Atmospheric Concentrations and Inventory of Krypton-85 in 1973

Abstract. *Measurements of the concentrations of ^{85}Kr in the atmosphere were made in 1973 between 1.5 and 20 kilometers. The global inventory of ^{85}Kr for 1973 calculated from these data is 53 megacuries. The stratospheric distribution of ^{85}Kr concentrations shows a maximum in the lower equatorial region of each hemisphere and a minimum concentration in the polar regions. This distribution tends to confirm the mean stratospheric circulation as deduced from other radioactive tracer data. Both ^{85}Kr and Fluorocarbon-11 (CCl_3F, here designated as F-11) are inert gases released principally into the surface air in the temperate latitudes of the Northern Hemisphere. The stratospheric distribution of ^{85}Kr is very similar to that of F-11. Above 20 kilometers a photodissociation of fluorocarbons may take place, producing atomic chlorine which may act as a catalyst in the destruction of stratospheric ozone. Since ^{85}Kr has no known significant sinks and its half-life is known, measurements of the concentration of both of these gases above 20 kilometers may provide insight regarding the rate of dissociation of F-11.*

The radioisotope ^{85}Kr of the noble gas krypton (half-life, 10.76 years) is produced by nuclear fission mainly as a result of the operation of nuclear reactors and, to a lesser extent, in nuclear test explosions. Almost all the ^{85}Kr produced in nuclear reactors is released to the atmosphere, not at the reactor site but in the course of the dissolution of the spent fuel at nuclear reprocessing plants. The main source region for ^{85}Kr is the temperate 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 (*1, 2*). Pannetier (*3*) has reported measurements of ^{85}Kr at the surface which indicate that it is fairly well mixed between hemispheres with the average Southern Hemisphere concentration being about 0.8 of the average Northern Hemisphere concentration during 1964. Pannetier has also obtained some upper tropospheric measurements, principally in the Northern Hemisphere, which suggest little vertical gradient of concentration.

Since ^{85}Kr is an inert gas and has a relatively long half-life, it has been possible to make projections of its accumulation in the atmosphere at future times (*4*). Other investigators have used regional and global scale dispersion models for estimating population exposure (*5*). Recently Schröder and Roether (*2*) estimated the global inventory of ^{85}Kr at the end of 1973 and the fraction of the total derived from nuclear power production, weapon plutonium production, and weapons tests.

Measurements of ^{85}Kr between 1.5 and 20 km were made during 1973 (*6*). The purpose of this report is to alert other investigators to these data and to furnish some insight into atmospheric circulation

