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## Mercury in air and soil on an urban-rural transect in East Africa†

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There are large knowledge gaps concerning concentrations, sources, emissions, and spatial trends of mercury (Hg) in the atmosphere in developing regions of the Southern Hemisphere, particularly in urban areas. Filling these gaps is a prerequisite for assessing the effectiveness of international regulation and for enabling a better understanding of the global transport of Hg in the environment. Here we use a passive sampling technique to study the spatial distribution of gaseous elemental Hg (Hg(0), GEM) and assess emission sources in and around Dar es Salaam, Tanzania's largest city. Included in the study were the city's main municipal waste dumpsite and an e-waste processing facility as potential sources of GEM. To complement the GEM data and for a better overview of the Hg contamination status of Dar es Salaam, soil samples were collected from the same locations where passive air samplers were deployed and analysed for total Hg. Overall, GEM concentrations ranged between <math>0.0067</math> and <math>5.34</math> ng m<sup>-3</sup>, indicating significant local sources within the urban area. The municipal waste dumpsite and e-waste site had GEM concentrations elevated above the background, at 2.41 and 1.77 ng m<sup>-3</sup>, respectively. Hg concentrations in soil in the region (range 0.0067 to 0.098 mg kg<sup>-1</sup>) were low compared to those of other urban areas and were not correlated with atmospheric GEM concentrations. This study demonstrates that GEM is a significant environmental issue in the urban region of Dar es Salaam. Further studies from urban areas in the Global South are needed to better identify sources of GEM.

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### Environmental significance

Most research on gaseous elemental mercury (GEM) in the atmosphere has been conducted in the Northern Hemisphere, while the available data on atmospheric GEM concentrations from the Southern Hemisphere, particularly from urban areas in sub-Saharan Africa, are very limited. At the same time, global emission estimates indicate increasing GEM emissions in Africa. We present here for the first time spatially resolved GEM concentrations across an urban area in the Southern Hemisphere (Dar es Salaam, Tanzania), determined using a passive sampling technique. Signs of significant GEM sources were observed in the urban area, but they remain unidentified. We therefore recommend further studies in this area and other urban areas in sub-Saharan Africa to gain further insight into emissions and sources of GEM.

## 1 Introduction

Harmful effects of mercury (Hg) on human health and ecosystems have been thoroughly documented and have resulted in national, regional, and international regulation *e.g.*, in the form of the Minamata Convention.<sup>1</sup> This has led to declining Hg emissions to the environment in Europe and North America.<sup>2</sup> On the global scale however, these declines are being offset by increased emissions in Asia and Africa.<sup>2</sup> These temporal and spatial trends in Hg emissions are reflected in atmospheric Hg concentrations, where North America and Europe have seen declines.<sup>2,3</sup> On the other hand, Hg concentrations at Cape Point in South Africa show signs of increasing concentrations during recent years.<sup>4,5</sup>

Primary sources of Hg to the atmosphere include natural processes (*i.e.*, rock weathering and volcanism), though these

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municipal waste and e-waste as sources of GEM. The first study component was addressed by sampling at 24 locations along an urban-rural transect across the city of Dar es Salaam as well as 40 and 60 km upwind and downwind, respectively. The sampling locations within Dar es Salaam were selected to cover different urbanized areas, including commercial districts downtown, heavily trafficked areas, heavy industrial areas, and more and less affluent residential areas. Samples were additionally collected in the vicinity of two cement production facilities, one located 30 km south and one 20 km north of Dar es Salaam. These samplers were collected as close to the facilities as achievable (within ~1000 m) within site access restrictions. For the second study component, eight samples were collected along a transect (12 km) covering the city's main municipal waste dumpsite and an e-waste recycling facility 20 km west of Dar es Salaam. Soil samples were collected from 28 locations in February 2019, as described in Nipen *et al.*<sup>40</sup> Table S1 in the ESI† provides detailed information on sampling locations.

