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Estimating chemical footprint on high-resolution geospatial grid

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Abstract

We need models to make decisions regarding minimization of chemical impacts on the environment. These models must allow us to make estimations with a minimum amount of data. The USEtox is an example of such a model.

However, the Russian Federation (as its districts and some regions) is large in area and contains a wide variety of topography and climatic features. As a result, estimations at the country, district, or region scales, on whole, might lead to incorrect administrative measures. Each administrative unit requires specific assessment and management decisions owing to the peculiarities of the state structure that take into account the above-mentioned features. In addition, there are many point sources causing significant chemical pollution (for example, chemical plants) in territories under consideration. These sources are often located a considerable distance from other sources.

Considering all of the above, we developed an algorithm to calculate the coefficients of chemicals transported between the compartments of the environment on a geospatial grid. The algorithm is based on the UNEP-SETAC scientific consensus USEtox model and a geographic information system (GIS) that provides data of water flows and airflows. We describe the application of the algorithm to the study of Hg^{2+} transport between Federal Districts of the Russian Federation on a low-resolution grid and to the study of Al^{3+} transport in the Leningrad Region on a high-resolution 0.5° x 0.5° geospatial grid.

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1. Introduction

Reliable methods for the prediction of chemical distribution in the biosphere are of great importance for the achievement of the Sustainable Development Goals (SDGs) set by the UN, for example, SDG 12, Item 12.4: «By 2020, achieve the environmentally sound management of chemicals and all wastes throughout their life cycle, in accordance with agreed international frameworks, and significantly reduce their release to air, water and soil in order to minimize their adverse impacts on human health and the environment» [1].

The chemical footprint (ChF) is an approach to the assessment of cumulative environmental impacts of chemical pollution and expresses the output as a "polluted volume" and as a pollution index [2].

Mathematical models that describe the fate of chemicals in the environment have been elaborated [3] and are used to express calculations of the transfer of chemicals released within countries [4], or even entire regions [2]. Several mathematical models, such as CalTox [5], Simple Box [6], BETR [7], and the UNEP-SETAC scientific consensus USEtox model [8], are incorporated into software products.

Spatial inhomogeneity of the environment should be taken into account by models to improve the accuracy of predictions [9], as the opposite leads to a decrease in the reliability of the life cycle of chemicals [10, 11], because different parts of large territories, such as countries and regions, may have varying characteristics [12, 13].

Models that consider spatial inhomogeneity have been applied for the global assessment of impacts caused by the emissions of inorganics, such as phosphorus [14, 15].

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Similar types of models are required for the calculation of the ChF and the impact assessment of chemicals of high concern. Some researchers recommend using the USEtox model for the calculation of the ChF [4]. They have reimplemented a combination of the environmental processes defined in the USEtox model [16] and have spatialized them on a high-resolution geospatial grid to capture spatial details [17]. In this paper, we present the algorithms for calculation of mass transfer of chemicals with water flows and airflows (while not running the model) and the accumulation of these chemicals in various environmental subsystems at global and regional scales on a grid with $0.5^{\circ} \times 0.5^{\circ}$ resolution.

2. Methods

We used the USEtox model [16] for the description of the chemical transport in the environment. We combined spatial peculiarities of the fate of the chemicals with the structure of the USEtox model. The USEtox model was chosen because 1) it was developed for the comparative assessment of chemicals and therefore is devoid of conservative assumptions; 2) it is widely accepted and applied as a UNEP-SETAC scientific consensus model [8] in life cycle impact assessment [18]; and 3) thanks to its database (v.2.0), for the time being, the USEtox model is capable of providing calculations for about 3000 organic chemical compounds and 25 elements [19], in particular, Hg²⁺ and Al³⁺, the case studies presented in this paper. Other advantages of the model are the availability of the system in the public domain; the presence of an open source code, which makes it easy to use this system together with other tools, e.g. GIS; and the existence of the informed approach, in which this model is recommended for calculating the ChF [4].

The USEtox model describes the environment as a system at the continental and global levels (Fig. 1). The continental level (level C) contains the following six compartments of the environment: urban air, rural air, seawater, fresh water, agricultural soil, and natural soil. The global level (level G) contains five compartments of the environment: atmospheric air, fresh water, ocean, agricultural soil, and natural soil.



Fig. 1. The USEtox model [8].

Given substance data and landscape data, the USEtox model for the continental and global levels calculates the

11×11 matrix **A** of mass balance rate constants $[day^{-1}]$ between the compartments of the environment (we did not consider the indoor level). Given the 11-dim vector **b** of the substance emission [kg/day] to the compartments, the USEtox model calculates the 11-dim vector **x** of the substance masses in the compartments [kg]. The vector of masses **x** is the solution to the **Ax+b=0** system of linear algebraic equations.

