Radioxenon prototype trap validation and controlled source experiments under the EU Council Decision VI project

J. Baré[1], M. Kalinowski[1], C. Gueibe[2], J. Rutten[2], J. Camps[2], K. van der Meer[2], D. Moyaux[3], B. Deconninck[3], J. Kusmierczyk-Michulec[1], M. Schoeppner[1]

[1] Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO), International Data Center, Vienna, Austria
[2] Belgian Nuclear Research Centre (SCK•CEN), Mol, Belgium
[3] Institute for Radioelements (IRE), Fleurus, Belgium

Email: jonathan.bare@ctbto.org

Abstract

The monitoring capabilities of the IMS noble gas systems may benefit from reductions of radioactive noble gas emissions. If these are achieved at nuclear facilities that have high radioxenon releases in normal operation, this could significantly enhance CTBT's verification capability.

In this framework, the SCK•CEN was contracted by the CTBTO to design a technical solution for the reduction of radioxenon emissions from civil nuclear facilities. A follow-up of this project was necessary to carry on the work already performed on emission reduction and to utilize it for controlled source experiments with field measurements, supported by high resolution ATM.

As a follow up of this project, both the SCK•CEN and the IRE were tasked by the CTBTO to:

- analyze the scale-up and the long term behavior of the prototype developed under EU Council Decision V;
- perform design studies at additional facilities;
- investigate further the stack releases at IRE, through observations at various distances and comparison with ATM results.

This project is funded under EU Council Decision VI. The current progress and the expected outcomes of the project are presented.

Context and scope of the project

Context

Radioxenon emissions from civil nuclear facilities significantly contribute to the background at the IMS noble gas detection systems. Although these emissions are within the relevant health regulatory limits, they affect the capability of the noble gas network to detect nuclear explosions. Reduction of emissions from civil nuclear facilities would significantly enhance the detection capability of the systems.

With a contribution received from the European Union within the framework of the EU Council Decision V (CFSP/2012/41/CTBTO V, 2015 – 2015), a mobile system which can be used for reducing radioxenon emissions from civil nuclear facilities was developed. The study was carried out by the Belgian Nuclear Research Center (SCK•CEN) in partnership with the Institute for Radioelements (IRE). From this collaboration, a retention system prototype was developed and designed to fit into the process line of the IRE. This first prototype demonstrated promising results and was donated to the IRE at the end of EU V [1].

Scope

Further developments and testing of the retention system prototype are needed. The current project, funded under EU Council Decision VI (CFSP/2015/1837 of 12 October 2015), is divided into 3 phases:

Phase I - Scaling up of the prototype system

- Test the system prototype and evaluate its long term stability;
- Study the long term behavior of the silver zeolites.

Phase II – Design studies

- Development of design studies for three nuclear facilities taking into account:
  - the characteristic features of the operational process;
  - the most suitable and efficient location for a retention system

Phase III – Controlled source experiment

- Field measurements around IRE supported with high resolution ATM

Phase I

Integration of the prototype system in the IRE process line

The prototype was placed in a shielded glove box (GB1) between the production hot cells (C1) and the decay tanks (cf. Fig. 1). Additional radiation detection and safety equipment were installed.

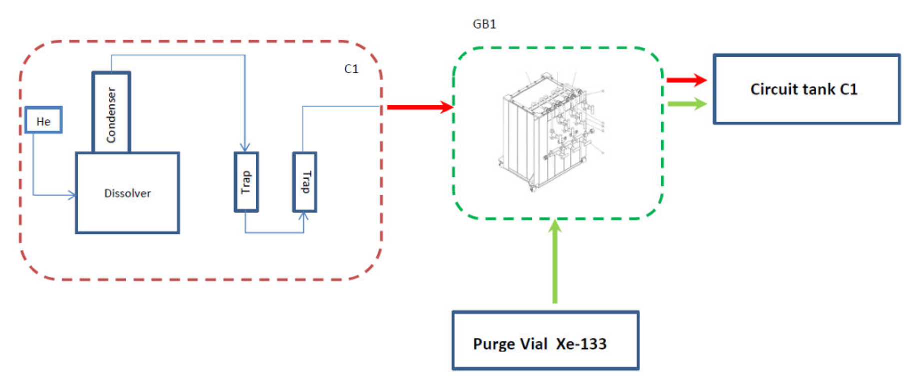


Fig. 1. Integration of the prototype into the LEU process line between the production hot cell and the decay tanks.

