Lung Types of Thoron Daughters in Monazite Storage Facility[1]

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Abstract. The dissolution of thoron daughters, sampled in workplace of monazite storage facility, in imitation of body fluids is studied. The dissolution of real ^{212}Pb aerosols can be described by superposition of two exponential dependencies with the rate of fast dissolution λ_f = 145 day^{-1}; rate of slow dissolution λ_s = 2 day^{-1} and the part of activity with fast rate of dissolution k = 0.3. The estimated values of dose conversion factor from thoron daughters exposure to effective dose 80–150 nSv per Bq·h·m^{-3} considerably (at least 3–5 times) exceeds the DCF values recommended by UNSCEAR. It is shown that the most perspective monitoring method of inhalation intake of ^{212}Pb aerosols is the direct measurements of whole body nuclide activity

1. Introduction

Traditionally the general attention in radon exposure investigation is paid to ^{222}Rn and its daughters exposure. To the other radon isotope ^{220}Rn, considerably less attention is paid and usually it's supposed that the thoron daughter contribution to population and workers exposure is practically negligible. Nevertheless in some specific situations the inhalation of thoron daughters (especially the most long lived nuclide ^{212}Pb) can be the dominant source of internal exposure in workplaces.

The example of such a workplace is the monazite storage facility situated in the South East part of Sverdlovsk region (Russia). Monazite contents 5–10% of ThO_2 (sometimes up to 30%) and 0.2% of U_3O_8. Since 1960 more than 82 000 t of monazite concentrate are stored on this facility in 19 wooden warehouses (former granaries) and 4 hangars [1]. The warehouses were built in 1940 and most of them are in the need of serious repair now. The extremely high values of thoron volume activity and thoron equivalent equilibrium concentration (EEC) do not allow using the standard techniques of radon and radon daughter measurements. The specially designed nuclear track detectors were used for monitoring of radon and thoron volume activity. Thoron EEC was measured by grab sampling of radioactive aerosols on filter. The radioactive monazite dust concentration in air also was measured by air sampling on the filter.

2. Method and results

The main sources of radiation exposure of storage facility workers are: external gamma-radiation (~0.15 mSv/h inside the warehouse, 0.07 mSv/h on the outside wall and 0.012 mSv/h at the distance 10 m from the warehouse), inhalation of radon and thoron daughters (EEC_{Rn} = 45–250 Bq/m^3; EEC_{Tn} = 60–700 Bq/m^3), direct inhalation of thoron gas (thoron activity concentration 1600–14 000 Bq/m^3), inhalation of monazite dust.

TLD and electronic direct reading alarm detectors are used for the monitoring of external exposure. At the same time, the monitoring for internal exposure is considerably more complicated. The problem of internal exposure monitoring is especially important considering the perspectives of reconstruction of storage facility and the processing of monazite concentrate for thorium and rare earth extraction.

At present the values of dose conversion factor (DCF) from thoron EEC exposure to effective dose, recommended by UNSCEAR are in the range 32–40 nSv per Bq·h·m^{-3} [2, 3]. In general such DCF value is correspond to Type F materials according to their rates of absorption from the respiratory tract to body fluids [1]. It was shown that DCF value is considerably depends on absorption rate (Type F or Type M) of thoron daughters [1]. Also it was stressed out that the experimental data on real lung type of thoron daughters (especially ^{212}Pb) in workplaces are needed.

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In assessment of dissolution rate of radioactive aerosols it is very important to provide the correspondence of chemical properties of aerosol in experiment to properties of aerosol in real workplaces. So the aerosol sampling in closed laboratory boxes isn't suitable for practical assessment of aerosol types. On the other hand, the thoron daughter concentrations in the ordinary workplaces or dwellings are too small for experimental purposes.

