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Long term Effect on the Ozone Layer of Nitrogen Oxides produced by thermonuclear Explosions in the Atmosphere

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FOREWORD

The paper entitled "Long term Effect on the Ozone Layer of Nitrogen Oxides produced by thermonuclear Explosions in the Atmosphere" has been submitted to the "Annales de Géophysique".

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LONG TERM EFFECT ON THE OZONE LAYER OF NITROGEN OXIDES PRODUCED BY THERMONUCLEAR EXPLOSIONS IN THE ATMOSPHERE

by

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Abstract

The explosion of thermonuclear bombs in the atmosphere leads to the formation of nitrogen oxides. In order to try to derive any significant variations of total ozone related to the injection of the NO_x molecules during the nuclear test series of 1961 and 1962, an analysis of the ozone records is presented. The data provided by the Arosa station since 1932 and by the ozone world network since 1957 are considered. No long term ozone depletion can be found. However, if the buldges observed in the ozone records at low and high latitudes are statistically significant, they could be attributed to a chemical chain producing ozone in presence of HO_2 and NO.

Résumé

Des oxydes d'azote sont produits à l'occasion des explosions thermonucléaires dans l'atmosphère. Ces molécules sont susceptibles d'altérer l'ozonosphère. Aussi, les observations d'ozone ont été analysées afin d'examiner son comportement durant et après les campagnes d'explosions nucléaires en 1961 et 1962. A cette fin, on porte son attention sur les données fournies d'une part par la station d'Arosa (Suisse) depuis 1932 et d'autre part par le réseau mondial d'observation de l'ozone depuis 1957. Aucune diminution de la quantité d'ozone n'est observée. Par contre, on remarque au lendemain des campagnes d'explosions, un accroissement de la teneur en ozone qui, s'il est significatif, pourrait être expliqué par une chaîne chimique productrice d'ozone en présence de HO_2 et NO.

Samenvatting

Tijdens thermonucleaire explosies worden stikstofoxydes gevormd. Deze molekulen kunnen de ozonosfeer wijzigen. Ten einde de variaties in ozon te onderzoeken die optraden tijdens en na de kernproefnemingen van 1961 en 1962, werden de waarnemingen gedaan te Arosa sinds 1932 en het ozon wereldnet sinds 1957, onderzocht. Geen enkele vermindering van de ozonhoeveelheid werd vastgesteld. Men merkt echter op dat juist na de kernexplosies, een toename van de ozonconcentratie optreedt, die, indien zij belangrijk is, verklaard kan worden door een chemische keten die ozon produceert in de aanwezigheid van HO_2 en NO.

Zusammenfassung

Stickstoffoxyden werden während thermonuklearen Explosionen in der Atmosphäre produziert. Diese Teilchen können die Ozonsphäre verändern. Die Ozonbeobachtungen werden deswegen analysiert um das Ozonbetragen während und nach den nuklearen Explosionen in 1961 und 1962 zu studieren. Die Daten kommen einerseits von der Station in Arosa (Schweiz) seit 1932 und andererseits vom Weldnetz der Ozonbeobachtungen seit 1957. Keine Verminderung in Ozon ist beobachtet. Man sieht ober nach den Explosionen ein Steigen im Ozoninhalt, das durch eine chemische Produktionskette von Ozon in Anwesenheit von HO_2 und NO erklärt werden kann, wenn solches Steigen bedeutsam ist.

INTRODUCTION

The possible action of nitrogen oxides on the ozone layer has been first pointed out by Crutzen (1970) and Johnston (1971) who suggested that the catalytical cycle

$$\begin{aligned} NO + O_3 &\rightarrow NO_2 + O_2 \\ NO_2 + O &\rightarrow NO + O_2 \\ \hline O_3 + O &\rightarrow O_2 + O_2 \end{aligned}$$

could be the most important destruction process of stratospheric ozone. These considerations were based on laboratory determinations only and in order to establish the efficiency in the real atmosphere of this chemical mechanism, different large perturbations leading to the production of NO (such as the explosion of thermonuclear weapons or the appearence of PCA events) were considered by various authors.

