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Impact of operational changes on the scaling factors of radioactive wastes from the IEA-R1 research reactor

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Abstract Nowadays, the scaling factor methodology is widely used in order to estimate the activity concentration of difficult to measure nuclides in low- and intermediate-level waste from nuclear reactors. However, very few experimental studies evaluate how operational changes in the reactors affect scaling factors. The present work examines the impact of operational changes on the scaling factors that were determined for spent ion-exchange resins and spent activated charcoal permanently withdrawn as radioactive wastes from the water cleanup system of the IEA-R1 nuclear research reactor.

Keywords Research reactors · Radioactive waste assay · Difficult to measure nuclides · Scaling factor methodology

Introduction

The IEA-R1 is a 5 MW pool-type nuclear research reactor, moderated and cooled by light water, which achieved its first criticality in September 1957. The core of the IEA-R1 reactor employs four fork-type control rods of Ag–In–Cd alloy, in proportion of 80, 15, 5% respectively, with a thin cladding of metallic Ni. Located at the Nuclear and Energy Research Institute (IPEN/CNEN-SP), this reactor is still currently in use for scientific research and production of radioisotopes [1–4].

It is important to highlight the significant changes that occurred in the IEA-R1 reactor along its operational history. Despite having been designed to operate at 5 MW, until 1961 the reactor operation schedule was irregular and the power varied between 200 kW and 2 MW. From 1961 onward, the reactor operated at 2 MW during business hours. Between 1971 and 1991, several modifications were introduced in order to adapt the facility to the latest safety standards. In 1995, it was decided that the reactor should be enabled to operate at the design power of 5 MW. After several refurbishments and modernization projects, the reactor began to operate weekly during 64 consecutive hours at steady power in the range from 3 up to 5 MW [5].

All these changes in operating power and shift length overlapped over the years with several changes in the nuclear fuel regarding chemical composition, density and U enrichment [1, 2, 5]. On the other hand, occurrences of nuclear fuel failures were rare, with only one major event recorded in 2001 [6–8].

The most significant recent changes in the IEA-R1 reactor were carried out between mid-2002 and early 2003, when control rods with breached cladding were replaced by flawless new ones and some graphite reflectors with Al cladding were replaced by Be reflectors [5].

These broad operational changes are expected to influence strongly the activity concentrations of nuclides in the radioactive wastes. However, since the beginning of the IEA-R1 reactor operation, ion-exchange resins and activated charcoal beds were replaced only twice, in 1993 and 2003, generating the radioactive wastes under consideration in this work [5, 9]. Consequently, the activity concentrations measured in each waste stream would reflect

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the average of many variations that occurred during those long periods.

Recently, the inventory of radionuclides in these spent ion-exchange resins and spent activated charcoal beds was estimated successfully in the scope of a radioactive waste characterization program [5, 9] employing the scaling factor methodology [10–13]. A detailed analysis of the results shows that significant operational changes in the reactor (power, shift length, nuclear fuel type, fuel failure event, control rods with breached cladding) do have influence on the average values of scaling factors [5, 9].

This work describes and analyses the impact of operational changes on the average values of scaling factors determined for radioactive wastes consisting of spent ion-exchange resins and spent activated charcoal that were permanently withdrawn from the water cleanup system of the IEA-R1 nuclear research reactor.

Methods

Scaling factor methodology

Scaling factor is defined as the ratio of A_{DTM} and A_{KN}, which are respectively the activity concentrations of a difficult to measure nuclide (DTM) and of a key nuclide (KN) [5, 9–13]:

$$SF = \frac{A_{DTM}}{A_{KN}}. \quad (1)$$

Difficult to measure nuclide is the generic designation of a radionuclide that present at least one of the following characteristics [9]: (a) emit no photons (gamma-rays or X-rays) at all; (b) emit photons with low energy and/or low absolute emission intensity; (c) have very low activity among the radioactive waste. On the contrary, key nuclide designates a radionuclide whose radioactive decay is accompanied by emission of higher energy gamma-rays and which is present among the radioactive waste with an activity high enough to be detected outside each package enclosing radioactive waste. Therefore, in most cases, the determination of A_{DTM} requires radiochemical analyses of radioactive waste samples, whereas A_{KN} can be determined directly by gamma-ray spectrometry [9].

Usually, a difficult to measure nuclide that is generated as activation product is correlated to the key nuclide ^{60}Co and a difficult to measure nuclide that is generated as fission product or is an isotope of U or an isotope of a transuranic element is correlated to the key nuclide ^{137}Cs. However, it should be emphasized that, beyond production route and physicochemical similarity, also complex transport phenomena affect activity concentrations of radionuclides in radioactive wastes [13]. This is the reason why scaling factors are empirically obtained correlations specific for each nuclear facility, operational regime, radioactive waste stream and pair of DTM/KN nuclides [9, 13].

