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Chlorinated paraffins and dechloranes in free-range chicken eggs and soil around waste disposal sites in Tanzania

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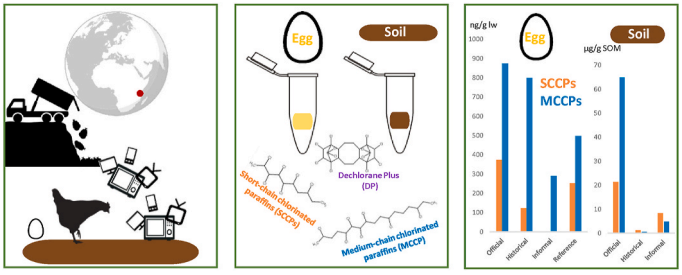
HIGHLIGHTS

- We quantify chlorinated paraffins and dechloranes in Tanzanian soil and chicken eggs.
- No clear association in contaminant levels or patterns between soil and eggs.
- Elevated contaminant levels in soil from e-waste location, but not in eggs.
- Risk assessment suggest concern for SCCP exposure via egg consumption at one site.

GRAPHICAL ABSTRACT



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ABSTRACT

Electronic waste is a source of both legacy and emerging flame retardants to the environment, especially in regions where sufficient waste handling systems are lacking. In the present study, we quantified the occurrence of short- and medium chain chlorinated paraffins (SCCPs and MCCPs) and dechloranes in household chicken (*Gallus domesticus*) eggs and soil collected near waste disposal sites on Zanzibar and the Tanzanian mainland. Sampling locations included an e-waste facility and the active dumpsite of Dar es Salaam, a historical dumpsite in Dar es Salaam, and an informal dumpsite on Zanzibar. We compared concentrations and contaminant profiles between soil and eggs, as free-range chickens ingest a considerable amount of soil during foraging, with potential for maternal transfer to the eggs. We found no correlation between soil and egg concentrations or patterns of dechloranes or CPs. CPs with shorter chain lengths and higher chlorination degree were associated with soil, while longer chain lengths and lower chlorination degree were associated with eggs. MCCPs dominated the CP profile in eggs, with median concentrations ranging from 500 to 900 ng/g lipid weight (lw) among locations. SCCP concentrations in eggs ranged from below the detection limit (LOD) to 370 ng/g lw. Dechlorane Plus was the dominating dechlorane compound in all egg samples, with median concentrations ranging from 0.5 to 2.8 ng/g lw. SCCPs dominated in the soil samples (400–21300 ng/g soil organic matter, SOM), except at the official

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dumpsite where MCCPs were highest (65000 ng/g SOM). Concentrations of dechloranes in soil ranged from below LOD to 240 ng/g SOM, and the dominating compounds were Dechlorane Plus and Dechlorane 603. Risk assessment of CP levels gave margins of exposure (MOE) close to or below 1000 for SCCPs at one location.

1. Introduction

Chemical flame retardants are added to a wide range of consumer goods, such as electronic products, that eventually become obsolete. Lack of sound handling and recycling of waste in developing regions in Africa represents an emerging environmental concern (White et al., 2020; Nipen et al., 2022b). Electronic waste (e-waste) is a source of both legacy, e.g. polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs), as well as emerging flame retardants, e.g. chlorinated paraffins (CPs) and dechloranes, that are replacing regulated contaminants (Abbasi et al., 2019; Nevondo and Okonkwo, 2021; White et al., 2020). Apart from some production of short chain chlorinated paraffins (SCCPs) in South Africa (10,000 tons year-1 as per 2015) and Egypt, other production of these contaminants has not been reported for Africa (Breivik et al., 2002; Abbasi et al., 2019; Hansen et al., 2020; Nevondo and Okonkwo, 2021). Thus, their increased environmental occurrence in this region is largely attributed to increased import and use of consumer goods along with lack of sound treatment of end-of-life products and waste (Breivik et al., 2015; Nipen et al., 2022b).

Declining levels of atmospheric legacy contaminants seen in industrialized regions as a result of reduced emissions is generally not found in developing countries (Gioia et al., 2011; Wong et al., 2021). Temporal trend studies using dated sediment cores show that concentrations of both legacy and emerging contaminants often peak in surface sediment layers in developing and newly industrialized regions, suggesting recent sources, compared to regions that have remained industrialized for some time, where peak concentrations in sediment layers correlate with peak in global production (Nipen et al., 2022b). A spatial shift in contaminant sources is thus occurring, from source regions where production and use have taken place historically, to source regions at the receiving end of obsolete products and waste (Breivik et al., 2011; Abbasi et al., 2015; Nipen et al., 2022b). This spatial shift can be attributed to increasing economic development and industrialization as well as a range of socio-economic factors (Nipen et al., 2022b), and stresses the need for expanding the research efforts on environmental contamination to developing regions in the Global South. Tanzania is experiencing rapid economic and population growth, expanding industry, and increasing import of consumer goods and potentially also waste. With limited capacity to safely manage an increasing stream of waste, both industrial and domestic, Tanzania faces a challenge regarding environmental contamination (Yhdego, 2017; Mahenge et al., 2018).

CPs are a large group of polychlorinated alkanes that encompass thousands of different congeners that are used in metal-working fluids, paints and sealants, and added to a wide range of products as plasticizers and flame retardants (Glüge et al., 2016, 2018). CPs have become ubiquitous in the environment since their introduction in the 1930s, and annual production is today estimated to exceed 1 million tons (Glüge et al., 2016, 2018; Wei et al., 2016; Vorkamp et al., 2019). Based on the carbon chain length, CPs can be classified into three groups: short chain CPs (C10–C13, SCCPs), medium chain CPs (C14–C17, MCCPs), and long chain CPs (>C18, LCCPs). They cover a wide range of physical-chemical properties, with octanol-water partitioning coefficients (Kow) varying from log Kow 5 to 7.5 for SCCPs and 6 to 10 for MCCPs (Glüge et al., 2013). Since the listing of SCCPs under Annex A of the Stockholm Convention on Persistent Organic Pollutants in 2017 (UNEP, 2017), MCCPs have increasingly been introduced as replacement substances (Glüge et al., 2018). However, studies indicate that MCCPs have similar Persistent, Bioaccumulative and Toxic (PBT) properties as the regulated SCCPs (Castro et al., 2019), and are currently listed for consideration under the European Chemicals Agency (ECHA, 2021). Dechloranes are another group of high production volume chlorinated flame retardants that includes Dechlorane Plus (DP) and Dechlorane 602 and 603. DP is used in plastic and electronic products and is currently listed for consideration under the Stockholm Convention due to its bioaccumulative properties and ability to undergo long-range transport (Hansen et al., 2020; Schuster et al., 2020). Estimated annual production of DP is around 750–6000 tons, while less is known about the chemical properties and production volumes for Dechlorane 602 and 603 (Wang et al., 2016; Hansen et al., 2020). The EFSA Panel on Contaminants in the Food Chain (CONTAM) recently performed benchmark dose (BMD) modelling on data from various studies on the effects of CPs on rodents, and selected as reference points a BMDL10 of 2.3 × 106 ng/kg body weight/day and 36 × 106 ng/kg body weight/day for SCCPs and MCCPs, respectively (EFSA CONTAM Panel et al., 2020). Similar reference points for risk assessment of dechloranes in food for human consumption are not available.

