MODELLING OF THE ATMOSPHERIC DISPERSION OF RADIONUCLIDES FROM THE URANIUM PROCESSING FACILITIES OF THE FORMER PRIDNEPROVSKY CHEMICAL PLANT IN UKRAINE

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Abstract: The modelling studies of the radioactive atmospheric contamination at the industrial site of the former Pridneprosky Chemical Plant is presented.

Key words: *uranium tailings, radon, building demolition, emission factors, environmental safety assessment.*

INTRODUCTION

Pridneprovsky Chemical Plant (PChP) was one of the most powerful enterprises dealing with uranium processing in USSR. The uranium ores had been processed at PChP from 1948. In the early 1990s with the crash of USSR PChP was split into several separate companies and processing of uranium was stopped. Now the radioactive waste originating from uranium treatment is stored in 9 tailings, their total radioactivity is estimated as $3.2 \cdot 10^{15}$ Bq and some of the tailings are not properly covered (Voitsekovhych and Lavrova, 2009). Besides the radioactivity stored in uranium tailings, some buildings at the industrial site of PChP are highly contaminated while the closest distance from the PChP to living zone is less than 1 km (Fig. 1) and the working enterprises still exist at the former territory of PChP. The basic information on the current contamination of the PChP industrial site, remediation planning and decontamination of the former uranium extraction facilities is reviewed by Lavrova and Voitsekhovych (2013).

TAILINGS INFLUENCE ON RADON CONTAMINATION

Contamination by radon originating as product of radioactive decay from uranium tailings could be one of essential exposure pathways and hence assessment of present-time and future radon distributions in atmosphere around the PChP is necessary. We consider 3 tailings shown at Figure 1: “Zapadnoe” (hereafter refereed as “West”), “Central Yar” and “Yugo-Vostochnoe” (hereafter refereed as “South-East”) since other tailings are either properly covered or located quite far from the PChP industrial site.

The modeling study was performed on the basis of measurement data (radon concentrations in air and in contaminated buildings and radon fluxes from the territory of tailings) collected during previous measurement campaigns at the territory of PChP (Lavrova and Voitsekhovych, 2013). The values of emission rate spanned range from 0.05 to 2 $\mathrm{Bq/m^2}s$ at the territory of West tailing, from 0.7 to 12.5 $\mathrm{Bq/m^2}s$ at the territory of Central Yar tailing and were on average 3 $\mathrm{Bq/m^2}s$ at the territory of South-East tailing (measurements taken in 2008 were used in present study; later the emission rate at South-East tailing was significantly decreased by improvement of cover). Interpretation of emission rate measurements had been enhanced by the expert subdivision of tailing’s surface onto sectors which take into account particular properties of the tailings territories. For instance West tailing was subdivided onto Northern part of tailing which was recultivated, Southern asphalted part, and Dam, separating tailing from the outside territory. Emission rate measurements were averaged in frame of each sector and this average value had been considered as representative for the whole sector. Those averaged values of emission rates were further taken as constant emission rate from the corresponding sector in simulation runs.

Simulations had been performed with the CALMET-CALPUFF model (Scire et al., 2000) through the 5-year period from 2005-2009 using the observations of the nearby surface synoptic station located in

Dnipropetrovsk (measured each 3 hours) and the nearest radiosonde data (measured each 12 hours). Since the computational domain had been quite small (2x8 km^2) radon had been considered as tracer.

