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

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Outline of presentation

ÅPrinciple of flux measurement of nanometer-size particles above the ground

ÅLaboratory experiment for performance tests of developed system

ÅField experiment of flux measurements

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Introduction

In terms of atmospheric environment:

Å 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).

Å 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.

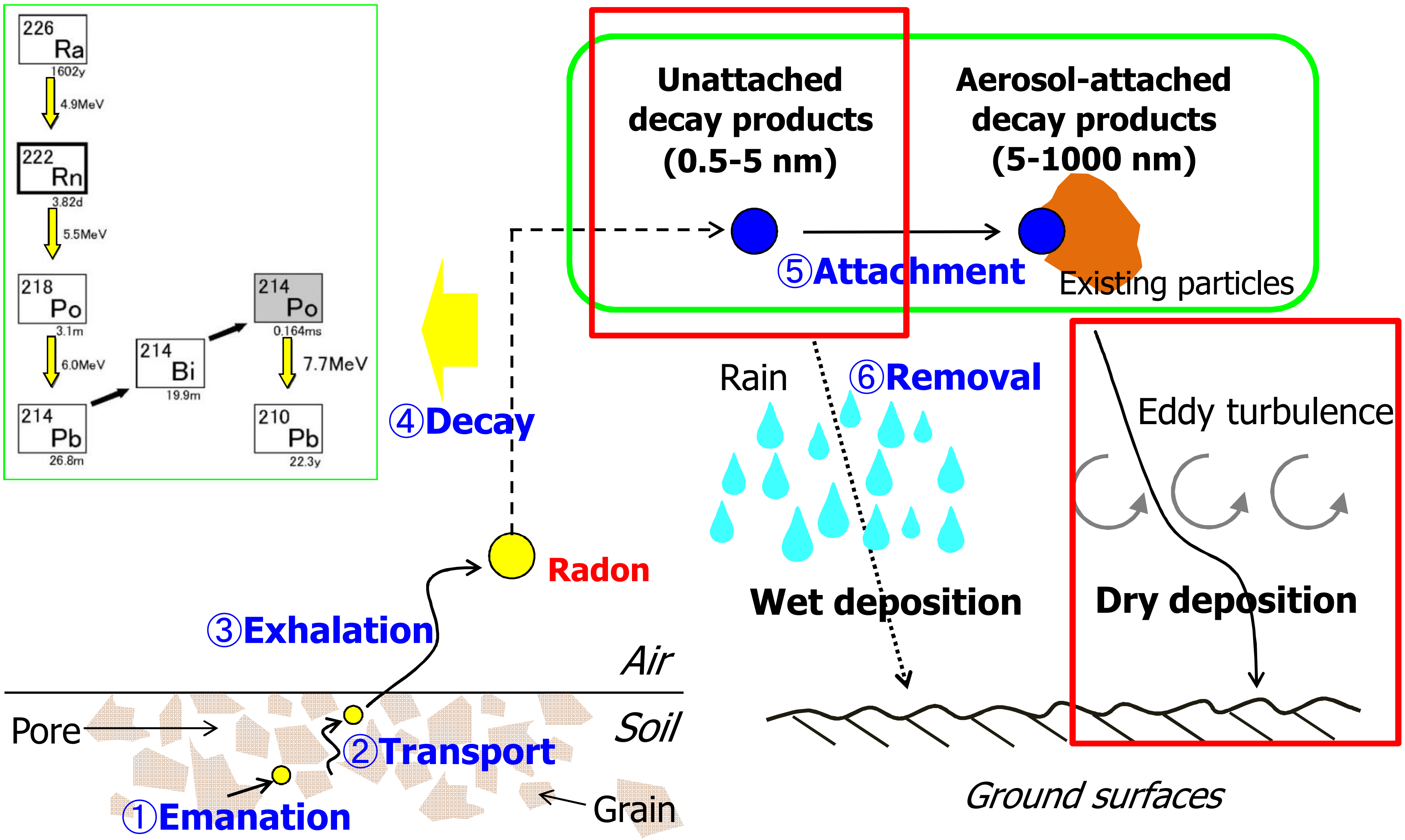
In terms of dosimetry:

Å 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}Rn decay products in the environment



Flux measurement methods based on atmospheric concentration of interest and meteorological data