Most studies reporting occurrence of CPs and dechloranes in the environment are from the Northern hemisphere, and China is the main producer of these chemicals (Houde et al., 2008; Clement et al., 2012; Wei et al., 2016; Zhang et al., 2019). Very few studies have investigated CPs and dechloranes in the sub-Saharan African environment (Moeckel et al., 2020; Nevondo and Okonkwo, 2021; Nipen et al., 2022a). Concentrations and composition profiles of CPs and dechloranes in various environmental matrices are governed by complex processes and mechanisms including distance from primary sources, temperature and season, and distance from urban and industrialized areas (Wang et al., 2013; Nipen et al., 2022a). Due to relatively high Kow, these substances will partition to hydrophobic matrices such as organic matter in soil and lipids in organisms, which also affect their environmental distribution and accumulation (Castro et al., 2019; Hansen et al., 2020). In a tropical climate, semi-volatile organic substances, like PCBs, dechloranes and CPs, will more readily evaporate and could be transported and deposited at higher latitudes (Breivik et al., 2011; Vorkamp et al., 2019). Recently, elevated concentrations of CPs and dechloranes were found in air and soil in urban areas and around waste and e-waste disposal sites in Dar es Salaam, Tanzania (Nipen et al., 2022a). CPs were also detected in human milk samples from Africa, with concentrations exceeding concentrations of PCBs, and with a dominance of MCCPs over SCCPs suggesting a shift towards non-regulated compounds (Krätschmer et al., 2021). Prior to our study, no peer-reviewed literature exists on CPs and dechloranes in African biota, but SCCP concentrations in chicken eggs from selected sites in Africa, including Tanzania, are reported by the International Pollutants Elimination Network (IPEN) showing elevated levels around waste and e-waste disposal sites (Petrlik et al., 2021). Eggs from free-range household chickens (*Gallus domesticus*) are ideal indicators for contaminated soils, and eggs are a commonly included matrix in environmental monitoring programs globally (Polder et al., 2016; Petrlik et al., 2022). In addition to commercial feed, invertebrates, seeds and household leftovers, free-range chickens ingest considerable amounts of soil during foraging, which can lead to accumulation of contaminants in the tissues of the hen and further transfer into the egg (De Vries et al., 2006; Waegeneers et al., 2009; Polder et al., 2016; Petrlik et al., 2022). Chickens also accumulate CPs from feed (Ueberschär et al., 2007), and mixtures of both short- and medium chained CPs were found to be transferred from chicken to egg with increasing accumulation ratios with increasing carbon chain length and chlorine content (Mézière et al., 2021).

Few studies have previously quantified emerging contaminants in tropical regions far from production and use, where occurrence of these contaminants is expected to increase due to economic development and

industrialization. The aim of the present study was to assess the occurrence of CPs and dechloranes in soil and chicken eggs around waste disposal sites on mainland Tanzania and Zanzibar and to investigate if and how concentrations and congener group profiles vary among locations and between matrices. Collection of soil samples were conducted during an initial screening study of contaminant occurrence around waste disposal sites. As these initial analyses of soil samples showed elevated concentrations of CPs and dechloranes, this motivated collection of eggs from free-range, household chickens from the same locations and one presumed reference site in later fieldwork campaigns. Using the reference dose levels set for CPs and a margin of exposure (MOE) approach, risk assessment of CP exposure via egg consumption was conducted.

2. Materials and methods

2.1. Sample sites

In our initial screening sampling in January 2018, soil from waste disposal sites on the Tanzanian mainland and Zanzibar were specifically targeted to assess CP and dechlorane contamination from waste in general, and from electronic waste in particular (Fig. 1). The two sample sites in Dar es Salaam consisted of one historical dumpsite and the current official dumpsite. The historic Mtoni dumpsite in Dar es Salaam (−6.8730°, 39.2832°) was closed in 2009 after operating for around six years and is located around 8 km from Dar es Salaam city center. This historical dumpsite is in close vicinity of the Kizinga River flowing into Kurasini Creek, surrounded by residential areas. The currently used, official dumpsite at Pugu Kinyamwezi (−6.9278°, 39.12785°) is located around 25 km from the city center. Despite initial plans for building a sanitary landfill for Dar es Salaam, Pugu is today receiving industrial, agricultural, domestic, commercial, medical and other hazardous wastes (e.g. e-waste) without restrictions or regulations regarding leachate and gas management, fencing, soil covering and worker safety measures (Yhdego, 2017). A licensed e-waste dealer is located next to the official dumpsite, where e-waste collection, sorting and dismantling take place. On Zanzibar, the Maruhubi dumpsite (−6.1442°, 39.2101°) is an informal dumpsite receiving solid wastes from Zanzibar municipality. It is located around 3 km from Zanzibar city center, and close to mangrove forests. From these same sites, chicken eggs were sampled in August 2018 (Zanzibar) and February 2019 (Tanzanian mainland/Dar es Salaam). In addition, eggs were collected from an area away from any known waste disposal sites, a residential area (Bumbwini Makoba, −5.9500°, 39.2000°) around 25 km North of Zanzibar city. Soil samples were not obtained from the residential area on Zanzibar. However, soil samples collected at rural locations around Dar es Salaam as part of a parallel study by Nipen et al. (2022a) are used for reference.

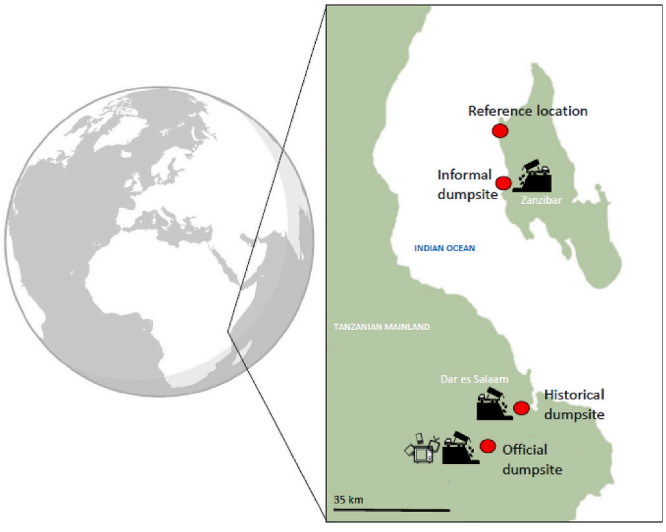

Fig. 1. Sampling locations on mainland Tanzania and Unguja island of Zanzibar. Free-range household chicken eggs and soil were collected at three waste disposal sites: The official dumpsite of Dar es Salaam located next to a small e-waste handling facility, a historical dumpsite in Dar es Salaam, no longer in use, and an informal dumpsite on Zanzibar, where unofficial waste disposal takes place. Only egg samples were collected from the residential area, located away from any waste disposal sites.

