

NANO AEROSOLS IN THE POSTOJNA CAVE

NANO AEROSOLI V POSTOJNSKI JAMI

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Abstract UDC 911:551.44:539.16(497.4 Postojna)
Ivan Iskra, Norbert Kávási & Janja Vaupotič: Nano aerosols in the Postojnska jama

At the lowest point in the Postojnska Jama (jama = cave), concentration and size distribution of non-radioactive aerosols in the size range of 10–1,000 nm were measured and their concentrations are compared with those of radioactive aerosols carrying radon short-lived decay products (²¹⁸Po, ²¹⁴Pb and ²¹⁴Bi) obtained previously. Concentration of non-radioactive aerosols during morning hours was in the range 600–2,750 cm⁻³, with about 90% of particles smaller than 50 nm. On the other hand, concentration of radioactive aerosols smaller than 50 nm was several radionuclide atoms per 1 cm³ and the bigger ones, less than 1 atom per 1 cm³.

Keywords: nano aerosols, radon decay products, unattached, attached, Postojnska Jama.

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Na najnižji točki turistične poti v Postojnski jami smo merili koncentracijo in velikostno porazdelitev ne-radioaktivnih aerosolov v območju 10–1.000 nm. Njihove koncentracije smo primerjali s koncentracijami radioaktivnih aerosolov radonovih kratkoživih razpadnih produktov (²¹⁸Po, ²¹⁴Pb in ²¹⁴Bi). Koncentracija ne-radioaktivnih aerosolov je bila v jutranjih urah v območju 600–2.750 cm⁻³, od tega je bilo 90% delcev manjših od 50 nm. Koncentracija radioaktivnih aerosolov, manjših od 50 nm, je bila le nekaj atomov radionuklida na 1 cm³, večjih pa manj kot 1 atom na 1 cm³.

Ključne besede: nano aerosoli, radonovi razpadni produkti, nevezani, vezani, Postojnska jama.

INTRODUCTION

In last decade, the word 'nano' has been frequently used in our everyday life, e. g., in relation to medicines, pharmaceuticals, cosmetics, and various sprays and paints. It is also included in our discussions on a number of scientific, technological and environmental topics. The initial enthusiasm on all the advantages of using nano particles was soon followed by a fear of harmful effects they may cause to our health. This fear has appeared to be justified in a number of cases (Brouwer *et al.* 2004) and great sci-

entific efforts has been paid in order to better understand these negative effects. To be specific: these are non-radioactive nano particles.

On the other hand, radioactive nano particles and their effects have been known for decades. These are aerosols carrying atoms of the following radionuclides: ²¹⁸Po (α -decay, half-life $t_{1/2} = 3.05$ min), ²¹⁴Pb (β/γ -decay, $t_{1/2} = 26.8$ min), ²¹⁴Bi (β/γ -decay, $t_{1/2} = 19.7$ min) and ²¹⁴Po (α -decay, $t_{1/2} = 164$ ns). They are ubiquitously present in

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environmental air as radon decay products (RnDP), created by radioactive transformation of radioactive noble gas radon (^{222}Rn , α -decay, $t_{1/2} = 3.82$ days), and are of great social concern because they contribute more than half to the radiation dose a member of the general public receives from all natural radioactivity, and are a major cause of lung cancer, second only to cigarette smoking. Initially, RnDPs are positive metal ions which sooner or later, depending on the environmental conditions, are neutralized and attach to non-radioactive aerosols. They appear as aerosols, bimodally distributed in the 1–10 (unattached RnDP) and 200–800 nm size ranges (attached RnDP), of which the former is crucial with regards to detrimental health effect.

