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Long-range transport of gaseous ^{131}I and other radionuclides from Fukushima accident to Southern Poland

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HIGHLIGHTS

- Only volatile gamma emitters were found in Fukushima plume in Poland.
- Original method of measurements for gaseous ^{131}I is presented.
- Results suggest Fukushima release below 10% of that from Chernobyl.
- The exchange between gaseous and aerosol fraction of ^{131}I affects activity ratios.

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ABSTRACT

A serious accident at Fukushima Dai-Ichi NPP triggered radioactive emission to the atmosphere on 12 March 2011. The results of gamma spectrometric measurements of both gaseous and aerosol fraction of the air, collected in Krakow over the period from March 21 till the end of May 2011, as well as wet and dry deposition recorded from March till the end of October 2011, are presented in this paper. Krakow happened to be the first Polish location where radioactive isotopes characteristic for reactor releases, such as ^{131}I, ^{132}I, ^{129m}Te, ^{132}Te, ^{134}Cs, ^{136}Cs, and ^{137}Cs, were detected. The maximum activity for aerosols equal to (5.73 ± 0.35) mBq/m^3, (0.461 ± 0.041) mBq/m^3 and (0.436 ± 0.038) mBq/m^3 for ^{131}I, ^{134}Cs and ^{137}Cs, respectively, was recorded for March 29, 2011. The data on the fallout are also given. The results of the radiochemical analysis of aerosol samples showed no traces of plutonium or americium isotopes associated with the disaster to be detected. The results of air activity concentration from Fukushima accident observed in Central Europe, Poland, in comparison to those of Chernobyl accident observed in Japan are presented and discussed. The comparison has revealed a discrepancy in the recognized relative scale of both accidents, and important difference in long distance transport of contamination, to exist. An attempt to explain the variation in the activity ratios between the aerosol fraction for ^{131}I and ^{137}Cs as resulting from exchange between the gaseous and aerosol fractions of ^{131}I while the contamination had been propagating, is made.

1. Introduction

A strong earthquake near Japan Islands, followed by enormous high tsunami on March 11, 2011 resulted in a serious overheating damage to have occurred in 3 reactors of the Fukushima Daiichi Nuclear Power Plant (Tanaka, 2012). Radioactive emission to the atmosphere began on March, 12 and lasted for over 2 weeks (IAEA, 2011). A radioactive plume migrated across the North Pacific, Northern America, Arctic and arrived over Europe from the north-western direction (Masson et al., 2011, MacMullin et al., 2012). According to meteorological modelling available on the Internet, already on March 21–23 it was predicted for the cloud to be present over central Europe between March 24–27 (NOAA, 2011).

Though at that time, the data on the release were scarce, from the spectra published in the open media one could expect only radioisotopes of volatile elements such as iodine, tellurium, and caesium to be detected. Among them iodine was the most interesting, as due to its complex chemistry, it can be transported both on aerosol and in gaseous fractions. A long-distance transportation, which is definitely the case for Japan release detectable in Central

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Table 1
Results of gamma spectrometric measurements of ground air activity concentration in Krakow following Fukushima accident for one day sampling cycle.

Sampling start/stop[a]	V [m^3]	Activity concentration [μBq/m^3]						
		^{131}I aerosols	^{131}I gas	^{137}Cs	^{136}Cs	^{134}Cs	^{132}Te/^{132}I	^{7}Be
21/24.03	17 957	105 ± 5	135 ± 42	<9	<7	<5	<4	3810 ± 270
24/25.03	5975	84 ± 7	286 ± 103	<15	<11	<11	<8	4290 ± 310
25/26.03	6386	<12	<48	<9	<7	<36	<3	3800 ± 280
26/27.03	5256	840 ± 37	604 ± 170	<17	<13	16 ± 8	<9	3310 ± 240
27/28.03	7468	1570 ± 60	2040 ± 860	83 ± 11	22 ± 10	105 ± 20	45 ± 10	4680 ± 350
28/29.03	5385	3600 ± 200	5200 ± 1200	254 ± 27	32 ± 15	206 ± 35	109 ± 17	4980 ± 390
29/30.03	5763	5730 ± 350	5220 ± 340	436 ± 38	28 ± 16	461 ± 41	124 ± 39	5690 ± 440
30/31.03	7264	2910 ± 120	3360 ± 260	296 ± 25	24 ± 13	307 ± 28	<9	6040 ± 450
31.03/1.04	6177	908 ± 42	1120 ± 110	36 ± 6	<11	35 ± 8	<8	6330 ± 450
1/2.04	6396	668 ± 33	1150 ± 230	41 ± 10	<19	75 ± 22	<10	5720 ± 430
2/3.04	6628	718 ± 35	950 ± 150	42 ± 11	<18	54 ± 17	<10	4340 ± 330
3/4.04	6230	2151 ± 90	1520 ± 410	411 ± 30	27 ± 5	420 ± 50	41 ± 10	7500 ± 540
4/5.04	8201	413 ± 18	625 ± 95	45 ± 5	<8	56 ± 7	<6	5220 ± 370
5/6.04	4261	494 ± 27	510 ± 220	43 ± 15	<28	44 ± 13	<15	3760 ± 310
6/7.04	6689	650 ± 27	930 ± 250	46 ± 6	<10	45 ± 8	<7	5490 ± 390
7/8.04	6554	438 ± 48	680 ± 320	60 ± 15	<20	67 ± 27	<10	4840 ± 390
8/9.04	6067	308 ± 20	775 ± 90	82 ± 14	<20	64 ± 21	<11	3960 ± 310
9/10.04	6788	182 ± 16	940 ± 190	<24	<18	25 ± 10	<9	2410 ± 210
10/11.04	6625	239 ± 15	1060 ± 360	42 ± 5	<7	45 ± 16	<3	2740 ± 650
11/13.04	12 882	157 ± 13	402 ± 59	40 ± 8	<10	37 ± 11	<5	3190 ± 240
13/15.04	13 106	91 ± 7	249 ± 32	<18	<15	18 ± 5	<4	2420 ± 180
15/18.04	18 233	126 ± 14	167 ± 27	28 ± 6	<11	32 ± 6	<8	2500 ± 190
18/21.04	14 775	45 ± 3	88 ± 28	23 ± 3	2.4 ± 1.1	14 ± 2	11 ± 7	7650 ± 540
21/26.04	29 154	11 ± 1	65 ± 18	5.3 ± 0.8	<1	6.5 ± 0.8	3 ± 2	6040 ± 430

