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THE MULTIBOX REACTIVE PLUME MODEL WITH VARIABILITY OF METEOROLOGICAL PARAMETERS TAKEN INTO ACCOUNT. PART I. MODEL FORMULATION

A multibox reactive plume model for simulation of the behaviour of chemically reactive pollutants in a plume emitted from a single, high-level point source is formulated. The model allows us to take into account the change of meteorological parameters in time. The model development and application are presented in two parts: part I – model formulation and part II – simulation of the behaviour of a reactive plume. In this paper, the model formulation is described.

1. INTRODUCTION

At present, fossil fuels are basic energy resources which can enable us to cope with the increasing demand for electric power.

Emissions from the power plants burning fossil fuels consist of large quantities of gaseous pollutants. Nitrogen oxides and sulphur dioxide are the main constituents of stack gases.

The pollutants forming stack plume while dispersing in the atmosphere react with all the species present in the ambient air. This concerns also the pollutants released from other emission sources. The evolution of the power plant plumes is strongly influenced by the presence of hydrocarbons in the atmosphere. Under the sunshine and warm weather conditions complex chemical reactions occurring in the stack plumes result in the formation of the secondary species such as ozone, PAN, nitric

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and sulphuric acids. These secondary pollutants are now thought to be more harmful to the environment and people health than their precursor gases.

Gaussian plume models are not appropriate to describe the characteristics of chemically reactive plumes emitted from point sources as their use is restricted to inert and linearly reactive pollutants.

It is also difficult to simulate the transport, dispersion and chemistry of emissions released from point sources using the conventional numerical grid models, in which the chemical submodel is built in, due to small and spatially changeable size of the plumes in comparison to the model grid cell.

A different modelling approach has to be used to treat the reactive plumes emitted from the point sources. The study described below is one of such undertakings. This study follows the approaches outlined by STEWARD and LIU [5], as well as DERWENT and HOV [18].

The model development and its application are presented in 2 parts:

1. Formulation of the model.
2. Simulation of the behaviour of a reactive plume.

In this paper, the model formulation is presented. First, model assumptions are characterised. Next, the basic mathematical relations are described. Finally, the possible model applications are discussed.

2. MODEL FORMULATION

2.1. BASIC ASSUMPTIONS

The model developed simulates the behaviour of reactive pollutants in a plume released from a single, high-level point source. It takes into account change of meteorological conditions in time. It is designed to study the transport, dispersion and chemistry of reactive plumes within the distances of few tens of km from the stack.

The model allows calculation of instantaneous concentrations of pollutants in the cross section of the plume within the Lagrangian framework, i.e. as the plume moves downwind. The concentrations calculated represent spatial averages over the subdivision of the plume cross section.

The formulation of the developed model bases on a concept of the multibox reactive plume model. The model employs the following assumptions [14]:

1. Meteorological conditions are spatially uniform. Change of meteorological conditions takes place in time every time step Δt . The following parameters undergo change: an atmospheric stability, an air temperature, a mixing layer and a wind velocity. In this version of the model a straight wind trajectory and the case of an expanding mixing layer are considered.

2. Emissions of pollutants from a point source and release parameters change in time in the way similar to the meteorological conditions, i.e. with every step Δt , and they are constant over the time interval Δt .

3. Primary pollutants released from the point source during the interval Δt are represented by the cross section of the plume. This implies that the continuous plume is approximated by means of the series of plume cross sections released regularly at the beginning of the time step Δt . In this version of the model only one plume cross section is traced as the plume moves downwind.

4. The plume cross section is visualised as an array of boxes perpendicular to the plume centreline. Its horizontal and vertical dimensions are bounded by 2.15 of the horizontal and vertical plume dispersion coefficients counting from the plume axis. The height of the plume centreline is determined by the height of effective emission. The boxes have equal volumes. During the initial time interval Δt chemical reactions in the plume are neglected. The initial distribution of the primary pollutants in the plume cross section in the horizontal direction follows the Gaussian characteristic. The material is uniformly distributed in each box. The schematic representation of this concept is given in figure 1.

5. Every time step Δt the array of boxes is transported along the plume trajectory. The vector of translation is determined by the mean wind. As the plume moves downwind the horizontal and vertical dimensions of the array expand and so do the boxes. The schematic representation of the expansion of the plume as the plume travels downwind is shown in figure 2.

6. The exchange of the material across the plume boundaries is the effect of two processes: the expansion of the box into another one and the horizontal diffusion. Values of the coefficients of eddy diffusivities are adjusted such that for the inert species there is no net flux across the box boundaries, i.e. the box boundaries act like impermeable membranes.

