Atmospheric Radioactivity in Bulgaria and Finland Following the Fukushima Nuclear Accident

Jussi Paatero[1] , Blagorodka Veleva[2], Juha Hatakka[1] and Elena Hristova[2]
1. Finnish Meteorological Institute, Helsinki, Finland
2. National Institute of Meteorology and Hydrology, Sofia, Bulgaria

Jussi.Paatero@fmi.fi

Abstract

A huge earthquake with a magnitude of 9.0 occurred east of the Honshu Island, Japan on 11 March 2011 05:46 UTC. The earthquake and especially the resulting tsunami wave caused a loss of thousands of human lives and massive material damages in Japan. One of the worst hit regions was the coast of the Fukushima prefecture. The nuclear power plant in the area, Fukushima Dai-ichi, was hit by the tsunami wave leading to the loss of cooling of reactors and pools of spent nuclear fuel. This caused releases of radioactivity to both the atmosphere and the ocean. There is an extended experience in atmospheric radioactivity monitoring and studies in FMI-Finland and NIMH-Bulgaria since the period of global nuclear fallout. The knowledge of the features of global atmospheric circulation suggested that the air masses, *contaminated with fission and activation products, travel in eastern direction due to the prevailing westerly winds in the northern temperate regions. Eventually, when they reach Europe significant dilution due to diffusion and removal processes occur.*

In northern Finland the atmospheric emissions from Fukushima were first observed from an aerosol sample collected between 21 March 06 UTC and 22 March 06 UTC, and from a sample collected a day later in Helsinki, southern Finland. The samples were measured for total beta activity and also with semiconductor gamma spectrometry. Several fission products were identified from the gamma spectra, for example iodine-131 and cesium isotopes 134 and 137. In Bulgaria the first traces of Fukushima emissions were detected on the peak Mussala on the aerosol sample from March 23 (http://beo-db.inrne.bas.bg/moussala). The concentrations of I-131 above detection limits were reported in Sofia first on 25th of March by National center for Radiation Protection and Radiobiology (NCRPR). Measurements of total beta radioactivity of aerosol show no distinguished increase during the period of direct influence of the tagged air masses. The fission products I-131, Cs-137 and in some samples Cs-134 were detected in weekly precipitation samples in Sofia, Burgas and some other stations in Northern Bulgaria during the periods 24-30.03.2011; 31.03-6.04.2011; 7.04-13.04. The total deposition of I-131 in Sofia, NIMH were for example 2.12±0.14; 1.20±0.46; 1.00±0.18 Bq.m^{-2}week^{-1} during the mentioned periods.

In Europe the radioactive plume from the Fukushima nuclear power plant did not pose a radiological risk to humans or the environment. The measured signal of I-131 and Cs-137 over the northern hemisphere in March and April 2011 might be useful further in diffusion parameterization and in deposition processes studies.

Keywords: *Fukushima nuclear accident, fission products, aerosols, deposition*

Introduction

The human being and the biosphere have lived with the environmental radioactivity all over the times/epochs since the earth exists. More of that, the natural radioactivity in the past was greater because the activity of the primordial radionuclides and their daughters from the decay chains have decreased due to radioactive decay.

Since the discovery of the artificial radioactivity in 1934 by Irene and Frederic Joliot-Curie, the start of operation of the first nuclear reactor in Chicago, 1942 and first nuclear explosion, 1945 a great number more than 2500 of man-made radionuclides have been produced, between them with a half-life time $T_{1/2}$ > 1 hour – more than 550 radionuclides. Parts of the man-made radionuclides were dispersed in the environment. The main source of artificial radioactive contamination of the atmosphere remains

the radioactive fission and fusion product from the nuclear tests in the atmosphere. There were altogether 543 tests. Comparative assessment of the airborne radioactivity in Bulgaria and Finland, the global bomb fallout variations and Chernobyl accident contamination is previously discussed in (1,2). Nowadays the atmospheric radioactivity is dominated by the radon emanation from the earth crust (soil, rock) and radon short lived daughters, usually attached to the aerosol particulates.

