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AIR CONCENTRATIONS OF KRYPTON-85 IN THE MIDWEST UNITED STATES DURING JANUARY–MAY 1974

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Abstract—Twice-daily air samples were collected at thirteen sites in the mid-western U.S. from 24 January to 20 May, 1974 in a study of the long-range transport and dilution of the ^{85}Kr plume emitted at the National Reactor Testing Station (NRTS) in Idaho. The NRTS plume (about 1000 Ci day^{-1}) proved to have very little effect on the ^{85}Kr background concentrations in the midwest (1500–2500 km from the source). The mean concentration during the month prior to the start of emissions was 14.0 compared to 13.8 pCi/SCM^{-1} during the most active emission period. Almost all of the 2500 samples collected had ^{85}Kr concentrations within 5% of the ambient background. Meteorological analysis indicated marked changes in ^{85}Kr concentrations from one air mass to another with relatively little fluctuation within an air mass. Surface dewpoint temperature, a quasi-conservative air mass property, was found to be well correlated with ^{85}Kr fluctuations. Occasional high concentrations of ^{85}Kr at Indianapolis and Detroit were found to be caused by plumes from the Savannah River Plant in South Carolina. Two episodes of anomalously high concentrations at Minneapolis are attributed to unknown local sources.

1. INTRODUCTION

The radioisotope ^{85}Kr of the noble gas krypton (half-life of 10.76 y) is produced by nuclear fission, mainly as a result of the operation of nuclear reactors and, to a lesser extent, in nuclear explosions. Other sources of ^{85}Kr are negligible (NCRP, 1975). Almost all ^{85}Kr produced in reactors is released to the atmosphere, not at the reactor site, but in the course of dissolution of the spent fuel at nuclear fuel reprocessing plants. The main source region of ^{85}Kr is the temperature latitudes of the Northern Hemisphere.

The concentration of ^{85}Kr in the atmosphere has been increasing with the growth of the nuclear industry during the past two decades (Pannetier, 1968; Schröder and Roether, 1974; United Nations, 1972). Since ^{85}Kr is an inert gas and has relatively long half-life, projections of its accumulation in the atmosphere have been made (Coleman and Liberace, 1962; Diethorn and Stockho, 1972). Others have used regional and global scale dispersion models for estimating population exposure (Knox and Peterson, 1972; Machta *et al.*, 1974). Recently Schröeder and Roethér (1974) and Telegadas and Ferber (1975) estimated the global atmospheric inventory of ^{85}Kr at the end of 1973 to be about 53 MCi.

This paper discusses the ^{85}Kr concentrations measured in surface air over the midwestern U.S. from 24 January to 20 May, 1974. Over 2500 samples were analyzed in an experimental program intended to study long range (1500–2500 km) atmospheric transport and dilution using, as a source of opportunity, the ^{85}Kr plume emitted from the fuel reprocessing plant at the Idaho National Engineering Laboratory, formerly the National Reactor Testing Station (NRTS).

The first month of sampling provided background data as there were no emissions from NRTS. From 28 February to 8 May an average of about 1000 Ci day^{-1} of ^{85}Kr were emitted from NRTS. There were no emissions after 8 May.

Calculations using the NOAA-ARL transport and diffusion model (Heffter *et al.*, 1975) indicated that the NRTS plume would produce peak concentrations about 25% above background at the sampling sites. Actually, very few samples showed ^{85}Kr concentrations more than 5% above the ambient background. The NRTS plume had no discernible effect on mean ^{85}Kr concentrations in the midwest during this period. The mean ^{85}Kr concentration for samples collected from 25 January to 27 February (a non-emission period) was 14.0 pCi/SCM^{-1} with a standard deviation of 0.5 pCi/SCM. During the most active emission period, 28 February–24 April the mean was 13.8 ± 0.3 pCi/Scm^{-1}.

