Available online at www.sciencedirect.com
SCIENCE DIRECT
ELSEVIER
Journal of Environmental Radioactivity 80 (2005) 183–215
JOURNAL OF ENVIRONMENTAL RADIOACTIVITY
www.elsevier.com/locate/jenvrad

A new compilation of the atmospheric ^{85}krypton inventories from 1945 to 2000 and its evaluation in a global transport model

K. Winger[a,*], J. Feichter[a], M.B. Kalinowski[b,1,2], H. Sartorius[c], C. Schlosser[c]

[a] *Max-Planck-Institut für Meteorologie, Bundesstraße 55, D-20146 Hamburg, Germany*
[b] *IANUS, Darmstadt University of Technology, Hochschulstr. 4a, D-64289 Darmstadt, Germany*
[c] *Bundesamt für Strahlenschutz (BfS), P.O. Box 10 01 49, D-38201 Salzgitter, Germany*

Received 24 September 2003; received in revised form 2 September 2004; accepted 29 September 2004

Abstract

This paper gives the yearly ^{85}Kr emissions of all known reprocessing facilities, which are the main sources of ^{85}Kr in the atmosphere since 1945, for the years 1945 until 2000. According to this inventory 10,600 PBq (Peta = 10^{15}) of ^{85}Kr have been globally emitted from the year 1945 until the end of 2000. The global atmospheric inventory at the end of the year 2000 amounts to 4800 PBq. These emissions have been incorporated into the ECHAM4 atmospheric general circulation model as point sources. Monthly mean model results are compared with measurements made at different locations and times. The influence of each source on the measured concentrations at various locations is studied. The calculated concentrations are found to give reasonably good agreement with the observations, indicating that the emission inventory is realistic. Although, at all northern hemispheric observation sites the model tends to slightly overestimate the concentrations. A possible reason for this

* Corresponding author. Ouranos, 550 Sherbrooke West, 19th floor, Tour West, Montréal, Québec, H3A 1B9, Canada. Fax: +1 514 288 7131.
E-mail address: katja.winger@ec.gc.ca (K. Winger).

[1] Current address: Provisional Technical Secretariat of the CTBTO Preparatory Commission, P.O. Box 1250, A-1400, Vienna, Austria.

[2] The views expressed herein are those of the authors and do not necessarily reflect the views of the CTBTO Preparatory Commission.

0265-931X/$ - see front matter
doi:10.1016/j.jenvrad.2004.09.005

overestimation can be found in model features (coarse resolution in time and space). The most prominent discrepancy that is consistently repeated at all northern hemispheric stations occurs in the early 1990s. This could most likely be related to an overestimate of sources. Possibly, the Russian emissions declined earlier than assumed in the current database. Another discrepancy between observations and simulations indicating an incompleteness of the release data is found at some southern hemispheric sites. The variability of their observations could only be explained by regional sources. However, several spikes occur after 1992 when no reprocessing facility is known to be in operation in the southern hemisphere. Production of isotopes for radiopharmaceuticals like technetium-99m from highly enriched uranium is the most likely explanation.

Keywords: Krypton-85; Atmospheric global transport; Wide-area environmental monitoring; Nuclear fuel reprocessing; Interhemispheric exchange time

1. Introduction

^{85}Kr is a radioactive noble gas with a half-life of 10.76 years. The major sources for atmospheric ^{85}Kr are reprocessing facilities for nuclear fuel, which are mainly located in the northern hemisphere. The relatively well known distribution of the sources and the fact that the only significant sink in the atmosphere is natural decay, which makes this tracer ideal to assess the characteristics of the large-scale horizontal and the interhemispheric transport as depicted in numerical atmospheric circulation models. For instance, Jacob et al. (1987) and Zimmermann et al. (1988) simulated the global ^{85}Kr distribution using a three-dimensional tropospheric model while Rath (1988) used a two-dimensional model.

Although the solubility of ^{85}Kr (as all other inert gases) in water is very low (the ratio of ^{85}Kr to water is 1.85×10^{-10} g/g at equilibrium) and the gas exchange rates of atmosphere–ocean and surface and deep water indicate that the ocean dissolves not more than 0.1 percent of the annual input of ^{85}Kr (Izrael et al., 1982), ^{85}Kr is also a very useful tracer for relatively short term (decadal) atmosphere–ocean exchanges and mixing, for the dating of ground water, and it can be used to determine the age distribution of the water as described by Loosli (1992). The ^{85}Kr distribution in the atmosphere can also be used as an indicator for clandestine separation of plutonium for building nuclear weapons.

The basis for these studies is the knowledge of the atmospheric ^{85}Kr input function. The first comprehensive emission inventory of ^{85}Kr was published by von Hippel et al. (1986) and expanded by Rath (1988). However, these inventories lack some major sources, especially those located in Russia, and they only include the period from 1945 to 1986.

In this paper, an atmospheric ^{85}Kr emission database has been compiled for all known nuclear fuel reprocessing facilities (RFs) from 1945 until 2000. To verify these emissions, a global three-dimensional tropospheric and stratospheric transport model is applied to this database and the results are validated with global measurements.

Measurements of atmospheric concentration of ^{85}Kr at the surface have been made for example by Różański (1979), Achkasov et al. (1991), Weiss et al. (1992), and Igarashi et al. (2001).

2. Major sources of atmospheric ^{85}Kr

Natural sources feed a stable equilibrium content of 0.09 PBq (Peta = 10^{15}) of ^{85}Kr in the atmosphere (Schröter and Roether, 1975). Anthropogenic sources generate an atmospheric inventory that is at least 4 orders of magnitude larger. The main amount of ^{85}Kr in the atmosphere is emitted by nuclear fuel reprocessing facilities during the extraction of plutonium for either civilian or military purposes. ^{85}Kr is produced as a fission product predominantly by neutron induced fission of ^{235}U and ^{239}Pu in nuclear power plants, in research reactors or in dedicated plutonium production reactors. Practically all the ^{85}Kr remains in the fuel rods. As a rule, irradiated rods are stored for half a year or more before they are reprocessed to reduce the activity of the fission products. At RFs, the spent fuel rods are first cut into smaller pieces and then dissolved whereby all ^{85}Kr is released into the atmosphere.

Krypton can technically be held back, but this has not yet been accomplished on a large scale basis for economic reasons. Only small amounts of ^{85}Kr are retained for technical applications and research.

Close to 100% of the ^{85}Kr released at reprocessing plants is emitted in the gaseous phase and only a negligible amount is contained in the aqueous effluents. The amount of activity released depends not only on the reprocessing system and the specific gas treatment of the reprocessing plant, but also on the type of fuel reprocessed, its irradiation history and cooling time. Approximately 130–1800 TBq of ^{85}Kr and 6–10 kg of plutonium are produced per ton of spent fuel (Kalinowski, 1997). Estimates for the ^{85}Kr yield from the reprocessing of 1 kg plutonium range from 10 to 35 TBq (Kalinowski et al., 2004).

Historic nuclear weapon tests in the atmosphere, in water and below ground contributed another significant source of ^{85}Kr. Most atmospheric nuclear tests took place before 1963 and the last one was conducted in 1980. The contribution from underground tests is very small. The last took place in 1998. An assessment by Telegades and Ferber (1975) indicates that less than 2 percent of the total ^{85}Kr world inventory in 1973 could be attributed to nuclear weapon tests conducted up to that time. Their contribution is estimated to have produced 111–185 PBq (Izrael et al., 1982; Hilbert, 1975). In 2000, 13–20 PBq of this amount still remains in the atmosphere. Finally, ^{85}Kr can also be emitted through accidents at nuclear power stations. The largest incident was the explosion in Chernobyl in 1986, which set about 35 PBq of ^{85}Kr free (Bradley, 1997).

3. The emission database

The ^{85}Kr emission database compiled in this paper contains only ^{85}Kr emitted by RFs. The RFs are mainly located in the northern hemisphere between 30°N and

60°N, most of them in Europe. In tropical latitudes, only three (small) sources exist, all of them in India. In the southern hemisphere only two (very small) sources are known, namely in Argentina and South Africa (Fig. 1).

Information on ^{85}Kr release rates has been, and in some cases still is, classified information, especially regarding military facilities. Emission estimated for time periods shorter than one year are rarely available. La Hague is the only RF that reports monthly emissions since 1994 (see Fig. 3). Besides of La Hague, all data are annual data. None of the annual data was modulated by an assumed seasonal variation. Annual ^{85}Kr emission amounts of all known RFs are given in Table 1.

^{85}Kr emissions are estimated using the most accurate available data. One of the following bases is used which are ordered by increasing uncertainty: (a) measurements taken at the exhaust stack (assumed accuracy $\pm$ 1%); (b) the amount of reprocessed plutonium ($\pm$10%); (c) power produced by the nuclear reactors ($\pm$10%); (d) comparison between global emitted ^{85}Kr and measurements, the discrepancy is ascribed to the Russian facilities ($\pm$20%); and (e) operation time and capacity of the facility (could be by a factor of 10 bigger or smaller). An f is used in the table for entries with unknown uncertainty.

Detailed information about the origin of the emission data for the respective RFs can be found in Table 2. In these estimates it is always assumed that no ^{85}Kr is

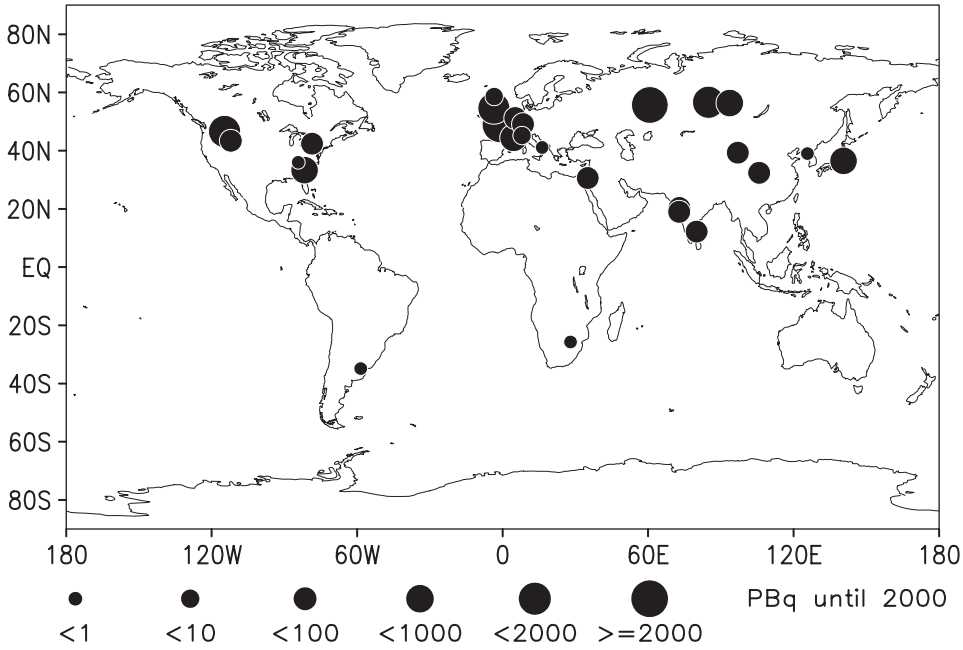


Fig. 1. Sites of reprocessing facilities operating between 1945 until 2000. The total amount of ^{85}Kr emitted until 2000 is shown by the size of the circles.

