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MODELLING OF AIR QUALITY AND ATMOSPHERIC DEPOSITION IN HUNGARY

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Abstract: Annual average atmospheric concentrations and depositions of sulfur and nitrogen species, as well as trace metals in Hungary and its surrounding regions were estimated by model computations. Spatial and temporal variations of the distributions are presented in the paper. The temporal variation of atmospheric sulfur budget was calculated based on a simple atmospheric box model. In the case of atmospheric lead, the cumulative atmospheric depositions were also estimated for a sixty years long period until 2015 using historical emission factors for the past decades as well as emission scenarios for the future. It was found that Hungary belongs to the moderately polluted regions in Europe. A model hierarachy was developed in order to perform multi-scale modelling studies on the air pollution processes at different spatial and temporal scales in Hungary. A set of environmental indicators was defined to characterize the actual state of the atmospheric environment.

Introduction

For the past decades, energy generation, industrial production and transportation have caused serious environmental contamination in east-central Europe. The rate of contamination can vary from place to place as a function of source densities and intensities of pollutant fluxes as well as meteorological conditions. The pattern of pollution may not only be characterized by local, highly concentrated sites such as densely populated urban areas, but also by lower concentrations of pollution widely dispersed over the landscape including agricultural regions, forests and surface waters. Trace gases and aerosol particles can be transported far away from their sources before being deposited on the surface. Due to the deep economic changes in this region during the past 15 years, its energy and industry structures were reorganized, which resulted in a significant decrease of pollutant emission. Out-of-date industrial technologies are being replaced by less energy consumer and more environment friendly ones. Thermal power plants are being equipped with efficient sulfur and dust filters. Leaded gasoline has been phased out of use. The rates of changes in pollutant emission, however, were different in the countries of the region. As a result of these changes, now the region is coping with less serious atmospheric environmental problems than 10–20 years ago.

Emission of pollutants

Due to the geographical size and location of the country, local air quality is highly influenced by both Hungarian and transboundary sources. At the same time, Hungarian sources have a strong effect on the air quality and rates of atmospheric deposition in its surrounding regions. In Hungary, the SO_2 emission decreased significantly during the

past decade in all important source categories: energy, industry, transportation, agriculture, home heating and other services. The release of 1,020 ktonnes estimated for 1990 was reduced to 480 ktonnes in 2000. In the case of nitrogen-oxides, the annual emission of 240 ktonnes in 1990 was reduced to 180 ktonnes within 10 years. It should be noted, however, that the relative contribution of transportation to the total emission increased during the same period. The emission rates of heavy metals have also decreased between 1990–2000. The most significant decrease was detected in the case of atmospheric lead (from 695,000 kg to 37,000 kg). The corresponding annual emission rates in 1990 and 2000 are 5,600 kg and 2,700 kg for Cd, 43,000 kg and 37,000 kg for Ni, 6,800 kg and 4,200 kg for Hg, 150,000 kg and 110,000 kg for V.

