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Accounting for local meteorological effects in the ozone time-series of Lovozero (Kola Peninsula)

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Abstract

The impact of local meteorological conditions on surface ozone was studied by means of regression models creation. Ozone and meteorological parameters measured at Lovozero site (250 m a.s.l., 68.5° N, 35.0° E, Kola Peninsula) for the period of 1999–2000 were used. The regression model of daily mean ozone concentrations on the meteorological parameters like temperature, relative humidity, and wind speed can explain up to 70% of the ozone variability, if the seasonal cycle is also considered. A regression model was created for separated time scales of the variables. The separation of short-term, synoptical and seasonal components was done by means of Kolmogorov-Zurbenko filtering. The synoptical scale variations were chosen as the most informative from the point of their relation with meteorological parameters. About 40% of synoptical scale variations of surface ozone can be explained by regression model on separated meteo parameters that is 30% more efficient than ozone residuals usage.

1. Introduction

Surface ozone levels and their changes are of the great interest since harmful effects of O_3 and positive trends of its concentration were established at a number of northern hemispheric locations (Scheel et al., 1997; Roemer, 2001). Since many processes like photochemical generation in the reactions with precursors, vertical and horizontal transport, deposition and some others are affecting ozone concentration, it is difficult to isolate long-term ozone changes connected to changes in the chemical composition of the atmosphere from changes driven by meteorological processes of different time scales.

It is well known that chemical ozone generation is strongly affected by meteorological conditions. For example, ozone levels tend to be higher under hot, sunny conditions favorable for photochemical ozone production. Conversely, wet, rainy weather with

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high relative humidity is typically associated with the low ozone levels provided by wet ozone deposition on the water droplets. Windy weather can influence ozone concentration near the surface differently. In particular, strong wind reflects the increased intensity of the vertical transport. If the boundary layer acts as a source of ozone due to chemical generation, the growth of the wind speed leads to the decreasing of ozone concentration cause of the vertical mixing. Conversely, if the ozone chemical budget in the boundary layer is negative, vertical transport transfers ozone-reach air from aloft downward, and surface ozone concentrations correlate positively with the wind speed.

An investigation of the influence of local meteorology on the surface ozone for a particular site is complicated by advection. Air masses with different history possess different ozone concentrations. For example, Moody et al. (1995) found that up to 50% of spring-time surface ozone day-to-day variations on Bermuda is explained by changes of transport patterns.

Although the relationship between some meteorological parameters and surface ozone has been statistically established for some sites, both in urban and remote conditions (Bloomenfield et al., 1996; Xu et al., 1996), the physical background of the statistical results is not completely understood yet. The first attempt to use separated scales was made by Flaum et al. (1996), who used the separated seasonal component of ozone variations for a regression model creation on the basis of air temperature and dew point temperature dependence.

In this paper we present results of the investigation of meteorological effects on the surface ozone time-series of a site located in the northwestern part of Russia. Together with the regression analysis of the original deseasonalized data, separation of the time scales was applied. Decomposition of the original data into the different time scale components could isolate the periods which contribute most to the bulk correlation. This information is important for a better understanding of the processes that underlay the statistical relationships between surface ozone and meteorological parameters. This could also be of importance for the construction of a regression model of surface ozone variations.

2. Measurements

The Lovozero site (250 m a.s.l., 68.5° N, 35.0° E) is located on the Kola Peninsula plateau on the bank of Lake Lovozero. This station is located away from the strong pollutant sources and it can thus be considered as a background site. Only a slight influence of the industrial towns of Apatity or Monchegorsk located at the distances of 80 km to the southwest or 120 km to the west, respectively, is possible. During the warm periods, this influence is very small, because northern and eastern winds are prevailing. Lovozero station is suitable for the comparison of the polluted air transport from Europe with the clean air transport from the Arctic.

