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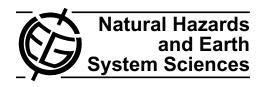
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## Anomalous radon emanation linked to preseismic electromagnetic phenomena

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**Abstract.** Anomalous emanation of radon (<sup>222</sup>Rn) was observed preceding large earthquakes and is considered to be linked to preseismic electromagnetic phenomena (e.g. great changes of atmospheric electric field and ionospheric disturbances). Here we analyze atmospheric radon concentration and estimate changes of electrical conditions in atmosphere due to preseismic radon anomaly. The increase of radon emanation obeys crustal damage evolution, following a power-law of time-to-earthquake. Moreover, the radon emanation decreases the atmospheric electric field by 40%, besides influencing the maximum strength of atmospheric electric field by  $10^4$ – $10^5$  V/m enough to trigger ionospheric disturbances. These changes are within the ranges observed or explaining electromagnetic phenomena associated with large earthquakes.

### 1 Introduction

Electromagnetic phenomena associated with large earth-quakes have been observed for many years. For example, earthquake light (Derr, 1973; Kamogawa et al., 2005), electromagnetic radiations (Gokhberg et al., 1982; Nagao et al., 2002), great changes of atmospheric electric field (Kondo, 1968; Vershinin et al., 1999) and ionospheric perturbations (Molchanov and Hayakawa, 1998; Liu et al., 2000) are well known. Among them, ionospheric perturbations have attracted many researchers and have been investigated for earthquake predictions (Hayakawa et al., 2004; Pulinets and Boyarchuk, 2004). Gokhberg et al. (1989) and Molchanov and Hayakawa (1998) pointed out the precursory ionospheric anomalies associated with large earthquakes, and Maekawa et al. (2006) emphasized the verification of the existence from a statistical analysis of the subionospheric LF propa-

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gation data (Kamogawa, 2007). Since lithosphere does not have a direct connection with the ionosphere, problems related to seismo-ionospheric disturbances are treated by considering the interaction among the lithosphere, atmosphere and the ionosphere, i.e., the LAI coupling (Hayakawa et al., 2004; Kamogawa, 2006).

On the other hand, for modeling ionospheric disturbances within the regime of the LAI coupling, radon emanation that causes changes in the atmospheric electrical properties has been focused on recently (e.g. Sorokin et al., 2001; Hayakawa et al., 2004; Pulinets and Boyarchuk, 2004; Rapoport et al., 2004; Kamogawa, 2006; Pulinets, 2007). Many investigations have revealed the anomalous radon emanation out of the lithosphere into the near-earth atmosphere (King, 1978, 1986) and the ground water (King, 1986; Igarashi et al., 1995), and United Nations Scientific Committee on the Effects of Atomic Radiation (2000) also reported the same things. In case of radon in the atmosphere, Okabe (1956) observed the increase of radon content at the near-surface atmosphere after earthquakes at Tottori, Japan, while nobody clearly shows preseismic variations of radon which are required for estimating the atmospheric electrical properties. Although Pierce (1976) pointed out the importance of the radon behavior to the preseismic change of the atmospheric electric field, there is no quantitative analysis of radon concentration measured in the atmosphere, among the various studies dealing with radon effects as part of the LAI coupling. Thus it is necessary to verify radon emanation into atmosphere, enough to trigger ionospheric perturbations as part of the LAI coupling, prior to earthquakes.

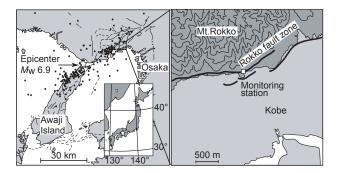
In this paper, we clarify whether the radon emanation triggers preseismic electromagnetic phenomena such as an anomaly in the atmospheric electric field causing ionospheric perturbations. First, we show that anomalous radon emanation reflects the damage process in the crust associated with large earthquakes. Next, we introduce the knowledge of atmospheric electricity, and investigate the atmospheric

