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Key Points:
- We describe a new phenomenon in atmospheric physics—Rn-222 progeny circulation during thunderstorms
- The Rn progeny lifted to the atmosphere by a near-surface electric field are returned back to the ground by rain
- The enhancement of the natural gamma radiation during thunderstorms was measured with precise gamma ray spectrometers

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Circulation of Radon Progeny in the Terrestrial Atmosphere During Thunderstorms

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Abstract We describe a new phenomenon in atmospheric physics—^{222}Rn progeny circulation during thunderstorms. The enhancement of the natural gamma radiation during thunderstorms was measured with precise gamma ray spectrometers. Results of measurements performed at Aragats mountain in Armenia during summer 2020 demonstrate the Rn progeny lifted to the atmosphere by a near-surface electric field are returned backward to the ground by rain precipitation. Thus, thunderstorms not only return negative charge to the Earth by lightning flashes but also maintain Rn progeny circulation in the atmosphere; it this way, significantly enlarging natural gamma radiation above the Earth surface and Radon concentration in the atmosphere.

Plain Language Summary Radon gas and its daughter products account for more than half of the total human radiation dose from natural and man-made radionuclides in the environment. Radioactive radiation contributes to the processes that lead to lung cancer. According to the WHO, radon is the second leading cause of lung cancer after tobacco smoking. In the letter, we describe a new phenomenon in the terrestrial atmosphere- the circulation of Radon and its progenies during thunderstorms. Measurements performed at Aragats mountain in Armenia during summer 2020 demonstrate the identity of the Rn progeny lifted to the atmosphere by a near-surface electric field, and progenies returned to the ground with rain droplets. Thus, thunderstorms not only return negative charge to the Earth but also maintain Radon progeny circulation in the atmosphere during thunderstorms; it this way, significantly enlarging natural gamma radiation.

1. Introduction

The terrestrial atmosphere is host to various sources of electric currents and gamma radiation ranging from the fair-weather current of picoamperes to hundreds of kiloampere lightning currents. Particle fluxes range from the single particles of the ambient population of secondary cosmic rays to huge particle showers from interactions of primary high-energy proton or nuclei with terrestrial atmosphere and electron-gamma ray avalanches from the electron accelerator operated in the thundercloud. Gamma radiation from the primordial radionuclides have the half-life comparable with the age of the Earth, and they contribute significantly to natural gamma radiation (NGR) at considerably low energies (<3 MeV).

Radionuclides derived from the Earth crust can influence the electrical properties of the atmosphere and can influence human illness and death rates, result in DNA alterations, and chromosomal aberrations and weakening of immunity (Hunting et al., 2020).

The static electric field in the lower atmosphere is modulated by the mobile particles carrying electrical charges, i.e., different types of hydrometeors, aerosols, small ions, and progeny of radioactive isotopes. The charge separation initiated by the updraft of moisture generates an electric field between differently charged layers emerging in the thundercloud; potential drop (voltage) in the cloud can reach hundreds of megavolts. Emerging near-surface electric field lifts charged aerosols with attached ^{222}Rn isotope and its progeny to the atmosphere. Correspondingly, the concentration of ^{222}Rn at surface decreases 10 times (Roffman, 1972; Wilkcning et al., 1966); the small ions and aerosols with attached ^{222}Rn are lifted up in seconds to tens of meters due to their large mobility. These gamma emitters significantly enhance low-energy natural gamma radiation measured by spectrometers located several meters above the ground (Chilingarian, 2018; Chilingarian et al., 2019a, 2019b). The rain returns long-lived progeny to the Earth recovering and somewhat enhancing the surface radiation (washout process, Barbosa et al., 2017; Chilingarian et al., 2020; Fabro et al., 2016; McCarthy & Parks, 1992; Reuveni and Yair, 2017).

We present the measurement of the gamma radiation performed on Aragats mountain, 3,200 m above sea level during summer thunderstorms. We estimate the intensity of the different ^{222}Rn progenies in the rainwater; clarify the washout process and estimate the percentage of isotopes returned by the rain to the Earth surface. For measurements, we use the precise ORTEC firm gamma spectrometer (NaI(Tl), FWHM ~7.7% at 0.6 MeV, see details in Hossain et al., 2012) surrounded with lead filters. Simulations of the cosmic radiation, radon progeny radiation, and detector response function calculation were performed with the aid of the EXPACS code (Sato, 2018).