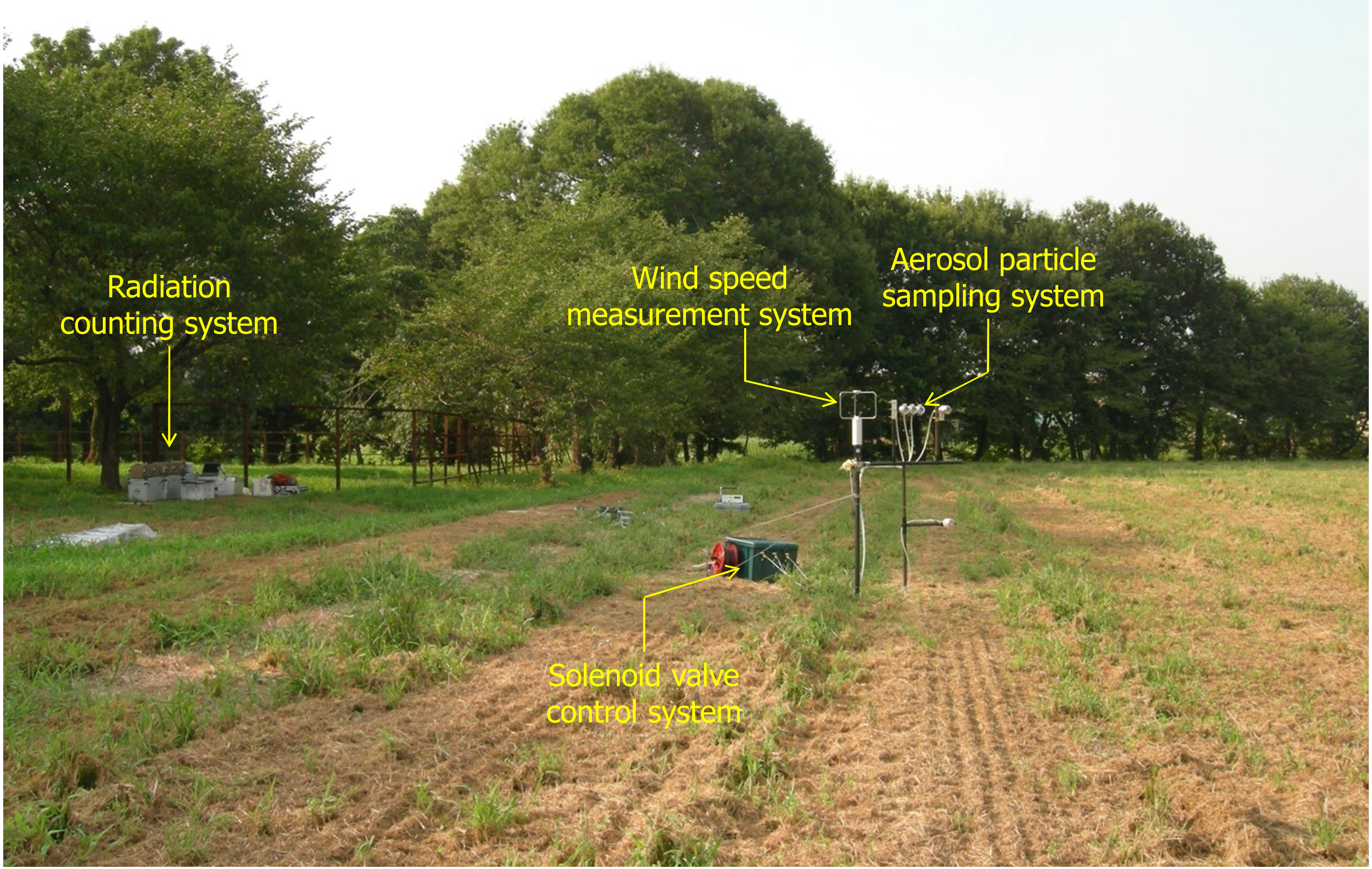
Methodology	Atmospheric concentration	Meteorological data	Advantage	Disadvantage
Aerodynamic gradient (AG) method	ÅHeight: more than 2 levels ÅMeasurement interval: 10 min-some weeks	ÅHeight: 1 level ÅMeasurement interval: 5-10 Hz	ÅComparatively long measurement is possible	ÅUncertainty due to reaction between 2 heights and averaging for long-term measurement
Eddy correlation (EC) method	ÅHeight: 1 level ÅMeasurement interval: 5-10 Hz	ÅHeight: 1 level ÅMeasurement interval: 5-10 Hz	ÅThe best estimation precision	ÅDifficulty of fast measurement of concentration and their data analysis
Relaxed eddy accumulation (REA) method	ÅHeight: 1 level ÅMeasurement interval: 10 min-some days	ÅHeight: 1 level ÅMeasurement interval: 5-10 Hz	ÅComparatively long measurement is possible	ÅDifficulty of separately measuring upward and downward concentrations

