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DEHYDRATION MECHANISM IN THE ANTARCTIC STRATOSPHERE DURING WINTER

V. Ramaswamy

Atmospheric and Oceanic Sciences Program, Princeton University

Abstract. The growth of ice nuclei through deposition of water vapor at temperatures below frost point is investigated in the context of the Antarctic winter stratosphere. The altitude and the ambient water vapor mixing ratio, as well as the size of the nuclei determine the ice particle growth rate, with higher altitudes requiring colder temperatures for ice deposition. The magnitude of the temperature decrease below the frost point and its evolution over the winter determine the residence time of the growing ice particles and the loss of water vapor at any altitude. A winter-long simulation, using the observed South Pole daily temperatures, suggests that, in the limit of weak latitudinal mixing over the austral winter, considerable dehydration can occur within the polar vortex, with the higher altitudes (above 22 km.) experiencing the least losses.

Introduction

The Antarctic lower stratosphere undergoes a long period of radiative cooling over the course of the austral winter (Mahlman and Fels, 1986). As temperatures get progressively colder, condensation events begin to occur, leading to the formation of Polar Stratospheric Clouds, as inferred from satellite extinction measurements (McCormick and Trepte, 1987). Interpretations of remote sensing and in-situ measurements have led to the suggestion that particles of different types and sizes may be present in these clouds (Poole and McCormick, 1988; Rosen et al., 1988).

It has also been suggested (Douglass and Stanford, 1982) that the Antarctic lower stratosphere may represent a sink for water vapor during the austral winter through the freeze-drying mechanism. The sources of this vapor include the contributions from the tropical and midlatitude tropospheres as well as that due to the oxidation of methane. In this paper, the quantitative aspects of the microphysical processes that occur at the cold Antarctic stratospheric temperatures are investigated, using particle growth and terminal velocity equations, and a column model of the stratosphere. Specifically, we examine the deposition of water vapor onto a distribution of ice nuclei at temperatures below the frost point, and the effects due to the vapor-ice phase interactions.

Ice Phase Growth

The process of deposition (or sublimation) of vapor onto (or from) ice nuclei depends on the contact angle between the nuclei and the water film, roughness or irregularity of the nuclei and their chemical composition (Pruppacher and Klett, Chapter 9); all the parameters for different types of nuclei are not well-known. In addition, ice crystal habits can take on a variety of different forms such as trigonal plates, solid or hollow columns and polycrystalline shapes (Heymsfield, 1986); the relative proportion of each form for the duration of the polar stratospheric winter is difficult to quantify at present. In this paper, we assume that the contact angle is zero, that the growing particles are spherical and smooth, and that the chemical composition is favorable for deposition of water vapor as ice. The size distributions considered here are assumed to consist of ice particles while the temperatures of interest are those below the frost point of water.

The mass growth rate (dM_i/dt) due to diffusion of water vapor for the i^{th}-sized nuclei can be approximated as, for the sizes in the present study (Pruppacher and Klett, Chapter 13; Ramaswamy and Detwiler, 1986),

$$dM_i/dt = K\, R_i\, (S - 1 - A/R_i) \qquad (1)$$

where R_i is the radius of the nuclei; the parameter K involves particle characteristics (Pruppacher and Klett (1978, Chapter 13)), $S = e/e_s$ is the saturation ratio, with e the ambient vapor pressure and e_s the saturation vapor pressure with respect to ice; A is a constant that is independent of the size and nature of the nucleus. The term A/R_i is the Kelvin term and represents the effect of the curvature of the nuclei. The ambient vapor pressure depends on the water vapor mixing ratio and the temperature; the saturation vapor pressure depends only on the temperature via the Clausius-Clapeyron equation. As temperatures decrease to below the frost point of water, the flux of water vapor onto an initial ice nuclei distribution enables the particle mass to be composed of primarily water ice, irrespective of the initial composition. Further, owing to the large differences in the mixing ratios in the lower stratosphere, the flux of water vapor exceeds those of other species (e.g. nitric, hydrochloric and sulfuric acids) substantially once temperatures are below the frost point of water (Toon et al., 1986). Effects on growth rates due to radiational cooling are small and, hence, ignored.

