Using Gamma-ray Spectrometry In The Nuclear Power Plants

NKS Gamma workshop – training 26.9.2023

This presentation tries to give answers to these questions:

- What is gamma spectrometry used for in a NPP
- What type of instrumentation is used
- What are the main nuclides that can be seen inside a NPP / in the environment
- What kind of training requirements are set for the personnel

Disclaimer: the examples are taken from the Finnish BWR/VVER/PWR power plants, but very similar solutions are found around the world.

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Why are gamma measurements done in a NPP

Gamma measurements are an important tool for understanding the state of the radiochemistry of the plant.

- Activity results give information about:
  - **The integrity of the fuel** (rise of the fission products).
  - **State of corrosion** (the level of corrosion products, release of crud).
  - **Leakages between systems** (primary-secondary leak, finding radioactivity in a system usually clean from radioactivity, etc).
  - The impurities of chemicals used in the reactor circuit, introduction of oxygen in the reactor circuit, foreign objects, etc (rise of activation products).
- "Scientific accuracy" (= absolute value) is not sought or needed→ **TRENDS** are important to see the changes in activity levels and understand the phenomena behind it.

But – discharge measurements need to be sensitive, to obtain the required level of detection stated in the regulation, e.g. 2004/2/Euratom.

What different sample types are measured

- **Water** (nuclides with half-lives of > 30min)
  - Water samples are either measured as it is or filtered (filtrate + filter = dissolved + undissolved)
  - Depending on the sample it might have to be measured a few times to obtain short and long lived nuclides in different measurements
- **Gas** (nuclides with half-lives of > 10 min)
  - Separating the gaseous nuclides from the reactor water (e.g. by bubbling) gives the most sensitive sample for finding nuclear fuel leakages.
- **Air** (nuclides with halfli-ves of > 10 min)
  - A grab sample of discharge air is measured to obtain the noble gas discharges (1-4L Marinelli)
  - Iodine- and aerosol filters are collected continuously in the release stack or obtained using a portable collector
- **Ion exchange resins** (nuclides with halflives of > 1 day)
  - Resins used to purify the reactor water are measured after removal to obtain information about the activation/corrosion/fission products inside the reactor

What nuclides are seen in a nuclear reactor (1/2)

Activation and corrosion products

- Any material that is made radioactive by the process of neutron activation is called an activation product. They are formed inside the reactor/reactor building from materials that are inside the neutron flux (e.g. air, concrete, piping, chemicals/impurities,…)
- Example nuclides (and their source):

  Na-24 and K-42 (chemicals and impurities), Ar-41 (Ar-40 in air), Ag-110m (Ag-materials), Sb-122/124 (Sb in materials)
- Specific activation products, that are originating from the corrosion of metal components are called **corrosion products**. Corrosion can be minimised by using appropriate chemistry and choosing suitable materials.
- Example nuclides (and their source):

  Cr-51 (Cr-materials), Mn-54 and Fe-59 (Fe-materials), Co-58 (Ni-materials), Co-60 (Co-materials), Zn-65 (Zn-materials)

What nuclides are seen in a nuclear reactor (2/2)

