Eduard Hanslík

Removal of natural radionuclides by water treatment processes, consequences with occupational health

T. G. MASARYK WATER RESEARCH INSTITUTE

T. G. Masaryk Water Research Institute, p.r.i.
Podbabská 2582/30, 160 00 Prague 6, Czech Republic | +420 220 197 111 | info@vuv.cz, www.vuv.cz
Brno Branch | Mojmírovo náměstí 16, 612 00 Brno | +420 541 126 311 | info_brno@vuv.cz
Ostrava Branch | Macharova 5, 702 00 Ostrava | +420 595 134 800 | info_ostrava@vuv.cz

Goals

Sources of natural radioactivity in groundwater used for drinking purposes in Czech Republic

Legal framework

Treatment technologies for removal radionuclides from groundwater

Case study – Treatment plant at Central Bohemia

Consequences with occupational health

Sources of drinking water at Czech Republic

Sources of public water supply:

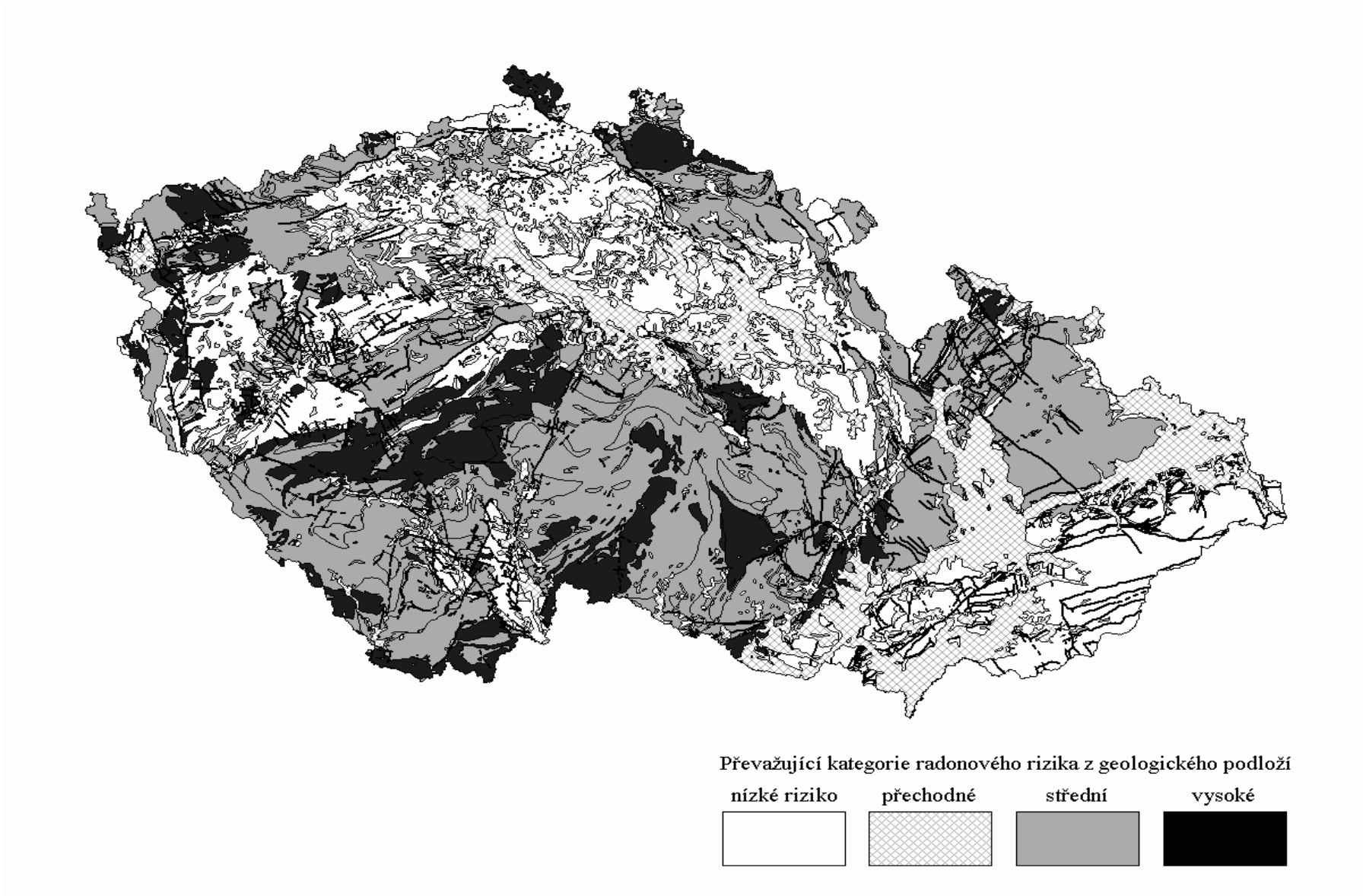
- **Surface water ~ 60%**
- **Ground water ~ 40 %**

Natural radioactivity of surface water (average values)

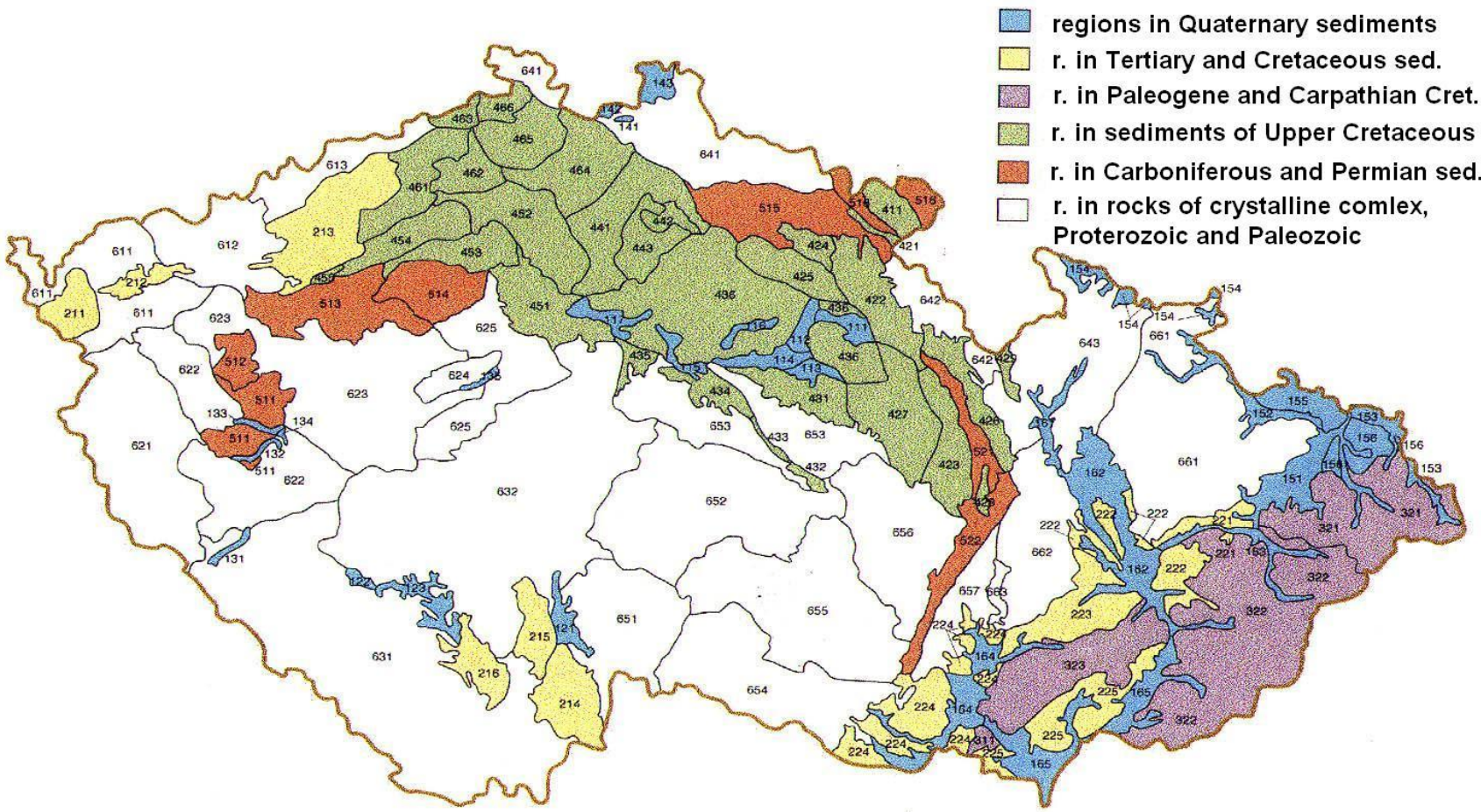
Radon: 2 Bq/l
Uranium: less than 2 μg/l
Ra-226, Ra-228: less than 10 mBq/l
K-40: 140 mBq/l
K: 5 mg/l

Natural radionuclides in treated surface water perform negligible impact on occupational health.

