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The assessment of the atmospheric air quality in Tomsk with the use of a mesoscale photochemical model

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Abstract. The paper presents the results of studying the influence of the development of meteorological situation in clear weather with a weak wind on the level of atmospheric surface air pollution in the city of Tomsk (Russia). Mesoscale meteorological and photochemical models with a new kinetic scheme were applied to the analysis. The main local weather conditions were identified, in which numerical calculations show increased values of the air pollution index. The numerical predictions have confirmed an increase in the ground-level concentration of pollutants in the presence of temperature inversion or isothermy.

1. Introduction

Processes occurring in the lower part of the atmosphere, i.e. in the atmospheric boundary layer (ABL) significantly influence the human life and activity. Special attention must be paid to the deterioration of the surface air quality due to an increase in the volume of pollutants as a result of natural disasters, anthropogenic emissions (the results of intensive human economic activity in large urban areas, accidental environmental pollution). A part of the incoming pollutants enter into chemical and photochemical reactions can lead to the generation of the urban smog. Many components thereof are characterized by a high toxicity and reduce visibility. Due to the requirements to ensure the environmental safety of major population centers, research into the causes and rate of the air pollution is becoming of particular importance. A significant effect that should be taken into account when studying urbanized territories is an anthropogenic atmospheric phenomenon urban "heat island" (UHI) which means a temperature difference of several degrees for a city and a suburb. It is such temperature differences due to which typical of the city circulation of air masses is formed under conditions of a weak transfer in the ABL. This may lead to adverse local environmental conditions within large cities or their suburbs. Therefore, one of the current most urgent fundamental problems of the Earth Sciences is the problem of the research of the basic laws of the atmospheric processes with the use of mathematical modeling methods in order to provide monitoring and forecasting of meteorological and chemical parameters of the surface layer state over populated areas with a specific exchange of mass, momentum and energy between the air and land [1].

Currently, there is a large number of studies on mathematical modeling of air quality in cities. Based on the review of these works, we can identify the following main methods of studying the meteorological regime and atmospheric pollution [2]: empirical-statistical, statistical, analog modeling, mathematical modeling.

For example, empirical-statistical and statistical methods associate various meteorological parameters and properties of the "underlying" terrain. Mathematical models used to assess air quality can be divided into two categories: energy (statistical) and hydrodynamic models. Energy models are designed to study meteorological regime in the surface air over the city. They are based on the heat balance equation. The hydrodynamic modeling method is characterized by some authors as the most fruitful and promising. It is based on the solution of a system of equations describing the meteorological regime of air flow formation in the urban environment depending on the horizontal and vertical components of wind speed, temperature, specific humidity, etc. [3].

Both Russian and foreign scientists work within the framework of the given classification. For example, in [4], the authors study an advective-diffusion model to study the distribution of pollutants in the atmosphere in order to determine whether pollutants are mostly concentrated at the ground level or near the source of emission. This model takes into account wind, diffusion coefficient, and temperature. Numerical solution of this model is carried out by the explicit finite difference method. The calculations carried out by the authors show that the atmospheric conditions affecting the concentration of pollutants are not final since model parameters also affect the atmospheric state.

The article [5] is devoted to the study of atmospheric pollution from the mathematical point of view. The authors describe spatial and temporal distribution of pollutants released into the atmosphere and perform mathematical modeling in the FlexPDE software package. This mathematical model can be a useful tool to estimate air pollution in industrial areas.

In [6], the authors used the weather research and forecasting model coupled with chemistry (WRF-CHEM) which includes anthropogenic and biogenic emissions in Southeast Asia. The article presents a comparison of the results of the model-based computations and ground-based measurements which shows that the WRF-CHEM model simulates meteorological parameters quite well and predicts O3, NO2 and CO. However, the value of the simulated NO2 concentration was underestimated in comparison with satellite observations. This could affect O3 modeling on the surface. The authors suggest that if we correct the underestimated NO2 values the model will probably simulate the values of O3 concentrations more accurately.

Based on the above review of research by modern authors, we can conclude that there is a large variety of mathematical models, but they all have their own areas of applicability and methods of solution. Moreover, the choice of the model and its solution method depends on many factors: available input data, geographical location of the city (environmental conditions, terrain, etc.), weather conditions, etc.

