Interrelation between Dynamics of Gas Composition and Meteorological Parameters in the Region of Tomsk

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Abstract—In this paper, we studied the interrelation between the variations in CH_4, CO, CO_2, NO, NO_2, O_3, and SO_2 concentrations, and the number concentration of aerosol with particle diameters larger than $0.4\ \mu m$, and the following meteorological parameters: air temperature, atmospheric pressure, wind direction and speed, total solar radiation and ultraviolet radiation in the wavelength range of 295–320 nm, relative humidity, and partial water vapor pressure. For this, we used data from monitoring the air composition (for the period 1993–2018) performed at the Tropospheric Ozone Research (TOR) station in the region of Tomsk Akademgorodok.

INTRODUCTION

The present-day global climate change, largely due to growing anthropogenic emissions [1], may, through feedback from the climate system, cause an increase in the concentrations of gas and aerosol constituents in air of not only industrial but also background regions [2–9]. Therefore, quite a few works have been devoted to studying the relationship between the content of certain compounds in the atmosphere and changes in meteorological parameters.

In work [10] it is shown that rising air temperature increases the productivity of forest vegetation, leading to assimilation of a greater amount of CO_2 and a greater emission of volatile organic compounds. Karol and Reshetnikov note that the concentrations of all greenhouse gases grow in the warming climate [11]. The authors of work [12] indicate that soil moisture, and not only air temperature, influences the efficiency of uptake of carbon dioxide by plants. In work [13] it is recorded that rising air temperature triggers the intensification of photosynthesis and, hence, reduces the CO_2 concentration. Relying upon a phenomenological approach, the authors of work [14] showed that the rising air temperature extends the growing season and, thereby, increases the assimilation of carbon dioxide. However, based on these data, the authors of [15] found that the increase in the seasonal amplitude of the CO_2 concentration slowed down with increasing temperature. An even stronger effect was revealed in work [16], where it is shown that at very high temperatures photosynthesis shuts down, leading to increase in the concentration of carbon dioxide. The authors of work [17] note the considerable interannual variations in the CO_2 concentration, depending on the changes in meteorological parameters. In addition to carbon dioxide, the concentrations of other gas and aerosol constituents of air were found to depend on meteorological parameters. This is demonstrated by the examples of ozone [18–20], methane [21, 22], nitrogen oxides [23–25], carbon monoxide [26, 27], and sulfur dioxide [28]. Moreover, there are publications where the effect of meteorological conditions is studied for a few gas admixtures [29–33].

All the abovementioned works, reflecting the effect of meteorological conditions on the air composition, relied upon the results from special experiments or short-term measurements. At the same time, multiyear studies of the air composition, of which there are many [34–45], usually disregard the effect of time variations of meteorological parameters. Therefore, the purpose of this work is to analyze the relationship between variations in concentrations of atmospheric

Fig. 1. Multiyear behaviors of O_3, CO_2, SO_2 concentrations and meteorological parameters in the region of Tomsk.

admixtures and meteorological parameters in the multiyear context.

INITIAL DATA

The study was carried out using data from monitoring air composition, obtained from 1993 to 2018 at the Tropospheric Ozone Research (TOR) station, located in the region of Tomsk Akademgorodok [46]. The measurement complex during this time has been constantly modernized to extend the parameters monitored. The modern state of the complex was presented in [47].

The analysis was carried out using monthly average values to eliminate short-period weather variations. We studied the interrelations between the concentrations of atmospheric admixtures (CH_4, CO, CO_2, NO, NO_2, O_3, SO_2, and aerosols with particle diameters larger than 0.4 μm) and the following meteorological parameters: air temperature (T), pressure (P), wind speed (V) and direction (d), total solar radiation (SR) and ultraviolet radiation in the wavelength range 295–320 nm (UV−B), relative air humidity (U), and partial water vapor pressure (e).

RESULTS AND DISCUSSION

Comparisons between multiyear behaviors of atmospheric admixture concentrations and meteoro-

logical parameters show that all diversity of the probable relationships can be divided into three groups: almost synchronous behaviors (Fig. 1a), anticorrelation, antiphase variations (Fig. 1b), and independent (or random) behaviors (Fig. 1c). It should be noted that the plots for Fig. 1 were not selected specially; there are several in each group.

The correlation coefficients were calculated to estimate the character and closeness of the interrelation between meteorological parameters and atmospheric constituents; they are presented in Table 1. The observation time series differ in length; therefore, the number of cases analyzed is indicated in the results, presented here.