Geological map for prediction of Radon Risk



HYDROGEOLOGICAL REGIONS OF CZECH REPUBLIC



Zpracoval: CHMU a Výzkumný ústav vodohospodářský TGM, 1997
Kartografické zpracování: AKVA-KART, spol. s r. o., 1997

Artificial radioactivity in surface water on the level of 2013 (average values)

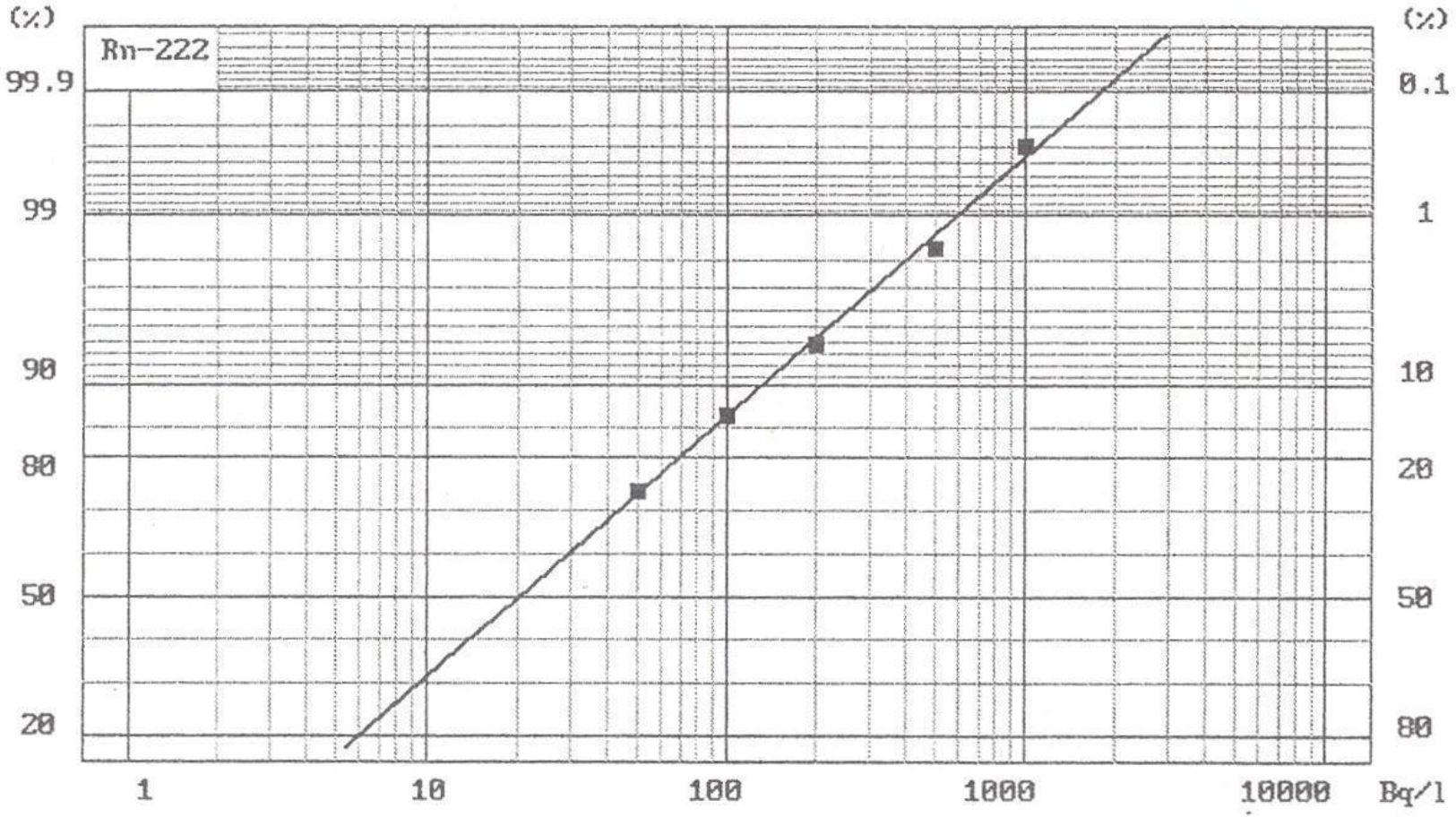
Tritium: 1 Bq/l (background) decreasing trend
5 – 100 Bq/l below nuclear devices
Strontium-90: about 1 mBq/l decreasing trend
Caesium-137: about 1 mBq/l decreasing trend

Artificial radionuclides in treated surface water perform negligible impact on occupational health.

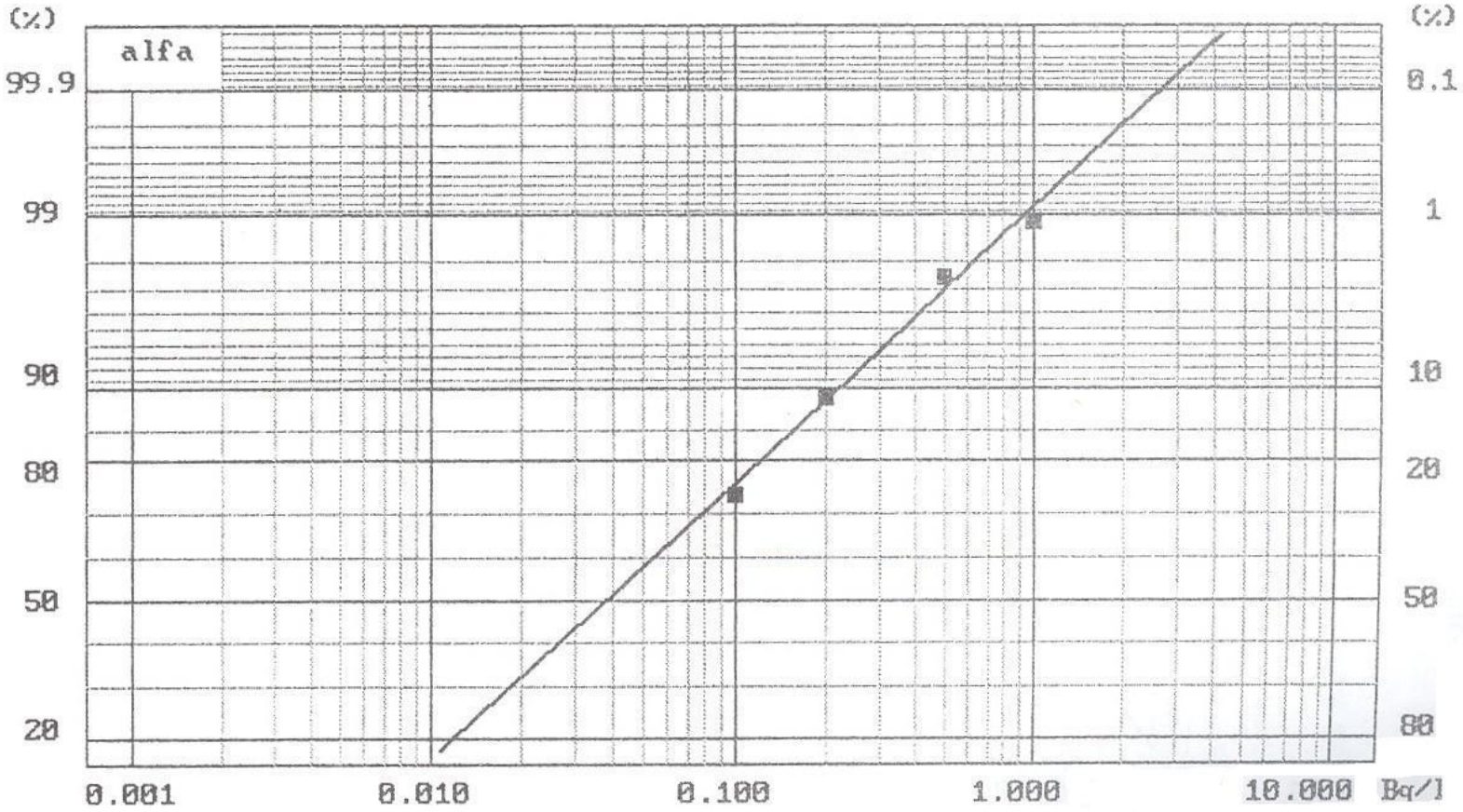
Natural radioactivity at ground water (range of values)

