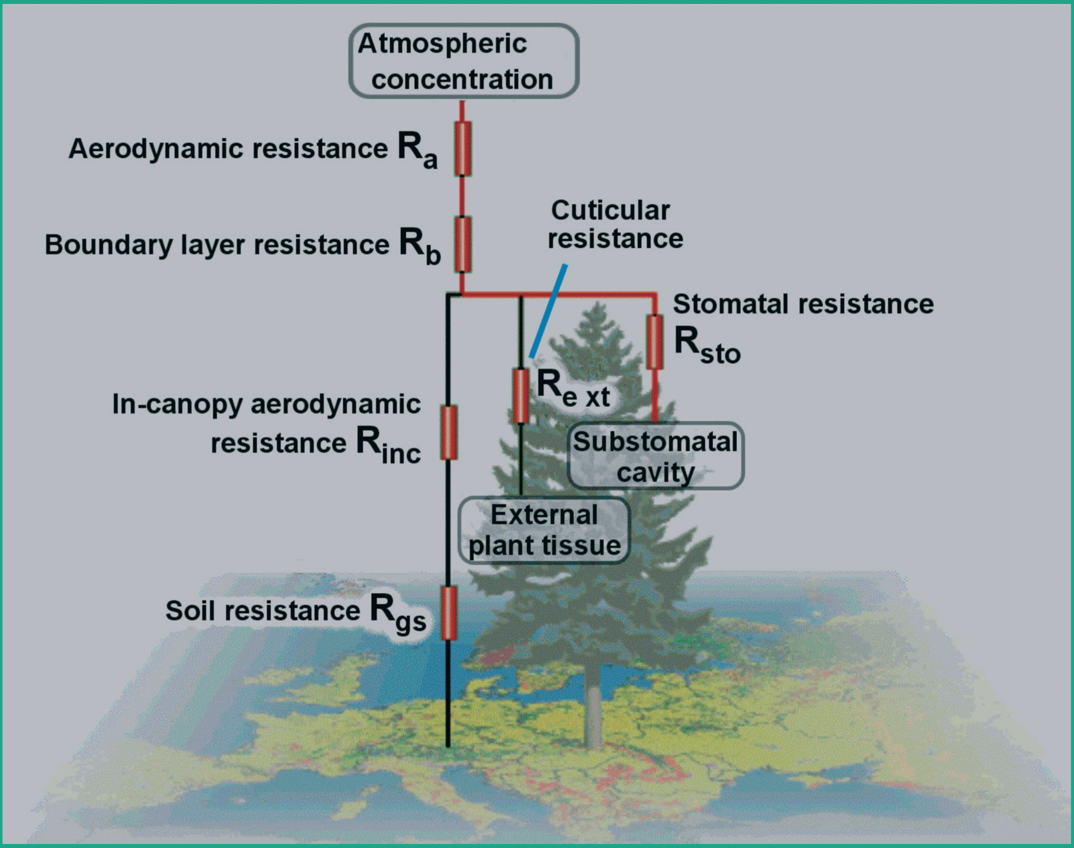
Mapping Manual 2004



Chapter 2 is an update and extension of the 1996 Mapping Manual. It has been re-edited by a revision group led by David Fowler and Ron Smith.

www.icpmapping.org

2.1 General remarks and objectives

The purpose of this chapter is to provide guidance to the participating countries in the generation of concentration level and deposition load maps for a range of pollutants for comparison with critical level/load maps. The document is intended as a general reference, with links into the recent literature. While specific recommendations are provided, the procedures are described in outline and the reader is referred to specialist publications for the measurenent and modelling approaches described here.

Total deposition is the sum of dry (turbulent flux of gases and particles to the surface), wet (via rain, snow or hail) and fog and cloud water deposition. All three pathways should be accounted for, but these deposition pathways differ so fundamentally that it is proposed to determine them separately and combine the quantifications to total deposition estimates (Hicks et al. 1993).

There are two main objectives for Europe-wide mapping of concentrations and depositions:

The first aim is to construct exceedance maps relative to critical levels and loads, which are then allocated to emissions in different countries. The thus obtained transfer coefficients between emission in all European countries and exceedances in each grid cell of the Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Euerope (EMEP) is particularly well suited to provide scientific results for a) implementation of, and compliance with, existing LRTAP Convention protocols and b) their review and extension. Such data are crucial for Integrated Assessment Modelling, and concentration and deposition maps from EMEP model calculations are designed for this purpose. Considering all the uncertainties inherent in the discussions of future emission-deposition relationships, the countries´ economies and energy demands, and our knowledge of environmental effects, maps provided at a scale of 150 x 150 km² have proved adequate for the development of international protocols. The new EMEP Eulerian model provides higher resolution concentration and deposition fields at 50 x 50 km².

The second aim is to map concentrations and depositions which can be used for effects assessments in specific ecosystems. Such data are needed with a much better spatial resolution than required for the Integrated Assessment Modelling. National Focal Centers should aim at a sufficient spatial resolution for the assessment process, making use of national models and measurement networks. EMEP will continue to provide background, long-range transported air components which can be used as boundary conditions for such national models. The chapter provides a range of different techniques for the provision of maps of concentration and deposition, depending on the resources and ambition of the country, with the EMEP values regarded as default data allowing the assessment process to be completed everywhere.

Within the countries of Europe, the expertise and facilities for measurement of concentrations and fluxes of pollutants is very variable. The extent to which the methods presented can be applied is therefore variable and it is necessary to show the range of options available. It is important to stress that involvement in the measurement and modelling activities is highly desirable as a part of the cooperation in the assessment process. The development of satisfactory strategies for control of pollutant emissions requires full participation in the underlying science as well as the political process.

The spatial scale aimed at within the Mapping Programme differs from the site-oriented approach in the ECE International Cooperative Programmes (ICPs) on Integrated Monitoring and on Forests (Level II): what is needed is not data for isolated sites but ecosystem-specific regional estimates for all of Europe. Therefore, the focus of this chapter is on methods that are capable of producing critical level/load exceedance maps for whole countries, using long-range transport models in combination with concentration measurements, small-scale dry deposition modelling and interpolated wet deposition measurement data from

(ICP, EMEP, national, regional) monitoring sites. Site-level measurements of dry deposition and canopy throughfall for example are intended for use for deriving process parametrisations (mainly micrometeorological measurements) and for independent model validation (mainly throughfall measurements; see Chapter 2.3.1, 2.3.2 and 2.3.10).

Consequently, this chapter is much less detailed in its description of field and laboratory methods than the respective sections in these ICPs´ Manuals and in the EMEP publications on monitoring methodology (EDC 1993 (ICP Integrated Monitoring); UNECE ICP Forests 1999; EMEP/CCC 1996).

A range of publications is available with detailed descriptions of the underlying theory and methods as well as modelling. These include proceedings from several ECE Workshops dealing with this subject, most notably the 1992 “Workshop on Deposition” in Göteborg, Sweden (Lövblad et al. 1993), the 1993 “Workshop on the Accuracy of Measurements” with WMO sponsored sessions on “Determining the Representativeness of Measured Parameters in a Given Grid Square as Compared to Model Calculations” in Passau, Germany (Berg and Schaug 1994), and in Erisman and Draaijers (1995), Sutton et al. (1998), Slanina (1996), Fowler et al. (1995a, 2001a) and ICP Forests Manual (UNECE 1999). Supplementary information can be found in other workshop proceedings and in scientific journals.

NFCs are strongly advised to ensure that the monitoring and modelling methodologies described in the publications listed above are documented in the development or validation of a database for national concentration and deposition (and critical level and load exceedance) maps. Compatibility of these maps with other national maps within the Mapping Programme as well as with monitoring methods employed within other deposition monitoring programmes under the Convention (ICPs on Forests and on Integrated Monitoring; EMEP) is very important.

2.1.1 Mapping resolution and application of Critical Loads

The use of deposition data with Critical Loads data very often involves different scales of the different datasources. In most cases the Critical Loads data are provided at a finer resolution than the deposition data. To avoid misleading the reader it is essential that the different scales be noted in the legend. However, it is important to note that the application of deposition data at a coarse scale relative to the high resolution Critial Load data usually gives exceedances which are systematically underestimated (see 2.3.2).

2.2 Mapped items

The following items are to be mapped for each country:

For critical level exceedance maps:
- ozone concentration ($AOT40$ values) and ozone flux,
- sulfur dioxide concentration,
- nitrogen dioxide concentration,
- ammonia concentration.

As input into deposition and critical load computations:
- precipitation amount and other meteorological parameters (as required),
- wet, dry, cloudwater/fog and aerosol deposition.

For critical load exceedance maps:
- oxidized sulfur (SO_x) deposition (total and non-sea-salt),
- oxidized nitrogen (NO_y) deposition,
- reduced nitrogen (NH_x) deposition,
- base cation and chloride deposition (total and non-sea-salt),
- total nitrogen deposition,
- total potential acid deposition.

Heavy Metal deposition (pending agreement on critical loads):

- aerosol and wet deposition of lead *(Pb)*, cadmium *(Cd)*, zinc *(Zn)*, and copper *(Cu)* (suggested as a minimum),
- total deposition of mercury *(Hg)*.

For all maps, the most recent available data should be used, not going further back in time than five years.

The mapping items concerning gaseous pollutant levels to be mapped are listed in detail in Chapter 3 of this manual. The concentrations and averaging periods are based upon the findings of the ECE workshops on critical levels held at Bad Harzburg (Germany) in 1988, in Egham (U.K.) in 1992, Berne (Switzerland) in 1993, St. Gallen (Switzerland) in 1995, Kuopio (Finland) in 1996, Harrogate (UK) in 2002, and Göteborg (Sweden) in 2002.

2.3 Methods of mapping, their underlying assumptions and data requirements

2.3.1 Linkage to emission inventories

Several methods are available to estimate boundary layer atmospheric concentrations and wet, dry, and cloud-water/fog deposition on different scales of time and space.

Only some of these methods are linked to emission inventories (see Table 2.2 / Page II-20): those that are based on emission inventories (group A: EMEP and national long-range transport modelling, also combined with small-scale dry deposition models) can be distinguished from those that are independent from emission inventories (group B: EMEP and national monitoring of air concentration and wet deposition, site-level micrometeorological and throughfall measurements).

The objectives of methods in *group A* (linked to emissions) are (1) regional present and past situation analysis, and (2) providing a basis for scenario analysis and therefore emission reduction negotiations. The objectives of the measurement activities in *group B* (not linked to emissions) are (1) model evaluation, and (2) site-specific effects analysis (see Chapter 2.1 and Table 2.2).

2.3.2 Quantification and mapping methods: Scales of time and space

For the ***time scale***, annual deposition rates are sufficient in order to determine critical load exceedances, whereas for critical level exceedances, short-term information is sometimes needed (see Chapters 3 and 4). However, there can be substantial variability from year to year with deposition, for example with changes in rainfall amount, and it is recommended that a 3 year average deposition is an appropriate time average for calculation of critical load exceedances.