Figure 1. ORTEC firm gamma spectrometer (NaI(Tl), FWHM ~7.7% at 0.6 MeV, see details in Hossain et al., 2012), surrounded by 4-cm thick lead filters. The spectrometer is positioned in the experimental hall on Mt Aragats (3,200 m MSL) which is 3-m high and located under a metallic tilt roof of 0.6-mm thickness.

2. Methods

Gamma radiation measured on the Earth surface comes from the ground and from the atmosphere. The largest surface contribution is from gamma rays originating in the mineral grain, in their crystal lattices, and in the construction materials. The radiation is stable because the concentration of radionuclides in minerals and construction materials is constant due to long half-lives of their parent isotopes (^{40}K, ^{238}U, ^{232}Th, see details in Chilingarian et al., 2019a). Therefore, to investigate Radon progeny circulation (lifted by the near-surface electric field and returned through precipitation from rain) in the atmosphere we need to take into account and filter as much as possible this more-or-less stable contribution of the radionuclides from the surface. Gamma spectrometers are positioned on Aragats in the experimental hall which is 3-m high and located under a metallic tilt roof of 0.6-mm thickness. By surrounding the ORTEC spectrometer with the 4-cm thick lead filter (see Figure 1) we suppress the Radon progeny gamma radiation ≈12 times; the count rate of the spectrometer decreases from 12,600 ± 112 to 1,080 ± 34.

In Figure 2, we show the energy spectrum measured by ORTEC spectrometer for 6 h (normalized to 1-min count rate). Most pronounced are the positron annihilation peak (0.511 MeV), ^{40}K peak (1.46 MeV), and ^{208}Tl peak (2.61 MeV). Bismuth isotopes also are present in smaller quantities, see Table 1.

In the first column of Table 1, we present the count rate of registered particles normalized to a 1-min time span. Next is the absolute enhancement and its share of important isotopes from radon and thoron chains, potassium isotope peak, the 511 keV positron annihilation line, as well a small amount ^{137}Cs trace due to contamination of the ORTEC spectrometer during calibration with Cs isotope. In the last column, we show the overwhelming portion of the continuous spectrum induced by the secondary cosmic rays (CR) and Compton scattering of the gamma rays in the body of the NaI(Tl) crystal of ORTEC spectrometer.

To measure the share of Rn progeny in the rainwater, we should subtract the background spectrum from the spectrum measured during exposing rainwater to the spectrometer (see details of technique in Chilingarian et al., 2019b). As we see in the last column of Table 1, the lead filter suppresses background down to a few percent; >90% of the count rate comes from the stable flux of high-energy cosmic rays penetrating the lead filter. Simulations of the detector response function with cosmic radiation, radon progeny radiation, were performed with EXPACS code (Sato, 2018) and coincide rather well with the experimentally measured count rate.

Table 1
Composition of the Background Gamma Radiation Measured by ORTEC Spectrometer Surrounded From All Sides by the 4-cm Thick Lead Filter

Total intensity 0.3–3 MeV	^{214}Pb (354 keV)	511 keV	^{137}Cs (662 keV)	^{214}Bi (768 keV)	^{228}Ac (911 keV)	^{214}Bi (1.12 MeV)	^{214}Bi (1.76 MeV)	^{214}Bi (2.2 MeV)	^{40}K (1.46 MeV)	^{208}Tl (2.6 MeV)	CR and Compton scattered
912	8	31	2	4	4	3	3	3	22	4	828
100%	0.9	3.4	0.2	0.4	0.4	0.3	0.3	0.3	2.4	0.4	90.8

