

On the Effects of Air Pressure Variations on Radon Exhalation into the Atmosphere

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Abstract—A model is studied which claims to describe the effects of air pressure variations on the outflow of radon into the atmosphere from loose deposits that are treated as a porous medium having the gas dynamic properties described by Darcy's law. It is shown that the model results are in satisfactory agreement with the experimental data acquired from a network for monitoring subsoil radon in the Petropavlovsk-Kamchatskii Geodynamic Site in 1997–2006. A technique has been developed for estimating the Darcy coefficient based on radon concentration data in loose deposits at various depths.

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INTRODUCTION

The subsoil air contains a mixture of air and geologic gas; this contains such gases as H₂, CO₂, CH₄, He, etc., and also radon (²²²Rn), which is a natural ionizer of near-ground air. Variations in the 3D activity of radon (3DA Rn) affect the electrical characteristics of the near-ground atmosphere, which may be immediate-term precursors (with advance times of a day or less) before large Kamchatka earthquakes [6, 7]. It is therefore of great interest to obtain information on all the factors that affect the outflow of Rn into the atmosphere. The possible mechanisms operative in the mass transfer of geologic gases are reviewed in detail in [12]. The focus in the present paper is on the study of how air pressure variations affect the exhalation of radon from loose deposits.

The effects of air pressure on the dynamics of subsoil Rn were noted in the first emanation surveys which became very popular in the 1950s as a tool in the search for radioactive ore. The many available publications on this topic [1, 2, 5, 13] provide explanations for why air pressure variations should affect the exhalation of radon from loose homogeneous deposits, the mass transfer of gases in loose deposits being described by the “gas attack equation” which involves both a diffusion term and a convective (advective) one corresponding to free flow. There is no free flow in a porous medium (where pore size is small compared with the typical size of the medium itself) which is here assumed to be eluvial-deluvial deposits that are typical of radon monitoring points in the Petropavlovsk-Kamchatskii Geodynamic Site. It is the velocity which is proportional to the pressure gradient in such a medium rather

than the acceleration, as is the case with free flow. The contribution due to the force of gravity is small too, because the length of perturbation waves in a porous medium is small compared with the height of a homogeneous atmosphere. Here we attempt to refine our understanding of the physical nature of that relation when eluvial-deluvial rock is the porous medium. The air pressure variations can be taken into account with the help of statistical analysis [4], similarly to the way this is done in [10], but it is also advisable to consider a model based on a physical cause–effect relationship, which may prove useful in the monitoring of subsoil radon with a view to the prediction of large Kamchatka earthquakes.

A BRIEF REVIEW OF THE OBSERVATION SYSTEM. THE RAW DATA

The network of observation stations for monitoring the dynamics of radon in subsoil air is deployed in the Petropavlovsk-Kamchatskii Geodynamic Site. The instrumentation and measurement procedures are described in detail in [10, 11]. Gas-discharge counters for beta radiation at nearly all sites were installed at two depths, one and two meters, which allowed monitoring of the time-dependent behavior of kinematic and dynamic parameters that characterize the Rn content in subsoil air at different depths and enabled us to compare them with air pressure variations. In addition, two stations (Paratunka, denoted PRT, and Institute, denoted INS) recorded radon concentration at the ground surface, as well as air pressure and temperature. The sampling rate for all parameters recorded at PRT was 2.0 h^{−1}.

