



Increasing Trends of Legacy and Emerging Organic Contaminants in a Dated Sediment Core From East-Africa

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Temporal trends of industrial organic contaminants can show how environmental burdens respond to changes in production, regulation, and other anthropogenic and environmental factors. Numerous studies have documented such trends from the Northern Hemisphere, while there is very limited data in the literature from sub-Saharan Africa. We hypothesized that the temporal trends of legacy and contemporary industrial contaminants in sub-Saharan Africa could greatly differ from the regions in which many of these chemicals were initially produced and more extensively used. For this purpose, a dated sediment core covering six decades from a floodplain system in urban Dar es Salaam, Tanzania, was analysed. The samples were analysed for selected legacy persistent organic pollutants (POPs) [polychlorinated biphenyls (PCBs) and polybrominated biphenyl ethers (PBDEs)] and chemicals of emerging concern (CECs) [alternative brominated flame retardants (aBFRs), chlorinated paraffins (CPs), and dechloranes]. All groups of chemicals showed a steep increase in concentrations towards the uppermost sediment layers reflecting the more recent years. Concentrations of the individual compound groups in surface sediment were found in the order CPs >> aBFRs ~ \sum_{25} PBDEs > dechloranes ~ \sum_{32} PCBs. Time trends for the individual compounds and compound groups differed, with \sum_{32} PCBs showing presence in sediments since at least the early 1960s, while some CECs first occurred in sediments corresponding to the last decade. Investigations into potential drivers for the observed trends showed that socioeconomic factors related to growth in population, economy, and waste generation have contributed to increasing concentrations of PBDEs, aBFRs, CPs, and Dechlorane Plus. Further monitoring of temporal trends of industrial organic contaminants in urban areas in the Global South is recommended.

Keywords: temporal trends, urban environment, PCBs, PBDEs, flame retardants, dechloranes, chlorinated paraffins

INTRODUCTION

Legacy persistent organic pollutants (POPs), like polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs), have physicochemical properties which makes them persistent, bioaccumulative, toxic, and have a potential for long-range environmental transport. Some chemicals of emerging concern (CECs), like alternative brominated flame-retardants (aBFRs), chlorinated paraffins (CPs), and dechloranes, also share some of these properties. Time trends for production of these contaminants vary. PCBs and CPs have been produced since the 1930s, Dechlorane Plus (DP) from the 1960s, and PBDEs from the 1970s, while some aBFRs were not produced before the 1980s and 1990s (Breivik et al., 2002; Hoh et al., 2006; Covaci et al., 2011; Glüge et al., 2016; Abbasi et al., 2019). International regulation ensured an end to production of PCBs by the 1990s and PBDEs by the 2010s (Breivik et al., 2002; UNEP, 2017; Abbasi et al., 2019), while the CECs largely remain unregulated at an international level and production remains high or is increasing (Glüge et al., 2016; Xiong et al., 2019; Hansen et al., 2020). Both legacy POPs and CECs have been and are still used extensively in consumer products with different functions and user lifetimes. PCBs were applied in long-life electrical equipment, such as capacitors and transformers (Voogt and Brinkman, 1989), while PBDEs, aBFRs, CPs, and dechloranes have been, or still are used as flame retardants in e.g., electronics, building materials, and textiles. Many of the contemporary products containing CECs have shorter use lifetimes. Emissions to the environment may occur during production, but also during use of products containing legacy POPs and CECs, and from their disposal and recycling when these products reach their end of life.

Different histories for legacy POPs and CECs in terms of production, use, regulation, and waste disposal are thus likely to be reflected in long-term time trends of contaminant burdens in the physical environment. Research on time trends of these contaminants is therefore important to assess the effects of regulation and other emission-reducing measures, and to identify compounds that may merit further attention. Most studies on long-term temporal trends of POPs and CECs are from the Northern Hemisphere, where most of these chemicals have been more extensively produced and used. For instance, PCBs were mainly produced and/or used in North America, Europe, and Japan (Breivik et al., 2002). In these regions, environmental burdens peak around the time of peak production (Bogdal et al., 2008; Arp et al., 2011). Monitoring of atmospheric concentrations have documented declining primary emissions in recent decades (Schuster et al., 2010; Hung et al., 2016; Halvorsen et al., 2021; Wong et al., 2021).

However, elevated atmospheric concentrations of PCBs in some developing and newly industrialized regions in the Global South, including sub-Saharan Africa, indicate ongoing emissions (Klánová et al., 2009; Gioia et al., 2011). Unfortunately, studies on environmental levels of legacy POPs and CECs from sub-Saharan Africa are limited, particularly involving monitoring of long-term time trends (UNEP, 2021). The population and consumption is growing in urban areas in sub-Saharan Africa, though there is a low capacity for environmentally sound waste



FIGURE 1 | Sampling location for sediment cores marked with arrow. Map from Google Earth.

and e-waste handling. These regions are thus conceived to receive disproportionately large burdens of emissions at the end of the lifetime of products containing legacy POPs and CECs (Hoornweg and Bhada-Tata, 2012). Furthermore, elevated ambient temperatures may lead to enhanced emission of semi-volatile POPs and CECs from products in use and from waste (Breivik et al., 2011). These factors are likely to result in diverging long-term time trends compared to temperate regions in the industrialized Northern Hemisphere, indicating a need for further research in sub-Saharan Africa.

Studies assessing time trends are often based on monitoring of atmospheric levels. However, assessments of long-term time trends require monitoring over decades, yet atmospheric monitoring programs have only recently included CECs (Wong et al., 2021). Instead, sequentially settled and undisturbed sediments in e.g., fluvial floodplain systems may serve as archives documenting how levels of persistent contaminants in the physical environment have changed in response to changing emissions. This is possible as sediments function as a sink for legacy POPs and CECs, and individual

sediment layers can be dated using radioisotopes (Korosi et al., 2015). The collection and analysis of sediment cores is therefore a relevant technique to assess long-term temporal trends of both legacy POPs and CECs in regions where long-term monitoring has been lacking.

