CONTRIBUTION TO THE STUDY OF ATMOSPHERIC DISPERSION OF A MESOSCALE POLLUTANT: USE OF KRYPTON-85 RELEASED BY THE COGEMA LA HAGUE NUCLEAR SPENT FUEL REPROCESSING PLANT AS ATMOSPHERIC TRACER

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INTRODUCTION

Operational atmospheric dispersion models (Gaussian models) are normally valid for releases on the ground, in the near-field where the topography is simple and up to a distance of a few tens of kilometres based on topography complexity. In fact few model validation campaigns have been conducted in the near-field for at-height releases and in complex topography or beyond a few tens of kilometres. IRSN/LRC has therefore conducted a study on the atmospheric dispersion of mesoscale pollutants, using krypton-85 (^{85}Kr) released by the COGEMA nuclear spent fuel reprocessing plant at La Hague (north-western France) as the plume tracer (Maro *et al.*, 2001, 2002). ^{85}Kr is emitted chronically by two 100 metre-high chimneys 200 metres apart. During a six months campaign, it was measured in real time in the air using proportional counters or plastic scintillators at two stations, one at the Laboratoire de Radioécologie de Cherbourg-Octeville (LRC) (18 km south-east of the release point) and the other at the La Hougue signal station (45.4 km south-east of the release point).

This paper aims to present the ^{85}Kr measurement results obtained from both stations and how they have been interpreted by the atmospheric dispersion operational models developed by Pasquill (1974) and Doury (1976).

EQUIPMENT AND METHOD

For this study of mesoscale atmospheric dispersion, two systems measuring ^{85}Kr concentrations in the air have been installed, at LRC in June 2003 and at the La Hougue signal station in November 2003 (Figure 1).

LRC is 18 km from the release point at COGEMA La Hague based on an angle of 106° from geographic north. The equipment is a plastic scintillator (SEM 1000-Munchener Apparatebau). The La Hougue signal station is 45.4 km from the release point at COGEMA La Hague based on an angle of 104° from geographic north. The equipment is a proportional counter (LB111-Berthold).

The measurements taken when the ^{85}Kr is carried towards LRC and La Hougue signal station are used to determine the Atmospheric Transfer Coefficients (ATC) based on the micro-metrological conditions (wind speed, atmospheric stability).

The micro-meteorological data used in this study is provided by the weather station at the COGEMA La Hague plant.

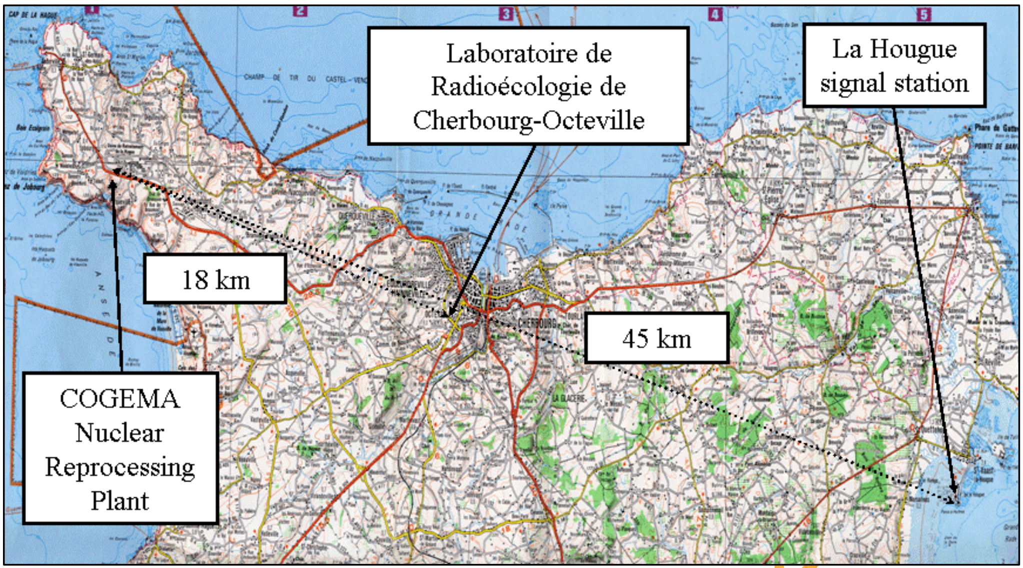

Figure 1: Positions of measurement stations (LRC, La Hougue) and the release point of krypton-85 from COGEMA La Hague.

The ATC is the ratio between the ^{85}Kr concentration integrated into the transit time of the plume at the observation point and the corresponding total amount emitted:

$$ATC = \frac{\int_{t0}^{t1} X(M,t).dt}{\int_{t'0}^{t'1} q(t).dt}$$

where:
- X(M,t): activity concentration at measurement point (M) at time t ($Bq.m^{-3}$),
- q(t): emission rate from the source ($Bq.s^{-1}$),
- t'0, t'1: emission start and end time from the source (s),
- t0, t1: measurement start and end time (s).