Measurements of surface ozone concentrations are carried out since January 1999. For these measurements DASIBI 1008-AH analyzer with automatic temperature and pressure correction is used. The instrument was calibrated against the reference generator of the Laboratory of Ecological Control (St. Petersburg in 1998) and compared with DASIBI 1008-RS instrument (bought in 1996). The device was also compared with the similar instrument operating at the Russian ozone-measuring network and with the instrument used in the international experiment TROICA (Crutzen et al., 1998). The samples uptake is made every 10 s, and the stored data have a resolution of 1 min, from which hourly, daily and monthly average are calculated.

The Lovozero meteorological station with a resolution of 3 h provides the standard meteorological parameters. They include temperature, relative humidity, wind speed and direction, and precipitation.

3. Regression for daily mean ozone

In this chapter we present the results of regression modeling of the surface ozone variation related to the variability of local meteorological parameters. We chose temperature, wind speed and relative humidity as proxies for the model, since they are the usual proxies for the regression models of the surface ozone (Bloomenfield et al.,

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1996; Roemer, 2001). Despite the fact that these parameters are not completely independent, we suppose that these variables are somewhat complementary because, as it was discussed in the Introduction, all of them influence ozone through different processes. We try also to include in the model the surface pressure which can account for the synoptic scale ozone variations connected to the passages of the different synoptic systems.

Some of the meteorological variables of interest, as well as ozone, have a strong seasonal cycle. Therefore the seasonal variations should be removed from the variables before the regression analysis. To describe the seasonal cycle, we used here the same technique as Zvyagintsev et al. (1996). The annual component was described as a sum of sine and cosine terms, both of them having a period of 365.25 days. To fit better the seasonal behavior, which is not a pure monochromatic oscillation, the semiannual component was also included into the model of the seasonal cycle. This was also described using the same technique, with the sine and the cosine having a period of 182.625 days. These trigonometric functions were applied to the variables using the least square method. In the ozone case, the sum of the annual and the semiannual components accounts for about 50% of the total variance of ozone. The technique was applied to all of the variables, and resulting periodical functions were removed from them. The correlation coefficients between deseasonalized ozone and the meteorological parameters are presented in Table 1.

It can be seen that the correlation coefficients change during the year. Ozone correlation with temperature is positive and statistically significant from winter till summer, but decreases in autumn. The mechanism of this relation is not clear yet. The correlation of ozone with relative humidity is negative in all seasons except winter, with the strongest negative correlation in autumn. The correlation of ozone with wind speed changes remarkably between autumn- winter and spring-summer. In autumn-winter it is about 0.5, suggesting that ozone in the boundary layer is destroyed in the cold half of the year. The small correlation coefficients in the spring-summer mean that vertical gradients of ozone are weak during this part of the year. Taking into account that destruction of ozone on the ground is even larger in spring and summer than on the snow during the winter, one can conclude that there should be a source of ozone near the ground that balances the deposition during spring-summer. One of the possible sources can be photochemical ozone generation from the precursors in the boundary layer. The correlation coefficient of ozone with pressure is seen to be small throughout the year. It implies, probably, that the origin of the synoptical systems is more significant than the systems themselves. For example, anticyclones coming to Kola Peninsula from the south differ strongly in ozone concentration from the arctic ones coming from the north. Probably, the separation of the ozone data set according to the transport patterns is necessary before a surface pressure dependence can be established.

Next we used the stepwise method to construct a multiple linear regression of the ozone concentration. The models were constructed for the complete data set and for different seasons separately. Obtained results are presented in Table 2 and on Fig. 1. It can be seen that the stepwise regression method did not include the pressure into any model as it does not improve the model quality.

Two different measures of the effectiveness of the models are presented in the last two columns of Table 2. The coefficients of the model for the complete data set are presented at the bottom of the table. It can be seen that this regression model is not very effective as decreasing of the ozone variance reaches only the factor of 1.25. This means that ozone residuals variance is almost the same as the variance of the input ozone data. The model can explain only 17% of the ozone variance. Such a low effectiveness of the model originates, probably, from the fact that relations between ozone and meteorological parameters differ significantly during the year and can even change a sign (see Table 1).