The temperatures below which ice is deposited from the vapor phase onto a plane surface (critical temperature T_{crit}, or equivalently, the frost point) are shown in Figure 1 as a function of the water vapor mixing ratio at various altitudes for the sub-Arctic winter profile of McClatchey et al. (1972). For any mixing ratio, colder temperatures are required at higher altitudes for deposition to occur; NMC

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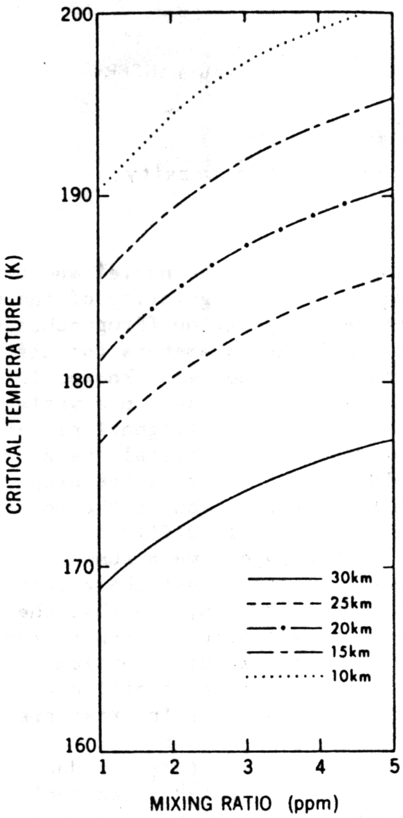

Fig. 1. Critical temperature (or frost point) for the deposition of water vapor as ice onto a plane surface as a function of water vapor mixing ratio (in ppm by mass) at various altitudes in a sub-Arctic winter atmospheric profile.

temperatures for South Pole (1980-1987) indicate that temperatures reach critical values (for 3 ppmm) often only below 25 km.

The existence of saturations greater than unity is not sufficient by itself for growth of particle sizes. From Equation (1), the condition for $dM_i/dt > 0$ for any size involves the Kelvin effect. Because of the inverse dependence on radius within the parenthesis in Equation (1), a higher saturation is required for the growth of smaller particles (< 0.05μ) than for the larger ones; from the Clausius-Clapeyron relation for a fixed vapor amount, this means that colder temperatures are required for the growth of the smaller sizes. Thus, unless temperatures become less than T_{crit}, vapor will not be deposited over all the particles in a nuclei distribution (e.g., sizes less than 0.05μ in a lognormal distribution with mode radius 0.08μ). Other factors being the same, a colder temperature would be required for deposition at non-zero contact angles (Pruppacher and Klett, 1978, Chapter 9).

The quantity ($S-1-A/R_i$) is the thermodynamic forcing that determines the growth/sublimation of particles of different sizes. For radii exceeding 0.1 micron, the Kelvin effect becomes negligible, and the forcing is determined by the saturation, which, in turn, depends on the temperature and water vapor. In the limit that the winter polar latitudes can be thought of as isolated regions with weak dynamical transport from the lower latitudes, there will be little supply of water vapor after winter has set in (especially within the polar vortex) and as the region cools radiatively. Under this assumption, the rate of change of temperature with time during the Antarctic winter (from, say, June 1) and subsidence constitute the direct thermodynamic forcing. Only the effects due to the former are considered in this study.

Model for Ice Particle Evolution

The growth of ice particles below the critical temperature, starting from an initial spectrum of ice nuclei is examined in the context of a column model that extends from 100mb (15km.) to 1 mb (50km.), with a resolution (1 km. upto an altitude of 25 km. and 5km. above) and profile corresponding to sub-Arctic winter (McClatchey et al. (1972)). Water vapor mixing ratio is assumed to be the same throughout the stratosphere at the beginning of the simulations.

Growth is assumed to occur on ice nuclei that are lognormally distributed initially. The initial concentrations, sizes and the vertical profile follow Hofmann and Rosen's (1982) data (mode radius of 0.08 micron, standard deviation of 1.6). Consistent with the objective of this paper ,species whose effects are small relative to that of water vapor at temperatures below T_{crit} but whose effects may be crucial at temperatures higher than T_{crit}, are ignored. Considerations of these species suggest that the initial size distribution can be different from that assumed here (Toon et al., 1986). Sensitivities with respect to different initial size distributions are stated later.

The initial particle size spectrum is discretized into 40 specific size categories (bins), with equal size separation. Particle concentration in any bin is invariant with time during the simulations discussed below. Thus, even though the size in any bin may change over time, the concentration at all times in that bin corresponds to the initial value. Initially, the first bin contains particles with radii a factor of ten less than the mode radius while the last bin contains particles with radii four times the mode radius. Cutoff of the lognormal distribution according to the above prescription was chosen as a trade-off between including all the relevant sizes and dictates of computational convenience. The results are unaffected by a finer discretization of the initial size spectrum. The initial binning of sizes, as described, is performed for each altitude level in the model. An optimal time step of 1 minute was selected.