2.1. Model on a geospatial grid

As an approach to the just mentioned model, we considered a model that may contain many local levels and many continental levels, with the latter corresponding to the parts of the world, and the former belonging to the system shown in Fig. 2.



For this model, the parameters of level C of the USEtox model were calibrated to the squares of the corresponding local level cells, and the mass balance rate constants matrix was calculated (Fig. 3).

Local levels	1	2		i	Continental level
Locui levels	1	2	1952	1	continentariever
1	K matrix from USEtox for level C	Zero values		Zero values	Constants (USEtox) between compartments levels C and G
2	Zero values	K matrix from USEtox for level C		Zero values	Constants (USEtox) between compartments levels C and G
J	Zero values	Zero values		K matrix from USEtox for level C	Constants (USEtox) between compartments levels C and G
Continental level	Constants (USEtox) between compartments levels C and G	Constants (USEtox) between compartments levels C and G		Constants (USEtox) between compartments levels C and G	K matrix (USEtox) for level G

Fig. 3. Block mass balance rate constants for j local (yellow) and continental (green) levels

To create this matrix, we used the rate constants. The rate constants calculation was based on the processes described in the USEtox model.

2.2. Mass balance rate constants between cells

We removed the seawater compartment from the cells/territories where it was not physically present. However, for the simplicity of computer processing, for the time being, the compartment was excluded by zeroing all the constants of transport of substances from one compartment to another (mathematically correct). The following modifications of matrix A (Fig. 3) were made in our calculation to describe the mass transfer of chemicals with fresh water flows between different cells:

- The area of seawater for the local cells from which transport to other local cells is possible was rendered null.
- The area of seawater for the local cells from which transport to global cells is possible was increased according to the area of upcoming fresh water (catchment areas).
- For the local cells without the sea, the following rate constants were rendered null: from the atmosphere to the sea, from the ocean to the sea, from the sea to the atmosphere, from the sea to the ocean, from fresh water to the sea.
- For the adjacent cells, the rate constant of mass transfer with fresh water was set equal to the rate constant of mass transfer from fresh water to the sea.

2.3. Landscape data and geographical information systems

We used geographic information systems that provided data for our planet on a $0.5^{\circ} \times 0.5^{\circ}$ grid:

- The topology of the rivers was extracted from the global Simulated Topological Network at 30-minute spatial resolution (STN-30p) [20-22].
- The average multiyear values of the moving mass of water (discharge) per year (m³), average annual rainfall (mm/year), and irrigation (m³/year) and data on rural and urban population for 0.5°×0.5° cells are available on the website of the University of New Hampshire [23].
- The average temperature (K) and wind speed (m/s) for a 0.5°×0.5° grid are available on the European Centre for Medium-Range Weather Forecast website [24].
- The ratio of agricultural soils was taken from the website of the EarthStat project [25].
- The water volumes in lakes and reservoirs and the share of the area occupied by these ponds for a grid with cells of 0.5°×0.5° also are available on the University of New Hampshire website [23].

Another task to accomplish was the estimation of the j-th cell square occupied by water bodies and the depths of water bodies. The total area occupied by water in the j-th cell A_j^{total} (km²) was determined as the sum of the volumes of river water A_j^{riv} (km²), lake water A_j^{lake} (km²), and reservoir water A_j^{res} (km²). Data on lakes and reservoirs are available on the University of New Hampshire website [23].

The determination of river water content in the j-th cell required additional calculations. Some scientists propose calculating the volume of river water by multiplying some conditional parameters, i.e. length L_j (km), width W_j (km), and depth D_j (km) of a hypothetic river located in the j-th cell [14]. In the most simple case, the length of the conventional river (assuming that it flows from one side of the j-th cell to the other) may be calculated as $\sqrt{A_j}$, but taking into account the sinuosity of most rivers, the dimensionless correction factor of the sinuosity $S_{b,j}$ is included in the calculations and the length of the conventional river is determined according to the equation 1:

$$L_j = S_{b,j} \sqrt{A_j} \tag{1}$$

where: A_j - square for the j-th cell calculated for the grid with resolution of $0.5^{\circ} \times 0.5^{\circ}$ depending on its location (km²).

The factor of sinuosity for the j-th cell $S_{b,j}$ was calculated depending on the river course in accordance with relevant tables composed for different continents [29]. The width W_j and the depth D_j of the conventional river in the j-th cell are calculated using the following empiric equations 2 and 3 [30]:

$$W_j = K_{w1} \times \left(Q_j\right)^{K_{w2}} \tag{2}$$

$$D_j = K_{d1} \times \left(Q_j\right)^{K_{d2}} \tag{3}$$

where: K_{wl} , K_{w2} , K_{dl} , K_{d2} - empiric constants, which, for global calculations, are equal to: $K_{wl}=5.01^{*}10^{-2}$, $K_{w2}=0.52$, $K_{d1}=1.04^{*}10^{-3}$, and $K_{d2}=0.37$, respectively [14]; Q_j - average multiyear values of the moving mass of water (discharge) per year (m³) for j-th cell, with data for the grid with $0.5^{\circ} \times 0.5^{\circ}$ resolution available on the University of New Hampshire website [23].

