



report

IVL Swedish Environmental Research Institute

Emissions, transport, deposition and effects of base cations in relation to acidification

Report from the UNECE LRTAP workshop in
Gothenburg November 2003



A MISTRA programme



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Summary <p>The workshop on base cation deposition took place in Gothenburg on 26-28 November 2003. It was an official workshop of the Co-operative Programme for Monitoring and Evaluation of the Long Range Transboundary Air Pollution, EMEP and Working Group on Effects under the UNECE/CLRTAP. It was organised by the Swedish Environmental Research Institute (IVL) and the Swedish Research Programme on International and National Abatement Strategies for Transboundary Air Pollution (ASTA).</p> <p>The workshop report summarises the state-of-knowledge of emissions, dispersion and deposition of base cations over Europe. The workshop evaluated in particular the knowledge with respect to mapping the base cation deposition over Europe as a basis for the application of dynamic models in the coming air pollution abatement strategies in Europe. The knowledge reflects the present situation, historical development and prospects for the coming 10-20 years.</p>	
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I Introduction

Data on base cation deposition are important for studying the effects of acidification of soils and waters, since it, together with weathering, provides ions for neutralising input of acidity. After sufficient reductions of acidifying deposition, it will be a source for replenishing lost base cation pools. The deposition of base cations from the atmosphere is large enough to significantly influence the critical loads of acidity and their exceedances. For the integrated assessment modelling to support strategies of pollution abatement over Europe there is a need for reliable data on the base cation deposition. Consequently, the uncertainties in data used will be of importance for the abatement strategies for acidification over Europe.

During 2004 the base cation deposition over Europe will be mapped in a Nordic mapping activity. For this purpose it is essential to summarise data needed and data available, as well as any other on-going activity which may be of value to co-operate and co-ordinate activities with. Mapping will be made using the EMEP model and by a data assimilation technique using monitoring data on base cations mainly in air and precipitation.

Data are needed on anthropogenic emissions and their changes since the 1800s including projections until 2020. Particles containing base cations are emitted from combustion of coal and wood fuels, industrial processes, etc. A few studies of base cation emissions have been made so far and some data are available. To make inventories, statistics are needed on energy production, industrial production, and other aspects. Significant infra-structural changes in society over the period must also be considered. Emissions are of anthropogenic and natural origin. Assumptions have to be made on the development of different sources. Also natural sources are influenced by human activities, such as land use, agricultural practises, etc. The climate change may influence the erosion and transport of soil containing base cations via changes in storm frequencies, precipitation and draughts. Sea salt is of significant importance for the input of base cations along the European West Coast and counteracts acidification in sensitive areas in particular in North-western Europe. Another important source, which is presently not well quantified, is soil dust.

The mapping of base cations depends on good quality monitoring data. This is essential when estimating base cation deposition by data assimilation as well as for the validation of model calculations based on emission data. Trends in deposition are possible to follow using the long time series available in some monitoring networks. The EMEP monitoring network as well as other networks have monitored base cations in precipitation for several decades and these data may help us estimating the integrated deposition over the last century. However, also data on base cations in air, throughfall and on surrogate surfaces are valuable for quantifying total deposition is of value.

For the Convention on Long Range Transboundary Air Pollution and the strategies to combat acidification and to allow the ecosystems to recover, it is essential to map the deposition of base cations in relation to the contributing sources. It is of importance to know whether the base cations originate from anthropogenic or natural sources, not least from the aim of making prognoses for future. The EMEP model is available for calculating the fate of particle emissions, as long as the emission data are available.

The workshop on base cation deposition took place in Gothenburg on 26-28 November 2003. It was an official workshop of the Co-operative Programme for Monitoring and Evaluation of the Long Range Transboundary Air Pollution, EMEP and Working Group on Effects under the UNECE/CLRTAP. The programme is presented in appendix 1. It was organised by the Swedish Environmental Research Institute (IVL) and the Swedish Research Programme on International and National Abatement Strategies for Transboundary Air Pollution (ASTA).

The workshop summarised the state-of-knowledge of emissions, dispersion and deposition of base cations over Europe. The workshop evaluated in particular the knowledge with respect to mapping the base cation deposition over Europe as a basis for the application of dynamic models in the coming air pollution abatement strategies in Europe. The knowledge reflects the present situation, historical development and prospects for the coming 10-20 years.

The workshop was attended by 23 experts from the following Parties to the Convention: Czech Republic, Estonia, Finland, Germany, Italy, Latvia, the Netherlands, Norway, Sweden and the United Kingdom. The International Co-operative Programme (ICP) Modelling and Mapping, the Joint Expert Group on Dynamic Modelling, EMEP/MSC-W and the secretariat to the Convention were also represented. A list of participants is available in appendix 2.

A background document on base cation deposition mapping in the Nordic countries was presented for the workshop. The report is available on <http://asta.ivl.se>.

The workshop was financed by the ASTA-programme and the Nordic Council of Ministers.

II Aims of the workshop

The scope of the workshop was to:

- Summarise knowledge on the state-of-the art of large-scale monitoring and modelling of base cation deposition in Europe,
- enhance the co-operation between effects oriented research, WGE and EMEP on the issue of base cation deposition,
- make recommendations for shorter and longer term approaches to obtain quantified data on base cation deposition,
- and explicitly address the needs of critical loads calculations and the requirements of dynamic models used in integrated assessment models.

III Conclusions from the workshop

Base cations in the effect oriented air pollution research refer normally to the most prevalent nutrient cations (except NH_4^+) in vegetation, also occurring as exchangeable cations in the soil, contributing to the base saturation, e. g. ions of calcium (Ca^{2+}), magnesium (Mg^{2+}), potassium (K^+) and in some cases sodium (Na^+). There are different approaches to the inclusion of base cations originating from sea salt in national critical load calculations. The methods should be harmonised or at least clearly described if sodium and the marine part of other base cations are included in the calculations.

In addition, chloride (Cl^-) is often used as tracer for the sea-salt correction. In general, effects work requires total (wet plus dry) base cation deposition input to the ecosystem considered. However, various fractions are needed for different purposes: For example, the base cation depositions used in steady state critical load calculations should exclude sea-salt and anthropogenic contributions, while the total base cation deposition is included in dynamic modelling.

Deposition rates of total base cations are important for the calculation of critical loads, dynamic modelling and other ecosystem effect assessments. They are usually of an order of magnitude, which is comparable to the base cation weathering and the net uptake by vegetation, although there is a strong variation between regions and ecosystem types.

The main part of the base cation deposition data is available on bulk deposition. The bulk samplers in most areas capture wet deposition well and in addition an unknown part of the dry deposition. The relation between the bulk and wet-only deposition varies strongly over Europe, as demonstrated by various presentations on national studies. It is important to consider that the bulk collector gives neither the wet-only fraction nor the total deposition.

Wet (wet-only) deposition of base cations over Europe is only fairly well known.

Dry deposition comprises a large contribution to the total base cation deposition in many parts of Europe. There are almost no measurements of dry deposition and only few data on air concentrations, which could be used in connection with inferential models (dry deposition velocities for different land covers) to estimate dry deposition amounts. Throughfall data, which are more frequently available, can be used in combination with canopy budget models to assess site-specific total deposition rates.

The dry deposition fraction is significantly affected by the land cover, and consequently is the land use classification, which should characterise the ecosystem properties, important for deposition estimates. Therefore, the mapping of dry and total deposition always requires appropriate land cover data.

The reliability of wet base cation deposition is relatively high. As for dry deposition, the uncertainties are very large.

The National Focal Centres (NFCs) under the ICP Mapping and Modelling (ICP M&M) and its Co-ordination Center for Effects (CCE) would benefit from an available European default total (ecosystem-specific) base cation deposition map. The map should be based on the joint

data and evaluation of the experts under the Working Group on Effects and the EMEP Steering Body. Such a map should preferably be at same geographical resolution as the other data in the effects oriented work. For mapping critical loads, recent long-term meteorological data (e.g. 3 - 5 years) are better than data for a single year in order to avoid potential biases due to inter-annual meteorological variations. No need is anticipated for data representing time periods shorter than one year, except maybe for specific validation purposes.

There is also a need for historical base cation deposition estimates to be used for dynamic modelling. So far there are no monitoring data or estimates available for the base cation deposition before 1950. There are methods to support any (modelled) estimates to acquire relative historical changes, such as tree ring analyses or lake sediment data.

The particulate matter emission inventory and modelling work carried out by EMEP/MSC-W was considered to provide a good framework for base cation deposition modelling. The base cation element contents can be incorporated into the chemical composition scheme for anthropogenic particulate matter emissions. However, there are no inventories made on natural (mainly sea-salt and soil erosion) emissions yet.

The preliminary results of comparing EMEP modelled deposition with throughfall measured at ICP Forests Level II sites (see summary of presentation by Westling in appendix 3) showed good agreement for ecosystem-specific total sulphur deposition and for nitrogen wet deposition. This method could be promising to apply on a base cation deposition assessment as well, provided that canopy budget models are valid to be applied on a European scale.

To validate model calculated wet deposition, the wet deposition fraction of measured bulk deposition should be estimated. As mentioned, the latter contains always some dry deposition. This could be done, for example, by using parallel wet-only and bulk deposition measurements from specific sites and apply those results region by region. Independent direct measurements of air concentrations and of dry deposition are necessary for validation of modelled dry deposition.

General decreasing trends of base cation deposition in Europe during the last 30 years were shown by several presentations. The anthropogenic fraction of base cation deposition has decreased a lot due to reduced particle emissions, even if the periods for largest decreases vary in different regions.

There is no clear evidence on the relative contributions from natural and anthropogenic emissions to present total base cation deposition. However, the natural emissions were considered to be roughly equally important compared to anthropogenic emissions, with large regional variation. There is a need for more knowledge on natural emissions (mainly wind blown dust from soil erosion).

