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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 of Air Pollutants in Europe (EMEP) (Twenty-fifth session, Geneva, 3-5 September 2001) Item 4(a) of the Provisional Agenda

MEASUREMENTS AND MODELLING

<u>Progress report prepared by the Chairman of the</u> <u>Task Force on Measurements and Modelling in collaboration with the secretariat</u>

Introduction

1. This report presents progress on atmospheric measurements and modelling. It includes the results of the first and second meetings of the Task Force on Measurements and Modelling, held in Vienna on 23-25 October 2000 and in Portoroz (Slovenia) from 30 May to 1 June 2001. On 30 May 2001, a joint session of the sixth Workshop on Air Quality Management and Assessment of the European Environment Information and Observations Network (EIONET) and the second meeting of the Task Force on Measurements and Modelling was held.

2. This report presents proposals from the Task Force for its terms of reference and its work programme for the next 3-5 years. It also presents the progress in the preparation of an assessment report on the changes in the transboundary fluxes, depositions and concentrations. Furthermore, the Task Force has reviewed the status of work on measurement and modelling of fine particulate matter and of heavy metals, and the report presents recommendations to the EMEP Steering Body for this work. The Task Force discussions on the transition from Lagrangian to Eulerian modelling are also reported.

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3. The presentations made at the second meeting of the Task Force are available on the Internet at <u>www.ubavie.gv.at/tfmm</u>.

4. At the first meeting, experts from Armenia, Austria, Belgium, Croatia, the Czech Republic, Denmark, Finland, France, Georgia, Germany, Hungary, Ireland, Italy, Latvia, Malta, the Netherlands, Norway, the Russian Federation, Slovenia, Sweden, Switzerland, the former Yugoslav Republic of Macedonia, the United Kingdom and the European Community (EC) participated. Representatives from the World Meteorological Organization (WMO), the four EMEP centres (Centre for Integrated Assessment Modelling (CIAM), Chemical Coordinating Centre (CCC), Meteorological Synthesizing Centre-East (MSC-E), and Meteorological Synthesizing Centre-West (MSC-W)) and the EC Joint Research Centre (JRC) also attended. Mr. Jürgen SCHNEIDER (Austria) and Mr. John MILLER (WMO) co-chaired the first meeting.

5. At the second meeting, experts from Austria, Belarus, Bosnia and Herzegovina, Croatia, the Czech Republic, Cyprus, Denmark, Estonia, Finland, France, Germany, Hungary, Iceland, Ireland, Italy, Latvia, Lithuania, the Netherlands, Norway, Poland, Slovakia, Slovenia, Sweden, Switzerland, the former Yugoslav Republic of Macedonia, the United Kingdom and Yugoslavia participated. Representatives from the four EMEP Centres, CIAM, CCC, MSC-E, and MSC-W, the European Environment Agency (EEA), JRC and the WMO also attended. Mr. Jürgen SCHNEIDER (Austria) and Ms. Liisa JALKANEN (WMO) co-chaired the second meeting.

I. ASSESSMENT OF CHANGES IN TRANSBOUNDARY FLUXES;

DEPOSITIONS AND CONCENTRATIONS

A. <u>Planning of the preparation of the assessment report</u>

6. The Task Force on Measurements and Modelling discussed the preparation of an assessment report intended to serve national needs and to provide a basis for the next round of negotiations. The Task Force takes the main responsibility for this report. The assessment report will be a joint report that should be prepared by the national experts associated with the work of EMEP and the EMEP centres. The following structure for the report was agreed provisionally:

Part 1. Overall European perspective

Introduction The regional air pollution problems and strategies to abate them The environmental problems Acidification Eutrophication Photo-oxidant pollution Heavy metal pollution Persistent organic compound pollution Trends Transboundary fluxes Non-linearities Conclusions Recommendations for the new EMEP Strategy

Part 2: The development in individual countries

Emissions. National measures and prospects for 2010 Concentration and deposition trends Transboundary fluxes (import-export) Perspective for 2010 National activities (scientific research in relation to EMEP)

7. Experts at the Swedish Environmental Research Institute (IVL), funded within the framework of the Swedish ASTA programme, (International and National Abatement Strategies for Transboundary Air Pollution), offered to serve as focal point for the work on the finalization of the assessment report. All national experts were invited to take an active part in its production. The Task Force agreed that the whole process was very demanding and should be closely followed.

8. For the implementation of the new EMEP strategy and the finalization of the assessment report it is necessary to obtain:

- Full support by the Executive Body and the EMEP Steering Body;
- Full awareness and support by national authorities; and
- Financial support from as many sources as possible, including the centres, the Parties and national and international research programmes.
- 9. CCC and MSC-W have made data available to facilitate the work by national experts:
 - Metadata of stations (currently available from 5 countries, 73 sites in total);
 - A consistent system of data flagging;
 - All daily acid deposition data from reported EMEP sites in an easy downloadable form;
 - Daily modelled data at EMEP sites calculated with the Lagrangian acid deposition model for the period 1985- 1996 in the same downloadable form as measured data;
 - Air mass classification (from 1985 1996) for several sites;
 - Software tools (non-commercial) for analysing trends;
 - A report on trends observed at EMEP stations (CCC Report 7-2000).