The results for the ratio shown in Eq. (1) may differ by orders of magnitude. Therefore, it is necessary to calculate the scaling factor average value as the geometric mean of a series of N measurements [5, 9–13]:

$$\overline{SF} = e^{\left(\frac{\sum_{i=1}^{N} \ln (SF)_i}{N}\right)}, \quad (2)$$

whereas the 2σ-dispersion of the results around the geometric mean is calculated by the equation [5, 9–13]:

$$D_{2\sigma} = e^{2\cdot\left(\sqrt{\frac{\sum_{i=1}^{N} [\ln (SF)_i - \ln (\overline{SF})]^2}{N-1}}\right)}, \quad (3)$$

A scaling factor is considered valid if $D_{2\sigma} \leq 10$ [10, 11], which means that 95.5% of the measurements results must lie within the range [5, 9–13]:

$$\frac{\overline{SF}}{10} \leq (SF)_i \leq \overline{SF} \cdot 10. \quad (4)$$

When the scaling factor that has been obtained can not be considered valid, a correlation function (CF) between the activity concentrations of the difficult to measure nuclide and of the corresponding key nuclide is searched, through regression analysis of logarithms, according to the equation [5, 9, 11–13]:

$$A_{DTM} = a \cdot (A_{KN})^b, \quad (5)$$

that can be rewritten in the form [5, 9, 11–13]:

$$\ln(A_{DTM}) = \ln(a) + b \cdot \ln(A_{KN}), \quad (6)$$

where $\ln(a)$ and b are the regression coefficients. Using the notations $x_i \equiv \ln(A_{KN})_i$ and $y_i \equiv \ln(A_{DTM})_i$ in Eq. (6), a measure of the degree of correlation between the two activity concentrations involved is most properly provided by a quantity called the correlation coefficient [5, 9, 14]:

$$r = \frac{\sum_{i=1}^{N} x_i y_i - \frac{\left(\sum_{i=1}^{N} x_i\right)\cdot\left(\sum_{i=1}^{N} y_i\right)}{N}}{\sqrt{\left[\left(\sum_{i=1}^{N} x_i^2 - \frac{\left(\sum_{i=1}^{N} x_i\right)^2}{N}\right)\right]\cdot\left[\left(\sum_{i=1}^{N} y_i^2 - \frac{\left(\sum_{i=1}^{N} y_i\right)^2}{N}\right)\right]}}. \quad (7)$$

The correlation function is considered valid only if the correlation coefficient for the fit is $r \geq 0.60$ [9, 13].

However, if the correlation coefficient $r < 0.60$, it is assumed that there is no correlation between the activity concentrations of the difficult to measure and the key nuclide. In this case, the arithmetic mean of the results

should be used as an estimate of the activity concentration of the difficult to measure nuclide in that radioactive waste stream [9, 13].

In order to attain statistically significant scaling factors or correlation functions for a given representative data set, the recommended number N of A_{DTM}/A_{KN} measurements is at least 15 [15] and ideally in the range 20–40 [16].

Impact of reactor operational changes on scaling factors

Although there are theoretical models that describe the impact of reactor operational changes on some scaling factors [13, 17–20], experimental information about this subject is scarce. The very few measurements available [21, 22] are restricted to some radioactive waste streams from the two most common nuclear power reactors: pressurized water reactors (PWR) and boiling water reactors (BWR). These measurements demonstrate that significant operational changes in a given nuclear reactor do have the capacity to change the average value of some scaling factors [21, 22]. However, the impacts are almost always within the overall precision of the scaling factor methodology, which means that the 2σ-dispersion of results is generally lower than or equal to ten ($D_{2\sigma} \leq 10$) [21, 22].

A similar behavior is observed in scaling factors determined for radioactive wastes from the IEA-R1 reactor [5, 9]. Regarding spent activated charcoal, the ratio between the average values of scaling factors determined for the 1993 and 2003 waste batches is lower than 10 for all 15 difficult to measure nuclides researched (^{14}C, ^{55}Fe, ^{59}Ni, ^{63}Ni, ^{90}Sr, ^{108m}Ag, ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{238}Pu, $^{239+240}Pu$, ^{241}Pu and ^{241}Am), with exception of ^{238}U [5, 9]. Impacts of reactor operational changes on average values of scaling factors are more notable in spent ion-exchange resins, although the ratio between these values for the 1993 and 2003 batches exceeds 10 only for ^{234}U, $^{235+236}U$, ^{238}U and ^{241}Am among all 12 difficult to measure nuclides researched (^{59}Ni, ^{63}Ni, ^{90}Sr, ^{234}U, $^{235+236}U$, ^{238}U, ^{238}Pu, $^{239+240}Pu$, ^{241}Pu and ^{241}Am) [5, 9]. For just one difficult to measure nuclide researched in spent ion-exchange resins, ^{55}Fe, no valid scaling factors were found and only correlation functions have been obtained [5, 9].

