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Abstract/Sammandrag

A pilot project is presented, where the dispersion model MATCH is tested for studies of dispersion and deposition of lindane (γ-HCH) over Europe. Comparisons between rough model calculations and long-term measurements at stations around the Baltic Sea indicate a positive correlation both for concentration in air and for wet deposition, but the model results are a factor of 3-10 larger than the measurements. The MATCH model simulations seem to agree better with observations from the Swedish West-coast and North Sea areas. Since the model calculations in this pilot project are performed in a very simplified way we cannot exclude that uncertainties in our calculations can cause the detected differences. However, we think that also emission data, especially for Sweden and Finland, and the respresentativity of measurements should be investigated further in a future study.

Key words/sök-, nyckelord

Lindane, POP, dispersion, model, concentration, air, precipitation, deposition, measurements, Europe, Baltic Sea, Kattegat

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1. INTRODUCTION

Lindane (γ-HCH) is a pesticide which has been, and to some extent still is used in Europe. It is a compound which is regarded as a Persistent Organic Pollutant (POP), which means that it is comparatively resistant to photolytic, biological and chemical degradation. For POPs the degradation time scale is much larger than the transport time scale. POPs also have a tendency to accumulate in living organisms, which can cause toxic effects. Several and quite extensive Dutch studies have recently presented estimates of emissions of lindane, which have been combined with long-term mean dispersion calculations (Hout, 1994), (Baart, Berdowski and Jaarsveld, 1995), (Pul, Jaarsveld and Jacobs, 1996) and (Jaarsveld, 1996).

This project is aimed as a pilot project, where the dispersion model MATCH is used in a first test for studies of POPs. We have tried to simulate, with a time resolution of 1-h, the dispersion of lindane over Europe for four different 10-day periods during 1993 and 1994. The main purpose has been to calculate the concentration in the air, assuming two different emission scenarios, and then compare the obtained results with measured 6-h mean values of lindane in the air at 5 stations in the Baltic States and one station in southern Sweden. Based on model results from the four studied periods also rough estimates of annual mean concentration in air and annual deposition have been obtained and some crude comparisons are made to long-term measurements of lindane.

Lindane has been chosen since it is one of the POPs for which, at least to some extent, estimates of emission data are available. It is important to note that in this pilot study, only a very simplified treatment of the reemission processes from the ground and sea surface has been included.

2. SHORT DESCRIPTION OF DISPERSION MODEL

The MATCH (Mesoscale Atmospheric Transport and CHemistry) model has been developed at SMHI as a tool for air pollution assessment studies on different geographical scales, see e. g. (Persson, Langner and Robertson, 1995 and 1996) and (Langner, Persson and Robertson, 1996). Model versions covering Europe, Sweden and subregions of Sweden have been used as a basis for decision making concerning environmental protection in Sweden. MATCH is an Eulerian atmospheric dispersion model, including physical and chemical processes governing sources, atmospheric transport and sinks of oxidised sulphur and oxidised and reduced nitrogen. With the MATCH system, air pollution contributions from different source types like traffic, industry, shipping, farming etc. can also be obtained. In this study we have tried to apply the European scale MATCH-model to the dispersion of lindane over Europe.

2.1 Transport and chemical degradation

The model version used here has eight layers in the vertical. The first layer has a height of 75 m and the thickness of the layers above increase with height. The top of the model is taken to be about 7 km. It is easy to add additional layers if necessary, but for this application eight layers was judged to be sufficient. The horizontal resolution for the calculations is 55x55 km.

Horizontal advection is calculated using a fourth order flux correction scheme (Bott 1989a, 1989b). Vertical advection is calculated using an upstream scheme. Vertical transport is also induced by turbulent vertical diffusion and the spatial and temporal variations of the mixing height. For the numerical solution of the combined horizontal and vertical transport, degradation and deposition an operator split time integration scheme is used.

For lindane, being a POP, the degradation time scale is much larger than the atmospheric transport time scale and should be of little importance for the present study. Anyhow, we have applied a degradation constant for lindane in the air (2.5×10^{-7} s^{-1}), which is equal to the value used by Pul et al (1995).

2.2 Wet deposition

Wet scavenging of the different species in the MATCH model is proportional to the precipitation rate and a species specific scavenging coefficient. For lindane, which has a relatively high solubility in water, we have in this study used a value of 1.4×10^{-5} (s^{-1}) at a precipitation rate of 1 mm/h, which is a value close to what has been used in the Dutch studies refered to above. We are, however, aware of that much more complex chemical and physical processes may have influence on the wet deposition and should be investigated in future studies.

2.3 Exchange with soil and water

The MATCH model has, so far, only been developed for pollutants where no reemission processes from the ground have to be considered. This means that the dry deposition is proportional to the concentration in the air and a species specific dry deposition velocity at 1 m height above ground. Since the lowest model layer has a height of 75 m, the dry deposition flux calculation is transformed to the middle of that layer using standard similarity theory for the atmospheric surface layer.

Lindane and many other POPs are semi-volatile at normal atmospheric temperatures. Thus lindane can be deposited to the ground and water surfaces and later reemitted to the atmosphere again. Thus the deposition should be calculated as a net-deposition, which is the sum of the (downward) deposition and the (upward) reemission fluxes. These questions have been studied in some detail by (Pul, Jaarsveld and Jacobs, 1995) and (Jaarsveld, 1995), where the lowest layer in their dispersion model describes the concentration in the soil and water surfaces below the model atmosphere.

In the present preliminary study using the MATCH model, we have not included a separate description of the reemission, but rather used a simplified net-deposition velocity for lindane which is assumed to describe both the dry deposition and the reemission. The net-deposition velocity value to ground has in this study been put to 0.03 cm/s. For water surfaces a much higher value, 1.5 cm/s, has been used, since the deposition of lindane to water is mainly controlled by the atmospheric resistance (Pul, Jaarsveld and Jacobs, 1995). These net-deposition velocities used should be of the right order of magnitude for northern Europe, but for soils with a large content of lindane the net-deposition value can be much smaller, even negative.

3. METEOROLOGICAL DATA

The MATCH dispersion model requires meteorological data to calculate transport and deposition processes. Also for the trajectory calculations, discussed below, meteorological data are needed. In this study all meteorological data have been obtained from weather analyses and forecasts within the European scale HIRLAM model, which is run in routine at SMHI. The grid resolution for HIRLAM is in this case 55x55 km. Meteorological fields are available for every 3rd hour and interpolated to every hour. It is important to note that also precipitation data are obtained from HIRLAM, which causes a somewhat larger uncertainty in the calculated wet deposition compared to the use of a large number of precipitation stations over the model area.

4. EMISSION

Emission data for lindane are very uncertain. For this study we have therefore made model calculations for two different emission scenarios. In scenario-1 yearly emission data for each country was obtained from Baart, Berdowski and Jaarsveld (1995), assuming a total emission of 1930 tonnes in Europe for the year 1990. A rough geographical distribution of the emission within each country was then obtained assuming proportionality between the lindane emission and the EMEP-database for agricultural emission of ammonia (Sandnes, 1993). The assumed geographical distribution of lindane emission in scenario-1 is shown in Figure 1. According to Voldner and Li (1995) the usage of lindane was banned in the three Baltic states in 1992 and severely restricted in the rest of the former Soviet union. In emission scenario-2 we have therefore assumed no emissions from the Baltic States and only 10% of the earlier emission from the rest of the former Soviet union, while all other countries in Europe have the same emission as in scenario-1. The total assumed lindane emission from Europe in scenario-2 is about 1060 tonnes and the geographical emission distribution is shown in Figure 2. No influence from emission of lindane outside Europe is considered.

