Proc. Estonian Acad. Sci. Ecol., 1996, 6, 1/2, 41—51
https://doi.org/10.3176/ecol.1996.1/2.04

ATMOSPHERIC TRANSPORT AND DEPOSITION OF TECHNOGENIC CALCIUM: MODEL ESTIMATION AND FIELD MEASUREMENT

Marko KAASIK

Department of Environmental Physics, University of Tartu, Tähe St. 4, EE-2400 Tartu, Estonia

Presented by J. Martin

Received 17 April 1995, revised version received 20 October 1995, accepted 18 December 1995

Abstract. Atmospheric calcium deposition with technogenic dust and fly ash in North-East Estonia was investigated. For deposition calculations the AEROPOL model, developed by the author, was used. The model allows of the estimation of concentrations and deposition loads of aerosols and gases emitted from elevated point sources over a plain terrain. The model is based on the Gaussian plume concept. Both dry and wet deposition were included. To take weather conditions into account, Pasquill stability classes were used (routine meteorological data needed). Model estimations were compared with the models of the same type developed at the Norwegian Institute for Air Research and with field measurement data from snow cover studies. The methods of geoinformatics were used. Fair agreement has been found. The calcium loads were modelled for time intervals not covered by field measurements.

Key words: atmospheric deposition, calcium load, cement dust, fly ash, Gaussian dispersion model, *Sphagnum* moss.

INTRODUCTION

Oil-shale fly ash and cement dust constitute more than a half of the technogenic emissions into the atmosphere in the North-East Estonian industrial region. Owing to their strong alkaline reaction and high nutrient content they have a substantial impact on the ecosystems. Below the geographical distribution of the calcium load is estimated. As earlier investigations show, under the conditions of high pH values the calcium load is the main damaging factor for *Sphagnum* mosses, covering large territories (more than 30%) in this region (Karofeld, 1994). An attempt was made to estimate the critical load for *Sphagnum* (and thus, for the bog ecosystem), using an atmospheric dispersion and deposition model. The results of field measurements, obtained in this region by different authors, do not constitute representative time series for independent deposition estimations, but could be used for model verification.

The development of a new dispersion model AEROPOL was urged by insufficiency of available models for the above-mentioned purpose. The program package Efir, based on works by Berlyand (Берлянд, 1985) and used earlier for air quality estimations in North-East Estonia, does not enable to describe actual dry and wet deposition rates, although the dependence of deposition loads on maximum concentrations was observed (Liblik & Rätsep, 1993). The box-model EMEST (Klimova, 1993), used for deposition estimations, has insufficient spatial resolution (10×10 km) for immediate neighbourhood of industrial enterprises. It should be pointed out that dry deposition properties of large particles are not yet well known, which produces high uncertainty in model estimations (Models and Methods..., 1993).

The Gaussian plume concept, used in the AEROPOL model, is the most widely used modelling algorithm: from 175 regulatory and operative air pollution models, registered in the World Meteorological Organization, 117 belong to this type (Scepesi, 1989). In developing the AEROPOL model, special attention was paid to removal mechanisms of admixtures (scavenging by rain and snow, adsorption, and gravitational sedimentation); a balance of removed and deposited matter is assumed. The full adsorbtion scheme of the Gaussian plume of large particles on the underlying surface has been used and verified. Thanks to Pasquill stability classes, used for dispersion calculations, for meteorological description only the data collected from observational stations of the state meteorological network are required.

MATERIALS AND METHODS

Research site. For a complete description North-East Estonia was considered together with the frontier region of Slantsy in Russia (Fig. 1). In the northern part of this area the landscape has been damaged by oil-shale mining, ash plateaus, and peat industry on large territories (Vallner & Sepp, 1993). The southern part is mainly forested, rural settlement is rare. Major pollutant sources are oil-shale heated power plants (TPP), chemical plants (CP), and cement industries (CI). The heights of the stacks considered vary from 60 m (the Kohtla-Järve TPP before reconstruction in the end of the 1980s) to 250 m (Estonian TPP). Because of its high content of alkaline oxides (CaO, K_2O, MgO) technogenic dust causes alkalization of naturally acidic soils, such as peaty soils. This process leads to the destruction of the swamp ecosystem. On the other hand, alkaline dust balances acidification caused by sulphur dioxide, emitted from the same sources.

Fig. 1. Model estimation region (filled rectangle).