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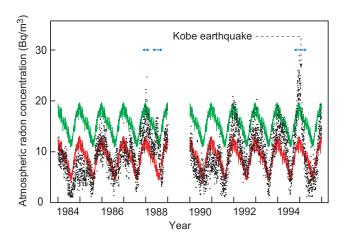
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**Fig. 1.** Location of the radon monitoring station and the distribution of active faults and epicenters in Kobe. Square shows the radon monitoring station. Lines, star and solid circles indicate active faults and epicenters of the Kobe earthquake and its aftershocks (magnitude  $M \ge 3.0$ ) until 31 January 1995, respectively. The distribution of active faults and the epicenters of aftershocks were obtained from the Research Group for Active Faults of Japan (1991) and Japan University Network Earthquake Catalog Hypocenters Files (http://wwweic.eri.u-tokyo.ac.jp/db/junec/index-j.html), respectively.



**Fig. 2.** Temporal variation of the atmospheric radon concentration. The data were sampled from 1984 to 1995, except for 1989 and during the 11 days just after the Kobe earthquake. The filled circles depict the time variation of the daily mean of the hourly radon concentration. The red and green lines indicate the temporal change in the averages of the daily mean values from January 1984 to December 1993 except for 1989 and their  $3\sigma$  range, respectively. Blue two-headed arrows represent seismic related anomaly in atmospheric radon concentration.

electrical conditions that contain the atmospheric electric field altered by preseismic radon emanation. For this purpose, the data on atmospheric radon concentration measured around the time of the Kobe earthquake (17 January 1995, Japan; moment magnitude  $M_w$ =6.9) are used.

### 2 Location and measurement procedure

The site investigated is located in Kobe immediately above the Rokko fault zone, which is a part of the fault zones that caused the Kobe earthquake and the aftershocks, situated about 25 km away from the epicenter (Fig. 1). The northern part of the site around the monitoring station is known as the Rokko mountain. It is composed mainly of the Nunobiki granodiorite and the Rokko granite (Huzita and Kasama, 1982), generally containing more <sup>238</sup>U (Wollenberg and Smith, 1987). <sup>238</sup>U is a parent nuclide of <sup>226</sup>Ra which decays into radon. Therefore, the monitoring station is situated in a region where radon gas is released along the fault into atmosphere.

The measurements of atmospheric radon concentration had been running since 1984, except for the lack of observations in 1989 and also during the 11 days just after the Kobe earthquake. In order to determine the atmospheric radon concentration, we used a flow-type ionization chamber having a volume of  $1.8 \times 10^{-2}$  m³ (Model NAG513, Fuji Electric Systems Co., Ltd.). Air, 5 m above the ground, was passed through a prefilter and a high efficiency particulate air filter with a ventilation rate of  $15.6 \, \text{m}^3 / \text{min}$ , for retrieving the radon progenies which form the aerosols. A part of the filtered air was pumped into the ionization chamber with a flow rate of  $6.7 \times 10^{-3} \, \text{m}^3 / \text{min}$ . The measurements were carried out every hour, continuously and automatically (Yasuoka and Shinogi, 1997; Yasuoka et al., 2006).

### 3 Data analysis and discussions

### 3.1 Preseismic radon emanation into atmosphere

Figure 2 shows the temporal variation in the atmospheric radon concentration between 1984 and 1995. One can see that the daily mean of the radon concentration presents a seasonal change, having a minimum in summer and a maximum in winter. This is due to atmospheric turbulence (Gesell, 1983) and the annual wind rose (Porstendörfer, 1994). The former points to the fact that the vertical mixing is smaller due to low solar heating in winter, than in summer (Liu et al., 1984). The latter indicates that continental air mass such as the Siberian air mass in Japan brings radon, originated elsewhere than Kobe, during winter.

