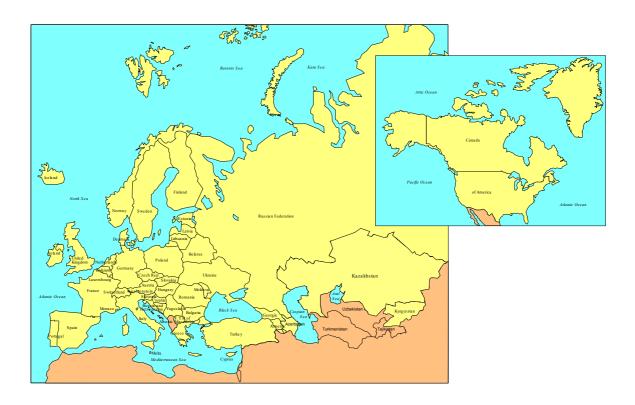
### UNECE CONVENTION ON LONG-RANGE TRANSBOUNDARY AIR POLLUTION



**EMEP** Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe

# **The EMEP monitoring strategy 2004-2009**

# Background document with justification and specification of the EMEP monitoring programme 2004-2009





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## Preface

The start of the long-term data series of air and precipitation chemistry observations, which are still available for EMEP, was established through the OECD-project LRTAP in the early 1970s. EMEP's role in continuing the observation programme, since 1979 under the Convention on Long-range Transboundary air Pollution, has ensured the long-term continuity of this unique data set. The monitoring programme has formed one of the basic pillars on which EMEP and the Convention itself rest, providing high quality data on the state of the environment, for model validation and national air quality assessments, national involvement and for independent validation of abatement measures. It has been essential to assure that measurements were made using comparable and reliable methodologies across Europe. For this reason the EMEP monitoring programme has employed fairly simple, robust and cost efficient methods. These have the additional advantage of being fairly easy to assess in terms of precision and reproducibility. Even globally, the EMEP data set is unique in these respects.

During the evolution of the programme new topics and priorities in air pollution control policies have entered the arena, but have generally not been associated with additional funding. This has resulted in large differences between different regions of Europe in their ability to implement the programme either in full or in part and to provide data of adequate quality. A shortcoming of the Convention and its protocols might be seen as a lack of specific requirements for the number of sites or parameters to be monitored; they provide no guidance to what is required to meet the programme's and the Convention's needs. One major objective of the new strategy is to identify clearly the monitoring requirements needed to underpin the work under Convention by defining minimum monitoring requirements for Parties. The strategy, also aims to describe the need for collaboration with other conventions (e.g. HELCOM, OSPARCOM) and organisations (e.g. AMAP), the European Union, especially with regard to its Air Quality Daughter Directive, and the Research Community at large in order to ensure that the efforts of all complement one another.

The EMEP monitoring strategy aims at preserving the long-term perspective. Tracking the changes over time is a major objective, through ensuring the continuation of the existing data series. At the same time, new priorities for policy-making have appeared in particular related to air quality and human health, to climate change and to the role of air pollution in changing biodiversity.

EMEP faces new challenges for its monitoring and modelling activities. New priorities place additional demands on the monitoring programme both for the number of parameters to be monitored and also for an increased density of sites. We need, for example, better spatial resolution to evaluate the influence of regional input to suburban and urban areas, site-specific exposure estimates to ecosystems, and additional parameters to understand the importance of hemispheric scale transport and the behaviour of atmospheric particulate matter. New technologies and tools are becoming available that can significantly improve our basis for making sound decisions on air pollution abatement, specifically earth

observation systems and data assimilation. As these techniques develop, further revisions of the strategy will be needed. Resources and requirements must, however, be balanced.

The strategy specifically addresses:

- The need for a revised monitoring activity by reviewing EMEP's strengths, weaknesses, opportunities and threats in relation to its objectives and the requirements of the Convention (Chapters 1-4);
- The importance of EMEP data for national as well as other regional or global programmes in order to improve the harmonisation and effective use of resources (Chapter 5);
- The need to establish obligatory monitoring requirements for those participating by applying a level approach (Chapter 6);
- The activities needed for individual topics under EMEP (Chapter 7) while monitoring requirements for individual Parties is presented in Chapter 8.

This report presents the monitoring strategy and the detailed monitoring requirements of EMEP as needed by the Convention for the period 2004-2009. A shorter version of the draft strategy will be presented for the EMEP Steering body in September 2003 (EB.AIR/GE.1/2003/3/Add.1, see Appendix A).

The strategy as presented has been prepared by the EMEP-CCC with contributions from the EMEP bureau, MSC-W and MSC-E and the national experts through the Task Force on Measurements and Modelling. Many individuals and institutions have thus assisted in the preparation of the strategy. We would in particular like to thank Wenche Aas, Urs Baltensperger, Knut Breivik, Jan Willem Erisman, Alexey Ryaboshapko, Martin Schlabach, Sverre Solberg, Till Spranger, Juha-Pekka Tuovinen and Mark Sutton for their valuable contributions.

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# The EMEP monitoring strategy 2004-2009 Background document with justification and specification of the EMEP monitoring programme 2004-2009

#### 1. Introduction

The "Cooperative programme for monitoring and evaluation of long-range transmission of air pollutants in Europe" (EMEP) was launched in 1977 as a response to the growing concern over the effects on the environment caused by acid deposition. EMEP was originally organized under the auspices of the United Nations Economic Commission for Europe (UNECE). Since 1979 it has been an integral component of the Convention on Long-range Transboundary Air Pollution with the EMEP Steering Body being one of the main subsidiary bodies of the Convention answering directly to the Convention's ruling body, the Executive Body.

International air pollution agreements depend on scientific credibility. The Convention, the United Nations Framework Convention for Climate Change (UNFCCC) and other conventions have developed their own ways of achieving this. Under the Convention, a system has evolved over the two decades since the original adoption of the Convention in 1979. It consists today of an integrated network of working groups, task forces and programme centres through which the scientific credibility and technical underpinning is established. Within the network workshops, assessments and scientific evaluations have been important ingredients in the consensus forming process.

One of the main objectives of EMEP is to provide Parties to the Convention with information on depositions and concentrations of air pollutants, as well as on the quantity and significance of long-range transmission of pollutants and transboundary fluxes. The programme has four main elements: emission data, measurements of air and precipitation quality, atmospheric chemistry transport modelling and integrated assessment modelling.

The vision of EMEP is to be the main science-based and policy-driven instrument for international cooperation in atmospheric monitoring and modelling, emission inventories and projections, and integrated assessment to help solve transboundary air pollution problems in Europe. To achieve this EMEP seeks to develop: SCIENCE – EMEP establishes sound scientific evidence and provides guidance to underpin, develop and evaluate environmental policies; PARTNERSHIP – EMEP fosters international partnership to find solutions to environmental problems; OPENNESS – EMEP encourages the open use of intellectual resources and products; SHARING – EMEP is transparent and shares information and expertise with research programmes, expert institutions, national and international organizations, and environmental agreements; ORGANIZATION – EMEP is organized to integrate information on emissions, environmental quality, effects and abatement options, and to provide the basis for solutions. The EMEP strategy can be found at <u>www.unece.org/env/emep/</u> while the complete programme is described at <u>www.emep.int/</u>.

Most countries in Europe have implemented monitoring programmes for air and precipitation chemistry and it is in their national interests to make sure that the data quality is as good as possible and comparable with similar observations over a larger geographical area. These measurements may be seen as just indications of ambient air quality, but they may also be used to address other objectives like understanding the causes of changes in the atmospheric composition including the effects of legislation and abatement measures. Due to the fact that air pollutants move across national boundaries, EMEP has played a central role for international cooperation in this field in Europe over the last 25 years. Priority has been placed on meeting national interests through bottom up structures and increasing the understanding and awareness on the national level by providing information that is transparent and of high quality.

The EMEP monitoring data forms one of the basic pillars on which EMEP, and the Convention itself, rest. It provides high quality data on the state of the environment, for model validation and national air quality assessments, national involvement and for independent validation of abatement measures. The data are essential for the technical underpinning of demonstrating the effectiveness of the implementation of current air pollution legislation, and for the work to revise and improve current policy. The EMEP observations are essential to establish a reliable picture of the air pollution situation in Europe, even in urban areas.

In the past EMEP has had substantial success in bringing East and West Europe together. Now an important challenge for EMEP is to extend its activity further eastwards into the newly independent states to ensure a monitoring system that can help quantify the movement eastwards of European pollution. EMEP has a particularly important role going beyond the political boundaries of the EU.

Following the Convention's adoption of its 1998 Protocols on Heavy Metals and Persistent Organic Pollutants and its 1999 Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone, the main priorities for the Convention are now a) the review and extension of existing protocols and b) the implementation of, and compliance with, existing agreements.

The EMEP data are used by a range of conventions and organisations. As well as reporting results through the EMEP Steering Body to the Executive Body for the Convention the results are especially important to the Convention's Working Group on Effects and its International Cooperative Programmes (in particular the ICPs on Forests, Integrated Monitoring, Vegetation, Waters, Materials and Mapping). Outside the Convention many organizations and Conventions use EMEP data: World Meteorological Organization (WMO); World Health Organization (WHO); Arctic Monitoring and Assessment Programme (AMAP); European Environment Agency (EEA); the marine conventions (HELCOM, OSPAR, MEDPOL/Barcelona Convention); Intergovernmental Panel on Climate Change (IPCC) as well as non-governmental organizations and national governments.

EMEP data are also widely used by the individual citizens and the atmospheric research community. While no detailed overview is possible, EMEP data have been used in thousands of papers and scientific reports and have thus been fundamental for improved scientific understanding, policy formulation and public awareness in relation to transboundary air pollution issues.

The EMEP observing and modelling system serves the national interests of Parties to the Convention by allowing them to assess the regional component of their air quality problems, and hence to arrive at cost-effective air pollution abatement.

The EMEP data are also being increasingly used by new groups of scientists such as the earth observation community and the climate modelling community as there is a strong requirement for high quality observational data on atmospheric composition also in climate research, chemical weather forecasting, assessment of air quality, and terrestrial and aquatic ecosystem change. In this way, EMEP data support regulations and directives for sustainable environment in a broad sense. For instance, the effects of greenhouse gases and aerosols on climate are similar in magnitude but work in opposite directions. Also the effect of aerosols on climate has one of the largest uncertainties (IPCC). It is thus obvious that the compounds regulated by the Convention's protocols also contribute to other air pollution issues ranging from urban air quality to climate change and associated effects (biodiversity etc.). Aerosols and tropospheric ozone are good examples. The EMEP region constitutes an important part of the global atmospheric environment so EMEP has a heavy responsibility to be a driving force in global monitoring as judged from the history of anthropogenic emissions and economic strength.

The new monitoring strategy considers the current strengths, weaknesses, opportunities and threats of the programme, including the level of reporting by Parties, new requirements from the users of data and recent technical developments. Special areas for discussion include the linking of scales (hemispheric-regional-local), improvements in the ability to estimate site-specific deposition and exposure are requested in order to better assess the negative effects to ecosystems and human health; improvements in the cooperation within the Convention, especially between EMEP and the Working Group on Effects and their programmes, the availability of new techniques, such as data assimilation, the use of remote sensing techniques and flux monitoring which have the potential to significantly improve our capabilities for implementing sound abatement measures.

#### 2. Objectives of the monitoring programme

Observations are fundamental to the progress of our understanding of atmospheric chemistry, in the estimation of regional emissions of pollutants, in the follow-up of emission reduction policies and in the assessment of regional concentrations and deposition of pollutants and their associated effects. Monitoring may in this context be defined as long-term measurements of various parameters to provide information of the geographical variations and changes by time in the atmospheric chemical composition.

The purpose of the EMEP measurement programme is to provide necessary air concentration and deposition data for the model development and improvement of the understanding of large-scale atmospheric dispersion and deposition processes. While no strict priority between of the various objectives has been given the monitoring serves to meet at least the following objectives;

- Establish pollutant concentrations, deposition, emissions and transboundary fluxes on the regional scale, including intercontinental transport and boundary conditions for urban air quality;
- Identify the trends with time as well as their sensitivity to European emission reductions;
- Assess the success of international abatement strategies for atmospheric pollutants;
- Improve the understanding of atmospheric chemical and physical processes and provide data for the validation of models;
- Provide data which, in conjunction with models, are the basis for the assessment of environmental problems related to air pollution including comparison with effect thresholds and exposure levels;
- Provide measurements required to assess the effects of atmospheric pollutants;
- Serve to explore the environmental concentrations of new substances and support the development of cost-effective abatement strategies.

The monitoring activities also serve to raise awareness in the participating countries and to provide relevant information to the public as well as to the atmospheric research community. The national monitoring within EMEP is also instrumental in building up national competence in atmospheric chemistry and in understanding the environmental impact of atmospheric pollution. The active involvement by national experts has been essential for the success of EMEP and this needs to be continued. In the future, the use of monitoring data to estimate emissions will be an important element for evaluating compliance with protocols.

## 3. Requirements for meeting the objectives

In order to address the objectives specified above some general requirements to the monitoring activities are obvious and these are presented in this chapter. Specific requirements for addressing the individual objectives are given in Chapter 7.

### 3.1 Data quality

In order to meet the EMEP objectives, the monitoring methods must satisfy certain criteria. It is essential that the measurements reflect the air quality in a representative manner and that the methods applied are consistent and free of artefacts. Long-term monitoring, in particular, requires that measurement series can be continued consistently for many years.

The purpose of the monitoring must be reflected in the measurement method. If observations are the basis for the warning of the public, or for assuring that air quality is acceptable in relation to recommended criteria, then it is particularly important that the measurement at or above the air quality limit are reproducible. and with an acceptable accuracy, and that the measurements are available in near real time. If, however, the purpose is long-term monitoring for detection of trends, the mean and certain percentile concentrations are more appropriate. These concentrations are generally much lower than the air quality limit values. Reproducible measurements without artefact or interference are needed. In order to indicate trends over several (5-10) years, precision of individual measurements should be within  $\pm 10\%$ , and any systematic change over time should be less than the expected trend (1% year<sup>-1</sup>). This can be hard to achieve with automatic monitors requiring periodic calibration. An example here is the experience from ozone monitoring in Europe. Only few sites have provided data of a satisfactory quality in order to assess the trends, and the main problem is missing information about calibrations and changes of instruments. This is an example that the EMEP data are subject to stricter quality requirements than data aiming mainly at assessing exceedance of air quality guidelines, where such shifts obviously are less critical. The situation is better for chemical or gravimetric methods, but procedures and details needs to be rigorously followed and documented.

For assessing long-term trends, quality assurance, high precision and consistency in the data records are of the utmost importance. Measurement sites should not be subject to changes in surroundings, or to changes in instrumentation, unless the impact of changes is carefully evaluated and documented. Similarly, any changes in sampling and analysis procedures should be documented and evaluated, and sampling period and data completeness should meet the existing data quality objectives. The data quality objectives of EMEP can be found in the EMEP manual for sampling and chemical analysis (EMEP/CCC, 1996).

Experience has shown that measurements should be standardized as far as possible to obtain data that are comparable and of sufficient quality. In addition, quality assurance has to be carried out on both the national level and by the CCC to ensure satisfactory data quality. This applies to individual samples and to long-term aggregated values, such as seasonal or yearly mean values. It is particularly important to avoid systematic errors and undefined changes in the data quality over time, which may cause problems in trend analyses.

For the majority of the methods, the necessary quality assurance is facilitated by a combination of simple and robust sampling techniques with well-described sampling equipment, and use of synthetic control samples for assessing the quality of the chemical analyses.

#### 3.2 Site representativity and spatial resolution

The representativeness of a given site needs to be evaluated. This can only be determined in relation to the purpose of the measurements. For EMEP the site must be located so that the measurements of air quality and the precipitation chemistry parameters are representative of a larger region. In order for a site to be representative, influences and contamination from local sources must be avoided. In regions where local emission sources are abundant and make it difficult to find locations satisfying the site criteria, the sites should be "typical" for the region. Obviously, monitoring data from regions more directly affected by local emission sources areas may provide essential information in order to evaluate their relative

contribution. It is not recommended that the existing EMEP sites are relocated to measure in such regions, since this is for some parameters done in support of the EC Air Quality Daughter Directives and the data are thus available to EMEP. For parameters being subject to local emission sources at rural sites (like ammonia) monitoring should be complemented with a programme making it possible to assess the relative importance of the local contribution.

To map concentrations and exposure on a European level, data are required at a resolution matching the spatial variability of any given pollutant and reflecting the resolution of the models being applied. With respect to secondary pollutants, with low spatial variability and low formation rates, the current EMEP network density is sufficient in large parts of Europe, but insufficient in eastern and Mediterranean parts of the Continent. For more spatially variable species, there is a need for denser monitoring as well as integration of data from different programmes (e.g. for ammonia, wet deposition and ozone measurements). For some substances, the variability is expected to be much larger than can be resolved by integrating even all available measurements, and the studies need to be supported by assessment of local scale variability, e.g. by passive samplers or other low cost methods.

Networks for trend evaluation need to cover in a representative way the climatic zones across the domain. In order to understand the temporal evolution (trends) there is also a particular need for high quality measurements at sites with little influence from local and regional emission sources.

#### 3.2.1 Improving air quality in populated areas

In spite of the fact that the emission levels in Europe as a whole have been steadily decreasing during the last 10 to 15 years, and that further reductions are expected in the years to come, air pollution will still remain a (potential) health problem in most European cities (EEA, 2003). Presently, Chemistry-Transport Models (CTMs) are used, together with air quality measurements, both in air quality assessments and for planning abatement strategies. It has been realized that application of urban scale models or urban measurements alone is not adequate for assessing the air quality on the local scale. Urban scale models are highly sensitive to what is prescribed at the model boundaries. To remedy these problems it is common to couple (or nest together) regional CTMs with urban CTMs. The proper way of building complex modelling systems, relay heavily on availability of high quality measurement data. Since these coupled models cover both regional and urban areas, measurements representative for these scales are needed as well. Moreover, a research area within this field of air quality modelling is presently to use (or assimilate) available observations as input to the CTMs during the model simulations, a technique that evidently will lead to a growing demand for appropriate observational data. EMEP observations are thus important for validation of the regional CTMs used for providing the boundary conditions for urban scale models. Often, EMEP observations are also used directly to indicate the regional background when being representative.