The presence of the unique monazite storage facility allows estimation of the aerosol type of ^{212}Pb for real aerosols sampled in workplace. The aerosol sampling was conducted during 3 hours in monazite warehouse. Due to need of samples transportation the time interval between the finishing of sampling and beginning of experiment was approximately 3 h. To estimate the type of ^{212}Pb aerosols their dissolution from aerosol filter in imitation of body fluids was studied. The filter was placed in Ringer solution at 37°C for some time. Then the filter was rinsed by clean Ringer solution at the same temperature and its activity was determined by gamma spectrometer. After activity measurements the filter again was placed in solution and the cycles of dissolution and activity measurements were continued. It was obtained that the decrease of ^{212}Pb activity on the filter can be described by superposition of two exponential dependencies

$$\frac{A(t)}{A_0} = k \cdot \exp[-(\lambda_0 + \lambda_f)] + (1 - k) \cdot \exp[-(\lambda_0 + \lambda_s)] \qquad (1)$$

where

A_0 is the initial filter activity

$A(t)$ is the filter activity after dissolution during time t,

λ_0 is the ^{212}Pb decay constant,

λ_f is the rate of fast dissolution,

λ_s is the rate of slow dissolution

k is the part of activity with fast rate of dissolution.

The parameters of dissolution kinetic are presented in the Table 1.

TABLE 1. PARAMETERS OF ^{212}Pb AEROSOL DISSOLUTION FROM FILTER IN RINGER SOLUTION AT 37°C (8 SAMPLES)

Parameter	Average	Minimum	Maximum	Standard deviation
λ_f, day^{-1}	145	79	278	60
λ_s, day^{-1}	2.0	1.1	3.9	1.0
k	0.30	0.21	0.45	0.08

The ICRP model of nuclide transfer from respiratory tract to body fluids is presented in Fig. 1 [4]. It was supposed that the kinetic of ^{212}Pb absorption from the respiratory tract to body fluids can be described by the same mechanism as ^{212}Pb dissolution from aerosol filter to Ringer solution. So for the calculations of ^{212}Pb dynamic in respiratory tract the values $f_r = k = 0.3$; $s_r = \lambda_f = 145$ day^{-1}; $s_s = \lambda_s = 2$ day^{-1} were assumed. The aerosol deposition in respiratory tract in dependence on AMAD, type of breathing (nose or mouth breather) and intensity of work was taken from ICRP tables [4]. The nuclide dynamic and absorbed and equivalent doses in organs and tissues were calculated using standard WinAct and DCAL 8.4 software. The estimated values of dose coefficients (Sv/Bq) for inhalation intake of ^{212}Pb aerosols are presented in Tables 2–4 (notation E-09 means $\cdot 10^{-9}$). The estimated values of dose coefficients for real ^{212}Pb aerosols are between the reference values for Type F and Type M materials [5].

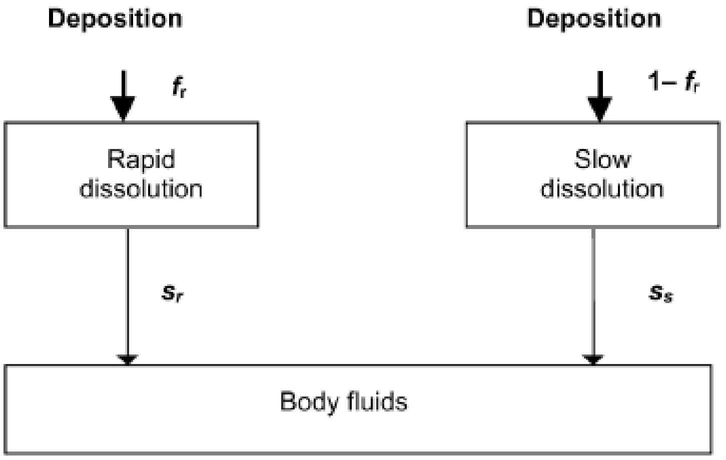

Fig. 1. ICRP model of nuclide transfer from respiratory tract to body fluids [4]

TABLE 2. DOSE COEFFICIENTS FOR INHALATION INTAKE OF REAL ^{212}Pb AEROSOLS, AMAD = 0.3 μm (Sv/Bq)