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

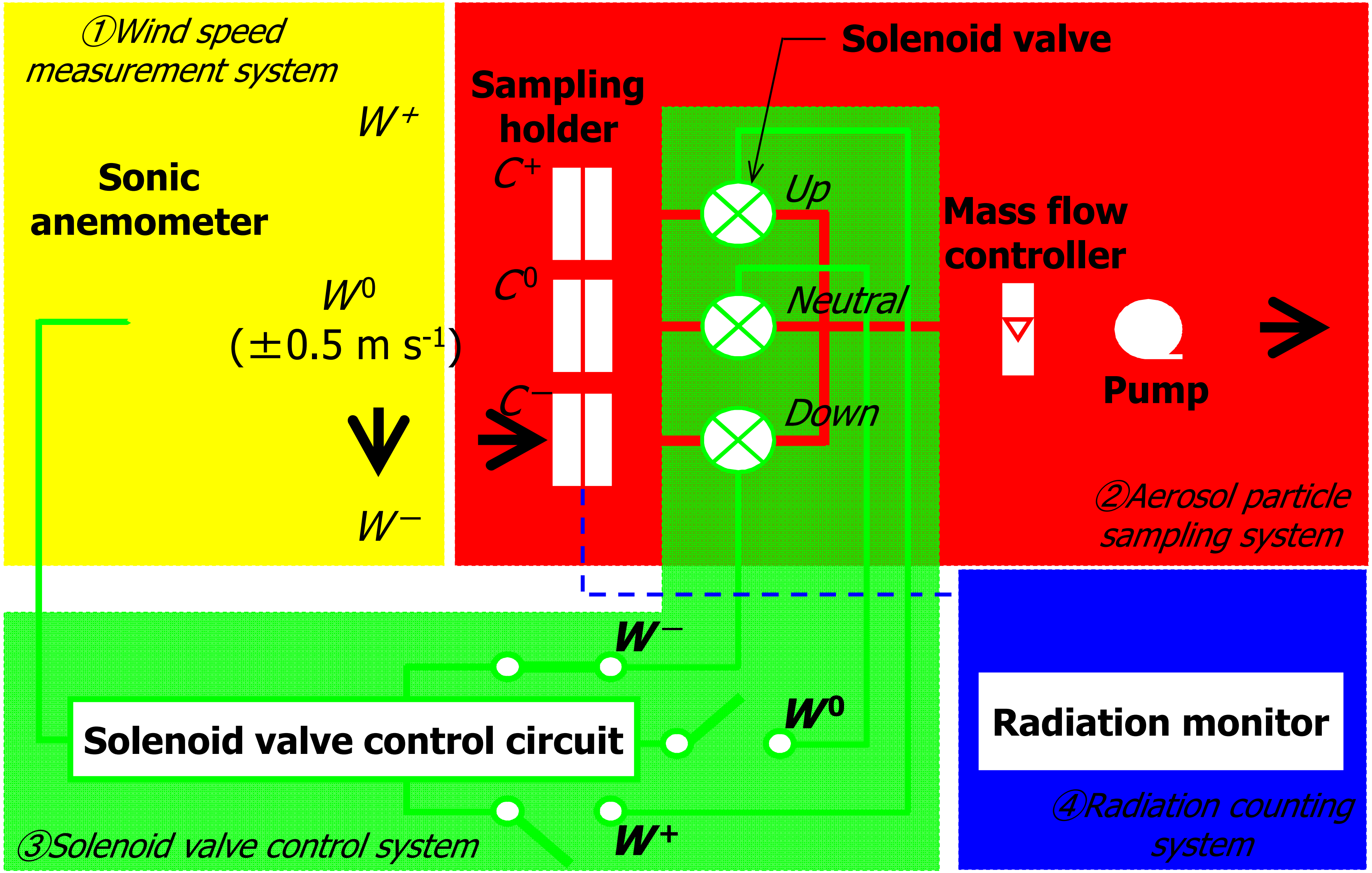
Advantages of the REA technique to the flux measurement in this study are:
Åhe trace air samples can be accumulated in the collectors and then they can be analyzed because their concentrations in ambient air are possibly low, and
Åhe ease of particle detection instead of traditional aerosol particle 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).

