Identifying civil Xe-emissions: from source to receptor

A. Bollhöfer[1], P. De Meutter[2,3,4], F. Gubernator[5], Benoît Deconninck[6], C. Schlosser[1], U. Stöhlker[1], G. Kirchner[5], C. Strobl[1], A. Delcloo[3], J. Camps[2]

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[4] Ghent University, B-9000 Ghent, Belgium
[5] University Hamburg, Carl Friedrich von Weizsäcker-Centre for Science and Peace Research (ZNF)
[6] Institute for Radioelements, Fleurus, Belgium

- Environmental Monitoring
- Atmospheric Transport Modelling
- Stack monitoring

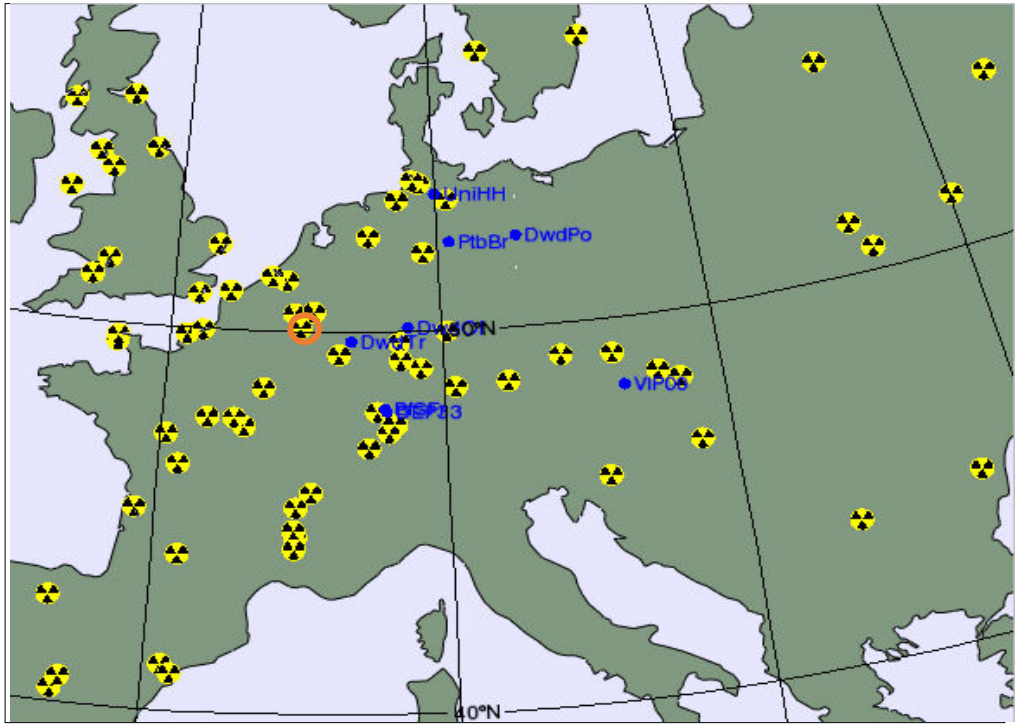
The International Monitoring System

Sources of radioxenons

Radioxenons are fission products:

- **Nuclear explosions**
- **Nuclear power plants and research reactors**
- **Medical isotope production (^{99}Mo/^{99}Tc; ^{131}I)**

Nuclide	$T_{1/2}$
^{131m}Xe	11.84 d
^{133}Xe	5.25 d
^{133m}Xe	2.19 d
^{135}Xe	9.10 h