Figure 2 also shows the anomalous increase in radon concentration during three periods; winter in 1987–1988, summer in 1988 and winter in 1994–1995 just before the Kobe earthquake. The former two correspond to a somewhat seismic-activated period. Mogi (1995) pointed out that the seismic activity had decreased, i.e., a seismic gap had been formed in Kobe area in the mid 1960s or later. However, the activity with magnitude (M) 3 began to increase in the late 1980 (Mogi, 1995), especially, that was remarkable in the northern part of Awaji Island in 1986-1989 (Katao and

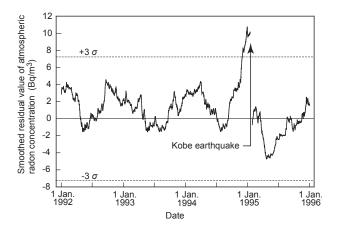
Ando, 1996; Watanabe, 1998). On the other hand, during the third period, i.e., just before the Kobe earthquake, the atmospheric radon concentration can be seen to be located far beyond  $3\sigma$  range ( $\sigma$ : standard deviation) for the successive days during the two months. Moreover, the similar tendency is shown from the analysis of the daily minimums (Fig. 2 in Kawada et al., 2007). Therefore we consider the anomaly during the three periods to be seismic related. Below, we handle the example in the 1995 Kobe earthquake.

For quantifying the unusual concentration of radon in the atmosphere, we analyzed the change in smoothed residual and cumulative values of concentration, from 1 September 1994 to 16 January 1995 (Figs. 3 and 4). By the use of the exponential smoothing method, the meteorological fluctuations of atmospheric radon are minimized. In addition, for excluding the seasonal effect, the residual is defined as the difference between the daily minimum and the smoothed normal variation of the minimums of atmospheric radon concentration for each day (Yasuoka et al., 2006) where the daily minimum represents the background radon concentration. The smoothed residual values are plotted above the  $3\sigma$  range, as well as the daily averages preceding the Kobe earthquake. During the period from 24 December 1994 to the time of the earthquake, the residual values can be seen to have attained magnitudes of up to about 10 Bq/m<sup>3</sup> (Fig. 3). Figure 4 shows the temporal variation of the cumulative residual values of atmospheric radon. Each cumulative residual value is expressed by summing up the residual value from 1 September 1994 to each day and finally to 16 January 1995. The variation of the cumulative value has a power-law dependence formulated as

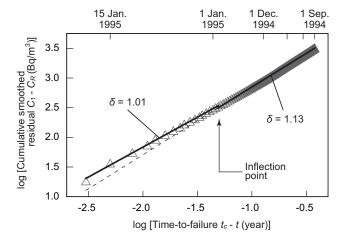
$$\sum (C_1 - C_R) = C_2 (t_c - t)^{\delta}, \tag{1}$$

or 
$$\sum C_R = C_3 - C_2 (t_c - t)^{\delta}$$
, (1')

where  $C_R$ , t, and  $t_c$  are the smoothed residual value of atmospheric radon concentration, time, and the occurrence time of the Kobe earthquake, respectively (see also Kawada et al., 2007). The other variables,  $C_1$ ,  $C_2$ ,  $C_3$ , and  $\delta$  are constants. These constants can be seen to change approximately 17 days prior to the earthquake (each value of  $t_c$ ,  $C_1$ ,  $C_2$ ,  $C_3$ , and  $\delta$  is denoted in the caption of Fig. 4). Since the smoothed residual value excludes the seasonal effects (Yasuoka et al., 2006), the increase following a power-law reflects crustal deformation. This temporal power-law change is similar to cumulative Benioff strain release which represents the summation of the square root of the energy released for sequential fracture events, prior to the time to the collapse. The change relates to the damage process in the crust (e.g. Ben-Zion and Lyakhovsky, 2002; Turcotte et al., 2003), and being a critical phenomenon prior to large earthquakes, shows a time-scale invariance, i.e., the temporal power-law (Kawada and Nagahama, 2006; Kawada et al., 2007). Furthermore, the inflection point in Fig. 4 is emerged, and the determination of this



**Fig. 3.** Temporal variation of the smoothed residual value of atmospheric radon concentration. Broken lines represent  $\pm 3\sigma$  range. This figure is modified from Kawada et al. (2007).