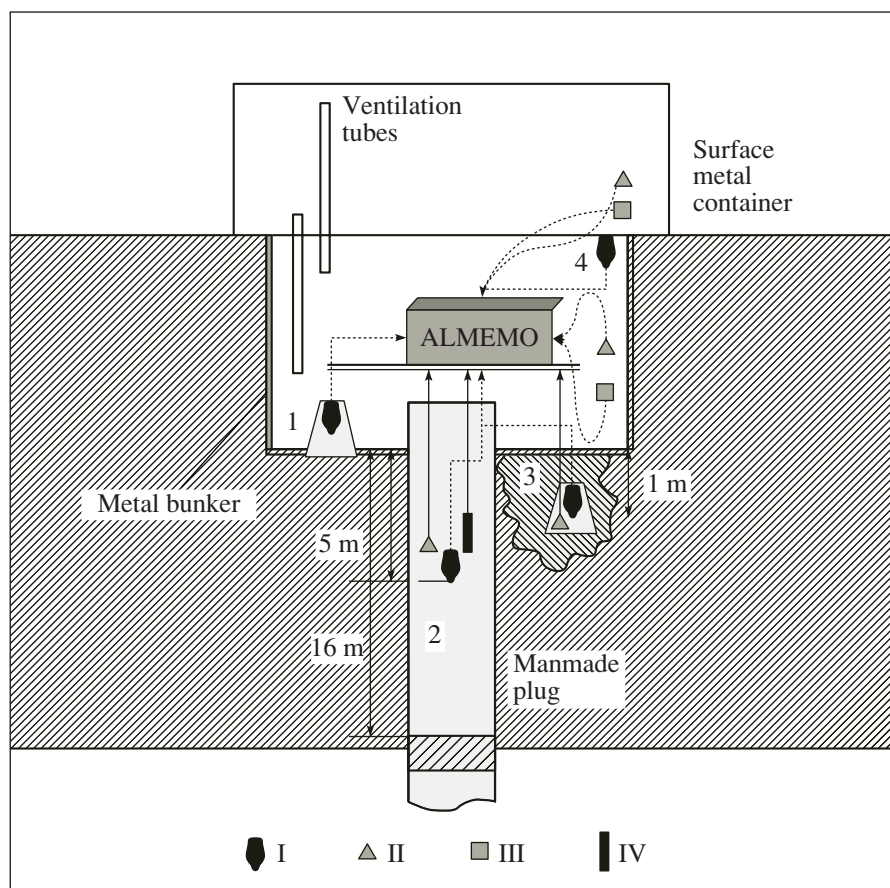


Fig. 1. Sensor locations in the NIS-1 well: (I) gas-discharge counters for beta radiation, (II) temperature sensors, (III) pressure sensors, (IV) carbon dioxide sensor.

Of special interest in this study of air pressure variations as affecting radon exhalation from loose deposits is the observation site INS which used the NIS-1 well near the building of the Institute of Volcanology and Seismology, Far East Division, Russian Academy of Sciences. As to the tectonic and geological position of the well (depth 350 m), it is drilled within the Petropavlovsk horst in a buried depression that is overlain by Late Pleistocene volcanogenic sediments. The well is in the rock mass deposited by a directional explosion of Avacha Volcano (0–180 m depth) and in marine boulder-pebble deposits (180–330 m). The well is encased with steel tubes throughout its depth and an aperture filter is installed in the depth range of 265–295 m. The water is hydrocarbonate and contains sodium and calcium. During the period of December 1979 to June 1993 the water level was fluctuating within 51.0–52.5 m, this variation being seasonal (oral communication by Yu.M. Khatkevich).

At present a manmade “plug” is present at a depth of 16 m in the well. A $2 \times 2 \times 2$ m³ underground bunker has been built above the well cap with a metal container installed above this on the ground surface. The bunker is ventilated by natural air convection through

the container and a set of tubes (Fig. 1). The erection of this structure was supervised by V.A. Gavrilov and O.P. Rulenko.

Four gas-discharge counters were installed in the bunker to study radon mass transfer. The sensors were located in the NIS-1 well as shown in Fig. 1:

- (1) in the well shaft at a depth of 2.5 m from the cap edge;
- (2) in the zone of aeration at a depth of one meter from the floor;
- (3) in a storage tank installed on the floor;
- (4) in the air of the underground bunker.

In addition, a sensor was installed at a depth of one meter in the well shaft to record the concentration of carbon dioxide. The surface container has a temperature sensor and an air pressure sensor, while the bunker contains three temperature sensors and a pressure sensor.

All the parameters were recorded by two measuring devices, ALMEMO 2390-8 and ALMEMO 2590-9, at a sampling rate of 6.0 h⁻¹.

A MODEL DESCRIBING THE EFFECT OF ATMOSPHERIC PRESSURE VARIATIONS ON THE OUTFLOW OF RADON INTO THE ATMOSPHERE

We consider the loose deposits as a 1D porous medium filled with subsoil air (geologic gas) whose rate of flow obeys Darcy's law [3, 8].

$$V = -K/\mu \times \partial P / \partial z, \quad (1)$$

where K is the coefficient of permeability for the loose deposits, μ the dynamic viscosity of air, $\partial P / \partial z$ the pressure gradient in the subsoil air, provided $\partial P \ll P_0$, where P_0 is the air pressure at the ground surface.

