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)

DEI / Service d'Étude du Comportement des Radionucléides dans les Écosystèmes

Contexte

The atmosphere is a major transfer path of pollutants released in gaseous form or in aerosols form from an industry to the land 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 substratum 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.

Objective

On May 18th, 2001, the spent fuel reprocessing plant of COGEMA La Hague (North West of France) released into the atmosphere, radioactive aerosols (^{106}Ru-Rh) which marked the near environment of the site.

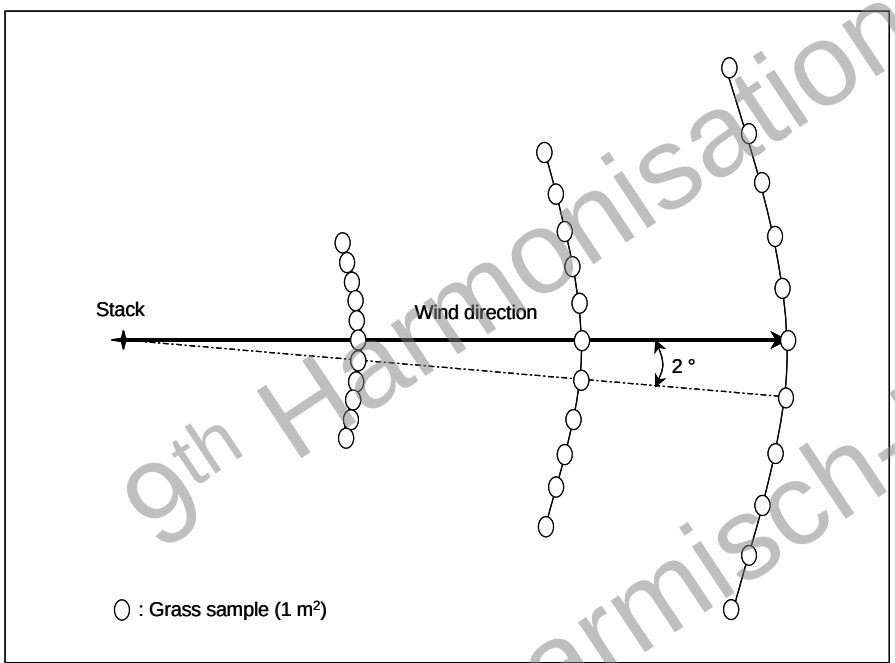
The purpose of my presention is to show the results of ^{106}Ru-Rh measurements carried out on grass, and the interpretation made regarding the operational deposition models.

Equipment and method

A failure in a gas-treatment line (on May 18th, 2001) 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. ^{106}Ru and ^{106}Rh are beta- and gamma-emitting radioelements.

Following this release, grass was sampled by different laboratories (COGEMA, ACRO, OPRI and IRSN) under wind of the facility.

Position of sample points regarding the discharge point and the average wind direction during the release



Grass was sampled on both sides of the average wind direction during the release between 700 m and 6,700 m from the discharge point.

