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Composition, size distribution and lung deposition distribution of aerosols collected in the atmosphere of a speleotherapeutic cave situated below Budapest, Hungary

Zs. Kertész [a,b,*], I. Balásházy [c], I. Borbély-Kiss [b], W. Hofmann [d], I. Hunyadi [b], I. Salma [e], R. Winkler-Heil [d]

[a] *Max-Planck-Institut für Kernphysik Heidelberg, Pf. 103980, D-69029 Heidelberg, Germany*
[b] *Institute of Nuclear Research of the Hungarian Academy of Sciences, H-4001 Debrecen, P.O. Box 51, Hungary*
[c] *KFKI Atomic Energy Research Institute, H-1525 Budapest, P.O. Box 49, Hungary*
[d] *Institute of Physics and Biophysics, University of Salzburg, Hellbrunner Strasse 34, A-5020 Salzburg, Austria*
[e] *Department of Chemical Technology and Environmental Chemistry, L. Eötvös University, H-1518 Budapest, P.O. Box 32, Hungary*

Abstract

Elemental composition and mass size distribution of cave aerosols were determined by PIXE on seven-stage cascade impactor samples collected in two different sites of the Szemlőhegy-cave, a speleotherapeutic cave situated below Budapest, Hungary. In addition, individual particle analysis was also performed on about 450 aerosol particles. Significant differences were found between the two sampling sites and also in comparison with the external air in both the size distribution and in the composition of the aerosol. On the basis of the obtained data a stochastic lung deposition model was used to calculate total and regional deposition efficiencies of the different types of particles along the human respiratory system. One can conclude that the extrathoracic deposition is quite significant and its role increasing with increasing respiratory minute volume. The regional thoracic deposition is not very sensitive to the size distribution and it has a maximum around the 15–20th airway generations. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

The more accurate and complete knowledge of the climate parameters of cool karstic caves – among which cave aerosol plays a specific role – leads to the description of the interaction between the cave and its environment, and might help the better understanding of cave forming processes and of the healing effect of these caves.

The Szemlőhegy-cave, the location of this study, is the most well known and frequently visited hydrothermal cave of Budapest, Hungary. It is situated together with numerous highly protected caves under the densely populated Rózsadomb thermal karst area of the Buda Hills. Speleotherapy

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[*] Corresponding author. Address: Max-Planck-Institut für Kernphysik Heidelberg, Pf. 103980, D-69029 Heidelberg, Germany. Tel.: +49-6221-516591; fax: +49-6221-516540.
E-mail address: zsofia.kertesz@mpi-hd.mpg.de (Zs. Kertész).

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of respiratory diseases was started here more than ten years ago, and very favourable healing rates have been detected in several hundred adult and child patients treated annually by the Pulmonology Department of the St. János Hospital [1]. The cases of respiratory diseases caused by the increasingly polluted city environment are increasing significantly, and nowadays in Hungary this disease is considered as endemic. The conventional medicinal treatment is not effective enough, and has disadvantageous side effects. On the other hand, it was observed that a treatment-like stay in a cave could significantly improve the respiratory function of both healthy and diseased individuals. The curative influence of cool karstic caves with dripping water was experienced empirically, and its mechanism is still fairly unknown (few correlated healing factors thought to be the following: the germ- and pollen-free atmosphere, the high CO_2 content, the high Rn activity concentration, the antiphlogistic Ca^{++} and Mg^{++} ion content of small droplets originating form the dripping water, and the favourable psychosomatic effect of the cave environment) [2].

In order to reach a better understanding of the possible health effects of cave aerosols – either positive healing effects or the radiation exposure due to radon daughters attached to aerosol particles – it is important to know their composition, size distribution and deposition probabilities along the human airways.

In our previous work [3] we have already investigated the interaction between this cave and its environment through measuring the elemental composition, spatial distribution and seasonal variation of cave aerosol with special regard to the spread of the outer air pollution. Here we deal with the size distribution and the possible healing effects of solid aerosols of the cave atmospheres in more detail by determining their elemental composition, mass size distribution and their deposition in the human respiratory system.