This work by CCC and MCS-W was not fully covered by the EMEP budget and was very timeconsuming. It was therefore highly appreciated by the Task Force. 10. In the near future, in addition, the two centres will make available:

- Metadata of stations for all countries;
- Air mass classification (from 1985-1996) for all EMEP sites.

Moreover, MSC-W will continue working on the development of an Internet-accessible version of the Lagrangian model. Model results from the Eulerian acid deposition model for 1980, 1985, 1990, 1995 and 2000 are expected to be available by the end of 2002.

11. Ms. Gun Lövblad (IVL), presented the plans for the preparation of the assessment report and a timetable for the initial phase. For the national assessments, IVL will establish contacts with national experts to get an overview of the work that has already been carried out, obtain information about national plans for the assessment and find out whether any discussions have been initiated with neighbouring countries regarding joint activities. A proposed minimum list of tasks, which should be carried out by national experts, as well as a guidance document on methods will be drawn up. A work-plan will be drafted in time for the twenty-fifth session of the EMEP Steering Body in September 2001.

12. For the European assessment, IVL will continue the dialogue with the group of experts taking part in the overall assessment and prepare a plan on how to share the tasks. This plan will include a proposal for an editorial committee. The dialogue with CCC and MSC-W will continue. In collaboration with MSC-E, a proposal will be prepared on how to deal with the assessment of heavy metals and persistent organic pollutants (POPs).

13. The Task Force agreed that national experts should initiate or continue work to examine the data for their countries that were available on the EMEP homepage. They should especially check the quality and completeness of data, and start the assessment work. The focus of the work should be on:

- The quality and completeness of the measured data, possibly with recommendations on correction factors useful for the assessment of trends;
- The analysis of the representativeness of trends by the use of other national network data and modelled results;
- The evaluation of the origin of the observed and modelled trends (source allocation).

The national assessment by the countries should provide conclusions on:

- The result of emission reduction measures within the country and internationally;
- The present status in relation to the desired environmental quality;
- The need for further actions to reduce pollution levels.

B. <u>Results of work by national experts</u>

14. In Canada, four assessment reports on acidification were published in the past 20 years. Each report was divided into two parts - an executive summary containing the main

conclusions in an easily understood and concise form, and a comprehensive scientific background document. This format has proven very successful.

15. Work done in Denmark on trends in air concentrations and depositions of certain pollutants from 1978 to 1997 showed a significant downwards trend in sulphur, while the situation for nitrogen compounds was less clear. The complex relationships between ammonia, ammonium and sulphur compounds were highlighted.

16. Work done in Yugoslavia underlined the great importance of proper checking of air quality data available on the EMEP/CCC web site. Within the trend assessment, a clear drop in sulphur concentrations was detected from the 1980s to the 1990s.

17. In Slovenia, a comprehensive study on trends and the origin of air pollutants was conducted in cooperation with a Canadian expert, using backward trajectories. Since the results seem representative for the whole region, other countries could benefit from this analysis.

18. In Lithuania, measurement data from one EMEP station from 1981 to 1999 were examined with a special focus on the seasonality of trends. Winter SO₂ concentrations showed a sharper decline during the past decade than summer concentrations.

19. By performing parallel measurements at some German EMEP sites, systematic differences between the results of SO_2 and NO_2 measurements using manual methods and automated monitors were detected. The results measured by the automated monitors were on average 50% higher than those obtained with manual methods. The Task Force recognized that it was necessary to apply a correction factor to the data on the EMEP/CCC web site and to flag the data for German EMEP sites accordingly.

20. In Belarus, the first monitoring station for precipitation was established in the early 1960s. Sulphur deposition in the past decades, while the situation for reduced and oxidized nitrogen was less clear.

21. In the Czech Republic, SO_2 emissions, concentrations and dry deposition have declined significantly over the past decade. However, wet deposition has been rather constant during that period.

22. In the United Kingdom, a study on wet and dry deposition trends of sulphur between 1986 and 1997 has been conducted. The results show that, while gas concentrations declined over the decade, trends for sulphate and nitrate in rainfall varied in the different regions. Monitoring sites were grouped according to data alone and these groupings confirmed significant downward trends close to emission sources but little or no trend in the west and northwest, areas with high deposition from orographic enhancement of rainfall. These results indicate that recovery from acidification may take longer than expected. Investigation of the non-linearity between emission and deposition had identified issues for modelling, including the need for time- or concentration-dependent canopy resistances.

23. The Task Force welcomed and highly appreciated the work performed by the national experts. The work presented clearly showed that an in-depth scrutiny of the consistency and quality of data by national experts was indispensable before a trend assessment.

C. <u>Assessment of ozone trends</u>

24. The German Federal Environment Agency organized a workshop on ozone trends in November 2000. A short summary of the results from that workshop was presented to the Task Force. The workshop had reached the following conclusions: Long data series (> 10 years) of validated high-quality data were needed for trend assessment of ozone. To assess the effect of emission changes on ozone levels, non-anthropogenic factors like weather conditions should be filtered out. There were different ozone trends for areas with different ozone trend regimes and different indicators. Therefore, the workshop recommended including NOx and NMVOC in the trend assessment. Within some west European countries, low percentiles showed an upward trend, while high percentiles seemed to decrease.

