Long-term trends in the mass concentration of near-surface aerosol at Zvenigorod Scientific Station of the A.M. Obukhov Institute of Atmospheric Physics RAS from 1991-2019 measurements

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Abstract. Estimates of seasonally dependent trends in the mass concentration of the near-surface aerosol are presented on the base of results of 29-year measurements at the Zvenigorod Scientific Station of the A. M. Obukhov Institute of atmospheric physics. The station is located in the woody countryside, 50 km west to Moscow. The multiple regression method is used for the analysis, in which autocorrelation of measurement data on large time scales is taken into account. Two periods are considered: 1991–2002 and 2013–2019. Excluded is the period 2003–2012 when abnormally high aerosol concentrations of unknown origin were often observed. For the periods 1991–2002 and 2013–2018, statistically significant negative trends were obtained for spring and summer seasons, respectively, and for the entire year. The spring trend in logarithm of concentration in 1991–2002 was −2.6% per year, and the annual trend was about −0.7% per year. For 2013–2019 a strong negative trend of about −6% per year was obtained in the summer season, and the annual trend estimate was −2.3% per year. The probable cause of the negative aerosol trend in the spring seasons of 1991–2002 is long-term changes in atmospheric transport. The negative trend in the summer seasons of 2013–2019 is probably due to the negative trend of summer temperature, drying and felling of spruce forest.

1. Introduction
Atmospheric aerosol has a significant impact on climate, air quality and human health. Regular measurements of the near-surface aerosol concentration began in the 1990s at separate sites. Currently, measurements of the mass concentration of aerosol particles (particulate matter) with aerodynamic diameters up to 2.5 and 10 microns (PM$_{2.5}$ and PM$_{10}$, respectively) are mainly performed. In most regions with long-term measurements of mass concentration, its decrease was detected [1–12]. However in some regions an increase was observed [9–12]. A more detail review of trends in near-surface aerosol can be found in [13].

Regular long-term aerosol measurements on the territory of Russia are carried out at the Zvenigorod Scientific Station of the A. M. Obukhov Institute of Atmospheric Physics (since 1991) and at the Aerosol Station of the V. E. Zuev Institute of Atmospheric Optics near Tomsk, West Siberia (since 1997). Results of trend analysis of near-surface aerosol concentration at the two stations are presented in [13–14]. In our work [13] we estimated trends in the mass concentration of aerosol at the
Zvenigorod Station and found a strong negative trend in the summers of 2013–2018, with maximum modulo trend in June. One proposed reason of the summer trend is a gradual cooling of summers observed in the region in the 2010s. Another probable reason is drying and felling of spruce forest.

One of summer months, July, was record cold in 2019 in the Moscow region. If the proposed association of the summer aerosol trend with the summer cooling is true, then additional year with cold summer could contribute to the trend. Consequently the subject of this work is the analysis of aerosol trends at the Zvenigorod Station for the extended period. The station is located in the woody countryside, 50 km west to Moscow.

2. Data
Regular measurements of the mass concentration of near-surface aerosol at the Zvenigorod Station started in 1991. Measurements were done by different instruments: by an original flow nephelometer in 1991–2003, by an original spectropolarimeter in 2001–2012, and, since 2012 to the present, by a commercially available domestic photoelectrical nephelometer named FAN. Measurements by the nephelometer were round-the-clock, while measurements after 2003 were done only around 9 h in the morning.

The mass concentration of aerosol is estimated by multiplying the coefficient of the directed light scattering at 45º angle by a scale coefficient. The value of the coefficient was determined using results of concurrent measurements of the PM$_{2.5}$ concentration at the same site. Description of instrument calibration and the method of deriving the mass concentration are described in [13]. Daily values of the aerosol concentration, $M$, and its decimal logarithm are shown in Fig. 1. The period from 2003 to 2012 is characterized by frequent strong anomalies of aerosol. The cause of the anomalies has not yet been established. We exclude this period from further analysis.

The dashed lines in Fig. 1 are linear regressions indicating decreases in the aerosol concentration in 1991–2002 and 2013–2019. However we will use the multiple linear regression method to analyze trends.