In particular several studies have been attempted to investigate the short term local effects (Christie, 1976, Miller, Krueger, Prabhakara and Hilsenrath, 1976, Heath, Krueger and Crutzen, 1977, Johnston, 1977) of nuclear bombs. Moreover, the long term and large scale action of the nitrogen oxides produced during the Soviet and American nuclear test series of 1961 and 1962 has been investigated. Foley and Ruderman (1972), who have pointed out that this type of analysis permitted a partial simulation of the injection of NO_x by a fleet of stratospheric aircraft, did not find in the ozone records any significant trend which could be related to the 1961-1962 explosions serie. This conclusion has been confirmed by other authors, namely Goldsmith, Tuck, Foot, Simmons and Newson (1973), Angell and Korshover (1973, 1976) and Bauer and Gilmore (1975). However, Johnston, Whitten and Birks (1973), after a careful examination of the data provided by 30 ozone stations around the world, reported an averaged 3.3 percent ozone reduction between 1960 and 1962 followed by a gradual increase for 1963-1970.

The purpose of this article is to present a new analysis of the ozone records and to try to derive any significant variations of the amount of ozone which could be related to the thermonuclear explosions.

FORMATION OF NITRIC OXIDE BY ATMOSPHERIC NUCLEAR EXPLOSIONS

The tremendous amount of heat liberated at the occurence of a thermonuclear blast dissociates most atmospheric molecules and particulary O_2, N_2 and N_2O in the atmospheric region where the temperature becomes larger than 2000 K. Table 1 (CIAP, 1974) reproduces the dominant reactions involved in the high temperature air dissociation. As the air cools off gradually, the odd nitrogen particles recombine with a rate depending critically on the temperature so that, below 2000 K, the relative NO concentration stabilizes at about 8×10^{-3}. The problem of the formation of NO molecules related to thermonuclear explosions has been treated by a number of authors. Their conclusions concerning the amount of NO produced per megaton do not quite agree. Considering the investigations of Zeldovich and Raizer (1967), Foley and Ruderman (1972), Johnston *et al* (1973), Goldsmith *et al* (1973) and Gilmore (1974), it appears that the understanding of the formation and injection of NO in the stratosphere is not yet complete and that the overall yield should be included in the range of (0.15 to 1.5) x 10^{32} NO molecules per Mt.

During 1961-1962, intensive thermonuclear tests have been carried out in the high latitude regions (Novaya Zemlya - USSR, 72-77°N - 52-58°E) corresponding to a total intensity of about 300 Mt (Glasstone, 1964). In 1962, different US explosions took place in the equatorial and tropical zone (Christmas Island, 15°N - 157°W; Johnston Island, 16°5N - 169°W) with a total intensity of 37 Mt (Glasstone, 1964). Considering that the stratospheric residence time is of the order of 1 to 5 years (Reiter and Bauer, 1974) a simple calculation shows that the atmospheric burden of NO due to the nuclear explosions during the 1961-1962 test series should have been of the same order of magnitude as the amount of NO present in the natural stratosphere. However, the distribution of the artificial NO (or NO_2 and HNO_3 after conversion) should be related to the specific location of the nuclear tests areas. Therefore, before mixing processes have spread out the injected molecules in the whole hemisphere, most of the anthropogenic NO_x is expected to be located in the high latitude regions.

TABLE 1 : Dominant reactions for high-temperature air dissociation.

$$O_2 + M \rightleftharpoons O + O + M$$
$$N_2 + M \rightleftharpoons N + N + M$$
$$NO + M \rightleftharpoons N + O + M$$
$$O + N_2 \rightleftharpoons NO + N$$
$$N + O_2 \rightleftharpoons NO + O$$
$$N_2O + O \rightleftharpoons NO + NO$$

ANALYSIS OF THE OZONE RECORDS IN RELATION WITH THE THERMONUCLEAR EXPLOSIONS

Before the International Geophysical Year (1957-1958) only a very few stations have provided long term observations of total atmospheric ozone. For example, the station at Arosa, Switzerland ($\varphi = 46.46^0$ N, $\lambda = 9^0.40$ E) has reported fairly continuous data since 1932. Their analysis provides some information concerning the variation of ozone over a specific location during half a century. On the other hand, the data reported by the ozone world network operating since 1957 (ozone data for the world, Meteorological Branch, Dept. of Transportation. Canada) give indications concerning the behavior of the global ozone around the Earth but only during a short period of time. The two different aspects will be examined here.

Total ozone has been recorded over Arosa by a Dobson instrument. Like most meteorological data, it shows variability on a variety of scales of time. In order to remove the short term fluctuations, the monthly and yearly mean values have been calculated (fig. 1 and 2), Fig. 1 clearly shows the well-known annual variation which also appears to be superimposed by long term trends. Fig. 2 indicates that the annual mean can markedly differ from one year to the other. For example, the averaged values of total ozone was equal to 328 Dobson units (corresponding a height of 3.28 mm of ozone at STP conditions) in 1939 and 361 Dobson units in 1940, which corresponds to an increase of 10 percent in one year.

