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ABSTRACT

A thermodynamic exclusion principle is used to determine those regions in the earth's upper atmosphere where the formation of clouds due to condensation or sublimation of water vapor is or is not possible. The probability of occurrence of such clouds is then determined from model atmospheres as a function of altitude for different latitudes and seasons. The theoretical results correspond well with actually observed locations, frequencies, and altitudes of noctilucent clouds. It is shown that statistical analysis of certain noctilucent cloud data should not only permit experimental tests of the theoretical study, but also provide, by inference, information about variations of the mesopause altitude with latitude, season, and solar activity.

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INTRODUCTION

In 1961, Hesstvedt(1) published a note on the nature of noctilucent clouds, where he made use of available information on the temperature structure of the upper atmosphere. Many additional data about atmospheric conditions near the mesopause level have been accumulated since. Although our understanding of the physical processes taking place there is still quite limited, there are a number of theoretical deductions which can be made with regard to *a priori* probabilities of cloud formation at high altitudes in the earth's atmosphere.

First, we shall postulate that noctilucent clouds consist of H_2O adsorbed to dust nuclei. We shall then investigate whether and where conditions in the upper atmosphere are such that H_2O cloud formation is possible, or at least probable. Finally, we shall compare the theoretically deduced conditions with actual cloud observations and make some predictions that can be checked by future experiments, thus shedding some light on the validity of the original postulate.

THEORETICAL BASIS

One of the requirements of our postulate, namely the presence of dust particles in the upper atmosphere that can act as condensation or sublimation nuclei and thus facilitate cloud formation, appears easily fulfilled. There is no need here to quote the extensive literature on the subject; refer, for example, to recent work by Whipple(2), Link(3), or Colombo and Fiocco(4).

The other requirement is the presence of sufficient water vapor to permit the formation of H_2O clouds. But we have no experimental data whatever on water vapor amounts above some 30 km altitude.

Let us therefore make use of an exclusion principle, based on simple thermodynamic laws and formulated in a recent study.(5) In brief, every model planetary atmosphere establishes unique altitude distributions for the saturation vapor pressures of its minor constituents. This is done solely by relating ambient atmospheric pressures and temperatures to altitude. But the partial pressure of a minor constituent can never be larger than the total ambient pressure; that is, the sum of the partial pressures of all minor constituents plus the carrier gas. Yet there may be altitude levels where the values of the computed saturation vapor pressures do exceed the ambient pressure. It is therefore possible to determine for any model atmosphere those altitude regions where cloud formation due to condensation or sublimation of any of its minor constituents can or cannot occur. Most important, this can be done independently of the actual amounts of the minor constituent present.

This is illustrated in Figure 1, which shows the thermodynamic pressure-temperature diagrams for water and carbon dioxide. The solid lines are the sublimation, evaporation, and melting curves for H_2O, and the sublimation curve for CO_2, respectively. They simply show the dependence of the saturation vapor pressure on temperature. The dashed curve superimposes the U.S. Standard Atmosphere 1962(6) in terms of total ambient air pressure versus temperature on the same scale. Note that in the shaded areas, the water vapor pressure values required to achieve saturation are larger than the ambient air pressure. Hence, water- or ice-cloud formation cannot occur there, regardless of the amount of water vapor present in the earth's atmosphere.

In this example, for the average conditions of the U.S. Standard Atmosphere, H_2O clouds can therefore only form between the surface and 43 km altitude, and in the region between 67 and 94 km, respectively.

THEORETICAL ANALYSIS

Let us now apply this principle to atmospheric conditions at different latitudes and seasons. We have available the so-called supplemental atmospheres(7) of the U.S. Standard Atmosphere(6), which tabulate atmospheric pressure and temperature to altitudes of 90 km between latitudes 60° north and south, for summer and winter. We can thus derive absolute limits for the regions where H_2O clouds could conceivably form as a function of latitude and season, still without any knowledge of the actual amount of water vapor present.

But we can go a step further.