Three groups of methods exist for various ***spatial scales:***

Long-range transport models - the most widely used source of deposition data, providing inputs at a range of spatial scales (50 x 50 km² for EMEP, 5 x 5 km² for country scale models)

Nested high-resolution models - are used to provide higher spatial resolution (1 x 1 km²)

Site or catchment specific measurements - providing local deposition estimates (1 to 1000 ha); methods include:

Wet deposition collectors (point or field scale)

Micrometeorology (field scale)

Throughfall methods (canopy scale)

Catchment mass balance (landscape scale).

Long-range transport* (LRT) *models provide average estimates of concentrations and deposition rates for large grid squares (typically 5 x 5 km² to the EMEP Eulerian model´s 50 x 50 km²). They belong to group A as defined above: they are based on emission inventories and are therefore most suitable for scenario analyses and country to country budgets (‘blame matrices’) used in emission reduction negotiations (if the model domain is more than one country). Long-range transport model results are available for the European UNECE region from EMEP and can be taken as default or reference model outputs.

Standard multiannual concentrations from EMEP are given as one number per year per component per 50 x 50 km² grid square and deposition fluxes are provided either as average deposition to the 50 x 50 km² grid square or as ecosystem specific deposition estimates. EMEP model output can be provided for shorter time periods, but with the overall constraint that one of the major inputs, the emission inventory, is often provided only as an annual total.

The scale at which critical levels/loads and concentration/deposition are mapped greatly influences the magnitude of exceedance values (Spranger et al. 2001, Bak 2001, Lövblad 1996, Smith et al. 1995). For example, if the average value from a 50 x 50 km² grid square is matched to critical loads on the 250 squares of 1 x 1 km² within the 50 x 50 km² grid square, there will be generally be less critical load exceedance than if the deposition were available at the 1 x 1 km² scale. The only circumstance in which this underestimate would not occur would be if the high deposition locations matched the high critical load locations. Over many areas of Europe, exactly the opposite occurs. In many areas of complex terrain the parts of the landscape receiving the largest deposition, such as the higher areas in the mountains of North-West Europe, are also the most sensitive to the effects of deposition, for example acidification. The same holds true for forested areas, which tend to correlate with poor soils in large parts of Europe. This problem is worse for components with low local sources (NH_3, NO_x) because the within-grid distribution of sources is not reflected in the grid average estimation from a LRT model, but does markedly increase the within-grid variability of deposition and hence increases the critical load exceedances. As the current deposition estimates from EMEP are provided at a scale which is much larger than the scale of this spatial variability, the critical loads exceedances for these areas are underestimated.

These effects are minimised by estimating deposition to the smallest spatial scale possible. However, there is an underlying relationship that the critical load exceedances will increase as the spatial resolution of the deposition gets closer to that of the critical loads.

High resolution modelling.
A second group of methods tries to overcome these scale problems by applying smaller-scale "*inferential*" models using large-scale meteorology and concentration fields either obtained from LRT models (see above) or by interpolation of sufficiently dense measurement networks (see below).

Dry deposition is inferred by multiplying the concentration with the deposition velocity of the component of interest (Hicks et al. 1987, 1993). The latter is calculated using a resistance model in which the transport to and absorption or uptake of the component by the surface is described. Resistances are modelled using observations of meteorological parameters and parametrisation of surface exchange processes for different receptor surfaces and pollution climates as described in Erisman et al. (1994a), Smith et al. (2000), Nemitz et al. (2001), Emberson et al. (2000), Grünhage and Haenel (1997), Gauger et al. (2003). The deposition velocities of cloudwater/fog droplets can be similarly estimated by modelling momentum transfer (Fowler et al. 1993) and a similar technique has been used to estimate base cation deposition (Draaijers et al. 1995). Parameters determining the deposition velocity include atmospheric parameters (e.g. wind speed, temperature, radiation, relative humidity, atmospheric stability, cloud and/or fog frequency) and surface conditions (e.g. roughness, wetness, stomatal response, soil water). Unfortunately, up to now no reliable European-wide cloudwater/fog concentration fields are available, hampering cloudwater/fog deposition estimation on a European scale.

The land use maps used for this deposition modelling should be identical to the stockat-risk maps used for critical levels/loads mapping (see Chapter 6). In addition to the geographical position of sensitive ecosystems, land use type/vegetation type, vegetation height and crown coverage should be mapped as well on a scale that allows for correct allocation of deposition to all ecosystem types in the model domain.

Uncertainties of inferential deposition models are described in Chapter 2.3.10.

Site or catchment specific measurements. All methods based on *point measurements* (e.g., wet deposition measurements, micrometeorological dry deposition measurements, throughfall measurements) belong to group B, as they cannot be directly connected to emission inventories. Maps can only be produced directly from these measurements if the network is dense enough to account for spatial (and temporal) variations. This may be the case for networks measuring air concentrations of compounds with little spatial variation or for measurements of wet deposition in areas of simple terrain. Network (point) measurements should be interpolated using the kriging technique and it may be helpful to include monitoring data from neighbouring countries for interpolation. For some air concentrations, such as ammonia, or for rain concentrations in complex terrain, the required density of the measurement network could be too dense for practical application. In these cases, it is recomended that concentrations are obtained from less dense networks and that simple models are used to assist the interpolation, e.g. using altitude dependences. It is preferable to interpolate concentrations in rain or in air and then calculate the deposition at the receptor site using local estimates of rainfall and land-use specific ground-level dry deposition rates (see above).

Additional monitoring of air concentrations of gases in order to create a dense network as a basis for mapping can be made with diffusive samplers. These samplers, can be used for a number of gases (ozone, sulphur dioxide, nitrogen dioxide, nitrogen oxides (NO_x), ammonia, nitric acid, mercury, hydrogen chloride, etc.) The sampler provides average concentrations over a time period, normally from one week to one month. It is a simple and cheap complementary method, to be used in parallel to other methods providing also the temporal variability (Ferm and Svanberg 1998, Ferm 2001, Sjöberg et al. 2001).

Wet deposition. In most cases, the long-term spatial variation in wet deposition within regions is determined mostly by variations in precipitation amount and less by variations in concentrations in rain or snow. In addition, precipitation amounts are mostly available from relatively dense meteorological networks. Therefore, if concentration variations are small, maps of annual wet deposition rates should not be drawn by interpolating measured wet deposition rates; it is recommended to interpolate measured solute concentrations and estimate the wet deposition as the product of the mapped solute concentration and the precipitation amount, the latter provided by the meteorological service for the country. This is an important step because the precipitation fields are defined by dense networks of collectors. An additional and very important enhancement of wet deposition occurs in the uplands of Northern Europe, due to wash out of orographic cloud by falling rain or snow. As networks do not generally measure at high elevation in complex terrain, these effects are generally omitted from network measurements. The underlying physical process is well documented and the effects may be modelled using the network data (Dore et al. 1992, Fowler et al. 1995b).

Dry and cloudwater/fog deposition can be estimated from concentration measurements of airborne substances by *micrometeorological measurements* at the process level (for SO_2: Fowler et al. 2001c; for NH_3: Flechard and Fowler 1998; for cloud: Beswick et al. 1991). During the last decade it has become possible to make micrometeorologically based long-term flux measurements (i.e. continuous flux measurements over more than a year). This has been demonstrated for O_3, NO_x and SO_2 (LIFE project, Erisman et al. 1998a) and for CO_2 and H_2O (Aubinet et al. 2000). Such measurements give information about the seasonal and interannual variability in the fluxes.

These measurements of deposition fluxes have therefore become straightforward and can be applied to all pollutant gases. However, the primary purpose of the measurements is to provide the parameters for modelling as the tool for extrapolation over the landscape as the measurement stations are expensive to operate. Thus the number of dry deposition stations could not realistically be sufficiently large to interpolate fluxes over spatial scales directly. There are low cost micrometeorological methods, such as the Time Averaged Gradient (TAG) system (Fowler et al. 2001b). These will provide the means of obtaining deposition parameters for many more representative terrestrial surfaces in Europe.

The methods mentioned here only work if stringent prerequisites concerning micrometeorological variables (e.g., surface homogeneity) are fulfilled. They cannot be directly extrapolated, but the process knowledge obtained from such measurements can be parametrized in inferential models and fluxes can be mapped using this information (see high resolution modelling above).

Throughfall and stemflow measurements can be used to estimate the site-specific total deposition of sulfur to plant canopies, mainly forests (wet plus dry plus cloudwater/fog). The data are useful for parallel effect studies, in order to estimate deposition rates on the basis of field data available from existing monitoring programmes and to validate other deposition estimates. They will also provide knowledge on the seasonal variation and the trends of deposition. In many cases throughfall monitoring is considered to be sufficient, and stemflow is only measured for some tree species, for which it is known to be of importance (e.g. beech trees) (UNECE 1999). In practice, it is not generally possible to determine the total deposition of substances for which uptake or leaching within the canopy is large relative to the deposited amounts (e.g. nitrate, ammonia, and calcium, potassium and magnesium) from throughfall measurements.

Throughfall measurements are cheaper and generally easier to perform than micrometeorological measurements. They also give a good overview of the deposition situation in the forest, not only for sulfur but also for nitrogen compounds. Recent Swedish experiences have highlighted the problems with comparing throughfall measurements with wet deposition when the dry deposition contribution to the total is very low (Westling *pers. comm.*), as is now the case for sulphur in many areas of Europe. Large uncertainties in wet deposition at wind-exposed sites have been shown with field intercomparison studies (Draaijers et al. 2001). Even if it is not possible to estimate the total deposition of nitrogen with this method, a lower limit can be set. Sampling considerations (e.g. location of collectors, species composition, spatialy variability) are very important for achieving good results and sampling requirements are described in detail in the ICP Forests Manual (UNECE 1999) and in review articles such as Draaijers et al. (1996a) and Erisman et al. (1994b).

In order to interpret the data, the relation between total deposition and throughfall can be expressed:

(2.1)

$$Total\ DEP = DRY + WET + Cl/Fog = THF - CEX$$

where:

THF = Flux in throughfall (plus stemflow)

$DRY, WET, Cl/Fog$ = dry, wet, cloudwater/fog deposition

CEX = canopy exchange; $CEX > 0$ for leaching, $CEX < 0$ for uptake

When $CEX=0$, the dry deposition can be estimated as the difference between total flux in throughfall and independent measurements of wet and cloudwater/fog deposition. If CEX differs from 0, dry deposition cannot be distinguished from internal cycling. This method can give large overestimates of the true deposition flux ($CEX>0$), due to canopy leaching (for some base cations), or large underestimates of the true deposition flux ($CEX<0$), due to canopy uptake (e.g., for nitrogen compounds and protons). Therefore, throughfall plus stemflow fluxes should be interpreted as upper bounds of total base cation deposition and as lower bounds of total nitrogen and proton deposition.

