



Radioisotopes in Effluents from PWR NPP during its Lifetime

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ABSTRACT

The level of radioactivity in discharges from a nuclear power plant (NPP) is a subject of strict control during the operation of an NPP as well as during its decommissioning. The control over radioisotopes in effluents of NPPs in the European Union (EU) is a subject of the Euratom Treaty signed in 1957. In addition, the European Commission published in 2004 a recommendation on standardised information on radioactive airborne and liquid discharges into the environment from nuclear power plant reactors and reprocessing plants in normal operation.

In 2004, 157 NPPs were in operation in the EU, the majority of these NPPs, namely 86 were pressurised water reactors (PWRs). On average they entered operation 22 years ago. These reactors already went through a few steps of the lifecycle of an NPP: start – up phase, modernisation, upgrading etc. The effect of those steps on effluents has been studied for four Westinghouse NPPs in four different states in Europe, namely Slovenia, Spain, Sweden and Switzerland. Great discrepancies in activities of group of radionuclides in airborne and liquid effluents were found for a particular year. Only for specific radionuclides such as C-14 in airborne effluents and H-3 in liquid effluents these discrepancies were not found out.

The study of the lifetime cycle from 1983 till 2004 of the Krško NPP is reflected on its effluents. Five periods in its lifetime are identified showing also that due to the leakage of fuel elements in the years from 1993 to 1997 a substantial increase of activity of noble gases in effluents could be observed in that period.

The operational regime of an NPP as well as the availability of best available technologies for treatment of effluents could drastically influence the absolute values of activities of some radionuclides in effluents. While no general study of operational regimes, pre-treatment technologies and subsequent effluents for NPPs in Europe is available yet, a first study identifying the lifecycle of an NPP and effluents could be done. In addition, it should be emphasised that no harmonised approach to the measurement techniques including sampling procedures for determination of effluents in NPPs is in place and neither harmonised reporting. Furthermore also a scope of a control over effluents has not been harmonised yet.

1 INTRODUCTION

The level of radioactivity in discharges from a nuclear power plant (NPP) is a subject of strict control during the operation of an NPP as well as during its decommissioning. Liquid and gaseous radioactive effluents from nuclear power reactors are also very good performance indicators of the plant operation [1]. Several groups of radionuclides are usually monitored in liquid and gaseous discharges, namely tritium, carbon C-14, iodine isotopes, noble gases, particulates (activation and fission products) and alpha emitters, while at some types of

nuclear power reactors some specific radionuclides in effluents are also important as for example S-35 from gas cooled reactors.

The control over radioisotopes effluents of NPPs in the European Union (EU) is a subject of the Euratom Treaty signed in 1957 [2]. In addition, the European Commission (EC) published in 2004 a recommendation on standardised information on radioactive airborne and liquid discharges into the environment from nuclear power plant reactors and reprocessing plants in normal operation [3]. The recommendation was published to “ensure minimum standards for the analysis methods”. The EC also publishes periodical reports on releases of radioactive materials to the environment in the member states from NPPs and from nuclear fuel reprocessing plants. The reports [4, 5] are based on the Commission Recommendation given in [3]. The eleventh in the series [5] deals with the period 1999-2003 including data from NPPs with a capacity larger than 50 MWe. The reports enable a comprehensive assessment of levels and radionuclide distribution in liquid and gaseous effluents from NPPs in the EU.

According to the data given in [6] 157 NPPs were located in EU in 2004. The majority of the NPPs, namely 86 were pressurised water reactors (PWRs). On average they entered operation 22 years ago. These reactors already went through a few steps of the lifecycle of an NPP: start – up phase, modernisation, upgrading etc. The average capacity of these PWR NPPs is 1065 MWe [7].

During the lifetime of a PWR NPP some non-uniformity in ratio of radioisotopes in discharges or in absolute values of discharged activities can be usually observed. The aim of the article is to present these non-uniformities as well as to encourage the study of operational experiences which can influence them.

2 REGULATORY CONTROL OF EFFLUENTS

The monitoring of discharges into the environment comprises two types of releases, namely gaseous as well as liquid effluents. Furthermore transport of solid waste from an NPP site as well as discharges of the solid material based on clearance levels are carefully monitored prior to transport or releases, very often using on line monitors at the exit of an NPP site. The control of effluent releases is in many cases also a comprehensive task taking into account two principles, namely ALARA and BAT options [8, 9]. A comprehensive review of international conventions related to effluents from NPPs, many of them signed by the members of the EU is given in [9].