2. SAMPLING PROGRAM

Cryogenic air samplers were installed at 13 of NOAA's National Weather Service (NWS) stations in the midwest (Fig. 1) to collect two 10-h samples each day. The samplers, designed by AIRCO, Inc. (1972, 1973), concentrate the krypton from an abundance of about one part per million in the ambient air to about one part per 100 in a 900 cm^3 sample cylinder. This is done by continuous liquefaction of the incoming air, using liquid nitrogen (about 25 l.

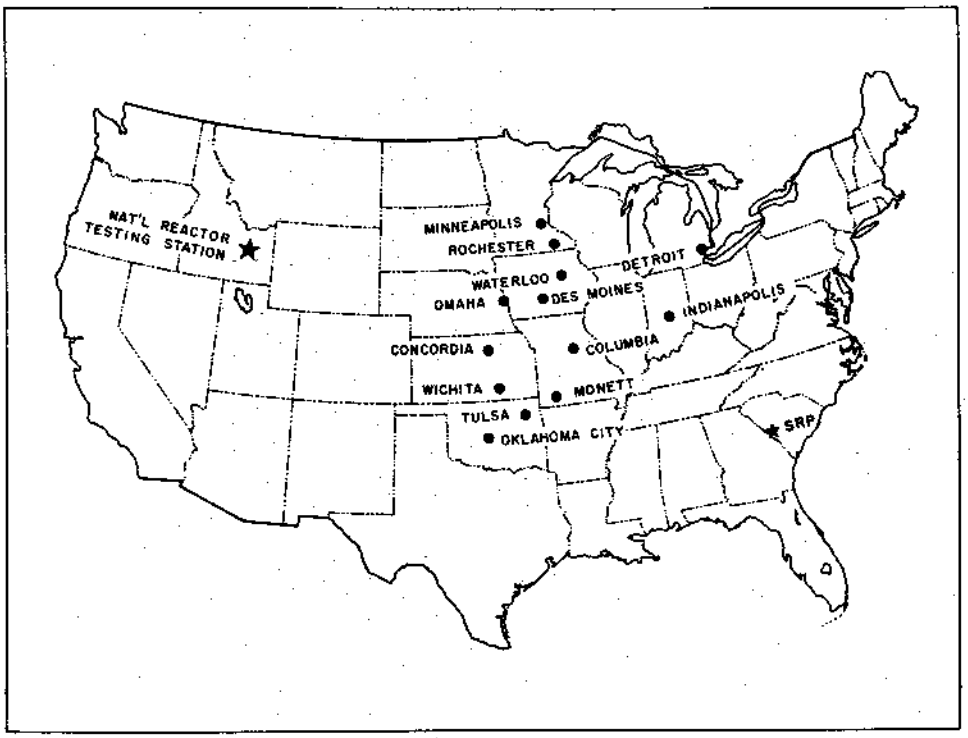

Fig. 1. Location of cryogenic air sampling stations, the National Reactor Testing Station (NRTS), and the Savannah River Plant (SRP).

