



Soil pollution at a major West African E-waste recycling site: Contamination pathways and implications for potential mitigation strategies



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ABSTRACT

Organic contaminants (polycyclic aromatic hydrocarbons (PAHs), polybrominated diphenyl ethers (PBDEs), polychlorinated biphenyls (PCBs), and chlorinated paraffins (CPs)) and heavy metals and metalloids (Ag, Cd, Co, Cr, Cu, Hg, Ni, Pb, Sb, Zn) were analysed in surface soil samples from the Agbogbloshie e-waste processing and dumping site in Accra (Ghana). In order to identify which of the pollutants are likely to be linked specifically to handling of e-waste, samples were also collected from the Kingdom general waste site in Freetown (Sierra Leone). The results were compared using principal component analyses (PCA). PBDE congeners found in technical octa-BDE mixtures, highly chlorinated PCBs and several heavy metals (Cu, Pb, Ni, Cd, Ag and Hg) showed elevated concentrations in the soils that are likely due to contamination by e-waste. PCAs associated those compounds with pyrogenic PAHs, suggesting that burning of e-waste, a common practice to isolate valuable metals, may cause this contamination. Moreover, other contamination pathways, especially incorporation of waste fragments into the soil, also appeared to play an important role in determining concentrations of some of the pollutants in the soil. Concentrations of several of these compounds were extremely high (especially PBDEs, heavy metals and SCCPs) and in some cases exceeded action guideline levels for soil. This indicates that exposure to these contaminants via the soil alone is potentially harmful to the recyclers and their families living on waste sites. Many organic contaminants and other exposure pathways such as inhalation are not yet included in such guidelines but may also be significant, given that deposition from the air following waste burning was identified as a major pollutant source.

1. Introduction

Large, informal recycling sites for electronic waste (e-waste) are located in a number of developing countries where environmental regulations are much less strict than in many developed countries. The e-waste processed at these sites often consists of the non-reusable fraction of large quantities of redundant electronic devices that are donated by developed countries. Electrical and electronic equipment (EEE) is known to contain large amounts of potentially toxic chemicals. This includes polybrominated diphenyl ethers (PBDEs) used as flame retardants in circuit boards, plastic casings and wire and cable insulation, polychlorinated biphenyls (PCBs) used as dielectric fluids, lubricants and coolants in older capacitors, transformers and generators, chlorinated paraffins (CPs) in wire and cable insulations, lead in printed circuit boards and cathode ray tubes (CRTs), chromium in anticorrosion coatings, cadmium in printed circuit boards, photocopying machines, CRTs and mobile phones, zinc and nickel in batteries, mercury in lamps, printed circuit boards and sensors (Perkins et al., 2014; Wang et al.,

2018). The waste is processed in very simple ways, such as dismantling with bare hands, open burning and acid digestion to extract valuable metals from EEE that contain often large amounts of plastic materials. These procedures combined with unsystematic and unprotected disposal of the remaining waste have become a large concern from an environmental pollution and human health perspective. Studies conducted at Chinese, Indian, Nigerian and Ghanaian e-waste sites found large concentrations of heavy metals and organic contaminants such as PBDEs, PCBs and polycyclic aromatic hydrocarbons (PAHs) in the air, in dust, soil, vegetation and in the blood of workers and residents on and in the vicinity of the sites (Adetunde et al., 2014; Isimekhai et al., 2017; Tue et al., 2016; Labunsk et al., 2013; Daso et al., 2016; Wang et al., 2012; Tang et al., 2010; Akortia et al., 2017; Amankwaa et al., 2017; Chakraborty et al., 2018; Zhang et al., 2012; Hogarh et al., 2018; Caravanos et al., 2011).

It has been estimated that 44,700 kt of e-waste were produced globally in 2016, with an expected growth rate of 3–4% per year (Baldé et al., 2017). This figure also includes potentially reusable items.

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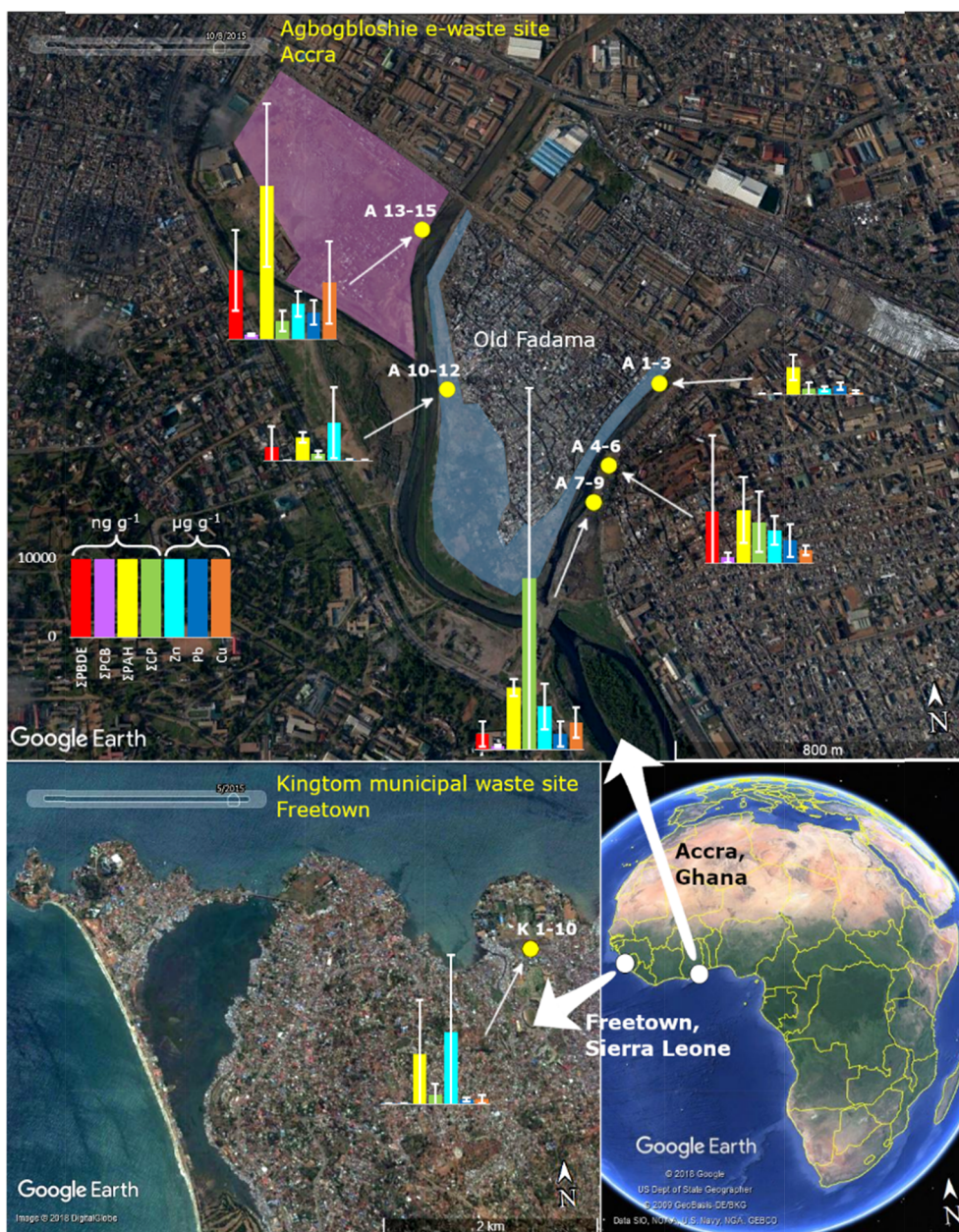


