Mem. Natl Inst. Polar Res., Spec. Issue, **40**, 462–466, 1986

THE DISTRIBUTION OF ^{85}Kr IN THE AIR OVER THE NORTH AND SOUTH PACIFIC OCEAN

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Abstract: The north-south section of the atmospheric concentration of ^{85}Kr over the Pacific is first reported. The most striking feature of the section is a large concentration difference at the geographical position of the intertropical convergence. The ^{85}Kr concentration in the air over the North Pacific ranged from 22 to 23 pCi m^{-3} and in the South Pacific 16 pCi m^{-3}. The concentration differences between two hemispheres is about 6 pCi m^{-3}. Five year records from land stations in Japan indicate that the annual increase is about 1 pCi m^{-3}y^{-1}. It seems that the amount of the southward transport of ^{85}Kr from the northern hemisphere is nearly equilibrated with that of radioactive disintegration and/or dissolution of ^{85}Kr into the ocean in the southern hemisphere.

1. Introduction

^{85}Kr is a radioactive noble gas (β-emitter, 10.75 y half-life) which is a nuclear fission product. It is released mainly from a nuclear fuel reprocessing plant (SITTKUS and STOCKBURGER, 1976), and in a lesser extent by the nuclear weapon testing or a nuclear power plant. It is being added to the atmosphere corresponding to the increase of the nuclear power generation and nuclear weapon production in the world.

Because of its properties as a noble gas, most of krypton are not effectively removed from the atmosphere, and consequently it would be accumulated into the atmosphere. The source area of ^{85}Kr is located mainly in the temperate zone of the northern hemisphere, between 30 and 50°N (HILBERT, 1974), and its release into the atmosphere might have been started since the latter half of the 1940s.

The major sink of ^{85}Kr in the atmosphere is radioactive disintegration and absorption into the ocean water. Therefore, the time-dependent distribution of ^{85}Kr in the air over the Earth may serve as an effective transient tracer for the evaluation of dispersion and diffusion processes in the atmospheric environment (PANNETIER, 1968; KAROL *et al.*, 1976; WEISS *et al.*, 1983).

According to the results of previous studies (PANNETIER, 1968; WEISS *et al.*, 1983), about 1 pCi m^{-3}y^{-1} of increase on an average was observed in recent year. However, up to now, little is known on the concentration and distribution of ^{85}Kr in the air over the Pacific Ocean.

In this paper, the present authors intend to give the first report on the recent trend of horizontal distribution of ^{85}Kr in the surface air over the Pacific Ocean, together with the results of determination of ^{85}Kr at five stations in Japan in recent years. The

results of study indicate that the concentration differences between the northern and southern hemispheres are about 6 pCi m^{-3} and the concentration in the southern hemisphere is relatively lower than that in the northern hemisphere. And the recent increase rate of ^{85}Kr in the surface air in Japan also maintained the annual rate of about 1 pCi $m^{-3}y^{-1}$.

2. Samples and the Method of Analysis

The procedure consists of compressed air sampling, cryogenic distillation and liquid scintillation counting.

The sample air is collected on board by using a small air compressor to fill an evacuated air cyclinder of 14 *l* at 120 atm during the cruise of R. V. HAKUHO MARU which belongs to the Ocean Research Institute, University of Tokyo (BIOMASS 1983, from November 1983 to February 1984) from the North Pacific to the Southern Ocean. The track of the cruise is shown in Fig. 1.

In the laboratory on land, separation of ^{85}Kr from the air sample was done by the cryogenic distillation method as follows: The cryogenic vacuum apparatus consisted of an all glass system containing a vacuum manifold and a series of traps as shown in

