

DIURNAL AND SPATIAL VARIATIONS OF RADON CONCENTRATION AND ITS INFLUENCE ON IONIZATION OF AIR

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Abstract: The most abundant and efficient source of air ionization in the lower layer of the atmosphere is radon. As an alpha emitter, radon plays a crucial role in the earth's atmospheric electricity. Besides the physical, radon and ions have a significant biological role concerning human health: radon is a health hazard while the ions are beneficial ingredients of the air we breathe. In this study, we examined the dynamics of radon and air ions diurnal change in houses with different floor and windows insulations. Measurements were made using continual radon monitor Rad-7 and air ion counter CDI-06. Diurnal and spatial variations of both atmospheric constituents are mutually related and dependent mostly on radon exhalation potential, meteorological parameters, aerosol concentration and formation of the temperature inversion layer. Indoor concentrations are related to the potential for accumulation of radon that is coming from the ground beneath the foundation and also influenced by external radon concentration that is diffusing through the walls, doors, and windows. Level of diffusion is depending on insulation. The difference in the paths by which radon enters the home can be seen by analyzing changes during diurnal continuous measurements.

Keywords: radon, air ions, ionization, atmosphere, air, natural radioactivity.

1. INTRODUCTION

Radon (^{222}Rn) is radioactive noble gas generated in the decay chain of the primordial elements uranium and thorium which can be found in the soil worldwide. ^{222}Rn decays emitting α -particle with half-life 3.82 days and it is followed by a series of four further decays (2 α and 2 β) with much shorter half-lives. After its ancestor decay, ^{222}Rn penetrates through the ground and moves up by diffusion and convection toward the surface and into the air. ^{222}Rn exhalation rate, at the surface, is driven by weather conditions such as precipitation, air pressure, the temperature of air and also permeability, thermal gradient, and humidity of the soil [1]. Further movement of ^{222}Rn through the atmosphere is determined by thermal processes. Diurnal ^{222}Rn concentration change is driven by surface soil and air temperature ratio which is resulting in the highest concentrations during

temperature inversion during the night and lowest in the afternoon when upward gas diffusion is highest. When exhaling from the soil into the air ^{222}Rn can enter the house interior by pressure-driven flow of soil gas through openings through the floor and foundations. If there is no communication with the outside environment, then it accumulates and can reach concentrations up to a few of the tens of thousands of Bq m^{-3} . Indoor ^{222}Rn concentration is a consequence of ^{222}Rn exhalation from the soil and much less from certain building materials. The ratio of these two sources depends on underlying soil and building material. Indoor ^{222}Rn concentration is depending on floor insulation from underground gasses and also from diffusion coefficients of walls, doors, and windows. Measurements showed that houses with poor floor insulation and new doors and windows have very high potential for ^{222}Rn accumulation. On the other hand, during the night, near-ground air is rich with ^{222}Rn and as a

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consequence the inversion air layer is forming. If doors and windows insulation is poor than ^{222}Rn rich air can penetrate inside the house. Those two cases have different dynamics of diurnal ^{222}Rn change that is shown in this study.

^{222}Rn is 7.5 times heavier than air and when inhaled, short-lived ^{222}Rn progeny in the lungs results in the deposition of α -energy in the cells of the bronchial epithelium [2]. According to many epidemiological studies, long term inhalation of ^{222}Rn can be one of the causes of lung cancer [3]. Energy of ^{222}Rn and ^{224}Ra decays are 5.49 and 6.29 MeV, respectively. Both generate more than 10^5 ion pairs in the air per each α -particle. Those primary ions evolve within microseconds by process of hydration into complexes that are called small air ions, cluster ions or charged nano-aerosols (≥ 1 nm). They consist of one layer of water molecules (4–12) around central ion that is usually some inorganic molecule. Those are the smallest ions and the most important ones due to their mobility and relative long lifetime which is up to 5–60 s depending on air pollution [4]. The size categorization of air ions in small cluster ions 0.36–0.85 nm, big cluster ions 0.85–1.6 nm, intermediate 1.6–7.4 and large ions 7.4–79 nm are provided [4] and [5]. Small air ions are stable only when carrying only a single elementary charge that relates mobility with mass.

