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Temporal Variability of Aerosol Wet Deposition Velocity in the Sevastopol Region: Observational Data

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Abstract

Purpose. The study is purposed at identifying both the features of seasonal and interannual variability of the aerosol wet deposition velocity and the factors that determine this variability on the time scales under study.

Methods and Results. The deposition velocity in 2012–2020 was estimated using the field data on temporal variability of the ^{7}Be concentration on atmospheric aerosols and the ^{7}Be "wet" deposition fluxes. The correlation analysis permitted to assess quantitatively the influence of the precipitation amount and frequency upon the seasonal and interannual variability of the deposition velocity. The multiple regression analysis was applied for constructing the regression models.

Conclusions. The deposition velocity varies from 0.21 to 1.40 cm·s^{-1} and averages 0.62 ± 0.29 cm s^{-1}. It has been established that its seasonal variability is conditioned by the amount and frequency of precipitation, whereas its interannual variability – only by the precipitation amount. Based on the obtained results, two regression models were been proposed. The first model describes seasonal variability of the deposition velocity, while the second one – the interannual variability of this parameter. The corresponding time series of precipitation variability data are used in both models as predictors. The validation results indicate that the errors in the obtained estimates constitute 21.1 and 12.9% for the seasonal and annual values of wet deposition velocity, respectively.

Keywords: Beryllium-7 (^{7}Be), precipitation, wet deposition velocity, atmospheric aerosol, wet deposition flux

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Introduction

Atmospheric aerosol is solid and liquid particles suspended in the atmosphere. It can be of natural (e.g. pollen, soil erosion and volcanic eruptions) and man-caused (e.g. soot, industrial emissions) origin. Atmospheric aerosol contains organic and inorganic substances [1]. Aerosol is removed from the atmosphere because of gravitational settling and washing out by precipitation. Aerosol flux from the atmosphere to the sea surface is an important source of many substances (including nutrients, radionuclides and geotracers) entering the marine environment [2–8]. In particular, this flux leads to phytoplankton production increase, promotes nitrogen fixation [9–13] and also affects biogeochemical processes in the World Ocean waters [14, 15].

Atmospheric aerosol wet deposition by precipitation is the dominant mechanism for its removal from the atmosphere [16]. The wet deposition velocity is a parameter that relates the concentration of an aerosol in the atmosphere (or a substance contained on an aerosol) with its flux to the underlying surface. Thus, by estimating the wet deposition velocity and knowing the studied substance in the atmosphere, it is possible to calculate its flux from the atmosphere into the marine environment. Direct measurements of the aerosol wet deposition velocity are not possible. Traditionally, radionuclides are convenient tracers in studies of various processes occurring in the atmosphere, including the atmospheric aerosol wet deposition [16]. One of these radionuclides is beryllium-7 (^{7}Be) [6, 17].

Beryllium-7 is a natural radionuclide of cosmogenic origin, continuously produced mainly in the upper layers of the atmosphere, its half-life is ~ 53 days. After formation, this radionuclide is adsorbed on submicron atmospheric aerosol and transported with it to the lower layers of the atmosphere. ^{7}Be is removed from the atmosphere mainly (80–90%) by the aerosol wet deposition by "wet" atmospheric deposition [16].

Studies of the aerosol wet deposition velocity in the Black Sea region have not been carried out before.

The present paper is aimed to reveal the features of the seasonal and interannual variability of the aerosol wet deposition velocity and to identify the factors that determine this variability on the time scales under study. To achieve this aim, long-term series of field data on the temporal variability of the ^{7}Be concentration in the atmosphere and its fluxes with rainfall are presented and analyzed. These data were used to obtain quantitative estimates of the aerosol wet deposition velocity.