### 2.3 Sample preparation and analysis

**2.3.1 Analysis of PASs for Hg.** PASs for GEM were analysed at the University of Toronto Scarborough following US EPA method 7473,<sup>41</sup> thermal combustion, amalgamation, and atomic absorption spectroscopy. Detailed descriptions of the procedures are given in McLagan *et al.*<sup>23,42</sup> Briefly, the sorbent was weighed, placed in ceramic sample boats, and covered with sodium carbonate (VWR Chemicals BDH®), to prevent sulphur poisoning of the instrument catalyst tube.<sup>42</sup> The entire mass of the sorbent in each PAS was analysed (in two aliquots) to account for possible heterogeneity of Hg distribution within the sorbent. Analysis was performed using a Mercury Analyzer-3000 (Nippon Instruments).

The measured Hg content in the PAS was converted to volumetric GEM air concentrations by dividing the Hg mass by the sampling time (days) and a sampling rate (m<sup>3</sup> per day). A global calibration study determined the PAS sampling rate to be 0.135 ± 0.016 m<sup>3</sup> per day.<sup>23</sup> This sampling rate was adjusted for temperature and wind speed at each site (average for the duration of the sampling period) according to equations given in McLagan *et al.*<sup>43</sup> Location specific meteorological data were extracted from the ERA5-dataset from ECMWF (European Centre for Medium-Range Weather Forecasts).<sup>44</sup>

### 2.3.2 Analysis of soil

The soil samples were analysed at the University of Oslo, Norway. The samples were air dried at room temperature (21 °C) for between two days and one week until constant weight. The dry soils were carefully crushed before being sifted through a 2 mm sieve.<sup>45</sup>

The soil samples (~50 mg) were analysed for total Hg content using a Direct Mercury Analyser (DMA-80) (Milestone, Italy), with a similar method to the PAS analysis.<sup>45</sup> Nickel sample boats were used, and no sodium carbonate addition was necessary given the lower content of sulphur in the analysed soil compared to the PAS sorbent. Soil pH was determined

potentiometrically according to ISO10390, and effective cation exchange capacity (CEC) was determined using barium chloride adhering to ISO11260. Soil organic matter (SOM) content was determined gravimetrically by loss on ignition (LOI) at 550 °C. LOI was subsequently corrected for structural water loss according to Hoogsteen *et al.*<sup>46</sup> to determine OM content. Clay content was roughly estimated according to Ritchey *et al.*<sup>47</sup>

### 2.4 QA/QC

For the analysis of GEM in the PAS, three types of standards were analysed between the samples. Liquid standards ( $N = 14$ ) showed 98 ± 1% recovery, the NIST standard reference material (Bituminous coal; SRM-2685c) ( $N = 8$ ) showed 93 ± 2% recovery, while an in-house reference material (PAS sorbent loaded with Hg) ( $N = 6$ ) showed 102 ± 5% recovery. Concentration of Hg in field blanks ( $N = 5$ ) represented on average 12% of the Hg concentration in the samples (Table S3†). The samples were blank corrected based on the average of field blanks, after one field blank was omitted as an outlier. Detection and quantification limits (LOD and LOQ) were calculated as three and 10 times the standard deviation of the field blanks, respectively, and correspond to 0.26 ng m<sup>-3</sup> and 0.86 ng m<sup>-3</sup> using the average sampling volume. The LOD and LOQ reported here are comparable to the LODs and LOQs reported by Quant *et al.*,<sup>48</sup> and in the lower range of the LODs and LOQs reported by Snow *et al.*,<sup>41</sup> but higher than those reported by McLagan *et al.*<sup>22</sup> The two duplicate samples showed 6% and 9% deviation. This is comparable to the precision achieved by Quant *et al.*,<sup>48</sup> and at the lower end of precision achieved by McLagan *et al.*<sup>23</sup> using the same type of PAS.