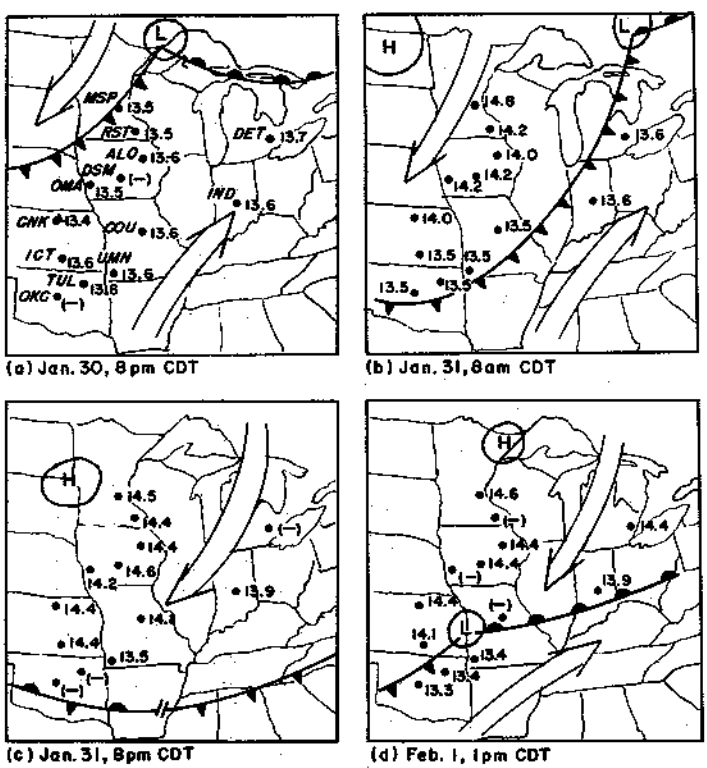

Fig. 2. Krypton–85 concentrations (pCi/Scm^{-1}) in relation to a frontal passage. (–) indicates missing ^{85}Kr data. Broad arrows show the surface air flow.

per day) as the cold source, and allowing the more volatile atmospheric components to boil off.

Each unit was programmed to start sampling at 09:00 and 21:00 h Local Daylight Time (LDT) each day. A compressor pumped air through the sampler at a rate of about 1 $m^3 h^{-1}$ and about 5–10 cm^3 of krypton were collected in each sample. The steel sample cylinders were shipped to AIRCO where the krypton was isolated in a highly purified state (at least 99.98% pure) by a gas chromatographic separation technique. The amount of ^{85}Kr contained in the sample was then determined by measuring its radioactivity with special Geiger counters over a 15-h period. Each sample was counted twice (in two different counters) to increase precision (statistical counting error was generally less than 1%).

Eleven stations were located about 1500 km from the source at NRTS. Two additional samplers were set up at Indianapolis and Detroit to attempt to detect traces of the plume about 2500 km from the source. The station identifiers are listed below:

Station	Call letters	Lat. (°N)
Indianapolis, IN	IND	39.7
Detroit, MI	DET	42.2
Minneapolis, MI	MSP	44.8
Rochester, MI	RST	43.9
Waterloo, I	ALO	42.6
Des Moines, I	DSM	41.5
Omaha, NB	OMA	41.2
Concordia, KS	CNK	39.6
Columbia, MO	COU	39.0
Wichita, KS	ICT	37.6
Monett, MO	UMN	37.0
Tulsa, OK	TUL	36.1
Oklahoma City, OK	OKC	35.3

3. VARIATION OF ^{85}KR WITH AIR MASS

In general, the background concentration of ^{85}Kr has been increasing from year to year. There is a seasonal variation, generally decreasing from winter to summer, and a latitudinal gradient with higher values to the north. Much of the fluctuation of ^{85}Kr concentration over the sampling network can be explained by the influx of air masses from the north or south. A good example of this can be seen (Fig. 2) in the concentrations from 30 January–1 February 1974. All ^{85}Kr concentrations are in picocuries per standard cubic meter of air computed at 76 cm Hg and 15°C.

The surface weather map (Fig. 2a) for 19:00 h Central Daylight Time (CDT) on the 30th, the end of the daytime sampling period, shows a low pressure center at the Canadian border with a cold front extending southwestward through Minnesota and Nebraska, and a warm front extending eastward across the northern Great Lakes. The cold front marks the leading edge of an intensely cold air mass moving southward out of northwestern Canada. At this time all sampling stations are in a relatively warm air mass with southerly flow. The daytime samples (09:00–19:00 h CDT) on the 30th (Fig. 2a) all show relatively low concentrations ranging from 13.4 to 13.8 pCi/Scm.

Figure 2(b) shows the weather map at 07:00 h CDT on 31 January the end of the nighttime sampling period. The cold air has moved rapidly southward with the front as far as Oklahoma. Indianapolis and Detroit are still in the southerly flow to the east of the front and concentrations are correspondingly low (13.6 pCi/Scm^{-1}). The other stations are all in the northerly flow by the end of this sampling period and the stations from Minneapolis to Concordia show significant increases in ^{85}Kr (values ranging from 14.0–14.8 pCi/Scm^{-1}). However, the stations from Columbia southward still have low values; probably because the southernmost portion of the cold air mass has undergone modification through mixing with the warm air (and low krypton) to the south as evidenced by the relatively high temperatures (not shown) in the region just behind the front. The temperature at Concordia was 25°F while Wichita had 34°F. Another factor is that the front passed the southern stations very late in the sampling period.

