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Daily variations of indoor air-ion and radon concentrations

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ABSTRACT

Air-ions and radon are two atmospheric trace constituents which have two opposite effects on human health: the ions are beneficial, and radon gas is potentially lethal as it increases the risk of lung cancer. In the lower troposphere, radon is the most important generator of the air-ions. Ionization by cosmic rays and radioactive minerals is almost constant in daily cycles, and variation of air-ion concentrations is attributed to changes of the radon activity.

Air-ion and radon concentrations in outdoor and indoor space and their vertical gradients in residential buildings were measured. Gerdien type air-ion detector "CDI-06" made in our laboratory and radon monitor "RAD7" were utilized for these measurements. Correlation coefficient between positive air-ion and Rn indoor concentrations was approximately 0.7. Outdoor and indoor peak values were simultaneous while vertical gradient of concentrations in indoor measurements was evident. The indoor experiments showed that positive air-ion concentration could be an alternative method of radon activity concentration evaluation.

1. Introduction

Radon (Rn) and air-ions are two highly correlated biologically active constituents of outdoor and indoor air. Air-ion concentrations ($n^{\pm}$) in the lower troposphere are determined mostly by airborne radionuclides and external sources, i.e., cosmic radiation and radioactivity that originates from the radioactive minerals from the earth's crust (Laakso et al., 2004; Chandrashekara et al., 2006). At sea level up to a few hundred meters, cosmic rays contribute approximately 20% of total surface air-ion production rate that is typically $10\,\mathrm{cm}^{-3}\,\mathrm{s}^{-1}$. According to UNSCEAR (2000, p. 87) a representative shielding factor of buildings on cosmic radiation charged particles and photons is 0.8 after the radiation has passed through a substantial ceiling. The second external source of air-ions is γ radiation from ^{238}U, ^{232}Th and ^{235}U decay series and primordial radionuclide as ^{40}K. These are the most stable sources of air-ion production and considered as almost constant during the day, because their variation is very small compared to Rn and its progenies decay (Hirsikko et al., 2007; Israelson and Tammet, 2001; Carslaw et al., 2002). Major inducement of variation of the air-ion pair production is due to changes in Rn exhalation and atmospheric mixing processes (UNSCEAR, 2000, p. 99).

1.1. Radon

^{222}Rn is an inert and radioactive soil gas descending from uranium ^{238}U decay series with half-life 3.82 days. ^{222}Rn α-decay is followed by a series of four further decays with half-lives less than 30 min each: ^{218}Po (α particle), ^{214}Pb (α particle), ^{214}Bi (β particle) and ^{214}Po (β particle). The radioactive decay of inhaled short-lived radon progeny in the respiratory tract results in the deposition of α-energy in the cells of the bronchial epithelium (Hopke et al., 1995). Decay products can cause direct effect on the DNA structure and indirect effect due to the production of active chemical radicals in the vicinity of DNA (UNSCEAR, 1993). The risk estimates obtained in the study of Field et al. (2000) suggest that cumulative Rn exposure in the residential environment is significantly associated with lung cancer risk. It is estimated that Rn contributes around 50% of background radiation dose received by general population (Groves-Kirkby et al., 2008).

Radon is a gas 7.5 times heavier than air; when generated in Earth's crust, it penetrates the pores in the ground and moves upward by diffusion and convection toward the surface and into the air. This process is called exhalation and its rate depends on air pressure and also permeability, thermal gradient and humidity of the soil (Dueñas et al., 1997). In the atmosphere, Rn appears mostly in the vicinity of its source, i.e., ground, and its transport is determined by thermal processes. When exhaling in the indoor space, Rn is prone to accumulation. Rn entrance and accumulation in residences and offices is related to many local and time dependent factors such as uranium content of the underlying soil,

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construction material, permeability and number of cracks in the basement shell, ventilation conditions, radioactivity in the air outdoors (Abbady et al., 2004; Janssen, 2003) and meteorological parameters (Kitto, 2005). Indoor sources of Rn are soil or rocks under or surrounding the buildings, construction materials, water supplies, natural gas and outdoor air (UNSCEAR, 1988). Annual indoor action level of Rn concentration above which remediation should be considered varies from country to country. According to ICRP (International Commission on Radiological Protection) recommendation, it is between 200 and 600 Bq/m^3 (WHO, 2006), where 1 Bq = 1 decay/s and Rn activity concentration is activity/volume. Evaluation of the Rn concentration in indoor spaces is very important where the underlying geology, soils or building materials are intensive Rn sources. High Rn concentrations are measured in the old houses without concrete floor and hydro-insulation especially if ventilation is not intensive. Also, construction materials can be sources of Rn and thoron radionuclides in the indoor air (UNSCEAR, 1993; Porstendőrfer, 1994).

