BALTIC SEA ENVIRONMENT PROCEEDINGS

No.68

REPORT ON THE EVALUATION OF THE RELATION OF ATMOSPHERIC DEPOSITION TO RIVERINE INPUT OF NITROGEN TO THE BALTIC SEA

HELSINKI COMMISSION
Baltic Marine Environment Protection Commission

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For bibliographic purposes this document should be cited as:
HELCOM, 1997
Report on the evaluation of the relation of atmospheric deposition to riverine input of nitrogen to the Baltic Sea
Balt. Sea Environ. Proc. No. 68

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ISSN 0357-2994

Publishing Center «Meteorology and Hydrology»
Moscow, Russia

LIST OF CONTENTS

Page

PREFACE 1

PART I. ANTHROPOGENIC NITROGEN DEPOSITION ON THE BALTIC SEA AND ITS WATERSHED DURING THE PERIOD OF 1987–1991 2
1. INTRODUCTION 2
2. GENERAL CHARACTERISTICS OF MSC-E MODELS 4
3. CALCULATION RESULTS 7
4. VALIDATION OF THE RESULTS 9
5. ANALYSIS OF OBSERVED AND CALCULATED DATA FOR GRID CELLS WITH MONITORING SITES 10
6. ESTIMATES OF REGIONAL DEPOSITIONS 15
7. SUMMARY 16
8. CONCLUSIONS 16
REFERENCES 17

PART II. ASSESSMENT OF THE RATIO BETWEEN DEPOSITED NITROGEN RUNOFF AND ITS RIVERINE INPUT TO THE BALTIC SEA . . 18
SUMMARY 18
1. INTRODUCTION 18
2. BRIEF DESCRIPTION OF THE BALTIC SEA DRAINAGE AREA 19
3. GENERAL CONCEPTS OF CALCULATION 20
4. MODEL STRUCTURE 20
5. CALCULATIONS OF NITROGEN DEPOSITION RUNOFF 21
5.1. Initial Information 21
5.2. Direct Calculations of Nitrogen Runoff (Method 1) 22
5.2.1. Nitrogen runoff from terrestrial ecosystems 22
5.2.2. Nitrogen retention in freshwater ecosystems 25
5.3. Indirect Calculation of Nitrogen Runoff (Method 2) 27
5.3.1. Nitrogen retention in terrestrial ecosystems 27
5.4. Fertilizer-Based Calculation of Nitrogen Runoff (Method 3) 29
6. RESULTS AND DISCUSSION 30
6.1. Computed Values of Atmospheric Deposition Nitrogen Runoff 30
6.2. Comparison of Computed and Experimental Results for Forested Ecosystems . . 38
7. UNCERTAINTY ESTIMATES 42
7.1. Nitrogen Runoff from Terrestrial Ecosystems 42
7.2. Nitrogen Retention in Aquatic Ecosystems 43
7.3. Nitrogen Fertilizer Application 43
7.4. Relation of Atmospheric Deposition to Riverine Input of Nitrogen 43
8. THE ASSESSMENT OF THE RELATION OF ATMOSPHERIC DEPOSITION TO RIVERINE INPUT OF NITROGEN TO THE BALTIC SEA 44
9. CONCLUSIONS 46

REFERENCES 46

ANNEX A. DESCRIPTION OF THE MSC-E ACID MODELS APPLIED TO THE BALTIC WATERSHED DEPOSITION CALCULATIONS 52
THE MODEL ADVECTION SCHEME 52
VERTICAL DISTRIBUTION 53
DRY AND WET DEPOSITION 55
THE MODEL CHEMICAL SCHEME 57
REFERENCES 60

ANNEX B. MAPS OF THE EMISSIONS AND CALCULATION RESULTS 62
ANNEX C. RESULTS OF MODEL-MEASUREMENT AND MODEL-MODEL COMPARISON 68

BALTIC SEA ENVIRONMENT PROCEEDINGS 76

PREFACE

This study is dedicated to the Baltic Sea and its watershed pollution with nitrogen compounds of atmospheric origin entering the sea directly and with riverine runoff. This work was initiated by Coalition Clean Baltic (CCB) and supported by Helsinki Commission (HELCOM). In accordance with the decision of the 16th Meeting of Helsinki Commission MSC-E of the Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe (EMEP) was authorized with this work.

This study consists of two parts:

1. Evaluation of anthropogenic nitrogen deposition on the Baltic Sea and its watershed during the period of 1987–1991, prepared by Dr. L. K. Erdman, I. S. Dedkova, S. A. Grigoryan, S. R. Subbotin, T. V. Cheshukina, Dr. M. A. Sofiev (Meteorological Synthesizing Centre-East of EMEP, Moscow).
2. Assessment of the ratio between deposited nitrogen runoff and riverine input of nitrogen to the Baltic Sea, prepared by Prof. V. N. Bashkin, Dr. M. Ya. Kozlov, A. Yu. Abramychev, Dr. I. V. Priputina, A. Tankanag, I. Chekhina (Institute of Soil Science and Photosynthesis, Russian Academy of Sciences) in co-operation with MSC-E.

The draft versions of this report was considered and amended by relevant experts in the framework of TC INPUT prior to release. The editorial work was prepared by the Meteorological Synthesizing Centre — East of EMEP.

* The report presents results of a scientific study therefore they can be disputed.

PART I

ANTHROPOGENIC NITROGEN DEPOSITION ON THE BALTIC SEA AND ITS WATERSHED DURING THE PERIOD OF 1987–1991

1. INTRODUCTION

An evaluation of nitrogen deposition on the Baltic Sea and its catchment area for 1987–1991 is considered. Two runs of MSC-E model, RUN92 and RUN96, were carried out using different MSC-E model versions and different emission data sets for the EMEP grid (Fig. 1).

