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Top-down constraints to aerosol emissions from open biomass burning: the role of gas-particle partitioning and secondary organic aerosol formation

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Open biomass burning (BB), including wildfires and controlled burns in agriculture and foresty, is known to provide an important contribution to organic aerosol (OA) and black carbon (BC) emissions on the global scale. However, quantitative estimates of BB aerosol emissions and their effects on climate and environment remain rather uncertain. A useful way to constrain the OA&BC emissions involves using atmospheric measurements in the framework of the inverse modeling approach. In such an approach, the relationship between the emissions and the measurements is simulated by a chemistry transport model; this means that top-down estimates may be sensitive to possible model uncertainties.

As a result of assimilation of satellite measurements of aerosol optical depth, several recent studies (e.g. [1,2]) indicated that aerosol emissions provided by bottom-up emission inventories may be strongly underestimated relative to emissions of gaseous species (such as CO). Meanwhile, it was earlier shown (e.g. [3]) that the relationship between primary organic aerosol emissions and aerosol concentration in the atmosphere can be significantly affected by gas-particle partitioning and oxidation of lower-volatility organic emissions; these processes are usually not taken into account in typical chemistry transport models.

The main goal of this study was to examine to what degree the discrepancy between the OA&BC/CO emission ratios predicted by the bottom-up inventories and derived from satellite observations can be associated with the mentioned processes and explained in the framework of the volatility basis set approach (VBS) [3] to OA modelling. To achieve this goal, a VBS scheme, which was recently implemented in the CHIMERE chemistry transport model (CTM), was first modified to account for OA emissions from biomass burning. An ensemble of simulations with the CHIMERE CTM was then performed for the case of the 2010 mega-fire event in European Russia [4]; each of the simulations featured a unique set of the control parameter values, which were selected within the range of values reported in experimental studies of OA emissions. In addition, a model run was performed with a "standard" OA modelling scheme. The simulations were analysed in combination with data of both ground based and satellite observations of aerosol and CO in order to derive corresponding top-down BB emission estimates. These estimates together with their uncertainties were used to formally test a hypothesis that the discrepancy between the OA&BC/CO emission ratios given by bottom-up emission inventories (such as GFED3.1) and those derived from atmospheric measurements can be entirely explained by statistical uncertainties (in particular, those associated with the uncertainties in CO and OA emission factors). The results of the hypothesis testing are presented for each model configuration in terms of the confidence level, and the best configuration (featuring the highest confidence level that the above hypothesis is true) is selected. The obtained results confirm that gas-particle partitioning and oxidation of lower-volatility organic emissions can play a very important role in evolution of BB aerosol. Implications of the results of this study for validation of fire emission inventories and for inverse modelling of BB aerosol emissions are discussed.

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