

Radioactivity of the atmospheric aerosols measured in Poland following the accident in the Fukushima Dai-ichi nuclear power plant in 2011

Marian Fujak,
Krzysztof Isajenko,
Paweł Lipiński,
Barbara Piotrowska,
Izabela Kwiatkowska

Abstract. The data were collected by the network of the ASS-500 stations, aimed at the detection and monitoring of the presence and activity concentration of radionuclides in the ground-level air in both normal and emergency situation. Results of the monitoring of radionuclide content in the ground-level atmosphere in Poland during the accident in Fukushima Dai-ichi nuclear power plant (FD-NPP) in 2011 are presented. In particular, the results of the measurements cover the period of March–May 2011 focusing on the activity concentration of Cs-137, Cs-136, Cs-134, I-131, I-132 and Te-132. The elevated concentration of radionuclides in the atmosphere was initially detected on 23rd March. The maximum activity concentrations of I-131 – 5.40 mBq/m³ and Cs-137 – 0.73 mBq/m³ were measured on 30th March in Łódź. The ratio of the Cs-134/Cs-137 – 0.82 in Warsaw was calculated for the period 28–30 March. The Fukushima data are compared with the data obtained during the Chernobyl accident and the subsequent period. After the accident in Chernobyl, the maximum measured activity concentration of I-131 reached 180 Bq/m³ on 29th April and Cs-137 – 18 Bq/m³ on 30th April 1986 in Warsaw, however the Cs-134/Cs-137 ratio amounted to 0.49 indicating the presence of the spent fuel of different origin than that of the Fukushima accident. The deposition of the radionuclides from the Fukushima accident does not affect neither Polish people health nor the environment in Poland.

Key words: ASS-500 • CTBTO • Chernobyl accident • Fukushima accident • radioactive aerosols

Introduction

On 11th March 2011, the high magnitude 9.0 earthquake occurred about 130 km off the eastern Pacific coast of Japan's main island Honshu, followed by a large tsunami. These events caused the loss of many lives and huge damage. One of the consequences was the station blackout (total loss of AC electric power) at the Fukushima Dai-ichi nuclear power plant (FD-NPP). The plant, which is operated by the Tokyo Electric Power Company (TEPCO). The station blackout developed into a disaster leaving four of the six FD-NPP units heavily damaged [21]. The radioactivity released into the atmosphere from the damaged reactor cores was dominated by volatile fission products as a result of the containment venting (reducing gaseous pressure), hydrogen explosions and the discharge of coolant water into the sea [3, 5, 6].

The radioactive materials were dispersed mainly in the northern hemisphere. Until early May, the levels of activity concentration remained significant throughout the whole northern hemisphere, but by mid June 2011 at most stations of the International Monitoring System (IMS) of the CTBTO (Comprehensive Nuclear-Test Ban Treaty Organization) network returned to the background level [20].

The emission of radionuclides to the atmosphere started on 12th March to the vicinity of the NPP [4]. The

M. Fujak[✉], K. Isajenko, P. Lipiński, B. Piotrowska,
I. Kwiatkowska
Central Laboratory for Radiological Protection,
7 Konwaliowa Str., 03-194 Warsaw, Poland,
Tel.: +48 22 811 0011 ext. 128, Fax: +48 22 811 1616,
E-mail: fujak@clor.waw.pl

