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The Effect of Nuclear Explosions on Stratospheric Nitric Oxide and Ozone

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Abstract

This article reviews the derivation by Foley and Ruderman of the injection of nitric oxide into the stratosphere by nuclear bomb tests and compares it with other similar studies. Upper and lower limits of this pollutant are estimated by us and compared with the amount and distribution of nitric oxide in the stratosphere possible from supersonic transports. The effect on ozone of any artificial nitric oxide in the stratosphere depends on the distribution as well as the quantity. The distribution of ^{90}Sr in the stratosphere was measured by balloons and planes after the 1961-62 nuclear tests, and there is a linear relation (with known proportionality constants) between bomb-produced ^{90}Sr and bomb-produced nitric oxide. In this way the actual distribution (within the six-fold range of uncertainty that connects NO formation to bomb yield) of NO in the stratosphere is presented. Most of this bomb-produced NO lay low in the stratosphere and far to the north such that the maximum expected ozone reduction would be matter of a few percent. The total-ozone data for the world for 1960-70 inclusive have been examined in detail. There appears to be a real (about 5%) increase of ozone over the period 1963-70; any other trends appear to be lost in fluctuations of the data and to be unobtainable because of the few ozone-observing stations before 1957. The increase in ozone 1963-70 is roughly parallel to the decrease of bomb-produced ^{90}Sr and thus bomb-produced NO; and hence this increase of ozone may be the stratosphere returning to normal after the nitric oxide injections by the nuclear bomb tests of 1952-62.

INTRODUCTION

Foley and Ruderman (1973) examined the question of the formation of nitric oxide from air heated by nuclear explosions during the period of large scale atmospheric testing, 1952-1962. They evaluated the upper and lower limits of the nitric oxide injection into the stratosphere and compared the upper limit to the production of NO by supersonic transports (SST). Foley and Ruderman examined the ozone data for the world during the period 1960-65, and they found no large reductions of ozone that correlated with the nuclear explosions. They did not look for small effects.

In this article we review the derivation of the rate of formation of NO from nuclear explosions. We agree with Foley and Ruderman's upper limit, but we disagree with their estimated lower limit. There is a linear relation with known coefficients between strontium-90 produced by the bombs and the nitric oxide (within the range of lower-to-upper limit) produced by the bombs. The observed distributions of ^{90}Sr in units of radioactive disintegrations per thousand standard cubic feet of sampled air can thus be directly translated into mole fractions of bomb-added nitrogen oxides in the stratosphere. Most of the added nitrogen oxide was at low elevations and in the far northern stratosphere where ozone photochemistry is not important, and the maximum expected effect is a matter of a few percent reduction of ozone, spread out over a number of years. The ozone data of the world are subject to daily variations and to seasonal, biennial, and perhaps 11 year cycles, and it would require 10 years of daily

observations to establish to the 95% confidence level a systematic 5% reduction of ozone (Pittock, 1972). The expected small changes of ozone may be lost in the noisy data. Careful analysis of 10 years of daily ozone observations show a systematic increase of about 5% in the northern hemisphere between 1963-197. The timing and distribution of ^{90}Sr in the stratosphere and our calculations estimating the effect of the related nitrogen oxides from nuclear bombs are consistent with these ozone data.

Although the large nitric oxide injections in the stratosphere from the nuclear bomb tests did not attain a distribution conducive to large ozone reductions, the long-term, steady flights of 500 American SST could establish a different distribution capable of much larger reduction of ozone.

THERMODYNAMICS

Foley and Ruderman (1973) pointed out that the detailed, quantitative study of the nuclear fireball by Brode (1968) can be expressed in relatively simple terms: one third of the bomb energy goes to radiation transmitted by air, one third of the energy is carried away as the directed kinetic energy of a strong shock wave, and one third remains in the fireball as thermal energy. The initial nuclear explosion is at temperatures far above 10^5°K, and the black-body emission of radiation is largely in the region of soft X-rays, which are strongly absorbed by air to produce high temperatures. The fireball grows by emission and absorption of short wave-length radiation, and it cools by emission of radiation

weakly absorbed by air, that above about 185 nm. A strong shock wave forms which both heats the air near the explosion and which carries away directed kinetic energy in the air. Within a few seconds the shock wave has carried away about one-third of the energy. The residual fireball at an average temperature of about 6000°K contains one-third of the energy. The equilibrium distribution of the components of air at several temperatures between 298 and 6000°K is given in Table 1 (derived from Gaydon and Hurle, 1963; and JANAF Thermochemical Tables). At one atmosphere pressure and 6000°K oxygen (and water, which is not included in Table 1), is almost completely dissociated, nitrogen is about 4 per cent dissociated, and the equilibrium concentration of nitric oxide is just above one per cent. The enthalpy function, $H^\circ_T - H^\circ_{298}$, in kcal mole^{-1} and the enthalpy including the standard heat of formation, $\Delta_f H^\circ_{298}$, is given for each component of air in Table 1. The sum of $\Delta_f H^\circ_{298}$ and $H^\circ_T - H^\circ_{298}$ over all species gives the total enthalpy of air; this total enthalpy is the energy required to heat one mole of air from 298°K and one atmosphere pressure to the temperature T, including the energy required to bring about the endothermic reactions. Heat capacities and the heat capacity ratio γ are included in Table 1.

Glasstone (1964) gives the energy of the nuclear bomb as

$$E_{total} = 10^{12} \text{ kcal } Y_{MT}$$

If one third of the energy goes to form hot air at 6000°K, the yield of hot air is this energy divided by the total enthalpy of air (Table 1)

Table 1. Thermodynamic properties of air between 298°K and 6000°K. Enthalpy functions refer to the quantity of material in one mole of air at 298°K.

T °K	298	2000	3000	4000	5000	6000
A. Relative composition (approx. 1 atm total pressure)						
N_2	0.78	0.78	0.76	0.75	0.75	0.735
O_2	0.21	0.21	0.165	0.038	0.0036	4×10^{-4}
NO	-	0.008	0.044	0.052	0.026	0.014
Ar	0.01	0.01	0.01	0.01	0.01	0.01
O	-	3×10^{-4}	0.046	0.29	0.39	0.41
N	-	-	-	0.002	0.028	0.075
B. $H^\circ_T - H^\circ_{298}$, kcal mole^{-1}						
N_2	0	13.42	22.16	31.08	40.12	49.24
O_2	0	14.15	23.45	33.20	43.26	53.48
NO	0	13.84	22.72	31.77	40.92	50.19
Ar	0	8.45	13.43	18.38	23.34	28.30
O	0	8.45	13.52	18.57	23.72	28.98
N	0	8.45	13.44	18.53	23.92	29.79
C. $\Delta_f H^\circ_{298} + H^\circ_T - H^\circ_{298}$, kcal mole^{-1}						
NO	21.45	35.29	44.17	53.22	62.37	71.64
O	58.98	67.52	72.50	77.55	82.70	87.96
N	112.6	121.0	126.0	131.1	136.5	142.3
D. Total standard enthalpy of air, kcal mole^{-1}						
	0	13.8	26.1	50.3	68.2	84.2

Table 1. Continued

E. Heat capacities, cal $mole^{-1}$ deg^{-1}, and $\gamma = C_p/(C_p - R)$

	298	2000	3000	4000	5000	6000
$C_p(N_2)$	6.96	8.601	8.86			
$C_p(O_2)$	7.02	9.029	9.55			
$\gamma(N_2)$	1.40	1.300	1.30			
$\gamma(O_2)$	1.40	1.282	1.27			

$$n\ (\text{air, } 6000°\text{K}) = (10^{12}\ Y_{MT}/3\times 84.2)\ \text{moles}$$

$$N\ (\text{air, } 6000°\text{K}) = 2.38\times 10^{33}\ Y_{MT}\ \text{molecules} \qquad (1)$$

As this air is cooled by adiabatic expansion, further radiative cooling, and a mixture with ambient air, it is expected (Foley and Ruderman) to have frozen-in a nitric oxide composition corresponding to equilibrium at 2000°K, which is a mole fraction of 0.008. The temperature of "frozen equilibrium" depends on the cooling rate, and the cooling rate of the rising fireball corresponds to a freeze-out temperature of 2000°K. The air in the original fireball at 6000°K and cooled down (at the appropriate rate) is sure to form about 0.008 mole fraction of nitric oxide, that is, the minimum production of nitric oxide is

$$N_{NO}(\text{minimum}) = (0.008)(2.38\times 10^{32}\ Y_{MT})$$

$$= 0.19\times 10^{32}\ Y_{MT}\ \text{molecules} \qquad (2)$$

CHEMICAL KINETICS

There is an additional thermal source of nitric oxide from the rising fireball. Ambient air mixed into the hot fireball will be heated up as the fireball itself is cooled down. The air outside the primary fireball is heated up and then cooled down shortly afterwards. The production of nitric oxide is kinetically controlled by a process with a very high activation energy, and it is enormously sensitive to the precise history of temperature and time. If the coolant air never reaches 2000°K (by virtue of a large ratio of cold air to hot air in the entrainment process),

there would be no nitric oxide production during the cooling process itself. On the other hand, if the cooling process occurs by stirring in relatively small amounts of outside air so that the temperature of all the coolant air exceeded 2000°K for a few tens of seconds, then the maximum amount of nitric oxide formation would occur. The two extreme cases are similar to the familiar chemical problem of mixing sulfuric acid with water or of mixing water with sulfuric acid; the temperature history of the water is remarkably different in the two cases.

The excess enthalpy $H^\circ_T - H^\circ_{298}$ of air including the energy of dissociation to form oxygen and nitrogen atoms, is 84.2 at 6000°K and 13.8 at 2000°K, Table 1. The ratio is 6.1. It thus requires 5 moles of air at 298°K to mix with one mole (29 grams) of air at 6000°K to produce 6 moles of air at 2000°K. The fireball must have risen to a substantial height in the atmosphere before cooling to 2000°K. There are small but significant contributions of adiabatic expansion to the cooling process, and the work of lifting the air in the earth's gravitational field contributes to the energy balance. These terms contribute approximately 15 per cent to the cooling of the fireball. The maximum thermal yield of nitric oxide is thus

$$N_{NO}\ (\text{maximum}) = 0.19(84.2/13.8)(0.85)\times 10^{32}\ Y_{MT}$$

$$= 0.99\times 10^{32}\ Y_{MT} \qquad (3)$$

Thus this analysis gives the range of production of nitric oxide from thermal sources as

$$N_{NO} = (0.19 \text{ to } 0.99)\times 10^{32}\ Y_{MT} \qquad (4)$$

Foley and Ruderman give the maximum yield of nitric oxide from the thermal source as 1.0×10^{32} Y_{MT} in agreement with our value. They give the minimum nitric oxide yield as a factor of three below the maximum value. Although the ratio of absolute temperature between 6000°K and 2000°K is a factor of three, the enthalpy content of air at 6000°K (high because of dissociated molecules) is 6.1 times that at 2000°K. Thus the range between maximum nitric oxide formation (mixing cold air with excess hot air) and minimum nitric oxide formation (mixing hot air with excess cold air) is a factor of 6, not 3.

An independent evaluation of the expected yield of nitric oxide from nuclear explosions was made in an unpublished Livermore Laboratory report by Bonner (1971)

> "I have made an estimate of the maximum credible production of NO_x. The reaction producing NO is $N_2 + O_2 = 2NO$. At room temperature the equilibrium is far to the left, i.e., N_2 and O_2 do not react. NO does not decompose into N_2 and O_2 at ordinary temperatures, however, because the reaction is very slow. In a situation such as a fireball where the temperature is changing fairly rapidly, the equilibrium tends to be frozen at about 2000°K. Above this temperature the system is in equilibrium, and below it the reaction rate is so slow that one can assume no reaction occurs.
>
> "I made the assumption that the 2000° equilibrium concentration exists throughout the volume of air which had been heated (by shock or otherwise) to at least

2000°. For 1 MT this corresponds to a radius of ∿ 3000 ft (H. Brode- Annual Reviews of Nuclear Science, 1968).

"At 2000°K the equilibrium concentration of NO in air is .007 mole fraction..."

This analysis gives

$$N_{NO} = 0.90\times10^{32}\ Y_{MT} \text{ molecules} \tag{5}$$

as the "maximum ... yield" of nitric oxide from air which was heated "by shock or otherwise" by the nuclear bombs. This figure is in excellent agreement with that of Foley and Ruderman (1.0×10^{32}) and the thermodynamic analysis (0.99×10^{32}) given above.

One of the mechanisms of nitric oxide formation discussed by Foley and Ruderman is the direct formation in the shock wave. They quoted a calculation based on 20 kilotons of TNT, and they scaled it linearly to megaton bombs. It should be pointed out that shock waves are highly non-linear phenomena and simple scaling factors are inadequate in some cases. In particular, strong shock waves are strongly damped by the endothermic dissociation of the molecules of air. A major fraction of the energy of 6000° air is in the form of dissociation of O_2 and N_2. Realistic considerations of shock waves in air must consider the interaction of endothermic chemistry with the gas dynamics of the shock waves (Gaydon and Hurle, 1963). These are very large effects and can change NO production in the shock wave by several orders of magnitude.

There is a factor that acts to reduce the amount of NO_x actually introduced into the stratosphere by nuclear bombs.

The initial fireball at 6000°K is diluted a factor of 6 (compare Table 1) in cooling to 2000°K, and it is diluted a further factor of 1700 (Bonner, 1971) in rising into the stratosphere. After cooling to 2000°K, the cloud continues to entrain large volumes of tropospheric air. The rising bomb containing moist tropospheric air eventually cools to stratospheric temperatures. The bomb cloud acts like a thunder-storm cloud, and copious quantities of water condense out and much of it precipitates out of the lower stratosphere and into the troposphere (Glasstone, 1964). The oxides of nitrogen, especially NO_2, coprecipitated NO and NO_2, and HNO_3, are highly soluble in water, and these would be expected to be washed out of the stratosphere more strongly than ^{90}Sr, ^{14}C, or tritium (that is, the highly soluble forms of NO_x would be more thoroughly rained out than water itself).

The injection of nitric oxide into the stratosphere is a much more complex problem than indicated above. The nature of some of these complications is given by further quotations from the memorandum by Bonner (1971):

> "...the only real data that emerged were the NRL spectral measurements on fireballs in the 1953 series at NTS. These were summarized in LAMS 1935 by DeWitt.
>
> "1) In LAMS 1935, DeWitt summarizes the data on NO_x. The presence of NO is not observed, but NO_2 and HNO_2 are observed. (The absence of NO is perhaps not surprising since there is about 20 times more ozone produced than NO_2. Ozone reacts rapidly with NO, so the presence of ozone almost automatically rules out NO.)

"For several shots of about 10 kt the light absorption by NO_2 bands reached a maximum at about the time of the temperature minimum. The amount of NO_2 was equivalent to roughly a 0.4 mm column of NO_2 at standard conditions (1 atm, 0°C)."

This quantity of NO_x observed spectroscopically is a factor of 8000 less than that calculated by Foley and Ruderman. It appears that these observations discussed by Bonner (1971) represent the NO_x produced by the shock wave, not by freeze-out cooling of the rising fireball. The time of observation of maximum NO_x appears to be after the shock wave has moved away but before the fireball has undergone significant mixing by air entrainment. The endothermic dissociation of air (including water) acts to reduce the temperature rise of the shock wave, which is a relatively minor direct source of nitric oxide from the nuclear bombs.

Foley and Ruderman discuss the role of residual radioactivity from the bomb debris in forming and in destroying nitric oxide in the stratosphere. These effects are smaller than the thermal effects in the nitric oxide balance. Some ionic processes produce nitric oxide and some destroy nitric oxide. Although there are factors that tend to decrease the yield of NO_x (rainout, destruction by fission products), we regard it as a reasonable postulate that large nuclear bombs formed NO_x in the atmosphere more or less within the range

$$N_{NO} = (\tfrac{1}{6} \text{ to } 1)\times 10^{32}\ Y_{MT} \qquad (6)$$

According to tables given by Foley and Ruderman, the total nuclear bomb yield from 1952 to 1962 was 513 MT. Thus the total number of molecules of nitric oxide produced by nuclear bombs between 1952 and 1962 is expected to be between the limits

$$N_{NO} = (0.8 - 5.1)\times 10^{34} \text{ molecules} \tag{7}$$

One American SST operating normally for one year would produce 4.8×10^{31} molecules of NO. Thus the decade of bomb testing had the average effect equivalent to the number of SST

$$N_{SST} \text{ (10 year average)} = 18 \text{ to } 103 \tag{8}$$

Alternatively it is as if SST flights had started in 1952 and increased linearly until 1962 to reach the total

$$N_{SST} \text{ (1962 max.)} = 36 \text{ to } 206 \tag{9}$$

and then all flights stopped. (The Concorde uses about one-third as much fuel as the proposed American SST, and thus these figures should be multiplied by three for comparison with the Concorde). In further discussions, these limits of Equation 6 are referred to as the "minimum" and "maximum" yields; it is recognized that these are not absolute limits. This range of values gives reasonable upper and lower limits, which should be examined for observable consequencies on the ozone of the stratosphere. One should consider separately the short-term, local effects and the long-term, global effects.

SHORT-TERM, LOCAL EFFECTS

Foley and Ruderman give equations for calculating the expected height and size of the cloud following nuclear bomb tests in tropical areas. For a time 30 minutes after detonation, the cloud sizes for 0.5, 5.0, and 50 megaton (MT) explosions are indicated by Figure 1, which includes a standard ozone profile at 45° latitude. The overlap of the ozone profile and the stabilized bomb cloud is strongly dependent on the yield of the bomb. For a standard ozone profile at the equator, this overlap is indicated by cross hatches in Figure 2. A bomb of 0.2 MT remains essentially within the tropical troposphere. A 1 MT bomb overlaps the ozone profile in the lower stratosphere, but it does not reach the level of maximum ozone concentration. A 10 MT bomb overlaps the maximum ozone. A 58 MT bomb, on the other hand, goes far above the ozone maximum, and it overlaps only a relatively small band of ozone. For short-term effects in the equatorial band, the maximum ozone reduction should be given by a 10 MT bomb, where a 65 percent overlap of the ozone column is indicated. With consideration of the finite rate of the catalytic destruction of ozone by NO_x, one could expect approximately a 30 to 50 per cent reduction of ozone within the bomb cloud during the first week or two of its existence. There would be a smaller effect for both larger and smaller bombs.

As the fireball cooled by radiation-emission, that radiation between 185 and 242 nm would dissociate oxygen and make ozone in a broad region surrounding the point of detonation. For bombs

fired well above the ground, this mechanism would form some ozone directly in the stratosphere, and for large bombs the artificial ozone layer would lie under the stabilized cloud. This layer of ozone would act to confuse the information to be expected on the ground directly under a bomb cloud during its first few days of existence. We carried out a numerical integration of the distribution of radiation from the cooling fireball of a 20 kiloton bomb, and the energy emitted as radiation between 186 and 242 nm was about one percent of the total energy of the bomb. We assume that this quantity scales linearly with bomb yield. In that case, the amount of ozone generated from ultraviolet radiation in the Herzberg continuum would be

$$N_{ozone} \approx 3\times10^{31}\ y_{MT} \tag{10}$$

Thus about one third as many molecules of ozone are produced as the upper limit estimate of nitric oxide production. This ozone would not make a significant contribution to the total global amount, but it is 40 percent of the ozone column subtended by a 30 minute old bomb cloud. Thus the ozone directly formed by radiation from the fireball is comparable to the maximum amount of ozone reduction expected in the newly stablized bomb cloud.

The principal USSR atmospheric tests were at Novaya Zemlya at 75°N. The largest tests were in late fall and winter so that there was little or no sunlight to support the photochemical reactions that destroy ozone. Ozone in the polar regions is formed in tropical and temperate zones and brought there by transport. Thus the ozone reductions from the big USSR tests are

expected to be delayed until high latitude NO has time to be moved into the temperate zones and the ozone formed there moved back to the polar region at a lower elevation.

The size of a 30 minute old bomb cloud is only about 10^{-4} the area of the earth. Thus the chance of a given ozone station observing a young cloud is small. The ozone collar photochemically produced by the bomb itself would confuse the information anyhow. Winds would be expected to shear the bomb cloud and move sheets of it in different directions at different elevations. Once the cloud is sheared and spread around the world at one latitude, the concentration of NO is reduced to such values that the effect of NO reduction of ozone is slow (months). Thus one should look for long term global effects of the nuclear bomb tests on stratospheric ozone, not for short-term local effects.

LONG TERM GLOBAL EFFECTS

When the nitric oxide yield equation, $(\frac{1}{6}$ to $1)\times10^{32}\ Y_{MT}$, is multiplied by the total explosion yields, especially over the periods 1952-54, 1957-58, and 1961-62, one obtains large values for the quantity of nitric oxide produced by nuclear explosives and injected into the stratosphere, see Foley and Ruderman's Table 3 for the upper limit values. However, the <u>distribution</u> of NO_x in the stratosphere is as important a variable as the <u>quantity</u> of NO_x there (Johnston, 1971). For example, it was shown (Johnston, 1971) that with a natural, non-uniform background of 8.6×10^{34} molecules of NO_x in the stratosphere, the addition of 4.4×10^{34}

molecules of artificial NO_x from supersonic transports could reduce a local (45° latitude) ozone shield anywhere between 3% and 50%, depending on the distribution of the added NO_x. Foley and Ruderman gave 3×10^{34} molecules of NO_x as the upper limit of the artificial increment of NO_x from the 1961-62 nuclear tests, but they only looked for large effects on the ozone shield. As noted (Johnston, 1971), the addition of 4.4×10^{34} molecules of NO_x could reduce the ozone shield by only 3% if it was distributed in one particular way. For the nitric oxide from nuclear bombs, it is necessary to consider the distribution of NO_x, not just the amount.

One aspect of the distribution is the elevation of the bomb debris in the stratosphere. The catalytic destruction of ozone by nitric oxide

$$\begin{array}{l} NO + O_3 \rightarrow NO_2 + O_2 \\ NO_2 + O \rightarrow NO + O_2 \\ \hline \text{net: } O + O_3 \rightarrow O_2 + O_2 \end{array} \qquad (11)$$

is a photochemical process; the formation of oxygen atoms is driven by solar radiation. The average concentration of oxygen atoms at 45° latitude in the summer varies approximately as: 10^4 molecules cm^{-3} at 10 km; 10^5 at 15 km; 10^6 at 20 km; 10^7 at 26 km; 10^8 at 35 km. Thus, the catalytic rate is 10 times faster at 20 km than at 15 km by virtue of the different oxygen atom concentration. Thus being "in the stratosphere" is only half the problem; the spread over elevation is an essential part of the problem.

Another essential aspect of the distribution of bomb-produced NO_x in the stratosphere is the geographical location. Over 90 per cent of the 1961-62 series (in terms of bomb yield) was conducted at the station at Novaya Zemlya at 75°N latitude. Three-quarters of these tests were conducted in the months September through December, just before or during the dark arctic winter. The NO_x catalyzed destruction of ozone is a photochemical process, which of course is inoperative during the polar night. Telegadas and List (1969) noted that radioactive debris is much more rapidly swept out of polar regions than tropical regions of the stratosphere, and a substantial degree of removal of bomb debris and NO_x surely occurred between deposition in the fall and winter and the return of an active sun the next summer. The NO_x increment (0.5 to 3)$\times 10^{34}$ molecules from the 1961-62 tests must be examined in terms of its distribution (latitude and elevation) before any conclusion can be stated as to whether one should expect a large effect or a small effect on the ozone shield.

During and after the 1961-62 bomb series, it appears that no one analyzed the stratosphere for nitric oxide, but very extensive analyses were made of strontium-90, excess carbon-14, and other radioactive products from the bombs (Glasstone, 1964; Telegadas, 1967, 1971; Krey and Kajewski, 1971). According to Telegadas and List (1969), the particulate ^{90}Sr samples and the gaseous ^{14}C (excess) gave very nearly the same record of nuclear bomb clouds, and particulate settling was "insignificant on the time scale of a year or so". An extensive report summarizing much

of these data was prepared by Seitz, Davidson, Friend, and Feely (1968). This report, is quoted here to illustrate several important points:

> "During the High Altitude Sampling Program and Project Stardust it often has been observed that interceptions of radioactive debris from specific tests did not substantiate the existence of a simple relationship between distribution of the debris with height and yield...Debris from the highest yield test in the 1961 Soviet series was encountered in high concentrations at 20 km by mid-1962. There was no evidence from balloon sampling program results that debris from this test ever reached higher than about 24 km...The basic premise, therefore, in assigning a vertical distribution of material to the model injections, particularly the polar injections, was that radioactive material was where you found it, and probably never was in regions which were sampled without finding it".