2.2. Sample collection and preparation

At each location, three soil cores (approximately 5 cm deep) were collected within a range of three to five square meters using a stainless-steel hand-held corer. The three cores from each site were pooled into one composite sample. Soil organic matter content (SOM) was estimated by loss on ignition using dried sample material. SOM in the soil samples varied among locations, from dry, sandy soil at the official dumpsite (SOM 3.2%), to 12% SOM at the informal dumpsite on Zanzibar. Two composite soil samples were collected at the historical dumpsite, one from a location influenced by run-off from the old dumpsite and one around 200 m away from the run-off location. The latter had lower SOM content (4.1%) compared to the area influenced by run-off from the old dumpsite (8.3% SOM).

The chickens were kept outside picking food from the local grounds, but owners also reported giving them additional feed such as food scraps and industrial feed. Six eggs were collected from each location, in total 24 eggs. The egg-laying sequence of the sampled eggs is not known. The whole egg content was collected in falcon tubes and kept frozen (−20 °C) until laboratory analyses.

2.3. Sample extraction

Composite soil samples (around 7 g) were dried and homogenized together with sodium sulphate with a mortar and pestle to a fine powder. Fresh samples of whole eggs (around 1 g) were used for extraction after homogenization with sodium sulphate. Soil samples were extracted using accelerated solvent extraction, while egg samples were extracted using a cold column technique, both extractions using acetone and n-hexane (1:1) after the addition of internal standards (13C - labelled 1,5,5,6,6,10 – hexachlorodecane, DP syn and Dec 602). Extracts were concentrated using a Turbovap system (Zymark) and sulphuric acid was used for removal of lipids. Extracts were cleaned with 4 g activated silica and a 1 cm layer of sodium sulphate packed in a column eluted with diethyl ether in n-hexane (1:9). Solvent was changed to isooctane, volume concentrated to 100 μL, and 1,2,3,4-tetrachloronaphtalene (TCN) was added as recovery standard. Extracts were analyzed for CPs, 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 capturenegative ionization (ECNI) mode. 34 SCCPs and 28 MCCPs congener groups (groups of CPs sharing the same molecular formula) were identified and quantified. A total of 22 egg samples were analyzed for CPs (two samples were lost during sample preparation), and 24 egg samples were analyzed for dechloranes. Lipid content in the egg samples was determined gravimetrically.

2.4. Quantification of chlorinated paraffins

The quantification of CPs in soil in the initial screening study was conducted using the method described by Tomy et al. (1997), where the technical mixture with the closest matching chlorination degree and congener group pattern is used for quantification of each sample. As the chemical analysis of chlorinated paraffins was a field in rapid development at the time of our study, the laboratory further improved its methods and updated to the deconvolution method developed by Bogdal

et al. (2015), by the time of the follow up study with egg analysis. The congener group profiles of CPs are comparable across the two methods of quantification as the same instrument was used for both soil and egg samples. Unfortunately we were not able to re-quantify the initial screening samples based on the Bogdal method, but our soil data from the official dumpsite/e-waste facility of Dar es Salaam were comparable to CPs quantified in soil samples collected from the same location the following year using the deconvolution method (Nipen et al., 2022a) (See quality assurance below).

The deconvolution method by Bogdal et al. (2015) involves quantification of CPs using technical mixtures with different chlorination degree (SCCPs: C10 – C13 standards with 51%, 55% and 63% Cl; MCCPs: C14 – C17 standards of 42%, 52%, and 57% Cl, Dr. Ehrenstorfer, Germany) and standards with single carbon chain lengths as calibration standards (SCCPs: C10, C11, C12, and C13 (50% and 65% Cl); MCCPs: C14 with 52% Cl, Dr. Ehrenstorfer, Germany). A mathematical deconvolution method reconstructs the congener group pattern in the samples using a linear combination of the congener group patterns in the calibration standards. Match between sample and standard pattern was assessed with the goodness of fit where R2 above 0.5 was considered acceptable. Average R2 for SCCPs were 0.91 while average R2 for MCCPs was 0.89 (Table A1).

2.5. Quality assurance & quality control

Despite using different quantification methods of soil CP occurrence in the present study and the soil transect study by Nipen et al. (2022a), the concentrations are comparable in the two soil samples from same site (the official dumpsite/e-waste facility in Dar es Salaam: present study: 670 ng/g dw SCCPs, 2100 ng/g MCCPs, and 7.6 ng/g dw DP. Sample W-23 from Nipen et al., 2022a: 480 ng/g dw SCCPs, 2700 ng/g dw MCCPs, and 6 ng/g dw DP). This confirms the quality and representativeness of the quantification methods used in the present study.

Substantial measures were done to avoid contamination during sample preparation and extraction, including use of high-quality grade solvents, glassware heat-treated at 450 °C and rinsed with solvents immediately prior to use, and sample preparation procedures performed in a laminar flow cabinet when possible. For every sample batch (samples were run in five batches), 1–2 blank samples were included to account for any background signal or contamination of CPs during the extraction. In total, eight blank samples were run to assure representative values of the background signal. The limit of detection (LOD) was defined as three times standard deviation of the blanks. The LODs for CPs in eggs were 16.2 ng/g ww and 65.0 ng/g ww for SCCPs and MCCPs, respectively, and 11 ng/g dw and 19 ng/g dw for SCCPs and MCCPs, respectively, in soil. LODs for dechloranes in eggs were 0.01 ng/g ww for Dechlorane 602 and 603, and 0.02 for Dechlorane Plus. The LODs for dechloranes in soil were 0.01 and 0.001 ng/g dw for Dechlorane Plus, and Dechlorane 602 and 603, respectively. The ratios of TCN and 13C labelled internal standards were compared against the ratios of TCN and internal standards in quantification mixtures (which were not subject to extraction and clean-up) to determine the recovery of internal standards. Recovery of CPs in soil samples were between 54% and 57% and between 71% and 87% for dechloranes. Recovery of CPs in eggs were between 83% and 130% (Table A1). Average recoveries of dechloranes in eggs were between 84% and 147% (calculated from two radiolabeled congeners used for recovery calculations, Table A2). Quantification of both CPs and dechloranes involves addition of internal standards prior to sample extraction and clean up, and thus recovery percentage is automatically corrected for.

