1.19 VALIDATION OF DRY DEPOSITION MODELS FOR SUBMICRONIC AND MICRONIC AEROSOLS

Denis Maro[1], Denis Boulaud[2], Alexis Coppalle[3], Pierre Germain[1], Didier Hebert[1] and Lionel Tenailleau[4]

[1]Institut de Radioprotection et de Sûreté Nucléaire, DEI/SECRE/LRC, BP 10, F-50130 Cherbourg-Octeville (France)

[2]Institut de Radioprotection et de Sûreté Nucléaire, DESTQ/Dir, BP17, F-92262 Fontenay aux Roses (France)

[3] UMR 6614 CORIA, Campus du Madrillet, F-76801 Saint Etienne du Rouvray (France)

[4] Marine Nationale, EAMEA/GEA, BP 19, F-50115 Cherbourg-Armées (France)

INTRODUCTION

The atmosphere is a major transfer path of pollutants released in gaseous form or in aerosols form from an industry to the land and/or sea environment and, consequently, to humans. In order to estimate the impacts of an atmospheric release on human and on the environment, it is necessary to assess the dispersion and deposition (wet and dry) of these pollutants. Dry deposition was studied from experimental campaigns carried out *in situ* and in laboratory which allowed understanding globally the phenomena governing it. However, it still remains uncertainties as for the assessment of dry deposition velocity, in particular for submicronic and micronic aerosols. The dry deposition velocity depends on numerous factors such as micro-meteorological conditions, pollutant and substrate properties. This implies that dry deposition velocity cannot be accurately assessed without *in situ* measurements. Generally, a conservative value of $5\ 10^{-3}$ $m.s^{-1}$ is used in operational models due to a lack of specific knowledge of the site being studied.

On May 18th, 2001, the spent fuel reprocessing plant of COGEMA La Hague released into the atmosphere, Ruthenium and Rhodium-106 (^{106}Ru-Rh) aerosols which marked the near environment of the site. Following this release, grass was sampled by different laboratories (*GRNC*, 2002) (*Crabol and Maro*, 2001) and the results obtained were analyzed (*Maro et al.*, 2002).

The purpose of this document is to show the results of ^{106}Ru-Rh measurements carried out on grass, and the interpretation made regarding the operational deposition models.

EQUIPMENT AND METHOD

On May 18th, 2001, a failure in a gas-treatment line of the spent fuel reprocessing plant of COGEMA La Hague resulted in a ^{106}Ru and ^{106}Rh release into the atmosphere. The activity released during 1 hour was estimated to $4.5\ 10^{9}$ Bq (*COGEMA*, 2001). ^{106}Ru and ^{106}Rh are beta- and gamma-emitting radioelements. ^{106}Ru and ^{106}Rh are in radioactive equilibrium with respectively radioactive time periods of 372.6 days and 30 s. Following this release, grass was sampled by COGEMA, ACRO, OPRI and IRSN (*GRNC*, 2002) (*Crabol and Maro*, 2001) (*Maro et al.*, 2002) under wind of the facility in the wind direction 320°. Grass was sampled on both sides of the average wind direction between 700 m and 6,700 m from the discharge point (figure 1).

Grass samples were then dried to 60°C then ^{106}Ru-Rh concentrations were measured using gamma spectrometry in the Roule Mountain underground laboratory at Cherbourg (French Navy) with a very low background noise deeply improving measures reliability.

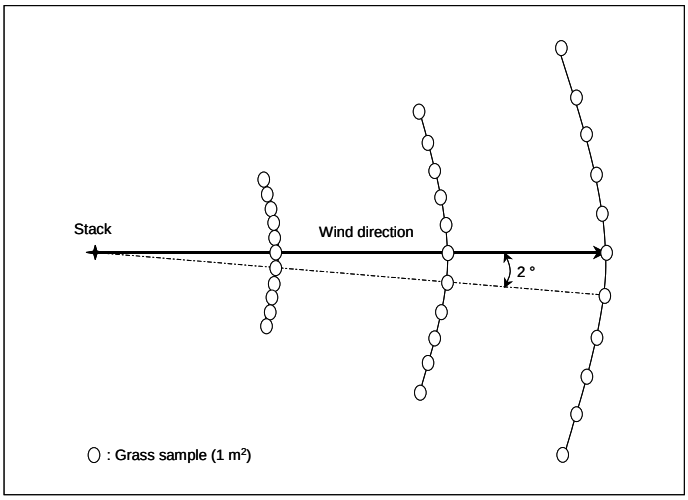

Figure 1. Position of sample points regarding the discharge point and the average wind direction

RESULTS AND DISCUSSION

Results of ^{106}Ru-Rh measurements in grass samples

Measurement results are displayed in figure 2 depending on the distance from the discharge point (UP2-800 stack).

