ENVIRONMENTAL POLLUTION AND DEGRADATION MODELS

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Contents

Summary

A numerical model for the transport of persistent organic pollutants (POP) in the Northern Hemisphere was developed. The model admits spatial and temporal variability of POP in the Northern Hemisphere to be studied on the basis of European-source emissions and aids assessment of the percentile distributions in different environmental media. The interaction of lindane with soil and water surface was simulated using surface and stratified atmospheric boundary layer models.

The transformation of POP in the atmosphere and soil was taken into account with the help of special modules describing its degradation in soil and atmosphere as well as accumulation in water. Also, considered are migration in soil and dry and wet deposition processes. The numerical calculations were performed for a period of one year, 1992, using the European Center for Medium Range Weather Forecasts (ECMWF) meteorological data and lindane emission data for Europe.

1. Introduction

A great deal of anthropogenic pollutants emitted into the atmosphere and toxic chemicals and mineral fertilizers applied to soils enter into the biogeochemical cycles with a considerable impact on them. Soil is the most vulnerable to the anthropogenic impacts, and an important factor in the biogeochemical cycles is the transformation of organic species. The emissions of gaseous species are subject to a series of physical and chemical transformations leading to their degradation in the environment. Among these, POPs are of particular importance. These pollutants are industrial chemical products, products and by-products of combustion having low volatility, high stability and bioaccumulation. Also, they have low solubility (influencing the adsorption processes in soil and on atmospheric aerosols) and are able to transport over long distances in the atmosphere. The importance of studying different aspects of POPs concerning emissions, properties, and modeling approaches is widely accepted.

There are a number of attempts to simulate the behavior of persistent organic pollutants both on the regional and global scales. Here we describe a tool for numerical modeling of the transport and transformation of a persistent organic pollutant (namely lindane) in the Northern Hemisphere. Lindane (the γ-isomer of HCH) is available in different forms and used generally as insecticide and fumigant. Lindane present in the atmosphere is subjected to processes of dry and wet deposition and degradation. It also interacts with water media through different types of transformations. In the air, lindane can persist for long time periods and degrade finally with the help of bacteria to less toxic species.

2. Mathematical Model for Global Transport of Persistent Organic Pollutants in the Northern Hemisphere

In order to assess the significance of the influence of remote pollution sources on the ecological state of a given region, the equations of pollutant transport in the atmosphere are used with due regard to the turbulent exchange and interaction of pollutant with the underlying surface.

We formulate the model assuming the Earth's surface to be spherical. A coordinate system (λ, ψ, z) is used where λ is the longitude, ψ is the complement of latitude and z is the altitude measured from the underlying surface. The main equation of pollutant transport on a sphere is expressed in the following form

$$
\frac{\partial \varphi}{\partial t} + \frac{u}{a \sin \psi} \frac{\partial \varphi}{\partial \lambda} + \frac{v}{a} \frac{\partial \varphi}{\partial \psi} + w \frac{\partial \varphi}{\partial z} =
$$

$$
F - P_1 \varphi - P_2 \varphi + \frac{\partial}{\partial z} \nu \frac{\partial \varphi}{\partial z} + \frac{1}{a^2 \sin^2 \psi} \frac{\partial}{\partial \lambda} \mu \frac{\partial \varphi}{\partial \lambda}
$$

$$
+ \frac{1}{a^2 \sin \psi} \frac{\partial}{\partial \psi} \mu \sin \psi \frac{\partial \varphi}{\partial \psi}.
\tag{1}
$$

Here $\varphi = \varphi(\lambda, \psi, z, t)$ is the pollutant concentration; $\mathbf{u} = (u, v, w)$ is the wind velocity

vector along λ, ψ, z directions, respectively; μ, ν are the horizontal and vertical turbulent exchange coefficients, respectively; $F = F(\lambda, \psi, z, t)$ is a function representing emission source magnitudes; P_1 describes the washout of lindane from the atmosphere; P_2 describes the degradation of lindane in the atmosphere; and a is the average radius of the Earth. To account for the topography of the terrain, we turn from the spherical coordinate systems (λ, ψ, z) to curvilinear one $(\lambda, \psi, \tilde{z})$: $\tilde{z} = z - \delta(\lambda, \psi)$, where $\delta(\lambda, \psi)$ is a function describing the topography of the terrain. Then the vertical velocity in the new system of coordinates $(\tilde{w})$ is expressed as follows: $\tilde{w} = w - u\delta_\lambda - v\delta_\psi$ where $u\delta_\lambda$ and $v\delta_\psi$ are slope angles. w is determined by the analog of the vertical velocity in the p-system of coordinates and geopotential.