The ATC measured at LRC and the La Hougue signal station are compared with those obtained using the Gaussian models developed by Pasquill (1974) and Doury (1976). Nevertheless, the calculated ATC depend strongly on the trajectory followed by the plume between the emission point and the measurement points. The measurements and the Gaussian models (Pasquill, Doury) are compared for a direct trajectory from the COGEMA La Hague plant up to the measurement points.

RESULTS AND DISCUSSION

^{85}Kr measurements at LRC and the La Hougue signal station

An analysis of the results reveals that during the study period, the ^{85}Kr plume was detected 75 times at LRC and 14 times at the La Hougue signal station.

At LRC, the average ^{85}Kr concentrations noted for all episodes are between 320 $Bq.m^{-3}$ and 6500 $Bq.m^{-3}$. Wind speeds at 10 metres at the release point varied between 1 $m.s^{-1}$ and 9 $m.s^{-1}$.

At the La Hougue signal station, average ^{85}Kr concentrations noted for all episodes are between 300 Bq.m^{-3} and 1200 Bq.m^{-3}. Wind speeds at 10 metres at the release point varied between 3 m.s^{-1} and 9 m.s^{-1}.

Determination of the atmospheric stability and ATC calculation at LRC and the La Hougue signal station

Atmospheric stability is determined with the method recommended by the EPA (2000) and using the horizontal wind speed and standard deviation of the vertical component in wind direction. During the study period, 75 ATC were measured at LRC and 14 at the La Hougue signal station. 7 ATC were obtained in meteorological conditions corresponding to Pasquill's class B, 13 in class C, 21 in class D and 48 in class E, i.e. 41 measurements in normal diffusion class (Doury) and 48 in low diffusion class. No measurements were observed in class A or class F (Pasquill). The ATC obtained at the La Hougue signal station were observed only for stability classes D or E (Pasquill).

At LRC, the average ATC measured for classes B, C, D and E are respectively 6.1 10^{-8} s.m^{-3} (average concentration of 1121 Bq.m^{-3}), 1.2 10^{-7} s.m^{-3} (average concentration of 1635 Bq.m^{-3}), 1.2 10^{-7} s.m^{-3} (average concentration of 1573 Bq.m^{-3}) and 2.4 10^{-7} s.m^{-3} (average concentration of 2703 Bq.m^{-3}). At the La Hougue signal station, the average ATC measured for classes D and E are equal to 3.5 10^{-8} s.m^{-3} (average concentration of 507 Bq.m^{-3} in class D and 594 Bq.m^{-3} in class E) (Tables 1 and 2).

Table 1: Average concentrations and ATC obtained at LRC as a function of atmospheric stability.

Stability class	Average concentration (Bq.m^{-3})	Average ATC (s.m^{-3})
B	1121	6.1 10^{-8}
C	1635	1.2 10^{-7}
D	1573	1.2 10^{-7}
E	2703	2.4 10^{-7}

Table 2: Average concentrations and ATC obtained at the La Hougue signal station as a function of atmospheric stability.

Stability class	Average concentration (Bq.m^{-3})	Average ATC (s.m^{-3})
D	507	3.5 10^{-8}
E	594	3.5 10^{-8}

Comparison of results obtained at LRC and the La Hougue signal station with Gaussian atmospheric dispersion models

Figures 2 and 3 compare the measured and calculated ATC for the direct trajectories from the COGEMA La Hague plant to the measurement points.

For the direct trajectories, the calculated ATC must be higher than or equal to the measured ATC because the actual trajectory has not been taken into account. If the calculated ATC are lower than the measured ATC, the model underestimates reality. If the calculated ATC are higher than the measured ATC, two possibilities exist: either the model overestimates reality or the plume trajectory is not direct.

