Variation of the total beta activity in the surface air layer in Bulgaria and Finland during the last decades

Blagorodka Veleva[1], Jussi Paatero[2]
[1]National Institute of Meteorology and Hydrology (NIMH), Sofia, Bulgaria
[2]Finnish Meteorological Institute (FMI), Helsinki, Finland

Source of manmade radionuclides in atmosphere:

1)Nuclear test in the atmosphere

- 1962 a moratorium on atmospheric tests explosions.
- France and China continued during the 1960s and 1970s.
- The last atmospheric nuclear explosion-by China on 16.10.1980.
- UNSCEAR (1988): during 1945-1980 **423** atmospheric nuclear tests.
- Recent information - almost 500 (Geoscience Australia, 2005).

2) other sources - accidental releases mainly

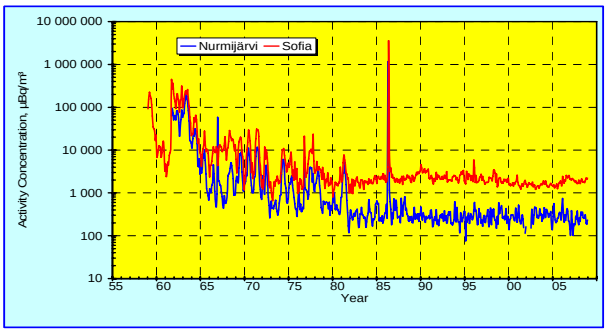
- satellite burning in reentering atmosphere (SNAP 9-A, 1964, Cosmos 954, 1978);
- waste storage (Kyshtym accident, 1957);
- nuclear reactor accidents (Windscale, 1957; Chernobyl, 1986).

3) minor source – operational releases NPPs

ABSTRACT

The atmospheric radioactivity close to the earth surface is dominated by the natural radon isotopes and their daughters' radionuclides. The other group of cosmogenic radionuclides is naturally formed in collisions of the cosmic rays with the atoms of the air gases. Since the beginning of nuclear epoch in 1940's new manmade radionuclides started to be released to the environment. The main source of global contamination with technogenic radionuclides, the prevailing part of which are beta emitters, has been the testing of nuclear weapons in the atmosphere in 1950's, early 1960s'. Therefore both in Bulgaria and in Finland the national networks for airborne radioactivity monitoring were developed within the national weather services (FMI in Finland, and NIMH in Bulgaria). The approaches and methods in NIMH and FMI are similar concerning total beta activity. There are data records available for many years, since 1959 for Sofia and since 1960 for Finnish stations for beta radioactivity of the air particulate and atmospheric deposition measured 120 hours after sampling.

A comparison of results from Finland and Bulgaria indicate that the long-term temporal behaviour of monthly average airborne total beta activity is very similar in these countries. However, a few cases of different behaviour can be observed. The air masses transport and precipitation during the passing of the radioactive plume played the most important role in degree of contamination in different regions during the period of Chernobyl accident in 1986.

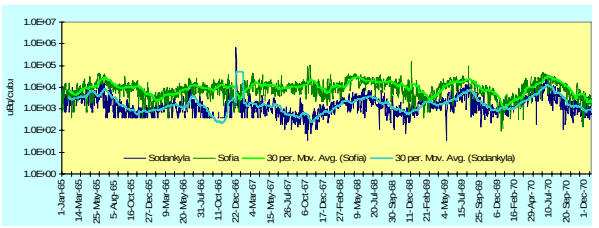


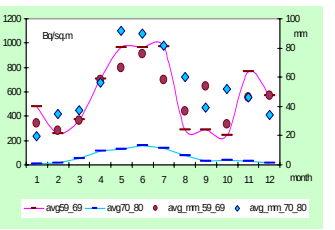
Airborne total beta activity (μBq/m³) in Nurmijärvi (60°30´N, 24°39´E), southern Finland and Sofia (42°41´N, 23°20´E), Bulgaria, monthly mean values. High values in the 1960s and the 1970' are due to the atmospheric nuclear tests and the peak in 1986 due to the Chernobyl accident. In the 1990s and the 2000s the activity is on a natural level (mainly due to lead-210).

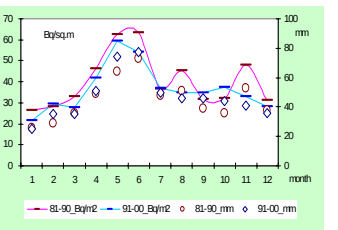
Experimental methods

The measurements of atmospheric radioactivity started at **NIMH**, Sofia, Bulgaria in 1958. The aerosol and precipitation total beta activity is measured initially 72 hours and since 1965 five days after end of sampling. During the years the monitoring station network expanded and since 1969 a network of 5 daily aerosol stations has been operated. The aerosol samples are collected on paper filters. 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 radiation detectors have changed from GM counters to proportional counters and, in the beginning of the 1980s, to plastic scintillator detectors [1].

The **FMI** began measurements of total beta activity in the air in the early 1960's. Daily and weekly aerosol samples have been collected onto glass fiber or paper filters at several stations. 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 [2]. The detector arrangement used in the analyzers consists of five large-area (600 cm²) gas-flow proportional counters. The flow gas is a mixture of argon (90 %) and methane (10 %). Background samples (unexposed filters) and reference samples are measured daily. The total 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.







