A Comparison of Ice Nucleus and Ozone Concentrations in Stratospheric Air

ROGER F. REINKING[1] AND JAMES E. LOVILL

Dept. of Atmospheric Science, Colorado State University, Fort Collins

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ABSTRACT

The variations of ice nucleus and ozone concentrations were monitored at a high mountain observatory in the Colorado Rockies. Because the observatory is at an altitude of nearly 4 km MSL, increases in ozone at this station are in most instances related to intrusions of stratospheric air into the troposphere. An increase of ice nuclei simultaneously with an increase of ozone in such cases would therefore be indicative of a transport of ice nuclei from the stratosphere to the earth's surface. High ice nucleus concentrations in stratospheric air would substantiate the possibility of an extraterrestrial nucleus source. A negative correlation between the ozone and ice nuclei existed during the time of the case study presented. It is concluded that at least during this period there was not an influx of ice nuclei simultaneously with a downward transport of stratospheric air. Hypotheses for a ground origin of ice nuclei are supported.

1. Introduction

A pressing question in cloud physics is the role played by extraterrestrial materials as ice nuclei. If extraterrestrial particulates are truly active in such a capacity, their effects will be seen in the clouds of the troposphere. However, in their descent to these clouds, the particulates must, of course, pass through the stratosphere. From the lower stratosphere the material may be transported downward primarily by dynamic mechanisms. The existence of such transport mechanisms for intrusion of stratospheric air into the tropo-

[1] Present affiliation: Department of Meteorology, San Jose State College, San Jose, Calif.

sphere and to the earth's surface is well established (Danielsen, 1959, 1968; Reiter, 1963; Reiter and Mahlman, 1965). This information makes it clear that 1) the stratosphere may act as a temporary storage reservoir and thus as a source of the extraterrestrial particles, and 2) particulate ice nuclei, if present, will reach the troposphere and the earth's surface in greatest concentrations if they accompany the intruding stratospheric air.

The definition of ice nucleus population differences in stratospheric and tropospheric air has been sought by several investigators. Bigg *et al.* (1961) measured relatively high nucleus concentrations in the stratosphere by sampling with filters on balloons. The filter method is subject to a "volume effect" caused by non-nucleating particulates which decrease the nucleus count (Mossop, 1965). This effect would be small or absent in the relatively clean stratospheric air and could, therefore, very definitely give rise to high nucleus counts relative to those made in "dirtier" air near the earth's surface. Thus, the results of the balloon sampling are open to question.

Ice nucleus measurements over an Australian surface network have revealed "storms" of persistently high nucleus populations sometimes coincident with stratospheric intrusions (Droessler, 1964; Bigg and Miles, 1964). In these cases an extraterrestrial source of nuclei was suggested, but the possibility of entrainment of soil nucleants from the surface due to observed turbulent mixing was not eliminated.

Evidence against an extraterrestrial origin of nuclei has recently emerged from an airborne investigation by Cadle *et al.* (1969) in which much more uniform and lower nucleus concentrations were measured in the stratosphere than in the troposphere. More evidence in this direction has been convincingly supplied by Gagin (1969) who has found a seasonal coincidence of 1) a surface maximum of radioactive products (Be_7, Ce_{144}) which originated in the lower stratosphere, 2) favorable conditions for intrusion of stratospheric air into the troposphere, and 3) a winter *minimum* of ice nucleus concentrations.

By using radioactive particles as tracers of stratospheric air, Gagin gained a significant advantage which was very valuable to his interpretations. Ozone may also serve as a tracer of stratospheric air. Most surface ozone other than that in urban areas originates in the stratosphere and is brought into the troposphere and to the surface by the same intrusions that transport particulates across the tropopause (Danielsen, 1959, 1968; Reiter, 1963; Reiter and Mahlman, 1965; Lovill and Miller, 1968; Lovill, 1969, 1970a,b). Thus, trends and variations in O_3 and ice nucleus concentrations should be parallel if the nuclei also come from the stratosphere. The existence of this parallelity is investigated here.

