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EDACS: European Deposition maps of Acidifying Components on a Small scale

W.A.J. van Pul, C.J.M. Potma, E.P. van Leeuwen, G.P.J. Draaijers, J.W. Erisman

Laboratory for Air Research, LLO
National Institute of Public Health and Environmental Protection, RIVM
P.O. Box 1, 3720 BA Bilthoven, the Netherlands

Abstract
In this paper a description is given of the EDACS model (European Deposition of Acidifying Components on Small scale), with which the deposition of acidifying components on a small scale over Europe is calculated for 1989. The acidifying components considered in EDACS are sulphur and reduced and oxidized nitrogen compounds. Dry deposition is estimated with the inference method i.e the deposition at the surface is inferred from the concentration and the deposition velocity at the same height. The deposition velocity is calculated using a resistance model in which the transport to and absorption or uptake of a component by the surface are described. Dry deposition velocity fields over Europe are constructed from a detailed land-use map ($1/6^{o}$x$1/6^{o}$ lat/long grid, made by RIVM) and meteorological information using a detailed parameterization of the dry deposition process. These small-scale dry deposition velocity fields are combined with concentration fields from the EMEP Lagrangian long range transport model to yield dry deposition amounts on a small scale. Wet deposition is also estimated, based on measurements, to obtain a total acidifying deposition map at a European scale. These deposition fields clearly reflect the spatial detailed land-use information and the large-scale concentration pattern over Europe.
The maps of the acidifying components over Europe on a small scale are made in cooperation with the EMEP\MSC-W, Oslo, Norway.

1 Introduction

In Europe, sulphur and reduced and oxidized nitrogen compounds are found to acidify soils and surface waters. Furthermore, nitrogen deposition causes eutrophication. The effects of acidification and eutrophication have been described extensively in the literature (e.g. Heij and Schneider, 1991, Grennfelt and Thörnelöf, 1992). Especially the effects on ecosystems have obtained great attention due to their vulnerability. In order to protect these ecosystems critical loads have been defined above which there is an increased risk of damage to the ecosystems.

The main atmospheric pathways via which emitted acidifying components reach the earth's surface are dry and wet deposition. The acidifying components can be transported over a long range up to 1000 km or more dependent on the component properties and the dry and wet deposition processes. Several models exist for estimating long-range transport of acidifying components on a European scale (e.g. EMEP: Sandnes, 1993, TREND: van Jaarsveld and Onderdelinden, 1994). The purpose of these models is to describe the relation between source

and receptor. The model results are used in quantifying country to country budgets, as a basis for the sulphur and nitrogen protocols and in assessments of the effects of acidification. The horizontal spatial scale on which these models operate is typically 50x50 km. However, describing the effects of acidification on the level of ecosystems, the acid load should be available at the scale of ecosystems or at scales which allow for comparison with critical loads (i.e. typically in the order of a 1x1 km resolution, Hettelingh *et al.*, 1991). So the above model resolutions are not appropriate to describe the acidifying load at the level of ecosystems.
A method was presented by van Pul *et al.*, 1992a, with which the deposition of acidifying components on a small scale over Europe can be mapped. Their method was discussed at the ECE-EMEP/BIATEX Workshops on deposition at Göteborg, Sweden (Lövblad *et al.*, 1993) and Aveiro, Portugal (Slanina *et al.*, 1993) and accepted as currently the best available method to describe local acid deposition fluxes.
In this paper the method is described and the first, preliminary, maps of small-scale fluxes of acidifying components over Europe are presented. The calculations are made with the EDACS model (European Deposition of Acidifying Components on Small scale) in which the method is adopted. Recommendations made during the Workshops are incorporated in the model.
The emphasis in the method is on modelling local scale dry deposition fluxes. A detailed parameterization of the dry deposition process for each acidifying component is based on available experimental results (EUROTRAC/BIATEX project) and literature (Erisman *et al.* 1994a). Wet deposition is also estimated to obtain a total acidifying deposition map over Europe (van Leeuwen *et al.*, 1994). The dry deposition maps were presented at the EUROTRAC symposium at Garmisch Partenkirchen, April 1994 by Erisman *et al.*(1994b) and van Pul *et al.*(1994a). The maps of the acidifying components over Europe on a small scale are made in cooperation with the EMEP/MSC-W, Oslo, Norway.

2 General description of EDACS

An overview of the input for and calculation scheme of EDACS is presented in Figure 1.
In EDACS the dry deposition is estimated with the inference method (Hicks, 1986). The deposition at the surface is inferred from the concentration and the deposition velocity at the same height. The deposition velocity is calculated using a resistance model in which the transport to and absorption or uptake by the surface of a component are described (see Section 3). The parameterizations are dependent on surface characteristics and other environmental and meteorological conditions. Dry deposition velocity fields, on a 6-hourly basis, are constructed from a detailed land-use map using these parameterizations along with the meteorological information (Potma, 1993). Here a RIVM data base is used which contains land-use data on a $1/6^{o}$x$1/6^{o}$ lat/long grid over Europe (van de Velde *et al.* 1994). Finally the dry deposition amounts are calculated by multiplying these dry deposition velocity fields with concentration fields.

