Radioiodine accumulation on soil and reconstruction of doses from iodine exposure on the territory contaminated after the Chernobyl accident

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The paper presents distributions of external doses and absorbed doses from internal exposure to the thyroid on the territory of the former USSR due to exposure of ^{131}I and ^{132}I released into the environment as a result of the Chernobyl accident. Dose assessments were based on measurements of daily depositions of ^{131}I and ^{132}Te from the atmosphere made by SPA "Typhoon". The main data base of available measurements were added with calculations of ^{131}I depositions from space-time correlations and results of measurements of total β-activity, ^{137}Cs etc. Based on these data calculations were made of ^{131}I and ^{132}I accumulation on soil surface, exposure dose rate and exposure dose from γ-irradiation ^{131}I and ^{132}I from the soil surface, and absorbed thyroid doses from incorporated ^{131}I and ^{132}I. Estimated errors in calculations are also presented.

The also paper includes generated maps of ^{131}I contamination of the soil by 15 May 1986 and distribution of external and internal thyroid exposure doses from incorporated ^{131}I and ^{132}I accumulated by 1 September 1986.

The dominant radiation load from ^{131}I and ^{132}I has been shown to be due to internal thyroid exposure to incorporated ^{131}I. If there had been no iodine prophylaxis and restrictions in the diet of the residents, it could have been three orders of magnitude higher than the doses from external exposure.

In different points of the former USSR, the maximum contamination was reported on different dates from 28 April to 3 May 1986. By estimation, the highest radiation loads due in the major cities to ^{131}I occurred in Gomel where by 1 June the exposure dose was 134 mR and the thyroid exposure dose was 166 cSv.

Introduction

The Chernobyl accident which occurred at 1.23 a.m. on 26 April 1986 had a significant effect on the radiation situation on the territory of the former USSR. Prior to the accident, short-lived radionuclides of the technogenic origin practically did not occur in the atmosphere.

The reactor accident itself was an explosion with a release of radioactive materials accumulated in the reactor during its operation primarily to the lower troposphere. High temperatures resulted in the graphite burning in the reactor which was accompanied by evaporation and oxidation of structural materials and, as a consequence, radioactive materials continued to be released to the atmosphere even after the explosion.

The measures taken to smother the reactor with inert materials first led to a decrease in the radioactive release rate ***Q(t)*** from 27.04 to 01.05.86, but at the same time, the temperature in the graphite burning source started to grow and because of this ***Q(t)*** began to grow again after 02.05.86. After 6.05.86 the temperature lowered and ***Q(t)*** began to fall and by 23.05.86 decreased by 5-6 orders of magnitude. This decrease was not monotonic and was associated with considerable variations in ***Q***. Later on, the release rate, on the average, continued to decrease slowly, though on separate days fluctuations with considerable amplitude were observed.

If inert radioactive gases are neglected , the largest activity of the release was due to short-lived ^{131}I, $^{132}Te + ^{132}I$ and $^{140}Ba + ^{140}La$ and $^{95}Zr + ^{95}Nb$.

A major radiation hazard immediately after the accident was posed by ^{131}I. Together with other radioactive materials of the accident, ^{131}I spread over an extensive area in a short time contaminating air, fields, forests, water bodies and agricultural lands. In particular, it deposited from the atmosphere and contaminated pastures and then was taken in by pasturing animals with grass. Further, along the food chain "grass-cow-milk" ^{131}I entered humans and accumulated in the critical organ - thyroid. Due to the selectivity of ^{131}I accumulation, the absorbed dose in the thyroid could be much higher than the whole body dose in the population living on the contaminated areas.

This paper considers accumulation of ^{131}I on the soil surface and distribution of possible external and internal doses in the population due to ^{131}I exposure in the European part of the former USSR after the Chernobyl accident. These calculations are only estimates as they are based on experimental data on daily atmospheric depositions of ^{131}I and do not allow for the complicated pattern of volatile iodine behaviour in the system "soil-vegetation cover-atmosphere" which is dependent on weather conditions, type of underlying surface, type of vegetation, ratio of physico-chemical forms of iodine and many other factors.

Another important goal of our work was to verify the primary set of measurements of ^{131}I, ^{132}I in the atmospheric deposition samples. For this purpose, results of several measurements of ^{131}I, ^{132}I in the same sample or a sample divided into parts were analysed and some data were rejected. The same was done with the primary array of measurements of total β-activity.

1. Method of studies and physico-chemical characteristics of ^{131}I

To reconstruct population doses from exposure to iodine after the Chernobyl accident, it is necessary to trace the dynamics of its accumulation on soil, which can be done by daily measurements of radioiodine depositions on the underlying surface. Such observations were conducted during the accident in question based on the constantly operating system of radiation monitoring of Roshydromet.

The samples of radioactive depositions were collected daily at meteorological stations using the methodology described in [1] and delivered quickly to SPA "Typhoon" to the laboratory of environmental radiation monitoring, in which they were first sorted by activity with a dosimeter and then measured for total β-activity in non-incinerated form. After that, the samples were compressed with a hydraulic press and without incineration were measured at a gamma-spectrometer, which permitted determination of content of volatile iodine radioisotopes and other radionuclides. Occasionally, the sample was measured some time later again. Then the samples were incinerated at 450 °C, their total β-activity was measured again and if necessary, gamma-spectrometric analysis was repeated after the decay of short-lived radionuclides. Radiochemical analysis of β- and α-emitting isotopes was performed selectively.

The total β-activity of short- and long-lived radionuclides was measured separately. In our work we used data on long-lived total β-activity of depositions from measurements made not earlier than the fifth day after the sampling. The calibration was conducted with $^{90}Sr + {}^{90}Y$.

When collecting radioiodine depositions from the atmosphere, the question arises about the efficiency of the sampling means which was a gauze collector. Earlier in [2] using a large body of statistical data, we determined the efficiency of capture of the aerosol fraction of global radioactive depositions by arranging a parallel sampling using a gauze collector and a high-wall collector the bottom of which was covered with a layer of distilled water with introduced stable isotopes-carriers. The efficiency of the high-wall collector was taken to be 100%. The comparison was done by measuring the content of ^{90}Sr, ^{137}Cs and long-lived total β-activity of radionuclides in samples. The efficiency of the gauze collector was determined to be 70% and this value was used in the calculations of radioactive depositions from the atmosphere [1, 3]. The efficient deposition rate for radioactive aerosols from the atmosphere V_g usually determined as a ratio of deposition rate f to volumetric concentration of aerosols in the air q was, on the average, 0.7 cm/s. The highest depositions and concentrations of ^{131}I in the air in Podmoskovje (Obninsk) with the dispersion of radioactive air masses from the Chernobyl accident area were reported on 30.04 - 01.05.1986: $f = 459$ Bq/(m^2·day) and $q = 0.669$ Bq/m^3 [4] which corresponds to $V_g = 0.8$ cm/s. This value is very similar to the above mentioned value of V_g for radioactive aerosols of global origin. This agreement seems to indicate that it is the aerosol fraction of ^{131}I which was picked up by the gauze collector.

Radioiodine isotopes occur in aerosol, molecular and organic forms. The interaction of theses physico-chemical forms of radioiodine with the underlying surface and their transformation during the atmospheric transport are still poorly understood. The ratio between the activities of the above radioiodine forms in the release from the damaged reactor was changing with time. While during the explosion a large amount of ^{131}I released was in the aerosol form, during the smothering with inert materials, the graphite stack burning and later on, the proportion of ^{131}I in the gaseous phase was to be increasing.

So, it follows from data of work [5] that at the height of 300 m above the damaged reactor the proportion of ^{131}I in the aerosol form was 69% on 8 May and on 29 August at 30 m above the reactor it was only 10%. At a distance of about 1000 km north of the Chernobyl NPP the proportion of the aerosol fraction of ^{131}I at 2 km height was 45-55% on 14-15 May, on 17 May - 24% and on 19 May - 10%. These individual measurements, however, do not mean that the proportion of the aerosol fraction of ^{131}I was changing everywhere monotonically from 55% to 10% from 14 to 19 May. In other areas this value could be different. On the part of the route of a research vessel from the English Channel to St.-Petersburg the proportion of the aerosol fraction was haphazardly changing within 10-27% from 4 to 11 May, in Vilnyus from 30 April to 10 May within 24-50% [6]. In the first days in Finland it was 15%, in France - 20-50%, in USA - 20-30%. Work [6] includes a summary of data collected in the first days after the accident in different areas of 11 countries of the north hemisphere, suggesting that on the average, the proportion of the aerosol fraction of ^{131}I in the near-surface layer was 25%.

In Vilnyus the aerosol, molecular and organic forms of ^{131}I were determined. The highest concentration of ^{131}I in the air was reported on 30 April, the percentage of the above forms being 24:22:54 and on the average from 30 April to 10 May - 35:14:51. The extreme values of the proportion of the aerosol form differed by a factor of 2, that of the molecular form by a factor of 3 and the organic form - only by 18%, i.e. the organic form was most stable. This is also confirmed by the low deposition rate of ^{131}I in the organic form on the underlying surface- when methyl iodide deposits on the grass surface V_g is 0.01 cm/s,

whereas the rate of deposition of elemental iodine on dry grass, averaged over the pasture period, is 2 cm/s and 3 cm/s on the wet grass [7]. Based on the ratio between the physico-chemical forms of ^{131}I observed in Vilnyus the weighted average deposition rate of total ^{131}I in dry weather will be 0.6 cm/s and in wet weather - 0.9 cm/s, which is close to the above value V_g = 0.8 cm/s for ^{131}I in Obninsk and V_g = 0.7 cm/s for radioactive aerosols of global origin.

Similar experimental data on effective deposition rates of ^{131}I observed after the accident and in global

sunny weather the deposited ^{131}I will evaporate from collector surface, while in the air it will pass to the aerosol and gaseous forms. The opposite sorption processes are also a possibility with the gaseous form being converted to the aerosol one. That is why, on individual days depending on weather conditions the inflow of ^{131}I from the air to the soil surface as well as soil and vegetation contamination density may vary significantly. Quantitatively, these effects have not been estimated, so the results of measuring radioiodine depositions from the air are approximate. In calculation of the accumulation of radioiodine on soil and doses, data are summed up over considerable time periods during which different weather conditions are observed. So, the indicated opposite processes of radioiodine migration are largely compensated and therefore, the results of calculating radioiodine accumulation on soil and doses are more reliable than the starting daily data on radioiodine depositions.

2. Verification and preparation of initial data on ^{131}I depositions from the air

The starting data for calculating ^{131}I accumulation on soil, dose rate and accumulated dose are daily ^{131}I depositions from the air shown in Table A1 (Attachment to the article).

We had at our disposal data on 71 observational points fitted with gauze collectors for collecting samples of daily radioactive depositions from the air. The collectors were exposed every day from 8 a.m to 8 a.m. the following day. For the cities in which there were less than 6 daily values of ^{131}I depositions (Kandalaksha, Orel, Bryansk and other) or data are not available for April - early May (Kaluga), the calculation was not done and these cities were not included in Table 1.

Some explanations to Table A1 of Attachment.

In some cases when the deposition sample was divided into two parts, in which ^{131}I content was determined separately, Table A1 contains both values - measurement 1 and measurement 2.

The same Table gives two values for the same exposure interval, if the same deposition sample was measured twice at different time. In doing this, the measured activity was, of course, assigned to the date of the collector exposure.

In those calculations where there were different possibilities of selecting the starting data, the preference was given to measurements with a gamma-

radioactive materials for which the efficiency of the gauze collector was determined allows us to use the value of this efficiency for ^{131}I in the Chernobyl depositions.

