Diurnal and Seasonal Variations of ^{222}Rn Concentration Profile in Soil

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(Received on November 27, 2006)

(Accepted on February 2, 2007)

The ^{222}Rn concentrations in soil and ^{222}Rn flux were measured in the campus of Nagoya University. The measurements were carried out discontinuously between December 1999 and March 2002 at depths of 15, 30, 50, 80, 120, and 200 cm. The ^{222}Rn concentrations in soil were analyzed for seasonal-, day-to-day-, and diurnal variations. The average values of ^{222}Rn concentrations in soil were 1.6, 6.9, 13.6, 24.2, 27.7, and $34.3 \text{ kBq} \cdot \text{m}^{-3}$ for soil depths of 15, 30, 50, 80, 120, and 200 cm, respectively. The ^{222}Rn flux ranged from 2.5 to $14 \text{ mBq} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ with the average $7.2 \text{ mBq} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$. The variations of ^{222}Rn concentrations in soil were larger in shallow depths. It seems that the ^{222}Rn concentration in soil during night-time was slight lower than day-time especially at depth 15 cm. The precipitation may have affected the ^{222}Rn concentration in soil differently depending on its amounts. The large decrease of air pressure during one day increased the ^{222}Rn concentration in soil at the depth 15, 30, and 50 cm. The effect of soil water content on ^{222}Rn concentration in soil was also observed.

KEY WORDS: ^{222}Rn profile, ^{222}Rn concentrations in soil, ^{222}Rn flux, seasonal variation, diurnal, precipitation, pressure decrease.

I INTRODUCTION

Natural material consists of a variety of elements, from the lightest element hydrogen to the heaviest element uranium. Some of these elements are radioactive ones belonging to the four natural occurring decay series. Radon-222 (^{222}Rn) with a half-life of 3.82 d is one of the radioactive elements in the uranium-238 series. It is an inert gas and hence has little affinity to get chemically bound. After produced by alpha decay of radium-226 (^{226}Ra), the ^{222}Rn atom possesses a kinetic energy of 86 keV and stop after a short distance of recoil depending on the density and composition of the material, e.g. in com-

mon minerals $0.02 - 0.07 \mu\text{m}$, and in water and air $0.1 \mu\text{m}$ and $63 \mu\text{m}$, respectively.¹⁾

Radon-222 and its short-lived decay products in the atmosphere are the most important contributors to human radiation exposure from natural radiation sources. The inhalation of these short-lived decay products through the bronchial tree provides the main pathway for radiation exposure to the lung causing the lung cancer risk. Indoor ^{222}Rn concentration in various countries yields average concentrations ranging from 10 to $100 \text{ Bq} \cdot \text{m}^{-3}$, with the values of 30 and $40 \text{ Bq} \cdot \text{m}^{-3}$ for geometric and arithmetic means of the worldwide distribution, respectively.^{2,3)}

The significant correlation between the ^{222}Rn concentration in the soil beneath buildings and indoor ^{222}Rn concentration has been found by Albering et al.⁴⁾ and Reimer.⁵⁾ Varley and Flowers⁶⁾ found this significant correlation only in the region with high ^{222}Rn concentration in soil. Radon-222 concentration in the soil can be used to predict whether indoor ^{222}Rn concentration might exceed some action level.

For most houses, the source of indoor ^{222}Rn is from the soil air entering the house through cracks and unsealed openings in the building which have direct contact to the ground. This transport is largely controlled by advection driven by the pressure difference between the building and the ground around the foundation. This pressure difference was produced by the higher temperature in building, mechanical ventilation, and wind blowing around the building.²⁾

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Within the soil, two different mechanisms of ^{222}Rn transport should be distinguished: (1) diffusion, in which the ^{222}Rn moves with respect to the concentration gradients in the soil, and (2) advection, in which the fluid itself moves through the porous medium and carries the ^{222}Rn along with it. Either or both mechanisms may be important in a given place.⁷⁾ Therefore, the ^{222}Rn flux from the soil surface will depend not only on the ^{226}Ra concentration, but also on several meteorological conditions and soil parameters, such as precipitation,⁸⁾ wind,⁹⁾ air pressure,^{10,11)} soil temperature,^{12,13)} soil moisture,^{14,15)} soil porosity and soil permeability.^{1, 16, 17)}

