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

<u>M. Beekmann</u><sup>1</sup>, H. Petetin<sup>1,10</sup>, Q.J. Zhang<sup>1,11</sup>, A. S. H. Prevot<sup>2</sup>, J. Sciare<sup>3</sup>, V. Gros<sup>3</sup>, V. Ghersi<sup>4</sup>, A.Rosso<sup>4</sup>, M. Crippa<sup>2</sup>, P.Zotter<sup>2,12</sup>, F. Freutel<sup>5</sup>, L. Poulain<sup>6</sup>, E. Freney, K. Sellegri, F. Drewnick<sup>5</sup>, A. Borbon<sup>1,7</sup>, H. A. C. Denier van der Gon<sup>8</sup>, A. Wiedensohler<sup>6</sup>, S. N Pandis<sup>9</sup>, and U. Baltensperger<sup>2</sup>

and the MEGAPOLI Paris, PARTICULES and FRANCIPOL - campaign teams

<sup>1</sup>LISA/IPSL,Créteil, France, <sup>2</sup>PSI, Villigen,Switzerland,<sup>34</sup>LSCE/IPSL,France. <sup>4</sup>AIRPARIF, Paris, France, <sup>5</sup>MPI-Chemistry, Mainz, Germany. <sup>6</sup>IfT, Leipzig, Germany, <sup>7</sup>LaMP, Clermont-Ferrand, France, <sup>8</sup>TNO, Utrecht, Netherlands, <sup>9</sup>FORTH, Patras, Greece, <sup>10</sup>now Laboratoire d'Aérologie, Toulouse; France, <sup>11</sup>now at Aria Technologies, Boulogne-Billancourt, France, <sup>12</sup>now at HSLU, Horw, Switzerland.

Keywords: secondary organic aerosol, carbonaceous aerosol, urban aerosol, source apportionment Presenting author email: matthias.beekmann@lisa.u-pec.fr

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 (PARTICULES and FRANCIPOL), and from satellite observations, 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 (Beekmann et al., 2015).

This data set of urban local and advected PM concentrations in the Paris area was 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 NH<sub>3</sub>, because (i) they underestimate the urban background NH<sub>3</sub> levels, probably due to neglecting enhanced NH<sub>3</sub> emissions for larger temperatures, and because they overestimate HNO<sub>3</sub> (Petetin et al., 2015). However, from an ensemble of mobile Max-DOAS NO<sub>2</sub> column and airborne NOy measurements around Paris, no clear sign on a NOx bias in the TNO-Airparif emission inventory 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 organic aerosol scheme into the CHIMERE air quality 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 within the Paris plume from anthropogenic precursors was also correctly simulated during summer, 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.

Finally, the comprehensive results on aerosol origin in the Paris Megacity are placed into a larger perspective of urban aerosol pollution in megacities worldwide. Even if, due to its geographical location, its post-industrial character, and past emission reduction measures, particulate matter levels in the Paris megacity are smaller than those in many other megacities, they still pose a challenge for air quality and health.

## References:

- Beekmann, M., et al.: In situ, satellite measurement and model evidence on the dominant regional contribution to fine particulate matter levels in the Paris megacity, Atmos. Chem. Phys., 15, 9577-9591, 2015.
- Petetin, H., Sciare, J., Bressi, M., Rosso, A., anchez, O., Sarda-Estève, R., Petit, J.-E., and Beekmann, M.: Assessing the ammonium nitrate formation regime in the Paris megacity and its representation in the CHIMERE model, Atmos. Chem. Phys. Discuss., 15, 23731-23794, doi:10.5194/acpd-15-23731-2015, 2015.
- Zhang, Q. J., Beekmann, M., Freney, E., Sellegri, K., Pichon, J. M., Schwarzenboeck, A., Colomb, A., Bourrianne, T., Michoud, V., and Borbon, A.: Formation of secondary organic aerosol in the Paris pollution plume and its impact on surrounding regions, Atmos. Chem. Phys., 15, 13973-13992, doi:10.5194/acp-15-13973-2015, 2015.