### KRYPTON-85: A TOOL FOR INVESTIGATING NEAR FIELD ATMOSPHERIC DISPERSION FOR ELEVATED EMISSIONS AROUND LA HAGUE SPENT FUEL NUCLEAR REPROCESSING PLANT

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### **INTRODUCTION**

Gaussian atmospheric dispersion models of a plume can be used to estimate the dispersion of ground-level atmospheric emissions or, in the case of high elevation emissions, distances far enough away from the source point for the plume to be close to the ground. These models have been essentially validated with ground-level or low height emission measurements campaigns (typically 10 m). The IRSN is conducting *in situ* near field (< 4 km) atmospheric dispersion investigations around COGEMA's La Hague spent fuel nuclear reprocessing plant (France), by using krypton-85 ( $^{85}$ Kr), the chemically inert gas, released in the gaseous waste from release stack 100 m high as an experimental atmospheric dispersion research device. This research aims to extend the scope of application of operational models.

Between 1997 and 1998, fourteen <sup>85</sup>Kr measurements campaigns were conducted at ground level and in the center of the plume for a variety of distances from the discharge point and meteorological conditions (Maro *et al.*, 2001). The various results confirm that <sup>85</sup>Kr is a good tool for the proposed investigation and that at short distances there are significant deviations between the models (Doury, 1976) (Pasquill - Briggs, 1974, 1976) and the experimental results, whence the need to acquire additional data on near field plume dispersion, particularly in relation to its vertical component. In this paper, the results of more recent measurements campaigns are presented and compared with the Gaussian atmospheric dispersion model using Doury (1976) and Pasquill - Briggs (1974, 1976) standard deviations, in order to reduce the uncertainties surrounding their areas of validity.

### EQUIPMENT AND METHOD

The IRSN is conducting fieldwork using the <sup>85</sup>Kr, released in La Hague plant gaseous waste to trace atmospheric dispersion. Bearing in mind that as a result of how COGEMA's La Hague plant operates, <sup>85</sup>Kr releases and kinetics are sequential, the Atmospheric Transfer Coefficients (ATC) for a given location during each shearing/dissolution of a fuel element in a bucket can be derived. By calculating the integrated <sup>85</sup>Kr concentration ratio to corresponding total emission quantity, over the whole period taken by the plume to reach the observation point, we arrive at the ATC:

$$ATC = \frac{\int_{t_0}^{t_1} X(M,t).dt}{\int_{t_0}^{t_1} q(t).dt}$$

where: - X(M,t): Radioactivity concentration at measuring point (M) at instant t (Bq.m<sup>-3</sup>), - q(t): Flow of the source activity (Bq.s<sup>-1</sup>),

- t'<sub>0</sub>, t'<sub>1</sub>: Instant of the beginning and end of source emission,

- t<sub>0</sub>, t<sub>1</sub>: Instant of the beginning and end of measurement.

Sets of ground-level readings are used to calculate the ATCs and determine horizontal distribution according to the distance from the source and meteorological conditions, essentially atmospheric turbulence. These campaigns are followed up by sets of altitude readings, under a purpose-designed tethered balloon (maximum flight altitude of 500 m), to estimate the vertical shape of the plume and the ATCs at various altitudes. The ground and above ground level measurements campaigns were not conducted at the same time. The campaigns took place during the daytime (in the time slot from one hour after sunrise to one hour before sunset), namely for atmospheric stability situations forecast to range from neutral to unstable conditions.

# GROUND-LEVEL AND ALTITUDE <sup>85</sup>KR MEASUREMENTS CAMPAIGNS

# Ground-level <sup>85</sup>Kr measurements campaigns

Following the 14 measurements campaigns conducted at ground level and in the center of the plume between 1997 and 1998, seven measurements campaigns were conducted with specific equipment between 21/05/01 and 27/06/01 for distances ranging from 300 - 3000 m from the discharge point (Table 1) to determine the ATCs and shape of the plume at ground level on either side of the wind axis.