On a large scale, the high variability of ^{222}Rn concentration in soil is due to the variable concentration of ^{226}Ra in the soil. However, even in small scale, there is considerable spatial and temporal variability of ^{222}Rn in soil. Quantitative information on this small scale variability is required to estimate realistically the mean annual ^{222}Rn concentration in soil from a limited number of measurements.^{12, 18)}

The objective of this study is to enhance the understanding of the factors influencing the diurnal and seasonal variations of ^{222}Rn concentration in soil. The results could be used to improve the prediction of the sampling time and method of the measurement of ^{222}Rn concentration in soil that will be used to estimate the mean annual ^{222}Rn concentration.

II BASIC BEHAVIOUR OF RADON IN SOIL

There are two main groups of factors that determine the concentration of ^{222}Rn in soil:

- (1) the parameters that determine the source potential of the soil, and
- (2) radon transport that determine the movement of radon in the soil.

One of the equations to calculate the ^{222}Rn concentration in soil¹⁹⁾ is given by:

$$(a+L\theta) \frac{\partial C_a}{\partial t} = \frac{\partial}{\partial t} \left[D_b \frac{\partial C_a}{\partial z} \right] - \frac{K_p}{\nu} \left(\frac{\partial P}{\partial z} \right) \left(\frac{\partial C_a}{\partial z} \right) - L\theta K(\Psi_m) \left(\frac{\partial \Psi_m}{\partial z} \right) \left(\frac{\partial C_a}{\partial z} \right) - (a+L\theta) \lambda C_a + R \lambda \rho E, \quad (2.1)$$

where a is the volumetric air-filled pore space in soil, θ is the volumetric water-filled pore space in soil, L is the ^{222}Rn equilibrium distribution coefficient (Henry's Law constant in dimensionless form), C_a is the ^{222}Rn concentration in air-filled pore space ($\text{Bq} \cdot \text{m}^{-3}$), D_b is the bulk ^{222}Rn diffusion coefficient in soil ($\text{m}^2 \cdot \text{s}^{-1}$), K_p is the bulk material air permeability (m^2), ν is the dynamic viscosity of air ($\text{Pa} \cdot \text{s}$), P is the pore air gas pressure (Pa), R is the ^{226}Ra concentration in solid material ($\text{Bq} \cdot \text{kg}^{-1}$ dry mass), λ is the ^{222}Rn decay constant (s^{-1}), ρ is the bulk dry density ($\text{kg} \cdot \text{m}^{-3}$), and E is the coefficient of ^{222}Rn emanation into the soil pore. The factor of $(a+L\theta)$ is physically corresponds to the effective porosity in which ^{222}Rn is distributed in the gas-phase and liquid-phase components. The first term in the right hand side in Eq.2.1 represents diffusion in the bulk soil, and second term accounts for advection by the soil air. The third term represents the advection of

^{222}Rn dissolved in the soil water and $K(\Psi_m) \left(\frac{\partial \Psi_m}{\partial z} \right)$ is the advective velocity of the pore water. The fourth and fifth terms describe the radioactive decay and sources of ^{222}Rn in soil.

The simplest deduction of the Eq.2.1 is a steady state situation where no transport takes place. With these assumptions, the equation can be simplified to yield:

$$(a+L\theta)C_a = R\rho E, \quad (2.2)$$

where only the last two terms in Eq.2.1 remain. From this equation, we can evaluate an equilibrium concentration C_{eq} for the situation where emanating radon decays locally.

$$C_{eq} = \frac{R\rho E}{a+L\theta}, \quad (2.3)$$

This equation implies that the equilibrium concentration is proportional with radium content and emanation coefficient, and inversely proportional with the pore space in soil for radon to reside.

III EXPERIMENTAL METHODS

The measurements were carried out over the period of more than two years since December 1999. In order to know the variations in a few weeks, the measurements of ^{222}Rn concentration in several depths were done once or twice per day for at least eight days long. The measurement was carried out every two months from December 2000 to December 2001. By taking the soil air samples every four hours at least for one day long, the diurnal variations of ^{222}Rn concentration in several depths were observed at 10 campaigns from December 1999 to March 2002. All of these measured data could be applied to analyze the seasonal variations of ^{222}Rn concentration in soil.

