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Some results of determining the source and reasons for the appearance of ^{106}Ru in Russia in September-October 2017

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Abstract. Probable locations of the sources of ruthenium-106 in the atmosphere in September-October 2017 have been considered. Taking into account the monoisotopic nature of the increase in ruthenium-106 concentration in the ground level air and atmospheric precipitation, the most probable version is substantiated, which suggests that ruthenium-106 came from a ground source located in the Southern or the Middle Urals. In order to locate the possible point of Ru-106 emission into the atmosphere, calculations of the time-inversed travel of air masses at the heights of 500-1500 m from the Ru-106 concentration measurement locations at European points during the relevant observation period were used. To confirm the correctness of the source location, calculations were performed that involved simulating regional transport and propagation of radioactivity in the near zone in case of a hypothetical emission of ruthenium-106 from a conventional source in the Southern Urals, in order to compare the simulation results with the available daily ruthenium-106 drop-out measurement data.

1. Introduction

According to an IAEA report [1, 2], in the period from September 25 to October 9, 2017, a radionuclide of ruthenium-106 in concentrations from 10 mBq/m^3 to 100 mBq/m^3 was detected in air samples of aerosols in the atmospheric air in the territory of European countries. The maximum concentration of 145 mBq/m^3 was registered in Romania. Such concentrations of ruthenium-106 in the atmospheric air do not pose a radiation hazard to the population and the environment. Nevertheless, the established international practice in the field of radiation monitoring related to such situations requires clarification of sources and reasons for the increase in the content of radionuclides in the environment. It should also be mentioned that the concentration of ruthenium-106 in air samples largely depends on the sampling time (exposure time). In this case, the exposure time at different observation points varied widely from several hours to several days and even weeks, making it difficult to perform a quantitative comparison of the available observational data. The prolonged nature of sample exposure makes it impossible to determine the exact maximum concentration of ruthenium-106 in the ground level air aerosols and the period of time during which it was observed.

There are various situations in which radionuclides may enter the environment. This may be an abnormal situation at the radiation-dangerous object associated with the loss of control over the release of volatile radionuclide compounds, or accidental disposal of a radioactive source, for example, when remelting metal scrap. However, with regard to the case under consideration, not a single enterprise, neither in Europe nor in Russia, declared an abnormal emission of ruthenium-106 during its production.

In the absence of information about the source, German and French specialists calculated so-called reverse trajectories from the points where ruthenium-106 was registered in Europe and estimated the location of the possible source. Based on the data obtained, it was stated that the emission point of ruthenium-106 is located in Russia, in the Urals.

In this situation, Roshydromet which coordinates activities related to radiation monitoring carried out work to assess the situation of the appearance of ruthenium-106 in the ground layers of the atmosphere and its distribution across Russia and Europe.



2. Analysis of data obtained from monitoring the content of ruthenium-106 in the ground level air aerosols and atmospheric drop-out in the territory of Russia

The main sources of information on the radiation situation in the territory of the Russian Federation are the observational systems of Roshydromet and Rosatom State Corporation. The most informative type of observation is the observation of radioactive air aerosols by means of air-filtering units (AFU). Roshydromet AFUs are located around nearly all first category radiation-dangerous objects in the territory of Russia. Aerosols are sampled with an exposure of 5 days, mainly. Radioactive drop-outs are sampled using horizontal plates with a daily exposure.

Starting from September 23, 2017, an increase in the total activity of beta-emitting radionuclides ($\Sigma\beta$) was observed in the territory of the Chelyabinsk region in the atmospheric aerosols samples, and spectrometric analysis of these samples revealed increased content of ruthenium-106. At the end of September and during the first ten days of October, the presence of ruthenium-106 in the ground level air and atmospheric drop-out was recorded at observation points in the territory of the Chelyabinsk region, Bashkiria, Tatarstan, Ulyanovsk, Samara, Saratov, Volgograd, Leningrad, Murmansk, Kursk, Voronezh and Rostov regions and near Krasnoyarsk city.

3. Analysis of possible ground sources of ruthenium-106 in Russia and in European countries

High levels of ruthenium-106 in the ground level atmosphere were observed across large areas, and therefore the source can be located both in the territory where the maximum levels were observed and at a considerable distance from these territories, due to the result of regional or transboundary transition.

To estimate location of the possible source, calculations were made, which consisted in simulating time-reversed transition of tracers from virtual sources located at the measurement sites. The model tracks the trajectories of time-reversed motion of the tracer particles emitted during the active period of the virtual source. The concentration of tracer particles at a given time interval reflects the probability of the location of the source of contamination acting during this interval.