Field measurements. Observational estimations of deposition loads of Ca^{2+} are based on snow samples collected during the winters of 1984/85 (Voll et al., 1989; Mandre, 1989; 80 samples after 110 days of permanent snow cover), and 1993/94 (by the author, 20 samples, after 35 days of snow cover). The background deposition value, 3 $mg \cdot m^{-2}$ per day (an average of six snow samples in rural areas of Central Estonia in winter 1984/85) was taken into account in model estimations. Nearly identical deposition loads were observed at the same time in the most remote western and southern areas of the North-East Estonian research site (probably caused by local sources, such as domestic heating, traffic, etc.).

Model. The concentration of an admixture is expressed as

$$C = B_c B_W (1 - P) C_0, \tag{1}$$

where C_0 is the concentration obtained considering only Gaussian dispersion and reflections, P is the penetration coefficient through the inversion layer, B_c and B_W are coefficients that constitute the corrections of the chemical reactions and wet deposition, respectively. The deposition load F consists of the dry deposition load F_d and wet deposition load F_W:

$$F = F_d + F_W. \tag{2}$$

Coefficients B_c, B_W, F_c, and F_W are specified below.

An aerosol plume, transported by the wind, disperses simultaneously in vertical and horizontal (perpendicular to the wind) directions, constituting a two-dimensional Gaussian distribution (Stern et al., 1984):

$$C_0 = \frac{Q}{2\pi\sigma_y\sigma_z u} \exp\left(-\frac{y^2}{2\sigma_y^2}\right) G_z, \tag{3}$$

where the dispersion in vertical direction is described by a sum G_z, where two reflections from the underlying surface and capping inversion (see below) are considered:

$$G_z = \sum_{n=-2}^{2} a_n \exp\left[-\frac{(z - H + 2nL)^2}{2\sigma_z^2}\right] + a_{n+1} \exp\left[-\frac{(z + H + 2nL)^2}{2\sigma_z^2}\right]. \tag{4}$$

The terms in Eq. (4) are grouped into pairs with respect to the recent reflection from the underlying surface: reflected n times (first term) and reflected $n+1$ times (second term).

In Eqs. (3) and (4) the following symbols are used: a_n, a_{n+1} — the reflection coefficients indicating the fraction of the admixture that was left before and after the last reflection, Q — the intensity of the source (the mass emitted per unit time), u — wind velocity, L — the height of the first inversion, y and z — vertical and horizontal coordinates.

To find the efficient height of the source H, we take into consideration the stack height and the initial plume rise. The latter depends on the diameter of the stack opening and the initial velocity and temperature of the emitted gases and the gravitational fall (for heavy aerosols). Dispersion parameters σ_y and σ_z increase with the distance x from the source (in the direction of the wind vector). The dispersion speed of pollutant plume, emitted from the point source, depends on the intensity of the atmospheric turbulence: in strongly convective conditions (a sunny day in summer) the plume disperses many times faster than in stable conditions (a cold night with weak wind). To consider these differences, all weather conditions are divided into six Pasquill classes, depending on the solar radiation intensity and the wind speed (Pasquill & Smith, 1983). Certain empirically determined dependences of dispersion parameters on x (different for rural and urban landscape) are related to each class. Due to their physically well-accepted behaviour Briggs' formulae for $\delta_y(x)$ and $\delta_z(x)$ (Stern et al., 1984) are used with correction for long distances ($x>10$ km) according to Sivertsen (1993).

Above the atmospheric convective boundary layer (at a height $L\approx1$ km) usually lies an inversion layer (capping inversion). In this layer the diffusion velocity is many times smaller than in the boundary layer. We assume that the turbulent flux of a pollutant reflects completely from the inversion layer and partially from the underlying surface. Further we assume that near the ground surface a heavy pollutant, e.g. fly ash with the particle diameter about 10 μm (Кикас et al., 1968), deposits completely on the vegetation and soil (the observed deposition velocities are bigger than the ordinary diffusion velocities (McMahon & Denison, 1979). Thus, $a_0=1$ and for $n\neq0$ all $a_n=0$. So formula (4) will obtain the following shape:

$$G_z=\exp\left[-\frac{(z-H)^2}{2\sigma_z^2}\right]+\exp\left[-\frac{(z+H-2L)^2}{2\sigma_z^2}\right]. \qquad (5)$$

For gaseous compounds partial adsorption (the rest being reflected from the surface) is considered, multiple reflections are assumed (Kaasik, 1994).

For high bouyancy and momentum plumes the penetration through the capping inversion is included, using empirical formulae for penetration coefficient P (Weil & Bower, 1984). In the case of stable stratification in the boundary layer, the inverison layer is based immediately on the underlying surface and no strict boundary with free atmosphere can be pointed out. In this case the second additive term in Eq. (5) is not taken into account.