1. Measurement of ^{222}Rn concentration in soil

The experiments were carried out in the campus of Nagoya University ($35^\circ 09' \text{ N}$, $136^\circ 58' \text{ E}$). The sampling point is about 2 m higher and 3 m far from a car park and about 10 m far from the nearest building. **Figure 1** illustrates the schematic diagram of the equipment to take samples of ^{222}Rn gas in soil. Stainless steel probes of inside diameter of 8.9 mm were installed in a square area of $1 \text{ m} \times 1 \text{ m}$ at depths of 15, 30, 50, 80, 120 and 200 cm at the beginning of December, 1999. The top of each probe was plugged with plastic cap. Soil gas was collected by a portable diaphragm pump into sampling bag (Tedlar® Polyvinyl fluoride (PVF) or aluminum bags, GL Sciences Inc.) via the stainless steel probes. The flow rate of collecting samples was $1 \text{ L} \cdot \text{m}^{-1}$, and the sample time was 20s. Before sampling air into bag, the pump was turned on for a few seconds to evacuate the air in the tube. The soil gas collected in each sampling bag was then filled into an evacuated scintillation cell (Pylon Model 300 A, Pylon Electronic Inc). After 3.5 hours for equilibrium between ^{222}Rn and its decay products, the alpha counts of the scintillation cell were measured with a portable radiation monitor (Pylon Model AB-5, Pylon Electronics Inc.).

Fig. 1 The schematic diagram of the equipment to measure the ^{222}Rn concentration in soil.

2. Measurement of ^{222}Rn flux and soil parameters

In addition to the measurements of ^{222}Rn concentration in soil, some parameters correlating to ^{222}Rn concentration in soil were also measured. Radon-222 flux was measured by the accumulating method with a chamber ($L50\text{ cm} \times W25\text{ cm} \times H20\text{ cm}$). Soil moisture was measured at the depth of 15 cm by a TRIME FM soil moisture meter (IMKO Micromodultechnik GmbH). The meteorological conditions such as wind speed, air temperature, humidity, rain fall, etc. were observed by the Gro Weather System (Davis Instruments Corp).

The soil parameters of bulk density, particle density, porosity and water content were measured by using Nakano's procedure.²⁰⁾ The organic contents in soil were measured from the weight reduction by heating at 450°C . The ^{226}Ra concentration was measured by gamma spectrometry with a Gamma-X HP Ge Detector (EG&G Ortec). The soil permeability was measured using a portable permeability probe by the same procedure of Damkjær.²¹⁾ The ^{222}Rn diffusion coefficients in soil were measured using a soil ^{222}Rn diffusion monitor, in which a soil sample was placed between two detectors.²²⁾

IV RESULTS AND DISCUSSIONS

1. Soil Parameters

Table 1 shows the soil parameters at Nagoya University measured using the methods as described in section III.2. The particle density was uniform from 0 cm to 65 cm in depth. The bulk density, porosity, and organic content show also uniform

distributions until 65 cm except 0 to 5 cm depth. The surface soil had relatively large porosity and organic content. The ^{226}Ra concentration was relatively uniform in the range of 15.6 to $18.7\text{ Bq}\cdot\text{kg}^{-1}$ in the depths until 65 cm.

2. Profiles of ^{222}Rn in the soil

The profiles of monthly average of ^{222}Rn concentrations in soil as the function of soil depths from December 1999 to March 2002 were shown in **Fig. 2** and **Table 2**. The monthly average of the ^{222}Rn concentration in soil was the average value of the concentrations measured in each month, and the values were not taken from the same year. **Table 2** shows the number of samples and the number of days in each month for calculation of the average values of ^{222}Rn in soil, for example in December the experiments were done for 19 days with 46 samples.

The average of ^{222}Rn concentrations in soil for all measurements were 1.6 , 6.9 , 13.6 , 24.2 , 27.7 , and $34.3\text{ kBq}\cdot\text{m}^{-3}$ at soil depth of 15, 30, 50, 80, 120, and 200 cm, respectively. The ^{222}Rn flux ranges from 2.5 to $14\text{ mBq}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ with the average $7.2\text{ mBq}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$.