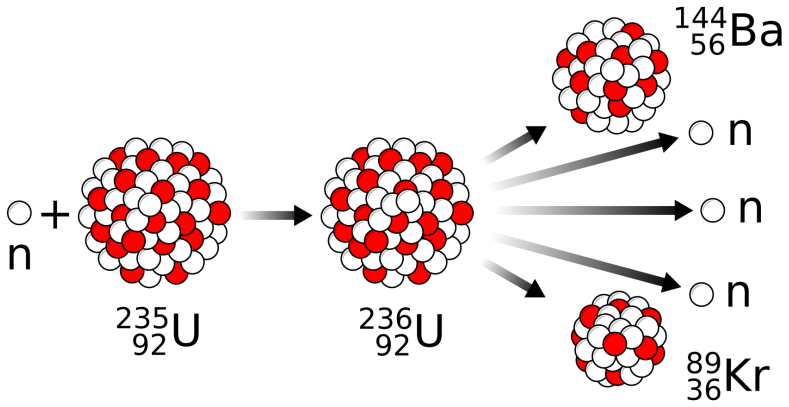
Fission products

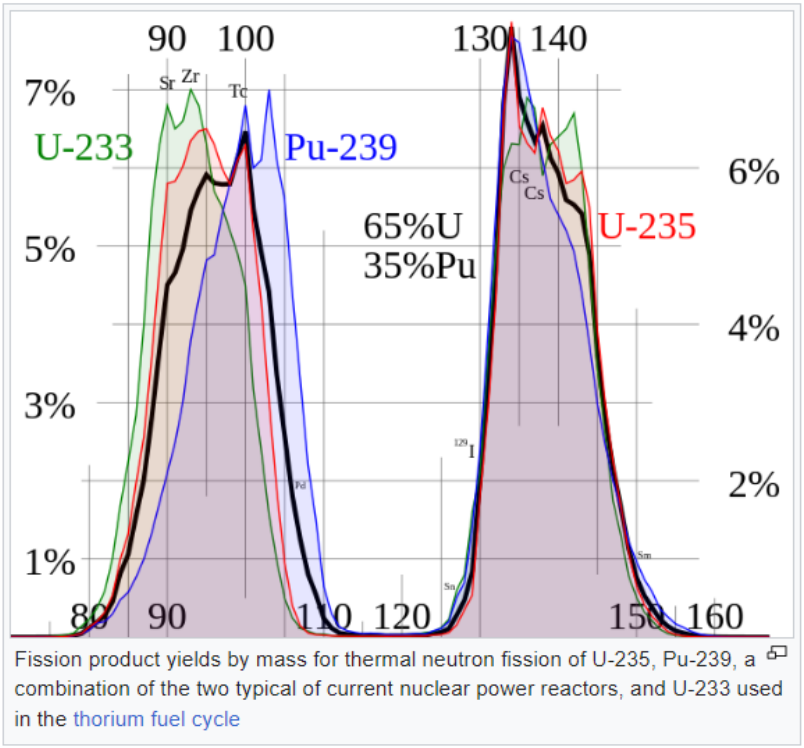
Nuclear fission products are the atomic fragments left after a large atomic nucleus undergoes nuclear fission. Fission products are formed either inside the fuel elements or from the uranium on the surfaces of the reactor (tramp uranium).

Examples: Kr-87, Kr-88, I-131, I-133, Xe-133, Xe-135, Cs-134, Cs-137, Ba-139

Fission product yields from Wikipedia:

basically “anything goes” between mass numbers 85-105 and 125-145





Fission product yields by mass for thermal neutron fission of U-235, Pu-239, a combination of the two typical of current nuclear power reactors, and U-233 used in the thorium fuel cycle

What nuclides are seen in NPP discharges

- Any nuclide produced inside the NPP can potentially be released to the environment, though there are numerous purification/delay systems in use
- Discharges are controlled by various continuous measurements and representative samples are taken on a weekly basis (air) or from each release batch (liquid)
- Typical nuclides in discharges to the air are:
  - (H-3, C-14), Ar-41, Cr-51, Mn-54, Co-58, Co-60, Sb-124, I-131, I-133, Xe-133, Xe-135, Cs-137
- Typical nuclides in liquid discharges are:
  - (H-3), Cr-51, Mn-54, Co-58, Co-60, Ag-110m, Sb-124, I-131, Cs-137, Ba-140, Ce-141

