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ASGM Mercury Discharges in Tropical Basins: Assessment of the Criticality of Their Geographical Distribution

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Abstract: The global-scale impacts of mercury discharged from Artisanal Small-scale Gold Mining (ASGM) on soils have been poorly studied, unlike atmospheric emissions. This is a key point to understand the potential exposure to mercury pollution of ecosystems and populations living in the river basins where ASGM is practiced, since the largest fraction of the pollutant is poured into soil, independently of the amalgamation technique employed. ASGM activities emit into the atmosphere mercury in its elemental form, which reacts very slowly with the major oxidants. Therefore, the exact location of the ASGM sites has a limited impact on the atmospheric mercury fate. Conversely, this cannot be applied to the mercury discharged on top of the soil. Two ASGM inventories and the related distributions available in the literature along with two novel distributions based on the intersection of zones characterized by low population density and rural areas were compared using a newly introduced Vulnerability Index. The results from this comparison showed that a precise distribution of ASGM is crucial to effectively evaluate the fate of mercury, and therefore the resulting effects on the local ecosystems.

Keywords: mercury; vulnerability; hydrological basin



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1. Introduction

The problem related to the presence of Artisanal Small-Scale Gold Mining (ASGM) in a territory is multifaceted. ASGM could reenter “green crimes”, as unauthorized acts committed by an individual or an organization in violation of a law, endangering the environment and people’s health [1]. Indeed ASGM is often practiced in the poorest areas of South America, Asia and Africa, where in the absence of mining regulation, informal workers increase their production since these activities are fundamental for the local and global economy [2]. Despite that, workers do not have licenses or knowledge about the best safe mining practices to minimize accidents [3]. These activities are almost always managed by criminal organizations, and 77% of ASGM miners reported an increase in their overall incomes as a result of illegal mining [4].

The attention to ASGM started a few decades ago and is increasing because it is a constantly expanding sector with various risks associated [5,6]. From an environmental point of view, before starting the extraction activities, the territory undergoes radical deforestation, because wood is needed to reinforce the excavations [7]. This further affects biodiversity, accelerates erosion, and potentially triggers landslides [8]. Other risks are linked to human diseases, since mercury (Hg), in its elemental form (Hg^0), used to amalgamate and extract gold, is a potent neurotoxicant [9]. For its toxic effect, Hg emissions from ASGM are covered by the Minamata Convention (www.mercuryconvention.org, accessed on 9 January 2023) [10]. Hg leakages into the environment greatly depend on the type of mineral processed. If secondary minerals (alluvial, colluvial, and eluvial) are processed, the Concentration Amalgamation (CA) technique is generally used, which employs less Hg than the Whole Ore Amalgamation (WOA) technique, in which the entire mineral is processed without crushing [11].

Consequently, these techniques pose risks to human health either by the direct inhalation of Hg emissions or via Hg discharged into soil or water that can be methylated and enter the food chain of river and lake biota [12]. Indeed MeHg is highly toxic for humans [13,14], and the areas where ASGM is practiced further represent an ethical problem due to the great presence of pregnant women and children [15]. Further, “post mining water bodies” have great methylation potential, exposing the ecosystems to biodiversity losses [16].

All aspects described above make the working activities of the mining community inconsistent with the concept of a sustainable livelihood [15], requiring adequate policies and actions.

Until now, studies on Hg from ASGM, especially those on a global scale, focused mainly on the estimates of atmospheric emissions, and no one has investigated Hg discharges. From the atmospheric point of view, the exact location of ASGM has a limited impact on the resulting Hg deposition, since most of the Hg^0 emitted by this activity likely contributes to the global pool. Contrarily, the reach of the Hg discharged by both CA and WOA amalgamation techniques, which greatly exceeds the amount released into the atmosphere [11,17], strongly depends on the precise location of the leakage.

For these reasons, the geographical distribution of the precise locations of Hg discharges related to ASGM is crucial to understanding its effects on public health. The rationale of this study is to urge the scientific and policy communities to improve their knowledge about this aspect, showing how different and reasonable geographical distributions of ASGM Hg discharge can potentially lead to very different impacts on people and ecosystems. To demonstrate this, ASGM Hg discharges as estimated in Bruno et al. [18] were considered and then distributed using four different geographical distributions, two from the literature and two developed for this study.

In this regard, in this study, an Environmental Index (IVm) was defined to identify the effects of the discharges in the most important basins of the tropical countries for which ASGM Hg emissions have been estimated in Bruno et al. [18]. This index is functional to study the potential impact of Hg on habitats of rivers and lakes influenced by ASGM locations, where Hg is captured by sediment, fixed in aquatic food-chain, therefore endangering humans and animals that eat contaminated fish [19]. Next, this Environmental Index was applied to four different potential geographic distributions of ASGM activities.

Identifying the mining sites is difficult since these are often illegal works [20,21]. Moreover, the ASGM location is sometimes complex as the sites are not fixed. The workers often use the gold resources until depletion and then they move to neighboring zones, leaving obvious traces on ecosystems [22]. If “inactive” mining zones, or zones that are “active” for a long time, are somewhat easy to recognize due to visible alterations of morphology and vegetation and due to the presence of intense erosion [23], on the other hand, identifying some “active” mining zones can be challenging, as well as the oldest inactive ASGM sites, where traces are covered by vegetation. However, general features of ASGM activities have been somewhat common over the different times and continents. Indeed, due to its illegal nature, ASGM is generally practiced in remote and rural areas of underdeveloped tropical countries where gold can represent an important resource.

Exploiting these aspects, in this study, a methodology has also been proposed to identify rural areas where the ASGM could be located by cross-referencing data, in a GIS environment, regarding agriculture, social and geological aspects, like land use, population density, and morphology.

2. Materials and Methods

2.1. Global Inventories of ASGM as Anthropogenic Activity

In this study, the dataset of Bruno et al. [18] has been used as the reference for the estimates of Hg emissions and discharges from ASGM.

Two other global inventories for anthropogenic Hg emissions, the Global Mercury Assessment (GMA, [10]), and the Database for Global Atmospheric Research ([24], <https://www.dgar.ac.cn/>).

[//edgar.jrc.ec.europa.eu](https://edgar.jrc.ec.europa.eu), accessed on 6 February 2023) were used to extrapolate the geographical distribution of Hg emissions, and then used to proxy the Hg discharges.

The Global Mercury Assessment GMA 2018 is the fourth assessment by the United Nations Environment Programme, after the reports of 2002, 2008, and 2013. The assessment is produced by the UN Environment in collaboration with the Arctic Monitoring and Evaluation Program (AMAP) and it is supported by documents prepared by a team of experts [25]. These documents provided the scientific support for the Minamata Mercury Convention, adopted in October 2013 and entered into force in August 2017. The 2018 Global Mercury Assessment technical report represents the update of the estimates made in 2015 [10]. In this report, the estimates of Hg emissions are calculated using available data from regional Hg consumption, as national data are not available in most cases. The applied method allowed the estimation of Hg emissions linked to different production processes, such as ASGM, for 11 regions of the world.

