
Pollution of atmospheric air above the city highways.

Olena Voloshkina Doctor of Science, Professor. ORCID: 0000-0002-3671-4449¹

Rostyslav Sipakov Ph.D. student. ORCID: 0000-0002-0862-5043^{1*}

Dmytro Varavin Ph.D. student. ORCID: 0000-0002-4161-9702¹

Yevheniia Anpilova Senior Research Scientist. ORCID: 0000-0002-4107-0617²

Tetiana Kryvomaz Doctor of Science, Professor. ORCID: 0000-0001-7426-8745¹

Julia Bereznitska Ph.D., Associate Professor. ORCID: 0000-0001-7953-3974¹



¹ Kyiv National University of Construction and Architecture, Vozduhoflotsky Avenue 31, Kyiv, 03680, Ukraine

² Institute of Telecommunications and Global Information Space, National Academy of Sciences of Ukraine, Chokolivsky Boulevard, 13, Kyiv, 03087, Ukraine

*Corresponding author e-mail: sipakov.rv@knuba.edu.ua

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ABSTRACT

The study is devoted to the estimation of pollution deposit caused by vehicles into the city total greenhouse gases emissions and to the forecast of the secondary air pollution with formaldehyde.

The present study contains mathematic model which allows getting the estimation of vehicles emission in the total amount of polluted atmospheric air. The model consists of two interconnected parts: a dynamic and a kinetic one. The first part of the model is based on the Theory of Convective Jet and Thermals in the Atmosphere and allows to get the parameters of the contaminated air which is being formed over the highway. The Dynamic part of the model allows to get the total amount of hydrocarbons emission depending on the number of vehicles which is present at the same time on the highway. We also obtain the data on concentration of pollution and temperature increasing in the narrowest section of the thermal jet.

The kinetic part of the model allows to determine the concentration of the secondary atmospheric air contamination with formaldehyde as a result of photochemical transformation of hydrocarbons which are the part of emission of internal combustion engine. The speed of the transformation reaction was determined by the Can't Hoff-Arrhenius deferential equation. The given mathematical model was tested with the help of the system of regular observation over the polluted atmospheric air in Kyiv city. The comparison of the estimated value of formaldehyde concentration and the observation data doesn't exceed 5 % above the maximum values. According to average monthly rates there is a false positive error (to + 20 % in the warmest months).

Basing on the given model the calculator of concentration of pollution by vehicles was developed. This calculator allows to obtain rapidly the values of formaldehyde concentration, depending on air temperature, solar radiation and number of vehicles on the highway. The amount of hydrocarbon emission from the vehicles, according the suggested model, allows to estimate the deposit of vehicles into the total amount of greenhouse gases of the city. The obtained results could be useful in forming of ecological policy of the cities and in managerial decision making.

1. Introduction.

The constant population increasing with the uneven simultaneous geographical distribution of the population density lead to global increasing impact of the urbanized territories on environment. It was stated that big cities significantly affect air pollution even at large distances [1-6]. According to the preliminary estimation of the Global Report on Human Settlements UN-HABITAT, the deposit of the cities into the climate changes of the atmosphere can reach from 40 to 70 per cent of greenhouse gases (GHG) emission. The increased dependence on personal transport is making it to be the main reason of the GHG emission in most cities, especially in developing countries. There is a direct dependence between population well-being growth and the number of personal transports. According to the forecast of the Program UN-HABITAT on people settlements, the number of cars will increase, and until 2050 it will reach 2,6 billion, that is 50% if compared to 2011. To find out the role of cars emissions in big cities in contamination of the atmosphere on condition of global climate changes we should make a large-scale research with attraction of the regional municipality data. We need interdisciplinary integrated estimation tools for optimizing measures to reduce the impact of megacities on human health and the climate [5].

This study relates to the assessment of mutual influence of temperature increasing in megacities as a result of global climate changes and the level of contamination of the atmospheric air from vehicle emissions.

