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Negative correlation between the number of sunspots and the occurrence of ^{7}Be and ^{22}Na in the surface air and their contribution to radiation doses

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This article presents yearly mean concentrations of cosmogenic radionuclides ^{7}Be and ^{22}Na occurring in dry and wet depositions (fallout) and aerosols. Time dependencies negatively correlated with the yearly mean number of sunspots. Activity concentrations of ^{7}Be and ^{22}Na in aerosols in the surface air had a correlation of near-unity. ^{7}Be in aerosols exhibited a smoother time dependence than ^{22}Na, implying that the production of ^{22}Na is more sensitive to the solar activity than the production of ^{7}Be. The effect of the measured doses on the general population through internal and external exposure to radiation from cosmogenic radionuclides was small.

KEY WORDS: aerosols; assessed radiation doses; cosmogenic radionuclides; dry deposition; systematic influences; wet deposition

^{7}Be and ^{22}Na are naturally occurring radionuclides that are produced in the stratosphere and upper troposphere by interactions between the galactic cosmic rays and secondary neutrons with the atomic nuclei of nitrogen, oxygen, and argon (1–4). These radionuclides attach to aerosol particles with diameters of 0.3–0.6 μm and reach the ground-level (surface) air through convection. Their residence time in the atmosphere is around 20 days (5, 6). The presence of ^{7}Be and ^{22}Na is detected regularly in aerosols, rainwater (wet fallout), and dry deposition through environmental radioactivity monitoring (ERM) programmes (7–9). These ERMs are designed to assess exposure doses for the general population through various sources of external radiation and pathways of radionuclide intake. They are also used to verify reported emissions from sources of radioactive contamination. Current sampling methods and measurements are not sensitive enough to assess spatial and temporal distribution of ^{7}Be and ^{22}Na in the surface air at high resolution. However, by averaging activity concentrations and dry or wet deposits over the results measured at more locations over several time points it is still possible assess long-term dependencies of activity concentrations of ^{7}Be and ^{22}Na in the surface air.

The aim of this study was to demonstrate the ability long-term ERM programs currently implemented in Slovenia to measure cosmogenic radionuclides, assess exposure doses for the general population, and assess how ^{7}Be and ^{22}Na in the surface air are related to the number of sunspots. To this end, we averaged the measurement results across sampling locations over a one-year period. The production of these radionuclides depends on the spectrum of the cosmic rays, which is influenced by the solar wind inducing a deflection of the cosmic rays from the inner parts of the solar system. The effect of the solar wind can decrease the flux of low-energy cosmic rays (below 1 TeV/nucleon) in the Earth's orbit by an order of magnitude, whereas at higher energies the influence is weaker (4). Since the solar wind depends on the solar activity, which is reflected in the number of sunspots, the concentrations of ^{7}Be and ^{22}Na are expected to negatively correlate with the occurrence of sunspots.

In this paper we present the calculated means of the measurement results and discuss the one-year averages in terms of this production mechanism.

MATERIALS AND METHODS

Slovenia has set up an environmental radioactivity monitoring network operating around the nuclear power plant Krško and at a reference location in Ljubljana. This monitoring network comprises sampling stations of rainwater (wet fallout), dry fallout, and aerosols (7).

Rainwater is collected at four locations every month, with the collectors having an effective sampling surface of

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0.25 m^2. Rainwater samples are added boron and oxalic acid and the solutions evaporated to dryness (10, 11). The dry residues are then transferred to plastic canisters, pressed and measured on high-resolution gamma-ray spectrometers for two days.

Dry fallout is collected at nine locations with Plexiglas plates (with a surface area of 0.3 m^2) covered with a thin layer of Vaseline and placed at a height between 1.8 and 2 m above ground for one month. The collection efficiency is 20 % (10). After the month is over, Vaseline is removed from the plate by scraping, transferred to plastic canisters, and melted at a temperature of 60 °C to obtain a consistent sample structure with a well-defined geometry. The samples are then measured on high-resolution gamma-ray spectrometers for approximately one day.

Aerosols are collected with aerosol samplers at nine locations. However, only two locations have high-volume samplers installed (since 2006), with a typical sampling volume between 110,000 and 150,000 m^3 of air per month. Other sampling sites operate with low-volume samplers. Aerosol filters are collected monthly, squeezed into plastic canisters, and pressed. The actual thickness of filter material is determined on the next day to allow filter material to relax. Samples are measured on high-resolution gamma-ray spectrometers four days after the end of sampling period to allow thoron daughters to decay.