The application of lindane, mainly in farming, is assumed to be concentrated to spring and early summer. Therefore, in the model, the main part (65%) of the yearly emission to the atmosphere is assumed to take place during April-July. The rest of the yearly emission is assumed to be emitted slowly from the ground surface with the lowest emission rate during winter. It is, however, important to note that no positive net reemission of lindane, originating from earlier years deposition, is included in the calculations in this study. We have assumed no daily variation of the emission, although in reality the emission probably is larger during day than night.

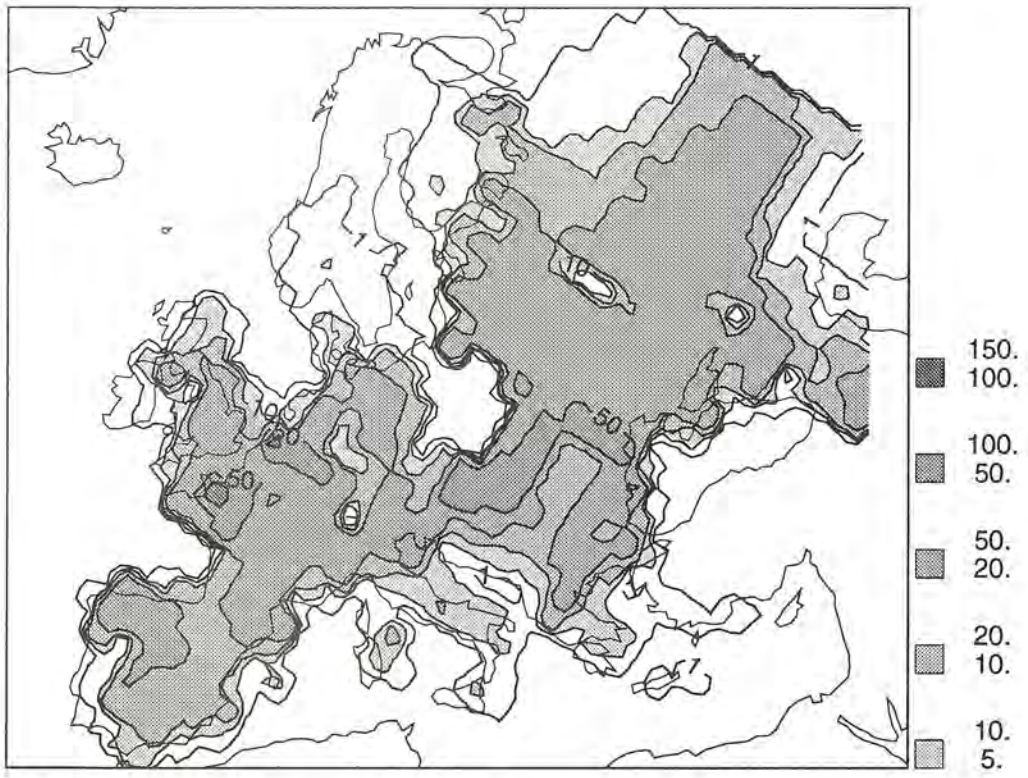


Figure 1. *Geographical distribution of lindane emission (10 g/km^2) assumed in scenario-1 (see text). Total emission in Europe is 1930 tonnes/year.*

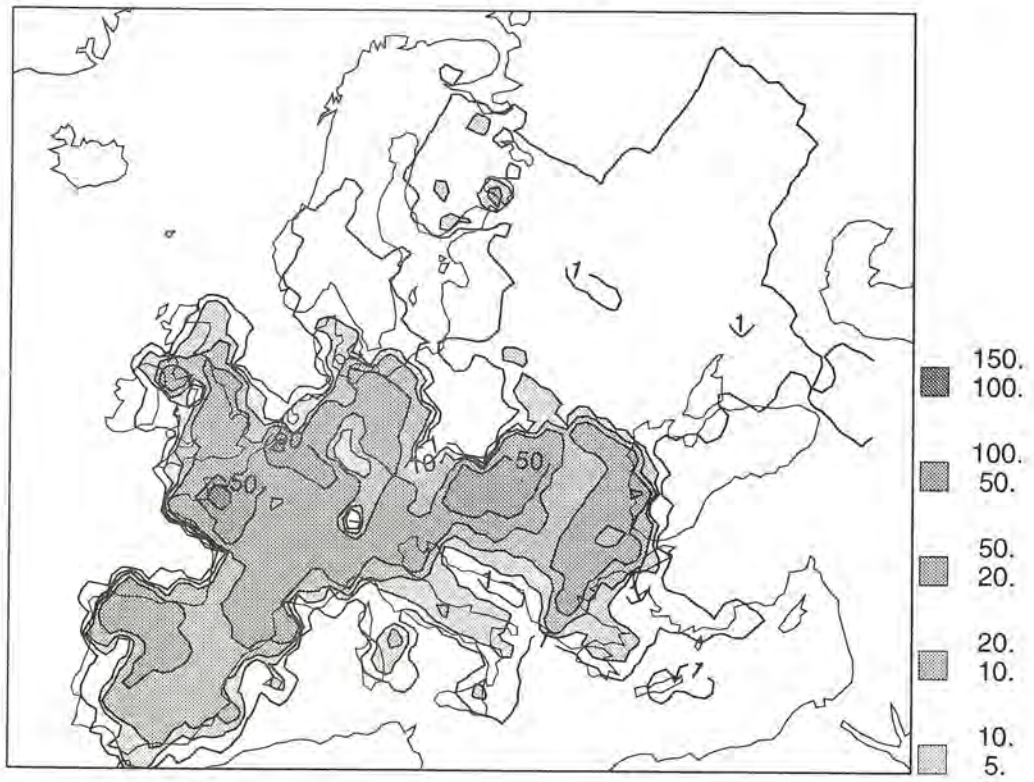


Figure 2. *Geographical distribution of lindane emission (10 g/km^2) assumed in scenario-2 (see text). Total emission in Europe is 1060 tonnes/year.*

5. MEASUREMENTS OF LINDANE IN AIR AND PRECIPITATION

Lindane measurement data from five different studies have been used in our model comparisons.

a) Short-time (6-h) air concentration values from five stations in the Baltic States and one station in southern Sweden have been measured by Department of Chemical Ecology, University of Lund (Agrell, pers. com.). Sampling have been performed during four 6-day periods: 15-20 November 1993, 15-20 February, 15-20 April and 15-20 July 1994. Samples were taken between 9-15 UTC (day-values) and 21-03 UTC (night-values) during each day. So far, analysed lindane concentrations are available only for the November and February periods.

b) Measurements of long-term mean values of concentration in air and wet deposition at 16 stations (see Figure 3) around the Baltic Sea made by Department of Chemical Ecology, University of Lund (Agrell et. al., 1996 submitted). The measurements have been performed for a number of 2-week periods covering about 12 months for each station during 1991-1993. Somewhat different periods have been used for the different stations. Annual values have been calculated from these measurements. For wet deposition the annual value for each station was based on the measured median value, since it was regarded as more representative (Agrell priv. com., 1995) than the mean value.

c) Lindane concentrations from snow samples taken on the ice of the Bothnian Bay during winter 1991 (Rahm et. al., 1995).

d) Lindane concentrations in precipitation during 1990 (PARCOM, 1991) for some stations around the North Sea.

e) Measurements of long-term mean values of concentration in air and wet deposition at 4 stations on the Swedish West-coast towards the Kattegat (Brorström-Lundén, 1995). The measurements have been performed during a number campaigns of 1-2 weeks length, at different seasons, during the years 1991-1994. Annual mean values have been estimated from the measurements.

6. TRAJECTORY CALCULATIONS

A trajectory describes the transport path of a certain air parcel and can be used to trace sources of contaminated air arriving at air pollution measurement stations. In order to support the work with the analyses of lindane, SMHI made trajectory calculations in real time twice a day during the four measurement periods under a) above. Trajectories, with a length of 72 hours, arriving 00 and 12 UTC each day to the stations Vilsandi, Salaspils and Torna-Hällestad were calculated and sent to the University of Lund. In the discussion below some examples of calculated trajectories are shown.