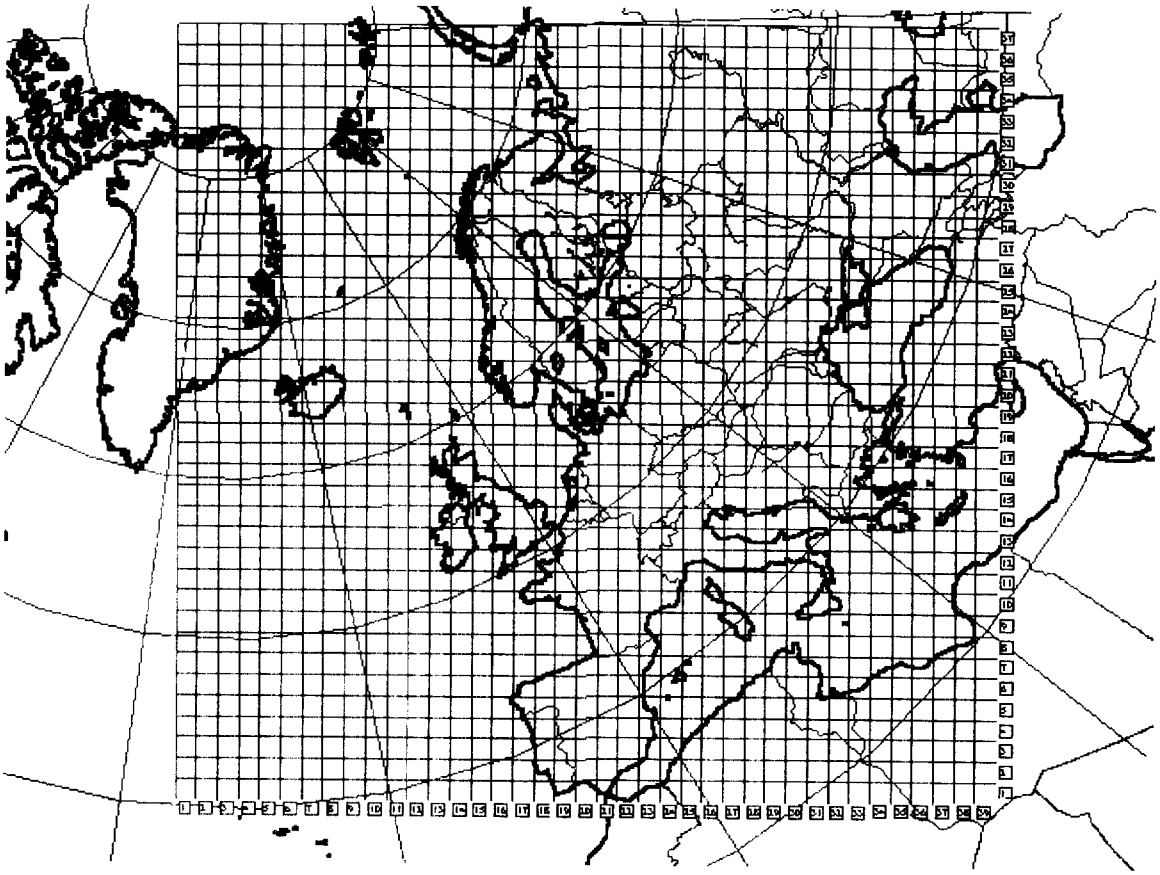


Fig. 1. The EMEP grid

The region covering the Baltic Sea with its division to subregions and the catchment area is presented in [*Laane and Wilhelms*, 1993] (Fig. 2).

RUN92 was carried out in 1992 and its results were used for a long time as a basis for the development of run-off model in order to keep self-consistency of the input data for this run-off model. RUN96 was made in spring 1996 on the basis of new emission data kindly submitted by MSC-W.

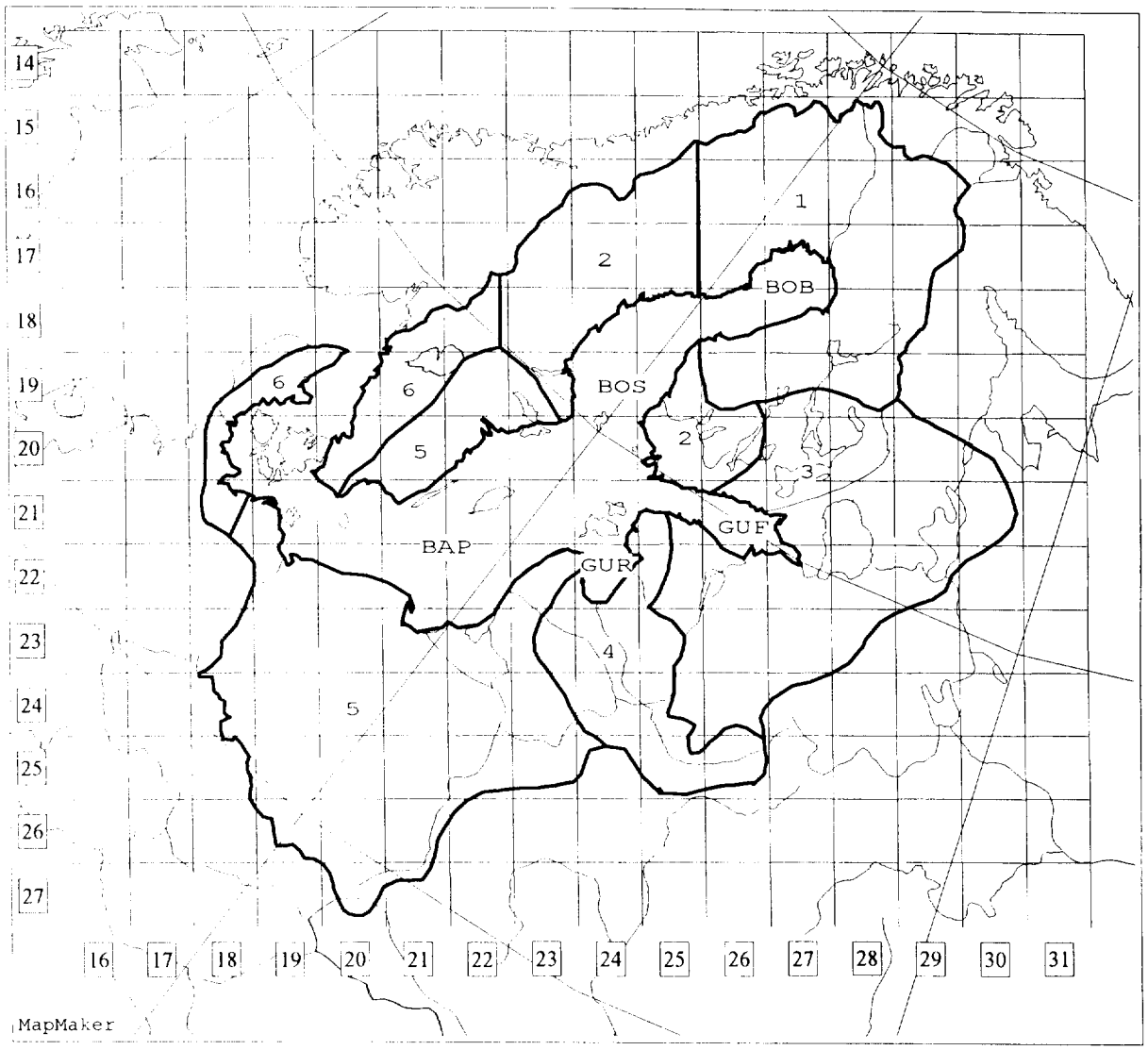


Fig. 2. The Baltic Sea watershed and its subregions.
Thick solid lines limit the Baltic Sea watershed and its subregion:

1. BOB — Bothnian Bay
2. BOS — Bothnian Sea
3. GUF — Gulf of Finland
4. GUR — Gulf of Riga
5. BAP — Baltic Proper
6. SOU — South

KAT — Kattegat
WEB — Western Bays

2. GENERAL CHARACTERISTICS OF MSC-E MODELS

Operational models developed by MSC-E in 1991–1996 under the EMEP programme were used for the evaluation of depositions on the Baltic Seaand its catchment area.The description of the models can be found in Annex A.

The model input consists of geographical, meteorological and emission data. Measurement data (if available) are used only for the evaluation of the quality of modelling results.

Geographical information includes geographical co-ordinates, surface type (sea, land, large fresh water basins, coastal line) and the height above the sea level. Together with emission the allocation of certain receivers (or their parts) to cells is also introduced to the model. Each country, territory or water basin can be represented as an area source, receiver or both. In particular a source can have no area (point sources).

Meteorological information used (1987–1991) includes:

— orthogonal wind components at 850 and 1000 mbar levels;
— precipitation amount;
— temperature (at 2 m above surface).

Averaging interval is 6 hours, spatial resolution — 150 × 150 km^2. The data are prepared by Russian Hydrometeorological Centre using real meteorological data processed with a special objective analysis [*Shapiro*, 1981]. In areas where the observational information is not available (ocean, desert, etc.) a combination of extrapolation/interpolation algorithms with climatological data is used.

Using above mentioned meteorological data and methodology elaborated in co-operation with Russian Hydrometeorological Centre the models make estimates of the state of the underlying surface and other physical parameters important for the calculations.

Emission data

For RUN92 gridded official emission data available in 1992 with 150 × 150 km^2 resolution and indication of the country or region-source are used. Emission from marine areas is taken from international shipping trade data. In cases when official data are not available expert estimates are used. Table 1 and Table 2 contain total values of emission from countries considered in the calculations. Countries made a small contribution to the Baltic Sea pollution (less than 2.5%) are combined in the line "Others". Maps of mean NO_x and NH_x emissions for 1987–1991 are presented in Annex B.

