

Concentration variability of atmospheric radon and gaseous pollutants at background area of Korea between 2017 and 2018

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Abstract: The concentrations of radon in the atmosphere were measured at the Gosan site of Jeju Island during 2017-2018, in order to investigate the time-series variation characteristics and the dependency of airflow transport pathways. The mean ²²²Rn concentration was 2,480 mBq m⁻³, and its monthly concentration in November was 3,262 mBq m⁻³, more than twice as that in July (1,459 mBq m⁻³). The diurnal radon concentrations increased throughout the nighttime to the maximum (2,862 mBq m⁻³) at around 7 a.m., then gradually decreased throughout the daytime by the minimum (1,997 mBq m⁻³) at around 3 p.m. The seasonal and monthly variations of CO, NO₂, O₃ showed a roughly similar pattern to that of radon for the same period, as high in winter and low in summer. The cluster back trajectory analysis described that about 60 % of overall airflow pathways was influenced by the airflow from China. The concentrations of radon and gaseous pollutants were relatively high as the airflow was influenced by China continent, but comparatively much lower as influenced by the northern Pacific Ocean.

Key words: atmospheric radon, gaseous pollutant, Gosan site, cluster back trajectory, airflow pathways

1. Introduction

Radon (²²²Rn) is a major natural radioactive element and distributed widely in nature such as in rocks, soil, and underground water, unlike other artificial radioactive elements.^{1,2} Also the inert gas is chemically inactive, and has a characteristics to disperse extensively into the atmosphere by convection.³ ²²²Rn is generated during the decay chain process of ²³⁸U, directly by alpha decay of ²²⁶Ra, and become to be stable into

²⁰⁶Pb through each 4 times α - and β -decays with 3.82 day half-life. Its major isotopes are ²²⁰Rn (thoron) and ²¹⁹Rn (actinon) that are generated from the decay processes of ²³²Th and ²³⁵U, respectively. However the half-lives of those isotopes are so short, respectively 55 seconds and 4 seconds, so the atmospheric radon exists mostly as ²²²Rn and sinks from the atmosphere through the radioactive reaction.⁴

²¹⁴Po and ²¹⁸Po nucleic progenies occurred from the radon decay process remain in the lung for a long

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period and may induce the radioactivity to cause lung cancers.¹ ^{222}Rn progenies can also lead to tissue damages and lung cancer by generating the radioactivity, because they are easily adsorbed on fine particles and deposited in the lung by inhalation.⁵ The International Agency for Research on Cancer (IARC) of the World Health Organization (WHO) has classified the radon as a first class carcinogen identified by its human oncogenicity.⁶ Moreover, the Environment Protection Agency (EPA) has specified the radon as the second high risk substance, followed by the cigarette smoking, causing the lung cancer, and the National Academy of Sciences (NAS) has reported 15,000~25,000 deaths are caused by the lung cancer stemmed from radon yearly.^{7,8}

Korean peninsula is situated in downwind direction of the prevailing westerlies at the northeastern Asia continent region, so is affected significantly by the atmospheric pollutants moved from China. Especially, the atmosphere in Jeju island is influenced as much as about 75 % yearly by the airflow from the Asia continent.⁴ Therefore, it would be very effective to apply the radon as a useful tracer for understanding the long-range transport pathways of the atmospheric pollutants. The radon monitoring at the Gosan site of Jeju Island can be practically useful for the application of radon as a transport tracer of pollutants.^{9,10}

The Australian Nuclear Science and Technology Organisation (ANSTO) has established the radon monitoring networks at major 30 stations around the world with the basis of World Meteorological Organization/Global Atmospheric Watch (WMO/GAW),^{9,11} and has been monitoring the atmospheric radon concentrations continuously for a long time. The monitoring of the atmospheric radon in Korea had been initiated at the Gosan station in Jeju Island as a part of ACE-Asia program in 2001.¹² Since then, the monitoring of the background aerosols as well as radon has been accomplished continuously for over 20 years.¹³ In this study, the radon concentrations had been monitored at the Gosan site of Jeju island during 2017-2018 in real time basis. And from the observation results, the real-time variation characteristics of radon concentrations has been studied, and the

long-range transport pathways of atmospheric pollutants have been investigated by comparing the concentrations and mutual relevancies of radon and gaseous pollutants.