Note that a positive correlation of this type can be expected only if the nuclei are sufficiently small to move with the air. If they are large enough to settle relative to the air any correlation would probably be due to chance. Measurements of the size distribution of natural ice nuclei taken at the same site and during the same week as the measurements analyzed below reveal a size range of 0.1–1 μ with model Stoke's diameters of 0.2, 0.3 and 0.6 μ for three separate occasions (Gerber *et al.*, 1970). For spherical particle of specific gravity 2 and diameters of 0.1, 0.5 and 1 μ, approximate settling rates in stagnant air are only 2, 7 and 24 cm hr^{-1}, respectively. Nuclei having these essentially negligible terminal fallspeeds may be expected to follow the slightest rising and/or falling air motions and thus be carried downward in stratospheric intrusions.

2. Instrumentation

For the present investigation ice nucleus and ozone concentrations were simultaneously and continuously monitored over a 5-day period in September 1969. The measurements were made at the Chalk Mountain Observatory, elevation 3658m (mean pressure, 643mb) near Climax, Colo. An advantage was gained in operating at this high altitude because of the proximity to any stratospheric source of particulates or O_3. Instrumentation included a Komhyr ECC ozone sensor (Komhyr, 1969) and a NCAR ice nucleus counter (Langer *et al.*, 1967; Steele *et al.*, 1967). The NCAR counter provided a continuous relative measure of the concentration of

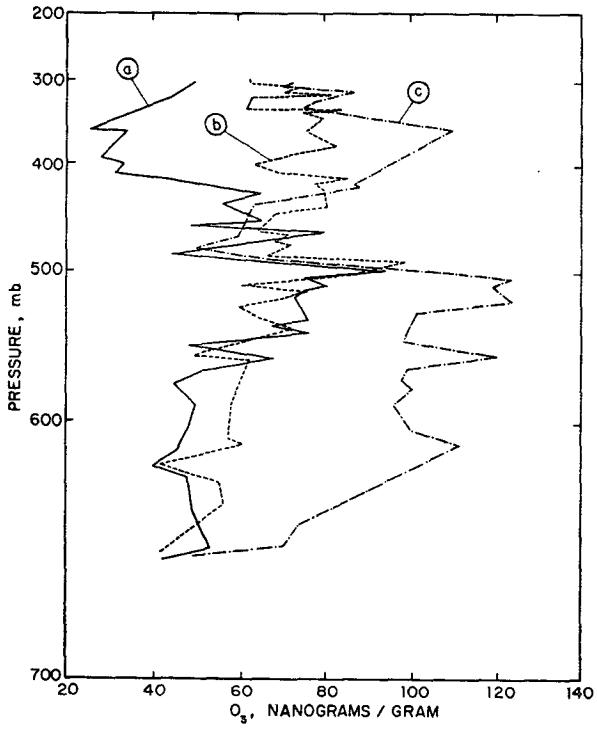

FIG. 1. The vertical distribution of ozone over the Chalk Mountain Observatory. The ozonesondes were released at 2200 MST 24 September 1969, a., 0100 MST 25 September 1969, b., and 0800 MST 25 September 1969, c.

nuclei active at −20C. These surface observations were supplemented by an intensive ozonesonde sounding program in the atmosphere above the observatory on 24 and 25 September. The radar tracked soundings provided readings on temperature, the u, v and w components of the wind, and ozone concentration.

3. Analysis

Special analyses were performed on the simultaneous surface ozone and ice nucleus data (see e.g., Blackman and Tukey, 1958). A strong peak in the O_3 spectra was observed at a frequency $f=1.4\times10^{-1}$ cycle hr^{-1} [period $\lambda(t)=4$ hr]. A spectral gap was observed at $\lambda=8$ hr. The ice nucleus spectra indicated peaks at 3.5–4 hr and at 8 hr. A spectral gap in ice nuclei was found at 5 hr. Thus, at approximately 3.5–4 hr there appeared to be an increase of both O_3 and ice nuclei. However, at 8 hr a decrease of ozone occurred simultaneously with an increase of ice nuclei. These data suggest that there was no physical correlation between the patterns of occurrence of O_3 and ice nucleus concentrations.