Materials and methods

Rainfall sampling. In the period of January 2012 – December 2020, 405 rainfall samples were taken and processed. They were taken from the Marine Hydrophysical Institute (MHI) of RAS (Sevastopol) rooftop using an enameled cuvette (area 0.64 m^2), located at the height of ~ 1.6 m relative to the roof level and connected to a plastic 50-liter container. This sampler design enables efficient collection of a large amount of rainwater and minimization of its loss due to evaporation. In case of rainfall on weekdays, samples were taken once a day at approximately 11:00 local time. In the event of rainfall over the weekend, rainwater was accumulated. This water was sampled on the next working day. Before sampling the sediments, the cuvette was washed with 300 ml of a 1–2% nitric or sulfuric acid and 300 ml of distilled water solution. This procedure makes it possible to minimize the losses of ^{7}Be as a result of its sorption on the sampler walls. The pH value of a rainwater sample in a plastic container was lowered to about two with a 30% solution of nitric or sulfuric acid. The water was poured from the container and then after 8–24 h transported to the laboratory. Acidification of the sample and a time delay between sampling and its transportation to the laboratory enabled to minimize possible losses of ^{7}Be as a result of its sorption on the container walls. In the laboratory, the sample was first filtered from insoluble impurities using paper filters, then its volume was measured and rainwater was passed at a rate of 5–50 ml·min^{-1} through two columns filled

with Dowex HCR-S/S cation exchanger. Each column contained ~ 14 ml of the cation exchanger. The ^{7}Be activity distribution between two columns with a cation exchange resin was used to determine the radionuclide extraction efficiency from a rainfall sample. The ^{7}Be activity in the cation exchanger was measured either in a plastic Petri dish 52 mm in diameter and 14 mm high, or in a plastic vial 28 mm in diameter and 70 mm high.

Atmospheric aerosol sampling. During the specified period, 2,056 samples of atmospheric aerosols were selected and processed. The sampling technique for atmospheric aerosols is described in detail in [18]. This paper provides a brief summary of it. Aerosol samples were taken from the rooftop of the institute building with a high-performance (volumetric pumping rate ~ 525 $m^3 \cdot h^{-1}$) air filtration unit using a Petryanov fiber filter (FPP-15-1.5). This fiber filter collects aerosols 0.2 μm in size with 99% efficiency. On weekdays, the filter was changed once a day at approximately 11:00 local time. The filter was not changed over the weekend. Thus, in the usual mode, four daily samples and one three-day sample were taken per week. At the end of sampling, the filter was pressed into a tablet with a diameter of 52 mm and a height of 5 mm. Measurement of ^{7}Be activity in the sample was carried out 7–10 days after the sample was taken. This time delay makes it possible to reduce the activity of short-lived gamma-active radionuclides (decay products of radon and thoron) in the sample by orders of magnitude. Reducing the activity of these radionuclides considerably simplifies the gamma spectrum form and its analysis procedure.

Measurements of ^{7}Be activity in the selected samples were carried out using two low-background gamma spectrometers with NaI(Tl) scintillation detectors. The first gamma spectrometer had a crystal 63 mm in diameter and 63 mm high, with a resolution of 7% for the ^{137}Cs peak. This detector was protected by 15 cm lead, 5 mm cadmium, 3 mm copper and 1 cm Plexiglas. The second gamma ray spectrometer had a crystal 100 mm in diameter and 100 mm high, with a well 30 mm in diameter and 60 mm high, with a resolution of 7% for the ^{137}Cs peak. The second detector was protected by 14 cm of lead and 15 cm of cast iron. The measurement time for a single sample varied in the range of 5–24 h and depended on the ^{7}Be activity in the sample. The measurement error of ^{7}Be activity in rainwater and atmospheric aerosol samples usually did not exceed 15 and 10%, respectively.

Precipitation data. Precipitation amount estimates were obtained by normalizing the sampled precipitation volume to the sampler area. Precipitation frequency refers to the number of days with precipitation during the time period under consideration.