The purpose of this work is to numerically study, using mathematical models, the influence of the evolvement of the meteorological situation in clear weather with a weak wind on the level of atmospheric surface air pollution with primary and secondary pollutants as a result of emissions from industrial enterprises and vehicles of the city. Tomsk (Russia) with population of more than 500 000 people is taken as an example for the calculation.

2. Mesoscale photochemical model

The Eulerian model of turbulent diffusion is used to calculate the concentration of pollutant components taking into account the chemical interactions between them. The model includes transport equations describing advection, turbulent diffusion, and chemical reactions [7]:

$$\frac{\partial C_i}{\partial t}+\frac{\partial UC_i}{\partial x}+\frac{\partial VC_i}{\partial y}+\frac{\partial WC_i}{\partial z}=-\frac{\partial}{\partial x}\langle c_i u\rangle-\frac{\partial}{\partial y}\langle c_i v\rangle-\frac{\partial}{\partial z}\langle c_i w\rangle-\sigma_i C_i+S_i+R_i, i=1,..,n_s. \quad (1)$$

Here C_i, c_i are the averaged and pulsation components of the concentration of the i-th component of the pollutant; U, V, u, v are the averaged and turbulent pulsation components of the horizontal wind speed vector; W, w are the averaged and turbulent pulsation vertical component of the pollutant velocity; $\langle\ \rangle$ – denotes Reynolds averaging; S_i is the source term representing emissions of pollutant components into the atmosphere due to industrial pipes and ground vehicles; R_i describes the formation and transformation of the substance due to chemical and photochemical reactions in the presence of pollutant components; σ_i is the rate of wet deposition of pollutants due to precipitation; n_s

is the number of chemical components of the pollutant whose concentration is to be determined; x, y are horizontal coordinates, the Ox axis is directed Eastward, Oy is Northward, z is the vertical coordinate, t is time, T is the period of modeling. The computational domain is a parallelepiped , L_x, L_y are horizontal dimensions of the area, H is the height of the research area, hz(x, y) is the terrain height above sea level; $-L_x/2 \le x \le L_x/2$, $-L_y/2 \le y \le L_y/2$, $hz(x,y) \le z \le H$, $0 \le t \le T$.

Equations (1) are not closed-form ones since in addition to the C_i concentration which is required to be determined there are additional unknown correlations $\langle c_i u\rangle$, $\langle c_i v\rangle$, $\langle c_i w\rangle$ which model the turbulent diffusion of the pollutant. The closing relations are obtained within thr equilibrium approximation for differential equations of transfer of turbulent momentum, heat and mass fluxes under conditions of local homogeneity of the atmospheric boundary layer [8].

Currently, emissions from vehicles make a major contribution to the formation of secondary air pollutants in cities including generation of aerosol particles of several microns in size which jeopardize human health. Two approaches are mainly used in mathematical modeling of secondary aerosol particle formation. The first [9] approach describes the processes of nucleation, condensation and coagulation that lead to the formation of aerosol particles in detail. The second one [10] represents aerosol formation as overall reactions whose rates are determined empirically in laboratory facilities. In this paper, the second approach is chosen due to its lower computational complexity. For this purpose, the semi-empirical kinetic mechanism GRS (General Reactions Set) [10] developed in CSIRO was added to the 3D photochemical mesoscale model [6,7,11] available to the team of authors instead of the shortcut kinetic scheme of the formation of secondary urban air pollutants with photochemical reaction products developed at the Danish Meteorological Institute [12]. Along with the photochemical reactions of ozone formation in the city atmosphere, the GRS also considers generalized reactions of atmospheric aerosol formation (inert non-gaseous carbon compounds *SNGOC*, nitrogen *SNGN* and sulfur *SNGS)*. In the developed modification of the kinetic mechanism, the reactions of ground-level ozone formation are presented in more detail: instead of two reactions of the basic GRS mechanism, six reactions are considered where nitrogen dioxide and monoxide, various states of atomic oxygen (excited and ground) are involved apart from ozone.

The advantage of the proposed modified semi-empirical kinetic mechanism is that, along with a more detailed description of gaseous chemical reactions, the overall reactions of formation of aerosol particles of 10 μm and 2.5 μm in the urban atmosphere are also quite compactly described.

Smokestacks of industrial enterprises (about 300) located in Tomsk and the Tomsk region, as well as city roads were considered as sources of air pollutants. The diurnal traffic emission rate was modeled according to the following law: *max (0.05+0.95 sin (π(t[h]-6)/18),0.0).*

Table 1. Chemical reactions of the modified kinetic mechanism of aerosol formation.

