

1 **Gaseous Elemental Mercury Emissions from Informal E-Waste Recycling Facilities in**
2 **Pakistan**

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26 **Abstract**

27 Detrimental effects of mercury (Hg) on ecosystems and human health have been well-documented.
28 Whereas emissions of gaseous elemental mercury (GEM) from e-waste recycling have been
29 reported in developed countries, much less is known about the situation in the Global South. Using
30 a total of 132 passive air samplers, seasonally resolved concentrations of GEM in air were
31 measured continuously at 32 informal e-waste recycling facilities and background location in
32 Pakistan for a period of one year between September 2020 and December 2021. Annual average
33 GEM concentrations at the studied locations ranged from 1.8 to 92 ng m⁻³. Among the studied
34 cities, higher concentrations were measured in Karachi (mean ± s.d: 17 ± 22 , range: 4.2 - 92 ng
35 m⁻³), Lahore (16 ± 4.2, 8.2 - 22 ng m⁻³) and Peshawar (15 ± 17, 4.9 - 80 ng m⁻³), while lower levels
36 were measured in Hyderabad (6.9 ± 6.2, 3.1 - 25 ng m⁻³), consistent with a higher rate of informal
37 recycling activities in metropolitan areas. Seasonally, higher GEM levels occurred during autumn
38 (15 ± 16: 3.3 – 92 ng m⁻³) and summer (13 ± 8.7: 1.8 – 80 ng m⁻³) than in winter (12 ± 8.4: 2.5 –
39 49 ng m⁻³) and spring (9.2 ± 7.3: 1.8 - 80 ng m⁻³), possibly reflecting enhanced volatilization at
40 higher temperatures and/or varying magnitude of recycling operations in different seasons.
41 Policies and strict regulations related to e-waste management should be developed and
42 implemented urgently in the country.

43 **Keywords:** GEM; E-waste; Informal recycling; Passive sampling; Spatio-temporal variations

44 **1. Introduction**

45 With technological advancements and replacement of old electrical and electronic equipment
46 (Forti et al., 2020), there is an increasing number of recycling operations designed to collect
47 precious metals and enable secondary use of valuable resources. Globally, there is widespread
48 export of e-wastes from some developed countries to developing countries, where informal and
49 unregulated handling of e-waste can result in high worker exposure to mercury (Hg) and other
50 hazardous substances (Wilson et al., 2018; Gravel et al., 2020).

51 Although Hg emissions to the environment have been reduced in some parts of the world, i.e.,
52 Europe and North America, it has risen in others, including Africa and Asia (Streets et al., 2019).
53 Worldwide anthropogenic emissions of Hg to the atmosphere have been estimated to amount to
54 2225 tons per year in 2015 (Bagnato et al., 2015) and have raised Hg concentrations manifold
55 above natural levels. Activities leading to these emissions include mining, extraction of precious
56 metals, coal combustion, chemical production, manufacturing of Hg-added products and informal
57 e-waste processing (Pirrone et al., 2010; Pacyna et al., 2010; Moody et al., 2020; Anselm et al.,
58 2021; Amponsah et al., 2022).

59 In the environment, Hg occurs in its elemental form (Hg^0), but also oxidized in inorganic (Hg^{2+} -
60 mercuric, Hg^+ - mercurous) and organic (methyl/ethyl-Hg) forms. In the atmosphere, Hg exists in
61 three forms, namely, gaseous elemental mercury (GEM, Hg^0), gaseous oxidized mercury (GOM,
62 mostly Hg^{2+} and Hg^+), and particle-bound mercury (PBM). Its chemical form determines the
63 temporal and spatial dispersion and, ultimately, its transfer to other environmental compartments.
64 GEM may have a long residence time in the atmosphere (up to one year) and can be transported
65 over long distances, dispersing to remote ecosystems. In contrast, GOM and PBM have a shorter
66 atmospheric residence time and are readily deposited locally and regionally to terrestrial and
67 aquatic ecosystems (Pirrone et al., 2010; Driscoll et al., 2013).

68 It is critical to study the atmospheric emission of GEM due to its wide dispersion and persistent
69 nature in the atmosphere. Passive samplers have been shown to provide reliable long-term average
70 GEM concentrations in air (Jeon et al., 2020; Naccarato et al., 2021). Because they are relatively
71 inexpensive, easy to handle, and need no power supply, these samplers may be deployed
72 simultaneously and in large numbers, enabling measurements of GEM air concentrations at high
73 spatial resolution at and near potential sources (McLagan et al., 2018a). This in turn allows for the
74 identification of GEM sources to the atmosphere (Tao et al., 2017; Streets et al., 2017) and the
75 assessment of GEM emissions (McLagan et al., 2019).

76 Pakistan is a signatory of the Minamata Convention (signed October 10, 2013) but lack of baseline
77 data and regulatory measures has hampered implementation of Hg reduction efforts (Sattar, 2020;
78 Hina et al., 2021). In recent years, a few monitoring studies have assessed some potential sources
79 of Hg in Pakistan, namely a chlor-alkali plant near Lahore (Jamil et al., 2015), dental clinics
80 (Khwaja and Abbasi, 2014), gold mining sites (Khan et al., 2012; Biber et al., 2014) and a ship-
81 breaking yard in Gadani (Baluchistan Coast) (Kakar et al., 2021). However, the literature on
82 atmospheric monitoring and source identification of GEM in Pakistan is still very limited.

83 Recently, it has been reported that e-waste and its informal recycling could potentially be an
84 important source of GEM emissions to the environment (Nipen et al., 2022; Snow et al., 2021).
85 Because of its rapidly growing economy, population, and consumption, Pakistan suffers from
86 insufficient systems for handling e-waste (Iqbal et al., 2017; Sajid et al., 2019). There are no listed
87 formal recycling sites in Pakistan and the processes are carried out in informal ways without any
88 guidelines and safety measures (Iqbal et al., 2017; Umair et al., 2016). Therefore, this study was
89 conceived to monitor and assess ambient GEM levels and emissions from informal e-waste
90 recycling facilities in Pakistan. This study provides the spatio-temporal determination of GEM

91 levels across major cities in Pakistan, developing a baseline dataset of GEM emissions from
92 Pakistan and constraining the contribution of informal e-waste recycling facilities to the GEM
93 pollution load in the country.

