OBSERVATION OF ATMOSPHERIC RADON-222 WITH A NEW MEASURING SYSTEM AT THE WMO/GAW MONITORING STATIONS IN JAPAN

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Topics

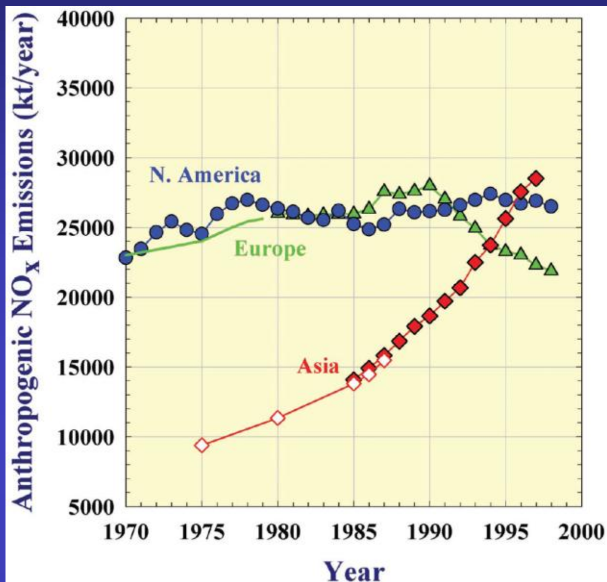
1) Briefly overview for the implications of Rn observation.

2) Briefly introduction of newly developed atmospheric Rn measuring system.

3) Observed results for temporal and spatial variations of Rn over the western North Pacific

Background and Implications

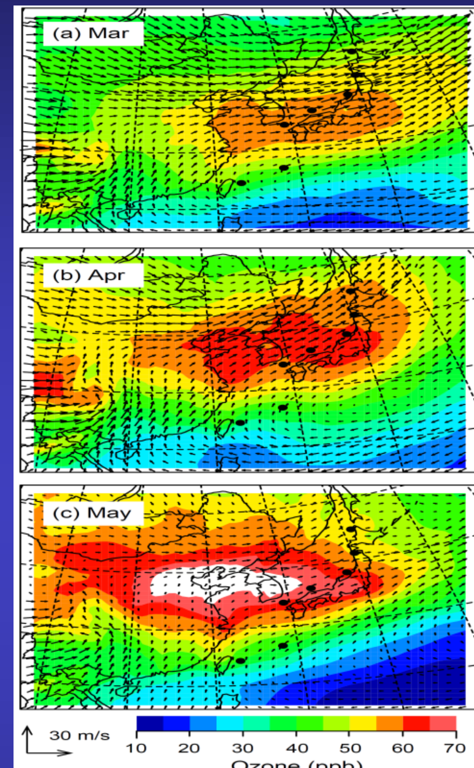
Increase of Anthropogenic Emissions



Akimoto (2003) Science, 302, 1716-1719

In the Asian regions, anthropogenic emissions are rapidly increasing due to the economic growth. The Asia has a large potential for future emissions of greenhouse gases and other pollutions.

Widespread Ozone



Tanimoto (2005) GRL, 32, L21805, Doi:10.10292005GL023514.

As shown in the model, widespread increased ozone with Asian outflow is produced by the photochemistry.

Various Impacts

1 ） Human Health

2 ） Agriculture

3 ） Regional Climate

4 ） Biological Diversit

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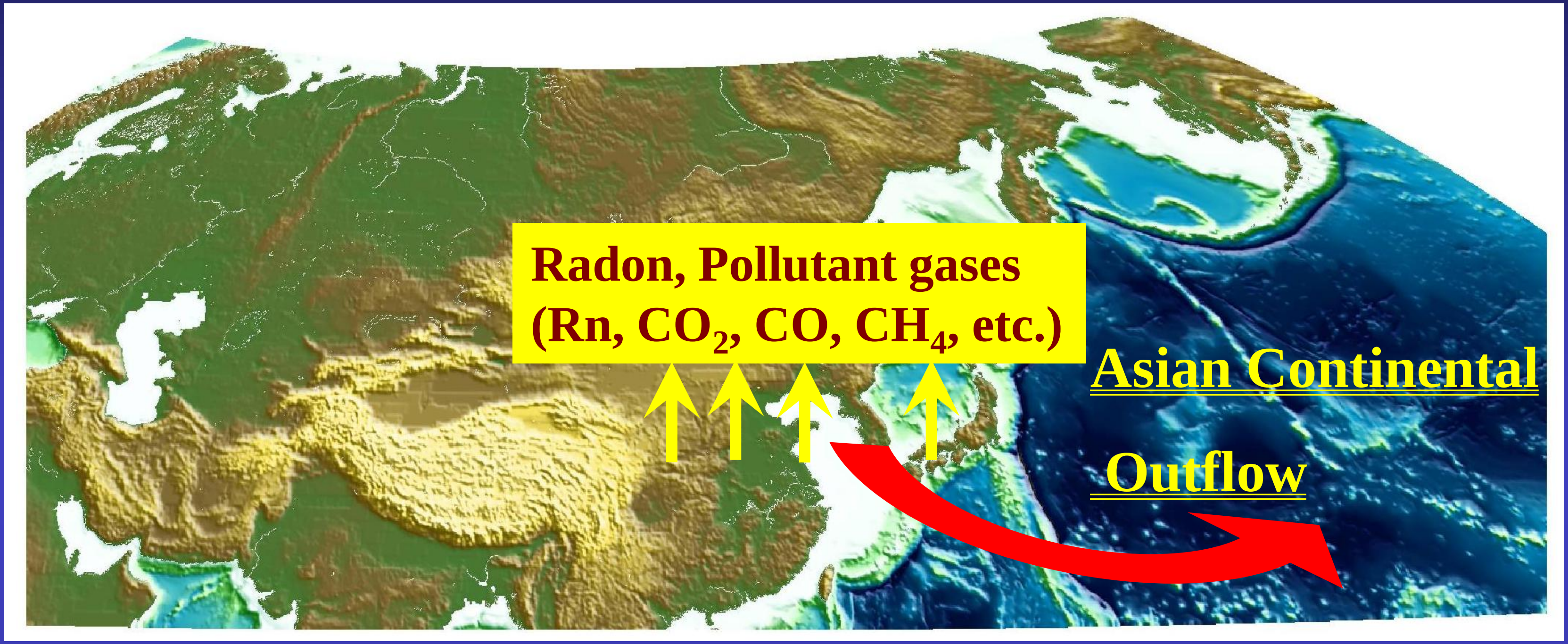
The air quality reduction causes the various impacts such as human health, agriculture, and regional climate.

For chemical weather forecasting, more understandings of transport mechanism is required.