#### 3.2.2 Ecosystem deposition fluxes

Deposition of air pollutants can cause severe damage to ecosystems. On the local scale, deposition amounts will depend on precipitation patterns and surface

characteristics. It is a major challenge to quantify so-called site-specific deposition amounts. In order to develop effective emission reduction policies and to monitor how the deposition levels change, a combination of modelling and measurement activities are used, where measurements serve as independent checks on the modelled concentration and deposition fields. Wet deposition is routinely monitored and provides spatial aggregated fluxes, while dry deposition of gases and particulate matter is more difficult to measure and it is not possible to provide reliable area specific fluxes. The approach to estimate deposition of sulphur, nitrogen, ozone and base cations must therefore be based on a combination of detailed monitoring at some (super) sites providing process information, with less demanding methods applied at a larger number of sites in combination with deposition models.

The EMEP/WMO workshop on Monitoring Strategies (EMEP/CCC, 1997) (see www.nilu.no/projects/ccc/aspenes98/) produced a number of conclusions and recommendations of relevance here. With respect to spatial resolution and representativity of measurements it was stated that: *Effects-related monitoring is* needed to define exposure-effect relationships at experimental sites, to provide high-resolution regional maps of exposure, and to help validate atmospheric transport models. The spatial coverage of sites has to be improved, especially in Southern and Eastern Europe. POPs are inadequately measured. Base cation concentration measurements in air are needed. EMEP should aim to estimate dry deposition at a large number of sites by using inferential modelling and at a small number of sites by continuous measurements. The latter should be used primarily to develop and test inferential models. Techniques need to be developed to monitor cloud concentrations and deposition in order to estimate exposure to them. Monitoring at experimental sites is performed at an international level by ICPs, as well as by national effects assessment projects. These data, mostly relving on throughfall measurements in forests, should be used by EMEP, but considering their lack of representativity (see also Chapter 5.1). Passive samplers should be applied to support high-resolution models and measurement networks for substances with a high spatial variability. Concentration measurements and high resolution inferential modelling should be combined. Sub-grid evaluations are of importance in all regions including those where critical loads are exceeded. This is because of the scale dependence of critical load exceedances, how they changes in the future and because they are necessary for dynamic modelling, independent from whether critical loads are presently exceeded. These recommendations are still valid.

#### 3.2.3 Intercontinental transport

Intercontinental transport of pollutants and emissions from marine and atmospheric sources outside Europe influence the air pollution levels and thus also abatement policies within Europe. The importance of this transport varies between the different compounds due to differences in their atmospheric residence times. The hemispheric scale concentrations of ozone and particulate matter are of particular importance, since they may severely influence the control needs and strategies on a regional scale. The same also holds for some POPs and heavy metals, for which it can be shown that more than 50% of the pollution load in Europe might originate from non-European sources. Even the hemispheric scale

deposition of sulphur and nitrogen is large enough to influence the control needs for the achievement of environmental objectives in Europe (critical loads).

Hemispheric concentrations of ozone, particles as well as of sulphur and nitrogen compounds are also of importance for global policies on climate change. It would thus be important to extend the modelling and monitoring domain in order to address importance of intercontinental transport and changes in the global atmospheric chemistry. This issue is also strongly related to the driving forces for climate change: the concentration changes of methane, tropospheric ozone, particulate matter and its composition, and even  $CO_2$  since the biogeochemical cycle of  $CO_2$  is influenced by e.g. changes in nitrogen cycle.

In order to properly address the issue of intercontinental transport the measurement programme needs further developments. This includes establishing sites in areas not yet sufficiently covered. In particular it would be important to establish new sites in the far east of the EMEP region (Kazakhstan etc.), on the African coast of the Mediterranean and in the Eastern Mediterranean. In addition, monitoring networks should integrate to form "multi purpose" networks linking urban (surface), rural (transboundary surface and low local influence) and global sites (surface, low local/regional influence).

For compounds with significant influence from intercontinental scale transport (PM,  $O_3$ ,  $NO_y$ ,  $SO_x$ ), improved understanding of the vertical gradients, as well as the free troposphere concentrations are required. A significant fraction of the pollutant transport can occur between continents as well as within the continent. EMEP surface measurements need to be complemented with vertical ozone soundings or aircraft measurement programmes. Ozone soundings networks are presently in operation under other initiatives and data could be made available for EMEP use.

To provide an observational basis to analyse the intercontinental transport of air pollution, EMEP need to work with earth observation from satellites, remote sensing from the ground, in-situ measurements from sondes and aircraft including routine observations from commercial airliners, surface measurements including mountain peak and remote region sites. A collaborative structure needs to be reinforced to extract the information content from observations, and also to make progress instrumentation, quality control and data assimilation.

#### 3.3 Need for complementary data

In EMEP observation data and chemistry transport modelling needs further integration to establish the relation between emissions and deposition fluxes or exposure levels. To properly evaluate model performance, measurements of the relevant species involved are required both in air and precipitation. An important illustration is  $PM_{10}$  which can only be modelled considering the emissions of the various gaseous precursors, particles emitted from primary sources, their atmospheric intermediates, reaction rates and finally their removal mechanisms. Thus, only by measuring a wide range of compounds (i.e. the major inorganic compounds in gaseous and in particulate form, their physical characterisation and precipitation chemistry routinely measured at a relatively large number of sites) can a theoretical model calculation be validated. A list of parameters is proposed

as mandatory requirement data at EMEP Level 1 and level 2 sites (as defined in Chapter 6), while a brief introduction to the monitoring requirements for the individual topics is given in Chapter 7.

#### 3.4 Dry deposition and non-linearities

Pollutants with a rapid exchange with the ground surface, such as ozone,  $HNO_3$ ,  $SO_2$  and  $NH_3$ , exhibit large vertical concentration gradients above the surface. The sampling height needs to be known when measurements are used for modelling, i.e. calculating fluxes using deposition velocities. In the case of  $NH_3$ , the vertical gradient can vary widely because surfaces may act both as sources and sinks. To limit the influence of upward  $NH_3$  gradients, monitoring should take place over grass without fertilisers or grazing. For slowly exchanging species, such as particulates, this issue is of less concern.

In large areas of Europe, dry deposition of sulphur and nitrogen species, as well as of base cations is larger than wet deposition, but dry deposition calculations by models are generally not well validated. Only very few sites in Europe measure dry deposition continuously to allow for model validation, and further effort needs to be made in this area. EMEP must aim to monitor dry deposition at a limited number of sites by using flux based monitoring for comparison with inferential approaches.

It is often assumed that deposition is proportional to the emissions. This is the case on the continental scale, but may not be the case on the local scale where the deposition-emission relationship may be non-linear. There are many observations indicating that such non-linearities are important in particular for sulphur and nitrogen (e.g. Fowler et al., 2003). Some of the processes responsible for these non-linearities are understood, but the scale of the effects and their consequence for policy development has not been evaluated over Europe. It is known that there are important non-linearities between ambient concentrations of SO<sub>2</sub> and NH<sub>3</sub> on the dry deposition of these gases. For reduced nitrogen, non-linearities occur locally where high ambient NH<sub>3</sub> concentrations suppress the deposition rate. These non-linearities are usually not accounted for in the source-receptor modelling and this introduces considerable uncertainty in the calculated amount of deposited nitrogen and consequently in the impact evaluation. Until recently, monitoring methods for dry deposition fluxes were expensive and only applied for research purposes at a limited number of sites. The EUROTRAC2 – BIATEX2 community has however developed methodologies proven to be applicable also for long term monitoring at a low cost, provided the sites satisfy specific site criteria (see also Chapter 4.2.2.1).

Ozone concentrations at canopy level are needed for assessing its impact on vegetation and steps are taken to adopt a flux-based approach. In that case fluxes need to be measured in order to determine the uptake through stomata.

#### 3.5 Supersites

As described in Chapter 3.3, a detailed understanding of the atmospheric physical and chemical processes are required in order to evaluate and further develop atmospheric models. Detailed observations of atmospheric processes are of particular importance, but can for a variety of practical reasons not be established at a large number of sites. State-of the-art measurements of this character need the support of research groups and include new techniques and platforms such as flux based monitoring, remote sensing and earth observations. Such research activities forms part of the EMEP monitoring strategy and take place on supersites in cooperation with the research community. Supersites can include sites established for short-term research campaigns as for some parameters even discontinuous data would be useful for process studies.

EMEP supersites will be nominated according to topic and each site would not necessarily need to cover all topics. It is advised that countries cooperate for cost sharing purposes. A number of so called "Large scale facilities" funded by the EU already exists and these need further development to cover the whole range of advanced parameters relevant to EMEP. A close cooperation with GAW on this issue is essential and the establishment of joint supersites for particulate matter is currently being implemented.

Sites that represent regional or global concentrations are encouraged to develop into supersites. High quality EMEP stations are encouraged to become supersites by implementing extended measurement programmes or specialised programmes for a particular pollutant, documenting adequate quality and technical staff. The supersites also need to satisfy the Data Quality Objectives defined for EMEP.

#### 3.6 Accompanying meteorological data

As mentioned above, EMEP monitoring sites are ideally representative of a larger region and not much affected by local conditions. In practice, however, the local conditions to a certain extent influences the measurements at a given site. Accompanying meteorological measurements at the EMEP sites are needed to evaluate the local influence of the station. Such data could help reveal both any local emissions and the importance of local meteorological conditions.

To evaluate the influence of local emissions, at least wind speed and direction is needed. Thermal stability measured by the vertical temperature gradient (e.g. by the temperature difference between 2 m and 10 m) is a measure of the mixing processes near the ground. Many of the EMEP sites experience frequent situations when a shallow boundary layer is "decoupled" from the troposphere above, particularly during night-time inversions, and then the measurements are only of local value. Such situations complicate the comparisons with regional scale models significantly, as the models usually do not resolve small-scale inversions. Local meteorological data can also be used for the direct evaluation of the other chemical observations at the site.

# 4. Strengths, weaknesses and opportunities of the EMEP measurement programme

This chapter summarizes the status of the EMEP measurement programme in relation to its objectives, and explains the need to improve the measurements and the network to meet its objectives.

The EMEP project organization is set up to detect trends in pollutant concentrations and deposition. A network of sites is in operation, instrument requirements are specified and quality assurance and quality control systems are in place though in most countries improvements are required. It is essential that all Parties to the Convention comply with the current requirements of the measurement programme and their future revisions, including instrumentation, quality assurance and quality control systems.

Long time series of measurements in Europe are available for specific chemical constituents of aerosol, such as particulate sulphate, and for precipitation chemistry. The particulate sulphate series go back to 1972, and although the equipment and the data quality have improved over the years, some of the data series are fully consistent over this long time period. Much less information is available for nitrate and ammonium because these measurements are much more susceptible to sampling artefacts, measurements were initiated much later and at fewer sites. Some EMEP observations are of sufficient quality to derive trends in SO<sub>2</sub>, particulate sulphate, NO<sub>2</sub>, ozone and heavy metals, while for VOCs, POPs and particulate matter, time series are short and the spatial coverage is still poor.

The performance of the laboratories is documented in the annual interlaboratory exercises. In general there has been a significant improvement in laboratory performance during the last years and at present, the performances are generally good for sulphate and nitrate in precipitation and for nitrogen dioxide in air. Some laboratories experience problems with the determination of ammonium and calcium. This may be due to contamination problems. The concentration levels for calcium in the test samples are low relative to the concentrations at EMEP sites in these countries. The determination of pH is also less accurate than for the other parameters, and the criteria for acceptable results for pH may have to be relaxed somewhat. However, laboratories are also reminded that pH measurements may need particular attention, checking the performance of electrodes with appropriate test solutions at regular intervals. The performance is strongly related to available equipment and resources.

Even if the laboratory performance is satisfactory, results at many EMEP sites are poor because inadequate sampling methods are used. The number of days with concentration values below the respective detection limits is given in the annexes to the annual EMEP data reports (e.g. Hjellbrekke, 2002). However, these detection limits are generally lower than the typical deviation experienced in the interlaboratory tests. Therefore, it is likely that some of the reported air concentration data may not be satisfactory, even if the laboratory performance is acceptable.

The laboratories serving the sites in North, Western and Central Europe are generally well equipped and perform satisfactorily in the laboratory intercomparisons. Many of these sites also have excellent long-term records of particular value for trend analyses. France, Spain and Portugal have applied methods for determining sulphur dioxide and nitrogen dioxide (mainly wet absorption solutions) that are not sensitive enough for the low concentrations usually experienced at the EMEP sites. In Spain and Portugal, measurements often deviate widely from model results. The ambient concentrations in these countries are to a large extent caused by indigenous emissions, and the concentration and deposition fields are not well described by the current EMEP models. This is due partly to the climatic conditions, and also to the high proportion of the emissions from large point sources. Data quality is, however, also an issue (Aas et al., 2002).

The countries on the Balkan Peninsula have had unstable political and economical conditions during the past ten years. Croatia, Serbia and Montenegro, and the former Yugoslavian Republic of Macedonia, have to different degrees maintained their network of measurement sites, and some of the countries are in a real need of resources and equipment. Bulgaria has not established EMEP sites, and Romania has not achieved satisfactory laboratory performance and has not reported data in recent years. All these countries have the necessary infrastructure to operate a network i.e. through their national hydrometeorological services, however.

Italy has failed to establish a proper national network of rural background stations for the EMEP programme. The two current sites are operated by a research organization and the European Union's Joint Research Centre in Ispra. Both sites are located near these two research centres, and are not satisfying the site criteria for rural background sites. Greece has established one EMEP site, but the laboratory performance has never been satisfactory. Turkey operates one EMEP site, with a good performance record, and is establishing two more sites: one closer to the Mediterranean Sea and one in the northeast. A new site has been established in Cyprus, and Malta has shown interest in joining the network.

In Russia, Belarus, and the Ukraine the measurement networks are suffering because of the general deterioration in public services. It is also difficult to arrange the transport of samples from the sampling sites to the laboratory, which is well equipped with ion chromatographs. Consequently, there is only one monitoring site in operation between the Baltic countries and the Urals. This is a severe deficiency of the EMEP network. The three Baltic countries have improved their monitoring activities significantly in recent years, but improvements are still needed.

The situation in the Balkans and in the Baltic countries calls for a general transfer of technology, including both equipment and training of personnel. Relatively modest investments are needed, but it is important that the national governments welcome such efforts, and that the facilities will also be used to serve national interests in addition to the countries' EMEP involvement. The strategy towards improving the situation in the Russian Federation, Belarus, and Ukraine is much less obvious. First priority should be given to the establishment of 3-5 additional properly located sampling sites, and to the operation of these sites, with proper transport of samples and materials. Then the central laboratory or laboratories must be given the necessary resources for the chemical analyses and for quality assurance. Technical cooperation between the three countries should be encouraged. For the remaining countries, priority should be given to maintaining and documenting the quality of the existing time series. In some cases, improvements in sampling methods and chemical analyses may still be necessary.

#### 4.2 Methods and new developments

This chapter discuss methods in use and other available methods, as well as new techniques that can significantly improve the observational basis for EMEP.

#### 4.2.1 Established methods

EMEP monitoring has always been based on fairly simple, robust and cost efficient methods. In an international programme where concentration and deposition level are compared across national boundaries and with many Parties involved, it is essential that the quality of the methods employed can be properly evaluated.

When EMEP was established, the monitoring procedures developed during the OECD-project were maintained. The OECD-project mainly focused on atmospheric dynamics and interpreting pollution episodes. To maintain long-term time series was of secondary importance. Whilst analysis of precipitation samples and particulate loaded filters is fairly straight forward, impregnated filters were a major innovation when they were introduced in the late 1970s. Most countries did, however, not change their methods until about 1988, and only few consistent data series for  $SO_2$  go further back in time.

A brief evaluation of the various candidate methods for monitoring air chemistry is given below. Precipitation chemistry is not further discussed as the methods in use can still be considered as state-of-the-art.

#### 4.2.1.1 Manual methods

Filter packs have traditionally been the reference method for sampling inorganic major compounds in both gaseous form and in particles. They have the advantage of being suitable for simultaneous sampling of both gases and particles (which then are directly comparable), they can be combined with size segregated sampling of PM, in combination with advanced analytical instrumentation it is reliable even at very low concentration levels and with fairly short time resolution (daily). The performance can also easily be documented by intercomparisons (field or laboratory). In addition it is easy to control the measurement uncertainty by taking field blank samples. The method is easy to employ, requires little training for field operators. The method is also quite cheap as a number of compounds can be determined at once. While the method is subject to artefacts in the separation of semi-volatile species (like HNO<sub>3</sub>/NO<sub>3</sub>, NH<sub>3</sub>/NH<sub>4</sub>), it can be combined with denuders to deliver results free from artefacts. Experience has shown however that in temperate regions, the gas to particle ratio as seen on impregnated filters does not deviate significantly from the ratio determined with artefact free methods. Some manual labour is needed for preparing the filters and for chemical analysis. The method also requires a pump and air volume recording.

- Recently, a low cost diffusion denuder system have been developed for ammonia sampling (DEnuder for Long Term Ammonia (DELTA method) (Sutton et al., 2001). The DELTA method may also be used to determine HNO<sub>3</sub>, NO<sub>3</sub><sup>-</sup>, SO<sub>2</sub>, SO<sub>4</sub><sup>2-</sup>, HCl, Cl<sup>-</sup> and base cation concentration in combination with filter pack sampling. By sampling at a low flow rate using relatively short and robust denuders these can be employed at a large number of sites. This may allow new measurements to be established in regions where data are currently missing due to cost/resource restraints.
- Passive samplers have attracted a growing interest over the last years. While retaining many of the advantages of the filter-pack, this method is particularly attractive as it does not require electricity, pump or recording of air volume and can easily be employed in large numbers. The latter makes the method particularly well suited for representativeness studies. It thus provides a relatively cheap alternative for gaseous compounds. Detection limits are, however, significantly higher and thus a longer sampling time compared to the filter-pack method are required (typically weekly or bi-weekly). A number of samplers also do not follow the theoretical geometry and thus requires empirical scaling. Some of the methods are not freely available. Countries have experienced mixed success with passive samplers, and quality control issues are critical.