The share of the area of the j-th cell under consideration, occupied by water $f_{i}^{A}{}_{w,i}$ is calculated using the equation 4:

$$fr_{w,j}^{A} = \frac{L_{j} \times W_{j} + fr_{j}^{A, lake + res} \times A_{j}}{A_{j}}$$

$$\tag{4}$$

where: $f_i^{A,lake+res}_j$ - the share of the area occupied by lakes and reservoirs for the j-th cell, with data for the grid with $0.5^{\circ} \times 0.5^{\circ}$ resolution available on the University of New Hampshire website [23].

The overall depth of water bodies in the j-th cell D^{total}_{i} is estimated using equation 5:

$$D_j^{total} = \frac{V_j^{total}}{f r_{w,j}^A * A_j} \tag{5}$$

where: V_j^{total} - the total volume of water in the cell, obtained by summing the volumes of water in lakes, reservoirs, and rivers. The data for the volumes of water in lakes and reservoirs for the grid with $0.5^{\circ} \times 0.5^{\circ}$ resolution are available on the University of New Hampshire website [23]. The volume of water in rivers in the j-th cell V_j^{riv} is estimated using equation 6:

$$V_j^{riv} = L_j \times W_j \times D_j \tag{6}$$

Furthermore, data of the Dai YAMAZAKI website [26] is planned to use to estimate river depth, and data of the website Lake Model Flake [27], or data of the more actual database: HydroSHEDS (Hydrological data and maps based on Shuttle Elevation Derivatives at Multiple Scales) [28], is planned to use to o estimate water body depth to hopefully improve the accuracy of the calculations significantly.

3. Case studies

We tested the model described above for two case studies of different elements (mercury and aluminium). Mercury was chosen, as this element is of great importance for the Russian Federation after it joined the Minamata Convention. Pollution by aluminium is important for the Leningrad region because several aluminium enterprises are located there.

In the first case study, we tested our model at the federal district level. We needed to consider the fact that the mercury discharged in most Federal Districts is drawn into the sea, except for the mercury from the Central Federal District and the Volga Federal District, which is transferred to the South Federal District.

In the second case study, we used the same algorithm for calculation of mass transfer of chemicals with water flows and the accumulation of these chemicals in various environmental subsystems, but we used GIS data for s grid with $0.5^{\circ} \times 0.5^{\circ}$ resolution.

In these two case studies:

- We tested the efficiency of the algorithm for the calculation of the mass transfer of chemicals with water flows and the accumulation of these chemicals in various environmental subsystems.
- We tested the efficiency of the regional scale model for a grid with 0.5°×0.5° resolution with the use of GIS data.

Because the inventory data are only available for total metallic form, it is assumed that mercury and aluminum are considered as Hg^{2+} and Al^{3+} in the case studies. Although the numerical values of the results could thus be questioned (speciation of the metals should be considered), this assumption does not compromise the testing and demonstration of the model application as intended in this study.

3.1. Hg²⁺ in the Federal Districts of the Russian Federation

We applied the model to Hg^{2+} fate for eight Federal Districts of the Russian Federation. For the calculations, in this study, we assumed that all mercury emitted was transformed into the divalent form. As the potential impacts caused by divalent mercury are much higher compared to those of metallic mercury, the results obtained corresponded to the worst scenario for the inorganic forms of mercury emitted into the environment. We have described eight local levels and two continental levels:

• Europe, including the Central, Northwest, South, and Volga Federal Districts

• Asia, including the North Caucasian, Siberian, Far-Eastern, and Ural Federal Districts

It was found out that pollutants from most of the Federal Districts were transported to the sea, but substances from the Central Federal District and the Volga Federal District were primarily transferred to the South Federal District, which was considered in the calculations:

- In the Central Federal District and the Volga Federal District, the sea areas were rendered null, and the sea area in the South Federal District increased in proportion to the sum of the areas of the South Federal District, the Central Federal District, and the Volga Federal District.
- In the Central Federal District and the Volga Federal District, the following indices were rendered null: rate constants of the transfer from the atmosphere to the sea (airC sea.waterC), from the ocean to the sea (oceanG sea.waterC), from the sea to the atmosphere (sea.waterC airC), from the sea to the ocean (sea.waterC oceanG) and from fresh water to the sea (fr.waterC sea.waterC).
- Rate constant (fr.waterC (the Central Federal District) fr.waterC (the South Federal District)) was set equal to the rate constant of transfer (fr.waterC (the Central Federal District) – sea.waterC (the South Federal District); similarly, the rate constant (fr.waterC (the Volga Federal District) - fr.water (the South Federal District)) was set equal to the rate constant of transfer (fr.waterC (the Volga Federal District) - sea.water (the South Federal district)).

