8. Atmospheric Supply of Lindane to the Baltic Sea in 2001

Atmospheric input of lindane to the Baltic Sea and its catchment area, was evaluated for 2001 using MSC-E Eulerian multimedia POP transport model MSCE-POP. The MSCE-POP model is used for the computations of long-range transport and exchange with underlying surface within the EMEP region of selected POPs with horizontal resolution 50x50 km^2 (Shatalov et al., 2003). This chapter presents a short description of lindane emission data used for modelling, computed annual depositions of lindane to the Baltic Sea sub-basins and catchments along with their monthly variations. Obtained results were compared with available monitoring data of lindane concentrations in air and precipitation in the Baltic Sea region.

8.1 Lindane emissions

Evaluation of lindane long-range transport and depositions to the Baltic Sea area was based on computations for the period 1990-2001. Computations for long period were carried out to take into account long-term accumulation of lindane in soil and sea water. Modelling was performed using the expert estimates of POPCYCLING-Baltic project (Pacyna et al., 1999) and available emission data officially reported by EMEP countries to the UN ECE Secretariat. In particular, beginning with 1990 available official data on lindane emissions and expert estimates based on officially reported usage of technical HCH or 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 taken equal to last available emission value. Expert estimates of POPCYCLING-Baltic project are ended with 1996. Therefore for countries, which have not submited their emission data for 1997-2001, lindane emission was taken equal to the value of expert estimate for 1996.

According to available publications the usage of lindane in most of European countries in recent decade was banned or restricted. 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 the atmosphere in Denmark, Finland, and Sweden was equal to zero in period 1990-2001. Due to the lack of official data for Estonia, Latvia, Lithuania, and Poland the expert estimates (Pacyna et al., 1999) were used for these countries. In Russian Federation the usage of lindane was prohibited in 1990. In spite of prohibition, remaining quantities of lindane in agriculture, forest and municipal economy were used until 1996 (Fedorov and Yablokov, 1999, List of chemical and biological …, 1994). Thus, the use of lindane in Russia starting from 1997 was assumed to be zero. The emission of lindane in the European part of Russia in 1990 was estimated equal to 462 tonnes on the basis of the data on pesticide application in this region (Revich et al., 1999). For subsequent years of the period 1990-1997 it was assumed that lindane emission of Russian Federation linearly decreased.

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 from Germany was equal to zero. However, it should be noted that these data were not available at the moment of calculation with MSCE-POP model for this report. In model runs for the period 1990-1993 expert estimates (Pacyna et al., 1999) were used and for the subsequent years previously reported official German emission for 1994, equal to 15 tonnes, was applied. New time-series of lindane emissions will be used in modeling for the next Joint EMEP Report for HELCOM.

Total values of lindane emission from HELCOM countries and total emission within the EMEP region for 2001 are given in the Table 8.1. Additional information on lindane emissions for other years in the period 1990-2001 can be found in the Annex E.

Table 8.1. Annual emissions of lindane in the HELCOM countries and in the entire EMEP area for 2001. Units: tonnes per year

Country	2001
Denmark	0
Estonia	0.003
Finland	0
Germany	0*
Latvia	0.003
Lithuania	0.002
Poland	0.28
Russian Federation	0
Sweden	0
TOTAL – HELCOM countries	**0.288**
TOTAL - EMEP	**101**

* - latest available officially submitted data. It should be noted that computations of lindane transport and depositions with MSCE-POP model for this report were done on the basis of previously reported data (Annex E).

Outside the HELCOM region the following countries are the most significant emitters of lindane: the United Kingdom (27 tonnes/year), Portugal (13 tonnes/year), Spain (10 tonnes/year), and Belgium (9.6 tonnes/year).

For model computations it is assumed that the use of lindane varies over the year and is concentrated in the growing season (from February to June). Seasonal variations of lindane emission to the atmosphere connected with its application mainly in spring and earlier summer are applied in the model (10% of annual emission – in February, 15% - in March, 25% - in April-June each).