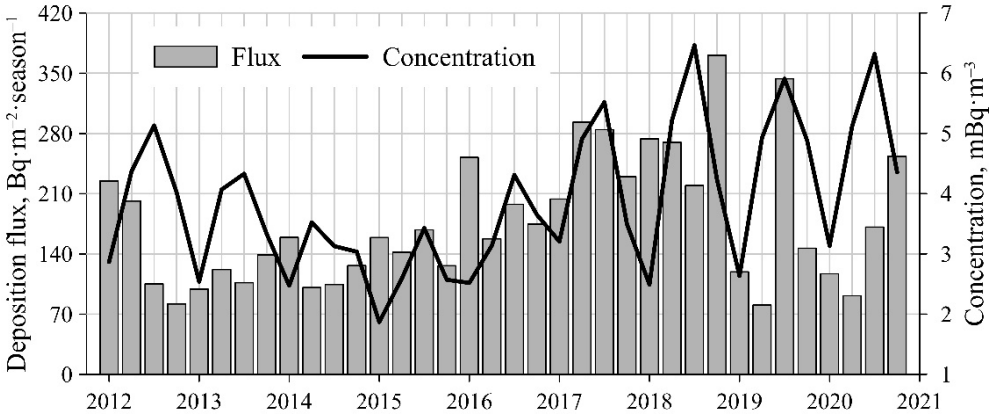
The aerosol wet deposition velocity from the atmosphere was calculated using the following formula:

$$V_d = \frac{F}{C_a}, \tag{1}$$

where V_d is wet deposition velocity, $cm \cdot s^{-1}$; F is seasonally or annually averaged ^{7}Be flux from the atmosphere, $Bq \cdot cm^{-2} \cdot s^{-1}$; C_a – seasonally or annually averaged ^{7}Be concentration in the atmosphere, $Bq \cdot cm^{-3}$.

Results and discussion

Statistical characteristics of data series on the ^{7}Be flux and concentration. The obtained values of the ^{7}Be flux and its concentration in the atmosphere were used to calculate the seasonal and annual characteristics of these parameters. For that, the flux data were summed for each individual season and year, and the concentration data were averaged. The time series of ^{7}Be seasonal fluxes and concentrations are shown in Fig. 1.



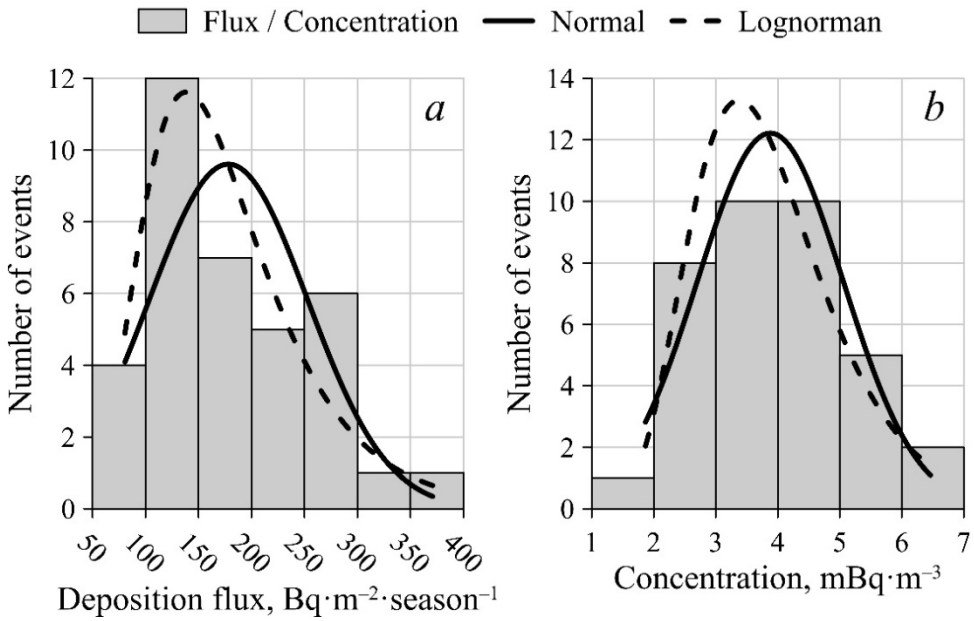
F i g. 1. Seasonal variability of the ^{7}Be flux and concentration

The ^{7}Be flux values with rainfall and its concentration in the atmosphere varied in the ranges of 81–371 $Bq \cdot m^{-2} \cdot season^{-1}$ and 1.9–6.5 $mBq \cdot m^{-3}$, respectively. The mean values were 178 ± 76 $Bq \cdot m^{-2} \cdot season^{-1}$ and 3.9 ± 1.2 $mBq \cdot m^{-3}$ for flux and concentration. The frequency distribution of the discussed series data is shown in Fig. 2. The studied series distribution is closer to lognormal than to normal, which, according to the published data in [19–21], is typical for this kind of data. The results of the Shapiro-Wilk and Anderson-Darling normality tests show that the frequency distributions of both parameters differ from the normal not statistically significantly at the 99% confidence level. The coefficients of variation were 43 and 31% for the flux and concentration data, respectively, indicating the presence of significant temporal variability in the studied series.

