RELATIONS OF THE LONG-TERM VARIABILITY OF THE NEAR SURFACE OZONE CONCENTRATION TO THE SOLAR ACTIVITY AND GLOBAL ATMOSPHERIC CIRCULATION ACCORDING TO DATA FROM EUROPEAN OZONOMETRIC STATIONS

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Time series of monthly mean surface ozone concentration obtained at the European ozonometric stations are analyzed in relation to the series of solar activity and the North Atlantic oscillations. The statistically significant relations between them have been revealed. For the ozonometric station in Hohenpeissenberg (Germany), the contributions to the long-term total variability of the surface ozone concentration from the trend, the North Atlantic oscillations, and the solar activity are about 0.16, 0.11, *and* 0.015, *respectively; the autoregression contribution adds* 0.18 *more.*

It is well known[1–3] that temporal behavior of the total ozone content (TOC) and the vertical distribution of ozone (VDO) over an observation site is qualitatively determined, to a noticeable extent, by numerous predictors. Among these predictors there are the regional ones (temperature and geopotential at certain levels, content of other trace gases and aerosols, etc.) and the global (quasiperiodic) perturbations. The global perturbations extend their influence for many thousands of kilometers. They are related to the solar activity, quasi-two-year oscillations (hereinafter, QTO) of the zonal circulation in the equatorial stratosphere, and parameters of the climate-forming centers of the atmospheric activity (in particular, the North Atlantic oscillation[4,5]).

Such a separation of the predictors into regional and global ones, though being conditional and simplified, reflects significant differences in the spatial and temporal scales of the forcings under study. The surface ozone concentration (SOC) is very sensitive to local influences, such as emissions of trace gases and aerosols from local sources and to meteorological situations in the surface layer. However, it is reasonable to expect that the time series of SOC is also dependent on global oscillations, which appreciably determine the global atmospheric circulation.

From analysis of such a dependence, we can find additional information about the nature of the surface ozone and its variability (for example, relation between the surface and stratospheric ozone). At the same time, such relations are practically unstudied. Only the studies of time series of the tropospheric ozone based on data acquired with the balloonborne ozonesondes are known.[6] However, the quality of these measurements and the completeness of the time series are far from being perfect.

The regional predictors are related to the high-frequency (the frequency above 1 month^{-1}) SOC variability, while the global ones are related to the low-frequency variability. The effect of the regional predictors (in particular, temperature, humidity, and others) on the SOC has been discussed in many papers,[7,8] but the effect of global predictors is not quantitatively understood yet,[5] although their influence on the long-term TOC variability has been studied rather comprehensively.[2,3] In this paper we analyze the time series of SOC obtained at the European ozonometric stations in order to estimate quantitatively the characteristics of SOC relation to the solar activity and to the parameters of global atmospheric circulation. To do this, we apply the approach described in our earlier papers.[5,8–11]

In this paper we deal with the monthly mean values of SOC observed since 1976 to 1995 at the stations of the worldwide ozonometric network.[12] Among the observation data, the time series obtained in Hohenpeissenberg stands out. These series are generally recognized as sufficiently long and complete, and they are of superior importance for analysis of time behavior of the parameters describing the ozone layer. The quality of experimental material collected in the Hohenpeissenberg series is high, and possible errors due to instrumental uncertainties and influence of neighboring anthropogenic sources of atmospheric pollution are sufficiently well analyzed.[13]

The series obtained at the station Neuglobson are also long enough for the analysis of the long-term variability. This station is a part of the BAPMoN network (the WMO network) for the background monitoring of air pollution. The data obtained at other stations,[12] including all non-European stations, are unfortunately either insufficiently long (shorter

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than 10 years) or discontinued (for example, Sibton). These data were used only in calculation of the parameters of the SOC annual behavior. (Note that the time behavior of SOC recorded at the high-mountain stations, for example, German ones – Zugspitze and Wank, – is characteristic of the free troposphere. It significantly differs from the time behavior of SOC recorded at stations in plain lands,[14] in particular, in diurnal variation. Besides, these series are not long enough. That is why we did not use them in our analysis). Note also that failure to detect statistically significant relations may be indicative of their low significance (as compared to measurement errors and other factors), rather than their absence. On the contrary, their detection with some confidence level is an evidence of the existence of such relations.