Therefore, as far as the radioactive wastes from the IEA-R1 reactor are concerned, the following directive was adopted [5].

In the spent ion-exchange resins, generally a larger variation in the average values of scaling factors was observed [5, 9], leading to the decision to use the results of each waste batch separately to estimate the radionuclide inventory of this waste stream [5]. The exceptions in this case are the correlation functions involving ^{55}Fe, which have been obtained considering both batches together, because this was the only manner to correlate this difficult to measure nuclide with the key nuclides [5, 9].

On the other hand, a smaller variation in the average values of scaling factors was observed in the spent activated charcoal [5, 9]. This fact, together with the smaller number of data from the 2003 waste batch, leaded to the decision to use the average result of both batches to estimate the radionuclide inventory of this waste stream [5].

Comparative statistical analysis

Aiming to perform a more detailed analysis of the impact that operational changes exert on average value of scaling factors, an additional approach was adopted in this work to study the results obtained by the scaling factor methodology for radioactive wastes from the IEA-R1 reactor. In this approach, the differences between the data sets of the two radioactive waste batches, generated in the two campaigns of replacement of the reactor water cleanup media in 1993 and 2003, were analyzed with the statistical tools 'analysis of sample variance' and 'graph of sequential control' [5].

The analysis of sample variance consists of comparing two different estimates of the variance common to two data sets using the F-function and the probability P that the average data of each set are equal. A small probability value ($P < 0.05$) leads the hypothesis of equal average to be rejected, while a great probability value ($P \geq 0.05$) indicates that the hypothesis of equal average can not be excluded.

Another statistical tool used in this work is the graph of sequential control, which shows the values of a given data set over time [23]. This graph has a center line, as well as a lower and an upper limit of control. The central line represents the average value of the measurements, while the control limits, indicated by horizontal dashed lines, are employed to separate and identify any unusual points. This graph is used in order to observe the properties of the data set and, therefore, to verify whether two data sets are stable or change over time. Therefore, each point in the abscissa axis is the number of a radioactive waste sample and the dashed vertical line that divides this axis indicates which samples belong to a given batch (either 1993 or 2003). A data set or a process is considered statistically stable only if it presents natural variation, without any cycles or pattern [24].

In this work, the STATISTICA software [25] was employed to perform such comparative statistical analysis with the data obtained for each activity concentrations ratio involving a difficult to measure nuclide and a key nuclide (either ^{60}Co or ^{137}Cs)—designated as DTM/KN ratio—taking into account separately waste batches (1993 and 2003) and waste streams (spent ion-exchange resins and spent activated charcoal) from the IEA-R1 reactor. The analysis of sample variance compared the means of

two data sets using box diagrams that indicated the average data for the DTM/KN ratios and the values of the probability P of equality for the two batches and the two streams of radioactive waste. The graphs of sequential control were obtained from the data organized chronologically and provided information on the stability of the DTM/KN ratios based on the observed variation of average data.

Results and discussion

Regarding spent ion-exchange resins, the evaluation of the data by means of the two previously mentioned statistical tools indicated that, for most of the 20 DTM/KN ratios measured, the average value changed between 1993 and 2003 waste batches. Exceptions were ^{59}Ni/^{60}Co, ^{59}Ni/^{137}Cs, ^{63}Ni/^{60}Co, ^{63}Ni/^{137}Cs, ^{238}Pu/^{60}Co, ^{238}Pu/^{137}Cs and ^{241}Pu/^{137}Cs. The average values of all the DTM/KN ratios in the 1993 waste batch were higher than the corresponding average values in the 2003 waste batch, primarily due to the increase in operating power of the IEA-R1 reactor during the period of the 2003 batch [5, 9].

The most representative box diagrams and graphs of sequential control obtained for spent ion-exchange resins are shown side by side in Figs. 1 and 2, presenting the DTM/KN ratios: (a) with the highest probability P (^{63}Ni/^{60}Co and ^{63}Ni/^{137}Cs); (b) that include a fission product for which a valid scaling factor was obtained (^{90}Sr/^{60}Co and ^{90}Sr/^{137}Cs) and (c) for which the greatest difference between the two waste batches was found (^{238}U/^{60}Co and ^{238}U/^{137}Cs) [5, 9].