In order to take wet deposition into consideration, the washing-out coefficient Λ (individual for each pollutant, depending on the rainfall rate) is introduced, according to the exponential decay law for wet deposition coefficient B_w:

$$B_w=e^{-\Lambda(x/u)}. \qquad (6)$$

The first-order chemical reactions are included in the same way. The corresponding coefficient is expressed as

$$B_c=e^{-k_r(x/u)} \qquad (7)$$

for a primary emission and as

$$B_c=1-e^{-k_r(x/u)} \qquad (8)$$

for a reaction product, where k_r is the reaction speed.

The washing-out coefficients and deposition velocities of solid particles, SO_2, and some other pollutants presented by McMahon & Denison (1979) were used here.

According to field measurement data, the washing-out process for aerosol particles by snow is 25 times more efficient than by rain (Graedel & Franey, 1975): $\Lambda_{\text{snow}}=25\Lambda_{\text{rain}}$.

The wet deposition load is calculated integrating the washed-out mass of the admixture over the tilted path of raindrops or snowflakes from the mixing height to the underlying surface:

$$F_w=\int_0^L \Lambda C[x(z),y,z]\,dz, \qquad (9)$$

where the integrating path is defined by the equation

$$x(z)=z\tan\beta=z\frac{v_p}{u},\qquad (10)$$

where β is the tilting angle of the precipitation fall and v_p is the velocity of its gravitational fall.

The dry deposition load is derived from the mass conservation assumption in the deposition process:

$$F_{\mathrm{d}}=v_{\mathrm{d}}C_{\mathrm{s}},\qquad (11)$$

where C_{s} is the concentration of an admixture near the underlying surface and v_{d} is the deposition velocity. In the case of complete adsorbtion the deposition velocity is equal to the diffusion velocity:

$$v_{\mathrm{d}}=u\frac{H}{\sigma_z}\frac{\mathrm{d}\sigma_z}{\mathrm{d}x}.\qquad (12)$$

To guarantee the mass conservation in the case of high diffusion velocity, for gaseous admixtures the larger from diffusion velocity and empirical adsorption velocity is chosen.

Modelling the deposition loads during some time period or long-term average, the depositions from individual Gaussian plumes were averaged (corresponding to time sequence or long-term classification of meteorological conditions). For model calculations the following data are used.

1. Meteorological data: wind speed and direction, rainfall (snowfall) rate, air temperature, sun height, cloud amount. The climatic averages for long-term calculations are obtained from a published material (Eesti NSV Kliimaatlas, 1969), for certain winter seasons (1984/85 and 1993/94) from the archive of the Estonian Institute of Meteorology and Hydrology.
2. Source data: source intensity (the mass emitted per time unit), stack height, opening diameter, temperature and the speed of the gas jet from the stack, obtained from Laigna & Potashnik, 1990 and Liblik & Rätsep, 1993.
3. Microphysical data: adsorption velocity and washing-out coefficient of admixture (from McMahon & Denison, 1979), for aerosol particles also their density and diameter (from Кикас et al., 1968). The calcium content of fly ash and cement dust are respectively 22% (Пец et al., 1985) and 30% (Annuka, 1994).

The computing code of the AEROPOL model is written in TurboPascal 7.0. For a successful run at least a 386DX computer is needed. The output of the model is formatted as a raster map of the concentration or deposition load of the pollutant. Each numeric element of the matrix represents the value of the concentration or deposition load on a certain grid point. The output is compatible with geographical analysis system IDRISI (Eastman, 1992).

RESULTS

Model verification. The results of dispersion calculations obtained using the AEROPOL model were compared with the results from Gaussian model CONCX, developed at the Norwegian Institute for Air Research (NILU) (Bøhler, 1987), and with field observations (snow samples).

Comparing the computed concentrations of fly ash, emitted from a point source, estimated by the AEROPOL and the short-term dispersion model CONCX, a fair agreement can be pointed out (Fig. 2).

The summary statistics of the modelled and measured loads at snow sample sites are given in the Table. The correlation coefficient is statistically significant at confidence level 99%.