Table 1
Yearly ^{85}Kr emissions of reprocessing facilities in PBq per year

Country	La Hague, France	Chelyabinsk, Russia	Tomsk, Russia	Krasnoyarsk, Russia	Sellafield, UK	Dounray, UK	Hanford, USA
Latitude	49.0	55.7	56.63	56.37	54.6	58.6	46.6
Longitude	−0.9	60.7	84.92	93.68	−3.6	−3.5	−114.7
Tracer #	1, 12–15	2	2	2	3	3	4
1945	0.00	0.00	0.00	0.00	0.00	0.00	0.30
1946	0.00	0.00	0.00	0.00	0.00	0.00	1.04
1947	0.00	0.00	0.00	0.00	0.00	0.00	2.66
1948	0.00	0.00	0.00	0.00	0.00	0.00	2.00
1949	0.00	0.00	0.00	0.00	0.00	0.00	2.55
1950	0.00	6.00	0.00	0.00	0.00	0.00	3.22
1951	0.00	9.00	0.00	0.00	0.00	0.00	4.26
1952	0.00	4.00	0.00	0.00	0.41	0.00	6.92
1953	0.00	5.00	0.00	0.00	0.85	0.00	13.80
1954	0.00	5.00	0.00	0.00	0.97	0.00	17.24
1955	0.00	10.95	0.05	0.00	1.04	0.00	20.72
1956	0.00	18.25	0.75	0.00	1.20	0.00	31.04
1957	0.00	14.22	0.78	0.00	1.36	0.00	34.52
1958	0.00	8.43	1.05	0.52	2.05	0.00	46.92
1959	0.00	20.62	4.64	4.75	2.27	0.00	40.03
1960	0.00	3.66	3.47	19.87	4.60	0.00	58.65
1961	0.00	8.64	2.21	2.16	6.46	0.00	65.56
1962	0.00	1.25	0.34	0.41	6.77	0.00	59.35
1963	0.00	32.08	12.90	12.02	4.84	0.00	62.12
1964	0.00	25.57	10.94	11.49	9.22	0.00	67.64
1965	0.45	36.95	20.69	19.36	11.67	0.00	58.65
1966	1.02	39.14	22.39	22.47	16.64	0.00	52.13
1967	1.47	26.73	18.26	16.02	17.39	0.00	59.20
1968	4.10	30.67	20.95	18.38	28.91	0.00	41.29
1969	2.90	23.14	16.99	13.87	33.86	0.00	33.60
1970	4.90	34.21	21.36	17.43	36.36	0.00	37.78
1971	6.00	49.63	28.84	23.53	39.53	0.00	17.80
1972	8.88	45.80	24.89	20.31	41.87	0.00	0.00
1973	8.51	49.82	27.08	22.09	34.09	0.00	0.00
1974	26.64	49.45	26.88	21.93	25.37	0.00	0.00
1975	24.42	113.79	61.85	50.46	38.69	0.00	0.00
1976	12.95	86.68	47.12	38.44	35.05	0.07	0.00
1977	24.75	53.85	29.27	23.88	28.46	0.03	0.00
1978	29.08	54.86	29.82	24.33	25.90	0.05	0.00
1979	23.75	65.93	35.84	29.24	35.00	0.05	0.00
1980	30.53	56.87	30.91	25.22	31.00	0.11	0.00
1981	35.85	88.45	48.08	39.22	52.00	0.00	0.00
1982	45.14	89.42	48.60	39.65	44.00	0.00	0.00
1983	50.17	76.42	41.54	33.89	41.80	0.00	0.00
1984	27.01	70.69	38.42	31.35	37.10	0.00	12.14
1985	70.30	65.00	49.77	40.61	23.80	0.00	5.72
1986	64.30	53.27	40.79	33.28	53.30	0.00	12.31
1987	79.50	53.71	35.64	29.07	34.00	0.00	18.17
1988	61.60	64.78	40.70	33.20	39.70	0.00	6.07

(*continued on next page*)

(Table 1 *continued*)

Country	La Hague, France	Chelyabinsk, Russia	Tomsk, Russia	Krasnoyarsk, Russia	Sellafield, UK	Dounray, UK	Hanford, USA
Latitude	49.0	55.7	56.63	56.37	54.6	58.6	46.6
Longitude	−0.9	60.7	84.92	93.68	−3.6	−3.5	−114.7
Tracer #	1, 12–15	2	2	2	3	3	4
1989	42.00	63.65	39.45	32.19	51.70	0.57	0.00
1990	63.00	40.00	28.55	22.45	37.60	0.57	0.00
1991	100.00	70.68	44.54	40.33	44.60	0.57	0.00
1992	95.20	66.03	36.85	18.84	27.40	0.57	0.00
1993	115.00	101.57	42.64	28.87	57.00	0.00	0.00
1994	180.00	106.49	48.98	30.26	38.20	0.00	0.00
1995	230.00	60.00	30.00	20.00	97.00	0.00	0.00
1996	259.00	30.00	20.00	15.00	100.00	0.00	0.00
1997	298.00	0.00	13.00	10.00	95.00	0.00	0.00
1998	319.00	0.00	13.00	10.00	99.00	0.00	0.00
1999	295.00	0.00	13.00	10.00	100.00	0.00	0.00
2000	233.00	0.00	13.00	10.00	95.00	0.00	0.00

Country	Idaho, USA	Savannah, USA	West Valley, USA	Oak Ridge, USA	Mol, Belgium	Karlsruhe, Germany	Saluggia, Italy
Latitude	43.4	33.3	42.4	36.0	51.2	49.1	45.2
Longitude	−112.1	−81.7	−78.6	−84.3	5.2	8.4	8.0
Tracer #	4	5	5	5	6	6	6
1945	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1946	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1947	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1948	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1949	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1950	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1951	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1952	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1953	0.56	0.00	0.00	0.00	0.00	0.00	0.00
1954	1.37	0.00	0.00	0.00	0.00	0.00	0.00
1955	1.92	4.07	0.00	0.00	0.00	0.00	0.00
1956	3.07	7.03	0.00	0.00	0.00	0.00	0.00
1957	2.78	16.98	0.00	0.00	0.00	0.00	0.00
1958	2.78	25.39	0.00	0.00	0.00	0.00	0.00
1959	2.78	28.71	0.00	0.00	0.00	0.00	0.00
1960	2.78	40.30	0.00	0.00	0.00	0.00	0.00
1961	2.78	43.47	0.00	0.00	0.00	0.00	0.00
1962	2.78	44.85	0.00	0.00	0.00	0.00	0.00
1963	2.78	43.47	0.00	0.00	0.00	0.00	0.00
1964	3.14	43.06	0.00	0.00	0.00	0.00	0.00
1965	1.70	44.85	0.00	0.00	0.00	0.00	0.00
1966	1.96	29.53	4.14	0.00	3.70	0.00	0.00
1967	0.78	30.50	4.01	0.00	3.70	0.00	0.00
1968	3.18	36.30	0.00	0.00	3.70	0.00	0.00
1969	4.11	34.64	10.51	0.00	3.70	0.00	0.00
1970	5.48	24.15	1.58	0.00	3.70	0.00	0.00

Table 1 (*continued*)

Country	Idaho, USA	Savannah, USA	West Valley, USA	Oak Ridge, USA	Mol, Belgium	Karlsruhe, Germany	Saluggia, Italy
Latitude	43.4	33.3	42.4	36.0	51.2	49.1	45.2
Longitude	−112.1	−81.7	−78.6	−84.3	5.2	8.4	8.0
Tracer #	4	5	5	5	6	6	6
1971	5.07	20.56	6.04	0.00	4.68	0.60	0.09
1972	1.70	22.20	0.00	0.00	7.40	2.52	0.00
1973	0.19	28.49	0.00	0.00	8.14	0.91	0.17
1974	9.62	18.50	0.00	0.00	3.70	0.03	0.16
1975	0.89	19.24	0.00	0.00	0.00	1.16	0.00
1976	1.22	27.38	0.00	0.00	0.00	3.17	0.00
1977	4.11	16.28	0.00	0.00	0.00	4.25	0.00
1978	3.74	19.61	0.00	0.00	0.00	1.25	0.00
1979	0.00	17.76	0.00	0.00	0.00	1.87	0.00
1980	3.40	21.46	0.00	0.33	0.00	1.20	0.10
1981	2.18	31.0.8	0.00	0.25	0.00	0.11	0.10
1982	0.33	19.06	0.00	0.00	0.00	0.61	0.00
1983	0.11	25.83	0.00	0.00	0.00	2.79	0.10
1984	0.00	25.83	0.00	0.00	0.00	1.16	0.00
1985	0.00	25.90	0.00	0.00	0.00	3.40	0.00
1986	0.00	24.00	0.00	0.00	0.00	3.17	0.00
1987	0.00	12.00	0.00	0.00	0.00	2.71	0.00
1988	0.00	4.00	0.00	0.00	0.00	1.66	0.00
1989	0.00	0.00	0.00	0.00	0.00	0.76	0.00
1990	0.00	0.00	0.00	0.00	0.00	0.94	0.00
1991	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1992	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1993	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1994	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1995	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1996	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1997	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1998	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1999	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2000	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Country	Marcoule, France	Rotondella, Italy	Dimona, Israel	Yongbyon, North Korea	Tokai Mura, Japan	Tarapur, India
Latitude	44.4	41.1	30.6	39.0	36.5	20.3
Longitude	4.5	16.3	35.1	125.7	140.6	72.9
Tracer #	7	7	7	8	8	9
1945	0.00	0.00	0.00	0.00	0.00	0.00
1946	0.00	0.00	0.00	0.00	0.00	0.00
1947	0.00	0.00	0.00	0.00	0.00	0.00
1948	0.00	0.00	0.00	0.00	0.00	0.00
1949	0.00	0.00	0.00	0.00	0.00	0.00
1950	0.00	0.00	0.00	0.00	0.00	0.00
1951	0.00	0.00	0.00	0.00	0.00	0.00
1952	0.00	0.00	0.00	0.00	0.00	0.00

(*continued on next page*)

Table 1 (*continued*)

Country	Marcoule, France	Rotondella, Italy	Dimona, Israel	Yongbyon, North Korea	Tokai Mura, Japan	Tarapur, India
Latitude	44.4	41.1	30.6	39.0	36.5	20.3
Longitude	4.5	16.3	35.1	125.7	140.6	72.9
Tracer #	7	7	7	8	8	9
1953	0.00	0.00	0.00	0.00	0.00	0.00
1954	0.00	0.00	0.00	0.00	0.00	0.00
1955	0.00	0.00	0.00	0.00	0.00	0.00
1956	0.00	0.00	0.00	0.00	0.00	0.00
1957	0.00	0.00	0.00	0.00	0.00	0.00
1958	0.00	0.00	0.00	0.00	0.00	0.00
1959	0.30	0.00	0.00	0.00	0.00	0.00
1960	0.19	0.00	0.00	0.00	0.00	0.00
1961	0.48	0.00	0.00	0.00	0.00	0.00
1962	1.85	0.00	0.00	0.00	0.00	0.00
1963	1.85	0.00	0.00	0.00	0.00	0.00
1964	3.33	0.00	0.00	0.00	0.00	0.00
1965	4.07	0.00	0.23	0.00	0.00	0.00
1966	3.70	0.00	0.23	0.00	0.00	0.00
1967	5.55	0.00	0.23	0.00	0.00	0.00
1968	4.07	0.00	0.23	0.00	0.00	0.00
1969	5.55	0.00	0.23	0.00	0.00	0.00
1970	3.33	0.00	0.23	0.00	0.00	0.00
1971	3.70	0.00	0.23	0.00	0.00	0.00
1972	1.74	0.00	0.23	0.00	0.00	0.00
1973	4.81	0.00	0.23	0.00	0.00	0.00
1974	4.07	0.00	0.23	0.00	0.00	0.00
1975	3.70	0.10	0.23	0.10	0.00	0.00
1976	3.40	0.10	0.41	0.00	0.00	0.00
1977	4.33	0.10	0.41	0.00	0.20	0.00
1978	11.40	0.10	0.41	0.00	2.41	0.00
1979	10.36	0.10	0.41	0.00	0.67	0.23
1980	19.80	0.10	0.41	0.00	6.63	0.23
1981	11.47	0.00	0.41	0.00	6.09	0.23
1982	11.47	0.00	0.41	0.00	9.75	0.23
1983	22.94	0.00	0.41	0.00	0.51	0.23
1984	23.00	0.00	0.41	0.00	0.01	0.15
1985	23.00	0.00	0.41	0.00	11.73	0.15
1986	23.00	0.00	0.41	0.00	8.86	0.15
1987	23.00	0.00	0.41	0.00	10.95	0.15
1988	23.00	0.00	0.41	0.00	7.75	0.15
1989	23.00	0.00	0.41	0.10	4.21	0.15
1990	23.00	0.00	0.41	0.10	15.36	0.15
1991	23.00	0.00	0.41	0.10	14.45	0.23
1992	23.00	0.00	0.41	0.10	12.82	0.23
1993	23.00	0.00	0.41	0.00	5.23	0.23
1994	23.00	0.00	0.41	0.00	13.77	0.23
1995	23.00	0.00	0.41	0.00	12.64	0.23
1996	23.00	0.00	0.41	0.00	9.14	0.23
1997	23.00	0.00	0.41	0.00	2.87	0.23

Table 1 (*continued*)

Country	Marcoule, France	Rotondella, Italy	Dimona, Israel	Yongbyon, North Korea	Tokai Mura, Japan	Tarapur, India
Latitude	44.4	41.1	30.6	39.0	36.5	20.3
Longitude	4.5	16.3	35.1	125.7	140.6	72.9
Tracer #	7	7	7	8	8	9
1998	0.00	0.00	0.41	0.00	0.00	0.23
1999	0.00	0.00	0.41	0.00	0.00	0.23
2000	0.00	0.00	0.41	0.00	0.00	0.23