Thoron (^{220}Rn) is noble gas from thorium (^{232}Th) decay chain with relatively short half-life of 55.6 s. Thoron exhales from the ground and walls in the same way as radon but in general has a lower concentration. In indoor space, thoron exhails from building materials that contain radium (^{224}Ra). Although thoron concentration decreases exponentially from the source (Urošević et al., 2008), its contribution to air-ion pair production due to α-decay remains.

1.2. Air-ions

Energy of α particle from ^{222}Rn decay is 5.49 MeV; that is enough to create 10^5 air-ion pairs on its trajectory through the air (Burcham, 1973). There are three basic groups of the air-ions classified by their mobilities or linear dimensions: small (0.36–1.6 nm), intermediate (1.6–7.4 nm) and large ions (7.4–79 nm) (Horrak, 2001; Jamieson et al., 2007). Only small air-ions are considered in this work because of their significant electrical and biological activity in the atmosphere. They are composed of the central primary ion and surrounding 4–10 molecules of water, depending on humidity, temperature, actual concentration of trace gasses (Luts and Parts, 2002) and polarity of the ions. The structure of dominant air-ions in indoor air is determined by water, ammonia, nitrogen oxides and NO$_x$ radicals (Parts, 1996). Central negative air-ions (n^-) attract lower number of water molecules than positive (n^+), so that their mass is lower and mobility is higher. Consequently, higher mobility enables larger probability for ion neutralization, so concentration of negative air-ions is usually lower than the positive, where n^+/n^- is called the coefficient of unipolarity. In the air-ion research only singly charged ions are found since the multiple charged ions rapidly undergo charge transfer reaction with surrounding neutral molecules to result in singly charged ions. The major mechanisms of air-ions neutralization near the ground level are self-recombination (characterized by a coefficient α) and attachment onto aerosol particles (coefficient β). The simplified equation of the air-ion equilibrium is determined as follows:

$$\frac{dn^\pm}{dt} = q - \alpha n^+ n^- - \beta^\pm N n^\pm - \delta^\pm n^\pm, \qquad (1)$$

where q is air-ion production rate, N large aerosol number concentration, and δ electrostatic deposition rate coefficient of the air-ions occurring only in indoor air (Graeffe and Keskinen, 1989). Coefficient δ depends on existing insulating surfaces and relative humidity. If $\beta^\pm N n^\pm \gg \alpha n^+ n^- + \delta^\pm n^\pm$ the air-ion concentration in equilibrium is given by

$$n^\pm = \frac{q}{\beta^\pm N}. \qquad (2)$$

In troposphere air, about 97% of the air-ions attach to aerosol particles (Luts and Parts, 2002). According to Horrak (2001), the mean natural mobilities of small positive and negative air-ions is 1.36 and 1.53 cm^2/V s, respectively. In indoor air average mobility of positive small air-ions is 1.19 and 1.66 cm^2/V s for small negative air-ions (Parts, 1999). Typical positive and negative air-ion concentrations outdoor, near the ground, is ~500–1000 ions/cm^3, having in mind that average number of atoms and molecules in the air is $\sim 2.7 \times 10^{19}$ cm^{-3}. Their typical lifetime is in the range from several seconds to several minutes (Guedalia et al., 1972) depending on the source of the ions (their composition), air pollution, relative humidity of the air and altitude. During their lifetime in indoor air, air-ions act as aerosol removals (Grinshpun et al., 2005; Uk Lee et al., 2004) and also they have an important biological influence on various micro-organisms (Shargawi et al., 1999) and positive effects on human organism (Krueger and Reed, 1976; Nakane et al., 2002; Takahashi et al., 2008). Measuring of the air-ion concentrations in this work is performed by using their electrical properties in our Gerdien aspirated condenser.

