Abstract

Top-down atmospheric inversions infer surface-atmosphere fluxes from spatially distributed observations of atmospheric compositions in order to quantify anthropogenic and natural emissions. In this study, we developed a Regional multi-Air Pollutant Assimilation System (RAPAS v1.0) based on the Weather Research and Forecasting/Community Multiscale Air Quality Modelling System (WRF/CMAQ) model, the three-dimensional variational (3DVAR) algorithm, and the ensemble square root filter (EnSRF) algorithm. This system can simultaneously assimilate hourly *in-situ* CO, SO_2, NO_2, $PM_{2.5}$ and PM_{10} observations to infer gridded emissions of CO, SO_2, NO_x, primary $PM_{2.5}$ ($PPM_{2.5}$), and coarse PM_{10} (PMC) on a regional scale. In each data assimilation window, we use a "two-step" scheme, in which the emissions are inferred first, and then input into the CMAQ model to simulate initial conditions (IC) of the next window. The posterior emissions are transferred to the next window as the prior emissions, and the original emission inventory is only used in the first window. Additionally, a "super-observation" approach is implemented to decrease the computational costs, observation error correlations, and influence of representative errors. Using this system, we estimated the emissions of CO, SO_2, NO_x, $PPM_{2.5}$, and PMC in December and July 2016 over China using nationwide surface observations. The results show that compared to the prior emissions (MEIC 2016), the posterior emissions of CO, SO_2, NO_x, $PPM_{2.5}$, and PMC in December 2016 increased by 129%, 20%, 5%, 95%, and 1045%, respectively, and the emission uncertainties decreased by 44%, 45%, 34%, 52%, and 56%, respectively. With the inverted emissions, the RMSE of simulated concentrations decreased by 40–56%. Sensitivity tests were conducted with different prior emissions, prior uncertainties, and observation errors. The results showed that the "two-step" scheme employed in RAPAS is robust in estimating emissions using nationwide surface observations over China. This study offers a useful tool for accurately quantifying multi-species anthropogenic emissions at large scales and in near real time.