Under our postulate, favorable conditions for the formation of noctilucent clouds will be present when:

(a) the atmospheric temperature is sufficiently low to permit sublimation under given ambient pressures;

(b) the amount of water vapor is sufficiently high to permit sublimation under these given ambient temperatures and pressures; and

(c) sufficient numbers of dust nuclei are present to facilitate sublimation.

At any altitude level, then, the probability for H_2O cloud formation will be inversely proportional to the given value of the saturation mixing ratio - as determined from ambient pressure and temperature data of the supplemental atmosphere - and will be directly proportional to the actual amount of water vapor present - expressed, say, in mixing ratios of gram of water vapor per gram of dry air.

Figure 2 illustrates these probabilities for H_2O cloud formation in the region near 80 km altitude. It shows water vapor mixing ratios required to achieve saturation at different latitudes and seasons, determined for the supplemental atmospheres.(7) For the altitude region shown, no cloud formation at all is possible at latitudes polewards of 30° in winter; these curves have been omitted. But note that a required mixing ratio of 1 g/g, for example, corresponds to an implausible mixture of equal amounts of water vapor and dry air. More reasonable values of the possible mixing ratio are of the order of 10^{-3} g/g(8) or 10^{-4} g/g(9). Therefore, a likely probability for saturation to occur is found only at latitudes north of 45° in summer.

Figure 2 illustrates an important theoretical result. For a wide range of possible water vapor mixing ratios, H_2O cloud formation is likely to occur only in exceedingly narrow and sharply defined altitude regions. For the saturation mixing ratio to change by one order of magnitude, a temperature change of about 12° to 15° is necessary near the 80 km level. But even considerable changes in either temperature or amount of water vapor present will shift the altitude boundaries, within which clouds can form, by only a very few kilometers.

The mesopause temperatures for the curves shown in Figure 2 range from 191°K at 30° latitude in winter to 171°K at 60° latitude in summer. The recent Kronograd experiments(10), however, recorded temperatures as low as 150°K near 80 km altitude, and rocket soundings in the tropics(11) have given temperatures between 150°K and 180°K near 90 km. Such departures from the values given by the supplemental atmospheres would shift the curves in Figure 2 to the right. This would result in high probabilities for saturation to occur even at rather low amounts of water vapor present, even for such low mixing ratios as 5×10^{-5} g/g calculated by Hesstvedt (9) just recently.

The above considerations can be graphically expanded to depict the likely variations of favorable conditions for cloud formation with latitude and season. The result is shown in Figure 3, based again on the supplemental atmospheres.(7) A priori, we know that H_2O clouds can only form within the areas bounded by the line denoted by ∞, which corresponds to pure water vapor (an infinite saturation mixing ratio). Higher probabilities of cloud formation are indicated by the lines of equal mixing ratios necessary to achieve saturation over ice. It is unlikely that water vapor mixing ratios much higher than 0.001 g/g can be found in the upper atmosphere, although a considerable amount of controversy exists in this regard.

This is easily seen by comparing, for example, studies by Hesstvedt(9), Gutnick(12), Mastenbrook and Dinger(13), and Murcray and his collaborators.(14) Thus we would anticipate that H_2O

clouds can form in the earth's upper atmosphere only in exceedingly narrow areas that are limited in both latitude and season, as indicated in Figure 3.

DISCUSSION

We have so far limited ourselves to a purely theoretical treatment of favorable conditions for H_2O clouds to form in the earth's upper atmosphere. The supplemental atmospheres(7) are based primarily on temperature, density, and wind measurements by means of sounding rockets and searchlight probes. There is no evidence that their computation was quantitatively influenced by consideration of noctilucent cloud observations.

We can, therefore, compare the above results with the observed locations, frequencies, and altitudes of occurrence of noctilucent clouds. We find, of course, that they occur indeed in sharply defined, narrow height intervals between 60 to 90 km altitude, and that they are observed only during the summer months in northern latitudes. (Refer, for example to publications by Witt(15) and Gromova(16).)