It can be seen from Table 2 that the seasonal models fit the observations better. Meteorological parameters have the strongest impact on the surface ozone concentration in spring and autumn explaining 38% and 37% of the ozone variance, respectively. We decided to combine the seasonal regression models into one so that the correspond-

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ing seasonal regression coefficients $KT_{(s_i)}$, $KH_{(s_i)}$, $KW_{(s_i)}$ with i = 1...4 are different for each season. The resulting model can explain about 32% of the ozone variance. This means that it is almost twice more effective than the model which does not take into consideration the seasonal dependence of the coefficients. Taking into account the model of seasonal ozone cycle, the surface ozone concentration $C_{O_3}(d)$ can be expressed in the following form:

$$C_{O_3}(d) = C_0 + \sum_i [A_i \cos(2\pi d/T) + B_i \sin(2\pi d/T)]$$
$$+K_T{}^*T(d) + K_H{}^*H(d) + K_W{}^*W(d) + R(d), \quad (1)$$

where C_0 is a constant, the sum within parenthesis describes the seasonal cycle, which includes annual and semiannual components, K_T, K_H, K_W are corresponding regression coefficients with temperature $T(d)$, humidity $H(d)$ and wind speed $W(d)$, $R(d)$ are residuals after regression on the meteorological parameters application, and d corresponds to the day of the year. This model explains about 70% of the total variance of the ozone.

The results of two models with season-dependent and constant coefficients are presented on Fig. 1. To show better the capability of the model to capture day-to-day ozone variations, only three months of the spring 1999 are presented in the plot. As it can be seen the first one fits the observations better.

4. Time scales separation

Its very important to estimate how the relation between surface ozone variations and meteorological parameters changes on different time scales.

Time scale separation can be reached by the application of different filters (Randel, 1994; Marple, 1990). It was shown that the simplest way to reach a suitable degree of time scales separation is Kolmogorov-Zurbenko (KZ) filtering (Rao et al., 1997). Other filters can also be applied (Roemer and Tarasova, 2002), but they are not so efficient in

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the wide range of frequencies. Another advantage of KZ method is its non-sensitivity to gaps in the original data sets.

The KZ filter is based on running means applied to the time series several times. The output of the previous step serves as input for the next step. Repeated application of the filter provides the necessary noise suppression.

The square transfer function of KZ(m, k) is given by (Rao et al., 1997):

$$\left|\phi_{m,k}(\varpi)\right|^2 = \left[\frac{1}{m}\frac{\sin(\pi m\varpi)}{\sin(\pi\varpi)}\right]^{2k},$$

where ϖ has units of cycles per day (frequency). The parameter k controls the level of noise suppression. Ones k is fixed, m is chosen such that

$$\varpi_0 \approx \frac{\sqrt{6}}{\pi}\sqrt{\frac{1-(1/2)^{1/2k}}{m^2-(1/2)^{1/2k}}},$$

where ϖ_0 is the desired separating frequency. The equation represents the approximate solution of the equation

$$\left|\phi_{m,k}(\varpi)\right|^2 = \frac{1}{2}.$$

To indicate the most informative part of the spectrum, which is most suitable for regression modeling, the following procedure was applied: KZ filtering with the step of 3 points was applied subsequently to time-series of original ozone and of the meteorological parameters. This yielded several separated data sets. In spite of the high degree of correlation between the separated time scales of each component, the correlation of separated ozone and meteo parameters changes substantially for each scale. The changes of the correlation coefficient for different periods are presented in Fig. 2. As it can be seen the most informative part of the spectrum from the point of mutual correlation between surface ozone and meteorological conditions lays in the range of the short periods between 11 days and 1.5 months. This led us to the following approach.

In the case of our measurements as the time series is restricted in time by a 2-year period, the following decomposition was suggested:

$$SH(t) = O_3 - KZ_{3,3}(O_3)$$

$$SY(t) = KZ_{3,3}(O_3) - KZ_{15,3}(O_3)$$

$$SE(t) = KZ_{15,3}(O_3),$$

where SH(t) represents the short term component with the periods shorter than 11 days, SY(t) represents the synoptical scale variations from 11 days to 2 months, and SE(t) represents the seasonal component.