For the analysis of Hg in soils, a light sandy soil JRC reference material (BCR-142) was analysed with 95.7% compliance with the certified concentration. Blank levels for the soils were low, representing <1% of the average Hg content in the soil samples (Table S6†). Soil Hg concentrations were therefore not blank corrected, and the LOD (0.0003 mg kg<sup>-1</sup>) and LOQ (0.0005 mg kg<sup>-1</sup>) were determined as the average plus three and 10 times the standard deviation of blanks ( $N = 10$ ), respectively, divided by the average sample amount. One rural sample (R-1, average concentration 0.012 mg kg<sup>-1</sup>) was analysed in five parallels, yielding a relative standard deviation of 13%.

### 2.5 Emission estimates for the urban area

GEM emissions from the urban area were estimated using a simple inflow–outflow box model of the urban atmosphere. As the Dar es Salaam region has varying degrees of urbanization, only the most central 37 of the ~90 wards of Dar es Salaam were defined as urban in this study, covering an area of approximately 100 km<sup>2</sup>. Assuming an atmospheric boundary layer at 1.2 km, the total volume of the urban atmosphere in the model was 120 km<sup>3</sup>. We assumed steady-state conditions and a well-mixed atmosphere. The background GEM concentration (air inflow) was based on Slemr *et al.*<sup>49</sup> For urban GEM concentrations (air outflow), we used the mean GEM concentrations of the urban locations found in this study after removal of outliers. Wind speed used in the model represented the mean for all



urban sampling locations in Dar es Salaam.<sup>44</sup> Table S4† provides the parameters and equations used for the urban emission model. Uncertainty in the emissions was estimated using a Monte Carlo approach including likely variability in the atmospheric boundary layer height, wind speed, and GEM concentration in inflowing and outflowing air (further described in the ESI). Predicted annual emissions from urban Dar es Salaam based on measured concentrations were compared to the gridded AMAP/UNEP global Hg emission inventory for 2010.<sup>50</sup>

## 2.6 Statistical analysis

Data treatment was conducted using Microsoft Excel and R studio version 4.0.5. Differences between location categories were tested using the non-parametric Wilcoxon rank sum test. Correlations between GEM, Hg in soil, and soil properties were determined using the Spearman rank correlation.

# 3 Results and discussion

## 3.1 General overview

The LOQ for GEM obtained in this study ( $0.86 \text{ ng m}^{-3}$ ) lies at the lower end of annual mean background concentrations in air reported for the Southern Hemisphere between 2007 and 2013 (range:  $0.85\text{--}1.05 \text{ ng m}^{-3}$ ).<sup>49</sup> Two rural/suburban locations had GEM concentrations between the LOD and the LOQ. GEM concentrations below the LOQ are reported in italics in the following text. One sample from the municipal waste/e-waste transect had a GEM concentration considerably lower than those of the blanks. This sample has thus likely been subject to some unknown instrumental error and was therefore excluded from the study. An additional PAS from the outer edge of the municipal waste/e-waste transect was lost during transport.

GEM concentrations over the study area as a whole ranged between  $0.79$  and  $5.34 \text{ ng m}^{-3}$  for the two-month sampling period (Fig. 1 and Table 1). Concentration data are provided in Table S2.†

Concentrations of Hg in soil in the present study ranged from  $0.0067$  to  $0.098 \text{ mg kg}^{-1}$  (Table 2 and S5†). Globally, background Hg content in soil varies, but reported values lie in the range of  $0.01$  to  $0.1 \text{ mg kg}^{-1}$  (Beckers and Rinklebe<sup>51</sup> and references therein). As such, the range of Hg concentrations in soil found in the region is comparable to background concentrations. Hg concentrations in soil were poorly correlated with GEM concentrations in air (Spearman's  $\rho$   $0.34$ ;  $p = 0.076$ ).