The nuclear power reactors are the dominant producers of the man-made radioactivity and therefore the nuclear safety and radiation protection of the men and environment are of great concern. In normal operation conditions the emissions through ventilation and stacks are limited by regulations in order not to exceed a limit, 1mSv.y-1 in Bulgaria, for members of public from all artificial radioactive sources and pathways (Law on the safe use of nuclear energy, LSUNE).

The production of radioactivity is a function of the energy produced in the reactor, which is the product of the reactor power output and operating time. A typical operating 1000 MWe light water reactor (LWR) will contain about 5.55 x 10^{20} Bq of activity, mostly short lived fission gases during operation; the activity will decrease to a fraction within few seconds after shutdown. In the Table 1 are summarized the radioactive releases as a result of the major radiation accidents following (3-9).

Table 1. Chronology of the accidents with off-site releases of radionuclides to the environment. The releases of Radioactive Noble Gases (RNG), ^{131}I and ^{137}Cs are given in PBq (1.10^{15} Bq)

	Date	RNG PBq	I-131 PBq	Cs-137 PBq	Others
Chelyabinsk-40 plutonium plant, Kyshtym, USSR*	September 1957			3.6 E-03	100 PBq long lived beta radionuclides 5.4 PBq ^{90}Sr+^{90}Y
Windscale accident, Northern England *	October 1957	14 ^{133}Xe	0.74-1.2	0.046	0.2 TBq ^{90}Sr 1.6 TBq ^{239}Pu
SL-1 Reactor, Idaho falls, USA *	January 1961		2.6E-03		
Rocky Flats plant, Denver, USA	1969				0.22 TBq Pu
Three Mile Island, Unit 2, USA	March 1979	77.7	5.55E-04		
Chernobyl NPP accident, former USSR	26-April 1986	6533 ^{133}Xe+^{85}Kr	1760	86	
Tokaimura nuclear fuel processing plant, Japan	30.09. 1999				~200 persons up to 21 mGy - n, γ
Fukushima, Dai -ichi NPP, Japan	March 2011	~1000	150	15	

*the accidents at weapon production facilities

The values in the table above might be compared to the global release of ^{131}I and ^{137}Cs from atmospheric nuclear weapon testing, 675 000 PBq and 948 PBq, respectively (10).

The evaluation of the the International Nuclear Event Scale for prompt communication of safety significance describes the highest level 7 as MAJOR ACCIDENT, (www.iaea.org):“External release of a large fraction of the radioactive material in a large facility (e.g. the core of a power reactor). This would typically involve mixture of short and long lived radioactive fission products (in quantities radiologically equivalent to **more than tens of thousands of terabecquerels, or > ~10^{16} Bq, of ^{131}I**). Such a release would result in the possibility of acute health effects; delayed health effects over a wide area, possibly involving more than one country; long term environmental consequences”. Till

March 2011 the only example of such an accident given was that at Chernobyl nuclear power plant, USSR, 1986. On 27 April TEPCO provided an update of the estimated percentage of core damage for **Units 1, 2 and 3** following an assessment (the values assessed previously on 15 March are given in parentheses): **Unit 1**: 55% core damage (70%); **Unit 2**: 35% core damage (30%); **Unit 3**: 30% core damage (25%). *(IAEA Briefing on Fukushima Nuclear Accident (2 May 2011, 19:50 UTC.*

The emissions of radioactive material to the atmosphere from Fukushima Dai-ichi reactors started on March 12 with the hydrogen explosion on Unit 1, due to the loss of reactor cooling. On March 13 and 14 explosions occurred on Unit 2 and Unit 3. The emissions consist mainly of RNG and volatile isotopes of Iodine, Cesium, Tellurium. Some limited emissions continued during April This way a complex and prolonged source term was formed. The contaminated air masses crossed the Pacific ocean, North America, Atlantic and reached Europe. A comprehensive investigation of the passage of the airborne radionuclides from Fukushima Dai-ichi nuclear reactors over Europe is presented by (11). The whole Northern hemisphere was affected within 4 weeks after the accident and after April 13 the radioactive traces were detected in South-East Pasific region – in CTBTO International Monitoring Stations (IMS) in Australia, Malaysia, Fiji and Papua New Guinea (12). The trace quantities of ^{131}I and ^{137}Cs and ^{134} Cs were also measurable (above Minimum Detectable Activity, MDA) in air particulate: in Finland since March 21 and in Bulgaria since March 23, first in peak Mussala (J. Penev, personal communication) and since March 25 in Sofia (13).