25. For the United Kingdom, an assessment of ozone trends was conducted with special emphasis on the impact of global atmospheric transport of pollution. A nested model approach was selected to examine the differences in responses to changes in NOx and VOC emissions at selected monitoring sites. The study showed that efforts to reduce the potential for ozone formation inside Europe may be partially offset by the global ozone build-up in the future. This influence alone may be more severe for mean ozone and AOT40 exposure than for exceedances of the 60 ppb level. The future global increases in tropospheric ozone concentrations are driven by emissions both inside and outside of Europe (in Asia and North America).

26. In France, ozone simulations are made with the CHIMERE model, which covers western Europe. The model has been compared with measurements, including those from EMEP stations, and its performance appears satisfactory. It is used to forecast daily ozone concentrations over western Europe. Forecasts are routinely compared with measurements at selected sites. Using the model, a study of the sensitivity of ozone to changes in NOx and VOC emissions was conducted for different areas. It was planned to compare the model results with those from the EMEP model.

II. MODELLING AND MEASUREMENTS OF PARTICULATE MATTER

A. <u>Modelling of fine particulates</u>

27. Modelling of fine particulate matter (PM) was discussed during the first meeting of the Task Force.

1. Emission data

28. Currently available emission inventories for PM and for compounds involved in the formation of secondary PM are not satisfactory:

- Ammonia. Knowledge about emission rates as a function of time of day and season is important. The work of the ammonia expert panel under the Task Force on Emission Inventories and Projections is relevant to this and close links between Task Forces have been established;

- Heavy metals and persistent organic pollutants (POPs). Most heavy metals and some POPs are emitted as aerosols. Efforts should be made to link emission inventories for primary PM with those of heavy metals and POPs;

- **Terpenes**. Work is ongoing and improvement of emission inventories is expected within the next few years. A number of different land-use and vegetation-cover databases are currently used to estimate natural emissions by different institutions in Europe. Harmonization and linking of information in these databases would increase possibilities for developing accurate emission inventories;

- **Primary aerosols**. A Coordinated European Programme on Particulate Matter Emission Inventories, Projections and Guidance (CEPMEIP) has been set up, coordinated by EEA in close cooperation with EMEP. The work is conducted by TNO, which has prepared a speciated European inventory for 1995. CIAM is developing an emissions and costs model for primary aerosols, which will enable the calculation of sectoral emissions for both past and future years. The results of CEPMEIP are to be revised by the Parties before they can be fully used in the MSC-W, MSC-E and CIAM modelling;

- **Natural emissions from sea salt, Sahara dust, Asian desert dust, etc**. The EC Joint Research centre (JRC) has offered to provide a global inventory. The third assessment report of the Intergovernmental Panel on Climate Change (IPCC) is available at <u>www.ipcc.ch</u>.

29. It is very important that work on emission inventories is also continued after the current activities are completed. Emission inventories need to be checked and compared with models and observations.

2. Scientific approach of models

30. A number of models are currently being developed or used. Intercomparison and testing of these models is necessary:

- A workshop on model intercomparison will be organized by MSC-W in October 2001 in cooperation with the University of Helsinki. The model intercomparison will focus on a fixed time period (August 1997 and/or 1998) for which extensive field measurement data are available. Participants will use their own chemistry, emission inventory, meteorology, etc. Several EUROTRAC/GLOREAM modellers will be invited to contribute to the workshop;

- In addition to the ongoing development of long-range transport models for particles (e.g. at MSC-W), there is a need for chemical mass balance modelling. These types of models can provide complementary information on source apportionment;

- Preliminary results of optimal interpolation (measurement/modelling data) and data assimilation are encouraging and should be further developed.

3. Model output in relation to policy needs

31. The current focus on emissions and long-rang transport of particles is motivated by reports of serious effects on human health occurring at air concentrations commonly observed in many areas of Europe and elsewhere. At present it is not possible to identify the most important parameter affecting human health (total mass, specific size fraction, specific chemical speciation, etc.). For this reason, models need to be comprehensive and flexible and not focus on the output of only one parameter.

32. Cooperation of modelling activities between and within the framework of the Convention (transboundary transport) and the EC Air Quality Directives (regional to local scale) should be further strengthened. Possibilities for the nesting of models (global-regional-local scales) will be investigated.

B. <u>A monitoring programme for particulate matter</u>

33. A draft monitoring programme was discussed at the first and second meetings of the Task Force. The results of the Interlaken workshop and EMEP reports prepared since then provided the basis for the discussion.

34. The Task Force recognized that PM_{10} and $PM_{2.5}$ should now be measured at EMEP sites. The proposed method should be in line with the reference method of the European Community. Chemical characterization of the PM composition is required for the evaluation of source contribution. The purpose of chemical characterization is decoupled from mass measurements.

35. At rural sites in Switzerland, $PM_{2.5}$ amounts to about three quarters of PM_{10} . In the Netherlands, automated monitors such as the TEOM devices used in the networks, underestimate $PM_{2.5}$ and PM_{10} by 30-50% compared to the gravimetric method described in European standard EN 12341. The correction factor is a complex function of relative humidity and temperature.

36. It is important to map the spatial and temporal distribution of the concentration of nitrate aerosol over Europe. This is important for several reasons, including radiative forcing, acidification, eutrophication and because nitrate aerosol is an oxidation product of NO_x and is therefore of significant relevance for the distribution of tropospheric O_3 . The concentration of nitrate aerosol is linked to traffic emissions, and is rising in many places. Denuders are required for monitoring it. Data on concentration distributions are very sparse and are at present a weak point in the characterization of transboundary pollution that need improvement.