In principle the concentration data can originate from measurements, model calculations or a combination of both. However, the spatial resolution of the operational European and national networks (ECE-EMEP, EUROTRAC) is too coarse to provide the necessary data and not every component is measured. This will lead to large uncertainties in the interpolated concentration fields. On a local scale, national or local networks may provide the concentration data. When the local maps are aggregated into one European map, problems

may arise about the inconsistency between the networks and it is foreseen that still not a full coverage of Europe can be obtained. However, for parts of Europe with small horizontal concentration gradients this can be done e.g. for UK:UK Review group on acid rain, 1990; Sweden: Lövblad *et al.* 1993, The Netherlands; Erisman 1992).

In this paper the concentration data of the EMEP Lagrangian long-range transport model (hence EMEP-LRT) on a 150x150 km scale were used (as described in Sandnes, 1993) to obtain a consistent concentration field over Europe. Using calculated concentration fields, the relation between emissions and deposition is maintained and assessments or scenario studies can be made at different scales.

In the inference method it is assumed that a constant flux layer is present between the reference height and the surface i.e. the atmospheric surface layer. This assumption implies that there is no significant advection in the layer and the air flow is well-adapted to the surface properties of the depositing surface and chemical reactions are not present. In that

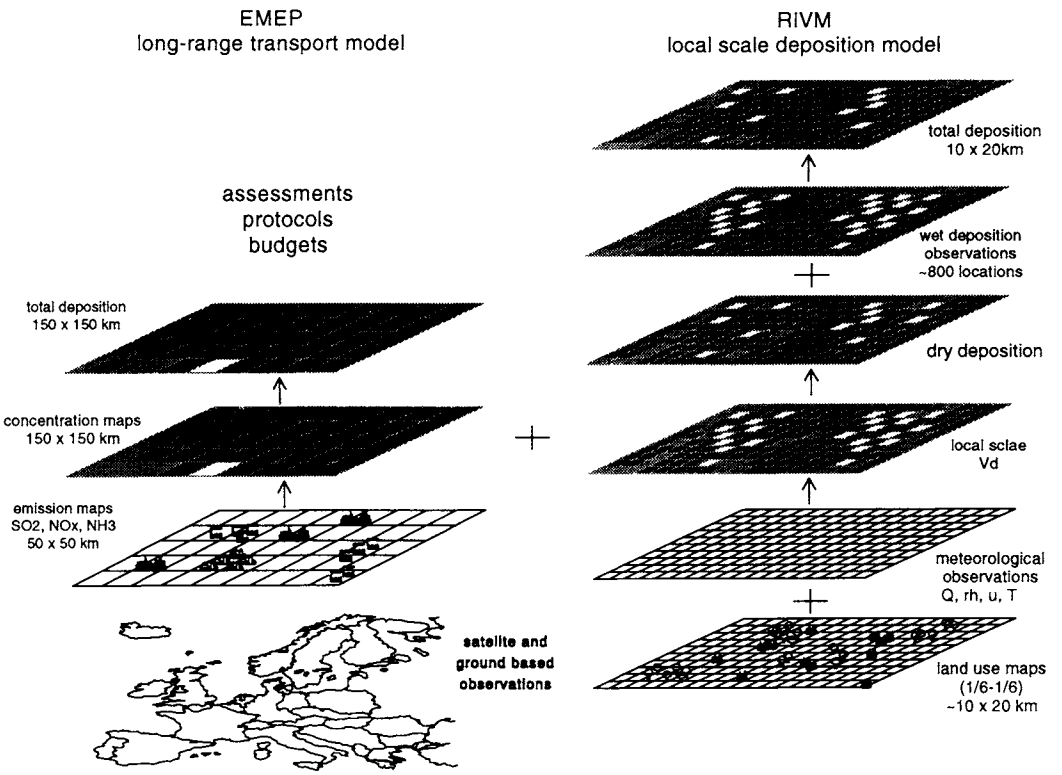

Figure 1 Overview of the input for and calculation scheme of EDACS. For explanation see text.

case the deposition flux at the reference height equals the deposition flux at the surface. The adaptation of the air flow to the surface is strongly dependent on the surface roughness and the stability of the air. The choice of the reference height is a compromise between the height where the concentration is not severely affected by local deposition and is below the surface layer height. In EDACS the concentration at 50m is taken which is the lowest LRT model level above the surface. This concentration then is assumed to be representative for a certain area, here an EMEP LRT gridcell of 150x150 km, and consequently can be used in estimating the deposition to surfaces within this area.

