

## ANALYSIS OF EXPERIMENTAL CAMPAIGNS ON ATMOSPHERIC TRANSFERS AROUND THE AREVA NC SPENT NUCLEAR FUEL REPROCESSING PLANT AT LA HAGUE: COMPARISON BETWEEN OPERATIONAL MODELS AND MEASUREMENTS

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### INTRODUCTION

The harmfulness of pollutants emitted into the atmosphere depends mainly on their ability to become fixed and their concentration in the air. This depends of course on the quantities released, but the state of the atmosphere which conditions the future of these pollutants may also be significant: they can be dispersed (vertically and horizontally), transformed by physical or chemical reactions and deposited (dry and wet depositions).

Institute for Radiological Protection and Nuclear Safety (IRSN) is developing its own dispersion and atmospheric transfer tools for its operational needs during an emergency radiological situation. It is also performing field experiments like around the AREVA NC spent nuclear fuel reprocessing plant at La Hague. The ultimate aim of the experiments is to reduce the uncertainties over the models of atmospheric dispersion and deposition of gases and aerosols emitted by the nuclear industry in normal operating or accident situations.

Series of environmental measurements have been carried out to determine the atmospheric concentration of Krypton-85 (<sup>85</sup>Kr), a rare gas released when reprocessing spent nuclear fuels at La Hague. In addition, bioindicator sampling campaigns (furze, grass) have been performed to have information of the surface activity of the flora for various radionuclides (iodine-129, tritium and carbon-14) released in the form of gases and aerosols. The campaign results are compared with the releases from the AREVA NC La Hague plant and provide a basis for estimating the atmospheric dispersion parameters and the deposition parameters that may be used in the operational models for the La Hague site.

The purpose of this paper is to present the results of the radionuclide concentrations obtained during the field campaigns and to compare them with the results from the dispersion and deposition operational models.

### METHOD

IRSN performed a series of fourteen <sup>85</sup>Kr measurement campaigns around the AREVA NC spent fuel processing plant site at La Hague between June 1997 and April 1998, to determine the Atmospheric Transfer Coefficients (ATC) at the centre of the plume (Maro *et al.*, 2001), followed by a further seven campaigns in May and June 2001 to determine the standard deviation at ground level (Maro *et al.*, 2002). The ATC is the ratio between the concentration in the air and the release rate ( $\text{s}\cdot\text{m}^{-3}$ ). These campaigns were performed for variable distances and meteorological conditions. The environmental measurements were taken between 500 and 4,500 metres from the release points with wind speeds at 100 metres above the La Hague plateau, which were between 4.5 and 16.9  $\text{m}\cdot\text{s}^{-1}$  (the release points of AREVA NC La Hague were 100 metres above the ground). The diffusion atmospheric conditions during the sampling were mainly "neutral or slightly unstable" in the Pasquill classification (1974) (class

C or D). A single measurement campaign was performed during an episode of high stability (class F).

The <sup>85</sup>Kr provides a good assessment of atmospheric concentrations, as a noble gas it is chemically neutral and is not subject to deposit phenomena.

The measurement results were used to determine the ATC and standard deviation of the plume at ground level for the distances in relation to the release point and for the meteorological conditions corresponding to these campaigns.

In addition, furze samples were taken at a point per square kilometre in an area of 70 square kilometre around the La Hague spent fuel processing plant in February 1997, January 1998 and March 1999 (Maro *et al.*, 2000). The last fifteen centimetres of each branch were sampled, corresponding to a growth period of one year in the La Hague region. Subsequently, in April 2000, grass samples were collected with the same geographical distribution. For these four years, the samples were analysed to determine their activity in <sup>129</sup>I, <sup>14</sup>C and <sup>3</sup>H. As the atmospheric releases from AREVA NC La Hague and the meteorological conditions in the La Hague region are known, the results of the atmospheric transfer models (GRNC, 1999) and the measurements were compared. The approach applied is described in Figure 1.