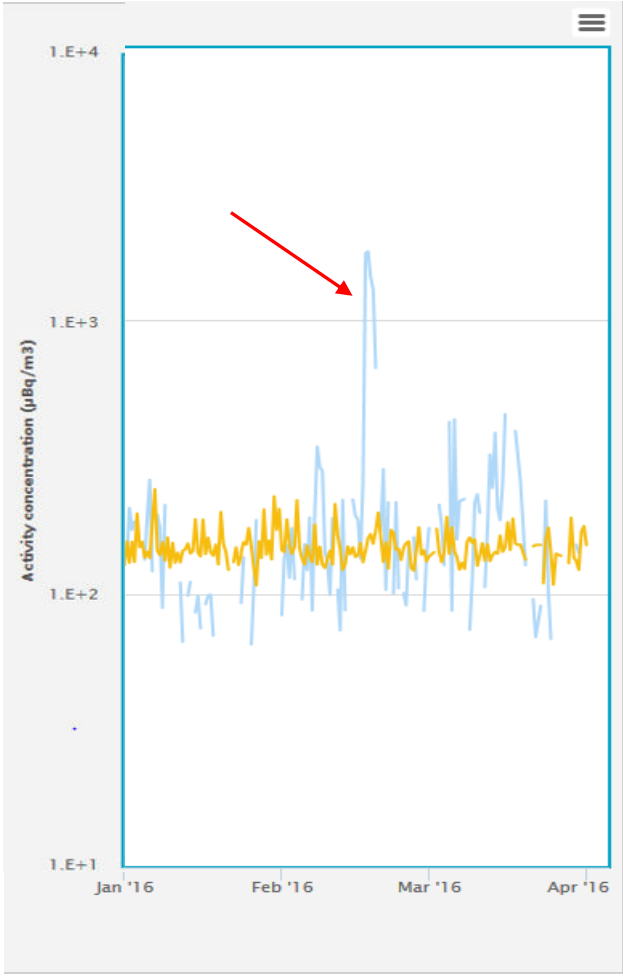


Underground test (#4), DPRK, Jan 2016

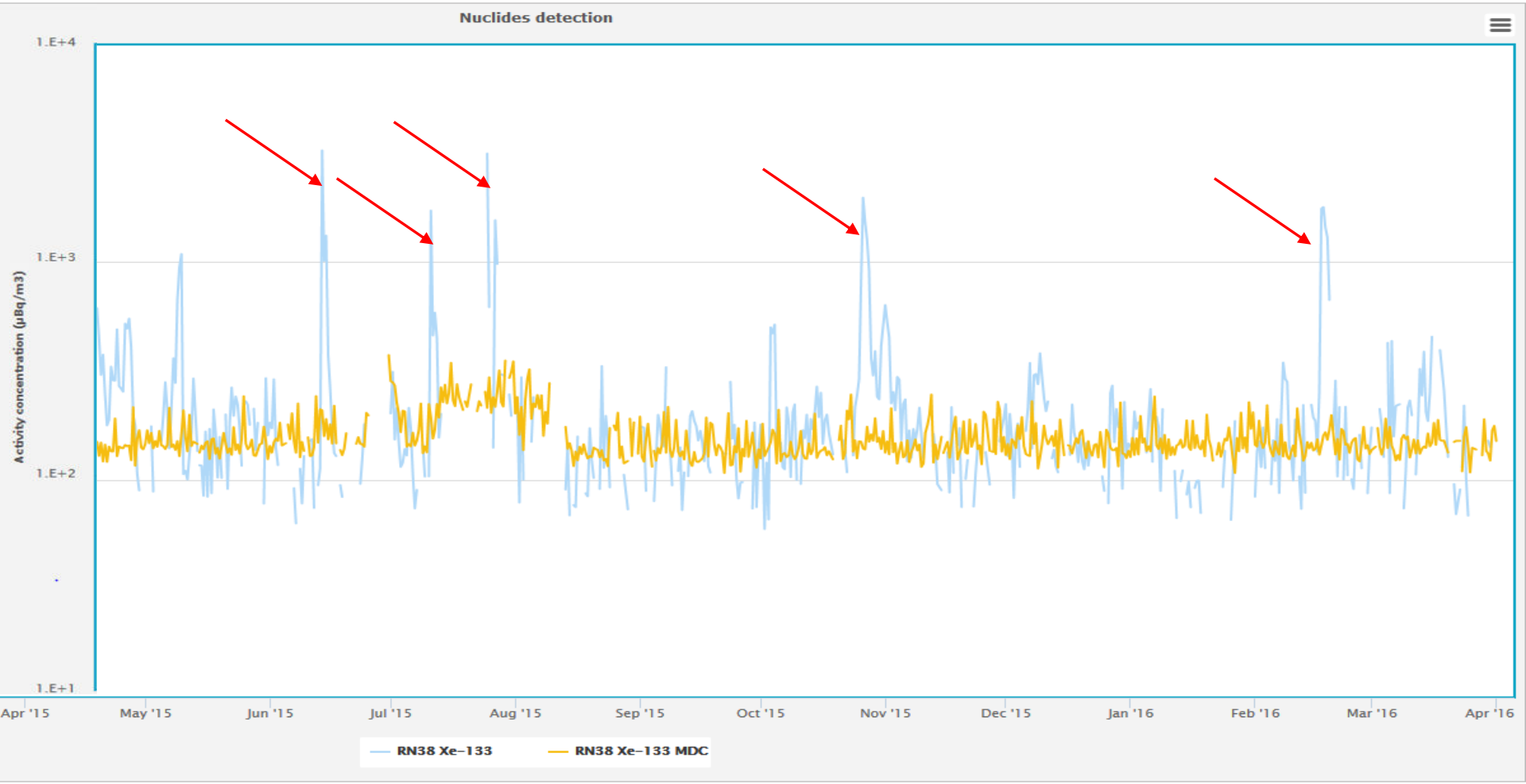
- **Successful nuclear weapons test announced, 6 Jan 2016**

- **"Waiting" for the smoking gun**

- **From Feb 17 elevated Xe-133 activity (2-3 days) at station Takasaki, JP38, but no other isotopes > MDC**