Country	Kalpakkam, India	Guangyan, China	Subei, China	Ezeiza, Argentina	Trombay, India	Pelindaba, South Africa
Latitude	12.2	32.4	39.3	−34.85	19.5	−25.75
Longitude	80.0	105.8	97.0	−58.53	72.8	28.0
Tracer #	9	10	10	11	16	17
1945	0.00	0.00	0.00	0.00	0.00	0.00
1946	0.00	0.00	0.00	0.00	0.00	0.00
1947	0.00	0.00	0.00	0.00	0.00	0.00
1948	0.00	0.00	0.00	0.00	0.00	0.00
1949	0.00	0.00	0.00	0.00	0.00	0.00
1950	0.00	0.00	0.00	0.00	0.00	0.00
1951	0.00	0.00	0.00	0.00	0.00	0.00
1952	0.00	0.00	0.00	0.00	0.00	0.00
1953	0.00	0.00	0.00	0.00	0.00	0.00
1954	0.00	0.00	0.00	0.00	0.00	0.00
1955	0.00	0.00	0.00	0.00	0.00	0.00
1956	0.00	0.00	0.00	0.00	0.00	0.00
1957	0.00	0.00	0.00	0.00	0.00	0.00
1958	0.00	0.00	0.00	0.00	0.00	0.00
1959	0.00	0.00	0.00	0.00	0.00	0.00
1960	0.00	0.00	0.00	0.00	0.00	0.00
1961	0.00	0.00	0.00	0.00	0.00	0.00
1962	0.00	0.00	0.00	0.00	0.00	0.00
1963	0.00	0.00	0.00	0.00	0.00	0.00
1964	0.00	0.00	0.00	0.00	0.08	0.00
1965	0.00	0.00	0.00	0.00	1.11	0.00
1966	0.00	0.00	0.00	0.00	1.12	0.00
1967	0.00	0.00	0.00	0.00	1.15	0.00
1968	0.00	0.10	0.10	0.00	1.15	0.00
1969	0.00	0.10	0.10	0.00	1.15	0.00
1970	0.00	1.00	3.00	0.00	1.15	0.00
1971	0.00	1.00	3.00	0.00	1.15	0.00
1972	0.00	1.00	3.00	0.00	1.15	0.00
1973	0.00	1.00	3.00	0.00	1.15	0.00
1974	0.00	1.00	3.00	0.00	1.15	0.00
1975	0.00	1.00	3.00	0.00	0.00	0.00
1976	0.00	1.00	3.00	0.00	0.00	0.00
1977	0.00	1.00	3.00	0.00	0.00	0.00
1978	0.00	1.00	3.00	0.00	0.00	0.00
1979	0.00	1.00	3.00	0.00	0.00	0.00

(*continued on next page*)

Table 1 (*continued*)

Country	Kalpakkam, India	Guangyan, China	Subei, China	Ezeiza, Argentina	Trombay, India	Pelindaba, South Africa
Latitude	12.2	32.4	39.3	−34.85	19.5	−25.75
Longitude	80.0	105.8	97.0	−58.53	72.8	28.0
Tracer #	9	10	10	11	16	17
1980	0.00	1.00	3.00	0.00	0.00	0.00
1981	0.00	1.00	3.00	0.00	0.00	0.00
1982	0.00	1.00	3.00	0.00	0.00	0.00
1983	0.00	1.00	3.00	0.00	0.00	0.00
1984	0.00	1.00	3.00	0.00	0.08	0.00
1985	0.00	1.00	3.00	0.00	0.08	0.00
1986	0.00	1.00	3.00	0.00	0.08	0.01
1987	0.00	1.00	3.00	0.00	0.15	0.01
1988	0.00	1.00	3.00	0.00	0.15	0.01
1989	0.00	1.00	3.00	0.10	0.15	0.01
1990	0.00	1.00	3.00	0.10	0.15	0.01
1991	0.00	1.00	3.00	0.00	0.15	0.01
1992	0.00	1.00	3.00	0.00	0.15	0.01
1993	0.00	1.00	3.00	0.00	0.15	0.00
1994	0.00	1.00	3.00	0.00	0.23	0.00
1995	0.00	1.00	3.00	0.00	0.23	0.00
1996	2.00	1.00	3.00	0.00	0.23	0.00
1997	2.00	1.00	3.00	0.00	0.23	0.00
1998	2.00	1.00	3.00	0.00	0.23	0.00
1999	2.00	1.00	3.00	0.00	0.23	0.00
2000	2.00	1.00	3.00	0.00	0.23	0.00

held back and known or assumed storage times are accounted for by decay correction.

Although the emission database was only compiled for ^{85}Kr emitted from RFs, it has to be taken into account that until the end of 1973 the ^{85}Kr ascribed to the Russian sources by Zimmermann et al. (1988) also includes the amount of ^{85}Kr emitted by nuclear weapon tests globally conducted in the respective years.

The global annual ^{85}Kr emission and the annual ^{85}Kr emissions of the major source countries are shown in Fig. 2.

The total atmospheric inventory of ^{85}Kr is calculated as the decay-corrected sum of all emissions in the preceding years. Beginning in 1945 with the first significant anthropogenic emissions, the total amount of ^{85}Kr in the atmosphere increased nearly exponentially up to the sixties (see Fig. 4).

According to this inventory 10,600 PBq of ^{85}Kr have been globally emitted from the year 1945 until the end of 2000. The global ^{85}Kr emissions exhibited a steep increase from 1945 until about 1960 and then stabilized in the following years. Since 1960, the growth rate was almost linear between 50 and 100 PBq per year. Very strong increases (up to 150 PBq per year) occurred in 1975 and 1982 due to high Russian emissions. This was followed first by an almost constantly high annual increase and then by a decrease until 1991. In the early 1990s, emissions increased by

Table 2
References of ^{85}Kr emissions from reprocessing facilities

Facility	Years	Reference	Q[a]	Commentary
Hanford (USA)	45–71	von Hippel et al., 1986	c	^{85}Kr release derived from reactor performance in Tera watt days, 13.8 PBq ^{85}Kr per TWtD. Assumed emission time lag of one year
	72–83	von Hippel et al., 1986		Shut down during 1972, restarted Nov 1983
	84–88	Albright et al., 1997	b	Given Pu production, 19.45 TBq[Kr-85]/kg[Pu] by von Hippel et al., 1986
Idaho (USA)	53–56	US At. En. Com., 1972	f	Cited in von Hippel et al., 1986
	57–63	von Hippel et al., 1986	f	1.85–3.7 PBq per year assumed
	64–72	US At. En. Com., 1972	f	Cited in von Hippel et al., 1986
	73–82	EG&G, 1982	f	Cited in von Hippel et al., 1986
	83	US Dep. Energy, 1984	f	Cited in von Hippel et al., 1986
Savannah (USA)	55	Rath, 1988	f	
	56–70	von Hippel et al., 1986	c	^{85}Kr release derived from reactor performance in Tera watt days, 13.8 PBq ^{85}Kr per TWtD. Assumed emission time lag of one year
	71–81	Ashley and Zeigler, 1980	f	Data compiled by Franke and Alvarez, 1983. Cited in von Hippel et al., 1986
	82–83	von Hippel et al., 1986	c	Like the years 56–70
	84–88	WMO, 1995	f	Emission data received from the USA
West Valley (USA)	66–71	von Hippel et al., 1986	f	Assumed emission time lag of five years
Oak Ridge (USA)	80–81	US Dep. Energy, 1982 and 1982a	f	Cited in Rath, 1988
USSR	50–73	Zimmermann et al., 1988	d	Values based on spot measurements from several North–South Atlantic cruises, aircraft samples over the American continent and the Atlantic, and some balloon flights over Europe, to calculate hemispheric ^{85}Kr mean inventories using a simple two-box model of the atmosphere. The difference between the estimated global ^{85}Kr input and release rates reported from the non-Soviet nuclear fuel reprocessing plants were ascribed to the Russian plants
	74–76	Novichkov	f	Priv. com. to H. Sartorius
	77–80	Kalinowski	c	Pu production in Cochran and Norris, 1993
	81–89	Novichkov	f	Priv. com. to H. Sartorius
	90			Interpolated
	91–94	Novichkov	f	Priv. com. to H. Sartorius
	95–00	Kalinowski	e	Estimations based on information of the operating time of Pu producing facilities from different sources

Rough annual Pu production estimated for individual facilities in Cochran and Norris, 1993. According to these production rates and detailed information for 1992 from M. Kalinowski ^{85}Kr was distributed on three facilities

(*continued on next page*)

Table 2 (*continued*)

Facility	Years	Reference	Q[a]	Commentary
Sellafield (UK)	51–77	Jackson et al., 1998	b	Discharges based on calculated releases
Windscale (UK)	78–83	Jackson et al., 1998	a	Discharges based on measurements
	84–95	Jackson et al., 1998 and BNFL, 1985–96	a	Discharges based on measurements
	96–99	BNFL, 1996–00	a	Discharges based on measurements
	00		e	Extrapolation based on information about the operation of the THORP facility
Dounray (UK)	76, 77	Luykx and Fraser, 1980	f	Cited in Rath, 1988. Data provided by the Department of the Environment
	78, 79			Interpolated
	80	Luykx and Fraser, 1983	f	Cited in Rath, 1988
	89–92	WISE News Com., 1994	e	Maximum values assumed
La Hague (France)	65–93	Albright et al., 1997	b	Pu production, 22.6 PBq[Kr-85]/kg[Pu] assumed
	94–00	Cogema-La Hague	a	Gaseous release monitored by the Ministries of Health and of Labor
Marcoule (France)	59–71	von Hippel et al., 1986	c	^{85}Kr release from estimated tonnage and average burnups from Syndicate CFDT de l'Energie Atomique, 1980. Formula: PBq ^{85}Kr = {[TWtD − 0.98 × (tones Pu fissioned)] × 0.405/0.96 + 0.177 × (tones ^{239}Pu fissioned)} × 37
	72–80	Luykx and Fraser, 1978, 1980, 1983	f	Data provided by the Comité technique interministériel pour l'EURATOM
	81–82	von Hippel et al., 1986	e	^{85}Kr release from estimated tonnage and average burnups
	83	von Hippel et al., 1986	f	According to Nuclear Fuel, December 31, 1984
	84–97	Kalinowski	e	Estimated due to information about facility operation
Mol (Belgium)	66–71	Rath, 1988	f	
	72–76	Luykx and Fraser, 1978	f	Data provided by the Ministére belge de ls Santé publique et de la Famille, Brussel
Karlsruhe (Germany)	71–91	Kernforschungszentrum Karlsruhe, 1971–1991	a	Discharges based on measurements
Saluggia (Italy)	71	Rath, 1988	f	
	72–76	Luykx and Fraser, 1978	f	Data provided by the Comotato Nazionale per l'Energia Nucleare, Rome
	80–83	Gili, 1996	f	Email to M. Kalinowski
Rotondella (Italy)	75–80	Kalinowski	e	Estimated. Date of shutdown (1980) by Lippolis, 1996 in letter to M. Kalinowski
Dimona (Israel)	65–94	Kalinowski	e	Estimated, based on scenario C for Pu production in the Dimona reactor
	95–00		e	Assuming unchanged Pu production
India	64–95	Kalinowski	b	Estimations based on Pu production given in Albright et al., 1997, assuming 15 TBq[Kr-85]/kg[Pu]
	96–00		e	Assuming the same Pu production as in 1995

Table 2 (*continued*)

Facility	Years	Reference	Q[a]	Commentary
Tokai Mura (Japan)	77–98	Japan Nuclear Cycle	a	Data based on measurements at the exhaust stack. Given to H. Sartorius by Y. Igarashi
Current data are available in the www: www.jnc.go.jp/ztokai/kankyo/houkan/torendo_7nichihyouji_a.html; www.jnc.go.jp/ztokai/kankyo/houkan/torendo_24jikanhyouji1_a.html				
Argentina, South Africa		Kalinowski	e	Rough estimations based on information of the operating time of relevant facilities (reactors and reprocessing plants) and their production capacities
China, North Korea		Kalinowski	b	Rough estimations based on Pu production given in Albright et al., 1997, assuming 22 TBq[Kr-85]/kg[Pu]

[a] Q: Quality, accuracy of the data (see Section 3): (a) measurements taken at the exhaust stack (assumed accuracy ± 1%); (b) the amount of reprocessed plutonium (±10%); (c) power produced by the nuclear reactors (±10%); (d) comparison between global emitted ^{85}Kr and measurements, the discrepancy is ascribed to the Russian facilities (±20%); (e) operation time and capacity of the facility (could be by a factor of 10 bigger or smaller); and (f) the certainty is unknown.

about 50% and showed the peak value in 1994. The strongest source between the years 1945 and 1970 was the reprocessing facilities in the U.S., between 1970 and 1990 in the USSR and in the 1990s in France.

For the period from 1955 to 1984, inventory estimates are available from previous studies for comparison. All these data agree with this study within their error estimates. The largest discrepancy is seen in comparison with the values for 1973 and 1974 as given by Styra and Butkus (1990) (see Fig. 4). They assume an increase in annual ^{85}Kr discharges while in fact they decreased. We took over many data as published before, if no more reliable estimates were available. After 1984 all input data are completely new. The current ^{85}Kr inventory is also the first study that gives a separate estimate of the Soviet Union's ^{85}Kr releases since 1974. Original release data or independent estimates are used for these years (see Table 2). Previous studies just derived these releases as the difference between the total inventory increase and the contribution from all known sources without using real input data. The total annual increase was determined from observations using a simple model of the global atmosphere. In this study the values derived by Zimmermann et al. (1988) were used as input data for the Russian sources up to 1973.