The wet deposition of acidifying components is based on measurements of the concentrations in precipitation and rain amounts (van Leeuwen *et al.*, 1994). A concentration map over Europe was constructed from these data by kriging. A data set with interpolated values of long-term yearly precipitation amounts was used to calculate the wet deposition.

The components considered here are SO_2 and SO_4^{2-}-aerosol (SO_x), NO, NO_2 (NO_x), HNO_3 and NO_3^--aerosol and NH_3 and NH_4^+-aerosol (NH_x). NH_x is considered to be acidifying because of the nitrification processes in the soil in which H^+ is produced (van Breemen *et al.* 1982). If all above components which are deposited produce one equivalent of acid this leads to a total potential acid load which is estimated from: potential acid = 2 SO_x + NO_y + NH_x. The actual acid load differs from the potential load because of an incomplete nitrification of NH_3 and by neutralization of the acidity by base cations. HONO, PAN and HNO_2 are not taken into account. However, the contribution of these components to the total acidifying deposition is very small (e.g. Lövblad et al., p 19).

3 Parameterization of the dry deposition velocity

The dry deposition flux of gases and particles from the atmosphere to a receptor surface is governed by the concentration in air and turbulent transport processes in the boundary layer, by the chemical and physical nature of the depositing species and by the efficiency of the surface to capture or absorb gases and particles. The flux of a trace gas is given as:

$$F = V_d(z)\ c(z) \qquad (1)$$

where c(z) is the concentration at height z and V_d is the dry deposition velocity (Chamberlain, 1966). *z* is the reference height above the surface: here taken as 50m. If the surface is covered with vegetation, a zero-plane displacement, d, is included: z=z-d. The absorbing surface is often assumed to have zero surface concentration. This holds only for depositing gases and not for gases that might also be emitted, such as NH_3 and NO. For these gases a non-zero surface concentration, a compensation point c_p, might exist, which can be higher than the ambient concentration, in which case the gas is emitted. Here the concentration at the various surfaces, c_s, is assumed to be zero for all components because of insufficient knowledge of the compensation point.
The parameterization of the dry deposition velocity is based on a description of this process via a resistance analogy or Big Leaf Model (see e.g. Thom, 1975, Hicks *et al.*, 1987, Fowler, 1978). In this resistance model the most important deposition pathways via which the component is transported and subsequently destroyed at, or taken up by the surface, are parameterized. The resistance model used here is shown in Figure 2.

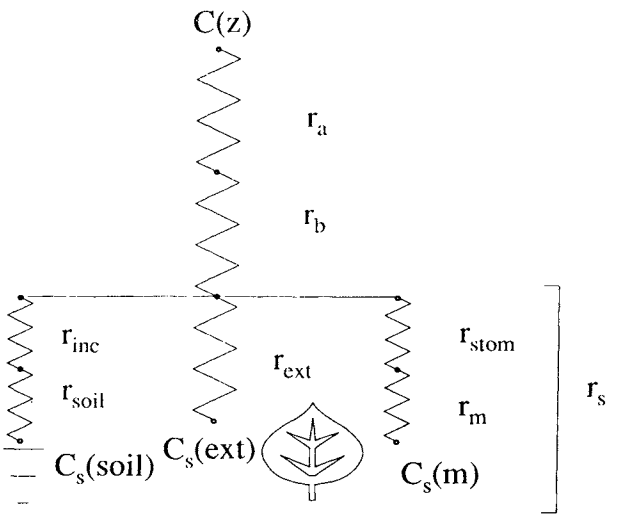

Figure 2 Resistance or Big Leaf model used in EDACS. c_s(soil), c_s(ext) and c_s(m) denote the concentration at the soil, external or mesophyl surface respectively, for explanation of the other symbols see text).

V_d is the inverse of three resistances:

$$V_d = (r_a + r_b + r_s)^{-1} \quad (2)$$

These three resistances indicate the three stages of transport: the aerodynamic resistance, r_a, represents the resistance against turbulent transport of the component close to the surface; the quasi-laminar sublayer resistance, r_b, accounts for the transport of the component through a laminar layer adjacent to the surface by molecular diffusion and the surface resistance r_s for the uptake or destruction at the surface. This surface resistance is composed of the resistances of the various destruction or uptake processes at the surface.
For a surface covered with vegetation this is:
-the stomatal resistance, r_{stom}, the resistance to the transport through the stomata of leaves and needles;
-the mesophyl resistance, r_m, the resistance of the internal plant tissues against the uptake or destruction (in a chemical way);
-the cuticle resistance, r_{cut}, or external surface resistance, r_{ext}, the resistance of the exterior plant parts against the uptake or destruction of the component;
-the r_{inc} the in-canopy aerodynamic resistance to account for the transport of air above the vegetation towards the soil and lower plant parts;
-r_{soil}, the soil resistance, the resistance against destruction or absorption at the soil surface;
These resistances which act in parallel or series are summed up to yield a (total) surface resistance, r_s:

$$r_s = \left[(r_{inc} + r_{soil})^{-1} + r_{ext}^{-1} + (r_m + r_{stom})^{-1} \right]^{-1} \quad (3)$$

For a water surface: $r_s = r_{wat}$, where r_{wat} is the resistance against the solution of gases in water.
For bare soil: $r_s = r_{soil}$ and for urban areas: $r_s = r_{urban}$. When the surface is covered with snow $r_s = r_{snow}$.

