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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Dry deposition fluxes and deposition velocities of seven trace metal species at five sites in Central Taiwan – a summary of surrogate surface measurements and a comparison with model estimation" *by* L. Zhang et al.

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We appreciate all the comments, which will help our future studies and to some extent the present study. Our responses are detailed below.

RC- Reviewer's Comments; AC – Authors' Comments

RC: In this study, the authors measured the dry deposition fluxes and deposition veloc-

ities of seven trace metals at five sites in central Taiwan. They reported that seasonal variations in deposition fluxes are mostly determined by those of air concentrations and to a less extent by those of deposition velocities. Moreover, it was found that the measured dry deposition fluxes can be reproduced reasonably well using the size-segregated particle dry deposition model if the mass fractions of trace metals in PM2.5, PM2.5-10 and PM10+ were known. I think that this is an important contribution to the dry deposition of trace metals, because such data is scarce.

AC: We are happy to know that the reviewer thinks that the study is an important contribution. We would like to point out that the study is important not simply because the data on this topic is scarce. There are actually some (although very limited) measurement data of metal dry deposition (mostly using surrogate surfaces) in literature. However, few studies quantified (or even qualitatively assessed) the uncertainties in this type of measurements and no study has been attributed to identify the most appropriate way of modeling the metal dry deposition. The preset study is designed to fill these gaps; that is to explore the potential uncertainties in measurements and modeling of metal dry deposition and to propose a realistic approach for future measurements and modeling studies.

RC: In this study, however, there are major problems, as described below. Hence, I cannot recommend the publication of this paper.

AC: We suspect that this recommendation was caused by the misreading of the focus of the paper. We hope the reviewer would think differently after reading our detailed explanations below.

RC: 1. The authors used a HNO3 digestion to measure the concentrations of trace metals in atmospheric PM and dry deposition samples. However, the bulk of crustal elements, such as Fe and Al, are not dissolved in HNO3. This may be one reason why the dry deposition velocity of Fe is smaller than those of other metals. It is generally expected that Fe in PM has much higher dry deposition velocity, because Fe is contained

primarily in soil particles with larger particle sizes.

AC: As mentioned briefly in the Supporting Document, all metals were analyzed using HCl combined with HNO3 (not just HNO3), a common method used in previous studies (e.g., Hieu and Lee, 2010; Fang et al., 2004, 2007). Using the same method, we also observed high Vd for Fe at several different location in Taiwan (Fang et al., 2004; 2007). The small Vd observed in this study can be caused by various reasons. Measurement uncertainty could be one reason. It is also possible that at this site Fe was mostly in PM2.5-10, still in coarse particles but with relatively low Vd, as supported by the modeling results presented in this study.

We admit that we do not have a good knowledge on Fe size distribution at the sites presented in this study and this can be improved in our future measurements. In fact, we have recently started measuring the size distributing of major metal species at these sites, as a result of the findings from the present study. We hope the publication of the paper can also help fellow researchers to improve their measurement designs.

RC: 2. The sources of trace metals at each site are unclear. The authors should evaluate their sources using several methods, such as principal component analysis, back trajectory analysis and lead isotope analysis. Also, the authors should discuss the contribution of long range transport of air pollutants from the China continent along with the contribution of domestic sources.

AC: We appreciate all the suggestions which provide additional research topics using our available data. In fact, substantial efforts have been ongoing on mercury species related to source-receptor study in our two search groups (Prof. Fang in Taiwan and Dr. Zhang in Canada) (e.g., Cheng et al., 2011; Huang et al., 2012). Source-receptor study on metal species at these sites is also planned in the near future. We like to point out that source-receptor study results will be presented in separate papers and are out of the scope of this paper.

As mentioned above, this paper focuses on dry deposition, not on emission sources.

More specifically, the paper focuses on exploring the potential uncertainties in measured dry deposition and identify the possibilities of modeling the dry deposition of these metals. Results generated from this study will be very useful to the scientific community of designing their measurement studies, as well as of choosing proper approaches to modeling the dry deposition of these metal species in chemical transport models.

RC: 3. The dry deposition flux is defined as the product of the atmospheric concentration and the deposition velocity for each constituent. Hence, it varies depending on these two parameters. The authors investigated the correlation between the measured dry deposition fluxes and atmospheric PM concentrations (shown in Fig.3). They concluded that the lack of correlation for several metals at several sites is due to large uncertainties in flux measurements. However, does not the lack of correlation suggest that the dry deposition flux is dependent on the dry deposition velocity rather than on the atmospheric PM concentration? It is well known that the dry deposition velocities are primarily dependent on the deposition of larger particles in PM. I think that the particle size distributions of trace metals in the atmosphere are likely the most important factor controlling their dry deposition fluxes. Further research, including intensive dry deposition and particle size distribution measurements, is required to better understand the relationship between the dry deposition fluxes and the particle size distribution for each metal.

AC: We totally agree with this comment. The present study actually confirms most of the above statements provide by the reviewer, e.g., the importance of size distribution, the dominance of largest PM (PM>10) on total dry deposition. As mentioned in our replies above, measurement of size distribution together with dry deposition has been started at these locations (again due to the new findings from the present study).

Regarding the correlation: flux (F) depends on both concentration (C) and deposition velocity (Vd). That is why, theoretically, a good correlation should be found simultaneously between F and C and between F and Vd. Of course, under some extremely

conditions, the correlations can be weak, e.g., if every low C corresponds to a large Vd (and vice versa). Such extreme conditions are unlikely in real world. Thus, a weak correlation between F and C would most probably be caused by a combination of large measurement uncertainties and large variation in particle size distributions. Sicne the measured Vd varied in a small range (implying the small change in particle size distribution), the very low correlation between F and C was most probably caused by measurement uncertainties. The above rationale is now added in the revised paper.

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 32847, 2011.