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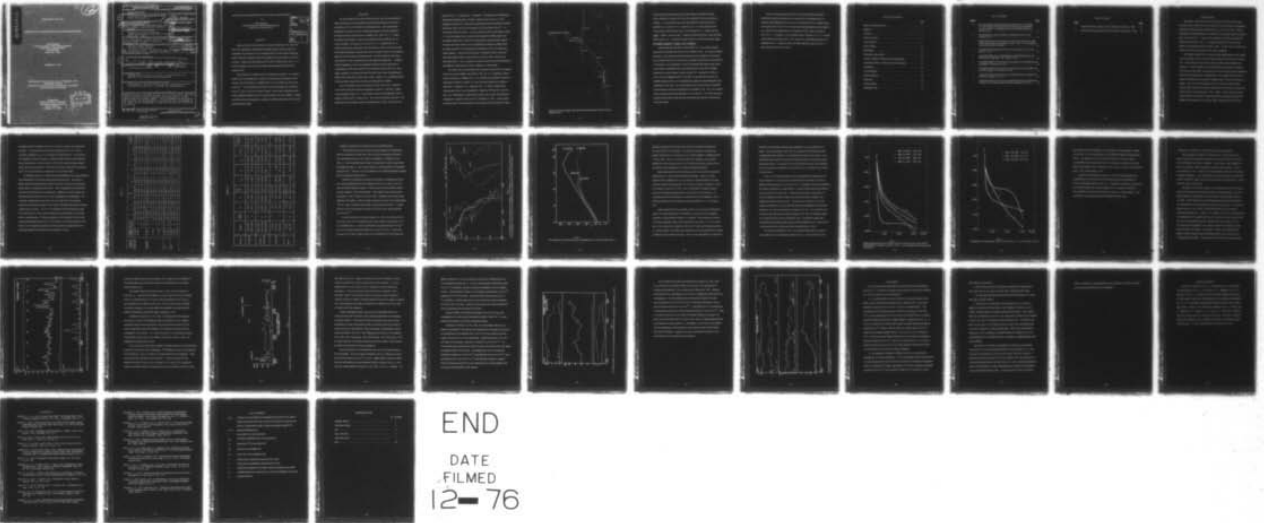
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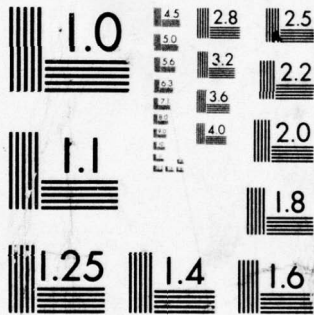
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AITKEN NUCLEI MEASUREMENTS IN THE LOWER STRATOSPHERE

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September 27, 1976

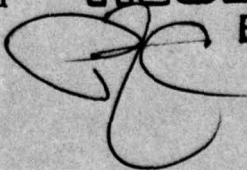
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Aitken nuclei (AN) were measured during 18 flights between 6 km and 19 km over the U.S.A., Central and South America with the new Stratospheric Aitken Nuclei Detection System (SANDS). The instrument was designed by the staff of the GE laboratory in Pittsfield, MA, calibrated in the laboratory of GCCPR-UMR in Rolla and installed on the WB-57F aircraft.		

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AITKEN NUCLEI MEASUREMENTS IN THE LOWER STRATOSPHERE

by

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ABSTRACT

Aitken nuclei (AN) measurements have been made between 6 km and 19 km with a new Stratospheric Aitken Nuclei Detection System (SANDS), which was calibrated in the laboratory of The Graduate Center for Cloud Physics Research at the University of Missouri-Rolla and installed in the nose section of a WB-57F aircraft. Samples were taken between 48°N and 9°S latitudes above the U. S. A., the Gulf of Mexico, and Central and South America between March 1974 and February 1975.

The evaluation of 18 flights leads to the following conclusions: The vertical profiles of AN concentrations are similar to those found by Junge et al. (1961); however, the concentrations at altitudes above 17 km are higher by several tens of AN cm⁻³. AN concentrations recorded on horizontal flights are low (below 20 AN cm⁻³), above 18 km in the tropical regions, and the Rocky Mountains and the Andes Mountains influence on the AN measurements at the 18 km altitude. Usually, several km above the tropopause, a negative correlation between the AN and ozone concentrations is found.

FOREWORD

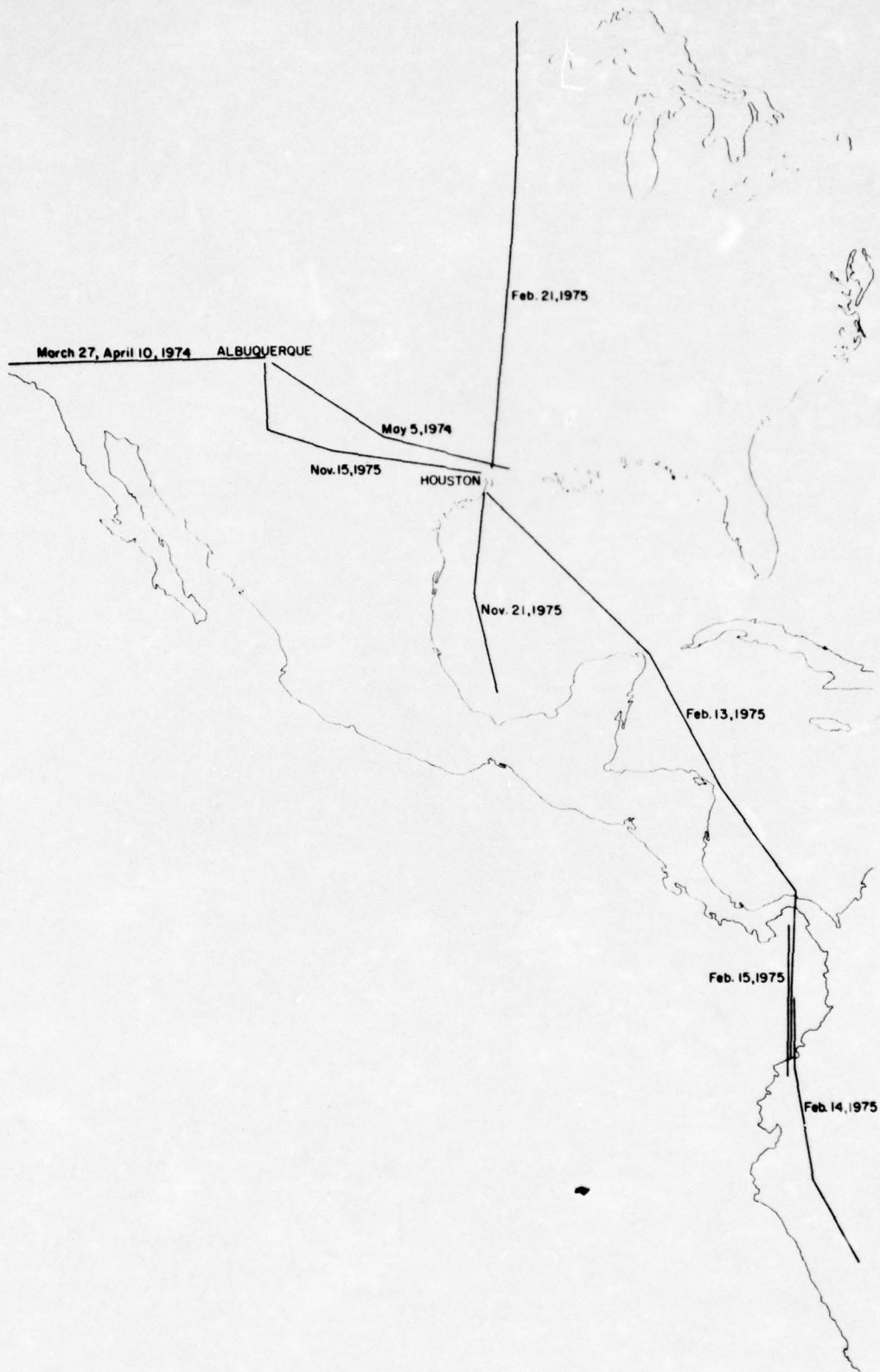
The main results of the measurements that were made of concentrations of Aitken nuclei in the lower stratosphere during the period between March 1974 and February 1975 are recorded in the following report. The measurements themselves were taken in a well orchestrated program of several institutions: the General Electric Laboratories in Pittsfield, MA, where Mr. J. B. Haberl developed the Stratospheric Aitken Nuclei Detection System (SANDS), the Los Alamos Scientific Laboratories, NM, where Dr. W. A. Sedlacek and Mr. P. Guthals supervised the installation of the instrument on board a WB-57F aircraft and monitored the flight measurements; and the Graduate Center for Cloud Physics Research of the University of Missouri-Rolla, (GCCPR-UMR) where the author and his fellow workers calibrated SANDS and analyzed the flight data. In addition to these institutions, which were supported by grants from the Department of Transportation (DOT) and the Office of Naval Research (ONR) of the Navy, many pilots, navigators, and technicians from the USAF 58th Weather Reconnaissance Squadron and of the NASA Johnson Space Center, Houston, TX, contributed greatly to the successful measurement of Aitken nuclei during the 18 flights.