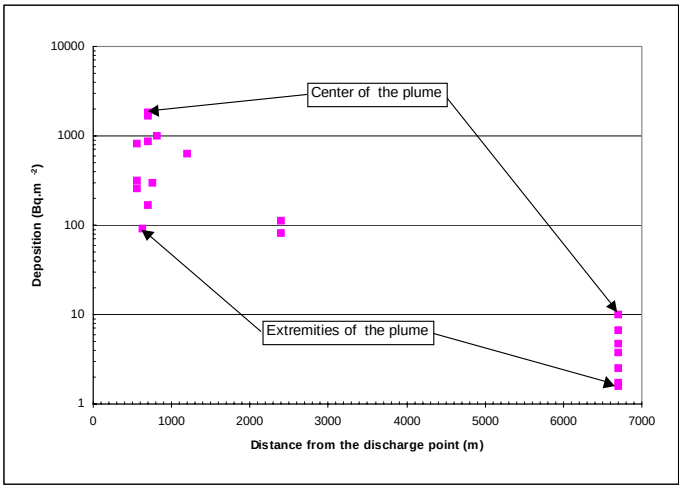

Figure 2. Evolution of ^{106}Ru-Rh deposition in environment depending on the distance from the discharge point

These results show a marking of the grass, up to a few thousands of Bq.m^{-2} of ^{106}Ru-Rh, at 700 m from the discharge stack, in the wind direction. This released activity reduces by 10 (100 Bq.m^{-2}) at 2,400 m and by 100 at least (10 Bq.m^{-2}) at 6,700 m.

Comparison of measurement results with calculation results using a constant dry deposition velocity of 5 10^{-3} m.s^{-1} generally used for operational models

An atmospheric plume may progressively deplete in the wind direction, as aerosols stick to surfaces, such as leaves and soil. The released aerosol quantity may be determined from a term called dry deposition velocity. This parameter of which dimension is a velocity (m.s^{-1}), is the quotient of the dry deposition flux density on the soil (Bq.m^{-2}.s^{-1}) and the atmospheric concentration at the soil level (Bq.m^{-3}). The dry deposition velocity values vary with the

atmospheric stability, the wind velocity and the surface condition, but also the aerosol granulometry. The most used hypothesis for operational atmospheric dispersion models, is a constant dry deposition velocity for a given physico-chemical form. With no precise data on the physical characteristics of the released aerosols, the typical value of this deposition velocity is 5 10^{-3} $m.s^{-1}$.

Basing on the Atmospheric Transfert Coefficients (ATC) measured at 700 m from the IRSN atmospheric dispersion study campaigns (experimental ATC) (*Maro et al.*, 1999) and the ^{106}Ru-Rh quantity released, assessed from ^{106}Ru-Rh measurements of COGEMA La Hague, the dry deposition velocity of aerosols may be estimated to 5.7 10^{-2} $m.s^{-1}$ (Figure 3).

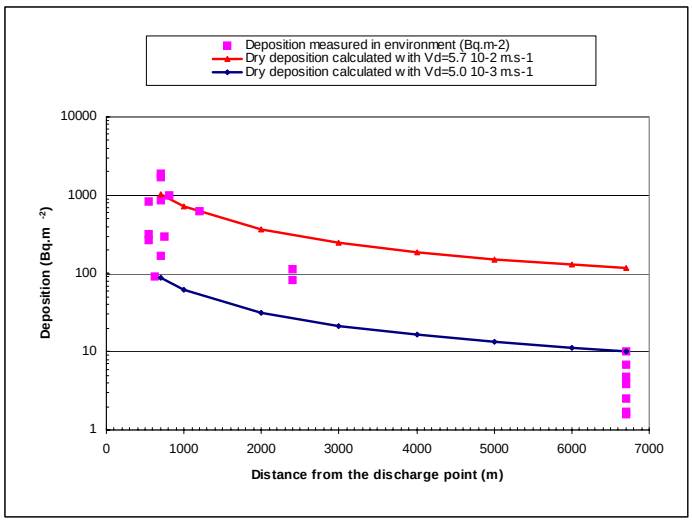

Figure 3. Comparison between ^{106}Ru-Rh deposition in environment with deposition calculated with dry deposition velocities of 5 10^{-3} $m.s^{-1}$ and 5.7 10^{-2} $m.s^{-1}$

This deposition velocity is higher (factor of 10) than the deposition velocity of 5 10^{-3} $m.s^{-1}$, generally used for operational models. An hypothesis to explain this significant value of the deposition velocity would be the non incorporation of the electrostatic field in the deposition velocity assessment for very low granulometry aerosols (*Gensdarmes*, 2002). Another hypothesis is that the significant value of deposition velocity may also result from the presence of strong local turbulence which may increase the deposition velocity of fine aerosols. Besides, during this incident, it is likely that ruthenium was released in gaseous form, RuO_4, then was progressively reduced in the form of RuO_2 aerosols. One may think that this reduction of RuO_4 into RuO_2 in the facility resulted from homogeneous nucleation which produced ultrafine aerosols (a few nanometers).

