

# STRATOSPHERIC O<sub>3</sub>-INTRUSIONS CAUSE TROPOSPHERIC O<sub>3</sub>-BACKGROUND IN MIDDLE EUROPE

by

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## 1. Summery

Simultaneous measurements of air pollutants and meteorological components by telemetrical measuring systems have shown, that cold weather fronts of cyclones, coming from western directions, are nearly always accompanied by increasing Ozone concentrations in Central Europe. This Ozone is a contamination of stratospheric cold and dry air masses, which are transported by jet streams into the biosphere during tropospheric air folding. It comes from lower stratospheric regions and forms mainly the tropospheric O<sub>3</sub>-background. Therefore the biosphere was already in pre-industrial times exposed by O<sub>3</sub>. This known O<sub>3</sub>-production seems to be underestimated. In the following paper some examples of measurements by the ground level telemetrical measuring system ZIMEN in Germany show that these natural O<sub>3</sub>-Intrusions are producing monthly averages of at least 10 to 40 µg/m<sup>3</sup>. Short time fluctuations of this intruded stratospheric Ozone have concentrations which are within the range of the lower limiting values to protect plants. Therefor this phenomenon should be considered by a new definition of Ozone limiting values to protect human beings and the nature.

## 2. Telemetrical Measuring Network ZIMEN of Rheinland-Pfalz.

To control the development of air pollution, telemetric networks have been installed in all sixteen federal countries of Germany. In the country Rheinland-Pfalz there are located 32 measuring stations. The control centre of this system, called ZIMEN, is located in the capital Mainz. The measuring network gives information about expansion, transportation and sources of air pollution. Measurements have been started first in industrialised towns in 1977 to get information about the development of air pollution in urban areas. To study the causes of forest decline, measurements of air pollution have been started 1984 at five sites in the forested highlands of Rheinland-Pfalz. Long time results of ZIMEN are published 1998 at 11.th Clean Air Congress in Durban /1/. Short time data are published in monthly reports since 1978 /2/. Actual data **from all 17 German air pollution networks** you can get by internet calling "<http://www.umad.de>".

## 3. Long-time Measurement Results of Ozone (O<sub>3</sub>)

Since about 1982 Ozone is considered as an important air pollutant. O<sub>3</sub> is one of the strongest gaseous oxidiser. It attacks the needles of conifers and reduces the assimilation of CO<sub>2</sub>. Therefore the first measurements of O<sub>3</sub> have been started to study the cause of forest decline in Germany. Fig. 1 shows monthly averages of O<sub>3</sub>, averaged about five forested places and six urban stations. Seasonal alternations of O<sub>3</sub> correspond with the curves of global radiation and air temperature. The mechanisms of the photochemical production and transportation of O<sub>3</sub> are well known: Tropospheric O<sub>3</sub> mainly origins by photolysis of NO<sub>2</sub> in the presence of peroxides produced from mainly anthropogeneous hydrocarbons. These precursors NO and hydrocarbons are mainly produced by traffic, gas power plants and households.

O<sub>3</sub> is a harmful respiration gas and attacks the lungs. Therefore O<sub>3</sub> has been measured in towns too. Sliding annual concentrations of O<sub>3</sub> in forests are nearly twice as high as in urban regions (Fig. 1).

It is remarkable, that in winter-times with relative small photochemical production of tropospheric O<sub>3</sub> due to low global radiation and at sites far away from sources of anthropogenic precursors, the monthly averages are relative high (between 30 and 50 µg/m<sup>3</sup>).