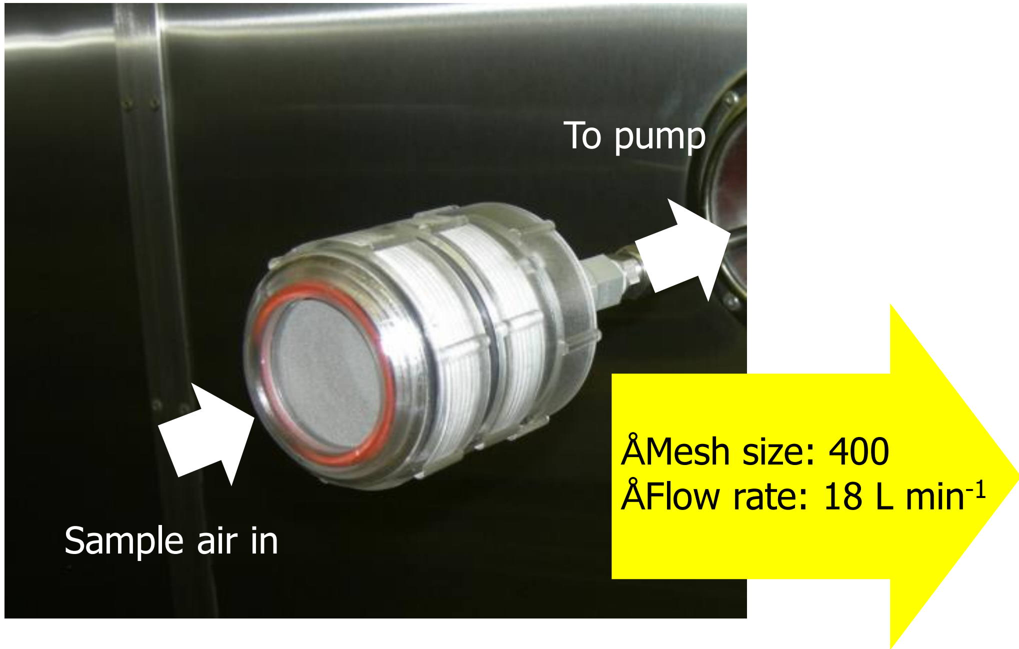
Setup of relaxed eddy accumulation (REA) system



A schematic diagram of the REA system



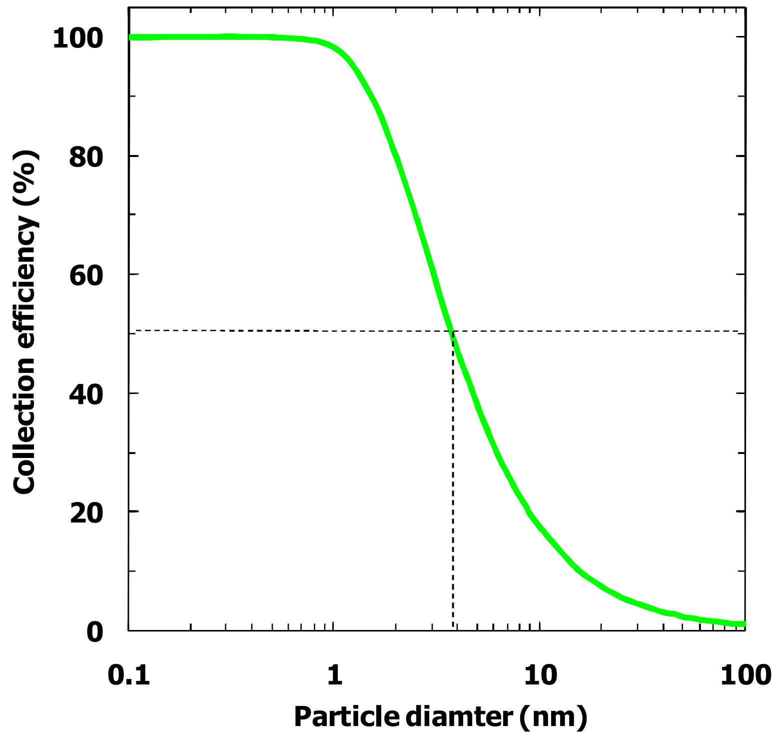
Collection of unattached ^{222}Rn decay products using metal wire screen



☐ Collection method of ^{222}Rn decay products (RnDP)

Å**Unattached RnDP**: Metal wire screen

Å**Aerosol-attached RnDP**: Filter



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

Å 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).

Å The equilibrium equivalent ^{222}Rn concentrations (EERC, Bq m^{-3}) are calculated:

$$\mathbf{EECR = (0.105\,C_1 + 0.516\,C_2 + 0.380\,C_3)}$$

where C_i (i = 1, 2 and 3) is concentrations of ^{218}Po, ^{214}Pb and ^{214}Bi in the air, respectively.