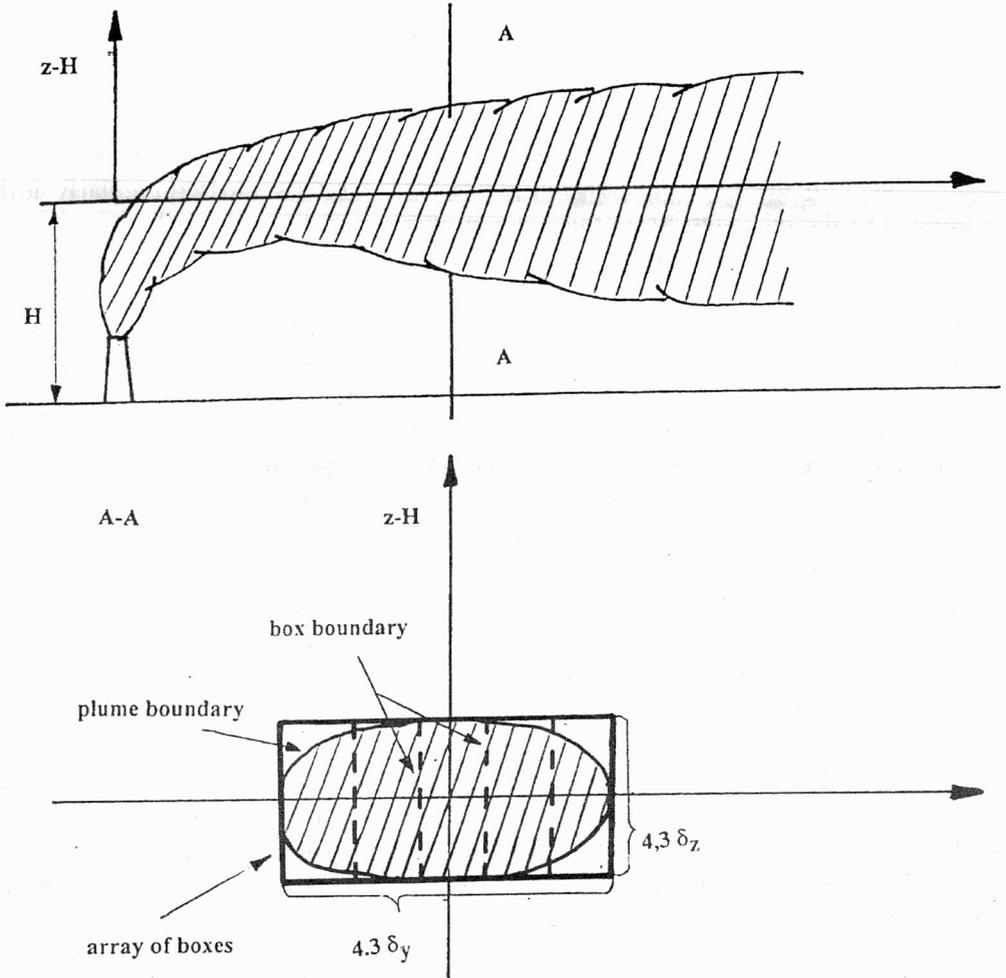
7. The ground limits the vertical dispersion of pollutants at the plume bottom. If the plume touches the ground the pollutants from ground level area are injected into the plume. The material expands vertically over the whole mixing layer across the whole depth of the plume.

8. The upper inversion layer limits the vertical dispersion of the pollutants at the upper part of the plume. The growth of the inversion layer allows for the further vertical dispersion of the plume.

9. The ambient air enters into the plume from the left and right edges as well as from the top and bottom of the plume. Ambient concentrations of chemical species in the mixing layer or the layer aloft, i.e. above the mixing layer, are calculated using a simple model.

10. Possible sinks of the pollutants in the atmosphere comprise: chemical gas-phase reactions and dry deposition. The dry deposition of the material takes place beyond the point where the plume reaches the ground.

11. Emissions of primary pollutants from ground level area sources vary spatially.



1. Schematic representation of the plume in the MRPM model. The plume cross section is visualized as an array of equal-volume boxes whose dimensions in horizontal and vertical directions are bounded by 4.3 of the horizontal and vertical dispersion coefficients, respectively. The initial distribution of the primary pollutants in the cross section of the plume follows the Gaussian characteristic. The material is uniformly mixed in each box.

12. Change of concentrations in the cross section of the plume as the plume travels downwind is calculated based on the set of mass conservation equations. Terms which account for the horizontal and vertical expansion of the plume, the entrainment of the ambient air to the plume as well as the chemical reactions and dry deposition of pollutants are included. There is one equation per species per box.