Fig. 1. Overview of dry soil weight-based concentrations of selected contaminants in waste site soils from Agbogbloshie (Ghana) and Kingtom (Sierra Leone). The scale of the y-axis in all graphs is the same as in the legend. Pink-shaded area: Agbogbloshie e-waste site, blue-shaded area: June 2015 eviction zone. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Around 70% of this waste is thought to have been exported to China in 2005, while India, Nigeria, Ghana, Ivory Coast, Benin and Liberia were also considered as major e-waste importing nations (Breivik et al., 2014). Abassi et al. (2019) estimated that over half of the used EEE imported to the major receiving regions is subjected to inappropriate (i.e. unsafe or uncontrolled) waste treatment (Abassi et al., 2019). According to Amoyaw-Osei et al. (2011), the Agbogbloshie e-waste site in Accra, Ghana, received 150.5 kt of used EEE generated within Ghana and in other (mostly developed) countries in 2009. About 15% of this could not be used anymore and was declared waste immediately (Amoyaw-Osei et al., 2011). Since this waste is processed on the site in the ways described above, its export to Ghana is in contravention of the Basel Convention on the control of transboundary movements of hazardous wastes and their disposal (de Sadeleer, 2016).

In developing countries without strict regulations, it can be assumed that especially domestic e-waste and industrial wastes are also disposed

of at ordinary municipal waste sites. One such site is the Kingtom dumpsite in Freetown, Sierra Leone, which receives approximately 740 tons per day. Of this, 84% is biodegradable organic waste from residential areas. Whilst e-waste may be present, it is thought that it comprises only a small percentage of the total. Importantly, this waste is not systematically burnt or acid-treated to isolate valuable metals.

Several studies have shown high concentrations of heavy metals, PBDEs or PAHs in soil and vegetation at Agbogbloshie (Daso et al., 2016; Akortia, et al., 2017; Fosu-Mensah et al., 2017; Oteng-Ababio et al., 2014). This is especially concerning since many of the workers and their families, including children and pregnant women, live on the site and are, therefore, constantly exposed to high amounts of these toxins. Not surprisingly, elevated blood levels were found in e-waste workers and other residents (Amankwaa et al., 2017; Wittsiepe et al., 2017), causing a health risk in addition to inadequate sanitation, hazards for physical injury and poverty in general. In order to recommend

measures that can reduce the pollution burden caused by e-waste handling, it is necessary to identify the site-specific sources and processes causing it.

The purpose of this study was to (i) assess environmental concentrations of PAHs, PBDEs, PCBs, chlorinated paraffins, and selected heavy metals in an international context, (ii) distinguish between pollution caused by e-waste and general waste, and identify e-waste-related processes that determine the pollutant concentrations and (iii) indicate the pollutants posing the highest risk to people living and working at the sites.

Soil was chosen as sample medium in this study because it has the capacity to store persistent pollutants that deposit to it. Soils, therefore, provide a time-integrated picture of the pollutant burdens without being heavily influenced by day-to-day differences in polluting activities such as burning of waste. Especially at waste sites, soils are also likely to be contaminated directly by pollutant-containing materials dumped there. People are exposed to pollutants in the soil by direct ingestion, inhalation of particles or outgassing compounds, and ingestion of products from animals that have been feeding on polluted soils. Contaminated soils may also act as an ongoing source of pollutants to the atmosphere. Moreover, guidelines are available for acceptable levels of many organic and inorganic pollutants in soil, depending on the intended land use.

2. Methods

2.1. Soil sampling

The present study included two different waste dumps for comparison: the Agbogbloshie e-waste site and the Kingtom domestic dumpsite. No protective lining to prevent leachates from entering surface water bodies and the groundwater has been installed at either site (Caravanos et al., 2011; Kallon 2008). At Agbogbloshie, in August 2015, a total of 15 soil samples (A-1 to A-15) were collected at five sites at the edge of settlements, partly on e-waste dumping grounds, where often livestock was found grazing. At each site, triplicates were collected ca. 1 m apart from each other. Samples A-1 to A-9 were taken along the southeastern edge of Old Fadama, the residential area immediately adjacent to the Agbogbloshie e-waste site. Samples A-13 to A-15 were collected within e-waste trading, dismantling, burning and dumping grounds, close to the Odaw River, and A-10 to A-12 from a former residential area, that was demolished in June 2015, within 160 m of the e-waste site (Fig. 1). Old Fadama itself is a slum and the adjacent river floodplains were also used for informal housing in addition to waste dumping (including e-waste) until the area was cleared after evictions two months before the sampling took place (Daum et al., 2017). At Kingtom a total of 10 soil samples (K-1 to K-10) were collected across the site, providing a broad coverage including areas where waste is processed or scavenged and close to residential areas. At both sites, a soil auger was used to collect soil to a depth of 10 cm.

2.2. Sample analysis and data processing

The soil samples were analysed for total organic carbon (TOC), PAHs, PCBs, PBDEs, chlorinated paraffins and heavy metals and metalloids according to established methods (see SI for details). The means of subsets were compared by Student's *t*-test. To investigate relationships between different compound (sub-)groups that may point at similar origins, principal component analysis (PCA) was carried out on the complete dataset and on subsets (as mentioned in the text) using XLSTAT. Details are provided in the SI.