Control over effluents is further assured by:

- regular reporting of values of specific radioisotopes in effluents to a regulatory authority
- regular reporting of values of specific radioisotopes in effluents to international organisation as for example EU or other countries
- independent measurements provided by a regulatory authority.

The process of authorisation of effluents from an NPP as well as the monitoring process of effluents is explained in details in [7]. Table 1 from [7] shows basic parameters i.e. activity of radionuclides or activities of a group of radionuclides in a different medium which are reported for the PWR NPPs in EU, annual limits related to liquid and gaseous discharges from NPPs as well as the ranges in which the reported values of annual effluent activities were measured. The data are taken from [4, 5]. When comparing the data from different NPP units, the fact that reported limits related to discharges could be related to the operation of one or more reactors should be taken into account. It should be emphasised that the harmonisation of scope of measurements, measurements of physical parameters of effluents including sampling procedures and reporting within the EU countries has not been achieved yet.

In addition, it should be also taken into account that in cases where no data are given the measurements were not performed, a reported value was zero or measurement results are below “detection limit/decision threshold” as stated in [5].

Table 1: The reported limits of liquid and gaseous effluents from NPPs in the EU in the period 1995- 1994 and reported interval of measured effluents in the period 1995-2003 taken from [4, 5].

Type of Discharge	Group of Radionuclides	Limit of the Activity [4] (GBq/a)	Reported Activity of the Released Effluents [4, 5] (GBq/a)	Generic Annual Discharges [8] (GBq/a)	Normalised Annual Discharges [8] (GBq/GWa)
Liquid Discharge	H-3	2.96E+04 - 1.66E+05 *	5.00E+01- 1.13E+05	1.8E+04	1.6E+04
	Total Beta/Gamma without H-3	3.70E+01 - 2.04E+04	9.40E-06- 8.12E+01	×	×
	Alpha Emitters	2.00E-01	9.10E-07- 6.51E-03	×	×
Airborne Discharge	H-3	2.00E+03- 8.88E+05	2.36E+00- 7.66E+03	9.0E+02	8.4E+02
	Total Beta/Gamma without H-3	4.63E+00- 1.48E+02	6.60E-05- 8.10E-01	×	×
	Total Noble Gas	8.88E+05- 2.96E+06	2.60E+01- 4.67E+04	×	×
	I-131	3.70E+00- 1.63E+01	9.90E-06- 7.75E-01	1.6E-02	1.5E-02
	C-14	3.00E+02- 6.00E+02	1.23E+01- 9.02E+02	2.2E+02	2.0E+02

* The value 1.85E+05 GBq/a was not taken into account due to the fact that it is related just to on NPP unit, namely Burgey 1+2+3+4 and it is well above other reported values.

As stated in [7] the limits for liquid effluents are usually set for activity of H-3 and the total activity of beta/gamma emitters without H-3 which are discharged in a certain period of time. Furthermore, the information related to alpha activity is usually also measured either as the total alpha activity or through key nuclides i.e. Pu-239, Pu-240, Am-241.

The limits of airborne effluents are usually set for activity of H-3, the total activity of particulates excluding iodines, the total activity of noble gases as well as the total activity of iodine isotopes and of C-14. At some NPPs the limits of airborne effluents were related to two kinds of measurements, i.e. measurements of activities of halogens and aerosols and measurements of activity of H-3 and noble gases as it is the case of French NPPs in the reported period 1995-1999 [4]. Generic annual discharges for PWRs from [8] as well as normalised annual discharges are also given in the table. The values are given for electricity generation of 1.07 GWa. The generic discharges for specific radioisotopes in addition to H-3, I-131 and C-14 are also given in [8].

As shown in the table the typical limiting values as well as the typical annual activities of effluents could be within the broad intervals which sometimes span over a few orders of

magnitude. The reason for the width of these intervals could lie in the site specific characteristics which regulate an influence of the operation of an NPP on the population. Furthermore, different methodological approaches applied by the regulatory authority in the assessment of this effluence could be the reason for the observed width of intervals.

In general, the actual activities of annual discharges are usually a few orders of magnitude lower than the prescribed limits except in case of H-3 in liquids and C-14 in air effluents.