2. Instrumentation

2.1. Air-ions detector

The aspirated Gerdien condenser is a widely utilized instrument for the air-ion concentrations and mobility measurements. Cylindrical detector of air-ions (CDI-06) was made in our laboratory in the Institute of Physics, Belgrade (Kolarž et al., 2005). It is a fully automated portable instrument with ability to alternatively measure concentrations of positive and negative air-ions, temperature (T), pressure (P) and relative humidity (RH).

Sensor part of the CDI-06 consists of three cylindrical and coaxially arranged electrodes, i.e., measuring (central), polarizing and shielding electrode (Fig. 1). The space between the electrodes is ventilated by a fan. In the inter-electrode space air-ions of desired polarity and mobility are forced by electric field to deliver their charge to the measuring electrode. The charge collected on this electrode is measured by an electrometer, recalculated and

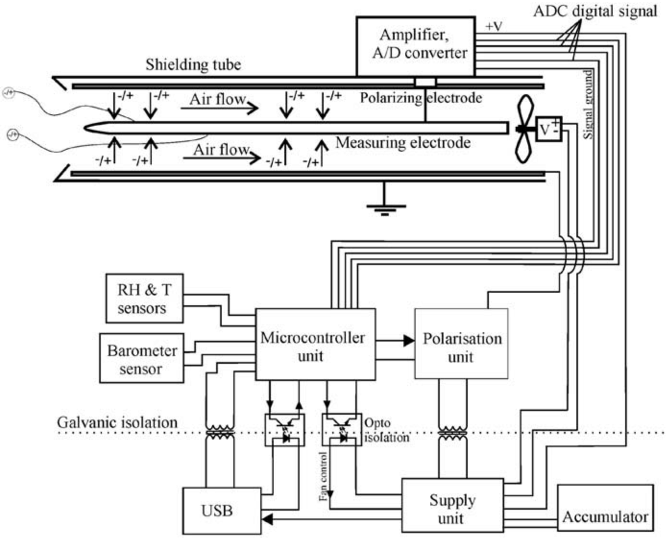

Fig. 1. Block diagram of automated cylindrical detector of air-ions (CDI-06).

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represented as concentration of air-ions in units of cm^{-3} with desired time resolution and averaging. The mobility of the ions is determined by the applied polarizing voltage and air-flow velocity. Polarizing voltage is set in a way that only small air-ions are collected on the central electrode. Larger ions, according to their kinetic energy, could not be deflected and they pass through electrodes. Air-flow (Q) is $8.7\times10^{-3}\,\mathrm{m}^3/\mathrm{s}$, which is relatively high for this kind of instrument. This provides lower inherent statistical and instrument (signal amplification) uncertainty. Air-ion concentration (n_J) is

$$n_J = \frac{I_J}{q_J Q}, \tag{3}$$

where $q_J = \pm e = \pm 1.602\times10^{-19}\,\mathrm{C}$, I_J is current through the central electrode. Absolute calibration of current and air flow enables absolute measurements of the air-ion concentrations.

The most important feature of CDI-06 is that the ion generated current signal is amplified, digitalized and zeroed (Kolarž et al., 2005). Zeroing of the system is performed periodically in order to compensate the noise sources induced by the changes of T, RH, external electromagnetic waves and high voltage peaks induced by polarity change or fan stopping/starting. Zeroing indicates that the signal measured during the zero conductivity (MacGorman and Rust, 1998) is subtracted from further measurements until next zeroing. Zero conductivity regimes imply that the polarizing voltage and the fan are off, and under these circumstances, measuring electrode collects only ions that are led by Brownian motion, and output signal should be about a few ions/cm^3. Digitalization of CDI output signal enables programmed auto-zeroing and automatic long term measurements without the presence of human operator. Five identical copies of CDI-06 were made so that they all could be mutually compared and tested, so any kind of systematic error due to instrumentation is avoided.

The sources of measuring uncertainties of CDI-06 in indoor conditions are components of the systematic uncertainty arising from air-flow uncertainty 3%, input calibration error 1.4%, temperature drift of feedback resistor (50 ppm/°C, i.e., 0.2%), gain error of operational amplifier (30 ppm/°C, 0.12%) and voltage reference error (50 ppm/°C, 0.2%). Leakage currents of the amplifier (within 2 fA) and within electrodes are compensated by the zeroing system. Overall uncertainty of the CDI-06 is in the range of 5% if the ambient humidity is below 80%. Measuring uncertainty in outdoor conditions is mostly subjected to intensity and direction of wind and high relative humidity, so it can be evaluated up to 30%.