In the research which is described in this article, the authors considered the following points:

-the analysis of the monitoring data of a number of megacities showed that along with increasing of hydrocarbon emission from vehicles there is an increasing of the most powerful pollutant of the atmospheric air - formaldehyde (secondary pollution), the formation of which depends on meteoconditions of the given area [7-12];

- the formaldehyde pollution of the atmosphere is an indicator of photochemical smog formation in big cities of the world. As the confirmation of this fact there is the smog formation over the highways, which is observed on neutral atmospheric conditions as well as with high temperatures. For example, for big cities in Ukraine this index is 3-5 M.A.C of the given toxic substance in summer months [10 - 14].

According to these statements the authors suggest mathematical model which consists two parts, which, in turn, describe the dynamics and kinetics of the given process. The first part considers the amount of convective air contamination above the highways with particular concentration of the emitted hydrocarbon, the second one - is based on the known laws of chemical kinetics. The developed calculator based on the given model allows to define the amount of emitted hydrocarbons when inventing greenhouse gases on urbanized territories and the secondary pollution with formaldehyde in consequence of chemical transformation.

2. Bibliographic data analysis and problem statement.

The most significant by-products of human activity in big cities are gases and aerosols emissions into the atmosphere, such as: carbon dioxide (CO₂), nitrogen oxides (NO_x), volatile compounds (VOCs), ammonia (NH₃), sulphur dioxide (SO₂), black and organic carbon (BC, OC). These gases and pollutants have an impact as on composition of the atmosphere as on the climate. It should be noted that numerous researches on modelling and monitoring on cities emissions impact on air quality and on climate in regional and global scale have been conducted [4,5,7,8,10,11,12,14,15].

As it is stated in [1], the first research on impact of emissions in the cities on global quality of the air was published by Mayer M. with co-author in 2000. Though it doesn't describe the impact of megacities on the global atmospheric composition, the connection between urbanization and key composition of the atmosphere was analyzed, such as NO_x, ozone and hydroxyl radical.

Many works (by Gurjar, Guttikunda Lawrence, Butler, Folberth, Marlier etc) are devoted to the researches on analysis of impact of urbancenosis on the quality of the air on a global and regional scale. The review of these authors' works are presented in details in the following resources [1,4,5,6].

Atmospheric cleaning and climate effects as well as the integrated tools for estimation and mitigating of consequences are presented by the European project which includes 23 research groups from 11 countries. The main attention in this project is paid to the estimation of megacities impact and hot points on air contamination on local, regional and global scales as well as to quantitative feedback assessment between air quality and climate [5].

Mega cities can affect the environment in several ways. Additional impacts on environment are happening because of goods and waste transportation in the cities and beyond. The impact of megacities and their growth can be described in different forms that can be used on local, regional and even global scale [1].

Dispersion trace of megacities is designed to quantify the outflow of gases and aerosols from their point sources (minimum, and on global scale). In other words, the dispersion traces describe the maximum distance from the source, i.e. megacity, over which the significant levels of pollution concentration can be revealed. Two main factors determine the efficiency of the pollution outflow on the regional level: 1) geographical location and 2) lifetime of the pollutant

The lifetime of atmospheric pollution types can last as for hours (for example, some volatile organic compounds) as for years (methane, oxide and, of course, carbon dioxide). The longer lifetime the bigger chance for particular gas or aerosol traces to become an object of long-term transportation. On the other side, meteorological conditions determine the outflow strength and depend on geographical location of the city and the season, during which the pollutants emission is happening.

The impacts and effects of the cities emissions/pollutants of the air have non-linear interaction between each other. As it is noted in the research of David D. Parrish and other, there is an interaction between mitigating effects on climate changing and air condition improving. So, in [4] it is stated that the summer photochemical smog was firstly determined as ecological problem in Los Angeles in 1950s. Emission standards for cars introduced since 1966 led to necessity of cs in cars, to clean unleaded fuel and emission minimization. These measures allowed to significantly reduce emissions of GHG despite the expansion of car trade and traffic in megacities [4].

So, the technological solutions that include the transition to the renewable heating in the residential sector and promotion of catalytic converters developing as well as more efficient car engines along with the implementing of the best traffic management systems - is the way to reducing of urban environment pollution and GHG emission.

