Radon parameters in outdoor air

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Abstract. For dose estimation by inhalation of the short lived radon progeny in outdoor air, the equilibrium factor (F), the unattached fraction (f_p), and the activity size distribution of the radon progeny were measured. Besides the radon parameter the meteorological parameter like temperature, wind speed, and rainfall intensity were registered. The measurements were carried out continuously for several weeks to find out the variation with time (day/night) and for different weather conditions. The radon gas, the unattached and aerosol-attached radon progenies were measured with an monitor developed for continuous measurements in outdoor air with low activity concentrations. For the determination of the activity size distribution a low pressure online alpha cascade impactor was used. The measured values of the equilibrium factor varied between 0.5–0.8 depending on weather conditions and time of the day. For high pressure weather conditions a diurnal variation of the F-factor was obtained. A lower average value (F = 0.25) was registered during rainy days. The obtained f_p-values varied between 0.04 and 0.12. They were higher than expected. The measured activity size distribution of the radon progeny averaged over a measurement period of three weeks can be approximated by a sum of three log-normal distributions. The greatest activity fraction is adsorbed on aerosol particles in the accumulation size range (100–1000 nm) with activity median diameters and geometric standard deviation values between 250–450 nm and 1.5–3.0, respectively. The activity median diameter of this accummulation mode in outdoor air was significantly greater than in indoor air (150–250 nm). An influence of the weather conditions on the activity of the accumulation particles was not significant. In contrast to the results of measurements in houses a small but significant fraction of the radon progeny (average value: 2%) is attached on coarse particles (>1000 nm). This fraction varied between 0–10%. 20%–40% of the activity is attached on particles in the nucleation size range (<100 nm). A diurnal variation was registered with a higher activity fraction during the day time caused by the higher particle number concentration of the atmosphere during the day hours.

Introduction

Not the radon gas ^{222}Rn but its short lived decay products ^{218}Po, ^{214}Pb, ^{214}Bi/ ^{214}Po, in the breathing air cause the greatest fraction of natural radiation dose of the human. The dominant parameter related to dose is the activity size distribution of the decay products in the air because the original deposition destination and amount of the inhaled activity deposited in lung depend on the particle size. For practical reasons related to measurements instead of the activity size distribution the characteristics

- equilibrium factor (F-factor),
- unattached fraction (f_p-factor),
- relative activity size distribution

for dose estimation were used.

The concentration of radon and its progeny in the atmosphere depends on the place, time, height above the ground, and meteorological conditions. The spread of radon in the atmosphere after their exhalation from the ground is mainly caused by turbulent diffusion and is limited only by the radioactive decay. In contrast to radon, the distribution of the decay products is also influenced by the removal into the earth's surface.

Jacobi and André [1] estimated the vertical airborne activity concentration of radon and its short lived progeny in model calculations. The results of such model calculations show the great variation of the radon concentration at ground level (about a factor 100) for different meteorological conditions (Table 1). Several investigators measured such great variations of the radon concentration and its progeny (e.g. [2] [3] [4] [5] [6] [7]) as predicted by model calculations.

Table 1. Radon concentration (C_o) and the equilibrium factor (F) in 1 m above the ground for different mixing conditions of the atmosphere after model calculations of Jacobi and André [1]. Assumed exhalation rate: 0.021 Bq m^{-2} s^{-1}

MIXING CONDITION	**^{222}Rn C_o [Bq/m^3]**	**EQUILIBRIUM FACTOR F**
INVERSION (INN)	103	0.71
WEAK MIXING (WNN)	14.5	0.54
NORMAL MIXING (NNN)	4.0	0.76
STRONG MIXING (SSN)	1.5	0.85

Table 2. Estimation of the unattached fraction (f_p), taking into account the attachment rate (X), obtained from measurements in the open air [8]

PARTICLE CONCENTRATION Z [x10^4 cm^{-3}]	ATTACHMENT RATE X [h^{-1}]	UNATTACHED FRACTION f_p
0.5	10	0.13
1	20	0.08
2.5	50	0.04
5	100	0.02

Theoretical considerations show, the dominant parameter, which influences the unattached radon progeny is the attachment rate to the atmospheric aerosol. The values of the unattached fraction of the free atmosphere can be estimated taking into account the attachment rate (X) as function of the aerosol particle concentration (Z) obtained from measurements with outdoor air [8]. After that a variation of the particle concentration outdoors between $1*10^4$ and $5*10^4$ particles/cm^3 corresponds unattached fractions between 8% and 2% (Table 2).

Simultaneous measurements of radon and its progeny for the determination of the F-factor and the measurement of unattached fraction (f_p) over a longer time period are scarce in the literature. Porstendörfer [9] reported a mean F-factor of about 0.6, with a variation of between 0.4–0.8 in a height of 1.5 m above the ground, and a mean value of the unattached fraction of about 2% [10].

The activity size distribution of the progeny aerosol C(d) and the number size distribution Z(d) of the aerosol are different, because the attachment probability ß(d) is a function of particle diameter d. The correlation between both size distributions can be expressed by [11]

$$C(d) = \frac{C}{X}\,\beta(d)\,Z(d)$$

C is the activity concentration and X is the attachment rate, which characterise the adsorption velocity of the decay product to the atmospheric aerosol. The equation above describes the activity size distribution for all four short lived decay products attached on aerosol particles, because the differences between the individual size distributions are small [12].