^{222}Rn: **10 – 4000 Bq/l**
α: **0.1 – 10 Bq/l**
β: **0.1 - 10 Bq/l**
^{226}Ra: **0.02 – 0.25 Bq/l**
^{228}R: **0.02 – 0.25 Bq/l**
Uranium: **1 – 100 µg/l**

Natural radionuclides in treated groundwater perform for occupational health impact from ^{222}Rn inhalation and dose rate from filter media and sludge.



Distribution of public water suply in Czech Republic according to Radon concentration in water (SÚJB 1996)



Distribution of public water upply in CR according to gross alpha concentration (SÚJB 1996)

Decree No. 307/2002 Coll. on Radiation Protection

Table 4
The guidance levels of volume activities and TID in drinking water

Indicator of radioactivity	The guidance levels		
	Packaged infant water	Packaged water, water intended for public supply	Packaged natural mineral water
Radon	20 Bq/l	50 Bq/l	100 Bq/l
Gross alpha	0,1 Bq/l	0,2 Bq/l	0,5 Bq/l
Tritium		100 Bq/l	
TID *)		0,1 mSv/y	
Gross beta excluding radioactivity of ^{40}K	0,1 Bq/l	0,5 Bq/l	1,0 Bq/l

*) TID is assumed to be less than the parametric indicator value of 0,1 mSv/y, when the gross alpha and the gross beta are less than the guidance levels

Table 5
The maximum permitted levels of volume activities upon the exceeding of which water must not be supplied

Radionuclide	The maximum permitted levels [Bq/l]		
	Packaged infant water	Packaged water, water intended for public supply	Packaged natural mineral water
Ra-226	0.4	1.5	3
Ra-228	0.1	0.5	1

Workplaces with a Possibility of Significantly Increased Exposure to Natural Sources

…

c) workplaces, namely pumping stations, spa facilities, filling rooms, **water treatment plants where underground water is handled by pumping, collecting or by other method**;

d) all workplaces where radon concentration of 400 Bq/m^3 has demonstrably been exceeded;

…

Investigative and guidance levels for exposure from natural sources

Investigative levels for workplaces with a possibility of significantly increased exposure to natural sources are laid down:

- average radon concentration of 400 Bq/m^3 for a work activity of the persons performing work at these workplaces
- 1 mSv for an effective dose per calendar year above the natural background, besides radon and his products

Guidance level is 6 mSv for an effective dose per calendar year; if this level could be exceeded, these are workplaces with significantly increased exposure to natural sources, than the radiation protection is ensured as in controlled area

Release of Natural Radionuclides from Workplaces with a Possibility of Significantly Increased Exposure to Natural Sources…

(1) During release of natural radionuclides from the workplaces with a possibility of significantly increased exposure to natural sources, the following shall preferably be monitored:

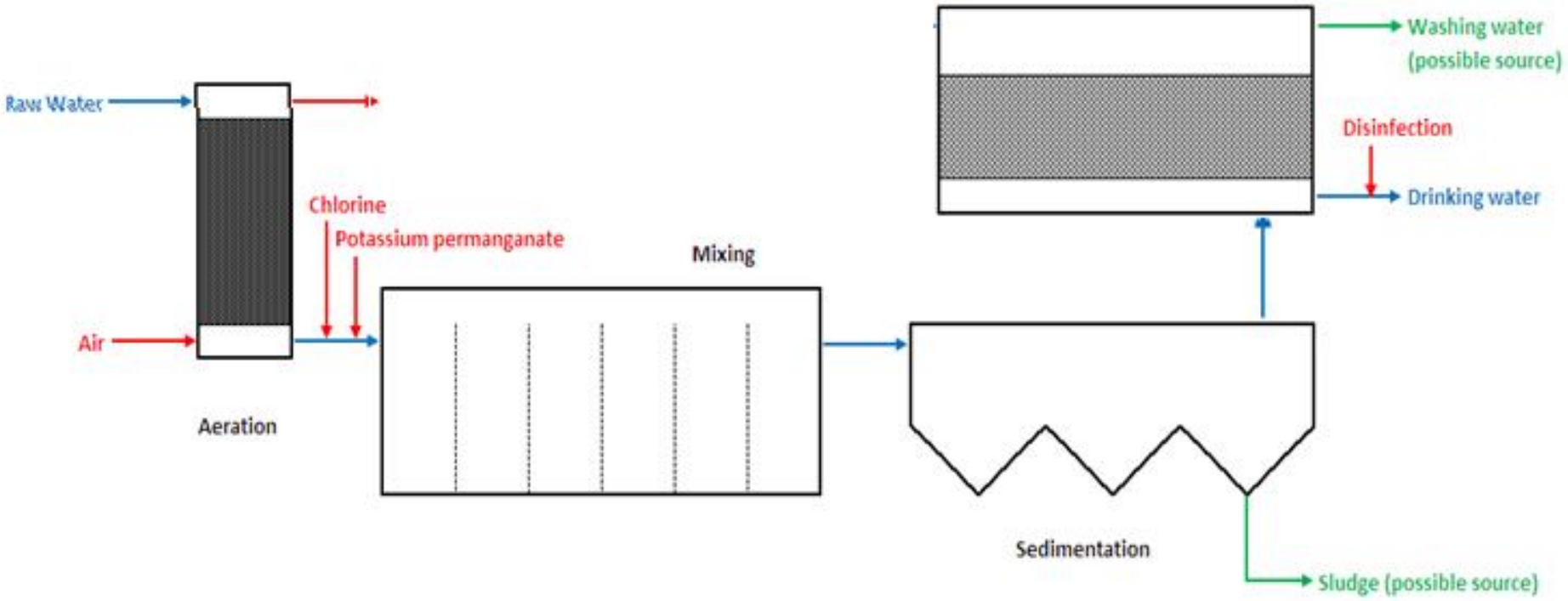
a) sediments and sludge in piping and storage systems, for example, in pumps, fittings, valves, collectors and separators;