In some cases, the total deposition to plant canopies can be deduced from throughfall and precipitation measurements in the open field using empirical *canopy budget models.* These models are not always applicable and should if used be applied with care. The method by Ulrich (1983), which uses the relation of dry vs. total deposition of sodium as an indicator of dry vs. total deposition of other elements, is used most widely. However, some of its assumptions, such as the constant (in time and with respect to substances) ratio of dry particulate deposition vs. wet deposition rates, are questionable. For several reasons (e.g., different deposition and canopy uptake processes), it cannot be applied for determining total nitrogen deposition to the forest ecosystem (Bredemeier 1988, Spranger 1992). A modification (Beier et al. 1992) and an extension of the model (Draaijers and Erisman 1995) mitigate some of the methodological problems, even though they have not yet been properly evaluated under all circumstances (see Draaijers et al. 1996a,b), but there remains an issue with canopy modelling that, in many cases, a cation surplus exists in the throughfall solutions indicating that major substances have not been measured.

The calibrated watershed method integrates deposition fluxes over a scale compatible to critical load computations for example for lakes and surface waters. However, major fluxes to the groundwater and soil exchange have to be accounted for. It is most useful for conservative elements (e.g., S, Na, Cl) in areas with clearly delineated watersheds. The data are useful to validate deposition estimates derived from modelling.

Concluding remarks. LRT Models will normally be used to calculate patterns of concentration and deposition across Europe. High resolution inferential models may be used to calculate patterns within countries or regions. The role of measurements in the process of mapping is twofold. Low cost devices, such as passive samplers or bulk samplers, may be used to observe regional patterns of concentrations and wet deposition. If the measurement strategy is good, these measured patterns will be more reliable than those calculated. However for dry deposition the situation is more complex. The deposition rate depends on atmospheric conditions as well as on ecosystem type. Recent developments of low cost dry deposition monitors (Fowler et al. 2001b) will make it possible to determine regional patterns of dry deposition in the near future. But still the application in dense networks is expected to be limited. The limitations of using throughfall measurement networks for estimating regional patterns of total deposition have been described above.

The results of measurements on the other hand are necessary to test, validate and improve model parameterisations. Defendable estimates of the deposition need to be validated by measurements to relevant European ecosystems. It is recommended to establish a network across Europe where detailed measurements of dry and wet deposition will be made. This may be linked to existing monitoring networks such as the EMEP network and the Level II Programme of ICP Forests.

2.3.3 Mapping meteorological parameters

Meteorological parameters are required inputs for most critical levels or critical loads calculations. The data requirements and data provision will vary from country to country. Data are generally avaialable from national weather services. European data can be obtained from European Centre for Medium-Range Weather Forecasts (ECMWF, website: www.ecmwf.int), who provide modelled data based on observations within Europe, and there are other sources for some data such as the US EPA/NCAR global precipitation database.

Precipitation amounts are needed for critical load computations, for wet deposition mapping (see Chapter 2.3.2), and for surface wetness parametrisations (also for materials: “time of wetness”; see Chapter 4).

Fog and cloud occurence is needed for cloudwater/fog deposition estimates.

Wind speed, temperature and *radiation* are basic requirements for the inferential modelling of dry deposition. Additionally, *relative humidity*, *soil water deficit*, and *atmospheric stability* are often required.

The availability of accurate local meteorological data is often a constraint to detailled local high resolution modelling, and therefore the success of models in improving deposition estimates to specific ecosystems may depend as much on the availability of quality meteorological data as on the quality of the local concentration estimates or measurements.

2.3.4 Mapping ozone (O_3) concentrations and deposition

Ozone concentration data are required to generate maps showing areas where the critical level is exceeded (see Chapter 3.2.4). Data may be available from photochemistry/transport modelling (see *(a.)* and *(b.)* below) or from monitoring networks (see *(c.)* below).

The concentrations of ozone close to terrestrial surfaces (e.g. within 1 m) show a large spatial variability in both rural and urban areas. For urban areas, this variability is mainly caused by chemical consumption of ozone by NO, which is locally emitted. For rural areas away from local sources, this variability is largely caused by spatial and temporal changes in the degree to which individual sites are vertically 'connected' to the main reservoir of ozone in the boundary layer. Like in urban areas O_3 might be consumed by the reaction with NO which can be emitted from bacterial processes in the soil (PORG 1997)

To provide a spatial resolution of the ozone exposure on a horizontal scale which reflects the variations in the orography it is helpful to produce the ozone concentration field at a grid of 1 x 1 km² cell-size at least (see *(b.)* and *(c.)* below). As the critical levels are based on the concentration measured in the turbulent layer near the receptor, ozone levels modelled or measured at higher distances from the ground are not directly related to the observed effects. Therefore a surface-type specific correction should be applied for assessing exceedances of critical levels, but it is hardly possible to quantify the correction from monitoring data to dose-effect data at present (Fuhrer 2002).

The supply of ozone to vegetation is provided by atmospheric turbulence and hence wind speed and the thermal structure of air close to the ground. The deposition of ozone on terrestrial surfaces and vegetation causes a vertical gradient of the ozone concentration, which is largely determined by the sink activity of the soil-vegetation system. Maps of O_3 deposition can be produced from inferential modelling based on parameters obtained from long-term measurements and land-use information (Emberson at al 2000).

(a.) <u>*LRT model results*</u>

EMEP/MSC-W has available calculations of ozone concentrations in 50 x 50 km² gridsquares with hourly time-resolution and also deposition to specified land cover types, e.g. forests, arable crops, etc. The model calculations are available for a notional height of 45 m above ground and scaling algorithms are available to provide output at lower heights, particularly 3 m and 1 m. $AOT40$ for crops and forests is also available, as well as their changes in each grid square per unit of changed country-emissions of VOC and NO_x. The calculations are based on new EMEP/CORINAIR emissions with 11 source sectors and VOC speciation specified for each sector. The model also includes biogenic VOC emissions from forests.

(b.) <u>*High resolution modelling*</u>

The low spatial resolution of LRT models does not match with the resolution required for the evaluation of ozone exposure of forests ecosystems, and estimates of ozone exposure can be improved by local scale modelling within the 50 x 50 km² EMEP square. The necessary concentration values at receptor level can be obtained at high resolution from the large scale model average values by correcting them for local emission of nitrogen oxides, orography and deposition. The computation of deposition velocities and deposition fluxes requires land-use maps (see Annex II), as well as meteorological data.

One method of adjusting for local scale effects is to adjust the diurnal cycle in concentrations from the LRT by accounting for the dependence of ozone concentrations on local orography (PORG 1997, Coyle et al. 2002). The elevation of a particular location determines the extent to which it experiences the influence of air from the free troposphere and from the boundary layer. Based on data from the EUROTRAC-TOR and EMEP monitoring programs as well as on results in literature, this dependence can be modelled and combined with small scale orographical data.

High-resolution $AOT40$ maps can also be computed by using other atmospheric transport and photochemistry models, provided that the output data are high-resolution results modelled over a long (for AOT_F: April-September), continuous time period, and that the model results are evaluated with measurements as well as with MSC-W model results.

(c.) Monitoring and interpolating ozone concentrations and fluxes

The monitoring of ozone is necessary to establish or to validate exceedance maps as well as for the verification of the long range transport and chemistry models.

Over large parts of Europe and particularly in South and East Europe there are very few available data. Efforts should be made to obtain data where monitoring stations exist. Elsewhere, the establishment of a network of monitoring stations is strongly recommended. Stations should be linked to the EMEP network.

To provide data from such a network that are representative for an extensive area it is recommended that the monitoring stations be sited at rural locations avoiding local sources of the oxides of nitrogen such as roads. Anshelm and Gauger (2001) developed a method to classify the suitability of monitoring sites for mapping concentrations.

Some countries have already a sufficiently ‘dense’ network. If the number of monitoring stations needs to be increased, it is recommended to install them at various altitudes and/or at various distances from the emitters of ozone precursors. Stations at urban locations are not representative for extensive areas, but they may be required for differentiating areas with rural and with urban pollution conditions and for population exposure assessments through mapping exceedances of air quality guidelines based on human health.

The preferred sampling height is 3-5 m and the monitoring station requires an open aspect without the presence of trees or other tall vegetation in the proximity of the sample intake. Appropriate recommendations for sampling and calibration are available from the Chemical Coordinating Centre of EMEP.

More or less simple interpolation procedures exist to obtain an estimate of the exposure of terrestrial surfaces to ozone using topographic and other information. Altitude may be used here as an indicator for the degree to which areas are ‘connected’ to the ozone reservoir in the boundary layer (see also *b.)*).

One possibility is to interpolate the measured hourly ozone values and then to compute seasonal dose-parameters such as $AOT40$ (see chapter 3.2.4) on the basis of those hourly maps (Loibl and Smidt 1996). But in general it will be easier to calculate the values of the required dose-parameters for each monitoring station first and then to use these values for the spatial application of a regression model. For the interpolation of $AOT40$ values relationships with altitude (height above mean sea level) have been used in the UK (Fowler et al. 1995c) and in the Nordic countries (Lövblad et al. 1996). Relationships with relative height (the height of the site of interest above the valley ground within a certain distance) are applied in Alpine regions (Loibl and Smidt 1996). Such relationships are reasonably consistent on the regional scale (100 to 500 km) with the exception of coastal and urban areas, but must be established from relatively dense monitoring networks. Dividing the study area into subregions and evaluating region-specific influences on the O_3 concentration or exposure may enhance the quality of the interpolation. Data assimilation, combining observed and LRT modelled concentrations, is another method recently applied to provide improved ozone concentration fields (Flemming 2003).

2.3.5 Mapping sulfur dioxide (SO_2) concentrations and oxidised sulfur (SO_x) deposition

Data on SO_2 gas concentrations, sulfate (SO_4^{2-}) aerosol concentrations and SO_4^{2-} concentrations in rain are required to generate maps showing areas where the critical level is exceeded. Data are available from long-range transport modelling, possibly coupled to small-scale modelling (see *(a.)* and *(b.)* below), or from monitoring networks (see *(c.)* below).

SO_2, in contrast to ozone or sulfate aerosol, is a primary pollutant. It is emitted by both high (e.g. power plants) and low (e.g. households) sources. Therefore the spatial variability of concentrations tends to be higher than that of ozone and sulfate aerosol but lower than that of ammonia. Close to urban areas, the concentrations of rural sulfur dioxide are elevated and this effect should be modelled explicitly where possible, for example by using urban concentration measurements and areas of urbanisation to model the urban effect (a similar method was used by Stedman et al. (1997) to model NO_x and NO_2 near roads).

For rural areas away from local sources, spatial variability is largely caused by spatial and temporal changes in the degree to which individual sites are vertically 'connected' to the main reservoir in the boundary layer (see preceding subchapter on ozone).

As for ozone, the SO_2 levels measured 3-5 m above ground are not directly related to the observed effects, since dry deposition causes a systematic vertical concentration gradient towards the surface, while the critical levels are based on the concentration measured close to the receptor. However, surface-type specific corrections are not generally applied and measured/modelled values usually taken uncorrected.

Non-sea-salt inputs of sulfur are needed in the critical loads framework, since critical loads are generally compared with anthropogenic S (and N) (see Chapter 5.3.2). Consequently, the base cation and chloride deposition in the charge balance - from which critical loads are derived with the SMB model - have to be corrected for sea salt contributions as well. Natural marine emissions of reduced sulfur compounds (especially Dimethylsulfate, DMS) are included into the EMEP emission data base (and therefore EMEP model results), whereas sea-salt emissions are not.