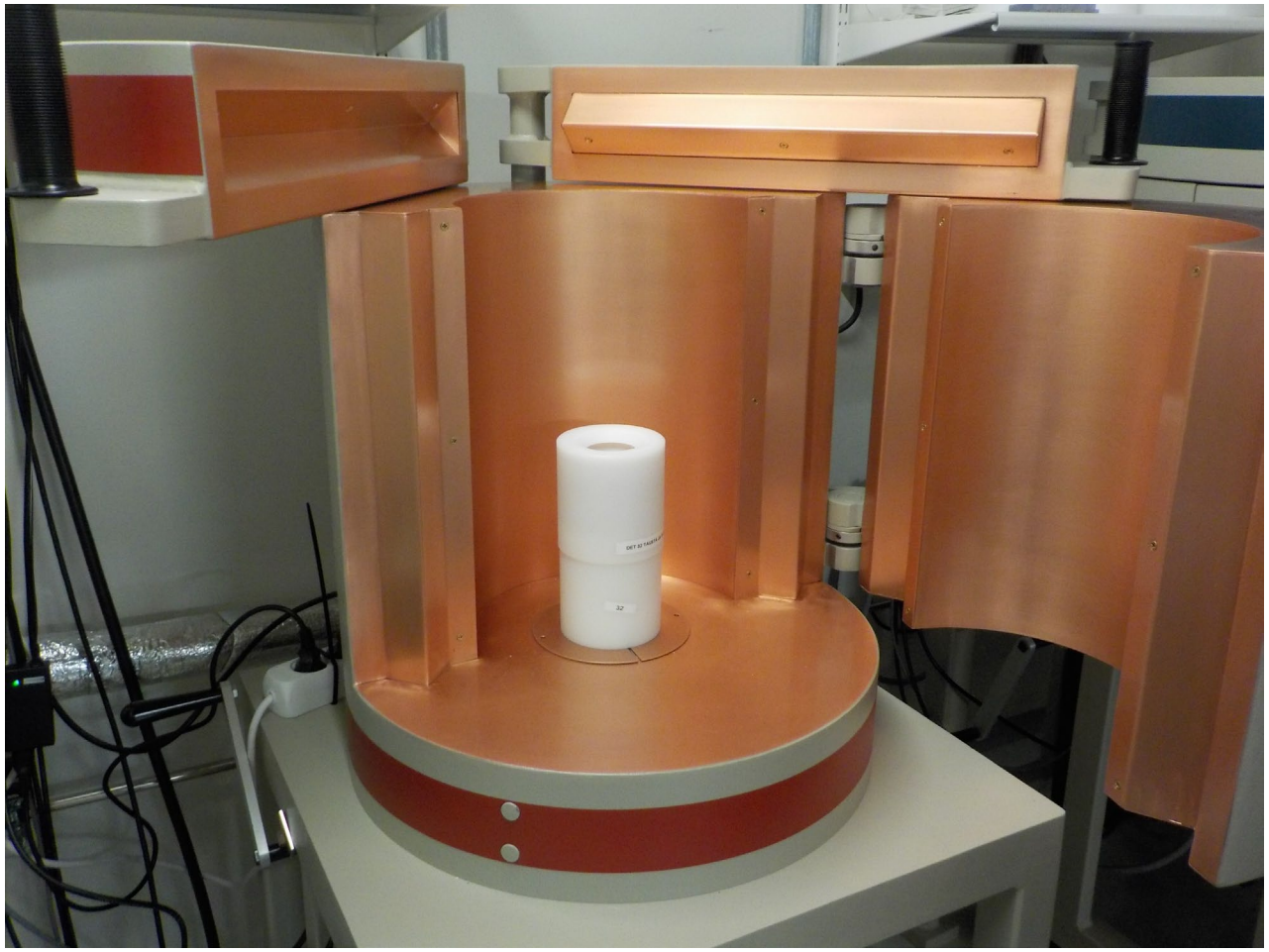
- The European Commission has a discharge database RADD (1995 →, nuclear site specific data) https://europa.eu/radd/
- The IAEA has a discharge database DIRATA https://dirata.iaea.org/ is being developed, it will have a longer time scale, when it is finished

Instrumentation requirements

- What is required from the instrumentation
  - **Many different geometries for different sample types and distances** (e.g. 0 – 30 cm)
  - Several HPGe detectors with different efficiencies (typically 15 – 65 %)
  - Some detectors should also measure lower energies (below 100 keV)
  - **A good automatic spectrum analyser** (e.g. analysis sequence in APEX/Genie)
  - **A broad measurement range ~1E-4 – 1E8 Bq/L**
    - Discharge samples and active process samples are measured with the same instrumentation in the same room on the same day
    - Prevention of cross contamination of samples is needed
    - The measuring chambers need to be kept clean from contamination!
    - The background level of the measuring room needs to be kept low (MDA requirements for discharge sample analyses)
  - As gamma spectrometry is needed on a daily basis to fulfil the requirements set by the plant (process control) or the regulator (discharge control), **"down time" e.g. due to power cuts needs to be minimised** (e.g. by using CryoCycle type cooling system).