3. Results and discussion

3.1. Pollutant concentrations at Agbogbloshie and Kingtom – Comparison to background, urban and e-waste site data worldwide

Concentrations of all chemicals analysed in the soil samples in our study are presented in Fig. 1 and Tab. SI-7–SI-11. For comparison, literature data from Agbogbloshie, other e-waste and general dump sites, urban and rural/background areas are also shown.

PAHs: The total concentration of the 16 USEPA PAHs (Σ_{16} PAHs, see Tab. SI-7) ranged from 130 to 5,200 ng/g in the Kingtom soil samples and from 850 to 10,000 ng/g in those from Agbogbloshie. Total concentrations of all PAHs measured (Σ PAH) were 260 to 14,000 ng/g in Kingtom and 1,300 to 16,000 ng/g in Agbogbloshie samples. In Agbogbloshie the lowest and highest Σ PAH concentrations were found in sample 12 and 14 respectively. The Σ_{16} PAHs concentrations found at both waste sites are similar to those found at other (e-)waste burning sites and in large cities, yet up to four magnitudes higher than concentrations measured in background soils (Tab. SI-7). This could be expected, possibly both as a result of waste burning and domestic burning at the waste sites and a wide variety of combustion processes in the nearby major cities of Freetown (ca. 1 056 000 inhabitants in 2015) and Accra (ca. 2 500 000 inhabitants in 2019). All samples show, according to Wilcke (2000) PAH profiles typical for tropical soils, with 3- and 4-ring PAHs dominating. However, with 41–81% vs. 12–44%, the contribution of high molecular weight (HMW, 4- to 7-ring) PAHs to total PAHs is significantly higher in the Agbogbloshie than in the Kingtom samples ($p = 10^{-6}$). This may indicate that pyrogenic sources play a larger role at Agbogbloshie than at Kingtom (Stogiannidis and Laane, 2015). To investigate this, several indicator ratios have been compared between the two waste sites. According to Katsoyiannis et al. (2011) and references therein, the results indicate that both types of sources have contributed to the burdens found, Ant/(Ant + Phe) suggests dominance of petrogenic sources at Kingtom while other ratios (BaA/(BaA + Chr), Flt/(Flt + Pyr), IDP/(IDP + BghiP) rather point at a contribution from both or dominance of pyrogenic sources. Comparison of the averages of each ratio between Kingtom and Agbogbloshie soils mostly supports the initial assumption that concentrations are affected more strongly by combustion in Agbogbloshie than in Kingtom. Except for Flt/(Flt + Pyr) all ratios at Kingtom are significantly smaller ($p < 0.003$), i.e. closer to values typical for petrogenic sources, than at Agbogbloshie. However, the ratios between methylated and parent PAHs (Phe and Ant), which would be expected to be higher where petrogenic sources play a larger role, are not significantly different between Kingtom and Agbogbloshie (see Tab. SI-12). For other e-waste sites, similar PAH profiles were reported (Leung et al., 2015; Wang et al., 2012; Tang et al., 2010; Wang et al., 2017), although in some cases pointing slightly more clearly at a domination of pyrogenic sources (Leung et al., 2015; Wang et al., 2017). Despite the relatively small differences in Σ PAH concentrations between Kingtom and Agbogbloshie, it is worth noting that total PAH-derived toxic equivalent concentrations (TEQ) (Nisbet and LagGoy, 1992) are significantly higher at Agbogbloshie than at Kingtom (230 vs. 41 ng/g on average, $p = 0.0021$) (see SI text and Tab. SI-13 for details). The PAH-related toxicity is therefore expected to be higher for the Agbogbloshie than for the Kingtom soils. Since evidence for pyrogenic sources was found we will use PAHs (especially high molecular weight ones) later as markers to assess burning of waste as possible emission source of other pollutants measured.

PBDEs: With concentrations ranging from 6.3 to 7,700 ng/g, Agbogbloshie samples showed significantly higher ($p = 0.010$) total PBDE concentrations than Kingtom samples where 1.2–100 ng/g were found. The concentrations at both sites are much higher than those reported for background areas in Kenya and Tanzania (Tab. SI-8). Reasons for this could be the influence of the waste dumps but also the vicinity to the cities of Freetown and Accra, respectively, where PBDEs

may be emitted to the atmosphere from various sources. PBDE concentrations and congener composition in Kingtom soils are similar to those found in urban areas around the world (see Tab. SI-8), whereas the burdens found in Agboglobshie soils are most comparable to other e-waste sites, e.g. at Guiyu, Taizhou and Qingyuan in China (Labunsk et al., 2013; Ma et al., 2009; Wang et al., 2015). In all but one Kingtom and one Agboglobshie sample where BDE-209 results are available, concentrations of this congener were higher than those of any other BDE congener or homologous group. The same was observed at many e-waste sites and in urban areas (see Tab. SI-8) although in a study of soils from the Agboglobshie site published earlier by Akortia et al. (2017), BDE-28 was found to be the most abundant congener. At Kingtom, on average, the next most abundant congener groups after BDE-209 are penta- and tetra-BDEs, followed by hexa- and nona-congeners. By contrast, in Agboglobshie soils, nona-BDEs are most abundant after BDE-209, followed by tetra and penta-congeners.

