

# Spatial and Temporal Variations of Atmospheric $^{85}\text{Kr}$ Observed During 1995–2001 in Japan: Estimation of Atmospheric $^{85}\text{Kr}$ Inventory in the Northern Hemisphere

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## Atmospheric $^{85}\text{Kr}$ /Annual growth rate/Nuclear fuel reprocessing plant/Emission/Inventory

Atmospheric  $^{85}\text{Kr}$  concentrations have been continuously monitored since 1995 at the Meteorological Research Institute (MRI) in Tsukuba, Japan. They have also been observed once a year at several stations over the Japanese islands since 1995. The annual growth rate of the background atmospheric  $^{85}\text{Kr}$  concentrations in Tsukuba was  $0.03 \text{ Bq}\cdot\text{m}^{-3}\cdot\text{yr}^{-1}$  during 1996–2001. The atmospheric  $^{85}\text{Kr}$  concentrations at several stations over Japan were within the range of the annual variations in Tsukuba. However, higher and lower  $^{85}\text{Kr}$  concentrations in early winter, compared with those in Tsukuba ( $36.1^\circ\text{N}$ ,  $140.1^\circ\text{E}$ ), occurred in Sapporo ( $43.1^\circ\text{N}$ ,  $141.3^\circ\text{E}$ ) and Ishigaki ( $24.3^\circ\text{N}$ ,  $124.2^\circ\text{E}$ ), respectively. The reason for this is that Sapporo is covered by a continental air mass, some from European sources, whereas Ishigaki is still covered by a subtropical air mass. The Northern Hemispheric background  $^{85}\text{Kr}$  concentrations from 1994 to 2001 was calculated from the  $^{85}\text{Kr}$  inventory and the release rate of  $^{85}\text{Kr}$  from the nuclear fuel reprocessing plants in Europe. Calculated  $^{85}\text{Kr}$  concentrations in surface air were in good agreement with annual average observed values at the MRI, Tsukuba. The global atmospheric inventory of  $^{85}\text{Kr}$  in December 2001 was also estimated to be approximately 5 EBq by using observed data in Tsukuba.

## INTRODUCTION

$^{85}\text{Kr}$  is a radioactive noble gas that is a beta emitter with a half-life of 10.76 years. Natural sources of  $^{85}\text{Kr}$  are nuclear reactions by cosmic radiation in the upper atmosphere and spontaneous fission of the heavy elements in the Earth's crust<sup>1</sup>. However most of the  $^{85}\text{Kr}$  in atmosphere is derived from anthropogenic sources, i.e., nuclear weapons tests and nuclear reactors. At present, the major sources of  $^{85}\text{Kr}$  are releases from the nuclear fuel reprocessing plants in Europe<sup>2,3,4</sup>. Because of its inert property, the only sink of

$^{85}\text{Kr}$  in the earth's surface is a radioactive decay at the rate of about 6% a year. The unbalance between sinks and sources of  $^{85}\text{Kr}$  changes its global inventory. The amount of released  $^{85}\text{Kr}$  from nuclear fuel reprocessing plants in Europe is now 300–400 PBq per year<sup>5</sup>. In fact, the atmospheric  $^{85}\text{Kr}$  concentrations are increasing year by year<sup>6,7</sup>.

At the Meteorological Research Institute (MRI) in Tsukuba, Japan, atmospheric  $^{85}\text{Kr}$  concentrations have been observed since 1995 in collaboration with the Bundesamt für Strahlenschutz (BfS), Germany<sup>6</sup>. Recently Igarashi *et al.* (2001)<sup>8</sup> have developed a  $^{85}\text{Kr}$  measuring system based on the BfS method. It is important to monitor the background level of the atmospheric  $^{85}\text{Kr}$  concentrations in Japan because the operation of a new commercial nuclear fuel reprocessing plant is scheduled in Aomori Prefecture in the northernmost part of Honshu Island, Japan, from 2005<sup>9</sup>. The emission of  $^{85}\text{Kr}$  at the new plant will be at the same level as those at European plants and can influence the background level of atmospheric  $^{85}\text{Kr}$  concentrations in the Northern Hemisphere<sup>6</sup>.

In this paper, we discuss the following two subjects:

- (1) The annual growth rate and the seasonal variations of the background atmospheric  $^{85}\text{Kr}$  concentrations in the

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**Table 1.** Sampling locations and related information.

Station	Location	Sampling period	Frequency of sampling	Height of intake
Tsukuba	36.05°N, 140.13°E	1 week	every week	40 m
Sapporo	43.06°N, 141.33°E	3 days	once a year	17.2 m
Ishigaki	24.33°N, 124.16°E	3 days	once a year	5.7 m
Wakkanai	45.41°N, 141.68°E	2 hours	once a year	10.6 m
Sendai	38.26°N, 140.90°E	2 hours	once a year	38.9 m
Osaka	34.68°N, 135.52°E	2 hours	once a year	92.6 m
Fukuoka	33.58°N, 130.38°E	2 hours	once a year	17.1 m

middle of Japan as well as the spatial distributions of the background  $^{85}\text{Kr}$  concentrations throughout Japan.

- (2) The atmospheric background  $^{85}\text{Kr}$  concentrations in the Northern Hemisphere from the emissions of  $^{85}\text{Kr}$  at the nuclear fuel reprocessing plants in Europe.

## MATERIALS AND METHODS

### Sampling stations

Air sampling has been carried out at the MRI in Tsukuba, about 60 km northeast from Tokyo. As an adjacent source of  $^{85}\text{Kr}$ , a nuclear fuel reprocessing plant has been operated by the Japan Nuclear Cycle Development Institute (JNC) at Tokai, about 60 km northeast from the MRI.