Only few data are available on the activity size distribution of radon progeny aerosol in the free atmosphere ([13] [14] [15] [16]) because of the difficulties encountered in measuring aerosol particles in a broad diameter range from 10 nm up to 10000 nm and at low activity concentrations outdoors.

The first direct measurements ([13] [14] [15] [17]) of activity size distributions could best be approximated by one lognormal size distribution with an activity median diameter (AMD) between 100–400 nm and geometric standard deviations between 1.5 and 3.0. The measurements of Becker et al. [14], Reineking et al. [16], and Zock [18] showed, that the AMD values of the outdoor air (300–400 nm) are significant greater than the values obtained in low-ventilated (< 0.5 h^{-1}) rooms. In addition, Reineking et al. [16] and Zock [18] could show, that besides the accumulation mode with the greatest fraction of activity two further modes of the activity size distribution of the progeny exists. 10%–40% of the activity is attached on aerosol particles between 10 nm and 100 nm (nucleation mode) and 0–10% are adsorbed on particles > 1000 nm (coarse mode).

In the following the results of measurement campaigns over some weeks are reported. The aim was to find out the average values of F, f_p and the relative activity size distribution of the radon progeny in the free atmosphere and their variation with time (day/night) and for different weather conditions.

Measurement techniques

A monitor for measuring the concentrations of radon gas, the unattached and on aerosol attached radon decay products in air separately, simultaneously and continuously by α-spectroscopy has been developed.

Fig. 1 gives a schematic diagram of the monitor. The instrument is composed of a measuring head, an electronic-unit and a personal computer for data acquisition and analysis. The measuring head itself consists of three different units. An airflow of 600 l/h is sucked through the first two units by a pump and the unattached activities are sampled in the first unit on a screen and the attached ones in the second unit on a membrane filter. The number of α-decays of ^{218}Po and ^{214}Po-decays are measured with a surface barrier detector in front of the screen and another detector in front of the filter respectively. Furthermore the number of α-decays of ^{212}Po can be detected. With its amount the number of interfering ^{212}Bi-decays in comparison to the ^{218}Po-decays can be calculated.

After passing the first two units and the pump the flow of 600 l/h is divided in two single streams of 60 l/h and 540 l/h. The smaller one passes over an air drier ($CaCl_2$) into the third unit where the radon gas concentration is measured by the principle of electrostatic precipitation [19].

In all three units passivated implanted planar silicon (PIPS) detectors for α-spectroscopy are used. The signals of the detectors are feed in three preamplifier/amplifier combinations and a multi-channel analyser. The data are stored in form of an energy spectrum of 1024 channels and analysed with a personal computer.

For measuring the unattached radon progeny the on-screen measurement technique was chosen. The collected decay products on screen are measured directly by means of alpha-spectrometry. By selection of the right type and size of the screen and the flow rate the monitor was optimised to have a 50% penetration for particles with 3.5 nm diameters and low entrance losses (10%) for the unattached radon progeny clusters.

The disadvantage of the screen measurement technique is that not only the unattached but also a certain fraction on aerosol particles attached progeny were collected on screen. To avoid this error (for atmospheric aerosol outdoors up to 100%) all the obtained f_p-values were corrected as proposed by Reineking and Porstendörfer [10].

