Experiments concerning Rn-measurement using HV sampling of Po-218

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Abstract

The aim of experiments was to check the influence of humidity and radon activity concentration on the calibration factor of a measuring chamber and to estimate rates of neutralisation of Po-218 ions as well. The chamber consists of a cylinder with 3.6 l volume. Experiments were carried out in flow through mode. For reference measurements a scintillation chamber (lucas chamber) was used. To change the humidity a $CaCl_2$ dryer column in bypass to the air stream was used. It was found that the calibration factor increased significantly with increasing humidity as suggested. The increase of calibration factor depends of activity concentration. In the range from dry air up to absolute humidity of a.H. =10 g/kg (dry air) the calibration factor changes of about factor 3.5 at low activity concentrations (< 2.5 Bq/l) and about factor 1.9 for higher activity concentrations.

At low absolute humidity (< 2.5 g/kg) no influence of the Rn-222-activity concentration on calibration factor was found.

Additionally the measured results were taken to calculate neutralisation rates of Po-218-ions. Using a spherical model of the electrical field the neutralisation rates were calculated for low and high humidity. Values of neutralisation rate **n** of about **n**= 5 s^{-1} for dry air and about **n**= 20 s^{-1} for an absolute humidity of about 10 g/kg (dry air) was found.

1 Introduction

HV based Rn-measurement is a common method used in a lot of instruments. Hereby Po-218-ions formed by Rn-decay are driven by an electric field onto the surface of a detector, sensitive for alpha radiation. The disadvantage of this method is the dependence of measuring results from climatic conditions, especially humidity of air. Additionally there is a dependence of calibration factor from different trace gases. This fact is well known. The influence of humidity and some gases like NOx or organic compounds on neutralisation rate of Po-218 is the reason for the dependencies. Chu/Hopke {1} published detailed investigations about the mechanism of neutralisation and found quantitative relations between gas concentrations and neutralisation rates. Hopke {2} used this results for practical advises for the construction of measuring chambers. The maximal distances of Po-218 ions between the formation by Rn decay and the neutralisation in dependence of gas concentrations and electric field strength were published in this paper. The numerical relation between air humidity and neutralisation rate was found to be

$$n = 1.14 \cdot \sqrt{[H_2O]} \tag{1}$$

were:

n: neutralisation rate s^{-1} and

$[H_2O]$: water vapour concentration in ppm.

Equation (1) is valid for concentrations < 1800 ppm or about < 1.12 g/kg (dry air). Assuming a temperature of 20 degrees Celsius this value corresponds with a relative humidity of 7.6 %. Most of natural occurring condition corresponds with higher humidity. Following Chu/Hopke {1} a constant neutralisation rate of **$n=47.9$ s^{-1}** is valid for this conditions.

Pagelsdorf/Porstendörfer {3} published neutralisation rates found by investigations done for estimation of ionised part of decay products in typical environmental conditions carried out in radon chamber of 12 m^3. Neutralization rates for different values of humidity and radon concentrations in a range of **0,011 s^{-1}** up to **0,083 s^{-1}** are published.

Because of this big discrepancy I carried out some own measurements to deduce values of the neutralisation rate using a measuring chamber in typical geometry and environmental air (soil air was used).

Moreover the dependency of calibration factor from humidity and radon activity concentration for the used experimental setup should be estimated.

2 Experimental setup

Figure 1: Scheme of the setup.