PCBs: Sum concentrations of all PCBs analysed ranged from 0.74 to 43 ng/g in Kingtom, significantly ($p = 0.003$) lower than in Agboglobshie (6.5–830 ng/g). Concentrations at both sites are elevated compared to the scarce background data available for this region (Tab. SI-9). The levels found in Kingtom are within the range of those in other urban areas of Ghana, Nigeria, South Africa and India (Tab. SI-9). Concentrations at Agboglobshie exceed many of those found in other cities and are more similar to the ranges that were observed at waste (recycling) sites in China (Tab. SI-9). The contribution of the different homologous groups to the total PCB concentration is very similar between Kingtom and Agboglobshie. With RSDs below 50% for the five most abundant homologous groups, the percentages are also much more consistent between individual samples than the absolute concentrations. Hexachlorinated PCBs are the most abundant homologue group and account for nearly 30% at both sides, followed by tri- and penta-congeners. This pattern differs from that observed for the 90°S – 30°N zone in a global study, and even more pronounced in the 30°N – 60°N zone where past PCB usage was highest (Meijer et al., 2003). As in our samples, hexa-CBs were also the most abundant homologue group there, but they were followed by penta- and hepta-CBs. This order of abundance was also observed in other studies of rural soils (Schuster et al., 2011; Gluge et al., 2016). Low molecular weight PCBs (including tri-CBs) showed very low concentrations in those studies, while they accounted on average for 24% of the total PCB burden in our soils both from Kingtom and Agboglobshie. A similar trend has been seen for instance at the e-waste recycling sites in Taizhou and Longtang, China (Liu et al., 2013; Tue et al., 2016), in urban and coastal soils of the central region of Ghana (Bentum et al., 2016) and in urban soils from Lagos, Nigeria (Alani et al., 2013). Soils from major Indian cities (Chakraborty et al., 2016) and suburban areas around Nairobi, Kenya (Sun et al., 2016) also showed similar tri-to-hepta -CB ratios as in our samples, but here tetra- and penta-CPs dominated, respectively.

S/MCCPs: SCCPs were found at concentrations between 69 and 1600 ng/g in the Kingtom and between 145 and 28,000 ng/g in the Agboglobshie samples, while MCCP concentrations ranged from < MDL to 1400 ng/g and < MDL to 1300 ng/g, respectively. Unlike in many soil samples from China (see Tab. SI-10), SCCP concentrations exceed MCCP concentrations in all samples in the current study except for one from Kingtom. Concentrations at both sites vary strongly with RSDs of up to 210% and neither the concentrations of SCCPs nor MCCPs are significantly different between Kingtom and Agboglobshie samples. Comparison with literature data on chlorinated paraffins in soils is difficult as data is still scarce and most studies have been conducted in China so far (Tab. SI-10). The lowest SCCP concentrations found in the Agboglobshie and Kingtom samples are similar to or only slightly higher than concentrations reported for ambient soils in Dalian, China and in some background soils from the UK and Norway (Xu et al., 2016; Halse et al., 2015). Urban and suburban soils from China often show similar concentrations (Tab. SI-10). The highest SCCP concentrations in Kingtom samples are similar to concentrations

reported in farmland soils irrigated with wastewater (Zeng et al., 2011) and in soils close to a plant where chlorinated paraffins are produced, whereas the highest concentrations found in Agboglobshie samples (above 2000 ng/g) were higher than most concentrations reported in soils to date. Only one study on soils collected at the Taizhou e-waste site in China in 2017 (Xu et al., 2019) found partly similar and partly almost one magnitude higher SCCP concentrations. MCCP concentrations found at both Agboglobshie and Kingtom were within ranges found in Chinese urban, suburban and ambient soils but over three magnitudes lower than those in the Taizhou e-waste site soils sampled by Xu et al. (2019) in 2017 (see Tab. SI-10). Our analysis suggests that the chlorine content of SCCP was higher in the Agboglobshie than in the Kingtom samples (see SI text under 6.3 for details). This may indicate that the SCCP at the two sites originated from different sources or applications.

Metals and metalloids: Soils from both Kingtom and Agboglobshie showed similar patterns of metal/metalloid concentrations with zinc being the most abundant element (5200 µg/g and 2300 µg/g on average), followed by copper, lead and chromium (on average 360 µg/g, 300 µg/g and 490 µg/g in Kingtom and 1500 µg/g, 1100 µg/g and 110 µg/g in Agboglobshie). These were also the most abundant metals in urban soils from Kumasi (Ghana) and Ibadan (Nigeria) (Darko et al., 2017; Odewande and Abimbola, 2008) but the concentrations of these and most other elements analysed in the current study exceeded the values reported for urban, suburban and solid waste dump site soils in West Africa (see Tab. SI-11). The naturally occurring heavy metal contents in soils in Ghana and Sierra Leone are very low (Sillanpää and Jansson, 1992) and can therefore be excluded as a reason for the very high zinc, lead and copper burdens found in the waste site soils. The average Pb, Cu and Hg concentrations were significantly higher at Agboglobshie ($p = 0.0065$, 0.049 and 0.0023) while chromium showed significantly higher levels at Kingtom ($p = 0.024$). The concentrations of the other metals analysed did on average not differ significantly between the two sites. Zinc concentrations as high as in Kingtom and Agboglobshie soils, and lead and copper concentrations high as those found in Agboglobshie are usually only found in areas affected by mining and e-waste recycling (see Tab. SI-11).

3.2. Possible contamination pathways - Agboglobshie (e-waste) vs. Kingtom (general waste)

Systematic burning of e-waste for the purpose of isolating valuable metals, is often considered to be a major source of many organic and inorganic environmental pollutants (Isimekhai et al., 2017; Tue et al., 2016; Labunsk et al., 2013; Wang et al., 2012). The spatial distribution of pollutant concentrations in the soils may provide information as to how strongly the immediate vicinity of the e-waste dismantling and burning areas and the Old Fadama area where many e-waste workers and their families live, are affected by e-waste burning-related pollution. Comparison with the data from the Kingtom site will help to distinguish pollution related to e-waste handling and dumping from that caused by dumping of general waste.

Compared to many other urban and suburban soils, both the general waste site at Kingtom and the Agboglobshie e-waste site soils show elevated concentrations of PAHs, PBDEs, CPs, Cu, Cd, Pb, Zn. Especially concentrations of HMW-PAHs, PBDEs, PCBs, Pb and Cu are significantly higher at Agboglobshie than at Kingtom ($p \leq 0.025$) and individual Agboglobshie samples contained remarkably high levels of SCCPs. This suggests that a large fraction of the soil burden of these compounds is related to e-waste (processing), while LMW-PAHs, and the remaining metals/metalloids may to a large extent be related to waste processing in general and/or other sources found in urban areas, such as traffic, various industries and domestic burning or chemical usage.