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first detection by the IMS network was on 13th March at Takasaki (about 200 km away from Fukushima). During the following period, 15th March and 24th March, the strong westerly stream blew over the Pacific Ocean from Japan to California, and over the Atlantic Ocean – from North America to Iceland. The flow was then directed towards the North Pole because of a high pressure over Europe [20]. In Europe, the first signs of the releases were detected 7 days later while the first peak of activity level was observed between March 28th and March 30th. A rough estimate of the total I-131 inventory that has passed over Europe during this period was $< 1\%$ of the released amount [17]. The high pressure structure established over the Western Europe then led the flow to the Central Europe. The radionuclides suspended in the atmosphere reached the Eastern Europe before reaching the Western Europe (e.g., 21st and 24th March), because the downdraft was located on the Eastern Europe. The radioactive isotopes released into the atmosphere were rapidly transported around the globe, and achieved the circumnavigation in 2–3 weeks [20]. The IMS station detected releases of I-131, I-133, Te-129m, Te-129, Te-131m, Te-132, Cs-132, Cs-134, Cs-136, Cs-137, less volatile radionuclides such as Zr-95, Nb-95, Ba-140, La-140, Mo-99, and Tc-99m were detected too, but on a lower level. The highest level of I-131 at the IMS station reached 100 Bq/m^3 [20].

The total atmospheric release of Cs-137 and I-131 was estimated by TEPCO at 1.5×10^{16} and 1.6×10^{17} Bq (for aerosols), respectively [3]. During March–April period, about 80% of the total radioisotope mass was estimated to have blown out over the ocean [21]. Atmospheric transport simulations suggest that the main air emissions have occurred during the events of 14th March, 2011 (UTC) and that no major release occurred after 23rd March. The radioactivity emitted into the atmosphere could represent 10% of the Chernobyl accident releases for I-131 and Cs-137 [1]. The measurement data were published by the Ministry of Economy, Trade and Industry in Japan (METI) [3] and showed that the emissions from FD-NPP caused strongly elevated levels of radioactivity in the Fukushima prefecture and other parts of Japan. Deposited Cs-134 and Cs-137 were the main factors influencing a long-term dose [8].

Sampling of aerosols

The aerosol sampling station ASS-500 is used for the routine environmental monitoring of the ground-level atmosphere in Poland and operated since 1992 (first station has worked since the 70s of the last century in Warsaw). The station is a stand-alone, all-weather instrument for continuous collection of air aerosols. The airflow rate higher than $500 \text{ m}^3/\text{h}$ through a chlorinated polyvinylchloride or organic fiber filters allows representative aerosol samples to be collected with volumes of up to about $1.2 \times 10^5 \text{ m}^3$, and enables accurate γ -spectrometric measurements of natural and artificial airborne radionuclides, covering a wide range of activity concentrations with the minimum detectable activity (MDA), starting from $1 \text{ } \mu\text{Bq/m}^3$ for Cs-137. The efficiency of the Petrianov filter type FPP 15–1.5 for aerosols with diameters between 0.3 and $1.25 \text{ } \mu\text{m}$, at linear air

velocities through the filter varying from 0.25 to 4 m/s with pressure drop through the filter Δp 0.5–9.3 kPa, is between 96 and 99%. In normal radiological situation the recommended sampling period is one week. The volume of the air depends on the aerodynamic burden of the filter with the aerosols collected on it. This volume ranges from 5×10^4 to $1.2 \times 10^5 \text{ m}^3$. In the emergency situation or for check-up reasons the period of the exchange of the filters can be increased as needed (down to several minutes when severe contamination is present/expected). All stations are equipped with NaI(Tl) detectors placed above the filter allowing on-line radioactivity control. The ASS-500 aerosol sampling stations are located in 12 different points of Poland [9].

The measurement of the activity concentration of the radionuclides deposited on the filter, (which was pressed to the proper geometry), is performed at a low-background laboratory by means of an high purity germanium (HPGe) spectrometer of 40% relative efficiency. The efficiency calibration of the detector is performed using a volume standard mix- γ source containing several isotopes emitting gamma radiation in the range 59–1836 keV. The standard source is a cylinder of dimensions: 2" diameter and 1/8" thickness. In order to minimize outside gamma background the detector should be placed in a shielding house made of 100 mm lead with two inner layers of 2 mm Cu and 1 mm Cd [9].

