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## Research article



# Heavy metal contamination of faecal sludge for agricultural production in Phnom Penh, Cambodia

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#### ABSTRACT

To achieve the universal target of 'safely managed sanitation' set out in UN Sustainable Development Goal 6, the world needs to increase its rate of progress, since e.g. Phnom Penh, the capital of Cambodia, currently has zero percent safely managed sanitation. One way to promote safer faecal sludge management is to shift to a more circular system with nutrient recycling, but this carries the risk of heavy metal accumulation in the environment. This study analysed the concentrations of heavy metals in raw faecal sludge from various sources and assessed the appropriateness of resource recovery and reuse in relation to the heavy metal and nutrient loads in faecal sludge. A total of 42 samples collected from sludge disposal sites in Phnom Penh during the dry and rainy seasons were analysed for heavy metals and physicochemical parameters. Mean measured concentrations of heavy metals in faecal sludge samples decreased in the order Zn > Cu > Pb > Cr > Ni > Hg > As > Cd in both seasons but were higher in the rainy season, probably due partly to inflow from stormwater drains and run-off from roads during storm events. All elements analysed were within the permissible limits for application to land according to EU standards and USEPA. However, Hg and Zn concentrations exceeded the tolerance limits for local organic fertiliser and Swedish limits for compost. Faecal sludge is thus not an appropriate fertiliser considering the risk of heavy metal accumulation in relation to phosphorus recovered. Options to avoid recirculating pollutants to the environment include upstream prevention of pollution, source separation of household wastewater fractions and use of biosolids as a soil conditioner together with other fertilisers or for soil production. Additional studies are needed on these options if sanitation stakeholders are to close the nutrient loop.

## 1. Introduction

The world will likely fail to achieve universal access to safely managed sanitation coverage in 2030, as set out in UN Sustainable Development Goal 6, target 6.2, if it continues with its current rate of progress, with 2.8 billion people still without a safe sanitation service (WHO & UNICEF, 2021). Onsite sanitation currently plays a key role in meeting the sanitation needs of around 2.7 billion of the world's population and this number is expected to reach 5 billion by 2030 (Strande, 2014). Approximately 1 billion onsite facilities worldwide are in urban areas (Strande, 2014). Indiscriminate disposal of the large quantities of faecal sludge generated by these facilities could lead to outbreaks of disease, as well as causing environmental pollution, eutrophication of waters and loss of the aesthetic beauty of nature (Kuffour et al., 2013). Therefore, safe management of faecal sludge is needed to avoid negative impacts on public health and the environment (Zewde et al., 2021). However, safe management of faecal sludge involves addressing the whole sanitation service chain, including collection, emptying and transportation, processing and safe disposal (Strande et al., 2014; Boot and Scott, 2008).

Many cities in low- and middle-income countries have poor faecal sludge management, with no legal framework and almost no services. For example, Phnom Penh, the capital of Cambodia, has 0% safely managed faecal sludge generated from onsite sanitation facilities and

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the sludge often ends up in the immediate urban environment, posing risks to public health and the environment (PPCA, 2021). For instance, diarrhoea is one of the primary contributors to global disease burden and faecal contamination attributed 88% of the cases (Otoo et al., 2015). A recent study in Phnom Penh revealed that households only empty their sludge container when it is overfilled or clogged (Eliyan et al., 2022a). This indicates a lack of regular emptying, which is a requirement to ensure proper performance of septic tanks, leading to poor performance of the system. Only 22% of Phnom Penh city dwellers report having their container emptied, while only 12% of the sludge collected reaches the authorised disposal site (Peal et al., 2015). The indiscriminate dumping of untreated faecal sludge contributes significantly to surface water and ground water pollution (Krithika et al., 2017), e. g eutrophication from excess nutrients can negatively impact the functions of natural ecosystem (Andersson et al., 2016).

Viewing human waste as a potential resource could enable a paradigm shift towards sustainable sanitation and faecal sludge management, thus avoiding associated environmental and health risks. Instead of indiscriminate disposal in the open environment and in local neighbourhoods, safe faecal sludge management through recovery of nutrients could improve sanitation and benefit the agriculture sector (Chandana and Rao, 2022). Sludge treatment is urgently needed to prevent pollution and to handle the large quantities of faecal sludge generated (Michael Steiner et al., 2002). In addition, different forms of treatment end-products could be recovered from faecal sludge, e.g. its high organic matter content makes it suitable for biogas production through anaerobic digestion (Ahmed et al., 2019) or as a soil amendment in crop production (Zewde et al., 2021). Its high calorific value is adequate for bioenergy generation or use as biofuel (Hafford et al., 2018). Other forms of resource that can be recovered from faecal sludge include building materials, protein, animal feedstuffs and water for irrigation (Andriessen et al., 2019; Diener et al., 2014).