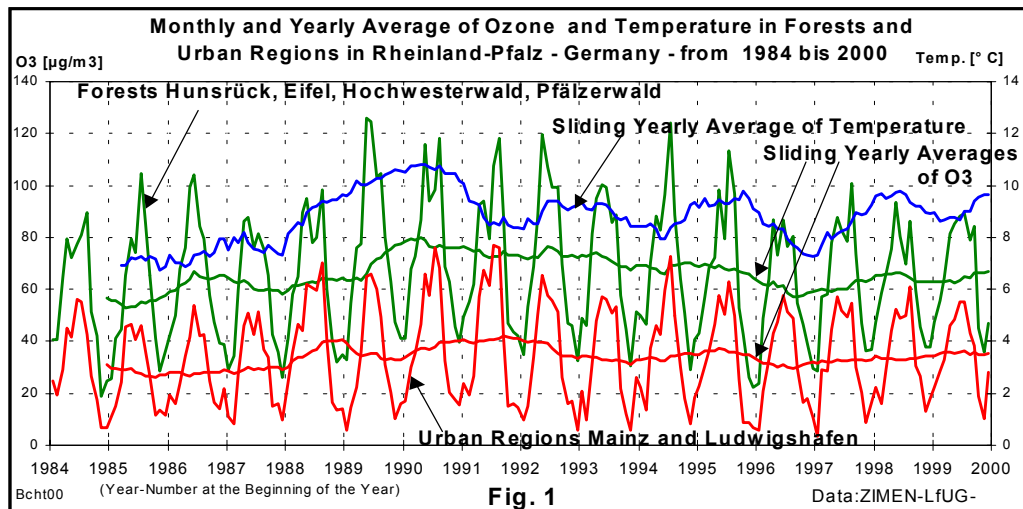


Fig.1: Monthly and Annual Averages of O<sub>3</sub>- Concentrations in Towns and Forests

Therefore it is of interest to look for the cause of this phenomena. **It was found, that during every intrusion of cold and dry western winds, beginning with increasing pressure after its minimum, relative high Ozone-concentrations occur.** As an example Fig.2 shows the course of half hour averages of O<sub>3</sub>, pressure, absolute humidity and wind-direction in the month November 1991, measured in a suburb of Mainz.

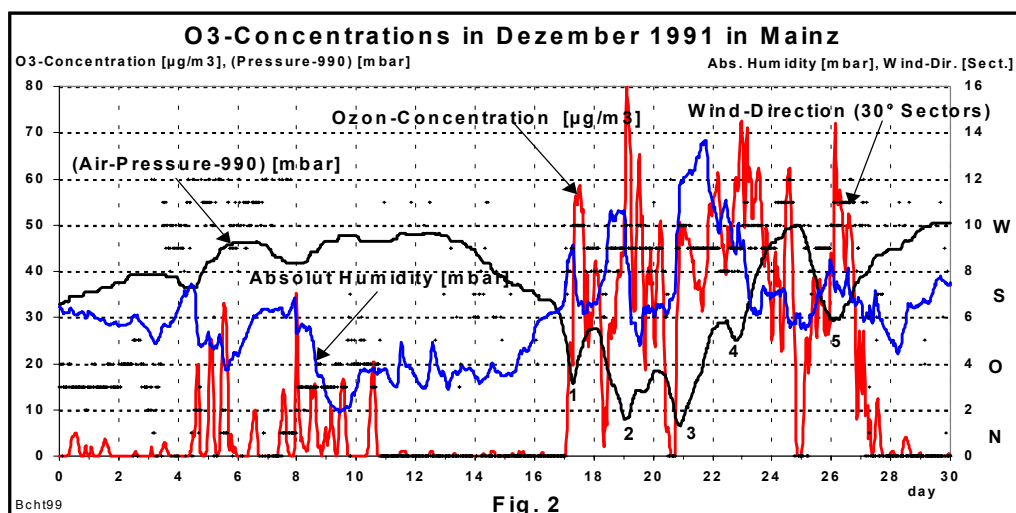


Fig.2: Half-hour Averages of Ozone, NO<sub>2</sub>, air-pressure, humidity and Wind-direction.

A “family” of 5 cyclones appear with intrusions of dry and cold western winds. Each cyclone is indicated by a minimum of pressure. **Generally at this points the O<sub>3</sub>-concentrations increase.** Such O<sub>3</sub> - Intrusions are observed as well in urban regions as in forests: Cold dry air coming during the cold-front of the cyclone by the yet stream from stratospheric regions, is contaminated with Ozone /3/. Such frontal-

cyclones are often shown in the daily weather forecast at television. Cyclones occur when cold air masses coming from the north-western region to eastern directions are meeting warm air masses, which are coming from south west directions to Europe /4/.