In the Postojnska Jama (jama = cave), as also at some other living and working environments in Slovenia, RnDPs have been monitored systematically for years in order to estimate radiation doses of the personnel and to keep them below an acceptable level (Vaupotič 2008;

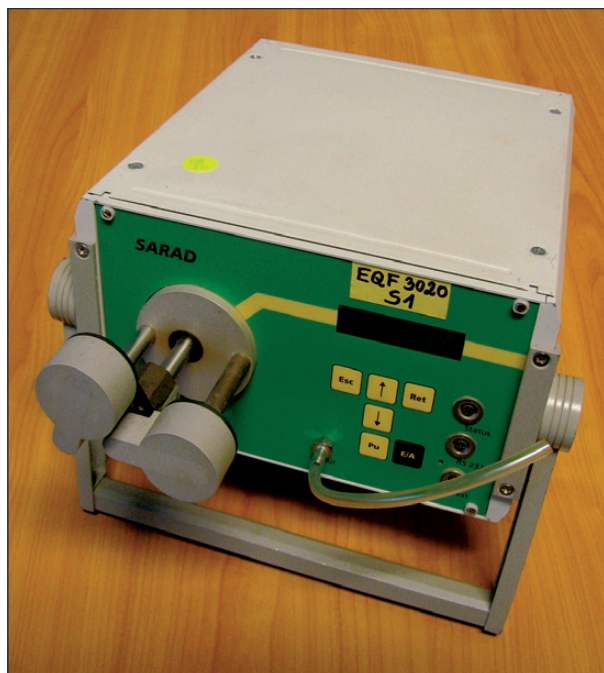
Vaupotič & Kobal 2007). The cave environment has been found as exceptional because of much higher concentration of the unattached RnDPs than anywhere else. In order to reveal whether this is a consequence of a very low concentration of non-radioactive nano aerosols, we initiated measurements of concentration and size distribution of non-radioactive aerosols in the 10–1,000 nm size range. In this way, we for the first time use a unique opportunity to study together both radioactive and non-radioactive nano aerosols.

The study is aimed at showing the levels of the nano-size non-radioactive aerosols in the cave and their dependence on the environmental conditions (i. e., barometric pressure and outdoor air temperature, as well as the working regime in the cave), and, consequently, at explaining the concentration levels of the 1–10 nm fraction of radioactive aerosols. In this paper, our measurements are described and preliminary results presented and commented on.

MATERIALS AND METHODS

RADIOACTIVE NANO AEROSOLS

Individual activity concentrations of ^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po (in Bq m^{-3} , 1 Bq is one radioactive transformation in a second) have been measured using the EQF3020 and EQF3020-2 devices (Sarad, Germany) (Fig. 1). Air



is pumped for 6 minutes at a flow rate of $2.4 \text{ dm}^3 \text{ min}^{-1}$ over a metal mesh grid on which aerosols smaller than 50 nm (considered as unattached RnDPs) are separated from those above this size (considered as attached RnDPs) and the two fractions are deposited electrostatically on two 150 mm^2 semiconductor detectors. Gross alpha activity is measured during three consecutive intervals within 110 minutes after the end of pumping and, applying the Markov method (Markov 1962; Streil *et al.* 1996), individual activity concentrations of radionuclides in both fractions are obtained in units of Bq m^{-3} . In order to facilitate comparison of concentrations of non-radioactive and radioactive aerosols, the activities (A in $\text{Bq} \equiv \text{s}^{-1}$, i. e., 1 Bq equals 1 radioactive transformation per second) of radionuclides were converted into their numbers (N), using their either decay constants ($\lambda = \ln 2 / t_{1/2}$) in the radioactivity law equation:

$$-\frac{dN}{dt} = A = \lambda \times N = \frac{\ln 2 \times N}{t_{1/2}} \rightarrow N = \frac{A \times t_{1/2}}{\ln 2}.$$

Number concentrations (in cm^{-3} , i. e., number of atoms in 1 cm^3) of ^{218}Po , ^{214}Pb and ^{214}Bi are denoted by

Fig. 1: Sarad EQF3020-2 device to measure activity concentrations of the unattached and attached radon decay products (Photo: J. Vaupotič).