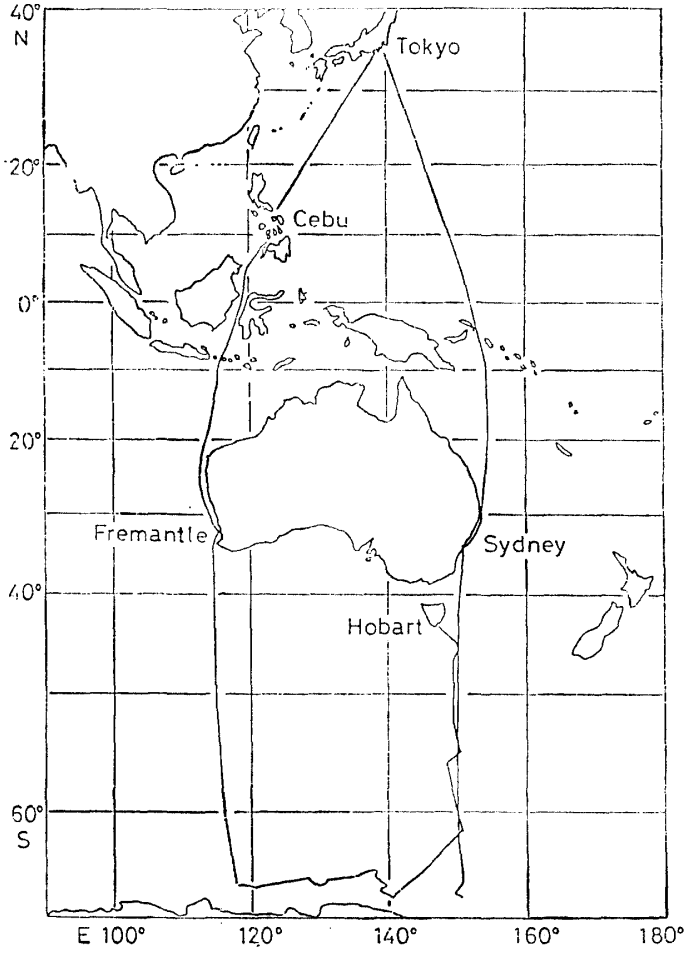

Fig. 1. Track of BIOMASS 1983 cruise in the Pacific Ocean.

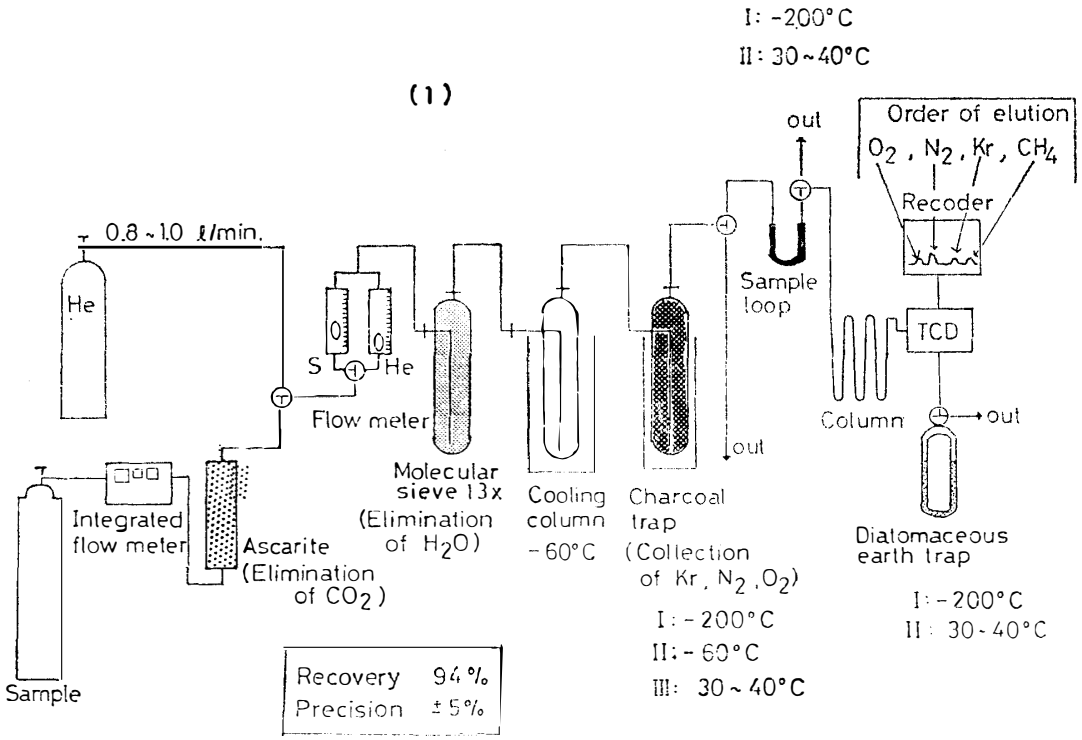

Fig. 2. A schematic diagram of ^{85}Kr separation system.

Fig. 2. The final separation of ^{85}Kr was done on the TCD gas chromatographic analyzer.

The detailed analytical procedure will be given elsewhere but in this paper only a brief explanation of the method is given as follows:

The cylinder is connected to the inlet of the cryogenic vacuum system through an integrated flow meter. The sample is leaked into the system using a reduced pressure. The gas flow is passed through an ascarite column, a trap containing molecular sieve 13X, a precooling coil at $-60°C$, and finally enters an active charcoal trap immersed in liquid nitrogen.