It was considered important to have model-calculated base cation deposition maps available during next year for comparison with monitored data. However, it was acknowledged that the emission data might not be fully evaluated and available for comprehensive analysis within this timeframe.

The forthcoming activities, grouped in short, medium and long term goals, are presented in more detail in the recommendations.

IV Recommendations

The workshop recommended the providers and users of base cation deposition always to clearly define the exact type of base cation deposition they use or require.

The workshop recommended providing the critical loads mapping community with one commonly agreed European default base cation deposition map. The workshop distinguished the needs of base cation deposition data to be of short, medium and long term type, depending on the availability and possibilities to include new findings.

Short-term deposition approach - until March 2004

Short-term map on base cation deposition

A short-term deliverable of base cation deposition data is necessary already in the beginning of 2004 in order to distribute the product well before the time frame set by the ICP on Modelling and Mapping and Co-ordination and its Centre for Effects for delivering national critical load data - the end of March 2004. Taking this time constraint into account, the most straightforward approach was considered to be the use of existing data.

Wet deposition

The map should be based on commonly agreed wet-only base cation using EMEP/CCC monitoring data. The map should be derived for the year 2000 (the reference year for the forthcoming protocol revision) or for a multi-year average around it:

- Year 2000 (or possibly 3-year average),
- 50 x 50 km,
- element specific (Ca, Mg, Na, K; Cl), with and without sea-salt correction,
- interpolated with a feasible method,
- and if above not possible, then the 1996 total base cation deposition map (modelled based on measurements) could be distributed which relates to the early 1990s.

Total deposition

To derive the total deposition, results obtained with the EMEP Unified Model on estimated regionalised land-use specific ratios between dry and wet deposition should be used in combination with the wet deposition monitoring data. The data should be concurrent with wet deposition data, i.e. year 2000 with element and ecosystem specific data in 50 x 50 km grid, and PM size segregated (2.5 and 10 µm), if relevant.

The data should be made available to national focal centres (NFC) via CCE, with a clear documentation attached presenting what the data represent, how the data were produced and how they can be used. The data should be used by NFCs as default database over Europe-

wide base cation deposition data as a comparison to national maps derived e.g. from national monitoring networks, and if no other appropriate base cation deposition data are available for a country, for use in calculations replacing past NFC estimates. An assessment of the default data, compared to their national data, should be made by NFCs. The assessments will be reported to CCE or ICP-MM at the Task Force meeting and workshop 24-28 May 2004.

Other short-term recommendations

It was considered important that the countries should follow the EMEP recommendation to report to EMEP also the base cation contents from filterpack measurements.

Medium-term scale deposition estimate - during 2004

Historic deposition to the dynamic modelling

The above-mentioned approach does not fulfil the needs expressed by the Joint Expert Group on Dynamic Modelling. A possible approach towards estimating the historical depositions would be to deliver similar maps based on EMEP/CCC data from past years, e.g. in 5-year intervals from 1980. To reach estimates for earlier years methods based on historical measurements, or relative historical changes of the sulphur deposition history. However, the official status of these approaches should be agreed jointly between main actors in WGE and EMEP. Such results should be considered as an ad hoc solution and data should be used with precaution.

Historic and future depositions (1860-2100) on both acidifying and neutralising compounds are required by the dynamic modelling. If a product cannot be delivered in a short to medium term, appropriate guidelines to fulfil the needs of dynamic model inputs should be provided by EMEP as soon as possible.

EMEP map on base cation deposition

Anthropogenic emissions can be quantified and included in the current particulate matter emission inventory, possibly in a manner comparable to the “dust” component (?). It was strongly encouraged that countries would submit any data on the base cation element contents in their sectoral particulate matter emissions, including the size distribution. The base cation emission data should be compiled and used with the atmospheric dispersion models.

The resulting modelled depositions should be compared with monitoring data, such as the EMEP precipitation and air concentration data, ICP-Forest, ICP-IM and ICP-M&M data. It is anticipated that the discrepancies between modelled and measured total depositions would be higher in southern to south-eastern Europe than elsewhere, mainly due to Saharan dust influence.

The needs for size segregated measurements should be re-evaluated. At the moment, it is possible to rely on the filterpack data from the EMEP monitoring network. There are some

data on particle size ranges including their chemical speciation from Europe. Large particles are expected to play a role, however, it is currently not easy to evaluate their contribution over Europe.

EMEP/MSC-W can deliver the sea-salt deposition (calculated within the EMEP Unified Model), if these data are found useful in effect assessments. The users of these data should be identified before the deliverance via e.g. the CCE.

The research on the soil dust originated base cation deposition should have a priority and preferably made as a Europe-wide assessment. This could be possible with the EMEP Unified model, as a proposed task of EMEP/MSC-W in 2004.

Land cover data

The land cover has to be taken into account when mapping deposition, since the spatial variability of total deposition largely varies with land cover. Land cover data should be identical to or compatible with those used in effects oriented work under the Convention. This has been underlined by the ICP Modelling and Mapping which recommended to use the CORINE Land Cover (or compatible national) database for spatial allocation and the EUNIS classification for receptor definition. This workshop noted that European land cover data should be made freely available to the Parties for the work under the Convention.

Long-term scale deposition estimate

Gaps of knowledge to be filled

On a longer time-scale, the default official total base cation deposition map will be revised with new research findings.

There is a need to further compilation and evaluation of dry deposition velocity data at national and international levels.

Validation of deposition estimate

There is a need for the development of additional methods for validation. These include the use of surrogate surfaces. This could be applied largely in Europe at sites where base cation air concentrations are measured, and with models that allow the use of throughfall measurements. Potential sites include many ICP-Forest and ICP-IM sites. More research on methods is needed.

Appendix 1 Programme

Programme for

Workshop on emissions, transport, deposition and effects of base cations in relation to acidification

Gothenburg 26-28 November 2003

November 26

13.00 – 14.20 **Introductory session**

Welcome and introduction – The aim and expected outcome of the workshop

Peringe Grennfelt, IVL Swedish Environmental Research Institute and ASTA project, Gothenburg, Sweden

The need for base cation deposition data of interest for evaluating the effects situation in Europe

Matti Johansson, UNECE Geneva, Switzerland

Till Spranger, ICP Modelling & Mapping

14.00 – 14.20 COFFEE BREAK

14.20 – 15.00 **Emission data as input to modelling of base cation deposition**

Emissions of base cations – how are they possible to estimate on a European scale?

Arne Semb, Norwegian Institute for Air Research, Lillestrøm, Norway

The emission trend perspective – Swedish experience

Gun Lövblad, IVL Swedish Environmental Research Institute, Gothenburg, Sweden

15.00 – 15.30 Discussion and conclusions regarding emission data available and methods possible to use for the emission estimate, the importance of natural sources and future scenarios for base cation emissions

15.30 – 18.00 **Monitoring base cations in air and monitoring base cation deposition as input data to modelling – National experiences**

Mapping base cation deposition in Germany

Till Spranger, Umweltbundesamt, Berlin

Dry deposition of base cations at two geochemically contrasting sites in the Czech Republic

Pavel Kram, Czech Geological Survey, Prague, Czech Republic

Base cation deposition in Finland

Tuija Ruoho-Airola, Finnish Meteorological Institute, Helsinki, Finland

Base cation deposition in UK

Ron Smith, Centre for Environment and Hydrology, Edinburgh, Scotland

Local and large scale deposition of base cations in Estonia

Katrin Pajuste, Estonian Environmental Research Centre, Tallinn, Estonia

Trend of base cation deposition at the Czech EMEP and ICP-IM sites

Milan Vana, Czech Hydrometeorological Institute, Kosetice, Czech Republic

Surrogate surface methods to estimate base cation deposition data

Martin Ferm, IVL Swedish Environmental Research Institute, Gothenburg, Sweden

Input and output of base cations in a forested catchment

Hans Hultberg, IVL Swedish Environmental Research Institute, Gothenburg, Sweden

Base cations in air and deposition in the Netherlands

Hans van Jaarsveld, RIVM, Bilthoven, The Netherlands

Base cations measured in the European Air and Chemistry Network 1950-1980

Lennart Granat, Stockholm University, Dep. of Meteorology, Stockholm, Sweden

November 27

09.00 – 09.30 **Monitoring base cations in air and deposition as input data to modelling - the European perspective**

Data available over Europe for base cation deposition mapping – the ICP Forests data

Olle Westling, IVL Swedish Environmental Research Institute, Gothenburg, Sweden

Discussion and conclusions regarding base cations in air, precipitation, throughfall and other monitoring data available on a European scale and methods possible to use for estimating deposition, how to deal with local deposition, etc.

10.00-10.30 COFFEE BREAK

10.30 – 12.00 **Modelling base cation deposition from emission data and from air quality and wet deposition data**

Using the EMEP model for base cation deposition

Leonor Tarrason, Meteorological Institute, Oslo, Norway

Case study - The Nordic mapping of base cation deposition using data assimilation

Christer Persson, Swedish Meteorological and Hydrological Institute, Norrköping, Sweden

12.00-13.00 LUNCH

- 13.00-13.45 **Modelling base cation deposition from emission data and from air quality and wet deposition data – continued**
- Discussion and conclusions regarding base cations in air, precipitation, throughfall and other monitoring data available on a European scale and methods possible to use for estimating deposition, how to deal with local deposition, sea salt deposition, model validation, etc.
- 13.45-15.00 **The inclusion of base cation deposition data into the mapping of critical loads**
- Need for data in the critical load calculations and in calculating ecosystem recovery
- Filip Moldan, IVL Swedish Environmental Research Institute*
- 15.00-15.30 COFFEE BREAK
- Discussion and conclusions regarding the need for data on base cation deposition, need for data accuracy, need for time periods to be used in dynamic modelling, the effect of accurate data in relation to available CL data, and the influence on the need for pollution abatement in protocols. Are there other important requirements e.g. on temporal and spatial scale and requirements due to model type used? Are there different needs for studying forest soils and surface waters and for steady-state and dynamic modelling.
- 15.30-17.00 Working groups to summarise recommendations on the data available, methods, models for the mapping of base cation deposition over Europe as input data to the Convention on Long-Range Transboundary Air Pollution
- 18.00 Transport to Eastindia ship and dinner
- November 28
- 9.00-12.00 Conclusions of the working group recommendations
- 10.00-10.30 COFFEE BREAK
- 12.00 Closing the workshop
- 12.00- LUNCH

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Appendix 3 Presentations

Introductory session: The need for base cation deposition data of interest for evaluating the effect situation in Europe

Progress under the Convention on Long-range Transboundary Air Pollution (CLRTAP)

Matti Johansson, Secretary to the Working Group on Effects

Matti Johansson informed the participants of relevant ongoing activities under the Convention and work of the ICPs and task forces under the Working Group on Effects. He also deliberated on the historical development of base cation deposition and potential general ways to approach the problem in a systematic manner with different spatial and temporal scales, observational and modelling methods.