The plume cross section is defined by the following parameters:

- a) localisation of the plume cross section,
- b) height of the plume centreline,
- c) width of the plume,
- d) heights of the upper and lower edges of the plume,
- e) height of the mixing layer,
- f) rates of the expansion of the plume in the vertical and horizontal directions,
- g) rate of the expansion of the mixing layer,
- h) wind velocity averaged over the layer bounded by the ground and the height of plume centreline.

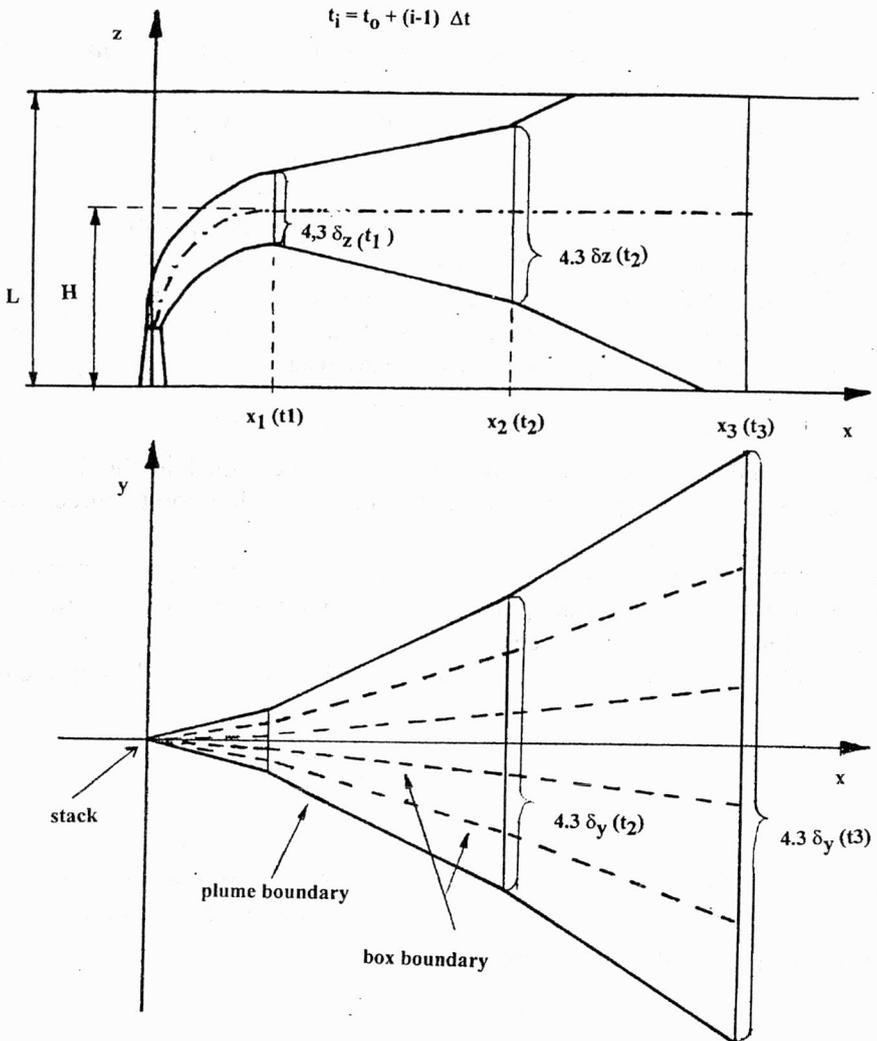


Fig 2. Schematic representation of the plume expansion in the MRPM model as plume travels downwind

The time interval Δt , during which the steady state is assumed, has the value of 15–30 minutes.

The model developed, which fulfils the assumptions stated above, is called MRPM (multibox reactive plume model).

2.2. MATHEMATICAL DESCRIPTION

2.2.1. STAGES OF CALCULATION PROCEDURE

Determination of the concentrations of pollutants in the plume cross section as the plume moves downwind is a 2-stage procedure.

The first stage involves calculation of the initial distribution of primary pollutants in the plume cross section and estimation of the parameters of the plume cross section. These parameters are estimated at the time: $t_{l+1} = t_0 + l\Delta t$, where t_0 is the time of the release of the cross section and l is the number of time intervals Δt , $l = 0, 1, 2, \dots (n - 1)$.

In the second stage, proper calculations are done, i.e. the distribution of concentrations of reactive species across the plume as the function of the travel time are determined. Values of the plume cross section parameters at the time: $t_l < t < t_{l+1}$ are estimated based on the values of these parameters at the time t_l and t_{l+1} . For most of the parameters the linear interpolation formulae are used.