In order to define the part of cars emission when inverting the total GHG emission of the big city, we should consider the way how the urban environment climate is formed.

Since the late XX century a lot of works have been devoted to the researches of features of urban climatic system developing.

One of the main factors of urban climate developing is the anthropogenic heat flows over the thermal spots in the city which increase the temperature and form the so called "heat islands" and "city stone chimneys" And, though lately the industrial enterprises are located in the suburbs, the indicators of technogenic heat flux isn't decreased, that can be explained by the following factors:

- the character of urban area construction which determines the airing and absorption of solar energy from the surface of the territory.
- car numbers increasing that work on gasoline (lately the traffic jams on big crossroads and highways have become commonplace for big cities and they make a significant deposit into technogenic heat flows formation by adding more heat)
- regional occurrence of climatic processes and local climatic effects.

It should be noted that the last factor has a significant impact on the dynamics and structure of the main life-saving resources and energy consumption of the city and it must be taken into account while developing the socio-economic concept of the city.

The issue of pollution depending on the temperature of urban environment are considered in detail in the studies by Richard C., John H., Brigman H. et al. [7,8,18 et al.]. In the studies of many authors this issue has been considered basing on the long-term monitoring data analysis [9-12,14,17,19]; other authors applied the approaches which were based on chemical kinetics laws [7,15,18].

But all of them have proved the dependence of the atmospheric air pollutants and the photochemical processes with the air temperature, and this is not in doubt. But the existing methodology of modelling as of the special distribution of pollution as of the model of atmospheric diffusion cannot be unified because there are substantial differences of the big cities under consideration, their microclimate, socio-economical conditions of development, and all this must be included into the model with large number of factors. Beside this,

Modern methods of assessment and forecast give opportunities to realize numerically pollution of the atmospheric air of the urbosystems from stationary and mobile sources together, without proper assessment of the separate source [20,21]. Now the most known, widespread and effective methods are considered to be the ones that are based on realization of mathematical models which, in their turn, are based on equations of mathematical physics with the conjugation of statistical models [15,18]. As a rule, such forecasts can be used only for particular territories under consideration because of their natural and climatic features and the level of socio-ecological structures development. As an example, we can use the model of the local

forecast on pollution of the atmosphere with formaldehyde in the city of Tomsk [20], the model of atmosphere pollution in Ust-Kamianohirsk [21], the model of atmosphere pollution based on the program complex Land Use Regression [22] and other. Besides, the possibilities of the existing models allow to get only the assessment of the "deposits" of separate sources (including vehicles) into the total pollution of the city atmosphere (in percentage terms).

But in practice it is difficult to justify and provide the necessary up-to-date information to the managerial decision-making authorities for assessment and forecast of smog situation emergence taking into account meteorological conditions of large accumulation of vehicles. In addition, to provide the stable development of a megacity there is a need of control of GHG emission by mobile sources.

An integral part of such assessment is the calculating part which could give an opportunity to determine the hydrocarbon emission concentration from cars engines and the level of their transformation into formaldehyde, which, as it is known, is an indicator of photochemical smog emergence from vehicles in many cities. The realization of such model will allow to make an assessment and forecast in the tasks of GHG inventory in big cities.

3. Purpose and tasks of the study.

The purpose of the study is the definition of hydrocarbon emission over the highways in urban environment and to define the secondary pollution with formaldehyde depending on climatic conditions of the area.

The research tasks:

to develop mathematical model which includes two subsystems - dynamic and kinetic, which will allow to determine the formation of air contamination dome over the crossroads and formaldehyde concentration in the air depending on the quantity of car engines and on-air temperature in stable conditions;

to conduct the testing of the received model;

to design the calculator for atmospheric air pollution assessment depending on climatic conditions and the quantity of hydrocarbons emission over the highway;

to investigate how the increasing of the cars number which work on gasoline and diesel fuel effect on the volume of GHG emission.

