A METHOD FOR FIELD DETERMINATIONS OF THE CHEMICAL AND PHYSICAL CHARACTERISTICS OF RADIONUCLIDES AFTER RELEASE INTO THE RIVER WATER

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The paper describes the field methods for the assessment of concentration levels and physical and chemical forms of radionuclides released by nuclear facilities, fall-out and medical uses into the river water. From the extended studies performed on the Po river, results of an experimental check carried out near E. Fermi, ENEL nuclear power plant (PWR) are presented and briefly discussed: samples simultaneously collected in a low rad waste tank, in the first mixing step and in river water at 10 Km downstream were characterized in order to investigate the behaviour of radionuclides into the river water.

EXPERIMENTAL

The system employed[1,2] carries out the separation of suspended matter by a sequential filtration equipment; the last step is a 0.3 um membrane filter. The recovery of ionic and non-ionic forms of radionuclides is obtained by ion exchange beds, alumina and NCFC, selective adsorbers for caesium. The system is able to treat large volumes of fresh water during a few hours; filters, resins and adsorbers are analysed by low background γ spectrometry with detection limit of a few pCi per sample.

RESULTS AND DISCUSSION

A typical situation without radioactive discharges is reported in table I from which the high sensitivities to fall-out and other radionuclide sources is pointed out. In table II, III, IV the distributions of the physical and chemical forms of radionuclides in the samples simultaneously collected during the radioactive waste release are presented. In table II one can see that ^{134}Cs and ^{137}Cs are almost completely in the cationic form; ^{54}Mn and mostly ^{60}Co are bound on solid material, whose composition turned out to be a mixture of amorphous oxides (CRUD). Suitable studies on the CRUD behaviour in the river water showed a full independence of water chemical-physical parameters. Table III reports concentration values and distributions at the first mixing with the river water after 30 min. of contact time and dilution factor 200. From table III and II one can see that caesium isotopes shift from the cationic species toward the solid one and ^{54}Mn, ^{60}Co almost maintain the same distribution. The distribution coefficient (Kd) of table III were calculated without CRUD activity that is assumed not to participate in the exchange mechanisms between water and suspended material. Table IV shows concentration values and distributions of samples collected 10 Km downstream (dilution factor = 140,000; 3 hours of contact time). A progressive shift of cationic forms toward the particulate turns out of table IV; ^{60}Co behaviour is complex owing to significant amounts of particulate, cationic, anionic and non-ionic forms. ^{103}Ru and ^{106}Ru come out in anionic and particulate forms; ^{95}Zr and ^{95}Nb are mostly bound on suspended matter (see table I too).

The adsorption mechanisms of radionuclides on suspended matter were carefully studied (see also paper No. 236 of this Congress) and the relation between specific activity and particle size was investigated: as the adsorption is roughly proportional to the specific area of particles a linear relation between log of radionuclide concentration and log of particle size should be found[3]. The speci-

fic activity of 5 granulometric classes of suspended matter evidenced this relation; for ^{137}Cs the coefficient of linear regression turns out to be -0.8. Then most ^{137}Cs appears to be transported by 5-10 μm particles in suspended sediment of different granulometric distributions (see fig. 1).

The system for field measurements of radionuclides in river water showed high sensitivity for natural and artificial radionuclides in order to follow the shift of physical and ionic characteristics. The fate of most radionuclides and of ^{137}Cs particularly is connected with the suspended sediment transport in the Po river. The granulometric class more significant for caesium transport has 5-10 μm particle size.

REFERENCES

1) D.E. Robertson et al. "Transport and depletion of radionuclides in the Columbia River"in Radioactive Contamination of the Marine Environment IAEA, Vienna 1973, p. 141-155.
2) A. Battaglia et al. "Sistema per il prelevamento e la determinazione contemporanea di un ampio spettro di radionuclidi in acque naturali". Simposio sulle Metodologie Radiometriche e Radiochimiche nella Radioprotezione. Pavia 1980.
3) W.W. Sayre et al. ('1963) with reference to J.L. Glenn in "Relations among Radionuclide Content and Physical Chemical and Mineral Characteristics of Columbia River Sediments". Report TID-25786 (1971).

