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IMPACT OF STRATOSPHERIC AIRCRAFT ON CALCULATIONS OF NITRIC ACID TRIHYDRATE CLOUD SURFACE AREA DENSITIES USING NMC TEMPERATURES AND 2D MODEL CONSTITUENT DISTRIBUTIONS

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ABSTRACT

A parameterization of NAT (nitric acid trihydrate) clouds is developed for use in 2D models of the stratosphere. The parameterization uses model distributions of HNO_3 and H_2O to determine critical temperatures for NAT formation as a function of latitude and pressure. National Meteorological Center temperature fields are then used to determine monthly temperature frequency distributions, also as a function of latitude and pressure. The fractions of these distributions which fall below the critical temperatures for NAT formation are then used to determine the NAT cloud surface area density for each location in the model grid. By specifying heterogeneous reaction rates as functions of the surface area density, it is then possible to assess the effects of the NAT clouds on model constituent distributions. We also consider the increase in the NAT cloud formation in the presence of a fleet of stratospheric aircraft. The stratospheric aircraft NO_x and H_2O perturbations result in increased HNO_3 as well as H_2O. This increases the probability of NAT formation substantially, especially if it is assumed that the aircraft perturbations are confined to a corridor region.

1. INTRODUCTION

Two dimensional photochemical models of the stratosphere are important tools for the study of stratospheric dynamics and chemistry. They are also the major method for evaluating human influences on the stratosphere because they require less computer time to run and contain a more complete photochemistry than the more complicated three dimensional models. In particular, they are being used to assess the potential effects of a fleet of stratospheric aircraft on stratospheric ozone [e.g., Jackman *et al.*,1991]. It has been found that model predictions of aircraft-induced ozone depletion differ substantially, depending on whether or not the models include a parameterization of sulfate aerosol heterogeneous processes. Purely gas-phase models generally predict a depletion on the order of several percent. If stratospheric sulfate aerosol effects are added, the predicted depletions are much less, or actually result in a slight increase in stratospheric ozone [Weisenstein *et al.*, 1991].

Heterogeneous reactions are also catalyzed by NAT clouds in the stratosphere. NAT clouds are different from the sulfate aerosols in that the temperature at which they will form depends on the amount of HNO_3 and H_2O in the stratosphere, and are not a ubiquitous feature of the lower stratosphere. NAT clouds are essential components of polar stratospheric chemistry and the properties of such clouds have been studied extensively [e.g., Turco *et al.*, 1989].

Implementing NAT cloud distributions and the heterogeneous reactions they catalyze into zonally averaged models of the stratosphere is difficult. This is becuase zonally averaged temperatures are generally too high to allow for NAT clouds to form. If one denotes by $\langle c \rangle$ the zonally averaged cloud surface area density, and the zonally averaged temperature by $\langle T \rangle$, then $\langle c(T) \rangle \neq c(\langle T \rangle)$. To get around this difficulty while remaining consistent with the philosopy of a 2D model, we use temperature probability distributions characterizing a particular latitude and pressure level in the model. These distributions are obtained using National Meteorological Center temperature data.

NAT cloud formation depends on the concentrations of HNO_3 and H_2O in addition to the temperature. Because stratospheric aircraft inject both into the stratosphere, it is likely that a stratospheric aircraft fleet will increase the probability of NAT cloud formation. Peter *et al.*[1991] attempted to assess the magnitude of the effect at $70^\circ N$, and concluded that the probability of finding an NAT cloud at that latitude might be doubled with the introduction of such a fleet. This paper is in the same spirit as their work, but attempts to develop an annual NAT surface area density climatology for the latitudes and pressure levels of the GSFC 2D model. We do this both with and without the assumption of a stratospheric aircraft fleet in order to allow a 2D model assessment of the chemical effects of stratospheric aircraft on the atmosphere.

