MEASUREMENT OF SPECIFIC RADIONUCLIDES IN GASEOUS EFFLUENTS FROM NUCLEAR POWER PLANTS AND THEIR CONTRIBUTION TO RADIATION EXPOSURE

I. Winkelmann, K. Vogl
Institut für Strahlenhygiene
des Bundesgesundheitsamtes
Neuherberg, F.R.G.

INTRODUCTION

Requirements for the surveillance of radionuclides in gaseous and liquid effluents from nuclear power plants are outlined in regulatory guidelines. In addition to these nuclides which are measured by the operators there exist in the effluents from nuclear power plants other radionuclides which require special measuring techniques due to their mode of decay.

This paper describes methods for sampling and measuring P-32, S-35, Fe-55, Ni-63 and Tc-99 which are not measured on a routine basis. Results of measurements of samples taken at the stack of nuclear power plants in the Federal Republic of Germany are given. Also presented are measurement results of C-14 in the gaseous effluents from nuclear power plants and research reactors. An assessment is made of the radiation exposure to the public resulting from the emissions of these radionuclides.

SAMPLING AND MEASURING OF SPECIFIC RADIONUCLIDES

C-14 may be contained in the effluent air from nuclear power plants in the form of carbon monoxide, carbon dioxide, methane and other hydrocarbon compounds. Since in most cases, direct measurement of the beta emitter C-14 in the gaseous effluents of a nuclear power plant will hardly be possible because of the high activity of radioactive noble gases, continuous sampling and discrete measurements in the laboratory have to be relied on. Molecular sieves can be used for this purpose. This procedure also permits the continuous sampling of gaseous compounds of phosphorus and sulphur for determining the radionuclides P-32 and S-35. After sampling, these three radionuclides are eluted and precipitated in order to prepare measurement samples. The activity is determined using a liquid scintillation spectrometer.

Since 1982 the Federal Health Office has been conducting measurements of technetium emissions in effluent air from nuclear power plants. For the purpose of selecting a suitable measurement technique for the detection of this nuclide in liquid and gaseous forms and as a particulate, a series of separation methods was first examined.

Sampling of Fe-55 and Ni-63 in the effluent air from nuclear power plants is done by continuous separation of particulates on filters in a bypass of the stack. After ashing the filter samples, iron and nickel are precipitated as carbonates. The iron is extracted as a chlorine complex and nickel is precipitated with dimethylglyoxim from the remaining solution. The samples are measured by liquid scintillation spectrometry.

RESULTS AND DISCUSSION

From 1978 to 1982 carbon dioxide-bound C-14 emissions from nuclear power plants have been measured by the Federal Health Office. Additional measurements of C-14 as carbon monoxide or organically bound C-14 were performed. Table 1 shows the results of C-14 measurements for the years 1978 to 1982. Where as in boiling water reactors nearly the entire C-14 activity is emitted as carbon dioxide, pressurized water reactors emit only approx. 5 - 50 % as carbon dioxide. With this data collected from 1978 to 1982 a yearly mean total C-14 emission of 200 GBq/GWe can be calculated for a nuclear power plant with a pressurized water reactor and 500 GBq/GWe for a plant with a boiling water reactor. In addition, during the years 1980 to 1982 C-14 samples were taken from research reactors for several weeks, respectively. Assuming a constant power, the yearly emissions from research reactors were estimated and compiled in table 2.

Since the beginning of 1983 the emissions of the pure beta-emitters P-32 and S-35 in the gaseous effluents have been determined. Table 3 contains the results of P-32 and S-35 measurements up to date. Results of technetium measurements can be tentatively expected by 1984. From 1979 to 1982 the emissions of Fe-55 and Ni-63 were determined in the gaseous effluents from nuclear power plants. The results from these measurements are compiled in table 4 and 5.

RADIATION EXPOSURE

The radiation exposure was calculated on the basis of averaged annual releases of C-14, Fe-55 and Ni-63 with gaseous effluents. A similar calculation was performed for P-32 and S-32 using the data from table 3. The results are given in table 6 and are below 1 µSv/a.