The results and discussion

Around 23rd March the air masses from the Fukushima NPP reached over Poland. The eight of the twelve ASS-500 stations exchanged filters twice a week during the period 21rd to 28rd March. The filters were exchanged for the first time on Friday (25th March) and immediately after the replacement the filters were pressed and measured. The relatively high concentrations of artificial radionuclides (fission or activation products) – especially I-131, Cs-134, Cs-137, I-132, Te-132 – were detected and the highest activity concentration of I-131 was measured in southern (Katowice – $86.6 \pm 3.6 \text{ } \mu\text{Bq/m}^3$) and central Poland (Warsaw – $83.9 \pm 2.2 \text{ } \mu\text{Bq/m}^3$), the lowest in northern (Gdynia – $52.9 \pm 2.6 \text{ } \mu\text{Bq/m}^3$) and east-southern Poland (Sanok – $53.5 \pm 2.2 \text{ } \mu\text{Bq/m}^3$). Eight stations exchanged the filters three times during the period 28th March to 4th April, and the highest value of the radionuclide activity concentration was for the period 28–30 March. All of the stations found the following isotopes: I-131, I-132, Cs-134, Cs-136, Cs-137, Te-132, and some of the stations detected Te-129m, La-140, Nb-95 as well. The paper presents the data obtained from four stations (Gdynia, Warsaw, Łódź and Katowice), which are representative for the whole territory of Poland.

The iodine I-131 was present on the territory of Poland since 25th March to 23th April 2011 (Katowice). The highest concentration reached 5.4 mBq/m^3 as observed on 30th March in Łódź. The concentration of radioactive iodine I-131 at other sites of Poland was smaller (Katowice – 4.4 mBq/m^3 , Warsaw – 3.2 mBq/m^3) (Fig. 1, Table 1) [10]. Figure 2 shows the average weekly activity concentration of the I-131 during the period 28th March to 4th April 2011 measured at twelve ASS-500

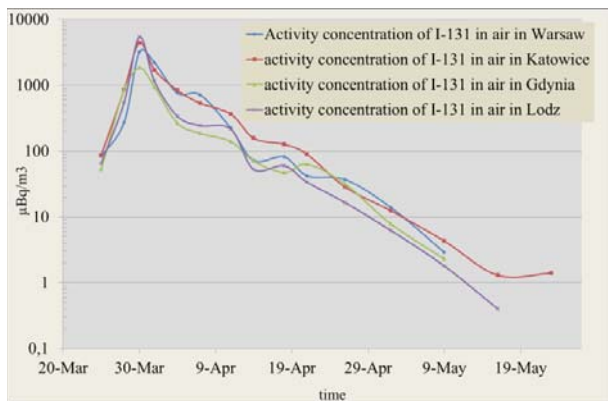


Fig. 1. The activity concentration of I-131 in the aerosols in Poland during the Fukushima accident.

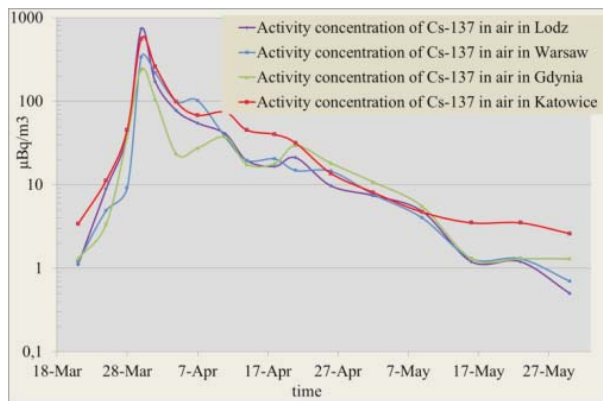


Fig. 3. The activity concentration of Cs-137 in the aerosols in Poland during the Fukushima accident.

stations in Poland. The maximum weekly average value is present in the central regions of Poland.

The maximum activity concentration of the Cs-137 – 0.73 mBq/m³, as shown in Fig. 3 and Table 2 was measured on 30th March in Łódź. The radiocesium is bound mainly to the inorganic accumulation mode aerosols fraction e.g. ammonium, sulphate and nitrate [11, 15]. Significant release occurred first time

on 14th March, the Cs-137 was attached to the aerosols of diameter 0.1–2.0 µm, which is proven by the simultaneous measurements of the sulphate aerosols [12, 15]. The sulphate aerosols grow by coagulation with other particles during the transport and are removed by wet and dry deposition [11, 15].