In order to make the long term variations more apparent, a running mean of the data, represented by fig. 1, has been calculated. The result is represented by fig. 3 where a four year period has been chosen over which the running mean is computed. It appears that the depletion of the ozone amount occuring after 1960 is not more significant than the other variations appearing in the 1940's and the 1950's. A regular variation which seems to be related to the undecennal solar cycle (Angell and Korshover, 1973; Ruderman and Chamberlain, 1975) is also apparent. In fact a large part of the ozone decrease observed at Arosa during the nuclear test series could be attributed to the reduction of the solar activity.

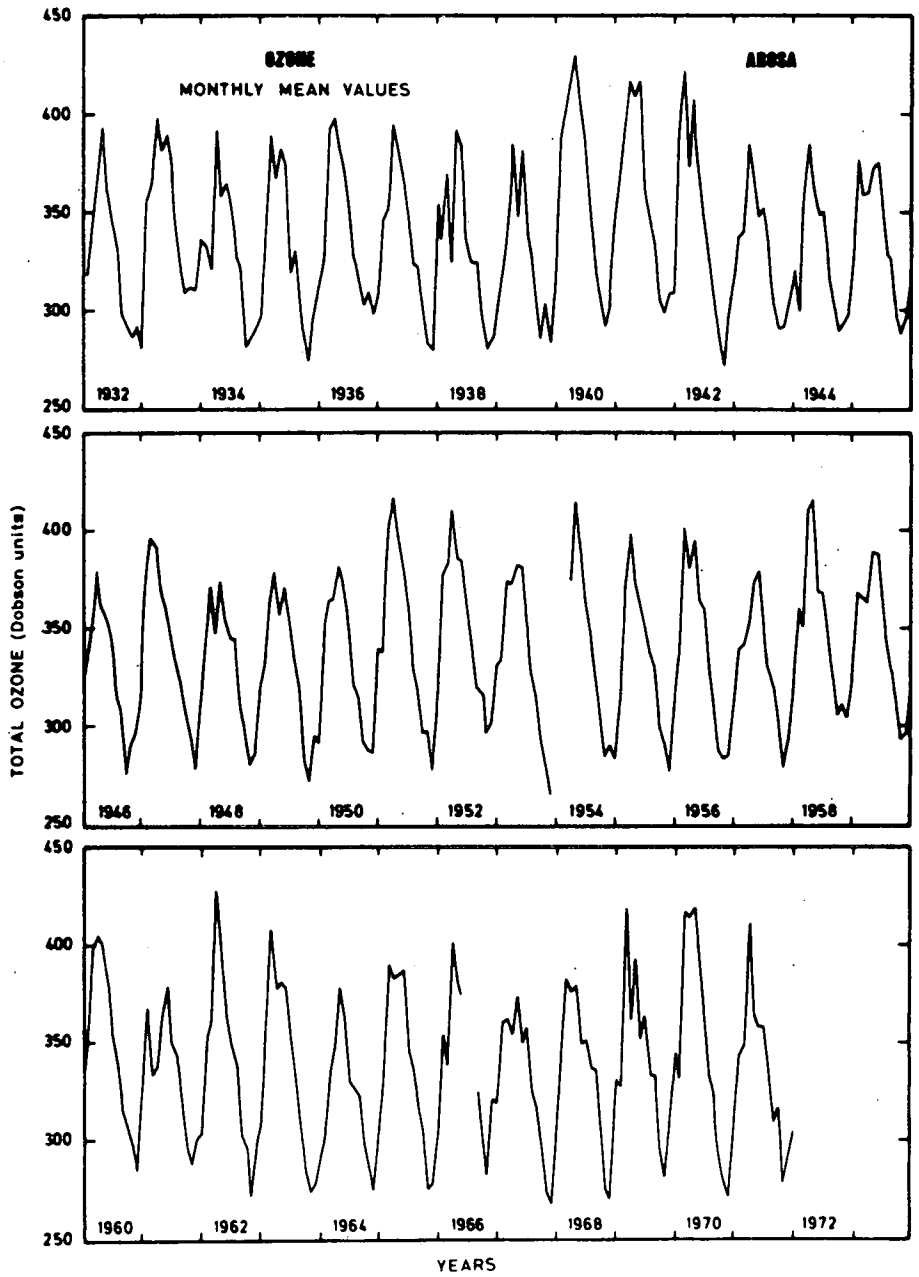

Fig. 1.- Monthly mean values of total ozone between 1932 and 1971 over Arosa (Switzerland).