C_{Po}^{un} , C_{Pb}^{un} and C_{Bi}^{un} , respectively, for the unattached, and C_{Po}^{att} , C_{Pb}^{att} and C_{Bi}^{att} , respectively, for the attached form. The fractions of unattached atoms of each radionuclide is defined as: $x_{Po}^{un} = C_{Po}^{un} / (C_{Po}^{un} + C_{Po}^{att})$, $x_{Pb}^{un} = C_{Pb}^{un} / (C_{Pb}^{un} + C_{Pb}^{att})$ and $x_{Bi}^{un} = C_{Bi}^{un} / (C_{Bi}^{un} + C_{Bi}^{att})$.

NON-RADIOACTIVE NANO AEROSOLS

Nanoparticle concentration and their size distribution have been measured with Grimm Aerosol SMPS+C instrument (Series 5.400) (Fig. 2). For that purpose, the long DMA unit was used, designed for the 10–1,100 nm size range. The DMA unit separates charged particles based on their electrical mobility. The unit is a cylindrical capacitor, consisting of an inner (HV-Rod) and an outer electrode (ground). The electrical mobility depends mainly on the particle size and electrical charge: the smaller the particle and the higher its electrical charge the higher is its mobility. The particles enter the CPC unit. It contains a heater saturator, in which alcohol vapour molecules condense onto the entering particles, thus causing them to grow into droplets. These droplets are then detected with a laser beam (DLS detection) and counted. Number concentrations (in cm^{-3}) is denoted by C_{nr} . Because the EQF device distinguishes between unattached and attached RnDPs at the size of 50 nm, concentration of non-radioactive aerosol smaller than 50 nm ($C_{nr}^{<50}$) and bigger than that $C_{nr}^{>50}$, as well as the fraction of the smaller ones, defined as $x_{nr}^{<50} = C_{nr}^{<50} / (C_{nr}^{<50} + C_{nr}^{>50})$, are calculated.

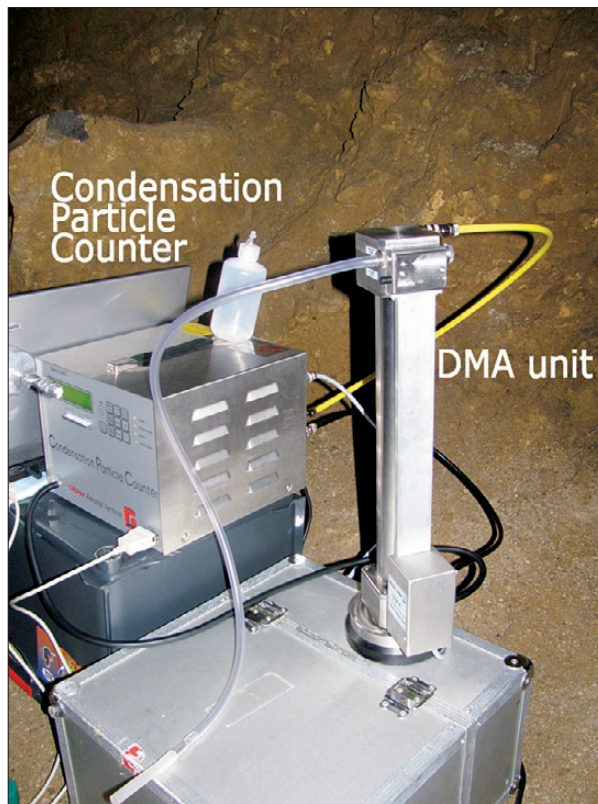


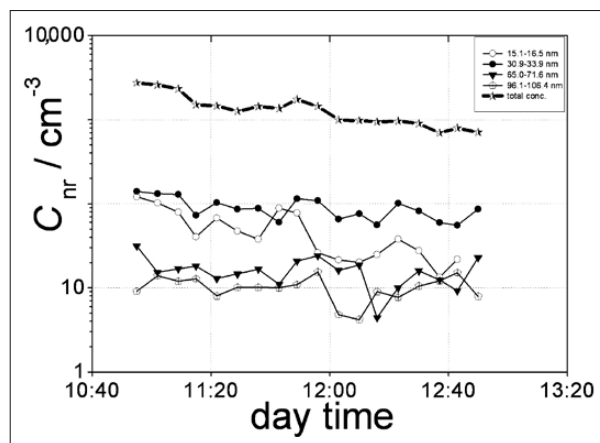
Fig. 2: Grimm Aerosol SMPS+C instrument, Series 5.400, to measure concentration and size distribution of aerosols in the size range 10–1,000 nm (Photo: I. Iskra).