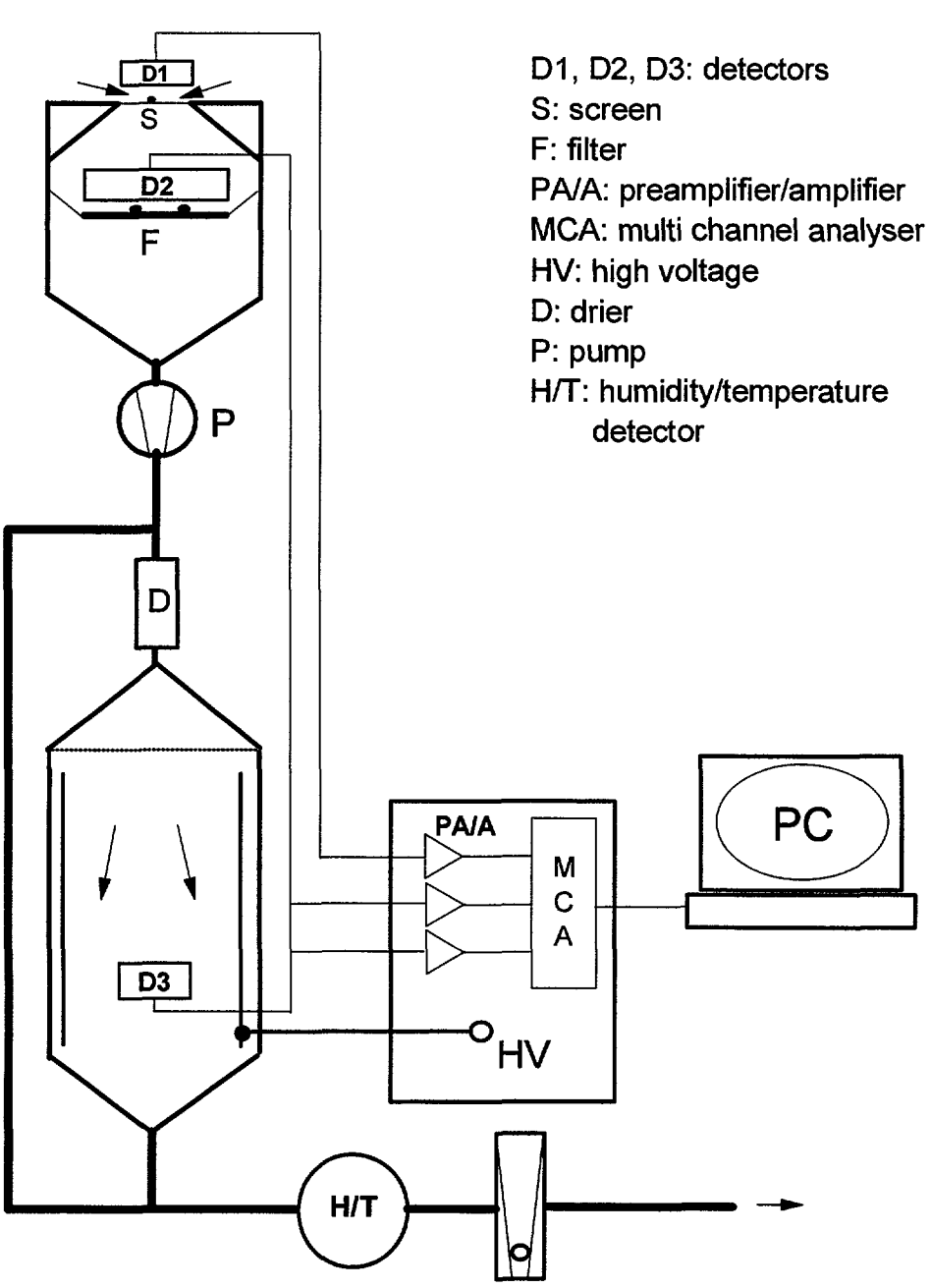

Fig. 1. Schematic diagram of the F-f_p-monitor.

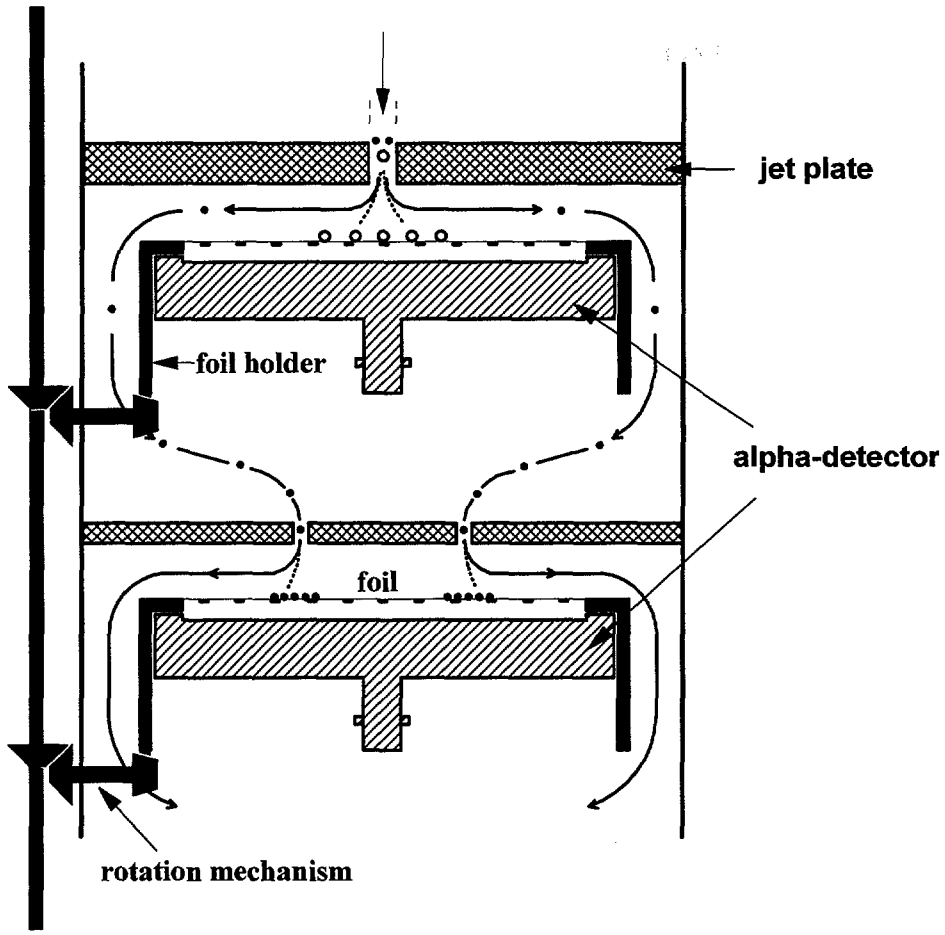

Fig. 2. The arrangement of jet plate collection foil and alpha detector of the online alpha cascade impactor.

The determination of the activity size distribution of the radon progeny was carried out with a low pressure online alpha cascade impactor (OLACI). This impactor was developed to measure continuously the size distribution of the radon progeny over longer time periods (1–2 weeks) [12]. The online impactor has nine stages with 50% cut-off diameters of 16000 nm, 8082 nm, 4242 nm, 2356 nm, 1135 nm, 589 nm, 292 nm, 150 nm and 60 nm. Particle sizes < 60 nm were collected and measured on a backup filter.