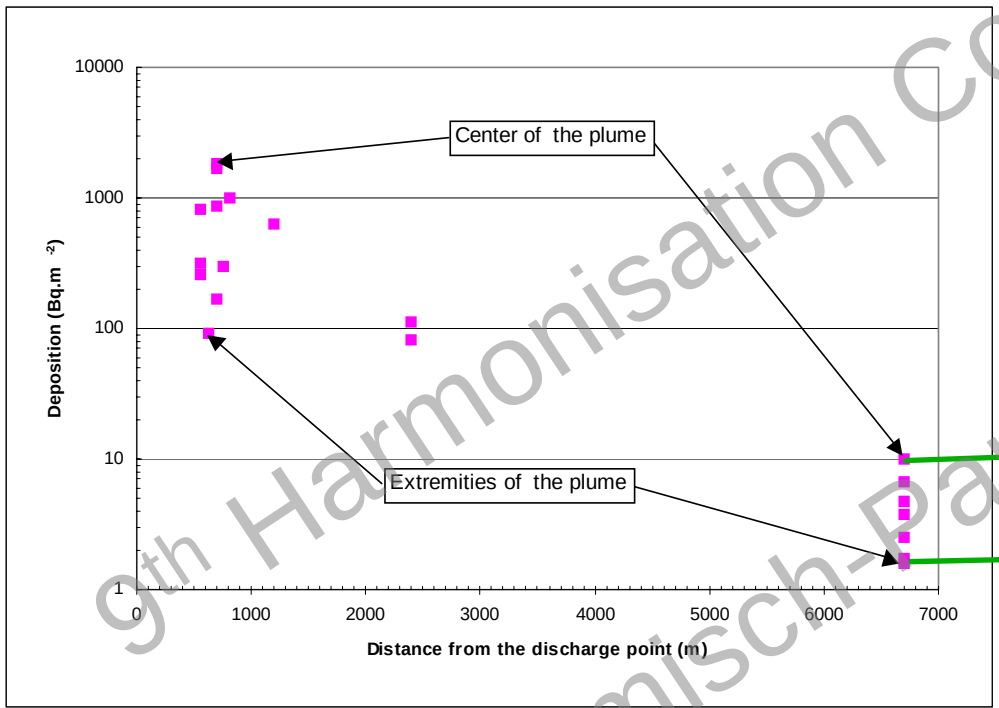
Measurements

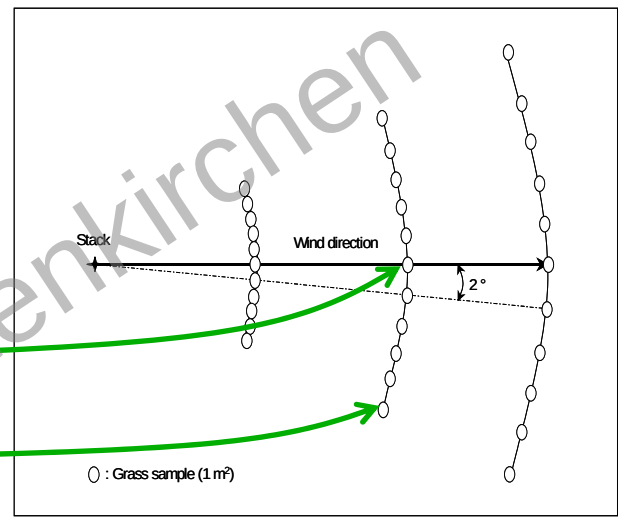
Grass samples were then dried to 60°C then ^{106}Ru-Rh concentrations were measured using gamma spectrometry in the Roule Mountain underground laboratory at Cherbourg (French Navy) with a very low background noise.

Roule underground laboratory (Cherbourg)