Sulfur and nitrogen species

Due to the significant reduction of SO_2 emission during the past decade, the annual average concentrations have also decreased. Based on EMEP model calculations (EMEP, 2003), local maxima could be detected in the northern part of western Hungary and in the northern part of eastern Hungary, where still lignite based powerplants are in operation. The model also shows, that Hungarian sulfur emitters contributes to the total atmospheric sulfur deposition in Hungary at a rate of 43%, the remaining part is transported from abroad. The atmospheric sulfur budget of Hungary, however, is still negative, so higher amount of sulfur is emitted than deposited by dry and wet atmospheric processes in the country. The rate of Hungarian SO_2 emission in 2000 was 0.24 Tg (S) yr^{-1}, while the rates of dry and wet deposition were 0.07 Tg (S) yr^{-1} and 0.04 Tg (S) yr^{-1}, respectively, thus the total atmospheric sulfur budget was –0.13 Tg (S) yr^{-1}. The value of the sulfur budget in 1990 was –0.38 Tg (S) yr^{-1}, so a significant improvement could be estimated for the last decade of the past century. Based on the measurements at the regional background stations in Hungary, the annual average SO_2 concentration decreased by 50% between 1990 and the first years of this century. The annual average NO_2 concentration under regional background conditions were in the range of 2.5–4.0 g (N) m^{-3} in 1990, which decreased into the range of 0.5–3.0 g (N) m^{-3} in Hungary. It should be noted, however, that in the vicinity of major roads and within urban areas, where traffic is the dominating NO_2 source, the average concentrations are much higher. Total atmospheric nitrogen deposition is also calculated by the EMEP model. It includes the dry and wet deposition of gases and aerosol particles containing nitrogen. Its range is between 400–1500 mg N m^{-2} yr^{-1}. Local maxima were detected in the northern and western part of the country. Concerning the atmospheric nitrogen budget of Hungary, it can be stated that it is more or less balanced for both reduced and oxidised nitrogen compounds**.** Modelling pilot studies were performed for the region of Lake Balaton (Figure 1. and 2.)

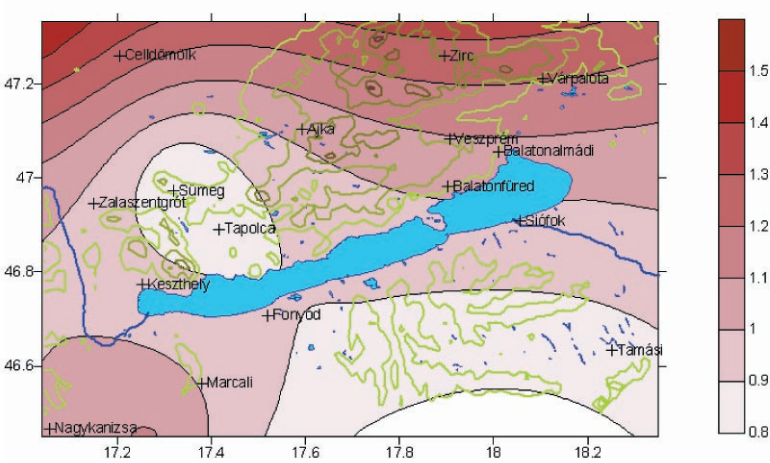

Figure 1. Regional background NO_2 concentration in the vicinity of Lake Balaton (μg N m^{-3})
1. ábra A NO_2 koncentráció (μg N m^{-3}) regionális háttere a Balaton környékén

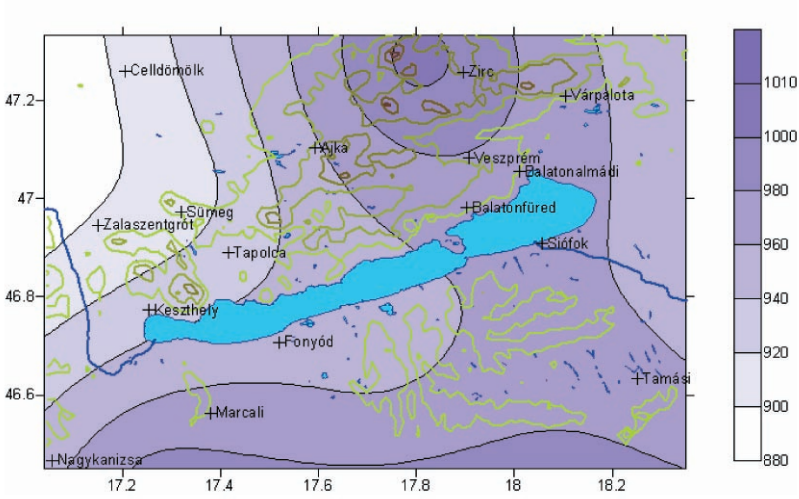

Figure 2. Total deposition of N species in the vicinity of Lake Balaton (mg N m^{-2} a^{-1})
2. ábra Összes nitrogénlerakódás a Balaton környékén (mg N m^{-2} a^{-1})

