A forward microphysical model to predict the size-distribution parameters of laboratory generated (mimic) stratospheric aerosols

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ABSTRACT

A Fast Aerosol Microphysical Model for the UTLS (FAMMUS) is applied to predict the size-distribution parameters of laboratory generated stratospheric aerosols. FAMMUS simulates the condensation and coagulation of binary H_2SO_4-H_2O particles using environmental and chamber thermodynamic data. The predicted final number concentration and size distribution parameters match the retrieved parameters within 10 % after optimising the initial conditions.

INTRODUCTION

The existence of polar stratospheric clouds (PSC) plays a fundamental role in chemical ozone loss during polar spring, over the Antarctic and the Arctic. The persistence and type of PSC particles dictate the extent and severity of ozone depletion. Thus understanding the formation and evolution of PSCs is key to predict any future trends in ozone depletion. Particle size distribution parameters are derived from extinction spectra of mimic stratospheric aerosols created in the Rutherford-Appleton Laboratory (RAL) aerosol cell. Here we attempt to numerically simulate the evolution of these particles using FAMMUS.

METHODOLOGY

Coagulation: Aerosol coagulation is important because it alters the composition and size distribution of stratospheric particles, primarily those smaller than one micron in diameter. Since most of the aerosol particles formed in the RAL aerosol cell (Bass, 2002) are of this size it is necessary to include coagulation within FAMMUS. Particles collide as a result of Brownian motion, differences in fall velocities, turbulent motions and inter-particle forces. The intermediate particles between two size bins are partitioned using fractions given by Jacobson (1999). The generalised Brownian coagulation kernel is taken from Jacobson (1999).

Condensation and evaporation: Apart from coagulation, condensation is another mechanism that affects the size distribution. The method we use to simulate condensation growth is taken from Jacobson (1999). While estimating the changes in aerosol particle size distribution due to condensational growth we have taken into account the Kelvin effect. We have approximated the particle density and surface tension by polynomial functions in weight percentage of sulphuric acid and temperature given by Steele and Hamill (1981). Aerosols are conditioned by passing them over a bath of sulphuric acid-the relevant water vapour pressure is calculated using the thermodynamic data provided by Zeleznik (1991). Numerical scheme used to couple the condensation and coagulation is discussed in Tripathi et al. (2002).

RESULTS AND DISCUSSION

Type of experiment		Initial Size distribution parameters			Final size distribution parameters		
		r_m, r (μm)	σ	N	r_m (μm)	σ	N (cm^{-3})
Numerical	Exp 1	0.1	0	$2x10^6$	0.9	1.20	$1.06x10^6$
	Exp 2	0.5	0	$4x10^6$	1.1	1.18	$.75x10^6$
	Exp3	0.5	0	$1.5x10^6$	.95	1.15	$.82x10^6$

