

Daysimetric mapping of mercury emissions from contaminated sites

S. Cinnirella¹, N. Pirrone², M. Horvat³ and D. Kocman³

¹ CNR-Institute of Atmospheric Pollution Research, Division of Rende, Rende, Italy, s.cinnirella@iia.cnr.it

² CNR-Institute of Atmospheric Pollution Research, Monterotondo Scalo, Italy

³ Jožef Stefan Institute, Ljubljana, Slovenia

Abstract. In this abstract we describe the Dasymetric Mapping (DM) technique applied to mercury contaminated sites. The DM is an intelligent disaggregation procedure that incorporates ancillary data to facilitate the areal interpolation process. It differs from choropleth mapping in that areas are defined based on the actual spatial distribution of the variable being mapped, rather than administrative or other arbitrary units. The disaggregation weight was derived from soil carbon content, as it was found that increases in total mercury (and methylmercury) concentrations are related to increasing fractions of organic carbon (OC) in the soil. The DM technique was implemented in ArcGis® by creating a model that uses emissions from contaminated sites, an area-class map, and a sampling strategy set out as binary.

Key words: gridded emission, contaminated soils, soil organic carbon

Introduction

In the last decade, considerable progress has been made in better understanding the sources, transport routes and behaviour of mercury in the global environment (Pirrone and Mason, 2010). While anthropogenic sources have been the object of several papers, only recently have natural sources received growing attention, as they have the potential to make significant contributions to regional atmospheric mercury budgets and overall global mercury pollution. (Pacyna et al., 2010; Pirrone et al., 2010).

In addition, much effort has been given to modelling assessments of the global mercury cycle (Pirrone et al., 2001; Hedgecock et al., 2006; Pacyna et al., 2006; Bullock and Jaegle, 2009; Dastoor and Davignon, 2009; Friedli et al., 2009; Jaegle et al., 2009; Jung et al., 2009; Seigneur et al., 2009; Travnikov and Ilyin, 2009). The most up-to-date assessments of global scales have shown that anthropogenic sources in 2005 contributed between 1926 and 2320 Mg yr⁻¹ (Pacyna et al., 2010; Pirrone et al., 2010), whereas emissions from natural sources may represent a larger contribution (up to 5200 Mg yr⁻¹) to the global atmospheric mercury budget (Pirrone et al., 2010). However, in these assessments, contributions from contaminated areas surrounding industrial and mining activities are usually not taken into full account, or they are neglected entirely. One recent assessment (Kockman et al., 2012) demonstrated that contaminated sites make a substantial contribution to the global budget. One

weakness of this work is that it did not consider the gridded distribution of mercury emission from contaminated sites. To improve assessments of contaminated sites, we adopted a rather new mapping technique that successfully made gridded emissions from contaminated sites available to modellers.

The prominent method of areal interpolation we adopted is known as Dasymetric Mapping (DM). An intelligent disaggregation procedure that incorporates ancillary data to facilitate the areal interpolation process, Dasymetric Mapping differs from choropleth mapping in that areas are defined based on the actual spatial distribution of the variable being mapped, rather than other arbitrary units. This technique has gained popularity as satellite imagery has become increasingly available, and as methods of utilising Earth Observation (EO) data in a GIS have steadily improved.

Materials and Methods

Dasymetric mapping is defined as the use of an ancillary data set to disaggregate a variable data from a coarse resolution to a finer one (Eicher and Brewer 2001). This mapping technique can be considered one approach to the polygon overlay areal interpolation problem, which seeks to improve on areal weighting by establishing a relationship between the underlying statistical surface and the different classes contained within the area-class map. The technique developed by Mennis and Hultgren (2006)

takes count data mapped to a set of source zones and a categorical ancillary data set as input, and redistributes the data to a set of target zones formed by an intersection of the source and ancillary zones. Data are redistributed based on a combination of areal weighting and the relative densities of ancillary classes (Mennis 2003).

The estimated count for a given target zone \hat{y}_t is calculated as:

$$\hat{y}_t = \sum_{s=1}^n \frac{y_s A_{s \cap z}}{A_s}$$

where y_s is the count of the source zone, $A_{s \cap z}$ the area of the intersection between the source and target zone; A_s the area of the source zone and n the number of source zones with which z overlaps.