The main objective of this study is to describe long-term temporal trends of selected legacy POPs and CECs in a dated sediment core from a sub-Saharan African urban region. Dar es Salaam, Tanzania, was chosen as the study area. This city has experienced growing consumption and population yet suffers from a limited capacity for handling municipal waste and e-waste (Yhdego, 2017; Mahenge et al., 2018). PCBs, PBDEs, aBFRs, CPs, and dechloranes were selected as target analytes as they represent divergent histories related to production, use, and international regulation. None of the selected chemicals are known to have been produced in the region. Their long-term trends in the dated sediment core are compared with results from similar studies from other parts of the world, with particular attention to potential differences between industrialized and developing regions. Finally, the likely socioeconomic and biogeochemical drivers affecting the observed trends are assessed.

MATERIALS AND METHODS

Sampling Site

An urban area was targeted for sampling as the aim was to assess contaminants from local sources rather than contaminants from e.g., long-range transport. Sediment cores were thus collected from a tributary to the Msimbazi River in Tabata ward in Dar es Salaam (6° 49' 17" S, 39° 13' 29" E) (Figure 1) in February 2019. The size of the tributary river watershed is approximately 30 km². Soil textures within the catchment are mainly sand and sandy clay (Igulu and Mshiu, 2020) while the sampled section of the riverbed mainly consisted of clay. A floodplain area was chosen for sampling since such terrains favour gravitational settling. The sediments are also less prone to be eroded due to the slow water movement (Heim and Schwarzbauer, 2013). Dar es Salaam has a wet tropical climate, with an annual rainfall of more than 1000 mm, mainly occurring during two rain seasons; the "long rains" in April-May and the "short rains" in October-November. Within the watershed, sources of legacy POPs and CECs to sediments are expected to include surface runoff, untreated wastewater, atmospheric deposition, and seepage from unsolicited scattered dumping of domestic waste.

Sample Collection

Two sediment cores, one for dating and one for legacy POPs and CECs analysis, were collected within ~30 cm of each other using the inner tube (inner diameter 65 mm) of a gravity corer. Both cores were 39 cm in length, of which the top 15 cm were cut in 1 cm layers, while the lower sections were cut into 2 cm layers. Sectioning was done on-site, and slicing tools were rinsed in stream water between each slicing. Sediment layers were placed in pre-cleaned glass containers with aluminium foil-lined lids, transported in cooling bags, and subsequently stored in freezer after arrival at the laboratory.

Dating of Sediments

Layers from the sediment core collected for dating were shipped to GEL Laboratories, Charleston SC, United States, for radioisotope analysis and dating. ²¹⁰Pb was determined by Alpha spectrometric analysis of its granddaughter in the ²³⁸U series, ²¹⁰Po, which provide more accurate estimates of ²¹⁰Pb compared to its direct analysis. ²²⁶Ra and ¹³⁷Cs were determined using gamma spectrometry (Kim and Burnett, 1986). Sedimentation rate was modelled based on ²¹⁰Pb that was not from the decay of ²²⁶Ra in the sediments (i.e., excess ²¹⁰Pb) using the constant rate of supply (CRS) model and the constant flux: constant sedimentation (CF:CS) model as detailed in Appleby and Oldfield (1992).

Sample Preparation and Instrumental Analysis

Methods used for sample preparation and chemical analysis are detailed in the supplementary information (SI) and previously described for soils in Nipen et al. (2021). Only a summary is provided here. Approximately 2 g of wet sediment from each layer of the sediment core collected for legacy POPs and CECs analysis were dried and homogenized by mixing with sodium sulphate in a mortar. ¹³C labelled internal standards were added to each sediment layer before accelerated solvent extraction (ASE) using acetone/n-hexane (1:1). Approximately 2 g of activated copper powder was added to the extraction cells to remove elemental sulphur. Extracts were volume reduced, before concentrated sulphuric acid was added to remove lipids and other acid-labile potential instrumental interferences, followed by column chromatography clean-up with activated silica eluted with diethyl ether/n-hexane (1:9). The solvent was changed to isooctane, and volume reduced to 100 µl using a gentle stream of N₂. A recovery standard of 1,2,3,4-tetrachloronaphthalene was added before instrumental analysis.

Sediment concentrations of 32 PCBs, 25 PBDEs, and 14 aBFRs were determined using gas chromatography high-resolution mass spectrometry (GC-HRMS) (AutoSpec, Waters, Manchester, United Kingdom). Concentrations of 34 short chain CPs (SCCPs) congener groups, 28 medium chain CPs (MCCPs) congener groups, and nine dechlorane compounds were analysed using gas chromatography quadrupole time-of-flight high-resolution mass spectrometry (GC/Q-TOF) (Agilent, Santa Clara, United States). CPs were quantified using a pattern deconvolution procedure (Bogdal et al., 2015). A full inventory of congeners, congener groups, and acronyms used in the text is provided in **Supplementary Table S1**.

Water and organic matter (OM) content in sediment layers were determined gravimetrically by loss on drying at 110°C and loss on ignition (LOI) at 550°C, respectively. OM was determined by LOI followed by correction for loss of structural water in clay (Hoogsteen et al., 2015). Water content, which varied from 31% in the lowest sediment layer to 75% in the top layer, was used to calculate dry weight (dw) concentrations and bulk density.

Quality Assurance

All glassware were baked overnight at 450°C and rinsed with acetone and n-hexane before use. Components for ASE cells were sonicated in acetone twice, followed by sonication in n-hexane. High purity grade solvents were used throughout laboratory procedures. Field blanks ($N = 3$) and method blanks ($N = 5$) were analysed along with the sediment samples. Average blank levels were typically low, except for DBDPE, SCCPs, and MCCPs. High concentrations of these compounds in sediment samples ensured average blank concentrations only represented 6, 9, and 4% of average detected sample concentrations, respectively. Detailed information on blank levels is provided in **Supplementary Table S2**. Sample concentrations were blank corrected based on field and method blanks run with each sample batch. No significant difference was detected between field and method blanks ($p < 0.05$), so both were used to determine method detection limits (LODs). LODs were determined as three times the standard deviation of the blanks normalized to the average sample amount. LODs ranged between 0.0028 and 0.069 ng/g dw for PCBs, 0.0002 and 0.19 ng/g dw for PBDEs, and 0.0011 and 0.043 ng/g dw for aBFRs other than DBDPE. DBDPE had a higher detection limit of 1.7 ng/g dw. LOD for both SCCPs and MCCPs were 22 ng/g dw. No dechloranes were detected in blanks. Instrument detection limits, which ranged from 0.0041 to 0.083 ng/g dw, were therefore used for these compounds. For PCBs, PBDEs, and aBFRs two isotope m/z ratios were analysed for each compound, and results were to be excluded if the ratio diverged from the theoretical isotope ratio by more than 20%. However, no diverging ratios were found in samples with concentrations above the LODs.