[a] Year 2011.

Europe, i.e. over a 10 000 km distance, can result in a fractionation between gas and aerosol (ie. the ratio of the two fractions observed in Europe is not necessarily the same as was released in Japan). This ratio can be affected either by chemical reactions which may occur due to oxidation conditions in the air, sunlight, or exposition to UV radiation, also the physical removing of gasses and aerosols from radioactive cloud is different as well as their mutual exchange can happen. Thus it seems very interesting to study both aerosol and gaseous fractions of iodine isotopes in the nuclear accident cloud. Traces of all other nuclides in relation to iodine could also shed some light onto understanding differences in the air transport mechanism for different radionuclides. From the dosimetric point of view apart from measuring particular fraction of iodine from nuclear accidents, it is of vital importance to measure its gaseous fraction, since it can produce significant part of the dose from iodine. It is of greater importance for shorter transportation distances where higher iodine activity concentration in the air can be detected, so developing the method of gaseous iodine detection seems particularly important.

2. Material and methods

2.1. Sample collection

Air sampling was carried out at one place, namely the Institute of Nuclear Physics in Krakow – IFJ PAN, 50.04 N, 19.58 E, 215 m a.s.l, with two aerosol samplers and one atmospheric precipitation collector.

2.1.1. Aerosol sampling

The IFJ PAN operates two high volume aerosol samplers, namely the Aerosol Sampling Station–500 and MASS-500. The first one is a standard ASS-500 with the nominal airflow rate of 500 m^3/h, designed and produced by CLOR (Central Laboratory for Radiation Protection, Warsaw). It works within the Polish Monitoring Network of Gamma-Ray Emitters in Ground-Level Air, coordinated by CLOR and Polish National Atomic Agency (PAA). The second one is an older version of ASS-500 high volume sampler, extensively reconstructed and renamed as MASS-500 for Modified ASS-500, and it is IFJ property. All the aerosol samples were collected on Petryanov filters FPP-15-1.5 (polyvinyl chloride) displaying high good aerosol collecting properties (Bysiek et al., 2000).

At the night of March 23, 2011 we received an informal communication that in Scandinavia iodine had been found (Masson et al., 2011), so in the morning of March 24, we changed the filters on MASS-500 and began an emergency mode operation; the filters were thus replaced every day till April 12. Then the sampling was run on a longer base, and ended with a whole week measurements at the end of April. The entire Polish network monitoring system consisting of 12 ASS-500 stations, turned from the week routine cycle to the emergency two-days-cycle on March 25 (Isajenko et al., 2011) and later, from April 1 till the end of the month, all the stations, including Krakow's ASS-500, operated under a half-week-cycle. The measurements using MASS-500 produced 20 samples of aerosols deposited on the air filters from IFJ PAN (Table 1).

2.1.2. Gaseous sampling

The gaseous fraction of the air was trapped in a double cartridge installed additionally inside the inlet of the MASS-500 station, behind the aerosol filter. This resulted in decreasing the flow rate from 500 m^3/h to about 250 m^3/h. The cartridge was tested with a smaller aerosol sampler HVS-30 produced by Atmoservice, Poznań, Poland, in an endocrinological hospital to measure ^{131}I concentration in the air above septic tanks (Mietelski et al., 2005). The cartridge consists of two identical cassettes in a form of rectangular aluminium frames, filled with granular activated carbon impregnated with KI (IBJ-6, mesh size 2 mm, produced by Gryskand, Hajnówka, Poland). The carbon granulate is bordered both at the top and bottom of each cassette with fine stainless steel nets. Between the cassettes a rubber seal is inserted. The efficiency for gaseous iodine adsorption is calculated from the ratio of the activity found in the first and the second cassette under assumption that iodine was absorbed identically in both cassettes. Hence, with an infinite series of such cassettes, the absorbed activity would