However, for this deposition velocity of 5.7 10^{-2} $m.s^{-1}$ determined from measurements at 700 m from the discharge point, the activity settled in ^{106}Ru-Rh should be 120 $Bq.m^{-2}$ at 6,700 m, whereas the activity measured in environment at this distance is about 5 $Bq.m^{-2}$ (Figure 3). We tried to determine if this deviation between measurements and model might be explained by taking into account the coagulation phenomenon of ^{106}Ru-Rh aerosols on atmospheric aerosols in the atmospheric dispersion modelling.

Comparison of measurement results with calculation results taking into account the coagulation phenomenon of ^{106}Ru-Rh aerosols on atmospheric aerosols

In order to take into account the aerosol coagulation phenomenon (*Boulaud and Renoux*, 1998) in the modelling, we considered a population of atmospheric aerosol with R_1 radius comprised between 10^{-2} and 1 µm and a population of ultra-fine ^{106}Ru-Rh aerosols with

R_2 radius comprised between 1 and 10 nanometers. The concentration of atmospheric aerosols N_1 is considered as equal to 5,000 particles per cm^3 (measuring campaign in La Hague dated March 11th, 2002). The coagulation phenomenon of R_1 radius particles with R_2 radius particles is represented by a coagulation coefficient noted K_{12}. K_{12} is therefore the total number of collisions, within one cm^3 and per s, between R_1 radius particles and R_2 radius particles, for a concentration of 1 particle per cm^3 of each type. To follow the evolution of the number of N_2 particles with R_2 radius regarding the population of the number of N_1 particles with R_1 radius, resolve the following differential equation (1):

$$\frac{dN_2}{dt} = -\frac{1}{2}.K_{12}.N_1.N_2 \qquad (1)$$

By integration, we obtain equation 2:

$$N_2 = N_{02}.Exp(-\frac{1}{2}.K_{12}.N_1.t) \qquad (2)$$

with N_{02}: initial concentration of R_2 radius particles.

The N_2 population would have reduced by half after a $T_{1/2} = -\frac{2.LN(0.5)}{K_{12}.N_1}$ (equation 3) time period.

The initial deposition velocities for atmospheric particles and ^{106}Ru-Rh particles are respectively 1 10^{-4} and 5,7 10^{-2} $m.s^{-1}$. Then, from measurements performed in environment, we determined, by mathematic adjustment (figure 4), to which coagulation coefficient (K_{12}) would correspond this deposition evolution depending on the distance regarding the discharge point.

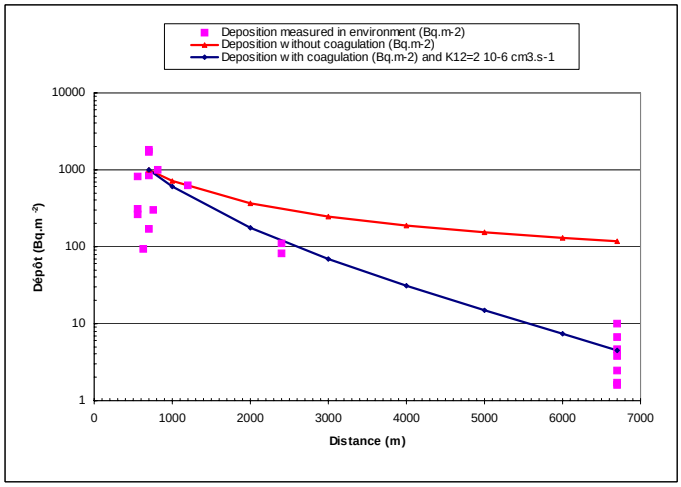

Figure 4. Comparison between ^{106}Ru-Rh deposition in environment and deposition calculated without and with coagulation phenomenon aerosols

