Wall losses during secondary organic aerosol formation and aging in an aluminium simulation chamber

H. Saathoff, C. Mohr, W. Huang, X. Shen, K.-H. Naumann

Institute for Meteorology and Climate Research, Karlsruhe Institute of Technology, Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany

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Presenting author email: harald.saathoff@kit.edu

Wall effects are always an important issue for the understanding of simulation chamber experiments. They strongly depend on the nature of the experiment, the type of wall surface and the size of the chamber. For the determination of yields or partitioning coefficients in secondary organic aerosol (SOA) experiments consequently wall effects have to be understood very well (Pierce et al., 2008; Saathoff et al., 2009; Matsunaga & Ziemann 2010; Zhang et al., 2014). Typically, the wall effects contribute more for lower concentration levels, and thus for experimental conditions more similar to the unpolluted ambient atmosphere.

Aerosol models coupling particle dynamics with aerosol chemistry and physics are indispensable tools for the analysis of simulation chamber experiments. In combination with dedicated measurements such models can be used to determine not only particle losses e.g. due to sedimentation or diffusion to the walls but also wall losses of gas molecules of different volatility. Aim of this work is to quantify wall losses in the 84.5 m^3 aluminium chamber AIDA (Aerosols Interactions and Dynamics in the Atmosphere) (Saathoff et al., 2009) by comprehensive analysis of long term SOA experiments with an aerosol dynamic model.

Experiments lasting up to 64 hours were done for this purpose since they have proven to be most sensitive to wall loss processes. The experiments started by ozonolysis of α-pinene leading to organic aerosol formation followed by subsequent aging by OH radical reactions. The evolutions of the particle chemical composition, mass, size, and number as well as dedicated trace gases were monitored by chemical ionisation mass spectrometry (CIMS), by aerosol mass spectrometry (HR-TOF-AMS), SMPS, CPC, and several trace gas monitors. After the experiments aluminium foils which had been exposed inside the chamber were analysed for deposited aerosol components by CIMS.

The experimentally observed data were analysed using the sectional aerosol behaviour code COSIMA (Naumann, 2003), supplemented by a SOA module (Saathoff et al., 2009) and using the master chemical mechanism (MCM 3.1) to describe the trace gas kinetics. The physical aerosol processes treated in COSIMA-SOA include particle diffusion to the walls and sedimentational deposition, coagulation, condensation and evaporation, wall losses of trace gases and dilution effects due to sampling.

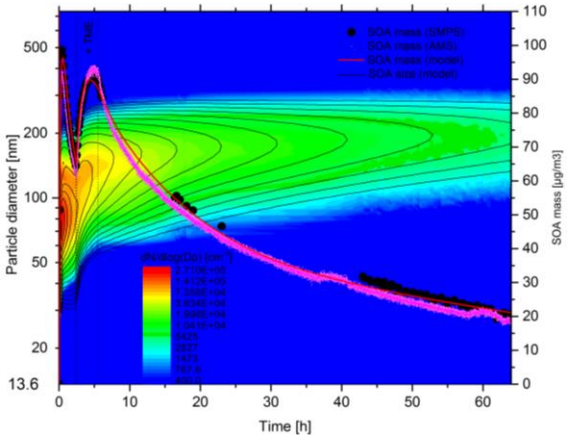

Figure 1. Evolution of measured (contour) and calculated (lines) aerosol particle size and mass during ozonolysis of 60 ppb of α-pinene and subsequent OH radical reactions at 294 K and 56 % RH in the AIDA simulation chamber.

COSIMA-SOA quantitatively reproduces the observed behaviour of trace gases and particles in AIDA (cf. Figure 1). Wall loss rate constants range between 10^{-3} and 10^{-4} s^{-1} for condensable organic trace gases and about 10^{-3} s^{-1} (nucleation mode) to 10^{-6} s^{-1} for SOA particles. Clearly, SOA yield measurements in the AIDA chamber have to be supplemented by detailed kinetic process modelling to account for losses – mainly from the gas phase - to the aluminium walls. Combining comprehensive measurement techniques and theoretical methods, such long term experiments provide a quantitatively reliable basis to evaluate AIDA SOA experiments even at lowest aerosol concentration levels.

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