#### 4.2.1.2 Monitors

- The use of continuous recording monitors has grown rapidly in air quality • monitoring networks. Their major advantages include high time resolution (typically 1 hour), rapid data acquisition and presentation, and that some quality control can be performed "on-line". Monitors are thus particularly suited for limit value exceedance assessment and the short time resolution may be useful in process studies or to detect temporary influence by local emission sources. Monitors do not necessarily require access and support from a chemical laboratory. Monitors do, however, generally have high detection limits, though "trace level monitors" are available. A monitor generally also gives information for one component only and the chemical composition only for gases. It is more difficult to intercompare and quality control them for an external body. It is evident from the EMEP field intercomparisons that monitors have a poorer performance compared to the manual methods (e.g. Aas et al., 2003). It is also our experience that monitors tend to be overvalued by inexperienced users and that maintenance often is insufficient. This also may lead to costs being underestimated for operation with the required attention and maintenance. Monitors are also more unreliable at low concentration levels and that makes trend assessments over long time-spans more difficult compared with manual methods. Monitors do also require housing, electrical power, etc. However, for ozone, monitors are the only widely available and reliable monitoring method.
- Spectroscopic methods (DOAS) have also received growing interest over the last years. Their advantages include some of those given above for monitors. In addition they integrate over a long path and thus are less influenced by local sources and features. The absorption spectrum from

DOAS instruments can be analysed for several gaseous components at once. DOAS instruments are, however, subject to significant uncertainties, which make them less attractive for application in EMEP. First of all they are difficult to calibrate and also the possible interference with other factors may be difficult to assess in most commercial systems. DOAS instruments are more difficult to intercompare with other measurements due to the different "siting criteria". Experience has also shown that many commercial systems often provide low data capture and with poor quality

As can be seen from the above, the filter-pack method seems most appropriate for the simultaneous determination of the major inorganic compounds on a daily basis (i.e. SO<sub>2</sub>, SO<sub>4</sub>, NH<sub>3</sub>/NH<sub>4</sub>, HNO/NO<sub>3</sub>, Na, Mg, Ca, K). The DELTA method resembles many of the advantages of the filter pack method, and in addition it allows for determining the correct distribution between gas and particulate phase. The DELTA method is thus proposed established on a monthly basis at EMEP sites in combination with the traditional filter pack measurements. Passive samplers have a great potential for improving some aspects of relevance for EMEP. These include site representativeness studies, studies in source areas, assessment of gas/particle distribution in combination with the filter pack method.

Continuous recording monitors comprise a useful supplement to the manual methods and should be operated at a limited number of sites.

#### 4.2.2 New methods and techniques

assurance.

#### 4.2.2.1 Dry deposition monitoring

While wet deposition can reliably be monitored, dry deposition fluxes are much more difficult to quantify. Micrometeorological methods have been available but these are generally not applicable for long-term monitoring. Over the last years, however, low-cost methods have been developed which have the potential of being applied at a selected number of EMEP sites. In particular the COnditional Time Averaged Gradient method (COTAG) (Sutton et al., 2001) have been demonstrated to provide reliable data for inorganic gases and particles (see also Chapter 4.2.1.1). It is thus recommended that EMEP establishes COTAG measurements at a selected number of sites or recruits some new sites (e.g. the Life sites (Erisman et al., 1998)) meeting the special criteria for site and surroundings. Flux measurements sites obviously also requires highly experienced staff. For ozone, more information is needed about dry deposition fluxes, and in particular in relation to uptake through the stomata. For this purpose, traditional micrometeorological gradient and eddy covariance methods will be applied.

#### 4.2.2.2 Earth observation

Earth observation systems can significantly improve our capabilities for atmospheric monitoring, at the same time ground observations are essential for calibration and validation of the remote sensing data. EMEP has to contribute to the development of satellite and other remote sensing observations by providing surface based measurements, and by this contribute to the European part of "Global Monitoring for Environment and Security" (GMES), Integrated Global Observing Strategy (IGOS) or other activities now being established. Space-borne sensors add information about key parameters and with wide spatial coverage. Individual observing systems are in general incomplete and insufficient, while the combination of ground-based and space-borne observations improves the observation of the chemical composition at the regional scale. Satellite observations further provides a connection between the various surface sites and provide information on spatial variability for network data interpretation. Auxiliary data as provided by the networks and models increase the value of the retrieved parameters substantially, and continuous ground-truth efforts will allow assessing the quality of the satellite data products.

# 4.3 Integration of observations and model calculations through data assimilation

A closer integration between the modelled and the observed values obviously seems as a natural way to proceed. In practice this is not trivial, and several approaches and levels are possible. Data assimilation is a well-known and widely applied technique in numerical weather forecasting, implying that the models are run in an iterative procedure to finally produce a result that optimises the model performance with respect to the observed meteorological data. For a CTM this becomes complicated due to the large number of physio-chemical processes and their non-linearities. Four-dimensional data assimilation of the EMEP model is thus presently a long-term aim foreseen for the future and at the moment limited by computer resources, programming effort and general knowledge in numeric and optimisation.

This should, on the other hand not prevent improvements in the synthesis of the modelled and measured data on a simpler level. There is a clear potential in the "harmonisation" or merging of observational and calculated regional data fields, e.g. by identifying regional differences in uncertainties and bias. This can in turn can be used for further model developments or modifications of the monitoring network. Furthermore, preparation of combined modelled-measured regional fields could be used for downscaling, i.e. producing maps with a finer resolution than the model grid. Clearly, additional data sets than EMEP, such as satellite data, vertical soundings and lidar data could be useful for this purpose.

### 5. Cooperation with other programmes in Europe

Through harmonisation with other monitoring activities EMEP shall develop a cost-efficient and multi-purpose monitoring strategy by integration with other air quality networks in Europe. This not only includes national networks and other programmes under the Convention (e.g. the International Cooperative Programmes (ICPs)) but also the Arctic Monitoring and Assessment Programme (AMAP), the Convention for the Protection of the Marine Environment of North-East Atlantic (OSPAR), the Convention on the Protection of the Marine Environment of the Baltic Sea Area (HELCOM), the monitoring performed under EC Air Quality Framework Directive, the Global Atmospheric Watch (GAW) programme of the World Meteorological Organisation (WMO) and others. Such interactions shall ensure an efficient use of resources by avoiding duplication of efforts.

National authorities are challenged to consider all national monitoring activities together to ensure that the demands imposed by the Convention, EU, and other international bodies are served in a cost-effective way. If the revision of national monitoring networks is dealt with in such a comprehensive way, modifications and even cost reductions may be identified without compromising the value and strength of the information.

The cooperation may also include data exchange, training procedures and quality assurance/quality control activities.

#### 5.1 The Convention – Working group on Effects and its International Cooperative Programmes (ICPs)

The 1997 EMEP/WMO Workshop at Aspenäs (cited in Ch. 3.3.2) recommended that "... EMEP should strengthen the observational basis by taking advantage of other air quality networks in Europe, e.g. (...) International Co-operative Programmes (ICPs)". The cooperation between the various effect programmes and EMEP has been improved and it is encouraged to make use of common methodologies, site sharing etc. A number of EMEP sites thus directly supports the effect work and vice-versa. The use of ICP observation data by EMEP can, however, still be improved.

- The ICP-Forest's Level II programme comprises bulk/wet deposition measurements and throughfall/stemflow measurements with canopy budget modelling at several hundred sites. In addition, inferential models have been applied by the Forest Intensive Monitoring Coordinating Institute (FIMCI) at many sites (see <a href="http://www.icp-forests.org/Programme.htm#FIMCI">http://www.icp-forests.org/Programme.htm#FIMCI</a>).
- Within ICP-Vegetation's monitoring programme heavy metals bulk/wet deposition rates and ozone concentrations are measured (see <u>http://icpvegetation.ceh.ac.uk/</u>).
- ICP-Integrated Monitoring performs bulk deposition and throughfall measurements at all of its sites; canopy budget models or inferential models are not regularly applied (see <a href="http://www.unece.org/env/wge/im.htm">http://www.unece.org/env/wge/im.htm</a>).
- ICP Modelling and Mapping (<u>http://www.icpmapping.org/</u>) does not have a formally installed deposition network. However, national measurement and modelling networks are applied by National Focal Centres, especially for the mapping of base cation deposition rates needed for critical loads assessment, and for mapping critical loads exceedances at a national scale.

#### 5.2 HELCOM

HELCOM is the governing body of the "Convention on the Protection of the Marine Environment of the Baltic Sea Area" - also known as the Helsinki Convention. HELCOM supports the protection of the marine environment of the Baltic Sea from all sources of pollution through intergovernmental co-operation between Denmark, Estonia, the European Community, Finland, Germany, Latvia, Lithuania, Poland, Russia and Sweden; which are all Contracting Parties to the Convention.

Atmospheric emissions and atmospheric deposition into the sea are mainly monitored under HELCOM's Pollution Load Compilation Programmes (PLC-Air). A close relationship between EMEP and HELCOM has been established which includes use of the EMEP infrastructure for monitoring data reporting as well as the results of the EMEP models and officially reported emissions. All data reported in support of HELCOM are thus available also for EMEP use.

Measurements of nitrogen compounds in air are available from about 16 sites, heavy metals is measured at 11 sites, while lindane is measured at three sites. All HELCOM sites are integrated with the EMEP network and provide a very good example on how national interest could be served through coordination of measurement activities.

### 5.3 OSPAR

The Convention for the Protection of the Marine Environment of the North-East Atlantic ("OSPAR Convention") was opened for signature at the Ministerial Meeting of the Oslo and Paris Commissions in Paris on 22 September 1992. The Convention has been signed and ratified by all of the Contracting Parties to the Oslo or Paris Conventions (Belgium, Denmark, the Commission of the European Communities, Finland, France, Germany, Iceland, Ireland, the Netherlands, Norway, Portugal, Spain, Sweden and the United Kingdom of Great Britain and Northern Ireland) and by Luxembourg and Switzerland.

OSPAR Comprehensive Atmospheric Monitoring Programme (CAMP) lists mandatory and voluntary components to be observed at background stations not more than 10 km from the coastline. These are:

	Mandatory	Voluntary
Precipitation	As, Cd, Cr, Cu, Pb, Hg, Ni, Zn, γ-HCH, NH4, NO3	PCB 28,52,101,118,138,153,180 Phenanthrene, anthracene, flouranthene, pyrene, benzo( <i>a</i> )anthracene, chrysene, benzo( <i>a</i> )pyrene, benzo( <i>ghi</i> )perylene, indeno( <i>1,2,3-cd</i> )pyrene
Airborne	NO <sub>2</sub> , HNO <sub>3</sub> , NH <sub>3</sub> , NH <sub>4</sub> , NO <sub>3</sub>	As, Cd, Cr, Cu, Pb, Hg, Ni, Zn, $\gamma$ -HCH, PCB 28,52,101,118,138,153,180, Phenanthrene, anthracene, flouranthene, pyrene, benzo( <i>a</i> )anthracene, chrysene, benzo( <i>a</i> )pyrene, benzo( <i>ghi</i> )perylene, indeno( <i>1,2,3-cd</i> )pyrene, NO

In 1999, twenty-one stations reported data, most of these report heavy metals in precipitation while a number also report nitrogen concentrations in precipitation.  $\gamma$ -HCH was reported from only three stations. There is a large overlap with EMEP sites. CAMP currently does not have a policy to put all data freely accessible on the Internet, but the data from sites reported also for EMEP will be available. NILU act as the data consultant for storing the CAMP data, but has no

responsibilities for quality assurance activities like in EMEP. All data flow is, however, according to EMEP infrastructure (file formats etc).

#### 5.4 MEDPOL

The Programme for the Assessment and Control of Pollution in the Mediterranean region (MEDPOL) was initiated in 1975 as the environmental assessment component of the Mediterranean Action Plan (MAP) and is now in Phase III. Its task is to assist Mediterranean countries in the implementation of pollution-assessment programmes (marine pollution trend monitoring, compliance monitoring and biological effects monitoring). The countries which signed the Barcelona Convention are: Albania, Algeria, Bosnia and Herzegovina, Croatia, Cyprus, Egypt, France, Greece, Israel, Italy, Lebanon, Libya, Malta, Monaco, Morocco, Slovenia, Spain, Syria, Tunisia, Turkey and the European Union.

The programme deals mainly with measurements of various pollutants in marine biota and sediments, in effluents and direct discharges to the sea as well as eutrophication parameters (N and P components) in seawater. There are also some activities to measure trace metals (as well as N and P) in precipitation and in air (on filters of high volume samplers) to assess the atmospheric input of these pollutants into the Mediterranean Sea. Some reports are available. At the present time there are several MEDPOL monitoring stations in Slovenia, Croatia, Turkey, Israel, Italy and France. Most of these countries participate in EMEP. Unfortunately no raw data from most of these stations have ever been reported to the MEDPOL office in Athens, Greece. Formats for data submission have been discussed but no information about the outcome of this is available.

#### 5.5 AMAP

AMAP's current objective is "providing reliable and sufficient information on the status of, and threats to, the Arctic environment, and providing scientific advice on actions to be taken in order to support Arctic governments in their efforts to take remedial and preventive actions relating to contaminants".

The monitoring work within AMAP is based, as far as possible, on existing national and international monitoring and research programs, aiming to harmonize these to the extent possible. Each country defines its own National Implementation Plan (NIP) to meet the AMAP monitoring objectives. Monitoring projects are carried out within each of the participating countries and across borders under bilateral and multilateral cooperations. Efforts continue to be made to harmonize existing and new programs with respect to methodologies and quality assurance.

AMAP's assessments are based to a large extent on information and results from recent (largely unpublished) monitoring and research work. Data from such activities are compiled together with routine monitoring data within AMAP Thematic Data Centres (TDCs). Data are made available from the TDCs to scientists engaged in AMAP assessments under strict conditions that protect the rights of data originators. These conditions are described in AMAP's data policy documentation. Consideration of quality assurance issues is an integral component of the AMAP monitoring and assessment process.

AMAP Thematic Data Centres have been established to meet the following objectives:

- to provide access to data from recent monitoring and research activities conducted as part of the AMAP NIPs;
- to provide a means to ensure that data are treated in a consistent manner, undergo uniform statistical analysis, etc., including application of objective quality assurance procedures;
- to begin the process of establishing a long-term archive of Arctic-relevant monitoring data, for use in future assessments of, e.g. temporal trends, etc.; and
- to meet the terms of reference of the Ministerial declarations, charging AMAP with establishing databases of sources, types, and levels of radionuclide contamination of the atmospheric, aquatic and terrestrial environments of the Arctic and northern areas.

The TDC for atmospheric contaminants is operated by NILU and employs EMEP infrastructure and data formats for its data flow.

#### 5.6 Monitoring in support of the European Community Air Quality Framework Directive

The EC aim has been to develop an overall strategy through the setting of longterm air quality objectives through its Air Quality Framework Directive (AQFD). In 1996, the Environment Council adopted Framework Directive 96/62/EC on ambient air quality assessment and management. This Directive covers the revision of previously existing legislation and the introduction of new air quality standards for previously unregulated air pollutants, setting the timetable for the development of daughter directives on a range of pollutants. A series of Daughter Directives has been introduced to control levels of certain pollutants and to monitor their concentrations in the air. The atmospheric pollutants to be considered include sulphur dioxide, nitrogen dioxide, particulate matter, lead and ozone – pollutants governed by already existing ambient air quality objectivesand benzene, carbon monoxide, poly-aromatic hydrocarbons, cadmium, arsenic, nickel and mercury.

The Daughter Directives set the numerical limit values, or target values in the case of ozone, target values for each of the identified pollutants. Besides setting air quality limit and alert thresholds, the objectives of the daughter directives are to harmonize monitoring strategies, measuring methods, calibration and quality assessment methods to arrive at comparable measurements throughout the EU and to provide for good public information. The Framework Directive, as well as its Daughter Directives, requires the assessment of the ambient air quality existing in member states on the basis of common methods and criteria.

EU has established a Community-wide procedure for the exchange of information and data on ambient air quality in the European Union by the Council Decision 97/101/EC of 27 January 1997 as amended by Commission Decision 2001/752/EC. It applies to:

- detailed information on networks and stations describing the air pollution monitoring networks and stations operating in the Member States,
- measurements of air quality obtained from stations: the exchange covers data calculated from measurements of air pollution by stations in the member states.

EU regulatory monitoring is mainly concerned with the exceedance of air quality standards for public information and warning. It is generally based on monitoring in locations where people live, though also rural sites are being established. It would be an advantage if the monitoring in rural areas were closely integrated with the EMEP work nationally. Data quality is an important issue here as the methodologies applied are less suited for meeting the EMEP objectives (e.g. precision and comparability at low concentration levels).

The European Community has also formulated strategies to combat acidification, ozone and eutrophication, notably via the Directive on national emission ceilings (NEC), which is an important directive parallel to the AQFD. The emission ceilings specified by the NEC has been based on the technical infrastructure developed under EMEP and the Convention.

The European community has initiated the programme Clean Air for Europe (CAFÉ) which aims to establishing a long-term, integrated strategy to tackle air pollution and to protect against its effects on human health and the environment. In particular it aims to:

- Develop, collect and validate scientific information on the effects of air pollution (including validation of emission inventories, air quality assessment, projections, cost-effectiveness studies and integrated assessment modelling);
- Support the implementation and review the effectiveness of existing legislation and to develop new proposals as and when necessary;
- Ensure that the requisite measures are taken at the relevant level, and to develop structural links with the relevant policy areas;
- Determine an integrated strategy (by 2004 at the latest) to include appropriate objectives and cost-effective measures. The objectives of the first programme phase are: particulate matter, tropospheric ozone, acidification, eutrophication and damage to cultural heritage;
- Disseminate to the general public the information arising from the programme.

Strong and effective links between CAFÉ and the Convention are seen as crucial in order to add real value to policy-making, to avoid duplication of effort, and to exploit synergies for resource efficiency purposes. A Technical Analysis Group has been set up to help ensure the technical analytical work is well coordinated between CAFE and the Convention. The Bureau of the Executive Body of the Convention has established a High Level Coordination Group with the European Commission's CAFÉ secretariat.

EIONET is a collaborative network of the European Environment Agency and its Member Countries, connecting National Focal Points in the EU and accession countries, European Topic Centres, National Reference Centres, and Main Component Elements. These organisations jointly provide the information that is used for making decisions for improving the state of environment in Europe and making EU policies more effective.