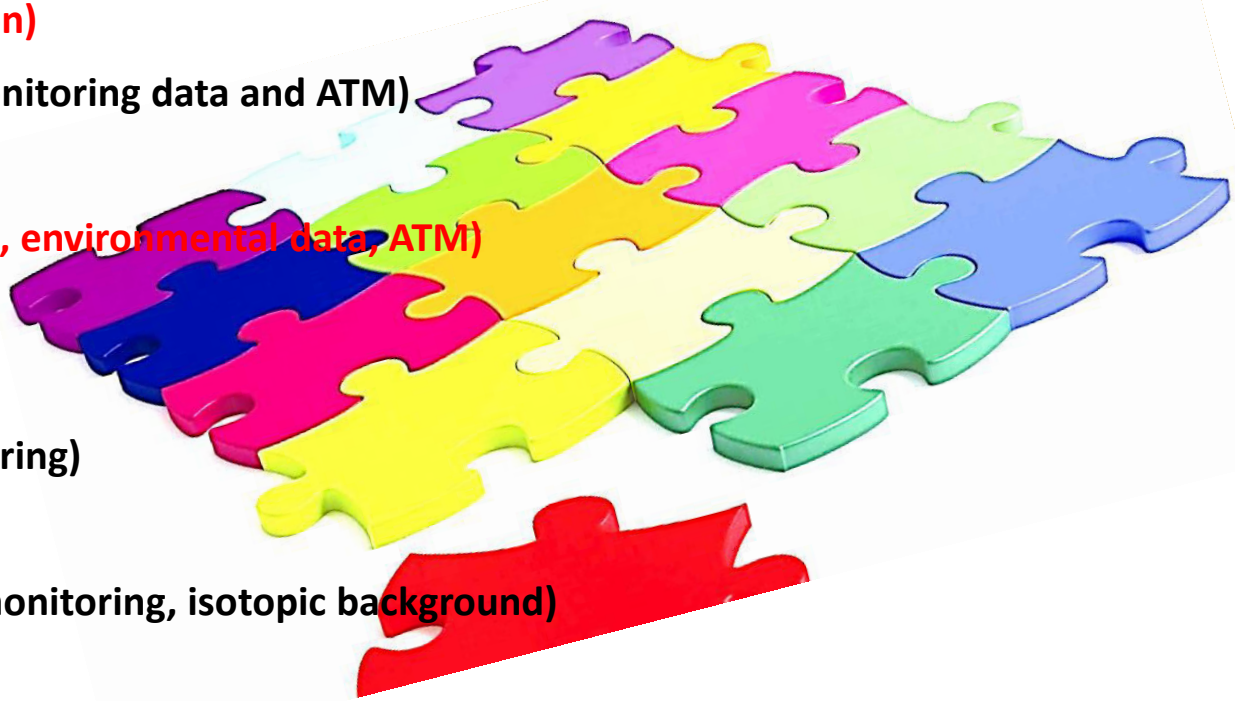
Underground test (#4), DPRK, Jan 2016



IAEA - Project CRP F23031

Sharing and Developing Protocols to Further Minimize Radioactive Gaseous Releases to the Environment in the Manufacture of Medical Radioisotopes, as Good Manufacturing Practice

- **Belgium (ATM, Xe-mitigation)**
- **Canada (CNL emissions, monitoring data and ATM)**
- **Egypt (Xe-mitigation)**
- **Germany (stack monitoring, environmental data, ATM)**
- **Indonesia (Xe-mitigation)**
- **Korea (Xe-mitigation)**
- **Pakistan (emissions monitoring)**
- **Poland (Xe-mitigation)**
- **USA (Xe mitigation, stack monitoring, isotopic background)**



Stack and near field monitoring

1) **Masters**' University Hamburg, BfS

Development and test of a compact spectrometric stack monitoring system

- Characterise emissions
  -> compare with IRE System

2) Investigate sensitivity of compact spectrometric systems ($LaBr_3$, CdZT) in the vicinity of IRE, Fleurus

- Xe-environmental monitoring
- Comparison with Belgian Telerad. Measurement campaign Q3 2017

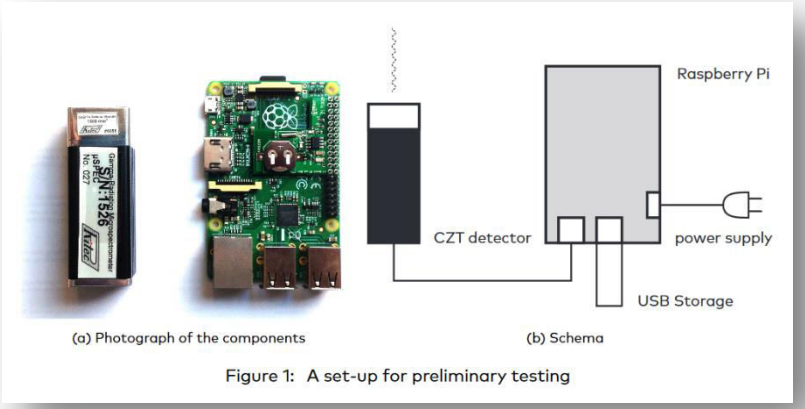


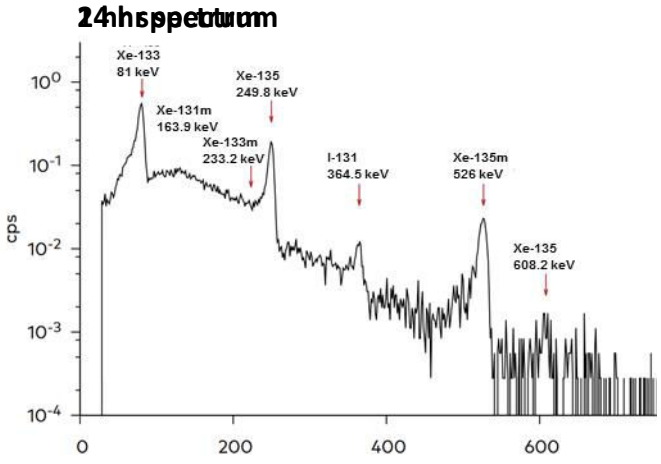
Figure 1: A set-up for preliminary testing