Table 2 shows in detail the database used for the current estimate.

4. Model description

The global model ECHAM4 used for this study was developed from the weather forecast model of the European Centre for Medium-Range Weather Forecasts (ECMWF). It is a spectral model based on the primitive equations. The prognostic variables are the logarithm of surface pressure, temperature, vorticity and divergence of the horizontal wind, water vapor, and liquid water mixing ratio. In addition,

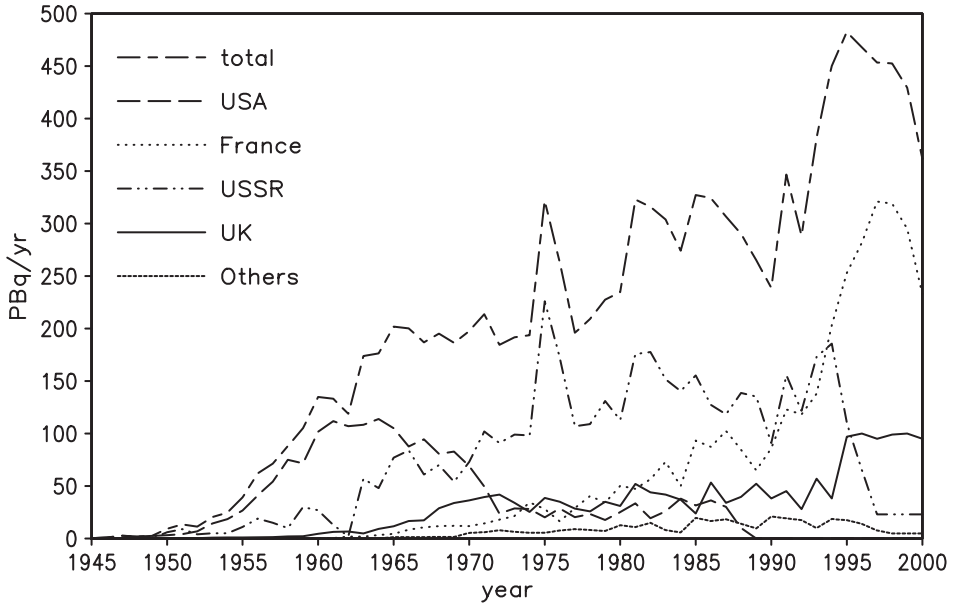


Fig. 2. Annual ^{85}Kr emissions of major source countries and global emissions.

a number of passive tracers can be transported. Water vapor, cloud water, and tracers (like ^{85}Kr) are advected by a multi-dimensional flux form (in principle conservative) scheme that is shape preserving called SPlit Implementation of Transport using Flux Integral REpresentations (Spitfire) (Rasch and Lawrence, 1998). The other prognostic variables of ECHAM are transported in the spectral space. A horizontal spectral resolution of T30 is used for the current study. T30

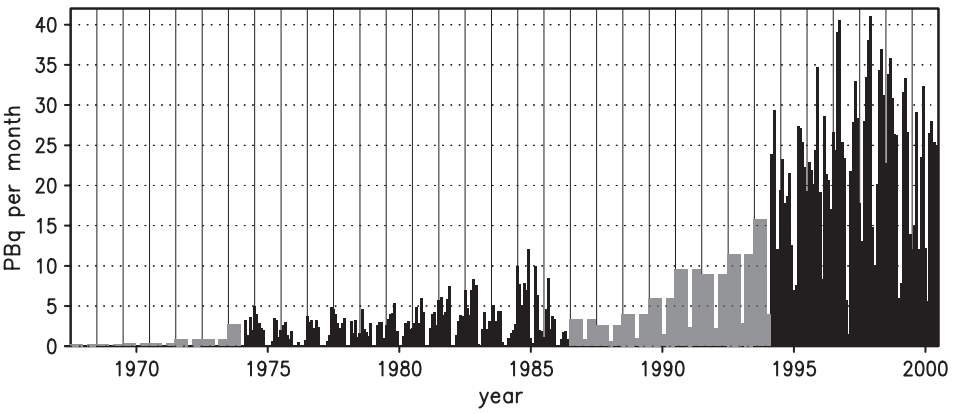


Fig. 3. Monthly ^{85}Kr emissions from La Hague (real values – black; calculated values from yearly sum – grey).

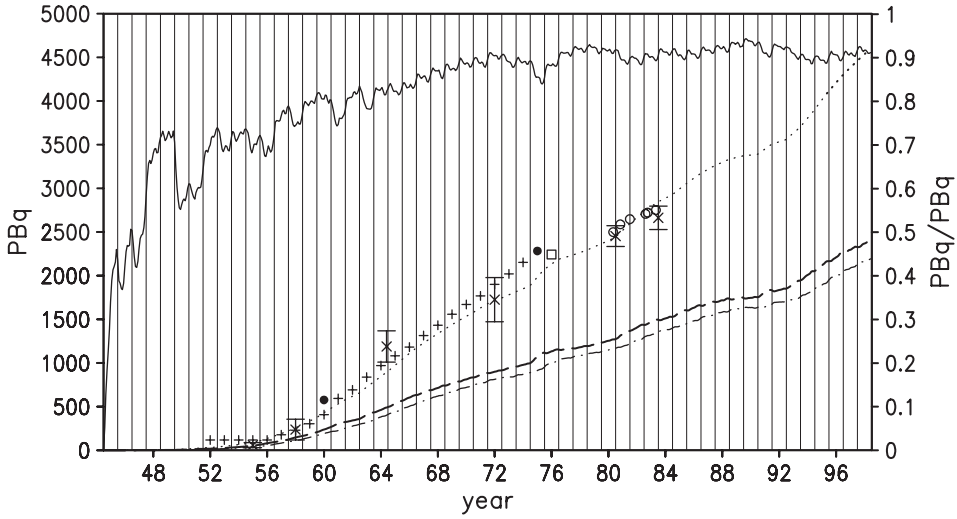


Fig. 4. Temporal development of the global and hemispheric ^{85}Kr inventories (global – dots, NH – long dash, SH – dot dash), left scale and the ratio of the SH and NH inventories (solid) right scale. Inventories calculated by von Hippel et al. (1986) (X with error bars), Jacob et al. (1987) (open circles), Sittkus and Stockburger (1976) (closed circles), Różański (1979) (square), and Styra and Butkus (1990) (crosses).

refers to a triangular truncation with the maximum total wavenumber on the sphere being 30. A triangular truncation is one for which the zonal wavenumber, for a given total wavenumber n, has values between $-n$ and $+n$ inclusively. The semi-implicit leap-frog time stepping scheme employs a time step of 30 min. The model includes a state-of-the-art parameterization package for unresolved dynamical and physical processes, including radiation, cumulus convection, stratiform clouds, gravity wave drag, vertical turbulent diffusion and surface fluxes, land surface processes, and horizontal diffusion. The advection scheme and the parameterized physical processes are calculated on the associated Gaussian transform grid of approximately $3.75^\circ \times 3.75^\circ$ in latitude and longitude. Which is about 417 km $\times$ 417 km at the equator and 273 km $\times$ 417 km at the latitude of La Hague. The characteristics of the ECHAM4 model are described comprehensively by Roeckner et al. (1996). ECHAM4 features that were adopted from previous ECHAM versions or from the original ECMWF forecast model can be found in Roeckner et al. (1992) and Simmons et al. (1989). To simulate weather conditions that are representative for specific years, the monthly mean sea surface temperatures of the specific years are prescribed, as analyzed from observations by the Hadley Centre (Rayner et al., 1996).

The ^{85}Kr emissions have been introduced into the ECHAM4 atmospheric general circulation model, which is equipped with 23 ^{85}Kr tracer species to distinguish the influence of individual source regions on the simulated ^{85}Kr concentrations. The simulation starts in 1945 and ends in 1998.

Up to three of the 26 known ^{85}Kr sources were combined to one tracer in order to avoid excessive memory and CPU use (see row 'Tracer #' in Table 1). To study the

different influences of constant ^{85}Kr emissions over a period of one year versus monthly varying releases, four tracers were added for the RF in La Hague.

4.1. Model results

By comparing the increase of the northern hemisphere (NH) inventory to the one of the southern hemisphere (SH), one can derive the characteristic time scale of the interhemispheric exchange in the model (see Fig. 4). Looking at the strong increase of the ^{85}Kr emissions in 1975 which caused a strong increase of the inventory, one can see that the increase of the hemispheric inventory shows a time-lag of nine months between the NH and the SH. This value is at the lower end of previous estimates (e.g. 1.0 year, Rath, 1988; 1.1 years, Jacob et al., 1987; 1.6 years, Weiss et al., 1992). Low characteristic exchange times indicate a fast cross-equator transport. However, this is in contradiction to the findings shown in Fig. 7. Fig. 7 shows the observed and the calculated ^{85}Kr concentrations taken at a ship-cruise over the Atlantic. Calculated concentrations in the SH agree very well with the observed ones, but concentrations in the NH are over predicted. This indicates a too slow cross-equator transport. Interhemispheric exchange times reported in literature have been calculated by chemistry transport models which use 12- or 6-hourly meteorological input whereas a general circulation model (GCM) as used in the current study creates meteorological fields every 30 min. Because the relevant transport processes across the equator are small-scale the temporal resolution of the driving meteorology may have an impact on the exchange time.

Fig. 5 shows the ^{85}Kr concentration at ground level, in January and July 1987, a year in which a comparatively large number of RFs were operational and, hence, this was a year with many active sources. The sources in the NH are easily recognized by plumes of higher concentrations. The distribution in the SH on the other hand is remarkably homogeneous. Differences between the winter and the summer months primarily reflect the seasonality of transport, but they also include a small increase in the global inventory of 1.47% between January and July.

In January the plumes are transported much more eastward and pole ward than in July because of the stronger winds and the lack of vertical exchange due to convective activity. One can also discern the East Asian winter monsoon transporting ^{85}Kr from the eastern Russian sources south east towards Japan. At the Himalayas and the Rocky Mountain region, there is always a local minimum because of the high altitude of the surface level.

The characteristic of the vertical structure of the ^{85}Kr distribution is shown by the zonal mean cross-section in Fig. 6, again for January and July 1987. It also shows the relatively uniform distribution in the SH and strong gradients in the NH. The ^{85}Kr transport into the SH occurs mainly in the upper troposphere. As a consequence, an inverse vertical gradient in the SH can be observed. This shape of the vertical distribution is in very good agreement with observations of other long-lived species, which are mainly emitted at ground level at the NH, like carbon dioxide, carbon monoxide and methane (Avery et al., 2001).

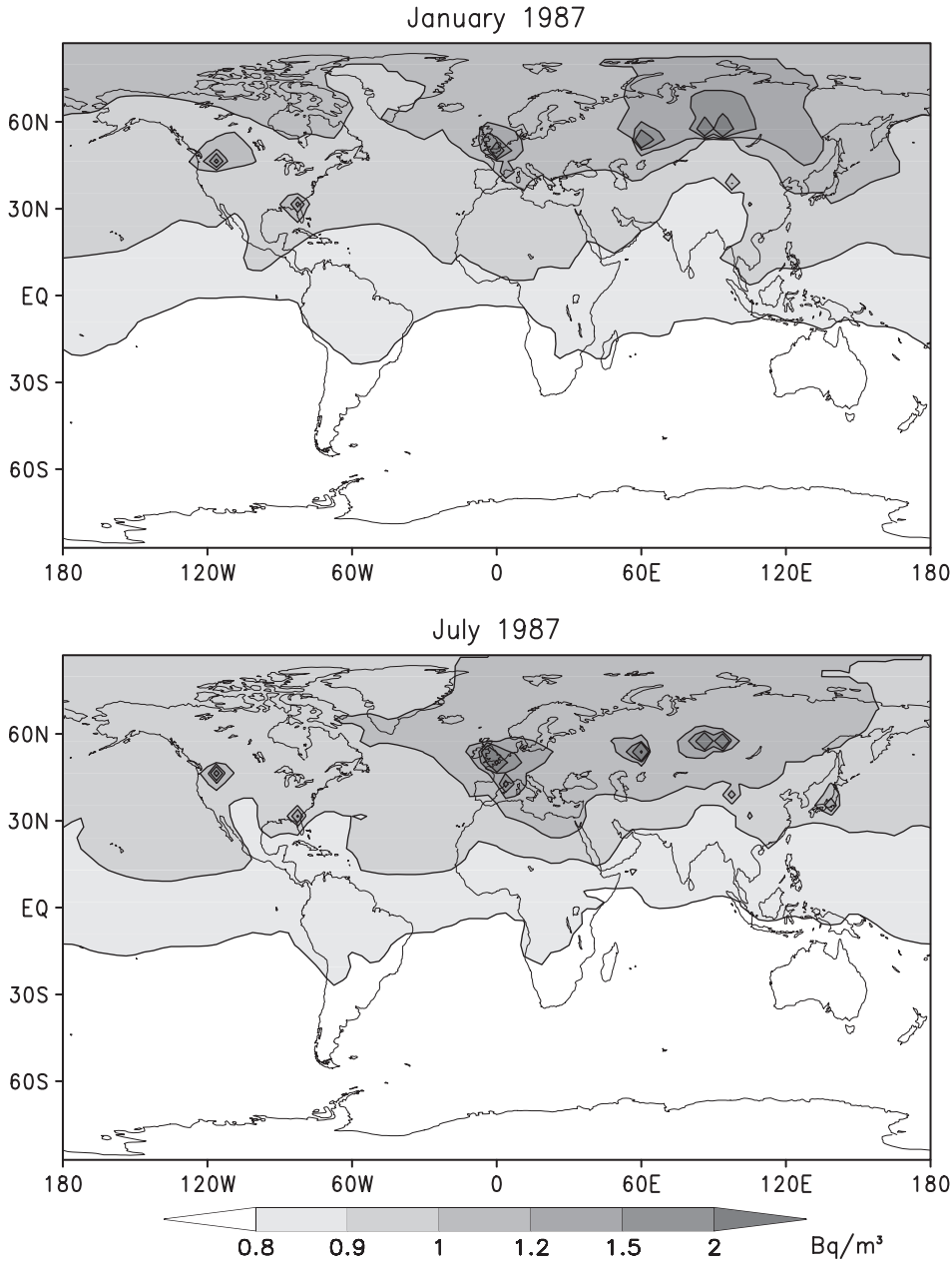


Fig. 5. ^{85}Kr monthly mean surface distribution. (a) January 1987, (b) July 1987.