There are 10 high-purity Germanium (HPGe) gamma-ray spectrometers available with the efficiency between 24 and 70 % relative to that of a 3.3 inch NaI crystal, 0.7-1.0 keV resolution at low energies and 1.9-2.2 keV at 1333 keV, and graded shields (12-14). The spectrometers are connected to the Canberra's GenieESP spectroscopy platform (Meriden, CT, USA) (15). The acquired spectra are analysed automatically, taking into account the actual sample dimensions, matrix, and density (16). Coincidence-summing corrections for ^{22}Na are performed according to the method proposed by McCallum and Coote (17) as described by Korun and Martinčič (18).

RESULTS

Figures 1–4 show average yearly means of ^{7}Be and ^{22}Na activity concentrations over the sampling sites and the incidence of sunspots (19). The activity concentrations of ^{7}Be are presented without uncertainties, since the dispersion of points from a smooth curve originates from the dispersion of yearly means calculated for different sampling sites. For samples of dry residue the collection efficiency is influenced by the saturation of the Vaseline surface and the washout. In other words, dispersion largely reflects transient sampling conditions at various locations rather than the uncertainties originating from sample preparation and measurement.

Figure 3 shows the time dependence of yearly mean deposition obtained from rainwater samples. Scattered time dependence indicates the influence of systematic effects, which are not effectively taken into account. On the other hand, uncertainties in ^{22}Na activities reflect those in counting statistics and peak evaluation. The time covered by these measurements extends only to the past 12 years, during which aerosol sampling has been effective enough to allow the collected ^{22}Na activities to exceed the decision threshold. This, however, occurs only during the warm season. During the cold season, lasting six months, ^{22}Na is not detected, which has a systematic influence on ^{22}Na activity means. This could be due not only to the weaker transport of ^{22}Na to the surface air in the cold season but also due to the weaker transport of argon into the stratosphere, which decreases its concentration there. Systematic influences of the missing results on yearly means were assessed according

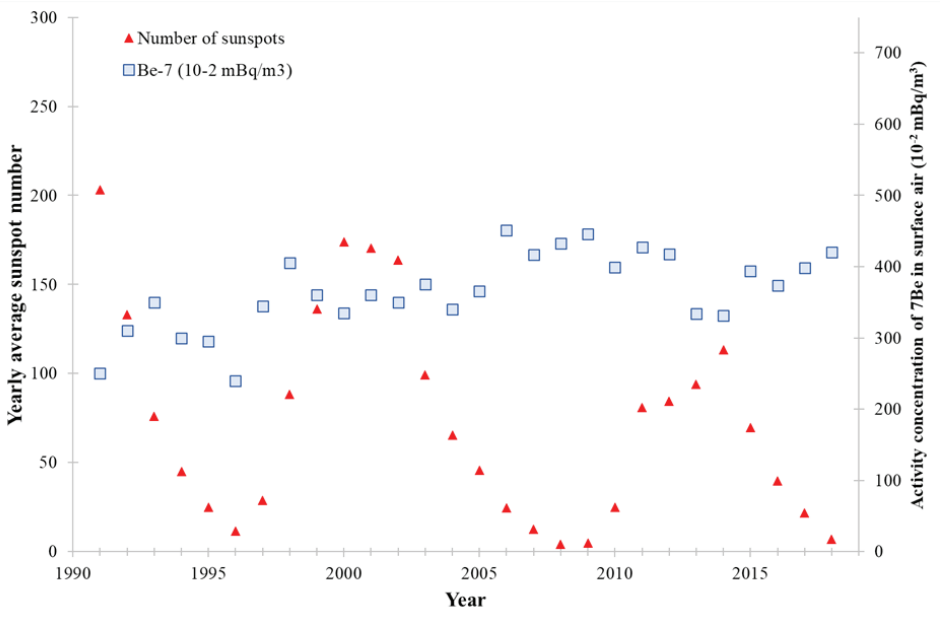

Figure 1 Yearly means of ^{7}Be concentrations in aerosols, averaged over the sampling locations