From data, presented in Table 1, it follows that ozone shows a significant positive or negative correlation with almost all analyzed meteorological parameters, except wind direction. This latter means that either there are no sources of ozone-forming substances, or no conditions for in situ ozone production, in the neighborhood of the TOR station [49]. Similar interrelations for certain meteorological parameters were also recorded in other regions of the planet [50–53]. The positive correlation of O_3 with solar radiation and the ultraviolet part of its spectrum is easily explainable because most of the tropospheric ozone is produced from precursor gases, exposed to UV−B radiation, mainly in the boundary layer [54,

Table 1. Correlation coefficients between monthly average values of meteorological parameters and aerosol/gas concentrations

Meteorological parameter	O_3	CO_2	CH_4	NO_2	SO_2	CO	NO	Aerosols
SR	**0.504**	**−0.461**	*−0.387*	*−0.197*	−0.124	0.046	0.058	*−0.237*
UV−B	**0.305**	**−0.624**	*−0.513*	*−0.163*	−0.164	−0.120	*−0.200*	**−0.337**
T	**0.326**	**−0.626**	**−0.586**	**−0.301**	*−0.230*	−0.032	−0.057	**−0.419**
P	**−0.332**	**0.594**	**0.590**	**0.329**	*0.211*	0.126	−0.057	**0.525**
e	**0.425**	**−0.621**	*−0.387*	*−0.178*	*−0.231*	0.047	−0.081	*−0.309*
U	**−0.591**	*0.245*	*0.421*	0.100	−0.076	−0.110	0.103	0.190
V	**−0.238**	*0.282*	0.051	**−0.301**	*−0.301*	−0.103	−0.184	−0.056
d	0.030	0.103	−0.009	−0.123	*0.271*	0.110	0.000	*−0.316*
N	311	73	30	158	93	206	116	104
$R\ (Q = 0.05)$	0.11	0.24	0.36	0.16	0.21	0.14	0.19	0.20
$R\ (Q = 0.001)$	0.19	0.39	0.57	0.27	0.34	0.23	0.31	0.32

N is the number of cases in the calculation; $R\ (Q = 0.05)$ and $R\ (Q = 0.001)$ are the significance levels of the correlation coefficient for a given N and confidence probability Q, taken from [48]; entries in semibold highlight the coefficients significant at the 0.001 level, and those in italic, at the 0.05 level.

55]. As was shown in [56, 57], air temperature strongly determines the ozone production in the atmosphere, just as reflected in the positive correlation with this parameter. The high correlation coefficient between O_3 and e is, most probably, because the partial water vapor pressure depends strongly on air temperature [58], while the direct cause-and-effect relationship is absent in this case. The anticorrelation between the relative humidity and ozone stems from the fact that, in the wide temperature range (+30 to −30°C), the rising temperature influences negatively the O_3 formation rate in the atmosphere [59]. The significant opposite interrelation between O_3 and V is probably because, the larger the wind speed, the greater the turbulence, leading to more intense dispersal of admixtures over the entire atmospheric boundary layer [60, 61]. The relationship between O_3 and P, as well as the relationships of other constituents, will be addressed a little later in the text since the atmospheric pressure turns out to be correlated significantly with changes in other air constituents.

The high negative correlations between carbon dioxide and solar and UV−B radiation, temperature, and partial water vapor pressure in Table 1 are quite readily explained. With respect to these meteorological parameters, CO_2 shows an opposite annual behavior [62, 63]. When solar radiative influx and, hence, UV−B radiation grow, air temperature and moisture content start to increase, an intense carbon uptake by plants begins during their photosynthesis, and the vertical air mixing in the troposphere intensifies. As a result, the higher the values of the most meteorological parameters, the lower the CO_2 concentration in the near-surface atmospheric layer.

Since the seasonal variations of methane and carbon dioxide concentrations show similar structures [62, 64], this is reflected in the character of interrelation between CH_4 and meteorological parameters; like CO_2, methane is characterized by a significant negative correlation coefficient (Table 1).

In addition to the positive dependence on pressure, nitrogen dioxide is characterized by significant negative correlations with air temperature and wind speed (Table 1). Probably, like for ozone, the anticorrelation with the wind speed can be interpreted by increased dispersal due to turbulence. At the same time, the decrease in the NO_2 concentration with the rising temperature seems to be caused by the participation of nitrogen oxides in the cycles of ozone formation. As is well known [65], the $NO-O_3-NO_2$ triple balance is established in clean air. Independent study of the territory around TOR station indicates that it is under background conditions for most of the time [66, 67].

Like gas-phase air admixtures, atmospheric aerosol is steadily and positively correlated with air pressure and negatively correlated with the total solar radiation, UV−B radiation, air temperature, and partial water vapor pressure. This result seems to be justified considering that the annual behavior of aerosol number concentration ($d > 0.4\ \mu m$) is opposite to the behavior of these meteorological parameters [47, 68].