1: Measuring instrument for air flow
2: Air pump
3: Valves
4: air dryer column
5: Vessel
6: T-r.H. probe
7: Aerosol filter
8: Scintillation chamber
9: Measuring chamber
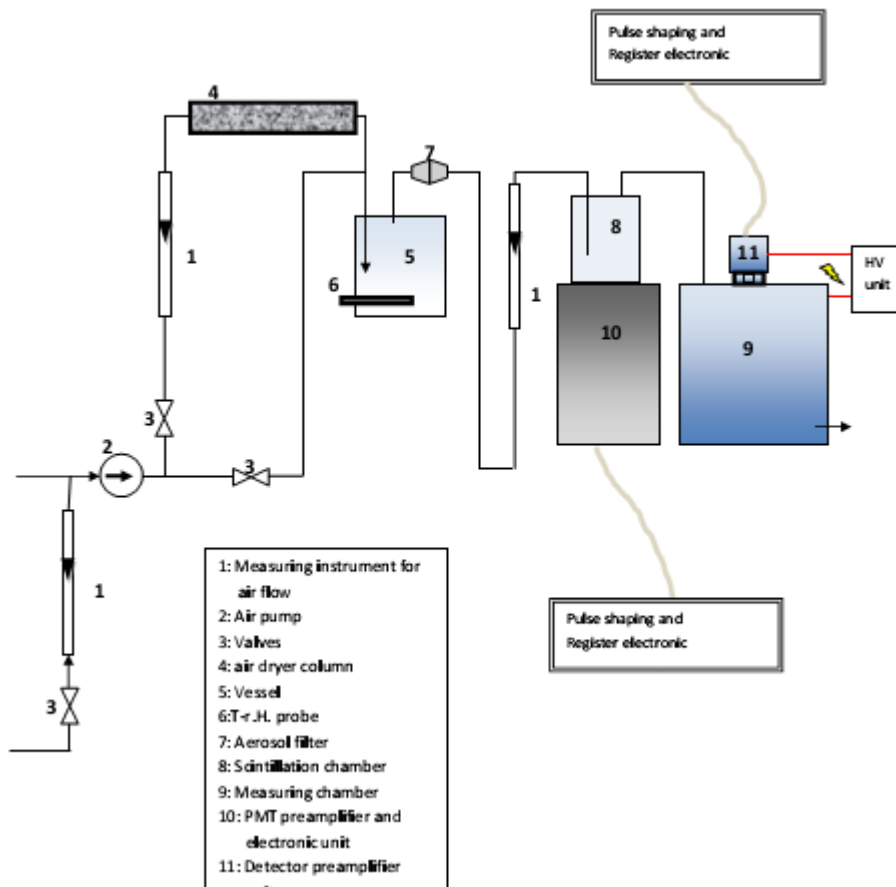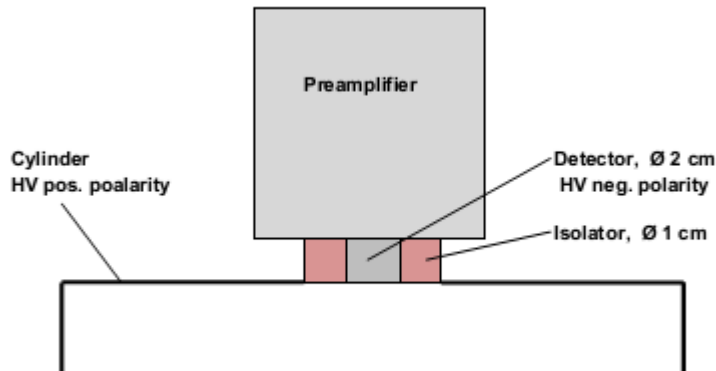10: PMT preamplifier and electronic unit
11: Detector preamplifier

Fig 2 top of the measuring chamber, detail of detector geometry

The air with radon content was pumped from a pipe located in soil because no other radon source was available. A disadvantage of this technique is the change of activity concentration during experiments. But using times of relative constant activity concentration this disadvantage can be minimized. The rel. humidity of soil gas was in the range of 80 -90 %. The humidity was reduced by a CaCl2 drying column according to Figure 1.

To realize small activity concentrations room air could be mixed into the measuring gas using a bypass.

The air flow through scintillation chamber and HV-measuring chamber was about 20 l/h.

