Flux measurements of nanometer-size particles using unattached radon decay products

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Outline of presentation

- Principle of flux measurement of nanometer-size particles above the ground
- Laboratory experiment for performance tests of developed system
- Field experiment of flux measurements
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Introduction

In terms of atmospheric environment:
• More information is needed on fluxes of nanometer-size particles, both to provide further tests of dry deposition models and to provide a better understanding of the formation and evolution of the nuclei mode of aerosol particles (less than 100 nm).
• In addition, the deposition velocity depends on the particle size, atmospheric stability, and turbulence. The deposition velocity values have been reported in the literature for particles larger than 100 nm.
  □ It is important to estimate the flux and deposition velocity of nanometer-size particles due to lack of such information.

In terms of dosimetry:
• The natural occurring radioactive materials are important for dose assessment.
  □ To better understand the mechanisms and phenomenon of radon and its decay products in the environment is necessary.

□ The purpose of this study: We develop a flux measurement system of nanometer-size particles using unattached $^{222}$Rn decay products as a tracer and show the preliminary results of laboratory and field experiments.
Generation and removal mechanisms of $^{222}\text{Rn}$ decay products in the environment

1. Emanation
   - Grain
   - Pore

2. Transport
   - Grain
   - Soil

3. Exhalation
   - Air

4. Decay
   - $^{226}\text{Ra}$
   - $^{222}\text{Rn}$
   - $^{218}\text{Po}$
   - $^{214}\text{Bi}$
   - $^{214}\text{Pb}$

5. Attachment
   - Unattached decay products (0.5-5 nm)
   - Aerosol-attached decay products (5-1000 nm)

6. Removal
   - Wet deposition
   - Dry deposition
   - Eddy turbulence

- $^{160}\text{y}$
- 4.9 MeV
- 5.5 MeV
- 3.1 MeV
- 6.0 MeV
- 7.7 MeV
- 0.164 ns
Flux measurement methods based on atmospheric concentration of interest and meteorological data

<table>
<thead>
<tr>
<th>Methodology</th>
<th>Atmospheric concentration</th>
<th>Meteorological data</th>
<th>Advantage</th>
<th>Disadvantage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aerodynamic gradient (AG) method</td>
<td>Height: more than 2 levels</td>
<td>Height: 1 level</td>
<td>Comparatively long measurement is possible</td>
<td>Uncertainty due to reaction between 2 heights and averaging for long-term measurement</td>
</tr>
<tr>
<td></td>
<td>Measurement interval: 10 min-some weeks</td>
<td>Measurement interval: 5-10 Hz</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Eddy correlation (EC) method</td>
<td>Height: 1 level</td>
<td>Height: 1 level</td>
<td>The best estimation precision</td>
<td>Difficulty of fast measurement of concentration and their data analysis</td>
</tr>
<tr>
<td></td>
<td>Measurement interval: 5-10 Hz</td>
<td>Measurement interval: 5-10 Hz</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Relaxed eddy accumulation (REA) method</td>
<td>Height: 1 level</td>
<td>Height: 1 level</td>
<td>Comparatively long measurement is possible</td>
<td>Difficulty of separately measuring upward and downward concentrations</td>
</tr>
<tr>
<td></td>
<td>Measurement interval: 10 min-some days</td>
<td>Measurement interval: 5-10 Hz</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

- The relaxed eddy accumulation (REA) technique is better to measure the nanometer-size particles using unattached $^{222}$Rn decay products.

Advantages of the REA technique to the flux measurement in this study are:
- The trace air samples can be accumulated in the collectors and then they can be analyzed because their concentrations in ambient air are possibly low, and
- The ease of particle detection instead of traditional aerosol particle counter due to difficulty of nanometer-size particles.

Businger and Oncley (1990) proposed the REA technique, where the air sampling into updraft and downdraft air collection devices (filter, reservoir, and so on) is done at constant flow rate, based on the signs of the vertical wind (updraft, neutral and downdraft).
Setup of relaxed eddy accumulation (REA) system

- Radiation counting system
- Wind speed measurement system
- Aerosol particle sampling system
- Solenoid valve control system
A schematic diagram of the REA system

1. Wind speed measurement system
   - Sonic anemometer
   - Wind speed measurement
   - Wind speed range: \((\pm 0.5 \text{ m s}^{-1})\)

2. Aerosol particle sampling system
   - Solenoid valve control system
   - Mass flow controller
   - Aerosol particle sampling