The selected radionuclides mainly of radiotoxicity I and II [10] in actual airborne effluents and liquid effluents from PWR NPPs in EU in 2003 were analyses elsewhere [7]. As shown in [7] the total activity of activation product Co-60 discharged in 2003 in liquid effluents was 21.96 GBq and two orders of magnitude less in airborne effluents, namely 7.08E-02 GBq. The total discharge of C-14 in airborne effluent was 14.7E+03 GBq. The total activity of fission product I-131 in airborne effluents was 1.8 GBq and 2.32 GBq in liquid effluents in that year. The total activity of H-3 in liquid effluents was 137 E+04 GBq and two orders of magnitude lower, namely 50.3E+03, in airborne effluents. The total activity of noble gases in effluents was 153.03 E+03 GBq.

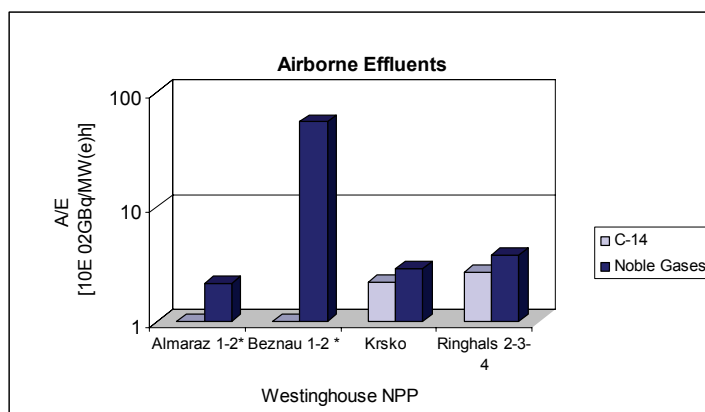
3 EFFLUENTS OF PWR NPP

Besides the type of an NPP, also specific characteristics related to its design could also influence the effluents from an NPP. Today, 11 Westinghouse NPPs are sited in the EU. For comparison the effluents [5] from NPPs from three different states are selected, namely NPP Krško from Slovenia, NPP Almaraz 1-2 from Spain and NPP Ringhals 2-3-4 from Sweden. In addition, the data for the NPP Beznau 1- 2 sited in Switzerland are also taken into account [11]. The comparison is performed for the year 2003 when no major modernisation took place in none of mentioned NPPs. In all of them the outage took place with the average time of 25 days. The details related to the operation of these NPPs are given in table 2. They are taken from [12].

Table 2: The details of the selected NPPs taken for the comparison of effluents in 2003.

NPP	State	Start of Commercial Operation	Maximum Net Capacity in 2003 (MW _e)	Energy Production in 2003 (GW(e)h)
Almaraz 1-2	Spain	1983, 1984	944.0, 953.0	7499.1, 6627.9
Beznau 1-2	Switzerland	1969, 1971	365.0, 365.0	3061.8, 2920.3
Ringhals 2-3-4	Sweden	1975, 1981, 1983	875.0, 915.0, 915.0	6811.5, 6714.6, 6996.5
Krško	Slovenia	1983	676.0	4963.3

The comparison of activity of selected radionuclides in airborne effluents in 2003 is shown in figures 1 and 2. The values are normalised with the production of electricity in that year. Figure 1 shows C-14 and noble gases and figure 2 shows I-131. Activities of C-14 are not reported for Almaraz1-2 and Beznau 1-2. As shown the reported activities for noble gases can span over an order of magnitude. The reported values for activity of C-14 for Krško and Ringhals 2-3-4 are close. Noble gases for both NPPs are also quite close. It can be emphasised that the Beznau NPPs started with operation approximately a decade earlier than the Krško and Ringhals 3-4 NPPs.



* No data are reported for C-14.

Figure 1: The normalised activities of C-14 and noble gases in airborne effluents for selected NPPs in 2003.

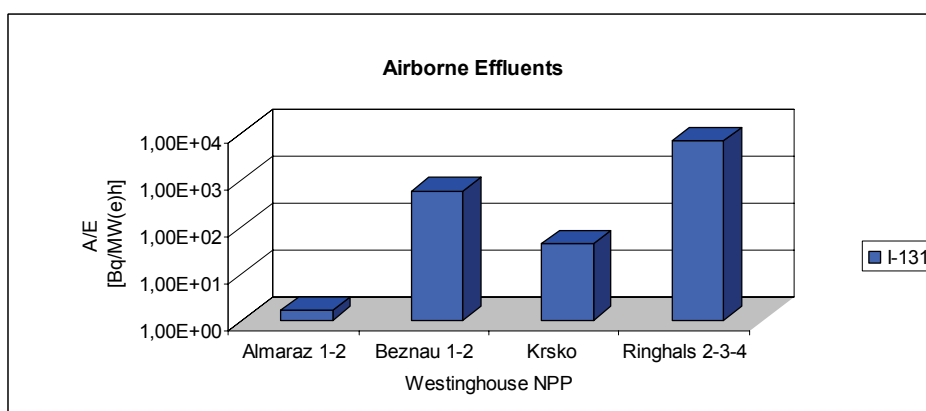


Figure 2: The normalised activities of I-131 in airborne effluents for selected NPPs in 2003.

