# 環境放射能-測定の基礎と応用

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# 内容

（0. 放射性核種とは？　放射能とは？）

1. 環境中に存在する放射性核種

2. 環境放射能とは？　なぜ測定が必要か？

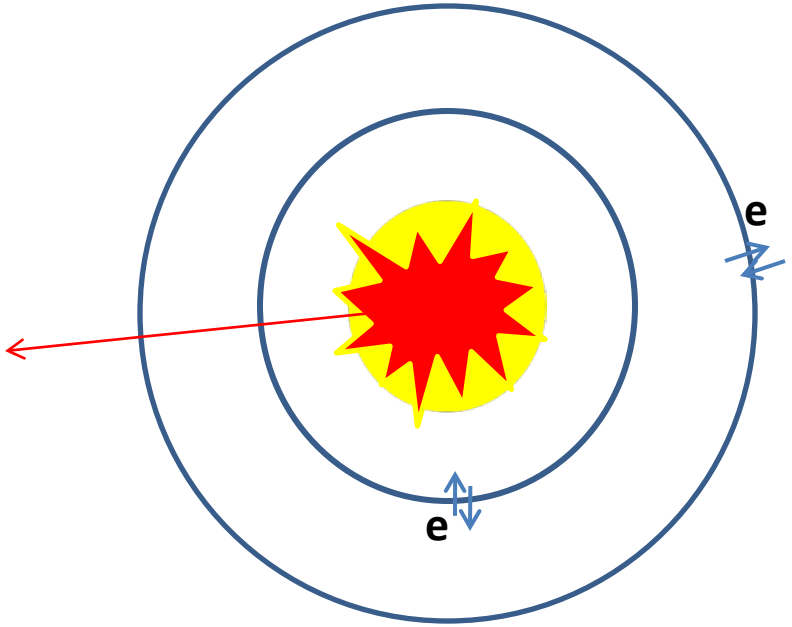
3. 放射能測定の基礎（サンプリングからデータ解析まで）

4. 放射能測定（モニタリング）の応用例

# 放射性核種とは？

不安定な（エネルギー状態の高い）原子核

放射線 (α, β, γ 線)




**壊変**

放射性核種のID

放出放射線　種類

エネルギー

放出の割合

半減期

# 放射能とは？

放射性核種が単位時間 (1s) に壊変する数
単位：ベクレル（Bq）

$$\frac{dN}{dt} = -\lambda N$$

放射能

N: 放射性核種の数（時間 t での）
t: 時間 (s)
λ: 壊変定数

# 環境中に存在する放射性核種

## 環境放射性核種とは？

起源により分類

- 天然放射性核種 (natural radionuclides)

  **岩石起源** (lithogenic radionuclides)

  ウラン ($^{238}$U)系列、トリウム ($^{232}$Th)系列、アクチニウム ($^{235}$U)系列、カリウム ($^{40}$K)

  **宇宙起源** (cosmogenic radionuclides)

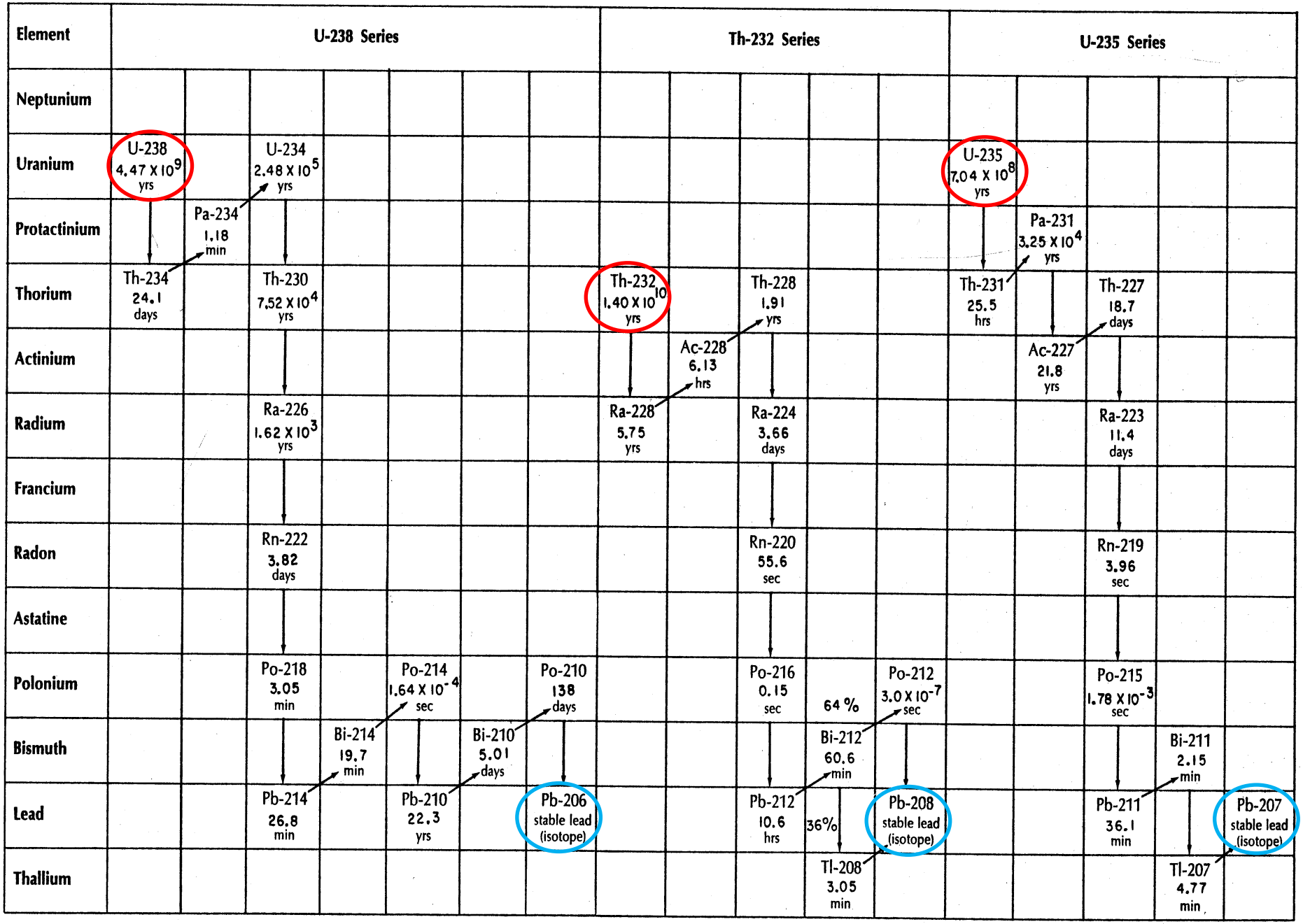
  トリチウム ($^{3}$H)、ベリリウム ($^{7}$B)、炭素 ($^{14}$C)、ナトリウム ($^{22}$Na)、アルミニウム ($^{26}$Al)、リン ($^{32}$P)、塩素 ($^{36}$Cl)、アルゴン ($^{37}$Ar)

- 人工放射性核種 (anthropogenic radionuclides)

  ストロンチウム ($^{90}$Sr)、ジルコニウム ($^{95}$Zr)、モリブデン ($^{99}$Mo)、テクネチウム ($^{99m}$Tc)、ルテニウム ($^{103}$Ru)、ヨウ素 ($^{131}$I)、セシウム ($^{137}$Cs)

  新規核種が環境中に放出される可能性あり！

# 系列を作る天然放射性核種 ($^{238}U$, $^{232}Th$, $^{235}U$)

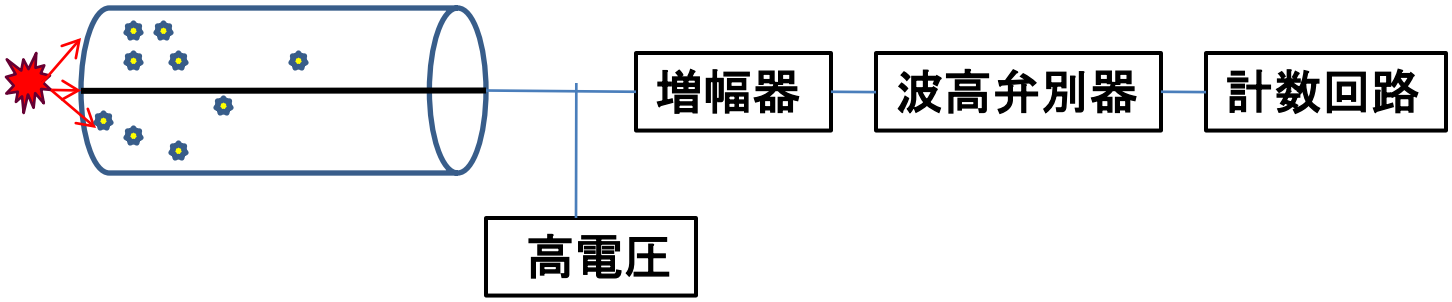


# 放射線の検出

**目に見えない！**

原理：　放射線と物質の相互作用
対象：　α 線、β 線、γ 線（中性子）
検出器：　気体、液体、固体

例　　ガイガーミュラー (GM)カウンタ



# 放射能の測定

例） ガンマ線スペクトロメトリ

検出器: 高純度ゲルマニウム半導体（同軸型、井戸型、平板型）

測定系: 高電圧電源、プレアンプ、リニアアンプ、マルチチャンネルアナライザ (4096 チャンネル)、データ解析ソフト

測定対象: ガンマ線

測定環境: 液体窒素 (77 K)、遮蔽体（古い鉛>数100ｙ）

校正:

エネルギー校正（$^{109}$Cd, $^{133}$Ba, $^{137}$Cs, $^{60}$Co, $^{40}$K）

効率校正（IAEA 標準物質）

# 高純度ゲルマニウム半導体検出装置

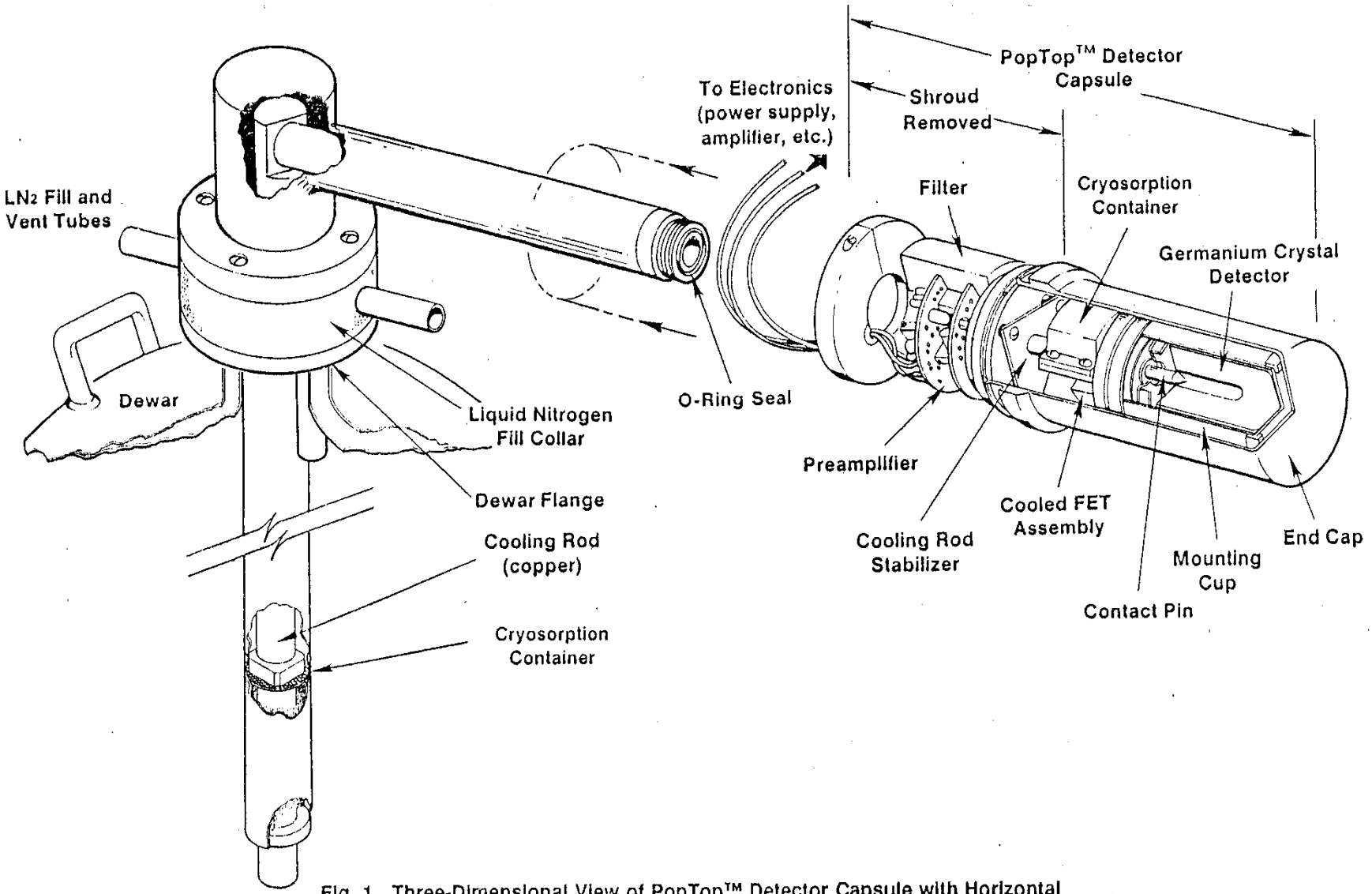



Fig. 1. Three-Dimensional View of PopTop™ Detector Capsule with Horizontal Dipstick Cryostat and 30-Liter Dewar.

