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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Correlation slopes of GEM / CO, GEM / CO_2, and GEM / CH_4 and estimated mercury emissions in China, South Asia, Indochinese Peninsula, and Central Asia derived from observations in northwest and southwest China" *by* X. W. Fu et al.

X. W. Fu et al.

fengxinbin@vip.skleg.cn

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Reponse to J. Pacyna

We thank Prof. J. Pacyna for providing the comments regarding the validity of the correlation slope method. We would like to to elaborate how the method works and its associated limitations.

This is an interesting paper adding to the discussion on amount of mercury emissions, particularly in a gaseous elemental form, emitted from various sources in China. This is the region where emission of mercury is the highest compared to other parts of the world. Therefore, it is of particular importance to assess these emissions in China as accurate and complete as possible. The reviewed paper contributes to this process.

Comments: My major comment relates to the application of correlation slopes of GEM/CO2, GEM/CO, and GEM/CH4. I am a bit concerned with the value of such analysis. All four gases are emitted from different sources. The major source of CO2 and mercury is the combustion of fossil fuels in large power plants, mostly coal-fired power plants. The CO emissions are mostly generated in incomplete combustion processes, basically selected industrial processes with poor industrial installations. Methane has a completely different sources/ processes generating emissions, mainly fermentation processes. How the above mentioned ratios of these gases could serve to explain the amount of these emissions? Or even to be compared with the ratios of the same gases measured in certain receptors in different regions? The authors may wish to provide more explanation on this issue.

Reply: The correlation method was firstly applied by Jaffe et al. (2005) to assess the Hg emissions in Asia using the slope of GEM/CO regression line. Later, Slemr et al. (2006) and Brunke et al., 2012 extended the use of the slopes between GEM and other atmospheric trace gases. The principles and assumptions have been discussed extensively by Jaffe et al. (2005) and the other two studies and the community can directly refer to the original development works. We have expanded the discussion regarding the limitations and uncertainties related to the method in the manuscript (line 527-552 on page 17).

First, it is true that GEM and CO/CO2/CH4 may be emitted from different sources with distinct emission characteristics. However, once these different sources are collocated, the pollutants emitted from different sources will well mixed fast and emission ratios are expected to be reflected by the observed correlation slopes of these long-lived

trace gases. Due to the different sources and emission rates, the correlation slopes exhibit both temporal and spatial. To obtain a comprehensive overview on the emission ratios, it is necessary to observe the correlation slopes in different seasons and regions, which was accomplished in this study. Therefore, we are confident that the estimates obtained in this study are representative and reasonably accurate.

One of the major uncertainties, though, is the accuracy of the emission estimates of CO, CO2, and CH4 in the two regions. However, many recent advances have been made in this regard. It is therefore perceived that the uncertainties of CO and CO2 emission estimates are less than 50% (Zhao et al., 2012a;Zhao et al., 2012b;Olivier et al., 2001). Such an uncertainty level is slightly lower than that of anthropogenic GEM (Pacyna et al., 2010;Pirrone et al., 2010). However, the uncertainties of CH4 emission estimates are expected to be larger, which would results in a greater uncertainty in the estimate of GEM emissions using GEM/CH4 ratios. Previous studies have suggested that the correlation slopes may give an estimate of total GEM emissions (including both anthropogenic and natural emissions). These estimates based on correlation slopes are consistent with the results from inverse modeling (Pan et al., 2007). This is important because the current understanding on natural emissions of GEM is poor and the knowledge on total GEM emissions is crucial for global Hg modeling.

Comments: Just to even complicate this issue, the two closest gases in the context of common emission sources, such as coal-fired power plants, namely CO2 and GEM may not be that close in the whole coal energy sector. The point is that GEM emissions from coal-fired power plants are dependent on the type and efficiency of flue gas desulfirization installation (FGDs), while CO2 emissions are not dependent on this factor. So, even within the coal energy sector one would expect rather large ranges of GEM/CO2 ratios. How to use this information for the assessment of GEM emissions from this sector in various regions of Asia or the world? Some more discussion on this application shall be added in the paper with explanation of limitations in such application.

Reply: First, we would like to point out that the emission factors of GEM, CO, CO2, and CH4 for emissions sources used in the present study were cited from most updated studies (line 371-375 on page 12). These emission factors were calculated on the basis of comprehensive field observations and the control measures can represent the predominant technologies used in industrial emission sources. Therefore they represent the most recently updated emission factors.

We agree that there may be significant variations in the emission factors. However, the emission factors in different emission sources varied more significantly than the emission factors among common sources and therefore can be used to trace the sources using observed correlation slopes. For example, the emission ratios of GEM/CO and GEM/CO2 for lead, zinc, mercury, gold productions were two orders of magnitudes higher than that of industrial coal combustion and cement productions. Pollution control devices may change the emission ratios, but the magnitudes of changes are much smaller than the emission ratio differences between different sources.

The correlation slopes method in the present study cannot be used to estimate GEM emissions from a certain emission source. It was used to estimate GEM emissions from different regions, where many sources are located in a given geographic boundary.

References

Olivier, J. G. J., Berdowski, J. J. M., Peters, J. A. H., Bakker, J., Visschedijk, A. J. H., and Bloos, J. P. J.: Applications of EDGAR including a description of EDGAR 3.2: reference database with trend data for 1970-1995, RIVM, Bilthoven. RIVM report 773301001/NRP report 410 200 051., 2001 Pacyna, E. G., Pacyna, J. M., Sundseth, K., Munthe, J., Kindbom, K., Wilson, S., Steenhuisen, F., and Maxson, P.: Global emission of mercury to the atmosphere from anthropogenic sources in 2005 and projections to 2020, Atmos Environ, 44, 2487-2499, DOI 10.1016/j.atmosenv.2009.06.009, 2010. Pan, L., Chai, T. F., Carmichael, G. R., Tang,

Y. H., Streets, D., Woo, J. H., Friedli, H. R., and Radke, L. F.: Top-down estimate of mercury emissions in China using four-dimensional variational data assimilation, Atmos Environ, 41, 2804-2819, DOI 10.1016/j.atmosenv.2006.11.048, 2007. Pirrone, N., Cinnirella, S., Feng, X., Finkelman, R. B., Friedli, H. R., Leaner, J., Mason, R., Mukherjee, A. B., Stracher, G. B., Streets, D. G., and Telmer, K.: Global mercury emissions to the atmosphere from anthropogenic and natural sources, Atmos Chem Phys, 10, 5951-5964, DOI 10.5194/acp-10-5951-2010, 2010. Zhao, Y., Nielsen, C. P., and McElroy, M. B.: China's CO2 emissions estimated from the bottom up: Recent trends, spatial distributions, and quantification of uncertainties, Atmos Environ, 59, 214-223, DOI 10.1016/j.atmosenv.2012.05.027, 2012a. Zhao, Y., Nielsen, C. P., McElroy, M. B., Zhang, L., and Zhang, J.: CO emissions in China: Uncertainties and implications of improved energy efficiency and emission control, Atmos Environ, 49, 103-113, DOI 10.1016/j.atmosenv.2011.12.015, 2012b.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/14/C10175/2014/acpd-14-C10175-2014-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 24985, 2014.