Figure 2(c) shows the weather map at 19:00 h CDT on the 31st; the end of the daytime sampling period. All stations, including Indianapolis and Detroit are now in the cold air mass. Columbia (14.1 pCi/Scm^{-1}) and Wichita (14.4 pCi/Scm^{-1}) now show concentrations characteristic of this air mass but Monett is still lower (13.5 pCi/Scm^{-1}) than we might expect.

For most of the next sampling period (not shown) the stations remained under the influence of the Canadian air mass as it moved eastward and all stations showed relatively high values (13.9–14.8 pCi/Scm^{-1}). Toward the end of the period, southerly flow in the warm sector of a low pressure region replaced the Canadian air over the stations in Oklahoma and Missouri. This is reflected in the return to low concentrations (13.3–13.4 pCi/Scm^{-1}) in the 1 February daytime sampling period at OKC, TUL and UMN in Fig. 2(d) which shows the weather map at 14:00 h CDT, in the middle of this sampling period.

4. ^{85}KR AND DEW-POINT TEMPERATURE

Since it appears that different air masses tend to have characteristic ^{85}Kr concentrations, we would expect ^{85}Kr to be correlated with other quasiconservative air mass properties. One such property, for which data were readily available at the sampling stations, is surface dew-point temperature. Dew point is a conservative property of a parcel of air for radiative heating or cooling to the point of saturation. It is not conservative when evaporation or condensation occur nor for turbulent mixing with air of different water vapor content.

The average dew point temperature for each sampling period was determined from the 3-hourly observations taken at all locations except Monett, MO,

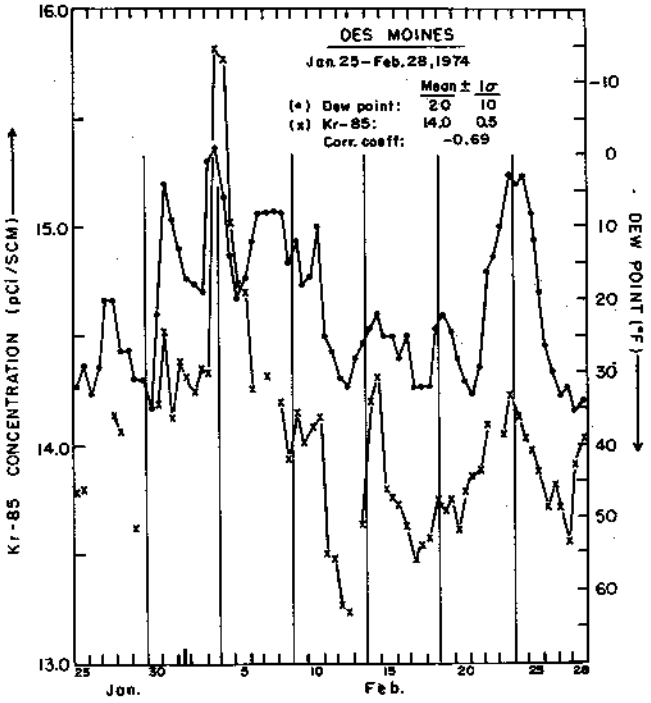

Fig. 3. Twice-daily ^{85}Kr concentrations and dew point temperatures for Des Moines Iowa (note inverted dew point scale).

where data from Springfield, MO, approx. 60 km NE of Monett, were used.

A comparison of ^{85}Kr concentrations with dew point temperatures for 25 January–28 February 1974 at Des Moines, Iowa is shown in Fig. 3. During this period there were no known emissions from NRTS. An obvious negative correlation exists with relatively high concentrations of ^{85}Kr associated with low average dew point temperatures. The changes from one sampling period to the next also tend to follow this pattern rather well; i.e. an increase in ^{85}Kr is usually associated with a drop in dew point.