2.2. *Rn detector*

Rn activity was measured with state of the art commercial solid state alpha detector Electronic Radon Detector-RAD7 (Durridge Co., 2000), calibrated on April 14, 2008. In contrast to classical widely used charcoal Rn adsorption devices which measure three or more day averages, this instrument can measure hour concentrations. Magnitude of uncertainty of this instrument depends on duration of measuring period and Rn activity.

3. Experiments

A typical five flat building in Belgrade was chosen for measuring vertical gradients of the air-ion and Rn concentrations. The building was at the final stage of construction, i.e., finished but unsettled. Before and during all measurements, apartments were undisturbed, windows and doors closed, so there was no forced air motion besides diffusion through the walls and floors. Forecasted meteorological conditions for whole measuring period were moderate and calm.

Since the building was new, all modern structural standards of hydro and thermo insulation were respected, so that Rn levels are within permissive limits. All apartments were empty and there were no electrostatic surfaces that could cause significant air-ion electrostatic deposition. Calibration of the air flow and current to signal ratio of the CDI-06 detectors was performed before the measurements. In the air-ion measurements there is no possibility for direct testing of the instruments; therefore inter-comparison of the two air-ion detectors used in experiments was performed. Air-ion concentrations were measured for two days with two identical instruments close to each other on the second floor of the building. During the first day, window was slightly opened and the next day it was closed (Fig. 2). Comparison of the two CDI-06 (Fig. 2) showed that average measuring result disagreement was within ±4% for both polarities which was more than satisfying for this kind of measurement.

To get the clear picture of the Rn exhalation, and consequently the air-ion concentrations changes over the typical soil of our surroundings, we performed an outdoor experiment. The measurement was made in Belgrade in outdoor and natural surrounding during fine weather conditions (Dolezalek, 1982), where Rn exhalation is not attenuated with concrete and asphalt. Sampling, averaging and change of electrode polarity on CDI-06 were set at 5 min for each polarity measurements while readings of RAD7 were set at 1 h.

Gradient measurements indoors were performed in the same positioned apartments of every floor, one above the other, starting from the ground-floor, within 24 h intervals. During measurements, all floors along the vertical axis were closed, same as measuring place itself, so that diurnal changes in Rn and air-ion concentrations could originate only from Rn exhalation from the ground and its diffusion and advection through the concrete or different gaps upwards. Otherwise, Rn concentration could change due to open windows or sudden changes of temperature could cause pressure differences between floors and forced diffusion of Rn. Instruments were located 1 m above the floor, close to each other in the middle of the room.

4. Results

Outdoor behaviors of the Rn and air-ions are presented in Fig. 3. Most of the Rn and air-ion peaks were about dawn, and minimal concentrations during the afternoon. Averaged multiplication factor (3 h averages) between daily maximal and minimal values was about 9 and 6 for the Rn and air-ions, respectively. Averaged Rn (A(Rn)) and air-ion ($n^{\pm}$) quotients were: $n^-/A(\mathrm{Rn}) = 37\times10^6$ ions/Bq and $n^+/A(\mathrm{Rn}) = 40\times10^6$ ions/Bq. Coefficient of the unipolarity was $n^+/n^- = 1.06$.

To acquire the best evaluation of results Pearson correlation coefficient was used,

$$r = \frac{\Delta xy}{\Delta x \Delta y}, \tag{4}$$

where $\Delta xy = \langle (x-\langle x\rangle)(y-\langle y\rangle)\rangle$ is a covariance, and $\Delta\xi = (\langle(\xi-\langle\xi\rangle)^2\rangle)^{1/2}$, $(\xi = x, y)$ are the standard deviations of the parameters x and y, respectively. The Pearson correlation coefficient (Eq. (4)) between air-ions and Rn concentration was 0.30 for negative and 0.24 for positive air-ions, which was above expectations concerning Rn dispersion in the atmosphere. Correlation factor between negative and positive air-ion concentrations was 0.94.

