# Half a century of atmospheric krypton-85 measurements

**Introduction** – sources and uses of atmospheric $^{85}Kr$
**The BfS Lab** – sampling and measurement of $^{85}Kr$
**Results** - Central Europe and the Southern Hemisphere
**Conclusions** - Data availability and Outlook

# History

**02.08.1939**: Start of “Manhattan Project”, Findings classified as “secret”

**02.12. 1942**: First critical reactor in Chicago (CP-1)

**1944**: Douglas A-26 airplanes from the US Air Force sample air above Europe in search of **radioxenon** of German nuclear reactors/weapons programme

**16.07.1945**: First nuclear weapon test

**Aug 1945**: Hiroshima & Nagasaki

**Late 40s**: US & British intelligence program to detect & monitor nuclear operations and estimate Pu stock pile of foreign countries (“global” **$^{85}$Kr** monitoring program)

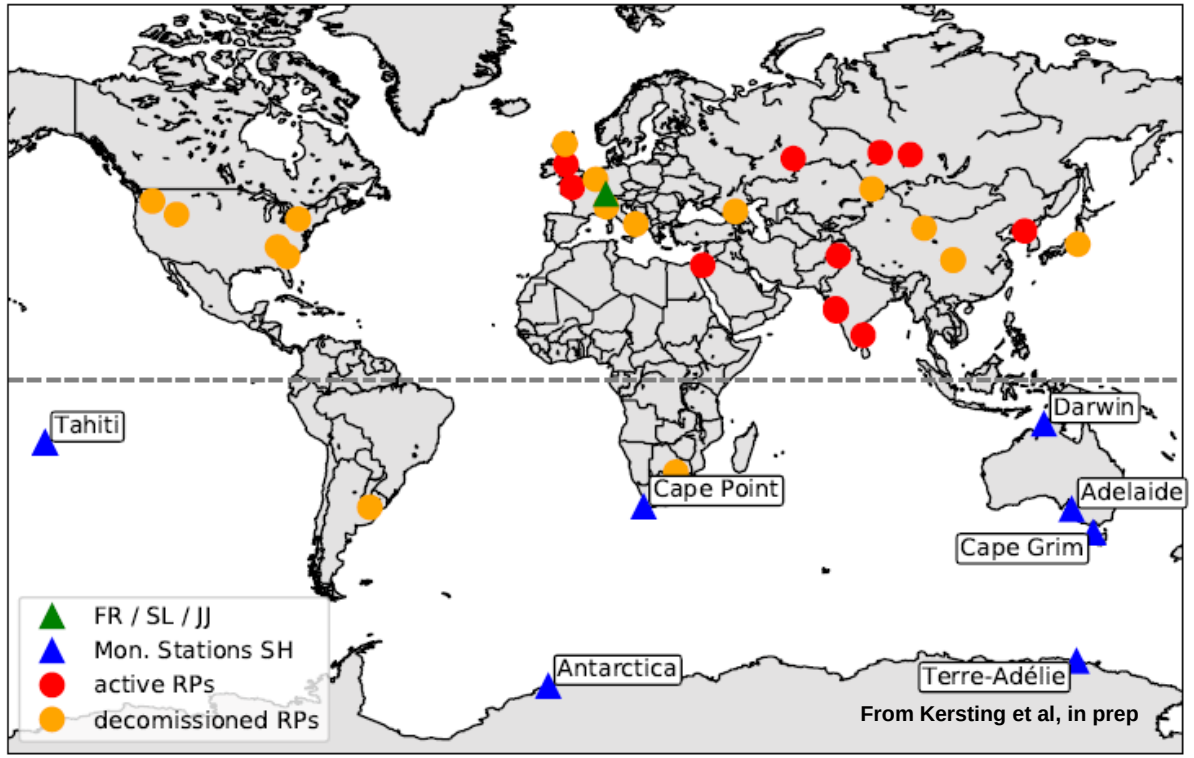
**Early 60s**: start of measurements ($^{85}$Kr and $^{133}$Xe) in Germany, continuous since 1972