2. Sampling and analysis

Cascade impactor samples were collected in the Szemlőhegy-cave in the framework of an aerosol sampling campaign carried out in March 1998. Two sampling sites were chosen: one was situated deep in the cave (Giant-gallery), where the speleotherapy itself takes place (the patients spend few hours here sitting or walking every day during the 3–6 weeks long treatment), and the other location was in a large hall (Loam-pit), closer to the surface, which is designated for complementary gymnastics activity (Fig. 1). The 48 h samplings were performed 1.5 m above the floor level of the cave with a seven-stage PIXE International cascade impactor, which allowed the separation of the aerosol within the size range of 0.25–32 μm into seven fractions: particles with 0.25–0.5, 0.5–1, 1–2, 2–4, 4–8, 8–16 and 16 μm or larger aerodynamic diameter. Due to the nearly 100% relative humidity of the cave environment, an after-filter was not used.

The elemental concentrations of the samples were measured by PIXE in the Institute of Nuclear

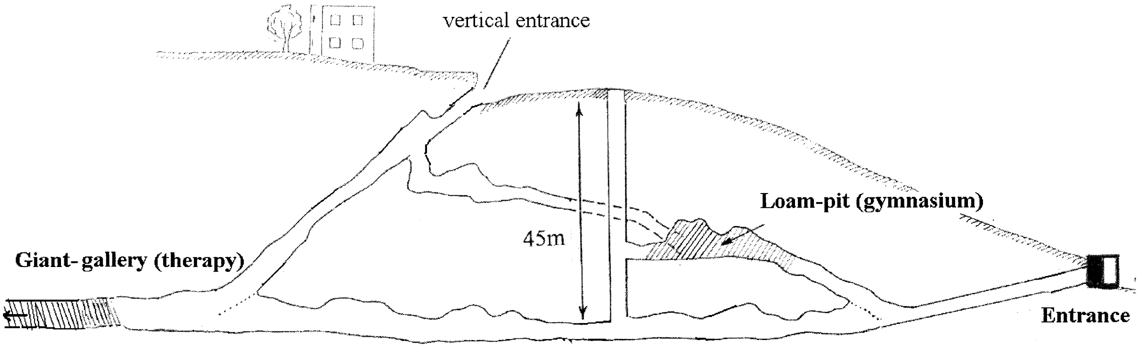

Fig. 1. Schematic map of the Szemlőhegy-cave with the two sampling sites.

Research of the Hungarian Academy of Sciences, Debrecen. A 2 MeV proton beam from the 5 MeV Van de Graaff accelerator was used to irradiate the samples [4].

In order to determine the sources and the possible chemical composition of cave aerosol, individual particle analysis was also carried out on the same samples, using the scanning proton microprobe of the ATOMKI [5]. The experimental arrangement and the detail of the measurement are described in [6]. Elemental maps and PIXE spectra of about 450 aerosol particles with aerodynamic diameter larger than 0.5 μm, were acquired.

The obtained spectra were evaluated with the PIXYKLM programme package [7]. Concentrations of the following elements were determined: Na, Mg, Al, Si, P, S, Cl, K, Ca, Sc, Ti, V, Mn, Cr, Fe, Ni, Cu, Zn, As, Ba, Mo and Pb. Hierarchical cluster analysis was performed to group the particles into clusters of similar composition.

On the basis of this data set, total and regional deposition efficiencies of the different types of particles along the human respiratory system were calculated by applying the stochastic lung deposition model of Koblinger and Hofmann [8,9] for the case of adult female, male and 5 years old child.

3. Results and discussion

As a result of the cluster analysis and correlation studies of elemental composition of individual aerosol particles, four main sources of solid cave aerosols could be identified: (1) the cave walls covered with carbonate minerals like calcite and aragonite crystals ($CaCO_3$); (2) the clay layer, which is a mixture of clay minerals (different aluminosilicate minerals, detrital quartz and feldspar) and iron oxides; (3) the external air of Budapest; and (4) "geo-aerosols" with high Zn and sometimes Cu content originating from warm streaming points in different locations of the cave.