The best adjustment between measurements and modelling is determined for a K_{12} value of 2 10^{-6} $cm^3.s^{-1}$. For example, a K_{12} coagulation coefficient of 2 10^{-6} $cm^3.s^{-1}$ may be obtained for atmospheric aerosols with 0.15 µm radius and ^{106}Ru-Rh aerosols with 1 nm radius or an atmospheric aerosols with 0.55 µm radius and ^{106}Ru-Rh aerosols with 2 nm radius (table 1). Therefore, it can be noted that ^{106}Ru-Rh aerosols with nanometric radius and atmospheric aerosols for which radius corresponds to the accumulation mode (0.1 – 0.2 µm), the evolution of the ruthenium deposition, following the incident of May 18th, 2002, may be explained. The coagulation phenomenon will increase the size of ^{106}Ru-Rh aerosols initially released and therefore reduce the dry deposition velocity of them. Besides, for aerosols of which radius is comprised between a few nanometers and some thousands of nanometers, the deposition

velocity is inversely proportional to the particle radius. Particles thus stuck to the atmospheric aerosol may then be conducted over long distances as their deposition velocity is very low.

Table 1. Values of atmospheric aerosol radius and ^{106}Ru-Rh aerosol radius determined from sampling campaign carried out in La Hague

Atmospheric release of ^{106}Ru-Rh (Bq)	Dry deposition velocity of the atmospheric aerosols ($m.s^{-1}$)	Dry deposition velocity of the ^{106}Ru-Rh aerosols ($m.s^{-1}$)	Coagulation coefficient $K_{12}.10^{10}$ ($cm^3.s^{-1}$)	Time period $T_{1/2}$ (s)	Radius of the atmospheric aerosols (µm)	Radius of the ^{106}Ru-Rh aerosols (µm)
$4,5\ 10^{9}$	$1\ 10^{-4}$	$5.7\ 10^{-2}$	20000	140	0.15	$1\ 10^{-3}$
$4,5\ 10^{9}$	$1\ 10^{-4}$	$5.7\ 10^{-2}$	20000	140	0.55	$2\ 10^{-3}$

CONCLUSION

This study shows that the consideration of the aerosol coagulation phenomenon in the modelling of the atmospheric dispersion and deposition allows explaining the ^{106}Ru-Rh deposition measured in environment following the incident of May 18th in the COGEMA La Hague facility. To do so, one must take into account the evolution of two different aerosol size distributions and dry deposition velocities. Moreover, the consideration in the modelling of local phenomena (electric field, turbulence…) should allow explaining the significant value of the dry deposition velocity ($5,7\ 10^{-2}$ $m.s^{-1}$) observed in near field (700 m). Lastly, these various hypotheses shall be invalidated or validated using field experiments carried out producing fluorescein monodispersed aerosols as tracer. This technique is being developed by IRSN.

REFERENCES

Boulaud D. and Renoux A., 1998: Les aérosols, Physique et Métrologie, Lavoisier TEC DOC, 291 p.

COGEMA., 2001 : Rapport trimestriel de COGEMA La Hague, HAG055200121259.

Crabol B. and Maro D., 2001: Rejet dans l'environnement de ruthénium consécutif à l'incident du 18 mai 2001 sur l'établissement COGEMA de La Hague, Bilan et interprétation des mesures dans l'environnement, Rapport IPSN/DPRE/SERNAT n° 2001-36, 15 p.

Gensdarmes F., 2002: Dépôts des aérosols ultra-fins soumis à un champ électrique, Rapport IRSN/DPEA/SERAC n° 02-21, 10 p.

GRNC., 2002: Rapport du groupe de travail « Ruthénium », Groupe Radioécologie Nord Cotentin Eds, 75p.

Maro D., Baron Y., Germain P., Crabol B., Hebert D. and Solier L., 1999: Utilisation du krypton 85 rejeté dans l'environnement par l'usine de retraitement de La Hague comme outil d'étude de la dispersion atmosphérique: Bilan de 14 campagnes de mesures, Rapport IPSN/SERNAT/99-14, 18 p.

Maro D., Boulaud D., Germain. P., Hebert D. and Baron Y., 2002: Importance de la coagulation sur l'évolution de la granulométrie des aérosols : Application au rejet dans l'environnement de ^{106}Ru-Rh consécutif à l'incident du 18 mai 2001 sur l'établissement COGEMA de La Hague, Rapport IRSN/DPRE/SERNAT/2002-20, 17p.

ACKNOWLEDGEMENTS

We wish to thank Mrs Le Bar and Schgier, Ms Fitamant and the whole of the team at COGEMA's La Hague plant for their help in ensuring that this measurement campaign went smoothly and for forwarding the meteorological and ^{106}Ru-Rh emission readings. We would also like to thank Mr Baron of the French Navy for having taken very low-level measurements at the Roule mountain underground laboratory at Cherbourg.