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Bayesian Monte Carlo analysis applied to regional scale inverse emission modeling for reactive trace gases: from daytime to climatologic periods

L. Deguillaume (1), M. Beekmann (1), L. Menut (2) and I. Pison (2)

(1) Laboratoire InterUniversitaire des Systèmes Atmosphériques, UMR CNRS 7583, Universités Paris 7 et Paris 12, 94010, Créteil Cedex, France, (2) Institut Pierre Simon Laplace/Laboratoire de Météorologie Dynamique, Ecole Polytechnique 91128 Palaiseau Cedex, France (deguillaume@lisa.univ-paris12.fr)

Uncertainties in deterministic atmospheric chemistry models stems from uncertainties in both input parameters and in the model formulation itself. Several studies pointed out the uncertainty in the sensitivity of ozone buildup to VOC and NOx emissions (Hanna et al., 2001; Moore and Londergan, 2001).

Monte Carlo analysis allows evaluating model uncertainties related to input parameters and parameterization of a model. This method consists in performing a large number of successive simulations with the same model but with a distinct set of model input parameters at each time (VOCs and NOx emissions, meteorological parameters, actinic flux, photolysis rates, and rate constants). In the present work, an extension of this method so called “Bayesian Monte Carlo analysis” is used. It attributes a posteriori weight to individual Monte Carlo simulation by comparing them with observations. This method is therefore able to reduce the uncertainty in both model input and output parameters if the uncertainty propagating from model input parameters into model output is larger than that of the constraining observations. In this work, we apply this mathematical analysis to a regional-scale chemistry transport model, CHIMERE. The objective of the present work is to evaluate how observations can reduce uncertainties on NOx and VOCs emissions. The Île de France region has been chosen since it represents an ideal framework for such an analysis as concentrated anthropogenic emissions in the Paris region frequently lead to the formation of urban plumes. Measurements used for constraining our simulations result from the ESQUIF campaign performed during summer 1998 and 1999 and from the AIRPARIF air quality network sites. We present results from specific intensive observations periods during the ESQUIF campaign characterized by the formation of photochemical ozone buildup and we extend our study to climatologic periods over the summers 1998 and 1999 period. The results show that: (1) the regional emission cadastre in the Île de France region does not show any significant bias; (2) the uncertainty in a posterior emissions could be determined; at 1 sigma level, and for average summer values, it is 24% for NOx emissions, 32% for VOC emissions, and 37% for the VOC/NOx emission ratio; (3) this means that observations (ozone, NO) exert a strong constraint in particular on NOx emissions and the VOC/NOx emission ratio, their uncertainty is reduced by about 60% with respect to the fixed a priori values.

In conclusion, the Bayesian Monte Carlo framework is found to give very useful results in particular for the assessment of emission uncertainties.