Averaged over the entire period of observations, the annual total value of the ^{7}Be flux with "wet" atmospheric depositions is 712 ± 227 $Bq \cdot m^{-2} \cdot yr^{-1}$. The resulting mean flux is consistent with the published data: 527 $Bq \cdot m^{-2} \cdot yr^{-1}$ in Damascus, Syria [22]; 738 $Bq \cdot m^{-2} \cdot yr^{-1}$ in Riso, Denmark [23]; 736 $Bq \cdot m^{-2} \cdot yr^{-1}$ in Thessaloniki, Greece [24]; 785 $Bq \cdot m^{-2} \cdot yr^{-1}$ in Alexandria, Egypt [25]. For comparison, the data used were from those stations, where close annual precipitation totals were observed.

The mean annual value of the ^{7}Be concentration in the atmosphere averaged over the entire observation period is 3.9 ± 0.8 $mBq \cdot m^{-3}$. This value is in good

agreement with the published data for stations located in the middle latitudes of the Northern Hemisphere: 4.7 mBq·m^{-3} in Valencia, Spain (39.4°N) [26]; 4.2 mBq·m^{-3} in Malaga, Spain (36.7°N) [27]; 3.8 mBq·m^{-3} in Barcelona, Spain (41.3°N) [28]; 3.2 mBq·m^{-3} in Bilbao, Spain (43.1°N) [20]; 3.7 mBq·m^{-3} in Ljubljana, Slovenia (46.0°N) [28].

