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## ON FLUCTUATIONS OF OZONE IN THE TROPOSPHERE AND STRATOSPHERE

Following is a translation of an address presented by Erich Regener to a convention of the JGGU in Brussels on 30 August 1951. The address was published in the German language in the periodical Journal of Atmospheric and Terrestrial Physics, 1952, Vol 2, pages 173-182, Pergamon Press Ltd, London.

### 1. Abstract

The complexity of the problem of atmospheric ozone is exhibited in the fluctuations of the ozone concentration at all heights. In the layer of air immediately adjoining the ground, the ozone content always sinks to zero when the air stagnates, due to the effect of the ground, which is very destructive to ozone. In the troposphere, advection plays the main part in causing fluctuations. Widely varying vertical ozone distributions have been found recently by means of the spectrographic method in balloon ascents. To explain this, large-scale horizontal and vertical movements of air at great heights must be assumed.

### 2. Body of Address

To speak of fluctuations of ozone means to speak of the entire complicated problem of atmospheric ozone. We know that the fluctuations vary according to time and locality. They occur at all altitudes where ozone is present at all. The amplitude of the fluctuations is different in all places and if periodic variations are involved, daily and annual time influences are superimposed upon local effects. We can therefore expect that all factors which are material for the entire ozone problem are more or less effective in relation to the fluctuations: the rate of formation and of the destruction of ozone, its geographic origin and even its history. Given these conditions, it would seem advisable to assemble as much observational data as possible and to group and interpret individually noteworthy situations. In the following, then, let us discuss in summary the relationships based upon old and new measured data, progressing from lower to higher altitudes.

The fluctuations of the ozone content of air layers near the surface of the earth seem to us today to be the most simple. Percentage-wise, the fluctuations are also greatest here; for here the ozone content value of zero occurs at least for periods of time but quite frequently and under certain conditions even regularly. This is an indication that the surface of the earth is the scene of considerable ozone destruction. In view of the chemical structure of ozone, this is to be expected. Everywhere on the surface of the earth, particularly there where there is vegetation, oxidizable substances are present which consume the ozone. Dust which may be present in the air and the industrial gases of our cities function in the same manner. Ozone is also consumed in catalytic roles upon coming into contact with completely inactive solid bodies. There is a spontaneous decomposition of ozone even in a completely clean glass vessel which naturally increases greatly with the temperature. The same applies in the free and completely clean atmosphere.

When ozone is found in the air near the earth, therefore, it must come from above -- from those layers of air wherein a balanced concentration of ozone is forced through the combined effects of ozone-building and ozone-destroying wave lengths of sunlight and by purely thermic, and therefore greatly dependent upon temperature, ozone decomposition. Observations show that the theoretically calculated ozone distribution can be changed locally to a great degree through advection and convection at all altitudes. But at altitudes below 20 kilometers the relative ozone content of certain moving masses of air changes only very gradually, becoming noticeable only after several days (1). The ozone content drops rapidly only when in contact with the surface of the earth or with dust -- a condition which of course cannot exist in clean air over extensive bodies of water. According to our present knowledge and opinion, a photochemical formation of ozone in the vicinity of the earth's surface is not possible. Only in strong electrical fields such as occur in storms is the formation of ozone possible through occasional point discharges. These occur so seldom, however, that they could hardly play a role in the ozone balance.

The transportation of ozone from air strata which are not influenced by the destructive effects of the earth's surface down to earth apparently is brought about by turbulent exchanges of air. This turbulence can vary very much in intensity and it can be very local or wide-spread in extent. But it is from the relationships between the type and the intensity of the turbulence and the decrease in ozone concentration near to the earth's surface that one must draw conclusions as to the strength of the ozone-destructive effects of the earth. Observations show that the ozone content can drop to zero in a very short time -- in 1-3 hours -- not only in stagnant, completely stationary air, but also in the presence of weak, laminar winds passing slowly over flat terrain.

Observations also show that ozone destruction at the surface of the earth takes place particularly rapidly if during the night, because of an inversion near the earth, a thin strata of air prevents exchange with higher layers. In such a case, as soon as the sun appears over the horizon in the morning, the small and localized thermal currents which result are sufficient to destroy the inversion near the earth relatively quickly and to allow uninfluenced, ozone-rich air masses to descend. Above flat terrain, the ozone destroying effects of the earth probably do not reach up very far, possibly 50-100 meters, naturally depending upon the degree of air mixing. When temperature inversions extend over large areas and where large areas of cold air settle in valleys, the effects of course could extend upwards much further.