3. Method of analysis
The multiple regression model includes the free member, the linear term and the North Atlantic

![Figure 1](https://example.com/figure1.png)

*Figure 1.* (a) Aerosol mass concentration and (b) its decimal logarithm. White dashed lines are linear regression.
Figure 2. Spectra of correlated (blue) and uncorrelated (red) errors for the intervals (a) 1991–2002 and (b) 2013–2019.

Oscillation (NAO) index as predictors (independent variables), and the residual term (error). All regression coefficients in the regression model are expanded into Fourier pairs corresponding to the annual and semi-annual harmonics to account for seasonal dependence of trends.

The residual (error) in the regression problem is often supposed to be like white noise obeying the Gauss distribution. However in practice the residual is not usually consist of random independent values. The residual series is usually correlated. To solve the system of the regression equations we use the method suggested in [15–16], which allows taking into account the serial correlation (autocorrelation) on long time scales. Importance of taking into account the long serial correlation is demonstrated by Fig. 2 that shows power spectra of the correlated and uncorrelated residuals (errors) for the two periods of measurements. The slope (scaling) of the spectra of the correlated error indicates the presence of memory in the residual time series meaning that the values of the residual are not independent from each other. Further, the spectra of the correlated errors have maxima in the vicinity of the 2.5-year period.

According to [17], accounting for the serial correlation allows getting corrections to regression coefficients and their confidence intervals, and an uncorrelated error can be determined that is associated with a correlated error. The spectra of the uncorrelated errors in Fig. 2 have zero slopes peculiar to spectra of white noise and do not contain maxima at the quasi-biennial period. See [13] for detail information about parameters of uncorrelated errors.

4. Results and their discussion
Conventional method of estimating confidence of regression coefficients assumes Gaussian distribution of the residual. Probability distribution of aerosol concentration follows lognormal low. Therefore if the regression model is applied to the concentration of aerosol, then the distribution of the residual should differ from normal. This is demonstrated by Fig. 3a, b showing probability distributions of the residual. The accounting for the autocorrelation does not affect significantly the form of the uncorrelated error distribution which remains far from the Gaussian.

If the multiregression model is applied to the logarithm of the aerosol concentration then the distribution of the residual follows the Gauss distribution well (Fig. 3c, d). For this reason, we prefer the analysis of the logarithm of concentration. It is more robust and, according to [13], gives estimates of aerosol trends that are less (modulo) than trends estimated from analysis of the aerosol concentration.

The annual, monthly and seasonal estimates of the linear trends in the aerosol concentration logarithm for the two periods are shown in Fig. 4. The trend units are percent change per year. Percentage value is calculated relative to the multiyear annual mean of the concentration logarithm for the appropriate time interval. Units on the right vertical axis are the annual change in the aerosol concentration associated with the linear trend in the concentration logarithm. Note that this change cannot be linearly extrapolated for periods longer than one year.
Figure 3. Histograms of correlated (red) and uncorrelated (blue) errors in the regression representation of (a, b) aerosol concentration and its logarithm (c, d) in the intervals (a, c) 1991-2002 and (b, d) 2013-2019. Black curves are approximating Gaussians.

Figure 4 shows that the aerosol trends have a strongly pronounced seasonal dependence. In general, the decrease in the aerosol concentration logarithm (and in the concentration itself as well) dominates. The annual estimates of the trends are negative and amounted to \(-0.7\%\) per year in 1991–2002 and \(-2.3\%\) per year in 2013–2019. In winter and autumn the trends are usually small and statistically insignificant.

Negative, statistically significant trends are observed in the spring seasons in 1991–2002 and in period from late spring to early autumn in 2013–2019. The maximum modulo trend in 1991–2002 is \(-2.6\%\) per year. The associated annual change in the aerosol concentration is \(-1\) mcg·m\(^{-3}\). Note that the strongest negative trend for this period obtained in [13] falls on April as well.

A strong negative trend exceeding modulo 5% per year is observed in the summer seasons in 2013–2019. The maximum (modulo) trend, approaching \(-6\%\) per year, falls on July. The associated annual change in the aerosol concentration is \(-1.4\) mcg·m\(^{-3}\).