Oxygen, nitrogen and argon which are collected together with ^{85}Kr are removed from the charcoal trap by raising the trap temperature with a $-60°C$ bath and purging them with the helium stream at a flow rate of 1 $l\,min^{-1}$ and 1 atm for 1 h. Through this procedure, most of nitrogen and oxygen are removed from the condensate retaining krypton in the trap. The remainder of the sample condensate in the trap is transferred to the sample loop immersed in liquid nitrogen by raising the trap temperature with hot water to 40°C under the helium stream.

Further removal of krypton from other elements is carried out on the gas-chromatographic system with the thermal conductivity detector (TCD) and separation column containing 0.4 g active charcoal and 16 g of molecular sieve 5A (8 mm dia. and 2 m long at 40°C). Argon, oxygen and nitrogen are eluted from the first and second fractions, and finally krypton is eluted from the column with the retention time of about 13 min.

The krypton fraction is collected in a diatomaceous earth trap at liquid nitrogen temperature, and at the time of counting it is transferred into a glass counting vial

containing 3 g of silica gel at liquid nitrogen temperature. After the complete transfer of krypton into the vial, 22 m*l* of scintillation solution (5 g of PPO and 300 mg of POPOP in toluene) was added to the vial to dissolve the krypton condensate.

The measurement of low energy beta activity of ^{85}Kr was done on a low background liquid scintillation counter for 50 min. The recovery in the whole procedure starting from 1 m^3 air is estimated to be 94±5% by the duplicate analysis of the same air sample.

3. Results and Discussion

The results of determination of ^{85}Kr in the air over the ocean from Japan to Antarctica are shown in Fig. 3. As shown in the figure, the concentration in the air over the North Pacific ranged from 22 to 23 pCi m^{-3}, and crossing the geographical location of the intertropical convergence (ITCZ) around 5°S the concentration decreased to 16 pCi m^{-3}. Over the South Pacific, the concentration showed a fairly uniform distribution and a slight increase was observed in the Southern Ocean area. It appeared that the concentration difference between the two hemispheres is about 6 pCi m^{-3}.

The similar distribution of ^{85}Kr in the air over the Atlantic Ocean was reported by WEISS *et al.* (1983) for the cruise of M. S. METEOR in 1980 to 1981. However, the concentration difference at ITCZ is only 1.5 pCi m^{-3}.

The differences of the observed values between two cruises are quite reasonable considering the annual increase rate of ^{85}Kr in the air recently in Japan. According to the results of determination of ^{85}Kr in the air at five stations in Japan from 1979 to 1983, the concentration increased from 16.4±0.4 pCi m^{-3} in 1979 to 21.9±1.1 pCi m^{-3} in 1983 with the average increasing rate of 1.4 pCi m^{-3}y^{-1}.

The results of the present study of ^{85}Kr in the air over Japan are shown in Fig. 4 together with those of previous works in the northern hemisphere. In the same figure, the total capacity of nuclear power generation in the world is given based on the information in the IAEA reports.