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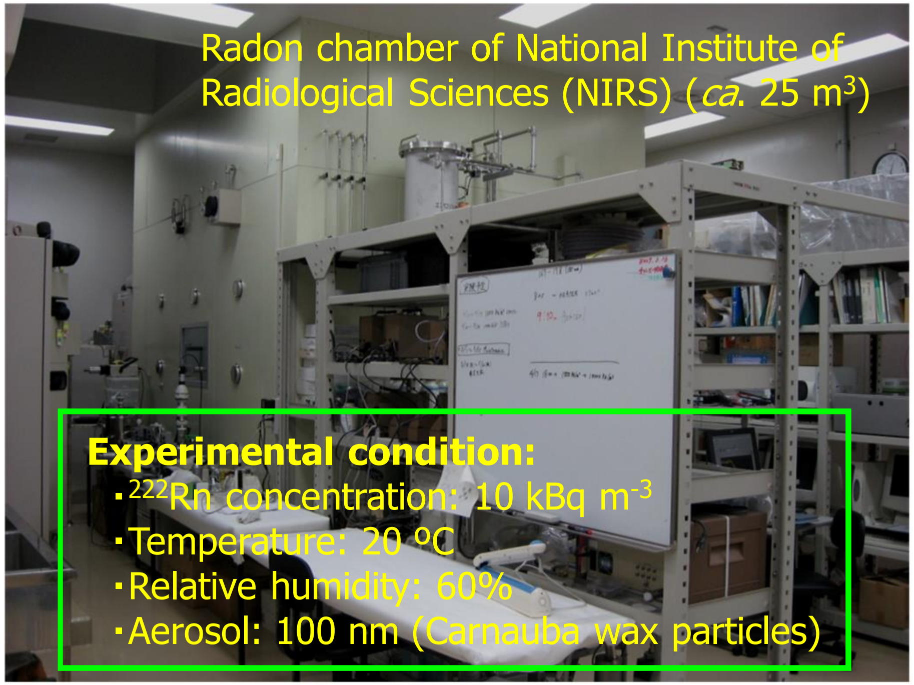
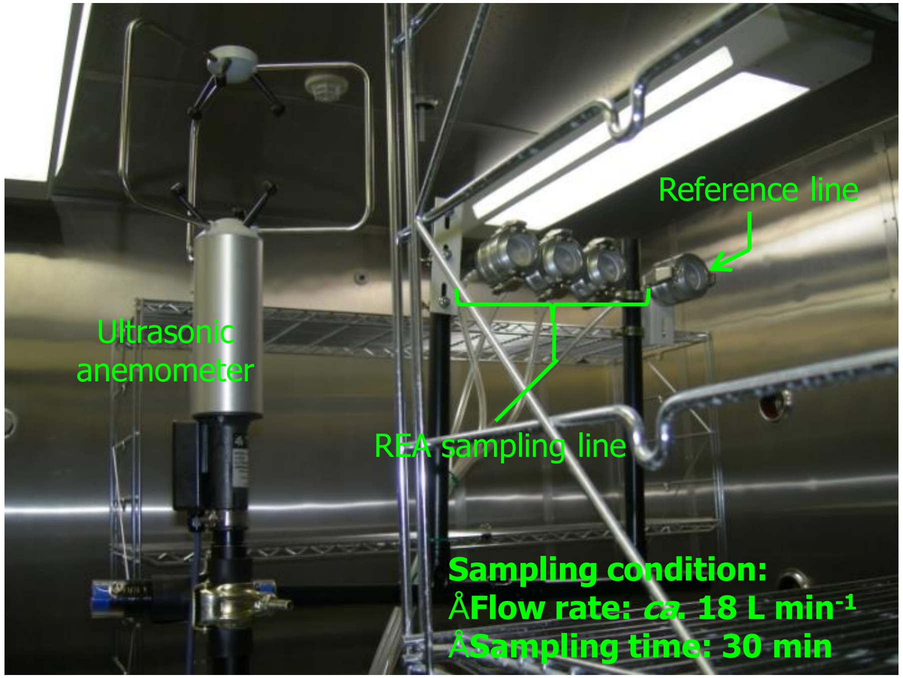