RESULTS AND DISCUSSION

Results of our twenty 7-minute consecutive measurements of concentration and size distribution of the non-radioactive aerosols, carried out at the lowest point along the guided tourist route in the Postojnska Jama (about 5 m off the guided path) during normal visits of tourist on April 28, 2009, are plotted in Fig. 3. Although the total concentration of aerosols varies from one measurement to another, it is mostly contributed by the particles with diameter about 30 nm. Total concentration in the range of 600–2,740 cm^{-3} , is higher than that previously found in a ‘clean room’ (100 cm^{-3}), but lower than in our radon laboratory (9,500 cm^{-3}) at the Jožef Stefan Institute. At

the moment, it is not clear whether these aerosols are solid dust particulates, or merely clusters of water mol-

Fig. 3: Total concentration and size distribution (d: 10–1,000 nm) of non-radioactive aerosols at the lowest point along the guided tourist route in the Postojnska Jama during morning hours.



ecules in almost 100% humid cave air. Collection of aerosols on filters and their analysis will be performed in near future.

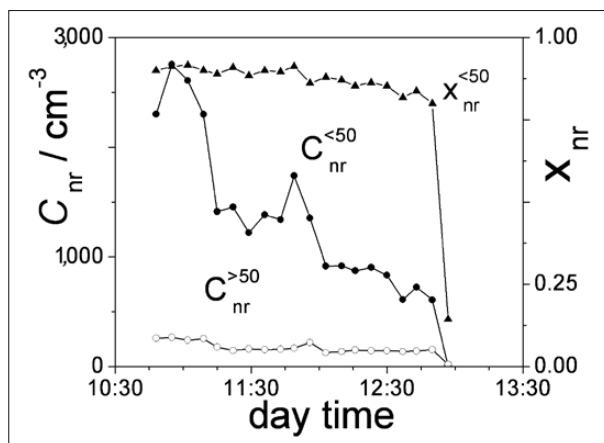


Fig. 4: Change of $C_{nr}^{<50}$, $C_{nr}^{>50}$ and $X_{nr}^{<50}$ (see the text for definition) at the lowest point along the guided tourist route in the Postojnska Jama during morning hours.

In relation to the RnDP aerosols, we are not so much interested in the total concentration of non-radioactive aerosols, but rather in the $C_{nr}^{<50}$ (the fraction carrying unattached RnDPs), $C_{nr}^{>50}$ and $x_{nr}^{<50}$ values. They are plotted in Fig. 4. The run of $x_{nr}^{<50}$ shows that $C_{nr}^{<50}$ is from five to ten times higher than $C_{nr}^{>50}$. During visits, the decrease of $C_{nr}^{<50}$ is substantially faster than that of $C_{nr}^{>50}$, probably because the deposition of smaller particulates is faster, and smaller fraction is preferentially both caught by cloths and deposited in visitor's lung (Hofmann *et al.* 1996).