In the open atmosphere, air ions are generated by natural sources such as cosmic rays, the radioactive decay of ^{222}Rn and other radioactive minerals in the ground (most often ^{40}K). The average ionization rate in continental areas is approximately 10 ion pairs $\text{cm}^{-3}\text{s}^{-1}$ at height of 1m above the ground. The contribution of cosmic radiation to the air ion pair generation rate is approximately 20%, ^{222}Rn contributing 65% and residue is attributed to the decay of mentioned radioactive elements in the ground [6]. The near-ground ionization rate caused by background ionization in the open atmosphere on continental earth is about 10 ion pair's $\text{cm}^{-3}\text{s}^{-1}$ [7]. The atmospheric ion concentration (n^\pm) is determined by the following balance equation:

$$\frac{dn^\pm}{dt} = q - \alpha n^\pm n^\mp - n^\pm \beta Z \quad (1)$$

where q is the volumetric production rate, Z is the aerosol number concentration, α is an ion to ion recombination coefficient and β is an effective ion-aerosol attachment coefficient [8] and [9]. Measuring of air ions is very confident indicator of ^{222}Rn concentration and its short term changes [10], especially in indoor air when there are no other ionizing sources. Positive vs. negative ions ratio is known as „coefficient of unipolarity” with average value of $n^+/n^- = 1.12$ [11,12]. Imbalance between

positive and negative small air ion concentrations is the consequence of the atmospheric electric electrode effect in near ground layer and higher mobility of negative small air ions and thus higher probability for their removal.

2. MEASURING METHODS AND INSTRUMENTS

Cylindrical Gerdien condenser is the most common instrument for the air-ion concentration measurements. In this experiment Cylindrical Detector of Air-Ions (CDI-06, Figure 1) made in the Institute of Physics Belgrade, was used.



Figure 1. Cylindrical Detector of Air-Ions (CDI-06)

It is a fully automated portable instrument with the ability to alternatively measure concentrations of positive and negative air-ions, temperature (T), pressure (P) and relative humidity (RH). Instrument consists of sensor and power/control parts. The sensor part of the CDI-06 consists of 3 cylindrical and coaxially arranged stainless steel electrodes: measuring (central), polarizing and shielding electrode. Inter-electrode space is ventilated by a fan and air-ions of desired polarity and mobility are forced by an electric field to deflect to the central measuring electrode and deliver their charge which is then measured by the current amplifier. Polarizing voltage is set in a way that only small air-ions are collected on the central electrode. Larger ions fly through the electrodes since their kinetic energy is higher than the deflecting potential of polarizing electrode [12] and [13]. Concentration of air-ions (n_j) is:

$$n_j = \frac{I_j}{q_j Q} \quad (2)$$

where $q_j = \pm e = \pm 1.602 \times 10^{-19}$ C, Q is the air-flow I_j is current through the central electrode. Absolute

calibration of current (by electrometer) and airflow (by hot-wire anemometer) enables absolute measurements of the air-ion concentrations. Generated current is amplified, digitalized and zeroed [13]. Digitalization of CDI output signal enables programmed auto-zeroing and automatic long term measurements. Air ion concentrations are measured in real time and can be used as tracer for ^{222}Rn activity concentration. Since the results are instantaneous, it is possible to see a fine structure of changes in concentrations of ^{222}Rn their causes.

Instrument was set to measure hour ^{222}Rn concentrations.

The uncertainty of this instrument depends on the measuring period and the level of ^{222}Rn activity. Longest measurement and higher concentration of ^{222}Rn resulting in lower uncertainty. Shortest ^{222}Rn concentration measurement is 5 minutes in so-called Sniff mode. This mode is with lowest accuracy but can be used as ^{222}Rn concentration indicator. Measurements that take an hour or longer are much more accurate.



Figure 2. Radon continual monitor, Rad7- Durridge Co, USA

2.1. Rn detector

^{222}Rn activity concentration was measured using a commercial solid-state alpha detector Electronic Radon Detector - RAD7 (Durrigde Company, 2000).

2. MEASUREMENTS AND RESULTS

Measurements were carried out in 3 different measuring places: an open atmosphere, a house with poor floor and window insulation (no vapor membrane) and a house with good insulation of floor and windows. Aim was to demonstrate air ions and ^{222}Rn concentration variations depending on the quality of floor and windows insulation in dwellings.