Recoveries of internal standards were acceptable for the methods used. They varied between $48 \pm 13\%$ and $59 \pm 6\%$ for PCBs, $29 \pm 7\%$ and $51 \pm 10\%$ for PBDEs, $35 \pm 11\%$ and $48 \pm 11\%$ for aBFRs, $79 \pm 15\%$ and $90 \pm 21\%$ for dechloranes, and $71 \pm 18\%$ for CPs. Samples with internal standard recovery $<15\%$ were re-analysed. All legacy POPs and CECs were quantified using the isotope dilution principle, hence intrinsically correcting for recovery. The standard reference material SRM 1944 of an urban marine sediment, from the National Institute for Standards and Technology, was analysed for PCBs and PBDEs with $92 \pm 14\%$ and $89 \pm 14\%$ compliance, respectively.

RESULTS AND DISCUSSION

Sediment Core Dating

The sediment layers contained low activities of excess ^{210}Pb , rendering core dating challenging. However, a reasonably good exponential decay of ^{210}Pb was measured in a mid-section of the core (8–20 cm depth). For this section, the sediment accumulation rate (SAR) was determined to be 0.27 ± 0.07 cm/year using the CRS model. This corresponds well with SAR determined using the CF:CS model (0.18 ± 0.03 cm/year). Dates for the top- and mid-section of the core were thus assigned based on the rate of sedimentation

(CRS model) in the mid-section of the core, covering the time interval 1962 to 2019. Sediment layers below the mid-section of the core did not show exponential decay of ^{210}Pb . This section could therefore not be dated with any confidence and is thus referred to as pre-1962. Activities of ^{137}Cs were generally below or near detection limits and could thus not confirm the dating.

Concentrations and Time Trends of Legacy POPs and CECs

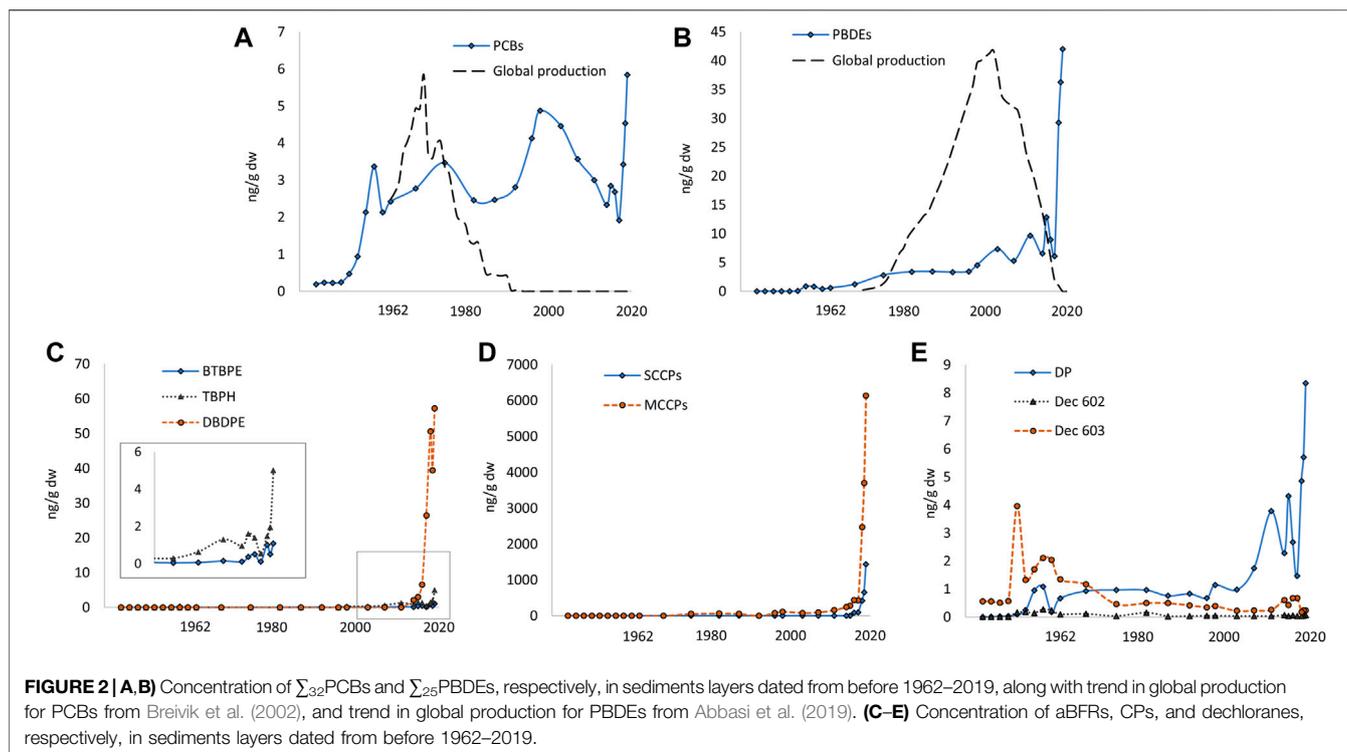
Temporal trends in concentrations of $\sum_{32}\text{PCBs}$, $\sum_{25}\text{PBDEs}$, aBFRs, CPs, and dechloranes with estimated age of sediment layers are shown in **Figure 2**. Concentrations and congener profiles in each sediment layer are provided in **Supplementary Table S6** and **Supplementary Figures S4–S8**. All analysed PCBs and PBDEs congeners, and both SCCPs and MCCPs, were detected in one or more sediment layer. Of the 14 analysed aBFRs, the following were detected in one or more sediment layers: DBDPE, BTBPE, TBPH, ATE, a-TBECH, b-TBECH, g/d-TBECH, HBB, and PBBZ. Of these, only three (DBDPE, BTBPE, and TBPH) were detected in notable amounts (concentrations >0.1 ng/g dw) and with discernible time trends and are therefore included in further discussions. Of the nine analysed dechloranes, only DP (syn- and anti-isomers), Dec 602, and Dec 603 were detected. Overall, the highest concentrations were found for CPs, followed by aBFRs and $\sum_{25}\text{PBDEs}$ with concentrations two orders of magnitude lower than CPs, and dechloranes and $\sum_{32}\text{PCBs}$ with concentrations three orders of magnitude lower than CPs.

