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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Implementation of a Markov Chain Monte Carlo Method to inorganic aerosol modeling of observations from the MCMA-2003 Campaign. Part I: Model description and application to the La Merced Site" *by* F. M. San Martini et al.

**Anonymous Referee #1**

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Overall review

Inorganic aerosol equilibrium models have been applied to many ambient data sets. This manuscript introduces the Markov Chain Monte Carlo (MCMC) enhancement in such an application. The MCMC-ISORROPIA model seems to perform quite well for this dataset. The subject matter is quite interesting to the atmospheric community, and the authors did an adequate job explaining the model to an audience who may have



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Interactive Discussion
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both aerosol and advanced statistics background. A few suggestions are given below that may help highlight the contribution of the work and strengthen the presentation of the model to members of the atmospheric community who may be less comfortable with Monte Carlo methods. The paper is acceptable for publication in ACP.

Detailed comments

(1) Abstract/introduction. Clearly state the goal of the study. (a) Describe the research questions being asked. o Is this a model evaluation study to find out how well the MCMC-ISORROPIA model predicts ambient partition? o Is MCMC-ISORROPIA a tool that can be used to discriminate between measurements from different instruments for NH3? o MCMC can be used to predict gas-phase concentrations where they are not available. To support this statement, the application to predict HCl, for which measurements are not available, should be highlighted. Results for NH3 and HNO3 should be discussed for measurement periods where they are missing, if any. (b) Specify what "prior knowledge" is incorporated - do you mean the equilibrium relationships described within ISORROPIA? (c) What are the values added by using the MCMC-enhanced model vs. standard ISORROPIA? o How different are the "most likely concentrations" compared with the deterministic values predicted by ISORROPIA using nominal measured values (and assumed values in the case of Na and HCl)?

(2) Bayesian approach. (a) In Equation 2, are "Data" and "theta" scalars or vectors? It is not clear if the definition of theta is constant (e.g., Appendix A) throughout the paper. If so, please move the definition in Appendix A into the text. (b) State that the posterior in Equation 2 is the quantity of interest. (c) In this implementation of the Bayes' theorem, theta is assumed to be a Markov Chain. How good is that assumption? Here, theta is the set of concentrations corresponding to some simultaneous measurements, so there is no time element. How are the different random samples of theta related to a Markov Chain? Please explain any assumption used in the representation of theta and Markov Chains. (d) How is the initial guess defined in this work? How is theta0 related to theta? (e) Does the acceptance probability alpha have any physical meaning? Why is



ACPD
6, S2679–S2682, 2006





EGU

20% an optimal value for this quantity (Appendix A)? (f) Is MCMC-ISORROPIA applied for each measurement period (hourly)? How many samples are analyzed? What is the minimum set of available measurements needed for a sample to be analyzed? How many have missing HNO3, NH3, or AMS measurements?

(3) Prior (a) Is the same prior distributions applied for all time periods? (b) Should the prior be a function of meteorology, chemical regime (e.g., ammonium-rich vs. sulfate-rich)? Should the prior distribution of inorganic compounds be correlated? (c) How sensitive are the posterior estimates to the prior distributions?

(4) Likelihood (a) How are below-detection (or negative) observations treated? (b) The notations of [] and = (three bars) need to be defined. (c) For NH3, theta seems to be defined as a two-dimensional quantity consisting of the FTIR and TILDAS measurements. Is that correct? This definition is different from the definition in Appendix A, where theta is defined as a set of 9 continuous + 1 binary variables. (d) How should the reader interpret p(MTILDAS)+p(MFTIR) = 1 when MTILDAS and MFTIR are different? Is one of them right and the other wrong? Is one right some time and the other right some other time? What if they are both wrong? (e) In equation 19, is “Data” a scalar or a vector? What data are you referring to here? (f) Remove NOz from Figure 11 if it is not used in the MCMC analysis.

(5) Results (a) Please clarify the sentence “...the TILDAS observations are more consistent with the observations.” by specifying which observations the TILDAS observations are consistent with. (b) The TILDAS measurements are more likely correct than the FTIR observations, which are still within 95% confidence interval. Under what conditions would FTIR be more probable? Elaborate on the conditions on 26 April that correspond to FTIR being the more likely correct value than TILDAS. (c) A plot of the posterior median value +/- 33 percentile (equivalent to mean +/- standard deviation for normal distribution) against the measurements +/- error will be a useful tool to discriminate between measurements of NH3. (d) Present the results of MCMC-ISORROPIA vs. standard (deterministic) ISORROPIA to highlight the value added using the MCMC



ACPD
6, S2679–S2682, 2006





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method. Quantify the improvements of MCMC method over standard deterministic applications.

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6, S2679–S2682, 2006

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