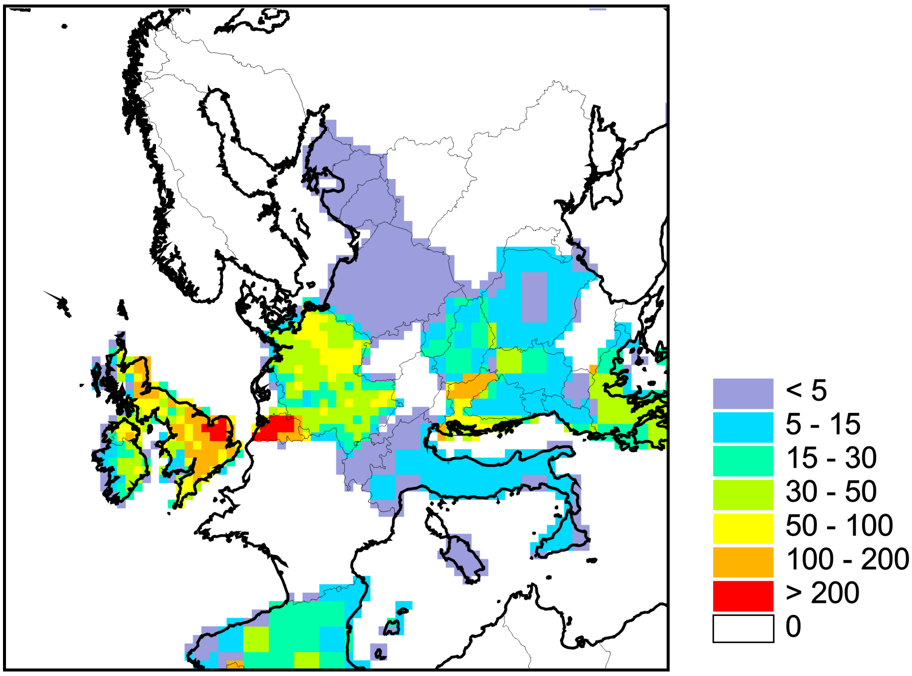


Figure 8.1. Spatial distribution of lindane emission in the EMEP domain in 2001 with resolution 50x50 km^2. Units: g/km^2/year

8.2 Annual deposition of lindane

Annual and monthly depositions and exchange of lindane between the atmosphere and the underlying surface for 2001 were computed for six sub-basins and six catchments of the Baltic Sea. 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.

Spatial distribution of net gaseous flux of lindane over the Baltic Sea region for 2001 is given in Figure 8.2.

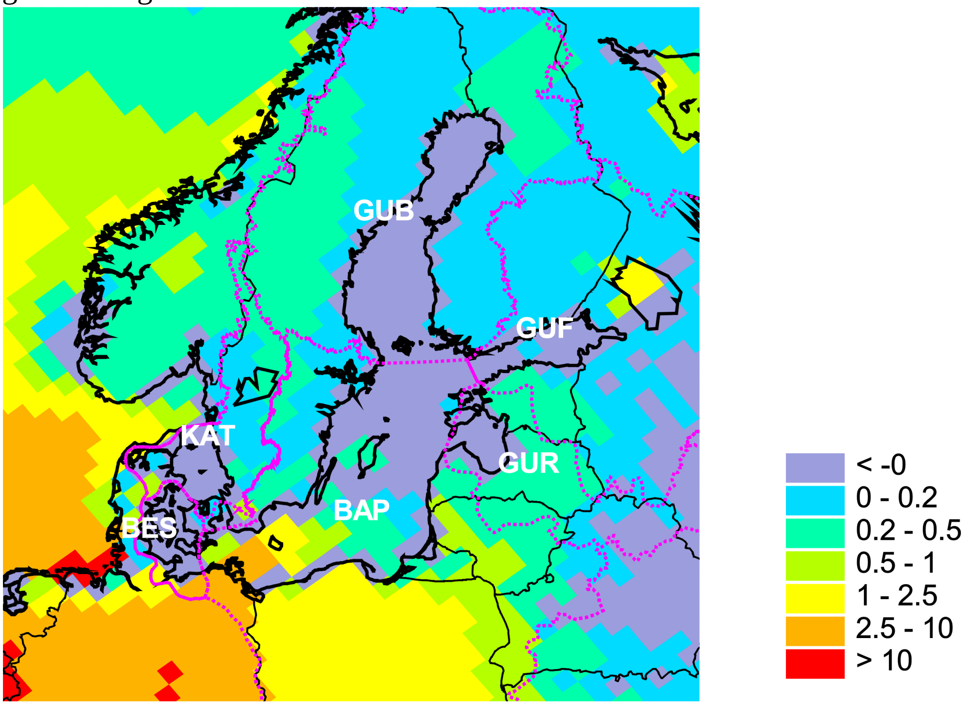


Figure 8.2. Spatial distribution of lindane net gaseous flux in the Baltic Sea region for 2001 with resolution 50x50 km^2. Units: g/km^2/year

Negative values of net gaseous flux can be noted for most of sub-basins of the Baltic Sea. This indicates that re-emission process takes place exceeding the dry and wet deposition fluxes. In the southern part of the Baltic Proper sub-basin (BAP) wet and dry depositions dominate over re-emission of lindane. Tables 8.2 and 8.3 present the distribution of total lindane depositions to the Baltic Sea sub-basins and catchments respectively.