Table 1: Annual releases of C-14 with gaseous effluents from nuclear power plants

Nuclear power plant	Annual releases in GBq/a* 1978	1979	1980	1981	1982
Brunsbüttel	170	7.4	30	240	59
Gundremmingen	3.7	3.7	0.74	0.26	-
Isar	-	-	-	180	4.8
Würgassen	230	160	350	200	130
Philippsburg	-	-	-	6.3	93
Biblis A	22 (56)	-	-	-	52(110)
Biblis B	15 (15)	-	-	-	22
Neckarwestheim	7.4(140)	22(160)	11(96)	22(110)	44
Obrigheim	19 (15)	74(33)	22	37	26
Stade	44	56(70)	41(78)	210	78
Unterweser	-	19(230)	56	33	41

* The results from measurements of carbon monoxide- and hydrocarbon-bound C-14 are listed in brackets

Table 2: Extrapolated annual releases of C-14 from research reactors

Research reactor	Type	Power in MW	Annual releases of C-14 in GBq/a*
Neuherberg (FRN)	LWR	0.14	0.081
Karlsruhe (MZFR)	HWR	200	1400(1400)
Karlsruhe (FR2)	HWR	44	78(48)
Jülich (AVR)	GCR	46	41(22)
München (FRM)	LWR	4.15	- (1,6)

* Values of C-14 as carbondioxide are listed in brackets

Table 3: Releases of S-35 and P-32 with gaseous effluents during the first half of 1983

Nuclear power plant	Activity releases (MBq) S-35		P-32	
	I. Quarter	II. Quarter	I. Quarter	II. Quarter
Brunsbüttel	<100	<100	<100	<100
Stade	1000	750	<100	<100
Unterweser	230	320	<100	<100
Würgassen	<100	<100	<100	<100

Table 4: Annual releases of Ni-63 with gaseous effluents from nuclear power plants

Nuclear power plant	Annual releases (MBq/a) 1979	1980	1981	1982
Brunsbüttel	22 (3)	1.2 (3)	0.06 (1)	0.15
Gundremmingen	4.2	0.11 (3)	0.053 (2)	0.15
Isar	1.0 (3)	0.37 (2)	0,38 (2)	6.68
Kahl	0.014 (3)	0.083 (3)	0.001 (2)	0.01
Lingen	0.08	0.31 (2)	0.02 (3)	0.04
Philippsburg	0.05 (1)	0.21 (3)	0.39 (2)	0.71
Biblis A	0.04	0.066 (2)	0.01 (2)	0.03 (3)
Biblis B	0.04	0.02 (3)	0.008 (1)	0.04
Neckarwestheim	0.28 (3)	1.1 (2)	0.45 (3)	0.58
Obrigheim	11	11 (3)	-	13.6
Stade	1.1 (3)	1.3	0.066 (2)	0.09
Unterweser	0.46 (1)	0.46 (1)	0.32 (1)	0.04
Grafenrheinfeld	-	-	-	0.1

(1) Value of one quarter
(2) Value of two quarters
(3) Value of three quarters

Table 5: Annual releases of Fe-55 with gaseous effluents from nuclear power plants

Nuclear power plant	Annual releases (MBq/a) 1979	1980	1981	1982
Brunsbüttel	1200 (3)	13 (3)	28 (3)	44
Gundremmingen	34	2.5	3.7	
Isar	452	250	570	600
Kahl	0.13	0.27(2)	0.15(3)	0.01(2)
Lingen	1.6	8.0 (3)	8.6	2.0
Philippsburg	18	23	42	6.5 (3)
Biblis A	1.0	4.9	9.1	13
Biblis B	1.1	8.7 (3)	2.9 (3)	3.2
Neckarwestheim	8.8	12 (3)	81	15.8
Obrigheim	25	20	2.5 (1)	1.6 (1)
Stade	12	7.3	1.2 (2)	3.0
Unterweser	2.8	4.8 (3)	3.4 (3)	22
Grafenrheinfeld	-	-	-	9

(1) Value of one quarter
(2) Value of two quarters
(3) Value of three quarters

Table 6: Radiation exposure by inhalation and ingestion from the emission of 100 GBq/a C-14, 100 MBq/a P-32, 100 MBq/a S-35, 10 MBq/a Fe-55 and 1 MBq/a Ni-63 in gaseous effluents from nuclear power plants.
(a longterm dispersion factor of $1,7\ 10^{-7}$ is assumed)

Organ	Exposure (nSv/a) C-14	P-32	S-35	Fe-55	Ni-63
Total body	54	0,8	0,5	5E-3	3E-3
Bone	270	27	2,2	2E-2	0,2
Liver	54	1,4	-	3E-2	1E-2
Thyroid	54	-	-	-	-
Kedneys	54	-	-	-	-
Lungs	54	-	-	1E-2	-
G-I-Tract	54	14	1,9	2E-2	3E-3