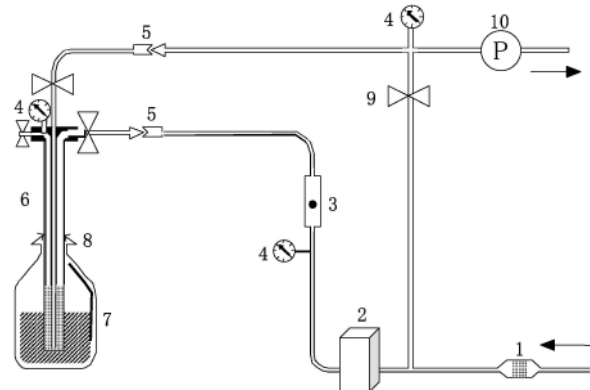
We observed the atmospheric  $^{85}\text{Kr}$  concentrations at several stations (Ishigaki Island, Fukuoka, Osaka, Sendai, Sapporo, and Wakkanai) that cover the area of Japan from south to north. Details of the sampling stations are shown in Table 1.

### Method of sampling and analysis

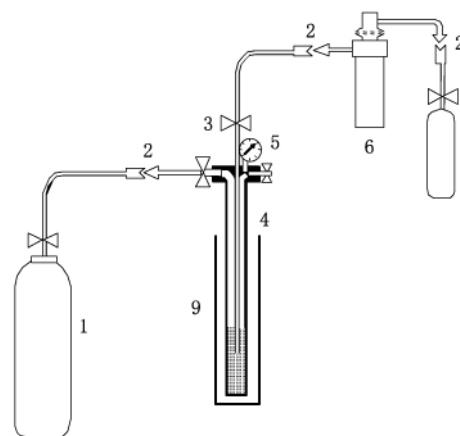
The method of sampling atmospheric  $^{85}\text{Kr}$ , based on the BfS, is briefly described below. The details are described elsewhere<sup>8,10,11</sup>.

Figure 1 shows a diagram of the  $^{85}\text{Kr}$  sampling system. Surface air was passed through the glass fiber filter (Whatman International Ltd., Whatman®, 47 mmφ, GF/F). Water vapor was removed by the cooling trap at 8°C. Dry air was introduced into the metal vessel that contained about 200 g of the activated charcoal (Merck Co., Ltd., 35–50 mesh ASTM) at liquid nitrogen temperature. At this temperature,  $^{85}\text{Kr}$  was adsorbed onto the activated charcoal. To vent excess air, a bypass line was attached in the center of the sampling system. The flow rate of sample air was kept at 1 l·min<sup>-1</sup> and about 10 m<sup>3</sup> of air was passed through the adsorbent in a week.

After the collection of the sample, the adsorbent was warmed to the room temperature. To transfer Kr fraction into a 1 l aluminum sampling bottle (Linde Co., Ltd., Minican®), the sampling bottle and a desiccant holder containing silica gel were set on the adsorbent (see Fig. 2). The adsorbent was



**Fig. 1.** Diagram of the  $^{85}\text{Kr}$  sampling system. 1: Air filter (Whatman®, GF/F 47 mmφ) 2: Cooling trap 3: Flowmeter 4: Manometer 5: Quick-snap 6: Adsorbent 7: Dewar (30 l) 8: Rubber collar 9: Bypass valve 10: Pump



**Fig. 2.** Diagram of the transfer system. 1: Helium gas cylinder 2: Quick-snap 3: Stopcock 4: Adsorbent 5: Manometer 6: Desiccant holder 7: Needle valve 8: Aluminum bottle 9: Heating unit

heated at 300°C for 1 hour, and the Kr fraction adsorbed on the activated charcoal was transferred into the sampling bottle through the desiccant holder. After heating, pure helium gas (purity is >99.9999%) was passed through the adsorbent and injected into the sampling bottle up to 0.4 MPa to transfer the Kr fraction to the sampling bottle.

The sampling procedures in Sapporo and Ishigaki were the same as in Tsukuba. But the sampling period was four days shorter. In Wakkanai, Sendai, Osaka, and Fukuoka, Tedler<sup>®</sup> bags (GL Science Inc.) made of polyvinyl fluoride film or balloons (Daiwa Advertise Corp.) made of polyvinyl chloride were used for each sampling. The volumes of the Tedler<sup>®</sup> bag and the balloon were approximately 0.4 m<sup>3</sup> and 0.6 m<sup>3</sup>, respectively. The air sample was injected into three of the Tedler<sup>®</sup> bags or the balloons at each sampling station by using a diaphragm pump at a rate of 20 l·min<sup>-1</sup>. The sampling time was 2 hours. These air samples were sent to the MRI, and Kr was recovered by using the adsorbent and then transferred to the aluminum sampling bottle.

Air samples have been collected once a week at the MRI, Tsukuba, throughout a year and once a year at other stations in various seasons.

All the sampling bottles were sent to the BfS and subjected for a determination of the concentrations of  $^{85}\text{Kr}$ . Details of the  $^{85}\text{Kr}$  measuring system at the BfS is described in a WMO report (1996)<sup>11</sup>. The atmospheric  $^{85}\text{Kr}$  concentrations were calculated from the specific activity (the activity of  $^{85}\text{Kr}$  over the volume of stable Kr) multiplied by the fraction of Kr in the atmosphere (1.14 ppmv). The precision of the measurement is about 1%, and the detection limit is less than 0.01 Bq·m<sup>-3</sup><sup>11</sup>.