At the lowest point along the guided tourist route in the cave, individual activity concentrations of ^{218}Po , ^{214}Pb and ^{214}Bi , both in unattached and attached form were continuously monitored previously, under different meteorological conditions and working regime (Vaupotič & Kobal 2007). Here, only results are discussed on, which had been obtained at the same time and season. Their number concentrations during morning hours are for both attached and unattached forms shown in Figs. 5a and 5b. These values are for orders of magnitudes lower than those for the non-radioactive aerosols, often less than one particle in 1 cm^3 . Therefore, one may speculate that only one radionuclide atom is attached to a non-radioactive aerosol particulate and, hence, the number of radionuclide atoms is equal to the number of radioactive particles detected by EQF device. As for the non-radioactive aerosols, also here all concentrations are decreasing during morning hours of visits, presumably because air movement enhances their deposition. Among the attached radionuclides (Fig. 5b), concentration of ^{214}Pb is highest, that of ^{214}Bi lowest and

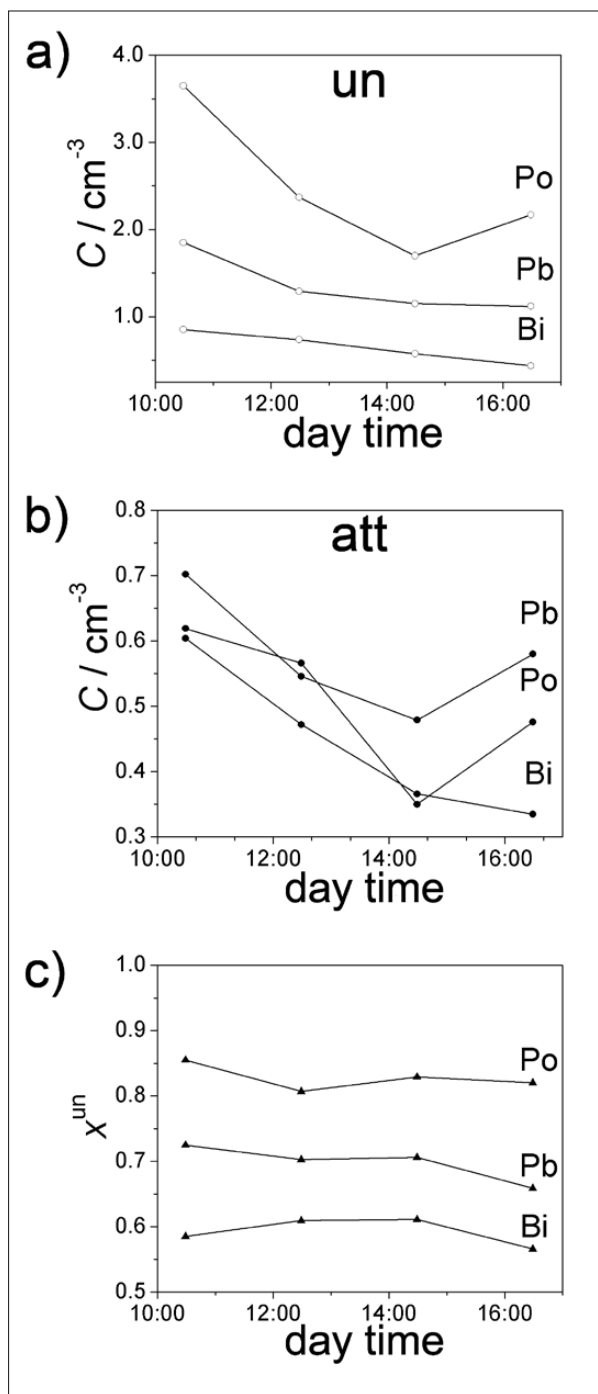


Fig. 5: Change of concentrations (C) of ^{218}Po , ^{214}Pb , ^{214}Bi : a) in the attached form and b) unattached form, and c) change of their unattached fractions x^{un} (see the text for definition) at the lowest point along the guided tourist route in the Postojnska Jama during morning hours.

that of ^{218}Po in between. On the other hand, among the unattached radionuclides (Fig. 5a), ^{218}Po appears at the highest levels. Fig. 5c shows the fractions of unattached

