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Letter

Concurrently Measured Concentrations of Atmospheric Mercury in Indoor (household) and Outdoor Air of Basel, Switzerland

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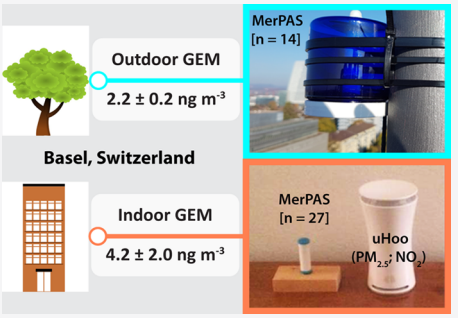
ABSTRACT: Indoor air pollution can be a major health risk because urban populations spend up to 90% of their time in closed rooms. Gaseous elemental mercury (GEM) has not been measured as routinely as other indoor air pollutants due to the high costs and limited mobility of active Hg analyzers. However, household GEM concentrations may exceed Hg air quality guidelines as a result of potential indoor GEM sources like broken Hg thermometers. Here we deploy novel low-cost mercury passive air samplers (MerPAS) in 27 households (7 days) and at 14 outdoor locations (29–31 days) in Basel, Switzerland. Average Hg concentrations ranged from 2.0 to 10.8 ng m^{-3} indoors and from 1.8 to 2.5 ng m^{-3} outdoors. These results reveal that households are a net source of Hg to the urban atmosphere and exceed outdoor Hg levels by a factor of 2 on average. We estimated an average weekly intake rate of 0.01 μg of Hg/kg of body weight for adult residents in Basel, which is usually lower than Hg exposure of people with dental amalgam fillings. Our campaign demonstrates that air monitoring programs can easily be complemented by straightforward Hg measurements using MerPAS.



INTRODUCTION

Air pollution is one of the greatest environmental risks to human health, causing ~7 million deaths globally each year.[1] In Europe, the health of urban populations is particularly affected by the major air pollutants, including particulate matter (PM) largely from combustion, dust from construction and natural sources, and nitrogen dioxide (NO_2) derived mainly from road traffic.[2] Indoor sources of these pollutants include smoking, cooking, and burning wood or candles.[3] Emissions of toxic metals (arsenic, cadmium, nickel, lead, and mercury) add to the atmospheric burden of air pollutants.

Mercury (Hg) is a global pollutant that is emitted to the atmosphere and subject to long-range global transport.[4] Atmospheric Hg is either emitted from anthropogenic Hg sources (~2.5 × 10^6 kg $year^{-1}$), mobilized naturally from the Earth's crust through volcanic eruptions and rock weathering (~0.5 × 10^6 kg $year^{-1}$), or re-emitted from ocean and land surfaces.[5,6] Urban centers contribute to the global atmospheric Hg burden mainly through fossil fuel combustion (e.g., coal combustion in power plants and residential heaters[7,8]), waste incineration, hospitals/dental facilities, and cremation.[9−11] Mean urban atmospheric Hg concentrations have been found to range from 1.46 ng m^{-3} in Toronto, Canada,[9] to 9.72 ng m^{-3} in Guiyang, China[7] (see Table S1 in section S1). The northern hemispheric background concentration is approximately 1.5 ng m^{-3}.[12]

Indoor Hg air concentrations can be elevated due to Hg evaporating from past spills of liquid Hg contained in thermometers or fluorescent light bulbs and Hg switches[13] or from Hg-containing biocide added to paint employed in buildings between 1950 and 1990.[14] A previous study estimated that the level of gaseous Hg in 10% of U.S. households exceeds the U.S. EPA reference concentration of 300 ng m^{-3}.[15] Previous studies of Hg pollution of indoor air, largely conducted in workplaces, revealed maximum Hg levels of 28.5 ng m^{-3} in Toronto,[16] 522 ng m^{-3} in New York,[15] and 1293 ng m^{-3} in Chongqing.[17] Chronic exposure to elevated ambient Hg concentrations may produce harmful effects on the nervous, digestive, and immune systems, the lungs, and the kidneys.[18]

While a few studies reported indoor Hg levels at workplaces,[15−17,19] Hg concentrations in households are usually not systematically investigated. Comprehensive studies of household Hg pollution are lacking because of the high costs and limited mobility of active measurement systems, which require a power supply and in some cases compressed carrier gas. Active systems are therefore ill-suited for Hg measurements in multiple households simultaneously.[9] An alternative approach to measuring gaseous elemental mercury (GEM) in households is deploying novel mercury passive air samplers (MerPAS), which provide the necessary accuracy and precision for atmospheric Hg monitoring.[20,21] The MerPAS can be