TABLE I - Concentration (pCi/m^3 ± 2 σ) and physical chemical distributions of radionuclides (June 12, 1981). Suspended matter = 125 mg/l. Flowrate = = 605 m^3/sec.

	^{7}Be	^{95}Nb	^{95}Zr	^{103}Ru	^{106}Ru	^{125}Sb	^{131}I	^{177}Cs
Particulate	136±18	127.6±4.8	82.3±4.4	23.0±2.4	58±15	5.1±4.5	S.S.(*)	64.5±3.7
dissolved	S.S.(*)	15.4±2.1	12.2±2.0	36.1±2.6	76±13	2.2±2.0	14.4±1.9	7.6±1.4
% Particulate	100	89	87	39	43	70	-	89
% Cationic	-	2	2	1	5	-	-	9
% Anionic	-	6	7	48	42	30	100	2
% Non-Ionic	-	3	4	12	10	-	-	-
Kd (cm^3/g)	-	66,000	54,000	5,100	6,100	∿18,000	-	68,000

(*) S.S. = below the detection limit.

In the particulate form were moreover measured (pCi/m^3): ^{59}Fe 54.3±5.0, ^{60}Co 3.6±2.0, ^{203}Hg 66.3±4.0.

TABLE II - Concentration (pCi/l = 2σ) and physical chemical distributions of radionuclides in a low rad waste tank (June 10, 1981).

	^{54}Mn	^{60}Co	^{125}Sb	^{134}Cs	^{137}Cs
Particulate	399±35	16,110±110	309±43	283±38	2,620±75
dissolved	1,696±151	12,390±150	87±69	9,110±210	69,610±390
% Particulate	19	56.0	78	4	4
% Cationic	81	34.0	-	96	96
% Anionic	-	8.4	15	-	-
% Non-Ionic	-	0.7	7	-	-

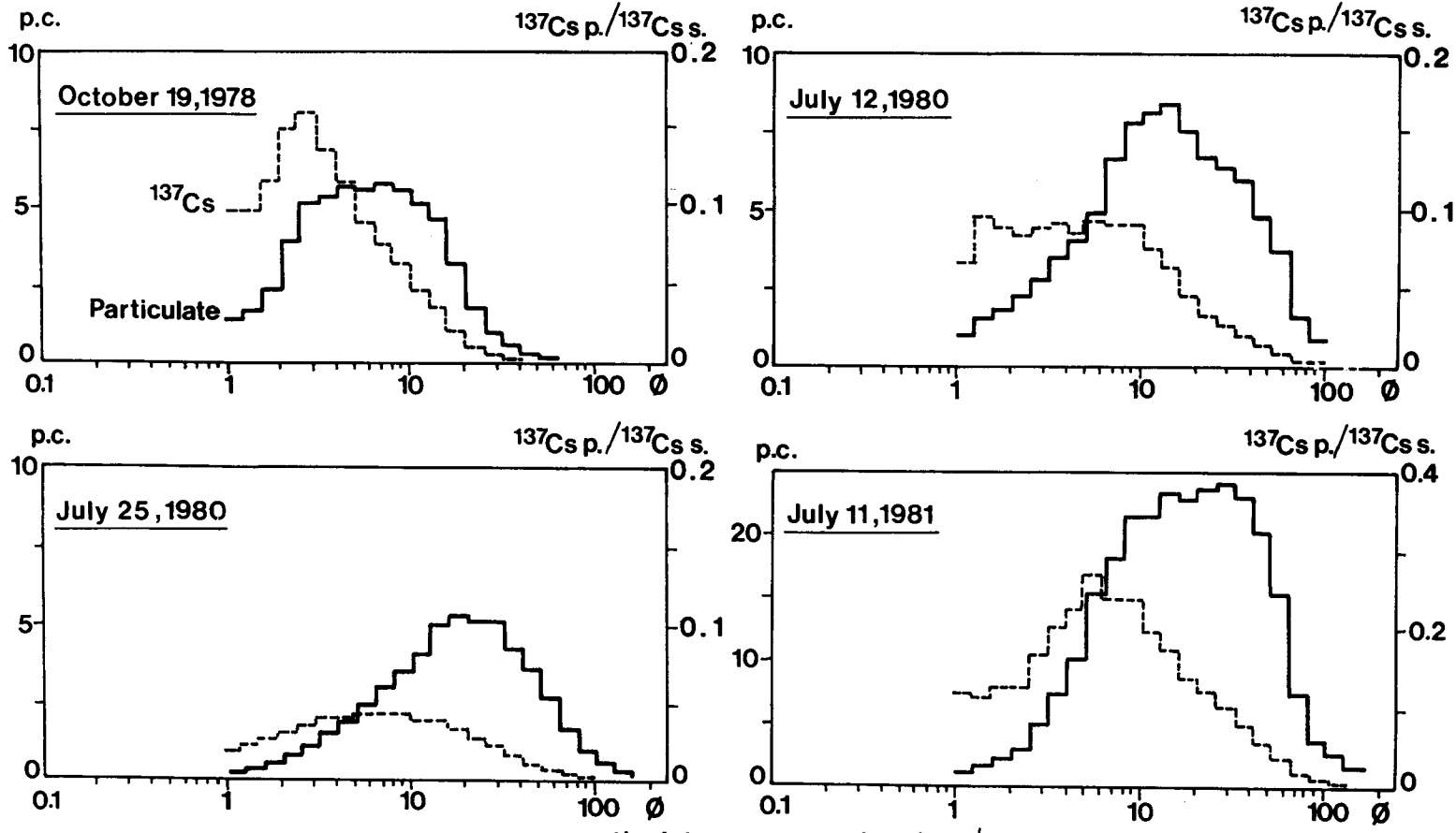
TABLE III - Concentration (pCi/l ± 2σ) and physical chemical distributions of radionuclides at the first mixing with river water (June 10, 1981), Suspended matter = 111 mg/l.

