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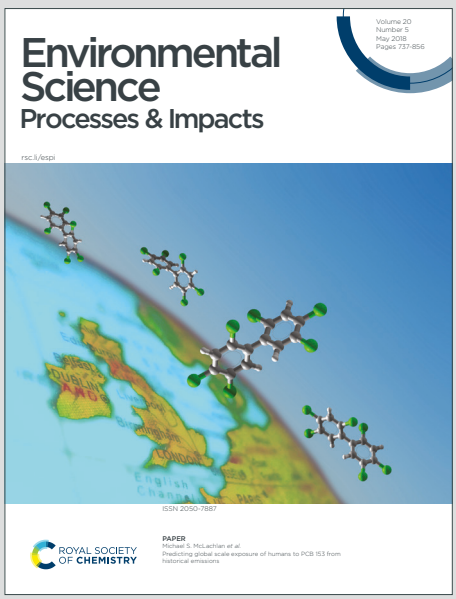

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Characterization of Inhalation Exposure to Gaseous Elemental Mercury During Artisanal Gold Mining and E-Waste Recycling Through Combined Stationary and Personal Passive Sampling

Melanie A. Snow,[1] Godfred Darko,[2] Opoku Gyamfi,[2] Eugene Ansah,[2] Knut Breivik,[3,4] Christopher Hoang,[1] Ying Duan Lei,[1] Frank Wania[1,*]

[1]*Department of Physical and Environmental Sciences, University of Toronto Scarborough, 1265 Military Trail, Toronto, Ontario, Canada M1C 1A4*

[2]*Department of Chemistry, Kwame Nkrumah University of Science & Technology, Kumasi, Ghana*

[3]*Norwegian Institute for Air Research, P.O. Box 100, NO-2027 Kjeller, Norway*

[4]*Department of Chemistry, University of Oslo, P.O. Box 1033, NO-0315 Oslo, Norway*

[*]Corresponding author: frank.wania@utoronto.ca, Tel. +1-416-287-7225

Abstract

While occupational inhalation exposure to gaseous elemental mercury (GEM) has decreased in many workplaces as mercury is being removed from many products and processes, it continues to be a concern for those engaged in artisanal and small-scale gold mining or in recycling mercury-containing products. Recently, stationary and personal passive air samplers based on activated carbon sorbents and radial diffusive barriers have been shown to be suitable for measuring GEM concentrations across the range relevant for chronic health effects. Here, we used a combination of stationary and personal passive samplers to characterize the inhalation exposure to gaseous elemental mercury of individuals living and working in two Ghanaian gold mining communities and working at a Norwegian e-waste recycling facility. Exposure concentrations ranging from < 7 ng/m^3 to >500 μg/m^3 were observed, with the higher end of the range occurring in one gold mining community. Large differences in the GEM exposure averaged over the length of a workday between individuals can be rationalized by their activity and proximity to mercury sources. In each of the three settings, the measured exposure of the highest exposed individuals exceeded the highest concentration recorded with a stationary sampler, presumably because those individuals were engaged in an activity that generated or involved GEM vapors. High day-to-day variability in exposure for those who participated on more than one day, suggest the need for sampling over multiple days for reliable exposure characterization. Overall, a combination of personal and stationary passive sampling is a cost-effective approach that cannot only provide information on exposure levels relative to regulatory thresholds, but also can identify emission hotspots and therefore guide mitigation measures.

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Environmental Significance Statement

Artisanal and small-scale gold mining (ASGM) has an economic impact on millions of people in the developing world. While the exposure of miners and their family members to mercury is widely recognized, inhalation exposure to gaseous elemental mercury is rarely assessed because existing measurement techniques are too costly, cumbersome and delicate for deployment in the challenging environments where mining is conducted. We demonstrate here that a passive air sampler-based approach can provide quantitative inhalation exposure data, simply, reliably and inexpensively. This approach should facilitate comprehensive exposure assessments and support efforts to reduce and mitigate mercury exposure in ASGM communities worldwide.

Introduction

Mercury (Hg) is a neurotoxin linked to numerous adverse health effects. While dietary uptake of methylmercury is the exposure pathway of primary concern for most people, there are sub-populations for which inhalation of gaseous elemental mercury (GEM) is the dominant route of exposure to Hg. While historically such inhalation exposure was a concern in many industrial workplaces (e.g. chloralkali plants[1], mercury mining operations[2,3] as well dental and medical facilities[4-6]), stricter regulation on the use of mercury in many processes and products has greatly reduced the number of occupational environments in advanced, industrial economies that carry the risk of hazardous exposure to GEM in the atmosphere.

Exceptions to this trend are e-waste recycling facilities, where the manual and automatic dismantling of discarded electrical and electronic equipment seeks to recover valuable materials. A large number of Hg-containing products, including fluorescent tubes, compact fluorescent lights (CFLs), batteries, and computers, are being phased out or are reaching their end-of-life. Dismantling these items generates vapors, including GEM, that could result in inhalation exposure of those working in e-waste facilities. Trade disperses e-waste globally, with large quantities being transferred from affluent to less affluent regions,[7,8] where recycling generally occurs in more informal settings, often involving open waste burning, which has the potential to release more GEM. Those living within an informal e-waste recycling region are at increased risk of heavy metal exposure.[9-12] Individuals who work in e-waste recycling plants in particular are at risk for increased Hg exposure.[13,14]

Artisanal and small-scale gold mining (ASGM) is another activity where GEM inhalation is an increasing rather than a diminishing exposure concern, as ASGM continues to grow.[15,16] ASG miners frequently use Hg as it is cheap, easily acquired, and binds with gold to form an amalgam which can be readily separated from the remaining ore.[17,18] While there is some potential for GEM inhalation during the handling of the elemental Hg liquid, the primary

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exposure concern revolves around the burning of the amalgam to remove Hg from the gold.[19,20] This is often completed using a torch or other heated device openly in public spaces. The resulting porous sponge gold contains impurities and residual Hg and is mostly sold to gold shops for further refining. Gold shops are likely to also be a significant source of GEM emissions due to the burning of residual Hg.[21] GEM emissions can be greatly reduced through the use of Hg vapor collection devices such as simple retorts although efforts to implement these in ASGM communities have yet to be successful.[22-24]