The Emissions Database for Global Atmospheric Research EDGAR (EDGARv4.tox2) is a worldwide database of anthropogenic emissions of greenhouse gases responsible for air pollution on Earth since 1970. EDGAR uses international statistics and provides both emissions in terms of total national share, as well as grid maps at a resolution of 0.1×0.1 degrees globally, with annual, monthly, and hourly data. Regarding the ASGM, the estimates are conceived based on gold production, using data from scientific literature for the period 1970–2012 [24,26].

While the previous two databases considered all industrial sectors, the dataset of Bruno et al. [18] was limited to the ASGM sector. Differences between previous databases concern the observation time intervals and the investigation scale. To estimate the related Hg emissions and discharges for countries of the tropical and subtropical region for the years 2006–2019, Au production and the two extraction methods CA and WOA have been used. A multi-step procedure was then applied to reduce uncertainties providing reliable estimates of Hg emissions and their confidence intervals.

2.2. Hg Discharges Distributions

As introduced above, the first two distributions of ASGM are based on those of AMAP and EDGAR. In particular, for each country for which an estimate of Hg discharges from ASGM exists in the inventory of Bruno et al. [18], these discharges have been mapped following the ASGM Hg emission distributions of AMAP and EDGAR for that country.

The remaining two were carried out as explained in the following paragraphs.

Distribution Based on Demography

This distribution has been accomplished on the basis only of demographic distribution. Demographic information in this work was obtained starting from the data available at sedac.ciesin.columbia.edu/data/set/grump-v1-urban-extents/maps (accessed on 13 March 2023) and transformed into a raster format. The information was vectored, obtaining a global mapping to differentiate the class of the rural population from all the rest. In the various continents, the rural population density index varies from a minimum of 0.5 hab/km² in Scotland, to less than 300 hab/km² in Brazil [27]. Since a site with a population density lower than 150 hab/km² (enrd.ec.europa.eu, accessed on 16 March 2023), can be defined as a rural municipality, for this work this threshold was considered.

Distribution Based on Demography Plus Rural Areas

The other and more complete distribution has been accomplished considering both demographic distribution and rural areas, which intrinsically contain different environmental information. Currently, the process of defining rural areas is very complex [28]. These are generally areas of open countryside, with small and poor settlements, which depend on agriculture and natural resources, and are highly vulnerable to climate change and extreme events [29]. The rural areas are very different from each other and therefore it is very difficult to give a single definition for all. Certainly, there are some fundamental

elements common to all rural areas of the world: physical discomfort, as the complexity to move; environmental vulnerability, as the predisposition to suffer permanent impacts for external pressures; socio-economic disadvantages, intended as a weak economic structure with difficult access to services and urban economies (enrd.ec.europa.eu/, accessed on 16 March 2023). On an international scale, there are various methods to define rural areas. For example, some rely on nocturnal light spotting, population density, and urbanization [30], others on the definition of morphological spatiotemporal patterns identified by satellite images of different periods [31]. In addition, on a local scale, there are other remarkable methodologies: the “Organisation for Economic Co-operation and Development” (OECD) methodology, which reclassifies the territory based on only population density; the 2007–2013 PSN “Piano Strategico Nazionale per lo Sviluppo Rurale” methodology, where an additional variation linked to the total agricultural area is added to the index of population density; the Emilia-Romagna PSR 2007–2013 methodology, where rural clusters are identified based on socio-economic, agricultural, industrial, and service-related index [32] (www.reterurale.it/flex/cm/pages/ServeBLOB.php/L/IT/IDPagina/11182, accessed on 11 March 2024).

In this work, to define the rural areas, information on land use was obtained (<https://land.copernicus.eu>, accessed on 9 February 2023) by downloading it in raster format n. 135 GeoTIFF files for 20×20 degrees of 2015 “epoch”, based on input for one year before and one year after. These images describe 23 discrete land use classes at 100 m, from evergreen forest to ocean, and finally no data [33]. In Qgis 3.22, a single raster with land use coverage on a global scale has been created. Each cell was automatically associated with a band corresponding to a particular land use class. The global raster was vectored to leave only the attributes that characterize the environments that transit from “forest” to “herbaceous vegetation”, excluding territories with perennial ice. The most rugged landforms were discarded leaving only the topographically flatter areas.

Finally, the land use and population levels have been intersected with the vector layer of the countries (www.naturalearthdata.com, accessed on 8 March 2023). For the resulting operation, in each country, the areas where ASGM activities could be located were identified. The single Hg emission value [18] for the whole country was distributed and scaled according to the population density, obtaining an Hg emission and discharge value for each single point of the layer vector in the rural areas.

2.3. Vulnerability Factors

In ASGM activities, Hg is widely used for a mix of reasons: it is easy to use by a single operator and it is cheap. Often miners are not aware of the risks, whereas in other cases it is imposed by the bosses [34].

Hg used in gold mining enters into the soil and stays for a long time through organic matter. Hg that has accumulated in this matrix may be slowly discharged into surface waters or soil over time. Hg is not biodegradable and can damage the ecosystem by its diffusion through wastewater, surface water, and groundwater [35].

Vegetation also plays an important role in regional Hg cycles. Leaf litter is important for the deposition of Hg in forested areas, although differences exist across different forest types [36]. Furthermore, Hg in the soil can be leached by rainwater, cross the vadose zone, and end up polluting aquifers and all agricultural products. Regarding this aspect, populations that feed predominantly rice can experience various health effects in Hg pollution areas, due to the methylmercury (MeHg) enrichment of the rice grains [37,38]. Although domestic and agricultural irrigation in many underdeveloped countries depends on groundwater and is, therefore, less sensitive to pollution, there is often a mismanagement of surface water bodies [39]. The concentrations of Hg on all continents often exceed the limits allowed by the WHO. It has been already found that a great part of water bodies in the world are highly polluted by toxic metals with a remarkable impact on the health of people [40]. Indeed, in recent studies, in Asia and South America Hg concentrations were above WHO guidelines in drinking water [40]. Furthermore, the surface waters of many

African basins are heavily polluted due to the exploitation of mineral resources. Despite this, there is still no correct regulation regarding environmental pollutants, and also where standards exist, they are not considered and/or applied [41]. Here, the focus has been placed on the largest hydrographic basin because of the effects Hg discharges from ASGM can have on biota living in the mining areas [16]. Hg can also be discharged directly into water, accumulating in river sediments, where it can be converted to MeHg and rapidly incorporated into the aquatic biota [42]. Fish is the most easily accessible animal protein in rural areas where there are river networks, guaranteeing the nutritional factors to the populations. Fishes in these areas are directly affected by Hg, causing serious damage to various animals, other aquatic organisms, and humans.