Speciated gas and aerosol data for HNO₃, NO₃⁻, NH₄⁺ and NH₃ are currently lacking but urgently needed. The currently used filter-pack sampling for the sum of HNO₃ + NO₃⁻ and NH₃ + NH₄⁺ may give some indirect information.

37. It should be noted that as SO_2 concentrations fall, less NH_4^+ is tied up as $(NH_4)_2SO_4$ and therefore becomes available to form NH_4NO_3 , which is more volatile. This changeover increases the complexity of PM sampling and chemical characterization.

38. A working group of the European Committee for Standardization (CEN TC264/WG15) is now organizing an intercomparison of about 10 monitors and samplers for $PM_{2.5}$ in 9 European cities as part of the development of a European standard for $PM_{2.5}$. The European reference method on $PM_{2.5}$ is expected to be published in 2004 at the earliest. It will most probably be a manual gravimetric method.

39. There is an important link between the rural and urban distributions of particulate matter. PM (and NO₂) affect health and, therefore, hence have an important urban component. On an annual basis the PM_{10} regional concentrations are comparable to the urban background levels. The long-range transported component of fine particles contributes significantly to urban PM. Urban PM is reported to the database AIRBASE hosted by the Netherlands National Institute of Public Health and the Environment (RIVM) for EEA. AIRBASE is transparent to the EMEP database at the Norwegian Institute for Air Research (NILU). Unified technical reporting and data flow is a goal for EMEP and EEA. For the link between fine particle distribution on the regional and global scales, collaboration with the WMO Global Atmospheric Watch (GAW) is important.

40. For time resolution of PM measurements in EMEP, 24 hours is realistic for manual methods. Model validation and emission assessments require higher time resolution. Higher time resolution requires automated monitors; these can be used when shown to be equivalent to manual reference method(s). To start with, a 24-hour sampling time and seven days per week measurements are recommended for PM. Later it may be possible to simplify this to 1-3 24-hour samples/week. Monitors reduce manpower requirements and improve time resolution and speed of data availability.

41. CCC has now drafted a new version of chapters 3.12 and 4.19 of the EMEP Manual for Sampling and Chemical Analysis (EMEP/CCC Report 1/95) for measurement of PM_{10} and chemical speciation of aerosol particles. The draft is based on CEN standard EN 12341. National experts are invited to review the draft available on the EMEP homepage and provide comments to CCC as soon as possible.

42. For 1999 only a few Parties reported on PM measurements: 4 countries covering 24 sites (Germany, Italy, Spain and Switzerland) reported on PM_{10} mass; 27 countries (81 sites) reported on sulphate; 20 countries (37 sites, gas/particle distribution at 21 sites) reported on nitrogen compounds; 1 country (6 sites) reported on base cation and sea salts; and no country reported on elemented carbon or organic carbon.

43. The Task Force recommends the EMEP Steering Body to adopt the draft PM measurement programme, as set out in annex I below. The programme defines three levels of

monitoring: Level 1 could eventually cover all EMEP sites, but Parties should start monitoring at least at one of their sites; Level 2 consists of a subset of 5-10 EMEP sites with a good distribution over Europe (to be recommended by EMEP in consultation with WMO-GAW); and level 3 refers to research projects and experimental campaigns.

44. To speed up the introduction of PM monitoring, CCC proposed to prepare a questionnaire on the implementation of the PM measurement programme. National experts would be invited to present their plans for PM monitoring and highlight any potential problems in implementing the minimum requirements (level 1) of the PM measurement programme. They would also be asked to present their potential contribution to levels 2 and 3 of the programme.

III. MONITORING AND MODELLING OF HEAVY METALS

A. <u>Manual for sampling and chemical analysis of heavy metals</u>

45. CCC, in cooperation with other experts and in consultation with the Task Force, drafted two new chapters to the EMEP Manual for Sampling and Chemical Analysis (EMEP/CCC Report 1/95). The objective of these chapters was to enhance the monitoring of heavy metals work in line with the objectives and requirements of the Protocol on Heavy Metals. The chapters were prepared using the work presented at, and the conclusions from, the EMEP and WMO-GAW workshops in Durham (1993), Beekbergen (1996), Moscow (1997) and Aspenäs (1997). An earlier draft had been discussed at a special session of the Task Force at its first meeting. Due to the special properties of mercury, different sampling techniques than for other metals are required. Therefore, mercury is covered by a separate chapter containing guidelines for the sampling and analysis of mercury in air and precipitation. This chapter is based on work prepared by IVL for the Commission for the Protection of the Marine Environment of the North-East Atlantic (OSPARCOM).

46. The Task Force:

(a) Agreed that these chapters were now ready for adoption;

(b) Requested CCC to add a table giving an overview of the monitoring requirements to it; and

(c) Recommended the EMEP Steering Body to adopt them for inclusion into the EMEP Manual for Sampling and Chemical Analysis and to request Parties to use this manual for heavy metals monitoring.

47. The Task Force noted that the sampling frequency proposed in the Manual might not be sufficient for some purposes. It stressed that Parties should understand that the analyses of weekly samples, indicated in the Manual as a minimum requirement, were introduced in view of the resource requirements. It recommended Parties, if possible, to conduct daily measurements.