The  $^{85}\text{Kr}$  measuring system at the MRI was established in 2000<sup>8</sup>, and duplicate samples have been collected at the MRI since April 2000. One sample was analyzed at the MRI and the other at the BfS in Freiburg. The results indicated that  $^{85}\text{Kr}$  concentrations determined by the MRI were in good agreement with those obtained by the BfS, within 7% and with 4 exceptions for 86 duplicate samples during April 2000 and December 2001. In this report, the data measured by the BfS were used.

## RESULTS

We have observed the weekly average atmospheric  $^{85}\text{Kr}$  concentrations in Tsukuba since May 1995. Monthly averaged  $^{85}\text{Kr}$  concentrations are shown in Table 2. During the period that the nuclear fuel reprocessing plant at Tokai was in operation (before May 1997 and after June 2000), high  $^{85}\text{Kr}$  concentrations exceeding 2 Bq·m<sup>-3</sup> were observed. On the other hand, low  $^{85}\text{Kr}$  concentrations of less than 1.5 Bq·m<sup>-3</sup> were observed for the period of operation suspension at the Tokai plant, as described later. The monthly emissions of  $^{85}\text{Kr}$  from the Tokai plant are shown in Table 3<sup>12,13</sup>. Compared with reprocessing plants in Europe, the magnitude of the emission at the Tokai plant is less than 10% of those in Europe<sup>5</sup>.

**Table 2.** Monthly averaged atmospheric  $^{85}\text{Kr}$  concentrations in Tsukuba

	unit: Bq·m <sup>-3</sup>						
	1995	1996	1997	1998	1999	2000	2001
Jan	no data	1.26	1.26	1.32	1.37	1.35	1.36
Feb	no data	1.24	3.34	1.31	1.36	1.37	1.34
Mar	no data	1.23	1.29	1.29	1.37	1.36	1.92
Apr	no data	2.44	1.27	1.27	1.37	1.38	3.13
May	6.88	4.35	1.31	1.29	1.36	1.32	2.48
Jun	2.11	2.29	1.26	1.25	1.32	1.33	2.85
Jul	1.08	1.16	1.23	1.21	1.25	2.02	1.29
Aug	1.06	1.16	1.19	1.21	1.24	1.25	1.33
Sep	5.30	1.99	1.23	1.22	1.30	1.30	1.39
Oct	4.29	5.03	1.27	1.32	1.39	1.37	3.18
Nov	1.76	2.20	1.33	1.35	1.37	1.46	1.85
Dec	1.33	1.23	1.36	1.40	1.41	1.39	1.51

Counting error (1 $\sigma$ ) was less than 0.80%.

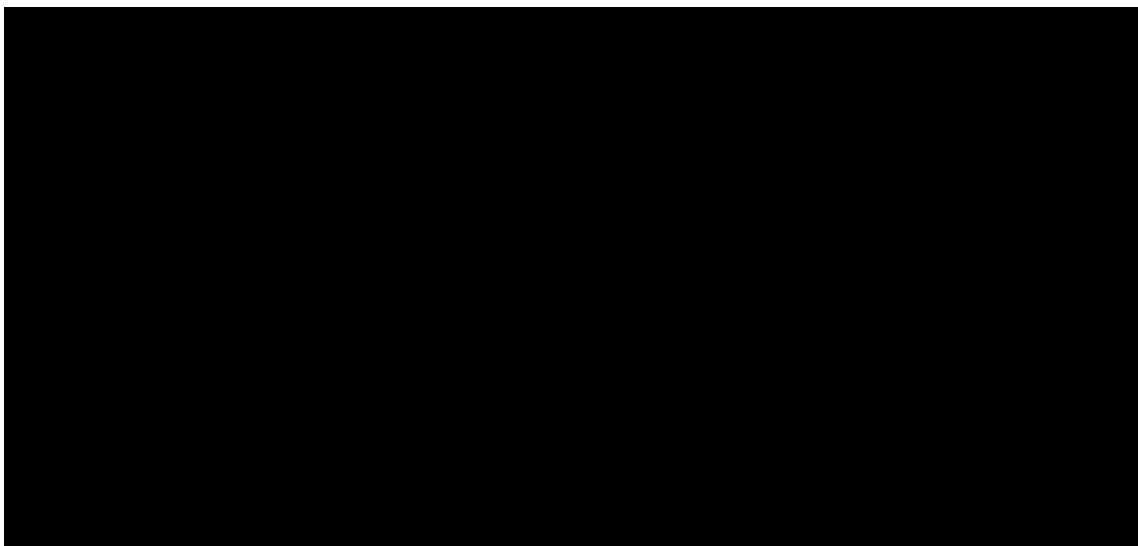
**Table 3.** Monthly  $^{85}\text{Kr}$  emissions at the Tokai nuclear fuel reprocessing plant

	unit: PBq						
	1995	1996	1997	1998	1999	2000	2001
Jan	0.16	0.00	0.00	0.00	0.00	0.00	0.00
Feb	3.02	0.00	1.88	0.00	0.00	0.00	0.00
Mar	0.93	0.00	0.97	0.00	0.00	0.00	0.77
Apr	1.92	1.30	0.00	0.00	0.00	0.00	0.79
May	2.60	1.82	0.00	0.00	0.00	0.00	1.13
Jun	0.49	0.38	0.00	0.00	0.00	0.07	0.29
Jul	0.00	0.00	0.00	0.00	0.00	0.55	0.00
Aug	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Sep	0.45	0.66	0.00	0.00	0.00	0.00	0.00
Oct	1.59	3.45	0.00	0.00	0.00	0.00	0.81
Nov	0.71	1.60	0.00	0.00	0.00	0.13	0.57
Dec	0.80	0.00	0.00	0.00	0.00	0.12	0.00
Total	12.66	9.22	2.86	0.00	0.00	0.86	4.35

