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### **133**Xe in Surface Air of St. Petersburg

Yu. S. Popov, Yu. V. Dubasov, N. M. Kazarinov, V. V. Mishurinskii, V. Yu. Popov, and N. V. Skirda

Khlopin Radium Institute, Research and Production Association, St. Petersburg, Russia

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**Abstract**—In the course of testing new devices for measuring atmospheric concentrations of radioactive noble gases and also of the monitoring techniques, large number (116) of determinations of Xe in surface air of St. Petersburg was made (100 in the period from December 1997 to December 1998 and 16 in the second half of March 2000). In 1998, samples were taken at 6-h to 7-day intervals with documentation of data on the wind direction, and in 2000, at 1-day intervals, but without fixing the wind direction. Statistical treatment of data obtained in 1998 gives the mean volume activity of $2.3 \pm 0.6 \text{ mBq m}^{-3}$. In 1998, this value was exceeded in three cases by a factor of 2–3 and in six cases by a factor of 5–16. In March 2000, even higher values were observed, exceeding by a factor of up to 43, which was caused essentially by releases from the Leningrad NPP. Data obtained in 1998 also show that the specific activity of $^{133}$Xe depends on the wind direction.

Radionuclides of Xe are formed in power and research nuclear reactors and, being chemically inert, enter the environment. Monitoring of the Xe concentration in air is important, since it characterizes the safety of the reactor operation. Therefore, it seems necessary, along with the system of monitoring of radioactive aerosols now existing in Russia, to organize monitoring of radioxenon, like that realized, for example, in Germany, Sweden, and some other West-European countries.

In the period from 1998 to 2000, we measured the concentrations of radioactive noble gases in St. Petersburg with the aim of testing new measuring systems and monitoring techniques. Here we report data on $^{133}$Xe.

**EXPERIMENTAL**

The specific activity of $^{133}$Xe in air of St. Petersburg was measured with BRIG and ARIKS sampling and measuring systems. Each of them consists of two major units: a sampling and sample preparation unit and a measuring unit. Using the measuring block of the BRIG system, we generally determined $^{85}$Kr ($T_{1/2} = 10.7$ years), $^{133}$Xe ($T_{1/2} = 5.25$ days), and $^{135}$Xe ($T_{1/2} = 9.09$ h), and with ARIKS system, $^{131m}$Xe ($T_{1/2} = 11.97$ days) and $^{133m}$Xe ($T_{1/2} = 2.19$ days) in the Xe preparations. The Xe activity was measured on $\gamma$-ray spectrometers operating in the $\beta-\gamma$-coincidence mode.

BRIG and ARIKS systems are highly sensitive: the $^{133}$Xe detection limits are 1.0 and 0.5 mBq m$^{-3}$, respectively. The principal feature of ARIKS is that, in contrast to BRIG, it is a fully automatic system, which can operate for a long time without human intervention. Air sampling proceeds continuously and analysis data are reported through a selected communication channel.

The procedure of determination of Xe radionuclides includes three steps. First, Xe is separated from air. Second, the sample is treated to remove admixtures, primarily, the concomitant $^{222}$Rn. The third step includes $\gamma$-spectrometric determination of Xe radionuclides, gas-chromatographic determination of stable Xe, and, finally, estimation of the specific activities of radionuclides in air.

Separation of Xe from air is carried out by low-temperature adsorption on a solid sorbent. Air flow pretreated to remove moisture and CO$_2$ is cooled to the temperature of adsorption (about $-170^\circ$C) using a MOV autonomous microcryogenic system operating by a closed loop and providing a process temperature of 80 K. The sampling step takes 5–7 h and 12 h in BRIG and ARIKS systems, respectively (in this time, up to 8 and 10 m$^3$ of air, respectively, is processed). After completion of this step, preparative gas chromatography is used to separate and further concentrate the Xe fraction from adsorbed gases. The final product has a volume of 8–9 cm$^3$ (normal conditions) and contains about 0.6 cm$^3$ of Xe in the BRIG system and no less than 0.8 cm$^3$ in ARIKS (helium is the major gas constituent of the product). Preparation of a Xe sample from the adsorbed gases takes about 4 h. From 75 to 85% of the final product goes to spectrometric
analysis, and the residual part is used for determination of the Xe concentration in the product by gas-chromatographic analysis. Knowledge of this value allows estimation of the volume of processed air and, therefore, the specific activities of the radionuclides in air.

After transfer of the Xe fraction to analysis, the sampling and sample preparation unit is regenerated for the next sampling cycle.

The $^{133}$Xe activity in samples prepared using the BRIG system was determined relative to a reference source of $^{133}$Xe using Eq. (1).

$$A_x = A_r (S_{x} - S_{br})/(S_{r} - S_{br}), \quad (1)$$

where $A_r$ is the $^{133}$Xe activity in the reference sample (Bq); $S_{x}$ and $S_{r}$ are the areas under the photopeaks at 30 and 80 keV in the $\gamma$-ray spectra of the test and reference samples, respectively (with no allowance for the baseline), registered in the $\beta-\gamma$-coincidence mode (counts); and $S_{x}$ and $S_{br}$, areas under the baselines in the spectra of the test and reference samples (before introduction of the samples) (counts).

The parameters $A_r$ and $S_{r}$ are set to the computed memory in calibration of the spectrometer. The areas contained in Eq. (1) were determined using a data processor connected to the spectrometer. Finally, the specific activity of the radionuclide was calculated by Eq. (2)

$$C_x = \frac{\nu A_x}{V_x} \frac{\omega}{1 - \omega} \exp(\nu \Delta t), \quad (2)$$

where $\nu$ is the volume concentration of Xe in $1 \text{ m}^3$ of surface air ($8.7 \times 10^{-2} \text{ cm}^3$ under normal conditions); $V_x$, volume concentration of Xe in the sample under normal conditions, $\text{cm}^3$; $\omega$, volume fraction of the sample taken for gas-chromatographic analysis (from 0.15 to 0.25); $A_x$, $^{133}$Xe activity in the sample, Bq; $\Delta t$, time elapsed from the completion of the sampling step to the beginning of the spectrometric analysis of the sample, h; and $\lambda$, radioactive decay constant, $\text{h}^{-1}$.

The error of determination of $C_x$ is a sum of errors of the parameters contained in Eq. (2). The error of determination of $A_r$ depends also on the recording time of the spectrum. In measurements of the specific activity of $^{135}$Xe with the BRIG system, the exposure was varied from a few hours to 16 h, depending on the wind direction and the presence of $^{135}$Xe.

In 2000, in the course of two-week tests of the ARIKS system, the samples were taken once a day. The duration of sampling was fixed (12 h) as well as the exposure in $\gamma$-spectrometric measurements (10 h).

The results of 1998 are summarized in Fig. 1 and Table 1 (data are arranged in accordance with the wind direction during sampling). The results of 2000 are given in Table 2.

Statistical treatment of all measurements of 1998 gives the mean volume activity of 2.3 ± 0.6 mBq m$^{-3}$. In 1998, this value was exceeded in three cases by a