AirBase is the air quality information system of the EEA. It contains a database carrying information submitted by participating countries from across Europe. This information comprises air quality data for a selection of stations and a number of components, and meta-data information on air quality monitoring networks and stations. The current database contains information, which was transmitted by EIONET partner states in the framework of 'Exchange of Information' (EoI) Decisions, or as part of EuroAirnet. The AirBase information system is developed and maintained by the European Topic Centre on Air Quality and Climate Change on behalf of the European Environment Agency.

# 5.7 World Meteorological Organisation – Global Atmosphere Watch (GAW)

The purpose and long term objective of the GAW is to provide data and other information on the atmospheric chemical composition and related physical characteristics of the background atmosphere from all parts of the globe required to improve the understanding of the behaviour of the atmosphere and its interactions with the oceans and the biosphere, and to enable predictions of the future states of the Earth system. The objectives of GAW are thus in part identical with those of EMEP, and most of the GAW parameters are included in the EMEP programme. Further, many of the sites are associated with both networks.

GAW is organized in cooperation with other international programmes and a close cooperation has been established between EMEP and GAW both on an administrative level as well as on a technical level. EMEP makes use of the recommendations given by the GAW Scientific Advisory Groups (and participates with a representative in the SAG on Precipitation Chemistry), and the technical manuals are similar between the programmes. Also with respect to Quality Assurance activities cooperation is in place. The EMEP Task Force on Measurements and Modelling (TFMM) is co-chaired by the WMO Atmospheric Research and Environment Programme (AREP) Environment Division secretariat. The development of the EMEP monitoring strategy for particulate matter also was based on the joint EMEP-GAW workshop in Interlaken 1999 and later discussions at the TFMM.

Still, further efforts could be made to harmonise the two networks. In the selection of joint supersites there is a good potential for serving the needs of both networks. Regional sites operated in support of GAW should report their data to EMEP, and duplication of efforts between EMEP and GAW should be avoided in particular in relation to data submission and storage. Recently, an agreement between EMEP and GAW-World Data Centre for Aerosols (WDCA) has been made in relation to the dataflow for aerosol measurements in Europe. This includes that regional sites performing monitoring according to level 2 and level 3 (defined below) shall report their data only to EMEP/CCC using the EMEP data reporting formats.

These sites will be nominated as joint EMEP/GAW supersites, and the data will also be made available and regularly updated in the WDCA. Data from level 1 sites will only be stored in the EMEP database.

There are nine major types of measurement parameters in the GAW programme, of these five are also covered in the EMEP programme, namely ozone, precipitation chemistry, chemical and physical properties of aerosols (including optical depth), reactive gases (SO<sub>2</sub>, NO<sub>x</sub> and VOC), POPs and heavy metals (greenhouse gases, solar radiation and radionucleides are not part of the EMEP programme. Both programmes recommend meteorological parameters to be monitored.

#### 5.8 National and EU funded research projects

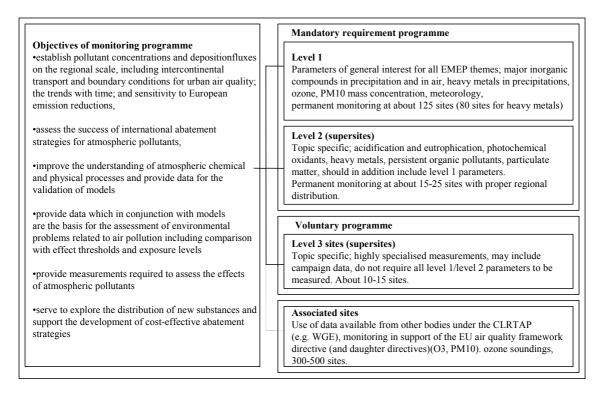
EU-DG Research (FP4 and FP5) funded research has been essential for establishing the current scientific understanding of atmospheric processes. The EUROTRAC and EUROTRAC-2 programmes have coordinated international research on issues relevant for EMEP. These activities range from controlled laboratory experiments to large-scale measurement campaigns. Without these activities the technical capabilities of EMEP would have been significantly less developed. It is essential for EMEP that research activities are continued in support of EMEP needs. EMEP shall continue to exploit the scientific results from national and international research projects. In particular short-term campaigns including advanced and comprehensive measurement programmes are important.

# 6. Definition of the level system and the monitoring requirement programme

There is no rigorous verification of the national implementation of the monitoring requirements stated in the CLRTAP, and history shows that the rather informal implementation of national monitoring within EMEP has left data coverage and data comparability at a level that is less than satisfactory in many places even after more than 20 years of operation. The new monitoring strategy points out in a rigorous manner the minimum requirements for national commitment as a party to CLRTAP.

The monitoring strategy is based on a "level" approach and introduces the term Mandatory Requirement Programme. The level approach is taken to distinguish several levels of ambition with respect to site densities and number of parameters being monitored. Levels 1 and 2 together represent the Mandatory requirement programme, while level 3 is considered voluntary. In addition, data from associated sites will be employed. A schematic overview of the levels is given in Figure 1.

The activity defined for EMEP mandatory requirement sites reflects the minimum requirements for monitoring to be performed in order to underpin the traditional objectives of EMEP. It is important to note that new priorities, such as improved spatial resolution and site specific deposition, linking of scales (local vs. regional vs. global scale), flux based monitoring, model improvement, etc., cannot be supported by the mandatory requirement sites alone and thus need complementary data.



*Figure 1: Schematic overview of the levels defined by the EMEP monitoring strategy.* 

**Level 1**; Level 1 sites will comprise a relatively large number of sites (>125) with a complete programme covering major inorganic compounds in air and precipitation, ozone and particulate matter mass. A minimum site density of 1 site per 50.000 km<sup>2</sup> is required, and a higher resolution is required in complex terrain. Level 1 is mandatory for all Parties and countries with a geographical area exceeding 25.000 km<sup>2</sup> are required to operate at least one level 1 site.

The objective of the EMEP Level 1 sites is to support the evaluation of spatial and temporal trends and is a prerequisite for the validation of models. These objectives put requirements on the network, such as good geographical coverage, continuous sampling, and long-term operation. Measurements are generally required with a temporal resolution of 24 hours, as this allows for linking observations with air mass trajectories. In addition, the level 1 sites involve all EMEP parties and ensure that there is a permanent and operational activity that links all EMEP countries and the Centres together. The monitoring thus serves as an essential element of the national participation in the EMEP work. The monitoring also directly serves additional national interest and needs.

Level 2; Level 2 sites comprise more technically or economically demanding measurements than what is realistic to expect at all EMEP sites. The minimum site density is 1 site per  $100.000 \text{ km}^2$  and varies between topics depending on region (photo-oxidants and particles have a higher site density in the south, while acidification and POPs have a higher density towards north and east). Some parameters are more process oriented compared to level 1. They also provide a basis for the analysis of spatial and temporal trends and thus serve the same general objectives as the level 1 sites.

The level 2 sites are not necessarily the same for all topics in EMEP (acidification and eutrophication, photochemical oxidants, heavy metals, persistent organic pollutants and particulate matter). Level 2 sites need in addition to comply with the monitoring requirements defined for Level 1 sites.

There will be a need for regional cooperation for providing a sufficient number of level 2 sites in order to minimise costs, and a cost sharing option is encouraged. Monitoring performed in support of the marine conventions (HELCOM, OSPAR and AMAP) will contribute significantly to level 2 sites for heavy metals and for POPs.

Level 3 sites; EMEP also needs specialised measurements generally only available from state-of-the-art monitoring sites either continuously operated or data from research experiments (see also Chapter 3.7). Level 3 sites collect research data for process studies or particularly demanding methodologies. For this reason, level 3 sites also include data from campaigns.

Level 2 and level 3 sites will be nominated as "EMEP supersites", as this would be an important motivation factor and provides appropriate recognition to the data providers. Supersites could be topic specific and would not need to cover all substances. The geographic distribution of level 2 and 3 sites should provide a good regional coverage. Supersites should be encouraged to support both EMEP as well as GAW.

#### Associated sites;

EMEP has in the past and will also in the future make use of data available from other bodies of the Convention, and from other national and international legislation. In most countries these resources are well coordinated nationally. Most measurement sites operated through HELCOM, OSPAR and AMAP are excellent examples of this, and they share the same organizational set-up with respect to technical solutions. These are thus candidates to serve as joint level 2 sites for e.g. POPs and HM. Currently, UNEP considers developing a global POP monitoring network. To be cost effective this activity must be harmonised with the efforts made by EMEP, HELCOM, AMAP and OSPAR.

EMEP will further complement its monitoring requirements by using data available from the national monitoring performed in support of European Community Air Quality Directives, which can improve the data coverage significantly for a few compounds. It should be noted that the networks operated under the air quality directives of the European Union have their main focus on populated areas to provide ambient concentrations for comparison with limit and target values for the protection of human health. However, there are also limit and target values for the protection of ecosystems and vegetation (NO<sub>x</sub>; SO<sub>2</sub>; O<sub>3</sub>), which require measurements and assessment on a regional scale. Further harmonisation of the rural sites between EMEP and the sites operated under the air quality daughter directives is important, and these data can support improvements of the spatial resolution and the linking of scales. The quality control and quality assurance of monitoring under the EU air quality directives is, however, an issue here.

#### Campaigns

EMEP has arranged a number of campaigns to provide data for parameters for which the monitoring technology is too expensive or demanding to be part of the regular programme. Examples here are the "Pilot measurements of nitrogen containing species in air" in 1993-1994, "the EC/OC-campaign" during 2002-2003, the centralised analysis of VOCs by the EMEP CCC in the 1990s and others. Such campaigns provide useful insight and information and should be considered also in the future as a necessary complement to the continuous sampling.

## 7. Specific requirements for individual topics

This chapter introduces the monitoring requirements for the individual topics of EMEP (i.e. acidification, eutrophication, photochemical oxidants, heavy metals, persistent organic pollutants and particulate matter). For each topic, an introduction to the importance of the various compounds is outlined. Further, the current status of the programme is evaluated before the monitoring requirements at the various levels are given.

#### 7.1 Acidification and Eutrophication

#### 7.1.1 Introduction;

Acidification results from long-term deposition of sulphur and nitrogen species in ecosystems. Acidification results from the vertical transport of so-called mobile anions through the soil profiles associated with a depletion of base cations and increased soil acidity. Further, surface water chemistry is affected yielding lowered pH, decreased alkalinity and high concentration of labile aluminium affecting fish stocks. Sulphate has traditionally been the main mobile anion, but in areas with high nitrogen deposition, nitrate is also contributing to the acidification. Atmospheric deposition of base cations (and associated anions) adds on the other hand buffering capacity to the soils. During the recent years it has been realised that sea salt episodes play a very important role in surface water acidification through the "sea-salt effect", by temporarily increasing concentrations of H<sup>+</sup> and Al in run-off water after ion exchange with sodium from sea-salt.

Eutrophication results from the increased deposition of nutrients (nitrogen compounds, phosphorous and base cations). Ammonia and nitrogen oxides are key components in the air pollution strategies and their role for acidification and eutrophication needs thorough evaluation. Fossil fuel combustion, mineral fertilizers and livestock manures all provide major sources of fixed reactive nitrogen (N). This leads to a cascade of effects as the N is transported and transformed through the environment. Excess nitrogen deposition also may increase the nitrate concentrations in surface water, thus also directly contributing to acidification.

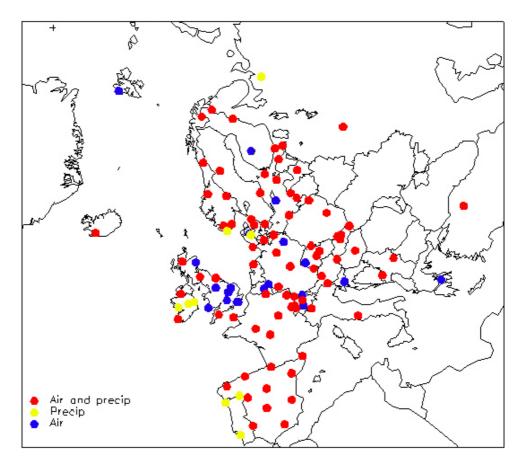
The aspects of acidification and eutrophication are closely linked in terms of atmospheric chemistry and deposition as well as effects in the ecosystems. Acidifying and eutrophying compounds affect biodiversity. Also when it comes to developing abatement policies, acidifying and eutrophying compounds are closely related, but still these are also dealt with in very different frameworks, e.g. the EU

Nitrates Directive dealing with eutrophication of ground and fresh waters from agriculture, the UNECE Convention on Long-Range Transboundary Air Pollution (CLRTAP) and the EU National Emissions Ceilings (NEC) Directive dealing with regional air pollution (acidification, eutrophication and photochemical oxidants), and the Framework Convention on Climate Change (FCCC) dealing with emissions of nitrous oxide as a greenhouse gas.

#### 7.1.2 Current situation

Beginning on 1<sup>st</sup> January 1978, the first measurement phase of EMEP initiated monitoring of acidity and sulphur compounds in air and precipitation across 14 countries and at 46 stations. Since then, a total of about 150 stations in 35 countries have produced data at various times as part of the acidification/ eutrophication network. The acidification network itself has extended beyond sulphur to include oxidised and reduced nitrogen compounds in air and precipitation, as well as base cations and sea-salts. Currently, the network consists of about 100 sites measuring air and precipitation chemistry, but of these only about 60 sites measure nitrogen compounds in air and very few measure base cations and sea-salt concentrations in air. An evaluation of the geographical coverage can be found in Chapter 4.1.

Figure 2 shows the location of sites performing air and precipitation chemistry measurements (main components) (in 2000).



*Figure 2:* The location of sites measuring performing air and precipitation chemistry measurements (main components) (in 2000).

#### 7.1.3 Monitoring requirements

Precipitation sampling and analysis is still important as it provides key information on removal mechanisms and deposition rates over large regions. Long-term data series of precipitation chemistry are fundamental for assessing changes in emission rates and associated changes in deposition amounts. Wet deposition represents the major deposition flux in large areas of Europe, and it can fairly reliably be quantified (as compared to dry deposition). Still there are large local variations in precipitation amounts which current models are not able to resolve. Representative regional sites may thus be combined with observations from more dense meteorological networks to provide wet deposition estimates for critical load exceedance calculations.

Precipitation chemistry data are valuable because of their low cost and their reproducibility. It is also recognised as important that precipitation data integrate vertically, so that the spatial representativeness of precipitation chemistry measurements may be used to infer ambient air concentration gradients, in particular for highly reactive species. This is not the case for ammonium in precipitation, which may be strongly influenced by local ammonia concentrations. Precipitation scavenging is the most important sink for fine particulate matter and measurements are thus required for developing and validating particulate matter models.

In order for the measurements to be useful for validation of models of long-range transport and deposition of air pollutants, the site for precipitation collection should be chosen, and the collection of rain and snow for analyses should be made in such a way that the concentrations are representative of rainfall composition over a larger area. These elements are thoroughly discussed in the EMEP manual (EMEP/CCC, 1996). Parameters to be measured in precipitation are all major inorganic ions including sulphate, nitrate, ammonium, chloride, sodium, magnesium, potassium, calcium as well as precipitation amount, pH and conductivity.

Monitoring trends in wet deposition amounts for assessments of effects on ecosystems and materials can be based on weekly samples. On the other hand, to meet the requirements to support model development and validation as well as to assess the emission changes (including evaluation of the importance of legislation and for compliance), measurements need to be taken on a daily basis. This also improves the data quality in general, and it reduces the influence from dry deposition to the precipitation samplers. Daily sampling also allows for assessing the air mass origin and thus the importance of emission changes in different regions (including the effect of legislation). Also air samples are required with a daily time resolution. Having identical time resolution for air concentration measurements as for precipitation chemistry measurements allows for studying scavenging efficiencies, which is fundamental for improving models as well as for assessing potential contamination of the samples etc.

Gaseous and particulate nitrogen species need to be measured separately at more sites to assess the transport distance and ecosystem effect of various nitrogen species. The gaseous and particulate fractions of NH<sub>3</sub>/NH<sub>4</sub> and HNO<sub>3</sub>/NO<sub>3</sub> should be reported separately from sites employing the reference method (realising that

the gas/particle distribution is subject to artefact). This should be combined with low cost methods for the gaseous fraction with a monthly time resolution using the DELTA method (see Chapter 4.2.1). At level 2 sites the gas-particle distribution should also be measured using denuder/filter pack or other artefact free methods (e.g. using AMANDA and SJAC combinations, as developed by ECN, The Netherlands).

Low cost methods for NH<sub>3</sub> (e.g. the DELTA method or passive samplers) should be employed with high spatial resolution in regions with significant local emission rates (Northern France, The Netherlands, UK, Ireland, Denmark and others) to provide information on their importance for trends of other parameters (see also Chapter 3.4). Such networks may need a significantly higher site density compared with level 1 or level 2 sites and could be considered as a level 3 activity. There is a need for a limited number of sites also monitoring dry deposition fluxes of sulphur and nitrogen species. These will assist in assessing non-linear effects in deposition fluxes compared to changes in ambient concentrations (see also Chapter 3.4). Measurements of NH<sub>3</sub> and HNO<sub>3</sub> concentrations on an hourly basis could be initiated at a limited number of level 3 sites. Because of the relatively short lifetime of the highly reactive nitrogen compounds, measuring these species is particularly challenging. This is especially the case for gaseous nitric acid, which is even more rapidly dry-deposited than ammonia.

It should be noted that passive samplers can be useful as a complement to active sampling methods in EMEP. Such methods should only be implemented where their accuracy is continuously monitored alongside an active reference. Given this and the fact that passive samplers do not require electricity, they may be used to investigate local variability and to establish site representativity.

In many areas, atmospheric deposition of anthropogenic base cations is comparable to the release from weathering of soil minerals. The base cation deposition is therefore important for determining the exceedance of critical loads. There is, however, a lack of measurements of aerosol concentrations of base cations, which are currently derived solely from calculations based on wet deposition, which is highly uncertain. Base cations and sea salts also contribute to aerosol mass and are important for chemical mass closure. Sea salt concentrations (Na, Mg (Cl)) allows for estimating the fraction of aerosol SO<sub>4</sub>, Ca and K originating from sea spray. The lack of information of the sea-salt contribution to EMEP data has been commented in several papers where data have been used for validation of global scale models (e.g. Chin et al., 2000).