Organs and tissues	Heavy work		Light work	
	Mouth breathing	Nose breathing	Mouth breathing	Nose breathing
Stomach wall	1.87E-09	1.91E-09	1.90E-09	2.04E-09
Small intestine wall	1.94E-09	2.06E-09	1.97E-09	2.19E-09
Upper large intestine	2.32E-09	2.78E-09	2.35E-09	2.93E-09
Lower large intestine	2.38E-09	2.92E-09	2.41E-09	3.07E-09
Kidneys	4.29E-08	4.25E-08	4.35E-08	4.55E-08
Liver	1.19E-08	1.19E-08	1.20E-08	1.27E-08
Extra thoracic region ET1	4.07E-05	1.51E-04	4.36E-05	1.60E-04
Extra thoracic region ET2	1.24E-08	1.89E-08	1.28E-08	1.96E-08
Lymph nodes extra thoracic	1.99E-09	2.44E-09	2.03E-09	2.60E-09
Bronchial region (basal)	5.16E-07	4.43E-07	4.85E-07	4.21E-07
Bronchial region (secretory)	9.49E-07	8.15E-07	8.95E-07	7.77E-07
Bronchiolar region	7.42E-07	6.89E-07	8.45E-07	7.93E-07
Alveolar region	5.47E-08	5.12E-08	5.47E-08	5.48E-08
Lymph nodes thoracic	1.88E-09	1.88E-09	1.91E-09	2.01E-09
Lungs at whole	5.09E-07	4.56E-07	5.29E-07	4.82E-07
Bone Surfaces	4.47E-08	4.47E-08	4.54E-08	4.78E-08
Red Marrow	5.68E-09	5.68E-09	5.76E-09	6.08E-09
Effective dose	6.42E-08	5.90E-08	6.66E-08	6.24E-08

TABLE 3. DOSE COEFFICIENTS FOR INHALATION INTAKE OF REAL ^{212}Pb AEROSOLS, AMAD = 1 μm (Sv/Bq)

Organs and tissues	Heavy work		Light work	
	Mouth breathing	Nose breathing	Mouth breathing	Nose breathing
Stomach wall	2.38E-09	2.26E-09	2.38E-09	2.30E-09
Small intestine wall	2.61E-09	2.76E-09	2.60E-09	2.79E-09
Upper large intestine	3.74E-09	5.02E-09	3.65E-09	5.03E-09
Lower large intestine	3.96E-09	5.50E-09	3.86E-09	5.51E-09
Kidneys	5.18E-08	4.47E-08	5.22E-08	4.57E-08
Liver	2.95E-08	1.29E-08	1.47E-08	1.32E-08
Extra thoracic region ET1	1.28E-04	4.65E-04	1.37E-04	4.94E-04
Extra thoracic region ET2	3.32E-08	4.59E-08	3.00E-08	4.93E-08
Lymph nodes extra thoracic	2.76E-09	3.88E-09	2.81E-09	4.04E-09
Bronchial region (basal)	1.74E-06	9.86E-07	1.28E-06	6.14E-07
Bronchial region (secretory)	3.19E-06	1.80E-06	2.34E-06	1.12E-06
Bronchiolar region	6.25E-07	4.15E-07	6.78E-07	4.39E-07
Alveolar region	5.51E-08	3.72E-08	5.87E-08	4.08E-08
Lymph nodes thoracic	2.31E-09	2.03E-09	2.32E-09	2.07E-09
Lungs at whole	1.05E-06	6.15E-07	8.49E-07	4.49E-07
Bone Surfaces	5.49E-08	4.89E-08	5.52E-08	4.99E-08
Red Marrow	6.97E-09	6.21E-09	7.01E-09	6.32E-09
Effective dose	1.31E-07	8.12E-08	1.07E-07	6.15E-08

TABLE 4. DOSE COEFFICIENTS FOR INHALATION INTAKE OF REAL ^{212}Pb AEROSOLS, AMAD = 5 μm (Sv/Bq)

