# Supporting Information

# Concurrently measured concentrations of atmospheric mercury in indoor (household) and outdoor air of Basel, Switzerland

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## S1. Overview of outdoor atmospheric Hg concentrations in urban areas

Table S1 provides an overview of atmospheric Hg concentrations measured in different urban areas in recent years. Hg air concentrations determined in Toronto[1] are methodically best comparable to our study as it is the only other example of simultaneous deployment of multiple MerPAS. Multiple MerPAS deployments in a city decrease spatially biased assessment of urban GEM concentrations.

**Table S1:** Overview of atmospheric Hg concentrations in urban areas globally

| Location | Hg speciation | Hg (mean ± SD) (ng $m^{-3}$) | Measurement period | Reference |
|---|---|---|---|---|
| Toronto; Canada | GEM | 1.46 ± 0.23 | July/Aug 2016 | [1] |
| Taoyuan; Taiwan | GEM | 2.61 ± 6.47 | Oct 2017 – Sept 2018 | [2] |
| Da Nang; Vietnam | GEM | 3.86 ± 1.46 | March – April 2010 | [3] |
| Seoul; Korea | TGM | 3.72 ± 2.96 | 2006 - 2009 | [4] |
| Bronx; NY, USA | GEM | 1.92 ± 0.59 | Jan 2013 – Nov 2014 | [5] |
| Rochester; NY, USA | GEM | 1.66 ± 0.86 | Jan 2013 – Nov 2014 | [5] |
| Beltsville; MD, USA | GEM | 1.41 ± 0.23 | 2007 - 2015 | [6] |
| Shanghai, Qingpu; China | GEM | 2.77 ± 1.36 | June 2015 – May 2016 | [7] |
| Guiyang; China | GEM | 9.72 ± 10.2 | Aug – Dec 2009 | [8] |
| Chicago; IL, USA | GEM | 2.50 ± 1.50 | July – Nov 2007 | [9] |
| Toronto; Canada | GEM | 1.46 ± 0.23 | July/Aug 2016 | [1] |
| Taoyuan; Taiwan | GEM | 2.61 ± 6.47 | Oct 2017 – Sept 2018 | [2] |
| Da Nang; Vietnam | GEM | 3.86 ± 1.46 | March – April 2010 | [3] |
| Seoul; Korea | TGM | 3.72 ± 2.96 | 2006 - 2009 | [4] |

## S2. Gaseous Elemental Mercury (GEM): The MerPAS sampled analyte

Previously, the sampled analyte of the MerPAS was described as gaseous Hg despite the strong hypothesis that reactive gaseous oxidized Hg (GOM) would not pass through the diffusive barrier. This was based on reviewers' suggestions in McLagan et al.[10] given that this had not yet been proven. However, two recent, as yet unpublished studies[11,12], have categorically defined the sampled analyte of the MerPAS instrument to be GEM. Both these studies were presented at the 14th International Conference on Mercury as a Global Pollutant (ICMGP) held in Krakow, Poland in 2019. Both studies are currently in the process of being completed for submission. Their results are outlined here. The first study showed near zero GOM when just the diffusive barrier (no sorbent inside the diffusive barrier) was placed on the inlet of a Tekran 2537/1130/1135 speciation system under elevated GOM conditions during atmospheric Hg depletion events (AMDEs) at Alert Station in the Canadian High Arctic[11]. A second, Tekran 2537/1130/1135 system was simultaneously measuring Hg speciation at the same location and this system did measure high GOM[11]. Thus, the system with the diffusive barrier on the inlet was removing GOM (and PBM), which suggests MerPAS samples only GEM. The second study analyzed Hg stable isotopes using different MerPAS setups with and without the diffusive barrier again during AMDEs at Alert[12]. The setup without the Radiello® diffusive barrier show distinctive Hg isotope signatures of gaseous oxidized Hg that were not present when the Radiello® diffusive barrier was used [12]. Despite the fact that these two studies are not published, we utilize the terminology "GEM" in this study as it represents the more accurate description of the target analyte.

**S3. Analysis of sulfur-impregnated activated carbon (AC)**

The average amount of sulfur-impregnated activated carbon (AC) contained in each stainless steel mesh cylinder inside the MerPAS white Radiello® sampler was 620 mg ± 83 mg. Since the DMA-80 combustion system allows only smaller samples sizes the AC was analyzed in three aliquots of approximately 200 mg. In order to mitigate sulfur contamination of the DMA-80 catalyst 100 mg of $Na_2CO_3$ was added to each aliquot following a recommendation[13]. The DMA-80 analysis method for solids included ramp heating to 750 °C for 1 minute and constant combustion at 750 °C for 4 minutes. During the analysis we used both liquid primary reference standards (PRS) and solid secondary reference materials (SRM). Liquid PRS consisted of a solution of 50 mg of 100 ng $g^{-1}$ NIST-3133 stabilized in 1% BrCl with which the DMA-80 had been calibrated. Before starting the measurement of samples we run a quality-control pre-sequence consisting of three PRS in order to check the daily performance of the instrument. All measurement data were corrected accordingly if the measured PRS were within 90% to 110% of the expected value. If the PRS were outside of this range the instrument was re-calibrated.