Gamma detector in the underground laboratory

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

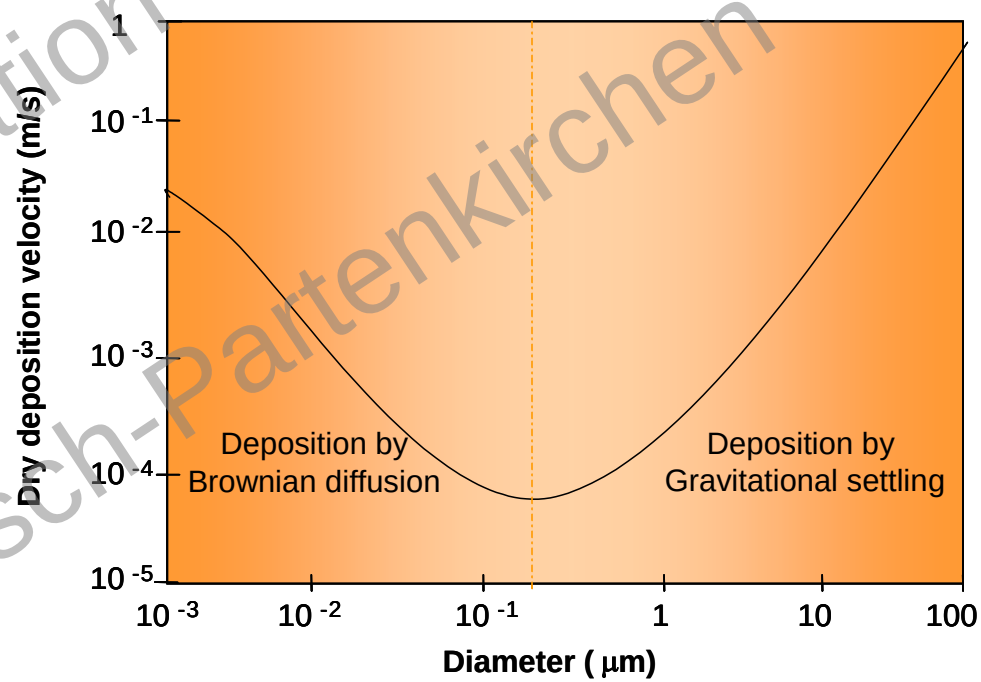




These results show a marking of the grass, up to a few thousands of Bq.m^{-2} of ^{106}Ru-Rh, at 700 m from the discharge stack, in the wind direction. This deposed activity reduces by 10 (100 Bq.m^{-2}) at 2,400 m and by 100 at least (10 Bq.m^{-2}) at 6,700 m.

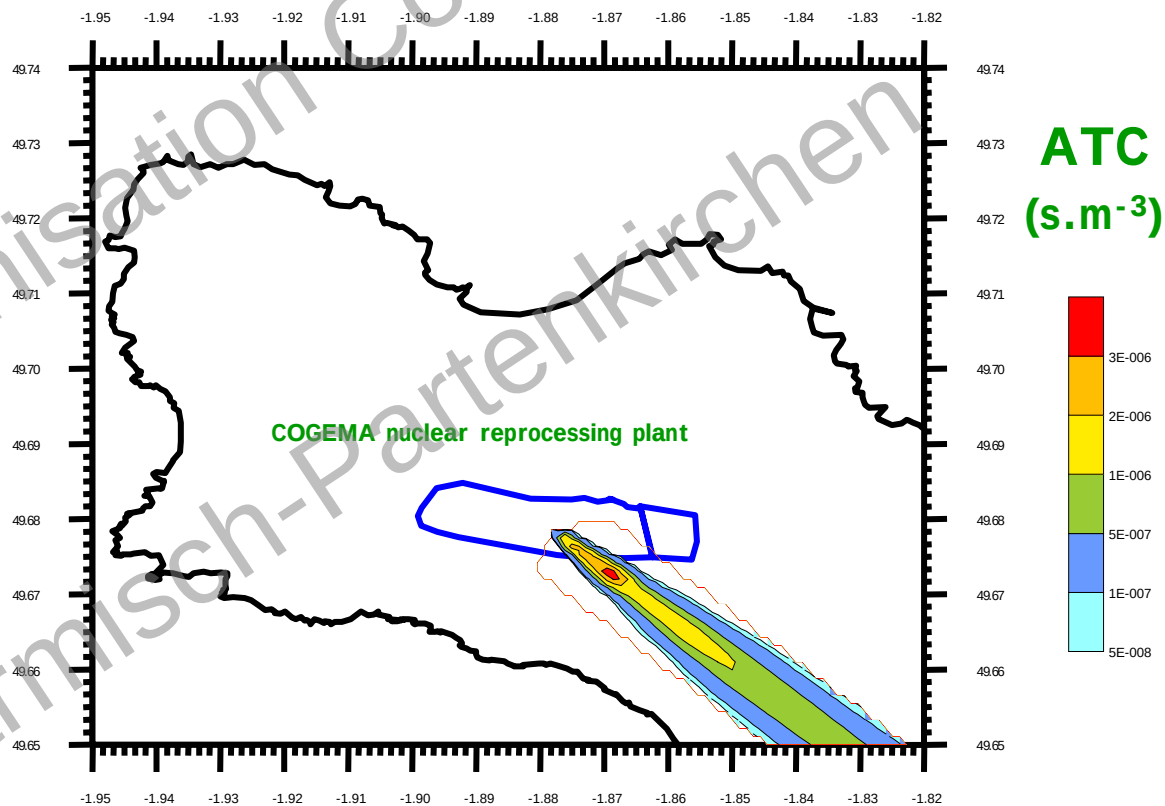
Dry deposition velocities

An atmospheric plume may progressively deplete in the wind direction, as aerosols stick to surfaces, such as leaves and soil. The deposition quantity may be determined from a term called dry deposition velocity. The dry deposition velocity values vary with the atmospheric stability, the surface condition, but also the aerosol granulometry. The most used hypothesis for operational atmospheric dispersion models, is a constant dry deposition. With no precise data the typical value of this deposition velocity is 5 10^{-3} $m.s^{-1}$.