Figure 4 indicates the Cs-134/Cs-137 ratio being in the range 0.6–0.8 during March–April. This differs

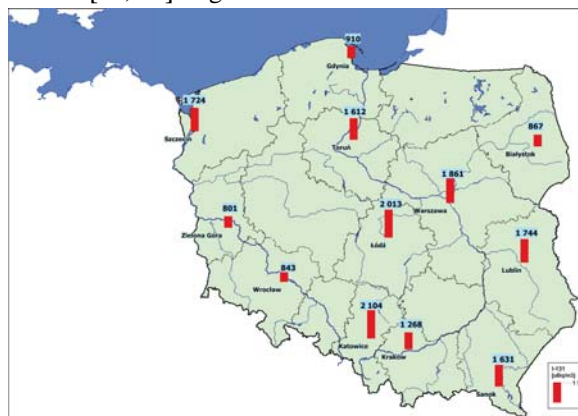


Fig. 2. The weekly average activity concentration of I-131 in aerosols measured at 12 stations ASS-500 during the period 28 March – 4 April 2011 in Poland.

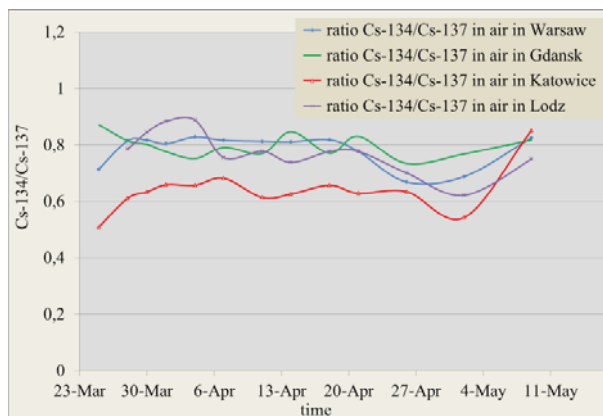


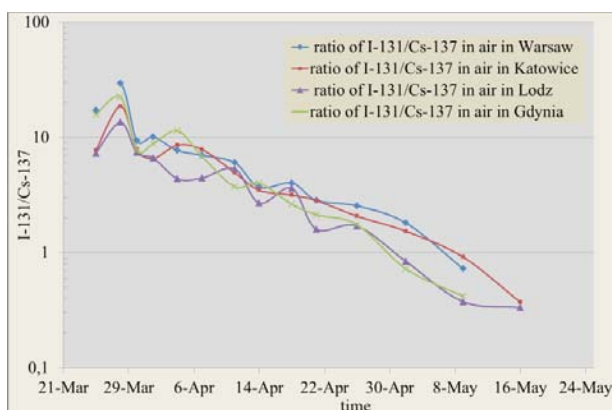
Fig. 4. The ratio of Cs-134/Cs-137 in Poland during the Fukushima accident.

