8. Atmospheric Supply of Lindane to the Baltic Sea in 2002

This chapter presents a short description of model evaluation of lindane atmospheric input to the Baltic Sea, its sub-basins and catchment area in 2002. Modelling of lindane atmospheric transport and depositions was carried out using MSC-E Eulerian multimedia POP transport model MSCE-POP (Shatalov et al., 2004). Latest available official information on lindane emission from HELCOM countries and other European countries was used in computations. Based on these data levels of annual and monthly lindane depositions to the Baltic Sea region have been obtained and their monhly variations are estimated. Model results were compared with observed levels of lindane concentrations in air and precipitation measured at monitoring sites around the Baltic Sea.

8.1 Lindane emissions

To evaluate long-range transport and depositions of lindane to the Baltic Sea area computations were carried out for the period 1990-2002. Computations for long period were carried out to take into account long-term accumulation of lindane in soil and sea water. Expert estimates of POPCYCLING-Baltic project (Pacyna et al., 1999) and available emission data officially submitted by EMEP countries to the UN ECE Secretariat were used. In particular, available official data on lindane emissions and expert estimates based on officially reported usage of lindane were included in computations. For countries, which reported emissions only for some years, emissions for missing years were interpolated between available official data or expert estimate or were taken equal to the last available emission value. Expert estimates of POPCYCLING-Baltic project are ended with 1996. Therefore for countries, which have not submit their emission data for 1997-2002, lindane emission was taken equal to the value of expert estimate for 1996.

Usage of lindane in most of European countries in recent decade was banned or restricted (Breivik et al., 1999). However official information on lindane emission including absence of its application for most of the countries is lacking. Following officially reported data lindane emission to air from Denmark, Finland, and Sweden was equal to zero in period 1990-2002. Due to the lack of official data for Latvia, Lithuania, and Poland the expert estimates (Pacyna et al., 1999) were used in computations. As in computations for the previous report no lindane emissions is assumed in Russian Federation from 1997. Latest available official data from Germany contain time-series of lindane emission from 1990 to 2002. According to this information lindane emission from Germany decreased from 14.9 tonnes in 1990 to 3.6 tonnes in 1997. Starting from 1998 lindane emission of Germany was equal to zero. Zero emission of lindane in 2002 is reported for Estonia. Total values of lindane emission of HELCOM countries and total emission within the EMEP region for 2002 are presented in the Table 8.1.

Table 8.1. Annual emissions of lindane in the HELCOM countries and in the entire EMEP area, used in computations for 2002. Units: tonnes per year

Country	2002
Denmark	0
Estonia	0
Finland	0
Germany	0
Latvia	0.003
Lithuania	0.002
Poland	0.28
Russian Federation	0
Sweden	0
TOTAL – HELCOM countries	**0.285**
TOTAL - EMEP	**64**

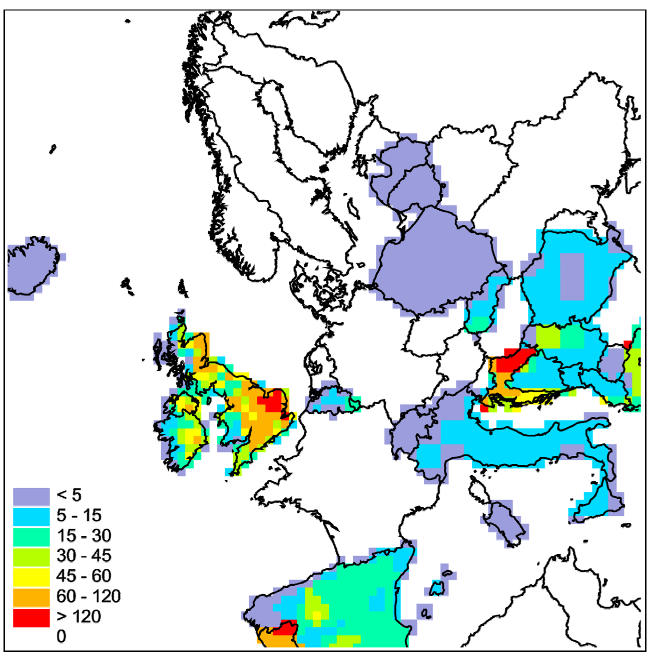


Figure 8.1. Spatial distribution of lindane emission in the EMEP domain in 2002 with resolution 50x50 km^2. Units: $g/km^2/year$