3.2.2. LOCALISATION OF THE PLUME CROSS SECTION

The localisation of the plume cross section is determined in the Cartesian coordinate system XOY , in which the origin is connected with a point source, and the x axis is parallel to the wind direction.

Taking into account that in this version of the model a straight wind trajectory is considered, the localisation of the plume for a given wind direction is defined by one parameter, which is a travel distance.

The travel distance, S_{l+1} at the $l + 1$ time interval Δt is calculated using the expression:

$$S_{l+1} \equiv x_{l+1} = x_l + \bar{u}_{l+1} \Delta t \text{ [m]} \quad (1)$$

$$x_0 \equiv 0$$

where x_l, x_{l+1} are the x -co-ordinates of the plume cross section at the time t_l and t_{l+1} , respectively [m]; \bar{u}_{l+1} is the mean wind velocity during the $(l+1)$ time interval [m/s], and Δt is the duration of the time interval [s].

2.2.3. HEIGHT OF THE PLUME CENTRELIN

The height of the plume centreline is determined by the height of the effective emission H

$$H = h + \Delta h \text{ [m]} \quad (2)$$

where h is the stack height [m] and Δh is the plume rise [m].

In the case of stacks whose heat emission is less than 20 MJ/s, the Holland formula is used [15] and for emission sources whose heat emission is greater, the CONCAWE formula is applied [1].

The height of the plume centreline is calculated based on the meteorological data for the time interval Δt during which the pollutants are released from the stack, i.e. for the time: $t_0 < t < t_1$. The height of the plume centreline does not change as the cross-section moves downwind.

2.2.4. WIDTH OF THE PLUME

The width of the plume, w_{l+1} , at the $l + 1$ time interval is calculated from the expression:

$$w_{l+1} = w_l + \alpha_{w,l+1} \Delta t \text{ [m]} \quad (3)$$

where $\alpha_{w,l+1}$ is the plume expansion rate in the horizontal direction during the $(l + 1)$ time interval [m/s].

The value of the rate $\alpha_{w,l}$ is determined based on the values of the horizontal dispersion coefficients, $\delta_{y,l}$, $\delta_{y,l+1}$, at the time t_l and t_{l+1}

$$\alpha_{w,l+1} = 4.3 (\delta_{y,l+1} - \delta_{y,l}) / \Delta t \text{ [m/s]}. \quad (4)$$

2.2.5. HEIGHTS OF THE UPPER AND LOWER EDGES OF THE PLUME

The heights of the upper and lower edges of the plume, $h_{u,l+1}$ and $h_{g,l+1}$, at the $(l + 1)$ time interval are calculated from the formulae:

$$h_{u,l+1} = h_{u,l} + \alpha_{u,l+1} \Delta t \text{ [m]}, \quad (5)$$

$$h_{g,l+1} = h_{g,l} - \alpha_{g,l+1} \Delta t \text{ [m]} \quad (6)$$

where $\alpha_{u,l+1}$ and $\alpha_{g,l+1}$ are the plume expansion rates for the upper and lower plume edges during the $(l+1)$ time interval, respectively [m/s].

The value of the rate $\alpha_{u,l}$ for the time: $t < t_u$, where t_u is the time when the plume touches the inversion layer, is calculated based on the values of vertical dispersion coefficients $\delta_{z,l+1}$, $\delta_{z,l}$ at the time t_l and t_{l+1} . Afterwards $\alpha_{u,l+1}$ is estimated based on the values of the mixing layer L_{l+1} , L_l .

$$t < t_u: \alpha_{u,l+1} = 2.15(\delta_{z,l+1} - \delta_{z,l})/\Delta t \text{ [m/s]}, \quad (7)$$

$$t \geq t_u: \alpha_{u,l+1} = (L_{l+1} - L_l)/\Delta t \text{ [m/s]}. \quad (8)$$

The value of the rate $\alpha_{g,l+1}$ for the time $t < t_g$, where t_g is the time when the plume touches the ground, is calculated based on the values of the vertical dispersion coefficients, $\delta_{z,l+1}$ and $\delta_{z,l}$ at the time t_l, t_{l+1} . Afterwards $\alpha_{g,l}$ is equal to 0:

$$t < t_g: \alpha_{g,l+1} = 2.15(\delta_{z,l+1} - \delta_{z,l})/\Delta t \text{ [m/s]}, \quad (9)$$

$$t \geq t_g: \alpha_{g,l+1} = 0. \quad (10)$$

2.2.6. ATMOSPHERIC DISPERSION COEFFICIENTS

The dispersion coefficients in the vertical and horizontal directions, δ_y and δ_z , are determined based on Nowicki's formulae [16]:

$$\delta_y = Ax^a \text{ [m]}, \quad (11)$$

$$\delta_z = Bx^b \text{ [m]} \quad (12)$$

where x is the travel distance of the plume from the point source [m]; A, B are the coefficients which depend on the atmospheric stability, effective height of emission and surface roughness, and a, b are the coefficients depending on the atmospheric stability.