Measurements of GEM inhalation exposure typically rely on two types of approaches: Short-term measurements with portable mercury analyzers or personal samplers. For example, Li et al.[3] used a portable Lumex RA-915 Zeeman mercury analyzer to record GEM levels at various locations of the Wuchuan mercury mining area in Southwest China. Xu et al.[25] used the same instrument to map the GEM concentration in the abandoned Wanshan mercury mining area based on 30-minute measurements at 67 outdoor locations. Gyamfi et al.[26] used this approach to make instantaneous measurements of a few minutes' length at different locations within rooms in 175 households of an ASGM community in Ghana. While this approach can measure GEM over a very wide concentration range from atmospheric background to 50,000 ng/m^3, it lacks the ability for time averaging and personal sampling.

Personal sampling on the other hand provides (i) concentration values over extended periods, e.g. for the length of a work day, which is of more relevance for chronic health effects than peak concentrations, and (ii) an individual's inhalation exposure, if the sampler is wearable within the breathing zone. Pumped personal samplers require electrical power and are too cumbersome to be practical in many workplace applications, especially in challenging environments such as ASGM in developing countries. Therefore, passive personal samplers are often the preferred option. A passive sampler does not use a pump to draw air through a sorbent, but relies on the diffusion of the target compound to the sorbent. In the context of ASGM, personal passive samplers have been previously used to record GEM inhalation exposure in Burkina Faso during open burn events.[27,28] Black et al.[27] used personal Hg vapor badges, whereas de Barros Santos et al.[28] used porous nanostructures of Vycor glass, which undergo a measurable color change upon mercury uptake. Whereas the latter only provide semi-quantitative information on GEM concentrations, the former study recorded extremely high GEM concentrations, often in excess of 1,000,000 ng/m^3. These samplers are designed for assessing exposure in the vicinity of the thresholds of acute health concern but are not well suited for lower exposure levels that may still be relevant for chronic health effects and that may affect a much larger population than those participating in open amalgam burning events. Snow et al.[29] recently introduced a personal passive sampler that can achieve much lower limits of detection than the ones used previously in ASGM communities. This sampler relies on a commercial Radiello® diffusive barrier to constrain the rate of GEM diffusion to a metal mesh

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cylinder filled with sulfur-impregnated activated carbon acting as a high capacity sorbent sorbent.[29] It was shown to achieve limits of quantification lower than the lowest relevant occupational threshold during deployments lasting eight hours, while achieving accuracy and precision on par with personal pumps.[29]

Here we present measurements that were undertaken in two ASGM communities in Ghana, as well as within a regulated e-waste handling facility in Norway, in order to establish GEM concentrations in the local atmosphere and to assess the personal inhalation exposure of workers and community members to GEM. We employed two types of passive air samplers (PASs) that are based on similar design principles and rely on the same sorbent and quantification technique, namely the stationary PAS described by McLagan et al.[30] and the personal PAS introduced by Snow et al.[29]. The objectives of these field studies were (i) to test the field-worthiness and establish performance characteristics of the new PAS[29] in challenging, real-life applications, (ii) to explore the benefits of a combined deployment of stationary and personal samplers, and (iii) to provide valuable information on the chronic GEM exposure that e-waste workers and ASG miners and other community members may experience. While there have been measurements of trace elements, including mercury, in atmospheric particles collected in e-waste areas,[11,13] we believe to present here the first measurements of personal GEM exposure in the formal or informal e-waste recycling sector. With respect to ASGM, we provide the first time-averaged GEM exposure concentrations during "normal" mining operations, i.e. not focused on the open burning events during which the highest levels are expected to occur.[27,28]

Methods

Sampling Campaigns. Both stationary and personal passive samplers were deployed in two ASGM communities in Ghana and one e-waste recycling facility in Norway. In the first ASGM community, hard-rock deposits are mined within an area where individuals not participating in the ASGM sector live and work, including many women and youth. In the second community, the mining of alluvial deposits occurs approximately 1 km away from the residential area. Miners at both sites use concentrate amalgamation to separate gold from the ore. Photographs of the mining operations in the two communities are shown in the supplementary material (Figure S1 and S2). The e-waste recycling facility is a large complex that includes industrial indoor spaces, including belts, a shredder and sorting areas, as well as offices and outdoor waste storage areas. It is surrounded by forested areas. Photographs of the facility are shown in Figure S3.

Details of the three sampling campaigns are summarised in Table 1. Between 10 and 21 stationary passive samplers[30] were deployed for periods of 1 to 4 days in order to establish the GEM concentration variability in the atmosphere within and around the three sites. This PAS has an established track record as a stationary sampler for mapping spatial variability of GEM in

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outdoor environments.[31-33] At the same time, personal passive samplers[29] were worn for the length of a workday (~7 to 8 hours) by 8 to 36 workers and community members in order to record their personal inhalation exposure. Because some participants wore samplers for up to three consecutive days, the total number of personal sampling periods was higher than the number of participants. Approval for field studies with human participants was obtained from the University of Toronto Research Ethics Board (protocol number: 11365).

At the end of the sampling period, all samplers were closed with a lid and tightly sealed with Parafilm®.[29] All PASs deployed in Ghana were stored for a few days at the Kwame Nkrumah University of Science and Technology in Kumasi, Ghana before being shipped back to the University of Toronto for analysis. Samplers from Norway were shipped back to the University of Toronto immediately after the end of the campaign.

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Table 1 Description of the stationary and personal sampling conducted during three field campaigns.