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} (1/3)

Basing on the Atmospheric Transfert Coefficients (ATC) measured at 700 m from the IRSN atmospheric dispersion of ^{85}Kr study campaigns (experimental ATC) 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 at 5.7 10^{-2} m.s^{-1}.

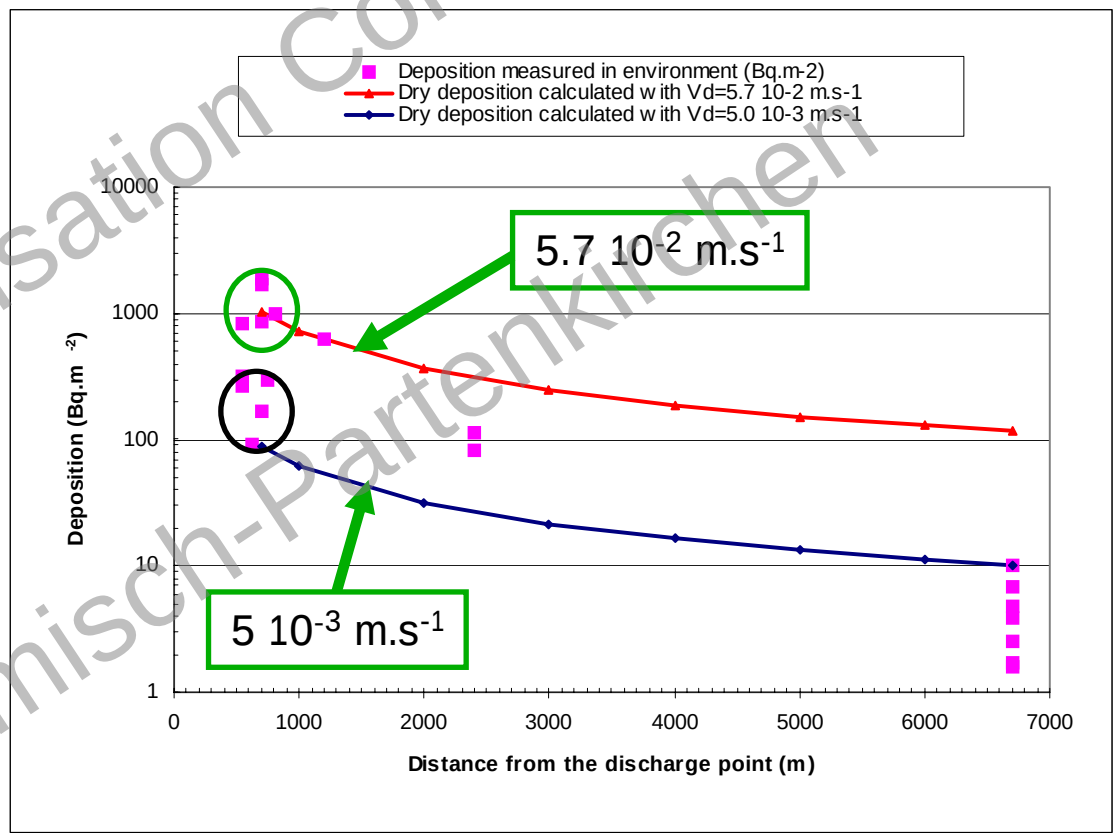


Comparison between ^{106}Ru-Rh deposition in environment with deposition calculated with dry deposition velocities of 5 10^{-3} m.s^{-1} and 5.7 10^{-2} m.s^{-1} (2/3)

This deposition velocity is higher (factor of 10) at 700 m than the deposition velocity of 5.10^{-3} m.s^{-1}, generally used for operational models.