In the lower troposphere the vertical concentration gradients in the NH are steeper in January than in July due to the weaker vertical exchange. In January there is more transport towards the north pole. In July enhanced convective activity over the continents increases the vertical mixing.

5. Evaluation

The measurements used in this paper for the validation of simulated concentrations were made by the Bundesamt für Strahlenschutz (BfS) in Freiburg, Germany. The ^{86}Kr activity concentrations data at the station Tsukuba, Japan are the result of a collaboration between the Meteorological Research Institute in Tsukuba and the BfS in Freiburg (Igarashi et al., 2000, 2001). The required precision (about 1%) is achieved through a high degree of standardization for all the enrichment and purification steps (Weiss et al., 1992).

5.1. Latitudinal surface distribution

Latitudinal surface distribution of ^{85}Kr concentration was measured during several Atlantic ship-cruises between 1980 and 1985. The observed data from eight of these sampling cruises are shown in Fig. 7 together with simulated data. The observations of all sampling cruises show a similar pattern of the latitudinal distribution and a global increase with time. In the SH, changes in the concentrations are in the order of the accuracy of the measurements (about $\pm 1\%$) whereas the NH is characterized by a strong spatial variability. Two maxima are observed indicating latitudinal bands with high emission sources; one between 20°N and 30°N and another stronger one between 45°N and 60°N. The highest concentration is observed at the same latitude as the main active RFs during this time: Sellafield, La Hague, and the three Russian facilities. Another predominant feature is the pronounced drop in concentration (about 15%) at the Intertropical Convergence Zone (ITCZ).

The simulated concentrations show a similar latitudinal gradient as the observations, but the model overestimates the concentrations, particularly in the NH (up to 20%). The agreement in the SH is very good until the year 1983. This indicates that the models interhemispheric exchange may be too weak. After 1983 the good agreement of observations and model results in the SH deteriorates. There is no conclusive evidence whether it could be due to an underestimation of the SH sources or due to a problem with interhemispheric transport simulation. However, since changes in the SH follow those in the NH with a delay of about one year, and the overestimate in the NH increases after 1983, the bias in the SH may be caused by an overestimate of NH sources in the mid 1980s.

The too high concentrations between 50°N and 55°N are due to the coarse resolution of the model. When the observation point is located in the same grid box as a RF, as in sampling cruise (e) and (f), the simulated concentration is much higher than the observed. Even when a sample is taken in a grid box just beside one containing a RF, the simulated concentration is still too high, like at sampling cruise

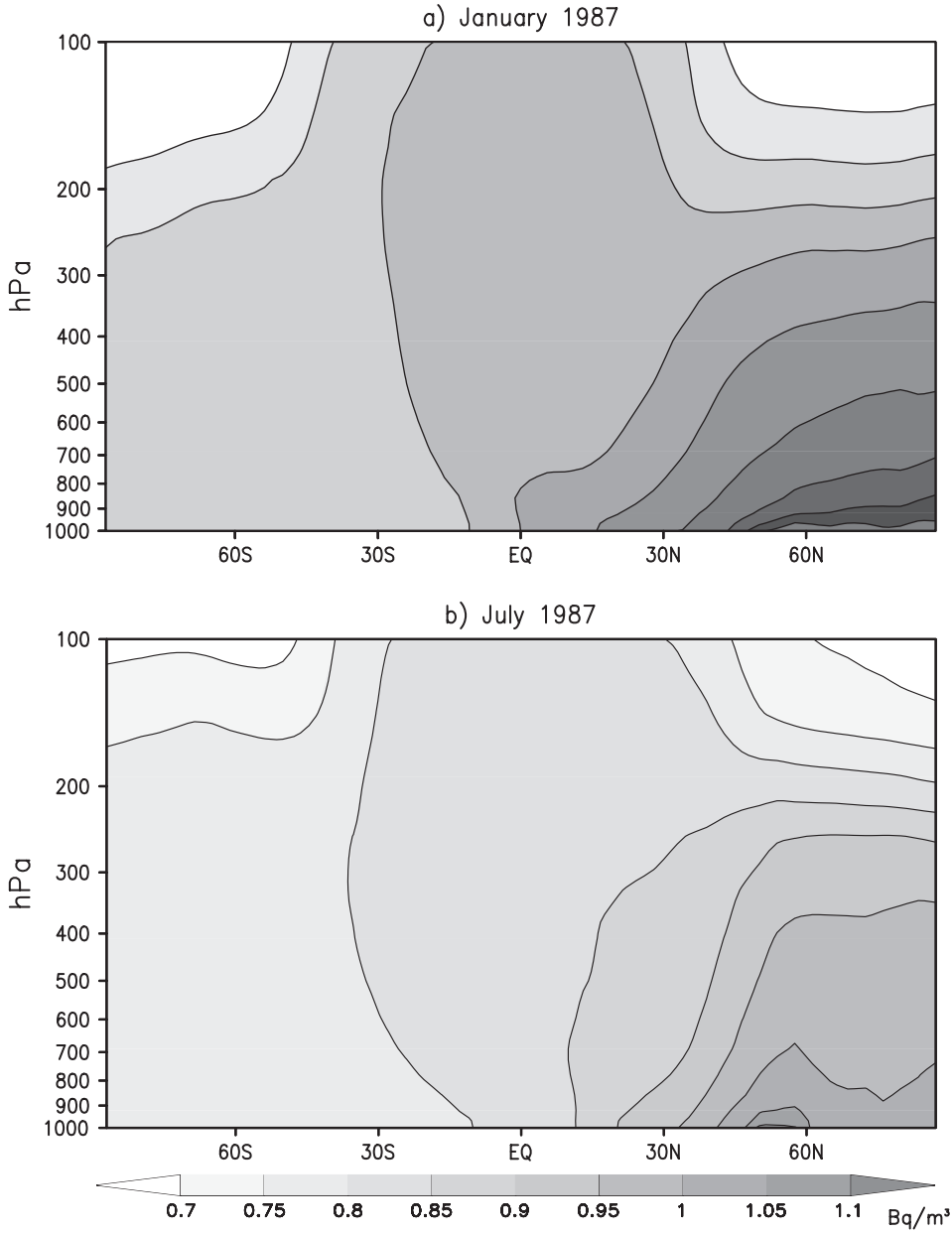


Fig. 6. ^{85}Kr zonal mean distribution in Bq/m^3. (a) January 1987, (b) July 1987.

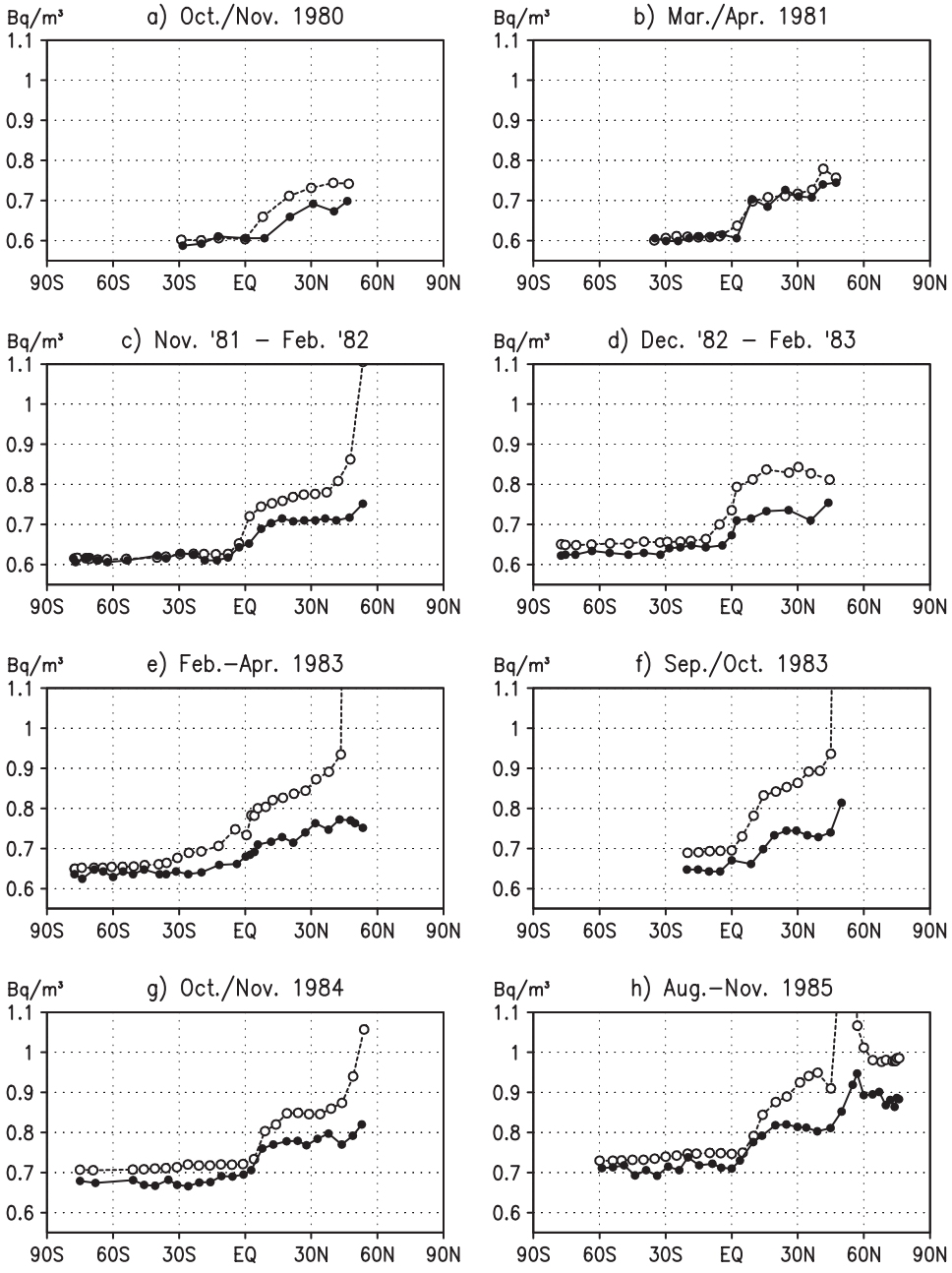


Fig. 7. Meridional ^{85}Kr profiles, observed on Atlantic ship-cruises (filled circles), and corresponding simulated ^{85}Kr concentrations (open circles). The profiles are connected for clarity.

Table 3
Location and operating period of observation stations

Station	Latitude	Longitude	Observation period
Heidelberg	49°24′N	8°42′E	1985–1996
Freiburg	48°00′N	7°51′E	1973–2000
Schauinsland	47°55′N	7°54′E	1976–2000
Madrid	40°25′N	3°43′W	1981–2000
Cracow	50°03′N	19°55′E	1981–1988
Tsukuba	36°03′N	140°08′E	1995–2002
Miami	25°45′N	80°15′W	1981–1998
Cape Point	34°21′S	18°29′E	1985–1997
Cape Grim	40°38′S	144°37′E	1987–1996
Antarctica	70°37′S	8°22′W	1982–1993

(c), (g), and (h). Sampling cruises far away from the continent like (a), (b), and (d) show a distinctly better agreement with the model calculation than sampling cruises very close to the continent.