#### 4. O<sub>3</sub>-Intrusion during Frontal-Cyclones in Detail

The following Fig. 3 and 4 show a typical example of intrusions of stratospheric Ozone during two frontal-cyclones (No. 1 and 2 in Fig. 2) together with simultaneously measured meteorological parameters at urban and forested measuring sites.

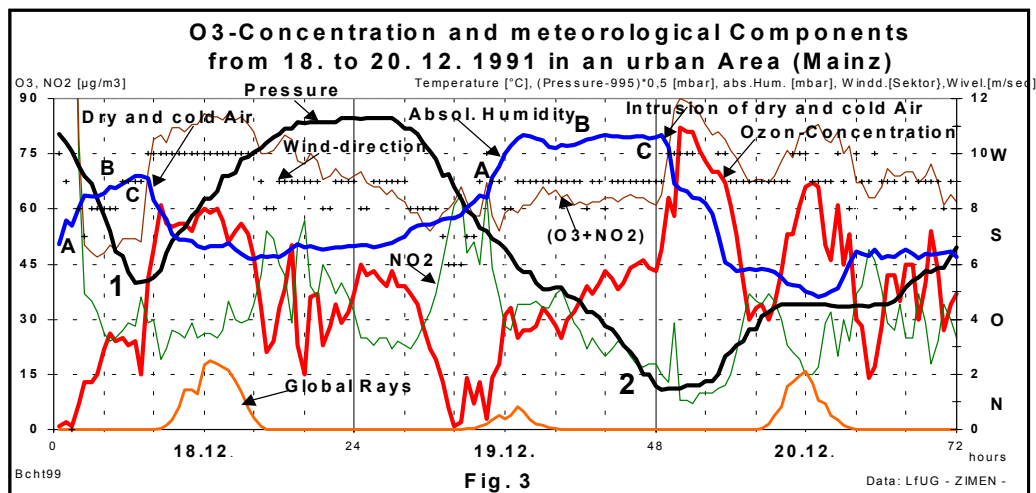


Fig. 3: O<sub>3</sub>-Intrusion during two frontal-cyclones, measured at the suburb station Mainz

Frontal-cyclones start with slow increasing temperature and absolute humidity. The air comes mainly from south-western directions (sector 8) into the country and is accompanied by some precipitation (Warm-front at Point A). Usually a region with nearly constant warm and humid air masses is following (Warm-region B). This region ends again with precipitation. Then follows the intrusion of cold air masses with increasing and changing winds from more western directions (sector 9 to 12) accompanied by rising O<sub>3</sub>-concentrations (Point C). Temperature and absolute humidity are falling very strong (about 2 °C/h resp. 2 mbar/h). Air-pressure is at this moment at its minimum and starts to increase with about 2 mbar per hour.

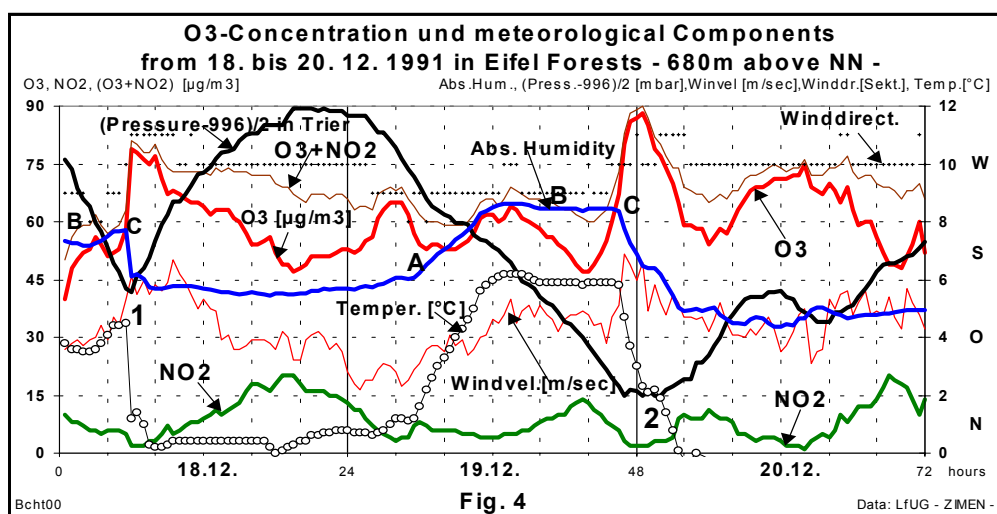


Fig. 4: O<sub>3</sub>-Intrusion during two frontal-cyclones, measured at the Forests Eifel

