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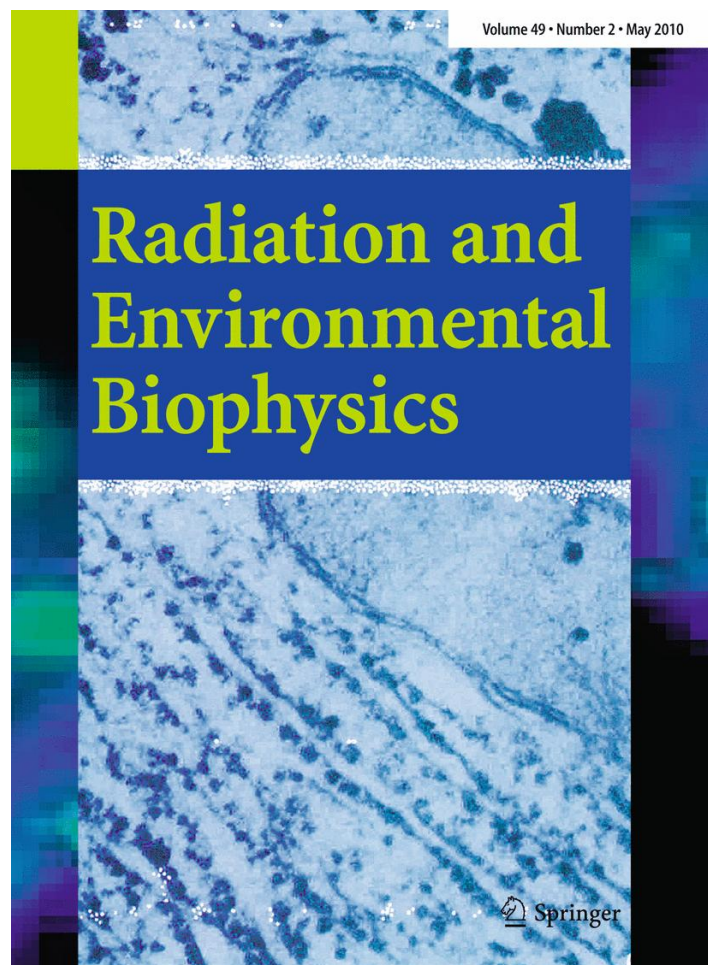
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ISSN 0301-634X, Volume 49, Number 2



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Deposition of radon progeny on skin surfaces and resulting radiation doses in radon therapy

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Received: 3 July 2009 / Accepted: 20 February 2010 / Published online: 17 March 2010
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Abstract In the Gastein valley, Austria, radon-rich thermal water and air have been used for decades for the treatment of various diseases. To explore the exposure pathway of radon progeny adsorbed to the skin, progeny activities on the skin of patients exposed to thermal water (in a bathtub) and hot vapour (in a vapour chamber) were measured by alpha spectrometry. Average total alpha activities on the patients' skin varied from 1.2 to 4.1 Bq/cm² in the bathtub, and from 1.1 to 2.6 Bq/cm² in the vapour bath. Water pH-value and ion concentration did affect radon progeny adsorption on the skin, whereas skin greasiness and blood circulation did not. Measurements of the penetration of deposited radon progeny into the skin revealed a roughly exponential activity distribution in the upper layers of the skin. Based on the radon progeny surface activity concentrations and their depth distributions, equivalent doses to different layers of the skin, in particular to the Langerhans cells located in the epidermis, ranged from 0.12 mSv in the thermal bath to 0.33 mSv in the vapour bath, exceeding equivalent doses to the inner organs (kidneys) by inhaled radon and progeny by about a factor 3, except for the lung,

which receives the highest doses via inhalation. These results suggest that radon progeny attachment on skin surfaces may play a major role in the dosimetry for both thermal water and hot vapour treatment schemes.

Introduction

In Bad Gastein and Bad Hofgastein, Austria, located in the province of Salzburg at the Northern slope of the Alps, natural radon-rich thermal water and radon vapour have been used for decades for the treatment of various rheumatic diseases such as ankylosing spondylitis, a painful autoimmune damage of the spinal cord (van Tubergen et al. 2001). In fact, several clinical studies have proved the effectiveness of radon treatment, especially its long-term pain relief after the treatment (Bernatzky et al. 1994; Falkenbach 2000; Falkenbach et al. 2005; Franke et al. 2000; Pratzel et al. 1999; Schwarzmeier and Shehata 2006; Shehata et al. 2006; van Tubergen et al. 2001). However, knowledge of the physiological and molecular mechanisms and their time sequence triggered by the radon exposure is still too fragmentary for a complete mechanistic understanding. In addition, our current knowledge of the relative efficiencies of different treatment regimes, and of the biophysical mechanisms of radon and radon progeny uptake and related radiation doses is still insufficient.

Two different therapeutic exposure regimes are presently used in Bad Gastein and Bad Hofgastein: (1) bathing in warm thermal water, and (2) exposure to hot radon vapour in an exposure chamber ("Dunstbad") or in the thermal gallery ("Gasteiner Heilstollen"), a former gold and silver mine. In the thermal bath, patients are sitting in a bathtub (37°C) for a period of 20 min (usually nine times for a complete treatment), with a radon concentration of

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about 900 Bq/l. Measurements of the radon concentration in air exhaled by patients indicated that a certain fraction of the radon in the water was taken up by the blood after diffusion through the skin, transported to the lungs (and other organs) via the bloodstream, and then exhaled (Grunewald et al. 1999; Hofmann et al. 1999). In addition, short-lived radon progeny (^{218}Po , ^{214}Pb and $^{214}\text{Bi}/^{214}\text{Po}$) were adsorbed on the skin of the patients (von Philipsborn 1997).

In the vapour bath (37°C), patients are exposed to radon in a small chamber for a period of 20 min (usually 10 times for a complete treatment), with a radon concentration ranging from 30 to 200 Bq/l (average value: 90 Bq/l). These exposure conditions are similar to those found in the thermal gallery (usually 10 sessions of 1 h for a complete treatment), with the notable exception that the head of the patient remains outside of the exposure chamber, thus avoiding inhalation of radon and its progeny (note: inhalation of radon is the primary source of the organ doses in the thermal gallery). In addition, short-lived radon progeny are adsorbed on the skin of the patients.