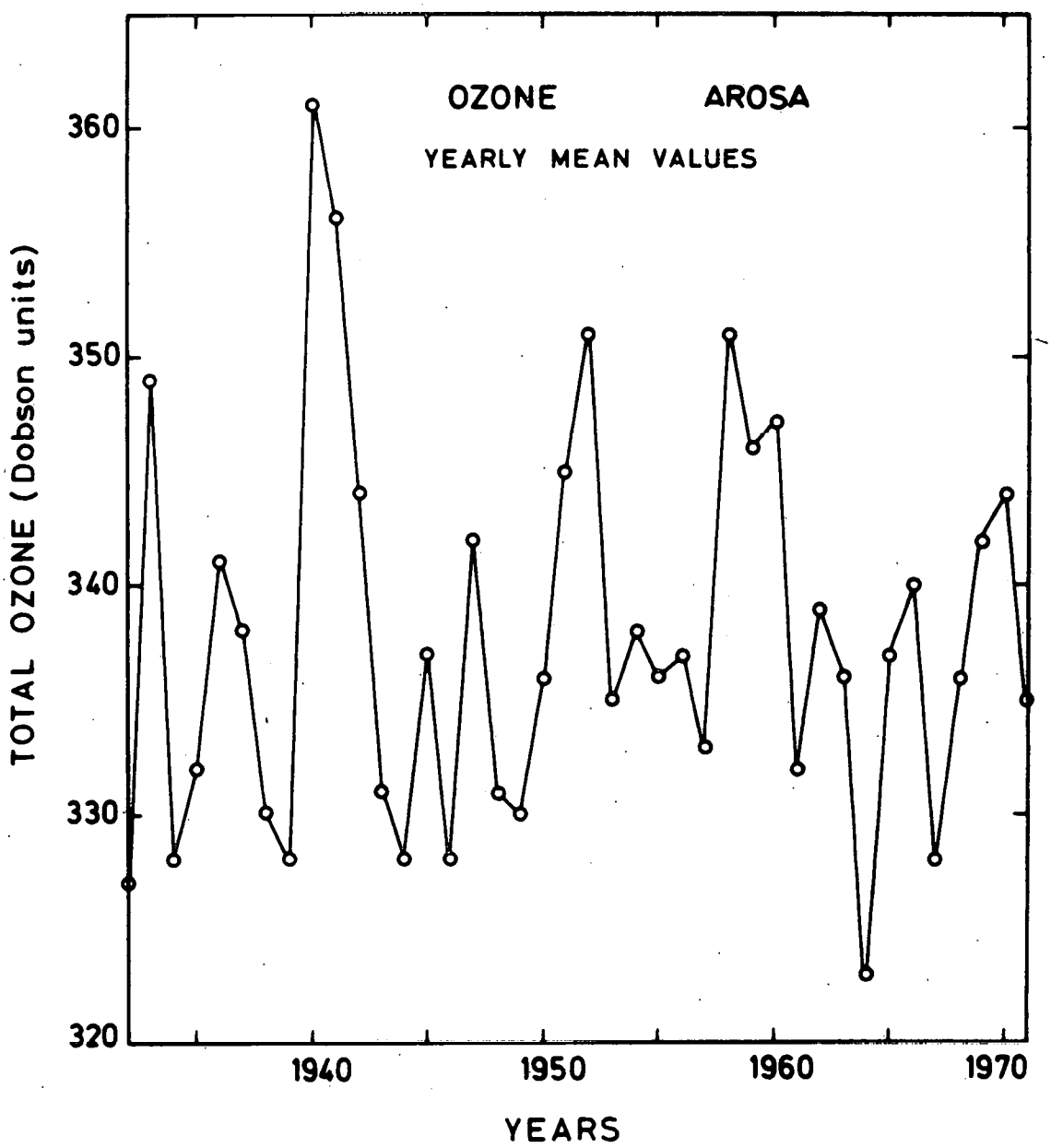

Fig. 2.- Yearly mean values of total ozone between 1932 and 1971 over Arosa (Switzerland).