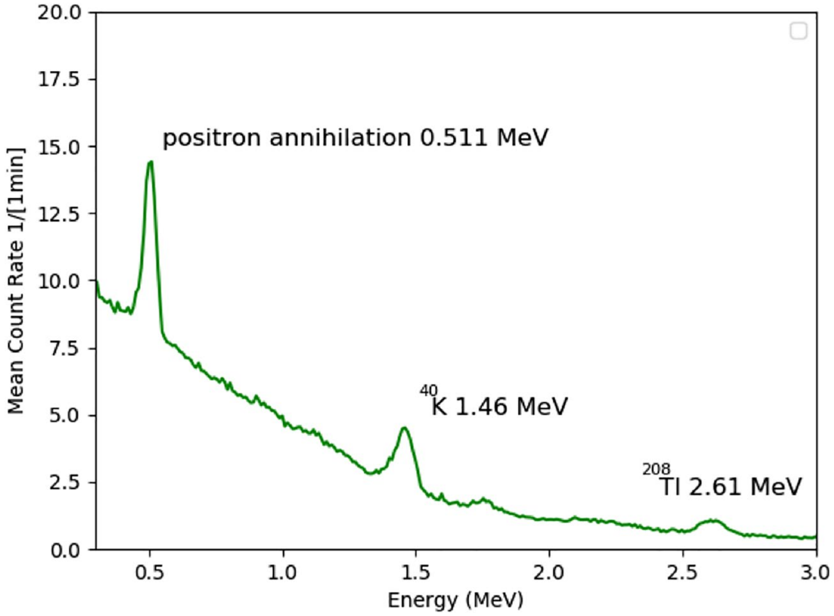

Figure 2. Energy spectra of the background measured by the ORTEC spectrometer surrounded by lead filter.

3. Results

In 2020, the first rain on Aragats was in June and rain showers were only during July, which is when they fille special container in a few minutes. In Figure 3, we show four episodes of the radiation measurements (after subtracting the background). In Table 2, we show the count rates of gamma emitters including radioactive isotopes, positron annihilation, and continuous spectrum of secondary cosmic rays (mostly muons) and gamma rays scattered in the body of the NaI crystal (continuum to the right of each spectral line).

As it was expected from previous measurements, the most pronounced peaks are ^{214}Pb and ^{214}Bi (Chilingarian et al., 2019b, 2019c, 2020), and as we show in Table 2 the share of different gamma-emitting isotopes in the atmosphere measured by the same spectrometer well coincides with the spectra measured from the rainwater (see Table 2 in Chilingarian et al., 2020). Sure, the electrified atmosphere introduces changes in

Table 2
Summary of the Gamma Radiation Measurements From the Rainwater by the ORTEC Spectrometer Covered by the 4-cm Thick Lead Filter From All Sides

	Intensity 0.3–3 MeV	^{214}Pb (354 keV)	511 keV	^{214}Bi (609 keV)	^{214}Bi (768 keV)	^{228}Ac (911 keV)	^{214}Bi (1.12 MeV)	^{214}Bi (1.76 MeV)	^{214}Bi (2.2 MeV)	CR + Compton scattered
23 July mean count rate [12:32–12:48] 1/min	585	147	5	109	45	26	40	32	7	174
%		25.1	0.9	18.6	7.7	4.4	6.8	5.5	1.2	29.7
23 July mean count rate [18:27–18:42] 1/min	531	123	6	102	32	43	38	23	8	156
%		23.2	1.1	19.2	6.0	8.1	7.2	4.3	1.5	29.4
24 July mean count rate [17:26–17:41] 1/min	814	191	8	161	46	41	60	42	12	253
%		23.4	1.0	19.7	5.7	5.0	7.4	5.2	1.5	31.1
1 August mean count rate [14:16–14:31] 1/min	343	91	9	63	19	13	28	13	8	99
%		26.5	2.6	18.4	5.5	3.8	8.2	3.8	2.3	28.9
Mean %		**24.6 ± 1.5**	**1.4 ± 0.8**	**19 ± 0.6**	**6.2 ± 1**	**5.3 ± 1.9**	**7.4 ± 0.6**	**4.7 ± 0.8**	**1.6 ± 0.5**	**29.8 ± 0.9**