**Fig. 4.** Temporal variation of the cumulative smoothed residual of atmospheric radon concentration. The fit follows power-laws given in Eq. (1) or (1'), and the constants change on 31 December 1994. The constants of the respective equations are  $C_1$ =2.86×10 Bq/m³,  $C_2$ =9.58×10³ Bq/m³/year³,  $C_3$ =3.95×10³ Bq/m³,  $\delta$ =1.13 and  $\delta$ =1.13 and  $\delta$ =1.13 and  $\delta$ =1.13 bq/m³/year³,  $\delta$ =1.13 and  $\delta$ =1.14 bq/m³/year³,  $\delta$ =1.15 and  $\delta$ =1.16 bq/m³/year³,  $\delta$ =1.17 and  $\delta$ =1.18 bq/m³/year³,  $\delta$ =1.19 bq/m³/year³/y

is likely to enable us in predicting earthquakes. The crustal damage evolution in the critical state promotes radon migration and emanation into the atmosphere, constrained the power-law increase of the atmospheric radon concentration.

## 3.2 Changes of atmospheric electrical conditions due to radon emanation

Radon emanation into the atmosphere can cause ionization of atmospheric gases. The atmospheric electrical conditions are basically constrained by the concentration of small ions (Chalmers, 1957; Pierce, 1976) and these can be altered near

**Table 1.** The values of the constants in the text.

Constants	Values	References
e	1.6×10 <sup>−19</sup> C	
k	$1.5 \times 10^{-4} \text{ m}^2/\text{V/s}$	Pierce (1976)
N	$10^{10}/\text{m}^3$	Junge (1963)
α	$(1.4-1.6)\times10^{-12}\mathrm{m}^3/\mathrm{s}$	Pierce (1976), and
		Kulkarni and Kamra (2001)
β	$(3.0-4.0)\times10^{-12}\mathrm{m}^3/\mathrm{s}$	Pierce (1976), and
		Kulkarni and Kamra (2001)
$\varepsilon_0$	$8.9 \times 10^{-12} \text{F/m}$	
R	$\leq 10 \mu\mathrm{m}$	Liperovsky et al. (2005)
$\rho_a$	$(1.0-2.0)\times10^3 \text{ kg/m}^3$	Junge (1963)
$N_a$	$10^9 / \text{m}^3$	Sorokin et al. (2001), and
		Liperovsky et al. (2005)
Q	2000e	Liperovsky et al. (2005)
g	$9.8  \text{m/s}^2$	
$\eta$	$1.7 \times 10^{-5} \text{ kg/m/s}$	

ground by radon, its progenies and radioactivity in the earth, producing ion-pairs in air. We now discuss the concentration of small ions and the atmospheric conditions such as the atmospheric conductivity and the electric field preceding the Kobe earthquake below.

#### 3.2.1 The concentration of small ions

The concentration of small ions near ground is controlled by the ionization rate, the recombination of positive and negative ions, and the attachment of small ions to the charged aerosols (e.g. Chalmers, 1957; Pierce, 1976; Liperovsky et al., 2005). Hence, one can construct the equation regarding the temporal change of small ion concentration as

$$\frac{\partial n}{\partial t} = q - \alpha n^2 - \beta n N,\tag{2}$$

and the estimate for the small ion concentration in the steady state as

$$n = \frac{-\beta N + \sqrt{(\beta N)^2 + 4\alpha q}}{2\alpha}.$$
 (3)

Here q is the ionization rate obtained from cosmic rays, radioactivity in the atmosphere including radon and the radiation from soil, N is the aerosol density,  $\alpha$  is the recombination coefficient and  $\beta$  is the attachment coefficient of small ions to charged aerosols (see also Table 1). We take the ionization rate of cosmic rays to be  $1.9\times10^6$  ion-pairs/m³/s at 35° N and that of the radiation from soil to be  $4.5\times10^6$  ion-pairs/m³/s (Mohnen, 1977). A radon concentration of 10 Bq/m³ corresponds to an ionization rate of  $(3.9-5.5)\times10^6$  ion-pairs/m³/s, from observations (e.g. Nagaraja et al., 2003) and calculation (see Appendix A). Then, at Kobe, the normal concentration of small ions in the atmosphere  $(n_0)$  was evaluated to be  $(2.6-3.9)\times10^8/\text{m}^3$ , from an atmospheric radon