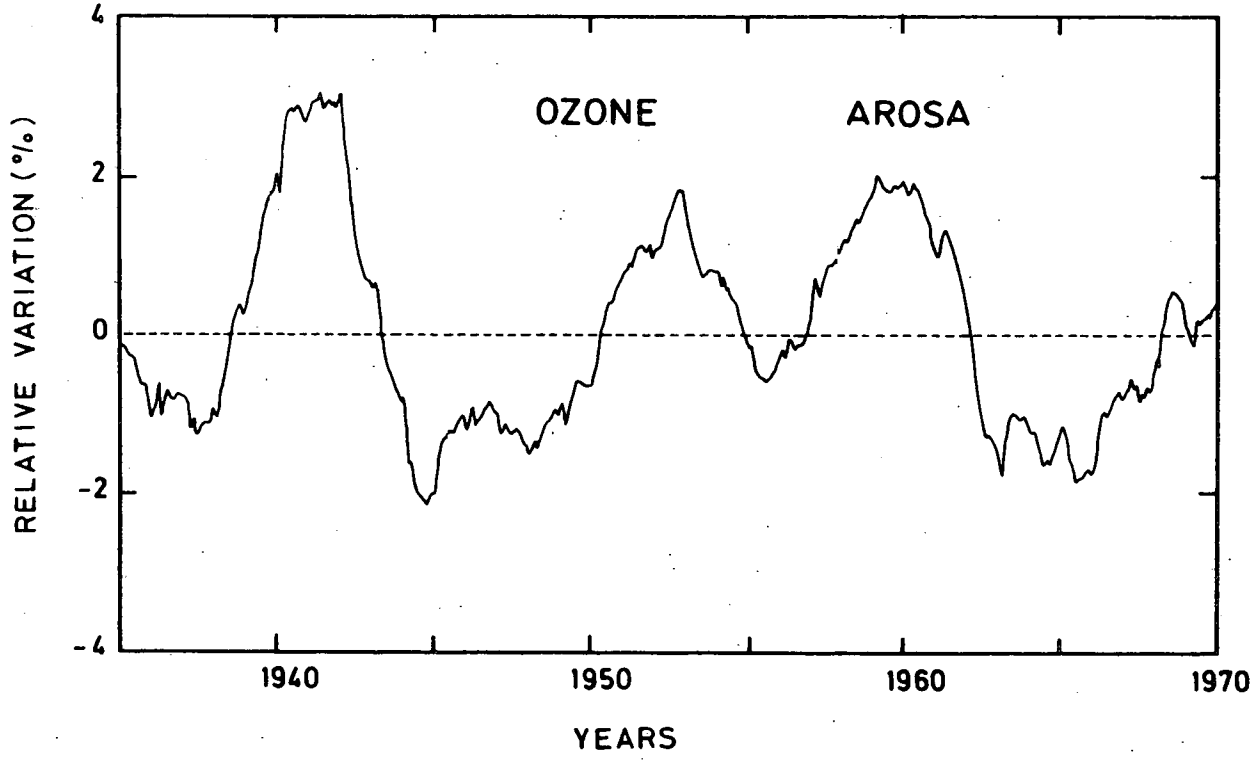

Fig. 3.- Four years running mean of the relative variation of total ozone over Arosa between 1934 and 1969.

In order to analyse the behavior of ozone on a planetary scale, 38 stations (Table 2) reporting systematic observations in the 1960's have been selected. Thirty of them are located in the northern hemisphere with a high proportion in the mid-latitude regions. The averaged number of reported monthly mean values by an individual station is equal to 126 during a period of observation of 162 months. Since it is necessary to avoid assigning a disproportionate weight to some stations according to their location on the Earth's surface, the contribution of the data collected at the 4 different latitudinal zones, namely the 0-30° N, 30°-60° N, 60°-90° N regions and the whole southern hemisphere, have been treated separately. In each of these latitudinal bands, an effective evolution of the ozone amount is derived by determining, for each month, the averaged value of the monthly mean values proper to each station included in this band. When, for a specific station, a monthly mean value is not available, it is replaced by a value obtained by averaging, for that month, the corresponding monthly means over the period 1957-1970. Fig. 4 shows the temporal evolution of the effective integrated ozone concentration in the three Northern hemispheric regions and in the Southern hemisphere. The well known characteristics of the behavior of O_3 are obvious on this graph : a larger amount of ozone and larger annual variations appear at higher latitude.

Since our purpose is to isolate a possible effect due to the thermonuclear explosions in the beginning of the 1960's, the most important natural variations have been filtered out of the data. The day-to-day fluctuations related to the meteorological situation do not appear since the data which are used represents monthly averages of observations carried out at different geographic locations. The annual variation, which is of no interest in this analysis, can be extracted by determining a running mean of the data over 12 months or a multiple of this period. We have plotted the 3 year running mean on Fig. 5 which shows only the long term trends of the total ozone. This technique also eliminates partially the quasi biennial oscillation in the ozone content.