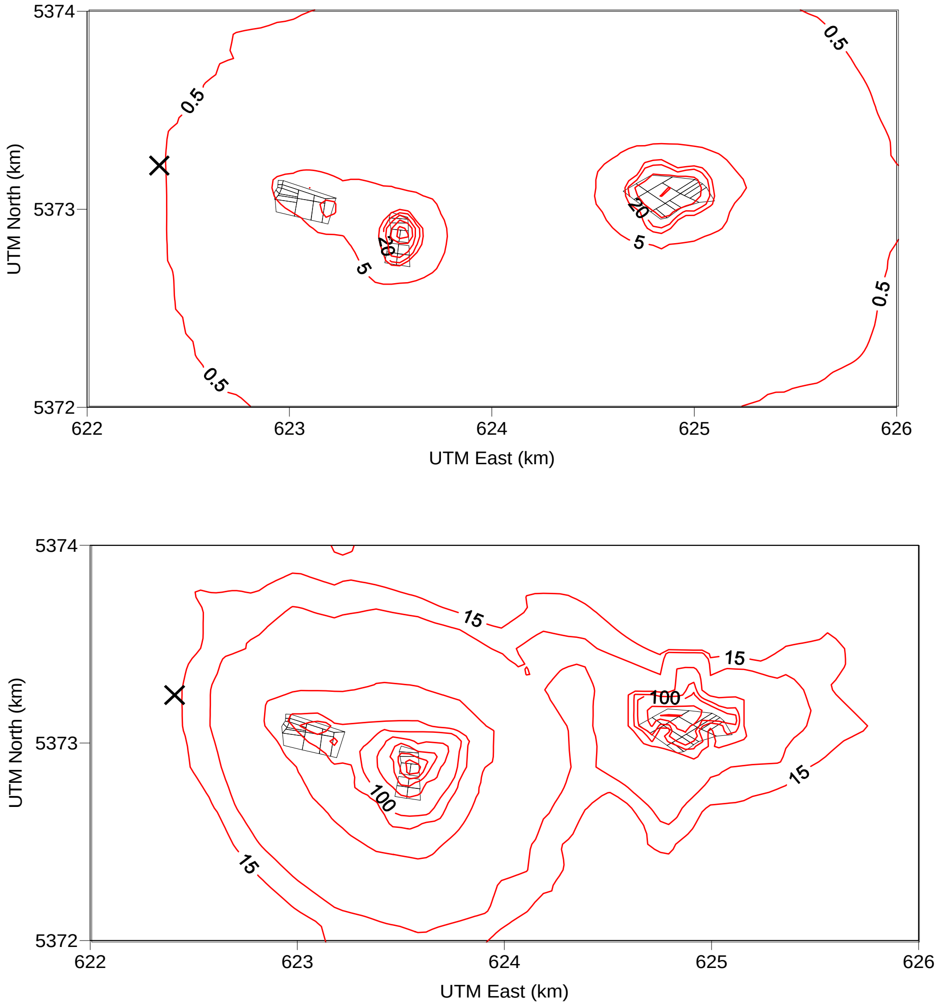

Figure 1. Average and maximum over 5 years daily concentration of radon created by the “West”, “Central Yar” and “South-East” tailings. Concentration isolines are shown for the values: 0.5, 5, 20, 50, 80, 95 Bq/m^3 and 15, 50, 100, 150, 200, 250, 300, 340 Bq/m^3. Symbol ✕ shows the nearest point in the inhabited area.

As it was shown by calculations (Figure 1, upper) the average over 5 years radon concentrations created by the tailings are less than the background levels of radon concentration (20 Bq/m^3) at the distances greater than 100 m from the tailing’s border. Maximum daily concentrations (Figure 2, bottom) are of course greater than the average concentrations but still small at the border of inhabited area (15 Bq/m^3).

The simulated concentrations had been compared with concentration measurements collected at the territory of the tailings. On average the simulated concentrations had been highly underestimated. For instance concentration measurements collected at the territory of the West tailing reached 200 Bq/m^3 in 2008 and 400 Bq/m^3 in 2007. On average the simulated concentrations for that tailing had been underestimated by the factor of 5. Similar results had been obtained for the South-East tailing. Sensitivity tests had been performed with respect to different assumptions of source rate distribution and meteorological parameterizations. In particular the 2-layer soil model of radon emission from the tailing had been used together with the measurements of geomorphological parameters of the West tailing and measurements of soil moisture and contamination inside tailing. However all attempts had not been successful in explanation of the disagreement of model results and measurements. Therefore it was concluded that radon concentrations above the West and South-East tailings are created mostly by the external sources, i.e. by contamination of the surrounding territories at the PChP industrial site.

Better agreement between measured and calculated radon concentrations had been reached at the territory of the Central Yar tailing where the simulated results were underestimated on average by the factor of 1.5 as compared to measurements. As it was mentioned above radon emission rates at the Central Yar tailing are significantly greater than those at the West tailing and therefore it is clear that the relative contribution of the 'external' sources of radon contamination to the concentrations at the territory of that tailing is also less significant.

CONTAMINATED BUILDINGS

One of the most significant potential sources of contamination at the industrial site of PChP are buildings No. 103 and No. 104 where the uranium extraction operations had been held. Even though the above buildings are presently closed, in a very close vicinity (of about 15 meters) the working enterprise is located (Building No. 102, Figure 2). Therefore the following questions arise:

1. What will be the possible consequences if buildings No. 103 and 104 "remain as they are"?
2. What levels of contamination will be reached if those buildings are dismantled?