Meanwhile, the data in Table 1 reveal no significant correlations between meteorological parameters and such gas-phase constituents as SO_2, CO, and NO.

From analysis of the data it follows that for certain species, stable correlations exist between their concentrations and meteorological parameters. Figure 1 indicates that the closeness and sign of correlations are determined mainly by specific features of annual

Fig. 2. Multiyear behavior of atmospheric pressure and CO_2 and O_3 concentrations in the region of Tomsk.

behaviors of admixtures and meteorological parameters. At the same time, pressure evidently has no distinct annual behavior [58]. Therefore, the correlations of O_3, CO_2, CH_4, NO_2, and aerosol number concentration with air pressure, revealed here, have another nature. It is noteworthy that the concentrations of all gas-phase admixtures were reduced to the normal conditions (0°C, 1013 hPa) before processing.

From the synoptic meteorology it is well known that the main changes in the atmospheric pressure are associated with the passage of cyclones and anticyclone, ridges and troughs through the observation site [69]. As a rule, at midlatitudes the cyclones pass as series of 3−5 objects, followed by the concluding anticyclone [70]. Thus, the waves of low and high pressure will alternate at the observation site (Fig. 2).

The concentration of carbon dioxide grows with increasing atmospheric pressure. This is just what is reflected in the positive correlation coefficient. A negative correlation coefficient between ozone and pressure time series is evident in Table 1. From Fig. 2b it can be readily seen that this pair is characterized by almost opposite variations. An increase of pressure leads to decrease of ozone concentration, making it possible to hypothesize that the significant correlations thus revealed are caused by the circulation processes. Earlier [71], we showed that the gas composition is quite homogeneous in each air mass and varies in jumplike fashion in passing from one air mass to another [71]. The occurrence of cyclones on the territory of observations strongly changes the diurnal behavior of the CO_2 amplitude [72]. Some other authors [73−75] who studied how synoptic processes influence the air composition also argue in favor this hypothesis.

To test this hypothesis, we calculated the correlation coefficients between monthly average gas/aerosol concentrations and the frequency of occurrence of cyclones, anticyclones, troughs, and ridges. Data, presented in Table 2, support partially (not for all admixtures) this hypothesis. For instance, ozone is significantly correlated with almost all synoptic objects. The concentrations of NO_2, CO, and aerosols are closely correlated with the total frequency of occurrence of synoptic structures.

Possibly some other characteristic, and not the frequency of occurrence, should be used for analysis because synoptic objects strongly differ both in the pressure difference, and in the size, speed of motion, and place of origin. Nonetheless, even such an approach makes it possible to identify partially the effect of atmospheric circulation on the change of air composition.

All results presented above were obtained from monthly average values, characterized by the presence of annual behavior. Many meteorological parameters and air constituents show similar annual behaviors. Therefore, the effect of this factor should be excluded in analysis. For this, annually averaged parameters will be used.

Data in Table 3 show that interrelations between annually averaged concentrations of gases/aerosols and meteorological parameters are significantly different in character. First, the correlation coefficients have a much lower significance level. The correlation coefficient is significant at the 0.001 level only for NO and solar radiation, probably reflecting NO_2 photolysis, during which nitric oxide is formed [65]. Second, the air composition in the multiyear behavior barely correlates with solar radiation, temperature, and air pressure. It is noteworthy that several gas admixtures (O_3, CO_2, SO_2) and aerosols exhibit the presence of a relationship with water vapor (e, U) and wind speed. The correlation between CO/aerosol concentrations

Table 2. Correlation coefficients between monthly average values of the frequency of occurrence of synoptic situations and gas/aerosol concentrations

Synoptic object	O_3	CO_2	CH_4	NO_2	NO	CO	SO_2	Aerosols
Frequency of occurrence of cyclones	−0.139	0.042	−0.032	0.026	−0.101	−0.018	−0.033	−0.143
Frequency of occurrence of anticyclones	−0.145	0.081	−0.033	0.004	−0.188	0.154	−0.062	0.195
Frequency of occurrence of cyclones and anticyclones in total	**−0.196**	0.106	0.305	0.017	−0.207	0.114	−0.081	0.062
Frequency of occurrence of cyclones and troughs	**−0.160**	0.158	−0.136	0.142	0.109	**−0.245**	−0.012	0.012
Frequency of occurrence of anticyclones and ridges	−0.078	0.012	0.212	0.065	−0.032	0.065	−0.043	−0.021
Frequency of occurrence of cyclones, troughs of anticyclones and ridges in total	**−0.198**	0.157	0.079	0.172	0.057	−0.153	−0.042	**−0.535**
N	311	73	30	158	93	206	116	105
$Q = 0.05$	0.11	0.24	0.36	0.16	0.19	0.14	0.21	0.20
$Q = 0.001$	0.19	0.39	0.57	0.27	0.31	0.23	0.34	0.32