#### 7.2 Photochemical oxidants

#### 7.2.1 Introduction

Ozone is a natural constituent of the atmosphere and plays a vital role in many atmospheric processes. However, man-made emissions of volatile organic compounds and nitrogen oxides have increased the photochemical formation of ozone in the troposphere. Episodes of high concentrations of ground-level ozone occur over most parts of the continent every summer. During these episodes the ozone concentrations can reach values above ambient air quality standards over large regions and lead to adverse effects for human health and vegetation. Historical records of ozone measurements in Europe and North America indicate that in the last part of the nineteenth century the values were only about half of the average surface ozone concentrations measured in the same regions during the last 10-15 years.

In defining the harmful effects of ozone exposure to plants, attention must be given to the physiological response to ozone. Ozone is generally taken up through the stomata, and reacts with a number of enzymes and antioxidants. Several studies have shown that plants respond by reduced carbon dioxide uptake, and other symptoms of damage to the respiration system. The response is highly correlated with the ozone exposure above a certain threshold. The effect varies between plant species, cultivars, and phenological development.

A revision of the critical levels for ozone is presently being finalised. A level 2 approach taking into account soil-moisture, ambient temperature etc to estimate the harmful part of the ozone that is deposited to the vegetation is being developed as part of this work.

#### 7.2.2 Current situation and further needs

The ozone monitoring was established in 1985 in the fourth phase of EMEP but due to financial reasons a systematic collection and checking of ozone data did not start until 1987. Presently, the ozone measurement network consisting of about 100 stations is insufficient in several areas of the continent, particularly in the south and east, but at the same time there are totally about thousand sites in operation in Europe. Improved cooperation and exchange with other bodies and programmes are needed.

The EMEP objectives set strict requirements in terms of instrument precision and quality control for ozone monitoring. A recent survey of the applied QA procedures for ozone monitoring showed that there are still significant gaps compared with the recommendations in the Manual. Long-term trend evaluations of ozone set particularly strong requirements on the precision of ozone monitoring, and stronger focus on the QA requirements is thus needed. A selection of sites should be designated to detect long-term changes in ozone concentrations.

Future revisions in the critical levels for ozone necessitate direct ozone flux measurements at certain experimental locations. This will be essential to improve the estimates of harmful effects on vegetation and to support the ongoing modelling work on ozone deposition.

Support to the model development is an important objective of the photo-oxidant monitoring programme. This requires monitoring of other constituents than those required for effect assessment, as e.g. NO, RO<sub>2</sub>, H<sub>2</sub>O<sub>2</sub>, PAN, carbonyls etc, and this is one reason for establishing super sites. Process studies also require monitoring of the vertical distribution of ozone, e.g. by vertical soundings. Data and results from EUROTRAC and EU/DG XII funded research projects are important sources of such information.

The EMEP VOC monitoring programme was initiated at the EMEP Workshop on Measurements of Hydrocarbons/VOC in Lindau, 1989 and the monitoring started in 1992-1993. Due to the control of vehicle exhaust and evaporative emissions, concentrations of the light VOCs have decreased quickly across much of Northwestern Europe in line with the inventories. In terms of compliance monitoring, the current VOC monitoring has worked well and has been cost effective.

In the future, control action under the EU Solvents Directives, Decorative Paints Directive and under the UNECE Gothenburg Protocol will increasingly tackle stationary sources of VOCs. In the speciated emission inventories for stationary VOC sources, much of the mass emitted is contained in  $C_6$ - $C_{12}$  species, so the overlap is poor with the current EMEP VOC monitoring. Thus verification of the VOC emissions from stationary sources is an urgent priority.

#### 7.2.3 Monitoring requirements

A network of surface background monitoring sites of ozone is the backbone of the photo-oxidant monitoring. It is essential to improve the geographical coverage of the present network, particularly to the south and east. Vertical ozone soundings at additional super sites will make an important complement to the surface monitoring. Direct flux monitoring of ozone to the surface is needed at appropriate experimental sites (super sites).

Continuation and expansion of the VOC monitoring network is needed for compliance monitoring and model support. As the small VOC monitoring network has changed several times, only very few sites have long-term reliable VOC measurements, and it is crucial to continue the VOC activity at these sites. A site at the main inflow boundary of Europe (such as Mace Head, Ireland) would be highly valuable. VOC sites at rural areas in Southern Europe (Spain, Italy, Greece, Balkan) are particularly needed, not only to assess geographical variations in the general concentration levels, but also in the VOC *speciation*, i.e. the mix of individual species, as this is likely to differ from Northern Europe due to a higher influence of biogenic emissions (terpenes, isoprene).

The VOC monitoring consists of measurements of light hydrocarbons by 10-20 min grab samples in canisters and measurements of carbonyls by 8 h sampling in DNPH tubes. The measurements are carried out on the same days, normally twice a week through the year. For the stations with sufficiently long time series of VOC it is recommended to continue this practice. For compliance monitoring purposes it is essential to maintain homogeneous long-term monitoring data of VOC without breaks in the procedures.

New sites should have the possibility to choose another monitoring practice when appropriate. Data from a continuous monitor (CH05 Rigi) is already being reported to EMEP. Furthermore, equipment for extended sampling time (to e.g. 8 hours) of hydrocarbons in canisters is available. New sites should also have the possibility for other sampling periods during the year. Dense monitoring of hydrocarbons during winter and of carbonyls during summer is an option with several advantages compared to the present practice.

The QA activity of the VOCs should be emphasised and campaigns with parallel sampling and analyses are recommended. An intercalibration exercise for carbonyls is also recommended.

VOCs are today measured in many (sub)urban areas as part of national monitoring of air quality as well as within campaign research projects in EU DGXII, and a closer link with these bodies is recommended. The EC Daughter Directive 2002/3/EC for ozone requires VOCs and other precursors to be measured at least at one site (suburban or rural) in each country. In many cases it will be advantageous if this activity rather was performed at one of the national EMEP sites as the formation of peroxiradicals from emitted VOCs occurs mainly on the regional scale. This would provide an improved basis for understanding the ozone formation and its changes with time.

#### • VOC speciation

At the Lindau workshop in 1989, a list of required and desirable VOC components for reporting was defined and is given below. All the required components are actually reported today (with the exception of trimethylbenzenes), and the desirable compounds are partly reported.

In addition to the species listed in Table 1, increased attention on measurements of solvent species in the  $C_6$ - $C_{12}$  range is an urgent priority. This requires method development as the traditional steel canisters sampling procedure is not well suited for these heavier hydrocarbons.

	required	desirable	
Alkanes	ethane	hexane	
	propane	branched hexanes	
	i-butane	heptane	
	n-butane	branched heptanes	
	i-pentane	octane	
	n-pentane		
Alkenes	ethene	butenes	
	propene	pentenes	
	isoprene		
Alkynes	acetylene		
Aromatics	benzene	styrene	
	toluene	propylbenzenes	
	o-xylene	ethyltoluenes	
	m,p-xylene		
	ethylbenzene		
	trimethylbenzenes		
Aldehydes	formaldehyde	propionaldehyde	
-	acetaldehyde	,	
Ketones	acetone	methylethylketone	
		methylvinylketone	

Table 1:	Original list of volatile organic compounds classified as "required"
	or "desirable" to measure within the EMEP programme as defined at
	the initiating EMEP Workshop in Lindau, 1989.

#### 7.3 Heavy metals

#### 7.3.1 Introduction

On a global scale and even more so within Europe, anthropogenic releases of the priority metals considerably outweigh natural emissions. These metals are all toxic to humans/biota. Metal and metalloid compounds can be incorporated to biota in the ecosystems, either directly via deposition, or indirectly via uptake from soil. Effects caused by the exposure of organisms to heavy metal deposition may be related either to current deposition rates or to accumulated amounts in the ecosystems. The knowledge of possible effects of some of the HMs like arsenic, cadmium and nickel, on ecosystems is still rather limited. The HM species occurring in ambient air, but also their physico-chemical properties, have not been properly characterized so far by measurements. Consequently, the assessment of effects is impaired by considerable uncertainties, and more information on speciation is needed.

During the seventh phase of EMEP (EB.AIR/GE.1/1998/8) it was recommended that the Convention should concentrate on eight priority heavy metal elements: lead (Pb), mercury (Hg), cadmium (Cd), chromium (Cr), nickel (Ni), zinc (Zn), copper (Cu) and arsenic (As). Particular attention should be paid to the first three elements, which are the target elements under the 1998 Aarhus Protocol on Heavy Metals.

Lead is enriched in the fine particle mode about  $2 \mu m$ , and can penetrate deeply into the respiratory system. Lead deposited from the atmosphere can enter ecosystems/food chains. Additives to petrol containing lead are still the main anthropogenic source.

Cadmium is enriched in the fine particle mode about or below  $1 \mu m$  and, consequently, can penetrate deeply into the respiratory system and have long residence times in the atmosphere. Cadmium is carcinogenic and act as systemic pollutants and the transfer into the food chain is of particular relevance. Non-ferrous metal industry is the main source.

Unlike the other priority metals, mercury is in the atmosphere mainly found in its elemental form, which is relatively un-reactive. However, close to anthropogenic sources the main species can be so-called reactive gaseous mercury and/or particulate mercury which are much more reactive and have a considerably higher deposition rate than elemental mercury. These species may also be found in the Arctic during special occasions. Methyl mercury is bio-accumulative. Anthropogenic sources are combustion of fossil fuels. Some of the processes involving elemental mercury in the atmosphere can be very quick (for example, Arctic depletion) and the temporal resolution should be sufficient to examine the rate of such processes (e.g. by applying modern TGM analysers).

Arsenic is enriched in the fine particle mode about or below  $1 \mu m$  and, consequently, can penetrate deeply into the respiratory system and have long residence times in the atmosphere. Methylation of inorganic arsenic is known to occur in water and soil and minor amounts of methylated species might be present in air as well. Arsenics transfer into the food chain is of particular relevance.

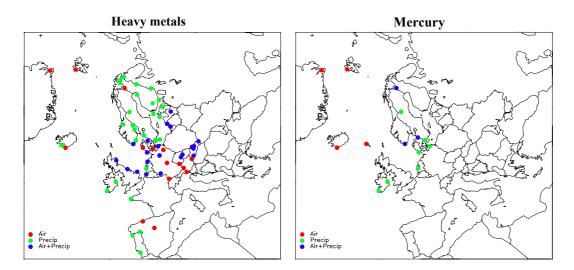
Arsenic is carcinogenic. Anthropogenic sources are non-ferrous-industry and partly combustion of fossil fuels.

In contrast to arsenic, lead and cadmium, up to 30% of the total nickel compounds may be found in the coarse mode. Several nickel compounds are classified as human carcinogens in the EU system. Anthropogenic sources are combustion of fossil fuels.

#### 7.3.2 Current situation and further needs

From 1999, heavy metals have been part of the EMEP programme. The recommendations and main conclusions for future monitoring are based on the EMEP and WMO-GAW workshops in Durham, Beekbergen, Moscow and Aspenäs, and the two first meetings in the task force on measurements and modelling (TFMM).

For 2001 the measurement network of heavy metals is seen in Figure 3. There are 67 heavy metal sites in Europe connected to either the EMEP, OSPAR (CAMP) or HELCOM monitoring programme. 22 of these measure both heavy metals in air and precipitation. For mercury the site density is lower, and only 15 sites in Europe measure at least one form of mercury.



*Figure 3: Measurements of heavy metals in air and precipitation at EMEP, CAMP and/or HELCOM sites in 2001.* 

The spatial distribution of sites measuring heavy metals is not satisfactory, as seen in Figure 3, since the sites are mainly distributed in Northern and Central Europe. A better site coverage would greatly improve the ability to validate the modelling results.

It is important to have information of HM in air and precipitation at the same time to improve model parameterisation of HM scavenging with precipitation. It is believed that 20 stations with a full programme covering both air- and precipitation chemistry would be enough to provide modelling deposition fields within a factor-2 uncertainty over the whole domain, assuming a good geographic distribution. In addition more sites with wet deposition measurements are needed for model validation and for more accurate deposition estimates.

Furthermore, there should be additional information from "background" areas on the borders of the domain. Recently experience has shown that in unpolluted areas (for example Northern Scandinavia) external sources of HMs can provide significant contributions to pollution levels. For the domain as a whole, external sources can contribute as much as internal anthropogenic sources in Europe. E.g. the region of South-Eastern Asia is going through a period of booming industrialisation. Already now the region makes the highest contribution to worldwide mercury emissions. So, to improve reliability of modelling results on a global scale it is important to get monitoring information from the whole region. In this respect it might be an idea to expand the EMEP database to also cover the Northern hemisphere. This should be further discussed also in cooperation with UNEP. Today the information on HM concentrations in the atmosphere and in the other geospheres is not coordinated. Creation of a unified database could give an opportunity to assess pollution levels and their trends more completely and easily.

The Arctic is a region of special interest and additional stations are needed. It is especially important for mercury, which can be accumulated in vulnerable Arctic ecosystems. Three forms of mercury should be measured. Until now only total gaseous mercury (TGM) is measured on a routine basis. However, it is widely recognised now that reactive gaseous mercury (RGM) and total particulate mercury (TPM) contribute significantly to pollution levels in industrialised areas.

In addition to the analysis of air and precipitation samples, there should be a closer cooperation between programmes monitoring HM concentrations in different environmental compartments (sea water, soils, etc.). Development of multi-compartment models for mercury requires knowledge of mercury contents in different media: fresh water, seawater, bottom sediments, and soils.

Further steps need to be taken to evaluate the overall data quality of the heavy metal measurements. Until now analytical methods are being checked in the annual laboratory comparisons. Uncertainties contributed by sampling, shipping, sample processing also need to be assessed.

#### 7.3.3 Monitoring requirements

#### Level 1 sites

A minimum activity comprising the measurement of heavy metals in precipitation is required at a relatively large number of sites, but it is probably not realistic to include these measurements at all level 1 sites. Thus the recommendation is to sample heavy metals at an intermediate number of sites compared to Core sites level 1 and Core sites level 2, about 80 sites At these sites there should be weekly sampling of at least the two first-priority compounds Pb and Cd, using a wet only collector.

#### Level 2 sites

Weekly sampling of the first and second priority compounds Pb, Cd, As, Ni, Cr, Zn and Cu in both air and precipitation is required using a  $PM_{10}$  sampler and wet only collector, respectively.

Monthly sampling of mercury in precipitation is required using a collector made of borosilicate class or halocarbon. Also weekly measurements of total gaseous mercury (TGM) is required with 24 hours sampling period using a gold trap, or if possible continuous measurements with an automatic TGM monitor.

#### Level 3 sites

At level 3 sites, measurement campaigns should be carried out to meet the requirements for improving the understanding of mechanisms in order to support further model development. These measurements could e.g. include high time resolution measurements of special compounds, defined fraction of compounds and measurements that are not yet well enough developed and tested for routine monitoring (e.g. reactive gaseous mercury, chromium species). Elemental mercury should be measured with a time resolution adequate for detecting any depletion processes wherever relevant (Arctic). In addition, mercury measurements in environments other than the atmosphere and fluxes between the compartments (e.g. air, oceans, soil, vegetation) are necessary to correct evaluation of contamination levels and complete descriptions of source-receptor relationships.

#### 7.4 Persistent Organic Pollutants

#### 7.4.1 Introduction

Persistent organic pollutants (POPs) are organic compounds of anthropogenic origin, which resist photolytic, biological and chemical degradation, leading to bioaccumulation in the food chain. They can be transported over long distances in the atmosphere resulting in a widespread distribution across the earth, including regions where they have never been emitted. Due to their toxic characteristics they pose a threat to humans and the environment. Therefore, in recent years the international community has called for urgent global actions to identify their possible risk to human health and the environment and to reduce and eliminate the release of POPs (EB.AIR/WG.1/2002/14).

DDT, DDE and DDD are semi-volatile and have a potential for long-range transboundary air pollution (LRTAP). They are lipophilic and have a significant potential for biomagnification. The breakdown products of DDT, DDD and DDE, are present virtually everywhere in the environment and are more persistent than the parent compound.

Chlordanes are semi-volatile and have a potential for long-range transboundary air pollution (LRTAP). They are lipophilic and have a significant potential for biomagnification.

 $\alpha$ - and  $\gamma$ -HCH are more soluble in water than other typical POPs and have a lesser bioconcentration potential.  $\gamma$ -HCH is very prevalent in marine environment and soils, but low levels are found in biota. A minor constituent of Lindane is  $\beta$ -HCH, this isomer has reduced water solubility and hence has a higher bioconcentration potential than  $\gamma$ -HCH. HCH residues are found in water and air samples all over the world. Often higher concentrations are found in the waters of northern regions than in major source regions in the mid latitudes. The presence of HCH in environments far away from the sources is considered to be due to LRTAP. The presence of large quantities of  $\gamma$ -HCH in the oceans and lakes introduces a delay in the response of atmospheric concentrations to the decreases in emissions.

Hexachlorobenzene is very persistent in the environment due to its chemical stability and resistance to biodegradation. Long-range transport plays a significant role as a mean of redistribution of HCB throughout the environment via atmospheric or oceanic systems. If released to the atmosphere, hexachlorobenzene exists primarily in the vapour phase and degradation is extremely slow.

PCDD/Fs are characterised by their semi-volatility and resistance to degradation. They are very lipophilic, highly persistent and have LRTAP potential. They intensively adsorb onto particles in air, soil and sediment and accumulate in fatcontaining tissues. The strong adsorption causes their mobility in soil and sediments to be negligible. Air is probably the most significant compartment for environmental distribution. They have the ability to bio-concentrate and biomagnify, thereby potentially achieving toxicologically relevant concentrations.

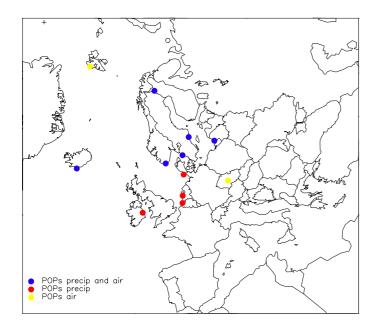
PCBs are characterised by their semi-volatility and resistance to degradation. They are very lipophilic, highly persistent and have LRTAP potential. They intensively adsorb onto particles in air, soil and sediment and accumulate in fatcontaining tissues. The strong adsorption causes their mobility in soil and sediments to be negligible. Air is probably the most significant compartment for environmental distribution. They have the ability to bio-concentrate and biomagnify, thereby potentially achieving toxicologically relevant concentrations.