**S4. Quality assurance and quality control of AC analysis**

In order to produce activated carbon (AC) SRM we made use of a mercury vapor primary calibration unit (Tekran® Model 2505, Inc. Toronto, Canada). The calibration unit delivers mercury vapor from a temperature controlled mercury reservoir. We extracted 500 µl of Hg vapor stabilized at 20°C from the calibration unit using a gas tight syringe (Hamilton Company) which corresponds to 6.6 ng of GEM. We injected the Hg vapor into 12 separate 10 ml glass vials (VWR) sealed with aluminum crimps (Thermo Scientific) and PTFE septa (VWR). Each vial contained 200 mg of the same AC that had been filled into the Radiello® cylinder of MerPAS prior to their deployment. After injection of Hg vapor using the syringe we shook the vials with the AC for two minutes and subsequently left them standing for 24 hours. The AC of each vial was used as SRMs during the analytical procedure to test recovery every 5–8 MerPAS AC samples. Recovery of SRMs was 98 ± 11%.

Blanks were measured among samples: Seven lab AC blanks from the same stock used to fill the MerPAS and one procedural (field) AC blank inside a closed MerPAS stored in the same box together with deployed MerPAS before and after deployment until analysis including transport during travel to outdoor sampling locations. The mean Hg concentration of lab blanks was 1.15 ± 0.4 ng $g^{-1}$ and 0.93 ng $g^{-1}$ for procedural blanks. All samples were blank corrected by multiplying the average Hg concentration of lab blanks and procedural blank (1.04 ng $g^{-1}$) with the AC mass in each MerPAS and subtracting the result from the measured total Hg content of each MerPAS.

**S5. Uncertainty estimate for indoor MerPAS measurements**

The uncertainty of indoor deployments of MerPAS was higher than outdoors because the deployed samplers were mainly subject to wind speeds <1 m $s^{-1}$ [10]. At wind speeds below 1 m $s^{-1}$ wind has a greater impact on the MerPAS sampling rate[14]. McLagan et al. [10] reported that the replicate precision indoors was twice as high compared to the outdoor precision for the setup used in this study. Thus, if

we double the precision and overall uncertainty of outdoor deployments from [15], precision and uncertainty for indoor deployments constitute 8% and 18%, respectively. However, this is the first application of this indoor sampling method using MerPAS. A more thorough calibration of the indoor sampling method at more than one site with co-located actively measured concentrations would help to further constrain uncertainty. We do acknowledge that there may be additional, as yet undescribed, error associated with this method. A previous study showed the thicker, less porous, yellow Radiello® to produce higher precision (less overall uncertainty) than the white Radiello®[14]. However, the sampling rate is ~40 – 50 % lower for the yellow Radiello® than for the white Radiello®[5], which requires increased length of deployment times to reach method detection/quantification limits. We used the white Radiello® for GEM measurements in this study for this reason and also the limited budget of the project (yellow Radiello® is approximately double the cost of the white Radiello®).

**S6. Indoor measurements of air pollutants and GEM**

The uHoo comprises nine low-cost sensors, measuring volatile organic compounds (VOC: 10-10'000 ppb), fine particulate matter ($PM_{2.5}$: 0-200 μg $m^{-3}$), carbon monoxide (CO: 0-1000 ppm), carbon dioxide ($CO_2$: 400-10000 ppm), ozone ($O_3$: 10-10000 ppb), nitrogen dioxide ($NO_2$: 0-1000ppb), temperature (-40 to 85 °C), humidity (0 to 100%) and air pressure (300-1100 mbar). The temporal resolution of the measurements was 1 minute. While the sensors used in the device do not provide the same accuracy as reference measurements, they still provide good relative information about the observed air quality within a household[16]. In this study, we only present data from $PM_{2.5}$ and $NO_2$ concentration measurements.