Admittedly, we know little about the variation of atmospheric temperature with altitude, season, latitude, solar activity, or time of day, near the mesopause. We know nothing about the actual amounts of H_2O present at these heights. However, with the postulate of the ice crystal theory, statistical analysis of noctilucent cloud data should provide interesting indications about the variation of mesopause temperatures and altitudes with latitude and season.

For example, Vestine(17) some 30 years ago, has pointed out that an inverse correlation seemed to exist between the frequency of occurrence of noctilucent clouds and sunspot activity between 1885 and 1932. There are already indications that as we approach the minimum of the current solar cycle, the frequency of occurrence of noctilucent clouds is beginning to increase. This becomes apparent from very recent Alaskan data published by Fogle(18). In the light of our earlier statements, this leads to the inference that during years of sunspot minimum either the height of the lower boundary of the mesopause or the temperature of the mesopause should be lower. Such inferences, in turn, should lend themselves to independent experimental tests in connection with observations of noctilucent clouds and future atmospheric soundings.

CONCLUSIONS

The purpose of the present note was to illustrate the application of a basic principle of thermodynamics to one specific phenomenon in the earth's upper atmosphere. There are other useful applications to be found in the study of the atmosphere of other planets, where we lack physical information. We hope to have been able to show that use of this principle in the context of the phenomenon of noctilucent clouds may yield profitable conclusions, and, more important, might perhaps facilitate the planning and conduct of experimental investigation and data analysis in the years ahead.

Let me add that I presented here a condensed version of Dr. Schilling's recent work. The complete paper has been submitted for publication in the Journal of Geophysical Research.