Name	Date	Distance from discharge point (m)	Wind speed at 100 m (m.s <sup>-1</sup> )	Wind direction at 100 m (°)	Atmospheric stability according to Doury	Atmospheric stability according to Pasquill
DIAPEG1.1	21/05/01	1000	12.2	64.8	Normal diffusion	Class D
DIAPEG1.2	21/05/01	1000	11.8	66.8	Normal diffusion	Class D
DIAPEG2.1	22/05/01	300	11.7	59.2	Normal diffusion	Class D
DIAPEG3.1	20/06/01	1300	5.1	253.3	Normal diffusion	Class C
DIAPEG4.1	26/06/01	600	9.0	177.0	Normal diffusion	Class D
DIAPEG4.2	26/06/01	2200	6.8	255.5	Normal diffusion	Class D
DIAPEG5.1	27/06/01	3000	7.1	260.7	Normal diffusion	Class C

*Table 1. Ground-level measurements campaigns* 

The horizontal wind speeds, measured at a height of 100 m from the La Hague plateau are spread between 5.1 and 12.2 m.s<sup>-1</sup>. The meteorological diffusion conditions throughout the sampling are of the "normal diffusion" type according to Doury's classification and of "neutral or slightly unstable" type according to Pasquill (classes C and D).

## High-level <sup>85</sup>Kr measurements campaigns

Twelve flights were carried out since the end of 1999, to perfect equipment and to measure <sup>85</sup>Kr in altitude. All in all six upper-level measurements campaigns took place between 15/06/00 and

23/08/01 for distances ranging from 300 - 1800 m (Table 2) from the discharge point. The horizontal wind speeds, measured at a height of 100 m from the La Hague plateau, are spread between 2.6 and 7.7 m.s<sup>-1</sup>. The meteorological diffusion conditions throughout the sampling are of the "normal diffusion" type according to Doury's classification or of "neutral or unstable" type according to Pasquill (classes B, C and D).

Name	Date	Distance from discharge point (m)	Wind speed at 100 m (m.s <sup>-1</sup> )	Wind direction at 100 m (°)	Atmospheric stability according to Doury	Atmospheric stability according to Pasquill
BIPV4	15/06/00	1800	4.1	274.2	Normal diffusion	Class C
BIPV6	01/02/01	600	6.3	149.2	Normal diffusion	Class D
BIPV8	16/02/01	300	7.7	10.8	Normal diffusion	Class D
BIPV10	18/06/01	300	2.6	42.8	Normal diffusion	Class B
BIPV11	03/07/01	600	3.4	159.0	Normal diffusion	Class C
BIPV12	23/08/01	600	5.2	191.7	Normal diffusion	Class C

 Table 2. High-level measurements campaigns

### **RESULTS AND DISCUSSION**

### Ground-level measurements campaigns

The shape of the plume at ground level is found to be Gaussian in all campaigns. ATC maxima at the plume center are very similar for the seven campaigns, varying between  $1.1 \ 10^{-6} \text{ s.m}^{-3}$  at 3000 m and  $4.3 \ 10^{-6} \text{ s.m}^{-3}$  at 1000 m (Table 3).

 Table 3. Comparison of ATC maxima established during the ground-level measurements campaigns and the ATCs calculated by Doury and Pasquill models