Depositions of base cations, sulphur and chloride (given in equivalents) are corrected by assuming that either all sodium or all chloride is derived from sea salts, and that the relations between ions are the same as in sea water (after Lyman and Fleming 1940, cited in Sverdrup 1946):

(2.2)

$$X^*_{dep} = X_{dep} - r_{XY} \cdot Y_{dep}$$

where

X = Ca, Mg, K, Na, Cl or SO_4,

Y = Na or Cl,

r_{XY} = is the ratio of ions X to Y in seawater and the star denotes the sea-salt corrected deposition. Ratios r_{XY} are shown in Table 2.1 with 3-decimal accuracy.

Table 2.1: Ion ratios $r_{XY}=[X]/[Y]$ (in eq/eq) in seawater

Y	X					
	Ca	Mg	K	Na	Cl	SO_4
Na	0.043	0.228	0.021	1	1.166	0.120
Cl	0.037	0.195	0.018	0.858	1	0.103

Note that for arbitrary ions X, Y and Z the relationships $r_{YX} = 1/r_{XY}$ and $r_{XY} \cdot r_{YZ} = r_{XZ}$ hold. If Na (Cl) is chosen to correct for sea salts, $Na^*_{dep}=0$ $(Cl^*_{dep}=0)$.

Using such a correction will only yield reliable estimates on non-sea-salt S, Mg, Ca, K, and Cl in areas where sea-salt is the only source of Na in ambient air. This generally will be the case in western and northern Europe. In some parts of southern and south-eastern Europe, however, significant quantities of Na in the atmosphere originate from wind-blown evaporates and applying the sea-salt correction there will result in underestimated non-sea-salt concentrations.

(a.) LRT model results

As for ozone, EMEP/MSC-W has available calculations of sulfur dioxide concentrations and sulfate aerosol concentrations in 50 x 50 km² grid-squares with hourly time-resolution and also deposition to specified land cover types, e.g. forests, arable crops, etc. The model calculations are available for a notional height of 45 m above ground and scaling algorithms are available to provide output at lower heights, particularly 3 m and 1 m. Daily concentrations and deposition of sulfate in rain are also available. Concentrations allocated to emissions in separate countries are available with monthly time-resolution.

(b.) High resolution modelling

Procedures as the ones described for ozone in the preceding subchapter can be applied for SO_2. However, since SO_2 is a primary pollutant, the most important factor causing variability is local emission, which has to be accounted for in the high-resolution model. SO_2 deposition velocities depend mostly on stomatal opening (stomatal pathway: to be parametrized using vegetation type/land use, and meteorology data), on surface wetness, and on NH_3 concentrations. When surfaces are wet, and at humidities >90%, surface resistances to deposition become very low and the flux is mostly determined by atmospheric resistances (Erisman et al. 1994a).

For SO_4^{2-} aerosol, dry deposition is highest for forests or other rough surfaces that are far from SO_2 emission sources (Gallagher et al. 1997). Sulfate deposition velocities can be estimated using Slinn´s (1982) or a similar simple particle deposition model (e.g., Erisman et al. 1995, Ruijgrok et al. 1996) but there is still significant research effort focussed on improving these estimates and it is reasonable to use site specific models to improve on the LRT sulfate aerosol deposition.

Wet deposition maps can be produced from monitoring data according to the methods described in Chapter 2.3.2, including orographic effects where applicable. The most important factor in improving wet deposition estimation at the local scale is the availability of rainfall maps derived from dense networks of rainfall collectors. If the density of the concentration monitoring network is not sufficient, EMEP/MSC-W modelled concentration data can be combined with local rainfall maps for improved estimates of wet deposition.

c.) Monitoring and interpolating concentrations

Measurement stations for SO_2 should be set up in areas that are not directly affected by local emitters. As for the EMEP network, and contrary to regional, national or EU health-related programmes, the main objective of a measurement network is not necessarily to determine the highest ambient concentrations (leading to high local critical level exceedances) but rather the large-scale concentrations that are due to long-range transport. Individual measurement stations should be representative for a maximum area, thus making interpolation and mapping of concentrations unaffected by local sources possible. Criteria for siting measurement stations are listed in, e.g., EMEP/CCC (1996).

Due to the fact that SO_2 is a primary pollutant, the measurement network density, especially in emitter areas such as Central Europe, has to be high so that the interpolation error (determined, e.g., by variogram analysis when using kriging) is minimal relative to the measured values. The same is true for mountainous areas due to the vertical gradients present. As secondary pollutants with relatively slowly varying concentrations, the measurement network density for $SO4^{2-}$ aerosol and for SO_4^{2-} in rain can be much lower than for SO_2. Methods to determine (and data on) representativity of measurements, as well as their precision and accuracy, are listed in Berg and Schaug (1994).

Maps can be produced from measurements by interpolation if all criteria mentioned above (accuracy/precision, representativity) are fulfilled. For some applications a blending height approach is appropriate, where ground-level observations are extrapolated to 50 m height (the blending height) above the ground, using a resistance model. At this height the concentration is less dependent on the surface processes and can be interpolated over larger areas (Erisman and Draaijers 1995). The preferred interpolation procedure is kriging, which also provides an estimated interpolation error.

2.3.6 Mapping nitrogen oxides (NO_x) concentrations and oxidised nitrogen (NO_y) deposition

Data are available from long-range transport modelling, possibly coupled to small-scale modelling (see *(a.)* and *(b.)* below), or from monitoring networks (see *(c.)* below).

NO_x $(=NO+NO_2)$, like SO_2, is emitted by both high (e.g. power plants) and low (e.g. traffic) sources, mostly as NO. The spatial variability of NO_x concentrations tends to be higher than that of ozone and nitrate but lower than that of, e.g., ammonia, due to conversion of NO by reaction with O_3. In rural areas emission of NO from soils (both agricultural and semi-natural) can likewise contribute to local NO_2 levels. Many national modelling activities are able to provide estimates of surface concentrations of NO_2 at a higher resolution than 50 x 50 km², for example at 5 x 5 km² in the Netherlands and in the UK (the UK Air Quality data base is at www.airquality.co.uk/archive/) and these can incorporate models to adjust concentrations for local emissions, for example by using distance to major roads.

(a.) LRT model results

As for SO_x, EMEP/MSC-W has available concentrations and deposition of NO_x (NO and NO_2), NO_3^- in aerosol and in rainfall, and HNO_3.

(b.) High resolution modelling

Procedures as the ones described for SO_x can be applied with the following comments. For inferential dry deposition modelling, NO_3^- aerosol and HNO_3 (ideally also $HONO$) concentration maps are needed besides the NO_x concentration maps. Since measurements are too scarce to be interpolated in most countries, they generally will have to be estimated from atmospheric models. The most important factor causing variability in NO_x is local emission which has to be accounted for in the high-resolution model. NO_2 deposition velocities depend almost exclusively on stomatal opening (stomatal pathway: to be parametrized using vegetation type/land use, and meteorology data) and the importance of surface wetness to SO_2 deposition does not hold for NO_2. For the aerosol fraction, HNO_3 deposition is determined by atmospheric resistances because the surface resistance is very low (surface roughness and windspeed are most important) and the NO_3^- aerosol deposition velocities are estimated similarly to SO_4^{2-} aerosol.

As was metioned above, ozone gradients may be affected by fast chemical reactions between O_3, NO_2 and NO. These reactions will also affect the gradients and hence the uptake of NO_2. In LRT models this effect is not taken into account. Although no firm evidence is available, it is felt that the uptake by low vegetation is only marginally affected. (Duyzer et al. 1995). On the other hand, the effect on uptake of NO_2 and O_3 by forests could be influenced by chemical reactions taking place in the canopy (Walton et al. 1997).

(c.) Monitoring and interpolating concentrations

Measurement stations for NO_x should be set up in areas that are not directly affected by local emitters (most importantly not near road traffic). Criteria for siting measurement stations are similar to those for SO_x. Due to the fact that NO is a primary pollutant and the reaction to NO_2 is relatively fast, the measurement network density has to be high, as for SO_2. The other nitrogen compounds are assumed to have slowly varying concentrations and the network density can be reduced accordingly.

2.3.7 Mapping ammonia (NH_3) concentration, reduced nitrogen (NH_x) deposition and total nitrogen deposition

Ammonia is emitted primarily from low level agricultural sources with varying source strengths. Gaseous NH_3 has a short atmospheric residence time (Erisman and Draaijers 1995) and as a result its concentrations in air may show steep horizontal and vertical gradients (Asman et al. 1988). Even in areas not affected by strong local sources, the ambient concentration of ammonia may vary by a factor of three to four on scales less then a few kilometres.

The very localised pattern of ammonia concentration, and also of ammonia dry deposition, has consequences for mapping procedures. Mapping of ammonia concentrations by interpolation from measurements alone is not treated explicitly here, as the required measurement network density would be extremely high and the method is only feasible over small areas. However, the critical level of ammonia is so high that except very near sources (farms) exceedances are not very likely.

A long-range transport model with, for example a 50 x 50 km² spatial resolution, will not resolve these large variations either for ammonia concentrations or for the dry deposition of ammonia which will be the major fraction of total reduced nitrogen deposition close to an ammonia source. So assessments of the exceedances of critical loads will be biased when using LRT models. In the absence of very detailed emission data (on the level of the individual farm), measurements in a dense network are needed to obtain accurate exceedance levels (Asman et al. 1988).

It is also important to note that ammonia may be emitted as well as deposited onto vegetation, and therefore surface–atmosphere exchange modelling must be used to quantify the net exchange over the landscape. The background developments to allow these processes to be simulated use a compensation point approach (Schjorring et al. 1998; Sutton et al. 2000).

(a.) Long-range transport modelling

As for SO_x, EMEP/MSC-W has available concentrations and deposition of NH_3 and NH_4^+ in aerosol and in rainfall. However, the interpretation of the concentration and dry deposition of ammonia estimates for ammonia must be qualified because of the spatial resolution of the model and the effects of local sources. Improvements in the ammonia component of the EMEP/MSC-W LRT model are currently being developed.

(b.) High-resolution modelling

In the LRT model results from EMEP/MSC-W, it is assumed that the concentration distribution of ammonia within a grid cell is homogeneous, whereas generally sub-grid concentration variations will be present. The spatial resolution on which concentrations can be modelled strongly depends on the resolution of the available emission estimates. Where these are available at spatial scales of the order of 1 x 1 km², considerable improvements to the estimates from the EMEP/MSC-W LRT 50 x 50 km² model are possible. Models are available for these more detailed calculations, such as the OPS model for the Netherlands (see below) and the FRAME model for the UK (Singles et al. 1998).