Table 3. Correlation coefficients between annually average values of gas/aerosol concentrations and meteorological parameters

Meteorological quantity	O_3	CO_2	NO_2	SO_2	CO	NO	Aerosols
SR	−0.038	−0.078	−0.203	0.567	−0.120	**0.579**	−0.169
UV–B	−0.257	−0.615	0.106	−0.686	−0.246	−0.080	0.132
T	−0.208	0.426	**−0.483**	0.139	0.154	−0.277	0.004
P	**−0.400**	−0.318	0.173	0.462	0.025	−0.239	−0.303
e	**−0.589**	**−0.656**	0.063	**−0.752**	0.052	−0.073	**−0.415**
U	**−0.447**	**−0.690**	0.190	**−0.837**	−0.136	0.075	**−0.587**
V	**−0.495**	**−0.669**	0.018	**−0.787**	−0.096	−0.153	**−0.432**
d	0.025	0.360	0.061	0.306	**0.449**	−0.189	**0.394**
Frequency of occurrence of cyclones	−0.251	0.266	0.213	−0.398	0.315	−0.287	−0.153
Frequency of occurrence of anticyclones	0.364	0.536	−0.090	−0.340	0.200	0.493	**−0.397**
Frequency of occurrence of cyclones and anticyclones in total	−0.339	0.500	0.022	−0.386	0.270	0.055	−0.314
N	26	10	17	8	21	13	26
$R (Q = 0.05)$	0.39	0.63	0.48	0.71	0.43	0.55	0.39
$R (Q = 0.001)$	0.61	0.87	0.73	0.93	0.67	0.80	0.61

Entries in semibold highlight the correlation coefficients in interval between the 0.05 and 0.001 probabilities.

and wind direction may be because the sources of these constituents are present near the measurement site. The opposite interrelation between NO_2 and air temperature is explained by the nonlinear dependence of the photochemical interaction rate of nitrogen dioxide with other gas-phase air constituents [76].

The multiyear behavior itself exhibits no constant synchronous or asynchronous interrelations, as is evident from Fig. 2.

From the plots in Fig. 3 it can be seen that there are asynchronous variations in ozone/aerosol concentrations and meteorological parameters; however, they do not encompass the whole period analyzed. For instance, we can state that ozone concentration varies in antiphase with the water vapor content, with the change of phases occurring in 1995 and 2001 (Fig. 3a). Aerosol is characterized by shorter transitions between phases. The changes of phases were observed in 1994, 2004, 2012, and 2016 (Fig. 3b).

CONCLUSIONS

The analysis of relationships between concentrations of gas/aerosol constituents of air and meteoro-

Fig. 3. Annually average values: (a) of ozone concentration and absolute humidity; and (b) of aerosol number concentration and relative humidity.

logical parameters, performed using monthly average data, shows that their entire diversity is divided into three groups: almost synchronous variations, anticorrelation, and independent (or random) behavior.

Ozone shows significant positive or negative correlation with all analyzed meteorological parameters, except wind direction. Carbon dioxide is found to anticorrelate strongly with total solar radiation and UV−B radiation, temperature, and partial water vapor pressure. Similar correlations with the same meteorological parameters are also recorded for methane. In addition to its positive dependence on pressure, nitrogen dioxide exhibits two significant negative correlations with air temperature and wind speed. Like for ozone, the anticorrelation with the wind speed can probably be attributed to increased dispersal due to turbulence. Like the gas-phase air admixtures, atmospheric aerosol shows a stable positive correlation with air pressure and negative correlations with solar radiation, UV−B radiation, air temperature, and partial water vapor pressure. No significant correlations are found between meteorological parameters and such constituents as SO_2, CO, and NO.

The closeness and sign of correlations are determined mainly by specific features of annual behaviors of admixtures and meteorological parameters. The found correlations of O_3, CO_2, CH_4, NO_2, and aerosol number concentrations with air pressure have a different nature. They are attributed to circulation processes.

The interrelation between annually averaged gas/aerosol concentrations and meteorological parameters is significantly different in character: lower significance level of the correlation coefficients and fewer interrelated variables. Air composition in the multiyear record is almost uncorrelated with solar radiation, temperature, and air pressure. Many gases show interrelation with water vapor and wind speed. The parameters analyzed exhibit asynchronous variations; however, they do not encompass the entire period analyzed.

Our analysis allows us to conclude that air composition in the multiyear record is determined by large-scale atmospheric processes.

CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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