For RUN96 the emission data were provided by MSC-W at the beginning of 1996. Table 3 and Table 4 present total emission values for countries influencing BSCA [*Barret et al.*, 1995]. Emission maps are also included in Annex B.

By the moment of publication of this report Germany has submitted new estimates of NO_x emission for the whole period (namely they are 966, 927, 879, 802 and 763 kt N/yr for 1987–1991 correspondingly). These figures are about 10% lower than those used in the presented calculations.

Table 1

Emission of nitrogen oxides (1000 t N/yr)

Emitters	1987	1988	1989	1990	1991*	Mean
Czech and Slovak	307	321	341	341	341	330
Denmark	80	76	76	76	76	77
Finland	82	84	84	84	84	84
France	496	492	539	539	539	521
Germany	1104	1086	1038	1038	1038	1060
The Netherlands	170	168	168	168	168	168
Poland	466	472	450	390	390	433
Russian Federation**	699	706	777	813	813	762
Sweden	119	119	117	117	117	118
Great Britain	739	755	765	765	765	757
Others	2637	2655	2527	2623	2623	2634
Total**	6899	6934	6982	6954	6954	6946

* For 1991 the last available emission data (1990) were used.
** Emission within the EMEP grid.

Table 2

Emission of ammonia (1000 t N/yr)

Emitters	1987	1988	1989	1990	1991*	Mean
Denmark	110	106	103	103	103	105
Finland	35	35	35	35	35	35
Germany	512	512	512	512	512	512
The Netherlands	209	205	201	201	201	204
Poland	394	394	394	394	394	394
Sweden	51	51	51	51	51	51
Great Britain	394	394	394	394	394	394
Russian Federation**	1367	1367	1367	1367	1367	1367
Estonia	25	25	25	25	25	25
Latvia	51	51	51	51	51	51
Others	4472	4469	4473	4473	4473	4472
Total**	7620	7609	7606	7606	7606	7609

* For 1991 the last available emission data (1990) were used.
** Emission within the EMEP grid.

Table 3

Emission of nitrogen oxides (1000 t N/yr)

Emitters	1987	1988	1989	1990	1991	Mean
Czech and Slovak	308	326	349	356	385	325
Denmark	92	89	83	82	97	89
Finland	82	84	86	86	87	85
France	496	492	539	482	493	500
Germany	1050	1027	980	923	893	975
The Netherlands	182	182	177	173	171	177
Poland	466	472	450	390	367	429
Russian Federation*	807	718	777	814	782	780
Sweden	132	125	123	121	121	124
Great Britain	800	823	838	831	801	819
Others	2563	2549	3061	2834	2820	2766
Total*	6978	6887	7463	7092	6918	7068

* Emission within the EMEP grid.

Table 4

Emission of ammonia (1000 t N/yr)

Emitters	1987	1988	1989	1990	1991	Mean
Denmark	125	123	120	115	110	119
Finland	37	36	35	34	34	35
Germany	690	684	674	625	548	644
The Netherlands	206	199	193	177	182	191
Poland	453	453	453	453	387	440
Sweden	61	61	61	61	60	61
Great Britain	315	315	315	315	315	315
Russian Federation*	1052	1045	1036	981	956	1014
Estonia	26	26	26	26	26	26
Latvia	50	50	50	50	50	50
Others	3343	3348	3354	3268	3188	3300
Total*	6358	6340	6317	6105	5856	6195

* Emission within the EMEP grid.

3. CALCULATION RESULTS

Calculated deposition maps obtained in RUN92 and RUN96 are presented in Annex B. Some results of RUN92 calculations for the whole EMEP grid have been already published in [*Afinogenova et al.*, 1992; *Dedkova et al.*, 1993; *Erdman et al.*, 1993].

Fig. 3 and Fig. 4 present charts of calculated annual nitrogen deposition onto the Baltic Sea Catchment Area for 1987–1991 for RUN92 and RUN96 correspondingly.

It should be noted that for the whole considered region the discrepancy between total estimates of two model results is rather small. At the same time there are some redistributions between subregions and certain differences for some years (especially for 1991) probably caused by differences in emissions (especially for NH_x) as well as by considerable improvements of the model between two runs.

For 1991 seasonal variations of reduced, oxidised, and total nitrogen deposition on the considered region are presented in charts of Fig. 5. These charts reflect the results of RUN92.

Brief overview of these charts shows that the most significant seasonal variations are observed for NH_x depositions which almost completely determine the seasonal trend lines for the bound nitrogen deposition.

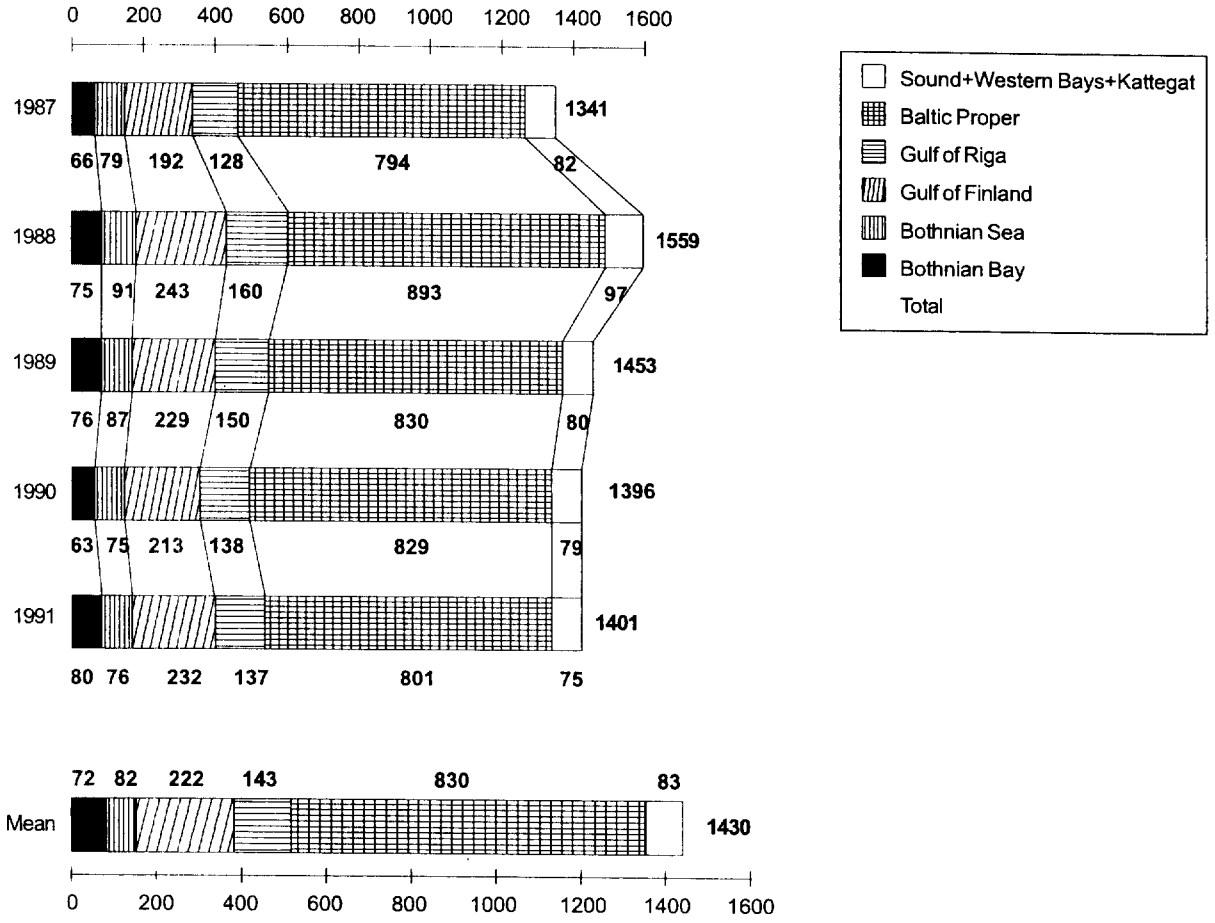


Fig. 3 Annual deposition of nitrogen on the Baltic Sea catchment area and its subregions (in 1000 t N/yr), RUN92 results