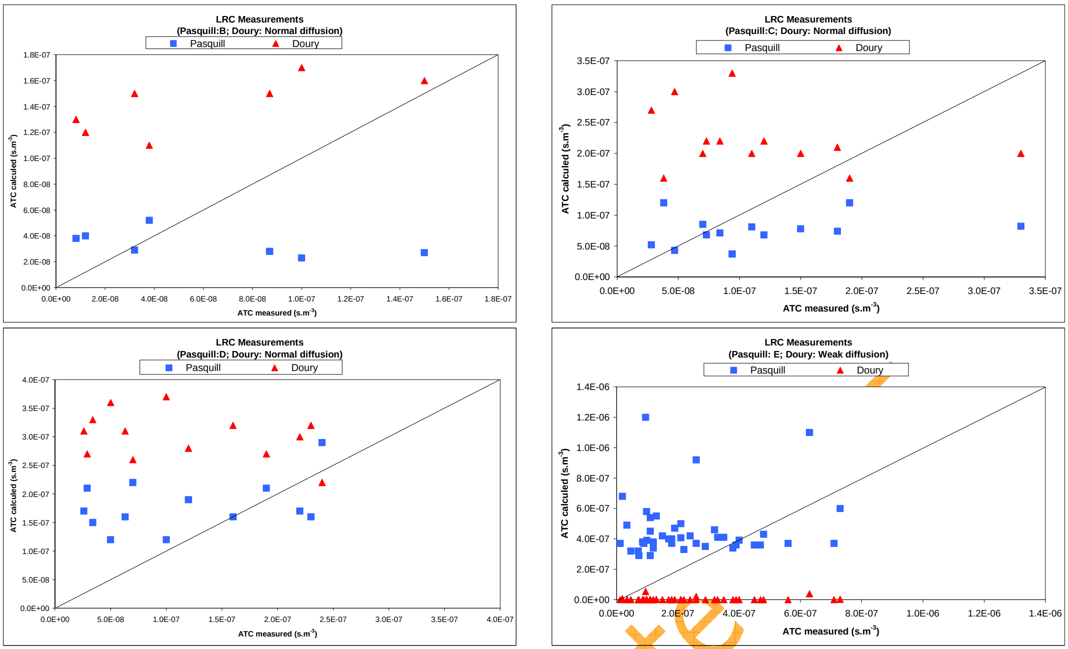

Figure 2: Comparison between ATC measured at LRC and calculated ATC for the various models as a function of atmospheric stability.

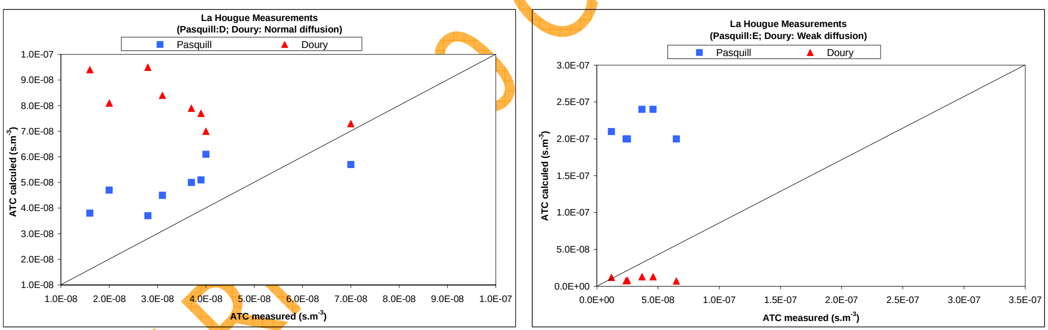

Figure 3: Comparison between ATC measured at the La Hougue signal station and calculated ATC for the various models as a function of atmospheric stability.

At LRC and on average, for stability class B (7 ATC), the ATC calculated using the Pasquill model underestimate the measured ATC by a factor of 2 and the ATC calculated using the Doury method overestimate the measured ATC by a factor of 3. For stability class C (13 ATC), the ATC calculated using the Pasquill method underestimate the measured ATC by a factor of 2 whereas the ATC calculated using the Doury method overestimate them by a factor of 2. For stability class D (13 ATC), the ATC calculated using the Pasquill method overestimate the measured ATC by a factor of 2 whereas the ATC calculated using the Doury method overestimate them by a factor of 3. For stability class E (42 ATC), the ATC calculated using the Pasquill method overestimate the measured ATC by a factor of 2 whereas the ATC calculated using the Doury method underestimate them by more than a factor of 1000.

At the La Hougue signal station and on average, for stability class D (8 ATC), the ATC calculated using the Pasquill method overestimate the measured ATC by a factor of 2 and the ATC calculated using the Doury method overestimate them by a factor of 3. For stability class E (6 ATC), the ATC calculated using the Pasquill method overestimate the measured ATC by a factor of 5 whereas the ATC calculated using the Doury method underestimate them by a factor of 4.

CONCLUSION

An analysis of the results reveals that during the study period (six months), the ^{85}Kr plume was detected 75 times at LRC and 14 times at the La Hougue signal station for different conditions of atmospheric stability. The results obtained have been compared with the results from the Pasquill and Doury operational atmospheric dispersion models. For a distance of 18 km from the release (LRC measurement point), the Pasquill model provides good estimations of the dispersion of a plume whatever the stability conditions. The Doury model, however, only provides a good estimation of the dispersion of a plume in unstable and neutral conditions. For a distance of 45.4 km from the release (La Hougue signal station), the Pasquill and Doury models provide good estimations of the dispersion of a plume for unstable and neutral conditions. Divergences appear seemingly in conditions of high stability.

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