	ASGM Community 1	**ASGM Community 2**	**E-waste Recycling**
Dates of sampling	Dec. 6 to 8, 2018	Dec. 14 to 15, 2018	June 3 to 7, 2019
Stationary Sampling			
Length of deployments	~ 2 days	~ 1 day	~ 4 days
Number of samples	n=20 (#311 to #330)	n=10 (#335 to #345)	n=21 (#1 to #21)
Personal Sampling			
Participants	25 total 10 for 3 days 9 for 2 days 6 for one day	36 total 14 for 2 days 22 for 1 day	8 total 5 for 3 days 3 for 2 days
Sampling periods	n=54	n=50 (- 2 lost[a])	n=21 (+ 1 stationary[b])
Length of sampling	7 hours average (6 to 10.5 hours)	7.3 hours average (~5.5 to 9 hours)	8.5 hours average (~7 to 12 hours)
Type of participants	miners (drilling, digging, washing, sieving, burning) community members (weavers, seamstresses, drivers, bar operators). Two underage children	miners (mostly diggers, crushers, soil carriers) community members (shop keeper, food seller, researchers, farmers)	e-waste sorters machine operators, driver office worker

[a]Two samplers could not be analyzed quantitatively, as all or part of the carbon sorbent was lost.
[b]One personal sampler was not worn by a person but was instead used as a stationary sampler.

Stationary Sampling. Stationary PASs were deployed by attaching them to existing structures, fences or trees, generally at a height of approx. 1.3 to 2 m above the ground, following established routines.[31,32] In Norway, 26 PASs were placed at 20 unique sampling sites (numbered #1 to #20), which included an office (#13A and #13B), four sites in industrial indoor spaces, including belts and sorting areas (#1, #2, #11, #12), and six sites in outdoor locations on the facility. One site at the shredder located in the centre of the facility (#9), and five mounted on the fence surrounding the entire facility (#3 to #7), and five sites within the vicinity of the facility mounted on trees (#14 to #18). Sample #20 was deployed furthest away from the facility to measure the atmospheric background, whereas sample #19 was deployed in a nearby town to measure the rural background. In Ghana, the sampling sites were for the most part outdoors covering both mining and residential areas. At each site, a few of the deployments were duplicated and a number of field blanks for both personal and stationary PASs were collected

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by opening a sampler for a period of approximately 10 seconds and immediately sealing it again.

Personal Sampling. Personal inhalation exposure PASs attached to commercially available sampling plates[29] were distributed each morning by researchers on-site and the deployment time was recorded by the participant on a participant card (Figure S4). Study participants were instructed to continue with a workday as they normally would if not wearing a device. If for any reason participants needed to bathe the PAS was briefly removed. Participants were also instructed to avoid touching the PAS throughout the day. Upon retrieval the end time of the sampling period was denoted on the respective participant card and samplers were placed in a storage container that was tightly sealed with Parafilm®.

Instrument Calibration and Sample Analysis. Thermal decomposition, amalgamation and atomic absorption spectroscopy (US EPA Method 7473) were used to quantify the amount of total mercury sorbed to the granulated activated carbon sorbent (Calgon Carbon Corporation, HGR-AC) in a PAS. Samples from Ghana were analyzed using the AMA-254 trace mercury analyzer (Leco Instruments Ltd., Ontario, Canada) while samples from Norway used the MA-3000 direct thermal decomposition mercury analyzer (Nippon Instruments Corporation, Tokyo, Japan). Details of the analytical technique, including the approach adopted to analyze samples with high and low expected amounts of Hg in the HGR-AC, are given in the SI.

Quality assurance and quality control measures included the analysis, every 5-10 samples, of reference materials, i.e., either a high sulfur, bituminous coal standard reference material, NIST 2685c (National Institute of Standards and Technology, Maryland USA) or an in-house prepared powdered HGR-AC sorbent loaded with mercury. Calibration checks were run every 5-10 samples by alternating the analysis of 5 and 10 ng of Hg using the 0.1 mg/L calibration standard. Clean HGR-AC was used as the analytical blank. All samples were blank corrected by multiplying the mean field blank Hg concentration of a particular experiment by the mass of HGR-AC in each sample and subtracting this value from the mass of Hg (ng) found in that sample. For each of the three field studies and both types of samplers, the method detection limit (MDL) and practical quantification limit (PQL) in ng of Hg were defined as three and ten times the standard deviation of the field blanks, respectively.

Calculation of volumetric air concentrations. Air concentrations were calculated by dividing the blank-corrected amount of Hg quantified in the carbon sorbent of a passive sampler (in ng) by the product of a sampling rate (in m^3/day) and the deployment length (in days). The sampling rate for stationary and personal samplers was taken to be 0.135 m^3/day and 0.070 m^3/day. These rates were adjusted for the local meteorological conditions during deployment using equations given in McLagan et al.[34] and Snow et al.[29]. Average temperature and wind speeds for the sampling sites were 31 °C and 3.9 m/s and 32 °C and 1.8 m/s for ASGM communities 1 and 2 respectively, and were taken from https://www.worldweatheronline.com/. The average

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temperature during the sampling at the e-waste facility was 13.5 °C, windspeed data was not available for this location, values were taken from https://seklima.met.no/observations/. The limits of detection and quantification (LOD and LOQ) in unit of ng/m^3 were defined as the MDL and PQL, respectively, divided by the sampling rate multiplied by the average deployment time. The adjusted sampling rates, MDLs, PQLs, LODs and LOQs are given in Table 2.

Results

Quality Assurance/Quality Control. Recoveries for liquid standards and standard reference materials ranging from 95-101 % indicate quantitative accuracy was maintained throughout the analysis of the samplers (Table 2). Average field blank concentrations for both ASGM communities were larger than values obtained during past studies using the sampler, while field blank concentrations from the e-waste facility were within a comparable range (Table 2).[30,31,32] The higher field blank contamination in Ghana can be attributed to the very high atmospheric GEM concentrations in the study area. While even a closed sampler can take up some GEM,[29,33] the amount of GEM that can diffuse to the sampler sorbent during the short time that field blank samplers were exposed to the ambient atmosphere is not negligible. Despite the high field blank levels, LODs and LOQs obtained for stationary PASs in both ASGM communities remained below 200 ng/m^3, which is the lowest GEM exposure thresholds of interest. In community 1, the LOD and LOQ for personal PASs exceeded the ATSDR and WHO minimum risk level (MRL) of 200 ng/m^3 but remained lower than all other thresholds. As there was only one stationary PAS field blank in community 2, it was assumed that a typical relative standard deviation of field blanks is 50 %, and a standard deviation of 1.6 ng was estimated using this value. Here, only the LOQ was found to exceed the lowest threshold. The LOD and LOQ are similar to those of community 1 as the shorter deployment period (1 instead of 2 days) cancels the lower field blank contamination. Despite the relatively large LOD and LOQ values obtained for the ASGM communities, the values remain reasonable given the high concentrations of GEM observed in the area. LODs and LOQs for stationary and personal PASs at the e-waste facility were below all thresholds of interest.