Protecting public health is a precondition for reuse of faecal sludge in agriculture (Tayler, 2018). The WHO and United States Environmental Protection Agency (USEPA) have established guidelines for using treated waste (biosolids) that require components defined as Class A and Class B biosolids to meet limits for pathogen removal and pollutant concentrations (WHO, 2006; USEPA, 1994). Pathogen removal can be achieved by use of appropriate treatment technologies, e.g. some studies report almost 100% pathogen inactivation and low helminth egg viability through thermophilic composting of faecal sludge (Strande et al., 2014; WHO, 2006). In contrast, heavy metals are very challenging to remove during treatment of faecal sludge and pose threats to human health and the environment if they accumulate in soil (Shamuyarira and Gumbo, 2014). Even in middle and high income countries where pathogenic hazards from wastewater are likely controlled, there is growing concerns about heavy metals and other chemicals contaminants in reuse systems (Manzoor Qadir et al., 2015). Therefore, information on the concentrations of heavy metals in faecal sludge in relation to the concentrations of available nutrients is necessary when planning resource

Previous studies have found that concentrations of heavy metals in faecal sludge vary based on season and source. For example, a study in Dar es Salaam, Tanzania, found a significant difference between the dry and rainy seasons in conductivity value and total solids concentration in pit latrine sludge (Doglas et al., 2021). Yet, it may be difficult to extrapolate data from African studies to the Asian context. The majority (58%) of onsite containment systems in African cities are pit latrines whereas Asian cities commonly have waterborne containment systems that discharge to open drain (Peal et al., 2020). For instance, water based toilet connected to onsite containment units is predominantly used by residents in Phnom Penh (Eliyan et al., 2022a). Therefore, local data are needed for proper planning and to ensure there are no potential risks associated with changing from current practices to a circular

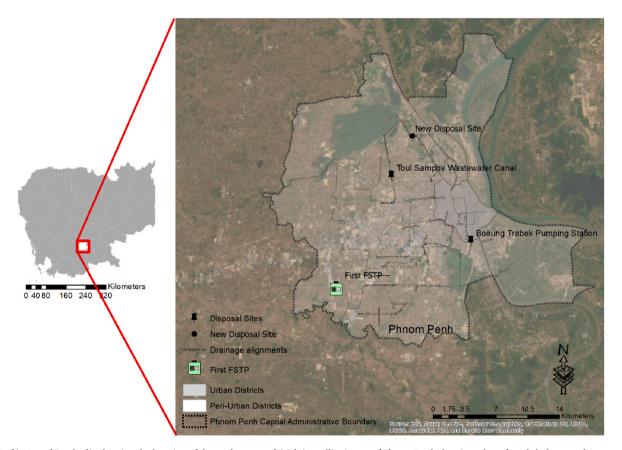


Fig. 1. (Left) Map of Cambodia showing the location of the study area and (right) satellite image of Phnom Penh showing where faecal sludge samples were collected during the rainy season (Toul Sampov wastewater canal and Boeung Trabek pumping station) and dry season (new disposal site and Boeung Trabek pumping station).

system (Chandana and Rao, 2022).

The aims of the present study were to determine the concentrations of heavy metals in raw faecal sludge from various sources and to assess the appropriateness of resource recovery and reuse in relation to heavy metal and nutrient concentrations in faecal sludge. Specific objectives were to: 1) identify whether there are seasonal variations in heavy metal concentrations in faecal sludge from different sources in Phnom Penh; 2) assess whether faecal sludge can be classified as biosolids based on national and international standards for heavy metal concentrations; and 3) provide baseline data on heavy metal concentrations in faecal sludge in relation to the plant nutrient content, to support sanitation stakeholders in selecting appropriate faecal sludge management technologies for resource recovery in Phnom Penh.

#### 2. Methods

Faecal sludge samples for the study were collected from disposal sites identified in a previous study as the main locations receiving sludge collected from household containment units in Phnom Penh (Eliyan et al., 2022a). The characteristics of the samples obtained were analysed both onsite and at the laboratory.

#### 2.1. Study area

The study area was the city of Phnom Penh, located at 11°34″N, 104°55″E on the Mekong floodplain, above the confluence of the Mekong, Tonle Sap and Bassac rivers (JICA, 2016). The city is undergoing rapid development and is now divided into 14 districts classified as urban areas (5 districts) and peri-urban areas (9 districts). The total population of Phnom Penh is around 2 million, with approximately 500, 000 households (NIS, 2020). There are typically two seasons in Cambodia, a rainy season (June to October) and a dry season (November to May). According to mean monthly rainfall data for the period 2004–2013, February had the lowest precipitation level (8.1 mm) and September had the highest (272.4 mm), followed by October (244 mm) (JICA, 2016). The wettest month shifted to October in 2020, while February remained the driest month, according to reports from the Cambodian Ministry of Water Resources and Meteorology.

Onsite sanitation is the predominant sanitation system used in Phnom Penh (Peal et al., 2015; Frenoux et al., 2011). There are two types of onsite containment system, cesspits and septic tanks. Cesspits are by far the most common system, serving over 90% of households in both urban and peri-urban districts of Phnom Penh. Regardless of the type of containment system they use, 95% of households in urban districts have the overflow from their containment system connected to a sewer network, while only 63% of peri-urban households have such a connection (Eliyan et al., 2022a). There is no legal requirement on regular emptying of containment systems and most Phnom Penh households have their containment unit emptied only when it is clogged (57%) or full (35%).

There is only one faecal sludge treatment plant in Phnom Penh at present, and it was opened only recently (19 May 2023). Previously, Boeung Trabek pumping station (Cheung Ek wetland) was the only authorised faecal sludge disposal site (JICA, 2016). However, a recent study by Eliyan et al. (2022a) identified further disposal sites (possibly unauthorised) in addition to Boeung Trabek pumping station, such as public manholes near households, fields around the Kob Srov area, Toul Sampov wastewater canal and a smaller canal (approximately 1 km length) connected to Toul Sampov wastewater canal. That study concluded that wherever faecal sludge is disposed of within the drainage network, it ends up in the two main receiving reservoirs (Cheung Ek and Kob Srov wetlands). Such indiscriminate disposal of faecal sludge in Phnom Penh has also been reported in other studies (PPCA, 2021).