2. Experimental

2.1. Monitoring of radon and gaseous pollutants

The concentrations of the atmospheric radon were measured at the Gosan site of Jeju Island between January 2017 and December 2018, by 30-minutes real-time intervals, using a high sensitive radon detector (Model D1500, 1500 L dual flow loop, two filter radon detector) designed by the Australian Nuclear Science and Technology Organisation (ANSTO). This radon detector measures the alpha particles emitted from radon and its progenies which are collected through the second filter. It is designed to count the number of lightning flashes which are generated by the reaction of alpha particles with ZnS(Ag), using a photomultiplier tube (PMT) detector. The sensitivity of radon detector system used in this study was $0.28\text{--}0.29\text{ counts}\cdot\text{sec}^{-1}/\text{Bq m}^{-3}$, and the low limit of detection was approximately $25\text{--}30\text{ mBq m}^{-3}$.¹⁴

The intake of ambient air was carried out through a inlet pipe (50 mm diameter, HDPE materials) installed at the height of 10 m above the ground level (Fig. 1), and the air flow rate was normally maintained as to approximately 60 L min^{-1} .

The instrumental calibrations for the radon observation were carried out once every month for 5 hours each time. ^{226}Ra source ($18.5\pm 4\%$ kBq, Pylon Electronic Inc., model RN-2000A, Canada) was used as a standard for the monthly calibration of the instrument. The background level of radon concentration was determined by measuring the baseline values of the detector instrument, keeping up with the cut-off of air flow, for 24 hours in every 3 month interval. The background values increase gradually in proportional to the amount of radon progenies such as ^{210}Pb (half-life 22.3 yr.) captured on the secondary filter of the detector as the time progress.

The data sets of gaseous air pollutants (SO_2 , CO , O_3 , NO_2) concentrations were obtained from the 'Air

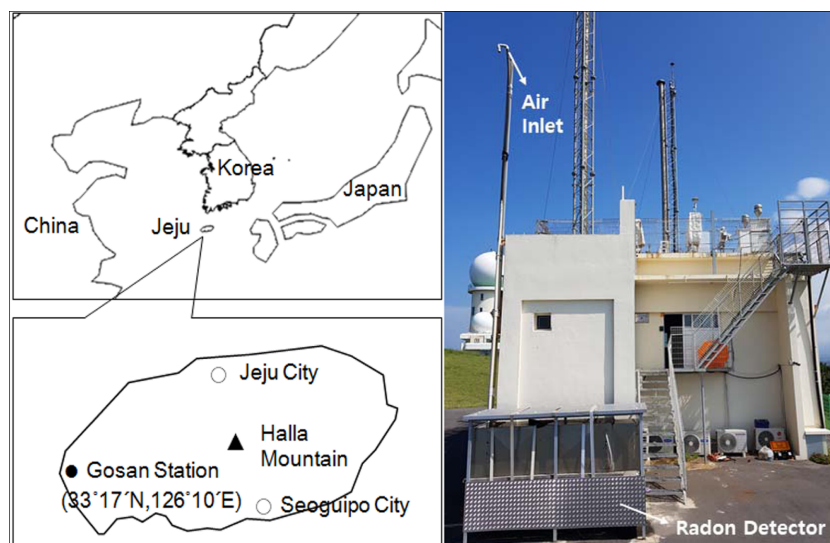


Fig. 1. Location of the Gosan site in Jeju Island and installation of the radon detector (ANSTO, Model D1500).

Korea' web site (<http://www.airkorea.or.kr>), the national air quality information system provided by the Korean Ministry of Environment (MOE) and the Korea Environment Corporation (KECO).

2.2. Back trajectory analysis

The back trajectory analysis has been performed in order to derive the air inflow patterns moved into the Gosan site, with relation to the inflow pathways of atmospheric radon and gaseous pollutants. The back trajectories were calculated by means of the HYSPLIT 4.0 (HYbrid Single-Particle Lagrangian Integrated Trajectory) model, provided by NOAA/ARL (National Oceanic and Atmospheric Administration/Air Resources Laboratory). The meteorological data of GDAS (global data assimilation system), $1^\circ \times 1^\circ$ resolution meteorological model (MRF) of NCEP (National Center for Environmental Prediction), were used for the HYSPLIT back trajectories.¹⁵ The 120-hour back trajectories were carried out starting from the Gosan site ($33^\circ 17'N$, $126^\circ 10'E$) at the altitude of 72 meter above ground level and every 00 UTC (KST 09:00), corresponding to each date of observation for atmospheric radon and gaseous pollutants.¹³ Then the airflow pathways have been classified into 5 clusters by cluster back trajectory.

