

## Temporal Variability of the Beryllium-7 ( $^7\text{Be}$ ) Scavenging Ratio in the Sevastopol Region

D. A. Kremenchutskii

*Marine Hydrophysical Institute of RAS, Sevastopol, Russian Federation*  
✉ [d.kremenchutskii@mhi-ras.ru](mailto:d.kremenchutskii@mhi-ras.ru)

### Abstract

**Purpose.** The study is purposed at identifying the features of temporal variability in the relation of  $^7\text{Be}$  scavenging ratio at the seasonal and interannual time intervals depending on the amount and frequency of precipitation.

**Methods and Results.** The scavenging ratio was assessed based on the field data on the  $^7\text{Be}$  concentration values in the atmosphere and precipitation in 2012–2020. Application of ANOVA made it possible to examine the relationship between the scavenging ratio and the variations in  $^7\text{Be}$  concentrations in the atmosphere and precipitation. The relationship between the scavenging ratio and the precipitation amount and frequency was studied by the correlation method of analysis.

**Conclusions.** The scavenging ratio values averaged over a season and a year varied within the ranges 423–1286 and 508–919, respectively. The geometric mean value of the scavenging ratio was  $719_{-173}^{+227}$ .

The intraannual variability of the scavenging ratio is absent at the 95% confidence level. The variability in average annual values of the scavenging ratio demonstrates a decrease in 2013 (as compared to 2012) from 664 to 508, an increase to 919 in 2016 and again a decrease to 516 in 2020. The ANOVA results indicate that variability of the scavenging ratio values averaged over a season and a year is due to the variation in  $^7\text{Be}$  concentration in precipitation by 90 and 74%, respectively. In its turn, the long-term seasonal variability of  $^7\text{Be}$  concentration in precipitation is conditioned by the variations in  $^7\text{Be}$  concentration in the atmosphere ( $r = 0.64$ ) and in the precipitation amount ( $r = -0.50$ ). The relationship between the annual values of  $^7\text{Be}$  concentration in precipitation and its concentration in the atmosphere or with the precipitation parameters has not been revealed. The correlation analysis results indicate that the variability of precipitation parameters (amount and frequency) produces no statistically significant effect at the 95% confidence level upon the variability of scavenging ratio values at seasonal and annual time intervals.

**Keywords:** Beryllium-7 ( $^7\text{Be}$ ), precipitation, atmospheric aerosol, scavenging ratio

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### Introduction

Beryllium-7 ( $^7\text{Be}$ ) is a radionuclide of cosmogenic origin characterized by a relatively short half-life ( $\sim 53$  days). It is formed in the atmosphere: approximately 2/3 in the stratosphere and the remaining 1/3 in the troposphere [1].  $^7\text{Be}$  enters the marine environment mainly (80–90%) with precipitation [2].



The remaining 10–20% accrue to the dry deposition flux. Works discussing the possibility of using [3–6] or using [7–9] this radionuclide as a tracer to obtain quantitative estimates of the entry of other substances (primarily so-called geotracers) from the atmosphere into the marine environment have been issued since the last decade. For this purpose, the parameters for removing substances from the atmosphere based on data on the  $^7\text{Be}$  concentration in the atmosphere and precipitation (or marine environment) are first calculated and then their flux is estimated. Such parameters can be deposition velocity and scavenging ratio according to [5, 10–12]. The deposition velocity relates the substance flux to its concentration in the atmosphere while the scavenging ratio relates the substance concentration in precipitation to its concentration in the atmosphere. Although each of these two parameters can be used to obtain quantitative estimates of both substance flux from the atmosphere with precipitation and its concentration in precipitation, the first parameter (deposition velocity) receives objectively more attention than the second one (scavenging ratio). In particular, the available literature contains information on the monthly, seasonal and annual average values of the  $^7\text{Be}$  deposition velocity, their temporal variability and factors determining it [11, 13–15]. At the same time, the depth of research into the scavenging ratio is limited by calculating this parameter value [11, 16–18].

The study is purposed at identifying the features of temporal variability in the relation of  $^7\text{Be}$  scavenging ratio in rainfall to its concentration in the atmosphere at the seasonal and interannual time intervals depending on the amount and frequency of precipitation.

## Materials and methods

**Determination of  $^7\text{Be}$  concentration in the surface atmosphere and in atmospheric precipitation.** The methods used for collecting and processing samples of atmospheric precipitation and aerosols with the subsequent determination of  $^7\text{Be}$  activity in them are described in detail in [15, 19, 20]. A summary of these methods is given below.