# $^{85}$Kr sources

**$^{85}$Kr: $T_{1/2}$=10.74 a**

**Main source: Nuclear Reprocessing (20*10$^{12}$ Bq $^{85}$Kr/kg weapon grade Pu)**



# Sampling Sites (BfS network)

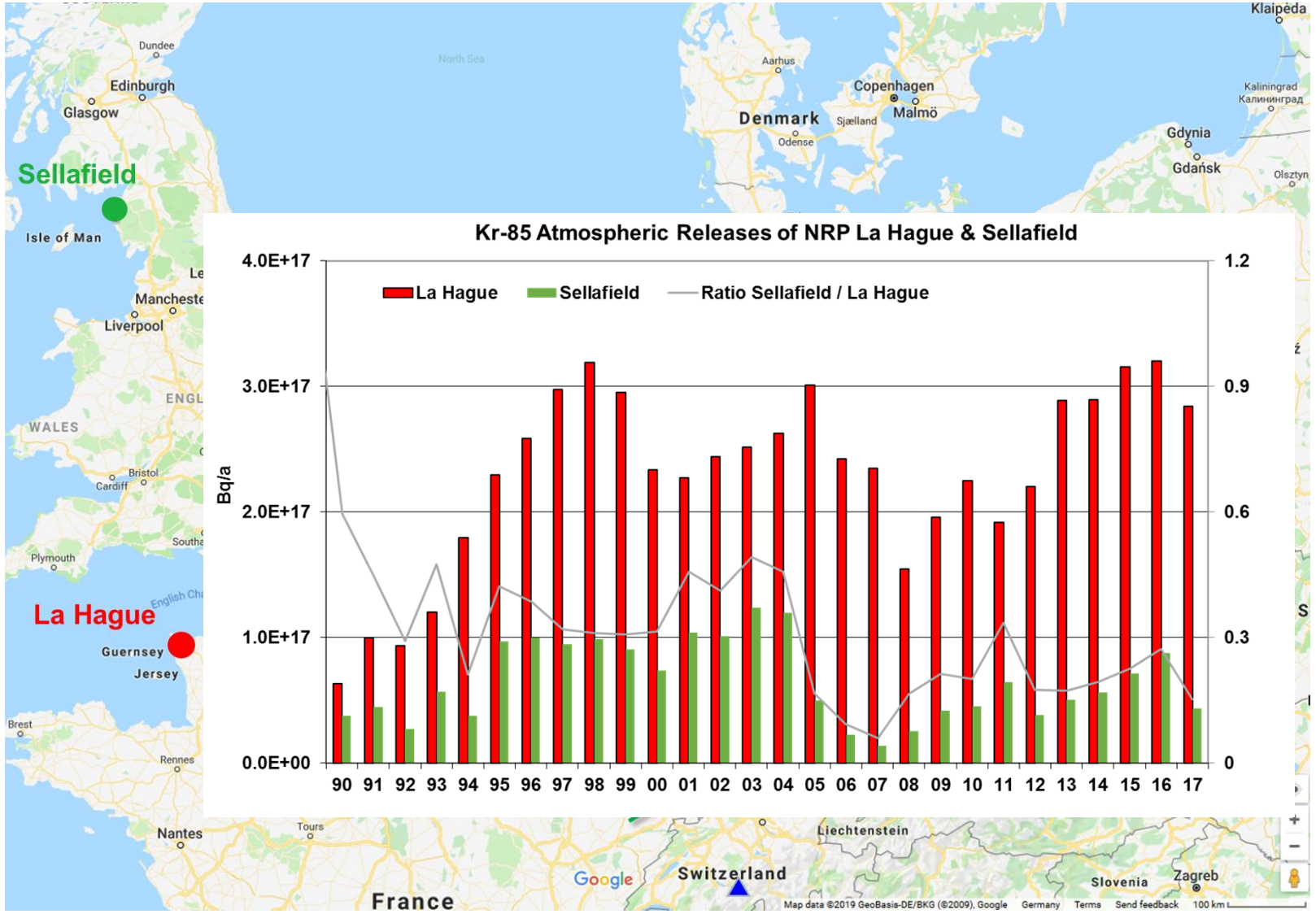
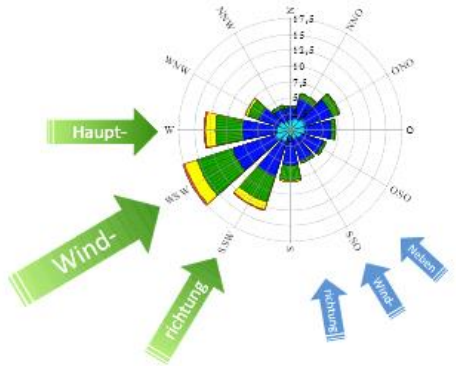


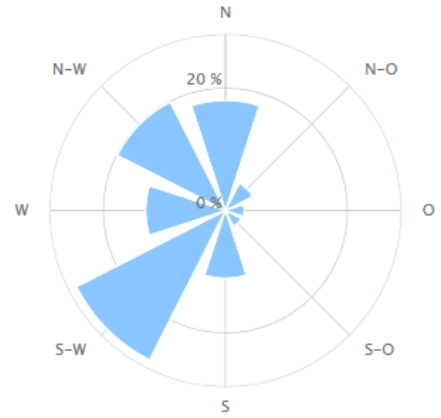




**Table 1**
Selected sampling locations, sampling periods and number of samples collected.

| Site | Location | | Sampling | | N |
|---|---|---|---|---|---|
| | Lat. | Long. | from | to | |
| Freiburg | 47.998 N | 7.846 E | 06/1973 | 01/2019 + | 2309 |
| Schauinsland | 47.915 N | 7.907 E | 02/1976 | 01/2019 + | 2182 |
| Jungfraujoch (CH) | 46.545 N | 7.987 E | 01/1990 | 01/2019 + | 1467 |
| Braunschweig | 52.296 N | 10.463 E | 10/1987 | 01/2019 + | 1554 |
| Offenbach | 50.102 N | 8.746 E | 01/1996 | 01/2019 + | 1171 |
| Berlin | 52.533 N | 13.417 E | 04/1997 | 04/2010 | 658 |
| Perl | 49.467 N | 6.397 E | 08/1989 | 10/2012 | 1067 |
| Crakow (POL) | 50.050 N | 19.917 E | 07/1981 | 04/1988 | 275 |
| Trier | 49.748 N | 6.657 E | 02/2013 | 01/2019 + | 303 |
| Potsdam | 52.382 N | 13.063 E | 05/2010 | 01/2019 + | 445 |
| Hamburg | 53.550 N | 9.983 E | 02/2008 | 01/2019 + | 545 |