3. Results and Discussion

3.1. Atmospheric radon concentrations

The atmospheric radon concentrations have been measured by real-time monitoring basis at the Gosan site of Jeju Island between January 2017 and December 2018, and their observation results are shown in Fig. 2. The overall atmospheric mean radon concentration was $2,480 \pm 1,275$ mBq m^{-3} (0.067 pCi/L), which is 29.2 times lower than the mean indoor radon concentration in domestic area of Korea (72.4 Bq m^{-3} , 2.0 pCi/L) and even 27.5 times lower than the indoor radon concentration in Jeju area (68.2 Bq m^{-3} , 1.8 pCi/L).¹⁶

The domestic radon observations in Korea have

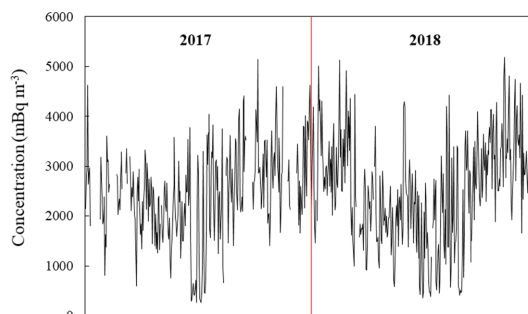


Fig. 2. Variation of the daily mean ^{222}Rn concentrations between January 2017 and December 2018.

focused mostly on monitoring the indoor and the underground area, but the ambient outdoor radon data are not sufficiently reported up to recently, making hard to compare them with those of other domestic places. Unavoidably, we had to compare our observation data with those which had been obtained from the measurements by active-type Electrostatic Radon Monitor at Seoul during December 1999 and January 2002.¹⁷ The radon concentration in Seoul area during that period was $7,620 \pm 4,110 \text{ mBq m}^{-3}$, about three times higher than the observation value at the Gosan site. Furthermore, the radon concentration in Gosan was 3.8, 3.9, 16.2 times lower respectively than those in King's Park Meteorological Station of Hong Kong, L'Aquila of Italy, and the southern capital Bucharest of Rumania,¹⁸⁻²⁰ showing the background area characteristics of quite low radon concentrations. On the other hand, in comparison with other foreign background areas, the radon concentration in the Gosan site was 24.3 times higher than that in Mauna Loa of Hawaii but 2.3 times lower than that in Hok Tsui of Hong Kong.^{21,22} The exceedingly low radon concentration in Hawaii may be reasoned as that the Mauna Loa observation station is located at 3,397 meter high altitude and it is the clean background region quite far from the main continent.

3.2. Time-series variations of radon concentrations

The seasonal mean radon concentrations were in the order of fall ($3,017 \text{ mBq m}^{-3}$) > winter ($2,937$

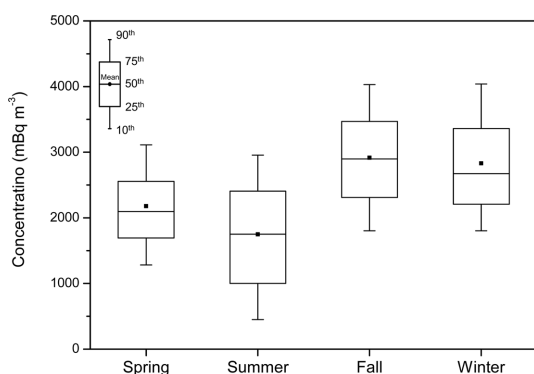


Fig. 3. Seasonal comparison of ²²²Rn concentrations at Gosan site.

mBq m^{-3}) > spring ($2,166 \text{ mBq m}^{-3}$) > summer ($1,939 \text{ mBq m}^{-3}$) during the study period, as shown in Fig. 3. This seasonal discrepancy of radon concentrations could be explained by the fact that the air masses influenced by northwesterly wind would be moved mostly from the China continent during fall and winter seasons, on the other hand, those by southeasterly wind would be moved from the Pacific Ocean during summer.