7. RESULTS AND DISCUSSION

MATCH model simulations have been performed for four different 10-day periods: 11-20 November 1993, 11-20 February 1994, 11-20 April 1994 and 11-20 July 1994. The first two days in each period have been used to spin-up the MATCH model, only results from the following days have been used in the study.

We show model results obtained from applications of emission scenario-1 as well as scenario-2. But, since emission scenario-2 seems to be most realistic, below we only discuss simulations for emission scenario-2 in the comparisons with measurements.

Figure 3. *Map showing air and precipitation stations for lindane, which have been used in the model comparisons for annual values. The stations have been run by Department of Chemical Ecology, University of Lund (Agrell et. al., 1996 submitted).*

7.1 Lindane concentration in air

It can be of interest to compare concentrations of lindane in the air based on scenario-1, scenario-2 and measurements. In Figure 4 diagrams are showing the MATCH-calculated time-dependent concentrations of lindane, based on emission scenario-1, at three different stations. In Figure 5 similar diagrams are presented for emission scenario-2 together with observed lindane concentrations where measurements are available (November and February). The calculated concentration values for the stations in the Baltic States are, in most situations, much lower for scenario-2 compared to scenario-1. This is obviously caused by the much smaller emissions in eastern Europe for scenario-2. Comparisons between measurements and model simulated concentrations for scenario-2 show that at Torna-Hällestad, in southern Sweden, measurements and model results are of the same order of magnitude for the November period, while the model simulations exceed the measurements in February. For stations in the Baltic States the model concentrations are a factor of 3-10 larger than the observed lindane concentrations in the air, and there seems to be only a weak correlation between measured and model simulated concentrations. Unfortunately, no measurements are available for April- and July-periods when the emissions are assumed to be larger than in winter.

It can be of some interest to compare calculated trajectories with dispersion model simulations. In Figure 6 examples of trajectories are shown for 20 April 1994. The trajectories indicate a transport of air from rather important lindane source areas in northern Germany towards the station in southern Sweden, while clean air from northern Scandinavia is transported over the Baltic Sea (with no assumed emission) towards the stations in the Baltic States. In Figures 4 and 5 the model simulated concentrations are presented for that situation, showing a fast increase in southern Sweden at the same time as the simulated concentration at Vilsandi, on an Estonian island in the Baltic Sea, is very low. The calculated concentrations at Salaspils in Latvia depend to a large extent on the assumed emissions within the Baltic States, since the air reaching the station has been transported over parts of Lithuania and Latvia. Other large variations in time for the calculated concentrations, which are seen in Figures 4 and 5, can be explained in a similar way by transport from different source areas. Also variations in turbulent mixing and other meteorological conditions play of course a role for the obtained model concentrations.

Calculated mean concentration of lindane in air for each of the studied periods, as well as rough estimates of the annual mean concentrations in air, are shown in Figures 7 and 8. The annual mean values are obtained in a simplified way, assuming an extrapolation of the four studied periods.

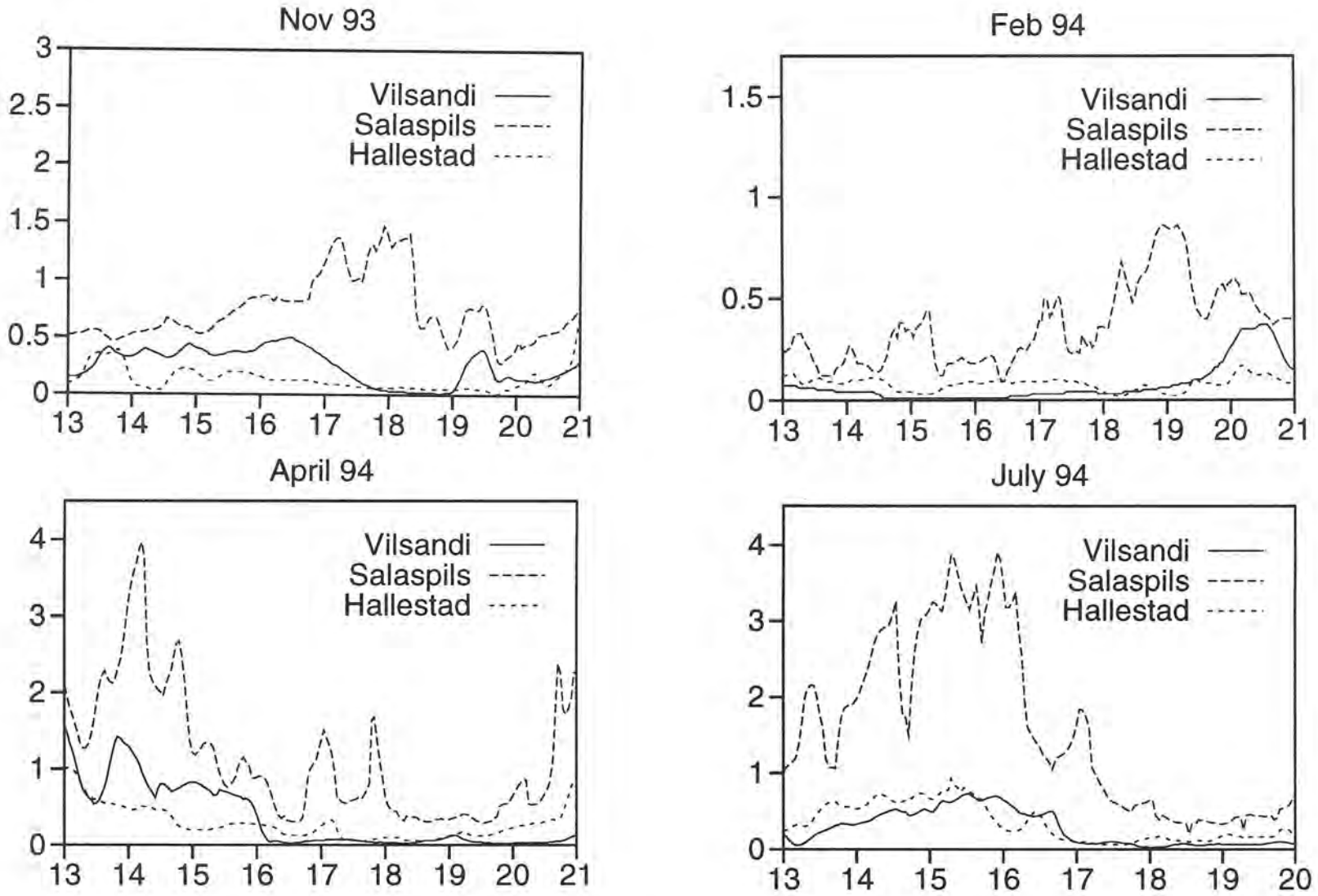


Figure 4. *Diagrams showing the calculated time-dependent concentrations of lindane (ng/m^3) during the studied periods, based on emission scenario-1, at the three stations Vilsandi (Estonia), Salaspils (Latvia) and Torna-Hällestad (southern Sweden).*