The work started in 1972 at the General Electric Laboratory in Pittsfield. The GCCPR-UMR staff assisted in designing the counter (J. Podzimek, "Report on the Feasibility Study of GE - Aitken Nuclei Counter for Stratospheric Levels", Report to ONR and DOT, October 1972). The work was carried on until 1975. The performance of the SANDS counter was checked twice in Rolla, once in 1974 and

again in 1975 (J. E. Wegrzyn and J. Podzimek: "Calibration of the Stratospheric Aitken Nuclei Detection System-SANDS", Report to ONR, March 19, 1975).

Beginning in March 1974, systematic measurements of the concentration of Aitken nuclei were made in the lower stratosphere over the territory of the U. S. A. and Central and South America. (The area covered is shown in the figure on page 5.) The main objective was to measure the background of Aitken nuclei in the stratosphere before SST commercial aircraft begin to operate in these regions. The other aim of the research was to investigate the exchange of Aitken nuclei over high mountains, in the Intertropical Convergence Zone, and around large frontal systems. There was some hope that a correlation could be made with other measurements taken on board the same aircraft, such as of ozone and large particulates. Also, an attempt was made to measure the size distribution of stratospheric Aitken nuclei from their mobility in an electric field.

The results of the measurements were evaluated in Rolla with the assistance of the Los Alamos Scientific Laboratories, NM, (Dr. W. A. Sedlacek), General Electric Laboratory, Pittsfield, MA, (J. B. Haberl) and The National Center for Atmospheric Research, Boulder, CO, and were summarized in two technical reports by J. Podzimek, W. A. Sedlacek, and J. B. Haberl ("Aitken Nuclei Measurements in the Lower Stratosphere", Reports to ONR and DOT, April 24, 1975, and "Peculiarities in Aitken Nuclei Counts in the Tropical and Subtropical Stratosphere", Reports to ONR and DOT, November 27, 1975). These reports formed the basis of an invited paper on the measurement of stratospheric Aitken



Survey of horizontal flights with SANDS performed between March 1974 and February 1975.

nuclei that was presented during the Climatic Impact Assessment Program (CIAP) conference in Boston (1975) and will be published in the conference proceedings. The geophysical aspects and interpretations of the studies will be published by J. Podzimek, W. A. Sedlacek, and J. B. Haberl under the title "Aitken Nuclei Measurements in the Lower Stratosphere" in a Swedish journal, Tellus. Finally, a special study, "Possible Correlation Between Ozone and Aitken Nuclei Counts", by J. Podzimek will appear this year in the Swiss journal, Pure and Applied Geophysics (Geofisica Pura e Applicata).

Aitken nuclei measurements with the SANDS was, in the author's opinion, prematurely terminated with the CIAP in the middle of 1975. As in other scientific programs, many questions remained unanswered at the completion of the project for the simple reason that many questions emerged during the actual measurements. For example, it was impossible to check the extremely high Aitken nuclei counts in the Alaskan stratosphere that were found during the whole series of flights performed over polar regions in April and May 1975. Because one of the two valves on the air supplying units of the SANDS was probably frozen, there was justified suspicion that the very interesting data might have been influenced by the malfunction of the valve. Few measurements were made around the well defined frontal systems and above the Intertropical Convergence Zone. Also, the measurements of Aitken nuclei size distribution are not conclusive. The author hopes that someone will use these rough data and incorporate them with new measurements in the near future.

There are many problems that have to be solved before the calibrated instrument can deliver the data that are necessary for formulating basic conclusions about existing pollution in the lower stratosphere and of a possible impact of commercial SST aircraft on the content of small particulates in the stratosphere. As a result of the understanding and wise guidance of the Program Manager of DOT, Dr. A. J. Grobecker, and his Deputy Program Manager, Dr. S. C. Coroniti, many problems have been solved. It is a pleasure to acknowledge the advice and assistance of Mr. J. Hughes of ONR, who supervised the program for the U. S. Navy (Contract N00014-75-C-0413).

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INTRODUCTION

The purpose of this final report is to summarize the 1974 and 1975 measurements made of Aitken nuclei (AN) with the new General Electric Stratospheric Aitken Nuclei Detection System (SANDS) flown on a WB-57F aircraft. Detailed descriptions of individual flights were presented in two separate reports (Podzimek et al. 1975a and 1975b). A report on the calibration of SANDS was prepared by Wegrzyn and Podzimek in 1975. The technical parameters of SANDS and its function were published elsewhere (Haberl, 1975). For this reason, only the main results of the SANDS calibration in Rolla will be mentioned.

This report is based on the evaluation of high altitude flights over the middle and lower latitudes of the American continents between March 1974 and February 1975. In total, nine vertical aircraft ascents and nine horizontal flights were evaluated. On several flights, ozone measurements accompanied AN counting. The main goal of this study, encompassed in the broad Climatic Impact Assessment Program of the Department of Transportation, was to supply data for the background concentrations of stratospheric pollutants before the SST aircraft begin to operate. Also, the AN measurements were to be compared with the balloon measurements of Junge and his fellow workers (1961) and with those of Rosen and Hofmann (1974) and Kasselau et al. (1974). Finally, SANDS in being carried by a high altitude aircraft offered a unique opportunity to investigate the space distribution and exchange of AN over large regions including those of the tropics.

CALIBRATION OF THE SANDS.

Calibration of the General Electric SANDS was performed by comparing its performance with that of the UMR Absolute Aitken Nuclei Counter (AANC) twice during the years of 1974 and 1975. The first comparison of both instruments was made at the beginning of 1974. The latter instrument has been described several times in the literature, e. g., Kassner (1968). In November of 1974, after being flown on the WB-57F aircraft for several months, the SANDS was slightly modified. This necessitated another calibration.

Both calibrations of the SANDS by the AANC were performed without the Pressurization Unit; however, the function of all parts of the SANDS was checked at stratospheric conditions in the environmental chamber at the Los Alamos Scientific Laboratories. Orientational measurements with room aerosol in the General Electric Laboratory showed that the nuclei losses in the Pressurization Unit of the SANDS did not surpass 15% of the measured concentration (see Wegrzyn and Podzimek, 1975, p. 11).

Six different kinds of test aerosol were used: room air, residual nuclei after the propane droplets evaporated, sodium chloride, silver iodide, nichrome, and gold aerosol. Silver iodide nuclei were generated by heating the substance with an electric wire coil. Sodium chloride nuclei were originally generated by an ultra-acoustic mist generator. During the second calibration sodium chloride nuclei were generated by bubbling the nitrogen gas through the salt solution. Nichrome nuclei were emitted from a heated wire in a nitrogen gas flow, and the gold particles

were generated by an exploding wire technique (a 9 μ F capacitor discharged at 4000 V through the wire). The modal size (diameter) of the gold aerosol was 0.025 μ m. The monodispersity factor, $\alpha = \sigma/\bar{r}$, was in the first case 0.532 and in the second 0.322. The nuclei size spectrum was obtained by evaluating the electron micrographs of the particles deposited in electrostatic precipitator.

After eliminating the coarse particles, the test aerosol was introduced into the 18 m³ storage chamber with an inflatable mylar bag inside. This large volume storage facility allowed both counters to be compared over long periods of time and enabled one to reach very low nuclei concentrations, which simulate stratospheric conditions.

During the first series of experiments, the SANDS, originally calibrated by a Nolan-Pollak counter, consistently showed lower counts than the AANC by more than 30%. After the adjustment of the instrument in the fall of 1974, the counts of both instruments were very close. Because of the apparent proportionality of the data from the SANDS (Y) to the AANC (X), a simple linear regression analysis was used in which the predicted value of the nuclei concentration measured by SANDS (\hat{Y}) is

$$\hat{Y} = b_0 + b_1 X,$$

in which

$$b_1 = \frac{\sum(X_i - \bar{X})(Y_i - \bar{Y})}{\sum(X_i - \bar{X})^2} \quad \text{and} \quad b_0 = \bar{Y} - b_1 \bar{X}.$$

All summations corresponding to the pairs of simultaneous measurements

by SANDS and AANC progress from $i=1$ to n , and X is a given value measured by the AANC. From these data, the estimated standard error (E. S. E.) and the correlation coefficient (r_{xy}) were calculated in a well-known way. The results of the calculation of these parameters for different kinds of nuclei and for different nuclei concentrations are summarized in Table I (first series of measurements) and in Table II (second series). Both tables are similar except that the calculated ratio \bar{X}/\bar{Y} of the second table corresponds to \bar{Y}/\bar{X} in the first table.

In conclusion, there was a satisfactory reproducibility of data, high sensitivity, and comparability of the SANDS measurements with the AANC. The counts of sodium chloride nuclei showed much better correlation with the AANC during the second series in comparison with the first. This is explained by the fact that the aerosol for the second series was prepared in a different way and in general was characterized by smaller sizes and a more monodisperse size distribution. The correlation coefficients for both series were very high for all concentration ranges and were generally higher for the smaller well defined particulates (propane residual nuclei and gold). The E. S. E. for different nuclei concentration ranges was smaller than 12% of the measured Aitken nuclei concentration in the second series of measurements. All these parameters, the low threshold of the SANDS measurements (approximately 10 AN cm^{-3}) and the very fast time sequence of the counts (2.5 air samples per second), were the necessary prerequisites for the successful operation of the SANDS in the stratospheric levels.