Table 1. Activity concentration of ¹³¹I in air during March–April 2011 in Poland

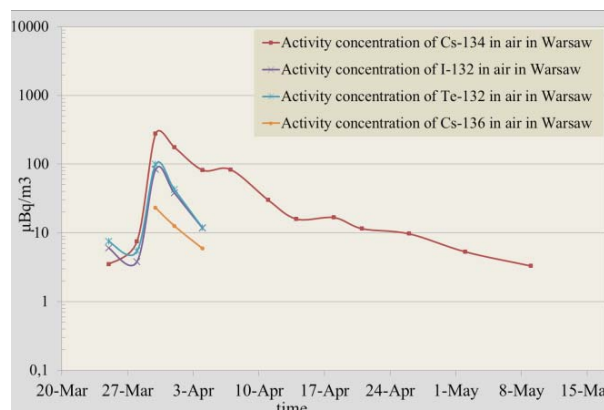
Date of deposition	Activity concentration of ¹³¹ I in air in Poland (µBq/m ³)			
	Gdynia	Warsaw	Łódź	Katowice
21 March	< 0.6	0.7 ± 0.2	< 0.22	< 0.57
25 March	52.9 ± 2.6	83.9 ± 2.0	65.1 ± 3.2	86.3 ± 3.6
28 March	845.2 ± 20.6	271.3 ± 5.5	542.6 ± 9.1	847.3 ± 43.6
30 March	1849.8 ± 40.9	3173.0 ± 57.5	5400.0 ± 95.0	4400.0 ± 226.1
1 April	939.4 ± 25.0	2196.9 ± 40.2	1139.0 ± 19.0	1709.6 ± 87.9
4 April	265.5 ± 9.7	763.1 ± 14.2	338.2 ± 5.6	837.9 ± 43.1
7 April	186.3 ± 4.1	712.6 ± 13.4	243.6 ± 5.1	532.7 ± 27.5
11 April	139.7 ± 6.2	222.5 ± 4.4	215.6 ± 3.7	365.6 ± 18.8
14 April	70.5 ± 3.9	72.1 ± 1.7	52.5 ± 1.0	157.1 ± 8.1
18 April	47.1 ± 2.6	81.9 ± 1.9	60.0 ± 2.4	126.3 ± 6.6
21 April	630.2 ± 3.3	42.2 ± 1.5	33.7 ± 0.7	89.1 ± 4.6
26 April	31.6 ± 1.8	36.9 ± 1.1	16.5 ± 0.5	28.4 ± 1.6
2 April	7.8 ± 0.7	13.9 ± 0.6	6.2 ± 0.3	12.4 ± 0.8
9 April	2.3 ± 0.3	2.9 ± 0.2	1.8 ± 0.1	4.3 ± 0.5
16 April	< 0.7	< 0.75	0.4 ± 0.1	1.3 ± 0.3
23 April	< 0.7	< 0.57	< 0.19	1.4 ± 0.3
30 April	< 0.6	1.7 ± 0.2	< 0.19	< 0.36

Table 2. Activity concentration of ^{137}Cs in air during March–April 2011 in Poland

Date of deposition	Activity concentration of ^{137}Cs in air in Poland ($\mu\text{Bq}/\text{m}^3$)			
	Gdynia	Warsaw	Łódź	Katowice
21 March	1.3 ± 0.2	1.2 ± 0.2	1.1 ± 0.1	3.4 ± 0.4
25 March	2.9 ± 0.2	4.9 ± 0.4	8.9 ± 2.8	11.2 ± 0.8
28 March	37.8 ± 1.2	9.2 ± 0.5	40.1 ± 0.7	45.5 ± 1.1
30 March	240.3 ± 5.6	339.4 ± 8.6	732.0 ± 12.5	559.1 ± 7.5
1 April	106.4 ± 2.9	217.8 ± 5.7	171.6 ± 2.2	260.5 ± 3.8
4 April	23.4 ± 0.9	99.0 ± 2.6	77.2 ± 1.0	97.8 ± 1.6
7 April	27.3 ± 0.5	102.0 ± 2.7	54.8 ± 1.5	68.0 ± 1.2
11 April	37.4 ± 0.9	36.9 ± 1.0	40.5 ± 0.6	74.0 ± 1.3
14 April	17.6 ± 0.7	19.6 ± 0.4	19.5 ± 0.4	45.2 ± 0.8
18 April	17.9 ± 0.6	20.4 ± 0.7	16.6 ± 0.4	40.0 ± 0.8
21 April	29.5 ± 1.0	14.8 ± 0.5	21.1 ± 0.5	31.5 ± 0.6
26 April	18.1 ± 0.6	14.5 ± 0.6	9.7 ± 0.3	13.7 ± 0.5
2 April	10.8 ± 0.4	7.7 ± 0.4	7.4 ± 0.2	8.1 ± 0.4
9 April	5.5 ± 0.3	4.0 ± 0.2	4.8 ± 0.1	4.7 ± 0.3
16 April	1.3 ± 0.2	1.3 ± 0.2	1.2 ± 0.1	3.5 ± 0.2
23 April	1.3 ± 0.2	1.3 ± 0.2	1.2 ± 0.2	3.5 ± 0.3
30 April	1.3 ± 0.2	0.7 ± 0.2	0.5 ± 0.1	2.6 ± 0.2