…

3. Treatment technologies for radionuclides removal from drinking water

^{222}Rn
^{226}Ra
^{228}Ra
Uranium

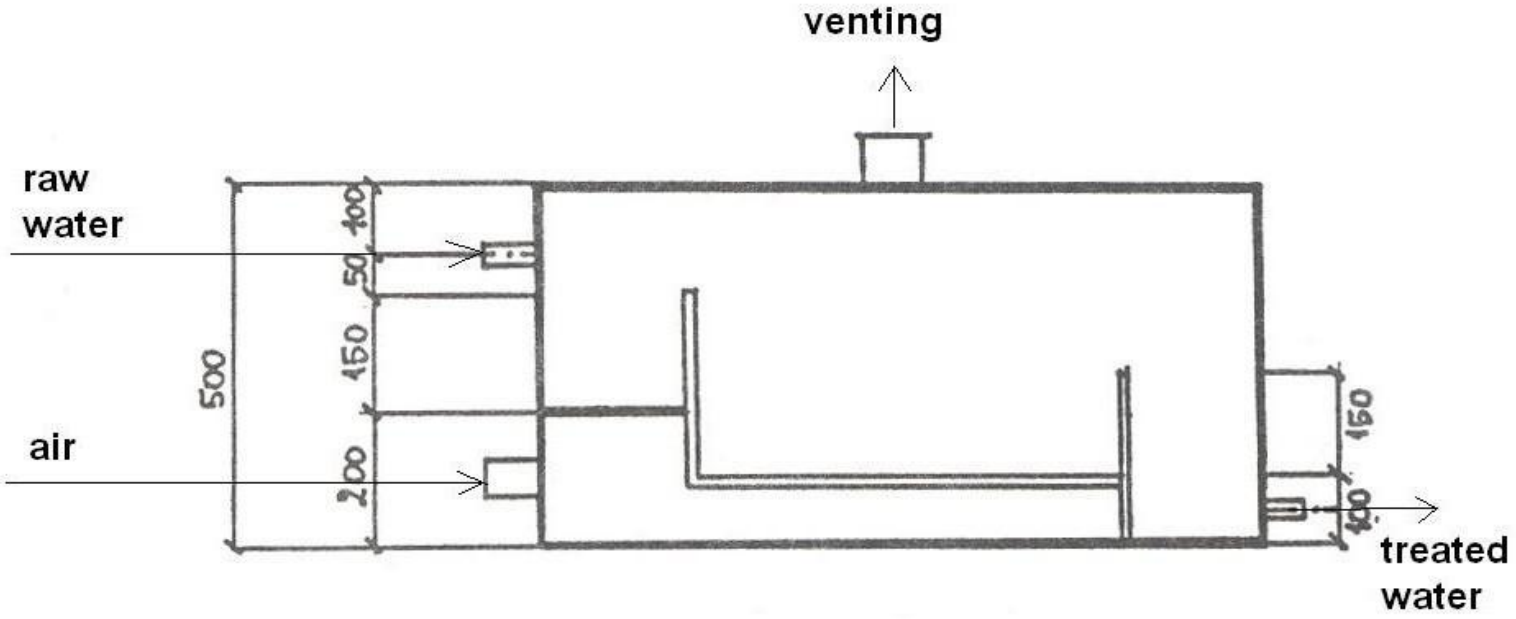
Water treatment technology – processes and device



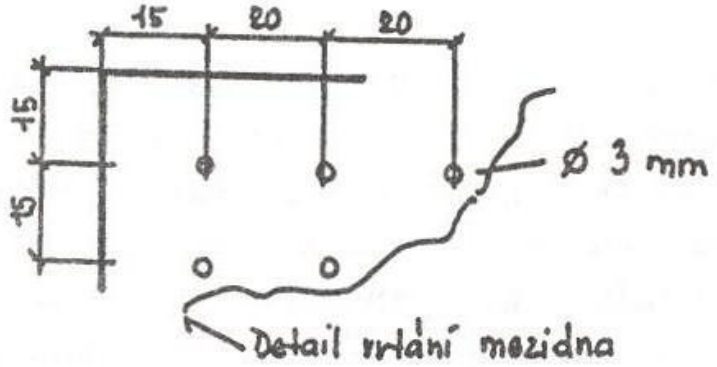
Radon – Aeration

<u>Aeration in shallow-water</u> **(bubble system)**
(INKA)

Efficiency: 90 %
Depends on: ratio Q_a/Q_w
mean residence time
diameter of bubbles
water temperature



perforating of
tray



Kinetics of first order

$$-\ln \frac{c_T}{c_R} = k_1 \frac{Q_a}{Q_w} + k_2$$

where

c_T	concentration of ^{222}Rn in treated water (Bq/l)
c_R	concentration of ^{222}Rn in raw water (Bq/l)
Q_a	flow rate on air (l/s)
Q_w	flow rate of water (l/s)
k_1,k_2	empirical parameters, end effect





Tower aeration

- **Parallel run of water and air 95 %**
- **Counter current of water and air**
  - **Efficiency: 98 %**
  - **Depends on: height of the tower**
    mean residence time