In most cases, patients receive such treatments once a year for 3 weeks, with about three sessions per week, sometimes combining thermal bath, vapour bath, thermal gallery and underwater therapy. In the special case of ankylosing spondylitis (Morbus Bechterew), a positive therapeutic effect has been reported already after the first treatment (Shehata et al. 2006), which may be repeated several times in the following years.

For decades, the therapeutic response—particularly for patients treated in the thermal gallery—was explained by the radiation doses incurred in different organs of the human body via inhalation of radon and its short-lived progeny (Pohl 1979; Hofmann 1992). However, an alternative explanation may be an immune response in the exposed patients, caused by irradiation of the deeper layers of the skin by radon progeny adsorbed to the skin (Andrejew 1990; Andrejew et al. 1990). Especially the so-called Langerhans cells located in the epidermis may be stimulated by irradiation to trigger an immune response (Pratzel et al. 1999; Flatscher 2006). However, apart from a few rough estimates of skin doses (Andrejew 1990; Andrejew et al. 1990; Bogoljubow et al. 1990) and radon progeny activities on the skin and hair of patients in the thermal gallery (Falkenbach et al. 2002), quantitative nuclide-specific information about the deposition of radon progeny in water and air on skin surfaces is still missing. For this reason, the present study aims at the investigation of the transport of radon progeny from radon-rich water and radon vapour to the epidermis of the skin. Resulting doses to sensitive target cells and tissues for specific exposure conditions typical for the radon spas in Bad Gastein and Bad Hofgastein are given.

Materials and methods

Radon in water and air

Radon activities in thermal water and radon vapour in the vapour bath and in the thermal gallery were measured by means of a 10-l ionisation chamber. The radon concentration in the thermal water was relatively constant at about 950 ± 73 Bq/l, while the radon concentrations in the vapour bath and in the thermal gallery ranged from 25 to 200 Bq/l. This variability is caused by temporal variations in radon production in the water wells due to weather-dependent variations in atmospheric pressure (Pohl-Rüling and Pohl 1969).

Radon progeny in water

Radon progeny (Rnp) activities in water were measured by exposing 4-cm² silver plates to 10 l of thermal water. The activities of the decay products ^{218}Po , ^{214}Pb and $^{214}\text{Bi}/^{214}\text{Po}$ on the plates were then determined from the spectra after 20 min of exposure, and decay curves were recorded by an 8-channel alpha spectrometer (Ortec Octete®) with 1,200 mm² detectors (Canberra CAM 1200 AM). The measured activities were compared to those measured in 10 l of Ra-standard water, which has been stored in a Rn-proof closed container overnight, to obtain equilibrium between radon and its progeny. In both Ra-standard water and thermal water, pH was adjusted to 7.0 to avoid any kind of precipitation. The results indicated that equilibrium between radon and its short-lived progeny was obtained already a few minutes after filling the bathtub. Since almost no radon progeny were found on the walls of the bathtubs, equilibrium conditions remain during the whole treatment period (Surbeck and Andrey 1999; Surbeck 2000).

Radon progeny deposition on skin surfaces

Patients were exposed to thermal water (thermal bath) or hot radon vapour (vapour bath) for a period of 20 min. Immediately after the patients had left the bathtub or the exposure chamber, the wet skin was tabbed with a towel. Alpha detectors were attached to four different locations of the body, one on the forearm, one on the lower leg and two on the abdomen. Alpha spectra and decay curves were then measured for 30 min in 1-min intervals to determine the Rnp surface activities (Lettner et al. 2005, 2006).

Individual factors were also investigated that may affect the adsorption process in thermal water and thus explain the apparent inter-subject variations: greasiness of the skin, blood circulation, and transpiration (Falkenbach et al. 2000): (1) The influence of skin greasiness on Rnp

adsorption was examined by comparing two pieces of pig skin—a normal skin and one with the fat removed prior to exposure. (2) A test person's arm was treated with a vasodilating ointment (Finalgon®, Boehringer-Ingelheim) to intensify blood circulation of the skin. (3) The influence of skin transpiration on adsorption was examined by exposing a test person after 15 min of intensive jogging; 5 h later, the same person was exposed without prior physical activity. Since transpiration during the bath increases the ion concentration in the thermal water, the influence of such an increase was examined by filling thermal radon water with different NaCl concentrations into 25-l containers. The temperature was held constant at 35°C. For comparison, one arm of a test person was exposed to the salt-enriched water, while the other was exposed to natural thermal radon water. After 20 min of exposure, the Rnp activity on the skin was measured.

The pH-value of water is likely to influence adsorption of waterborne ions (Ziechmann 1990). To investigate this effect, thermal radon water was filled into 25-l containers and the pH-value was varied between 3 and 9.5 by adding HCl or NaOH. For comparison, one arm of a test person was exposed to water with different pH-values, while the second arm was exposed to pH-neutral thermal radon water.

Depth distribution of radon progeny in the upper skin layers

To assess radiation doses to the deeper layers of the skin where the Langerhans cells are located that are presumed to stimulate an immune response, information on the vertical distribution of the Rnp in the upper layers of the skin is required. Such information is also necessary for the determination of the efficiency of the alpha spectrometer. Therefore, single cell layers were successively removed by attaching strips of Tesa-film® (area: 16 cm²) to the skin, pulling them off after a few seconds (Pratzel 1985). The Rnp activity at the treated spot was measured and compared with a second detector attached to the untreated skin. These measurements were repeated with 2, 4, 6 and 8 pull-offs. Microscopic analysis of eosin-stained cells indicated that two pull-offs remove on average one cell layer. Since the thickness of an epidermic corneocyte swollen in water is about 5 µm (Talreja et al. 2001), it was assumed that two pull-offs correspond to a depth of about 5 µm. After microscopic analysis of the pulled-off strips, a vertical activity distribution relative to the untreated skin could be plotted.

Radon progeny activity deposition during exposure

In the bathtub, radon progeny in the water are in equilibrium or at least close to equilibrium. According to the

experimental findings of Surbeck and Andrey (1999) and Surbeck (2000), Rnp adsorbed on the skin are desorbed again after their radioactive decay. In this case, the surface activity can be expressed by a simple build-up function which considers each radon progeny separately. Thus, the specific activity of the *i*-th Rnp on the skin surface, $C_{Rnp(s),i}$ (Bq/cm²), is controlled by the attachment velocity, φ_i (cm/s), the concentration of the *i*-th radionuclide in the water, $C_{Rnp(w),i}$ (Bq/cm³) and its decay constant λ (s⁻¹) according to Eq. 1:

$$C_{Rnp(s),i} = \frac{C_{Rnp(w),i} \cdot \varphi_i (1 - e^{-\lambda_i t})}{\lambda_i} \quad (1)$$

It can be assumed that the attachment velocities of the various Rnp are very similar, as all Rnp are heavy metals coated by a water layer. If equilibrium is achieved for all Rnp in water and their attachment velocities are equal, the saturation activity concentrations behave like the Rnp half-lives.