DM supports the ability to combine domain knowledge (using preset density values) with statistical estimation (using empirical sampling) to quantify this relationship. In our application of DM, disaggregation weight was derived from soil carbon content as it was found that increases in total mercury (and methylmercury) concentration is related to increasing fractions of organic carbon (OC) in the soil (Pant and Allen, 2007).

With the exclusion of naturally enriched substrates, soils receive most of the mercury from depositions. Deposited Hg normally appears as Hg^0 and Hg^{2+} , the latter in the form of complexes (Wang et al., 2009). In addition, the major fraction of Hg^{2+} is often bound in soil minerals or adsorbed on solid surfaces. Since $HgCl_2$ is only weakly retained by mineral matter, it can be assumed that Hg^{2+} is mainly attracted to organic matter in acid soils, whereas in neutral and slightly alkaline soils, mineral compounds are also active.

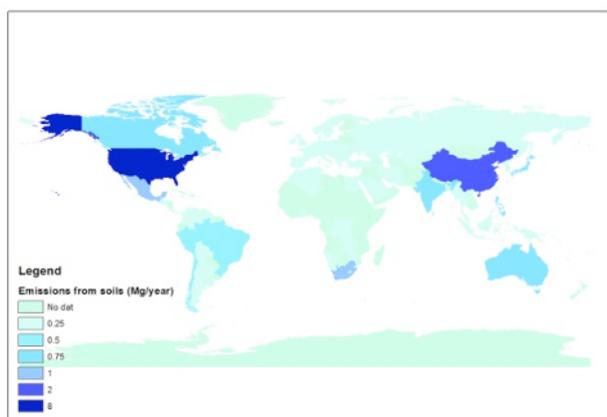


Fig. 1. Scaled mercury re-emissions from contaminated sites (by courtesy of Kocman et al., 2012)

This DM technique was applied to emissions from contaminated sites as reported in Kocman et al. (2012). For the first time, the authors were able to estimate the amount of mercury released from contaminated sites into the global mercury budget. They did so by using a geo-referenced database built on more than 3000 mercury

contaminated sites associated with mercury mining, precious metal processing, non-ferrous metal production, and various other polluted industrial sites. In their assessment, contaminated-site mercury releases to both the atmosphere and hydrosphere were considered; whereas in our work, we consider only mercury releases to the atmosphere (Figure 1).

The soil OC map was obtained from the Natural Resources Conservation Service (NRCS) of the U.S. Department of Agriculture (<http://soils.usda.gov/use/worldsoils/mapindex/soc.html>). It was based on reclassification of the FAO-UNESCO Soil Map of the World, and then combined with a soil climate map. The map has a 2 minute grid cell and six classes of soil organic carbon volume ranges (Figure 2).

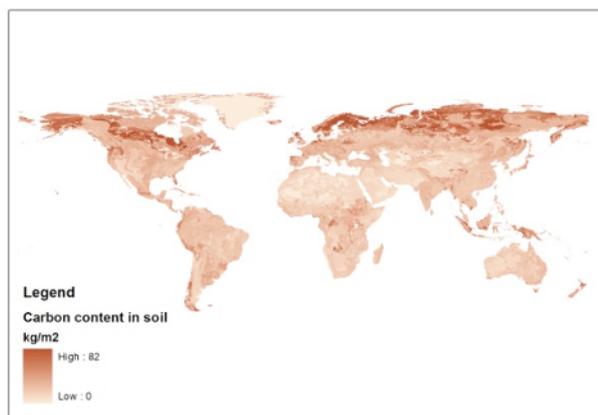


Fig. 2. Soil organic carbon content (from NRCS, 2007)

At the end of the interpolation process, the difference between the estimated and actual mercury content in soils was calculated, and the root mean square (RMS) error was calculated to assess error.

The dasymetric mapping technique was implemented in ArcGis® by creating a model that uses emissions from contaminated sites, an area-class map, and a sampling strategy set out as binary.

Results and Discussion

Figure 3 shows the map of total mercury emissions from soils produced using the dasymetric mapping technique. Clearly, the map offers a far more detailed depiction of overall mercury distribution in soils, as it gives a “pixeled” value - thereby avoiding uniform values for wide regions. As expected, greater emissions were found in those regions where mercury re-emission from soils occurred as a consequence of mercury mining, precious metal processing, non-ferrous metal production, and various polluted industrial and urban sites.