Trace metals

The most significant changes in the atmospheric environment of the past decades in the central-eastern European region is connected with the introduction and widespread use of unleaded gasoline. Lead emission decreased considerably in all countries of the region, resulting in decreasing atmospheric concentrations and total deposition rates, as well as in re-ranking the relative contributions of source categories. Long-range transport of atmospheric trace metals is not simply a clean air protection problem in

Europe but a major part of environmental policy as well: considering their characteristic residence time in the atmosphere (a few days) and the sizes of the countries in Europe, international co-operation is needed to control the transboundary flux and deposition of these pollutants. Model computations were performed based on the application of a Lagrangian-type long-range transport model, (Bozó, 2000), on the total deposition of lead and cadmium.

For the long-range transport computations a continental scale, climatological-type model (TRACE) was used. Input fields of emission inventory, meteorological data and deposition parameters are preprocessed for 50×50 km^2 spatial resolution EMEP grid system. The concentration of lead at a certain receptor point is given by a simple loss function:

$$c(x_r,y_r;x_e,y_e) = \beta\, E(x_e,y_e)\, R^{-1}\, (1\text{-}a)\, e^{-(kd+kw)t} \quad (1)$$

where:
c is lead concentration in the air in a certain receptor point as a result of an individual source point (ng m^{-3});

(x_r, y_r) and (x_e, y_e) are the spatial co-ordinates of source and receptor points;

E is the emission at the source point (ng s^{-1});

β is derived by setting the upward and downward atmospheric fluxes equal assuming mass conservation (s m^{-2})

R is distance between the source and receptor (m);

A is local emission coefficient (dimensionless);

kd and kw are loss parameters for dry and wet depositions (s^{-1});

t is atmospheric transport time between source and receptor (s);

The total concentration at the receptor point is computed from the sum of contributions coming from each emission source, weighted according to the frequency of 925 hPa backward trajectories, $F(s)$ coming from a particular sector, s:

$$c(x_r,y_r) = \sum_{s=1}^{8} F(s)\, c_s(x_r,y_r) \quad (2)$$

As indicated in (2) there are 8 spatial sectors considered during trajectory analyses. In the next step of the calculations, wet and dry deposition of the pollutant at the receptor

are computed from the atmospheric lead concentration:
$d_w = c(x_r,y_r)\ W\ P$ (3)

where:

W is the scavenging ratio (dimensionless);
P is the precipitation intensity (m s-1).

Dry deposition is expressed as:

$d_d = c(x_r,y_r)\ v_d$ (4)

where:
d_d is the dry deposition (ng m^{-2} s^{-1});
$c(x_r,y_r)$ is the lead concentration at the receptor point (ng m^{-3});
v_d is the dry deposition velocity (m s^{-1}).

Dry deposition velocities of lead are calculated separately for each grid element, depending on roughness length, friction velocity and size distribution of aerosol particles containing lead. Size distribution of particles were taken from MÉSZÁROS et al. (1997) whose research group carried out Berner-type cascade impactor sampling and trace metal measurements in Hungary.

For detailed model description, testing and validation see ALCAMO et al. (1992), BOZÓ (1996) and BOZÓ et al. (1992).

Even relatively low fluxes across the Earth's surface can result in accumulation of toxic trace metals in various soils. From the soils they can be taken up by the plants and may be leached out into the ground water.

Annual deposition in Hungary

The rate of annual lead deposition in 2002 over Hungary was found in the range of 600–2000 g km^{-2} yr^{-1}, much below the ecological threshold in Europe (250,000 g km^{-2} yr^{-1}). It was also estimated by model computations that ecological threshold of 5,000 g km^{-2} yr^{-1} for Cd was not exceeded in any parts of the region: it varies in the range of 20–300 g km^{-2} yr^{-1} Based on the comparisons with regional background measurements, it was found that our model simulations underestimate the rate of annual deposition by 10–20%. The reason for this could be the underestimations in emission data of trace elements and the uncertainty in the parameterization of deposition processes in the atmosphere. The rate of lead deposition varied significantly during the past decades. Results of a pilot model study are presented in Figure 3.