Most of the legacy POPs and CECs showed steep increases in concentrations in sediment layers corresponding to the late 2010s. This trend was particularly notable for the CECs, with CPs and aBFRs showing 10–15 times increase from sediment layers corresponding to 2016/2017 to 2019. Of the dechloranes, DP increased five times in the same layers, but the levels of Dec 602 and Dec 603 remained relatively constant. Of the legacy POPs, $\sum_{25}\text{PBDEs}$ concentrations increased seven times, while $\sum_{32}\text{PCBs}$ concentrations tripled. Although the increase in $\sum_{32}\text{PCBs}$ in the late 2010s was small compared to some CECs, it is notable considering that the global production of PCBs peaked five decades ago (Breivik et al., 2002).

Tables 1, 2 lists the approximate year of peak concentration in sediment cores from this study and other studies for comparison, organized by region (developing and newly industrialized countries (**Table 1**) and industrialized countries (**Table 2**)). Temporal trends for the compound groups are discussed below, followed by a discussion of potential drivers for the observed increasing trends.

Legacy POPs

$\sum_{32}\text{PCB}$ concentrations varied between 0.19 ng/g dw in the deepest sediment layer and 5.8 ng/g dw in the surface sediment layer (**Figure 2A**). Subsurface peaks of $\sum_{32}\text{PCBs}$ were found in layers corresponding to pre-1962 (3.4 ng/g dw), the 1970s (3.5 ng/g dw), and the late-1990s (4.9 ng/g dw). The dominating congeners in the sediments were PCB-138 (on



average 15%), PCB-153 (13%), PCB-149 (9.3%), PCB-101 (8.3%), PCB-118 (7.8%), PCB-180 (7.4%), PCB-28 (4.2%), and PCB-31 (3.0%). Individual PCB congeners had temporal trends which differed from each other (Illustrated for PCB-28 and PCB-180 in **Figure 3**). Concentrations of PCB-118, PCB-138, and PCB-180 peaked in subsurface layers, and the increase between 2017 and 2019 was less than factor 2. Lower chlorinated (e.g., PCB-28 and PCB-52) and higher chlorinated (e.g., PCB-206 and PCB-209) congeners peaked instead in the upper layers of the core, where concentrations increased by factors between 3 and 5 from layers corresponding to 2017–2019. The contribution of higher chlorinated congeners to \sum_{32} PCB was however minor (**Supplementary Figure S4**). Lower chlorinated congeners are less persistent, and they reached peak production later compared to higher chlorinated congeners (Breivik et al., 2002). Both these factors may affect comparative time trends.

The temporal trend in \sum_{32} PCBs (**Figure 2A**) in the studied sediment core was not consistent with trends found in sediment cores from industrialized countries. The PCBs loading in industrialized regions generally peak around 1970 (**Table 2**), reflecting the trends in global PCBs production (**Figure 2A**). In Tanzania, PCBs were mainly used in closed systems (e.g., electrical transformers) (IPEN, 2005) where emissions are likely to occur during or after decommissioning. This, in combination with long use-lives of transformers, may explain the comparatively low peak in PCBs concentrations observed in the 1970s followed by a higher peak in the late 1990s (**Figure 2A**). In this study, as in several other studies from developing and newly industrialized regions (**Table 1**), the highest PCBs concentrations are found in upper most recent

layers of sediments. We attribute this to emissions from products and waste containing PCBs, causing delayed emissions compared to industrialized regions. For example, a Chinese study reported a bimodal peak in PCBs in a dated sediment core, with the first peak corresponding to the 1970s, and the second (higher) peak to the 2000s. These peaks were explained by PCBs production and e-waste processing, respectively (Yang H. et al., 2012).

\sum_{25} PBDE concentrations ranged from below LOD in the deeper sediment layers to 42 ng/g dw in the surface layer (**Figure 2B**). Concentrations increased from the 1970s to around 3 ng/g dw in the 1980s, where it remained relatively constant until it increased again from the 2000s. From its first detection onwards, BDE-209, the main congener of the decaBDE mixture, dominated the \sum_{25} PBDE concentrations, comprising on average 81% of the total (range 71–90%). The relative contribution of BDE-209 increased with sediment depth ($p < 0.05$). This was an unexpected trend considering that penta- and octaBDE were regulated and phased out of production a decade earlier than decaBDE (Abbasi et al., 2019). Time trends for congeners associated with pentaBDE mixtures and octaBDE mixtures were relatively similar (**Supplementary Figure S5**). \sum_{25} PBDEs and \sum_{32} PCBs were strongly correlated ($p < 0.001$), and several individual PBDE and PCB congeners were also correlated (**Supplementary Table S7**). PBDEs are not known to have been produced before around 1970. Nevertheless, several PBDE congeners were detected in pre-1962 layers, though at low levels. Similar findings have been reported for PBDEs in sediment cores collected in China, the UK, and North America, with

TABLE 1 | Approximate year of peak concentration of PCBs, PBDEs, aBFRs, CPs, and dechloranes in the studied sediment core and in sediment cores from developing and newly industrialized regions, compiled from literature.