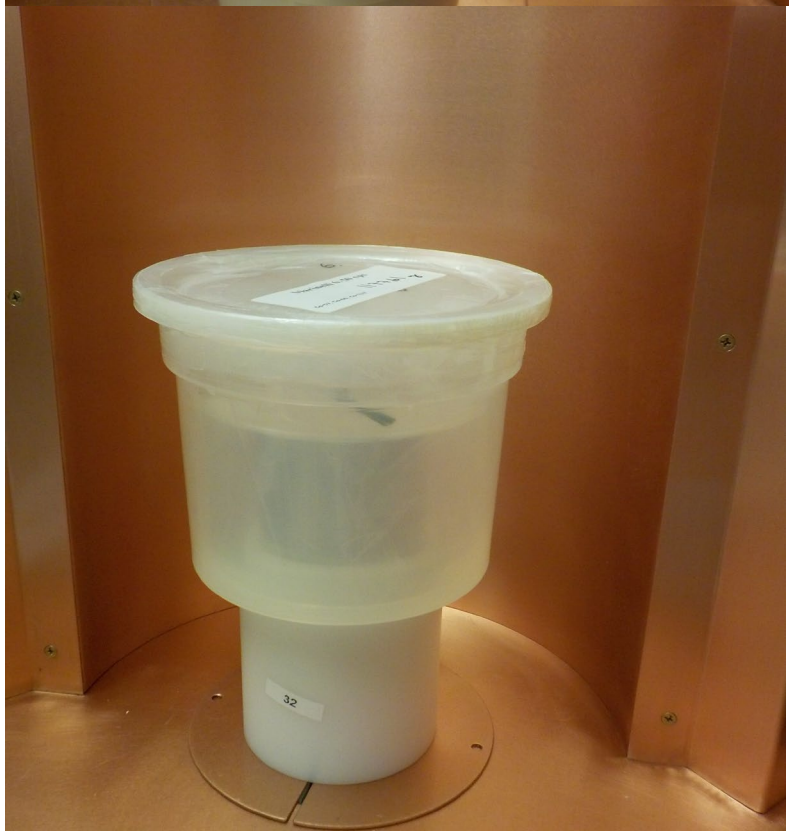
An example of a HPGe detector with thick lead+copper shielding.

The white sample holder allows for a precise positioning of the sample at different distances (more blocks can be stacked)