The main priorities under the Convention

Implementation of and compliance with protocols in force:

- 1985 and 1994 Sulphur Protocols
- 1988 NO_x Protocol
- 1991 VOC Protocol
- 1998 POP Protocol (entry in force 23 Oct 2003)

Review and extension of protocols:

- 1998 POPs Protocol (17 ratifications)
- 1998 Heavy Metals Protocol (17 ratifications)
- 1999 Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone (6 ratifications)

Current priorities under WGE

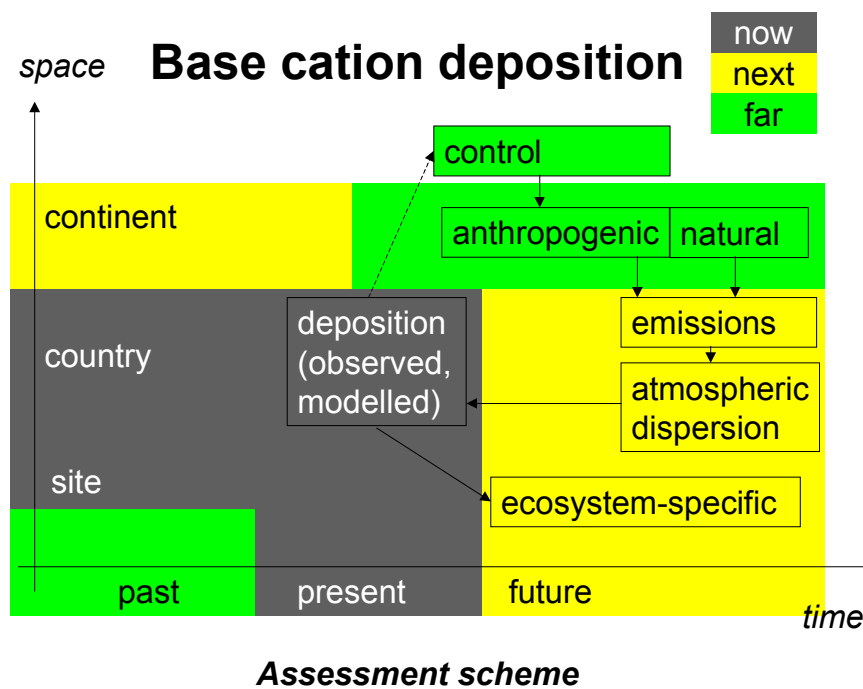
- a communication strategy including a substantive report of WGE and CLRTAP 25th anniversary
- increased focus on HM and POP
- collaboration between WGE and EMEP, including joint medium-term work plan, including BC deposition
- facilitating participation of all countries
- financing of core activities

Timing and content of Gothenburg Protocol review

The review could:

- commence next year (dependant on entry into force in 2004)
- cover PM and its health impacts
- make use of dynamic modelling results
- take account of hemispheric transport of air pollutants
- use 2015 / 2020 as possible target years, different for dynamic modelling (2010/2015/2030/2050/2100)
- be concurrent with EU/CAFE/NECD revision

The need for data is summarised in the figure below.



*Introductory session: The need for
base cation deposition data of interest
for evaluating the effect situation in
Europe*

The need and priorities for data

Till Spranger, Chairman of ICP Modelling and Mapping

The needs for BC deposition data

Bc deposition is part of the CL(Acidity) calculation. Bc deposition also affects N and C budgets (CL(nutrients) and C sequestration). Deposition rates are similar to Bc weathering and Bc net uptake. Land-use (ecosystem) specific rates are needed for CL calculations and Bc deposition histories (1860 \Rightarrow) and scenarios (\Rightarrow 2080) are needed for dynamic modelling. Present basis of base cation deposition for European CL map are different observational and modelling methods of the ICP M&M National Focal Centres (NFCs). A default map on base cation deposition would help harmonising the European critical load dataset in some regions. To this end, quantitative input from EMEP had been agreed in the Joint Medium-term Workplan of WGE and EMEP. Such a default map should be available to ICP M&M NFCs in time early 2004 in response to the call for data on critical loads and dynamic modelling (deadline end of March 2004).

Division of Work: Present

BC deposition maps have been provided by ICP Modelling & Mapping National Focal Centres to the CCE. EMEP has not been (directly) involved. The resulting data quality is mixed and merits European harmonisation. The 2003 CCE call for BC deposition data asked for:

- receptor specificity
- quantification methods
- interpolation/allocation

Additional site-specific trend data are available from:

- EMEP monitoring (bulk, wet)
- ICP Forests (TF, bulk, wet)
- ICP Integrated Monitoring (TF, bulk, wet)
- National and sub-national monitoring networks

The latest CCE Call for CL and dynamic modelling (TL) has the deadline 31 March 2004. It may be the final one for review / revision of Gothenburg Protocol and NEC Directive.

Division of work: Future

EMEP could provide European base cation deposition maps by 2003, according to “Joint Medium-Term Workplans of EMEP and WGE”. The JEG (on Dynamic Modelling, 2003) strongly urges EMEP to develop estimates of base cation deposition by grid square over time (1860-2000) In the absence of such estimates, for the first call these data will be estimated on a country by country basis.

Important discussion items at the BC workshop

- State-of-the-art in BC_{dep} models and maps
- Availability and use of observational data
- Inclusion of dry deposition
- Ecosystem / land cover specific allocation
- Feasible and necessary resolution
- Uncertainties
- One practical, usable and commonly agreed European map by EMEP?
- Role of country-specific maps (NFCs)
- Deposition history and scenarios (or surrogates) for dynamic modelling – by EMEP?
- Time constraints: GP and NEC Dir revision.

*Emission data as input to modelling of
base cation deposition*

**Emissions of base cations - how are they possible to estimate
on a European scale?**

Arne Semb from the Norwegian Institute for Air Research (NILU), Norway

Arne Semb presented results from comparison of base cation content in airborne particulate matter and emission-related activities and related crustal material contents and also other data which could be used for emission inventories.

He highlighted the importance of both wind blown dust and the anthropogenic source sectors, the latter of which have decreased relatively much in recent times. Emission surveys should be encouraged and made at national levels to ensure country specific details in certain sectors. Some work at European level is underway to include chemical composition in particulate matter emission database.

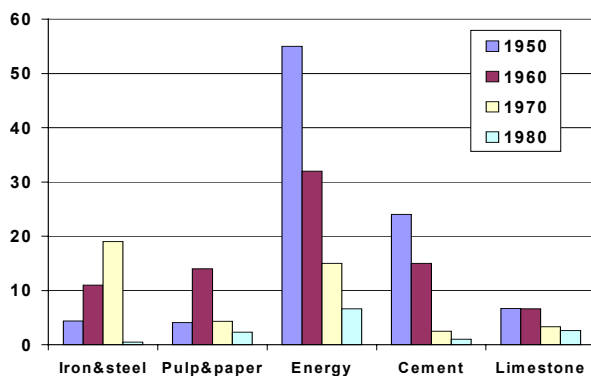
Emission data as input to modelling of base cation deposition

The emission trend perspective – Swedish experience

Gun Lövblad from the Swedish Environmental Research Institute (IVL), Sweden

Gun Lövblad presented emission studies in Sweden during 1980s and 1990s. Base cation emissions and deposition has been of interest in Sweden since 1980. Important issues and studies are summarised in five bullets referring to figures and tables below:

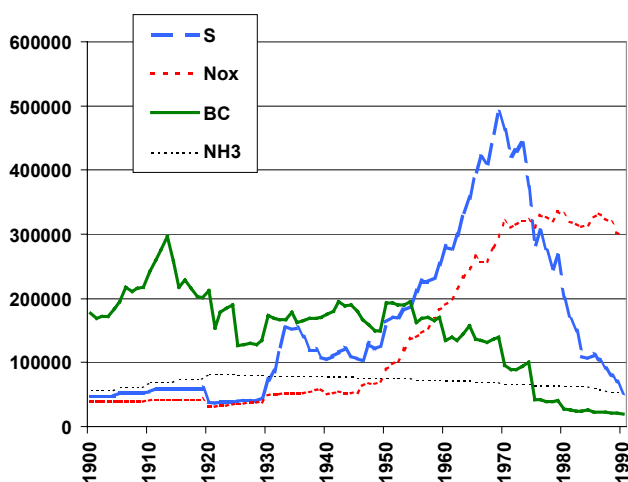
1. Alkali and BC emissions in Sweden, studies conducted in 1984, 1987
2. Accumulated load of acidity, conducted in 1993
3. Deposition of base cations in Sweden and emissions of base cations, conducted in 2000
4. Mapping deposition of base cations in the Nordic countries, conducted 2003, and
5. Finally planned for 2004, Mapping BC deposition in Europe



1. A decrease (a factor 5-10) in base cation emissions

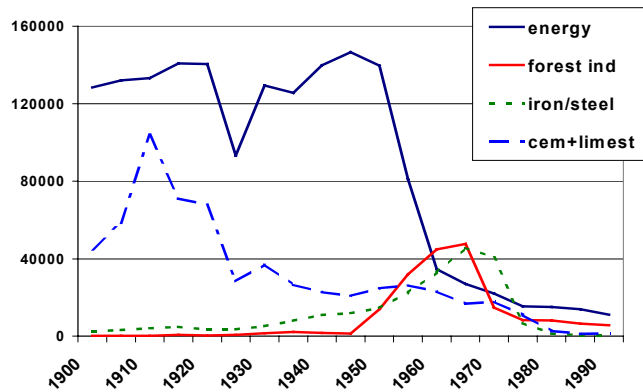
At the same time an increase in SO₂ (1000 ton SO₂-eq.).