3. Solenoid valve control system
   - Sampling holder
   - C+ (Up)
   - C0 (Neutral)
   - C- (Down)

4. Radiation counting system
   - Radiation monitor
   - Radiation measurement
Collection of unattached $^{222}$Rn decay products using metal wire screen

- Collection method of $^{222}$Rn decay products (RnDP)
  - Unattached RnDP: Metal wire screen
  - Aerosol-attached RnDP: Filter

Mesh size: 400
Flow rate: 18 L min$^{-1}$

50% collection for aerosol particles by metal wire screen based on theoretical assumptions regarding the screen

4 nm in diameter
Counting for alpha-particles emitted from $^{222}$Rn decay products

- ZnS(Ag) scintillation detecting system
  - After a 30-min sampling, alpha particles on the wire screen and filter were counted using the ZnS(Ag) scintillation detector. The progeny concentrations were estimated by the three-count method (Thomas method).
  - The equilibrium equivalent $^{222}$Rn concentrations (EERC, Bq m$^{-3}$) are calculated:
    \[
    \text{EECR} = (0.105 C_1 + 0.516 C_2 + 0.380 C_3)
    \]
    where $C_i$ ($i = 1, 2$ and $3$) is concentrations of $^{218}$Po, $^{214}$Pb and $^{214}$Bi in the air, respectively.
Outline of presentation

- Principle of flux measurement of nanometer-size particles above the ground
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**Experimental condition:**
- $^{222}$Rn concentration: 10 kBq m$^{-3}$
- Temperature: 20 ºC
- Relative humidity: 60%
- Aerosol: 100 nm (Carnauba wax particles)

**Laboratory experiment using the NIRS $^{222}$Rn chamber**

Concentration measured in the “Reference” line was estimated based on the continuous 30-min sampling.

Concentration measured in the “REA” sampling system was the average of three unattached $^{222}$Rn decay products concentrations ($C^+$, $C^0$ and $C^-$) when the “up”, “neutral” and “down” valves open, respectively.
Results of REA sampling system using unattached $^{222}$Rn decay products (u-RnDP)

Unattached $^{222}$Rn decay products using wire screen

$\text{u-RnDP in REA system (Bq m}^{-3}\text{)}$

Aerosol-attached $^{222}$Rn decay products using filter

$\text{a-RnDP in REA system (kBq m}^{-3}\text{)}$

The linear regression slope for unattached $^{222}$Rn decay products indicates that the REA sampling system losses of about 20% compared to the reference line.
Outline of presentation

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Field experiment

Sampling site: Saitama Prefectural Comprehensive Education Center

A: REA system
B: Counting system
# Measurement conditions

**Table. Summary of experimental and weather conditions**

<table>
<thead>
<tr>
<th>Experimental</th>
<th>June</th>
<th>August</th>
</tr>
</thead>
<tbody>
<tr>
<td>Measurement period (day)</td>
<td>2-4 (3 d)</td>
<td>24-27 (4 d)</td>
</tr>
<tr>
<td>Ground surface condition</td>
<td>Pasture</td>
<td>Pasture-patched soil</td>
</tr>
<tr>
<td>Height of pasture (m)</td>
<td>1</td>
<td>0.1-0.3</td>
</tr>
<tr>
<td>Rainfall event (daytime)</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>Temperature (°C) (daytime)</td>
<td>29 ± 2</td>
<td>39 ± 4</td>
</tr>
<tr>
<td>Relative humidity (%) (daytime)</td>
<td>33 ± 8</td>
<td>41 ± 11</td>
</tr>
<tr>
<td>Wind speed (m s⁻¹) (daytime)</td>
<td>2.0 ± 0.5</td>
<td>1.5 ± 0.3</td>
</tr>
</tbody>
</table>
Data screening of flux measurements of unattached $^{222}$Rn decay products using the REA system

Table. Summary of mean (± SD) concentrations of unattached $^{222}$Rn decay products in outdoor air

<table>
<thead>
<tr>
<th>City, country</th>
<th>Measurement site (height in m)</th>
<th>U-RnDP (n) (Bq m$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shanghai, China</td>
<td>Grass (1 m)</td>
<td>0.39 ± 0.23 (16)</td>
</tr>
<tr>
<td>Yanji, China</td>
<td>City (ca. 15 m)</td>
<td>1.09 ± 0.83 (15)</td>
</tr>
<tr>
<td>Hirosaki, Japan</td>
<td>City (ca. 15 m)</td>
<td>1.06 ± 0.41 (8)</td>
</tr>
<tr>
<td>Kumagaya, Japan</td>
<td>Pasture (0.1-1m)</td>
<td>0.16 ± 0.08 (15)</td>
</tr>
</tbody>
</table>

- The data were screened out according to two data screenings to ensure their credibility.