Atmospheric aerosol and precipitation samples were taken from the roof of the building of Marine Hydrophysical Institute of RAS. A Petryanov filter (FPP-15-1.5) was used to concentrate the aerosol. The average speed of air pumping through the filter was  $\sim 525 \text{ m}^3 \cdot \text{h}^{-1}$ . As a rule, four daily samples (Monday – Friday) and one three-day sample (Friday – Monday) were taken during the week. The filter was compressed into a tablet with a diameter of 5.2 cm and a height of 0.5 cm after the completion of air filtration. Moist atmospheric precipitation samples were collected using a  $0.64 \text{ m}^2$  cuvette connected to a 50 L plastic barrel. The precipitation in the barrel, if any, was collected immediately after sampling the atmospheric aerosol. Radionuclide concentration from wet atmospheric precipitation samples was carried out using Dowex HCR-S/S cation exchange resin. The resin was packaged in a Petri dish with a diameter of 5.2 cm and a height of 1.4 cm or in a test tube with a diameter of 2.8 cm and a height of 7 cm.

In all cases,  $^7\text{Be}$  activity measurements in the samples were carried out on a gamma spectrometer with one of two detectors: a NaI(Tl) crystal with a diameter and a height of 6.3 cm or a NaI(Tl) crystal with a diameter and a height of 10 cm with a well with a diameter of 3 cm and a height of 6 cm. Both crystals had a resolution of 7% for the  $^{137}\text{Cs}$  peak and were located in a low-background multilayer protection. The single sample activity was recorded for 5–24 hours. The total error in measuring  $^7\text{Be}$  activity in samples usually did not exceed 15%.

The total number of 2,056 samples of atmospheric aerosols and 405 samples of rainfall was collected and processed in 2012–2020. The obtained atmospheric and precipitation  $^7\text{Be}$  concentrations were averaged for each individual season and year under this study. As a result, two series of average seasonal and average annual values lasting 36 seasons and 9 years were obtained for each parameter.

**Meteorological parameters.** Precipitation amount data was obtained by normalizing the precipitation sample volume to the sampler area. Precipitation frequency data represents the number of days with precipitation in a particular season or year.

**Scavenging ratio of  $^7\text{Be}$**  was calculated as follows:

$$W = \kappa \frac{C_w}{C_a}, \quad (1)$$

where  $W$  is scavenging ratio;  $\kappa$  is correction taking into account the difference in density of water and air, equal to  $1.2 \cdot 10^{-3}$ ;  $C_w$  and  $C_a$  are seasonally averaged  $^7\text{Be}$  concentrations in moist atmospheric precipitation and on atmospheric aerosols, respectively,  $\text{Bq} \cdot \text{m}^{-3}$ .

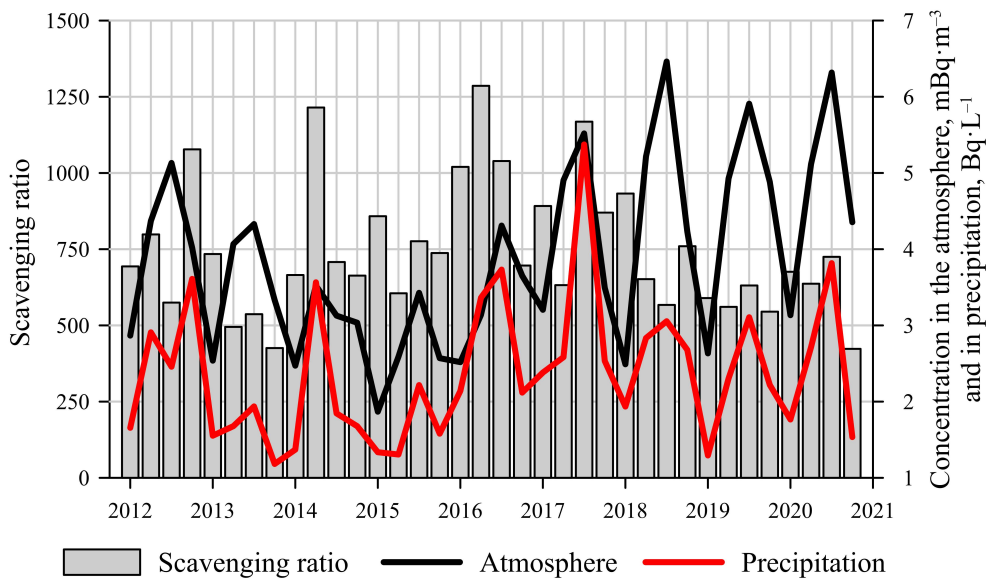
Knowing the scavenging ratio, it is possible to estimate the  $^7\text{Be}$  flux from the atmosphere with precipitation using the following equation:

$$F = \frac{WC_a Pr}{k}, \quad (2)$$

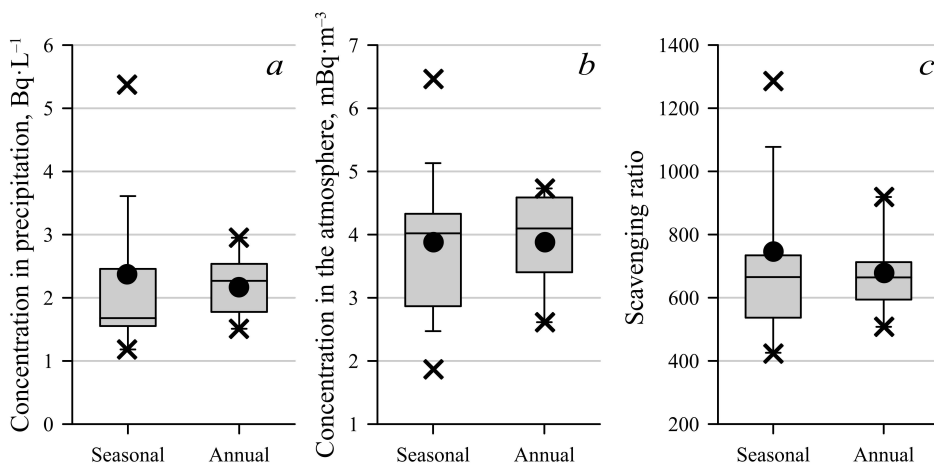
where  $F$  is flux,  $\text{Bq} \cdot \text{m}^{-2} \cdot \text{season}^{-1}$ ;  $Pr$  is precipitation amount,  $\text{mm} \cdot \text{season}^{-1}$ .

## Results and discussion

**Basic statistics.** Figure 1 shows the obtained series of observations of  $^7\text{Be}$  concentrations in the atmosphere and precipitation, as well as the scavenging ratio estimates. Seasonally and annually averaged values of  $^7\text{Be}$  concentration in precipitation varied in the ranges of 1.2–5.4 and 1.5–3.0  $\text{Bq} \cdot \text{L}^{-1}$ , respectively, in 2012–2020 (Fig. 2, *a*). The majority (80%) of the seasonally averaged concentration values were in the range of 1.4–3.6  $\text{Bq} \cdot \text{L}^{-1}$ . The 10th and 90th percentiles for average annual values coincide with the minimum and maximum of this series due to its relatively short duration for this series and the remaining two series shown in Fig. 2. Variation coefficients were 38.5 and 22.8% for seasonal and annual values. The arithmetic mean and median values of seasonally averaged  $^7\text{Be}$  concentrations were 2.4 and 2.2  $\text{Bq} \cdot \text{L}^{-1}$ , respectively. This indicates that the distribution of seasonal concentration values may be different from normal.



**Fig. 1.** Seasonal variability of  $^7\text{Be}$  concentrations in the atmosphere and precipitation, as well as their scavenging ratio



**Fig. 2.** Frequency distribution of the data on seasonal and annual values of  $^7\text{Be}$  concentrations: *a* – in precipitation, *b* – in the atmosphere; *c* – scavenging ratio

Seasonally and annually averaged values of  $^7\text{Be}$  concentrations in the atmosphere varied within the range of 1.9–6.5 and 2.6–4.7  $\text{mBq}\cdot\text{m}^{-3}$ , respectively (Fig. 2, *b*). The main variability (80%) of the seasonally averaged values occurred in the range of 2.5–5.5  $\text{mBq}\cdot\text{m}^{-3}$ . As in the case of  $^7\text{Be}$  concentration in atmospheric precipitation, the 10th and 90th percentile values for the average annual concentration of this radionuclide in the atmosphere coincide with the minimum and maximum, respectively. Variation coefficients were 30.7 and 19.5% for seasonal and annual values. The arithmetic mean and median values of seasonally averaged  $^7\text{Be}$

concentrations in the atmosphere were 3.9 and 3.6 mBq·m<sup>-3</sup>. They are close, but not equal to each other, which can indicate that their distribution is different from normal.

The seasonally and annually averaged values of the scavenging ratio varied in the ranges of 423–1286 and 508–919, respectively (Fig. 2, c), according to the estimates obtained using equation (1). The 10th and 90th percentile values were 537 and 1078 for the seasonal mean scavenging ratios. Variation coefficients were 28.8 and 19.8% for seasonal and annual values. The arithmetic mean (746) and median (695) values of seasonal ratios are close, but not equal to each other.

