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SIGNIFICANT I-131 DISCHARGES FROM THE RADIOPHARMACEUTICAL INDUSTRY UNDER NORMAL OPERATING AND INCIDENT CONDITIONS

MEMBRE DE ETSON

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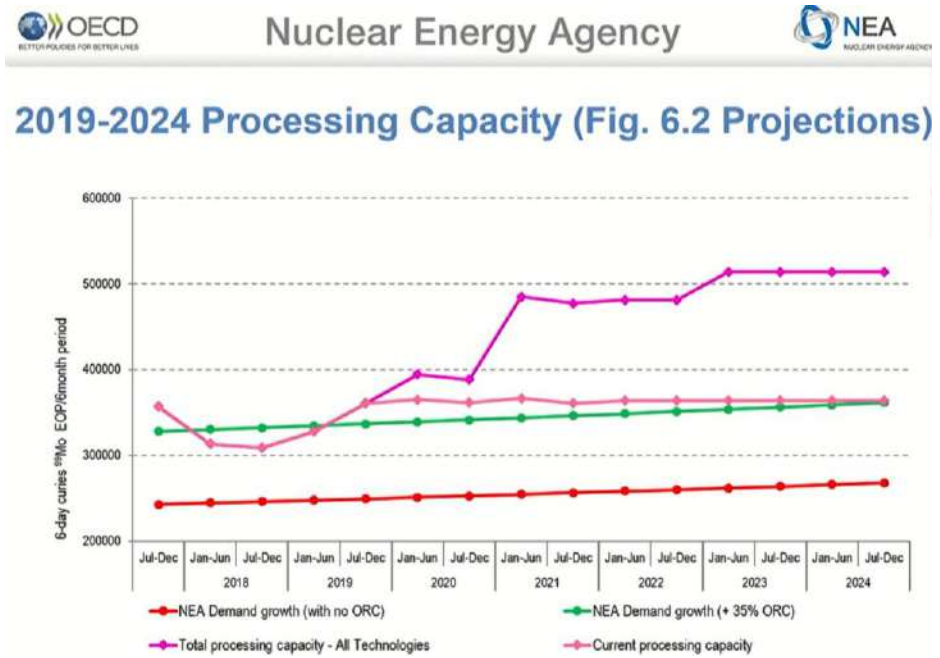
- To cope with an increasing demand of radiopharmaceuticals in nuclear medicine there is an increasing effort since several years and decades to increase Medical Isotope Production facilities (MIPFs) capabilities. This also results from the low number of facilities that can produce radiopharmaceuticals which thus concentrate the production on a low number of them

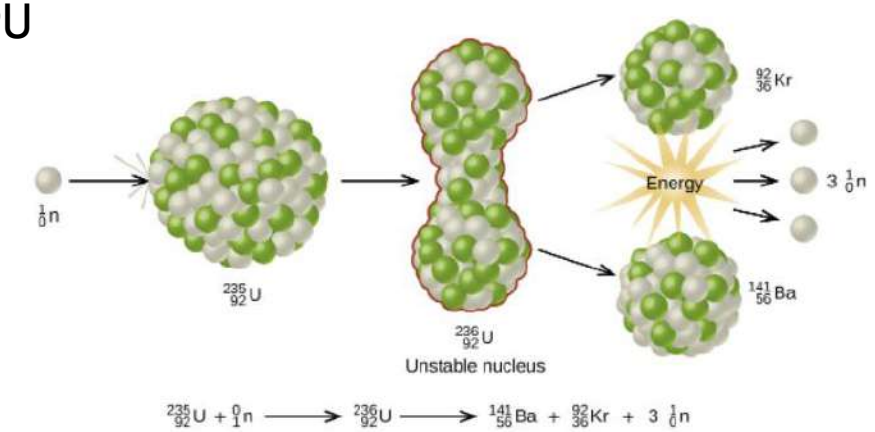


- Little growth is forecast for ^{99m}Tc in OECD countries through 2023 and 5% per year for developing markets. However, the global interest for radiopharmaceuticals indirectly results in a potentially not negligible release of radionuclides into the environment, both encompassing production and use
- The most commonly used reactor-produced isotope in medical applications is ^{99m}Tc (about 30 millions of nuclear diagnoses / year and 85% of all diagnostic scans worldwide), followed to a much lesser extent by ^{131}I, ^{32}P, ^{51}Cr, ^{89}Sr, ^{153}Sm, ^{186}Re and ^{177}Lu as well as ^{223}Ra and ^{227}Th

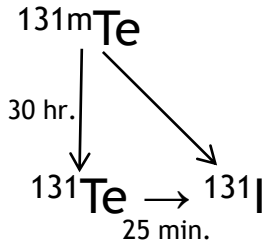
Among the 37 different radioiodine, 4 of them (^{123}I, ^{124}I, ^{125}I, and ^{131}I) are produced for medical applications
^{131}I is the most commonly used radioiodine isotope in the treatment of thyroid diseases (therapeutic purpose)

It is produced commercially in large quantities through nuclear fission, simultaneously with ^{99}Mo and ^{133}Xe. To somewhat extent, it can be considered as a by-product of the ^{99}Mo production. Depending on the reactor used, the obtained ^{99}Mo activity varies from 12 TBq to 19 TBq (post calibrated 8 days after unloading targets from the reactor). Complementary, more than 20 TBq ^{133}Xe and 5 TBq ^{131}I at the same calibration date are recuperated by side-processes developed in shielded boxes
Two main production routes : fission of ^{235}U and so-called (n, γ) reaction (neutron activation)

