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Monitoring of atmospheric emission of alpha emitters radionuclides at a nuclear fuel cycle facility in RJ, Brazil

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ABSTRACT

The Indústrias Nucleares do Brasil S/A (INB) in Resende, state of Rio de Janeiro, operate with open sources containing alpha emitters. This operation produces particulate material in the air. To reduce this impact, the plant operates with negative pressure using an exhaust equipment. Exhaust air is sent to an absolute filter system. In the exhaust stack, there is an alpha emitter detection module that operates monitoring the releases. Following the dose calculation model established by the National Commission for Nuclear Energy (CNEN), the authorized limit is fixed at $1.2 \cdot 10^7$ Bq.y^{-1} of alpha emitters by Nuclear Fuel Factory (FCN). The present work aims to evaluate the alpha-emitting radionuclides released by FCN in terms of total release (Bq) and activity concentration (AC, Bq·m^{-3}). In 2018, FCN released $1.41 \cdot 10^6$ Bq.y^{-1}(*i.e.* 34% of the 2016 releases). The largest mean for AC was 2.04 mBq·m^{-3}. The release data was almost an order of magnitude lower than the annual release limit, proving the adequation of the atmospheric effluents to the licensing requirements.

Keywords: **environmental Radioprotection, Alpha emitter, nuclear fuel cicle.**

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1. INTRODUCTION

Manufacturing facilities during their operations may impact the environment. In order to protect the environment, official agencies issue regulations defining the amounts of release that make the impact negligible, allowing the factory operation, nature conservation and quality of the surrounding environment, and the health of the surrounding population [1-5].

The Nulcear Fuel Factory, at its Unit II (NFF-II), operates in Resende (RJ) tons of uraniumn in a number of chemical forms. The various industrial operations generate particulate material containing uranium in its composition. As a Collective Protective Equipment (CPE), the plan operates at negative pressure, being able to remove 6 times by day the internal atmosphere of NFF-II [6]. With the used inputs, the environmental dispersion model and dose assessment among members of the public, pointed out the alpha-emitting radionuclides as critical radionuclides, i.e., the generators of higher doses. Beta-emitting radionuclides add traits in the addition of the doses received, being less than the uncertainty of the dispersion model and dose evaluation being, therefore, neglected from the radioprotection point of view [7].

In addition, there is a system that blows into the factory; the same amount of air that is exhaled by the exhaust system. The exhaust air goes through an absolute filter system. At the chimney outlet, there is an online sampling system that measures the activity concentration (AC) of alpha emitters. If the sample exceeds a certain restricted release value, then the exhaustion deflects the flow and passes through a gas scrubbing system to ensure immediate reduction of the concentrations of released activities [6].

The present work aims to evaluate the alpha emitters releases of NFF-II to the atmosphere, in terms of activity concentration and activity released during 2018, and compliance to the operational levels defined by the regulatory agency.

2. MATERIALS AND METHODS

The air from the Nuclear Fuel Factory (NFF-II) is released through a chimney and contains alpha emitters. On the top of the chimney samples of air are captured and analyzed by a Thermo Scientific equipment to measure the emitters. This equipment is endowed of a radiation detector based on "Alpha Energy Range Discrimination" (AERD) system [8], and the counting method employs primary and secondary proportional counter tubes separated by a foil and an electronic evaluation system to optimize the detection of alpha emitters in the air [6, 8].

Data for the year 2018 were organized in Table 1 by month containing the minimum, maximum amd average values od released actiivity concentration (Bq m^{-3}) and the released value of activiy in monthly terms. The dose estimate reported to the regulator must be expressed in terms of annual doses and the values are collected in real time. To estimate the annual average value, data are integrated in daily terms. Daily values are used to estimate the minimum, maximum and average monthly values. This approach generates an artifact in estimating the minimum. If the installation or equipment is not operating during a daily cycle (from 00:01 h to 24:00 h), then the zero value in the minimum value will be reported, meaning no values below the detectable minimum. Data comparison was carried out between released values (Bq) and estimated dose, in monthly terms with the year 2016 from data published by Pereira et al. [6].