The Operationele Prioritaire Stoffen model (OPS) developed at RIVM is able to calculate dispersion (and deposition) of NH_x on a 5 x 5 km² grid over The Netherlands (Asman and Van Jaarsveld 1992; Erisman et al. 1998b). The model is able to describe both short- and long-distance transport, average concentrations (and depositions) can be computed for time scales from 1 day to more than 10 years, and it can account for both point sources of various heights and area sources of various shapes and heights. The basis for the model on the local scale is formed by the Gaussian plume formulation for a point source. Computations are made for a limited number of meteorological situations (classes) with a representative meteorology for each class derived from actual observations. The uncertainty in emission values appeared to be the most important factor determining the uncertainty in concentrations. Model results and air concentration measurements in the Netherlands are reasonably well correlated, but substantial differences in the absolute values have yet to be explained quantitatively (Duyzer et al. 2001). Given the concentrations on a 5 x 5 km² scale, deposition of NH_x in the Netherlands was estimated and showed good agreement with results of throughfall (corrected for canopy exchange) and micrometeorological measurements (Erisman et al. 1995).

For aerosol deposition and wet deposition, the procedures as described for SO_x can be applied to NH_4^+.

(c.) Monitoring and interpolating concentrations

Accurate representative measurement of NH_3 concentrations, especially in high emission density areas, requires many measuring sites. Typically, most of the concentration gradient is present within a few km of the source and local scale monitoring is a valuable tool to understand the processes. With the developments in passive samplers, large-scale monitoring of ammonia concentrations has become possible and there are national ammonia monitoring networks in the Netherlands and in the UK (Sutton et al. 2001a,b). The local site conditions must be noted for NH_3 monitoring so that the data can be correctly interpreted. The main use of the measurement networks for ammonia is to support the models used to predict the local scale variations in concentration, as neither models nor measurements on their own can adequately predict concentrations.

Criteria for siting measurement stations for ammonium are similar to those for SO_x. Ammonium is assumed to have slowly varying concentrations and the network density can be relatively low.

The **deposition of total nitrogen** is needed for many applications in the critical load framework. It is defined as the sum of total deposition of reduced (NH_x) nitrogen [NH_3 dry deposition, NH_4^+ aerosol deposition, NH_4^+ wet deposition, NH_4^+ cloudwater/fog deposition] and oxidised (NO_y) nitrogen [NO_2 dry deposition, HNO_3 dry deposition, NO_3^- aerosol deposition, NO_3^- wet deposition, NO_3^- cloudwater/fog deposition]. The methodological considerations concerning NH_x and NO_y depositon mapping apply accordingly.

2.3.8 Mapping base cation and chloride deposition

The deposition of physiologically active basic cations ($Bc = Ca+Mg+K$; i.e. the sum of calcium, magnesium and potassium) counteracts impacts of acid deposition and can improve the nutrient status of ecosystems with respect to eutrophication by nitrogen inputs. Sodium (Na) fluxes are needed for estimating the sea-salt fraction of sulfur, chloride, and Bc inputs, and as a tracer for canopy and soil budget models. In addition, inputs of Bc as well as Na and chloride (Cl) determine the potential acidity of deposition.

As the aim of the Convention is to minimize acid deposition irrespective of other man-made emissions, base cation inputs not linked to emissions of acidifying compounds (for example from emissions of Sahara dust, large-scale wind erosion of basic topsoil particles, etc.) should in principle not be accounted for within the critical loads framework. The non-anthropogenic, non-sea-salt atmospheric input of base cations is defined as a property of the receptor ecosystem and indirectly enters the critical load equation for acidity (see Chapter 5). However, at present there is no method to differentiate anthropogenic from non-anthropogenic deposition of base cations due to the lack of emission inventories and long-range transport models for this task.

There are so far no emission inventories available and therefore base cation and chloride deposition is not yet estimated using “classical” LRT models. Work is going on to provide base cation and chloride deposition on a European scale with 50 x 50 km² resolution. As soon as EMEP models based on (anthropogenic) emission inventories are applied to base cations, the different sources of base cations should be identified and only those relevant for control of acid deposition included in critical loads calculations.

The wet deposition of non-sea-salt chloride (Cl^*) can be estimated by correcting site fluxes of Cl for the sea-salt fraction (see Chapter 2.3.5, eq. 2.2), then interpolating the derived Cl^* concentration (see the wet deposition section of Chapter 2.3.2). Similar procedures are applied to map the concentrations of the non-sea-salt base cations Ca^*, Mg^* and K^* to produce non-sea-salt base cation wet deposition.

Base cation particle deposition can been estimated from concentrations in wet deposition and empirical scavenging ratios (Eder and Dennis 1990, Draaijers et al. 1995). Dry deposition velocities can be inferred as for SO_4^{2-} aerosol and the obtained dry deposition estimates added to measured and interpolated wet deposition estimates (e.g., Gauger et al. 2003). A similar approach has been used for the UK (RGAR 1997, CLAG 1997).

Deposition of base cations have been estimated for the Nordic countries based on monitoring data on concentrations of base cations in precipitation and air-borne particles.

2.3.9 Mapping total potential acid deposition

Total Potential Acid Deposition is defined as the sum of total deposition of strong acid anions plus ammonium minus non-sea-salt base cations.

As stated in the preceding subchapter, most chloride inputs are assumed to be of sea-salt origin, and these are removed from the equation by removing all other sea-salt inputs (i.e. of base cations incl. Na and sulfate) using a “sea-salt correction” with Na as a tracer. The implicit assumption is that sea-salt is neutral and containing no carbonates.

Surplus chloride inputs (Cl^*_{dep}) are assumed to be due to anthropogenic HCl emissions.

The sum of critical load (for sulfur) and background (non-anthropogenic) base cation deposition has formerly been defined as critical (sulfur) deposition, as used for the negotiations for the Second Sulfur Protocol (Oslo, 1994). For comparison to $CL(S+N)$, as defined in Chapter 5.3.3 (eq. 5.16), only deposition values of S and N are needed. However, if the amount of total acid input is of interest (e.g., for comparison to $CL(Ac_{pot})$, as defined in Chapter 5.3.2), non-sea-salt base cation and chloride deposition has to be included into the input side of the potential acidity exceedance equation:

(2.3)

$$Ac(pot)_{dep} = SO^*_{x\,dep} + NO_{y\,dep} + NH_{x\,dep} - BC^*_{dep} + Cl^*_{dep}$$

where:

$SO^*_{x,\,dep}$ = non-sea-salt sulfate deposition

$NO_{y\,dep}$, $NH_{x\,dep}$ = total oxidized/reduced nitrogen deposition

BC^*_{dep}, Cl^*_{dep} = non-sea-salt base cation / chloride deposition

In areas strongly affected by sea spray (high sea-salt Na, Cl, S inputs), the "total potential acid" definition of Eq. 2.3 becomes problematic, since base cations have a beneficial nutrifying effect irrespective of their chemical form (e.g., $CaCl$ vs. $CaCO_3$). At the Grange-over-Sands Workshop 1994 it was concluded that *total* $Mg+Ca+K$ deposition rates should be used for the determination of critical loads for acidity (Sverdrup et al. 1995) (see Chapter 5.3.2).

As stated in Chapter 5.3.2, Eq. 2.3 assumes that deposited NH_x is completely nitrified and exported from the system as NO_3^-, thereby acidifying the system. Thus, with respect to soil acidification it is assumed that 1 mol of SO^*_x is forming 2 moles of H^+, and 1 mol of NO_y, NH_x and Cl^* each 1 mol of H^+.

It is important to be consistent when determining total acid inputs: If results are determined on a site and process level, and if H^+ deposition rates are determined separately, NH_4^+ inputs (max. 2 eq H^+ per mol) have to be distinguished from NH_3 inputs (max. 1 eq H^+ per mol). The same applies to SO_2 (2 eq H^+ per mol) vs. SO_4^{2-} (0 eq H^+ per mol). On a larger scale, this may be neglected: Note that the emission and subsequent deposition of 1 mol and 2 mol NH_3 yields the same potential acid deposition as the deposition of 1 mol of their reaction product $(NH_4)_2SO_4$, namely 4 eq.

2.3.10 Uncertainties of quantification and mapping methods

The errors concomitant with the different methods are strongly dependent on the scale considered and the availablity of data. The following analysis is focussed on the mapping of concentrations and depositions from the EMEP/MSC-W LRT model, inferential models and interpolated measurements.

Although it is expected that the revised EMEP/MSC-W 50 x 50 km² Eulerian LRT model will become the standard model from 2003, the results from the model have not been generally available for analysis. However, preliminary indications are that the discrepancies between model output and measurements will be no larger than those from the previous EMEP/MSC-W 150 x 150 km² Lagrangian model and significant improvements are expected with some components. From a critical loads/levels perspective, the change from a 150 x 150 km² scale to a 50 x 50 km² represents a major improvement in mapping the concentrations and deposition. The other notable change in the move from the Lagrangian model to the Eulerian model has been the inclusion of vegetation specific dry deposition fluxes within the EMEP/MSC-W model. This change takes the EMEP/MSC-W model part of the way towards a full local inferential model, as the output pollutant fluxes are now vegetation specific and the issue of grid-average values being inappropriately applied to specific vegetation types, e.g. forests, should no longer occur. However, the issue of scale is important as the concentrations and the meteorological inputs are still 'average' values for the whole 50 x 50 km² grid square. Where concentrations are expected to be slowly varying, e.g. sulphate aerosol, the 'average' concentration concept should not be an important issue, but the approach is still inadequate for the rapidly varying concentration fields associated with some primary pollutants. There will still be issues to resolve with local meteorology as grid 'average' values, e.g. wind speed, will not be correct for many of the ecosystems, e.g. forests on the higher altitude areas within the grid square. Therefore, the uncertainty in deposition from the Eulerian model should be improved from the Lagrangian model but will not match the levels of uncertainty which could be achieved by a local scale inferential model. A full analysis of uncertainty in the EMEP/MSC-W Eulerian model would be a substantial task.

There are a number of references comparing the EMEP/MSC-W Lagrangian model with available EMEP measurements (e.g. Barrett et al. 1995), and with the results of other models (Iversen 1991) and the previous version of this manual (UBA 1996). When evaluating model-measurement intercomparisons, it is important to recall that (a) there are also uncertainties with the measurements and (b) the model may be estimating something rather different from what is being measured, e.g. the NO_2 concentration at a single site in a 50 x 50 km² grid square is only an estimate from a sample of size one of the 'average' NO_2 concentration in the square, which is the value the EMEP/MSC-W model is attempting to match. An evaluation of the overall uncertainty of the model requires that some further information is available on the effects of the spatial distribution of measurement sites.

Inferential deposition models treat deposition as a one-dimensional (vertical) transfer to homogeneous surfaces with infinite length, assuming a constant flux layer. This means that the flux from the 50 m reference height is assumed to be equal to the flux at the surface. The reference height must be high enough so that the concentration is not severely affected by dry deposition, but it must be below the surface layer height. Fast chemical reactions as well as the impact of enhanced turbulent exchange induced by local roughness transitions (forest edges, hills, mountains) are not taken into account. Components whose deposition strongly depends on the aerodynamic resistance (e.g. HNO_3, aerosols and cloud water/fog droplets) will show higher deposition rates than modelled. The impact of transitions on dry deposition rates of components like NO_2, whose deposition is mainly determined by stomatal conductance, will be relatively small.