Consider a temperature decrease below T_{crit} in any layer. After each time step, if deposition of vapor has occurred, the growth rate in each bin is calculated, using Eq. (1). The vapor that has condensed is distributed among the particles, based on the magnitude of the thermodynamic forcing function for each bin. The concentrations in each bin are always conserved so that all particles in any bin grow to the same size. Since different layers in the atmosphere can experience different magnitudes of forcing, it is only at the initial time period that a specific bin number at different altitudes will contain similarly sized particles; at other times, this need not be true.

After each time step, the total mass of vapor that was deposited as ice is depleted from the vapor amount present during the previous time step. This implies that the ambient vapor density (ρ) decreases according to

$$d\rho / dt = - \Sigma\, dM_i / dt \qquad (2)$$

Thus, an initial decrease in temperature below T_{crit} results in an increase in saturation ratio. This, in turn, results in deposition of vapor as ice, causing an increase in particulate size and mass, as well as causing a decrease in the vapor mixing ratio. The saturation ratio is reduced and the growth rate slows down or may be completely halted. On the other hand, if the temperature continues to decrease, the growth rate may continue unabated. If, after an initial decrease, the temperature increases, the forcing function assumes the opposite sign and, in this case, the particulate sizes and mass will decrease while the vapor mixing ratio will increase.

Particle fall velocity expressions (applicable to sizes less than 50 microns) follow Pruppacher and Klett (1978, Chapter 10). The vertical distance fallen by the size that is present in each bin and in each layer is evaluated at each time step. Thus, over each time step, there is a downward displacement of each bin, which is controlled by the bin size during that time. After some time, the height fallen by the bin may reach the next level. Then, a new size category (i.e. a new bin) is created for the layer below, with the size and concentration corresponding to those of the bin that fell from the layer above; simultaneously, this bin is deleted in the layer above. The above procedure is repeated whenever any bin crosses any level. At subsequent time steps, the sizes in the new bins become another component in the thermodynamic forcing for the layer below. The particles which have grown and fallen into the layer below, depending on the saturation ratio there, may undergo further growth (or may sublimate), thus increasing (or decreasing) the vapor mixing ratio in that layer.

The overall effect of this particular model framework is that sizes are continuously monitored in all the layers at all times and, at any instant, any specific layer in the atmosphere can contain sizes that have been there since the initial time period as well as those that have fallen from the layer/s above.

Tests with decreases of temperature below T_{crit} in any layer indicate that particle sizes grow rapidly (<4days) from the submicron sizes to sizes greater than 2-3μ. The greater velocities at the larger sizes accelerates the vertical fall of the particles; once the particles fall out of a layer, there is an irreversible loss of vapor from that layer. Experiments with larger initial mode radii (up to 2μ) and larger initial concentrations show that the amount of vapor depleted is similar for all cases, suggesting that the dominating factor in the dehydration process is the magnitude of the temperature decrease below the critical values.

Dehydration at South Pole

The column microphysical model is used to investigate the growth and sedimentation