For these reasons it is crucial to understand how the people living in an area with ASGM are exposed to Hg considering their “vulnerability”, i.e., to be intended as the propensity of a site/person/ecosystem to suffer from damage following a specific event (<http://www.unisdr.org/eng/library/lib-terminology-eng%20home.htm>, accessed on 11 March 2024). Vulnerability is not an absolute characteristic, but rather a relative, non-measurable, and dimensionless property that indicates where an event is most likely to occur [43]. So, mapping the vulnerability of surface hydrological resources can prevent environmental deterioration impacting the entire ecosystem [44].

In the past, several works paid attention to the long-term health impacts of chronic Hg exposure on residents in areas highly polluted by Hg [38]. Some of the estimates have been based on the concentration of Hg in water and soil samples collected at single sites [45]. Other works have evaluated a health risk index relating to IQ, breathing, or walking problems based on workers’ nutrition [46]. Still, other works have calculated how the concentrations of Hg in blood, urine, and hair samples from women, children, and men, exceed legal limits in different communities [47].

2.3.1. The Vulnerability Index to Hg Pollution (IVm)

In this work, an index method was applied to assess the environmental vulnerability on the hydrographic basins based on the estimates from Bruno et al. [18] geographically mapped as for the different distributions described above. The Index of the Hg pollution Vulnerability (IVm) describes the potential impacts of ASGM activities on the population living in a basin through its potential flowing following the orography and surface waters. This index is useful to evaluate the effects of Hg not only on miner communities but also on the human population that lives downstream in these river basins. All the Hg discharge points in a given basin were considered and then followed the orography using a flooding-like algorithm to identify all the downward points interested by the flowing of the discharged Hg (Figure 1). Finally, the population living in each of these potentially polluted points has been considered as a factor to compute the effective impact of Hg discharged from ASGM.

Following is a more rigorous mathematical description.

First, within each basin, the set of points where the discharge of Hg from ASGM is greater than 0 was determined,

$$I = \{i_1, i_2, \dots, i_N\}.$$

Second, in the same basin, for each element of the set I , the set of the points affected by the flowing of Hg from the point i (i.e., all the points downward in the basins) was determined using a free-flooding algorithm,

$$J^i = \{j_1^i, j_2^i, \dots, j_M^i\}.$$

Then, the IVm for any single impacted point $j \in J^i$, $IVm_{j \in J^i}$ can be written as

$$IVm_{j \in J^i} = \frac{R_i}{\sum_Y A_i} \times D_j$$

where R_i is the amount of Hg discharged in the point i , D_j is the population density and A_j is the area of the affected point $j \in J^i$. Having this information is then possible to compute the IVm per country, $IVm_{country}$, by summing the contribution of each $IVm_{j \in J^i}$, for each $j \in C$, where C is the set of geographical points belonging to the country.

Since the set $I = \{i_1, i_2 \dots i_N\}$ depends on the Hg release distribution used, at the end four different IVms were obtained, one for each distribution used in this study: IVm_{AMAP} , for AMAP distribution; IVm_{EDGAR} , for EDGAR distribution; IVm_{DMG} , for distribution using only demographic information; $IVm_{DMG+RUA}$, considering land use, demography, as summarised in the following Table 1.

Table 1. Details of the Vulnerability Index to Hg pollution (IVm) calculated using four different distributions of the ASGM Hg discharges as estimated in Bruno et al. [18].

Vulnerability Index	Distribution Applied	Discharges
IVm_{AMAP}	AMAP distribution	from [18]
IVm_{EDGAR}	EDGAR distribution	from [18]
IVm_{DMG}	Demography	from [18]
$IVm_{DMG+RUA}$	Demography plus rural areas	from [18]

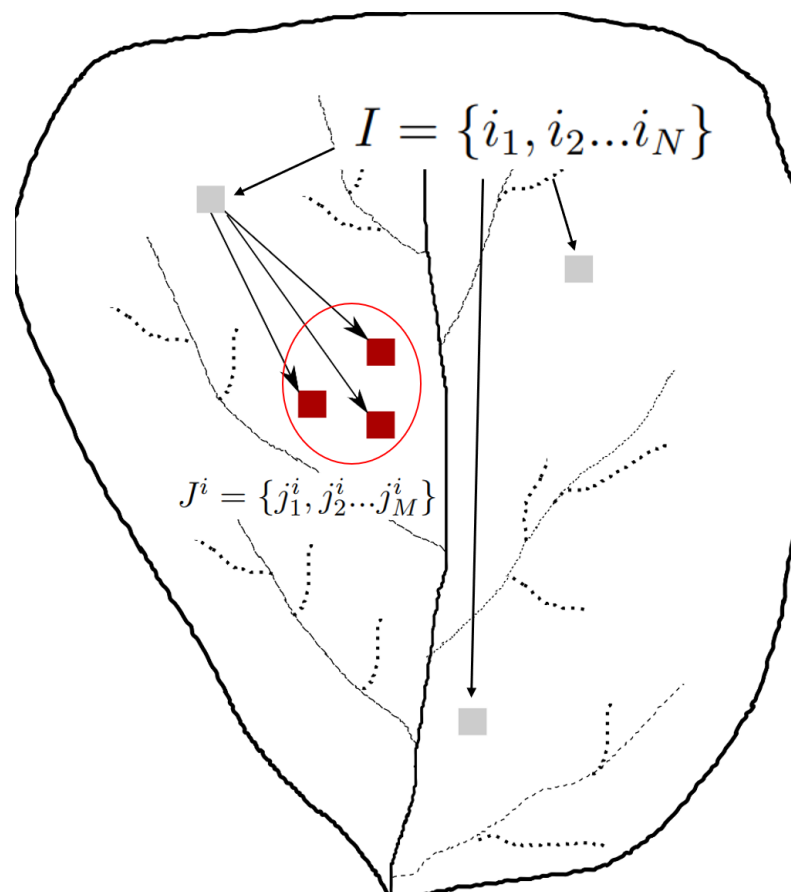


Figure 1. Determination of the Vulnerability Index IVm.

The algorithm was implemented in Matlab using a standard flooding-like algorithm. All required geo-referenced files, digital elevation, population density, and emissions from AMAP and EDGAR were downloaded from the respective sources. The files, where not available in netcdf, were all converted into netcdf on the same native grids using Matlab standard functions, to ensure being processed in a uniform way in the next steps. All grids were converted on the same regular grid at 0.025° making use of the Climate Data Operator, CDO [48], using the appropriate re-gridding algorithms: conservative for Density

and Emissions, and bilinear for Digital elevation. Finally, these files are then used in the computation. Since the nominal error of digital elevation was 8 m, this margin was considered to discern between the height differences among adjacent grid cells in the flooding algorithm.