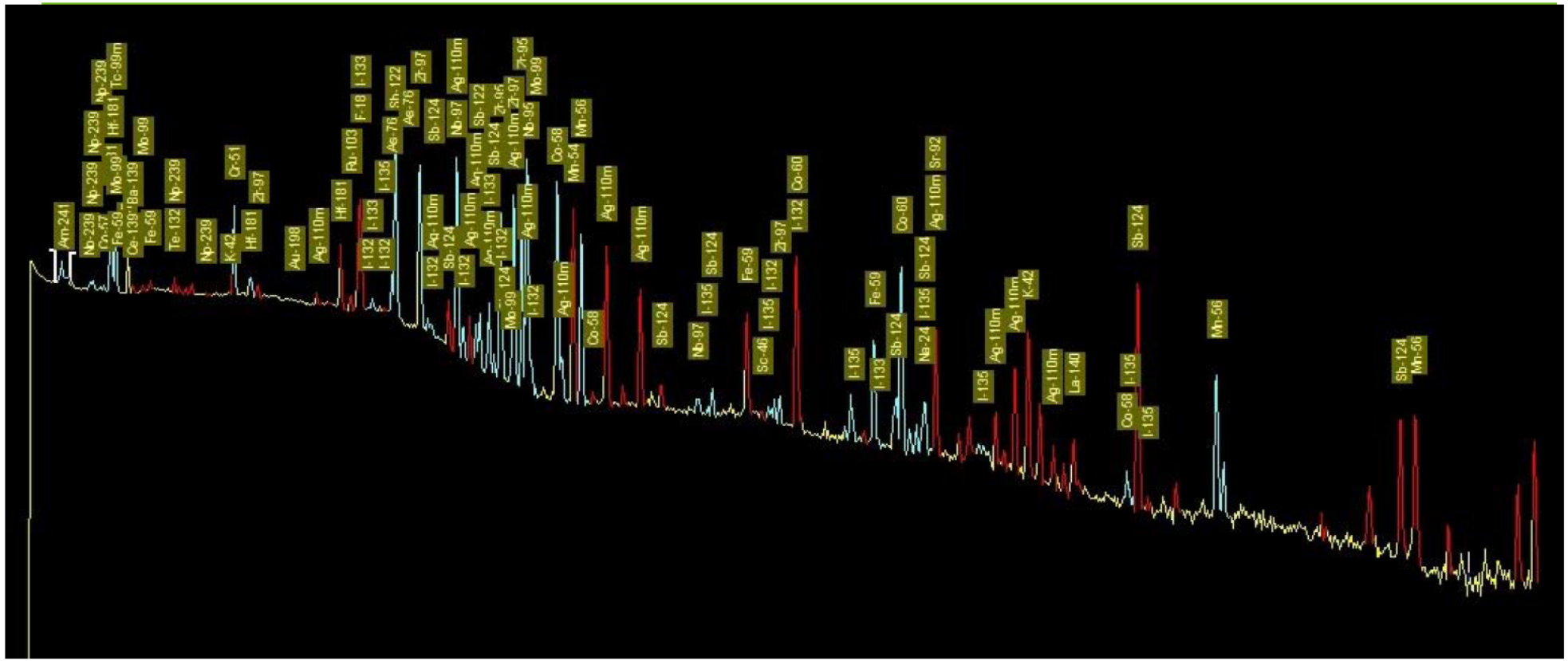
Particulate filter

Marinelli beaker



Pictures from OL3

LO2 particulate matter 3.10.2014



- **133 peaks** are found, 38 of them are identified as SUM or ESC peaks
- Peaks for 37 nuclides are initially identified, after interference correction and daugther/parent evaluation, results for **34 nuclides** are given.