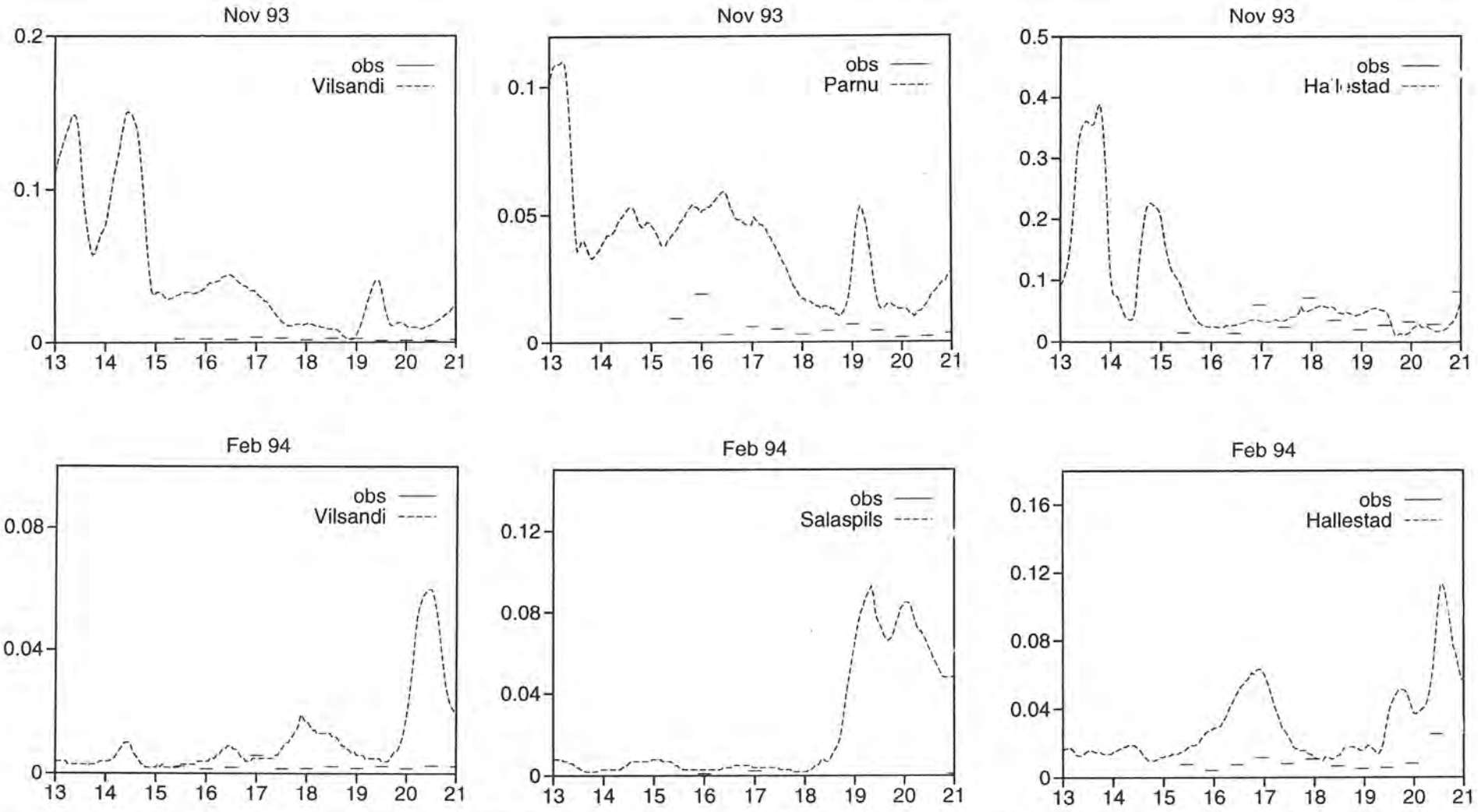


Figure 5. *Diagrams showing the calculated time-dependent concentrations of lindane (ng/m^3) during the studied periods, based on emission scenario-2, at the stations Vilsandi (Estonia), Salaspils (Latvia) or Pärnu (Estonia) and Torna-Hällestad (southern Sweden) together with available observed (obs) lindane concentrations.*

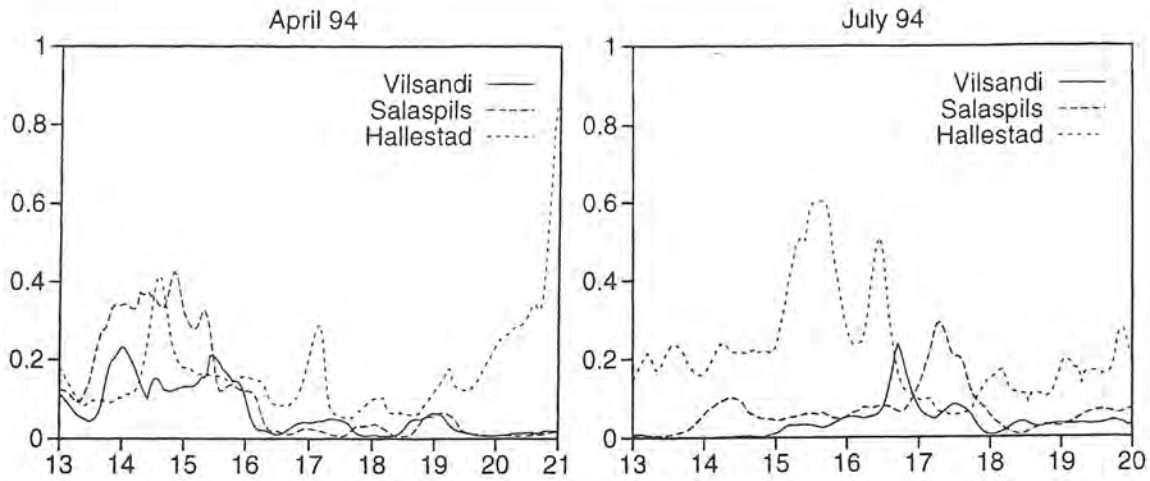


Figure 5. cont.