Motivation of Asian outflow study

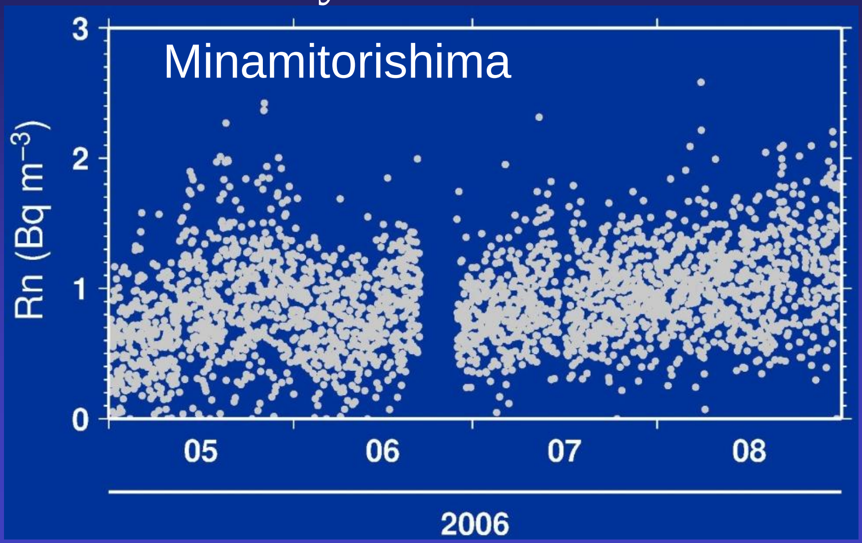


Radon : Useful chemical tracer for Asian continental outflow

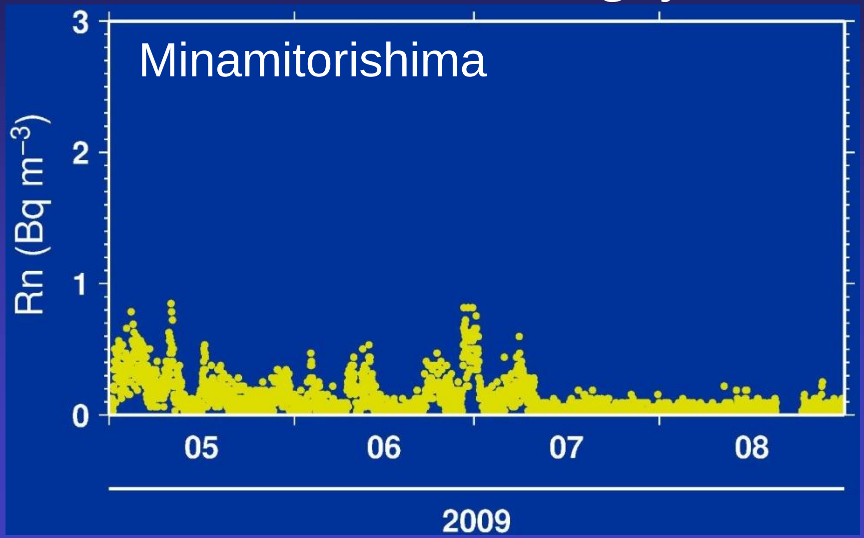
- Radon-222 (Rn-222) is a useful chemical tracer for identifying continental air masses.
- Because it releases from the soil to the atmosphere, and its half life time of 3.8 days for only radioactive decay is suitable for tracing the travel of the Asian outflow over the western North Pacific.

Development for New Radon Measuring System

Commercially available radon monitor



New radon measuring system

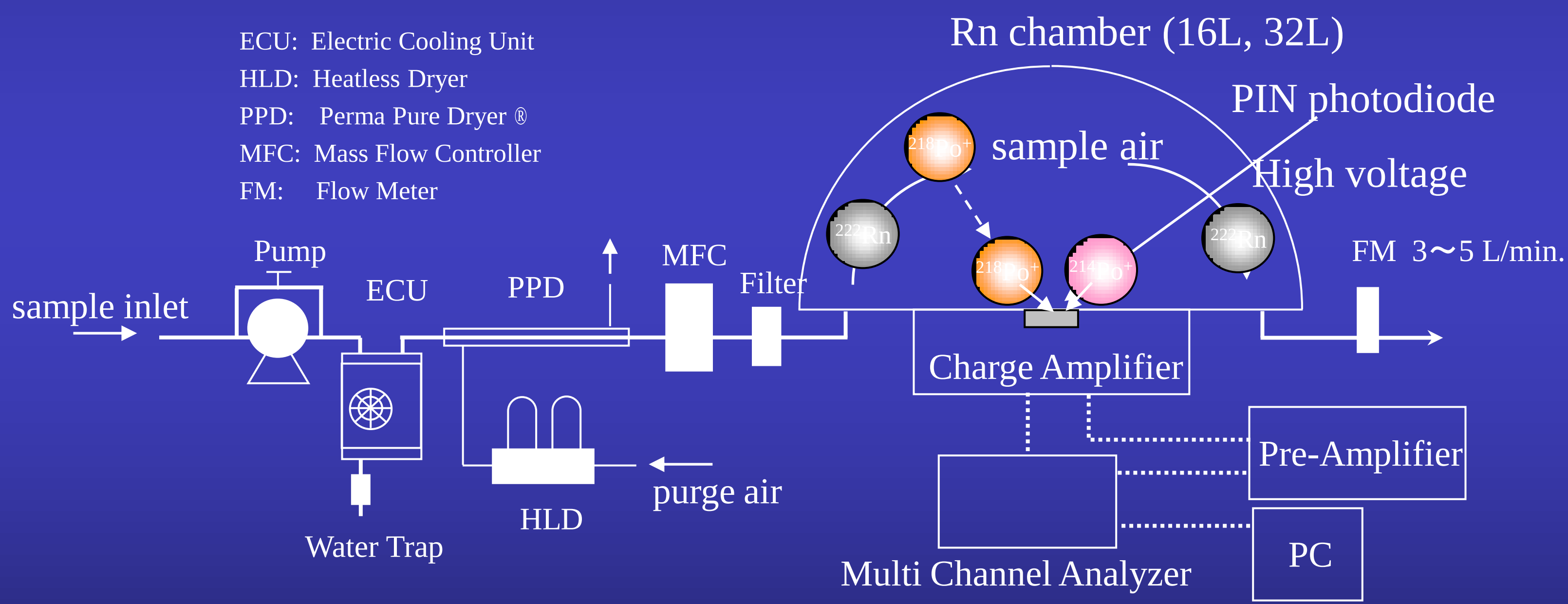


- **A commercially available radon monitor in Japan was insufficient for extremely low level at Minamitorishima station, where Rn background level is less than 0.5 Bq/m^3.**
- **We had developed a new radon measuring system using a high sensitive PIN photodiode by a co-operation research program with AIST and our Institute.**
- **As shown in this picture, our Rn system is relatively compact for installing in the limited space of the observatory, and routinely operated by automated observations.**

Electrostatic method with PIN photodiode

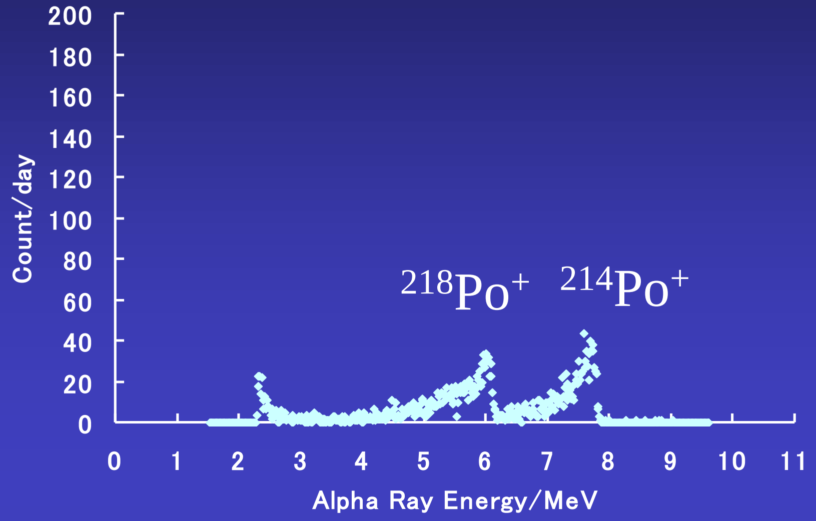
$$^{226}\mathrm{Ra} \xrightarrow[\alpha]{1.600\times 10^{3}\mathrm{y}} {}^{222}\mathbf{Rn} \xrightarrow[\alpha]{3.824\mathrm{d}} {}^{218}\mathbf{Po} \xrightarrow[\alpha]{3.10\mathrm{m}} {}^{214}\mathrm{Pb} \xrightarrow[\beta]{26.8\mathrm{m}} {}^{214}\mathrm{Bi} \xrightarrow[\beta]{19.9\mathrm{m}} {}^{214}\mathbf{Po}$$

$$\xrightarrow[\alpha]{164.3\mu\mathrm{s}} {}^{210}\mathrm{Pb}\,(\mathrm{Ra\text{-}D}) \xrightarrow[\beta]{22.3\mathrm{y}} {}^{210}\mathrm{Bi}\,(\mathrm{Ra\text{-}E}) \xrightarrow[\beta]{5.013\mathrm{d}} {}^{210}\mathbf{Po}\,(\mathrm{Ra\text{-}F}) \xrightarrow[\alpha]{138.4\mathrm{d}} {}^{206}\mathrm{Pb}\,(\mathrm{stable})$$