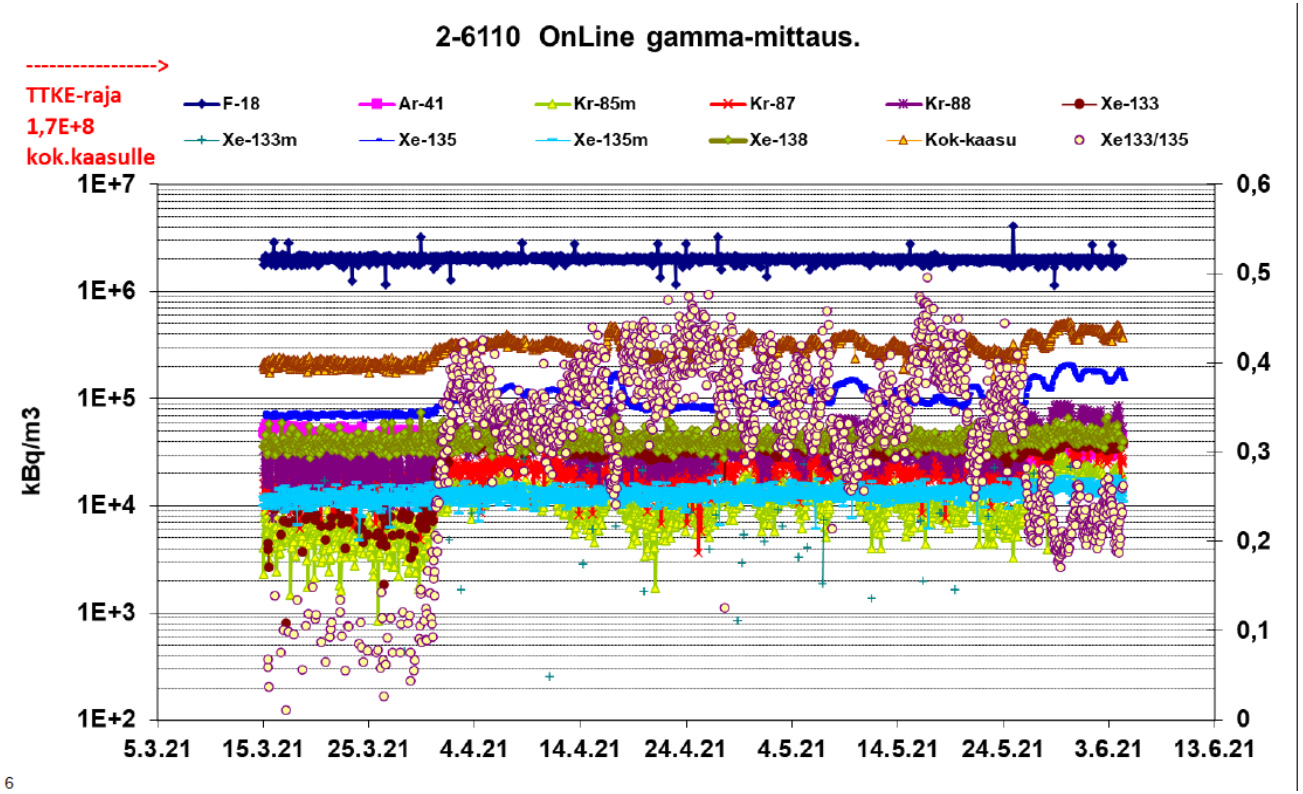
Knowledge requirements

- Good knowledge of gamma spectrometry is essential at a NPP
  - How the instrumentation works, how to solve problems with the instrumentation or the analysis programme, how the analysis sequences have been chosen (validation!), what can be seen in a spectra, how is the nuclide library formed, how is summing tackled, how are all the calibrations done, how is the quality control done, uncertainties etc…
  - The person responsible for the measurements should have a suitable educational background, like radiochemistry or physics.

- What is required from the personnel doing the daily measurements
  - Constant learning (e.g. new nuclides in samples and their meaning)
  - Attention and care (reliable results, no contamination of samples).
  - Understanding of what is “normal” in which process sample in which process situation.
  - Questioning attitude towards the automated spectrum analyser results (sufficient training!)
  - **A proof of competence!**

Continuously measuring devices

- Some plants have gamma spectrometric measurements attached to the process.
  - These give additional information about the state of the process, but are not precise enough for official reporting.
  - If there is an indication of a change in the process (e.g. rise of fission products in reactor water), a grab sample can be taken to confirm the situation.



The noble gas results of the online measurement showing a small but clear rise in fission products → fuel leakage