It should be remembered, however, that the dynamic equilibrium which sets in the atmosphere between the aerosol and gaseous forms of ^{131}I, is maintained only with space-time averaging. The local behaviour of ^{131}I in the air and ^{131}I deposited on the collector or soil and vegetation cover may change depending on weather conditions. In

spectrometer with a semiconductor detector. If results of measurements of two parts of the collector sample differed not more than by 10 Bq/m^2·day, an average rounded to ten was entered Table A1. If results differed by more than 10 Bq/m^2·day, both measurement results were entered the Table and in further calculations an arithmetic mean was used while the initial actual values of depositions were used to determine the range of possible values of calculated ^{131}I accumulation on soil or radiation doses. In those cases when data of direct measurements were scarce, the initial array was added with estimates of ^{131}I depositions from correlations with depositions of total β-activity, with depositions of volatile ^{137}Cs, with soil gamma-radiation dose rate etc.

We now consider in more detail the preparation of initial array of data for estimation of radiation doses. In Table A1 the mark "v" means that in the considered time interval direct measurements of^{131}I depositions are not available. The given value has been reconstructed with space-time averaging, i.e. with results of measured depositions in the nearest days in the given city with allowance for deposition value on the same day in the neighbouring cities. If there was no sharp increase or decrease in the deposition value, we take the average between the two values next to the missing values (rounded to hundred). If there are "jumps" in the value of significance, the smoothing is performed using daily maps of ^{131}I depositions by the closest isolines [4, 9, 10]. A possible error in such estimates does not exceed 50%. "Reconstruction" of data by depositions in the closest cities yields fairly good results. With low values of depositions the "reconstructed" data by several neighbouring cities are similar and do no go beyond the adopted spread of 50%, but what is most important is (due to smallness) that they do not affect significantly the final results of calculations (accumulated dose and spread in its values).

In those cases when the "reconstructed" values were close to maximum which was the case in three populated points: Baranovichi (27-28.04.86); Mariupol (1-2.05.86) and Sumy (30.04-1.05.86), their contribution to the results of calculation of the accumulated dose was significant and accordingly the spread increases, which is 30% for Baranovichi, for Mariupol - 17% and for Sumy - to 30%.

Let us consider these three cases in more detail. Table 1 shows ^{131}I daily depositions in Bq/m^2·day in

the above cities and some neighbouring ones. The question marks are gaps to be reconstructed.

By way of illustration Figures 1-3 show relations of ^{131}I depositions in Baranovichi and those in neighbouring points: Pinsk, Grodno and Minsk. The measured values are dots and the ranges of possible spread and reconstructed values are dashes. These plots were used to "reconstruct" ^{131}I depositions in Baranovichi: for Pinsk - 500, Grodno - 30 and Minsk - 4 kBq/m^2·day. By averaging these three values we get the ^{131}I deposition in Baranovichi of 268 Bq/m^2·day. On the daily maps of atmospheric depositions of ^{131}I Baranovichi lies between the outlines of 100 and 500 kBq/m^2·day, Sumy - not far from the outline 1 kBq/m^2 ·day; in the area of Mariupol no outlines were drawn and in the closest cities at that time a uniform decrease or slight increase in ^{131}I depositions was observed. Thus, the atmospheric depositions of ^{131}I on 27-28 April were: in Baranovichi 250∓125 kBq/m^2·day (spread of 50%); 1-2 May - in Mariupol - 10∓2 kBq/m^2·day (spread of 20%); on 30 April-1 May in Sumy - 20∓10kBq/m^2·day (spread of 50%).

For some cities there are data of ^{131}I depositions measurements in several points: within the city and in the vicinity. Let us consider such cases.

In Vilnyus the observations were conducted in the city itself and in the point Vilnyus-Zagorodny. Table A1 (Attachment) contains both values. In calculations, measurements in Vilnyus-Zagorodny were used as a second value and for those days on which observational data within the city were non-existent they were used as the only value for Vilnyus. Results of observations of ^{131}I depositions in St.-Petersburg, Moscow and Odessa and their vicinities are presented in Table 2. This Table also includes the first and second values of depositions used. The first value was result obtained with a gamma-spectrometer with a semiconductor detector (if available) If such results are more than two, an arithmetical average was taken. The second value was the maximum of all available results for a day under consideration.

For Odessa we primarily used data for the point Odessa - GMO. These data on individual days were in discrepancy with those for the points Odessa-Zavodskaya and Odessa-Chernomorskaya, which was taken into account in estimating the spread in calculated radiation doses. This issue is further discussed in the following section. Data of direct measurements of ^{131}I depositions in Odessa are not available after 7 May and Table 2 is added with data obtained by averaging of reconstructed deposition values for the closest cities: Izmail, Nikolaev and Kherson.

Table 1

Daily depositions of ^{131}I (Bq/m^2 · day) in selected cities of CIS with data missing in some time intervals

Populated point	Time interval, 1986		
	26-27.04	27-28.04	28-29.04
Baranovichi	26143	?	163181
Pinsk	2775	750000	-
Grodno	-	100341	68400
Minsk	47	2400	24021
Brest	15150	113736	-
	30.04-1.05	1-2.05	2-3.05.
Mariupol	8898	?	12000
Donetsk	6768	4380	4539
Zaporozhje	20300	4180	1562
Genichevsk	4470	5738	2387
Kerch	2892	9033	4715
	29-30.04	30.04-1.05	1.05-2.05.
Sumy	878	?	2358
Poltava	788	21000	2465
Kharkov	290	12970	-

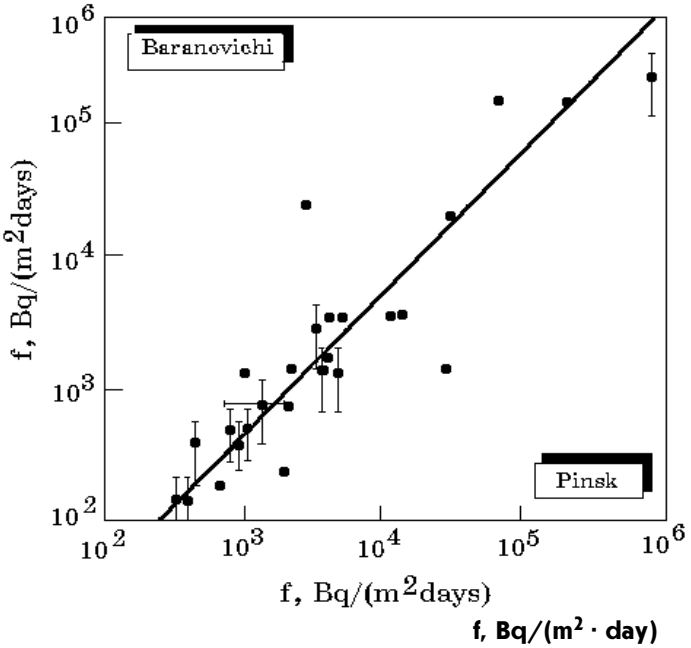

Fig. 1. Comparison of ^{131}I depositions in Baranovichi and Pinsk. Points - measurements; dashes - ranges of possible spread in measurements and reconstructed values.

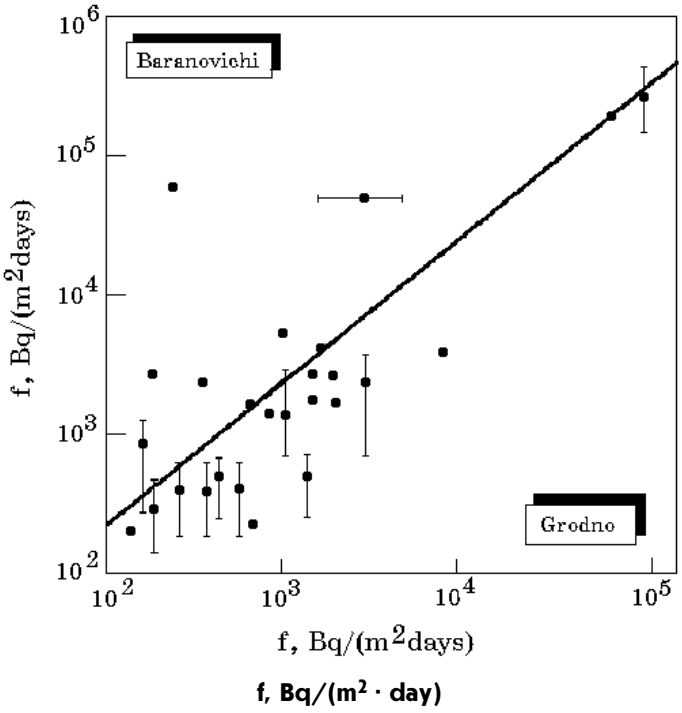

Fig. 2. Comparison of ^{131}I depositions in Baranovichi and Grodno. Points - measurements; dashes - ranges of possible spread in measurements and reconstructed values.

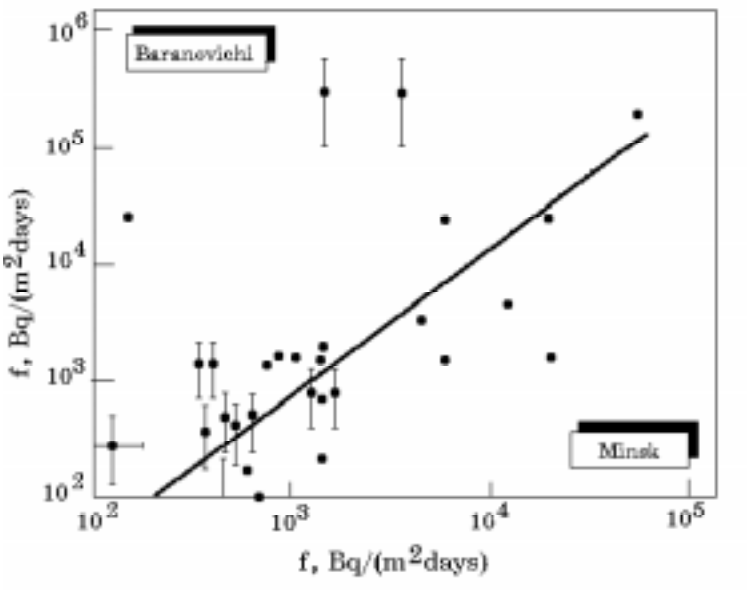

Fig. 3. Comparison of ^{131}I depositions in Baranovichi and Minsk.
Points - measurements; dashes - ranges of possible spread
in measurements and reconstructed values.