3. Results and Discussions

3.1. Emissions and Discharges on Rural Areas

As our first analysis, the Hg emissions from ASGM as estimated from AMAP using the native distribution (i.e., *AMAP* distribution) were compared with those estimated in Bruno et al. [18] and distributed using the complete *DGM + RUA* distribution. The map of the differences is presented in Figure 2. The chromatic scale of the legend indicates, from the coldest colors to the warmest colors the passage from the higher values of Bruno et al. [18] estimates to the opposite situation where the values of AMAP emissions are the greatest. In terms of Hg emitted into the atmosphere, for most points, the estimates of Bruno et al. [18] are higher. This trend is particularly evident in some African countries: Niger, Mali, Mozambique, Namibia, Madagascar, and Chad, for which 6.91 kg/cell against 0.000035 kg/cell is recorded. This tendency also applies to some areas of Asian and South American countries. The AMAP estimates are higher in Sudan and Brazil, where the maximum differences are, respectively, 79.28 kg/cell against 0.014 kg/cell and 66 kg/cell against 0.025 kg/cell.

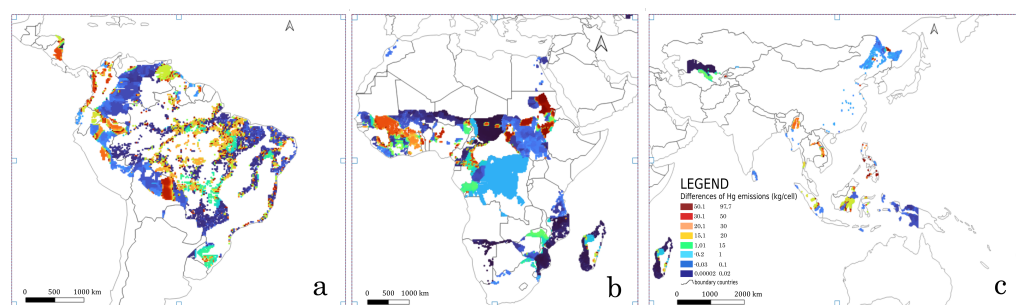


Figure 2. ASGM Hg emissions differences between AMAP estimates, using their native distribution, and Bruno et al. [18] estimates, distributed using *DGM + RUA* distribution. Details are provided for countries in South America (a), Africa (b), and Asian blocks (c).

The differences between the *AMAP* and *DGM + RUA* distributions can be attributed to the different proxies utilized, and to the relative “weights” assigned to them. The assumption for AMAP was that the roasting process took place mainly in city shops. The presence of gold resources, where available, was used to improve this proxy. In addition, AMAP uses the population as a proxy to define the distribution of Hg emissions, but it must be paired with other proxies to be functional [49]. The identification of active sites using satellite information generally presents a high level of uncertainty. In summary, AMAP proceeded to assume that the extension of the ASGM concerned larger areas with the awareness that the final mapping, on a national scale, is still incomplete [50]. Indeed, AMAP inventory was developed for the scope of covering the atmospheric emissions from ASGM. In this regard, the exact position of the Hg emission points is not critical for the atmospheric cycle of elemental Hg, since it reacts very slowly and is very likely to reach the global pool. Exceptions can apply for some areas where vegetation can have a role in absorbing the elemental Hg [51]; however, this is beyond the scope of this study.

Conversely, in this study, for the definition of *DGM + RUA* distribution, the intersection of different layers was used. In addition to the population density (less than 150 hab/km²), land use and the presence of water needed to process the raw material, were considered, as well as the appropriate topography necessary to carry out the artisanal mining activities.

In this regard, Figure 3 shows the geographical distribution of Hg emissions and discharges as estimated in Bruno et al. [18] with *DGM + RUA* distribution. Both emissions and discharges occur at the same locations. However, based on formulas proposed by O'Neill et al. [52] for the two possible amalgamation processes (i.e., CA and WOA) the ratio between Hg emissions and discharges can vary, see Telmer and Veiga [17] for the details, and therefore the values among the two distributions (Figure 3a,b).

In South America, emissions and discharges are higher along the entire western belt, where data on the demographic distribution are also higher. The same results can be observed for the countries of sub-Saharan Africa and for some areas of Indonesia and China. However, the geographical distribution also reflects the selection made initially on land use.

The discharges show the highest values in South America along a belt across Peru, Chile, Colombia, Bolivia, Guyana, and Suriname, whereas in Asia it coincides in the easternmost part of China. In Africa, the highest values (about 9×10^{-5} kg/cell) are reached in Chad and Madagascar.

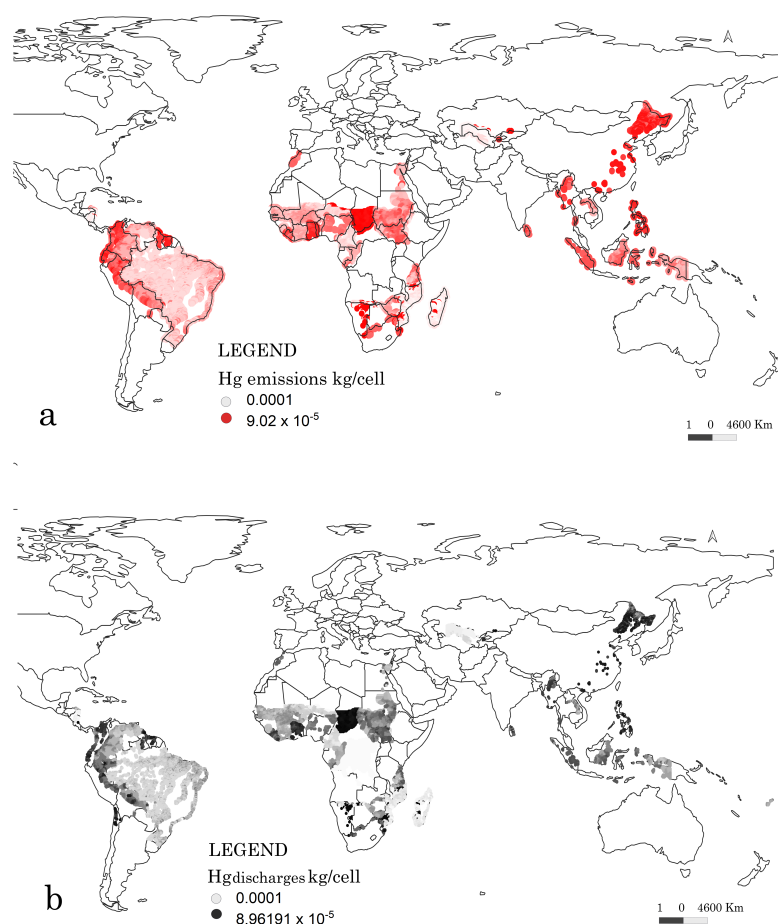


Figure 3. Geographical distributions of Hg emissions (a) and discharges (b) as estimated in Bruno et al. [18] and using the *DGM + RUA* distribution.