It follows from the equation of continuity that the rate of change for the number of molecules in subsoil air (n) is given by

$$\partial n / \partial t = -n_0 \operatorname{div} V. \quad (2)$$

Here, n is the density of air molecules in the loose deposits, equal to $n = p/C^2 M$, where C is the speed of sound in the atmosphere, M is the mean molecular mass of subsoil air, and n_0 the number of air molecules per unit volume at normal pressure. Denote by D the quantity $Kc^2 Mn_0/\mu = KC^2/\nu$, where $\nu = \mu/Mn_0$ is kinematic viscosity. Equation (1) can then be rewritten as

$$n_0 V = -D \partial n / \partial z. \quad (3)$$

Substitution of (3) in (2) yields the equation of one-dimensional diffusion with the diffusion coefficient D :

$$\partial n / \partial t = D \partial^2 n / \partial z^2. \quad (4)$$

The solution to (4) will be sought in the form

$$n = \Sigma [a_m \sin m(\omega t - kz) + b_m \cos m(\omega t - kz)], \quad (5)$$

where the summation is carried out over m from 0 to the end of the upper part of the time interval. Equation (5) can also be written in a more compact form:

$$n = \Sigma c_m \exp(iu_m), \quad (6)$$

where

$$c_m = (a_m^2 + b_m^2)^{1/2}, \quad u_m = [(m\omega t - kz) - \psi_m],$$

$$\tan \psi_m = b_m/a_m.$$

Substitution of (6) in (4) gives $i\omega m + Dk^2 = 0$, whence $k = (1 - i)(\omega m/2D)^{1/2}$.

Substituting (6) into (3), we find

$$n_0 V = \Sigma (D\omega m)^{1/2} c_m \exp i[(m\omega t - k_0 z) - \psi_m] \exp(-k_0 z), \quad (7)$$

where $k_0 = (\omega m/2D)^{1/2}$.

The above solution describes the propagation of flow rate pulsations for subsoil air in a porous medium.

The quantity $k_0^{-1} = z_0$ is the distance over which the rate of pulsation decays by a factor of e and ω/k_0 is the phase

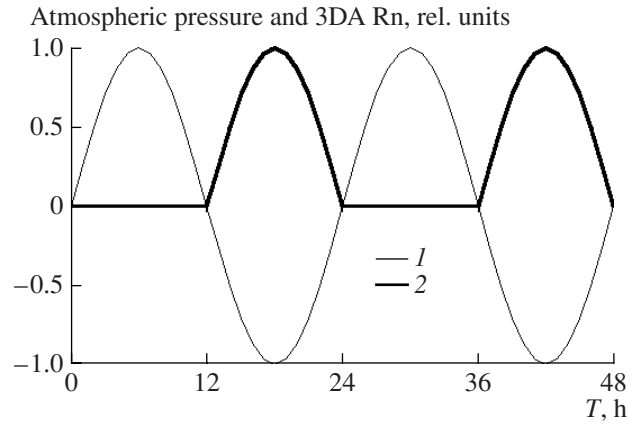


Fig. 2. Atmospheric pressure variation (1) and the response of geologic gas (2) at the ground surface as calculated from (10). The curves have been normalized by the maximum value.

velocity of pulsations in a porous medium along the z -axis:

$$V_{ph} = (2D\omega m)^{1/2} = c(2\omega K/\nu)^{1/2}. \quad (8)$$

The phase velocity for daily fluctuations with $K = 10^{-10} \text{ cm}^2$, the phase velocity is $\sim 10^{-2} \text{ cm/s}$. The mass velocity of subsoil air is

$$U = V_{ph} \times \delta P / P_0, \quad (9)$$

where $\delta P / P_0$ is the relative change in air pressure.

Geologic gas interacts with atmospheric air at the interface between the porous medium and the atmosphere. When atmospheric pressure increases, the air enters the pores and compresses the geologic gas, while when the pressure decreases, the air and some gas go from the pores. It follows that the average effect per period of atmospheric pressure change is to "pump" the geologic gas out to the atmosphere.

When the atmospheric pressure varies according to the law $\sim \sin \omega t > 0$, the amount of geologic gas in the air remains constant during the first half-period, but during the second the gas flows out into the atmosphere. In formal terms the process at the ground surface ($z = 0$) will be described by the equation

$$\begin{aligned} n_R U &= 0, \quad \text{if } \sin u_m^* > 0; \\ n_R U &= n_R V_{ph} (\delta P / P_0) \sin u_m^*, \quad \text{if } \sin u_m^* < 0, \end{aligned} \quad (10)$$

where $u_m^* = m\omega t - \psi_m$.