First measurements (Fig. 3) were performed in the open atmosphere in the city of Belgrade, nearby other two measuring points. Diurnal variation of both, ^{222}Rn and air ions of both polarities, is obvious and ^{222}Rn concentration is relatively low with an average value of about 12 Bq m^{-3} . This value is near to world average for near ground ionization. Night maxima are caused by a temperature inversion in the near-ground atmospheric layer which is visible almost every night. Open atmosphere is subjected to the influence of various parameters, so changes in the concentrations of ions and ^{222}Rn are not always simultaneous and interdependent (Fig. 3) as they are in the indoor air (Figure 4,5).

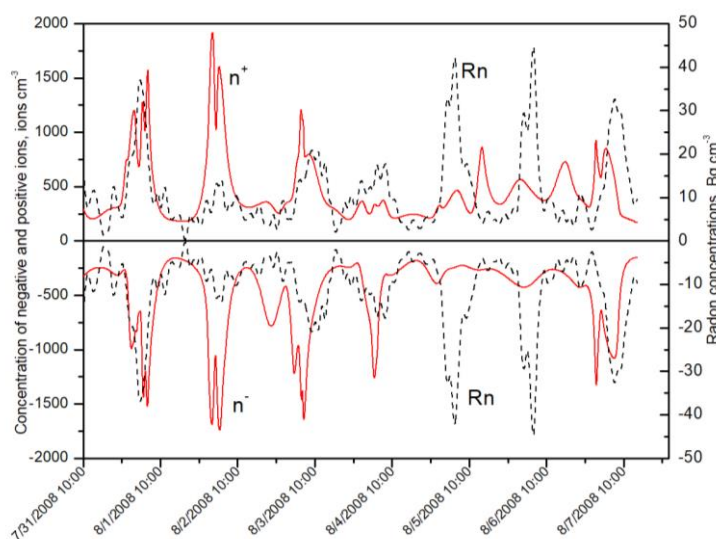


Figure 3. Typical daily outdoor ^{222}Rn and air ion variations

In the second part of the study, measurements were done in indoor air. First it was a ground floor house (Figure 4) which previously contained high concentrations of ^{222}Rn until remediation that was done 3 years ago. Remediation included concreting and floor insulation from water vapor which comprehends preventing of penetration of all other gases from under laying soil into the building. Also, all windows have been replaced with new ones (PVC) and walls were additionally insulated due to better thermal properties. All that provided better insulation from outdoor air. Since the house was occupied with inhabitants their activity can be seen on the graphs where short term changes during room ventilation are much clearer visible on air ion concentration lines on the graphs. Coefficient of unipolarity was 1.3, while during outdoor measurements it was 1.05. This implies on lower dust concentration outdoor since this coefficient is

influenced by higher coefficient of aerosol attachment of negative ions due to their higher mobility.

In Figure 5, measurements in the house with no floor insulation and poorly insulated windows are presented. Hence the assumption follows that the potential for ^{222}Rn exhalation in both houses is similar. Relatively high ^{222}Rn and air ion concentrations were measured with maxima at dawn and minima in the afternoon. Also, both measuring parameters are in high correlation. Except penetration of ^{222}Rn from the soil below the house, the influence of higher ^{222}Rn concentrations from the outside air is present. Weak insulation of the walls and windows is obvious due to low concentrations during the day. In well-insulated house, these differences are less pronounced. Coefficient of ion unipolarity was 1.2.

