Determination of the volatility distribution of exhaust aerosol from thermodenuder data using a genetic optimization algorithm

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Keywords: exhaust aerosol, evaporation, volatility, aerosol modelling.
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Understanding the evolution of exhaust aerosols is key to model their impacts on environment. Fresh emission particulate matter contains both non-volatile and semi-volatile compounds, and it is well known that the partitioning between the phases may change as the aerosol dilutes and ages (e.g. Robinson *et al*, 2007; Saha *et al*, 2018). For modelling, representing the different volatilities of the aerosol with a volatility distribution has shown to be effective (Donahue *et al*, 2006).

Determining the volatility distribution is not straightforward: for example, several different volatility distributions can produce similar evaporation results (Riipinen *et al*, 2010; Pandis *et al*, 2013). Vaporization enthalpy and mass accommodation coefficient have significant effect on the simulation output. Recently, Tikkanen *et al* (2019) and Yli-Juuti *et al* (2017) presented an approach using genetic optimization algorithm to determine the volatility distribution from isothermal evaporation experiments. We apply a similar approach for thermodenuder experiments, in which a fresh aerosol evaporates at different temperatures.

Our approach to model aerosol evaporation is similar to Riipinen *et al* (2010). An evaporation curve for an aerosol with an initial evaporation distribution is obtained by solving the saturation concentration driven mass transfer between gas and particle phases.

To solve the initial volatility distribution from evaporation data, a genetic algorithm is used. As in Tikkanen *et al* (2019), we generate an initial population with random volatility distributions and vaporization enthalpies and solve their individual evaporation curves. Comparing the remaining mass after evaporation to a target function gives a fitness value. The model randomly chooses two individuals to produce a child, which inherits its properties from the parents with a possibility of mutation. A child with a better fitness value than the worst individual enters the population. This step repeats over the whole population and over several generations. The algorithm attempts to improve the fitness of the individuals while keeping the population varied to keep the algorithm from converging to a local rather than global minimum.

To test the model against experimental results, Electrical Low Pressure Impactor (ELPI) provided the initial number distribution of an exhaust aerosol while the target function came from mass concentration measurements with Aerosol Mass Spectrometer (AMS) before and after a thermodenuder. The remaining organic mass after evaporation in 150°C and 265 °C was 37% and 14% suggesting that large fraction of the organic mass is non-volatile. Our simulated results agree with this as shown in Figure 1. The two least volatile bins comprise nearly 50% of the initial organic aerosol.

We also compare our approach against other methods inferring volatility distribution from thermodenuder data. Earlier methods often assume same vaporization enthalpies for all the volatility bins or solve them separately. Our method solves volatility distribution and vaporization enthalpy simultaneously.

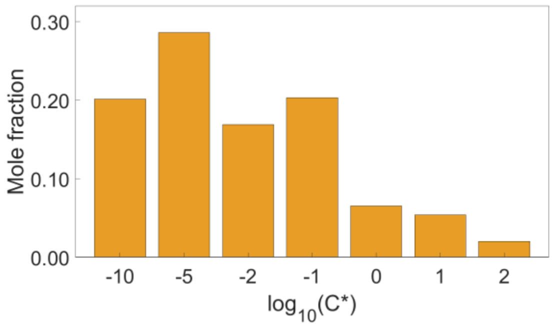

Figure 1. Modelled volatility distributions from exhaust aerosol measurements.

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