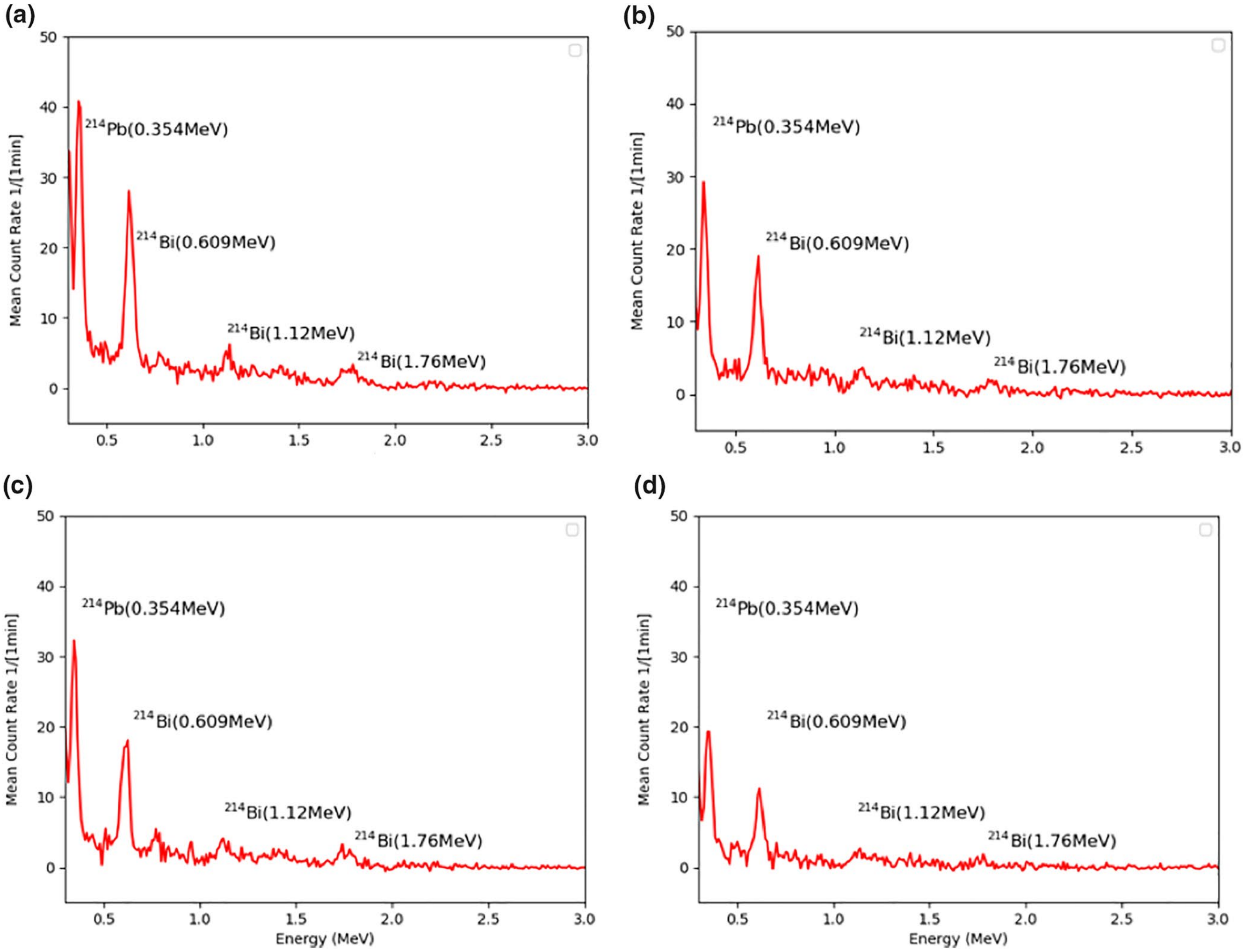

Figure 3. The energy spectra of the rainwater measured by the ORTEC spectrometer covered by 4-cm thick lead filter from all sides: (a) at 12:32 on 23 July; (b) at 18:27 on 23 July; (c) at 17:26 on 24 July; and (d) at 14:16 on 1 August.

the particle fluxes and slightly enlarges the positron annihilation share (see Figure 6 in Livesay et al., 2014); however, these changes are not large and do not exceed ≈1%. The potassium spectrum is very stable and does not influence the rainwater spectrum. In the last row of Table 2, we show the mean value of each gamma emitter share and error of the mean.

In Figure 4, we present the decay curve of the most abundant ^{214}Pb isotope. The intensity was measured every 30 min for a period of 15 min and then normalized to the 1-min count rate. Then the measured values were fitted with the exponential function and the half-life time calculated.

The derived mean value of the ^{214}Pb (354 keV) isotope half-life time (31.4 ± 2.5 min) is larger than the real value of half-life time (26.8 min). This shows that the collected rainwater contains the ^{222}Rn isotope and it continues to decay giving additional ^{214}Pb isotope, and in this way enlarging its half-life time. It means than rain washout not only gamma emitters (mostly Bismuth isotopes) but also ^{222}Rn itself.