This cold and dry air is the outcome of a tropospheric air folding and originates mainly from stratospheric cold regions, transported by a jet stream /5/.

Fig. 4. shows the same frontal-cyclones as Fig. 3 but measured in the forest region of the Eifel mountains.

The comparison shows that the averaged concentrations of  $O_3$  in the forests are about twice the concentrations in urban regions. **This fact is the consequence of the oxidation between the intruded  $O_3$  and mainly anthropogenic NO in the lower troposphere:** In forest regions, this influence of NO is relatively small. In urban regions this influence is high. Concentrations of the intruded stratospheric  $O_3$  are reduced by this relative quick oxidation with the velocity of about  $v = 27$  [1/(ppm\*min)]. **That means, the real  $O_3$ -concentration intruded into the lower troposphere is given by the sum ( $O_3 + NO_2$ ).** This is valid, if the measured  $NO_2$  is not produced direct within the source (ca 5% of the  $NO_x$ -Emission of the motor car) or by the quicker oxidation of NO with Peroxides (RO2) ( $v = .2300$  [1/(ppm\*min)]) produced by anthropogeneous hydrocarbons. So far as there is no other relevant  $NO_2$ -Production, the real intruded stratospheric  $O_3$ -concentration by yet streams is given by the sum ( $O_3+NO_2$ ). ( Using the units [ $\mu g/m^3$ ], the failure is lower than the statistical failure of the measurements of both components.)  $NO_2$ -production by the very slow oxidation of NO with  $O_2$  is not very relevant.

### 5. $O_3$ -Flux into the Lower Troposphere

To estimate the quantity of  $O_3$ -molecules transported by such events into the lower biosphere it can be assumed, that the  $O_3$ -concentration of the air masses at the measuring high of wind velocity at 10 m above ground (measuring point of wind components) is within 10 percent the same  $O_3$ -concentration as at measuring high of 3.5 m above ground (sampling point of the air pollutant monitors). Multiplying the half hour average of  $O_3$  with the averaged wind velocity, you get the current of  $O_3$  through a vertical square meter per second in units [ $g/(m^2*sec)$ ] at the high of 10 m. Fig. 5 shows that there is no relevant difference of the accumulated  $O_3$ -flux between forests and towns on the condition that anthropogenic NO is changing a part of the intruded  $O_3$  into  $NO_2$ . Only the relative high situated forest station in the Eifel shows some higher values. At this site the intruded  $O_3$  first appears with air masses from western directions.

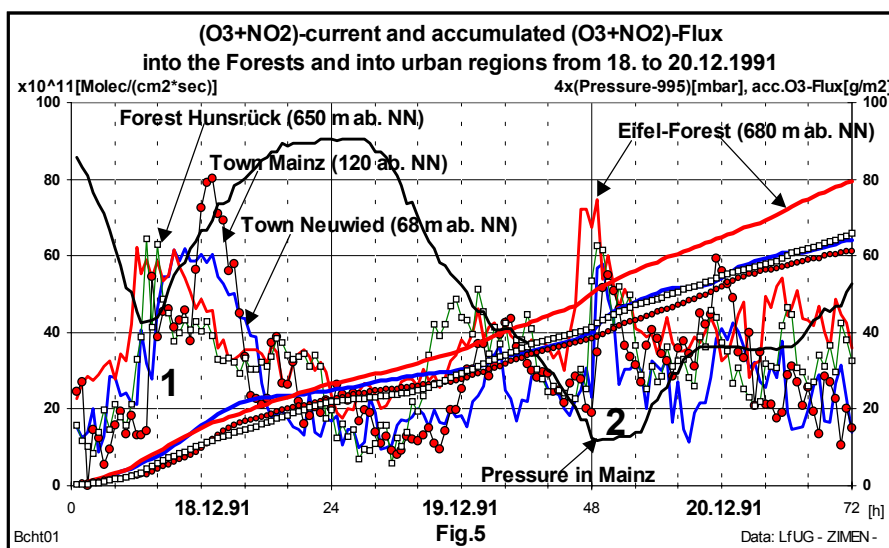


Fig. 5:  $O_3$ -Flux, caused by two frontal-cyclones into forest and urban region.