# 環境放射能測定の意義と必要性

1. バックグラウンド放射能レベル

2. 長期モニタリングによる変動（日、季節）

3. 緊急時等における異常値の検出

4. 公衆に対する自然放射線による被ばく線量（内部被ばく、外部被ばく）評価

# 環境試料放射能分析　その前に。。。
## 試料採取－最も重要なファーストステップ

0. 情報収集（気候、地質、地形、生態系など）
1. フィールド用採取装置および器具の準備
2. サイト選択（バックグラウンドデータ）
3. 採取ポイントおよび試料数（試料の代表性）
4. 試料採取（採取時の気象状況等）
5. 現場での前処理と汚染防止策
6. 実験室への輸送
7. 試料の保存（温度・圧力・酸化還元・pH条件等）

# スクリーニングか精密測定か？

## スクリーニング

メリット：　簡便な測定法
迅速なデータ取得
一度に多くの試料を処理可能

## 精密測定

メリット：　高精度の測定値
高い信頼性

# 計数の統計

## 放射性核種の壊変は確率の現象である！

### 放射能測定の不確定さ

1. 対象核種の半減期が短い場合の試料中の放射能
2. 放射性核種の壊変はランダム現象

### 計数の統計学的処理が必要

ポアソン分布－ランダム過程の記述法。すべての事象の総数は統計学的に小さく、その事象の起こる確率が一定である場合に適用可能。

### ポアソン分布適用のための前提条件

- すべての放射性核種について、ある時間内に壊変する確率は一定。
- ある核種の壊変過程は、別の核種の壊変によって影響されない。
- 放射性核種の総数および計数時間は十分に長い。
- 対象核種の半減期は計数時間よりも十分長い。

# ポアソン分布

ある放射性核種についてある時間内 t にn壊変が起こる確率 $P_n$ は、

$$P(n) = \frac{e^{-\mu}(\mu)^n}{n!}$$

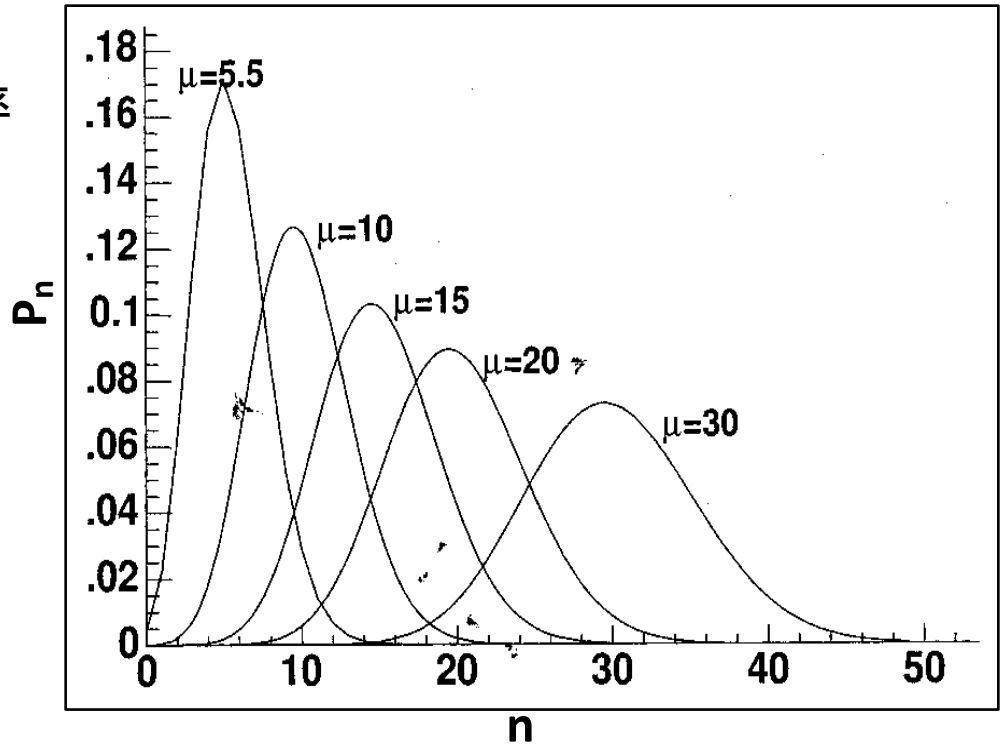
$P_n$ : 時間 t 内にn回壊変が起こる確率

μ : 時間 t の間に起こる壊変数 n の平均数 (μ = mean of n)

σ : 標準偏差

$$\mu = \sum_{n=0}^{\infty} nP(n)$$

$$\sigma^2 = \sum_{n=0}^{\infty} (n-\mu)^2 P(n) = \mu$$



# 標準物質

統一した測定体系の構築と相互比較可能なデータ蓄積のため

## IAEA（国際原子力機関）の役割

### 経緯）

標準物質の必要性　チェルノブイリ原子力発電所事故（1986年）
国際チェルノブイリプロジェクト（1989） 周辺地域の汚染状況把握や地域住民の健康被害評価を目的として

### 必要なリソース）

測定装置（定期的に校正）、熟練した技術者、標準物質、標準分析法、”in situ”分析、連続モニタリング

### 起こりうる問題）

マトリックス効果、測定下限、試料の代表性、前処理（濃縮と分離）、妨害物質の除去

# 低レベル放射能測定

## バックグラウンド計数の低減と検出感度向上への努力

### 低レベルとは？

β線、γ線測定 :　≤ 1 cpm
α線測定 :　≤ 0.1 cpm

### 試料の必要量

| | |
|---|---|
| 土壌・堆積物 | 数10～100 g |
| 植物・海産物 | 1～数 kg |
| 海水 | 数 10～100 ℓ |
| 大気・浮遊塵 | 数千～数万 $m^3$ |

### 前処理 - 濃縮、抽出、灰化等

# 得られたデータのゆくえ

- 放射線防護の立場から　　被ばく線量 (Sv)
  （実効線量係数）
- 生態学の立場から　　生物サイクル
  （濃縮係数）
- 地球科学の立場から　　地球科学的物質循環
  （分配定数、フラックス）
  自然災害予知
  年代推定

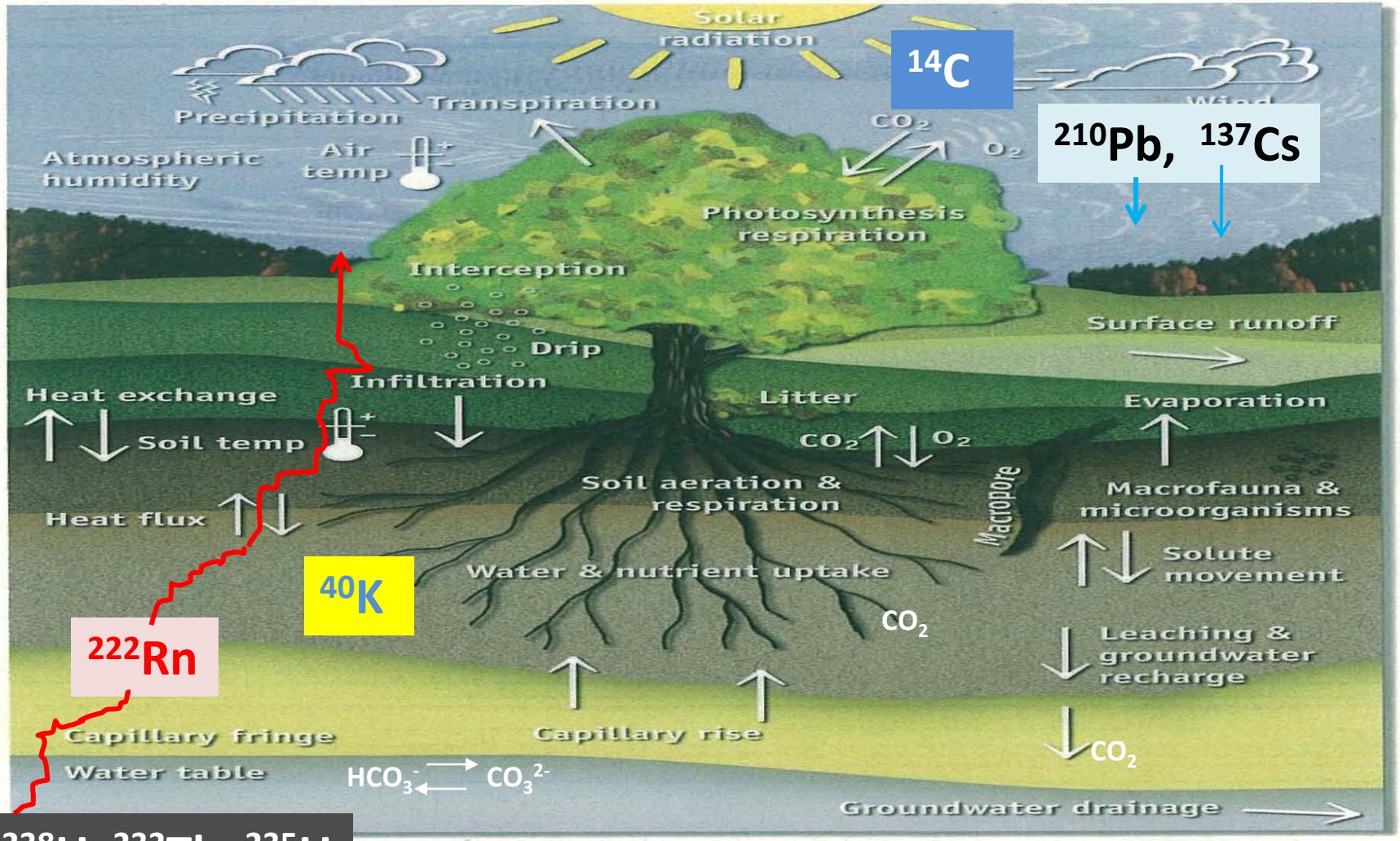
# 環境放射能測定（モニタリング）の応用例

森林土壌中のラドン ($^{222}Rn$) 、$CO_2$
およびその炭素同位体 ($\Delta^{14}C$, $\delta^{13}C$) 測定

森林土壌中の環境放射性核種 ($^{40}K$, $^{226}Ra$, $^{137}Cs$, $^{210}Pb$) の深度分布

# Mass and energy transfer in the atmosphere-soil system

## Where various environmental radionuclides are present




Cited from Hillel D. "Environmental Soil Physics, Academic Press (1998)

# Simultaneous monitoring of $^{222}Rn$ and $CO_2$ in soil air under a cool-temperate deciduous stand

Ryoko Fujiyoshi[1)], Yukihide Haraki[1)], Hironori kikuma[1)], Takashi Sumiyoshi[1)],Hikaru Amano[2)], Kobal Ivan[3)], Janja Vaupotič[3)]

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Oslo, Aug. 6 - 14, 2008

# Why carbon dioxide is to be monitored as well as $^{222}Rn$ in soil air?

## **Previous Results** 1 **Fujiyoshi et al., Environ. Geochem. Health 27, 539-547 (2005)**

1. **Soil radon ($^{222}$Rn)** was measured temporally with a scintillation Lucas cell in semi-natural woods on the campus of Hokkaido University, Japan. Its level was generally **low** in Sapporo where the surface geology is Quaternary deposit of alluvial sediments from the nearby rivers(Kotoni and Hassamu).

2. Changing air-filled porosity accompanied with **meteorological phenomena** may affects soil radon level and its variability.

3. **Sudden increase in $^{222}$Rn concentration** was observed, which appeared coincidentally with a big **earthquake** (M.8) occurred on Sep. 26, 2003 off the Coast of Hidaka in Hokkaido.

# Previous results 2 Fujiyoshi, Vaupotic & Kobal, Sci. Total Environ. 370, 224-234(2006)

1. **Continuous $^{222}$Rn monitoring** in soil air with a Barasol Probe (Algade, France) from Nov. 2004 to Mar. 2005 and from Sep.2005 to Mar.2006 in the same site investigated in the previous study.
2. Soil humidity and barometric pressure just above the ground surface were measured continuously.
3. Considering factors affecting variability of soil radon level, especially **meteorological ones**: **Temperature** was a dominant parameter under high atmospheric pressure during three seasons except in winter.
4. Soil radon level had been kept low at constant temperature (0°C) under thick snow cover (1m) in winter. **There appeared $^{222}$Rn peaks sporadically** which may be accompanied by changing **pressure in soil air** at a depth of **30 cm**.