The concentrations measured in Sofia are higher then in Finnish stations with only few exceptions. In December 1966, leaking underground nuclear test in Semipalatinsk, affected Finnish but not Bulgarian stations. The daily values in Sofia and Sodankyla (67°22´N, 26°39´E) are shown for comparison on Figure above. The meteorological situation combined with constant absolute vorticity trajectory calculations indicated that the source of the plume was in Central Asia. Based on the seismographic observations it was suggested that the radioactivity originated from an underground nuclear test at the Semipalatinsk test site 18 December 1966 04:58 UTC. The passage of the fission products through the soil into the atmosphere caused a high fractionation of the nuclides. Strontium-89 and caesium-137 were enriched in the nuclide mixture compared to their fission yields. These nuclides have noble gas precursors which facilitated their escape into the atmosphere. Owing to a stable inversion layer around the 850 hPa pressure level the vertical dilution of the plume was prevented thus preserving the high concentration of the fission products [5].

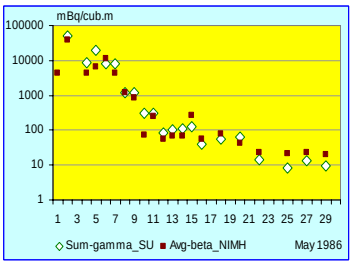
The deposition by precipitation is the main removal process for pollutants in the atmosphere. The monthly variations in the deposited total beta activity [Bq/m2] and in monthly precipitation [mm] in Sofia, four consequent 10 years periods, is presented. The highest activity is deposited during the period of global bomb fallout.

Table 1. Deposited by precipitation long-lived beta activity during the years with highest fallout in kBq/m2 per year.

Year	Sofia	Karnobat	Kjustendil	Gramada	Helsinki*	Ivalo*
1962	17.5	12.6	14.8	11.1	-	-
1963	28.7	15.8	13.9	10.5	-	9.6
1964	4.5	2.4	3	2.8	-	2.3
1986	14.4	15.3	11.1	0.7	6.0	0.8
1993	0.24	0.19	0.34	0.10	0.08	0.05

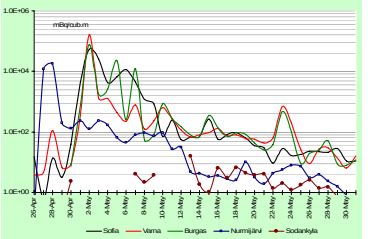
*The total monthly deposition is considered for Finish stations.

Year 1993 is given as a representative for the calm and almost free from artificial beta radionuclides atmosphere



The Chernobyl accident caused dispersion and deposition of a number of fission and activation radionuclides, in total, excluding RNG, of about 8.10^{18}Bq. The concentrations of long lived beta aerosol rose from background values of about or less than 1 mBq/m³ to 18 Bq/m³ in Nurmijarvi, 52.8 Bq/m³ in Sofia and up to 163 Bq/m³ in Varna, due to the enrichment of the sample with hot particles. The atmospheric radioactive contamination in Bulgaria and Finland due to Chernobyl releases is very inhomogeneous, [3] and [4]. *Daily values of long-lived beta activity in aerosol in 3 Bulgarian and 2 Finnish stations during the period of direct Chernobyl fallout.*

Beta activity (μBq/m³) in Sofia in May 1986 sampled and measured at NIMH compared to the sum of activity of gamma-emitting radionuclides, sampled and measured at Faculty of Chemistry of SU, Sofia. NIMH data are averaged over the period of sampling in SU if it cover 2-3days. Correlation coeff. = 0.966. The prevailing measured activity by gamma is due to beta decay of Zr-95, Mo-99, Ru-103, Ru-106, Sb-125, I-131, Te-132, I-132, Cs-134, Cs-137, Ba-140, Ce-141, Ce-144.



Conclusions

Monitoring of airborne total beta activity is often considered less important than gathering nuclide-specific data. Nonetheless, the total beta activity monitoring has been shown to be a reliable and a sensitive surveillance and screening method. The method is also cost-efficient as it can be utilised with relatively simple instruments and laboratory staff without academic degrees. These reasons have directed also the European Union to adapt the method for its routine radiation monitoring system.

References

[1] Mattsson, R.., Paatero, J. and Hatakka, J. Automatic Alpha/Beta Analyser for Air Filter Samples - Absolute Determination of Radon Progeny by Pseudo-coincidence Techniques. Radiation Protection Dosimetry, 1996. Vol. 63, pp. 133-139.
[2] Manolov, L. and Teneva, M. Bulletin of the Institute of Hydrology and Meteorology. Vol.1. Sofia: Publishing house of the Bulgarian Academy of Sciences, 1964.
[3] Paatero, J., Hatakka, J., Mattsson, R. and Viisanen, Y. Analysis of Daily Lead-210 Air Concentrations in Finland, 1967-1996. Radiation Protection Dosimetry, 1998. Vol. 77, pp. 191-198.
[4] Antonov, A., Veleva, B., Adjarova, L. and Kolarova, M. Time and area distribution of low level radioactivity of technogenic radionuclides in the surface air and fallout over the territory of Bulgaria. In: Povinec, P. (Ed.). Proc. of the 14th Europhysics Conference on Nuclear Physics "Rare Nuclear Processes", 22-26 Oct 1990, Bratislava. Singapore: World Scientific, 1992. pp. 407-419.
[5] Kauranen, P., Kulmala, A. and Mattsson, R. Fission Products of Unusual Composition in Finland. Nature, 1967. Vol. 216, pp. 238-241.
[6] Paatero, J., Mattsson, R. and Hatakka, J. Measurements of airborne radioactivity in Finland 1983-1990, and applications to air air quality studies. Publications on Air Quality No. 17. Helsinki: Finnish Meteorological Institute, 1994.
[7] Paatero, J. and Hatakka, J.. Measurements of Long-lived Radioactivity in the Air and Precipitation in Finland 1991-1994. Publications on Air Quality No. 26. Helsinki: Finnish Meteorological Institute, 1997.
[8] Results of investigation of the radiological situation in NR Bulgaria after the accident of the Chernobyl NPP, Committee for Use of Atomic Energy for peaceful purposes, November 1986, Sofia.