Location	System	PCBs	PBDEs	aBFRs	SCCPs	MCCPs	DP	Dec 602	Dec 603	Ref
Dar es Salaam, Tanzania	River	<i>2019</i>	<i>2019</i>	<i>2019</i>	<i>2019</i>	<i>2019</i>	<i>2019</i>	<1962	<1962	This study
Nador Lagoon, Morocco	Lagoon	<i>2009</i>								Giuliani et al. (2015)
Lake Victoria, Uganda	Lake	1986								Arinaitwe et al. (2016)
Lake Bujuku and Mahoma, Uganda	Lakes	<i>2003</i>								Arinaitwe et al. (2016)
East China Sea, China	Marine	2000								Yang et al. (2012a)
Bay of Manila, Philippines	Marine	<i>2009</i>	<i>2009</i>							Kwan et al. (2014)
Gulf of Thailand	Marine	1965	1995							Kwan et al. (2014)
Lake Chapala, Mexico	Lake	<i>2010</i>	<i>2010</i>							Ontiveros-Cuadras et al. (2019)
Nhat Le and Cua Dai, Vietnam	Estuaries	1968	<i>2015</i>							Tham et al. (2020)
East China Sea, Yellow Sea, China	Marine		<i>2010</i>							Li et al. (2016)
Thane Creek, Mumbai, India	Estuary		2005							Tiwari et al. (2018)
12 Chinese lakes	Lakes		<i>2006</i>	<i>2006</i>						Wu et al. (2012)
Dongjiang, China	River		sub-surface	surface						Zhang et al. (2009)
Daya Bay, China	Marine		2005	<i>2012</i>			<i>2012</i>			Liu et al. (2014)
Pearl River estuary, China	Estuary/mangrove			surface						Zhang et al. (2015)
Dongjiang, China	River						sub-surface			He et al. (2014)
Yellow Sea, China	Marine						<i>2010</i>			Wang et al. (2017)
East China Seas	Marine						1980			Wang et al. (2016)
Pearl River estuary, China	Estuary/mangrove						surface			Sun et al. (2016)
Beijing, China	Lake				surface					Zeng et al. (2011)
Dongjiang, China	River				surface	surface				Chen et al. (2011)
Nine Chinese lakes	Lakes				<i>2019</i>	<i>2019</i>				Zhang et al. (2019)
South China Sea, China	Marine				<i>2004</i>	<i>2004</i>				Zeng et al. (2017)
Yellow Sea, China	Marine				2005	2005				Zeng et al. (2013)

Years in italic indicate peak concentration in surface layer of the sediment core.

PBDEs detected in sediments dated to the 1960s and earlier (Wu et al., 2012; Liu et al., 2014; Yang et al., 2016; Da et al., 2019; Guo et al., 2020). The reason for detection of PBDEs in sediment layers corresponding to dates prior to its production is unknown. Potential explanations include contamination during core extrusion, uncertainty in dating of the older layers, or bioturbation of the sediments. These factors are also of relevance to the other analysed compound groups.

According to sediment core data from Western Europe, the environmental levels of PBDEs levelled off or decreased from the late 1990s and early 2000s (Bogdal et al., 2008; Vauclin et al., 2021). On the other hand, some studies from other industrialized countries conducted in the early 2000s and 2010s, documented increasing sediment concentrations of PBDEs (Table 2). This may reflect the continued presence of PBDEs in the anthroposphere. Contrary to flattening trends of PBDEs in Western Europe, and similarly to our study, Ontiveros-Cuadras et al. (2019) found increasing PBDEs concentrations in sediment cores from Mexico (Table 1). Li et al. (2016) also found increasing trends of PBDEs in sediment cores from the East China Sea. They explain this with shifts in industrial production towards China and import of waste containing PBDEs. The recent steep increase in PBDEs is

therefore likely best explained by increasing disposal and recycling of waste products containing PBDEs.

CECs

The concentrations of DBDPE, BTBPE and TBPH ranged from <1.7 to 57, <0.01 to 1.1, and <0.06–5.0 ng/g dw, respectively (Figure 2C). DBDPE was first detected in sediment layers corresponding to 2014 and increased up to 57 ng/g dw in the surface layer. Studies on sediment cores from North America (Yang R. et al., 2012), Japan (Hoang et al., 2021), and China (Zhang et al., 2009; Wu et al., 2012; Zhang et al., 2015) show similar increasing concentrations of DBDPE (Tables 1, 2), though DBDPE was generally detected in layers down to deposits from the 1980s or 1990s. BTBPE was detectable in the sediment down to layers corresponding to the early 1980s, though concentrations increased from the late 1990s, and reached the highest concentration in layers corresponding to the late 2010s. Sediment cores from North America show BTBPE concentrations increasing from the 1970s, and levelling off between the mid-1980s and the early 2000s (Hoh et al., 2005; Yang R. et al., 2012). In a study from China, however, BTBPE concentrations were generally found to continue increasing to the upper sediment layers (Zhang et al., 2015). TBPH was detected down to sediment layers corresponding to the late 1990s. Concentrations

TABLE 2 | Approximate year of peak concentration of PCBs, PBDEs, aBFRs, CPs, and dechloranes in sediment cores from regions which have remained industrialized for an extended time, compiled from literature.

