Supplementary material for manuscript "Total sulphate vs. sulphuric acid monomer in nucleation studies: which represents the "true" concentration?" by Neitola et al.

Losses of sulphuric acid to the particle phase were calculated using the DMPS measured size distribution. The total volume of the distribution was calculated and the particles were assumed to be composed of pure sulphuric acid. This assumption is not accurate, as sulphuric acid is very hygroscopic and draws water vapour from the humidified carrier gas. Also, contaminants will condense on the particles. At the highest saturator temperature, the size distribution of the particles extends beyond the measurement range of the DMPS (>250 nm) which causes the estimation of losses of sulphuric acid to the particles to be underestimated. This was the case when the temperature of the saturator was above 300 K.

Picture S1 presents the calculated losses of sulphuric acid to the particles (open stars) calculated from two independent measurement sets as a function of saturator temperature for MARGA and CIMS measurements at RH of 30%. Also, gas phase monomer concentration measured by CIMS (red filled circles), the sum of gas phase monomer and losses to the particles (red triangles), total sulphate concentration measured by MARGA (blue triangles) and predicted sulphuric acid concentration by K&L (black solid line). The losses to the particle phase agree well between the two data sets (MARGA and CIMS) and overlap when the data sets have same saturator temperatures. The sum of gas phase monomer and the losses to the particle phase is one to two orders of magnitude lower than the prediction by K&L or the measured total sulphate measured with MARGA. This "missing" part of the sulphuric acid is distributed between wall losses in the system, which are larger for gas phase monomer than for total sulphate (see Fig. 4) and sulphuric acid clusters containing base molecules, to some extend "hidden" to mass spectrometers.
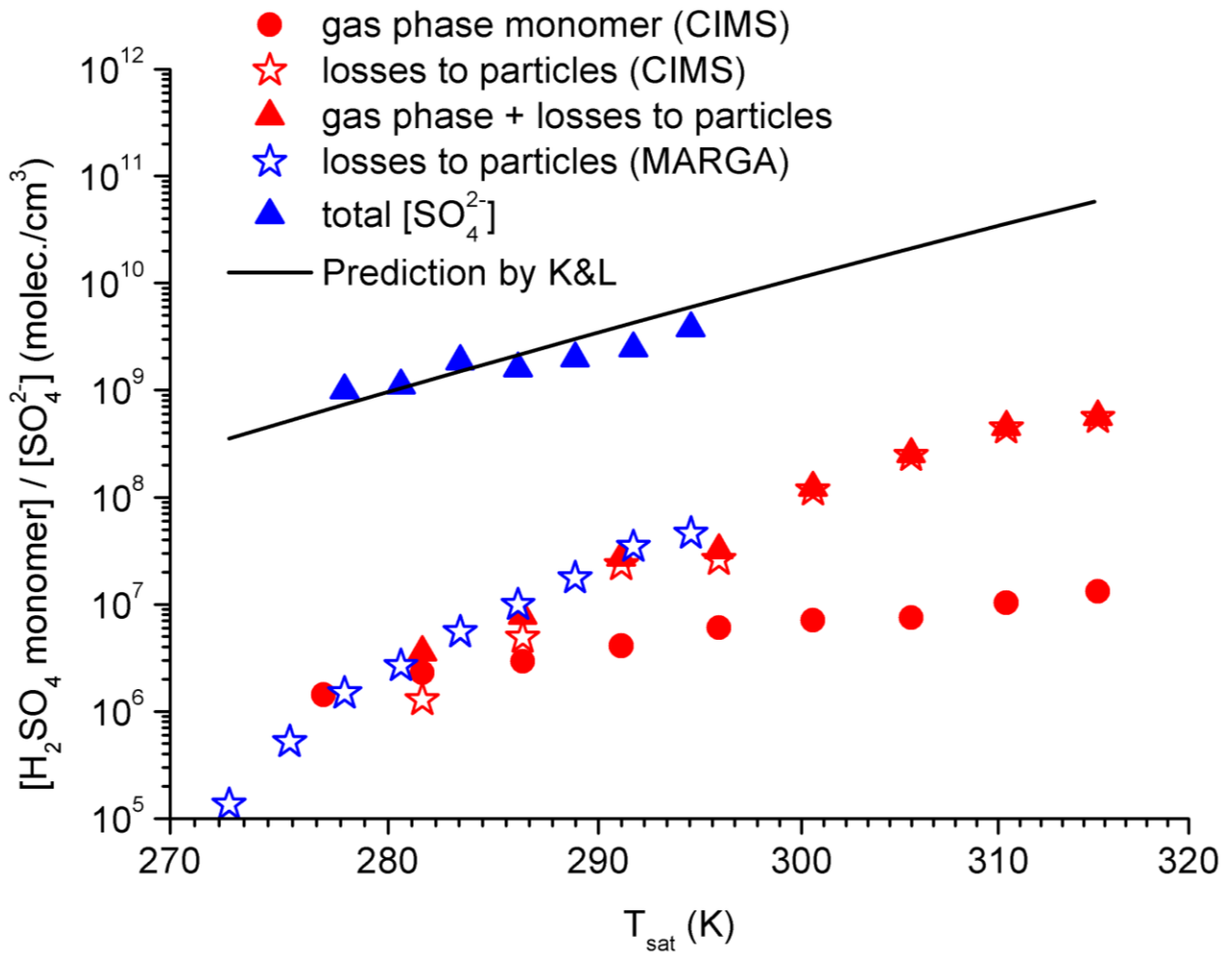
Figure S1. Calculated losses of sulphuric acid to the particles as a function of saturator temperature from two independent data sets for CIMS (open red stars) and MARGA (open blue stars) at RH of 30%. Presented are also gas phase monomer concentration measured with CIMS (filled red circles), sum of gas phase monomer and losses to the particles (red triangles), total sulphate measured with MARGA (blue triangles) and predicted sulphuric acid concentration by K&L (black solid line), all as a function of saturator temperature.