## 3.2 Urban-rural transect

**3.2.1 Air.** Urban locations in this study showed a median GEM concentration of  $1.55 \text{ ng m}^{-3}$  (mean  $2.13 \pm 1.57 \text{ ng m}^{-3}$ ). Variability in GEM concentrations within the urban area was large, with the range of  $1.07$  to  $5.34 \text{ ng m}^{-3}$ . GEM concentrations at all urban sites exceeded the highest background value ( $1.05 \text{ ng m}^{-3}$ ) of the range reported by Slemr *et al.*<sup>49</sup> for the Southern Hemisphere. The two highest GEM concentrations in the study ( $5.13$  and  $5.34 \text{ ng m}^{-3}$ ) were found at urban locations. Both these samples were collected in areas with mainly

residential character, but not in vicinity of each other, and with differences in *e.g.*, the traffic level, affluence, and proximity to industry.

The mean GEM concentrations found in urban areas in this study were compared to GEM concentrations found in other urban areas of the world in the last decade (Table 1). Only one study from the Southern Hemisphere including urban areas could be found in the peer reviewed literature, illustrating the need for the present study. This is of relevance when comparing concentrations between the Hemispheres, as background concentrations in air in the Northern Hemisphere ( $1.3$  to  $1.7 \text{ ng m}^{-3}$ ) are higher than in the Southern Hemisphere ( $0.85$  to  $1.05 \text{ ng m}^{-3}$ ).<sup>12,15,49</sup> The mean GEM concentration over urban locations found in our study was higher than GEM concentrations found in Toronto, Canada, and in Bronx, Rochester, and Beltsville, USA.<sup>16,52,53</sup> The GEM concentrations were comparable to GEM concentrations in Pretoria, South Africa; Basel, Switzerland; Taoyuan, Taiwan, and Chicago, USA,<sup>54–57</sup> and lower compared to Da Nang, Vietnam; Seoul, South Korea, and Shanghai, Beijing, Nanjing, and Guiyang, China.<sup>21,58–62</sup>

Studies assessing spatial trends of GEM using PASs in urban areas have, as far as we are aware, only been conducted in Toronto, Canada,<sup>16</sup> and in Basel, Switzerland,<sup>55</sup> Both these studies showed lower spatial variability in Hg concentrations in the urban environment (range Toronto:  $0.94$  to  $2.37 \text{ ng m}^{-3}$ ; range Basel:  $1.83$  to  $2.52 \text{ ng m}^{-3}$ ) compared to our study ( $1.07$  to  $5.34 \text{ ng m}^{-3}$ ). However, the Canadian study also contained a component where samples were collected along transects in the vicinity of industrial facilities registered as Hg emission sources. The highest measured GEM concentration amongst these samples was  $12.3 \text{ ng m}^{-3}$ , collected within a few metres of a facility handling disposal of Hg containing products. The GEM concentrations along the transects in the Canadian study decreased exponentially, approaching average urban concentrations after  $2000$  to  $3500 \text{ m}$ . The two locations in our study with GEM concentrations in excess of  $5 \text{ ng m}^{-3}$  thus suggest the presence of significant local sources of Hg. However, given the unknown nature of these sources and thus the unknown distance between the sampler and source, no quantitative estimate of the magnitude of GEM emissions from the sources can be made.

Using the mean urban GEM concentrations measured in this study (excluding the two highest concentrations as outliers), we estimated an annual emission of  $500 \pm 400 \text{ kg}$  for the  $100 \text{ km}^2$  Dar es Salaam urban area using a simple box model. Estimated annual Hg emissions from the AMAP/UNEP emission inventory for the grid cell corresponding to Dar es Salaam (covering  $3070 \text{ km}^2$ ) is lower than this, at  $20 \text{ kg}$ .<sup>50</sup> The AMAP/UNEP emission estimate for Dar es Salaam is also low compared to grid cells corresponding to other urban areas in Africa, *e.g.*, Nairobi, Kenya ( $170 \text{ kg}$ ), and Johannesburg, South Africa ( $1100 \text{ kg}$ ).<sup>50</sup> Fossil fuel combustion (notably coal combustion) and ASGM are considered the dominating Hg emission sources in Africa.<sup>2,7,63</sup> However, to our knowledge, there are no coal fired power plants and no major ASGM activity within or near Dar es Salaam. Despite the high associated uncertainty, our emission estimates based on measurements suggest that there are







Fig. 1 A: Spatial variation in the concentrations of GEM in Dar es Salaam and surrounding regions, time-averaged over a two-month period from February to April in 2019. B: Spatial variation in the concentrations of GEM along the municipal waste/e-waste transect, time-averaged over a two-month period from February to April in 2019. Base maps are produced in QGIS.

significant Hg sources within urban Dar es Salaam that are not accounted for in UNEP/AMAP emission estimates, which therefore could be biased low.