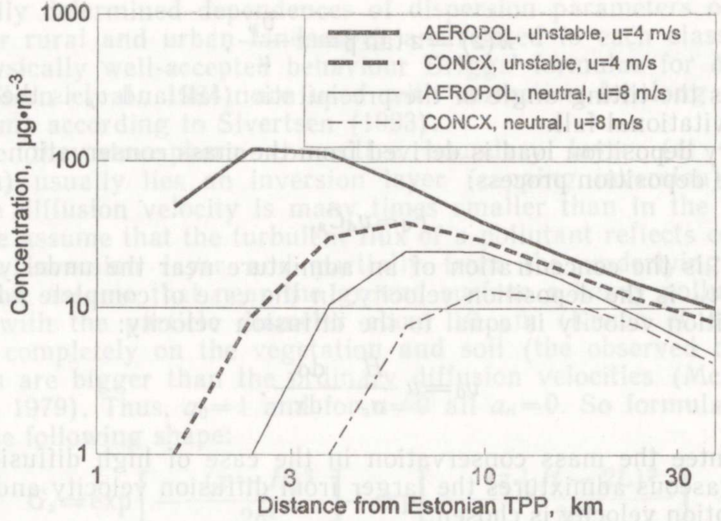

Fig. 2. Comparative model estimations of sector-averaged (30°) ground-level downwind concentrations of fly ash, emitted from one stack of the Estonian TPP ($Q=1670\,\mathrm{g\cdot s^{-1}}$) in different atmospheric stability and wind conditions.

Comparison of computed and measured (from snow samples) loads of calcium ($\mathrm{mg\cdot m^{-2}}$ per day) during the winter season 1984/85

	Source of data	
	AEROPOL model	Field measurement
Area of spatial averaging, km²	1	20—2000
Average Ca load	21.9	37.5
Minimum Ca load	3.6	2.6
Maximum Ca load	134.1	171.0
Standard deviation	21.0	35.7
Correlation coefficient	0.53	

A comparison of the geographical distribution of the loads calculated by the AEROPOL model with the measured ones revealed considerable systematic deviations. Near relatively low sources (e.g. Kohtla-Järve TPP before reconstruction) the computed calcium loads (Fig. 3) are higher than the measured ones (Fig. 4). Near high stacks (150—250 m) with high momentum and bouyancy plumes (the Baltic and the Estonian TPPs) an opposite tendency is seen. The distribution around sources agrees with the prevailing southwestern and northeastern winds during winter 1984/85 (Mandre, 1989). The considerably high loads (both observed and computed) to the north from Narva could be explained by the collective effect of the Estonian and the Baltic TPPs.

In the fixed direction from the Estonian TPP the observed calcium loads have a steeper gradient near the high stack than expected by the AEROPOL model (Fig. 5). Deposition profiles agree with a profile of the calcium content in the bog water near the pollution sources in North-East Estonia (Karofeld, 1994).

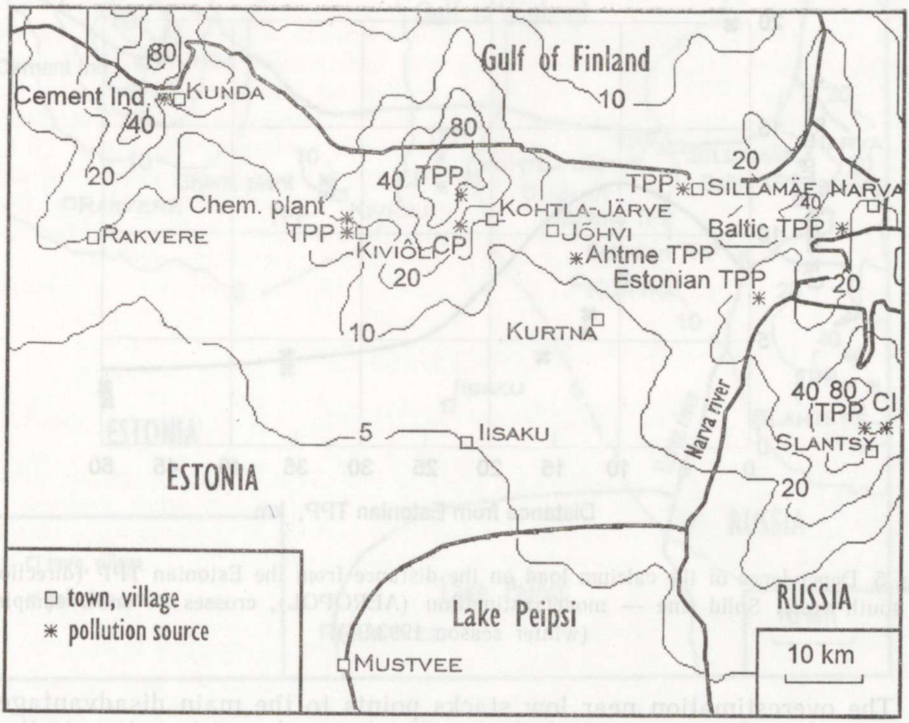

Fig. 3. Deposition load of calcium during the winter season 1984/85, mg · m^{-2} per day (modelled, using AEROPOL).