Prototype validation under controlled conditions and scaling up

In a first approach, the radioxenon was directly purged out of vials into the prototype. Breakthrough curves were studied and analyzed. Several assays were performed, and the theoretical model was adjusted (Fig. 2).

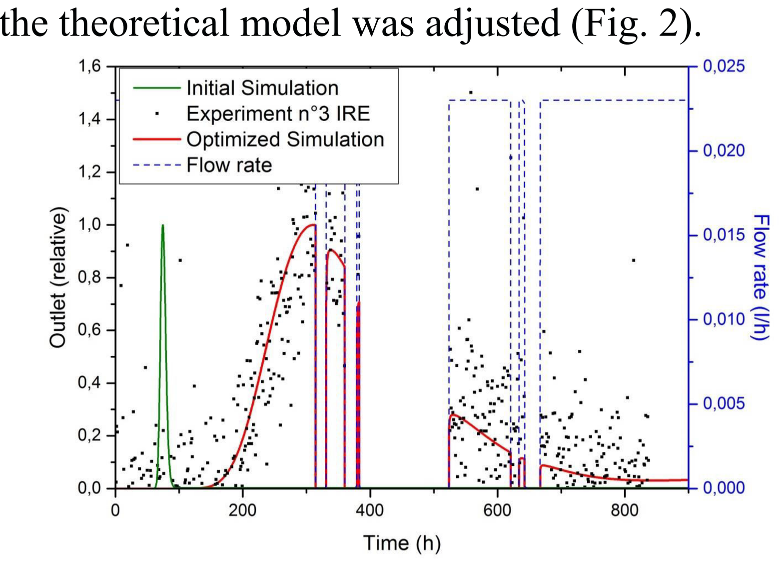


Fig. 2 – Simulated breakthrough peak from the initial (green) and optimized parameters (red) compared to the experimental one (black dots).

Results:

- The advection-diffusion-adsorption model was refined to better correspond to the experiments, and further validated;
- The current breakthrough experiment results are indicating that the prototype is well suited for operation in the process line;
- The regeneration of the material was performed after all breakthrough experiments and performed as expected;
- No degradation of the silver doped zeolite was observed for the moment;
- The xenon retention efficiency of a silver zeolites column was evaluated during the dissolution of LEU irradiated targets (scaling up). In preliminary experiments, the xenon trapping rate varies between 92% and 99%.

Phase II

Selection process

Facilities were selected in two stages, i.e.:

- a request for letters of interest was issued to 73 facilities worldwide;
- a dedicated web conference with the facilities that provided a letter of interest.

Selection criteria: technical feasibility, project planning and availability of the facility.

Result of the selection

Three facilities were selected for the design studies:

nordion · ansto · INUKI

The participation of the facilities in the project is on voluntary basis.

Design studies

For each of the selected facility, a design study will be performed taking into account the specific characteristic features of the operational process of the facility. The final design study will be donated to the facility to construct a prototype or a definitive trap (outside the project scope).

Phase III

Evaluation of the silver zeolites as retention system (ongoing)

The effluents from the main stack of the IRE are measured with activated charcoal using two parallel HPGe-based monitoring systems, allowing direct, accurate and continuous evaluation of the radioxenon releases in the atmosphere.