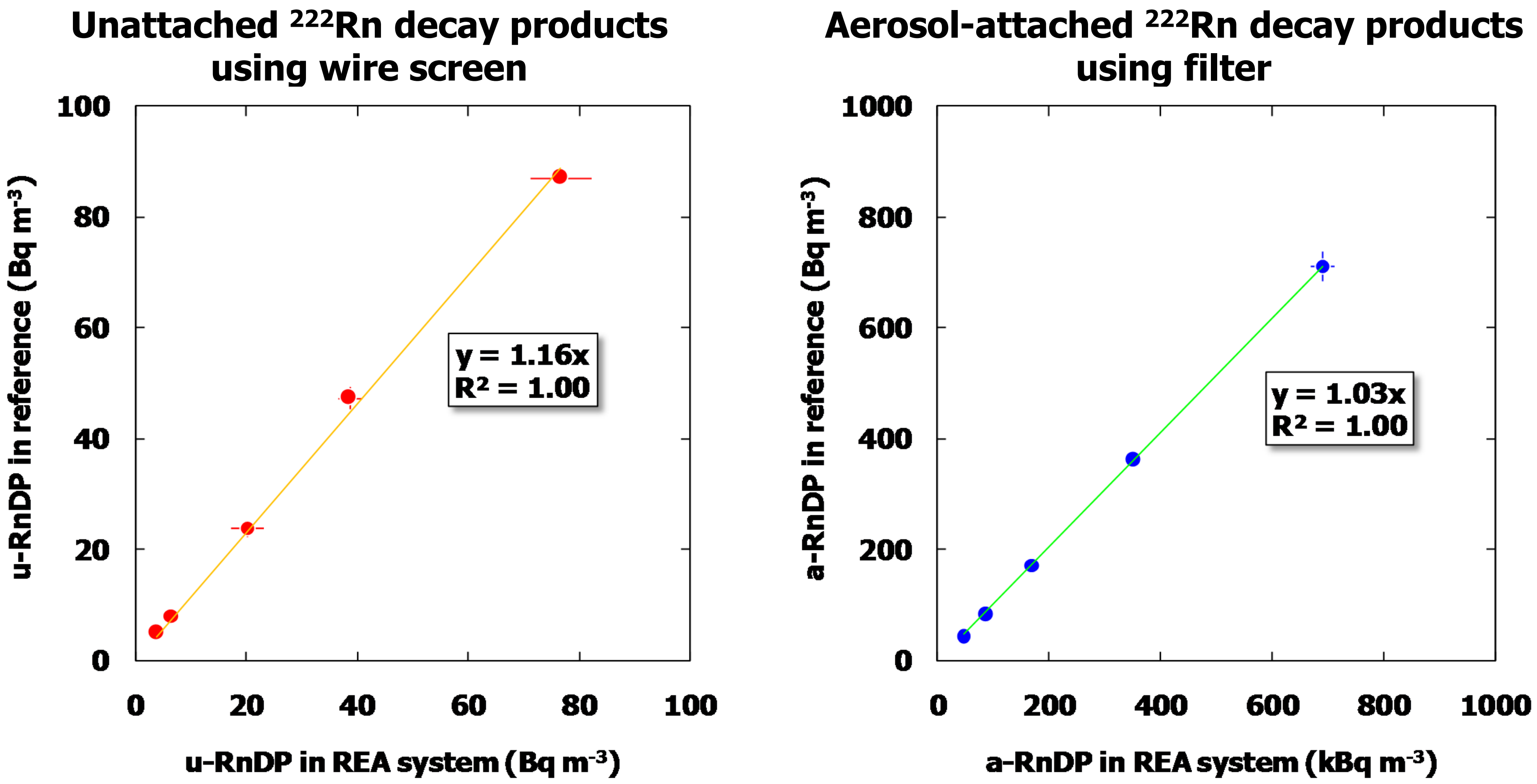
Laboratory experiment using the NIRS ^{222}Rn chamber





