CHARACTERISTICS OF RADON- AND THORON DAUGHTERS AEROSOLS

Project ID: BI7*0047

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Project details

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<th>Total cost:</th>
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<td>Not available</td>
<td>Funding scheme:</td>
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<td>CSC - Cost-sharing contracts</td>
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Objective

A radon chamber was designed to study the dynamics of cluster formation processes of radon progeny in controlled atmospheres. The major parts of this chamber were an electrostatic classifier and a rotating screen diffusion disk for measuring radioactive particles in the diameter size range between 0.5 and 10 nm. The dimension and the inlet of the classifier were designed to minimize diffusion losses of small particles.

It is planned to measure the inactive number size distribution by a commercial available condensation particle counter (CPC) (TSI, CPC Model 3025). The capacity of this CPC was tested with monodisperse sodium chloride and silver particles. A 50% registration efficiency of 2.9 to 3.1 nm was found and a slight decrease of efficiency from 1 to 0.96 in the particle concentration region above 60 000 cm\(^{-3}\). The influence of the chemical substance of the aerosol particles on the detection efficiency (hygroscopic or nonhygroscopic) can be neglected.

Activity size distributions of the radon decay products RaB and RaC are usually determined by conventional impactors using gamma-spectroscopic methods after air sampling. However, more detailed investigations require the measurement of the size distributions of the aerosol attached fraction of all nuclides RaA-RaC. For this purpose a low level cascade impactor was designed and constructed where the size fractionated activities can be measured during air sampling by alpha-spectroscopy. This impactor consists of 9 stages and a back up filter and works at a flow rate of 5 m\(^3\) h\(^{-1}\). The aerosol attached activities of each stage are deposited on surface barrier detectors with large areas (2000 mm\(^2\)) covered with thin foils (4 um). The energy resolution is sufficient to determine all size distributions of RaA-RaC from the emitted alpha particles. Rotating detector foils yield sampling times of some weeks for measurements in the human environment at low activity concentrations. The calibration of this impactor is in progress.

Size distribution measurements of the thoron decay product ThB lead-212 were continued in indoor and outdoor environments using a low pressure impactor (type BERNER). More than 80% of the activity was associated to aerosol particles in the accumulation mode with average diameters of 217 nm indoors and 330 nm outdoors, respectively. From these measurements it can be estimated that about 10% of the activity is attached to particles in the nucleation region with median diameters less than 82 nm. The ThB size distributions are similar to the size distributions of the short lived radon progeny.

The deposition rate constants of unattached polonium-218 and lead-214 were evaluated from the measured radon and decay product concentrations in a 1 m\(^3\) chamber as a function of the degree of turbulence. The turbulence is induced by ventilation and by generating heat. The progress of the deposition rate constants is the same for both decay products. The value for lead-214 is however significantly lower than that of polonium-218.

The theory of Crump and Seinfeld was fitted to deposition data and the coefficient, ke, and the exponent (n) in the definition of the eddy diffusion coefficient were examined. The coefficient ke was found to be proportional to lambda V\(^3\) (ventilation)
and $W^{3/2}$ (generated heat). With this data it is not possible to discriminate between the values of the exponent, $n$, reported in the literature.

A clear difference was found between the deposition rate constants of polonium-218 and lead-214. The corresponding difference in diffusion coefficient was about a factor of 3, which means that the diameter of the unattached lead-214 particle is twice as large as the diameter of the unattached polonium-218 particle. The difference could be due to the physical and chemical properties of the 2 elements.

The models currently used for predicting the concentrations of the decay products in the indoor environment assume equal deposition rate constants for polonium-218 and lead-214. However a significantly lower deposition rate constant for lead-214 would explain some of the difficulties found in fitting the model to the data collected during case studies in houses. Indeed, when the deposition rate constant of the unattached lead-214 is applied to the unattached polonium-218, the measured radon concentrations are systematically underestimated.

It is possible to design a measurement system based on wire screen penetration theory to simulate the deposition of the decay products in the nasal cavity and in the tracheobronchial region. The measurement system consists of 3 sampling heads, an opening faced filter giving the total airborne activity, a screen covering the filter yielding the nasal deposition and 5 screens covering the filter yielding nasal and bronchial deposition. The sampler is under construction. The 3 simultaneous measurements must be comparable within 1% in order to give meaningful results. Limiting orifices and a special design of the sampling head have been applied to meet this goal.

The experimental facility for radon studies at Lund Institute of Technology has been further developed and introductory studies regarding radon daughter aerosol particle interaction have been performed. A multiorifice impactor stage has been constructed and tested.