B. <u>Modelling of heavy metals</u>

48. MSC-E has made considerable progress in developing its heavy metals models. A major objective of the work is to ensure that EMEP will be in a position to comply with the requirements of article 7, paragraph 3, of the Protocol on Heavy Metals, i.e. that, in good time before the annual session of the Executive Body, EMEP can provide information on long-range transport and deposition of heavy metals. In addition, it will follow the goals given by the EMEP Strategy for 2000-2009:

- Quantify national emissions, minimizing uncertainties;
- Verify the emission reductions of the same substances;
- Calculate transboundary fluxes, deposition and source attribution; analyse trends;
 - Contribute to research on the effects on human health and the environment.

49. The model is used to calculate deposition and air-concentration fields, country-to-country budgets, and atmospheric heavy metal input to the regional seas. Work on assessing long-term deposition trends and their relation to emission reductions has started. Most of the results are presented for each Party and available on the EMEP homepage (http://www.emep.int). The information is given in the form of maps of total deposition in a country, total deposition originating from foreign sources, and deposition from national sources to other countries. The web page also contains information on trends and the spatial distributions of emissions as well as comparisons of modelled and measured air concentration.

50. The heavy metals model is applicable for cadmium, lead and mercury and calculates atmospheric transport, deposition and chemical transformation. The present version of the model takes into account pollutant transport by large-scale vertical fluxes up to 4km high. The model deals with four mercury species: elemental mercury; gaseous inorganic oxidized mercury; particulate mercury; and dimethyl mercury. The chemical scheme used in the model is based on the Tropospheric Chemical Module (TCM), developed in the German research centre Gesellschaft für Kernenergieverwertung in Schiffbau und Schiffahrt (GKSS). The physico-chemical scheme has been significantly simplified by introducing an analytical solution for the most important processes. In addition to TCM, the decay reaction of dimethyl mercury is considered.

51. Dry deposition velocity of elemental mercury is assumed to depend on the season and the type of underlying surface. Dry deposition of particulate mercury is calculated by analogy with lead because the mass median diameters of the two particles associated with each species seem to be similar. For divalent mercury, dry deposition velocity is assumed to be constant. Dry deposition velocity of particulate compounds is parameterized using friction velocity and roughness height. Wet removal of elemental mercury follows the chemical scheme. Wet deposition parameters for gaseous divalent mercury and for particulate mercury have been determined by analogy with nitric acid and sulphate particles, respectively. For lead and cadmium a seasonal dependence of the washout ratio was introduced.

52. The model uses a 50 km x 50 km resolution of anthropogenic and natural emissions of the three metals and re-emission fields for mercury. Slight seasonal variations of anthropogenic emissions as well as distributions with height are included in the model.

53. MSC-E has started work to evaluate the reliability and accuracy of model calculations. Three types of modelling uncertainty sources are distinguished:

- Uncertainty of input data;
- Inaccuracy of physical and chemical processes parameterization;
- Limitations of model realization (numerical scheme, simplified chemistry, etc.).

54. The first two types have been assessed by means of sensitivity studies and stochastic simulation. Emissions data introduce the most significant model errors. MSC-E results indicate that using expert estimates of heavy metals emissions as input data produces more reliable modelling results than when it uses the officially submitted emission data. Modelling results are also to a great extent influenced by meteorological and scavenging parameters. Under reasonable assumptions on the input uncertainties of these latter parameters (about 40%), each of them may introduce a relative error up to 15%. The overall uncertainty due to intrinsic model parameters (excluding emissions intensity) does not exceed 25%.

55. The third class of uncertainties can be estimated by comparing results of airborne pollutant transport modelling obtained from different scientific and operational models using the same input parameters. The first such intercomparison study dealing with lead was conducted in 1996. Seven atmospheric transport models with various numerical approaches and diverse representations of physical and chemical processes were involved. The comparison showed that the discrepancy of the modelling results did not exceed a factor of two for all the models and was less than 50% for models using similar approaches (Eulerian or Lagrangian). The second intercomparison campaign took place in 1998-99 and was devoted to cadmium atmospheric transport. Four regional transport models (both Eulerian and Lagrangian) participated in the intercomparison study and have also shown satisfactory agreement of modelling results. The differences between results again did not exceed a factor of two. The mercury intercomparison campaign was launched in 2000. It consists of four stages, and is to be completed by the end of 2002. The first stage was devoted to the comparison of chemical modules of mercury transformation in a cloud environment. This was finished in February 2001. Five national airborne mercury transport models took part in the first stage. Results of the comparison showed that all of the models applied similar basic physical and chemical principles for mercury transformations.

56. The model results for lead and cadmium have been validated by comparison against observations from about a dozen measurement stations. The agreement is better than $\pm 40\%$ for air concentrations and by about a factor of two for wet depositions. Wet deposition values are somewhat underestimated by the model. Only four stations measuring air concentrations of total gaseous mercury and eight stations measuring wet deposition were involved in the validation procedure. For total gaseous mercury, the agreement between the measured and modelled concentrations is within $\pm 30\%$. Wet deposition values agree within factor of two for most of the stations.

57. The regional modelling work cannot provide the level of detail that may be desirable to examine some specific national policy questions. Such detail could be developed through national modelling work. In order to make calculations on the national level, modellers need to know the contribution of transboundary transport. Combining models of different scales can do this. Such an

approach has been applied by MSC-E in cooperation with national experts from Bulgaria, Italy, Germany and the United Kingdom.