The observed (Seitz et al, 1968) distributions of ^{90}Sr as a function of elevation and latitude for May-August 1963 and for Jan.-April 1964 are given by Figure 3. The contours are in units of radioactive decompositions per minute per thousand standard (STP) cubic feet of air. The May-August 1963 record represents the maximum spread of ^{90}Sr at north temperate and tropical regions. There is a one-to-one correspondence between ^{90}Sr dpm/1000 SCF and the mole fraction of NO_x. This correspondence arises as

follows: one megacurie (3.7×10^{16} radioactive disentegrations per second) of ^{90}Sr is produced by 10 megatons of nuclear fission. In the 1961-62 series the ratio of total bomb yield to fission yield was 3.3 (Seitz et al, 1968).

$$1\ \mathrm{MC}\ ^{90}\mathrm{Sr} \leftarrow 10\ Y^{f}_{MT} \leftarrow 33\ Y_{MT} \qquad (12)$$
$$\downarrow$$
$$(\tfrac{1}{6}\ \text{to}\ 1)\,33\times10^{32}\ N_{NO}$$

When numerical values are put in, one finds

$$500\ \mathrm{dpm}/1000\ \mathrm{SCF} = (\tfrac{1}{6}\ \text{to}\ 1)\ \mathrm{ppb\ NO} \qquad (13)$$

where ppb is parts per billion (10^9) by volume. Thus the contour of maximum ^{90}Sr in Figure 3 of 1500 dpm/1000 SCF corresponds to 0.5 to 3 ppb of NO_x, and the map of ^{90}Sr in Figure 3 can be translated to NO_x distributions.

Profiles of the maximum NO_x inferred in this way are given for 0, 30, and 60°N in Figure 4. It can be seen that very large amounts of NO_x lie at low elevations and near polar latitudes, both factors that minimize the effect of NO_x on ozone. It is especially interesting to compare these NO_x distributions with recently observed distributions of NO, NO_2, and HNO_3 in the stratosphere, Figure 5. The NO from the nuclear bomb would be in the form of NO, NO_2, and HNO_3, and thus the bomb yields of NO should be compared with the sum of NO, NO_2, and HNO_3. The maximum and minimum bomb-produced NO_x is compared with the sum of the average NO_x reported by Ackerman (1972) and the HNO_3 for November as reported by Murcray et al (1973) in Figure 6. The observed curve was extrapolated to low elevations at the value of

3 ppb, and it was extrapolated above 30 kilometers by fitting to the observed ozone profile there. The maximum 45°N distribution is well below the natural background.

With the NO_x background shown in Figure 6 we calculated the steady-state reduction of ozone by the NO_x increments from nuclear bombs as inferred from the measured ^{90}Sr. A set of 31 reactions including H_2O, $O(^1D)$, HO, HOO, and HNO_3 were used in this steady-state calculation. The shortcomings of the steady-state calculation are recognized, but in this application this method has the merit of probably providing an upper limit to the amount of reduction of ozone. The atmospheric motions that complicate the use of this method would probably act to reduce the calculated effect. The calculated steady-state profiles indicate that the minimum NO yield from bombs ($\frac{1}{6} \times 10^{32}$ Y_{MT}) would have produced no more than a 1.2 per cent reduction of the ozone column, and the maximum NO yield from bombs (1×10^{32} Y_{MT}) would produce a steady-state reduction of no more than 6.3 per cent.

In their examination of the ozone data of the world, Foley and Ruderman only looked for large effects, that is, effects directly and conspicuously larger than a five per cent change. On the basis of observed ^{90}Sr from the bombs, the linear relation expected between ^{90}Sr and N_{NO}, and a steady-state analysis based on the recently observed natural background of NO_x ($NO + NO_2 + HNO_3$), we find the effect to be expected from the 1961-62 injections of NO from nuclear bombs is a matter of 1 to 6% in the temperate zone of the northern hemisphere.

If 3×10^{34} molecules of NO artificially introduced by nuclear bombs are expected to reduce the ozone column by no more than 6 per cent, does this imply that artificial injections by supersonic transports are expected necessarily to have a small effect on ozone? We carried out the following computation (with the assistance of Dr. Purna Patniak) as the first of a series designed to give some insight into questions of this sort. We included O, H, and N chemistry with one dimensional vertical eddy diffusion. The natural NO_x in the stratosphere was generated by the nitrous oxide mechanism (Crutzen, 1971; McElroy and McConnell, 1971; Nicolet and Vergison, 1971), which was allowed to "operate" for 30 years. Then an artificial source of NO was added at 20 km. This artificial source corresponds to the normal operation of 500 American SST at 45° latitude with a local traffic density three times the world average. The American SST was assigned an emission index of 14 g NO/kg fuel. The computation represented 5 years of SST operation under the artificial condition of perpetual spring equinox. The distribution of solar radiation was recomputed every day at the beginning of the calculation and every 100 days as the distributions approached a steady state. The steady operation of the SST produced a distribution of artificial NO_x spread out over considerably higher elevations than was attained by the 1963 distribution of nuclear debris as is shown in Figure 7. The SST distribution of NO, NO_2, and HNO_3 gave a substantially larger reduction of ozone than was given by the distribution of NO, NO_2, and HNO_3 from the nuclear bomb tests.

Table 2. Calculated steady-state profiles of ozone without and with added NO_x from nuclear bombs, compare Figure 6.

Latitude	Natural	Min. bomb increment*	max. bomb increment*
A. Calculated ozone column in units of 10^{18} molecules cm^{-2}			
60°N	4.50	4.46	4.30
45°N	5.62	5.56	5.30
30°N	6.40	6.33	6.05
15°N	6.54	6.46	6.13
0°	6.22	6.18	6.00
B. Relative ozone column, per cent			
60°N	100	99	96
45°N	100	99	94
30°N	100	99	94
15°N	100	99	94
0°	100	99	96
C. Percentage reduction of ozone			
60°N		1.0	4.4
45°N		1.1	5.7
30°N		1.1	5.4
15°N		1.2	6.3
0°		0.6	3.5

* Minimum and maximum bomb increments are the limiting values of the relation.

$$N_{NO} = (\tfrac{1}{6} \text{ to } 1.0) \times 10^{32}\ Y_{MT} \text{ molecules.}$$

(This computation derived a vertical profile of NO_x that is only about half that of Figure 6, and thus direct comparisons between the reduction of ozone by the nuclear bombs and the SST cannot be made. However, we found with the one-dimensional vertical eddy diffusion model a 34% reduction of ozone by the SST flights as indicated by Figure 7. This figure is a preliminary one, and it merely illustrate that large fleets of SST could have a large effect on ozone when vertical diffusion and H, N, O chemistry are considered.)

The total stratospheric burden of ^{90}Sr and excess ^{14}C from 1950 to 1970 is given in Figure 8 (Glasstone, 1964; Telegadas, 1967, 1971; Krey and Kajewski, 1971). The timing and magnitude of nuclear bomb testing is indicated on the figure. The global radioactive debris in the stratosphere was high and irregular between 1954 and 1961. During the nuclear bomb testing moratorium during 1959-1961, there was only a slight decrease of ^{90}Sr or ^{14}C. The major test series in 1961-62 caused an increase of ^{14}C to a value in early 1963 that was three times the previous peak (1959) and five times the minimum of 1961. It was 1970 before excess ^{14}C returned to its 1961 value. The global inventory of ^{90}Sr behaved similarly to ^{14}C except that its long term decay was somewhat faster than that of ^{14}C. The long-term behavior of stratospheric excess NO_x from nuclear bombs, as inferred from Figure 8, is an irregular slow increase from 1952 to early 1963 and a regular slow decrease from 1963 to 1970. The slow decrease of the gaseous ^{14}C from 1963-70 indicates that any effect of artificial NO_x from the 1952-62 bombs would require approximately 10 years after cessation of large scale testing in 1962 to be eliminate

OZONE DATA FOR THE WORLD

There are about one hundred stations, distributed in an irregular pattern over the world, that make daily observations of the total ozone column, and these data have been published since 1960 as Ozone Data for the World (ODW) by the Canadian Department of Transport. Two such stations, Moscow and Messina, are given by Figure 9. Approximately two-thirds of the stations of the world use the Dobson ozone instrument, a double quartz prism monochromator that measures the ratio of sunlight at pairs of wavelengths; and one-third of the stations use the Gustin filter ozonometer that measures the ratio of sunlight through a pair of filters (Bojkov, 1969). The Dobson instrument has relatively narrow spread in each wavelength band, 9 to 19 Å, and a separation of 200 Å. The Gustin meter has a wider wavelength spread, 480 to 590 Å, and a separation between center of 550 Å. It has been found by an extended side-by-side comparison of a Gustin and a Dobson meter that the filter instrument reports ozone systematically higher (up to 20 to 30 percent) than the prism meter at low solar angles (winter) and lower (about 6 percent) for nearly overhead sun (summer). The filter meter overestimates ozone (relative to the Dobson meter) under hazy conditions. Also, on a side-by-side, minute-to-minute comparison the filter meter gave a standard deviation of readings about five times as great as the Dobson meter (Bojkov, 1969).

The data from Moscow in Figure 9 were taken by the Gustin instrument and that from Messina by the Dobson method. Both stations show the usual maximum of ozone in the late winter and

early spring months and the minimum of ozone in the autumn months. However, the amplitude between maximum and minimum is much greater for Moscow (filter instrument) than for Messina (Dobson instrument). We found the standard deviation of all of the daily readings from Moscow to be 15.0% about the current seasonal average and that for Messina to be 7.8%. For a given station the average readings and the average trends over long periods of time have a certain validity for each method, but these considerations indicate that the pooling of data from stations using the two methods presents difficulties.

At a given observing station of total ozone, there is a substantial variation from day to day as illustrated by data (Dobson instrument) from Aspendale, Australia, Figure 10. There are systematic, rather well understood annual trends, with a peak in the early spring and a minimum in the fall, illustrated both in Figures 9 and 10. There are "quasi-biennial" variations of temperature, tropopause height, and total ozone column at a given location (Ramanathan, 1963; Angell and Korshover, 1964) although the phases of these oscillations vary over the globe. The biennial oscillations were particularly strong at Aspendale (1955-1963, although they ceased after 1963). There is a long record of discussion about whether or not the ozone data show a correlation with the sunspot cycle (Paetzold, 1972; London and Oltans, 1972).

With a large day-to-day noise, a large seasonal variation, a sometimes-present-sometimes-absent biennial oscillation, and perhaps an eleven year cycle, it is extremely difficult to prove

or disprove a small systematic change of the ozone level, such as the few percent decrease expected from the nuclear bomb model (with maximum NO input) from 1952 to 1962 and the few per cent increase expected after 1963. Pittock (1971) analyzed the detailed data at Aspendale, and by using the two-sided Students' t-test he calculated how many years (N) of observations would be required to establish or disprove (at the 95 per cent confidence level) the occurence of a real trend of magnitude b per cent per decade. His results are reproduced as Figure 11. Note that if there were a real increase or decrease of ozone at the rate of 30% per decade, it would require three years of observation to establish the trend with a 95% confidence level if one has the noise spectrum given by the observed ozone data at Aspendale. An extrapolation of Pittock's curve indicates that it would take almost 2 years to establish the presence of a real trend of 10 per cent per year.

Foley and Ruderman presented graphs of total ozone from five stations for the period 1960 and 1964 with ODW, and they reported that they saw no large effects to be correlated with nuclear bomb tests. These graphs are analogous to those given by Figures 9 and 10, and any possible systematic trend is superimposed on the strong seasonal variations. Pittock showed that it requires a careful statistical analysis with five years of observation to establish a trend of 20 per cent per decade to the 95 per cent confidence level. A visual inspection of the raw data is indequate to detect small effects such as those expected from the bomb tests. It is necessary to factor out the seasonal variations in order to detect real, long term, global trends of the expected size from the NO_x input from nuclear bomb tests.

A method of factoring out the seasonal trends has been presented by Komhyr et al (1971). They analyzed ozone data of the world for the time interval 1961-70 (inclusive) for 10 stations. They evaluated the "mean monthly total ozone deviations from monthly normals, based on the periods of record...The ozone trend lines [linear change with time] were fitted to the data by the least squares method". Table 3 reproduces their results, and in Table 3 the error range is two standard deviations (2σ) or three times the computed probable error. The seven stations show a linear increase in ozone between 2.4 and 10.0 per cent per decade, well within the 95 per cent confidence level (2σ).

We have adopted the method of Komhyr et al (1971), with one extension, to examine all the total-ozone data in the published volumes of Ozone Data for the World (1960-70). For the stations considered by Komhyr et al ozone was observed at least 25 days per month in almost every case. In considering all the data from all stations, there are some cases where ozone was observed only once or twice a month. In our extension of Komhyr's method, we weighted the monthly means by the number of observations made that month.

In pooling data from stations at different geographical locations and with different instruments, there is considerable uncertainty as to how to proceed. One could represent different zones of the earth by the stations there and attempt to present a global average of some sort, but the stations are so sparse in the tropical zone and the southern hemisphere that the expected error of estimate

Table 3. Summary of data analyses by Komhyr et al (1971)

Station	Mean O_3 m-atm-cm	% increase per decade (1961-70)
Huancayo, Peru	262	6.0±1.2
Kodaikanal, India	257	10.0±1.2
Mauna Loa, Hawaii	276	2.4±1.8
Brisbane, Australia	289	6.6±1.8
Nashville, USA	332	8.8±2.1
Arosa, Switzerland	333	4.1±3.0
Oxford, England	354	8.9±3.0

for some parts of the world would be much greater than for other parts. We chose to combine different stations not in terms of geography but in terms of weighting factors based on the standard deviation of the day-by-day readings at each station. We used the reciprocal of the square of the standard deviation as the weighting factor. This method suppresses the contribution from stations that covered a short time span, that had few observations, or that had a large scatter of points. The averages we give may not be interpreted as a "global average", rather they are "weighted average" of all the stations in the world.

In agreement with Komhyr et al (1971), we find a statistically significant (5.1±1.2%) increase in ozone for the period 1961-70 for the error-weighted average from 93 stations with 169,000 observation-days. We considered the increase of all stations for various time spans, 1960-70, 1961-70, 1962-70, etc. These are listed in Table 4. The last column gives the percentage increase in ozone over the various time intervals. In view of the time of resumption of nuclear tests (1961), it is suggestive to note that the average increase of ozone for the period 1961-70 was greater than the increase from 1960-70. However, such comparisons are complicated by the different number of stations in the various periods.

As can be seen from Figure 3, most of the nuclear bomb debris was in the northern hemisphere. We made a comparison between the weighted average ozone record in the northern hemisphere and the southern hemisphere, and also stations above 50°N latitude were averaged together, Table 5. This pooling of data from different

Table 4. Weighted average changes of ozone for various time intervals, b is rate of increase of ozone percent per decade.

Time period	Years	Stations	Observation days	b	2σ	Percent increase over time period
1960-70	11	93	178,000	4.5	1.2	4.9
1961-70	10	93	169,000	5.1	1.2	5.1
1962-70	9	92	159,000	4.8	1.3	4.3
1963-70	8	89	147,000	4.6	1.4	3.7
1964-70	7	89	132,000	4.9	1.6	3.4
1965-70	6	86	111,000	4.6	2.0	2.8

Table 5. Geographical variation of ozone changes for the two time intervals 1960-62 (inclusive), 1963-70.

Latitude	Time period	% increase per decade	2σ	No. of stations	No. of meas. days
50N-90N	1960-62	-12.6	10.6	20	11,000
	1963-70	+7.6	3.2	28	47,000
	1960-70	+6.5	2.4	29	57,000
0-90N	1960-62	-7.6	7.0	42	28,000
	1963-70	+5.6	1.5	74	129,000
	1960-70	+5.3	1.2	76	156,000
0-90S	1960-62	-3.6	18.8	9	4,200
	1963-70	-1.2	3.1	15	19,000
	1960-70	+0.2	2.3	17	23,000
All Stations	1960-62	-7.1	6.5	51	32,000
	1963-70	4.6	1.4	89	147,000
	1960-70	4.5	1.2	93	178,000

stations was weighted by the reciprocal of the square of the standard deviation of the daily readings. It gives the properly weighted average value for all the stations within the stated geographical boundaries; it does not try to give equal weight to equal sized geographical areas (the uneven distribution of the stations, and the difference in standard error between filter and prism instruments make it very difficult to carry out such a treatment). Table 5 shows no significant change in stratospheric ozone in the southern hemisphere over the time interval 1960-71. In the northern hemisphere, there is a decrease of ozone between 1960-62 (inclusive), the nominal change is -7.6±7.0% per decade. The trend (-7.6%) is greater than twice the standard deviation (7.0) by a small amount. It is recognized that biennial cycles could greatly distort three-year trends. The point to be made is that if the ozone data for 1960, 1961, 1962 show any trend at all, it is a decrease. On the other hand, the increases for the period 1963-70 (inclusive) are much larger than twice the standard deviation. There was a larger increase (+7.6±2% per decade) for the average of 28 stations located north of 50°N latitude than for the average of 74 stations in the northern hemisphere (+5.6±1.5% per decade). This difference is consistent with the distributions of ^{90}Sr, Figure 3.

As an alternative to averaging the various stations, one may look at the records of each and every individual station. A plot of monthly deviations (Komhyr method) against time, from 1960-1970 inclusive, for all stations is given in the appendix. The plots are organized in terms of latitude, starting at the

north pole and moving to the south pole. Certain general comments can be made. The noisy pattern of the filter instrument is obvious simply by inspection. There are several patterns visible to the eye: (1) an increase of ozone over essentially the entire period, for example, Dickson Island, Lerwick, Leningrad, Sverdlovsk, Oxford, Irkutsk, Kiev, Hradek Kralove, Cagliari-Elmas, Nashville, Kagoshima, New Delhi, Kodaikanal; (2) essentially constant ozone (within a noise pattern) over the interval, for example, Resolute, Reykjavik, Negaevo, Edmonton, Goose Bay, Caribou, Bismark, Arosa, Toronto, Green Bay, Vladivostok, Sapporo, Vigna di Valle, Naples, Tateno, Dum Dum, Brisbane, Aspendale, Macquarie Island; (3) decreasing ozone, for example, Val-Joyeux, Magny-les-Hameaux, Abustumani, Mirniy, Halley Bay; (4) high ozone in early 1960's, low ozone in mid-1960's, and high in late 1960's, for example, Aarhus, Murmansk, Yakutsk, Riga, Moscow, Kuibyshev, Mont-Louis, Alma Ata, Messina, Port-aux-Francaise. Oxford (increase), Arosa (constant), Val-Joyeaux (decrease), and Messina (minimum in mid-1960's) are all in the European temperate zone, and each used the Dobson instrument. This variety of patterns shown by individual stations indicates that no general conclusions can be drawn from the results of any single station or from a small number of stations.

During the years 1960-62, there were 30 ozone-observing stations that measured total ozone during at least 30 out of 36 months. These stations are entered in Table 6. At 23 out of 30 stations, ozone decreased during the years 1960, 61, 62, and it increased at only 7 stations. The average change per decade is -10.8 per cent. Of the 30 stations in Table 6, 27 of them had

Table 6. Stations with at least 30 months of observation in the period 1960-62, and in 1963-70 (k, ozone increase per decade, percent)

Station	1960-62 (inclusive)			1963-70		
	Months of obs.	b	2σ	Months of obs.	b	2σ
Aarhus, Denmark	36	-12.7	14.9	92	-8.9	7.6
Abustamani, USSR	35	-36.1	34.0	29		
Ahmedabad, India	36	+12.3	11.3	82	+8.0	3.7
Alma Ata, USSR	30	-9.1	23.5	91	+25.9	11.5
Arosa, Switzerland	36	-5.8	18.4	90	+4.6	5.0
Aspendale, Aust.	36	-3.7	17.7	92	-4.3	2.4
Brisbane, Aust.	36	-4.3	11.7	92	+1.2	2.4
Camborne, U.K.	36	-11.8	19.2	38	+17.2	9.2
Cagliari-Elmas, Italy	36	-3.4	12.5	92	+13.5	4.2
Edmonton, Canada	36	-16.0	13.6	92	+3.3	4.3
Eskdalemuis, U.K.	36	-32.8	23.3	7		
Fort Collins, U.S.A.	31	-5.8	9.6	36	+18.0	12.8
Kagoshima, Japan	36	+55.9	19.3	92	0.0	4.8
Karadag, USSR	31	-89.3	22.6	36	+6.9	37.4
Kiev, USSR	31	-11.4	29.8	92	+9.5	8.4
Kodaikanal, India	36	-0.6	8.4	90	+12.4	1.9
Leningrad, USSR	30	-7.0	29.3	84	+12.8	8.4
Lerwick, U.K.	36	-26.7	15.9	79	+7.5	4.7
Marcus Is., Japan	36	-38.8	16.1	6		
Messina, Italy	36	-32.5	11.3	92	+8.2	3.2
New Delhi, India	36	+15.0	16.1	92	-5.6	3.3
Oxford, U.K.	36	-18.2	17.9	92	+6.8	4.5
Port-Aux-Francaise	30	+2.0	24.8	70	+29.0	19.0
Quetta, Pakistan	36	-7.0	17.0	37	+4.5	10.1
Resolute, Canada	35	-32.1	19.1	83	+4.1	6.2
Sapporo, Japan	36	+1.0	16.6	92	+4.1	3.2
Tateno, Japan	36	+10.7	16.7	92	-0.9	4.2
Toronto, Canada	36	-11.6	18.8	88	+1.7	4.4
Tromso, Norway	30	+11.1	31.0	64	+10.6	7.8
Vigna di Valle, Italy	36	-15.2	14.6	90	+4.1	4.4
Average per decade %		-10.8			+7.2	
% in interval		-3.3			+5.8	

at least 30 months of observations (20 had over 80 months of observations) between 1963 and 1970. Of these 27 stations, 22 showed an increase in ozone. Of these 27 stations, two thirds of them showed both a decrease between 1960-62 and an increase 1963-70. The average increase for these stations is 7.2 per cent per decade. Generally speaking, the three-year (1960-62) trends in Table 6 are not statistically significant as shown by the standard deviation of the data. In addition, such a short period could be part of a quasi-biennial cycle, and the trend could arise largely from a portion of such a cycle or from any longer term trend.

On the other hand, the increase of ozone from 1963-1970 appears to be a real effect. The weighted average (Table 4) of all the Ozone Data for the World gives an increase of about 4 or 5 percent. The individual stations in Table 6 show an increase usually larger than 2 standard deviations. The reason for this pronounced increase is not apparent, and it must be regarded as unknown. However, the following is a plausible working hypothesis, which may provide the explanation for this trend: Large scale nuclear testing occurred between 1952 and 1962 and then stopped. The ^{14}C and ^{90}Sr built up in an irregular manner between 1952 and 1963 and then dropped back to 1953 levels by 1968 (Figure 8). The expected steady-state reduction of ozone by the 1963 distribution of NO_x from the nuclear bombs is between 1 and 6% (Table 2). The expected response (Figure 8) of stratospheric ozone to these perturbations would be constancy before 1950, an irregular decrease

of a few per cent between 1952 and 1963, and a slow steady increase of a few percent from 1963 to 1970. There were only three ozone-observing stations (Tromso, Oxford, and Arosa) with records from before 1950 up to 1970 (Angell and Korshover, 1964; ODW). These few stations are inadequate to establish the pattern before 1950. Between 1950 and 1956 ten more long-term stations were established, and between 1957-59 another ten stations were started. During the 1960's there were about 90 stations with extensive ozone data (see the appendix), and there appears to have been a real systematic increase of ozone during this period. It is a reasonable hypothesis that the increase of ozone between 1963 and 1970 is the stratosphere of the earth returning to normal after the addition of nitric oxide by nuclear bombs in the period 1952-62.

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TITLES TO FIGURES

Fig. 1. Elevation and dimension of nuclear bomb clouds 30 minutes after detonation according to formulas by Foley and Ruderman. A standard ozone profile at 45° latitude is shown.

Fig. 2. Overlap of standard tropical ozone profile with bomb clouds 20 minutes after detonation.

Fig. 3. Stratospheric distribution of strontium-90 based on the Atomic Energy Commission sampling program: (a) May-August, 1963; (b) Jan.-Apr. 1964. Contour lines are in units of observed decompositions per minute (dpm) per thousand standard cubic feet of air (SCF).