Cadmium Zinc Telluride detector

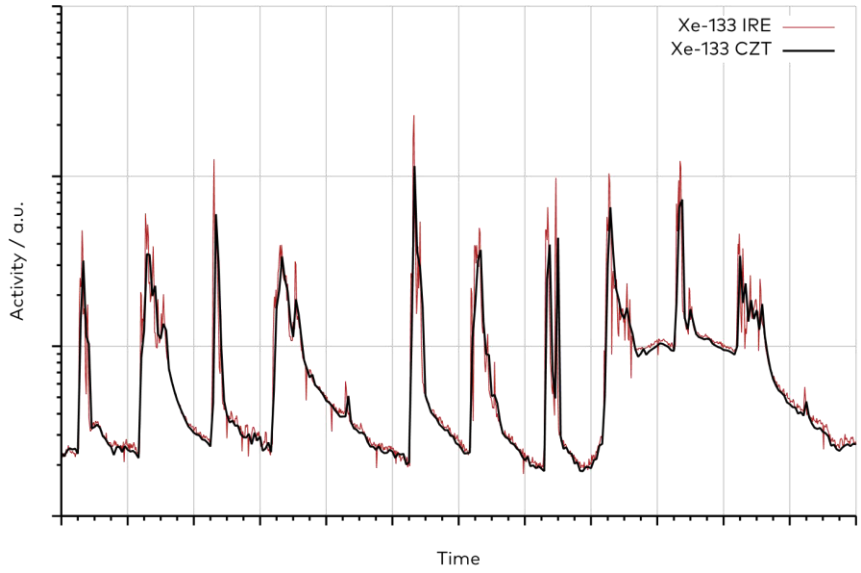
- Intergated electronics
- Compact
- Low energy usage
- FWHM ~2%, 662keV

Results: Stack monitoring IRE, Fleurus

Example spectrum IRE first campaign (day 1 + day 2)



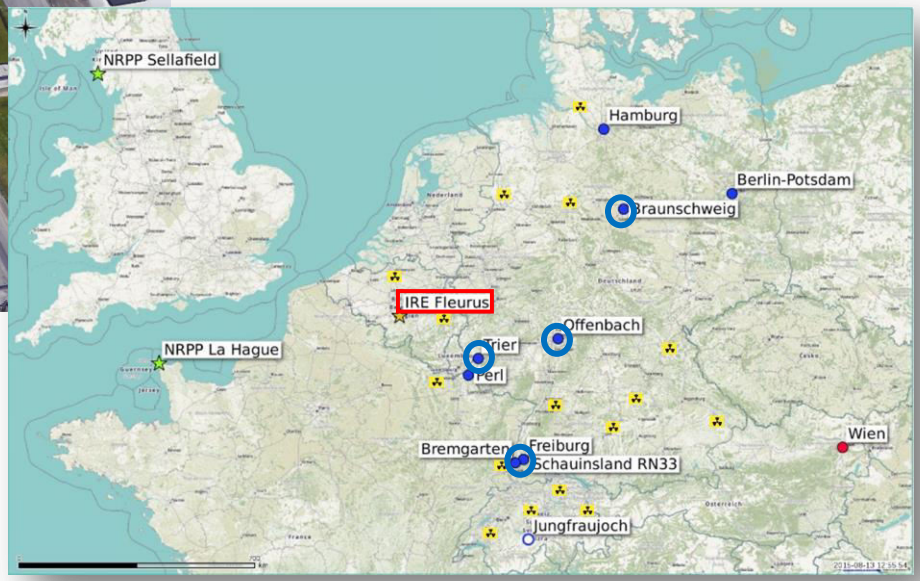
Nuclide	$T_{1/2}$	E_γ (P_γ)
^{131m}Xe	11.8 d	163.9 keV (1.95%)
^{133}Xe	5.25 d	81 keV (36.6%)
^{133m}Xe	2.19 d	233.2 keV (10.1%)
^{135}Xe	9.10 h	249.8 keV (90%)
^{135m}Xe	15.3 m	526.6 keV (80.5%)



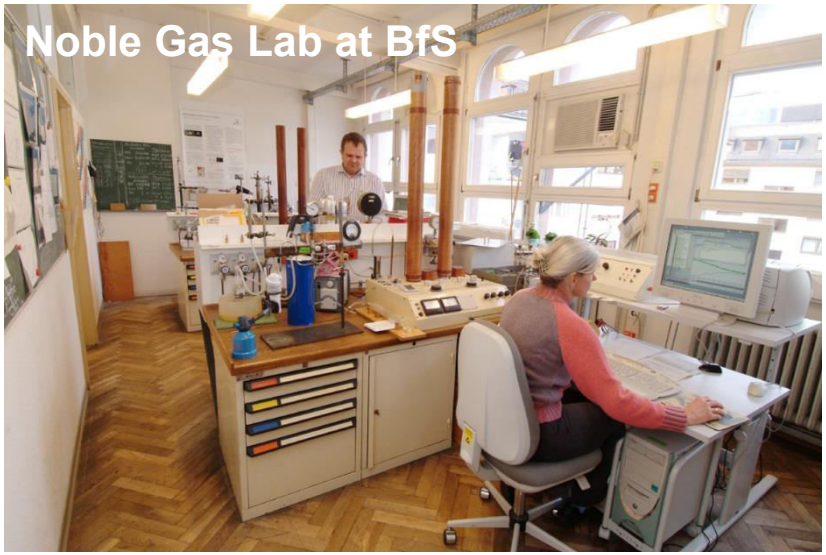
Conclusions: Stack and near field monitoring