Organs and tissues	Heavy work		Light work	
	Mouth breathing	Nose breathing	Mouth breathing	Nose breathing
Stomach wall	3.91E-09	2.78E-09	3.77E-09	2.63E-09
Small intestine wall	4.81E-09	3.74E-09	4.61E-09	3.57E-09
Upper large intestine	8.89E-09	7.99E-09	8.41E-09	7.73E-09
Lower large intestine	9.75E-09	8.91E-09	9.21E-09	8.63E-09
Kidneys	7.58E-08	4.92E-08	7.37E-08	4.62E-08
Liver	2.21E-08	1.48E-08	2.14E-08	1.39E-08
Extra thoracic region ET1	3.20E-04	9.88E-04	3.49E-04	9.88E-04
Extra thoracic region ET2	1.12E-07	9.60E-08	1.03E-07	8.93E-08
Lymph nodes extra thoracic	4.71E-09	6.29E-09	4.73E-09	6.15E-09
Bronchial region (basal)	3.46E-06	1.35E-06	2.75E-06	6.76E-07
Bronchial region (secretory)	6.34E-06	2.46E-06	5.05E-06	1.24E-06
Bronchiolar region	8.63E-07	3.08E-07	9.01E-07	2.77E-07
Alveolar region	4.57E-08	2.03E-08	4.91E-08	2.08E-08
Lymph nodes thoracic	3.52E-09	2.32E-09	3.40E-09	2.17E-09
Lungs at whole	1.93E-06	7.44E-07	1.62E-06	4.18E-07
Bone Surfaces	8.37E-08	5.61E-08	8.11E-08	5.28E-08
Red Marrow	1.06E-08	7.12E-09	1.03E-08	6.70E-09
Effective dose	2.41E-07	1.01E-07	2.03E-07	8.13E-08

The possible distributions of dose coefficient values for respiratory tract due to uncertainties of dissolution parameters were calculated by Monte-Carlo method using specially developed software. Two kinds of parameter distribution were considered: uniform distribution and normal distribution. For parameter λ_s the bounded normal distribution was used so only positive values of parameter were considered. The results are presented in Fig. 2.

The DCF values from thoron daughters exposure to effective dose in dependence on air exchange rate λ_υ and attachment rate of free atoms to aerosols λ_χ were calculated by method described in [1]. The dependences of DCF for adult workers as a function of air exchange rate λ_υ, attachment rate of free atoms to aerosols λ_χ and lung type ^{212}Bi aerosols are presented in Fig. 3. It should be noted that the DCF values, based on experimental assessment of ^{212}Pb absorption from the respiratory tract to body fluids, considerably exceed the range 32–40 nSv per Bq·h·m^{-3} recommended by UNSCEAR [2, 3]. Taking into account the reevaluated DCF values the internal exposure due to inhalation of thoron daughters in monazite storage facility is comparable with external exposure. In this case very important problem of radiological protection of workers is the individual monitoring of internal exposure due to inhalation intake of thoron daughters (especially ^{212}Pb). The typical techniques of individual monitoring of internal exposure are the estimation of nuclide inhalation intake by its retention (in lungs or whole body) or excretion (fecal or urine) [6].