The faecal sludge samples analysed in the present study were mainly collected at the two main disposal spots, i.e. Boeung Trabek pumping station and Toul Sampov wastewater canal (Fig. 1). However, during dry

season sampling of faecal sludge it was found that there were no incoming trucks discharging sludge into Toul Sampov wastewater canal, due to recent enforcement by the local authority of a ban on indiscriminate disposal of faecal sludge around the canal area, with a fine of 500 USD applied to operators contravening the ban. To overcome this challenge, local operators have begun to use a new disposal site located in northern Phnom Penh that is also connected to Kob Srov wetland, i.e. the wetland is still the recipient of all faecal sludge collected in the area. Dry season sampling of faecal sludge (28 January-12 February 2023; n = 7 samples) for the present study was conducted at the new disposal site, instead of the Toul Sampov wastewater canal site. Wet season sampling was conducted earlier (2–15 October 2022), when Toul Sampov was still an active disposal site.

## 2.2. Screening step before collecting faecal sludge samples

An initial screening was performed to determine whether samples needed be taken from all incoming trucks at each sampling site. Screening criteria were source of faecal sludge and consent from emptying and transportation operator. A decision was made to exclude industrial sources and a temporary containment system in the study area, since sludge from industrial sources was expected to differ significantly from that from other sources and since the temporary containment system served a construction site for a new shopping mall and would disappear once construction ended. If extracted faecal sludge arriving at the sampling sites was from non-industrial sources and from permanent containment systems and if consent was granted, a sample was taken.

#### 2.3. Faecal sludge sampling procedure

The sampling design took into account variations between the seasons in Phnom Penh. A total of 42 faecal sludge samples were collected, with 21 samples each in the rainy and dry seasons. Before actual sampling began, site observations were conducted to confirm whether the preliminary sampling plan and sites were applicable. Before rainy season sampling, a week of observations revealed that Boeung Trabek pumping station received more frequent discharges than Toul Sampov wastewater canal. This agreed with previous findings that Boeung Trabek pumping station receives around 60% of total faecal sludge discharge in Phnom Penh (Eliyan et al., 2022a). Therefore, in the rainy and dry seasons, the initial plan was to collect 28 samples from Boeung Trabek pumping station and 14 samples from Toul Sampov wastewater canal. Since the discharged ratio between Boeung Trabek pumping station and Toul Sampov wastewater canal is approximately 2:1, the sampling strategy is representative. When the new site for dry season sampling was identified, it replaced the seven dry-season samples that were planned to be collected from Toul Sampov wastewater canal. The faecal sludge dumped at the disposal sites comes from various sources, but this study focused only on sludge extracted from containment systems serving households, rented houses, apartments and restaurants.

Faecal sludge sampling was designed based on findings in a previous study in Phnom Penh that three factors significantly influence faecal sludge characteristics in the city: i) addition of water during emptying, ii) connection to urban drainage network and iii) type of wastewater captured by household containment systems (Eliyan et al., 2022b). Specific location (urban vs peri-urban) and type of containment unit (septic tank vs cesspit) were thus not critical factors contributing to the variation in faecal sludge characteristics in that study. Sampling was conducted at sludge disposal sites, since they are the key targets for construction of treatment plants (Koottatep et al., 2021). The first sampling round (rainy season) was performed during 2–15 October 2022 (since October was the wettest month in 2020) and the second sampling round (dry season) during 28 January-2 February 2023 (since February was the driest month in 2004–2013 and in 2020). Each sampling round consisted of seven alternate sampling days within the

Table 1
Physicochemical properties of faecal sludge samples collected in Phnom Penh in the dry and rainy seasons. SD = standard deviation, P-values <0.05 indicate statistically significant difference between dry and rainy seasons, (–) indicates data not available.

Parameter	Both seasons ( $n = 42$ )			Dry season ( $n = 21$ )					Rainy season ( $n = 21$ )					P-value
	Median	Mean	SD	Min	Max	Median	Mean	SD	Min	Max	Median	Mean	SD	
Conductivity (mS/cm)	1.5	1.8	0.99	0.62	4.9	1.5	1.8	1.0	0.64	4.5	1.5	1.7	0.98	0.586
DO (mg/L)	0.56	0.56	0.090	0.42	0.82	0.57	0.58	0.08	0.43	0.83	0.53	0.54	0.092	0.157
pH	7.1	6.9	0.60	5.4	8.3	7.1	7.0	0.62	5.5	7.6	7.0	6.9	0.59	0.484
Temp (°C)	29	29	1.4	27	33	30	30	1.7	28	32	29	29	1.1	0.266
TS (%)	1.9	2.9	2.9	0.52	10	2.1	2.9	2.4	0.30	13	1.4	2.9	3.5	0.261
VS (%)	69	62	18	33	84	72	64	17	13	90	63	61	19	0.517
NH <sub>4</sub> -N (g/L)	_	_	_	0.04	0.68	0.11	0.16	0.14	_	_	_	_	_	_
TN (g/L)	_	_	_	1.5	3.3	2.0	2.2	0.65	_	_	_	_	_	_
PO <sub>4</sub> -P (g/L)	_	-	_	0.01	0.68	0.61	0.10	0.14	_	_	_	-	_	_
TP (g/L)	_	_	_	0.22	2.5	0.23	0.47	0.65	_	_	_	_	_	_

selected two-week period. On each sampling day, the study team waited for incoming trucks at each disposal site and collected faecal sludge samples from any incoming trucks that met the screening criteria. During sampling, the study team asked the contractor to begin discharging the sludge as usual and, after 1 min of discharge, a sample was collected in a 1-L polypropylene bottle and subjected to recommended sample handling and preservation techniques (APHA, AWWA and WEF, 2017).