The same situation is seen in western Europe and USA.



2. Accumulated load of acidity in Sweden

Estimated anthropogenic emissions (tons) of sulphur dioxide, nitrogen oxides, ammonia and base cations (mill. eq.) in Sweden from 1900 to 1990.



Emissions (mill. eq.) of base cations from different anthropogenic sources in Sweden from 1900 to 1990.

3. Emissions of base cations from anthropogenic sources in Sweden

Ton/year	Ca	Mg	K	Na
Cement ind	230		5	5
Pulp & paper	480		30	820
Iron & steel	80		30	20
wood/biofuel	2200	210	540	65
Oil	180		45	30
Coal	120		25	25

Wind erosion of base cations

Ton/year	Ca	Mg	K	Na
Wind erosion	20-200	1.5-15	1.5-15	1.5-15

4. The Nordic mapping (Lövblad et al. 2003) made some recommendations for procedure on a European scale:

1. *Compilation of monitoring data*
2. *Compilation of emission data*
3. *Implementation of base cations in the EMEP Unified Eulerian model*
4. *Evaluation of uncertainties in the European mapping of base cation deposition*

5. Mapping on European scale will be initiated in 2004 via a Nordic project (NMR).

Support and co-operation with European groups would be favourable.

Reference:

The deposition of base cations in the Nordic countries. Report from a NMR project 2003 and background document at the workshop.

Authors: Gun Lövblad, Christer Persson, Thomas Klein, Tuija Ruoho-Airola, Mads Hovmand Leonor Tarrason, Kjetil Tørseth Lars Rapp, Thorjörn Larssen, Filip Moldan

Monitoring base cations in air and monitoring base cation deposition as input data to modelling – National experiences

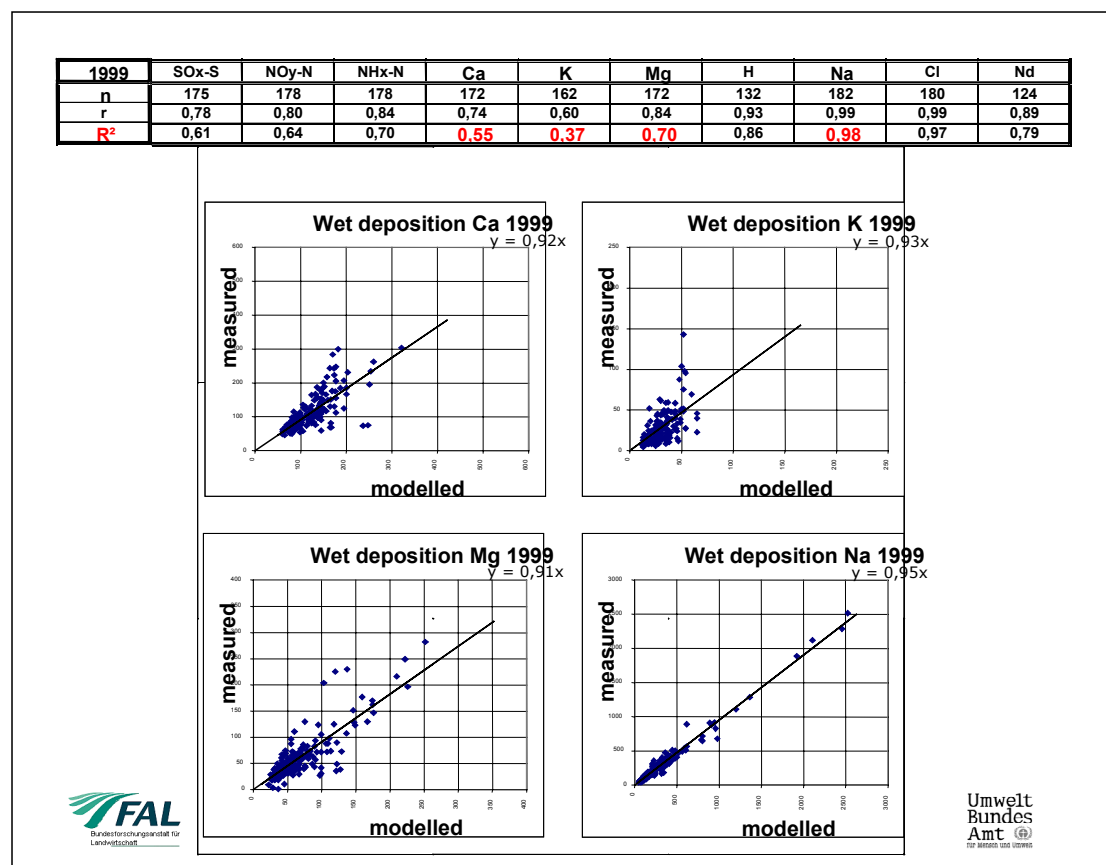
Mapping base cation deposition in Germany

Till Spranger from Umweltbundesamt, Berlin, Germany

Till Spranger presented base cation deposition mapping in Germany in 1990 - 1999. On a high resolution (1 km x 1 km) grid wet and dry depositions were handled differently. The land cover was found to have the main effect on dry deposition velocities and rates. Calcium dominated base cation deposition (e.g. 66% of total non-seasalt base cation deposition).

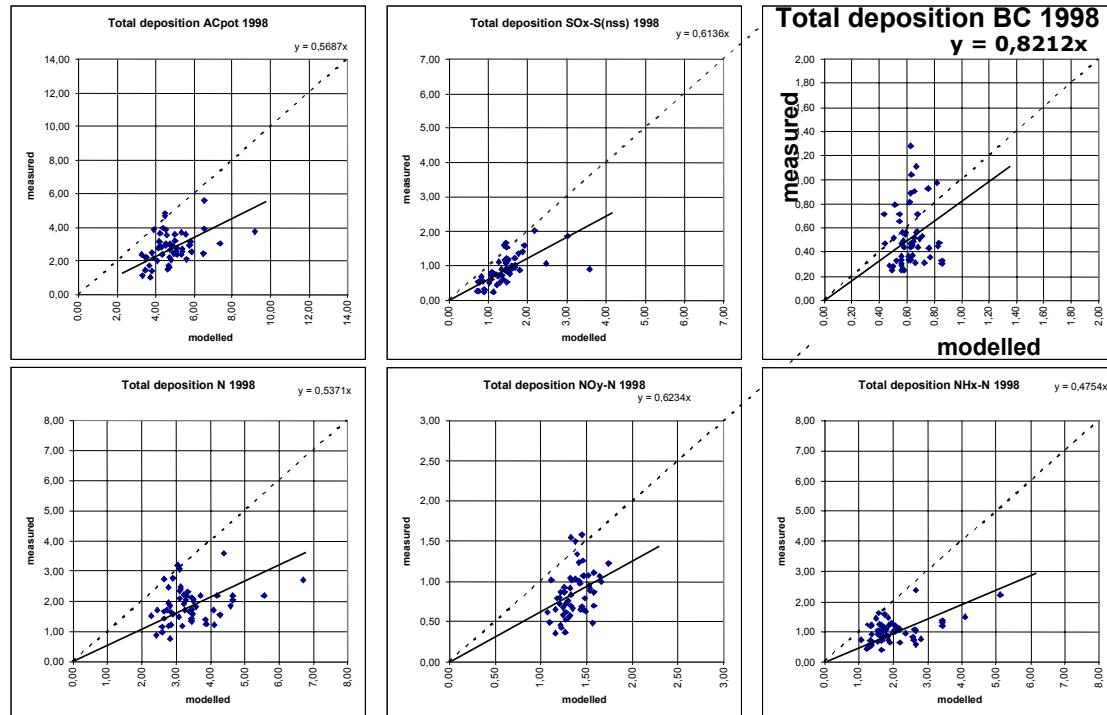
Dry deposition comprised 60% of total deposition at national level, and was even more important for forests (ca. 75%). The wet deposition compared well between measured and modelled values, however, it was noted that mapped total base cation deposition is difficult to validate. The total deposition has decreased 40% in 1990-1999.

Wet deposition loads: map values vs. measured values



Total deposition loads: map values vs. monitoring data

Validation of total deposition mapping results via data from canopy budget modelled (CBM) site measurements (ICP Forests Level II). Comparison of x = mapping result with y = (CBM).



Conclusions

- High resolution, ecosystem specific mapping of total base cation deposition is possible with reasonable accuracy
- Wet deposition should be based on monitoring, dry deposition on (monitoring, scavenging ratios and) inferential modelling
- Dry deposition is ~60% of total deposition even on national average (ca. 75% for forests)
- Total deposition rates in Germany have decreased by ~40% between 1990 and 1999

*Monitoring base cations in air and
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Dry and wet deposition and internal cycling of base cations at two geochemically contrasting sites in the Czech Republic

Pavel Kram and Jakub Hruska from the Czech Geological Survey, Czech Republic

Pavel Kram presented evaluation of atmospheric deposition at two study catchments in western Bohemia. Lysina catchment has Mg-poor soil underlain by leucogranite, however Pluhuv Bor catchment has Mg-rich soil underlain by serpentinite (Hruska and Kram 2003, Hydrol. Earth Syst. Sci. 7: 525). Both catchments are covered almost exclusively by Norway spruce plantations. Atmospheric deposition was monitored in a monthly time step using bulk deposition and throughfall collectors. Calculations were based on assumption that Na is not involved in internal cycling of trees and has the same dry deposition factor as the other three base cations which are important nutrients. Another used assumption was that all chloride is of marine origin and the proportion of base cations of marine origin in the deposition depends on the proportion in oceanic water.