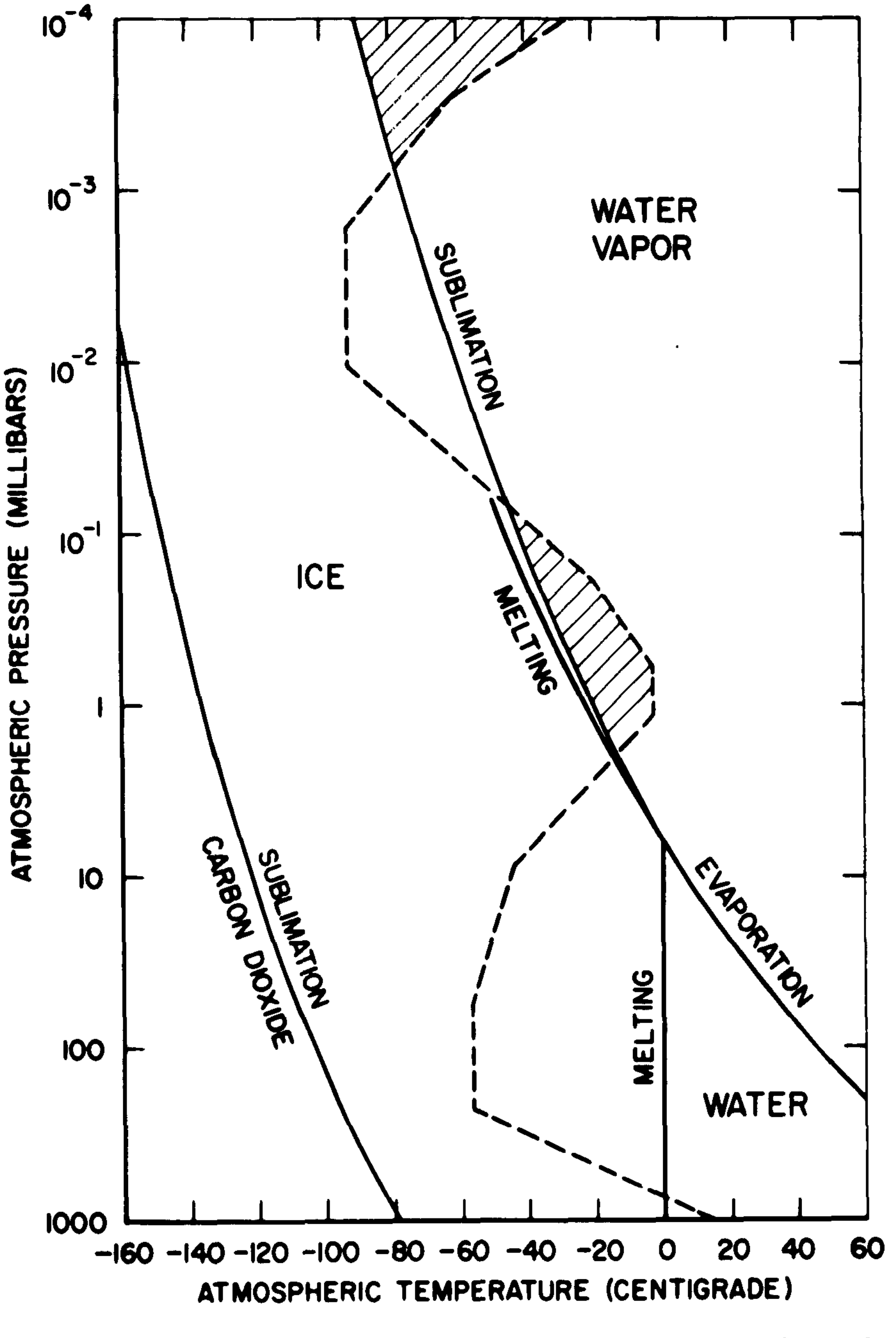

Fig. 1 Thermodynamic pressure--temperature relation for H_2O and CO_2. Dashed line is U.S. Standard Atmosphere (1962); H_2O cloud formation is not possible in shaded areas.

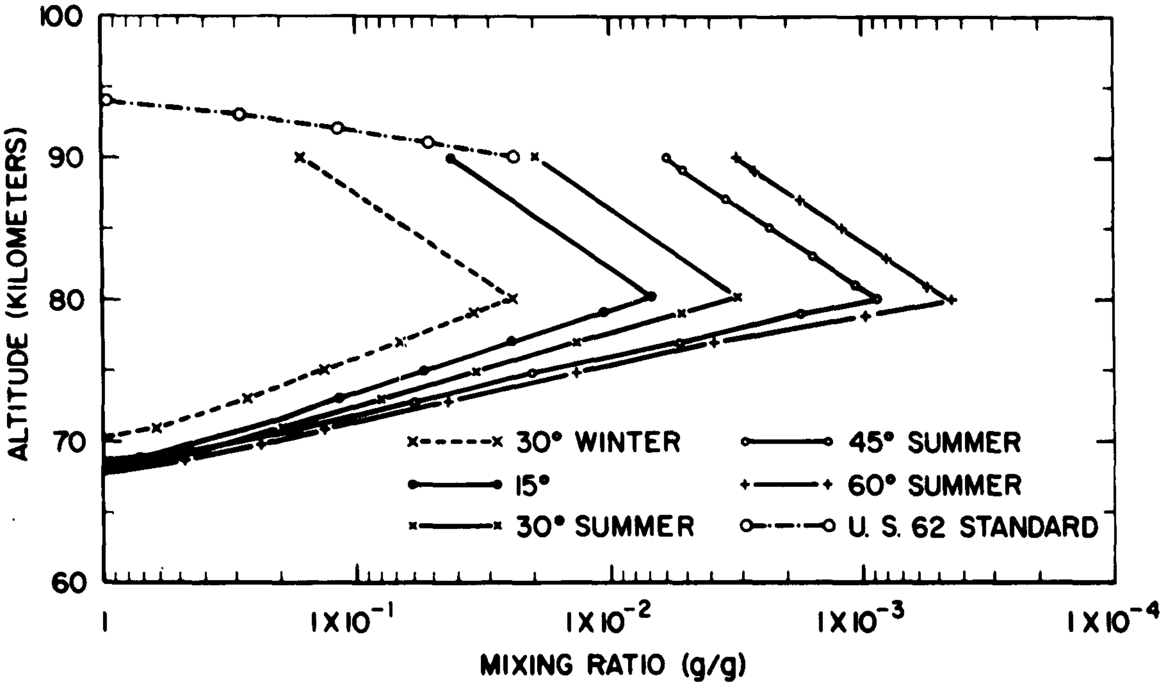

Fig. 2 Water vapor mixing ratios required to achieve saturation in the upper atmosphere at different latitudes and seasons, determined for supplemental atmospheres (Cole, et al., 1963).