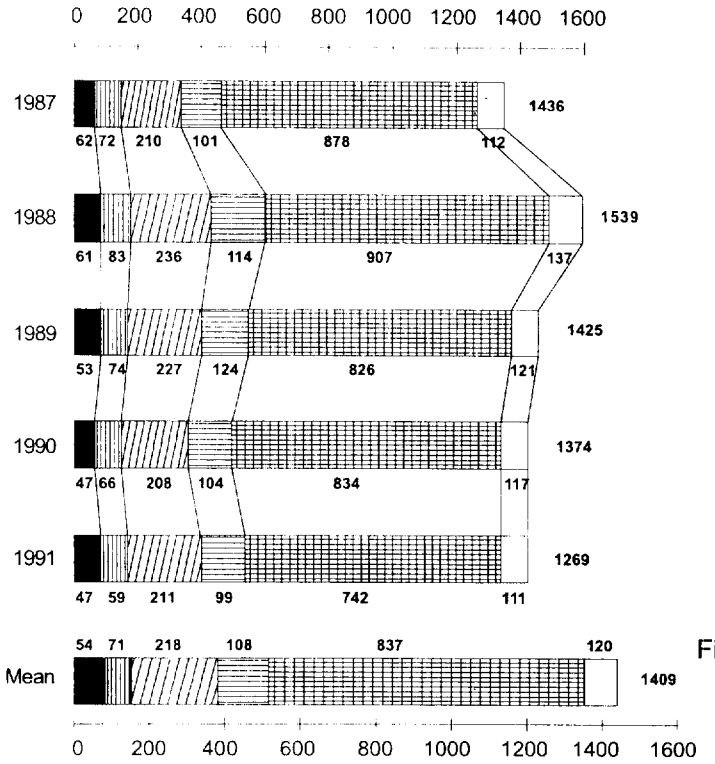
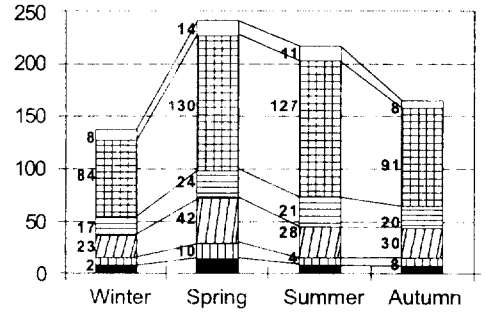
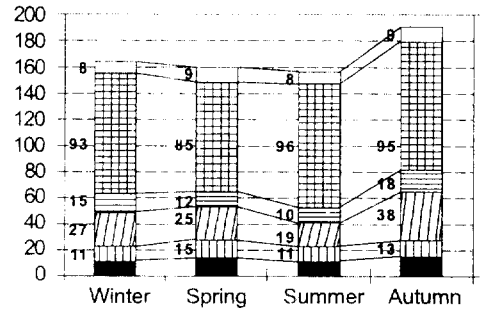


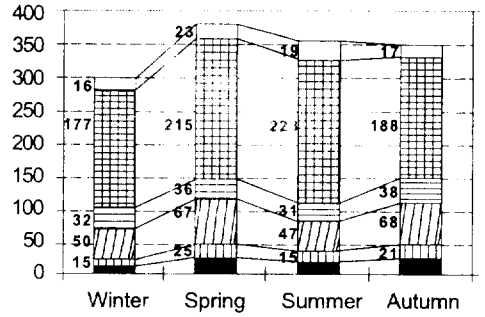
Fig. 4. Annual deposition of nitrogen on the Baltic Sea catchment area and its subregions (in 1000 t N/yr), RUN96 results



a) Reduced nitrogen deposition



b) Oxidised nitrogen deposition



c) Total nitrogen deposition

Fig. 5. Seasonal variations of reduced, oxidised, and total nitrogen deposition onto the Baltic catchment area and its subregions for 1991 (unit = 1000 t N/yr), RUN92 results

4. VALIDATION OF THE RESULTS

Some general information about the agreement of model results with measurements can be found in [*Dedkova et al.*, 1993; *Annual report*, 1993; *Sofiev et al.*, 1994] and in [*Sofiev and Grigoryan*, 1996]. The first, the second and the third reports describe the results of the model version of 1992 and 1993 (which is close to that used for RUN92). The last report contains results of the model applied to RUN96 (hereinafter it is referred as 3DWind-model).

This chapter is dedicated to the comparison of results of RUN92 and RUN96 with measurements, their intercomparison with results of the MSC-West model [*Barret et al.*, 1995] (further referred as W95).

For the multiparametrical quality assurance the scheme of validation presented in Fig. 6 was used. Full procedure consists of two parts.

The analysis is applied to several grid cells where monitoring stations are located. This part of the procedure has been made for wet deposition for two reasons:

— it is a directly measured parameter

— wet deposition on a grid cell is directly used in the run-off model as input data.

The second part of the intercomparison is connected with the analysis of depositions on some regions like Baltic Sea or HELCOM countries. For the regions only total depositions of NO_x and NH_x are available, hence this parameter was used instead of wet depositions.

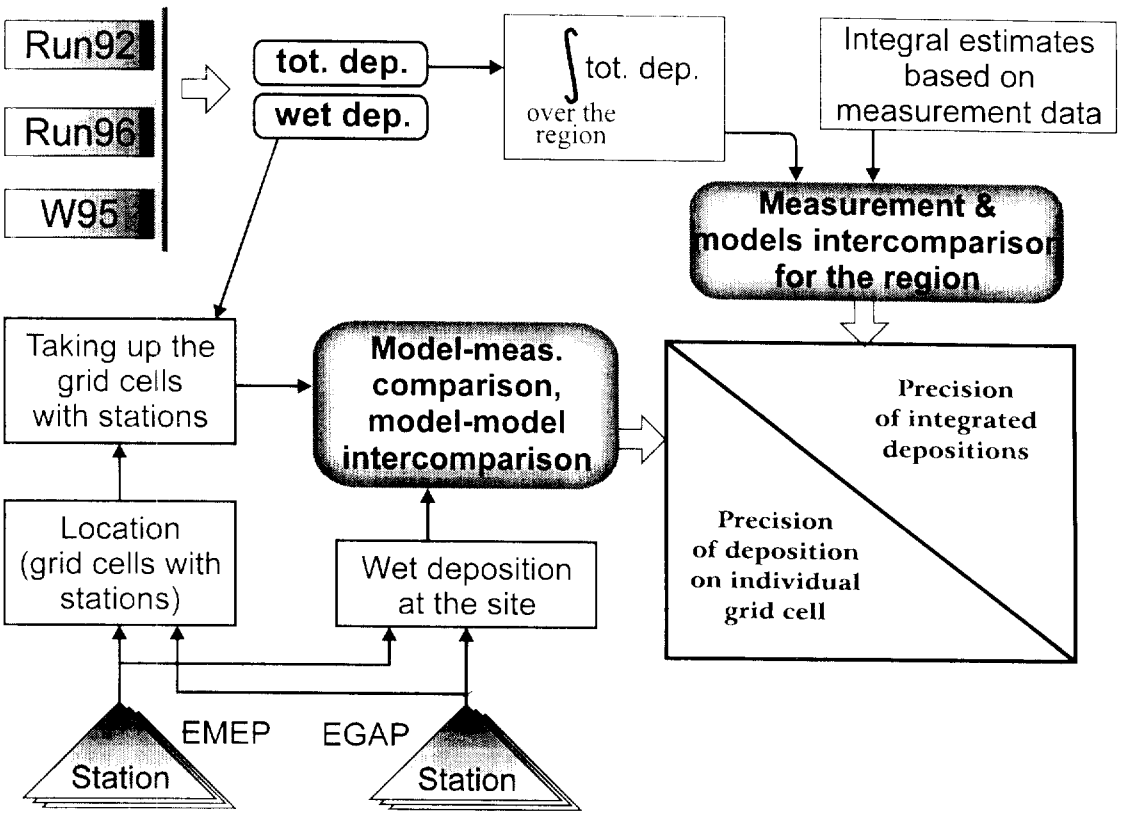


Fig. 6. Scheme of validation of calculation results

5. ANALYSIS OF OBSERVED AND CALCULATED DATA FOR GRID CELLS WITH MONITORING SITES

In this analysis data of EMEP and EGAP monitoring networks were used.