The time behavior of the amount of gas given away into the atmosphere $|\delta P / P_0|$ in relation to the phases of atmospheric pressure variations is shown in Fig. 2 for the simplest case in which equation (10) holds.

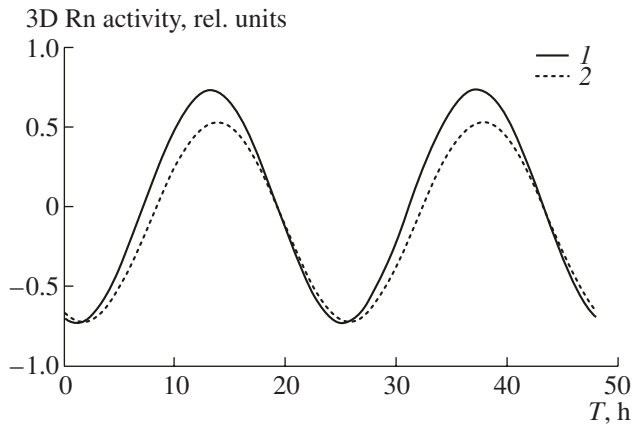


Fig. 3. Relative amplitude of 3D radon activity as recorded by two sensors installed at depths of one meter (1) and 1.5 meters (2), calculated on the model developed in this study. The phase and amplitude difference corresponds to $K \approx 10^{-10} \text{ cm}^2$. The curves have been centered and normalized by the maximum value.

Let $\chi = n_R/n_0$ be the average relative amount of geologic gas in subsoil air; the two equations (10) can then be replaced with the single formula

$$n_R V = \chi n_0 (\delta p/p_0) [\sin(\omega t - kz) + \sin(\omega t - kz)]/2. \quad (11)$$

If now we consider a single component in the geologic gas (radon), and assume the isolines of equal Rn concentration in the soil to be quasicontant, then atmospheric pressure variation is effectively a pump that continually forces Rn into the atmosphere from the zone of aeration h thick. The depth of that zone will be $\sim U \times \tau$, where U is the velocity of subsoil air and τ the radon disintegration constant. It is of interest to compare the latter with z_0 :

$$h/z_0 = (U/V)_{\omega\tau} = (\delta p/p_0)\omega\tau.$$

For daily atmospheric pressure variations of amplitude 10 millibars the thickness of the active layer is about $h \sim 0.25z_0$.

The amplitude of radon density variation at depth z is $\exp(-k_0 z)$ times smaller than that at the interface with the atmosphere, and its phase is different from the phase of the atmospheric pressure variation. We note that radon density variations are not due to atmospheric air mixing with radon, but to the lagging perturbation of geologic gas pressure caused by the pressure contrast across the surface of the porous medium. It is evident that, all other things being equal, the phase lag depends on the sensor depth. The phase shift between two sensors installed at different depths can be used to estimate the Darcy coefficient for a soil of interest. Figure 3 shows a modeled response of Rn concentration in loose deposits, assuming the Darcy coefficient to be equal to $\sim 10^{-14} \text{ m}^2$ at depths of 1 and 1.5 meters, to atmospheric pressure variations of period $T = 1$ day.

It is clearly seen in Fig. 3 that the signal at the upper sensor has a slightly larger amplitude, and is shifted about one hour in time. We are going to show that the shift is controlled by the value of K . The reasoning is as follows. Assuming curves 1 and 2 to have the same phase:

$$\omega t_1 - k_0 z_1 = \omega t_2 - k_0 z_2,$$

we get $k_0 = \omega(t_2 - t_1)/(z_2 - z_1)$. Since $k_0 = (\omega/2D)^{1/2} = (\omega\nu/KC^2)^{1/2}$, we have

$$K = (\nu/\omega c^2) [(z_2 - z_1)/(t_2 - t_1)]^2. \quad (12)$$

Here, z_1 and z_2 are the sensor depths and $t_2 - t_1 = \Delta t$ the phase shift in the daily variation of radon concentration.

Obviously, measurement of Δt allows monitoring of changes in the soil permeability coefficient. The use of a kinematic parameter in the dynamics of subsoil radon for monitoring the permeability of loose deposits will simplify the data processing, because no calibration by absolute values will then be required.