In the vapour bath and in the thermal gallery, Rnp in the atmosphere are not in equilibrium. For example, *F*-values determined in the gallery vary between 0.4 and 0.6 (Lettner et al. 1996). In addition, the activities of ²¹⁸Po, ²¹⁴Pb and ²¹⁴Bi/²¹⁴Po are different due to attachment processes and the chronological order of decay, suggesting that Rnp activities are ordered as $A(^{218}\text{Po}) > A(^{214}\text{Pb}) > A(^{214}\text{Bi}/^{214}\text{Po})$. Aerosol growth along with the reduction of the attachment velocity may further modify the activity flux onto the skin.

Unlike in the thermal water baths, Rnp attached to the skin in the vapour bath will not desorb. Instead, Rnp will further decay on the skin and contribute to the next radionuclide in the decay chain. Thus, the theoretical model has to consider the flux of a given Rnp, its decay and its contribution to the daughter Rnp. The specific activity of the *i*-th radionuclide on the surface, $C_{Rnp(s),i}$, is controlled by the attachment velocity, φ_i , the concentration of the *i*-th radionuclide in air and the decay of the precursor Rnp already deposited on the skin. The algorithm to model the decay chain by the solution of the Bateman equations was implemented in a MathCad computer code.

Results

Activity of radon progeny on the skin

To examine potential intra- and inter-individual variations of the Rnp surface activities, six selected patients (three males, three females) were measured three to four times within 1 week. The results of these measurements for both exposure conditions are listed in Tables 1 and 2. Significant differences among the six individuals could be

Table 1 Radon progeny activities on various body surfaces exposed to thermal water; exposure time: 20 min

Surface	Surface activities (Bq/cm ²)			
	²¹⁸ Po	²¹⁴ Pb	²¹⁴ Bi/ ²¹⁴ Po	Total
Forearm, females (<i>n</i> = 11)	0.20 ± 0.13	0.96 ± 0.17	0.80 ± 0.11	1.96 ± 0.82
Abdomen, females (<i>n</i> = 22)	0.22 ± 0.10	0.57 ± 0.07	0.56 ± 0.17	1.35 ± 0.19
Lower leg, females (<i>n</i> = 11)	0.20 ± 0.14	0.62 ± 0.20	0.42 ± 0.01	1.24 ± 0.24
Forearm, males (<i>n</i> = 10)	0.31 ± 0.13	1.97 ± 0.82	1.81 ± 1.02	4.09 ± 1.33
Abdomen, males (<i>n</i> = 20)	0.34 ± 1.18	1.12 ± 0.54	1.07 ± 0.60	3.53 ± 0.82
Lower leg, males (<i>n</i> = 10)	0.45 ± 0.23	1.30 ± 0.44	0.90 ± 0.33	2.65 ± 1.00
Average values*	0.29	1.09	0.93	2.47
Copper (<i>n</i> = 4)	0.18 ± 0.04	0.25 ± 0.07	0.23 ± 0.08	0.66 ± 0.11
PVC (<i>n</i> = 4)	0.10 ± 0.02	0.33 ± 0.08	0.13 ± 0.02	0.56 ± 0.08

* Averaged over all test persons and skin surfaces

Table 2 Radon progeny activities on various body surfaces exposed to radon vapour; exposure time: 20 min

Surface	Surface activities (Bq/cm ²)			
	²¹⁸ Po	²¹⁴ Pb	²¹⁴ Bi/ ²¹⁴ Po	Total
Forearm, females (<i>n</i> = 11)	1.09 ± 0.62	0.74 ± 0.37	0.32 ± 0.21	2.15 ± 0.75
Abdomen, females (<i>n</i> = 22)	0.77 ± 0.33	0.50 ± 0.20	0.18 ± 0.15	1.45 ± 0.41
Lower leg, females (<i>n</i> = 11)	0.50 ± 0.19	0.41 ± 0.17	0.14 ± 0.09	1.05 ± 0.27
Forearm, males (<i>n</i> = 10)	1.29 ± 0.51	0.96 ± 0.44	0.33 ± 0.18	2.58 ± 0.70
Abdomen, males (<i>n</i> = 20)	0.86 ± 0.38	0.49 ± 0.15	0.23 ± 0.13	1.58 ± 0.43
Lower leg, males (<i>n</i> = 10)	0.57 ± 0.22	0.40 ± 0.18	0.18 ± 0.08	1.15 ± 0.30
Average values*	0.85	0.58	0.23	1.66
Copper (<i>n</i> = 4)	1.43 ± 0.40	1.37 ± 0.57	0.58 ± 0.28	3.38 ± 1.25
PVC (<i>n</i> = 4)	1.63 ± 0.62	1.49 ± 0.65	0.53 ± 0.18	3.65 ± 1.45

* Averaged over all test persons and skin surfaces

observed. Despite the small sample size, males appear to adsorb radon progeny better than females in thermal water, while almost no differences were observed for radon vapour. Activities measured on artificial inorganic surfaces (copper and PVC), when exposed to thermal water, were on average about four times lower than those on the human skin. In contrast, Rnp activities measured on these surfaces, when exposed to vapour, were higher by about a factor 2 than those measured on the skin.

All measurements on human skin showed that ²¹⁸Po, ²¹⁴Pb and ²¹⁴Bi/²¹⁴Po are adsorbed from the thermal water in a ratio of about 1:4:3 after 20 min (see Table 1). The reason for this phenomenon is that daughters of adsorbed nuclides get desorbed from a wet surface by the decay of the mother nuclides. This desorption process following radioactive decay has been experimentally verified by Surbeck and Andrey (1999). At steady-state conditions, i.e. after about 2 h, the ratios of the Rnp activities adsorbed on a wet surface reflect corresponding differences in Rnp half-lives (Surbeck and Andrey 1999).