2.6. Risk assessment of CP intake via egg consumption

Human risk assessment of CP intake via eggs was assessed using the margin of exposure (MOE) approach. Egg consumption for the Tanzanian population is estimated to be 106 eggs per person per year (Ringo and Lekule, 2020), and average weight for all eggs sampled in the present study (46.3 g) was used to calculate daily egg intake in grams per day. Daily intake of CPs via consumption of eggs was calculated using the following formula:

$$\text{Egg intake} = (\text{ConcCP x ecd x lipidegg}) \ / \ \text{BW}$$

where ConcCP is SCCP or MCCP concentration in the egg samples (ng/g lw); ecd is the daily egg consumption (g/day); lipidegg the average lipid content (%) of eggs at each location in the present study; and average human body weight (BW), set at 60 kg. The MOE was calculated using the following formula:

$$\text{MOE} = \text{BMDL10} \ / \ \text{Egg intake}$$

where BMDL10 is 2.3 × 106 ng/kg bw/day and 36 × 106 ng/kg bw/day for SCCPs and MCCPs, respectively. The CONTAM Panel conclude that a MOE higher than 1000 indicate no health concern.

2.7. Data treatment and statistical analyses

All statistical analyses were conducted using R Studio. Compounds detected below LOD in more than 85% of the samples were removed from further data treatment. This included SCCPs in eggs from the informal dumpsite on Zanzibar (Table A1). Otherwise, values that were quantified but below LOD were included for data treatment and statistical analyses. This included three egg samples for SCCPs and five samples for MCCPs (Table A1). Due to low sample size, and because assumptions of normality and equal variance were not met, assessment of spatial variation of contaminant concentrations in eggs was conducted using the non-parametric Kruskal Wallis test.

Assessment of CP congener group profiles among sites and between matrices was done using principal component analyses (PCA) with the vegan package for R (Jari Oksanen et al., 2019). All variables were scaled to zero mean and unit standard deviation. For the multivariate assessment of CP congener group profiles, individual congener groups (e.g. C10H17Cl5) were normalized to total CP concentrations (SCCPs + MCCPs). For visualization of CP congener group profiles in Fig. 4, individual congener groups were normalized to total SCCP and MCCP concentrations, respectively.

3. Results

CPs was the dominant contaminant group in both egg and soil samples from the present study, with concentrations up to three orders of magnitude higher than for dechloranes (Table 1). Elevated concentrations of SCCPs, MCCPs and DP in soil were found at the official dumpsite compared to the other locations (Fig. 2), but there was no clear association between concentrations or congener group profiles between egg and soil samples. Contaminant concentrations in eggs did not differ between dumpsites and the reference location, and the standard deviation within sites was relatively large (Table 2). Contaminant concentrations on a wet weight/dry weight basis with LODs annotated can be found in the Appendix (Figure A1).

Mean lipid content in eggs ranged from 8.9% from the official dumpsite to 14.2% in eggs from the residential area. Due to the high lipid solubility of CPs and dechloranes and differences in lipid content among eggs from the different locations, the assessment of spatial variation in contaminant concentrations was conducted on a lipid weight basis to ensure that spatial variability in CP and dechloranes reflects variation beyond lipid content. SOM content in the soil samples ranged from 3% to 12%. Due to low sample size, statistical testing of differences in SOM content or contaminant concentrations among locations could not be performed, but data are presented normalized to SOM, also to allow comparison to the egg lipid normalized concentrations.

Table 1
Concentrations of short and medium chained chlorinated paraffins (SCCP, MCCP) and dechloranes (mean (median), range (standard deviation, SD), ng/g) in chicken eggs from four locations in Dar es Salaam (Tanzanian mainland) and Zanzibar (Unguja island).

Location	n	Lipid%	SCCP	MCCP	MCCP/SCCP	Dech Plus (DP)	Dech 602	Dech 603
		mean	**ng/g wet weight** *mean (median)* *min-max (SD)*					
Official, DAR	5/6[a]	8.9	38 (37) 32–49 (6)	81 (76) <LOD–110 (18)	2.7	0.04 (0.04) 0.03–0.05 (0)	<LOD	<LOD
Historical, DAR	6	10	40 (17) <LOD–115 (41)	130 (78) <LOD–305 (93)	4.4	0.3 (0.1) 0.1–0.6 (0.2)	<LOD	0.2 (0.1) 0.1–0.3 (0.1)
Informal, ZNZ	6	11.6	<LOD	64 (67) <LOD–75 (10)	6.6	0.3 (0.3) 0.2–0.5 (0.1)	0.02 (0.02) 0.01–0.02 (0.002)	0.2 (0.3) 0.2–0.3 (0.03)
Reference, ZNZ	5/6[a]	14.2	33 (28) 19–57 (13)	86 (70) <LOD–127 (28)	2.9	0.1 (0.1) 0.1–0.3 (0.1)	<LOD	0.2 (0.1) 0.03–0.9 (0.3)
		mean	**ng/g lipid weight** *mean (median)* *min-max*					
Official, DAR	5/6[a]	8.9	480 (370) 290–790 (185)	1100 (870) <LOD–2400 (655)	2.7	0.5 (0.5) 0.3–0.8 (0.2)	<LOD	<LOD
Historical, DAR	6	10	380 (220) <LOD–940 (325)	1200 (900) <LOD–2500 (683)	4.4	2.5 (1.5) 0.7–5 (1.8)	<LOD	1.6 (1.4) 0.9–2.7 (0.7)
Informal, ZNZ	6	11.6	<LOD	560 (560) <LOD–670 (91)	6.6	2.9 (2.8) 1.6–4.1 (1)	0.2 (0.2) 0.1–0.2 (0.02)	2.1 (2.2) 1.8–2.4 (0.2)
Reference, ZNZ	5/6[a]	14.2	240 (250) 130–390 (94)	650 (500) <LOD–1230 (323)	2.9	0.8 (0.7) 0.4–2 (0.6)	<LOD	1.5 (0.4) 0.3–6.2 (2.1)

[a] Five eggs analyzed for chlorinated paraffins, six eggs analyzed for dechloranes.

3.1. Concentrations of chlorinated paraffins in eggs and soil

SCCPs were detected above LOD in 13 of 22 egg samples, while MCCPs were above LOD in 16 of 22 egg samples. Median concentrations of SCCPs in eggs ranged from <LOD to 370 ng/g lipid weight (lw) among locations. SCCPs were below LOD in all eggs from the informal dumpsite on Zanzibar while highest concentrations were found at the official dumpsite ($p = 0.02$, $\chi 2$ Kruskal Wallis = 14.5, Fig. 2, Table 1). MCCP concentrations in eggs were higher compared to SCCPs at all locations (SCCP/MCCP ratio ranging from 0.3 to 0.5, Table 1). Concentrations ranged from 500 to 900 ng/g lw with no significant difference among locations ($p = 0.09$, $\chi 2$ Kruskal Wallis = 6.43, Fig. 2). The average MOE values ranged from 2400 to 3100 for SCCPs and 13,500 to 24,900 for MCCPs among locations (Table A3). At all locations, MOE for MCCPs exceeded 1,000, while MOE for SCCPs was below or close to 1000 (890 and 1300, Fig. 3) in two egg samples collected at the historical dumpsite in Dar es Salaam.