PAHs are present in the atmosphere in the gaseous phase or as sorbed to particulates with relatively low degradation rates. Fine particles can remain airborne for a few days or longer and can be transported over long distances, therefore a part of PAHs have LRTAP potential. Air is probably the most significant compartment for environmental distribution. The accumulation of PAHs in the soil is not significant. Bioaccumulation is limited and biomagnification has not been observed because most organisms have a high biotransformation potential for PAHs.

#### 7.4.2 Current situation and further needs

POP sampling sites in Europe are few and mostly found around the North- and Baltic Seas, in the Arctic and in northern Finland (Figure 4). In 2001 there were 6 sites measuring POPs in both compartments, and altogether there were 12 measurement sites in operation.

A major limitation in the development of an adequate monitoring programme for POPs is the need for advanced instrumentation, the associated costs and need for resources. To increase the number of sites measuring POPs, a central laboratory may carry out all the chemical analyses in the beginning to ensure appropriate procedures and quality as the national laboratories acquire sufficient skills to continue on their own.



*Figure 4: Measurements of POPs in air and precipitation at EMEP, CAMP and/or HELCOM sites in 2001.* 

As a minimum it is recommended to monitor PAHs, PCBs, HCB, chlordane,  $\gamma$ -HCH (lindane),  $\alpha$ -HCH and DDT. There are numerous different congeners for the different group of compounds, and the various monitoring programmes and organisations may have different priorities of species to be measured. For EMEP the recommendations are to a large extent harmonised with HELCOM, AMAP and OSPAR.

For PCB it is recommended to measure the 7 congeners "PCB7": 28, 52, 101, 118, 138, 153, 180. The PAH of highest priorities are benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, indeno(1,2,3-cd)pyrene, benzo(a)anthraxcene, benzo(ghi)perylene, and fluoranthene where the first four are indicators for emission inventories according to the UN/ECE POPs Protocol. In addition phenanthrene, anthracene, pyrene and chrycene are important PAHs and are part of the voluntary list of compounds in the CAMP programme. For chlordanes the cis- and trans chlordane and nonachlor are the most important analytes; and for DDT it is the 4,4'-DDE, -DDE, -DDT that are the most essential according to UNEP.

Congener specific measurements of dioxins and furans (PCDDs, PCDFs) should be included as a level 3 activity.

The aims of monitoring POPs as formulated by the various international conventions and programmes (CLRTAP, HELCOM, OSPAR, AMAP and UNEP) are to a large extent related. In particular, the following tasks are to be accomplished in the framework of monitoring activities:

- Revealing pathways of POP transport from sources to remote regions;
- Evaluation of level and spatial distribution of POP contamination in various environmental compartments;
- Establishing long-term trends of POP environmental contamination;
- Evaluation of POP long-range transport and source-receptor relationships;
- Evaluation of adverse effects of POP contamination on human health and the environment;
- Evaluation of media response to different emission scenarios;
- Evaluation of new substances to be included into international agreements.

For obtaining a more complete set of information on POP environmental contamination a complex measurement/modelling approach to monitoring is used under EMEP.

The requirements for measurement data from the viewpoint of modelling are formulated below on the basis of discussions at numerous international conferences and workshops. In particular these requirements were clearly summarised in conclusions and recommendations of the Geneva Workshop on modelling of atmospheric transport and deposition of persistent organic pollutants and heavy metals held in Geneva, Switzerland in November 1999.

1. Data of POP measurements from different parts of the globe (or at least of the Northern Hemisphere) are very important for global/hemispheric modelling of POP transport and accumulation in the environment.

The importance of measuring POP concentrations in the environment at the hemispheric scale is conditioned by high ability of some POPs to long-range transport all over the globe. Therefore for such POPs modelling is to be performed by hemispheric models. To validate these models it is necessary to have measurements in different regions of the Northern Hemisphere. Particular attention is to be paid to such regions as the Arctic and South-Eastern Asia. Measurements of POP concentrations in various environmental compartments in the Arctic are of special interest for evaluation of the impact of POP contamination to the vulnerable Arctic ecosystems. Asian regions give great contribution to the overall POP emissions, and POP measurements in these regions will raise the reliability of modelling results on a global scale.

2. It is important to have measurements in environmental compartments other than the atmosphere (soil, seawater, vegetation). Possibly, measurements in environmental media other than air and precipitation may be included into the measurement programme in the future.

The necessity of measuring POP concentrations in different environmental compartments is conditioned by the fact that these substances accumulated in these compartments during long time periods, can be re-emitted into the atmosphere and undergo further atmospheric transport (grasshopper effect). Thus POP transport modelling is to be performed by multicompartment

models. To validate such models it is necessary to have measurements in additional environmental compartments as soil, seawater and vegetation.

Apart from model validation purposes, pollution levels in soil, seawater and vegetation are important since these levels determine accumulation of POPs along food chains and, as a consequence, are considered as essential input information for exposure and risk assessment studies.

As mentioned above, the exchange processes play an important role in longrange transport of some POPs. To refine model descriptions of exchange processes and to validate the performance of corresponding model blocks, simultaneous measurements in pairs of media (atmosphere/soil, atmosphere/ seawater, atmosphere/vegetation) are needed. Simultaneous measurements of concentrations in the atmosphere and precipitation are important for correct model description of wet scavenging process.

3. Measurements of atmospheric concentrations are to be done separately for particulate and gaseous phases of a pollutant.

The transport potential of persistent organic pollutants in particulate and gaseous phases are quite different, due to differences in processes that contributes to their dispersion and fate in the environment. For instance, particulate phase POPs (e.g. more chlorinated PCBs) are more easily scavenged from the atmosphere via dry and wet deposition processes. For some of the more volatile POPs (e.g. HCHs) gaseous exchange processes between atmosphere and underlying surfaces are of particular importance. Therefore, discrimination between these two phases is an important parameter in the description of long-range transport, accumulation and fate of POPs in the environment.

- 4. Congener-specific measurements are important for complex chemical mixtures such as PCBs, PCDD/Fs, PAHs, etc. because of the substantial variation in physical-chemical properties, which affect their long-range transport potential. In addition, environmental lifetimes and toxicity are usually highly variable within groups of chemical mixtures. For example, among the 210 possible PCDD/F congeners, only 17 possess significant toxic properties, according to NATO toxicity equivalent system. Further, toxicity equivalents of different congeners are different. Besides, in the course of their transport, complex chemical mixtures can change their congener composition (fractionation). To assess the overall toxicity of complex chemical mixtures and to evaluate source-receptor relationships of such pollutants in the environment, it is therefore necessary to monitor the congener composition and its variability in space and time.
- 5. Weekly sampling is considered to be sufficient for EMEP objectives, considering the large costs associated with a shorter time resolution. The information on emissions that is reported is on the level of annual totals. Besides, in some cases information on seasonal variations (on the level of monthly data) is available. As a consequence, the results of long-range transport models are considered reliable on the level of annual/monthly averages. To obtain similar averages from measurement data weekly measure-

ments seem to be the best compromise between modellers' demands and feasibility.

6. The necessity of measurements in different regions of Europe is conditioned by behavioural peculiarities of POPs in the environment within different climatic zones. For most other pollutants, concentrations tend to decrease from source areas, due to dispersion, degradation and dilution. However, for some POPs, concentrations may be surprisingly high, due to effects such as prolonged persistence in cold climates and cold condensation. Because of the strong impact of temperature on environmental phase partitioning and environmental lifetimes, it is therefore necessary to extend the spatial coverage of POP monitoring to better facilitate model validation.

#### "New substances"

The selection of POPs for the different monitoring levels given above is based on the knowledge on emissions, transport potential and effects, which was available at that time. However, recent research results are pointing out more compounds as possible persistent organic pollutants. Such "new" POPs are some brominated flame retardants (BFRs), short and medium chain chlorinated parafins (SCCPs and MCCPs) or very recently perfluorinated alkylsulphonates (PFAS).

Brominated flame retardants (BFRs) are used in a lot of different technical applications to prevent the products from catching fire. There are several different brominated compounds, which are used for these applications. However, there is a growing interest both from environmental scientist and regulating authorities towards the following three major compound groups: Polybrominated diphenylethers (PBDE), tetrabromo bisphenol A (TBBPA), and hexabromo cyclododecane (HBCD). These three compound groups are the most applied BFRs and can be found in different environmental and human samples with often increasing concentrations. PBDE congeners have physical and chemical properties (lipophilic, low vapour pressure, high  $\log K_{ow}$ ), which make them generally resistant to environmental degradation, susceptible to long range transport processes and able to bioaccumulate. There is also an increasing evidence for different toxic effects of these compounds. These concerns have resulted in limitations and regulations for their use and disposal. In addition to the concern on the direct effects of the unchanged compounds, the BFRs can also during incineration undergo chemical reactions leading to the formation of brominated dioxins and furans (PBDD/Fs). There are very few data available on environmental transport and distribution. PBDD/Fs are more readily photochemically degraded compared to PCDD/Fs. Generally, the physicochemical properties of PBDD/Fs suggest similarities to PCDD/Fs. Therefore they would be expected to accumulate in carbon- and/or fat-rich compartments.

SCCPs are complex mixtures, which vary in chain lengths and in the degree of chlorination. The vapour pressure values, Henry's law constants and atmospheric half-live values are in the same range as for other persistent organic pollutants and imply a significant potential for long range atmospheric transport. SCCPs have been detected in Arctic air, biota and lake sediments, and in the water column around Bermuda Islands despite the absence of significant sources of SCCPs in these regions, which suggests that these residues are present due to long-range

atmospheric transport. SCCPs are clearly fulfilling the criteria for bioconcentration and there is some evidence for biomagnification.

### 7.4.3 Monitoring requirements for POPs

Level 1 sites; Due to the resource requirements associated with measurements of POPs such measurements are not recommended to be included at the level 1 sites.

Level 2 sites; Active sampling in both air and precipitation at 10-15 sites in Scandinavia/Baltic States, Northern Europe Atlantic region, Continental Europe, Mediterranean region and in the Southern Europe Atlantic region is required. It is further recommended to supplement with passive samplers at additional sites.

The weekly precipitation sampling should be done using a collector made of borosilicate class. For collecting the air components it is recommended to have 24 to 48 hours high volume sampling on a weekly (or biweekly) sampling interval

Level 3 sites; At level 3 sites, measurement campaigns should be carried out to meet the requirements for improving the understanding of mechanisms in order to support further model development. These measurements include high time resolution measurements of special compounds, and congener specific measurements. Furthermore, POP measurements in compartments other than the atmosphere and fluxes between the compartments (e.g. air, oceans, soil, vegetation) are necessary to correct evaluation of contamination levels and complete descriptions of source-receptor relationships. Also analysis for "new substances" is highly desirable at level 3 sites.

#### 7.5 Particulate matter

#### 7.5.1 Introduction

Particulate matter may cause a variety of negative effect on our environment including impacts on human health, reduced visibility, it affects cloud formation and cloud properties and thus the radiative properties of the atmosphere, it contributes to the deposition of chemical compounds to ecosystems, and it contributes to soiling of materials (Lazaridis et al., 1999).

The chemical composition and physical properties of aerosols contain information about the particles' sources. These properties also determine the aerosols' interaction with the human respiratory system, but also the transport, transformation, and deposition of particles. Aerosols can be classified according to their origin as either primary or secondary, or as natural or anthropogenic. Typical precursor gases are sulphur dioxide, nitrogen oxides, ammonia, and volatile organic compounds (VOC). The precursors can be of natural or anthropogenic origin, where the latter are usually predominant in Europe. The precursor gases can form particles either by homogeneous nucleation, by condensation onto existing particles, or by chemical and photochemical reactions. Primary natural aerosols originate from sea spray (salt aerosols), soil resuspension by wind (e.g. Saharan dust), or volcano emissions. The most important anthropogenic sources of aerosols or precursor gases of secondary particles are road transport, combustion sources, fossil fuel power plants, and agriculture. Particulate matter is not a single pollutant and its mass includes a mixture of many pollutants in a complex multiphase system (Noone et al., 2003). The multiphase system of aerosols and clouds is extremely challenging one to sample properly. In contrast to gases, characterisation of aerosols requires a number of different measurements to quantify their thermodynamic state. Aerosol particles vary in size from a few nanometers for particles just produced from the gas phase, to tens of micrometers for coarse material. The chemical composition of the aerosol varies as a function of size, as does the state of mixing of the particles (the degree to which the particles are *internally* or *externally* mixed). Also the physical properties of particles vary widely with size and composition, making it necessary to use a large number of different sampling and analysis techniques to characterize particulate matter. All of these properties vary with the relative humidity of the air.

Long-range transboundary transport is responsible for a significant fraction of the particulate pollution in European cities as well as in rural areas (EMEP-CCC, 1999) and particulate matter is also subject to intercontinental transport. Long-range transport of particulate matter was specifically added to the EMEP work programme in 1998.

The establishment of an adequate monitoring programme is a crucial step in the development of abatement strategies for particulate matter. After the inclusion of particulate matter in the work plan of EMEP, a monitoring strategy was developed in close cooperation with the scientific community and national experts through the Task Force on Measurements and Modelling. Only minor changes to this strategy are introduced here.

The mass measurement part is based on standard EN 12341 of the European Committee for Standardization (CEN) (CEN, 1998) and recommends employing the gravimetric method, which has proven to be the most accurate method. Gravimetric methods also have the advantage of allowing chemical analysis of the collected  $PM_{10}$  sample after weighing. The application of the methods and quality assurance procedures recommended by the manual is important in order to harmonise the ongoing  $PM_{10}$  measurements throughout the EMEP network.

Collection of aerosols on a suitable filter and weighing is reasonably straightforward, provided that a suitable size-discriminating air inlet is being used. The construction of the air inlet is not critical for the >10 micron size range. With respect to a lower size cut-off, it has been discussed whether this should be 2.5 or 1 micrometer. Problems are generally related to the volatility problem. As much as 30% of the sample weight may be ammonium nitrate in some areas (the Netherlands), but also many chemical compounds in the organic fraction have appreciable vapour pressures at ambient temperatures. We still need to know how these constituents behave in the "conditioning" process.

An alternative to weighing is quantification by chemical methods. Actually, long measurement series of measurements in Europe are available for specific chemical constituents of aerosol, e.g., for sulphate and to some extent for nitrate and some other species. The sulphate series go back to 1972, and although the equipment and the data quality have improved over the years, some of the data series are fully consistent over this long time period. Much less information is available for

nitrate, and these data are much more susceptible to sampling artefacts. Inorganic elemental analysis using several methods has also been performed, with a very high quality with respect to both sampling and chemical analysis, allowing quantification of the inorganic mineral mass, as well as the sea-salt contribution. If the organic and the elementary carbon fractions could be quantified by chemical analysis in the same way, simple weighing of the filters (or mass quantification by other means) could be replaced by chemical analyses, giving more specific as well as more precise information. However, as discussed above, these sampling and analysis techniques are still under development. Ideally, aerosol measurements should also be size-specified. However, cascade impactors require relatively long sampling periods and are expensive to run over extended periods.

More specifically, in relation to aerosol particles, the task is to provide data for both individual chemical components and for the contribution of sources and source areas to the total particulate mass. Providing a detailed chemical mass balance is not simple. Sampling and analysing the individual components should be consistent with the determination of the total mass. The problems related to volatility and water content are both of concern in this connection. Data on aerosol mass alone is thus by far not sufficient to improve the understanding of particulate matter behaviour in the atmosphere and the associated emission rates and exposure levels.

From evaporative losses during the collection of organic carbon it was deduced that a substantial fraction of this material is semi-volatile (Noone et al., 2003). However, since the molecular composition of the organics cannot be determined yet, it is not known which organic species have this semi-volatile character. This in turn makes focused investigations and resolution of the sampling artefacts impossible. A complication is that, at the same time that evaporation of semivolatiles occurs, gaseous material adsorbs to the collection substrates. Notorious in this respect are the quartz and especially the glass-fibre filters that are prescribed as the filter material in the EU guidelines for sampling of PM<sub>10</sub> and PM<sub>2.5</sub>. This leads to potentially large overestimates of the actual concentrations of these parameters. A way to overcome this positive interference is the removal of the adsorptive gaseous organics with so-called gas denuders. However, the proper adsorptive material has not yet been found. To assess the evaporative losses, filter packs, consisting of a series of filters, are being used. Such approaches would seem to be simple, but are very costly and prone to contamination by all the necessary handling. To prevent this handling and for short-term measurements a commercial carbon-monitor (ACPM) is in principle available, and it has been tested in a field intercomparison campaign.

A comprehensive description of the atmospheric particles also requires the evaluation of particle number, surface and volume distributions in addition to their mass and their chemical composition. Size distribution measurements combined with chemical speciation are also necessary for identifying the sources of atmospheric particulate matter.

#### 7.5.2 Current situation and further needs

A number of countries have initiated measurements of  $PM_{10}$  mass during 2001 and 2002. Measurements of parameters other than  $PM_{10}$  mass are still sparse and

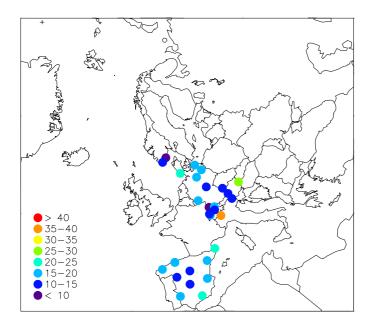
countries are strongly encouraged to expand their activities to meet the requirements. The EMEP monitoring programme thus still provides insufficient data for model validation. This is particularly the case for information on the chemical composition of the aerosol. Although secondary inorganic aerosol components such as nitrate and ammonium have been part of the programme for many years, only few countries report data obtained from measurements that allow for the separation of gas and aerosol phase for these compounds. Apart from their essential role in developing an improved description with respect to acidification and eutrophication, these compounds are also needed for the ongoing development of PM modelling.

For other parameters even less data are available. In particular for carbonaceous species information on the ambient concentration and chemical composition is generally lacking. Thus a comprehensive campaign aiming at determining concentrations of elemental and organic carbon concentrations at a number of sites across Europe has recently been conducted. Detailed information about this campaign can be found in EMEP/CCC-Report 4/2002. The experiences from the EC/OC campaign will give guidance on the further development of the monitoring strategy for these compounds.