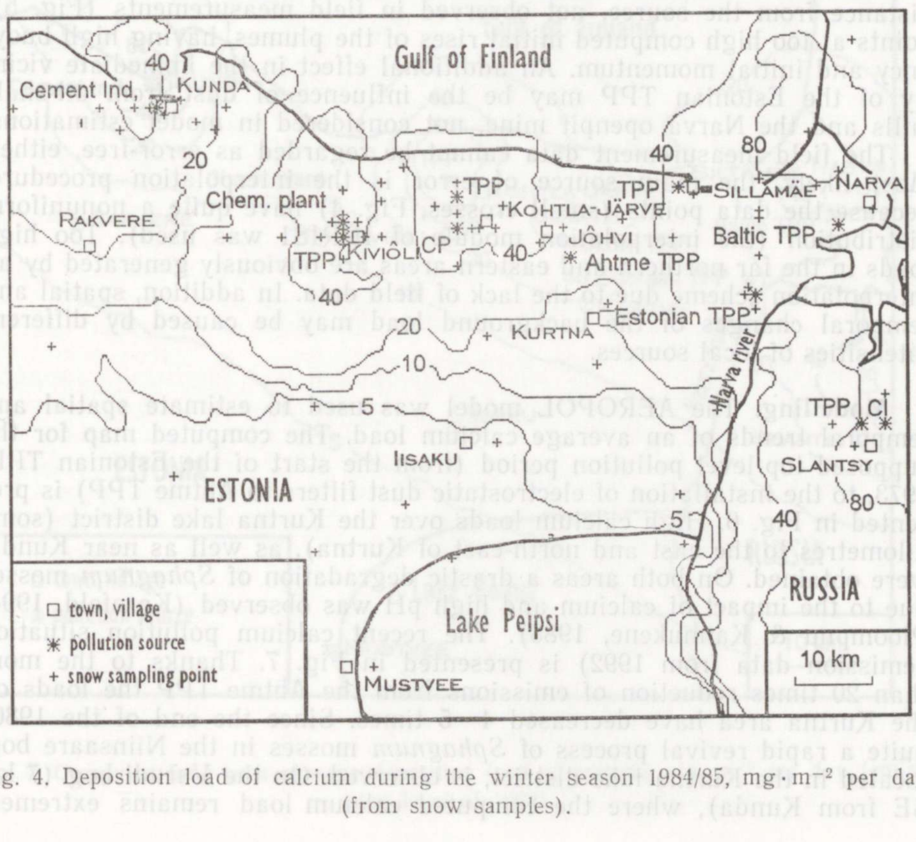

Fig. 4. Deposition load of calcium during the winter season 1984/85, mg m^{-2} per day (from snow samples).

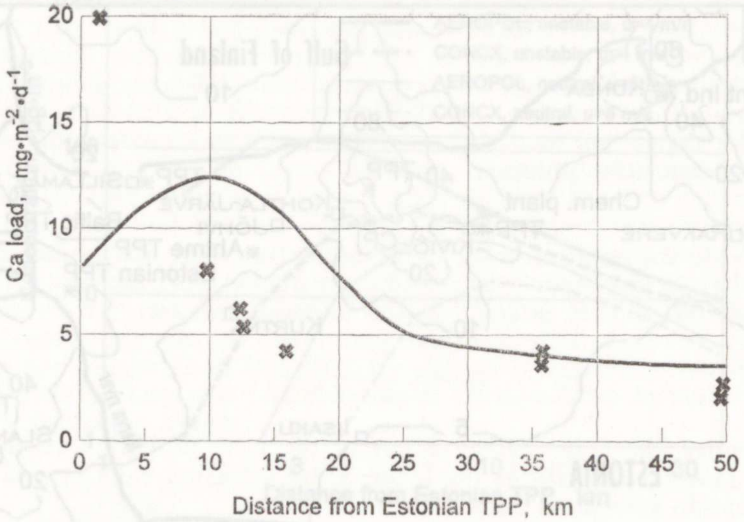

Fig. 5. Dependence of the calcium load on the distance from the Estonian TPP (direction to south-west). Solid line — model estimation (AEROPOL), crosses — snow samples (winter season 1993/94).