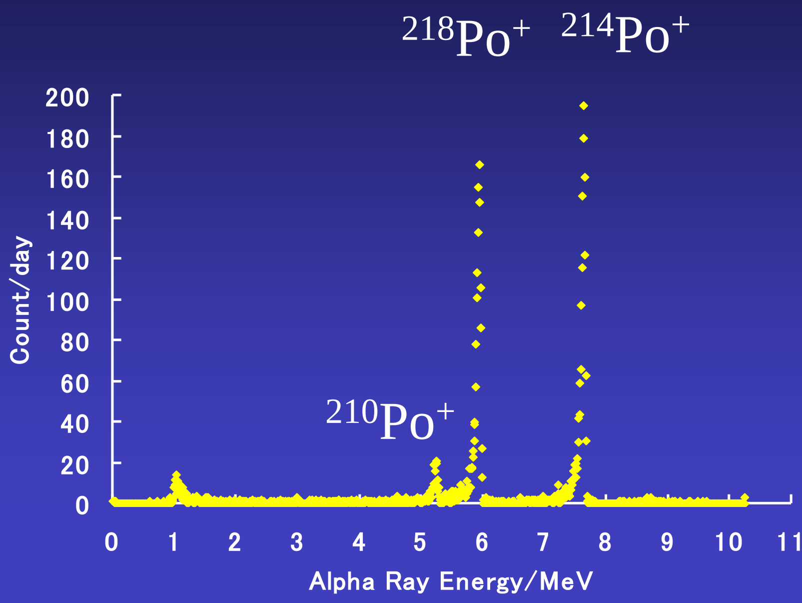


- In our electrostatic method, Po positive ions are collected on PIN photodiode by high negative voltage, where a-rays from respective radon progenies are separately counted depending on their energy.
- Counting rates of a-rays from radon progenies are used to calculate the Rn activity.
- Using this system, continuous and automated Rn measurement can be made.

Comparison of alpha spectrum



Commercially Available Radon Monitor



New Radon Monitor

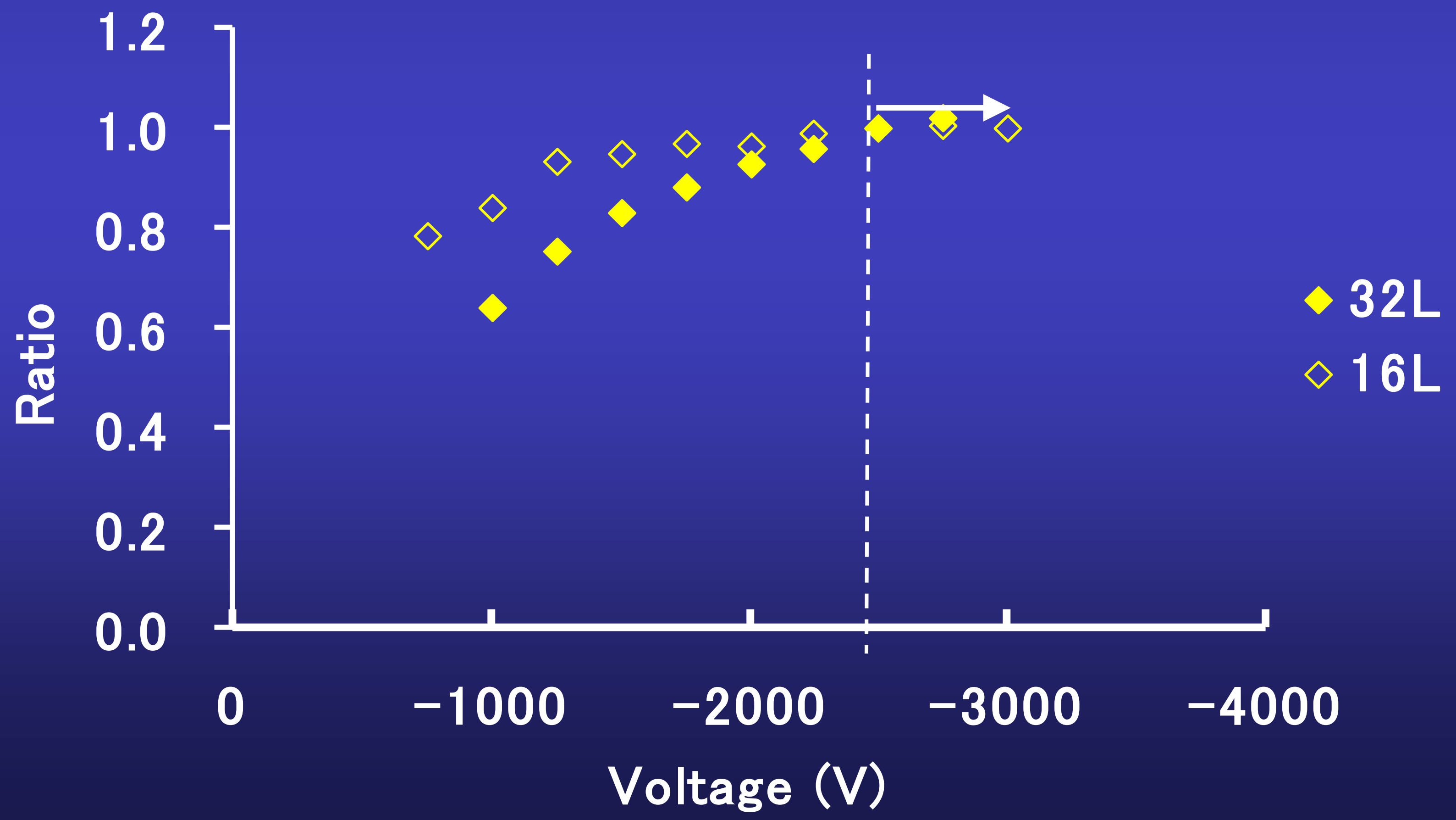
- Our Rn monitor can sharp-separately detect the peak of ^{218}Po with net short half life of 3.1 min after ^{222}Rn decay, which is useful for high time-resolution measurement.
- Our Rn monitor can distinguish ^{218}Po from ^{210}Po, with long total half life (~22 yr) accumulated on the detector.
- Such good peak separation reduces effects of ^{210}Po on the ^{218}Po counting rate, and prevents our Rn monitor from overestimating Rn activity even after long time use.
- We decided to calculate Rn concentration only from ^{218}Po counting rate in this study.

Dependence of calibration factor (count hr^{-1}) /(Bq m^{-3}) on high voltage

To collect Po positive ions, PIN photodiode is charged by high negative voltage.

Calibration factor is found to be almost constant at <-2500 V for both chambers.

Thus, we set the voltage of -2500 V for the both 32L and 16L chambers.