In the soil samples, CP concentrations were highest at the official dumpsite next to the e-waste handling facility in Dar es Salaam. MCCPs was the dominating CP group at the official dumpsite (SCCP/MCCP ratio 0.3, Table 1), while SCCPs dominated at the other locations. Concentrations among locations ranged from 400 to 21300 ng/g SOM for SCCPs and <LOD to 65000 ng/g SOM for MCCPs (Table 1).

3.2. Concentrations of dechloranes in eggs and soil

Dechlorane Plus (combining both syn- and anti-isomers) was detected in eggs from all locations, and was the dominant dechlorane group. Median DP concentrations ranged from 0.5 ng/g lw to 2.8 ng/g lw among locations, and highest concentrations were found at the informal dumpsite on Zanzibar ($p = 0.006$, $\chi 2$ Kruskal Wallis = 12.3, Fig. 2). Dechlorane 603 was detected above LOD in 18 of 24 samples, with median concentrations ranging from <LOD to 2.2 ng/g lw. Dechlorane 602 was only detected above LOD in eggs from the informal dumpsite on Zanzibar (Table 1).

Dechlorane 602 was detected at relatively low concentrations (below or close to LOD) in soil from all sites. However, relatively high concentration of Dechlorane 603 was found at the historical dumpsite, at the location away from the area affected by run-off from the old dumpsite. Here, concentration of Dech 603 was up to two orders of magnitude higher compared to the other locations, and higher than DP from the same site (230 and 12 ng/g SOM, respectively). Except from at this site, DP dominated the dechlorane pattern in soil, and the concentration was highest at the official dumpsite of Dar es Salaam and the informal dumpsite on Zanzibar, 240 and 70 ng/g SOM, respectively (Table 2).

3.3. CP congener group profiles in egg and soil

C10 and C14 homologues were evenly distributed among samples and locations, while C15–C17 homologues were generally more present in eggs compared to soil. SCCPs with higher chlorination degree dominated in the soil samples, while MCCPs with lower chlorination degree dominated in the egg samples (Fig. 4). The multivariate analyses of CP congener group profiles showed a gradient from high to low chlorination degree along the first principal component (PC1 28%) of the PCA plot (Fig. 5). Congener groups with more chlorine atoms pointed in the direction of the soil samples, and groups with fewer chlorine atoms point in the direction of the egg samples. There is a gradient from low to high carbon chain length along the second principal component (PC2, 21%), with increasing relative contribution of CPs with shorter carbon chain lengths in soil samples and longer carbon chain lengths in egg samples with increasing PC2 values. PC1 and PC2 together account for 49% of the total variation in the dataset, meaning that there might be other drivers of variation in the data not captured in this study. Some CPs had higher absolute loadings on the third axis, PC3 (C10Cl5, C10Cl6,

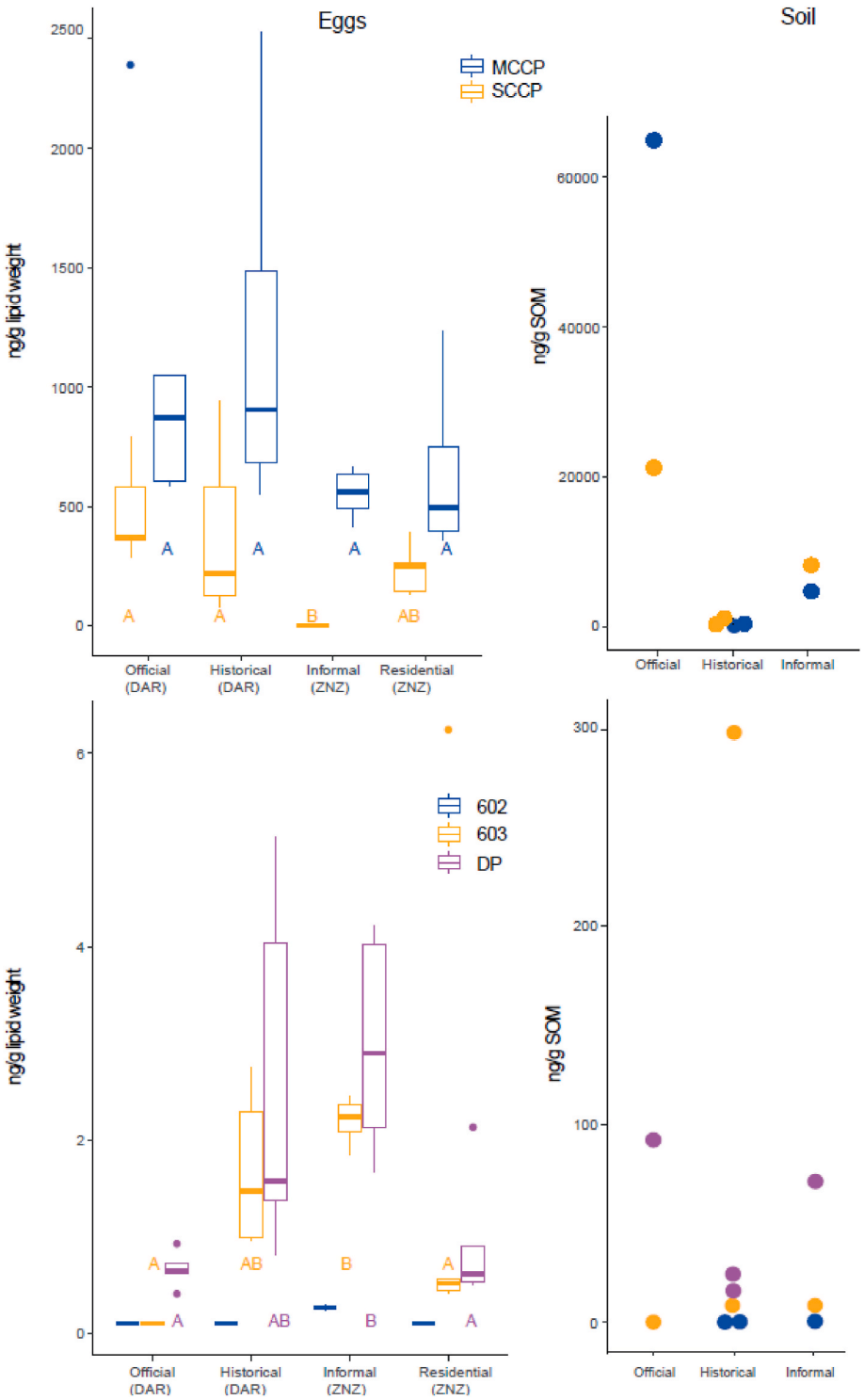

Fig. 2. Concentrations of chlorinated paraffins and dechloranes in eggs (ng/g lipid weight) and soil (ng/g SOM) from three dumpsites on mainland Tanzania and Zanzibar. Groups not significantly different from each other (Kruskal Wallis test) share letter code (A, B).