## 2.4. Faecal sludge analysis and quality control

The collected samples were immediately analysed onsite for the following parameters: Conductivity, dissolved oxygen (DO), pH and temperature (Temp), using a HORIBA-U-52G multi-parameter water meter. Regular calibration of the instrument was performed before each sampling day to ensure accurate measurement. The samples were then placed in an ice box and transported to the Department of Environmental Science Laboratory, Royal University of Phnom Penh, for further analysis. At the laboratory, gravimetric analysis method 2540G (drying, cooling, desiccating and weighing) was used for determination of total solids (TS) and volatile solids (VS) content. Sub-samples were then pretreated by drying at 103–105 °C and sent to Bureau Veritas (Cambodia) Limited Laboratory, where the concentrations of arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), mercury (Hg), nickel (Ni), lead (Pb) and zinc (Zn) were determined by microwave digestion (methods MARS 6 and ICPMS-7700), where the detection limit for As, Cd, Cr, Cu, Hg, Ni, Pb and Zn was 0.1, 1, 1, 2, 0.02, 1, 1 and 2  $\mu$ g/L, respectively. The blank spiked recovery rate of all elements in rainy season samples was: As 9.5%, Cd 90.1%, Cr 104% Cu 107%, Hg 120%, Ni 98%, Pb 114% and Zn 90%. In dry season samples it was: As 93.2%, Cd 97.7%, Cr 101% Cu 104% Hg 90.1%, Ni 93.7%, Pb 98.9% and Zn 98%.

The dry-season samples were preserved by adding H<sub>2</sub>SO<sub>4</sub> to obtain pH < 2 (APHA, AWWA and WEF, 2017) and frozen for transport to the laboratory at the Department of Energy and Technology, Swedish University of Agricultural Sciences (SLU), for nutrient analysis (ammonia nitrogen (NH<sub>4</sub>-N), total nitrogen (TN), phosphate-phosphorus (PO<sub>4</sub>-P) and total phosphorus (TP)). Merck test kits were used for all nutrient analyses, following the manufacturer's instructions. A two-step approach was employed for determination of TN. The first step was digestion (Spectroquant Crack set Cat. No. 1.14963) to transform organic and inorganic nitrogen compounds into nitrate according to Koroleff's method by using oxidising agents in a thermo-reactor at 120 °C for 1 h. According to the manufacturer's instructions, chemical oxygen demand (COD) acts as interference if it exceeds 0.35 g/L. Other studies have found that COD in faecal sludge can range from 8 to 122 g/L (Ward et al., 2021; Junglen et al., 2020; Ahmed et al., 2019; Afolabi and Sohail, 2017; Bassan et al., 2013). Thus the COD concentration in faecal sludge samples is generally high and a high dilution factor is needed to reduce it to a level that will not consume the oxidising agent, leaving organic and inorganic nitrogen compounds in the digested solution.

Here, the amount of oxidising reagent needed to be doubled to ensure complete transformation of nitrogenous compounds in the faecal sludge samples. The digestion solution was then analysed using the Spectroquant Nitrate test (Cat. No. 1.09713) and Spectroquant Ammonium cell test (Cat. No. 1.14559). Similarly, a two-step process was employed to analyse TP (Spectroquant Crack set Cat. No. 1.14687), where the digestion solution was analysed following  $PO_4$ –P analysis by colorimetric methods, using a Thermo Scientific Gallery<sup>TM</sup> discrete analyser.

#### 2.5. Statistical analysis

The analytical data obtained were computed in Microsoft Excel (2010), and R software version 4.0.4 (R Core Team, 2021) was used for data analysis. Descriptive statistic tools were used to assess faecal sludge characteristics across all samples and by season. According to the central limit theorem, the mean of observations could be assumed normally distributed when sample size is large enough (>20). The non-parametric method (Wilcoxon's rank sum test) was used to check for statistically significant differences between seasons for parameters that were not normally distributed, such as TS, VS, As, Cd, Cr, Cu, Hg, Ni, Pb and Zn, while the two sample *t*-test was applied for data with a normal distribution, such as Conductivity, DO, pH and Temp. Half the limit of quantification value was applied for calculation of heavy metal content in relation to P recovered.

## 3. Results

Households (including apartment and rented houses) were the dominant source of faecal sludge collected, representing 93% of total samples, while other source (apartments) made up 7%. Approximately 67% of faecal sludge samples collected were from peri-urban districts, while other locations including urban districts of Phnom Penh and two nearby cities (Oudong and Ta Khmau) contributed the remainder. Oudong, in Kampong Speu province, lies approximately 35 km northwest of Phnom Penh and Ta Khmau, the largest city in Kandal province, lies about 11 km south of Phnom Penh. The majority of sludge samples collected were black (62%), with other colours such as dark brown, brown, light brown and yellow together representing 38%.