EXPERIMENTAL DATA AND DISCUSSION OF RESULTS

Consider our experimental data in the light of the model developed above. Figure 4 shows variations in atmospheric pressure and Rn concentration in the zone of aeration at a depth of one meter and at the surface, point 1 of Paratunka site (PRT) for the period September 3 to October 13, 2004. One can discern a correlation between Rn concentration and atmospheric pressure, one cycle of the latter lasting 3–4 days. It should be noted that the highest transfer function was observed for this site, of the entire radon monitoring network; the function is $H = 400 \text{ Bq}/(\text{m}^3 \text{ hPa})$ in the frequency range 0.7–1.2 cycles per day [10].

Figure 4b shows the correlation coefficient between Rn concentration and atmospheric pressure for various curve shifts. The outflow of radon into the atmosphere increases after more than 0.5 days since the drop in atmospheric pressure began. The phase shift of the correlation coefficient between the zone of aeration and the ground surface is about one hour. Examination of Fig. 4c, where the curves of Rn concentration have been normalized by the maximum and made to coincide, shows that the time behavior of Rn concentration at the ground surface is similar to that of the inflow of geologic gas into the atmosphere as calculated theoretically in relation to atmospheric pressure variation (Fig. 2).

Figure 5a shows the dynamics of Rn concentration in the zone of aeration (1 m), at the ground, and in the bunker air, as well as the concentration of CO_2 in the well shaft at INS during the period July 17 to August 30, 2006. It can be seen in Fig. 5b that the bunker observa-

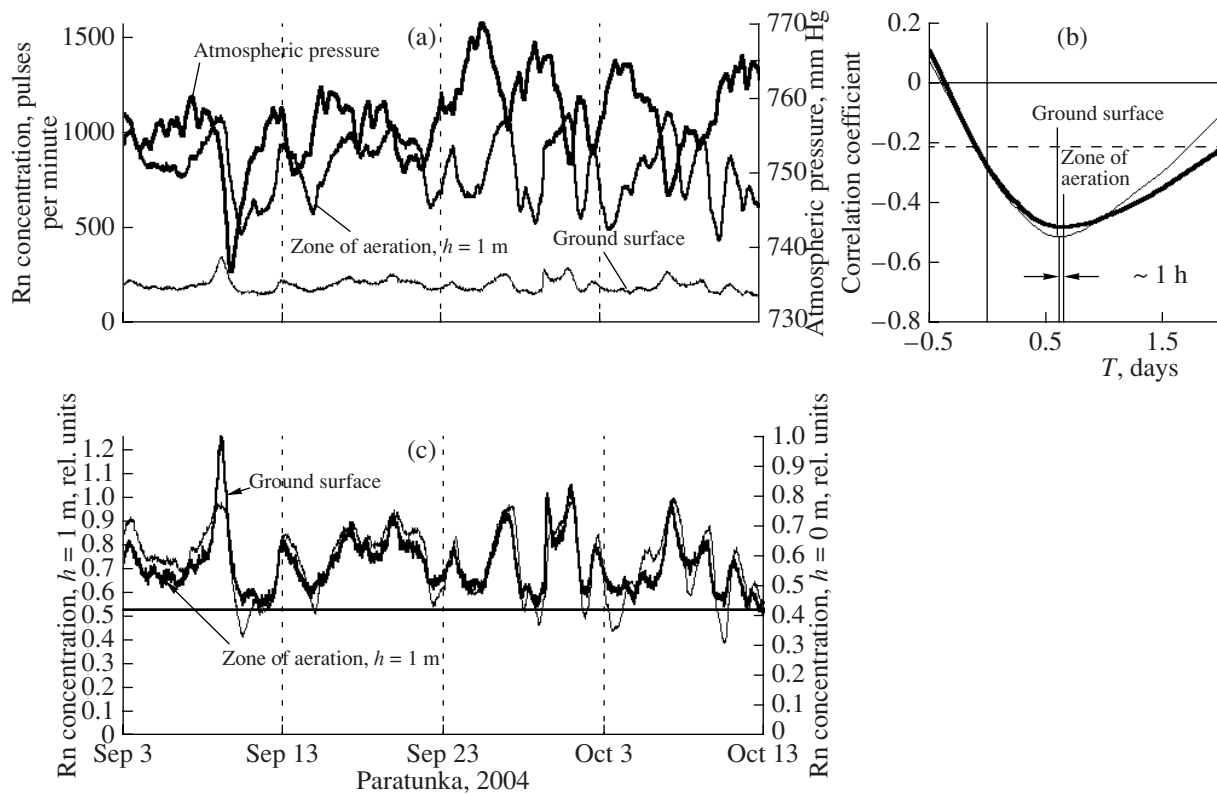


Fig. 4. Effects of atmospheric pressure variations on the time behavior of subsoil radon concentration: (a) curves of atmospheric pressure and Rn concentration in the zone of aeration ($H = 1$ m) and at the ground, site PR, (b) correlation coefficient between Rn concentration and atmospheric pressure for various shifts of curves, (c) curves of radon concentration in the zone of aeration and at the ground surface normalized by the maximum value.

