SnT 2025 8 SEPTEMBER ONLINE DAY 9 TO 12 SEPTEMBER AT HOFBURG PALACE, VIENNA & ONLINE
CTBT: SCIENCE AND TECHNOLOGY CONFERENCE

20 years of Krypton-85 measurements revisited

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INTRODUCTION AND MAIN RESULTS

Large quantities of the radioactive noble gas Kr-85 are released into the atmosphere as a result of reprocessing of used nuclear fuels. Reprocessing started in the 1940s mainly to separate plutonium for military purposes and the monitoring of Kr-85 has been suggested as an indicator for clandestine plutonium production. However, emissions from civil reprocessing activities have created a significant background.

Bundesamt für Strahlenschutz (BfS) has been operating a network with weekly air sample collection at various sites (26) and time periods in Germany and worldwide, with sampling going back to 1973. The samples are analyzed for Kr-85 at the BfS Noble Gas laboratory in Freiburg which is accredited according to DIN EN ISO/IEC 17025.

We analyze the data from 2005-2024 with backward atmospheric transport modelling (ATM) for 5 stations. Using high resolution emission data, we simulate the contribution of the reprocessing plant in La Hague. Considering this background, we discuss the possibility of distinguishing different emitters using the recent shutdown of the Sellafield nuclear reprocessing plants as an example.

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T3.6-700

Krypton

- Noble gas
- Stable Krypton content is constant in the atmosphere
- Kr-85 has a half-live of 10.76 y
- It is used for dating young groundwater and ice, and ideal for atmospheric tracer studies, e.g. the determination of the interhemispheric exchange time

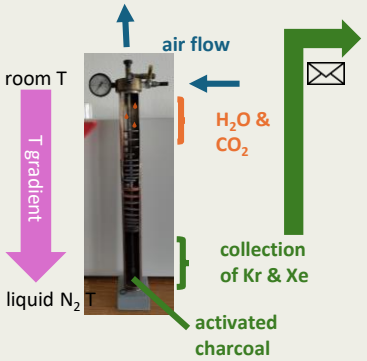
Motivation

- BfS has been measuring Kr-85 for more than 50 years. (Why) continue with weekly sampling in a dense network? Can we identify redundancies?
- The Fissile materials cut-off treaty is a proposed international treaty to prohibit the further production of fissile material for nuclear weapons. This treaty could be internationally monitored similar to the CTBT.

Is it possible to identify an unknown emitter, given
- The substantial background of Kr-85?
- The presence of large reprocessing facilities?

Here, we present a study case:
- Vast amount of weekly data
- Shutdown of reprocessing facilities in Sellafield, UK: THORP in 2018, MAGNOX in 2022
- while another large reprocessing facility is in operation “near by”: ORANO/La Hague