Eq. (**??**) is considered in the domain $D_t = G \times [(0, T])$, where

$G = S \times [0, H])$; $S = \{(\lambda, \psi) : 0 \leq \lambda \leq 2\pi, \ 0 \leq \psi \leq \pi/2\}$, H is the upper boundary of the numerical grid domain.

The boundary conditions are given according to periodicity conditions on space coordinates:

$$\varphi(0, \psi, z, t) = \varphi(2\pi, \psi, z, t)$$

$$\varphi(\lambda, -\psi, z, t) = \varphi(\lambda + \pi, \psi, z, t)$$

$$\varphi(\lambda, \pi + \psi, z, t) = \varphi(\lambda + \pi, \pi - \psi, z, t) \tag{2}$$

$$\frac{\partial \varphi}{\partial \psi} = 0 \quad \text{for} \quad \psi = \pi/2$$

2.1. Parameterization of the Planetary Boundary Layer

The vertical spatial resolution in global atmospheric models is rather coarse and it is difficult to reconstruct the fields in the lower layers of the atmosphere with adequate accuracy. Therefore we calculate meteorological characteristics of the surface and boundary layers with the help of a parameterization scheme for the planetary boundary layer. With the help of the model of the planetary boundary layer the following outer parameters are determined at every point of the horizontal computational grid by known values of velocity and temperature at the first computational level:

$$\text{Ro} = \frac{|u_{g0}|}{1 z_0}, \quad \mathcal{S}_T = \frac{\beta \delta \hat{\theta}}{1 |u_{g0}|}, \quad \eta_x = \frac{\text{æ}^2}{1} \frac{\partial u_g}{\partial z} = -\frac{\beta \text{æ}^2}{1^2} \frac{\partial \hat{\theta}}{\partial y},$$

$$\eta_y = \frac{\text{æ}^2}{1} \frac{\partial v_g}{\partial z} = \frac{\beta \text{æ}^2}{1^2} \frac{\partial \hat{\theta}}{\partial x} \tag{3}$$

where $x = a\,\sin\psi\,\cos\lambda$; $y = a\sin\psi\,\sin\lambda$; Ro is the Rossby number; S_T is the stratification parameter; u_g, v_g are the components of the geostrophic wind; $\left|u_{g0}\right|$ is absolute value of geostrophic wind near the underlying surface; l is the Coriolis parameter; η_x, η_y are the parameters of baroclinity; $\hat{\theta}$ is the potential temperature; $\beta = g/\hat{\theta}$ is the buoyancy parameter; g is the gravity acceleration; and $\delta\hat{\theta}$ is the difference between potential temperature at the top of the planetary boundary layer and the underlying surface. Using the values of $\mathrm{Ro}, S_T, \eta_x, \eta_y$ find the following values: $\varphi_g = \mathrm{u}_* / \left|u_g\right|$ (the geostrophic resistance coefficient); α (angle between turbulent stresses near the underlying surface and u_{g0}); $\mu = \mathrm{h}_0 / \mathrm{L}_0$ (the dimensionless internal parameter of stratification).

The following notation is adopted here: $\mathrm{h}_0 = æu_*/l$ is the internal scale for the boundary layer height; $L_0 = -c_p\rho u_*^3 / æ\beta\mathrm{q}_0$ is the Monin-Obukhov length scale; c_p is the air specific heat at constant pressure; ρ is the air density; q_0 is the surface heat flux near the surface.

Using φ_g, α and μ, the value of q_0 is computed; then u_* (by φ_g and u_g) and the turbulent exchange coefficients at heights $z \geq H$ are determined as follows

$$\mathrm{v}(\mu) = \frac{æ^2 u_*^2}{l^2} \begin{cases} \mathrm{h}/(1+10\mu\mathrm{h}) & \text{for} & \mu \geq 0, \\ \mathrm{h} & \text{for} & -2.33 \leq \mu \leq 0, \\ (-0.07/\mu)^{-1/3}\,\mathrm{h}^{4/3} & \text{for} & \mu \leq -2.33, \end{cases} \qquad (4)$$

where h is the height of the surface layer.

The wind velocity components are determined using the so called "velocity defect" concept.