	^{54}Mn	^{60}Co	^{125}Sb	^{134}Cs	^{137}Cs
Particulate	2.42±0.68	87.5±1.1	1.23±0.83	16.1±0.8	140.3±1.6
dissolved	8.17±0.54	65.6±2.3	0.75±0.17	27.1±1.7	210.7±11.6
% Particulate	23	57.1	62	37	40
% Cationic	77	37.1	-	63	60
% Anionic	-	4.3	38	-	-
% Non-Ionic	-	1.5	-	-	-
Kd (cm^3/g)	3,000	1,800	11,000	5,700	6,200

TABLE IV - Concentration (pCi/m^3 ± 2σ) and physical chemical distributions of radionuclides at 10 Km downstream (June 10, 1981). Suspended matter = = 111 mg/l. Flowrate = 650 m^3/sec.

	^{54}Mn	^{60}Co	^{95}Nb	^{95}Zr	^{103}Ru	^{106}Ru	^{134}Cs	^{137}Cs
Particulate	5.1±2.4	145.8±5.8	134.9±8.9	84.8±8.9	19.8±4.9	49±9	41.5±5.8	335.3±9.7
dissolved	9.5±2.2	68.4±3.4	16.3±2.7	8.5±1.9	36.0±3.2	68±15	27.0±1.5	221.0±3.3
% Particulate	35	68.1	89	91	35	42	61	60
% Cationic	65	21.1	2	-	5	-	39	40
% Anionic	-	8.1	6	6	46	44	-	-
% Non-Ionic	-	2.7	3	3	14	14	-	-
Kd (cm^3/g)	4,800	5,200	74,000	90,000	5,000	6,500	13,200	10,700

Were moreover measured (pCi/m^3): ^{7}Be 120±43; ^{131}I 15.3±3.2 (88% anionic and 12% non-ionic).



p.c. = particulate concentration (mg/l)

^{137}Cs p./^{137}Cs s. = ^{137}Cs particulate/^{137}Cs soluble

Ø = diameter (µm)