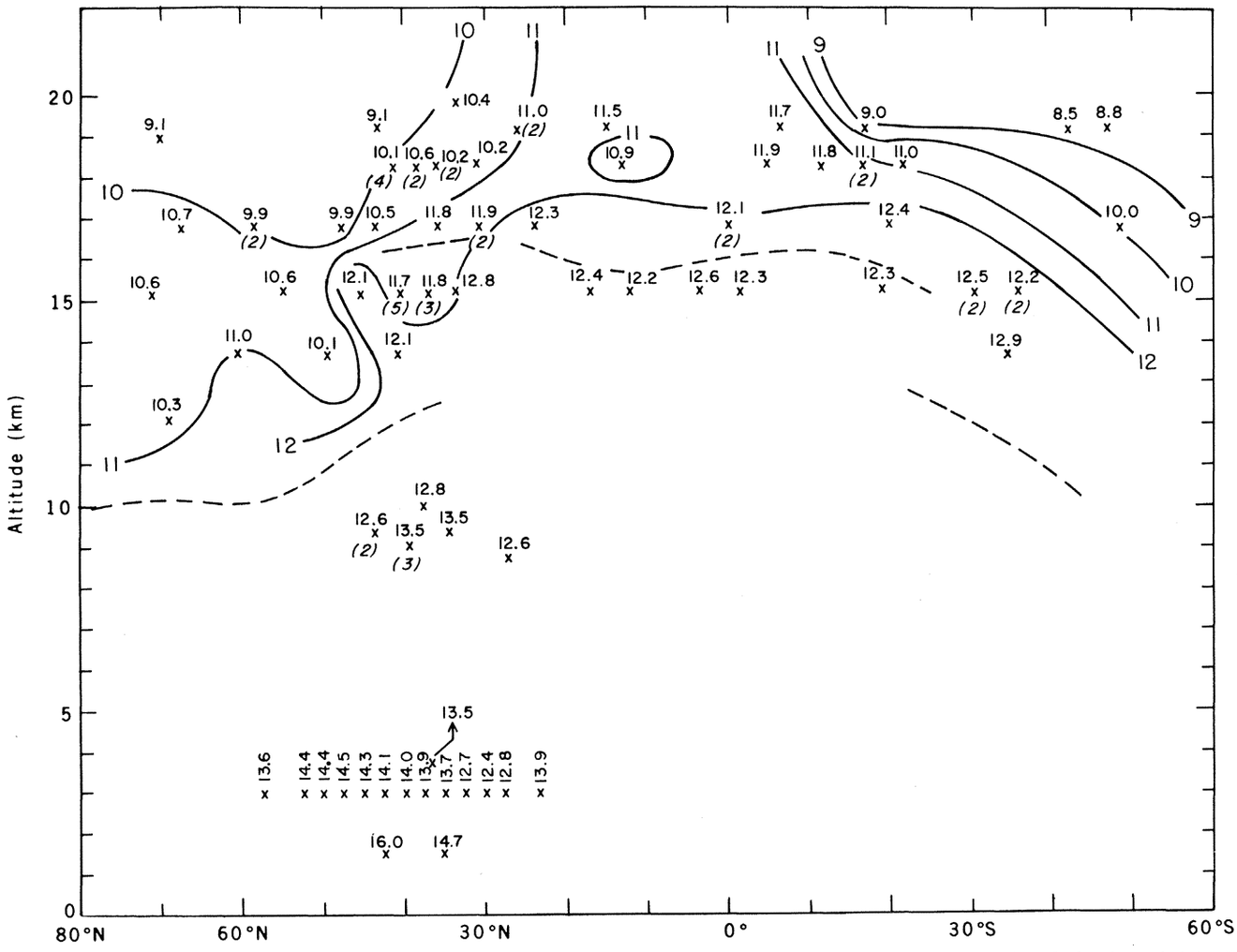

Fig. 1. Atmospheric distribution of ^{85}Kr in units of picocuries per standard cubic meter of air (computed at 76 cm-Hg and 15°C) for the period April through November 1973. The dashed lines represent the mean tropopause along the aircraft sampling corridor. Each number in parentheses indicates the number of samples used to determine the mean value given beside the cross.

patterns. All of the ^{85}Kr data for 1973 were plotted on a latitudinal cross section, and lines of equal concentration were drawn (Fig. 1). Because of the density of the data at 3 km, these data were averaged over a running 5° latitude band and plotted at 2.5° increments.

The highest observed ^{85}Kr concentrations are found in the lower troposphere at 40°N to 50°N as one would expect since this is the main source region. However, the highest observed stratospheric concentrations are in the lower equatorial stratosphere of each hemisphere. Inspection of Fig. 1 also indicates that during this period the lower stratosphere of both the Northern Hemisphere and the Southern Hemisphere exhibit very similar distributions, even though the principal source region is in the north temperate latitudes.

Tropospheric measurements of ^{85}Kr in 1973 were restricted to the Northern Hemisphere temperate latitudes. Figure 2 shows the vertical profile of ^{85}Kr based on data from 35°N to 45°N in Fig. 1. The average surface concentration in Fig. 2 is that given for January through June 1973 for ten stations in the contiguous United States (30°N to 45°N) reported by the Environmental Protection Agency (*7*). The decrease of the ^{85}Kr concentration with altitude is in qualitative agreement with a ground level source. The inset in Fig. 2 shows a smoothed profile of the ^{85}Kr concentration on a pressure scale with an average tropopause height estimated from Fig. 1 at about 150 mbar. An average tropospheric concentration of ^{85}Kr of 13.8 pc per standard cubic meter of air (SCM) was estimated from the inset. Using the stratospheric distribution given in Fig. 1 and the tropospheric distribution shown in Fig. 2, Telegadas and Ferber (*6*) estimated the total global burden of ^{85}Kr in the atmosphere for 1973 to be 53 Mc. The main sources of error are the lack of data in the Southern Hemisphere troposphere and, to a lesser extent, in the upper stratosphere (above 20 km) which contains about 5 percent of the total mass of the atmosphere. We estimate the uncertainty for the total inventory to be ± 5 Mc of ^{85}Kr. Schröder and Roether (*2*), using limited surface data, estimated a global atmospheric burden of 55 Mc of ^{85}Kr at the end of 1973. This is in agreement with our estimate based on considerably more data to an altitude of 20 km.