Given strong turbulence, especially with westerly gradient winds, the turbulent exchange takes place much more rapidly and also extends much further upward. Under these conditions, observations show that the transfer of ozone from upper stratas overcomes the destructive effects of the earth's surface and the amount of ozone at ground level is more or less equal to that in free air at small altitudes.

This process is naturally greatly dependent upon the local conditions and orography and can best be described by using examples. For this reason, I shall now give some measurements which were made by my associates in the vicinity of Friedrichshafen am Bodensee (Lake Constance) shortly before and also during the war. Conditions there where the large water surface meets the flat shore area are particularly simple and the individual effective factors can be clearly traced in the measurements.

Illustration 1 shows the daily progress of ozone content values measured by R. Aner on fair, windless fall days using a rapid-functioning potassium iodide method. A high ozone content is recognized during the daytime which drops to a non-detectable value after sunset, rising again after sunrise with increasing thermal effects in the air at the surface of the earth.

The graph prepared by A. Ehnert (3) also shows that it is the thermal turbulence in the lowest layers of air caused by the warmed earth which brings ozone down from above after the nightly minimum. This graph is shown in Illustration 2 and reveals a parallel between air temperature and ozone content with a maximum between 2 and 3 o'clock P.M. A striking feature of these curves, as well as of those in Illustration 1, is the rapid decrease of ozone content which accompanies the sinking of the sun towards evening. Apparently, a temperature inversion is quickly formed which lies close to the surface and which seals off a thin layer of air against exchanges with higher layers. The destruction of ozone can then take place rapidly in this thin layer. This effect can be reinforced by the reversal of the movement of air which carries ozone deficient air from land to the lake in the evening

and back again in the morning. Given conditions of minimal movement of air, one finds air in the morning which has been located over the lake all night and hardly exposed to ozone-destructive influences.

In Illustration 3, the upper curve shows higher ozone values and an absence of the nightly minima with strong westerly winds. The destruction of ozone on the ground is compensated for completely by ozone brought in from higher layers. The large fluctuations can apparently be connected with wide-spread conditions of turbulence of the air which brings masses of air of varying ozone content to the measuring site. In the vicinity of the Bodensee, this can be masses of lake and of land air.

During calm weather on the banks of the Bodensee, an anti-parallel course can be observed between turbulence and humidity (4). From this, the decrease in turbulence can be estimated from the increase in humidity. When the air is still, it quickly takes on a very high relative humidity. On the other hand, when thermal currents occur, dry air comes down from the higher layers. For this reason, we can see an inverse relationship (see Illustration 4) between humidity (which is reciprocal to turbulence) and the ozone content. In this curve, the sudden increase in the amount of ozone at 8:30 in the evening is interesting. At this point, a squall moved over the measuring station at a rate of 8 meters per second. In addition to oxygen, this brought dry air down from greater altitudes, as can be seen from the jagged humidity line at this point in time. As the squall passed, the ozone content soon returned to zero and the humidity again increased. Naturally, a condition of turbulence can also cause the ozone content at the surface of the earth to drop if the general weather conditions should bring down a layer of air which had a lower ozone content than prevailed near the earth.

Local conditions are not always so simple as on the Bodensee. As the configuration of the land changes, so must also the daily course of ozone content near the earth change. These changes depend upon the degree to which stagnated air and temperature inversions can form and the degree to which exchanges of air are influenced by the configuration of the terrain and local wind conditions. This is borne out especially by the many measurements of ozone which are carried out at resort areas for climatological reasons. In mountain areas it often occurs that there is a second ozone maximum during the night which can be explained as due to a katabatic wind which brings down ozone-rich air. Everywhere in valleys or low areas where night brings the formation of a cold pool with a temperature inversion, the nightly ozone minimum is to be found, however.

In order to determine the altitude to which the ozone-destructive effects of the earth extend in a single case, we have set

two measuring devices at our new research station at Weißenau (20 kilometers from the Bodensee). The devices are built on the new principle by A. Ehmert. One was placed in a meadow, the other atop a 20 meter mast (see Illustration 5). Although the landscape here is only gently rolling, the daily ozone behavior has proved to be by far not as simple, even during calm weather, as on the banks of the Bodensee at Friedrichshafen. The slight depressions in the earth's surface, the forest and a nearby river valley exert their influences upon the wind and turbulence and therefore also upon the daily ozone situation. Nonetheless, there are some days when the influence of the 20 meter difference in altitude between the two apparatuses is clearly manifested. Thus we see in Illustration 5 that the daytime ozone values are higher and at night the ozone concentration atop the tower usually does not fall as low as that at the base of the tower.