The results of indoor experiment (Fig. 4) showed that air-ion and Rn concentration on fifth floor reached about 60% of the

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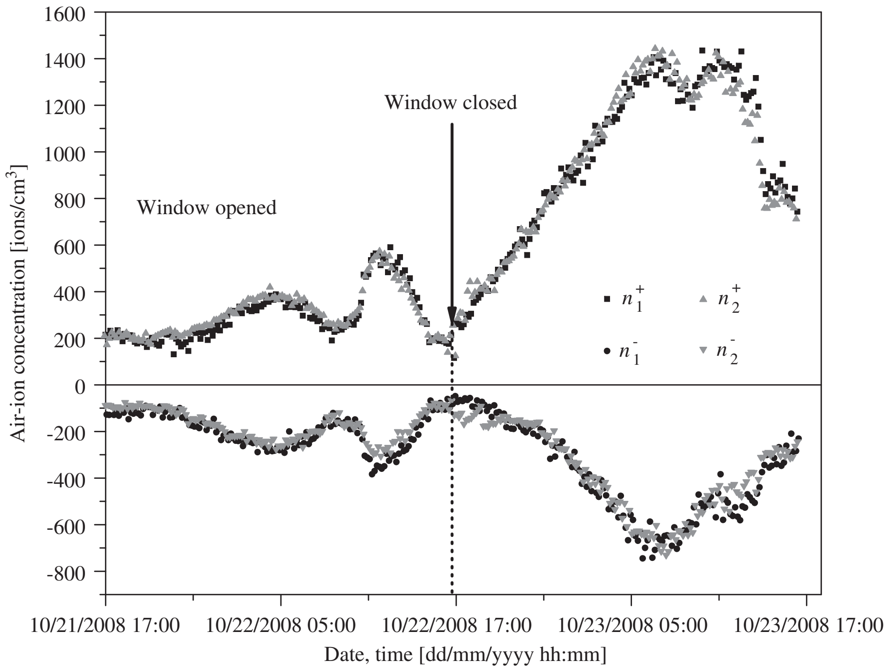

Fig. 2. Measuring results of the two air-ion detectors CDI-06 used for comparison and testing performed in Belgrade. Negative sign on *Y*-axis signifies negative air-ion concentration.

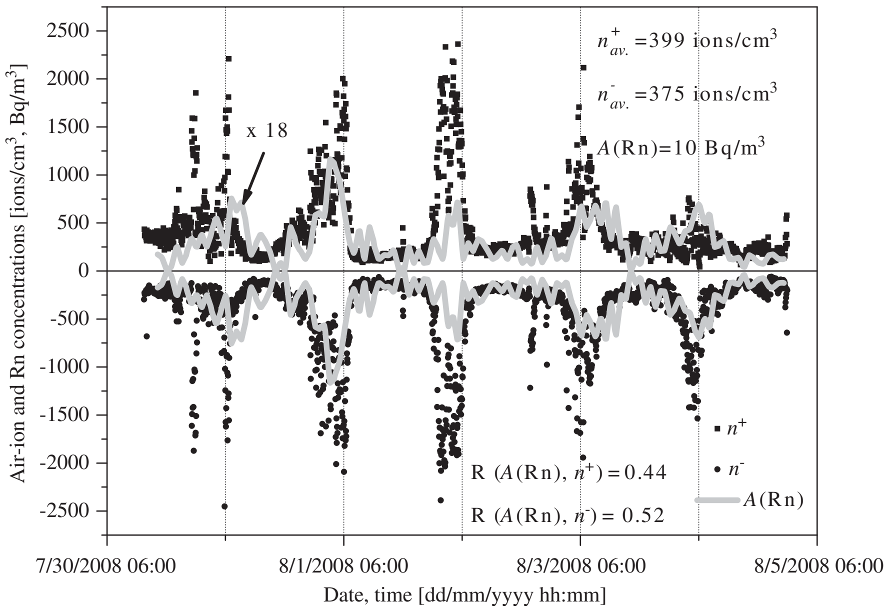

Fig. 3. Air-ion and Rn concentrations (standardized due to clarity) in six-day period measured outside in front of the Institute of Physics, Belgrade. Vertical dot grid lines indicating 6 h in the morning. $R(A(\mathrm{Rn}), n^{\pm})$ is the correlation coefficient. Measurements were performed outside the Institute, in early October 2008.

