MEASUREMENT CONCERNING THE CONVERSION OF ELEMENTAL IODINE DURING DISPERSION IN THE ATMOSPHERE

Karl Heinemann
Department of Safety and Radiation Protection
Kernforschungsanlage Jülich GmbH
Jülich

1. Introduction

Pollutants reach vegetation, soil and surface waters via deposition from waste air plumes and thus enter into the food chain. Dry deposition (fallout) is today conventionally described by the deposition velocity and deposition by precipitations (washout) by the washout constant.

As a critical nuclide, I 131 plays a particular role both in normal operation and also during accidents in nuclear engineering facilities. In the case of radioiodine a differentiation must be made between elemental, aerosol (attached to aerosols) and organically bound iodine, since according to /1/ the deposition velocities and the washout constants, and thus also the ingestion doses, vary by approximately one order of magnitude for each of these forms of iodine. Elemental iodine accordingly proves to be the radiologically most important form of iodine.

The composition of the forms of iodine was measured during emission /2,3,4/. No answer has yet been found to the question in how far elemental iodine is converted into other forms of iodine during transport in the atmosphere. Experimental studies into this problem are carried out at the Jülich Nuclear Research Centre.

2. Test Method

Iodine in elemental form was released from the 50 m platform of the meteorological tower at the Jülich Nuclear Research Centre and air samples were taken downwind at distances between 200 and 1000 m with a tripartite filter system.

In order to produce elemental iodine a maximum of 100 mCi of I 123 (half-life = 13.2 h), supplied in 5 ml of carrier-free KI solution, was put into a reaction vessel in which 5 ml of saturated $K_2Cr_2O_7$ solution and 1 mg I_2 had previously been evaporated to dryness. At the time of emission the reaction flask was heated up to approx. 300 °C and the resulting iodine was expelled with a gas flow. The additional iodine concentration in air thus generated is comparable to the natural concentration.

The two sampling stations were switched on at the beginning of emission and shut down again 5 minutes after ending emission. The air first flowed through an aerosol filter at a flow rate of about 10 m^3/h, then through an I_2 adsorber (Südchemie DMS 11)

and finally through activated charcoal treated with KI. The residence time of the air in the adsorbers was 0.1 s. First the aerosols, then elemental iodine and finally the remaining gaseous iodine were removed from the air stream in this filter system and each form of iodine was separately measured by means of gammaspectrometry after completing the experiment.

The residual gaseous iodine collected in the activated charcoal filter mainly consists of organically bound iodine. However, it may also contain other iodine compounds with similar physico-chemical properties, thus causing similar radiation exposures. In the following this iodine fraction is designated organic iodine.

The series of experiments, which has not yet been completed extends over several years for the following reasons. In the first place, the number of experiments per year is limited to 5 by the licensing authority. However, this number cannot even be completely exploited since the possible production dates for I 123 seldom coincide with suitable diffusion conditions.

By using short-lived iodine 123 the potential radiation exposure to the public is greatly reduced in comparison to I 131. On the other hand, the short half-life of I 123 leads to the fact that due to the limited measuring capacity during one experiment air samples can only be taken and measured at a maximum of two locations.

3. Results and Discussion

The results obtained up to now and the meteorological parameters measured at the same time are compiled in Table 1 and Table 2. Apart from the wind direction and wind velocity, determined at release height of 50 m, all other meteorological parameters refer as usual to measurements at 2 m above ground level.

The results of the iodine measurements already show iodine conversions at a very short distance. It can be seen from Table 1 that, as a rule, a larger and larger fraction of elemental iodine is converted with distance and that the percentage of organic iodine increases. However, conclusions cannot yet be drawn from the few measurements available about whether there is a distance after which an equilibrium distribution of the forms of iodine is established, where this distance is and what form the equilibrium distribution takes.

A suspected correlation of the degree of conversion e.g. with the air temperature or reaction time (last column of Table 1) resulting from the distance of the field point from the source and the wind velocity has not been established. All the measurements have therefore been combined and the mean values, standard deviations and fluctuation ranges given below were determined. As an average over all the measurements

Table 1 Results of the Iodine Measurements

Experiment No.	Date	Time	Distance (m)	Relative Iodine Fractions (%)[1]			Reaction Time (s)
				aerosol	elemental	organic	
1	16.12.80	12.00-13.00	290	47	41	<12	88
2	14.07.81	14.00-15.10	250	8	39	53	46
3	13.10.81	12.00-13.05	220	59	22	18	25
			850	10	12	78	96
4	30.06.82	11.20-12.25	400	28	45	27	93
			1100	45	45	10	256
5	21.09.82	12.50-13.50	1000	12	86	< 2	123
6	20.10.82	11.15-12.15	430	15	83	< 2	84
			915	16	58	<25	179
7	15.06.83	11.45-12.32	230	16	66	<18	38
8	13.09.83	14.21-15.00	370	31	64	5	69
			450	29	60	11	83
9	20.09.83	12.50-13.20	400	47	51	2	67
			480	45	43	11	80

1) "<" smaller than detection limit

Table 2 Meteorological Conditions During the Experiments

Experiment No.	Wind Direction (°)	Wind Velocity (m/s)	Pressure (Torr)	Temperature (°C)	Humidity (%)	Diffusion Category
1	289	3.3	760	5.0	68	D
2	265	5.4	756	21	67	C
3	242	8.9	749	7.2	78	D
4	244	4.3	759	17.1	63	C
5	221	8.1	747	20.3	60	D
6	213	5.1	756	14.9	60	C
7	290	6.1	758	14	57	C
8	248	5.4	754	13.9	76	C
9	263	6.0	761	16.4	59	B

only about 50 % elemental iodine was detected, whereas about 30 % was accounted for by aerosol and about 20 % by organic iodine.

iodine form	relative fractions (%)		
	arithmetic mean	standard deviation	fluctuation range
aerosol	29	17	8 - 59
elemental	51	13	12 - 86
organic	20	22	<2 - 78

During transportation of iodine in the atmosphere every form of iodine can in principle be converted into every other. An equilibrium is finally established which can be dependent, amongst other aspects, on the temperature of the air, the intensity of cosmic radiation but also on the reactants or catalysts present in the air. It is therefore to be expected that each measurement of forms of iodine in the vicinity of a point source will provide different results. This explains the large standard deviations.

4. References

/1/ Der Bundesminister des Innern, Allgemeine Berechnungsgrundlage für die Strahlenexposition bei radioaktiven Ableitungen mit der Abluft und in Oberflächengewässer, Gemeinsames Ministerialblatt 1979, 371

/2/ H. Deuber, Die physikalisch-chemischen Radiojodkomponenten in der Abluft eines Druckwasserreaktors, KFK Report 3207, 1981

/3/ H. Deuber, Die physiaklisch-chemischen Radiojodkomponenten in der Abluft eines Siedewasserreaktors, KfK Report 3424, 1982

/4/ K. Heinemann, Messung von elementarem, aerosolförmigem und organischem Jod in der Abluft, Proceedings of 5th Fachgespräch "Überwachung der Umweltradioaktivität", 22-24 March, 1983, in Karlsruhe, to be published