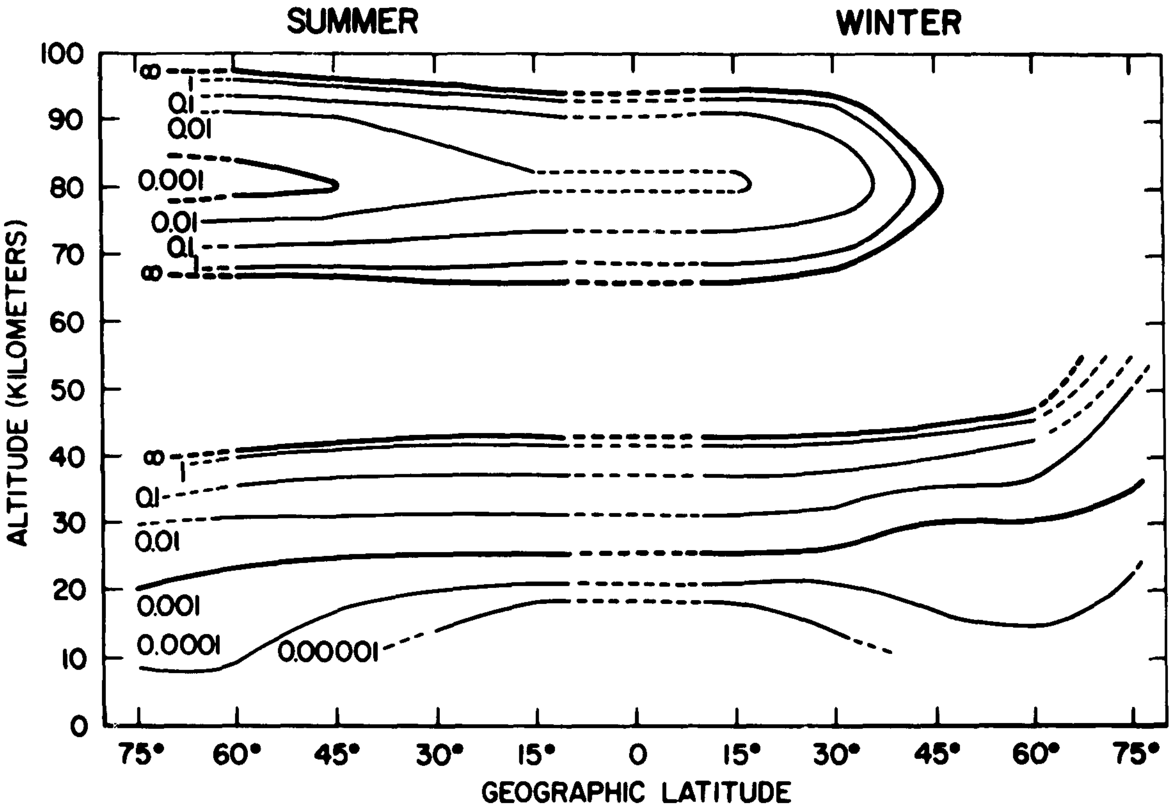

Fig. 3 Latitudinal and seasonal distribution of permitted regions for the formation of H_2O clouds in the earth's atmosphere, determined from supplemental atmospheres (Cole, et al., 1963). Lines connecting altitudes where saturation can occur are labeled with the required water vapor mixing ratio in g/g. (Values higher than 1×10^{-3} g/g are unlikely to be found frequently.)

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