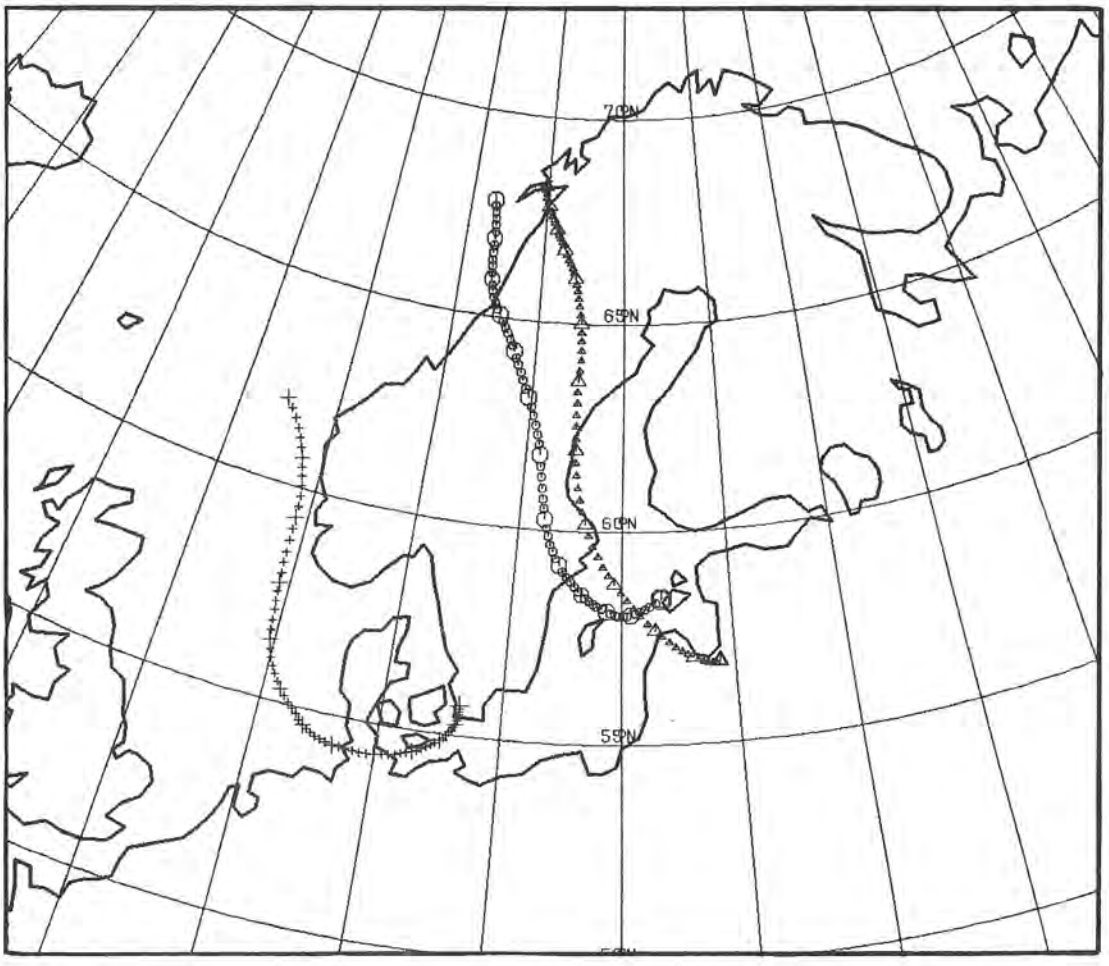


Figure 6. *Calculated trajectories showing 72h transport arriving at the stations Torna-Hällestad (southern Sweden), Vilsandi (Estonia) and Salaspils (Latvia) on 20 April 1994, 12 UTC. The trajectories are calculated for a height of about 650m.*

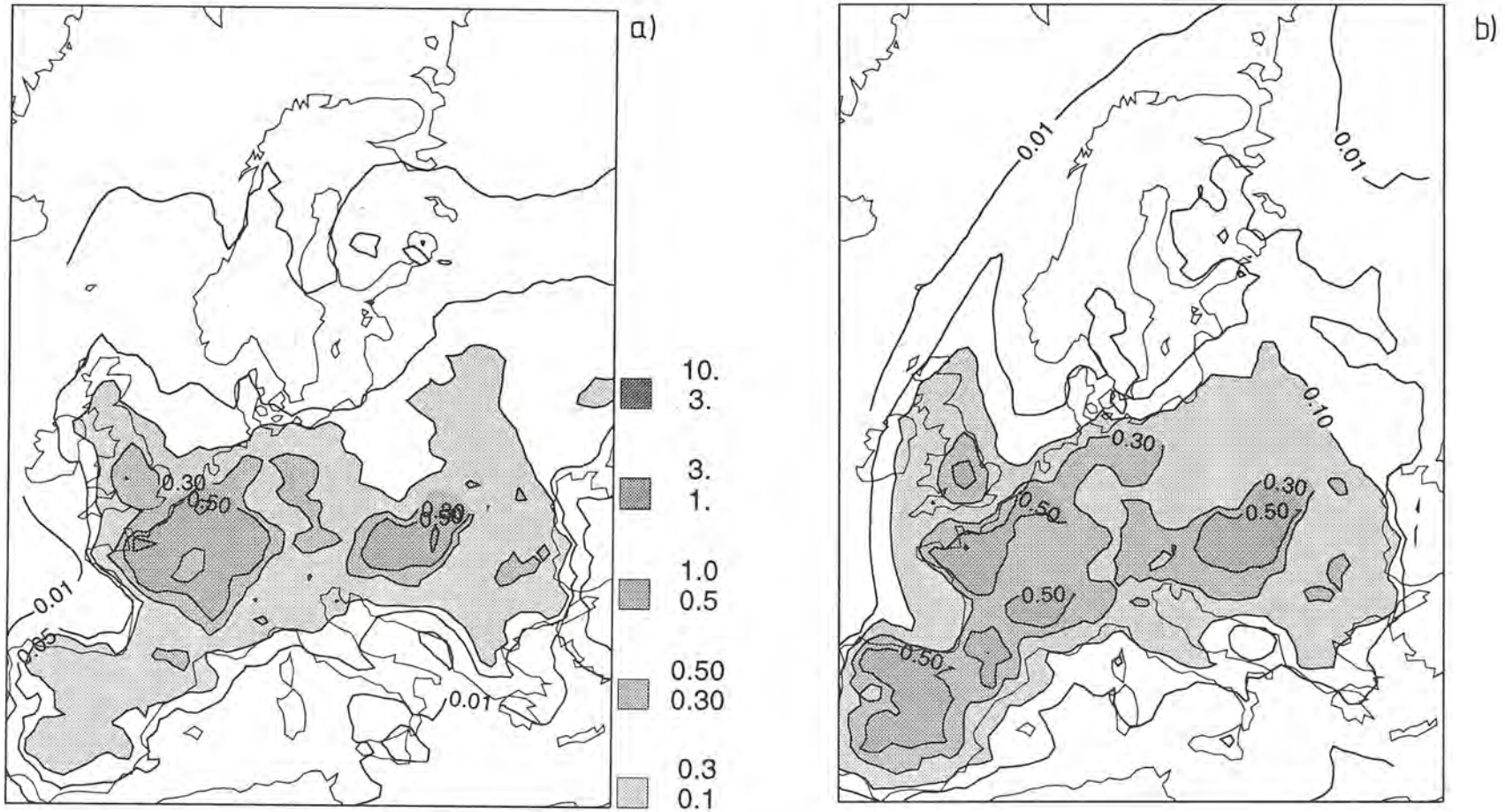
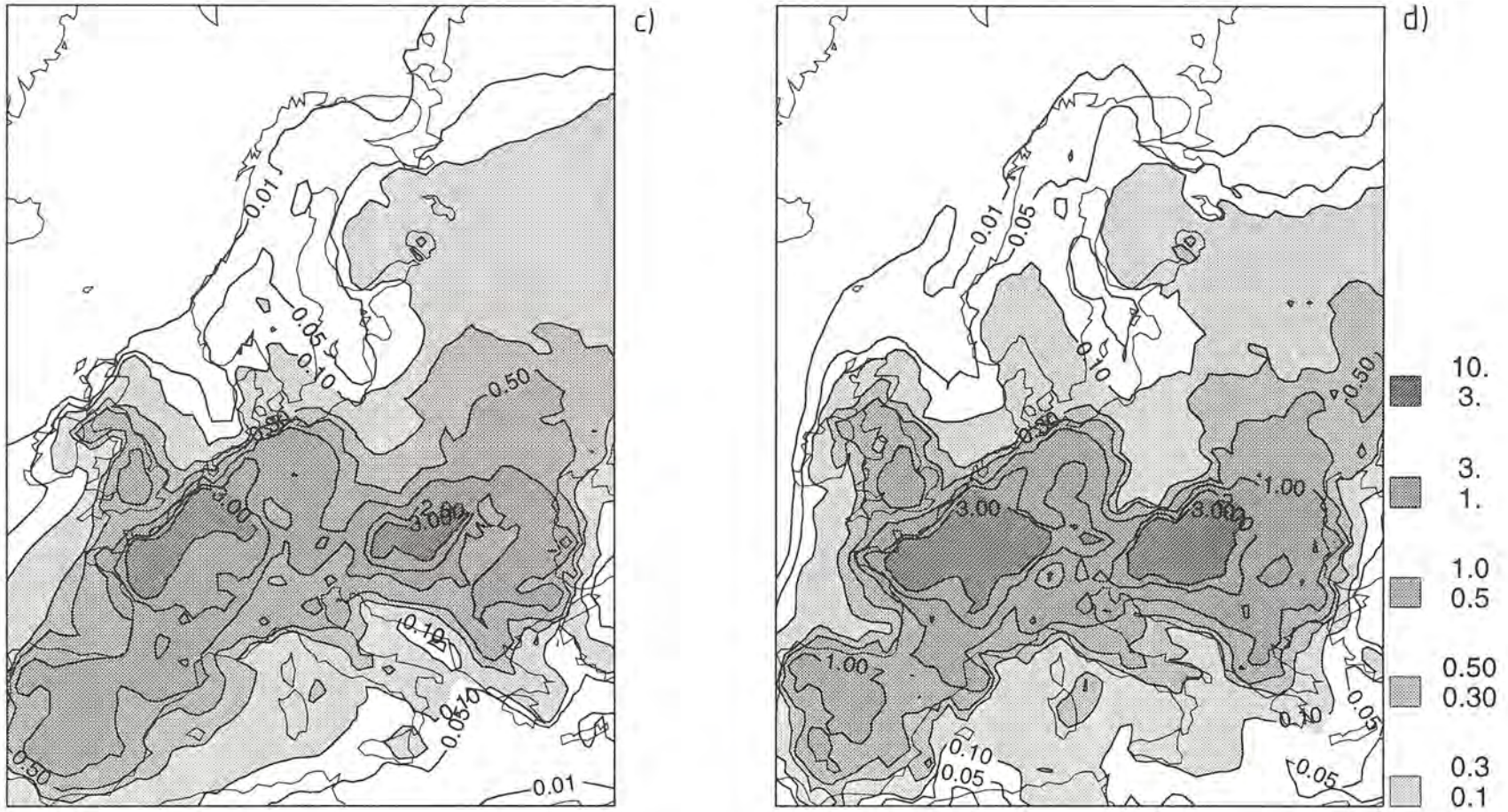