- **Set-up of a CZT detector and a Raspberry Pi**
  - **Simple**
  - **Mobile**
  - **Small**
  - **Cheap**
- **Capable of detecting Xe-133, Xe-135, Xe-135m**
- **Detection Xe-131m, Xe-133m needs further development**
- **Preliminary data (800m N-E of IRE) indicate Xe-133 and Xe-135 easily detectable in the near field ($LaBr_3$)**

Comparison: Environmental data - ATM

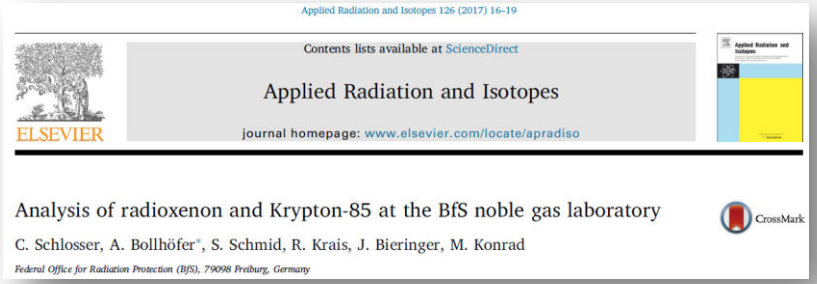


Xenon measurements in the environment



Manual system: ^{85}Kr & ^{133}Xe

- Cryogenic sampling (1 week, 10 m³ air)
- GC for processing and purification
- 7 proportional gas counters + anticoincidence
- MDA for ^{133}Xe: 0,01 Bq (2 m³ air)


Applied Radiation and Isotopes 126 (2017) 16–19

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journal homepage: www.elsevier.com/locate/apradiso

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Analysis of radioxenon and Krypton-85 at the BfS noble gas laboratory

CrossMark

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Federal Office for Radiation Protection (BfS), 79098 Freiburg, Germany

SAUNA Lab System: Xenons only

- Nuclide specific: ^{131m}Xe, ^{133}Xe, ^{133m}Xe, ^{135}Xe
- **β-γ coincidence detection system**
- MDAs (12 h meas.): 3 mBq

Atmospheric Transport Modelling

SCK-CEN, Ghent University, RMI Belgium: ATM as part of the collaboration

Stack monitoring data from IRE, Fleurus for 2014:

- 15 min Xe-133 emission data as model input

Meteorological data: ECMWF

- 3 hourly weather for 2014

Model: FLEXPART("FLEXible PARTicle dispersion model")

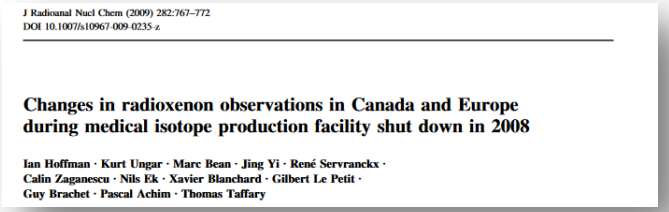
- Lagrange transport and dispersion model
- 0.5º horizontal resolution

Emissions NPP:

- Germany (quarterly), Switzerland (monthly),France, Belgium and Scandinavian countries (annually)

Emissions CNL:

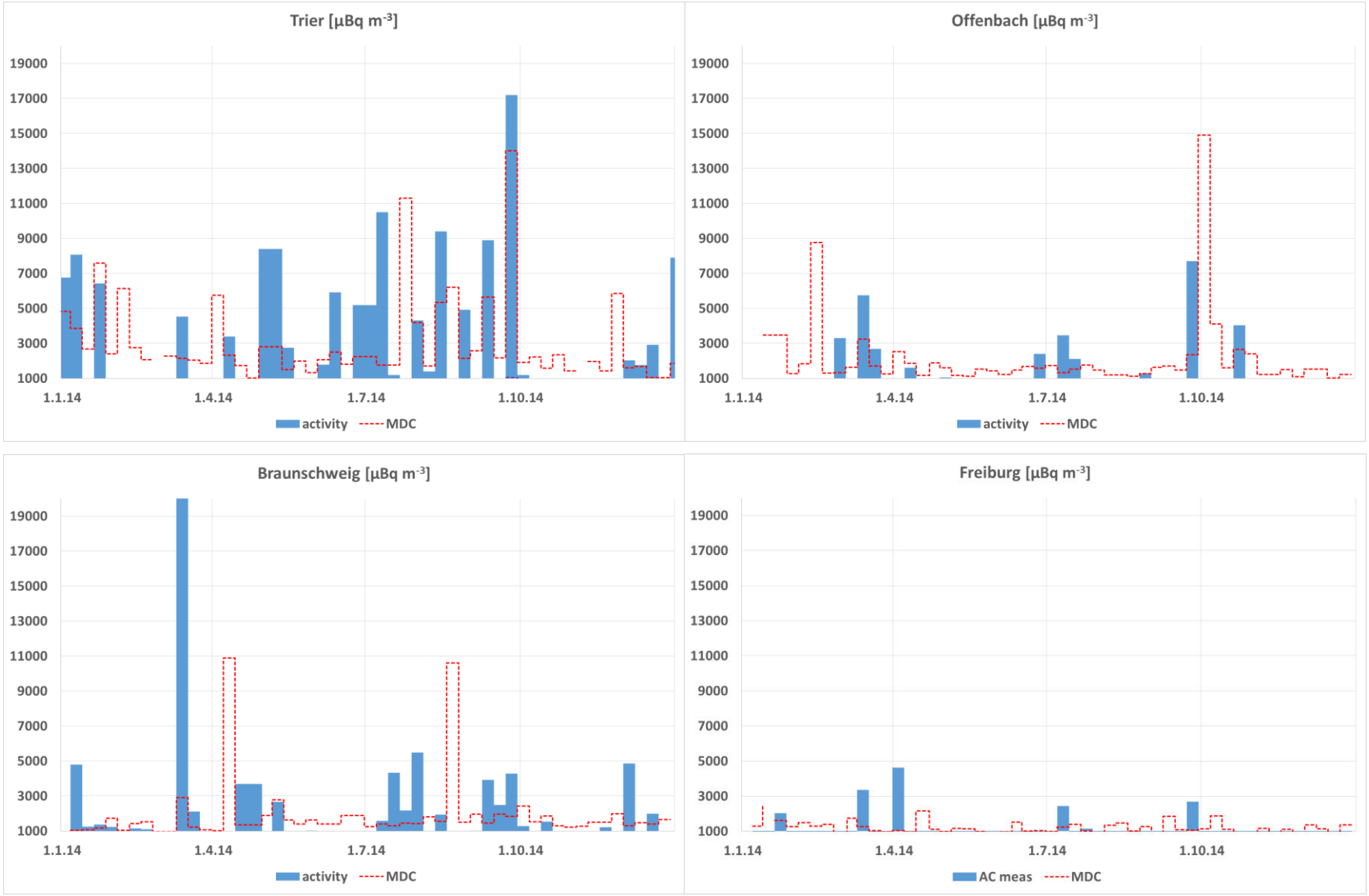
- Hoffman et al 2009


J Radioanal Nucl Chem (2009) 282:767–772
DOI 10.1007/s10967-009-0235-z

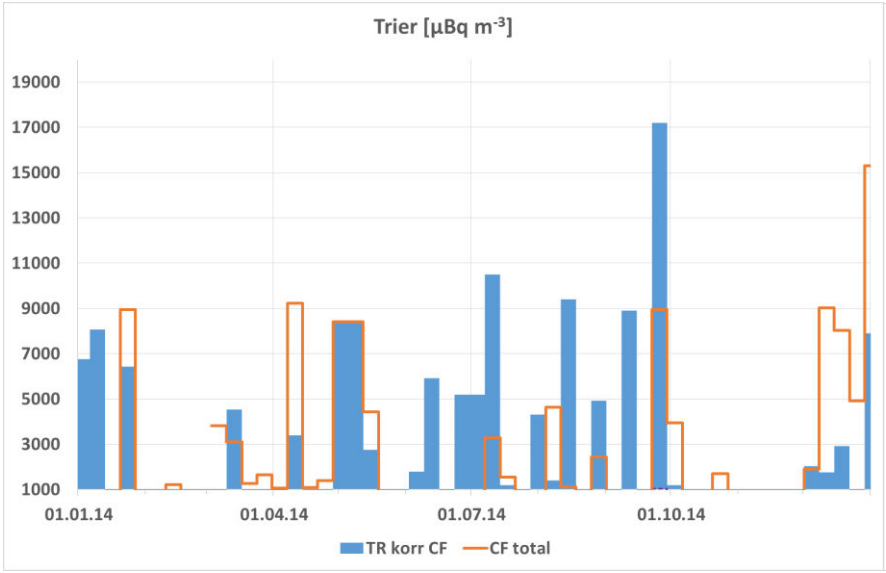
Changes in radioxenon observations in Canada and Europe during medical isotope production facility shut down in 2008

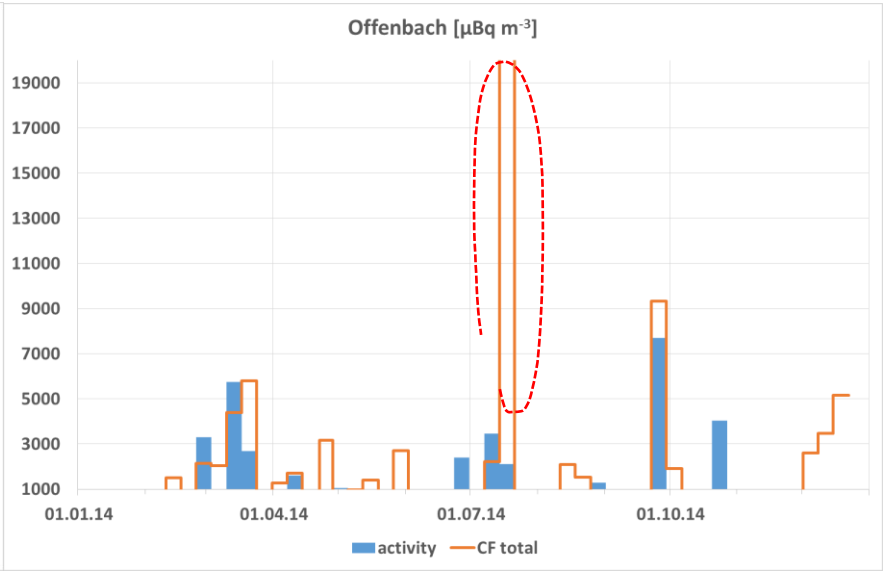
Ian Hoffman · Kurt Ungar · Marc Bean · Jing Yi · René Servranckx · Calin Zaganescu · Nils Ek · Xavier Blanchard · Gilbert Le Petit · Guy Brachet · Pascal Achim · Thomas Taffary