ground floor value. Despite expected decrease in Rn exhalation effects with height, significant diurnal differences in Rn and air-ion concentrations could be observed. Maxima of measured values were about dawn when Rn exhalation from underlying soil and walls was most intensive and minima were in the afternoon. Maximal values for Rn and air-ion concentrations were 2–3 times higher than minimal. The Pearson correlation coefficients between positive air-ions and Rn concentrations were mostly above 0.7 which proves a high dependence between these two physical parameters. Calculating coefficient of determination (r^2), 50% of observed air-ion concentration variability can be explained by Rn concentration. Averaged quotients of air-ion and Rn concentrations ($n^{\pm}/A(\mathrm{Rn})$) was $(5.5\pm2.0)\times10^6$ ions/Bq for negative and $(12.5\pm4.6)\times10^6$ ions/Bq for positive air-ions. The estimated uncertainties for these transfer quotients are 37% that include different measuring places and conditions. It is composed of uncertainty of Rn measurements based on counting statistics of the Rad7 (28.6%), systematic uncertainty of CDI-06 measurements (5%), inherent statistical uncertainty of the air-ion counting ($<1\%$) and systematic uncertainty of air-ion measurements due to different measuring conditions (20%). All uncertainties are summed using "root-sum-of-squares" method, and the last one is evaluated according to previous measuring experience (Taylor and Kuyatt, 1994). The uncertainty contribution based on counting

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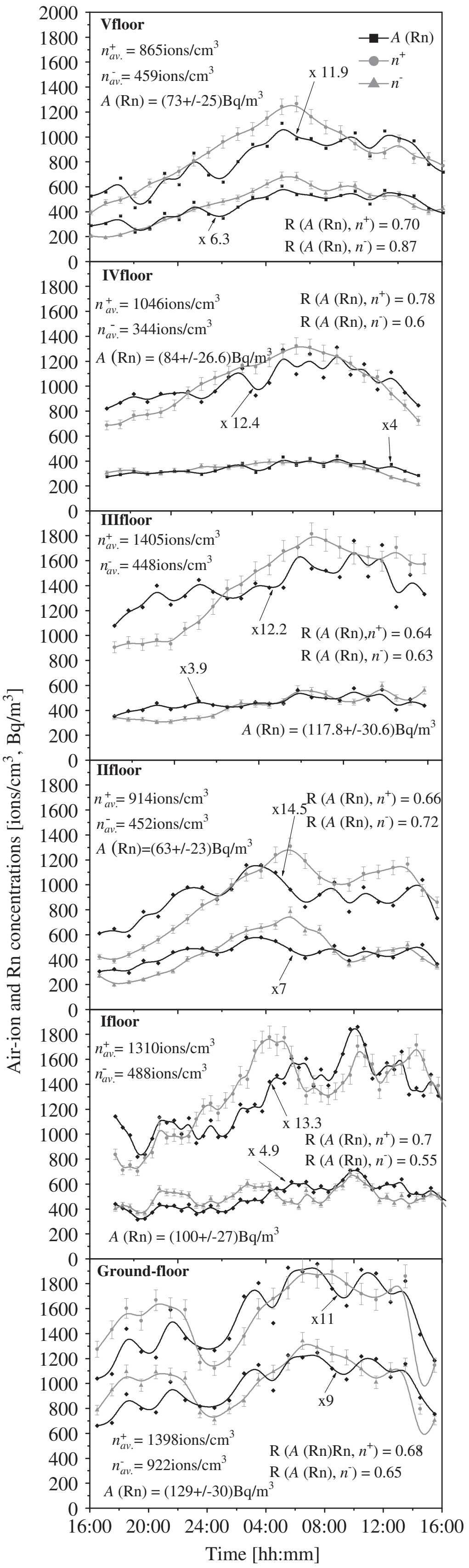

Fig. 4. Daily Rn (A(Rn)) and air-ion ($n^{\pm}$) concentrations from ground to fifth floor measured during October and November 2008 in Belgrade. $R(A(\mathrm{Rn}), n^{\pm})$ represent Pearsons correlation coefficient. Rn concentration error bars are omitted on graph because of their magnitude, instead mean measuring error is associated to mean Rn concentration A(Rn).

statistics of the Rad7 decreases with the increase of Rn concentration. If the radon activity concentration is as high as 3700 Bq/m^3, this uncertainty is 3.7% (instead of 28.6% used for 95 Bq/m^3).

Average coefficient of unipolarity (n^+/n^-) indoor was 2.7, which was 2.5 times higher than outdoor. During the measurements, weather conditions were stable, so T, RH and P measured inside the building showed almost constant values with minor daily changes.

