Xenon and Krypton-85 Radionuclides Monitoring in the Northwest Region of Russia

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INTRODUCTION
Xenon and krypton radionuclides are formed in large quantities during nuclear fission at nuclear power plants and nuclear explosions, krypton-85 is also emitted at reprocessing of spent nuclear fuel. The manufacture of radiopharmaceutical tracers for medicine are also xenon radionuclides sources.

In USSR-Russia $^{85}\text{Kr}$ monitoring was carried out in 1977-1993 by the employees of Institute of Experimental Meteorology– RPA “Typhoon” [Makhon’ko, 1990, 1992]. Soon after the USSR disintegration works on $^{85}\text{Kr}$ monitoring in Russia were ceased. $^{85}\text{Kr}$ monitoring in the world was carried out from the end of the 50-ths of the 20-th century. $^{85}\text{Kr}$ concentration during this period of time was steady increasing.

In late 80-ties – early 90-ties of the last century the employees of the Radium Institute carried out noble gases radionuclides monitoring both in the USSR and the neutral waters of the Atlantic Ocean. These works were performed under the guidance of prof. A. Krivokhatskyi. Large researches cycle on $^{85}\text{Kr}$ monitoring was carried out in Chelyabinsk area and in La Manche Channel (Pakhomov, 1990, 1991). $^{85}\text{Kr}$ volumetric activity was then about 0.9 Bq/m$^3$.

$^{85}\text{Kr}$ and xenon radionuclides monitoring was recommenced at V. G. Khlopin Radium Institute in 2006 in frames of 2133 ISIC Project.

Basic xenon radionuclides formed as a result of nuclear fission reaction are $^{133}\text{Xe}$ ($T_{1/2}$=5.245 day), $^{135}\text{Xe}$ ($T_{1/2}$=9.1 hour), $^{133m}\text{Xe}$ ($T_{1/2}$=2.19 day) and $^{131m}\text{Xe}$ ($T_{1/2}$=11.9 day). $^{133}\text{Xe}$ is most often among this isotope family because of its highest yield in fission, and its half-life is rather long, which makes it possible to register this radionuclide at the significant distances from a plume.

MATERIALS AND METHODS
For monitoring conduction samples of krypton-xenon fraction were again taken at one of the works for oxygen production in Cherepovets city (59°07′59″N и 037°55′26″E.) of Vologda province and analyzed at our laboratory. Krypton and xenon separation was carried out using low-temperature adsorption on activated charcoal SKT-3 and their subsequent concentration as spectrometric samples.

Gamma-spectrometers with Ge detectors were used as Kr-85 measuring means. As gamma-quants yield in $^{85}\text{Kr}$ beta-decay is only 0.435% (Golashvili, 2004), its measurement by gamma-spectrometry method is very difficult. Therefore it seems expedient to use krypton sample obtained after reprocessing of large volume atmospheric air sample for $^{85}\text{Kr}$ measurement. Krypton fraction obtained from reprocessing of air sample with $\sim$2000 m$^3$ of air was adsorbed on activated charcoal in hermetic measuring container. The krypton sample container was places on Ge detector; measurement duration was not less than 2 hours, each sample being measured 3-5 times. Gamma-spectrometer was calibrated using a standard
sample in the same container. $^{85}$Kr volumetric activity was calculated taking into account the fact, that stable krypton (volumetric) concentration in an atmospheric air is $1.14 \times 10^{-4} \%$.

Kr volume in measuring ampoule was from 500 to 1400 cm$^3$ (STP), which corresponds to the sample separated from the atmospheric air 450-1200 m$^3$ by volume. During a month some samples were taken, sample duration being about 24 hours.

High sensitivity technique was especially elaborated for xenon radionuclides monitoring. Xenon fraction separated from krypton-xenon mixture was absorbed on SKT-3 charcoal in small-size ampoule set up in HPGe-detector well. Minimum $^{133}$Xe activity detected by a spectrometer makes up 9 mBq for 24 hours’ measurement.

Technique elaborated here makes it possible to concentrate in measuring ampoule xenon sample obtained during air sample reprocessing having a volume up to 1500 m$^3$. Thus, at present this technique has record sensitivity, and using this technique $^{133}$Xe minimum detectable concentration (MDC) achieved its lowest value of 0.008 mBq/m$^3$ at a confidence level of 95%.

Technique for Xe and Kr radionuclides separation from liquid oxygen samples was also elaborated; it allowed to carry out monitoring in St-Petersburg. Kr and Xe in increased concentrations ($> 5 \times 10^{-4} \%$ in sum) as compared with the atmospheric air are known to be present in technical oxygen. Kr and Xe are separated from technical oxygen samples 5-7 l by volume, the volume of xenon fraction separated being equivalent to 25-90 m$^3$ of atmospheric air reprocessing. Measurements were carried out twice a week.