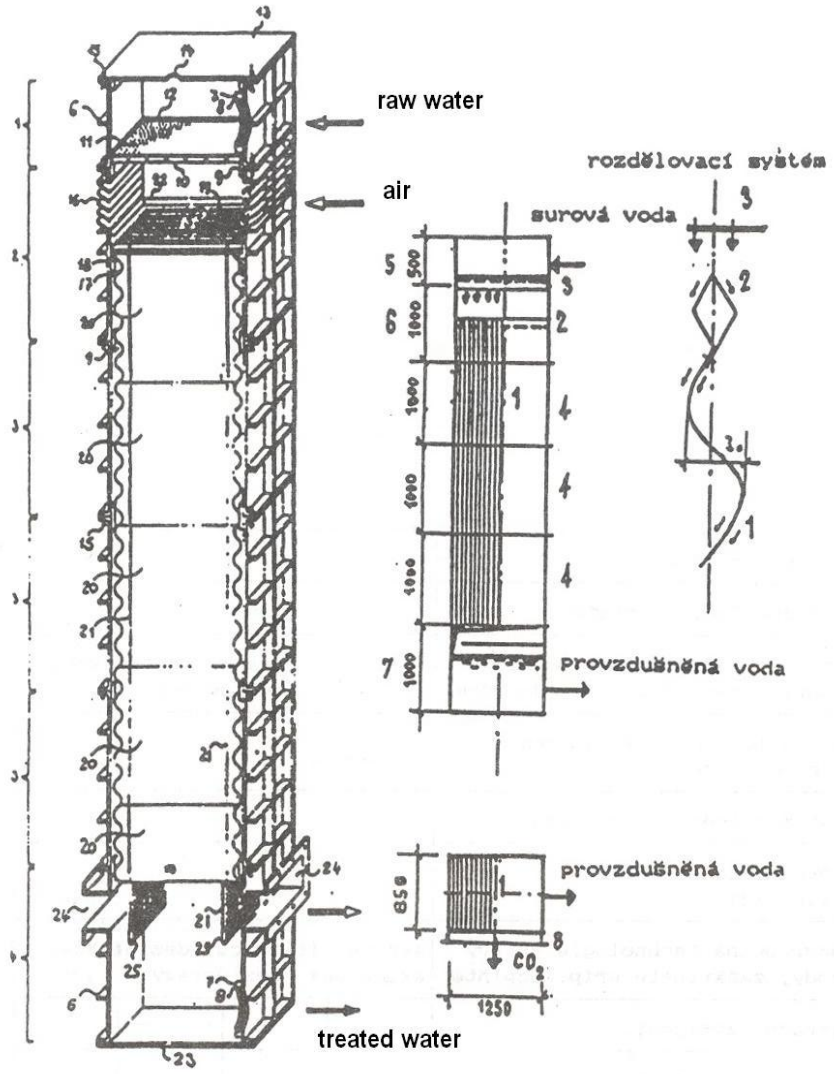
Height of aeration tower

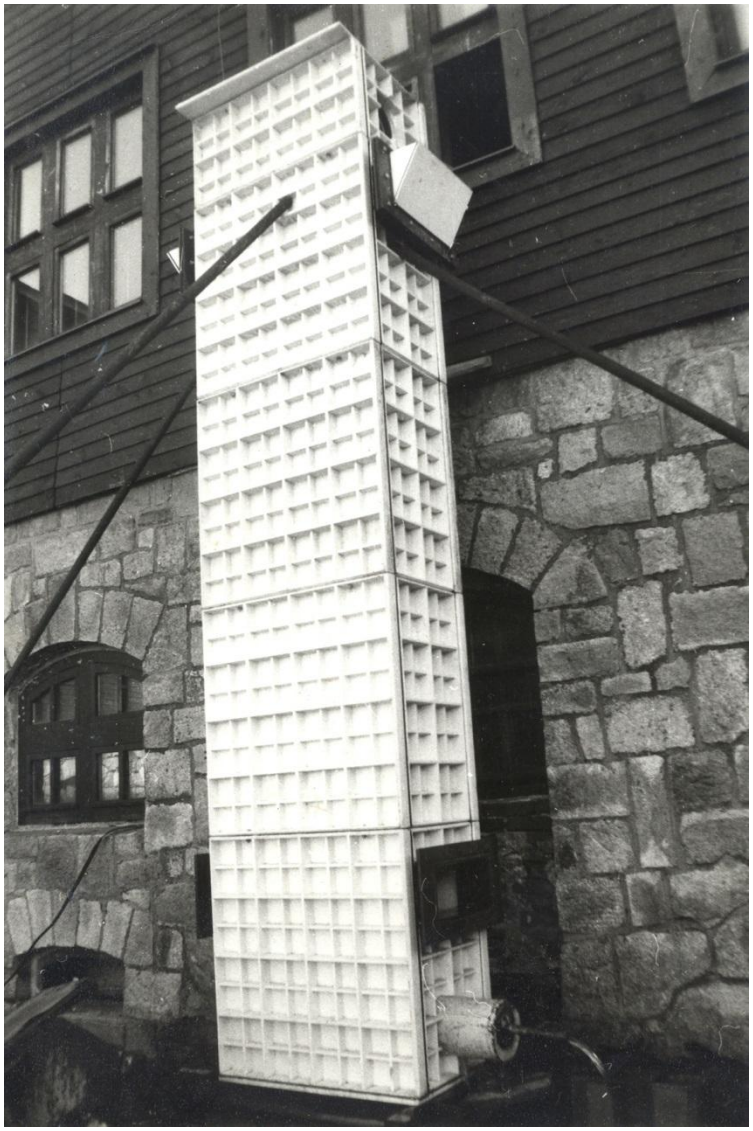
Equation used in chemical engineering

$$h = HTU.NTU$$

where	H	height of aeration tower (m)
	HTU	height of aeration unit (m)
	NTU	number of aeration unit

Parallel run of water and air







Aeration tower

Counter current of water and air



^{226}Ra, ^{228}Ra - Combined Radium and Iron removal

Removal of radium by filtration on sand coated by Fe and Mn oxides

Process of Iron and Manganese removal were used before knowledge of radium content in water and risk from intake by water consumption

Efficiency: 30 - 70 %

Depends on: water quality

Man made filter sand coated by Mn oxides has removal efficiency 70 % and more

Mechanism of radium isotopes sorption

$$M^{+} + x(=MeOH) \Leftrightarrow M(=MeO)x^{(n-x)^{+}} + xH^{+}$$

where	M	the sorbed metal ion
	(=MeOH), (=MeO)	hydroxides, oxides

Concentration of ^{226}Ra 0.2 Bq/l in raw water generated mass activity of filter media in equilibrium about 5000 Bq/kg.

Water treatment hall with open sand filters



Specific activity in filter media

Specific activity of ^{226}Ra and ^{228}Ra are in the range 1000 – 15000 Bq/kg

Risk for workers must be evaluated, study on water treatment plant show that dose rate increase from ^{226}Ra and ^{228}Ra is relatively small.

Uranium

Coagulation with Fe or Al hydroxides

Removal efficiency 80 % by pH about 6

Sorption - ion exchange

Removal efficiency 95 %

Capacity of uranium in filter media is approximately 5 g/kg

Anion exchange resin
Capacity for uranium 5 g/kg

$$R-(NR_2´.H_2O)+UO_2(CO_3)_2^{2-} \Leftrightarrow R-UO_2(CO_3)_2^{2-}+2H_2O$$

where	R	matrix of resins
	R´	hydrogen

Saturated resin is according Atomic act toxic matter, fissionable material respectively

Case study

Water treatment plant in Central Bohemia (Czech Republic)

6 open sand filters, each filter media has 30 m^3

Sand coated MnO_2 and Fe_2O_3

Methods

Raw and treated water samples were taken regularly, the filtration sand was sampled from the ibndividual filters.

A gammaspectrometric analysis, conducted according to the standard ČSN ISO 10703 (75 7630), determined concentration of ^{226}Ra and ^{228}Ra in water and sand samples. Canberra-Packard S 100 instrument with HpGe detector, was used.

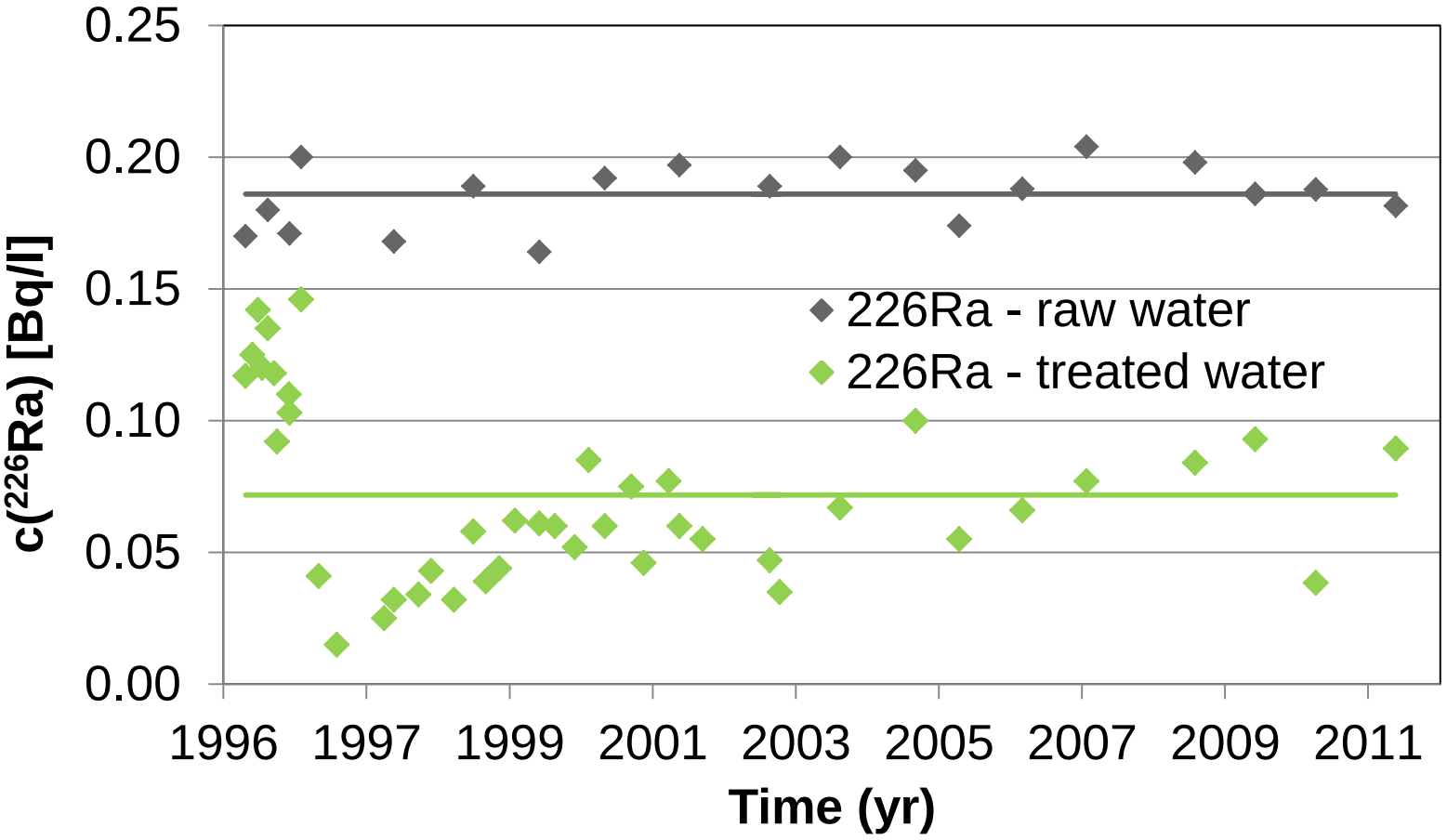
^{222}Ra concentration in the raw water and treated water was determined with the emanation method.