Observations of 30 EMEP stations were taken into account. The site location was the only criterion of the station selection — a station should be within the BSCA or at most 2 grid cells outside this region. Station Nida (SU3) was excluded because of irregularities of the submitted data for the considered period (1987–1991). The map of considered EMEP stations is shown in Fig. 7. All EGAP stations (but Nida SU3) were considered. The map of their locations is presented in Fig. 8.

Data of all stations and the model result sets were averaged over the period (1987–1990). If several stations are located in the same grid cell their data were averaged.

A full set of comparison results is given in Annex C. It includes model-measurement and model-model scatter plots, regression analysis results, etc.

Summary of the comparison is shown in Table 5 (Fig. 9) and Table 6 (Fig. 10) for EMEP and EGAP networks correspondingly. In addition, these tables include some analysis of the data discussed further.

Table 5

Annual wet deposition of N_{tot} at EMEP stations, 1987–1990. Unit = mg N m^{-2} yr^{-1}

Site	Observed	RUN92	RUN96	W95
A2	852	1003 (=)	1061 (=)	1094 (=)
CS1	1117	1646 (+)	1761 (+)	1835 (+)
CS3	914	1031 (=)	992 (=)	1655 (+)
D1	944	1011 (=)	979 (=)	1216 (+)
D2	936	1343 (+)	1298 (+)	1293 (+)
D5	1419	1621 (=)	1970 (+)	1634 (=)
DD1	905	1106 (=)	1306 (+)	948 (=)
DD2	1101	1199 (=)	1339 (=)	1092 (−)
DK3	836	1326 (+)	971 (=)	1388 (+)
DK5	847	1458 (+)	1444 (+)	1693 (+)
EE2	922	726 (=)	728 (=)	501 (−)
EE9	326	540 (+)	450 (+)	439 (+)
FI4	394	258 (−)	214 (−)	295 (−)
FI9	778	345 (−)	532 (−)	245 (−)
FI17	731	540 (−)	450 (−)	439 (−)
LV10	762	1043(+)	1021 (+)	865 (=)
NO15	266	186 (−)	80 (−)	241 (=)
NO30	84	97 (=)	59 (−)	55 (−)
NO39	224	239 (=)	353 (+)	166 (−)
NO41	393	394 (=)	281 (−)	394 (=)
NL2	1953	2263 (=)	2022 (=)	2902 (+)
PL1	836	1406 (+)	1147 (+)	969 (=)
PL2	850	1096 (+)	1440 (+)	1067 (+)
SE2	1023	749 (−)	846 (=)	824 (=)
SE3	673	355 (−)	624 (=)	501 (−)
SE5	257	181 (−)	202 (=)	220 (=)
SE8	979	547 (−)	748 (=)	476 (−)
SE13	1310	775 (−)	1081 (=)	909 (−)
SU3	523	410 (=)	530 (=)	457 (=)
RU1	56	139 (+)	61 (=)	84 (+)
Mean	774	834	866	863
Deviation of calculations from observations is within 25%	=	12	14	11
Overestimation	+	9	10	10
Underestimation	−	9	6	9

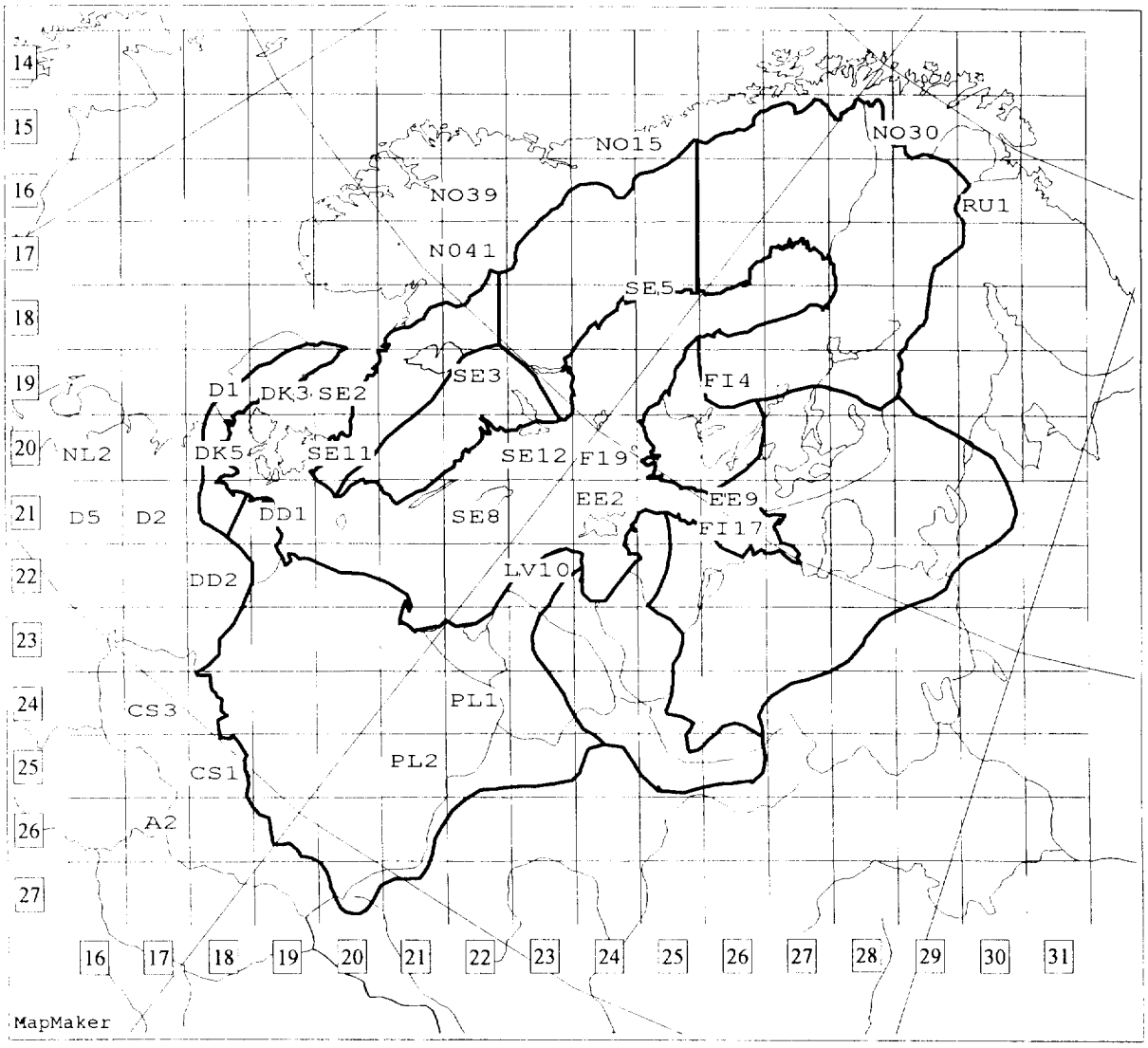


Fig. 7. Location of EMEP stations used in comparisons: measurements versus calculations and calculations versus calculations