**What's going on such a sudden increase in $^{222}Rn$ concentration in soil air under a thick snow cover in winter?**

**Trying to find source place(s) of $^{222}Rn$ in soil air**

**together with another "natural tracer", if any**

**Focusing on $CO_2$ and its carbon isotopes ($^{12}C$, $^{13}C$, $^{14}C$)**

**Carbon dioxide is much more complicated in behavior for tracing soil air due to various factors including biological activity!**

**In spite of that, $CO_2$ is still important to trace because of its ability as a carrier gas of radon**

**Carbon isotopes ($^{12}C$, $^{13}C$, $^{14}C$) of $CO_2$ could be used to estimate its source(s), especially in winter when most biological activity decreases compared with that in summer.**

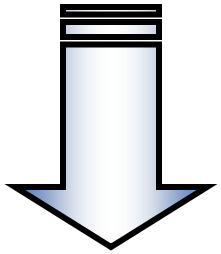
**AND some more information on $^{222}Rn$, too??**

# Outlines of this study

Continuous monitoring of $^{222}Rn$ and $CO_2$ in soil air in the semi-natural deciduous woods

Carbon isotope ($^{12}C$, $^{13}C$, $^{14}C$) analyses of soil gas $CO_2$

Monitoring of soil properties (humidity, density, organic content)



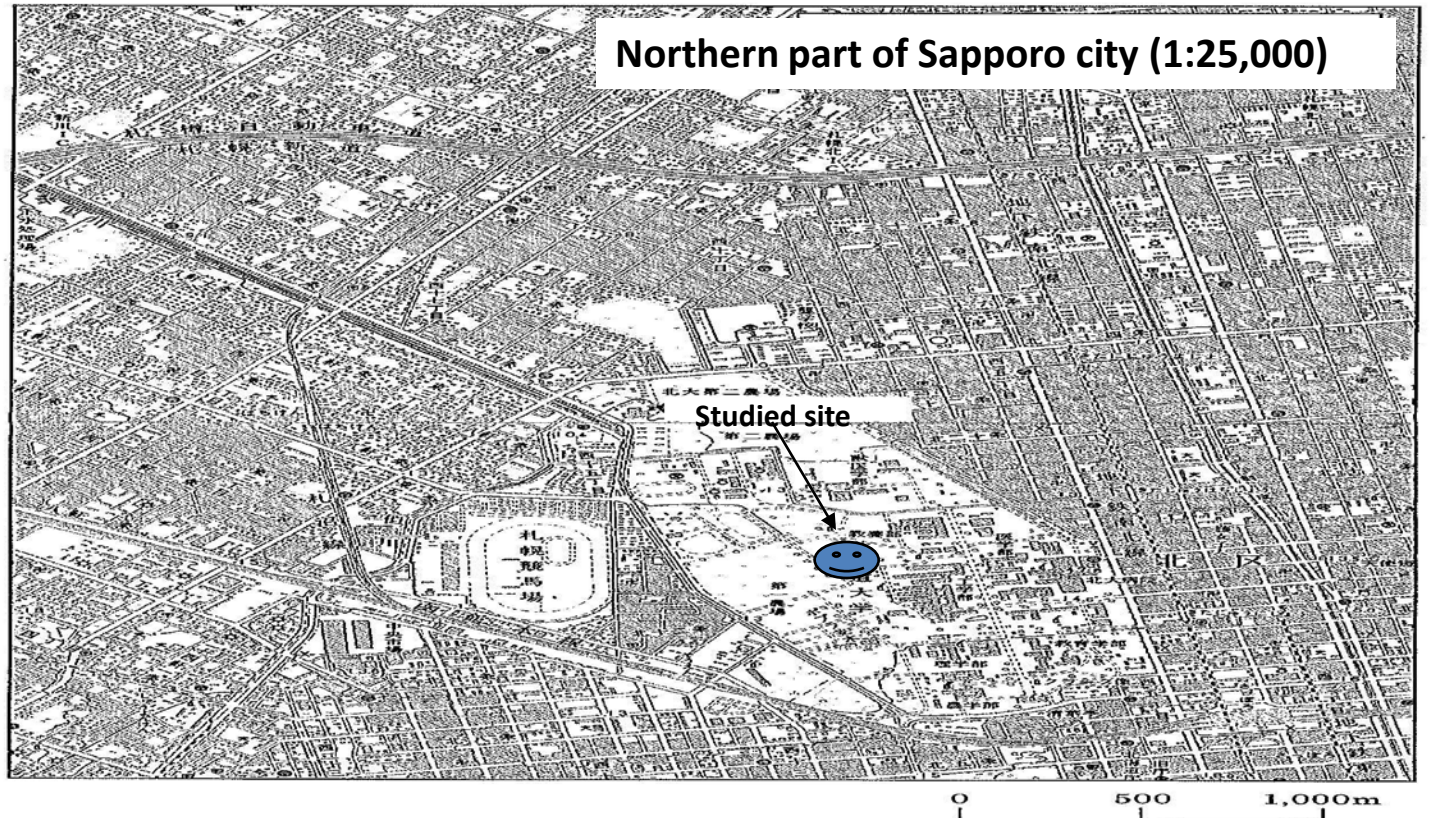
**Origin of Soil Air Components Using $^{222}Rn$ and $CO_2$ ($^{12}C$, $^{13}C$, $^{14}C$) as Natural Tracers**

# Location map of the site on the campus of Hokkaido University, Sapporo, Japan

**Climate: Annual mean temperature, 6-9˚C; Annual mean precipitation, 800-1600 mm**

**Geology: Alluvial sediment from Toyohira and Hassamu rivers**

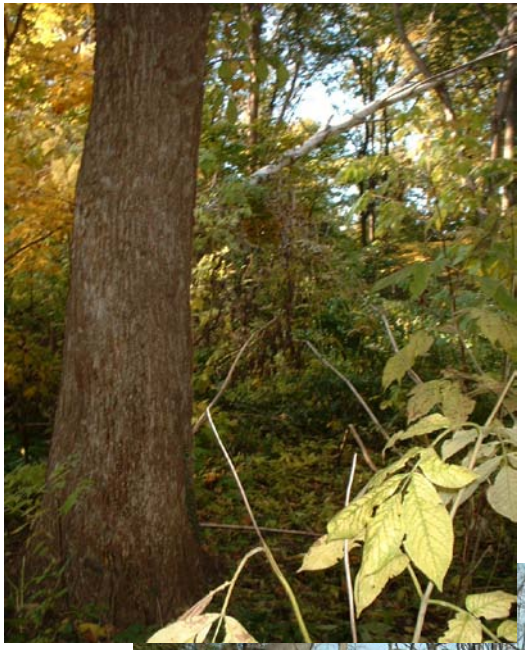
**Stand: cool temperate mixed deciduous stand**



# Seasonal view of the site on the campus of Hokkaido University

Early May

July



October

Early April

# *Methods*

## Soil sampling and preliminary task

- Point setting and drilling holes (30, and 100 cm in depth)
- Collecting soil samples from the surface to a depth of 50 cm
- Determining dry density, moisture and organic contents

## Experimental

- Elemental analyses of the soil using an X-ray fluorescence spectrometer (JSX-3220, JEOL)
- Gamma spectrometry with HPGe detectors (EG&G Ortec,USA)

  Nuclides of interest : $^{40}K$, $^{226}Ra$, $^{137}Cs$, $^{210}Pb$
- Determination of dissolved ions ($K^+$, $Na^+$, $Cl^-$ etc.) in soil solution using ion selective electrodes
- Sorption experiments with radiotracers ($^{54}Mn$, $^{65}Zn$, $^{210}Pb$)