# Sampling

**Manual system (1973): Measurement of atmospheric activity concentrations of $^{85}Kr$ (and $^{133}Xe$ )**

**World wide network and central lab in Freiburg**

**Cryogenic sampling**

**GC for processing & purification of NG**

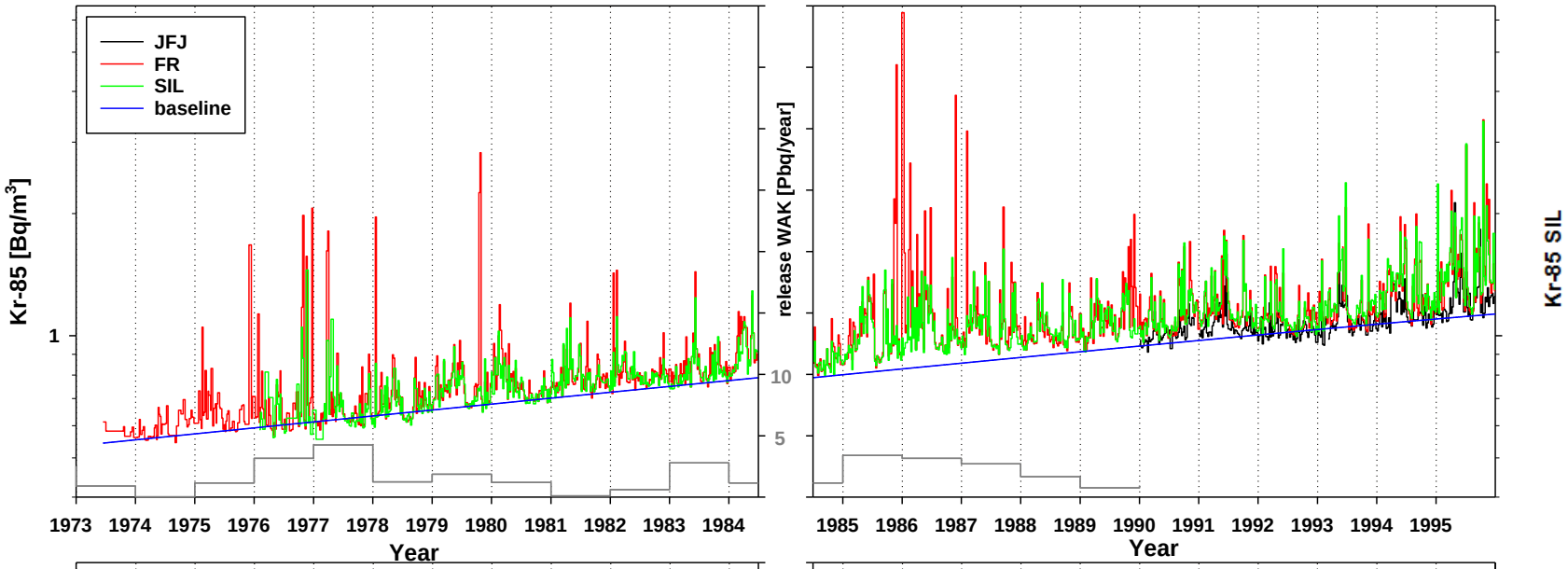
# Measurement

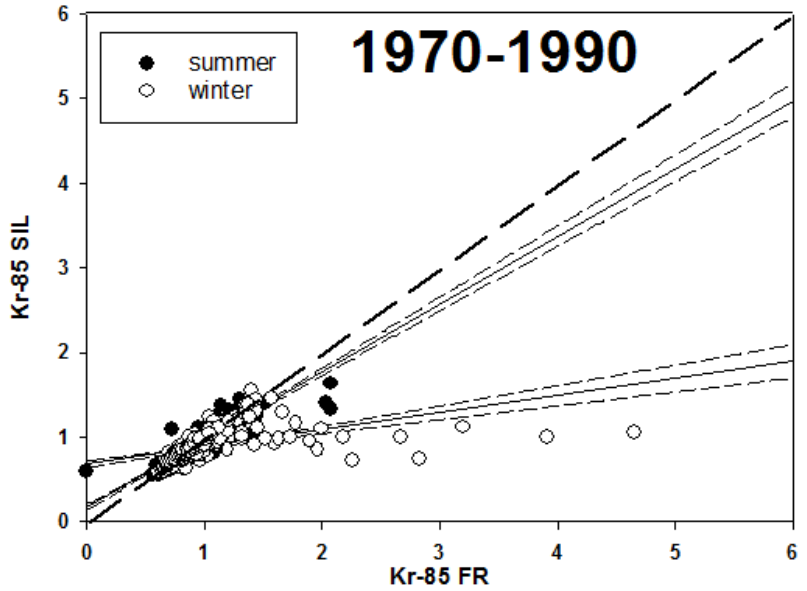
**8 proportional gas counters, Pb-shielding & anticoincidence counters**