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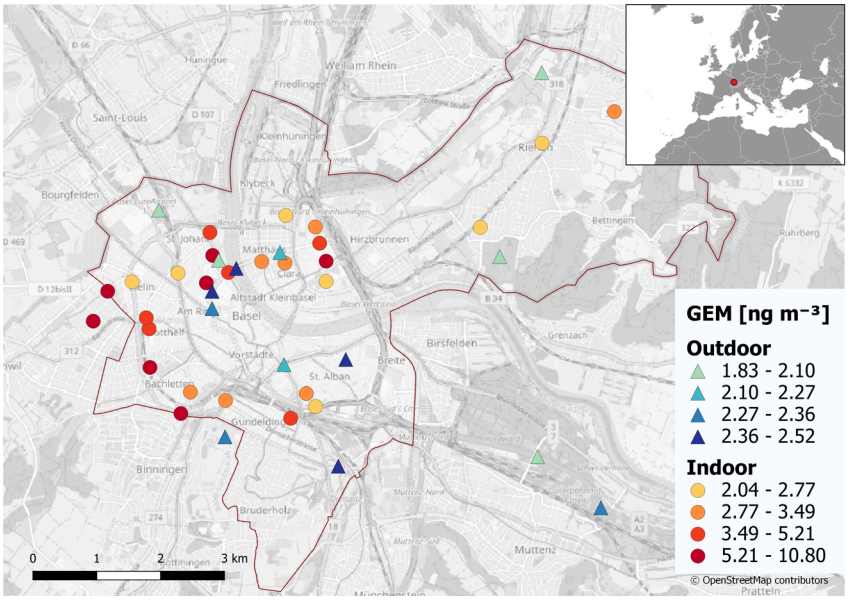

Figure 1. Sampling locations of air pollution measurements in the Basel metropolitan area. Indoor (circles) and outdoor (triangles) GEM concentrations (nanograms per cubic meter) are given in 25 percentile bins. The Basel city limits are indicated by the brown contour line. Base map reproduced with permission under a Creative Commons CC BY-SA 2.0 license from OpenStreetMap (accessed February 26, 2020).

exposed indoors or outdoors for extended periods (1 week to 1 year) without maintenance work and are therefore well suited for large-scale and long-term GEM measurement campaigns.

The objective of this study was to determine the average GEM exposure of residents in the city of Basel. To achieve this goal, we deployed MerPAS to study GEM concentrations in indoor air of multiple households and urban outdoor air simultaneously. In both indoor and outdoor settings, potential GEM sources were examined. We tested the potential of MerPAS to complement short-term air monitoring campaigns of major air pollutants with GEM measurements.

MATERIALS AND METHODS

We measured indoor and outdoor GEM concentrations using commercially available MerPAS.[20,21] MerPAS almost exclusively collect GEM because only ≤6% of the gaseous oxidized Hg (GOM) can pass through the diffuse barrier,[22] and GEM is the dominant atmospheric Hg species (typically >90% of total atmospheric Hg: GEM, GOM, and particulate Hg).[12] MerPAS are thus considered to collect GEM[22,23] (see section S2 for details). We determined the level of GEM in 27 households along with major air pollutants ($PM_{2.5}$ and NO_2) and at 14 outdoor sites in Basel, Switzerland (Figure 1). The greater triborder region of Basel has a population of about 852000 inhabitants and is located at the southern end of the Upper Rhine Valley.[24] The location of the 27 households was determined on the basis of recruitment into the EU Horizon 2020 project ICARUS (Integrated Climate forcing and Air pollution Reduction in Urban Systems) that aims to assess air pollution in nine European cities.[25] Recruitment of households did not follow a formal sampling design. Rather, households were recruited via advertisement on social media and the institute Web site, and flyers were distributed to households on selected streets in an attempt to broadly capture areas with both higher and lower ambient air pollution and socioeconomic status. For the indoor GEM measurements, we followed a previously described setup[21,26] of a sulfur-impregnated activated carbon (AC) sorbent with a white Radiello diffusive barrier. The white Radiello was screwed onto a wooden platform without a protective shield. Subsequently, MerPAS were deployed at heights of 1−1.5 m in 27 living rooms for 7 days. The average indoor air temperature was 21.3 °C. Along with GEM concentrations, major air pollutants were monitored in households using the commercial uHoo indoor air quality sensor (uHoo Limited, Hong Kong). See details about the uHoo sensor in AQMD[27] and in section S6. The NO_2 and $PM_{2.5}$ measurements from one household were not included due to device malfunction.