	Reactions	Reaction rate	Source
1.	$R_{smog} + hv \rightarrow RP + R_{smog} + \eta SNGOC$	$r_1 = k_1 C_{Rsmog}$	GRS
2.	$RP + NO \rightarrow NO_2$	$r_2 = k_2 C_{RP} C_{NO}$	GRS
3.	$RP + RP \rightarrow RP + \alpha H_2O_2$	$r_3 = k_3 C_{RP} C_{RP}$	GRS
4.	$RP + NO_2 \rightarrow SGN$	$r_4 = k_4 C_{RP} C_{NO_2}$	GRS
5.	$RP + NO_2 \rightarrow SNGN$	$r_5 = k_5 C_{RP} C_{NO_2}$	GRS
6.	$RP + SO_2 \rightarrow SNGS$	$r_6 = k_6 C_{RP} C_{SO_2}$	GRS
7.	$H_2O_2 + SO_2 \rightarrow SNGS$	$r_7 = k_7 C_{H_2O_2} C_{SO_2}$	GRS
8.	$O_3 + SO_2 \rightarrow SNGS$	$r_8 = k_8 C_{O_3} C_{SO_2}$	GRS
9.	$NO_2 + hv \rightarrow O(^3P) + NO$	$r_9 = k_9 C_{NO_2}$	DMI
10.	$O_3 + hv \rightarrow O(^1D) + O_2$	$r_{10} = k_{10} C_{O_3}$	DMI
11.	$O(^3P) + O_2 \rightarrow O_3$	$r_{11} = k_{11} C_{O(^3P)} C_{O_2}$	DMI
12.	$O(^1D) + N_2 \rightarrow O(^3P) + N_2$	$r_{12} = k_{12} C_{O(^1D)} C_{N_2}$	DMI
13.	$O(^1D) + O_2 \rightarrow O(^3P) + O_2$	$r_{13} = k_{13} C_{O(^1D)} C_{O_2}$	DMI

14. $O_3 + NO \rightarrow NO_2 + O_2$ $r_{14} = k_{14} C_{O_3} C_{NO}$ DMI

Where R_{smog} is the reacting part of the smog emitted by traffic and point sources, O(^{1}D) is the excited state of atomic oxygen, O(^{3}P) is the ground state of atomic oxygen, RP is radical pool, SNGOC is a stable non-gaseous organic carbon, SGN is stable gaseous nitrogen products, SNGN is stable non–gaseous nitrogen products, SNGS is stable non–gaseous sulphur products, Airborne Particulate Matter (APM) and Fine Farticulate Matter (FPM) that include secondary particulate concentrations consisting of (SNGOC), (SNGN) and (SNGS) [10].

3. Mesoscale meteorological model

The mesoscale meteorological model TSUNM3 (Tomsk State University Nonhydrostatical Mesoscale Meteorological Model) [13] is used to prepare meteorological fields for calculation of the distribution of primary and secondary components of the anthropogenic pollutant. The mesoscale TSUNM3 model predicts wind velocity components and temperature-humidity characteristics in the atmospheric boundary layer at 50 vertical levels (up to 9000 m) for the area of 200×200 with the nested domain of 50x50 km (grid spacing is 1 km with the center in Tomsk). The model is initialized based on the results of a numerical forecast based on the operational global model SL-AV of the Roshydromet of the Russian Federation [14]. The TSUNM3 model uses the non-hydrostatic quasi-compressible approximation for equations of motion in a coordinate system with variable vertical resolution that follows surface topography. The spatial and temporal trends of dependent variables generated by the model of a larger scale (SL-AV operational global model) are used as lateral boundary conditions for the components of horizontal velocity, temperature, and humidity. Interaction of the atmospheric boundary layer with the surface is modeled with the use of the empirical relations of the Monin-Obukhov similarity theory and the non-stationary equation of heat transfer in the near-surface soil layer. The model takes into account short-wave and long-wave radiation with cloud layer effects, as well as the formation of precipitation in the form of rain, snow, ice crystals, and graupel. Turbulent structure of the atmospheric boundary layer is simulated on the basis of the prognostic equation for the turbulence energy and algebraic relations for the turbulence scale and turbulent diffusion of heat and momentum. There is a parallel version of the mesoscale model developed using the parallel programming standard for distributed memory systems – Message Passing Interface.