Duplicate PAS were deployed during all three field studies. One duplicate deployed in community 1 was lost due to an error during analysis. Duplicates deployed in community 2 agreed within 18 % with each other, indicative of acceptable precision considering the short deployment time of only one day. Two sets of duplicates deployed at the e-waste facility agreed within 2 and 29 % with each other. The large variability among duplicates is likely due to the relatively short deployment times, as previously reported better replicate precision was for PASs that had been deployed for much longer sampling times (often 6 months to a year).[31,32,34]

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Table 2 Summary of stationary and personal PAS meteorological and quality assurance data for Ghana and Norway field experiments

Sampling Location	ASGM – Community 1		ASGM – Community 2		E-Waste Facility	
Average Temperature (°C)	31		32		13.5	
Average Wind Speed (m/s)	3.9		1.8			
Standard Recoveries (%):						
In-house carbon	97 ± 4, (n=34)		95 ± 5, (n=19)		95 ± 1, (n=14)	
NIST Standard Reference Material	96 ± 5, (n=5)		101 ± 4, (n=17)			
Continuing Calibration Verification	101 ± 3, (n=89)		98 ± 3, (n=44)		100 ± 1, (n=14)	
PAS Type	Personal	Stationary	Personal	Stationary	Personal	Stationary
Adjusted Sampling Rate (m^3/day)	0.076	0.156	0.076	0.151	0.067	0.139
Number of Samples	55	20	52	10	21	24
Replicate Precision (%)				18 %		2-29 %
Field blank Concentration (ng/g_{HGR-AC})	5 ± 2, n=3	15 ± 3, n=2	2 ± 1, n=3	3 ± 2, n=1	0.2 ± 0.1, n=6	0.4 ± 0.1, n=3
Method Detetion Limit (ng)	6	8	3	5	0.2	0.4
Practical Quantification Limit (ng)	20	27	10	16	0.6	1.4
Average Deployment Length (days)	0.3	2	0.3	1	0.4	4
Limit of Detection (ng/m^3)	267	25	126	26	7	0.8
Limit of Quantification (ng/m^3)	889	82	420	88	23	2.5

Stationary Sampling in Ghana. All stationary PASs deployed in community 1 recorded GEM levels well above the LOQ. The geometric mean concentration was ~900 ng/m^3, with most of the concentrations ranging between 500 and 1500 ng/m^3 (data for individual samplers provided in Tables S3); the latter value exceeds hemispheric background levels by three orders of magnitude. One sampler in an indoor location recorded levels in excess of 30,000 ng/m^3. Of the nine stationary samplers deployed in community 2 (Tables S4), three recorded volumetric air concentrations below the LOD and four had levels that are above the LOD, but below the LOQ. These concentrations thus should only be regarded as semi-quantitative measures. The two samplers with levels well above the LOQ recorded concentrations of 160 and 360 ng/m^3.

Personal Sampling in Ghana. In community 1, the concentrations recorded by personal samplers ranged from <LOD to in excess of 500,000 ng/m^3, with a geometric mean concentration of 5500 ng/m^3 (Tables S5). Only four of the 55 sampling periods from this community recorded levels below the LOQ, of which two were below the LOD. 33 out of 55 samples (60 %) had levels between 1000 and 10,000 ng/m^3, 8 (15 %) had levels between 10,000 and 100,000 ng/m^3, and 6 (11 %) recorded levels in excess of 100,000 ng/m^3. Only 8 (14 %) had levels below 1000 ng/m^3. In community 2, the personal samplers recorded concentrations ranging from below the LOD to 7000 ng/m^3, with a geometric mean concentration of ~250 ng/m^3 (Tables S6). 15 out of 52 samples (29 %) had levels below 100 ng/m^3, 9 (17 %) had levels above 1000 ng/m^3, with the remaining 28 samples (54 %) falling in the range between 100 and

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1000 ng/m^3. In community 2, however, 32 of 52 sampling periods had levels below the LOQ, 16 of which were also below the LOD.

Stationary Sampling in Norway. Concentrations for only 24 of 26 deployed samplers are reported (Table S7), because the sampler from site #10 was lost during deployment and one of the duplicates at site #14 was lost during analysis. The sampler at site #8 was accidentally deployed with the lid closed, but with the Parafilm® removed during the four days in the field. It contained 0.99 ng of Hg, i.e. slightly more than the field blanks, which is consistent with the absence of a Parafilm® seal. At all locations, recorded volumetric air concentrations were higher than the LOD. Four sites (#1, #2, #11, #12) located inside the e-waste facility as well as an outdoor site next to the shredder (#9) had the highest observed GEM concentrations ranging from 31 to 1140 ng/m^3. Five outdoor locations (#3, #4, #6, #7, #18) had concentration levels above the LOQ, falling all in the narrow range from 2.8 to 3.8 ng/m^3. This indicates that the facility also contributes to the Hg contamination of the outside ambient atmosphere. The remaining nine samplers, which sampled in the office and the outdoor locations away from the facility, recorded air concentrations below the LOQ of 2.5 ng/m^3, but still above the LOD of 0.8 ng/m^3. While these concentrations thus should only be regarded as semi-quantitative, it is clear that these are levels close to the expected hemispheric background of ~1.5 ng/m^3.

