COMPARISON of LAGRANGIAN ATMOSPHERIC DISPERSION MODELS (DIFPAR, SPRAY) WITH ^{85}KRYPTON MEASUREMENTS TAKEN AROUND LA HAgue SPENT FUEL REPROCESSING PLANT

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INTRODUCTION

In June 1997 the Institute for Radiological Protection and Nuclear Safety (IRSN) in collaboration with Météo-France began a joint *in situ* investigation around COGEMA's La Hague spent fuel reprocessing plant (France) with the aim of reducing the uncertainties surrounding near field (< 4 km) operational atmospheric dispersion models (Pasquill, 1974; Doury, 1976) for elevated emissions. The program was to look into krypton-85 (^{85}Kr), the chemically inert gas, released in the gaseous waste (release stack 100 m high) as a plume tracer (Maro, 2001).

Krypton-85 is a radioactive β^- and γ-emitting isotope with a radioactive half-life of 10.71 years, which is both naturally occurring and produced in nuclear reactors. Furthermore it has been released into the atmosphere following atomic bomb explosions, but most emissions are currently discharged by spent fuel reprocessing plants such as COGEMA's La Hague plant.

The goal of this investigation is to compare the Atmospheric Transfer Coefficients (ATC) obtained from ^{85}Kr measurements in La Hague with the findings of two Lagrangian atmospheric dispersion models: DIFPAR (developed by Electricité De France) and SPRAY (an ARIA Technologies product). The meteorological data is supplied by the meso-NH model, a non-hydrostatic meteorological model developed jointly by Météo-France and the Aerology Laboratory of the French National Scientific Research Center (CNRS). In this paper the computations of the two models are presented and compared with the ATCs measured in the environment over the course of two measurements campaigns held on 23 April 1998 and 15 June 2000.

EQUIPMENT AND METHOD

Atmospheric measurements of ^{85}Kr

The IRSN is conducting fieldwork using the ^{85}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, ^{85}Kr releases and kinetics are sequential, the ATCs for a given location during each shearing/dissolution of a fuel element in a bucket can be derived. By calculating the integrated ^{85}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.

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 tethered balloon (maximum flight altitude of 500 m), to estimate the vertical shape of the plume and the ATCs at various altitudes. The ground-level and altitude measurements campaigns were conducted at separate times. The 23 April 1998 and 15 June 2000 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.

Lagrangian DIFPAR and SPRAY atmospheric dispersion models
The models used are special codes of the DIFPAR (Electricité de France) and SPRAY (Aria Technologies) types.

DIFPAR (Wendum, 1998) describes atmospheric release by following a large number of particles whose various components are displaced according to the following form:

$$\delta x = U\delta t + B\sqrt{\delta t}\,\Omega_x$$

where U is the function of the wind and gradient diffusivity, B the diffusivity function, and Ω^x white noise. The meso-NH model provides the meteorological values directly, the vertical diffusivity coefficients are calculated from meso-NH by applying the Louis (1979) formulation.

SPRAY 2.1 (Tinarelli *et al.* 1994a, Tinarelli *et al.* 1994b) is a stochastic or Monte-Carlo (Hockney and Eastwood, 1981) type dispersion model, based on the hypothesis that turbulent atmospheric flux is characterized by random spatial and temporal variations of fluid dynamic variables. The version used in this study uses a meteorological preprocessor to recalculate turbulent fluxes from meteorological fields supplied by meso-NH.

RESULTS AND DISCUSSION

Campaign carried out on 23 April 1998
On this particular day, ^{85}Kr was measured at ground level at various distances from the discharge point. The readings taken over the course of the day are given in Figure 1. The maximum ^{85}Kr air concentration reached was 6.2 10^4 Bq.m^{-3} and the ATCs (Table 1) were in the range 8.1 10^{-7} - 1.3 10^{-6} s.m^{-3}. The ATCs varied only very slightly (a factor of 1.5 in the readings taken at 8:40 a.m. and 9:20 a.m.) over the same distance from the discharge point (1000 m).

Table 1. Meteorological readings at the discharge point and ATCs measured at ground level during the measurementscampaign of 28 April 1998.