In turn, these resistances are affected by meteorology, leaf area, stomatal physiology, soil and external leaf surface pH, and presence and chemistry of water drops and films. Especially the state of the leaf and soil surface i.e. the presence of water films and snow, is an important variable in the deposition of soluble gases such as SO_2 and NH_3.

The process of dry deposition of particles of acidifying components is not very well known compared to the gaseous counterparts (Ruijgrok *et al.*,1993). As a best estimate the dry deposition of particles is described using a parameterization by Wesely *et al.*(1985) and Erisman (1992). Recent information on the deposition of particles to forests has come available (Ruijgrok *et al.* 1994) and will be used in a future version of the model.

The scheme used here to derive the surface resistances for SO_2, NO_2, NO, HNO_3, NH_3 is described in Erisman *et al.*(1994a). This scheme is based on previous publications among others Wesely (1989), Lövblad *et al.*(1993) and recent dry deposition measurements (among others in the BIATEX project of EUROTRAC). More details on the actual parameterizations used in EDACS can be found in van Pul *et al.*(1995).

4 Results

The 6-hourly deposition velocity fields were averaged to daily values and multiplied with the daily EMEP/LRT concentrations. These daily dry deposition maps were summed to annual totals. In Figure 3 the dry deposition of total potential acid over Europe estimated with EDACS is shown. The dry deposition values for most components vary greatly over Europe. This is partly explained by variations in the deposition velocity caused by variations in land use and meteorological conditions over Europe. The concentration pattern of the components over Europe, which are associated with the distribution of emissions, introduce variations on a larger scale (150x150km i.e. the EMEP grid). Large emission areas can be detected in the maps e.g. for SO_2 and NO_2 this is the so-called black triangle (Eastern Germany - Poland - Czech Republic), for ammonia e.g. north western Europe (The Netherlands, Denmark).

The total potential acidification map which is the sum of the dry deposition and wet deposition is presented in Figure 4. This figure reflects the above mentioned variations. The relative contribution of dry and wet deposition can be observed. For instance in the Scandinavian countries the surface inhomogeneities are not represented due to the large contribution of the wet deposition which has a smooth distribution over this area.

In Figure 5 the standard deviation of the total deposition of EDACS cells in an EMEP-grid cell (about 100 EDACS cells in one EMEP-grid) is given as absolute values and values relative to the average per EMEP-grid. It can be seen that the largest absolute values of the standard deviation can be found in the above described emission areas. Whereas the relative standard deviation is largest in areas with a small deposition and so variations in land use, meteorology etcetera are reflected.