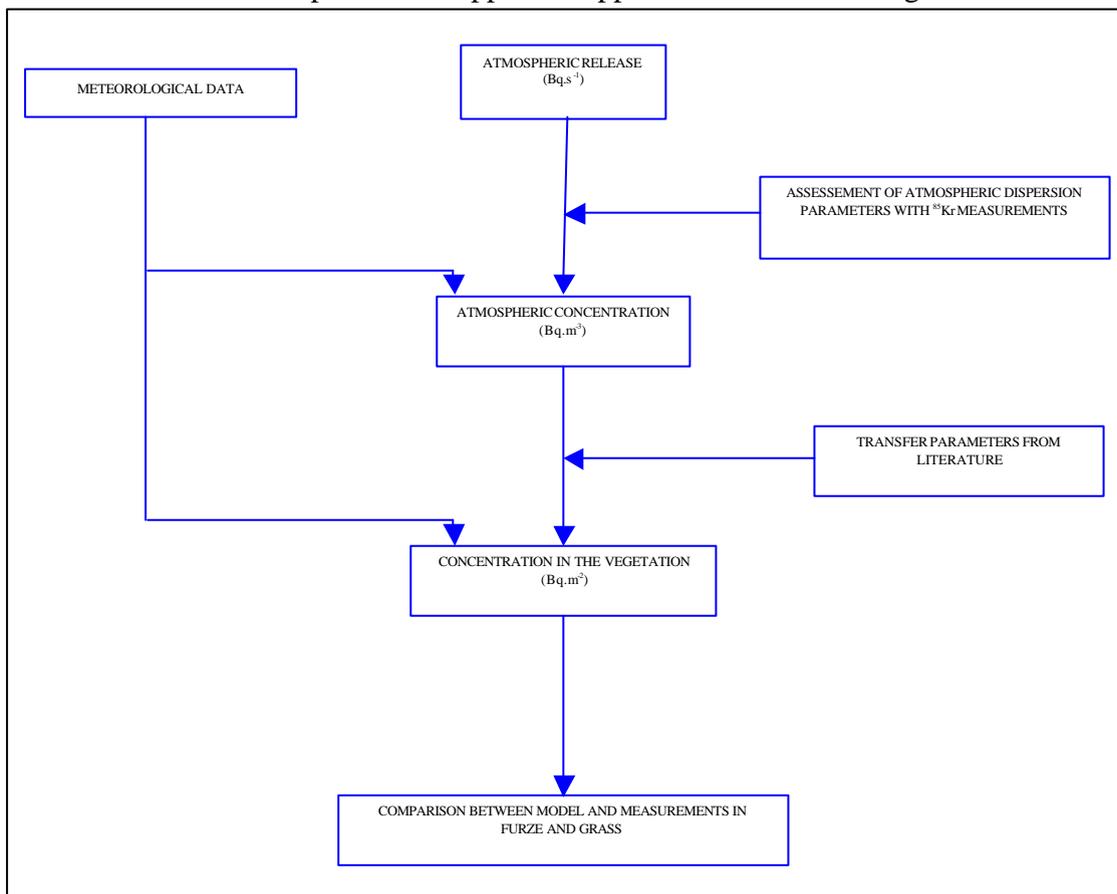


Fig. 1; General approach for model-measurement comparison (GRNC, 1999).

## RESULTS AND DISCUSSION

### Analysis of <sup>85</sup>Kr measurement campaigns

The <sup>85</sup>Kr measurement campaigns between 1997 and 2001 culminated in a set of data grouping the ATC measured according to the distance from the source, the stability class and

the wind speed at the release height. In addition, standard horizontal deviation for various distances covered from the source were calculated. Using the release distance, wind speed and stability class, it was thus possible to fit a Gaussian model to assess the atmospheric concentrations of radionuclides released. Figure 2 shows the comparison between the results of the model and the <sup>85</sup>Kr measurements as function of the distance. The <sup>85</sup>Kr measurements are realised especially where the furze and grass are sampled.