Materials and Methods (Experimental)

Air particulate sampling, Bulgaria

In Sofia, the NIMH started the measurements of atmospheric radioactivity in 1958. The aerosol total beta activity is measured immediately after sampling, three days (72h) and five days (120) after the end of sampling. The aerosol samples are collected on filters (Synpor, FPP-15) by pumps at 2 m high above a grass surface. The air volume is measured with a flow meter. The samples are changed every day at 6:00 UTC (8:00 Local Standard Time). The air volume of the samples is about 100m^3. During the years the monitoring station network of NIMH was developed and since 1969 a network of 5 daily aerosol stations has been operated and 4 radiometric laboratories were set in Plovdiv, Burgas, Varna and Pleven. The radiation detectors used since 1980's are plastic scintillator detectors, the same type beta radiometers are in operational use in 5 NIMH laboratories. Mainly due to the economical restrictions since 2009 the sampling during the weekends and holidays was stopped.

Atmospheric deposition sampling, Bulgaria

The 24 hours precipitation, collected in cylindrical container with the surface area of 0.2-1.0 m^2 is also sampled at 6:00 GMT. The container is washed every day (every working day since 2009) with distilled water to avoid influence of dry deposition in dry periods. The aliquot of 0.250l is evaporating slowly in glass beaker to few ml and then transferred to aluminum plate and evaporated to dry residue. Few ml of distilled water are used to wash the beaker, added and evaporated in Al plate. The source is measured usually 3-4 hours after sampling for short lived beta and after 120 hours for man made beta emitters and lead-210. The uncertainty due to the radiometry is estimated to be in the range of 6-7% for periods with higher concentrations and up to 50% in case of low concentrations.

The total (wet+dry) weekly and monthly fallout is collected in cylindrical containers at the height of 1m above ground with the vessel bottom covered with distilled water. The sampling of "weekly fallout" is done every Thursday and of monthly fallout - on every 1st date of the next month. The process of the liquid sample is the same as for the daily precipitation water samples.

In addition the network for deposition on the planchet samplers, S=0.3m^2, covered with cotton material was in use in Bulgaria (14). Because of the ashing of the sample its efficiency for volatile elements is limited and practically no iodine isotopes remain, but cesium-134 and cesium-137 were detected, as well as natural berilium-7.

Total beta and gamma-spectrometry measurements, March-May, 2011, Bulgaria

All the filters are measured for total beta activity: on the 5th and 60th minute after sampling, when the beta activity is mainly due to short lived ^{222}Rn daughters, and on 72h and 120h after the end of sampling. Composed samples of several days' filters or for the whole month are measured by gamma spectrometry in several low level laboratories, depending of the place of sampling and samples

processing. The High Purity Germanium detectors (HPGe) with relative efficiency from 17 to 50% and passive shielding are used in all laboratories, listed below: The gamma-spectrometric laboratories were calibrated in the frame of Bulgarian national calibration campaigns with the same geometry and density volume sources at least once.

List of Bulgaria Laboratories:

Laboratory of Radioanalitical Methods and Lab2, INRNE-BAS - Sofia;

Regional Laboratory of Ministry of Environment and Waters (MOeW) – Burgas

Regional Laboratory of NIMH – Pleven

Gamma-spectrometry laboratory of EAEW, MOeW – Sofia.

The wet deposition samples were divided in 2 parts: 1) aliquots of 0.25-0.5l for total beta measurement and 2) the rest quantity of the water was slowly pre-concentrated by evaporation to less volume, and transfer in the specific container geometry, different from lab to lab.