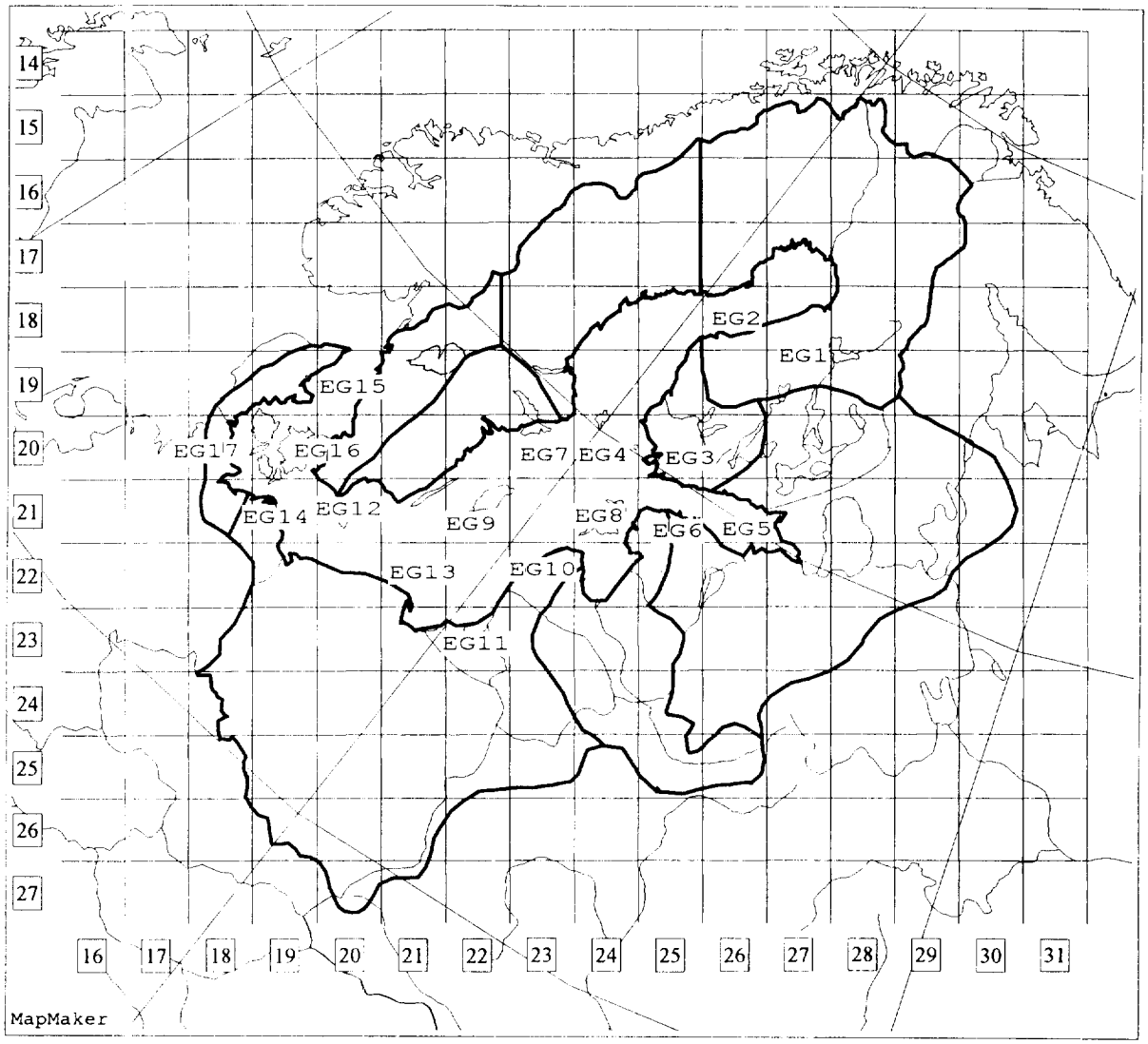


Fig. 8. Location of EGAP stations used in comparisons: measurements versus calculations and calculations versus calculations. Notations are included in Table 6

Summarising these data it can be stated that calculated results are in an acceptable agreement with measurements. For the EMEP network mean values calculated by three models are about 10% higher than measured ones, for the EGAP network the calculated results are 5–20% lower (about 5% for RUN96 and about 20% for RUN92 and W95).

A similar pattern is manifested in correlation coefficients. For EGAP data set they are varied from 0.57 up to 0.67. The agreement of all models with EMEP data is better and more uniform — the correlation coefficients are 0.83–0.88.

Table 6

Annual wet depositions of N_{tot} at EGAP stations, 1987–1990. Unit = mg N m^{-2} yr^{-1}

Stations		Observed	RUN92	RUN96	W95
EG1	Hailuoto, Rahja	342	166 (–)	351 (=)	190 (–)
EG2	Ricklea, Sulva, Ylimarkku	560	280 (–)	403 (–)	275 (–)
EG3	Korppoo	788	420 (–)	394 (–)	494 (–)
EG4	Jomala, Utö	876	345 (–)	532 (–)	245 (–)
EG5	Virolahti, Haapasaari, Sipoo, Lahemaa, Vanhankylänmaa	623	540 (=)	450 (–)	439 (–)
EG6	Tväminne	858	658 (=)	569 (–)	671 (=)
EG7	Aspvreten	468	410 (=)	530 (=)	457 (=)
EG8	Syrve	875	726 (=)	728 (=)	501 (–)
EG9	Hoburg	930	547 (–)	748 (=)	476 (–)
EG10	Rucava	705	1043 (+)	1021 (+)	865 (=)
EG12	Arup	1029	642 (–)	975 (=)	744 (–)
EG13	Leba	802	681 (=)	1008 (+)	682 (=)
EG14	Kap Arcona	1019	1106 (=)	1306 (+)	948 (=)
EG15	Rörvik	1025	749 (–)	846 (=)	824 (=)
EG16	Vavihill	1154	775 (–)	1081 (=)	909 (=)
EG17	Keldsnor, Dänisch-Nienhof	955	1458 (+)	1444 (+)	1693 (+)
	Mean	813	659	774	651
	Calculations deviate from measurements within 25%	=	6	7	7
	Overestimation	+	2	4	1
	Underestimation	–	8	5	8

The model-to-model intercomparison manifests a very good agreement for all three models. (as it was mentioned above, the regression scatter plots are presented in Annex C). Aggregated numbers are shown in Table 5 and Table 6 and Fig. 9 and Fig. 10. Like in model-measurement comparison only grid cells with monitoring sites (separately EGAP and EMEP ones) are considered.

It is well seen that mean values of different models deviate from each other less than by 4% for the EMEP data set and less than by 12% for the EGAP one. The correlation coefficients exceed 0.9.

One more qualitative illustration of the similarity between the model and measurements data can be made by the consideration of the following classifications (see bottom parts of Tables 5 and 6). Let mark the stations with less than 25% model-measurement deviation by the equal sign (=). If model underestimates the value by more than 25% than use sign "–", in case of overestimation more than 25% — sign "+". This classification diminishes small chaotic fluctuations of values and highlights the main pattern of differences.