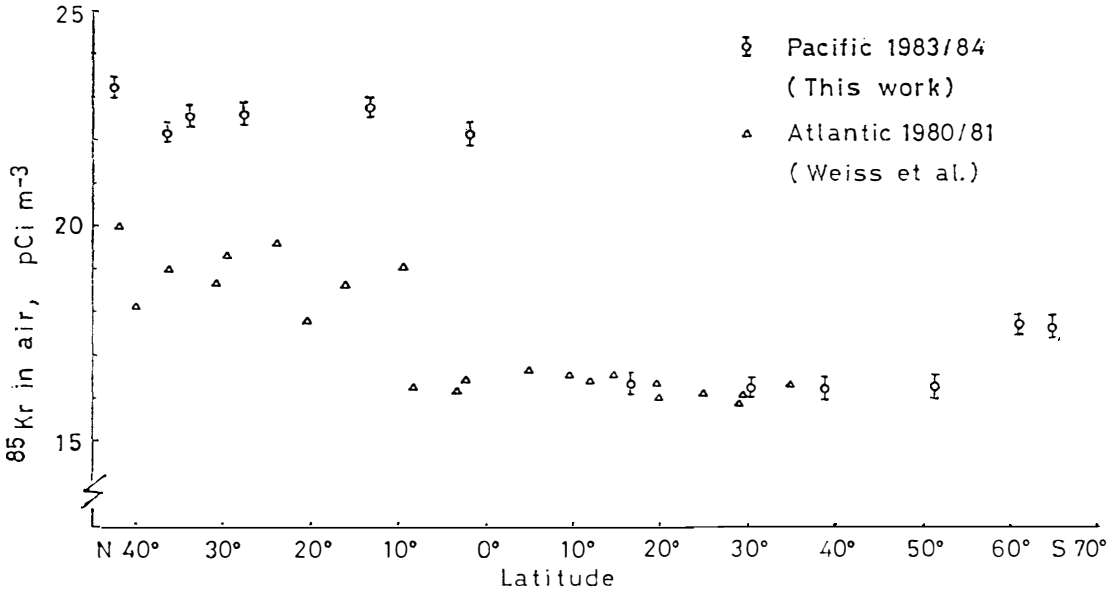

Fig. 3. Meridional distribution of ^{85}Kr in the surface air over the North and South Pacific.

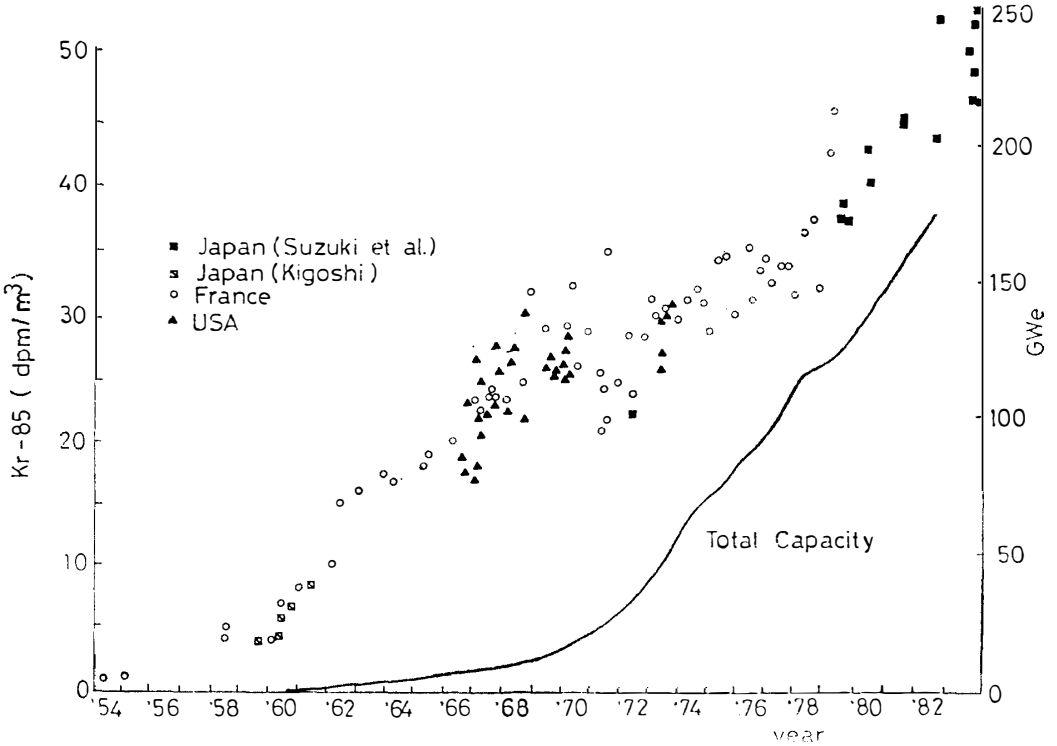

Fig. 4. The concentration of ^{85}Kr in the air in the northren hemisphere from 1960 to 1983, together with the total capacity of nuclear power generation in the world.

As seen in the figure, the concentration of ^{85}Kr in the air increased from 1960 to 1970 at the rate of 1.2 pCi $m^{-3}y^{-1}$. From 1970 to 1974, the increasing rate decreased slightly and again it increased to about 1 pCi $m^{-3}y^{-1}$ after 1974 to the present. The recent increase of the atmospheric ^{85}Kr is in fairly good agreement with that of the capacity of nuclear power generation in the world.

The observed pile-up of ^{85}Kr in the air in the northern hemisphere and the relatively slow increase in the southern hemisphere may indicate the relatively slower rate of north-south mixing of the surface air between the northern and the southern hemispheres as compared with those given by the previous researchers.

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(Received May 1, 1985; Revised manuscript received September 7, 1985)