Graphs were plotted for visual analysis in order to establish correlations between variables by Pearson's R^2 method and Student's t-test to compare averages [9].

3. RESULTS AND DISCUSSION

Statistical data on the release of alpha emitters into air by NFF-II during 2018 are described in Table 1. The minimum, maximum and average values of activity concentration, all in Bq·m^{-3}, and the amount released in GBq, can also be seen in Table 1. Figure 1 shows the lack of correlation between the monthly average AC and the monthly amount of alpha emitters released. Pearson R^2 value of 0.46, associated with a $P < 0.05$, points to a lack of statistical correlation between the variables, probably linked to a third variable “flow” that was not assessed in this work.

Table 1. Descriptive statistics for AC releases (minimum, maximum and average) in Bq·m^{-3} and for monthly release activity values in GBq, during 2018.

Month	Minimum (Bq·m^{-3})	Maximum (Bq·m^{-3})	Average (Bq·m^{-3})	Release (GBq)
Jan	0.00E+00	2.60E-03	8.22E-04	5.84E-06

Feb	1.00E-04	1.90E-03	4.43E-04	4.66E-06
Mar	0.00E+00	2.10E-03	7.21E-04	1.95E-05
Apr	0.00E+00	1.20E-02	1.07E-03	1.34E-05
May	0.00E+00	2.50E-03	7.69E-04	1.03E-05
Jun	0.00E+00	5.60E-03	9.28E-04	9.08E-06
Jul	0.00E+00	1.80E-02	1.39E-03	1.45E-05
Aug	2.00E-04	1.30E-02	1.96E-03	9.86E-06
Sep	0.00E+00	3.70E-03	1.36E-03	8.60E-06
Oct	4.00E-04	4.40E-03	1.29E-03	8.00E-06
Nov	2.00E-04	1.38E-02	1.62E-03	1.11E-05
Dec	1.00E-04	1.99E-02	2.04E-03	2.62E-05

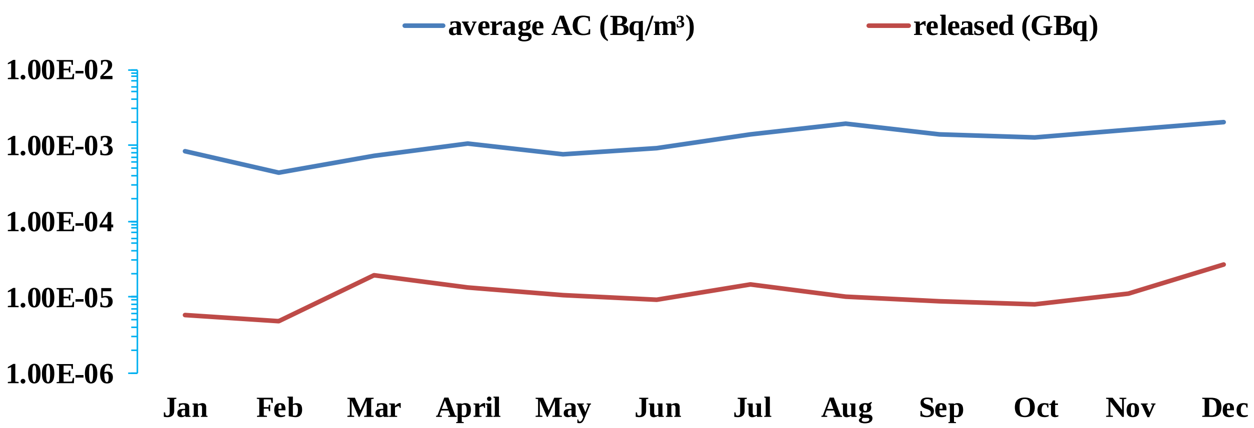

Figure 1 : Values of monthly average concentrations (Bq·m^{-3}) and activity releases (GBq).