2. CRITICAL TEMPERATURES FOR NAT FORMATION

According to Hanson and Mauersberger, [1988], the relationship between the NAT saturation temperature, T_s, the partial pressure of HNO_3, P_{HNO_3}, and the partial pressure of H_2O, P_{H_2O}, is given by.

$$\log_{10}(P_{HNO3}) = m(T_s)\log_{10}(P_{H2O}) + b(T_s), \quad (1)$$

and

$$m(T_s) = -2.7836 - 0.00088T_s \quad (2)$$

$$b(T_s) = 38.9855 - \frac{11,397.0}{T_s} + 0.009179T_s. \quad (3)$$

The stated range of validity of this equation is for temperatures between 180 K and 200 K. In this study, we assume that the equation is valid up to 210 K, and that NAT clouds will form wherever the temperature is below T_s. This assumption may not be correct, however. Peter et al.[1991] have discussed the possibility that a substantial supersaturation on the order of 3 K is required to nucleate the NAT clouds. If this is the case, then using Eq. 1 will result in a prediction of the critical temperatures for NAT cloud condensation that is too high.

Given Eq. 1, and model distributions of HNO_3 and H_2O, a critical temperature distribution can be calculated as a function of latitude and pressure, as shown in Figure 1. In this case, a subsonic aircraft fleet has been assumed, but no stratospheric aircraft fleet. The addition of stratospheric aircraft increases the critical temperatures. The amount of increase depends on the assumptions made about the zonal distribution of HNO_3. For instance, the dynamics of the stratosphere may be such that aircraft corridors are formed inside of which the consituent concentrations are significantly greater than the zonal mean values. Tracer simulations using the GSFC 3D chemistry and transport model and European Center for Medium Range Weather Forcasting (ECMWF) wind fields suggest maximum concentrations of up to 5 times the zonal mean during the winter [Douglass et al., 1992]. If one considers all of the injected HNO_3 and H_2O from the stratospheric aircraft to be concentrated in a corridor with a width of 72° longitude, the local concentration increase inside the corridor will be 5 times the perturbation to the zonal mean, while outside the corridor the contribution from the aircraft fleet will be 0. Using this assumption, the increase in NAT formation temperatures can be up to 6 K, as shown in Figure 2.