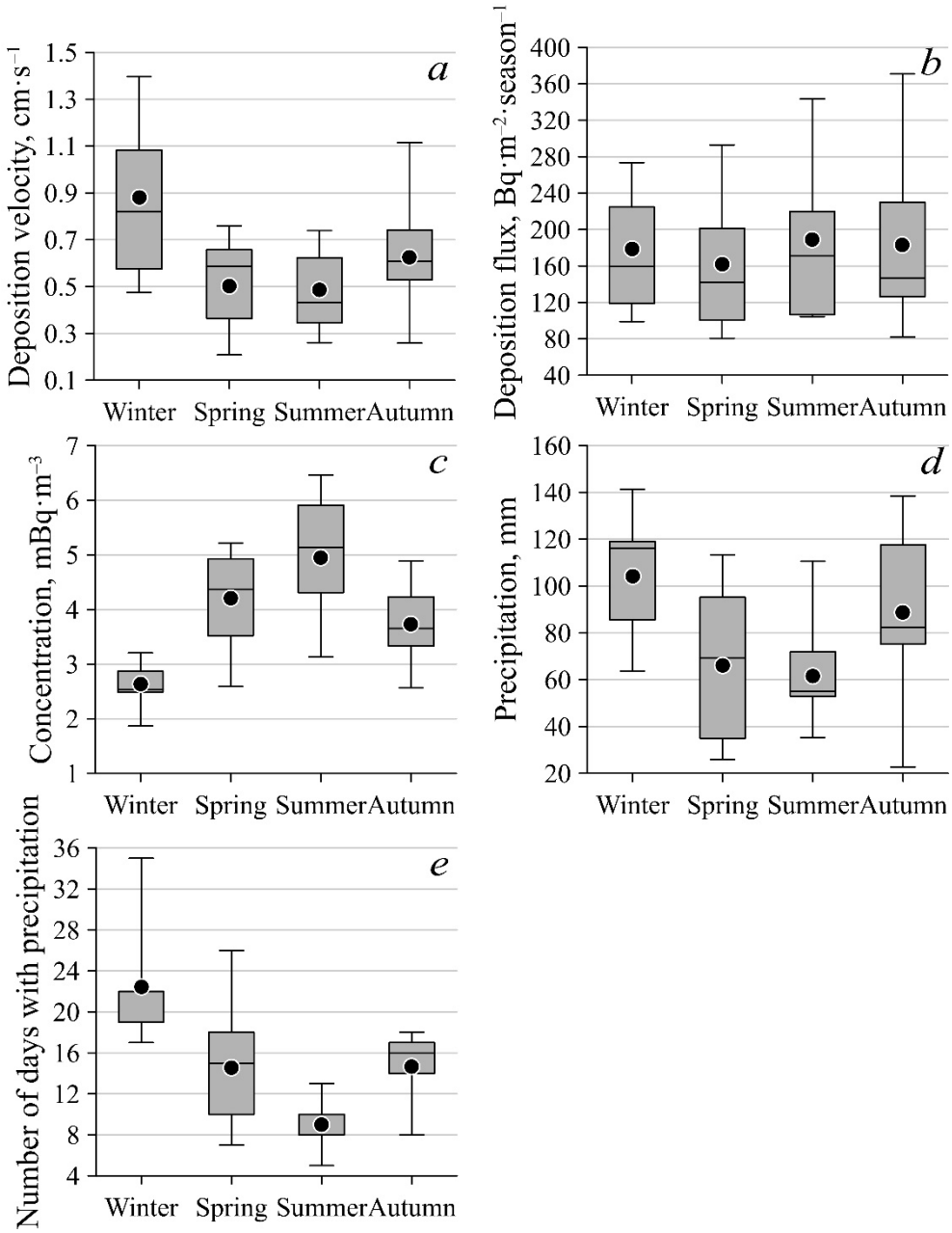


F i g. 2. Frequency distribution of the ^{7}Be flux (*a*) and concentration (*b*) data

Interannual variability of the wet deposition velocity. The season-averaged rate of ^{7}Be washout from the atmosphere varied over time within the range of 0.21–1.40 cm·s^{-1} and averaged 0.62 ± 0.29 cm·s^{-1}. The estimates obtained do not contradict the published data: 0.78 cm·s^{-1} in Brisbane, Australia [29]; 0.5 cm·s^{-1} in Huelva, Spain [30]; 0.5 cm·s^{-1} in Thessaloniki, Greece [17].

The interannual variability of the ^{7}Be wet deposition velocity, averaged over the entire observation period, is shown in Fig. 3, *a*. A pronounced seasonal variability is observed with lower values in the spring and summer seasons (0.50 and 0.49 cm·s^{-1}, respectively) and a maximum value in the winter season (0.88 cm·s^{-1}).

Multiple regression analysis was carried out to obtain quantitative estimates of the ^{7}Be flux and concentration influence on the seasonal variability of the wet deposition velocity. The analysis results show that the seasonal variability in the wet deposition velocity is determined by 69% of the flux variation and by 31% by the concentration variation. It should be noted that seasonal variability is absent in the time series of the ^{7}Be flux (Fig. 3, *b*), but is present in the series of its concentration (Fig. 3, *c*) with a maximum in the summer season (4.9 mBq·m^{-3}) and a minimum in winter (2.6 mBq·m^{-3}).



F i g. 3. Averaged over the whole observation period data on seasonal variability of the ^{7}Be wet deposition velocity, cm·s^{-1} (*a*), the ^{7}Be wet deposition flux, Bq·m^{-2}·season^{-1} (*b*), the ^{7}Be concentration in the atmosphere, mBq·m^{-3} (*c*), precipitation amount, mm·season^{-1} (*d*) and frequency, days (*e*)

According to the correlation analysis results (Table 1), the ^{7}Be flux is statistically significant at the 95% confidence level associated only with the precipitation amount (r = 0.64). There is no connection with the precipitation frequency and the ^{7}Be concentration in the atmosphere. Thus, on a seasonal time

scale, the more precipitation falls, the higher the ^{7}Be flux is. The correlation analysis results show that the seasonal variability of the ^{7}Be concentration in the atmosphere is affected by both the amount and the frequency of precipitation. An increase in the precipitation amount and frequency reduces the season-averaged ^{7}Be concentration in the atmosphere. Based on the absolute values of the correlation coefficients, it can be concluded that on a seasonal time scale, the precipitation distribution during the season under consideration is more important than the precipitation amount (correlation coefficients –0.51 and –0.38, respectively). It should also be noted that the amount and frequency of precipitation are interconnected: seasons with an increased amount of precipitation are characterized by an increased frequency (r = 0.61).