Personal Sampling in Norway. Personal inhalation exposure PASs were worn during working hours by eight individuals working at the facility for a three-day period. Five of these individuals did this for three consecutive days, the remaining three did this only for the first two of these days. As such, there were 21 sampling periods. One additional sampler was not worn by a person but was instead placed as a stationary sampler near a work area. The workers included those that sort e-waste most of the time, to those that alternate between sorting e-waste and operating machinery. It also included an office worker and a driver. 18 of the 21 samplers recorded levels above the LOQ. They ranged from 69 to 3800 ng/m^3 and had a geometric mean of ~500 ng/m^3 (Table S8).

Discussion

Comparison with measurements in other occupational settings. Table 3 compares the GEM concentration measured here with those that have been reported for various occupational and industrial settings previously. The levels in the Norwegian e-waste facility are comparable to those that have been measured in health care facilities in Poland[5] and Mongolia.[6] The GEM contamination in those settings are attributed to broken instruments and devices containing elemental Hg.[5,6] We similarly attribute the GEM in the Norwegian e-waste facility to broken devices such as fluorescent bulbs and tubes. In a residence where Hg from a manometer had been spilled, concentration of 750-3700 ng/m^3 and 220-1500 ng/m^3 prior and after clean-up operations, respectively have been reported.[5] It appears that average concentrations indoors in the 100s of ng/m^3 and peak time-weighted average (TWA) concentrations of a few 1000s of

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ng/m^3 can be expected as a result of broken equipment containing Hg, with the actual levels depending on the size of the source (number of broken items and their Hg content) and the room ventilation rate. Similar indoor air contamination might be expected from Hg use for cultural and religious practices.[35]

The concentrations in ASGM community 2 in Ghana are similar to the concentrations reported for former mercury mining areas in China[3,25] and Italy,[32] ranging from close to background levels at sites removed from sources to 10,000 ng/m^3 and above at the most contaminated sites (e.g. abandoned smelting workshops[3]). This is probably typical for outdoor environments that are heavily contaminated with elemental mercury, but where there are no or only limited activities that result in the volatilization of GEM at high temperature, such as occurs during active smelting of Hg containing ores or the open burning of amalgam.

The higher end of the concentrations in ASGM community 1 is comparable to what Mnizek[1] has reported as the exposure of workers within two active chloralkali plants in Poland. This suggests that those living in the parts of ASGM communities where regular amalgam burning is taking place can be exposed to time-weighted average GEM concentrations in the range of 10,000s to 100,000s of ng/m^3, with the higher end of the range more likely to occur in indoor environments.

Comparison with measurements reported previously in ASGM communities. Gyamfi et al.[26] had previously measured GEM concentrations in numerous households of community 1 using a portable Hg analyzer. Concentrations in households reporting burning of amalgam averaged ~14,000 ng/m^3 whereas those without such burning were much lower at around 500-800 ng/m^3. Concentrations recorded outside of burning and non-burning households averaged 2800 and 300 ng/m^3, respectively.[26] These values are entirely consistent with the values recorded by the stationary samplers here, with a geometric mean of 900 ng/m^3 and a range from 400-30,000 ng/m^3, whereby the maximum also was an indoor location.

Black et al.,[27] using SKC badges, recorded extremely high GEM inhalation exposure for gold miners in Burkina Faso during open burn events. Concentrations averaged over periods ranging from 1 to 4 hours were ~7,000,000 ng/m^3 for burners and ~1,400,000 ng/m^3 for bystanders. This is much higher than the values recorded by the personal samplers in this study, where the highest value was ~500,000 ng/m^3 and the geometric mean ~5000 ng/m^3. This difference can be explained by the different sampling strategies. Black et al.[27] aimed at recording the maximum expected, acute exposure levels of those engaged in open amalgam burning and those in their immediate vicinity. Our sampling was focussed on recording the day-to-day chronic exposure to GEM that those working and living in an ASGM community might expect.

Table 3 Comparison of the GEM concentrations measured in this study with those reported for occupational and industrial settings in the literature.

Type of environment	Sampling method	Range ng/m³	mean ng/m³	Reference
ASGM community 1, Ghana	Stationary passive sampler	360 to 31,400	900 (geomean)	This study
	Personal passive sampler	<267 to 530,000	5500 (geomean)	
ASGM community 2, Ghana	Stationary passive sampler	<26 to 360	42 (geomean)	
	Personal passive sampler	<126 to 6800	280 (geomean	
E-waste recycling, Norway	Stationary passive sampler	0.9 to 1,140	5 (geomean)	
	Personal passive sampler	<7 to 3,850	500 (geomean)	
Chloralkali plants, Poland	Personal active sampling	23,000 to 150,000	60,000 (geomean)	1
Hospital, Poland	Stationary sampling	<20 to 14,000	150 to 200 (medians)	5
Dental clinics, Sweden	Personal active sampling	600 to 10,400	1,800 to 2,100 (median)	4
Health care facilities, Mongolia	Portable Hg analyser	3 to 2,700	200	6
Mercury mining area, China	Portable Hg analyser	20 to 40,000		3
Mercury mining area, Italy	Stationary passive sampler	1.3 to 6,700	112 to 242	32
Mercury mining area, China	Portable Hg analyzer	1.3 to 800	16 (geomean)	25

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Variability in GEM levels and exposure between the two ASGM communities. The two ASGM communities were exposed to very different levels of GEM contamination. Stationary and personal sampling are consistent with each other in terms of showing that GEM levels in community 1 were much higher than in community 2. The lowest atmospheric concentration measured with a stationary PAS in community 1 was similar to the highest level in community 2 (360 ng/m^3). The average exposure recorded with personal PASs in community 1 (geomean > 5000 ng/m^3) was ~20 times higher than that observed in community 2 (geomean ~250 ng/m^3).