The aerosol particles of every impactor stage were collected on a thin aluminium-foil (4 μm) placed above an alpha detector (Passivated Implanted Planar Silicon (PIPS), active surface 2000 mm^2). Therefore the individual radon progeny can be measured by alpha-spectroscopy during air sampling. A rotating mechanism allows a homogenous activity and dust distribution on the foil (Fig. 2). In spite of the energy losses of the alpha particles in air and the collection foil, which cause an alpha spectra with an increased tailing, the energy separation between ^{218}Po and ^{214}Po was satisfactory. A tailing correction was made during data evaluation. The air sampling rate of 5.1 m^3/h made it possible to measure the activity size distribution also at low activity concentrations (1 Bq/m^3). For measurement of the size distribution of all three short lived radon decay products the alpha-counting during and after air sampling was carried out.

Besides the activity measurements the meteorological parameters like temperature, wind speed, and rainfall intensity were registered.

Results

The measurement campaigns were carried out in a suburban area of Göttingen. Each measurement campaign lasted for 2–3 weeks. The activity concentrations, the activity size distribution of the radon progeny, and the meteorological parameters like temperature, rain rate and wind velocity were measured continuously. For each quantity the results of only one measurement campaign are presented and discussed in the paper.

In Fig. 3 and Fig. 4 the measured values of radon, F and f_p as function of time are presented. The measurement period can be divided in two parts with different weather conditions. Some days with sunny weather and lower turbulent mixing during the night/morning hours follow cloudy days with higher turbulences during the whole day. This two different weather conditions are typically characterised by the diurnal variation of the radon concentration with higher activity concentration during the night/morning hours of a day (Fig. 4) [7] [9].

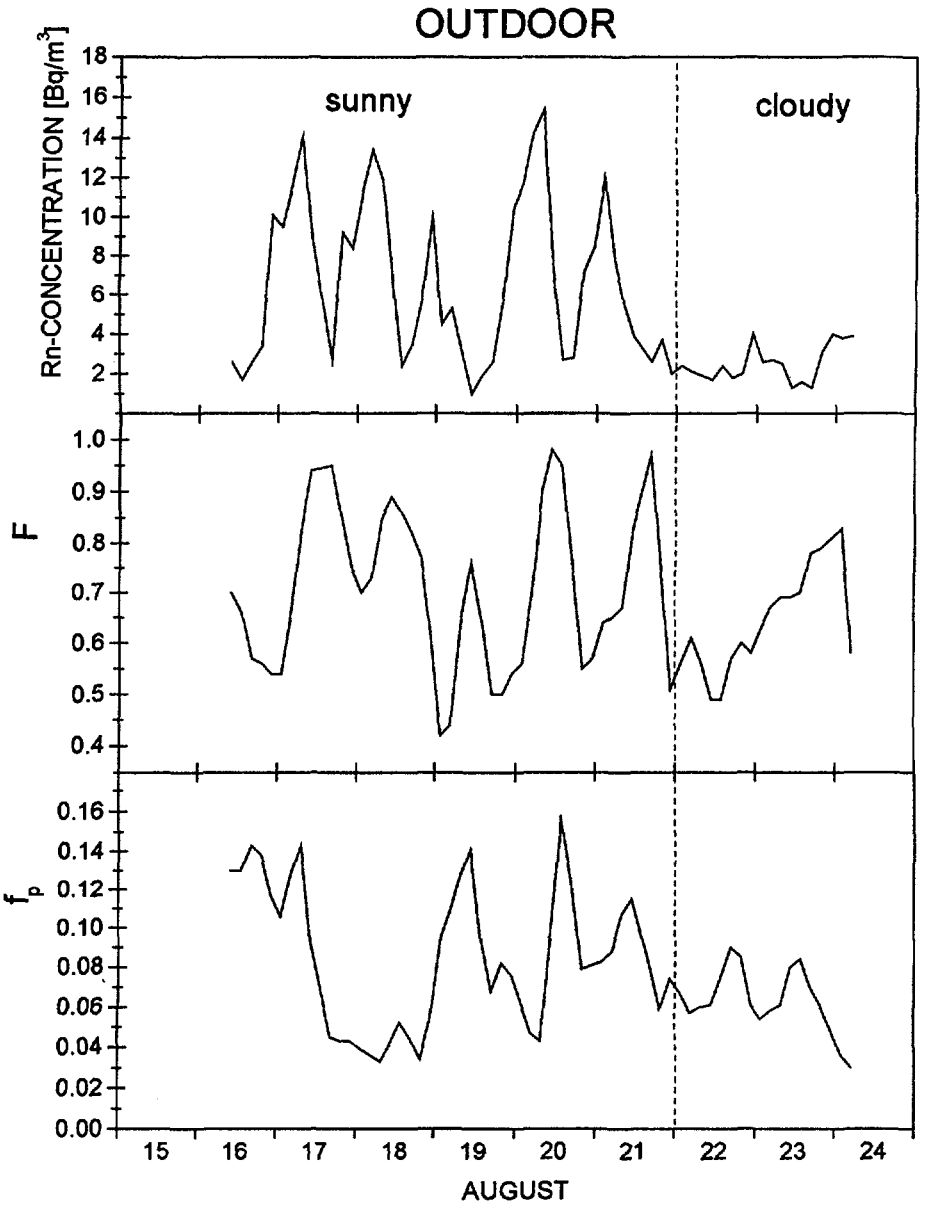

Fig. 3. Radon concentration, equilibrium factor (F), and the unattached fraction (f_p) in the atmosphere near the institute building 1 m above the ground.