Low dew points usually are indicative of more northerly, dry, colder air containing higher background ^{85}Kr concentrations than the more southerly, warm, moist air. The data in Fig. 3 give a correlation coefficient of -0.69 between ^{85}Kr and dew point.

5. SPACE AND TIME DISTRIBUTIONS OF ^{85}KR AND DEW POINT

The space and time distributions of ^{85}Kr and dew point were analyzed for the entire sampling period. Results for the period from 24 January–8 February are illustrated in Fig. 4. The horizontal scale is time beginning with the nighttime observation on 24 January followed by the day and night observations on 25 January and so on. The sampling sites are arranged from north (MSP) to south (OKC) along the vertical axis except that Indianapolis and Detroit are at the top and separated from the others by a horizontal line. The top portion of the figure shows the ^{85}Kr concentrations (pCi/Scm^{-1} *minus* 10 pCi/Scm^{-1}, to save space) while the lower portion shows the dew point temperatures.

The ^{85}Kr concentrations of 14 pCi Scm^{-1} (plotted as 4.0) or greater are shaded. Similarly a dew point temperature of 20°F or less is shaded. Note the correspondence between shaded areas for dew point and ^{85}Kr concentration. There were no known emissions from NRTS during this period. Figure 4 shows three periods, denoted by the capital letters, A, B, and C, where a deep southward penetration of high ^{85}Kr concentrations is accompanied by a surge of low dew point temperatures southward associated with intrusions of cold polar air across the sampling network. Northward extensions of relatively low concentrations, denoted by lower-case letters a, b, and c, are accompanied by high dew points.

During the entire sampling period (24 January–20 May, 1974) there were 15 cases where relatively high concentrations of ^{85}Kr associated with low dew points extended southward along the sampling network. There were 20 cases where relatively low ^{85}Kr concentrations accompanied by high dew points extended northward. Superimposed on these patterns, some anomalously high concentrations were observed both during non-active and active emission periods at NRTS.

6. ANOMALOUSLY HIGH ^{85}KR CONCENTRATIONS

Two episodes of very high ^{85}Kr concentrations occurred at Minneapolis about a month apart (18–19 February and 15–16 March). Concentrations in these