The comparisons of activity of selected radionuclides in liquid effluents in 2003 are shown in figures 3, 4 and 5. The values are also normalised with the production of electricity in that year. Figure 3 shows normalised activity of H-3 and figure 4 normalised activities of beta and gamma emitters without H-3. The data given in figure 4 for the Krško NPP is based on the identification of radionuclides. Figure 5 shows normalised activity of two selected fission products namely I-131 and Cs-137 as well as activity of activation product Co-60.

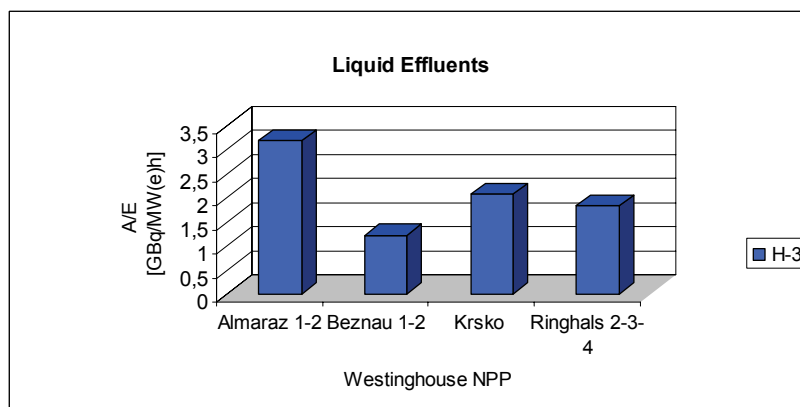


Figure 3: The normalised activities of H-3 in liquid effluents for selected NPPs in 2003.

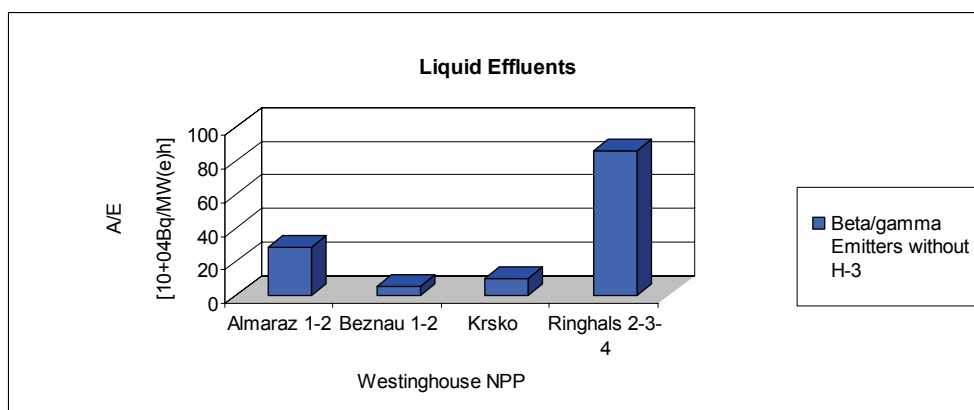
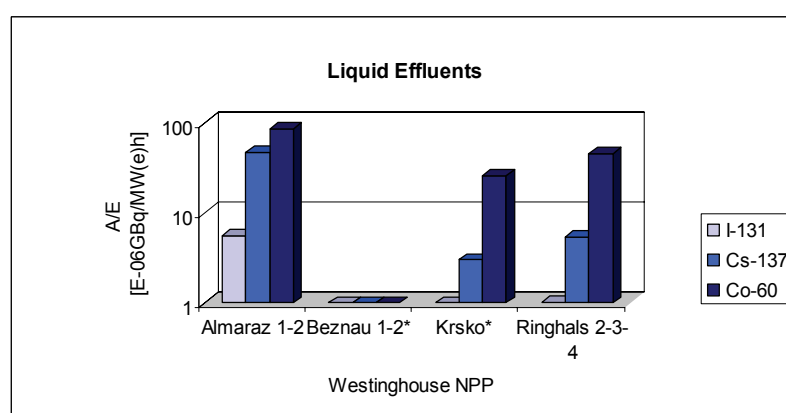


Figure 4: The normalised activities of beta/gamma emitters in liquid effluents for selected NPPs in 2003. The data for the Krško NPP is based on the identification of radionuclides.



