

## ABOUT SOME PECULIARITIES OF THE SPATIOTEMPORAL STRUCTURE OF ATMOSPHERIC OZONE

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*Fast variations of stratospheric ozone derived from the data of balloon measurements at the edge of a circumpolar vortex, as well as diurnal and faster variations of surface ozone in polar and tropical regions are considered.*

Ozone accumulated in the lower stratosphere and the troposphere is used as a tracer with the long lifetime. It allows one to track the motion of air masses. At the same time, strong variability of stratospheric and surface ozone is observed. Some examples of so variable ozone are given in the present paper.

### STRATOSPHERIC OZONE VARIATIONS

The ozone layer structure at high latitudes is determined by a number of physical-chemical parameters: air temperature, aerosol content, solar radiation intensity, availability of substances involved in the process of ozone decomposition, and dynamic processes. Thin layers with sharply different ozone content are observed at the edge of a circumpolar vortex in winter and spring. Differential advection, instability of the vortex edge, and wave motions in the stratosphere are responsible for these layers.<sup>1-4</sup>

The Polar Institute of Geophysics carried out balloon measurements of ozone at Swedish station in Kirune as part of the experiment EASOE in winter-spring of 1991-1992. A hemiluminescent ozonometer, briefly described in Ref. 5 together with the data of measurements performed on January 31 of 1992, was used as a detector. In this balloon flight, according to maps of the potential vortex constructed by the European Medium-Range Weather Forecast Center and calculated trajectories of air cells, the ascending balloon was outside the circumpolar vortex for altitudes below 19 km and inside the vortex within the 23-25.5 km altitude range.<sup>6</sup> After drifting at the maximum altitude (25.5 km) for about half an hour, the balloon started to descend with a velocity of 4 m/s close to that of its ascent. During balloon ascent and descent, we recorded strong variations of the partial ozone pressure, which was not surprising in the close proximity to the vortex edge when the balloon trajectory even intersected it.

The ozone pressure and the balloon flight altitude measured at the stage of balloon drift are shown in Fig. 1. Considerable variations of the partial ozone pressure are seen, even though the flight altitude

derived from radar measurements and determined from readings of a balloon-borne pressure sensor varied by 350 m at most about its average value. It should be taken into account that the drifting balloon has no motion with respect to surrounding air in stationary case and must record unchanged ozone content of course, if it remains constant.

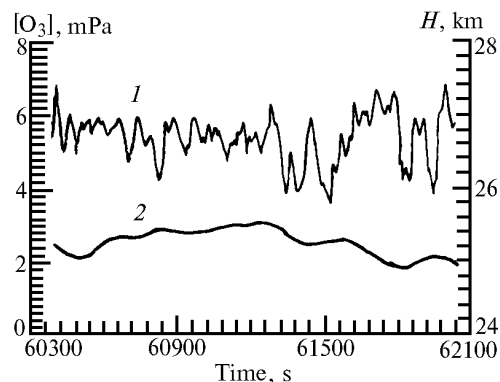


FIG. 1. Variations of the partial ozone pressure (1) in the stratosphere and altitude of the balloon drift (2) in Kirune on March 31, 1992.

The observed variations can be explained in two ways. First, we actually recorded the variations of the partial ozone pressure caused by a certain variable factor of only slightly mobile surrounding air. The factor of this kind may be the change in the temperature that decreased from  $-79$  to  $-82^{\circ}\text{C}$ , that is, down to the threshold temperature for the formation of polar stratospheric clouds from nitrogen oxide trihydrate. However, short periods of ozone decrease followed by that of its recovery have urged us to the second factor, namely, the variations of the ozone content due to wave motions of air surrounding the balloon when the ozone layer has a horizontal or vertical gradient. The balloon is an inertial device and hence it is brought into adjacent stratospheric zones and the ozonometer records the corresponding values of the ozone pressure.