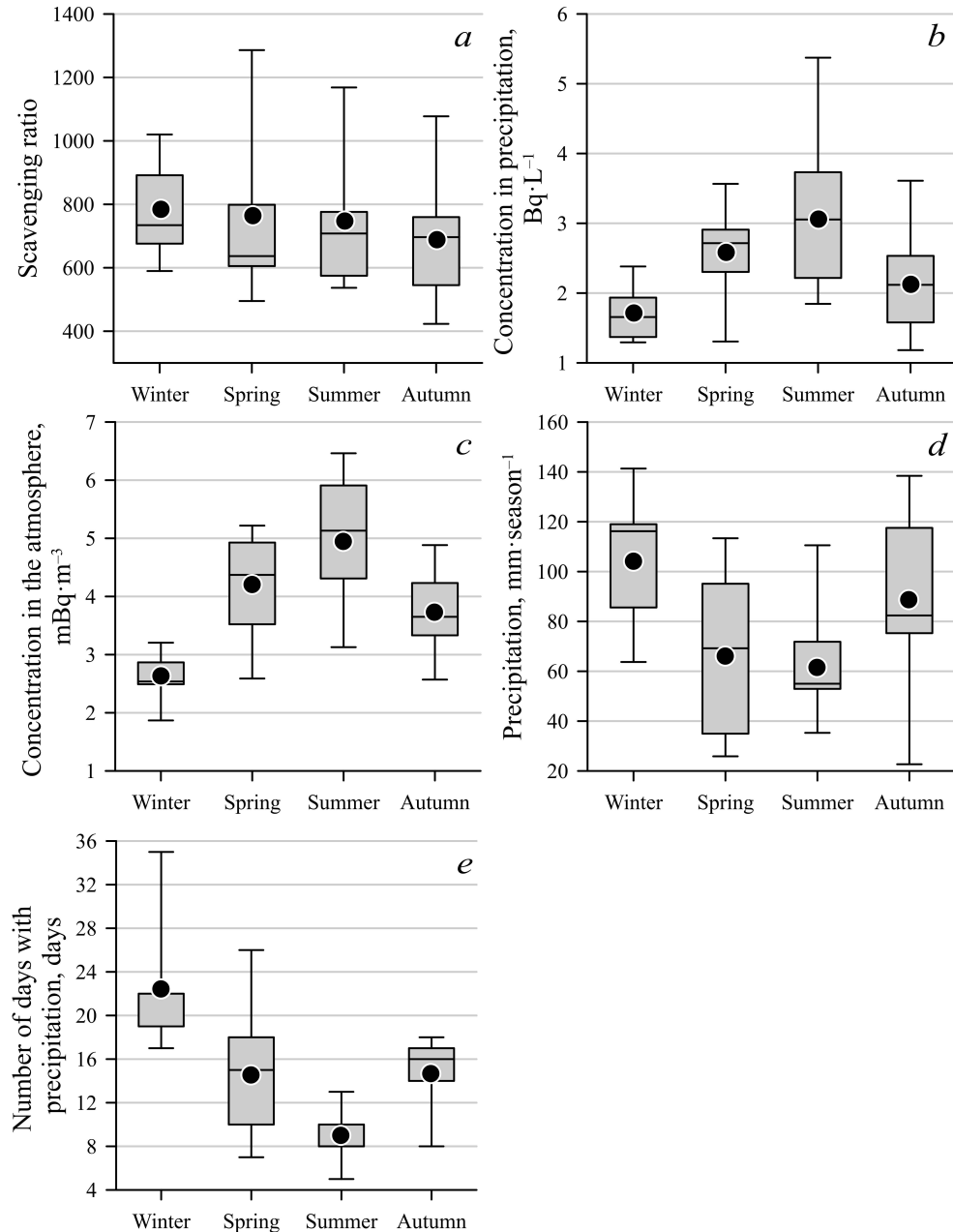
Analysis of the results of the Shapiro–Wilk tests for normality shows that the distributions of data on the <sup>7</sup>Be concentration in precipitation and on scavenging ratio are statistically significantly different from normal at the 95% confidence level. On the contrary, the distribution of data on the <sup>7</sup>Be concentration in the atmosphere is not statistically significantly different from normal at the 95% confidence level.