For a more detailed investigation of the link between e-waste burning and the concentrations of heavy metals and persistent organic pollutants in the soil, data from the two different types of waste

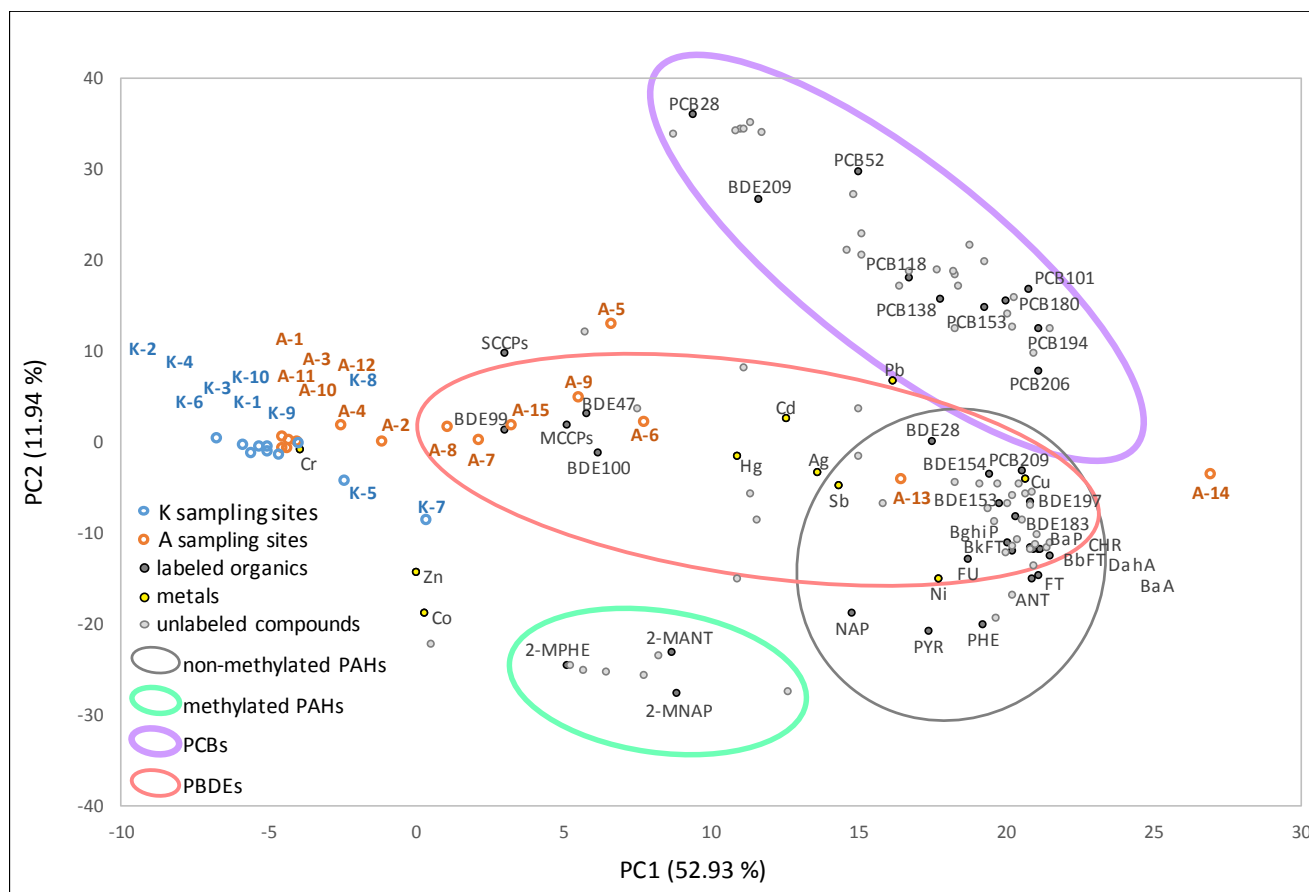


Fig. 2. PCA on all samples and including all compounds analysed. See Fig. SI-3 for labels on all compounds, including those unlabeled here.

recycling/dump sites were included in principal component analyses (see [Supplementary Information](#) for details). One aim was to differentiate between the effects of general waste management and e-waste recycling because there are unique processes associated with the handling of e-waste and more emphasis could be given to how specific activities could become cleaner, if they appear to cause a large proportion of the pollution. When carrying out a PCA on all samples and including all compounds analysed (Fig. 2) as well as for individual groups of chemicals (Fig. SI-5 - SI-9), all Kingtom (K-) samples form a relatively tight cluster while the Agboglobshie (A-) samples spread widely. This reflects the large variations in analyte concentrations within the Agboglobshie sample set compared to the Kingtom samples, and the higher pollutant burdens found at Agboglobshie compared to Kingtom for many samples.

Despite the close proximity between all the Kingtom and all the Agboglobshie sampling sites, there are large variations in the concentrations of most contaminants analysed, even between the triplicate samples taken at each Agboglobshie sampling site. Relative standard deviations within both sites were around 100% for PAHs, PCBs, metals and metalloids. Larger variations (partly above 200%) were found for PBDEs and chlorinated paraffins in both datasets and for Cu in the Agboglobshie set. This reflects the very heterogeneous nature of the soils, possibly with numerous small-scale hotspots. Before assessing possible contaminant sources, the soil organic carbon (SOC) content was tested to see if it could explain a considerable portion of this variation. Such a relationship should exist if airborne compounds were transferred to the soil mainly via vegetation, or if compounds evaporate again from the soil after they deposited. Normalising for SOC did not bring the triplicates closer to each other in a PCA, suggesting that the analyte concentrations did not depend primarily on the SOC content (Fig. SI-4). The discussion of possible contaminant sources will

therefore focus on soil dry weight-based concentrations. The amounts of pollutants humans are exposed to via the soil depends on those concentrations, in addition to the activities they carry out.

The following section discusses pollutant concentration patterns with regards to the main factors and processes identified as likely to affect and determine these concentrations: (i) contamination related to open burning of waste, (ii) incorporation of waste (fragments) into the soil and leaching of pollutants out of waste materials, (iii) deposition of compounds that have evaporated from sources on site or outside of the waste site (without burning), and (iv) volatilization, possibly resulting in transport to other areas.

3.2.1. Contamination related to open burning of waste

The Kingtom site in Freetown receives general waste mainly from Freetown, which will include some electronic products and batteries. It is likely that some accidental burning occurs due to the heat and landfill gasses generated during degradation of organic waste materials, and occasionally the waste is burned intentionally, but unlike at Agboglobshie this is not done in a systematic way. The Agboglobshie e-waste recycling site receives large amounts of electronic waste, while general waste from Accra is also taken to this site. Unlike at Kingtom, electronic products that cannot (at least partly) be re-used are burned or acid-treated systematically to isolate valuable metals. During incomplete burning processes, typically PAHs are produced, especially non-alkylated ones, whereas methylated PAHs are rather indicators of petrogenic sources. Therefore, to assess if the variation of any compound group can be linked to waste treatment by burning we carried out PCAs for PAHs alone and for each individual compound group combined with PAHs.