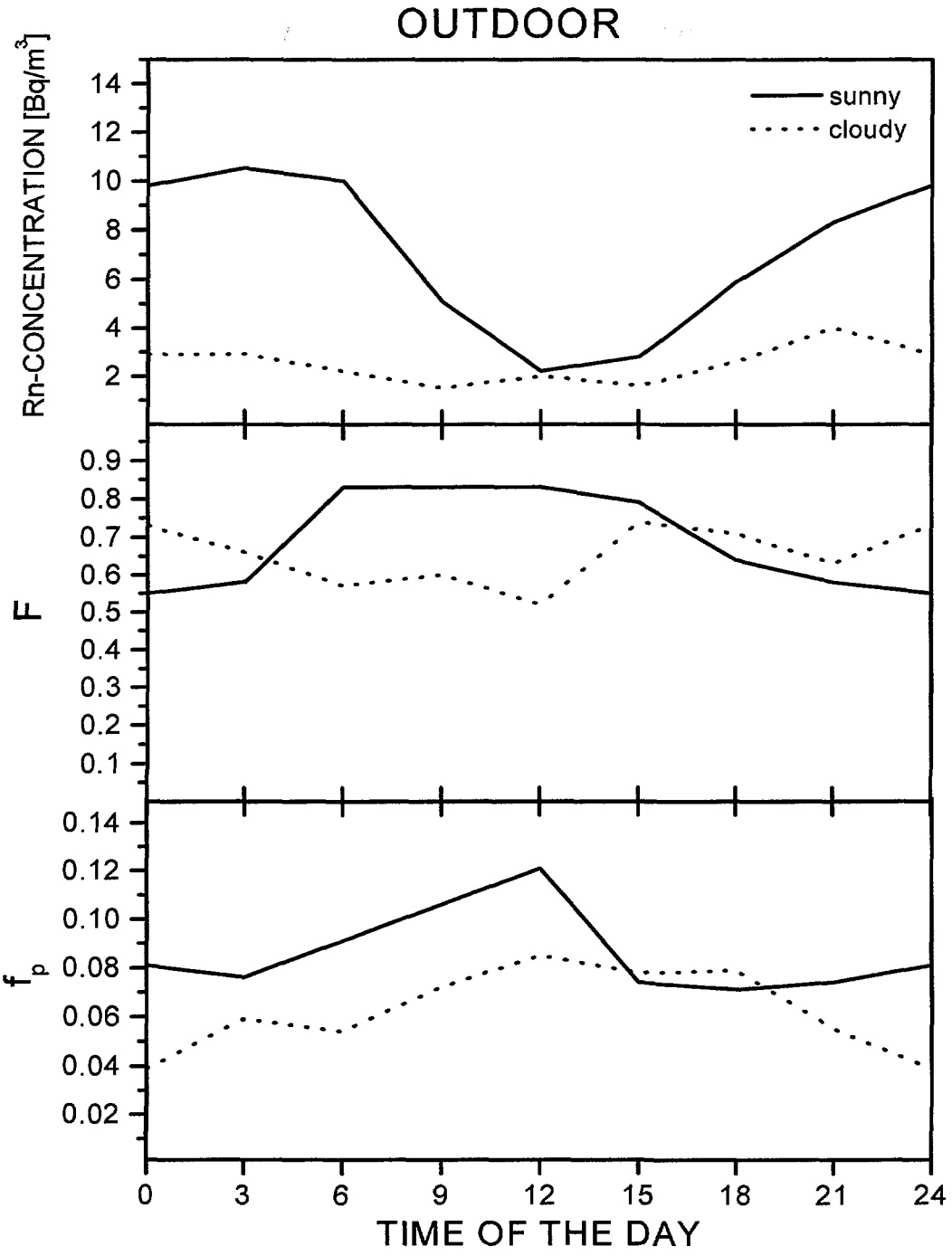

Fig. 4. Diurnal variation of the radon concentration, equilibrium factor (F), and the unattached fraction (f_p) obtain from measurements in the atmosphere near the institute building 1 m above the ground.

The results of the F-value vary between 0.5 and 0.8 with higher values during the day hours of the sunny high pressure days.

All f_p-values obtained from the measurements are fairly high (0.04–0.12) compared to the results of former measurements [10]. The maximum of the value during the sunny day hours cannot be understood. Because of the higher particle number concentrations during this daytime a minimum of unattached fraction should be expected. More measurements are needed. Especially the variation of the aerosol particle concentration should be registered to help the explanation of the results.