4. Some computation results

To test the developed set of models for the study of local meteorological conditions and atmospheric air quality over large localities, computations were compared with measurements made with the use of instruments of the Joint Use Center (JUC) "Atmosphere" of V.E. Zuev Institute of Atmospheric Optics of the Siberian Branch of the RAS (http://juc.iao.ru), as well as instruments of the meteorological station of Tomsk.

Figure 1 shows graphs of calculated and measured values of surface meteorological parameters, such as temperature and relative humidity at a height of 2 meters above the earth's surface, as well as wind speed and direction at a height of 10 meters. The calculations are compared with the measurements made by instruments of the TOR-station of the CUC "Atmosphere" and the Tomsk meteorological station. Figure 1 shows that on this day (July 3, 2013), a weak east wind was observed in Tomsk with changes in surface temperature and humidity during the day which is typical for low-cloud weather. There was no precipitation on that that (http://rp5.ru). Comparing the obtained values, it can be noted that the mesoscale meteorological model predicts surface wind characteristics numerically quite well. Some underestimation of the decrease in air temperature in the morning indicates the need to pay attention to the modeling of the interaction of the atmospheric boundary layer with the terrain. During the daytime and evening hours, the model reproduces changes in relative humidity and temperature quite well.

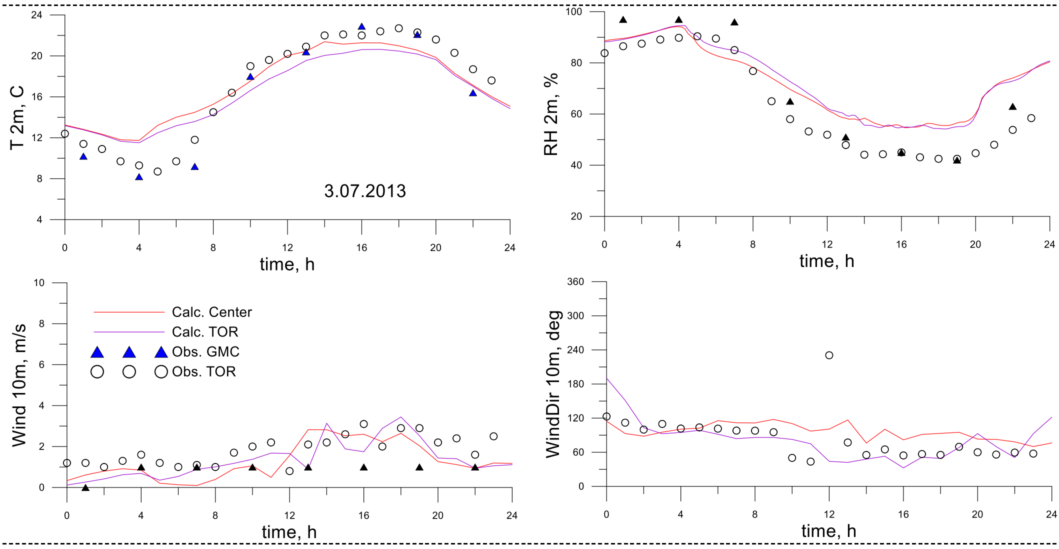

Figure 1. Near – surface calculated and measured meteorological parameters (air temperature and humidity at 2 meters, wind velocity and direction at 10 meters). Solid lines are predictions at the points where meteorological units are distributed. TOR – location of TOR-station of the JUC «Atmosphere», GMC–location of the meteorological station 'Tomsk', Center – a center of Tomsk city.

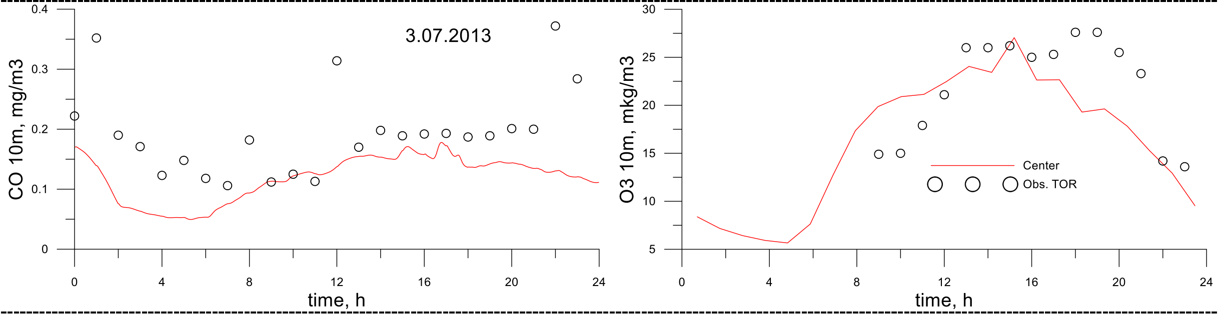

Figure 2. Near surface calculated and measured concentrations of carbon monoxide CO (mg/m2) and ozone O3 (μg/m3). The solid line is numerical predictions, signs are measured values.