Aerosol sampling and analysis in Finland

The FMI has been collecting daily aerosol samples onto glass-fibre filters in Helsinki, southern Finland and at Sodankylä, northern Finland since the 1960s. At first all the filters were measured in the FMI's laboratory with GM counters, but since 1982 two successive automatic alpha/beta analyzers have been used (15). The detector arrangement used in the analyzers consists of five large-area (600 cm^2) gas-flow proportional counters. The total alpha and beta activities of the samples are measured five days after the end of sampling, when the short-lived ^{222}Rn progeny have decayed into ^{210}Pb and the ^{220}Rn progeny have decayed into stable lead. The measured total beta activity consists mainly of ^{210}Pb/^{210}Bi and possible artificial beta emitters while the measured total alpha activity consists mainly of ^{210}Po. Some of the filter samples are measured also with HPGe gamma spectrometry (16).

Results and discussion

Particulate airborne radioactivity

The sampling for the total beta radioactivity in NIMH is done by using low volume samplers. Therefore the combined filter samples were measured by gamma spectrometry. It has to be pointed out that the concentrations of ^{131}I were low and some of the samples were measured several half-live times after the sampling mainly because available apparatus time was limited and the gamma spectrometric laboratories had their own measurements program. Because of the delayed measurements high uncertainty and high MDA are observed for some samples.

Table 2a. Results of gamma spectrometric measurements of combined filter samples Sofia-I (25,28,29,30, 31.03, 1.04.), and Sofia-II (4-8.04.), end of March and the beginning of April 2011. The measurements were performed in LRAM, INRNE. The DL for ^{137}Cs in this Lab is relatively higher.

Sample	Volume m^3	Date of meas.	Period of sampling	^{7}Be $mBq.m^{-3}$	± 1σ	^{137}Cs $mBq.m^{-3}$	^{131}I $mBq.m^{-3}$	± 1σ
Sofia-I	427.1	30.4.2011	25.03-1.04.2011	2.15	±0.21	≤0.052	0.616	±0.25
Sofia-II	431.5	29.4.2011	04.04-.8.04	5.42	±0.28	≤0.052	0.735	±0.15

Table 2b. Gamma emitting radionuclides in combined semi-monthly and monthly filter samples.

Sample	Volume m^3	Date of meas.	^{7}Be, $mBq.m^{-3}$	± 1σ	^{137}Cs, mBq.m-3	± 1σ	^{134}Cs, $mBq.m^{-3}$	± 1σ
Sofia-March	1884	March2011	4.45	± 0.11	0.015	± 0.005	0.006	± 0.0025
Sofia-April-I	898.2	1-14 April	5.21	± 0.15	0.087	± 0.011	0.071	± 0.007
Sofia-April-II	1251.6	15-30 April	5.93	± 0.09	0.014	± 0.007	0.018	± 0.004
Sofia-May	1789	May 2011	5.6	± 1.68	0.015	± 0.007	DL	
Sofia-June	1919.2	June 2011	5.73	± 1.07	0.014	± 0.008	DL	
Pleven April**	247.2	April 2011	4.52	± 0.23	0.069	± 0.02	0.06	± 0.017
Pleven-May	298	2-31.05	5.03	± 0.46	0.035	± 0.017	0.027	± 0.025

Because the monthly and semi-monthly samples were measured later, more than 3 iodine-131 half-live times, ^{131}I was detected only in sample "Pleven April**" as 0.23±0.15 $mBq.m^{-3}$. The method and equipment in use in Lab.2 of INRNE for measurement of the aerosol radioactivity, where NIMH samples from Sofia were measured is described in details in (17).