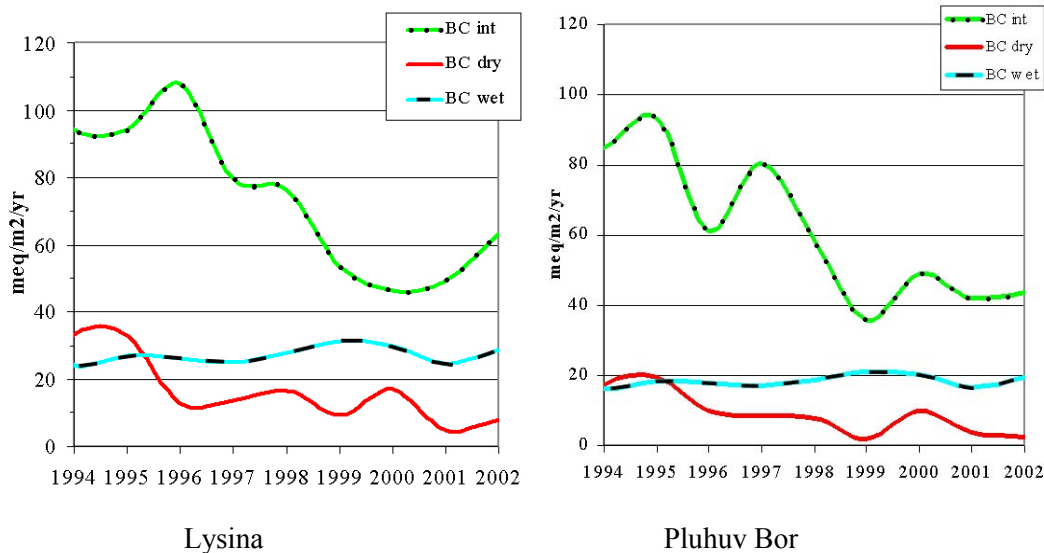
Long-term mean concentration of base cations in deposition was 43 ueq/l. Ca formed 46%, Na 31%, Mg 14% and K 9% of the total deposition. Large influence of marine salts on Na (100%) and Mg (69%) was apparent at the study sites despite the fact that they are situated in the middle of continent. On the other hand, the influence of marine salts on the other base cations (K 10%, Ca 4%) and sulphate (2%) was negligible. Almost identical proportions of Ca, K and Na individual fluxes in throughfall at the both sites were striking (see table below). However, very different patterns of Mg fluxes in throughfall were found. Much larger internal flux of Mg at Pluhuv Bor was caused by extraordinary availability of Mg in the corresponding eutrophic brown soil developed on Mg-silicate bedrock.

Long-term mean proportions of annual base cation fluxes in throughfall at the two studied catchments covered by Norway spruce in the Czech Republic in 1992-2002.

Element	Lysina			Pluhuv Bor			
	Proportion (%)	Internal cycling	Dry deposition	Wet deposition	Internal cycling	Dry deposition	Wet deposition
Ca		56	15	29	57	13	30
K		92	3	5	88	4	8
Na		0	35	65	0	31	69
Mg		54	16	30	88	4	8

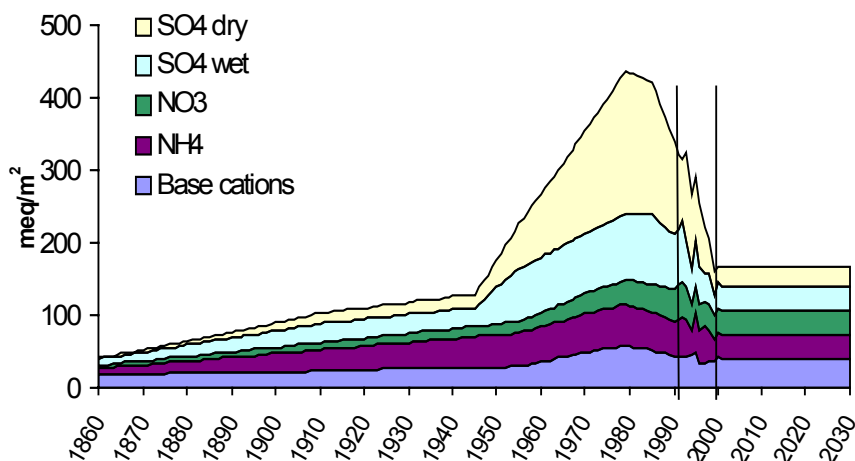
A decreasing trend in acidity of throughfall was evident at both sites. Values of pH ranged usually between 3.3 and 4.8 in the beginning of 1990s. However they ranged typically between 3.8 and 6.4 in the second half of 1990s and in the following years. This change

influenced leaching of base cations from the foliage of spruce trees in study sites. Internal cycling of base cations markedly decreased in the second half of the 1990s (see figure below). Lower dry deposition of base cations in the same period was probably caused by lower regional emissions of particulate matter. Dry deposition of base cations was higher than the wet deposition only before 1996. Interestingly, the wet deposition of base cations remained at the same level during the study period.



Annual internal cycling, dry and wet deposition fluxes of base cations in throughfall at the two studied catchments covered by Norway spruce in the Czech Republic.

Long-term input of annual atmospheric deposition including base cations was used as the major driving variable for dynamic modelling of streamwater chemistry at the examined catchments (see figure below).



Atmospheric Deposition Sequences used in the application of the MAGIC model at Lysina (Hruška et al. 2002, *Env. Poll.* 120: 261). For the period 1860-1990 estimated hindcast deposition was used, for 2001-2000 (depicted by vertical lines) measured annual data were applied, and for 2001-2030 the average chemical fluxes measured in 1999-2000 were used.

*Monitoring base cations in air and
monitoring base cation deposition as input
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Base cation deposition in Finland

Tuija Ruoho-Airola from the Finnish Meteorological Institute (FMI), Finland

Tuija Ruoho-Airola presented Finnish base cation deposition estimates based on the monitoring networks of the FMI. Many significant downward trends were observed from 1985/1988 to 2000. The results were in line with Nordic and other Finnish monitoring. The observed decreasing sulphur deposition trends occurred in larger regions than those of base cation deposition. In SE the wet deposition of base cations decreased almost as much as the wet deposition of sulphur.

Trends for base cation concentration in bulk precipitation.

Comp.	station	area	first year	trend sign.	slope/ year	nss trend
Ca ²⁺	Utö	SW	1988	**	-0.023	*
	Virolahti	SE	1988	***	-0.033	***
	Punkaharju	SE	1985	***	-0.018	***
	Kotinen	S	1988	***	-0.009	***
	Ähtäri	C	1988	*	-0.005	**
	Sodankylä	N	1985	*	-0.003	*
K ⁺	Kevo	N	1985	***	-0.005	
	Utö	SW	1988	***	-0.039	**
	Guttorp	SW	1985	*	-0.005	*
	Virolahti	SE	1988	**	-0.011	**
Mg ²⁺	Kevo	N	1985	**	-0.004	
	Virolahti	SE	1988	*	-0.002	**
	Punkaharju	SE	1985	**	-0.002	
	Kotinen	S	1988	**	-0.001	*
	Ähtäri	C	1988	*	-0.001	

unit: mg/l*year; last year 2000

Trends for base cation concentration in particles for 1989-1999

Comp.	station	area	trend sign.	slope/ year	nss trend
Ca ²⁺	Punkaharju	SE	**	0.0019	*
	Kevo	N	**	0.0006	*
K ⁺	Kevo	N	*	0.0004	
Na ⁺	Punkaharju	SE	*	0.0092	
Mg ²⁺	Punkaharju	SE	**	0.0005	
	Kevo	N			*

unit ng/m³*year

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Base cation deposition in UK

Ron Smith from Centre for Environment and Hydrology, United Kingdom

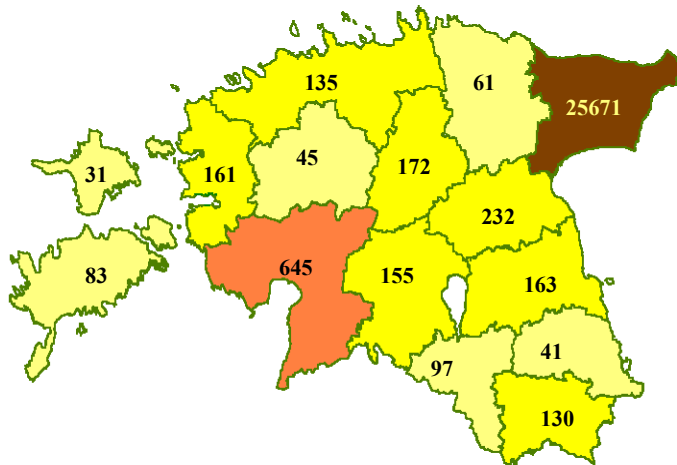
Ron Smith presented various national activities relating to base cation deposition. Several monitoring activities produce high-resolution maps (for Ca, Mg and Na). Wet deposition was found to be largest on a national average, cloud deposition smallest, but dry deposition was close to half of total deposition.