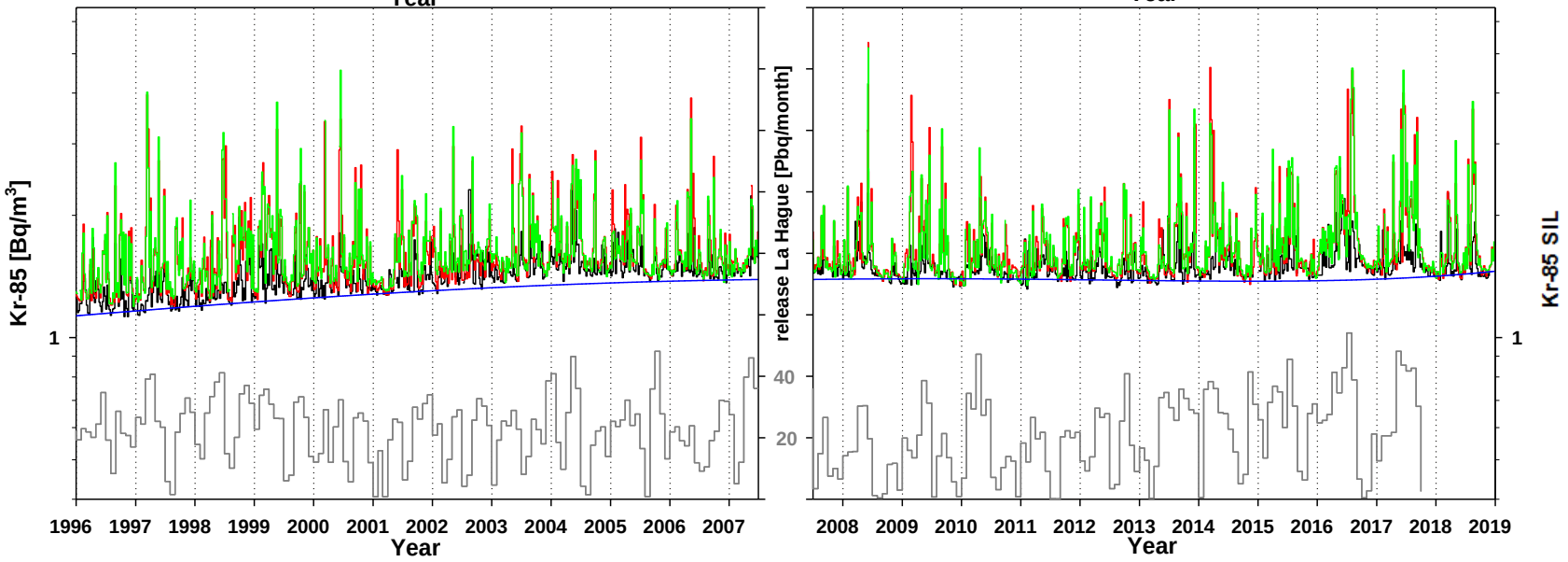
**MDA for $^{85}Kr$: 30 mBq (1 m³ air)**

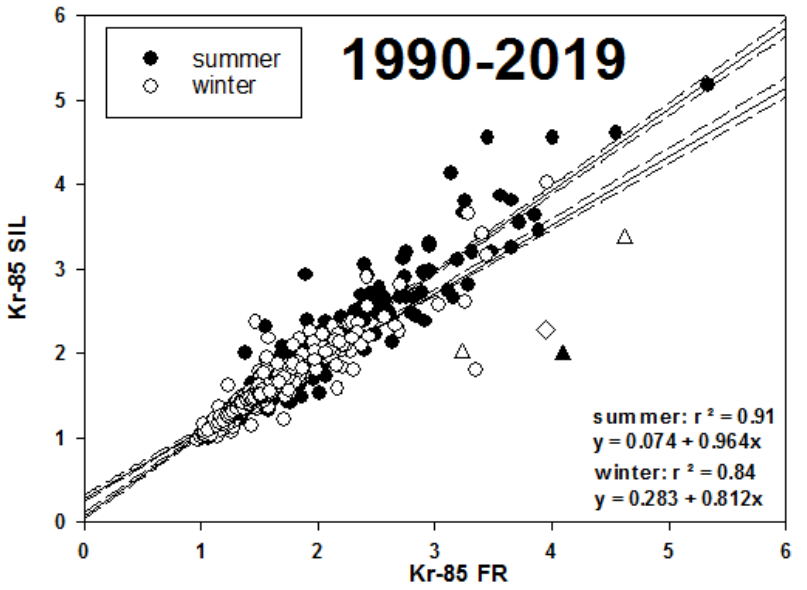
# Results – Freiburg, Schauinsland, Jungfraujoch