94 **2. Materials and Methods**

95 **2.1. *E-waste Streaming in Pakistan***

96 Details of the e-waste stream in Pakistan are presented in a flow chart in Figure 1. The two major
97 sources of e-waste in Pakistan include domestic generation and legal/illegal import of electrical
98 and electronics equipment (Iqbal et al., 2015). As documented by ‘The Global E-Waste Monitor
99 2020’, Pakistan produced 433 kt of e-waste in 2019 (Forti et al., 2020). In addition to the e-waste
100 generated locally, it has been estimated that Pakistan is currently importing (often illegally) over
101 50 kt of e-waste from developed countries each year (Khan, 2022), with Karachi's maritime port
102 as the major entry point (Imran et al., 2017).

103 Following disposal from these sources, e-waste is collected by scrapers and sellers, who may
104 disassemble the waste into parts, which are then sold to extractors and dismantlers. Dismantlers
105 and recyclers recover precious metals by informal methods such as physical dismantling, open
106 burning, acid baths, and the use of blow torches. To the best of our knowledge, there are no formal
107 e-waste recycling facilities in the country; all e-waste is treated informally (Hameed et al., 2020).

108 **2.2. *Sampling Method***

109 Details of the Passive Air Samplers (PAS) for the monitoring of GEM levels used in the current
110 study have been described previously (McLagan et al., 2016). Briefly, a stainless-steel mesh
111 cylinder is filled with a sorbent (sulfur-impregnated activated carbon with a high capacity to sorb
112 Hg) and placed into a porous polyethylene diffusive barrier (white Radiello[®]), which controls the
113 rate of uptake by standardizing the distance GEM is diffusing from the atmosphere to the sorbent.

114 The Radiello[®] is housed inside a polypropylene jar protecting it from wind and rain during
115 deployments, but also doubling as a transport and storage container. Further details, including the
116 effect of temperature on the sampling rate, and the PAS's precision and accuracy, have been
117 reported previously (McLagan et al., 2016, 2017a, 2018b, Naccarato et al. 2021).

118 **2.3. Study Area and Sampling Scheme**

119 Initially, informal e-waste recycling sites were identified based on: (1) field surveys; (2) national
120 entry points; (3) potential for generation, collection, and markets; (4) e-waste repairing,
121 dismantling and refurbishment sites; and (5) as reported in previous studies (Umair et al., 2016;
122 Iqbal et al., 2015, 2017; Imran et al., 2017; Sajid et al., 2019; Shaikh, 2021). Between September
123 2020 and December 2021, PAS were deployed for four sequential seasonal deployments (Autumn,
124 Winter, Spring & Summer) at a total of 32 informal e-waste recycling facilities in nine major cities
125 of Pakistan including Rawalpindi, Faisalabad, Lahore, Gujranwala, and Multan in Punjab
126 Province, Karachi and Hyderabad from Sindh Province, Quetta from Baluchistan Province, and
127 Peshawar from Khyber Pakhtunkhwa (KPK) Province (Figure 2). One background site was located
128 on a COMSATS university campus in Islamabad. Details, including site name, site code,
129 geographical coordinates and a detailed description are summarized for each sampling location in
130 Table S1. Details on sampling duration and site-specific information are presented in Table S2.
131 The PAS were deployed at different distances from, but within a radius of ~ 200 m of e-waste
132 recycling sites.

133 Deployment lengths of PAS ranged from 62 to 135 days and averaged 100 days. Due to Covid-19
134 travelling restrictions, sampling periods occasionally exceeded the target of 3 months. Samplers
135 were deployed on the rooftop of double story buildings (~10m height). PASs were harvested at the
136 sampling locations by closing the polypropylene jar with a lid, sealing them tightly with

137 polytetrafluoroethylene tape, placing the sealed samplers in Ziplock bags and a closed container
138 for transfer to COMSATS University Islamabad, where they were stored in a clean room until
139 being shipped to Bursa Technical University in Turkey for analysis.

140 **2.4. GEM Analysis**

141 The Hg collected on the sorbent was quantified with a Direct Mercury Analyzer (DMA-1;
142 Milestone Srl, Italy), which is based on the principles of thermal decomposition, Hg amalgamation
143 and atomic adsorption detection. The sorbent in the stainless-steel mesh cylinder was emptied in a
144 pre-weighed Eppendorf tube (2 mL) and then the amount of the sorbent was weighed. A subsample
145 of sorbent was transferred into the quartz cell of the DMA-1 instrument to measure Hg. In the
146 analysis, the sample which is weighed into a quartz vial is dried in the sample cell of the instrument
147 and then thermally decomposed in an oxygen-rich stream. Mercury and other combustion products
148 are released from the sample and passed through a catalyst tube, where all interfering substances
149 are eliminated. The gold amalgamator selectively traps the Hg with other combustion products
150 being flushed from the system. The amalgamation furnace is heated to rapidly release the Hg and
151 carry it into multiple measuring cells positioned along the optical path of the spectrophotometer,
152 and quantified by atomic absorption at 253.65 nm.

153 **2.5 Calculation of Atmospheric GEM Concentrations**

154 Volumetric GEM concentrations C (ng m^{-3}) were calculated by dividing the blank-corrected mass
155 of sorbed Hg m (ng) by the product of a sampling rate SR ($\text{m}^3 \text{ day}^{-1}$) and the deployment time t
156 (day):

$$157 \quad C = m / (SR t) \quad (\text{Eq. 1})$$

158 Although, relative humidity has no effect on the SR , it does increase slightly with temperature
159 ($0.001 \text{ m}^3 \text{ day}^{-1}$ or 0.7% increase in SR for every 1 K increase), because temperature affects GEM's
160 molecular diffusivity. For wind speeds over 1 m s^{-1} , the SR increases by $0.003 \text{ m}^3/\text{day}^{-1}$ for an
161 increase by 1 m s^{-1} (McLagan et al., 2017b). Wind speed influences the diffusion path length by
162 controlling the thickness of the boundary layer surrounding the Radiello diffusive barrier (Zhang
163 et al., 2013).