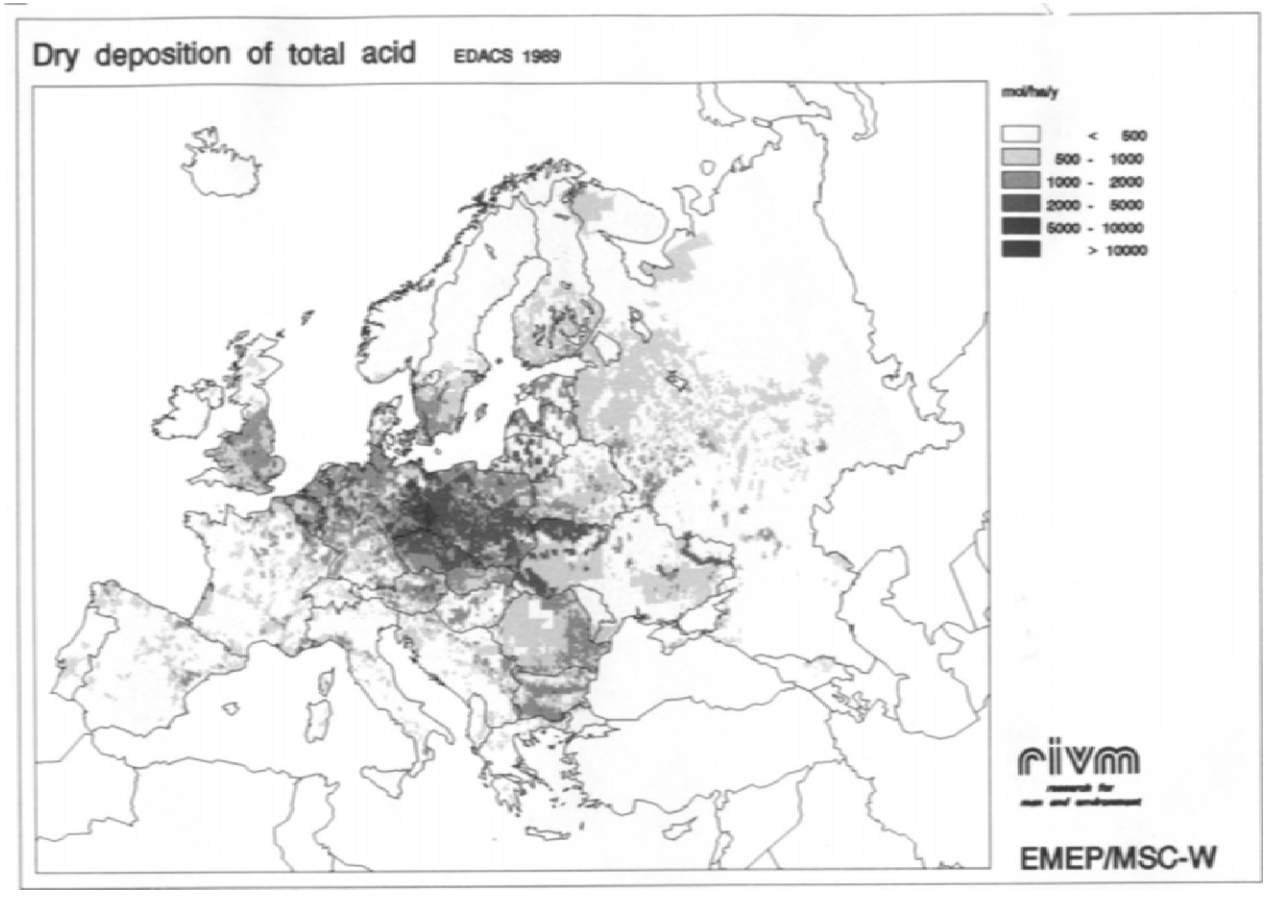

Figure 3 Dry deposition of total potential acid over Europe on 1/6°x1/6° lat/long grid for 1989 (mol ha^{-1} $year^{-1}$).