The operational limit stipulated for the release of alpha emitters into the atmosphere by the regulatory agency is annual, being 1.2·10^{7} Bq·year^{-1}. The cumulative release assessment can be seen in Figure 2, which shows that the released amounts of alpha emitting radionuclides were approximately one order of magnitude below the upper cited authorized limit. Releasing a total annual amount of 1.4·10^{6} Bq, it was clear that NFF-II meets indeed the operational limit imposed in terms of release. Again, Person's R^2-based correlation was below significance. Thus, monthly releases vary and are not correlated with the cumulative release variable.

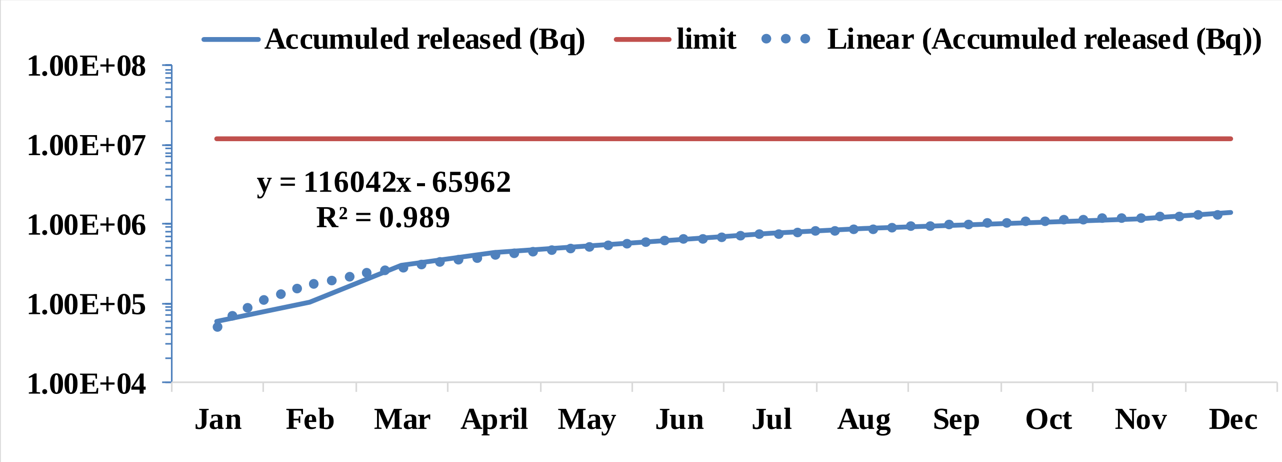

Figure 2 : Monthly releases, cumulative release and operational limit in 2018.

Comparing the monthly release values of 2018 with those of 2016 reported by Pereira et al. [6], it can be seen that the 2018 monthly values were below the 2016 values (see Table 2). The R^2 of Pearson of 0.23 and $P < 0.01$, obtained from monthly values, demonstrated that the releases are not statistically correlated (see Figure 3). The use of Student's t-test with T_{cal} of 45 and $P < 0.01$ shows the statistical veracity of this inference. The 2018 release was lower than the 2016 one.

Table 2 : Monthly averages of alpha emitters released in 2016 and 2018 and operational limit.

Month	Released_2016 (Bq)	Released_2018 (Bq)	Operational limit(Bq/y)
Jan	2.25E+05	5.84E+04	
Feb	3.54E+05	4.66E+04	
Mar	2.83E+05	1.95E+05	
Apr	4.55E+05	1.34E+05	
May	2.86E+05	1.03E+05	
Jun	2.76E+05	9.08E+04	1.20E+07
Jul	4.71E+05	1.45E+05	
Aug	5.68E+05	9.86E+04	
Sep	2.93E+05	8.60E+04	
Oct	2.70E+05	8.00E+04	
Nov	2.75E+05	1.11E+05	
Dec	3.85E+05	2.62E+05	