In order to analyse these variations let us first consider the case of the mid-latitude regions. The evolution of the quantity of ozone appears to be remarkably constant in the first half of the 1960's and a rather sharp positive trend can be observed at the end of the decade. Actually no feature appears in the data that can be attributed to the thermonuclear explosions of 1961-1962.

TABLE 2 : Observation stations considered for the study of the ozone variations.

A. Northern Hemisphere : High latitudes φ = 60° - 90° N

N°	Station	Latitude	Longitude
1	Resolute	74.43 N	94.59 W
2	Dikson Island	73.30 N	80.14 E
3	Tromsø	69.39 N	18.57 E
4	Fairbanks	64.49 N	147.52 W
5	Reykjavik	64.08 N	21.54 W
6	Lerwick	60.08 N	1.11 W
7	Leningrad	59.58 N	30.18 E

B. Northern Hemisphere : Mid-latitudes φ = 30° - 60° N

N°	Station	Latitude	Longitude
8	Aarhus	56.10 N	10.13 E
9	Moscou	55.45 N	37.34 E
10	Edmonton	53.33 N	114.06 W
11	Irkutsk	52.16 N	104.21 E
12	Oxford	51.45 N	1.11 W
13	Kiev	50.24 N	30.27 E
14	Arosa	46.46 N	9.40 E
15	Toronto	43.43 N	79.14 W
16	Vladivostok	43.07 N	131.54 E
17	Sapporo	43.03 N	141.20 E
18	Alma Ata	43.14 N	76.56 E
19	Mont Louis	42.30 N	2.07 E
20	Vigna di Valle	42.05 N	12.13 E

TABLE 2 : (continued).

N°	Station	Latitude	Longitude
21	Boulder	40.01 N	105.15 W
22	Cagliari	39.15 N	9.03 E
23	Messina	38.12 N	15.33 E
24	Nashville	36.15 N	86.34 E
25	Tateno	36.03 N	140.08 E
26	Albuquerque	35.05 N	106.37 W
27	Kagoshima	31.38 N	130.36 E

C Northern Hemisphere : Low latitudes φ = 0° - 30° N

N°	Station	Latitude	Longitude
28	New Delhi	28.38 N	77.13 E
29	Dum Dum	22.39 N	88.27 E
30	Kodaikanal	10.14 N	77.28 E

D. Southern Hemisphere φ = 0° - 90° S

N°	Station	Latitude	Longitude
31	Huangayo	12.03 S	75.19 W
32	Pretoria	25.45 S	28.14 E
33	Brisbane	27.28 S	153.02 E
34	Aspendale	38.02 S	145.06 E
35	Port aux Français	49.21 S	70.17 E
36	Halley Bay	75.31 S	26.44 W
37	Byrd	80.01 S	119.31 W
38	Amundson	89.59 S	24.48 W