Table 2

An example of selection ^{131}I daily depositions used in radiation dose estimation for cities with several observational points

Observational point	25-26	26-27	27-28	28-29	29-30	30-01	01-02	02-03	03-04	04-05	05-06	06-07	07-08
Moscow													
	April 1986						May 1986						
Balchug	0	0	0	365		617	41	2709					
Lenino-Dachnoe						1975	101	1438					
EEA					1070		32	2385					
CHS							79	2788					
Novo-Ierusalim											98	61	66
1-st value	0	0	0	365	1070	1975	51	2144	60	50	98	61	66
2-nd value						617	101	2788	180v	150v			
Odessa													
GMO	11c		131c	178	609	14978	37949	17882			656c	138c	184c
Zavodskaya	2849	128c	107c	123c	120c		3975c	65c	4671c	1773c	137c		
Chernomorskaya	820	16c	102c	32	106c	265c	5095c	15615	3484	940	259c		43c
Chernomorskaya								15638	4616	1448			
1-st value	2849	128c	131c	178	609	14978	37949	17882	4050	1387	656c	138c	184c
2-nd value	820	16c	107c	123c	110c	265c	3975c	7000	4671c	1773c	200c		43c
Sankt-Petersburg													
St.-Petersburg				384	4709								
Nevskaya						136c				61			
ITsP				544c	7953	151				247			
AMTs			275		4916		2410						
Lodeynoe Pole					4280								
1-st value	5	15	275	384	4859	151	2410	250	50	247	100b		
2-nd value	15v	45v		544c	7953	136c		750v		61			

In Moscow deposition samples were collected downtown near the hotel "Balchug" at the Central helicopter station (CHS) in the territory of the Central Air Terminal between the metro stations Airport and Dynamo and in the territory of Exhibition of Economic Achievements (EEA), in the proximity of the ring road in the point Lenino-Dachnoe and in the settlement Novo-Ierusalim. The data in Table 2 for these points are results of sample analysis performed in SPA "Typhoon" in Obninsk. Unfortunately, in the following days measurements of ^{131}I content in depositions samples collected in these points were terminated because they were passed over to be measured in Institute of Applied Geophysics (IAG) located in Moscow. The decision was, in principle, proper and aimed at shortening the time of delivery of samples for measurements, but given the post-accidental confusion, it led to breaking the established schedule and a result IAG measured only concentration of ^{131}I in the air at CHS, while deposition samples were not measured. For this reason, Table 2 for Moscow includes either data on ^{131}I depositions in Novo-Ierusalim which continued to be delivered for measurements to Obninsk or data calculated with ^{131}I concentrations in Moscow from the ratio between ^{131}I depositions and concentrations for the same day in Obninsk (100 km from Moscow). The values of ^{131}I for Moscow estimated in this way are marked in Table A1 (Attachment) with letter "k".

For most cities the acceptable length of series of continuous observations of ^{131}I from the atmosphere together with individual reconstructed values is available for the period of highest values of depositions from 26 April to 7 May 1986. In separate points longer observational series are available, for example, for Kiev - to 13 May, for Minsk - to 16 May, for Obninsk - to 19 June (after this date ^{131}I depositions were below the sensitivity threshold of the methodology). However, for some cities the data series were shorter. For example, for Lugansk there are 9 values of depositions (of them 1 is "reconstructed"), for St.-Petersburg -10 (3 - "reconstructed"). These are the shortest data series, but even they include the highest values of depositions, which allows estimation of accumulation of ^{131}I on the soil and dose from gamma exposure, since later on the iodine decays quickly. For more accurate calculation the data on daily ^{131}I depositions should be extended to include the following months till the complete decay of the radionuclide. This was done by one of the methods described below or their combination.

In Table A1 (Attachment) in column "Type" the symbol "b" designates data on ^{131}I depositions calculated from the value of depositions of total β-activity in the same observational point. For this purpose, for separate regions (for example, Baltic areas, Podmoskovje, Pricarpatje etc.) selected with consideration of their location and distance to the Chernobyl NPP, we constructed correlations of ^{131}I depositions and depositions of total β-activity in each point of the region. An example of such correlation for Gomel is given in Figure 4. Before measurement of the total β-activity the sample was incinerated, whereas ^{131}I was determined in the unincinerated sample.

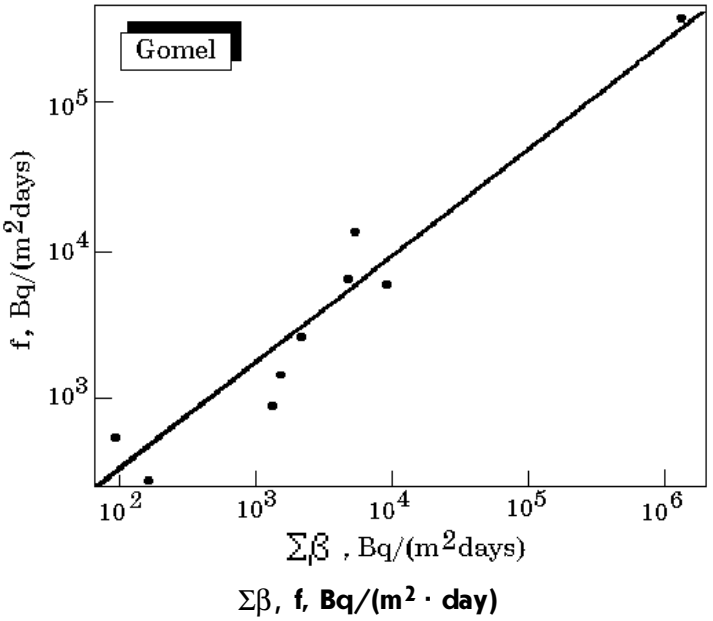

Fig. 4. Comparison of ^{131}I daily depositions (the ordinate axis) and total β-activity in Gomel.

By similar plots constructed for separate cities or groups of cities forming a region it was possible to reconstruct the value of ^{131}I depositions in the time period from mid-May to mid-June. In this region, the relation between the ^{131}I depositions and total β-activity in the bilogarithmic scale was almost linear. In the region of small values (less than 50-200 Bq/m^2·day) depending on the distance from the accident point there were deviations from the linear relationship. This region was not included in our calculations and is not shown in Figure 4.

Whereas the time of delivery of different deposition samples for measurements was varying in a wide range, the relation between ^{131}I depositions and total β-activity was impossible to be determined because of a significant spread of points. A wide spread was also observed in the region of low values of the activity due to the increased measurement error.

The relation between daily depositions of ^{131}I and ^{137}Cs depositions from the air was much less obvious. The attempts to establish a similar relation with ^{90}Sr depositions were to no avail.

In addition to the mentioned reasons, the relation between ^{131}I depositions and total β-activity depends on weather conditions. That is why, when possible, results of reconstruction of ^{131}I depositions from total β-activity were corrected. Such data in "Type" column of Table A1 (Attachment) are marked by symbol "bp".

The samples from some cities after 10 May to 1 June were combined over 5-10 consecutive days prior to ^{131}I determination. For example, samples from Klaipeda: from 11 May to 20 May; from Baltiysk - from 11 to 15 May; from Vilnyus - from 11 to 20 May and from 21 to 30 May. In order one could estimate daily ^{131}I depositions from five-day or ten-day depositions of ^{131}I the resulting activity value was distributed among the days included in the averaging in proportion to daily depositions of total β-activity. With this, the value of ratio of these depositions was within $^{131}I/\Sigma\beta = 0.5\text{-}1$. The ^{131}I daily depositions derived in this way appeared in some cases 2-4 times lower those derived from the plots similar to the one shown in Figure 4 because the resulting regression dependence was mainly affected by measurements of the samples collected prior to 10 May when there were more "short-lived" radionuclides in the atmosphere. Therefore, in all the cases when after 14 May ^{131}I depositions were estimated from the dependence similar to that shown in Figure 4 and the relation $^{131}I/\Sigma\beta > 1$ was derived, the corresponding values of depositions were reduced so that the condition be met: $^{131}I/\Sigma\beta = 0.5\text{-}1$. If within a ten day period the whole series of ^{131}I depositions meets this condition and 1-3 values were slightly higher, they were used without any changes.

The reconstructed values of ^{131}I depositions without adjustment were not rejected in further analysis, but were used for estimation of maximum possible value of accumulation of ^{131}I on the soil and radiation dose.

Determination of ^{131}I from depositions of total β-activity was used only to the end of May 1986 when the levels of β-activity were high and the jumps in the depositions still occurred. At low levels of β-activity and when impossible to reconstruct ^{131}I depositions

from the regression dependence, the further series of daily depositions was calculated under the assumption of exponential decrease in ^{131}I depositions. These kind of data are not given in Table A1. After 1 June all ^{131}I depositions were estimated only by the exponential law. In doing this, separate large values of total β-activity of depositions samples were not taken into account as they could be due to "hot" particles which got on the collector during dust resuspension from the soil surface and do not contain volatile ^{131}I.

When estimating ^{131}I depositions by the exponential law we discriminated two time intervals: approximately from 15 May to 1 June and from 1 June to 1 September 1986.

For the first period we assumed that the influence of self-cleaning of the atmosphere on the time decrease in ^{131}I depositions, in the first approximation, can be neglected (the data obtained by the exponential law were close to those derived from depositions of total β-activity with a correction). The reason for this assumption was that at that period ^{131}I releases from the damaged reactor continued, which, to a certain extent, made up for the loss of this isotope in the atmosphere due to its self-cleaning. It may be assumed that during this period the decrease in ^{131}I atmospheric depositions occurred in accordance with the law of radioactive decay.

In the second period (from 1 June to 1 September 1986) the self-cleaning of the atmosphere from ^{131}I was taken into account. The period of half-life of passive material from the surface atmosphere is, on the average, 7 days [11] with the removal constant λ_0. Considering the radioactive decay with the constant λ, the effective constant λ_1 of ^{131}I decrease in the near-surface atmosphere in this case can be determined with an obvious formula: $\lambda_1 = \lambda_0 + \lambda$.

In the first period, when filling in gaps in the data, as the most probable were taken ^{131}I depositions calculated by the exponential law with allowance for the radioactive decay only. Depositions estimated for this period by the exponential law with consideration of decay and self-cleaning were used as a possible lower bound.

In the second period, when filling in gaps in the data, as the most probable were taken, on the contrary, ^{131}I depositions calculated by the exponential law with allowance for atmospheric self-cleaning. Depositions estimated for this period by the exponential law without consideration of self-cleaning were used as a possible upper bound.

After such preliminary processing of the starting array of data we estimated possible doses from radioiodine for the population of 43 cities of the European territory of the former USSR (ETU).

3. Accumulation of ^{131}I on the soil surface

After the Chernobyl accident external doses in ETU was formed primarily due to gamma-radiation of radioactive materials dispersed in the area. The gamma-irradiation of technogenic radioactive materials always occurs against the background of gamma-irradiation of natural radioactive elements occurring in the soil and mountain rocks and ionizing component of the space radiation. For the flatland part of the former USSR this total natural "gamma-background" is, on the average, 8.7 μR/hour, it is about 12 μR/hour for mountain area; up to 30-60 μR/hour for high mountains and up to 100 μR/hour for high mountain peaks.

Prior to the Chernobyl accident the dose rate from gamma-irradiation of technogenic radionuclides occurring in the soil was primarily due to irradiation of ^{137}Cs accumulated from atmospheric depositions of products of nuclear explosions and the country-averaged value was 0.24 μR/hour. Since this value could not be measured directly in the presence of natural gamma-radiation, this estimate was derived using the methodology described in [12] based on data about ^{137}Cs in soil (partially published in [3, 13]), its vertical distribution along the soil profile with allowance for the influence of arable lands, screening of gamma-radiation by snow cover and contribution of different geographic zones. So, prior to the Chernobyl accident the contribution of technogenic radioactive materials to the total gamma-radiation dose rate, averaged over the country, was negligibly small.

In the first month after the accident the main contribution to formation of external dose was made by short-lived radioisotopes: ^{140}Ba+^{140}La, ^{132}Te+^{132}I and ^{131}I. The external dose from ^{131}I radiation from the soil surface can be calculated by accumulation of its total depositions on the surface of soil and vegetation cover. Concurrent with the ^{131}I inflow from the atmosphere to the soil surface, the radioactive decay of the radionuclide, its wash-off with surface waters, migration down the soil profile, evaporation from the surface of soil and vegetation cover to the atmosphere, removal with the yield and other processes occur. In the first approximation we consider only ^{131}I depositions from the atmosphere and its radioactive decay, neglecting secondary effects, i.e. we derive the upper estimate of the radionuclide accumulation. The formula for calculating the ^{131}I accumulation on the surface of soil and vegetation cover P from measurements of daily depositions of this isotope from the atmosphere f can be written as

$$P_i = P_{i-1} \cdot e^{-\lambda \cdot \Delta t} + f_i \cdot \Delta t, \qquad (1)$$

where P_i, P_{i-1} is accumulation on soil of ^{131}I depositions by the end of the i-th, $(i-1)$-th days;

f_i is ^{131}I deposition from the atmosphere for the i-th day;

Δt is collector exposure time (1 day);

λ is ^{131}I radioactive decay constant.