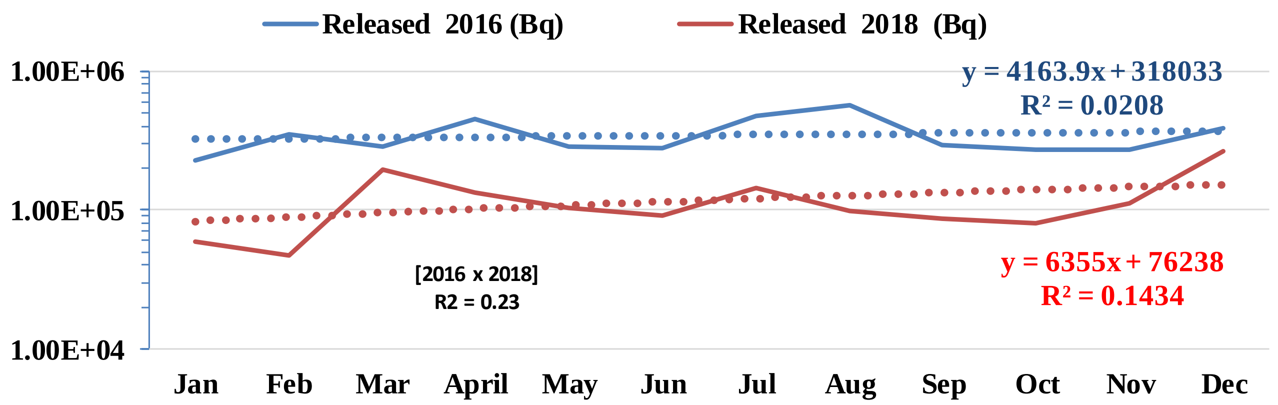

Figure 3 : Monthly variation between releases of alpha emitters in 2016 and 2018, in Bq.

To assess the estimated inhalation doses in the critical group, based on the total release of alpha emitters in NFF-II, the regulatory agency accepted the modeling proposed by INB where a linear relationship between release and dose is stated. In this modeling, the release of $1.20{\cdot}10^7$ Bq·year^{-1} corresponds to 0.3 mSv·year^{-1}. The cumulative dose estimation data for 2018 (this paper) and 2016 [6] can be seen in Table 3. On the whole, the monthly averages for 2016 were 3 times higher than for 2018.

Considering the linear dose-release relationship proposed above, monthly average doses and releases respond in the same way. Therefore, these doses are linearly related. Thus, based on the release analysis, there is no linear relationship between the doses in the critical group and the doses in 2016 are higher than in 2018.

Table 3 : Estimated monthly doses in 2016 [6] and 2018 (this paper).

Month	Accumulated dose rate 2016	Accumulated dose rate 2018
Jan	6.61E-03	1.72E-03
Feb	1.70E-02	3.09E-03
Mar	2.53E-02	8.82E-03
Apr	3.87E-02	1.28E-02
May	4.72E-02	1.58E-02
Jun	5.53E-02	1.85E-02
Jul	6.91E-02	2.27E-02
Aug	8.58E-02	2.56E-02
Sep	9.44E-02	2.82E-02
Oct	1.02E-01	3.05E-02

Nov	1.10E-01	3.38E-02
Dec	1.22E-01	4.15E-02

Evaluating the evolution of the accumulated dose along the year, the correlation between the years becomes irrefutable. With R^2 of 0.99 and P >> 1 % (see Figure 4), the two variables showed a close correlation, showing that, during the year, the evolution of both variables followed the same trend, but the 2018 values were, in the mean, 3 times lower than those of 2016.

Factory production values, which could help to explain this synchronized variation, are not available because they are restricted availability items.