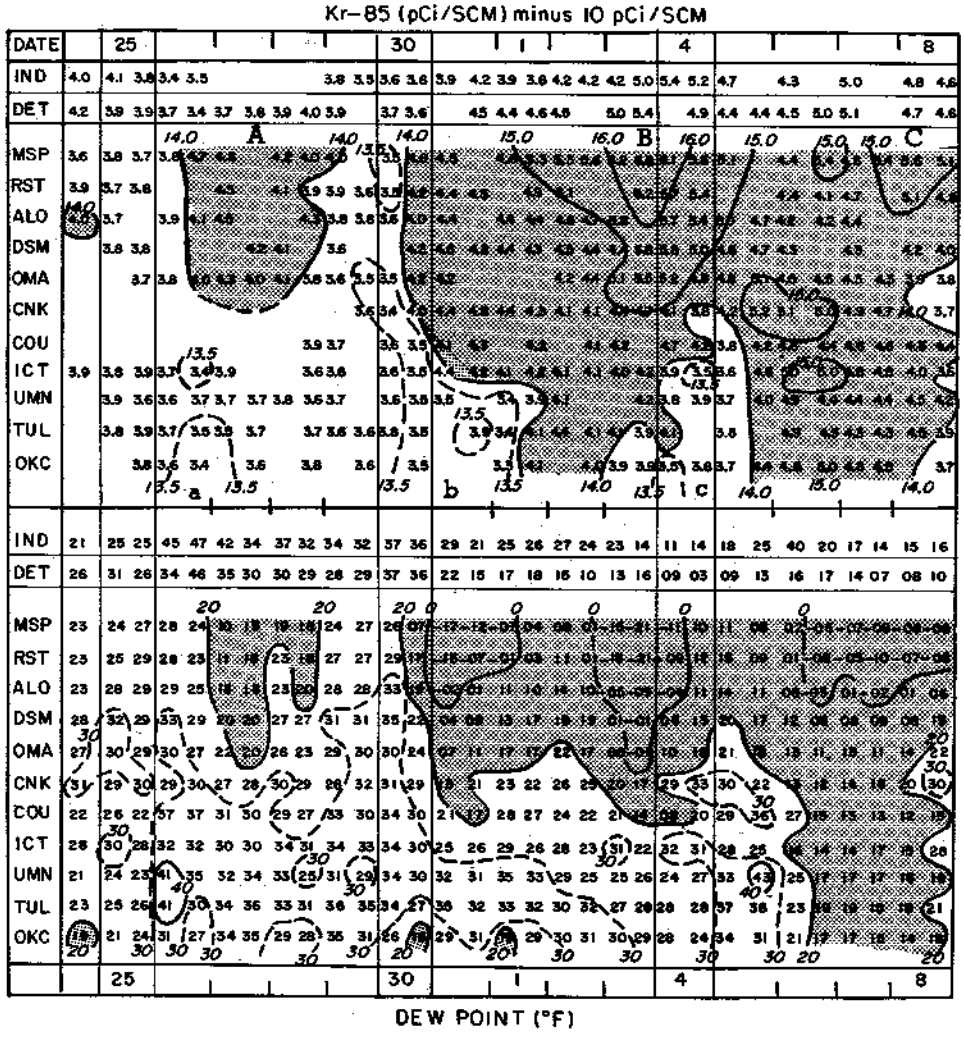

Fig. 4. Distribution of ^{85}Kr concentration and dew point temperature for all sampling stations from 24 January to 8 February, 1974.

samples are given in Table 1. In the first instance, prior to the start of pluming at NRTS, the background concentration in the Minneapolis to Waterloo region was about 14 pCi/Scm^{-1}. Three consecutive samples with ^{85}Kr concentrations ranging from 15.5–20.0 pCi/Scm^{-1} were observed. Although the high concentrations occur after a cold front passage, they are much too high to be accounted for by the air mass change. The air moved in from the Pacific and covered the entire sampling arc. All stations show a drop in dew point and a rise in ^{85}Kr during the period but concentrations at Rochester and Waterloo suggest that the air mass background at Minneapolis should be no higher than about 14.5 pCi/Scm^{-1}. If the high concentrations had been due to arrival of a plume from a distant source, we would expect that diffusion would have spread the plume sufficiently that Rochester would have had similar high concentrations. The most likely cause would appear to be a local source of ^{85}Kr.

Table 1. Episodes of High ^{85}Kr at Minneapolis

Date	Sample starting time	^{85}Kr (pCi/Scm^{-1})
18 Feb.	09:00 h	15.5
18	21:00 h	16.8
19	09:00 h	20.0
15 Mar.	21:00 h	26.7
16	09:00 h	16.3
16	21:00 h	17.4

In the second episode (15–16 March) three consecutive samples with ^{85}Kr concentrations ranging from 16.3–26.7 pCi/Scm^{-1} were measured while the background concentration from Rochester south to Concordia was no more than 14.5 pCi/Scm^{-1}. The high values occurred after the arrival of a Canadian air mass from the north but, once again, the fact that very high concentrations are localized at a single station suggests a local rather than a distant source. The NRTS source was in operation at the time but model calculations indicate the plume did not pass very close to Minneapolis. Even if the plume has

passed directly over Minneapolis, very little of it would be likely to mix into the cold air mass at ground level.

The similarity in air concentration and duration suggests a similar cause for both episodes; possibly a local ground-level source, but this has not been verified.

Five episodes of anomalously high ^{85}Kr concentrations occurred at Indianapolis and Detroit. Meteorological analysis suggests that the Savannah River nuclear fuel reprocessing plant in South Carolina about 900 km to the SSE was the probable source. Estimates of ^{85}Kr emissions were obtained from the Savannah River Plant (SRP) and the NOAA-ARL model was used to calculate plume trajectories and expected sample concentrations from this source. In every case, the high concentrations at these two stations could be attributed to SRP plumes. These cases are of interest from the standpoint of long-range transport and diffusion model verification and are discussed in a separate paper, now in preparation.