230 m asl 1205 m asl 3454 m asl

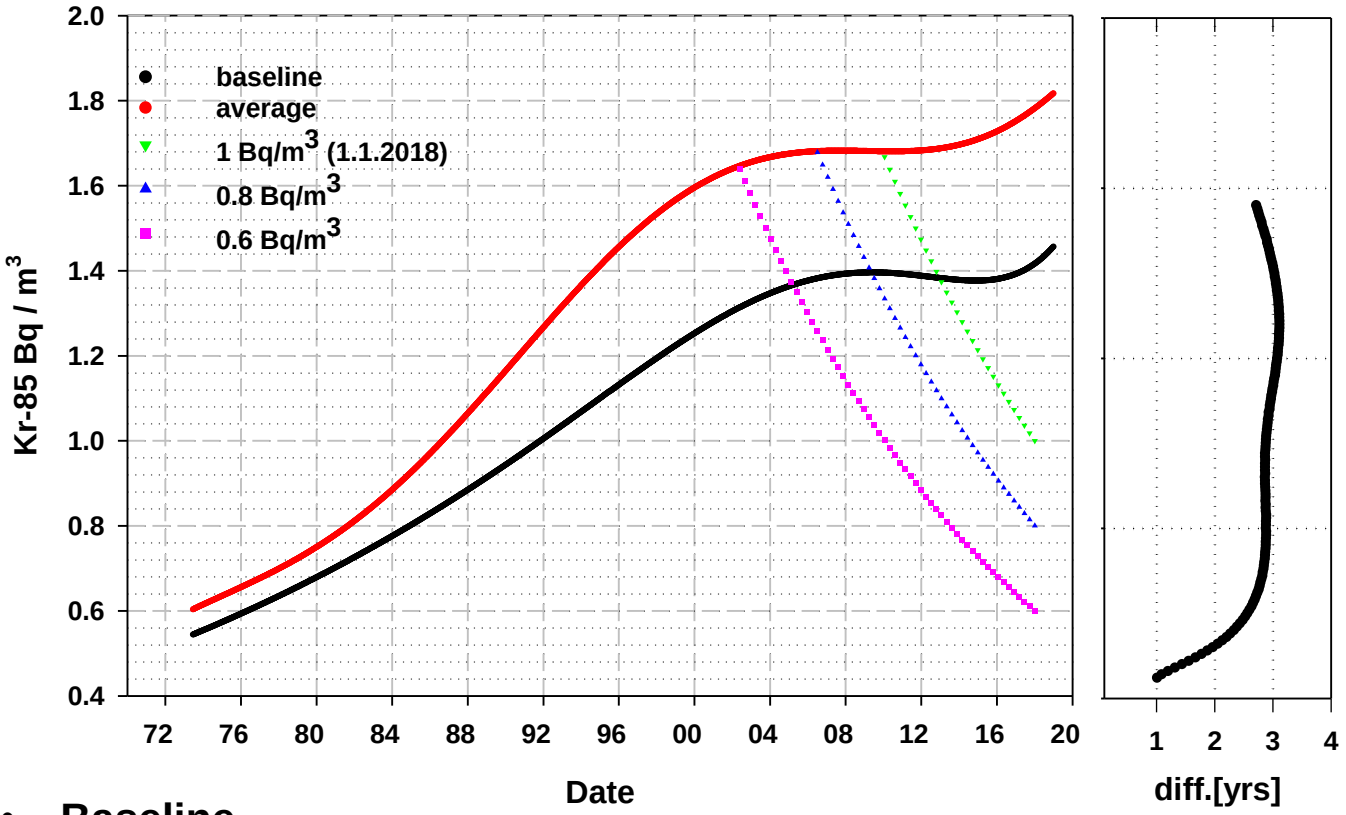


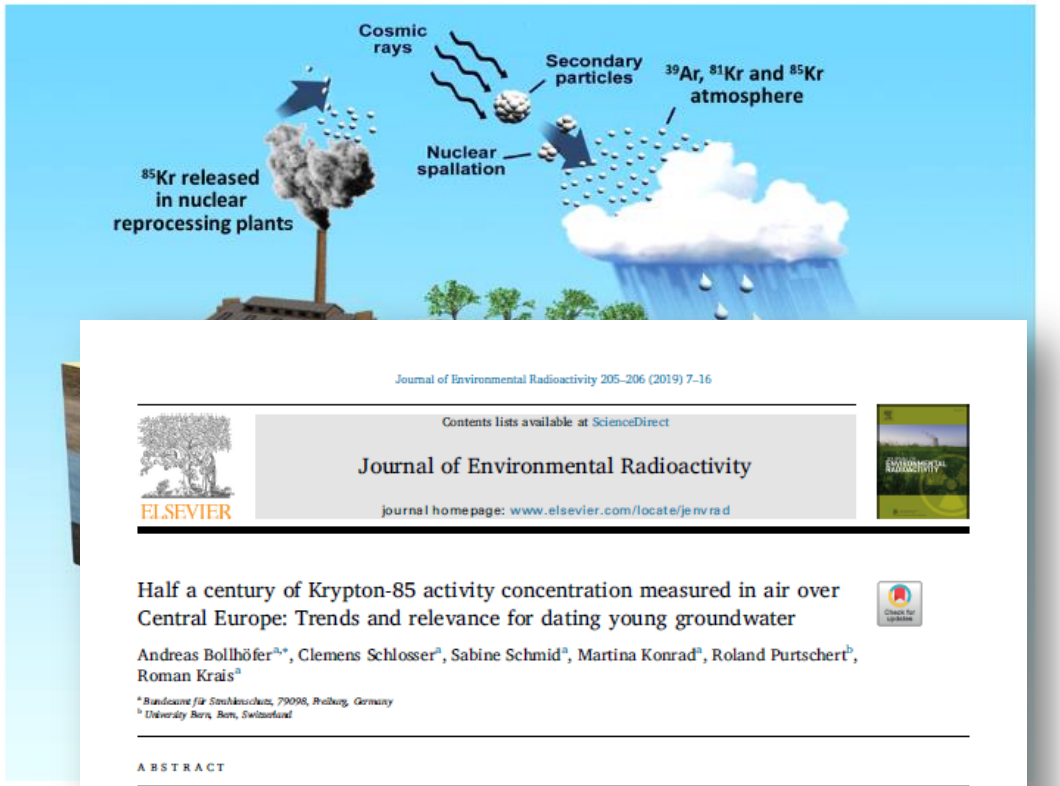



# Results – Freiburg, Schauinsland, Jungfraujoch




Picture modified from C.E. Aalseth, 2015



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Half a century of Krypton-85 activity concentration measured in air over Central Europe: Trends and relevance for dating young groundwater