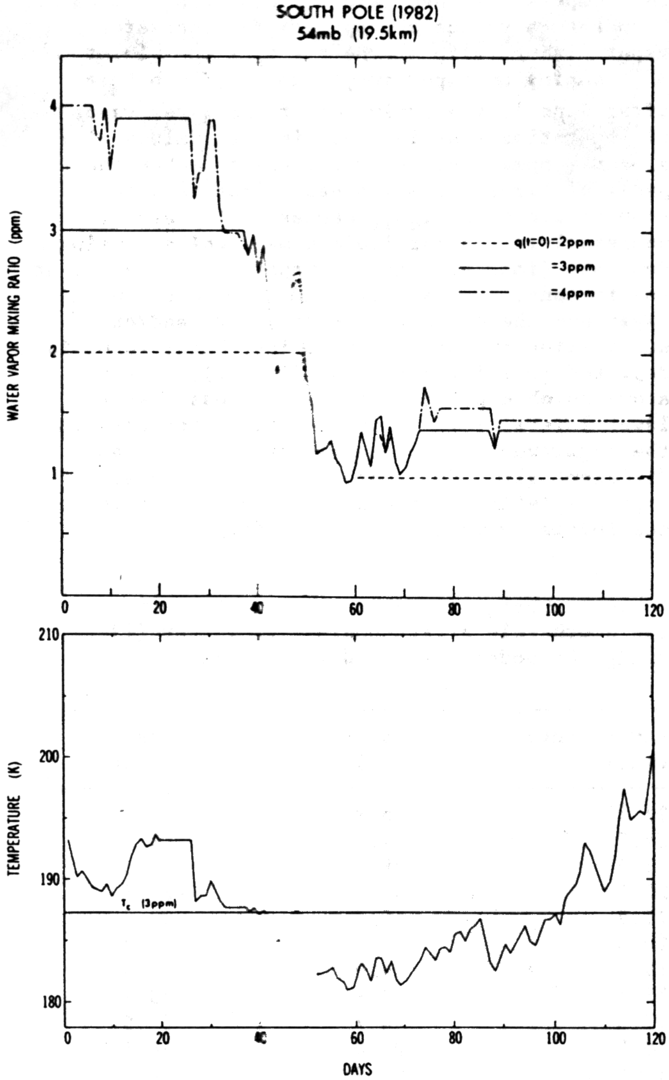

Fig. 2 (Bottom panel) Evolution of temperature in the layer centred at 54 mb over winter (June 1 to September 28, 1982). Solid line indicates the critical temperature corresponding to 3 ppmm. (Top panel) Evolution of water vapor mixing ratio as a consequence of the changes in temperature and ice microphysical processes. Different initial (at t=0) water vapor mixing ratios are assumed.

processes, using the NMC daily observations (Cayford and Hamilton, 1987) at South Pole from June 1 to September 28, 1982 (the data is interpolated onto the model grid). The 54 mb layer (~19.5 km.) temperature evolution is shown in the bottom panel of Figure 2. Note that if the supply of water vapor were such that a mixing ratio of 3ppmm is maintained, there would be several days when the temperature can fall to below the critical values at 54 mb. The frequency of occurrence of temperatures below the critical values is less in the layers above 54 mb. The temperature fluctuations (Figure 2) can be expected to render the growth and sedimentation processes a disorderly one, since particles in any bin can not only be retarded in their growth but can also be reduced in size. In the framework of the present model, ice particle growth and significant vapor deposition begin to occur when the temperatures decrease to below the critical temperature values appropriate for the water vapor mixing ratio at the start of the winter.

Assuming no vapor supply to the South Pole after June 1, the evolution of the water vapor mixing ratios starting from initial values of 2, 3, and 4 ppmm, respectively, are shown in the top panel of Figure 2. A decrease (increase) in temperature below T_{crit} causes an increase (decrease) in the particulate mass while causing a decrease (increase) in the vapor mixing ratio. At any instant, the particle and vapor mass in any layer are the consequences of growth and/or sublimation and/or sedimentation. Water vapor is depleted in all the layers (Table 1); layers above 54 mb experience a smaller loss than the lower layers. This is due to the differences in the magnitude of the temperature decrease at different altitudes over the austral winter. Vapor depletion in the 54 mb layer occurs for all the initial values chosen here (Figure 2).

Table 1. Change in water vapor content in the South Pole stratosphere over winter (1982), using the model described in the text.

Layer center (km.)	Pressure (mb)	Change (percent)
27.5	15.9	0
24.5	24.5	0
23.5	28.8	-21.6
22.5	34.8	-26.2
21.5	39.7	-39.0
20.5	46.5	-50.3
19.5	54.0	-54.6
18.5	63.8	-53.2
17.5	74.7	-61.5
16.5	87.0	-46.5
15.5	102.2	-58.3

The irreversibility of the dehydration mechanism through growth of ice particles and their subsequent settling holds true only if mixing is negligible into the column. The process would be inefficient if the vertical velocity in the ambient atmosphere transports the particles out of a preferred growth region in the stratosphere. The existence of favorable ice nuclei and temperatures below frost point constitute principal elements for the growth-cum-sedimentation and dehydration mechanisms; nonspherical ice crystal forms with different mass growth rates can also be expected to exhibit similar features. Finally, simulations with the South Pole temperature data from other years also exhibit dehydration of the stratosphere. The reductions in the lower stratospheric water vapor mixing ratios imply a depletion of the solube chemical species in the stratosphere over the austral winter and could, therefore, be a significant factor in the 'ozone hole' phenomenon.

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V. Ramaswamy, AOS Program, Princeton University, Princeton, NJ 08542.

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