**Fig. 5.** The ratio of I-131/Cs-137 in Poland during the Fukushima accident.

from the ratios reported by UNSCEAR 2000 during Chernobyl, which were in the range 0.50–0.63 [22]. The relatively small fluctuation of the Cs-134/Cs-137 ratio suggests that the radioactive material was predominantly released from the same type of source material.

**Fig. 6.** The activity concentration of Cs-134, I-132, Te-132 and Cs-136 in the ground-level atmosphere in Warsaw during the Fukushima accident.

The decrease of the I-131/Cs-137 ratio (Fig. 5) is due mainly to the decay of the short-lived I-131 isotope. Kristiansen *et al.* [15] calculated the removal times from the air for the Cs-137 to be 10–13.9 days and for the

Table 3. Activity concentration of ^{134}Cs in air during March–April 2011 in Poland

Date of deposition	Activity concentration of ^{134}Cs in air in Poland ($\mu\text{Bq}/\text{m}^3$)			
	Gdynia	Warsaw	Łódź	Katowice
25 March	3.4 ± 0.3	3.5 ± 0.2	–	5.7 ± 0.3
28 March	30.8 ± 0.7	7.5 ± 0.3	31.5 ± 0.6	27.8 ± 0.5
30 March	192.9 ± 2.3	277.4 ± 5.3	618.5 ± 6.5	354.4 ± 2.8
1 April	82.6 ± 1.8	175.3 ± 3.4	151.7 ± 1.7	171.7 ± 1.7
4 April	17.6 ± 0.5	82.0 ± 1.6	68.6 ± 0.8	64.3 ± 0.9
7 April	21.6 ± 0.4	83.3 ± 1.6	41.4 ± 0.8	46.4 ± 0.6
11 April	28.8 ± 0.5	30.0 ± 0.6	31.5 ± 0.5	45.4 ± 0.6
14 April	14.9 ± 0.5	15.9 ± 0.3	14.4 ± 0.3	28.3 ± 0.4
18 April	13.8 ± 0.4	16.7 ± 0.4	12.9 ± 0.3	26.3 ± 0.5
21 April	24.5 ± 0.6	11.5 ± 0.3	16.4 ± 0.4	19.8 ± 0.3
26 April	13.3 ± 0.3	9.7 ± 0.3	6.8 ± 0.2	8.7 ± 0.3
2 April	8.3 ± 0.3	5.3 ± 0.2	4.6 ± 0.2	4.4 ± 0.2
9 April	4.5 ± 0.2	3.3 ± 0.1	3.6 ± 0.1	4.0 ± 0.1
16 April	< 0.41	< 0.57	0.3 ± 0.1	1.0 ± 0.1
23 April	< 0.38	< 0.30	< 0.12	< 0.19
30 April	< 0.31	< 0.44	< 0.07	< 0.17

Table 4. Ratios of the radionuclides detected in Warsaw during the period 28–30 March 2011

Ratio	I-131/Cs-137	I-132/Cs-137	Cs-134/Cs-137	Cs-136/Cs-137	Te-132/Cs-137
Value	9.32	0.24	0.82	0.067	0.29

I-131 of 17.1–24.2 days during April and May 2011. The longer removal time for I-131 than for Cs-137 is expected, because I-131 has a large gaseous fraction, and the gas to particle conversion leads to the formation of new particulate iodine from gaseous iodine during the transport. The gas-to-particle conversion for I-131 typically occurs on time scales of about 2–3 weeks [15, 17].