As a model for description of the time series of SOC $C(m)$, we have taken the model, which is usually used to describe time behavior of TOC and VDO:

$$C(m) = Annual(m) + Trend\ m +$$
$$+ \Sigma\ (k_i\ Regr_i(m)) + R(m), \qquad (1)$$

where m is the running number of a month starting from January of 1976 ($m = 1$); $Annual(m) = C_0 + \Sigma\ [A_i \cos(2\pi\, i\, m/12) + B \sin(2\pi\, i\, m/12)]$ is the normal annual behavior (three annual harmonics is enough for its good description[4]); $Trend$ is the linear trend; $k_i\ Regr_i(m)$ is the term describing the influence of the ith predictor quantitatively characterized by the value of $Regr_i(m)$; k_i is the corresponding coefficient; $R(m)$ is the remainder, which can be additionally presented as an autoregression series of the first order:

$$R(m) = AR(1)\ R(m - 1) + Noise(m),$$

where $AR(1)$ is the coefficient of autoregression of the first order (the order of autoregression is caused mainly by the duration of the synoptic cycle, which is about one week in midlatitudes of the Northern hemisphere; deviations of SOC from the climatic norm hold during such a cycle, hence the autoregression coefficient must be from 0.2 to 0.4); $Noise(m)$ is the white noise.

The index $F_{10.7}$, of solar activity, the characteristics of QTO: monthly average values of the equatorial wind in Singapore at the level of 30 hPa (with regard for the phase, as described in Ref. 1), the index of the southern oscillation describing the El-Ninio/Southern oscillation[2] phenomenon, and parameters of the North Atlantic oscillation[4,5] have been tested as possible predictors. The atmospheric pressure at the centers of the Azores anticyclone and the Iceland cyclone, the coordinates of these centers, differences in the pressure and in the corresponding coordinates, as well as the index of the North Atlantic oscillation (the normalized difference in atmospheric pressure at fixed points – on the Azores and in Iceland) were used as the parameters of the North Atlantic oscillation (NAO).

The QTO characteristics and the index of the Southern oscillation turned out to be statistically insignificant predictors (with the confidence level $P = 0.95$). For this reason they were excluded from further calculations. On the contrary, it turned out that SOC in Hohenpeissenberg could be presented in the form of regression series over practically all the above listed parameters of the north Atlantic oscillation. The regression coefficients at the parameters of the Iceland cyclone and their statistical significance have larger weights than the corresponding coefficients at the parameters of the Azores anticyclone. For further calculations we have selected the parameters, which are most efficient predictors having clear physical meaning. These parameters are the following: differences $P_{\mathrm{AI}}(m)$ between the atmospheric pressure at the centers of the Azores anticyclone and the Iceland cyclone (characterizing the intensity of the oscillation) and between the latitudes of these centers $\psi_{\mathrm{AI}}(m)$ (characterizing the “directionalityB of the oscillation). The time behavior of these predictors is shown in Fig. 1. They (especially, the pressure difference) experience statistically significant long-term variations. It is likely, that these are just the factors that determine (at least, partially) a part of the observed SOC trend.

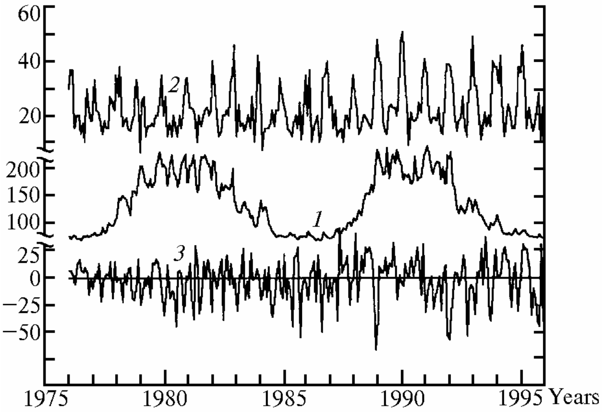

FIG. 1. Time behavior of different Parameters used in the regression analysis: the index of solar activity $F_{10.7}$ *(1); the difference in the atmosPheric Pressure at the sea level in the centers of the Azores anticyclone and the Iceland cyclone* $P_{\mathrm{AI}}(m)$, *in* hPa *(2); the difference between the latitudes of the centers of the Azores anticyclone and the Iceland cyclone* $\psi_{\mathrm{AI}}(m)$ *(3).*