- 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)



Å 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.

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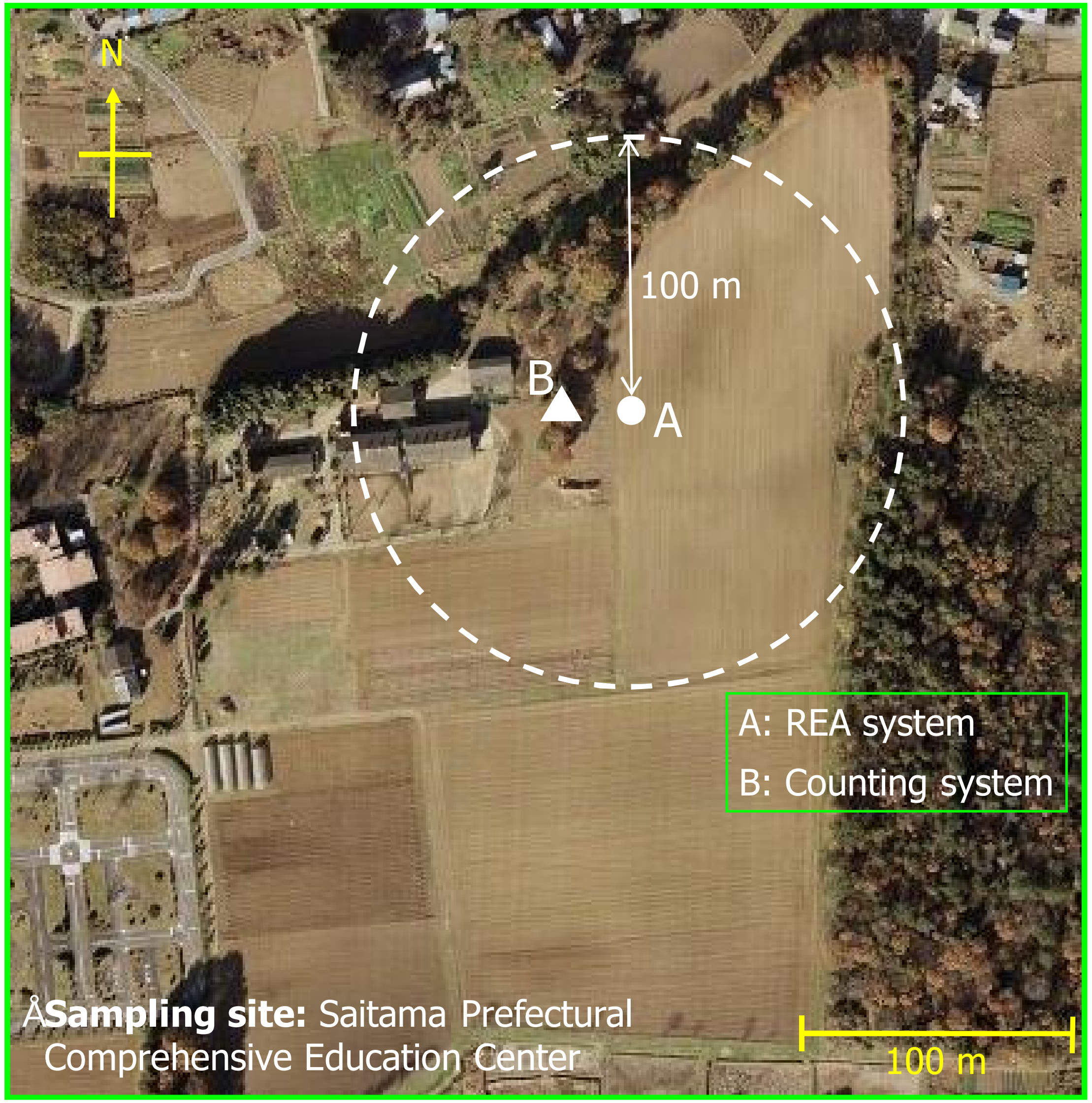
Field experiment



Measurement conditions

Table. Summary of experimental and weather conditions

Experimental	June	August
Measurement period (day)	2-4 (3 d)	24-27 (4 d)
Ground surface condition	Pasture	Pasture-patched soil
Height of pasture (m)	1	0.1-0.3
Rainfall event (daytime)	No	No
Temperature (°C) (daytime)	29 ± 2	39 ± 4
Relative humidity (%) (daytime)	33 ± 8	41 ± 11
Wind speed (m s^{-1}) (daytime)	2.0 ± 0.5	1.5 ± 0.3

Data screening of flux measurements of unattached ^{222}Rn decay products using the REA system