However, the main uncertainties in dry deposition of sulfur and nitrogen compounds in the inferential framework are due to (1) uncertainties in surface resistance parametrisations which are not always available for all vegetation species and surface types, and (2) uncertainties in the concentration estimates, which for all the reactive gases show a scale of spatial variation which is too great to quantify from measurement activities. Surface wetness, which is one of the major factors determining dry deposition of soluble gases (NO_2, NH_3), is up to now parametrized very roughly only. The overall uncertainty in surface resistance varies between 20% and 100%, depending on component and surface type (van Pul et al. 1995).

For the deposition of nitrogen compounds, the main sources of uncertainty were described by Lövblad and Erisman (1992) to be uncertainties in emissions, concentrations and surface resistances to dry deposition, as well as surface wetness and particle deposition for NH_x especially in NW and Central Europe.

Using error propagation methods and assuming that the above mentioned uncertainties represent random errors, the total uncertainty in dry deposition of acidifying compounds for an average 10x20 km² grid is estimated at 50-100%. Systematic errors in dry deposition may arise from neglecting complex terrain effects in the parametrisation of the deposition velocity, and from other simplifications (see Chapter 2.3.2). As these calculations are scale dependent and are based on a relatively coarse spatial resolution, the uncertainty in reality may be much larger. For NH_3, for example, much larger uncertainties in even small areas have been shown (Dragosits et al. 2002).

Additional uncertainties arise for estimates of dry deposition for base cations (i) in parametrising the deposition velocity, (ii) in the precipitation concentration maps and (iii) in the scavenging ratios used to derive surface-level airborne particulate concentrations from concentrations in precipitation.

The overall uncertainty in modelled dry deposition velocities integrated over the particle size distribution representative for alkaline particles at the Speulder forest (The Netherlands) was found to equal 60%. For other sites (and for regions) additional uncertainties will arise due to limited availability and accuracy of relevant information on land use and on meteorology. The uncertainty in deposition velocity caused by variations in the size distribution of alkaline particles amounts to 30-50%, assuming a mass median diameter (MMD) of 5 µm and taking a geometric standard deviation of 2-3 to represent the variation (Ruijgrok et al. 1996). The MMD of particles depends on the distance to sources and on relative humidity.

Also scavenging ratios vary with particle diameter, and assuming the same MMD and standard deviation as above, the uncertainty in estimated ambient air concentrations caused by variation in size distribution is estimated at 50-100%. Large uncertainties in air concentrations can be expected very close or far from major sources and/or in areas with strongly deviating precipitation climatology.

Systematic errors arise from (i) using scavenging ratios based on a limited set of simultaneous ambient air and precipitation concentration measurements, (ii) neglecting complex terrain effects in the parametrisation of the deposition velocity, (iii) using annual mean air concentrations and deposition velocities for flux calculation, thereby neglecting temporal correlations. The total uncertainty in dry deposition of base cations for an average 10x20 km² grid, caused by random errors in deposition velocities and air concentrations, is estimated to be 80-120% (Draaijers et al. 1995).

Errors on wet deposition maps are due to (i) limited accuracy of measurements and (ii) the non-representativity of measurement sites. An in-depth analysis of methods to minimize and to quantify these errors can be found in the proceedings of an EMEP Workshop on these topics (Berg and Schaug 1994) and will not be repeated here. The uncertainty of the wet deposition rate for a 50x50 km² grid square, based on interpolated measurements of precipitation concentration and precipitation amount, is estimated at 50% on average. Larger uncertainties (about 70%) were found by van Leeuwen et al. (1995) in mountaineous areas and complex terrain; the same is to be expected if the measurement network is not as dense as in Northwest Europe (Schaug et al. 1993). Using a spatial scale of 5 x 5 km² over the UK, uncertainties were reported as ±35%, reflecting the improved information available by using a detailed rainfall map with an appropriate orographic model (Smith and Fowler 2001).

The uncertainty in total deposition is determined by the uncertainty in wet, dry and cloud and fog deposition. The latter is not taken into account by most models and rarely measured. As described above, the uncertainty of dry deposition is generally much larger than that of wet deposition. Total deposition estimates are more uncertain in areas with complex terrain or with strong horizontal concentration gradients. The uncertainty in total deposition (grid square average) of acidifying compounds $(N + S)$ and base cations can be estimated to be 70-120% and 90-140%, respectively for grid sizes of the order of 10 x 10 km².

Errors of sulfur deposition rates determined from throughfall measurements vs. inferential models for a forest in the Netherlands were estimated by Draaijers and Erisman (1993). Error estimates for sulfur and other substances are provided by Erisman and Draaijers (1995).

These large uncertainties illustrate the need to validate model results by measurements of airborne concentrations, wet deposition, dry deposition and throughfall, as stated in Chapter 2.3.2.

This, and the scale-specific qualities of the methods, should be kept in mind when reading the substance-specific listing of individual methods in Chapters 2.3.3-2.3.9.

2.4 Use of deposition load and concentration maps

These maps are designed to be used in combination with critical loads and critical levels maps to show where and by how much critical loads and critical levels are exceeded. The use of deposition data with critical loads data very often involves different scales of the different data sources and, in most cases, the critical loads data are provided at a finer resolution than the deposition data resulting in an underestimation of the critical load exceedance. These issues have been discussed above and improved deposition estimates, for example by using national models at a finer spatial resolution, can improve the quality of the critical load exceedances. One important point re-iterated here is that it is essential to note any different scales in the legends to figures and maps.

National maps and model outputs may be compared with data from the EMEP model, since the EMEP data are used for the Integrated Assessment Modelling activities and for the protocol developments. If the national datasets deviate strongly from EMEP model data, EMEP should be notified and scientifically based improvements of the EMEP models should be developed. National datasets may always be used for national purposes, and national model outputs should be calibrated with monitoring results at international (e.g., EMEP), national and subnational levels. This should also ensure that it is possible to compare national maps where they meet at international borders, and these activities at various levels (regional, national, international) should be coordinated by comparing results at EMEP or other workshops.

Table 2.2: Summary of methods for atmospheric concentration and deposition mapping (see Chapter 2.3), their linkage to emission inventories, and their suitability for mapping and site-level validation (1=good, 2=useful, 3=should not be used) (see text!)

Parameter	Method	Link to emission inventories	Mapping Suitability	Site-Level Validation Suitability	Comments / Requirements	Data sources	Selected References
O_3 concentration (AOT40 values)	EMEP model results	yes	1/2	3	Suitability for mapping dependent on topography and emission structure	MSC-West	Barrett et al. (1995), Malik et al. (1996)
	(interpolated) measurements	no	1/2/3	1	same as above; dependent on measurement network density	EMEP and national monitoring	CCC(1996), Barrett et al. (1995)
SO_2 concentration	EMEP model results	yes	1/2	3	Suitability for mapping mostly dependent on emission structure	MSC-West	Barrett et al. (1995)
	(interpolated) measurements	no	1/2/3	1	same as above; dependent on measurement network density	EMEP and national monitoring	CCC(1996), Barrett et al. (1995)
NO_x concentration	EMEP model results	yes	1/2	3	Suitability for mapping mostly dependent on emission structure	MSC-West	Barrett et al. (1995)
	(interpolated) measurements	no	1/2/3	1	same as above; dependent on measurement network density	EMEP and national monitoring	CCC(1996), Barrett et al. (1995)
NH_3 concentration	EMEP model results	yes	2/3	3	EMEP emission inventories not very reliable, large local variations	MSC-West	Barrett et al. (1995)
	(interpolated) measurements	no	2/3	1	Suitability for mapping mostly dependent on emission structure; high measurement network density required	EMEP and national monitoring	CCC(1996), Barrett et al. (1995)
Meteorol. parameters	(interpolated) measurements	n.a.	1/2	1/2	precipitation volume, fog and cloud occurence, net radiation, temperature, wind speed and direction	national weather services, ECMWF, EPA/NCAR	Potma (1993)
SO_x deposition	EMEP model results	yes	1/2	3	dependent mostly on quality of emission data, spatial resolution	MSC-West	Barrett et al. (1995)
	throughfall measurements	no	3	1	Should not be extrapolated	ICP Forest Level II and national monitoring	ICP Forests (1994), EDC (1993), Lövblad et al. (1993), Draaijers et al. (1996a)
	site-level inferential modelling	yes/no	3	1/2	dependent on data quality and adequate parametrization	RIVM, ITE, ...	Lövblad et al. (1993), Erisman et al. (1994)
NO_y deposition	EMEP model results	yes	1/2	3	dependent mostly on quality of emission data, spatial resolution	MSC-West	Barrett et al. (1995)
	throughfall measurements combined with canopy budget modelling	no	3	2	Should not be extrapolated	ICP Forest Level II and national monitoring	ICP Forests (1994), EDC (1993), Lövblad et al. (1993), Draaijers et al. (1996a)
	site-level inferential modelling	yes/no	3	1/2	dependent on data quality and adequate parametrization	RIVM, ITE, ...	Lövblad et al. (1993) Erisman et al. (1994)

NH_x deposition	EMEP model results	yes	2	3	EMEP emission inventories not very reliable, large local variations	MSC-West	Barrett et al. (1995)
	throughfall measurements combined with canopy budget modelling	no	3	2	Should not be extrapolated	ICP Forest Level II and national monitoring	ICP Forests (1994), EDC (1993), Lövblad et al. (1993), Draaijers et al. (1996a)
	site-level inferential modelling	yes/no	3	1/2	dependent on data quality and adequate parametrization	RIVM, ITE, ...	Lövblad et al. (1993) Erisman et al. (1994)
base cation deposition	interpolated wet deposition measurements plus model-derived dry deposition fields	no	2	3	no emission dataset available yet; concentration field (for dry deposition modelling) based on uncertain assumptions	RIVM	Draaijers et al. (1996c)
	throughfall measurements combined with canopy budget modelling	no	3	1	Should not be extrapolated	ICP Forest Level II and national monitoring	ICP Forests (1994), EDC (1993), Lövblad et al. (1993), Draaijers et al. (1996a)
	site-level inferential modelling	no	3	2	dependent on data quality and adequate parametrization	RIVM	Erisman et al. (1995)
total potential acid deposition	EMEP model results	yes	1/2	3	dependent mostly on quality of emission data, spatial resolution	MSC-West	Barrett et al. (1995)
	throughfall measurements combined with canopy budget modelling	no	3	2	Should not be extrapolated	ICP Forest Level II and national monitoring	ICP Forests (1994), EDC (1993), Lövblad et al. (1993), Draaijers et al. (1996a)
	site-level inferential modelling	yes/no	3	2	dependent on data quality and adequate parametrization	RIVM, ITE, ...	Lövblad et al. (1993) Erisman et al. (1994)

References

Anshelm F, Gauger Th (2001) Mapping of ecosystem specific long-term trends in deposition loads and concentrations of air pollutants in Germany and their comparison with Critical Loads and Critical Levels. Part 2: Mapping Critical Levels exceedances. Final Report of Project 29942210. Inst. for Navigation, Stuttgart University and Umweltbundesamt.

Asman WAH, Van Jaarsveld HA (1992) A variable-resolution transport model applied for NHx in Europe. Atmos. Environ. Part A-General Topics 26: 445-464.