This difference between the two communities could potentially be attributed to sampling location and community practices. Community 2 is close to a bigger city, where the miners bring their amalgams to be burnt in gold shops, while community 1 is remote and therefore miners have to do the amalgam burning themselves. We also believe that the GEM concentrations obtained in the second ASGM community may be biased low. ASGM, or galamsey as it is referred to by the locals, was under intense regulation from local authorities during our visit. Although the community members agreed to participate, they were reluctant to talk about or admit to the process of Hg burning. Researchers observed large quantities of Hg in use during the sampling campaign which does not appear to be reflected in sampling results. One explanation for this could be that individuals from community 2 completed the burning of amalgam at night when there was no personal sampling being completed. Although stationary samplers remained overnight, community reception of the project limited the placement and deployment of these samplers. This is in sharp contrast with the reception in community 1, where the project was openly welcomed with curiosity and a willingness to participate. This likely allowed the results obtained to more accurately reflect GEM exposure in the community. This difference in the extent of community participation highlights the challenge of conducting effective exposure characterization when emission-generating activities have to be conducted clandestinely to avoid repression by authorities.

Spatial variability within concentrations measured by stationary samplers. Figure 1 shows the concentrations measured with the stationary samplers on a map of community 1 and also displays the types of activities that are taking place in different areas of the community. It indicates that the highest levels are prevalent in the Southwest of the area. This overlaps with the area where most of the community members spend their time. Notably, the mining areas in the Southeast of community 1 had relatively lower Hg concentrations. The high concentrations are likely associated with the parts of the community where the burning of amalgam occurs most frequently. In community 2 the two samplers with the highest recorded levels were also within the community and near a location where one individual was seen burning amalgam.

In the e-waste facility, GEM concentrations recorded by the stationary PASs suggest that the main source of gaseous mercury contamination is found in the indoor e-waste sorting area (probably related to the presence of broken, Hg-containing fluorescent tubes and CFL bulbs),

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whereas much lower values prevail outdoors. The one outdoor location with high GEM levels was immediately adjacent to the shredder.

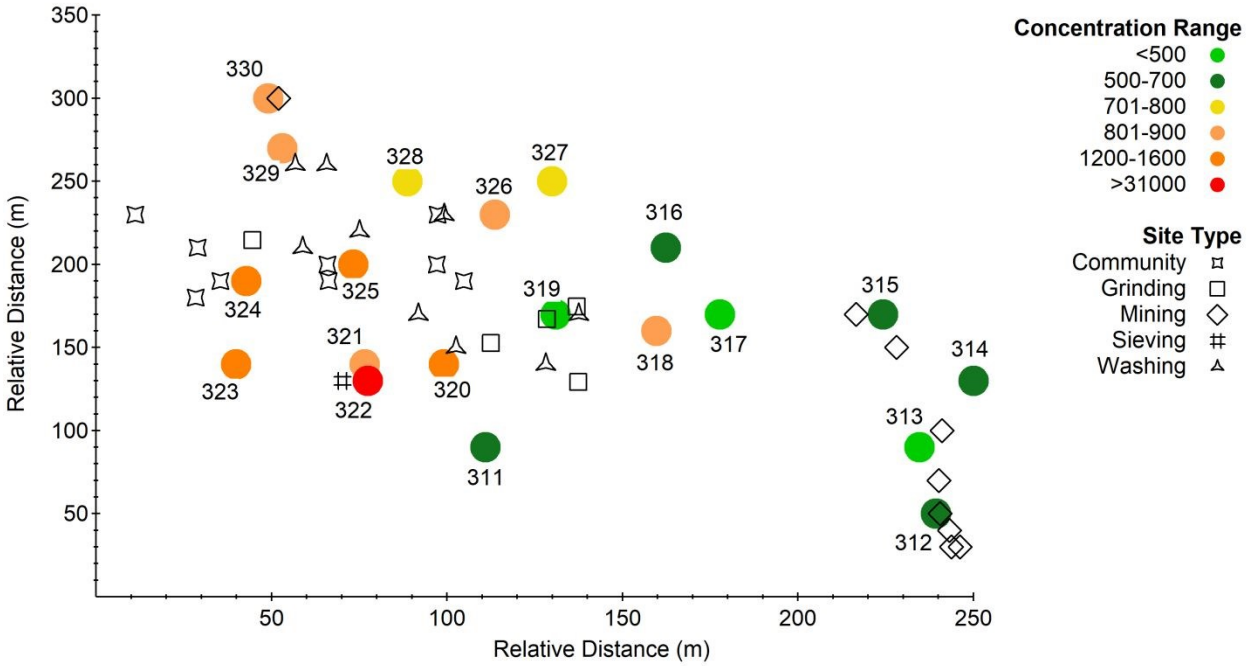

Figure 1 Map of measured GEM air concentrations in community 1 outlining the distribution of mining activities in the community. The numbers indicate sampler ID.

Personal exposure variability within communities. Not only between, but also within the ASGM communities the measured personal exposure varied widely. The variability can to a large extent be related to the activities in which community members are engaged. In community 1, exposure levels in the 1000 to 10,000 ng/m^3 range were prevalent for those describing their work as “sieving”, “seamstress”, “supervisor” and “bar operator”. Higher exposure levels in excess of 10,000 ng/m^3 were more commonly encountered among those engaged in “digging, washing” or “burning, crushing, digging”. However, among the most exposed in the community counted the “driver” and, remarkably, one of the students. Only two professions (“chop bar operator”, “drilling”) were associated with exposure consistently below 1000 ng/m^3. In ASGM community 2, those who reported to be engaged in crushing and digging were more likely to have inhalation exposure in excess of 1000 ng/m^3, than those describing their work as “soil carrier” or those not engaged in gold mining activities.

Entirely consistent with the findings from the stationary PASs, in Norway those working directly with the sorting of e-waste experience higher exposure to gaseous mercury than those working more in other parts of the facility, in particular in the outdoors. The concentrations recorded by the three samplers worn by the office worker were all below the LOD. The individual, who was mostly a driver within the facility and did not participate in the waste sorting activities was

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clearly exposed to lower concentrations (geometric mean over three days of ~100 ng/m^3) than the other workers (geometric mean of 737 ng/m^3, n=15). Whether those who only participate on alternating days in waste sorting activities had a lower exposure level than those who do this every working day could not be shown with statistical significance, mostly because of large day-to-day variability in measured exposure (see next section). One such individual, though, had low exposure levels on days 1 and 3 (< 82 ng/m^3) yet very high exposure on day 2 (~2750 ng/m^3).