## 3.1. Nutrient content in faecal sludge

The parameters measured in the field (Conductivity, DO, pH, Temp) did not show statistically significant differences between seasons (Table 1). However, the mean value of all parameters was generally lower in rainy-season than in dry-season samples. The range of values obtained for dry-season samples was: Conductivity 0.62–4.9 mS/cm, DO 0.42–0.82 mg/L, pH 5.4–8.3 and Temp 29–33 °C. Similar ranges were found for all parameters in rainy-season samples.

The concentrations of TS, VS,  $NH_4$ –N, TN,  $PO_4$ –P and TP showed high variation, uneven distribution and high standard deviation

Table 2 Summary statistics on heavy metal concentrations (mg/kg) in faecal sludge samples collected in Phnom Penh in the dry and rainy seasons. All values expressed on a dry mass basis (mg/kg total solids, TS). Values in bold indicate significant difference between the seasons (P < 0.05).

Parameter	Dry seaso	n (n = 21)				Rainy seaso	P-value				
	Min	Max	Median	Mean	SD	Min	Max	Median	Mean	SD	
Non-essential l	neavy metals										
As	0.28	5.7	1.1	1.7	1.6	2.8	15	4.4	5.4	3.0	< 0.001
Cd	<1ª	2.3	<1ª	_	_	<1 <sup>a</sup>	2.2	1.1	_	_	0.011
Hg	1.1	16	3.7	4.9	3.6	$< 0.02^{a}$	16	3.1	5.3	5.1	0.654
Pb	1.4	17	10	9.2	3.9	9.1	112	46	49	27	< 0.001
Essential heavy	metals										
Cr	<1ª	13	3.8	4.1	2.7	22	72	40	43	13	< 0.001
Cu	15	670	79	120	130	67	280	160	150	61	0.0062
Ni	2.5	57	14	21	17	12	30	18	19	5.3	0.102
Zn	810	1900	1300	1200	330	320	4300	1600	1600	900	0.184

<sup>&</sup>lt;sup>a</sup> Below limit of quantification.

(Table 1). The standard deviation was almost as high as the mean value of the respective parameter in all cases. The TS and VS content in faecal sludge samples did not differ significantly between the rainy-season and dry-season samples. In dry-season samples, the TS content as 0.52–0% and the VS content 33–84%TS, while in rainy-season samples the range was 0.30–13% for TS and 13–90%TS for VS. The mean values of both TS and VS were generally lower in rainy-season samples. Only dry season data were available for NH<sub>4</sub>–N, TN, PO<sub>4</sub>–P and TP concentrations, so comparison between the seasons was not applicable. The mean concentration ( $\pm$ standard deviation) of NH<sub>4</sub>–N and TN was 0.16  $\pm$  0.14 g/L and 2.2  $\pm$  0.65 g/L, respectively (range 0.04–0.68 g/L and 1.5–3.3 g/L, respectively). The mean concentration ( $\pm$ standard deviation) of PO<sub>4</sub>–P and TP was 0.10  $\pm$  0.14 g/L and 0.47  $\pm$  0.65 g/L, respectively.

#### 3.2. Seasonal variation in heavy metal loads in faecal sludge

Summary statistics on measured heavy metal loads in faecal sludge in Phnom Penh are presented in Table 2. As seen for chemical parameters, the concentrations of the different heavy metals analysed (classified into two groups, essential and non-essential metals) also showed high variation, non-normal distribution and large standard deviation (almost equal to the mean value). Mean and median values were very different, but the mean concentration ( $\pm$ standard deviation) of all heavy metals analysed was generally higher in rainy-season than in dry-season samples. The concentration ranges (mg/kg TS) in dry season samples were: As 0.28–5.7, Cd < 1–2.3, Hg 1.1–16, Pb 1.4–17, Cr < 1–13, Cu15-680, Ni 2.5–57, Zn 810–1900, while those in rainy-season samples were: As 2.8–15, Cd < 1–2.2, Hg < 0.02–16, Pb 9.1–112, Cr 22–72, Cu 67–280, Ni

12–30, Zn 320–4300. The two metals present in the highest concentration were Zn and Cu in both seasons, while the two present in the lowest concentrations were As and Cd in both seasons. The concentrations of Pb and Cr were higher than those of Ni and Hg in rainy-season samples, while a trend for the reverse was observed in dry-season samples, where the Cr concentration was among the lowest recorded.

#### 4. Discussion

The mean concentration of TN in faecal sludge samples in this study was higher than values reported in the literature (Eliyan et al., 2022b; Ahmed et al., 2019; Afolabi and Sohail, 2017). This discrepancy could derive from analytical errors arising when using spectrophotometric methods for TN analysis, e.g. from insufficient addition of oxidation reagent leading to organic and inorganic N remaining in samples after digestion owing to the high COD in faecal sludge samples. This interference could be the reason why NH<sub>4</sub>-N was reported to be higher than TN in one study (Ahmed et al., 2019), when theoretically it should be lower, and why total Kjeldahl nitrogen (TKN) was higher than TN in another study (Afolabi and Sohail, 2017), when it should be lower than or equal to TN as nitrates are not detected in that method. A similar issue was found in a previous study in Phnom Penh (Eliyan et al., 2022b). However, the TN concentrations found in the present study were in line with previously reported values in a study of septic tank contents in Hanoi, Vietnam, and Kampala, Uganda (Englund et al., 2020) and in household pit latrine faecal sludge in Kampala, Uganda (Strande et al.,

Significant differences in concentrations in faecal sludge between the  $\,$ 

Table 3

Summary of mean values of all parameters in dry-season and rainy-season faecal sludge (FS) samples in this study and mean values from other studies, and permissible limits of heavy metal concentration in the Cambodian standard for organic fertiliser and other international standards and guidelines on use of biosolids for land application. Bold type indicate that a value exceeds either the Cambodian standard for organic fertiliser or the Swedish limit for compost, while bold and italics indicate that it exceeds both. All values are in mg/kg total solids (TS), unless otherwise indicated. Value after  $\pm$  signs are standard deviation. (-) data not available. The grey shaded panel shows values from different standards and guidelines for comparison.