58. The mercury modelling conducted at GKSS shows that significant progress has been made. For mercury, however, a number of questions and areas of uncertainty remain. Elemental mercury, due to its long atmospheric lifetime (> 6 months), is obviously a hemispheric or even a global problem. The process of natural emissions or the re-emission of previously deposited mercury needs to be better understood. There is still significant disagreement between modelled and measured data for oxidized mercury and the same is true for particulate mercury concentrations.

59. Heavy metals measurements have been conducted in Slovakia since 1985 at six stations, four of which are EMEP stations. The stations have participated in several intercomparison studies by EMEP and by WMO/GAW. The work is a good example of valuable measurement time series data available nationally that have not been fully used by EMEP. Parties are encouraged to make any heavy metals measurement data available to CCC and MCS-E to support the modelling work.

60. National atmospheric modelling at a fine spatial resolution (5 km x 5 km) has been conducted in the United Kingdom. The objective was to derive deposition fields that can be used to examine the exceedance of critical loads. The modelling work included eight heavy metals. It used a multi-layer Lagrangian atmospheric transport model. The model results for lead and cadmium show good agreement with deposition estimates derived from a national moss survey. Deposition maps for the deposition from foreign sources, provided for lead and cadmium by MSC-E, were very useful in defining the transboundary component. A comparison of the results with deposition maps prepared by MSC-E shows that, although totals for lead and cadmium are similar, the MSC-E model does not reproduce all high deposition areas and could be improved by using the national spatially disaggregated emissions.

61. The Task Force noted that the MSC-E model was in principle operational for lead and cadmium. Once it will have been possible to reduce the emission data uncertainties, modelling results can be applied for policy purposes.

62. The following areas for further work are highlighted:

(a) Mercury modelling should be extended to the hemispheric and, eventually, global scales;

(b) There is a need to improve the understanding of natural emissions or reemissions of previously deposited mercury;

(c) Emissions data are the most important source of uncertainty. MSC-E in collaboration with the Task Force on Emission Inventories and Projections will organize a workshop on heavy metal emission data and factors in Moscow on 21-23 November 2001. The Task Force on Measurements and Modelling requested MSC-E to present more detailed results, comparing modelling results using officially submitted data with results using expert estimates, to identify specific problem areas;

(d) There is need for better assessment of depositions, especially dry depositions;

(e) More and better measurements should receive high priority. Efforts should be redoubled to compare modelling results with all available measurement data. MSC-E should clearly define the monitoring data needs for that purpose and report back to the Task Force.

63. The acting Chairman of the Working Group on Effects informed the Task Force about progress in heavy metals impact assessment and, in particular, the progress in developing an accepted methodology for heavy metals critical load mapping. The Task Force recognized that there was a need to get a clear perspective on when the critical load data would become available in order to set the priorities for the atmospheric modelling work.

IV. MOVING FROM LANGRANGIAN TO EULERIAN MODELLING

64. The transition from Lagrangian to Eulerian modelling at MSC-W has been completed and the Eulerian model is fully operational for acidification, eutrophication and ground-level ozone. Results from both models were compared to measured data. In addition, source-receptor matrices were calculated.

65. The performance of the EMEP Eulerian and Lagrangian models has been compared with observations for the year 1996, for which data from all models are now available. The table below shows an overview of the statistical performance of the acid deposition model. The Eulerian model provides on average a better correlation with measured nitrogen compounds and ozone but not with measured sulphur compounds. The reason for the better performance is mainly related to the increased resolution (both horizontally and vertically) of the Eulerian model and improvements in the chemical scheme for nitrate. The systematic underestimation of sulphate and ammonium by the Eulerian model is a subject for further research and has consequences for the calculation of the transport of atmospheric particles. Concerning ozone, the concentrations calculated with the Eulerian model for the Mediterranean region seem more reliable than those using the Lagrangian model.

	Observed	Modelled mean		Relative bias		Annual correlation	
	Mean	(µg/m3)					
Component	(µg/m3)	Lagrangian	Eulerian	Lagrangian	Eulerian	Lagrangian	Eulerian
SO_2	1.79	2.05	2.16	+ 14%	+ 21%	84%	82%
$\mathrm{SO_4}^{2-}$	1.04	1.18	0.64	+ 13%	- 37%	80%	76%
NO ₂	1.96	1.16	1.94	+ 19%	- 1%	70%	69%
$HNO_3 + NO_3$	0.50	1.11	0.36	+122%	+ 28%	90%	81%
$NH_3 + NH_4^+$	1.24	1.48	1.04	+ 19%	- 16%	82%	83%
$SO_4^{-2}(1)$	0.60	0.51	0.46	- 15%	- 23%	61%	60%
NO ₃ -(1)	0.40	0.37	0.27	- 10%	- 33%	69%	72%
$\mathrm{NH_4}^+(\mathrm{l})$	0.50	0.34	0.32	- 32%	- 36%	75%	70%

Table: Comparing Eulerian and Lagrangian model performance (year 1996)

66. The transposition from Lagrangian to Eulerian models also has consequences for the calculations of source-receptor relationships. It is important to note that the Lagrangian models provided a valid conservative estimate of the source-receptor relationships for acidification, eutrophication and ground-level ozone. The models behave similarly in assessing source-receptor relationships but the Eulerian model generally provides higher estimates of export and import deposition. This is because the Eulerian model is mass conservative and able to allocate a larger fraction of the emissions inside the EMEP area, including also the "indeterminate deposition" derived from the Lagrangian models.

