INPUT OF ARTIFICIAL RADIONUCLIDES FROM FRENCH RIVERS TO THE ATLANTIC AND MEDITERRANEAN

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Résumé

Cette étude présente l'évaluation des apports dissous et particulaires en radionucléides artificiels à l'Océan Atlantique et à la Méditerranée par les quatre grands fleuves français. Elle insiste plus particulièrement sur le comportement géochimique du $^{239-240}Pu$ et du ^{137}Cs en milieu estuarien.

^{238}Pu, $^{239-240}Pu$, ^{241}Am, ^{137}Cs, ^{144}Ce, ^{106}Ru, ^{125}Sb and ^{131}I have been measured in major French rivers and estuaries (Seine, Loire, Gironde and Rhône). $^{239-240}Pu$ specific activity in suspended river sediments averages 5-10 fCi/g. There are no significant differences between the Garonne and Seine river, only contaminated by atmospheric fall-out, and the Loire and Rhône where nuclear power plants are located. Conversely ^{137}Cs varies from 0.5 pCi/g in the Garonne river to a maximum of 5 pCi/g in the Rhône river. For both isotopes, distribution coefficient (K_D) and percentage of transported dissolved radionuclides are very close from one river to the other. The majority of ^{137}Cs and mainly $^{239-240}Pu$ flux in the rivers occurs in the particulate phase. More than 40% of the total French rivers discharge of ^{137}Cs and $^{239-240}Pu$ is supplied by Mediterranean rivers.

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$^{238}/^{239-240}$Pu activity ratios in suspended river sediment is unexpectedly high (0.1 - 0.25). These values can be ascribed to nuclear wastes in the Seine and possibly in the Loire and Rhône estuaries but a fractionation mechanism of plutonium isotopes during weathering processes is required for Garonne and Seine river samples. Particulate ^{137}Cs and $^{239-240}$Pu activities average respectively 0.3 pCi/g and 17 fCi/g in the Gironde estuary but reach 1 pCi/g and 65 fCi/g in the Seine estuary : these higher values, as well as ^{125}Sb, ^{144}Ce and ^{106}Ru activities, cannot be ascribed only to the atmospheric fall-out. In the Seine estuary, these radionuclides are likely to originate from Windscale and essentially La Hague reprocessing plant effluents.

A systematic survey of ^{137}Cs and $^{239-240}$Pu behaviour along with chlorinity shows a different picture for each nuclide. Particulate ^{137}Cs is constant in the Gironde where it is likely to be in a "lattice-held" position, decreasing seaward in the Loire estuary where it is ion exchangeable and increasing seaward in the Seine estuary owing to marine contamination. Dissolved ^{137}Cs activities increase seaward in all estuaries. Particulate $^{239-240}$Pu increase between river and low salinity areas is observed in all cases, whereas dissolved activities remain nearly constant (0.05 - 0.2 fCi/l) despite a possible removal. Various mechanisms, including a "coagulation" of riverine "dissolved" plutonium as well as an adsorption of marine plutonium are likely in the Gironde and Loire estuaries. In the Seine estuary, these mechanisms are magnified by a landward transport of marine contaminated sediment. Taking into account the various transfer processes which

have been observed and a sedimentation of 90% of river solid discharge in nearshore areas, the actual flux of riverine radionuclides to the open sea is predominantly particulate for plutonium 239-240 (>80%) and dissolved for cesium 137 (less than 20% in particulate form). These fluxes are very low as compared to direct atmospheric input over the Mediterranean sea.

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"Input of artificial radionuclides from French rivers to the Atlantic and Mediterranean"

Paper presented by J.-C. Guary (France)

Discussion

S.W. FOWLER: How do you explain the high $^{238}Pu/^{239,240}Pu$ ratios measured in the area upstream evidentally above any nuclear activities?

J.-C. GUARY: For the moment we do not have a satisfactory explanation for this observation.

C. BADIE: Quelles sont les caractéristiques de la technique de filtration utilisée?

J.-C. GUARY: Des volumes d'eau de 200 à 600 l sont filtrés sous pression sur des filtres Sartorius de diamètre 293 mm et de pores 0,45 µm.