4. Conclusions

We measured the gamma radiation of ^{222}Rn progeny during thunderstorms by precise gamma spectrometers located within the lead filter. The gamma radiation was measured from the rainwater collected during four summer storms on Aragats. The concentration the most abundant gamma emitters in the rainwater

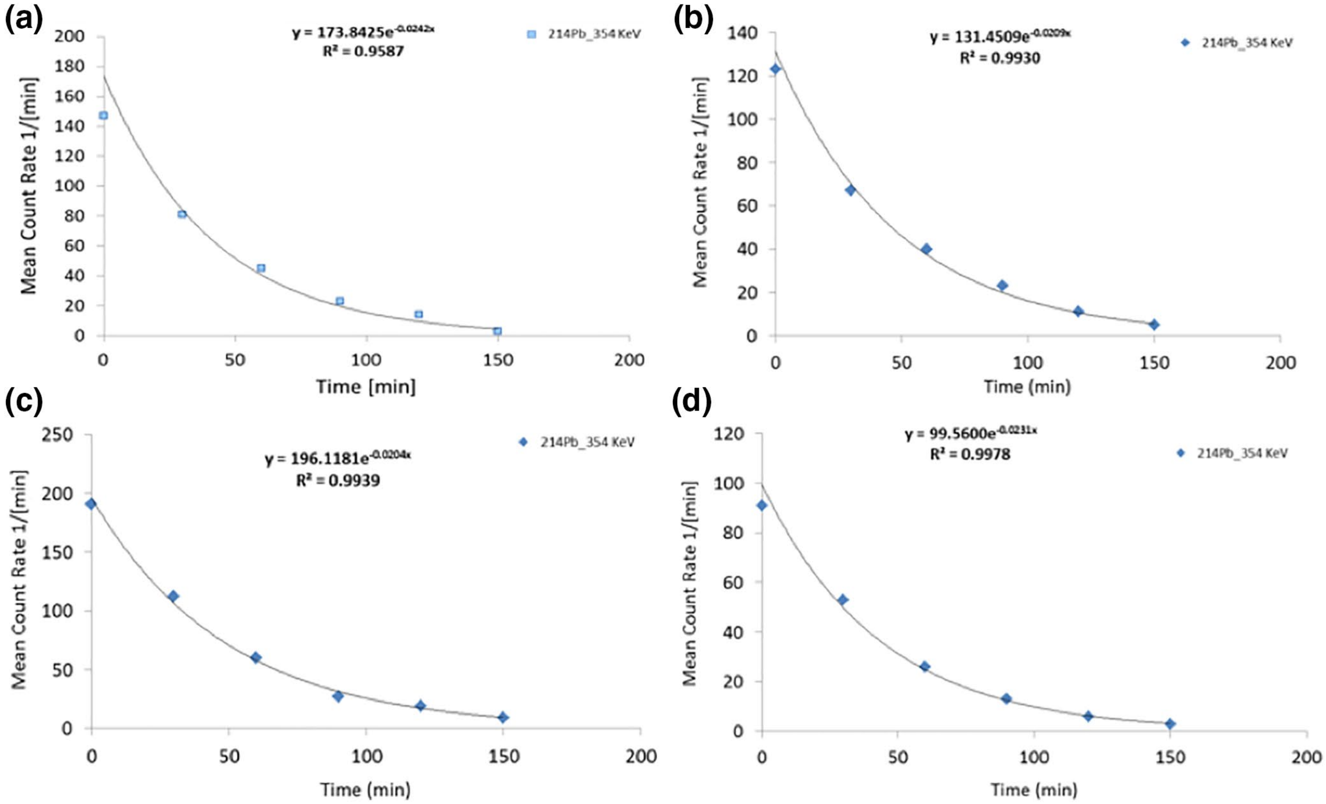

Figure 4. The exponential fit of the decay of the ^{214}Pb isotope. The intensity of the 354 keV line was measured each half-of-hour during 150 min of measurements. Solid line—the exponential fit.

^{214}Pb, ^{214}Bi (609 keV), and ^{214}Bi (1.12 MeV) was 25.3 ± 0.8%, 19.5 ± 1%, and 7.5 ± 0.2% in the first minute of the exposing of the rainwater to the ORTEC spectrometer. In the last, 150th minute of exposition, the concentration of these isotopes changed to 13.5 ± 0.7%, 25.6 ± 1.8%, and 17.1 ± 2.8% accordingly. The overall composition of the ^{222}Rn progeny in rainwater coincides well with one recovered from the registered gamma radiation of the atmospheric origin. Thus, near-surface electric field lifts the ^{222}Rn and its progeny up in the atmosphere and the rain return it backward in this way providing the circulation of the radioactive isotopes and enlarging surface radioactivity during thunderstorms.

Data Availability Statement

The data for this study are available on the WEB page of the Cosmic Ray Division (CRD) of the Yerevan Physics Institute, http://adei.crd.yerphi.am/adei.

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