There is an ongoing discussion whether  $PM_1$  mass should be preferred rather than  $PM_{2.5}$  mass measurements. While  $PM_1$  better resolves the accumulation mode from the coarse mode, is less affected from re-suspension resulting in improved spatial correlation and regional representativeness of the measurement data,  $PM_{2.5}$  is the parameter the health effects community is considering for establishing new air quality guideline values. From the perspective of improving the modelling capabilities,  $PM_1$  data would likely be better. The WMO-GAW programme, however, recommends  $PM_{2.5}$  to be measured. It is therefore at this stage not given preference to any to the size cut-offs in EMEP.

The initial testing of a particle dynamic module in the EMEP Unified model also highlights the need for accurate descriptions of size dependent dry and wet removal processes. Further, the model validation requires sufficient measurement data both in terms of site density, data quality and chemical/physical parameters determined. Modelling long-range transport of aerosols requires computations in a model atmosphere consisting of multiple layers. Mass and number concentrations of particles on the ground and in upper layers influence each other in various ways. A comprehensive validation of the EMEP unified model should therefore aim in the future at taking information on vertical distributions of particles into account. Mass and particle concentrations and chemical composition of aerosols measured on the ground may not be representative for the corresponding particle characteristics at higher altitudes. Various kinds of optical measurements can serve to retrieve information on the distribution and temporal evolution of particles at higher altitudes (i.e. Lidar instruments, sun photometers). Relevant parameters include aerosol optical depth, scattering coefficient, scattering ratio, depolarisation ration and others.

Figure 5 shows the number of sites measuring  $PM_{10}$  in 2001.



*Figure 5:* Annual averages of  $PM_{10}$  concentrations in 2001.

#### 7.5.3 Monitoring requirements for particulate matter

The strategy aims at improving the chemical mass closure i.e. full characterisation of inorganic compounds, EC/OC determination at selected sites, improved OC characterisation, chemical speciation as a function of size as well as physical characterisation (number size distribution and surface area distribution).

#### Level 1 sites;

The core data for assessing particulate matter is the daily measurements of inorganic compounds by filter pack as also required for acidification and eutrophication (see also Chapter 7.1.3) requiring determination of SO<sub>4</sub>, NH<sub>4</sub>, NO<sub>3</sub>, Na, Mg, Ca, K (Cl) (NH<sub>4</sub> and NO<sub>3</sub> to be estimated using values from filter packs, gas and particle fractions should be reported separately) and values for the gaseous fraction measured by low cost methods on monthly basis. Further, information about the  $PM_{10}$  mass is required to evaluate the chemical mass closure.

#### Level 2 sites;

At level 2 sites, more advanced measurements are required, including in addition to level 1 parameters, also;  $PM_{2.5}$  and/or  $PM_1$  mass, (daily), speciation of inorganics by size (two ranges ( $PM_{2.5}$  and  $PM_{10}$ ), mineral dust, elemental carbon (EC), Organic Carbon (OC) (all with a time resolution of one sample per week or longer). The gas/particle distribution for  $NH_3/NH_4$  and  $HNO_3/NO_3$  with denuder-filter pack method as defined for acidification and eutrophication is of high value.

#### Level 3 sites;

Parameters required at level 3 sites include Size/number distribution (hourly resolution, size 20-600 nm), light scattering (hourly), OC-speciation (grab samples or weekly samples), BC concentrations (hourly) and vertical profiles from using lidar.

## 7.6 Monitoring requirements at the various levels

# Table 2:Monitoring requirements for the various levels specified by the EMEP<br/>monitoring strategy.

Programme	rogramme Parameters	
Level 1 sites (all parameters a	re required to be monitored)	period/Frequency
Inorganic compounds in precipitation	$SO_4^-$ , $NO_3^-$ , $NH_4^+$ , $H^+$ (pH), $Na^+$ , $K^+$ , $Ca^{++}$ , $Mg^{++}$ , $CI^-$ , (cond)	24h/daily
Inorganic compounds in air	SO <sub>2</sub> , SO <sub>4</sub> <sup></sup> , NO <sub>3</sub> <sup></sup> , HNO <sub>3</sub> , NH <sub>4</sub> <sup>+</sup> , NH <sub>3</sub> , (sNO <sub>3</sub> , sNH <sub>4</sub> ), Na <sup>+</sup> , K <sup>+</sup> , Ca <sup>++</sup> , Mg <sup>++</sup> , NO <sub>2</sub>	24h/daily
Gas particle ratio Heavy metals in precipitation $PM_{10}$ mass concentration Ozone Meteorology	NH <sub>3</sub> , HNO <sub>3</sub> (in combination with filter pack sampling) Cd, Pb ( $1^{st}$ pri.ority), Cu, Zn, As, Cr, Ni (2nd priority) PM <sub>10</sub> O <sub>3</sub> Precipitation, temperature, wind direction/velocity, relative humidity, atmospheric pressure	monthly/monthly weekly 24h/daily continuous/hourly continuous/hourly
Level 2 sites (in addition to level		
Acidification and Eutrophicat Gas particle ratio Ammonia in emission areas (high spatial resolution)	tion NH <sub>3</sub> /NH <sub>4</sub> , HNO <sub>3</sub> /NO <sub>3</sub> (artefact free methods) NH <sub>3</sub> (low cost methods)	24h/daily monthly/monthly
<b>Photochemical oxidants</b> NO <sub>x</sub> Light hydrocarbons Carbonyls	NO, NO <sub>2</sub> C <sub>2</sub> -C <sub>7</sub> Aldehydes and ketones	continuous/hourly grab samp. or c/h 8h/2 days/week
Heavy metals Mercury in precipitation Mercury in air HMs in air	Hg Hg (TGM), Cd, Pb (1st priority.), Cu, Zn, As, Cr, Ni (2nd priority)	monthly 24h/weekly weekly/weekly
Persistent organic pollutants POP in precip. POP in air	PAHs, PCBs, HCB, chlordane, HCHs, DDT/DDE PAHs, PCBs, HCB, chlordane, HCHs, DDT/DDE	weekly/weekly 48h/weekly
<b>Particulate matter</b> PM mass Gas particle ratio	PM <sub>1</sub> , PM <sub>2.5</sub> NH <sub>3</sub> /NH <sub>4</sub> , HNO <sub>3</sub> /NO <sub>3</sub>	24h/daily 24h/daily
Speciation vs. size (PM2,5 and PM10) Mineral dust EC/OC	SO <sub>4</sub> <sup></sup> , NO <sub>3</sub> <sup>-</sup> , NH <sub>4</sub> , Na <sup>+</sup> , K <sup>+</sup> , Ca <sup>++</sup> , Mg <sup>+</sup> (Cl <sup>-</sup> ) Si EC, OC	weekly/weekly weekly/weekly weekly/weekly
Level 3 sites (do not require a	ll level 1 and level 2 parameters)	
Dry deposition flux of sulphur and nitrogen species	SO <sub>2</sub> , NH <sub>3</sub> , HNO <sub>3</sub> (SO <sub>4</sub> <sup></sup> , NH <sub>4</sub> <sup>+</sup> , NO <sub>3</sub> <sup></sup> )	
Dry deposition flux of O <sub>3</sub> Hydrocarbons Vertical profiles NO <sub>y</sub> chemistry Mercury speciation Congener specific POPs	O <sub>3</sub> C <sub>6</sub> -C <sub>12</sub> O3 soundings, PM Lidar, NO, NO <sub>2</sub> , PAN, organic nitrates TGM, RGM and TPM PCBs, PAHs, PCDDs and PCDFs	
Multi compartment (air, soil,	POPs and Hg	
water) Size/number distribution Light scattering OC-speciation "Black Carbon"	dN/dlogDp Aerosol Optical Depth, other optical parameters water soluble and water insoluble OC BC	

# 8. National monitoring requirements for the Parties for monitoring at the various levels

Table 3:Number of monitoring sites required at level 1 and proposal for<br/>numbers for level-2 sites for the different themes. Countries grouped<br/>by location to identify possible cooperation on level-2 sites. For<br/>regions with insufficient level-2 sites, an additional site requirement<br/>has been proposed ("# shared sites"). "Level-2 sites" takes into<br/>account the current monitoring.

		level 1		Level 2 (status + need)						
				POPs						
	Current #	Additional	# of	Acidification/	Photo-	Heavy metals		Particulate		
Country (*=party to EMEP)	of sites	need	sites	eutrophication		(precip + air)	air)	matter		
Iceland	1	neeu	1	cutiophication	Oxidants	1	1	matter		
Norway *	7		7		2	2	2	2		
Ireland *	4		4	0	1	1	1	1		
United Kingdom *	8		8	2	1	3	1	1		
Sweden *	5		5			1	2	1		
Finland *	4	1	5		1	1	1	2		
Estonia *	2		2							
Denmark *	4		4			2	1	1		
#shared sites				1	1			1		
Lithuania	1		1			1	1			
Latvia *	2		2			2				
#shared sites								1		
Russian Federation *	4	4	8		1	1	1	1		
Belgium *	0	1	1			1		1		
Liechtenstein *	0	I	1			I		I		
Luxembourg *	0									
			0		0	4	4	4		
France *	8		8		2	1	1	1		
Netherlands *	2		2		1	1	1	1		
Germany *	8		8	-	2	4	1	2		
#shared sites				2	1		1			
Hungary *	1		1	1				1		
Czech Republic *	2		2		1	2	1	1		
Austria *	3		3					1		
Switzerland *	5		5					3		
#shared sites										
Belarus *	0	1	1					1		
Republic of Moldova	Ő	1	1					I		
Romania	0	2	2							
Ukraine *	0	2	2							
		3						4		
Poland *	4		4			_		1		
Slovakia *	5		5			5		1		
#shared sites				1	1		1			
Armenia	0	1	1							
Azerbaijan	0	1	1							
Kazakhstan	0	2	2							
Georgia	0	1	1							
Turkey *	1	1	2					1		
Kyrgyzstan	1		1							
#shared sites				1	1	1	1	1		
Spain *	10		10					1		
Portugal *	3		3		1			1		
#shared sites			5	1	I	1	1	I		
Monaco *	0			· ·		1	1			
San Marino	0	4	~		4			0		
Italy *	2	1	3	1	1			2		
Slovenia *	1		1	ļ ,						
#shared sites				1	1	1	1	1		
Bosnia and Herzegovina *	0	1	1							
The FYR of Macedonia	0	1	1							
Malta *	0	1	1							
Bulgaria *	0	1	1							
Cyprus *	0	1	1							
Greece *	ů 1	1	2		1			1		
Serbia and Montenegro	2		2							
Croatia *	2		2							
#shared sites			2	1	1	1	1	1		
#Silaieu Siles			100							
	103		130	10	20	32	20	33		

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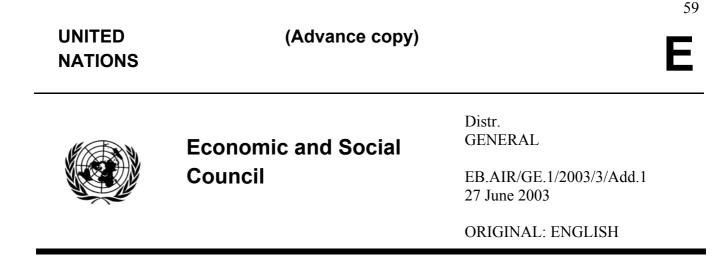
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# Appendix A

# Draft EMEP monitoring strategy and measurement programme 2004-2009 prepared by the Chemical Coordinating Centre in consultation with the Bureau and with the assistance of the secretariat

Advance copy presented for the EMEP Steering Body, September 2003 (EB.AIR/GE.1/2003/3/Add.1)



## ECONOMIC COMMISSION FOR EUROPE

EXECUTIVE BODY FOR THE CONVENTION ON LONG-RANGE TRANSBOUNDARY AIR POLLUTION

Steering Body to the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission for Air Pollutants in Europe (EMEP) (Twenty-seventh session, Geneva, 8-10 September 2003) Item 4 (e) of the provisional agenda

#### **MEASUREMENTS AND MODELLING**

#### Addendum

### Draft EMEP monitoring strategy and measurement programme 2004-2009 prepared by the Chemical Coordinating Centre in consultation with the Bureau and with the assistance of the secretariat

#### **Introduction**

1. EMEP is the Convention's Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollution in Europe. Most Parties to the Convention are Parties to the Protocol on Long-term Financing of EMEP, which defines the obligations and financial contributions of the Parties to the programme. The EMEP monitoring programme is an integral, essential part of EMEP.

2. The EMEP Steering Body at its twenty-sixth session requested the EMEP centres to further elaborate, in close collaboration with national experts, a new monitoring strategy, which would be the basis for the measurement programme of EMEP in the coming years.

Documents prepared under the auspices or at the request of the Executive Body for the Convention on Long-range Transboundary Air Pollution for GENERAL circulation should be considered provisional unless APPROVED by the Executive Body.

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3. In spring 2003, the Task Force on Measurements and Modelling and the programme centres reviewed the EMEP monitoring strategy and considered its revision. In developing the new draft monitoring strategy, due consideration was given to the current strengths and weaknesses of the programme as well as the opportunities for its development and the threats to its future. The strategy takes account of the required level of reporting by Parties, new requirements from the users of data, and recent technical developments. A comprehensive description of these elements can be found in a report by the Chemical Coordinating Centre (CCC) -Report 9/2003.

4. The main EMEP objective is to provide the Convention with technical information on depositions and concentrations of air pollutants, and on quantities and source allocation of long-range transmission of pollutants and transboundary fluxes, related to acidification, eutrophication, photo-oxidants, particulate matter, heavy metals and persistent organic pollutants. Such information is also made available to other interested bodies and institutions. This information is the basis for policy development and implementation of the Convention. Under the activities of EMEP, annual emission data are collected, air quality and deposition are assessed through measurements performed within the programme, and models for atmospheric chemistry and transport and integrated assessment are developed and operated. The EMEP observations and model calculations are essential to establish a reliable picture of the air pollution situation in Europe, and to build up the confidence required for policy development on transboundary pollution issues, including the links both to global- and to urban-scale transport.

## I. OBJECTIVES OF EMEP MONITORING

5. The EMEP monitoring programme must provide the observational underpinning to:

(a) Establish pollutant concentrations, deposition, emissions and transboundary fluxes on the regional scale, including intercontinental transport and boundary conditions for urban air quality;

(b) Identify the trends with time as well as their sensitivity to European emission reductions;

(c) Assess the success of international abatement strategies for atmospheric pollutants;

(d) Improve the understanding of atmospheric chemical and physical processes and provide data for the validation of models;

(e) Provide data which, in conjunction with models, are the basis for the assessment of environmental problems related to air pollution, including comparison with effect thresholds and exposure levels;

(f) Provide the measurements required to assess the effects of atmospheric pollutants;

(g) Serve to explore the environmental concentrations of new substances and support the development of cost-effective abatement strategies.

6. In addition, EMEP should make increasing contributions to provide cost-effective measurements also related to environmental issues partly outside the focus of the Convention, e.g. urban air quality; climate; atmospheric deposition in relation to biodiversity and water quality. Such issues fall under European Union (EU) legislation such as the Directive on Ambient Air Quality Assessment and Management ("Air Quality Framework Directive") and its daughter directives and the National Emissions Ceilings Directive, as well as under the United Nations Framework Convention for Climate Change, the Helsinki Commission of the Convention on the Protection of the Marine Environment of the Baltic Sea Area (HELCOM), the Oslo and Paris Commissions for the Convention for the Protection of the Marine Environment of the North-East Atlantic (OSPARCOM) and the Arctic Monitoring and Assessment Programme (AMAP) as well as other international regulations and agreements.

## **II. STATUS OF THE MONITORING NETWORK TODAY**

7. The EMEP monitoring network has, for more than 25 years, provided high-quality data on atmospheric concentrations and deposition of transboundary air pollution. The network has been successful in bringing together a large number of countries performing measurements using common methodologies and with quality assurance and control systems including site visits and intercomparison studies. From its early priorities of sulphur and nitrogen compounds, the network has developed to include the new themes and priorities of air pollution policies, including ozone, volatile organic compounds (VOCs), particulate matter, heavy metals and persistent organic compounds (POPs). The openness of the programme and the general availability of the data have been of great importance for creating a common basis for policy development.

8. However, the development of the programme has not generally benefited from national funding in proportion to the increased requirements. This has resulted in large variations, especially between different regions of Europe, in the ability to implement fully or even partly the programme or to provide data of adequate quality. For all parameters, the monitoring activity is less than satisfactory in the Mediterranean area and in Eastern Europe. In Central, Western and Northern Europe the situation is better, but here too significant improvements to the monitoring network for specific themes are required, e.g. particulate matter (PM), POPs and VOCs (see CCC Report 9/2003). There is a general obligation, under article 9 of the Convention, regarding the need to implement and further develop EMEP, including emphasis on aspects of a monitoring programme. For such a monitoring programme to be fully effective in supporting the Convention, national monitoring should be at least in line with the minimum requirements described below.

#### A. <u>Quality assurance/quality control including data storage and dissemination</u>

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9. EMEP has a well-recognized history of quality assurance and quality control with strong bottom-up and top-down approaches to monitoring. This has resulted in good links between national data producers and central data collection and dissemination. Some data series established through EMEP range over several decades and are essential for assessing the success of abatement measures, including future ones.

#### B. Challenges

10. To address topics of political interest, such as the intercontinental transport of air pollutants, the measurement programme needs to be extended to new regions. This includes establishing sites in areas not sufficiently covered, in particular beyond the political boundaries of the European Community to the far Eastern part of the EMEP region, including Central Asia. It would also be useful to have monitoring sites in North Africa and in the Eastern Mediterranean region. A transfer of technology, including equipment and training, is required for many countries. For this, modest investments will be needed.

11. Monitoring in EMEP is essential for the technical underpinning of the Convention in the calculation of the deposition of acidifying and eutrophying compounds, photo-oxidants, base cations, heavy metals and persistent organic compounds. The data also provide essential information on the regional concentration of health-relevant pollutants including particulate matter, O<sub>3</sub> and NO<sub>2</sub>. In recent years, it has become clear that the monitoring data are essential inputs to studies on biodiversity and changes in water quality, as well as for determining temporal and spatial variability in the radiative climate forcing resulting from regional pollutants, where particulate matter is especially important. In view of its high costs, it is evident that monitoring should be multi-purpose and cost-effective. Full advantage should be taken of other monitoring frameworks in Europe, such as the Global Atmosphere Watch (GAW), the national monitoring networks reporting to the European Commission under the European Union's air quality directives and the Exchange of Information (EoI) Decision as well as those under the EUROAIRNET umbrella of the European Environment Agency, as well as national and local monitoring efforts.