Formula (1) describes time integration of depositions with step Δt, which allows tracing the dynamics of ^{131}I accumulation on the soil surface. By way of illustration Figure 5 shows observational data on ^{131}I depositions from the atmosphere in Gomel (Belarus) and Obninsk (Podmoskovje). Figure 6 presents outlines of ^{131}I accumulation on the surface of soil and

vegetation cover calculated by these data; for comparison, it also shows accumulation curves for Kiev and Kaunas.

In the Attachment to the article Table A1 and Figure A1 present data on the dynamics of ^{131}I depositions and accumulation on soil surface in each of 43 cities of the former USSR.

It can be seen from Figure 6 that the maximum soil contamination with ^{131}I in the indicated points was observed at different times from 28 April to 3 May. Summing up data on accumulation of ^{131}I depositions in each individual point enabled construction of the first map of radioactive contamination of the European territory of the former USSR with this isotope [4]. Figure 7 presents a map of ^{131}I contamination of soil surface by 15 May 1986. In the territories where ^{131}I depositions were not measured: east of Sumy-Bryansk - the ^{131}I deposition outlines were drawn which are estimated from ^{137}Cs depositions using an empirical formula constructed with data of Figure 8:

$$P_I = 15.03 \cdot \Delta P_{Cs}^{1.14}, \quad (2)$$

at $0.01 < \Delta P_{Cs} < 1$ Ci/km^2,

where P_I is accumulation of ^{131}I depositions, Ci/km^2 as of 15 May 1986;

ΔP_{Cs} is accumulation of ^{137}Cs depositions due to the Chernobyl accident, Ci/km^2. In calculations we used the ^{137}Cs global background contamination level for the ETU - 0.056 Ci/km^2 [13].

When constructing the map, in those cases when the collectors network was too sparse, measurements of gamma-radiation dose rate from the soil surface were used for interpolation. The highest contamination with ^{131}I was reported in the north Ukraine, east Belarus and adjacent areas of central regions of Russia. Separate contamination "spots" were found near the west border of Russia in the vicinity of Baltiysk-Kaliningrad and in the area between Gomel and Bryansk and south of Tula. The presence of such "spots" is explained by localized zones of atmospheric precipitation during dispersion of radioactive clouds and spatial heterogeneity of radioactive air masses in the atmosphere.

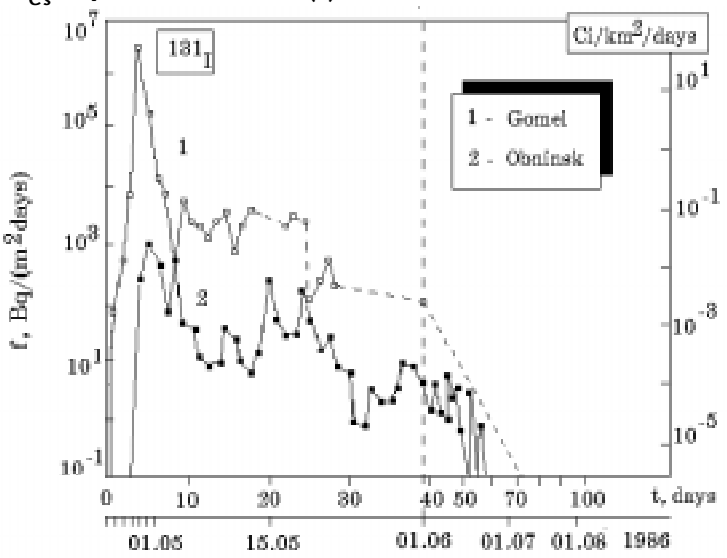

Fig. 5. Dependence of ^{131}I daily depositions from the atmosphere on the soil surface on time t after the Chernobyl accident for Gomel (Belarus) and Obninsk (Russia).

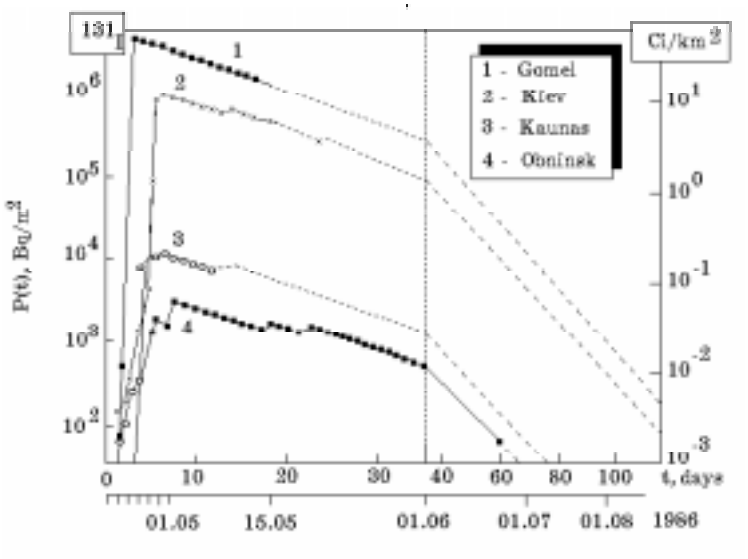

Fig. 6. Dynamics of ^{131}I accumulation on the soil surface in some cities of the former USSR.

To define the spatial distribution of ^{131}I in the territory of CIS we also used measurements of ^{131}I and ^{137}Cs contamination density in soil samples (about 100) which were collected in May-June 1986 by SPA "Typhoon". The correlation coefficient between densities of soil contamination with the accidental ^{131}I and ^{137}Cs, including data of calculations of cumulative depositions, was 0.91 in the region $0.01 < \Delta P_{Cs} <$ 200 Ci/km^2. The high correlation coefficient gives grounds to construct a regression model similar to (2) permitting estimation of ^{131}I soil contamination density by 15 May 1986 based on data about ^{137}Cs contamination [14, 16]:

$$P_I = 3.77 \cdot \Delta P_{Cs}^{0.847}, \qquad (3)$$

at $0.01 < \Delta P_{Cs} < 200$ Ci/km^2.

As can be seen, relation (2) in the range of contamination density values ΔP_{Cs} - 0.01-1 Ci/km^2 can be considered as an upper estimate of the ^{131}I contamination density, as the ratio $P_I(2)/P_I(3)$ grows from 1 to 3.9 with increase of ΔP_{Cs} in the considered range. The location of the outlines on the map shown in Figure 7 ($P_I < 5$ Ci/km^2), however, has not changed in the used scale.

It should also be noted that the closeness of the power to unity in formulae (2) and (3) justifies to a certain extent estimation of ^{131}I soil contamination by ^{137}Cs content using linear relations [17].

Figure 9 presents outlines of ^{131}I contamination density - 4 Ci/km^2 by 15 May 1986 in the territory of the former USSR calculated with formula (3) using the latest results of aerial spectrometric survey of ^{137}Cs contamination density, results of which are published in [18].

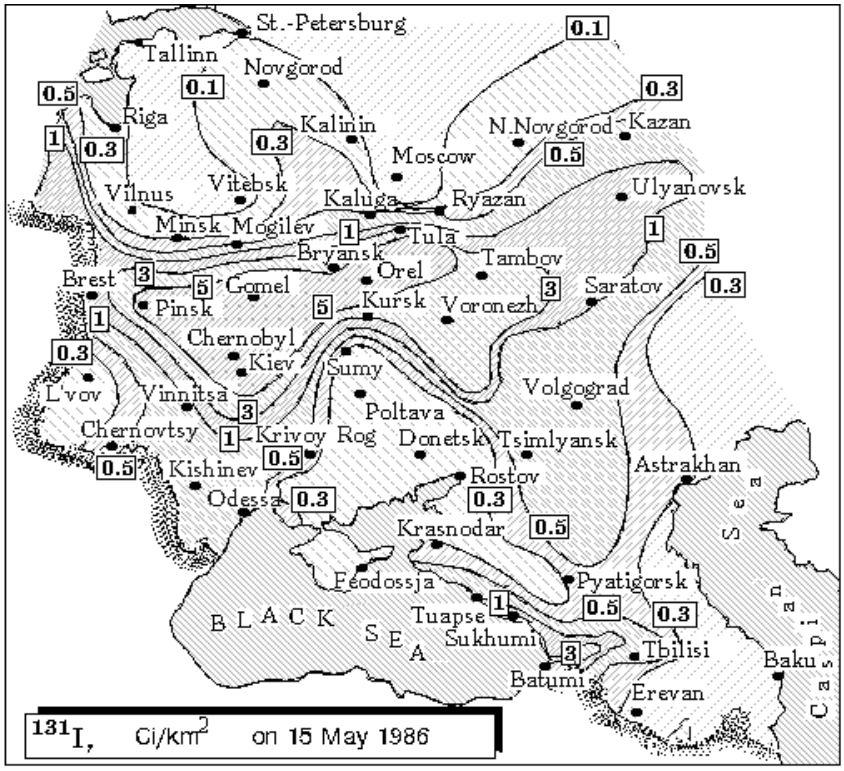

Fig. 7. Map of ^{131}I contamination of the territory of ETU constructed based on measurements of ^{131}I depositions on collectors and results of reconstruction from ^{137}Cs depositions - outlines east of Sumy-Bryansk.

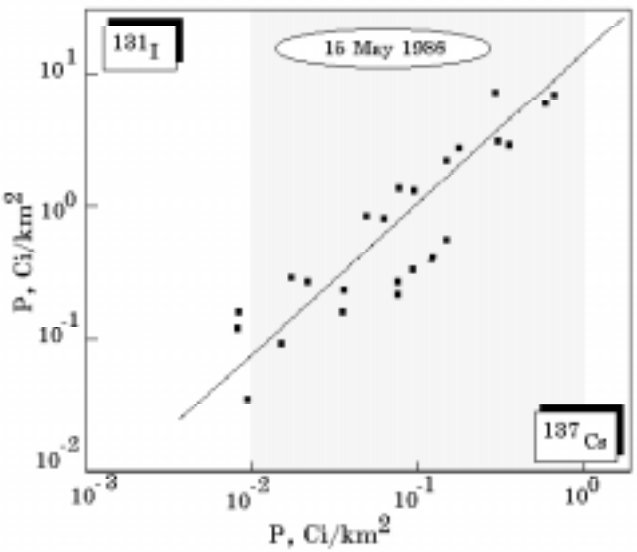

Fig. 8. Comparison of accumulation of depositions of ^{131}I (as of 15 May 1986) on the soil surface - (the ordinate) and accumulation of the accidental ^{137}Cs - (the abscissa).
The watched area is the area in which relation (2) is applicable.