Results of Calibration

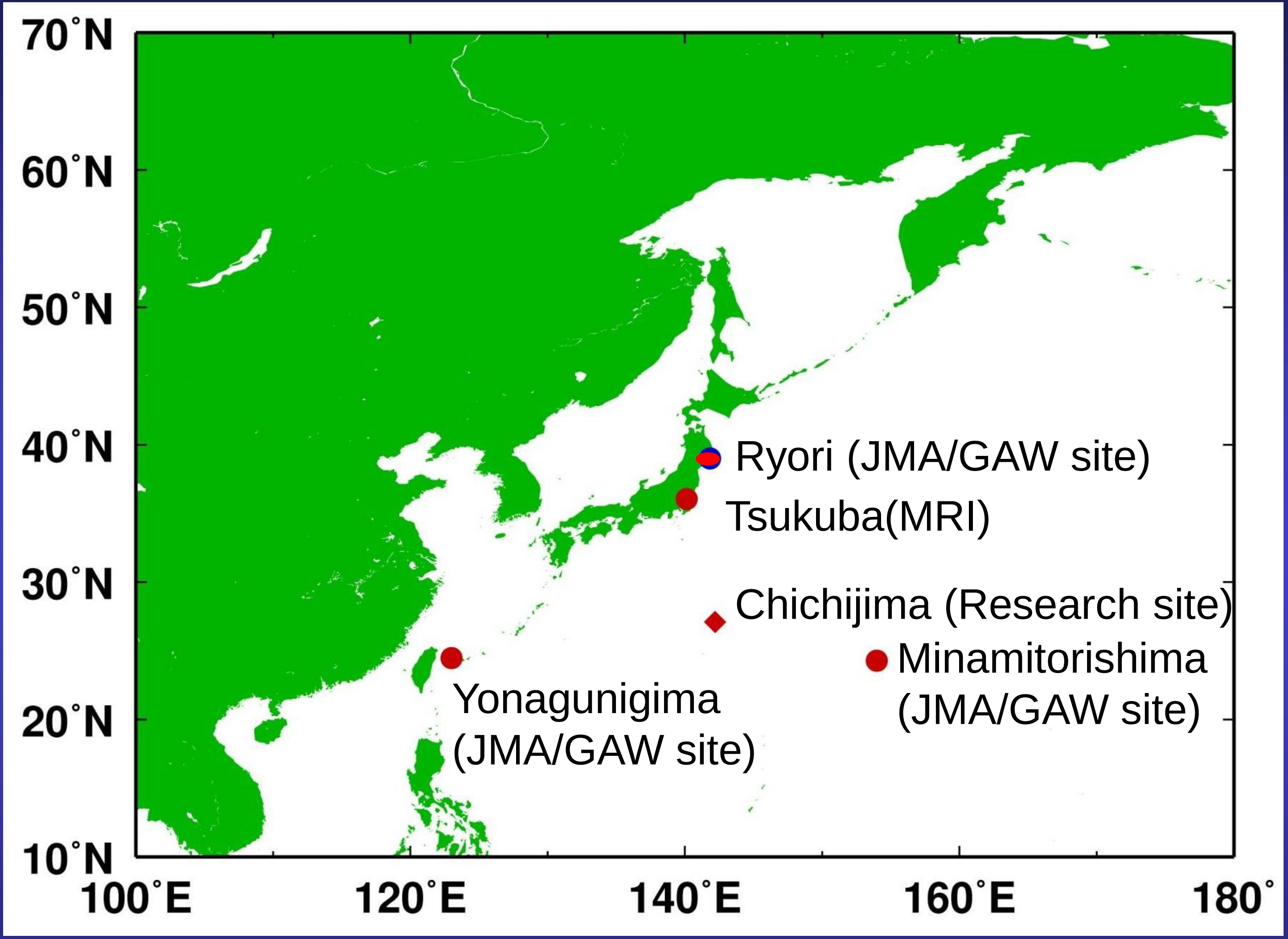
	32L	16L
Calibration Factor/counts $h^{-1}(Bq\ m^{-3})^{-1}$	31.8	19.6
Counting Efficiency	0.27	0.32
Limit of detection (LD) /Bqm^{-3} for 1h	0.16	0.20

This table shows the results of the calibration at Nagoya University in Japan.

It was found that Calibration factor is better for 32 L than for 16 L , reflecting its volume dependence.

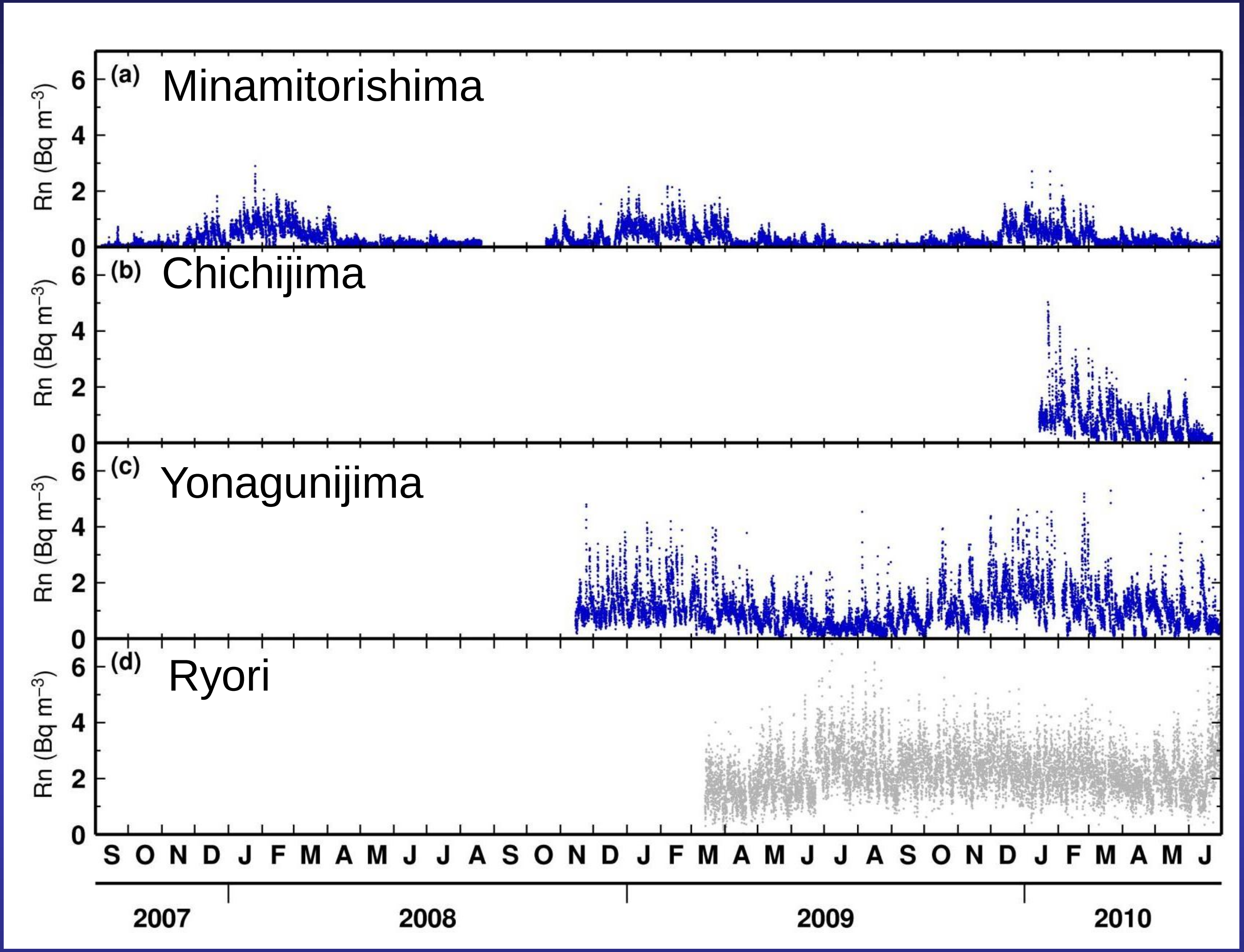
Our compact Rn 222 measuring system has the potential to be widely used for high time-resolution measurements of low-level Rn222.