On condition that these O<sub>3</sub>-contaminated air masses will reach the soil, the deposition of O<sub>3</sub> lies between 4 and 40 g/m<sup>2</sup> per frontal-cyclone in Central Europe. In this example O<sub>3</sub>-currents from stratosphere into biosphere by jet streams reach values up to 80x10<sup>11</sup> [molec./(cm<sup>2</sup>\*sec)] That is a little above ranges of earlier published estimations /6/.

## 6. The Monthly Concentration of Stratospheric Ozone-Background

To estimate the toxic relevance of this intruded O<sub>3</sub> the resulting monthly background is to be calculated. The smallest possible monthly O<sub>3</sub>-background concentrations are given by the measuring values of O<sub>3</sub> and NO<sub>2</sub> which correlate with decreasing **absolute** humidity and which come from **western directions** (wind sector 8 to 12 of the 30 degree wind scale between 1 (North) and 12, marked by bigger points in Fig. 6).

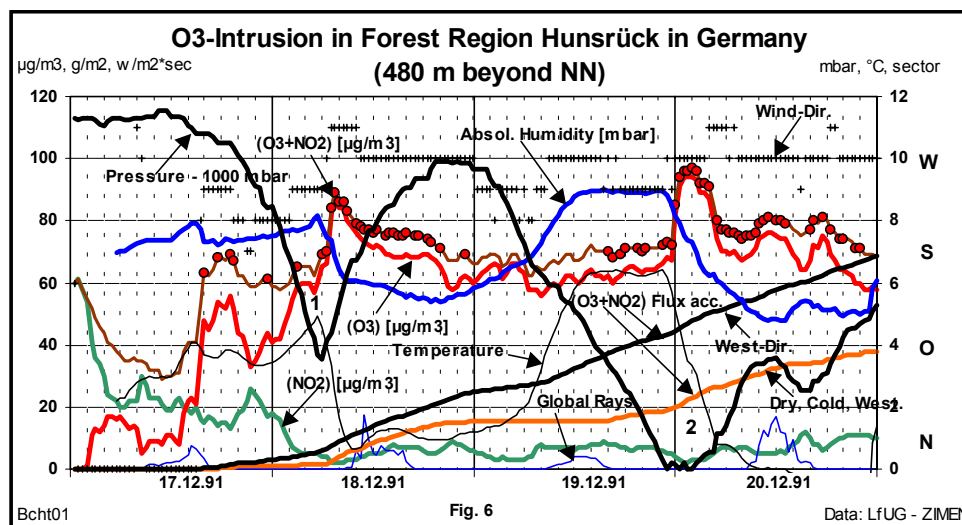


Fig. 6: O<sub>3</sub>-Intrusion in Forest Region Hunsrück in Germany

From this one must conclude that intrusions of stratospheric Ozone by frontal cyclones into the troposphere produce a natural O<sub>3</sub>-background with monthly averages of at least 10 to 30 µg/m<sup>3</sup> countrywide (Fig. 7). Surely the natural O<sub>3</sub>-background can be higher because the stratospheric air masses of the jet stream get mixed partly with air masses of the next upcoming western warm-front, which is on this way transporting rests of stratospheric O<sub>3</sub> into the biosphere.