In order to study the time dependence of the decay product adsorption process, the same six patients were exposed to thermal water and radon vapour, respectively, for 10, 20 (the normal exposure time), 30, 40 and 60 min. Radon activities were measured on the forearm of each test person. Following each exposure, radon progeny decay curves were recorded over a period of 30 min. The time dependence of the adsorption process on the patients' skin is illustrated in Figs. 1 and 2. The figures demonstrate that ²¹⁸Po saturates after less than 20 min because of its half-life of 3.05 min. Due to the longer half-lives of the other progeny, steady-state is not yet reached after 1 h of exposure.

Table 3 compares Rnp activities on clean pig skin, greasy pig skin, and human skin treated with Finalgon®, after 20 min in thermal water (based on five measurements each). The low radon progeny activity on the pig skin suggests that biological mechanisms may play an important role in the adsorption process, while greasiness and blood circulation do not seem to be decisive factors.

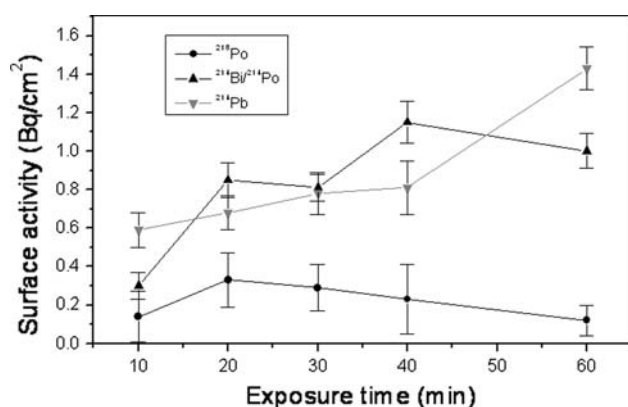


Fig. 1 Time dependence of radon progeny attachment onto the skin of the forearm in a thermal water bathtub (6 test persons: 3 females and 3 males)

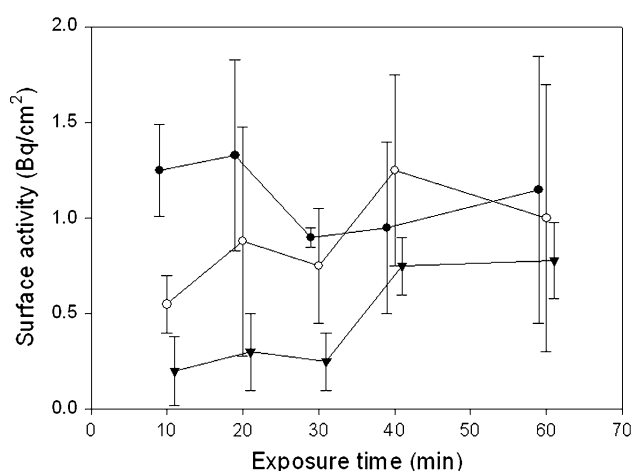


Fig. 2 Time dependence of radon progeny attachment onto skin of the forearm in a radon vapour bath (6 test persons: 3 females and 3 males). Radionuclides: ^{218}Po (filled circle), ^{214}Pb (open circle) and $^{214}\text{Bi}/^{214}\text{Po}$ (filled inverted triangle)

Transpiration at higher physical activities can increase the salt concentration in the thermal water. The influence of salt concentration on Rnp adsorption is plotted in Fig. 3. For comparison, one arm of a test person was exposed to water with different pH-values, while the second arm was exposed to pH-neutral thermal radon water. The figure clearly shows that adsorption of radon progeny is reduced in basic and acidic environments. Reduction in a basic environment can be explained by the formation of hydroxides, while the decrease in an acidic environment may be explained by a shift of the adsorption equilibrium from Rnp to protons both competing for surface binding sites (Ziechmann 1990).

To compare the salt concentrations in the therapy bath to the salt concentrations in the experiment, the conductivity as a direct measure of the ion concentration, measured in the thermal water 0.5 cm above the skin surface, was $499 \pm 12 \mu\text{S}/\text{cm}$ ($n = 12$). During a 20-min bath, this value increased only by about 2–5%. In the experiment with a NaCl concentration of 0.04 M, the conductivity measured was 4.5 mS/cm, while at 1 M it was 99.6 mS/cm. This suggests that there is an effect of the salt concentration in

Table 3 Radon progeny activities on various types of skin: clean pig skin, greasy pig skin, and human skin treated with Finalgon®; exposure time in thermal water: 20 min

Nuclide	Surface activity (Bq/cm^2)		
	Clean pig skin	Greasy pig skin	Human skin (Finalgon)
^{218}Po	0.07 ± 0.04	0.89 ± 0.25	0.98 ± 0.11
^{214}Pb	0.14 ± 0.10	0.91 ± 0.19	0.91 ± 0.22
$^{214}\text{Bi}/^{214}\text{Po}$	0.13 ± 0.18	1.11 ± 0.25	1.20 ± 0.33

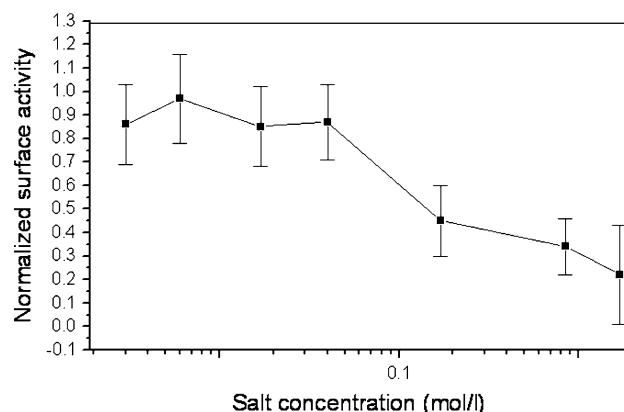


Fig. 3 Radon progeny attachment onto skin of the forearm in a thermal water bathtub as a function of NaCl concentration, normalized to the deposited activity in the untreated thermal water (6 test persons: 3 females and 3 males). Duration of exposure: 30 min

water on deposition of radon progeny, but that this effect is negligible at salt concentrations typically for therapy baths.

The dependence of Rnp adsorption on pH-value is plotted in Fig. 4. For comparison, one arm of a test person was exposed to water with different pH-values, while the second arm was exposed to pH-neutral thermal radon water. The figure clearly shows that adsorption of radon progeny is reduced in basic and acidic environments. Reduction in a basic environment can be explained by the formation of hydroxides, while the decrease in an acidic environment may be explained by a shift of the adsorption equilibrium from Rnp to protons both competing for surface binding sites (Ziechmann 1990).