tions show well-pronounced daily variations in atmospheric pressure with an amplitude of about 10 hPa, the maximum in the daily variations that affect components of the geologic gas occurring around the greatest rate of change for atmospheric pressure (Fig. 5c). These daily variations are due to some features in the structure of the observing station. The metal container above the bunker is heated in daytime and cools during nighttime, the temperature range being as great as 15°C, thus daily pressure variations occur due to convection currents. Daily pressure variations at the point can be regarded as a sounding signal, while the evolution of the response of Rn concentration to these variations can provide information on the state of the geologic medium at the point.

Figure 6 shows the correlation coefficient between the concentrations of CO₂ and Rn at the various recording points at INS and atmospheric pressure as a function of time shift between the two series. A shift of 100 min was observed between the zone of aeration and the ground surface. Assuming $\nu = 1.4 \text{ cm}^2 \text{ s}^{-1}$ as the dynamic viscosity for air [9], and $C = 330 \text{ m/s}$, $\omega = 7.27 \times 10^{-5} \text{ s}^{-1}$, we use (12) to get $K = 1.7 \times 10^{-4} \text{ m}^2$ for

$t_2 - t_1 = 100 \text{ min}$. This value of the Darcy coefficient is suitable for sandstone or settled loam [8].

It is of interest to examine relations between the kinematic and dynamic parameters of the Rn concentration in the zone of aeration and at the ground surface during the time period of study, considering that two earthquakes of magnitude 5.9 and 5.2 occurred off Kamchatka on July 17 and 20, respectively, following a two-month seismic quiescence as reported by the Kamchatka Branch, Geophysical Service, Russian Academy of Sciences. The events took place in the north (55.54° N) and in the south (49.6° N) of the Kamchatka Peninsula about 350 km from the observing station. The largest earthquake ($M_L = 6.2$) occurred 190 km from PRT in southern Kamchatka (51.19° N) on August 24, and a sequence of earthquakes was recorded September 1–2 in the southern Kronotskii Bay and in the Avacha Bay. The epicenter of the largest shock (magnitude 5.6) was 110 km from the observing station. The seismicity increase along the entire northern flank of the Kuril–Kamchatka island arc suggests an increased rate of motion for the Pacific plate, which may have caused a rearrangement of the regional stress field. Consider the response of the Darcy coefficient to the seismicity in