In addition, it is necessary to consider the mixing depth effect. The mixing depth is the mixing layer height at which the planetary boundary layer is in convectional state and the concentrations of gaseous components are on a vertically homogeneous distribution, and it serves a significant role for accumulation, diffusion and dilution of various atmospheric pollutants.²³⁻²⁴ The mixing depth tends to vary with seasons, generally high in spring and summer and low in fall and winter, due to the temperature variations. And the radon in atmosphere is presumed to show the concentrations reciprocally from the mixing depth variations, well matching to this study results.

Fig. 4 shows the monthly comparison results of radon mean concentrations during the monitoring periods. Additionally in this figure, the median, 90th, 10th percentile concentrations were compared each other to verify the high and low concentration variations. The monthly mean concentrations of radon were in the order of November > January > October > December > February > September > March > June > April > August > May > July, showing the highest

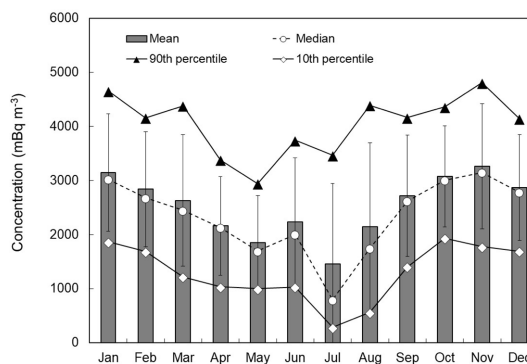


Fig. 4. Monthly variations of ²²²Rn concentrations at Gosan site.

in November ($3,262 \text{ mBq m}^{-3}$) and the lowest in July ($1,459 \text{ mBq m}^{-3}$). The time-series variations from 90th and 10th percentile radon concentrations showed very similar pattern as the mean, except in the case of 90th percentile concentrations in March and August.

The diurnal variations of radon mean concentrations showed the highest ($2,862 \text{ mBq m}^{-3}$) at around 7 a.m. and the lowest ($1,997 \text{ mBq m}^{-3}$) at around 3 p.m., gradually descending from the morning until around 3 p.m. afternoon then again ascending through the evening till the dawn, which was very much similar variation patterns as those at L'Aquila of Italy.¹⁸ This diurnal variation pattern could be inferred to be affected by the change of atmospheric mixing depth and flux. The mixing depth in atmospheric boundary layer (ABL) is high during daytime, and the atmospheric pollutants within this mixing depth are spread as a vertical distribution uniformly by convection. By contrast, during night time, a stable radiation inversion layer lies in the near ground, and then the mixing depth is low due to the relatively small air fluctuation and vertical mixing.²³ Consequently, it could be deduced that the radon concentration was high in daytime due to the maximum dispersion, but conversely it was low in nighttime. Similar to the seasonal and monthly variations, the diurnal variation patterns of radon concentrations were inferred by dispersion and compression of radon in accordance with the mixing depth variations.

The seasonal comparison of diurnal radon variations

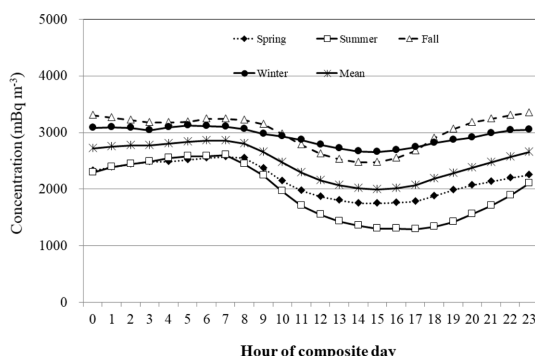


Fig. 5. Seasonal breakdown of the diurnal ^{222}Rn cycles at Gosan site expressed as mean hourly values for the 2-year composite.

has shown that the large discrepancy between daily high and low radon concentrations is exposed during summer. Generally, the temperature difference between day and night is large in summer season, and it causes the high deviation between extension and contraction of the mixing depth, and so comparatively large gaps between the day and night concentrations. In winter season, the concentration differences between day and night were not so big, and it could be reasoned by the opposite accounts, as shown in Fig. 5.²⁵

3.3. Concentration variations of radon and gaseous pollutants

The half-life of ^{222}Rn is 3.82 days, quite similar life time as of major gaseous reactants (SO_2 , CO , O_3 , NO_2 , etc.).⁹ The radon is stable in humidity, temperature and irradiation, and insoluble in water, so that it exhibits quite inert chemical properties. Therefore, the radon is used as a tracer to understand the transport pathways of gaseous air pollutants.