Depth distribution of radon progeny in the upper skin layers

The vertical Rnp activity distribution relative to the untreated skin is shown in Figs. 5 and 6, after exposure to thermal water and radon vapour, respectively. The observed exponential decrease of the Rnp activity with depth for both treatment regimes may be caused by diffusion and transport through microcrevices and/or along hair capillary tubes. Fitting the measured depth profiles by

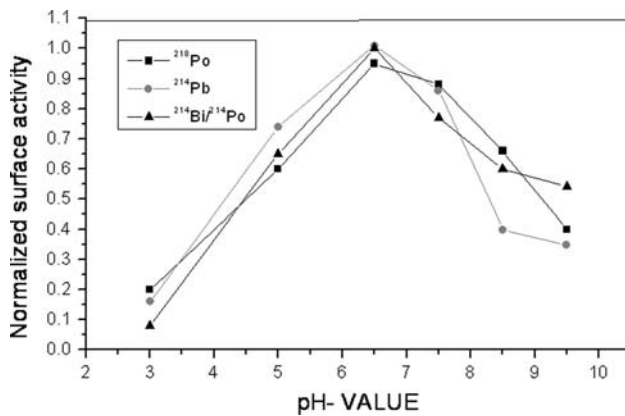


Fig. 4 Radon progeny attachment onto skin of the forearm in a thermal water bathtub as a function of pH-value, normalized to the radionuclide activities in the untreated thermal water (6 test persons: 3 females and 3 males). Duration of exposure: 30 min

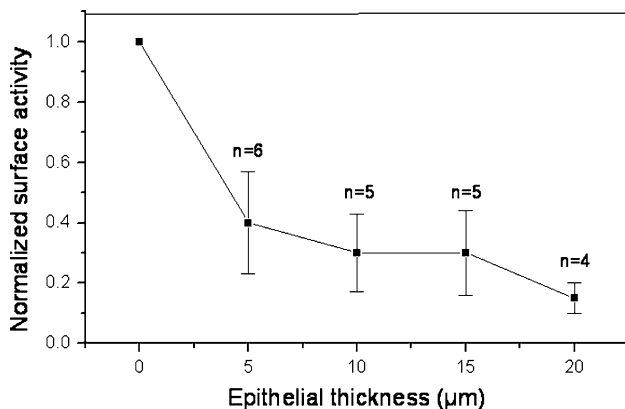


Fig. 5 Depth distribution of radon progeny in the upper layers of the skin after 20 min of exposure to thermal radon water, normalized to the activity on the surface of the skin

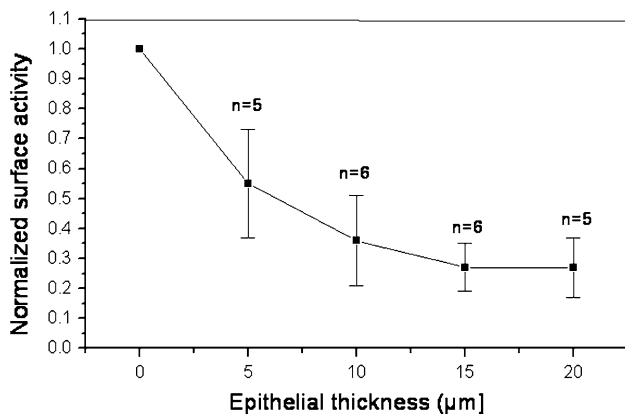


Fig. 6 Depth distribution of radon progeny in the upper layers of the skin after 20 min of exposure to radon vapour, normalized to the activity on the surface of the skin

exponential functions resulted in half-value depths of $6.8 \pm 1.7 \mu\text{m}$ in the thermal bath and 8.1 ± 1.3 in the vapour bath, respectively.

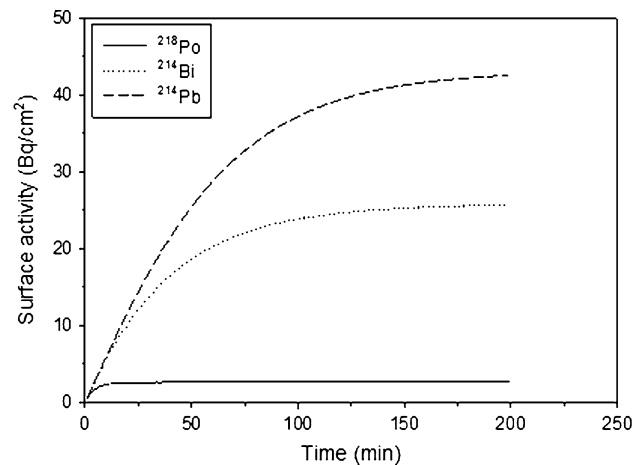


Fig. 7 Theoretical time dependence of the Rnp activity concentration on the skin (Bq/cm^2) during a 20-min radon exposure in the thermal bath, assuming equal flux ϕ_i for all Rnp from water onto the skin ($\phi_i = 0.01 \text{ Bq}/(\text{cm}^2 \text{ s})$)

Skin dosimetry

The theoretical time dependence of the Rnp activity on the skin surface (see Eq. 1) is plotted in Fig. 7, based on equal Rnp activity concentrations of $1 \times 10^{-2} \text{ Bq}/\text{cm}^3$ and equal attachment velocities of $1 \text{ cm}/\text{s}$, producing a flux ϕ_i of $0.01 \text{ Bq}/(\text{cm}^2 \text{ s})$. These values were selected for illustrative purposes, to demonstrate the theoretical dependence of radon progeny activities with time.