4. Materials and methods of the research.

The common scheme of the ways of transfer and transformation of natural and anthropogenic emissions is presented by the authors of the following studies [8,12,16]. According to the given classification

they distinguish six main points: I - sedimentation on unstable meteoconditions by the index of turbulence of the atmosphere and by the gradient of air temperature distribution with respect to height; II - emission into the zone of below-cloud scavenging by the rains; III - emission into the cloud cover area; IV - emission into the atmosphere open to solar rays; V - rainfall from acid (polluted) clouds; VI - raising with drops transformation.

Pollutants dispersion by the way I is estimated analytically with the expected concentration in the atmospheric air which is connected with the amount of substances emitted per one-time unit and is specified by unfavorable meteoconditions. The process is very rapid and covers the territory with radius of several hundred meters from the source. The way (II) with the below-cloud scavenging is characteristic to the stable conditions (inversion and low turbulence) with long-term precipitation. Clouds dissolving in water (III) is related to neutral meteoconditions, it is accompanied by longer period of staying in the atmosphere and is ended or by precipitation (V) or by raising into the upper layers of troposphere (VI) or by the drops transformation into ice and by their freezing with the release of small particles like SO_4^{2-} , NH_4^+ , NO_3^- , and by their inclusion to the global transfer.

Let us consider the photochemical transformation (IV) under influence of sunlight on stable or neutral conditions in the absence of clouds which make a specific atmosphere pollution during the light period of the day.

The processes of transfer and transformation of pollution with clouds as well as meteorological factors for neutral atmospheric conditions formation are described in details in the study by S. Kavelt [16].

The streams with their own temperature which differs from the one of atmospheric surrounding gain their own buoyancy. On normal meteoconditions atmosphere is characterized by negative vertical temperature gradient $G = dt/dH$ (usually it is -1°C per 100 m of height).

The change of stream pollution concentration is determined by the wind of average strength (dilution on the stream way), by turbulence (forced convection on directions perpendicular to the average speed) and by buoyancy characteristics (free convection) in vertical direction. Reynolds number give some certainty to these processes (as criterion of turbulence) and the criterion of Richardson Ri .

When using the Richardson criterion Ri , they differentiate stable ($Ri > 1$), unstable ($Ri < 1$) and neutral conditions on which Ri approximately is equal to destabilization and stabilization of atmosphere and become more specified if the middle wind is decreased. The pollution raising is slowed down by gradual decrease of difference in density of the jet and the ambient air. The combination of the described phenomena leads to the formation of pollution dome at the upper height of the jet. The process of formation of the dome will be more distinct if it comes to the counter-cyclical process "top-down" on certain synoptic conditions.

4.1 Dynamic part of the model.

Mathematical model, which would consider all the processes of formation of secondary pollution of atmospheric air with formaldehyde over the highways, consists of two blocks which describe accordingly the dynamics and kinetics of the given process.

The dynamic part of the model defines the basic parameters of the polluted layer of warm air, which is formed over highways and crossroads. The theory of convective jet is used by the authors when the polluted air layer is formed.

The amount of heat in relation to the highway or crossroad S , m with conditional diameter D , is determined by the dependence:

$$Q_s = \pi D^2 / 4 (R_a + R_c + R_t) Q_s, \quad (1)$$

when R_a , R_c and R_t - accordingly, are the scattered and direct radiation of the area which is under consideration for the particular month, and the heat which is emitted from automotive emissions at the crossroad, MJ/m^2 .

The heat emitted by cars we can obtain with the dependence:

$$Q_a = q_k \cdot N \cdot 40000 \text{ J} \quad (2)$$

where q_k - fuel consumption of one car for 1 m of the road, l .

By the number of lines, we define the number of cars N . In peak hours we should make an assumption according to the interval between cars $L = 20 \text{ m}$, and during this period there are 40 cars per 1 km.

$$N = n \cdot L' / (l_{auto} + L) \quad (3)$$

when l_{auto} - is the length of the car, m ; n - number of car lines on the road, L' - the Leghorn of one line, m .

According to the source [16] we can find the typical composition of exhaust fumes (hydrocarbons $CH_{1,85}$) by the engine type. So, for carburetor engine this value is $7,5 \frac{g}{kW \cdot h}$.

When calculating average emission by vehicles we accept the average capacity of one car as 100 kW and even arrival and departure of cars on transport hub at a speed of 60 km/h.