Parameter	This study		Mean hear	vy metal concen		Values from different standards and guidelines					
	Mean	FS in Nakuru,	FS in peri-urban	FS in	Dewatered	Dry	Wet	Cambodia	Permissible	Council	USEPA
	value	Kenya <sup>(1)</sup>	Ashanti region,	Limpopo,	FS in	dewatered FS	dewatered FS	National	limit for	Directive	Limits for
	from both		Ghana <sup>(2)</sup>	South	Kampala,	in Greater	in Greater	standard for	Compost in	86/278	EQ and PC
	seasons			Africa <sup>(3)</sup>	Uganda <sup>(4)</sup>	Accra	Accra	organic	Sweden <sup>(7)</sup>	EEC(8)	Biosolids(9)
					-	region <sup>(5)</sup>	region <sup>(5)</sup>	fertiliser(6)			
	Non-essential heavy metals										
As	3.6±3.0	(4)	0.152±0.027	-	-	10.82±0.46	10.02±3.26	10	-	-	41
Cd	<1ª	2.3±0.1	0.045±0.005	0.32	0.9±0.4	0.60±0.11	0.86±0.04	5	3	20-40	39
Hg	5.1±4.3	CHI CHI	-	-	-	~	12	0.15	3	16-25	17
Pb	29±28	13.9±1.2	0.160±0.053	17.96	73.3±29.9	6.69±1.10	18.77±1.45	100	150	750-1200	300
					Esser	tial heavy metals					
Cr	23±22	42.4±4.3	-	44.13	46.9±36.2	42.53±6.23	26.33±11.94	50	150	-	1200
Cu	130±100	68.5±4.0	3.978±0.439	80.80	113.7±41.1	125.93±14.37	185.30±3.24	300	150	1000-1750	1500
Ni	20±12		-	18.89	15.3±6.8	25.12±4.05	22.04±1.82	50	50	300-400	420
Zn	1 400±690	51.6±4.8	2.235±0.102	303.83	448.7±151.9	694±72.56	923.77±86.65	1000	500	2500-4000	2800

<sup>a</sup>Median value. For abbreviations, see text. (1)(Moturi et al., 2018). (2)(Appiah-Effah et al., 2015), units mg/L. (3)(Shamuyarira & Gumbo, 2014). (4)(Manga et al., 2022). (5)(Ahmed et al., 2019). (6)(MAFF, 2012). (7)(Sharma et al., 2017). (8)(CD, 1986). (9)(USEPA, 1994)

dry and rainy seasons were detected for almost all metals analysed except Hg, Ni and Zn. In general, the concentration of all metals was higher in rainy-season samples than in samples collected in the dry seasons. One source of the greater heavy metal loads during the rainy season could be surface run-off during storm events. A previous study examining the levels of heavy metals in the Chan Thnal reservoir in Cambodia also concluded that the Cd, Cu, Zn and Pb concentrations were higher in the rainy season than in the dry season (Chheang et al., 2021). Higher loads of Cd during the rainy season have also been reported in a study in Eastern Cape province, South Africa (Agoro et al., 2020). However, all 42 samples collected in the present study were independent samples, so is difficult to compare the results obtained given the many possible influential variables, including types of containment systems, user habits and management practices. A study on pit latrine sludge in a peri-urban residential area of Zimbabwe found that user habits and management practices made each containment unique in terms of faecal sludge characteristics (Changara et al., 2018). Another factor that contributes significantly to the variation in physicochemical characteristics of faecal sludge in Phnom Penh is the type of wastewater captured by the containment systems (only blackwater or mixed household wastewater) (Elivan et al., 2022b). Those factors were not assessed in the present study, but could contribute to the presence of heavy metal in faecal sludge and greywater is the main contributor of heavy metals (Vinnerås et al., 2006). In this study, sampling was conducted at disposal sites and the research team had only a limited time to talk to operators during emptying events, so their responses were not used. In any cities that have onsite containment units capturing mixture of domestic wastewater, there would also be a chance that faecal sludge contains higher concentration of heavy metal than the permissible limit to be used as fertiliser.

Heavy metal concentrations found in faecal sludge in different studies are generally low (Afolabi and Sohail, 2017), as confirmed in this study. The mean concentration of all metals in both seasons followed the order Zn > Cu > Pb > Cr > Ni > Hg > As > Cd (Table 3). A similar decreasing trend in heavy metal concentrations is reported in the literature, e.g. Zn was the most common element found in faecal sludge from peri-urban areas in the Ashanti region, Ghana (Appiah-Effah et al., 2015); from Limpopo, South Africa (Shamuyarira and Gumbo, 2014); from dewatered faecal sludge in Kampala, Uganda (Manga et al., 2022); and from wet and dry dewatered faecal sludge in the Greater Accra region, Ghana (Ahmed et al., 2019). However, a study on Cd, Pb, Cu and Zn concentrations in sewage sludge in Nakuru, Kenya, found that Cu was present in the highest concentrations, followed by Zn (Moturi et al., 2018).