The ^{222}Rn concentration in air was measured directly in the drinking water treatment building, using an automatic monitor of radon concentration course. The minimal detectable activity is 30 Bq/m^3 (for the measuring time 1 hour, the statistical error 20 %).

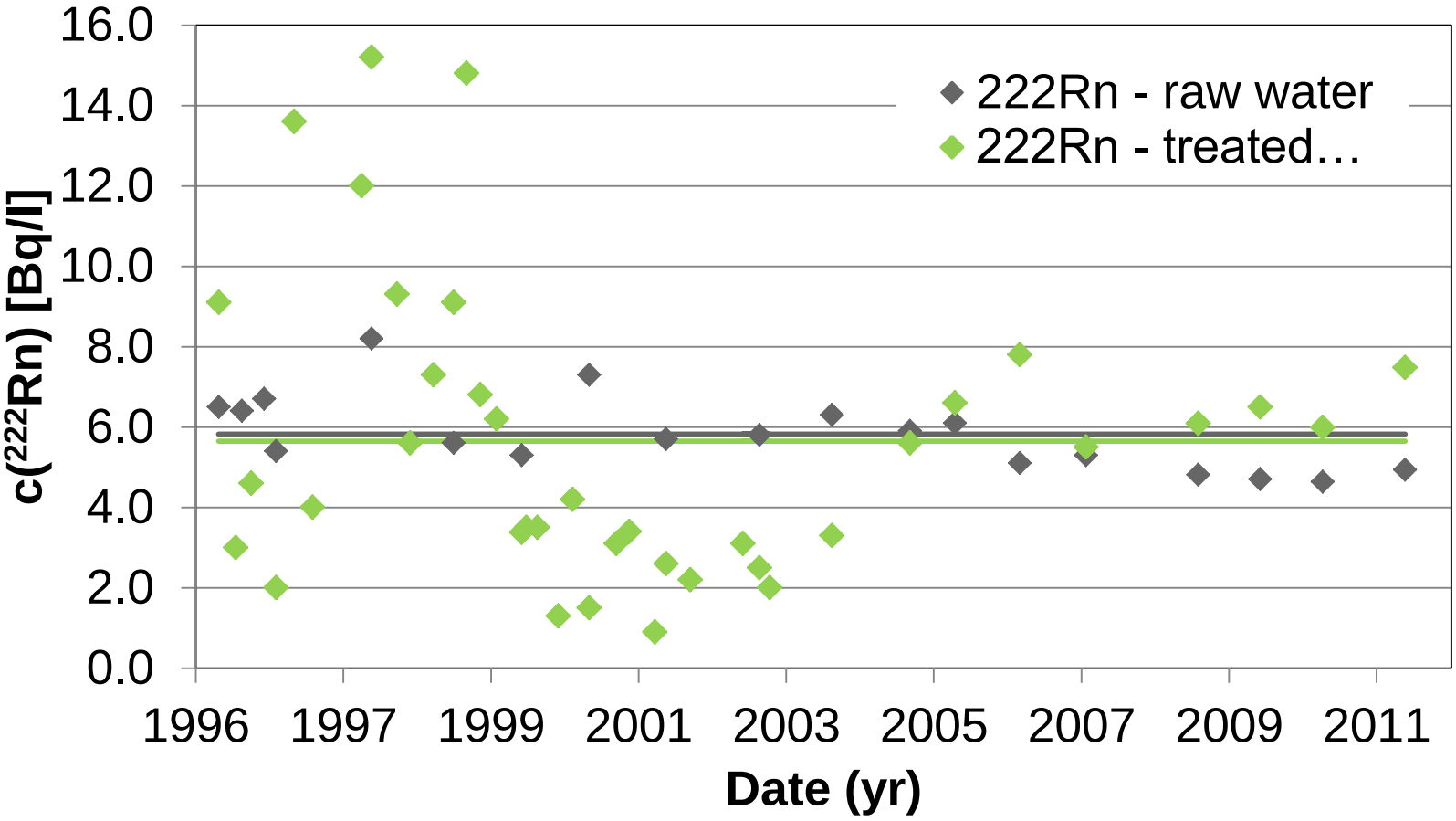
Dose rate was measured with monitor enables to measure the dose rates in range from 0.01 nGy/s to 30000 nGy/s.

The measuring instruments are regularly verified, as the legislation requires, by the Czech Metrological Institute.

Development of ^{226}Ra concentrations in raw (0.186 Bq/l) and treated (0.072 Bq/l) water in the monitored period, average removal efficiency 61 %



^{222}Rn concentrations in raw (5.8 Bq/l) and treated (5.65 Bq/l) water in the monitored period. Removal after 2. stage aeration 3 %



Contamination of water by ^{222}Rn emanating from filter sand

The magnitude of the secondary ^{222}Rn contamination of the treated water depends on the specific ^{226}Ra activity in the filter media, filter loading, medium detention time and emanation coefficient.

Assuming that the total ^{222}Rn, produced by the ^{226}Ra decay, is emanate into filtered water, the ^{222}Rn concentration in water treated can be calculated as:



where

c_{Rn}	is concentration of ^{222}Rn in water after filtration (Bq/l)
a_{Ra}	specific activity of ^{226}Ra in filtration sand (Bq/kg)
λ_{Rn}	decay constant of ^{222}Rn (0.00755/h)
t	time (h)
L	filter loading (l/kgh)
t_{det}	medium detention time of water in filter (h)
c_{0Rn}	concentration of ^{222}Rn in water before filtration (Bq/l)

1st member 2nd member



The first member of the simplified equation characterizes the emanation of ^{222}Rn by the decay of ^{226}Ra, retained in the filtration sand, and its simultaneous decay.

The second member of the equation describes the spontaneous radioactive decay of the ^{222}Rn, entering the gravity filter with raw water.

^{222}Rn concentrations in out flow water from filter measured and calculated for the period of 24 h following the filter washing