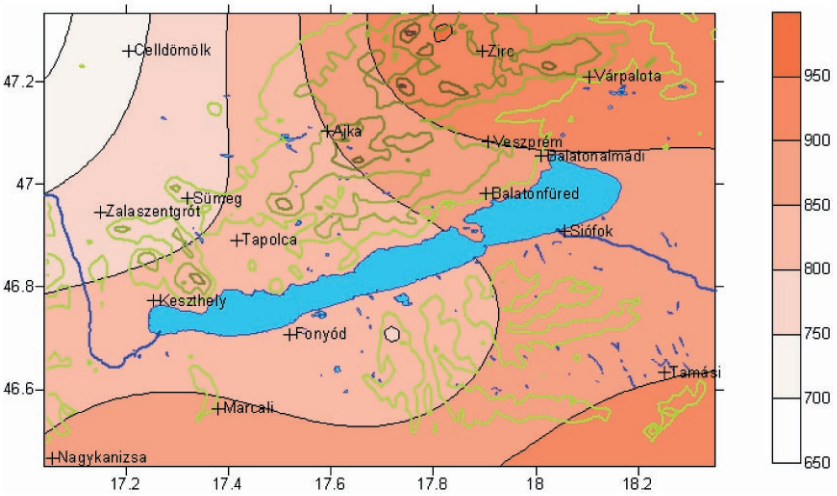

Figure 3. Total deposition of atmospheric lead (Pb) in the vicinity of Lake Balaton (g km^{-2} a^{-1})
3. ábra A legköri ólom (Pb) összes lerakódása a Balaton környékén (g km^{-2} a^{-1})

Cumulative lead deposition

Due to the cumulative characteristics of lead in our environment, it is advisible to estimate the cumulative lead deposition in Hungary for the past 50 years and to provide some quantitative estimates for the next decade. This type of simulation was also done by means of TRACE model computations. Historical emission data were taken from Olendrzynski et al. (1995), while future scenarios are based on the calculations of Berdowski et al. (1998). For comparisons, the target of model simulations was not only Hungary but a few other countries in different regions of Europe – United Kingdom, The Netherlands, Spain, Austria, Romania and Poland. It is not surprising that cumulative lead deposition was much higher during the 30 years of the period 1955–1985 than that of 1985–2015. Regarding Hungary, the rate of total lead deposition was 320 mg m^{-2} during 1955–1985, while on the basis of model computations it is expected that it will not exceed 95 mg m^{-2} during the consecutive 30 years period (1985–2015). It can also be stated that in some selected countries (e.g. The Netherlands or Austria) the cumulative lead deposition rate was higher than in Hungary, while in the case of Romania and Spain lower cumulative lead deposition rates were estimated.

Tropospheric regional background ozone

In general, the concentration of atmospheric pollutants is lower under regional background conditions than in urban areas. Usually, this is not true for surface ozone. It has no direct natural and anthropogenic sources, tropospheric ozone is formed from NO_2 via complicated chemical processes in the presence of precursors and solar radiation. Volatile organic compounds (VOC's) speed up the rate of tropospheric ozone formation.

It is typical, that the ozone peaks occur within 50–100 km distance downwind from highly polluted urban areas. Most of the countries in the east-central European region are coping with high ozone concentrations and threshold exceedences during summer period. Based on our regional background measurements carried out at K-puszta regional background air pollution monitoring station, Hungary, it was demonstrated that both 1 h and 8 h thresholds were exceeded: the numbers of annual exceedences – i.e., the number of the averaging periods with concentration values above the corresponding limit values in each year – were in the range of 2–37 for 1 h averages, and 60–138 for 8 h averages during the period of 1996–2003. The problems of regulations of ozone level can be demonstrated by model computations of HAVASI et al. (2001). Using the Danish Eulerian Model it was estimated, that even if the anthropogenic ozone precursors' emissions in Hungary were equal to zero, the AOT40 values would be at a level of 77–95% of their present values. The reason for it is that transboundary transport of precursors and tropospheric ozone itself contribute significantly to the level of ozone concentration in Hungary. In addition, precursors of natural origin, have also a considerable effect on ozone formation.