Heavy metals are categorised as essential, i.e. required in the diet and with a biological use, e.g. in enzymes (Cr, Cu, Ni, Zn) and non-essential, i.e. toxic to organisms even in trace amounts (Slobodian et al., 2021) (As, Cd, Pb and Hg). However, exposure to high amounts of any heavy metal, whether essential or non-essential, can have adverse health effects (Slobodian et al., 2021; Vinnerås et al., 2006). In general, the level in human excreta per person per day is: Zn 9-16 mg, Cu 1.4-1.5 mg, Ni 0.3 mg, Cd 0.02-0.03 mg, Pb 0.07-0.14 and Hg 0.01 mg (Schouw et al., 2002). The concentrations mainly reflect the use of the different metals in society, with some local deviation due to background content in the environment, e. g As from the groundwater. The major contributor of Cd could be the use of Cd in rubber tyres and heavy traffic, from where Cd can enter stormwater drains and run-off from nearby roads (Agoro et al., 2020; Shamuyarira and Gumbo, 2014). Phnom Penh is a city with heavy traffic on the roads and generating Cd-contaminated stormwater that enters the urban drainage system. During the rainy season and especially during severe storm events, the city is flooded and overflows of stormwater possibly enter faecal sludge containment systems. It has been reported that around 77% of residents in Phnom Penh have their faecal sludge containment unit emptied in the rainy season, when it probably becomes visibly full due to backflow or overflow (PPCA, 2021). Other non-essential metals such as As are found in groundwater in the study

region, with peri-urban areas of Phnom Penh being As risk areas. A groundwater study by UNICEF and other collaborating agencies revealed that these high concentrations of As in groundwater are strongly associated with the floodplains of the Bassac, Mekong and Tonle Sap rivers (Berg et al., 2007). The source of Hg could potentially be use of Hg-containing batteries in households and amalgam in dentistry. Dental amalgam (via teeth brushing and defecation) contributes more than 80% of Hg in domestic wastewater, while other possible sources of Hg contamination are common household and toiletry products (Friedler et al., 2019). In addition to the diet, Cu can also derive from other sources, such as corrosion of water pipes. Zinc is a constituent of galvanised steel, which is used in pipes in the drinking water distribution network, so piped water supply could be contaminated with Cu and Zn. Use of cleaning materials containing Cu and Zn is another possible source. A main source of Zn and Cu is in-house plumbing and greywater, which contribute 90% of the total metal load (Friedler et al., 2019). Household effluent such as greywater may contribute strongly to the total heavy metal content in faecal sludge (WHO, 2006).

Different sources contribute to the higher concentration of heavy metals in Phnom Penh and pose a concern that would limit the reuse potential of faecal sludge. Such concerns could possibly be relevant for other cities in similar context. For instance, any cities that face with frequent flooding, have onsite containment units connected to urban drainage network and have water based toilets connected to containment units. It is important to identify all sources of pollutants in order to implement measure upstream to limit heavy metal loads entering wastewater, because once the heavy metals are present in wastewater they are costly and difficult to remove and pose serious health issues.

Cambodia divides fertilisers into five different types, based on the nutrient and micronutrient content. These are: inorganic or chemical fertiliser, organic fertiliser, bio-fertiliser, soil conditioner and raw material. The classification is based on the primary nutrient (N, P, K) content in normal inorganic fertiliser (MAFF, 2012). Almost all individual heavy metal concentrations in the faecal sludge samples analysed in the present study were below the permissible limits in the Cambodia standard for organic fertiliser and the Swedish limits for compost (Sharma et al., 2017), with the exception of Hg and Zn (Table 3). There is currently no standard limit for total heavy metal load in faecal sludge in Cambodia, but all samples analysed fell within the acceptable range in biosolids based on the USEPA limits for exceptional quality for land application and the EU standard for sludge for use in agriculture (USEPA, 1994; CD, 1986).

Given the high concentrations of Hg and Zn in faecal sludge, exceeding the Cambodian standard for organic fertiliser, treated faecal sludge biosolids should only be used as a soil conditioner and not as a complete substitute for fertiliser, meaning that it should only be applied to some selected crops or applied only in limited amounts. To ensure safe reuse, further studies should seek to identify crops that may not absorb heavy metals, especially As, Pb, Ni and Zn, and to determine the amount of faecal sludge that may be safe to apply considering the amount of plant nutrients needed and heavy metal accumulation in soil and uptake by plants. Alternatively, treated faecal sludge should be used as fertiliser in soil production for non-edible plants such as grass and flowers in Phnom Penh and nearby city parks.

Another alternative could be to introduce a source separation system for faeces and urine. This would be beneficial for biological treatment (Rose et al., 2015), given the high nutrient content in urine and greater heavy metal load in faeces (Schouw et al., 2002). Urine is the highest contributor of nutrients to domestic wastewater (79% of N, 47% of P) and greywater is the lowest (Friedler et al., 2019). Most heavy metals in domestic wastewater (e.g. Zn, Cu, Ni, Cd, Pb, and Hg) derive from faeces (Vinnerås et al., 2006; Schouw et al., 2002). Therefore, urine is the most valuable resource that can be recovered from domestic wastewater.