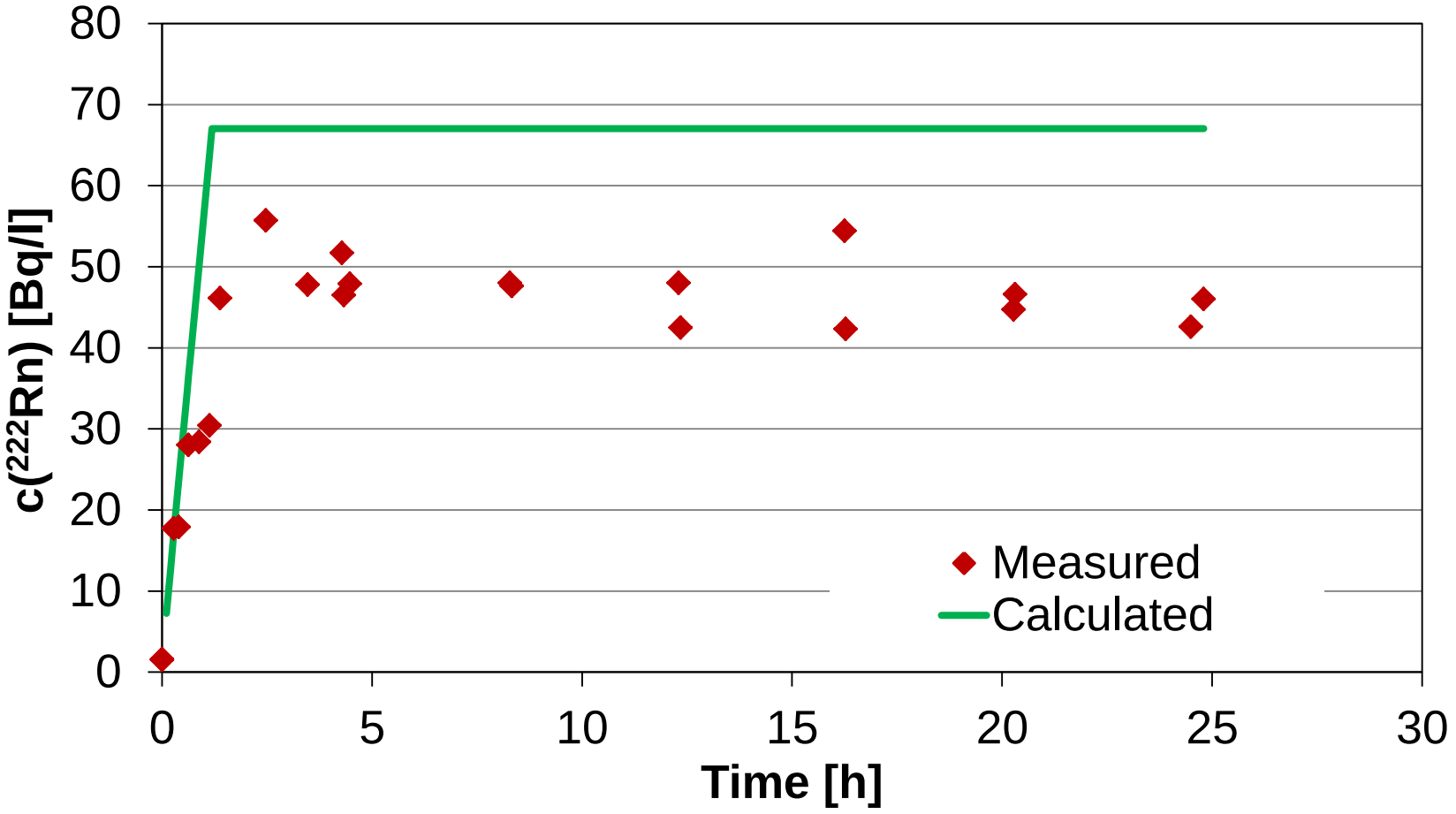


Figure shows that the theoretically calculated equilibrium values are higher than the actually measured ones. The average value of the measured equilibrium ^{222}Rn concentrations was 47.4 Bq/l, the corresponding calculated value was 67.0 Bq/l.

$$K_e = \frac{c_{Rn,measured} - c_{0Rn}}{c_{Rn,calculated} - c_{0Rn}}$$

where K_e is emanation coefficient of the filtration sand

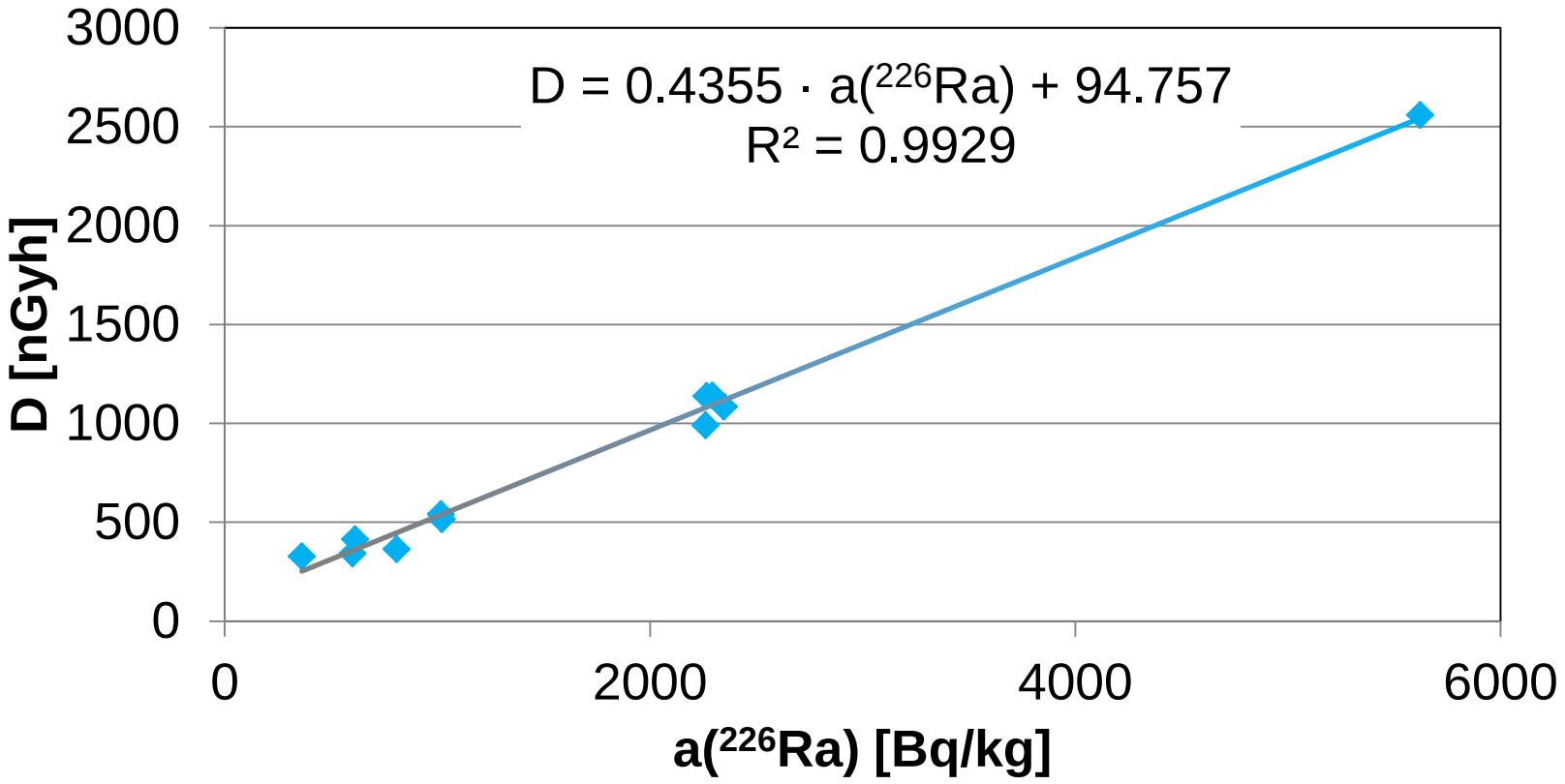
$c_{Rn,measured}$ concentration of ^{222}Rn in water, measured (Bq/l)

$c_{Rn,calculated}$ concentration of ^{222}Rn in water, calculated (Bq/l)

c_{0Rn} concentration of ^{222}Rn in water before filtration (Bq/l)

Emanation coefficient in case study was about 70 %.

The relation between the dose rate on surface of individual filters and the ^{226}Ra content in their filter sand was fitted with a linear regression. It can be noted from the figure that the relation is very tight.