The difference between average air temperature on the surface of the transport hub and the narrowest section of thermal jet that is raising upward as well as the average air speed we can obtain by the formulas of convective heat transfer.

$$\Delta t_{ycp} = \frac{41 \cdot Q_k^{2/3}}{(y-y_0)^{5/3}} \text{ grad}, \quad (4)$$

$$V_y = 0,56(Q_k/y - y_0)^{0,33} \quad (5)$$

where $Q_k = R_a + R_c + R_t$, MJ/m²,

$y-y_0$ - the distance from the earth surface to the narrowest thermal convective jet that is raising upward, m.

Hydrocarbons concentration at the given number of vehicles C_{CH} we can obtain in the narrowest section of the polluted jet.

4.2 Kinetic part of the model.

The second part of the model refers to photochemical transformations of hydrocarbons (C_2H_4) of different origin, which are described in detail in many studies [7,8,18]; for example, methane, hydrocarbons of biogenic origin - isoprene and alkenes which are included into emissions of internal combustion engines.

Attribution of emissions in the atmosphere to a particular structural constituent needs to take into account number of factors of the given area including climatic, relief, and chemical ones. As for formaldehyde, apparently it can be treated to as ozone or peroxyacetyl nitrate (PAN) if emissions develop as in IV way [12].

The background concentrations of CHOH are assume as such that are formed by the constant factors: soil (CH) and vegetation (C_2H_4).

The correlation connection between concentration of CO and CHOH in the air near the highways and crossroads confirms the presumption of many scientists about the fact that formaldehyde concentration in the air of some big cities can serve as an indicator of photochemical smog emergence in the air over the highways with big vehicles density [4,7].

Formaldehyde concentration (secondary pollution) we can write as the following equation:

$$C_{CHOH} = K \cdot C_{CH} \quad (6)$$

when K - coefficient which characterizes the reaction speed, or which is a constant of transformation and depends on air temperature, time interval, sunlight intensity etc.

Basing on the laws of chemical kinetics which are described in detail in the studies [7,8,18], the molecule of gas can react only when they approach each other for direct energy exchange, that can lead to the destruction of bonds and rearrangement of atoms. As the two molecules collision is a necessary condition

of the reaction for chemical bonds breakdown there must be enough of energy. The speed of reaction is determined by the formula which includes the frequency of collisions as well as the share of the required energy excess.

$$K = A(T) \cdot \exp(-E/RT), \quad (7)$$

when K - constant of the reaction speed, h^{-1} ; T - absolute temperature, K ; R - unified gas constant, $\text{J}/(\text{mole} \cdot \text{K})$; E - energy of activation, constant of the given reaction J/mole .

In the formula (1) the predefined parameter $A(T)$ characterizes the frequency of collision of the number of reacting molecules and it is considered to be a constant as there is a weak dependence from the temperature (estimation of this parameter in the temperature range from 200°C to 300°C leads to the change of collision frequency A only by 10%). According to [23] the dependence of constant of reaction speed from temperature in general case is described by differential Vant-Hoff-Arrhenius equation which integral solution has the following form:

$$\ln K = -(E/RT) + C \quad (8)$$

when C - constant of integration.

The mentioned equation allows to determine effective energy of activation of the process of molecules transformation basing on monitoring data.

5. Research results.

Linear approximation of the dependence (8), which is based on experimental data, allows to find the average value of integrating constant by standard method (least squares method) and to determine analytical form of the dependence of speed constant K .

Analytical form of the speed constant dependency for Kyiv city is:

$$\ln K = -3784,6/T + 8,96 \quad (9)$$

Calculations of the most polluted highway near the Central Bus station in Kyiv are shown in table 1.

Table 1 Calculations and maximum monthly concentrations of atmospheric air pollution with formaldehyde in Kyiv.