3.2. Vulnerability Index to Hg Pollution (IVm)

The values of the Vulnerability Index to Hg pollution (IVm) as calculated for the four different distributions of ASGM Hg discharges (see Table 1), are presented in the following Tables. In particular, Table 2 reports IVm values for those countries for which the inventory of Bruno et al. [18] estimates Hg discharges different from zero, whereas Table 3 reports IVm values for those countries for which the inventory of Bruno et al. [18] estimates Hg discharges equal to zero.

Table 2. The values of IVm as calculated for the different four distributions of ASGM Hg discharges considered IVm_{DMG} , IVm_{AMAP} , IVm_{EDGAR} , $IVm_{DMG+RUA}$ for those countries having estimates of Hg discharges different from zero according to Bruno et al. [18]. Numbers in the bracket report the quartile of the IVm.

Country	Discharge	IVm_{DMG}	IVm_{AMAP}	IVm_{EDGAR}	$IVm_{DMG+RUA}$
Algeria	0.13	9.62×10^{-01} (2)	$0.00 \times 10^{+00}$ (1)	2.45×10^{-02} (1)	1.93×10^{-02} (1)
Bolivia	34.31	$1.14 \times 10^{+03}$ (4)	$5.23 \times 10^{+02}$ (4)	$1.17 \times 10^{+03}$ (4)	$4.26 \times 10^{+02}$ (4)
Brazil	25.94	$8.84 \times 10^{+19}$ (4)	$3.46 \times 10^{+20}$ (4)	$1.00 \times 10^{+20}$ (4)	$1.10 \times 10^{+20}$ (4)
BurkinaFaso	6.31	$4.82 \times 10^{+02}$ (4)	$3.43 \times 10^{+02}$ (4)	$1.03 \times 10^{+03}$ (4)	$4.82 \times 10^{+02}$ (4)
Burundi	0.62	$5.45 \times 10^{+01}$ (3)	$1.51 \times 10^{+02}$ (3)	$3.60 \times 10^{+02}$ (4)	$0.00 \times 10^{+00}$ (1)
Cameroon	0.58	$3.16 \times 10^{+01}$ (3)	$1.25 \times 10^{+01}$ (2)	$4.46 \times 10^{+01}$ (3)	$3.09 \times 10^{+01}$ (3)
Chad	0.02	$5.03 \times 10^{+01}$ (3)	$1.71 \times 10^{+01}$ (2)	$5.54 \times 10^{+00}$ (2)	$5.49 \times 10^{+01}$ (3)
Chile	17.76	$7.48 \times 10^{+01}$ (3)	$2.25 \times 10^{+01}$ (3)	3.41×10^{-04} (1)	$0.00 \times 10^{+00}$ (1)
China	353.25	$2.78 \times 10^{+04}$ (4)	$1.80 \times 10^{+20}$ (4)	$4.15 \times 10^{+04}$ (4)	$3.73 \times 10^{+04}$ (4)
Colombia	74.02	$4.39 \times 10^{+03}$ (4)	$7.35 \times 10^{+22}$ (4)	$1.25 \times 10^{+17}$ (4)	$3.13 \times 10^{+03}$ (4)
Dem. Rep. Congo	11.59	$6.56 \times 10^{+19}$ (4)	$5.11 \times 10^{+18}$ (4)	$7.38 \times 10^{+19}$ (4)	$1.25 \times 10^{+20}$ (4)
Ecuador	22.81	$5.74 \times 10^{+02}$ (4)	$3.22 \times 10^{+02}$ (4)	$6.11 \times 10^{+02}$ (4)	$1.65 \times 10^{+02}$ (3)
Egypt	2.99	$1.37 \times 10^{+20}$ (4)	1.10×10^{-03} (1)	$0.00 \times 10^{+00}$ (1)	$2.77 \times 10^{+02}$ (4)
EquatorialGuinea	0.46	5.28×10^{-01} (2)	4.24×10^{-01} (2)	1.76×10^{-01} (1)	5.30×10^{-01} (2)
Ethiopia	0.2	$3.63 \times 10^{+01}$ (3)	$6.61 \times 10^{+01}$ (3)	$2.18 \times 10^{+01}$ (3)	$2.18 \times 10^{+01}$ (3)
France	32.06	$4.62 \times 10^{+01}$ (3)	$2.77 \times 10^{+01}$ (3)	$3.47 \times 10^{+01}$ (3)	$5.77 \times 10^{+01}$ (3)
Ghana	59.44	$2.36 \times 10^{+03}$ (4)	$8.86 \times 10^{+02}$ (4)	$4.04 \times 10^{+02}$ (4)	$2.36 \times 10^{+03}$ (4)
Guinea-Bissau	26.81	$2.11 \times 10^{+02}$ (3)	$1.31 \times 10^{+02}$ (3)	$1.48 \times 10^{+02}$ (3)	$2.31 \times 10^{+02}$ (3)
Guyana	21.13	$2.63 \times 10^{+01}$ (3)	$1.65 \times 10^{+01}$ (2)	$8.56 \times 10^{+00}$ (2)	$1.33 \times 10^{+01}$ (2)
Honduras	0.39	$1.53 \times 10^{+00}$ (2)	8.14×10^{-01} (2)	$1.11 \times 10^{+00}$ (2)	1.15×10^{-01} (1)
Indonesia	49.68	$3.40 \times 10^{+02}$ (4)	$4.14 \times 10^{+02}$ (4)	$8.87 \times 10^{+01}$ (3)	$3.06 \times 10^{+02}$ (4)
IvoryCoast	3.84	$2.21 \times 10^{+02}$ (3)	$1.82 \times 10^{+02}$ (3)	$2.50 \times 10^{+02}$ (4)	$2.22 \times 10^{+02}$ (3)
Kenya	0.46	$4.55 \times 10^{+01}$ (3)	$3.96 \times 10^{+01}$ (3)	$7.77 \times 10^{+01}$ (3)	$5.58 \times 10^{+00}$ (2)