164 The generic SR of $0.135 \text{ m}^3 \text{ day}^{-1}$ obtained during a global-scale calibration study of the PAS
165 (McLagan et al., 2018) was therefore adjusted for temperature and wind speed (McLagan et al.,
166 2017a; McLagan et al., 2018) using:

$$167 \quad SR_{adj} = SR + (T - 9.89 \text{ }^\circ\text{C}) \cdot 0.0009 \frac{\text{m}^3}{\text{day}\cdot^\circ\text{C}} + \left(WS - 3.41 \frac{\text{m}}{\text{s}} \right) \cdot 0.003 \frac{\text{s}\cdot\text{m}^2}{\text{day}} \quad (\text{Eq. 2})$$

168 where T and WS are the average temperature ($^\circ\text{C}$) and wind speed (m s^{-1}) during the deployment
169 period of each PAS. The meteorological data, reported for each deployment in Table S2, were
170 taken from the Meteorological Department of Pakistan (PMD). The adjusted SR ranged from 0.132
171 to $0.179 \text{ m}^3 \text{ day}^{-1}$.

172 **2.6. Statistical Analysis**

173 Statistical analyses were performed using SPSS Statistics version 22. Analysis of variance
174 (ANOVA) was used to examine differences in GEM mean concentrations between sites and
175 between different seasons at one site. Linear regression models were used for correlations between
176 GEM levels, background concentrations and meteorological parameters.

177 **2.7. Quality Assurance and Quality Control (QA/QC)**

178 QA/QC measures include auto calibration of DMA-1 instrument, proper clean-up of quartz sample
179 cells, monitoring of blanks (field and laboratory) and sample replicates. The instrument run in

180 autocalibration mode automatically when Hg level in each sample was ≥ 100 ng, otherwise user
181 preferred autocalibration was performed after every 10 samples. Field and laboratory blanks were
182 prepared using sulfur-impregnated activated carbon. Laboratory blanks were prepared by weighing
183 ~ 0.011 g of AC taken out of the bulk AC from the supplier into the quartz sample cells. Field
184 blanks were the PAS that were exposed to air for approx. 1 min. during the deployment of the
185 samplers in the field, then were sealed properly and were brought back to laboratory. Average Hg
186 level (ng Hg g⁻¹ AC) in four field (0.72 \pm 0.16; 0.62-0.95) and five laboratory blanks (1.01 \pm 0.35;
187 0.58-1.4) were similar and their average (0.88 \pm 0.30) is in the lower part of the range of blank levels
188 (0.38 \pm 0.08 to 36 \pm 17) reported for previous studies with this sampler (Hoang et al., 2023). Blank
189 correction of samples was performed by subtracting the average concentration in blanks (in ng Hg
190 g⁻¹ AC) multiplied with the mass of the sorbent in a sampler (in g AC) from the amount measured
191 in the sorbent from that sampler (in ng Hg). Instrument detection limit (IDL) was 50% of the lowest
192 level of calibration curve (0.001 ng). The method detection limit (MDL) (0.041 ng m⁻³) was
193 derived as three times the standard deviation of the concentration in the blank and converted to ng
194 m⁻³ applying average of the adjusted SR (0.154 m³day⁻¹) and an average deployment duration of
195 100 days.

196 Quartz sample cells were cleaned prior to analysis by soaking in 5 % nitric acid overnight, then
197 rinsed with deionized water and heated to 550 °C for several hours to remove any traces of Hg.
198 Before analysis of each sample, Hg residue on empty quartz cells was measured by placing the
199 cells in the DMA-1 instrument and applying the same method as during sample analysis. The
200 process was repeated 3 times and the cells were cleaned in 5 % nitric acid overnight if average Hg
201 residue level of triplicate analysis was higher than 50% of average level detected in blanks (0.0045

202 ng). Two aliquots of each sample were analyzed and duplicate analyses showed differences
203 ranging from 3.5 % to 11 %; the average was used to calculate the Hg amount in the total sample.
204 The DMA-1 instrument has a wide dynamic range and is suitable for a variety of different matrices
205 (DMA-1 Manufacturer's Brochure). We tested the accuracy of the results by i) spiking known
206 amounts of Hg on quartz filters and ii) analyzing NIST CRM 1648a urban particulate matter. For
207 the first approach, we spiked 20 ng (n=4) and 50 ng (n=4) Hg (from a solution with a concentration
208 of 10 ng/ μ L in 5% nitric acid solution) on ceramic fiber disks (which are used to hold the samples
209 in quartz cells of the instrument). Measured Hg amount in the spiked samples were 20.3 ± 0.83 ng
210 (19.5 ng-21.2 ng, RSD% between spiked amount and measured amount ranges between 1.9% and
211 6.2%) and 50.9 ± 2.25 ng (48.9 ng-53.1 ng, RSD% ranging 1.5% and 6.3%). The average Hg level
212 measured in three subsamples of NIST CRM 1648a was 1.33 ± 0.078 (1.27 mg/kg-1.42 mg/kg),
213 with RSD% from the certified value of 1.323 ± 0.064 mg kg⁻¹ ranging between 1.2%-7.4%. Overall,
214 these RSD% values were judged to be in an acceptable range.

215 **3. Results and Discussion**

216 **3.1. Average GEM Concentrations at Background and E-Waste Sites**

217 GEM concentrations levels measured during four deployments periods at all studied sites are given
218 in Table S3. At present, due to the lack of national environmental monitoring, there are no data
219 available to assess typical background GEM concentrations in Pakistan. In this study,
220 concentration measured at the background location in Rawalpindi (3.1 ± 0.81 ng m⁻³) and ranged
221 seasonally from 1.9 to 3.8 ng m⁻³. This is about double and triple the global background
222 concentration in the Northern (1.5 - 1.7 ng m⁻³) and Southern (1.1- 1.3 ng m⁻³) hemisphere,
223 respectively (Venter et al., 2015; Sprovieri et al., 2016). For example, GEM levels of 1.5 ng m⁻³
224 have been measured at a remote mountain peak station in Kodaikanal, India (Karthik *et al.*, 2017;

225 Karuppasamy et al., 2020). The concentration level in Rawalpindi ($3.1 \pm 0.81 \text{ ng m}^{-3}$; Table 1) is
226 similar to what has been reported for background sites in Southern China (2.8 ng m^{-3}) (Fu et al.,
227 2010).