To include the change of meteorological conditions as the plume travels downwind, an effective travel distance x_{eff} in the place of the travel distance x is introduced. The value of x_{eff} is estimated based on the formula [13]:

$$x_{\text{eff},l} = (\delta_l/G_{l+1})/g_{l+1} \text{ [m]} \quad (13)$$

where δ_l is the dispersion coefficient in vertical or horizontal direction at the time t_l ; G_{l+1}, g_{l+1} are the constants A, a or B, b at the $(l+1)$ time interval.

At each $(l+1)$ time interval the dispersion coefficients are calculated using the virtual distance concept:

$$\delta_{l+1} = G_{l+1}(x_{\text{eff},l} + \bar{u}_{l+1} \Delta t) g_{l+1} \text{ [m]}. \quad (14)$$

At the time $t = t_0$ it is assumed that $\delta_0 \equiv 0$.

For the treatment of relative diffusion only, the dispersion coefficients δ_y, δ_z are multiplied by the rate $r = 0.75$ which accounts for the reduction of the values of the dispersion coefficients [17].

2.2.7. LIMITATIONS OF THE VERTICAL DISPERSION OF THE PLUME

The dispersion of the plume in a vertical direction is limited by the ground and the upper inversion layer. This implies that the minimum height of the lower plume boundary is $h_g = 0$ and the maximum height of the upper plume boundary is $h_u = L$,

where L is the depth of the mixing layer. The value of the depth of the mixing layer at the time $t_l < t < t_{l+1}$ is interpolated linearly, i.e.

$$L = L_l + (L_{l+1} - L_l) (t - t_l) / \Delta t \text{ [m]} \quad (15)$$

where L_l, L_{l+1} are the depths of the mixing layer at the time t_l, t_{l+1} , respectively.

2.2.8. MEAN WIND VELOCITY

The mean wind velocity, i.e. averaged over the layer bounded by the ground surface and the height of the effective emission, is calculated from the formula [2]:

$$\bar{u} = u_a / (m+1) (H/h_a)^m \text{ [m/s]} \quad (16)$$

where u_a is the wind velocity at the height h_a [m/s], and m is the meteorological coefficient.

2.2.9. PARAMETERISATION OF CHEMICAL REACTIONS

The parameterisation of chemical transformation processes is based on the scheme originally developed by DERWENT and HOV [4], [5] and simplified by HOUGH [8], [9]. It describes the homogeneous gas-phase processes. The chemical system includes: CO, NO_x, SO₂ and representative hydrocarbons among the primary species. The overall number of chemical reactions exceeds one hundred. The reaction which is responsible for the transformation of NO to NO₂ in the case of high NO concentrations has been added [11].

Lists of all inorganic reactions and initial organic reactions and the associated rate coefficients used in the model are presented in table 1 and table 2.

Table 1

Inorganic reactions and rate coefficients in the MRPM model

Chemical reaction Inorganic chemistry	Rate coefficient [cm ³ /(molecule s)]
1	2
O(3p) + O ₂ + M → O ₃ + M	5.8 × 10 ⁻³⁴ (T/300) ^{-2.6}
O(3p) + NO ₂ → NO	9.0 × 10 ⁻³²
O(3p) + NO ₂ + M → NO ₃	9.3 × 10 ⁻¹²
O(3p) + NO + M → NO ₂ + M	1.1 × 10 ⁻³¹ (T/300) ^{-1.8}
O(¹ D) + M → O(3p) + M	2.1 × 10 ⁻¹¹ exp(+100/T)
O(¹ D) + H ₂ O → 2OH	2.2 × 10 ⁻¹⁰
O ₃ + NO → NO ₂ + O ₂	1.8 × 10 ⁻¹² exp(-1370/T)
O ₃ + NO ₂ → NO ₃ + O ₂	1.2 × 10 ⁻¹³ exp(-450/T)
O ₃ + OH → HO ₂ + O ₂	1.9 × 10 ⁻¹² exp(-1000/T)
O ₃ + HO ₂ → OH + 2O ₂	1.4 × 10 ⁻¹⁴ exp(-600/T)