Following officially submitted information the most significant emissions of lindane outside the HELCOM region belong to the following countries: the United Kingdom (17 tonnes/year), Portugal (13 tonnes/year), Spain (10 tonnes/year), and Croatia (7 tonnes/year).

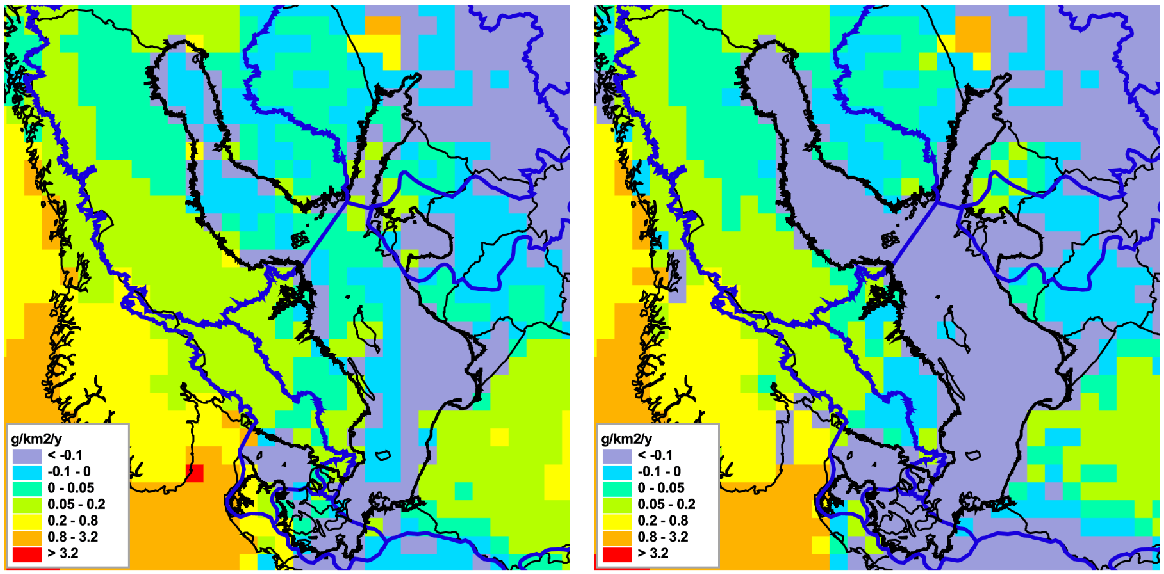
Some information on emissions submitted by countries to the UN ECE Secretariat in 2004 differed significantly from expert estimates and data in literature (Pacyna et al., 1999; Breivik et al., 1999). In particular, France reported that HCH not occurred in the period 1990-2002. However, there was other information that France was the most significant user of lindane in Europe until 1998. Therefore to take this into account in model computations two emission scenario was collected for 1990-2002: the first one assuming no use of lindane in France in period 1990-2002, and the second one with lindane emissions for France based on the expert estimates (Pacyna et al., 1999). The difference between these two runs and the influence of France emissions will be discussed below.

For modeling of lindane transport it was assumed that its application varies over the year and is concentrated in the growing season. Seasonal variation of lindane emission to the atmosphere is assumed to have maximum in spring and earlier summer. Following this the annual lindane emission is didtributed in following way: 10% of annual emission – in February, 15% - in March, 25% - in April-June each.