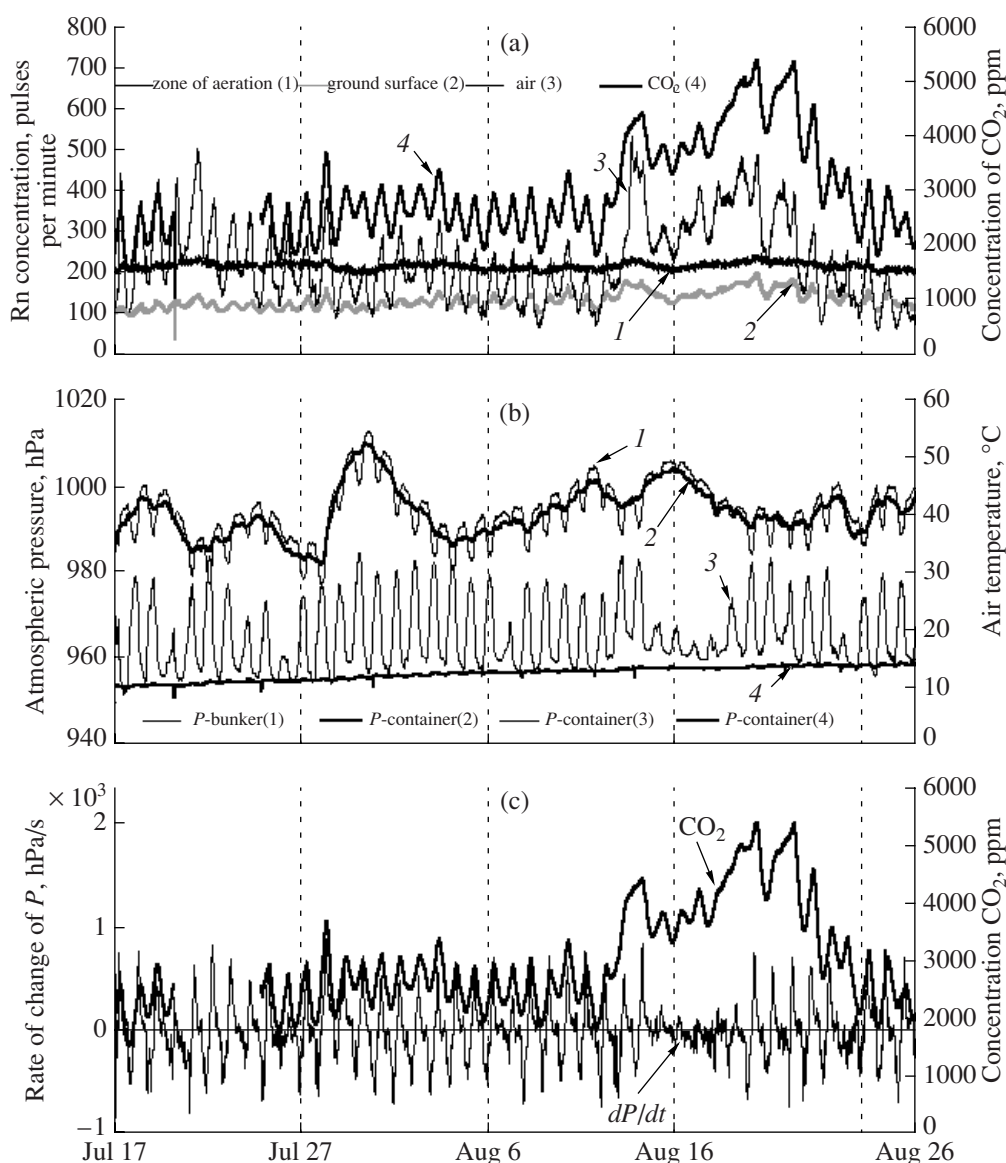


Fig. 5. Relationships between the time behavior of subsoil gas and several meteorological quantities: (a) concentrations of Rn and CO₂ at INS, (b) meteorological quantities (pressure P and temperature T) in the bunker and in the container, (c) concentration of CO₂ in the well and the rate of change of atmospheric pressure dP/dt .

the northern flank of the Kuril–Kamchatka island arc (Fig. 7).

We investigated correlative relationships for Rn concentration as measured in the zone of aeration and at the ground surface by using a bandpass filter bounded by the end frequencies $f_{1\text{end}} = 0.03$ and $f_{2\text{end}} = 0.05$ cycles per hour, resulting in identification of a daily harmonic (Fig. 7, top curves).

The maximum of the correlation coefficient was sought in five-day intervals and was ascribed to the endpoint of the interval. It can be seen from Fig. 7 that the shift of the highest correlation coefficient is within 80–180 min. The Darcy coefficient was found from (12) to lie in the range 6×10^{-14} to 2.7×10^{-13} m².

Indeed, the Darcy coefficient decreased by a factor of nearly four during the seismicity increase (August 14–19). Beginning from August 13, one notes that the concentrations of Rn at all channels and of CO₂ (Fig. 5a) have increased; as can be seen from Fig. 5b, this is unrelated to the meteorological parameters. Figure 7 also shows the ratio of Rn concentration in the zone of aeration and at the ground surface (N_a/N_{surf}) which clearly involves a daily component whose maximum occurs at the minimum in the rate of change of atmospheric pressure.

This observation can be treated as follows: the outflow of Rn from the ground surface exhibits the greatest sensitivity to atmospheric pressure variations compared

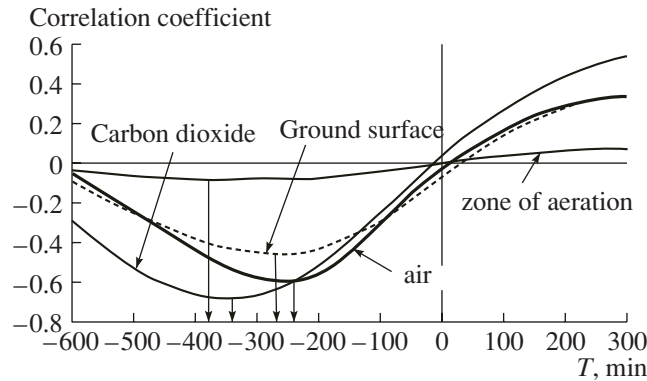


Fig. 6. Correlation coefficient between Rn concentration at various INS observation points and atmospheric pressure for several different shifts between these data series for the period July 17 to August 30, 2006. Arrows mark maxima in the correlation coefficient.