A picture of vertical ozone transport and simultaneous variations in ice nucleus concentrations is provided by the ozonesonde data. The O_3 mixing ratio is conserved in a given air parcel in the free atmosphere. Thus, downward or upward transport of the air may be discerned by following the trends with time of this property.

Four ozonesondes were released at approximately 3.2-hr intervals between 2200 (all times Mountain Standard) on 24 September and 0800 on 25 September. (The third ozonesonde released at 0534 experienced a malfunction in the ozone sensor shortly after release.) Fig. 1 shows vertical distributions of the O_3 mixing ratios from three of the soundings. These profiles were analyzed in time sequence and compared with the surface ozone and ice nucleus concentration variations.

The 2200 profile of ozone has a maximum at 5600 m (495 mb) of approximately 100 ng gm^{-1} (nanograms per gram). A secondary peak should be noted at 6000 m (465 mb). In general, the mixing ratio decreases from the primary maximum to the surface. In agreement with this, the curve of the surface O_3 continuous sampler (Fig. 2) indicates a mixing ratio of 35 ng gm^{-1} which is low relative to the value of the maximum aloft. An ice nucleus count of 2 (relative units) is also indicated in Fig. 2. This level is near the average for the period. [Typical ozone mixing ratios at this time of year for the lower stratosphere (~150 mb) can be determined from the ozone soundings taken at Boulder, Colo., from 1963–65 (Dütsch *et al.*, 1970). Typical values at this level were approximately ~100–200 ng gm^{-1}. Typical O_3 mixing ratios at the 500–600 mb level were ~21 ng gm^{-1} during this period as determined from 30 ozone soundings.]

The 0100 ozone profile (Fig. 1) shows the primary maximum still at 5600 m. However, two changes occurred during the 3-hr interval since the first sounding. First, O_3 values below 5000 m (540 mb) increased (values also increased above 6000 m or 475 mb). Second, the peak seen on the previous O_3 profile at

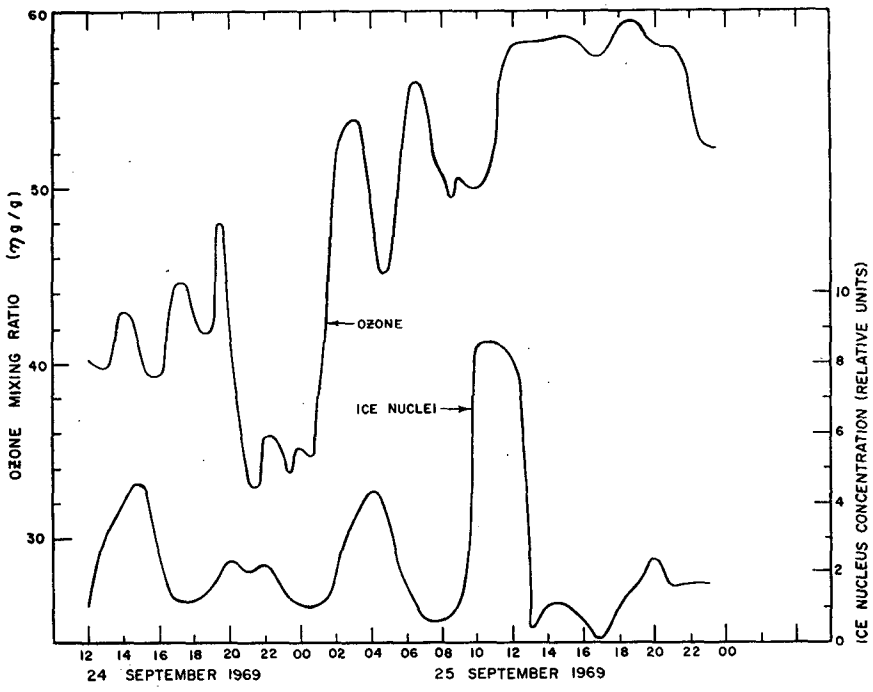

FIG. 2. Ozone mixing ratio (ng gm^{-1}) and ice nuclei (relative units) variations with time at the Chalk Mountain Observatory, Colo.