8.2 Annual deposition of lindane

Annual and monthly depositions and exchange of lindane between the air and the underlying surface for 2002 were computed for six sub-basins and six catchments of the Baltic Sea. Spatial distribution of net gaseous flux of lindane over the Baltic Sea region for 2002 is given in Figure 8.2 for two scenarios described above. The removal of lindane from the atmosphere occurs due to wet deposition and gaseous exchange which can be directed from air to the underlying surface (dry deposition) and backward (re-emission). In latter case accumulated lindane re-volatilizes back to the atmosphere. Net gaseous flux represents the sum of wet deposition and gaseous flux which is the sum of dry deposition flux, directed downward from air to the underlying surface, and re-emission flux, directed upward from the underlying surface to air. Net gaseous flux can be negative or positive depending on the values of both fluxes. Negative values of the flux indicate that re-emission process takes place.

As it can be seen the most significant differences in net depositions are obtained over the Baltic Sea. The sea and soil compartments in the second scenario run have accumulated more lindane which is resulted in more significant re-emission from seawater and also from soils. Negative values of net gaseous flux can be noted for all sub-basins of the Baltic Sea. In model run for the first scenario the re-emission from the Baltic Sea is less pronounced though it takes place in southern part of the Baltic Proper sub-basin (BAP).



(a) (b)

Figure 8.2. Spatial distribution of lindane net gaseous flux over the Baltic Sea region for 2002 obtained in model run for the first scenario (a) and for the second scenario (b) with resolution 50x50 km^2. Units: g/km^2/year

Tables 8.2 and 8.3 present the distribution of total lindane depositions to the Baltic Sea sub-basins and catchment area in model run for the first scenario. Tables 8.4 and 8.5 present the same distribution in model run for the second scenario. In tables along with the total depositions, values of deposition fluxes and contributions of wet and net dry depositions are given. Comparing these four tables it can be seen that the most significant differences are obtained for the dry depositions over the Baltic Sea sub-basins. Over the catchment area the difference in dry depositions is smaller. Wet depositions in computations for the second scenario are approximately 30% higher than in the second scenario run both for the Baltic Sea and its catchment area. Re-emission fluxes of lindane are dominant for all sub-basins and its catchment area with the maximum in the Belt Sea sub-basin (BES). These differences in deposition fluxes between two model runs are caused by the influence of lindane accumulated in sea and soil compartments.

Table 8.2. Annual wet deposition (Wet), gaseous exchange (Dry), sum of wet deposition and gaseous exchange (Total) of lindane (kg/year) and net gaseous flux (g/km^2/year) to the Baltic Sea sub-basins in 2002 in model run for the first scenario

	GUB	GUF	GUR	BAP	BES	KAT	Baltic Sea
Wet	18	9	6	66	8	9	116
Dry	-265	-123	-82	-719	-116	-88	-1393
Total	-247	-114	-76	-653	-108	-79	-1277
Flux	-2.1	-3.8	-4.1	-3.1	-5.3	-3.4	-3.0

Table 8.3. Annual wet deposition (Wet), gaseous exchange (Dry), sum of wet deposition and gaseous exchange (Total) of lindane (kg/year) and net gaseous flux (g/km^2/year) to the Baltic Sea catchment area in 2002 in model run for the first scenario

	GUB	GUF	GUR	BAP	BES	KAT	Catchment area
Wet	88	91	34	233	10	32	489
Dry	-876	-699	-283	-1510	-108	-247	-3724
Total	-788	-608	-249	-1277	-99	-215	-3235
Flux	-1.6	-1.4	-1.8	-2.3	-3.8	-2.5	-1.9

Table 8.4. Annual wet deposition (Wet), gaseous exchange (Dry), sum of wet deposition and gaseous exchange (Total) of lindane (kg/year) and net gaseous flux (g/km^2/year) to the Baltic Sea sub-basins in 2002 in model run for the second scenario