## DISCUSSION

### *The atmospheric $^{85}\text{Kr}$ concentrations in Tsukuba*

Figure 3 depicts the atmospheric  $^{85}\text{Kr}$  concentrations in Tsukuba observed from May 1995 to December 2001. Sporadic high  $^{85}\text{Kr}$  concentrations are due to the effect of the release of  $^{85}\text{Kr}$  from a nuclear fuel reprocessing plant at



**Fig. 3.** Weekly averaged atmospheric  $^{85}\text{Kr}$  concentrations in Tsukuba during 1995–2001. March 1997; fire and explosion accident at the nuclear fuel reprocessing plant at Tokai. September 30, 1999; JCO accident. July 2000; The experimental operation started at the nuclear fuel reprocessing plant at Tokai. March 2001; The routine operation started at the nuclear reprocessing plant at Tokai.

Tokai operated by the JNC, 60 km northeast from the MRI<sup>6,14</sup>). Because of a fire and explosion accident in March 1997<sup>6,15,16,17</sup>), the operation of the nuclear fuel reprocessing plant at Tokai had been stopped up to summer in 2000. In that period, no sporadic high  $^{85}\text{Kr}$  concentrations in surface air were observed in Tsukuba. The experimental operation of the nuclear fuel reprocessing plant was conducted in summer 2000, and its routine operation was restarted in spring 2001. Since then, sporadic increases of the atmospheric  $^{85}\text{Kr}$  concentrations have occurred again in Tsukuba.

#### *The background level of the atmospheric $^{85}\text{Kr}$ concentrations in Tsukuba*

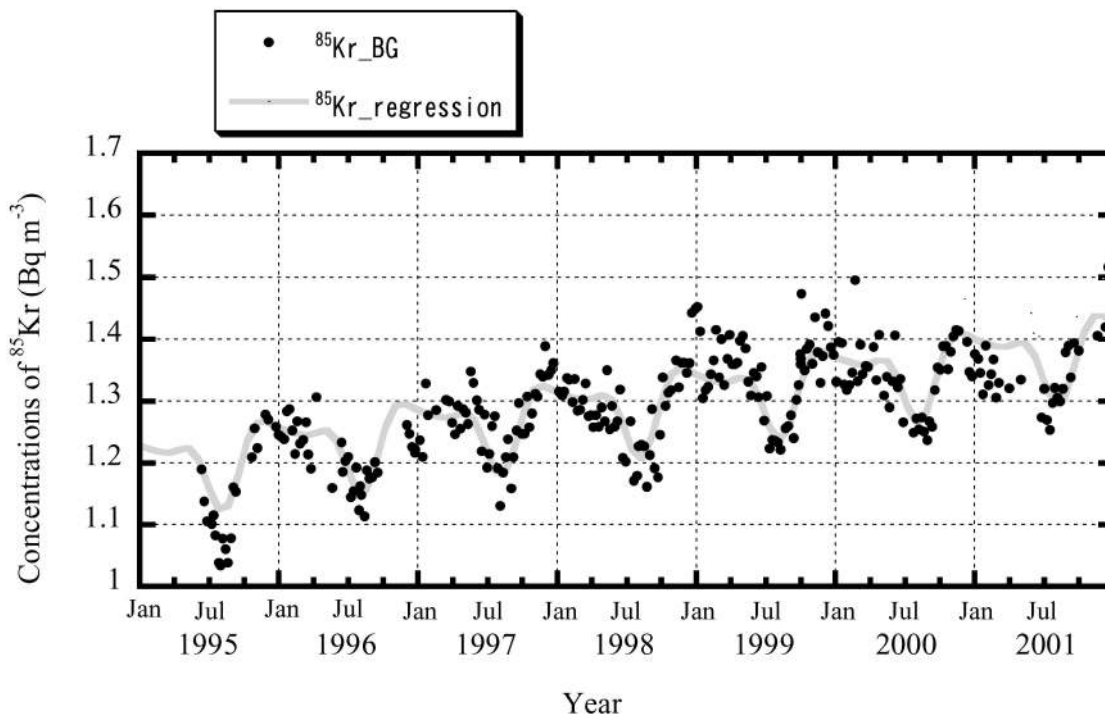
For the public's radiation protection, it is important to know the background level of the atmospheric  $^{85}\text{Kr}$  concentrations. For the background level of atmospheric  $^{85}\text{Kr}$  concentrations in Tsukuba to be known, data influenced by  $^{85}\text{Kr}$  discharge from the Tokai plant should be removed. Since the effect of  $^{85}\text{Kr}$  release from the Tokai plant to atmospheric  $^{85}\text{Kr}$  in Tsukuba depends on wind direction, wind speed, and daily  $^{85}\text{Kr}$  release rate from the Tokai plant<sup>6</sup>), all of the observed data in the sampling period, including the days when the Tokai plant has been in operation, were removed regardless of the  $^{85}\text{Kr}$  concentration measured at the MRI. The background data obtained were plotted in Fig. 4. The background atmospheric  $^{85}\text{Kr}$  concentrations in Tsukuba have been increasing, accompanied with seasonal variations, from 1995 to 2001. Assuming that the annual growth rate and seasonal variation are given by a linear function and three harmonics, respectively, we applied a curve fitting to the data<sup>6</sup>) from January 1996 to December 2001. The following equation was used:

$$\text{[Blacked out equation]} \quad (1).$$

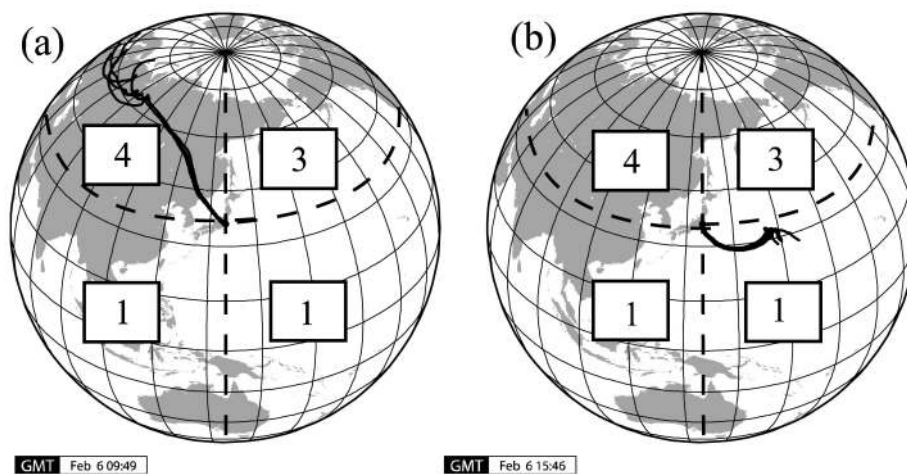
Where  $A(t)$  is the atmospheric  $^{85}\text{Kr}$  concentration at the time of  $t$ , expressed as a unit of year. The second term of Equation (1),  $a_1$ , represents the annual growth rate. As a result, the annual growth rate from 1996 to 2001 in Tsukuba was calculated to be  $0.03 \text{ Bq}\cdot\text{m}^{-3}\cdot\text{yr}^{-1}$ . This rate is at the same level as a previous estimate for the period of 1996 – 1998<sup>6</sup>). However, it must be noted that the growth rate varied year by year. The mean annual growth rate of  $0.03 \text{ Bq}\cdot\text{m}^{-3}\cdot\text{yr}^{-1}$  is independently observed at all other stations of the global BfS noble gas network.

Pollard *et al.* (1997)<sup>7</sup>) reported the atmospheric  $^{85}\text{Kr}$  concentrations at Clonskeagh, Dublin, Ireland, between 1994 and 1996. Excluding the data exceeding  $2.5 \text{ Bq}\cdot\text{m}^{-3}$  as outliers, the mean annual  $^{85}\text{Kr}$  concentration was  $1.12 \text{ Bq}\cdot\text{m}^{-3}$  during 1994 and  $1.30 \text{ Bq}\cdot\text{m}^{-3}$  during 1996, and an increasing trend of approximately  $0.1 \text{ Bq}\cdot\text{m}^{-3}\cdot\text{yr}^{-1}$  was observed. The atmospheric  $^{85}\text{Kr}$  concentrations in Dublin from 1994 to 1996 were in agreement with those observed in winter at the MRI, Tsukuba.

The background atmospheric  $^{85}\text{Kr}$  concentrations showed a clear seasonal variation, which is characterized as low in summer and high in winter. To explain this pattern of seasonal variation, we carried out the backward trajectory analysis by using  $^{85}\text{Kr}$  data observed in Tsukuba in 1999. We used GANAL compiled by the Japan Meteorological Agency for wind data analysis. A total of 240 hours backward data (starting at an altitude of 1,500 m, about 850 hPa; 00 UTC) was calculated every 5 days. Typical examples of the backward trajectory in winter and in summer are shown in Fig. 5a and 5b, respectively. In this analysis, the origin of air



**Fig. 4.** Background of the atmospheric  $^{85}\text{Kr}$  concentrations in Tsukuba. Solid circles are observed data of the weekly averaged atmospheric  $^{85}\text{Kr}$  concentrations in Tsukuba, and the solid line is a fitting curve that considers annual growth rate and seasonal variations by using the data from January 1996 to December 2001.



**Fig. 5.** Typical charts of backward trajectory in winter and in summer. The point for each category (see Table 4) is shown on the globe. (a) the chart for 10 days from January 5, 1999. (b) the chart for 10 days from August 3, 1999.

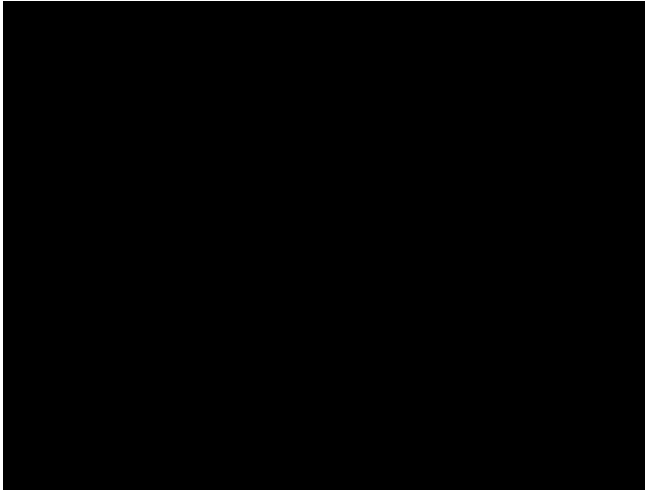
mass transported to Tsukuba was estimated. The origins were grouped by four categories centering on Tsukuba, and we introduced the “Trajectory Index” of air masses that reflects the relative distribution of the global atmospheric  $^{85}\text{Kr}$  concentrations. This index is defined as a mean of the points during the sampling period, in which the point in each day is assigned from the category of the trajectory (Table 4 and Fig. 5). Since most of the nuclear fuel reprocessing plants are in Europe, it is considered that the area classified

into category 1 shows the highest concentrations of  $^{85}\text{Kr}$ , i.e., the highest point. The areas classified into categories 3 and 4 show the lowest concentrations of  $^{85}\text{Kr}$ , i.e., the lowest point. This reflects a gradient of the atmospheric  $^{85}\text{Kr}$  concentrations between northern and southern air masses, which is in agreement with the observations reported by Weiss *et al.* (1992)<sup>4</sup>.