The obtained mean <sup>7</sup>Be concentrations in precipitation (geometric mean – 2.2<sup>+1.0</sup><sub>-0.7</sub> Bq·L<sup>-1</sup>) and in the atmosphere (arithmetic mean 3.9 ± 1.2 mBq·m<sup>-3</sup>), and scavenging ratio (geometric mean – 719<sup>+227</sup><sub>-173</sub>) coincide well with the literature data. The available literature shows the following values concerning <sup>7</sup>Be concentration in precipitation (Bq·L<sup>-1</sup>): 2.9 – for Detroit, Michigan, USA [16]; 2.5 – for Huelva, Spain [11]; 2.6 – for Malaga, Spain [17]; 1.1<sup>+1.8</sup><sub>-0.7</sub> – for Mangalore, India [18]. The following values are given concerning <sup>7</sup>Be concentration in the atmosphere (mBq·m<sup>-3</sup>): 4.8 – for Detroit, Michigan, USA [16]; 4.9 – for Huelva, Spain [11]; 4.0 – for Lisbon, Portugal [21]; 4.2 – for Malaga, Spain [17]; 6.9<sup>+2.1</sup><sub>-1.6</sub> – for Mangalore, India [18]. The following estimates have been published regarding the scavenging ratios: 948 – for Detroit, Michigan, USA [16]; 496 ± 180 – for Huelva, Spain [11]; 840 – for Malaga, Spain [17]; 553<sup>+719</sup><sub>-312</sub> – for Mangalore, India [18].

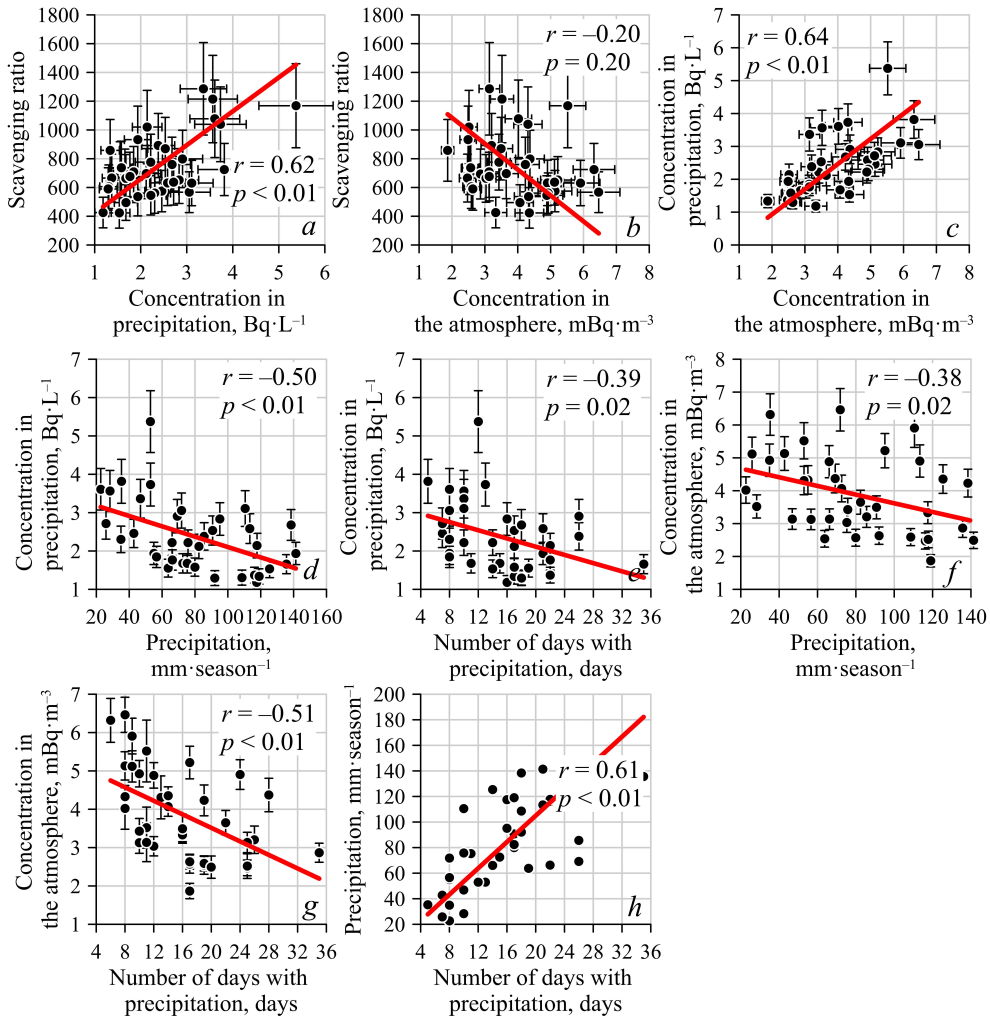
**Seasonal variability of the scavenging ratio** averaged over the entire observation period is shown in Fig. 3, a. No pronounced seasonal variability can be seen in the series under consideration. An ANOVA was carried out in order to confirm that the difference in seasonal mean scavenging ratios was not statistically significant. Its results show that the seasonal average values differ from each other statistically insignificantly at the 95% confidence level. Thus, it can be concluded that the period under study revealed no seasonal variability in the scavenging ratio.