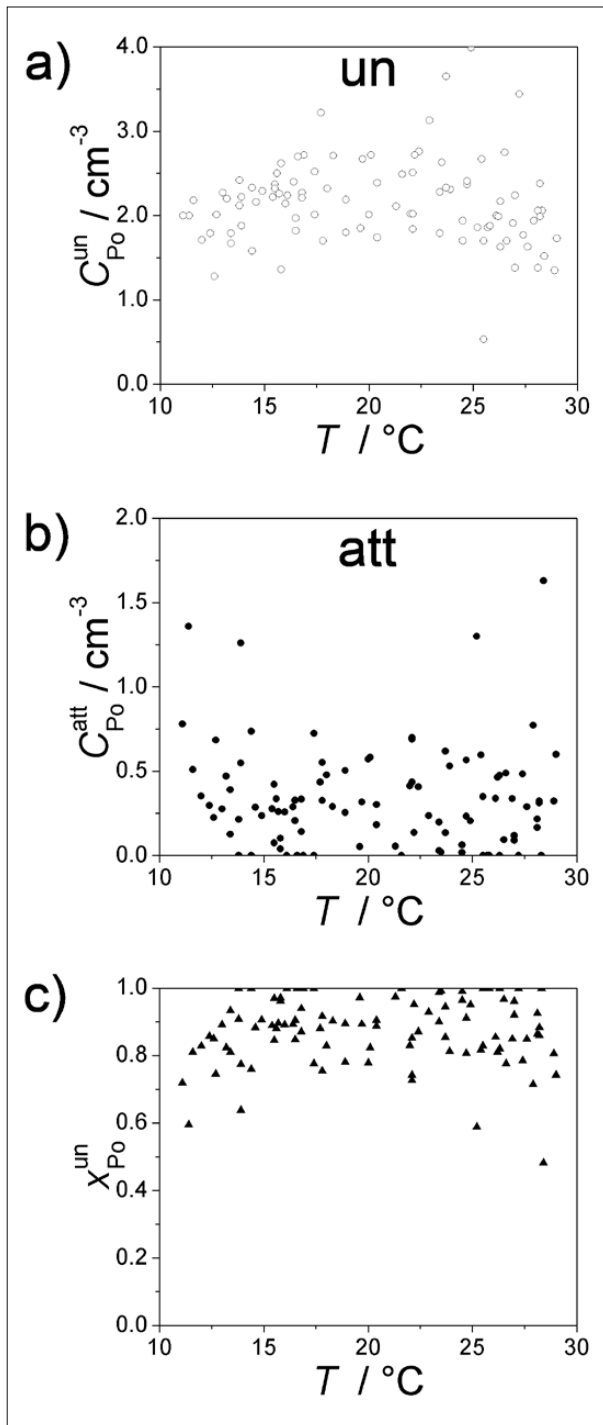


Fig. 6: The influence of the outdoor air temperature on: a) ^{218}Po concentration in the unattached form ($C_{\text{Po}}^{\text{un}}$), b) ^{218}Po concentration in the attached form, and c) the unattached fraction of ^{218}Po ($x_{\text{Po}}^{\text{un}}$).

RnDPs, with the highest values for ^{218}Po . This is very important from the dosimetry point of view because this