Local scale modelling

Long-range transport models (LRTMs) are operating over a 50×50 km^2 grid which is suitable to handle transboundary processes in the atmosphere. It is obvious, however, that better spatial resolution should be used for modeling of local scale air quality problems. LRTMs are providing the boundary conditions (e.g. background concentration or deposition field) for lower scale models. In Hungary, the adaptation of AIRMOD model was selected for this purpose. AERMOD is a steady-state plume model. In the stable boundary layer (SBL), it assumes the concentration distribution to be Gaussian in both the vertical and horizontal. In the convective boundary layer (CBL), the horizontal distribution is also assumed to be Gaussian, but the vertical distribution is described with a bi-Gaussian probability density function (pdf). This behavior of the concentration distributions in the CBL was demonstrated by WILLIS and DEARDORFF (1981). Additionally, in the CBL, AERMOD treats "plume lofting," whereby a portion of plume mass, released from a buoyant source, rises to and remains near the top of the boundary layer before becoming mixed into the CBL. AERMOD also tracks any plume mass that penetrates into the elevated stable layer, and then allows it to re-enter the boundary layer when and if appropriate. For sources in both the CBL and the SBL AERMOD treats the enhancement of lateral dispersion resulting from plume meander.

Using a relatively simple approach, AERMOD incorporates current concepts about flow and dispersion in complex terrain. Where appropriate, the plume is modelled as either impacting and/or following the terrain. This approach has been designed to be physically realistic and simple to implement while avoiding the need to distinguish among simple, intermediate and complex terrain, as required by other regulatory models. As a result, AERMOD removes the need for defining complex terrain regimes. All terrain is handled in a consistent and continuous manner while considering the dividing streamline concept (SNYDER et al. 1985) in stably stratified conditions.

One of the major improvements that AERMOD brings to applied dispersion modelling is its ability to characterize the PBL through both surface and mixed layer scaling. AERMOD constructs vertical profiles of required meteorological variables based on measurements and extrapolations of those measurements using similarity (scaling) relationships. Vertical profiles of wind speed, wind direction, turbulence, temperature, and temperature gradient are estimated using all available meteorological observations. The structure of the modelling system is shown in Figure 4.

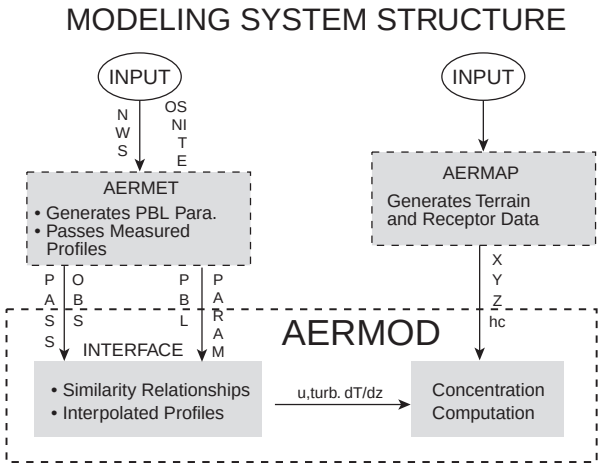

Figure 4. Data flow in the AERMOD system
4. ábra Adatáramlás az AERMOD rendszerben

That is the reason why we decided to replace our old dispersion model TRANSMISSION 1.0 (very simple Gaussian model) by AERMOD. We hope that AERMOD will give us much more results in our regulatory modelling calculations. Surface characteristics in the form of albedo, surface roughness and Bowen ratio, plus standard meteorological observations (wind speed, wind direction, temperature, and cloud cover), are input to AERMET. AERMET then calculates the PBL parameters: friction velocity (u^*), Monin-Obukhov length (L), convective velocity scale (w^*), temperature scale (Θ^*), mixing height (z_i), and surface heat flux (H). These parameters are then passed to the INTERFACE (which is within AERMOD) where similarity expressions (in conjunction with measurements) are used to calculate vertical profiles of wind speed (u), lateral and vertical turbulent fluctuations (δ_V, δ_W), potential temperature gradient ($d\Theta T/dz$), and potential temperature (Θ).