The intensity of virtual sources corresponds to the measurement results at each point, i.e. the total emission of tracers is proportional to the concentration integrated over time. The time when measurements start and end at each point determines the start and end of the active period of the corresponding virtual source, the duration of each virtual source is equal to the samples averaging time at this point. Tracer properties (sedimentation rate, deposition rate, decay constant and leaching coefficient) are equal to 0.

The calculation is performed using the model of atmospheric diffusion with a predetermined time step Δt . At each step, the field of the average (for the Δt interval) tracer concentration $C(x, y, h)$ is calculated.

The amount of tracer in a random elementary volume ΔV of the calculation area

$$q = \int_0^{\Delta V} C(x, y, h) \times dv$$

At the same time, the total number of tracer in the calculation area at this step

$$Q = \int_0^V C(x, y, h) \times dv$$

The value of $p = q/Q$ can be interpreted as conditional probability that the emission point is located in a given elementary cell at a given time interval, provided that the source is in the calculation area.

The calculation results may be conveniently presented in the form of a field of values of source location probability density.

$$f(x, y) = \frac{1}{Q} \int_0^H C(x, y, h) \times dh$$

where H is the upper limit of the calculation area.

Then, by integrating the values of the $f(x, y)$ field over the area of a randomly specified area, we can calculate the conditional probability of source location in the specified area.

In this case, the $f(x, y)$ integral over the area of the entire calculation region

$$\int_0^{S_0} f(x, y) \times ds = 1$$

i.e. the source is located in the calculation area.

Figure 1 shows an example of calculation results concerning the location of the source of ruthenium-106 as of September 25, 2017. The dots indicate the measurement points (virtual sources) providing the data according to which the calculation was carried out. The most significant measurement data were used with an exposure of not more than 2 days from 20 locations in Europe (according to IAEA data) and Roshydromet data (Argayash, Novogorny, Volgograd, Tsimlyansk) with an exposure of 5 days.

Using the calculation results it is possible to outline areas of the Southern and Middle Urals as a probable location of the source of Ru-106 release into the atmosphere.

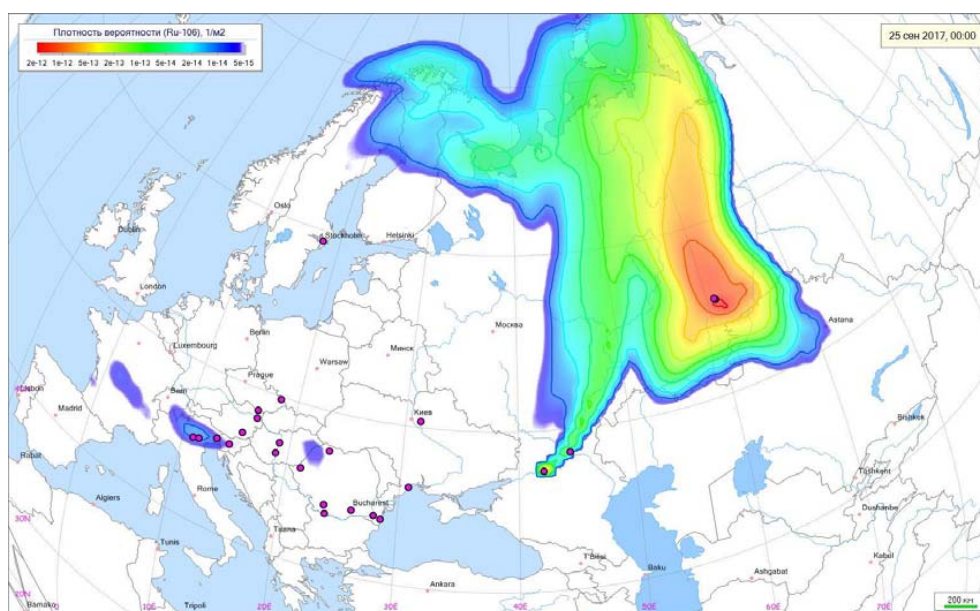


Figure 1 – Probability density of ruthenium-106 source location on September 25, 2017, based on measurements in 20 points in Europe and in 4 points of Roshydromet.

4. Modelling dissemination of ruthenium-106 in the atmosphere from a hypothetical ground source located in the Urals

Scenarios were prepared and a series of calculations was performed to simulate dissemination of radioactivity in case of a hypothetical emission of Ru-106 from conventional sources in the Southern and Middle Urals in order to find the best match between the modelling results with the available data on Ru-106 concentrations at points in Russia and Europe.

The ruthenium-106 emission parameters varied in a wide range, namely: it began to release on September 23–24, 2017, the duration of the emission was from 2 to 7 days, the emission intensity was from 40 TBq/day to 250 TBq/day, the height of the emission was from 100 to 500 m.