Mapped mercury releases from contaminated sites to the global mercury budget indicates that, from a global perspective, most mercury contaminated sites that were identified (>70%) are concentrated in industrial regions of Southern Europe, Western North America, and China. It must be noted, however, that the map presented here is associated with a large degree of uncertainty, mostly due

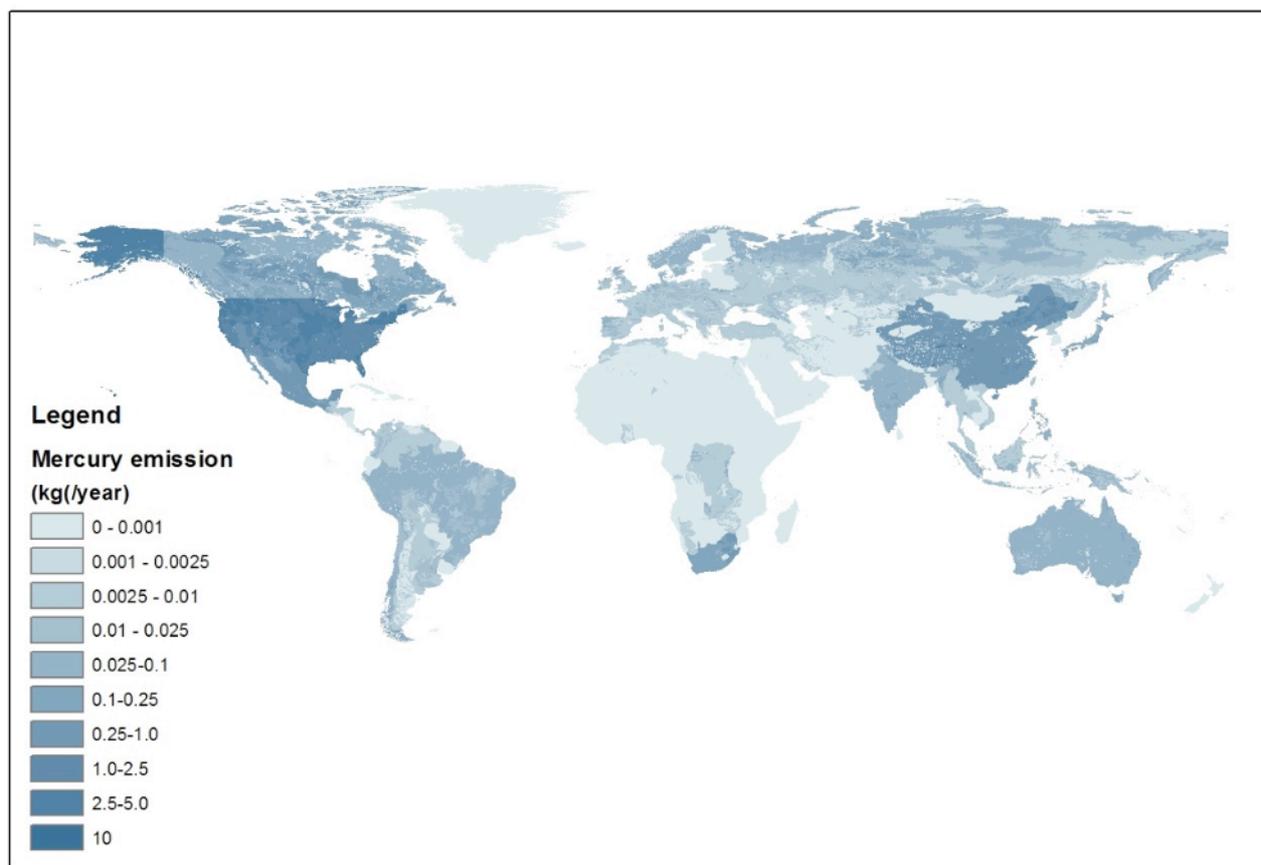


Fig. 3. The gridded mercury emission map for contaminated soils.

to the scarcity of data on mercury releases from contaminated sites, as was stated in the original estimate of mercury re-emission from soils.