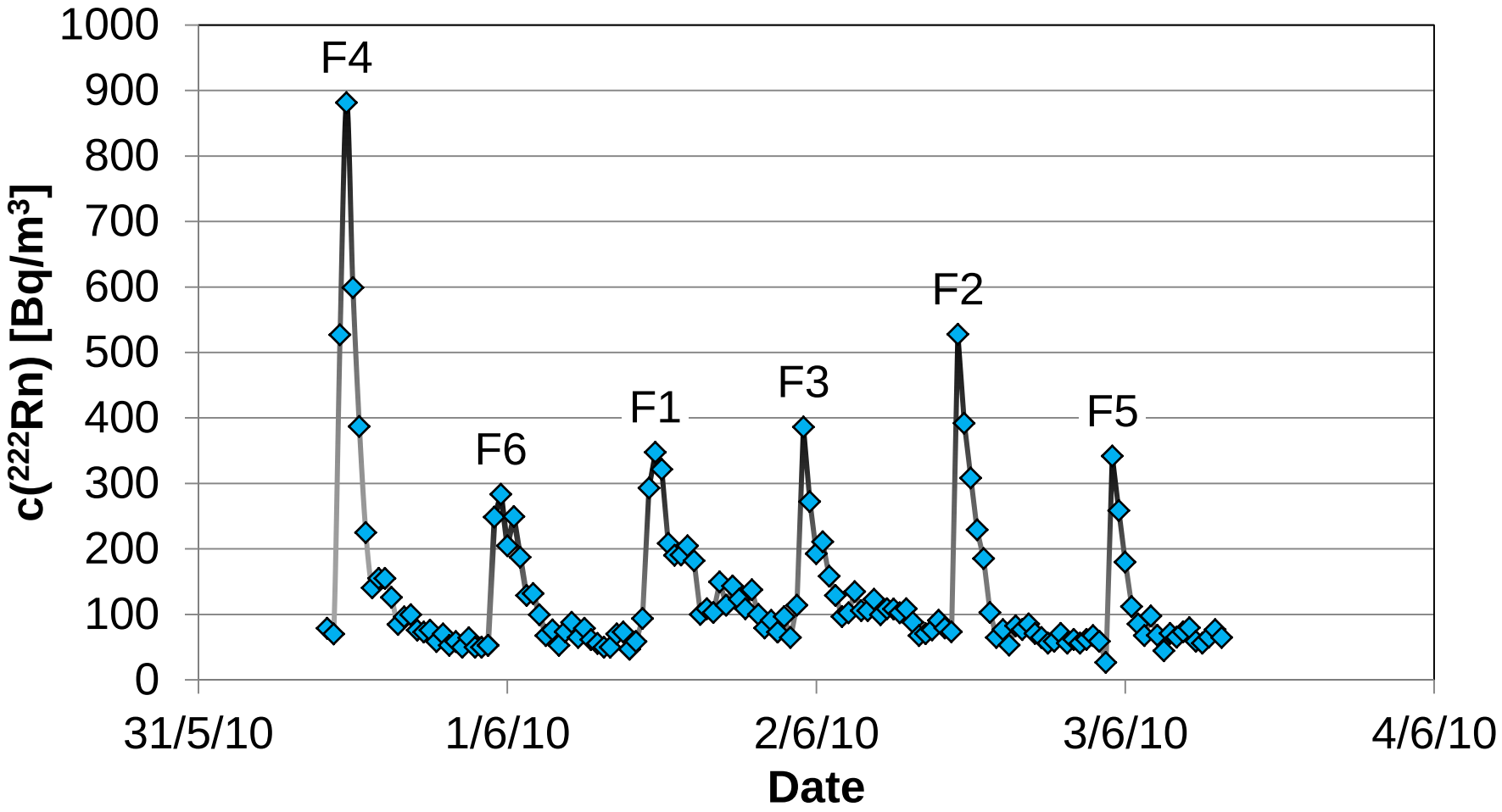
In the Plant, slow filtration is alternated with filter washing. Duration of all six filters washing cycle is 3 days. The filters are washed with a strong torrent creating a turbulent flow. During this process, the ^{222}Rn is vigorously released into the air. Rapid increase in the ^{222}Rn air concentration was detected, when a filter was being washed. When the filter washing process was finished, the ^{222}Rn concentration values dropped back quickly to the primary values.

The highest peaks were detected in the filtration hall itself.

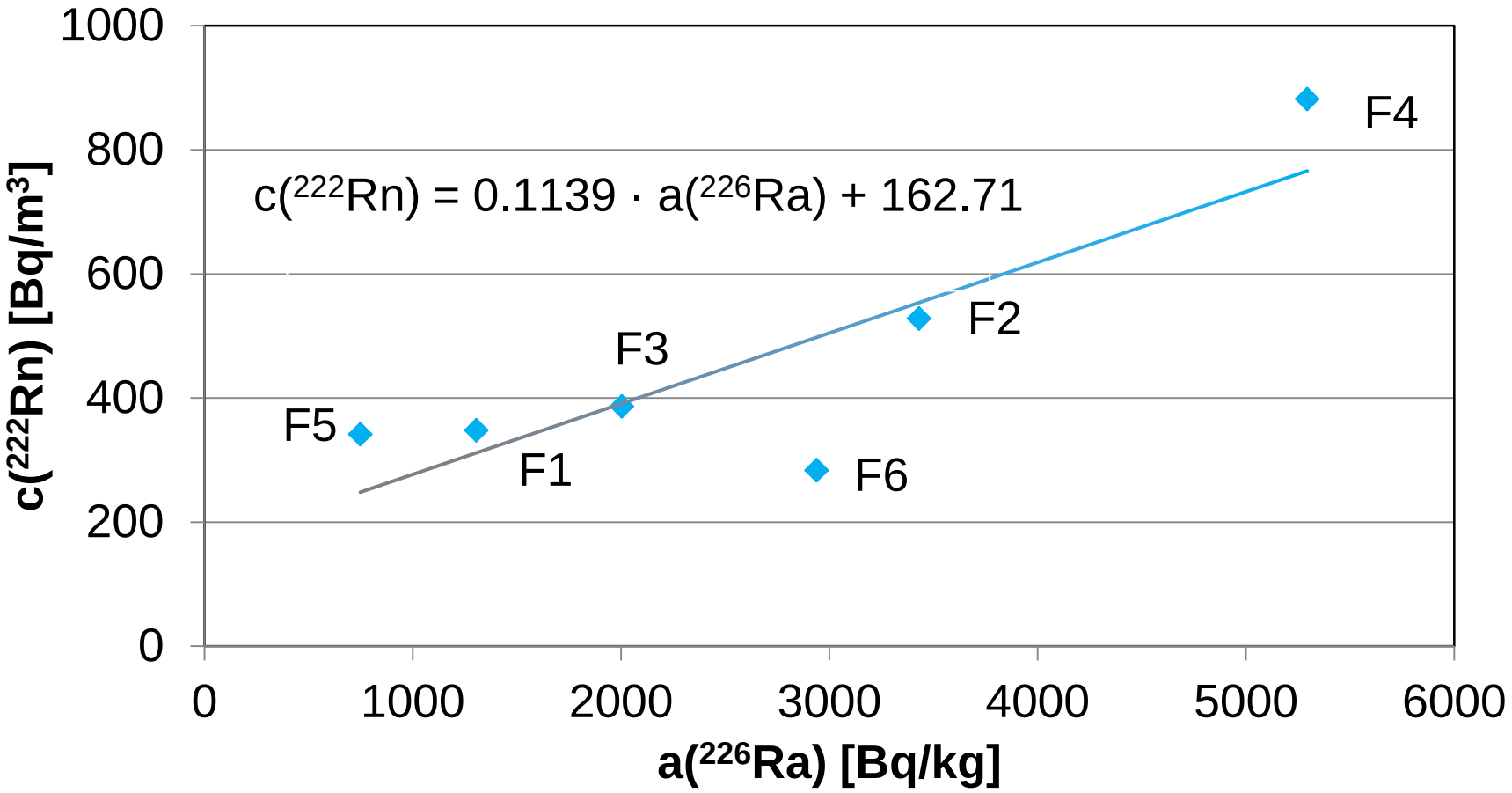
The maximal detected concentration of ^{222}Rn in air was 2 163 Bq/m^3. The average radon concentration was 141 Bq/m^3.

Further, the study assesses a relation between maximal ^{222}Rn concentrations in the air of the filtration hall and ^{226}Ra content in filter media of individual filters (F1 – F6).

^{222}Rn concentration in air in the filtration hall in the monitored period



Relation between the maximal ^{222}Rn air concentrations and ^{226}Ra activities in filters in short (3-day) period



Conclusion

Increased concentration of ^{222}Rn in air and ^{226}Ra and ^{228}Ra in filters represent the main health risk for the personnel of the ground water treatment plants.

The risk caused by ^{222}Rn inhalation can be significantly reduced by suitable ventilation (e.g. blowing the air out of the water treatment hall) and proper management of the personnel occupation time in the plant premises.

The risk of irradiation should be assessed, before manipulating with the filter media and sludge.