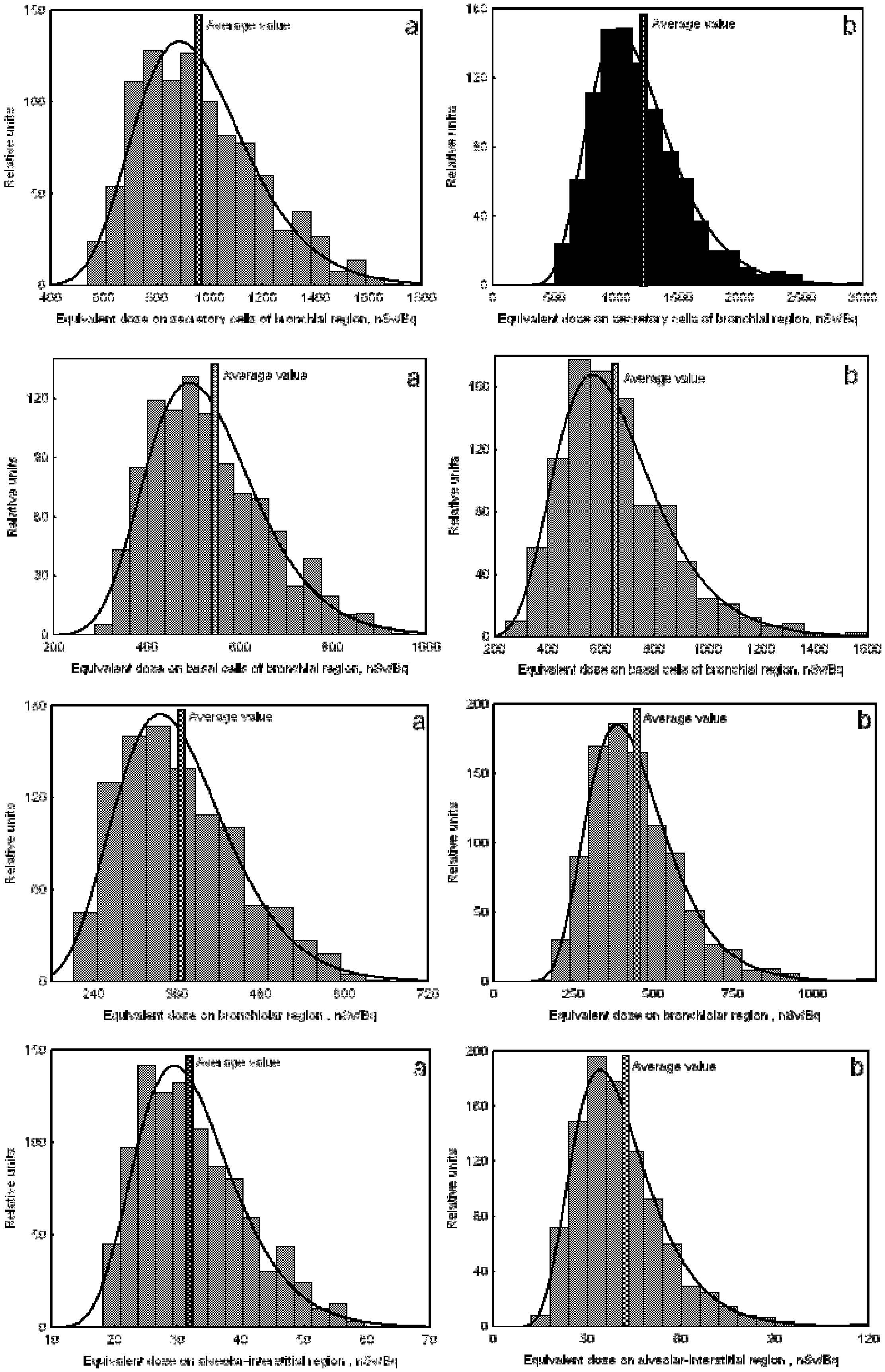

FIG. 2. Distributions of dose coefficients for respiratory tract due to uncertainties of ^{212}Pb dissolution parameters (AMAD=1 μm, nose breather, light work); a – uniform distribution of parameters, b – normal distribution of parameters