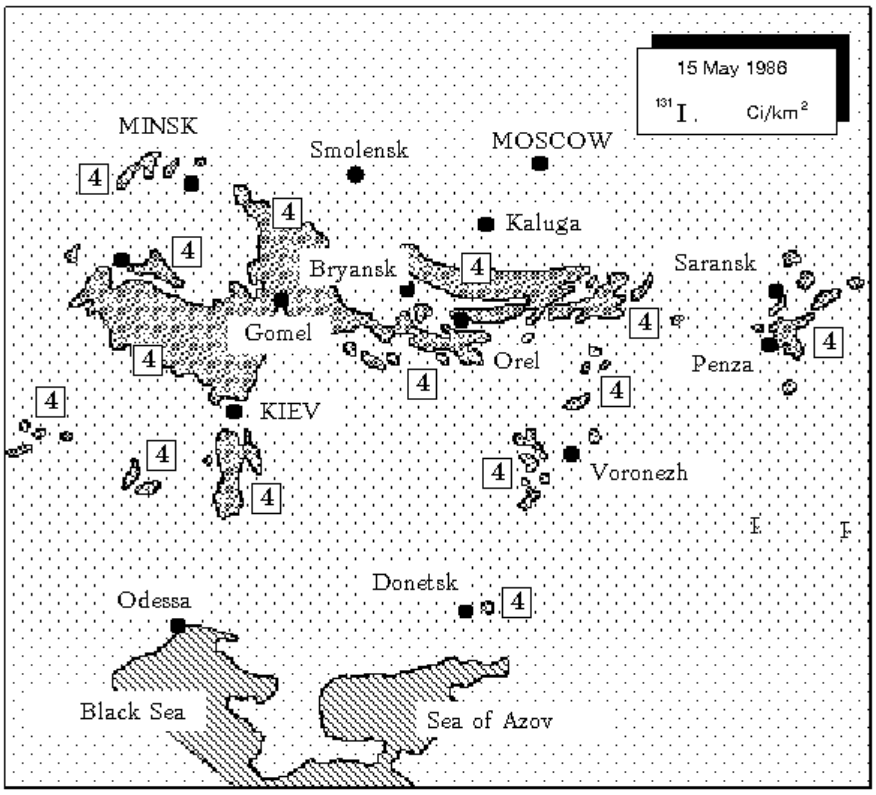

Fig. 9. Position of the outline of ^{131}I contamination density - 4 Ci/km^2 (as of 15 May 1986) reconstructed by relation (3) using data of aerial gamma-spectrometric determination of ^{137}Cs soil contamination density (as of 1 January 1993).

The comparison of Figures 7 and 9 suggests a fairly good general agreement in the density outlines of ^{131}I contamination, in spite of the fact that Figure 9 is more detailed. Because of a relatively sparse network of meteostations, the map in Figure 7 constructed with data on ^{131}I atmospheric depositions is rather general. Using data on depositions from a sparse network of meteostations does not make it possible to identify the fine pattern of the contamination. For construction of a detailed map, in May-June 1986 a comprehensive aircraft gamma-spectrometric survey should have been arranged with small distances between routes and land reference routes with sampling and determination of ^{131}I. However, such detailed mapping of soil contamination with short-lived ^{131}I to the complete decay of the radionuclide was not possible.

When constructing the ^{131}I contamination maps, data on the radionuclide content in the soil obtained at different times were recalculated by the law of radioactive decay to the same date - 15 May 1986. This date was chosen because by this time the soil contamination with ^{131}I deposited from the atmosphere had been mostly formed, i.e. the contributions of the following depositions to the soil contamination can be neglected. The recalculation of depositions accumulated prior to 15 May to the indicated date results in underestimation due to continuing ^{131}I releases to the atmosphere and after 15 May - on the opposite, overestimation, as ^{131}I went on to deposit from the atmosphere to the soil.

A relative error in estimating the true soil contamination P_0 at time moment t_0 (15 May 1986) from measurements of ^{131}I content in soil P_t at time moment t can be calculated by the formula:

$$\frac{\Delta P}{P_0} = 1 - \frac{P_t}{P_0} \cdot e^{-\lambda \cdot t}, \qquad (4)$$

where $\Delta P = P_t - P_0$; λ is ^{131}I decay constant. The time interval t was counted off from the time of the accident.

Depending on the type of changes in ^{131}I depositions with time in different geographical points the considered error can differ significantly. For example, Figure 10 shows outlines of change in relative error (4) for Gomel, Brest and Minsk depending on time of soil sampling when recalculation of ^{131}I activity by 15 May by the radioactive decay law is done without consideration of factual depositions of this radionuclide from the atmosphere.

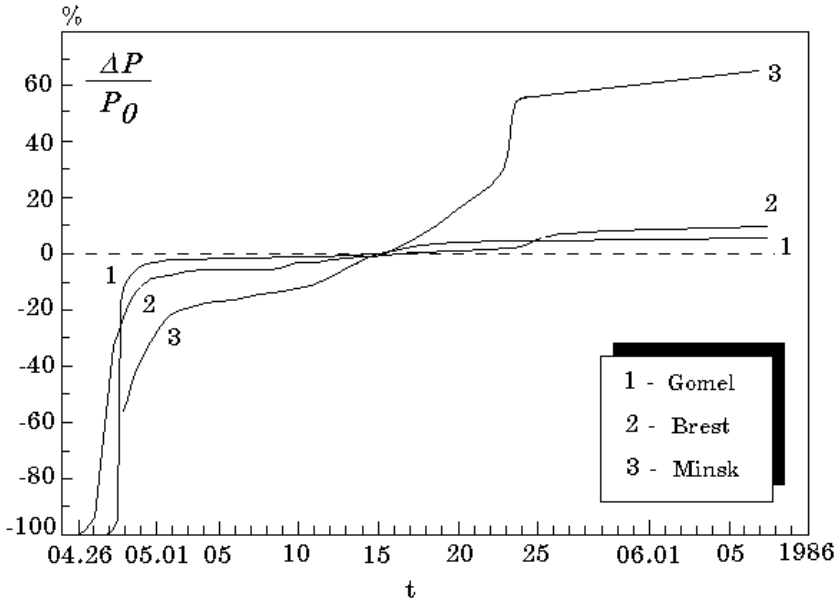

Fig. 10. Relative error in referring the ^{131}I soil contamination density to 15 May 1986 by the law of radioactive decay depending on date of soil sampling.

In calculations with formula (4) the density P_0 of the soil contamination was taken to be equal to the measured cumulative depositions of ^{131}I. It can be seen from Figure 10 that in May the considered relative error of recalculation did not exceed several percents for Gomel, 10% for Brest, but was rather high for Minsk - as high as 60% [14].

4. Radiation dose from ^{131}I gamma-irradiation

Gamma-irradiation of ^{131}I accumulating on the soil surface every past day leads to dose rate R_i (μR/day) in the air at the height 1 m above the ground which can be calculated by the formula:

$$R_i = k \cdot P_i , \qquad (5)$$

where k is the coefficient of conversion from ^{131}I surface contamination density to the air radiation dose at the height 1 m.

For "short-lived" ^{131}I it can be assumed that after the accident the bulk of the isotope was deposited on the grass and in the upper surface soil layer, i.e. the contamination was film type. For an infinite uniformly contaminated plane the factor k in (5) for ^{131}I is 6.8 (μR/hour)(Ci/km^2) [19]. As always the population migrates, we decided to sum up doses accumulated for each day spent in the given day and not use the value of the dose factor cited in [8].

Because ^{131}I decays almost completely with time, those living subsequently in previously contaminated areas are not exposed to further a increase in the accumulated external dose from ^{131}I. Table 3 gives the date on which external dose after the Chernobyl accident from ^{131}I gamma-radiation from soil surface was 90% of the dose accumulated by 1 September 1986. By way of demonstration, Figure 11 shows the outlines of accumulation of dose from ^{131}I gamma-radiation for Gomel and Kiev. It can be clearly seen that the outlines reach saturation at the end of June and as soon as July-August the accumulated dose does not increase. The 90% level of the external dose was reached by the end of May 1986 and it does not go beyond the date of 3 June 1986 in any of the considered population points.

A possible spread in the calculated external doses depended on the initial array of data on ^{131}I depositions from the air and was determined in the following way.

As was mentioned in the previous section, a critical analysis of measurements was performed for each city and three continuous series of ^{131}I depositions were constructed comprising the most probable maximum and minimum values. For each of these series the accumulation of dose with time was estimated. Table 4 below presents accumulated external doses starting from the accident time to 1 September 1986 calculated from the most probable deposition values and doses as of 1 June and 1 September calculated by maximum and minimum values of the depositions series.

Table 3

Dates (1986) by which accumulated gamma-radiation dose from ^{131}I depositions on the soil surface reached 90% of the dose accumulated by 1 September 1986

Populated point	Date	Populated point	Date	Populated point	Date
Baltiysk	25.05	Ivano-Frankovsk	02.06	Moscow	30.05
Baranovichi	24.05	Izmail	30.05	Nikolaev	01.06
Baryshevka	31.05	Kaliningrad	24.05	Obninsk	02.06
Brest	25.05	Kaunas	28.05	Odessa	29.05
St.-Petersburg	29.05	Kherson	31.05	Pinsk	24.05
Cherkassy	29.05	Kerch	31.05	Poltava	30.05
Chernovtsy	01.06	Kiev	30.05	Riga	30.05
Dnepropetrovsk	29.05	Klaipeda	25.05	Sumy	31.05
Donetsk	29.05	Krivoy Rog	29.05	Uzhgorod	02.06
Dukshtas	01.06	Lugansk	01.06	Vilnyus	29.05
Feodosiya	30.05	Lutsk	26.05	Vinnitsa	31.05
Genichevsk	29.05	Lvov	02.06	Vitebsk	03.06
Gomel	25.05	Mariupol	30.05	Zaporozhje	28.05
Grodno	25.05	Minsk	31.05		
Kharkov	30.05	Mogilev	27.05		

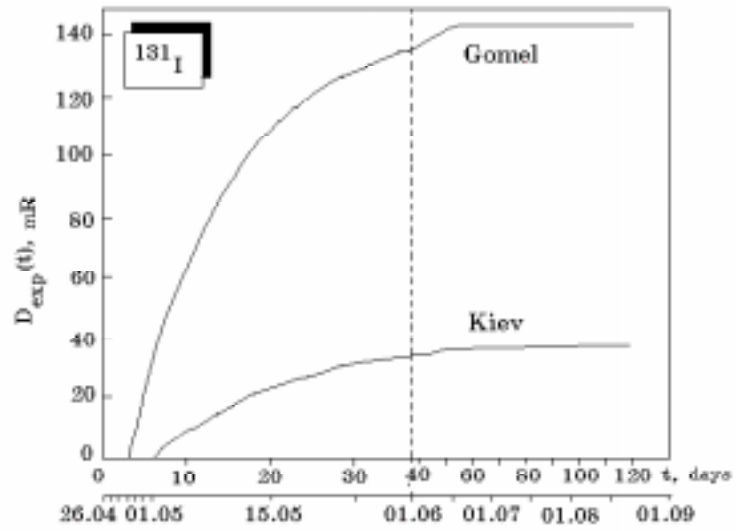

Fig. 11. Accumulation of ^{131}I gamma-radiation dose as a function of time by measurements of depositions at meteorological stations in Gomel and Kiev.

For most cities the spread in the calculated value of external dose did not exceed 20% and only in individual cases was more than 20%. It should be emphasized that the presented estimates should be treated with caution because the ^{131}I daily depositions from the air used in the calculations, strictly speaking, refer only to the meteorological site where they were observed. The above mentioned example of observations in Odessa indicates that the spatial fluctuations of the value could be significant when the rainfall pattern within a city was spotty. The doses calculated for the first 13 days for Odessa - GMO and Odessa Zavodskaya differ almost by a factor of 5. Based on the concept of the worst case selection from the observational series in dose calculation , we took the data for Odessa - GMO as most probable and the rest were considered in estimating the spread in dose values. Therefore, for Odessa the spread in radiation dose accumulated by 1 September has been estimated to be 70%.

In estimation of radiation effects of ^{131}I exposure on man, it should be remembered that we did not take into account behaviour, in particular, the screening effects of dwellings and working premises. To that extent , the calculation results should be considered as an upper estimate, as has been said above.