The experimental facility comprises room (volume = 19.8 m$^3$) housed in a trailer. The walls are made of urethane, covered with fibreglass, plastic and plastic paint. Inside, there are 12 supply terminals and 12 extract terminals along the sides of the room.

Pressurized air from a compressor is filtered and dried and then supplied to the room. Various aerosols can be added and mixed in the glove box. The instruments for monitoring the concentrations of radon, radon progeny and nitrogen oxide, are located outside the room. The size distribution of the passive and the active aerosol is also measured. Plate out is measured with a large area pulse ionization chamber. The unattached fraction is measured with the single screen and filter technique.

Measurements show that the room can provide an atmosphere with an unattached fraction ranging from 0 to 100% and an equilibrium factor ranging from 0.1 to 0.7 when the room is ventilated at a rate of 0.5 h$^{-1}$.

An impactor stage has been constructed and an experimental set up for testing and calibrating impactors for submicron particles has been designed.

In order to optimize the impactor design, the collection characteristics have been studied as a function of jet to plate distance and of the Reynold's number of the jet. The current impactor stage consists of 2700 laser drilled orifices. the diameter of the orifice plate is 26 mm. The orifice diameter is 50 mm. A cut off diameter between 50 and 100 nm can be achieved. The air flow is about 30 l/min and the pressure drop over the stage is about 20 kPa.

In order to identify sources of radon-222 and to characterize simultaneously indoor radon daughters (potential alpha energy, equilibrium factor, size distribution, unattached fraction) in houses with high radon level, studies have been carried out in 5 houses in Brittany. The houses were selected using the following criteria: radon concentration greater than 400 Bq m$^{-3}$, PAEC greater than 1 uJ m$^{-3}$, thermoluminescent dosimetry (TLD) gamma dose greater than 100 mrad y$^{-1}$. Corresponding values were obtained during field campaign of 1985 to 1989 in the living part of these houses.

From measurements of radon emanation rates through walls and ground, in conjunction with PAEC and ground level measurements, it has been possible to locate sources on mud floors in the basement of houses. Indeed, radon emanation rates were measured in the 5-200 E4 atmospheres m$^{-2}$ s$^{-1}$ range which is 5 to 10 times more than the usually encountered value. In those same houses, radon daughter size distributions have also been investigated and compared with natural particle size distribution.

Concerning the unattached fraction (fp) and activity mean diameter (AMD) measurements, values measured seem to be in agreement with those from other groups. However AMD of individual radon daughters appeared to be divergent. This can be explained by the fact that, because statistical errors of alpha counting have a significant effect on radon daughter concentration calculated on each sample by the Tsivoglou method, data treatment becomes more inconsistent. It is clear that effort has to be spent on improving counting and analysis procedures.

Experimental studies of the fraction of inhaled radon daughters deposited in the human airways have been carried out with a combination of 2 different techniques. In parallel with the exposure of the subject, measurements of the air borne radon daughters has been performed with a system of fine mesh wire screens that simulates the penetration and deposition in nasal and tracheobronchial region.

A radon chamber with a volume of 30 m$^3$ was used for the exposure. The concentration of radon-222 and radon-222 decay products, temperature aerosol concentration, ventilation and humidity were continuously recorded during the exposures. 2 extreme exposure conditions were chosen, namely high aerosol concentration giving virtually no airborne unattached fraction and low aerosol concentration where the unattached fraction dominated. The high aerosol concentration was achieved by burning organic material and running a condensation aerosol generator. The low aerosol was achieved by closing the ventilation system of the radon chamber and running an air cleaner inside the room.
Mouth breathing under resting condition with high aerosol concentration (less than 0.5% unattached fraction) was found to give a total deposition of inhaled radon daughters of 25%.

Nose breathing under resting condition with low aerosol concentration (approximately % unattached fraction) was found to give a total deposition of 85%. The radon daughters were found deposited in the head and bronchial part of the airways, with about half of the deposition in the nasal region. It is not possible to give more exact values due to experimental difficulties. This is due mainly to the relatively low levels of activity to be measured, but is also influenced by the poor spatial resolution of the lung detector due to relatively high energies of the gamma lines to be measured.

It was also found that exposure to only radon-222 without daughter products contribute to the gamma-emission from the body.

Additonal exposures and variation of the measuring geometry for the lung counter is planned to improve the accuracy of the regional deposition.

The instrumentation to measure the airborne radon daughters and the unattached fraction has been calibrated and an intercomparison has been carried out with the Isotopenlaboratorium in Goettinge. Very good agreement was obtained.