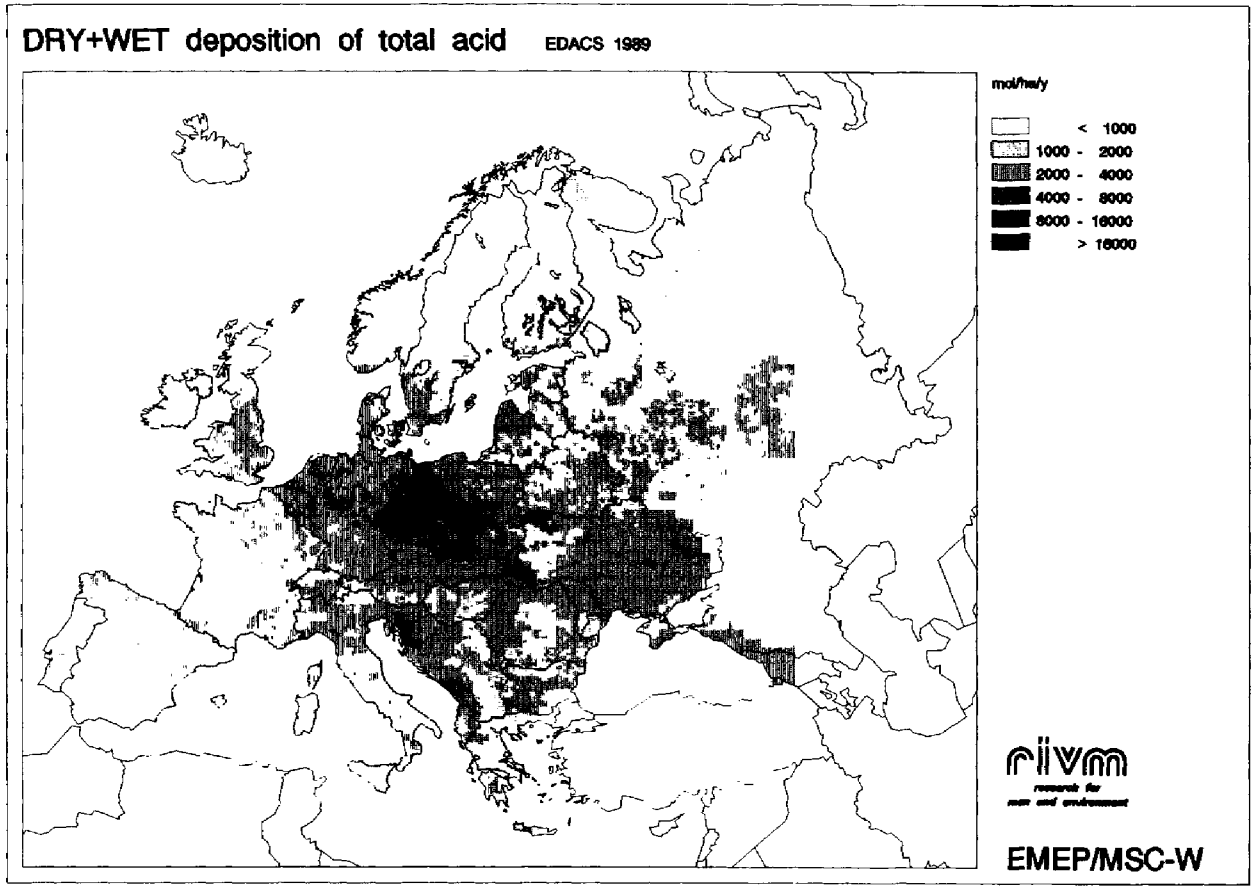

Figure 4 Potential acidification map over Europe on 1/6°x1/6° lat/long grid for 1989 (mol ha^{-1} $year^{-1}$).

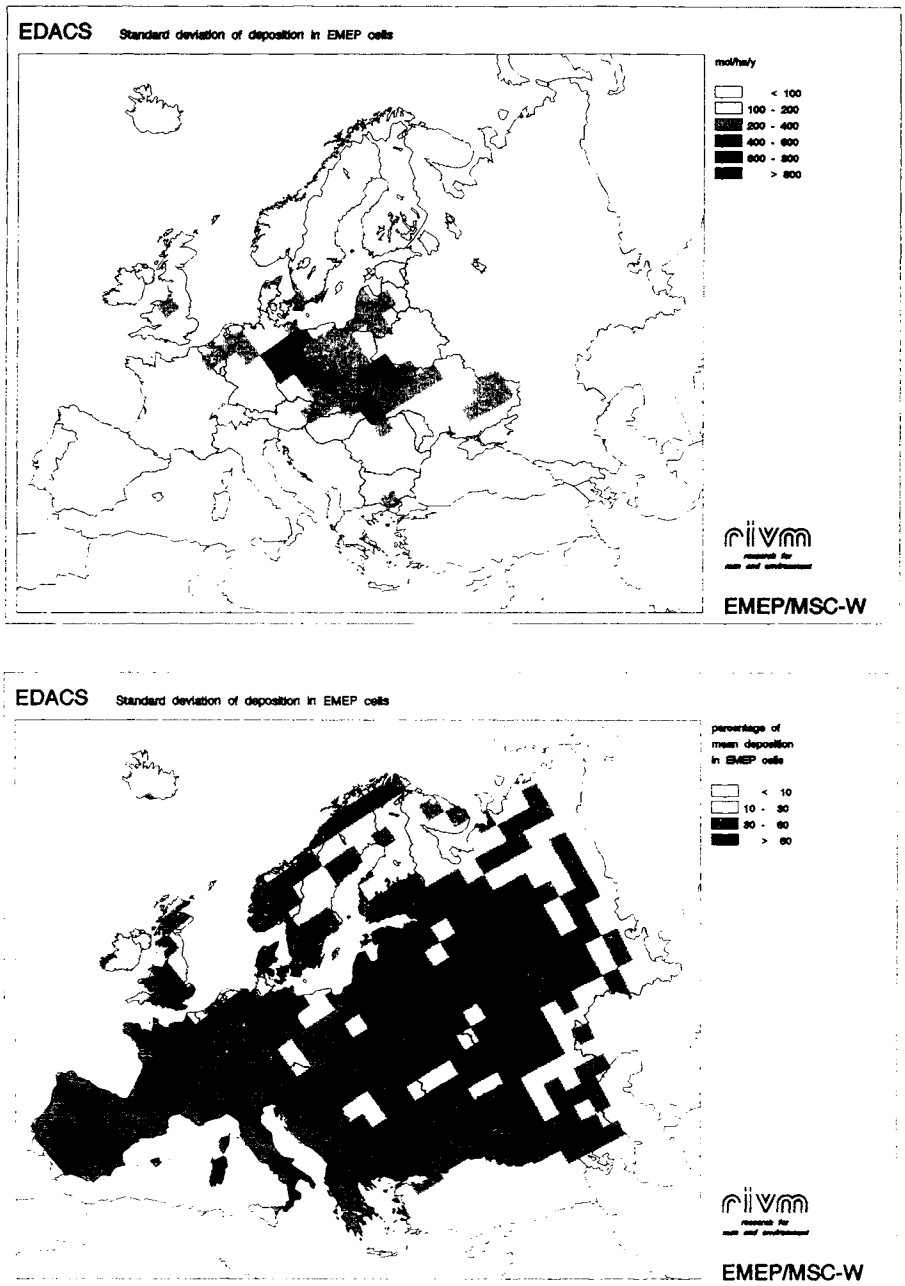

Figure 5 Standard deviation of the total deposition of EDACS grid cells per EMEP grid in absolute values in mol ha^{-1} $year^{-1}$ (above) and values relative to the total deposition per EMEP-grid cell in % (below).