3 Measuring results for calibration factor and discussion

Results of measurements were related to calibration factor (*CF*). *CF* is the inverse of the sensitivity of the chamber and defined from the equation

$$C_{Rn} = CF \cdot I \tag{2}$$

were

C_{Rn} : activity concentration of Rn-222 and

I : count rate of alpha pulses.

The dependency from measuring conditions was expressed as *CF* as function of absolute humidity (*a.H.*) or Rn-activity concentration C_{Rn}.

$$CF = f(a.H.) \text{ or} \tag{3}$$

$$CF = f(C_{Rn}). \tag{4}$$

To allow a more detailed interpretation of the influence of humidity and activity concentration the tests was grouped into classes of the quantities separately.

The function $CF = f(C_{Rn})$ was grouped into the 3 classes of abs. humidity $a.H.$ [1]

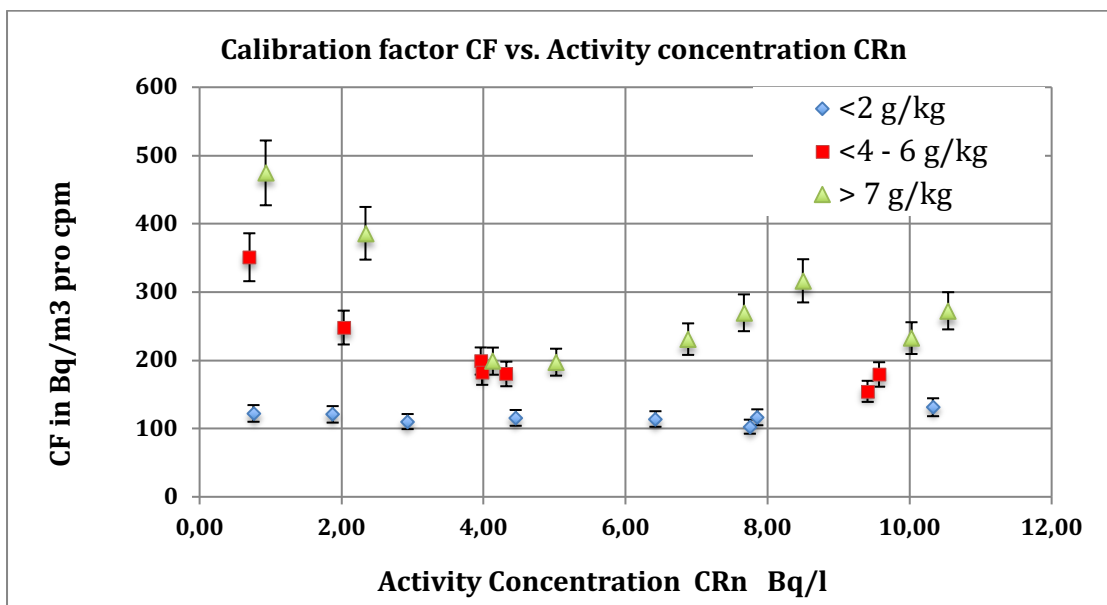a.H. < 2   g/kg,

a.H. 4 – 6   g/kg and

a.H. > 7   g/kg.

The function $CF = f(a.H.)$ was grouped into the 3 classes of activity concentration C_{Rn}

C_{Rn} < 2.5   Bq/l,

C_{Rn} 2.5 – 8   Bq/l and

C_{Rn} > 8   Bq/l.

