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Study of medical isotope production facility stack emissions and noble gas isotopic signature using automatic gamma-spectra analysis platform

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The nuclear industry emissions of the four CTBT (Comprehensive Nuclear-Test-Ban Treaty) relevant radioxenon isotopes are unavoidably detected by the IMS along with possible treaty violations. Another civil source of radioxenon emissions which contributes to the global background is radiopharmaceutical production companies. To better understand the source terms of these background emissions, a joint project between HC, ANSTO, PNNL and CRL was formed to install real-time detection systems to support 135Xe, 133Xe, 131mXe and 133mXe measurements at the ANSTO and CRL 99Mo production facility stacks as well as the CANDU (CANada Deuterium Uranium) primary coolant monitoring system at CRL. At each site, high resolution gamma spectra were collected every 15 minutes using a HPGe detector to continuously monitor a bypass feed from the stack or CANDU primary coolant system as it passed through a sampling cell. HC also conducted atmospheric monitoring for radioxenon at approximately 200 km distant from CRL.

A program was written to transfer each spectrum into a text file format suitable for the automatic gamma-spectra analysis platform and then email the file to a server. Once the email was received by the server, it was automatically analysed with the gamma-spectrum software UniSampo/Shaman to perform radionuclide identification and activity calculation for a large number of gamma-spectra in a short period of time (less than 10 seconds per spectrum). The results of nuclide activity together with other spectrum parameters were saved into the Linssi database. This database contains a large amount of radionuclide information which is a valuable resource for the analysis of radionuclide distribution within the noble gas fission product emissions. The results could be useful to identify the specific mechanisms of the activity release.

The isotopic signatures of the various radioxenon species can be determined as a function of release time. Comparison of 133mXe and 133Xe activity ratios showed distinct differences between the closed CANDU primary coolant system and radiopharmaceutical production releases. According to the concept proposed by Kalinowski and Pistner (2006), the relationship between different isotopic activity ratios based on three or four radioxenon isotopes was plotted in a log-log diagram for source characterisation (civil vs. nuclear test). The multiple isotopic activity ratios were distributed in three distinct areas: HC atmospheric monitoring ratios extended to far left; the CANDU primary coolant system ratios lay in the middle; and 99Mo stack monitoring ratios for ANSTO and CRL were located on the right. The closed CANDU primary coolant has the lowest logarithmic mean ratio that represents the nuclear power reactor operation. The HC atmospheric monitoring exhibited a broad range of ratios spreading over several orders of magnitude. In contrast, the ANSTO and CRL stack emissions showed the smallest range of ratios but the results indicate at least two processes involved in the 99Mo productions. Overall, most measurements were found to be shifted towards the reactor domain. The hypothesis is that this is due to an accumulation of the isotope 131mXe in the stack or atmospheric background as it has the longest half-life and extra 131mXe emissions from the decay of 131I. The contribution of older 131mXe to a fresh release shifts the ratio of 133mXe/131mXe to the left. It was also very interesting to note that there were some situations where isotopic ratios from 99Mo production emissions fell within the nuclear test domain. This is due to operational variability, such as shorter target irradiation times.

Martin B. Kalinowski and Christoph Pistner, (2006), Isotopic signature of atmospheric xenon released from light water reactors, Journal of Environmental Radioactivity, 88, 215-235.