# Continuous monitoring of $^{222}Rn$ activity concentration in soil air at a depth of 80cm

**Probe: Barasol (Algade, France)**

**Detector: Solid silicon detector (Depleted depth, 100 μm; Useful area, 450 $mm^2$; BG, < $7 \times 10^{-4}$ cpm)**

**Measurement time interval: 60 min.**

**Interval of data collection: 1-4 months**

**Passive technique**

# Continuous monitoring of $CO_2$ (humidity and temperature) in soil air at 30 and 100 cm in depth

**Probe: IAQ probe (TESTO-435, TESTO, Germany)**

**Detection: Non-dispersive IR absorption**

**Measurable concentration range: 0 to10,000 ppm**

**Resolution: 1 ppm**

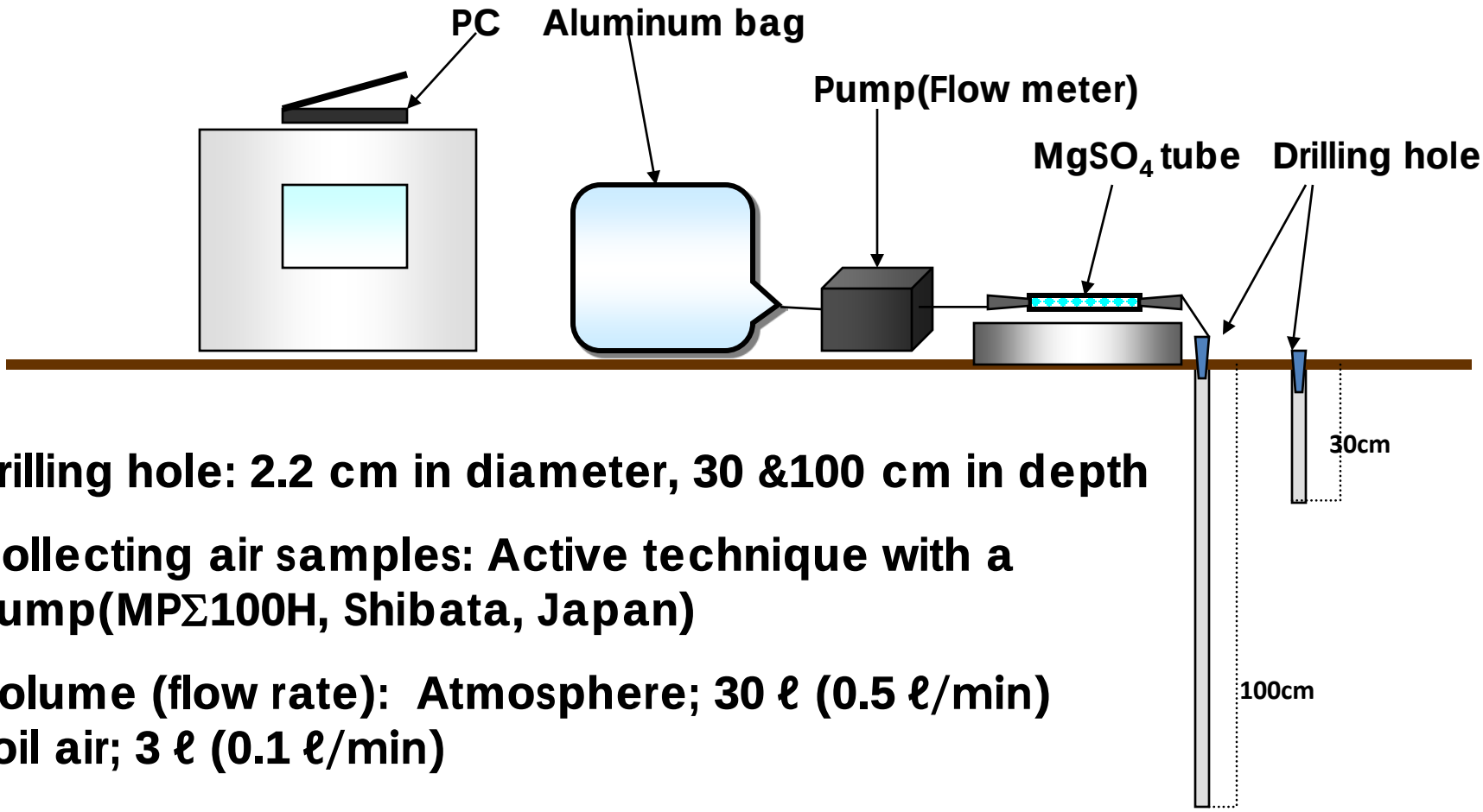
**Measurement time interval: 10 min**

**Interval of data collection: 24 hrs**

**Passive technique**

# Carbon isotopic analysis of $CO_2$ in soil air by AMS

## 1. Soil air sampling



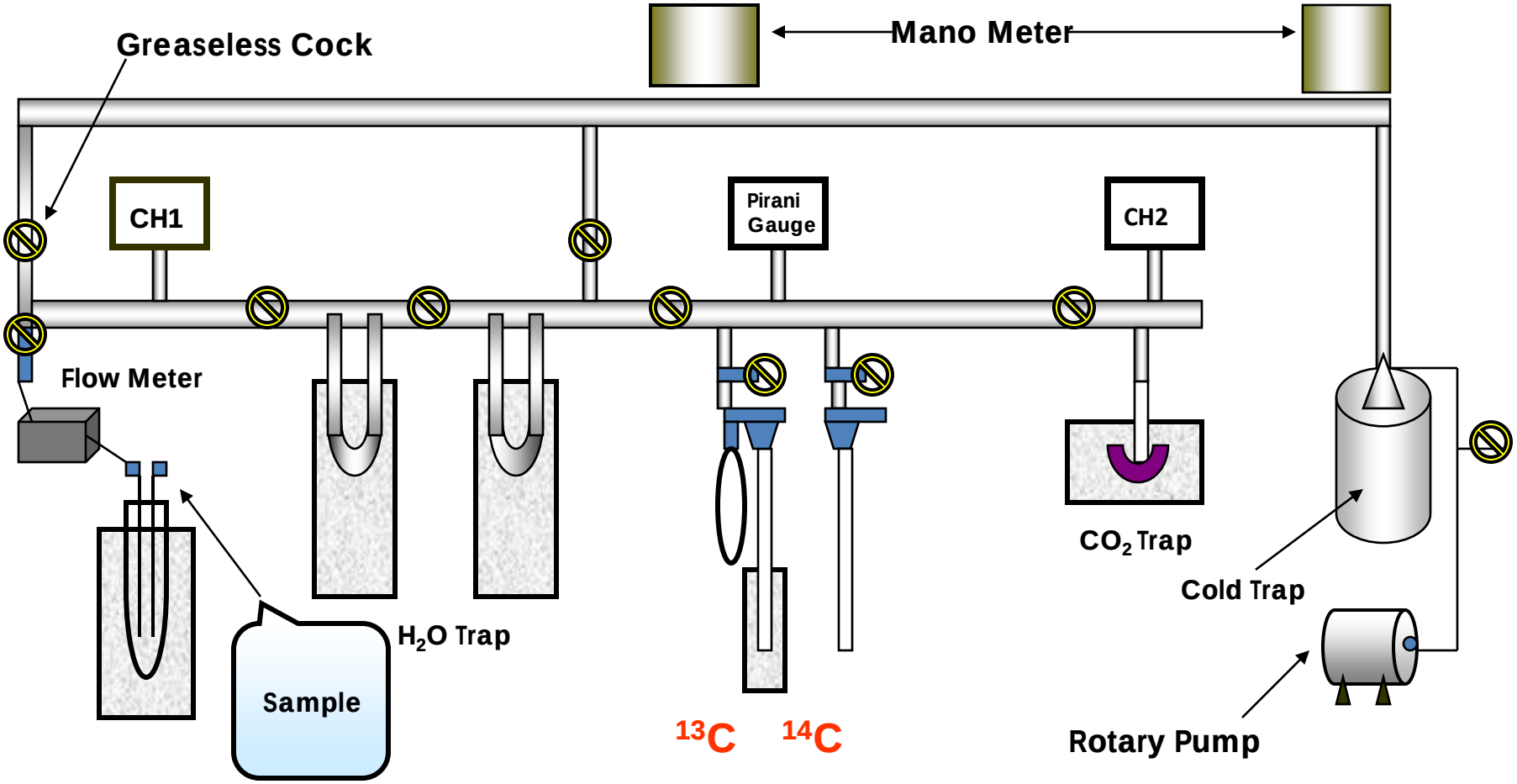

**Drilling hole: 2.2 cm in diameter, 30 &100 cm in depth**

**Collecting air samples: Active technique with a pump(MPΣ100H, Shibata, Japan)**

**Volume (flow rate): Atmosphere; 30 ℓ (0.5 ℓ/min)**
**Soil air; 3 ℓ (0.1 ℓ/min)**

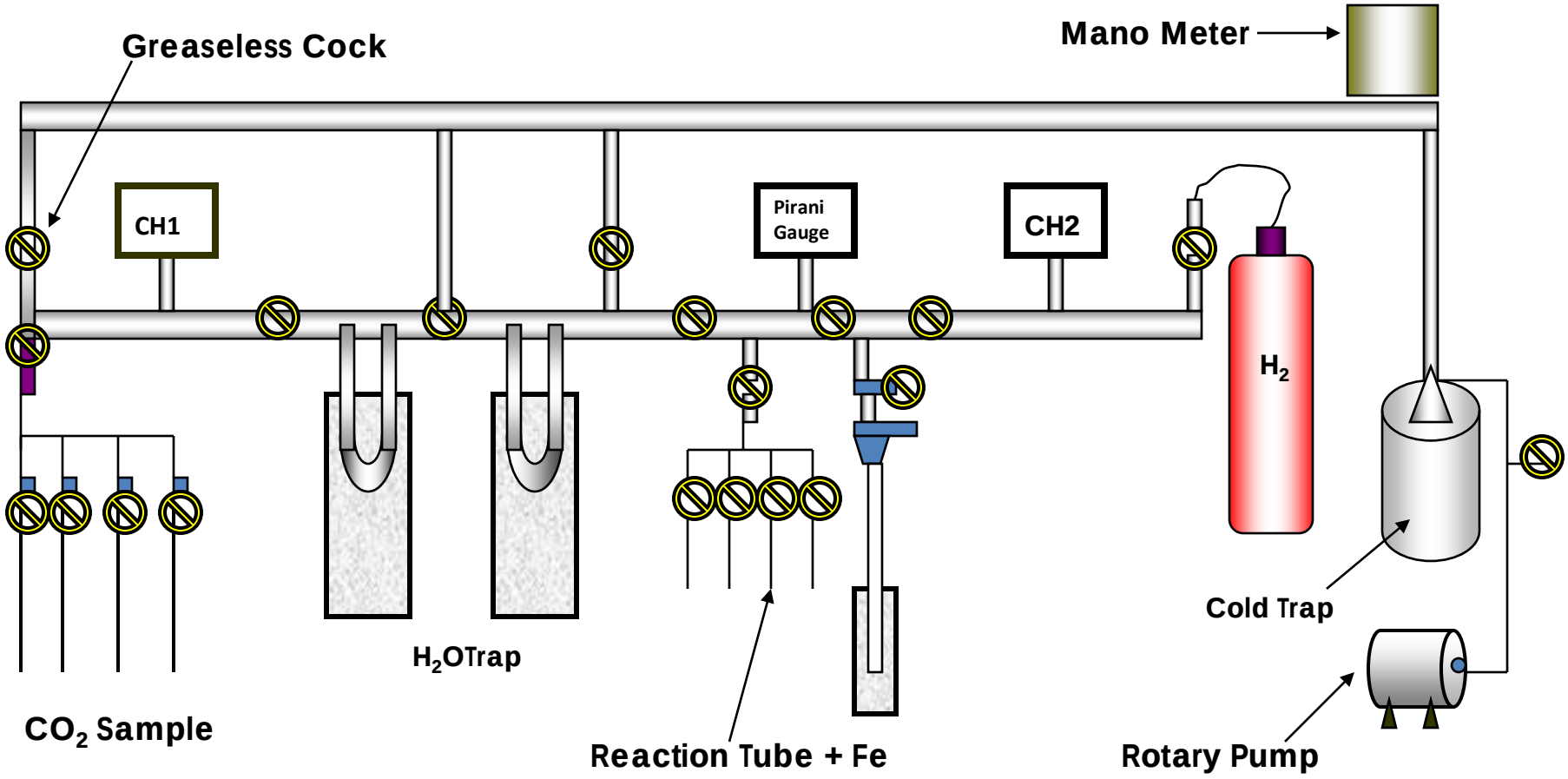
# Carbon isotopic analysis of $CO_2$ in soil air by AMS

## 2. Pretreatment of the samples(dehydration, purification)

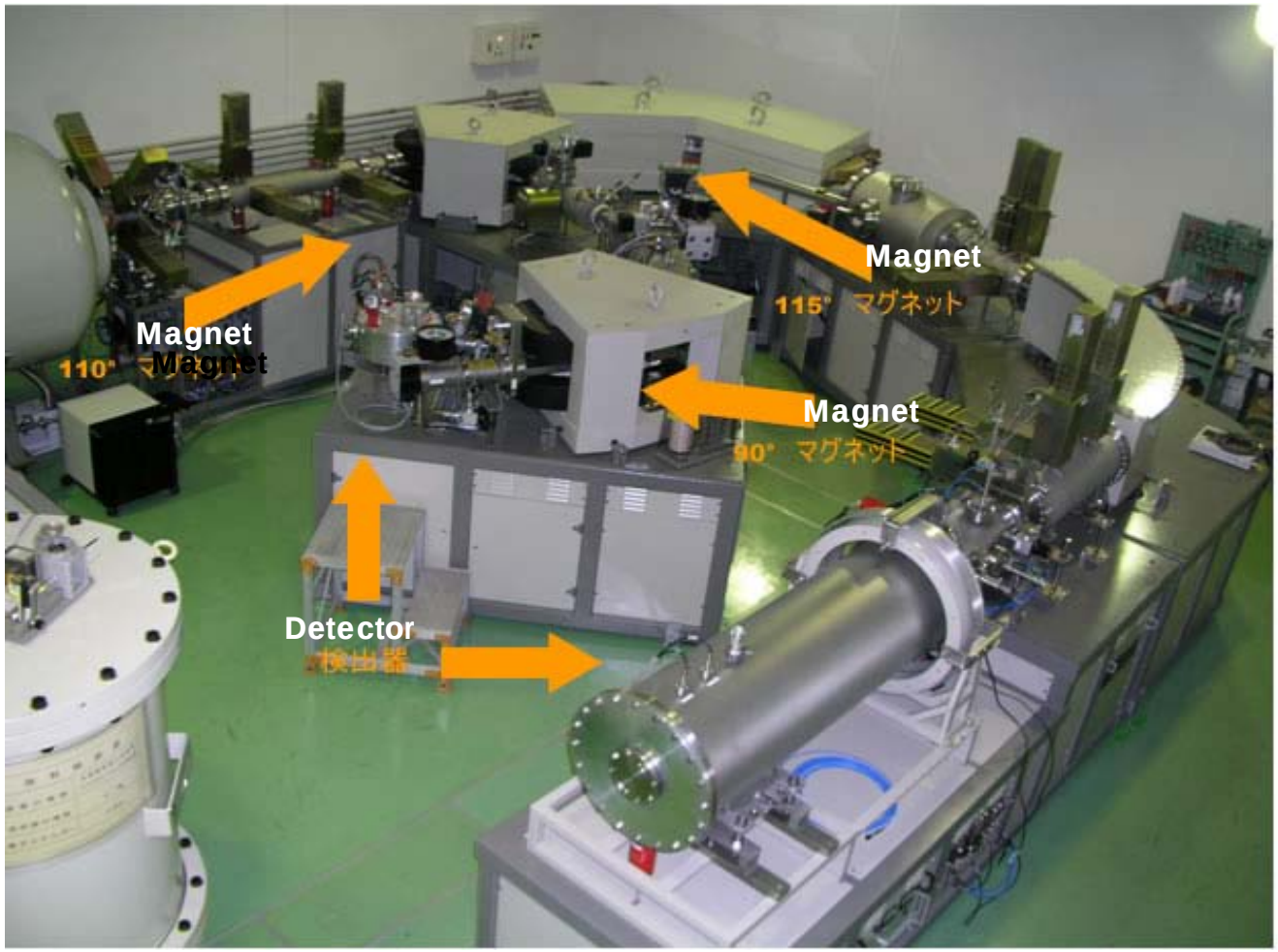


# Carbon isotopic analysis of $CO_2$ in soil air by AMS

### 3. Pretreatment of the samples(Reduction to graphite)



# Accelerator mass spectrometer (AMS) installed at Mutsu Office, Japan Atomic Energy Agency (JAEA), Japan




View of the detection system

# Carbon isotopic ratio ($\delta^{13}C$, $\Delta^{14}C$)

The amount of carbon isotopes ($^{13}C$ and $^{14}C$) in geochemical samples is represented by isotopic ratio as follows:

$$\delta^{13}C(‰) = (R_X/R_{st} - 1) \times 1000$$

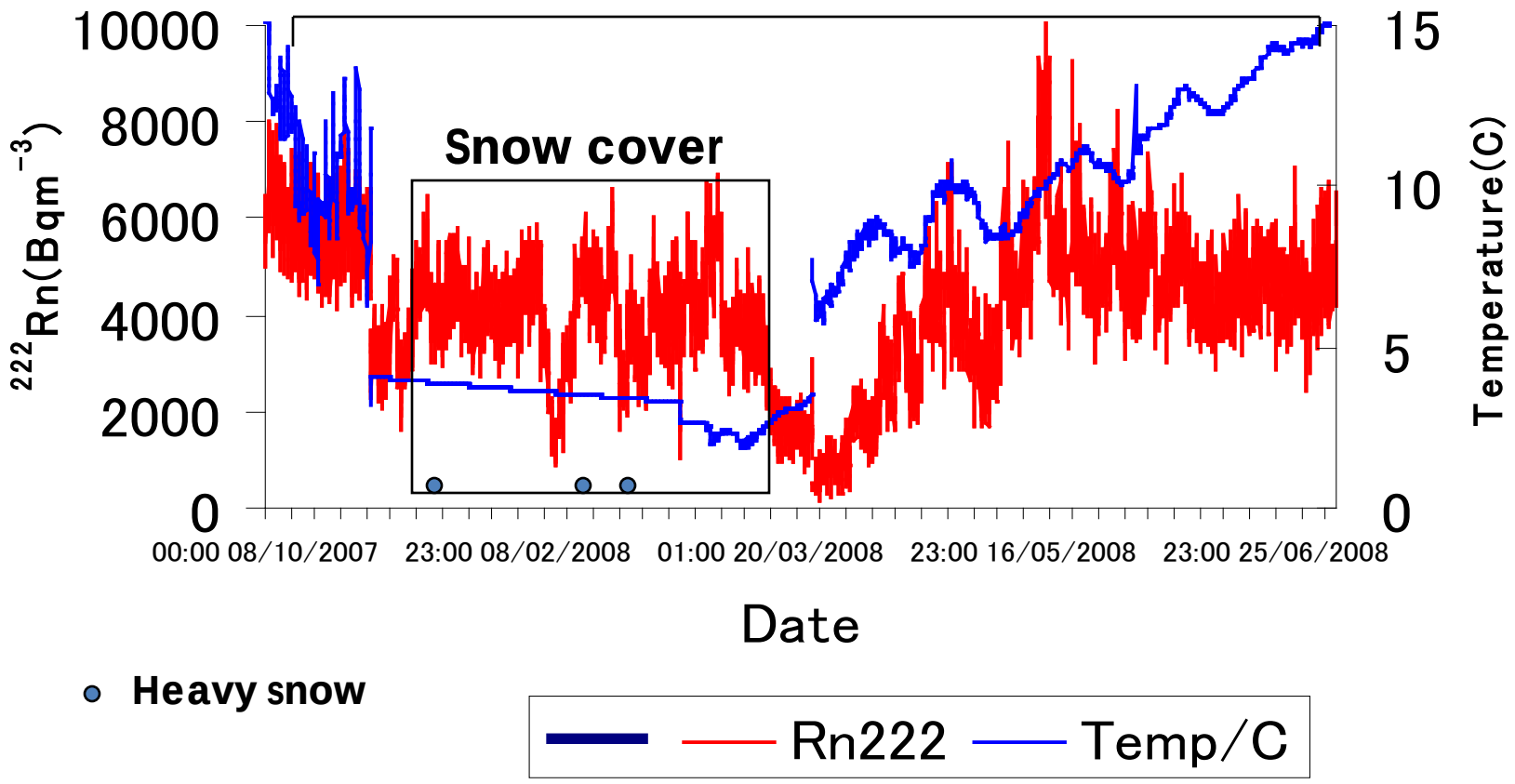
$$\Delta^{14}C\ (‰) = (R_X/R_{st} - 1) \times 1000$$

$R_X$: isotopic ratio of a sample ($^{13}C/\ ^{12}C$ or $^{14}C/\ ^{12}C$ )