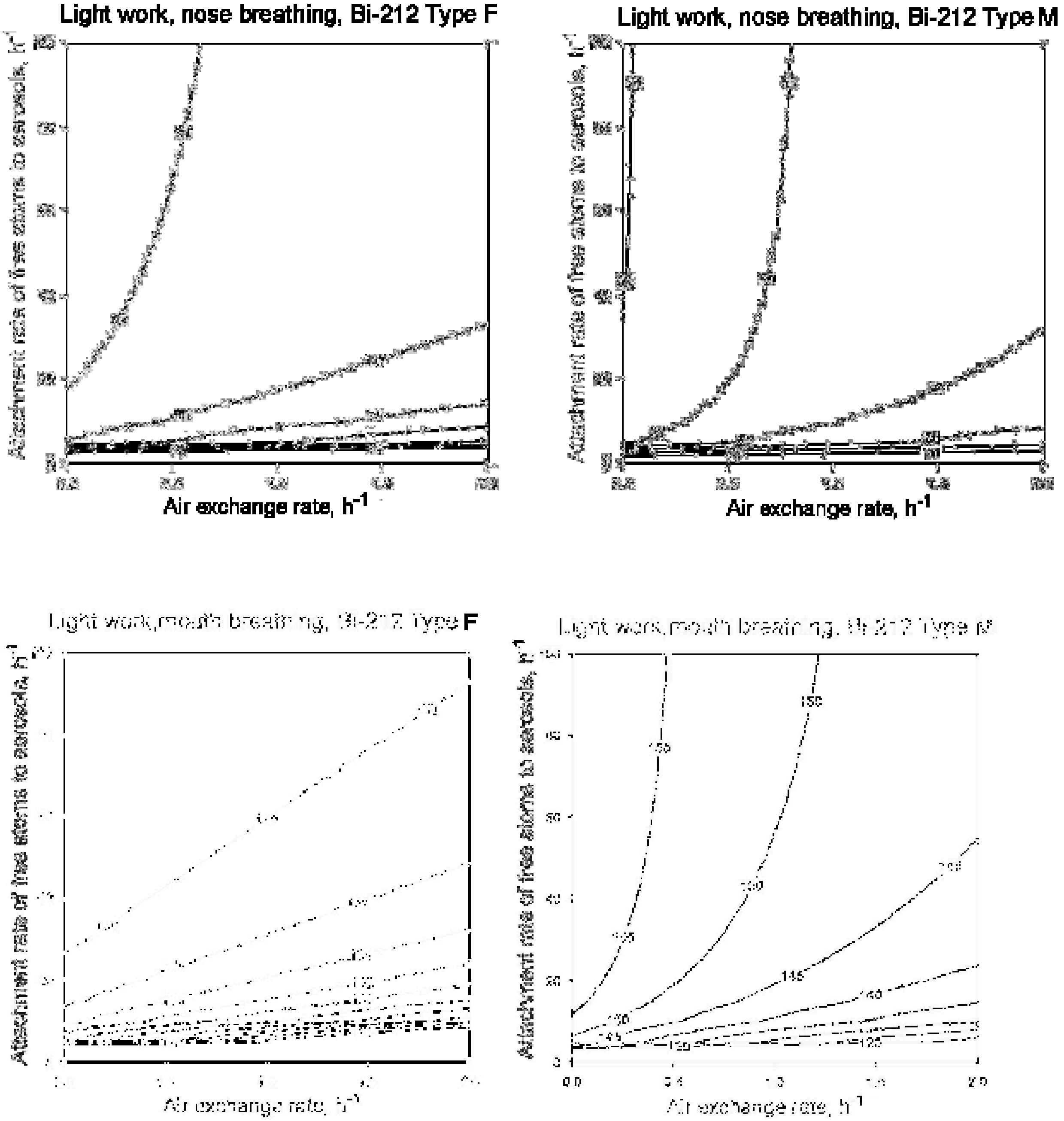

Fig. 3. The dependences of DCF from thoron daughters exposure to effective dose for adult workers as a function of air exchange rate λ_{υ} and attachment rate of free atoms to aerosols λ_{χ}, nSv per Bq·h·m^{-3} (AMAD = 1 μm)

The estimated value of ^{212}Pb inhalation intake during the working shift in monazite storage facility is nearly 1000 Bq. The half-life of ^{212}Pb $T_{1/2}$ = 10.6 h is comparable with working shift duration. So the traditional calculation of nuclide dynamics for single intake is not suitable for internal monitoring program purposes. All calculations of nuclide dynamic for estimated ^{212}Pb absorption rate from the respiratory tract to body fluids were performed for uniform inhalation intake during 6 h working shift. During the calculations the next factors were considered:

- The aerosol dispersion: AMAD 0.3 (AMTD 0.19), 1 and 5 μm;
- The type of breathing: nose and mouth breather;
- The intensity of work: light exercise (breathing rate 1.2 m^3/h) and heavy exercise (breathing rate 1.7 m^3/h).

The system of differential equations describing the nuclide dynamic in respiratory and gastro-intestinal tracts, organs and tissues of body [4, 7, 8] was solved. The samples of nuclide retention as well as faecal and urine excretion dynamics are presented in Fig. 4 and Table 5. The calculation results are presented as a part of total uniform intake during a 6 h working shift. The beginning of time countdown corresponds to the beginning of working shift.

