PRESENTATION AND INTERPRETATION OF FIELD EXPERIMENTS OF GASEOUS UF6 RELEASES IN THE ATMOSPHERE

B. CRABOL, D. BOULAUD, G. DEVILLE-CAVELIN
CEA / INSTITUT DE PROTECTION ET DE SURETE NUCLEAIRE
Département de Protection de l'Environnement et des Installations
BP 6 92265 Fontenay-Aux-Roses Cédex FRANCE

C. GEISSE, L. IACONA
EURODIF / PRODUCTION
Site de Tricastin
BP 175 26702 Pierrelatte Cédex FRANCE

ABSTRACT

An experimental programme concerning the behaviour of UF_6 released in gaseous phase in the atmosphere has been conducted in the years 1986-1989 by the french Atomic Energy Commission and Eurodif. Three field tests have been performed on the CEA/CESTA experimental site. These experiments permitted to get informations about the kinetics of the hydrolysis reaction of the UF_6, the behaviour of the hydrolysis products in the atmosphere and the granulometry of the solid particles.

RESUME

Un programme d'expérimentation sur site a été mené entre 1986 et 1989 par le Commissariat à l'Energie Atomique et Eurodif afin de mieux évaluer les risques pour l'environnement auxquels conduirait un rejet accidentel d'UF_6 sous forme gazeuse dans l'atmosphère. Trois essais ont été réalisés sur le terrain d'expérimentation extérieur du CEA/CESTA. Ces essais ont permis d'obtenir des informations sur la cinétique de la réaction d'hydrolyse de l'UF_6, sur la diffusion des produits d'hydrolyse dans l'atmosphère et la granulométrie des particules formées.

1. Introduction

The UF_6, used in the nuclear fuel processing, reacts with the water vapour of the air to give a solid product UO_2F_2 and a toxic gas, HF:

$$UF_6 + 2H_2O \text{ -----> } UO_2F_2 + 4HF$$

The numerical prediction of the consequences of an accidental release of UF_6 in the atmosphere needs therefore the following points to be considered :

. what is the kinetics of the hydrolysis reaction,

. how do UO_2F_2 and HF diffuse in the air, what are their chemical-physical forms and how do they deposit on the ground.

In order to answer to these questions, EURODIF, the operator of the uranium enrichment plant of Tricastin in the Rhône Valley, and the Institute for Protection and Nuclear Safety of the CEA, in cooperation with the U.S. Department of Energy (D.O.E.), decided some years ago to undertake an in-field experimental programme on UF_6 atmospheric dispersion. Three releases of gaseous UF_6 have been accomplished in 1986, 1987 and 1989 on the CEA/CESTA test site, near Bordeaux. This paper summarizes the main results obtained in these experiments.

2. Results and interpretation

2.1 - Concentrations in the atmosphere

The figure 1 presents the maximum values of the Atmospheric Transfer Coefficient (ATC, i.e., the ratio of the time integrated concentration at a given point over the released quantity) versus distance. It is observed that the ATC values in U and F are very close to each other. On the same figure, are also reported the SF6 results (SF6 is used in these experiments as a reference tracer). It can be observed that these values are very near to the previous ones, except at short distance from the source (it is difficult to attribute this discrepancy to a particular behaviour of the UF_6 plume in the first meters or only to the differences, even small, in the release conditions). It can be concluded that U and F diffuse similarly, and diffuse like a passive contaminant. Particularly, a major finding of these experiments is that no detectable depletion of the aerosols of UO_2F_2 in the plume due to the ground deposition occurs.

For analysis of the kinetics of the hydrolysis reaction, the atmospheric concentrations of U trapped on the particle filters in the first test have been measured : they were found to be close to the values collected at the same points in the bubblers, meaning that, after some seconds of travel time, all uranium was in form of particles UO_2F_2 and that the hydrolysis reaction had already ended. A confirmation of this fact is brought hereafter.

2.2 - Ground deposition

The figure 2 presents the deposition velocity of the uranium versus distance, on the points where the measured atmospheric concentrations were maximum on each cross-section. In both experiments, the velocity lies between 10^{-4} and 4.10^{-4} ms^{-1}, in good agreement with the sedimentation velocity which can be expected according to the measured particle sizes (see § 2.3). The velocity does not increase with the distance (on the contrary, it decreases), confirming that the hydrolysis is complete after a few seconds : if it was not the case, the creation of particles of UO_2F_2 during the transfer would increase the deposition velocity.

The mass ratio U/F measured in the deposits is presented on the figure 3 for the points in the wind axis. In the second test, this ratio is near by 6.3, which is the theoretical ratio in the UO_2F_2 compound. It means that only UO_2F_2 deposits on the ground, and indicates that no mixed compound UO_2F_2,HF is present in significant proportion in the plume. However, in the third test, a lower ratio is observed, particularly at distances greater than 40 m. These measurements do not seem significant as sand was found in important quantity in several of the Petri boxes where samples were collected, disturbing the analysis (a clear correlation is observed between the boxes where sand was found and the low value of the ratio). The sand could be easily transported by the wind due to the relatively low air humidity during this experiment.

2.3 - Granulometry

The granulometric distribution (aerodynamic diameters) in uranium is presented on the figure 4. The results in both experiments are very similar : they show a major mass fraction between 1 and 3 μm at all distances. Moreover, the distributions in F is very comparable to the one in U (which is not a surprise according to the preceding results concerning the solid compound formed during the transfer).