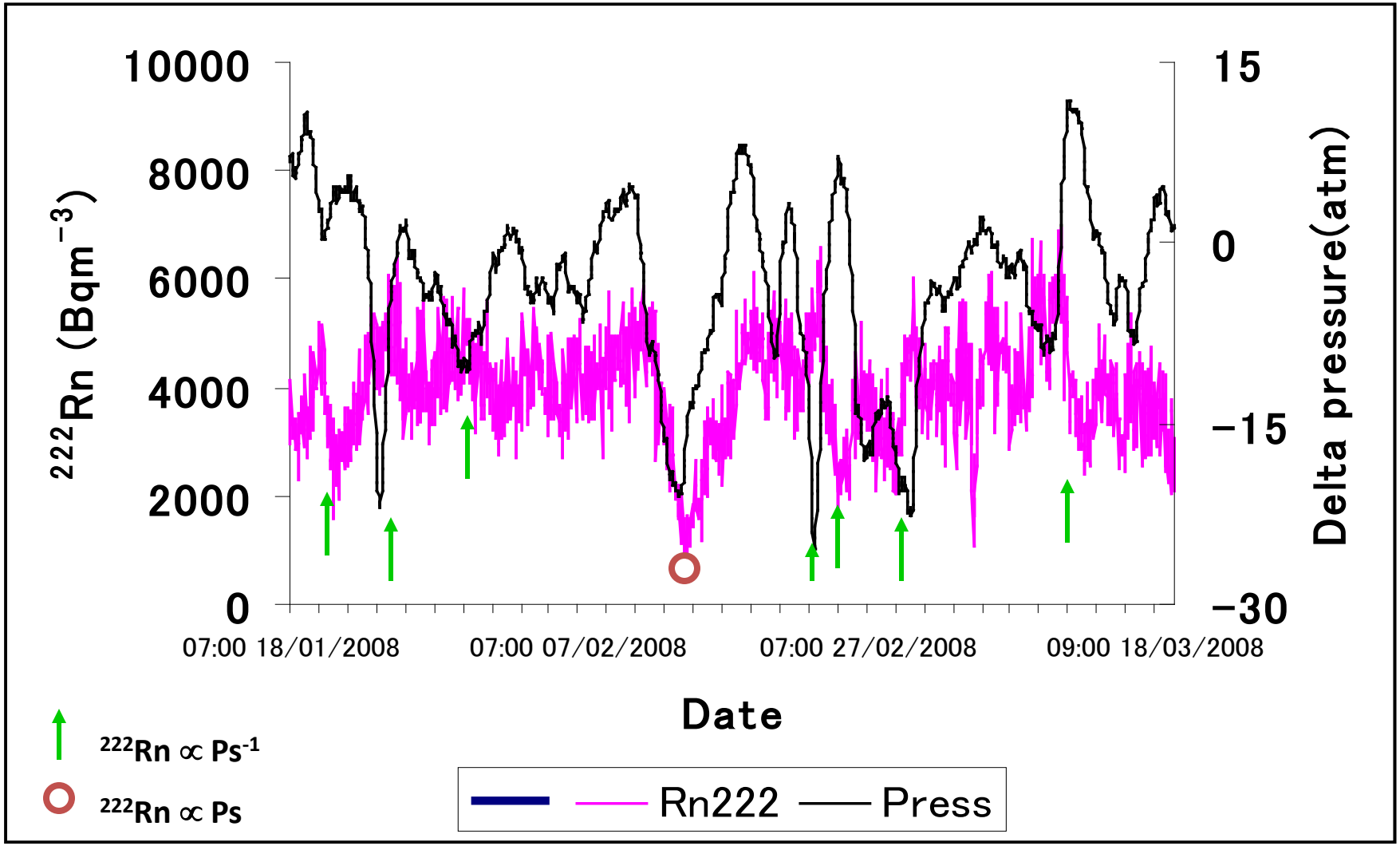
$R_{st}$: isotopic ratio of a standard

Standard materials: $^{13}C$ (belemnite carbonate from the Pee Dee formation, South Carolina, USA), $^{14}C$ (Oxalic acid distributed by National Institute of Standards and Technology, NIST)

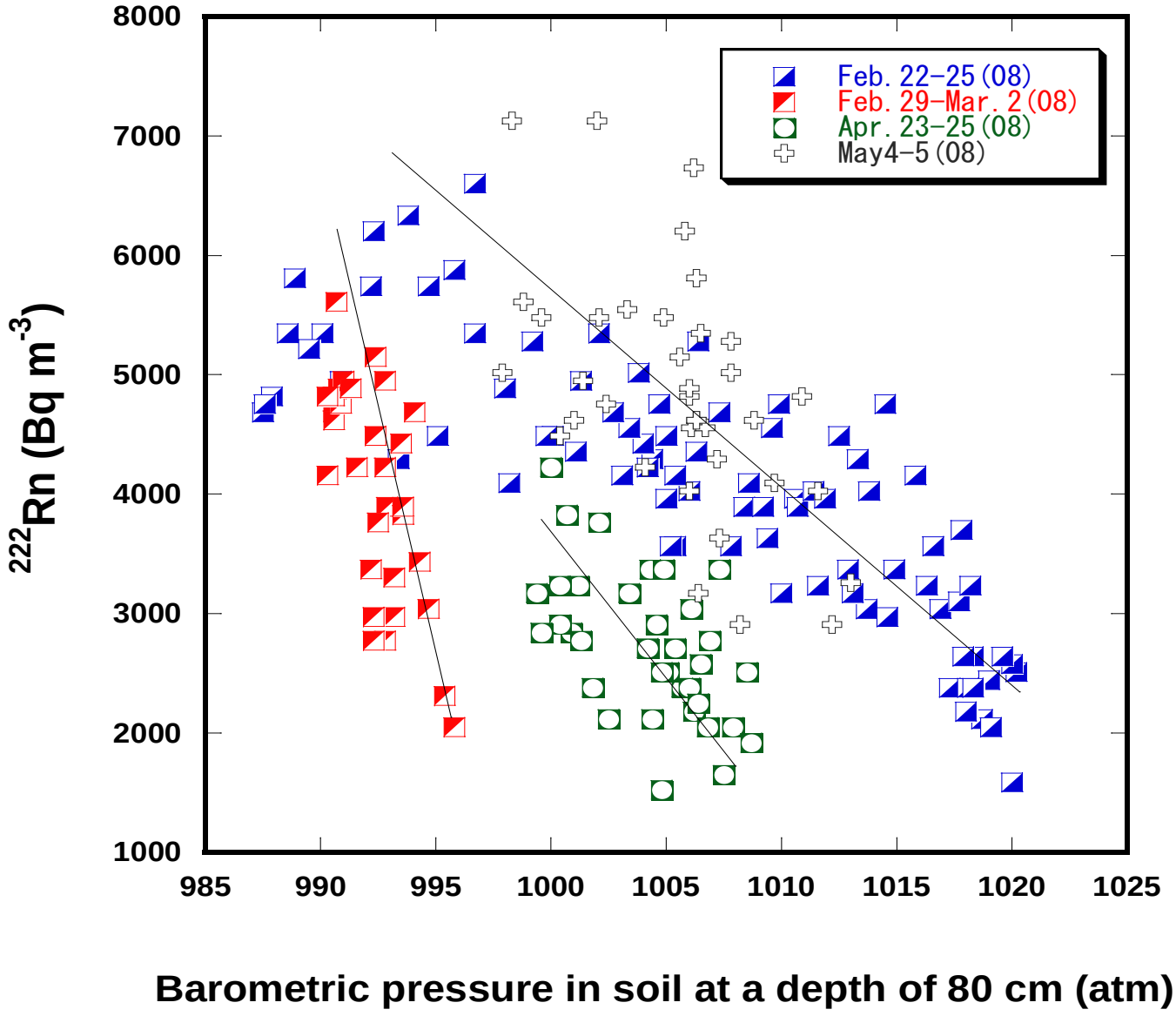
## *Result 1($^{222}Rn$)*



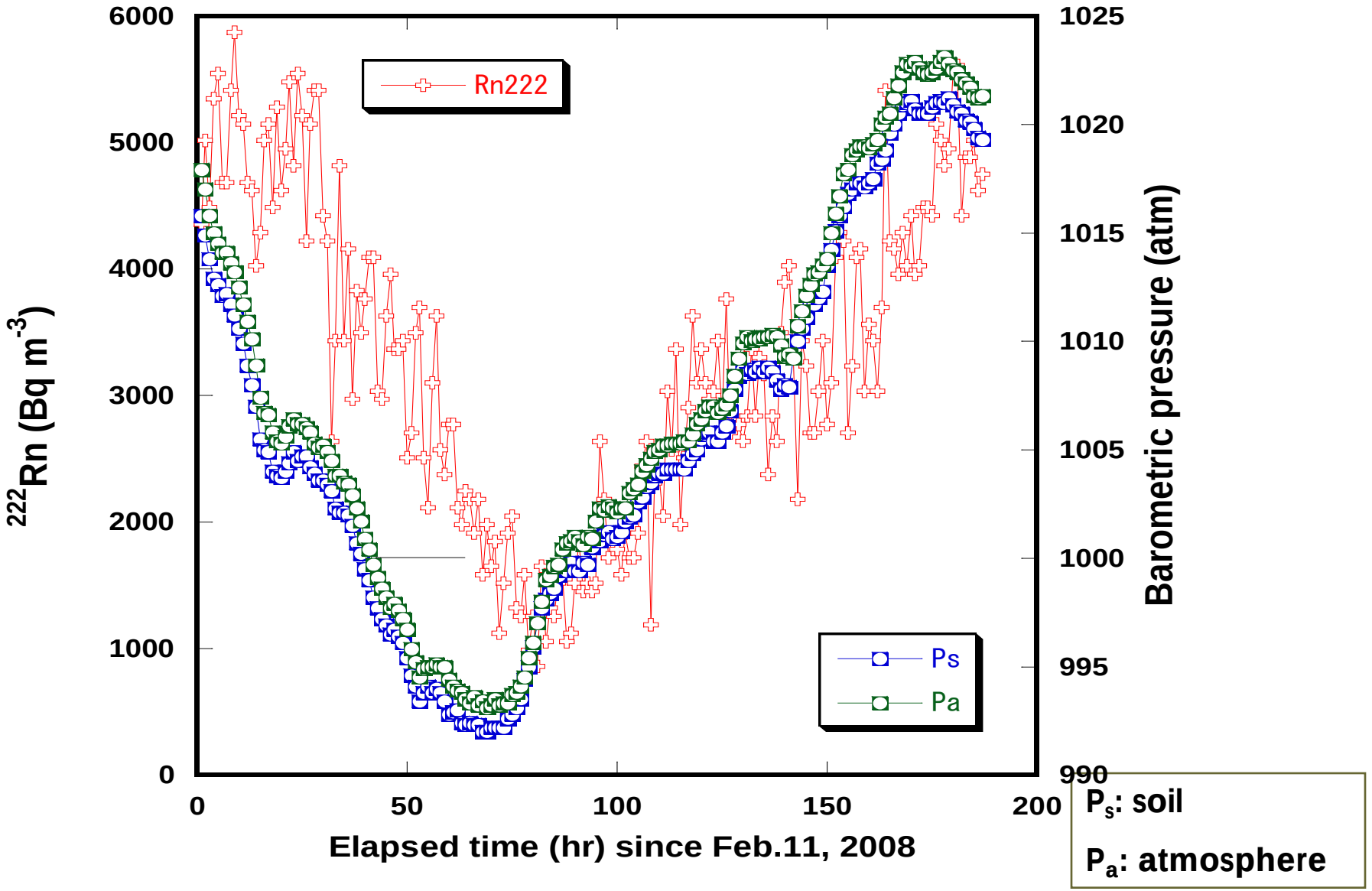
# Time series plots of $^{222}Rn$ activity concentration and barometric pressure in soil at a depth of 80 cm during lingering snow in winter



# Increase in $^{222}$Rn activity concentration with decreasing pressure in soil at a depth of 80 cm



# Change in $^{222}Rn$ activity concentration with barometric pressure in the atmosphere and in soil at a depth of 80 cm



# Comparing sets of $^{222}Rn$ data obtained in winter at different depths (30 and 80 cm)

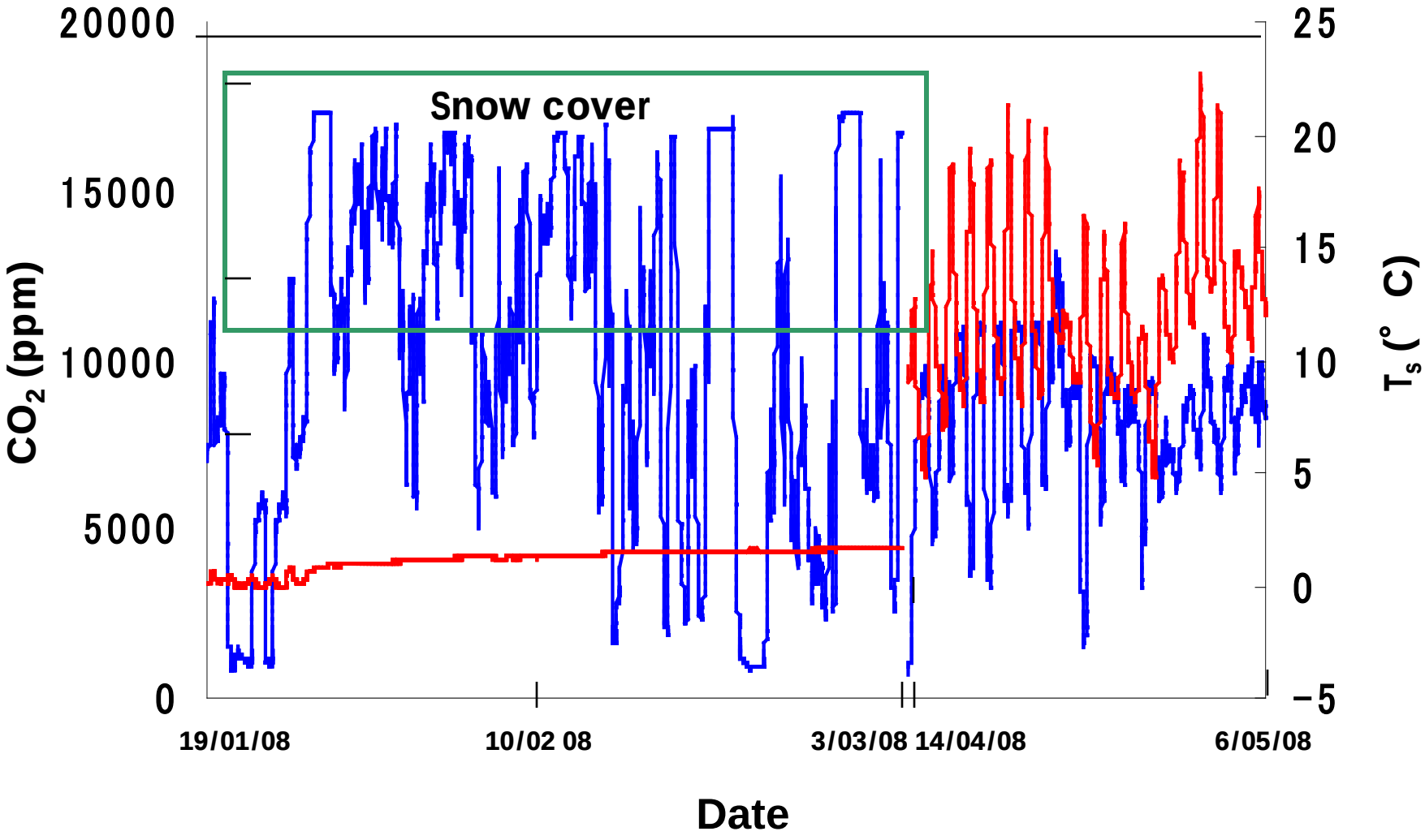
1. Activity concentration of $^{222}Rn$ in soil air was consistently low (1000~1500 Bq $m^{-3}$) at a depth of 30 cm where the temperature had been kept constant about 0 °C. The values were 4000~5000 Bq $m^{-3}$ at a depth of 80 cm with mean temperature of 3 °C.