An analogous evaluation was performed for spent activated charcoal, with results indicating likely changes in the average for most of the 28 DTM/KN ratios, except ^{14}C/^{60}Co, ^{14}C/^{137}Cs, ^{59}Ni/^{60}Co, ^{59}Ni/^{137}Cs, ^{63}Ni/^{60}Co and ^{63}Ni/^{137}Cs. The graphs of sequential control showed variation between batches, but due to the wide dispersion of results and fewer data for the 2003 waste batch, the changes are not as pronounced as in spent ion-exchange resins.

The most representative box diagrams and graphs of sequential control obtained for spent activated charcoal are shown side by side in Figs. 3 and 4, presenting the DTM/KN ratios: (a) with the highest probability P (^{63}Ni/^{60}Co and ^{63}Ni/^{137}Cs); (b) with the activation product whose scaling factors are considered [21] the most sensitive to operational changes in nuclear power plants (^{55}Fe/^{60}Co and ^{55}Fe/^{137}Cs) and (c) for which this study found a marked increase in the 2003 waste batch compared with the 1993 waste batch (^{108m}Ag/^{60}Co and ^{108m}Ag/^{137}Cs) [5, 9].

Some remarkable aspects of these results are discussed below.

The stability of the average value of the scaling factor of ^{63}Ni to ^{60}Co was observed in both waste streams and batches considered in this work. Such characteristic is in good agreement with a trend already observed in radioactive waste from PWR and BWR nuclear power reactors, in which the scaling factor of ^{63}Ni to ^{60}Co is considered valid for all radioactive waste streams generated in a specific nuclear power plant [10]. This stability evidences that the Ni and Co isotopes have a common main origin [11], which in the case of the IEA-R1 reactor is the thin metallic Ni cladding that coats the control rods [5, 9, 26].

In the case of the ^{90}Sr/^{60}Co ratio, there are significant differences between the chemical behavior of Sr and Co that become more evident as a result of changes in reactor operation power attained within a short period of time (transient), as observed in PWR reactors in France [27]. A marked difference in scaling factors of ^{90}Sr to ^{60}Co between the 1993 and the 2003 waste batches is clearly observed on spent ion-exchange resins from the IEA-R1 reactor [5]. This difference reveals the contrast between the regularity of the reactor operation up to 1993 (operation power of 2 MW during business hours) and the variation of the reactor operating regime mainly after 1995 (operation power varying between 3 and 5 MW in weekly shifts of 64 consecutive hours) [5].

All scaling factors involving U isotopes presented a marked decrease in the 2003 waste batch compared with the 1993 waste batch, whereas for Pu and Am isotopes this variation was much smaller [5, 9]. As mentioned previously, after 1995 there was an increase in both operating power and shift length of the IEA-R1 reactor. At the same time, there were no significant occurrences regarding the nuclear fuel, either in terms of failures or in the observed rates of surface contamination, when both remained very low. Moreover, in that period, there were no changes in the general operating conditions of the water cleanup system. Under these circumstances, the activity concentrations of U isotopes decrease because they are practically only consumed during irradiation, while the activity concentrations of Pu and Am isotopes, as well as of the key nuclides ^{60}Co and ^{137}Cs, increase because they are more produced than consumed during irradiation [28]. Therefore, the trend observed in the obtained scaling factors in the 2003 waste batch, as compared to the 1993 waste batch, is a marked decline of the DTM/KN ratios for the nuclides ^{234}U, ^{235}U, ^{236}U, $^{235+236}$U and ^{238}U, whereas a less marked variation arises in the DTM/KN ratios for the nuclides ^{238}Pu, $^{239+240}$Pu, ^{241}Pu and ^{241}Am.

Although ^{55}Fe and ^{63}Ni are both activation products, a comparison between the two batches of spent activated charcoal from the IEA-R1 reactor showed that the variation