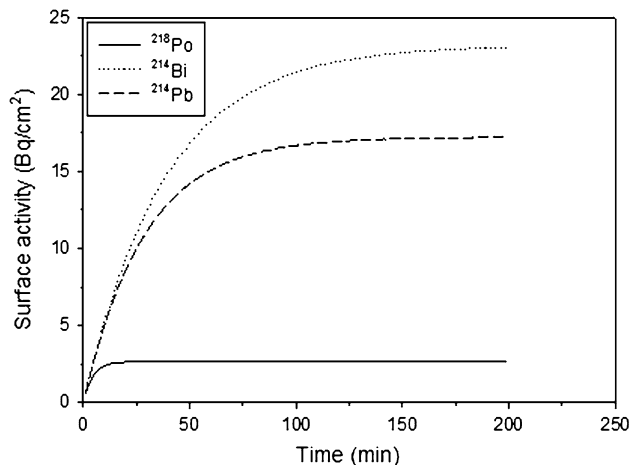
Based on the Rnp surface activities measured immediately after 20-min radon exposure in the thermal bath (see Table 1: ^{218}Po : $0.35 \text{ Bq}/\text{cm}^2$, ^{214}Pb : $0.86 \text{ Bq}/\text{cm}^2$, $^{214}\text{Bi}/^{214}\text{Po}$: $0.68 \text{ Bq}/\text{cm}^2$), the radionuclide flux, ϕ_i , defined as the product of attachment velocity (cm/s) and the Rnp-concentration (Bq/cm^3), $\phi_{\text{Rnp},i} = v_{\text{Rnp},i} C_{\text{Rnp},i}$, was computed using Eq. 1 and data in Table 1. Within the statistical uncertainties, the calculated deposition fluxes for the three nuclides are indeed relatively similar to each other. The resulting total activity deposited during the exposure serves then as the source term for the subsequent dose calculations (see Table 4).

For the dose calculations it must be considered that, for example, ^{218}Po deposited on the skin during the bathing phase will only deliver the emitted α -energy of 6.8 MeV and not the total potential alpha energy concentration (PAEC), whereas after the bathing phase the total PAEC will be deposited, as no desorption occurs any longer.

The theoretical time dependence of the Rnp activity concentration on the skin surface during a 20-min radon exposure in a vapour bath assuming equal flux ($\phi_i = 0.01 \text{ Bq}/(\text{cm}^2 \text{ s})$) for all Rnp from ambient air onto the skin is plotted in Fig. 8. As ^{218}Po contributes to ^{214}Pb and $^{214}\text{Bi}/^{214}\text{Po}$, and ^{214}Pb to $^{214}\text{Bi}/^{214}\text{Po}$, the activities of ^{214}Pb and $^{214}\text{Bi}/^{214}\text{Po}$ are larger than those calculated for

Table 4 Measured Rnp activities on the skin, resulting deposition fluxes and total activities deposited during an exposure time of 20 min in thermal water

Nuclide	Initial activity on skin after bathing (Bq/cm ²)	Flux onto skin Bq/(cm ² s) $\times 10^{-3}$	Total deposited activity during exposure (Bq/cm ²)
²¹⁸ Po	0.29	1.11	1.33
²¹⁴ Pb	1.09	1.16	1.40
²¹⁴ Bi/ ²¹⁴ Po	0.93	1.07	1.29

**Fig. 8** Theoretical time dependence of the Rnp activity concentration on the skin (Bq/cm²) during a 20-min radon exposure in the vapour bath, assuming equal flux ϕ_i for all Rnp from ambient air onto the skin ($\phi_i = 0.01$ Bq/(cm² s))

the water bath. Based on the Rnp surface activities measured immediately after the radon exposure in the vapour bath (see Fig. 2—²¹⁸Po: 1.33 Bq/cm², ²¹⁴Pb: 0.95 Bq/cm², ²¹⁴Bi/²¹⁴Po: 0.33 Bq/cm²), the computed radionuclide flux and the total activity deposited during the exposure time of 20 min are compiled in Table 5. Note that the flux decreases from ²¹⁸Po to ²¹⁴Bi/²¹⁴Po by roughly two orders of magnitude. This behaviour reflects, on the one hand, the decreasing activity concentration in the ambient air, and, on the other hand, the growth of carrier aerosols Rnp are attached to, which drastically reduces their deposition velocity (Morawska and Jamriska 1996).

Dose to sensitive skin cells

Based on stopping power and alpha particle ranges in the skin (ICRU 1993), absorbed doses were calculated as a function of depth for ²¹⁸Po and ²¹⁴Po alpha particles emitted from the skin surface by using equations provided by Eatough and Henshaw (1992) and Eatough (1997). Considering the measured depth distributions of the radon progeny throughout the epidermis (Figs. 5, 6), the resulting absorbed doses as a function of depth are plotted in Figs. 9 (thermal water) and 10 (hot vapour), normalized to a surface activity of 1 Bq/cm² on the surface of the skin

(depth = 0). For the conversion of absorbed doses to equivalent doses, a radiation weighting factor of 20 was used (ICRP 2007).

The thickness of the skin, ranging from the skin surface to the bottom of the basal cell layer, depends on body location and varies from individual to individual. It includes the stratum corneum (a horny layer) and the epidermis (Zhen et al. 1999; Charles 2007a). For our dose calculations, the following skin model was adopted (Eatough 1997): 15 μ m for the stratum corneum and 40 μ m for the epidermis, resulting in a skin thickness of 55 μ m. In case of the thermal bath, microscopic analysis of eosin-stained cells indicated that the stratum corneum swells from about 15 to 25 μ m, leading to a total skin thickness of 65 μ m.

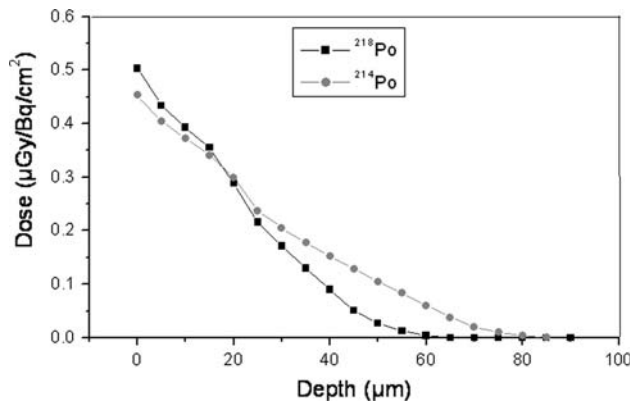
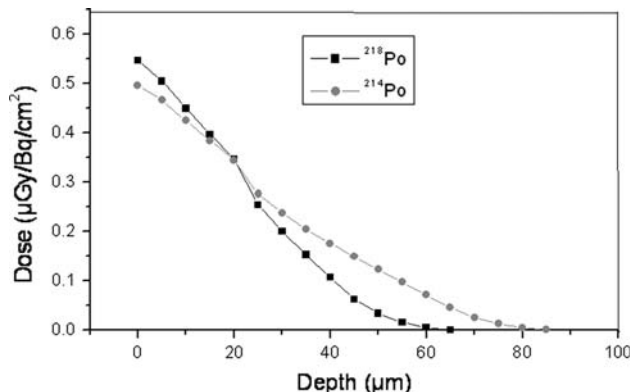
It is currently assumed that the Langerhans cells are the primary target cells for the stimulation of the immune system treatment of rheumatic diseases (Pratzel et al. 1999; Grom 2004; Flatscher 2006; Gaston et al. 2009). Since the Langerhans cells can be found everywhere in the epidermis, a uniform distribution was assumed for the dose calculations. The thickness of the epidermis indicates that these cells are located within the range of alpha particles emitted from the radon progeny. Because the concentration of the radon gas in the skin is much smaller than that of the Rnp, its contribution to the skin dose can safely be neglected.