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ABSTRACT

For almost half a century weekly samples for the measurement of krypton-85 ($^{85}$Kr) activity concentrations in surface air have been collected by the Bundesamt für Strahlenschutz (BfS), Germany. Sampling started at Freiburg (230m asl) in 1973, Mt Schauinsland (1205m asl) in 1976 and Mt Jungfraujoch in Switzerland (3454 asl) in 1990. Distinct maxima in the time series of atmospheric $^{85}$Kr activity concentration are caused by emissions from nuclear reprocessing plants in Europe, mainly the La Hague, France, and Sellafield, UK, reprocessing plants. Between 1970 and 1990 peak activity concentrations measured in winter along the Rhine Rift in Freiburg are often higher than at Mt Schauinsland, due to emissions from the operating pilot reprocessing plant in Karlsruhe - approximately 130 km to the north - and large-scale inversions that inhibit exchange of air masses within the Rhine Rift with those at higher altitudes. From the early 1990s onwards, after the shut-down of the pilot plant, differences between Freiburg and Schauinsland are much smaller. Activity concentrations measured at Jungfraujoch are generally lower and close to baseline levels, due to its location in the free troposphere. Weekly baseline and average $^{85}$Kr activity concentration in the atmosphere in Central Europe were modelled from almost 12,000 individual measurements at 11 stations. The baseline and average have continuously increased, interrupted by a relatively stable period between 2009 and the end of 2014 with a baseline activity concentration of about 1.39 Bq/m$^3$. Depending on the geographical location and hydrological conditions, the modelled baseline or average $^{85}$Kr activity concentration time series can be used as input functions for the dating of young groundwater.

- **Baseline**
  - **quarterly minima or average all Central Europe stations**
  - **Poly-fit, 7th order**
- **Input function for dating groundwater**
  - **Simple piston flow model**
  - **Age difference: ~ 3 years**

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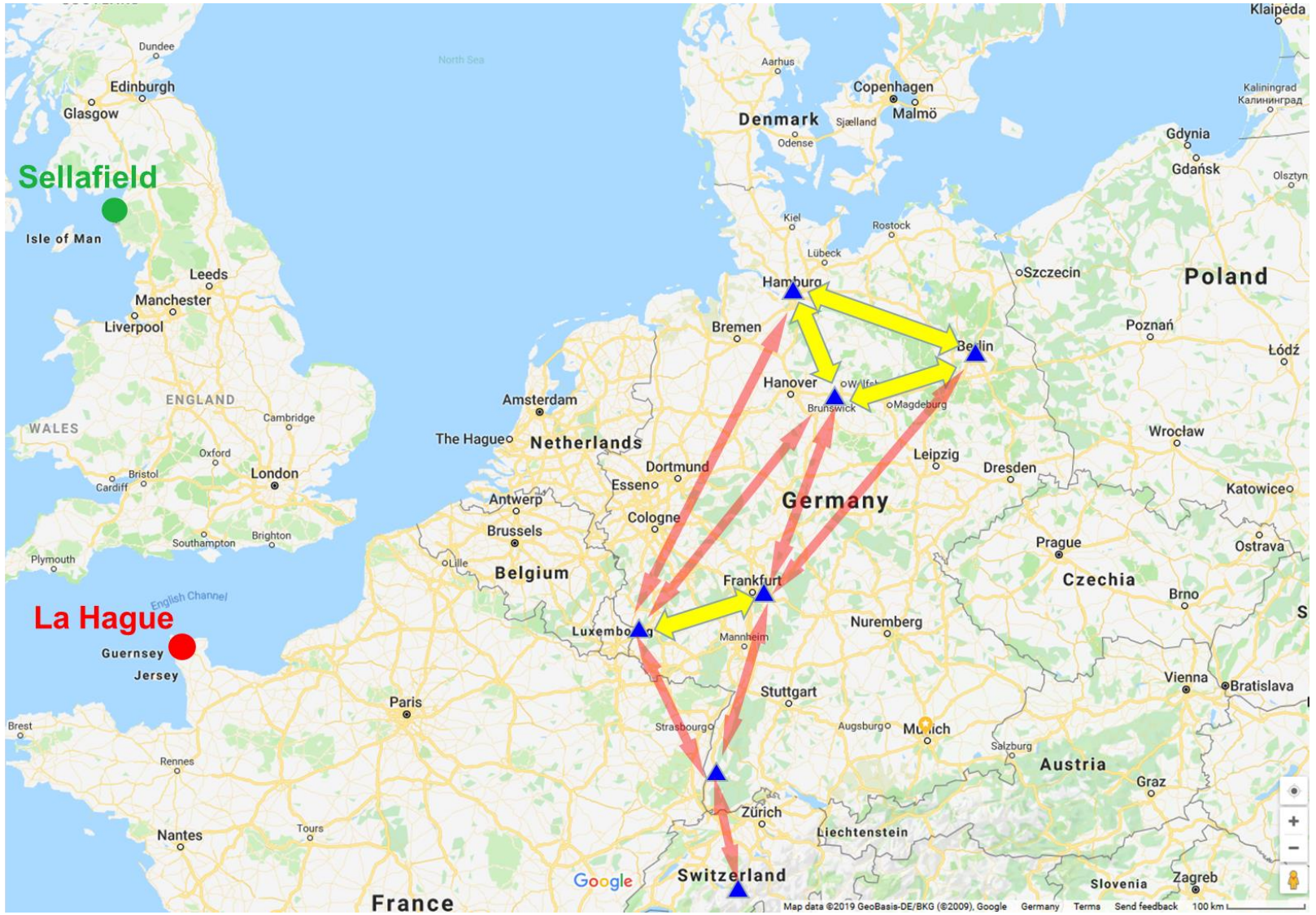
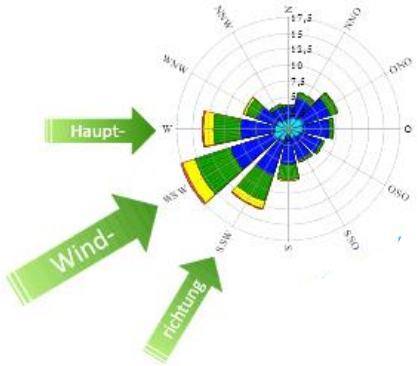
# Results - Central Europe