	GUB	GUF	GUR	BAP	BES	KAT	Baltic Sea
Wet	24	12	8	88	10	11	152
Dry	-484	-210	-140	-1412	-268	-181	-2695
Total	-460	-199	-132	-1324	-258	-170	-2543
Flux	-3.9	-6.6	-7.2	-6.3	-12.5	-7.3	-6.0

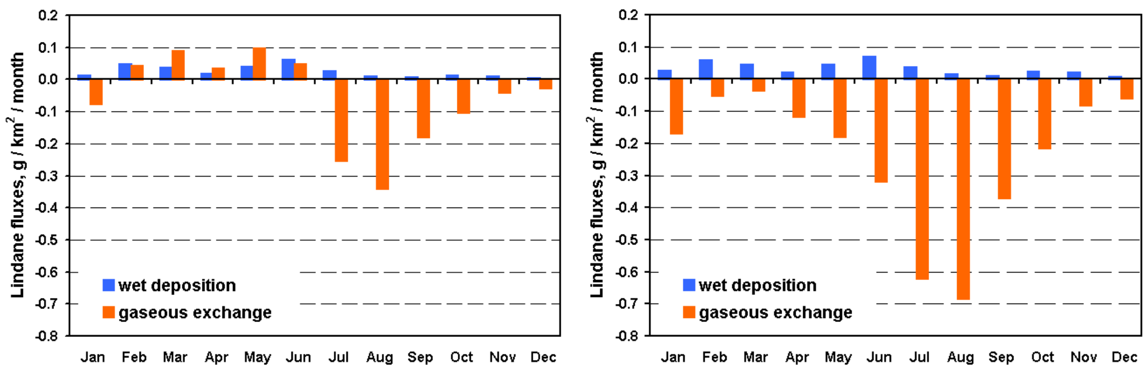
Table 8.5. Annual wet deposition (Wet), gaseous exchange (Dry), sum of wet deposition and gaseous exchange (Total) of lindane (kg/year) and net gaseous flux (g/km^2/year) to the Baltic Sea catchment area in 2002 in model run for the second scenario

	GUB	GUF	GUR	BAP	BES	KAT	Catchment area
Wet	113	120	45	281	13	39	611
Dry	-1190	-941	-391	-2062	-210	-346	-5141
Total	-1077	-821	-346	-1781	-198	-307	-4530
Flux	-2.2	-2.0	-2.5	-3.2	-7.6	-3.6	-2.6

8.3 Monthly depositions of lindane

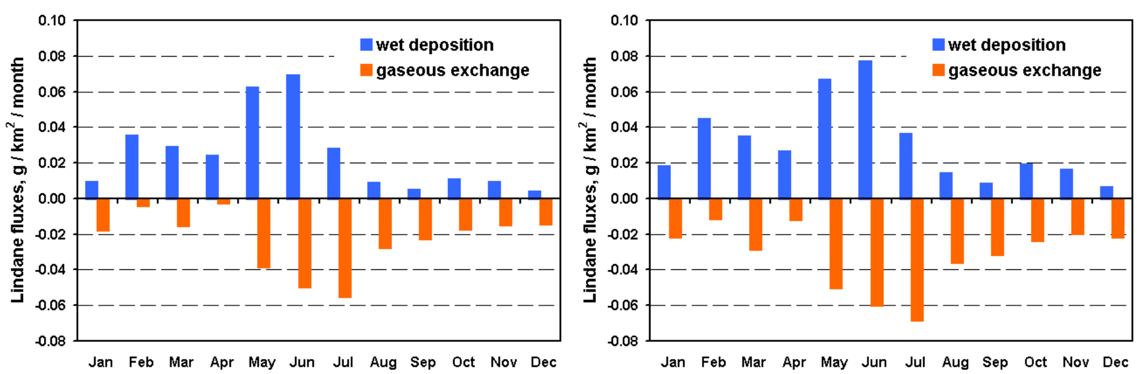
Monthly variations of lindane wet deposition and gaseous exchange fluxes for the Baltic Sea and its catchment area in computations for 2002 are shown in Figures 8.3 and 8.4 respectively. These two figures compare monthly variations obtained in model runs for two scenarios of lindane emissions in period 1990-2002.