concentration of  $10 \, \text{Bq/m}^3$ , during winter (Fig. 2). This estimation is comparable to the concentration measured in Kobe (Tsunoda and Satsutani, 1976). When the remarkable radon emanation occurred before the Kobe earthquake, i.e., during the period of more than  $10 \, \text{Bq/m}^3$  increase in the atmosphere (Fig. 3), the small ion concentration was seen to have varied from  $(2.6–3.9)\times10^8 \, / \text{m}^3$  to  $(3.5–5.7)\times10^8 / \text{m}^3$ . This increment was supported by the increase of small ion density by  $(1.0–1.4)\times10^8 / \text{m}^3$  before the earthquake (Satsutani, 1996, 1999).

### 3.2.2 The atmospheric electrical conditions

The basic elements of atmospheric electricity are the atmospheric conductivity, the atmospheric electric field and the air-earth current. The atmospheric conductivity affects the atmospheric electric field and air-earth current (Makino and Ogawa, 1984). The relationship among them has Ohm's law (Chalmers, 1957; Pierce, 1976). Hence, the atmospheric conductivity is useful for discussing atmospheric electrical conditions prior to the Kobe earthquake.

Because the small ions migrate easier than the large ones, the atmospheric conductivity is proportional to the concentration of small ions and given by

$$\lambda \approx 2ekn,$$
 (4)

where e is the electron charge, and k is the mobility of the small ions at the ground level (Pierce, 1976 and see Table 1). From Eq. (4) and the small ion concentration mentioned above, the atmospheric conductivity in normal winter  $(\lambda_0)$  and preceding the Kobe earthquake  $(\lambda)$  are (1.2- $1.9 \times 10^{-14} / \Omega / m$  and  $(1.7-2.7) \times 10^{-14} / \Omega / m$ , respectively. That is, the preseismic radon emanation increases the atmospheric conductivity by about 40%. And in turn, according to the constancy of air-earth current in Ohm's law, the atmospheric electric field decreases by about 40%. The Matsushiro earthquake swarm (1965, Japan) formed many fractures, and anomalous variation in atmospheric electric field was observed (Kondo, 1968), as well as earthquake lights (Derr, 1973). High densities of the positive ions were proposed to influence the abnormal animal behavior preceding earthquakes (Tributsch, 1978). Crustal damage evolutions prompt radon emanation into the open air, which is responsible for the alteration of the atmospheric electric field and the densities of positive ions by ionizing the aerial gases.

On the other hand, radon emanation induces temporal evolution of large atmospheric electric fields under non-stationary conditions as compared to stationary. We can define these non-stationary and stationary conditions as having the presence and absence of a characteristic time, as denoted by

$$\tau \approx \frac{\varepsilon_0 n_0}{\lambda_0 n},\tag{5}$$

where  $\varepsilon_0$  is the permittivity constant,  $\lambda_0$  is the undisturbed atmospheric conductivity near the ground and n is the dis-

turbed value of ion concentration, that depends on the ionization rate, respectively. The difference in the mobility and the size distribution of the positive and negative ions generate the pulses of local electric fields (Liperovsky et al., 2005). One can quantify the maximum amplitude of the electric field pulses by

$$E_g = \frac{2R^2 \rho_a N_a Qg}{9\eta \left\{ \lambda_0 \frac{n}{n_0} + \frac{Q^2 N_a}{6\pi \eta R} \right\}},\tag{6}$$