Fig. 4. Maximum bomb-produced NO_x ($NO + NO_2 + HNO_3$) at three latitudes as deduced from Fig. 3 with the relation 500 dpm/1000 SCF = ($\frac{1}{5}$ to 1) ppb NO_x. The contour lines are based on the upper limit.

Fig. 5. Observed values of natural NO_x in the stratosphere. I, NO and NO_2, measured as easily ionized species; ⟷, NO_2 at sunset, equals total $NO+NO_2$, infrared absorption spectrum from balloons; continuous curves, HNO_3, infrared observation from balloons.

Fig. 6. Comparison of natural (Fig. 5) and bomb-produced NO_x at 45° latitude as deduced from Fig. 3 with the relations: for minimum NO_x, 1 ppb corresponds to 3000 dpm/1000 SCF of ^{90}Sr; for maximum NO_x, 1 ppb corresponds to 500 dpm/1000 SCF of ^{90}Sr. The "observed background" is the sum of HNO_3 for Nov. (Fig. 5) and the central point in the error bar for $NO + NO_2$ by Ackerman and Muller (Fig. 5).

Fig. 7. A distribution of NO_x ($NO + NO_2 + HNO_3$) from 500 American-type SST at a region of high traffic (three times the world-wide average) calculated from a model of one-dimensional vertical eddy diffusion; the estimated maximum and minimum distribution NO_x from nuclear bombs at 45°N (Fig. 6).

Fig. 8. The variation with time of total stratospheric inventory of strontium-90 and carbon-14, with periods and yields (MT) of nuclear explosives.

Fig. 9. Total ozone columns at Moscow and at Messina.

Fig. 10. Total ozone columns at Aspendale showing daily fluctuations and seasonal variations.

Fig. 11. Graph showing the number of years N of observation required to determine a trend b (% ozone change per decade) to the 95% confidence level at Aspendale, Australia (Pittock, 1972).

APPENDIX

Figs. 12-16. Contour maps of zonal average, excess carbon 14 in the stratosphere in units of 10^5 atoms of carbon-14 per gram of air.

Figs. 17-35. Monthly deviations of total ozone column for all stations in ODW (1960-1970). The stations are listed in order of latitude from 90°N to 90°S.

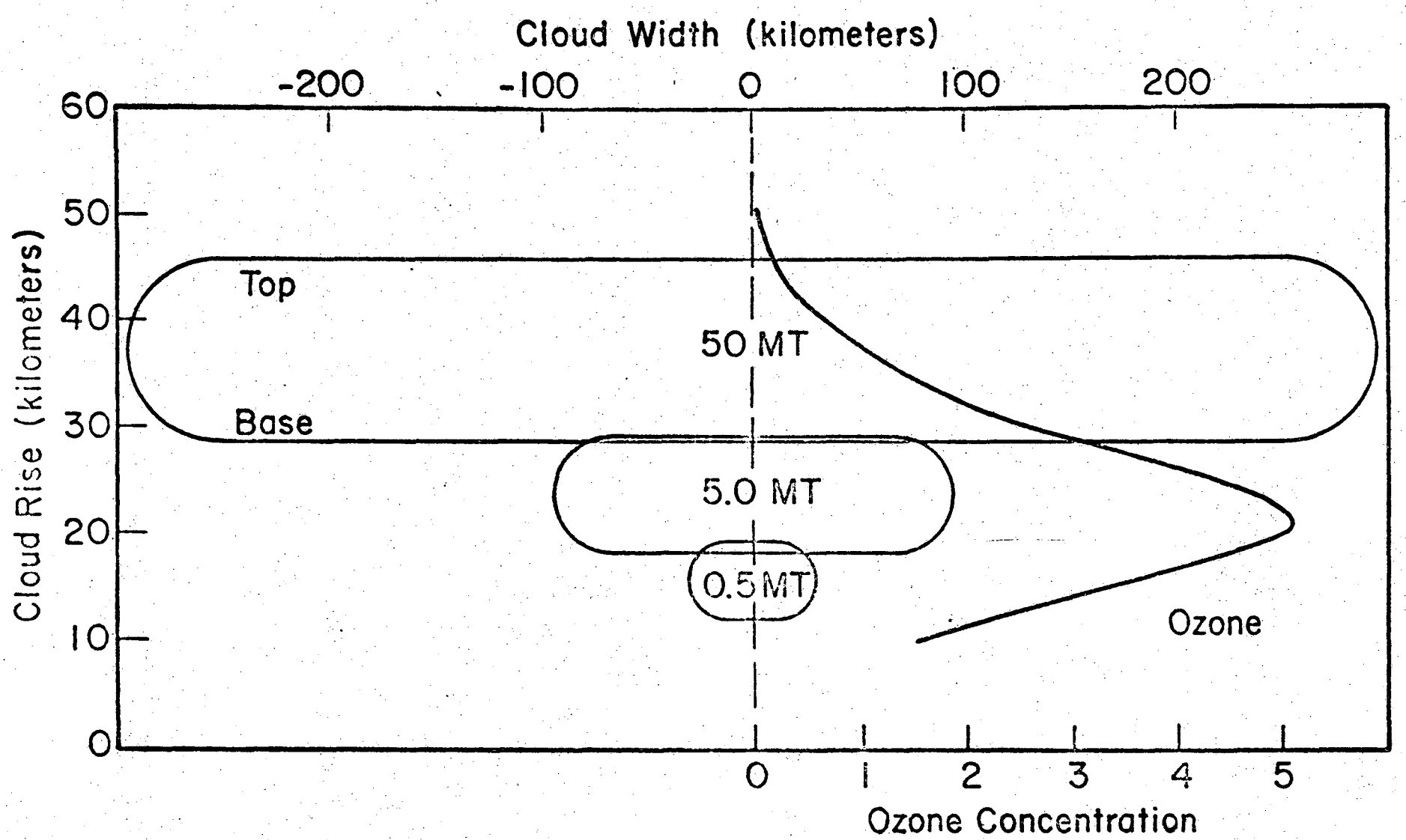
Cloud Width (kilometers)
-200
-100
0
100
200
60
50
40
30
20
10
0
Cloud Rise (kilometers)
Top
Base
50 MT
5.0 MT
0.5 MT
Ozone
0
1
2
3
4
5
Ozone Concentration

Fig. 1

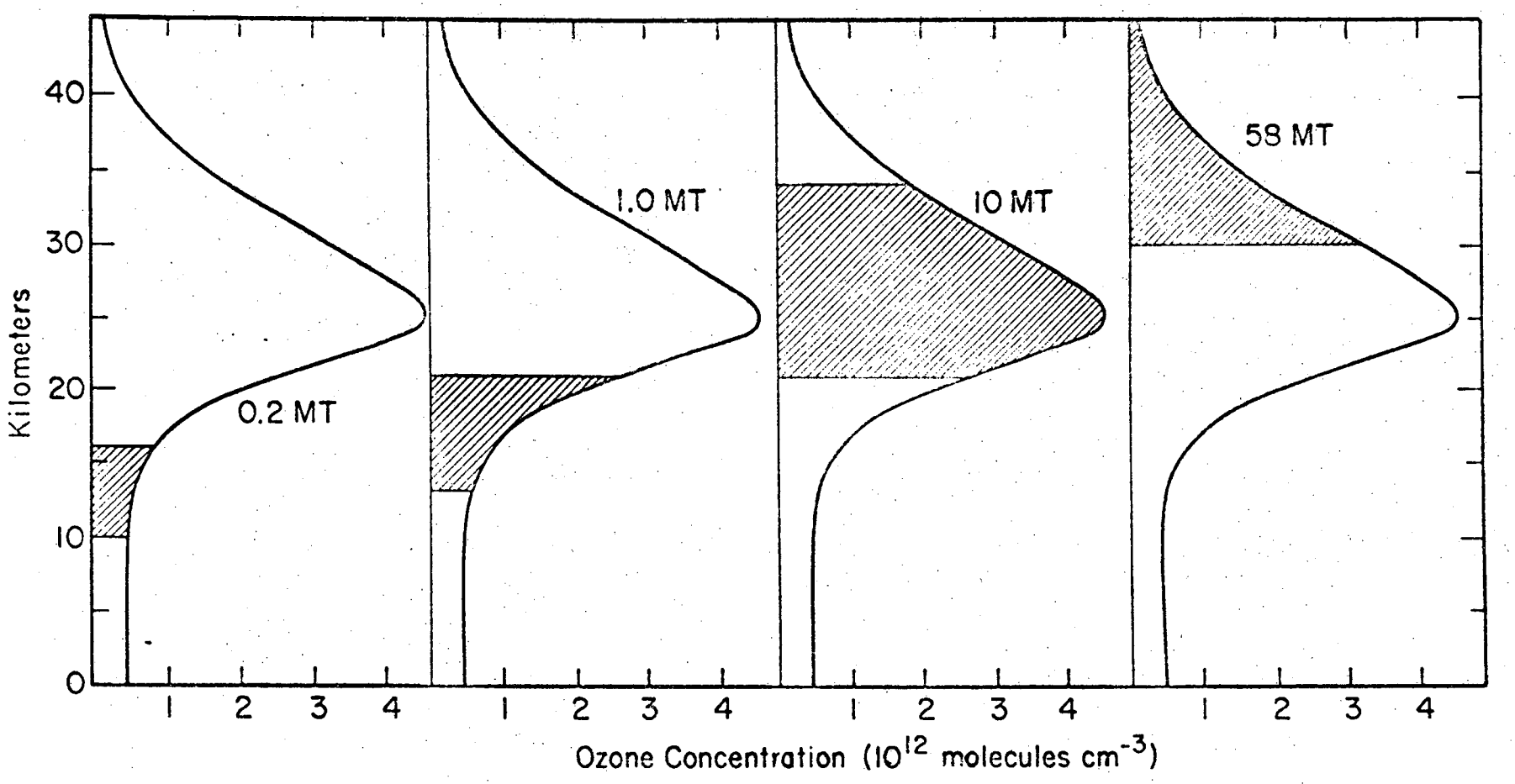
Kilometers
40
30
20
10
0
0.2 MT
1.0 MT
10 MT
58 MT
1 2 3 4
1 2 3 4
1 2 3 4
1 2 3 4
Ozone Concentration (10^{12} molecules cm^{-3})

Fig. 2

STRATOSPHERIC DISTRIBUTION OF STRONTIUM-90 (dpm/1000 SCF)

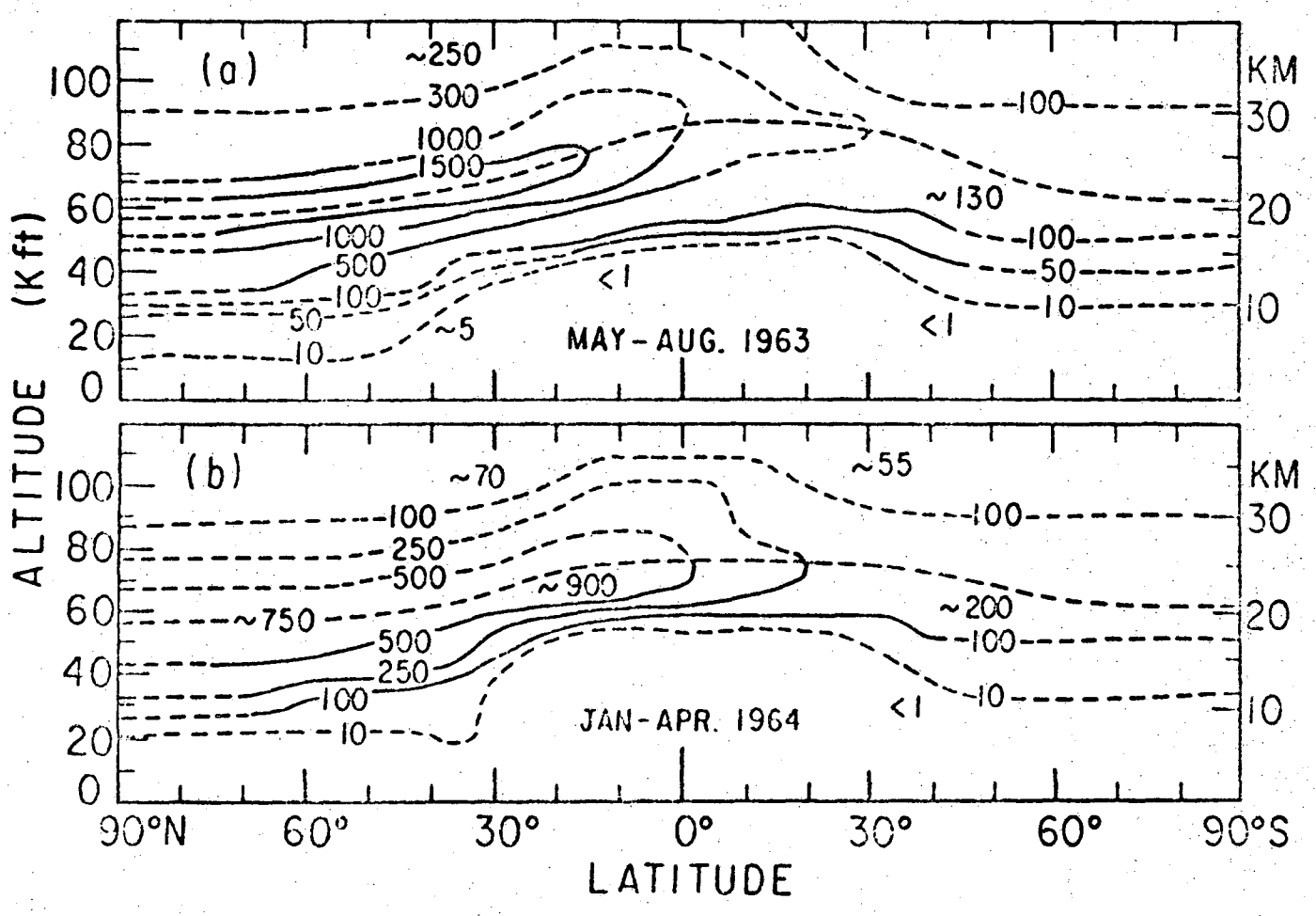

Fig. 3

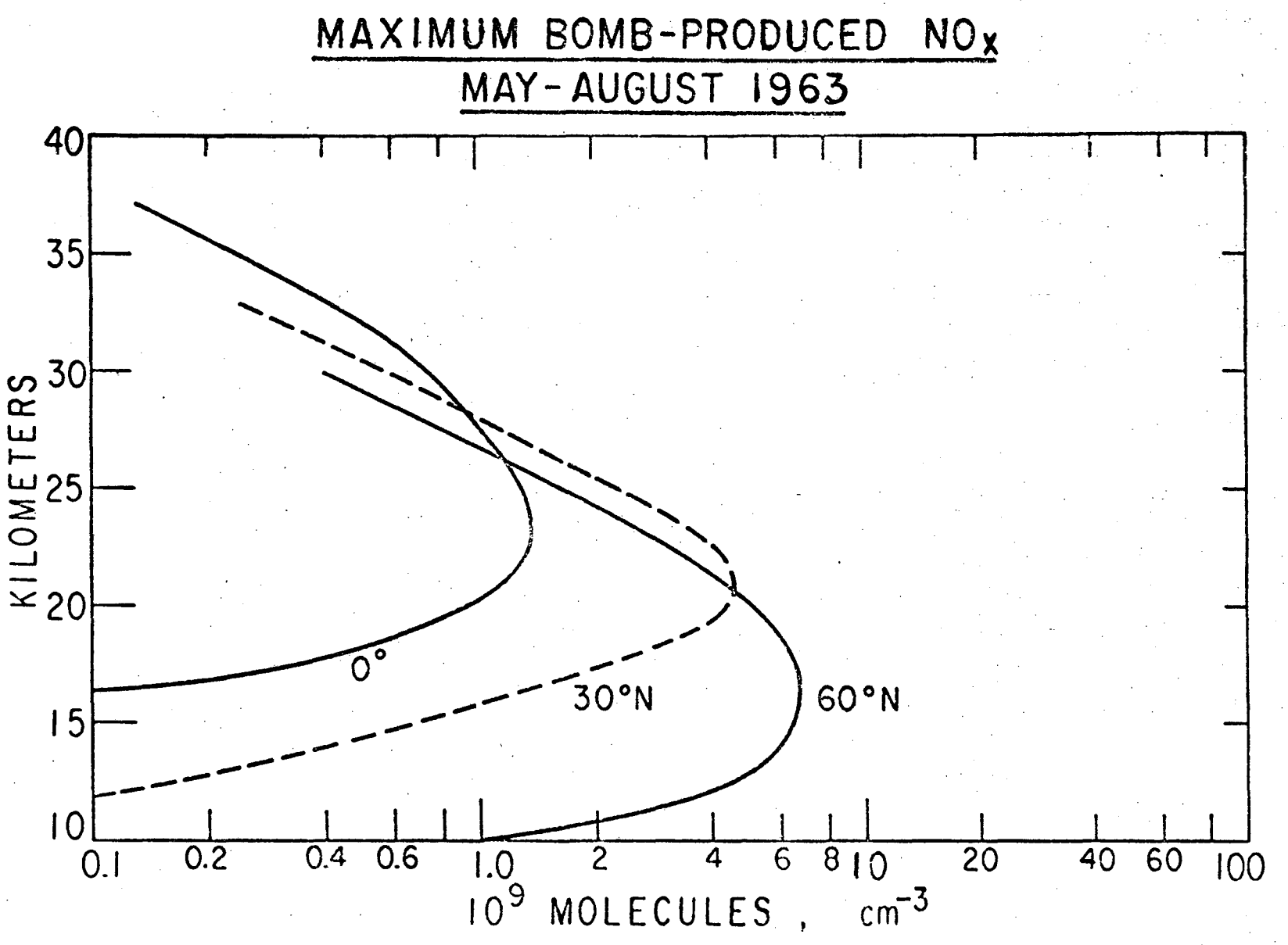
MAXIMUM BOMB-PRODUCED NO_x
MAY-AUGUST 1963
KILOMETERS
40
35
30
25
20
15
10
0°
30°N
60°N
0.1
0.2
0.4
0.6
1.0
2
4
6
8
10
20
40
60
100
10^9 MOLECULES , cm^{-3}

Fig. 4

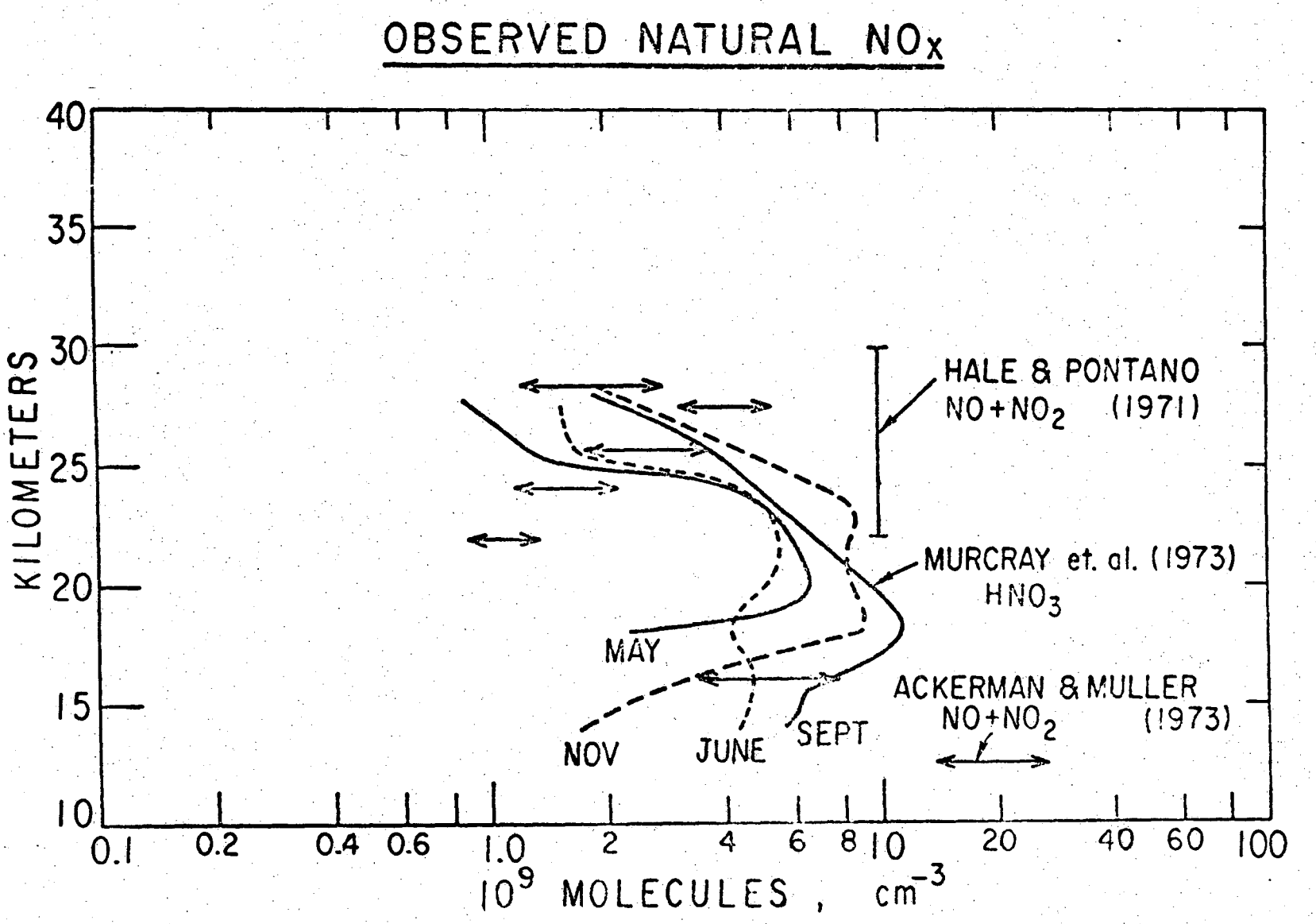
OBSERVED NATURAL NOx
KILOMETERS
40
35
30
25
20
15
10
0.1
0.2
0.4
0.6
1.0
2
4
6
8
10
20
40
60
100
10^9 MOLECULES , cm^{-3}
HALE & PONTANO
$NO+NO_2$ (1971)
MURCRAY et. al. (1973)
HNO_3
ACKERMAN & MULLER
$NO+NO_2$ (1973)
MAY
NOV
JUNE
SEPT