Table 1 Summary of the different experiments performed

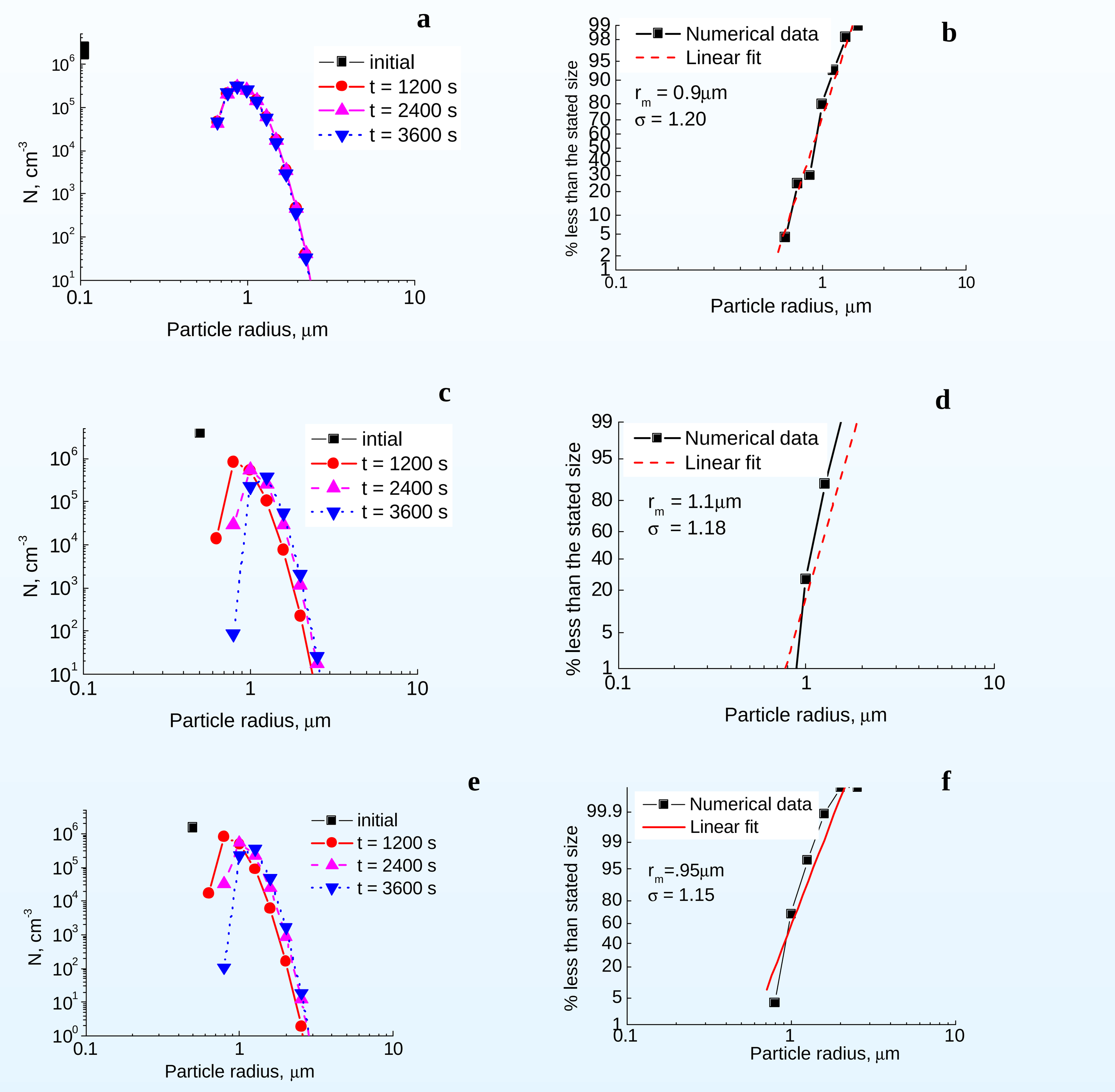

Fig. 1 (a) Evolution of cell particle size distribution undergoing condensational growth and coagulation when particles are cooled down from 295 K to 190 K. Initial size distribution parameters are those of numerical exp.1 in Table 1 (b) Cumulative percentage less than stated size vs particle radius for evolved distribution after one hour. The parameters for log normal size distribution are r_m *and* s.(c) Same as (a) except that the initial conditions are those of numerical experiment 2 in table 1 and droplets are cooled from 295 to 240 K. (d) same as (b) but for evolved distribution after one hour of fig (c). (e) Same as (c) except that the initial conditions are those of numerical experiment 3 in table 1.(f) same as (d) but for evolved distribution after 40 m of fig (e).

From figure 1 it can be seen that in the three experiments FAMMUS is successful in producing a narrow log-normal particle size distribution starting from a monodisperse size distribution. However, depending upon the initial size and number concentration of particles the final parameters of size-distribution vary. In the first experiment particles are cooled from 295 K to 190 K. Although the size distribution parameters of final distribution match closely the retrieved values, the water vapour concentration does not fall steadily showing the need for inclusion of nitric acid in the droplets. In the second experiment the mean size of the final distribution is slightly larger than the retrieved value. The particles are resident for half hour in aerosol cell. In the last experiment final parameters deduced evolving the distribution for 40 minutes results in the closest match with the retrieved values. The initial size distribution parameters chosen in these experiments are almost identical to those in similar calculations taking only thermodynamic considerations into account.

References

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