Kyrgyzstan	4.07	$2.32 \times 10^{+02}$ (3)	$8.67 \times 10^{+01}$ (3)	$8.43 \times 10^{+01}$ (3)	$4.31 \times 10^{+02}$ (4)
Laos	1.91	$8.32 \times 10^{+01}$ (3)	$5.47 \times 10^{+01}$ (3)	$4.76 \times 10^{+01}$ (3)	$1.16 \times 10^{+02}$ (3)
Liberia	18.06	$3.16 \times 10^{+02}$ (4)	$2.44 \times 10^{+02}$ (3)	$9.27 \times 10^{+02}$ (4)	$3.16 \times 10^{+02}$ (4)
Madagascar	0.15	$4.05 \times 10^{+00}$ (2)	$1.87 \times 10^{+00}$ (2)	$3.95 \times 10^{+00}$ (2)	3.99×10^{-01} (2)
Mali	10.54	$3.09 \times 10^{+02}$ (4)	$5.86 \times 10^{+02}$ (4)	$8.75 \times 10^{+02}$ (4)	$3.15 \times 10^{+02}$ (4)
Mongolia	6.44	$4.13 \times 10^{+01}$ (3)	$2.64 \times 10^{+01}$ (3)	$3.49 \times 10^{+00}$ (2)	$0.00 \times 10^{+00}$ (1)
Morocco	3.15	$6.80 \times 10^{+00}$ (2)	$0.00 \times 10^{+00}$ (1)	$0.00 \times 10^{+00}$ (1)	0.00 (NA)
Mozambique	0.33	$1.02 \times 10^{+01}$ (2)	$5.33 \times 10^{+00}$ (2)	$3.13 \times 10^{+00}$ (2)	$9.86 \times 10^{+00}$ (2)
Myanmar	21.86	$1.40 \times 10^{+03}$ (4)	$1.04 \times 10^{+03}$ (4)	$6.06 \times 10^{+02}$ (4)	$1.75 \times 10^{+03}$ (4)
Namibia	0.06	3.19×10^{-01} (1)	1.06×10^{-01} (1)	$0.00 \times 10^{+00}$ (1)	8.39×10^{-01} (2)
Nicaragua	0.02	4.58×10^{-01} (2)	6.19×10^{-01} (2)	2.87×10^{-01} (1)	1.77×10^{-01} (2)
Niger	0.57	$4.67 \times 10^{+01}$ (3)	$1.58 \times 10^{+01}$ (2)	$5.78 \times 10^{+01}$ (3)	$4.85 \times 10^{+01}$ (3)
Nigeria	13.91	$1.24 \times 10^{+03}$ (4)	$3.15 \times 10^{+03}$ (4)	$3.58 \times 10^{+03}$ (4)	$1.25 \times 10^{+03}$ (4)
PapuaNewGuinea	21.32	$5.17 \times 10^{+05}$ (4)	$2.17 \times 10^{+02}$ (3)	$7.10 \times 10^{+01}$ (3)	$1.65 \times 10^{+06}$ (4)
Peru	93.41	$2.18 \times 10^{+03}$ (4)	$8.57 \times 10^{+02}$ (4)	$1.51 \times 10^{+03}$ (4)	$1.71 \times 10^{+03}$ (4)
RepublicoftheCongo	11.59	$4.52 \times 10^{+01}$ (3)	$5.21 \times 10^{+01}$ (3)	$2.38 \times 10^{+03}$ (4)	$4.82 \times 10^{+01}$ (3)
Rwanda	0.02	$6.16 \times 10^{+01}$ (3)	$2.33 \times 10^{+02}$ (3)	$5.01 \times 10^{+02}$ (4)	$0.00 \times 10^{+00}$ (1)
Senegal	3.36	$1.00 \times 10^{+02}$ (3)	$7.55 \times 10^{+01}$ (3)	$1.53 \times 10^{+02}$ (3)	$1.08 \times 10^{+02}$ (3)
SierraLeone	0.25	$1.23 \times 10^{+01}$ (2)	$1.71 \times 10^{+01}$ (2)	$6.42 \times 10^{+00}$ (2)	$1.23 \times 10^{+01}$ (2)
Somalia	18.45	$5.92 \times 10^{+20}$ (4)	$1.06 \times 10^{+02}$ (3)	$0.00 \times 10^{+00}$ (1)	$0.00 \times 10^{+00}$ (1)
SouthAfrica	50.83	$1.66 \times 10^{+03}$ (4)	$5.11 \times 10^{+03}$ (4)	$4.02 \times 10^{+03}$ (4)	$2.06 \times 10^{+03}$ (4)
Sudan	72.25	$1.49 \times 10^{+03}$ (4)	$9.13 \times 10^{+02}$ (4)	$3.70 \times 10^{+04}$ (4)	$1.55 \times 10^{+03}$ (4)
Suriname	25.58	$9.55 \times 10^{+00}$ (2)	$8.62 \times 10^{+00}$ (2)	$1.90 \times 10^{+01}$ (3)	$7.09 \times 10^{+00}$ (2)
Tajikistan	1.09	$9.98 \times 10^{+01}$ (3)	$7.93 \times 10^{+01}$ (3)	$4.88 \times 10^{+01}$ (3)	$1.10 \times 10^{+02}$ (3)
Togo	9.83	$5.90 \times 10^{+02}$ (4)	$5.45 \times 10^{+02}$ (4)	$5.00 \times 10^{+02}$ (4)	$5.90 \times 10^{+02}$ (4)
Uganda	0.55	$1.24 \times 10^{+02}$ (3)	$3.68 \times 10^{+02}$ (4)	$5.55 \times 10^{+02}$ (4)	$0.00 \times 10^{+00}$ (1)
UnitedRepublicofTanzania	10.14	$3.57 \times 10^{+02}$ (4)	$9.95 \times 10^{+02}$ (4)	$4.61 \times 10^{+02}$ (4)	$1.80 \times 10^{+02}$ (3)
Uzbekistan	0.26	$1.92 \times 10^{+01}$ (3)	$6.08 \times 10^{+01}$ (3)	$3.90 \times 10^{+01}$ (3)	$1.55 \times 10^{+01}$ (2)
Venezuela	6.96	$1.65 \times 10^{+02}$ (3)	$6.48 \times 10^{+01}$ (3)	$2.44 \times 10^{+01}$ (3)	$1.74 \times 10^{+02}$ (3)
Zimbabwe	5.4	$2.11 \times 10^{+02}$ (3)	$3.11 \times 10^{+02}$ (4)	$3.39 \times 10^{+02}$ (4)	$1.27 \times 10^{+02}$ (3)