In this study, the diurnal concentration variations of radon and gaseous pollutants (Fig. 6) showed the reciprocally opposite patterns between radon and ozone, that was very similar pattern as in L'Aquila of Italy.¹⁸ On the other hand, the concentrations of CO

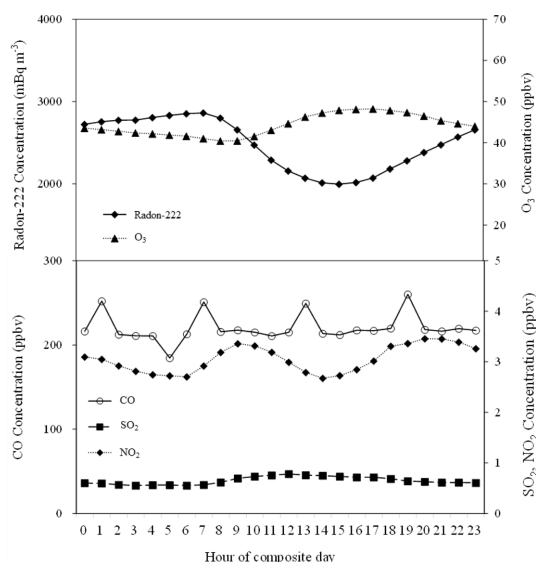


Fig. 6. Concentration variations of the diurnal atmospheric ^{222}Rn and gaseous pollutants.

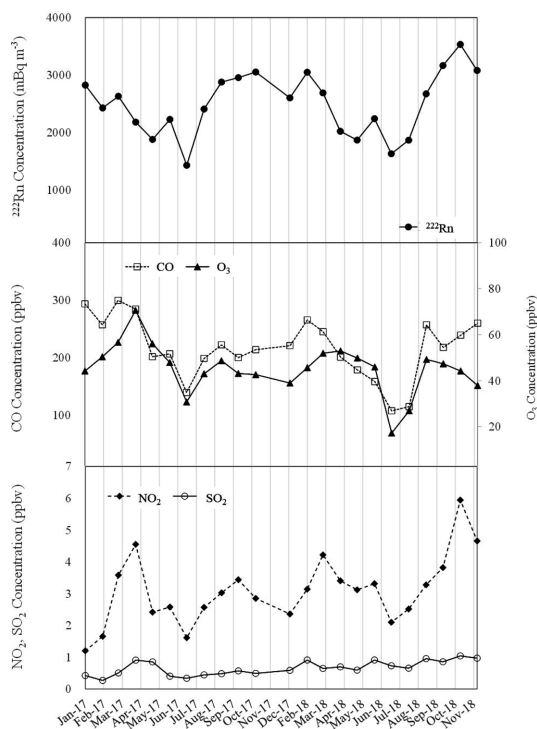


Fig. 7. Monthly concentration variations of atmospheric ²²²Rn and gaseous pollutants at Gosan site during 2017-2018.

and NO₂ had much similar variation pattern as radon concentrations. The monthly concentration variations (Fig. 7) showed the overall resemblance between CO, O₃, NO₂ and ²²²Rn with a little exceptional divergence. The correlation coefficients of radon with CO and NO₂ were 0.63 and 0.52 respectively, showing good correlation relations. Especially, the correlation coefficient of radon with NO₂ excluding the three months data (January-March, 2017) was improved to 0.66, showing quite high correlations. However, the correlations of radon with SO₂ and O₃ were not so high comparatively. Consequently, the monitoring of atmospheric radon concentrations could be applicable effectually as a tracer of possible transport pathways of gaseous pollutants such as CO and NO₂.