The same decomposition was applied to the time series of the meteorological parameters: daily mean temperature, relative humidity, pressure and wind speed. The separated time series of surface ozone are presented on Fig. 3.

To check the quality of separation the following technique was used. The autocorrelation functions of the separated time series were calculated and then subjected to fast Fourier transformation. Then the ratio of summary power of useful/noise harmonics was estimated showing the part of energy in the selected range of periods. The obtained estimates are given in Table 3. As it can be seen the separation is quite poor for most of the components. This is mainly due to the short length of the analyzed time series and small time resolution. That causes strong aliasing and ringing effects in the analyzed spectra. When comparing the separation of the log transformed ozone and raw time-series it is seen that the seasonal component can be obtained better using ozone filtering, whereas for the other components the log transformation is preferable. Table 3 also shows that the best separation of the seasonal component is obtained for the parameters having strong seasonal behavior like ozone or temperature. Poor separation of the seasonal component is observed for pressure.

5. Regression model for the synoptical component

The correlation coefficients for all separated components (Table 4) were compared with the ones obtained for the residuals in the previous chapter. It is obvious that the highest correlation is observed between the seasonal courses of all separated components. We note that if the correlation between ozone and temperature short and synoptical components is positive, and it gets negative for the seasonal one. The correlation coefficients of seasonal ozone component and meteorological parameters are substantially higher than the same ones for the annual residuals.

As a new approach the regression model for the synoptical ozone component was created. For the model the filtered ozone, temperature, humidity, wind speed and pressure were used. All of them were processed by the same algorithm. As in the modeling of residuals we constructed the model for the whole time series with the same coefficients and with the seasonal coefficients. As before the effectiveness of the models is presented in Table 5.

A comparison of the created regression models shows the following interesting features. Usage of the separated scales is 10% more effective than application of the simple regression on the deseasonalized ozone time series. This is probably connected with the fact that the residuals contain not only a synoptical component, which is the most informative from the point of relation between ozone and meteo parameters, but also a short term component.

There is very weak dependence of the synoptical scale ozone on temperature in winter, and most of the variability is described by wind speed. This relationship is much stronger than in simple regression. The dispersion is reduced nearly to the factor of two.

The regression coefficients are very similar for simple and synoptical scale regression in spring. In summer humidity works much better for separated scales, and the situation is opposite for the wind speed dependence. In autumn the temperature coefficient in the separated scales is stronger, having the same sign as in simple regression

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and opposite to the other seasons. Using the non-seasonal regression of synoptical scale ozone variations can explain 22% of the variability. Application of the seasonal regression shows that meteorological parameters can explain 42% of the synoptical scale variability of the surface ozone.

The results of modeling are presented in Fig. 4. As it can be seen the models tends to underestimate the amplitude of the synoptical scale variation.

6. Conclusions

In the paper the possibilities of the surface ozone description in terms of the empiric regression models on the local meteorological parameters is considered. Two different approaches were applied.

The regression model of ozone residuals after the subscription of the seasonal variations as a parametric functions let us explain about 30% of the surface ozone variability on the basis of the local meteorological condition changes. The model with included seasonal cycle can explain up to 70% of the surface ozone variability.

As a second approach time scale separation was applied. It was shown that the highest correlation in the range of the short periods is observed for the periods between 11 days and 2 months.

Three components separation was done for ozone and local meteorological parameters (temperature, relative humidity, pressure and wind speed). The synoptical scale variations were chosen for regression modeling as the most information in mutual correlation.

Regression model of synoptical scale ozone component of the meteorological parameters can explain about 40% of variation in terms of local meteorological condition changes of the same time scale. This model is 30% more efficient that the one on the base of residuals.