228 There could be various reasons for such elevated background concentrations of GEM in Pakistan,
229 including coal production (Ali et al., 2017), coal combustion (Joy and Qureshi, 2023), chlor-alkali
230 plants (Jamil et al., 2015), and gold mining activities (Riaz et al., 2018). Other possible sources
231 could be traffic-related emissions in urban areas (Yue et al., 2021; Cabassi et al., 2022) and
232 atmospheric transport from other regions particularly from nearby countries with high mercury
233 emissions such as India (Lin et al., 2019).

234 The GEM concentration averaged over all 32 e-waste recycling sites and seasons was 12 ng m^{-3}
235 and thus four times higher than the levels recorded at the background site. It is also approximately
236 double what has been reported for coastal/urban air in Chennai, India (4.7 ng m^{-3}) (Karuppasamy
237 et al., 2020), urban air in Mexico City (Morton-Bermea et al., 2021; Schiavo et al., 2022), and the
238 Pearl River Delta in China (Chen et al., 2013) (Table 1). The levels are also comparable or slightly
239 higher than what has been measured in Guiyang, China (Feng et al., 2003). In general, GEM
240 contamination in those settings has been attributed to high industrialization, with coal fired power
241 and cement production plants as key contributors to Hg emission. These greatly elevated GEM
242 levels observed in Pakistan urban areas suggest that e-waste recycling is potentially a major
243 contributor to Hg emissions, even if other possible sources exist. They are supporting earlier
244 studies from Pakistan that identified informal e-waste recycling facilities as a possible source of
245 GEM emission (Iqbal et al., 2015, 2017; Umair et al., 2016; Imran et al., 2017; Sajid et al., 2019).

246 **3.2. Spatial Trends**

247 An inter-city comparison of individual recycling sites is presented in Table S3 and in Figure 3.
248 While serious contamination with GEM is apparent at all sampling sites, the annual average at
249 those sites ranged from 4.4 to 56 ng m⁻³, i.e., from slightly above background to well over one
250 order of magnitude above background. This variation likely reflects the range of sources and
251 source strengths in different city environments, as well as the location of the sampling sites relative
252 to the GEM generating activity (upwind vs. downwind, distance) (Ding et al., 2007). In particular,
253 the concentration levels are related to the nature and quantity of the recycling processes responsible
254 for emission. Higher concentrations (in units of ng m⁻³) were measured in samples from Karachi
255 (mean ± s.d, 17 ± 22, range 4.2–92), Lahore (16 ± 4.2, 8.2–22) and Peshawar (15 ± 17, 4.9–80),
256 consistent with the high rate of informal recycling in these metropolitan areas.

257 With ~17 million residents, Karachi is the city with the highest population in Pakistan, the
258 country's major business center, and South Asia's largest and busiest seaport, annually receiving
259 85 kt of imported e-waste (89 % of the nation's total) (Imran et al., 2017). In particular, Shershah
260 (J14) is the hub of the recycling industry in Karachi (Hasan, 2002) along with Gulshan-e-Hadeed
261 (J17) (Rafeeq, 2020). High GEM concentrations of 56 ng m⁻³ (39–92 ng m⁻³) were measured at
262 site J14, considerably higher than at the other sampling sites in Karachi, (4.2–9.1 ng m⁻³).
263 Similarly, higher levels at Lahore and Peshawar are consistent with extensive e-waste recycling
264 activities and the illegal import of e-waste items along with other secondhand equipment from
265 across the Afghan border (Miankhel et al., 2016; Imran et al., 2017).

266 Among the studied cities, GEM concentrations in Gujranwala, Rawalpindi, Faisalabad, Multan,
267 Quetta and Hyderabad were lower, but still well above background. This may be due to less and
268 smaller-scale recycling activities in those cities when compared to Karachi, Lahore and Peshawar
269 (Imran et al., 2017), consistent with what has been reported by Shaikh et al. (2020) and Hameed

270 et al. (2020). According to Ilyas et al. (2017), small and medium-sized steel and metal extraction
271 industries operate in Gujranwala, suggesting that industrial emissions from chrome-plating
272 facilities, metal smelters and informal melting of e-waste for the extraction of precious metals
273 (Faiz et al., 2015) could be active source of emissions in this city.

274 **3.3. Seasonal Trends**

275 Pakistan has four well defined seasons, a warm and rainy summer (June to August), a dry autumn
276 (September to November), and a cold and dry winter (December to February) and spring (March
277 to May). Seasonal variations in the GEM levels are presented in Figure 4. Higher concentrations
278 (in units of ng m^{-3}) occurred during autumn (mean 15: range 3.3–92) followed by summer (13:
279 1.8–80), winter (12: 2.5–50), and spring (9.2: 3.1–39), possibly reflecting the variable magnitude
280 of recycling operations in different seasons. The small variations between seasons suggest minor
281 impact of meteorological conditions (Wan et al., 2009). Generally, higher concentrations were
282 measured during months with lower temperatures and lower ones in summer and post monsoon
283 months. An increase in coal and biomass combustion or reduced vertical atmospheric mixing
284 might contribute to the elevated atmospheric mercury levels during the cold seasons. Also, higher
285 rates of photochemical oxidation may reduce GEM levels in summer. This trend has been observed
286 in several studies, including Schleicher et al. (2015), Kumari and Kulshrestha (2018), Yi et al.
287 (2020), and Yeh et al. (2021).