The ANOVA was carried out in order to obtain quantitative estimates of the influence of variations in <sup>7</sup>Be concentration in the atmosphere and in precipitation on the seasonal variability of the scavenging ratio. Its results show that the temporal variability of the scavenging ratio is determined by variations in <sup>7</sup>Be concentration in precipitation and in the atmosphere by 90 and 10%, respectively. The correlation analysis results indicate a strong, statistically significant correlation at the 99% confidence level ( $r = 0.62$ ) between the scavenging ratio and <sup>7</sup>Be concentration values in precipitation (Fig. 4, a). No statistically significant correlation between the scavenging ratio variability and <sup>7</sup>Be concentration in the atmosphere can be observed even at the 90% confidence level

( $r = -0.20$ ) (Fig. 4, *b*). No relationship between the long-term variability of seasonal average scavenging ratio and the precipitation amount and frequency was confirmed as the correlation coefficients were  $-0.25$  ( $p = 0.14$ ) and  $0.004$  ( $p = 0.98$ ), respectively.



**Fig. 3.** Averaged over the observation period data on seasonal variability of: scavenging ratio (*a*), <sup>7</sup>Be concentration in precipitation (*b*), <sup>7</sup>Be concentration in the atmosphere (*c*), precipitation amount (*d*) and precipitation frequency (*e*)



**Fig. 4.** Relationship between the seasonal values of various parameters: *a* – scavenging ratio and  ${}^7\text{Be}$  concentration in precipitation; *b* – scavenging ratio and  ${}^7\text{Be}$  concentration in the atmosphere; *c* –  ${}^7\text{Be}$  concentrations in precipitation and atmosphere; *d* –  ${}^7\text{Be}$  concentration in precipitation and precipitation amount; *e* –  ${}^7\text{Be}$  concentration in precipitation and a number of days with precipitation, *f* –  ${}^7\text{Be}$  concentration in the atmosphere and precipitation amount; *g* –  ${}^7\text{Be}$  concentration in the atmosphere and precipitation frequency; and *h* – precipitation amount and precipitation frequency

Fig. 3, *b* shows the intraannual variability of  ${}^7\text{Be}$  concentration in precipitation averaged over the entire observation period. The ANOVA results confirm a statistically significant seasonal variability of  ${}^7\text{Be}$  concentration in precipitation with a maximum in the summer season ( $2.9 \text{ Bq}\cdot\text{L}^{-1}$ ) and a minimum in the winter season ( $1.7 \text{ Bq}\cdot\text{L}^{-1}$ ) at the 95% confidence level. The correlation analysis results show that the temporal variability of the seasonally averaged  ${}^7\text{Be}$  concentration in precipitation is statistically significantly associated at the 95% confidence level with the temporal variability of  ${}^7\text{Be}$  concentration in the atmosphere ( $r = 0.64$ ), as well as with the amount ( $r = -0.50$ ) and frequency ( $r = -0.39$ ) of precipitation

(Fig. 4, *c – e*). Note that the strongest connection is observed precisely with  $^7\text{Be}$  concentration in the atmosphere. Thus, an increase in the  $^7\text{Be}$  concentration in the atmosphere and a decrease in the precipitation amount and frequency are accompanied by an increase in the radionuclide concentration in precipitation.  $^7\text{Be}$  concentration in the atmosphere takes on a maximum value in the summer season (Fig. 3, *c*) and the precipitation amount and frequency is minimal (Fig. 3, *d, e*), which explains the observed maximum concentration of this radionuclide in precipitation in this season (Fig. 3, *b*).  $^7\text{Be}$  concentration in the atmosphere takes a minimum value in winter and the precipitation amount and frequency is maximum, which determines the minimum  $^7\text{Be}$  concentration in precipitation observed at this time. The authors of [17] noted the presence of a similar seasonal variation in  $^7\text{Be}$  concentration in precipitation for a station located in Malaga, Spain. They indicated that such seasonal variability is most likely due to the intraannual distribution of precipitation and  $^7\text{Be}$  concentrations in the atmosphere, which were similar to those noted in the present study.