“high“ correlation ($R^2$ > 0.5-0.8)

“low“ correlation ($R^2$ < 0.5)

**Dominance of La Hague from 2005 reflected in better correlation between sites**



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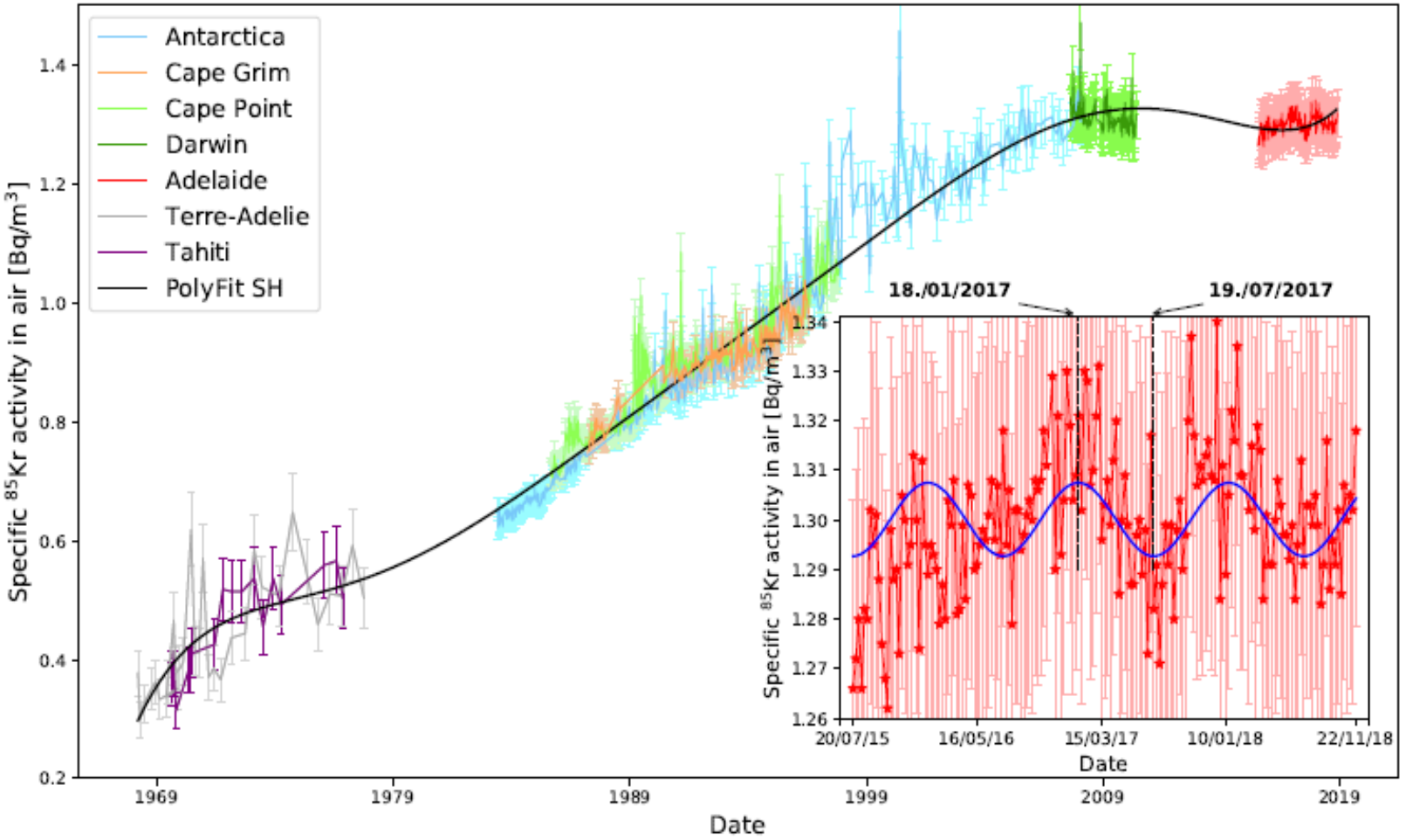
# Results – Southern Hemisphere