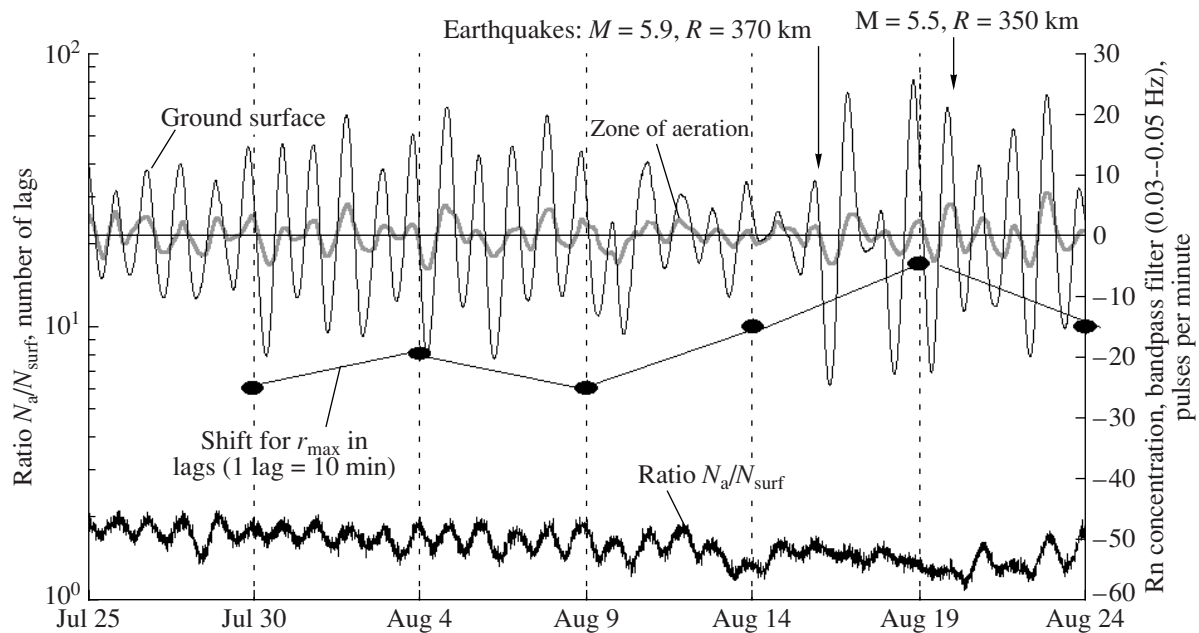


Fig. 7. Filtered curves of radon concentration (bandpass filter for 0.03–0.05 cycles per hour) at INS (the zone of aeration and the ground surface), the shift for maximum correlation between these curves in five-day intervals and the ratio of Rn concentration between the zone of aeration and the ground surface (N_a/N_{surf}).

with the zone of aeration, which leads to a relatively greater decrease in the surface concentration of Rn compared with the zone of aeration at the minimum rate of change of atmospheric pressure. The increased flow of subsoil Rn during August 14–19 was accompanied by a decrease in N_a/N_{surf} owing to a shift in the equilibrium state of Rn with the decay products at the surface.

Overall, the present analysis of dynamic and kinematic parameters for radon concentration at various depths in loose deposits provides a means for monitoring of changes in the state of stress and strain in the geologic medium.

CONCLUSION

The above analysis of the behavior of geologic gas due to the influence of external pressure variations is fairly well described within the framework of a porous earth model. Daily variations in the outflow of radon into the atmosphere due to atmospheric pressure variations may serve as a natural sounding signal in the monitoring of changes in the state of stress and strain in the geologic medium. The period of seismicity increase in the northern flank of the Kuril–Kamchatka island arc during August 17–30, 2006, was preceded and accompanied by increased concentrations of Rn and CO_2 at all

INS observation points during the period August 14 to 25 with a simultaneous decrease in the permeability coefficient for the upper part of the loose deposits around the observation site by a factor of more than 4 (6×10^{-14} – 2.7×10^{-13} m²).

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