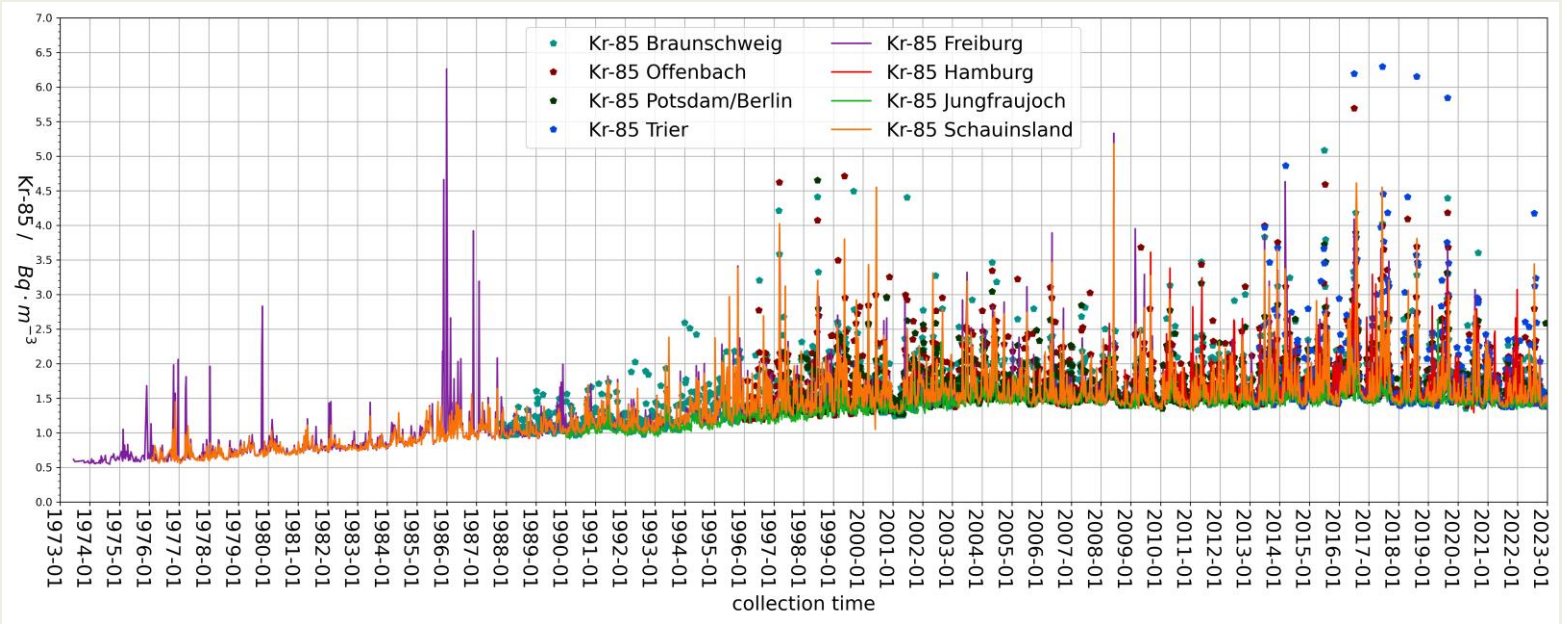
Sample collection & measurement



Laboratory (Freiburg):
2 proportional gas counter systems with Pb-shielding, anticoincidence counters and
GC volume determination;
Accredited according to DIN EN ISO/IEC 17025.

Measurement data

Shown here is weekly sampling data from seven German stations and the Swiss research station on Jungfraujoch (3400 m a.s.l.).
Two different features can be observed:
- Since measurements were started, the baseline constantly increased and is in a “steady state” since about 2005
- Distinct peaks are caused by near-by emitters (La Hague, Sellafield)



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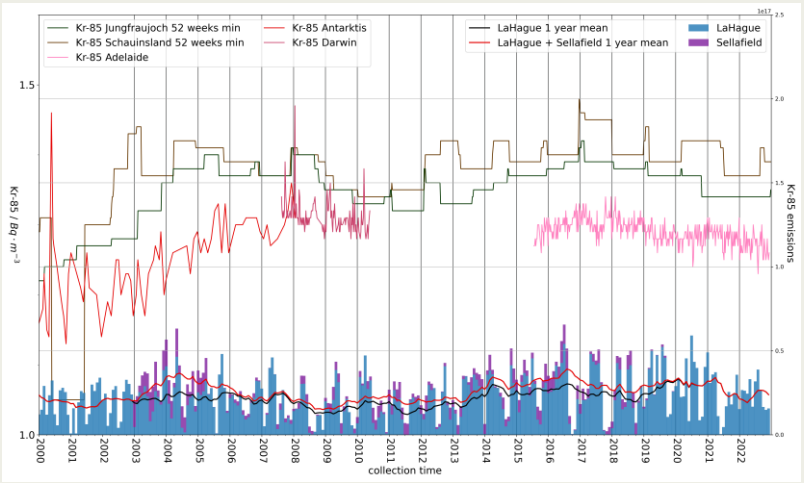
Baseline – closer look at „steady state“

A closer look at the supposedly constant level since 2005 reveals slight trends (2005 - 2011: ↘, 2011 - 2017: ↗, since 2017 ↘) and shows a correlation with the emissions from La Hague and Sellafield.