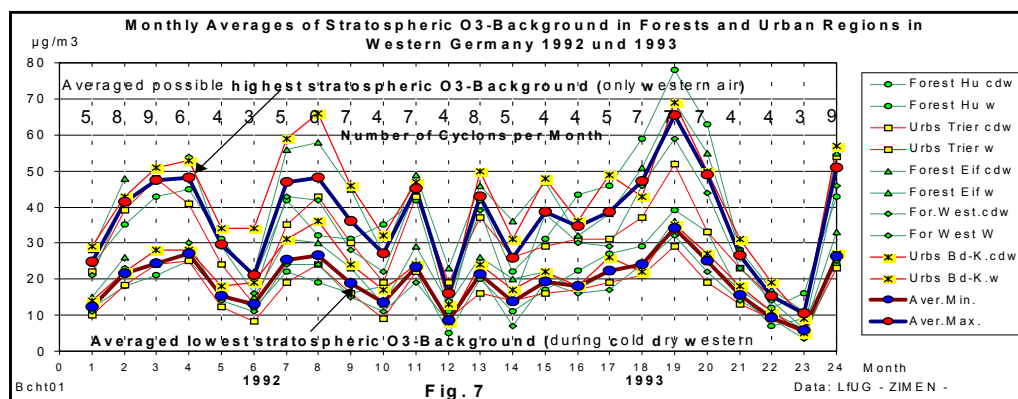


Fig. 7: Monthly averages of O<sub>3</sub>, produced by frontal cyclones in towns and forests.

This condition leads to an estimation of the possibly highest values of the O<sub>3</sub>-concentration produced by western frontal cyclones. This argumentation is supported

by the fact, that both, the lowest and the possibly highest values of these  $O_3$  are respectively nearly equal in forests and towns (Fig. 7). The Quantity of the monthly stratospheric  $O_3$ -background depends on the intensity and the number of frontal cyclones per month. Fig. 8 shows a correlation between the Number of frontal-cyclones and lowest resp. possible highest stratospheric  $O_3$ -background countrywide.

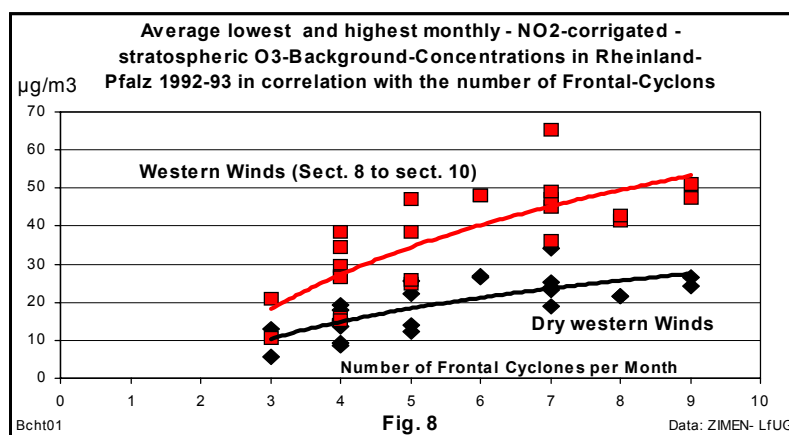


Fig. 8: Correlation between Number of frontal cyclones and stratospheric  $O_3$ -background

It seems, the more frontal cyclones occur per month, the lower is the intensity of intruded stratospheric  $O_3$  per frontal cyclone up to a certain saturation.

## 7. Conclusion

Measuring the concentrations of  $O_3$  and  $NO_2$  together with the meteorological components at several different sites in the lower troposphere (3,5 to 10 m above ground) we find a natural background of  $O_3$  with a monthly average between 10 to 50  $\mu\text{g}/\text{m}^3$ . This background is produced by cold and dry western air masses contaminated by stratospheric  $O_3$  which are transported into the biosphere by jet streams caused by tropospheric folding. In the lower troposphere the  $O_3$ -concentrations are reduced by anthropogenic  $NO$ , which is to consider when estimating the quantity of intruded stratospheric Ozone. Examples show that stratospheric  $O_3$  produces background concentrations of this kind all over the year. The magnitude depends on the frequency of frontal cyclones per month. This phenomenon already existed in pre-industrial times and exists surely everywhere in the world where frontal cyclones occur. This Ozone comes from the lower region of the stratosphere. It should be of interest to prove these results by correlation with cosmogenic radio nuclides /7/.

## 8. Literature

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