5 Uncertainties

The maps shown in Section 4 have a limited accuracy and are therefore preliminary. The aim of these maps is to show the variations of the deposition of acidifying components on a small scale. Several uncertainties and shortcomings are present which need some discussion. We will adress these items here and will suggest a quantification of the uncertainties. Also some recommendations for improvements of these maps are given.

One of the main uncertainties in the maps is in the simple resistance model and especially the surface resistance parameterization for estimating the dry deposition of different gases and particles. The resistance model is a simple approach for a highly variable process. It assumes a constant flux layer, i.e. there are no surface inhomogeneities, edge effects or chemical reactions. How much these simplifications contribute to the total uncertainty in the annual average deposition fluxes has not been investigated. The uncertainty in the surface resistance parameterization is the largest uncertainty in this simplified scheme. Therefore more, and more accurate, parameterizations are needed for various vegetations and surfaces. Moreover there is a lack of measurements on which these parameterizations can be based especially for southern and eastern European climates and surfaces. This is needed to obtain parameterizations for use in LRT models which are valid for the whole of Europe. Part of the uncertainty in the surface resistance parameterization is due to the mismatch between the available parameterizations for a limited number of landcover types and the landuse classifications used in the RIVM land-use data base.
Surface wetness is found to be one of the major factors influencing the deposition process. In the present version of EDACS only rain and an indication of dew is used. In the next version the dew amount will be modelled in more detail using appropriate surface properties. The evaporation of rain and dew will also be parameterized. This means that an administration of the available energy and moisture flux during the day has to be made. An indicator on the presence and condition of a snow layer will also be taken into account.
The overall uncertainty in the surface resistance due to the above factors is different for each component and surface type. This uncertainty, on a annual basis, is a few tens percent points but can easily exceed 100%.

In the current version of EDACS, the EMEP-LRT concentration maps on a 150x150 grid are used. The uncertainty in the concentrations are estimated at 40-70% by a statistical analysis with the EMEP measurements (Krüger, 1993). These concentrations represent the background situation in Europe. It is assumed that the concentration distribution within a grid is homogeneous. This is not the case in a grid which contains industrialised areas or many scattered sources such as of NH_3 and NO_x. For such conditions, subgrid concentration variations are present and will lead to underestimates of the deposition in that grid. To obtain an indication of the errors, a small-scale, short-range model can be useful here to resolve sub-grid concentration gradients for dense source areas. The uncertainty in the deposition in an EDACS grid cell due to these gradients is estimated at 25% (Berg and Schaug, 1994).

The deposition in EDACS is based on the EMEP-LRT concentrations which in turn are dependent on EMEP deposition estimates. The deposition in the EMEP-LRT model and in EDACS are calculated in different ways. By using other dry deposition velocities in EDACS a mass inconsistency, between the EMEP calculated deposition and the small scale maps by EDACS, is introduced. However, if the differences in the used deposition descriptions between the two models are not very large and non-systematic over a larger region, this will

not lead to large mass inconsistencies. In Figure 6 a comparison between the sulphur dry deposition per country estimated by EMEP and EDACS is shown. It can be seen that on average there is a good agreement indicating that for the model area the mass consistency is not violated to a large extent. However, for some countries the deviations can be as large as 50%. To avoid this mass inconsistency it is planned to implement the deposition module in the EMEP-LRT model. In this way the calculated concentration fields are consistent with the EDACS deposition description.

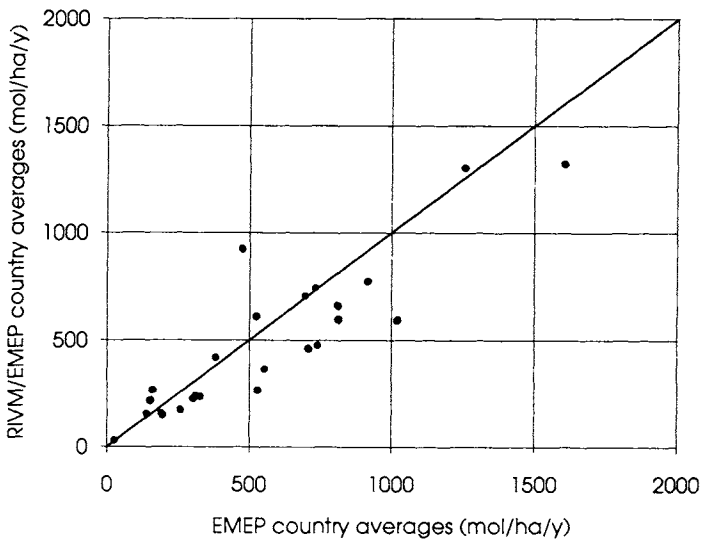

Figure 6 Comparison between the country averaged dry deposition of sulphur calculated with EMEP and EDACS (deposition in mol ha^{-1} $year^{-1}$).