Continuous ozone registrations are presently being made at four locations in southern Germany and at one place in Switzerland (Arosa) with the new ozone measuring device designed by Ehmert and he will report on these in this periodical in the near future.

The influence of great differences in altitude can be seen from Illustration 6 on which the ozone conditions as measured each day by Ehmert in Friedrichshafen at an altitude of 400 meters and at the eastern end of the Bodensee at 1064 meters on Pfändergipfel Peak are plotted. We note that the nightly decrease in ozone content which was observed in Friedrichshafen does not exist in the more disturbed air atop the Pfändergipfel which lies exposed to the West. Occasionally, as on 17 September 1940 at 10:00 o'clock P.M., more ozone could be found in Friedrichshafen, which again indicates a rotation of air masses.

The differences in daily ozone conditions under extensive area relationships between a valley and mountain station are shown even more clearly by two measurements which were made with a chemical method recently in New Mexico (J. G. Bowen and V. H. Regener (3)). The measurements were made simultaneously atop a somewhat isolated mountain -- Capilla Peak, 2800 meters high, 900-1500 meters higher than the surrounding area -- and at Station Acoma, which lies in a broad valley but is still 2000 meters high. Illustration 7 shows that fluctuations in ozone concentration caused by weather conditions occur only very irregularly at the mountain station, whereas regular strong ozone minima occur each night at the valley station. (The measurements were made during December.) In actuality, as shown by the temperature curve in Illustration 8, pronounced nightly temperature minima occur regularly at Acoma Station. These prevent an exchange of air with higher altitudes, but this is not the case at the mountain station (Illustration 9). These measurements show that the nightly decreases in ozone concentration also occur in a broad valley in mountainous terrain.

although this takes place somewhat more gradually because of the vast scale of the terrain. The ozone minimum therefore occurs towards morning, and on the third day of our observations even seemed to be displaced to the following afternoon. The dissociating effect of the earth's surface apparently extended to an enormous volume of air under the given local conditions. The balance brought about through turbulence apparently took longer. In general, it could be said that the areas having little or no ozone are larger and more persistent where ever where favorable local conditions exist for the formation of large pools of cold air at night.

Now as regards the fluctuating distribution of ozone in the free troposphere, I can only refer to the measurements of A. Knert (6) which he made in an aircraft during the war using the chemical method. The four measurements in Illustrations 10-13 were made at intervals of 2-3 days and, with one exception, reached to an altitude of 8.5 to 9 kilometers. On the first day (Illustration 10) we see first a minimum at an altitude of 4 kilometers, followed by a slight increase in the ozone/air ratio at 7 kilometers. Two days later (Illustration 11) we observe a pronounced maximum at an altitude of 2 kilometers which shifted to 4-5 kilometers two more days later (Illustration 12). Again three days later (Illustration 13), the maximum is seen to have become dissipated and a constant value for the ozone/air ratio existed between 2 and nearly 6 kilometers altitude -- an indication that extensive mixing had taken place in the air layers between those heights. The temperature line plotted on the left and the indicated clouds show the way in which this layer was isolated by inversions. In view of these plotted values, there can be no doubt that the vertical exchange of air (advection and turbulence) play a role in the distribution of ozone in the troposphere. Which factor has the greater effect in individual cases can only be determined if the vertical distribution of ozone in the troposphere could be measured simultaneously at several places.

