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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Measurements of atmospheric mercury in Shanghai during September 2009" *by* H. R. Friedli et al.

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This paper presents a study on a short-term variation (about 20 days) of atmospheric TGM concentrations at an urban site in Shanghai, East China. I wonder why the authors measured TGM concentrations for a very short duration. Shanghai is located in one of the most serious polluted areas in China, and it is also a costal area. Therefore, it could be expected that this area is affected by large atmospheric emissions from inland and fresh air masses from ocean. As pointed out in the paper, levels of atmospheric TGM concentrations were largely depended on wind direction, and low atmospheric TGM concentrations during the study period were because that the most of the air masses during the whole study period were originated from ocean. Hence, this study may be insufficient to evaluate the overall level of atmospheric TGM in Shanghai,

and this might lead to many uncertainties if the value reported here is used for comparison in future studies. Could the authors make an attempt to estimate the annual level of TGM in Shanghai by using some empirical models? Are there strong correlations between TGM and CO, SO2, and NOx? Many previous studies suggest that GEM is generally linear correlated with CO. Could the authors use these relationship and levels of these criteria pollutants to predict the annual mean of TGM in Shanghai.

Specific comments. In section 3.3, the authors speculated that CFPP were the major contributor to observed TGM during the plume events, which was mainly supported by the high NOx/SO2 ratio (2.78) for the major plume events. Since CFPP was the major contributor for plume events, why was the NOx/SO2 ratio for plume events higher than the emission ration for CFPP. Are the some other emission sources and atmospheric process evolved in these plume events? In section 3.4, measurements of atmospheric Hg in remote areas of China should be also included in the comparison. Table 3, the sampling periods of some studies in China were not presented correctly Figure 1, there is no map scale. Figure 2D, the wind dependence of TGM is not clearly shown.

Response:

First of all, we thank the reviewer for the helpful comments.

1). On short duration. The reviewer wonders why the experiment was of such short duration: this was an exploratory experiment limited by time and funding, and was to be extended at a later date. The pollution behavior was as expected: a composite of the interactions between an industrial center and an ocean. We are carefully avoiding the pitfalls of extrapolation September data to a full year or longer. We want to stress that our data should not be misconstrued for regulatory purposes nor for comparison with other local and global studies without stating the limitations. One of the highlights of this MS is to show the tremendous variability in emissions from SH.

We have stayed away from making annual estimates for TGM emissions for SH because of the large variability and uncertainties of emission ratios or factors. The re-

viewer suggested TGM/CO correlations which are well suited for Hg/biomass burning correlations but show a very large range of ER's for this part of the world, e.g. Friedli et al., 2004 (cited).

We note that in the Abstract, Introduction and Conclusion of the manuscript, we clearly stated the limitation of this work (e.g. "This is an underestimate for an annual-mean concentration because the meteorology in September favored predominantly easterly oceanic air, replaced in other seasons by airflow from industrial areas.")

We have also added a sentence in the Abstract as follows:

"...The September 2009 Shanghai measurements are lower than those reported for most other Chinese cities and Mexico City, and similar to concentrations found in some Asian and in North American cities. Such comparisons are tenuous because of differences in season and year of the respective measurements. Our results should not be used for regulatory purposes. ..."

We have also modified the sentence in the Conclusion as follows:

"To obtain a better assessment of variability and trends, mercury measurements (TGM, preferably augmented by speciated mercury and Hg analysis in aerosol) should be continued or initiated at multiple locations in and around Shanghai. Integrated and long-term measurements (NOX, SO2, CO and meteorology) are mandatory given the large spatio-temporal variability in the observed concentrations. Such data are useful for tracking progress in pollution abatement and as input to predictive models of atmospheric Hg."

2). On Section 3.3. We have rewritten Section 3.3 to reflect our better understanding of the NOX/SO2 relationship and sources. It now reads:

"SO2, NOX, and CO are the most relevant tracers for TGM. While SO2 and NOX are co-emitted with Hg from CFPPs and nonferrous smelting processes, their relative abundance vary greatly among industries, fuel types, degree of pollution abatement and

within each source categories (Lin et al., 2010; Lu et al., 2010). This difference in abundance provides a way to identify the dominance of coal combustion or smelting in the TGM plume that we observed in this study. High temperature combustion of coal in CFPP, in conjunction with increasing control technologies, results in relatively low SO2 and high NOX emissions. On the other hand, the high sulfur content in the ore during smelting results to relatively high SO2 emissions from large-scale smelters. For Shanghai in 2006, Zhang et al. (2009) reported anthropogenic NOX and SO2 emissions in the year 2006 of 631 and 618 Gg/year, respectively (ratio of 1.02). This is a shift from SO2 to NOX dominant source in Shanghai reported by an early study by Streets and Waldhoff, (2000) for the year 1995. Our data show that the NOX mass loading during the background period of this study is a factor of 11.6 higher than the SO2 mass loading. This factor is calculated using a Monte Carlo approach to account for the large variability in the mass loading (Table 1). In particular, a large sample of NOX and SO2 mass loading was drawn from a multivariate lognormal distribution using mean and standard deviation shown in Table 1 and correlation factors shown in Table 2. The high NOX suggests the dominance of combustion from the transportation sector as a local source (background) of NOX observed at the Pudong site. On the other hand, the ratio between the enhancement (relative to the background) in NOX and SO2 mass loading within the major plume is 1.4 ± 0.1 based on a similar Monte Carlo simulation. This ratio suggests a relatively NOX-rich pollution (on top of the local source from transportation) during the major plume event. This pollution is further characterized by a significantly high correlation between TGM and NOX (R=0.8) relative to the background (R=0.3). Because transportation is a minor contributor to TGM during the major plume event, our data indicates that NOX-dominant sources like CFPPs contribute largely to the observed enhancements in TGM relative to SO2-dominant pollution from smelters. The larger contribution of CFPP to observed TGM is supported by Streets et al. (2005) for Shanghai in September."

3). On Section 3.4. We have added new China data from X. Feng. (Feng, X.: Interactive comment on "Worldwide atmospheric mercury measurements: a review and

synthesis of spatial and temporal trends" by F. Sprovieri et al., Atmos. Chem. Phys. Discuss., 10, C81–C82, 2010).

We added in Section 3.4 the following sentence:

"Additional measurements for China can be found in Feng (2010) and Sprovieri et al. (2010)."

4). On Table 3. Thank you. We corrected the sampling periods.

5). On Figure 1. Thank you. We now have a scale in the Figure.

6). On Figure 2D. We modified its description in Section 3.1 as follows:

"As seen in Figure 2 (and in succeeding sections of this paper), the variability of TGM is strongly correlated with the wind direction."

Overall Response: The full MS has been edited and the above described changes incorporated into the final MS. Additions to the acknowledgments were made.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 30279, 2010.