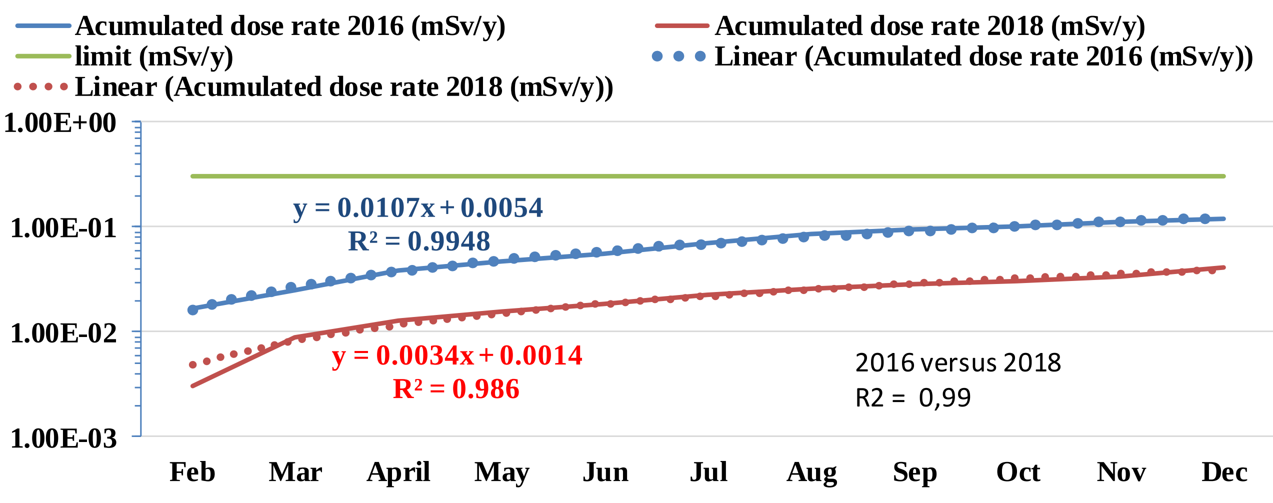

Figure 4. Accumulated dose in 2016 and 2018 and annual dose limit.

Finally, the estimated dose values for 2016 and 2018 were below the operational dose limit values, approximately 1/3 of it, as shown in Figure 5.

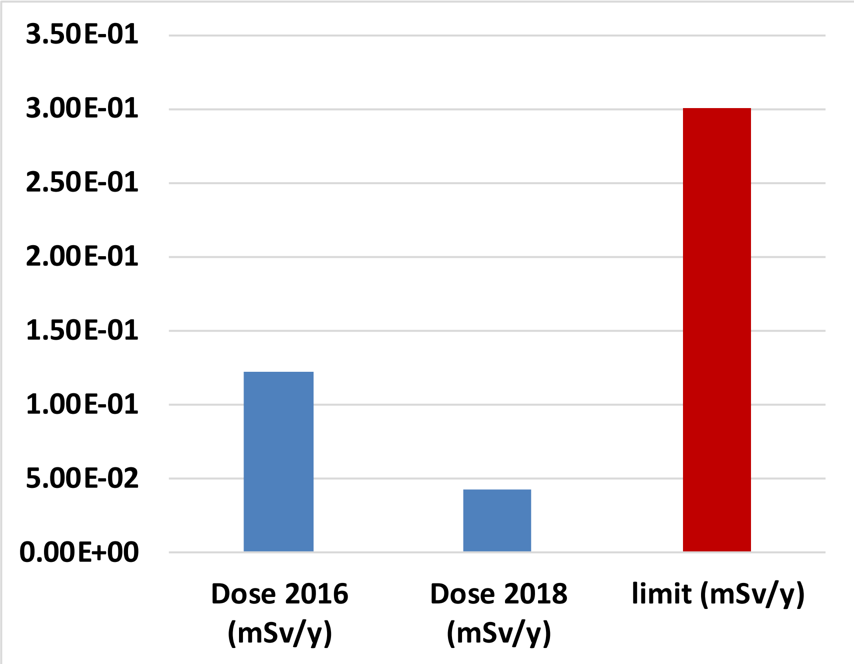

Figure 5. Annual dose rate in 2016 and 2018 compared to operational limit (annual dose rate).

4. CONCLUSION

The release of alpha emitters in 2018 was lower than in 2016. In both cases, the released values were below the operational limit, stipulated by the regulator, as well as the prevision of the doses received by the critical group. The average monthly releases in 2016 and 2018 were different.

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