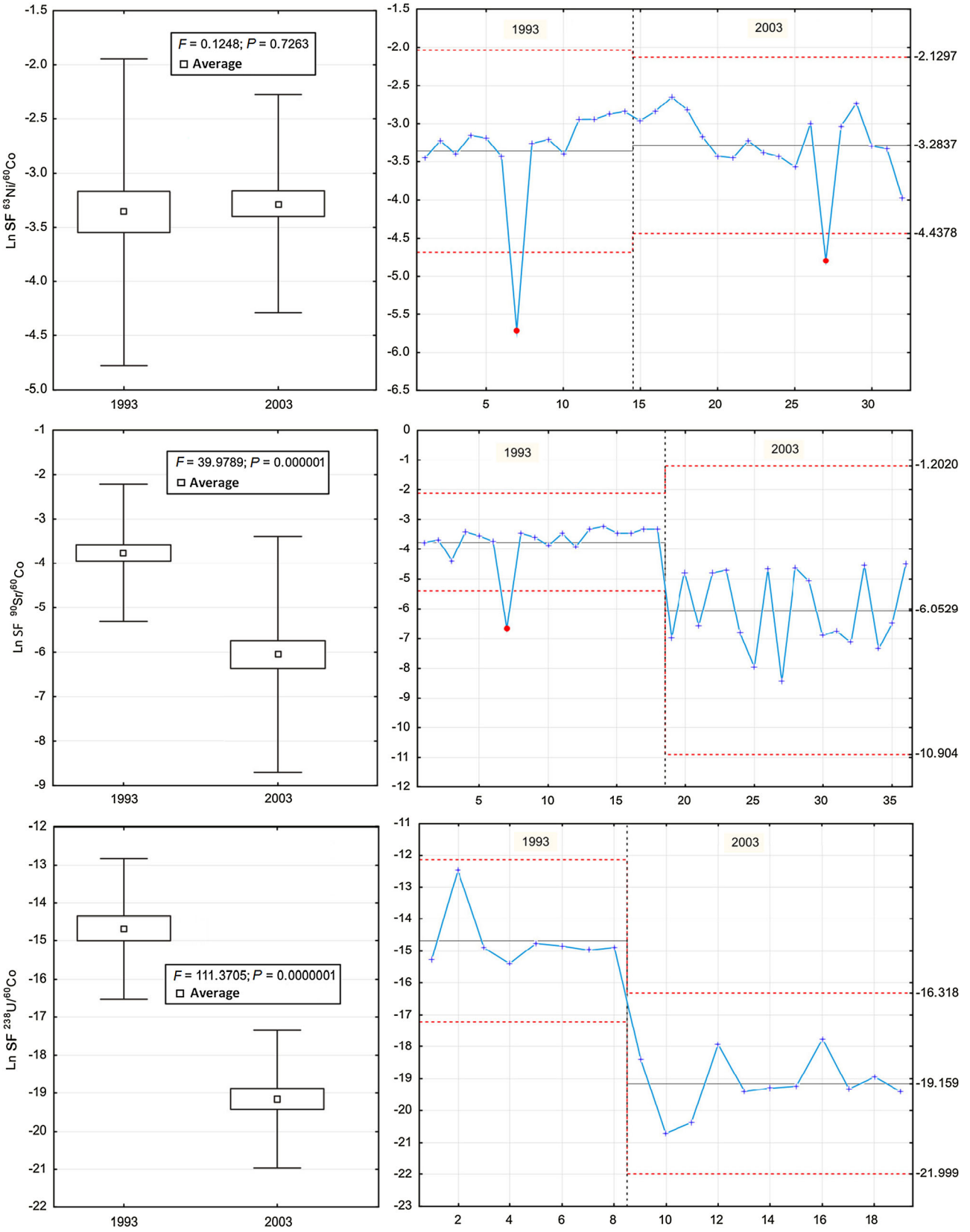

Fig. 1 *Box diagrams* and *graphs* of sequential control for the $^{63}Ni/^{60}Co$, $^{90}Sr/^{60}Co$ and $^{238}U/^{60}Co$ logarithmic ratios in spent ion-exchange resins including the two radioactive waste batches

