A letter of Response to Anonymous Referee #2

Manuscript Number: acp-2011-907
Manuscript Title: Development and uncertainty analysis of a high-resolution NH_3 emissions inventory and its implications with precipitation over the Pearl River Delta region, China

Referee comments

General Comments:

This work presents 2006 NH_3 emission inventory for PRD region and then compared the NH_3 emission trends with precipitation chemistry. Ammonia is one of the key precursors of secondary aerosols and this work is within the scope of Atmospheric Chemistry and Physics. I would recommend publication after following major comments were addressed.

Response: We thank the referee for favorable considerations of our manuscript. We have made major revisions as suggested by the referee, and details are described in our responses to the Specific Comments below.

Referee comments

Specific Comments:

(1) The relationship between NH_3 emissions and precipitation chemistry needs further examination. The correlations presented in Fig. 11-14 are plausible but may not pass statistical significance examination. I suggest the authors dig more on their data and consider further validation of their inventory, e.g., TES ammonia observations, etc.

Response: Thanks for the valuable suggestion! The aim of the comparisons of annual variations was to diagnose if there were possible correlations between NH_3 emission and precipitation NH_4^+. It is true that the relationship between NH_3 emission and NH_4^+ in precipitation was weak in this case (Fig. 11) due to complex atmospheric chemistry process and uncertainty in emissions and precipitation. We totally agreed with the referee's suggestion about the further validation of our inventory by using ammonia observations, and in fact this is the best way, ideally. Although there were few ammonia observations in the Pearl River Delta (PRD) region, in the revised version, we did some discussions based upon other studies (Shen et al., 2011; Meng et al., 2010) about the surface measurements of NH_3 in the gas phase, and have revised the manuscript by referring to the results (See Section 3.3 in the revised version). We also suggest that that the local measured NH_3 should be conducted in the PRD region (See Lines 16-18 on Page 25 in the revised version).

Meanwhile, combining with the other referee's suggestion we have removed Fig. 11, 12 in the previous version. Details can be found in the new Section 3.6.1 in the

revised version for our response to this comment.

Reference:

Meng, Z. Y., Xu, X. B., Wang, T., Zhang, X. Y., Yu, X. L, Wang, S. F., Lin, W. L., Chen, Y. Z., Jiang, Y. A., and An, X. Q.: Ambient sulfur dioxide, nitrogen dioxide, and ammonia at ten background and rural sites in china during 2007-2008, Atmos. Environ., 44, 2625-2631, 2010.

Shen, J. L., Liu, X. J., Zhang, Y., Fangmeier, A., Goulding, K., and Zhang, F. S.: Atmospheric ammonia and particulate ammonium from agricultural sources in the north China plain, Atmos. Environ., 45, 5033-5041, 2011.

(2) I couldn't understand how monthly variations of emissions were developed. It should be well documented in Sect. 2 as seasonality is one of top interests on NH_3 emissions.

Response: Thanks for comments. In order to improve the performance of air quality models, a timely updated emission inventory with temporal and spatial variations is crucial (Skjøth et al., 2011; Zheng et al., 2009). We conducted an initial investigation of temporal characteristics of major ammonia emission sources as much as possible. However, seasonal characteristics in agricultural source emissions depend strongly on both local human farming activities and climate conditions (Pinder et al., 2006). Nevertheless, it must be pointed out that currently, under the circumstances of lacking detailed agricultural registers and practices, reliable input parameters in the function of process driven description, and field NH_3 measurement data in the air quality monitoring network widely existed in China, sound temporal profiles are hard to develop. In this study, temporal profiles mainly referred to other similar studies (Streets et al., 2003; Chinkin et al., 2003; Meng et al., 2010), considering the seasonal characteristics of agriculture activities and meteorological conditions in the PRD region.

In the revised revision, we clarify how monthly variations in the agricultural sources were characterized (See the added Section 2.5 and Section 3.3 in the revised version) and recommended future research by considering the other referee's comment and the short comment in the Interactive Discussion (See the revision Lines 10-18 on Page 25).