The release of ^{85}Kr into the atmosphere can be viewed as a long-term global tracer experiment with a quasi-continuous release over the past two decades. The distribution in Fig. 1 represents the integrated effects of transport, diffusion, and decay processes over many years. A schematic representation of the stratospheric circulation as deduced from radioactive tracers that have been injected into the stratosphere over many years has been presented by List and Telegadas (*8*). The almost symmetrical distribution of ^{85}Kr in the lower equatorial stratosphere (Fig. 1), with a minimum concentration in the polar regions, is in accordance with their picture of the mean stratospheric circulation.

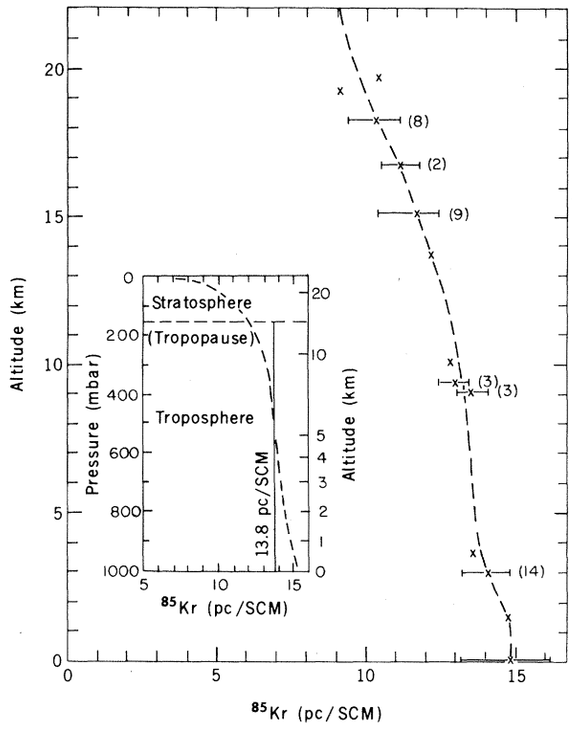

Fig. 2. The ^{85}Kr profile over 35°N to 45°N, April through November 1973. Each number in parentheses indicates the number of samples used to determine the mean value given beside the cross. Horizontal bars denote the range of the data sets. The inset diagram denotes the same profile on a pressure-altitude scale.

Additional measurements of ^{85}Kr in the stratosphere might shed light on questions concerning possible depletion of stratospheric O_3 by Fluorocarbon-11 (CCl_3F, here designated as F-11) and other halocarbons. It has been suggested that photodissociation of chlorine-containing compounds may take place above 20 km in the stratosphere (*9*). This would produce atomic chlorine which may act as a catalyst in reactions leading to the depletion of stratospheric O_3.

The compound F-11, used mainly as a propellant in spray cans, is released primarily in the temperate latitudes of the Northern Hemisphere. Like ^{85}Kr, F-11 is an inert gas with no known significant tropospheric sinks and its production and release to the atmosphere have increased rapidly over the past two decades. Krey and Lagomarsino (*10*), who have analyzed F-11 from gas samples taken primarily in the stratosphere of both hemispheres, reported a distribution very similar to that of ^{85}Kr presented in Fig. 1. However, if F-11 undergoes dissociation, its distribution above 20 km should differ from that of ^{85}Kr, which has no sink in this region. More measurements of both chlorine-containing compounds and ^{85}Kr should be made in the stratosphere to at least 40 km. The distributions of both of these components can then be compared to give some insight not only into meteorological processes but also into the O_3 depletion problem.

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