Outdoor GEM concentrations were measured at 14 sites using MerPAS with white Radiello with a protective shield[20,21] between November 5 and December 7, 2018. The average outdoor air temperature during this period was 6.6 °C. Data from simultaneous temperature and wind speed monitoring were available from eight outdoor MerPAS sites. Ten MerPAS were installed in the vicinity of possible Hg point sources, and four MerPAS in city outskirts and residential areas (see photodocumentation of the outdoor MerPAS setup in Figure S2 in section S8).

The total mass of Hg in the sulfur-impregnated activated carbon sorbent (AC) of each MerPAS was measured by thermal desorption, amalgamation, and atomic absorption spectroscopy using a DMA-80 instrument (Milestone Inc.). Sections S3 and S4 give details about the GEM analysis and respective quality control. The mean replicate precision and overall uncertainty of MerPAS outdoor deployments are 4% and 9%, respectively, which were determined through the analysis of hundreds of replicated samples deployed around the globe.[20] This is similar to the uncertainty of active sampling instruments.[28,29] The uncertainty of indoor deployments has been estimated to be double that of outdoor deployments because indoor deployments are predominantly subject to wind speeds of <1 m s^{-1}; wind speeds in this range are known to result in increased variability on the MerPAS sampling rate compared to wind speeds of >1 m s^{-1} typical of outdoor deployments.[26] We estimate the replicate precision and overall

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uncertainty of the indoor measurements to be 8% and 18%, respectively (see section S5).

We calculated GEM concentrations (nanograms per cubic meter) by dividing the quantified mass of Hg (nanograms) on the AC sorbents by the product of the corresponding deployment time (days) and sampling rate (SR, cubic meters per day). Indoor air concentrations were calculated using a SR of 0.156 m^3 day^{-1}.[21] The SR was not adjusted for temperature or wind speed for these deployments.[21] Outdoor GEM concentrations are based on a SR of 0.111 m^3 day^{-1}. The base sampling rate is recommended for commercially distributed MerPAS by Tekran Inc.[30,31] Sampling rates depend on the MerPAS configuration and are listed under the following link that will be updated over time: https://www.tekran.com/files/MerPAS-Config-Options.pdf (accessed March 13, 2020). At sites where temperature and wind speed data were available (B01−B08 and LH4), the SR was changed according to experimentally determined adjustments.[26] Adjustment of the SR using sampling site specific meteorological data increased the GEM concentration between 2% and 9%. For sites at which wind speed (LHA01−LHA03) and both temperature and wind speed data were missing (RFH and KVA), the sampling site specific SR was estimated by using site elevation and a linear interpolation between temperatures or wind speeds recorded at the other sites.[32] The higher SR for indoor deployments was attributed to the absence of the protective shield, which is expected to increase the diffusive path length of the samplers.[21,26] The protective shield was not used indoors for the following reasons. (i) There is no precipitation. (ii) There is no benefit from reducing wind speeds that are already typically <1 m s^{-1} indoors.[26] (iii) Given the short-term nature of the deployments, a higher sampling rate was desirable to maximize the mass of sorbed Hg in the sampler.

RESULTS AND DISCUSSION

The average indoor GEM concentration was 4.2 ± 2.0 ng m^{-3} (range of 2.0−10.8 ng m^{-3}) and significantly higher than the average outdoor concentration of 2.21 ± 0.20 ng m^{-3} (range of 1.83−2.52 ng m^{-3}) [Wilcoxon two-sample t test; $p < 0.01$ (Figure 2)]. Thus, GEM diffusion occurs from higher indoor to lower outdoor GEM levels. Consequently, households were a net source of Hg to the urban atmosphere. Mean indoor concentrations of simultaneously monitored $PM_{2.5}$ and NO_2 were 15.5 ± 10.3 and 25.4 ± 9.0 μg m^{-3}, respectively (Figure 2), with higher levels measured in homes where the survey revealed known sources such as smoking or burning candles. Indoor air pollution levels from the other case study cities in ICARUS, using the same protocol and measurement equipment, were not yet available for comparison. Average indoor $PM_{2.5}$ concentrations in Basel, however, were previously measured (21.0 ± 16.7 μg m^{-3}) in 41 households from October 1996 to March 1998.[33] Compared to those of other cities measured in the same study, the average $PM_{2.5}$ concentrations were considered moderate in Basel, low in Helsinki (9.5 μg m^{-3}), and remarkably higher in Prague (34.4 μg m^{-3}) and Athens (35.6 μg m^{-3}). Average indoor NO_2 concentrations, measured during a similar earlier time period, were 8.3 ppb (~15.6 μg m^{-3}) in Geneva, with levels among the 15 countries worldwide ranging from 10.3 to 117.9 μg m^{-3}.[34] Both of these earlier studies[33,34] used passive samplers deployed for 48 h, which may in part contribute to the lower values.