3.4. Concentration comparison related to the airflow pathways

Radon is naturally occurred by the radioactive decay of uranium existed in soil or rocks, and it is

transferred into the atmosphere with the airflow stream. It is chemically inactive in air, and poorly soluble in water, not so as to wash out by rain. The atmospheric radon concentrations are more than three times higher in the air above the land rather than above the oceanic area. Furthermore, the life time of ²²²Rn is similar to those of some atmospheric pollutants, so it can be a suitable tracer to investigate the transport pathways of NO₂, SO₂, CO, O₃, etc. in the air.^{9,19}

In this study, the long-range transport pathways of radon were investigated by 120-hour cluster back trajectory analysis with the application of NOAA HYSPLIT model and NCEP GDAS meteorological data, as shown in Fig. 8.

The airflow pathways moved into the Gosan site were classified to five parts according to the cluster back trajectories; Cluster 1 (northern China), Cluster 2 (Korean Peninsula), Cluster 3 (eastern China), Cluster 4 (Japan), and Cluster 5 (northern Pacific). In the results, the frequencies of each cluster out of overall trajectories were 29 % (156 days), 17 % (91 days), 31 % (165 days), 15 % (80 days), and 9 % (48 days), respectively, so that it has shown about 60 % of all airflow pathways might be from China continent during the study period.

The concentrations of ²²²Rn and major atmospheric pollutants (SO₂, CO, O₃, NO₂) were compared on the basis of back trajectory clusters analysis, as in Table 1. The concentrations of radon and SO₂ derived through the pathways of northern China (Cluster 1) were 2,849 mBq m⁻³ and 0.73 ppbv, respectively, high values compared to those of other pathways. The concentrations of CO, O₃, NO₂ derived through the eastern China (Cluster 3) were respectively 236.8 ppbv, 51.3 ppbv, and 3.6 ppbv, showing comparatively higher values than from other clusters. It was revealed that radon and other atmospheric pollutants showed high concentrations in the airflow pathways derived from China, but relatively low concentrations in the pathways from the northern Pacific Ocean. Consequently, it could be confirmed that the air quality of Jeju Island might be seriously affected by several gaseous pollutants from the China continent.