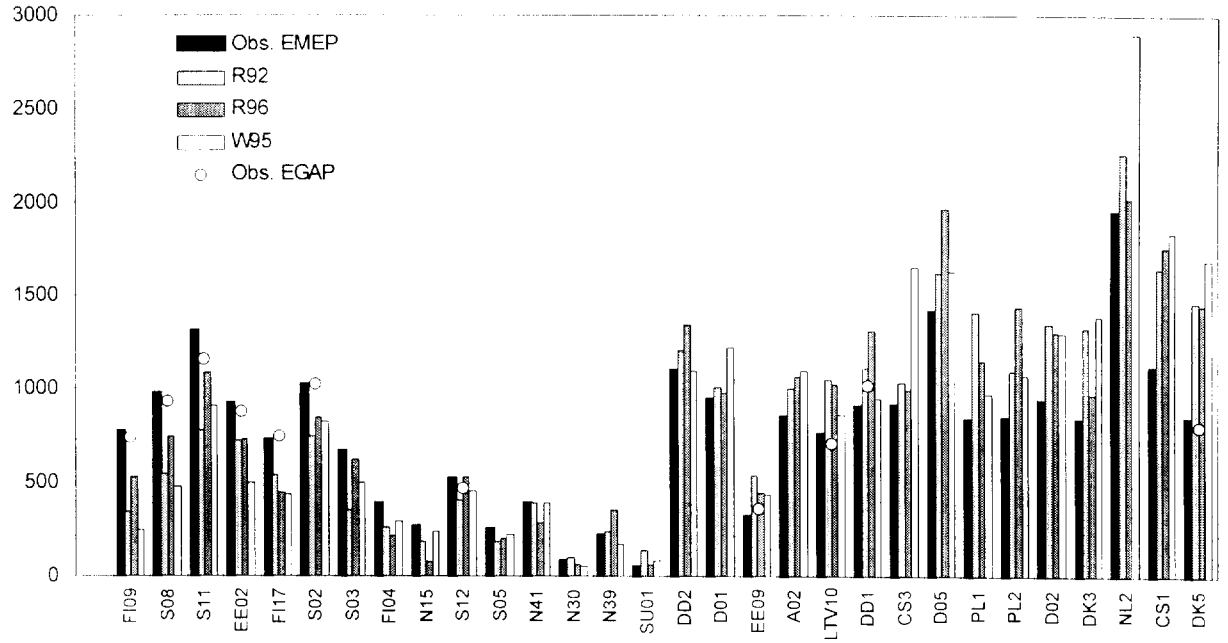


Fig. 9. Combined comparison of measured and calculated results [$N_{tot} = (N\hat{I}_x + NH_x)$] mean annual wet deposition in 1987–1990 at EMEP stations. Unit = mg N m^{-2} yr^{-1}

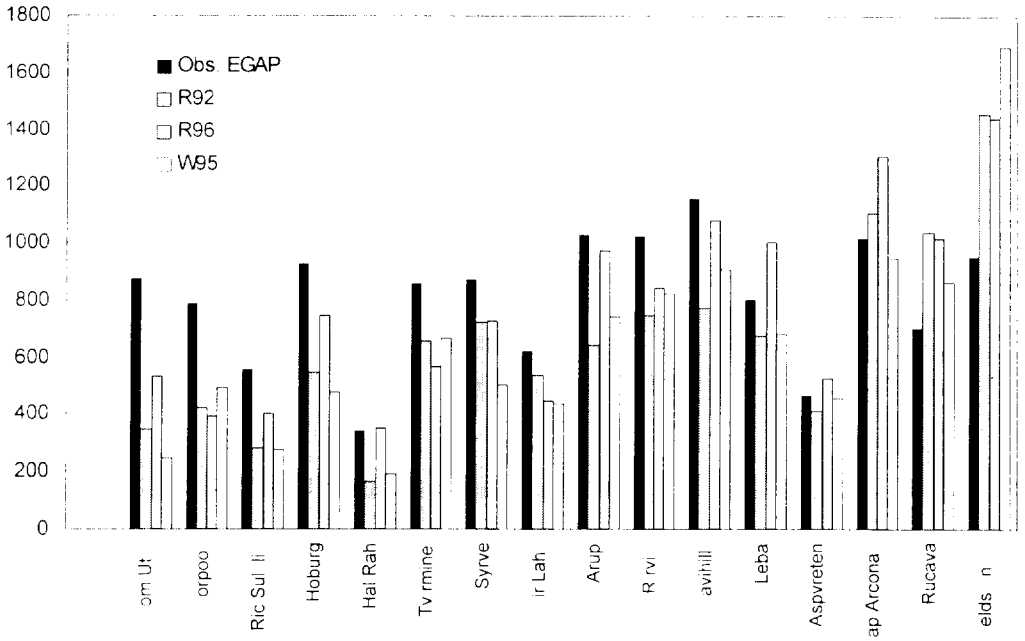


Fig. 10. Combined comparison of measured and calculated results [$N_{tot} = (N\hat{I}_x + NH_x)$] mean annual wet deposition in 1987–1990 at the EGAP stations. Unit = mg N m^{-2} yr^{-1}

The most important conclusion which can be drawn from this analysis is: the differences from particular station are of the same sign for all models. There is the only station (EMEP NO39) for which one model shows underestimation but another — overestimation. This consideration confirms a general conclusion that model results are much better agreed with each other than with measurements although measurement/calculation agreement is also acceptable.

The spatial analysis of deviations shows that all models tend to underestimate the wet depositions in Scandinavia and to overestimate in regions close to central Europe (Germany, Austria, Czech Republic, etc.).

Some reasons for the mentioned discrepancies are analysed in [*HELCOM,* 1991]. All of them (namely, uncertainties of emission and meteorology, difficulties with separation and consideration of point sources, re-emission of ammonia, etc.) are remained at this stage of investigations as well as inaccuracies of model parametrization.

Some rough estimates of the representativeness of the stations in the grid cells can be taken from [*Galperin & Sofiev,* 1993]. Corresponding standard deviation (about 25%) should be taken into account as an independent summand of the total distortions. The reduction of calculation grid cell size down to 50 km will probably lead to diminishing of this uncertainty.

In some cases the data reported to EMEP and EGAP data bases are inconsistent. For example, for Keldsnor (DK 5) wet deposition of NH_x and NO_x amounts to 729 and 468 mg N m^{-2} yr^{-1} in EMEP data base [*Pedersen et al.,* 1990, p. 52] and — 481 and 441 mg N m^{-2} yr^{-1} according to HELCOM data base [*HELCOM,* 1991, pp. 82, 83]. These discrepancies are also shown in Fig. 9 where circles indicate data submitted to EGAP by EMEP/EGAP stations.

6. ESTIMATES OF REGIONAL DEPOSITIONS

The W95 estimate of the deposition onto the Baltic Sea averaged over 1987–1991 amounts to about 282 kt N yr^{-1}. The RUN92 result is close to 315 kt N yr^{-1}. Both figures are completely within the range given in [*HELCOM,* 1991] which is 300 30 kt N yr^{-1}. The last estimate is based on the combination of measurement and modelling results.

There are no published data on the deposition onto the Baltic Sea Catchment Area (BSCA). Instead of this value the deposition on the HELCOM countries was used. The area of these countries covers about 95% of BSCA, thus this estimate is quite representative. The analysis period was limited by one year — 1991 since it is the only year in the five-year period (1987–1991) for which separated estimates for Baltic countries (Estonia, Latvia and Lithuania) and Russia are provided by W95. For earlier years only total figures for the Soviet Union are available.

Table 7 presents the summary of the comparison of model deposition values.

Discrepancies for individual countries are mostly within the range of 20–30% and discrepancies for countries of a small area are greater than those of a large area.

It should be noted that depositions onto the territory of Russia and Germany are not representative for the Baltic watershed since the Baltic Catchment area is a small part of their territories. As an illustration of this statement it can be mentioned that a considerable difference in depositions on Russia are mostly determined by drastic changes in Ukrainian and Belarussian emission values submitted and used in different model runs (for details see [*Afinogenova et al.,* 1992; *Barret et al.,* 1995]).