**RESULTS AND CONCLUSIONS**

**Kr-$^{85}$ monitoring**

First measurements results obtained in October-November, 2006, had shown $^{85}$Kr volumetric activity at an observation station at Cherepovets to be ~1.5 Bq/m$^3$, that is, substantially increased as compared with the 90-ties beginning. Available literature data (Hirota, 2004, Winger, 2005) show, that significant $^{85}$Kr concentration increase occurred in an atmosphere of both West Europe and Japan in the period from 1993 to 2004.

A plot of $^{85}$Kr concentration change in the atmospheric air of Cherepovets is shown in Fig.1. $^{85}$Kr volumetric activity in this period of time was changing from 1.41 to 1.94 Bq/m$^3$, mean value being 1.65±0.12 Bq/m$^3$. Really, volumetric activity in northwest region of Russia has increased in almost 50% for late 15 years.

**Xe radionuclides Monitoring**

Xenon radionuclides concentration measurements in an atmospheric air of Cherepovets situated in 200 km from Kalinin NPP were carried out from early August, 2006 to February, 2008.

A graph of $^{133}$Xe concentration change from the very offset of monitoring is presented in Fig.2. Its concentration was changing from a background value of 0.09 to 2.47 Bq/m$^3$ during this period, mean concentration value made up 0.79±0.48 mBq/m$^3$, and median was 0.68 mBq/m$^3$. It is evident, that mean value and median are close, and exceed minimum value almost 8 times. Backward trajectories of air mass transfer calculation makes it possible to suppose, that $^{133}$Xe increased concentration (up to 2.5 mBq/m$^3$) could be transported from the area of Kola peninsula in September, 2006, and on 15-16, January, 2008 (2 mBq/m$^3$) – from the area of Kalinin NPP (Udomlya town). However, in most cases it was possible to
determine the sources of $^{133}$Xe injection into the atmosphere. They were located both in CIS European territory and in west Europe. $^{133}$Xe arrivals were observed from Kola Peninsula region, Smolensk, Voronezh and Tver' provinces of Russia, from Ukraine, Lithuania, Finland, Scandinavia and West Europe. But each case should be analyzed.

$^{133}$Xe concentration in St.-Petersburg was changed from background values (0.2-0.3 mBq/m$^3$) to 185 mBq/m$^3$ in the period from March, 2007 to February, 2008 (Fig.3). $^{133}$Xe values exceeding 10 mBq/m$^3$ were observed in 20% cases. Median for this series of observations is 2.5 mBq/m$^3$.

**Fig.1.** $^{85}$Kr in Cherepovets atmospheric air during period September, 2006 –February 2008

**Fig.2.** $^{133}$Xe in Cherepovets atmospheric air during period August, 2006 –February 2008

**Fig.3** $^{133}$Xe volumetric activity in St.-Petersburg atmospheric air
It is to be noted that some cases were registered in this period when $^{135}$Xe was present in samples ($T_{1/2} = 9.14$ hour), $^{135}$Xe/$^{133}$Xe activity ratios changing from 0.033 to 3.5, whereas activity ratios of these radionuclides at equilibrium in the reactor zone is about 0.3-0.24 (Finkelstein, 2001). Correlation between xenon radionuclides concentrations was not found. When the increased concentrations of $^{133}$Xe and $^{135}$Xe were registered in atmospheric air, particles were transferred, as a rule, from western and south-western directions. The nearest nuclear facilities to the west of St.-Petersburg are the Leningrad NPP (Russia), two NPPs in Finland, and Swedish NPPs.

Previous years (2000, 2003, and 2004) there were also detected events of $^{133}$Xe and $^{135}$Xe radionuclides appearing in atmospheric air. At that there could be observed the repeated increases in regional background level for $^{133}$Xe (up to 100 mBq/m$^3$) and infrequently for $^{135}$Xe (up to 30 mBq/m$^3$) (Dubasov, 2004).

Thus, thanks to implementation of the ISTC Project # 2133 there has been recommenced the monitoring of $^{85}$Kr in the Russia, which indicates the growth of this radionuclide concentration, similar to that in Western Europe. The monitoring of Xe radionuclides in the north-west region of Russia has made it possible to reach the data on the artificial radioactivity in the atmosphere in monitoring points and provide estimation of injection region. The presence of short-lived $^{135}$Xe in the atmosphere indicates that the standard mode of NPP's gas treatment is changed and allows estimation of a distance to injection source.

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REFERENCES