Since the parameters of the North Atlantic oscillation themselves experience annual variability (it is well seen in Fig. 1, especially for the pressure difference), their deviations from the estimated normal annual behavior were used as predictors. The deviations were calculated using the method of least squares:

$$\Delta P_{\mathrm{AI}}(m) = P_{\mathrm{AI}}(m) - C_p - \Sigma\,[A \cos(2\pi\, i\, m/12) +$$
$$+ B \sin(2\pi\, i\, m/12)];$$
$$\Delta\psi_{\mathrm{AI}}(m) = \psi_{\mathrm{AI}}(m) - C_\psi - \Sigma\,[A \cos(2\pi\, i\, m/12) +$$
$$+ B \sin(2\pi\, i\, m/12)].$$

The designations here are similar to those in Eq. (1). The values of $p_{\mathrm{AI}}(m)$ and $\psi_{\mathrm{AI}}(m)$ averaged over the 1958–1995 period are 21.8 hPa and −0.3°, respectively; and their rms deviations are, respectively, 8 hPa and 21.3°, while the rms deviations of $\Delta p_{\mathrm{AI}}(m)$ and $\Delta\psi_{\mathrm{AI}}(m)$ are, respectively, −6.3 hPa and 20.7°.

The numerical characteristic of the effect of each factor (or their group), namely its (their) part in the total variability, can be estimated as follows[7,8]:

$$Q = 1 - \sigma_2^2/\sigma_1^2,$$

where σ_2^2 and σ_1^2 are the variance of the series after application of the regression expansion to a given factor (group of factors) and that of the initial series, respectively. The normal annual behavior of SOC at the European ozonometric stations is well described by the expansion over the first three harmonics of the annual behavior[11]; the corresponding efficiency Q of separation of the normal annual behavior is about 0.8 and higher. The results of determination of the model parameters (along with the corresponding errors calculated for the confidence level $P = 0.95$) for the time series of SOC at different stations are presented in Table I. Dashes in the Table stand for statistically insignificant values. The Table contains, in addition to the above-mentioned parameters, the rms deviations for the series $C(m) - \sigma_0$, $C(m) - Annual(m) - \sigma_r$, and $Noise(m) - \sigma_n$.

TABLE I. Parameters of models for the time series of monthly mean SOC.

Parameter	Station					
	Hohenpeissenberg	Arkona	Neuglobson	Sibton	Preila	Dolgoprudny
	Country					
	Germany			Gr. Britain	Lithuania	Russia
N	47°48′	54°41′	53°09′	51°30′	55°20′	55°45′
E	11°01′	13°26′	13°02′	00°07′	21°13′	37°34′
Period	1976–1995	1988–1990	1978–1991	1987–1989	1987–1994	1991–1996
C_0, mPa	3.07 ± 0.10	1.56 ± 0.68	1.4 ± 0.14	2.08 ± 0.33	2.08 ± 0.16	1.99 ± 0.13
σ_0, mPa	0.936	0.931	0.771	0.852	0.912	1.002
A_1, mPa	−1.20 ± 0.07	−1.22 ± 0.19	−0.78 ± 0.10	−1.09 ± 0.15	−1.16 ± 0.12	−1.33 ± 0.19
B_1, mPa	−0.04 ± 0.07	−0.05 ± 0.19	0.20 ± 0.10	0.30 ± 0.16	−0.22 ± 0.12	0.14 ± 0.19
A_2, mPa	−0.03 ± 0.07	0.01 ± 0.19	−0.05 ± 0.10	−0.10 ± 0.15	–	–
B_2, mPa	0.12 ± 0.07	−0.20 ± 0.19	−0.06 ± 0.10	−0.21 ± 0.16	–	–
A_3, mPa	0.14 ± 0.07	–	0.10 ± 0.10	–	0.14 ± 0.12	–
B_3, mPa	0.03 ± 0.07	–	0.06 ± 0.10	–	0.02 ± 0.12	–
σ_r, mPa	0.358	0.387	0.452	0.299	0.374	0.384
Trend, mPa·year^{-1}	0.029 ± 0.009		0.064 ± 0.019		0.069 ± 0.033	
$k_{F10.7}\cdot10^{22}$, mPa·W^{-1}·m^2·Hz	(9.2 ± 8.8)·10^{-4}		(12 ± 13)·10^{-4}		(19 ± 15)·10^{-4}	
$k_{\Delta p\mathrm{AI}}$, mPa/hPa	0.080 ± 0.53		–		–	
$k_{\Delta\psi\mathrm{AI}}$, mPa/deg	0.047 ± 0.024		–		–	
$AR(1)$	0.033 ± 0.016		0.043 ± 0.014		0.021 ± 0.020	
σ_n, mPa	0.338		0.394		0.347	