For the global transport, pollutants with small gravitational settling rates are of special interest. They remain in the atmosphere for long periods and are transported together with air masses. In this case, the principles of turbulence theory that are used in global circulation models for the description of turbulent exchange of heat and moisture could be applied to pollutants. In particular, the following model is used to determine the horizontal turbulent exchange coefficient:

$$\mu = æ_1^2 \Delta\mathcal{S} \,|\, \mathrm{D}_\mathrm{N}\,|, \mathrm{D}_\mathrm{N} = (\mathrm{D}_\mathrm{T}^2 + \mathrm{D}_\mathrm{S}^2)^{1/2},\ \mathrm{D}_\mathrm{T}$$
$$= \frac{1}{a\sin\psi}\frac{\partial u}{\partial\lambda} - \frac{\partial v}{2\partial\psi}, \mathrm{D}_\mathrm{S} = \frac{1}{a\sin\psi}\frac{\partial v}{\partial\lambda} - \frac{\partial u}{2\partial\psi}, \qquad (5)$$

where $\Delta\mathcal{S}$ is the area of an elementary grid cell and k_1 is a dimensionless parameter.

Now we describe the basic physical principles of lindane transformations in different media.

2.2. Flux of Lindane in the Atmospheric Surface Layer

Dry deposition is one of the important processes describing the distribution of atmospheric pollutants and particularly lindane in the system "atmosphere-soil-water". In the atmosphere it is influenced by a a number of factors including temperature stability of the atmosphere, type of pollutant, characteristics of the underlying surface. The dry deposition is described by the resistance of the pollutant with surface as a sum of the following three terms:

- Aerodynamic resistance (r_a) conditioned by the inner turbulence layer and depending essentially on the stability of the atmosphere;
- Quasi-laminar boundary layer resistance r_b, conditioned by the molecular diffusion process just near the earth's surface;
- Surface resistance (r_c) connected with the process on the deposition surface;

The surface flux of lindane can be written down as

$$\nu \frac{\partial \varphi}{\partial z} = \frac{1}{r_a + r_b + r_c}(\varphi - \varphi_{\text{surf}}) \tag{6}$$

where

$$r_b = \frac{2}{\text{æ} u_*}\left(\frac{S_c}{Pr}\right)^{2/3}$$

$$S_c = 1.5 \times 10^{-5}\,\text{m}^2\text{s}^{-1};\ Pr = 0.71 \text{ is the Prandtl number} \tag{7}$$

In what follows, we describe how r_a is determined. To set the boundary condition at $z = h$, the Monin-Obukhov theory is used stating that the turbulent flux of a pollutant (along with heat and momentum fluxes) can be taken constant by height. Then for $z \le h$ the following relationships hold

$$\frac{\partial \varphi}{\partial z} = \frac{\varphi_*}{z}\eta_\varphi\ (\zeta) \tag{8}$$

$$\varphi - \varphi_0 = \varphi_* \int_{\zeta_0}^{\zeta} \frac{\eta_\varphi(\zeta)}{\zeta}\,\mathrm{d}\zeta\ = \varphi_* \mathrm{f}_\varphi(\zeta, \zeta_0) \tag{9}$$

$$\mathrm{v}_\varphi(\zeta) = \frac{u_* \text{æ} z}{\eta_\varphi(\zeta)}, \quad u_* = \frac{\text{æ}\,|\vec{u}h\,|}{\mathrm{f}_u(\zeta_h, \zeta_0)} \equiv c_u\,|\vec{u}\,|_h, \quad c_u = \frac{\text{æ}}{\mathrm{f}_u(\zeta_h, \zeta_0)} \tag{10}$$

Where φ_* is the scale for concentration changes; η_φ, f_φ are universal continuous functions; $\zeta = z/L$ is dimensionless height characterizing atmospheric stability; indices $_0$ and $_h$ and related to levels $z = z_0$ and $z = h$ respectively; L is the length scale; z_0 is the roughness parameter; $æ$ is the von Karmann constant; ν_ϑ is the vertical turbulent exchange coefficient; u_* is the dynamic velocity. Because of lack of empirical data for functions $f_u, \varphi_*, \eta_\varphi$ and f_φ, it is supposed that, due to similarity of the physics of the turbulent exchange by heat and passive substances, the coefficients ν_φ and ν_ϑ can be taken to be approximately equal, implying that $\eta_\varphi = \eta_\vartheta$ and $f_\varphi = f_\vartheta$. Here η_ϑ and f_ϑ are the respective universal functions for temperature.