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Table 1. Correlation coefficients between the residuals of the surface ozone and meteorological parameters

	Correlation coefficients between surface ozone and ...			
	temperature	humidity	pressure	wind speed
Winter	0.48±0.08	0.17±0.10	−0.19±0.10	0.48±0.08
Spring	0.57±0.05	−0.25±0.07	−0.10±0.07	0.09±0.07
Summer	0.47±0.06	−0.17±0.07	0.09±0.08	0.19±0.07
Autumn	−0.15±0.07	−0.38±0.07	0.07±0.08	0.52±0.06
Year	0.33±0.04	−0.22±0.04	−0.04±0.04	0.30±0.04

Table 2. Regression coefficients with their errors, and different measures of regression models effectiveness, where (σ_R) and (σ_{RR}) are the standard deviation of the ozone before and after regression model application, respectively

	Temperature	Humidity	Wind speed	1-σ_{RR}^2/σ_R^2	σ_R^2/σ_{RR}^2
Winter	0.16±0.06	–	0.58±0.21	0.27	1.40
Spring	0.70±0.08	−0.14±0.04	–	0.38	1.58
Summer	0.64±0.09	–	0.92±0.30	0.25	1.35
Autumn	−0.35±0.09	−0.13±0.05	1.50±0.20	0.37	1.59
Year	0.31±0.04	−0.13±0.02	0.51±0.10	0.17	1.25

Table 3. The ratio "signal/noise" power in the spectrum of the separated components

Component	ozone	$\ln(O_3)$	relative humidity	temperature	wind speed	pressure
Seasonal	9.96	4.52	4.51	13.68	1.92	0.085
Synoptical	3.94	4.42	3.16	5.56	2.68	5.67
Short	2.43	2.92	4.46	1.95	5.01	1.58

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Table 4. Correlation coefficients between the separated time scales of the surface ozone and the meteorological parameters

	Correlation coefficients between surface ozone and ...			
	temperature	humidity	pressure	wind speed
Short-term	0.03±0.04	−0.29±0.03	0.05±0.04	0.26±0.04
Synoptical	0.28±0.04	−0.38±0.03	0.07±0.04	0.22±0.04
Seasonal	−0.44±0.03	−0.4±0.03	−0.57±0.03	0.61±0.03

Table 5. Regression coefficients for synoptical component

	Temperature	Humidity	Wind speed	1-σ_{RR}^2/σ_R^2	σ_R^2/σ_{RR}^2
Winter	–	0.10±0.09	1.19±0.27	0.48	1.92
Spring	0.63± 0.07	−0.22±0.04	–	0.42	1.73
Summer	0.61±0.01	−0.11±0.04	0.22±0.30	0.36	1.55
Autumn	−0.52±0.09	−0.12±0.07	1.92±0.26	0.45	1.83
Year	0.28±0.04	−0.21±0.03	0.44±0.12	0.22	1.28

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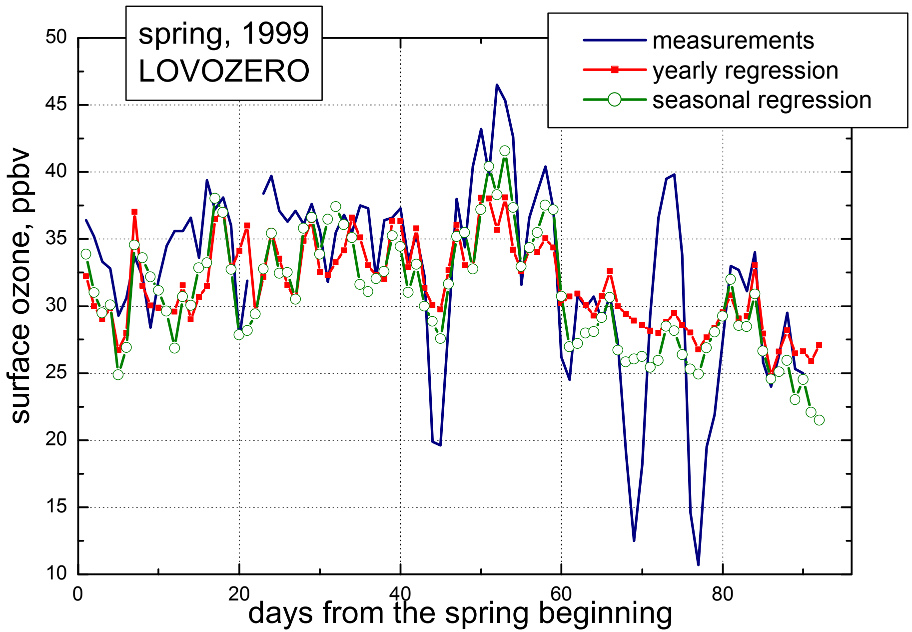