Fig. 5

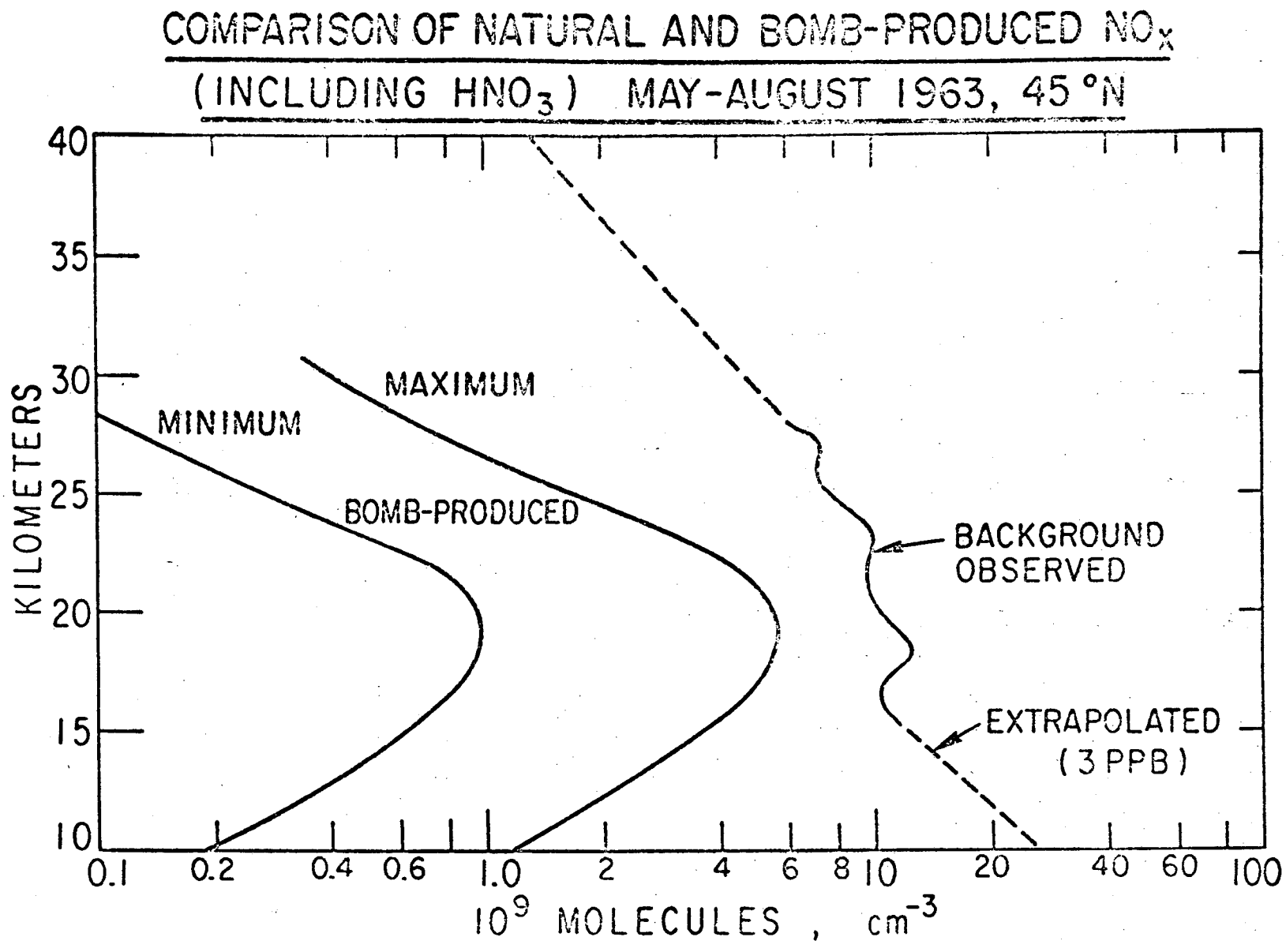
COMPARISON OF NATURAL AND BOMB-PRODUCED NO_x
(INCLUDING HNO_3) MAY-AUGUST 1963, 45 °N
KILOMETERS
40
35
30
25
20
15
10
MAXIMUM
MINIMUM
BOMB-PRODUCED
BACKGROUND OBSERVED
EXTRAPOLATED (3 PPB)
0.1
0.2
0.4
0.6
1.0
2
4
6
8
10
20
40
60
100
10^9 MOLECULES , cm^{-3}

Fig. 6=

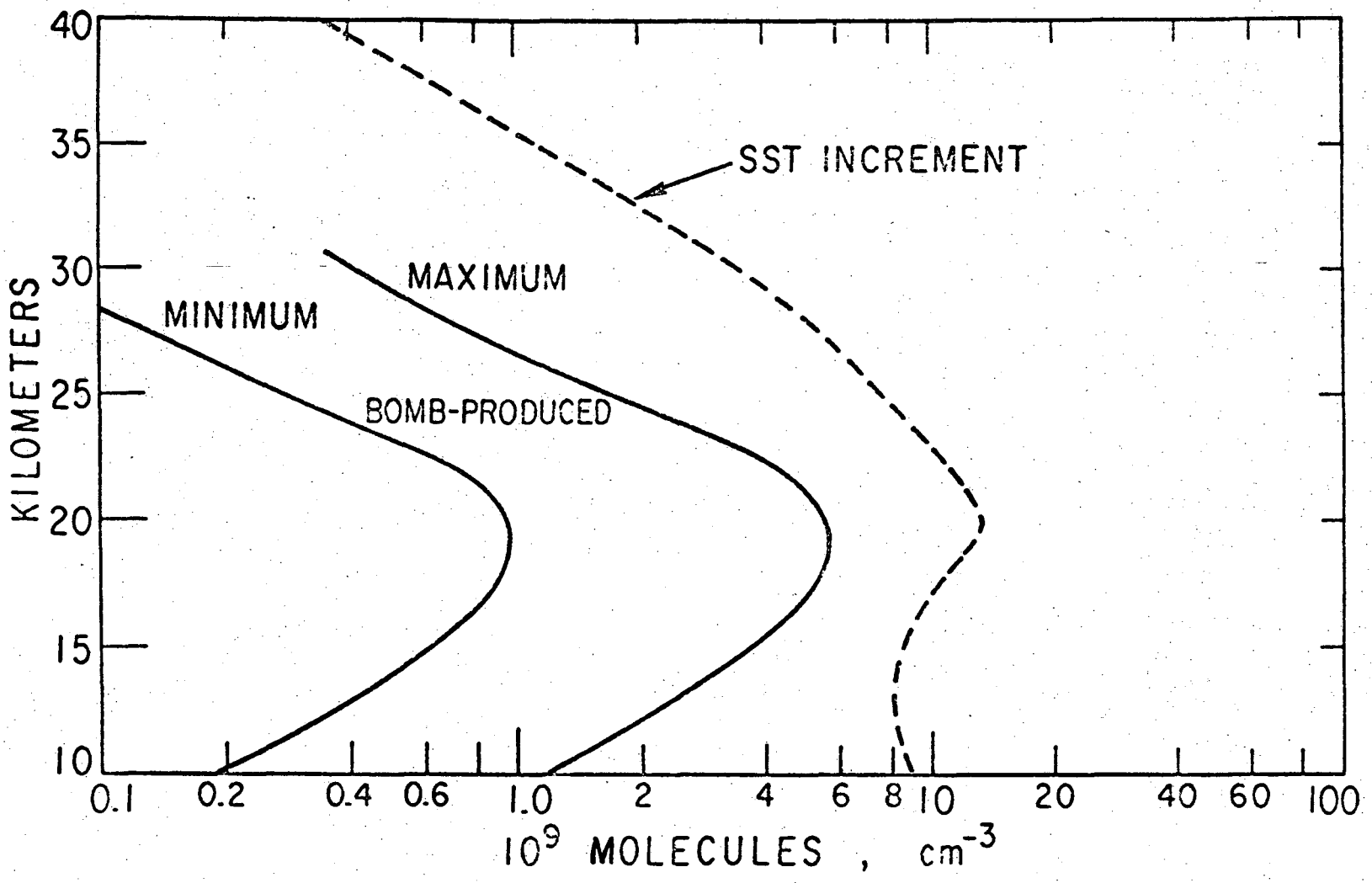
SST INCREMENT
MAXIMUM
MINIMUM
BOMB-PRODUCED
KILOMETERS
40
35
30
25
20
15
10
0.1
0.2
0.4
0.6
1.0
2
4
6
8
10
20
40
60
100
10^9 MOLECULES , cm^{-3}

Fig. 7

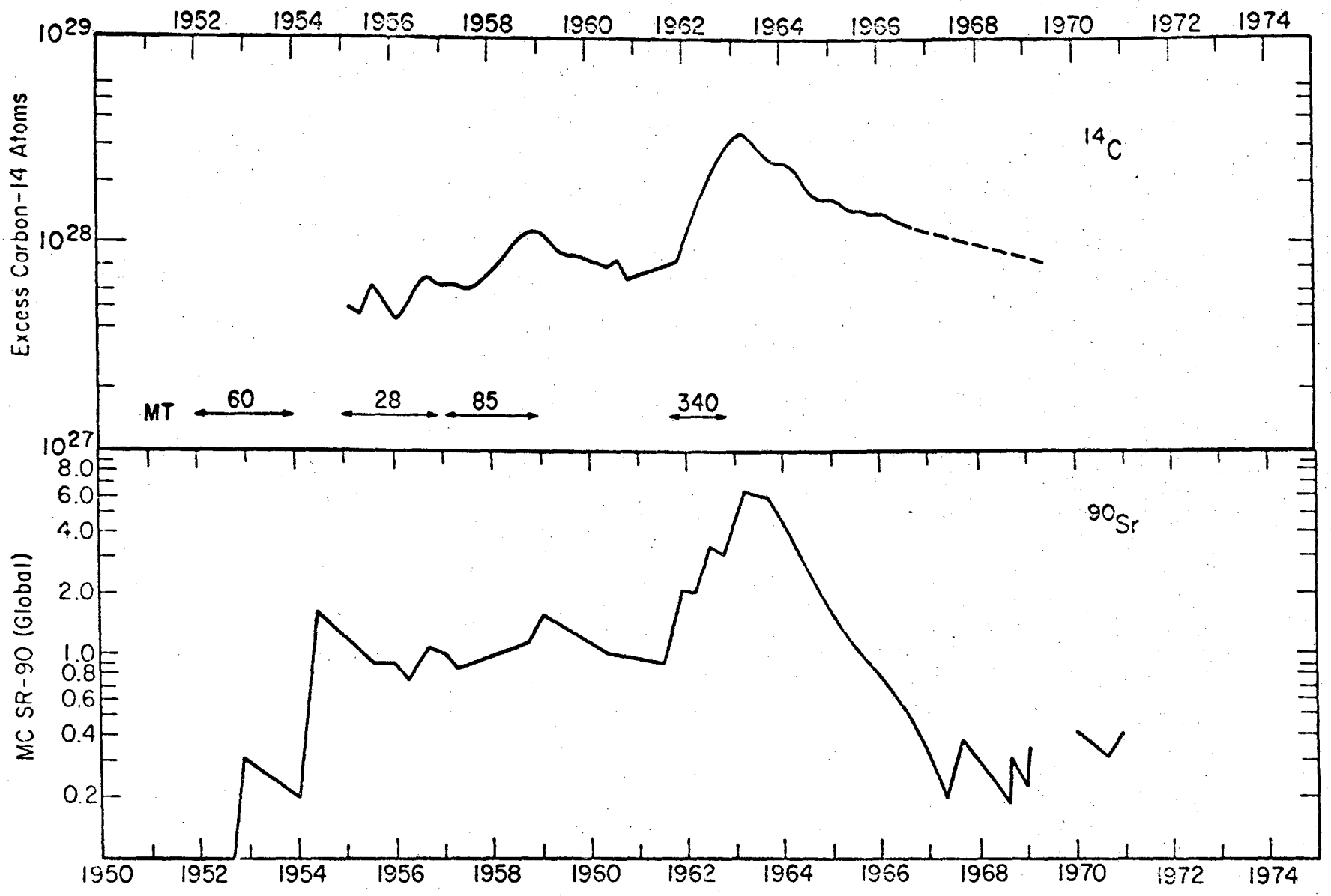
Excess Carbon-14 Atoms
10^{29}
10^{28}
10^{27}
^{14}C
MT
60
28
85
340
MC SR-90 (Global)
8.0
6.0
4.0
2.0
1.0
0.8
0.6
0.4
0.2
^{90}Sr
1950
1952
1954
1956
1958
1960
1962
1964
1966
1968
1970
1972
1974

Fig. 8

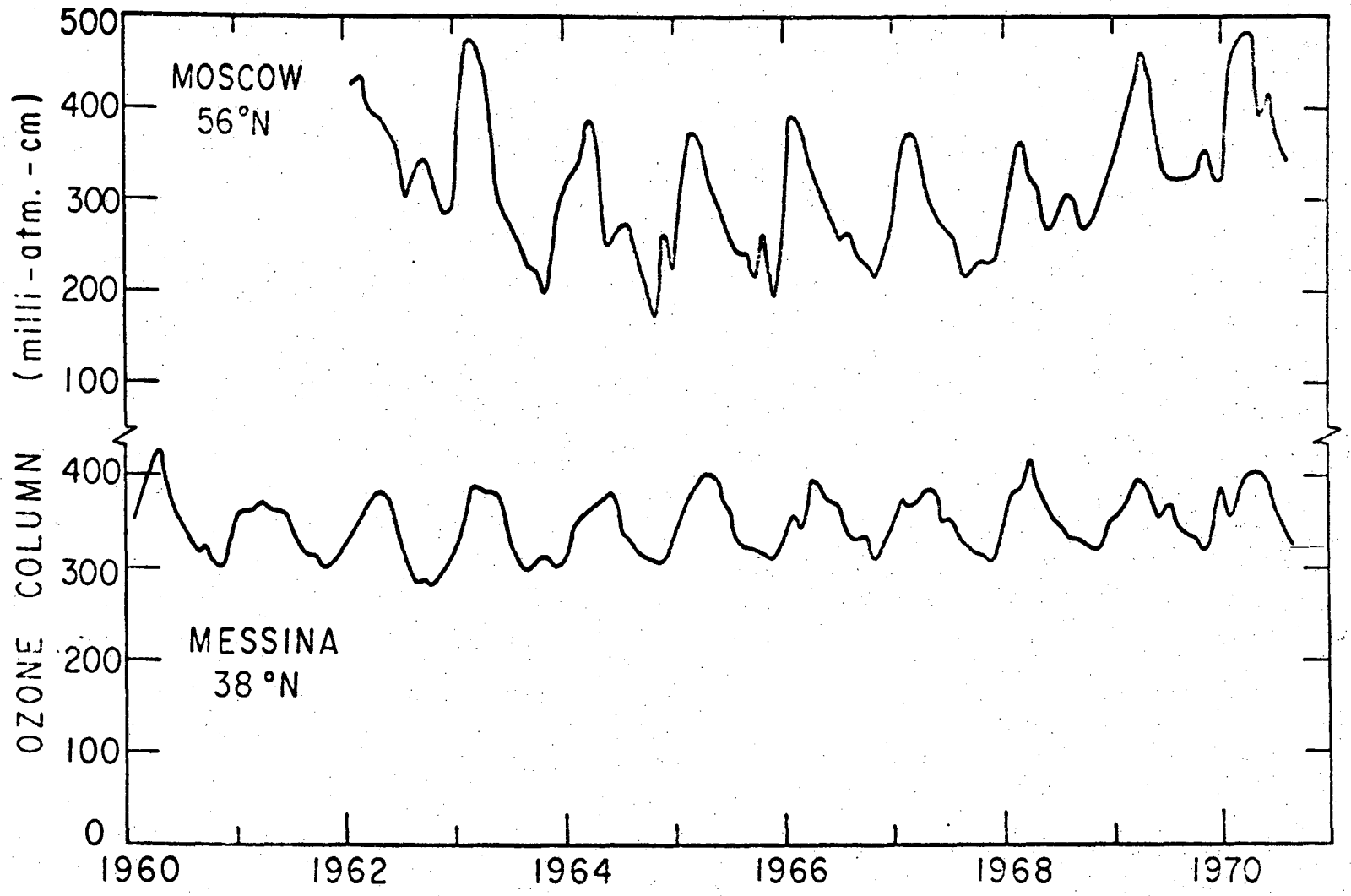
OZONE COLUMN (milli-atm.-cm)
500
400
300
200
100
MOSCOW
56°N
400
300
200
100
0
MESSINA
38 °N
1960
1962
1964
1966
1968
1970

Fig. 9

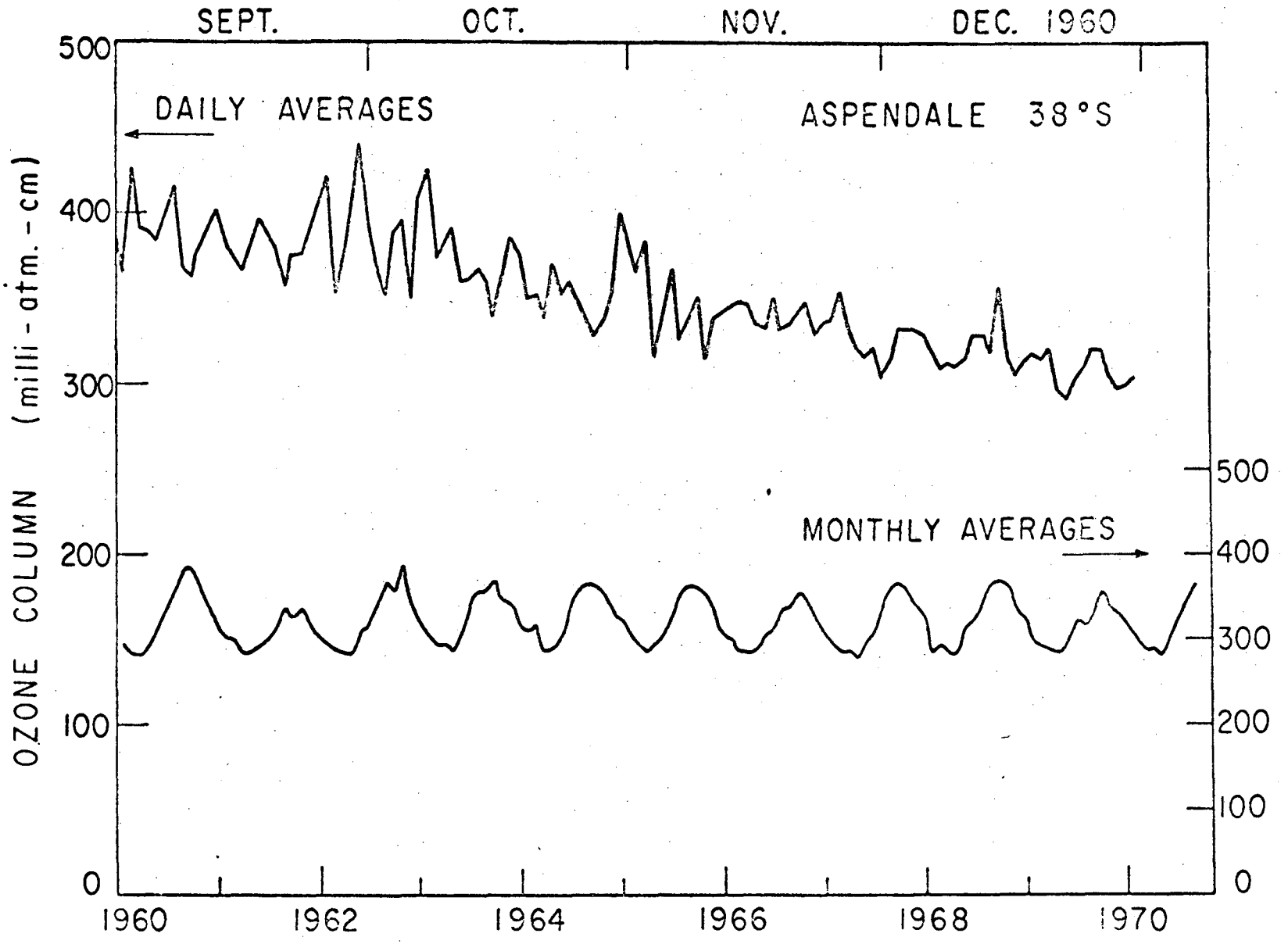
SEPT.
OCT.
NOV.
DEC. 1960
DAILY AVERAGES
ASPENDALE 38°S
MONTHLY AVERAGES
OZONE COLUMN (milli-atm.-cm)
500
400
300
200
100
0
1960
1962
1964
1966
1968
1970

Fig. 10

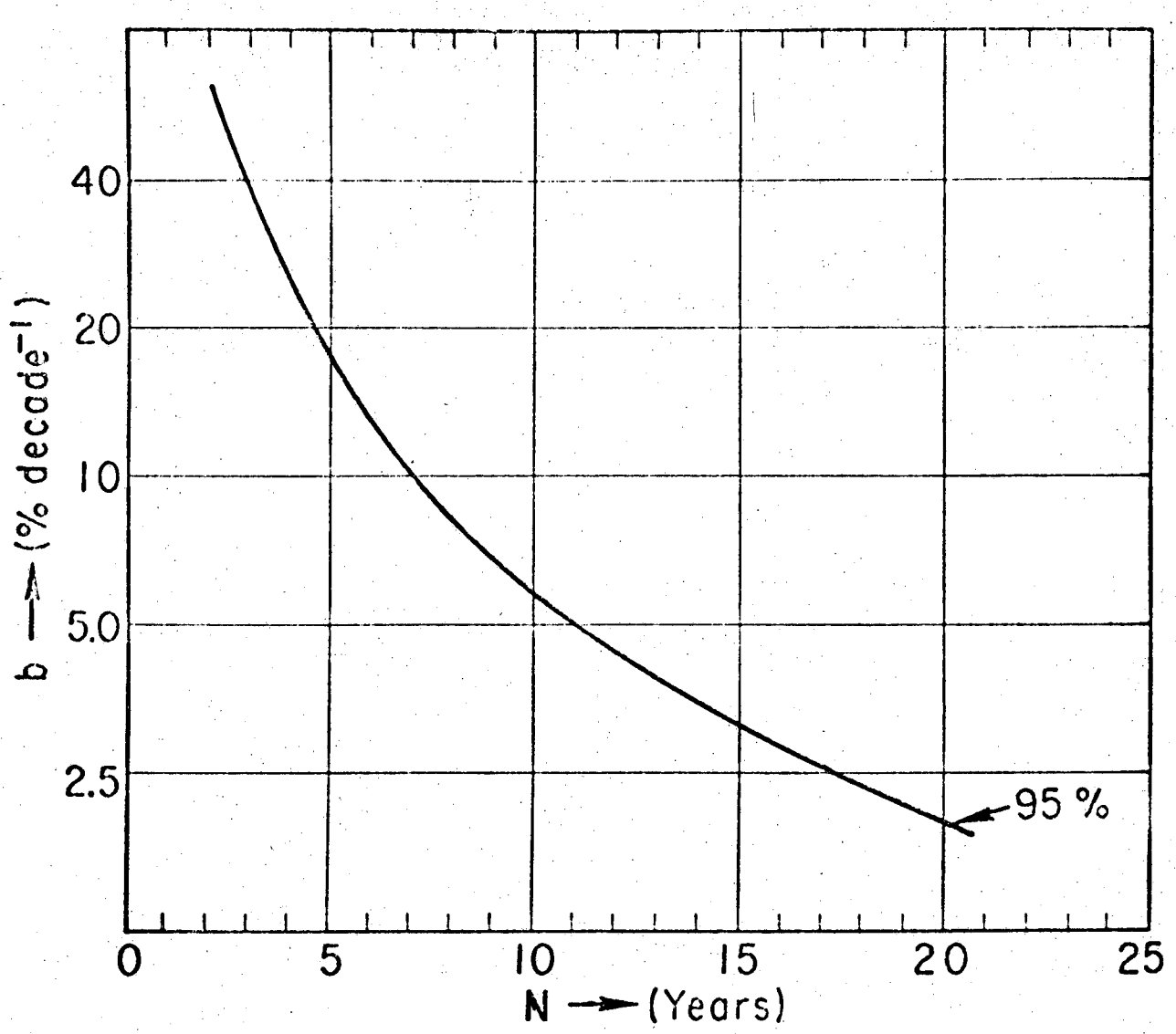

Fig. 11

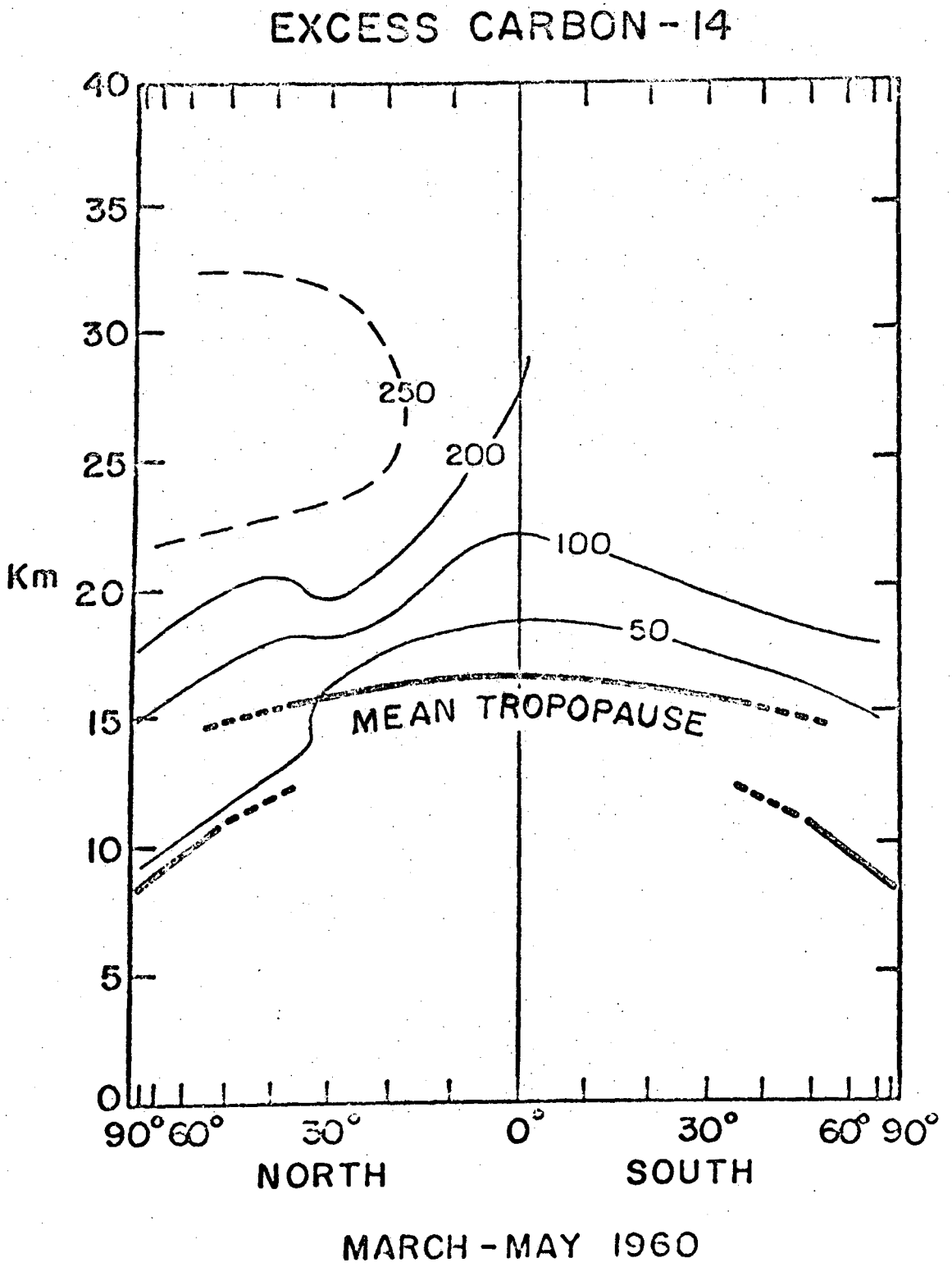
EXCESS CARBON-14
40
35
30
25
20
15
10
5
0
Km
250
200
100
50
MEAN TROPOPAUSE
90° 60° 30° 0° 30° 60° 90°
NORTH
SOUTH
MARCH-MAY 1960