Rn observation network



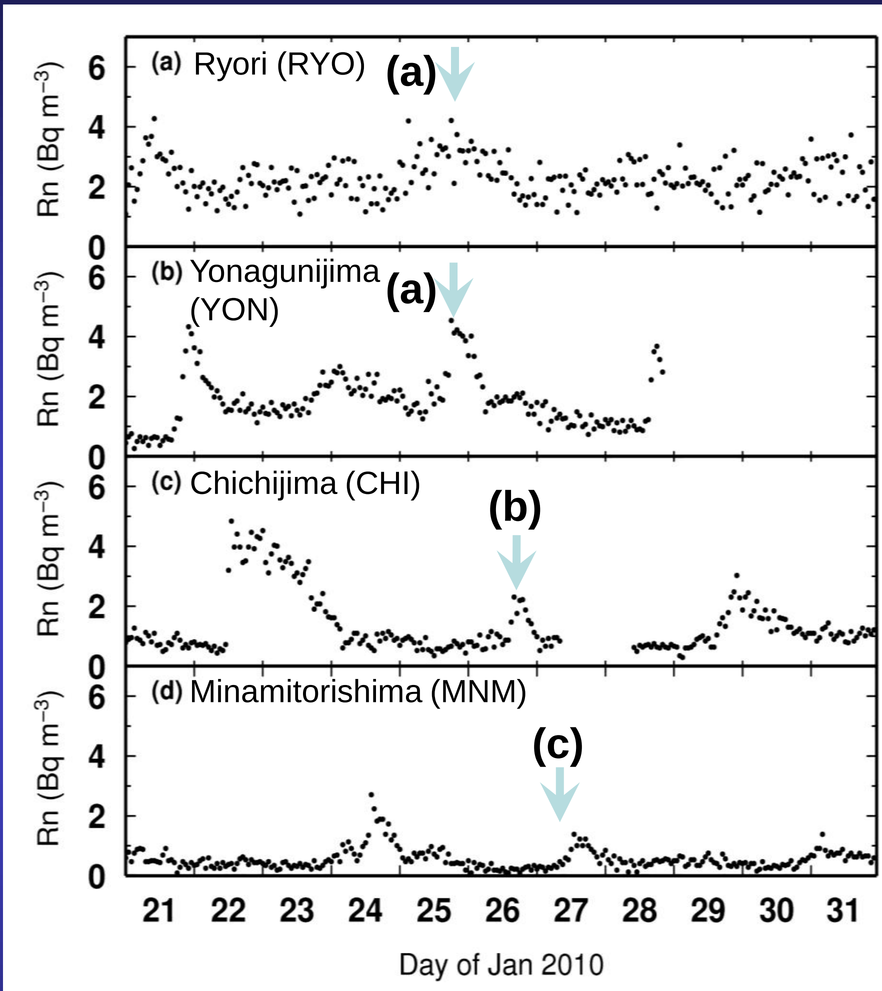
- In our Rn observation network, three Japanese GAW stations of Ryori, Yonagunijima and Minamitorishima are used, because various greenhouse gases are operationally monitored by JMA.
- Chichijima is a research site, where Rn measurements are sometimes made during the campaign observation.
- This network is suitable for the study of Asian continental outflow, because the distance from the continent is different each other.

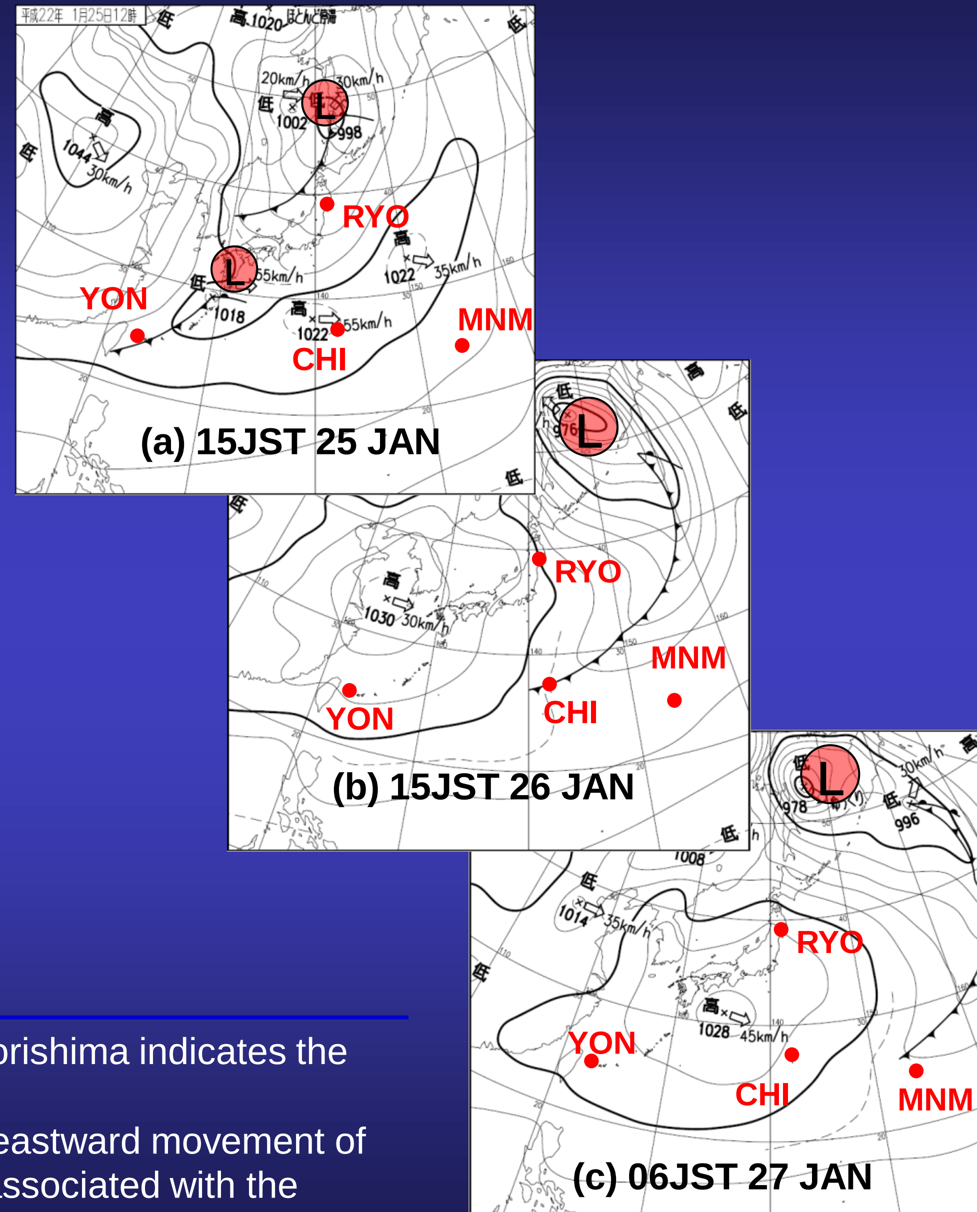
Rn variations observed at 4 stations



- The Rn observation started at Minamitorishima since 2007.
- After that, Rn measurements at other stations of Chichijima, Yonagunijima and Ryori are collected.
- For all of the stations, not only a distinct seasonal variation but also numerous Rn enhanced peaks are well captured.
- These Rn enhancement events indicate the signals of the Asian continental outlow.

Rn Plume associated with a moving cold front





- The time series of Rn peaks from Ryori to Minamitorishima indicates the Rn plume is moving toward the east.
- It is clearly revealed in the weather charts that the eastward movement of the Rn plume is driven by the cold front system associated with the development of the moving cyclone.
- We found that this frontal transport is responsible for most of the Rn events.