Different hypothesis can explained this significant value of deposition velocity:

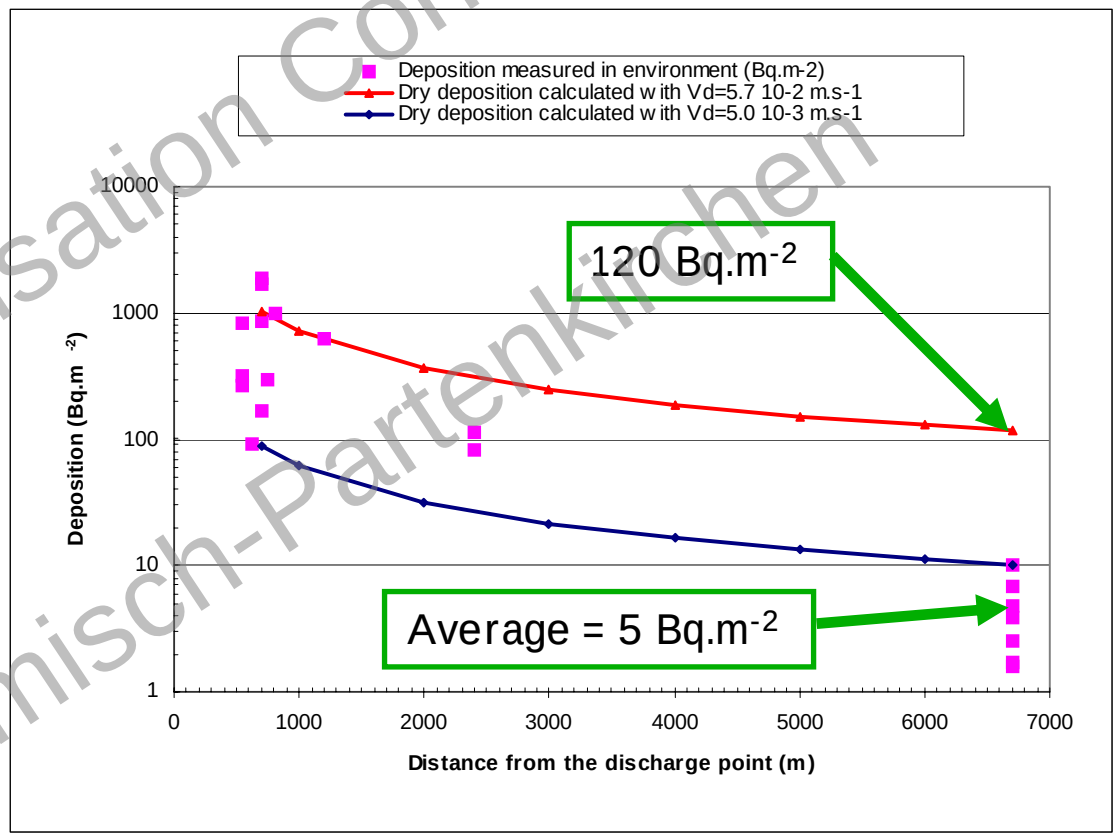
- electrostratic field,
- local turbulence,
- etc…



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} (3/3)