TABLE I.

TYPE OF AEROSOL	CONCEN. RANGE	NO. OF CASES	\bar{X}	\bar{Y}	$\frac{\bar{X}}{\bar{Y}}$	r_{xy}	b_0	b_1	E.S.E.	$\frac{E.S.E.}{\bar{Y}}$
ROOM AIR	0-100	9	83.4444	117.9111	70.7690	0.8731	47.4140	0.8448	12.7436	10.8078
	0-300	7	112.4286	163.8571	68.6138	0.8860	-68.2323	2.0643	39.6355	24.1890
		18	195.7778	256.1665	76.4260	0.7730	57.0347	1.0171	15.5684	6.0775
		16	199.4375	299.5625	66.5762	0.9193	-908.4529	6.0571	87.6869	29.2716
PROPANE	0-100	12	49.7083	71.1333	69.8805	0.9680	-6.4861	1.5615	12.4409	17.4896
		9	55.3333	88.6667	62.4060	0.9495	26.7550	1.1189	10.1477	11.4448
	0-300	4	185.0000	311.0000	59.4835	0.9909	-30.0776	1.8437	60.6955	19.5162
		9	213.3333	317.2222	67.2504	0.9372	38.4299	1.3068	49.9861	15.7574
	0-1000	15	565.8665	684.8665	82.6244	0.9732	124.2644	0.9907	52.0342	7.5977
		9	705.1111	976.5554	72.2039	0.9964	50.1504	1.3138	146.0613	14.9568
AgI	0-100	7	88.8571	145.0000	61.2808	0.8659	6.1535	1.5626	31.9411	22.0283
	0-300	13	179.4615	255.6923	70.1865	0.9901	25.9352	1.2803	21.1147	8.2579
NaCl	0-100	6	39.5000	72.6667	54.3578	0.9622	-19.7010	2.3384	28.5614	39.3047
	0-300	10	186.4000	317.2000	58.7642	0.9749	26.3367	1.5604	39.4253	12.4292
	0-1000	13	446.4614	651.2307	68.5566	0.7247	-178.2173	1.8578	51.4966	7.9076

TABLE II.

Aerosol	Range	No. cases	\bar{X}	\bar{Y}	$\frac{\bar{X}}{\bar{Y}}$	r_{xy}	b_0	b_1	E.S.E.	$\frac{\text{E.S.E.}}{\bar{Y}}$
Nichrome I	300-1000	10	627.3	598.0	104.9	0.94760	-115.99	1.13820	76.970	12.071
	1000-3000	4	1467.5	1395.0	105.2	0.99267	90.94	0.88863	164.255	11.775
Sodium Chloride	1000-3000	8	2730.6	2417.5	113.0	0.81009	543.60	0.68626	75.628	3.128
	3000-10000	21	6803.0	6619.0	102.8	0.98695	-290.60	1.01567	432.416	6.533
	10000-30000	11	11122.2	10854.5	102.3	0.80902	2495.7	0.75154	320.25	2.950
Nichrome	0-100	2	57.5	48.0	119.8					
	100-300		243.7	231.6	105.2	0.82046	7.99	0.91752	10.667	4.607
	300-1000	3	394.3	380.0	103.8	0.88815	157.44	0.56445	22.240	5.853
Gold	0-100	3	24.67	21.33	115.7					
	100-300	13	260.6	243.2	107.1	0.93234	46.24	0.75584	10.054	4.134

VERTICAL PROFILES OF AITKEN NUCLEI CONCENTRATIONS

The aircraft ascents in New Mexico and Texas yielded AN concentrations that are comparable to Junge's data (1961) around the tropopause and surpass his concentrations by one or two orders of magnitude at an altitude of 18 km (Fig. 1). A well expressed layer of high AN concentration is often located below the tropopause (Fig. 2). This usually amounts to several hundred to one thousand nuclei per cm^3 . This layer is more conspicuous over middle geographical latitudes than over equatorial regions (Fig. 1).

Vertical AN concentration profiles above Houston and Albuquerque usually show several polluted layers in the troposphere above 6 km, which is the level where the SANDS started to operate. These layers are most probably related to the local conditions of jet aircraft traffic. This is quite obvious above Houston (November 18, 1974, February 11 and 21, 1975). Qualitative proof of the strong influence of air traffic on the AN counts was given by the crew of the instrumented WB-57F, which crossed the wake behind a jet aircraft several times. A sudden increase in AN concentrations in the wake was always found, often surpassing 20,000 AN cm^{-3} .

Aircraft ascents above Panama (February 15, 1975) were characterized by the smooth curve of AN concentrations and almost constant counts between 6 and 12 km altitude (Fig. 1). The AN concentration was approximately 300 AN cm^{-3} up to 12 km and then decreased steeply to 10 AN cm^{-3} at 19 km. This smooth decrease of AN counts is typical of the lower stratosphere above the tropics and

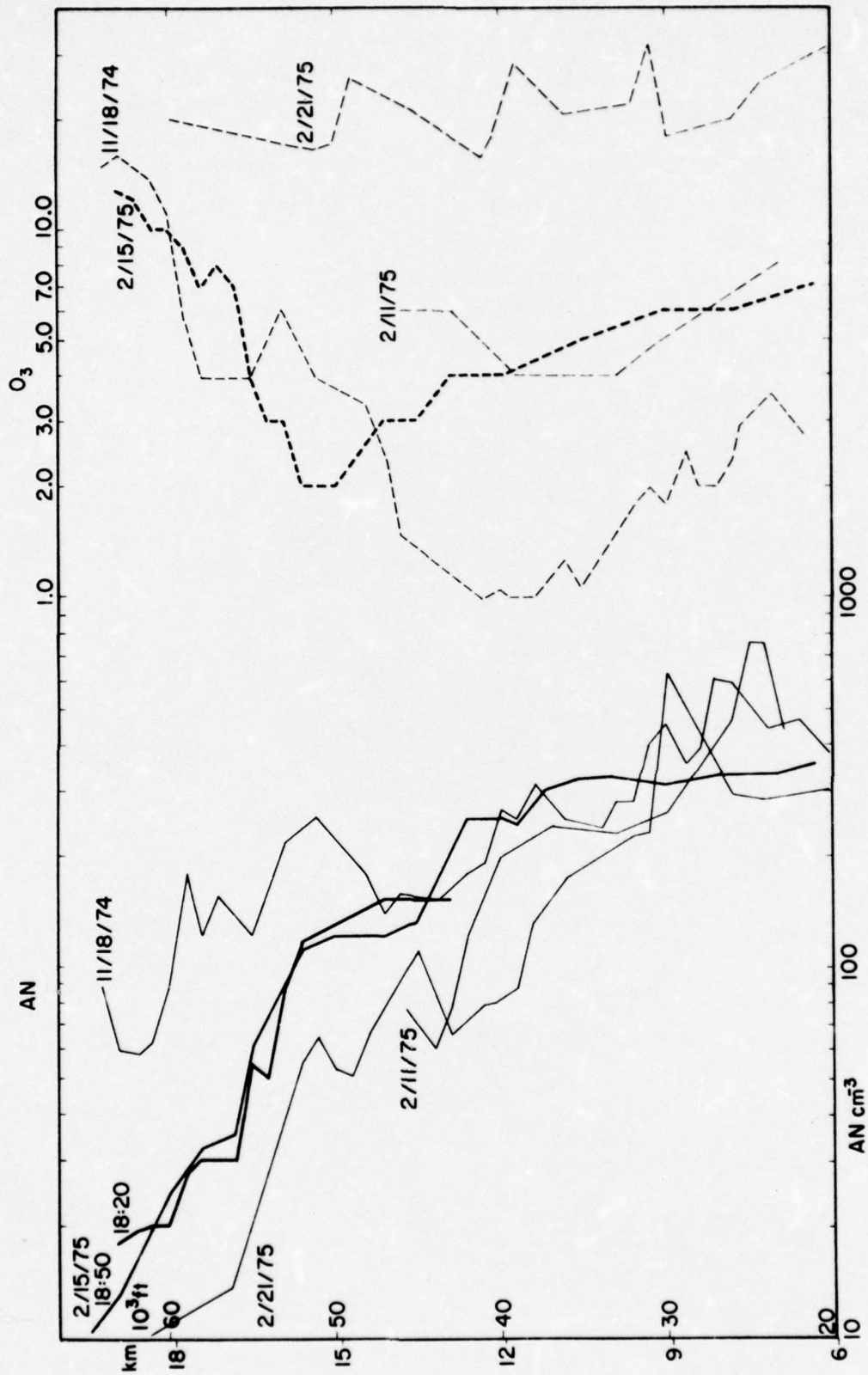


Fig. 1

AN concentration vertical profiles above Houston, TX, (November 18, 1974; February 11 and 21, 1975) and above Panama (February 15, 1975) with the corresponding ozone concentration profiles (in relative units).