The median GEM concentration measured at rural and suburban sites of  $1.05 \text{ ng m}^{-3}$  (range  $0.79$  to  $2.16 \text{ ng m}^{-3}$ ) is consistent with the Southern Hemisphere background concentrations reported by Slemr *et al.*<sup>49</sup> Elevated GEM

concentrations at three rural and suburban locations ( $1.99$ ,  $2.05$ , and  $2.16 \text{ ng m}^{-3}$ ) indicate the presence of local emissions. Two of these sites were at the outer edges of the urban/rural transect, excluding proximity to the urban environment as an explanatory factor for these elevated concentrations. Consequently, no significant difference was seen between urban locations and rural/suburban locations ( $p = 0.27$ ). Belelie *et al.*<sup>64</sup>





combination with the lack of correlation between GEM in air and Hg in soil, this suggests that historically contaminated soil in the urban area is not a dominating source of GEM.

In rural and suburban regions, the Hg concentrations in soil ranged from 0.0067 to 0.092 mg kg<sup>-1</sup>, with a median of 0.012 mg kg<sup>-1</sup>. More remote locations had Hg concentrations below 0.015 mg kg<sup>-1</sup>, while some suburban locations showed elevated concentrations. This was particularly the case at one location where the SOM content was also high. The presence of some high Hg concentrations in suburban regions and the presence of some lower Hg concentrations in urban regions ensured no significant difference in Hg concentrations between urban and rural/suburban regions in soil ( $p = 0.23$ ).

Soil physicochemical properties (pH, CEC, SOM and clay content) are known to influence soil capacity to bind Hg and thus its spatial distribution.<sup>54,75</sup> Of these properties, SOM showed the strongest positive correlation with Hg concentrations in soil (Table S9†) (Spearman's rho 0.44;  $p = 0.019$ ); however, this correlation was strongly influenced by one location. Soils in the region generally have low SOM content, but with relatively large variability (median 1.7%, range <0.01–9.6%). Overall, it seems likely that soil Hg concentrations are influenced by the physicochemical properties of the soil in the form of the SOM content, in combination with proximity to sources, given the presence of some higher concentrations in urban locations.

### 3.3 Municipal waste and e-waste transect

**3.3.1 Air.** GEM concentrations at sites along the municipal waste/e-waste transect had a median of 1.32 ng m<sup>-3</sup> and a range of 1.08 to 2.41 ng m<sup>-3</sup> (Fig. 1B). The highest GEM concentration within this transect was found at the municipal waste dumpsite, while the two locations furthest downwind in the transect showed only slightly elevated concentrations (1.12 and 1.15 ng m<sup>-3</sup>) compared to the range reported by Slemr *et al.*<sup>49</sup> for the Southern Hemisphere background (0.85 to 1.05 ng m<sup>-3</sup>). The e-waste handler (1.77 ng m<sup>-3</sup>) and most sites in the immediate vicinity of the e-waste handler and the municipal waste site (1.08, 1.72, and 1.32 ng m<sup>-3</sup>) had intermediate GEM concentrations, identifying the municipal waste dumpsite (2.41 ng m<sup>-3</sup>) rather than the e-waste handler as the main source of GEM along this transect. On the other hand, three urban locations within the urban-rural transect that were located near identified e-waste handling or recycling sites, all showed somewhat elevated GEM concentrations (1.44, 1.69, and 1.84 ng m<sup>-3</sup>). An urban location near a decommissioned municipal waste dumpsite did however not show an elevated GEM concentration (1.10 ng m<sup>-3</sup>).