No.	Temperature, T	Calculated concentration, C_p	Actual average monthly concentration, C_a	Maximum concentration, C_{max}
1	231,84	0,007	0,006	0,007
2	238,86	0,009	0,007	0,007
3	242,84	0,012	0,00	0,011
4	250,06	0,015	0,011	0,012
5	252,24	0,019	0,016	0,017
6	256,17	0,022	0,018	0,022
7	259,96	0,024	0,019	0,024
8	256,63	0,018	0,015	0,018
9	253,47	0,015	0,009	0,014
10	243,47	0,009	0,004	0,01
11	235,59	0,006	0,004	0,008
12	235,14	0,006	0,005	0,005

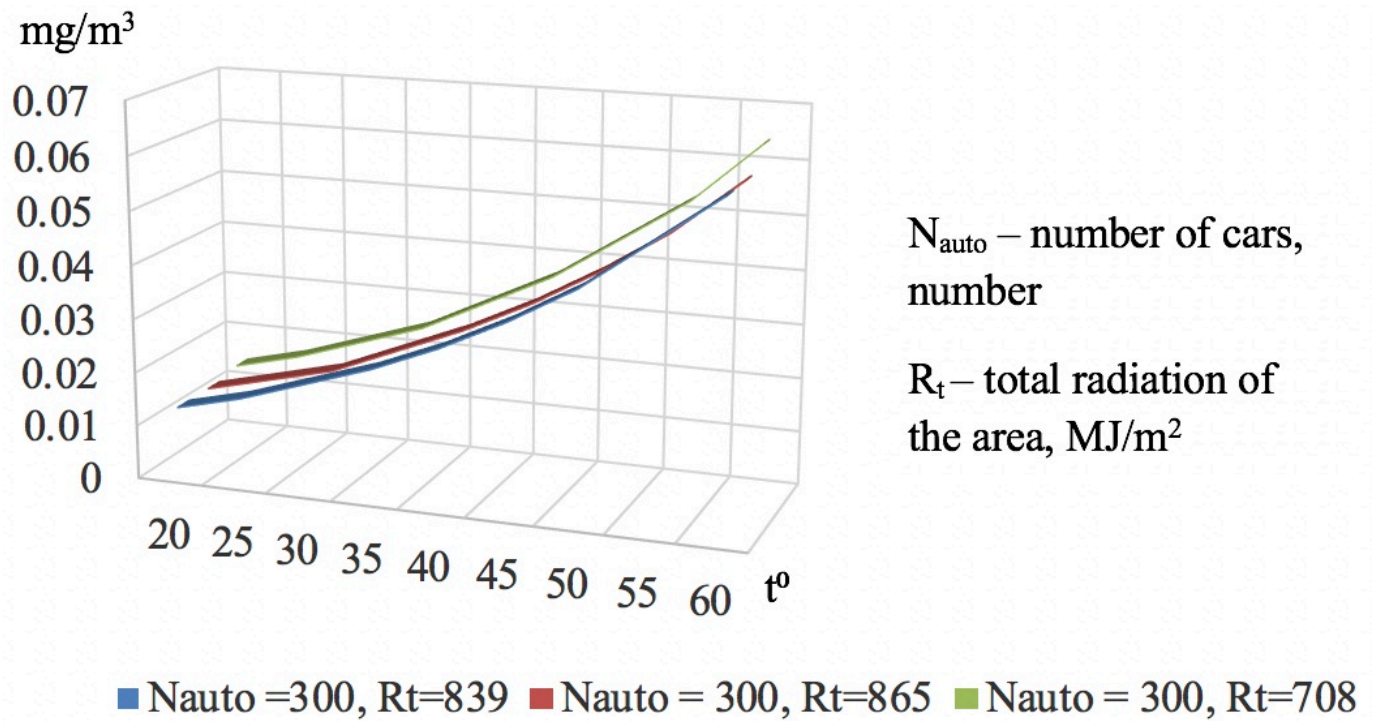
So, for Kyiv city the calculations by the method showed that the possible error with the maximum monitoring value of formaldehyde doesn't exceed 5%. As for comparison of the average monthly values of CHO₂ concentration in the atmospheric air - we observe the excess of the calculated values over the actual ones that can reach +20% in summer months. This error can be explained in such way: the main process of smog formation is emerging on the height of 200 m and more when the monitoring measurements of the magnitude C_{CHO_2} are occurring in the lower layer of atmospheric air on the height of 2m above the surface. This positive error has variation nature and it is also typical for a number of big cities.