Fission of ^{235}U



$$^{235}_{92}U + {}^{0}_{1}n \longrightarrow {}^{236}_{92}U \longrightarrow {}^{141}_{56}Ba + {}^{92}_{36}Kr + 3\,{}^{1}_{0}n$$



Another ^{131}I production route involves neutron activation of a stable target (e.g.^{130}Te)

Processing of radioactive gaseous wastes is a serious problem in the Mo extraction from fission products

Although most of I and Xe are recovered a small portion of organic iodides such as methyl iodide contained in the atmosphere is to be released in the exhaust system

To prevent major gaseous ^{131}I releases into the environment, mitigation processes (HEPA filters for ^{131}I-labeled aerosols and adsorbent beds (charcoal, zeolite, alumina…) are installed) but their efficiency despite being very high is not 100 %

As a result, the radiopharmaceutical ^{131}I production is the largest emitter of ^{131}I into the environment

Evidence for significant emissions of ^{131}I from Medical Isotope Production Facilities

In routine operations (authorization limits up to several hundreds GBq/yr. as compared to a total release of about 0.3 GBq/yr from 58 operating French NPPs in 2013 !)

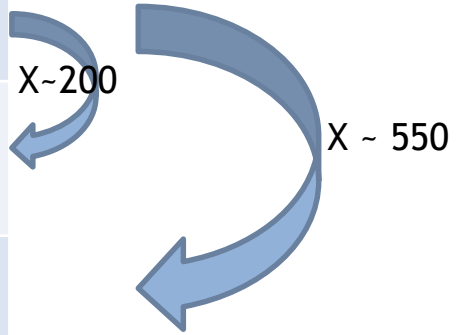
- Current MIPF discharges are significant (if not close) to their authorization limits. This likely results from the low number of facilities able to produce ^{99}Mo with radiopharmaceutical grade
- Regular ^{131}I detections at trace concentrations have been reported near MIPFs while this has never been pointed out for NPP
- Regular detections (^{131}I only) also in Scandinavia or at IMS stations in support to the CTBTO, downwind of MIPFs. During the 2009-2019 period (2011 left out because of the Fukushima accident), and except ^{137}Cs, ^{131}I was the most detected anthropogenic radionuclide in Finland,
- From January 2019 onwards more than 600 ^{131}I and about 80 ^{133}I trace detections events were reported by the IMS stations

More information

- Masson et al., (2018) Potential source apportionment and metereological conditions involved in airborne I-131 detections in anuary/February 2017 in Europe. Environ. Sci. Technol., 52 (15): 8488-8500
- Ageeva et al. (2015) Long-term monitoring airborne I-131 in the surface layer in Obninsk city, Kaluga region. Radiation & Risk, 24 (1), 96–107
- Goodwin et al. (2022) Analysis of radionuclide detection events on the International Monitoring System. J. Envir. Radioact., 242, 106789

Comparison of routine ^{131}I discharges from various nuclear activities

Atmospheric iodine discharges ($GBq.y^{-1}$) (Expressed in equivalent ^{131}I)	2014	2015	2016	2017	2018	2019	2020
IRE (MIPF)	1.772	1.637	1.973	1.594	2.023	1.64	1.906
Tihange NPP 3 PWRs (3000 MWe)	0.009	0.0078	0.008	0.0078	0.011	0.010	0.0115
SCK-CEN	0.0028	0.001	0.002	0.0028	0.00734	0.00264	0.00449
Sellafield (airborne ^{131}I discharge from SF of heavy nuclides)			0.42	0.37	0.39	0.23	0.12



For the sake of comparison, a ^{131}I capsule administrated for the treatment of thyroid disease can contain as much as 6 GBq of ^{131}I !

^{131}I releases from MIPFs in incident/accident conditions

Significant ^{131}I releases were detected on a continental scale in 2011 and 2017. At such a scale and at distance from the emission point, only a release on the order of magnitude of several tens GBq and more can lead to detectable concentrations (a fraction of µBq/m^3 to a few tens of µBq/m^3 range for the particulate fraction, 3 to 4-fold more for the gaseous fraction)

The two most significant reported ^{131}I-release events since 2000 were:

- 48 GBq in Belgium in summer 2008 (rated 3 on the INES scale because similar to the yearly authorization),
- 342 GBq in Hungary in fall 2011 (rated 1, about 5-fold < to the yearly authorization)

More information

- https://ec.europa.eu/energy/sites/ener/files/documents/tech_report_hungary_2012_en.pdf
- Masson et al., (2018) Potential source apportionment and metereological conditions involved in airborne I-131 detections in January/February 2017 in Europe. Environ. Sci. Technol., 52(15): 8488-8500
- IAEA (2017) Source of Iodine-131 in Europe Identified. http://www.iaea.org/newscenter/pressreleases/2011/prn201127.html
- Vandecasteele et al. (2011) Rejet accidentel d'iode-131 par l'IRE sur le site de Fleurus: Retour d'expérience de l'autorité de sûreté belge. Radioprotection 2011, 46 (2), 159–173

Thank you for your attention