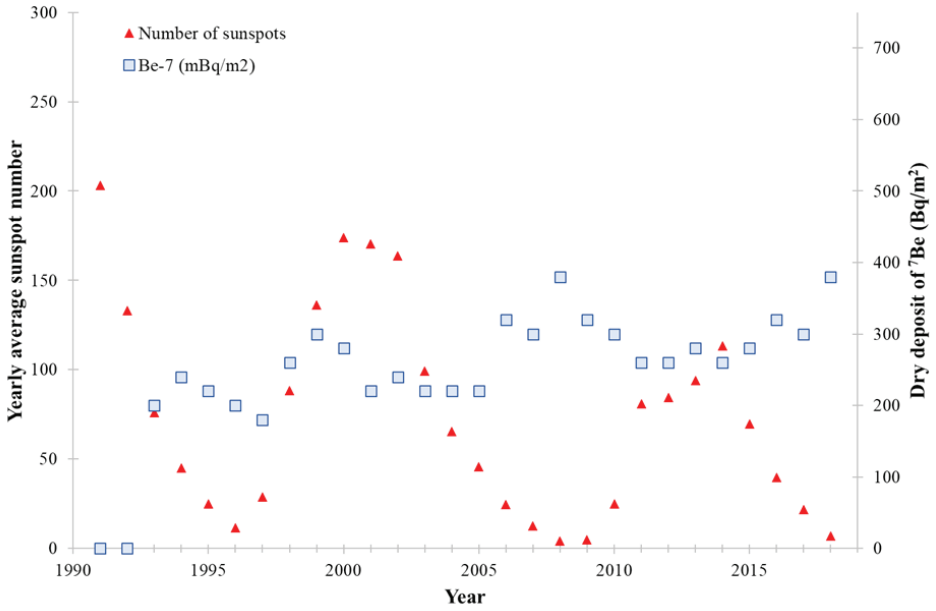

Figure 2 Yearly means of ^{7}Be concentrations in dry deposits, averaged over the sampling locations

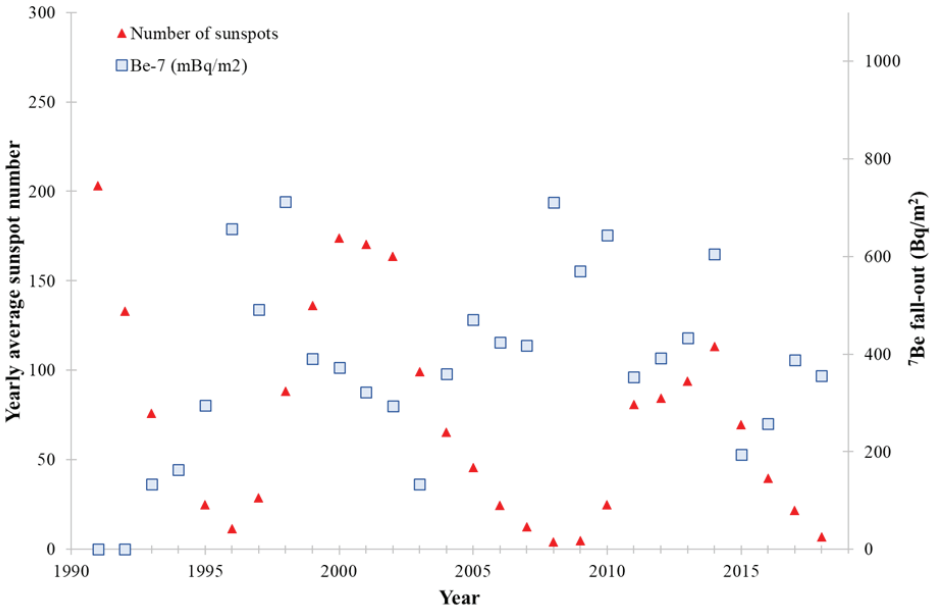

Figure 3 Yearly means of ^{7}Be concentrations in wet deposits, averaged over the sampling points

to the procedure recommended by the EU Commission (20), which says that measurement outcomes below the decision threshold should be conservatively substituted by one-half of the decision threshold. The testing of this procedure has shown that it introduces a smaller systematic influence on the means than the approaches based on best estimates (7). The systematic influence due to the missing results on the yearly averages amounts to about 150 nBq/m^{3}.

DISCUSSION

In order to arrive at reasonable conclusions, we averaged the recorded data across the sampling locations and over a one-year period. Table 1 confirms negative correlations between the mean number of sunspots and activity concentrations of ^{7}Be and ^{22}Na in aerosols and of ^{7}Be in dry and wet deposits.