Finally, as regards the distribution of ozone at still greater altitudes, I shall show here a collection of eight distributions (Illustration 14) which represent both old and newly obtained data and which were obtained, with but one exception (8), by means of balloon ascents with spectrographs. The variation of these curves as compared to our knowledge of 10 or more years ago is surprising. Beside the well known old curves which showed a maximum in the area of 20 kilometers or higher, we now see curves where the maximum lies either higher or lower, as well as some which show a second maximum at lower altitudes, such as those found via the reversal method (7). Worthy of particular note is the fact that the ratio ozone/air varies greatly above 20 kilometers. This can only be explained by assuming that a severe and rapid mixing must have taken place. The second maxima at lesser altitudes, which are usually found during the spring of the year, are most likely to be explained by advection of low-lying polar bodies of air with a high concentration of ozone. Probably the



the maximum which occurs at about 23 kilometers altitude can be connected with the photochemical theory as found by the first early balloon ascents. My associate H. K. Paetsold recomputed the vertical ozone distribution using the formulas given by Schröder (9), but varying the constants such as the (still not clearly understood) absorption coefficient of oxygen, which are applicable for ozone formation. He also tried changing Beer's Law as well as varying the assumed temperature at the altitudes which came into consideration for ozone formation. The results (Illustration 15) showed, however, that all this had little effect and that the curves still agreed for the most part.

Therefore, if the curves which are observed are found to vary, the difference between them must be considered to be due to the influence of advection or turbulence. In any case, the curves shown indicate the presence of sizeable vertical and horizontal movements at higher altitudes which have not been sufficiently studied to date.

Illustration 16 shows an attempt made by H. K. Paetsold to connect the shapes of four curves recently obtained with the spectrograph to the origin of the air masses. The origin of the air masses is naturally given with reservations, for the trajectories at an altitude of 16 kilometers were used, and the way in which air moves at 16 kilometers is not known. Nevertheless, a certain connection seems to be indicated in that for the two curves shown on the left which have a pointed maximum, the air came from the area of the Azores, whereas the trajectory for the lower right distribution curve, which has a pronounced maximum below 20 kilometers and shows indication of another at a greater altitude, pointed to North America. The curve at the upper right, for which the air at 16 kilometers altitude came from Greenland shows, in addition to a sharp maximum at 23 kilometers, quite constant and somewhat high ozone content between 10 and 20 kilometers. The curve which seems most to correspond to the photochemical balance and which also has a pointed maximum which is usually connected with a low overall concentration of ozone is that measured in equatorial air masses. The deformed curves seem to correspond to the polar air masses which occur here in Europe most often in the spring and which have a higher overall ozone content (10).

To give a better picture, four characteristic cases measured with the spectrograph are plotted together in Illustration 17 (11), (12).

### 3. Summary

The entire complexity of the problem of atmospheric ozone is reflected in the fluctuations of ozone concentration at all altitudes. In the layers of air lying immediately next to the surface of the earth, the ozone content always drops to zero in stagnated air because of the destructive effects of the earth's

surface. In the troposphere, advection is the major factor in ozone fluctuations. Recently greatly varying distribution of ozone at higher altitudes have been found using the spectrographic method in balloon ascents. Extensive horizontal and vertical movements of air must be assumed in order to explain this feature of ozone distribution at great altitudes.

#### 4. Bibliography

(1) Moser, Berichte des Deutschen Wetterdienst, US Zone (Reports of the German Weather Service, US Zone), No 11, page 28, 1949. (2) R. Auer, Gerlands Beiträge zur Geophysik (Gerland's Contributions to Geophysics), 1939, Vol 54, page 137. (3) A. Ehmert, Berichte des Deutschen Wetterdienstes US Zone (Reports of the German Weather Service, US Zone), No 11, page 47, 1949. (4) A. Ehmert, loc.cit.(3). (5) J. G. Bowen and V. H. Regener, Journal of Geophysical Research, September 1951. (6) A. Ehmert, loc.cit. (3), page 63. (7) Götts, F. W. P., Gerlands Beiträge zur Geophysik (Gerland's Contributions to Geophysics), 1931, Vol 31, page 119. (8) Curves by H. K. Paetzold, Zeitschrift für Naturforschung (Journal of Nature Research), 1950, Vol 5a, page 661. (9) E. Schröder, Berichte des Deutschen Wetterdienstes, US Zone (Reports of the German Weather Service, US Zone), No 11, page 15, 1940. (10) Cf. Moser, loc.cit.(1). (11) E. Regener, H. K. Paetzold and G. Pfozser, Naturwissenschaften (The Natural Sciences), 1950, Vol 37, page 359. (12) V. H. Regener, Nature, London, 1951, Vol 167, page 276.

