MEASUREMENT OF ATMOSPHERIC ^{85}KR IN GENT, BELGIUM: HISTORY AND PERSPECTIVES

Timothy De Clerck °, André Poffijn °, Gilbert Eggermont *

° University Gent, Subatomic and Radiation Physics Department, Proeftuinstraat 86, 9000 Gent, Belgium

*** SCK-CEN, Boeretang 200, 2400 Mol, Belgium**

The Ghent University started the study of ^{85}Kr atmospheric pollution in 1974. Nuclear prospects after the oil crisis were very optimistic, assuming complete reprocessing of all spent fuel in only a limited number of plants (1). The increase of atmospheric concentration at global level and high local concentrations near release points were estimated to be far from negligible in the beginning of this century (10% MPC).

The ICRP 26 concept modification in 1977 was giving marginal weight to skin effects. Moreover the slowing down of nuclear energy programmes and of military and civil reprocessing had reduced considerably the health concerns. Some environmental uncertainties regarding atmospheric interaction, increasing acidification (2) and climate impact were studied but finally received no priority policy attention.

The few remaining reprocessing centres in the world continued to release all ^{85}Kr in the atmosphere till now. The present release worldwide is less than 10^{18} Bq/y with an atmospheric inventory of less than 10^{19} Bq due to the 10y half-life time. If reprocessing continues at increasing burn-up and shorter cooling times, 10^{20}Bq could be reached only from 2020 on.

The atmospheric concentration was measured systematically in Gent from 1979. A system was set up starting with air sampling and cryogenic trapping on charcoal. The trapped gases are led into a small metal coil filled with molecular sieves, allowing sampling at different locations. After gas-chromatographic purification and separation the mass of 1 ppm of krypton in air is determined. The nuclear detection of ^{85}Kr β 's is realised by liquid scintillation counting of krypton condensed in a glass vial (3). The overall uncertainty for a typical 2m³ background air sample is better than 2,5% at 68% CL with a reproducibility of 1% (1).

Over the period 1980-1998 the results of atmospheric measurements in Gent have shown a steady increase of the background ranging from 0,7 Bq/m³ to over 1,4 Bq/m³ with peak values up to fifty times these values (4), see figure 1. Since 1999 the atmospheric concentration is stabilising, coinciding with a drop in emissions at the commercial reprocessing centres.

Local high concentrations can occur in inversion conditions at small distances of release points, while backtracking techniques can be applied for meso-scale distances. The Tsjernobyl ^{85}Kr

release in 1986 could even been measured at large distances. Some days after the accident elevated concentrations were measured with a wind direction opposite the usual sources (5).

It was realised and demonstrated that the atmospheric dispersion of the huge quantities of released radioactivity in only some sites could allow dedicated application of tracer studies.

This very sensitive measurement system has been used in Ireland and UK to follow releases at Sellafield and in order to validate dispersion calculations. The results of the longstanding cooperation with the Radiological Protection Institute of Ireland also proved the reliability of the two-stage approach.

As the replica of the air-sampling unit is difficult to move around, a new, more manageable sampling concept was developed, in which air is collected into diving bottles. To facilitate sampling in the field a portable air compressor is applied. The air is compressed up to a pressure of 200 bars in bottles of six litres in volume, permitting a sample volume of about 1,2 m^3 STP. The bottles can then be hooked up to the original air-sampling unit, without any major modification.

Release data from La Hague made available some years ago have allowed to confirm the tracer capacity of ^{85}Kr. The analysis of release data of Cogéma has shown considerable underestimations over the period 1984-96 while the annual release is now already about 50% of the authorised limit ($4.8\ 10^{17}$ Bq).

Currently the sampling method is being evaluated by means of a campaign in collaboration with the French Institute of Radiation protection and Nuclear Safety (IRSN) at Cherbourg. The use of our sensitive ^{85}Kr measurement system to validate atmospheric dispersion models and other measuring systems could offer perspectives.

The use of ^{85}Kr for safeguard purposes through environmental monitoring has been considered. This could greatly facilitate in the long run the monitoring of compliance with the non-proliferation treaty where a combined knowledge of existing releases and source terms is required together with a network of measurement systems (1).