6000 m (465 mb) is no longer evident (it may have moved downward and joined with the larger primary maximum). An abrupt increase in ozone concentration beginning between 2300 and 0030 is evident from examination of the continuous surface O_3 record. The 0100 point on the ice nucleus record marks a very low ice nucleus concentration and the beginning of a gradual increase with time that lags the O_3 increase by roughly one hour. At 0300 the ozone began a temporary decrease. After an hour lag, the nuclei followed this trend. However, this pattern subsequently dissolved.

The 0800 ozone profile (Fig. 1) indicates that a large increase in O_3 occurred below 5600 m (500 mb). Most significant is that the 100 ng gm^{-1} maximum moved downward and by this time was below 4000 m MSL (610 mb). This is less than 300 m above the Chalk Mountain Observatory. The surface trend in ice nuclei at this time no longer corresponded to that in O_3. The nucleus concentration of 0.6 at 0800 was even less than the concentration before the ozone began to increase. In contradiction to the lags previously noted, a large nucleus concentration increase at 0900 *preceded* a relatively small increase in O_3 at 1000.

Evident in all ozone profiles in Fig. 1 is a general vertical increase of O_3 from the surface to ~5600 m (500 mb). In this 2000 m layer the mixing ratio increases with height, by a factor of 2. This points to the relative effectiveness of the surface as an ozone sink, as previously demonstrated by Aldaz (1969), Regener and Aldaz (1969), Dütsch (1946), Junge (1963) and Paetzold (1955). In the 0800 profile the erosion of O_3 from the 100 ng gm^{-1} region downward is quite marked. At this time (0800) it is apparent that O_3 was being transported to the surface and at the same time was being destroyed at the ground. (A slight temperature inversion was evident immediately off the surface at 0800; the wind was calm.)

The O_3 profiles considered in sequence indicate a general increase in O_3 concentration and a lowering in altitude of the 100 ng gm^{-1} region in the lower troposphere during the 10-hr period. From the continuous surface ozone sensor a rapid increase in O_3 was seen at approximately 0100 (Fig. 2). This increase became more gradual after 0200, but continued for 17 hr. The evidence is very strong that this increase in O_3 at the surface is produced by an O_3 source originating in the stratosphere that 10 hr earlier was 2000 m above the station. The movement of ozone maxima in a similar manner has been described in previous literature (Kroening and Ney, 1962; Lovill and Miller, 1968).

There is no destructive mechanism acting on particulate ice nuclei in the air approaching the surface which corresponds to that affecting O_3. Thus, any nuclei accompanying the air traced by the ozone will be available to be counted at the surface. It also should be noted that the surface O_3 sink will not destroy trends in ozone concentration. Therefore, if the ice nuclei were of stratospheric, and ultimately of extraterrestrial, origin they should increase at the surface simultaneously with increases in ozone. A higher nucleus count would continue as long as the O_3 concentration continued to increase. Early in the sounding period very roughly parallel trends in nuclei and O_3 were observed, with the nuclei lagging O_3 by ~1 hr (Fig. 2). This correspondence, however, did not last. The nuclei cycled through maxima and minima with peaks at 0400 and 1100, while O_3 continued a general increase.

4. Summary and conclusion

An analysis of ozone and ice nucleus spectra was performed on simultaneous data to see if any similar variations and peaks in the spectral curves of these atmospheric constituents were evident. None were found. A brief period during which a number of vertical O_3 soundings were made was examined to determine whether trends in ice nuclei and ozone were comparable. It appeared that no relation existed between a downward transport of O_3 from the stratosphere and ice nucleus variations at the surface.

The conclusion can be drawn from these data that for the September period of observations no significant portion of ice nuclei observed at the surface came from the stratosphere or are of extraterrestrial origin. The data add support to the arguments by Cadle *et al.* (1969) and Gagin (1969) that ice nuclei originate at the earth's surface.

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