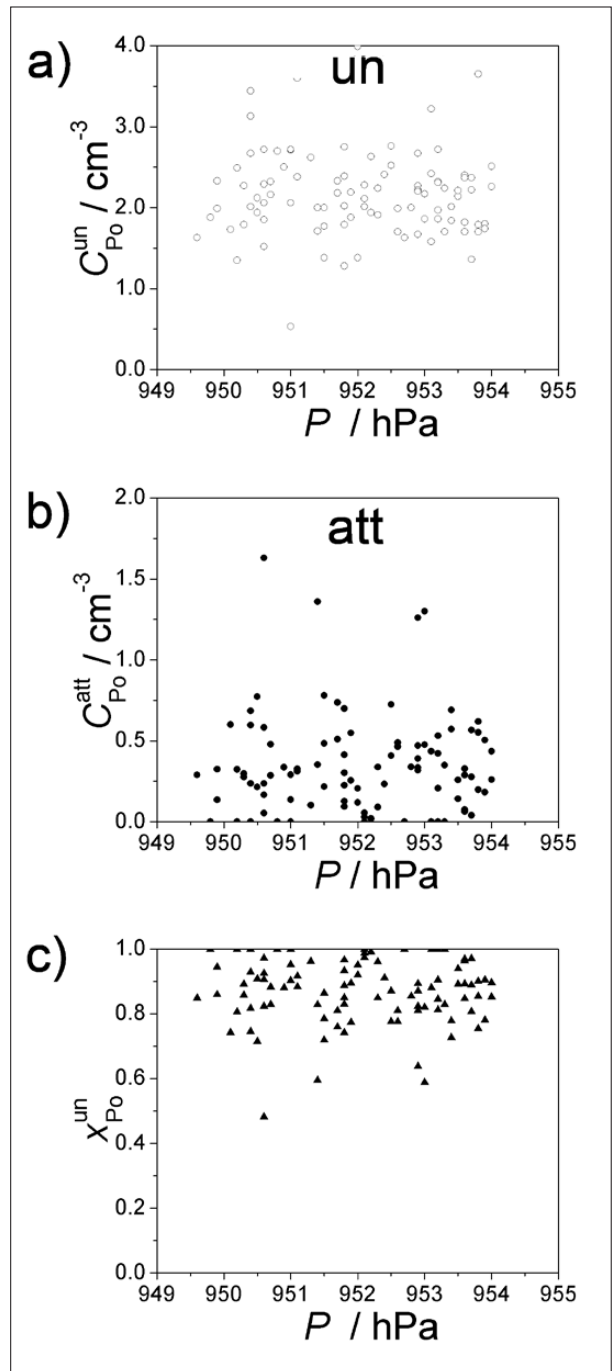


Fig. 7: The influence of the barometric pressure on: a) ^{218}Po concentration in the unattached form ($C_{\text{Po}}^{\text{un}}$), b) ^{218}Po concentration in the attached form ($C_{\text{Po}}^{\text{att}}$), and c) the unattached fraction of ^{218}Po ($x_{\text{Po}}^{\text{un}}$).

radionuclide in the unattached form causes a great detrimental effect on tissue cells and therefore has a major contribution to the dose conversion factor (Birchall & James 1994; Porstendörfer 1996).

These results may answer the question why the fraction of unattached RnDP in the Postojnska Jama is much higher than in other environments. Firstly, the concentration of non-radioactive aerosols to which RnDPs attach is lower than in other environments. And secondly, the concentration of smaller aerosols (<50 nm), carrying the unattached RnDPs, is ten times higher than that of bigger ones (>50 nm).

Although the activity concentration of ^{222}Rn , the source of RnDPs, in the Postojnska Jama has appeared to be influenced by both the barometric pressure and the

difference in air temperature in the cave and outdoors (Vaupotič 2008), no such influence on the number concentration of RnDPs has been observed. For sake of saving space, this is shown only for ^{218}Po , i. e.; dependence of $C_{\text{Po}}^{\text{un}}$, $C_{\text{Po}}^{\text{att}}$ and $x_{\text{Po}}^{\text{un}}$ on outdoor air temperature in Figs. 6a, 6b and 6c, and on barometric pressure in Figs. 7a, 7b and 7c. Further measurements will be carried out under different meteorological conditions (barometric pressure, outdoor air temperature) and working regime (with and without visitors) in order to provide a sound interpretation of these relationships.

CONCLUSION

Concentrations of non-radioactive aerosols during morning hours at the lowest point in the Postojnska Jama are by an order of magnitude lower than in the Radon laboratory. They amounted to 2,740 particles per cm^{-3} , with about 90% of particles smaller than 50 nm. Concentration of radioactive aerosols smaller than 50 nm was several atoms per 1 cm^3 and that of the bigger ones, less

than 1 atom per 1 cm^3 . The study will be continued in order to provide a sound interpretation of the attachment of radionuclides on the aerosol particulates and to improve our understanding of the influence of environmental parameters on the concentration and size distribution of aerosols in the Postojnska Jama.

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