3. TEMPERATURE PROBABILITY DISTRIBUTIONS

As mentioned above, zonally-averaged temperatures are generally too warm to dip below the NAT critical temperatures. If one considers the longitudinal distribution of temperatures at a particular latitude, however, it could be that a substantial fraction of the band is below the condensation temperature, and could support NAT cloud formation. To quantify this effect, we took a month's worth of NMC temperature data, and binned the temperatures into 10°-latitude bands. At each NMC pressure level, the temperatures in the band were used to produce a temperature probability density distribution for each month, $P_m(T, \theta_i, p_j)$, where $\theta_i = (-85, -75, ..., 85)$ are the latitude coordinates and $\{p_j\}$ are the NMC pressure levels. This distribution is normalized such that PdT gives the probability of finding a temperature between T and $T + dT$. For this paper, NMC temperatures for one year, from May, 1991 to April, 1992, were used. By consider-
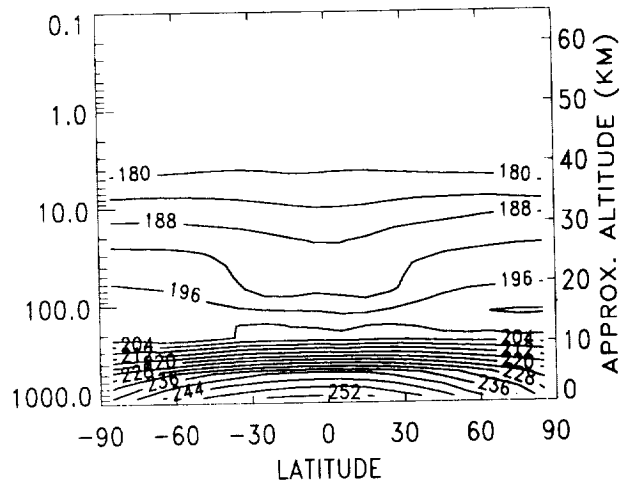
Fig. 1 Critical Temperatures for NAT formation: Assuming the relationship between saturation temperature and distributions of HNO_3 and H_2O given in Hanson and Mauersberger [1988], critical temperature distributions can be calculated. Shown is the January, 1992 distribution, where the HNO_3 and H_2O fields were taken from a model run including sulfate aerosol heterogenous processes and a subsonic aircraft fleet.
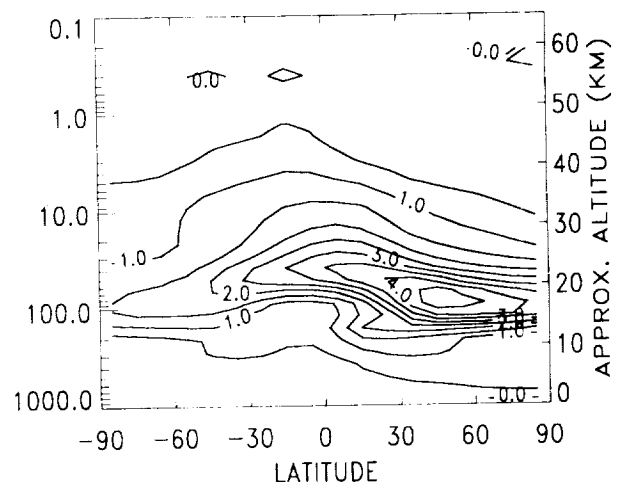
Fig. 2 Increase in critical temperatures from addition of a stratospheric aircraft fleet: The increases in stratospheric HNO_3 and H_2O from stratospheric aircraft can substantially increase the critical temperatures for NAT formation. This effect is on the order of 2 K if the aircraft perturbation is assumed to be distributed zonally. If the aircraft perturbation is assumed to be confined to a tight corridor, concentrations are higher and so is the temperature. The figure above was calculated assuming an aircraft corridor width of 72° longitude.
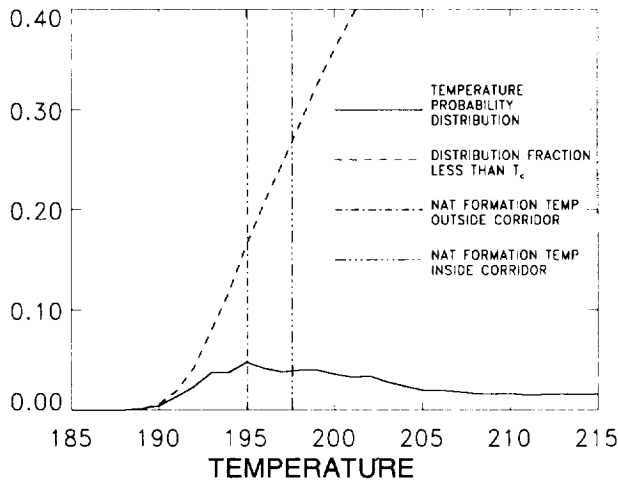
ing more than one year of NMC data, a better estimate of the climatological temperature distributions could be obtained.

With the critical temperatures calculated using the GSFC 2D model and the NMC temperature distributions, the probability that NAT clouds will be found can be calculated as a function of month, latitude, and pressure. Denoting this fraction by f,
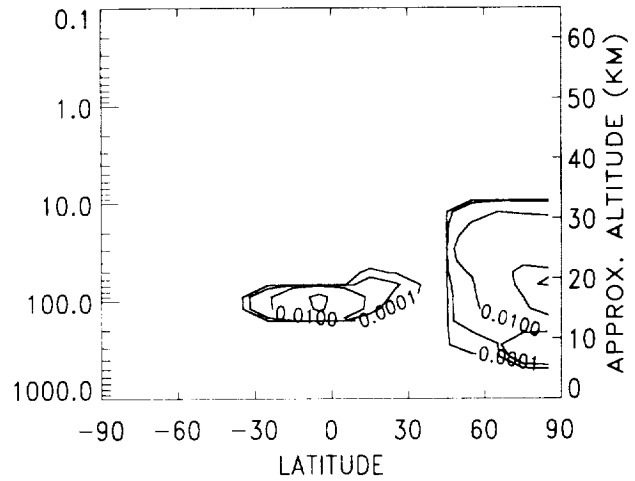
$$f_m(T_c, \theta_i, p_j) = \int_0^{T_c(\theta_i, p_j)} P_m(T, \theta_i, p_j) dT, \qquad (4)$$