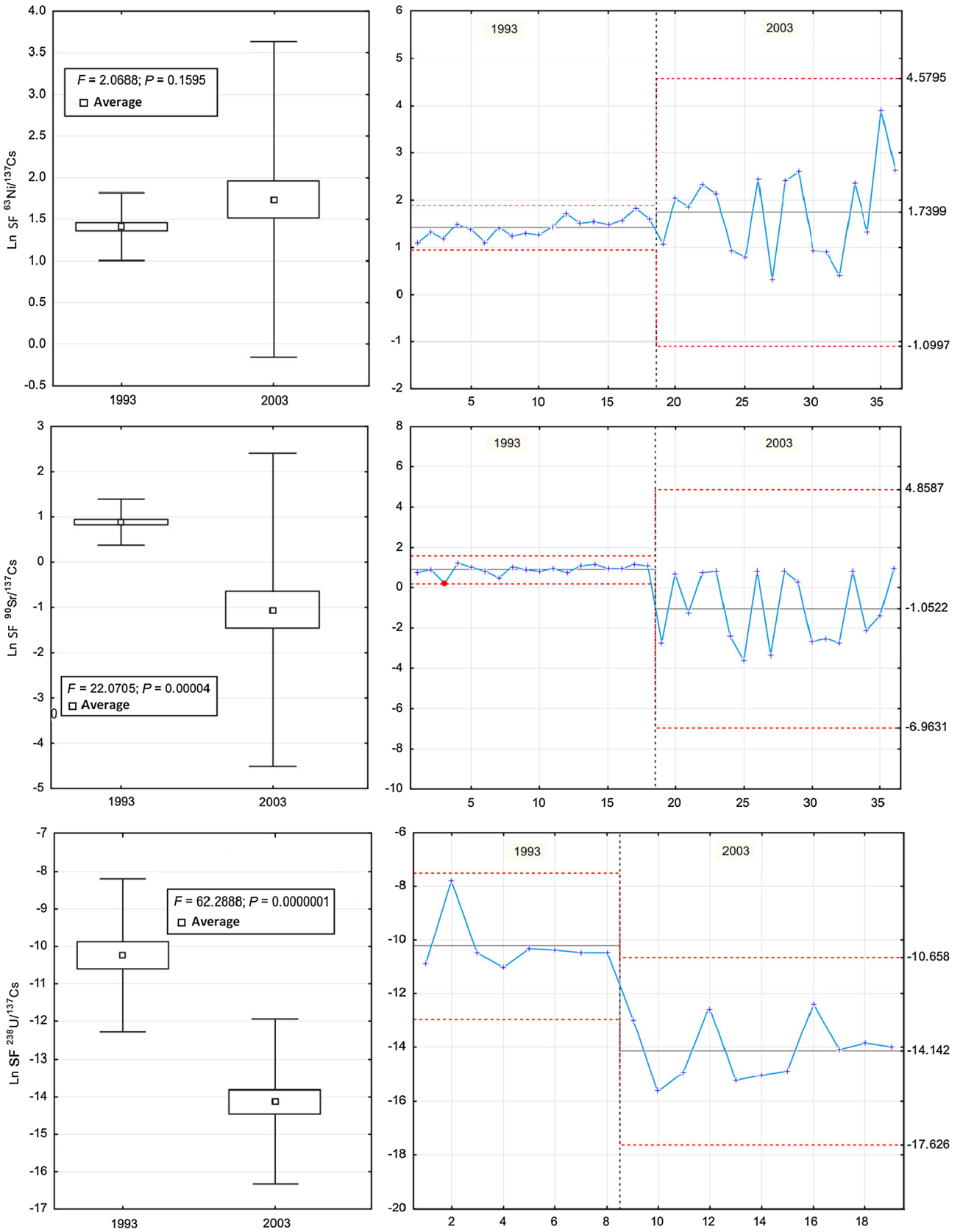

Fig. 2 *Box diagrams* and *graphs* of sequential control for the $^{63}Ni/^{137}Cs$, $^{90}Sr/^{137}Cs$ and $^{238}U/^{137}Cs$ logarithmic ratios in spent ion-exchange resins including the two radioactive waste batches