67. A study on "data assimilation" with Kalman filters and experience with the long-term ozone similation (LOTOS) model, developed within the framework of EUROTRAC, was conducted at the Netherlands consultancy firm TNO by Ms. M. van Loon and Mr. P. Builtjes. The LOTOS model is a three-dimensional Eulerian grid model, which covers Europe in grids of 0.25 latitudes x 0.5 longitude, with four vertical layers up to about 2-3 km. The aim of the study is to calculate hour-by-hour concentrations over periods of several years.

68. "Data assimilation" is a technique that combines two sources of information that both contain errors: the model results and the measurements. The technique uses errorstatistics of both sources of information to find an optimal estimate, and it does this in a physically consistent manner. In this way a better model-state is created for process studies, to improve the model, for instance its parameters, and the model input, for instance the emission data. The technique has been applied to modelled ozone and aerosol optical depth, both over August 1997. The results show that data assimilation is a working tool that provides a wealth of coherent information. Further work is needed to assess and understand the full capabilities of

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the method and TNO is seeking cooperation with other programmes, institutes and initiatives, including EMEP, Clean Air for Europe programme (CAFE), and EUROTRAC.

V. COOPERATION BETWEEN EMEP AND EIONET

69. On 30 May 2001, a joint session of the sixth EIONET Workshop on Air Quality Management and Assessment and the second meeting of the Task Force was held. The session was devoted to the harmonization and streamlining of the reporting of monitoring data in Europe. The participants were informed of the status of the European air quality monitoring network EUROAIRNET, the AIRBASE database, the AIRBASE Internet access facility AIRVIEW, and the Data Exchange Module (DEM). DEM is a data-reporting software now widely used for reporting air quality data to the European Commission and EEA.

70. EEA presented an outline proposal to streamline and harmonize European air quality reporting with the following elements:

- Using common data reporting software;
- Reporting once a year in September, synchronizing data reporting deadlines;
- Working towards one European report on air quality and deposition.

EEA announced that it was developing, in close coordination with CCC, a new version of DEM, fully compatible with EMEP data reporting. EEA intended to make it available in September 2001.

71. The Task Force agreed on the usefulness of this work and recommended that it should be pursued. Parties will receive information about the new DEM reporting tool and are invited to use DEM for data reporting to EMEP already in 2001. National experts and CCC should evaluate their experience with this new tool and report back to the Task Force at its next meeting. The Task Force recommended that the EMEP Steering Body should endorse these decisions.

72. The Task Force noted that reporting monitoring data once a year could help reduce the reporting burden on countries, particularly if reporting could be synchronized with the mandatory reporting under EC directives. CCC confirmed that a change to a single reporting of monitoring data once a year, for instance by 1 September, would not alter its reporting schedule to the EMEP Steering Body. EEA distributed a proposal for such a reporting schedule to be implemented on a voluntary basis. Parties, national experts and the EMEP centres were invited to consider this proposal for further discussion by the Task Force and the EMEP Steering Body.

VI. DRAFT TERMS OF REFERENCE AND WORK PROGRAMME

73. Taking into account the priorities for the work under the Convention adopted by the Executive Body in 1999 and the EMEP Strategy for 2000-2009 (ECE/EB.AIR/73), the Task Force adopted the proposal for its terms of reference (see annex II).

74. The Task Force was informed of the work of the ad hoc expert group on ammonia. This work is summarized in a report to the Working Group on Strategies and Review (EB.AIR/WG.5/2001/6). The expert group had discussed the link between emission control and the measurement of reduced nitrogen concentrations and deposition. It identified the problem of the difficulties detecting emission changes using monitoring data. The work showed that long-term trends (> 10 years) could be detected, whereas shorter-term (< 5 years) changes were more difficult to detect. Changes in SO₂ emissions affected the ammonia/ammonium balance and complicated detection of ammonia emission changes more. The work indicated the requirements for monitoring needed to detect ammonia emission changes, including long-term data and averaging over many sites and larger areas, and a need to increase monitoring in source regions.

75. Discussion of this work in the Task Force focused on the conclusion of the ad hoc ammonia expert group that current monitoring of reduced nitrogen concentrations was unsatisfactory, and that there was an urgent need to speciate between gaseous NH_3 and aerosol NH_4^+ . This may require reconsidering the EMEP monitoring strategy for reduced nitrogen. A possibility could be the complementary use of a low-cost denuder system to determine long-term trends at many sites. The Task Force recognized that this work of the ad hoc ammonia expert group was very relevant to its own work. It concluded that an appropriate link between this work and its own work plan should be established.

76. CCC informed the Task Force about progress in supporting POPs monitoring work. A laboratory intercomparison was performed in 2000. The results turned out to be very good for almost all compounds (with the exception of chlordane). CCC also performed a case study on α - and γ -HCH for the Baltic Sea, using the results of the POPCYCLING-BALTIC model conducted with EC funding. The modelling work illustrates well how multi-media fate and transport modelling can help to quantify the long-term fate of semi-volatile and persistent organic chemicals in a large aquatic system and its drainage basin. The Task Force welcomed the progress made through this work at CCC. It recognized that the modelling and monitoring of POPs should receive some focus in its further work.