Figure 2 shows graphs of diurnal variations of the values of the ground-level concentration of carbon monoxide – the main air pollutant emitted by industrial enterprises and traffic vehicles in the city, and the ground-level concentration of ozone, the intensity of which is significantly influenced by photochemical reactions shown in Table 1. Figure 2 shows that the level of predicted and measured values. Deviations in the distribution of the values under consideration are due to the fact that since there is no more detailed information about local changes in the intensity of emissions during a specific day, the computations use the values of carbon monoxide and ozone precursors emission calculated from the average annual estimates of the environmental services of Tomsk. However, the figure shows an increase in the concentration of CO and O3 in the surface air during the midday period of the day when there is the greatest activity of traffic vehicles and high intensity of solar radiation which stimulates photochemical reactions.

Appropriate dates of 2015 were selected to study the impact of the meteorological situation (clear weather with a weak wind) on the level of surface air pollution (with primary and secondary pollutants of the emissions from industrial enterprises and traffic vehicles of the city). The same values of industrial and motor vehicle emissions were used in calculations for the dates of different seasons. Emissions from heat power plants were not taken into account in the warm months (May-September). The following values were considered as background values of the main concentrations: CO_0=200ppb, $NO2_0$=20ppb, $O3_0$=20ppb, $SO2_0$=3,7ppb, $Rsmog_0$=0, NO_0=0,1ppb, APM_0=0 $\mu g/m^3$. Computations of pollution transport were carried out for two days on a grid with a step of 500 m in the 50x50 km area with a vertical resolution of 10m. To assess air quality, the Air Pollution Index (API) was used. The API values are calculated based on the calculated concentrations of CO, SO2, NO2, NO, O3 with the use of the following formula:

$$API = \sum_{i=1}^{n} \left(\frac{C_i}{C_{i\infty}} \right)^{a_i},$$

where C_i is the concentration of the *i*-th substance, µg/m^3; C_i is the short-term exposure limit of the *i*-th substance defined by the Ministry of Health Care and Social Development of the Russian Federation in µg/m^3; a_i is the dimensionless constant of reduction of the hazard of the *i*-th substance to the hazard of sulfur dioxide, which depends on the hazard class to which a pollutant belongs. a_i values are 1.5; 1.3; 1.0 and 0.85, respectively, for the first, second, third and fourth pollutant hazard classes.

Table 2. Conditions of Tomsk air quality assessment modeling.

Date	Temperature, ^{0}C	Humidity, %	Wind, m/s	Direction	Precipitation (mm)	Cloudiness, %
25.01.2015	min= -35.7 max= -28.5	min=65 max=72	quiet wind (1 m/s)	windless conditions, still air	no precipitation	20-30
21.02.2015	min= -23.7 max= -6.5	min=26 max=77	quiet wind (1 m/s)	East-North-East wind	no precipitation	50
8.04.2015	min= -3.0 max= 12.9	min=22 max=77	still air	windless conditions, still air	no precipitation	60
6.06.2015	min=9.9 max=26.1	min=29 max=84	quiet wind (1 m/s)	North-East wind	no precipitation	15
12.06.2015	min= 11.9 max=20.2	min=33 max=89	quiet wind (1 m/s)	West-South-West wind	no precipitation	40
18.07.2015	min=16.5 max=28.7	min=31 max=78	quiet wind (1 m/s)	East-North-East wind	no precipitation	0
18.08.2015	min= 13.3 max= 26.3	min=49 max=96	quiet wind (1 m/s)	North-East wind	no precipitation	65
8.09.2015	min= 4.6 max=18.0	min=39 max=95	quiet wind (1 m/s)	windless conditions, still air	no precipitation	40
21.09.2015	min=4.8 max=9.1	min=71 max=94	quiet wind (1 m/s)	North-East wind	no precipitation	100

Figure 3 shows graphs of changes in the maximum API values for the territory under consideration 50x50km^2 for each calculation hour during the day for the cold (October-April) and warm (May-September) periods of the year (Table 2).