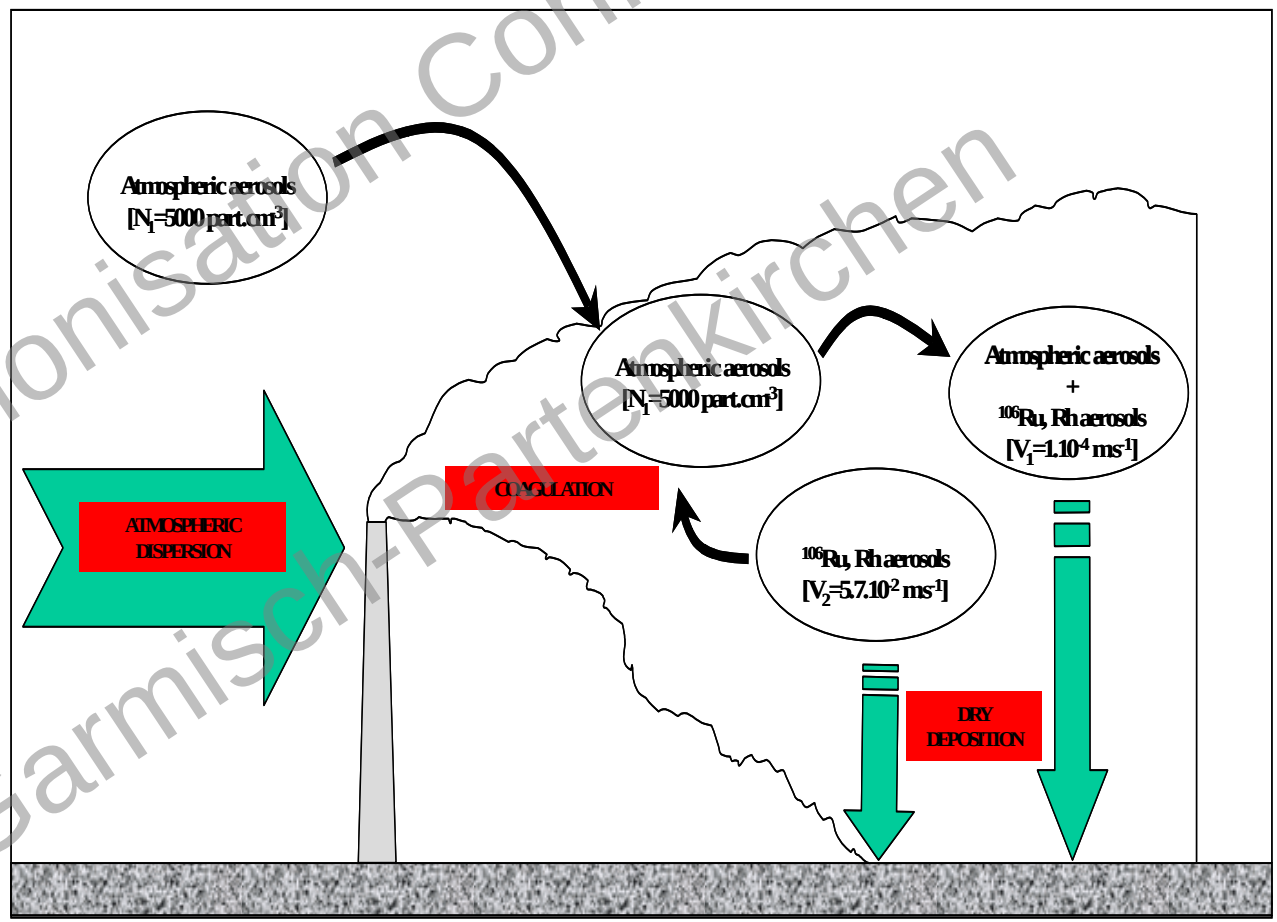
However, for this deposition velocity of 5.7 10^{-2} m.s^{-1} determined from measurements at 700 m from the discharge point, the activity settled in ^{106}Ru-Rh should be 120 Bq.m^{-2} at 6,700 m, whereas the activity measured in environment at this distance is about 5 Bq.m^{-2}.

We tried to determine if this deviation might be explained by the coagulation phenomenon of ^{106}Ru-Rh aerosols on atmospheric aerosols.



Comparison of measurement results with calculation results taking into account the coagulation phenomenon of ^{106}Ru-Rh aerosols on atmospheric aerosols (1/4)

In order to take into account the aerosol coagulation phenomenon 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 measured as equal to 5,000 particles per cm^3.