Reference:

Chinkin, L. R., Ryan, P. A., and Coe, D. L.: Recommended improvements to the CMU ammonia emission inventory model for use by LADCO, U.S. Lake Michigan Air Directors Consortium, 2003.

Pinder, R. W., Adams, P.J., Pandis, S. N., and Gilliland, A. B.: Temporally resolved ammonia emission inventories: Current estimates, evaluation tools, and measurement needs, J. Geophys. Res., 111, D16310, doi:10.1029/2005JD006603, 2006.

Skjøth, C. A., Geels, C., Berge, H., Gyldenkærne, S., Fagerli, H., Ellermann, T., Frohn, L. M., Christensen, J., Hansen, K. M., Hansen, K., and Hertel, O.: Spatial and temporal variations in

ammonia emissions - a freely accessible model code for Europe: Atmos. Chem. Phys., 11, 5221-5236., 2011.

Streets, D. G., Bond, T. C., Carmichael, G. R., Fernandes, S. D., Fu, D., Klimont, Z., Nelson, S.M., Tsai, N. Y., Wang, M. Q., Woo, J. H., and Yarber, K. F.: An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, 108, D21, 8809, doi:10.1029/2002JD003093, 2003.

Zheng, J. Y., Zhang, L. J., Che, W. W., Zheng, Z. Y., and Yin S. S.: A Highly Resolved Temporal and Spatial Air Pollutant Emission Inventory for the Pearl River Delta Region, China and its Uncertainty Assessment, Atmos. Environ., 43, 5112-5122, 2009.

(3) Sect. 2.5 is too general to me. It only contains conceptional description. I would see more details in uncertainty analysis, e.g., CV and distribution of key parameters.

Response: Accepted! In this study, quantitative uncertainties were characterized with the aid of the uncertainty analysis tool of AuvToolPro. The AuvToolPro is an extension version of AuvTool developed by Dr. Zheng and Frey (2002). The AuvToolpro was developed by Dr. Zheng, the corresponding author of this manuscript. It is able to conduct quantitative variability and uncertainty analysis in model inputs and outputs for any user-defined models with the use of bootstrap simulation and Monte Carlo simulation, and to identify key sources leading to uncertainty in model outputs using sensitivity analysis approaches (Lau et al, 2010; Zheng et al., 2011).

The conceptual framework for quantifying uncertainty emissions inventory is shown in the Fig. 1 below (Zheng et al., 2011). In AuvToolPro, uncertainties in model input parameters were first quantified for each sub-category through the use of statistical methods, and then uncertainties in emissions for livestock, N fertilizer application, human being, biomass burning, waste treatment, fuel combustion estimates and other sources were calculated with the use of Monte Carlo simulation. For each emission source, quantification of uncertainties in emission estimates mainly involves two aspects: (1) quantification of uncertainties in model inputs and (2) propagation of uncertainties in model inputs through emission equation (Frey et al., 2002). The quantification of uncertainties in model parameters is typically done with statistical methods, such as bootstrap simulation or expert judgment, depending on data availability. Details can be found in the followed articles (Cullen and Frey, 1999; Frey et al., 2002; Frey and Zheng, 2002; NARSTO, 2005).