Figure 5 shows a near-unity correlation between ^{7}Be and ^{22}Na activity concentrations in aerosols, which indicates not only their common origin but also their common pathways in the atmosphere.

It is clear that the obtained averages do not provide information about the transport mechanisms in the atmosphere. The time dependencies of ^{7}Be and ^{22}Na in aerosols in Figures 1 and 4 can be described as a wave with a length of 11 years, superimposed on a monotonous function representing the concentrations at high solar

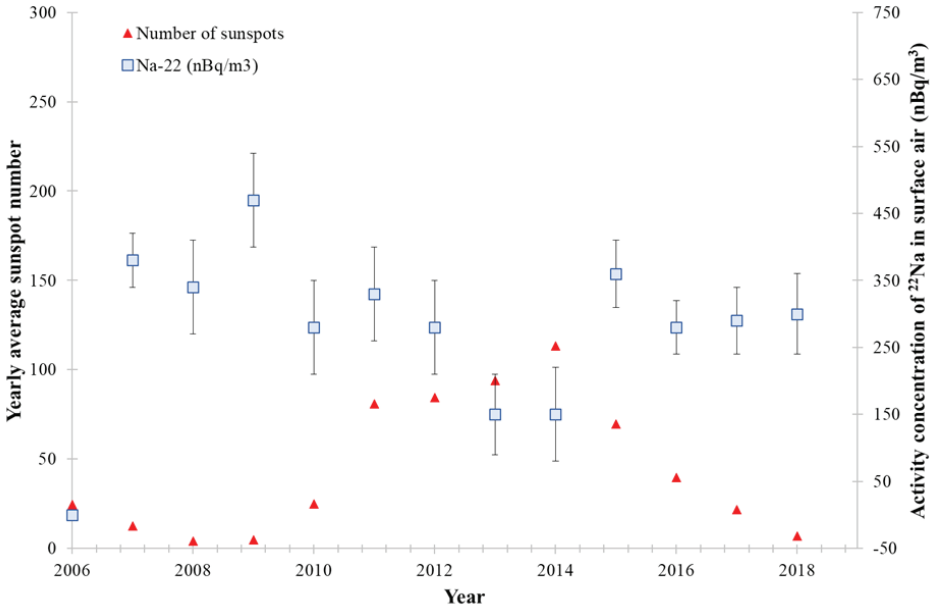

Figure 4 Yearly means of ^{22}Na concentration in aerosols, averaged over the sampling locations

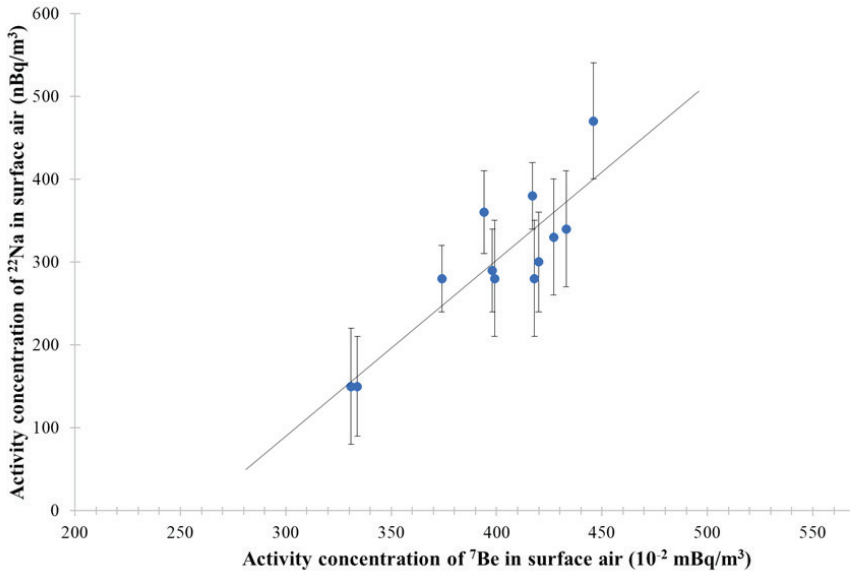

Figure 5 Correlation between yearly means of ^{7}Be and ^{22}Na concentrations in aerosols, averaged over the sampling locations