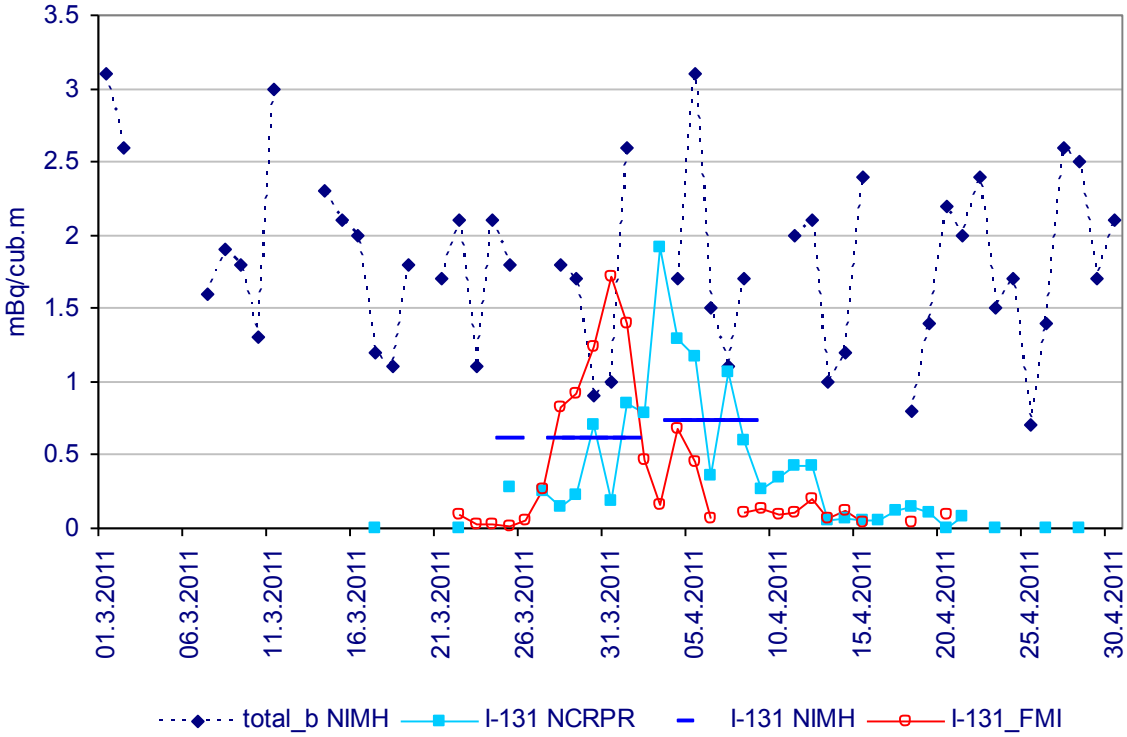


Figure 1. ^{131}I measurements in Helsinki and Sofia, NIMH, NCRPR, FMI results. Long lived total beta activity in NIMH-Sofia is given for comparison.

The natural variations in the total beta concentration in the surface air layer in Sofia is high, within factor of about 5, depending where the sampling station is influence by continental or marine air masses or whether there is penetration from high altitudes or stratospheric air masses. The observed levels of the newly emitted fission products from Fukushima was low and did not affect significantly monthly mean values in Bulgaria in contrast to Finland.

When compare the time variations of ^{131}I concentration it is clear that the signal arrived 2-3 days earlier in Helsinki, Finland then in Sofia, Bulgaria. Maximum of ^{131}I concentration of 1.72 mBq.m^{-3} was measured in Helsinki in the sample of 31.03-1.04.2011. In Bulgaria the maximum reported value of 1.92 mBq.m^{-3} is for April 3. The average value in NCRRP station, Sofia is 0.90 mBq.m^{-3} for the period April 4-8 and is not far from 0.735 mBq.m^{-3}, measured on combined filter sample from NIMH, Sofia. The distance between 2 sampling sides is about 8 km. NCRRP station is closer to the city center where air particulate concentration is usually higher.

In Athens, Greece the maximum air concentration of ^{131}I, on April 6 was 0.490±0.035 mBq.m^{-3} (18) i.e. lower than registered in Sofia, in Vilnius, Lithuania concentrations of ^{131}I ranged from 0.012 Bq.m^{-3} to 3.7 mBq.m^{-3}(19).