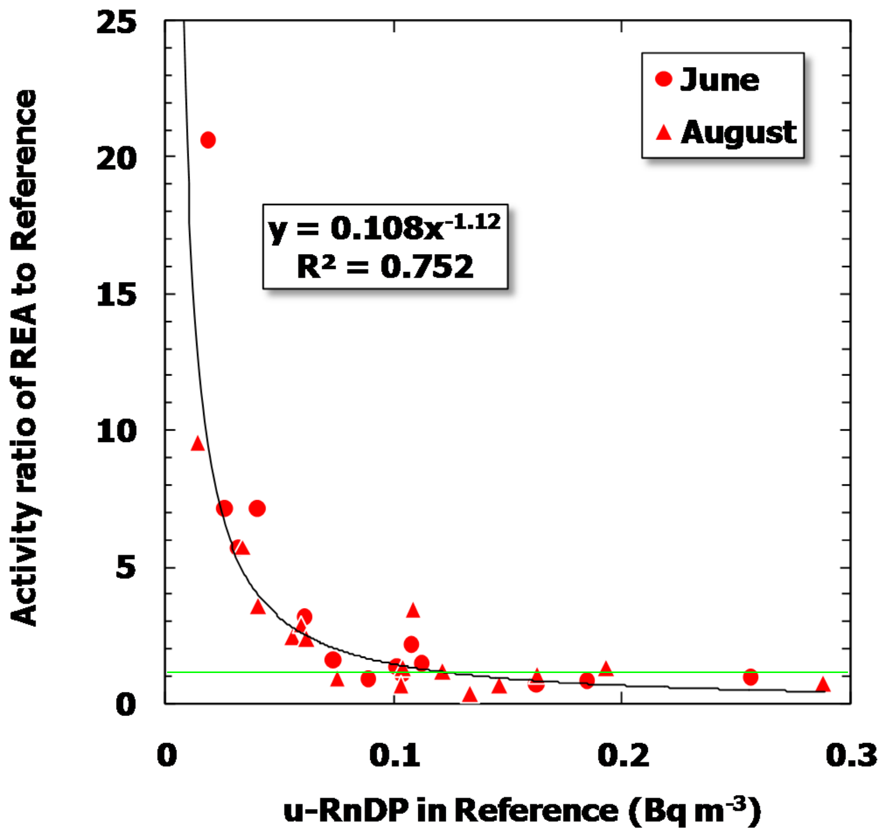


Fig. Activity concentration ratio of REA system to reference as a function of unattached ^{222}Rn decay products (u-RnDP) measured in field experiments

Increase of uncertainty in REA system with a decrease of unattached ^{222}Rn decay products concentration less than approximately 0.1 Bq m^{-3}

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

City, country	Measurement site (height in m)	U-RnDP (n) (Bq m^{-3})
Shanghai, China	Grass (1 m)	0.39 ± 0.23 (16)
Yanji, China	City (*ca.* 15 m)	1.09 ± 0.83 (15)
Hirosaki, Japan	City (*ca.* 15 m)	1.06 ± 0.41 (8)
Kumagaya, Japan	**Pasture (0.1-1m)**	**0.16 ± 0.08 (15)**

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

Table. Data screening of flux measurements

Data screening	Remaining data (%)	
	June	August
1. Wind direction	94	94
2. $C_{Ref} \geq 0.1$ Bq m^{-3}	38	53
The number of initial point	16	17

Results of flux measurements of unattached ^{222}Rn decay products using the REA system during the daytime

	Symbol	June		August	
		Emission	Deposition	Emission	Deposition
Flux ($\times 10^{-2}$ Bq m^{-2} s^{-1})	F	3.7 (2.6—4.9)	-2.4 (-4.8—0.3)	1.3 (0.4—2.0)	-4.4 (-8.2—-2.1)
Unattached RnDP concentration (Bq m^{-3})	C	0.13 (0.11—0.16)	0.15 (0.10—0.22)	0.13 (0.04—0.25)	0.23 (0.14—0.37)
Transfer velocity (cm s^{-1})	V_f	-27 (-31—-24)	18 (1.2—35)	-12 (-27—-4.3)	18 (16—22)
Number of data after the criteria (all data)		2 (16)	4 (16)	6 (17)	3 (17)
Radon concentration (Bq m^{-3})	Rn	4.4 ± 1.0		7.7 ± 1.6	

The flux (F) is calculated as follows:

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

b : empirical dimensionless coefficient

σ_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

Deposition surface (height in m)	Object substances	D_p (nm)	V_d (cm s^{-1})	z^* (m s^{-1})	n	Reference
Pasture (1)	**u-RnDP[a]**	**4[b]**	**18 (1.2－35)**	**0.2 (0.2－0.2)**	**4**	**This study (June)**
Pasture-patched soil (0.1-0.3)	**u-RnDP[a]**	**4[b]**	**18 (16－22)**	**0.1 (0.1－0.1)**	**3**	**This study (August)**
Semiarid desert covered with low grass (0.3)	u-RnDP[a]	2.7[b]	7.3 ± 2.5 (5－35)		22	Schery et al. (1998)

[a]Unattached ^{222}Rn decay product.

[b]Particle diameter for 50% penetration into wire mesh screens.

Conclusions

ÅThe 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.

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

ÅIn 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