Our calculations suggest that of the cities in which observations of ^{131}I depositions were conducted at meteorological stations, the highest levels of the radionuclide gamma-radiation from the soil surface was found in Gomel (Belarus). The radiation dose accumulated by 1 June in this city was 134 mR and by 1

September - 142 mR, but it did not increase later on. The latter value corresponds to the effective dose of 0.135 cSv, with the dose limit of the total external and internal irradiation for a limited contingent of the population (category B) being 0.5 cSv for the calendar year [20] or 23% of the dose limit for ^{131}I.

Let us now consider the spatial distribution of external doses from ^{131}I dose radiation from the soil surface contaminated after the Chernobyl accident.

We use the calculations of accumulated external dose D in individual points of the country to construct a distribution map. For those areas in which ^{131}I in the atmospheric depositions was not measured we use the relation between the dose from ^{131}I gamma-irradiation from the soil surface and ^{137}Cs soil contamination density ΔP_{Cs} shown in Figure 12.

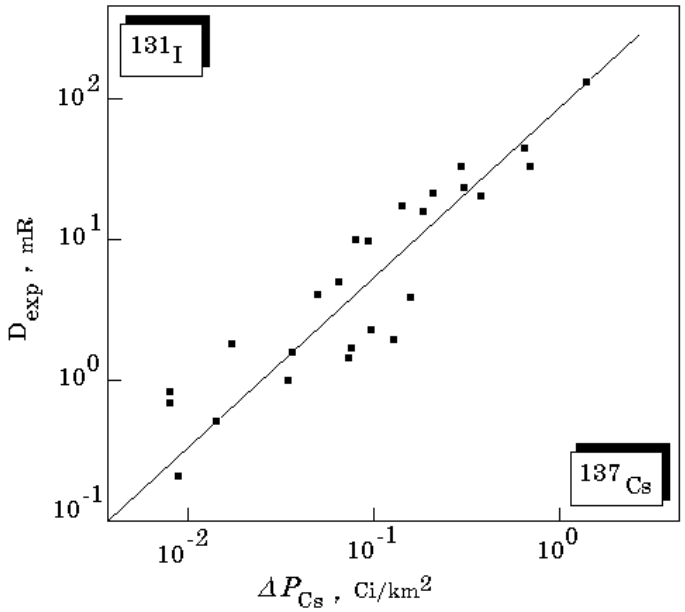

Fig. 12. Comparison of estimated external doses from ^{131}I and the accidental ^{137}Cs soil contamination density (the abscissa axis).

The empirical formula for calculating the exponential dose from ^{131}I gamma-irradiation from the soil surface D_{exp} from accumulation of the accidental ^{137}Cs on the soil has the form:

$$D_{exp} = 113 \cdot \Delta P_{Cs}^{1.26}, \text{ mR} \quad (6)$$

at $0.01 < \Delta P_{Cs} < 1$ Ci/km^2.

Figure 13 shows a map of distribution of radiation doses from ^{131}I gamma-radiation from the soil surface accumulated by 1 September 1986 for the ETU and Zakavkazje after the Chernobyl accident. As was said before, these doses primarily formed at the end of May - beginning of June and increased very little later on. As is in Figure 7, the isodoses east of Sumy-Bryansk have been reconstructed by ^{137}Cs soil contamination density. As can be seen from the present data by absolute value the external doses from ^{131}I are not high as compared to the natural gamma-background of the area and do not present a hazard for human health.

It should be noted that the isodoses in Figure 13 have been smoothed significantly in construction of the map. They just give an idea of the dose distribution pattern in ETU. A more detailed consideration of the distribution of the external dose from ^{131}I for separate areas and Kaluga and Bryansk regions (with resolution of 0.5 km) described in our work [15, 21, 22] suggests that the value in question is characterized by considerable spatial fluctuations. Outside populated points this is mostly associated with spatial heterogeneity of the radioactive deposition field. In the cities, the contamination is largely determined by current economic activities resulting in redistribution of this contamination with time and hence gamma-radiation field density within the city. Due to this, the reliability of detailed estimates of population doses in different parts of the city becomes lower and recalculation of the ^{137}Cs soil contamination density measured in the latest years to the ^{131}I soil contamination density in 1986 becomes questionable, since the extent of further anthropogenic impact on the initial soil contamination with ^{131}I is usually unknown. In work [22] for the communities of Klintsy of Bryansk, a significant heterogeneity of ^{137}Cs depositions field has been established. It appeared that the high contamination of

one place was due to the fact that this had been a storage site for peat transported from a highly contaminated areas. There is no way to determine now the amount of peat, the extent of radioactive contamination and the time of its transportation. For this reason, we did not construct a map of doses from ^{131}I for the territory of Klintsy.

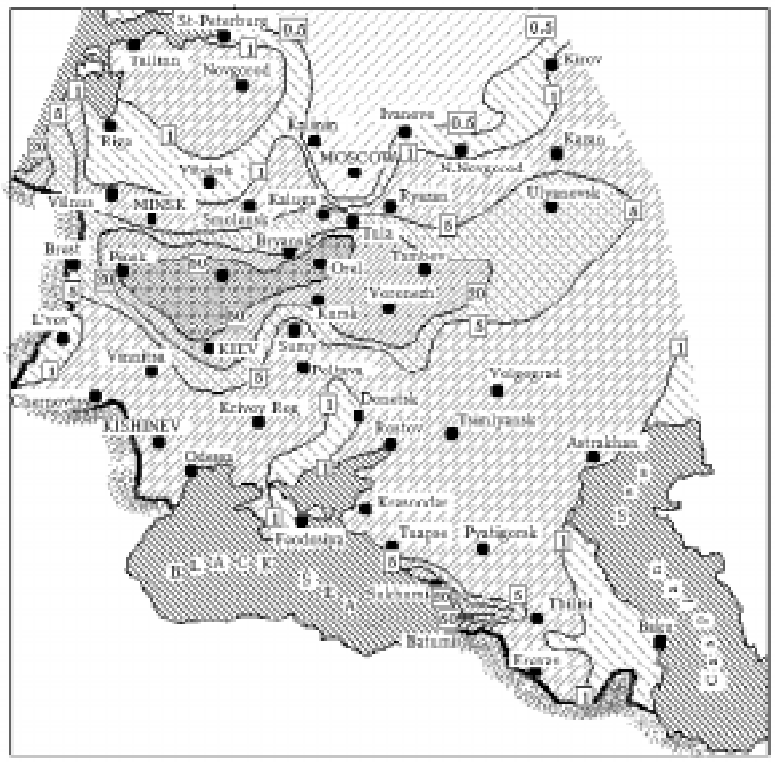

Fig. 13. Map of distribution of external radiation dose from ^{131}I deposited on the ETU after the Chernobyl accident (accumulated dose from the accident time to 1 September 1986).
Numbers in boxes on outlines (isodoses) are radiation doses in mR.
Isodoses east of Sumy-Bryansk are reconstruction by ^{137}Cs contamination density.

5. Estimation of internal thyroid dose from incorporated ^{131}I via the food-chain "grass-cow-milk"

The internal exposure of humans due to radionuclides intake with food stuffs, water and air is of major significance. The internal exposure was most dangerous in the first time after the Chernobyl accident when a large amount of "short-lived" isotopes occurred in the air and on the surface of the soil and vegetation cover. The most hazardous of them was ^{131}I accumulating in the thyroid by peroral and inhalation pathways [23, 24].

According to estimates in [25] made for Moscow after the Chernobyl accident, the thyroid dose from ^{131}I entering the body by inhalation was lower that due to peroral intake. In children of younger age the absorbed thyroid dose due to peroral intake of ^{131}I with milk was 20 times higher that from inhalation intake; for teenagers it was 10 times higher and for adults 2 times. Thus, the greatest hazard is posed by ^{131}I taken in through the chain "grass-cow-milk".

It may be noted that the air concentrations of ^{131}I and $^{132}Te+^{132}I$ cited in [25], exceed in absolute value than those obtained by SPA "Typhoon" in the first day after the accident and in IAG later, by more than an order of magnitude. If these discrepancies are related to the difference in the radioiodine measurement methodology, then the ratio between the ^{131}I content in milk and in the air determined by the same method in [25] is maintained and the above values are valid. But if only concentrations of iodine isotopes in the air are overestimated in [25], but in milk - they are not, then the actual role of the inhalation intake of iodine in formation of total internal radiation dose will be lower the above indicated values and our conclusion on the priority of food chain is enhanced.

The individual internal doses from ingestion intake of radionuclides will be calculated under the assumption that the population does not migrate and consumes only local food stuffs. Such an estimate gives

exposure levels which are highest possible in the given conditions, and these normally exceed actual doses, as part of the diet is made up by relatively "clean" imported food stuffs. However, for the rural areas, where local milk is consumed more than other products, given no bans on its consumption and neglecting the rules of iodine prophylaxis, results of our calculations may be quite close to the actual values.

The calculation of internal doses was done with the methodology [8]. The individual expected equivalent thyroid dose H^c from ^{131}I taken in via the chain "atmospheric depositions-grass-cow-milk" is written as:

$$H^c = P_s \cdot K_{fi} \cdot B_{ig}, \quad (7)$$

where P_s is the one-time deposition of ^{131}I from the air, Bq/m^2;

$K_{fi} = 1.3$ m^2 is coefficient of conversion of ^{131}I to human body from depositions on the soil through milk, with grazing milk cattle;

$B_{ig} = 5.1 \cdot 10^{-7}$ Sv/Bq is the coefficient of conversion from ^{131}I activity coming with consumed milk to the equivalent dose formed in human thyroid (the contribution of other organs to formation of dose is less than 10 %).

In the situation under study, the ^{131}I deposition from the air was not at one-time only and therefore the final internal dose will be determined by summation of doses from ^{131}I depositions on grass each separate day till the completely decay of the radionuclide. Assuming $P_s = P_i$ to be daily depositions of ^{131}I for the i-th day and introducing for brevity $K_{fi} \cdot B_{ig} = K_{FD}$ - the formula for calculating the total internal dose, we can write:

$$H = K_{FD} \cdot \sum_i P_i . \quad (8)$$

By inserting numerical values K_{fi} and B_{ig} cited above, we obtain for ^{131}I:

$K_{FD} = 6.63 \cdot 10^{-7}$ Sv/(Bq/m^2) = 2.45 rem/(Ci/km^2).

Depending on the value of K_{FD} substituted in (8) the value H is expressed either in Sv in the IS system or in rem by the old system. The radioactive decay of ^{131}I in (7) and (8) is taken into account implicitly by selecting numerical values of the coefficients included in the formulae.

Thus, formula (8) allows calculation of the internal dose from the daily deposition of ^{131}I, providing a person consumes only local milk and has been living in the given area from the time of the accident to the time of complete decay of ^{131}I - 1 September 1986. A possible decay of H values was calculated by the same way as were external doses D_{exp}.

Table 4 gives a selection of important data from the whole bulk of the performed calculations. In this table for each city considered the value of H and D_{exp} accumulated by different time is given.

It can be seen from Table 4 that a possible internal thyroid dose from consumption of local milk contaminated with ^{131}I is about three orders of magnitude higher the external radiation dose. Let us use data on the value of H in separate points of ETU for constructing a map of geographic distribution of the internal thyroid dose. For areas in which ^{131}I in depositions did not change, we take the empirical relation between H calculated by (8) and the accidental ^{137}Cs accumulation on the soil:

$$H = 132 \cdot \Delta P_{Cs}^{1.26}, \text{ cSv}$$

at $0.01 < \Delta P_{Cs} < 1$ Ci/km^2.

Figure 14 gives a comparison of the estimated equivalent thyroid dose H_{th} based on ^{131}I depositions measurements and density of accidental ^{137}Cs soil contamination. As can be seen from Figure 14 in the indicated range the values in question correlate fairly well.