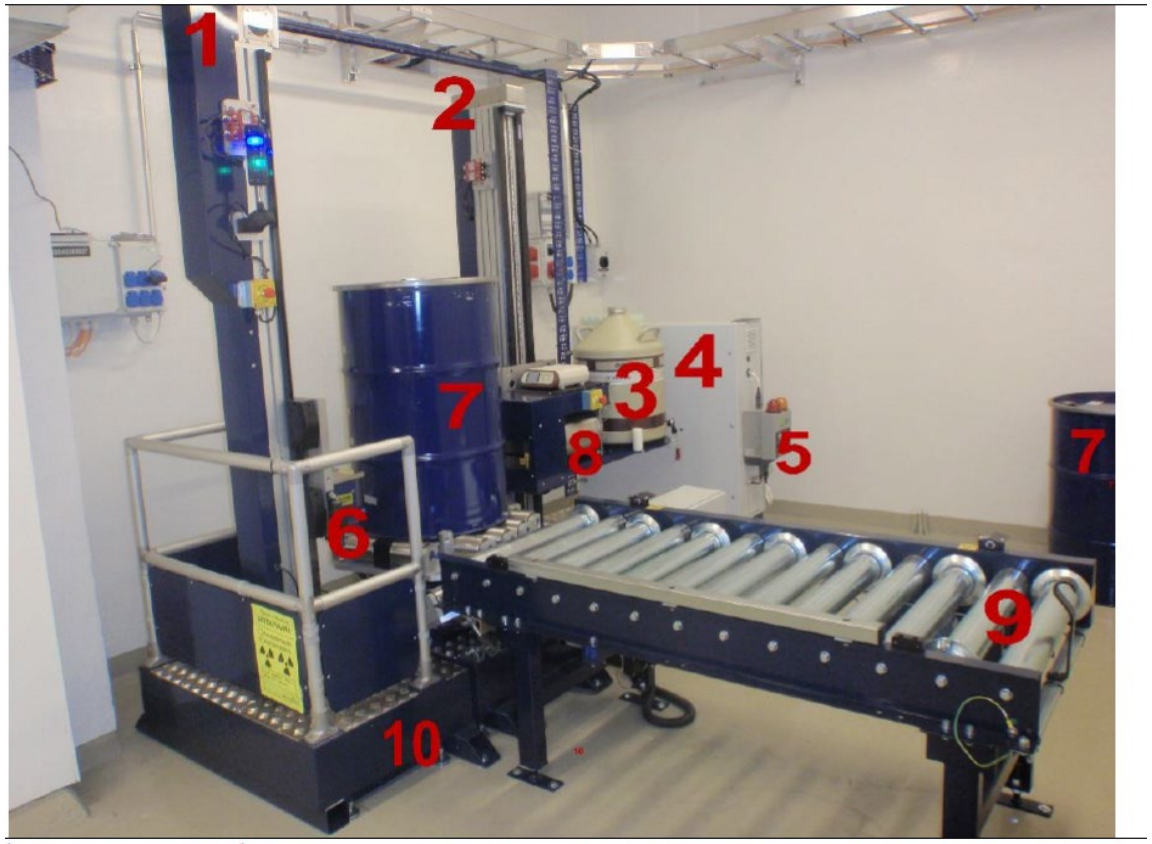
The online measurement instrumentation in Loviisa (electrically cooled HPGe)

Additional gamma spectrometry applications

- Gamma spectrometry is also needed to measure the waste drums in a NPP.
  - The waste generated inside the NPP has to be measured and activity records must be kept.
  - There are clearance levels stated for each nuclide in the regulation.
  - If the clearance levels are surpassed, the waste is stored and possibly released after sufficient aging. Otherwise it is put in the repository.

Calibration drum in Olkiluoto

Series 3500 Segmented Gamma Scanner in Loviisa



Additional gamma spectrometry applications

- Gamma spectrometry is also needed for
  - Measuring the contamination levels of various surfaces like steam generator piping and heat exchangers to see what nuclides are prominent (qualitative and quantitative)
  - Assessing the pellet location on used nuclear fuel rods under water
  - Looking for radiation sources/fallout nuclides in the environment