We examined the correlation between the “Trajectory Index” and the observed atmospheric  $^{85}\text{Kr}$  concentrations.

**Table 4.** Trajectory Index

Category	Location	Points
1	North of 36.05°N and West of 140.13°E	4
2	North of 36.05°N and East of 140.13°E	3
3	South of 36.05°N and West of 140.13°E	1
4	South of 36.05°N and East of 140.13°E	1

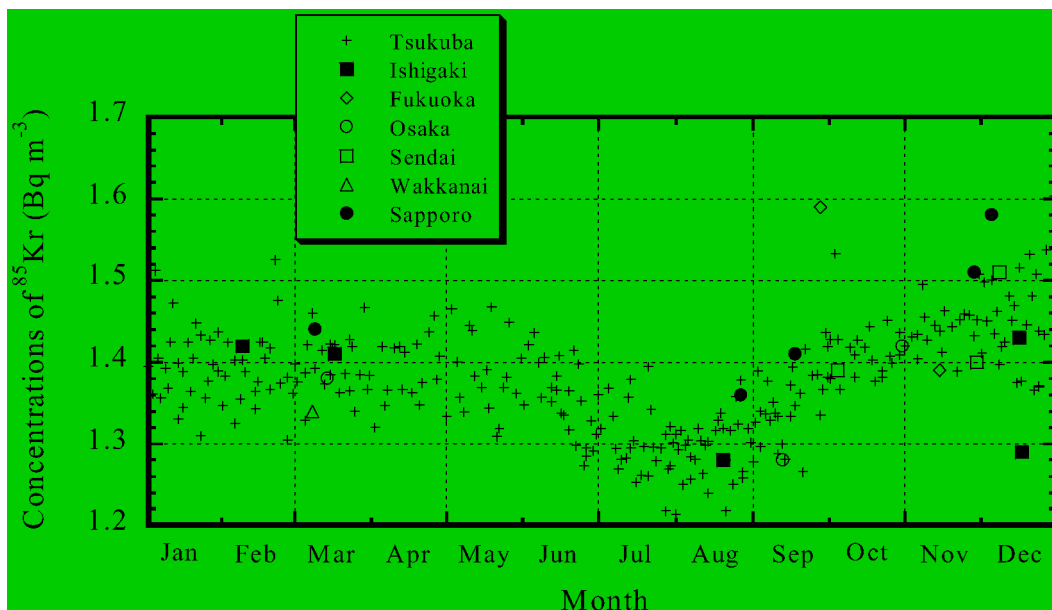


**Fig. 6.** Comparison between the observed  $^{85}\text{Kr}$  concentrations and the estimated ones from the “Trajectory Indexes” in Tsukuba in 1999. The solid line is the observed atmospheric  $^{85}\text{Kr}$  concentrations and the dashed line is the estimated ones calculated from the “Trajectory Indexes” during the sampling period.

As a result, a good correlation between the index and the observed  $^{85}\text{Kr}$  concentrations (correlation factor 0.67) was found. Using a linear regression obtained from this correlation, we estimated  $^{85}\text{Kr}$  concentrations in Tsukuba from the indexes. Figure 6 shows the comparison between the observed  $^{85}\text{Kr}$  concentrations and the estimated ones from the indexes in Tsukuba. The result suggests that the seasonal variation of the atmospheric  $^{85}\text{Kr}$  concentrations in Tsukuba, Japan, is mainly controlled by the transport of the air masses with different origins and that the high concentrations from October to May are attributable to the transport of the continental air mass directly affected by the European sources.