The AERMIC terrain pre-processor AERMAP uses gridded terrain data to calculate a representative terrain-influence height (h_c), also referred to as the terrain height scale. The terrain height scale h_c, which is uniquely defined for each receptor location, is used to calculate the dividing streamline height. The gridded data needed by AERMAP is selected from Digital Elevation Model (DEM) data. AERMAP is also used to create receptor grids. The elevation for each specified receptor is automatically assigned

through AERMAP. For each receptor, AERMAP passes the following information to AERMOD: the receptor's location (x_r, y_r), its height above mean sea level (z_r), and the receptor specific terrain height scale (h_c).

Results of a test run with AERMOD for a huge point source in SW part of Hungary are presented in Figure 5. It can be seen how the local orography influence the spatial distribution of pollutants.

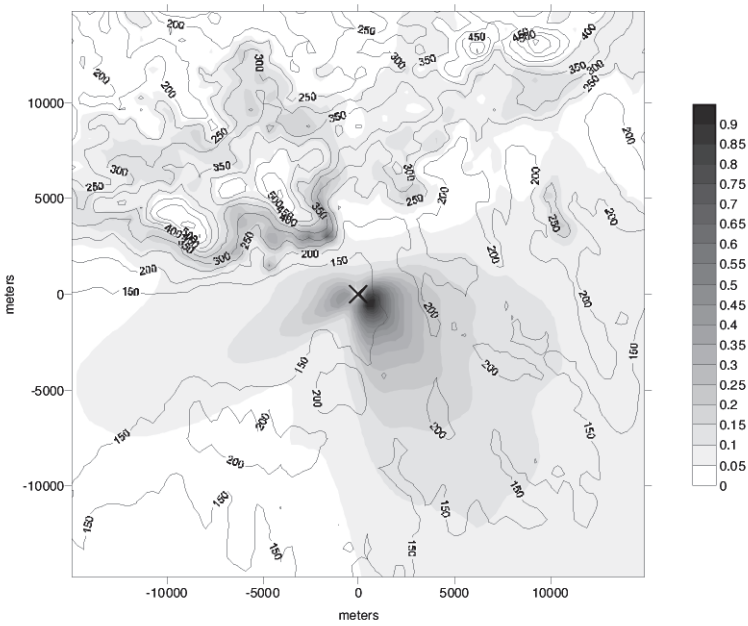

Figure 5. Yearly average NO_x concentration with elevated terrain. Units: μg m^{-3}.
The black isolines represent the terrain height. Contour intervals are at every 50 m.
5. ábra Átlagos évi NO_x koncentráció kiemelkedett domborzattal (μg m^{-3}).
A fekete izovonalak a terep magasságát jelölik. A kontúrvonalak között 50 m van.

Conclusions and future plans

Modelling tools developed allow us to simulate the dispersion and transport of atmospheric pollutants, as well as their dry and wet deposition processes from continental/ regional scales down to local scale. Temporal variations and future scenarios of air quality indicators (atmospheric concentration, acid deposition, toxic metal load, eutrophisation etc.) can also be prepared and evaluated. Validation of the models are performed with the data sets gained in national and international monitoring networks.The main advantage of the system presented is that depending on the purpose of the environmental study, the state of the atmospheric environment can be assessed at any location in Hungary. Output data computed by the atmospheric modelling system are to be forwarded to the connecting disciplines and are further applied and processed for the estimation of the state of soil, vegetation and surface waters, as well as the human

exposure in Hungary. It is planned that output matrices will be better resolved by means of statistical methods in the future. Detailed maps of environmental loads will also be prepared in the vicinity of most important stationary and line sources in Hungarian regions.