where T_c is the condensation temperature for NAT clouds (which we assume is equal to the Hanson and Mauersberger saturation temperature, Eqs. 1-3, above). As shown in Figure 3, which depicts the calculation of f at 50 mb, 65° for January, 1992, f can be a rapidly increasing function of T_c. If T_c increases slightly due to the introduction of stratospheric aircraft, f can increase substantially.
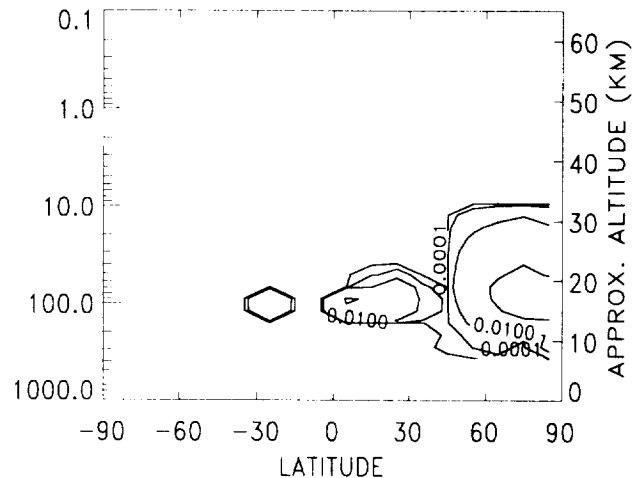
A plot of f for January 1992 is shown in Figure 4. In the case shown, no stratospheric aircraft fleet has been assumed. For this month, the peak probability that an NAT cloud will form occurs in the polar region, and has a magnitude of .2. From the figure, we might expect to see an NAT cloud forming at, say, 70° and 50 mb with a probability of about 0.1, or roughly three days out of the month. This appears to be in fair agreement with the results of the study of Peter et al.[1991], which also calculated such probabilities. The figure also indicates a substantial probability that NAT clouds will form in


Fig. 4 January 1992 NAT formation probabilities: No stratospheric aircraft are included in this calculation. The peak in the January formation probability occurs near the north pole, at approximately 70 mb. A significant tropical formation probability is also seen, which peaks a bit lower down at approximately 100 mb. Very small probabilities of NAT formation are found in the northern midlatitudes. Temperatures in the southern hemisphere are too warm for NAT clouds to exist there.

Fig. 3 Calculation of f from T_c and $P(T)$: The solid curve at the bottom of the figure is the temperature probability distribution at 65°N and 50 mb. the dashed line is the integral of this distribution up to the temperature T. The vertical dashed lines show the critical temperatures for NAT condensation inside and outside of an aircraft corridor with a longitudinal width of 72°. Because f can increase rapidly, $f(T_c)$ inside the corridor can be substantially larger than $f(T_c)$ outside of the corridor.

Fig. 5 Increase in January NAT formation probability from the addition of a stratospheric aircraft fleet. Shown is the increase assuming a 72° aircraft corridor, inside the corridor.

the tropics, peaking at about 100 mb, a little lower than the polar peak. The probability of cloud occurence is a minimum at northern midlatitudes, and is absent in the summer hemisphere, reflecting the warmer temperatures there.