Name	Date	Distance from discharge point (m)	Maximum ATC measured (s.m <sup>-3</sup> )	Doury ATC (s.m <sup>-3</sup> )	Pasquill ATC (s.m <sup>-3</sup> )
DIAPEG2-1	22/05/01	300	3.1 10 <sup>-06</sup>	2.5 10 <sup>-49</sup>	1.5 10 <sup>-14</sup>
DIAPEG4-1	26/05/01	600	2.8 10 <sup>-06</sup>	3.7 10-14	1.9 10 <sup>-08</sup>
DIAPEG1-1	21/05/01	1000	4.3 10 <sup>-06</sup>	1.1 10 <sup>-11</sup>	2.8 10-07
DIAPEG1-2	21/05/01	1000	4.3 10 <sup>-06</sup>	2.4 10-11	2.9 10-07
DIAPEG3-1	20/06/01	1300	1.4 10 <sup>-06</sup>	2.1 10 <sup>-06</sup>	2.8 10 <sup>-06</sup>
DIAPEG4-2	26/06/01	2200	1.5 10 <sup>-06</sup>	2.0 10 <sup>-06</sup>	1.3 10 <sup>-06</sup>
DIAPEG5-1	27/06/01	3000	1.1 10 <sup>-06</sup>	2.1 10 <sup>-06</sup>	7.1 10 <sup>-07</sup>

The ATCs established from the <sup>85</sup>Kr measurements is considerably underestimated by the models, both Doury and Pasquill, to the short distances from the discharge point (<1300 m), in the prevailing meteorological conditions. The smaller the distance between the source point and the calculation point, the greater the deviation between the models and the measurements. This is explained by the fact that the Doury and Pasquill models have not been validated for near field elevated emissions and for the type of complex topography that characterizes La Hague spent fuel nuclear reprocessing plant. The standard deviations calculated for this study vary from 43 – 210 m (Figure 1) for distances between the discharge point and the measuring point placed on the wind axis varying from 300 - 3000 m. The measured standard deviations diverge strongly from the standard deviations given by Doury on the whole of the area investigated. This contrasts with Pasquill standard deviations that compare well with the measured standard deviations between 3000 m and 1000 m but deviate below 1000 m. In fact the plumes calculated by Doury are much narrower than those estimated by Pasquill, and those measured at La Hague.



Figure 1. Horizontal standard deviations in relation to the distance from the discharge point for ground-level measurements campaigns and Doury and Pasquill models.

### High-level measurements campaigns

Taking all the measurements campaigns (Table 4) and meteorological conditions together, the ATCs vary from 2.1  $10^{-7}$  (4 m from the ground) to 4.0  $10^{-5}$  s.m<sup>-3</sup> (upper level measurements). These results also demonstrate that in near field the Doury model predicts narrower plumes at height than those actually measured. Pasquill's standard vertical deviations are greater than Doury's, and, in strong instability situations (BIPV10) and at a distance of 1800 m from the discharge point (BIPV4) are representative of the vertical dispersion of the plume. The ATCs measured at height and the ATCs calculated with the Pasquill model (ATC ~ 1  $10^{-5}$  s.m<sup>-3</sup>) at the center of the plume tally fairly well. The above differences stem from the fact that the gaussian approach considers that the medium is homogenous during the resolution of a pollutant's transport-diffusion equation. However the atmosphere is not a homogenous medium, especially in the vertical direction near the ground. In order to extend the scope of application of the near field operational models to upper level emissions, atmospheric turbulence quantification (a phenomenon representing the non-homogeneity of the atmosphere) will have to be perfected especially in its vertical dimension. The turbulence is induced by local phenomena (topography,

internal thermal boundary layer due to the land-sea contrast, etc...) or synoptic scale phenomena (wind field, subsidence, etc...).

Table 4. Comparison of ATC maxima established during upper level measurements campaigns and the ATCs calculated by the Doury and Pasquill models.