Notes: 1. All errors are given for the 95-% confidence interval (at the level ± 2σ). 2. Dashes stand for statistically insignificant values. 3. Empty cells mean that the corresponding parameters were not calculated.

The efficiency of separating trend, the parameters of the North Atlantic oscillation, and those of solar activity from the time series of SOC remainders in Hohenpeissenberg, for the period from 1976 to 1995, is 0.16, 0.12, and 0.015, respectively. The efficiency of joint separation of the trend and all of the above-indicated parameters is about 0.3.

To confirm the constant character of the described effects, the calculations were done not only for theentire period of observations in Hohenpeissenberg (1976–1995), but for the first and second halves of this period, separately. The results turned out to be similar, especially in the quantitative estimation of the NAO parameters. The initial time series of SOC and some of those obtained from analysis are shown in Fig. 2. Note that the remainder $R(m)$, the temporal behavior of which is described by curve *3* in Fig. 2, can likely be additionally presented as an expansion over the local predictors (in particular, the surface temperature and humidity[10]). This must result in additional, possibly, twofold decrease in the variance of the remainder time series.[7,10] However, this expansion requires invoking of the time series of the above-indicated meteorological parameters, what is beyond the scope of this paper.

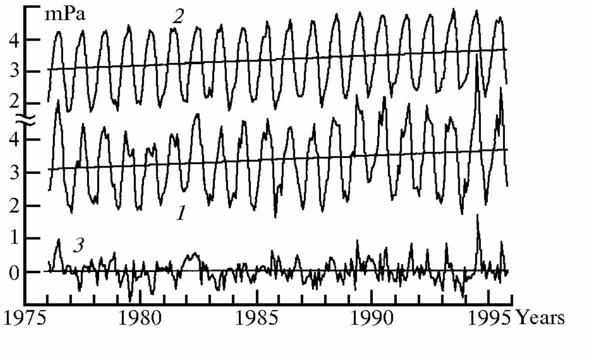

FIG. 2. TemPoral behavior of the monthly mean SOC, in mPa*, in HohenPeissenberg: the observed data (curve 1); the regression Presentation by EC. (1) (curve 2); the remainder, i.e. the difference between the curves 1 and 2 (curve 3). The straight lines along the curves 1 and 2 corresPond to the sum* $(C_0 + Trend\ m)$ *from EC. (1).*

The results obtained in this paper are indicative of the following:

1) There exists positive trend of the surface ozone in Europe for the entire period of regular measurements since the 1970s by about 1% per year, what confirms the results of earlier studies.[3,15]

2) The solar activity probably ($P = 0.95$) influences the surface ozone, and this influence is significant at practically all European ozonometric stations. This result confirms the results from Ref. 5.

3) The North Atlantic oscillation has a significant influence on the surface ozone in Europe, as well as on TOC.[16] (This influence may be caused, in particular, by NAO effect on the weather conditions there.[4]) This influence should be taken into account in quantitative description of the temporal behavior of the surface ozone concentration in Europe, because the NAO parameters experience climatic variations,[16] that may certainly be responsible for a part of the observed trend.

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