#### *Spatial distribution of the atmospheric $^{85}\text{Kr}$ concentrations in Japan*

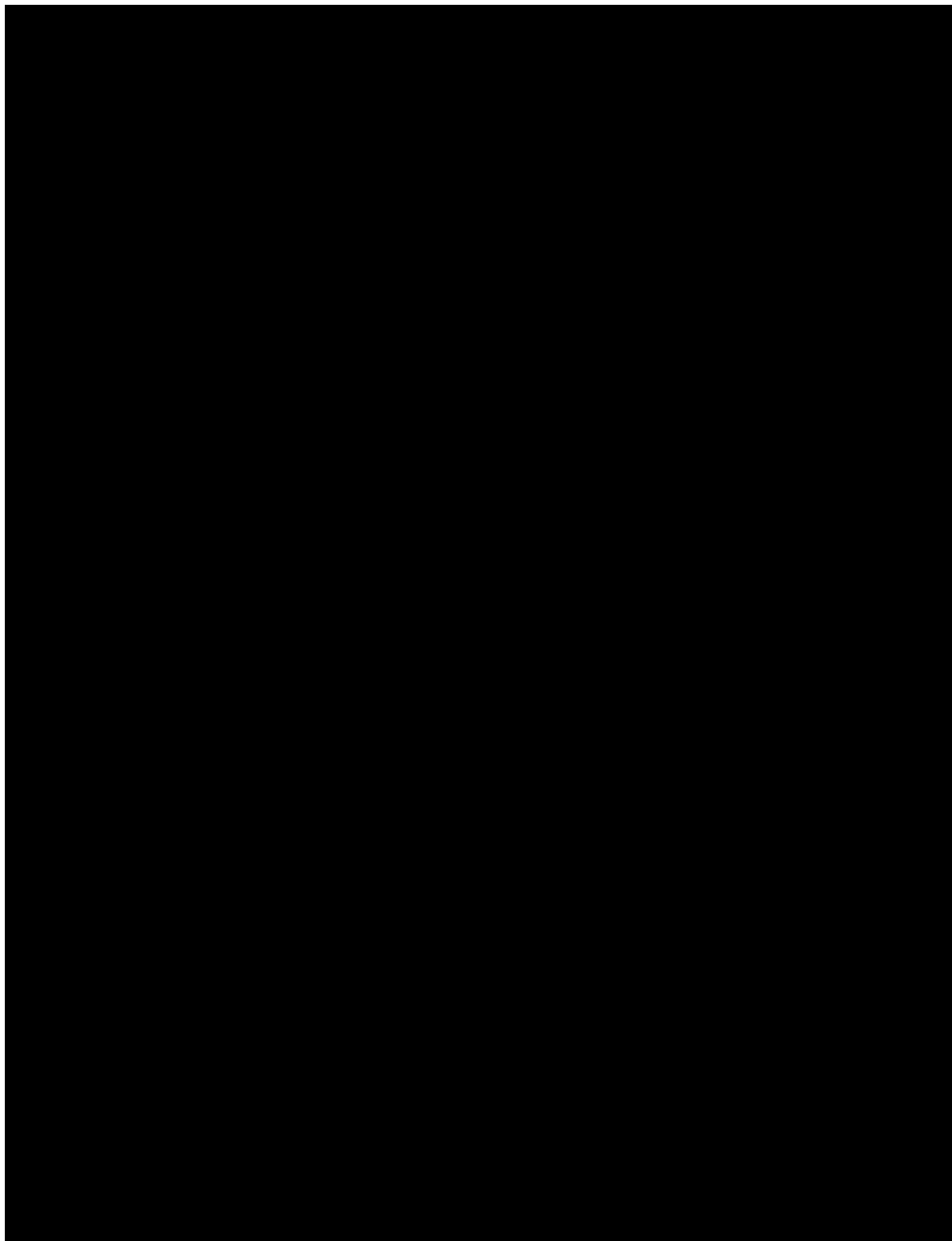
We examined the spatial distribution of the atmospheric  $^{85}\text{Kr}$  concentrations in Japan. However, the data of the atmospheric  $^{85}\text{Kr}$  in the six stations (Table 1), except Tsukuba, are scant. Therefore we introduced normalized  $^{85}\text{Kr}$  concentrations. All the background data observed from 1995 to 2001 at all stations in Japan were normalized to the concentration level of 2001 taken into account as the annual growth rate of  $0.03 \text{ Bq}\cdot\text{m}^{-3}\cdot\text{yr}^{-1}$ ; this enabled us to obtain information about the spatial distribution of the background atmospheric  $^{85}\text{Kr}$  concentrations regardless of the annual variations. Figure 7 shows the seasonal variations of the normalized atmospheric  $^{85}\text{Kr}$  concentrations in Tsukuba together with those observed at the six stations over the Japanese islands. As shown in Fig. 7, the atmospheric  $^{85}\text{Kr}$  concentrations from February to March at the stations of Wak-



**Fig. 7.** Seasonal variation of the atmospheric  $^{85}\text{Kr}$  concentrations at all stations in Japan. Data were obtained for the period from 1995 to 2001. All data were normalized to the concentration levels of 2001 on the assumption of an annual growth rate of  $0.03 \text{ Bq}\cdot\text{m}^{-3}\cdot\text{yr}^{-1}$ .

kanai, Sapporo, Osaka and Ishigaki were at the same levels as those in Tsukuba. On the other hand, a few data obtained in December in Sapporo and Ishigaki seem to be different from those in Tsukuba, which is explained as follows. Since Sapporo is at a higher latitude than Tsukuba, it was covered in early winter by the continental air mass directly affected by European sources. Therefore the  $^{85}\text{Kr}$  concentrations

were higher than in Tsukuba in December. On the other hand, Ishigaki, at a lower latitude than Tsukuba, was still covered by the Pacific air mass with lower  $^{85}\text{Kr}$ ; thus the  $^{85}\text{Kr}$  concentrations in Ishigaki were lower than in Tsukuba. It is suggested that the atmospheric  $^{85}\text{Kr}$  concentrations in early winter in Japan reflect global-scale latitudinal gradient.



**Fig. 8.** (a) Monthly  $^{85}\text{Kr}$  emission at La Hague and Sellafield, compiled by the BfS, and Tokai. (b) Estimation of the atmospheric  $^{85}\text{Kr}$  concentrations in the Northern and Southern Hemispheres by the use of  $^{85}\text{Kr}$  emission data of the nuclear fuel reprocessing plants in Europe. Calculated atmospheric  $^{85}\text{Kr}$  concentrations in the Northern Hemisphere (closed diamonds) and the Southern Hemisphere (open triangles) and observed  $^{85}\text{Kr}$  data in Tsukuba (crosses). The solid lines show the annual average of the observed data in Tsukuba.