The calculation of the concentration of ^{222}Rn in soil at the equilibrium state using Eq.2.3 with the soil parameters in **Table 1** and emanation power of 0.22 yields $42\text{ kBq}\cdot\text{m}^{-3}$. The value is in a good agreement with the measurement results which asymptotically approach to the value with depth.

Figure 2 also shows the results of the ^{222}Rn concentration profile in soil calculated using the Eq.2.1. The term for advection by the soil air were omitted in this calculation. The soil parameters used in the calculation has been shown in **Table 1** with soil water content of 0.11, emanation power of 0.22, and no rainfall. It should be noted that the parameters used in the simulation were for the situation with a relatively low soil moisture content because the diffusion coefficient used are for the near surface layer where porosity was high and water content was relatively low. This calculation result can be regarded as a reference profile. The measurement results lay on both sides, mostly on the high concentration side. From this profile, there seems that ^{222}Rn concentrations in soil have variations among the measurements. In the following section, the seasonal variations, day-to-day variations, and diurnal variations will be analyzed to find the factors that affect the ^{222}Rn concentration in soil and flux of ^{222}Rn .

3. Seasonal variations

From the **Table 2** the ^{222}Rn concentrations in soil have an annual cyclic for the soil depths of 80 to 200 cm. From January to June the concentration increased, then decreased until September. October had high concentration then decrease again until January. The highest concentrations were about $45\text{ kBq}\cdot\text{m}^{-3}$ at 200 cm on June and October and the lowest were about $25\text{ kBq}\cdot\text{m}^{-3}$ at 200 cm on August and September. This characteristic is correlated with the precipitation pattern. On June and October, the precipitations were high, while in July to September were low. Beside that, when it rains in summer from July to September, the water easily evaporates due to the high temperature and solar incidence. This results less water in the deeper soil.

Figure 2 also shows that the concentrations at 80 cm were

Table 1 Soil parameters in Nagoya.

Depth (cm)	Bulk density ($\text{g}\cdot\text{cm}^{-3}$)	Particle density ($\text{g}\cdot\text{cm}^{-3}$)	Porosity content (Vol.%)	Organic content (wt.%)	^{226}Ra coefficient ($\text{Bq}\cdot\text{kg}^{-1}$)	Permeability (m^2)	Diffusion ($\text{m}^2\cdot\text{s}^{-1}$)
0-5	1.58	2.78	43	8.37	15.6 ± 0.9	-	5.3×10^{-6}
15-20	1.83	2.72	33	3.87	18.7 ± 0.9	-	-
30-35	1.93	2.77	30	3.71	17.3 ± 0.9	-	-
50-55	1.86	2.75	33	3.75	17.7 ± 0.9	4.6×10^{-12}	-
60-65	1.76	2.73	36	2.75	17.9 ± 0.9	-	-
90						6.4×10^{-12}	-

Table 2 Number of profiles, monthly average of the ^{222}Rn concentration in soil ($\text{kBq}\cdot\text{m}^{-3}$) and the ^{222}Rn flux ($\text{mBq}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$). Number of days the measurement made is shown in parenthesis.

	No. of profiles (No. of days)	15 cm	30 cm	50 cm	80 cm	120 cm	200 cm	flux
Jan	6 (1 d)	1.9	6.3	11.1	16.8	27.6	30.7	
Feb	22 (10 d)	1.8	2.6	17.6	17.2	31.6		11.0
Mar	6 (1 d)	0.1	3.2	13.4	29.3	27.0	32.8	
Apr	18 (10 d)	0.3	3.2	7.9	25.2			4.9
May	12 (2 d)	0.4	12.1	16.5	29.8	32.1	34.2	
Jun	17 (9 d)	2.3	12.0	11.9	33.3	35.6	46.0	2.5
Jul	12 (2 d)	0.7	5.7	9.0	25.4	25.2	29.1	
Aug	15 (9 d)	2.6	4.9	11.4	19.8	17.8	29.1	4.6
Sep	6 (1 d)	1.2	7.0	11.1	14.8	15.4	24.1	
Oct	16 (10 d)	2.8	11.1	21.2	25.8	33.1	46.1	6.4
Dec	46 (19 d)	3.8	7.8	18.3	29.0	31.7	36.5	14.0
Average	176 (74 d)	1.6	6.9	13.6	24.2	27.7	34.3	7.2

sometimes higher than that at 120 cm. This characteristic may be due to discontinuous or layered structure of soil properties that affect the radon concentration such as soil porosity, water content, and ^{226}Ra concentration.

