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Origin of particulate matter and gaseous precursors in the Paris Megacity: Results from intensive campaigns, long term measurements and modelling

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Uncertainties on the origin of primary and secondary particulate matter and its gaseous precursors in megacities is still large and needs to be reduced. A detailed characterization of air quality in Paris (France), a megacity of more than 10 million inhabitants, during two one month intensive campaigns (MEGAPOLI) and from additional one year observations (PARTICULATE and FRANCIPOL), revealed that about 70% of the fine particulate matter (PM) at urban background is transported on average into the megacity from upwind regions. While advection of sulfate is well documented for other megacities, there was a surprisingly high contribution from long-range transport for both nitrate and organic aerosol. The data set of urban local and advected PM concentrations in the Paris area were used for a thorough evaluation of the CHIMERE model and revealed error compensation for the local and advected components of organic matter and nitrate. During spring time, CHIMERE simulations overestimate the sensitivity of ammonium nitrate peaks to NH3, because (i) they underestimate the urban background NH3 levels, probably due to neglecting enhanced NH3 emissions for larger temperatures, and because they overestimate HNO_3. However, from an ensemble of mobile Max-DOAS NO_2 column and airborne NO_y measurements around Paris, no clear sign on a NO_x emission bias in the TNO-Airparif data set was made evident.

The origin of organic PM was investigated by a comprehensive analysis of aerosol mass spectrometer (AMS), radiocarbon and tracer measurements during two intensive campaigns. Primary fossil fuel combustion emissions contributed less than 20% in winter and 40% in summer to carbonaceous fine PM, unexpectedly little for a megacity. Cooking activities and, during winter, residential wood burning are the major primary organic PM sources. This analysis suggests that the major part of secondary organic aerosol is of modern origin, i.e. from biogenic precursors and from wood burning. Implementation of different configurations of the volatility basis set into the CHIMERE model allowed correctly representing summertime organic aerosol (OA) peaks within the agglomeration and attributing them to biogenic secondary OA as a major source. OA build-up from anthropogenic precursors within the plume was also correctly simulated, but it was not possible to attribute it specifically to oxidation of aromatic or of semi/ intermediate volatile organic compounds. Plume build-up of PM significantly contributes to regional air quality around the Paris region.