Asman WAH, Drukker B, Janssen AJ (1988) Modeled historical concentrations and depositions of ammonia and ammonium in Europe. Atmos. Environ. 22: 725-735.

Aubinet M, Grelle A, Ibrom A, Rannik U, Moncrieff J, Foken T, Kowalski AS, Martin PH, Berbigier P, Bernhofer C, Clement R, Elbers J, Granier A, Grunwald T, Morgenstern K, Pilegaard K, Rebmann C, Snijders W, Valentini R, Vesala T (2000) Estimates of the annual net carbon and water exchange of forests: The EUROFLUX methodology. Adv. Ecol. Res. 30: 113-175.

Bak J (2001) Uncertainties in large scale assessments of critical loads exceedances. Water, Air and Soil Pollution Focus, Vol. 1 Nos.: 1-2, 265-280.

Barrett K, Seland Ø, Foss A, Mylona S, Sandnes H, Styve H, Tarrasòn L (1995) European transboundary acidifying air pollution. EMEP/MSC-W Report 1/95, The Norwegian Meteorological Institute, Oslo, Norway.

Beier C, Gundersen P, Rasmussen L (1992) A new method for estimation of dry deposition of particles based on throughfall measurements in a forest edge. Atmos. Environ. 26A: 1553-1559.

Berg T, Schaug J (eds.) (1994) Proceedings of the EMEP Workshop on the Accuracy of Measurements with WMO sponsored sessions on Determining the Representativeness of Measured Parameters in a Given Grid Square as Compared to Model Calculations, Passau, Germany, 22-26 Nov. 1993.

Beswick KM, Hargreaves K, Gallagher MW, Choularton TW, Fowler D (1991) Size resolved measurements of cloud droplet deposition velocity to a forest using an eddy correlation technique. Roy QJ Meteor. Soc. 117: 623-645.

Bredemeier M (1988) forest canopy transformation of atmospheric deposition. Water Air Soil Poll. 40: 121138.

CLAG (1997) Deposition fluxes in the United Kingdom: A compilation of the current deposition maps and mapping methods (1992-1994) used for Critical Loads exceedance assessment in the United Kingdom. Critical Loads Advisory Group Sub-group report on Depositon Fluxes. 45 pp. Penicuik: Institute of Terrestrial Ecology.

Coyle M, Smith RI, Stedman JR, Weston KJ, Fowler D (2002) Quantifying the spatial distribution of surface ozone concentration in the UK. Atmos. Environ. 36: 1013-1024.

Dore AJ, Choularton TW, Fowler D (1992) An improved wet deposition map of the United Kingdom incorporating the topographic dependence of rainfall concentrations. Atmos. Environ. 26A: 1375-1381.

Draaijers GPJ, Erisman JW (1993) Atmospheric sulfur deposition to forest stands - throughfall estimates compared to estimates from inference. Atmos. Environ. 27A: 43-55.

Draaijers GPJ, Erisman JW (1995) A canopy budget model to assess atmospheric deposition from throughfall measurements. Water Air Soil Pollut. 85: 2253-2258.

Draaijers GPJ, van Leeuwen EP, Potma C, van Pul WAJ, Erisman JW (1995) Mapping base cation deposition in Europe on a 10x20 km grid. Water Air Soil Pollut. 85: 2389-2394.

Draaijers GPJ, Erisman JW, Spranger T, Wyers GP (1996a) The application of throughfall measurements. Atmos. Environ. 30: 3349-3361.

Draaijers GPJ, Erisman JW, van Leeuwen NFM, Römer FG, te Winkel BH, Veltkamp AC, Vermeulen AT, Wyers GP (1996b) The impact of canopy exchange on differences observed between atmospheric deposition and throughfall fluxes. Atmos. Environ. 31: 387-397.

Draaijers GPJ, Bleeker A, van der Veen D, Erisman JW, Möls H, Fonteijn P, Geusenbrock M (2001) Field intecomparison of throughfall, stemflow and precipitation measurements performed within the framework of the Pan European Intensive Monitoring Programme of EU/ICP Forests TNO report R2001/140.

Dragosits U, Theobald MR, Place CJ, Lord E, Webb J, Hill J, ApSimon HM, Sutton MA (2002) Ammonia emission, deposition and impact assessment at the field scale: a case study of sub-grid spatial variablility. Environ. Pollut. 117: 147-158.

Duyzer JH, Deinum G, Baak J (1995) The interpretation of measurements of surface exchange of nitrogen oxides; corrections for chemical reactions. Philosophical Transactions of the Royal Society of London A 351: 1-18.

Duyzer J, Nijenhuis B, Weststrate H (2001) Monitoring and modelling of ammonia concentrations and deposition in agricultural areas of the Netherlands. Water, Air and Soil Pollution Focus, Vol 1 Nos. 5-6: 131-144.

EDC (Environmental Data Centre) (1993) Manual for Integrated Monitoring. Programme Phase 1993-1996. National Board of Waters and the Environment, Helsinki.

Eder BK, Dennis RL (1990) On the use of scavenging ratios for the inference of surface level concentration. Water Air Soil Pollut. 52: 197-215.

Emberson LD, Ashmore MR, Cambridge HM, Simpson D, Tuovinen JP (2000) Modelling stomatal ozone flux across Europe. Environ. Pollut. 109: 403-413.

EMEP/CCC (1996) EMEP Manual for Sampling and Analysis. EMEP/CCC Report 1/95.

Erisman JW, Draaijers G (1995) Atmospheric deposition in relation to acidification and eutrophication. Studies in Environmental Science 63, Elsevier, Amsterdam.

Erisman JW, van Pul A, Wyers P (1994a) Parameterization of dry deposition mechanisms for the quantification of atmospheric input to ecosystems. Atmos. Environ. 28: 2595-2607.

Erisman JW, Beier C, Draaijers GPJ, Lindberg S (1994b) Review of deposition monitoring methods. Tellus 46B: 79-93.

Erisman JW, Draaijers G, Duyzer J, Hofschreuder P, van Leeuwen N, Römer F, Ruijgrok W, Wyers P (1995) Particle deposition to forests. In: Heij GJ, Erisman JW (eds.): Proceedings of the conference “Acid rain research: do we have enough answers?”, `s-Hertogenbosch, The Netherlands, 10-12 Oct. 1994. Studies in Environmental Science 64: 115-126.

Erisman JW, Mennen MG, Fowler D, Flechard CR, Spindler G, Gr?ner A, Duyzer JH, Ruigrok W, Wyers GP (1998a) Deposition Monitoring in Europe. Environ. Monit. Assess. 53: 279-295.

Erisman JW, Bleeker A, van Jaarsveld H (1998b) Atmospheric deposition of ammonia to semi-natural vegetation in the Netherlands-methods for mapping and evaluation. Atmos. Environ. 32: 481-489.

Ferm M (2001) Validation of a diffusive sampler for ozone in workplace atmospheres according to EN838. Proceedings from International Conference Measuring Air Pollutants by Diffusive Sampling, Montpellier, France 26-28 September 2001: 298-303.

Ferm M, Svanberg PA (1998) Cost-efficient techniques for urban and background measurements of SO2 and NO2. Atmos. Environ. 32: 1377-1381.

Flechard CR, Fowler D (1998) Atmospheric ammonia at a moorland site. II: Long-term surface/atmosphere micrometeorological flux measurements. Roy QJ Meteor. Soc. 124: 733-757.

Flemming J (2003) Immissionsfelder aus Beobachtung, Modellierung und deren Kombination, PhD thesis, Free University Berlin, FB Geowissenschaften (in German).

Fowler D, Granat L, Koble R, Lovett G, Moldan F, Simmons C, Slanina SJ, Zapletal M (1993) Wet, cloud water and fog deposition. (Working Group report). In: Models and methods for the quantification of atmospheric input to ecosystems, edited by Lovblad G, Erisman JW, Fowler: 912. Copenhagen: Nordic Council of Ministers.

Fowler D, Jenkinson DS, Monteith JL, Unsworth MH (eds.) (1995a) The exchange of trace gases between land and atmosphere. Proceedings of a Royal Society discussion meeting. Philos. T. Roy. Soc. A 351 (1696), pp. 205-416.

Fowler D, Leith ID, Binnie J, Crossley A, Inglis DWF, Choularton TW, Gay M, Longhurst JWS, Conland DE (1995b) Orographic enhancement of wet deposition in the United Kingdom: continuouse monitoring. Water Air Soil Pollut. 85: 2107-2112.

Fowler D, Smith RI, Weston KJ (1995c) Quantifying the spatial distribution of surface ozone exposure at the 1kmx1km scale. In: Fuhrer, Achermann (eds.) (1995): 196-205.

Fowler D, Pitcairn CER, Erisman JW (eds.) (2001a) Air-Surface Exchange of Gases and Priticles (2000). Proceedings of the Sixth International Conference on Air-Surface Exchange of Gases and Particles, Edinburgh 3-7 July, 2000. Water Air and Soil Pollution Focus, Vol. 1 Nos. 5-6: 1-464.

Fowler D, Coyle M, Flechard C, Hargreaves K, Storeton-West R, Sutton M, Erisman JW (2001b) Advances in micrometeorological methods for the measurement and interpretation of gas and particles nitrogen fluxes. Plant Soil 228: 117-129.

Fowler D, Sutton MA, Flechard C, Cape JN, Storeton-West R, Coyle M, Smith RI (2001c) The Control of SO_2 dry deposition on to natural surfaces and its effects on regional deposition. Water, Air and Soil Pollution Focus, Vol. 1 Nos. 5-6: 39-48.

Fuhrer J (2002) Ozone impacts on vegetation. Ozone-Sci. Eng. 24: 69-74.

Gallagher M, Fontan J, Wyers P, Ruijgrok W, Duyzer J, Hummelsh P, Fowler D (1997) Atmospheric Particles and their Interactions with Natural Surfaces. In: Biosphere-Atmosphere Exchange of Pollutants and Trace Substances (ed. Slanina S.): 45-92. Springer-Verlag.

Gauger Th, Anshelm F, Schuster H, Erisman JW, Vermeulen AT, Draaijers GPJ, Bleeker A, Nagel HD (2003) Mapping of ecosystem specific long-term trends in deposition loads and concentrations of air pollutants in Germany and their comparison with Critical Loads and Critical Levels. Part 1: Deposition Loads 1990-1999. Final Report of Project 29942210. Inst. for Navigation, Stuttgart University and Umweltbundesamt.

Grunhage L, Haenel HD (1997) Platin (plant-atmosphere interaction) In: A model of plant-atmosphere interaction for estimating absorbed doses of gaseous air pollutants. Environ. Pollut. 98: 37-50.

Hicks BB, Baldocchi DD, Meyers TP, Matt DR, Hosker Jr RP (1987) A preliminary multiple resistance routine for deriving dry deposition velocities from measured quantities. Water Air Soil Pollut. 36: 311-330.

Hicks BB, McMillen R, Turner RS, Holdren Jr GR, Strickland TC (1993) A national critical loads framework for atmospheric deposition effects assessment: III. Deposition characterization. Environ. Manage. 17, No.3: 343-353.