1	2
$\text{NO} + \text{NO} + \text{O}_2 \rightarrow \text{NO}_2 + \text{NO}_2$	$3.3 \times 10^{-39} \exp(-530/T)$
$\text{NO} + \text{NO}_3 \rightarrow 2\text{NO}_2$	2.0×10^{-11}
$\text{NO} + \text{OH} + \text{M} \rightarrow \text{HNO}_2 + \text{M}$	4.0×10^{-12}
$\text{NO} + \text{NO}_2 + \text{H}_2\text{O} \rightarrow 2\text{HNO}_2$	8.2×10^{-39}
$\text{NO} + \text{HO}_2 \rightarrow \text{NO}_2 + \text{OH}$	$3.7 \times 10^{-12} \exp(+240/T)$
$\text{NO}_2 + \text{NO}_3 \rightarrow \text{NO} + \text{NO}_2 + \text{O}_2$	$2.3 \times 10^{-13} \exp(-1000/T)$
$\text{NO}_2 + \text{NO}_3 \rightarrow \text{N}_2\text{O}_5$	1.4×10^{-12}
$\text{NO}_2 + \text{OH} \rightarrow \text{HNO}_3$	1.5×10^{-11}
$\text{NO}_2 + \text{HO}_2 \rightarrow \text{HO}_2\text{NO}_2$	1.7×10^{-12}
$\text{HO}_2\text{NO}_2 \rightarrow \text{NO}_2 + \text{HO}_2$	$6.8 \times 10^{13} \exp(-10200/T)$
$\text{NO}_3 + \text{H}_2\text{O}_2 \rightarrow \text{HNO}_3 + \text{HO}_2$	4.1×10^{-16}
$\text{N}_2\text{O}_5 \rightarrow \text{NO}_2 + \text{NO}_3$	$8.5 \times 10^{14} \exp(-11080/T)$
$\text{N}_2\text{O}_5 + \text{H}_2\text{O} \rightarrow \text{HNO}_3 + \text{HNO}_3$	1.3×10^{-21}
$\text{OH} + \text{HO}_2 \rightarrow \text{H}_2\text{O} + \text{O}_2$	1.0×10^{-10}
$\text{OH} + \text{H}_2\text{O}_2 \rightarrow \text{HO}_2 + \text{H}_2\text{O}$	$2.9 \times 10^{-12} \exp(-160/T)$
$\text{OH} + \text{H}_2 \rightarrow \text{H}_2\text{O} + \text{H}$	$7.7 \times 10^{-12} \exp(-2100/T)$
$\text{OH} + \text{HNO}_2 \rightarrow \text{NO}_2 + \text{H}_2\text{O}$	6.6×10^{-12}
$\text{OH} + \text{HNO}_3 \rightarrow \text{NO}_3 + \text{H}_2\text{O}$	$9.4 \times 10^{-15} \exp(778/T)$
$\text{HO}_2 + \text{HO}_2 \rightarrow \text{H}_2\text{O}_2 + \text{O}_2$	$2.2 \times 10^{-13} \exp(600/T)$
$\text{HO}_2 + \text{HO}_2 + \text{M} \rightarrow \text{H}_2\text{O}_2 + \text{O}_2 + \text{M}$	$1.9 \times 10^{-33} \exp(980/T)$
$\text{HO}_2 + \text{HO}_2 + \text{H}_2\text{O} \rightarrow \text{H}_2\text{O}_2 + \text{O}_2 + \text{H}_2\text{O}$	$2.1 \times 10^{-38} \exp(5800/T)$
$\text{OH} + \text{SO}_2 \rightarrow \text{HSO}_3$	1.35×10^{-12}
$\text{CH}_3\text{O}_2 + \text{SO}_2 \rightarrow \text{SO}_3 + \text{CH}_3\text{O}$	4.0×10^{-17}
$\text{O}(3p) + \text{SO}_2 + \text{M} \rightarrow \text{SO}_3 + \text{M}$	$4.0 \times 10^{-32} \exp(-1000/T)$
$\text{HO}_2 + \text{SO}_2 \rightarrow \text{SO}_3 + \text{OH}$	1.0×10^{-18}
$\text{HSO}_3 + \text{O}_2 \rightarrow \text{SO}_3 + \text{HO}_2$	1.0×10^{-11}
$\text{SO}_3 + \text{H}_2\text{O} \rightarrow \text{sulphate aerosol}$	9.1×10^{-13}
$\text{N}_2\text{O}_5 \rightarrow 2 \text{ nitrate aerosol}$	3.0×10^{-5}
$\text{HNO}_3 \rightarrow \text{ nitrate aerosol}$	6.0×10^{-6}