A large fraction (85%) of the variation of PAH concentrations in the soil samples is explained by the first two PCs (Fig. SI-5). Especially

typical pyrogenic PAHs score highly positively on PC1, hence we can assume that this PC is linked to pyrogenic processes. They are little affected by PC2, where the more volatile LMW PAHs show positive values while the less volatile HMW PAHs score negatively and form a tight cluster on the plot. In PC1 direction, most of the K-samples group on the negative side of the plot whereas A-samples show a wide variation with some samples having highly positive loadings, indicating that pyrogenic sources contribute more strongly to the soil PAH concentrations at Agbogbloshe than at Kingtom. Fig. SI-5 suggests that alkylated PAHs were not linked to pyrogenic PAHs. This could be expected as they originate from petrogenic sources, e.g. petroleum-based fuels and oils and coal tars.

In a PCA in combination with PAHs (Fig. SI-10, including both A- and K-sites), technical octa-mixture (t-octa) PBDE congeners (i.e. hexa- to octa-BDEs) group with pyrogenic PAHs and score highly on PC1. This is even more pronounced when including A-sites only (compare Fig. SI-11 and SI-12). Technical penta-mixture (t-penta) PBDE congeners (i.e. tri- to penta-BDEs) separate from more highly brominated PBDEs and occupy a different space on the PCA plot than any of the PAHs. Their vectors are orthogonal to those of petrogenic PAHs, indicating that there is no link between those two subgroups. This may be due to the different applications the technical PBDE mixtures were used for. T-octa mixtures were more commonly used in plastic cases of electronic products than t-penta mixtures which are mostly found in soft furniture and textiles. This explains their high contribution to the total PBDE content both in the A-samples of the present study and in soils from other e-waste sites (see Tab. SI-8). Nona-brominated congeners may also have formed by debromination of BDE-209 during the burning of waste electronic products which have been shown to contain large amounts of this congener (UNEP, 2017). Generally, PC1 and PC2 do not explain a large proportion of the variation of the concentrations of those more volatile PBDEs. They are much better explained by a third PC, which plays no role for PAHs. These observations suggest that while the levels of t-octa congeners in the soils appear to be linked to burning activities, those of t-penta congeners may be driven by other processes. The lack of a complete overlap between t-octa PBDEs and PAHs especially in the Agbogbloshe PCA plot supports the theory that not all fires producing PAHs also release PBDEs and/or that a fraction of the PBDEs found in the soil is not linked to burning. The former can be expected when materials (including waste) that do not contain PBDEs are burned. The latter may be a result of other possible PBDE sources as identified below.

A similar behaviour can be seen for PCBs. In a PCA combined with PAHs (Fig. SI-13), PCBs found in more high-chlorinated technical mixtures such as Aroclors 1254 and 1260 score closer to pyrogenic PAHs than lighter congeners that are found in less highly chlorinated mixtures. Aroclors 1254 and 1260 have been used extensively in electrical equipment and cable insulations and may be released when such products are burned. Nearly orthogonal vectors between low-chlorinated PCBs and pyrogenic PAHs suggest that burning activities are not the dominating source for the more volatile PCBs. When all or only A-samples are included, there is no overlap between PCBs and PAHs except for PCB-209, indicating that apart from burning there are also other sources that affect concentrations of most PCB congeners. Apart from the release of existing PCBs they may also form during burning when both chlorine containing plastics such as PVC and materials acting as catalysts, e.g. copper, are present. However, unlike in other studies (Chakraborty et al., 2018), the congeners that have been linked to this process, such as dioxin-like PCBs 105, 114, 118, 123, 156, 157, 167 and 189, do not show elevated relative concentrations compared to Aroclor 1254 for instance. This suggests that, although formation of PCBs may occur, it is not likely to be a major source.

CPs show no strong link to pyrogenic PAHs. In a PCA with PAHs (Fig. SI-14) they score in a similar direction as pyrogenic PAHs but their vectors are much shorter than those of PAHs, indicating that CPs are not well explained by the first two PCs which explain the variation of PAHs

well. This suggests that burning of waste affects CP concentrations very little, both at Kingtom and Agbogbloshe. This picture remains when individual SCCP congener groups (C₁₁ with five to ten chlorine) are tentatively included in the PCA (see SI for details). The relative abundance of the different congener groups varies between technical SCCP mixtures with different overall chlorine contents (Yuan et al., 2017). Our results may therefore indicate that contamination pathways other than burning played a dominating role for all SCCPs in this study, even if they originated from technical mixtures with different chlorine contents.

A PCA that includes only metals and PAHs at all sites (Fig. SI-15) suggests that some of the metals/metalloids, namely Cu, Ni, Cd, Sb, Pb, Ag and Hg, are emitted during burning as they score close to pyrogenic PAHs. In contrast, Zn, Co and Cr do not appear to be related to those PAHs and may, therefore, not originate from the same sources. Compared to the Kingtom samples, the Agbogbloshe samples load highly in the pyrogenic PAH direction, especially A-13 and A-14. We can therefore conclude that burning of waste, including e-waste, is a significant contributor to the heavy metal burdens in Agbogbloshe soils, while it does not seem to play an important role at Kingtom.

3.2.2. Incorporation of waste fragments into the soil and leaching

Soils may contain fine particles or larger fragments originating from discarded, unburned waste materials that contain PBDEs, or dust that adhered to products containing PBDEs. Plastic particles may be shed during handling and processing of the waste, or as a result of aging of the materials, especially in the case of polymer foams. Whilst larger, easily visible plastic fragments were removed from the samples in this study, smaller particles could neither be distinguished nor removed from the soil. The potential presence of fine plastic particles and dust previously adhering to plastic products may explain the large variations in concentrations even between samples taken very close to each other. Indeed, two of the four samples with the highest sum(tri-nona) BDE concentrations have the highest tetra- and penta-BDE concentrations of all soil samples (A-9 and A-12) and load far from pyrogenic PAHs on the PCA plot. The other two (A-13 and A-14) have the highest hexa-, hepta- and nona-BDE concentrations of all soil samples and load close to pyrogenic PAHs on the PCA plot (Fig. SI-10). Waste fragments incorporated into the soil could explain both the large variation in CP concentrations between samples collected close to each other and the lack of a correlation between SCCP and MCCP concentrations as commonly only one of the two mixtures is added to products.