Figure 7. *Calculated mean concentration of lindane in the air (ng/m^3), assuming emission scenario-2, for each of the four studied periods (dates 15-20 each month). a) November 1993, b) February 1994, c) April 1994 and d) July 1994.*



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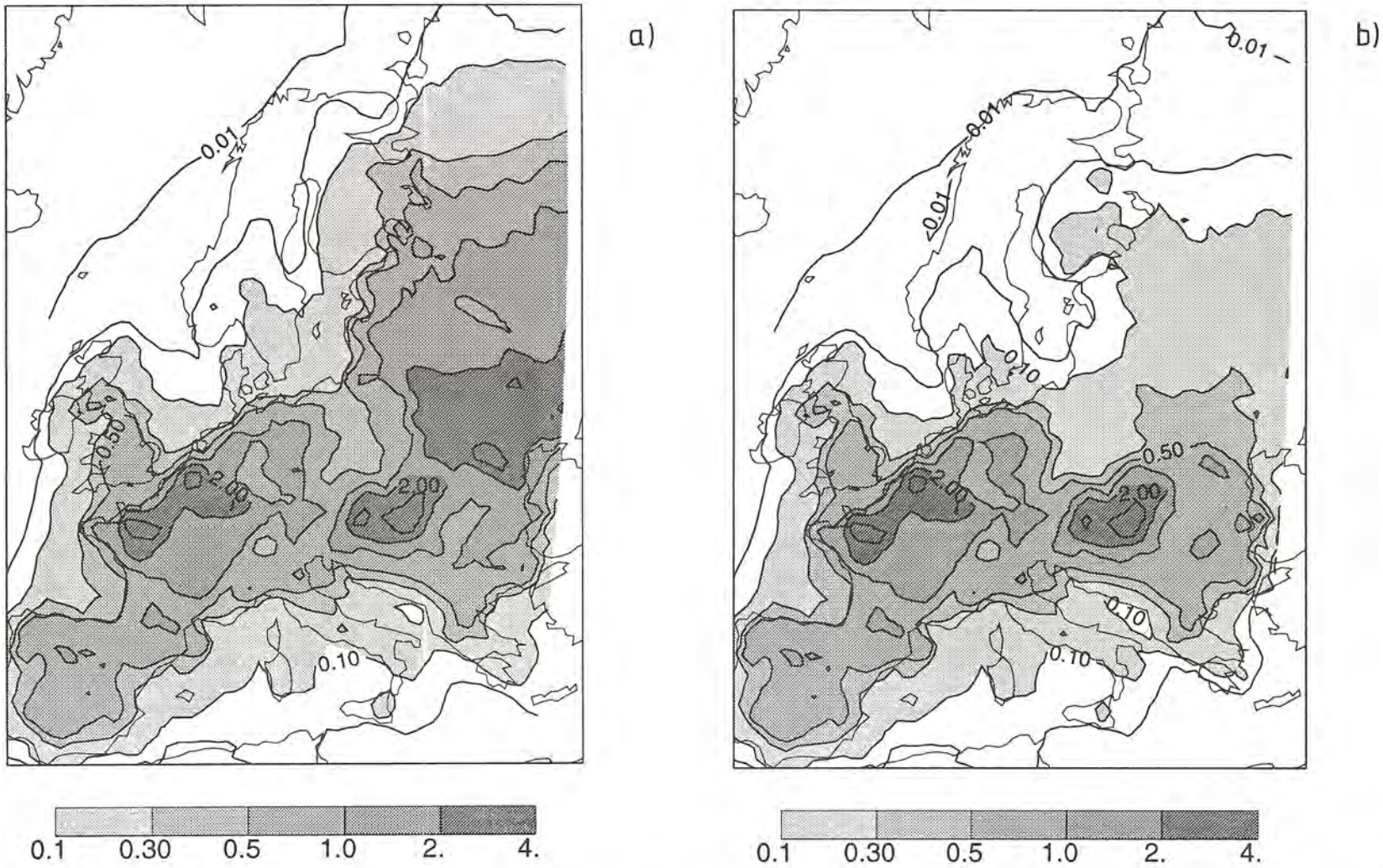

Figure 8. *Rough model estimates of the annual mean concentrations of lindane in the air (ng/m^3). Results for a) emission scenario 1 and b) emission scenario 2.*

Comparisons between rough model calculated (emission scenario-2) and measured annual mean concentrations in air are shown in Figure 9. For the 16 stations around the Baltic Sea (Agrell et. al., 1996 submitted) there is a positive correlation between measurements and model results, but with about a factor of 3 larger values in the model. The reported measurements from the Swedish West-coast (Brorström-Lundén, 1995), with mean concentrations of 0.12 ng/m^3 at Rörvik/Nidingen and 0.06 ng/m^3 at Gårdsjön, are close to the model estimate of about 0.1 ng/m^3 using emission scenario-2 (see Figure 8).

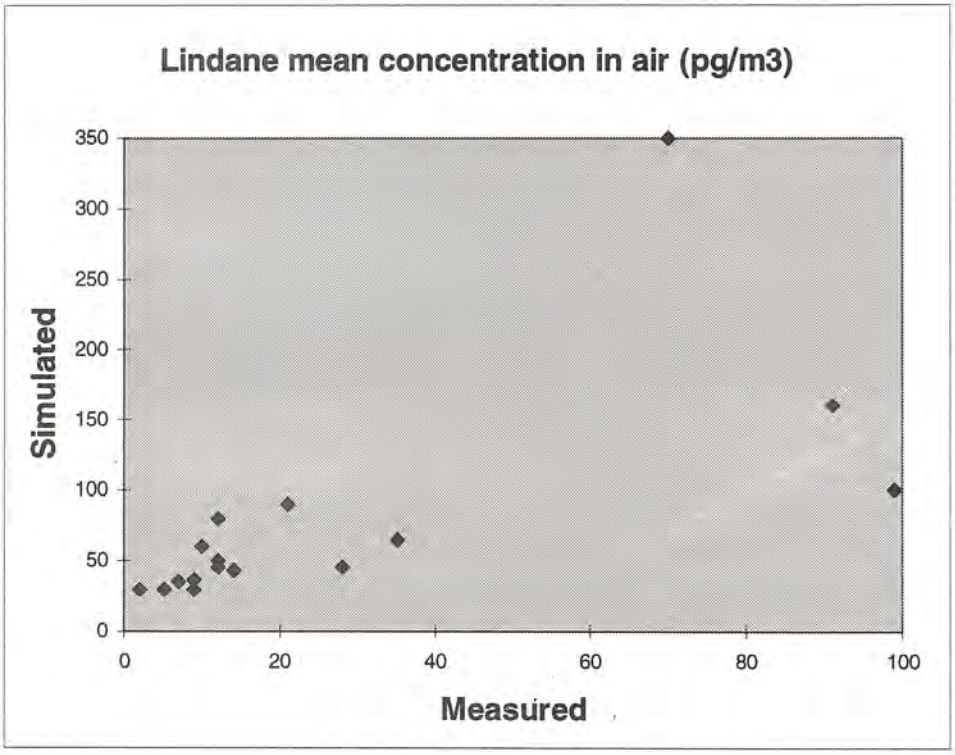

Figure 9. *Scatter plot comparing annual mean values for measured and model (emission scenario-2) simulated concentration in the air (ng/m^3) around the Baltic Sea.*