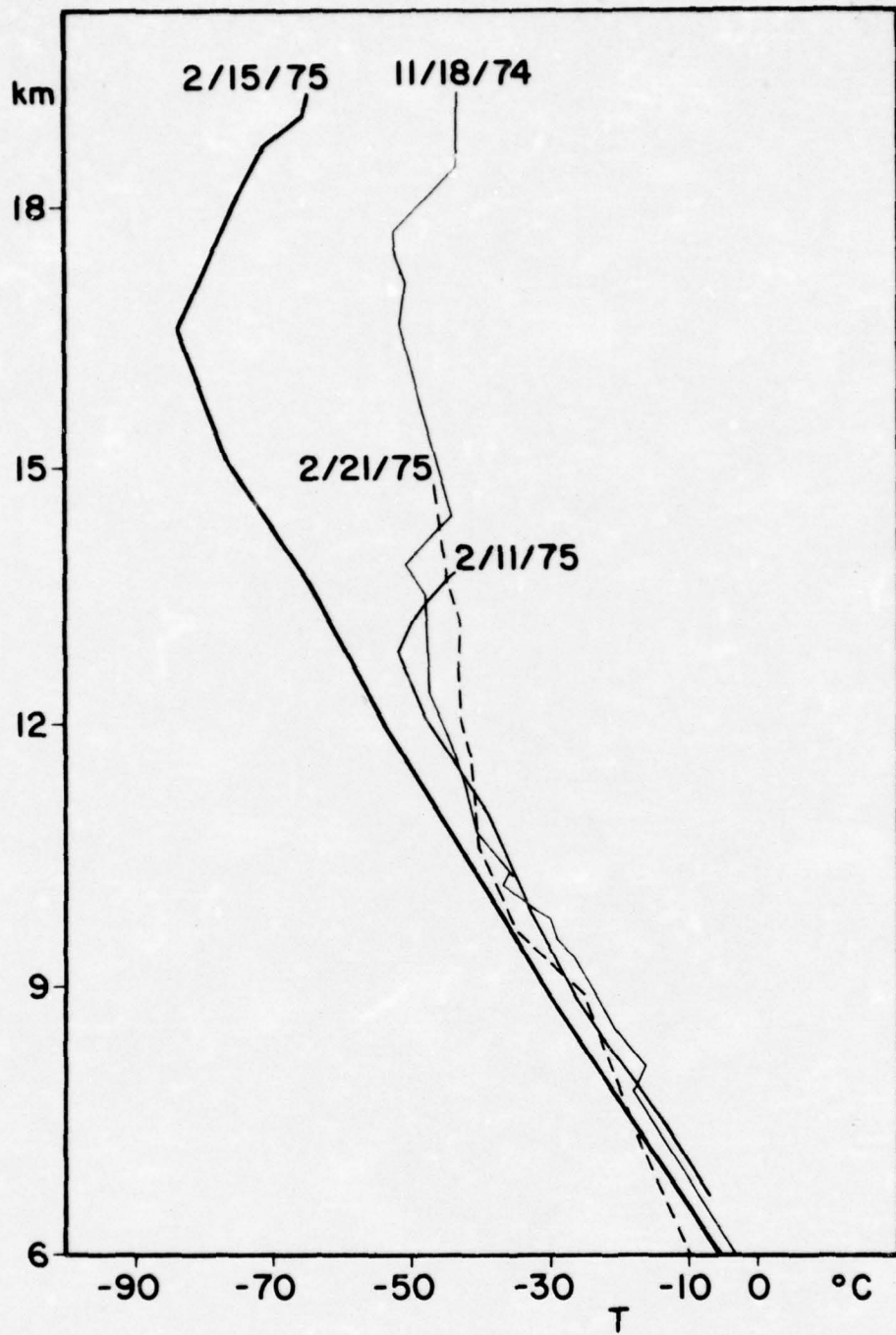


Fig. 2
 The temperature vertical profiles corresponding to the aircraft ascents in Fig. 1.

probably corresponds to the value of the vertical concentration gradient of 20 to 30 AN cm³ per 1 km of altitude. This value was reached, and actually surpassed, only during two aircraft ascents over middle latitudes at Albuquerque May 5 and 22, 1974. However, the extreme difficulty of obtaining a mean vertical gradient of AN concentrations from the curve indicates that many polluted layers are present in the stratosphere over middle geographical latitudes.

Ozone measurements were taken during several ascents with the instrument prepared by Dr. J. Rosen of the University of Wyoming. There is a strong indication that at altitudes above 17 km a negative correlation between AN counts and ozone concentration exists (Fig. 1, the ascent at Houston, November 18, 1974, or at Panama, February 15, 1975). Ozone concentration is plotted in relative units; 100 corresponds approximately to 350 nanobars of ozone pressure. No correlation between the parameters was found just above the tropopause, such as has been claimed by several authors for larger particulates, i. e., Rosen et al., 1974.

Rough estimates of the size spectrum of the AN were made by evaluating the number of charged nuclei that were deposited on the inner wall of a cylindrical condenser on which different voltages were imposed. The percentage of removed AN versus the applied voltage can be converted into the number of AN of a certain size, if one assumes the steadiness of the ratio of charged and uncharged particles at a certain altitude, the validity of the Boltzmann law for the distribution of charges on different sizes of nuclei (Rich et al., 1959), and the applicability of a steady flow

formula for the calculation of the particle deposition rate in the cylindrical condenser. The last assumption is far from real, because the SANDS operates with a pulsating flow when taking 2.5 air samples into the counter in one second. This explains the large scatter of recorded data that often makes an accurate evaluation impossible; however, a comparison of the samples taken under the same conditions and the measured size distribution curves does serve as an indication of the change in aerosol state.

In Fig. 3, four size distributions of AN are plotted from samples taken over Texas (November 15, 1974) and over the Gulf of Mexico (November 21, 1974) at different altitudes between 10.2 km and 18.2 km. The points on each curve represent the number of particles--in relative units, 1.0 means the total number of particles--that are larger than a certain size. Unlike the AN size spectra of November 15, 1974, the spectrum of nuclei measured over the Gulf of Mexico exhibits a very different character at the same level. The first curve (1) of the Nov. 21, 1974, sample was deduced from the measurement taken at the beginning of the horizontal flight (Fig. 7), where the aircraft just ascended to 18.2 km. The second curve (2) is based on the measurement around the latitude 27° N, where the dramatic change in AN counts and in the ozone concentration occur. This change is apparently responsible for the different shape of the AN size distribution curve, which reveals a large portion of very small nuclei with radii smaller than $0.02 \mu\text{m}$.

The ascent of November 18, 1974, over Houston offers another example of the importance of AN size spectrum analysis (Fig. 4). In this ascent, the layer

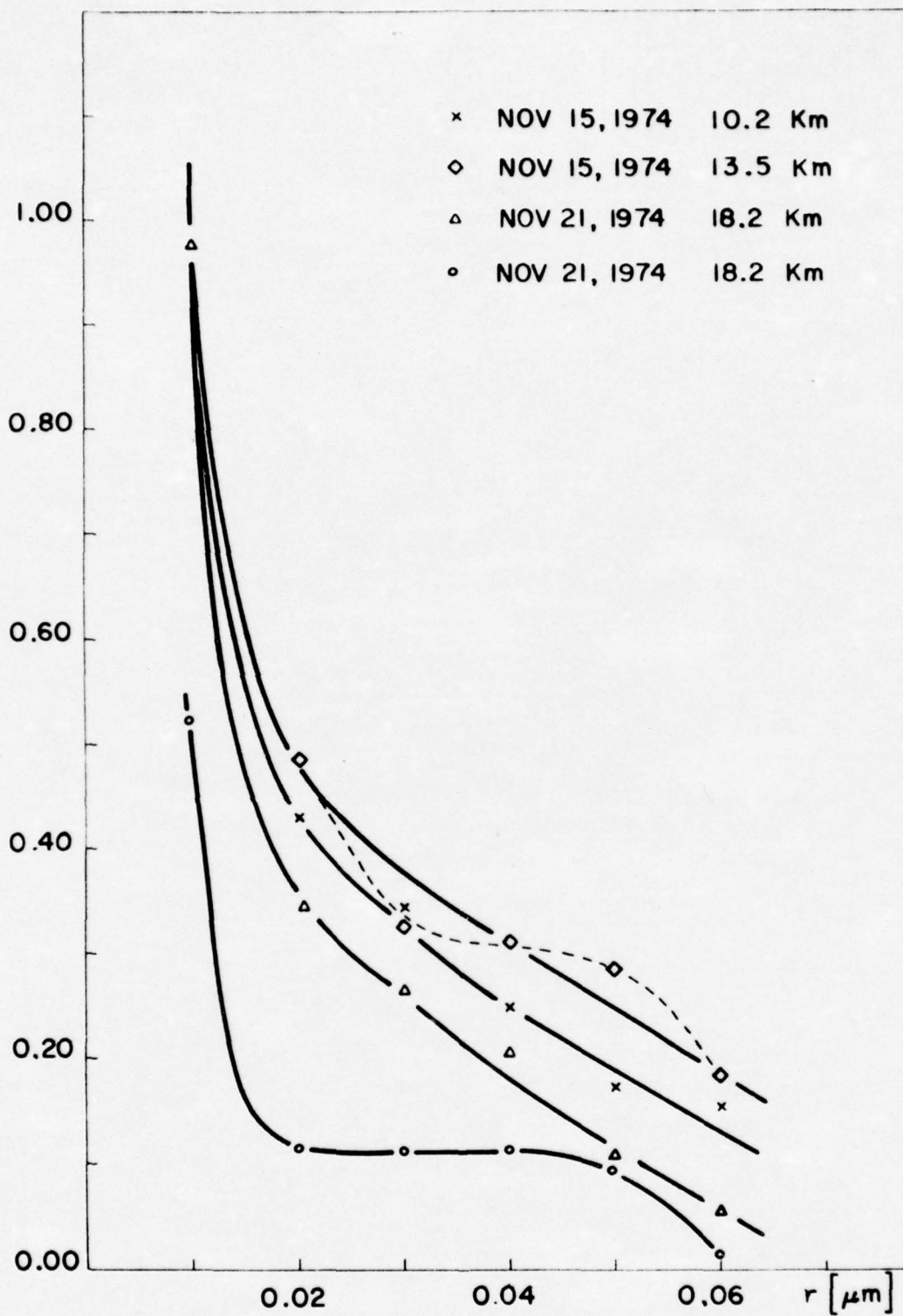


Fig. 3
 Rough estimates of the size spectrum of the AN measured over Texas and the Gulf of Mexico (November 15 and 21, 1974). (1.0 corresponds to the total number of particles)