Conclusion

In this research we applied a dasymetric mapping technique to grid of contaminated soils from mercury mining, precious metal processing, non-ferrous metal production, and various polluted industrial and urban sites. This prominent technique is generally used to improve accuracy estimates for population density, by means of land use classes; whereas its application to environmental parameters has never been tried. By adopting the organic carbon content of soils as an a priori driver, our work provides some clue to the real mercury content of soils by examining mercury associated with organic carbon. Nevertheless, more research is needed to characterize the relationship between deposition and mercury content in soils, as well as on mercury distribution around contaminated sites.

Acknowledgements

The authors would like to express their gratitude for the financial support provided by the GMOS Project (FP7-ENV-2010 No. 265113).

References

Bullock, O. R., and L. Jaeglé (2009), Importance of a global scale approach to using regional models in the assessment of source-receptor relationships for mercury, in *Mercury Fate and Transport in the Global Atmosphere: Emissions, Measurements, and Models*, edited by N. Pirrone and R. P. Mason, 503-517 pp., Springer, New York.

Dastoor, A. P., and D. Davignon (2009), Global mercury modeling at Environment Canada, in *Mercury Fate and Transport in the Global Atmosphere: Emissions, Measurements, and Models*, edited by N. Pirrone and R. P. Mason, 519-531 pp., Springer, New York.

Eicher, C L and Brewer, C A, 2001, "Dasymetric mapping and areal interpolation: implementation and evaluation" *Cartography and Geographic Information Science* 28 125-138

Friedli, H. R., et al. (2009), Mercury emissions from global biomass burning: spatial and temporal distribution, in *Mercury Fate and Transport in the Global Atmosphere: Emissions, Measurements, and Models*, edited by N. Pirrone and R. P. Mason, 193-221 pp., Springer, New York.

Hedgecock I.M., Pirrone N.,Trunfio G.A., Sprovieri F., 2006. Integrated mercury cycling, transport and air-water exchange (MECAWEx) model. *Journal of Geophysical Research*, 111 (D20302).

Kocman D., Horvat M., Pirrone N., Cinnirella S., 2012. Contribution of contaminated sites to the global

- mercury budget. *Environmental Research* (submitted).
- Mennis, J. 2003. Generating surface models of population using dasymetric mapping. *The Professional Geographer* 55: 31-42.
- Mennis, J., and T. Hultgren. 2006. D Intelligent Dasymetric Mapping and Its Application to Areal Interpolation. *Cartography and Geographic Information Science*, Vol. 33, No. 3, 2006, pp. 179-194.
- Pacyna, E.G., Pacyna, J.M., Steenhuisen F., Wilson S., 2006b. Global anthropogenic mercury emission inventory for 2000. *Atmospheric Environment*, 40: 4048-4063.
- Pacyna, E. G., et al., 2010. Global emission of mercury to the atmosphere from anthropogenic sources in 2005 and projections to 2020, *Atmospheric Environment*, 40(20): 2487-2499.
- Pant and Allen, 2007, *Bull Environ Contam Toxicol* 78: 539-542
- Pirrone N., Costa P., Pacyna J.M., Ferrara R., 2001. Mercury Emissions to the Atmosphere from Natural and Anthropogenic Sources in the Mediterranean region. *Atmospheric Environment*. Vol. 35, 2997-3006.
- Pirrone, N., and R. P. Mason (Eds.) (2010), *Mercury Fate and Transport in the Global Atmosphere: Emissions, Measurements, and Models*, 637 pp., Springer, New York
- Pirrone, N., et al. (2010), Global mercury emissions to the atmosphere from anthropogenic and natural sources, *Atmospheric Chemistry and Physics*, 10(13): 5951-5964.
- Seigneur, C., et al. (2009), The AER/EPRI global chemical transport model for mercury (CTM-HG), in *Mercury Fate and Transport in the Global Atmosphere: Emissions, Measurements, and Models*, edited by N. Pirrone and R. P. Mason, 589-601 pp., Springer, New York.
- Travnikov, O., and I. Ilyin (2009), The EMEP/MSC-E mercury modeling system, in *Mercury Fate and Transport in the Global Atmosphere: Emissions, Measurements, and Models*, edited by N. Pirrone and R. P. Mason, 571-587 pp., Springer, New York.
- Wang et al., 2003, *The Science of the Total Environment* 304: 209-214.