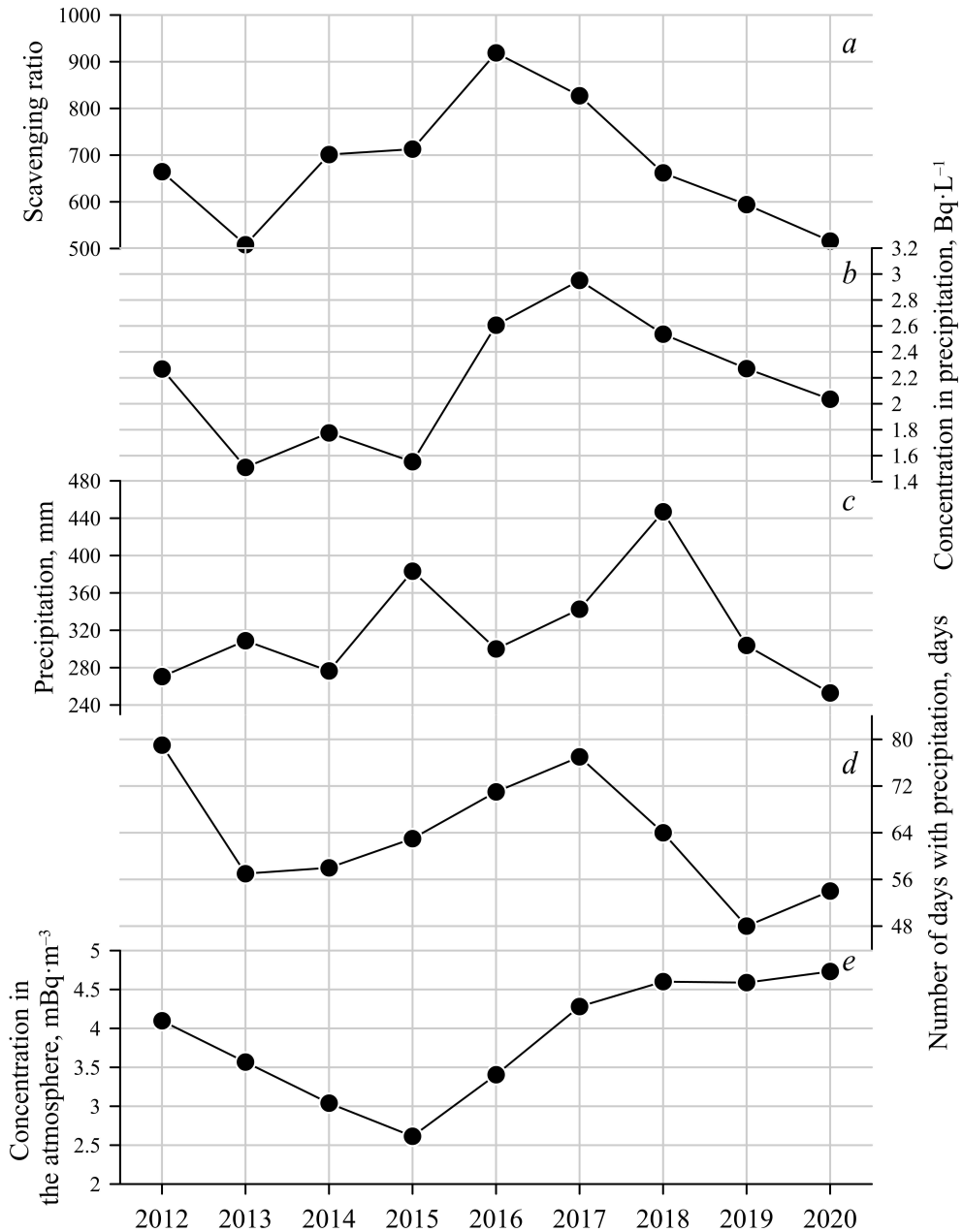
Temporal variability of seasonally averaged values of  $^7\text{Be}$  concentration in the atmosphere is statistically significantly associated with the precipitation amount ( $r = -0.38$ ) and frequency ( $r = -0.51$ ) at the 95% confidence level (Fig. 4, *f, g*). Thus, a decrease in the precipitation amount and frequency increases  $^7\text{Be}$  content in the atmosphere. In turn, the considered precipitation parameters (amount and frequency) are also interrelated. Thus, an increase in the frequency of precipitation is accompanied by an increase in its quantity with a correlation coefficient of 0.61 (Fig. 4, *h*). This relationship between the seasonal variability of  $^7\text{Be}$  concentration in the atmosphere and precipitation parameters, as well as the relationship between the parameters under consideration, has already been noted in recent studies [15, 22].

**Interannual variability of the scavenging ratio.** Figure 5 shows the resulting series of observations of interannual variability of the studied parameters.

A decrease in the scavenging ratio values from 664 to 508 was observed in 2012–2013, then followed by a long period of their growth to 919 in 2016, which then was followed by a long period of decrease to 516 in 2020. Thus, the average annual scavenging ratio values changed by 1.8 times during the period under study. Interannual variability of  $^7\text{Be}$  concentration in precipitation shows similar dynamics: a decrease from 2.3 to 1.5  $\text{Bq}\cdot\text{L}^{-1}$  was observed in 2012–2013, then followed by a short period of relatively small fluctuations (1.8 and 1.6  $\text{Bq}\cdot\text{L}^{-1}$  in 2014 and 2015), an increase in concentration to 3.0  $\text{Bq}\cdot\text{L}^{-1}$  in 2017 and then its decrease to 2.0  $\text{Bq}\cdot\text{L}^{-1}$  in 2020. Thus, the average annual values of  $^7\text{Be}$  concentration in precipitation changed by a factor of 2 in 2012–2020.