Fig. 12

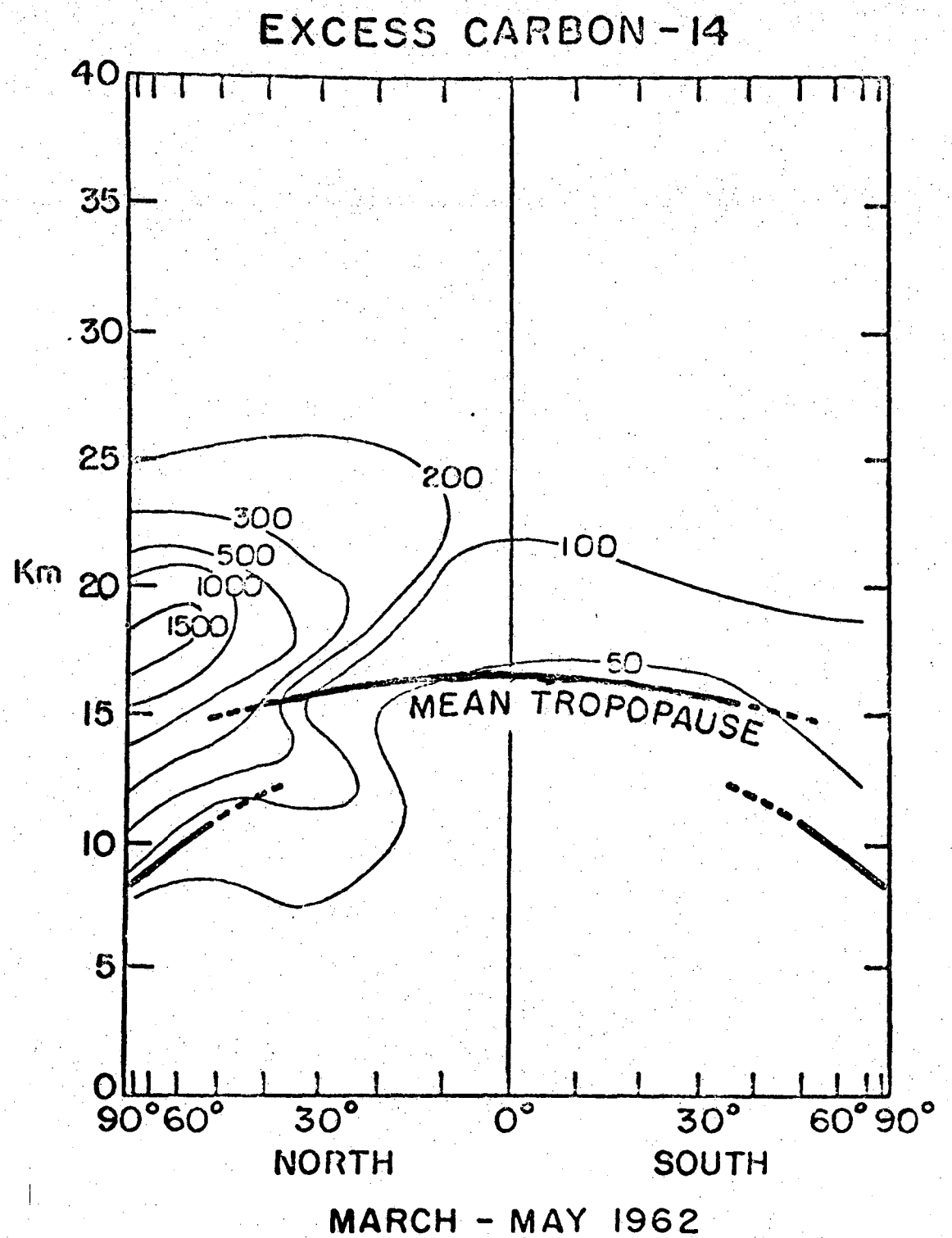
EXCESS CARBON-14
40
35
30
25
20
15
10
5
0
Km
200
300
500
1000
1500
100
50
MEAN TROPOPAUSE
90° 60° 30° 0° 30° 60° 90°
NORTH
SOUTH
MARCH - MAY 1962

Fig. 13

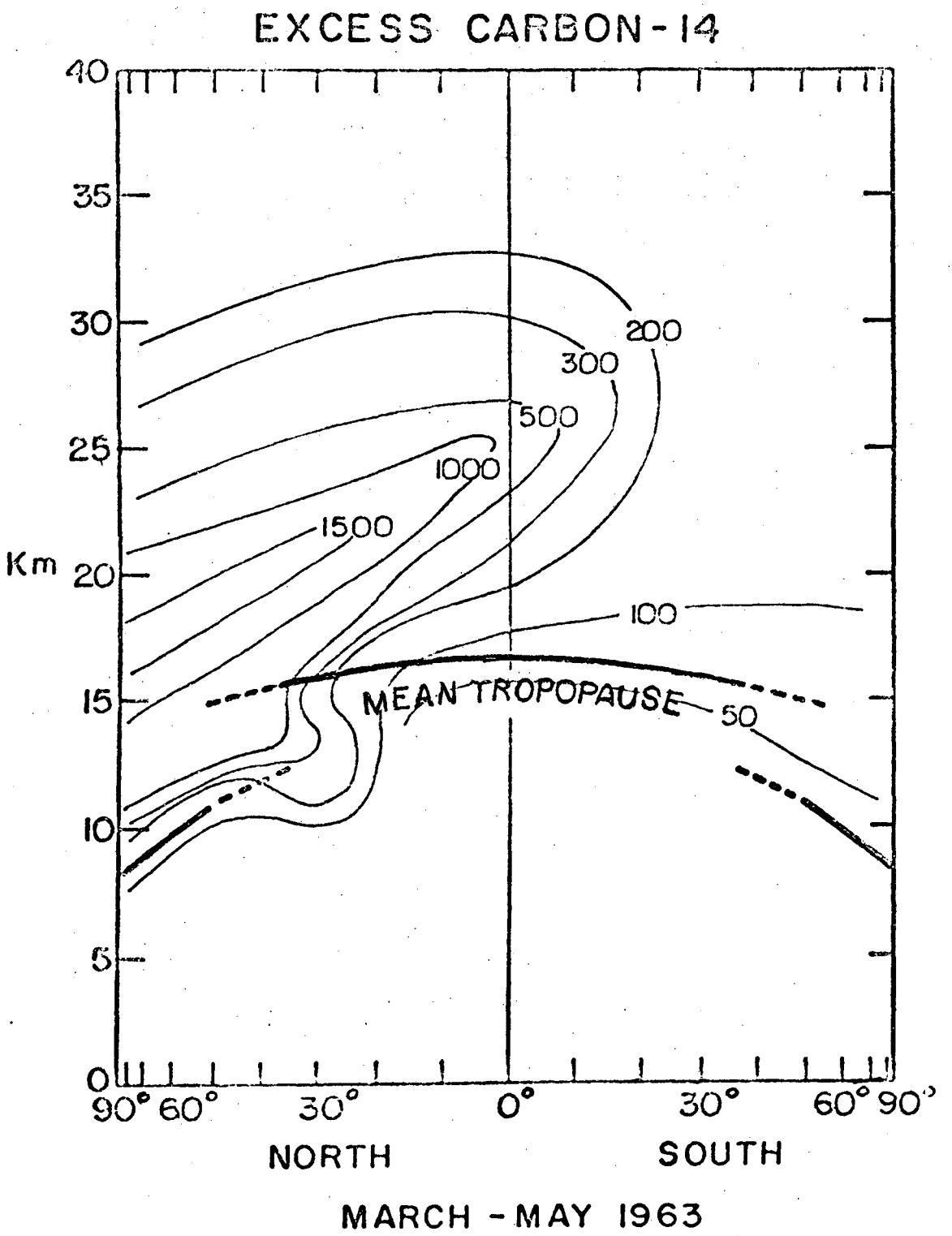
EXCESS CARBON-14
40
35
30
25
20
15
10
5
0
Km
200
300
500
1000
1500
100
50
MEAN TROPOPAUSE
90° 60° 30° 0° 30° 60° 90°
NORTH
SOUTH
MARCH - MAY 1963

Fig. 14

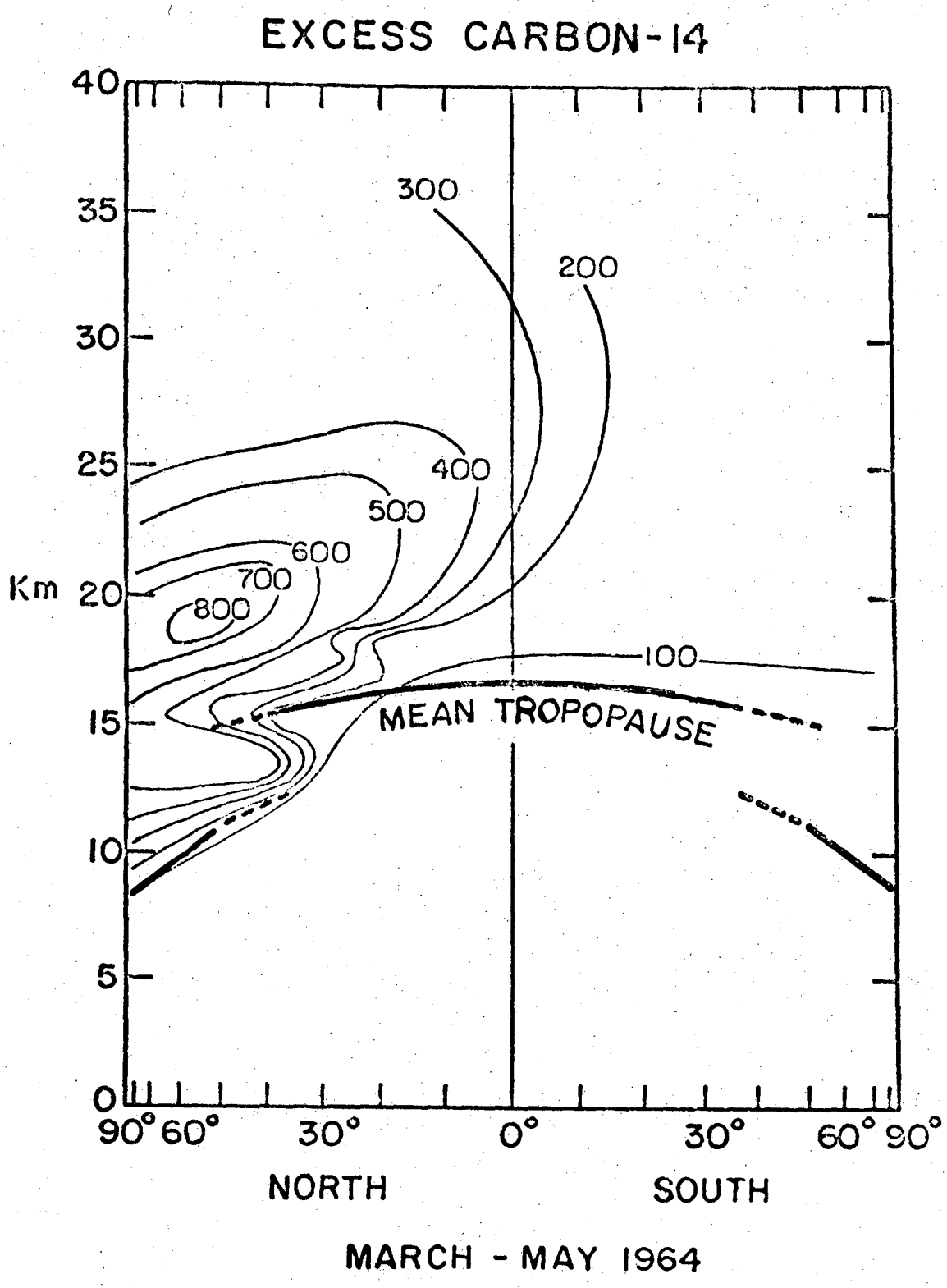
EXCESS CARBON-14
40
35
30
25
20
15
10
5
0
Km
300
200
400
500
600
700
800
100
MEAN TROPOPAUSE
90° 60° 30° 0° 30° 60° 90°
NORTH
SOUTH
MARCH - MAY 1964

Fig.15

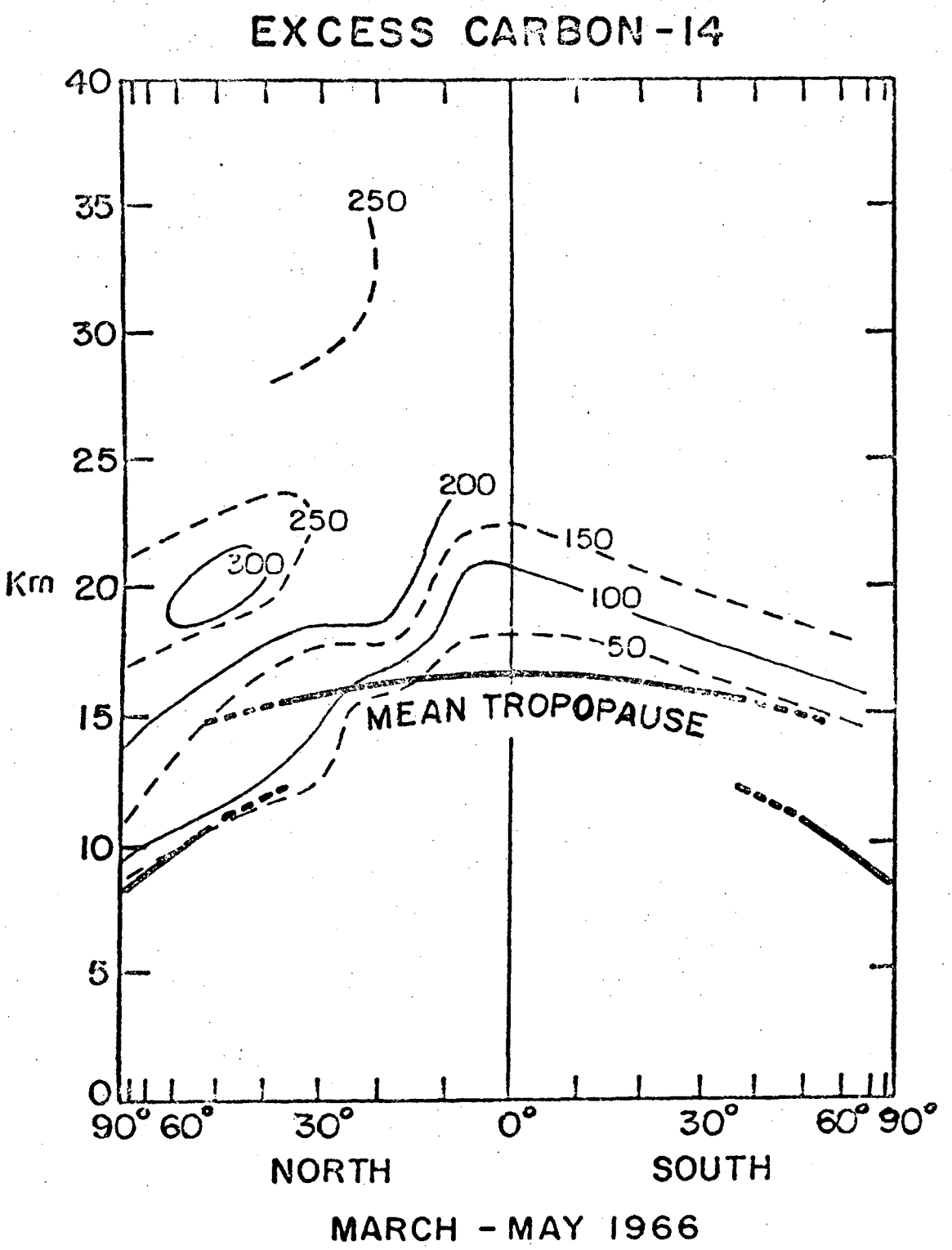
EXCESS CARBON-14
40
35
30
25
20
15
10
5
0
Km
250
200
250
300
150
100
50
MEAN TROPOPAUSE
90° 60° 30° 0° 30° 60° 90°
NORTH
SOUTH
MARCH - MAY 1966

Fig. 16

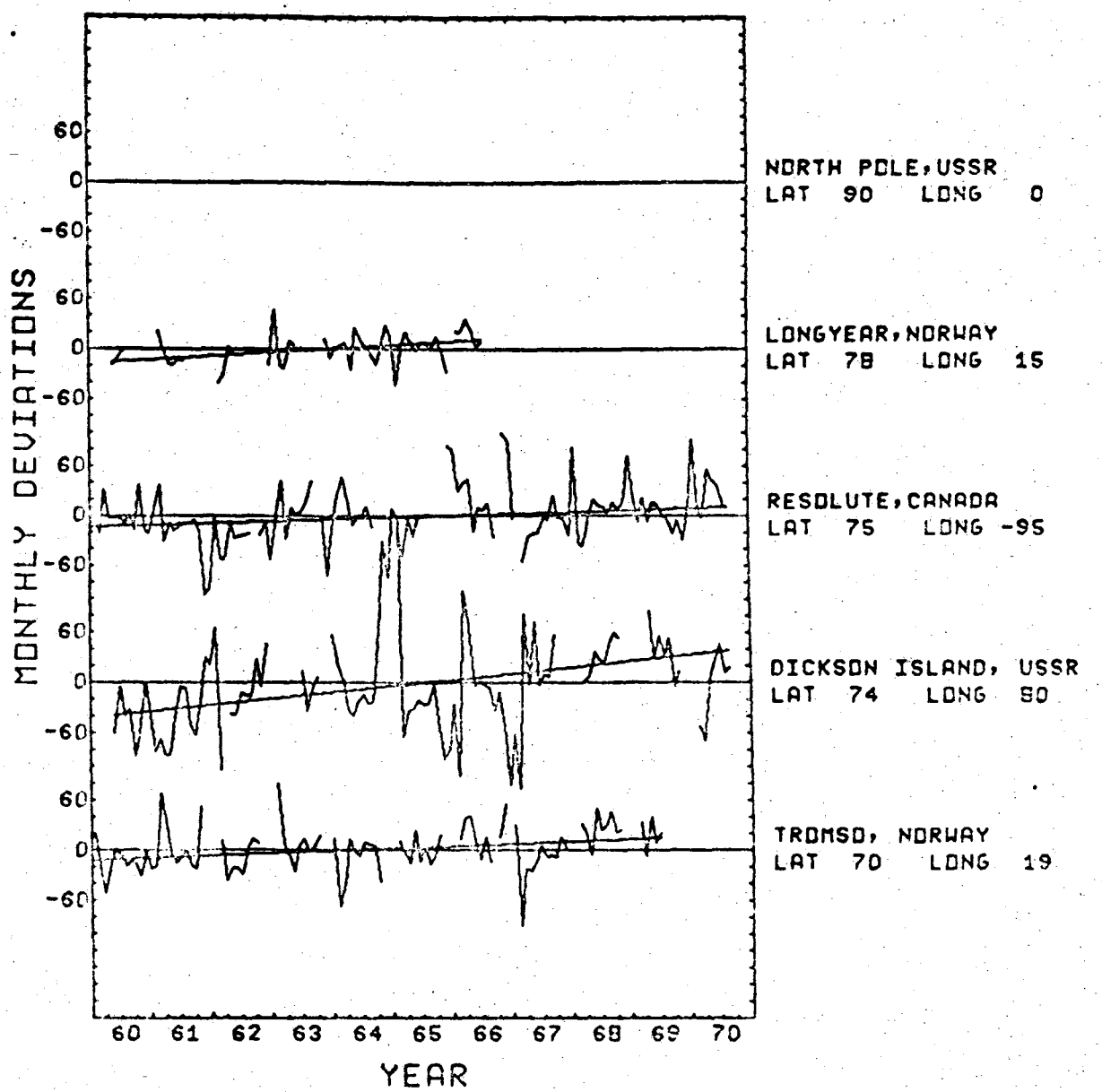
MONTHLY DEVIATIONS
60
0
-60
NORTH POLE, USSR
LAT 90 LONG 0
LONGYEAR, NORWAY
LAT 78 LONG 15
RESOLUTE, CANADA
LAT 75 LONG -95
DICKSON ISLAND, USSR
LAT 74 LONG 80
TROMSO, NORWAY
LAT 70 LONG 19
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig. 17

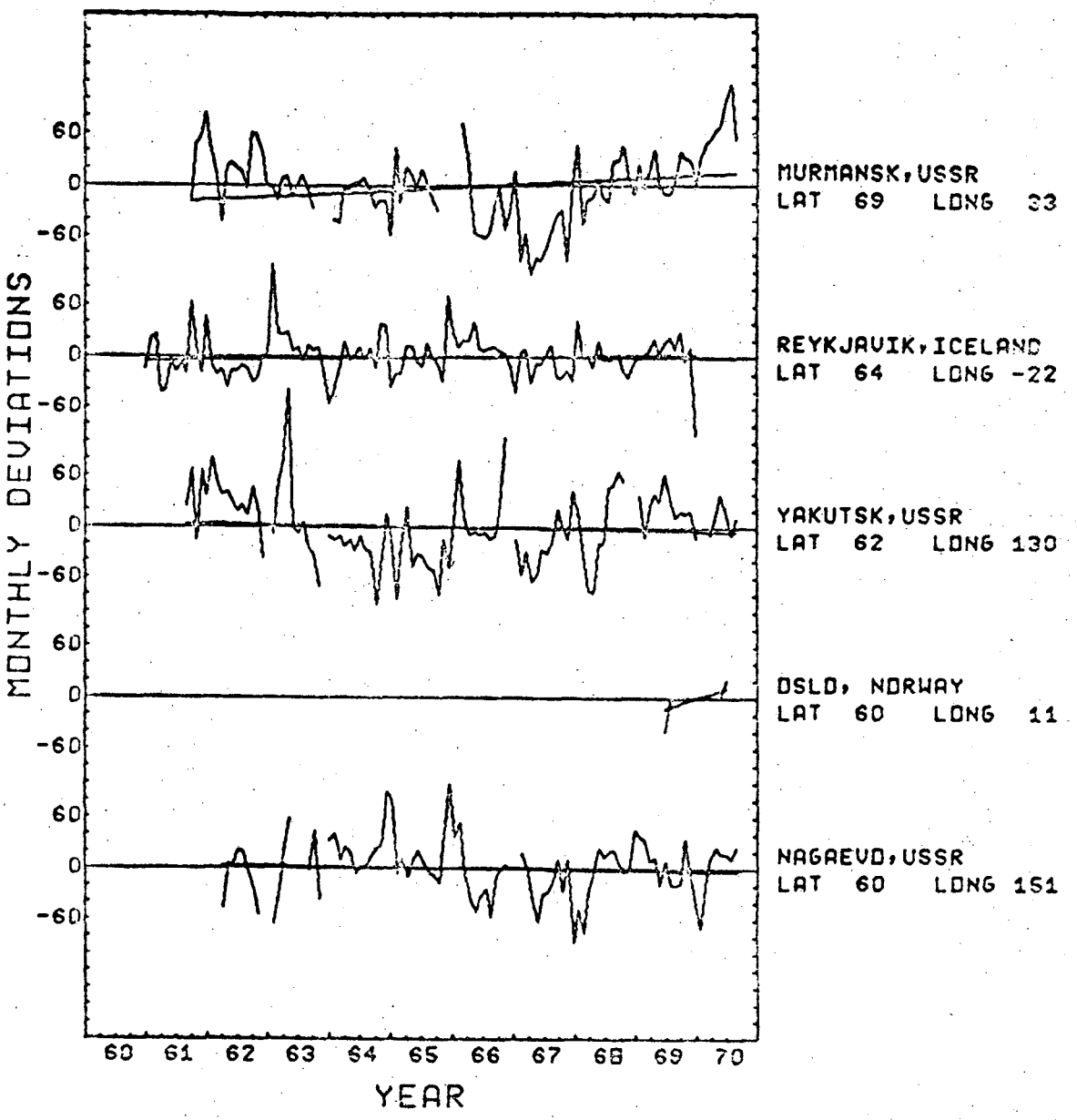
MONTHLY DEVIATIONS
60
0
-60
MURMANSK,USSR
LAT 69 LONG 33
REYKJAVIK,ICELAND
LAT 64 LONG -22
YAKUTSK,USSR
LAT 62 LONG 130
OSLO, NORWAY
LAT 60 LONG 11
NAGAEVO,USSR
LAT 60 LONG 151
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig. 18