7.2 Lindane deposition

In detailed studies of the net deposition of lindane, the reemission processes should be taken under careful considerations. However, in this pilot project the reemission is only considered through the use of a very simplified "net" dry deposition velocity (always positive). Still, it can be of some interest to present rough model estimates for the wet and total (wet+netdry) deposition of lindane and compare the simulated wet deposition values with available observations. For these estimates the calculated periods have been extrapolated to give the deposition for a whole year.

Estimates of the total annual deposition are shown in Figure 10 for emission scenarios 1 and 2. In Figure 11 a rough model estimate of the annual wet deposition is given for emission scenario-2. The four short periods, for which we have made model simulation, have been extrapolated to a whole year. We have then compared these calculated annual wet deposition values with observations from 13 stations around the Baltic Sea (Agrell et. al.,1996 submitted), although the measurements are not performed for the same periods as the model calculations. Figure 12 shows a scatter plot where the observed annual wet deposition values are compared with model simulations. There is a relatively strong positive correlation between measured and simulated wet deposition, with small wet deposition values around the north of the Baltic Sea (northern Bothnian Bay) and much larger values along the southern coast of the Baltic Sea. However, the model simulations give roughly a factor of 10 higher values than the observations.

Rough comparisons of the annual wet deposition can also be made for four stations on the Swedish West-coast (Brorström-Lundén, 1995). The measurements give annual values of 1.2-4.4 $\mu g/m^2$, with the lowest value at station Koster in the northern part of the area. The model estimates (scenario-2) of 1.5-3 $\mu g/m^2$ are close to those values and also the model indicates lower values to the north.

The comparisons between model calculated and measured wet deposition, which are given above, have a limitation since we have no explicit precipitation information available for the lindane measurement stations. Also for the model calculations it is difficult to obtain representative estimates of the annual precipitation amounts. These uncertainties in the precipitation amounts are probably of the order of a factor of two.

In many cases it is better to compare estimates of concentration in precipitation rather than wet deposition. But in this study we have to assume normal annual precipitation values at each lindane measuring station, since we have no explicit measured precipitation amounts available. Concentration estimates, based on measurements (Agrell et. al., 1996 submitted) around the Baltic Sea, show mean concentrations in precipitation in the range 0.1-2 ng/L, while our model simulations, for emission scenario-2, give concentrations of 3-16 ng/L in precipitation for the same geographical area. Thus the values based on measurements are about a factor of 10 lower than the model estimates. A mean lindane concentration of 0.16 ng/L was reported from snow samples on the ice on the Bothnian Bay during late winter 1991 (Rahm et al, 1995), which is also about a factor of 10 lower than estimated from our model calculations.

Based on measurements (Brorström-Lundén, 1995) for the Swedish West-coast area, annual mean concentrations in precipitation are in the range 2-7 ng/L, which is very close to our model simulations (emission scenario-2) of about 5 ng/L for that area.

Pul et al (1995) have made comparisons between model calculations and observed lindane concentrations in precipitation during 1990 (PARCOM, 1991) for stations around the North Sea. Both simulated and observed data were in the same range (3-60 ng/L), but the correlation was reported weak. MATCH model results from the present study, for emission scenario-2, indicate yearly mean concentrations in precipitation in the range 2-40 ng/L around the North Sea, thus of the same order of magnitude as given by Pul et al (1995).

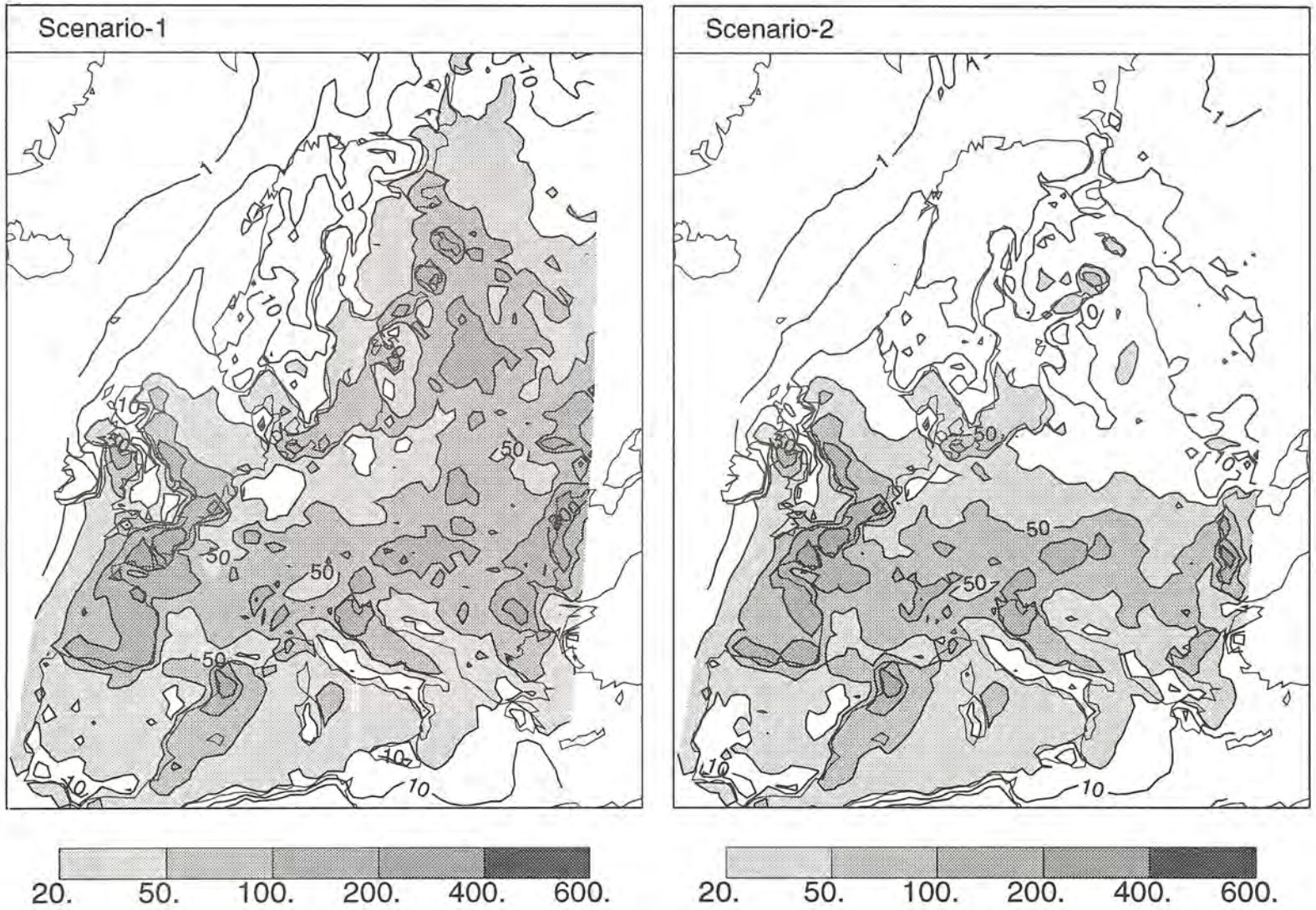

***Figure 10**. Simplified model estimates (emission scenarios 1 and 2) of the total annual deposition of lindane (µg/m²). Isolines for 1, 3, 5, 10, 20, 50 and 100 µg/m² are given.*