288 **3.4 Comparison with Measurements at Other E-Waste Recycling Sites**

289 Table 1 compares the annual average GEM concentration measured here with those that have
290 previously been reported for various e-waste recycling sites. The annual average GEM level (12
291 ng m^{-3}) at e-waste recycling sites in this study is lower than what has been reported in Norway
292 (Snow et al., 2021) and Taizhou, China (Tang et al., 2015) although the highest GEM levels we

293 recorded (at site J1 in Peshawar and site J14 in Karachi) are comparable to the average levels
294 recorded in these studies. A possible reason for higher levels could be placement of samplers closer
295 to the e-waste handling activities (5-20 m) compared to what was feasible in our study (~200 m).
296 Snow et al. (2021) reported GEM concentrations around an e-waste recycling facility in Norway
297 of 2.8 to 3.8 ng m⁻³, with much higher levels inside the enclosed facility (30 to >1000 ng m⁻³).
298 Snow et al. (2021) suggested that very high concentrations occurred close to certain types of Hg-
299 containing e-waste materials such as broken compact fluorescent lamp (CFL) bulbs and
300 fluorescent tubes. Nipen et al. (2022) reported GEM concentrations (5.3 ng m⁻³) close to at an e-
301 waste recycling site in Dar-e-Salaam, Tanzania, that are similar to the levels in this study (e.g.,
302 Hyderabad: 6.2 ng m⁻³).

303 **3.4 Implications for Human Inhalation Exposure to GEM**

304 According to the U.S. Agency for Toxic Substances and Disease Registry (USATSDR, 2015), the
305 Minimal Risk Level (MRL) for chronic inhalation exposure to GEM on a daily basis is 200 ng m⁻³
306 ³. The United States Environmental Protection Agency (USEPA) provides a reference value for
307 GEM inhalation of 300 ng m⁻³ (Palma et al., 1999), whereas the World Health Organization (WHO,
308 2000) and the International Programme on Chemical Safety (Fisher & WHO, 2003) suggest values
309 ranging from 100 to 200 ng m⁻³. Japan's Ministry of the Environment suggested occupational GEM
310 inhalation limits of 40 ng m⁻³ (MOE, 2003). The values measured here are consistently lower than
311 these thresholds, except for a handful of seasonal deployments at J14 and J1 that exceeded the very
312 strict Japanese limit. We nevertheless believe that our measurements raise serious concerns related
313 to human inhalation exposure to GEM.
314 First, because our measurements represent average concentrations over periods of approx. three
315 months, we can expect that much higher concentrations prevailed during shorter time periods.

316 More importantly, we believe that the GEM levels in inhaled air could be manifold higher than the
317 reported average annual levels (12 ng m^{-3}) because our sampling sites were generally $\sim 200 \text{ m}$
318 removed from the actual recycling operations. In particular, we should expect very steep spatial
319 concentration gradients between the site of the actual recycling operations and the sampling sites.
320 For example, Snow et al. (2021) observed significantly higher levels of GEM inside a Norwegian
321 e-waste recycling facility, with concentrations ranging from 31 to 1140 ng m^{-3} , compared to levels
322 of 2.8 to 3.8 ng m^{-3} at a distance of only 100 m from the facility. Similarly, Monaci et al. (2022)
323 reported long term average GEM concentrations near processing facilities of an abandoned Hg-
324 mine that ranged over more than two orders of magnitude (from 17 to $4,200 \text{ ng m}^{-3}$) over a distance
325 of a few 100 metres. Moreover, in Snow et al. (2021)'s study in the Norwegian e-waste recycling
326 facility and two artisanal gold mining communities in Ghana, which involved both stationary
327 sampling locations and personal wearable samplers, the latter had consistently higher GEM
328 concentrations, often by as much as an order of magnitude.

329 In summary, by measuring annual average concentrations as high as 56 ng m^{-3} at a fair distance
330 from actual recycling activities, we project personal inhalation exposures that will regularly and
331 consistently exceed MRLs for chronic inhalation exposure to GEM. Such exposures may occur
332 not only for those directly involved in the recycling activities, but also to community members in
333 the vicinity to these activities, including children.

334 **4. Conclusions**

335 A total of thirty-two informal e-waste recycling facilities across nine major cities of Pakistan were
336 selected for the monitoring of GEM concentrations using a passive air sampling technique. The
337 results indicated that uncontrolled and large-scale informal e-waste recycling practices were
338 associated with high GEM concentrations in metropolitan areas. These levels exceeded those

339 found at a national background site and are well above hemispheric background levels, suggesting
340 that emissions from e-waste handling and recycling operations were contributing to these elevated
341 levels. The levels remained high throughout the year, indicating that the emission sources were
342 stable. Practices such as dismantling, open burning, acid bath, and refurbishment of e-waste
343 without safety measures were commonly observed at the studied sites. Furthermore, these facilities
344 lacked proper ventilation and were often located in urbanized areas, posing health hazards related
345 to inhalation exposure not only for the those involved in the e-waste recycling operations but also
346 to the larger community. It is important for Pakistan, as a signatory of the Mina-Mata Convention,
347 to develop an e-waste management plan that will help reduce emissions and exposure levels.
348 Additional investigations are needed to better identify and characterize the e-waste handling
349 activities that result in the release of GEM, to determine the spatial concentration variability within
350 the informal e-waste processing sites, to quantify the human GEM inhalation exposure and to
351 investigate the potential health effect it may cause.

352 **Supplementary Information.** Supplementary information (SI) contains details on description of
353 sampling sites (Table S1), sampling periods, meteorological conditions and site sampling rates
354 (Table S2) and basic descriptive statistics of spatio-temporal concentrations level (Table S3)