* No data are reported for Beznau NPP. The measurement for I-131 for the Krško NPP is below MDA.

Figure 5: The normalised activities of I-131, Cs-137 and Co-60 in liquid effluents for selected NPPs in 2003.

The activities of H-3 are quite close in all NPPs while this is not the case for other radionuclides. It can be concluded that despite no major modifications in the year 2003 which could influence liquid effluents of studied Westinghouse NPPs, the activities of the total beta gamma emitters as well as I-131, Cs-137 and Co-60 can vary substantially. Besides different production of radionuclides in liquids in a specific year the water chemistry in addition to pre-treatment methodology related to BAT applied in specific NPP could also influence these results. Furthermore, measurement procedures as well as sampling techniques for the determination of radionuclide in liquid effluents could drastically influence the results.

4 EFFLUENTS IN THE LIFETIME OF THE KRŠKO NPP

The effluents related to the only NPP in Slovenia, namely the Krško NPP are limited in the licensing document. Additionally the values for specific radionuclides or a group of radionuclides in liquid and airborne effluents are given in the Krško NPP Updated Safety Analysis Report. The PWR Westinghouse NPP entered commercial operation in 1983. It was modernised and upgraded in the period from 1996 to 2000 when both steam generators were replaced. The upgrade of the NPP resulted in electrical output of 700 MW_e. During the refuelling period in 2006 an additional upgrade was accomplished.

The effluents from the Krško NPP are controlled by the Krško NPP laboratories and in addition by authorised laboratories. The reports of measurements are reported to the Slovenian Nuclear Safety Administration, the responsible regulatory body for the control of effluents from the nuclear facility. The results of the monitoring of effluents are also available to the public [13].

In view of prolonged lifetimes of many operating NPPs in the world it is important to recognise the effect of different operational experiences on effluents. Based on the data from [13] five lifetime periods can be identified for the Krško NPP:

- The start-up of the Krško NPP resulted in quite high values of some radioisotopes in liquid effluents, namely gamma/beta emitters without H-3, particularly activation product Co-60 and fission products Cs-137 as well as I-131. In addition, the highest value of C-14 in gaseous effluents in all lifetime of the NPP was achieved in the period which spans from 1983 to 1985.
- The period of stability in operation of the Krško NPP regarding effluents spans in the period 1986 - 1992.
- Due to the leakage of fuel elements in the years 1993 to 1997 a substantial increase of activity of noble gases could be seen in that period. The maximum value of the activity of noble gases was 11.5% of the prescribed annual limit, which is given to be 110 TBq of Xe-133 equivalent, was reached in 1995. In addition, the average value of activity of fission product I-131 in the liquid effluents could be observed.
- In the period of the main modernisation of the NPP no specific trends could be seen, but just after the modernisation a small increase of Co-60 could be detected.
- From 2003 to 2005, an increase of H-3 in liquid effluents can be seen. In that period a change in the duration of the fuel cycle took place which was step by step changed from 12 months to 18 months.

5 CONCLUSIONS

Big differences of discharged activities for certain radionuclides or a group of radionuclides from NPPs were observed. Several reasons could be identified.

It should be also emphasised that no harmonised approach to the measurement techniques including sampling procedures for the determination of effluents in NPPs is in place and no harmonised reporting is available. In addition, it was identified that the scope of the control of effluents has not been harmonised yet. First steps in harmonisation in the EU have been already done [3].

The main factor influencing the activity of radionuclides in effluents is a type of the reactor. In addition, the material used by a producer of an NPP or systems of an NPP can be the main factor for the existence of specific radionuclides in effluents. The operational regimes i.e. duration of a fuel cycle, leakages, outages as well as water chemistry and use of BAT for treatment of effluents could essentially influence the absolute values of activities of some radionuclides.

While no general study of operational regimes, pre-treatment technologies and subsequent effluents for NPPs in Europe is available yet, a first study identifying a lifecycle of an NPP in relation to effluents could be done. The detailed study of effluents from several NPPs could be a useful tool for operators of an NPP to further optimise the releases from the facilities.

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