Personal exposure variability between days. At all three field locations, personal exposure was measured on more than one day, allowing for an exploration of day-to-day variability. Such variability was apparent in all three cases. When comparing the different days of sampling in ASGM community 1, the 17 samplers deployed on the first day had a lower geometric mean of 3200 ng/m^3 when compared to each of the 19 samplers deployed on the second (6300 ng/m^3) and third day (7700 ng/m^3). Among those that participated on both days of sampling in ASGM community 2, some experienced widely different exposure on the two days. For example, the exposure of 4 out of 14 participants with two days of sampling deviated by more than an order of magnitude. For others, the exposure on the two days was similar; for example, the exposure of 5 out of 14 participants with two days of sampling was within a factor of 2. In Norway, there were large differences in the personal exposure measured on different days of sampling. Specifically, concentrations on day 2 (June 5, 2019, geometric mean of 2161 ng/m^3, n = 6) were much higher than on day 1 (June 4, 2019, 381 ng/m^3, n = 6) or day 3 (June 6, 2019, 321 ng/m^3, n = 3). This variability likely arises from day-to-day variability in the activities of the participants. For example, weather conditions in Norway were inclement on the second day of sampling, which may have resulted in more work being done inside than on the other two days, when weather was good. We might also expect day-to-day variability in the strength of the GEM sources. For example, in the e-waste facility, it may depend on the number and mercury content of items processed and broken during a particular day, whereas in the ASGM communities it likely is strongly influenced by the number of open burning events occurring during a day.

Comparison of measured exposure with chronic exposure thresholds. LOD and LOQ values obtained for each field study were low enough to allow for classification of the volumetric air concentrations obtained for each participant relative to the applicable regulatory threshold level for GEM (Figure 2). In community 1, all but one of the samples exceeded the ATSDR and WHO chronic inhalation MRL of 200 ng/m^3. 22 % of the samplers exceeded the ACGIH TLV of 25,000 ng/m^3, 18 % exceeded the NIOSH REL of 50,000 ng/m^3, and 11 % exceeded the OSHA PEL of 100,000 ng/m^3. In contrast to Black et al.,[27] we did no observe levels above 10,000,000 ng/m^3, which are considered Immediately Dangerous to Life and Health (IDLH).

More than half of the PASs in community 2 recorded levels above the chronic inhalation MRL of 200 ng/m^3; four PASs exceeded this limit by more than an order of magnitude. In the e-waste

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facility, most sampling periods (n=14) recorded levels above this MRL. Three sampling periods exceeded that level by more than an order of magnitude. The Norwegian recommended threshold value of 20,000 ng/m^3 was not reached during any of the personal sampling periods, although three of the PASs were within an order of magnitude of that value (i.e. exceeded 2000 ng/m^3).

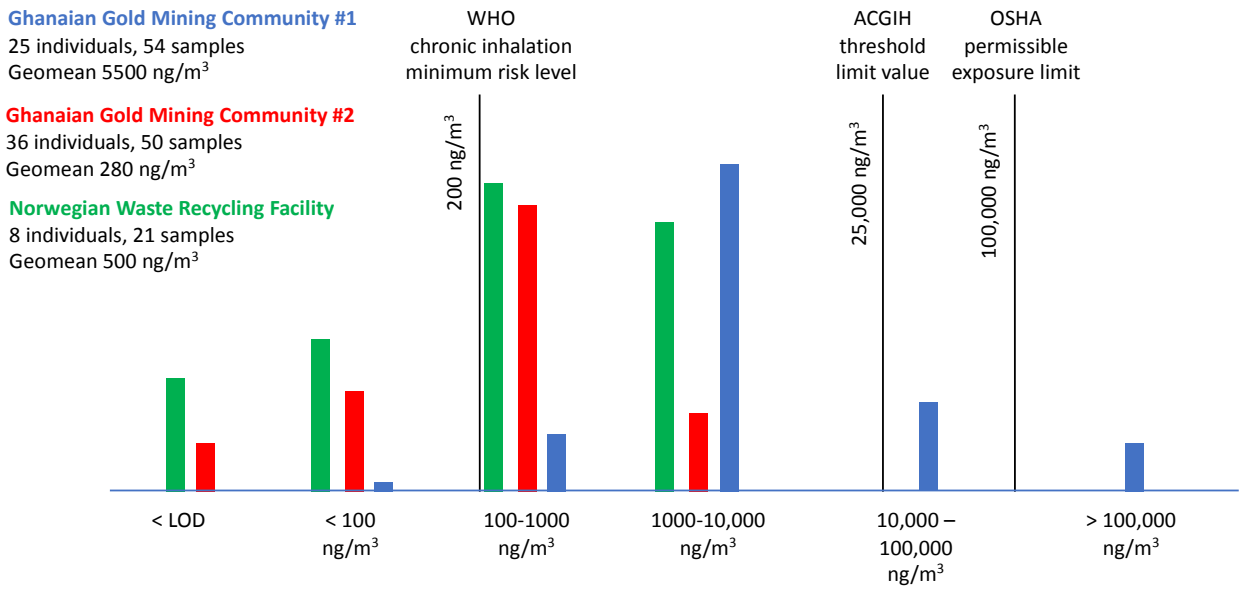

Figure 2 Frequency distribution of volumetric air concentrations measured with personal passive air samplers in ASGM communities compared to GEM occupational exposure limits

Comparing stationary and personal sampling. Interestingly, at all three investigated locations the concentrations recorded in personal PASs were considerably higher than in the stationary PASs. In the e-waste facility, the concentrations recorded in personal PASs often exceeded the highest concentration measured in a stationary sampler (1140 ng/m^3), in one case by a factor of three (~3500 ng/m^3). In both ASGM communities, only a single stationary PAS recorded a level higher than the geometric mean concentration of the personal PASs. The highest concentration recorded by a personal PAS exceeded the highest concentration measured by a stationary PAS by a factor of 19 in community 2 (6,800 vs. 370 ng/m^3) and a factor of 17 in community 1 (530,000 vs. 31,000 ng/m^3). This is important as it suggests that stationary sampling may strongly underestimate the personal inhalation exposure of those living in an ASGM community or those working in an industrial setting.