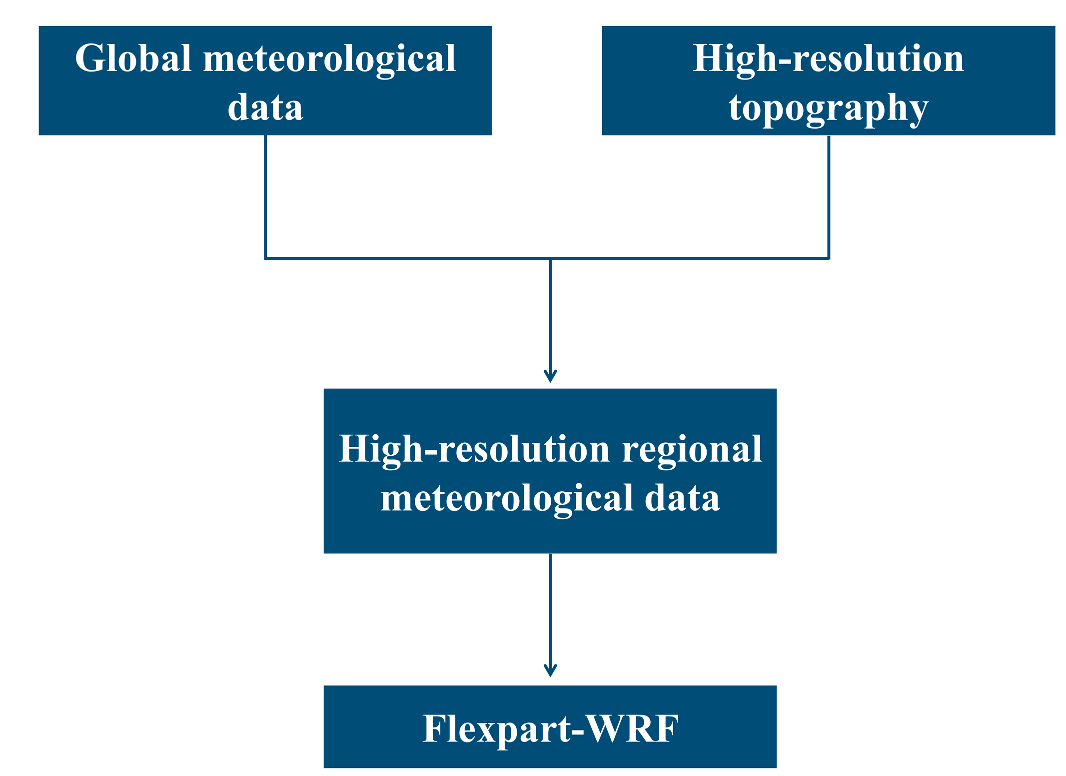
For the purpose of the project, the standard setup was modified (Fig. 3):

- a main monitoring system, measuring radioxenon with activated charcoal;
- a redundant monitoring system, measuring radioxenon with modified trapping setups.

Fig. 3 – The two monitoring systems running in parallel: the main system using charcoal in the background, the secondary system using silver zeolites in the foreground.

High-resolution ATM developments

The capability to conduct high-resolution (HR) atmospheric transport modelling (ATM) was recently developed at CTBTO [2]. In HR ATM, existing meteorological data are refined with a HR Earth's topography, creating HR data usable for regional scenarios with complex topography.



The HR-ATM software package delivers plotted meteorological data, plume dispersion and source-receptor sensitivity fields (Fig. 4) that can be used for post-processing and analysis.