Figure 3 shows the average of ^{222}Rn flux together with the variations of soil temperature, the precipitation, and water content in soil from February to December 2001. The value of ^{222}Rn flux in each month was the average values of all measurements conducted at that month. Numbers of observations at April, June, August, and October were the same as shown in **Table 2**, while at February and December were 16 observations (nine days) and 16 observations (eight days), respectively. The maximum value of ^{222}Rn flux was observed in December and the minimum value in June. These values were opposite with the ^{222}Rn concentration in soil. This may be explained that when the water blocked the ^{222}Rn in soil, especially in soil surface, the concentration of ^{222}Rn in soil would increase, but the ^{222}Rn flux would decrease.

Fig. 2 The monthly average of ^{222}Rn concentration profiles in soil.

Fig. 3 The flux of ^{222}Rn together with the amount of rainfall, water content, and soil temperature in 2001.

Fig. 4 The variations of ^{222}Rn concentration in soil depths in two weeks from 14 JST on 4 October 2001.

4. Day-to-day variations.

In this observation, the measurements were carried out every two months from December 2000 to December 2001. The ^{222}Rn concentrations in soil were measured once or twice a day at least eight days per campaign. **Figure 4** shows the result measured on 4–15 October 2001. The figure shows an interesting tendency after two times of rainfall. After the first rainfall, the ^{222}Rn concentrations at 15, 30 and 50 cm depths increased 9, 3, and 1.3 times, respectively, i.e. from 1, 6, and $20\text{ kBq}\cdot\text{m}^{-3}$ to 9, 19, and $26\text{ kBq}\cdot\text{m}^{-3}$, then decrease in the following days. The air pressure on 4 October 2001 at 14 JST was 101.0 and on 5 October 2001 at 11 JST was 100.9 kPa. The second rainfall in the figure affected the concentration differently from the first one. Although quantification of the change in the concentration due to the rain could not be done since no measurement has been carried out just before the precipitation, it is clear that the increase was not as large as that of the first rain. The precipitation of the first and second rain was 18.4 mm and 45.4 mm, respectively. The rainwater may have affected the ^{222}Rn concentration differently depending on its amounts. With large amount of precipitation, there is a possibility that the water could play a role to move easily ^{222}Rn in soil by advection processes; therefore the concentration was not as large as the first case with smaller amount of precipitation.

The influence of the precipitation and air pressure to ^{222}Rn

Fig. 5 The variations of ^{222}Rn concentrations in soil and ^{222}Rn flux due to the influence of precipitation and air pressure in December 2001.

concentration in soil was seen in the observation on December 2001. The air pressure suddenly dropped from 102.2 kPa at 22 JST on 12 December to 100.1 kPa at 14 JST on 13 December and gradually increased as shown in **Fig. 5**. The amount of precipitation at that time was 18 mm. The ^{222}Rn concentrations in soil at 15, 30, and 50 cm increased 15, 5, and 2 times, respectively, i.e., from 1, 4, and 16 $\text{kBq}\cdot\text{m}^{-3}$ at 22 JST on 12 December to 7, 15, 18 $\text{kBq}\cdot\text{m}^{-3}$ on 13 December, and still increased to 15, 20 and 27 $\text{kBq}\cdot\text{m}^{-3}$ on 14 December. After that the concentration at 30 and 50 cm suddenly dropped. The increase of ^{222}Rn concentration in soil for this period was larger than in the period of October 2001 as shown in **Fig. 4**, even the amount of rainfall was the same. This was probably due to the effect of air pressure. The effect of only air pressure

Fig. 6 The diurnal variations of ^{222}Rn concentrations at depths of 15, 30, 50, 80, 120, and 200 cm from 23 to 24 December 2001.

itself was difficult to identify since the depression is mostly followed by the rain. As described by KRANER et al.,⁸⁾ the change of air pressure will effect a short range displacement of soil gas since there is a pressure differential between the atmosphere and soil gas at depth. As the barometer rises, ^{222}Rn free air flows into the surface soil layers, displacing and forcing the soil gas initially in these surface layers to deeper soil. The opposite effect would occur during a falling barometer.