C13Cl5, C13Cl6, C14Cl5, C16Cl7, C16Cl8), but due to the low amount of variation explained by this axis (12%), PC3 was not considered important for interpretations of the main trends in the results.

4. Discussion

Elevated concentrations of SCCPs, MCCPs and DP in soil at the official dumpsite next to the e-waste facility in Dar es Salaam could indicate that waste and e-waste represent a contaminant source to the Tanzanian environment. However, due to the low spatial variation of CP concentrations in eggs and the large variation within sites, no clear conclusions regarding a waste signal can be made. Unknown contaminant sources at the residential area cannot be ruled out. Particularly high concentrations of SCCPs in two eggs from the historical dumpsite resulted in MOE close to or below the 1000-limit set by the CONTAM panel. This suggests that consumption of eggs from this location represent a health concern, which warrants further research on human exposure to CPs and their health effects.

Table 2
Concentrations of chlorinated paraffins and dechloranes in soil samples collected at three locations around Dar es Salaam and Zanzibar, Tanzania. Each soil sample consists of three composite samples.

Location	%SOM	SCCP	MCCP	SCCP/MCCP	Dech Plus (DP)	Dech 602	Dech 603
ng/g dry weight							
Official (DAR)	3.2	670	2100	0.3	7.6	0.004	<LOD
Historical (DAR)	8.3	100	40	2.5	0.7	0.01	0.4
Historical II (DAR)	4.1	17	<LOD	1.9	0.5	<LOD	10
Informal (ZNZ)	12.1	1010	580	1.7	8.4	0.1	0.3
ng/g SOM							
Official (DAR)	3.2	21300	65000	0.3	240	0.1	<LOD
Historical (DAR)	8.3	1250	500	2.5	9	0.1	4
Historical II (DAR)	4.1	400	<LOD	1.9	12	<LOD	230
Informal (ZNZ)	12.1	8300	4800	1.7	70	0.6	2.5

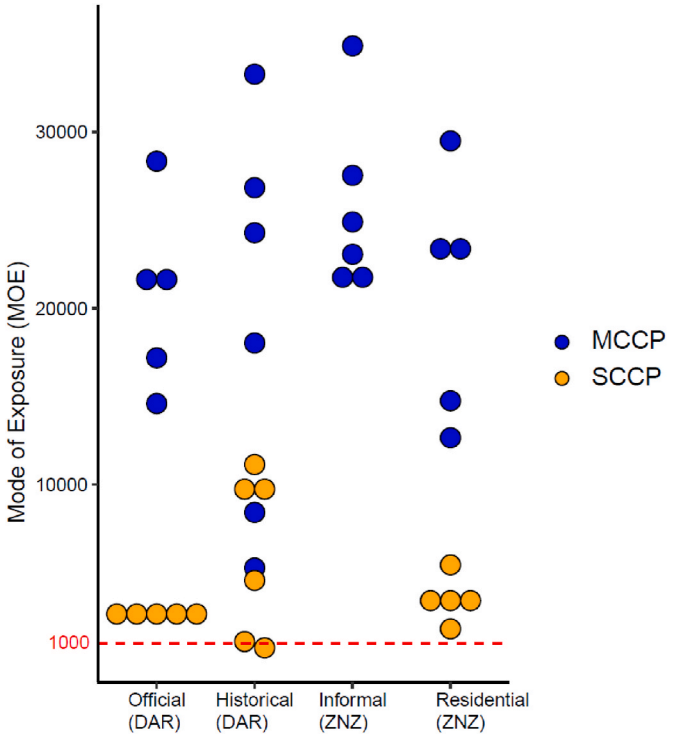

Fig. 3. Mode of Exposure (MOE) calculated for SCCPs and MCCPs in individual egg samples collected from locations on mainland Tanzania and Zanzibar. The MOE limit at 1000 (indicated by the red, dotted line) is set by the CONTAM panel for human exposure to CPs and MOE above this limit indicate no risk. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Despite limited amount of soil samples collected in the present study, differences in CP congener group patterns between soil and eggs were observed. While MCCPs was the dominant CP group in the egg samples, SCCPs dominated in two out of three soil samples. This may be due to higher Kow and affinity to lipid-rich tissues such as eggs for CPs with longer carbon chain lengths. Elevated concentrations of Dechlorane Plus in soil from the official and informal dumpsites, compared to the historical dumpsite, could also indicate a waste signal. As opposed to the historical dumpsite, the informal and official dumpsites are active and currently receiving household wastes, which might be represented by more modern products and consumer goods compared to what has been discarded at the historical dumpsite that was shut down in 2009. Elevated concentrations of Dechlorane 603 at the historical dumpsite but not at the official dumpsite suggest local use of pesticides rather than a waste signal, as this compound can occur as an impurity in the pesticides aldrin and dieldrin (Shen et al., 2011). Higher concentrations of MCCPs compared to SCCPs in soil samples at the official dumpsite next to the e-waste handling facility could be a consequence of more modern products containing MCCPs being disposed of at this location, as regulatory efforts have led to a dominance of MCCPs over SCCPs in environmental compartments (Gluge et al., 2018). The calculated MOEs for the CP exposure via consumption of eggs from the present study do not suggest a health concern for MCCPs (all MOEs >1000), whereas one egg from the historic waste site had SCCPs levels of consern with MOE values below 1000. However, this limited scenario does not account for differences in egg consumption or CP exposure from other sources, and further investigations should be made regarding human exposure to CPs and dechloranes and potential health risks.

In general, chlorinated paraffin and dechlorane concentrations in eggs and soil in the present study were higher compared to what has been reported from Europe, but several orders of magnitude lower compared to e-waste locations in China (Table 3). Due to few replicates in the present study, comparisons between other studies must be done with care. In addition, analytical challenges and uncertainties, especially regarding quantification of CPs, make comparisons among different laboratories challenging. Concentrations of CPs and DP in home-produced eggs from e-waste locations in China were up to three orders of magnitude higher compared to the present study (Zheng et al., 2012; Zeng et al., 2018). Concentrations of DP in eggs from the present study were in the same range as in eggs from Latvia (Zacs et al., 2021). SCCPs could not be detected in chicken eggs purchased from different suppliers from Belgium (LOQ: 0.5 ng/g ww), while MCCP concentrations were around ten times lower than concentrations in eggs from the present study (McGrath et al., 2021, Table 3). Concentrations of CPs in soil from an e-waste dismantling location in China were 200 times higher than the highest SCCP concentration found in the present study, and over three orders of magnitude higher compared to the highest MCCP concentration measured in the present study (Xu et al., 2019). In background soil samples collected from Norway and the UK in 2008, average concentrations of SCCPs were more than ten times lower than the lowest concentration measured in the present study (Halse et al., 2015). Soil samples collected at rural sites outside Dar es Salaam (Table 3) showed CP concentrations in the same range as samples collected in the present study from the historical dumpsite, and lower concentrations of DP (Nipen et al., 2022a). This supports the hypothesis that wastes containing CPs and dechloranes have not readily been discarded at the historical dumpsite, and that soil from this location does not represent a major contaminant source to household chickens foraging in the area.