Along with the total depositions, values of deposition fluxes and contributions of wet and net dry depositions are given in the tables. Over the Baltic Sea re-emission fluxes of lindane are dominant for all sub-basins with the maximum in the Belt Sea sub-basin (BES). For the catchment area of the Baltic Sea the distribution of fluxes is different. Contribution of wet deposition fluxes dominates in all catchments. Maximum total deposition flux is estimated for the Baltic Proper catchment (BAP).

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 2001

	GUB	GUF	GUR	BAP	BES	KAT	Baltic Sea
Wet	32	16	15	198	37	17	315
Dry	-242	-101	-49	-285	-140	-72	-889
Total	-210	-85	-34	-87	-103	-55	-575
Flux	-1.8	-2.8	-1.8	-0.4	-5.0	-2.4	-1.4

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 2001

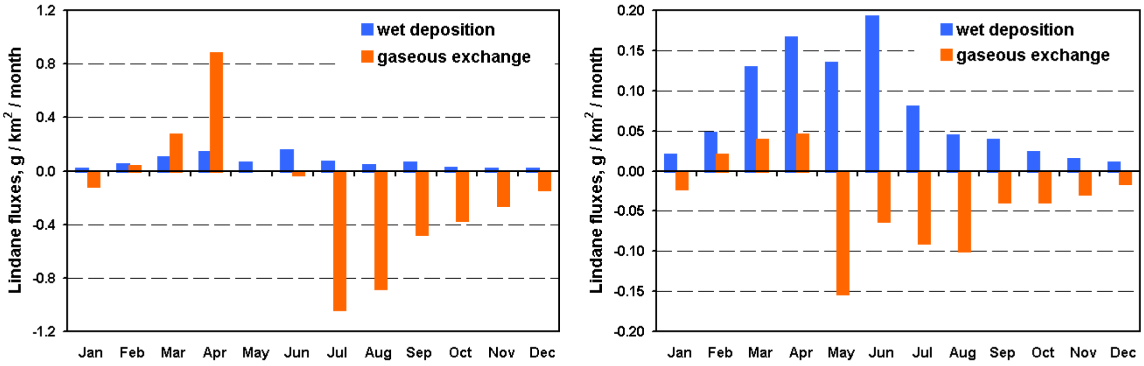
	GUB	GUF	GUR	BAP	BES	KAT	Catchment area
Wet	215	217	116	891	51	60	1550
Dry	-124	-181	-101	-304	-19	-37	-767
Total	91	37	15	587	32	22	784
Flux	0.2	0.1	0.1	1.1	0.3	0.3	0.5

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 2001 are shown in Figures 8.3a and 8.3b respectively. Due to pronounced temporal variation of emissions monthly wet deposition and gaseous exchange fluxes of lindane vary significantly within the year.

For the Baltic Sea gaseous exchange flux of lindane is more intensive comparing to wet deposition flux. During the application period atmospheric input of lindane to the Baltic Sea prevails the re-emission. For the second part of the year, when no application is assumed, lindane previously accumulated in sea water is re-volatilized to the atmosphere. Over the Baltic Sea catchment area wet deposition flux is more significant comparing to gaseous exchange flux. Maximum values of net gaseous fluxes of lindane both for the Baltic Sea

(Figure 8.3a) and its catchment area (Figure 8.3b) are obtained for the spring, in period of its application.



(a) (b)

Figure 8.3. Monthly variations of lindane wet deposition and gaseous exchange fluxes over the Baltic Sea (a) and its catchment area (b) in 2001, tonnes/month.

8.4 Comparison of model results with measurements

For the verification of model results available concentrations of lindane in 2001 measured at the EMEP monitoring stations were considered. Measurements of lindane obtained close to or within the Baltic Sea area were reported by Westerland (DE01), Zingst (DE09), Lista (NO99), De Zilk (NL91), Knokke (BE04). Comparison of measured and computed lindane concentrations and their ratios are presented in Table 8.6.

Table 8.6. Comparison of calculated and measured mean annual lindane concentrations in air and precipitation for 2001

Station code	Station name	Observed	Calculated	Obs / Calc
		Lindane concentrations in air (pg/m^3)		
NO99	Lista	15.7	26.9	0.6
		Lindane concentrations in precipitation (ng/l)		
BE04	Knokke	12.6	3.8	3.3
DE01	Westerland	3.3	2.6	1.2
DE09	Zingst	16.3	4.6	3.6
NL91	De Zilk	6.1	2.9	2.1
NO99	Lista	2.5	0.6	4.3

In comparison with observed mean annual air concentrations of lindane at the station Lista (NO99) computed concentrations are almost twice higher. Concentrations of lindane in precipitation measured at the selected stations are higher than computed ones by a factor of 1-4. Discrepancies between the model results and measurements can be caused by the uncertainties of lindane emission data and model parameterization.