*Monitoring base cations in air and
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Local and large scale deposition of base cations in Estonia

Katrin Pajuste from Estonian Environmental Research Centre, Estonia

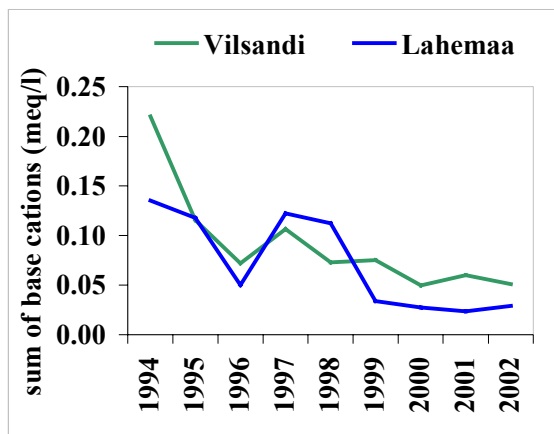
Katrin Pajuste presented local and large scale deposition of base cations in Estonia. The total suspended particle emissions are dominated by few sources, especially in north-eastern part of the country. The emissions have recently been reduced due to installed control technologies. A decreasing trend in base cation deposition was detected in 1994 to 2001, and the levels in the north-eastern part of the country remain still clearly higher than elsewhere.



Emissions of particulates (t) from stationary sources in energy sector in 2002. Total emission of particulates was 27823 t.

Region	Ca	Mg	Na	K	Total deposition	Non-sea-salt deposition
	mg/m ²					
N	710	137	289	169	48	45
NE	1806	247	529	373	113	106
E	688	140	255	238	51	48
S	595	117	243	169	45	42
W	312	44	380	182	27	20

Weighted mean deposition of Ca, Mg, Na, K (mg/m²) and deposition of summed cations (meq/m²) in some regions in 2000-2002.



Trends of base cations in background areas 1994-2002.

Conclusions

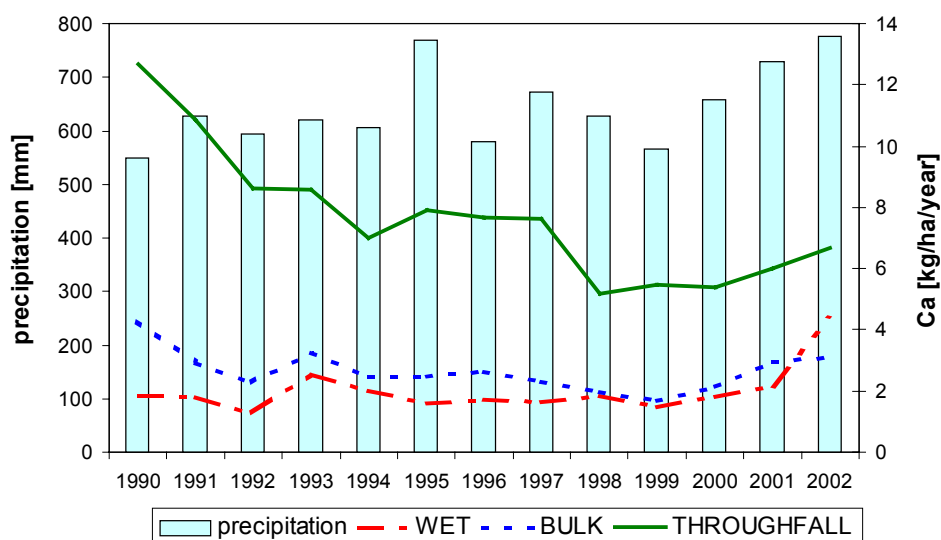
- Emissions have decreased and further decreasing is expected
- Base cation load are still relatively high in NE Estonia
- Sum of the base cation concentrations has decreased
- Future: normalisation or acidification of conditions in affected ecosystems?

*Monitoring base cations in air and
monitoring base cation deposition as input
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Trend of base cation deposition at the Czech EMEP and ICP-IM sites

Milan Vana from Czech Hydrometeorological Institute, the Czech Republic

Milan Vana presented base cation deposition in Czech EMEP and IM stations. Emissions have decreased significantly during 1980 to 2001, especially for solid particles. A decreasing trend of –50% for calcium was observed in 1990 to 2002.



Calcium deposition 1990-2002.

Conclusions

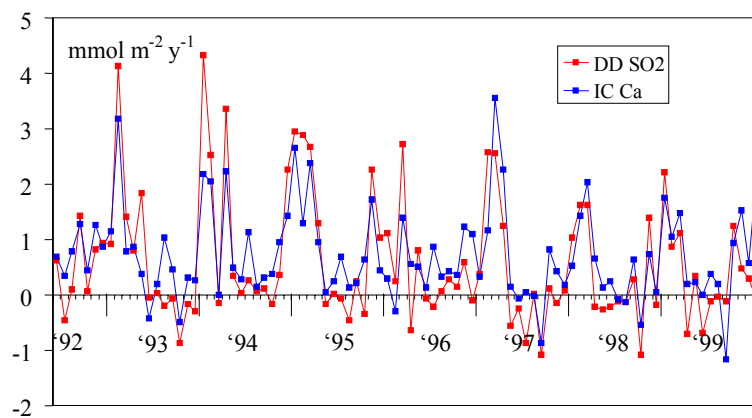
- Lower sulphur emissions in the Czech Republic have reduced the deposition significantly at the regional level. The largest difference can be observed in respect of throughfall, which implies a marked decrease in dry deposition.
- The NO_x emissions dropped in the period under review, but by far not so significantly as in the case of sulphur.
- Calcium deposition shows decreasing trend in throughfall from 14 kg/ha/year in the beginning of nineties to 7 kg/ha/year in the end of the period under review. No trend in bulk and wet deposition.
- A slightly downward trend has been identified for some other pollutants (Ca, Na, Mg).
- The value of pH increased continuously, the most significant trend is recorded in throughfall.

*Monitoring base cations in air and
monitoring base cation deposition as input
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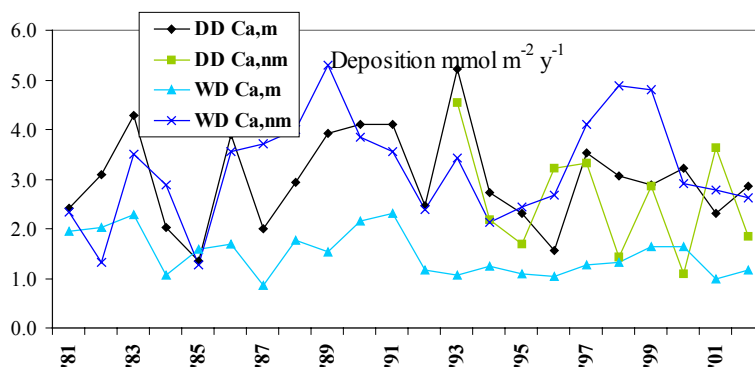
Surrogate surface methods to estimate base cation deposition data

Martin Ferm from the Swedish Environmental Research Institute (IVL), Sweden

Martin Ferm presented surrogate surface methods to estimate wet and dry base cation deposition data, including internal cycling, from open field and throughfall monitoring. The calculations show that the internal cycling of calcium in the tree canopy is correlated to dry deposition of SO_2 . Calculations with the surrogate surface method show no significant trends (tendency in wet deposition) since 1981 in wet and dry deposition of Ca in a spruce forest in SW Sweden



Internal cycling of Ca and dry deposition of SO_2 in a Norway spruce forest at Gårdsjön, SW Sweden during 1992 to 2000.



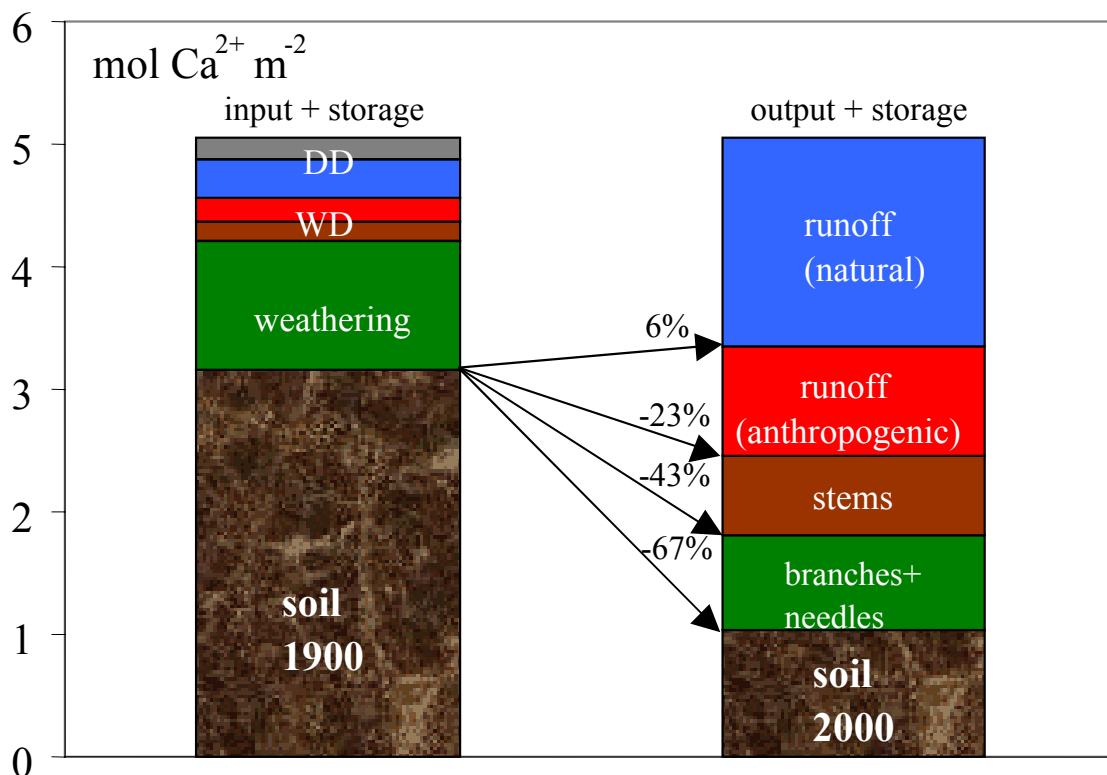
Calculated marine (m) and none marine (nm) dry (DD) and wet (WD) deposition in a Norway spruce forest at Gårdsjön, SW Sweden during 1981 to 2002.