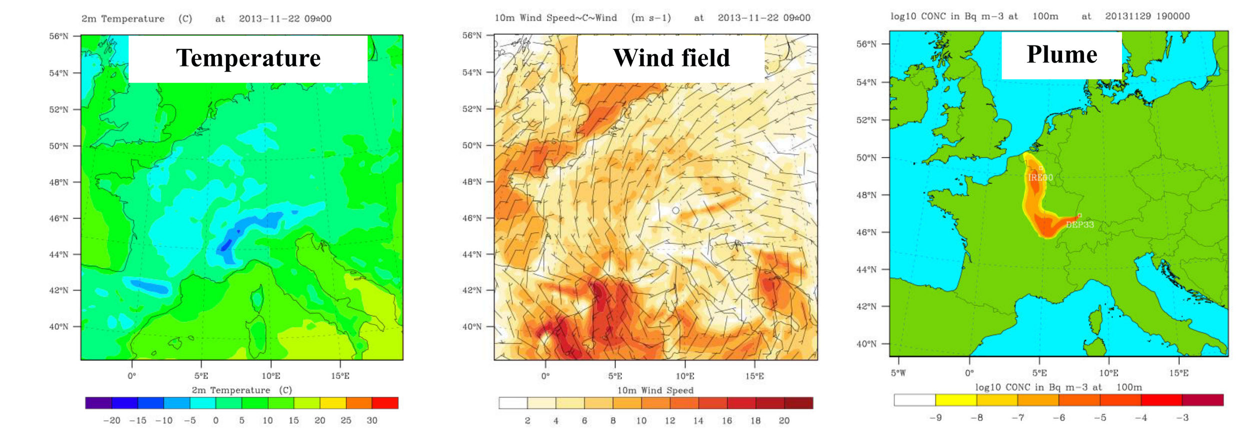


Fig. 4 – Examples of high-resolution meteorological data and plume dispersion based on HR-ATM.

The HR-ATM package has been applied on stack emission data. It was shown that radioxenon concentrations over complex terrains and various distances can be reliably reproduced (e.g. [3]). This capability will be of interest in the controlled source experiment, as environmental radioxenon will be measured in the complex surrounding area of the IRE.

Controlled source – receptor experiment (outlook)

Controlled source experiments with external atmospheric radioxenon field measurements will be organized around IRE to:

- assess the work performed on emission reductions;
- evaluate the potential interest of silver zeolites for detecting very low concentrations in the environment.

For the purpose of this measurement campaign:

- environmental radioxenon will be measured at spots of interest with customized silver zeolites cartridges equipped with additional air drying system;
- standard charcoal cartridges will be used in parallel and considered as a reference for the measurement;
- spots of interest will be determined based on meteorological forecast: several ATM models (including HR ATM as the local topography can be a key element of this measurement campaign) will be run;
- systematic plume hunting (measuring at sites where it is predicted the plume should pass) will be performed;
- all cartridges will be measured with HPGe detectors at IRE.

A full data set over short to medium distances (0 – 10 km) measurement spots is expected. The expected outcomes of this dynamic measurement campaign is the comparison of the radioxenon collected with the two materials (through activity measurements). Based on the results, the suitability of silver zeolites will be assessed for field measurements.

Conclusions and outcomes

Phase I: The prototype system was successfully installed in the production lines of IRE. The advection-diffusion-adsorption model was further refined. The prototype suitability for operational purposes was evaluated under controlled conditions and the material regeneration showed no degradation. In first experiments, it was shown that the xenon trapping rates is between 92% and 99%.

Phase II: Three facilities were selected for the design study, which will be performed based on the facility's operational characteristics features. A specific design study will be performed for each facility and donated for implementation.

Phase III: A controlled source experiment around IRE is in preparation. The retention capabilities of the silver zeolites is currently under evaluation. The measurement campaign will be supported by ATM, including the HR ATM model recently developed at CTBTO. A full set of data over short to medium distances is expected to demonstrate the potential suitability of silver zeolites for measurements of low activities of radioxenon in the environment.

References

[1] C. Gueibe et al., *Results and outcomes in xenon mitigation under the European Council Decision V*, poster T2.4-P10 at SnT 2017Conference, Vienna, Austria.
[2] M. Schoeppner et al., *Performance assessment of the high-resolution ATM at the IDC of the CTBTO,* poster T1.3-P19 at SnT 2017Conference, Vienna, Austria.
[3] C. Maurer et al, *2nd ATM challenge 2016* , oral presentation T1.3-O1 at SnT2017 Conference, Vienna, Austria.

Acknowledgement: this project in support of the Comprehensive Nuclear-Test-Ban Treaty is funded under the European Union Council Decision VI