Zn, Co and Cr, the metals that dominate in the K-samples and reach higher concentrations than in the A-samples, show no indication of being linked to burning. For Zn and Co, which are mostly associated with batteries (Zn in zinc-carbon and Co in Li-ion batteries) that can be found in household waste, leaching out of the waste may be the main contamination pathway. For Cr, which is found in anticorrosion coatings, incorporation of waste particles into the soil is also likely to play a role. Acid treatment of waste, which can result in the release of heavy metals, was not considered separately in this study because samples were taken in settlement areas in order to investigate pollutant concentrations in the soils where people live. Acid treatment to isolate valuable metals is conducted in designated areas. Soil in those areas, contaminated with heavy metals, can potentially be transported to settlement areas with the wind, during flooding or via the groundwater transport, but with the data available these processes cannot be distinguished from leaching or incorporation of metal-containing waste particles into the soil. Despite showing an association with pyrogenic PAHs, it is likely that some of the Pb contamination originated from leaking lead-acid batteries or broken CRT computer screens. This may for instance explain the highest lead concentration found in any of the samples at site A-4.

Methylated PAHs may also reach the soils as a result of spills. Very high levels were found in one K-sample (6900 ng/g) and another K- and three A-samples showed relatively high concentrations (between 1400

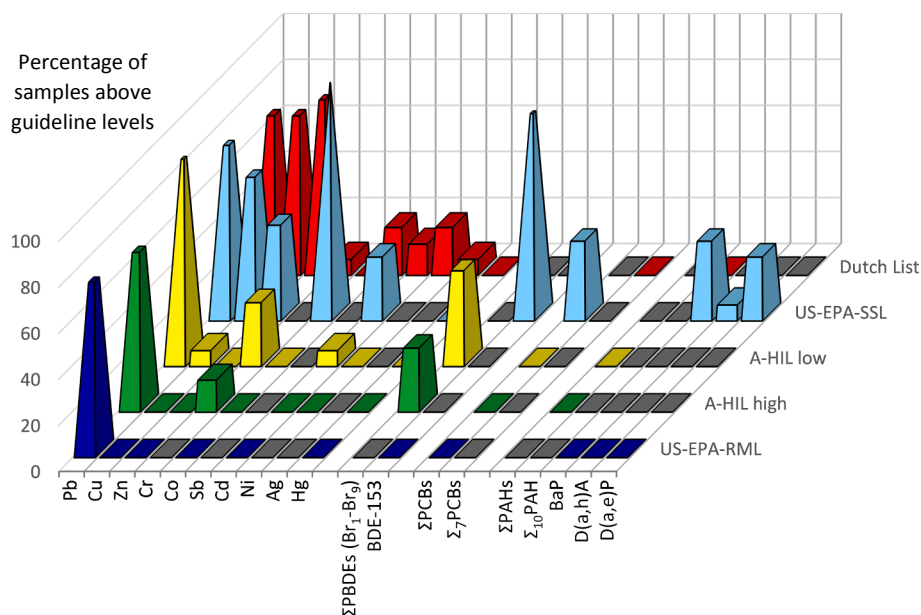


Fig 3. Percentage of Agbogbloshie samples with concentrations above guideline levels. Grey zero values mean that the chemical was not included in the corresponding guideline.

and 2400 ng/g) compared to the other samples. This indicates influence of small spills of petrogenic products while the other sampling sites are more likely to have received alkylated PAHs mainly via atmospheric deposition. Average concentrations of alkylated PAHs in the K-samples are higher than in the A-samples (1100 ng/g vs. 650 ng/g) due to the very polluted K-sample. Excluding this, the average concentrations become very similar between the two waste sites, suggesting that alkylated PAHs were not related to e-waste.

3.2.3. Deposition of compounds evaporated within or outside the waste sites

All of the organic contaminants studied are semi-volatile compounds and are therefore able to evaporate not only when exposed to strong heat during burning processes but also at ambient temperatures. Potential sources can be located either within or outside of the waste site, for instance in the surrounding city or adjacent agricultural areas. Only evaporation sources onsite or within a short distance of the waste site would be expected to be able to cause significant spatial differences in the contaminant amounts depositing to soils onsite. Especially the more volatile PCB congeners are likely to be affected by evaporation in the urban surroundings of the waste sites. The variation of their concentrations in samples collected in Agbogbloshie was low compared to PBDEs and CPs (Tab. SI-8–SI-10) and PCAs in combination with PAHs (Fig. 2 and SI-13) suggested no link to pyrogenic PAHs. Nevertheless, concentrations of the more volatile PCBs, which showed no association with pyrogenic PAHs in the A-samples, are over one order of magnitude higher in most A-samples compared to K-samples. Therefore, it can be assumed that a large proportion of lighter PCBs that deposited to the soil after outgassing originated from e-waste-related sources rather than from the surrounding city. A high contribution of tri-PCBs to the total PCB concentration, as found in our samples, was also observed in soils from other e-waste sites (Tang et al., 2010; Liu et al., 2013) and from urban areas ca. 50 km west of Agbogbloshie (Bentum et al., 2016). As suggested in these studies, it seems most plausible that they originate from transformers that contain for instance Aroclor 1242 as dielectric medium. PCBs may evaporate both from obsolete transformers and, as speculated by Bentum et al. (2016), from equipment handled at a transformer servicing center in Accra. Outgassing from materials containing the more volatile PBDEs and CPs (i.e. t-penta PBDEs and SCCPs) may also be a source of those compounds, which did not show a strong link to burning. The much higher concentrations found in the A-samples

compared to the K-samples suggest that also for those chemical groups at least some of the soil burden at Agbogbloshie is due to emissions from the waste dumped on site rather than having been transported from the wider surroundings via the atmosphere. For CPs this would also explain the higher contribution of highly chlorinated and hence less volatile congeners in A- compared to K-samples, as those are expected to travel less readily in the atmosphere. Methylated PAHs may also reach the soil by atmospheric deposition after volatilizing from sources on the waste site or elsewhere in the cities of Freetown or Accra.