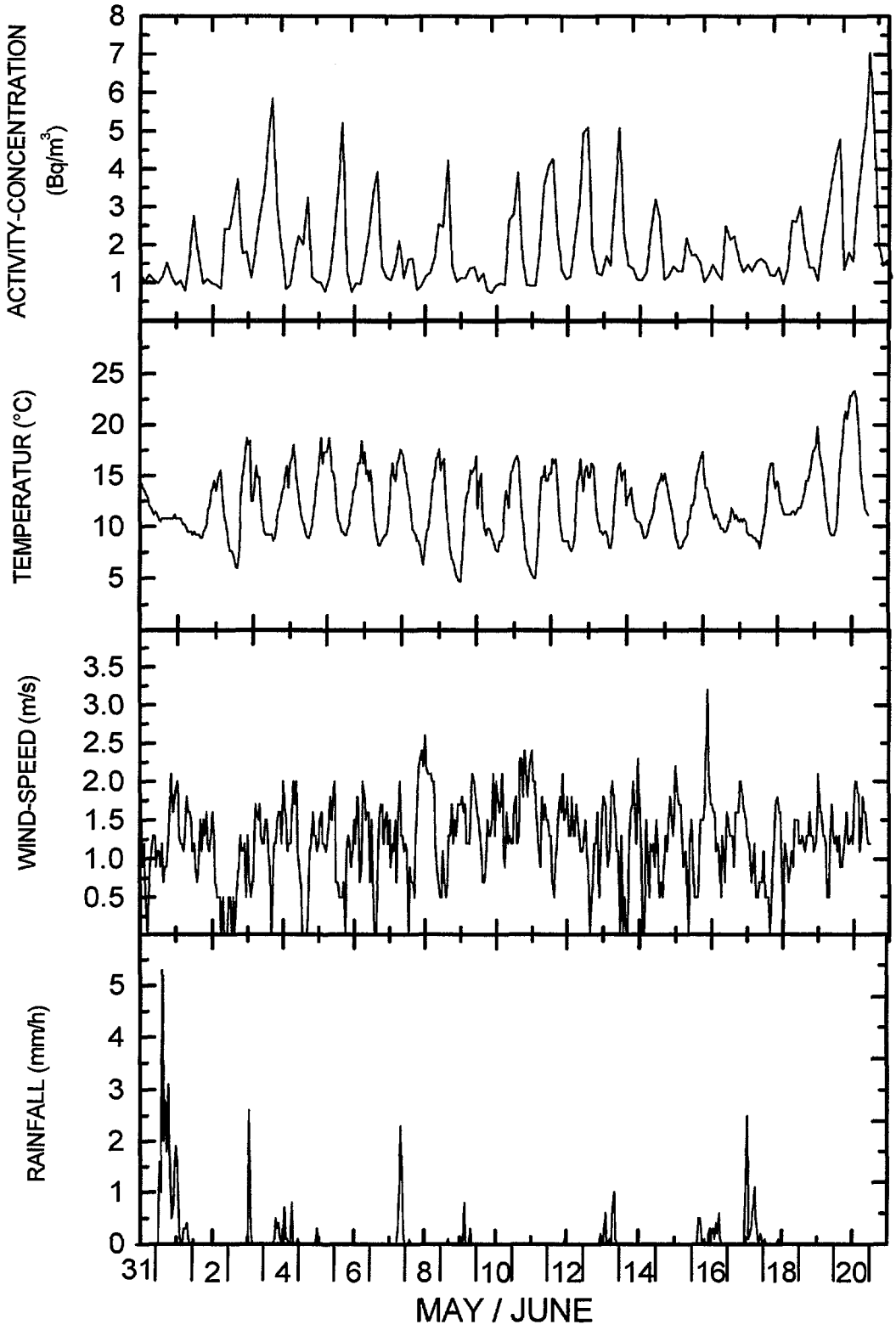

Fig. 5. The variation of the activity concentration, temperature, wind speed and rainfall intensity in the atmosphere during the measurement campaign.

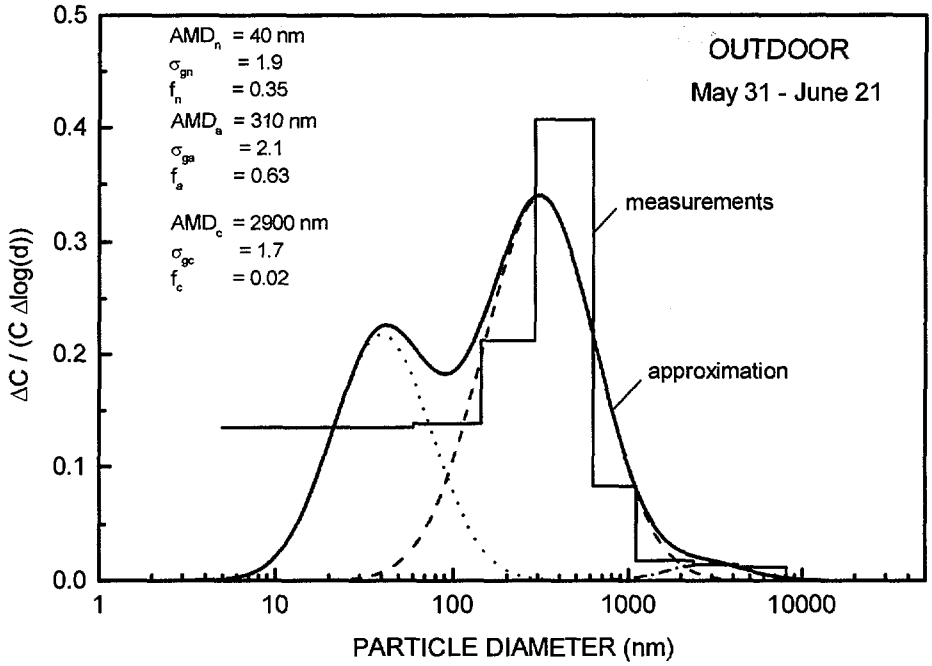

Fig. 6. The average activity size distribution of the radon progeny for the measurement campaign May 31–June 21.