3.1. Mass size distribution of cave aerosols

The mass size distributions at the two sampling sites obtained from the bulk PIXE measurement are shown in Fig. 2. In comparison mass size distributions of the same elements measured in the Budapest air are also shown. The most conspicuous difference between the urban and cave aerosols – besides the variation in the mass size distribution – is the presence of much more Si and Ca in the cave atmosphere. Regarding these distributions, significant differences have been found between the two locations within the cave.

The mass size distribution of elements originating from the clay layer of the cave like Al, Si, K, Ti, Fe and partly Ca, has a maximum at the 4–8 μm size range in the case of the Loam-pit, and at the >16 μm size in case of the Giant-gallery. This difference, and also the difference in their concentrations, can be attributed to the fact, that while in the Loam-pit the ground is clay and exploration works are also carried out, in the Giant-gallery the ground is covered with concrete and is cleaned regularly by water. So in this latter location there is no direct source of clay particles and due to hygroscopic growth in the nearly 100% relative humidity cave environment, their size increases.

Particles originating from the crystal cover of the cave walls characterized by Ca (calcite and aragonite), S (gypsum), Si (quartz) were found in the >16 μm size range.

Concentrations of elements coming from external anthropogenic pollution (like S, Cl, V, Zn, Cu and Pb) were found to decrease with increasing distance from the entrance (due to the cleaning effect of the cave) [3], and a growth in their size could be observed. At the site of the therapy (Giant-gallery) their presence was minimal, while in the Loam-pit, which is situated closer to the entrance and to the surface, much higher concentration values could be observed. Cl is a good example for this: while in the Loam-pit significant amount of Cl was measured with a size distribution very similar to S, in the Giant-gallery its concentration remained under the detection limit with the exception of the >16 μm size fraction.

In the 'warm' streaming points of the cave the 2 °C warmer air brings aerosol with itself, which contains a high amount of Zn, and some Cu, S, Ca, K, Cl and Fe. This 'geo-aerosol' can also influence the aerosol content of the cave atmosphere, particularly in the smaller size fractions.