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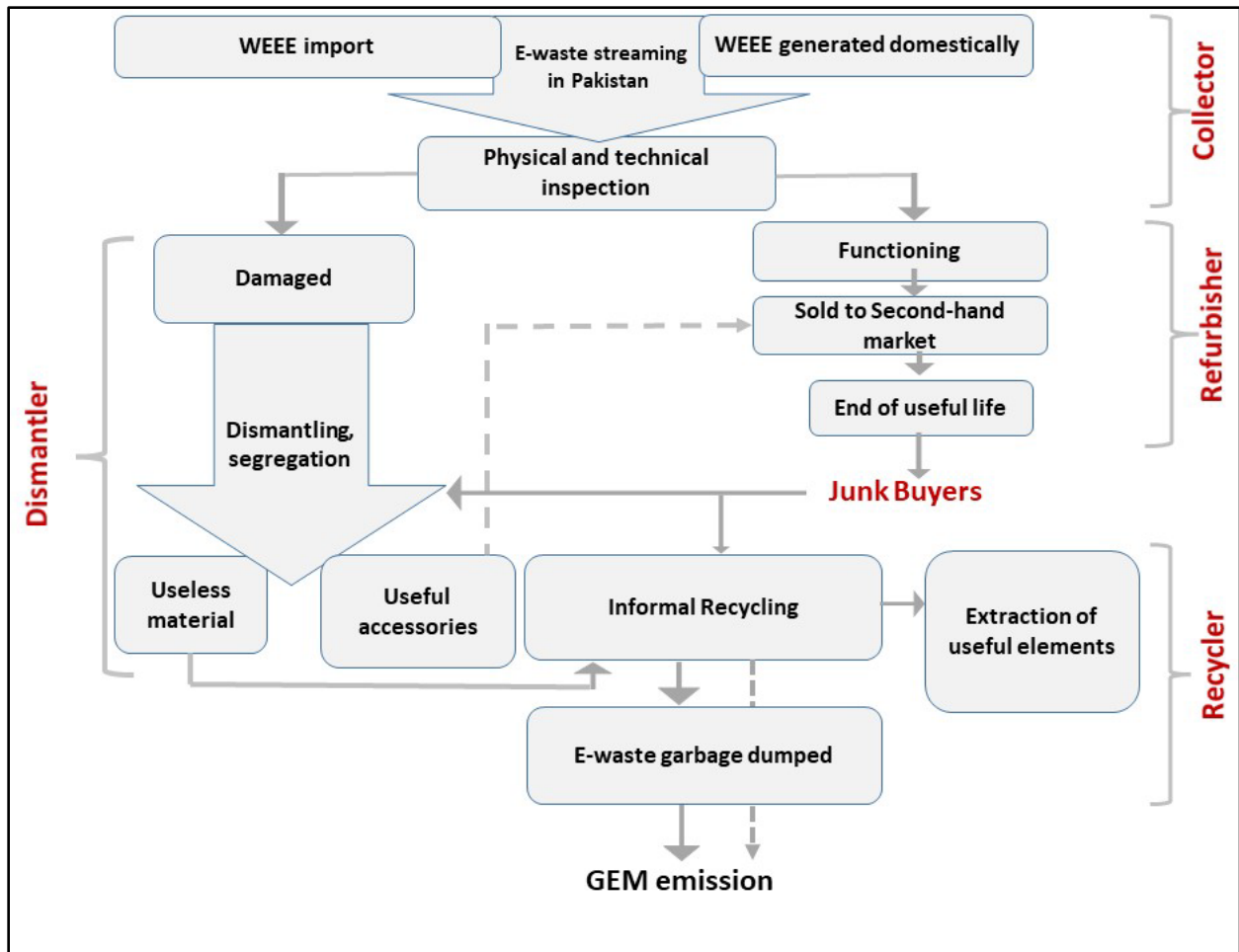