FIGURE APPENDIX

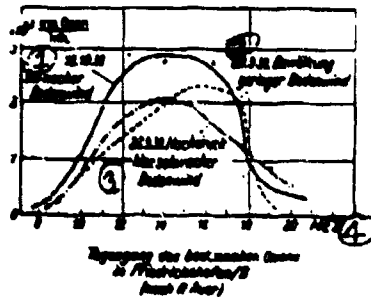


Illustration 1. Daily condition of ozone near the earth's surface in Friedrichshafen/B (according to R. Auer).

- Legend: ① 12 October 1938, weak ground wind  
 ② 29 September 1938, slight ground wind  
 ③ 26 September 1938, high pressure, clear, weak ground wind  
 ④ Central European Time

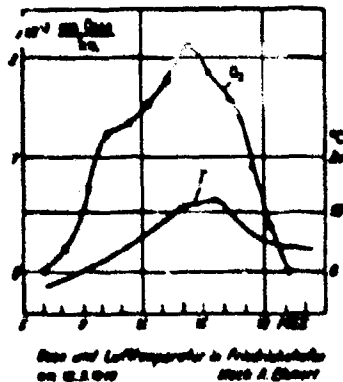


Illustration 2. Ozone and air temperature in Friedrichshafen on 12 March 1940, by A. Ehnert.

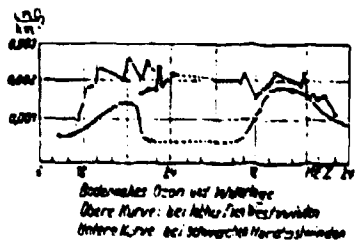


Illustration 3. Ozon und weather conditions near the earth's surface.  
 Upper line: with strong westerly winds  
 Lower line: with light northeasterly winds

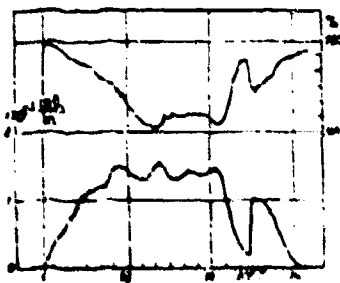


Illustration 4. Ozon (top) and humidity (bottom).

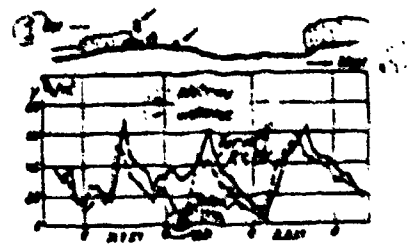


Illustration 5. Ozon situation at altitudes of 1.4 and 20 meters.

- Legend:
- ① East
  - ② West
  - ③ Cloudless at Weissenau
  - ④ Tower, 21.5 meters
  - ⑤ Ground, 1.4 meter

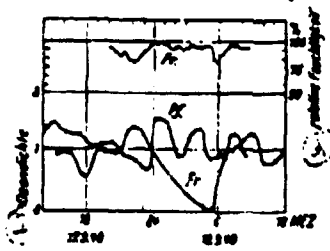


Illustration 6. Ozone situation atop Pfändergipfel Peak and in Friedrichshafen.

Legend: 1 Ozone density  
2 Relative humidity

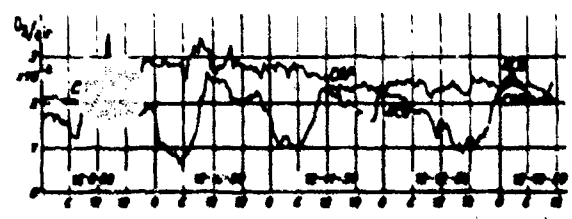


Illustration 7. Ozone situation on a mountain peak (CAP) and in a broad valley (ACO).

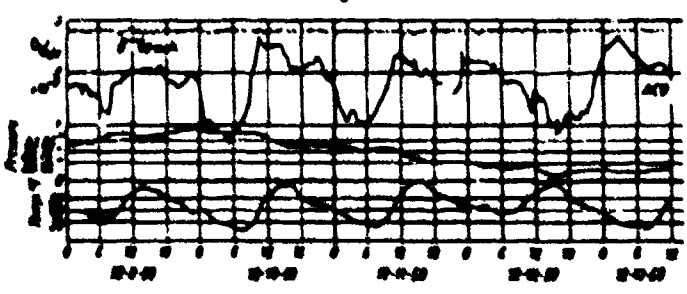


Illustration 8. Ozone situation, air pressure and temperature in a broad valley (ACO).