Table 1 Correlation coefficients between the number of sunspots and the activities of ^{7}Be and ^{22}Na in aerosols and dry and wet depositions

Activity	Value of the correlation coefficient
^{7}Be in aerosols	-0.40
^{22}Na in aerosols	-0.46
^{7}Be in dry deposition	-0.31
^{7}Be in wet deposition	-0.28

activity. For ^{7}Be, where the presented time interval amounts to more than two solar cycles, a linearly increasing time dependence is assumed, representing longer-term dependence. For ^{22}Na, in contrast, only a constant can be assumed, because the time interval is only one solar cycle long. By fitting the dependencies, we obtained the ratio of the amplitude of the wave and the height of the monotonous function in the middle of the interval of 0.075±0.021 for ^{7}Be and 0.19±0.06 for ^{22}Na, taking into account systematic influence due to the absence of data for the cold season. The higher ratio for ^{22}Na indicates that its production is more susceptible to solar activity than the production of ^{7}Be. Since solar activity influences the spectrum of the cosmic rays more at low than at high energies (21), it follows that the cosmic rays producing ^{22}Na have smaller energies than the cosmic rays producing ^{7}Be.

The activity concentrations of the cosmogenic radionuclei in the air and on the soil surface contribute to the yearly ambient dose equivalent. The dose equivalents induced by ^{7}Be range between 1.6 and 2.8 10^{-7} mSv in the air and between 2.6 and 4.9 10^{-7} mSv in dry deposition. The conversion coefficients used were based on the 1999 recommendations by the Joint Working Group of Radiation Protection (22). The assessed yearly effective doses to adults due to inhalation of ^{7}Be and ^{22}Na in the air are in the range of 0.9–1.5 10^{-6} mSv and 1.3–4.0 10^{-9} mSv, respectively. Here the dose conversion coefficients were taken from Slovene legislation (23). The doses assessed are many orders of magnitude below the doses from the terrestrial and cosmic background.

CONCLUSION

Long-term measurements, performed in the framework of an environmental radioactivity monitoring programme designed for assessing doses to the general population due to natural radioactivity and emissions of radioactive substances from the nuclear power plant Krško are sensitive enough to establish activity concentrations of cosmogenic radionuclides ^{7}Be and ^{22}Na in the atmosphere over many years. However, the sensitivity of spectrum analysis should be improved in order to determine ^{22}Na activity in the air during the cold season more reliably (24). The measured activities in aerosols (surface air) indicate that ^{7}Be and ^{22}Na follow the same pathways from the stratosphere to the surface air and that the production of ^{22}Na is more sensitive to the solar activity than the production of ^{7}Be. The stability of these results provides an opportunity to assess the quality of the analytical process as a whole, including sampling, sample preparation, and measurements. Our study has demonstrated that activity concentrations in aerosols and dry deposit can be measured consistently, whereas the measurement of activity concentrations in wet fallout suffers from influences that are not well controlled.

Finally, our measurements are reassuring in that the impact of cosmogenic radionuclides (^{7}Be, ^{22}Na) on the health of adult general population, either through inhalation or direct radiation, is low even at peak concentrations.

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Negativna korelacija med številom sončevih peg in koncetracijama aktivnosti ^{7}Be in ^{22}Na v površinskem zraku ter njun prispevek k dozi ionizirajočega sevanja

V članku prikazujemo letne povprečne koncentracije kozmogenih radionuklidov ^{7}Be in ^{22}Na v suhem in mokrem usedu in aerosolih od 1991. do 2018. Med koncentracijami aktivnosti ^{7}Be in ^{22}Na v aerosolih v površinskem zraku je zelo dobra korelacija. Časovni odvisnosti koncentracij aktivnosti kozmogenih radionuklidov sta negativno korelirani s povprečnim letnim številom sončevih peg. Časovna odvisnost ^{22}Na v aerosolih je bolj razgibana kot pri ^{7}Be, zato je nastajanje atomov ^{22}Na v atmosferi bolj povezano s časovnim ciklusom sončeve aktivnosti, kot to velja za atome ^{7}Be v zraku. Prispevka kozmogenih radionuklidov k celotni oceni vpliva na prebivalstvo zaradi notranjega obsevanja in zunanjega sevanja sta majhna.

KLJUČNE BESEDE: aerosoli; kozmogeni radionuklidi; mokri used; ocenjena doza ionizirajočega sevanja; sistematični vplivi; suhi used