The overestimation near low stacks points to the main disadvantages of the Gaussian model: rigidly defined dispersion parameters do not allow of consideration of the weakening of turbulence near the ground. Underestimation near high sources with the maximum of concentration at some distance from the source, not observed in field measurements (Fig. 5), points at too high computed initial rises of the plumes, having high buoyancy and initial momentum. An additional effect in the immediate vicinity of the Estonian TPP may be the influence of dust from oil-shale mills and the Narva openpit mine, not considered in model estimations.

The field measurement data cannot be regarded as error-free, either. Most likely, the main source of error is the interpolation procedure, because the data points (small crosses, Fig. 4) have quite a nonuniform distribution (the interpolation module of IDRISI was used). Too high loads in the far northern and eastern areas are obviously generated by an interpolation scheme due to the lack of field data. In addition, spatial and temporal changes of the background load may be caused by different intensities of local sources.

Modelling. The AEROPOL model was used to estimate spatial and temporal trends of an average calcium load. The computed map for the supposed top-level pollution period (from the start of the Estonian TPP, 1973, to the installation of electrostatic dust filters to Ahtme TPP) is presented in Fig. 6. High calcium loads over the Kurtna lake district (some kilometres to the east and north-east of Kurtna), as well as near Kunda, were obtained. On both areas a drastic degradation of *Sphagnum* mosses due to the impact of calcium and high pH was observed (Karofeld, 1994; Ploompuu & Kannukene, 1988). The recent calcium pollution situation (emission data from 1992) is presented in Fig. 7. Thanks to the more than 20 times reduction of emissions from the Ahtme TPP the loads on the Kurtna area have decreased 4—5 times. Since the end of the 1980s quite a rapid revival process of *Sphagnum* mosses in the Niinsaare bog, located in the Kurtna lake district, is observed. On the Varudi bog (7 km SE from Kunda), where the computed calcium load remains extremely

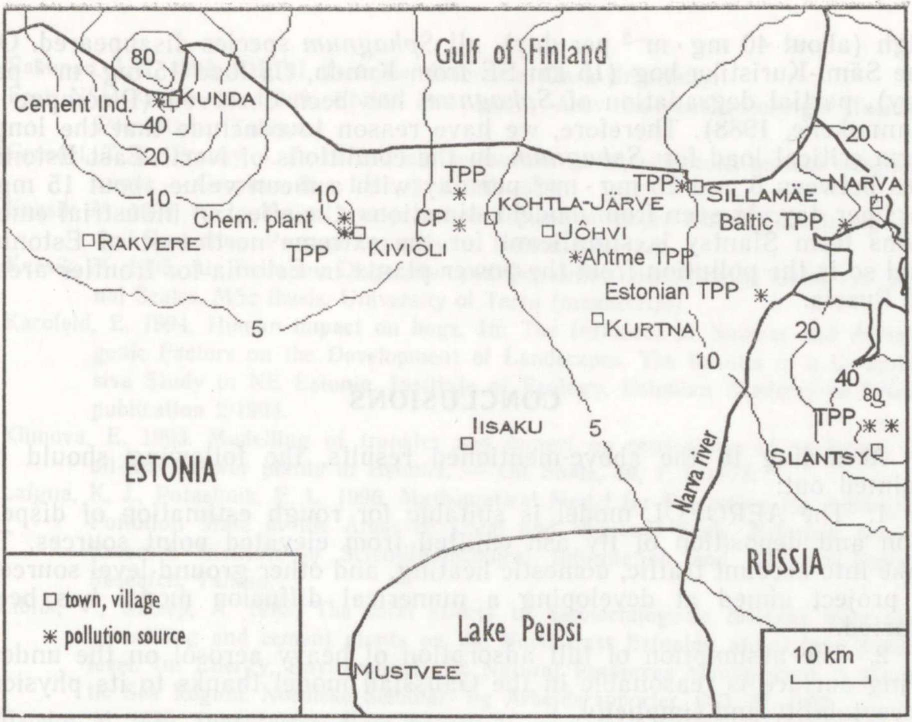

Fig. 6. Computed long-term average deposition load of calcium, mg · m^{-2} per day (emission level of the years 1973—79).