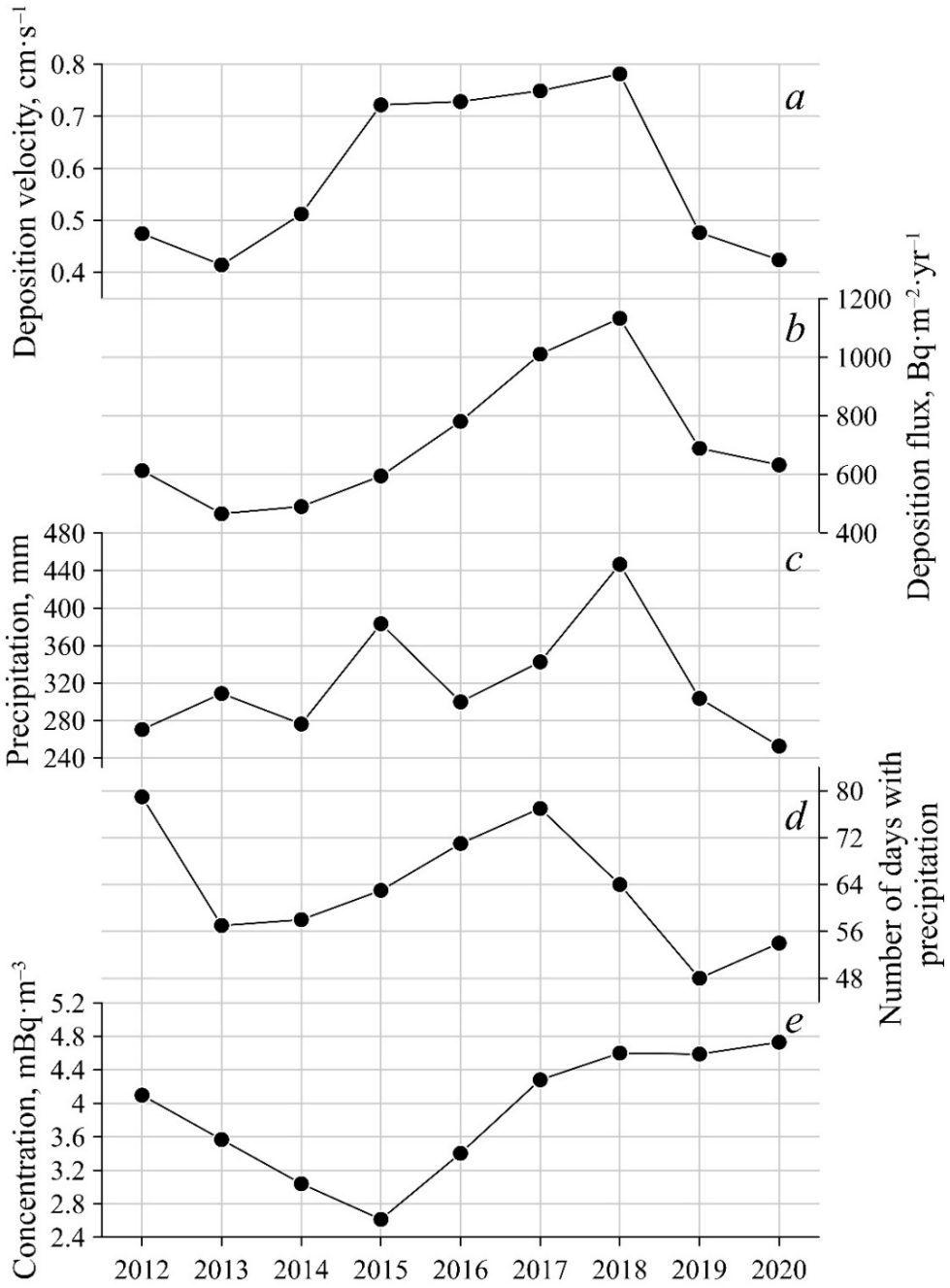
T a b l e 1

Pearson paired correlation coefficients between the seasonal values of the studied parameters

