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Radon decay products in the boundary layer of the atmosphere

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Abstract. The concentration of radon and its decay products in the boundary layer of the atmosphere varies and depends on many factors. One of the main factors leading to the variability of equilibrium equivalent concentration (EEC) of radon is the dynamic behavior of the atmosphere. Describing the behavior of radon concentration and EEC in the atmosphere is important for the estimation of natural human exposition (this was our original goal). The National Institute for Nuclear, Chemical and Biological Protection (SÚJCHBO, v.v.i.) has been investigating the characteristics of radon decay products in the boundary layer of the atmosphere since 1985. The measurements of EEC were carried out in places with different exhalation of radon from the subsoil. The grab sampling method, as well as continuous measurement, were applied in various areas. The determination of outdoor radon and radon daughter product (RnDP) is not easy due to very low levels of their concentrations: therefore, many methods and devices had to be modified for this type of measurement.

Key words: natural radioactivity • atmosphere • radon daughter product (RnDP) • equilibrium equivalent concentration (EEC) • solid-state nuclear track detector (SSNTD) • long-lived radionuclides

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Introduction

The primary reason for these measurements was to estimate the dose for inhabitants outdoors. The attempts to improve the knowledge of influence of climatic parameters on radon concentration started later.

The lowest layer of the atmosphere in which the majority of the weather system occurs and which contains about eighty percent of the total mass of the atmosphere is the troposphere. The lowest section of the troposphere has the properties that are directly influenced by the earth's surface and is known as the boundary layer or the mixing layer. The properties of the boundary layer have a major impact on the dispersion of radionuclides released within it, particularly over short to medium distances. All the experiments were performed in the ground layer, which is approximately 0 to 50 m above the ground. Figure 1 shows an image of the vertical range of the two main earthbound spheres of the troposphere.

The behavior of radon daughter products in the boundary layer of the troposphere is controlled by atmospheric processes, which also influence the releasing of radon from soil sphere. Short-lived radon decay products (^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po) – RnDP and other radionuclides (^{210}Pb) from the uranium family are the result of the radon gas decay. These radionuclides are mostly deposited on aerosol particles; they are so-called radioactive aerosols and are transported through the atmosphere.

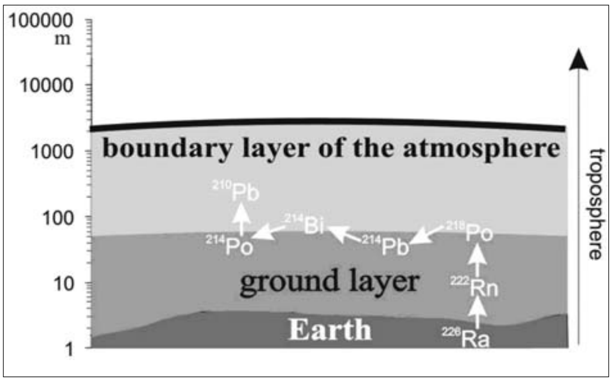

Fig. 1. Lower parts of troposphere.

We try to demonstrate the number of different entities – the average concentration of aerosol particles in the atmosphere is 6000 particles·cm^{-3}, the average concentration of radon is about 10 Bq·m^{-3} and EEC is 5 Bq·m^{-3} [4], so the ratio between the number of aerosol particles, radon, and RnDP is about 6000:5:0.001 in 1 cm^3.

There are two main factors that break the radioactive equilibrium between radon and RnDP. The deposition of atmospheric particles occurs as dry deposition due to gravity and inertia with impaction and gravitational settling. The second factor is wet deposition by rainfall events [3].

Materials and methods

EEC, radon concentration, and long-lived radionuclides concentration were measured in the ground layer.

EEC (20 minute mean) was estimated by the BUHS method that is based on sampling of airborne outdoor radioactivity to an open-face glass-fiber filter.

The continuous monitor of RnDP (TS-92), which is normally used indoors was modified for the outdoors (the sampling step is half-hour).

To estimate the concentration of long-lived radionuclides in the outdoor air, a more powerful sampling device has been used. The (open face) filter was afterwards measured by a 512 channel spectrometer Canberra.

For measurement of radon concentration outdoors, a high-volume scintillation chamber with a very low background was used. Also, the SSNTDs in diffusion chambers were used to measure the vertical profile in the atmosphere.

Results

The main part of experiments was performed in the Czech Republic, especially in Central Bohemia, but several measurements were also carried out abroad (e.g. Berlin, Timisoara, coast of Norway, Indianapolis). Near the sea the values of EEC were very low (it was expected).

EEC

Figure 2 shows the average EEC in Kamenná (where our Institute is located, about 50 km south of Praha).

