



Top-down estimate of black carbon emissions for city cluster using ground observations: A case study in southern Jiangsu, China

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We combined chemistry transport model (CTM), multiple regression model and available ground observation, to derive top-down estimate of black carbon (BC) emissions and to reduce deviations between simulations and observations for southern Jiangsu city cluster, a typical developed and polluted region in eastern China. Scaled from a high-resolution emission inventory for 2012 based on changes in activity levels, BC emissions in Jiangsu were calculated at 27.0 Gg /yr for 2015 (JS-prior). The annual mean concentration of BC at Xianlin Campus of Nanjing University (NJU, a rural site) was simulated at 3.4 $\mu\text{g}/\text{m}^3$, 11% lower than the observed 3.8 $\mu\text{g}/\text{m}^3$. In contrast, it was 3.9 $\mu\text{g}/\text{m}^3$ at Jiangsu Provincial Academy of Environmental Science (PAES, an urban site), 56% higher than the observed 2.5 $\mu\text{g}/\text{m}^3$. The discrepancies at the two sites implied uncertainty of bottom-up estimate of BC emissions and their spatial allocation. Multiple regression to fit surface hourly BC observations at the two sites Constrained with the top-down method, BC emissions were constrained at 13.4 Gg/yr (JS-posterior), 50% smaller than the bottom-up estimate, and stronger seasonal variation were found. Biases between simulations and observations were reduced for most months at the two sites when JS-posterior was applied. In particular, at PAES, the simulated annual mean was elevated to 3.1 $\mu\text{g}/\text{m}^3$ and the annual normalized mean error (NME) decreased to 24.8%. However, application of JS-posterior slightly enhanced NMEs for certain months at NJU where simulated concentrations with JS-prior were lower than observation, implying that reduction in total emissions could not correct CTM underestimation. The effects on top-down estimation were further quantified for numbers, and spatial representativeness of observation sites. Best CTM performance was obtained when observation of both sites were used for emission constraining, with their difference in spatial functions considered. Given the limited BC concentration data in the area, therefore, more measurements with better spatiotemporal coverage were recommended for constraining BC emissions effectively. Moreover, top-down estimates derived from JS-prior and the Multi-resolution Emission Inventory for China (MEIC) were compared to test the sensitivity of the method to initial emission input. The differences in emission levels, spatial distributions and CTM performances were largely reduced after constraining, implying that the impacts of initial inventories were limited on top-down estimate.