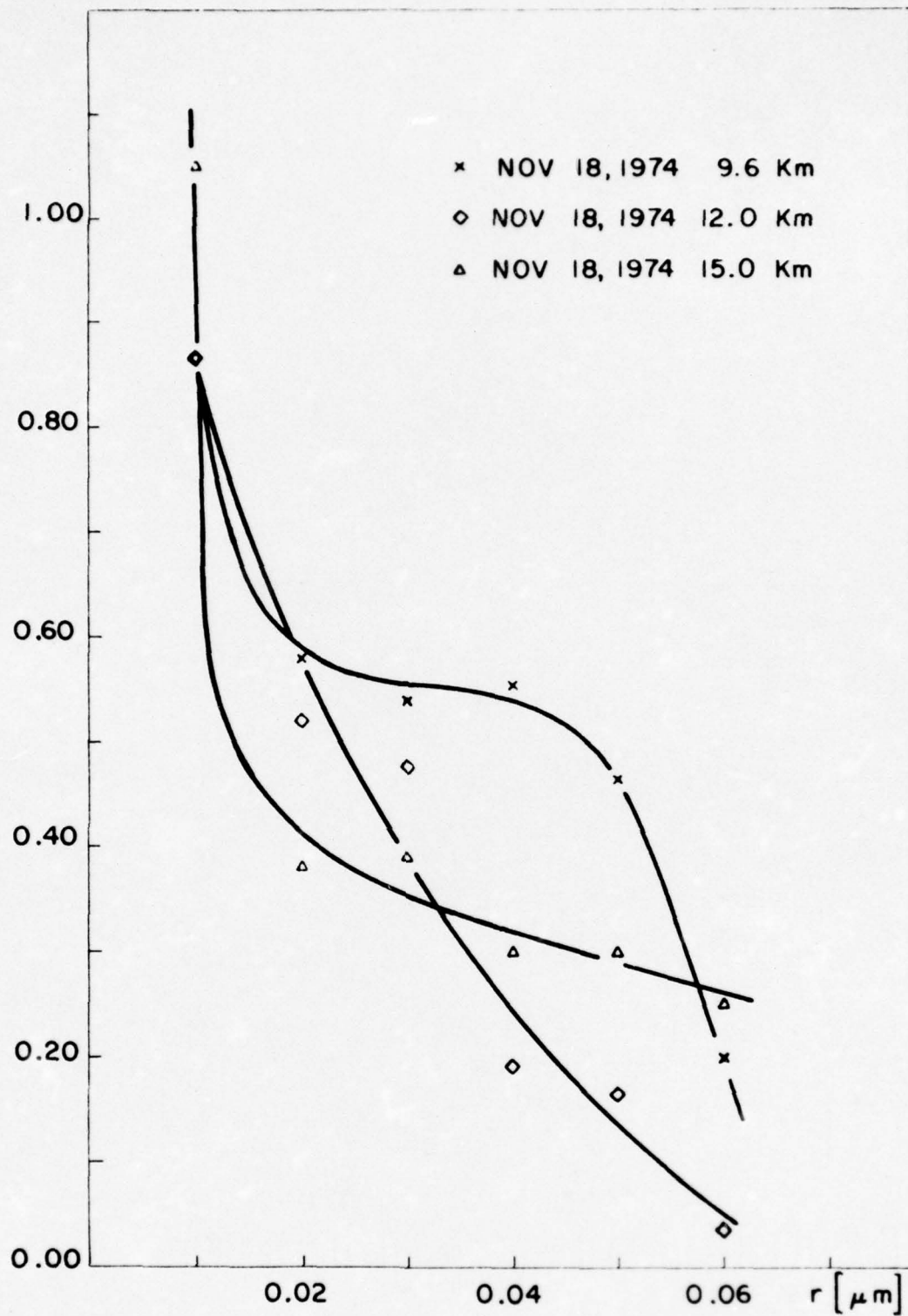


Fig. 4
 Size spectrum measurements of the AN above Houston, TX, on November 18, 1974.

type structure of the stratospheric AN concentration was accompanied by a different character of AN size distribution curves at different altitudes (Podzimek et al., 1975a). The explanation of the observed peculiarities in the AN size spectra is made difficult by the possible influence of contaminants emitted into the stratosphere by the eruption of the Guatemalen volcano, Fuego, in October 1975 (Podzimek et al., 1975a; Fujiwara et al., 1975).

In general, the evaluated size spectra of AN are not in disagreement with the hypothesis of Uchino and Hirono (1975) that the normal size distribution of AN in the stratosphere follows a power law. Exceptions are situations characterized by a particle injection after volcanic eruptions or air exchanges with higher or lower altitudes over large frontal systems, jet streams, or high mountains. This conclusion is not in disagreement with the findings of Briehl (1974).

HORIZONTAL PROFILES OF AITKEN NUCLEI CONCENTRATIONS

The main purpose of taking AN measurements at a certain level along a path covering several thousands of kilometers is to check the spatial AN concentration distribution and its time variability. In this respect, special attention is paid to the influence of high mountaineous massives, frontal systems, and the air exchange over tropical regions on the AN distribution in the lower stratosphere and higher troposphere. From this point of view, the horizontal AN concentration profiles described in detail in the two reports prepared by Podzimek *et al.*, (1975a, and 1975b) will be summarized.

The flights over the Rocky Mountains, the mountaineous part of New Mexico, and the Andes support the idea that the turbulent exchange and gravity waves over high mountains are responsible for strong variations and mean higher values of AN counts. These results have been recently supported by the measurements of AN concentrations near the tropopause by Cadle and Langer (1975). The horizontal flight of March 27, 1974, which was performed entirely in the stratosphere, documents this statement. In Figure 5, the altitude of the aircraft, the temperature at certain points of the aircraft's path (T_f), the wind vectors at the aircraft's path (W_f) and at the tropopause level (W_t), and the Aitken nuclei counts (N_{cm}^{-3}) in accordance with the longitude ($^{\circ}\lambda$) are plotted, because the aircraft flew almost exactly along the 35° latitude. At the bottom of Figure 5, the altitudes of the highest mountains situated along the flight path within a strip ± 25 km wide are very approximately plotted. Those that are north of the path are indicated with a