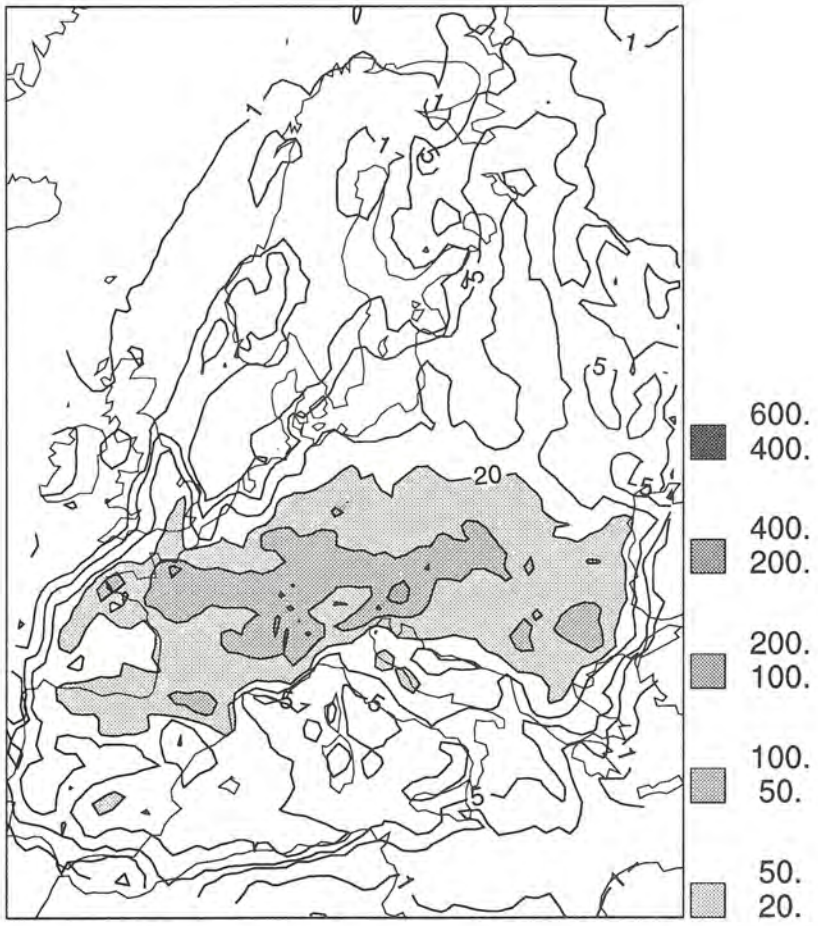

Figure 11. *Simplified model estimate (emission scenario-2) of the annual wet deposition of lindane (μg/m²). Isolines for 1, 3, 5, 10, 20, 50 and 100 μg/m² are given.*

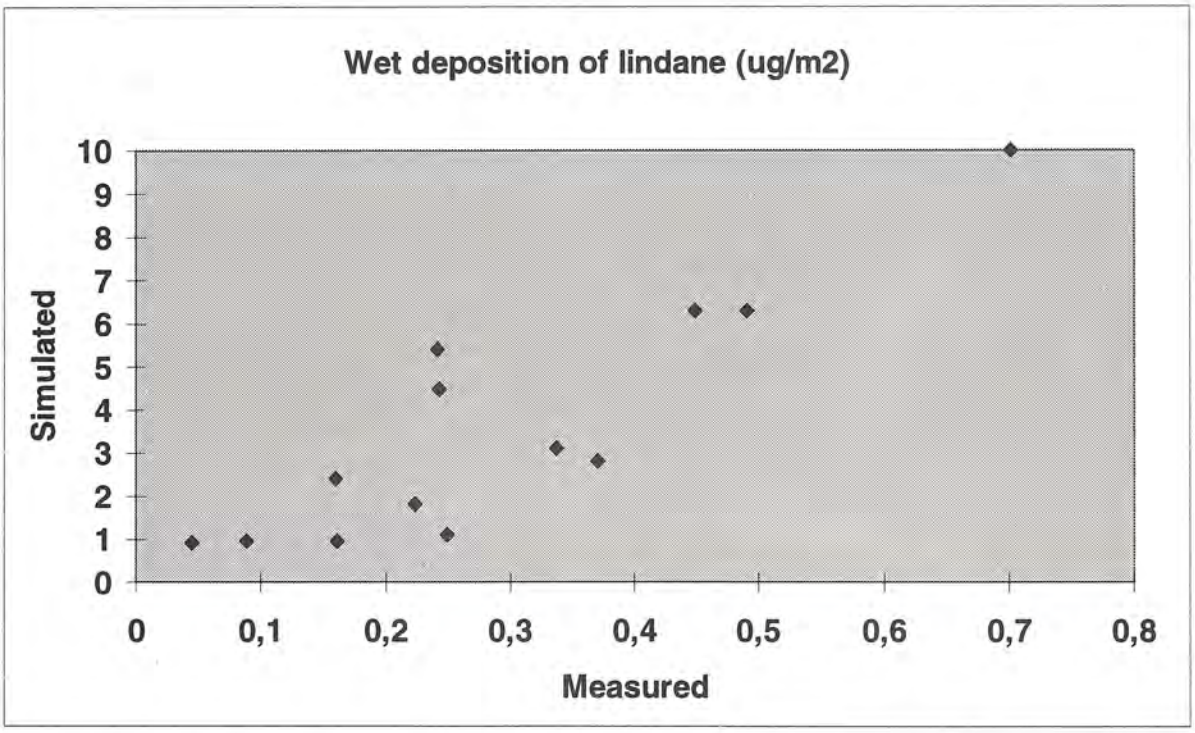

Figure 12. *Scatter plot comparing annual values for measured and simulated wet deposition of lindane (μg/m²) around the Baltic Sea.*

8. TENTATIVE CONCLUSIONS

This pilot study indicate that MATCH model calculations (based on emission scenario-2), give lindane concentrations in air and in precipitation, which are correlated to measurements made by Agrell et. al. (1996 submitted) in the Baltic Sea area during 1991-1994, but about 3-10 times larger than the observations. The MATCH model results seem to agree better with observations from the Swedish West-coast (during 1991-94) and North Sea (during 1990) areas.

Since the model calculations only include four 8-day periods and also the measurements are limited in time, any firm conclusions can not be drawn. We cannot, at this stage, exclude that uncertainties in our calculations of e.g. chemical degradation, deposition, reemission, precipitation and atmospheric transport can cause the detected differences. Especially, the difficulties to obtain representative estimates of precipitation amounts from both model calculations and measurements causes uncertainties in the comparisons for lindane concentration in precipitation and for annual wet deposition. However, since the agreement between measurements and model simulations are better for the Swedish West-coast and North Sea areas, we think that also emission data, especially for Sweden and Finland, and the representativity of measurements should be investigated further in a future study. In the emission data base valid for the year 1990, which has been used in this study, the annual lindane emission is assumed to be 4 tonnes from Sweden and 12 tonnes from Finland. A substantially smaller emission from these two countries should certainly reduce the simulated values in the Baltic Sea area. In a future study also detailed precipitation information should be added to the lindane measurements and the model simulations.

Acknowledgements

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S-60176 Norrköping, Sweden. Tel. +46 11 15 80 00. Telex 64400 smhi s.

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