12. The monitoring of atmospheric air pollutants in rural and background areas is a responsibility of EMEP. Unfortunately, there are pressures on the EMEP monitoring network. For example, the European Commission has its own monitoring requirements under the EU Air Quality Framework Directive and its daughter directives; for these it can impose sanctions for non-compliance. The resulting shift in emphasis of monitoring efforts by some Parties is of concern, since it causes increasing uncertainties in the evaluation of the effects of emission reduction measures. The EMEP network needs to be revised and strengthened in order to serve the needs of the Convention. Such a revision should take into account all other needs including those of the European Community, those of other international conventions (e.g. United Nations Framework Convention on Climate Change, Convention on Biological Diversity) and those of individual

countries. When such considerations are dealt with comprehensively, improvements or even cost reductions may be identified without compromising the value and strength of the information derived. This is essential to achieve the necessary technical underpinning for the implementation of current air pollution legislation, as well as for the work to develop new policies.

#### **III. THE NEW EMEP MONITORING STRATEGY**

#### A. Minimum requirements

13. To fulfil the EMEP responsibilities for reporting on concentrations and depositions, and for validating the EMEP models, the monitoring strategy sets minimum monitoring requirements for Parties to participate in the EMEP monitoring network. This minimum level of monitoring will provide the input to the EMEP mandatory requirement programme. Such monitoring is considered essential for an effective EMEP network to support the Convention. EMEP also needs to complement this monitoring with information from other related activities, such as those associated with other international agreements/regulations and with the research community at large.

14. The new monitoring strategy identifies different "levels" for monitoring. Levels 1 and 2 together represent the minimum monitoring requirements (the mandatory requirement programme). Measurements in this programme are separated into a large number (>125) of level-1 sites with a complete range of measurements of the major inorganic compounds (in air and precipitation), ozone, heavy metals (in precipitation), and particulate matter mass, and fewer (about 25) level-2 sites with more difficult or expensive measurements (table 1). The mandatory requirement programme should be harmonized with the requirements for rural measurements in the EU air quality daughter directives, though some differences will result from differences in objectives. Level-2 sites should provide relevant information for the themes addressed by EMEP (acidification and eutrophication; photochemical oxidants; heavy metals; POPs; and particulate matter).

15. A minimum number of level-1 sites is defined for each country, while for level-2 sites there is a preliminary proposal for countries as well as for regions (table 2). There is a need for regional cooperation to provide a sufficient number of level-2 sites to minimize costs; cost-sharing options are proposed. Monitoring in support of HELCOM, OSPARCOM and AMAP will contribute significantly to level-2 sites for heavy metals and POPs. EMEP also requires specialized measurements; these are available from state-of-the-art monitoring sites that are continuously operated or from research/field experiments (level-3 sites). A schematic overview of the various levels is given in the figure below.

16. EMEP will make use of relevant data from other networks either operated under other subsidiary bodies of the Convention, e.g. the Working Group of Effects, or available from monitoring activities performed under the air quality directives of the European Union (associated sites) or from GAW under the World Meteorological Organization (WMO) (the joint supersites).

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17. The following criteria have been defined for the various site levels:

(a) Level-1 sites. A minimum site density of one site per 50,000 km<sup>2</sup> is recommended, with a higher density in complex terrain. All countries with a geographical area exceeding 25,000 km<sup>2</sup> should operate at least one level-1 site. For large countries with a very low current site density, there is a realistic proposal for the number of sites that need to be established. Full implementation of level 1 is required for fulfilling the mandatory requirement programme. This will ensure participation of a large number of sites aimed at describing spatial trends, it will establish temporal trends and provide data for the development and validation of models. The network should have good spatial coverage and long-term operation is assumed. As the level-1 sites should involve all Parties, they should ensure that there is an ongoing operational activity linking all Parties and EMEP centres. It is seen as a very important way for individual countries to take an active part in the work of EMEP and the Convention. The level-1 activity will directly serve national interests and needs and will satisfy monitoring obligations at rural sites as specified by the EU Air Quality Framework Directive;

(b) Level-2 sites. A minimum density of one site per 100,000 km<sup>2</sup> is recommended (not necessarily for each individual theme); the density can vary between themes depending on region, e.g. photo-oxidants and particles should have a higher site density in Southern Europe, acidification and POPs a higher density towards the North and East. Within regions there should be cooperation to share costs. Level 2 covers advanced measurements that are more expensive and technically demanding at selected sites (table 1). Level-2 sites also need to measure all parameters measured at level 1. Level-1 and level-2 sites serve the same objectives and should provide good regional coverage;

(c) Level-3 sites relate to research data for process studies or demanding methodologies, including data from sources external to EMEP (table 1). Level-3 sites are voluntary. Level-2 and level-3 sites are called "EMEP supersites"; this is intended to be an important motivating factor and to provide appropriate recognition of the data providers. Level-3 sites are topic-specific and do not need to cover all substances defined for levels 1 and 2. Supersites should be encouraged to support both EMEP and GAW.

## B. Cost-efficient multi-purpose monitoring at urban to hemispheric spatial scales

18. To address questions related to the intercontinental transport of air pollutants, the measurement programme should be extended to new regions to establish sites in areas not sufficiently covered, in particular to the far eastern part of the EMEP region (e.g. Central Asia). Sites in North Africa and the Eastern Mediterranean would also be of value. Furthermore, EMEP should, where possible, integrate with other monitoring networks to form a "multi-purpose" network linking urban, rural and global sites. Remote sensing from satellites needs to become an integral part of the observations where the ground-based network provides surface and vertical

profile data for the calibration and validation of remote-sensing instruments. Only by doing this can new challenges such as "global change", "earth systems" and biogeochemical cycles (fluxes between air, water and soil) be addressed properly and cost-effectively. Such an approach will facilitate the combination of information from all relevant sources, and the provision of adequate data will be ensured by combining the resources from several major sources: (a) national budgets for funding EMEP monitoring; (b) funding associated with other parts of the Convention or other international agreements (HELCOM, OSPARCOM, AMAP, United Nations Framework Convention on Climate Change); or (c) national monitoring in support of the EU Air Quality Framework Directive. There may also be indirect support given to the scientific community including national and international research and monitoring programmes (WMO-GAW, the European Commission's research programmes). By combining resources from these areas, more efficient use of resources is ensured by avoiding duplication of effort, and a sound observations base for EMEP is maintained.

## C. Quality of observation data and instrumentation

19. EMEP will maintain and further improve its quality assurance programme to make sure that observation data are of known quality and adequate for their intended use. Field intercomparisons and laboratory ring tests are important, as well as the maintenance of good links between national data providers and data collection under EMEP. These activities can be strengthened by combining resources with the central quality assurance facilities in the EU and in the GAW system. The EMEP Manual for Sampling and Chemical Analysis gives the criteria that need to be satisfied for instrumentation and analytical methods. Other methods such as automatic monitors can replace manual methods when data quality is equivalent or better.

#### D. Data collection, value adding, dissemination and transparency

20. Data submission to CCC shall follow the protocols agreed upon. A transparent (Internetbased) system is in place for adding other types of data, notably meteorological information and, in collaboration with the EMEP centres, emissions data and model results. The system is being developed by consulting regularly with users of EMEP information.

21. The EMEP Centres will use the data reported to meet their reporting requirements, for assessing pollutant concentrations, deposition, emissions and transboundary fluxes within the EMEP region, to the EMEP Steering Body.

## **IV. IMPLEMENTATION AND FURTHER EVOLUTION OF THE STRATEGY**

22. The mandatory requirements programme at level 1 is already partly implemented in many countries. The remaining countries should follow without delay.

23. Some level-2 sites have already been established and, in addition, a number of potential sites

have been identified. In some regions there is a further need for level-2 sites and Parties should cooperate in order to reduce total costs. The selection of level-2 sites will be done in consultation with CCC.

24. Level-3 activities relate to research studies on processes together with the collection of data on parameters not included in levels 1 and 2, including data from sources external to EMEP. Level 3 may also include studies on developing methodologies. It is expected that some activities at level 3 sites will with time be moved into the mandatory requirements programme, primarily as level-2 activities. This applies to:

(a) Flux monitoring of SO<sub>2</sub>, NH<sub>3</sub> and HNO<sub>3</sub> using the conditional time averaged gradient (COTAG) method and of ozone and particulate matter using eddy correlation techniques;

(b) Improved cut-off in the characterization of size fractions of particulate matter, and chemical characterization at level-2 and level-1 sites;

(c) Inclusion of methane, carbon monoxide and other climate-relevant species in the measurement programme;

(d) Improvement of time resolution and data availability for users including the general public, as adequate monitors become available to replace manual methods at low concentrations;

(e) Measurement of three mercury forms: total gaseous mercury, reactive gaseous mercury and total particulate mercury;

(f) Improved measurement programme for POPs to include congener-specific compounds such as PCDDs and PCDFs;

(g) Extension of VOC monitoring to include heavier components (up to  $C_{12}$  compounds).

25. The EMEP monitoring network must be a dynamic system ready to adopt new needs and requirements without losing its ability to develop long-term trend data to illustrate compliance with agreed emission reductions. This requires that the work should be reviewed and revised at regular intervals.

### Figure. Schematic overview of the levels defined by the EMEP monitoring strategy

#### **Objectives of monitoring programme**

•establish pollutant concentrations and deposition fluxes on the regional scale, including intercontinental transport and boundary conditions for urban air quality; the trends with time; and sensitivity to European emission reductions

• assess the success of international abatement strategies for atmospheric pollutants

• improve the understanding of atmospheric chemical and physical processes and provide data for the validation of models

• provide data which in conjunction with models are the basis for the assessment of environmental problems related to air pollution including comparison with effect thresholds and exposure levels

• provide measurements required to assess the effects of atmospheric pollutants

•serve to explore the distribution of new substances and support the development of costeffective abatement strategies

#### Mandatory requirement programme

#### Level 1

Parameters of general interest for all EMEP themes; majc inorganic compounds in precipitation and in air, heavy metals in precipitations ozone, PM10 mass concentration meteorology, permanent monitoring at about 125 sites (80 sites for heavy metals)

#### Level 2 (supersites)

Topic-specific, acidification and eutrophication, photochemical oxidants, heavy metals, persistent organic pollutants, particulate matter, should in addition include level-1 parameters. Permanent monitoring at about 15-25 sites with proper regional distribution

#### Voluntary programme

#### Level 3 (supersites)

Topic-specific, highly specialized measurements, may include campaign data, do not require all level-1/level-2 parameters to be measured. About 10-15 sites

#### Associated sites

Use of data available from other bodies under the Convention (e.g. Working Group on Effects), monitoring in support of the EU Air Quality Framework Directive (and daughter directives)(03, PM10). Ozone soundings, 300-500 sites

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Table 1. Monitoring	requirements	for	the	various	levels	specified	by	the	EMEP	monitoring
strategy										

Programme	Parameters	Measurement period/Frequency
Level-1 sites (all parameters a	re required to be monitored)	, • <b></b>
Inorganic compounds in precipitation	SO <sub>4</sub> <sup></sup> , NO <sub>3</sub> <sup>-</sup> , NH <sub>4</sub> <sup>+</sup> , H <sup>+</sup> (pH), Na <sup>+</sup> , K <sup>+</sup> , Ca <sup>++</sup> , Mg <sup>++</sup> , Cl <sup>-</sup> (cond)	24h/daily
Inorganic compounds in air	SO <sub>2</sub> , SO <sub>4</sub> <sup></sup> , NO <sub>3</sub> <sup>-</sup> , HNO <sub>3</sub> , NH <sub>4</sub> <sup>+</sup> , NH <sub>3</sub> , (sNO <sub>3</sub> , sNH <sub>4</sub> ), Na <sup>+</sup> , K <sup>+</sup> , Ca <sup>++</sup> , Mg <sup>++</sup> , NO <sub>2</sub>	24h/daily
Gas particle ratio Heavy metals in precipitation $PM_{10}$ mass concentration Ozone	NH <sub>3</sub> , HNO <sub>3</sub> (in combination with filter pack sampling) Cd, Pb (1 <sup>st</sup> priority), Cu, Zn, As, Cr, Ni (2nd priority) PM <sub>10</sub> O <sub>3</sub>	monthly/monthly weekly 24h/daily continuous/hourly
Meteorology	Precipitation amount (RR), temperature (T), wind direction (dd), wind speed (ff), relative humidity (rh), atmospheric pressure (pr)	continuous/hourly
Level-2 sites (in addition to lev	vel-1 narameters):	
Acidification and eutrophicati		
Gas particle ratio	NH <sub>3</sub> /NH <sub>4</sub> , HNO <sub>3</sub> /NO <sub>3</sub> (artefact-free methods)	24h/daily
Ammonia in emission areas (high spatial resolution)	NH <sub>3</sub> (low-cost methods)	monthly/monthly
Photochemical oxidants		
NO <sub>x</sub>	NO, NO <sub>2</sub>	continuous/hourly
Light hydrocarbons	C <sub>2</sub> -C <sub>7</sub>	grab samp. or c/h
Carbonyls	Aldehydes and ketones	8h/2 days/week
Heavy metals		
Mercury in precipitation	Hg	monthly
Mercury in air	Hg (TGM), Cd. Dh (1-t priority) Co. Zo. A. Co. Ni (2nd priority)	24h/weekly
Heavy metals in air	Cd, Pb (1st priority.), Cu, Zn, As, Cr, Ni (2nd priority)	weekly/weekly
Persistent organic pollutants		11/11
POPs in precipitation POPs in air	PAHs, PCBs, HCB, chlordane, HCHs, DDT/DDE PAHs, PCBs, HCB, chlordane, HCHs, DDT/DDE	weekly/weekly 48h/weekly
Particulate matter		
PM mass	PM10 <sub>1</sub> , PM <sub>2.5</sub>	24h/daily
Gas particle ratio	NH <sub>3</sub> /NH <sub>4</sub> , HNO <sub>3</sub> /NO <sub>3</sub>	24h/daily
Speciation vs. size (PM2.5 and PM10)	SO <sub>4</sub> <sup></sup> , NO <sub>3</sub> <sup>-</sup> , NH <sub>4</sub> , Na <sup>+</sup> , K <sup>+</sup> , Ca <sup>++</sup> , Mg <sup>+</sup> (Cl <sup>-</sup> )	weekly/weekly
Mineral dust	Si	weekly/weekly
elemental carbon (EC)		weekly/weekly
organic carbon (OC)	EC, OC	5 5
	ll level-1 and level-2 parameters)	
Dry deposition flux of sulphur	$SO_2$ , $NH_3$ , $HNO_3$ ( $SO_4^{}$ , $NH_4^{++}$ , $NO_3^{}$ )	
and nitrogen species		
Dry deposition flux of O <sub>3</sub> Hydrocarbons	O <sub>3</sub> C <sub>6</sub> -C <sub>12</sub>	
Vertical profiles	O3 soundings, PM lidar,	
$NO_v$ chemistry	NO, NO <sub>2</sub> , PAN, organic nitrates	
Mercury speciation	TGM, RGM and TPM	
Congener-specific POPs	PCBs, PAHs, PCDDs and PCDFs	
Multi-compartment (air, soil, water)	POPs and Hg	
Size/number distribution	dN/dlogDp	
Light scattering	Aerosol optical depth	
OC speciation	water soluble and water insoluble OC	
"Black carbon"	BC	

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Table 2. Number of monitoring sites required at level 1 and proposal for numbers for level-2 sites for the different themes. Countries grouped by location to identify possible cooperation on level-2 sites. For regions with insufficient level-2 sites, an additional site requirement has been proposed ("# shared sites"). "Level-2 sites" takes into account the current monitoring

		level 1		Level 2 (status + need)						
				POPs						
Country (*=party to EMEP)	Current # of sites	Additional need	# of sites	Acidification/ eutrophication	Photo- oxidants	Heavy metals (precip + air)	(precip + air)	Particulate matter		
Iceland	1		1			1	1			
Norway *	7		7		2	2	2	2		
Ireland *	4		4		1	1	1	1		
United Kingdom *	8		8	2	1	3	1	1		
Sweden *	5		5			1	2	1		
Finland *	4	1	5		1	1	1	2		
Estonia *	2 4		2 4			0	4	4		
Denmark *	4		4	1	1	2	1	1 1		
#shared sites	1		1	1	I	1	1	I		
Latvia *	2		2			2	I			
#shared sites			2			2		1		
Russian Federation *	4	4	8		1	1	1	1		
Belgium *		1	1		•	1		1		
Liechtenstein *	0	ı	I			1		'		
Luxembourg *	0									
France *	8		8		2	1	1	1		
Netherlands *	2		2		1	1	1	1		
Germany *	8		8		2	4	1	2		
#shared sites	-			2	1		1			
Hungary *	1		1	1				1		
Czech Republic *	2		2		1	2	1	1		
Austria *	3		3					1		
Switzerland *	5		5					3		
#shared sites										
Belarus *	0	1	1					1		
Republic of Moldova	0	1	1							
Romania	0	2	2							
Ukraine *	0	3	3					4		
Poland * Slovakia *	4 5		4 5			5		1 1		
#shared sites	5		5	1	1	5	1	1		
Armenia	0	1	1	I	I		I			
Azerbaijan	0	1	1							
Kazakhstan	Õ	2	2							
Georgia	õ	1	1							
Turkey *	1	1	2					1		
Kyrgyzstan	1		1					-		
#shared sites				1	1	1	1	1		
Spain *	10		10					1		
Portugal *	3		3		1			1		
#shared sites				1		1	1			
Monaco *	0									
San Marino	0		_							
Italy *	2	1	3	1	1			2		
Slovenia *	1		1		,		<i>,</i>			
and #shared sites		4	4	1	1	1	1	1		
Bosnia and Herzegovina *	0	1	1							
The FYR of Macedonia	0 0	1 1	1							
Malta * Bulgaria *	0	1	1 1							
Cyprus *	0	1	1							
Greece *	0	1	2		1			1		
Serbia and Montenegro	2	I	2		I			I		
Croatia *	2		2							
#shared sites			~	1	1	1	1	1		
	103		130	10	20	32	20	33		
				10		52		50		