Parameter	F	Pr_a	Pr_f	C_a	V_d
F	1.0	0.64 $p < 0.01$	0.29 $p = 0.09$	0.26 $p = 0.12$	0.69 $p < 0.01$
Pr_a	–	1.0	0.61 $p < 0.01$	–0.38 $p = 0.02$	0.84 $p < 0.01$
Pr_f	–	–	1.0	–0.51 $p < 0.01$	0.60 $p < 0.01$
C_a	–	–	–	1.0	–0.46 $p < 0.01$
V_d	–	–	–	–	1.0

The correlation analysis results show that the strongest relationship is observed between the wet deposition velocity and the precipitation amount (r = 0.84): the precipitation amount growth increases the wet deposition velocity. An increase in the frequency of precipitation has a similar but lesser effect on the seasonal variability of the wet deposition velocity (r = 0.60).

Interannual variability of the wet deposition velocity. The long-term variability of the parameters considered in the work is shown in Fig. 4. The mean annual wet deposition velocity varied in the range of 0.41–0.78 cm·s^{-1} and averaged 0.59 ± 0.15 cm·s^{-1}. Increased values (0.72–0.78 cm·s^{-1}) were noted in 2015–2018, lower values (0.41–0.51 cm·s^{-1}) in 2012–2014 and in 2019–2020 (0.42–0.48 cm·s^{-1}). In 2012–2013 there was a decrease in the ^{7}Be flux from 613 to 466 Bq·m^{-2}·yr^{-1}, further in 2014–2018 there was an increase in the annual total flux to 1133 Bq·m^{-2}·yr^{-1}, after which, in 2019–2020, it was followed by a decrease to 633 Bq·m^{-2}·yr^{-1}. In 2012–2015 the ^{7}Be concentration in the atmosphere decreased from 4.1 to 2.3 mBq·m^{-3}, from 2016 to 2020 its growth from 3.4 to 4.7 mBq·m^{-3} was noted.



F i g. 4. Interannual variability of the ^{7}Be deposition velocity (*a*), the ^{7}Be wet deposition flux (*b*), precipitation amount (*c*) and frequency (*d*), and the ^{7}Be concentration in the atmosphere (*e*)

According to the correlation analysis results (Table 2), the interannual variability of the mean annual values of the ^{7}Be wet deposition velocity is determined by the temporal variability of its flux (r = 0.75). The temporal

variability of the ^{7}Be flux depends on the precipitation amount variability (r = 0.67). Thus, the interannual variability of mean annual values is controlled by the precipitation amount (r = 0.76): an increase in the annual amount of atmospheric precipitation leads both to an increase in the total value of the ^{7}Be flux over the year and to an increase in the annually averaged value of the ^{7}Be deposition velocity. Note that, according to the correlation analysis results (Table 2), there is no statistically significant correlation at the 95% confidence level between the interannual variability of the wet deposition velocity and the ^{7}Be concentration in the atmosphere. It is also worth noting that the interannual variability in the precipitation amount and frequency does not have a statistically significant effect at the 95% confidence level on the interannual variability in the ^{7}Be concentration in the atmosphere.