5. Discussion

The measurements of the air-ion and Rn concentrations showed that outdoor (Fig. 3) and indoor (Fig. 4) changes had similar trends with maximal values about dawn and lower during the afternoon. These trends are usually caused by temperature inversion conditions of the lower atmosphere. During the night, temperature increases with altitude up to several meters above the ground forming near-ground inversion layer and then declines with further increase in height. This process decreases vertical mixing ratio and tends to trap the Rn closer to the ground (Blaauboer and Smetsers, 1996; Nagaraja et al., 2003). During the day, the Sun warms the atmosphere, vertical mixing ratio rises and inversion layer disappears. This process occurs in the urban area in a similar manner as in the rural area. According to the atmospheric mixing processes outdoor Rn concentrations can vary diurnally by a factor of as much as 10 (UNSCEAR, 2000). Similar results for outdoor measurements of Rn and air-ion concentrations were obtained by Blaauboer and Smetsers (1996), Prasad et al. (2005), Chandrashekara et al. (2006) and Komsaare et al. (2004).

Measured averaged concentrations of the Rn outdoor were low compared to the indoor concentrations due to diffusion toward higher layers of the atmosphere. This feature disables air-ions to Rn transfer in outdoor conditions. During outdoor measurements, which were performed in suburban area, the coefficient of unipolarity was ~1 meaning that the air was almost aerosol free. Smoothed peaks of the indoor concentrations, compared to the outdoor, were consequence of slower Rn diffusion through construction materials and also practically constant T, RH and P parameters. According to Porstendőrfer (1994) transport of Rn isotopes through construction materials is influenced by convective flow generated by a pressure gradient between interior and exterior of the building.

Significant differences in indoor coefficients of unipolarity were caused by higher mobility of the negative air-ions and consequently higher rates of neutralization (Eq. (1)). Enhanced amount of aerosol particles, i.e., dust in the apartments, resulted in higher attachment coefficient (β) onto aerosol particles (Eq. (2)). Second possible reason was electrostatic depositions of the air-ions on insulating surfaces. According to Graeffe and Keskinen (1989) this effect can be neglected when no human activities are present. It can be concluded that if indoor air-ion concentration measurements are used for Rn concentration estimation, much better results would be attained with positive air-ion concentration due to much lower coefficient of neutralization. Similar results concerning indoor air measurements were obtained by Komsaare et al. (2004) and Graeffe and Keskinen (1989) concluding that βN and δ should be sufficiently constant over different houses and Rn concentrations above 500 Bq/m^3.

Diurnal variations of Rn concentration in residential buildings and non-linear exposure of occupants in their homes (Briggs et al., 2002) refer that short term continuous measurements are needed for actual exposure determination. Short term measurements of relatively low Rn activities are subjected to high measuring

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uncertainty. “Rad7” detector, one of the best short term commercial Rn monitors, has measuring uncertainty of 41% at 37 Bq/m^3 at 1 h readings (Durridge Co., 2000), while measuring uncertainty in sniff mode (5 min) could be much higher. Nevertheless, rate of Rn exhalation has significant peaks in short time periods and if signal integration is too long they remain invisible. The results in Fig. 3 show sharp peaks of very high air-ion concentrations and analogous but much lower Rn peaks. On the other hand, besides uncertainty of air-ion measuring instruments, presence of aerosols, electrostatic surfaces, changes of cosmic radiation and local factors as terrestrial radiation and thoron concentration could influence uncertainty in air-ion to Rn transfer. Our opinion is that specified uncertainties in this transfer do not exceed measuring uncertainties in short time Rn determination even in the lower Rn concentration conditions.

6. Conclusion

Air-ion concentration measurements are the source of crucial information concerning electrical and nuclear processes in the atmosphere. According to the high correlation factor ($r > 0.7$) between positive air-ions and Rn in indoor measurements (Fig. 4), measuring of the positive air-ions could be a confident method in the evaluation of Rn concentrations. Also, the measuring concept of the air-ion detectors provides immediate results and thus continuous tracking of Rn variations is possible.

A measurement of air-ions and Rn concentrations shows that their outdoor and indoor concentrations are subjected to high diurnal variations with maximums in the early morning hours. Indoor diurnal variations can be observed along the vertical axis on every floor, including moderate vertical gradient of concentrations.

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