For the case of Hg^{2+} fate, selected for the assessment, the resulting matrix of rate constants with the dimension of 46 by 46 (including 6 local and 2 continental levels) was converted using MS Excel and multiplied by the vector of sources of mercury entry into various environmental subsystems in all the Federal Districts. The comparison with calculations made using the USEtox model [31], showed a difference of 0.2% in the total mass of Hg^{2+} , distributed to local and continental levels.

3.2. Al^{3+} in the Leningrad region of the Russian Federation

For calculating the accumulation of Al^{2+} in different compartments of the environment of the Leningrad region, , we used data from the GIS of a $0.5^{\circ} \times 0.5^{\circ}$ grid.

The data on Al^{3+} content in the atmosphere and hydrosphere were taken from the registers of objects that have a negative impact on the environment (TVE, WONWAS). The results obtained are presented in the form of maps with $0.5^{\circ} \times 0.5^{\circ}$ grids (Figs. 4 and 5).

The difference between the total mass of AI^{3+} , calculated for the Leningrad plus Europe using the model under consideration, and the total mass of AI^{3+} , calculated using the USEtox model, was about 3.7%, which requires additional investigation.



Fig. 4. Al^{3*} content in fresh water in the Leningrad region (Russia) on the maps with $0.5^{\circ} \times 0.5^{\circ}$ grids overlaid with the Leningrad region map.



Fig. 5. Al^{3*} content in natural soil in the Leningrad region (Russia) on the maps with 0.5°×0.5° grids overlaid with the Leningrad region map.

4. Discussion

4.1. Comparison with the USEtox model.

The verification of the adequacy of our model was based on the observance of the mass balance. In the USEtox model, the sum mass is calculated as the sum of the mass of chemicals under consideration in all the compartments of the global and continental levels.

When testing our approach and model on a large scale (mercury at the federal level), we obtained approximately the same sum mass of mercury content distributed over the assessed compartments on several levels.

When we are using the grid approach, the differences between the sum masses calculated by the USEtox model and by our model were significant. This means that the USEtox model equations might not work correctly at the grid scale, and this problem needs further consideration.

As far as the Al^{3+} content is concerned, it decreased (Fig. 4) in the direction of the river currents, which means that our model reflected the phenomena of the dilution and the precipitation of the substance. In addition, based on Fig. 4, it might be concluded that the content of Al^{3+} increased when the watercourses merged. The model is demonstrating the

effect of the summation of the masses of the chemicals carried by the watercourses. The content of the AI^{3+} (Fig. 5) in the soil decreased with distance from the main sources of emissions. In this case, our model demonstrated the fact that, when the AI^{3+} was transferred from one cell to another, its content also decreased, because of its deposition.

4.2. Applicability of our approach and model at the global level.

We estimated the singular values of the 341×341 matrix for the Al³⁺ problem. The estimate of the smallest singular value was 3.474×10^{-8} , and the estimate of the largest singular value was 2.766×10^{11} . The condition number of the matrix was, consequently, about 7.962×10^{18} . This condition number is large, and thus one cannot guarantee fast convergence of the biconjugate gradient method we have used to solve the system of linear equations.

We also estimated the singular values of the 13×13 USEtox mass balance rate constants matrix for the Al problem. The estimate of the smallest singular value was $2.119*10^{-6}$, and the estimate of the largest singular value was $3.764*10^2$. The condition number of the system of linear equations was about $1.776*10^8$. This condition number of the USEtox mass balance rate constants matrix is fairly large for the 13×13 matrix.

The authors plan to investigate the reasons the matrices were ill conditioned, as well as the applicability of the approach and the model at the global level, in further works.

5. Conclusions and recommendations

The approach proposed in this paper for the assessment of the spatial differentiation of the residence time of chemicals in different environmental compartments produced good results in terms of mass balance when applied to large geographical areas. The combination of the USEtox model and GIS and the application of the proposed approach to the calculations for a grid with $0.5^{\circ} \times 0.5^{\circ}$ resolution resulted in discrepancies, which require further investigation of the method.

In the future, we plan to construct a mathematical model that contains no continental but many local levels, one level for each cell of a high-resolution grid. For example, a $0.5^{\circ} \times 0.5^{\circ}$ geospatial grid implies 259,200 cells with 6 compartments each and a system of linear equations with a sparse matrix of large dimension, i.e. 1,296,000×1,296,000.

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