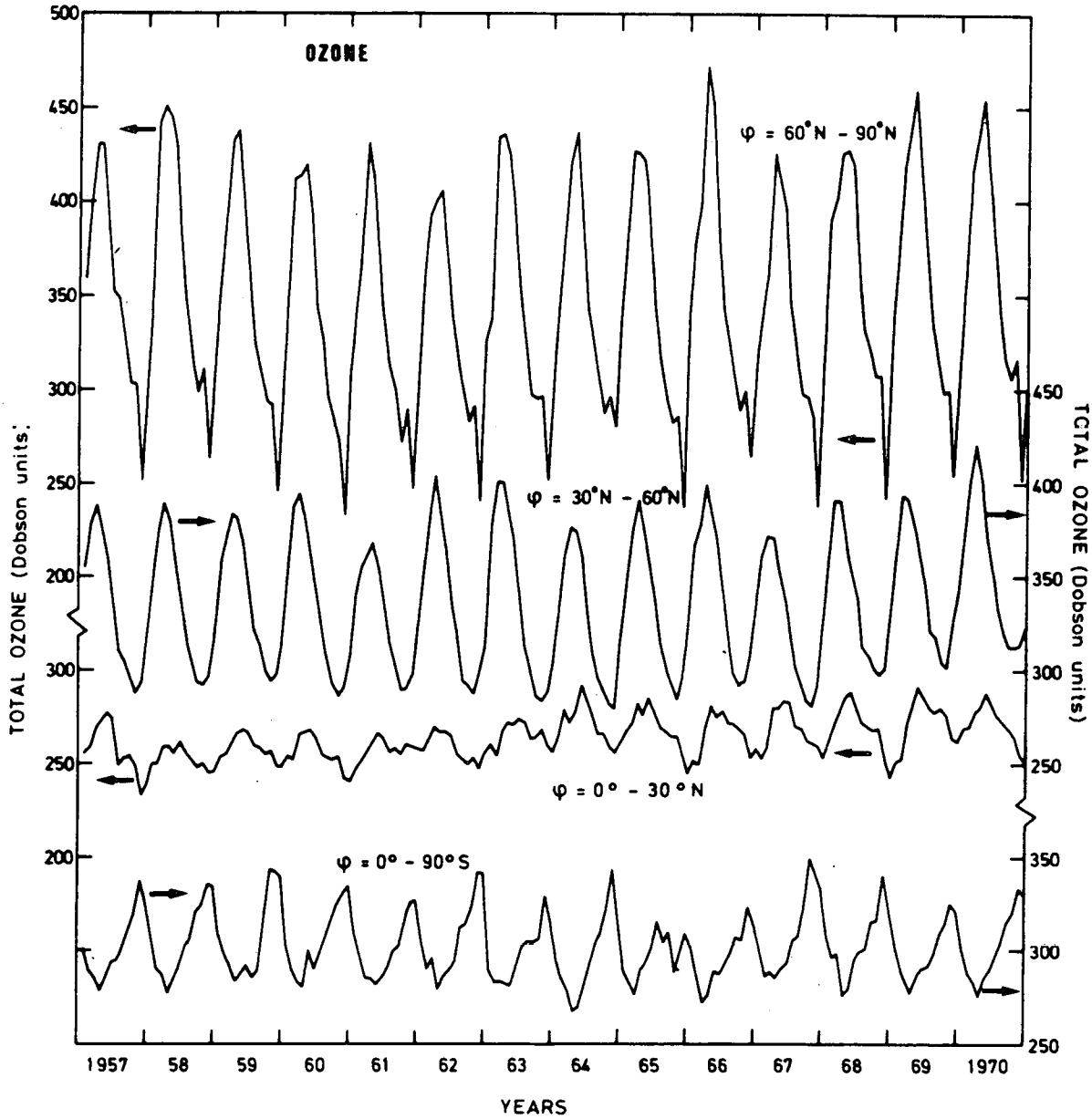

Fig. 4.- Variation of the averaged total ozone between 1957 and 1970 in four latitudinal regions. See text for explanation.