The figure shows that the maximum API values are observed from 10 to 17 o'clock with a low surface wind speed in cold months (Figure 3a, b, Table 2). And not one but two local extremes are possible in the period from 10 to 17 o'clock. In warm weather (Figure 3c, d; Table 2), there are usually two peaks: morning from 5 to 10 o'clock and evening – from 16 to 22. The closer the date under consideration (Table 2) to the cold period is, the closer these peak values are located with respect to time of appearance. The dates of April 8 and June 6, 2015 with the highest degree of air pollution in the research area predicted by the models were chosen to study the meteorological reasons for the numerical prediction of maximum API values by the photochemical mesoscale model. Vertical profiles of temperature and air pollution index were built for the position located in the center of the city of Tomsk (84.95^{o}E, 56.49^{o}N) for 0, 4, 8, 12, 16, 20, 24 o'clock of the local time of the specified dates. Figure 4 shows that the models predicted the formation of elevated concentrations of air pollutants at an altitude of about 100 m on the morning of April 8, 2015. The increase in the concentration of pollutants is explained by the presence of an elevated temperature inversion at 00 and 04 o'clock, with the lower boundary at the altitudes of about 100 and 50 m, respectively. As is known, inversion is a stable layer with no turbulent movements. Pollutants from high-rise sources (pipes or smokestacks of industrial enterprises) located within the city accumulate under the inversion layer. Note that at a height of 10m, the API values are relatively small. Starting at sunrise (8 o'clock), the surface layer is turbulized due to convection, which in low wind conditions leads to the alignment of the vertical API profile and an increase in its values at a height of 10 m, i.e. to surface air quality deterioration. At a later time, according to the computations, the pollutant is well mixed due to a high level of turbulence in the atmospheric boundary layer and is deposited in the urban area; and the value of the air pollution index at an altitude of 10 m decreases increasing again only in isothermy conditions (or slight temperature inversion) at 20 o'clock.