*Monitoring base cations in air and
monitoring base cation deposition as input
data to modelling – National experiences*

Input and output of base cations in a forested catchment

Hans Hultberg from the Swedish Environmental Research Institute (IVL), Sweden

Hans Hultberg presented the input and output of base cations in a forested catchment (Gårdsjön F1) in SW Sweden. The sulphur input is still above the critical load for sulphur, despite the large reduction in deposition. Studies of the present soil chemistry (year 2000) and estimates of output of Ca from soil via runoff and tree uptake during the last 100 years indicates a large decrease of exchangeable Ca in the soil compared to year 1900. The net loss of Ca from soil is caused by a combination of increased (anthropogenic) leaching and net uptake in trees regenerated about 100 years ago. The future Ca budget is dependent of the management methods. With whole tree harvest the Ca in stems, branches and needles will be permanently lost from the soil. The studied site is an example of an acid sensitive forest common in Sweden, due to thin and natural acid soils and slow weathering rate.



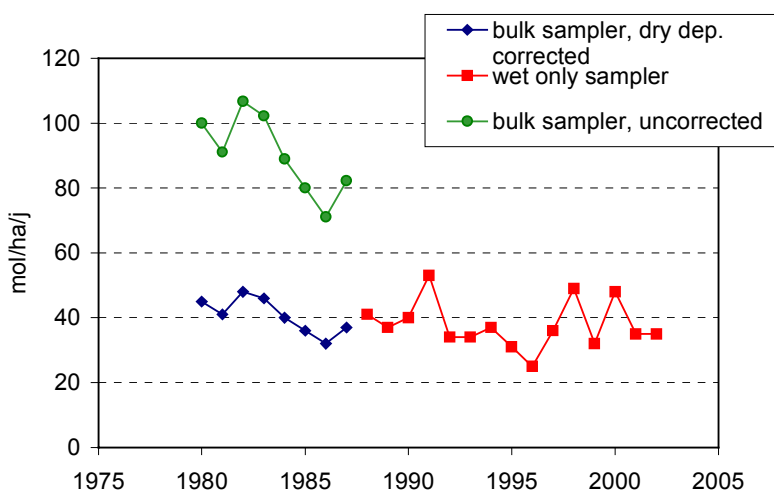
A calculation in a forested catchment in SW Sweden showing the balance between input (100 years of deposition and weathering) and the calculated storage in soil of Ca year 1900 (left column) and the 100 years of output and the measured storage in soil year 2000 (right column).

*Monitoring base cations in air and
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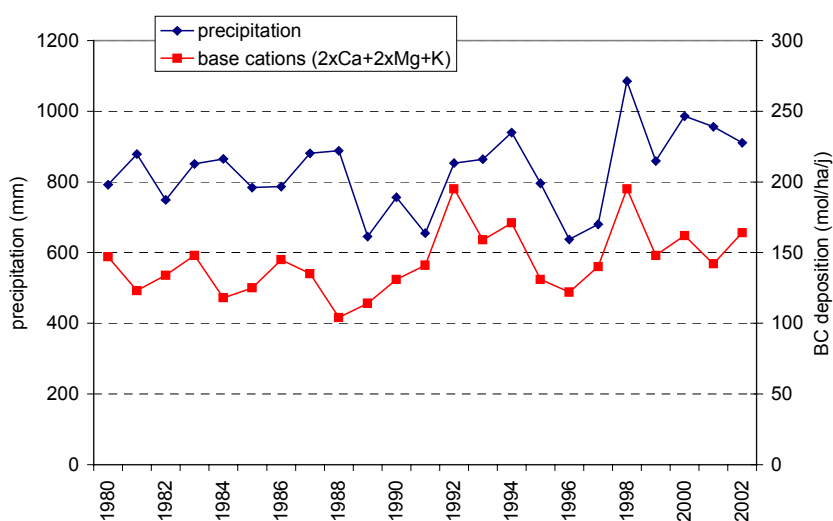
Base cations in air and deposition in the Netherlands

Hans van Jaarsveld from Netherlands Environmental Assessment at RIVM

Hans van Jaarsveld presented Dutch monitoring activities on both wet deposition and aerosols. For calcium, the bulk deposition was almost twice the wet-only deposition, and much less for other components. A decreasing trend was noted before the 1990s. Remaining temporal variations were presumed to be strongly related to natural sources and/or agricultural activities.



Example of trend in wet Ca²⁺ deposition; station De Bilt (seasalt corrected).



Trend in wet base cation deposition, mean of 14 stations in the Netherlands; sea salt corrected.

Conclusions

- Base cation deposition in the Netherlands mainly related to soil and/or agricultural activities
- High base cation wet depositions are related to local agricultural activities
- Bulk collectors give much higher values than wet- only collectors, especially for Ca^{2+}
- No clear trends in CA^{2+} since 1988; Wet deposition does not change while aerosol concentrations show a small decrease

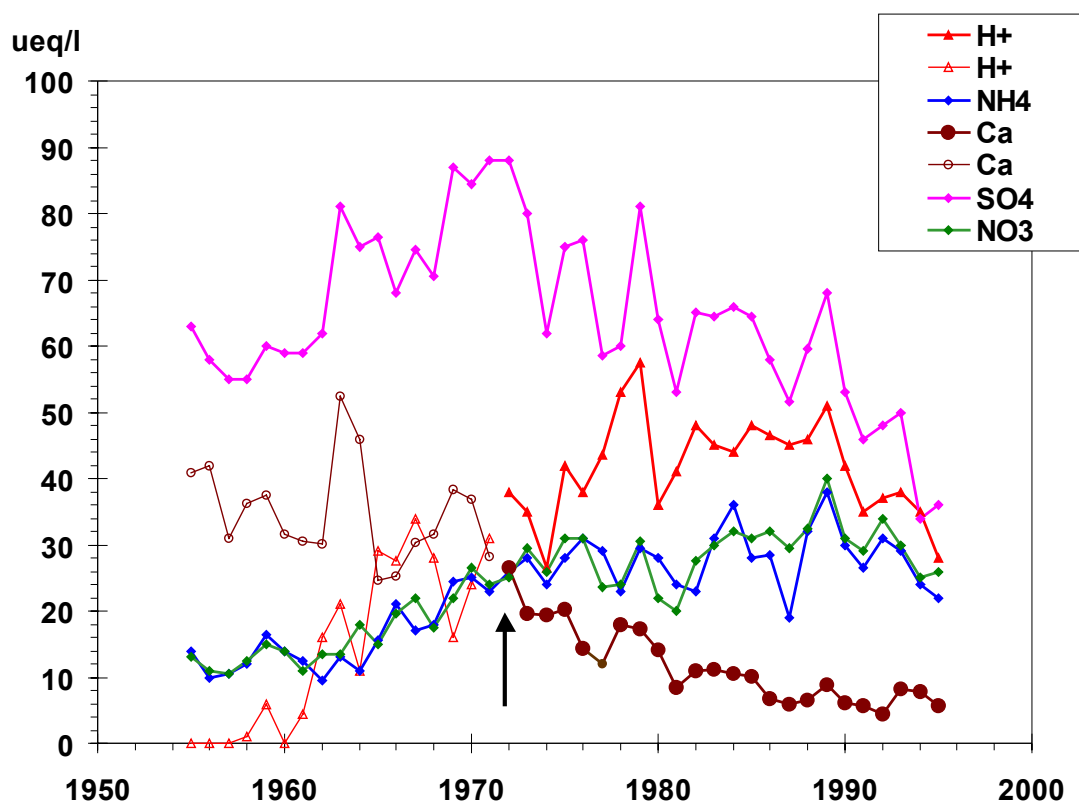
*Monitoring base cations in air and
monitoring base cation deposition as input
data to modelling – National experiences*

Base cations measured in the European Air Chemistry Network

Lennart Granat from Stockholm University, Sweden

Lennart Granat presented long term precipitation chemistry measurements from central Sweden. Measurements started at several locations all over Sweden in the mid 1950:s as part of the European Air Chemistry Network (EACN) and was gradually replaced with new and better located stations from the early 1970:ies. From 1983 a coherent network with some 30 well located stations over all of Sweden was in operation.

Traditionally the trends are based on data from many stations in an area of central Sweden with a notice that before around 1972 data on base cations and acidity are biased due to sampling equipment and –location, see the figure below.

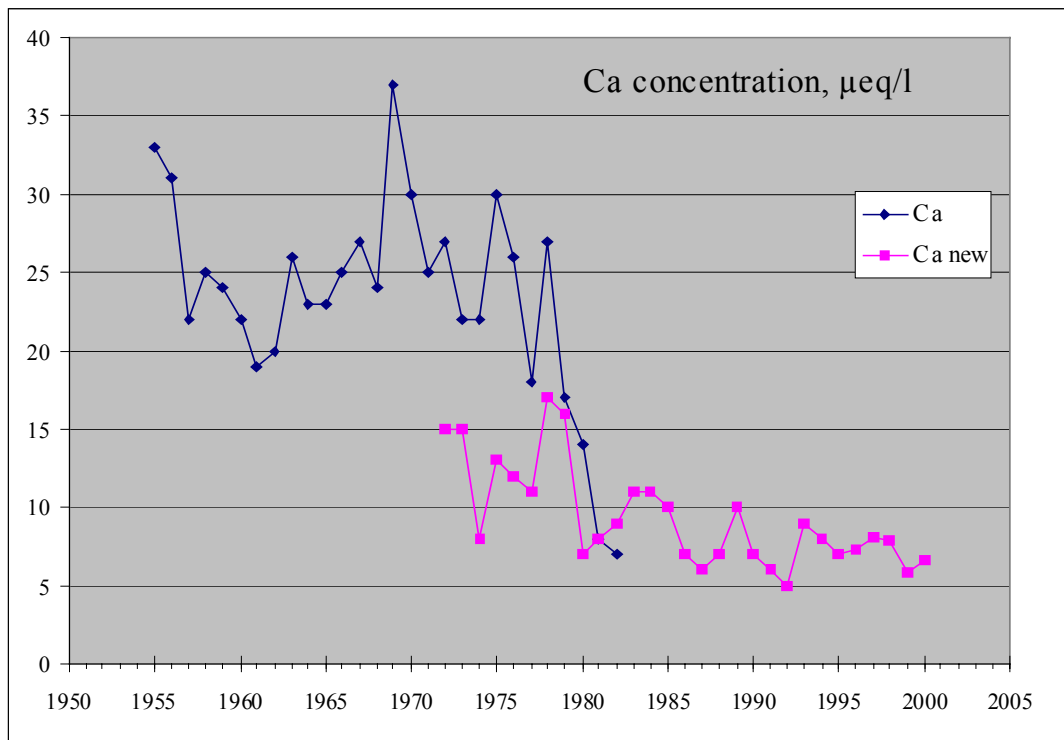


Long term changes in precipitation chemistry in Central Sweden. The arrow shows when locations and sampling equipment were changed.