PASs have previously been applied to determine GEM concentrations near a landfill known to contain Hg contaminated materials in Switzerland<sup>76</sup> and at an e-waste handling facility in Norway<sup>11</sup> (Table 1). The GEM concentration at the municipal waste dumpsite in our study was within the lower range of GEM measured in the Swiss study. Moreover, the GEM concentration at the e-waste site in our study was lower than the range of GEM concentrations in ambient air in the vicinity of

the Norwegian facility and comparable to ambient air at some distance from it.<sup>11</sup> Overall, the spatial distribution of GEM concentrations in the present study suggests that both general waste and e-waste are sources of GEM. The relatively low levels of GEM around the e-waste handler and municipal waste dumpsite compared to similar sites in industrialized countries could be explained by an overall lower volume of Hg containing products entering the waste stream in Tanzania.

**3.3.2 Soil.** Along the municipal waste/e-waste transect, Hg concentrations in soil ranged between 0.0069 and 0.065 mg kg<sup>-1</sup>, with a median of 0.015 mg kg<sup>-1</sup>. The soil at the e-waste site (0.033 mg kg<sup>-1</sup>) had a higher Hg concentration compared to the soil collected at the edge of the municipal waste site (0.022 mg kg<sup>-1</sup>). The highest soil Hg concentration was nevertheless found at a site towards the periphery of the transect (0.065 mg kg<sup>-1</sup>). Soil properties at this site differed from the rest of the transect, with *e.g.*, higher content of SOM. In general, the soil along the municipal waste/e-waste transect had lower SOM content and lower CEC compared to the soil in the urban-rural transect (Table S8†), indicating lower capacity to retain Hg. Along the urban-rural transect, soil was sampled at two urban locations in the vicinity of e-waste processing, one of which showed the highest Hg soil concentration in the present study, while the other showed a low Hg concentration (0.098 and 0.011 mg kg<sup>-1</sup>, respectively). An urban location near a decommissioned municipal waste dumpsite also had a low soil Hg concentration (0.018 mg kg<sup>-1</sup>).

The municipal waste dumpsite had a soil Hg concentration in the lower range of concentrations reported from a municipal waste dumpsite in Sierra Leone.<sup>77</sup> Moreover, the Hg concentration in the soil at the e-waste site was lower than the concentrations reported from an informal e-waste site in Ghana.<sup>77</sup>

### 3.4 Concluding remarks

This study shows that urban ambient GEM concentrations in Dar es Salaam are lower than those found in some urban areas in Asia. Nevertheless, GEM concentrations exceeded 5 ng m<sup>-3</sup> at two locations, demonstrating the presence of significant local sources. Further investigations using multiple PASs should be conducted in the vicinity of these locations to identify sources and to facilitate estimation of the magnitude of their emissions. The comparatively high estimated annual GEM emissions in Dar es Salaam further support the presence of significant unidentified sources. This raises questions regarding current emission estimates for Africa, as the sources expected to be most important in Africa, *i.e.*, AGSM and coal power plants, are not known to be relevant for Dar es Salaam. Further studies should also be carried out in other urban regions in Africa to gain further insight into urban emissions and sources for GEM. The PAS applied in this study has been shown to be suitable to carry out such investigations in urban areas in the Global South, given the quantification limits in the lower range of GEM background concentrations for the Southern Hemisphere. Longer deployment times could further lower the quantification limits.



In addition to disclosing unidentified sources of GEM in Dar es Salaam, our study shows that handling of municipal waste and e-waste represent sources of GEM at waste sites in the outskirts of the city and that e-waste also represents a source within Dar es Salaam. Further efforts in waste management at the regional level should be made, but this should also be addressed in regulatory efforts at the international level to reduce trade of products and waste which contain Hg.

## Conflicts of interest

There are no conflicts to declare.

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