Figure 15 presents a map of spatial distribution of possible thyroid doses from ^{131}I taken in with local milk after the Chernobyl accident. As before, the isodoses east of Sumy-Bryansk have been reconstructed by the ^{137}Cs soil contamination density. Considering the above, the presented map, constructed from rather conservative estimates of thyroid doses, is only a rough estimate because data on the diet of local population, ways of milk transportation to city dwellers, regimen for milk cattle and other information is not available.

Nevertheless, the cited data lead us to conclude that unless the bans on cattle grazing and local milk consumption had been introduced and prophylactic and protective measures had been taken, the population could have received a radiation dose above the admissible level of 1.5 rem a year by NRB 76/87 across a rather extensive area.

The conversion coefficients from ^{131}I soil depositions to equivalent thyroid dose cited in [8] make it possible to calculate threshold accumulation values for this isotopes on pastures resulting in receiving the maximum permissible dose.

Table 4

Accumulated external whole body doses and internal thyroid doses from exposure to ^{131}I and $^{132}Te+^{132}I$ estimated from measurements of depositions on collectors placed at meteorological stations in ETU

In brackets is the confidence interval for radiation dose and absorbed thyroid dose in case when its bounds are different from the tabulated value by more than 10%.

Meteostation in city:	^{131}I exposure					$^{132}Te+^{132}I$ exposure	
	Radiation dose (mR) on date: (second line - proportion of dose by 01.09 - 2-4 colums)				Equivalent thyroid dose, cSv	Radiation dose, mR	Equivalent thyroid dose, cSv
	08.05	15.05	01.06	01.09	01.09	01.06	01.06
Baltiysk	13 55%	17 75%	22 94%	23	27		
Baranovichi	15 59%	19 77%	24 95%	25 (17 - 33)	29 (20 - 38)	21	0.002
Baryshevka	13 36%	20 59%	31 90%	35 (34 - 50)	41 (40 - 58)	42	0.003
Brest	6 36%	8 76%	10 94%	11	12	28	0.002
Vilnyus	0.8 48%	1.1 70%	1.5 98%	1.6	1.9		
Vinnitsa	1.3 32%	2.5 61%	3.7 90%	4.1	4.8		
Vitebsk	0.18 35%	0.30 58%	0.46 87%	0.53	0.6		
Genichevsk	0.37 43%	0.57 67%	0.78 92%	0.85	1		
Gomel	81 57%	108 76%	134 94%	142	166	106	0.008
Grodno	5.8 57%	7.8 76%	9.6 95%	10.2	12		
Dnepropetrovsk	0.75 46%	1.1 68%	1.5 92%	1.6	1.9		
Donetsk	0.42 42%	0.67 66%	0.93 92%	1.0	1.2		
Dukshtas	0.19 37%	0.31 61%	0.45 89%	0.51 (0.46 - 0.6)	0.6 (0.5 - 0.7)		
Zaporozhje	0.73 45%	1.1 69%	1.5 92%	1.6	1.9		
Ivano-Frankovsk	0.68 32%	1.2 56%	1.9 89%	2.1 (1.9 - 2.5)	2.5 (2.3 - 2.9)		
Izmail	1.2 35%	2.1 63%	3.0 91%	3.3	3.9		
Kaliningrad	13 58%	17 77%	21 95%	22	26		
Kaunas	0.30 44%	0.47 68%	0.64 92%	0.69 (0.67 - 0.8)	0.8 (0.78 - 0.9)		
Kerch	0.48 40%	0.77 64%	1.08 90%	1.2 (1.19 - 1.3)	1.4 (1.39 - 1.6)		
Kiev	14 38%	23 63%	33 90%	36	42		
Klaipeda	9.4 55%	12.9 75%	16 94%	17	20		
Krivoy Rog	1.3 45%	1.9 68%	2.6 92%	2.8	3.3		
Lugansk	0.45 42%	0.7 65%	0/97 90%	1.1	1.3		
Lvov	0.33 41%	0.50 64%	0.70 89%	0.79 (0.72 - 0.92)	0.92 (0.84 - 1.08)		
Lutsk	2.1 51%	2.9 73%	3.8 94%	4.0 (3.6 - 4.3)	4.7 (4.2 - 5.0)		
Mariupol	0.88 44%	1.3 67%	1.8 91%	2.0 (1.9 - 2.34)	2.3 (2.2 - 2.7)		
Minsk	1.1 46%	1.6 65%	2.2 91%	2.4 (2.2 - 2.9)	2.9 (2.6 - 3.4)		
Mogilev	2.3 53%	3.1 73%	4.0 93%	4.3	5.0		

Table 4 (continued)

Meteostation in city:	^{131}I exposure					^{132}Te+^{132}I exposure	
	Radiation dose (mR) on date: (second line - proportion of dose by 01.09 - 2-4 colums)				Equivalent thyroid dose, cSv	Radiation dose, mR	Equivalent thyroid dose, cSv
	08.05	15.05	01.06	01.09	01.09	01.06	01.06
Moscow	0.14 0.39%	0.22 62%	0.32 91%	0.36 (0.3 - 0.42)	0.42 (0.34 - 0.5)		
Nikolaev	0.74 41%	1.2 65%	1.6 90%	1.8	2.1		
Obninsk	0.09 37%	0.13 57%	0.20 87%	0.23	0.27		
Odessa	0.19 40%	0.31 66%	4.3 91%	4.7 (1.1 - 4.9)	5.5 (1.3 - 5.7)		
Pinsk	30 59%	39 77%	48 95%	51	59	111	0.008
Poltava	0.67 45%	1.0 68%	1.4 91%	1.5	1.7		
Riga	0.29 40%	0.46 65%	0.65 91%	0.72	0.84		
St.-Petersburg	0.29 46%	0.43 68%	0.57 92%	0.62 (0.5 - 0.78)	0.7 (0.6 - 0.9)		
Sumy	0.71 44%	1.1 69%	1.4 88%	1.6 (1.6 - 2.4)	1.84 (1.8 - 2.8)		
Uzhgorod	0.23 29%	0.46 58%	0.71 89%	0.80	0.93		
Feodosiya	0.31 40%	0.50 65%	0.7 91%	0.77 (0.7 - 0.83)	0.9 (0.8 - 0.97)		
Kharkov	0.44 44%	0.67 66%	0.91 91%	1.0	1.2		
Kherson	0.9 34%	1.6 61%	2.4 90%	2.7	3.1		
Cherkassy	7.6 44%	11.7 68%	15.8 92%	17.2 (11 - 24)	20.1 (13 - 28)		
Chernovtsy	1.3 33%	2.3 59%	3.5 89%	4.0	4.6	≤ 200	≤ 0.01

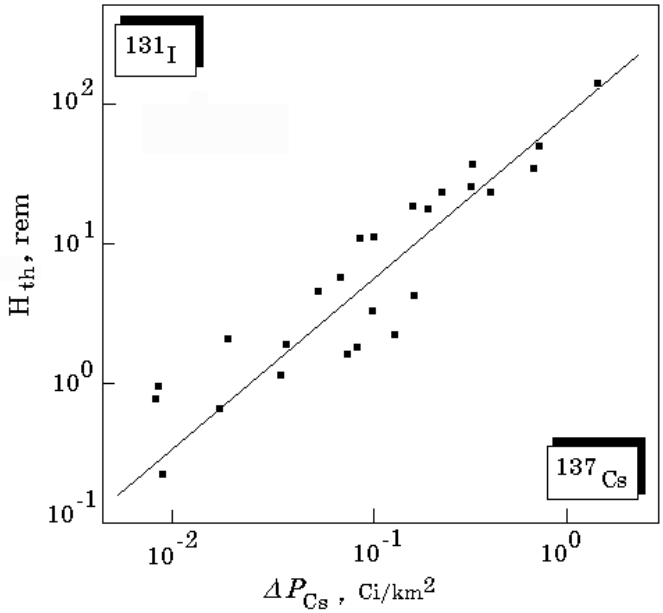

Fig. 14. Comparison of equivalent thyroid doses H_{th} estimated from measured depositions of ^{131}I and the accidental ^{137}Cs soil contamination density (the ordinate axis).

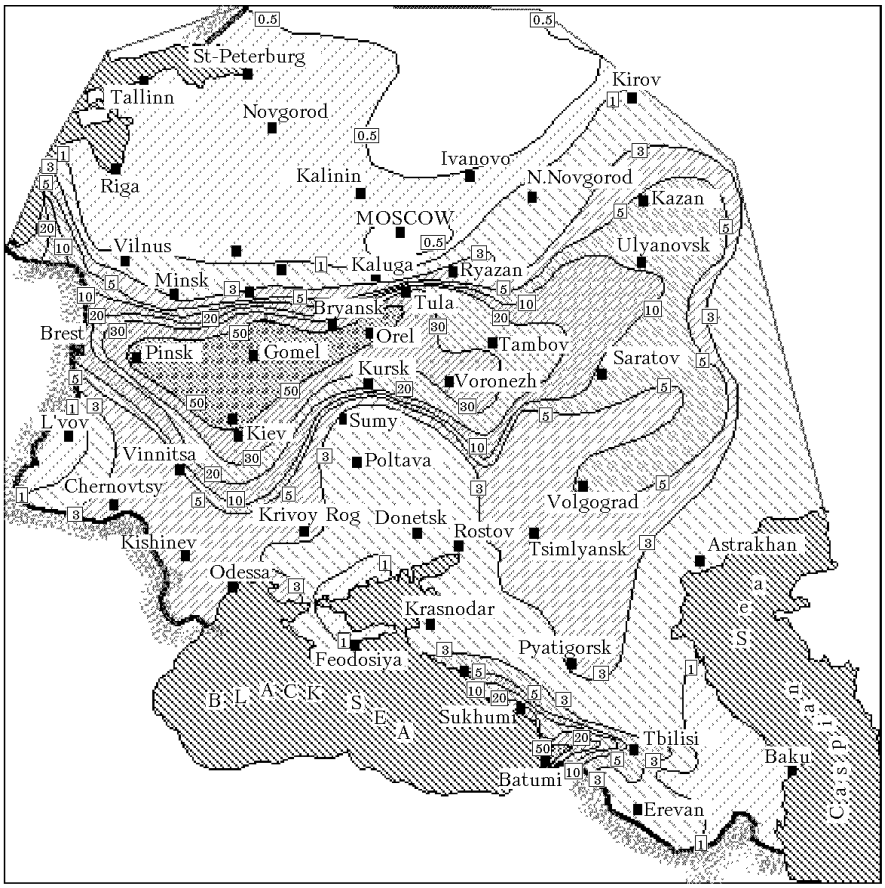

Fig. 15. Map of distribution of possible (maximum) equivalent thyroid doses from incorporated ^{131}I for the population living in 1986 in the areas contaminated after the Chernobyl accident.
Numbers on outlines in boxes are equivalent dose, cSv.
Outlines east of Sumy-Bryansk are doses estimated with results of reconstruction of ^{131}I.

According to temporary norms [26] in force during 1986 the values of permissible concentrations of radioactive materials in food stuffs were calculated from the condition that the permissible thyroid dose of 300 mGy for an adult be not exceeded and for children younger than 7 years old this value should be 10 times lower. Then the critical value of ^{131}I accumulation on pastures for the population of different age groups consuming local milk will be:

Age bounds in the group, years	0 - 7	7 - 13	13 - 19	19-70
Permissible absorbed thyroid dose, mGy	3	30	30	30
Threshold density of ^{131}I accumulation on soil, Ci/km^2	0.18	4.4	10.5	12.8

It follows from these data that the level of ^{131}I contamination in pastures grazed by milk cattle posing a hazard for adults is higher than that for children, particularly, younger ones. For children younger 7 years old, the threshold density of ^{131}I contamination is 0.18 Ci/km^2 which is 1.5 times lower that adopted in Sweden [27]. The resulting data enable us to estimate for different times the outlines of the region within which unrestricted grazing of cattle on natural pastures should have been temporarily banned, if the milk of these regions had been consumed by small children without limitations or protective measures.