Table 3. The values of IVm as calculated for the different four distributions of ASGM Hg discharges considered IVm_{DMG} , IVm_{AMAP} , IVm_{EDGAR} , $IVm_{DMG+RUA}$ for those countries having estimates of Hg discharges equal to zero according to Bruno et al. [18]. Numbers in the bracket report the quartile of the IVm.

Country	Discharge	IVm_{DMG}	IVm_{AMAP}	IVm_{EDGAR}	$IVm_{DMG+RUA}$
Afghanistan	0	1.83×10^{-01} (1)	1.09×10^{-01} (1)	$0.00 \times 10^{+00}$ (1)	1.64×10^{-01} (1)
Angola	0	$6.20 \times 10^{+00}$ (2)	4.04×10^{-01} (2)	9.24×10^{-02} (1)	$2.61 \times 10^{+00}$ (2)
Argentina	0	$3.11 \times 10^{+00}$ (2)	5.96×10^{-04} (1)	4.07×10^{-02} (1)	$4.65 \times 10^{+00}$ (2)
Benin	0	$4.87 \times 10^{+00}$ (2)	$4.02 \times 10^{+00}$ (2)	$1.93 \times 10^{+00}$ (2)	$4.89 \times 10^{+00}$ (2)
Bhutan	0	4.04×10^{-03} (1)	$0.00 \times 10^{+00}$ (1)	$0.00 \times 10^{+00}$ (1)	0.00 (NA)
Botswana	0	$1.11 \times 10^{+01}$ (2)	$1.69 \times 10^{+01}$ (2)	$1.78 \times 10^{+01}$ (2)	$9.82 \times 10^{+00}$ (2)
Cambodia	0	1.17×10^{-01} (1)	1.17×10^{-01} (1)	$0.00 \times 10^{+00}$ (1)	2.82×10^{-01} (2)
CentAfric.	0	$4.43 \times 10^{+00}$ (2)	$2.48 \times 10^{+01}$ (3)	$1.13 \times 10^{+00}$ (2)	$5.00 \times 10^{+00}$ (2)
CostaRica	0	3.65×10^{-03} (1)	4.12×10^{-03} (1)	$0.00 \times 10^{+00}$ (1)	1.15×10^{-02} (1)
Eritrea	0	$1.45 \times 10^{+01}$ (2)	$7.81 \times 10^{+00}$ (2)	9.49×10^{-04} (1)	$1.36 \times 10^{+01}$ (2)
eSwatini	0	$5.43 \times 10^{+00}$ (2)	$0.00 \times 10^{+00}$ (1)	$0.00 \times 10^{+00}$ (1)	1.29×10^{-03} (1)
Gabon	0	$1.95 \times 10^{+00}$ (2)	$2.23 \times 10^{+00}$ (2)	1.40×10^{-01} (1)	$1.95 \times 10^{+00}$ (2)
Gambia	0	3.50×10^{-01} (1)	1.71×10^{-02} (1)	2.95×10^{-01} (1)	3.68×10^{-01} (2)
Guinea	0	$1.63 \times 10^{+01}$ (2)	$5.82 \times 10^{+01}$ (3)	$2.38 \times 10^{+00}$ (2)	$1.67 \times 10^{+01}$ (3)
India	0	2.23×10^{-01} (1)	5.66×10^{-01} (2)	$0.00 \times 10^{+00}$ (1)	7.02×10^{-02} (1)
Kazakhstan	0	$1.39 \times 10^{+00}$ (2)	3.84×10^{-01} (2)	2.78×10^{-03} (1)	$8.56 \times 10^{+00}$ (2)
Lesotho	0	$2.39 \times 10^{+01}$ (3)	$1.07 \times 10^{+02}$ (3)	$1.13 \times 10^{+02}$ (3)	$0.00 \times 10^{+00}$ (1)
Malawi	0	$1.77 \times 10^{+00}$ (2)	1.25×10^{-01} (1)	5.14×10^{-07} (1)	9.54×10^{-02} (1)
Malaysia	0	7.45×10^{-01} (2)	9.15×10^{-01} (2)	$0.00 \times 10^{+00}$ (1)	7.46×10^{-02} (1)
Mauritania	0	$3.08 \times 10^{+00}$ (2)	1.18×10^{-01} (1)	$0.00 \times 10^{+00}$ (1)	$3.19 \times 10^{+00}$ (2)
Nepal	0	2.00×10^{-02} (1)	$0.00 \times 10^{+00}$ (1)	$0.00 \times 10^{+00}$ (1)	0.00 (NA)
NorthKorea	0	$6.87 \times 10^{+00}$ (2)	1.74×10^{-01} (1)	$0.00 \times 10^{+00}$ (1)	$0.00 \times 10^{+00}$ (1)
Pakistan	0	1.04×10^{-03} (1)	7.33×10^{-04} (1)	$0.00 \times 10^{+00}$ (1)	$0.00 \times 10^{+00}$ (1)
Paraguay	0	4.79×10^{-01} (2)	1.23×10^{-02} (1)	2.27×10^{-02} (1)	$1.27 \times 10^{+00}$ (2)
Russia	0	$1.12 \times 10^{+01}$ (2)	$1.67 \times 10^{+00}$ (2)	$2.68 \times 10^{+00}$ (2)	$4.94 \times 10^{+01}$ (3)
Somaliland	0	1.92×10^{-02} (1)	5.91×10^{-03} (1)	$0.00 \times 10^{+00}$ (1)	$0.00 \times 10^{+00}$ (1)
SouthSudan	0	$2.58 \times 10^{+02}$ (4)	$1.84 \times 10^{+02}$ (3)	$2.71 \times 10^{+01}$ (3)	$2.74 \times 10^{+02}$ (4)
Thailand	0	$7.97 \times 10^{+00}$ (2)	$6.41 \times 10^{+00}$ (2)	$1.77 \times 10^{+00}$ (2)	$9.19 \times 10^{+00}$ (2)
Turkmenistan'	0	2.13×10^{-01} (1)	1.02×10^{-01} (1)	$0.00 \times 10^{+00}$ (1)	3.39×10^{-01} (2)
Uruguay	0	8.07×10^{-02} (1)	3.84×10^{-03} (1)	2.28×10^{-06} (1)	1.49×10^{-01} (1)
Vietnam	0	$1.42 \times 10^{+01}$ (2)	1.22×10^{-01} (1)	$0.00 \times 10^{+00}$ (1)	$0.00 \times 10^{+00}$ (1)
Zambia	0	$8.95 \times 10^{+00}$ (2)	$1.03 \times 10^{+00}$ (2)	$1.30 \times 10^{+01}$ (2)	$1.67 \times 10^{+00}$ (2)
Zambia	0	$8.95 \times 10^{+00}$ (2)	$1.03 \times 10^{+00}$ (2)	$1.30 \times 10^{+01}$ (2)	$1.67 \times 10^{+00}$ (2)

The countries in Table 2 with the highest values of IVm are Brazil, China, and the Dem. Rep. of Congo, all having consistencies across the different distributions assessed. Instead, Colombia, Egypt, Papua New Guinea, Somalia, and Sudan have the highest IVm only for fewer distributions. Conversely, regarding the countries in Table 3, all fall in the lower classes of vulnerability. The only exception is South Sudan, falling in the highest quartile when the distributions *DMG* and *DMG + RUA* are applied.

The countries that present the higher variability regarding the related class across the different distributions are Burundi, Mongolia, Rwanda, Somalia, and Uganda, all belonging to three different quartiles based on the distribution applied.

Figure 4 plots the data of the different IVms against the Hg discharges (in Mg), as reported in Table 2.

Data are plotted in log-log to better visualize the pattern of the outliers. The IVms calculated for all the Hg distributions show, in general, a proportional relationship with Hg discharges (the dotted line represents the perfect linearity ($F(x) = x$)). However, important exceptions are present. In particular, for Egypt, the Dem. Rep. of Congo, Somalia, Brazil, Colombia, and China, the calculated impacts are exponentially greater than expected. For two of these countries, Brazil and the Dem. Rep. of Congo, all the distributions used lead to the highest IVms, showing that probably here the demographic distribution in the IVm algorithm plays a decisive role. For other countries, fewer distributions lead to the highest

IVm. This is the case for China, Colombia, Somalia, and Egypt. For Somalia and Egypt, in particular, this happens only with the simple DGM distribution, showing that some proxies for the distribution can lead to a misconception of the impact. It is noteworthy that Egypt is also the only country, together with Chile, that presents an IVm that is exponentially lower than expected according to the Hg discharges. This also demonstrates how different distributions of ASGM sites can lead to IVms differing many (in this case 23) orders of magnitude there-between.