The variation of the activity concentration of the ^{218}Po-aerosol and the meteorological parameters during the measurement campaign for determination the activity size distribution are presented in Fig. 5. The activity concentration of the progeny aerosol was obtained by the sum of the activities on all impactor stages and on the back up filter. Different weather conditions occur during the measurement period. The days with a high pressure weather are typically characterised by greater differences of temperature and activity concentration between day and night. In this case the night indicates a stable atmosphere with lower mixing and therefore higher activity concentration. During the rainy days there is higher mixing in the lower atmosphere, also during the night and morning hours. In this case no diurnal variation could be observed.

The relative activity size distribution of the radon progeny aerosol averaged over 22 days is presented in Fig. 6. The experimental data can be approximated by a sum of three lognormal distributions. Most of the activity (fraction f_a = 63%) is adsorbed on particles of the accumulation mode with an activity median diameter AMD$_a$ = 310 nm and a geometric standard deviation of σ_{ga} = 2.1. Remarkable is the fairly high amount of the activity on particles below 60 nm (collected on the backup filter). This activity fraction of the nucleation mode (f_n = 35%) can be approximated by an AMD$_n$ of about 40 nm. On average only 2% of the activity is adsorbed on coarse particles (>1000 nm) with an AMD$_c$ value of about 3000 nm. The fraction of the coarse particle mode mainly generated by resuspension and dispersion processes varied between 0–10%.

To see the variation of the activity size distribution during the measurement period, the AMD$_a$ value and the geometric standard deviation, σ_{ga}, of the accumulation mode and the activity fraction on the backup filter as function of time is presented in Fig. 7. The AMD$_a$ and the σ_{ga} values vary between 250–500 nm and 1.8–3.0, respectively, but a diurnal variation and an influence of the weather parameters were not significant.

A high activity fraction of 20%–45% (average 32%) on the backup filter (sizes <60 nm) was registered with the online alpha cascade impactor (Fig. 7). This activity fraction showed also a diurnal variation (Fig. 8). For the explanation some measurements of the number size distribution of the atmospheric aerosol with a differential electrostatic mobility analyser (DMA) and a laser aerosol spectrometer (LAS) were carried out. The results of these measurements showed a higher particle concentration with diameters <50 nm during the day hours. This is caused by the higher aerosol generation by the combustion processes and photochemical reactions during the daytime.