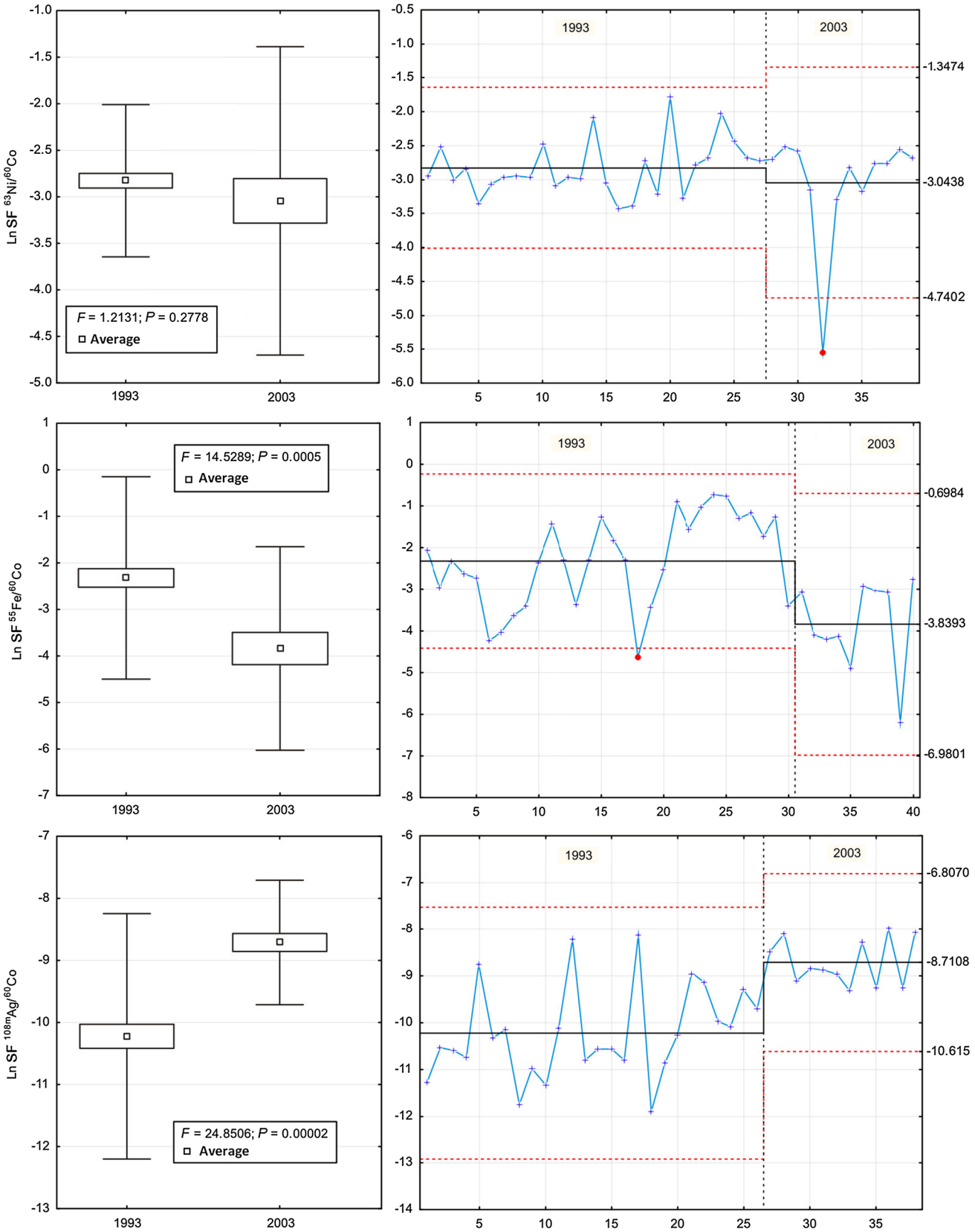

Fig. 3 *Box diagrams* and *graphs* of sequential control for the $^{63}Ni/^{60}Co$, $^{55}Fe/^{60}Co$ and $^{108m}Ag/^{60}Co$ logarithmic ratios in spent activated charcoal including the two radioactive waste batches