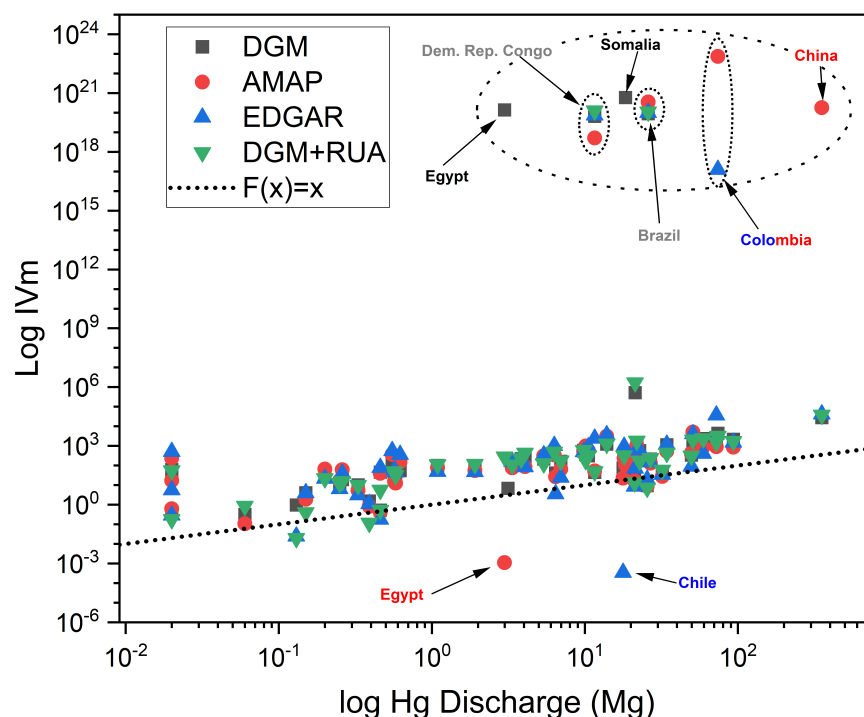


Figure 4. Comparison between all IVm considered: IVm_{AMAP} is the dataset with the most outliers, with respect to $IVm_{DGM+RUA}$. For Brazil, all IVm reenter in the outlier zone.

Considering concentrations greater than $2.5 \mu\text{g/g}$ as a proxy of high exposure levels [53], literature data on Hg concentrations in the hair of workers from some countries show consistency with our findings. In Brazil, several locations were found [54]; Brasilia, 24.7 ng/mg ; Jacareacanga 22.7 ng/mg ; Bocas, 31.4 ng/mg [55]. In Indonesia, precisely at Kalimantan, results showed $17.5 \mu\text{g/g}$ [56]. In Tanzania, $9.33 \mu\text{g/g}$ was found [56]. Consistency was also found in Peru, at Madre de Dios, $11.0 \mu\text{g/g}$ and at Boca Amigo $10.1 \mu\text{g/g}$ [57], and in Guyana (above $15 \mu\text{g/g}$) [58].

Conversely, inconsistency appears to be present in Senegal, belonging to the 3th quartile, but with sampled concentrations lower than $1 \mu\text{g/g}$, at Kedougou and Samekouta [59]. Another inconsistency was found in China, at Chong Qing and Hunan where concentrations resulted in being lower than $1 \mu\text{g/g}$.

4. Limitations and Future Development

As stated elsewhere, the scope of this study is to show how different geographical distributions of Hg discharges, all of these equally valid and reasonable, can lead to very different potential impacts on the environment and populations. The extent of the effects of the dispersion of Hg discharged on the ground by ASGM activities has never been addressed, especially at a global scale. It is precisely in this context that the measure designed here to evaluate such impact, i.e., the Vulnerability Index (IVm), has to be considered. It is a useful and simple tool that allows a quick assessment of a problem that is very challenging to face in a complete manner. Indeed, the exact processes linking Hg discharges in an ASGM site within a basin and its harmful effects on the final end-point (people) are

complex to simulate depending on many factors, including the different habits and diets of peoples [60].

For this work, land use and population density were considered, but obviously, other proxies can be included in the future. In particular, satellite products at different wavelengths and higher resolution can be employed to extract typical features of active/non-active ASGM sites at the local scale for particular terrain characteristics and then used to extrapolate a reliable mapping at larger regional scales.

5. Conclusions

In the context of environmental and health risks linked to Hg used in ASGM activities, this study focused on aspects that have not been adequately investigated until now at a large scale: the impact of Hg discharges in solid matrixes in hydrographic basins through the implementation of a Vulnerability Index. Although Hg emissions into the atmosphere from ASGM activities have been investigated and estimated in recent decades, the geographical distribution of the ASGM sites did not attract the due attention of researchers. Indeed, Hg is emitted from ASGM in a form that slowly interacts with atmospheric chemicals and therefore is very likely to reach the global pool, and therefore the precise locations of the emissions are not critical from an atmospheric point of view. However, this is not true for Hg discharged in the same processes, further considering that Hg emissions are only a fraction of the discharges. Therefore, a need exists for quantifying the impact different geographic distributions of Hg discharges would have on the environment. Using the Vulnerability Index (IVm) applied to two existing distributions, from EDGAR and AMAP inventories, in addition to two novel mapping procedures to identify more precisely the ASGM sites in hydrographic basins of tropical countries, the potential impact of the different geographic distributions of Hg discharges on the environment has been investigated.

Although this is a preliminary study with a qualitative index that only takes into account the surface movements of Hg, it shows that different geographical distributions lead to different impacts. In particular, it is important to note that some proxies used for mapping ASGM, if not properly chosen, can generate discrepancies between Hg discharges and expected impacts.

Furthermore, although the study does not aim to make a comparison between the different distributions used, the comparison with some experimental data, obtained from the literature regarding the Hg concentrations sampled in populations living in the basins, preliminary shows that a careful choice of proxies to map ASGM sites supports a better understanding of the associated risks.

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Abbreviations

The following abbreviations are used in this manuscript:

ASGM	Artisanal Small-scale Gold Mining
RUA	Rural Areas
DMG	Demography
CA	Concentration Amalgamam
WOA	Whole Ore Amalgamation
GMA	Global Mercury Assessment
AMAP	Arctic Monitoring and Assessment Programme
EDGAR	Emission Database for Global Atmospheric Research
CDO	Climate Data Operator
OECD	Organisation for Economic Co-operation and Development
PSN	Piano Strategico Nazionale per lo Sviluppo Rurale

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