Fig. 1. Regression model of daily mean surface ozone on the local meteorological parameters (spring of 1999).

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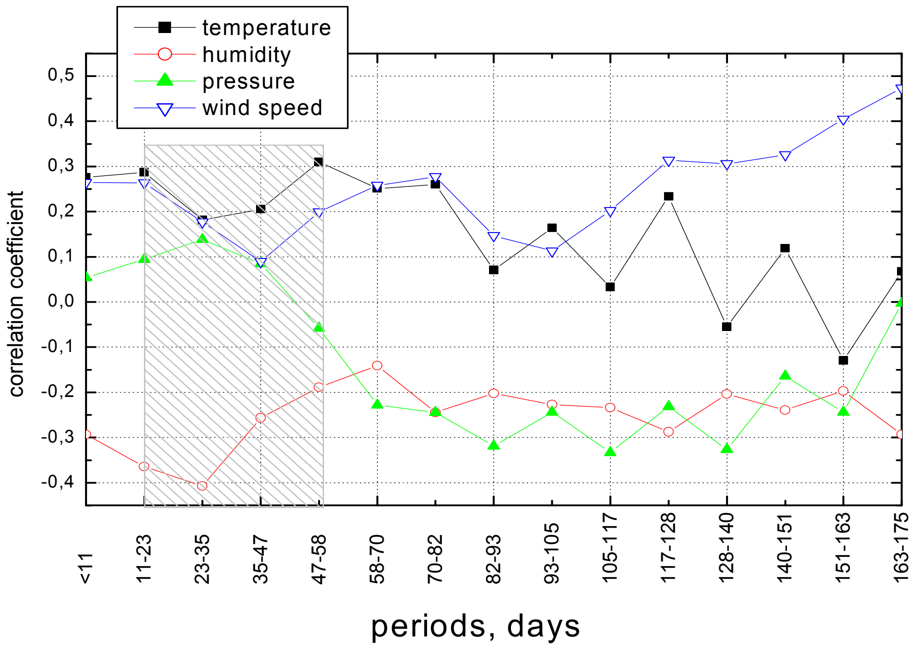

Fig. 2. Correlation coefficients for separated scales of surface ozone and meteorological parameters variations (synoptical periods are shadowed).