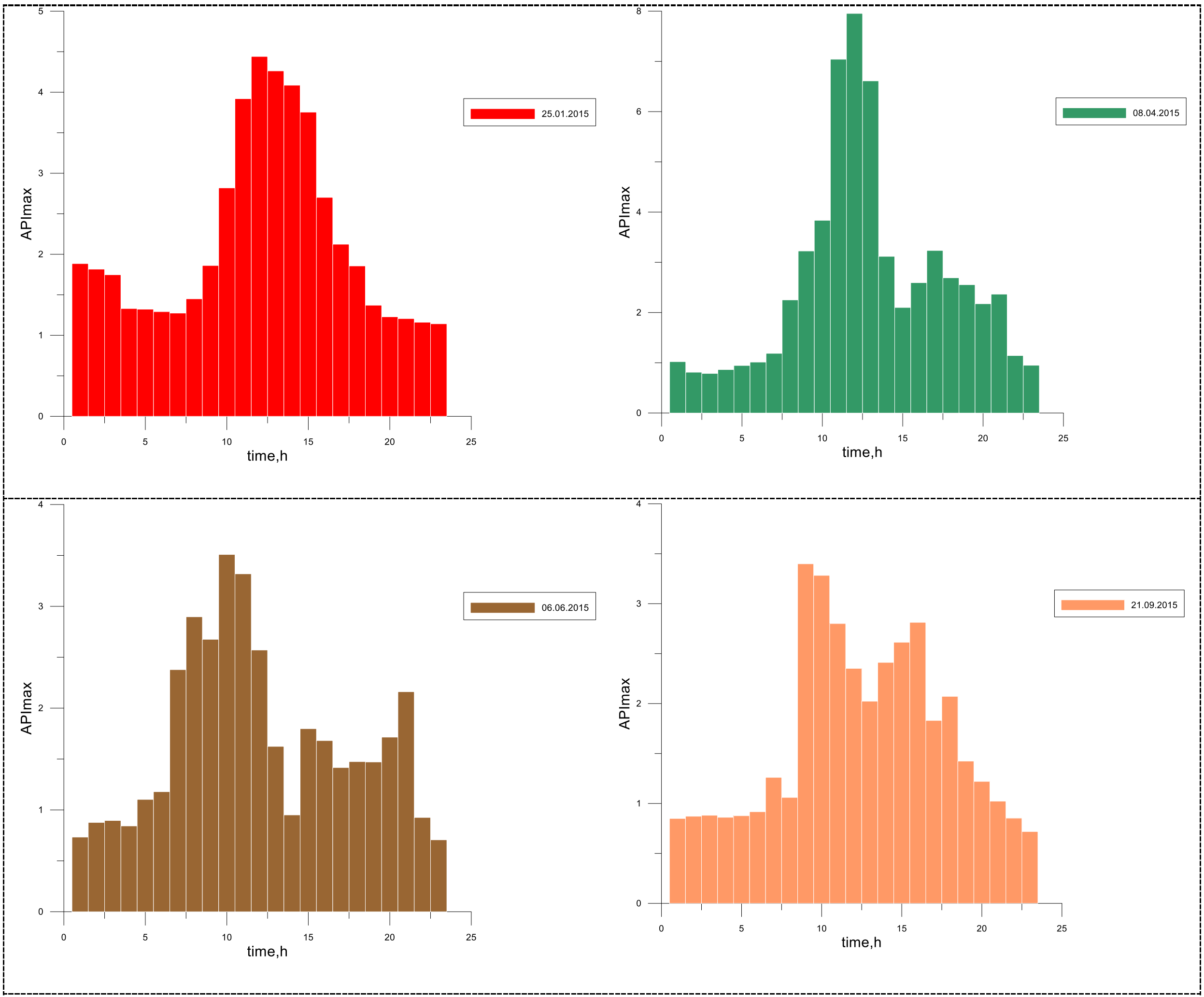

Figure 3. Time change of the hourly maximum values of air pollution indices over time for the research area. Time change of the hourly maximum values of air pollution indices for the research area for various historical dates: a) 2015/01/21; b) 2015/04/08; c) 2015/06/06; d) 2015/09/21

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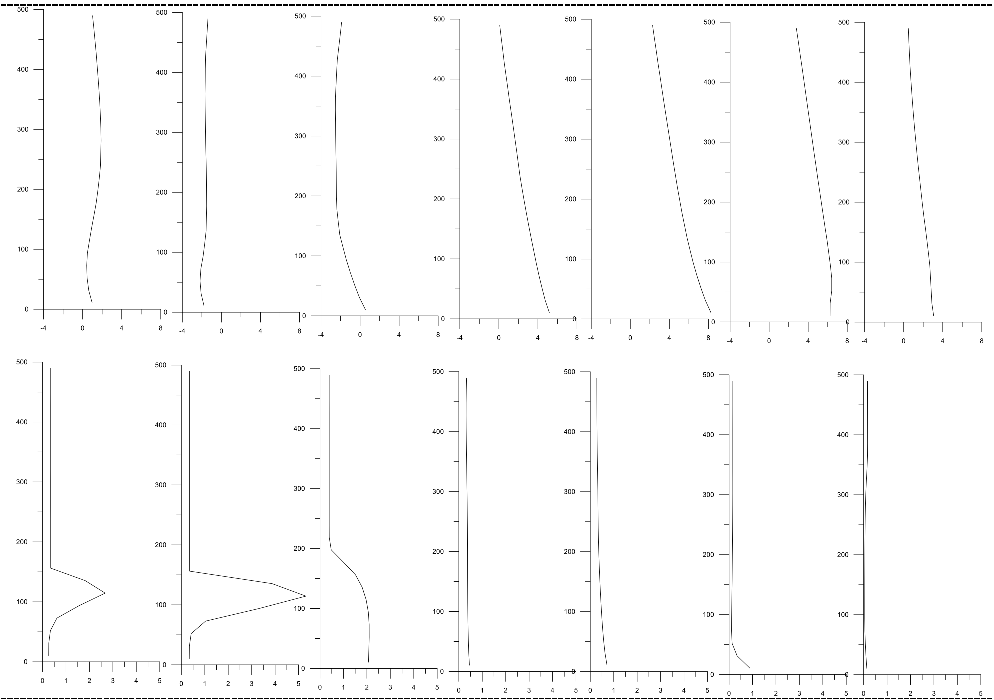

Figure 4. Calculated vertical profiles of temperature and air pollution index over the center of Tomsk with the course of time on April 8, 2015.