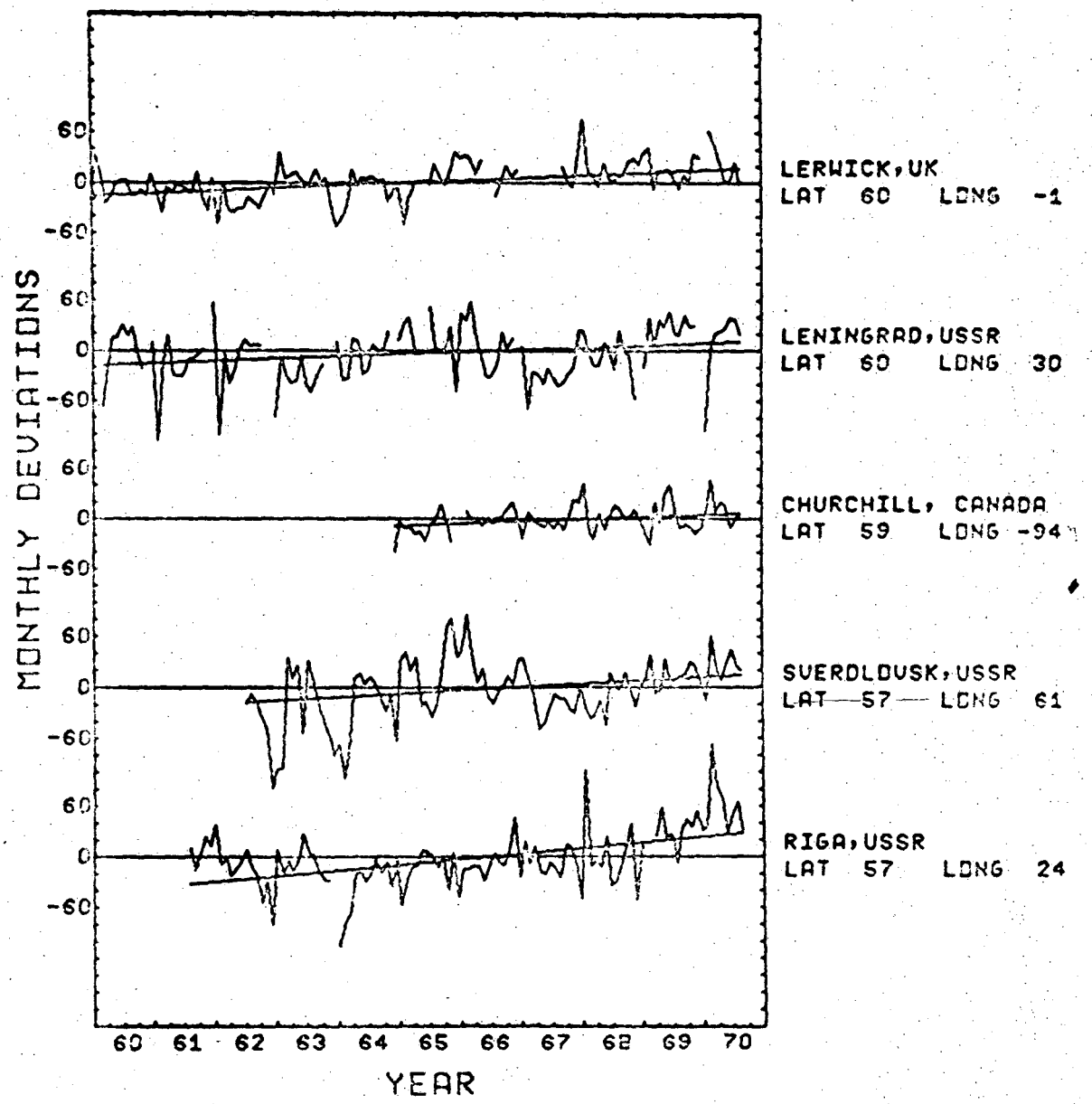
MONTHLY DEVIATIONS
60
0
-60
LERWICK,UK
LAT 60 LONG -1
LENINGRAD,USSR
LAT 60 LONG 30
CHURCHILL, CANADA
LAT 59 LONG -94
SVERDLOVSK,USSR
LAT 57 LONG 61
RIGA,USSR
LAT 57 LONG 24
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig. 19

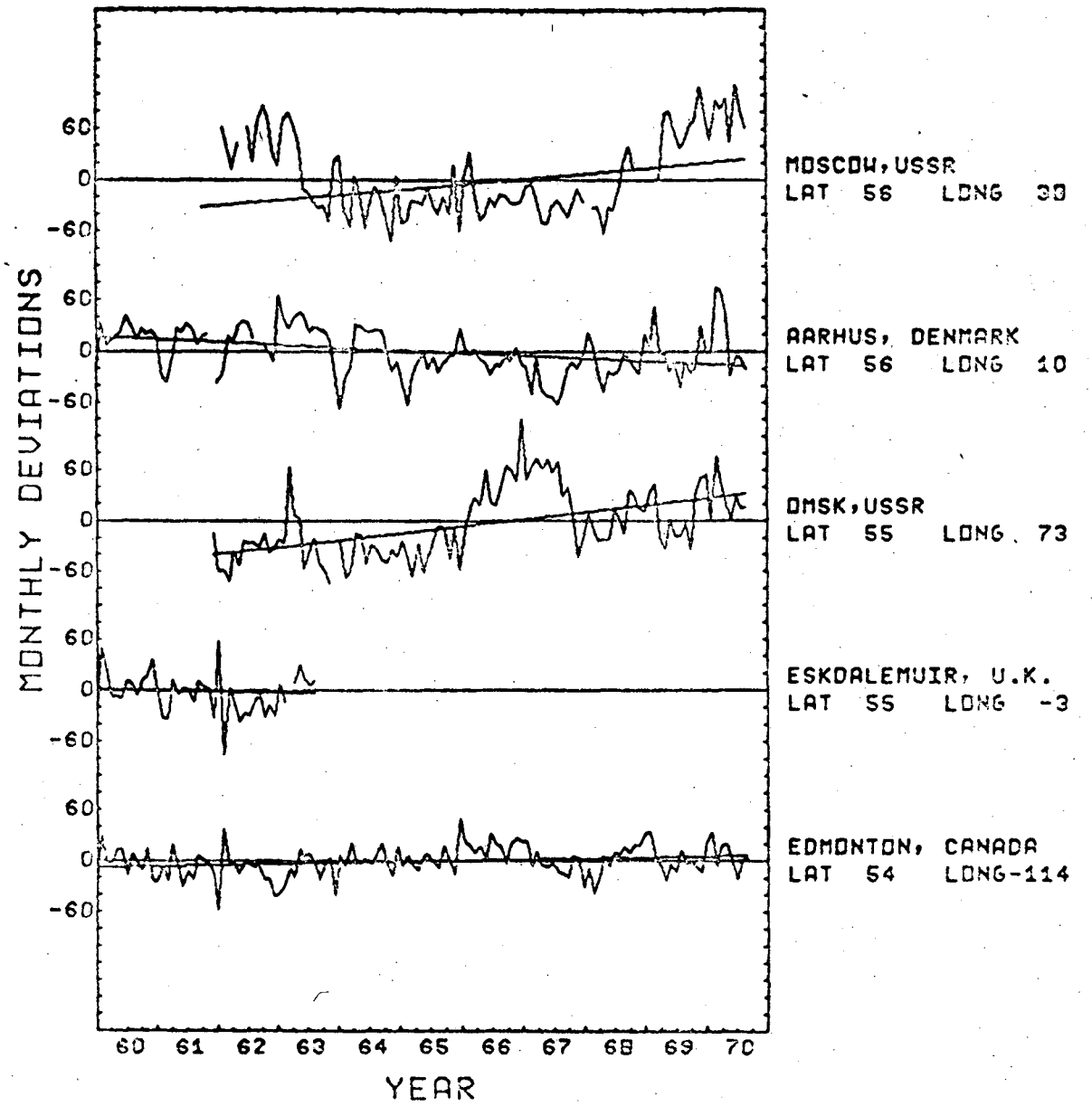
MONTHLY DEVIATIONS
60
0
-60
MOSCOW, USSR
LAT 56 LONG 30
AARHUS, DENMARK
LAT 56 LONG 10
OMSK, USSR
LAT 55 LONG 73
ESKDALEMUIR, U.K.
LAT 55 LONG -3
EDMONTON, CANADA
LAT 54 LONG-114
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig. 20

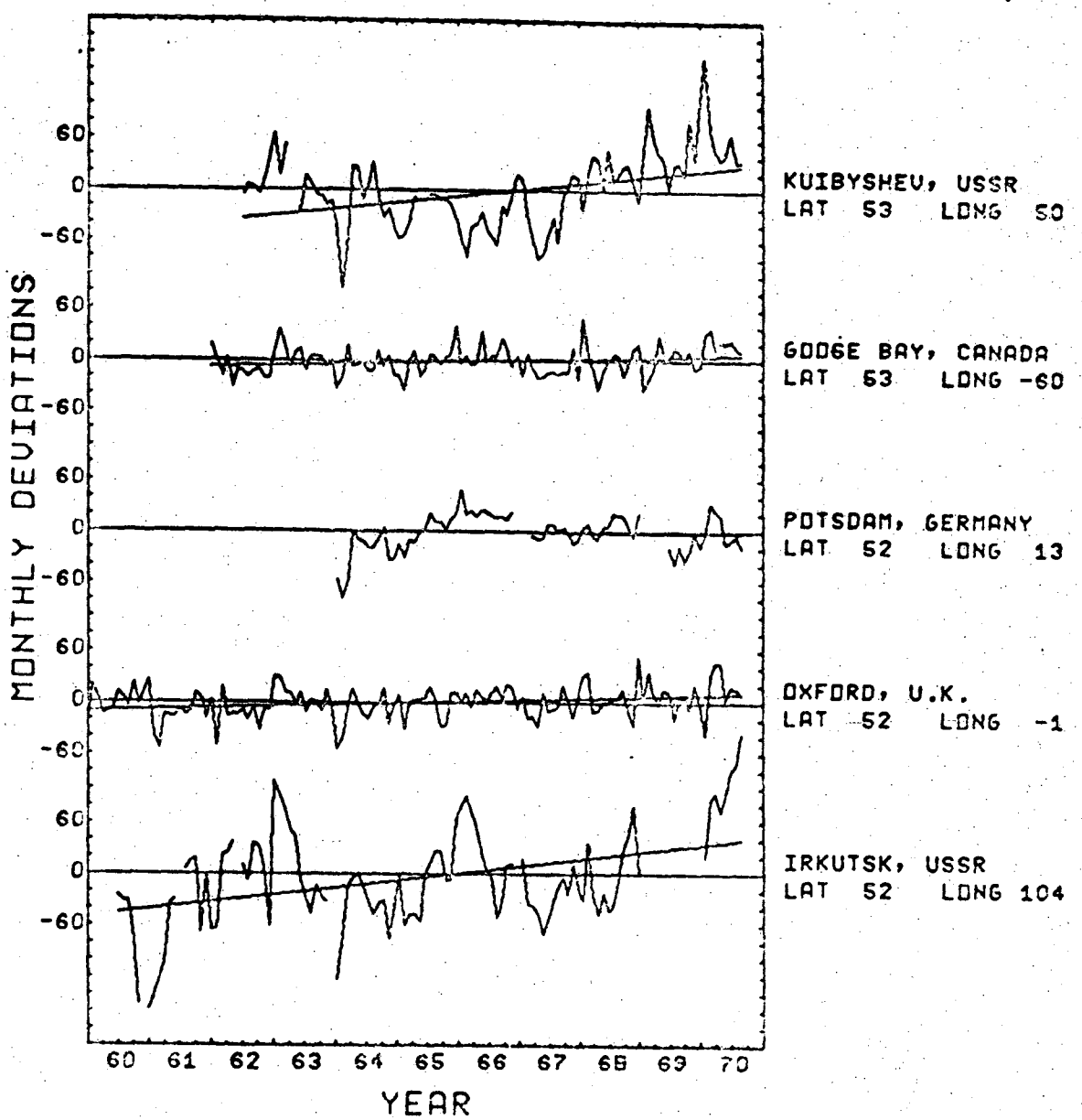
MONTHLY DEVIATIONS
60
0
-60
KUIBYSHEV, USSR
LAT 53 LONG 50
GOOSE BAY, CANADA
LAT 53 LONG -60
POTSDAM, GERMANY
LAT 52 LONG 13
OXFORD, U.K.
LAT 52 LONG -1
IRKUTSK, USSR
LAT 52 LONG 104
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig. 21

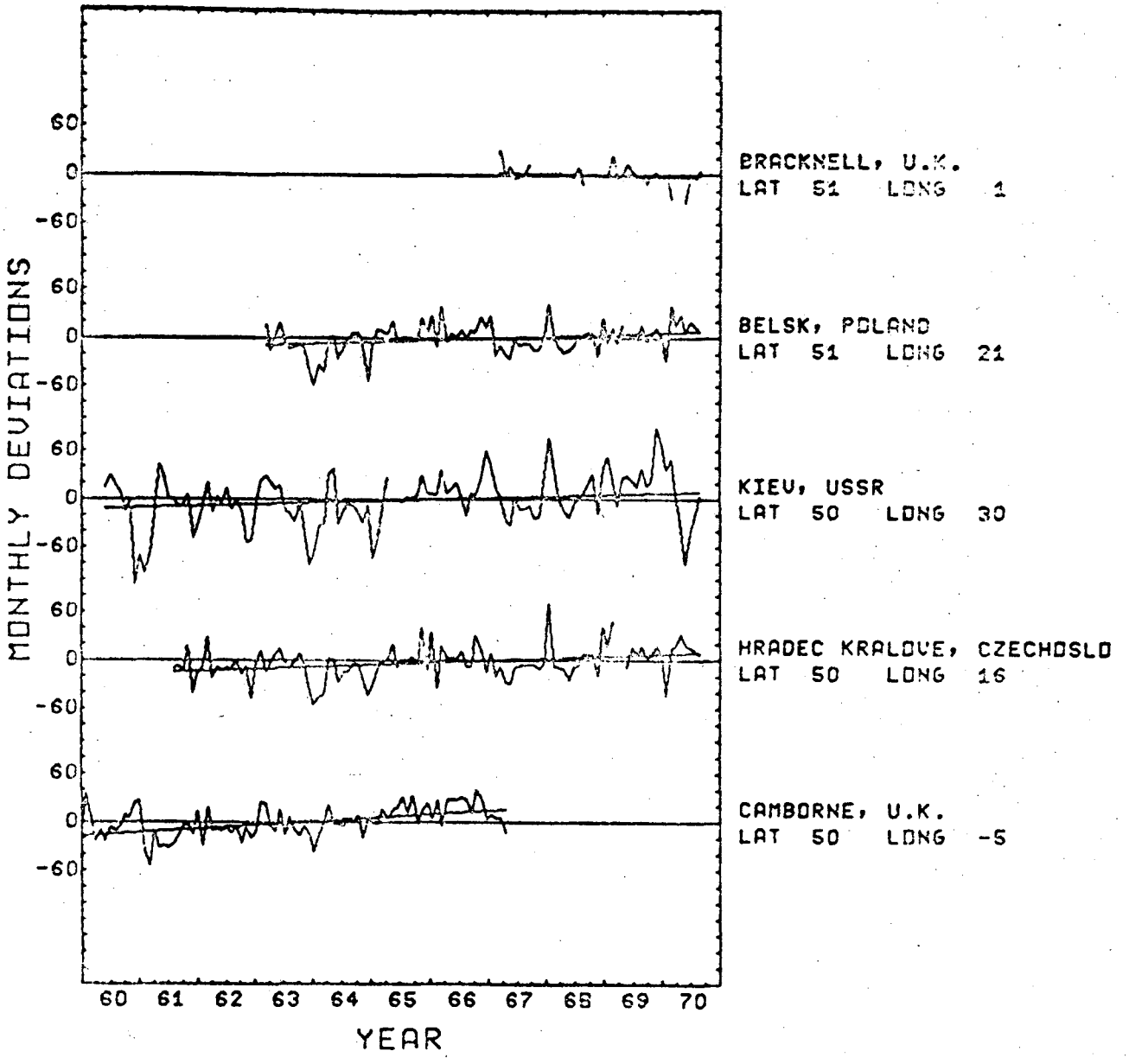
MONTHLY DEVIATIONS
60
0
-60
BRACKNELL, U.K.
LAT 51 LONG 1
BELSK, POLAND
LAT 51 LONG 21
KIEV, USSR
LAT 50 LONG 30
HRADEC KRALOVE, CZECHOSLO
LAT 50 LONG 16
CAMBORNE, U.K.
LAT 50 LONG -5
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig. 22

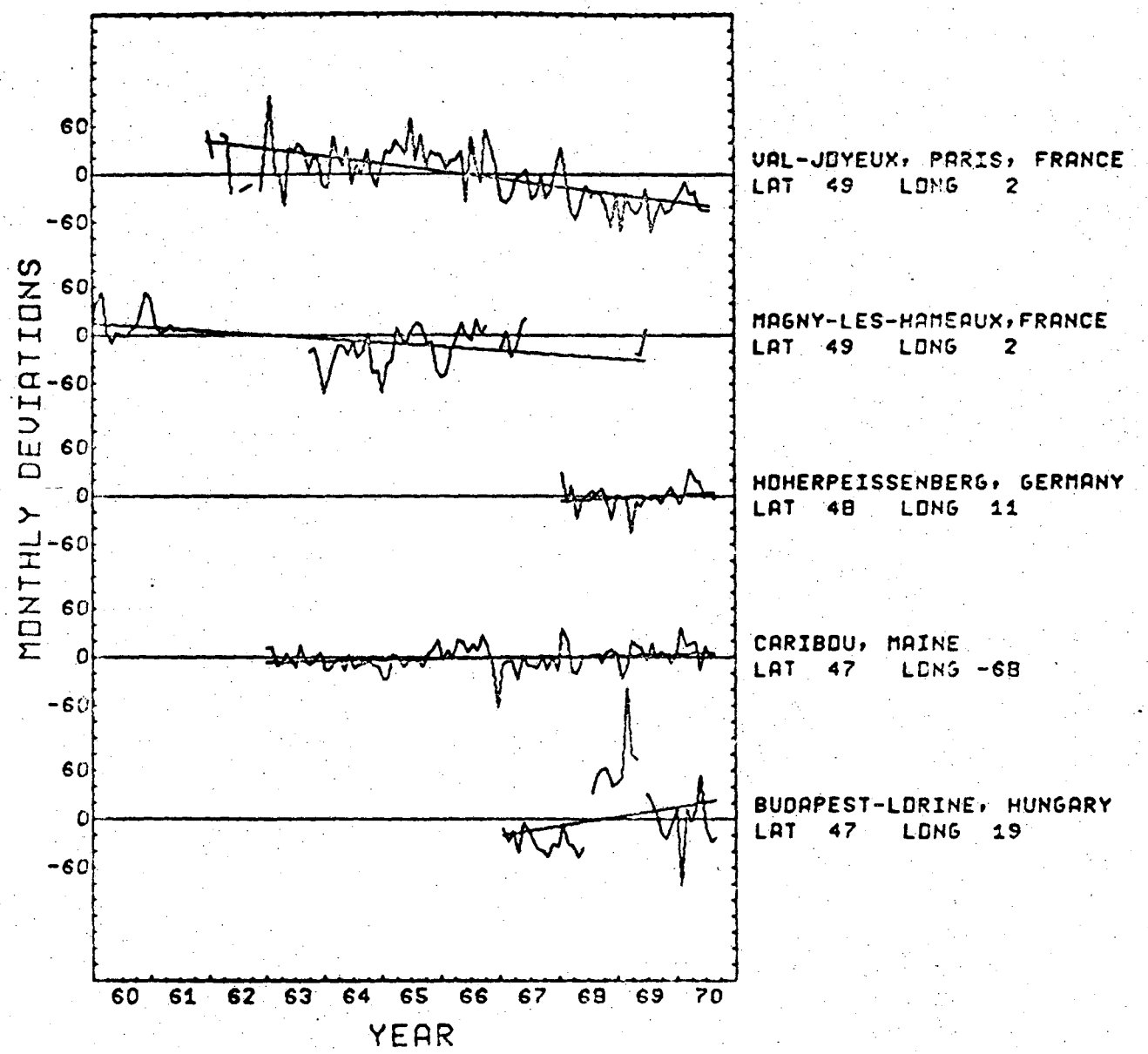
MONTHLY DEVIATIONS
60
0
-60
VAL-JOYEUX, PARIS, FRANCE
LAT 49 LONG 2
MAGNY-LES-HAMEAUX, FRANCE
LAT 49 LONG 2
HOHERPEISSENBERG, GERMANY
LAT 48 LONG 11
CARIBOU, MAINE
LAT 47 LONG -68
BUDAPEST-LORINE, HUNGARY
LAT 47 LONG 19
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig. 23