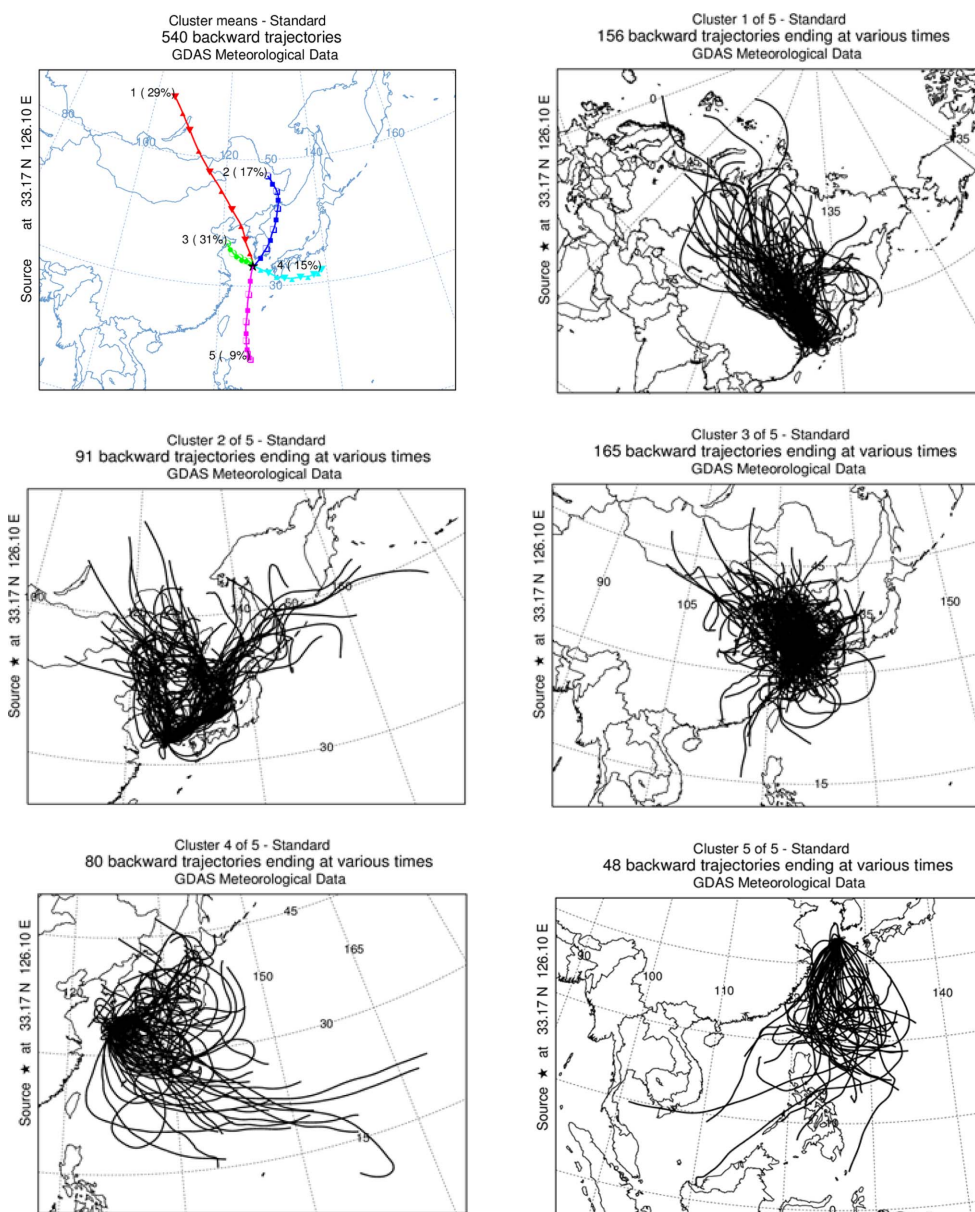


Fig. 8. Cluster back trajectories of air masses corresponding to ^{222}Rn monitoring data at Gosan site during 2017-2018.

Table 1. Statistics of atmospheric ^{222}Rn and gaseous pollutants concentrations classified by cluster back trajectory.

Cluster	^{222}Rn (mBq m^{-3})	SO_2 (ppbv)	CO (ppbv)	O_3 (ppbv)	NO_2 (ppbv)
Cluster 1 (n=156)	2,849	0.73	232.8	47.6	3.3
Cluster 2 (n=91)	2,597	0.69	192.6	45.1	3.4
Cluster 3 (n=165)	2,632	0.70	236.8	51.3	3.6
Cluster 4 (n=80)	1,884	0.66	152.0	33.6	2.8
Cluster 5 (n=48)	1,190	0.36	132.5	28.6	1.7

4. Conclusions

In this study, the concentrations of ^{222}Rn and gaseous atmospheric pollutants at the Gosan site of Jeju Island had been monitored for 2017-2018, and their concentration characteristics by time-series variation and airflow pathways has been investigated. The atmospheric radon concentration for the study period was $2,480 \text{ mBq m}^{-3}$, about 28 times lower than the indoor radon concentration in Jeju Island. The monthly variations showed the highest ($3,262 \text{ mBq m}^{-3}$) in November and the lowest ($1,459 \text{ mBq m}^{-3}$) in July, indicating about 2.2 factors difference between them. Seasonally, the high radon concentrations were observed during fall and winter, but relatively low for summer season. The diurnal variations showed the maximum radon concentrations at around 7 a.m. and the minimum at around 3 p.m., implying a close dependence upon the atmospheric mixing depths. The mean concentrations of gaseous pollutants SO_2 , CO , O_3 , NO_2 were respectively 0.7, 206.4, 44.5, 3.2 ppbv, showing roughly similar monthly pattern of radon except the variation of SO_2 . The diurnal variation of ozone concentrations was opposite to the radon characteristics.

The concentrations of radon and gaseous pollutants classified by the airflow pathways were compared by cluster back trajectory analysis, and it was revealed the high concentration values in the airflow pathways through China and Korean peninsula and the low concentrations in through the northern Pacific Ocean. From this study, it would be suggested that the atmospheric radon could be used as a useful tracer for identifying the transport pathways of gaseous pollutants moving into Jeju area.

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