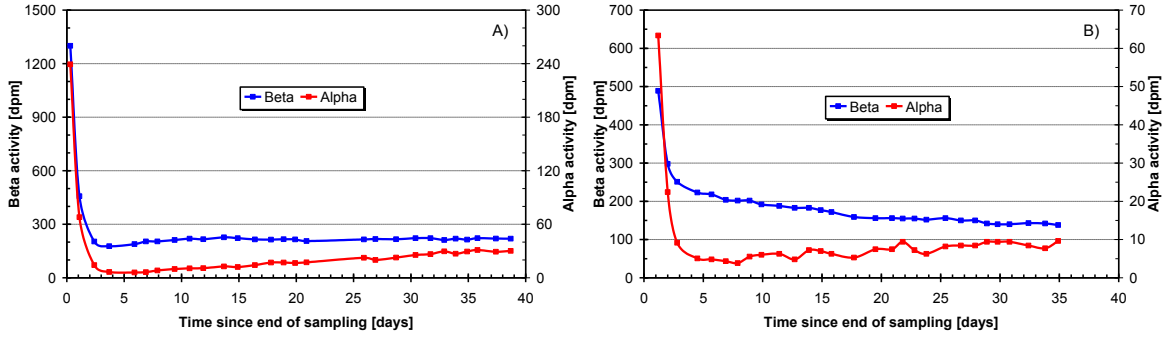
Total beta activity and iodine-131 in Finland

The fresh fission product nuclides from the Fukushima nuclear accident caused a clear change in the temporal behaviour of total beta activity collected onto aerosol filters. In case of negligible amounts of artificial radionuclides in the air both the alpha and beta activity on the filter first decrease rapidly owing to the decay of radon-220 progeny (Figure 2A). Five days after the end of sampling radon-220 progeny has decayed into stable lead and the measured beta activity consists mainly of bismuth-210 more or less in equilibrium with lead-210. Total alpha activity starts to gradually increase as polonium-210 grows in to the sample. In case of fresh fission products on the filter both the alpha and beta activity present on the filter first also decrease rapidly owing to the decay of radon-220 progeny (Figure 2B). The alpha activity starts to increase due to the in-growth of polonium-210 whereas the total beta activity gradually decreases due to the decay of short-lived fission products.

A sharp peak in the atmospheric total beta activity concentration was observed in Helsinki in conjunction with airborne iodine-131 (Figure 3). The highest total beta activity concentration values, 1800 µBq/m³, can in rare cases be of natural origin, i.e. caused by lead-210 (16). The activity concentration values are about twice the ones observed in the High Arctic (20). A comparison of Fukushima-related total beta activity concentrations to certain earlier incidents observed in Finland is presented in Table 3.

Table 3. Maximum daily total beta activity concentrations (µBq/m³) observed in Finland; NTS = Nuclear Test Site; NPP = Nuclear Power Plant (21-23).

Source	Year	Max. conc. µBq/m³
Atmospheric nuclear tests	1961	1040000
Leakage from Semipalatinsk NTS	1966	1000000
Leakage from Novaya Zemlya NTS	1987	2400
Chernobyl NPP accident	1986	18000000
Sosnovyi Bor NPP incident	1992	3300
Fukushima NPP accident	2011	1800



A) B)

Figure 2. Temporal behaviour of alpha and beta activity on aerosol filters from Helsinki; A) sampling 23-24 February 2011; B) sampling 4-5 April 2011.