Fig. 3 Calibration factor $CF = f(C_{Rn})$ as function of activity concentration for different groups of humidity [2]

Fig. 3 shows:

- At small values of humidity (< 2 g/kg) the dependency of calibration factor from activity concentration is not significant.
- Higher values of humidity (> 4 g/kg) causes significant higher calibration factors.
- For small activity concentrations the increase of calibration factor at humidity > 4 g/kg is greater than for higher one.
- Higher values of humidity (> 4 g/kg) causes significant more variation of calibration factor

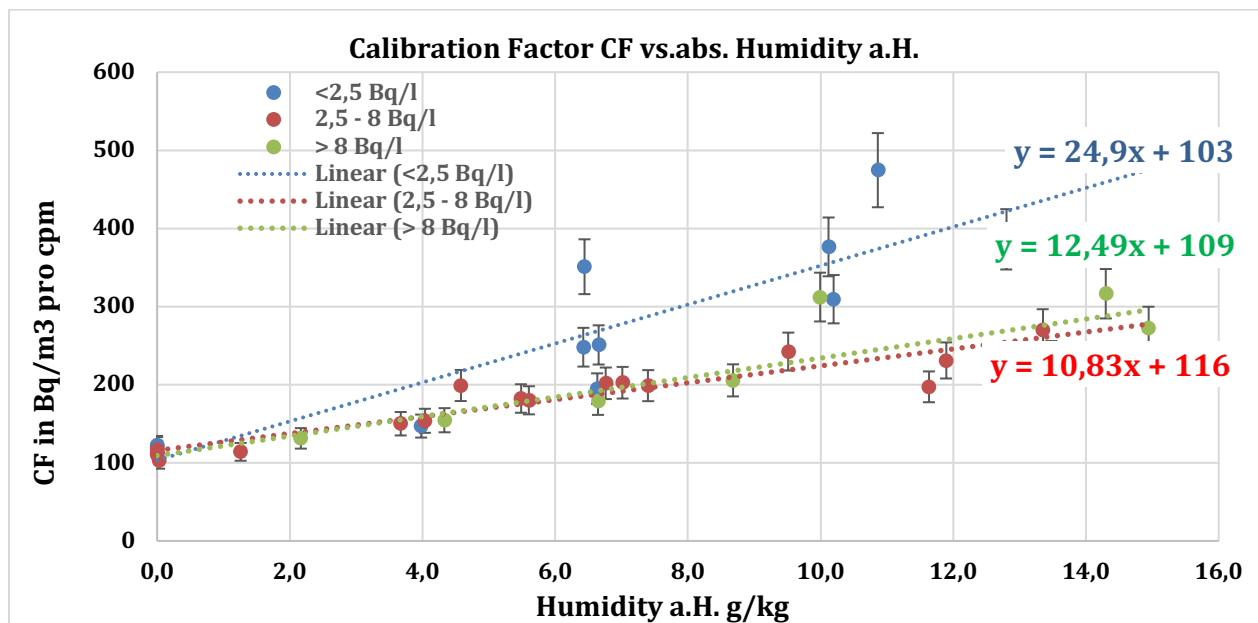**Fig. 4   Calibration factor CF for the different groups of Rn-activity concentration C_{Rn}**

Fig. 4 shows:

- The functions $CF = f(a.H.)$ can be fitted by a linear function.
- Higher humidity causes more variation and in consequence more uncertainty.
- the slope of the function $CF = f(a.H.)$ for small activity concentrations (< 2.5 Bq/l) is obvious higher than for higher activity concentrations. This is in accordance with the results of the function $CF = f(C_{Rn})$. The result is not in accordance with expected behaviour and cannot be explained with counting statistic.

Summery

i.   The calibration factor not depends from activity concentrations at low humidity (< 2 g/kg a.H.) in the measuring air.

ii. Higher humidity causes uncertainties and dependency of calibration factor from activity concentration.

iii. At low activity concentrations (< 2.5 Bq/m³) the increase of calibration factor with humidity is much more than for higher activity concentrations.

Discussion

The found results i. and ii. for the dependence of calibration factor from activity concentration and humidity (see topic 4) are not new findings but shows the need for the consideration of this dependencies for quality assurance for instruments used for the radon measurement based on HV enrichment of Po-218-ions.

The finding of results iii. is surprising but significant and not to explain with uncertainties of measurements. It would be fine when anyone can confirm this finding and search for a physical reason for this.

Quality assurance tests for measuring instruments used for radiation protection at environmental conditions are necessary furthermore.