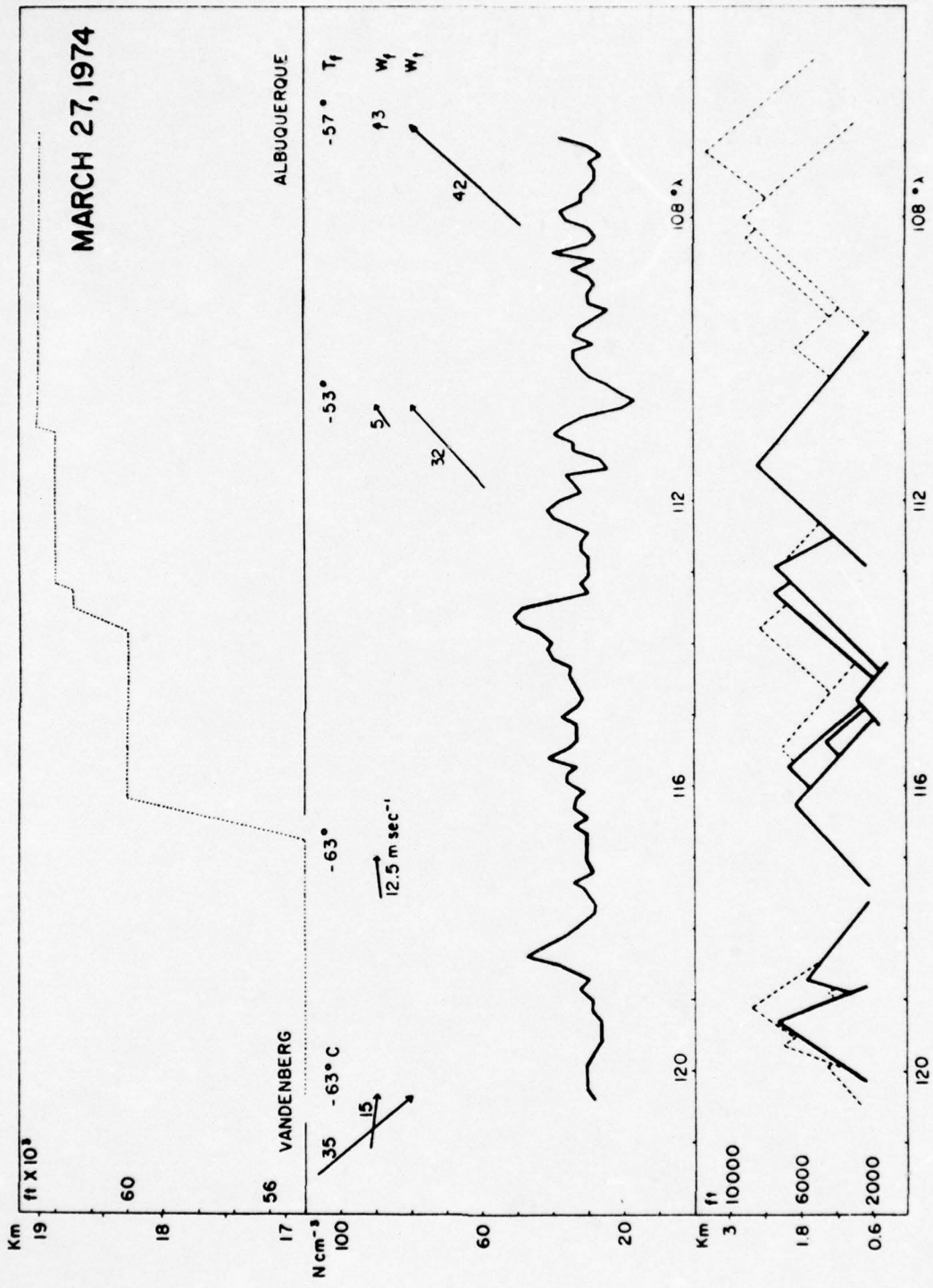


Fig. 5
 Horizontal profile of AN concentrations from the flight between Vandenberg and Albuquerque, NM, March 27, 1974.

dotted line, and those south of it with a solid line. The meteorological situation was characterized by very strong winds, which reached speeds of almost 50 m sec^{-1} above Vandenberg, Winslow, and Albuquerque at an altitude of 10 km. The first part of the flight was performed at an altitude of 16.5 km and the second around the 19.0 km level. The increase in AN counts over high Californian mountains, such as Reyes Peak (2,250 m) and Mt. Pinos (2,650 m), is conspicuous. Even at an altitude of 19 km, the AN concentration fluctuations very often surpass 50% of the mean value. This AN concentration profile contrasts strongly with those recorded on the flights over the flat plains of central Texas or over the Gulf of Mexico (Fig. 7).

The flights on April 10 and May 5, 1974, yielded a picture similar to the horizontal flight on March 27, 1974. The flight from New Mexico to the east of Texas on May 5, 1974, however, was not characteristic of the influence of high mountains, because the aircraft changed altitude several times between 14 and 18 km. On the other hand, the flight over the Andes in Ecuador and Peru on February 14, 1975, (Fig. 6) is much more supportive of the idea of the injection of tropospheric nuclei into the stratosphere above high mountaineous massives; however, the question of why the strong fluctuations of AN concentrations do not start over Ecuador, where many of the mountain peaks surpass 4.0 km, remains unanswered. In order to explain this discrepancy it is suggested that the existing secondary Intertropical Convergence Zone (Podzimek *et al.*, 1975b; Hubert *et al.*, 1969) extends from the Pacific through Peru to central Brazil. To support this hypothesis,

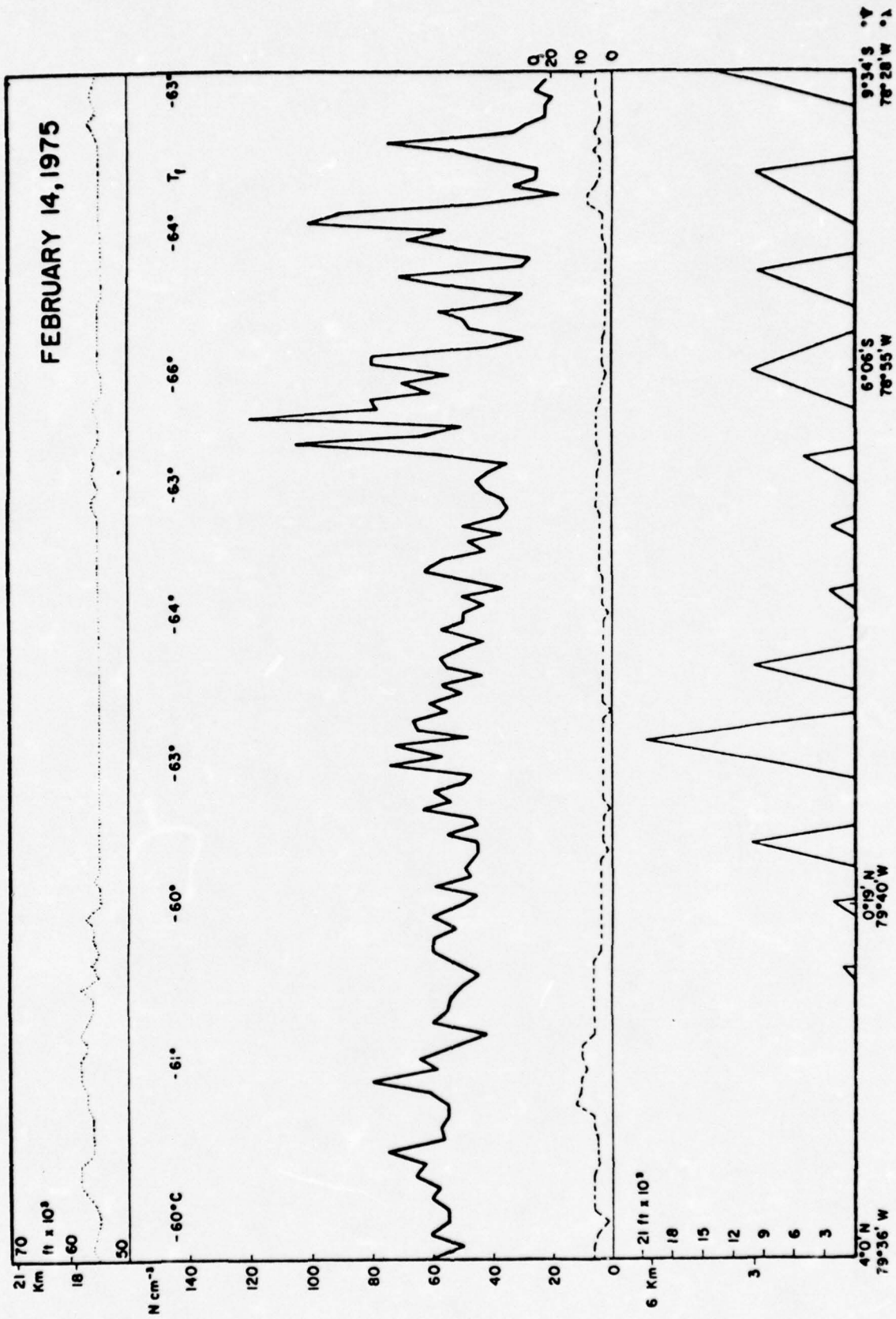


Fig. 6
Horizontal profile of AN and ozone concentrations from the flight across the equator on February 14, 1975.

it would be helpful to know the exact position of the airplane and the conditions of the meteorological parameters that existed in the stratosphere over the Andes on that particular day.

Two sunrise missions were flown on May 5, 1974, and on November 21, 1974 (Fig. 7). The first of these flights was made at night, the second at daylight. There is no indication that solar radiation strongly influenced the AN counts in the stratosphere at an altitude around 18 km. Because neither of the two flights covered the tropopause, one cannot exclude the possibility of a photochemically induced AN generation at this level, Vide, Friend et al., 1973.

The flight of November 21, 1974, (Fig. 7) was particularly interesting for two reasons: 1) the AN concentration showed at an altitude of 18 km a negative correlation with the ozone concentration, and 2) the dramatic change of AN and ozone concentration around 27°N latitude can be related to the general model of stratospheric air circulation. As a result of the high AN concentrations in general and the finding of other authors, the influence of the active volcano, Fuego, was suggested by Podzimek et al. (1975a).