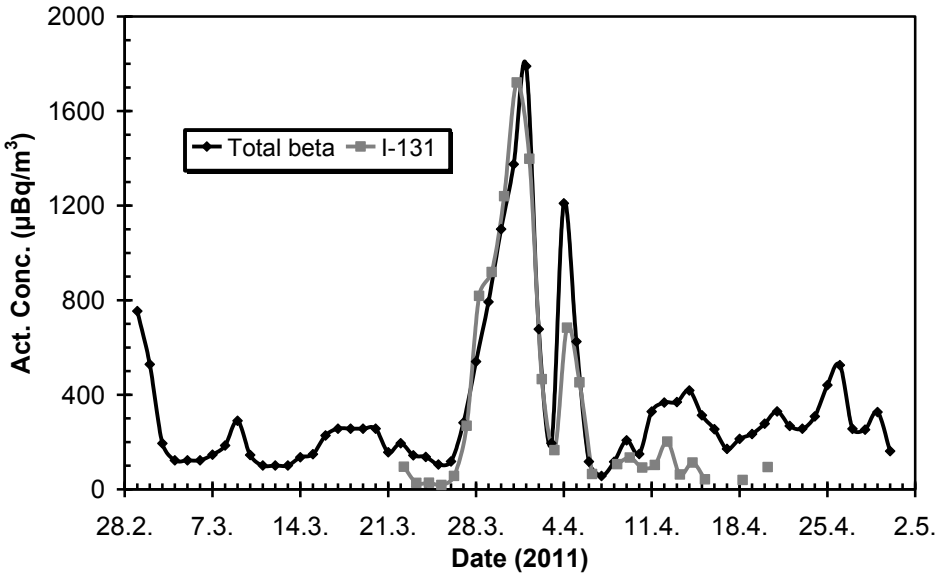


Figure 3. Daily activity concentration of total beta activity and particle-bound iodine-131 in Helsinki in March-April 2011.

Atmospheric deposition results

It is known that the radioactive Iodine isotopes are in different chemical forms. While the particle bounded ^{131}I is analyzed by air filters, to determine the remaining gaseous fraction require charcoal traps. The ratio between attached to the aerosol and gaseous fraction was reported in the close vicinity to the accident in Japan of about 0.40 and in Europe ~0.30 (11), showing that more iodine activity is in the gaseous form. The deposition over the water surface, as in case of NIMH wet deposition collectors is efficient both for aerosol and gaseous fraction but to unknown degree. The pre-concentration by evaporation might cause some loses of deposited iodine. Further studies are needed also to clarify the role of vegetation in deposition processes.

Weekly deposition samples were collected, pre-concentrated and measured for gamma-emitting radionuclides since March 17. The first analyzed weekly sample 17-23.03 show no ^{131}I activity above MDC (Minimum Detectable Concentration). The highest deposition observed in Sofia and Pleven was during the 2nd week – 24-30.03, see Fig.4. These samples were taken on March 31 at 8:00 LST. In some of the samples ^{134}Cs and ^{137}Cs isotopes were also measured: in Sofia, during the period 7-13.04 the deposited ^{137}Cs was 0.2 ±0.1 Bq.m^{-2}week^{-1}; during May -0.10 ±0.04 Bq.m^{-2}month^{-1} and in June - 0.06 ±0.03 Bq.m^{-2}month^{-1}. The ratio ^{134}Cs/ ^{137}Cs in Bulgarian samples vary within the range 0.8÷ 1.1, the uncertainty in activities of the both isotopes are high.

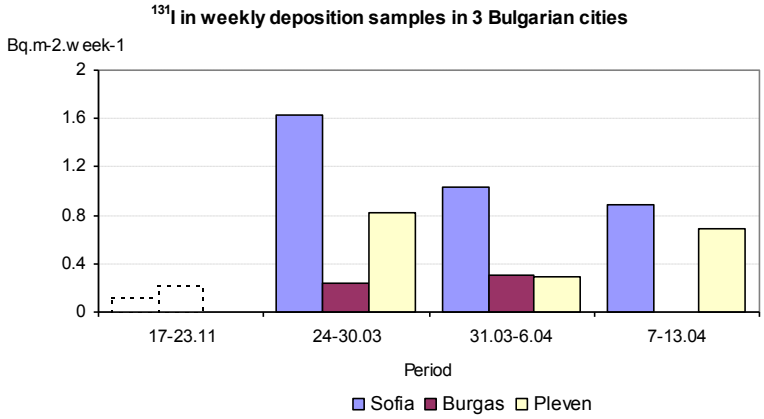


Figure 4. ^{131}I fallout from Fukushima releases in weekly deposition samples in March-April 2011. During the week 17-23.03 the deposition was below MDC.

Table 4. Gamma emiting radionuclides in monthly deposition samples in some Bulgarian cities.