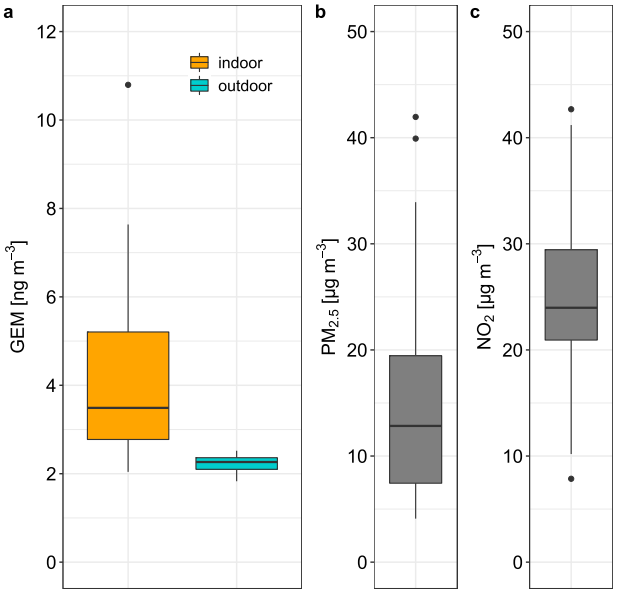

Figure 2. Summary of average (a) indoor (27 households) and outdoor (14 locations) gaseous elemental mercury (GEM) concentrations and average indoor concentrations of (b) particulate matter ($PM_{2.5}$) and (c) nitrogen dioxide (NO_2). $PM_{2.5}$ and NO_2 were measured in 26 households.

The highest household GEM concentration (10.8 ng m^{-3}) was 5-fold elevated compared to the average outdoor concentration (2.21 ng m^{-3}) (see Table S2 in section S6). The absolute difference between the average indoor and average outdoor GEM levels (2.0 ng m^{-3}) was smaller compared to those of studies in Chongqing (3.5 ng m^{-3} in winter), Toronto (13.5 ng m^{-3}), and New York (18.6 ng m^{-3}, difference between median indoor and outdoor GEM).[15−17] Variations in indoor and outdoor total gaseous mercury (TGM) concentrations at nine residential locations in downtown Chongqing, China, were measured to identify possible sources and evaluate diurnal and seasonal fluctuations.[17] Indoor TGM concentrations (eight buildings in summer and two in winter) were significantly elevated over outdoor concentrations. Overall, TGM concentrations were highly variable, likely as a result of changes in anthropogenic emissions (e.g., coal combustion, mobile sources, and iron refinery), fluctuations in atmospheric variables, and unique events (e.g., use of mercury-containing skin cream, presence of Hg-based dental amalgams, thermometer spill, etc.). Elevated indoor GEM concentrations are likely the result of current or past indoor use of Hg-containing appliances like thermometers, barometers, florescence tubes, or paints. A survey conducted among ICARUS residents to identify potential indoor Hg sources revealed that the average GEM concentration was not statistically different between households where Hg thermometers had been broken (2 yes, 17 no), where walls were painted within the last 3 years before measurements (11 yes, 11 no), or with residents having dental amalgam fillings (8 yes, 16 no) [Wilcoxon two-sample t test; $p > 0.01$ (see Figure S1 in section S7)]. Despite the fact that we could not identify specific household Hg sources, elevated indoor GEM concentrations still indicated the presence of diffuse sources (e.g., past spills or appliances containing Hg) of which residents might not be aware.[35]

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Outdoor GEM levels were ~50% higher than northern hemispheric background concentrations (1.5 ng m^{-3}) (see Table S3 in section S8). Outdoor GEM concentrations can be elevated during heating season in the winter when the extent of vertical mixing of air masses is reduced during high-pressure periods.[36] The average GEM concentration measured in this study (2.21 ± 0.20 ng m^{-3}) was lower than the GEM concentration measured 39 m above ground in the center of Basel in February 2012 (4.1 ng m^{-3}).[37] GEM concentrations were slightly elevated compared to GEM concentrations determined in downtown Zurich (median of 1.81 ng m^{-3}) from December 2013 to December 2015 as well as at the Zurich zoo on the outskirts of the city (median of 1.62 ng m^{-3}) over a period from January to February 2016.[36] Concurrent MerPAS deployments were also made across Toronto, Canada, and the average summertime GEM concentration (1.77 ± 0.28 ng m^{-3}) was lower than in the current study.[9] The spatial Hg concentration variability in Basel ranged from 1.83 ng m^{-3} in the Hörnli cemetery area (RFH) close to the crematory to 2.52 ng m^{-3} at a residential area (B07). GEM concentrations were not elevated close to potential emission sources such as the crematory, the waste incineration plant, dental offices, the Basel University Hospital, or the industrial area Schweizerhalle (see Table S3 in section S8).