'Ventilation' scenario

As for the first question even if accidental scenarios of building demolition are not considered the contamination of the surrounding territories would result in particular due to ventilation of the contaminated dust from the building. In the simulation scenario it was conservatively assumed that all windows in Building No. 103 are broken and the radionuclides are released due to ventilation.

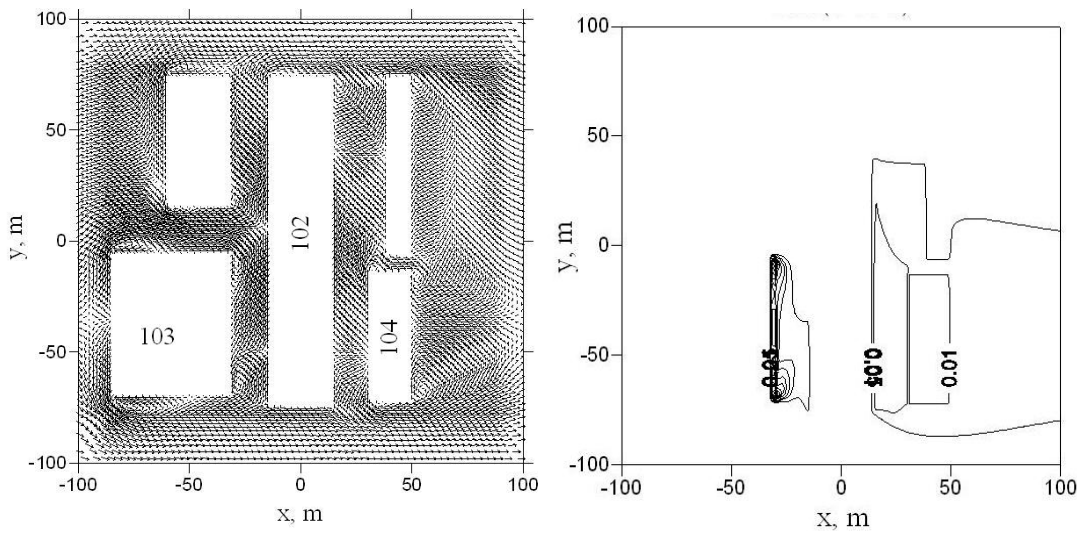

Figure 2. Horizontal distribution of the wind field and normalized concentration near ground for the considered scenario. Isolines are shown for the values of C/C_b=0,01; 0,05; 0,1; 0,2; 0,3; 0,4; 0,5; 0,6; 0,7; 0,8; 0,9; 1.

The calculations had been performed for the following meteorological scenario: ambient undisturbed wind of 5 m/s at height 10 m directed towards the positive direction of x-axis (Figure 2). Concentration of radionuclides in air inside building is set to constant and equal to C_b. The ventilation flux of concentration was parameterized as: $F = \left(\beta_h \left|u_h\right| + \beta_\perp \left|u_\perp\right|\right) C_b$, where $\beta_h, \beta_\perp$ are coefficients of ventilation efficiency with respect to horizontal and normal components of wind velocity ($\beta_h = 0.3, \beta_\perp = 0.8$ according to Santamouris, 2002).

Calculations had been performed with the CFD model of Kovalets and Maderich (2006). Figure 2 shows the calculated wind field around the buildings and normalized concentration near the ground. The values of normalized concentration reach levels of $C / C_b = 0.05$ near the Building No. 102, where the operating entherprise is located. Due to intense recirculation zones near the wall of the Building No. 103 the normalized concentration reach the value of 1. If radon is considered the corresponding dimensional concentrations will be 50÷300 Bq/m^3 near the building No. 102 and 1÷6·10^3 Bq/m^3 near the Building No. 103. The existing measurements in the vicinity of buildings reveal radon concentrations which are less but yet comparable to the above conservative estimates.

'Demolition' scenario

Activities connected with demolition and remediation of the territory of the Building No. 103 requires assessment of the possible consequences of those activities including contamination of environment. Such assessments require accurate estimations of possible releases which lead to the need of making complete inventory of possible sources of contamination inside building. Sources of contamination fall in 2 broad categories: 1) contamination of process equipment; 2) contamination of building materials, contaminated dust and spills. At current stage, when amount of radioactivity which is contained inside process equipment is not known, estimates of potential emissions resulting from demolition activities could be made only in assumption that contaminated equipment was taken out from the building. I.e. only emissions resulting from contamination of building materials, dust and spills were considered.