Validation of model by the observed Rn

3D Chemical Transport Model

NIRE-CTM-96 (S. Taguchi, 1996)

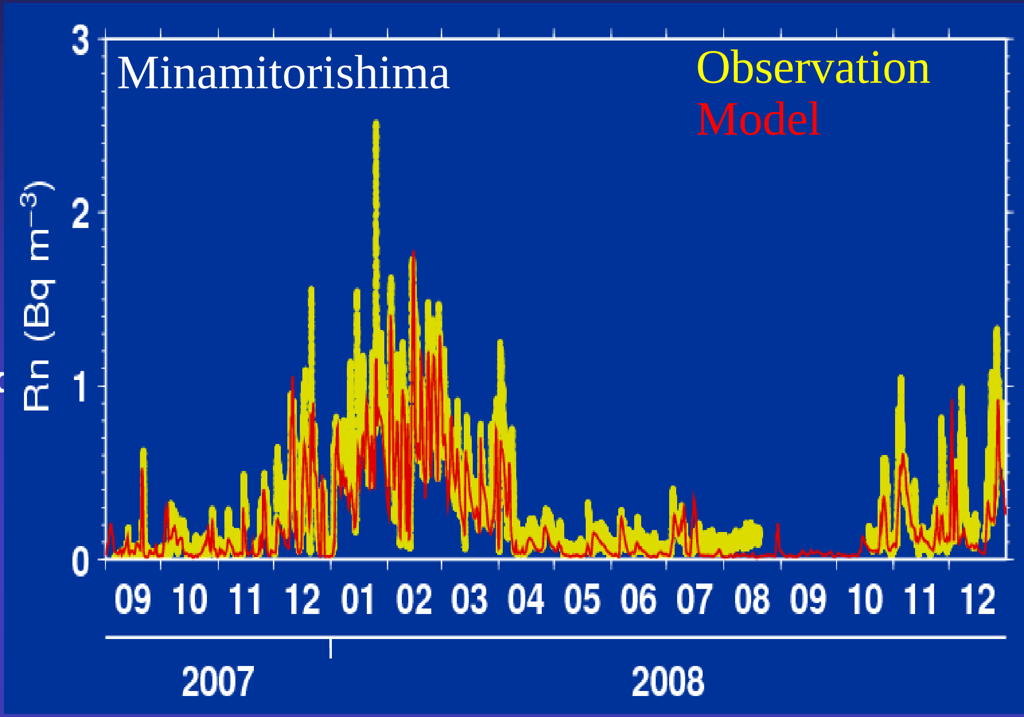
Driven by NCEP/NCAR reanalysis Data

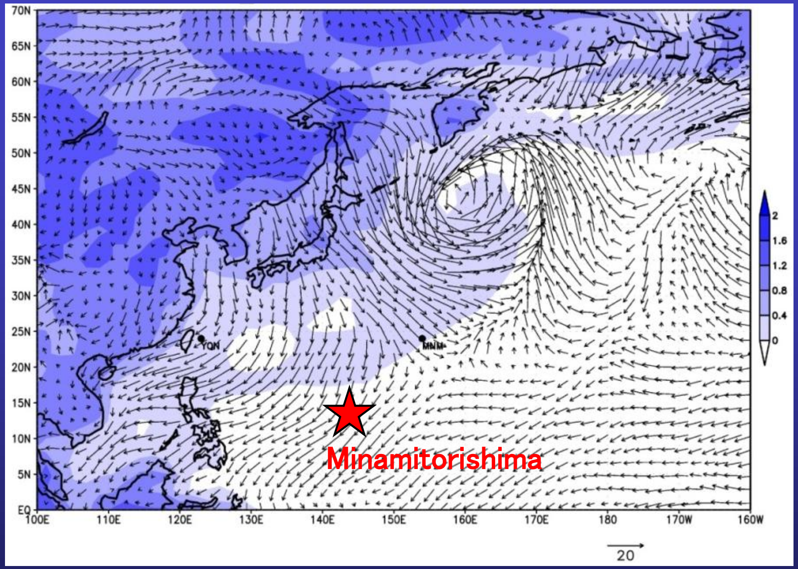
Radon Emission (D. Jacob et al, 1997)

1.0 atom cm^{-2} s^{-1} (60°S - 60°N)

0.5 atom cm^{-2} s^{-1} (60°N - 70°N, without Greenland

0 atom cm^{-2} s^{-1} (Ocean)

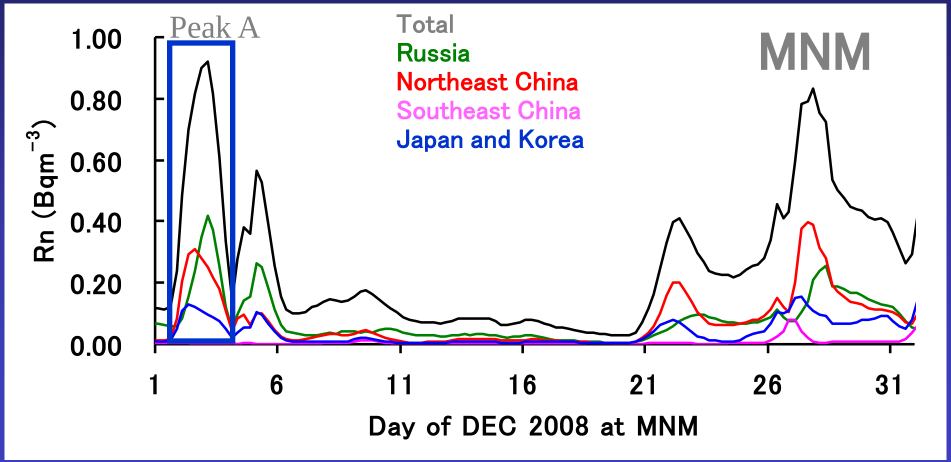


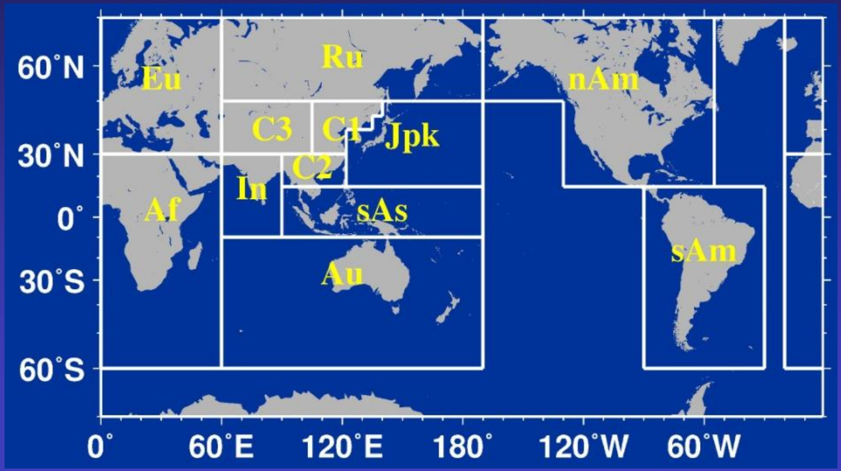


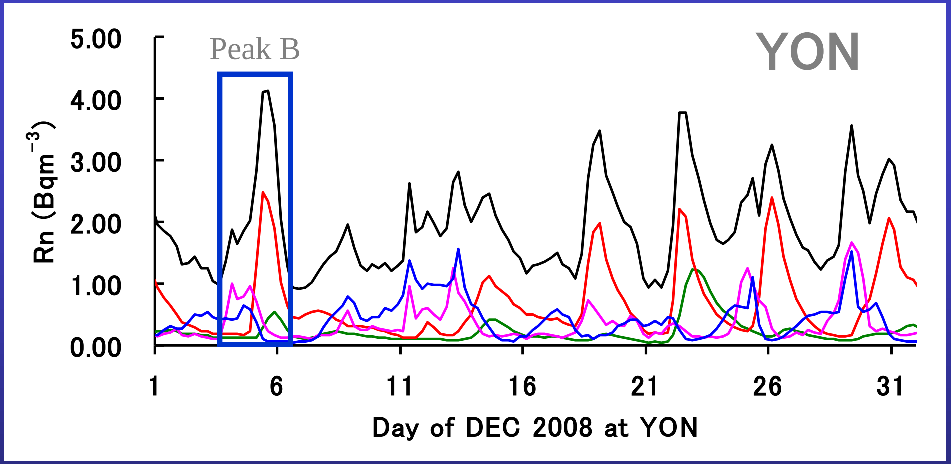
Surface spatial distribution of Rn by CTM on 25 JAN 2008 12UTC

- **To confirm the Rn increased events, CTM simulation was made to reproduce the observed Rn at Minamitorishima station, as shown in the upper panel.**
- **The left panel clearly shows that enhanced Rn is widespread along with the cold front over the western North Pacific.**
- **These results indicates that Rn measurements are useful for validating the transport processes of the 3-D model.**

Tagged tracer experiment by 3-D model







Eu: Europe Jpk: Japan and Korea
Af: Africa In: India
Ru: Russia sAs: South Asia
C1: Northeast China Au: Australia
C2: Southeast China nAm: North America
C3: West China sAm: South America

The tagged tracer experiment by using the model is also available for quantitatively estimating the source regions.