The total Hg emission to the atmosphere in Basel was estimated by the Swiss Pollutant Release and Transfer Register (PRTR). In 2017, the city of Basel reportedly emitted 14 kg of Hg to the atmosphere. This results in a per capita Hg emission of 0.08 g $year^{-1}$. Basel per capita Hg emissions were comparable to per capita emissions in Zurich (0.06−0.10 g $year^{-1}$) quantified by active measurements using a Tekran 2537X instrument from December 2013 to December 2015.[36] Mercury emissions in Basel, Zurich, or Toronto mainly originated from both diffuse and point sources.[9,36] Ambient GEM concentrations in cities are likely more variable, at least over short periods of time compared to background sites due to re-emission from buildings[19] and other artificial surfaces,[15] exhaust from heating,[38] and sinks (e.g., vegetation cover[39]). The complex vertical and horizontal transport mechanism caused by urban structures like surface type or building height and micrometeorological conditions further contribute to the variability of GEM concentrations in urban environments.[16,40,41] MerPAS in this study were installed within the broad vicinity but not directly next to potential Hg point sources. It is thus likely that MerPAS did not catch GEM exclusively from potential site specific Hg sources but rather from mixed sources. GEM concentrations from background sites at the city outskirts (2.34 ng m^{-3} at B01 and 2.10 ng m^{-3} at B02) were similar to the average outdoor GEM concentration of 2.21 ng m^{-3}.

The average GEM exposure of ICARUS residents was calculated on the basis of average outdoor (2.21 ng m^{-3}) and indoor (4.2 ng m^{-3}) as well as average workplace Hg concentrations of 15 ng m^{-3} (estimated for laboratory and office facilities).[42] On average, ICARUS residents (n = 46 in the full ICARUS sample) spent 14.9% of their time outdoors or in transit, 57.6% at home, 16.4% at an indoor workplace, and 11.2% at other indoor locations. The resulting intake of GEM was calculated as 0.01 μg kg^{-1} of body weight per week for adults (respiration rate of 20 m^3 day^{-1}, body weight of 70 kg). Such an exposure is small compared to exposure from dental amalgam of ~2−16 μg per adult and week.[43,44]

We conclude that indoor and outdoor air Hg concentrations in the city of Basel were clearly below the reference value given by Carpi and Chen[15] (300 ng m^{-3}) at least during 1 week in late autumn when the GEM concentrations are expected to be elevated compared to summertime. Consequently, the average inhalation exposure to GEM for Basel ICARUS residents is of no concern. However, the assessment of chronic toxic effects on local residents remains ambiguous. Our study demonstrates that low-cost MerPAS are suitable to complement measurement campaigns of GEM with other air pollutants. Indoor and outdoor GEM monitoring with MerPAS offers great potential to locate unknown Hg sources and to assess the contribution of indoor Hg sources to total urban emissions. Additionally, MerPAS can help to identify indoor Hg vapor exposure at the workplace or at home.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.0c00110.

An overview of recently measured atmospheric Hg concentrations in different cities (Table S1 in section S1), a paragraph about the sample analyte GEM (section S2), details about the sulfur-impregnated activated carbon analysis (section S3), including QA/QC (section S4), uncertainties for indoor MerPAS measurements (section S5), an overview of results from indoor GEM, $PM_{2.5}$, and NO_2 concentration measurements (Table S2 in section S6), the influence of potential Hg indoor sources on GEM concentration (Figure S1 in section S7), results from outdoor GEM measurements (Table S3), and photodocumentation (Figure S2) of the MerPAS outdoor deployment (Section S8) (PDF)

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Author Contributions

S.O. initiated and coordinated the study. L.W. and S.O. deployed outdoor MerPAS. B.F. and D.V. set up indoor

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MerPAS, provided uHoo data, and produced Figure 1. L.W. analyzed MerPAS and determined GEM concentrations. D.M. provided crucial methodological know-how. S.O. and L.W. wrote the manuscript with major contributions from D.M., D.V., and B.F.

Notes

The authors declare no competing financial interest.

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