BIPV8				BIPV10				BIPV6			
Altit de (m)	u Measured ATC (s.m <sup>-3</sup> )	Doury ATC (s.m <sup>-3</sup> )	Pasquill ATC (s.m <sup>-3</sup> )	Altitu de (m)	Measured ATC (s.m <sup>-3</sup> )	Doury ATC (s.m <sup>-3</sup> )	Pasquill ATC (s.m <sup>-3</sup> )	Altitu de (m)	Measured ATC (s.m <sup>-3</sup> )	Doury ATC (s.m <sup>-3</sup> )	Pasquill ATC (s.m <sup>-3</sup> )
4	2.1 10-07	1.4 10 <sup>-25</sup>	6.6 10 <sup>-14</sup>	4	2.7 10-06	2.9 10 <sup>-08</sup>	1.6 10-06	4	4.4 10-06	7.2 10-10	3.2 10 <sup>-08</sup>
51	2.5 10-05	6.2 10 <sup>-10</sup>	2.7 10-07	43	2.0 10-05	5.1 10 <sup>-06</sup>	1.0 10-05	51	3.9 10 <sup>-06</sup>	2.8 10-06	3.6 10 <sup>-06</sup>
73	2.8 10-05	4.2 10-06	1.1 10-05	57	4.0 10-05	1.8 10-05	1.8 10-05	135	3.5 10-06	1.2 10-05	8.5 10-06
90	9.4 10 <sup>-06</sup>	1.2 10-04	4.7 10-05	86	1.4 10-05	8.0 10-05	3.3 10-05				

Table 4 (continued). Comparison of ATC maxima established during upper level measurements campaigns and the ATCs calculated by the Doury and Pasquill models

BIPV11			BIPV12				BIPV4				
Altitu de (m)	Measured ATC (s.m <sup>-3</sup> )	Doury ATC (s.m <sup>-3</sup> )	Pasquill ATC (s.m <sup>-3</sup> )	Altitu de (m)	Measured ATC (s.m <sup>-3</sup> )	Doury ATC (s.m <sup>-3</sup> )	Pasquill ATC (s.m <sup>-3</sup> )	Altitu de (m)	Measured ATC (s.m <sup>-3</sup> )	Doury ATC (s.m <sup>-3</sup> )	Pasquill ATC (s.m <sup>-3</sup> )
4	2.5 10-06	8.3 10-07	2.9 10 <sup>-06</sup>	4	1.1 10 <sup>-06</sup>	1.4 10 <sup>-08</sup>	1.9 10 <sup>-06</sup>	4	5.6 10 <sup>-06</sup>	3.6 10-06	2.5 10-06
102	8.5 10-06	3.6 10-05	1.6 10 <sup>-05</sup>	101	2.5 10-05	4.8 10 <sup>-05</sup>	1.1 10 <sup>-05</sup>	40	5.1 10-06	4.4 10 <sup>-06</sup>	2.4 10 <sup>-06</sup>
				151	1.4 10 <sup>-06</sup>	4.6 10 <sup>-06</sup>	5.6 10 <sup>-06</sup>	71	4.9 10 <sup>-06</sup>	5.6 10 <sup>-06</sup>	2.3 10-06
								102	4.4 10-06	6.0 10 <sup>-06</sup>	2.2 10 <sup>-06</sup>

## CONCLUSION

Ground-level and upper level measurements campaigns enable us to gather data on the horizontal and vertical atmospheric dispersion of the <sup>85</sup>Kr, discharged by a stack 100 m high, around La Hague spent fuel nuclear reprocessing plant. The first findings of the measurements campaigns at ground level show that the plume has a gaussian shape at the ground with measured horizontal standard deviations varying from 43 - 210 m for distances, between the discharge point and the measuring point placed on the wind axis, ranging from 300 - 3000 m and for medium to strong turbulence situations. These standard deviations tally closely with Pasquill's but diverge strongly from Doury's standard deviations. The vertically measured plume shapes indicate that local phenomena (presence of site buildings, topography of La Hague region, the internal thermal boundary layer, etc...) or synoptic scale phenomena (wind field, subsidence, etc...) can have an influence on atmospheric dispersion. To improve near field atmospheric dispersion quantification for upper level emissions and increase the scope of validity of the operational models, further investigation will be carried out to analyze the influence these phenomena on atmospheric turbulence. It should adopt both an experimental approach (turbulence measurements linked to <sup>85</sup>Kr measurements campaigns) and a mathematical approach (calculations with grid models).

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