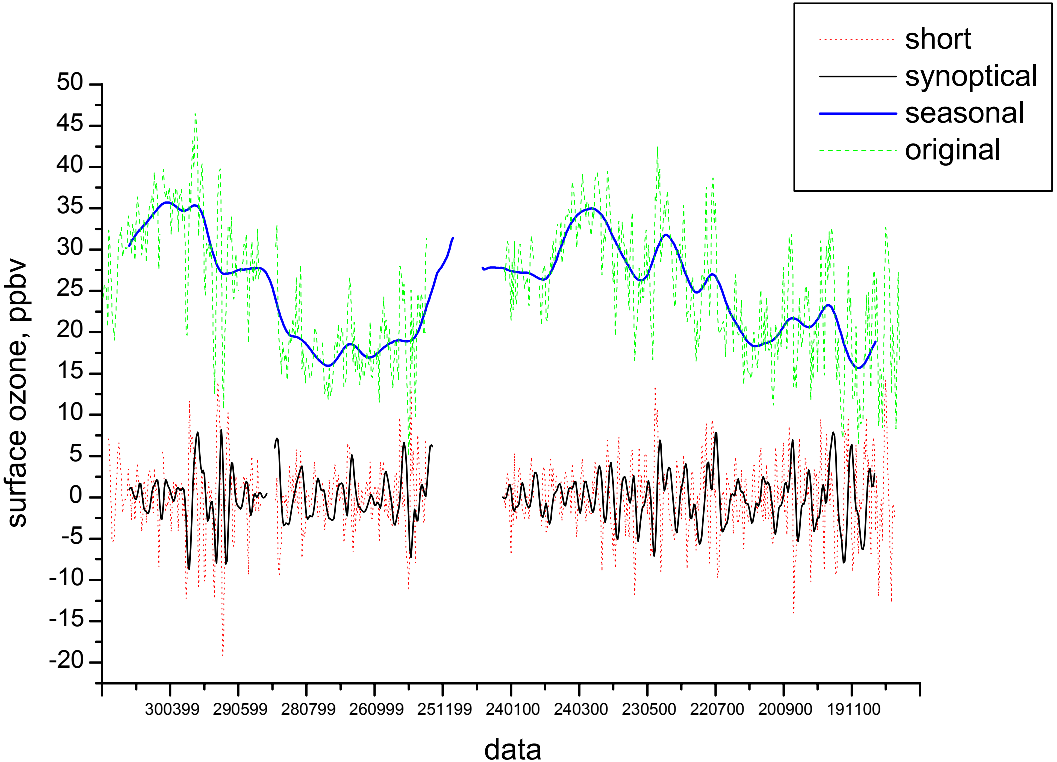

Fig. 3. Separation of the surface ozone time series.

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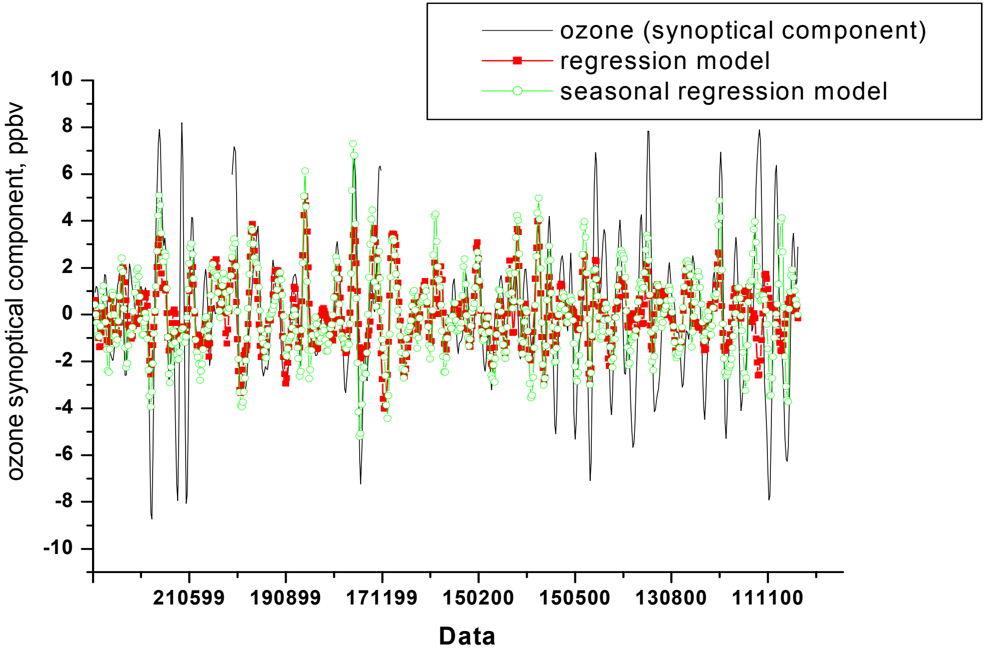

Fig. 4. Regression models application for ozone synoptical component.

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