Location	System	PCBs	PBDEs	aBFRs	SCCPs	MCCPs	DP	Dec 602	Dec 603	Ref
Baltic Sea, Sweden	Marine				1940	<i>2016</i>				Yuan et al. (2017)
Tokyo Bay, Japan	Marine				1990	1990				Zeng et al. (2017)
Lake Ontario, North America	Lake				1970					Marvin et al. (2003)
Lake Thun, Switzerland	Lake	1969			1986	<i>2004</i>				Iozza et al. (2008)
Oslo Harbour, Norway	Marine	1970								Arp et al. (2011)
Beppu Bay, Japan	Marine	1970								Takahashi et al. (2020)
Greifensee, Switzerland	Lake	1960								Zennegg et al. (2007)
Adriatic Sea, Italy	Marine	1985								Combi et al. (2016)
Rhone, France	River	1990								Desmet et al. (2012)
Seine, France	River	1970								Dendievel et al. (2020)
Five lakes, Texas, USA	Lakes/ reservoirs	1970								Van Metre and Mahler (2004)
Lake Superior, North America	Lake	1985	<i>2002</i>							Song et al. (2004)
Lake Thun, Switzerland	Lake	1965	<i>2004</i>							Bogdal et al. (2008)
Rhone, France	River			1995						Vauclin et al. (2021)
Drammensfjord, Norway	Marine			1999						Zegers et al. (2003)
Wadden Sea, Netherlands	Marine			1989						Zegers et al. (2003)
Lake Woserin, Germany	Lake			1990						Zegers et al. (2003)
Lake Erie, North America	Lake			1999						Zhu and Hites, (2005)
Lake Michigan, USA	Lake			1995						Zhu and Hites, (2005)
Sidney estuary, Australia	Estuary		<i>2014</i>							Drage et al. (2015)
Lake Ontario, North America	Lake		<i>2004</i>				1994			Qiu et al. (2007)
Lake Michigan, North America	Lake						1976			Hoh et al. (2006)
Lake Erie, North America	Lake						sub- surface			Sverko et al. (2008)
Five Great Lakes, North America	Lakes						1980	1990 ^a	1990 ^a	Yang et al. (2011)
Lake Ontario, North America	Lake						1980	1995	1980	Shen et al. (2011)
Lake Ontario, North America	Lake						1980	1980	1990	Shen et al. (2010b)
Three Great Lakes, North America	Lakes			2000	<i>2010</i>		1980			Guo et al. (2020)
Lake Michigan, North America	Lake		1995	1986						Hoh et al. (2005)
Five Great Lakes, North America	Lakes			<i>2007</i>						Yang et al. (2012b)
Beppu Bay, Japan	Marine			<i>2011</i>						Hoang et al. (2021)

Years in italic indicate peak concentration in surface layer of the sediment core.

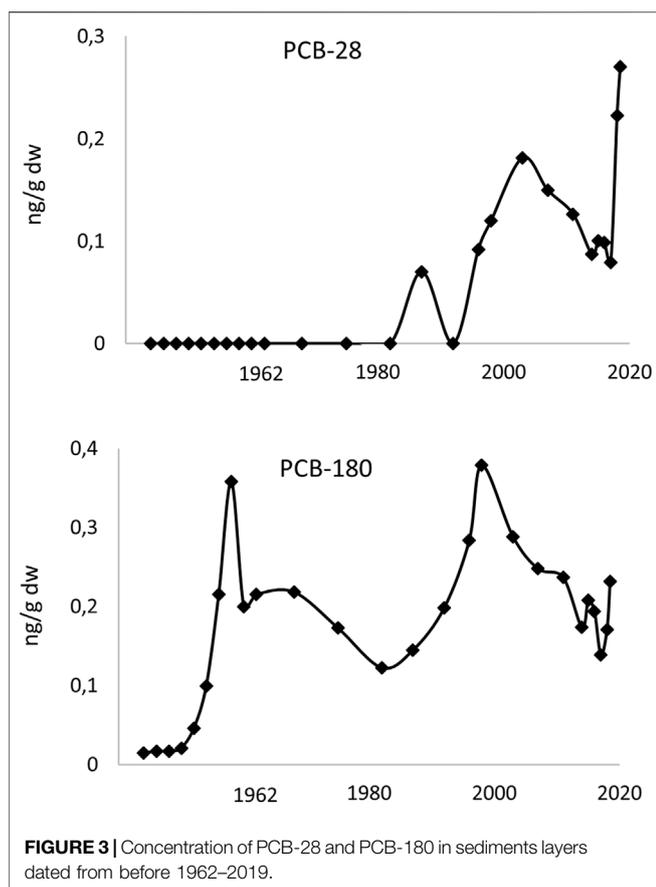
^aDec 602 and Dec 603 reported together.

remain relatively constant between 1 and 2 ng/g dw through the 2010s and increase to 5 ng/g dw in the surface sediment layer. A study from South China reported similar increasing concentrations of TBPH in sediments (Liu et al., 2014). The delayed appearance of DBDPE, BTBPE, and TBPH in the sediments from Dar es Salaam, compared to studies from North America and China, is likely due to differences in industrial activity. DBDPE, BTBPE, and TBPH all correlated strongly with \sum_{25} PBDEs and individual PBDEs congeners. BTBPE and TBPH also showed strong correlations to \sum_{32} PCBs ($p < 0.001$) (Supplementary Table S7).

MCCPs dominated over SCCPs with concentrations varying from <22 to 6,200 and to 1,400 ng/g dw, respectively. The highest CPs concentrations were found in the surface sediment layer, with strongest increase occurring between 2017 and 2019 (Figure 2D). MCCPs were detected in all sediment layers corresponding to the mid-1990s onwards, while SCCPs were first detected in layers corresponding to the mid-2010s. Time trends of SCCPs and MCCPs were strongly correlated to \sum_{25} PBDEs and aBFRs ($p < 0.001$) (Supplementary Table S7). In sediment cores from Western Europe, SCCPs concentrations peaked around the 1980s and 1990s, while MCCPs generally

show continued increasing trends (Iozza et al., 2008; Yuan et al., 2017). In sediment cores from China, which has sustained high CPs production, both SCCPs and MCCPs concentrations in general show the highest concentrations in surface layers (Zeng et al., 2012; Zhang et al., 2019) (Tables 1, 2). There is no known production of CPs in Tanzania, so the continued increasing trend is likely connected to the increased use and disposal of CPs containing products, possibly in combination with increased applications of CPs in metal industry. Elevated levels of CPs have been measured in air and soil in and around Dar es Salaam. Spatial trends suggested that municipal waste and e-waste were important sources (Nipen et al., 2021). Surface sediment concentrations of SCCPs and MCCPs were comparable to or higher than the highest soil concentration found in urban areas (1600 and 3200 ng/g dw, respectively, (Nipen et al., 2021). Moreover, congener group profiles of CPs in sediments were more comparable to urban soil than to urban air (SI Text 2).