Table. Data screening of flux measurements

<table>
<thead>
<tr>
<th>Data screening</th>
<th>Remaining data (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>June</td>
<td>94</td>
</tr>
<tr>
<td>August</td>
<td>94</td>
</tr>
<tr>
<td>1. Wind direction</td>
<td>94</td>
</tr>
<tr>
<td>2. $C_{\text{Ref}} \geq 0.1$ Bq m$^{-3}$</td>
<td>38</td>
</tr>
<tr>
<td>The number of initial point</td>
<td>16</td>
</tr>
</tbody>
</table>

Fig. Activity concentration ratio of REA system to reference as a function of unattached $^{222}$Rn decay products (u-RnDP) measured in field experiments

- Increase of uncertainty in REA system with a decrease of unattached $^{222}$Rn decay products concentration less than approximately 0.1 Bq m$^{-3}$
Results of flux measurements of unattached $^{222}\text{Rn}$ decay products using the REA system during the daytime

<table>
<thead>
<tr>
<th>Symbol</th>
<th>June</th>
<th>August</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Emission</td>
<td>Deposition</td>
</tr>
<tr>
<td>Flux ($\times 10^{-2}$ Bq m$^{-2}$ s$^{-1}$)</td>
<td>$F$</td>
<td>3.7 (2.6−4.9)</td>
</tr>
<tr>
<td>Unattached RnDP concentration (Bq m$^{-3}$)</td>
<td>$C$</td>
<td>0.13 (0.11−0.16)</td>
</tr>
<tr>
<td>Transfer velocity (cm s$^{-1}$)</td>
<td>$V_f$</td>
<td>-27 (-31−24)</td>
</tr>
<tr>
<td>Number of data after the criteria (all data)</td>
<td></td>
<td>2 (16)</td>
</tr>
<tr>
<td>Radon concentration (Bq m$^{-3}$)</td>
<td>Rn</td>
<td>4.4 ± 1.0</td>
</tr>
</tbody>
</table>

The flux ($F$) is calculated as follows:

$$F = b \sigma_w (C^+ - C^-)$$

$b$: empirical dimensionless coefficient

$\sigma_w$: standard deviation of vertical wind speed

$C^+$ and $C^-$: average concentrations of species sampled during updraft and downdraft, respectively

$$V_f = -\frac{F}{C}$$

$V_f > 0$: Deposition

$V_f < 0$: Emission
Comparison of deposition velocities ($V_d$) of nanometer-size particles

<table>
<thead>
<tr>
<th>Deposition surface</th>
<th>Object substances</th>
<th>$D_p$ (nm)</th>
<th>$V_d$ (cm s$^{-1}$)</th>
<th>$z^*$ (m s$^{-1}$)</th>
<th>n</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pasture (1)</td>
<td>u-RnDP$^a$</td>
<td>4$^b$</td>
<td>18 (1.2–35)</td>
<td>0.2 (0.2–0.2)</td>
<td>4</td>
<td>This study (June)</td>
</tr>
<tr>
<td>Pasture-patched soil (0.1–0.3)</td>
<td>u-RnDP$^a$</td>
<td>4$^b$</td>
<td>18 (16–22)</td>
<td>0.1 (0.1–0.1)</td>
<td>3</td>
<td>This study (August)</td>
</tr>
<tr>
<td>Semiarid desert covered with low grass (0.3)</td>
<td>u-RnDP$^a$</td>
<td>2.7$^b$</td>
<td>7.3 ± 2.5 (5–35)</td>
<td></td>
<td>22</td>
<td>Schery et al. (1998)</td>
</tr>
</tbody>
</table>

$^a$Unattached $^{222}$Rn decay product.

$^b$Particle diameter for 50% penetration into wire mesh screens.
The REA system was developed in order to measure the flux and deposition velocity of nanometer-size particles using unattached radon decay products as a tracer.

The performance of REA sampling system was tested in the laboratory and field experiments.

In the field experiment, deposition velocity of nanometer-size particles was a good agreement with the results reported in the literature.
Thank you for your kind attention