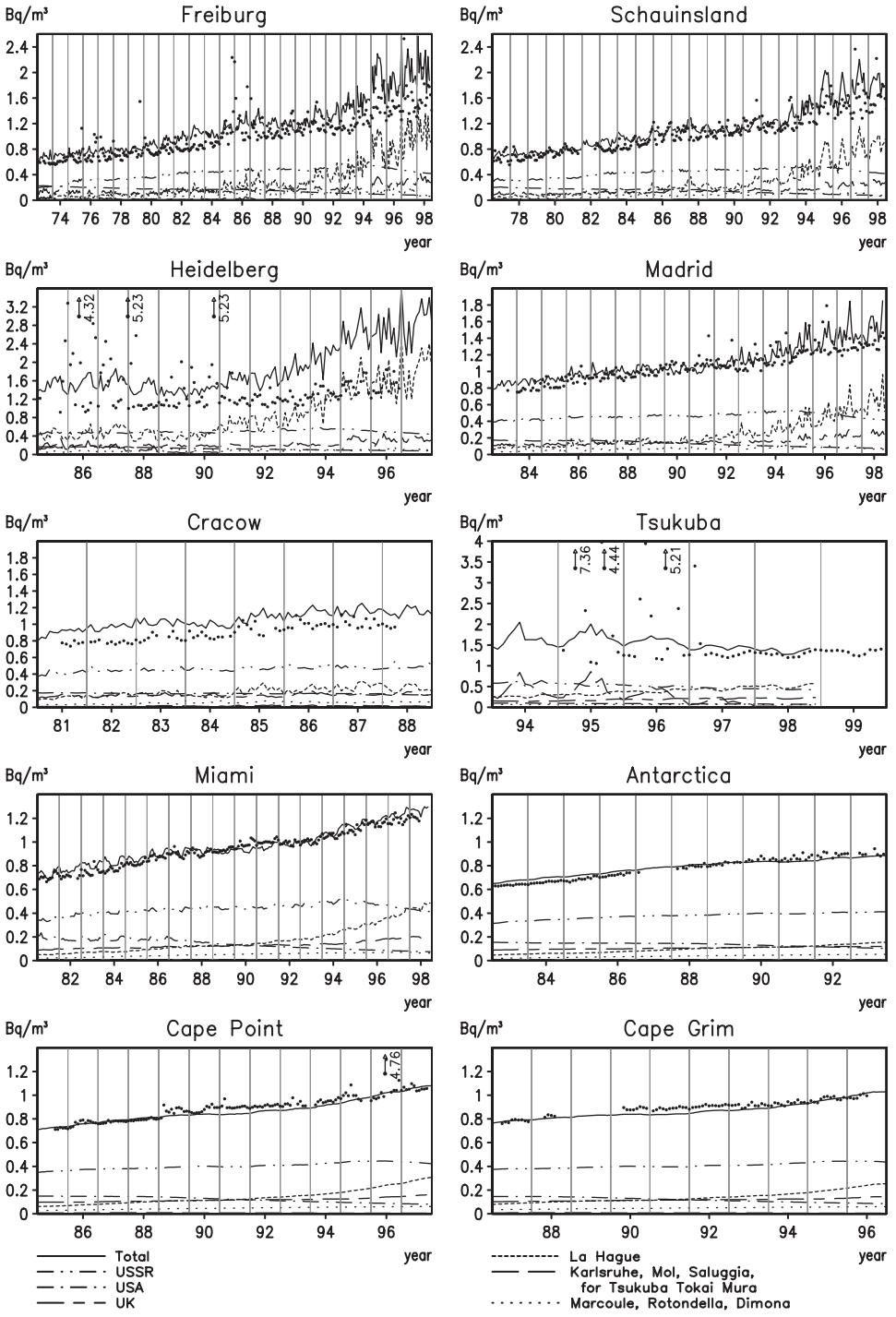
5.2. Temporal evolution

The calculated ^{85}Kr activities are also compared with observed concentrations at 10 selected monitoring sites listed in Table 3. The observed data are mainly provided as weekly averages. For all stations, monthly mean concentrations were calculated for comparison with monthly mean simulated concentrations.

5.2.1. Freiburg, Schauinsland, and Heidelberg, Germany

The observed ^{85}Kr concentrations of the three German observation stations in Heidelberg, Freiburg, and Schauinsland (Fig. 8) show a high variability. They are influenced by the European RFs, mainly La Hague, Karlsruhe, and Sellafield. In general, observed summer concentrations are lower at all three sites. This seasonal variability is not only controlled by transport but can also be caused by reduced emissions due to summer vacations.

Freiburg and Schauinsland, which are very close to each other in horizontal direction (but Schauinsland is about 940 m higher) have a similar background concentration but Freiburg exhibits a stronger variability especially until 1990. In 1991 and the following years, the variability of Freiburg and Schauinsland show a similar behavior. This might be an indication that the variability before 1991 was mainly caused by the influence of a different source than the variability after 1991. In fact, the RF in Karlsruhe stopped operating in 1991, whereas the emission from La Hague increased (from 63 to 100 PBq per year). The contribution of different sources to the observed concentrations is discussed further below. Since Schauinsland is on a mountain and in winter months the site is above the top of the boundary layer, it is not as much influenced by Karlsruhe as Freiburg is, and therefore it shows less variability before 1991 (see Kalinowski et al., 2004).


Freiburg
Schauinsland
Heidelberg
4.32
5.23
5.23
Madrid
Cracow
Tsukuba
7.36
4.44
5.21
Miami
Antarctica
Cape Point
4.76
Cape Grim
Bq/m³
year
Total
USSR
USA
UK
La Hague
Karlsruhe, Mol, Saluggia,
for Tsukuba Tokai Mura
Marcoule, Rotondella, Dimona

The observations taken in Heidelberg show the same pattern as in Freiburg and Schauinsland, except that the variability is much higher from the beginning of the observation until 1990. This result from the fact that Heidelberg is located just 40 km downwind of the main wind direction of the RF Karlsruhe. After 1991, the variability is caused by pulse-like emissions from La Hague. This is not reproduced by the model because the time resolution of the emission data is too coarse.

5.2.2. Impact of different sources

The calculated ^{85}Kr concentration from the North American and the Russian sources show a similar pattern at all European observation sites. According to the model results, ^{85}Kr originating from the USA is slightly decreasing in the displayed time period and well mixed. The fraction of the ^{85}Kr concentration originating from the Russian sources shows a higher variability and increases until 1995. Since 1995, the Russian ^{85}Kr load is decreasing and shows less variability than in the years before, caused by strong decrease of the Russian emissions after 1994. Until then, the highest contribution measured in Freiburg and Schauinsland was emitted by the Russian sources. Since 1995 the highest contribution results from La Hague. But for Heidelberg, La Hague became the most important source already in 1991. Some variability is caused by the Mediterranean and the British sources, as well as by Karlsruhe, Mol, and Saluggia. However, the influence of the source in La Hague explains most of the calculated variability.

5.2.3. Differences between simulation and observations

The general upward trend of observed concentrations in Germany is well reproduced by the mode, except between 1988 and 1990. However, during this period, the simulated concentrations decrease due to reduced emissions from La Hague. After 1993, the simulated concentrations are increasing even faster than the observed concentrations related to high emissions from La Hague. Probably, the influence from the quite close RF La Hague on the German observation stations is overestimated. We see the same behavior as before in the sampling cruises. The model overestimates ^{85}Kr concentrations in grid boxes containing a RF or being close to one. The ^{85}Kr gets distributed over the whole grid box immediately after emission.

At the same time, the model underestimates the influence of Karlsruhe. This can be observed especially at Heidelberg. This is attributed to the short distance between Karlsruhe and Heidelberg, which is lying in the same valley in main down wind direction from Karlsruhe. The ^{85}Kr emitted by the RF Karlsruhe is not far diluted when reaching Heidelberg. But in the model, emissions from Karlsruhe are immediately distributed over the whole grid box and are therefore calculated as much more diluted at Heidelberg, which is located in the same model grid box. At

Fig. 8. Measured monthly mean ^{85}Kr concentration (dots) and simulated monthly mean ^{85}Kr concentration (solid line) at the observation stations. The contribution of some single countries is displayed in the other line styles.

Freiburg and Schauinsland one recognizes still less variability in the simulated concentrations because they are located in a different grid box than Karlsruhe and Heidelberg.

5.2.4. Madrid, Spain

The observed ^{85}Kr background concentration in Madrid (Fig. 8) is a bit lower than the one of the other more northern European stations. It is less influenced by the European sources than the German stations. Especially until 1991, there is much less variability in the observation data because of the greater distance to a RF. Since 1991, when the emissions from La Hague became stronger, also the variability in Madrid increased, but it is still smaller than the variability measured at the German stations.

Until 1996, the most important sources influencing Madrid were the Russian ones, whereas La Hague dominates after 1997. The strongest variability is found in the ^{85}Kr concentrations from Marcoule, Sellafield, and La Hague, especially after 1992. The RF in Karlsruhe has almost no influence on the simulated concentrations in Madrid.

As for all the European stations, the simulated concentrations in Madrid are almost always higher than the observed concentrations. However in the years 1989 until 1991, the agreement between simulation and observation is fairly good, both in terms of the mean and the variability.

5.2.5. Cracow, Poland

In Cracow (Fig. 8), the observed background concentration is the same as in Heidelberg and Freiburg, but there is much less variability. Compared to the German stations, Madrid and Cracow show only little seasonal variability.

At the beginning of the observed time period in 1981, about 50% of the ^{85}Kr in Cracow came from Russia. Due to the increasing contributions from La Hague this rate decreases to about 40% until 1988. From all the European stations, Cracow is the most eastern one and therefore closer to the Russian sources. Hence, concentrations influenced from the Russian sources show a higher variability. But since 1985, even in Cracow the ^{85}Kr released from La Hague is responsible for most of the variability. ^{85}Kr from the Mediterranean and the British sources contributes less and the emissions from Karlsruhe almost nothing to the variability.

Of all available stations, the simulated concentration in Cracow is the one that is most severely overestimated. The difference between simulated and observed background levels is similar for all observed years: between 0.15 Bq/m^3 and 0.2 Bq/m^3. This implies that the increase rate is captured well by the model, but the influence of some sources or their source strengths are overestimated.

5.2.6. Tsukuba, Japan

With regard to the observations taken in Tsukuba (Fig. 8), two features are noteworthy: the constant ^{85}Kr background of 1.2–1.4 Bq/m^3 and several isolated ^{85}Kr peaks up to 8 Bq/m^3. These peaks vanish after the shutdown of the Tokai Mura facility in March 1997, which is a strong indication that they were indeed caused by this local source. (See also Igarashi et al., 2000.) The Tsukuba background data are similar to those observed during the same time period at similar latitudes in Europe.

During periods with no apparent influence of the RF in Tokai Mura, the background level is characterized by a quite slow increase with time and by strong seasonal variations. These are mainly caused by the Asian monsoon, bringing cleaner air from the ocean at summertime, and in winter more polluted air from the northwest.

All the calculated concentrations show this strong seasonal variability. The stronger and closer a source is, the more distinctly marked is the seasonal variability of its concentration. The closest source, Tokai Mura, shows the strongest variability of concentration. However, the seasonality is exactly the opposite of the seasonality of all other sources. There are high values in summer and low values in winter when there is more advection because of the strong winter monsoon.

Tsukuba is located in the same grid box as the emitter Tokai Mura and emissions are distributed in the whole grid box. Hence, as long as the plant in Tokai Mura was emitting, the simulated concentrations are much higher than the observed background level, but because of the assumption of constant emissions, the concentration shows less variability. This overestimate is substantially reduced after the operation of Tokai Mura has stopped in March 1997 (in the model at the end of the year). In 1998, the only year during the simulated and observed period without any emissions from Tokai Mura, the calculated concentration is only marginally higher than the observed and has almost the same seasonality.

5.2.7. Miami, USA

The observations in Miami (Fig. 8) do not show much variability. This indicates that Miami is not influenced by nearby sources. The background level is increasing, especially since 1994, when the emissions from La Hague increased. After the shutdown of the North American RFs in 1989 and before the increase of the emission in La Hague in 1993, the seasonal amplitude was quite small.

The shutdown of the last two North American RFs in 1989 is distinctly marked at the measurement site in Miami. After 1989 there is almost no short-time variability in the simulated ^{85}Kr concentrations of North America. The model's variability is mainly caused by the contributions from the USA, Russia and later on by the rapidly increasing contributions from La Hague. The seasonal variability is not caused by varying emissions, since the emissions are constant over a period of one year, but due to seasonal varying meteorology.

The agreement between observed and calculated concentrations in Miami is excellent and the seasonality is also captured very well. However, in the years 1990 and 1991 the simulation shows lower concentrations than the observation. In contrast to the European stations, Russian sources have the main impact at Tsukuba and Miami until the year 1997.