The authors have made a useful calculator in Excel program for calculation of concentration of pollution from vehicles.

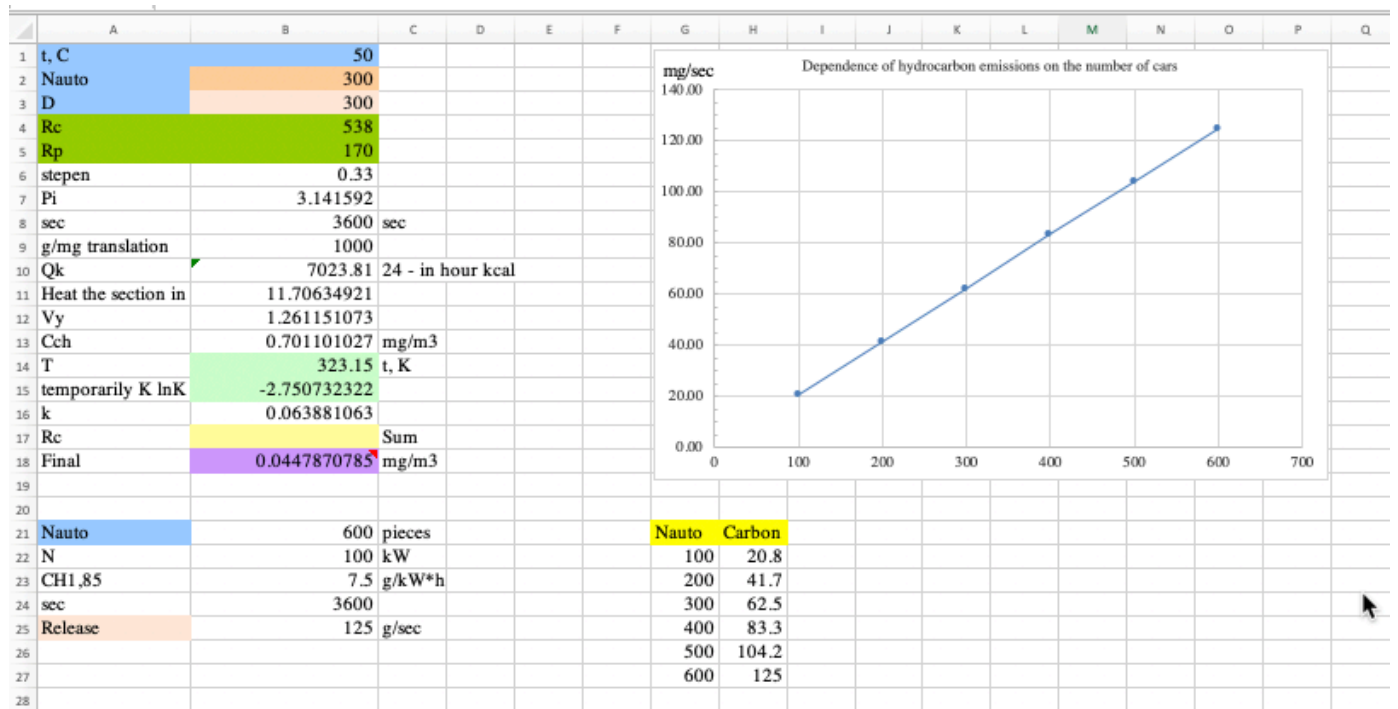
It allows to avoid mistakes caused by human factor and to quickly obtain the value of pollution concentration depending on the season, solar radiation and number of vehicles on the crossroad. Using the calculator, we can make experimental and scientific research on some unknown characteristics and get new knowledge on these characteristics by the planned monitoring of them, use the synthesized external influences with further processing of the results and further analysis of the consequences.

In our case we changed the output parameters and received variation data on impact on atmospheric air at different loads of traffic flow.

At pic.2 a screenshot of the calculator is presented with the dependence of GHG emission from amount of traffic on the road. This dependency allows us to forecast the personal transport deposit increasing in the context of global climate change.