Avian maternal transfer of CPs is lower compared to other POPs, such as PCBs and DDT (Choo et al., 2020; Guan et al., 2020; Knudtzon et al., 2021). However, the effects of CP chain length, chlorine content and chlorine distribution/positioning on bioaccumulation and maternal transfer are still not well understood (Bettina et al., 2011; Mézière et al., 2021). After dietary exposure to five technical mixtures, the maternal transfer of CPs with both low and high chlorine content from feed to chicken eggs was favored for SCCPs and MCCPs, whereas lower

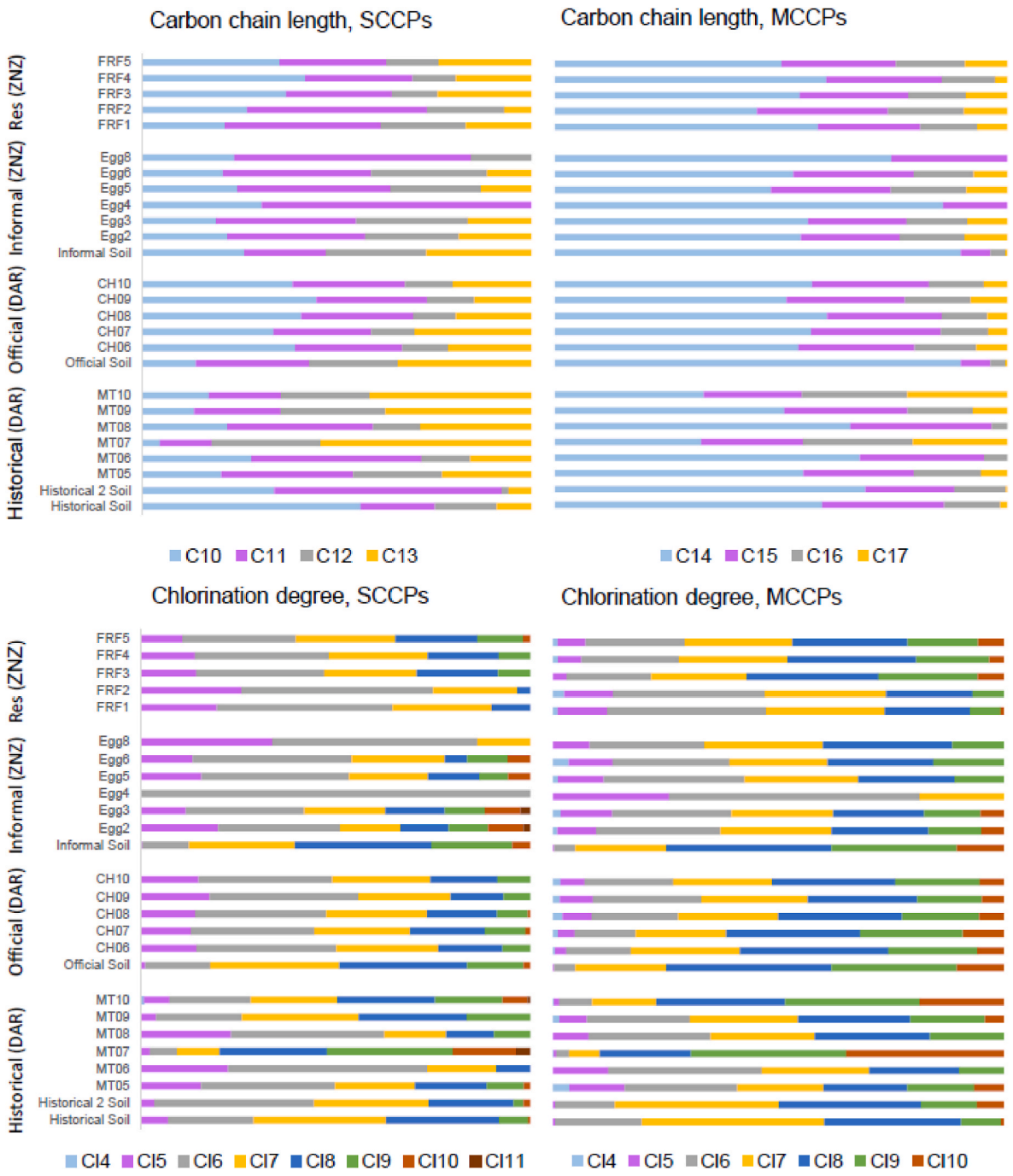

Fig. 4. Congener group profiles for chlorinated paraffins (SCCPs and MCCPs) in chicken egg and soil collected around various waste disposal sites on mainland Tanzania and Zanzibar.

chlorinated congeners were transferred for long chained chlorinated paraffins (LCCPs) (Mézière et al., 2021). Maternal transfer of CPs in watersnake, Enhydris chinensis, (egg to muscle ratio) decreased with increasing log Kow (ranging from log Kow 5 to 10) (Guan et al., 2020). Contrary, egg to liver ratio of CPs in the black-spotted frog, Pelophylax nigromaculatus, increased with log Kow and carbon chain length, suggesting that maternal transfer of CPs is related to the lipophilicity of the chemical. In addition, maternal transfer and chlorine content had a parabolic relationship, where egg to liver ratio increased up to chlorine content of 6–7, and then decreased as chlorine content increased further (Du et al., 2019). This parabolic relationship was also found for PBDEs in the rice frog, Rana limnocharis, with maternal transfer ratio peaking at 6–7 bromine atoms and log Kow at around 8 (Wu et al., 2009). A study by Zheng et al. (2018) also suggests highest maternal transfer rate for mid-halogenated POPs with log Kow of 6.5–7 in the Kentish Plover, Charadrius alexandrines. This seems in accordance with the present study, where CPs with 5–8 chlorine atoms and log Kow values around 6.5–9 were dominating in the egg samples. Dechlorane Plus is a relatively large molecule with a log Kow value above 9, and a high potential for maternal transfer in chickens compared to other organohalogen contaminants, including PBDEs and PCBs. However, the mechanisms for maternal transfer of DP are not clear (Li et al., 2021), and cannot be assessed further in the present study.