Analysing the trajectories of the movement of air masses, it is possible to determine the time interval of the hypothetical ground source of ruthenium emission in the Urals region which is the closest match with the existing measurement data.

Figure 2 shows the calculation results of the ground level concentration of Ru-106 integrated for 10 days from a conventional source located in the Chelyabinsk region near the town of Ozersk.

Table 1 shows the results of comparison of the measurement data obtained at some points in European countries with the calculated data obtained from simulating a hypothetical emission from a conventional source located in the Chelyabinsk region near the town of Ozersk.

Table 1. Comparison of calculation results and measurement data related to ruthenium-106 concentration in the air.

Country	Point	Start	End	Measurements (МБк/м ³)	Calculation, (МБк/м ³)
Austria	Graz	02.10, 8:25	03.10, 10:16	36	30
Austria	Graz	03.10, 0:20	04.10, 7:50	13	10
Austria	Graz	04.10, 8:00	05.10, 7:00	1,0	1
Czech Republic	Ostrava	02.10, 6:00	03.10, 15:00	40	40
Czech Republic	Ostrava	03.10, 5:00	04.10, 15:00	1,1	1
Hungary	Budapest	25.09, 0:00	26.09, 0:00	2,7	0
Hungary	Budapest	26.09, 0:00	27.09, 0:00	1,7	0
Hungary	Budapest	27.09, 0:00	28.09, 0:00	2,0	0
Hungary	Budapest	28.09, 0:00	29.09, 0:00	1,9	0
Hungary	Budapest	02.10, 0:00	03.10, 0:00	27,6	40
Hungary	Budapest	03.10, 0:00	04.10, 0:00	9,9	30
Hungary	Budapest	04.10, 0:00	05.10, 0:00	2,1	3
Italy	Udine	02.10, 0:00	03.10, 0:00	49,1	10
Italy	Udine	03.10, 0:00	03.10, 0:00	54,3	15
Italy	Udine	03.10, 0:00	04.10, 0:00	30	3
Italy	Udine	04.10, 0:00	05.10, 0:00	5,2	1
Italy	Udine	04.10, 0:00	04.10, 0:00	4,2	1
Romania	Bucuresti	29.09, 0:00	29.09, 0:00	37,9	40
Romania	Bucuresti	30.09, 0:00	30.09, 0:00	145	150
Romania	Bucuresti	01.10, 0:00	01.10, 0:00	18,1	20
Sweden	Stockholm	30.09, 8:42	01.10, 8:42	0	0
Sweden	Stockholm	01.10, 8:42	02.10, 8:42	17	8
Sweden	Stockholm	02.10, 8:42	03.10, 8:42	9,8	10
Switzerland	Lugano	02.10, 0:00	02.10, 23:59	0,1	0,1
Switzerland	Lugano	03.10, 0:00	03.10, 23:59	1,9	0,4
Switzerland	Lugano	04.10, 0:00	04.10, 23:59	0,3	1
Switzerland	Lugano	05.10, 0:00	05.10, 23:59	0,5	0

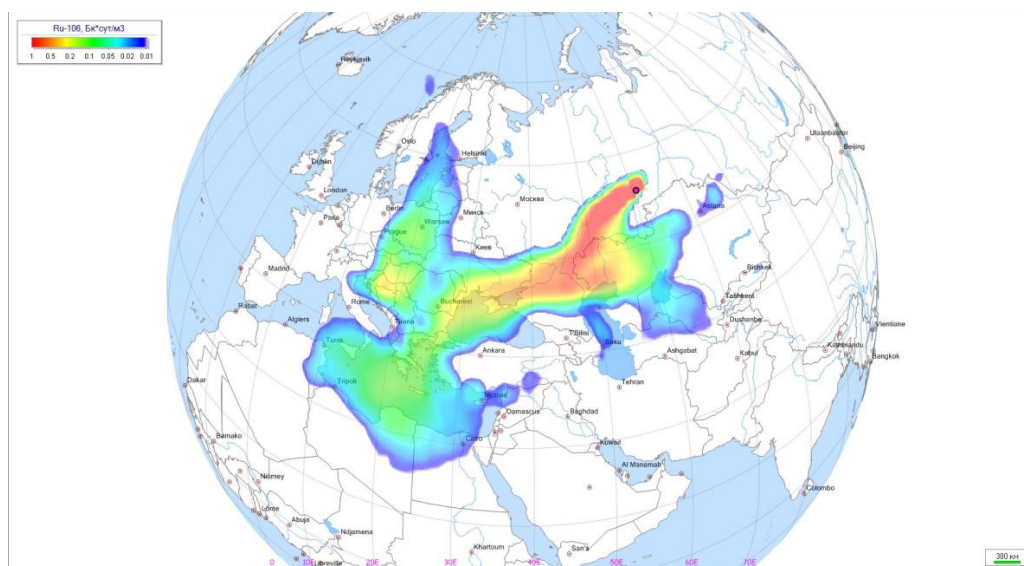


Figure 2. The ground level concentration of Ru-106 integrated over 10 days according to the results of simulating Ru-106 dissemination in the atmosphere.