2. Soil radon level was varied correspondingly with changing barometric pressure due to low pressure region passing through the site several times in whole winter in 2007-2008 (at a depth of 80 cm).

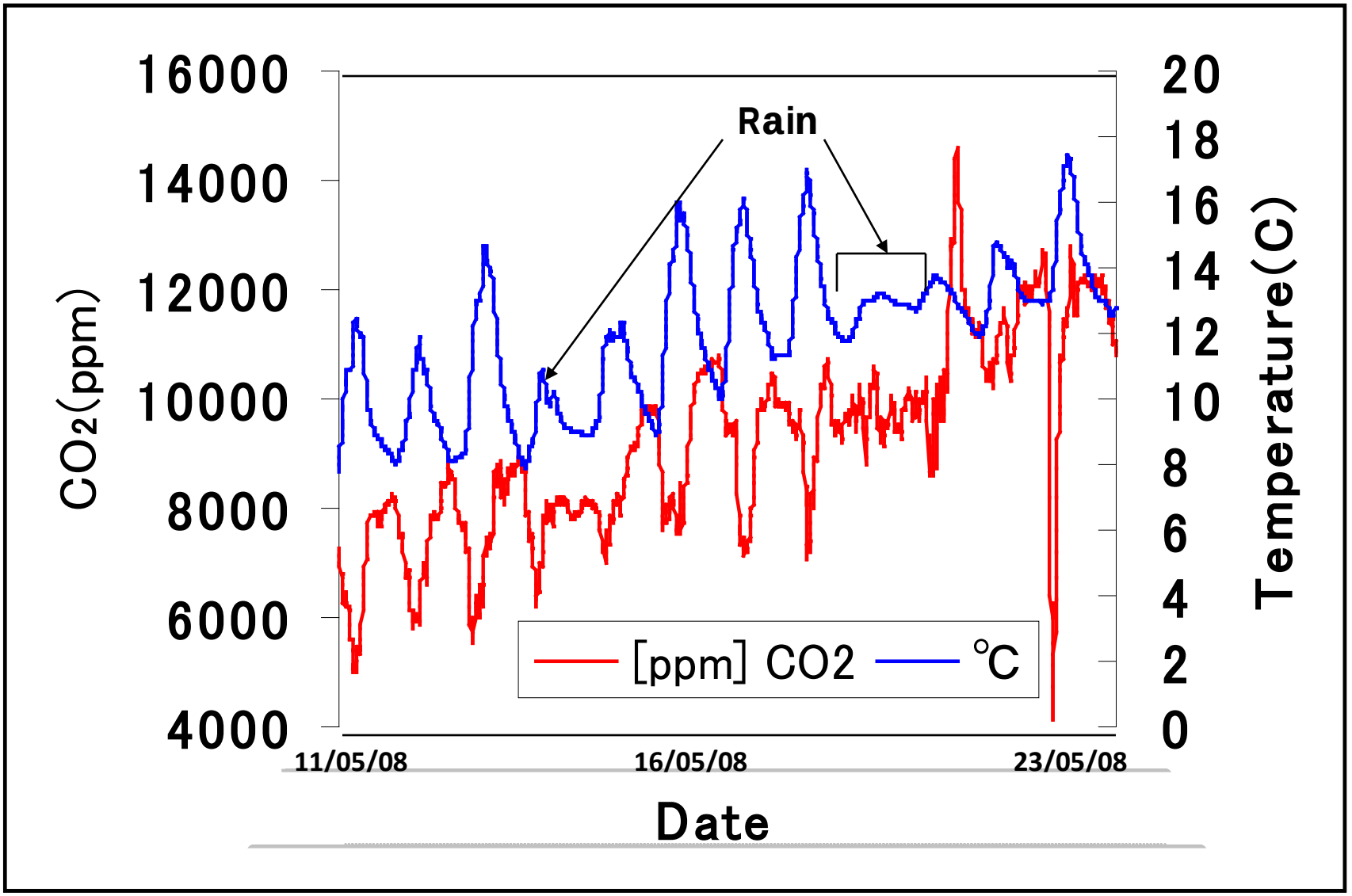
3. Sudden increase in $^{222}Rn$ activity concentration observed in 2004 did not appear this time, whereas $^{222}Rn$ level decreased and then was recovered slowly depending on unusual weather conditions.

Result 2 ($CO_2$)

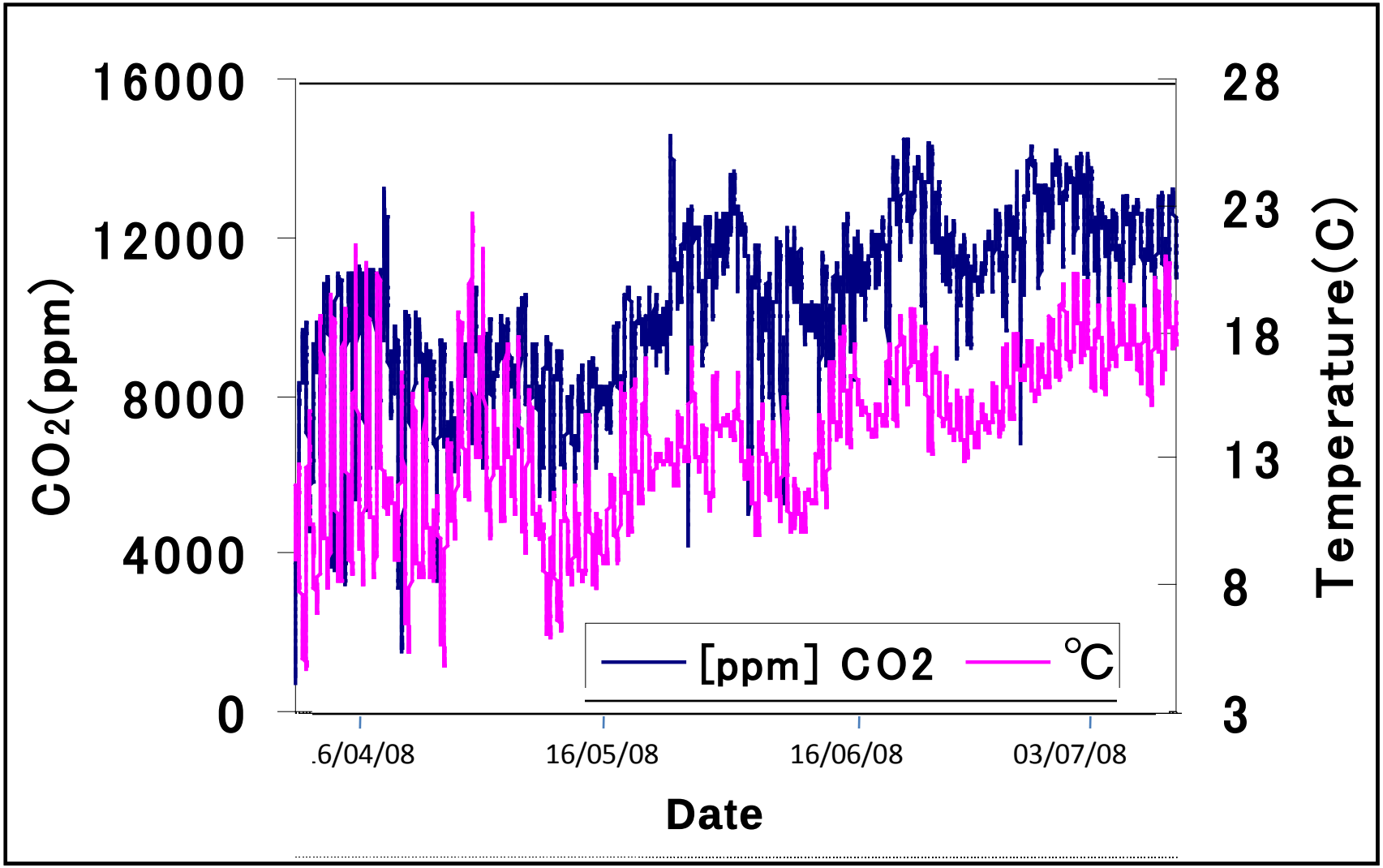
# Time series plots of $CO_2$ concentration and temperature in soil air at a depth of 100 cm from Jan. to May in 2008



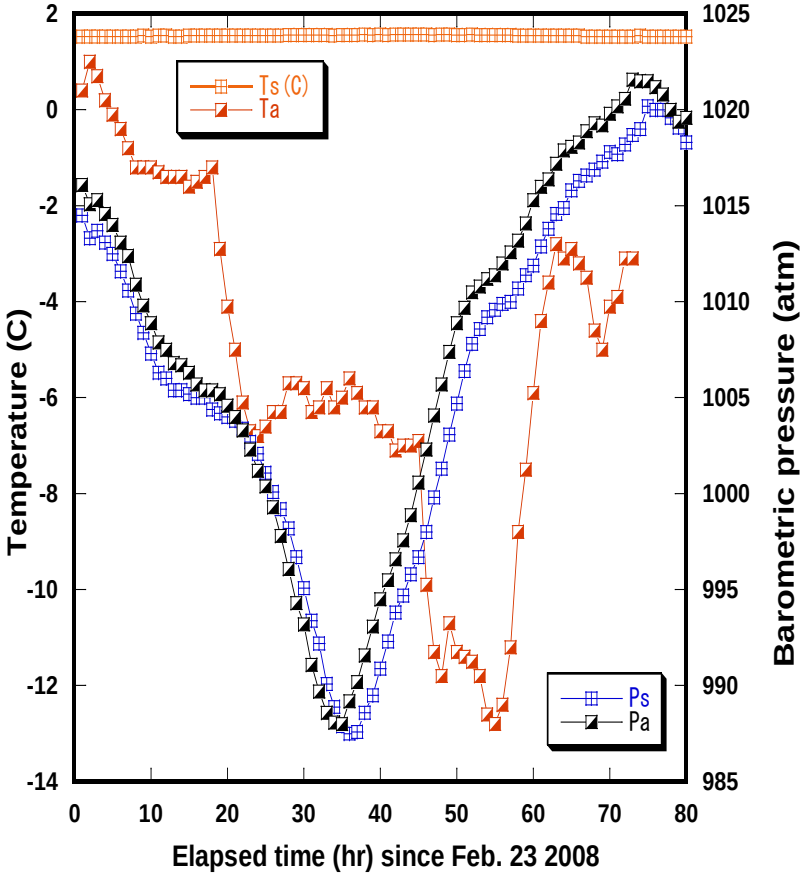
# Daily patterns of $CO_2$ (ppm) and temperature in soil air at a depth of 100 cm in May 2008