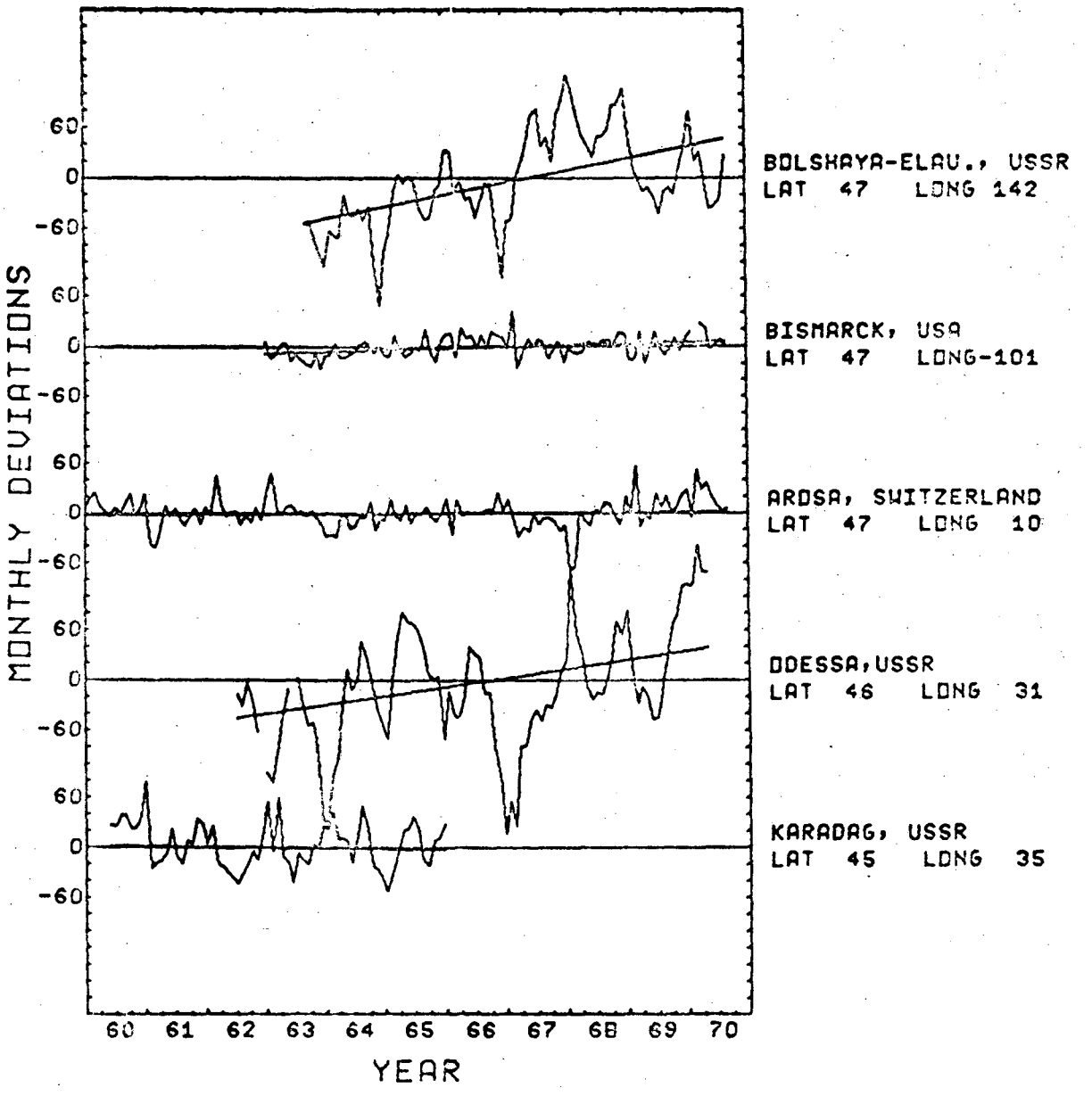
MONTHLY DEVIATIONS
60
0
-60
BOLSHAYA-ELAU., USSR
LAT 47 LONG 142
BISMARCK, USA
LAT 47 LONG-101
ARDSA, SWITZERLAND
LAT 47 LONG 10
ODESSA,USSR
LAT 46 LONG 31
KARADAG, USSR
LAT 45 LONG 35
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig. 24

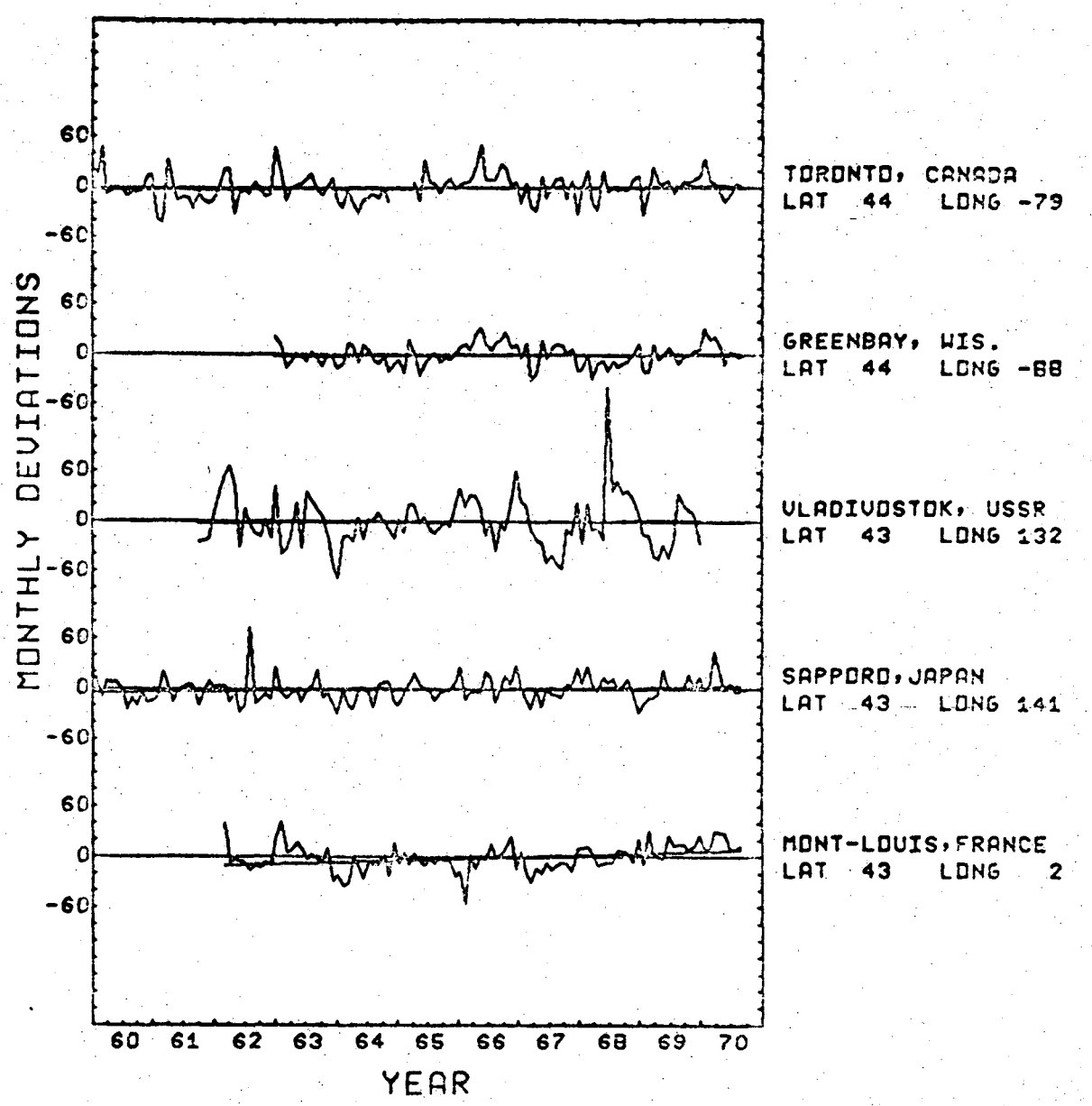
MONTHLY DEVIATIONS
60
0
-60
TORONTO, CANADA
LAT 44 LONG -79
GREENBAY, WIS.
LAT 44 LONG -88
VLADIVOSTOK, USSR
LAT 43 LONG 132
SAPPORO, JAPAN
LAT 43 LONG 141
MONT-LOUIS, FRANCE
LAT 43 LONG 2
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig. 25

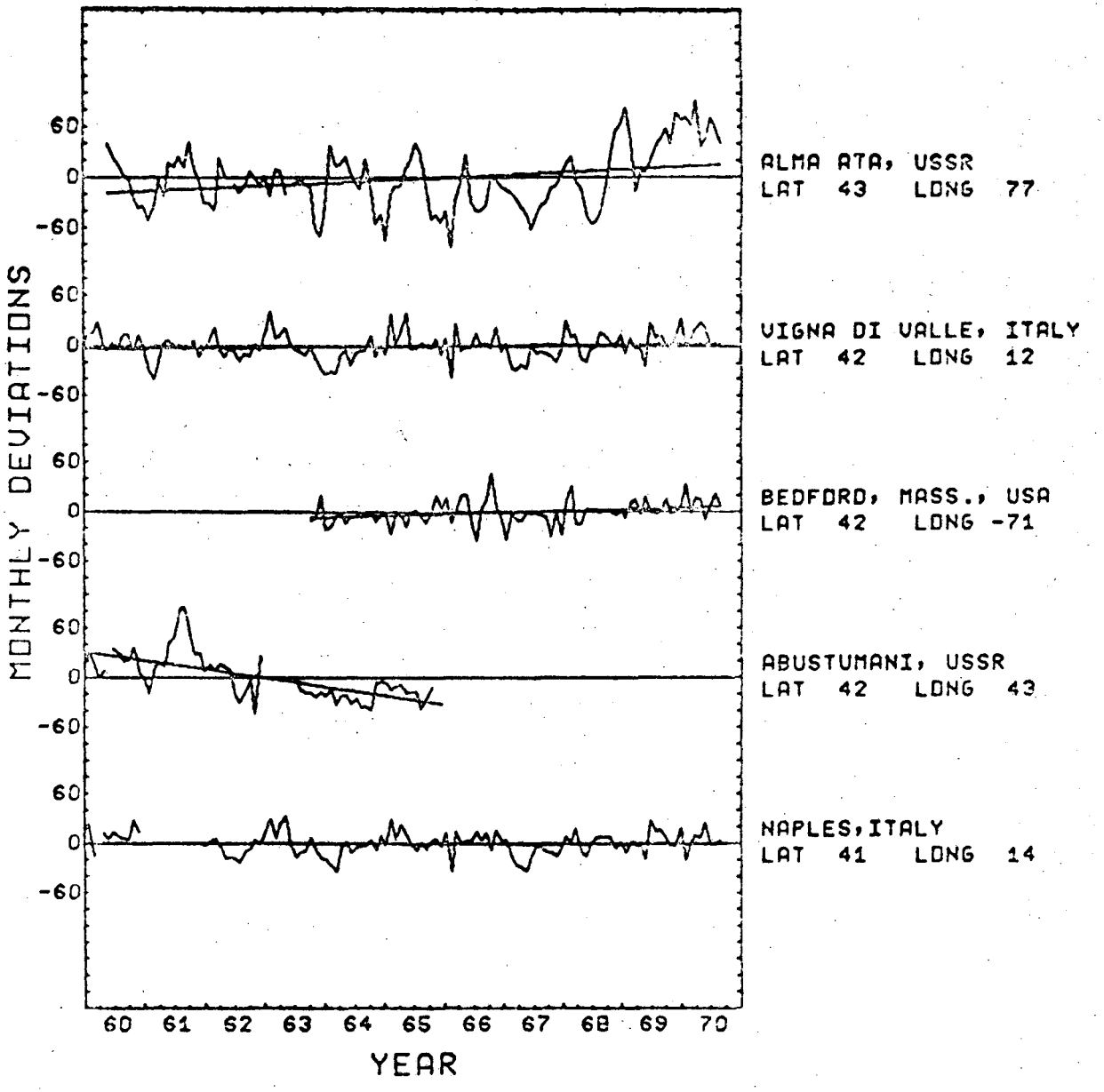
MONTHLY DEVIATIONS
60
0
-60
ALMA ATA, USSR
LAT 43 LONG 77
VIGNA DI VALLE, ITALY
LAT 42 LONG 12
BEDFORD, MASS., USA
LAT 42 LONG -71
ABUSTUMANI, USSR
LAT 42 LONG 43
NAPLES, ITALY
LAT 41 LONG 14
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig. 26

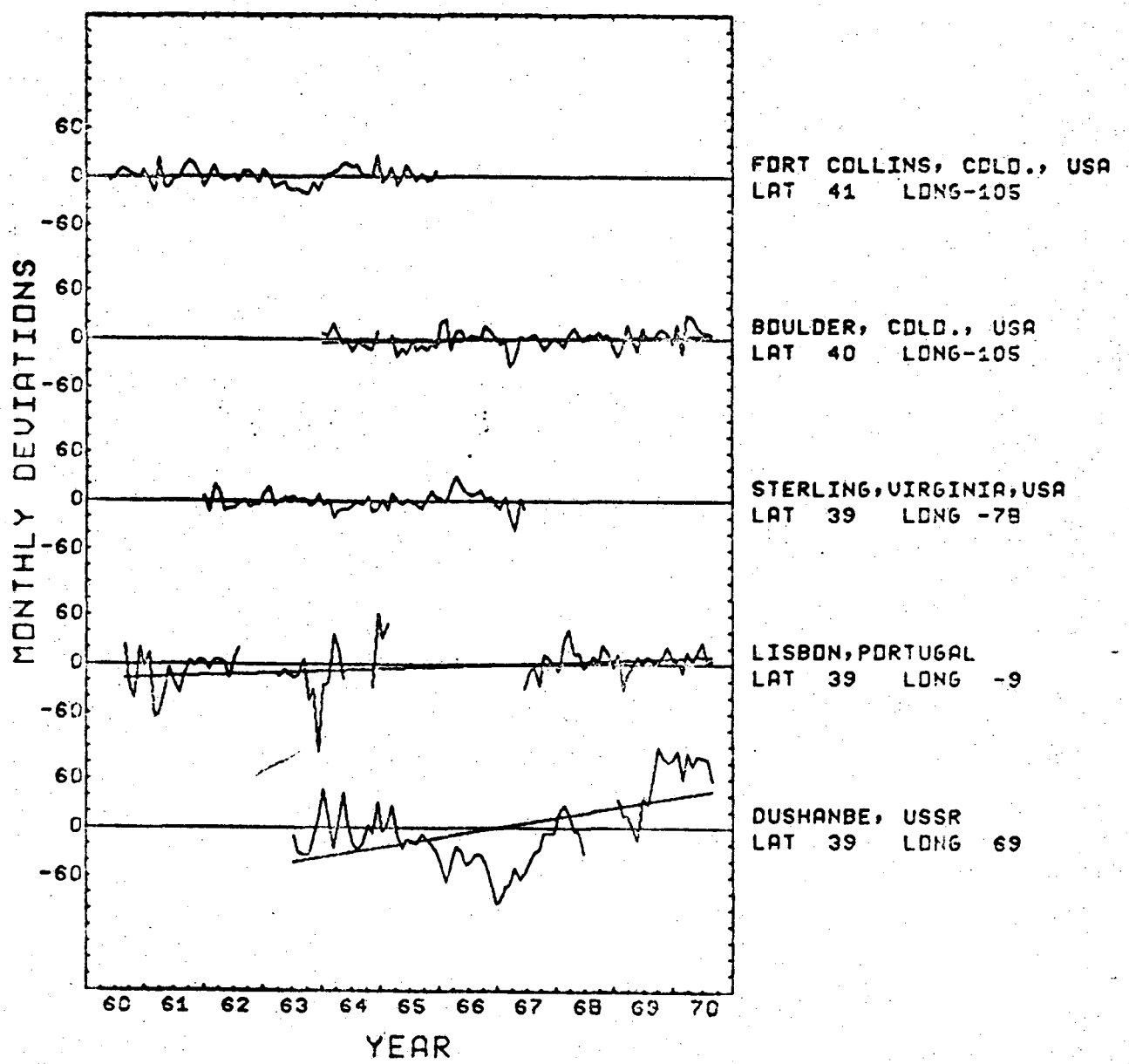
MONTHLY DEVIATIONS
60
0
-60
FORT COLLINS, COLO., USA
LAT 41 LONG-105
BOULDER, COLO., USA
LAT 40 LONG-105
STERLING,VIRGINIA,USA
LAT 39 LONG -78
LISBON,PORTUGAL
LAT 39 LONG -9
DUSHANBE, USSR
LAT 39 LONG 69
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig. 27

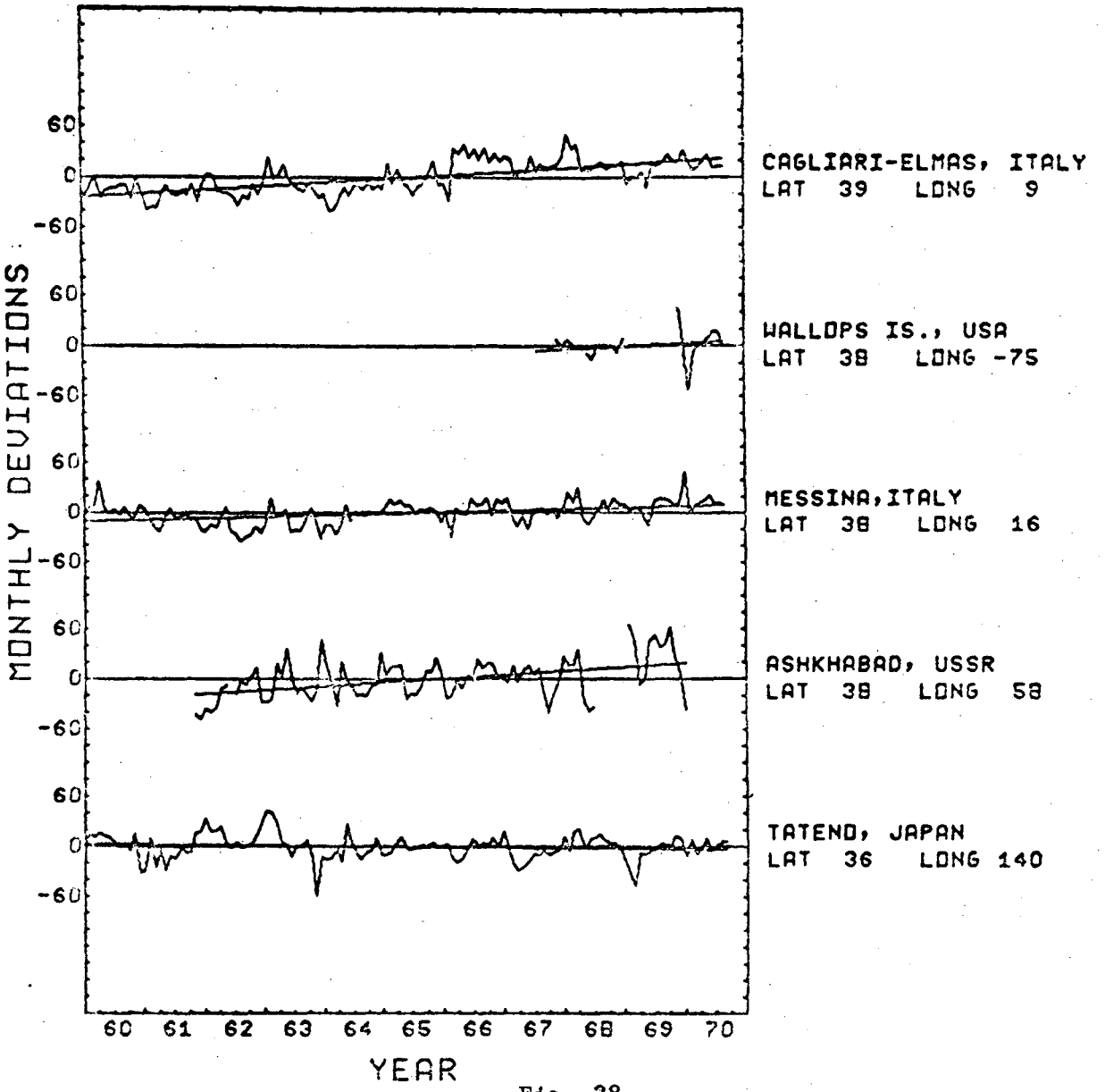
MONTHLY DEVIATIONS
60
0
-60
CAGLIARI-ELMAS, ITALY
LAT 39 LONG 9
WALLOPS IS., USA
LAT 38 LONG -75
MESSINA,ITALY
LAT 38 LONG 16
ASHKHABAD, USSR
LAT 38 LONG 58
TATENO, JAPAN
LAT 36 LONG 140
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig. 28

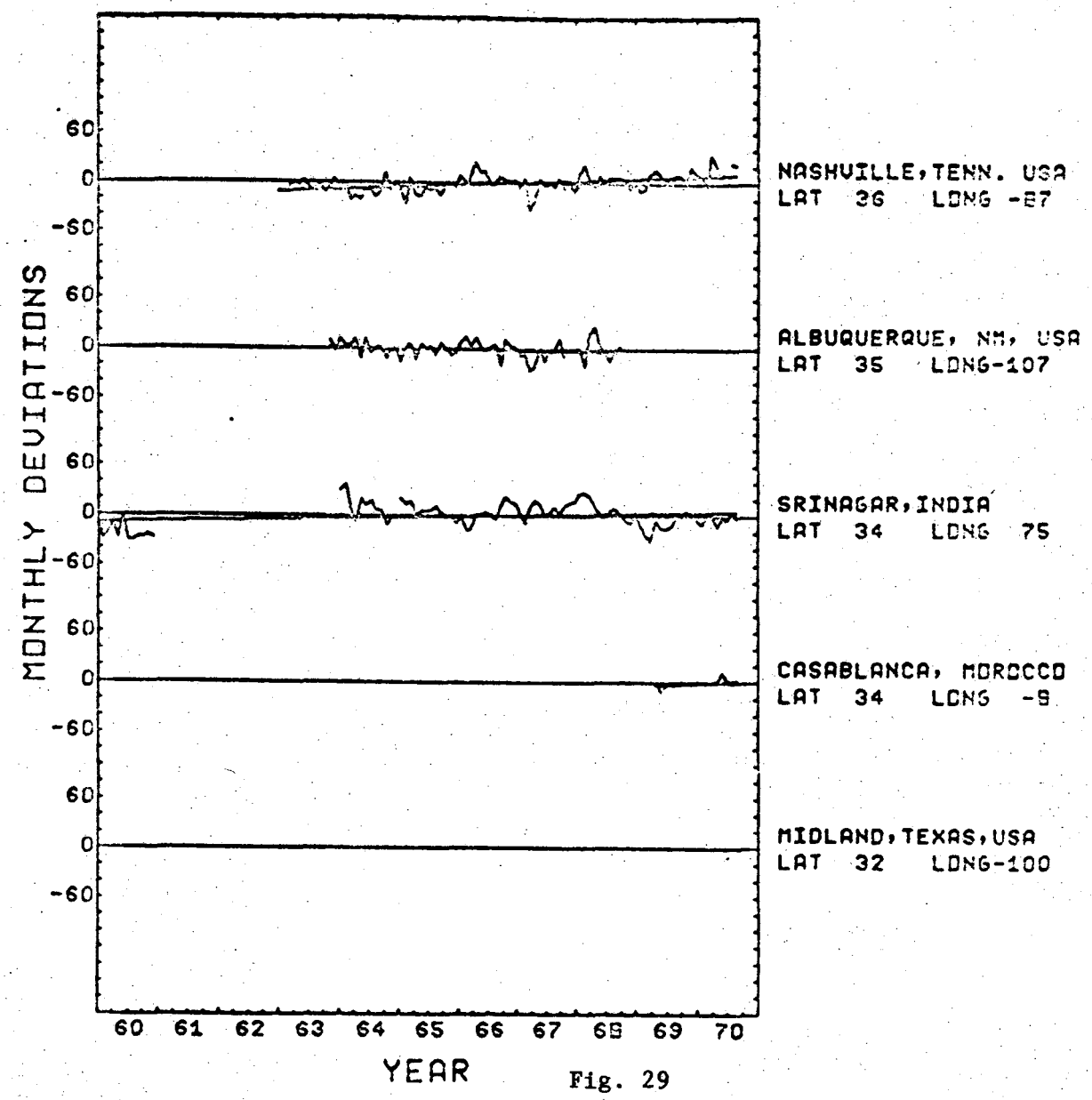
MONTHLY DEVIATIONS
60
0
-60
NASHVILLE,TENN. USA
LAT 36 LONG -87
ALBUQUERQUE, NM, USA
LAT 35 LONG-107
SRINAGAR,INDIA
LAT 34 LONG 75
CASABLANCA, MOROCCO
LAT 34 LONG -9
MIDLAND,TEXAS,USA
LAT 32 LONG-100
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig. 29