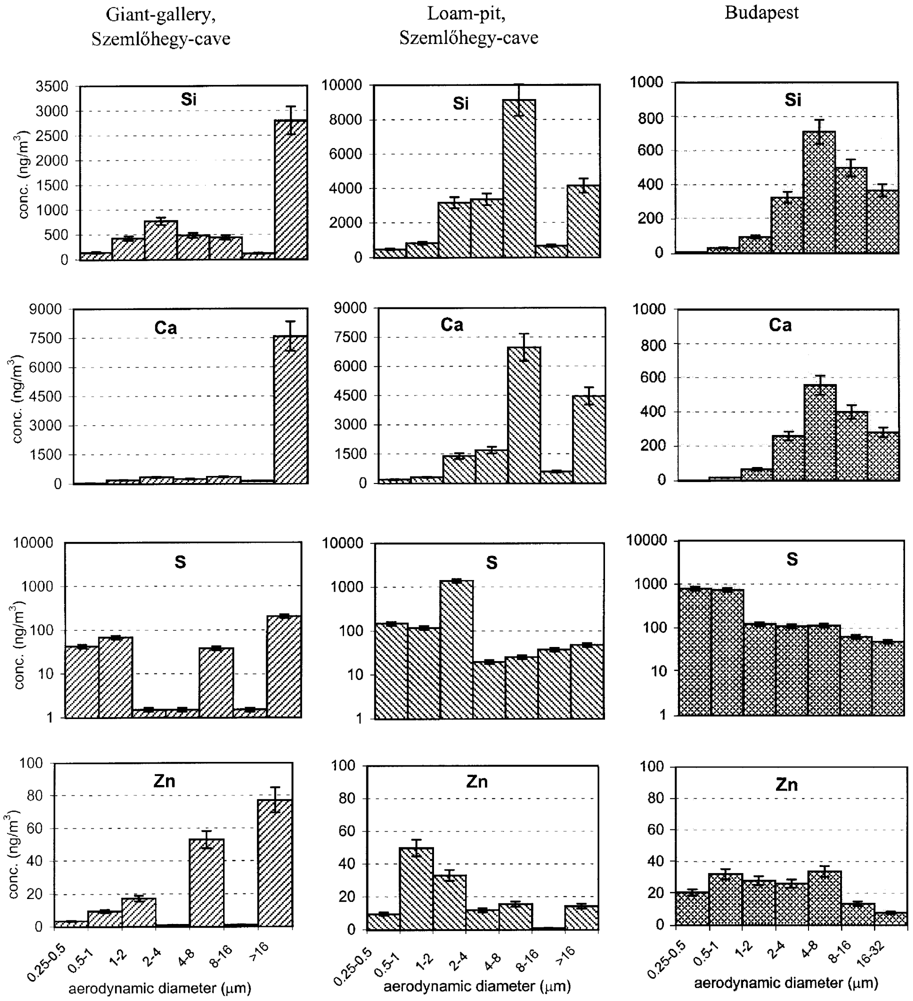

Fig. 2. Mass size distributions for some elements in the two sampling sites of the cave, and in the Budapest air too.

3.2. Lung deposition distributions

Deposition fractions, that is the ratio of deposited and simulated particles, in the human respiratory system were calculated with the above mentioned stochastic lung deposition model IDEAL4 [8,9]. For modelling purposes the respiratory system is divided into extrathoracic, tracheobronchial and acinar regions. The extrathoracic region contains nasal and oral passages and the throat. The tracheobronchial part consists of the so-called tracheobronchial tree which connecting dichotomic (slightly asymmetric) bifurcations conduct the air from the trachea to the alveolar regions. Here, an airway can be characterised by the airway generation number that is by the number of bifurcations from the trachea. After 12–21 generations, the alveoli appear on the airways representing the beginning of the acinar region where the direct gas exchange happens. To the end of the airway paths, the number of airway generations can be over 30 [8–10]. Results of the computations for some elements in the two sampling sites are presented in Table 1. The deposition fractions of the extrathoracic, tracheobronchial and the acinar regions were calculated for an adult

Table 1
Extrathoracic (eth.), tracheobronchial (bron.), acinar (acin.) and total ($D_{sum} = D_{eth} + D_{bron} + D_{acin}$) depositions in % for some elements in the two sampling sites of the Szemlőhegy-cave

	Adult male				Adult female				5-year-old child		
	eth.	bron.	acin.	sum.	eth.	bron.	acin.	sum.	eth.	bron.	acin.
Giant-gallery											
Si	70.4	2.07	4.43	76.9	68.1	2.1	3.26	73.5	67.1	1.45	1.05
Ca	91.9	0.741	1.29	93.9	91.1	0.87	0.949	92.9	90.5	0.69	0.316
S	68	1.48	2.86	72.3	67.1	1.55	2.02	70.7	66.1	1.09	0.76
Cu	68.5	4.55	5.35	78.4	63.6	5.19	4.75	73.5	60.3	3.78	1.97
Zn	76.6	2.69	3.8	83.1	73.6	3.0	3.17	79.8	71.9	2.19	1.2
Loam-pit											
Si	65.2	4.33	6.79	76.3	60.1	4.72	5.52	70.3	57.1	3.34	2.06
Ca	74.3	3.94	5.35	83.6	69.9	4.41	4.51	78.8	66.8	3.19	1.78
S	20.2	3.56	11.2	34.96	16.9	3.18	7.39	27.5	14.9	1.84	1.94
Cl	54.9	3.16	7.05	65.1	51.2	3.26	5.1	59.6	49.4	2.19	1.57
Cu	23.9	3.07	8.15	35.1	21.4	2.87	5.34	29.6	20	1.74	1.66
Zn	30.1	3.34	8.33	41.8	27.2	3.18	5.79	36.2	25.7	1.99	1.78

male, female and a 5-year-old child under sitting breathing conditions [10].