5.2.8. Southern hemisphere

Observed concentrations in the SH (Fig. 8) are much lower than in the NH. Furthermore, the concentration decreases from the equator to the pole by about 0.1 Bq/m^3.

The measurements show an almost steady linear increase of the background ^{85}Kr concentration throughout the observed time period at all three sites. The magnitude

of the variability gives information about the influence of nearby ^{85}Kr sources. In general, close to the sources, the variability is high due to pulse shaped emissions and varying local transport.

The simulated concentrations at the three stations are very similar. The concentrations exhibit a weak seasonality with higher values in the southern winter and lower values in summer. This seasonality is caused by the seasonal varying interhemispheric transport. In contrast to the sites in the NH, the Russian contribution remains the most important until the end of the simulation. The decline in the concentrations from Russian sources occurs about one year later than in the NH.

In the years 1983–1989, the calculated concentrations are overestimated in Antarctica. This corresponds to the model overestimations along the ship-cruises (see Fig. 7). The overestimate vanishes in 1989 and is not observed at the onset of the time series for Cape Point (starting 1985) and Cape Grim (starting 1987). Between 1989 and 1995, the model tends to underestimate the concentrations at all three sites. This could possibly be due to an underestimation of the ^{85}Kr emissions in the years where the measurements bump up (1989, 1990, 1995). Since these increases in the observations are relaxing very slowly, they must be due to a source lasting almost over the whole period where the model underestimates the observations. It seems likely that the SH sources which are reporting emissions during this period are underestimated. The higher concentrations measured in 1995 could then be due to an unknown source in the SH. A definite answer to this question could only be given by further research with a model that has a better resolution.

At the beginning of the simulation, variability from month to month is very low. However, variability is enhanced at Cape Point in 1989, in Antarctica in 1991, and at Cape Grim in 1993 whereas Cape Point exhibits the highest variability. There are some single months with higher concentrations, some periods with elevated concentrations as in 1986, 1989, and 1995, and single peaks in 1990 and 1996. The variability and in particular the single peaks which are about 10% higher than the background concentration, indicate the influence of a regional source.

According to available information, there are only two RFs in the SH, Ezeiza in Argentina and Pelindaba in South Africa. Ezeiza was reported to operate in the years 1989 and 1990 and Pelindaba during the years 1986–1992. Therefore, it may be that the peaks at Cape Point are caused by a SH source which is not included in our emission inventory.

5.3. Impact of the temporal resolution of the model emissions

Because the emission inventory contains, except for La Hague, just annual data, the emissions used in the model are held constant over a period of one year. However, in reality, the release of ^{85}Kr is not continuous at all. The release rates of the few plants reporting temporally better resolved emissions like La Hague (Fig. 3, monthly emissions), Karlsruhe (Kalinowski et al., 1998, weekly and daily emissions) and Tokai Mura (Igarashi et al., 2000, daily emissions) show substantial short term and seasonal variations due to the sporadic processing cycle. In order to assess the

influence of the temporal resolution of the emissions on the concentrations, an additional simulation was performed with monthly emission rates for the RF in La Hague, the only RF of which a monthly resolution of the emissions is available for a longer period. For the years for which no monthly emissions were available (1968–Aug l974 and 1987–Sep l994), the annual emission was evenly distributed over the months, assuming no emissions during the holiday season, in July and August, for the period 1968–1973 and reduced emissions in July and August for 1987–1994 (Fig. 3).

As expected, the variability of the concentrations calculated with emissions varying from month to month is higher than the variability calculated with emissions held constant over one year, and is in better agreement with the observations. For example, the comparison of the years 1994 to 1998 is shown in Fig. 9. Comparing the results at Heidelberg and Freiburg (Fig. 9), the background concentration calculated with the monthly emissions is nearly always lower than the background concentration calculated with the yearly emissions, whereas the variability from the monthly emissions is higher.

6. Summary and conclusions

A global-scale ^{85}Kr emission inventory has been compiled covering the period from the beginning of anthropogenic emissions in 1945 until the year 2000. Annual emissions for each reprocessing facility have been gathered from the literature. Estimates were made where no release data were available. According to this data set, 10,600 PBq (Peta = 10^{15}) of ^{85}Kr have been emitted globally by reprocessing facilities until the end of 2000. The global atmospheric inventory at the end of the year 2000 amounts to 4800 PBq.

The emissions have been incorporated into the ECHAM4 atmospheric general circulation model as point sources to calculate the ^{85}Kr distribution for the years 1945–1998. In order to study the impact of specific emitters on the ^{85}Kr distribution different tracers were used for different emissions. To lower the number of tracers some emissions were combined within a specific region. The resulting monthly mean atmospheric ^{85}Kr concentrations have been compared to observations taken at several observation sites and at Atlantic ship-cruises. At each observation site the temporal evolution of the observed and calculated concentrations, the contributions from different emitters and the variability of the concentrations have been analyzed. High variability has been considered as an indicator for influence of sources close to the receptor site. Based on this comparison the quality of the inventory has been assessed.

The calculated concentrations are found to give reasonably good agreement with the observations indicating that the emission inventory is reliable. However, at all northern hemispheric observation sites the model tends to slightly overestimate the concentrations, in particular at the German sites in the 1990s. The station Madrid/Spain shows a similar behavior, but the agreement is better than at the German sites. At Cracow/Poland the model overestimates the concentration by a rather constant value of about 0.15–0.20 Bq/m^3 throughout all the years for which observations are available (1980–1989). It is unlikely that a systematic bias in the measurements

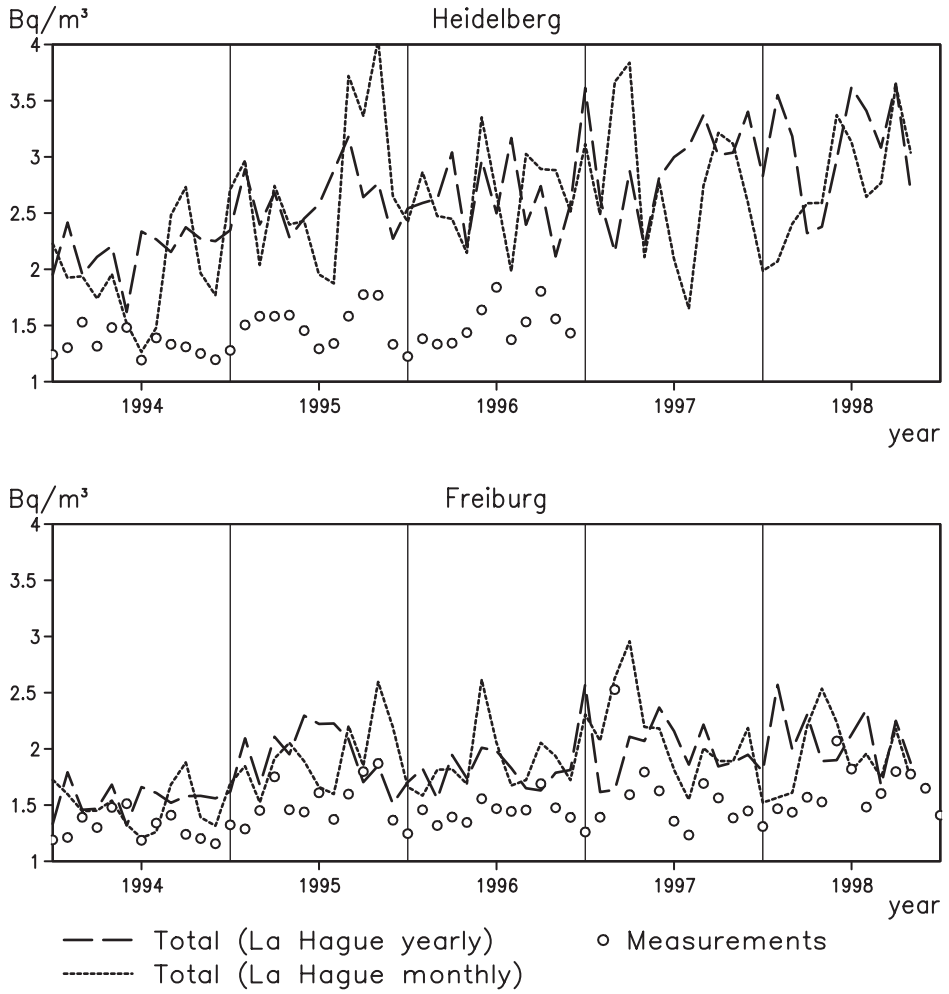


Fig. 9. Measured monthly mean ^{85}Kr concentration at the observation sites Heidelberg and Freiburg (circles) and simulated ^{85}Kr concentration using annually resolved (long dash) and using monthly (short dash) emission from the RF in La Hague.

explains this discrepancy. Since the emission data of La Hague are known with high certainty, the USSR/Russian source estimates could be a reason for this overestimation. Their simulated contribution to the concentrations found at Cracow is about two times as high as the discrepancy. No other source could be the reason because none of them has a simulated contribution larger than the discrepancy and a 100% error can be ruled out.

A sensitivity study with monthly varying emissions also yielded the result that the annual constant emissions used in the base run lead to higher concentration and less variability close to sources. Further away from sources, only very small differences in the concentrations from monthly varying, respectively, annual constant emissions

can be found. In reality, the emissions show a high intra-annual variability. Hence, this may explain to some extent the model's too high concentrations. In addition, the coarse resolution of the model worsens the agreement with observations in the vicinity of the sources. Observations close to emission sources cannot properly be reproduced with a $3.75^\circ \times 3.75^\circ$ grid since the characteristic spatial scale of a plume is much smaller than a grid box. This may explain why the largest discrepancies are found at the German stations which are the closest to the RF in La Hague.

In contrast to the European stations, model predictions at Tsukuba/Japan and Miami/US show a very good agreement with the observations. In Antarctica the model simulates somewhat higher concentrations than observed until the year 1990. After 1990 the simulations are too low compared to the observations. At Cape Point/South Africa and Cape Grim/Tasmania the agreement is almost perfect until the end of 1988, whereas between 1989 and 1995 the model underestimates the concentrations and exhibits a lower variability.

The relatively high variability in the observations at Cape Point requires some consideration. It persists until the end of the observation period in 1997. According to available information, the only regional reprocessing facility which could cause such variability, the RF Pelindaba in South Africa, was shut down in 1992. Higher variability indicates influence from a regional source, definitely originating from the same hemisphere. Therefore, it is likely that the spikes at Cape Point and similarly the variability at Cape Grim are caused by a SH source which is not included in the emission inventory. Possible sources are most likely to be found within a few hundred kilometers upwind from Cape Grim in the main wind direction. According to the simulated impact of the known source location in Argentina compared to the known source location in South Africa, the unknown source is probably in Africa, south of the equator. It is unlikely to be located in South America.

The nuclear weapon tests that France conducted in the SH during the relevant period were reported to be occurring under ground and their dates do not correlate with the observed spikes. Therefore, they can be excluded as possible sources. The most likely ^{85}Kr source results from radionuclide production based on nuclear fission. Technetium-99m (^{99m}Tc) is the most widely used isotope in radiopharmaceuticals for diagnostic and therapeutic nuclear medicine. It is the short-lived daughter product of molybdenum (^{99}Mo). Though this isotope could be produced by neutron capture in ^{98}Mo production, the most efficient method is utilizing nuclear fission in highly (80%) enriched uranium-235 (HEU). The Atomic Energy Corporation of South Africa Limited (AEC) is applying this method at its 20 MW SAFARI reactor at Pelindaba (see http://www.aec.co.za/moly99/index.htm). The same process produces ^{85}Kr which is inadvertently released during the extraction of ^{99}Mo from the irradiated targets. The simultaneous plutonium generation is low. In order to meet the annual consumption of ^{99m}Tc of a small industrialized country like Austria, the production would release less than 1 TBq of ^{85}Kr. The USA has an annual consumption that is three orders of magnitude larger. Therefore, an annual release of ^{85}Kr due to ^{99}Mo production from HEU could plausibly lie between 1 TBq and 1 PBq. Hence, it might explain the observed spikes at Cape Point. However, Pelindaba does not appear to be a likely location to explain these spikes, because in the period prior to 1993 there was

no correlation between reported emissions at that site and spikes registered at Cape Point. This can easily be understood from the large distance of about 1000 km and the fact that western winds predominate at that latitude and any release from Pelindaba would rather be moved away from the detector site.

Besides plutonium separation from irradiated fuel, there are other sources of ^{85}Kr which can be neglected in the northern hemisphere because they are dwarfed by the releases from RFs. However, in the absence of reprocessing in the southern hemisphere, these sources will dominate.

In the 1990s the bias between observations and calculations increases at almost all northern hemispheric sites. This is another predominant feature that requires discussion. It is not likely to be caused by the model and most likely reflects an inaccuracy of the release data. Russian sources are associated with the largest uncertainty. It might be that the decline in the Russian emissions assumed to occur in 1994 started already earlier. However, this is a period for which original release data are available. The hypothetical overestimation of the Russian emissions might be compensated by the underestimation of the southern hemispheric emissions, so that, as a result of this compensation, the calculated ^{85}Kr concentration at the three southern hemispheric stations is still close to the observations.