For $z = h$ eqs. (??)−(??) yield

$$\varphi = \varphi_0 \frac{f_\varphi(\zeta, \zeta_0)}{f_\varphi(\zeta_h, \zeta_0)} \tag{11}$$

$$h\frac{\partial \varphi}{\partial z} = \frac{\eta_\varphi(\zeta_h)}{f_\varphi(\zeta_h, \zeta_0)} \tag{12}$$

Multiplying Eq. (**??**) by $(\nu_\varphi)_h$ we arrive at

$$\nu_\varphi \frac{\partial \varphi}{\partial z} = c_u c_\vartheta (\varphi - \varphi_0)\,|\,\vec{u}\,|, \quad c_\vartheta = \frac{æ}{f_\vartheta(\zeta_h, \zeta_0)} \quad \text{for } z = h \tag{13}$$

which along with Eq. (**??**) can be used as boundary condition for $z \ge h$.

φ_0 is determined from the pollutant balance equation on the surface:

$$-\left(\nu_\eta \frac{\partial \varphi}{\partial z}\right)_0 = I, \tag{14}$$

where I is the emission on the earth's surface.

Note that in corresponding universal functions the molecular resistance r_b is also accounted for. Thus

$$r_a = \frac{1}{c_u c_\vartheta |\vec{u}|}. \tag{15}$$

To calculate r_a, the quasistationary surface layer model is used. For the strong instability area with no reliable measurement data, the so-called "-1/3"-law is adopted which results from similarity theory. For the strong stability area, a linearity law is assumed.

The surface resistance r_c is calculated by

$$\frac{1}{r_c} = \frac{1}{r_s + r_m} + \frac{1}{r_c + 1/r_{\text{cuticle}}}, \tag{16}$$

where r_s is the stomata resistance; r_m is the mesophyle resistance; and r_{cuticle} is the cuticle resistance.

The stomata resistance is a function of photosynthesis-active radiation, air temperature, leaf water potential, and vapor pressure deficit. The resistance to gas uptake by mesophile cells is determined by the area of the mesophile and gas solubility (≈ 0.1-0.5 cm/s). The cuticle resistance is linked with the gas uptake on leaf surface. This resistance depends on the chemical characteristics of gases and leaf surface area (≤ 200 cm/s).

2.3. Soil-atmosphere Exchange

In soil, lindane exists mainly in the following three states: water (dissolved) (φ_L); gaseous (φ_g); and absorbed (φ_s); the crystal state is not considered here.

The concentration of lindane can be so represented in the form:

$$\varphi_T = \rho_S \varphi_S + \vartheta \varphi_L + (\Phi - \vartheta)\, \varphi_g \tag{17}$$

where ρ_S is the volume density of soil; ϑ is the volume humidity of soil; Φ is soil porosity; $\varphi_S, \varphi_L,$ and φ_g are the concentrations in adsorbed, liquid, and vapor states, related to the soil solution mass, skeleton, and free vapor space, respectively.

The equilibrium of POP exchange between liquid and gas phases is quantitatively characterized by Henry's constant. This is the ratio of the concentration in the gas phase at equilibrium to that of a solution. Henry's constant (like saturated vapor pressure and solubility) depends on the chemical composition of the water phase and temperature. The adsorption of POP by atmospheric aerosols essentially depends on the relative humidity of air. At a definite level of the latter, mono-layer of adsorbed water molecules emerged in the soil surface. Increased numbers of these mono-layers lead to particles that have lost their initial features and turned to pure-water like ones. With increasing values of the relative humidity, the specific volume of the surface changes affecting the adsorption, leading to closure of different-size PORS, and decreasing the POP sorption. At the relative humidity values close to saturation, atmospheric aerosols are a three-phase system consisting of solid particles, water that covers them, and air. Interphase distribution of POP is determined by their solubility in water and by adsorption from the solution to the particle surface. The equilibrium of the first process is characterized by the Henry constant, and that of the second by the distribution coefficient related to the organic content.

Assuming that a POP obeys Henry's law, we get

$$\varphi_g = K_H \varphi_L \tag{18}$$

where K_H is the dimensionless Henry's constant. Since for most POPs, the characteristic time of concentration change in soil is greater than that of sorption and desorption, an instantaneous sorption equilibrium approximation can be assumed. In the linear approximation, the relationship between the adsorbed and dissolved phases can be represented as

$$\varphi_S = k_d \varphi_L \tag{19}$$

where k_d is the adsorption isotherm tilt, or distribution coefficient.