Figure 6 and Table 3 presents the results of the measurements of the activity concentration of Cs-134, I-132, Te-132 and Cs-136 in the ground-level atmosphere in Warsaw during the March–May 2011 period. The maximum activity concentrations of radionuclides were detected on 30th March – similar to other countries of Europe [17]. The measurement values decreased mainly due to the decay of short-lived isotopes.

The knowledge of the activity ratio of isotopes of different chemical elements is useful to derive information on the accident mechanisms at the time of the releases. The Cs-134/Cs-137 ratio characterizes the level on burn-up of fuel [13]. The variation in the Cs-136/Cs-137 ratio may indicate that the source material included a mixture of different irradiated batches of fuel, however, the volatility ratio Cs-136/Cs-137 may be due to underestimation of uncertainty Cs-136. The Te-132/Cs-137 ratio provides the information on the behaviour of the metallic elements, but it is dominated by decay of the short-lived radionuclide. The change of ratio I-131/Cs-137 can indicate time and condition of events [2]. Table 4 presents the ratios of the radionuclides I-131/Cs-137, I-132/Cs-137, Cs-134/Cs-137, Cs-136/Cs-137 and Te-132/Cs-137 in the air measured in Warsaw during the period 28–30 March.

Currently, the Cs-137 is detected by the ASS-500 network with concentrations close to the detection limit of about $1 \mu\text{Bq}/\text{m}^3$. At present, the detection of Cs-137 is due to the re-suspension of soil particles containing the traces from former depositions (fallout from nuclear weapon tests and from the Chernobyl and Fukushima accidents).

Comparison – Fukushima and Chernobyl

The temporal variations of artificial radionuclides such as Cs-137 in the ground-level atmospheric aerosols observed in Poland before the Chernobyl accident were due to the global fallout. The concentration of Cs-137 in the atmosphere showed a decreasing trend since 1963, when the maximum value was observed. The second maximum of Cs-137 activity in the ground-level air aerosols occurred in 1986, just after the Chernobyl accident [14], when about 85 PBq of this isotope was released to the atmosphere. The Chernobyl reactor accident involved both nuclear fuel melting and graphite fire [22]. The long-term radiological impact of the Chernobyl accident on the environment and humans due to released radionuclides, migration, re-suspension of deposited radionuclides has been studied over

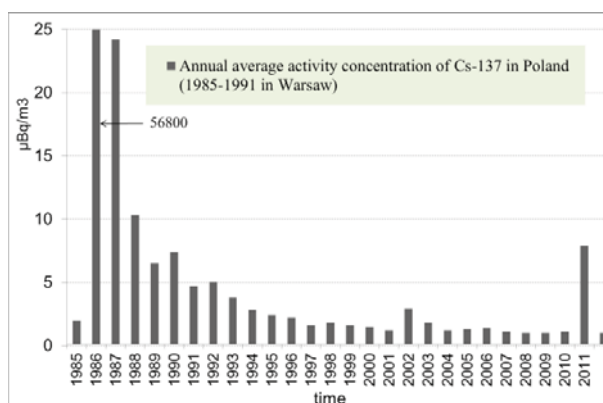


Fig. 7. The average annual activity concentration of Cs-137 in Poland in air 1985–2012 (the measurement performed by the Central Laboratory for Radiological Protection).