Uncertainties in the sampling rates of the two different sampler configurations are far too small to explain these divergent concentrations recorded by stationary and personal sampling.[29,34] One possible reason for this difference is that personal sampling was only done during the day, whereas the stationary sampling included nights. Lower levels during nighttime would cause personal PASs to give higher readings. It is very likely that GEM levels are higher during the day

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when the human activities responsible for Hg emissions take place (e.g. amalgam burning, e-waste sorting, breaking Hg-containing devices). While higher temperatures during the daytime may increase also surface emissions from the ground, this is unlikely to be important relative to the high primary anthropogenic emissions. Another reason for higher personal exposure concentrations is that people are likely to be closer to the main sources of GEM than the stationary sampling locations. This is plausible, because human activities generate gaseous Hg emissions, and the individuals for whom high personal exposure was recorded spend much of their day time in the locations with higher GEM concentrations, i.e. the indoor waste sorting area in Norway and gathering places and workplaces in Ghana (see also Figure 1).

A more ambitious sampling campaign could investigate the consistency between stationary and personal sampling more rigorously. For example, interpolating the concentrations recorded by a network of stationary passive sampler can be used to generate maps of GEM concentrations,[31-33] while personal passive samplers can record the exposure of persons within the same area. If the trajectories of persons moving within the space covered by the map can be recorded (e.g. with mobile phone data), it should be possible to directly compare the levels recorded by the personal samplers with the personal exposures calculated by weighing the concentrations in different locations with the time a person spends in these locations.[36] However, this assumes that temporal changes in the concentrations at a location are minor.

Conclusions

Field-worthiness and performance characteristics of the new PAS. The three field tests confirmed the suitability of the personal sampler introduced by Snow et al.[29] to be used in challenging, real-life applications. The sampler proved to be sufficiently robust and sturdy, yet also unobtrusive, to be worn during a wide variety of activities, including strenuous work in the outdoors, as frequently encountered during ASGM. The number of samplers were lost due to breakage or malfunction was very small (n=3 over all three study locations). Importantly, the sampler succeeded in recording exposure concentrations ranging from below 200 ng/m^3 to in excess of 500,000 ng/m^3, i.e. over more than four orders of magnitude and covering the entire range of exposure thresholds relevant for chronic health effects.

The divergence in the LOD and LOQ between the three different sites highlights the need to determine the LOD and LOQ of the PASs for each campaign and therefore the need for a suitable number of field blanks. The relatively high LOD and LOQ in the Ghanaian deployments were largely a result of the field blank procedure, which involved a short exposure of an opened PAS to the local atmosphere. In future deployments in environments with very high GEM concentrations, we recommend a field blank procedure that does not include opening the sampler, as this is likely to lead to much lower LODs and LOQs.

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We have not tested the PAS to measure the exposure to GEM of those actively engaged in amalgam burning, which is expected to be another one to two orders of magnitude higher than the highest exposure recorded here.[27] The highest concentration that can be determined with the PASs is constrained by the analysis of the HGR-AC. If the amount of Hg sorbed to the HGR-AC is too high, it could be outside of the range of the instrument and may in fact contaminate the instrument. This can be addressed by (i) analyzing only small aliquots of the sorbent within a sampler and (ii) by limiting the deployment length.

Benefits of a combined deployment of stationary and personal samplers. In all three deployment locations, stationary and personal passive samplers provided complementary, exposure-relevant information. The combination of the two approaches offers a way to obtain a comprehensive, spatially resolved map of background GEM concentrations while simultaneously obtaining individual exposure information. Stationary sampling alone is unlikely to provide realistic estimates of personal exposure, because of constraints on when and where the sampling takes place. This implies that personal sampling is advisable to obtain information on the exposure of workers and community members. On the other hand, stationary samplers can be used to identify the locations and therefore the activities or materials that contribute most to personal inhalation exposure. Clearly, this is a key piece of information required for mitigating any hazardous exposure that may be identified. For example, eliminating extended storage of the suspected main contaminant sources (e.g. broken fluorescent tubes or other items stored in open containers) in the e-waste facility would greatly reduce GEM concentrations. Similarly, conducting amalgam burning no longer indoors but in well-ventilated outdoor spaces away from community gathering places would be a very effective exposure reduction effort in ASGM community 1.[26]

The combination of stationary and personal passive sampling approaches based on the same sorbent and therefore the same analytical quantification technique, should allow for efficiencies in instrument acquisition, maintenance, and operator training. The fact that the stationary version of the PAS has already been widely tested and characterized adds confidence to the measurements obtained with the version designed for personal sampling.

Chronic GEM exposure of e-waste workers and ASG miners and other community members. We believe to have conducted the first measurements of the personal exposure to GEM of ASGM workers and community members, that are not directly engaged in amalgam burning activities. Inhalation exposure is often an acknowledged data gap when seeking to characterize the exposure of ASGM communities to mercury.[37] While we found large differences between two ASGM communities, in one of them we observed that almost everyone was exposed to time-weighted average levels in excess of the ATSDR and WHO chronic inhalation MRL of 200 ng/m^3, and many were exposed to much higher concentrations. Even within a regulated waste facility in Norway, 7 out of 21 time-averaged personal exposure measurements exceeded 1000

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ng/m^3 during a work day. Clearly, many GEM exposure concerns remain and there is thus a continued need for the monitoring of the inhalation exposure to GEM. We hope that the presented passive sampling techniques will aid the characterization of GEM inhalation exposure in future studies investigating the health effects of mercury use in ASGM communities. The results from this study may provide suitable guidance on an appropriate and representative sampling strategy, e.g. with respect the number of days to sample personal inhalation expsoure or where within a community stationary samplers should be placed.

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Conflict of Interest

Tekran Instruments Corp. has entered into a licensing agreement with the University of Toronto for the commercialization of the PAS used in this study under the *Mer*PAS® name, which results in some license fees being distributed to FW.

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