The variations values of ^{222}Rn concentration in soil for this day-to-day observations show that the largest variation was in the depths of 15 and 30 cm, and the smallest was in the depths 120 and 200 cm.

5. Diurnal variations

The diurnal variations of ^{222}Rn concentrations in soil were measured for more than 10 campaigns. In this observation, the ^{222}Rn concentrations were measured every four hours at least for a day at the depths of 15, 30, 50, 80, 120, and 200 cm. The soil temperatures and other meteorological data also were measured. **Figure 6** shows the one of the results of ^{222}Rn concentrations in soil conducted on 23–24 December 2001. The ^{222}Rn concentration in soil was about 5 $\text{kBq}\cdot\text{m}^{-3}$ at the depth of 15 cm and about 35 $\text{kBq}\cdot\text{m}^{-3}$ at the depth of 200 cm, and increase with the increasing the depths. Air temperatures ranged between 3 and 10°C, the relative humidity in air was between 40 and 80%, and the air pressure was from 101.45 to 101.9 kPa. It seems that the ^{222}Rn concentration in soil during night-time was slight lower than day-time especially at depth 15 cm.

The diurnal variation of ^{222}Rn concentration in soil was the largest in the depth of 15 cm, and decrease with the depth. This shows that in shallow depths the ^{222}Rn concentration was largely influenced by meteorological conditions, as wind, air pressure and air temperature. But, when each these parameters were correlated to the ^{222}Rn concentration, the results failed. This is because of the composite influence of meteorological parameters could mask the effect of any particular parameter.

V CONCLUSIONS

The transport mechanisms of ^{222}Rn in soil have been studied in the campus of Nagoya University. The profile of ^{222}Rn concentrations in soil and the ^{222}Rn flux together with meteorological conditions and soil parameters have been measured between December 1999 and March 2002 at the depths of 15, 30, 50, 80, 120, and 200 cm. The results showed that ^{222}Rn concentration in soil increased with depth. The calculated ^{222}Rn profile using the soil parameters showed similar profile with the measured profile. The results were found in this study as follows:

- (1) The average values of ^{222}Rn concentrations in soil for all observations were 1.6, 6.9, 13.6, 24.2, 27.7, and 34.3 $\text{kBq}\cdot\text{m}^{-3}$ for soil depths of 15, 30, 50, 80, 120, and 200 cm, respectively. The ^{222}Rn flux ranged from 2.5 to 14 $\text{mBq}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ with the average 7.2 $\text{mBq}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$.
- (2) The ^{222}Rn concentrations in soil were analyzed for seasonal-, day-to-day-, and diurnal variations. The variations of ^{222}Rn concentrations in soil depths were larger in shallow

depths.

- (3) The ^{222}Rn concentrations in soil had an annual cyclic for the soil depths of 80 to 200 cm. The highest concentrations were in June and October and the lowest in August and September. This characteristic was a good agreement with the precipitation pattern. The precipitation was high in June and October and low in August and September. The precipitation had an influence also on the ^{222}Rn flux. The precipitation may have affected the ^{222}Rn concentration differently depending on its amounts.
- (4) The large decreasing of air pressure during one day increased the ^{222}Rn concentration in soil at the depth 15, 30, and 50 cm. The effects of soil water content were also observed.
- (5) The diurnal variation of ^{222}Rn concentration in soil was the largest in the depth of 15 and 30 cm, and decrease with the depth. It seems that the ^{222}Rn concentration in soil during night-time was slight lower than day-time especially at depth 15 cm.

There were still a lot of problems to know exactly the transport mechanisms of ^{222}Rn in soil. The ^{222}Rn concentration is affected by the water movement in soil. To know the actual effect of this phenomenon, it is important to make the experiment related to the water movement in soil. These studies will give comprehensive understanding about the ^{222}Rn transport from soil into houses.

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