Date/time	Distance from discharge point (m)	Wind speed (m.s^{-1}) at an altitude of 100 m	Wind direction (°) at an altitude of 100 m	ATCs (s.m^{-3})
23/04/98: 8:40 a.m.	1000	15.3	181	1.2 10^{-6}
23/04/98: 9:20 a.m.	1000	16.5	176	8.1 10^{-7}
23/04/98: 12:50 p.m.	2275	15.1	211	1.3 10^{-6}
23/04/98: 2:40 p.m.	575	16.8	232	1.2 10^{-6}

The day's weather was characterized by the arrival of a westerly disturbance that was to generate instability, giving rise to cold air descending to the low layers with resulting precipitation. Table 2 presents the calculations obtained by the models. The meso-NH model accurately plots the flow for that day, except at 12:50 p.m. where there is a 20° error in the wind direction. This error is linked to a delay in the disturbance going through the meso-NH simulation, which by displacing the cloud in relation to the point of measurement affected the ATC values plotted by SPRAY and DIFPAR. The ATCs obtained using DIFPAR match the measurements (<factor of

4), whereas SPRAY underestimated the ATCs read off at ground level by a wide margin - a factor of 2 to 50.

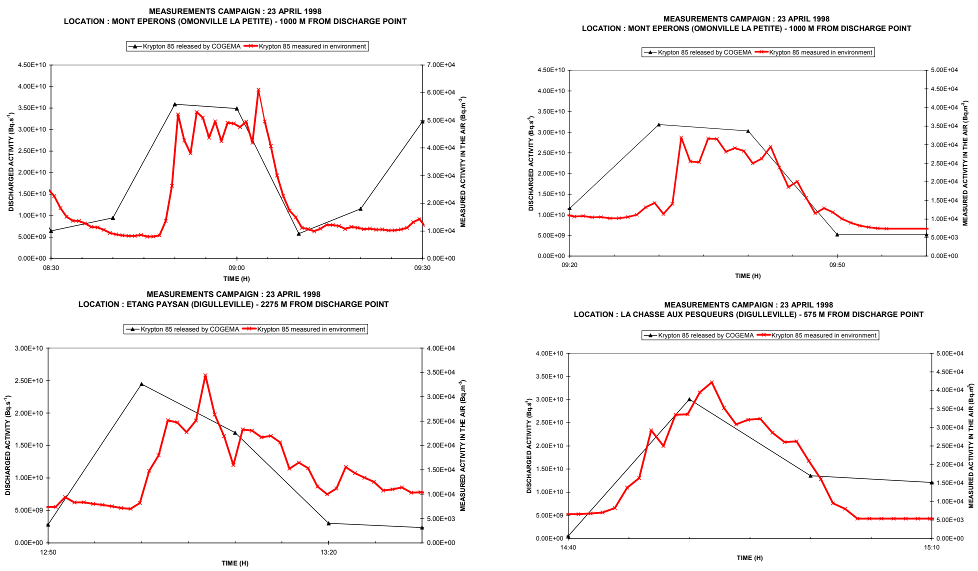

Figure 1. ^{85}Kr measurements at ground-level taken during the campaign of 23 April 1998.

Table 2. Wind modeled at the discharge point and ATCs simulated at ground-level for the measurements campaign of 28 April 1998.

Time	**8:40 a.m.**	**9:20 a.m.**	**12:50 p.m.**	**2:40 p.m.**
Meso-NH Wind Direction (°)	186	188	195	227
Meso-NH Wind Force (m.s^{-1})	14.3	15.0	17.8	14.7
DIFPAR ATC (s.m^{-3})	1.5 10^{-6}	1.5 10^{-6}	3.3 10^{-7}	1.6 10^{-6}
SPRAY ATC (s.m^{-3})	6.5 10^{-8}	3.7 10^{-7}	3.0 10^{-7}	2.5 10^{-8}

Campaign carried out on 15 June 2000

The activity levels measured at ground and above ground-level and at altitude were 2.0 10^5 and 1.2 10^5 Bq.m^{-3} respectively. During these sessions, the mean wind speed and direction were 4.1 m.s^{-1} and 274.2 ° (readings taken at an altitude of 100 m).

The ATCs measured up to an altitude of 100 m were in the range 5.6 10^{-6} s.m^{-3} at ground level and 4.5 10^{-6} s.m^{-3} at 100 m (figure 2). The plume shape, with maximum concentration at ground level, reveals the reflection of the plume on the ground.