The concentration of heavy metals in recycled fertiliser needs to be set in relation to the benefits of the fertiliser, since otherwise there is a risk of just diluting a polluted fertiliser with a clean material for

Table 4
Summary statistics on concentrations (mg/kg phosphorus (P)) of heavy metals in faecal sludge in this study and in other waste materials. Bold indicates that the value exceeds the average level in mineral P fertiliser. (–) not included/data not available. Grey shaded column shows mean heavy metal concentrations in mineral fertiliser samples in 12 European countries.

Parameter		Т	his study(1	)		Fresh urine(2)	Faeces(3)	Urine+faeces	Farmyard	Sewage	Mineral P
	Min	Max	Mean	Median	SD			mixed <sup>(3)</sup>	manure <sup>(3)</sup>	sludge <sup>(4)</sup>	fertiliser <sup>(5)</sup>
Non-essential heavy metals											
As	11	1 200	170	120	280	-	-	-	-	-	123
Cd	12	140	57	45	40	0.7	20	7	16	46.9	82.7
Hg	43	1 200	410	260	350	1	-	=	-	-	-
Pb	58	2 100	860	810	620	2	40	15	124	1108	55.3
	Essential heavy metals										
Cr	20	1 300	390	340	360	10	40	20	463	1 072	1100
Cu	370	60 000	10 000	5 400	13 000	101	2186	797	3537	13 360	-
Ni	100	7 700	1800	1 500	2000	7	148	54	427	617	190
Zn	20 000	360 000	111 000	89 000	80 000	45	21 312	7 146	18 049	19 793	2 290

(1)Faecal sludge density 1.001 kg/m3 (Radford & Sugden, 2014). (2)(Vinnerås et al., 2006). (3)(Jönsson et al., 2004); farmyard manure from organic cattle farms in Sweden. (4)(WHO, 2006). (5)Mineral fertiliser samples in 12 European countries (n = 196) (Nziguheba & Smolders, 2008)

recycling. The most simple correlation is to refer to the fertiliser value, e. g. mg heavy metal/kg P recovered. For sustainable fertiliser production, introduction of new heavy metals into the food cycle should be avoided as much as possible. The concentration of heavy metals in faecal sludge in this study and in other types of waste material in relation to the recovered P is shown in Table 4. The mean concentrations of As, Pb, Ni and Zn in faecal sludge were higher than in other waste sources and in mineral P fertiliser (Nziguheba and Smolders, 2008; WHO, 2006; Jönsson et al., 2004). However, if median concentration were used for the comparison, only Pb, Ni and Zn concentration in faecal sludge were higher than in other waste materials and in mineral P fertiliser. Farmyard manure and sewage sludge had higher Pb, Ni and Zn loads than mineral P fertiliser, while faeces and mixtures of urine and faeces had higher Zn loads. Fresh urine was the only waste fraction with lower reported heavy metal concentrations in relation to P recovered than mineral P fertiliser (Table 4). When recycling faecal sludge, it is therefore important to keep heavy metal concentrations as low as possible by reducing non-food related heavy metal loads entering the faecal sludge management system. Implementation of upstream prevention would require collaboration with stakeholders in other sectors that are the main contributors of heavy metals. In the specific case of Phnom Penh, transportation, stormwater runoff and use of household products likely contribute to heavy metal contamination of faecal sludge, but further studies are needed to identify the key contributors.

## 5. Conclusions

Heavy metal concentrations in faecal sludge samples collected in Phnom Penh were significantly higher in the rainy season than in the dry season, probably due to metal-containing inflow from stormwater drains and run-off from roads during the rainy season. Based on heavy metal load in relation to P recovered the sludge cannot be recommended to be used as fertiliser in agriculture. This present study also revealed that there is a potential of heavy metals contamination in faecal sludge in any settings that have similar context like Phnom Penh, therefore direct use of treated faecal sludge biosolids from such settings as fertiliser should be avoided to safeguard public health.

Since treated faecal sludge as biosolids would not be safe to be used as fertiliser in agriculture, sanitation stakeholders should consider different alternatives for closing nutrient loops, such as: i) source separation and reuse of different household wastewater fractions; ii) pollution prevention at upstream sources; and iii) use of biosolids as a soil conditioner together with other fertiliser for selected crops. As a short-term solution to the current lack of faecal sludge management in Phnom Penh, use of biosolids as a soil conditioner with other fertiliser or for soil production is a good alternative.

To ensure safe reuse, future studies should identify crop types that do not absorb heavy metals and suitable faecal sludge treatment methods

for Phnom Penh and other cities with similar settings. In a long term planning, potential for implementation of upstream source prevention and source separation should be further investigated since both options would have greater benefits, but also demand greater commitment and more efforts from all stakeholders to ensure successful and sustainable faecal sludge management, and thus safely managed sanitation services in Phnom Penh. Nevertheless, future research on technologies for heavy metals removal from faecal sludge should be conducted, thus making treated faecal sludge biosolids safe to be used as fertiliser.

## CRediT authorship contribution statement

Chea Eliyan: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing – original draft, Writing – review & editing. Jennifer McConville: Conceptualization, Investigation, Validation, Methodology, Supervision, Writing – review & editing. Christian Zurbrügg: Methodology, Supervision. Thammarat Koottatep: Methodology, Supervision, Writing – review & editing. Kok Sothea: Funding acquisition, Methodology, Supervision. Björn Vinnerås: Conceptualization, Funding acquisition, Investigation, Methodology, Project administration, Supervision, 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.

## Data availability

All relevant data are included in the paper.

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