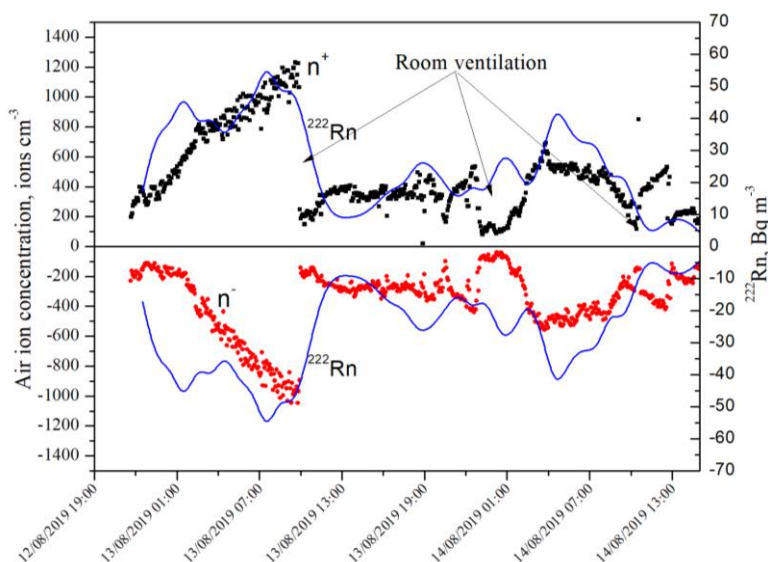


Figure 4. 2-day measurements in a ground floor house with insulated floor

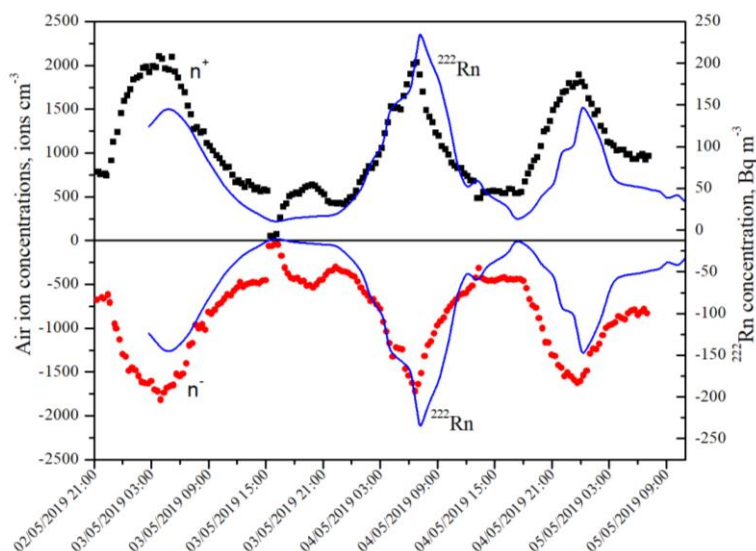


Figure 5. 3-day measurements in a ground floor house without floor insulation

Average concentrations of air ions in insulated house were 465 and 355 ions cm^{-3} , while ^{222}Rn concentration was 25 Bq m^{-3} . At the other hand, average ion concentrations in un-insulated house were 1004 and 836 ions cm^{-3} , while ^{222}Rn concentration was 79 Bq m^{-3} . ^{222}Rn concentration in both houses was relatively low and within acceptable limits, but ^{222}Rn less pronounced peaks and significantly higher concentration in un-insulated house is an obvious example of ^{222}Rn behavior in the indoor air.

4. CONCLUSION

Alpha particles from decay of ^{222}Rn and their descendants are the main source of the air ion generation in the lower atmosphere and thus the concentration of these two air constituents are highly correlated. This correlation is most visible in indoor air where the influence of interfering parameters is small. That way, using air ions as tracers, short term (order of a few seconds or more) changes of ^{222}Rn concentration can be detected. Diurnal and spatial variations of both values are mutually related with specific diurnal distribution in open atmosphere as well as in indoor air. House insulation from the gasses below the house and external air are crucial for lowering ^{222}Rn and air ion indoor concentrations. In insulated house ^{222}Rn concentration was lower and peaks were less pronounced. House with poor insulation from outdoor air was subjected to higher ^{222}Rn diffusion in and out, resulting in higher concentrations during the night and lower during the day. Old houses with typically bad floor insulation and new windows are prone to ^{222}Rn accumulation with small concentration amplitudes while same old houses with poorly insulating windows have high night ^{222}Rn concentration peaks.

5. REFERENCES

[1] C. Duenas, M. C. Fernandez, J. Carretero, E. Liger, M. Perez, *Release of ^{222}Rn from some soils*, Ann. Geophysicae, Vol. 15 (1997) 124–133.
[2] P. Hopke, B. Jensen, C. S. Li, N. Montassier, P. Wasiolek, A. J. Cavallo, K. Gatsby, R., Socolow, A. James, *Assessment of the Exposure to and Dose from Radon Decay Products in Normally Occupied Homes*, Environ. Sci. Technol., Vol. 29–5 (1995) 1359–1364.