It can be seen that the total deposition fraction decreases with decreasing respiratory minute volume, i.e. higher for male than for female, and higher for female than for child. In the Giant-gallery, where the majority of aerosol particles can be found in the coarse mode, the total deposition fraction is between 71 and 94%, and the extrathoracic deposition plays a significant role. This can be explained by the fact that the filtration mechanism of the extrathoracic part of the respiratory system for coarse mode particles is rather effective: most of these coarse particles are trapped before or in the large bronchial airways. In the Loam-pit, where the average diameter of the aerosol particles is smaller, higher thoracic deposition rates are found although the total deposition fractions for elements of natural origin are similar to the previous ones. For elements of anthropogenic origin (the size distribution of which the accumulation mode is the dominant) the total deposition is about 30–40%. This means that the majority of these particles remain airborne, and are exhaled. The deposition probabilities of the urban aerosol are very similar to those achieved in the case of the Loam-pit. More detailed description of this can be found in [11].

To illustrate the regional thoracic distribution, the deposition of Ca is shown in Fig. 3 as a function of bifurcation numbers for male, female

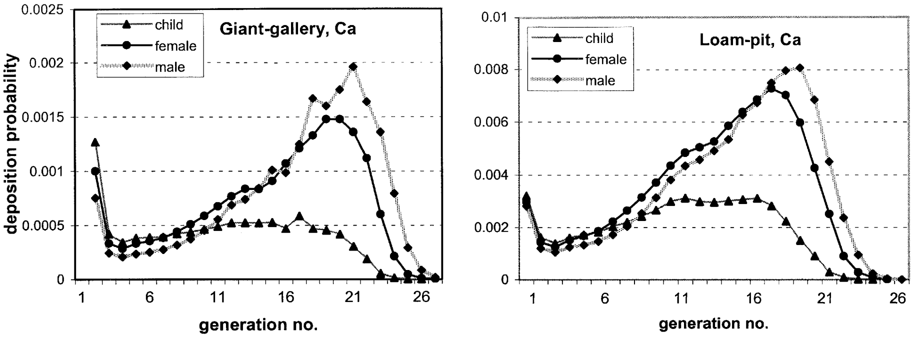

Fig. 3. Thoracic deposition of Ca as a function of airway generation number calculated for adult male, female and 5-year-old child under sitting breathing conditions in the two sampling sites of the Szemlőhegy-cave.

and child at the two sampling sites. The distribution of the thoracic deposition is not very sensitive to the size distribution of the aerosol particles, i.e. the shape of the curves is very similar for all particle types mentioned previously. The maximum deposition occurs between the 16th and the 23rd airway generations, that is already in the acinar region. In the Loam-pit, the deposition fractions are about four times higher than in the Giant-gallery representing the much cleaner atmosphere of the Giant-gallery. Deposition in the pulmonary regions is significantly lower for the 5-year-old child than for female or male adults, originating from the higher extrathoracic deposition in case of children which is based on the faster breathing frequency and thinner airway dimensions.

4. Conclusions

Despite the fact that the Szemlőhegy-cave is situated in a highly polluted environment, the atmospheric air deep in the cave is very clean, especially from the point of view of thoracic deposition of aerosol particles originating from outer anthropogenic pollution.

It was also confirmed by this study that the Loam-pit part of the cave is not appropriate for therapeutic purposes, since high concentration values were observed of the aerosol component originating from outer anthropogenic pollution, and also the lung deposition fractions were found to be 3–6 times higher than in the Giant-gallery. This hall lies too close to the surface, and the filtering effect of its covering karst layer is not sufficient enough – which was pointed out from previous radon and temperature measurements, as well [12].

The understanding of the healing mechanism of cool karstic caves and their unique atmosphere is very difficult, because of its multiple complexity. Besides the assumed contributing healing parameters, very little information can be found in the field of "speleo-aerosols", so this kind of study, which provides quantitative data, may help towards a better understanding of special health effects during speleotherapy.

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