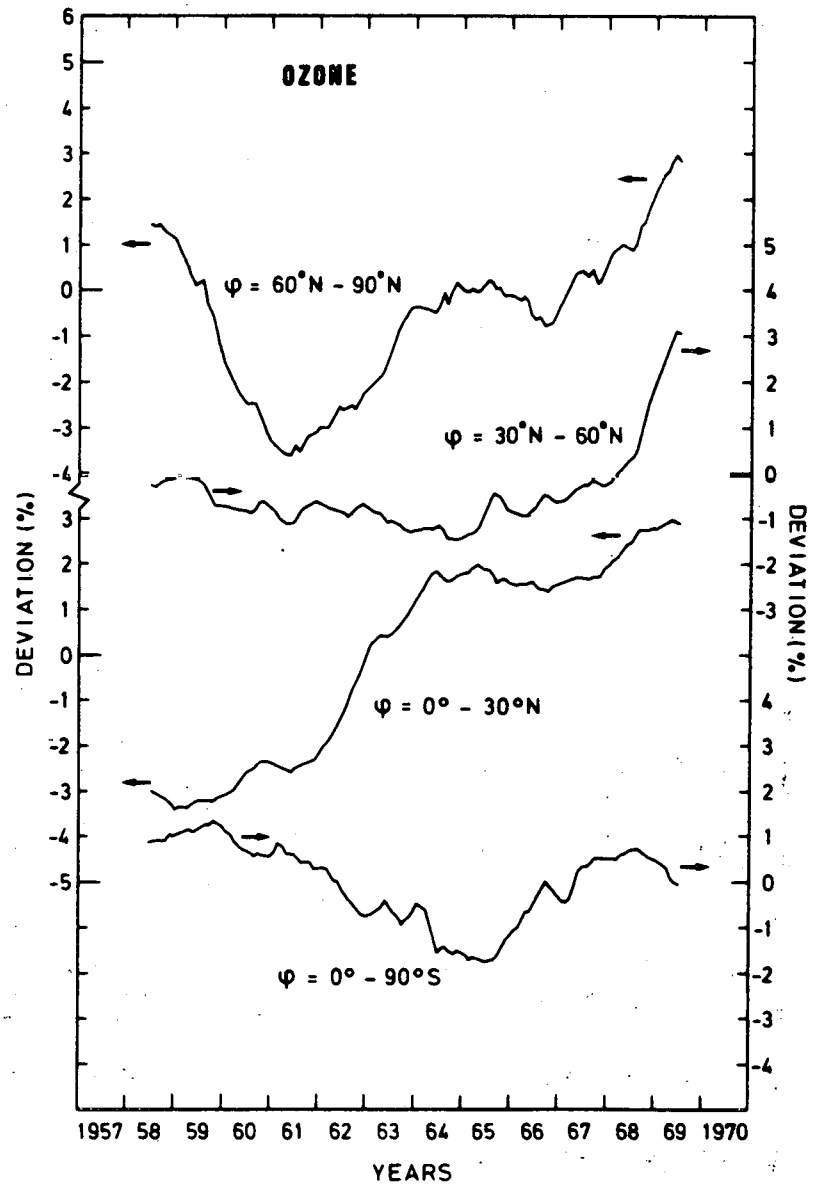

Fig. 5.- 36 months running mean of the relative variation of total ozone during the 60's in 4 latitudinal regions.

In the equatorial and tropical regions where the total amount of energy released in the atmosphere by nuclear blasts in 1962, corresponds to 37 Mt, a rather marked bulge, which lasts about 5 years, appears after the nuclear tests. Moreover it is superimposed on a general positive trend characterizing the 1958-1970 period. It is not quite clear if this bulge can be related to the explosions but again no destruction of ozone is evident.

The examination of the data obtained in the high latitude regions should lead to the clearest conclusions since most of the anthropogenic NO should have been injected in the polar regions. However it should be noted that the most of the USSR explosions took place during or just before the winter season (polar night) when the chemistry is not very efficient. Nevertheless the residence time of the injected molecules is larger than the winter period and an effect of NO on the ozone amount should be expected according to the theory. The curve on Fig. 5, corresponding to the polar regions, shows a rather rapid depletion at the end of the 1950's and an inversion of this trend after the nuclear test series. Again, the atmospheric explosions seem to be followed by the appearence of an ozone bulge which also lasts about 5 years and which is also superimposed on a general upward trend. It should be noted that most of the data in the high latitudes (Soviet Union) are provided by M 83 filter instruments whose precision is much weaker (Bojkov, 1969) than the accuracy of the Dobson instruments which are used in most parts of the world.

Finally, the only slight ozone depletion appears to be observed (Fig. 5) in the Southern hemisphere where no nuclear explosion took place during the period discussed. But the amount of data is not sufficient to derive any definitive conclusion.

CONCLUSIONS

To summarize, it seems difficult to derive from the analysis of the data provided by a specific station like Arosa or from a large amount of observations recorded all over the world any significant change in the ozone amount. The ground base data available during the 1957-1970 period is not homogenous enough nor large enough to provide an unambiguous interpretation and, in particular, is not enough to validate the efficiency of the catalytical destruction of ozone by NO_x. As the photochemical theory shows, the action of

nitrogen oxides on ozone is especially important in the middle and the upper stratosphere. In the vicinity of the tropopause where NO is injected, the transport plays a major role and makes the analysis very complicated. On the other hand the effect of an ozone depletion by the Crutzen catalytical cycle should compete with the following chemical chain leading to the formation of ozone in the daytime troposphere and lower stratosphere (Crutzen, 1973, Chameides and Walker, 1973, Nicolet, 1975)

$$\begin{aligned} NO + RO_2 &\rightarrow NO_2 + RO \\ NO_2 + h\nu &\rightarrow NO + O \\ O + O_2 + M &\rightarrow O_3 + M \\ \hline O_2 + RO_2 &\rightarrow O_3 + RO \end{aligned}$$

where R is a radical (e.g. H or CH_3). If the bulge observed in the data discussed previously is statisticly significant, it could be attributed to such a mechanism. In fact, this photochemical scheme, which was not taken into account in the model simulation by Chang *et al* (1973) and which was not considered in the analysis by Foley and Ruderman (1972, 1973) or Johnston *et al* (1973) could mask the expected depletion of total ozone by nitrogen oxides. This mechanism should be more efficient in the tropical and equatorial regions where the solar flux is more intense but it should not be rejected during the summer in the polar regions because of the much higher injection of NO_x in the high latitudes.

Finally further studies are needed before the action of nitrogen oxides on ozone in the real atmosphere can be completly understood. The use of satellites with their detailed global coverage will provide vital information on the behavior of ozone after future nuclear tests or at the occasion of PCA events (Crutzen, Isaksen and Reid, 1975) which should be responsible of odd nitrogen formation in the upper stratosphere.

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