Equivalent doses were calculated for the Langerhans cells, averaged over the whole thickness of the epidermis, as well as for the deeper lying basal cells, which are currently considered to be the most plausible target cells for the induction of skin carcinoma (Eatough 1997; Charles 2007a, b) (Table 6), for ten exposure sessions à 20 min as prescribed by the medical treatment plan for the thermal as well as the vapour bath. Although the highest doses are received by the horny cells located on the surface of the skin, it was assumed that these cells do not contribute to a therapeutic effect. The primary contributions to the cellular doses are produced by alpha particles emitted from ²¹⁸Po, because of their highest deposited activity and the emission of two alpha particles. Thus, the statistical errors of the doses listed in Table 6 are dominated by the statistical errors of the ²¹⁸Po surface activity measurements, estimated to be of the order of about 20–30%.

Radon levels and equilibrium factors in the thermal gallery are similar to those for the vapour bath (Falkenbach

Table 5 Measured Rnp activities on the skin and resulting deposition flux and total activity deposited during an exposure time of 20 min in the vapour bath

Nuclide	Initial activity on skin after bathing (Bq/cm ²)	Flux onto skin Bq/(cm ² s) × 10 ⁻³	Total deposited activity during exposure (Bq/cm ²)
²¹⁸ Po	0.85	3.25	3.90
²¹⁴ Pb	0.58	0.32	0.38
²¹⁴ Bi/ ²¹⁴ Po	0.23	0.08	0.10

**Fig. 9** Depth distribution of absorbed doses (in µGy) delivered by Rnp activities throughout the skin epithelium in the thermal water bath, normalized to 1 Bq/(cm² s) on the surface of the skin**Fig. 10** Depth distribution of absorbed doses (in µGy) delivered by Rnp activities throughout the skin epithelium in the vapour bath, normalized to 1 Bq/(cm² s) on the surface of the skin**Table 6** Average equivalent doses to Langerhans cells (epidermis), basal cells and horny cells on the surface of the skin for patients treated in the thermal bath, vapour bath and the thermal gallery during 10 exposure sessions

Treatment	Equivalent dose (mSv)		
	Skin surface	Epidermis	Basal cells
Thermal bath	0.48	0.12	0.03
Vapour bath	0.86	0.33	0.12
Thermal gallery	2.07	0.80	0.28

et al. 2002; Köstinger 2006). Hence, corresponding doses for 10 exposure sessions were estimated on the basis of the surface activity measurements in the vapour bath, considering that the exposure time in the gallery is 60 min, the average radon concentration is 45 Bq/l and the equilibrium factor $F = 0.4$ (Köstinger 2006). Because of the longer exposure times and thus higher deposited activities, but lower radon concentration, doses calculated for the thermal gallery are slightly higher when compared to those calculated for the vapour bath.

Discussion

To explore the dosimetric significance of radon progeny adsorbed to the skin, Rnp activities on the skin of patients exposed to thermal water and hot vapour were measured by alpha spectrometry. Average total alpha activity on the patients' skin varied from 1.2 to 4.1 Bq/cm² in the bathtub, and from 1.1 to 2.6 Bq/cm² in the vapour bath shortly after the end of the treatment. Extrapolating the vapour bath conditions to the thermal gallery and considering differences in exposure time and radon concentration, Rnp activities were estimated to range from 1.6 to 3.9 Bq/cm² in the thermal gallery.

Measurements of the Rnp activities on the skin of patients in a thermal bath were recently reported by Philipsborn (von Philipsborn 2006) for the radon spa Sybiltenbad, Germany, who measured the total alpha activity with a scintillation detector. The measured total alpha activity of about 1.1 Bq/cm² after 20 min of exposure is about a factor of 2 smaller than our values obtained for the thermal bath in Bad Hofgastein, which may be explained by differences in exposure conditions and uncertainties of the measurement techniques.

Results of measurements of total Rnp activities (in counts per min) on the skin of patients performed with a Geiger-Müller counter 25 min after exposure were published by Falkenbach et al. (2002) for the thermal gallery in Bad Gastein. Assuming that 1 Bq of total alpha activity corresponds to about 6 counts per min detected by the Geiger-Müller counter (Falkenbach et al. 2002), and an effective detector area of 16 cm², total Rnp activities for paraumbilical and paravertebral surfaces ranged from 2.6

to 4.7 Bq/cm², very similar to our extrapolated values of 1.6–3.9 Bq/cm². This agreement also indicates that our assumptions for the extrapolation of surface activities from the vapour bath to the thermal gallery were indeed reasonable.

Paatero (2000) reported a radon progeny deposition of 264 kBq/m² (or 26.4 Bq/cm²) due to atmospheric washout, which is about an order of magnitude greater than the measured surface activities in radon therapy. However, due to the much shorter exposure period in radon therapy (about 3 h when compared to about 5 months), resulting basal cell doses are about two orders of magnitude smaller than those incurred from atmospheric washout.

In the past, two hypotheses have been discussed to explain the therapeutic effects observed after exposure to radon and radon progeny: (1) a direct radiation effect in specific organs of the human body, e.g. regeneration of damaged tissue in the affected organ through incorporation of radon via skin or the lungs, and, (2) an indirect radiation effect such as an induced immune response through the irradiation of the deeper layers of the skin (epidermis) by radon progeny activities deposited on the skin surface. In Table 7, the doses to the skin incurred in the three exposure scenarios discussed in the present study are compared to kidney doses which are representative of doses to internal organs as they hardly differ from each other (Pohl 1979). Doses to internal organs following the exposure to radon and its progeny were computed by applying organ-specific dose-exposure conversion factors proposed by Pohl and Pohl-Rüling (1977) and Pohl (1979).