Table 2

Inorganic reactions and rate coefficients in the MRPM model

Chemical reaction Initial organic chemistry	Rate coefficient [cm ³ /(molecule s)]
1	2
$\text{CH}_4 + \text{OH} \rightarrow \text{CH}_3 + \text{H}_2\text{O}$	$2.4 \times 10^{-12} \exp(-1710/T)$
$\text{CH}_3\text{OH} + \text{OH} \xrightarrow{\text{O}_2} \text{HO}_2 + \text{HCHO} + \text{H}_2\text{O}$	1.0×10^{-12}
$\text{HCHO} + \text{OH} \xrightarrow{\text{O}_2} \text{HO}_2 + \text{CO} + \text{H}_2\text{O}$	1.1×10^{-11}
$\text{HCHO} + \text{NO}_3 \xrightarrow{\text{O}_2} \text{HO}_2 + \text{CO} + \text{HNO}_3$	3.2×10^{-16}
$\text{CO} + \text{OH} \rightarrow \text{CO}_2 + \text{H}$	2.2×10^{-13}

	1	2
$C_2H_6 + OH \xrightarrow{O_2} C_2H_5O_2 + H_2O$		$1.9 \times 10^{-11} \exp(-1260/T)$
$CH_3CHO + OH \xrightarrow{O_2} CH_3COO_2 + H_2O$		$6.9 \times 10^{-12} \exp(260/T)$
$n-C_4H_{10} + OH \xrightarrow{O_2} s-C_4H_9O_2 + H_2O$		$1.64 \times 10^{-11} \exp(-559/T)$
$CH_3COC_2H_5 + OH \xrightarrow{O_2} CH_3COCHO_2CH_3 + H_2O$		8.8×10^{-13}
$o\text{-xylene} + OH \xrightarrow{O_2} \text{additional product}$		1.1×10^{-11}
$\text{But-2-ene} + OH \xrightarrow{O_2} CH_3CHOHCHO_2CH_3$		$1.12 \times 10^{-11} \exp(549/T)$

In all cases, the values of the photolysis rates, J , are parameterised as a function of the solar zenith angle θ

$$J = a_{ph} \exp(b_{ph} \sec\theta) \quad (17)$$

where a_{ph} , b_{ph} are the coefficients of photolysis rate.

The list of photochemical reactions and the associated rate coefficients used in the model are presented in table 3.

Table 3

Photochemical processes and coefficients of photolysis rate in the MRPM model

Process	Photolysis rate coefficient	
	a_{ph}	b_{ph}
$O_3 \rightarrow O(3p)$	1.23×10^{-3}	-0.6
$O_3 \rightarrow O(^1D)$	2.0×10^{-4}	-1.4
$H_2O_2 \rightarrow 2OH$	2.2×10^{-5}	-0.75
$HNO_3 \rightarrow NO_2 + OH$	3.0×10^{-6}	-1.25
$NO_2 \rightarrow NO + O(3p)$	1.45×10^{-2}	-0.4
$N_2O_5 \rightarrow NO_2 + NO_3$	3.324×10^{-5}	-0.567
$HCHO \rightarrow H + CO + HO_2$	5.4×10^{-5}	-0.79
$HCHO \rightarrow H_2 + CO$	6.65×10^{-5}	-0.6
$NO_3 \rightarrow NO + O_2$	3.527×10^{-2}	-0.0809
$NO_3 \rightarrow NO_2 + O(3p)$	8.939×10^{-2}	-0.0591
$CH_3CHO \rightarrow CH_3 + HO_2 + CO$	1.35×10^{-5}	-0.94
$CH_3COC_2H_5 \xrightarrow{O_2} CH_3COO_2 + C_2H_5O_2$	2.425×10^{-5}	-0.877
$CH_3COCOH_3 \xrightarrow{O_2} 2CH_3COO_2$	5.4×10^{-4}	-0.79

2.2.10. DRY DEPOSITION PARAMETERISATION

The parameterisation of dry deposition processes is based on the deposition velocity v_d . The change of concentration due to deposition velocity in the vertically well-mixed plume is described by the following expression

$$P_i^j = C_i v_{d,i} / h_p \text{ [molecules/cm}^3 \text{ s]} \quad (18)$$

where C_i is the concentration of the species i [molecules/cm³ s] and h_p is the depth of the plume [m].

The deposition processes take place at the time $t > t_{gr}$, i.e. when the plume reaches the ground. The values of v_d of species: HNO₃, SO₂, NO₂, SO₂, O₃, PAN are presented in table 4. The values applied are based on the recommendation of GARLAND [6].

Table 4

Deposition velocities v_d used in the MRPM model

Species	Deposition velocity [m/s]
Ozone	0.006
Sulphur dioxide	0.008
Nitrogen dioxide	0.001
PAN	0.002
HNO ₃	0.008

2.2.11. PARAMETERISATION OF MASS TRANSFER ACROSS THE BOX BOUNDARIES

The transfer of net mass of pollutants across the expanding boundaries of the box j , E_i^j is expressed by the equation [4], [18]:

$$F_i^j = E_i^j - D_i^j \text{ [molecules/cm}^3 \text{ s]} \quad (19)$$

where D_i^j is the detrainment of mass of species i from the j -th box.