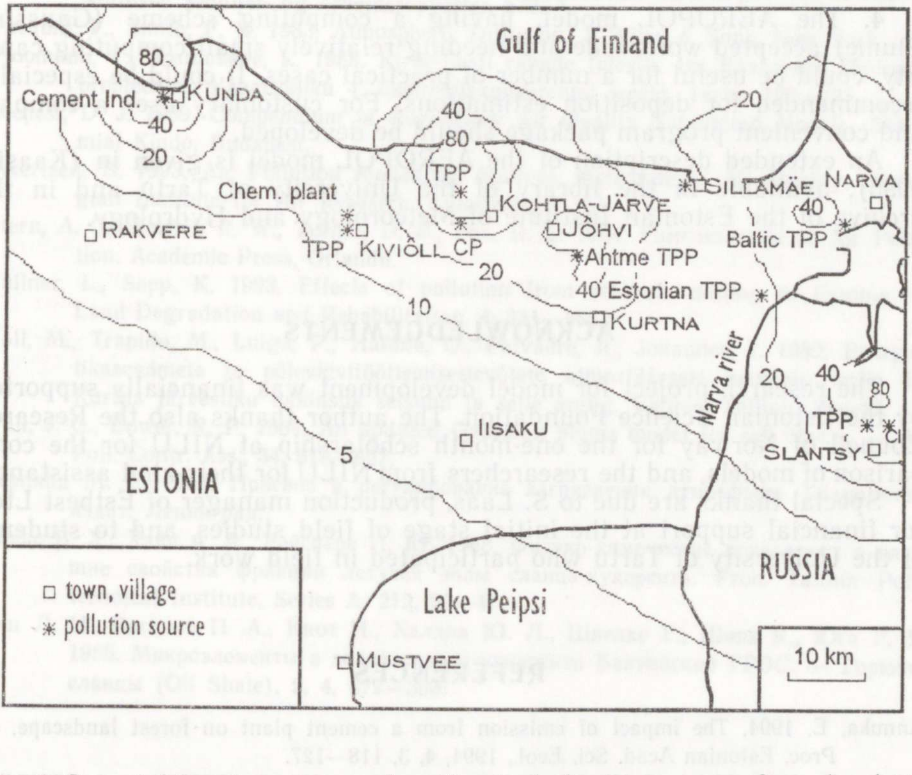

Fig. 7. Computed long-term average deposition load of calcium, mg · m^{-2} per day (emission level of 1992).

high (about 40 mg · m^{-2} per day), all *Sphagnum* species disappeared. On the Sämi-Kuristiku bog (15 km SE from Kunda, Ca load 15 mg · m^{-2} per day), partial degradation of *Sphagnum* has been observed (Ploompuu & Kannukene, 1988). Therefore, we have reason to conclude that the long-term critical load for *Sphagnum* in the conditions of North-East Estonia lies between 5 and 25 mg · m^{-2} per day with a mean value about 15 mg · m^{-2} per day. As seen from model estimations, the effect of industrial emissions from Slantsy is significant for the extreme north-east of Estonia, and so is the pollution from the power plants in Estonia for frontier areas of Russia.

CONCLUSIONS

According to the above-mentioned results, the following should be pointed out:

1. The AEROPOL model is suitable for rough estimation of dispersion and deposition of fly ash emitted from elevated point sources. To take into account traffic, domestic heating, and other ground-level sources, a project aimed at developing a numerical diffusion model has been started.
2. The assumption of full adsorption of heavy aerosol on the underlying surface is reasonable in the Gaussian model thanks to its physical acceptability and simplicity.
3. Model estimations of fly ash deposition give considerable quantitative knowledge about the ecological impact of technogenic emissions. For more precise studies an integrated model of the systems air—soil—plant cover should be developed.
4. The AEROPOL model, having a computing scheme (Gaussian Plume) accepted worldwide and needing relatively small computing capacity, could be useful for a number of practical cases. It could be especially recommended for deposition estimations. For customary user a compact and convenient program package should be developed.

An extended description of the AEROPOL model is given in (Kaasik, 1995), available in the library of the University of Tartu and in the archive of the Estonian Institute of Meteorology and Hydrology.

ACKNOWLEDGEMENTS

The research project for model development was financially supported by the Estonian Science Foundation. The author thanks also the Research Council of Norway for the one-month scholarship at NILU for the comparison of models, and the researchers from NILU for their kind assistance.

Special thanks are due to S. Laas, production manager of Estbest Ltd., for financial support at the initial stage of field studies, and to students of the University of Tartu who participated in field work.

REFERENCES

Annuka, E. 1994. The impact of emission from a cement plant on forest landscape. — Proc. Estonian Acad. Sci. Ecol., 1994, 4, 3, 118—127.