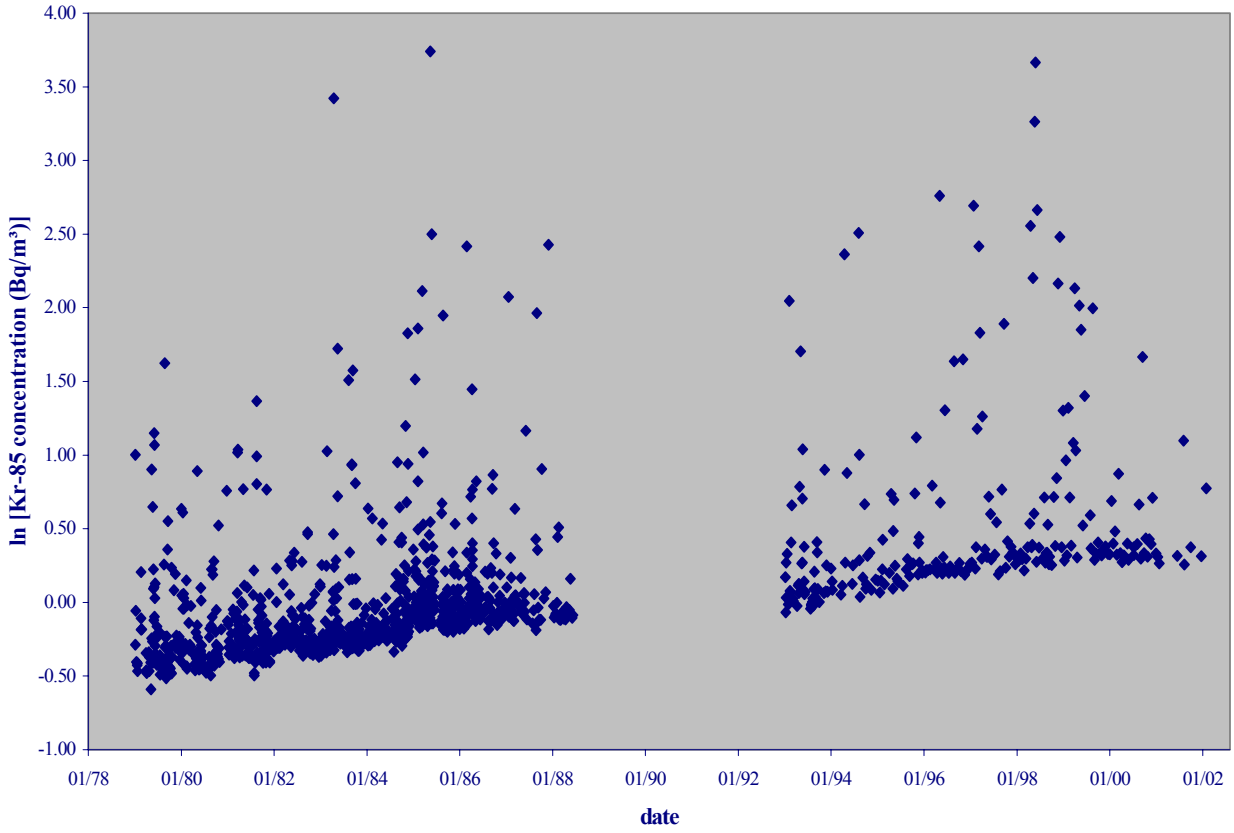

Figure 1: An overview of krypton-85 measurements from 1979 on

REFERENCES

[1] A. POFFIJN and G. EGGERMONT (Eds.), Proceedings of the ^{85}Kr Workshop, Brussels, 1998, SCK•CEN, BLG-835, Mol, 1999.

[2] F. RAES, A. JANSSENS and G. EGGERMONT, A synergism between UV- and γ - radiation in producing aerosol particles form SO_2-H_2SO_4 laden atmospheres, Atmospheric Environment, 20, 9, 1705, 1986.

[3] A. JANSSENS, J. BUYSSE, F. RAES and H. VANMARCKE, An improved method for the sampling of atmospheric ^{85}Kr, Nuclear Instruments and Methods in Physics Research, B17: 564-567, 1986.

[4] P. CAUWELS, J. BUYSSE, A. POFFIJN and G. EGGERMONT, Study of the atmospheric ^{85}Kr concentration growth in Gent between 1979 and 1999, Radiation Physics and Chemistry 61 (2001) 649-651, 2001.

[5] G. EGGERMONT, J. BUYSSE, P. BERKVENS and F. RAES, Atmospheric concentration measurements of ^{85}Kr – A tool for ecological standards, Proc. Int. Conf. Europ. IRPA Salzburg,1986