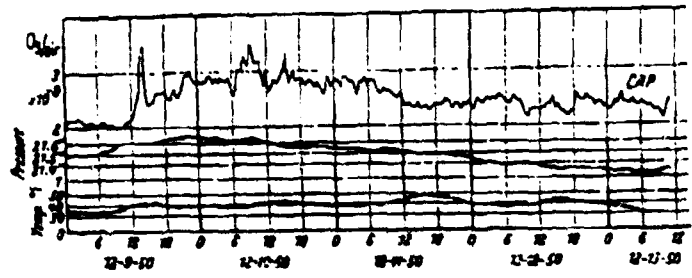


Illustration 9. Ozone situation, air pressure and temperature atop a mountain peak (CAP).

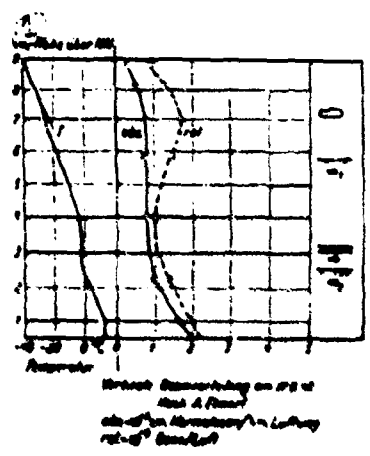


Illustration 10. Vertical ozone distribution on 17 August 1942. By A. Ehnert. abs (absolute) =  $10^{-3}$  on normal ozone/kilometer path of air. rel (relative) =  $10^{-6}$  ozone/air. Legend: (1) Height above sea level

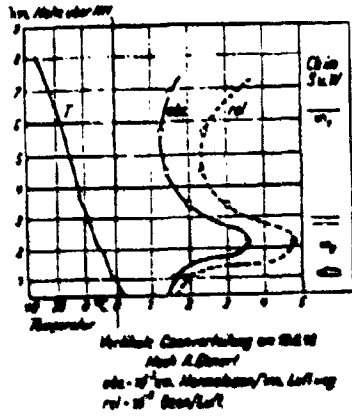


Illustration 11. Vertical ozone distribution on 19 August 1942.  
 By A. Ehnert. (Legend -- see Illustration 10.)

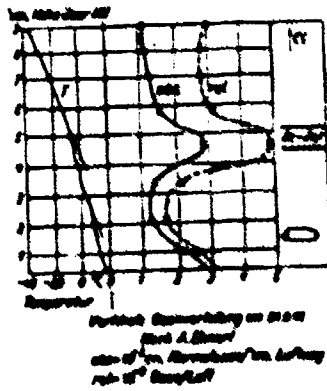


Illustration 12. Vertical Ozone distribution on 21 August 1942.  
 By A. Ehnert. (Legend -- see Illustration 10.)

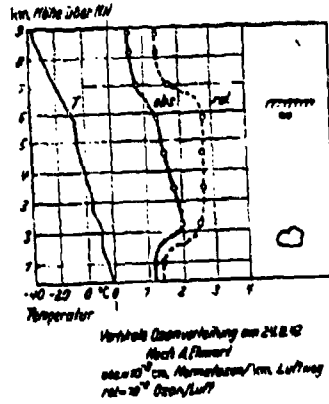


Illustration 13. Vertical ozone distribution on 24 August 1942.  
By A. Ehsert. (Legend -- see Illustration 10.)

