A-3.1.1 Laboratory Study on Processes of Generation and Extinction of Aerosol Particles

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Abstract Polar stratospheric clouds (PSCs) play important roles in the mechanism of ozone depletion. In order to clear the generation and extinction processes of PSCs, we developed an experimental apparatus and a method to generate the particles, which are the models of PSCs, and developed an in-situ measurement system of the particles in this subject.

The generator of model particles was designed based on the analysis of the condensation growth, coagulation growth and the deposition of particles, and was constructed. It is the laminar flow type system which consisted of a premixing room and a condenser tube. An numerical simulator to calculate simultaneously the changes of gases and particles in the generator by FDM was developed. As the results of the simulation, the experimental conditions were decided in which nitric acid vapor and water vapor would co-condense to solid particles. It was shown these conditions could be realized in the developed generator.

We also constructed an in-situ measurement system for the model particles by static/dynamic light scattering methods. We developed the ultra-high-frequency batch-type correlator system, which was used complementary with real-time correlator, in order to measure small and low-concentration particles. The fiber optical probe were also developed in order to enable measurements in the generator. It was shown that this system has enough performance to measure the model particles which would be generated.

As the results of this subject, it were realized that the condition in which the model particles of PSCs would be generated and that the equipment to measure the model particles.

Key Words Ozone layer, PSCs, Formation process, Particle generation, In-situ measurement

1. Introduction

It is known that the aerosols called polar stratospheric clouds play the important roles on ozone depletion. In order to estimate the ozone change, modeling of physical and chemical behaviors of aerosols is needed. By only observations in stratosphere, it does not have been got that the sufficient data of particles for quantitative valuation of parameters included in models. Recently, experimental approaches have been done in laboratories, but all of them have treated bulk or films. It might have effects on behaviors that differences between particles and bulk/films on the surface condition and others.

In this project, we designed and developed a generator and a method for making particles which are models of polar stratospheric clouds, and an in-situ measurement system for getting information about behaviors and states of particles in generation and extinction processes.

2. Development of a generator and a method for making model particles of PSCs

We planned to make both nitric acid trihydrate crystal particles and solid solution particles of ice and nitric acid in a laboratory. In the generator, 1) rapid growth by efficient condensation to particles must be occurred, 2) precise experimental conditions must be controlled, 3) the numerical analysis of phenomena should not be complex, and 4) the in-situ measurement should be possible. By comparison among generators of various types, the laminar flow type system termed a PSM consisted of a premixing room and a condenser tube was decided to use. In the premixing room, gases which include vapor, nitric acid vapor and nuclei respectively are mixed to make the supersaturations. Particles grow by the condensation in the condenser tube, of which the wall temperature is controlled.

In order to decide the outline of experiments, followings are important.

a) The growth of particles in the condenser tube: The condensation growth rate. The accommodation coefficient was treated as a parameter.

b) The deposition of gases and particles to the inside wall of the tube: The deposition ratio was evaluated on the assumption that the wall is perfect sink.

c) The effect of coagulation: The non-dimensional ratio the condensation growth rate to the coagulation growth rate was calculated.

Under various conditions (temperature, partial-pressure, total-pressure, flow rate, nuclei concentration, nuclei size and the size of the tube) where b) and c) were almost negligible, the particles size was estimated by integrating a) along stream lines.

The generator was designed based on results of this basic analysis, and constructed (Figure.1). It consisted of (1) a nuclei generator using electric furnace and DMA, (2) vaporizers using thermostat bath, (3) a gas premixer made of teflon, (4) a condenser tube made of glass, (5) a freezer which cool (3) and (4), and so on.

An numerical simulator to calculate simultaneously the changes of gases and particles in the generator by FDM was developed, in order to analyze the behaviors and states of particles more precisely than the above mentioned basic analysis. From the results of this simulation, the experimental conditions were determined in which nitric acid vapor and water vapor would co-condense to solid particles.

