

## **Observation of a SO<sub>2</sub> Induced Nucleation Event in the Atmosphere**

**Bart Verheggen and Michael Mozurkewich**  
Centre for Atmospheric Chemistry, York University, Toronto, Canada

The atmospheric aerosol has a substantial effect on the global climate. Both the direct forcing (scattering by particles) and indirect forcing (reflectivity of clouds) are dependent on the size distribution of the aerosol. This means that particle formation and growth are important factors in determining the climatic effects of the aerosol. Both processes are not yet fully understood, which is shown by the large discrepancies found between conventional nucleation theory and measured nucleation rates.

In this work data are presented that show a rapid conversion of SO<sub>2</sub> to sulphate aerosol. The change over time of the measured size distribution is used to quantify the formation and growth of particles. The results are compared with the condensational growth rate calculated using a photochemical model. The growth rates were used to infer the time at which the particles formed.

The data analyzed were obtained as part of SONTOS (Southern ONTario Oxidant Study). This was an extensive measurement program undertaken in 1992 and 1993 at a rural site near Hastings, Ontario, Canada. The number density and size distribution of particles have been measured using a Condensation Nuclei Counter (cut-off diameter ~6 nm) and Differential Mobility Particle Sizer (DMPS, cut-off diameter ~10 nm). A range of gas species, radicals (as a group) and meteorological parameters were measured as well.

During this field experiment, an isolated burst of particles took place on August 25<sup>th</sup>, 1993. On this day, all measured species (O<sub>3</sub>, SO<sub>2</sub>, NO<sub>x</sub>, CO and hydrocarbons) were present at relatively low concentrations. At 9:45 a.m. the SO<sub>2</sub> concentration rose strongly, increasing from 1.5 to 5 ppbv in less than one hour. After a time delay of 15-30 minutes the small particles (6-20 nm diameter) increased in number, and grew to larger sizes in the following two hours. All other species remained at their initial low levels. The time delay shows that these particles were not emitted simultaneously with the SO<sub>2</sub> but implies that they were formed by homogeneous nucleation.

The SO<sub>2</sub> was probably emitted around midnight by the industries near Sudbury (ca 400 km northwest of Hastings), as indicated by the air mass back trajectories. The SO<sub>2</sub> plume traveled above the nocturnal inversion and was transported to the measurement site followed by fumigation downwards. The arrival of the SO<sub>2</sub> plume coincides with the breakup of the nocturnal inversion, as indicated by the change in wind speed and direction over time.

Assuming fumigation of an SO<sub>2</sub> plume as the history of the air mass, calculations are performed on the formation and growth of particles. Both dilution with upper level air and growth of particles cause the size distribution to shift. The effect of dilution is accounted for by using SO<sub>2</sub> as a tracer. We can then create a reconstruction of the growth of the newly formed particles. Once the SO<sub>2</sub> has been fully entrained, the SO<sub>2</sub> and H<sub>2</sub>SO<sub>4</sub> are spatially homogeneous. This allows us to deduce quantitative

information on the growth directly from the change of the size distribution over time.

A 1-dimensional model is used to calculate the  $\text{H}_2\text{SO}_4$  that is produced from the oxidation of  $\text{SO}_2$  by the OH radical, which under clear sky conditions is the major agent for oxidation. These calculated  $\text{H}_2\text{SO}_4$  concentrations are used to calculate the growth of newly formed particles, considering condensation on existing particles to be the main growth process. These calculations indicate that the particles could reach the detection limit of the DMPS (10 nm) in 30 minutes. This fast growth rate is confirmed by interpreting the change in size distribution over time.

The time of formation of the small particles is calculated using the growth rate. The rate of nucleation is deduced from the formation times, and compared with the calculated gas phase  $\text{H}_2\text{SO}_4$  concentration. This gives a new insight on the quantitative relation between  $\text{H}_2\text{SO}_4$  and nucleation rates.