The different sampling times of egg and soil pairs (collected up to one year apart) from each location could affect the lack of correspondence in egg and soil concentrations in the present study. However, this is

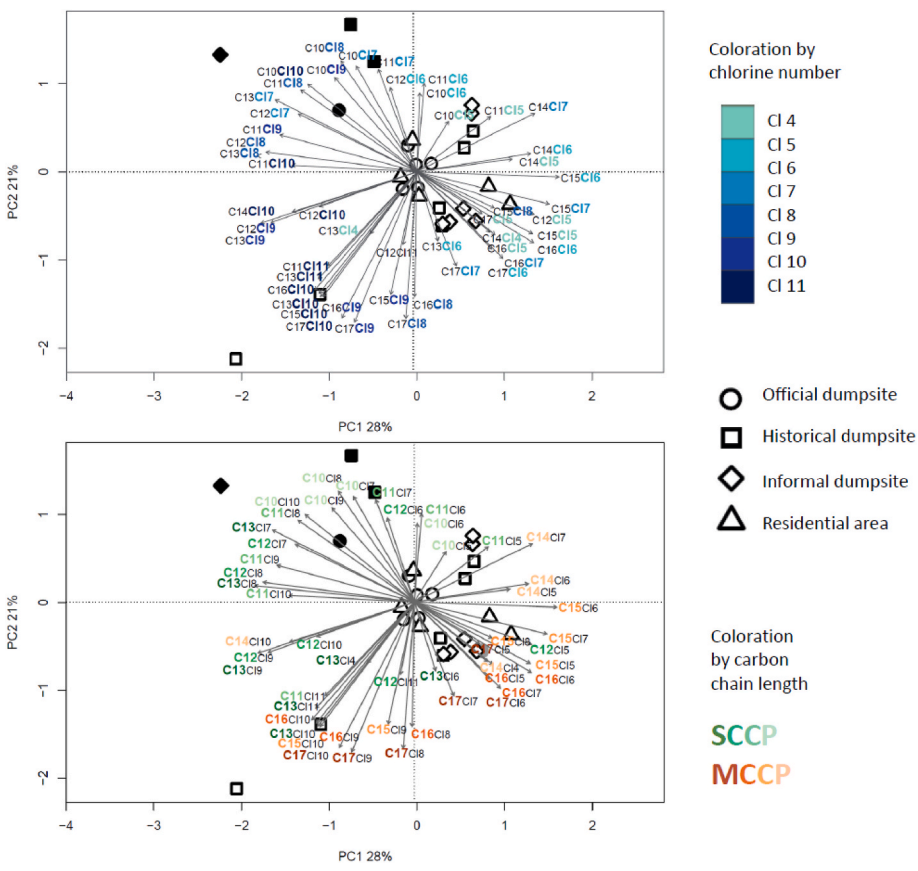

Fig. 5. Biplots of congener group profiles of chlorinated paraffins in eggs and soil from dumpsites on mainland Tanzania and Zanzibar. The different locations are shown as different symbols. Open symbols represent eggs, while filled symbols represent soil. Carbon chain length and chlorination degree of the different congener groups are highlighted in the two plots, respectively. Coloration of congener groups with light to darker blue with increasing chlorine number (above), and by carbon chain length (below) with darker green or orange for increasing chain length of short-chain and medium-chain CPs, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 3
Concentrations of CPs and dechloranes in chicken egg and soil reported from other studies. Concentrations are given in ng/g (different units used for normalization (lipid weight, SOM) is annotated). Mean (median) and range is reported when given in reference.

Location	n	Matrix	SCCP	MCCP	Dech Plus (DP)	Unit	Reference
Tanzania	1 (pooled)	egg	599 (–)	NA		lw	Petrlik et al. (2021)
China (e-waste)	30	egg	10,100 (1490) 611-111,000	6830 (999) 297-91,100		lw	Zeng et al. (2018)
Belgium	10	egg	ND	- (10) 3.3–16		ww	McGrath et al. (2021)
China (e-waste)	33	egg			- (–) 665–3290	lw	Zheng et al. (2012)
Latvia	8	egg			0.03	ww	Zacs et al. (2021)
China (e-waste)	7	soil	43000 (2300) 69-2,2x105	841000 (22600) 507-4,4x106		dw	Xu et al. (2019)
Norway & UK	58	soil	35 (–) <0.6–570	NA		ng/g SOM	Halse et al. (2015)
Tanzania (waste transect)	9	soil	670 (40) 11–5300	966 (160) <19-5100	0.9 (0.1) <0.01–6	dw	Nipen et al. (2022a)
Tanzania (rural transect)	6	soil	31 (58) <11-120	19 (19) <19-210	0,04 (0,04) 0,03–0,07	dw	Nipen et al. (2022a)

probably more relevant in regions where organisms experience large seasonal variation in lipid content (Warner et al., 2019). In addition to soil, industrial feed is also considered a significant source of CPs to domestic chickens (Dong et al., 2019). Future studies should include analyses of industrial feed, in addition to soil, to assess its importance as a source of CPs and dechloranes to domestic animals.

5. Conclusion

This study is one of very few studies to quantify levels of CPs and dechloranes in biota from Sub-Saharan Africa. A spatial shift in contaminant sources is expected and stresses the need for increasing the monitoring- and research efforts to developing regions in the Global South. Our results show higher levels of MCCPs compared to SCCPs in eggs, while SCCPs were dominating in two out of three soil samples. The detected levels of dechloranes were up to three orders of magnitude lower compared to CPs with DP as the dominant compound. Further, our results indicate that MCCPs congeners with lower chlorine content (5–8 chlorine atoms) seem to accumulate more readily in eggs compared to soil. This might be due to dechlorination of MCCPs in the hen or that larger congeners with higher log Kow are less bioavailable or more readily eliminated prior to maternal transfer. This would need to be confirmed by comparing the congener profiles in the eggs with profiles in other tissues from the mother, which is beyond the scope of the present study. Based on results from the present study, no clear conclusion can be made regarding waste and e-waste as a source of CPs

and dechloranes to the Tanzanian environment. Rather, results indicate that CPs and dechloranes are ubiquitous in the environment as a result of large-scale production and lack of regulations. Risk assessment of CPs shows that consumption of eggs from free-range chickens could represent a health concern regarding exposure to SCCPs, which warrants further research on the occurrence and sources of these chemicals in this region.

Credit author statement

Ane Haarr: Conceptualization, Investigation, Formal analysis, Visualization, writing – original draft preparation. **Maja Nipen:** Investigation, Resources – contaminant analyses, Writing - review and editing. **Eliezer B. Mwakalapa:** Investigation, Writing - review and editing. **Anders R. Borgen:** Resources – contaminant analyses, Writing - review and editing **Aviti J. Mmochi:** Resources – provision of laboratory facilities in Tanzania, Writing - review and editing **Katrine Borgå:** Supervision, Project administration, Conceptualization, 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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2023.138646.

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