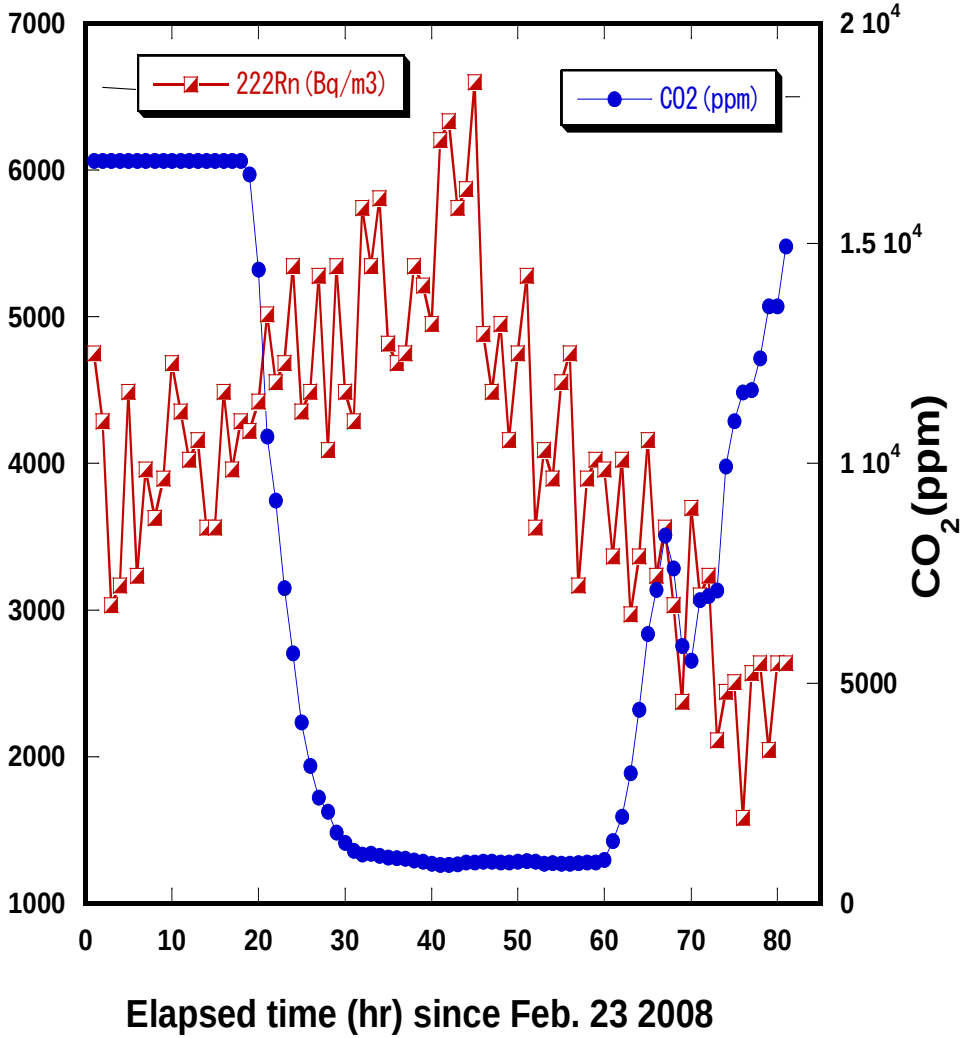


# Time series plots of $CO_2$ concentration and temperature in soil air at a depth of 100 cm from Apr.16 to July 3 2008

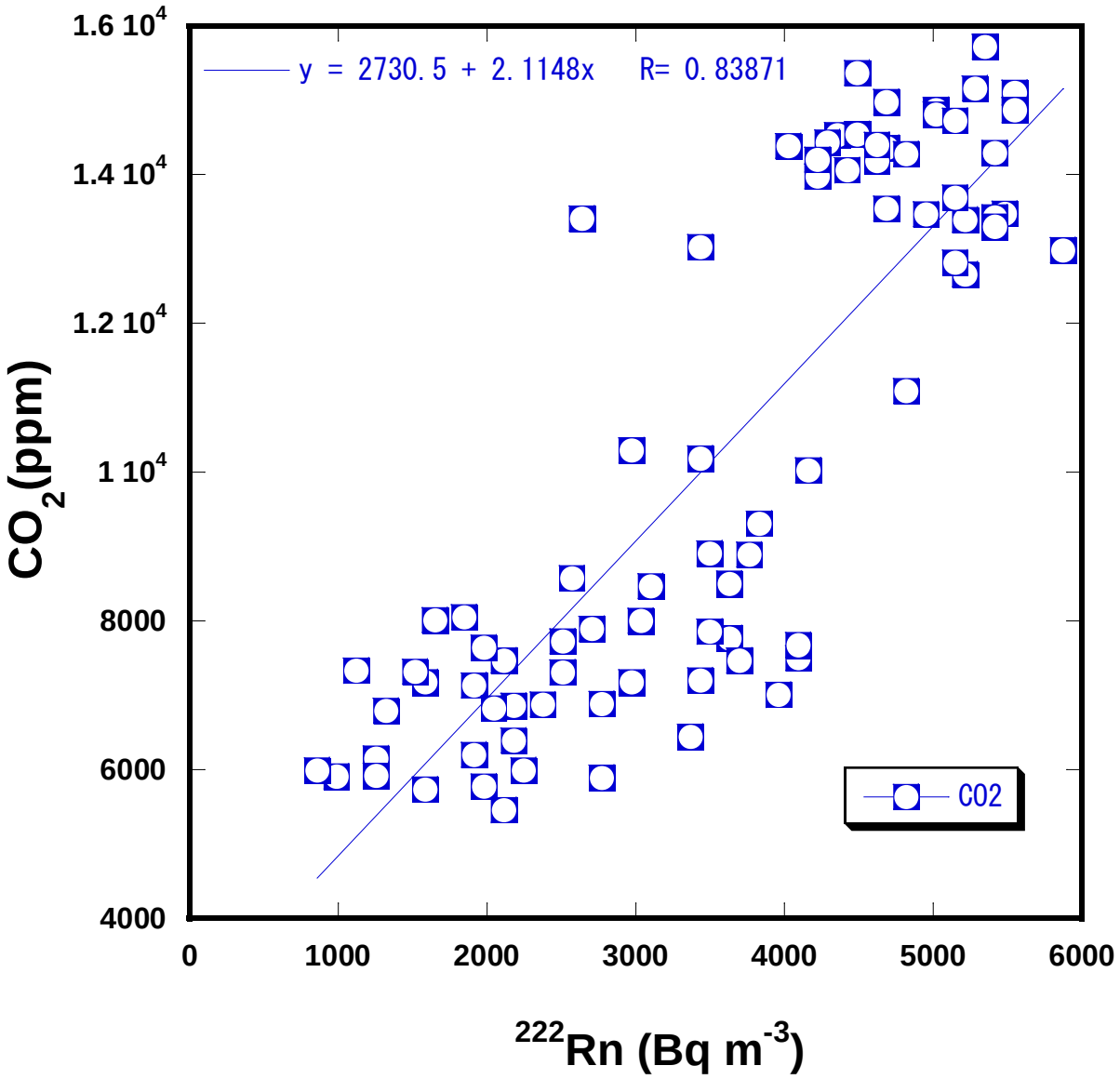


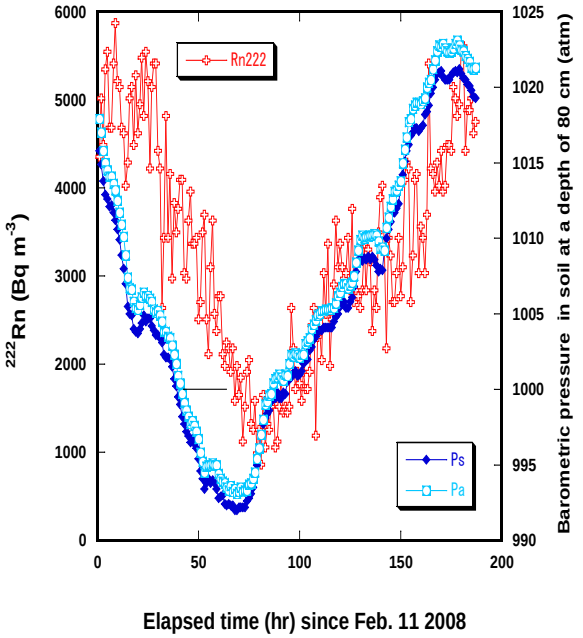
# Behavior of $^{222}Rn$ and $CO_2$ in soil air under thick snow cover (60~100 cm) in winter (1)

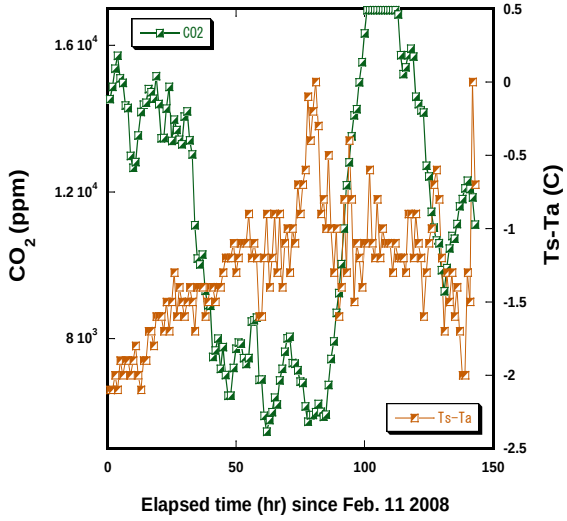


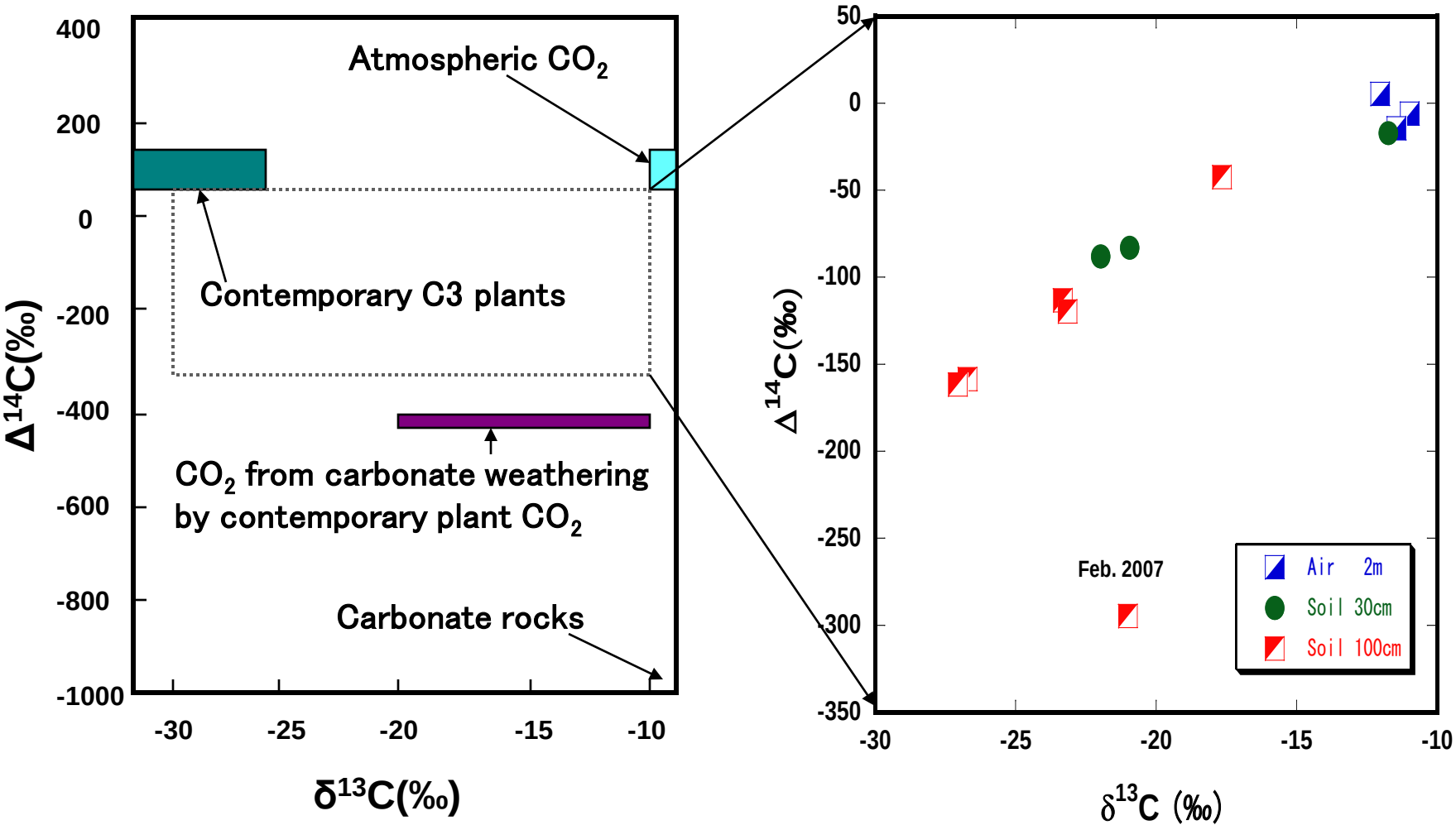
# Behavior of $^{222}Rn$ and $CO_2$ in soil air at a depth of 100 cm under thick snow cover (50~80 cm) in winter (2)