## SURFACE OZONE VARIATIONS

### 1. Fast variations and pulsations in the tropics

As known, ozone is the secondary pollutant in the troposphere formed by photochemical reactions in which nitrogen oxides, hydrocarbons, and other compounds are involved. These substances are basically of anthropogenic origin. The bulk of the experiments on the anthropogenic effect on surface ozone was performed at mid-latitudes of the northern hemisphere. It was found that the temperature increase and the enhanced concentration of nitrogen oxides led to intensive formation of ozone in the surface layer of the atmosphere and increased probability of ozone smog formation.

The number of measurements in polar regions is much less. Therefore, it is of great interest to elucidate how strong is the urbanistic effect of an individual city on the surface ozone concentration at high latitudes and to compare it with measurements in the tropics of the southern hemisphere.

The Polar Institute of Geophysics (PIG) performed measurements of the surface ozone concentration in collaboration with the Vatin Institute of Physics at the University of Campinas, São Paulo, Brazil in 1993. The measurements were carried out with a hemiluminescent ozonometer, developed at the PIG, for half a year. Half the measurement period fell within the dry period and its latter part fell within the pluvial period. Main results of this work were published in Ref. 5. We mention here only two of them. First, in this industrial state of Brazil, like in the industrial northern hemisphere, the marked increase of the ozone concentration of anthropogenic origin was often encountered accompanied with smog puffs, in which the photochemical ozone generation took place,<sup>7</sup> being formed as a result of industrial emissions and mass burning out of dead wood and sugar cane before harvesting. Second, a broad spectrum of temporal variations of the surface ozone concentration was revealed in the process of continuous computer-controlled data recording every one second. Examples of the temporal behavior of the surface ozone concentration given below were obtained in the suburbs of Campinas.

Figure 2 illustrates the diurnal behavior of the ozone concentration (one-minute averages) in the pluvial period. The first two sharp decays of concentration after 13 h, LT coincide with the start and intensification of a tropical shower. Further the direct dependence disappears: ozone variations observed after 18 h, LT do not coincide with the change in the meteorological situation. The dips in the dependence of the surface ozone concentration appeared from day to day without precipitation and visible change of the atmospheric conditions. The evening decay of the ozone concentration was often sharp and nonmonotonic by its nature. In addition to slow variations with a

period of 5–10 min seen in Fig. 2, faster pulsations with a period of about one minute are also seen typical of solar days with the enhanced ozone concentration. Spectral analysis of a large number of analogous episodes did not reveal any dominant frequency or several frequencies. Several maxima were recorded between 50 and 400 s. As a rule, the interval between 100 and 200 s was more pronounced.

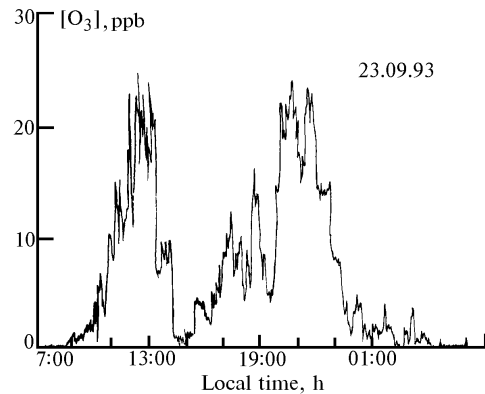


FIG. 2. Diurnal behavior of the ozone concentration at the station Campinas, Brazil on September 9, 1993.

Fig. 3 shows the example of registergram segments and corresponding periodograms obtained by the Lomb–Scargle least-squares method in Zhang version.<sup>8</sup>

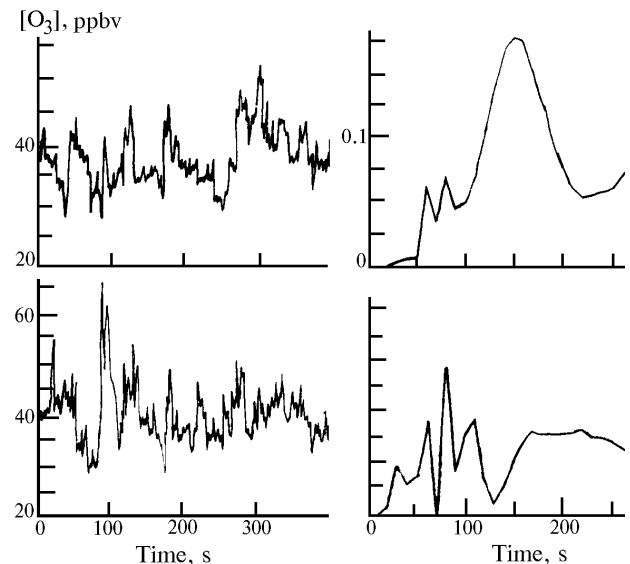


FIG. 3. At the left: two examples of recording of fast variations of the ozone concentration at the station Campinas. Data sampling period is 1 s. Measurement period is 400 s. At the right: periodograms for these records.