25 years [16]. The Chernobyl accident resulted in the contamination of large areas of Europe. Figure 7 presents the annual average of the activity concentration of the Cs-137 in Poland in the years 1985–2012 measured by the Central Laboratory for Radiological Protection (CLOR, Warsaw). The Chernobyl accident data come from the only working stationary ASS-500 station located in Warsaw. Nineteen measurements of air aerosols were carried out in Warsaw during the period since 28th to 30th April 1986 (with the air volumes from 50 to 2010 m³). The maximum measured activity concentration of I-131 was 186 Bq/m³ (29th April), Te-132 – 165 Bq/m³ (29 April), Cs-137 – 18.6 Bq/m³ (30th April). Figure 8 shows the percentage distribution of radionuclides in air in Poland during the Chernobyl accident estimated at the Central Laboratory for Radiological Protection after 1986. The contamination of the atmosphere by the radionuclides released from the Chernobyl NPP depended mostly upon the half-lives of radionuclides and their volatility [23]. The measurements of radionuclides in Poland performed by the Central Laboratory for Radiological Protection during the accidents in Chernobyl and Fukushima are included in Table 5. The calculated I-131/Cs-137 ratio (for three days period in Warsaw) from the Chernobyl accident was higher (at the first phase of deposition) than from Fukushima

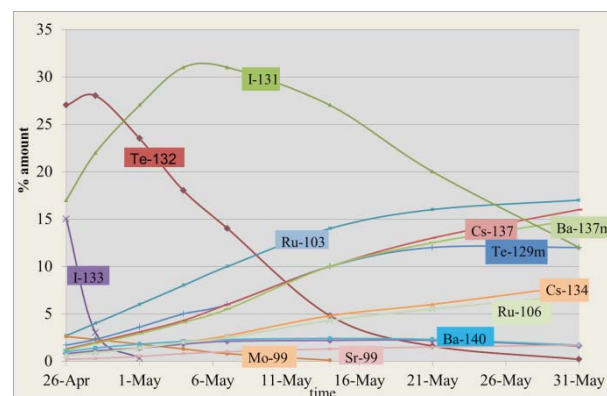


Fig. 8. The amount of radionuclides (in %) in air in Poland during the Chernobyl accident [19].

Table 5. Chernobyl vs. Fukushima in Warsaw (measurements performed by the Central Laboratory for Radiological Protection)

Radionuclide	Average value (mBq/m ³)	
	Chernobyl 28–30 April 1986	Fukushima 28–30 March 2011
¹³¹ I	68 700	3.17
¹³⁷ Cs	6 570	0.34
¹³⁴ Cs	3 230	0.28
¹³² Te	69 730	0.10
Less volatile isotopes:		
⁹⁵ Zr	240	not detected
¹⁰³ Ru	12 700	not detected

(Table 5). This can be the result of the various distances (decay of the iodine) and the changes of the gas-aerosols fraction. However, the ratio of the I-131/Cs-137 during the accident decreases with increasing of the level of the Cs-137 ground deposition and it can indicate the time of event (Fig. 8).

The consequences of the Fukushima accident were estimated to be close to that of Chernobyl then both are assigned to the level 7 on the international nuclear and radiobiological event scale (INES) [7]. The main difference between the release characteristics of the Fukushima and Chernobyl accidents is the extent of the release and non-significant presence of the less volatile elements (Zr, Sr, Ru, Mo and Tc) in the case of Fukushima releases [3]. The released fraction from Fukushima included a large amount of elements, otherwise than the Chernobyl fallout, where quite high activity concentration of non-volatile elements was present and “hot particles” in the environment were found (tiny fragments of the of fuel rods) [18, 22]. This indicates that the fuel temperature remained well below 2400°C during Fukushima accident. At such temperature, the release of refractory elements (e.g. Zr, Pu, Ru, Mo) is still limited to the few percent compared to those of the caesium [13, 19].

Conclusion

The emission of radionuclides during the Fukushima accident was high enough to be detected at every of the ASS-500 station in Poland. The concentration of radionuclides in the ground-level atmosphere was effectively diluted due to the decay of short-lived isotopes, dispersion and wet and dry deposition, and no longer posed a radiological risk to humans when the air masses reached Poland. In the period March–May 2011 mainly volatile radionuclides were detected. In June 2011, the activity concentrations of artificial radionuclides from Fukushima at all ASS-500 stations fell down to the background level.

The maximum daily measured concentration of the I-131 in the ground level air in Warsaw after the Fukushima accident was more than 30 000 times lower than during the Chernobyl accident.

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