In inferential modelling the concept is used that the surfaces, at which the deposition is calculated, should have a certain horizontal length. As an approximation this is about 100 times the reference height (Pasquill, 1972). The typical horizontal length scale of the surface, using 50m as the reference height, is 5 km. This means that only surfaces with larger length scales are modelled correctly. Variations in land cover on this scale are regularly present. At each surface transition the deposition is altered. Especially forest edges give rise to large enhancements of the deposition. This enhanced deposition can be dealt with in a very simplified way using correcting factors defined by e.g. van Pul *et al.* 1992b, Draaijers *et al.*, 1994. Since in this land-use data base only percentages of the land-use type per grid are given and not the geographical position, only a statistical approach of the uncertainty can be carried out. For instance, the enhanced deposition at forest edges as a whole for all forest stands in the Netherlands, is estimated at 10-30% (van Pul *et al.*, 1992b, Draaijers *et al.* 1994).

The accuracy of the presented results depends on the availability and quality of the input data such as the land-use map and the meteorological observations. In the gridded version of the RIVM land-use data base, forest is not subdivided into deciduous and coniferous. All forest is classified as coniferous forest. This will probably lead to overestimates of the deposition

velocity to deciduous forests for all components during winter. However, the stomatal resistance in winter will be large due to low temperatures. So this overestimate will be somewhat leveled out. A version in which the forest data are subdivided in the above categories will be available in 1995. In this new version the quality of the land-use data for some areas in Eastern Europe will also be improved.

The dry deposition is calculated on a daily basis. However, due to the daily averaging of the concentration and the deposition velocity, a loss in temporal correlation is introduced between the concentration and the deposition velocity. This error is component specific and is estimated to be smaller than 20% (van Pul *et al.*,1993). In the future the deposition will be calculated on a 6-hourly basis and the above error avoided.

The uncertainty in the wet deposition estimates is relatively small compared to the uncertainty in the dry deposition estimates. A comparison between the derived wet deposition maps and EMEP long-range transport model results was carried out by means of calculation of differences and ratios between the grids derived by the two methods (van Leeuwen *et al.*, 1994). In most parts of Europe deviations were found smaller than 200 mol/ha/year in absolute terms for individual components and smaller than 50% in relative terms. The uncertainty in the wet deposition is most important in areas where wet deposition is equal to or higher than dry deposition. In such areas, however, wet deposition usually shows a smooth pattern. This is not true for mountainous regions where additional deposition pathways such as fog and cloud deposition are present. Corrections can be applied to the wet deposition if local data on fog and cloud composition, occurrence and liquid water content are available (Fowler, 1991).

The overall uncertainty in the depostion maps consists of the above-mentioned uncertainties. However, the uncertainty in the surface resistance and the occurence of sub-grid concentration gradients will act as the largest uncertainty sources. Given these uncertainty estimates, the uncertainty in the deposition of a component of a 1/6°x1/6° lat/long gridcell is typically 100-200%. However, the uncertainty in the total potential acidification map is smaller because the total acidification consists of components such as wet deposition which have a smaller uncertainty .

6 Conclusions

In this paper a description is given of the EDACS model, with which the deposition of acidifying components on a small scale over Europe is calculated. Dry deposition velocity fields are constructed from a detailed land-use map (1/6°x1/6° lat/long, made by RIVM) and meteorological information using a detailed parameterization of the dry deposition process. These small-scale dry deposition velocity fields are combined with air-concentration fields (taken from the EMEP Long range transport model) to yield dry deposition amounts on a small scale. Wet deposition is also estimated to obtain a total acidifying deposition map over Europe (van Leeuwen *et al.*, 1994). These deposition fields clearly reflect the spatially detailed land-use information and the large-scale concentration pattern over Europe. With these fields a better match is obtained between the critical and actual loads when ecosystems are concerned.
The presented deposition fields are preliminary because of several shortcomings present in the method and data bases. An update of the deposition fields and calculations for more recent years will be available in 1995. A more thorough uncertainty analysis of the deposition maps,

a validation with (throughfall and micro-meteorological) measurements and corrections on the wet deposition caused by cloud and fog deposition will also be carried out.

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