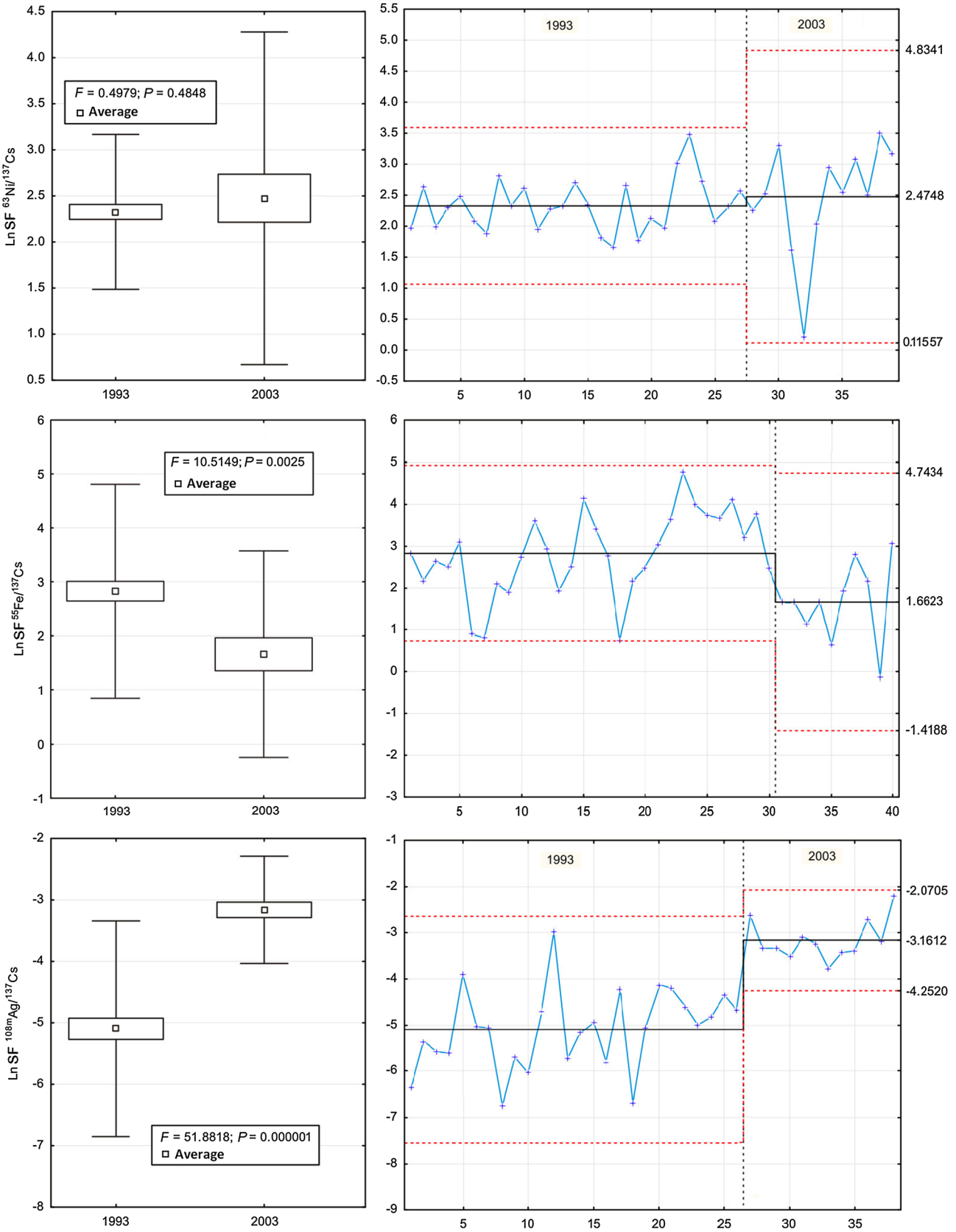

Fig. 4 *Box diagrams* and *graphs* of sequential control for the $^{63}Ni/^{137}Cs$, $^{55}Fe/^{137}Cs$ and $^{108m}Ag/^{137}Cs$ logarithmic ratios in spent activated charcoal including the two radioactive waste batches

of the scaling factor of ^{55}Fe to ^{60}Co was approximately 3.5 times higher than for the scaling factor of ^{63}Ni to ^{60}Co [5, 9]. Moreover, it should be emphasized that no valid scaling factors were found for ^{55}Fe in spent ion-exchange resins from the IEA-R1 reactor [5, 9]. These results agree with the behavior observed in PWR and BWR reactors, where ^{55}Fe is chosen as a surrogate for activation products since it is observed to be more sensitive to reactor operational changes and respond to them more consistently than ^{63}Ni [21].

The nuclide ^{108m}Ag is a special case in this work. It was the only difficult to measure nuclide with a pronounced increase in the average value of the scaling factors from the 1993 to the 2003 waste batches, while the overall trend for all the other radionuclides of its kind was a decrease. Such behavior can be explained by the flaking and pit corrosion in the thin Ni cladding of all the IEA-R1 reactor control rods—revealed during visual inspections performed in 1998, 2000 and 2001—and is consistent with gamma spectrometry measurements that detected ^{110m}Ag in the IEA-R1 reactor pool water along the same period [2]. These facts expedite the replacement of the flawed control rods in 2003 [5, 9].

Conclusion

A comparative statistical analysis, that was carried out for two batches of spent ion-exchange resins and spent activated charcoal permanently withdrawn as radioactive wastes from the water cleanup system of the IEA-R1 nuclear research reactor, evidenced the impact of reactor operational changes on scaling factors.

Each radioactive waste batch is associated to a distinct period of the IEA-R1 reactor operation regime, concerning mainly to the operating power and the length of the weekly operation shifts.

Many operational changes occurred simultaneously in the IEA-R1 reactor during each long period that corresponds to one of the radioactive waste batches. Due to this overlapping, it was not possible to assign quantitatively and exactly the impact on a given scaling factor to just one specific operational change. Nevertheless, qualitative evidences indicate the prevalence of impacts related to: (a) variations in reactor operation power attained within a short period of time on the scaling factor of ^{90}Sr to ^{60}Co, (b) increase in operating power and shift length on the scaling factors of U, Pu and Am isotopes and (c) flaking and pit corrosion in the thin cladding of reactor control rods on the scaling factors of ^{108m}Ag.

For most of the difficult to measure nuclides researched among radioactive wastes from the IEA-R1 reactor, variation in the average values of scaling factors was larger in spent ion-exchange resins than in spent activated charcoal. As a consequence, it was decided to use results of each waste batch separately in order to estimate the radionuclide inventory of the spent ion-exchange resins, whereas the average result of both waste batches was used in order to estimate the radionuclide inventory of the spent activated charcoal.

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