3. Conclusion

The experimental programme carried out in France in the years 1986-1989 allowed to get several important informations on the behaviour of UF_6 released in gaseous phase in the atmosphere :

- the hydrolysis reaction of the UF_6 is rapid, probably complete after some seconds;
- U and F diffuse similarly, without significant differences compared to the reference tracer;
- only, UO_2F_2 aerosols deposit on the ground;
- the major mass fraction of aerosols is observed for aerodynamic diameters between 1 and 3 μm;
- the deposition velocity of the aerosols lies between 1 and 4.10^{-4} ms^{-1}, values which are coherent with the expected sedimentation velocity for such particles.

Care must be taken not to extrapolate without caution all these conclusions to the cases of UF_6 released in liquid phase, for which, as it has been observed in different incidents, other and more complex phenomena may occur.

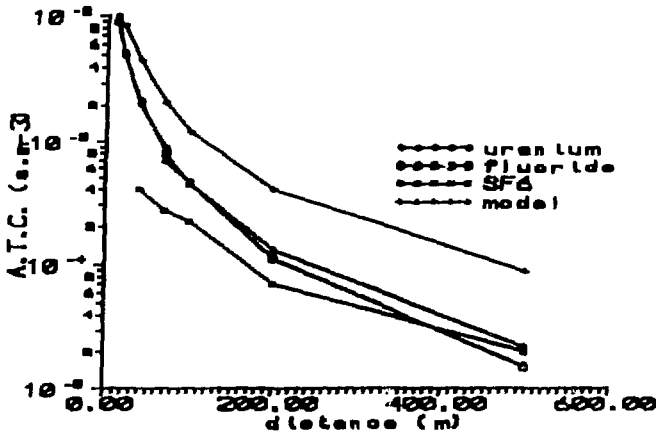

Figure 1. Second experiment. Atmospheric Transfer Coefficient in the wind axis versus distance

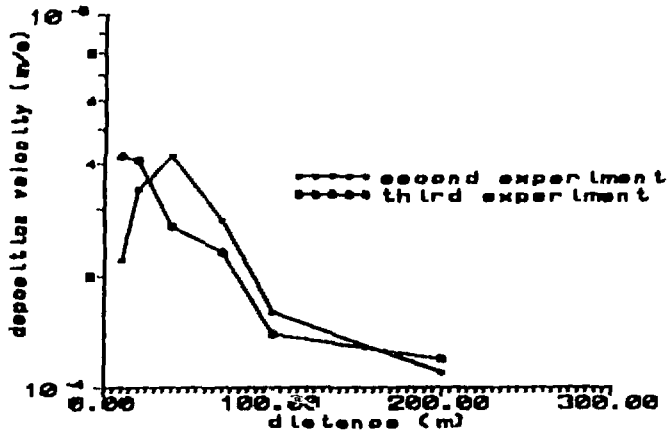

Figure 2. Deposition velocity of uranium in the wind axis

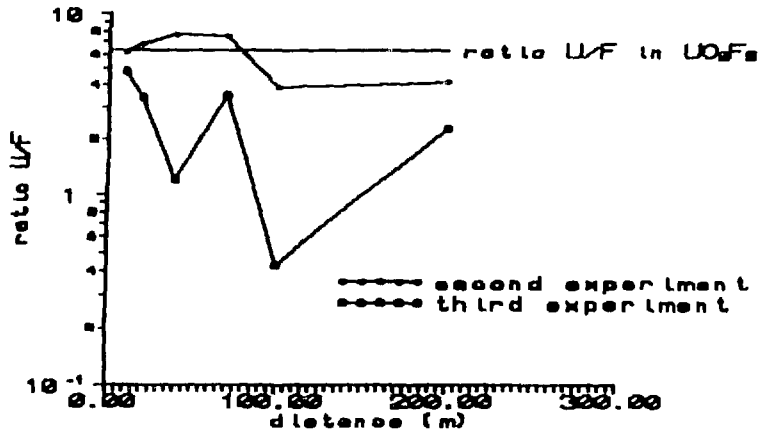

Figure 3. Mass ratio U/F in the ground deposit (wind axis)

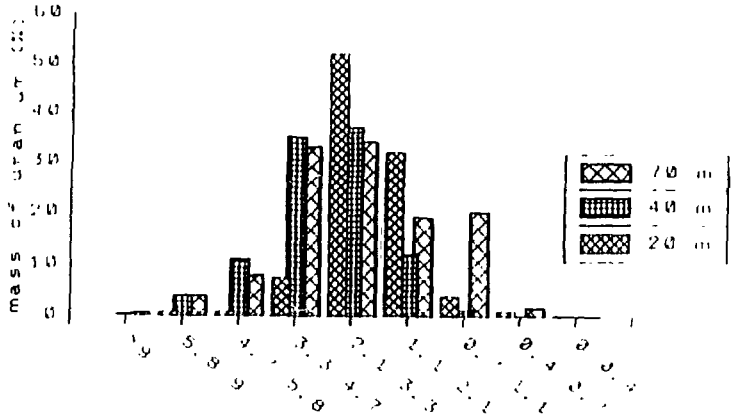

Figure 4. Second experiment. Distribution of the granulometry of the uranium