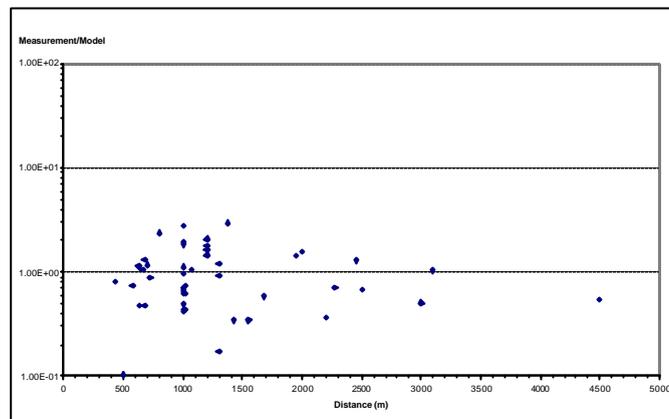


Fig. 2; Comparison between the results of the model and the <sup>85</sup>Kr measurements as function of the distance.

### Analysis of radionuclide measurement campaigns in the bioindicators

Nuclear Safety (GRNC, 1999) stipulates a dry deposition velocity equal to  $5 \cdot 10^{-3} \text{ m}\cdot\text{s}^{-1}$  and a washout coefficient for the wet deposit of  $1 \cdot 10^{-4} \text{ s}^{-1}$  regardless of the radioelement, except for <sup>3</sup>H and <sup>14</sup>C. On the other hand, for <sup>3</sup>H and <sup>14</sup>C, it is considered that there is equilibrium between the amounts of <sup>14</sup>C and <sup>3</sup>H present in the air and the vegetation. In other words, that the amount of <sup>14</sup>C per kilogram of carbon and the amount of <sup>3</sup>H per litre of water are identical in these two environments.

### <sup>3</sup>H

For <sup>3</sup>H, the mean ratio between the theoretical values and the experimental values is 1.3 for 1996, 1.6 for 1998 and 1.6 for 1999 (Figure 3 and Table 1). The model slightly over-estimates reality.

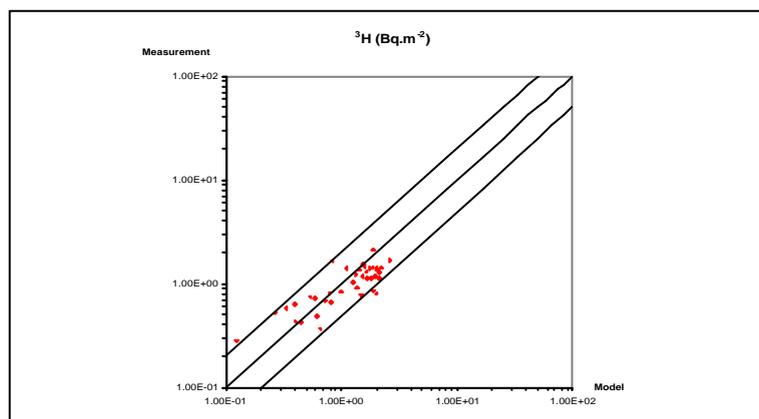


Fig. 3; Comparison between the results of the model and the <sup>3</sup>H measurements in the 1997 furze samples (the diagonal lines are equivalent to a factor of 2).

However, to estimate the tritium concentration in the samples, it is considered that there is equilibrium between the amount of <sup>3</sup>H present in the vegetation and in the air around this

vegetation. This model could be improved by considering the actual absorption kinetics of the water vapour in the vegetation (between night and day, for example).

Table 1. Model/Measurements ratio for various bioindicators and various years.

Year	Bioindicator	<sup>3</sup> H	<sup>129</sup> I	<sup>14</sup> C
1997	furze	1.3	4.1	1.8
1998	furze	1.6	4.1	2.8
1999	furze	1.6	4.0	2.7
2000	grass	not measured	1.5	not measured

### <sup>129</sup>I

For <sup>129</sup>I, the mean ratio between the theoretical values and the experimental values is 4.1 for 1997, 4.1 for 1998, 4.0 for 1999 and 1.5 for 2000 (Figure 4 and Table 1). The model over-estimates the activity found in the samples. Note that this over-estimation is greater for the furze than for the grass.

