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Impact of anthropogenic emissions from major population centers on global and regional aerosol budgets

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In urban areas, in particular in major population centers (MPCs), anthropogenic pollutants can dominate over natural emissions and cause severe air quality problems. We used emission annihilation scenario simulations in the atmospheric chemistry global circulation model EMAC to study the individual and cumulative impact of four major aerosol species from MPCs on the global and regional aerosol budgets. Black carbon, particulate organic matter, sulphur dioxide (SO_2), and nitrogen oxides (NO_x) were used to represent emissions of primary aerosols and of precursors gases for secondary aerosols sulphate and nitrate, respectively. Moreover, feedbacks resulting from changed emissions on other atmospheric constituents were assessed and the linearity in the burden changes due to the emission changes was discussed. Aerosol sulphate showed the strongest decrease in the global budget and also the most widespread changes in the tropospheric column density, whereas the smallest global decrease with only local changes was found for particulate organic matter. The maximum reduction was found around the emission sources and in downwind regions. The primary emitted aerosols showed almost no feedback on other species. In contrast, many gas-aerosol equilibria were affected when the SO_2 and NO_x emissions were reduced. In the case with the reduced MPC-NO_x emissions, many species participating in the NO_x–ozone (O_3) chemistry showed a response in their concentrations. In particular, ozone changed differently in extra-tropical and tropical cities, which is in accordance with findings of Butler and Lawrence (2009). Moreover, the oxidation capacity of the atmosphere was changed. The hydroxyl radical concentration changed similarly to O_3, which lead to an increase in the tropospheric carbon monoxide concentration and to locally greater SO_2 concentrations. Changes in the emissions for black carbon, particulate organic matter, and SO_2 resulted in almost linear responses of the corresponding atmospheric burdens. However, for NO_x and the aerosol nitrate this was only the case during boreal winter, but not during summer when the (ammonium-)nitrate burden depended not only on the NO_x emissions but also on the ambient temperature and the available sulphuric acid concentration.