The amount of the mass of the species i , which entrains per unit volume per unit time into the box j is estimated based on the expression:

$$E_i^j = \alpha_{w,l+1} (jC_i^{j+1} - (j-1)C_i^j) / (n_b w_b) \text{ [molecules/cm}^3 \text{ s]} \quad (20)$$

where $\alpha_{w,l+1}$ is the plume expansion rate in the horizontal direction during the $(l+1)$ time interval [m/s]; C_i^{j+1} , C_i^j are the concentrations of the species i in the boxes $(j+1)$ and j , respectively [molecules/cm³ s]; n_b is the number of boxes in the plume cross section and w_b is the box width [m].

The amount of the mass of the species i , which entrains per unit volume per unit time from the box j , is estimated based on the expression:

$$D_i^j = 2 \left[K^j (c_i^{j+1} - c_i^j) - K^{j-1} (c_i^j - c_i^{j-1}) \right] / w_b^2 \quad [\text{molecules/cm}^3 \text{ s}] \quad (21)$$

where K^j, K^{j-1} are the eddy diffusion coefficients for the boxes j and $(j-1)$, respectively [m^2/s].

The values of K are calculated from the recursive formula which was developed by imposing the conditions that for an inert species there is no net mass transfer [17].

2.2.12. GOVERNING EQUATIONS

The governing equations of the model developed are based on the mass conservation principle. The terms which account for the horizontal and vertical expansion of the plume, the entrainment of the ambient air, chemical reactions and dry deposition of the pollutants are included in the equation. There is one equation per species per one box. The change of concentration of the species i in the box j, C_i^j is calculated based on the formula:

$$\begin{aligned} dC_i^j/dt = & (\alpha_{w,j+1}/w_b n_b) C_i^j - (\alpha_{u,j+1}/h_b) (C_i^j - C_{i,u}) \\ & - (\alpha_{g,j+1}/h_b) (C_i^j - C_{i,g}) + F_i^j + R_i^j + P_i^j + S_i^j \end{aligned} \quad (22)$$

where $C_{i,u}$ and $C_{i,g}$ are the concentrations of the species i above the upper and below the lower plume edge [molecules/m^3]; w_b and $h_b = h_p$ are the width and height of the box [m]; $\alpha_{w,j+1}$ is the plume expansion rate in the horizontal direction [m/s]; $\alpha_{u,j+1}$ and $\alpha_{g,j+1}$ are the plume expansion rates of the upper and lower edges of the plume, respectively [m/s]. The last four terms on the right side of the equation are expressed in [$\text{molecules/cm}^3 \text{ s}$]. The term F_i^j describes the mass transfer of the species i across the expanding box boundaries. The terms: R_i^j and P_i^j characterise the change of concentration of the species i due to chemical reactions and due to dry deposition, respectively. And finally, S_i^j is the flux of the species i from the ground level area.

3. IMPLEMENTATION OF THE RPMM MODEL

The MRPM model has been implemented in FACSIMILE language on the DEC station 3100 computer [3], [12]. The Gear algorithm [7] is used to integrate the set of governing differential equations. This algorithm allows us to treat accurately a system

of stiff equations, i.e. a system involving both rapidly changing terms and slowly varying terms.

4. MODEL APPLICATIONS

The MRPM model developed allows us to trace the evolution of chemically reactive plumes emitted from the point sources. The fact that the MRPM model takes into account the variability of meteorological conditions extends its application in comparison to the other reactive plume models described in the literature.

The model can be used to study the evolution of the reactive plumes to few tens of km from the stack during the daytime conditions with the sunshine and warm weather.

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WIELOPUDEŁKOWY MODEL SMUGI REAKTYWNEJ CHEMICZNIE UWZGLĘDNIAJĄCY
ZMIENNOŚĆ PARAMETRÓW METEOROLOGICZNYCH.
CZĘŚĆ I. SFORMUŁOWANIE MODELU

W pierwszej części artykułu przedstawiono założenia, podstawy matematyczne i praktyczne zastosowanie modelu smugi reaktywnej chemicznie. Model ten opisuje rozprzestrzenianie się reaktywnych chemicznie zanieczyszczeń, które są emitowane z punktowego źródła emisji. W drugiej części artykułu opisano wyniki symulacji zachowania się smugi przeprowadzonej z zastosowaniem opracowanego modelu.