[3] F. Bochicchio, *Radon epidemiology and nuclear track detectors: Methods, results and perspectives*, Radiat. Meas., Vol. 40 (2005) 177–190.
[4] U. Hõrrak, J. Salm, H. Tammet, *Statistical characterization of air ion mobility spectra at Tahkuse Observatory: Classification of air ions*, J. Geophys. Res., Vol. 105–D7 (2000) 9291–9302.
[5] U. Hõrrak, *Air Ion Mobility Spectrum at a Rural Area*, PhD Thesis, University of Tartu, Estonia, 2001.
[6] L. Laakso, A. Hirsikko, T. Gronholm, M. Kulmala, A. Luts, T. E. Parts, *Waterfalls as sources of small charged aerosol particles*, Atmos. Chem. Phys. Discuss., Vol. 6 (2006) 9297–9314.
[7] J. A. Chalmers, *Atmospheric Electricity*, Pergamon Press, Oxford, London, 1967.
[8] L. Laakso, A. Hirsikko, T. Gronholm, M. Kulmala, A. Luts, T. E. Parts, *Waterfalls as sources of small charged aerosol particles*, Atmos. Chem. Phys., Vol 7 (2007) 2271–2275.
[9] U. Hõrrak, P. P. Aalto, J. Salm, K. Komsaare, H. J. Tammet, M. Mäkelä, L. Laakso, M. Kulmala, *Variation and balance of positive air ion concentrations in a boreal forest*, Atmos. Chem. Phys., Vol. 8 (2008) 655–675.
[10] P. Kolarž, Z. Čurguz, *Air ions as indicators of short-term indoor radon variations*, Applied Radiation and Isotopes, Vol. 99 (2015) 179–185.
[11] W. A. Hoppel, R. V. Anderson, J. C. Willett, *Atmospheric electricity in the planetary boundary layer*, in *The Earth's Electrical Environment*, Electrical Environment, National Academy Press, Washington, USA, 1986, 149–165.
[12] U. Hõrrak, *Air ion mobility spectrum at a rural area*, Dissertationes Geophysicales Universitat Tartuensis, Tartu, Estonia, 2001.
[13] P. Kolarž, B. P. Marinković, D. M. Filipović, *Zeroing and testing units developed for Gerdien atmospheric ion detectors*, Review of Scientific Instruments, Vol. 76 (2005) 046107–046109.
[12] P. M. Kolarž, D. M. Filipović, B. P. Marinković, *Daily variations of indoor air ion and radon concentrations*, Applied Radiation and Isotopes, Vol. 67 (2009) 2062–2067.
[13] P. Kolarž, B. Miljković, Z. Čurguz, *Air-ion counter and mobility spectrometer*, Nuclear Instruments and Methods in Physics Research B, Vol. 279 (2012) 219–222.



ДНЕВНЕ И ПРОСТОРНЕ ПРОМЕНЕ КОНЦЕНТРАЦИЈЕ РАДОНА И ЊЕГОВ УТИЦАЈ НА ЈОНИЗАЦИЈУ ВАЗДУХА

Сажетак: Најприсутнији и најефикаснији извор јонизације ваздуха у приземном слоју атмосфере је радон. Као алфа емитер, радон игра пресудну улогу у атмосферском наелектрисању. Поред физичке, радон и јони имају значајну биолошку улогу по здравље људи: високе концентрације радона представљају опасност по здравље, док су јони као нуспродукт распада радона корисни састојак ваздуха који удишемо. Мерења су вршена коришћењем континуалног радон монитора “Rad7” и бројача јона у ваздуху CDI-06. Дневне и просторне варијације оба атмосферска састојка међусобно су повезане и углавном зависе од потенцијала распада радона, метеоролошких параметара, концентрације аеросола и стварања температурног инверзног слоја у приземном слоју атмосфере. Концентрације радона у затвореном простору повезане су са акумулацијом радона који долази из тла испод зграде као и са спољашњом концентрацијом радона који дифундује кроз зидове, врата и прозоре. Разлика путева на који радон улази у кућу може се видети анализом промена током дневних континуалних мерења.

Кључне речи: радон, атмосферски јони, јонизација, атмосфера, ваздух, природна радиоактивност.



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