7. CORRELATION BETWEEN ^{85}KR AND DEW POINT

The correlation between ^{85}Kr concentrations and dew point temperatures for the total period of sampling at each sampling station is given in Table 2. The mean dew point temperature and ^{85}Kr concentration and their standard deviations are also shown.

Indianapolis, Detroit and Minneapolis show relatively low correlations while all the other stations indicate a negative correlation ranging from about −0.5 to −0.7. Seven samples at IND and four at DET were attributed to plumes from SRP. At MSP there were six samples with very high ^{85}Kr concentrations, possibly due to a local source. When these very high non-background samples are excluded from the statistics, the correlation coefficients fall into line with the others as indicated by the recomputed statistics at the bottom of Table 2.

Table 2. The mean dew point temperature (T_d) and ^{85}Kr concentration with their respective standard deviations (σ); and the T_d-^{85}Kr correlation coefficient (r) and number of samples (n) at each station.

Station	T_d (°F)	σ	^{85}Kr (pCi/Scm^{-1})	σ	r	n
IND	36	15	14.0	0.7	−0.24	190
DET	32	13	14.1	1.1	−0.05	190
MSP	23	16	14.3	1.1	−0.32	214
RST	27	15	14.1	0.4	−0.65	197
ALO	29	14	14.0	0.4	−0.62	185
DSM	32	14	13.9	0.4	−0.64	218
OMA	33	14	13.9	0.4	−0.68	204
CNK	37	13	13.8	0.4	−0.53	203
COU	36	14	13.8	0.3	−0.64	203
ICT	36	14	13.8	0.4	−0.57	181
UMN	38	12	13.7	0.3	−0.57	184
TUL	40	14	13.7	0.4	−0.70	164
OKC	42	15	13.6	0.4	−0.57	189
IND*	36	15	13.9	0.4	−0.72	183
DET*	32	13	14.0	0.3	−0.62	186
MSP†	23	16	14.1	0.5	−0.66	208

* Kr-85 concentrations attributed to Savannah River Plant eliminated.

† Kr-85 concentrations attributed to unknown source eliminated.

There is an increase in the average dew point temperature from north to south with an associated decrease in the average ^{85}Kr concentration as shown graphically in Fig. 5. The gradients in the dew point and ^{85}Kr indicate that the northerly stations experienced more outbreaks of Canadian air containing higher background ^{85}Kr concentrations than the more southerly stations. In Fig. 5, curves were fitted "by eye" through the ^{85}Kr and dew point data for those stations approx. 1500 km from NRTS. Data points for the furthest sampling stations, DET and IND, were added after the curves were drawn; they agree with the general latitudinal trend. These data show a ^{85}Kr gradient of about 0.5 pCi/Scm^{-1} over ten degrees of latitude.

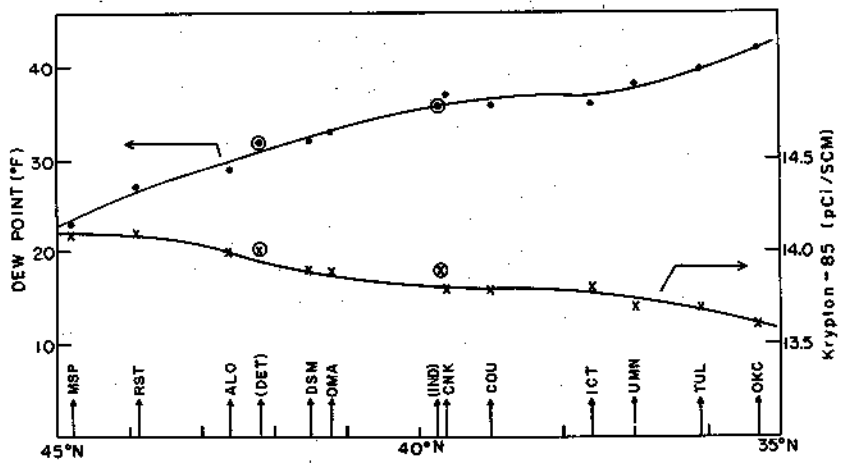

Fig. 5. Latitudinal distribution of the average ^{85}Kr concentration and average dew point temperature for the entire sampling period.