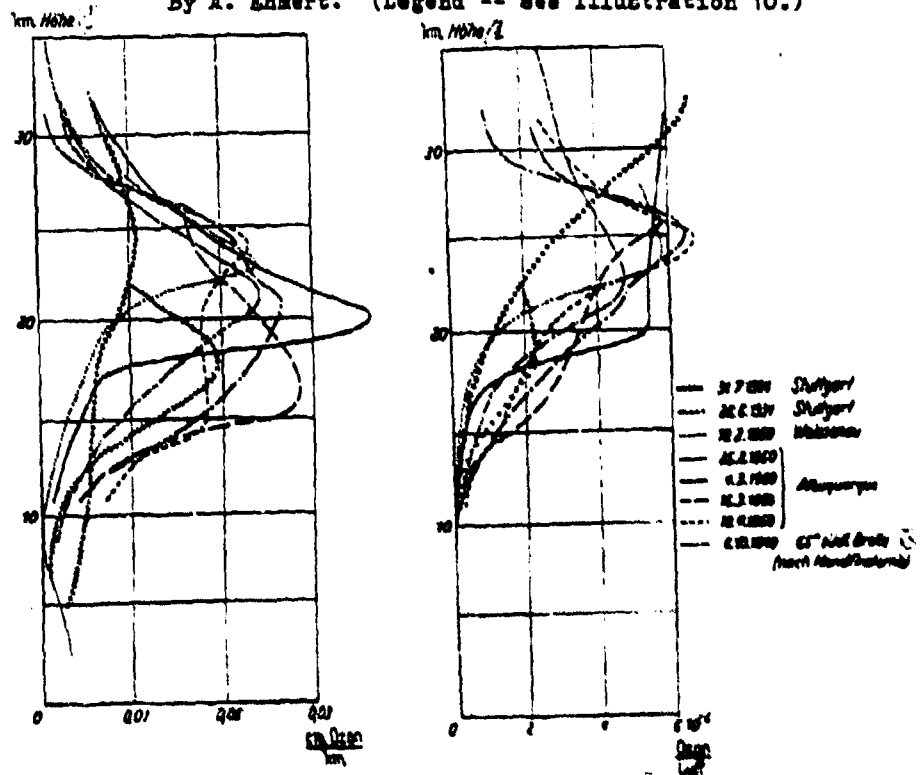


Illustration 14. Greatly varying vertical ozone distribution.  
Legend 1 Altitude 2 65° South latitude  
3 ozone/air (following lunar eclipse)



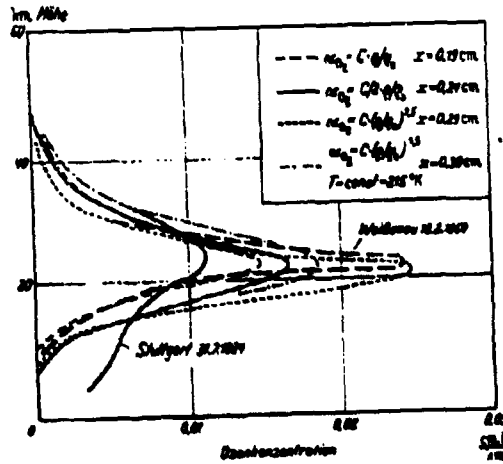


Illustration 15. Theoretical vertical ozone distribution.

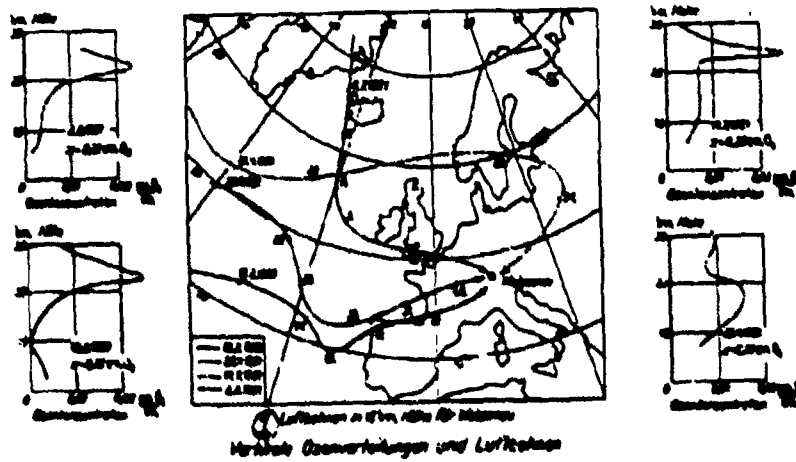


Illustration 16. Vertical ozone distribution and movements of air masses.

Legend: ① Routes of air masses at altitude of 16 kilometers for Weissenau

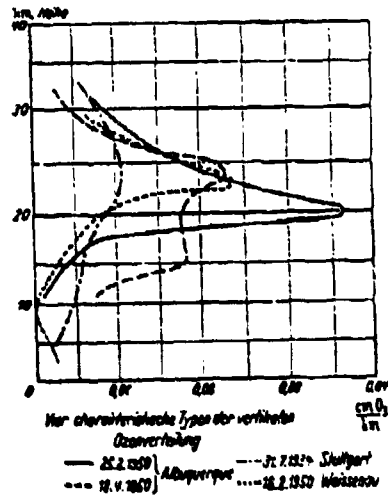


Illustration 17. Four characteristic types of vertical ozone distribution.