We tested the performance of each section of the generator. It was shown the conditions, in which the model particles of polar stratospheric clouds would grow, could be realized in a laboratory.

3. Development of advanced in-situ measurement system for particles

Aerosol particles are very sensitive to changes of ambient condition. Especially particles which are easy to evaporate like ice are difficult to be measure by usual measurement equipments which need sampling.

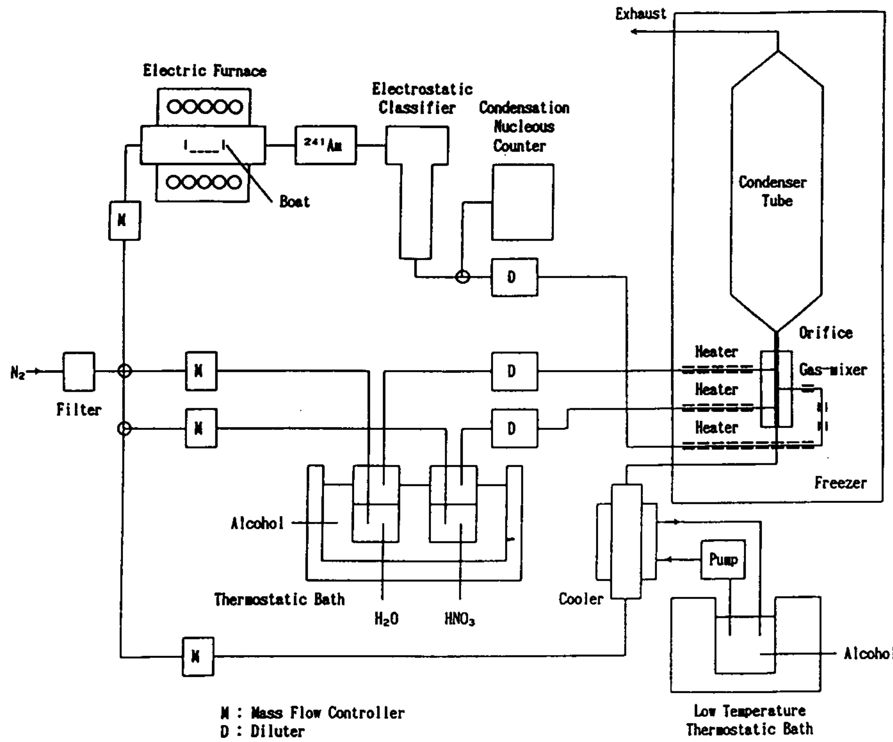

Figure.1 The experimental setup of particle generator

We had been studied an in-situ measurement system by photon correlation spectroscopy. The system had been examined by comparison with a measurement by DMA and CNC, and applied for the generation processes of NH_4Cl particles, which were from gas reaction $NH_3(g)$ + HCl(g), and Zn fumes, which were made by evaporation - condensation method. As it had been shown that the system was powerful to study the growth mechanisms, we decided to apply this system in this project.

The numerical analysis written in the preceding section showed the concentration of generated particles in this project would be much lower than it of the particles previously measured. It was suggested that some improvements of the system, as counterplans for the effect of fluctuation of concentration in measuring volume and for the weakness of the intensity of observed light, were necessary. Therefore, we designed and constructed the ultra-high-frequency batch-type correlator system using 200MHz logic analyzer, which was used complementary with 20MHz real-time correlator.

The probing units were also developed in order to apply the system in the generator in the freezer. These were consisted of polarization preserving optical fibers of which core diameter were 3 microns, polarization rotator, micro lenses and so on.

The improved measurement system was tested. Particles ranging in size from 0.02 to 10 micron could be measured by this system. This results shows that this system has enough performance to measure the model particles which would be generated.

4. Conclusion

It were realized that the condition in which the model particles of PSCs would be generated and that the equipment to measure the particles in growing process. Using the systems developed in this subject, we are going to try to clear micro physical processes of PSCs.