Figure 16 shows how the outlines of the region of unrestricted grazing of milk cattle change with time. This map has been constructed by the above described method. The data of Figure 16 indicate that in the first days of May the region in which unrestricted grazing was banned included the whole ETU, where

by that time fresh grass had grown and, probably, the whole Zakavkazje (though no direct measurements were performed there and the data are very approximate). As soon as 15 May, the region in which unrestricted grazing of cattle was banned was significantly reduced. In the north the boundary was south of Riga, went through Vilnyus, Vitebsk, Novgorod, Kaluga and in the south involved the areas of Lvov and Poltava, Donetsk, Rostov-on-Don and went approximately between Erevan and Baku towards Astrakhan. Later on the boundary of this region went on shrinking. By 15 July the areas prohibited for unrestricted grazing were only the zones north of Chernobyl and east of Gomel shown in Figure 16. Later on practically for all the regions, except the nearest zone, the iodine problem stopped to exist.

It should be stressed again that the above calculations are only preliminary estimates and besides, they do not take into account specific features of the diet in different parts of the country and assume complete absence of protective measures, in particular, iodine prophylaxis for the population.

6. Estimation of external and internal doses from ^{132}I

Let us now briefly dwell on the doses resulted from exposure to short-lived ^{132}I (half-life of 2.30 hour) which is a daughter product of ^{132}Te (half-life of 78.2 hour) in the atmospheric depositions after the Chernobyl accident.

The doses were calculated in a similar way from measurements of these isotopes in daily atmospheric depositions. As these radionuclides have a half-life shorter than that of ^{131}I, we took a shorter observational series - to 1 June 1986. The conversion coefficient k in formula (5) from the surface soil contamination density to the gamma-radiation dose rate for $^{132}Te+^{132}I$ is six times higher that for ^{131}I and is $k = 43$ $(\mu R/hour)/(Ci/km^2)$. However, the dose factor $K_{FD} = 3.88 \cdot 10^{-4}$ rem/(Ci/km^2) in formula (8) is almost six thousand times lower than for ^{131}I ($K_{fi} = 0.027$ m^2, $B_{ig} = 3.9 \cdot 10^{-9}$ Sv/Bq [8]).

Results of calculations of accumulation of total activity of $^{132}Te+^{132}I$ on the soil are shown in Table A2 of attachment and calculated external doses accumulated by 1 June 1986 and internal thyroid doses - in Table 4. It can be seen from Table 4 that the external doses from $^{132}Te+^{132}I$ for most compared points were higher those from gamma-irradiation of ^{131}I. It is worth noting that the spread in ratios of the internal doses from $^{132}Te+^{132}I$ and ^{131}I is fairly significant: from 1 (Baranovichi) to 136 (Vinnitsa) which suggests different type of transport of these radionuclides in the atmosphere. By absolute value, the external doses from $^{132}Te+^{132}I$ were insignificant and in the indicated cities they did not pose a hazard for the population similar to the external doses from ^{131}I.

The internal thyroid doses from ^{132}I transferred to humans via milk is lower those from ^{131}I by four orders of magnitude. Therefore, exposure of thyroid to ^{132}I can be neglected.

Conclusions

As a result of the radiation disaster at the Chernobyl NPP an extensive territory of ETU was contaminated with depositions of radioactive materials, in particular, iodine radionuclides.

Daily measurements of radioactive depositions at meteorological stations of Goscomhydromet in the former USSR allowed maps to be drawn quickly of daily depositions in ETU. Analysis of the obtained data has made it possible to construct a map of the ETU contamination with ^{131}I which was a major radiation threat in the first several months after the accident. In the present work, continuous series of daily data on atmospheric depositions of ^{131}I in separate points which have been critically analyzed, corrected and added with results of indirect calculations are used for estimating the accumulation of ^{131}I on soil and constructing a refined map of the ^{131}I soil contamination in ETU and Zakavkazje.

The same array of data on daily ^{131}I depositions is used for calculating and constructing maps of dose distribution of external γ-irradiation from the soil surface and maximum internal thyroid dose due to incorporated ^{131}I. The results of our calculations lead us to conclude that external doses from ^{131}I, by absolute value, across ETU and Zakavkazje, except the 30-km zone, were not high and did not present a hazard for the population. The main dose burden of significance was due to ^{131}I intake by humans via the food chain "grass-cow-milk". If there had been no iodine prophylaxis or food restrictions, the dose could have been 1000 times higher the doses from external exposure.

If the population consumes local milk without any restrictions or protective measures, bans should be introduced on unrestricted grazing of dairy cattle on natural pastures, so that the content of ^{131}I in milk would not reach hazardous levels. Considering ^{131}I decay, these bans should be temporary. The calculated map of time moving of outlines of unrestricted grazing of dairy cattle shows that in the beginning of May 1986 such bans were necessary to be introduced practically in the whole ETU and Zakavkazje, in the areas in which by that time new grass appeared and the cattle was moved from stalls to pastures. With time, this boundary was shifting and the area in question was shrinking. By the end of the summer ^{131}I broke down and the hazard of exposure to it ceased to exist.

The doses of internal thyroid exposure to $^{132}Te+^{132}I$ were much lower and they can be ignored.

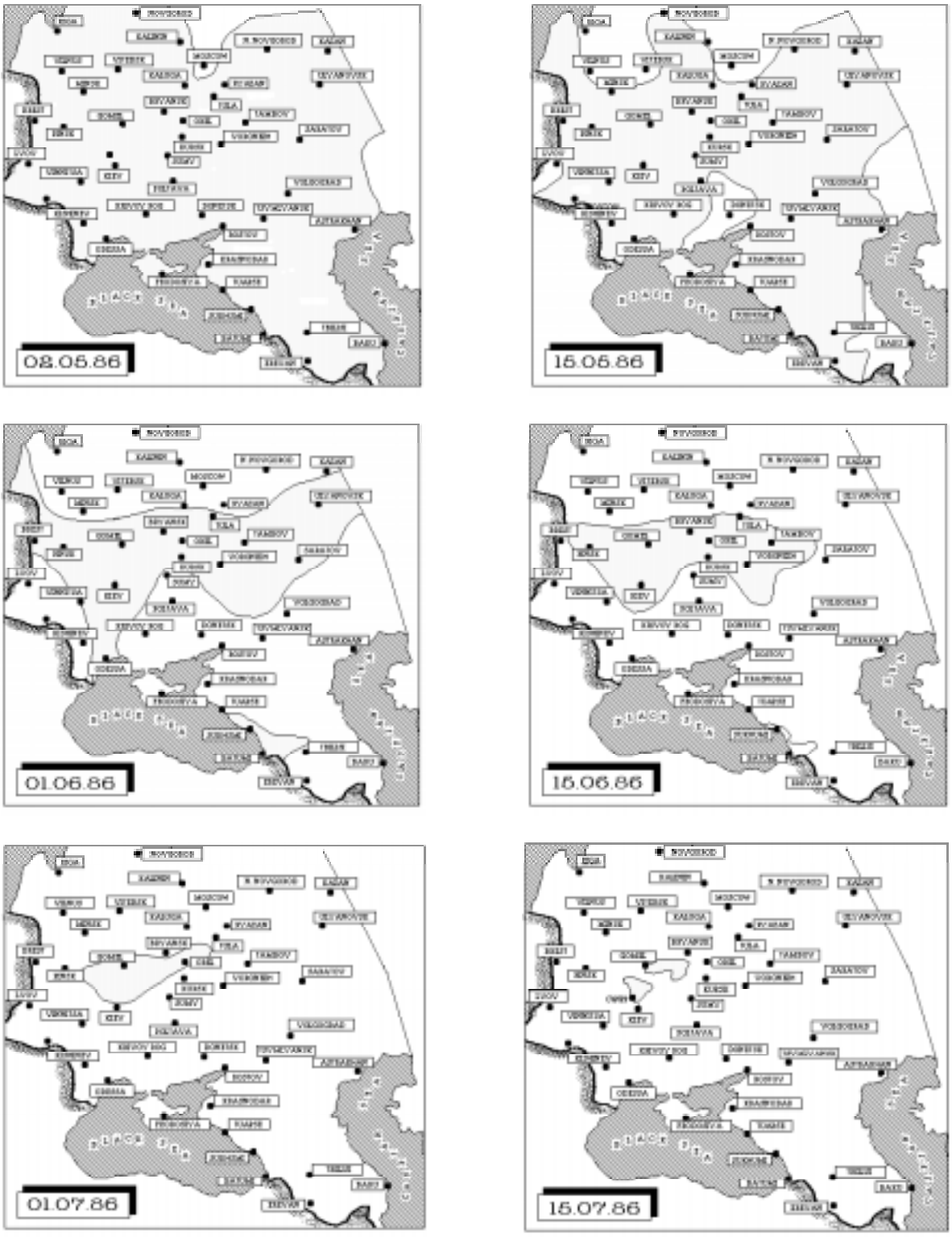

Fig. 16. Time change in ETU boundary within which milk from natural pastures might have increased ^{131}I content.
Estimates have been made for younger children whose diet included whole milk without any restrictions and protective measures (iodine prophylaxis).

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Attachment

Data of measurements of daily depositions of ^{131}I and $^{132}Te+^{132}I$ on collectors

Tables A1 and A2 show measurements of daily depositions of ^{131}I and $^{132}Te+^{132}I$ on collectors exposed at meteorological stations of the former USSR in April-June 1986.

Explanations to Table A1.

In those cases when the deposition sample was divided two parts and the ^{131}I content was measured in them separately Table A1 gives both values - measurement 1 and measurement 2.

The same table contains two values for the same exposure interval if the sample was measured twice at different times. The measured activity was, of course, assigned to the date of the collector exposure.

Table A1 includes data obtained by different ways - column "Type".

If any symbol is missing in the column "Type", it means that the activity was measured with a gamma-spectrometer with a semiconductor detector (SCD). These results are most accurate of all.

In other cases the following designations are used:

c - measurement has been performed with a gamma-spectrometer with a scintillation detector;

v - the value has been calculated using space-time correlations;

b - the value has been derived from the ratio of ^{131}I depositions and total β-activity;

bp - the value has been derived from the ratio of ^{131}I depositions and total β-activity with an adjustment, if results of the 1-st and 2-nd measurements of ^{131}I are different;

db - total (for 5-10 days) ^{131}I depositions are distributed with respect to days in proportion to β-activity of depositions on those days;

k - the value has been derived from the ratio between concentrations of ^{131}I and its depositions from the air.

In calculations, given a choice in selecting initial data, preference was given to measurements with a gamma-spectrometer with SCD. If results of separate measurements of two halves of the collector sample differed by not more than 10 $Bq/m^2 \cdot$ day, only the average value rounded off to ten was entered Table A1. If results differed by more than 10

$Bq/m^2 \cdot$ day, both measurements were entered the table, and in further calculations the arithmetic average was used, while the initial actual values of depositions were used for determining the range of possible values of the calculated quantity of ^{131}I accumulation on the soil and radiation dose. In those cases when data of direct measurements were scarce, the initial data array was added with estimates of ^{131}I depositions from space-time correlation relations with total beta-activity depositions, volatile ^{137}Cs depositions, soil gamma-radiation dose rate etc.

Graphical comparison of daily and cumulative depositions of ^{131}I and $^{132}Te+^{132}I$ on collectors

Plots of Figure A1 show continuous series of daily depositions of ^{131}I and $^{132}Te+^{132}I$ (see the paper text) and the time dependence of cumulative depositions of these radionuclides.