The bias was clearly demonstrated by detailed comparisons of sampling equipment and sampling sites and is, in the area, related to soil derived components. With this in mind a new graph was made based on available good sampling stations, from the beginning only 1 to 3 in number and then gradually increasing, see the figure below. In the same graph is also given data from old stations with their equipment. The bias in the old measurements is obvious and the magnitude about the same as was obtained in direct comparisons.

The graph suggests that Ca concentration in the region was around 10-15 $\mu\text{eq/l}$ during the measuring period from mid 1950:ies towards the later part of the 1970:ies. Then a decrease started and was continued until the present level was reached. The decrease after 1983 is well supported by data from the national network for all of Sweden.

For estimating historical input of base cations (after 1955) the numbers given here can be judged as the most likely ones for the region. For other parts of Sweden a geographical scaling can be applied based on the geographical distribution in the 1980:ies before the final decrease in Ca concentration took place. Although uncertain these estimates are likely to come closer to real conditions than using for instance present day data.



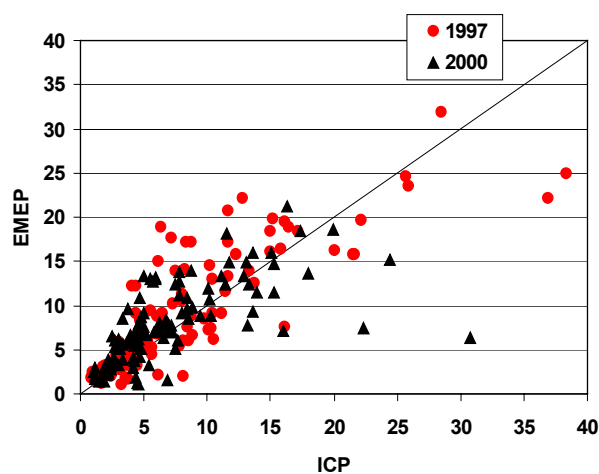
Ca concentration obtained from EACN stations and equipment (Ca, dark line) and from new stations with better location and sampling equipment (Ca new, light line). All stations were located in central Sweden.

*Monitoring base cations in air and deposition
as input data to modelling - the European
perspective*

Data available over Europe for base cation deposition mapping – the ICP Forests data

Olle Westling from the Swedish Environmental Research Institute (IVL), Sweden,

Olle Westling presented the possible use of ICP Forest database for a base cation deposition assessment. Good quality observational ICP Forest data from seven countries have recently been used to compare total deposition from measurements (open field and throughfall) and modelled data from the Unified EMEP-model (ecosystem specific deposition in the 50 km x 50 km grid) in 1997 and 2000. The comparison showed that the correlation was good for sulphur total deposition. Nitrogen compounds total deposition was not tested because of large uncertainties in canopy exchange. Wet deposition (bulk) measurements agreed well with modelled rates. The results were important as a model validation exercise due to good correlation and data independence. The findings of the study will encourage performing further studies of this type, e.g. measured and modelled base cation deposition.



Total deposition of SO₄-S in coniferous forests, kg per ha and year. EMEP_{modelled} wet + dry vs. ICP throughfall

Concentrations in precipitation		EMEP 1997 mg/L	ICP 1997 mg/L	EMEP 2000 mg/L	ICP 2000 mg/L
SO ₄ -S	average	0.64	0.63	0.49	0.52
SO ₄ -S	median	0.62	0.58	0.50	0.50
NO ₃ -N	average	0.44	0.45	0.40	0.41
NO ₃ -N	median	0.48	0.44	0.39	0.40
NH ₄ -N	average	0.43	0.49	0.42	0.43
NH ₄ -N	median	0.42	0.45	0.42	0.39

Volume weighted concentrations in precipitation (EMEP wet and ICP bulk deposition).

Modelling base cation deposition from emission data and from air quality and wet deposition data

Using the EMEP model for bas cation deposition

Leonor Tarrasón, Meteorological Institute, Oslo, Norway

Leonor Tarrasón, from EMEP/MSC-W, presented the use of the EMEP Unified Model for ecosystem-specific base cation deposition in Europe.

MSC-W participates in the current and forthcoming Nordic projects on base cation deposition. The monitoring data for base cations are available, but emission data are not inventoried yet, although they can be included in the particulate matter emission inventory. New data on natural sources will be compiled in EMEP in 2004, when initial results for long-range transported dust deposition are expected. However, the sea-salt deposition has already been computed and mapped, and could be delivered if necessary. A suggested new particulate matter task group under Task Force on Emission Inventories and Projections (TFEIP) would concentrate on chemical speciation, and could be useful in quantifying base cation content in emissions. The model validation continues in 2005, with expected improvements in dry deposition routines. Historical emission and consequent deposition estimates would be foreseen earliest in 2006.

In summary

Modelling base cation deposition is now beginning for good within EMEP (and conclusions from the BC workshop are crucial to that development)

2004:

- Initial results with respect to natural sources can be expected – estimates for present situation

2005:

- Validation of model results
- Improvement of anthropogenic emissions (?) included in the Unified EMEP model
- Improvements for dry deposition routines

2006 (and onwards):

- Historical emissions – requirements on climatology and land use changes for natural emissions (new projects may be necessary)

*Modelling base cation deposition from
emission data and from air quality and wet
deposition data*

**Case study - The Nordic mapping of base cation deposition
using data assimilation**

**Christer Persson from the Swedish Meteorological Hydrological Institute
(SMHI), Sweden**

Christer Persson presented MATCH model application to map Nordic base cation deposition using simplified data assimilation. The dry deposition fraction was estimated on the basis of few available concentration measurements to be ca. 20% of total base cation deposition. A detailed description of the results can be found in the background document of the BC workshop (Lövblad et al. 2003: The deposition of base cations in the Nordic countries, manuscript).

Conclusions

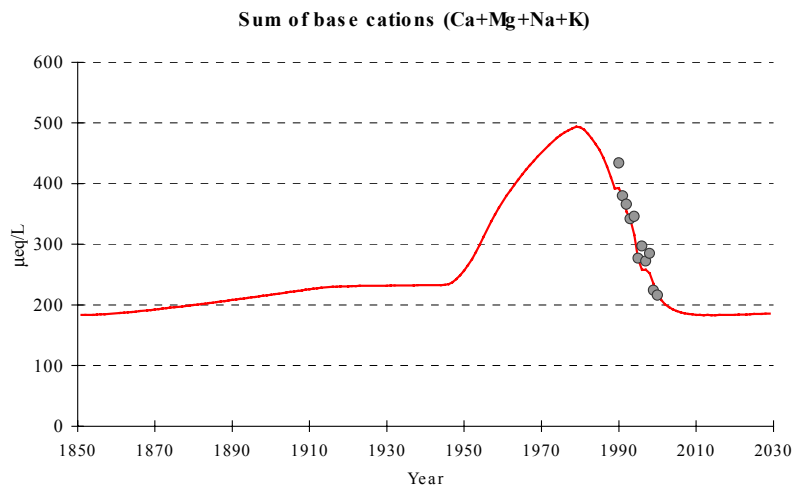
- Lack of information on air concentrations and dry deposition processes cause large uncertainties in mapped dry deposition
- Present results indicate that dry deposition is about 20% of total for Ca, Mg, Na
- Quality control of utilised atmospheric chemistry data is of large importance
- No consistent Nordic base cation emission database has been available for the project - but should be valuable for future studies
- For year 2000 and later necessary meteorological data is prepared for the whole of Northern Europe, all Europe can be included in the future

The inclusion of base cation deposition data into the mapping of critical loads

Need for data in the critical load calculations and in calculating ecosystem recovery

Filip Moldan from the Swedish Environmental Research Institute (IVL), Sweden

Filip Moldan presented the needs of base cation deposition for critical load calculations and recovery estimates with dynamic models. There are four base cation input parameters to models of about same magnitude and importance. Dynamic modelling results show fast and substantial recovery of lakes. Forest soils have only stopped acidifying without much recovery, though in relative terms some improvement can be seen. The derivation of acidifying and neutralising deposition inputs has varied between sites and nations, since there have been no official data or guidelines as requested from EMEP.



Example of modelled historic and future concentrations of base cations in runoff from the Lysina catchment, Czech Republic (Hruska et al., 1996) during 1850 to 2030. Dots are measured values

Conclusions

- BC deposition is very important for dynamic models, relative importance larger now than before because of higher demand for the precision of the modelling both past and future deposition important
- even small changes in BC budget important for the long term development of soils and consequently of waters
- time table for the regional assessment of recovery with dynamic models is set, the call is issued, and deadline for results is March 2004. If BC deposition is not provided, the countries must decide on their own.
- If the 50x50 km ecosystem specific 1850 - 2030 BC* deposition is unrealistic to achieve, guidelines would help.



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