## Southern Hemisphere (Kersting et al in prep)




- **Interhemispheric exchange**

$$c_{Kr}^{SH}(t) = c_{Kr}^{NH}(t - \tau_{ex}) \cdot exp(-\tau_{ex} \cdot \lambda_{Kr})$$

  - **Extrapolate NH baseline**
  - **Minimize $\chi^2$ fit of extrapolated NH baseline to SH data**

**$\tau_{ex}$ = 1.2 years** ($\chi^2$= 2.42)

- **Annual variability**
  - **movement of the Hadley Cell**
  - **ENSO cycle**



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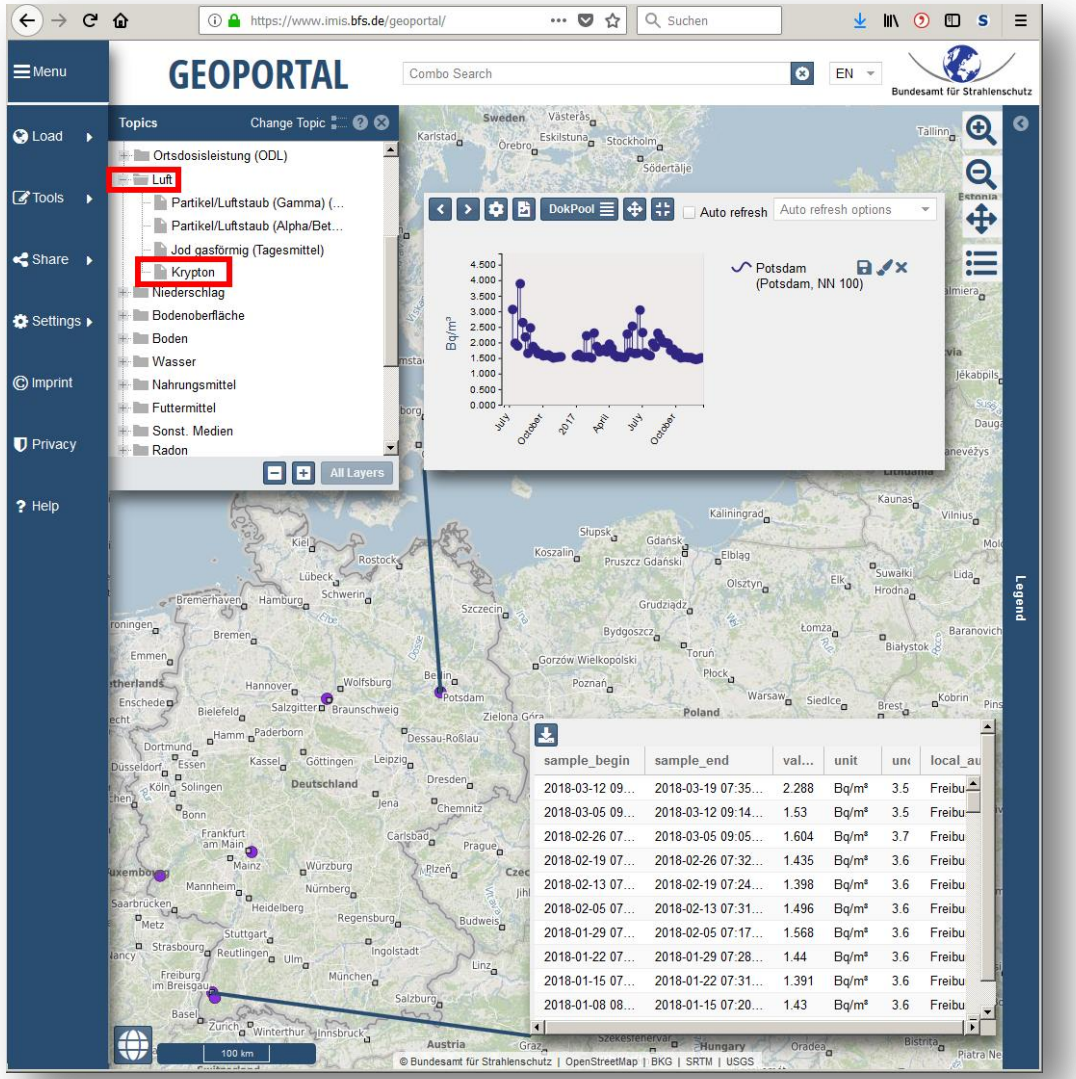
# Data-sharing and Outlook




**Freiburg, Schauinsland, Jungfraujoch and baseline/average available on:**
https://www.sciencedirect.com/science/article/pii/S0265931X19300888?via%3Dihub

**These data will be useful for dating groundwater**

**Central European data will be made available (and regularly updated) soon on BfS GEOPORTAL:**
https://www.imis.bfs.de/geoportal/

**ATTA development: precision ↑ sample size ↓ will help continue time-series and allow for additional groundwater applications/studies**

**Despite 'predictability' of SH baseline, monitoring still required for verification purposes**



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