Please be aware that the found dependencies are valid for big volumes of the measuring chamber. It can be assumed that smaller volumes show smaller dependencies.

4 Calculation of neutralisation rates

To asses neutralisation rates explaining the measured calibration factors for the used measuring chamber a model of the electric field strength was established. Therefore a spherical geometry was used. The principal approach is the same as used by Hopke {2} calculating the maximal distances of an ion in the mean lifetime of the ion. For the model it is assumed that the detector is located in the midpoint of the sphere. The volume contributing to sensitivity is limited by the radius of a sphere around the detector the ion can move to the detector before neutralisation. The parameters determining the radius are the electric field strength, the electrical mobility of the ion and the mean lifetime of the ion. In this way the maximal radius of the sphere contributing to sensitivity can be calculated for any lifetime (or neutralisation rate) using a known mobility.

Calculation:

Electric field strength of a sphere:

$$E = \frac{R_1 \cdot R_2}{(R_2 - R_1)} \cdot \frac{U}{r^2} \tag{7}$$

with

R_1: inner radius of the sphere,
R_2 outer radius of the sphere,
U : Voltage,
r : radial variable.

setting

$$a = \frac{R_1 \cdot R_2}{(R_2 - R_1)} \cdot U \tag{8}$$

the velocity of an ion will be

$$v(r) = B_{el} \cdot \frac{a}{r^2}. \tag{9}$$

The time T for a distance L at a variable velocity $v(r)$ is

$$T = \int_0^L \frac{dr}{v(r)} \tag{10}$$

using eq. ()

$$T = \frac{1}{a \cdot B_{el}} \int_0^L r^2 dr \tag{9.5}$$

integration

$$T = \frac{1}{3 \cdot a \cdot B_{el}} \cdot L^3 \tag{11}$$

changing to L

$$L = \sqrt[3]{3 \cdot a \cdot B_{el} \cdot T} \quad \text{and} \tag{12}$$

or

$$L = \sqrt[3]{3 \cdot \frac{R_1 \cdot R_2 \cdot U}{R_2 - R_1} \cdot B_{el} \cdot T} . \tag{13}$$

The distance L is the maximal available radius around the detector.

It has to taken into account that the model is strong valid only for a spherical detector with radius R1. The used geometry for experiments is a cylinder with a small detector area in the top of the cylinder.[3]
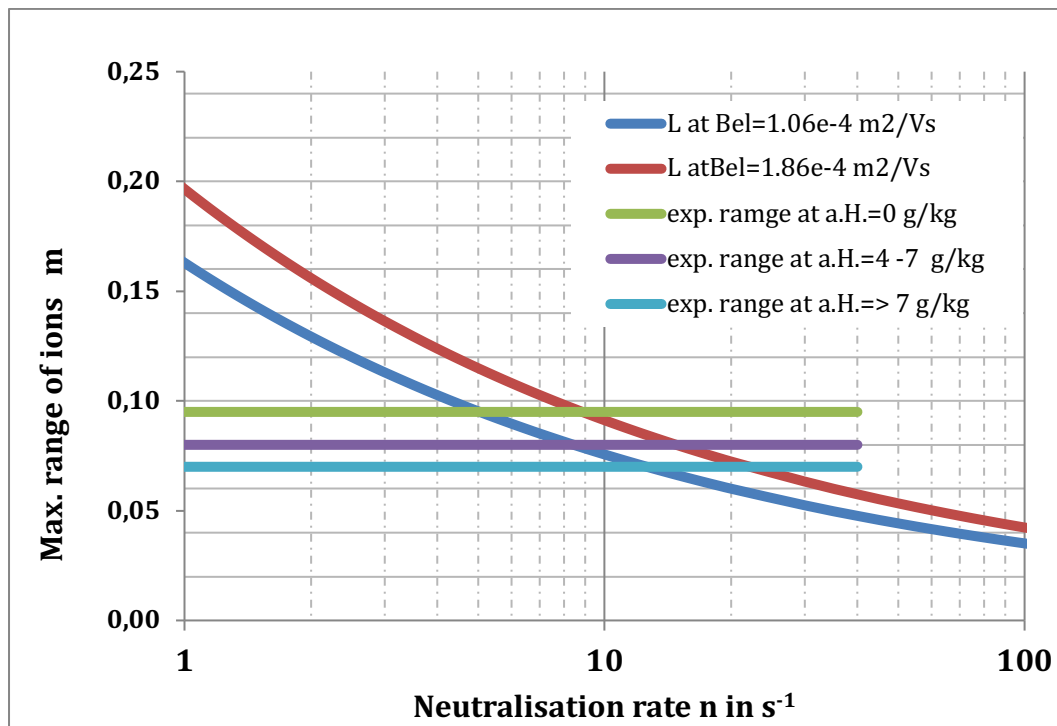
Figure 7 shows the results of calculation for two different values of electrical mobility. That's because some authors published different values for diffusion coefficient of Po-218 ion. On the other hand Chu/Hopke{1} pointed out that the diffusion coefficient depends from the age of Po-218. It is assumed that range of the different values of mobility represents the uncertainty of the results for neutralisation rate.