77. CCC is drawing up a proposal for a new EMEP data-flagging system consistent with the system used by WMO/GAW. It presented the modifications already finalized and its plans for further changes. It will inform national quality assurance managers about the new system once it is finalized and will invite experts to comment on the draft. The Task Force recognized the importance of a good and clear (easily understandable) data-flagging system. A change in flagging of past data may be very demanding, but is important for the work on the assessment report. CCC will keep the Task Force informed about progress.

78. The Joint Research Centre (JRC) in Ispra, Italy has taken the initiative to create and promote a platform for know-how and information exchange on air pollution monitoring and related research activities in the Mediterranean area. The Task Force recognized that this initiative could be very useful for the further work of the Task Force and of EMEP in general and suggests to the EMEP Steering Body to consider the proposal for cooperation in this initiative.

79. The Task Force on Measurements and Modelling discussed its work for the coming three years, outlining the following main tasks:

(a) Foster cooperation between the EMEP centres, other bodies within the Convention, the Parties and national and international research activities;

(b) Investigate the trends in transboundary fluxes, concentrations and depositions over the lifetime of EMEP in different regions, making use of measurements and modelling results; in particular, the Task Force will:

- (i) Assist countries in the application of tools to assess their data;
- (ii) Contribute to the preparation of a report summarizing the topics mentioned ('Assessment Report');
- (iii) Coordinate the input by national experts to the preparation of the report;

(c) Investigate further possibilities for the combined use of modelling results and measurement data;

(d) Provide a forum for technical/scientific discussions of work done within CCC, MSC-E and MSC-W;

(e) Review the current measurement strategy in all five thematic fields of EMEP:

- Acidifying and eutrophying substances;
- Photochemical air pollutants and its precursors;
- Particulate matter;
- Heavy metals; and
- Persistent organic pollutants.

Based on this review, the Task Force will, as appropriate, draw up proposals for adapting the measurement strategy to the needs within the Convention.

80. As a matter of priority the Task Force suggests focussing on the preparation of the assessment report, and on measurements and modelling of POPs, ammonia and particulate matter. It will further discuss the harmonization of reporting of monitoring data to different forums in Europe, and, as a first step, review the revised version of DEM and the reporting deadlines harmonization with EIONET.

81. The next meeting of the Task Force is scheduled for early 2002.

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<u>Annex I</u>

Level ^{#)}	1	2	3
	All EMEP sites,	5-10 EMEP sites, start	Research projects
	start in 2001, time	in 2002 ⁺⁾ , time	campaigns, time resolution
Parameter	resolution 24h	resolution 24h	at least once per 2-3hours
PM ₁₀	CEN 12341		
PM _{2.5} ⁺⁺⁾	CEN 2.5 ⁺⁺⁾		
PM _{1.0} ⁺⁺⁾			
Number size			(SMPS/OPC) Scanning
distribution			mobility particle sizer/optical
			particle counter
$SO_4^{=}, a$	EMEP manual		
NO_3 , a	EMEP filter pack	Denuders and filter pack	
$\mathrm{NH_4}^+$, a	EMEP filter pack	Denuders and filter pack	
Soluble base cations	Water extraction [*])		
$(Na^+, K^+, Ca^{2+}, Mg^{2+})$			
and plus Cl ⁻)			
Mineral dust		Method to be specified	
Elemental carbon (EC)	Quartz fibre filter ^{**)}		
	central analysis lab		
	(e.g. at CCC)		
Organic carbon (OC)	Analysed at CCC		OC specification and higher
			time resolution
Chemical speciation as			Steam-jet aerosol collector
a function of particle			(SJAC)
size			
Light scattering			Nephelometer ^{***)}

RECOMMENDED PM MONITORING PROGRAMME IN EMEP

^{#)} Level 1: could eventually cover all EMEP sites, but Parties should start monitoring at least at one of their sites; Level 2: a subset of 5-10 EMEP sites with a good distribution over Europe (to be recommended by EMEP in consultation with WMO-GAW); and level 3: research projects and experimental campaigns.

- ⁺⁾ To be recommended by EMEP in consultation with WMO-GAW.
- ⁺⁺⁾ Expected European reference method in 2004 (CEN TC264/WG15). PM_{2.5} and PM_{1.0} cannot start now and the recommendation depends on the revision of the EC daughter directive in 2003.
- *) Need special procedure description for filter handling to avoid contamination (addendum to EMEP Manual). Base cations and Cl cannot be analysed at all EMEP sites, but should at least be done when EC and OC are sampled.
- **) EC-OC instrument based on heating sample to 650K, conversion to CO₂, and infrared detection at selected sites throughout Europe. Sampling at least once a week.
- ***) Nephelometers can in principle be operated on a routine basis at a few sites and can also be classified as level 2 measurement.

Annex II

DRAFT TERMS OF REFERENCE

- 1. The Task Force on Measurements and Modelling:
 - (a) Supports the EMEP Steering Body and its Bureau by:
 - (i) Reviewing and assessing the scientific and operational activities of EMEP related to monitoring and modelling;
 - (ii) Evaluating their contribution and support to the effective implementation and further development of the Protocols;
 - (iii) Drawing up specific proposals for the EMEP work-plan;

(b) Provides for closer collaboration of Parties to the Convention, the EMEP centres, other bodies under the Convention, other international bodies and the scientific community in strengthening scientific communication and cooperation in air pollution monitoring and modelling.

2. The Task Force reports to the EMEP Steering Body and holds regular meetings.