Figure 1: Flowchart showing the e-waste streaming in Pakistan

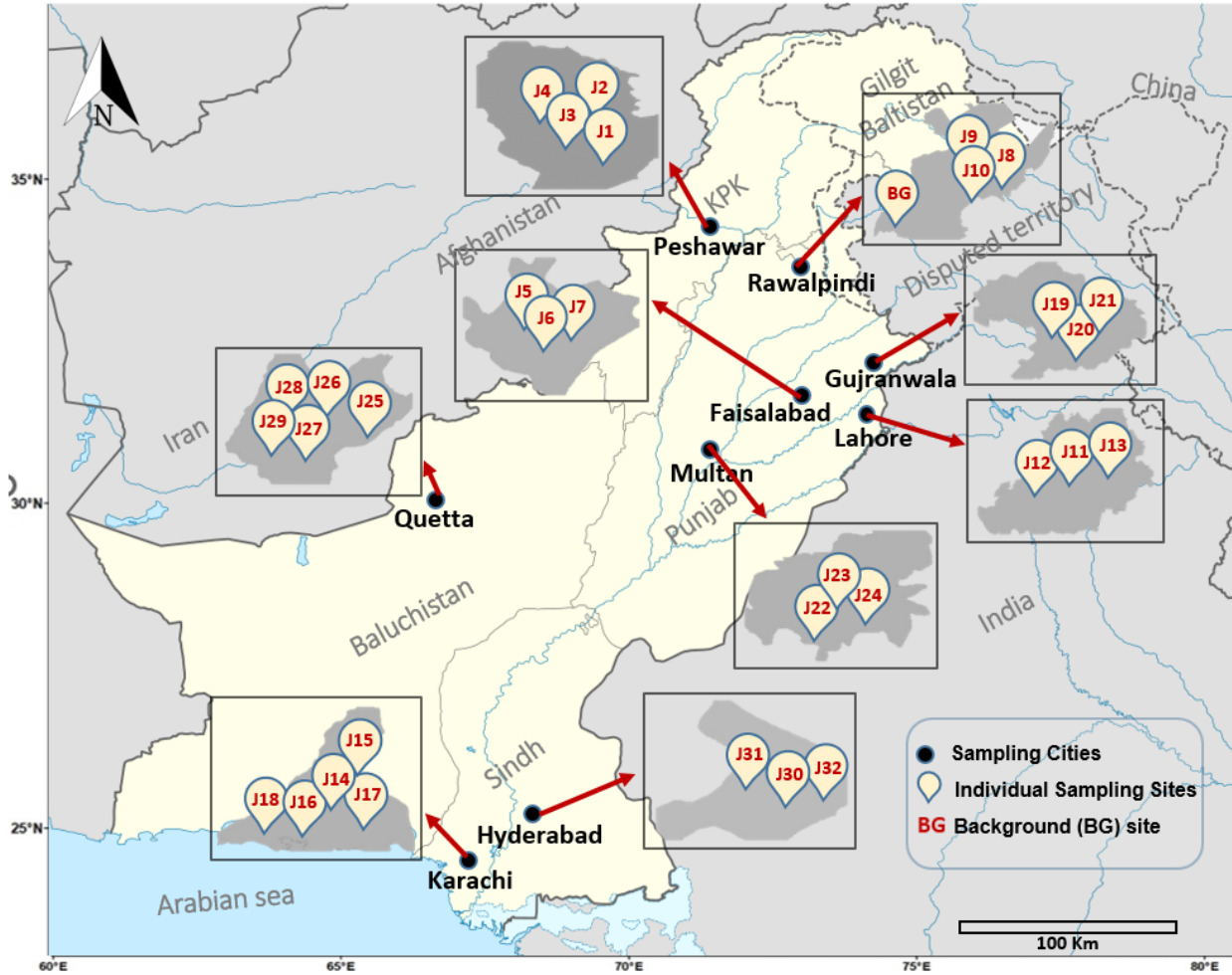


Figure 2: Individual e-waste recycling sites across major cities in Pakistan

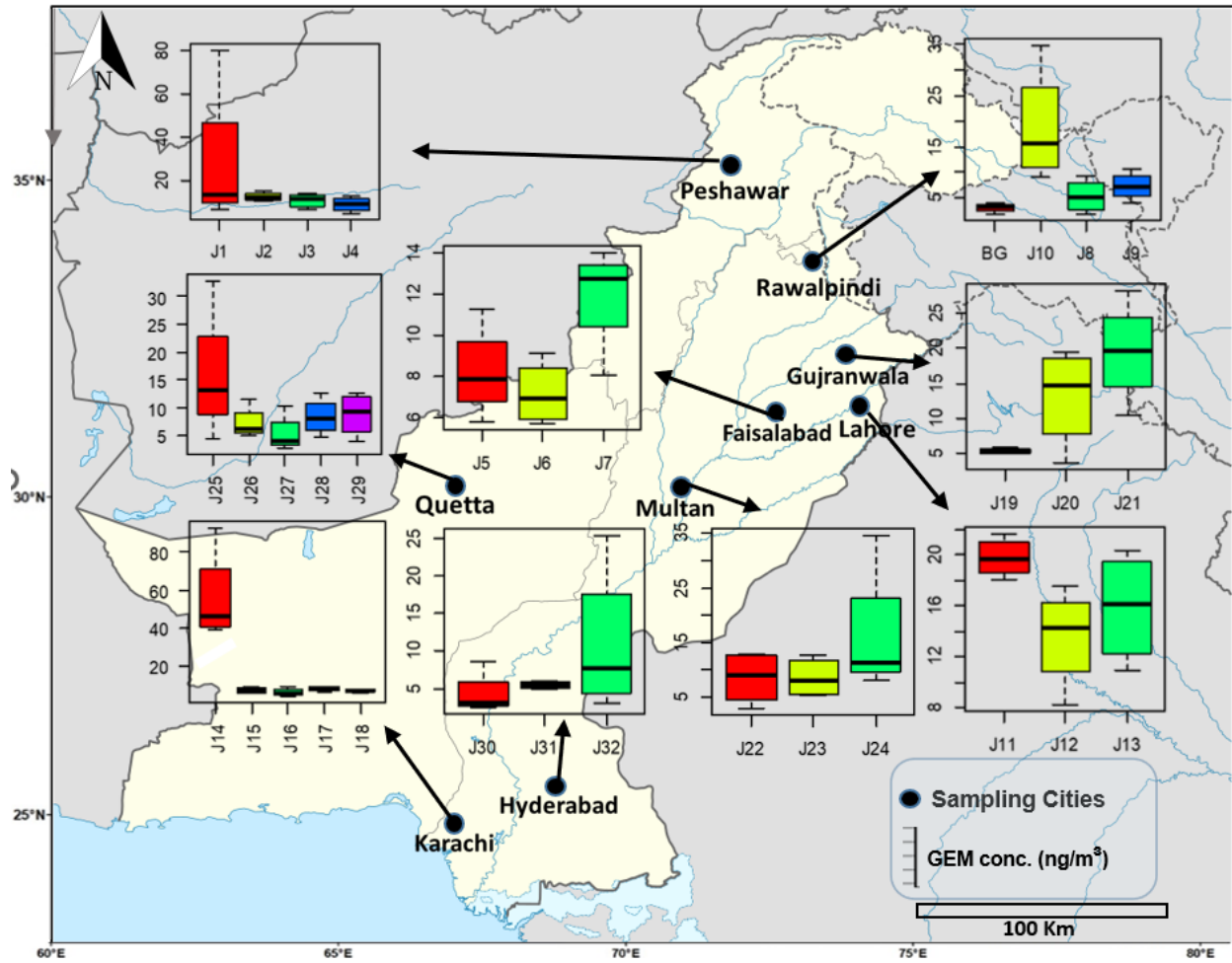


Figure 3: Yearly mean concentrations of GEM (ng m^{-3}) at individual e-waste recycling sites in Pakistan