Using Eqs. (**??**)-(**??**), we obtain

$$\varphi_T = R_L \varphi_L = R_S \varphi_S = R_g \varphi_g \tag{20}$$

where

$$R_L = \rho_s k_d + \vartheta + (\Phi - \Theta) K_H,$$
$$R_S = \rho_s + \frac{\vartheta}{k_d} + \frac{(\Phi - \Theta) K_H}{k_d},$$
$$R_g = \frac{\rho_s k_d}{K_H} + \frac{\vartheta}{K_H} + (\Phi - \Theta)$$

are the coefficients for different states of lindane. Henry's coefficient is given in the general case by the formula.

$$K_H = K_{H_0} e^{b_1\left(\frac{1}{T} - \frac{1}{T_0}\right)} \tag{21}$$

where $K_{H_0} = K_H(T_0)$ and b_1 is an empirical constant.

The adsorption of lindane in soil depends on soil properties, and, particularly, on organic matter content. To account for the latter, a coefficient for carbon distribution (K_{oc}) is introduced:

$$k_d = f_{oc} K_{oc} \tag{22}$$

where f_{oc} is the fraction of organic matter content in soil. In numerical calculations, organic content data of FAO global data base are used. The resolution is $1^\circ \times 1^\circ$, with 27 types of organic soil in total and 9 categories of organic content (ranging from 1% to 30%) for the upper 30-cm layer.

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Biographical Sketches

Guri Marchuk is a scientific counselor of the Presidium of the Russian Academy of Sciences. Between 1962 and 1980 he worked in the Siberian Branch of the USSR Academy of Sciences, first as a director of the Computer Center and then as the Chairman of the Siberian Branch and Vice-President of the USSR Academy of Sciences. Between 1980 and 1986 he was a Deputy Prime Minister of the USSR and Chairman of the State Committee of Science and Technology. From 1986 to 1991 he was the President of the USSR Academy of Sciences. Guri Marchuk is an outstanding scholar in the field of numerical and applied mathematics. He has received many honors, including the Fridman, Keldysh, and Karpinski prizes; he is a member of the Academies of Sciences of Bulgaria, Czechoslovakia, Europe, Finland, France, Germany, India, Poland, and Rumania, and an Honorary Professor of Calcutta, Houston, Karlov, Tel-Aviv, Toulouse, and Oregon Universities, and Budapest and Dresden Polytechnic Universities. He is also a member of the editorial boards of many international and several Russian journals, and the Editor-in-Chief of the Russian Journal of Numerical Analysis and Mathematical Modelling, published by the Institute of Numerical Mathematics, RAS, in the Netherlands. Guri Marchuk is the author of a series of monographs on numerical mathematics, numerical simulation of nuclear reactors, numerical methods for the problems in the atmosphere and ocean dynamics, immunology, medicine, and environmental protection. For notable progress in scientific and organizational activities, Guri Marchuk has been awarded with prestigious state honors.

Kiril Kondratyev is a member of the Russian Academy of Sciences, Department of Oceanology, Atmospheric Physics and Geography. He is the Editor in chief of the journal *Issledovanie Zemli iz Kosmosa (Remote Sensing)*.

Academician Kondratyev is the cochairman of the National Oceanographic Committee, a member of the Governmental Oceanographic Committee, the chief of the Navigation and Oceanography Headquarters of the Russian Defence Ministry. Between 1964 and 1970, he was the rector of the Leningrad University. Academician Kondratyev is an outstanding scientist in the field of atmospheric radiation, greenhouse effect, numerical simulation of the ecological problems. He is the author of a number of monographs, *including Longwave Radiation Transfer in the Atmosphere, Nuclear Winter, Multivariate Global Changes, Climate Effects of Aerosols and Clouds.*

Artash Aloyan is an highly qualified specialist in the field of numerical modeling of atmospheric dynamics and environmental protection problems. He is engaged in development of mathematical models for transport of gaseous and aerosol pollutants in the atmosphere accounting for their transformation (photochemistry, condensation, coagulation, nucleation). From 1969 to 1987, he was a senior researcher at the Computer Center of the Siberian Branch of the USSR Academy of Sciences (Novosybirsk). Artash Aloyan earned his Ph.D. degree in 1975 in the field of numerical modeling of mesometeorological problems. In 1985, he earned his Dr. Sci. degree in mathematical modeling of the atmosphere dynamics and environmental protection problems. From 1987 on, he is a leading research scientist at the Institute of Numerical Mathematics of the Russian Academy of Sciences in Moscow, Russia. During his past research years, Aloyan has published two monographs and more than 100 papers in different scientific journals.