Negative trends in near-surface aerosol at the Zvenigorod Station fit into the general picture of aerosol decrease in most geographical regions (see introduction). Comparing our trend estimates for 1991–2002 and 2013–2019, we note an increase in the rate of the aerosol decrease in 2013–2019. Difference of the annual trend estimates is as much as three times the value. Difference of the extreme values of the trends (in spring and summer, respectively) is also big in percentage units. In absolute values, the trends for the two periods are of the same order, but the trends for 2013–2018 are noticeably stronger.

An important common feature of the trends for the two periods is that they occur predominantly in certain seasons. Consider the possible causes of the trends. Change in the emission of aerosol particles and their precursor gases, which affects the concentration of aerosol in West Europe [5, 7–8], does not appear to affect aerosol trends at the Zvenigorod Station. Otherwise, one would expect trends in winter. Perhaps a decrease in the emission due to industrial degradation is roughly balanced by their increase due to motor transport.

Another likely cause of the trends is climate change acting through changes in atmospheric transport and environmental parameters. Long-term changes in the transport require special analysis.
Figure 4. Annual (dots in the left parts of plots), monthly (dots and curves in the middle parts of plots) and seasonal (dots and curves in the right parts of plots) estimates of the linear trend in the aerosol concentration logarithm and their 95% confidence intervals for periods (a) 1991–2002 and (b) 2013–2019 (black) and 2013–2018 (gray). Estimates for 2013–2018 are slightly shifted along horizontal axis for better distinguishing the two set of estimates. See text for explanation of the right vertical axis.

which is not carried out here. Another important natural factor influencing the aerosol content is temperature. In particular, a long-term trend in spring temperature can affect the melting of the snow cover and, as a consequence, could affect the influx of aerosol of soil origin into the atmosphere. A temperature trend during the growing season could modulate concentration of biogenic aerosol. An additional factor affecting the biogenic component of aerosol in the Moscow region may be the drying of spruce forests. According to our observations, the drying in the vicinity of Zvenigorod was the strongest in the 2000s and 2010s.

Figure 5 shows spring mean and summer mean near-surface temperatures according to local data of routine rawinsonde sounding near Moscow and to ERA-Interim reanalysis data averaged over the region around Zvenigorod with 5°-longitude 4°-latitude widths.

The spring temperature trend for 1991–2002 is zero according to the aerological data and weak positive (statistically insignificant) according to the reanalysis data. If this warming was actual and if, as a result, the snow cover and the beginning of the growing season shifted to earlier dates, then we should expect increase but not decrease in aerosol. We conclude that the interpretation of aerosol trends in the 1990s requires analysis of atmospheric circulation changes. Both the aerological and ERA-Interim data show the negative summer temperature trend of about 0.2°C·year⁻¹ in 2013–2019 while there is no any trend in the spring seasons during 1991–2002 (Fig.5). It was found that the emission of biogenic volatile organic compounds that are important precursors of biogenic aerosol in summer is highly sensitive to temperature in the range of summer temperature values [18–19]. Therefore the cooling of summers in 2013–2019 could lead to a decrease in the biogenic component of the near-surface aerosol. An additional contribution to the aerosol trend
Figure 5. Seasonal mean daytime (12 hour UT) temperature for spring (green) and summer (red) derived from radiosonde sounding and diurnally mean summer mean temperature in the Moscow region derived from ERA-Interim (blue). Dashed lines are linear trends for 1991–2002 and 2013–2019.

may be associated with the drying and felling of spruce forests, which, as part of the coniferous forests, are a source of monoterpenes [20].

Hypothesis about the temperature reason of the summer aerosol trend in the 2010s is confirmed by comparison of the aerosol trends estimated for periods 2013–2019 and 2013–2018 (Fig. 4b). In the summer of 2019, July was especially cold, and the trend extremum in Fig. 4b shifts from June to July after addition data for 2019 to the data for the period 2003–2018. Therefore the negative aerosol trend in the 2010s may be, at least in part, due to the summer cooling and the death of spruce forests in the Moscow region.

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