*The influence of the European nuclear fuel reprocessing plants on the background level of the atmospheric  $^{85}\text{Kr}$  concentrations in Tsukuba*

Since the current main source of  $^{85}\text{Kr}$  is the nuclear fuel reprocessing plants in Europe, we estimated the atmospheric  $^{85}\text{Kr}$  concentrations in the Northern and Southern Hemispheres by a 2-box model based on the  $^{85}\text{Kr}$  emission data of the nuclear fuel reprocessing plants. From 1995 to 2001, most of the  $^{85}\text{Kr}$  emitted from the nuclear fuel reprocessing plants originated from La Hague in France and Sellafield in the U.K. ( $>300 \text{ PBq}\cdot\text{yr}^{-1}$ ), whereas  $^{85}\text{Kr}$  emitted from the Tokai plant is not significant ( $<13 \text{ PBq}\cdot\text{yr}^{-1}$ ). Therefore we calculated the atmospheric  $^{85}\text{Kr}$  concentrations in the Northern and Southern Hemispheres by using the monthly  $^{85}\text{Kr}$  emission data at La Hague and Sellafield, whose emission data were compiled by the BfS (Fig. 8(a)). To estimate the atmospheric  $^{85}\text{Kr}$  concentrations in the Northern and Southern Hemispheres, we estimated the global  $^{85}\text{Kr}$  inventory by the following equations.

$$\begin{aligned} dA_{n,t} / dt &= E_t - (A_{n,t} - A_{s,t}) / \tau - \lambda A_{n,t} \\ dA_{s,t} / dt &= (A_{n,t} - A_{s,t}) / \tau - \lambda A_{s,t} \end{aligned} \quad (2)$$

In Equation (2),  $t$  is the time as a unit of month,  $A_{n,t}$  and  $A_{s,t}$  (Bq) are the inventories of  $^{85}\text{Kr}$  at  $t$  in the Northern and Southern Hemispheres, respectively;  $E_t$  ( $\text{Bq}\cdot\text{month}^{-1}$ ) is the monthly emission of  $^{85}\text{Kr}$  from the nuclear fuel reprocessing plants in Europe at  $t$ ,  $\tau$  (month) is the interhemispheric exchange time, and  $\lambda$  ( $\text{month}^{-1}$ ) is the radioactive decay constant of  $^{85}\text{Kr}$ . In the calculation, we set January 1994 as an initial time. Equation (2) was calculated by use of the method of finite differences with a time step of a month. The initial value of  $A_{n,t}$  was calculated from the global air mass ( $5.137 \text{ Ekg}^{18}$ ), density of surface air ( $1.2250 \text{ kg}\cdot\text{m}^{-3}$ , U.S. Standard Atmosphere 1976), and the concentration of  $^{85}\text{Kr}$  in Tsukuba in January 1994. It was assumed, however, that  $^{85}\text{Kr}$  existed only in the troposphere, which contained 80% of the global air mass. The concentration of  $^{85}\text{Kr}$  in January 1994 was extrapolated from the annual average concentration of  $^{85}\text{Kr}$  in 1998; in doing this, we took, into account the mean annual growth rate. This extrapolation was considered reasonable because the emission increased steadily from 1994 to 1998, as shown in Fig. 8(a). In this case, the annual average concentration of the atmospheric  $^{85}\text{Kr}$  in 1998 was used for two reasons: The  $^{85}\text{Kr}$  concentration in Tsukuba had a clear seasonal variation, and there was no release of  $^{85}\text{Kr}$  from the Tokai nuclear fuel reprocessing plant throughout this year. Thus the concentration of  $^{85}\text{Kr}$  in the Northern Hemisphere on January 1994 was estimated to be  $1.15 \text{ Bq}\cdot\text{m}^{-3}$ , which was considered approximately reasonable because Pollard *et al.* (1997)<sup>7</sup> reported that the mean annual  $^{85}\text{Kr}$  concentration in Dublin, Ireland, was  $1.12 \text{ Bq}\cdot\text{m}^{-3}$  during 1994. The initial value of  $0.92 \text{ Bq}\cdot\text{m}^{-3}$  for  $A_{s,t}$  was determined from the observed data during the first months in 1994 at the Georg von Neumeyer station in the Antarctica,

at Cape Grim and at Cape Point by the BfS. As the inter-hemispheric exchange time constant,  $\tau$ , 1.6 years (19.2 months, which was obtained by Levin and Hesshaimer<sup>19</sup>) was used.

Figure 8(b) shows calculated atmospheric  $^{85}\text{Kr}$  concentrations in the Northern Hemisphere (closed diamonds) and the Southern Hemisphere (open triangles) and observed  $^{85}\text{Kr}$  data in Tsukuba (crosses). The solid lines show the annual average of the observed data in Tsukuba. The estimated concentrations of  $^{85}\text{Kr}$  in the Northern Hemisphere were in good agreement with the annual averages of the atmospheric  $^{85}\text{Kr}$  observed in Tsukuba. It is concluded, therefore, that the background atmospheric  $^{85}\text{Kr}$  concentrations in Tsukuba are controlled by the emissions of the European nuclear fuel reprocessing plants and the variation of air mass transportation. After 1999, the increasing rate was lowered, which corresponded to the lower shift of the emission rate in Europe (Fig. 8(a)).

Furthermore, we estimated the global  $^{85}\text{Kr}$  inventory up to December 2001 to be about 5 EBq. If the global atmospheric  $^{85}\text{Kr}$  concentrations and its inventory are to be known from now, long-term observation and a global monitoring network should be maintained.

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