Flocco and Grams (1964) found a negative correlation between ozone concentration and aerosol counts in the lower stratosphere. This finding, however, is not supported by Hofmann et al., (1972), who found no correlation between both magnitudes. Little is known about the correlation of AN with ozone. Almost all the data from the flights on November 18 and 21, 1974, February 14, 15, and 21, 1975, support the negative correlation between AN counts and the ozone concentration when the aircraft

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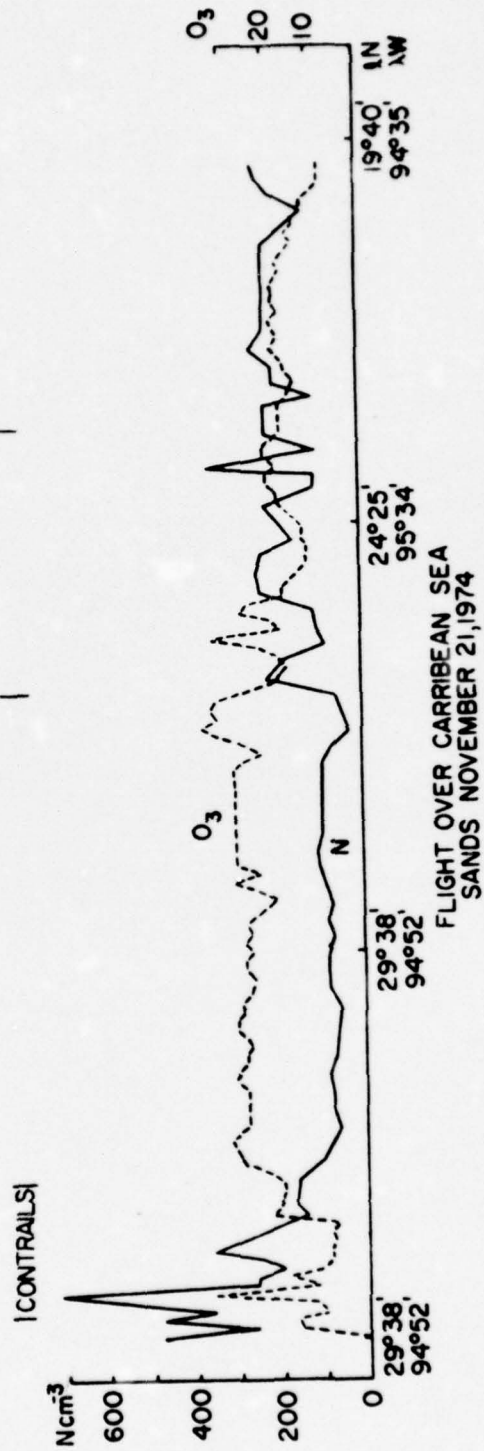


Fig. 7

Horizontal profile of AN and ozone concentrations during the flight over the Gulf of Mexico on November 21, 1974.

flew higher than 17 km. Negative correlation was weak on February 21, 1975, when the aircraft flew from the Canadian border back to Houston. At a lower altitude (below 16.5 km), there was also a negative correlation between the parameters. A separate article has dealt with this topic (Podzimek, 1976). Therefore, based on the presented material, one cannot accept the finding of Schaefer (1973) that a positive correlation between ozone and AN exists as a general rule in the subtropical and tropical atmosphere at an altitude of 39,000 ft, which is certainly below the tropopause.

Another challenging problem seems to be the relationship between AN counts and the general circulation of the atmosphere especially over subtropical and tropical regions. This subject has been investigated in the past for larger particulates by several investigators mainly in connection with the formation of Junge's layer and in explaining the origin and exchange of stratospheric aerosol (Junge and Manson, 1961; Ivlev, 1967; Pilipowskyj and Weinman, 1971; Blifford, 1971; Cadle, 1972; Friend et al., 1973; Hofmann et al., 1974; Rosen et al., 1974). AN nuclei counts are not directly included in most of the models, because there is few data from the tropical and polar atmospheres.

Several conclusions can be deduced from the AN measurements during the horizontal flights. Above the tropical tropopause, the AN concentrations remain low and rarely surpass 20 AN cm^{-3} at 18 km. Slightly higher counts are found in the lower stratosphere above the subtropical regions (February 13 and 15, 1975) and in the middle latitudes (February 21, 1975, Fig. 9) at the same altitude. The

flight of February 14, 1975, from Panama to Peru shows a different picture of AN concentration profile (Fig. 6). The AN counts fluctuate considerably around 50 AN cm^{-3} and suddenly increase in number and amplitude over the mountains in Peru. This broad band of higher AN concentrations (greater than 80 AN cm^{-3}) extends from 5°S to 9°S latitude. Because the flight was performed near the 17 km altitude, a possible explanation is that it entered the tropical troposphere in at least a part of its path. Besides this, the counts were certainly influenced by the air exchange above the peaks of the Andes.

During all flights in the tropical stratosphere (up to 19 km), the ozone concentration is low and scarcely surpasses the relative value of 10. It corresponds approximately to 35 manobars of partial pressure.

The flight on February 15, 1975, (Fig. 8) was intended to study in more detail the mechanism of AN exchange in the Intertropical Convergence Zone (ITCZ). It was performed at two different levels: above 18 km from Panama toward the equator and around 15 km on the return flight. Along the path above 18 km, the AN counts were recorded as uniformly low (between 10 to 20 AN cm^{-3}). The ozone concentration surpassed the value of 10 units on the relative scale only during the highest position of the aircraft. On the flight back, the AN concentration fluctuated strongly above 100 AN cm^{-3} , surpassing the level of 150 AN cm^{-3} above the position of ITCZ and of 200 AN cm^{-3} above the high mountains in Ecuador. The AN increase above the ITCZ is very noticeable and is easily identified when one uses the ESSA satellite's cloud pictures.

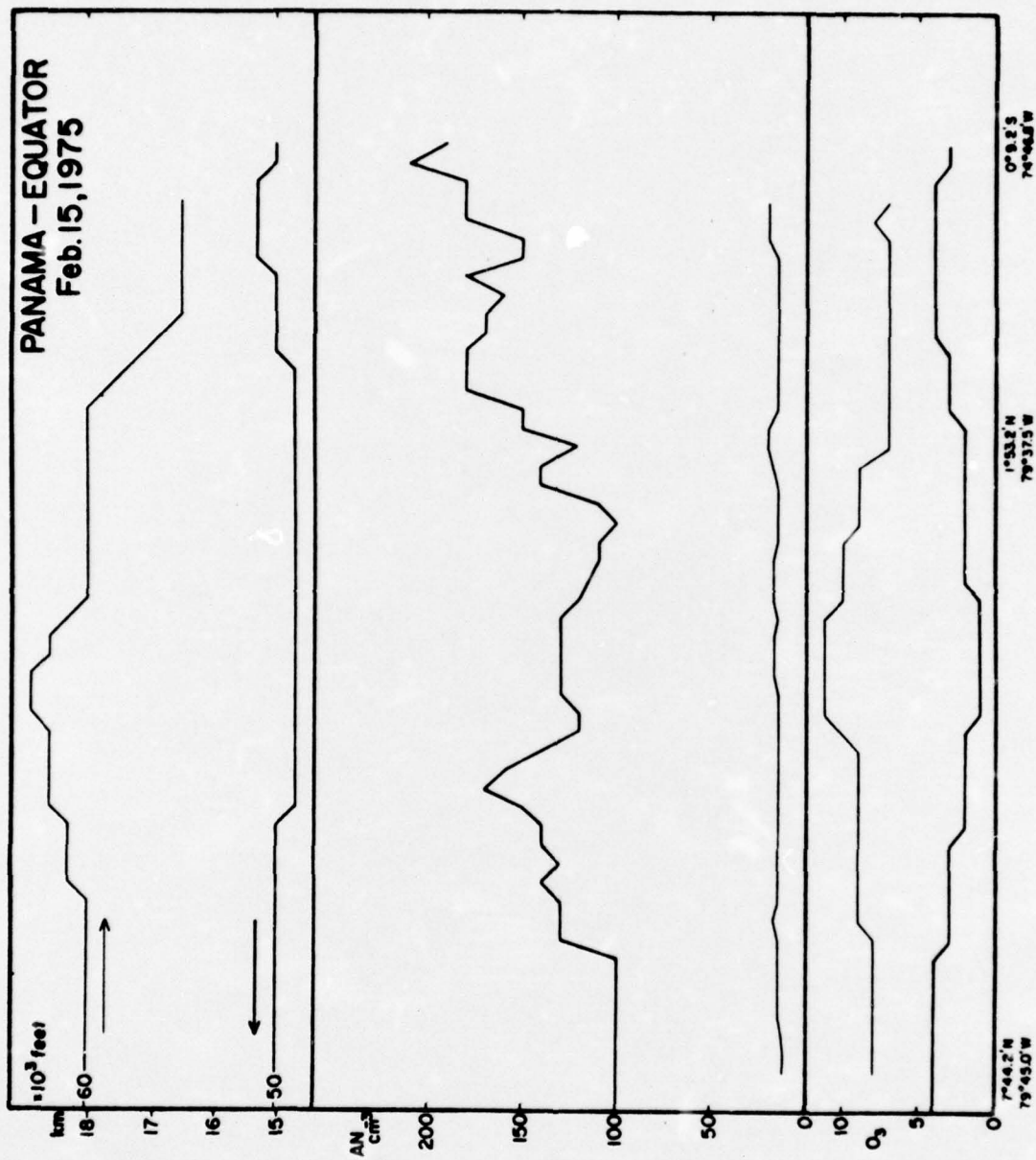


Fig. 8

Horizontal profile of AN and ozone concentrations between Panama and Equator on February 15, 1975.