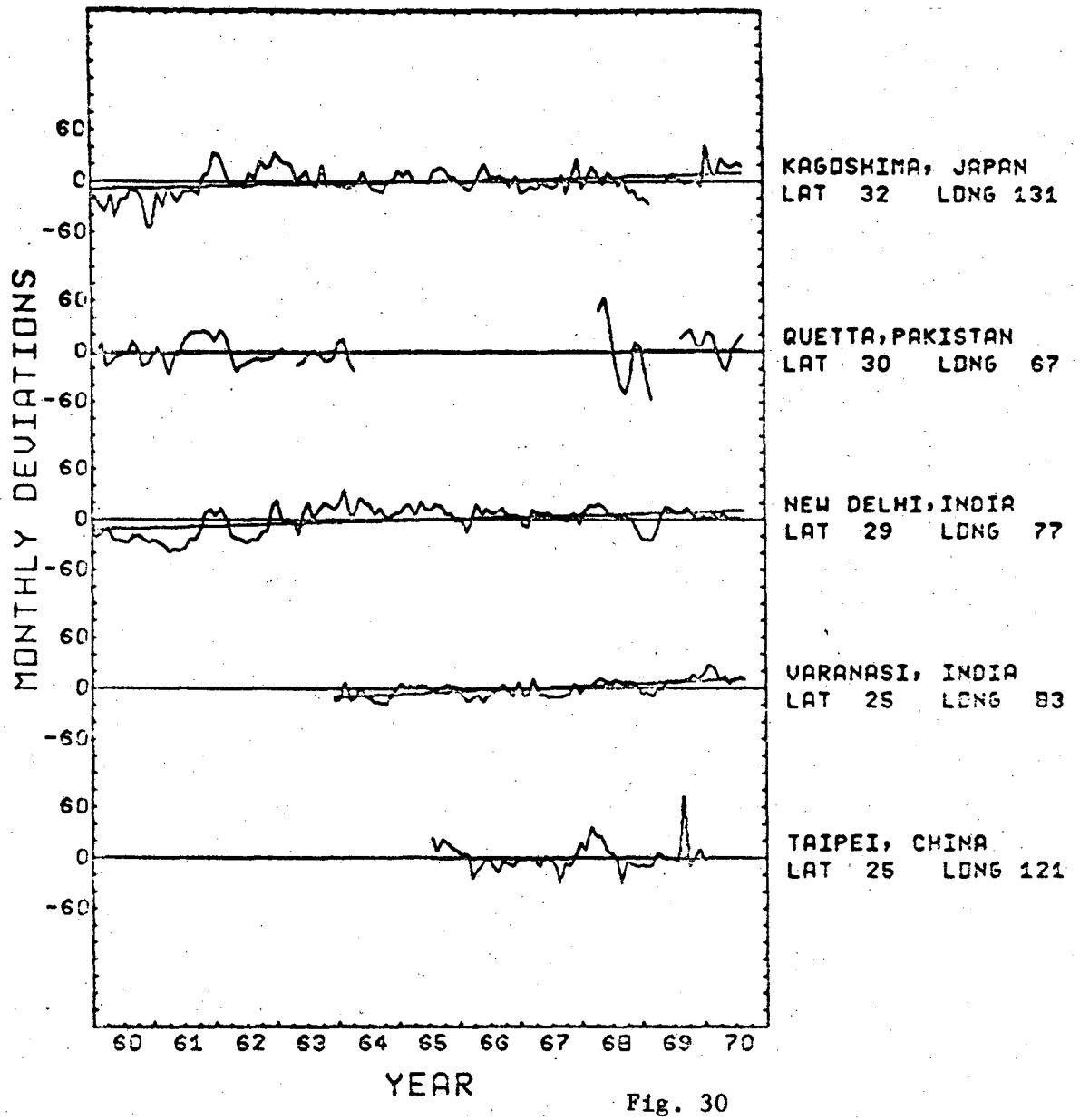
MONTHLY DEVIATIONS
60
0
-60
KAGOSHIMA, JAPAN
LAT 32 LONG 131
QUETTA, PAKISTAN
LAT 30 LONG 67
NEW DELHI, INDIA
LAT 29 LONG 77
VARANASI, INDIA
LAT 25 LONG 83
TAIPEI, CHINA
LAT 25 LONG 121
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig. 30

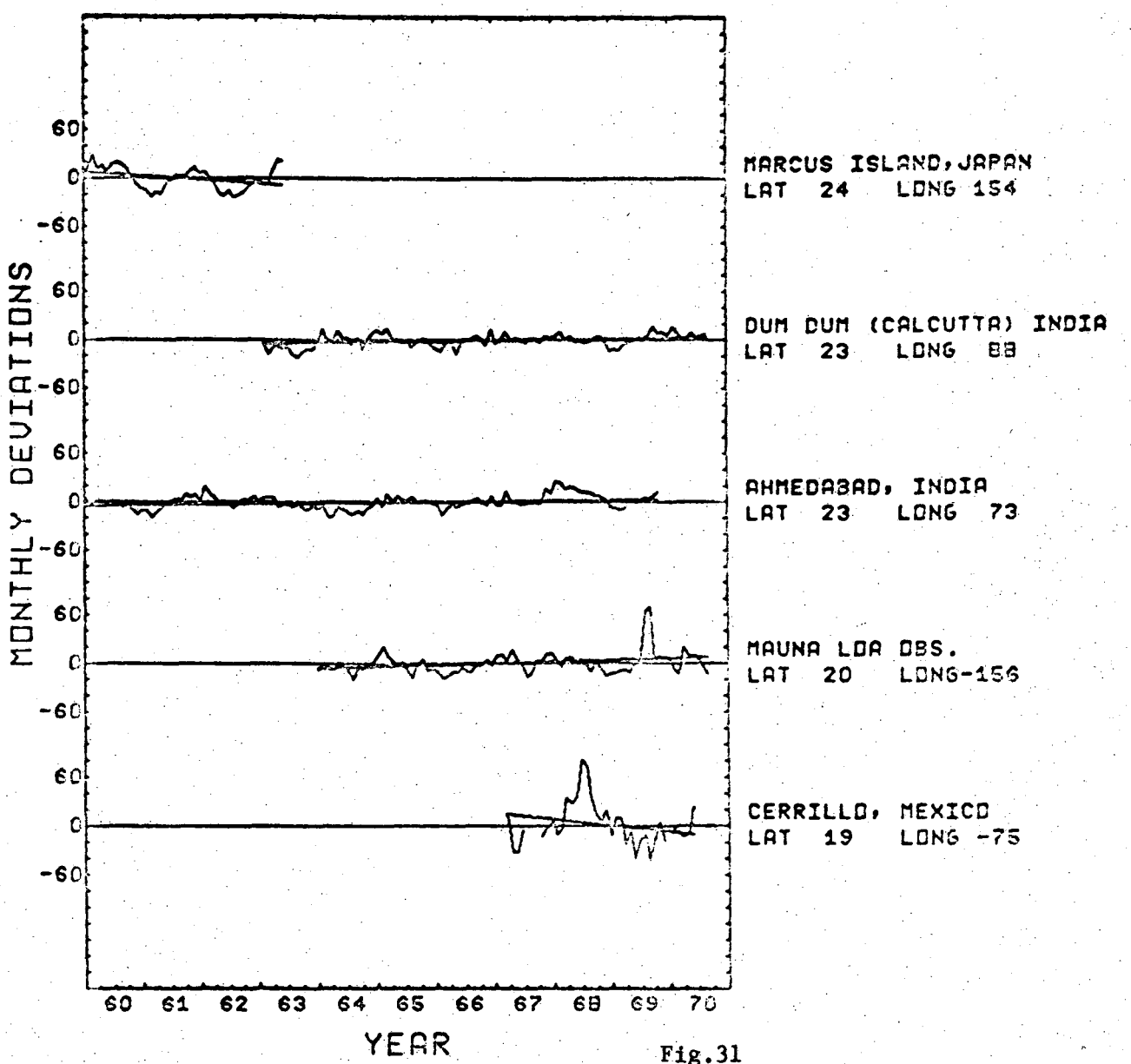
MONTHLY DEVIATIONS
60
0
-60
MARCUS ISLAND, JAPAN
LAT 24 LONG 154
DUM DUM (CALCUTTA) INDIA
LAT 23 LONG 88
AHMEDABAD, INDIA
LAT 23 LONG 73
MAUNA LOA OBS.
LAT 20 LONG-156
CERRILLO, MEXICO
LAT 19 LONG -75
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig.31

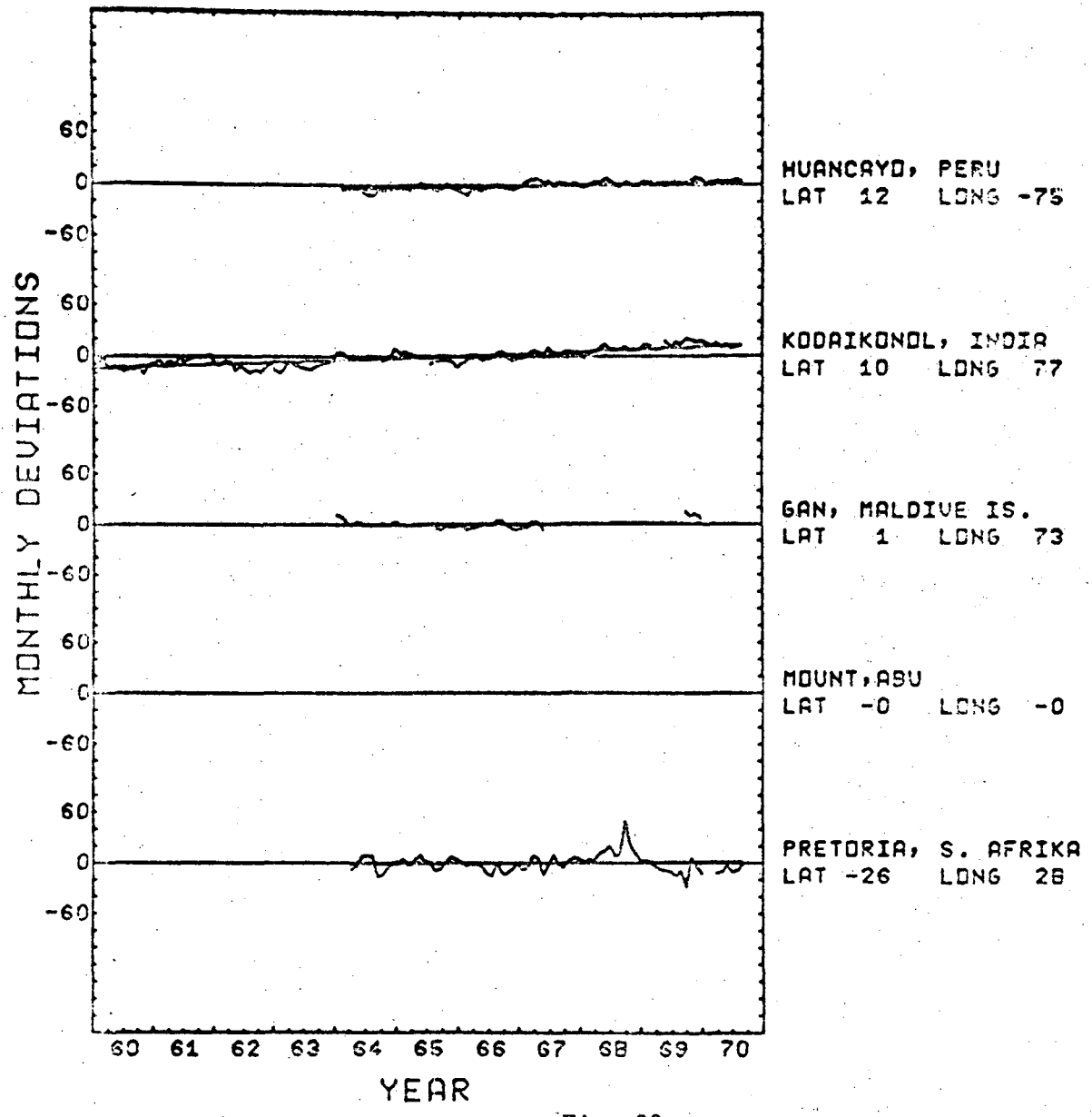
MONTHLY DEVIATIONS
60
0
-60
HUANCAYO, PERU
LAT 12 LONG -75
KODAIKONDL, INDIA
LAT 10 LONG 77
GAN, MALDIVE IS.
LAT 1 LONG 73
MOUNT,ABU
LAT -0 LONG -0
PRETORIA, S. AFRIKA
LAT -26 LONG 28
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig. 32

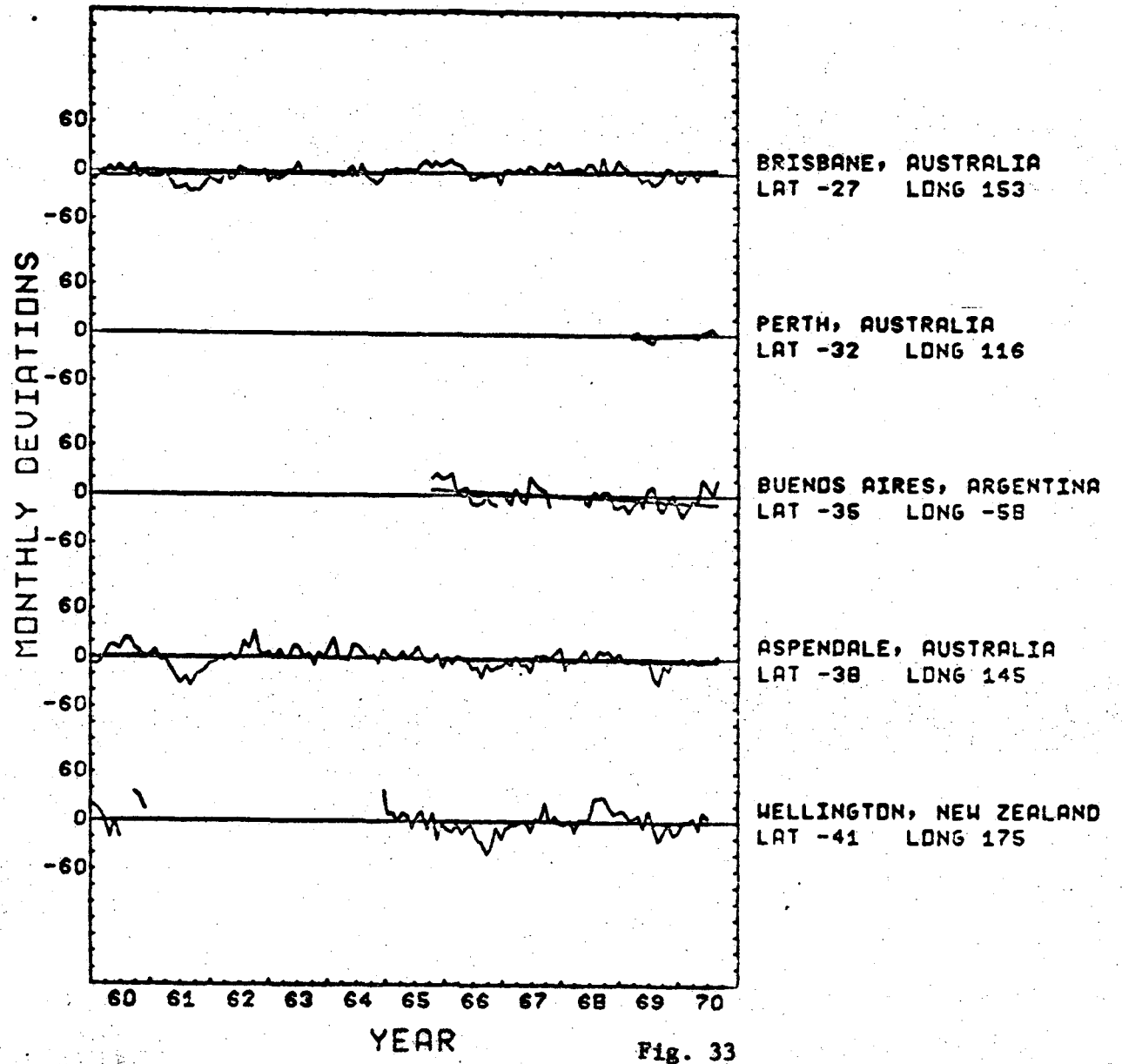
MONTHLY DEVIATIONS
60
0
-60
60
0
-60
60
0
-60
60
0
-60
60
0
-60
60 61 62 63 64 65 66 67 68 69 70
YEAR
BRISBANE, AUSTRALIA
LAT -27 LONG 153
PERTH, AUSTRALIA
LAT -32 LONG 116
BUENOS AIRES, ARGENTINA
LAT -35 LONG -58
ASPENDALE, AUSTRALIA
LAT -38 LONG 145
WELLINGTON, NEW ZEALAND
LAT -41 LONG 175

Fig. 33

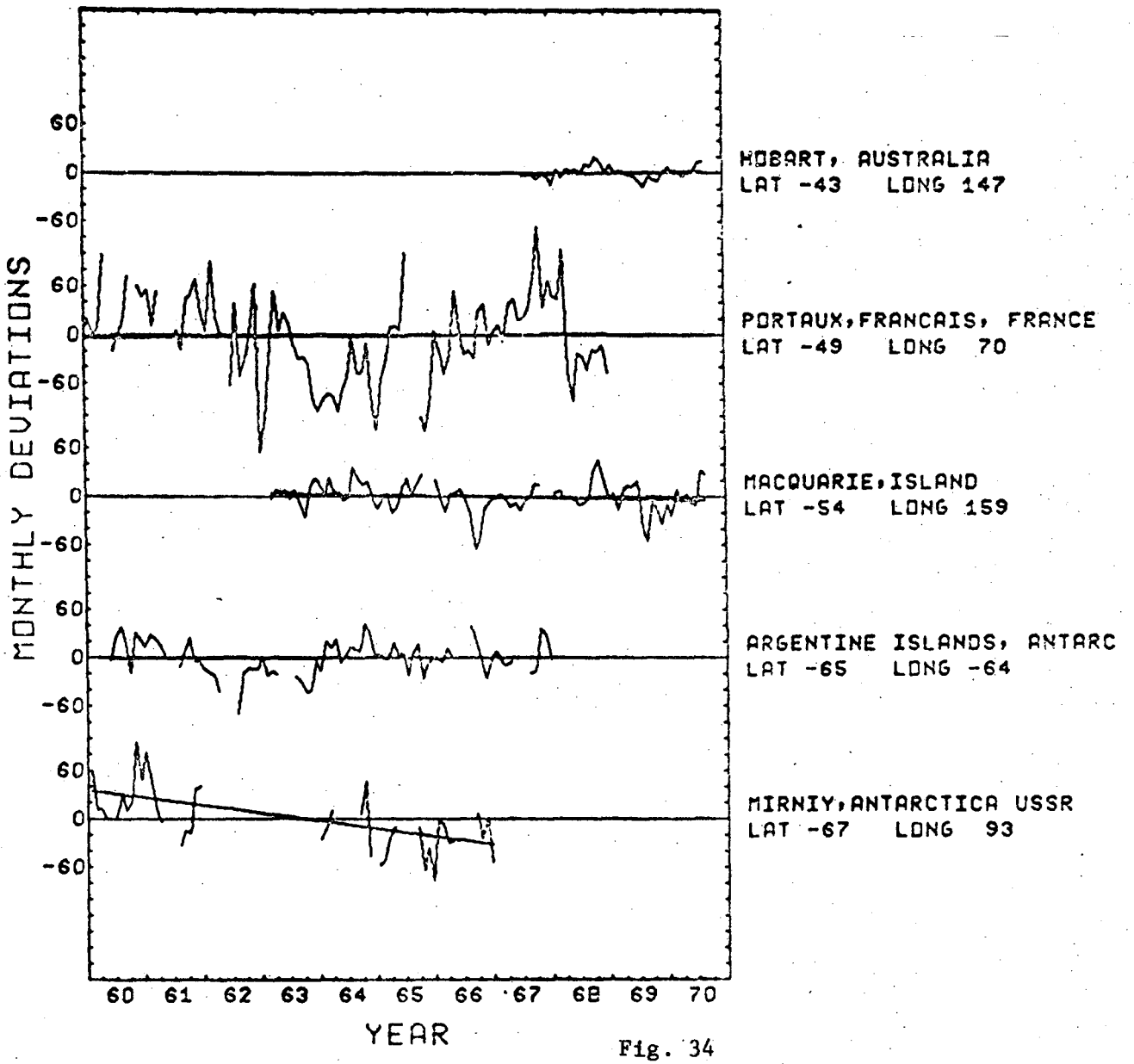
MONTHLY DEVIATIONS
60
0
-60
HOBART, AUSTRALIA
LAT -43 LONG 147
PORTAUX,FRANCAIS, FRANCE
LAT -49 LONG 70
MACQUARIE,ISLAND
LAT -54 LONG 159
ARGENTINE ISLANDS, ANTARC
LAT -65 LONG -64
MIRNIY,ANTARCTICA USSR
LAT -67 LONG 93
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig. 34

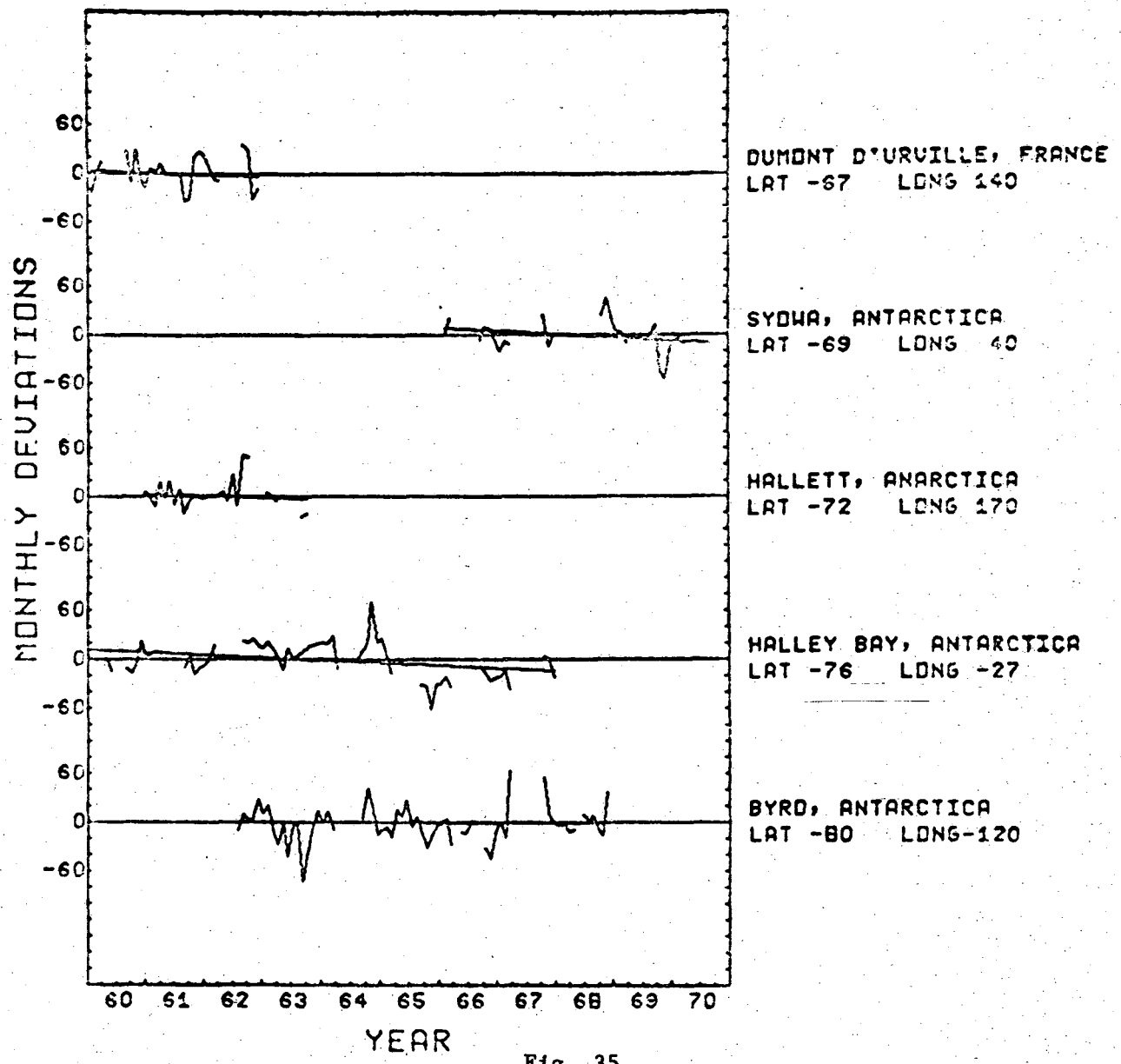
MONTHLY DEVIATIONS
60
0
-60
DUMONT D'URVILLE, FRANCE
LAT -67 LONG 140
SYOWA, ANTARCTICA
LAT -69 LONG 40
HALLETT, ANARCTICA
LAT -72 LONG 170
HALLEY BAY, ANTARCTICA
LAT -76 LONG -27
BYRD, ANTARCTICA
LAT -80 LONG-120
60 61 62 63 64 65 66 67 68 69 70
YEAR

Fig. 35

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