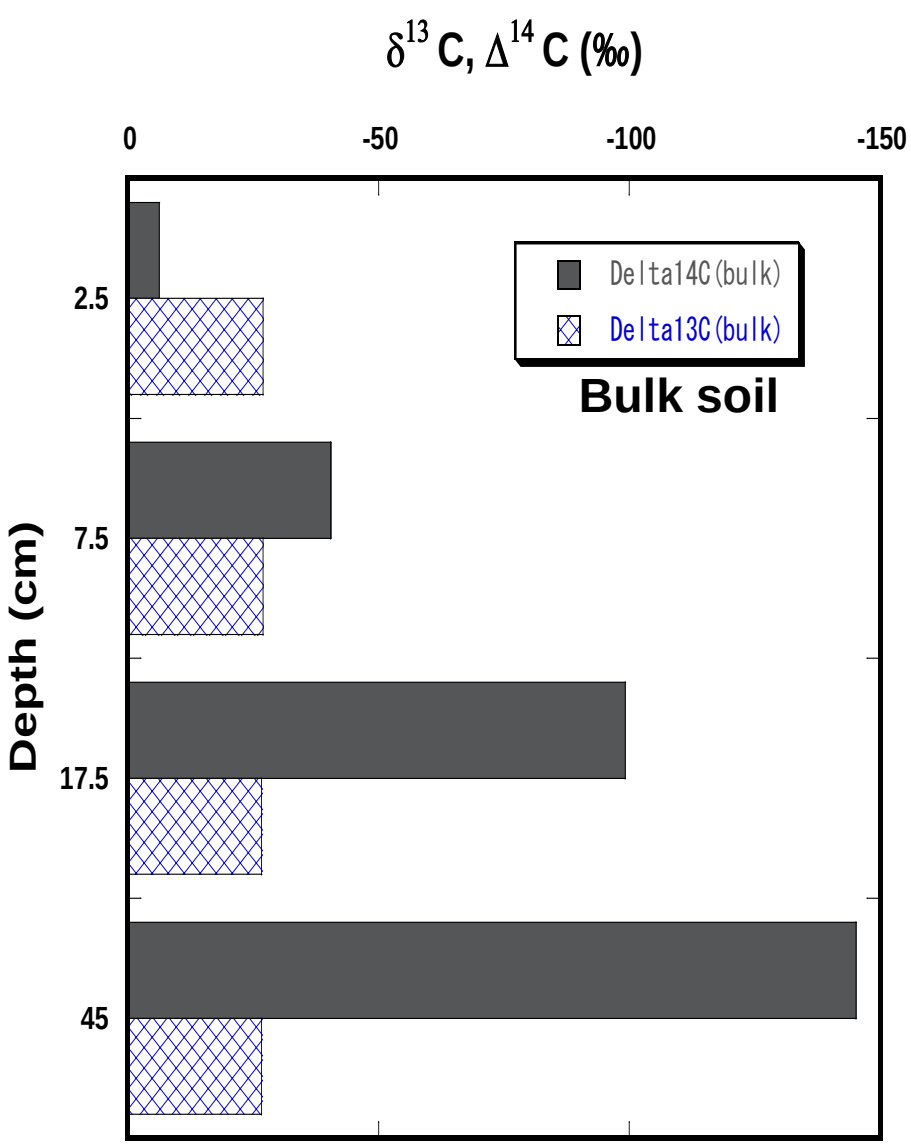



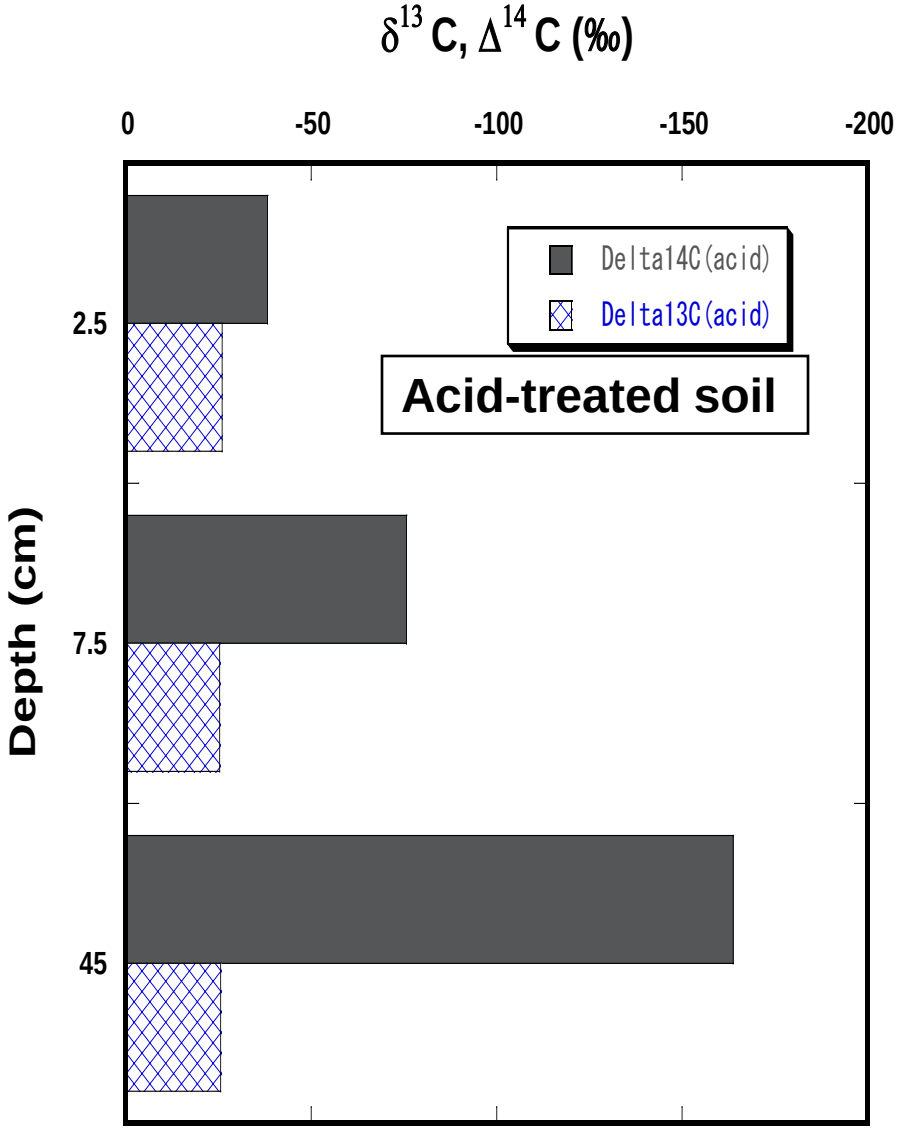



# Distribution of $^{13}C$ ($\delta^{13}C$) and $^{14}C$ ($\Delta^{14}C$) isotopes of atmospheric and soil $CO_2$ in the woods

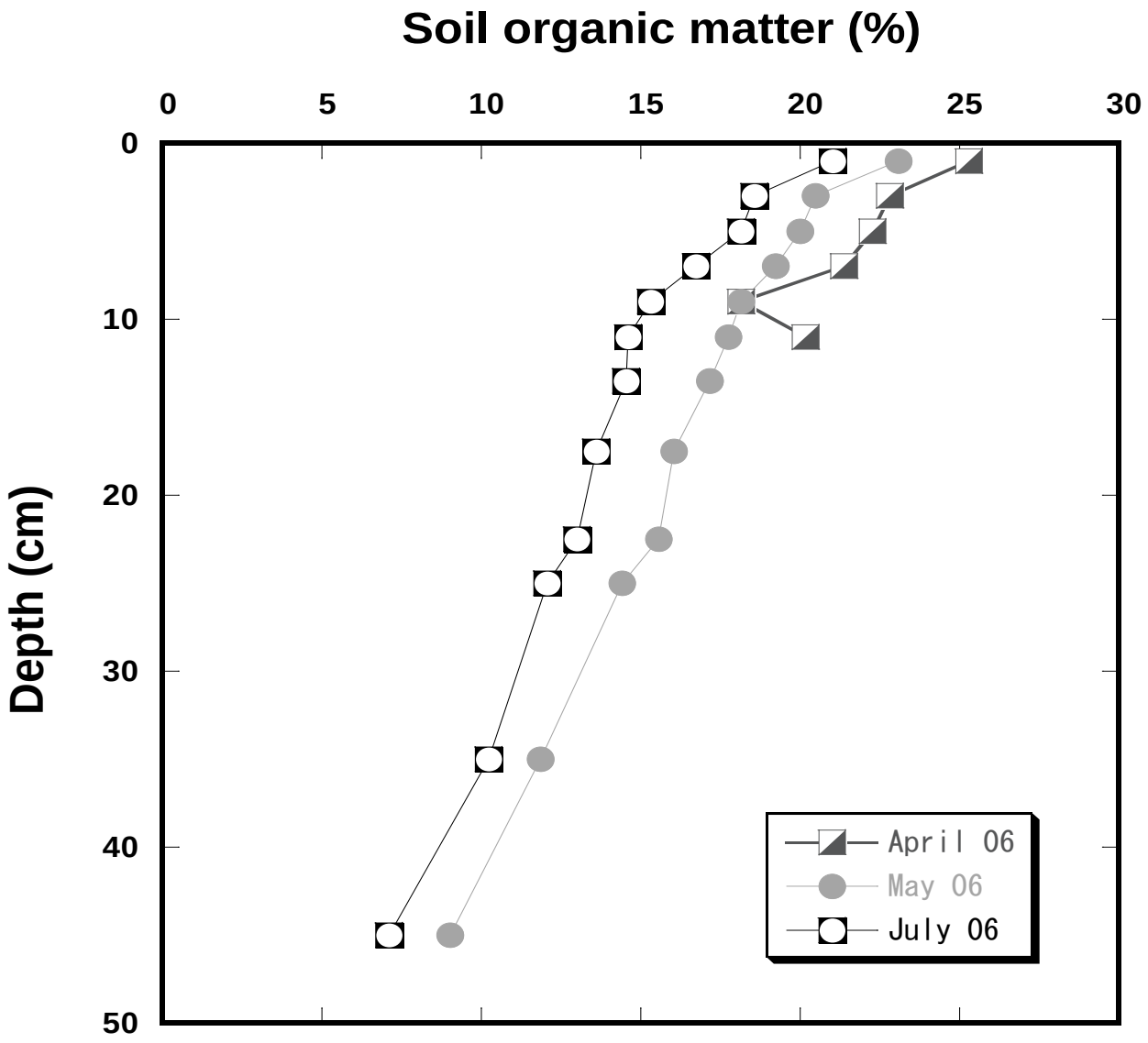


# Change in carbon isotopic ratio ($\delta^{13}C$ and $\Delta^{14}C$) of $CO_2$ in bulk (left) and acid-treated (right) soil with depth



# Seasonal variation of soil organic matter with depth



# Estimating origin of $CO_2$ in soil air

**$CO_2$ in soil air = Atmospheric air + New SOM + Old SOM+ Mineral**

$$\delta^{13}C_{obs} = a\delta^{13}C_{air} + b\delta^{13}C_{newSOM} + c\delta^{13}C_{oldSOM} + d\delta^{13}C_{mineral}$$

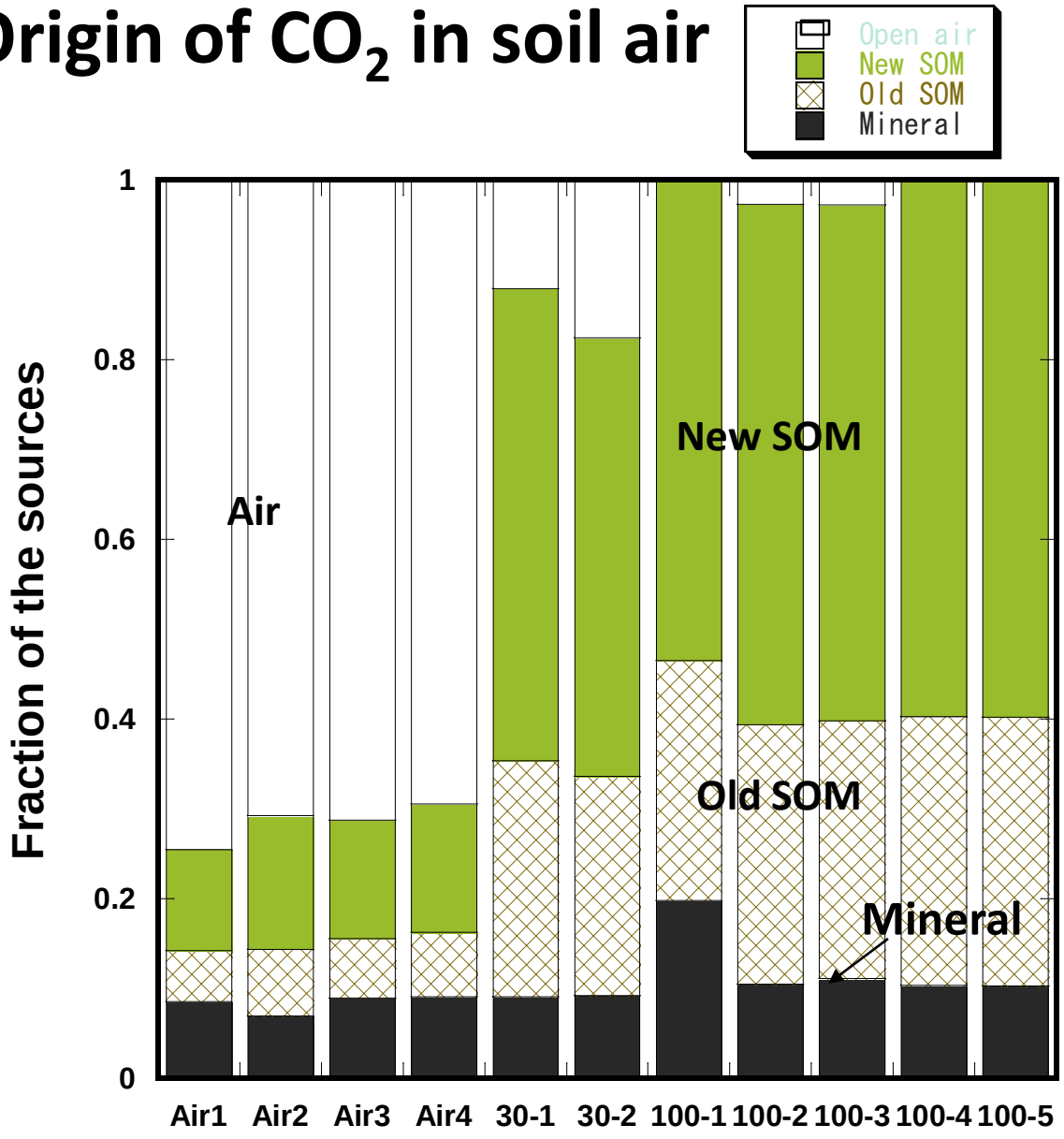
$$\Delta^{14}C_{obs} = a\Delta^{14}C_{air} + b\Delta^{14}C_{newSOM} + c\Delta^{14}C_{oldSOM} + d\Delta^{14}C_{mineral}$$

$$a + b + c + d = 1$$

$$b/c = \alpha$$

| | $\delta^{13}C$ | $\Delta^{14}C$ |
|---|---|---|
| Atmospheric air | -8.5 | 110 |
| Soil organic matter | | |
| young | -27 | 70 |
| old | -25 | -160 |
| Deep source $CO_2$ | -2 | -1000 |

# Origin of $CO_2$ in soil air




Forest air (Air 1, 2, 3 and 4) and soil air collected at 30 cm (30-1and 2) and 100 cm (100-1, 2, 3, 4 and 5) in depth

| Sample | Date |
|---|---|
| **Forest Air** | |
| Air 1 | Feb.22pm 07 |
| Air 2 | Sep. 6am 07 |
| Air 3 | Sep.6pm 07 |
| Air 4 | Mar.7am 08 |
| **Soil air** | |
| 30-1(30 cm) | Feb.22pm 07 |
| 30-2 (30 cm) | Sep.6pm 07 |
| 100-1(100cm) | Feb.20pm 07 |
| 100-2(100cm) | Sep.6am 07 |
| 100-3(100cm) | Sep. 6pm 07 |
| 100-4(100cm) | Mar.7am 08 |
| 100-5(100cm) | Mar.7pm 08 |

## Summary

**Radon ($^{222}Rn$) and $CO_2$ monitoring (and its carbon isotopic analyses) in soil air in the woods on the campus of Hokkaido University**

- **Daily and seasonal changes in $^{222}Rn$ and $CO_2$ concentrations were observed.**

- **Meteorological parameters influence both gaseous components in soil air even under thick snow cover in winter.**

- **Carbon isotopic analyses ($\delta^{13}C$, $\Delta^{14}C$) are applicable for estimating origin of $CO_2$ in soil air, especially in winter.**

最後に。。。

# 環境放射能測定ー将来の展望

- より低レベルの放射能測定法
- 新規核種の検出
- 緊急時における迅速測定系の確立
- グローバルスケールでのデータの互換性
- だれでも放射能測定

謝辞

本研究の一部は、日本原子力研究開発機構施設供用助成により実施されました。

同機構青森研究開発センターむつ事務所職員の皆様に感謝申し上げます。


2009年度バックエンド週末基礎講座
ありがとうございました。
藤吉亮子