

Interactive comment on “4D dispersion of total gaseous mercury derived from a mining source: identification of criteria to assess risks related with high concentrations of atmospheric mercury” by José M. Esbri et al.

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General comments: The manuscript “4D dispersion of total gaseous mercury derived from a mining source: identification of criteria to assess risks related with high concentrations of atmospheric mercury” by Esbri et al. discusses criteria and a minimum amount of information needed to efficiently characterize Hg contaminated site as a result of past mining activities. The authors suggest a novel monitoring design and evaluate it based on results obtained during measurement campaigns in the Almaden mercury mining districts. Overall, the manuscript brings new insights into specific path-

C1

ways of Hg at contaminated sites, as well as the methodology to determine risks associated. The paper is well written and structured, including visualizations, statistical treatment and the interpretation of the results. However, there are some parts of the manuscript that are a bit unclear in its present form and need to be revised and simplified, respectively. To this end, in the following they are some specific comments and suggestions to improve the quality of this work. Specific comments: - Abstract: In its present form the abstract contains too many details, the second and third paragraphs in particular. It is suggested to rewrite it, focusing on the main outcomes of this work, e.g. relevant criteria and data needs for characterization of contamination and associated risks in the spatio-temporal context. Done. We have rewrite second and third paragraph to accomplish reviewer suggestion: The vertical profiles revealed that higher Total Gaseous Mercury concentrations are present at lower altitude during nocturnal hours and at higher altitude at dawn and dusk. Horizontal profiles showed that the background values were close to 6 ng m⁻³ except in the spring months, when they rose to 13 ng m⁻³ and increased the area affected by mercury emissions to more than 4 km around the mining and metallurgical sites. On a daily basis the most important process involved in gaseous mercury movements is the mixing layer, which begins in the early morning and finishes at nightfall. Vertical transferences are predominant when this process is active, i.e., in all seasons except winter, while major sources act as constant suppliers of gaseous Hg to the mixing cell, thus producing Hg deposition at dusk. Conversely, horizontal transferences prevail during the hours of darkness and the main factors are major and minor sources, solar radiation, wind speed and topography. The study has shown that it is important: i) to identify the sources; ii) to get data about Hg movements in vertical and horizontal directions; iii) to extend the measurements over time in a sufficiently representative way, both daily and seasonally; iv) to determine the different populations of data to establish the background levels, this work proposes the use of Lepeltier graphs to do it. In terms of risk assessment, and based on the model constructed to infer atmospheric Hg concentrations based on micrometeorological parameters, the nights carry greater risk than the days in all seasons (54%

C2

in spring and winter, 72% in summer) except in autumn, when 99% of the hours of risk occurred during the day. The main factors involved in the creation of high-risk periods are those related to dilution (or its absence): namely wind speed and solar radiation at null levels. The extent of the area affected by an emission source is independent of its importance in terms of absolute emissions. The affected zone did not extend beyond 100 metres from the location of the source during the daytime period and 200 metres in the night-time. Under the worst micrometeorological conditions, it was predicted that the affected area would cover almost the entire town of Almadenejos, although these risk conditions only represent 11.34% of the hours in an annual period. - Line 46: Revise the definition of TGM Done. Now the sentence is “GEM and RGM species constitute ‘total gaseous mercury’ (TGM)”. - Line 64: “altitudes in the range 500-11,000 metres from background and contaminated locations;. . .”. Not clear. Revise and support with some references. Done. The sentence is now as follows: Most of the available information on this topic is on a kilometric scale, at high altitudes in the range 500–11,000 metres from background and contaminated locations (Slemr et al., 2018; Weigelt et al., 2016); - Lines 73-75: It is not clear what is meant by “Risk assessment”, “. . .worst theoretical conditions. . .” and “. . .the worst-case scenario. . .”. Revise and provide relevant details. Done. The sentence is now as follows: Risk assessments of areas with anthropic contamination of gaseous Hg are often carried out with scarce data, often corresponding to short periods of time, and these do not provide a representative view of the day-night contrast or the seasonality, not even at the level of hot and cold or dry and wet seasons (depending on the location of the case study). We have conducted studies based on sampling times selected in the worst theoretical conditions, with higher expected emission rates enhanced by temperature and solar radiation, with the aim of identifying the worst-case scenario in summer days without winds in a mining site in Almadén (Martínez-Coronado et al., 2011), in a mining complex in Mount Amiata-Italy (Vaselli et al., 2013) in a chloralkali plant in Tarragona (Esbri et al., 2015), in a chloralkali plant in Romania (Esbri et al., ; 2018a) and in a period of time with higher Hg metallurgical works in Almadén (Tejero et al., 2015), the evaluation