Comparison of measurement results with calculation results taking into account the coagulation phenomenon of ^{106}Ru-Rh aerosols on atmospheric aerosols (2/4)

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 :

$$\frac{dN_2}{dt} = -\frac{1}{2}.K_{12}.N_1.N_2 \quad \Leftrightarrow \quad \frac{dN_2}{N_2} = -\frac{1}{2}.K_{12}.N_1.dt$$

K_{12} is the coagulation constant

By integration, we obtain equation :

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

N_{02}: initial concentration of R_2 radius particles

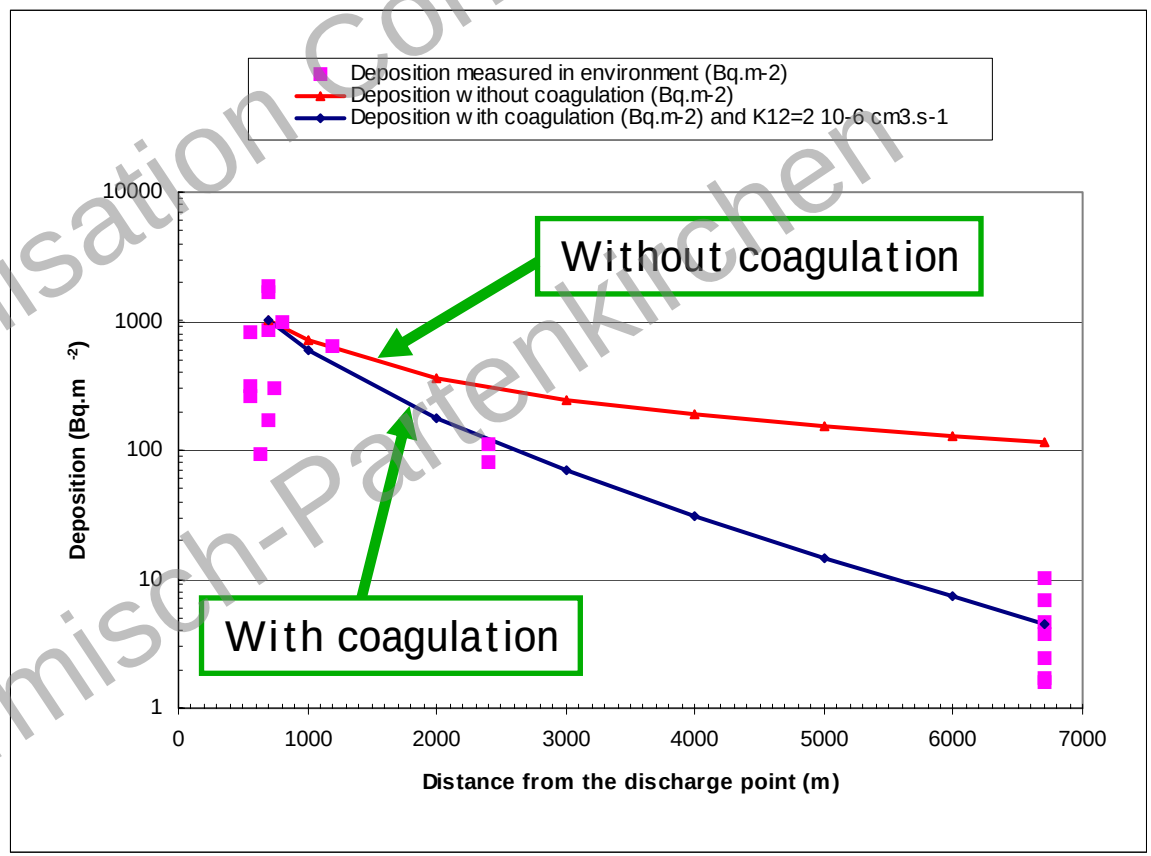
The N_2 population would have reduced by half after with the time period :

$$T_{1/2} = -\frac{2.LN(0.5)}{K_{12}.N_1}$$

Comparison of measurement results with calculation results taking into account the coagulation phenomenon of ^{106}Ru-Rh aerosols on atmospheric aerosols (3/4)

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, the best adjustment between measurements and modelling is determined for a K_{12} value of 2 10^{-6} $cm^3.s^{-1}$.



Comparison of measurement results with calculation results taking into account the coagulation phenomenon of ^{106}Ru-Rh aerosols on atmospheric aerosols (4/4)

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.

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 (1/2)

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 release 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).

Conclusion (2/2)

Lastly, these various hypotheses will be invalidated or validated using field experiments carried out producing fluorescein monodispersed aerosols as tracer. This technique is being developed by IRSN.