where R is the large aerosols' radius,  $\rho_a$  is the aerosol mass density based on the general chemical composition of natural aerosols,  $N_a$  is the large aerosol concentration, Q is the large aerosol charge, g is the gravitational acceleration, and  $\eta$  is the viscosity of air at 0°C (We modify the equation in Liperovsky et al., 2005. Each value of R,  $\rho_a$ ,  $N_a$ , Q, g and  $\eta$  is given in Table 1). Based on the radon concentration and the estimated ion density, we can show that the maximum amplitude  $(E_g)$  was up to about  $10^4$ - $10^5$ V/m. These ranges, even in fair weather, were found to be nearly equal to those under thunder clouds and the characteristic time  $(\tau)$  in Eq. (5) was  $(3.3-5.3)\times10^2$  s, for an atmospheric radon concentration of 10 Bq/m<sup>3</sup>, prior to the Kobe earthquake. During the earthquake, a decrease of the reflection height in the D region (around 50-90 km in ionospheric altitude) was observed, determined from VLF transmission anomaly (Molchanov and Hayakawa, 1998; Molchanov et al., 1998). When the atmospheric conductivity near the ground increases by a factor of 2, the ionospheric perturbation can be explained in terms of the induction of the electrostatic field by  $(0.5-1.5)\times10^3$  V/m (Grimalsky et al., 2003; Rapoport et al., 2004). These values were within our estimation, and thus the pulses can trigger lowering the reflection altitude. Therefore radon emanation is essential for the preseismic ionospheric disturbances and they are detectable via VLF transmission.

The calculated changes in the atmospheric conductivity and electric fields under stationary and non-stationary conditions were obtained only from the contribution of radon and its shorter half-life progenies. We ignore the variations of thoron (<sup>220</sup>Rn) and its daughter nuclides and the radiation from soil. Additional measurements of thoron and the radiation from soil would be required in future.

### 4 Conclusions

We have considered the effects of radon on the preseismic electromagnetic phenomena by use of the data on the anomalous concentration of atmospheric radon measured around the time of the Kobe earthquake. The cumulative residual values of the radon concentration were found to follow temopral power-law of time-to-the earthquake. This increase responded to the damage process in the crust associated with large earthquakes. In addition, our quotations for atmospheric electrical conditions altered due to radon emanation

were comparable with those from the acutual observations and the estimations in large earthquakes. Hence, seismic related anomaly in radon emanation can be linked to presesmic electromagnetic phenomena such as the great change of small ion concentrations and atmospheric electric field and ionospheric perturbations. These effects are applicable for the prediction of large earthquakes.

The present study indicates that ionization due to radon emanation into atmosphere plays a very important role in the LAI coupling scheme associated with earthquakes.

### Appendix A

### Ionization rate of radon

When radon emanates from the ground, they (radon and its progenies) produce ion-pairs in the atmosphere. It is polonium ( $^{218}$ Po,  $^{214}$ Po) that mainly contributes to ionizing aerial gases, from amongst the various daughter nuclides of radon because of its decay accompanied by  $\alpha$ -particles. Then, the ionization rate can be quantified by the ratio of the energy of  $\alpha$ -particles emitted by radon and its progenies to the average energy (35eV) required for producing an ion-pair in atmosphere. This can be formulated as

$$q_r = \frac{5.49 \times 10^6 C_r + 6.00 \times 10^6 C_{P1} + 7.69 \times 10^6 C_{P2}}{35},$$
 (A1)

where  $C_r$ ,  $C_{P1}$  and  $C_{P2}$  are the concentrations of radon, <sup>218</sup>Po and <sup>214</sup>Po, respectively. Assuming a secular equilibrium between <sup>214</sup>Bi and <sup>214</sup>Po, the ratio of  $C_r$ ,  $C_{P1}$  and  $C_{P2}$ , averaged over time, was evaluated to be 1:0.7:0.5 (see Fig. 11 in Porstendörfer, 1994). Because the range of  $\alpha$ -particles is much larger than the radius of the aerosols, the attachment of the nuclides to aerosols does not influence the ionization of atmosphere. Hence, when atmospheric radon concentration is  $10\,\mathrm{Bq/m^3}$ , we can estimate the ionization rate of radon to be  $3.9 \times 10^6$  ion-pairs/m³/s.

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