DP was the dominant dechlorane compound in the sediment (Figure 2E), with concentrations ranging from <0.02 to 8.4 ng/g dw. Dec 602 and Dec 603 concentrations ranged from <0.01 to



0.27, and 0.18–4.0 ng/g dw, respectively. DP was detected in pre-1962 layers and remained stable at around 1 ng/g dw until the early 2000s. From the mid-2000s, concentrations fluctuated over a general increase to peak concentration in the surface layer. As far as we are aware, this study is the first to report sediment core based long-term time trends for dechloranes from outside regions where they are produced (the North American Great Lakes region and China). In the Great Lakes region, DP concentrations have generally increased from the 1960s or 1970s, usually peaking in the 1980s or 1990s, although some sediment cores showed continued increasing trends into the upper layers (Qiu et al., 2007; Shen et al., 2010b; Shen et al., 2011; Yang et al., 2011) (Table 2). In China, the vertical profile of DP in sediments show increasing trends towards the surface layers (Table 1). Dec 603 concentration in the studied sediment peaked in layers corresponding to pre-1962 (Figure 2E), followed by comparatively low concentrations in subsequent sediment layers. In sediment cores from the Great Lakes region, Dec 603 increased from the 1950s or 1960s, and peaked around the 1980s or 1990s (Shen et al., 2010b; Shen et al., 2011) (Table 2). Shen et al. (2010a) describe Dec 603 as an impurity in the historically used pesticides Aldrin and Dieldrin. They observed spatial trends of Dec 603 in sediments in the Great Lakes corresponding to pesticide use. Historic pesticide use is also a plausible explanation for the trends for Dec 603 in this study (SI Text 3). The levels of Dec 602 also peaked in pre-1962 sediment

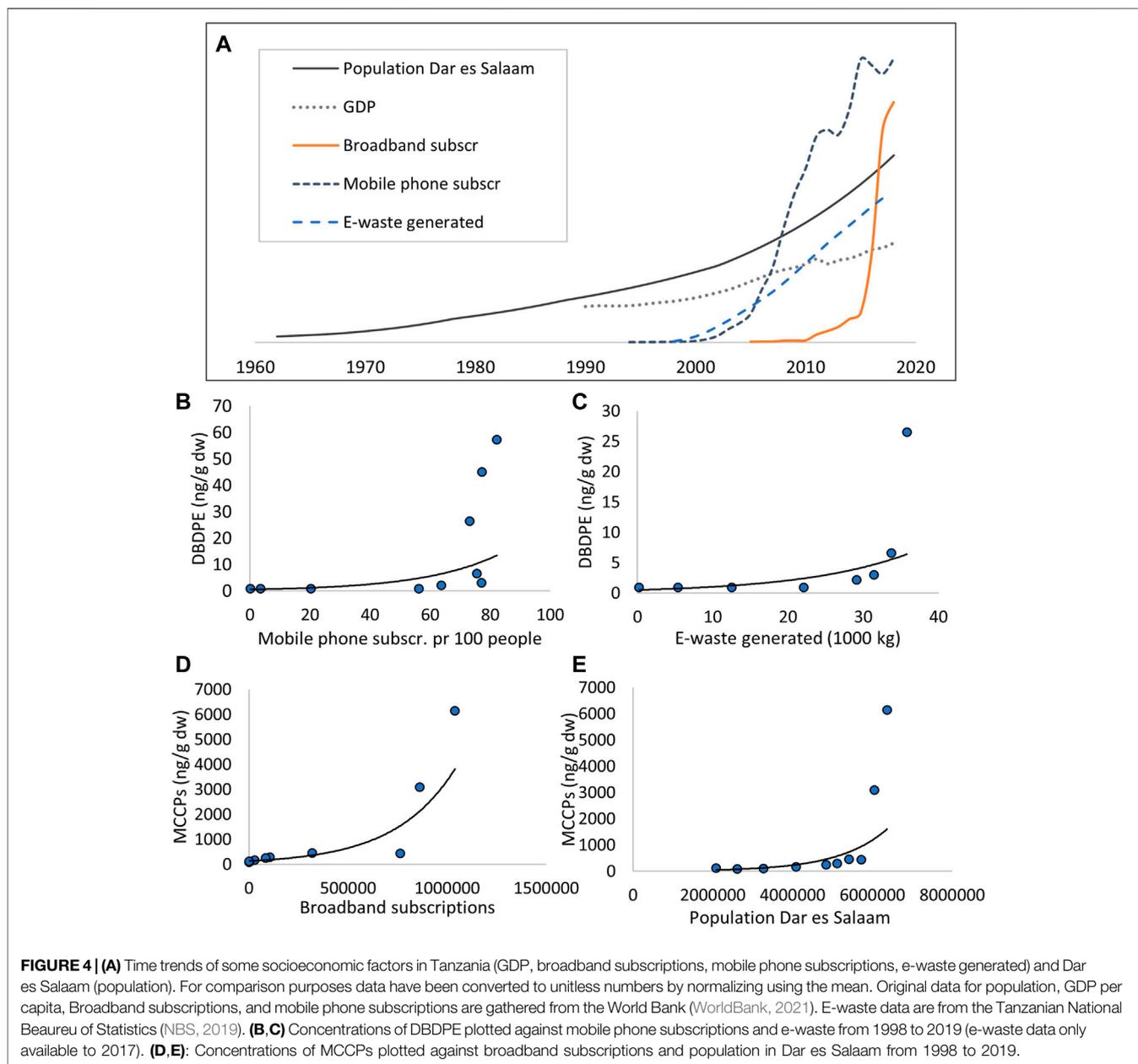
layers (Figure 2E). The Dec 602 peak was followed by declining concentrations, with stable low concentrations around or just below 0.05 ng/g dw from the mid-1980s up to the surface sediment layer. In sediment cores from the Great Lakes region, Dec 602 concentrations increased from the 1960s or 1970s and peaked around the 1980s and 1990s (Shen et al., 2010b; Shen et al., 2011). Dec 602 only showed a significant correlation with Dec 603 ($p < 0.01$), while correlations between Dec 603 and other CECs and legacy POPs were generally negative. Although DP showed strong positive correlations with both the legacy POPs ($\sum_{32}\text{PCBs}$ and $\sum_{25}\text{PBDEs}$) and the other CECs (aBFRs and CPs) ($p < 0.001$ for all), the continued presence of DP in sediments over decades demonstrate that it is not a “new” contaminant in this region.