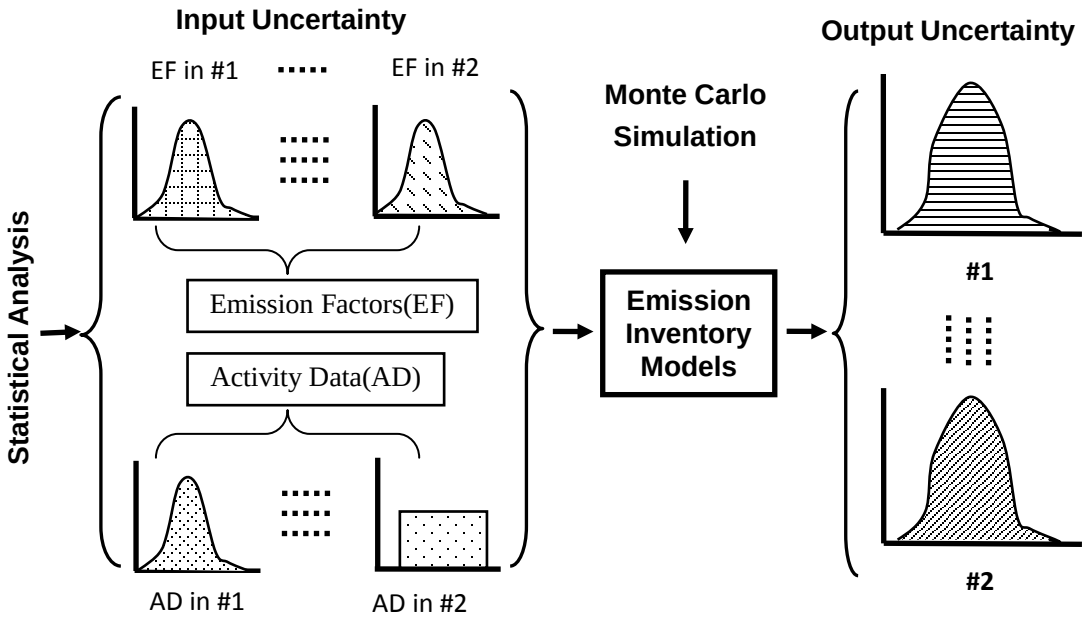

Fig. 1. The conceptual framework for quantifying uncertaity in emissions estimate

Note: EF: emission factor. AD: activity data

For response to the distribution of key parameters, since there was lot of information about uncertainty distributions used in this study, here we chose the emission models of biomass burning and livestock as examples. In the revised version, Table 7a summarized the probabilistic distributions representing uncertainties in emission factors of livestock. Table 7b summarized the probabilistic distributions representing uncertainties in emission factors, parameters used in the emission model for biomass burning. We hope that these substantial supplementary materials could be helpful for clarification.

Reference:

Cullen, A. C.and Frey, H. C.: The Use of Probabilistic Techniques in Exposure Assessment: a Handbook for Dealing with Variability and Uncertainty in Models and Inputs. Plenum, New York, 1999.

Frey, H. C. and Zheng, J. Y.: Quantification of variability and uncertainty in utility NOx emission inventories. Journal of the Air & Waste Management Association 52, 1083-1095, 2002.

Frey, H. C., Zheng, J. Y., Zhao, Y., Li, S., and Zhu, Y.: Technical Documentation of the AuvTool Software for Analysis of Variability and Uncertainty. Prepared by North Carolina State University for the Office of Research and Development. U.S. Environmental Protection Agency, Research Triangle Park, NC, 2002.

Lau A., Zheng J.Y., Lin B., et al.: Study on Analysis of Variability and Uncertainty for Hong Kong Air Pollutant Emission Inventories[R].Environmental Protection Department, HKSAR for Provision of Service, 2010.

NARSTO, 2005. Improving emission inventories for effective air quality management across north America: a NARSTO assessment. www.narsto.org.

Zheng J. Y., Wang S. S., Yu Y. F.: Development and demonstration of emission inventory uncertainty technology [R], Sub subject of the Chinese National 863 Key High-Tech Scientific Programs in the "Eleventh five-year Plan": Synthesized Prevention Techniques for Air Pollution Complex and Integrated Demonstration in Key City-Cluster Region, South China University of Technology, 2011(in Chinese).

Zheng, J.Y. and Frey, H.C.: AuvTool User's Guide, Prepared for Office of Research and Development U.S. Environmental Protection Agency Research Triangle Park, NC, 2002.

(4) Finally, I would draw authors' attention to the following in press paper, which may be relevant to their work.

Huang, X., Y. Song, M. Li, J. Li, Q. Huo, X. Cai, Z. Tong, and H. Zhang (2012), A high-resolution ammonia emission inventory in China, Global Biogeochem. Cycles, doi:10.1029/2011GB004161, in press

Response: We appreciated the recommended article from the reviewer. We did a careful review on this paper. We gratefully agreed that the study about the high-resolution ammonia emission inventory in China is very helpful and it improved the model-ready NH_3 emission inventory in China. We cited the paper, and introduced the important work in the revised version (See Lines 6 on Page 8 in the revised version).