**Table S2:** Statistical summary of indoor average GEM concentrations, average and standard deviation (SD) of temperature and average and 95$^{th}$ percentile (95$^{th}$) of particulate matter ($PM_{2.5}$) and nitrogen dioxide ($NO_2$) concentrations.

| ID | GEM | Temperature | | $PM_{2.5}$ | | $NO_2$ | |
|---|---|---|---|---|---|---|---|
| | | Mean | SD | Mean | 95$^{th}$ | Mean | 95$^{th}$ |
| | (ng $m^{-3}$) | (°C) | | (μg $m^{-3}$) | | (μg $m^{-3}$) | |
| BSL_W_H001 | 2.9 | - | - | - | - | - | - |
| BSL_W_H002 | 2.6 | 21.4 | 0.7 | 8.6 | 18.1 | 22.0 | 31.1 |
| BSL_W_H004 | 6.6 | 21.1 | 0.8 | 16.7 | 59.9 | 16.7 | 42.2 |
| BSL_W_H006 | 4.7 | 22.9 | 0.8 | 12.0 | 47.9 | 19.3 | 30.4 |
| BSL_W_H007 | 5.1 | 21.3 | 0.3 | 42.0 | 87.8 | 10.2 | 39.0 |
| BSL_W_H008 | 4.6 | 22.1 | 0.3 | 10.1 | 25.4 | 15.3 | 64.2 |
| BSL_W_H011 | 2.6 | 19.8 | 0.3 | 27.2 | 48.5 | 23.7 | 31.4 |
| BSL_W_H013 | 6.4 | 21.0 | 0.3 | 5.0 | 8.8 | 24.3 | 34.5 |
| BSL_W_H015 | 3.5 | 18.7 | 0.5 | 20.8 | 64.3 | 42.7 | 63.5 |
| BSL_W_H016 | 2.8 | 20.4 | 0.4 | 13.1 | 37.4 | 21.8 | 48.1 |
| BSL_W_H017 | 6.2 | 21.8 | 0.2 | 15.4 | 28.5 | 25.0 | 34.3 |
| BSL_W_H018 | 4.7 | 20.0 | 0.6 | 14.9 | 41.0 | 28.0 | 74.6 |
| BSL_W_H019 | 2.2 | 21.3 | 0.9 | 20.3 | 104.3 | 37.4 | 62.8 |
| BSL_W_H021 | 2.6 | 19.5 | 1.2 | 12.0 | 40.6 | 23.3 | 42.2 |
| BSL_W_H023 | 5.4 | 24.0 | 0.5 | 33.9 | 63.4 | 21.0 | 47.1 |
| BSL_W_H025 | 3.8 | 19.8 | 1.4 | 6.5 | 18.2 | 28.7 | 60.4 |
| BSL_W_H026 | 3.0 | 21.5 | 0.6 | 12.6 | 36.1 | 7.9 | 22.4 |
| BSL_W_H027 | 2.0 | 22.6 | 0.5 | 7.1 | 16.5 | 29.7 | 47.7 |
| BSL_W_H028 | 10.8 | 21.2 | 0.8 | 6.4 | 14.2 | 41.2 | 131.8 |
| BSL_W_H029 | 2.5 | 18.5 | 0.5 | 7.2 | 16.9 | 37.3 | 62.4 |
| BSL_W_H030 | 3.2 | 22.2 | 0.6 | 39.9 | 152.8 | 25.6 | 47.4 |
| BSL_W_H032 | 2.8 | 22.5 | 0.5 | 16.8 | 38.7 | 33.6 | 62.6 |
| BSL_W_H033 | 5.7 | 22.8 | 1.2 | 4.1 | 6.3 | 26.7 | 44.9 |
| BSL_W_H035 | 3.1 | 22.1 | 0.7 | 16.8 | 84.2 | 20.9 | 39.0 |
| BSL_W_H037 | 7.6 | 22.3 | 0.3 | 8.0 | 23.9 | 39.5 | 76.4 |
| BSL_W_H042 | 3.7 | 23.2 | 0.3 | 4.8 | 9.6 | 22.6 | 40.1 |
| BSL_W_H043 | 2.7 | 20.7 | 0.5 | 20.8 | 45.9 | 16.6 | 38.4 |

## S7. Influence of potential indoor Hg sources on GEM

A survey among ICARUS residents allowed us to pinpoint potential sources of GEM in each of the 27 investigated households. We asked the residents whether they recall 1) Hg spilling due to broken Hg thermometers or light bulbs (2 yes, 17 no), 2) whether residents have dental amalgam fillings (8 yes, 16 no) or 3) whether walls were painted within the last 3 years before measurements (11 yes, 11 no). There was no significant difference in GEM concentrations between "yes" and "no" answers (Fig. S1)