Iversen T (editor) (1991) Comparison of three models for long term photochemical oxidants in Europe. EMEP/MSC-W Report 3/91. The Norwegian Meteorological Institute, Oslo, Norway.

Loibl W, Smidt S (1996) Ozone exposure - Areas of potential ozone risk for selected tree species. Environ. Sci. Pollut. R. 3: 213-217.

Lövblad G (1996) In: Knoflacher M, Schneider J, Soja G (eds.): Exceedance of Critical Loads and Levels - Spatial and Temporal Interpretation of Elements in Landscape Sensitive to Atmospheric Pollutants. Report from a UNECE Workshop held in Vienna, Austria, 22-24 November 1995. Federal Ministry for Environment, Youth and Family, Austria, Conference Papers 15: 236.

Lövblad G, Erisman JW (1992) Deposition of Nitrogen in Europe. Background document; In Grennfelt P, Thörnelöf E (eds.) (1992) Critical Loads for Nitrogen; report from a workshop held at Lökeberg, Sweden, 6-10 April 1992. Nord 1992: 41.

Lövblad G, Erisman JW, Fowler D (eds.) (1993) Models and Methods for the Quantification of Atmospheric Input to Ecosystems. Proceedings of an international workshop on the deposition of acidifying substances, Göteborg 4-6 November 1992. Nordiske Seminar- og Arbejdsrapporter 1993: 573, Nordic Council of Ministers.

Lövblad G, Grennfelt P, Kärenlampi L, Laurila T, Mortensen L, Ojanperä K, Pleijel H, Semb A, Simpson D, Skärby L, Tuovinen JP, Tørseth K (1996) Ozone exposure mapping in the Nordic Countries, Report from a Nordic cooperation project finanaced by the Nordic Council of Miisters, Report Tema Nord 1996: 528.

Lyman J, Fleming RH (1940) Composition of sea water. Journal of Marine Research 3: 134-146.

Nemitz E, Milford C, Sutton MA (2001) A two-layer canopy compensation point model for describing bi-directional biosphere-atmosphere exchange of ammonia. Roy QJ Meteor. Soc. 127: 815-833.

PORG (1997) Ozone in the UK. Fourth report of the Photochemical Oxidants Review Group. 234pp. DETR (ITE Edinburgh).

RGAR (1997) Acid Deposition in the United Kingdom 1992-1994. Fourth Report of the Review Group on Acid Rain. 176 pp. HMSO, London.

Ruijgrok W, Tieben H, Eisinga P (1996) The dry deposition of particles to a forest canopy: a comparison of model and experimental results. Atmos. Environ. 31: 399-415.

Schaug, J., Iversen, T. and Pedersen, U. (1993): Comparison of measurements and model results for airborne sulfur and nitrogen components with Kriging. Atmos. Environ., 27A, 831-844.

Schjoerring JK, Husted S, Mattsson M (1998) Physiological parameters controlling plant-atmosphere ammonia exchange. Atmos. Environ. 32: 491-498.

Singles R, Sutton MA, Weston KJ (1998) A multi-layer model to describe the atmospheric transport and deposition of ammonia in Great Britain. Atmos. Environ. 32: 393-399.

Sjöberg K, Lövblad G, Ferm M, Ulrich E, Cecchini S, Dalstein L (2001) Ozone measurements at forest plots using diffusive samplers. Proc. from International Conference Measuring Air Pollutants by Diffusive Sampling, Montpellier, France 26-28 September 2001. p116-123.

Slanina S ed. (1997) Biosphere-Atmosphere Exchange of Pollutants and Trace Substances. Springer-Verlag.

Slinn WGN (1982) Predictions for particle deposition to vegetativ surfaces. Atmos. Environ. 16: 1785-1794.

Smith RI, Fowler D (2001) Uncertainty in estimation of wet deposition of sulphur. Water, Air and Soil Pollution Focus, Vol 1. Nos. 5-6: 341-354.

Smith RI, Fowler D, Bull KR (1995) Quantifying the scale dependence in estimates of wet and dry deposition and the implication for critical load exceedances. In: Heij GJ, Erisman JW (eds.): Proceedings of the conference “Acid rain research: do we have enough answers?”, `s-Hertogenbosch, The Netherlands, 10-12 Oct. 1994. Studies in Environmental Science 64: 175-186.

Smith RI, Fowler D, Sutton MA, Flechard C, Coyle M (2000) Regional estimation of pollutant gas dry deposition in the UK: model description, sensitivity analyses and outputs. Atmos. Environ. 34: 3757-3777.

Spranger T (1992) Erfassung und ökosystemare Bewertung der atmosphärischen Deposition und weiterer oberirdischer Stoffflüsse im Bereich der Bornhöveder Seenkette. Ph.D. thesis, Christian-Albrechts Universität, Kiel, Germany (in German).

Spranger T, Kunze F, Gauger Th, Nagel HD, Bleeker A, Draaijers GPJ (2001) Critical Loads exceedances in Germany and their dependence on the scale of input data. Water, Air and Soil Pollution Focus, Vol. 1 Nos. 1-2: 335-351.

Stedman JR, Vincent KJ, Campbell GW, Goodwin JWL, Downing CEH (1997) New high resolution maps of estimated background ambient Nox and NO2 concentrations in the UK. Atmos. Environ. 31: 3591-3602.

Sutton MA, Lee DS, Dollard GJ, Fowler D (eds.) (1998): International conference on atmospheric ammonia: emmison, deposition and environmental impacts. Atmos. Environ. 32: 269-594.

Sutton MA, Nemitz E, Fowler D, Wyers GP, Otjes RP, Schjoerring JK, Husted S, Nielsen KH, San José, Moreno J, Gallagher MW, Gut A (2000) Fluxes of ammonia over oilseed rape Overview of the EXAMINE experiment. Agr. Forest Meteorol. 105: 327-349.

Sutton MA, Miners B, Tang YS, Milford C, Wyers GP, Duyzer JH, Fowler D (2001a) Comparison of low-cost measurement techniques for long-term monitoring of atmospheric ammonia. J. Environ. Monitor. 3: 446-453.

Sutton MA, Tang YS, Dragosits U, Fournier N, Dore T, Smith RI, Weston KJ, Fowler D (2001b) A spat ial analysis of atmospheric ammonia and ammonium in the UK. The Scientific World, 1 (S2): 275-286.

Sverdrup H, De Vries W, Hornung M, Cresser M, Langan S, Reynolds B, Skeffington R, Robertson W (1995) Modification of the simple mass-balance equation for calculation of critical loads of acidity. In: Hornung M, Sutton MA, Wilson RB (eds.) Mapping and modelling of critical loads for nitrogen: a Workshop Report, Grange-over-Sands, October 1994. Penicuik: Instsitute of Terrestrial Ecology: 87-92.

Sverdrup HU, Johnson MW, Fleming RH (1946) The Oceans - Their Physics Chemistry and General Biology. Prentice-Hall, New York, 1087 pp.

UBA (1996) Manual on Methodologies and Criteria for Mapping Critical Levels/Loads and geographical areas where they are exceeded. UBA-Texte 71/96.

Ulrich B (1983) Interaction of forest canopies with atmospheric constituents: SO_2 , alkali and earth alkali cations and chloride. In: Ulrich B, Pankrath J (eds.): Effects of accumulation af air pollutants in forest ecosystems: 33-45.

UNECE ICP Forests (1999) Manual on methods and criteria for harmonized sampling, assessment, monitoring and analysis of the effects of air pollution on forests. Part VI: Measurement of Deposition and Air Pollution.

van Leeuwen EP, Erisman JW, Draaijers GPJ, Potma CJM, van Pul WAJ (1995) European wet deposition maps based on measurements. Report No. 722108006, National Institute of Public Health and Environmental Protection, Bilthoven, The Netherlands.

van Pul WAJ, Potma C, van Leeuwen E, Draaijers G, Erisman JW (1995): EDACS: European deposition maps of acidifying components on a small scale. Model description and preliminary results. RIVM Report no. 722401005, Bilthoven, The Netherlands.

Walton S, Gallagher MW, Duyzer JH (1997) Use of a detailed model to study the exchange of Nox and O3 above and below a deciduous canopy. Atmos. Environ. 31: 2915-2932.

Who do you ask for further advice ?

For questions on .... please contact:

EMEP long-range transport models:
Erik Berge, The Norwegian Meteorological Institute, P.O. Box 43 - Blindern, N - 0313 Oslo, Norway, Tel. +47 2296 - 3000; Fax. +47 2296 - 3050

High resolution modelling of dry and cloud/fog deposition, combination with maps of interpolated wet deposition:
Jan-Willem Erisman, RIVM-LLO, P.O. Box 1, 3720 BA Bilthoven, The Netherlands, Tel. +31-30-274-2824; Fax. +31-30-2287531
David Fowler, Centre for Ecology and Hydrology, Bush Estate, Penicuik, Midlothian, EH26 0QB, United Kingdom, Tel. +44-131-445-4343; Fax. +44-131-445-3943

Combination of high-resolution models with long-range transport models:
Jan-Willem Erisman, RIVM-LLO, P.O. Box 1, 3720 BA Bilthoven, The Netherlands; Tel. +31-30-274-2824; Fax. +31-30-2287531
Erik Berge, The Norwegian Meteorological Institute, P.O. Box 43 - Blindern, N - 0313 Oslo, Norway, Tel. +47 2296 3000; Fax. +47 2296 3050

Measurement and interpolation methodology (Ambient air concentrations, wet and bulk deposition):
EMEP CCC, NILU, Postbox 100, N-2007 Kjeller, Norway, Tel. +47-6389-8000; Fax. +47-6389-8050

Evaluating total deposition maps with throughfall measurements:
Gun Lövblad, Swedish Environmental Research Institute, Box 47086, 40258 Göteborg, Sweden; Tel. +46-31-725 6240, Fax. +46-31- 725 6290, gun.lovblad@ivl.se
Jan-Willem Erisman, RIVM-LLO, P.O. Box 1, 3720 BA Bilthoven, The Netherlands; Tel +31-30-274-2824; Fax +31-30-2287531

Diffusive samplers for air pollution monitoring:
Martin Fern, Swedish Environmental Research Institute, Box 47086, 40258 Göteborg, Sweden; Tel. +46-31-725 6224, Fax. +46-31-725 6290 martin.ferm@ivls.se

General information on mapping excercises can also be obtained by contacting the Coordination Center for Effects, CCE, Netherlands: Jean-Paul Hettelingh, Tel. +31-30-74 30 48; Fax. +31-30-74 29 71

General information on modelling:
Ron Smith, Centre for Ecology and Hydrology, Bush Estate, Penicuik, Midlothian EH26 0QB, Tel. +44 131 445 4343; Fax. +44 131 445 3943

General information on NO_x, NO_3:

Kim Pilegaard, Riso National Laboratory, PO Box 49, DK-4000 Roskilde, Denmark Tel. +45 4677 4677, Fax. +45 4677 4160
Jan Duyzer, TNO-MEP, Postbus 342, 7300 AH, Apeldoorn, The Netherlands, Tel. +31 55 549 3944; Fax. +31 55 549 3252