large populations potentially are exposed to high CECs concentrations. More data on CECs in these regions is also of importance to improve the understanding of environmental distribution and fate of these compounds.

This study showed that the monsoonal rain in the region affects the environmental fate of the studied CECs. For air concentrations, differences were seen compared to seasonal trends in temperate and subtropical regions. These findings have implications for future air monitoring strategies in regions with monsoonal rain. However, our results are based on limited data, and we therefore suggest future studies on CPs and dechloranes in air in the region include studies of seasonal variation at multiple sites.

Credit author statement

Maja Nipen: Conceptualization, Investigation, Formal analysis, Writing – Original Draft. Rolf David Vogt: Conceptualization, Supervision, Investigation, Writing – Review and Editing. Pernilla Bohlin-Nizzetto: Conceptualization, Supervision, Writing – Review and Editing. Katrine Borgå: Conceptualization, Supervision, Project Administration, Writing – Review and Editing. Eliezer Brown Mwakalapa: Investigation. Anders Røsrud Borgen: Supervision, Investigation, Writing – Review and Editing. Susanne Jøntvedt Jørgensen: Investigation. Samwel Moses Ntapanta: Investigation. Aviti John Mmochi: Resources, Writing – Review and Editing. Martin Schlabach: Supervision,

Resources. Knut Breivik: Conceptualization, Supervision, Writing – Review and Editing.

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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2021.118298.

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