The last flight of this series was performed on February 21, 1975, northward of Houston to the Canadian border and back (Fig. 9) for the purpose of checking the distribution of AN as on the flight of February 15, 1975. However, in this specific case, both flights at 18 km and 15 km altitude were performed above the tropopause. The lower flight level is characterized by higher AN counts above the Houston area (50 AN cm^{-3}) and by a gradually decreasing AN concentration toward the north (20 to 30 AN cm^{-3}). On the flight back, AN concentrations remained almost at the same value and fluctuated between 7 and 30 AN cm^{-3} . There is an indication that the low AN counts in the middle latitudes correspond to the high ozone concentration (above 70 relative units, which corresponds approximately to 245 nanobars of partial pressure). Unfortunately, a part of this record suffers from the malfunction of the instruments which explains the gap in data between 39°N and 44°N latitude. In general, the ozone concentration is higher at 18 km on the north than on the south of the aircraft path.

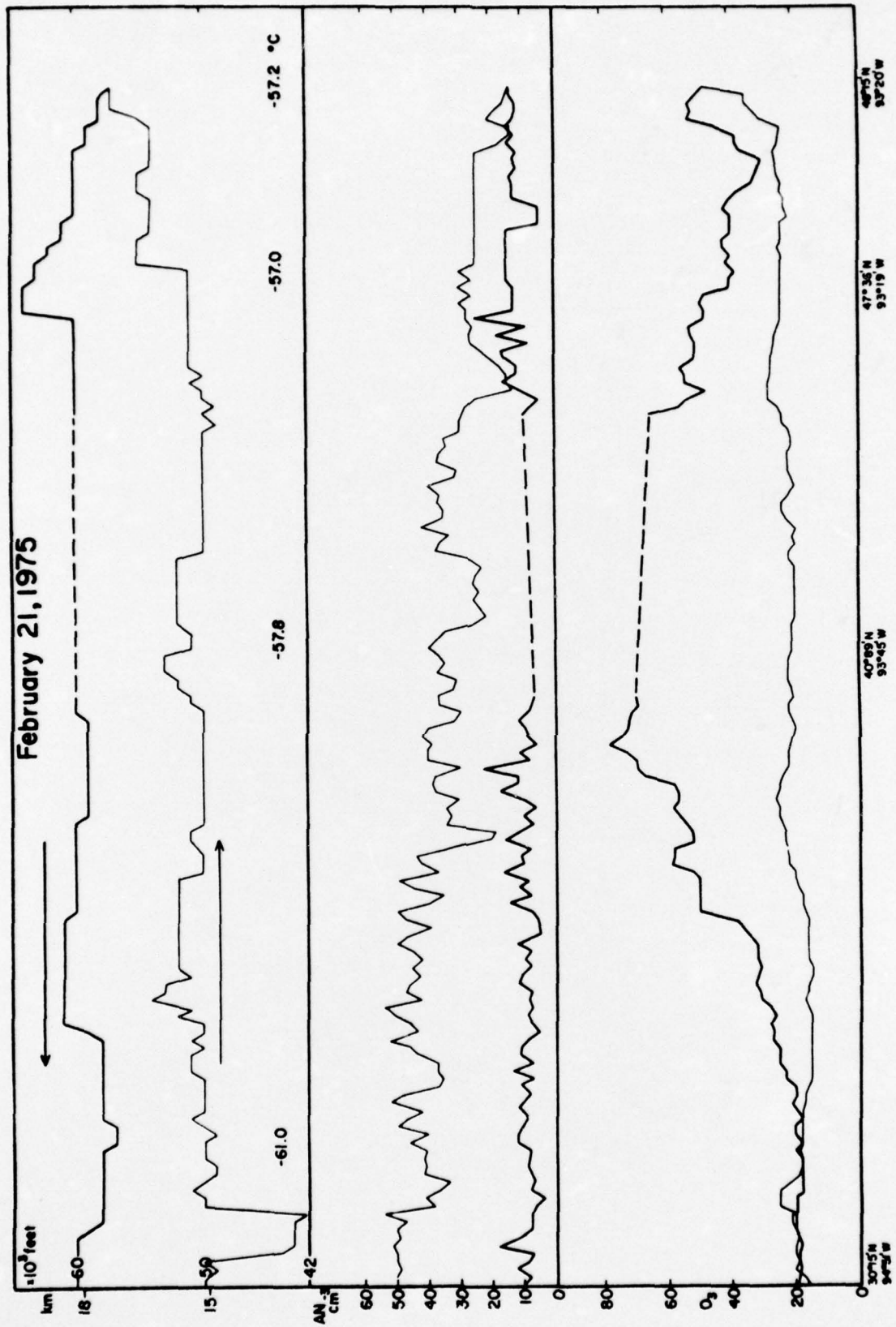


Fig. 9
 Horizontal profile of AN and ozone concentrations during the flight from Houston to the Canadian border and back on February 21, 1975.

CONCLUSIONS

The AN concentrations measured by the SANDS in the lower stratosphere over the U. S. A. and Central and South America between March 1974 and February 1975 can be characterized in the following way:

1) The vertical profiles are very similar to those found by Junge and his fellow workers (1961) with the exception that at altitudes above 17 km the data presented in this report are of one or two orders of magnitude higher. Also, compared with AN measurements made recently by Rosen et al. (1974) in the U. S. A. and by Kaselau et al. (1974) in Germany with balloon-borne AN counters, the data presented in this work are slightly higher for altitudes above 15 km. At 18 km, AN counts were recorded in the tropical and middle latitude stratosphere around 20 AN cm^{-3} ; however, several times they reached almost 70 AN cm^{-3} , mainly over the middle latitudes (March 5, 1974; May 22, 1974; November 18, 1974). Usually one can find high concentrations of AN just below the tropopause, where they surpass 500 AN cm^{-3} , and low concentrations above it. AN concentrations greater than 1000 AN cm^{-3} were found confined to relatively thin layers in areas with heavy air traffic in the upper troposphere.

2) The tropopause represents an efficient barrier to the penetration of tropospheric AN into the stratosphere. This is documented mainly by the horizontal AN concentration profiles above and below the tropical and subtropical stratosphere. There is an indication of a higher concentration of AN below the tropical tropopause (more than 100 AN cm^{-3}) and of moderate transport of AN through it by means

of the Hadley cell circulation.

3) High mountain masses act as agents in stimulating air exchange up to levels higher than 18 km, such as over the Rocky Mountains and the Andes.

4) No noticeable difference was found between the stratospheric AN counts taken before and after sunrise.

5) Most of the AN concentration measurements above 16.5 km show a negative correlation with ozone counts. Only the flight on February 14, 1975, yielded a slightly positive correlation between both parameters. This particular flight was characterized by very low ozone counts and was performed at an altitude around 17 km, which was apparently at the tropopause level. At altitudes lower than 16.5 km, there is no correlation between AN and ozone concentrations that can be characterized by a widely prevailing sign. High ozone concentrations on February 21, 1975, measured over middle latitudes at an altitude of 18 km are not paralleled by low AN counts that would correspond to a supposed perfect negative correlation.

This report is of necessity a presentation of rough data accompanied by observations on some interesting peculiarities of the stratospheric AN. It does not answer many questions, which must await answer until more material is gathered and analyzed; however, it can serve, together with other reports on AN and aerosol measurements in the atmosphere, for a more complex interpretation, which could contribute to the better understanding and checking of the feasibility of many existing theoretical models. An attempt has been made in a separate

article to contribute to this interpretative work (Podzimek, 1976) and to compare AN and ozone concentrations in the stratosphere.

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RECOMMENDATIONS

Several recommendations can be made on the basis of the past experience with the SANDS during the AN measurements in the laboratory and in the stratosphere:

1) The SANDS program should be continued in order to answer questions about the operation of the pressurization unit, humidification system, and the device for measuring the size spectrum of AN, mainly during a series of flights over Alaska. Also, SANDS operation should be rechecked in the laboratory, because some measurements indicated that the supersaturation in the SANDS might be lower than that claimed by the designer. The nuclei size spectrum measurement in the SANDS requires several improvements, according to the opinion of the author. First of all, the theoretical assumptions should be rechecked and an estimated error in the measurements should be calculated. Secondly, the current way of measuring and recording the percentage of deposited charged nuclei is not very accurate.

2) There still is a need to make a direct comparison of the Nolan-Pollak counter with the University of Missouri AANC. Several indirect comparisons reveal that the Nolan-Pollak counter when used as a standard by all producers in the U.S.A. often measures 20 to 30 percent less nuclei than the AANC at the same supersaturation.

3) The measurements of AN in the tropopause should be continued to establish a complete picture of the exchange of AN and pollutants over the ridges of high

mountains, around jet-streams, large thunderstorms, and well developed frontal systems. Special attention should be paid to the polar and tropical tropopause.

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LIST OF SYMBOLS

b_0, b_1	constants in the regression curve deduced from the pairs of AN concentrations measured by the AANC (X_i) and by the SANDS (Y_i) and from the mean AN concentrations within a certain concentration range (\bar{X}, \bar{Y})
E. S. E.	estimated standard error
\bar{r}	mean radius of an aerosol particle
r_{xy}	correlation coefficient of the AN concentrations
T_f	temperature ($^{\circ}\text{C}$) at the flight level
W_f	wind vector at the flight level
W_t	wind vector at the tropopause level
X	Aitken nuclei concentration measured by the AANC
Y	Aitken nuclei concentration measured by the SANDS
Y	predicted concentration of AN which would be measured by the SANDS
α	monodispersity factor (geometrical) of the size distribution of aerosols
σ	standard deviation

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