References

ALCAMO, J., BARTNICKI, J., OLENDRZYNSKI, K., PACYNA, J. 1992: Computing heavy metals in Europe's atmosphere – I. Model development and testing. Atmos. Environ. 26A, 3355–3370.

BERDOWSKI, J. J. M., PULLES, M. P. J., VISSCHEDIJK, A.J.H. 1998: Incremental cost and remaining emission in 2010 of Heavy Metals (HM) resulting from the implementation of the draft HM Protocol under UN ECE Convention on Long Range Transboundary Air Pollution. TNO Report – R 98/020.

BOZÓ, L. 1996: Long-range transport model estimations on the origin of atmospheric lead and cadmium deposition over Hungary. Időjárás 100: 43–49.

BOZÓ, L. 2000: Estimation of historical lead (Pb) deposition over Hungary. Időjárás 104: 161–172.

BOZÓ, L., ALCAMO, J., BARTNICKI, J., OLENDRZYNSKI, K. 1992: Total deposition and budgets of heavy metals over Eastern Europe. Időjárás 96: 61–80.

EMEP MSC-W, Transboundary acidification, eutrophication and ground level ozone in Europe. *MSC–W Status report* 1/2003, Part III.

HAVASI Á., BOZÓ L., ZLATEV Z. 2001: Model simulation on the transboundary contribution to the atmospheric sulfur concentration and deposition in Hungary. Időjárás 105: 135–144.

MÉSZÁROS, E., BARCZA, T., GELENCSÉR, A., HLAVAY, J., KISS, GY., KRIVÁCSY, Z., MOLNÁR, A., POLYÁK, K. 1997: Size distributions of inorganic and organic species in the atmospheric aerosol in Hungary. J. Aerosol Sci. 28:aaaa 1163–1175.

OLENDRZYNSKI, K., ANDERBERG, S., BARTNICKI, J., PACYNA, J. M., STIGLIANI, W. 1995: Atmospheric Emissions of Cadmium, Lead and Zink in Europe During the Period 1955-1987. IIASA Working paper WP-95-35.

SNYDER, W. H., THOMPSON, R. S., ESKRIDGE, R. E., LAWSON, R. E., CASTRO, I. P., LEE, J. T., HUNT, J. C. R., OGAWA, Y. 1985: The structure of the strongly stratified flow over hills: Dividing streamline concept. J. Fluid Mech., 152: 249–288.

WILLIS, G. E., DEARDORFF, J. W. 1981: A laboratory study of dispersion in the middle of the convectively mixed layer. Atmos. Environ., 15: 109–117.

A LEVEGŐMINŐSÉG ÉS A LÉGKÖRI LERAKÓDÁS MODELLEZÉSE MAGYARORSZÁGON

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Kulcsszavak: diszperzió és terjedés modellezése, légköri ülepedés, nagytávolságú légköri transzport, regionális és városi levegőminőség

Összefoglalás: Modellszámításokkal becsültük a légköri kén- és nitrogén-vegyületek, valamint a nehézfémek évi átlagos koncentrációját és ülepedését Magyarországon és közvetlen környezetében. Az eloszlások tér- és időbeli változásait mutatjuk be a dolgozatban. A légköri kén mérlegét egyszerű box-modellel határoztuk meg. A légköri ólom esetében a kumulatív ülepedési értékeket is kiszámítottuk egy 2015-ig tartó 60 éves periódusra, a történeti és az előrejelzett kibocsátások felhasználásával. Megállapítottuk, hogy Magyarország a közepesen szennyezett területek közé tartozik Európában. Multi-skálájú modellezési eljárás eredményeit is bemutatjuk a dolgozatban. Néhány környezeti indikátor meghatározásával jellemezzük a hazai levegőkörnyezet aktuális állapotát.