Contribution (%)

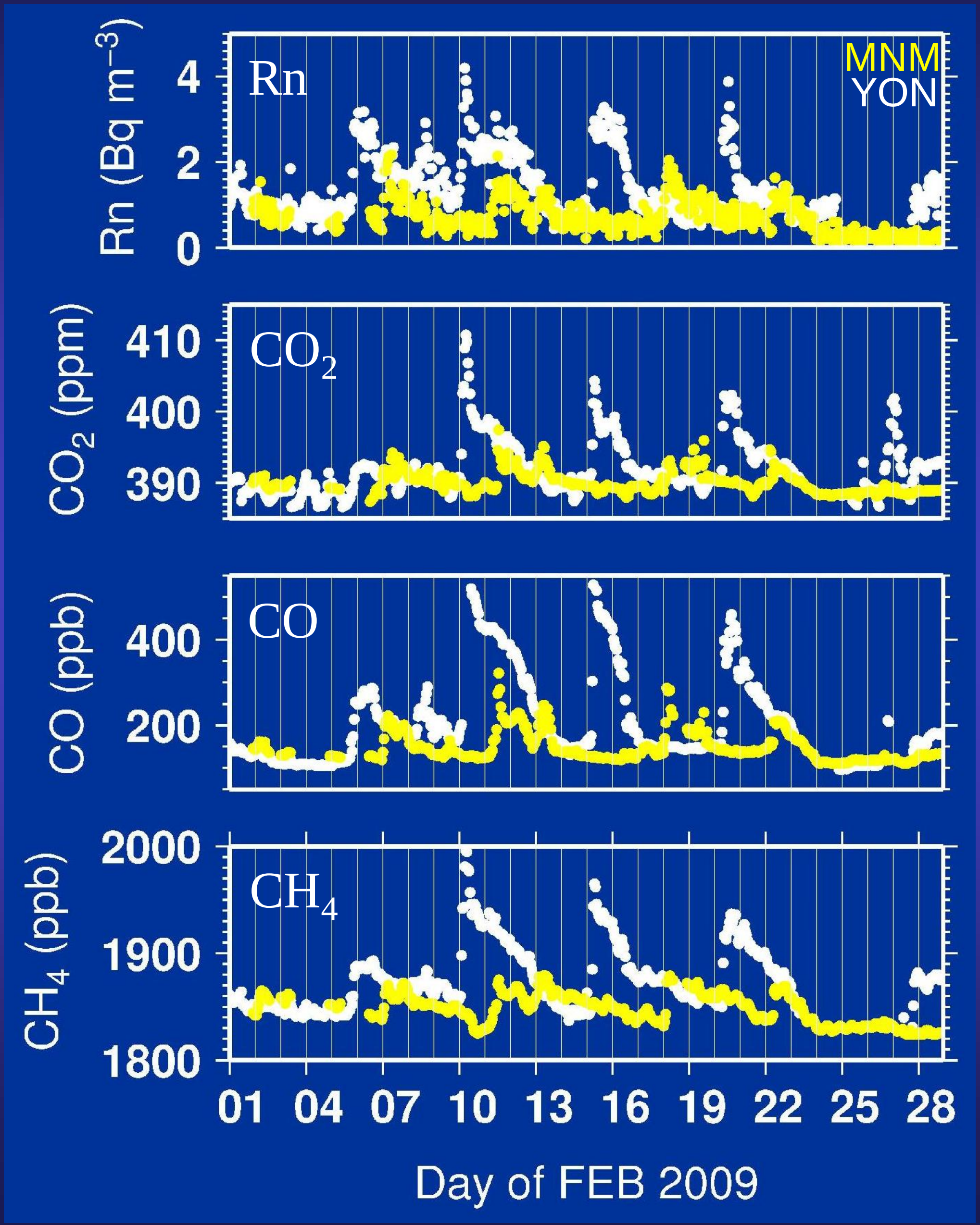
	Europe	Russia	NE China	SE China	W China	Japan & Korea
peak A	9.0	35.5	32.6	0.3	6.4	14.3
Peak B	4.0	10.2	40.0	21.2	12.2	11.8

Main source regions

MNM: Russia and NE China

YON: NE China and SE China

Comparison with Other Trace Gases



- High resolution measurements of Rn well captured a synoptic-scale increase due to the intrusion of continental air masses.
- It is also of interest that all of the Rn peaks are well coincided with the increased trace gases of CO2, CO and CH4.
- These tight relations demonstrate the transport of continental air masses passing over the anthropogenic-emissions regions over the continent.
- Thus, the enhancement ratios could provide information of the air quality over the continent.
- Also, more observations could allow us to clarify the long-term change in the Asian continental emissions

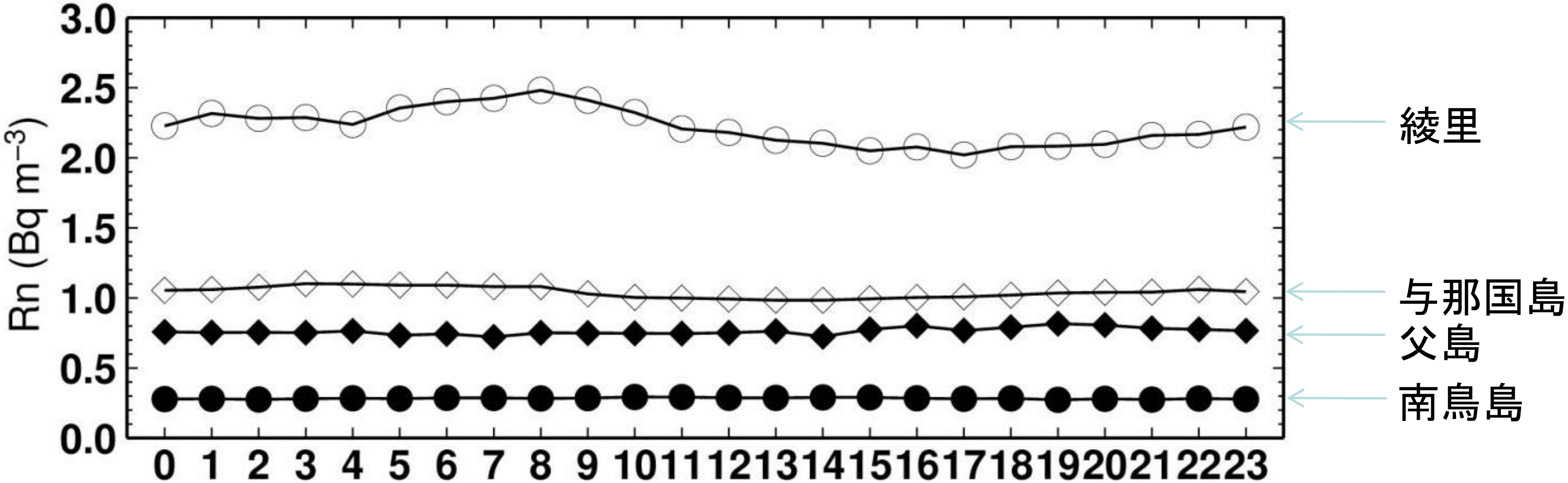
Conclusions

- High sensitive radon measuring system has been developed. This system detected successfully seasonal as well as synoptic-scale variations over the western North Pacific.
- Synoptic-scale variation of radon was brought by a passage of cold front associated with a moving cyclone.
- For the 3-D model simulation, widespread Rn from the Asian continental outflow is well illustrated.
- Tight relations between Rn and other trace gases could be used for flux estimations on the Asian continent source region.