A charcoal dosimeter, 'canister', incorporating a silica gel layer has been developed with the aim of controlling the effects of humidity. The effect of this parameter has been checked. Research has also been carried out into the determination of gamma thoron daughters emitters by high resolution gamma spectroscopy. Also, several alpha spectroscopy chain detectors have been set up for active measurement of short lived radon decay products concentration in the air and of the unattached fraction. An electrostatic chamber has been designed and furnished by the Isotopenlaboratorium from Goettingen for the determination of the radon and thoron in the air. Finally training courses have been made available at the Isotopenlaboratorium of the University of Goettingen.

In all dosimetric models the particle size of the aerosol attached and unattached activities of short lived radon and thoron daughters are important parameters for the estimation of the radiation exposure.

Experiments will be continued to determine aerosol size characteristics of radon and thoron decay products in real living indoor and outdoor atmospheres. The results will be compared with model calculations.

The physical and chemical interaction (particle growth, cluster formation, plate out rates, etc.) of the unattached radon and thoron daughters with trace gases (SO₂, NOₓ, humidity) and other aerosol particles will be studied in chambers under controlled conditions.

For these investigations the sensitivity and efficiency of different experimental techniques for measurements of the size distributions have to be improved and modified techniques will be developed (eg diffusion batteries, low pressure impactors, multijet impactors, electrostatic classifier). These techniques have to be calibrated with monodisperse aerosol particles. The different methods including various data evaluation methods have to be compared during joint exercises.

Isotopenlabor, University of Gottingen, DE:

With different calibrated experimental techniques (high volume screen diffusion batteries, low pressure cascade impactors) the measurement of size characteristics (size distribution, fp, F) of the short lived radon daughters will be continued in the ambient air and first measurements of properties of thoron gas and decay products will be performed. In a radon chamber the growth and cluster formation processes, the influence of trace gases (SO₂, NOₓ), humidity, and ambient air and the interaction with monodisperse aerosol particles will be investigated.

Different data evaluation methods concerning the size distribution measurements (Twomey, expectationAmaximation, Simplex and Monte Carlo methods) will be compared.

Nuclear Research Centre (SCK/CEN) Mol, BE and

State University of Ghent, BE:
The complex interactions between the radon decay products and the indoor aerosol determine the unattached fraction which is responsible for about half of the lung dose. The factors influencing the concentrations of attached and unattached decay products will be studied in laboratory and in building environments.

University of Lund, SV:

In a research programme funded by the Swedish Natural Research Council new techniques for characterization of radon daughters are being developed and tested. As part of the project a radon room (volume about 20 m³) has been calibrated. In this room it is possible to vary the radon concentration, the air exchange rate and the concentration and size distribution of the aerosol.

One objective of the programme is to develop alpha spectrometry using detection over larger areas. This will increase the sensitivity and be of great value to future plate out and diffusion battery measurements.

A second objective is to determine the activity distributions of the attached radon daughters by using two new techniques; a multijet impacter, which combines a low cutoff and a high flow rate and a combination of a differential mobility analyzer (TSI 3071) and track etch plastic film.

National Institute of Radiation Protection (SSI), Stockholm, SV:

In earlier work performed at SSI the respiratory tract deposition of radon progeny in humans has experimentally been studied in dwellings, mining atmospheres and under laboratory conditions.

SSI will improve the earlier technique and contribute with deposition studies including measurements of the unattached fraction. The inhalation studies will also include deposition studies on the inactive aerosols.

SSI will also contribute in the work to improve the technique to calibrate and measure the unattached fraction collected on fine mesh wire screens.

Laboratoire de Physique des Aerosols et de Radioactivite Atmospherique, University of Brest, FR:
Improvement of alpha activity data treatment given by SDI 2000 (inertial and diffusional sampling device) to obtain size distributions of each short lived radon daughters in houses.

Elaboration of a technique for measuring size distributions of the unattached activities of the radon progeny.

Measurements in houses of the Bretagne having radon gas levels above 400 Bq m⁻³:
- research of radon sources;
- general studies of indoor environments (fp, F) and size distributions of radon daughters.

Institut de Tecniques Energetiques, Universitat Politècnica de Catalunya, INTE(UPC), Barcelona, ES:
The INTE(UPC) will participate as a subcontractor of the Isotopenlabor, University Göttingen, DE. The INTE(UPC) has been doing its research activities on radiation protection for the last ten years. The radiation protection section is devoted to the fields of gamma dosimetry and is dedicated to the field of low level radioactivity measurements.

In this project different experimental setups will be built up, in close collaboration with the Isotopenlabor of Göttingen, for measuring air activity concentrations of radon and thoron gas and of short lived decay products and for measuring activity size distributions.

Measurements of aerosol size characteristics (size distributions, fp, F) will be performed in a limited number of houses under real living conditions.

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