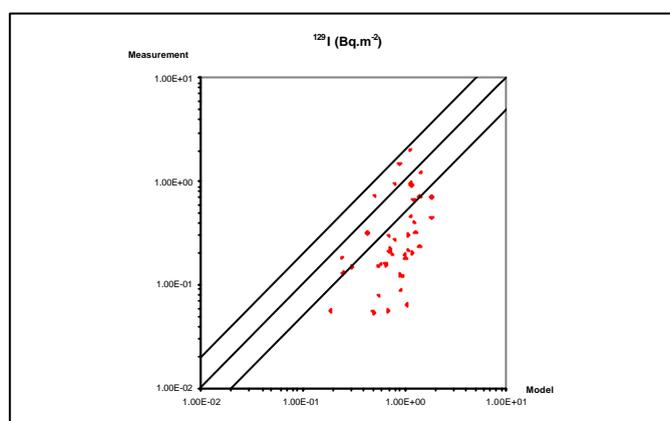


Fig. 4; Comparison between the results of the model and the <sup>129</sup>I measurements in the 1998 furze samples (the diagonals lines are equivalent to a factor of 2).

The differences between the model and the existing <sup>129</sup>I measurements can be explained by too high values for the dry and wet deposit parameters. For example, the dry deposition velocity stipulated by the GRNC (1999) is  $5 \cdot 10^{-3} \text{ m.s}^{-1}$  and a best estimate value was  $1.5 \cdot 10^{-3} \text{ m.s}^{-1}$ .

### <sup>14</sup>C

Regardless of the year, the model over-estimates the <sup>14</sup>C activity measured in the bioindicators (Figure 5 and Table 1).

The mean ratio between the theoretical values and the experimental values is 1.8 for 1997, 2.8 for 1998 and 2.7 for 1999. The differences existing between the model and measurements for the three years could come from the hypothesis of <sup>14</sup>C equilibrium between the atmosphere and the flora. As for <sup>3</sup>H, the <sup>14</sup>C model could be improved by taking into account the actual kinetics of carbon uptake in the plants (between night and day, for example).

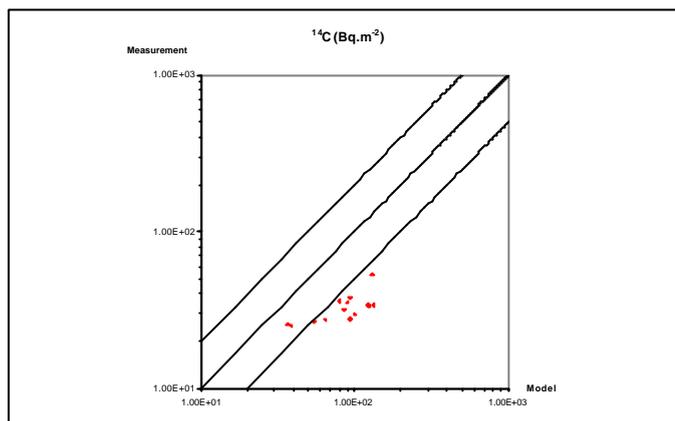


Fig. 5; Comparison between the results of the model and the  $^{14}\text{C}$  measurements in the 1999 furze samples (the diagonal lines are equivalent to a factor of 2).

## CONCLUSION

Following the analysis of  $^{85}\text{Kr}$  measurement campaigns carried out between 1997 and 2001 and grass and furze samples campaigns between 1997 and 2000, several parameters concerning atmospheric dispersion and deposit phenomena around the La Hague site could be assessed.

The  $^{85}\text{Kr}$  campaigns were used to fit a Gaussian model, to assess the atmospheric concentrations of released radionuclides. An atmospheric transfer model was then introduced, with its results being compared with data from the grass and furze campaigns. It emerges that the calculation chain employed produces results fairly close to the values measured for  $^3\text{H}$ , the  $^{14}\text{C}$  and the  $^{129}\text{I}$  (deviations lower than a factor of 5). The ratios for each of these radionuclides between the results of the model and the results of the measurements are of the same order of magnitude regardless of the year and bioindicator studied.

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