Sites of recording were changed several times, but the fine temporal structure remained unchanged. For this reason we abandoned the hypothesis that a local

source of ozone decomposition may exist. We seek an explanation in nonlinear character of ozone generation and decomposition in photochemical smog. Changes in the proportion between ozone and NO, illumination, and other factors may lead to termination of ozone generation and onset of its absorption. In this case, the process may be characterized as explosive instability.

It is natural to assume that at sunset the instability will be observed in local cells or jets rather than in large air volumes. Alternating cells and jets may result in the observed pattern of irregular pulsations.

## 2. Diurnal variations of the surface ozone concentration at high latitudes

Experiments were performed in Apatity (the Murmansk Region) in 1993–1995 (see Refs. 9 and 10). It was found that the ozone content decreased by several tens of per cent in the vicinity of sources of atmospheric pollution by nitrogen oxide and hydrocarbons (motor transport, heat-and-power stations, etc.) at temperatures below 15°C typical of high latitudes. Under certain conditions (calm and surface temperature inversion), ozone may be completely destroyed and there arises a situation that in this sense differs from that observed at mid-latitudes. In this case, the zone of urbanistic effect for a city with one hundred inhabitants extends approximately to 10 km from the city boundary and the plume of atmospheric pollution by nitrogen oxides may extend to several tens of kilometers.

Marked ozone generation occurs at large distances from sources of pollution for sufficiently strong ultraviolet solar illumination. In this case, experiments performed at two stations located in rural regions of Kol'skiy peninsular (the first station was to the southwest of Apatity at a distance of 15 km and the second station was to the east of Lovozero settlement, with about 10 000 inhabitants, at a distance of 7 km) in different seasons indicated that the surface ozone concentration during the polar day was primarily determined by its photochemical generation upon exposure to the ultraviolet solar radiation. Diurnal variations of the surface ozone concentration decrease sharply at the polar night. This is illustrated by Fig. 4. Here, curve 1 is for observations made at the first station in summer; curves 2 and 3 are for observations at the first and second stations, respectively, in winter; curves 4 and 7 describe diurnal variations of the intensity of directly transmitted solar radiation in June–July and March, respectively; and curves 5 and 6 are for observations at the first station in March of 1993 and 1994, respectively.

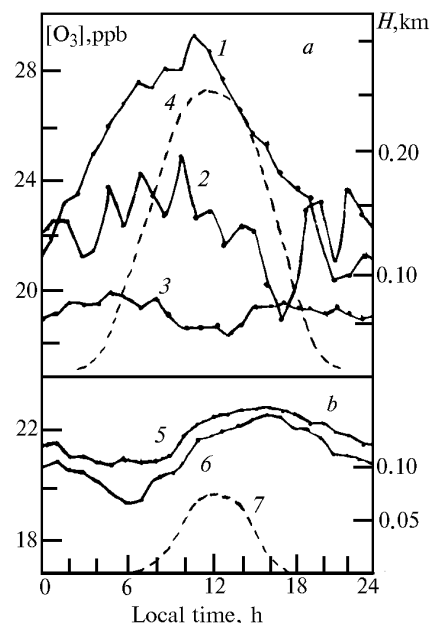


FIG. 4. Average daily variations of the surface ozone concentration in different seasons in the Arctic.

Thus, we can draw a conclusion that by polar day the photochemical ozone generation is so superior to ozone absorption and transport that the night decrease of the ozone concentration observed at midlatitudes is hardly seen in the diurnal ozone behavior recorded at the rural station.<sup>11</sup>

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