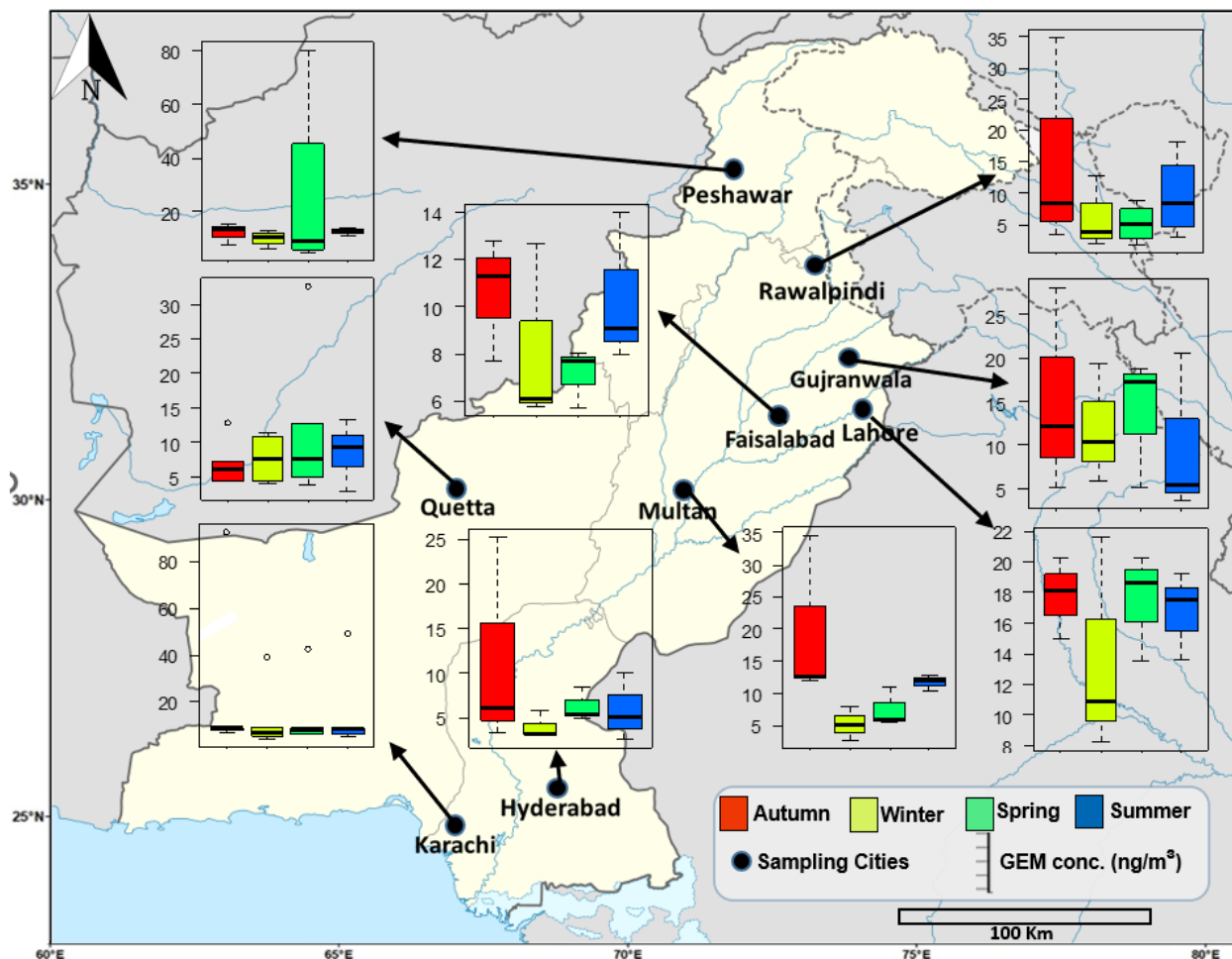


Fig 4: Seasonal GEM concentrations (ng m^{-3}) at informal e-waste recycling sites

1 **Table 1: Comparison of average GEM concentrations (ng m⁻³) with previous studies across the worldwide**

Location	Country	Sample site	Monitoring period	GEM (Mean ± SD) Range values	Reference
Regional Background Sites					
Rawalpindi	Pakistan	Background	Oct-2020 to Sept-2021	2.99±0.71 (1.94-3.84)	This study
Summit of Mt. Leigong	South China	Ambient Air	May-2008 to May 2009	2.80±1.51 1.88 to 3.59	Fu <i>et al.</i> , (2010)
Popocatepetl	Mexico	Volcanic/rural	Mar-2019	1.72 ± 0.83 0.51-5.5	<u>Schiavo <i>et al.</i>, (2020)</u>
Kodaikanal*	India	Rural	Nov-2012 to Sept-2013	1.53±0.25 0.83 to 3.25	<u>Karthik <i>et al.</i>, (2017)</u>
		Ambient air	Jan-2015 to Dec-2016	1.53 1.38-159	Karuppasamy <i>et al.</i> , (2020)
Southern Hemisphere		Background	2016	1.1-1.3 (range)	Sprovieri <i>et al.</i> , (2016)
Northern hemisphere		Background	2015	1.5-1.7 (range)	Venter <i>et al.</i> , (2015)
E-waste Recycling Sites					
Peshawar, Faisalabad, Lahore, Rawalpindi, Karachi, Multan, Gujranwala, Quetta & Hyderabad	Pakistan	informal facilities	Oct-2020 to Sept-2021	11.88±2.12 (1.78-92.07)	This study
Dar-es-Salaam city	Tanzania	informal sites	Feb-2019 to April 2019	2.13±1.57 0.79-5.34	Nipen <i>et al.</i> , 2022
E-waste recycling facility	Norway	formal recycling facility	Dec- 2018	5 Geomean 0.9 – 1140	Snow <i>et al.</i> , 2021
Taizhou*	China	informal recycling sites	2015	30.7 ± 9.9 16.7 to 43.4	Tang <i>et al.</i> , (2015)
Urban and Impacted Sites					
Mexico City	Mexico	Urban	Mar-2021 to Apr-2021	5.60 ± 2.33 0.20-30.23	Schiavo <i>et al.</i> , (2022)
Hefei	China	Urban	Mar-2016 to May-2016	2.53 ± 1.28 0.32-15.10	<u>Yue <i>et al.</i>, (2021)</u>
Mexico City	Mexico	Urban	May-2019,	3.80 ± 1.34 0.50-11.90	<u>Morton-Bermea <i>et al.</i>, (2021)</u>

			May-2020	5.04 ± 2.95 0.50-32.60	
Chennai*	India	Ambient air	Jan-2015 to Dec-2016	4.68 3.62-5.40	Karuppasamy <i>et al.</i> , 2020
Zhongshan*	China	Urban, fluorescent lamp manufacturing facilities	Jul-2019 to Aug-2019	2.4 ± 3.5 0.37-49	<u>Luo <i>et al.</i>, (2021)</u>
Abbadia San Salvatore	Italy	abandoned Hg-mining area	Jun-2016 to Jul-2016	1030±1420 17 to 4200	Monaci <i>et al.</i> , (2022)
Mexico's Pacific coast, Presidente Plutarco Elías Calles (CETEPEC)	Mexico	Coal-fired power plant Mt. Dinghu	2013	2.8 0.3-14	Garcia <i>et al.</i> , (2017)
Pearl River Delta	China	Guangzhou	Nov-2010 to Oct-2011	5.07 ± 2.89 1.87 to 29.9 4.60 ± 1.36 2.7 to 11	<u>Chen <i>et al.</i>, (2013)</u>
Guiyang*	China	Coal Fired Power Plant and Guizhou Cement Production Plant	Apr-2000 to Nov-2001	7.39 1.7 to 147	Feng <i>et al.</i> , (2003)

2 * Total gaseous mercury (TGM)