Atmospheric transport modelling

Backward simulation for weekly samples jan 05 - dec 24
For Freiburg, Schauinsland, Trier, Braunschweig, Jungfraujoch (CH)
Planned for Madrid, Offenbach, Hamburg, Guernsey

ATM specification (480 000 particles):

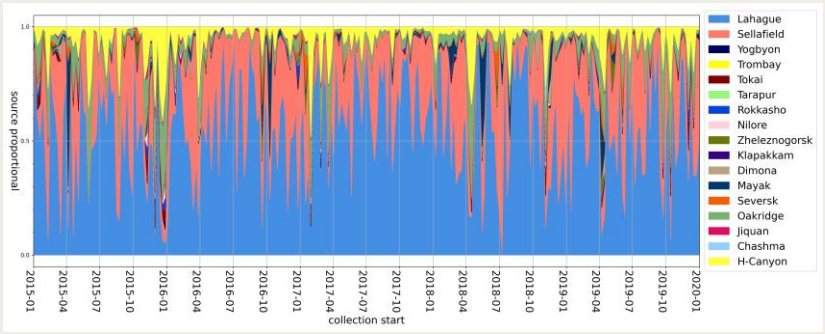
model:	HYSPLIT v5.3.3 (NOAA-ARL)
meteorological data:	GDAS 1°
simulated time:	480 h
grid resolution:	0.5° x 0.5°
vertical layer:	0-500 m AGL
output interval:	6 h

Possible sources of Kr-85

- Natural (traces): Kr-84 + cosmic ray
- Anthropogenic:
  - NPP (accidents)
  - Nuclear explosions
  - Reprocessing of nuclear fuel rods, production of Pu-239. There are 18 known reprocessing facilities in the northern hemisphere.

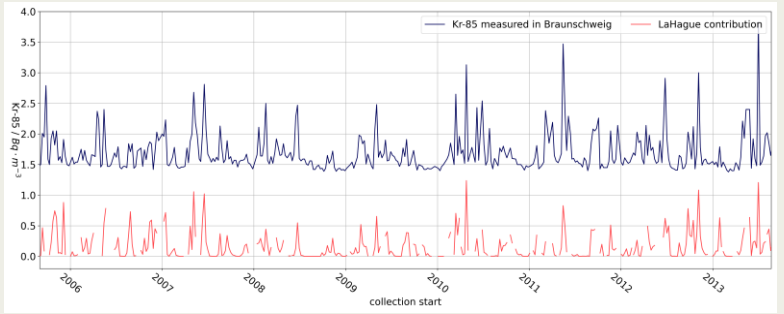
Source receptor sensitivities (SRS)

SRS of 5 stations and the 18 considered Kr-85 emitters were extracted. The origin of the air masses is calculated proportionally. Here we show – as an example – the station in Freiburg between 2015 and 2020: The vast amount of Kr-85 measured at this station is from La Hague (blue) and Sellafield (salmon). There is also a significant contribution from potential emitters in Northern America.



Simulation of La Hague contribution

High resolution emission data was provided by ORANO/ La Hague for 2006-2013. Together with the SRS, the expected contribution of the reprocessing plant to the weekly measured activity concentration can be simulated:



Summary & outlook

- Vast amount of Krypton-85 measurements
- Baseline + peaks to be considered separately
- 20 years of ATM for 5 stations (more coming)
- Known background + high resolution emission data used for Krypton simulation
- Is the identification of a new reprocessing facility possible?
- Kr inventory
- Information from occasional sampling (Guernsey)
- Effects of different time resolutions of emission data (monthly, weekly, hourly)
- Use of global model to estimate global emissions?

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