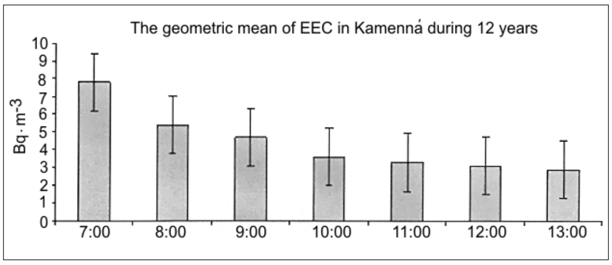

Fig. 2. The geometric mean of EEC during daytime.

The graph sums results of 12 years of regular measurements by using grab sampling methods. This database contains over 500 measurements.

A decreasing trend can be seen in the morning hours. A similar trend was observed in the case of other measurements at other places.

Another type of measurement consists of placing a continuous monitor at 1.8 m above the ground (in the breathing zone of a human). These measurements always continued for at least one month. From these results, it is also evident that the main factor to decrease of EEC in the atmosphere (in day run) is the mixing of air caused by temperature inhomogeneity.

This was realized by many simultaneously measurements in different places in the Czech Republic.

Results of these measurements show that at the same time the values of EEC are on a similar level. One example: a measurement was performed for two days in all district towns in the Czech Republic. The first day total mean was 3.2±1.5 Bq·m^{-3}, the second day 2.5±1.3 Bq·m^{-3} [2]. The measurements were realized mostly at 7 a.m. In both days, EEC in all places was atypically low.

The graphic visualization of results, which were mentioned above (measurement realized near our Institute), can be seen in Fig. 3. During the last 12 years, it can be seen a insignificant increase.

Long-lived radionuclides

UNSCEAR 2000 [4] presents a like reference value of ^{210}Pb in the atmosphere 500 μBq·m^{-3}. The range of concentrations of this radionuclide is 40–710 μBq·m^{-3} in Poland and 28–2250 μBq·m^{-3} in Germany. We have found that the concentration of ^{210}Pb in the Czech Republic is close to the lower part of the published range. From the limited number of our measurements, one can estimate the value to be about 35 μBq·m^{-3}.

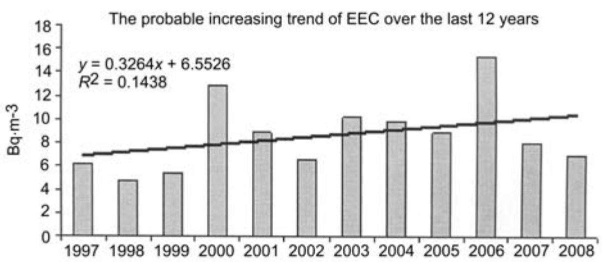

Fig. 3. The probable increasing trend of EEC over the last 12 years.

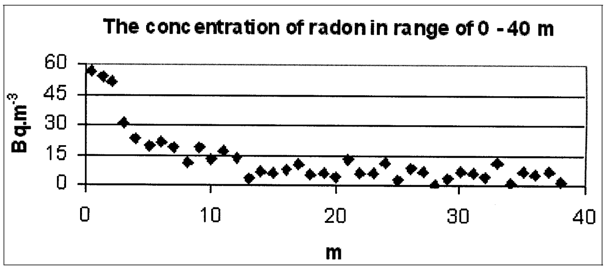

Fig. 4. The concentration of radon in the range of 0–40 m.

Radon vertical distribution

Radon gas concentration was measured by SSNTD in 0–40 m above the ground and the mean for one quarter of the year (in spring 2008) was 50 Bq·m^{-3} at a height of 1.8 m. The detectors were placed on a non-active chimney in the area of our Institute in the neighborhood of former uranium mine. Figure 4 shows that the stratification of concentration of radon at this height is striking.

The measurements described above show a substantial decrease in radon concentration when elevating – in 40 m above the ground, radon concentration was only 2 Bq·m^{-3}. The detailed measurements were realized also by using SSNTD at low heights. Detectors were placed on different levels of a 6 m high iron construction. The exposition time was three months. At these heights, the strong vertical stratification is not observed. See Fig. 5.

These types of experiments are not influenced by differences between night and day concentrations of radon. Both applications of SSNTDs were realized at a 100 m distance and in different year seasons.

There were also sets of grab samplings realized (not during nights) at levels 0.3–6 m. Morning EEC values in the same place and in different days did not depend on the high. See Fig. 6.

Discussion

The behavior of radon and EEC outdoors in the boundary layer of the atmosphere is affected mainly by two factors. Changes of barometric pressure cause a difference in emanation of radon from soil – this will be discussed in a future paper. The second factor influencing EEC is air turbulence, which homogenizes and decreases EEC in the boundary layer of the atmosphere.