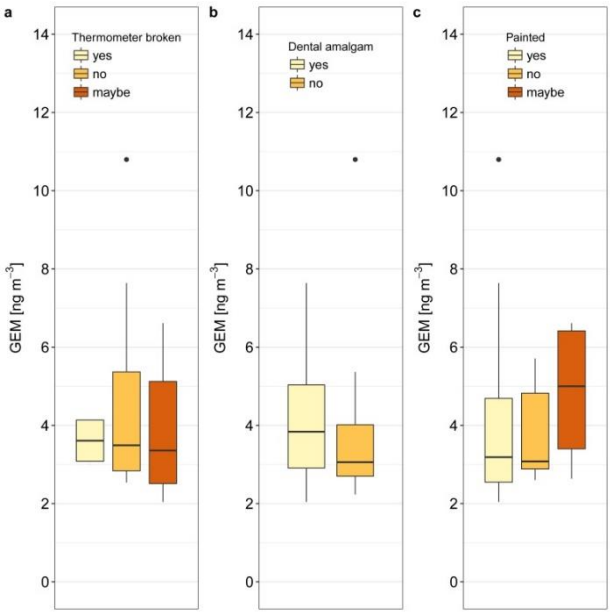


**Figure S1:** Results from survey among ICARUS residents to assess potential indoor Hg sources in the 27 Basel households. GEM concentrations are grouped for households where thermometer broke (a), residents had dental amalgam fillings (b) and walls have been painted within the past three years (c).

## S8. Outdoor GEM and photo-documentation of MerPAS deployment

Contaminated category

MerPAS close to potential point sources were deployed at

- RFA: a cemetery called Hörnli within 150 m of a crematory that represents a potential Hg source from incineration of dental amalgam [17]
- KVA: a waste incineration plant
- LHA4: in the center of the industrial area Schweizerhalle where multiple chemical companies are located
- LHA1 and B05: locations within 170 m and 120 m of dental offices, respectively
- LHA3: at the edge of a major highway
- B03 and B04: in the vicinity of the main building of the Basel University Hospital
- LHA2 and B08: close to busy city roads.

Background category

Sites of MerPAS in the background category constitute

- B07: a residential area
- B06: the middle of the Johanniter bridge crossing the Rhine river
- B01 and B02: two stations at the city of Basel outskirts

**Table S3:** Statistical summary of outdoor GEM concentrations.

| ID | Station name | Latitude (°N) | Longitude (°E) | SR adj. ($m^3$ $days^{-1}$) | GEM (ng $m^{-3}$) | Depl. (days) | Height (a.s.l.) | Height (a.g.l.) | Details |
|---|---|---|---|---|---|---|---|---|---|
| B01 | Basel-Binningen | 47.5411 | 7.5833 | 0.104 | 2.34 | 30.4 | 316 | 2 | city outskirts |
| B02 | Lange Erlen | 47.5922 | 7.6493 | 0.102 | 2.10 | 29.8 | 273 | 2 | city outskirts |
| B03 | Klingelbergstrasse roof | 47.5617 | 7.5805 | 0.104 | 2.44 | 30.1 | 286 | 21 | close to hospital |
| B04 | Klingelbergstrasse street | 47.5615 | 7.5807 | 0.100 | 2.28 | 30.1 | 264 | 2 | close to hospital |
| B05 | Aeschenplatz | 47.5512 | 7.5956 | 0.105 | 2.25 | 30.0 | 270 | 36 | close to dental office |
| B06 | Johanniter Bridge | 47.5648 | 7.5857 | 0.103 | 2.37 | 30.0 | 256 | 3 | bridge over Rhine river |
| B07 | Gellert street | 47.5519 | 7.6084 | 0.104 | 2.52 | 31.1 | 270 | 2 | residential area |
| B08 | Dreispitz street | 47.5369 | 7.6068 | 0.103 | 2.37 | 31.1 | 284 | 3 | busy city road |
| LHA1 | Basel St. Johann | 47.5659 | 7.5820 | 0.105 | 2.10 | 30.1 | 260 | 3 | close to dental office |
| LHA2 | Feldberg street | 47.5670 | 7.5948 | 0.106 | 2.13 | 30.1 | 256 | 3 | busy city road |
| LHA3 | A2 Hard | 47.5382 | 7.6482 | 0.105 | 2.03 | 30.1 | 275 | 3 | major highway |
| LHA4 | Schweizerhalle | 47.5310 | 7.6614 | 0.109 | 2.30 | 30.1 | 270 | 25 | industrial area |
| RFH | Hörnli cemetery | 47.5664 | 7.6405 | 0.104 | 1.83 | 28.9 | 274 | 2 | close to crematory |
| KVA | KVA Basel | 47.5730 | 7.5696 | 0.104 | 1.91 | 30.0 | 261 | 1.8 | close to waste incineration |

**a)** B01: Basel-Binningen

**b)** B02: Lange Erlen

**c)** B03: Klingelbergstrasse roof level

**d)** B04: Klingelbergstrasse street level

**e)** B05: Aeschenplatz

**f)** B06: Johanniter Bridge

**g)** B07: Gellert street

**h)** B08: Dreispitz street

**i)** LHA1: Basel St. Johann

**j)** LHA2: Feldberg street

**k)** LHA3: A2 Hard

**l)** LHA4: Schweizerhalle

**m)** RFH: Hörnli cemetery

**n)** KVA: Waste incineration plant

**Figure S2:** Photo-documentation of outdoor MerPAS deployment (a-n).

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