C3

of background conditions (Higuera et al., 2014), and comparison of the worst and best scenarios (Higuera et al., 2013). - Lines 80-88: Not clear how the mentioned reference (Deng et al., 2016) is linked with the rest of the paragraph. It is also suggested to shorten and simplify this whole paragraph. Thanks for the suggestion, but we think it is important to put into perspective the argument that it is necessary to rethink the representativeness of the data when doing a mercury-related risk analysis. In many cases sampling (o monitoring?) is not performed (o carried out) in the worst conditions due to ignorance, since the variations in environmental concentrations of mercury can be very large in space and time and may be due to local reasons, not predictable based on the scientist's previous experience. For these reasons, we think that cited a reference like Deng et al (among many others), is relevant to support the idea. - Line 97: Provide more details on the “exhaustive identification” of sources in the study area. By what means these sources were identified? Done. The sentence is now as follows: In this work we have tried to obtain the minimum information necessary about the emission, transport and deposition of atmospheric mercury to ensure the representativeness of such data with a minimum cost in terms of effort and money. Before designing the sampling locations, an exhaustive identification of the Almadenejos emission sources, represented in red in Fig. 1, was carried out with a Lumex RA-915M equipment in mobile monitoring mode using a car to cover the entire area. - In Lines 100-103 emission sources in the study area are ranked according to their importance. Based on what criteria? Done. A new sentence has been added: The importance of the sources has been established if the average concentrations are below 200 ng m⁻³ (low importance), in the range of 200-1000 ng m⁻³ (medium importance) or up to 1000 ng m⁻³ (high importance). - Line 109: Check if coordinates of AWTP are written in a correct format Checked. - Line 115: I suggest leaving out the sentence starting with “This situation gives. . .” Yes, we have deleted this sentence. - Lines 242-243: How were the background locations defined and separated from the rest? Background values were determined using Lepeltier graphs. Technical corrections: - Line 120: Check values indicated in brackets for Lower and Upper Gradient Yes, we have checked and they

C4

are correct. - Lines 174-177: In Figure 2 there are no A, B and C mentioned in the text Yes, the reference to the figure were incorrect. We have deleted A, B and C in the text. – Page 19: Location should be mentioned in Table 2 caption Done. We have changed the sentence: Table 1. Statistical summary of TGM levels at different heights (3, 2 and 0.5 metres) and total gradient (3–0.5 m), upper gradient (3–2 m) and lower gradient (2–0.5 m) in Almadenejos WWTP. All TGM data are in ng m^{-3} . - Figure 5: units are not shown for scale bar in Profile 1 and Profile 3, respectively Yes, the scale was missing. We have revised the figure, adding these scales. - Figure 9: scale bar is missing Done.

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C5

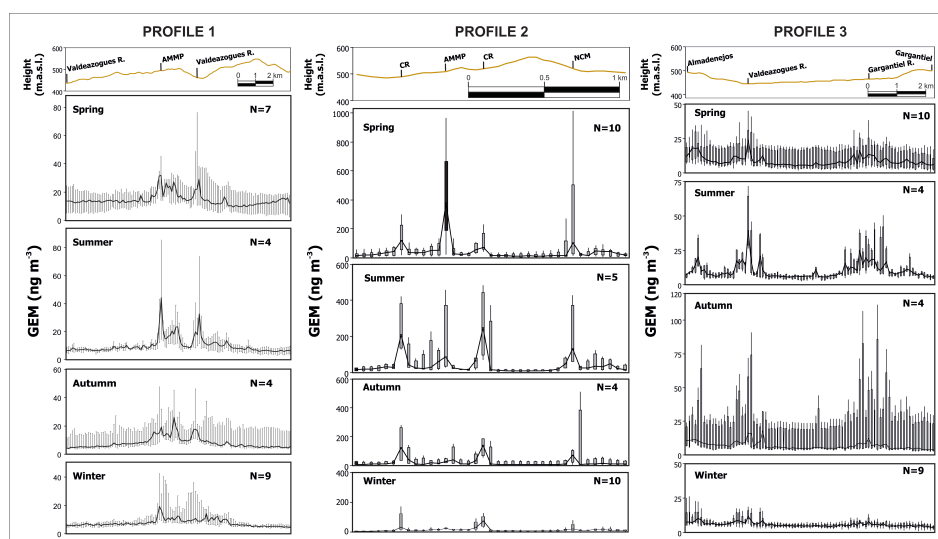


Fig. 1. Figure 5 revised

C6

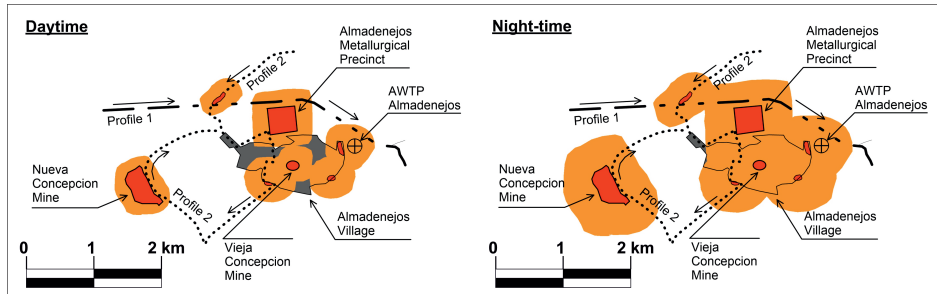


Fig. 2. Figure 9 revised