When the NO_x and H_2O perturbations from a stratospheric aircraft fleet are included in the calculations, the resulting probabilities of NAT cloud occurrence are increased. The increase in probability of cloud formation
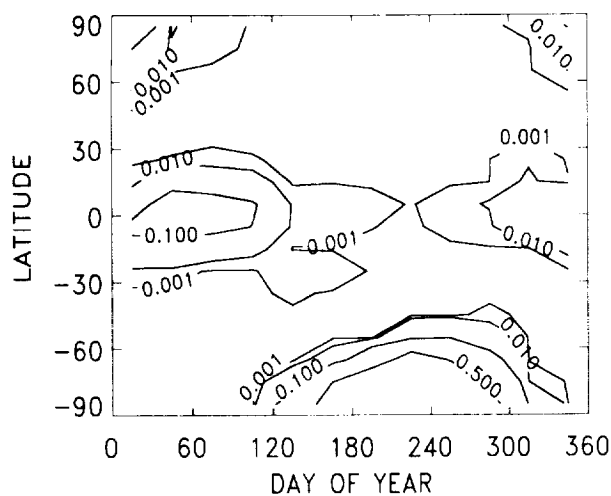
Fig. 6 Seasonal variation of NAT formation probability: Shown is the variation of f as a function of day and latitude, on the 90 mb surface. No stratospheric aircraft perturbation is included. The largest probabilities are in the south polar regions in the months of July, August, and September. Substantial probabilities of occurrence are also found in December/January in the northern high latitudes, and in the tropics in February/March. At 50 mb, no tropical cloud formation is predicted, but the north and south polar distributions are similar to those depicted here.

has somewhat the same morphology as f itself, and has a similar magnitude. If the perturbations are assumed to be confined to a corridor, the increases are larger than if the perturbations are assumed to be distributed zonally (Figure 5).

Distributions similar to Figure 4 can be obtained for each month. Figure 6 depicts the seasonal variation of NAT cloud occurrence probability on the 90 mb surface. The north polar peak in occurrence probability occurs in December/January. The south polar occurrence probability peaks in August, and is large in both July and September. Finally, the tropical probability peaks in February/March, and is at a minimum in October. If one moves to the 50 mb surface, one finds only very small probabilities of tropical cloud formation, while the northern and southern polar regions remain about the same.

If we assume that the NAT cloud that forms has a value of 10^{-8} cm^{-1}, as given by Turco et al., [1989], the distributions define a seasonally-varying NAT cloud surface area density distribution. (This assumption may introduce a substantial uncertainty into the calculation. It should be adequate for a 2D sensitivity study.) The surface area density distribution can then be introduced into the model, and used to calculate the effects of heterogeneous reactions catalyzed by the NAT clouds on constituent distributions. Since the NAT cloud freqency is increased with the addition of a stratospheric aircraft fleet, it appears that including NAT cloud processing in assessments of potential stratospheric aircraft effects on

the stratosphere may result in important changes. These changes remain to be assessed.

REFERENCES

Douglass, A. R., R. B. Rood, and C. H. Jackman, Three dimensional model calculations of the global dispersion of high speed aircraft exhaust and implications for stratospheric ozone loss, this volume, Proceedings of the Quadrennial Ozone Symposium, 1992

Hanson, D. and K. Mauersberger, Laboratory Studies of the Nitric Acid Trihydrate: Implications for the South Polar Stratosphere, Geophys. Res. Lett. 15, 855-858, 1988.

Jackman, C. H., A. R. Douglass, K. F. Brueske, and S. A. Klein, The Influence of Dynamics on Two-Dimensional Model Results: Simulations of ^{14}C and Stratospheric Aircraft NO$_x$ Injections, J. Geophys. Res. 96, 22559-22572, 1991.

Peter, Th., C. Bruhl, and P.J. Crutzen, Increase in the PSC-Formation Probability Caused by High-Flying Aircraft, Geophys. Res. Lett. 18, 1465-1468, 1991.

Turco, R. P., O. B. Toon, and P. Hamill, Heterogeneous Physicochemistry of the Polar Ozone Hole, J. Geophys. Res. 94, 16493-16510, 1989.

Weisenstein, D., M. K. W. Ko, J. M. Rodriguez, and N. D. Sze, Impact of heterogeneous chemistry on model-calculated ozone change due to HSCT aircraft Geophys. Res. Lett. 18, 1991-1994, 1991.

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