Pic. 1. Impact of air temperature on concentration of secondary pollution with formaldehyde depending on amount of total radiation from highway surface.



Pic.2 Dependence of hydrocarbons from number of vehicles on highway.

So, the modern urbanization and global climatic changes make substantial interaction in the part of increased density of anthropogenic thermal streams over highways and the formation of the city polluted dome of heat.

6. Discussion of results of the research of the secondary pollution of atmospheric air with formaldehyde method depending on temperature conditions.

Review of the results of monitoring studies allowed to detect logical processes on distribution of atmospheric air pollutants over urbanized territories. For big cities the most powerful pollutant is formaldehyde. This fact caused the necessity to single out photochemical transformation and to define the structure of possible variants of transfer. The formation of formaldehyde as prevailing factor had the following necessary conditions: the presence of predecessors (volatile hydrocarbons of different origin), atmosphere temperature (annual temperature and the warm period temperature), solar radiation (periods of clear and cloudy days).

For calculation of emissions in megacities it is necessary to sum up the emissions over all big crossroads and highways. The very construction of the model involves such calculation path as the model's condition includes an assumption about large accumulation of vehicles and their moving with low speed, and the number of vehicles which is included in the calculation - is the amount of vehicle which is present at the same time at the area $S \text{ m}^2$.

The obtained results of the modelling of atmospheric air pollution basing on the model of two parts (kinetic and dynamic) allow to make the estimation and forecast of interaction of temperature conditions of a city, solar radiation and concentration of formaldehyde which is formed as a result of photochemical reactions over the highway. The suggested in this study calculator allows to find optimal managerial decisions on municipal level for many scientific and practical tasks of the city, such as: estimation of GHG emission by vehicles; influence of the emissions in the context of climatic changes; calculation of ecological safety level of the city and risks for the population health, especially in residential areas; development of norms and technological solutions on climatic changes consequences softening and improving of air quality; development of plans and concepts of socio-economical development of the city.

With this we should mention the positive error between the calculations of formaldehyde concentration in the narrowest section of the polluted jet and monitoring data of the lower air layer. This error is explained by the fact that photochemical transformation of hydrocarbons take place on conditions of emission into the atmosphere opened to solar radiation on the height of more than two conditional diameters of the heat surface over which the warm jet of polluted air is rising. This fact reveals the possibility of usage of the calculation results of the secondary pollution with some "stock". We should also mention the necessity

of further investigations on other ways of emission in the context of pollution of the atmosphere and the development of the issue of estimation and forecast of photochemical smog emergence over the highways depending on main factors of the process, and, first of all, on temperature conditions. In further researches the work of the calculator in urbocenosis of different climatic zones should also be considered and there should be detected the algorithm of softening measures on climate changes because of GHG emission by vehicles.

Conclusions

So, the suggested mathematical model for estimation and forecast of atmospheric air pollution from vehicles has united two tasks, which of them can be solved with the help of separate part (dynamic and kinetic). The first part gives the estimation of hydrocarbons emission by the vehicles with the help of solving of convective jet theory equation, the second one gives the estimation of the secondary pollution of the atmospheric air with formaldehyde on stable meteoconditions and depending on air temperature and solar radiation of the earth surface.

Comparing of the calculating results using this model and the regular monitoring data in Kyiv city gave positive results (5%) with the maximum measured concentrations. The exceeding of the calculated values of CHO₂ over the annual monthly ones gives positive excess (which can reach 20% in some summer months) This fact points at the necessity to monitoring improvement with expansion of permanent observation posts.

The suggested calculator gives an opportunity not only to support the managerial decisions on development of rational ecological policy of the city at municipal level, but it also gives an opportunity to optimize the measures for reducing of urbanized territories impact on human's health and climate. In addition, with the help of the calculator you can estimate the population health risk and conduct identification of danger of atmospheric air pollution.

Dynamic part of the model allows to get the "deposit" from vehicles into the GHG emission in the context of the city as well as in the context of total pollution of the atmospheric air with formaldehyde.

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