5. Modelling dissemination of ruthenium-106 in the near zone from a hypothetical ground source located in the Southern Urals

Calculations were performed that involved simulating propagation of radioactivity in the near zone in case of a hypothetical emission of ruthenium-106 from a conventional source in the Southern Urals, in order to compare the simulation results with the available daily ruthenium-106 drop-out measurement data.

When calculating the following parameters of the conventional source were adopted:

- source coordinates: 60.804° E, 55.696° N;
- start of emission: September 23, 00:00 (TAE);
- duration: 7 days;
- initial height of the emission: uniform distribution in the 50-100 m interval;
- source intensity: 40 TBq/day;
- dry deposition rate: $1 \cdot 10^{-5}$ m/s;
- calculation period: 7 days.

Figure 3 shows the results of calculating the total drop-out of ruthenium-106 over 7 days under given conditions.

Table 2 compares results of the simulation according to the conditions of the above scenario with the daily ruthenium-106 drop-out measurement data.

Table 2. Comparison of calculation results and measurement data related to daily drop-out of ruthenium-106 obtained by Roshydromet RMN.

Point	Measurements ^a (Bq/m ²)	Calculation (Bq/m ²)
Kyshtym	51	50
Argayash	25	24
Novogorny	66	64
Khudaiberdinsky	32	41
Metlino	37	110

^a For the period of 7 days exposure (September 23-30, 2017).

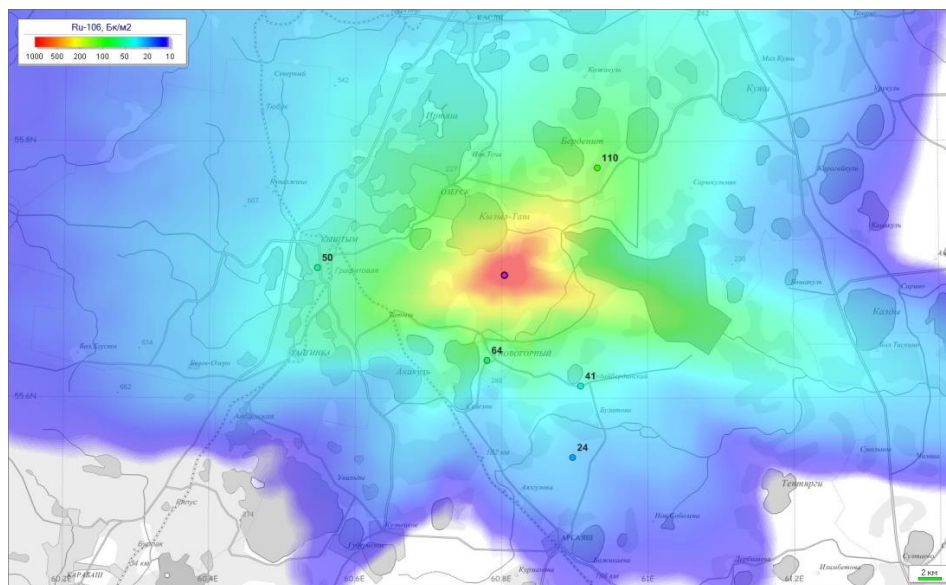


Figure 3. Integral drop-out of ruthenium-106 in the near zone over 7 days based on simulation results (the dots indicate the locations and values of ground measurements).

6. Conclusion

The results of modelling ruthenium-106 dissemination from conventional sources located in the Southern Urals (both on regional and local scale), taking into account the adopted values of the calculation parameters, show good agreement with the measurement data submitted by the IAEA and Roshydromet.

According to ruthenium-106 daily drop-out measurements, conducted by Roshydromet RMN (Radiation Monitoring Network), increased drop-outs were registered from September 23 to September 30, that is, the source remained active during this period. At the same time, the increased concentrations of ruthenium-106 in Europe (in the course of regional transition) are apparently due to the emission during the period from September 25 to September 26.

References

- [1] IAEA – International Atomic Energy Agency. Status of Measurements of Ru-106 in Europe. IAEA, Vienna, 2017. – 5 p.
- [2] IAEA – International Atomic Energy Agency. Updated Technical Attachment Status of Measurements of Ru-106 in Europe. IAEA, Vienna, 2017. – 19 p.