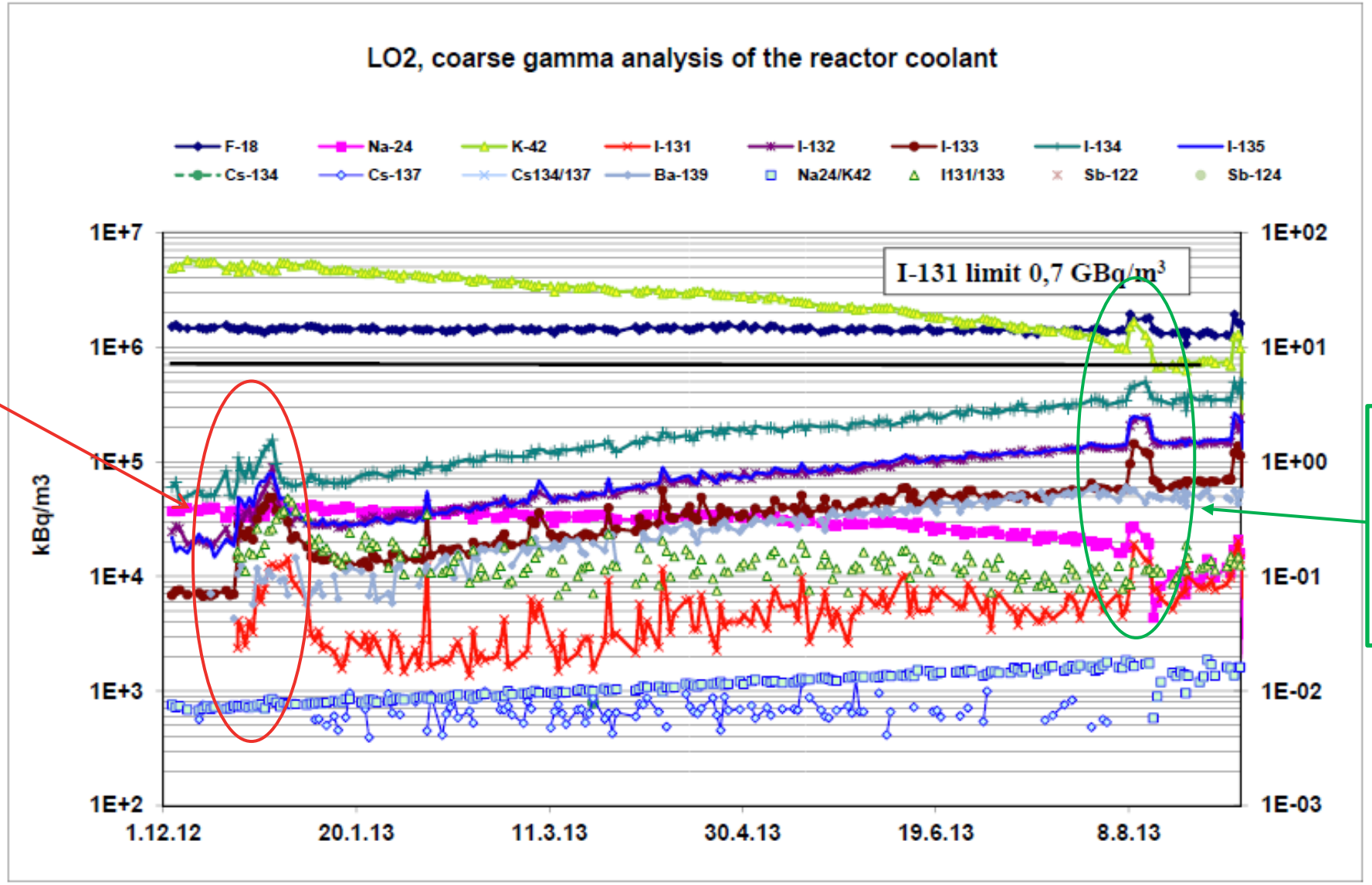
In-situ HPGe measurements from piping (2 m high) in Olkiluoto

$LaBr_3$ Backpack for source monitoring in the environment

InSpector™ 1000 Digital Hand-Held Multichannel Analyzer NaI

If there is time

- A look at the daily water sample analysis results from Loviisa, showing the rise of fission products, the behaviour of some activation products and the effect of the purification system on the activity levels. Trends are important, with activities like this, the absolute value is not needed. The presence or absence of a nuclide is also an important piece of information.



Fuel leakage detected, fission products rise

Purification resins are changed → all nuclides rise temporarily

F-18 is steady = steady power during the cycle

Activation products K-42 and Na-24 drop during the cycle

Fuel leakage detected, fission products rise

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