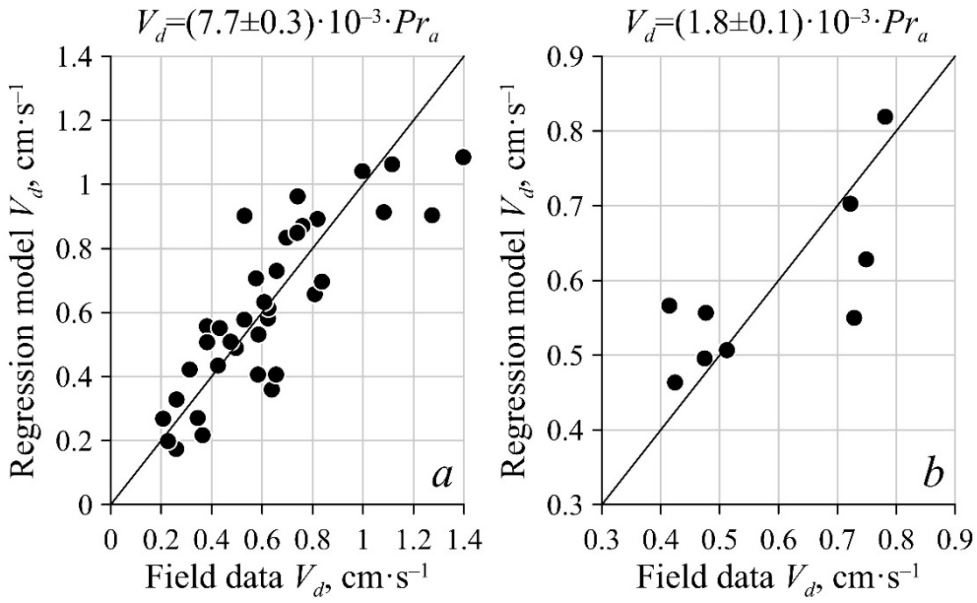
Table 2

Pearson paired correlation coefficients between the annual values of the studied parameters *

Parameter	F	Pr_a	Pr_f	C_a	V_d
F	1.0	0.67 p = 0.05	0.38 p = 0.32	0.51 p = 0.16	0.75 p = 0.02
Pr_a	–	1.0	0.12 p = 0.76	–0.03 p = 0.95	0.76 p = 0.02
Pr_f	–	–	1.0	–0.10 p = 0.80	0.49 p = 0.18
C_a	–	–	–	1.0	–0.17 p = 0.66
V_d	–	–	–	–	1.0

* See designations in Table 1.

Regression Model. Based on the results of multiple regression analysis, regression models are proposed. They allow to calculate the seasonal and interannual variability in the ^{7}Be wet deposition velocity. Taking into account the relationship between the parameters considered in the work (Tables 1 and 2), only the corresponding time series of precipitation amount are used as a predictor of the model of seasonal and interannual variability of the precipitation deposition velocity. Fig. 5 shows the results of a comparison of the wet deposition velocity values calculated from the regression models with those obtained from field data. The mean relative errors of the resulting estimates were 21.1 and 12.9% for the seasonal and annual values of the washout rate, respectively.



F i g. 5. Comparison of the seasonal (*a*) and annual (*b*) values of the ^{7}Be deposition velocity calculated using the corresponding regression model, with those resulted from the field data. Model equations are given at the fragments. The dots show the calculation results, the solid lines – the cases of full compliance of the calculated values with field data

Conclusions

According to the results of the study of seasonal and interannual variability of the aerosol deposition velocity in 2012–2020 the following conclusions were made.

1. In the Sevastopol region, there is seasonal and interannual variability in the aerosol wet deposition velocity. Seasonal values varied in the range of 0.21–1.40 cm·s^{-1}. Decreased values are typical for spring and summer, and increased values are typical for winter. The annual values varied within a narrower range of 0.41–0.78 cm·s^{-1}. Increased values were noted in 2015–2018, and decreased values were noted in other years. The mean annual deposition velocity, averaged over the entire observation period, was 0.59 ± 0.15 cm·s^{-1}.

2. The correlation analysis results show that seasonal variations in the wet deposition velocity are determined by the amount and frequency of precipitation, and the interannual variability of this parameter depends only on the precipitation amount.

3. According to the analysis results, two regression models are proposed. One model enables to describe the seasonal variability of the wet deposition velocity, while the other model describes the interannual variability of this parameter. Both models use the respective time series of rainfall variability as predictors. According to the validation results, the errors of the obtained estimates are 21.1 and 12.9% for seasonal and annual washout rates, respectively.

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