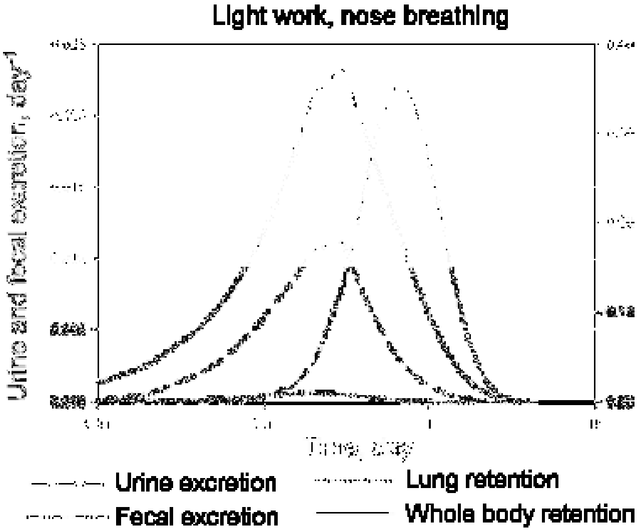

FIG. 4. Dynamics of ^{212}Pb whole body and lung retention and fecal and urine excretion for estimated nuclide absorption rate from the respiratory tract to body fluids (AMAD = 1 μm)

TABLE 5. FRACTION OF ^{212}Pb INTAKE (TAKING RADIOACTIVE DECAY INTO ACCOUNT) RETAINED IN THE BODY AND TRANSFERRED TO URINE AND FAECES DURING SAMPLING PERIOD τ

AMAD (μm)	Exercise	Breathing type	Whole body retention		Urinary excretion			Faecal excretion
			τ = 12 h	τ = 24 h	τ = 8 h	τ = 12 h	τ = 24 h	τ = 24 h
0.3	Heavy	Mouth	0.121	0.052	0.0017	0.0022	0.0017	0.0014
		Nose	0.147	0.060	0.0017	0.0022	0.0017	0.0028
	Light	Mouth	0.123	0.053	0.0017	0.0022	0.0017	0.0014
		Nose	0.156	0.064	0.0018	0.0024	0.0018	0.0029
1.0	Heavy	Mouth	0.164	0.069	0.0021	0.0027	0.0021	0.0030
		Nose	0.249	0.095	0.0021	0.0027	0.0019	0.0093
	Light	Mouth	0.180	0.075	0.0021	0.0028	0.0021	0.0042
		Nose	0.255	0.097	0.0021	0.0027	0.0020	0.0092
5.0	Heavy	Mouth	0.404	0.162	0.0048	0.0058	0.0040	0.0168
		Nose	0.401	0.145	0.0027	0.0033	0.0023	0.0176
	Light	Mouth	0.358	0.142	0.0035	0.0045	0.0032	0.0156
		Nose	0.391	0.141	0.0025	0.0032	0.0022	0.0172

3. Conclusions

1. The dissolution of real ^{212}Pb aerosols in imitation of body fluids can be described by superposition of two exponential dependencies with the rate of fast dissolution λ_f = 145 day^{-1}; rate of slow dissolution λ_s = 2 day^{-1} and the part of activity with fast rate of dissolution k = 0.3.
2. The estimated values of DCF from thoron daughter exposure to effective dose 80–150 nSv per Bq·h·m^{-3} at least 3–5 times exceeds the DCF values recommended by UNSCEAR.
3. Most perspective monitoring method of inhalation intake of ^{212}Pb aerosols is the direct measurements of whole body activity. Assessment of ^{212}Pb inhalation intake by urinary excretion is possible for sample collection time from 8 to 12 h. Even in this case the total activity of the sample will be in the range 1.5 – 3 Bq for total intake during the working shift ~1000 Bq. The estimation of inhalation intake of ^{212}Pb aerosols by measuring of fecal excretion can be efficient and non-depended on type of breathing and intensity of work only for coarse aerosols (AMAD ≈5 μm).

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