In contrast to the doses to the internal organs, represented here by the kidney doses, which are assumed to be responsible for the observed therapeutic effects in radon therapy, doses to basal and secretory cells in the bronchial epithelium may lead to lung cancer induction and thus represent an adverse health effect. For the calculation of doses to the kidneys and the bronchial epithelium, the following radon concentrations in ambient air were assumed: 3 kBq/m³ in the thermal bath, 15 kBq/m³ in the vapour bath, and 45 kBq/m³ in the thermal gallery (Tempfer et al. 2005).

Table 7 Equivalent doses to specific target organs, skin (epidermis), kidneys, and tracheobronchial (TB) region of the lungs, incurred in three different radon therapy treatment procedures

Treatment	Equivalent dose (mSv)			
	Skin	Kidneys	Lung (TB)	TEF
Thermal bath	0.12	0.04	0.13	0.92
Vapour bath	0.33	0.10	1.87	0.18
Thermal gallery	0.80	0.31	5.61	0.14

The therapeutic effectiveness factor (TEF) is defined as the ratio of skin to lung doses

While the thermal bath produces the lowest equivalent doses to the skin, the highest equivalent doses are incurred in the thermal gallery, with intermediate values for the vapour bath (Table 7). However, bronchial doses in the thermal gallery are also higher by nearly two orders of magnitude compared to those in the thermal bath. To quantify the relation of beneficial to detrimental effects, a therapeutic effectiveness factor was defined as the ratio of skin to bronchial equivalent doses (see Table 7). This factor is lowest for the thermal gallery and highest for the thermal bath, suggesting that the thermal bath may provide the preferable therapeutic treatment scenario.

In contrast to previous dose assessments, which practically neglected skin irradiation (Pohl 1979), the present study indicates that deposition of radon progeny onto the skin may produce a much higher dose to the skin than inhaled radon and progeny to typical internal organs such as the kidneys and thus may play an important role in the therapeutic response for both treatment schemes. While our investigations do not attempt to explain the cellular mechanisms leading to an observable therapeutic effect nor their dependence on dose, they provide the indispensable dosimetric basis for evaluating the relative significance of the different exposure scenarios.

Beneficial effects of radon balneotherapy have been documented for the treatment of various rheumatic diseases, such as ankylosing spondylitis, by stimulation of the immune system. However, in case of the endocrine system, Nagy et al. (2009) failed to demonstrate any substantial effect of thermal water with relatively low radon concentrations. While the interpretation of physiological and molecular mechanisms following exposure to radon progeny is beyond the scope of the present paper, it should be noted that the radon concentrations in the study of Nagy et al. (2009) were about an order of magnitude lower than those in the thermal baths in Gastein (80 vs. 950 Bq/l).

In the present study, equivalent doses were calculated for the Langerhans cells which were assumed to be uniformly distributed in the epidermis. If other potentially sensitive cells in the epidermis also contribute to the immune response, these cells will receive the same equivalent dose as the Langerhans cells. In addition, various types of lymphocytes in peripheral blood vessels may also exhibit an immune reaction, if reached by alpha particles (Tiepol et al. 2002). Moreover, recent in vitro experiments (Mitchell et al. 2004) and resulting model calculations (Fakir et al. 2009) have demonstrated that bystander effects may play an important role for alpha irradiation at low doses, i.e. sensitive target cells may not be hit at all but still exhibit a response. Thus, from a dosimetric as well as radiobiological point of view, the determination of the relevant target cell type causing an immune response in the skin may become less relevant.

The absorbed dose to the whole epidermis, which is numerically equivalent to the absorbed dose to the Langerhans cells, may represent all these different aspects.

The radon concentrations in the vapour bath (15 kBq/m³) or the thermal gallery (45 kBq/m³) are up to three orders of magnitude higher than the annual median indoor radon concentration of 51 Bq/m³ in the province of Salzburg (Friedmann 2005). Thus, if based on the ratio of the radon concentrations and neglecting other exposure differences, the environmental equivalent doses to Langerhans and basal cells incurred during the therapeutic treatment are between two and three orders of magnitude smaller than the corresponding therapeutic doses. In terms of total annual equivalent doses, the therapeutic equivalent doses contribute about 10% to the total doses for a patient, while 90% are caused by the environmental equivalent doses. Given the observed variability of the thickness of the skin as well as variations of environmental radon levels, a therapeutic treatment in the Gastein radon spa facilities hardly plays a role for radiation protection considerations.

Deposition of radon progeny on skin surfaces may also give rise to skin tumours (Charles 2007a, b). However, the rather low doses received by the basal cells, which are currently presumed to be the relevant target cells for the induction of skin carcinoma, suggest that the induction of skin cancer can safely be neglected for the spa patients.

With respect to lung cancer, which is the primary biological effect for radon progeny inhalation, it is important to emphasize that bronchial doses lie well within the natural fluctuations of the doses received by the general population, thus causing no additional hazard to the patients undergoing such treatment. For example, the annual effective bronchial dose to the population living in Salzburg City amounts to 2.8 mSv (Pohl 1979).

The reported beneficial therapeutic response in radon therapy has been attributed primarily to radiation doses produced by inhaled radon and radon progeny deposited on skin surfaces. However, from a radiobiological point of view, it still remains an open question, how such small doses, which are even too small to induce skin tumours, are able to produce the therapeutic effects observed in clinical studies. Thus, high humidity and temperatures in the vapour bath and the thermal gallery, and the mineral and ion content of the thermal water in the water bath may also contribute to the reported therapeutic response. In addition, the specific conditions during a spa treatment in the Gastein valley, such as altitude (about 1,000 m above sea level), clean mountain air, controlled diet, physical exercise (underwater therapy, walking), may also beneficially affect the health status of the patients. At present, however, there are no clinical and molecular studies available, which would allow a differentiation between these various factors. The fact that the equivalent epidermal doses incurred

in the different treatment schedules are below 1 mSv suggests that radiation alone may not be the sole therapeutic factor but that all these factors may act together, possibly in a synergistic fashion.

Acknowledgments This research was supported in part by the Research Institute Gastein through projects FPK 109 and 110, by a project of the FOI Gastein 2009, the Austrian Research Center Seibersdorf (HT, AS) and the Socrates exchange program between the University of Salzburg and Babes-Bolyai University, Cluj-Napoca (AD). The authors further thank Dr. W. Foisner (Bad Hofgastein) and Dr. J. Untner (Bad Gastein) for providing the patients and medical advice, and Dr. Ferdinand Steger from the Austrian Research Center Seibersdorf for providing PhD scholarships (HT, AS).

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