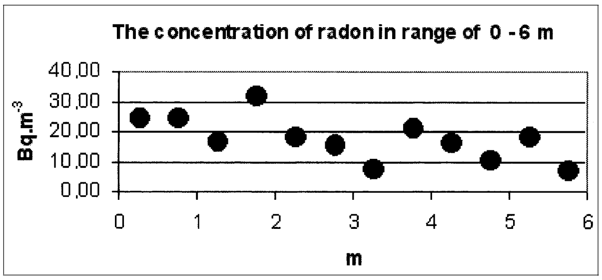

Fig. 5. The concentration of radon in the range of 0–6 m.

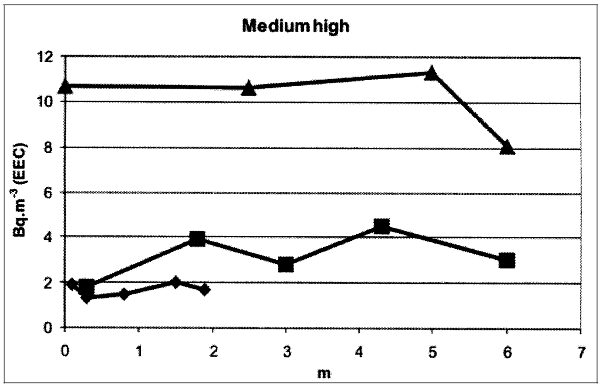

Fig. 6. EEC in the range of 0–6 m.

From some former studies [1, 2], it is possible to construct the hypothesis: levels of EEC in the entire Czech Republic are similar when measured at the same time. Part of our measurements was organized in little villages. We never saw a significant difference between values there and values collected in towns. These experiences are opposite to the following vision: there are essential differences in many factors of the atmosphere in towns and in the countryside (turbulent regime, concentration of aerosols, etc.). EEC for these reasons could be significantly different for forests and, e.g. in the downtown areas.

Figure 7 shows how EEC is affected by wind. This graph displays typical running of EEC, which was obtained by several monitoring of the atmosphere in breathing zone of man.

From this graph, it is also clear that EEC in the ground layer is mostly higher during the night. It is partly caused by a lower wind speed. In the morning, approximately between 7 and 8 a.m. EEC decreases as a result of turbulent flow due to sunshine and non-

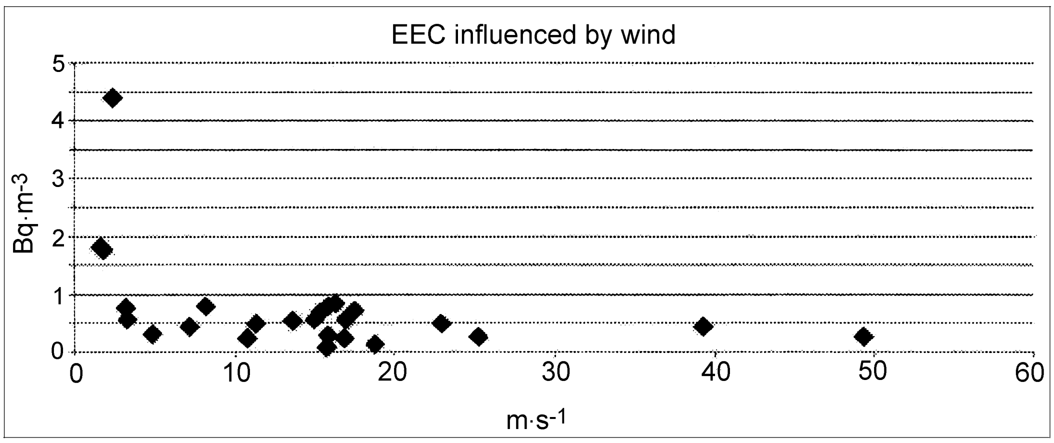

Fig. 7. The relation between EEC and wind speed.

-uniform warming of the earth's surface. This is one of two main factors influencing RnDP deposition on the earth's surface (buildings, trees, bushes, etc.) which cause the decreasing and homogenizing of EEC in the boundary level of the atmosphere.

Remarks

In 1988, EECs were measured in district towns of the former Czechoslovakia and the mean at 7 a.m. was 8.2 Bq·m^{-3} [1]. The method of estimation of EER in outdoor was similar to the methods used now. In the past, some conclusions were similar to those now presented – no important differences in values at 0.3 m and in the 1.8 m breathing zone were observed. The coefficient of correlation between morning EEC and temperature was relatively high and positive. A weak decrease in EEC with increasing pressure was also observed.

Our measurement results in the range from 200 Bq·m^{-3} to 1 MBq·m^{-3} are traceable to Physikalisch-Technische Bundesanstalt Braunschweig (Germany) with relative uncertainty less than 7%. However, the outdoor concentrations are much lower and, therefore, the uncertainties of the absolute values are rather difficult to estimate (there are less than 30%).

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