【Reference】

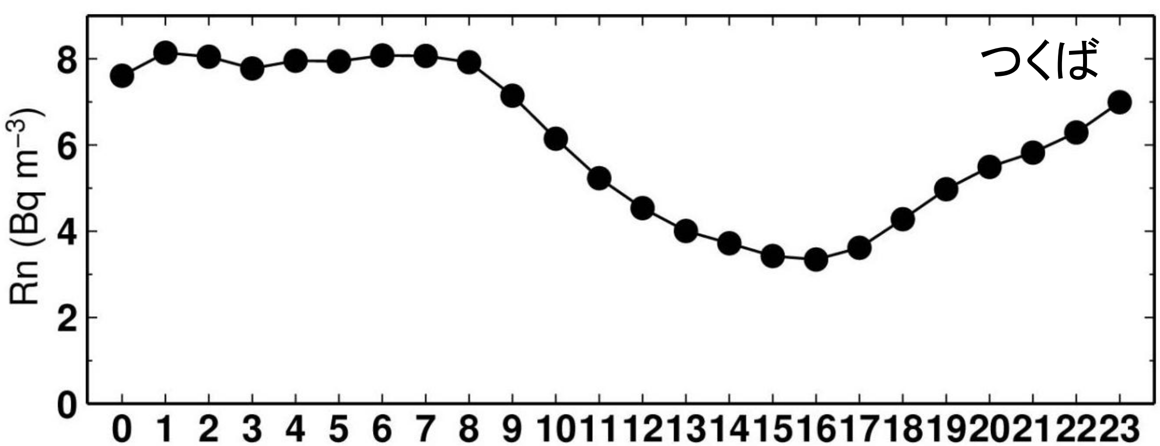
Akira WADA, Shohei MURAYAMA, Hiroaki KONDO, Hidekazu MATSUEDA, Yousuke SAWA and Kazuhiro TSUBOI: “Development of a Compact and Sensitive Electrostatic Radon-222 Measuring System for Use in Atmospheric Observation”. JMSJ, Vol. 88, 123-134. (2010) .

日変動



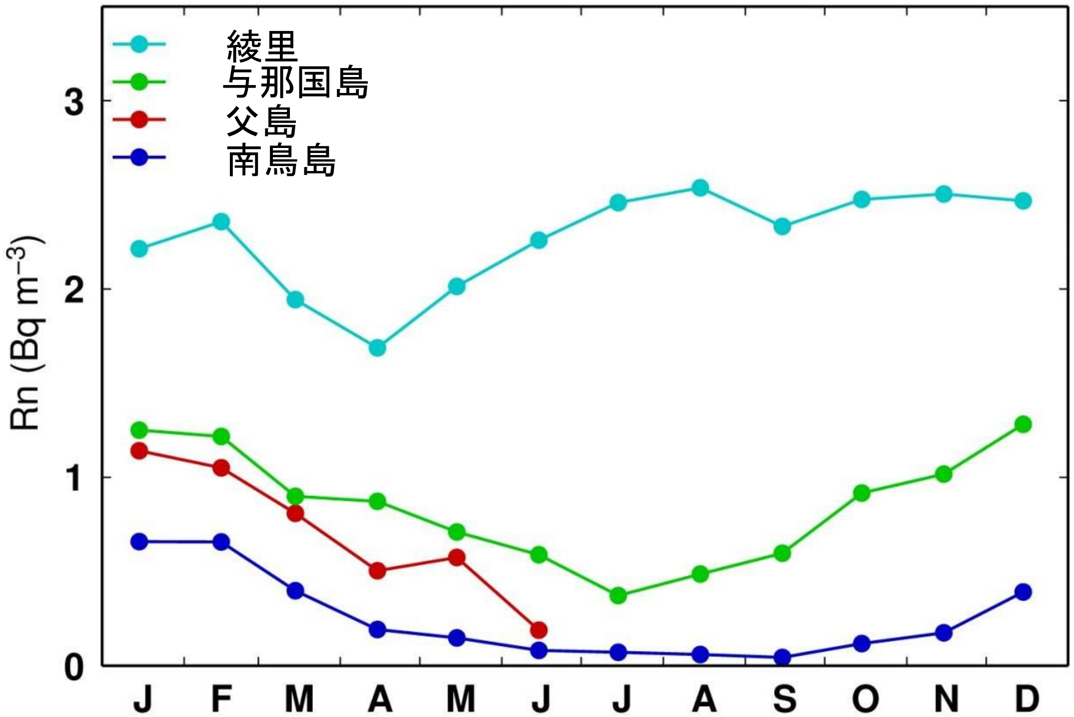
大気中ラドン濃度の平均日変動

つくばで観測される明瞭な日変動は、南鳥島、父島および与那国島ではみられない
綾里ではわずかに日変動がみとめられた

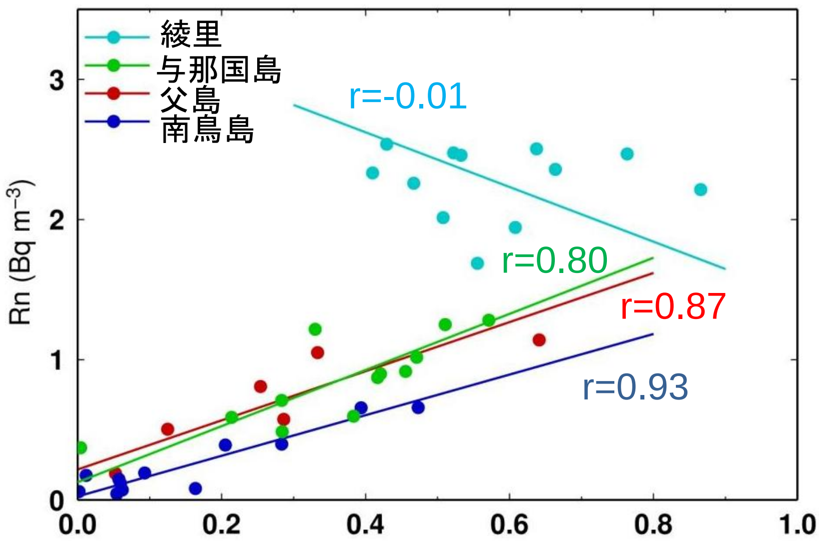


2010年1月につくばで観測された大気中ラドン濃度の平均日変動

季節変動



大気中ラドン濃度の月平均値



月平均ラドン濃度と北西風の割合との相関

南鳥島、父島、与那国島のラドンの季節変動は北西風の割合と高い相関がある。一方綾里では風向との相関はなかった。

Comparison with other Rn monitors

Institute	This Work	ANSTO** (Cape Grim)	Nagoya Univ.*
Method	Electrostatic	Two filter	Electrostatic
Detector	PIN photodiode	ZnS(Ag)	ZnS(Ag)
Counting efficiency	0.32 & 0.27	0.33	0.3
Separate detection of ^{218}Po	Yes	No	No
Chamber/L	16 & 32	9000	16.8
Flow rate/$Lmin^{-1}$	3～5	200	1
LD***/Bqm^{-3}	0.20 & 0.16	0.003**	0.35
Time resolution/h	0.5～1	>1.5	1

* T. Iida et al. 1996, Environ. Int.

** S. Whittlestone and W. Zahorowski, 1998, JGR.

*** LD represents limit of detection defined by Currie (1968).

- Counting efficiency: almost similar. LD: worse than ANSTO, better than Nagoya.
- Time resolution: better than other monitors.
- Compact system: small chamber, low flow rate with a small pump.
- Separately detect ^{218}Po: high time resolution measurement & long time use.