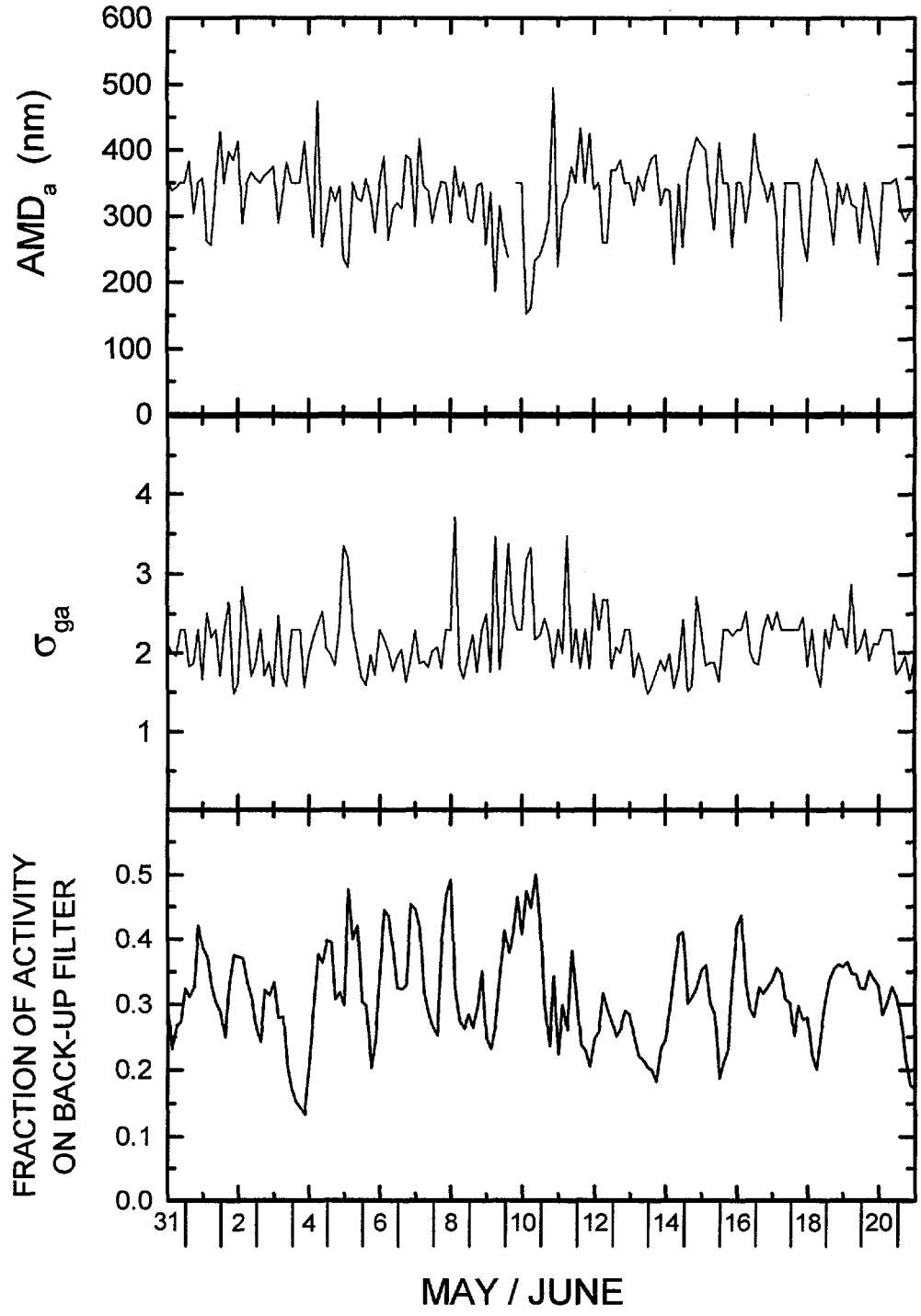

Fig. 7. The variation of the activity median diameter (AMD_a) and the geometric standard deviation (σ_{ga}) of the radon progeny in the accumulation size range and of the backup filter.

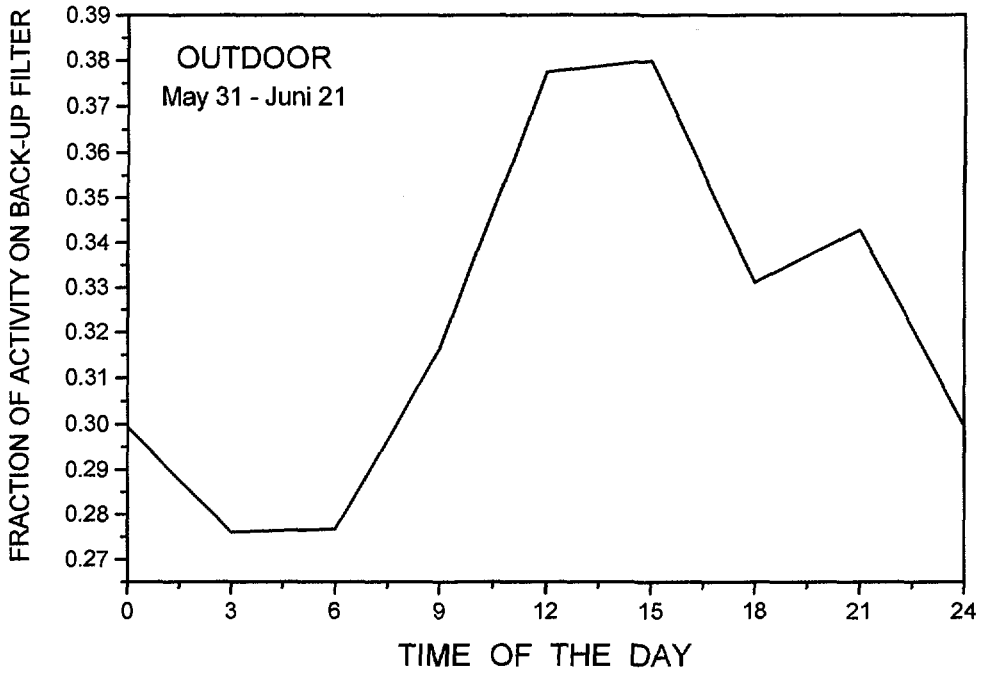

Fig. 8. The diurnal variation of the activity fraction on backup filter (<60 nm) averaged over the measurement campaign May 31–June 21.

Conclusion

The measured F-values vary between 0.5 and 0.8 in 1 m height above the ground depending on the daytime and the weather conditions. These are values which can be explained by model calculations.

The obtained f_p-values between 0.04 and 0.12 are higher than expected. In addition, the diurnal variation with a maximum during the sunny day hours cannot be explained.

The greatest activity fraction of the radon decay products is adsorbed on aerosol particles in the accumulation size range. The experimental data, approximated by a lognormal distribution give AMD_a and σ_{ga} values between 250–450 nm, and 1.5–3.0, respectively. This accumulation mode in outdoor air has significantly greater AMD_a values than in indoor air (AMD_a: 150–250 nm) in agreement with the results of former measurements [14] [16]. An influence of the weather conditions on the activity of the accumulation particles was not significant.

In contrary to the measurement values obtained in dwellings a small but significant fraction of the radon progenies (average value: 2%) is attached on coarse particles (>1000 nm). This fraction varied between 0 – 10% during the measurements.

The measurements with the online alpha cascade impactor yielt an activity fraction of 20%–40% in the nucleation size range (<100 nm). A diurnal variation was registered with a higher activity fraction during the daytime, caused by the higher aerosol particle concentration of the atmosphere during these day hours.

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