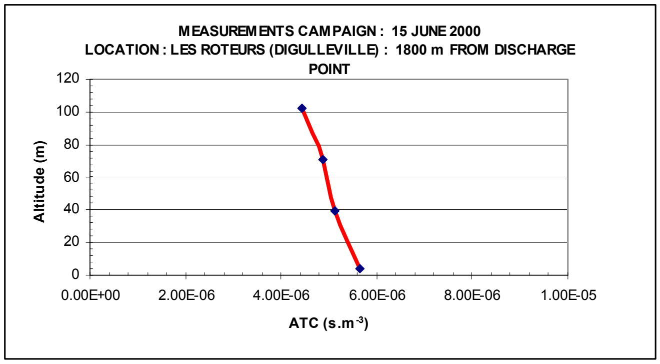

Figure 2. ATC ground-level and altitude measurements taken with the use of a tethered balloon during the campaign of 15 June 2000.

The day's weather was characterized by the arrival of a weak westerly flow with a mixed layer roughly 400 m thick over the area of investigation. The meso-NH values reproduced give a westerly wind (slightly south-westerly) of about 4.5 m.s^{-1} at the discharge.

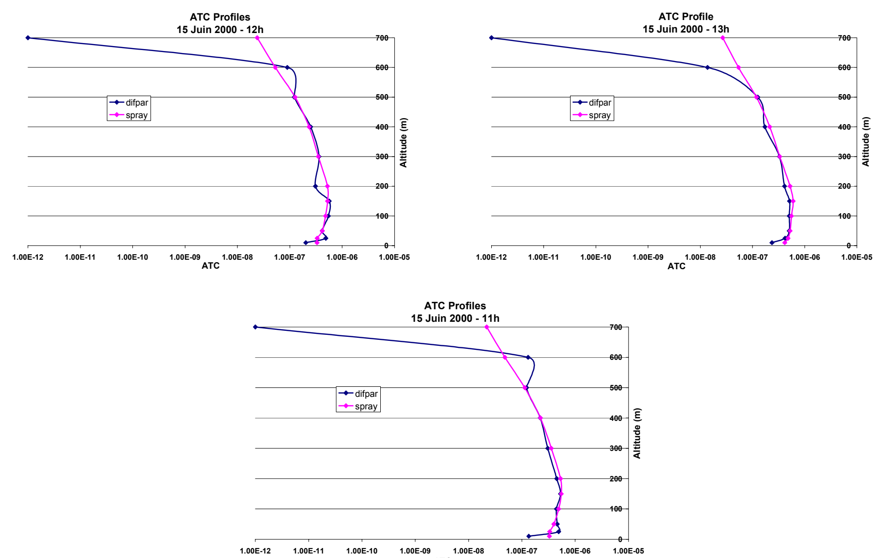

Figure 3. Vertical ATC profiles simulated by DIFPAR and SPRAY.

The ATCs obtained (figure 3) with the two codes are similar and underestimate the ground-level values by a factor of 10. We actually observe slight slippage (an angular error of 5°) between the simulated and observed ATC maxima. At ground level the simulated ATC maxima compare quite well with the observed values (<factor of 5). However, both DIFPAR and SPRAY posit a maximum ATC clustering at an elevation of about 50 meters and not at ground level as the measurements indicate.

CONCLUSION

The measurements campaigns carried out at ground-level on 23 April 1998 and above ground-level on 15 June 2000 yield data about ATCs according to distance from the point of emission and for above ground-level for the meteorological conditions prevailing at the time of the sessions. The indications are that DIFPAR stands comparison better with the ATCs measured at ground-level on 23 April 1998. While the behavior of SPRAY is fairly similar, it underestimates the ATC values by a factor of 2 - 50. The session held on 15 June 2000 is more interesting because it offers ground measurements and a vertical profile of experimental ATCs up to 100 m. The two models produce similar results but underestimate the values actually measured by a factor of 10. These results are encouraging, but if we want to characterize the differences and similarity of the two particular codes' behavior, we will need to simulate more of the ground- and above ground-level monitoring sessions already carried out by IRSN. Research around COGEMA's La Hague spent fuel reprocessing plant should enable us to assess atmospheric dispersion models, so that operational models can ultimately be improved.

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