Bøhler, T. 1987. Users Guide for the Gaussian Type Models CONCX and CONDEP. Norwegian Institute for Air Research, Lillestrøm.

Eastman, J. R. 1992. IDRISI. Technical Reference. Clark University.

Eesti NSV Kliimaatlas. 1969. ENSV MN Juures Asuv Hüdrometeoroloogia Teenistuse Peavalitsus, Tallinn.

Graedel, T. E., Franey, J. P. 1975. Field measurements of submicron aerosol washout by snow. — Geophys. Res. Lett., 2, 8, 325—328.

Kaasik, M. 1994. Air pollution transport and deposition model AEROPOL. In: Land and Soil Protection. Proc. of Symposium. Tallinn, 222—232.

Kaasik, M. 1995. Air Pollution Dispersion and Deposition Modelling in Local and Regional Scales. MSc thesis, University of Tartu (manuscript).

Karofeld, E. 1994. Human impact on bogs. In: The Influence of Natural and Anthropogenic Factors on the Development of Landscapes. The Results of a Comprehensive Study in NE Estonia. Institute of Ecology, Estonian Academy of Sciences, publication 2/1994.

Klimova, E. 1993. Modelling of transfer and impact on ecosystems of emissions from oil-shale power plants in Estonia. — Oil Shale, 10, 1, 67—78.

Laigna, K. J., Potashnik, E. L. 1990. Mathematical Model for Predicting the Atmospheric Pollution State in the Areas of Local and Regional Scale. Estonian Scientific Research Institute of Scientific-technical Information and Technical-Economic Research, Tallinn.

Liblik, V., Rätsep, A. 1993. The total effects of aerotechnogenic influxes from power generating and cement plants on the North-East Estonian atmosphere pollution level. In: Energy, Environment and Natural Resources Management in the Baltic Sea Region. Nordiske Seminar- og Arbejdsrapporter, 653, 91—102.

Mandre, M. 1989. Tolmusaaste: Mõju männile ja . . . — Eesti Loodus, 11, 723—731.

McMahon, T. A., Denison, P. J. 1979. Review paper. Empirical atmospheric deposition parameters — a survey. — Atmospheric Environment, 13, 571—585.

Models and Methods for the Quantification of Atmospheric Input to Ecosystems. 1993. Nordiske Seminar- og Arbejdsrapporter, 573.

Pasquill, F., Smith, F. B. 1983. Atmospheric Diffusion. J. Wiley & Sons, New York.

Ploompuu, T., Kannukene, L. 1988. Kirde-Eesti rabade tulevik. In: Kaasaegse ökoloogia probleemid. Vabariikliku 4. ökoloogiakonverentsi teesid. Tartu, 120—123.

Scepesi, D. J. 1989. Compendium of Regulatory Air Quality Simulation Models. Akademiai Kiado, Budapest.

Sivertsen, B. 1993. Air Pollution Modelling. Sources, Meteorology, Air Quality. Norwegian Institute for Air Research, Lillestrøm.

Stern, A. C., Boubel, R. W., Turner, D. B., Fox, D. L. 1984. Fundamentals of Air Pollution. Academic Press, Orlando.

Vallner, L., Sepp, K. 1993. Effects of pollution from oil-shale mining in Estonia. — Land Degradation and Rehabilitation, 4, 381—385.

Voll, M., Trapido, M., Luiga, P., Haldna, Ü., Palvadre, R., Johannes, I. 1989. Energeetikaseadmete ja põlevkivitöötlemisettevõtete atmosfäärsete heitmete levik. In: Kurtna järvestiku looduslik seisund ja selle areng. Valgus, Tallinn, 29—42.

Weil, J. C., Bower, R. P. 1984. An updated Gaussian plume model for tall stacks. — Air Poll. Contr. Ass., 34, 818—827.

Берлянд М. Е. 1985. Прогноз и регулирование загрязнения атмосферы. Гидрометеоиздат, Ленинград.

Кикас В. Х., Хайн А. А., Рейспере Н. Ю. 1968. Физико-химические показатели и вяжущие свойства фракции летучей золы сланца-кукерсита. Proc. Tallinn Polytechnical Institute, Series A, 212, 29—47.

Пец Л. И., Ваганов П .А., Кнот И., Халдна Ю. Л., Швенке Г., Шник К., Юга Р. Я. 1985. Микроэлементы в золах сланца-кукерсита Балтийской ГРЭС. — Горючие сланцы (Oil Shale), 2, 4, 379—390.