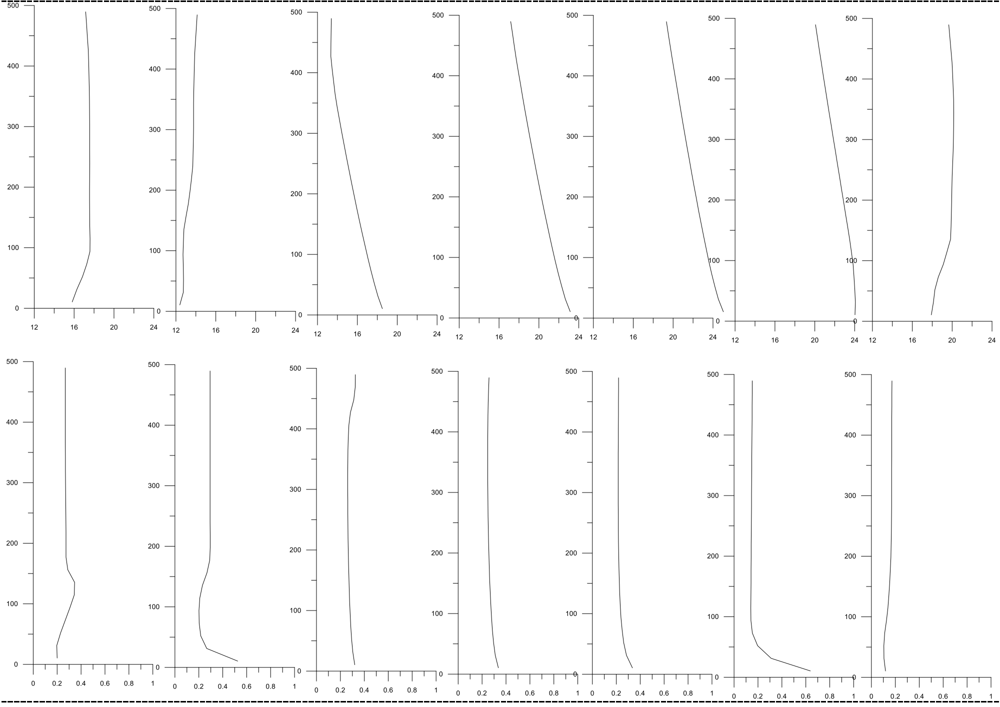

Figure 5. Calculated vertical profiles of temperature and air pollution index over the center of Tomsk with the course of time on June 6, 2015.

Figure 5 shows vertical profiles of temperature and air pollution index on June 6, 2015, for the position located in the center of Tomsk. The figure shows that the highest API values at a height of 10m are predicted by the model for 4 and 20 o'clock of the considered time period. It is during these hours that the calculated temperature profiles suggest temperature isothermy and even temperature inversion in the surface layer (stable stratification), whereby vertical turbulent mixing decreases and pollutant accumulates near the earth's surface. At 0 and 24 o'clock, the temperature profiles also show inversion. But due to the weak ingress of the pollutant from ground sources, its accumulation at an altitude of 10m is not numerically predicted. During the daytime, due to the intense flow of solar radiation, calculations show a well-developed convective boundary layer with turbulent mixing which, even with a weak wind and an increased level of air pollution emissions during daytime hours, provides safe conditions for air quality in the city.

5. Conclusion

The paper presents the mesoscale meteorological and photochemical models developed at Tomsk State University to study and forecast the development of the meteorological situation and assess air quality over the city. For inclusion in the photochemical model, a new kinetic scheme is proposed which is based on the kinetic schemes developed in CSIRO [10] and DMI [12]. Its feature is the use of a reduced semi-empirical description of chemical and photochemical reactions that lead to the formation of secondary pollutants as a result of anthropogenic emissions. The models were validate on the data of the measurements of meteorological parameters and ground – level concentrations of pollutants obtained by the Joint Use Center "Atmosphere" of the V.E. Zuev Institute of Atmospheric Optics of the Siberian Branch of the RAS. Using the models, the influence of meteorological parameters on air quality in the city of Tomsk at the weak wind and partly cloudy weather was studied. Meteorological conditions specific for the dates when the computations showed increased API values were identified.

Numerical computations confirmed an increase in the concentration of pollutants in the presence of temperature inversion (or isothermy): the largest amount of pollutants is registered under the inversion layer. The lower the base of the inversion is, the higher the level of pollutants at the earth's surface is. Lowering the inversion leads to an increase in the level of pollution at the Earth’s surface.

Acknowledgments

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