Acknowledgments

This paper is based on a Diploma thesis in meteorology by one of the authors (Winger, 2002). It was conducted at the Max-Planck-Institute for Meteorology in Hamburg, Germany.

The BfS acknowledges the support of many colleagues in operating ^{85}Kr sampling stations: C. Dick and N. Tindale (Cape Grim, Australia), I. Levin (Heidelberg, Germany), K. Różański (Cracow, Poland), C. Heras Iñiguez (Madrid, Spain), W. Brunke (Cape Point, South Africa) and R. Oleson and G. Östlund (Miami, USA). The ^{85}Kr samples from the Georg von Neumeyer station were collected by various members of staff of this German Antarctica station and transported to the BfS with the help of the Institute for Environmental Physics at the University in Heidelberg. We thank the Meteorological Research Institute in Tsukuba, Japan for the valuable co-operation in the field of noble gas measurements.

References

Achkasov, S.K., Gudkov, A.N., Zakharov, O.V., Krylov, A.Yu., Nekrasov, V.M., Novichkov, V.P., Serbulov, Yu.A., Ushakova, N.P., Zadorozhnyi, Yu.A., 1991. Monitoring the Contamination of the Atmosphere by ^{85}Kr. Moscow Engineering Physics Institute. Translated from Atomnaya Energiya 70 (4), 234–239 (April 1991).

Albright, D., Berkhout, F., Walker, W., 1997. Plutonium and Highly Enriched Uranium 1996, World Inventories, Capabilities and Policies. In Oxford University Press, ISBN: 0-19-828009-2.

Ashley, C., Zeigler, C.C., 1980. Releases of Radioactivity at the Savannah River Plant, 1954 through 1978, DPSPU 75-25-1 (1980); Environmental Monitoring at the Savannah River Plant, Annual Reports, DPSU 72-302 (1971), DPSPU 75-302 (1974), and Environmental Monitoring in the Vicinity of the

Savannah River Plant, Annual Reports, DPSPU 74-30-1 (1973), DPSU 75-30-1 (1974), DPSU 76-30-1 (1975), DPSPU 77-30-1 (1976), DPSPU 78-30-1 (1977), DPSPU 79-30-1 (1978), DPSPU 80-30-1 (1979), DPSPU 81-30-1 (1980), DPSPU 82-30-1 (1981).

Avery, M.A., Westberg, D.J., Fuelberg, H.E., Newell, R.E., Anderson, B.E., Vay, S.A., Sachse, G.W., Blacke, D.R., 2001. Chemical transport across the ITCZ in the central Pacific during an El Niño-Southern Oscillation cold phase event in March–April 1999. J. Geophys. Res. 106 (D23), 32539–32553.

Bradley, Don J., 1997. Behind the Nuclear Curtain: Radioactive Waste Management in the Former Soviet Union. Battelle Press, Columbus, OH, ISBN: 1574770225.

British Nuclear Fuels Limited (BNFL), Health and Safety Directorate, Risley (1985 until 1996). Annual report on radioactive discharges and monitoring of the environment 1984 (until 1995).

British Nuclear Fuels Limited (BNFL), Health and Safety Directorate, Risley (1996 until 2000). Annual report on radioactive discharges and monitoring of the environment 1995 (until 1999).

Cochran, T.B., Norris, R.S., 1993. Russian/Soviet Nuclear Warhead Production. Nuclear Weapons Databook, 93-1.

Cogema-La Hague. http://www.cogemalahague.com/servlet/ContentServer?pagename=cogema_en/common/gotopage&assetid=1039482706934&type=Page&callingpage=1039482706928.

EG&G Idaho, Inc. Radioactive Waste Management Information, 1982 Summary and Record-to-Date. IDO-10054(82).

Franke, B., Alvarez, R., 1983. Analysis of External [Gamma]-Radiation Monitoring Around the Savannah River Plant. Washington, D.C.: Environmental Policy Institute, Draft Report, 1983.

Gili, M., ENEA, 1996. Private communication to M. Kalinowski. Email dated 6 August 1996.

Hilbert, F., 1975. Erzeugung und Freisetzung von radioaktiven krypton- und xenon-isotopen durch Kernreaktoren und Wiederaufbereitungsanlagen und die voraussichtliche radiologische Belastung bis zum Jahr 2000. KFK 2035.

von Hippel, F., Albright, D.H., Levi, B.G., 1986. Quantities of Fissile Materials in US and Soviet Nuclear Weapons Arsenals. Center for Energy and Environmental Studies. The Engineering Quadrangle, Princeton University, PU/CEES Report No. 168.

Igarashi, Y., Sartorius, H., Miyao, T., Weiss, W., Fushimi, K., Aoyama, M., Hirose, K., Inoue, H.Y., 2000. ^{85}Kr and ^{133}Xe monitoring at MRI, Tsukuba and its importance. J. Environ. Radioactivity 48, 191–202.

Igarashi, Y., Aoyama, M., Nemeto, K., Hirose, K., Miyao, T., Fushimi, K., Suzuki, M., Yasui, S., Asai, Y., Aoki, I., Fujii, K., Yamamoto, S., Sartorius, H., Weiss, W., 2001. ^{85}Kr measurement system for continuous monitoring at the Meteorological Research Institute, Japan. J. Environ. Monitoring 3, 688–696.

Izrael, Y.A., Nazarov, I.M., Ryaboshapko, A.G., 1982. Release of man-made krypton 85 to the atmosphere. Meteorologiya i Gidrologiya No. 6, 5–15.

Jackson, D., Zimmermann, C.H., Gray, J., 1998. Discharges of krypton from Sellafield, 1951–1997, and the resultant doses to members of the public. J. Radiol. Prot. 18 (2), 111–118.

Jacob, D.J., Prather, M.J., Wofsy, S.C., McElroy, M.B., 1987. Atmospheric distribution of ^{85}Kr simulated with a general circulation model. J. Geophys. Res. 92 (D6), 6614–6626.

Kalinowski, M.B., 1997. Measurements and modeling of atmospheric krypton-85 as indicator for plutonium separation. International Workshop on the Status of Measurement Techniques for the Identification of Nuclear Signatures, Geel, 25–27 Feb. 1997.

Kalinowski, M.B., Sartorius, H., Uhl, S., Weiss, W., 1998. Rückschliessbarkeit auf Plutoniumabtrennungen. IANUS 3/1998, TU Darmstadt.

Kalinowski, M.B., Sartorius, H., Uhl, S., Weiss, W., 2004. Conclusions on plutonium separation from atmospheric krypton-85 measured at various distances from the Karlsruhe reprocessing plant. J. Environ. Radioactivity 73 (2), 203–222.

Kernforschungszentrum Karlsruhe, Abteilung Sicherheit. Jahresberichte 1971 until 1991.

Lippolis, G., ENEA, 1996. Private communication to M. Kalinowski. Letter dated 10 Oct. 1996.

Loosli, H.H., 1992. Applications of ^{37}Ar, ^{39}Ar and ^{85}Kr in hydrology, oceanography and atmospheric studies, Current state of the art. Technical Report of the Results of a Consultants Meeting 'Isotopes of Noble Gases as Tracers in Environmental Studies', IAEA, Vienna, Technical Report Series No. 332, pp. 73–85.

Luykx, F., Fraser, G., 1978. Radioactive effluents from nuclear power stations and nuclear fuel reprocessing plants in the European Community, Discharge data 1972–76, radiological aspects. Commission of the European Communities, Directorate-General Employment and Social Affairs, Health and Safety Directorate, Luxembourg, Report EUR 6088, EN, FR.

Luykx, F., Fraser, G., 1980. Radioactive effluents from nuclear power stations and nuclear fuel reprocessing plants in the European Community, Discharge data 1974–78, radiological aspects. Commission of the European Communities, Directorate-General Employment and Social Affairs, Health and Safety Directorate, Doc.Nr.- V/4116/80.

Luykx, F., Fraser, G., 1983. Radioactive effluents from nuclear power stations and nuclear fuel reprocessing plants in the European Community, Discharge data 1976–80, radiological aspects. Directorate-General Employment, Social Affairs and Education, Health and Safety Directorate, Luxembourg.

Novichkov, V. Moscow Physical Engineering Institute. Private communication to H. Sartorius (preliminary information).

Rasch, P.J., Lawrence, M., 1998. Recent development in transport methods at NCAR. Report No. 265, Max-Planck-Institut für Meteorologie, Hamburg, pp. 65–75.

Rath, H.-K., 1988. Simulation der globalen ^{85}Kr und $^{14}CO_2$ Verteilung mit Hilfe eines zeitabhängigen, zweidimensionalen Modells der Atmosphäre. Inaugural-Dissertation, Ruprecht-Karls Universität, Heidelberg.

Rayner, N.A., Horton, E.B., Parker, D.E., Folland, C.K., Hackett, R.B., 1996. Version 2.2 of the global sea-ice and sea surface temperature data set, 1903–1994, Clim. Res. Tech. Note 74. Hadley Centre, Bracknell, UK.

Roeckner, E., Arpe, K., Bengtsson, L., Brinkop, S., Dümenil, L., Esch, M., Kirk, E., Lunkeit, F., Ponater, M., Rockel, B., Sausen, R., Schlese, U., Schubert, S., Windelband, M., 1992. Simulation of the present-day climate with the ECHAM model: Impact of model physics and resolution. Report No. 93, Max-Planck-Institut für Meteorologie, Hamburg, Germany.

Roeckner, E., Arpe, K., Bengtsson, L., Christoph, M., Claussen, M., Dümenil, L., Esch, M., Giorgetta, M., Schlese, U., Schulzweida, U., 1996. The atmospheric general circulation model ECHAM-4: Model description and simulation of present-day climate. Report No. 218, Max-Planck-Institut für Meteorologie, Hamburg, Germany.

Różański, K., 1979. Krypton-85 in the atmosphere 1950–1977: a data review. Environ. Intern. 2 (13) 139–143.

Schröter, K.J.P., Roether, W., 1975. The release of krypton-85 and tritium to the environment and tritium to krypton-85 ratios as source indicators. Isotope ratios as pollutant source and behavior indicators, IAEA, Vienna.

Simmons, A., Burridge, D., Jarraud, M., Girard, C., Wergen, W., 1989. The ECMWF medium-range prediction models: development of the numerical formulations and the impact of increased resolution. Meteorol. Atmos. Phys. 40, 28–60.

Sittkus, A., Stockburger, H., 1976. Krypton-85 als Indikator des Kernbrennstoffver-brauchs. Naturwissenschaften 63, 266–272.

Styra, B., Butkus, D., 1990. Geophysical Problems of Krypton-85 in Atmosphere. Washington.

Syndicate CFDT de l'Energie Atomique, 1980. Le Dossier Electronucleaire. Editions du Seuil, pp. 188–190.

Telegades, K., Ferber, G.J., 1975. Atmospheric concentrations and inventory of krypton-85 in 1973. Science 190, 882–883.

US Atomic Energy Commission, Idaho Operations Office. National Reactor Testing Station Radioactive Waste Management Information, 1972. Summary and Record-to-Date. IDO-10054(72).

US Department of Energy, 1982. Summary of annual environmental Reports for CY 1980 (1981). Department of Energy Nuclear Sites, Assistant Secretary for Environmental Protection, Safety and Emergency Preparedness Office of Operational Safety, Washington D.C., DOE/EP-0038, DE82 020467 (DE83 008175).

US Department of Energy, Idaho Operations Office, 1984. 1983 Environmental Monitoring Program Report for Idaho National Engineering Laboratory Site. DOE/ID-12082-83, p. 17.

Weiss, W., Sartorius, H., Stockburger, H., 1992. Global Distribution of Atmospheric Krypton-85, A Data Base for the Verification of Transport and Mixing Models, Technical Report of the Results of a Consultants Meeting 'Isotopes of Noble Gases as Tracers in Environmental Studies', Technical Report Series No. 332, IAEA, Vienna, pp. 29–62.

Winger, K., 2002. Compilation and evaluation of Krypton-85 emission inventories from 1945 until 2000. Diplomarbeit, Max-Planck-Institut fü Meteorologie, Hamburg.

WISE News Communique, 1994. AEA Technology application to H.M. Industrial Pollution Inspectorate (HMIPI) 1993. No. 405, 28 January 1994.

WMO Global Atmosphere Watch, 1995. Report of an Expert Consultation on ^{85}Kr and ^{222}Rn: Measurements, Effects and Applications. Freiburg, Germany, No. 109, TD No. 733.

Zimmermann, P.H., Feichter, J., Rath, H.-K., Crutzen, P.J., Weiss, W., 1988. A global three-dimensional source-receptor model investigation using ^{85}Kr. Atmos. Environ. 23, 25–35.