Used parameters:

- detector radius R_1=1 cm;
- cylinder radius R_2= 8,5 cm;
- B_{el} (1) =1,86e-4 m²/Vs (used by Hopke{2})
- B_{el} (2) =1.06e-4 m²/Vs
- Voltage U=1200 V.

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[3] The difference between model and experiment seems to be negligible because the measured maximal distances from point of decay to detector are smaller or about equal than cylinder radius.

Fig. 7 Maximale distance of an ion in a spheric chamber as function of neutralisation rate

Using the results of experiments the maximal distances measured at different humidity was calculated. The estimated distances were

➢ L1:   9.5 cm for a humidity < 0.1 g/kg (dry air),
➢ L2:   8.0 cm for a humidity 4 -7 g/kg (dry air) and
➢ L3:   7.0 cm for a humidity of > 7 kg (dry air).

Figure 7 shows that the resulting neutralisation rate is:

➢ **5,0 s⁻¹ up to 8.75 s⁻¹** for a humidity < 0.1 g/kg (dry air),
➢ **8.5 s⁻¹ up to 15 s⁻¹** for a humidity 4 - 7g/kg (dry air) and
➢ **12 s⁻¹ up to 20 s⁻¹** for a humidity of > 7 kg (dry air).

Discussion

The estimated values of neutralisation rates n are between the values found by Chu/Hopke {1} of about n=48 s⁻¹ and Pagelkopf/Porstendörfer {3} n=0,011s⁻¹ up to n=0,087 s⁻¹. Despite of the uncertainties caused by uncertainty of used mobility of ions, differences between model of electric field and experiment and uncertainties of measured values of sensitivity as well the estimated values of neutralisation rates differ from this of Pagekopf/Porstendörfer {3} significantly and are near to the values of Chu/Hopke{1}. An interpretation of this finding using the experimental data is not possible. Maybe there is an obvious difference between the

experimental setup of Chu/Hopke{1} and Pagelsdorf/ Porstendörfer {3}. In the setup used by Pagelkopf/ Porstendörfer {3} no electric field is influencing the neutralisation process while neutralisation rate at the experiments of Chu/Hopke{1} and at the own experiments is influenced by an electric field. This findings show the need of further experiments to investigate physical processes in the measuring chamber.

References

- {1}: Kal-Dee Chu and Phillip K. Hopke; Environ. Sci. Technol. Vol. 22, No.6, 1988

- {2}: Phillip K. Hopke; Health Pys., Vol.57, No. 1, pp.39-42, 1989

- {3}: P- Pagelkopf, J. Porstendörfer; Atmospheric Environment 37 (2003) 1057-1064