Station	Period	^{131}I, Bq/m^2	± 1σ	^{7}Be, Bq/m^2	± 1σ	^{137}Cs, Bq/m^2	± 1σ	^{134}Cs, Bq/m^2	± 1σ
Burgas	1-31.03.2011	0.46	± 0.18	1.11	± 0.68	<0.98		-	
Karnobat	1-31.03.2011	1.37	± 0.37	<4.13				-	
Silistra	1-31.03.2011	2.81	± 0.32	18.5	± 1.63	0.3	± 0.18	-	
Razgrad	1-31.03.2011	4.41	± 1.45	63.41	± 2.02	<0.38		-	
Dobrich	1-31.03.2011	2.01	± 0.37	5.41	± 0.81	<0.27		-	
Pleven	1-30.04.2011	2.9	± 1.43	25.04	± 3.59	0.38	± 0.14	-	
Sofia	1-30.04.2011	<2.44		85.15	± 2.13	<0.26		-	
Pleven	2-31.05.2011	-		20.12	± 3.2	<0.41		<0.41	
Razgrad	1-31.05.2011	-		4.66	± 1.61	<0.41			
Varna	1-31.05.2011	-		140.47	± 3.38	0.47	± 0.14	0.4	± 0.16

The monthly deposition in March 2011 (sampling period 1-31.03), from several other Bulgarian stations was also analyzed. The results are summarized in Table 4.

The contamination of the air masses that reach Europe from Fukushima releases display much more homogeneous pattern than during Chernobyl accident. During Chernobyl fallout the deposition of beta radionuclides in Sofia was several times higher than in Pleven and other stations in Northeast Bulgaria. More or less the homogeneity of the Fukushima fission products was observed not only in horizontal direction, but also in elevation with sea level in Europe as a whole and in Bulgaria in particular, if compare ^{131}I concentration in Sofia (560 m.a.s.l) to that on peak Mussala (2925m) (11, 13). This is completely different to the radioactive deposition from Chernobyl accident. The maximum in daily deposited 120 h total beta activity, planshet sample, May 2 1986, in Mussala was 2 times higher than 11856 Bq.m^{-2}.day^{-1} measured in Sofia. The increase in deposition was more distinguished if compare total deposited beta for May 1986 in peak Mussala - 79692 Bq.m^{-2}.day^{-1} to 19791 Bq.m^{-2}.day^{-1} in Sofia (^{131}I activity is removed by ashing of the planshet samples).

The radioactivity of daily precipitation from peaks Mussala (Rila mountain) and Botev (Balkan mountains) measured as 0.2-0.45l aliquots was below detection (MDC) as follows:

Combined precipitation, 27-30.03.2011, peak Botev – ^{131}I<0.21Bq/l; ^{137}Cs<0.05Bq/l

Daily precipitation, 13-14.04; 16-17.04; 18-19.04, peak Mussala - ^{131}I<0.06Bq/l; ^{137}Cs<0.1Bq/l.

The ^{131}I I concentration in Sofia precipitation from 29-30.03 and 13-14.04 measured by (13) is given as 0.3 and 0.08 Bq/kg.

Conclusions

Both in Bulgaria and Finland the trace amount of radioactive releases of ^{131}I, ^{134}Cs and ^{137}Cs from the Fukushima Dai-ichi NPP accident were detected and evaluated in March and April 2011. The observed values are similar and in the medium range for the reported in Europe (11,18,19). There was no large inhomogeneity in the measured very low levels of contamination between the two countries and in different country regions in Bulgaria as it was recorded during the Chernobyl fallout (1, 2, 14, 23).

It was considered that a frequency for core damage accident for Light Water Reactors is about 5.10^{-5} y^{-1}. (24). Many reactors, about 100, are in construction or are planned to be built-up in the near future in the world. Most probably this severe accidents frequency will be re-estimated after the Fukushima accident. Even with the additional measures for Reactor's safety and radiation protection the accidental emissions to the environment and to the atmosphere in particular, can not be neglected. The changing reality requires Networks for atmospheric radioactivity monitoring, naturally connected to the national meteorological institutions as FMI and NIMH, to be continuously supported and developed.

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