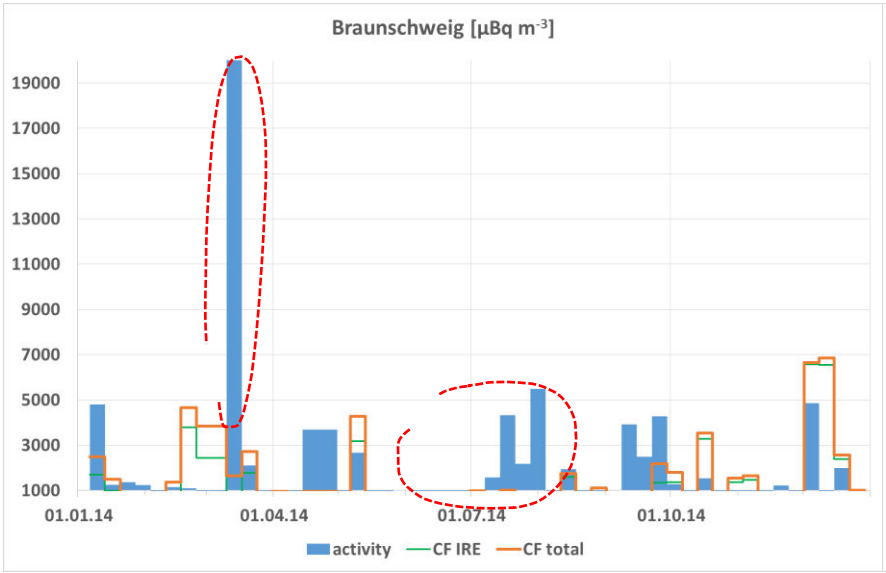
2014 (weekly samples)

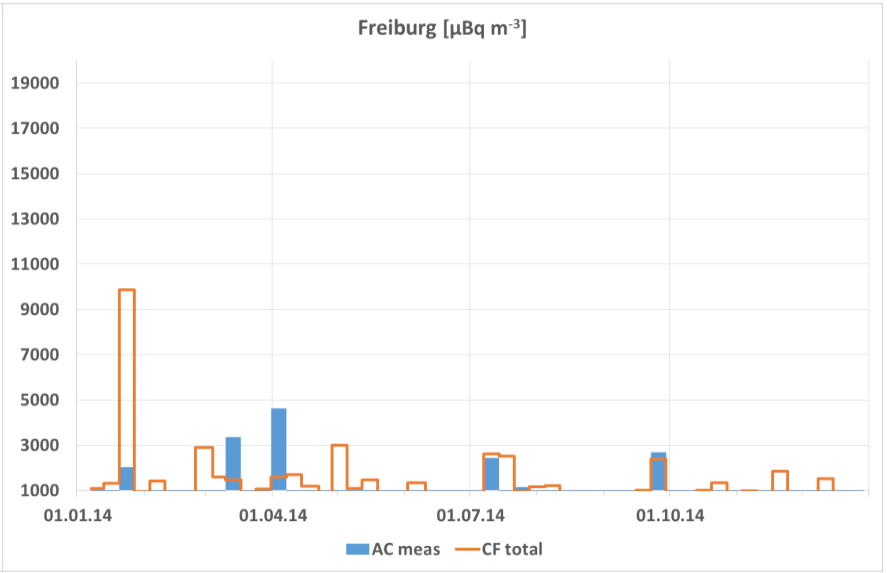


2014 (weekly samples)









Discussion & Future work

- **ATM reproduces general trends at the stations but:**
  - Periods with higher measured vs modelled data and vice versa

    (1) miss-representation of NPP's in a multi-source region
    due to temporal variability of emission (e.g. during revisions)
    (2) local sources
    (3) emission data IRE
    (4) uncertainties in dispersion modelling
    (?) . . .

- **Averaging reduces accuracy of measured and modelled data**
  - Weekly data depend on timing of arrival of cloud, Factor 2-3 difference. Daily sampling required
  - Maximum travel time only 14 days. CNL under-represented?