The standard deviations about the mean dew point temperatures and ^{85}Kr concentrations (Table 2) are essentially the same for all locations, lending confidence to the statistical significance of the correlation coefficients.

8. CONCLUSIONS

Over 2500 samples collected at 13 sites in the midwest over a 4-month period provided an opportunity to examine the variability of ^{85}Kr concentration on a much finer time and space-scale than had been possible previously. The influence of the NRTS plume from a nuclear fuel reprocessing plant was barely discernible at sites about 1500 km downwind.

Although almost all samples were within 5% of the ambient background, ^{85}Kr concentrations showed easily recognized changes from one air mass to another. Within an air mass there was usually relatively little fluctuation in the concentrations. Surface dew point temperature proved useful as an air mass indicator that is highly correlated with ^{85}Kr fluctuations.

On several occasions, plumes from the Savannah River Plant were identified at concentrations well above background levels at Indianapolis and Detroit, about 1000 km from the source. These cases will be reported in another paper dealing with the verification of the ARL transport and diffusion model.

It appears that ^{85}Kr emitted from a nuclear fuel reprocessing plant would be an excellent tracer for plume studies out to several hundred kilometers from a source. The samplers used in this study are now deployed at sites surrounding the Savannah River Plant to study the distribution of monthly, seasonal and annual air concentrations within 100 km of a quasi-continuous pollutant source.

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REFERENCES

AIRCO, Inc. (1972) Cryogenic air sampler, AIRCO, Inc., Murray Hill, NJ 07974, 5 pp.

AIRCO, Inc. (1973) Cryogenic air sampler, installation and maintenance manual, AIRCO, Inc., Murray Hill, NJ 07974, 33 pp.

Coleman J. R. and Liberace R. (1966) Nuclear power production and estimated Krypton-85 levels. *Radiat. Data Reports* **7**, 615–622.

Diethorn W. S. and Stockho W. L. (1972) The dose to man from atmospheric Kr-85. *Hlth Phys.* **13**, 653–662.

Heffter J. L., Taylor A. D. and Ferber G. J. (1975) A regional-continental scale transport, diffusion and deposition model. NOAA Tech. Memo., ERL-ARL-50, U.S. Dept. of Commerce, NOAA, Air Resources Laboratories, Silver Spring, MD 20910, 28 pp.

Knox J. B. and Peterson K. R. (1972) Estimates of dose to northern hemisphere population groups from Kr-85 emitted by a single nuclear fuel reprocessing plant. *Nucl. Safety* **13**, 130–135.

Machta L., Ferber G. J. and Heffter J. L. (1974) Regional and global scale dispersion of Kr-85 for population-dose calculations. Symposium, FAO/IAEA, *Physical Behaviour of Radioactive Contaminants in the Atmosphere.* IAEA-SM-181/7, pp. 411–426, Vienna.

National Council on Radiation Protection and Measurements (1975) Krypton-85 in the atmosphere. NCRP Report No. 44, Washington, D.C. 20014, 79 pp.

Pannetier R. (1968) Distribution, atmospheric transfer and balance of Krypton-85. Atomic Energy Commission, Fontenay-aux Roses; France, CEA-R-3591, 177 pp.

Schröder K. J. P. and Roether W. (1974) The release of Krypton-85 and tritium to the environment and Krypton-85 to tritium ratios as source indicators, FAO/IAEA Symposium, *Isotope Ratios as Pollutant Source and Behaviour Indicators*, pp. 231–253. IAEA-SM-191/30, Vienna.

Telegadas K. and Ferber G. J. (1975) Atmospheric concentrations and inventory of Krypton-85 in 1973. *Science* **190**, 882–883.

United Nations (1972) Ionizing radiation: levels and effects, United Nations publication Sales No. E.72.1X. 17, New York, 197 pp.