3.2.4. Volatilisation

Compounds that evaporate from sources on the waste site may deposit again nearby or undergo atmospheric transport over short or longer ranges. Our data does not allow for estimating the amounts of contaminants volatilizing from the Agbogbloshie area and the percentage that experiences long-range transport vs. that depositing within the Accra region.

However, given that PBDE concentrations in some of the Agbogbloshie soils are very high compared to any literature data available, volatilization from soil seems conceivable. There are no data available on PBDE concentrations in the air in Accra or at Agbogbloshie itself, but with the predominating wind from the south/southwest (i.e. from the sea), it is very likely that the waste site soils contribute to an increase in PBDE concentrations in the relatively uncontaminated air coming from the sea (Xie et al., 2011). Since the compounds released may reach remote regions far from the waste site, we believe that providing data that allows for quantifying the extent of this process is an important task future studies on the global impact of e-waste should focus on.

3.3. Contaminants of most concern for human health

To assess which of the pollutants analysed are of highest potential concern to human health we have compared the concentrations found in this study to the following guideline values: Australian health-based investigation levels (A-HILs) for different scenarios (see SI for details), US-EPA regional soil screening and removal management levels (US-EPA-SSL and US-EPA-RML), and the New Dutch List intervention levels (NEPC, 2011; U.S. EPA, 2019a; U.S. EPA 2019b; Rijkswaterstaat, 2013).

These guidelines were chosen as they include a wide range of organic and inorganic pollutants and are commonly used in many countries to assess soil contamination. Fig. 3 and SI-17 give the percentages of Agbogloboshi and Kingtom samples where the concentrations found exceeded the guideline concentrations for those chemical groups that showed high concentrations in the waste site soils compared to literature data, i.e. for PAHs, PBDEs, PCBs, and various heavy metals/metalloids. Fig. SI-18a-t provides a more detailed picture, showing the concentrations measured in individual samples in comparison with the different guideline levels.

For soils collected at the Kingtom site, the concentrations of the organic pollutants measured were mostly below the guideline values used for comparison. Several heavy metals on the other hand exceeded guideline screening/investigation levels in several samples, especially Cr, Cu, Zn, Co and Pb. In some samples Pb, Cu, Zn, Co, Sb and Ag were also found above removal management/intervention levels (Fig. SI-17, SI-18).

In the Agbogloboshie samples, concentrations of individual carcinogenic PAHs, PBDEs, and PCBs exceeded screening/investigation levels (Fig. 3b, SI-18). This suggests a potential but probably low risk to people from these compounds. Several heavy metals and metalloids reached concentrations above intervention/removal guidelines, most frequently Pb, Cu and Zn but also Sb, Cd, Cr, Ni and Ag. This observation is in agreement with the high levels of those metals recently found in blood, urine and hair of e-waste workers at Agbogloboshie (Amankwaa et al., 2017). No guidelines were available for chlorinated paraffins but a "Risk assessment of short-chain chlorinated paraffins in Japan" (Tsunemi, 2009) suggested a soil screening level of 10,000 µg/kg (wet weight). Wet weight-based concentrations are not available for our samples but with up to 28,000 µg/kg (dry weight-based) it can be assumed that the SCCP concentration in some samples would be above this suggested wet-weight screening level.

However, the guidelines available only reflect a part of the risk caused by the contaminants that people in such environments are exposed to, and may, therefore, underestimate the risk. PAHs of pyrogenic origin and other compounds associated with burning or evaporation from waste materials will reach the soil predominantly via deposition from the air. It should, therefore, be assessed whether inhalation of polluted air is another major uptake route for people living on or in the vicinity of e-waste sites. Soil guideline values however do not include this atmospheric exposure pathway, and some do also not consider the uptake of contaminants via products from animals reared on polluted soils which is commonplace at Agbogloboshie and Kingtom.

3.4. Concluding remarks and future suggestions

Several heavy metals, PBDEs, PAHs, and possibly SCCPs can be found at concerning high concentrations in dumpsite soils, with the majority of the burden originating from the handling and dumping e-waste and other waste materials. Some of those pollutants (Cu, Pb, Ni, Sb, Cd, Ag, Hg, octa-tech BDEs and highly chlorinated PCBs) are closely linked to burning and may be reduced by replacing burning as a method to isolate valuable metals. So far, in Agbogloboshie, campaigns have aimed at providing education and encouraging people to stop burning and use the provided manual cable stripping machines and personal protection equipment (Little, 2019). However, some of the pollutants (e.g. Zn, Cr, Co, t-penta BDEs, CPs) appear not to be associated with burning but rather evaporate or leach out of waste materials or get incorporated into the soil in association with waste fragments. Hence, reducing their soil burdens and human exposure will require other measures, e.g. disposing of unwanted materials in sealed pits rather than simply leaving them behind where the processing was conducted or to discard them in the Korle Lagoon (Akormedi et al., 2013). However, to date, large amounts of e-waste have already been piled on the ground, making efficient prevention of dispersal difficult. Therefore, financial support for e-waste workers to enable a stricter separation

between waste site and residential areas would be desirable.

For a more complete assessment of the pollution and exposure situation we recommend that:

- Additional compounds should be included in the analysis (e.g. organophosphorous and non-PBDE brominated flame retardants, dioxins/furans, arsenic) and the main sources of compounds showing harmful concentrations should be assessed.
- A larger number of samples should be taken as the pollutant concentrations vary largely, even between samples collected close to each other.
- Air samples and foodstuff should also be analysed, the former preferably in the places where e-waste workers and non-workers spend most of their time.

Efforts to reduce the impact of e-waste recycling on people and the environment should then take into account the main sources and exposure routes of contaminants.

CRedit authorship contribution statement

Claudia Moeckel: Data curation, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing - original draft, Writing - review & editing. **Knut Breivik:** Formal analysis, Funding acquisition, Investigation, Project administration, Resources, Supervision, Writing - original draft, Writing - review & editing. **Therese Haugdahl Nøst:** Methodology, Software, Visualization, Writing - original draft, Writing - review & editing. **Alhaji Sankoh:** Conceptualization, Investigation, Methodology, Project administration, Writing - original draft. **Kevin C. Jones:** Writing - original draft, Writing - review & editing. **Andrew Sweetman:** Conceptualization, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Supervision, Writing - original draft, Writing - review & 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.

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Appendix A. Supplementary material

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