Spatial trends and elevated levels of DP in air and soil in Dar es Salaam suggest e-waste as an important source (Nipen et al., 2021). The levels of DP in the surface sediment layer were higher than the highest concentrations found in urban soil (5.0 ng/g dw). Dec 602 and Dec 603 concentrations in surface sediment (0.07 and 0.2 ng/g dw, respectively) were comparable to the highest soil concentrations found in urban areas (0.05 and 0.3 ng/g dw, respectively). Moreover, the study by Nipen et al. suggested that the Dec 603 sources were of a historic nature, which is consistent with the findings in this study.

Potential Drivers for Time Trends of Legacy POPs and CECs

There are several conceptual explanations for the recent increasing trends in POPs and CECs, both in terms of the socioeconomic development in the region and due to shifts in environmental pressures on biogeochemical processes.

Growth in urban population and especially in per capita Gross Domestic Product (GDP) (Figure 4A) imply growth in consumption and general waste generation (Hoornweg and Bhada-Tata, 2012). Further, the very strong growth in mobile and broadband subscriptions, along with the amount of e-waste generated in the region, show that consumption has been particularly strong for electronics during the last decade. Strong correlations were found between these socioeconomic parameters and sediment concentrations of $\sum_{25}\text{PBDEs}$, aBFRs, DP, and CPs ($p < 0.001$). Dec 602, Dec 603, and $\sum_{32}\text{PCBs}$ were not correlated with any of the socioeconomic parameters, although some individual congeners (PCB-28 and PCB-206, $p < 0.001$) were (Supplementary Table S8). Strong temporal correlations between socioeconomic parameters and the level of flame retardants (PBDEs, aBFRs, DP, along with CPs) in the sediment layers, suggest that increases in consumption and waste generation of consumer goods containing these additives contribute to the increase in these pollutants during the late 2010s. PCB-28 and PCB-206 have been reported to occur as unintentional by-products in pigment production (Anh et al., 2020). Consequently, use and disposal of pigment-containing consumer products may be a contributing factor for the increasing trend for these congeners in the 2010s. These findings suggest that



inadequate handling and recycling of waste has led to increasing pollutant levels in the urban environment.

The exponential rather than linear relationship between CECs and socioeconomic factors (**Figures 4B–E**) suggests that additional factors may contribute synergistically to increasing concentrations. Elevated concentrations of organic pollutants in sediment layers may be due to erosion of contaminated topsoil or other contaminated surface reservoirs within the watershed caused by intense rainfall and flooding events (Lu et al., 2016). Correlations of rainfall versus legacy POPs and CECs were investigated for two different time intervals (1983–2018 and 2010–2018), using rainfall data from the Tanzanian Meteorological Authority (TMA, 2021). The longer and shorter time intervals were chosen based on the likelihood of

legacy POPs and CECs being present in topsoil, respectively. No significant correlations were found for the time interval 1983–2018 (**Supplementary Table S9**). For the time interval 2010 to 2018, the rainfall episodes between 2010 and 2012 were less intense, around average between 2013 and 2016, and stronger than average in 2017 and 2018. For the time interval 2010 to 2018, CPs and DBDPE were significantly correlated with rainfall ($p < 0.05$ and $p < 0.01$, respectively). Similarities in CPs congener group profiles in soil from urban areas and sediment, as well as comparable concentrations, support that increased erosion of contaminated soil has contributed to the increasing trend of CPs in sediments.

Strong associations are expected between OM content and the hydrophobic legacy POPs and CECs (**Supplementary Figure**

S11). OM content in the sediment varied between 1% in the lowest sediment layer to 9% in the top layer. The OM content was strongly positively correlated to sediment concentration of most PCB and PBDE congeners, \sum_{32} PCBs, \sum_{25} PBDE, TBPH, BTBPE, DP, and MCCPs ($p < 0.001$). OM content was also significantly correlated to PCB-209, DBDPE, and SCCPs ($p < 0.05$), while there were no or negative correlations between OM and Dec 602 and Dec 603. However, the main features of trends of legacy POPs and CECs, including strong increases in the late 2010s and subsurface peaks for PCBs, remain when concentrations are normalized to OM content (**Supplementary Figure S12**). If OM content was the main explanatory factor governing the time trends, there would have been a “smoothing out” of the concentration differences between the sediment layers. The trend in OM content is therefore not likely to be the main driver for the observed temporal trends.

Overall, these findings suggest that the increasing trends in legacy POPs and CECs in sediment layers corresponding to the late 2010s are mainly a consequence of increased primary emissions from products and waste in connection with the rapid socioeconomic development in the region. However, secondary emissions in the form of remobilized surface reservoirs of legacy POPs and CECs are likely to have contributed to the increasing trends, particularly for CPs and DBDPE.

CONCLUDING REMARKS

This study showed that concentrations of both legacy POPs and CECs, particularly CPs and DBDPE, have increased notably in sediment layers corresponding to the late 2010s. Other studies from industrialized regions, as well as other developing and newly industrialized regions, in general also show increasing trends for these CECs. This suggests that particular attention should be given to these compounds in future monitoring studies, both regionally and internationally. Comparisons of time trends of legacy POPs showed that the flattening and declining trends seen in most industrialized regions, as a response to regulation and emission controls, are not reflected in the studied region, nor in most other developing and newly industrialized regions. This underlines the importance of improved emissions controls in waste handling and recycling in these regions. This should be further prioritized at a regional level, but also in international regulatory efforts relating to trade in products, used products, and waste that contain these compounds.

More studies of sediment cores are needed to further document historical emission trends in developing and newly

industrialized regions. Monitoring of atmospheric concentrations of legacy POPs and CECs in urban areas in these regions may also be an important additional asset to assess the effect of emissions reductions, as atmospheric concentrations respond more rapidly to changes in emissions.

DATA AVAILABILITY STATEMENT

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

AUTHOR CONTRIBUTIONS

MN, RV, PB-N, KBo, and KBr conceptualized the study, MN wrote the original draft, while RV, PB-N, KBo, EM, AB, MS, GC, AM, and KBr contributed to review and editing of the manuscript. Formal analysis was conducted by MN. Investigation was conducted by MN, EM, AB, and GC. Supervision was provided by RV, PB-N, KBo, MS, and KBr, and KBo was project administrator.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/fenvs.2021.805544/full#supplementary-material>

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