General framework of source term estimations in case of demolitions and dismantling of contaminated facilities is presented in DOE (1994). According to DOE (1994) the release due to building demolition could be expressed by the following general relationship:

$$ST = MAR \cdot DR \cdot ARF \cdot RF \cdot LPF \quad (1)$$

where source term (ST) is the amount of respirable pollutants released to the atmosphere during the demolition; MAR is the material-at-risk which is equal to the amount of radioactivity available to be acted on by a given physical stress; DR is damage ratio which is equal to the fraction of the MAR actually impacted by the demolition conditions; ARF is airborne release fraction, i.e. the fraction of radioactivity suspended in air as an aerosol and thus available for transport due to a physical stress from a specific activity; RF is respirable fraction which is commonly assumed to include particles 10-μm aerodynamic equivalent diameter and less; leak path factor (LPF) is the fraction of the radionuclides in the aerosol transported through some confinement system and/or emission mitigation methods (e.g., misters or foggers).

The monitoring data related to building No. 103 and reviewed by Lavrova and Voitsekhovych (2013) could be used for assessment of MAR in formula (1). Other factors (DR, ARF, RF and LPF) depend on specific demolition plan and are specified in any case using empirical data presented in DOE, (1994). The same approach could be used also for assessment of accidental demolition scenarios.

The measurements which had been taken up to now inside Building No. 103 provided data of surface contamination by different nuclides (U-238, Ra-226, Pb-210, Po-210, Th-230, Th-228) in different parts of building. We consider the following release scenario: during dismantling activities large part of the bulding: roof or wall falls down leading to release of dust. This scenario also corresponds to accidental demolition of building for example, due earthquake. Since according to measurements contamination of wall materials is much less than contamination in dust and in spills, wall materials are further neglected.

For powders ARF and RF could be taken from DOE (1994) (chapter 4.4.3 Free-Fall Spill and Impaction Stress**).** According to data provided there the conservative estimate for the ARF=0.01 and RF=0.2. The total amount of contamination in dust is taken by multiplying data from average measured contamination of dust in industrial unit on area of building. For the case of spills presently we know only maximum area concentration densities in spills, while we still do not know total area of spills. Therefore at present stage we make quite voluntary assumption that spills cover 3% of the floor area. The release is considered to be instantaneous. The radionuclide mixture consists of: U-238 (51%), Th-230 (32%), Ra-226 (12%).

In simulations we performed series of instantaneous releases with different meteorological conditions which actually happened during 5 years. The calculations had been performed with the CALPUFF-CALMET model. The average and maximum calculated concentrations at different distances from the source are presented in Table 1.

Table 1. Statistics of maximum 3-h averaged concentrations at different distances from the source

Statistics	**50 m**	**100 m**	**300 m**
Average, Bq/m^3	1.3	0.45	0.07
Maximum, Bq/m^3	13.3	5.5	0.9

CONCLUSIONS

Results of calculation of radon contamination originating from the uranium tailings show that uranium tailings are not the only source of radon contamination at the industrial site of PChP. This follows from the fact that the sensitivity studies do not allow to explain the high level of underestimation of the calculated radon concentrations as compared to measurements by uncertainties in radon emission rates from the uranium tailings and/or by meteorological parameters. In particular, such additional sources of contamination can include the former uranium ore storage sites, as well as sites for storage of contaminated soils and construction materials at the PChP site. Therefore it is necessary to carry out further work on identification, characterization and documentation of such undocumented sources of radon exhalation at the PChP industrial site.

The detailed safety assessments of the contaminated buildings at the territory of industrial site of PChP require collection of additional data concerning the total area of spills inside buildings and the levels of contamination of the process equipment. Also the detailed dismantling scenario should be prepared prior to the assessments of the consequences of dismantling activities on the environmental contamination both at the industrial site and in the living zone. Therefore at present stage only some preliminary estimates were possible. It is shown that for the conservative scenario of ventilation of the radioactivity from inside buildings without building demolition the resulting levels of contamination in the vicinity could be several times more than currently observed levels of contamination. Accidental demolition such as falling down of the roof of building would result in instantaneous release and the corresponding airborne radioactivity may exceed maximum permissible concentrations of U-238, Th-230 and Ra-226 at distances of about 100 m.

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