Gaseous exchange fluxes of lindane over the Baltic Sea sub-basins are more intensive comparing to wet deposition fluxes. Over the Baltic Sea catchment area wet deposition fluxes are more significant comparing to gaseous exchange fluxes over soil.



(a) (b)

Figure 8.3. Monthly variations of lindane wet deposition and gaseous exchange fluxes over the Baltic Sea in computations for 2002 run I (a) and run II (b), g/km^2/month.



(a) (b)

Figure 8.4. Monthly variations of lindane wet deposition and gaseous exchange fluxes over the Baltic Sea catchment area in computations for 2002 run I (a) and run II (b), g/km^2/month.

As it can be seen re-emission fluxes over the Baltic Sea obtained in the model run for the second scenario are almost twice higher than the results for the first scenario. Over the catchment area the difference in fluxes of these two runs is about 30%.

8.4 Comparison of model results with measurements

Model results obtained in computations were compared with available measurements of lindane concentrations for 2002 of EMEP monitoring stations. Measurements of lindane for 2002 were reported by Westerland (DE01), Zingst (DE09), Lista (NO99), De Zilk (NL91), Knokke (BE04), CZ03 (Kosetice), Pallas (FI96), SE14 (Råö), and Aspvreten (SE12). Comparison of measured and computed lindane concentrations and their ratios are presented in Table 8.6.

Table 8.6. Comparison of observed mean annual lindane concentrations in air and precipitation with computed concentrations for 2002 in both scenarios of emission

Station code	Station name	Observed	Calculated I	Calculated II
Lindane concentrations in air (pg/m^3)				
CZ03	Kosetice	27.1	21.3	27.6
NO99	Lista	12.4	15.2	20.0
FI96	Pallas	7.3	3.4	4.4
SE14	Råö	11.6	14.5	23.7
SE12	Aspvreten	12.7	10.0	14.0
Lindane concentrations in precipitation (ng/l)				
BE04	Knokke	7.12	0.59	0.82
DE01	Westerland	2.41	0.60	0.76
DE09	Zingst	1.55	0.45	0.62
NL91	De Zilk	6.78	0.71	0.92
NO99	Lista	1.68	0.56	0.63
Lindane deposition flux (ng/m2/y)				
FI96	Pallas	161	154	201
SE14	Råö	26	363	447
SE12	Aspvreten	310	222	286

It should be noted that for model verification it is of importance to use parallel obsrvations of concentrations in air and in precipitation performed at the site. Among selected sites parallel measurements were carried out only at Lista (NO99), Pallas (FI96), Råö (SE14), and Aspvreten (SE12). The latter three sites presented the results of precipitation measurements as deposition fluxes which were formed by wet deposition and partly by dry deposition process. Therefore it resulted in the additional uncertainty in the comparison with modelling results.

In comparison with observed mean annual air concentrations of lindane at selected stations computed concentrations are slightly lower in model run for the first scenario and higher by almost 30% in model run for the second one. Concentrations of lindane in precipitation measured at Knokke (BE04), Westerland (DE01), and De Zilk (NL91) are significantly higher than computed values. The reason for this difference can be connected with the influence of local sources not taken into account in the emission data and also of residues of lindane in soil. Deposition fluxes measured at Pallas and Aspvreten are close to computed values of wet deposition fluxes. At the same time the difference for Råö (SE14) is rather high which can be caused by the uncertainties of lindane emission data as for 2002 and also for previous years and by the uncertainties in model parameterization.