- **Time resolved emission data needed as input into ATM**
  - Further development and refinement of easy to use CSMS
  - Phase out PC -> isotope specific Xe measurements using SAUNA only
  - Continue collaboration: stack - near-field - far-field monitoring, Q3 2017

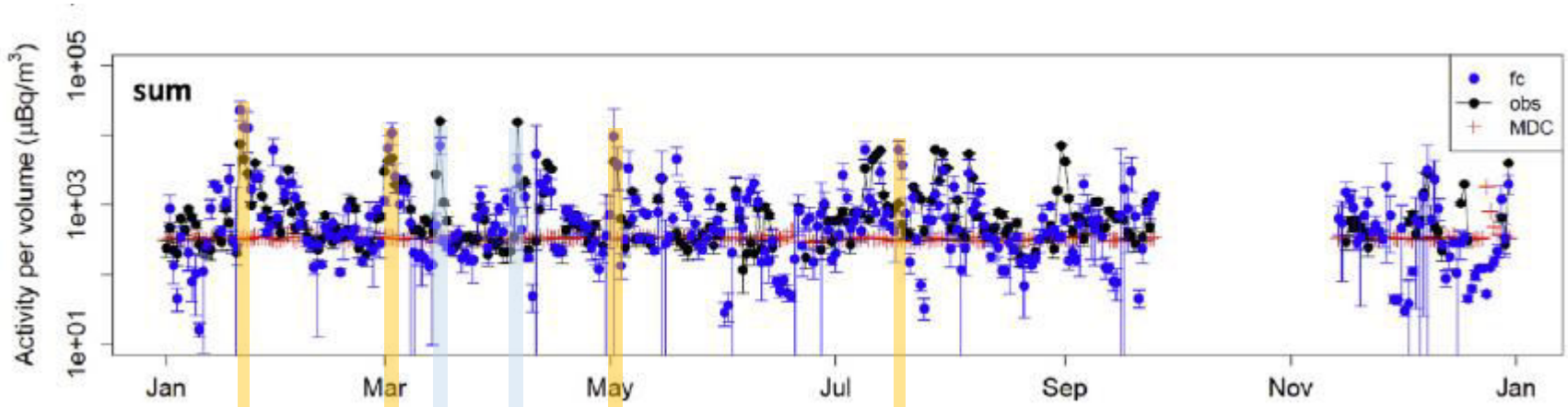
Questions?

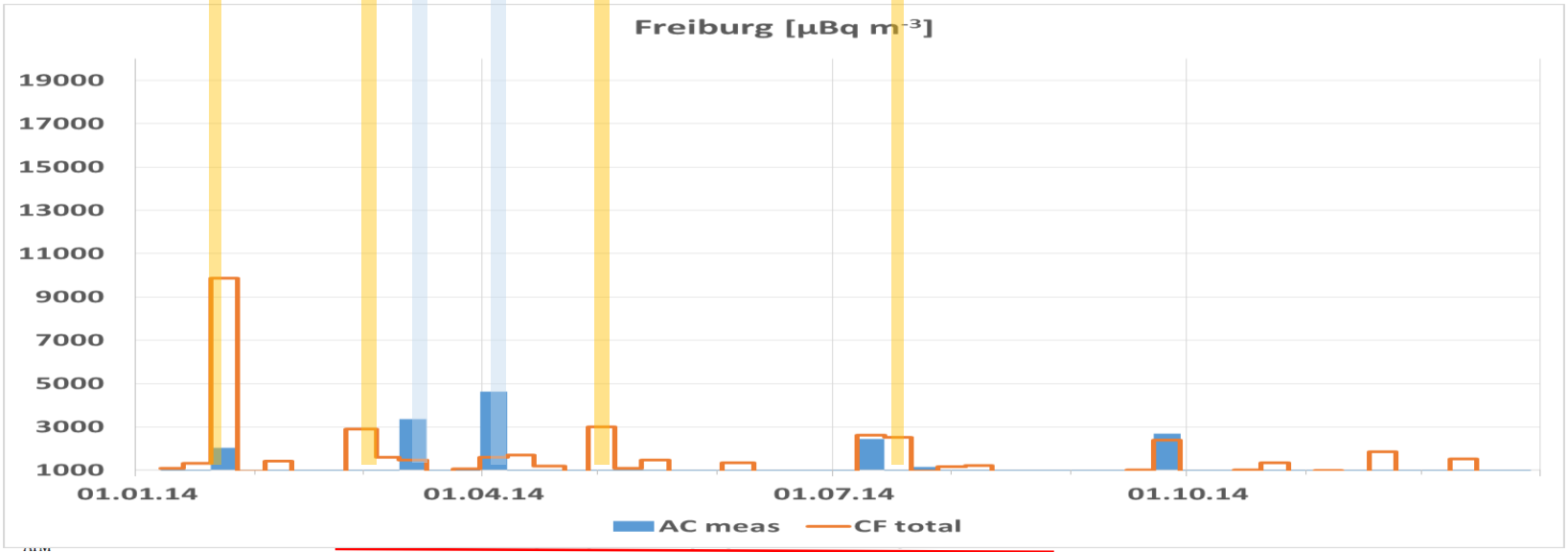

SMOKING GUN? WHAT SMOKING GUN?..
...YOU MEAN "ALLEGED SMOKING GUN..."
COUGH COUGH
...YOU MEAN "RUMORS OF AN ALLEGED SMOKING GUN"...
..."ALLEGED RUMORS OF AN ALLEGED SMOKING GUN"...

Campaign planned for September 2017

- MIPF Fleurus: ~ 10-16 September 2017
- 15‘ emissions data
- Measurements in near field (GDR + spectrometric)
- Daily noble gas samples: Freiburg, Schauinsland, Trier
- ATM (including near-field)
- Time-resolved data from German NPPs

2014 (daily samples) RN33 – weekly samples Freiberg





Uncertainty quantification
CTBT verification
Radioxenon monitoring

background.