The ANOVA results show that the temporal variability of the average annual scavenging ratio values is determined by 74% by  $^7\text{Be}$  concentration variations in precipitation and by 26% in the atmosphere. Thus, the contribution of variability in  $^7\text{Be}$  concentration in the atmosphere to the variation in average annual scavenging ratios increased by 2.6 times compared to the same for seasonal values.





**Fig. 5.** Temporal variability of annual values of: scavenging ratio (a), <sup>7</sup>Be concentration in precipitation (b), precipitation amount (c), precipitation frequency (d) and <sup>7</sup>Be concentration in the atmosphere (e)

The correlation analysis results show no statistically significant relationships at the 95% confidence level between the variability of annual concentration ratios and the amount ( $r = 0.19$ ,  $p = 0.63$ ) or frequency ( $r = 0.66$ ,  $p = 0.06$ ) of precipitation. A strong, statistically significant correlation at the 90% confidence level between the scavenging ratio and the number of days with precipitation (the more rainy days in a year, the higher the average annual value of the scavenging ratio) is a rather

unexpected result. Formula (1) shows that the scavenging ratio value is directly proportional to  $^7\text{Be}$  concentration in precipitation and inversely proportional to its concentration in the atmosphere. On a synoptic time scale, an increase in the precipitation frequency can lead to a decrease in the  $^7\text{Be}$  concentration in the atmosphere due to the fact that it takes time for its concentration to recover after precipitation (1–2 days) [23]. As a result of a decrease in  $^7\text{Be}$  concentration in the atmosphere, its concentration in precipitation decreases proportionally (all other precipitation parameters being equal). Thus, no relationship between precipitation frequency and scavenging ratios is to be expected (the lack of such a relationship for temporal variability in seasonal values is reported above). Physical processes that cause such a relationship between annual values are not obvious.

No statistically significant correlations at the 90% confidence level between  $^7\text{Be}$  concentration in the atmosphere and the studied precipitation parameters were indicated. Thus, the obtained values of the correlation coefficients between  $^7\text{Be}$  concentration in the atmosphere and the precipitation amount and frequency were 0.18 ( $p = 0.64$ ) and 0.53 ( $p = 0.14$ ), respectively.

### Conclusions

This paper presents field data on the temporal variability of  $^7\text{Be}$  concentrations in the atmosphere and precipitation in 2012–2020. Seasonally averaged concentrations of this radionuclide in the atmosphere and precipitation varied in the ranges of 1.9–6.5  $\text{mBq}\cdot\text{m}^{-3}$  and 1.2–5.4  $\text{mBq}\cdot\text{L}^{-1}$ . These data made it possible to obtain quantitative estimates of the seasonal and annual scavenging ratio values, which were 423–1286 and 508–919, respectively, with a geometric mean of  $719_{-173}^{+227}$ . The ANOVA results show that temporal fluctuations in seasonal and annual scavenging ratio values are determined to a greater extent by variations in  $^7\text{Be}$  concentration in precipitation (90 and 74%, respectively) than by variations in its concentration in the atmosphere. The correlation analysis results indicate that fluctuations in seasonal values of  $^7\text{Be}$  concentration in precipitation are associated with variations in its concentration in the atmosphere ( $r = 0.64$ ,  $p < 0.01$ ) and with variability in precipitation amount ( $r = -0.50$ ,  $p < 0.01$ ). The variability of annual values of  $^7\text{Be}$  concentration in precipitation is statistically significant at the 95% confidence level and is not associated with variations in the considered precipitation parameters (amount and frequency). The analysis results indicate that there is no relation between time series of concentration ratios and precipitation parameters at seasonal and annual time intervals. In other words, it is not possible to estimate the seasonal or annual average scavenging ratio using precipitation amount and frequency data only.

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*About the author:*

**Dmitrii A. Kremenchutskii**, Senior Researcher, Marine Hydrophysical Institute of RAS (2 Kapitanskaya Str., Sevastopol, 299011, Russian Federation), CSc (Geogr.), **ORCID ID: 0000-0002-8747-6612**, **ResearchID: AAC-1673-2020**, [d.kremenchutskii@mhi-ras.ru](mailto:d.kremenchutskii@mhi-ras.ru)

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