Table 7

Deposition of nitrogen compounds on HELCOM countries in 1991 (in 1000 t N)

Countries	Compounds								
	NO_x			NH_x			N_{tot}		
	RUN92	RUN96	W95	RUN92	RUN96	W95	RUN92	RUN96	W95
Denmark	30	21	25	41	27	58	71	48	83
Estonia	18	15	18	19	15	21	37	30	39
Finland	77	53	72	45	33	47	122	86	119
Germany	400	420	317	403	395	430	803	815	747
Latvia	32	20	26	38	28	39	70	48	65
Lithuania	38	27	30	53	39	52	91	66	82
Poland	278	261	224	288	239	287	566	500	511
Russia	943	826	722	1383	825	838	2326	1651	1560
Sweden	106	101	115	60	67	78	166	168	193

Note: While the analysis of the table it should be taken into account that for RUN96 values are calculated by a new version of computer GIS software with improved geography data base.

7. SUMMARY

Summarizing results of the comparison of MSC-E two model runs with each other and with other data:

1. Estimates of depositions over large areas (the Baltic Sea or HELCOM countries within BSCA) made by different models are rather close, the discrepancy is mainly within 20–30%. Estimates of depositions on the Baltic Sea made on the basis of EGAP measurement data are also in a good agreement with model results.
2. The discrepancy between mean calculated values of all three models and mean values of measurements averaged over all sites of the EMEP and EGAP monitoring networks (over 40 points in total) does not exceed 20%. Correlation coefficients are within the limits of 0.57–0.88. Standard deviations of measurement values are about 25%. The sum of these figures gives to 30% of standard deviation in grid cells.
3. The intercomparison of deposition values obtained by various models for monitoring sites of the EGAP/EMEP networks points to very high similarity between model results.
4. In general it can be concluded that reliable estimates of depositions on the BSCA area are in the range of 20% deviation from the presented results. At the same time deposition data on the northern subregions can be somewhat underestimated and in southern ones — somewhat overestimated.

8. CONCLUSIONS

1. In accordance with the presented results and [*HELCOM*, 1991] data mean nitrogen deposition on the Baltic Sea can be estimated as 300 ± 30 kt N per year for the period of 1987–1991.
2. For the same period mean annual nitrogen deposition on the Baltic Sea Catchment Area can be evaluated as 1400 ± 250 kt N per year.

3. Evaluations of the reliability of the above values indicate that different models show very similar results. Almost without exception the deviation of calculated results from available measurements is of the same sign and order of magnitude.
4. In order to obtain more reliable estimates of atmospheric nitrogen runoff from BSCA it is desirable to operate with longer calculation period and with higher spatial resolution (e.g., 50 × 50 km^2).

Acknowledgements

We are grateful to Dr. Till Spranger and Dr. Carl-Ellis Bostrm for a fruitful discussion of initial results and provision of necessary information, to Dr. Sindre Langaas — for operational transmission of required BGIS data, to Dr. Herata Heike who helped with data questionnaires.

We are also grateful to Dr. Sofia Milona (EMEP/MSC-W) for providing emission data on sulphur and nitrogen and consultations as well as to Dr. Erik Berge and Dr. Kevin Barrett (EMEP/MSC-W) for providing calculated data on nitrogen deposition in 1987–1990.

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PART II

ASSESSMENT OF THE RATIO BETWEEN DEPOSITED NITROGEN RUNOFF AND ITS RIVERINE INPUT TO THE BALTIC SEA

SUMMARY

Evaluation of the relation of atmospheric deposition to riverine input of nitrogen to the Baltic Sea from its watershed has been carried out for the period of 1987–1991. It is shown that the deposition of nitrogen compounds on the watershed is c. 5 times higher than their direct deposition on the sea surface. An assessment of runoff of nitrogen to the Baltic Sea from its watershed has been carried out by three methods and the results of calculations were very similar. It was demonstrated that the main sources of uncertainties are connected with values of nitrogen runoff and retention versus different types of land use types. Depending on subregions the relation between airborne nitrogen input to the Baltic Sea and nitrogen deposition on the catchment area changed from 5 up to 10%. Minimum values were obtained for northern subregions and maximum — for southern and western ones. For the whole basin of the Baltic Sea during 1987–1991 period runoff values were within the limits of 8.4–8.6% of the total amount (1268–1539 kt) of atmospheric nitrogen deposition on the watershed. The input of atmospheric deposition to the riverine input to the Baltic Sea was 21% (118.6 kt) in 1990.

1. INTRODUCTION

At present nitrogen input into the Baltic Sea basin occurs due to river runoff, direct urban and industrial waste discharge as well as by nitrogen deposition directly into sea waters. It is very difficult to distinguish the role of atmospheric nitrogen in its biogeochemical balance, especially that part of nitrogen which reaches the Baltic Sea basin being deposited on its drainage area. These difficulties are related, firstly, to many uncertainties of parameters characterizing nitrogen input, transformation in terrestrial and freshwater ecosystems, corresponding retention, volatilization, denitrification, runoff etc. [*The role...*,1992].

The Baltic Sea was found to be rather sensitive to the anthropogenic pollution due to its poor water mixing with the ocean, low depth and low salinity. According to existing data [*Airborne Pollution...*, 1991; *Dedkova et al.*, 1993; *Erdman et al.*, 1993; *Enell and Fejes*, 1995, see also part I] nitrogen deposition on the sea basin is about 300 30 kt N/yr. However, the lowest estimates are of 190 kt N/yr [*Agren*, 1996]. The nitrogen loading with river runoff, direct urban and industrial discharge to the sea amounts to 650–980 kt/yr depending on the calculation method and reference year [*Laane and Wilhelms*, 1993; *Nutrient Transport...*, 1991; *Enell and Fejes*, 1995]. Up till now there is no quantitative assessment of nitrogen deposition on the Baltic Sea watershed and an inflow to the Baltic Sea with riverine discharge and direct coastal surface runoff.

For example, the evaluation of natural and anthropogenic contributions to riverine fluxes, *L.M.Svendsen et al.* [1995] does not provide the quantitative assessment of N atmospheric deposition on its riverine fluxes.

Furthermore, assuming nitrogen deposition to be the only source of this element in the natural landscape, the background loads of nitrogen to riverine fluxes can represent the conditions in the watershed not effected by human activity (atmospheric deposition input exepted). At present no watersheds meet these conditions since the amount of atmospheric deposition is increasing due to human activity. Thus, the best estimate of background loads is found in small, scarcely populated catchments with low human activity giving an opportunity to compare computed values with experimental monitoring of N runoff.

2. BRIEF DESCRIPTION OF THE BALTIC SEA DRAINAGE AREA

The Baltic Sea drainage area is about 1 733 850 km^2 (Table 1). Table 1 and Fig. 2, Part 1 show also the division of drainage area between the countries and sub-regions. The main parts of this area belong to Finland, Russia, Poland and Sweden, being in sum 1 398 200 km^2 or 80% of the whole watershed as well as the area of Bothnian Bay (BOB), Bothnian Sea (BOS), Gulf of Finland (GUF) and Baltic Proper (BAP) is totally 1 463 800 km^2 or 84%. Gotland Island is not included in BAP drainage area.

Table 1

Division of the Baltic Sea Drainage Area between the Baltic States and Baltic Sub-regions, km^2*

Country	Subregion						
	Bothnian Bay (BOB)	Bothnian Sea (BOS)	Gulf of Finland (GUF)	Gulf of Riga (GUR)	Baltic Proper (BAP)	Western** Bays (SOU)	Total
Baltic Sea contacting countries							
Finland	146000	48000	107300	–	–	–	301300
Russia	–	–	276100	23700	15000	–	314800
Estonia	–	–	26400	17600	1100	–	45100
Latvia	–	–	3500	48500	12600	–	64600
Lithuania	–	–	–	16500	48800	–	65300
Poland	–	–	–	–	311900	–	311900
Germany	–	–	–	–	12600	10950	23550
Denmark	–	–	–	–	1200	29900	31100
Sweden	131000	180100	–	–	84900	74200***	470200****
Baltic Sea non-contacting countries							
Belarus	–	–	–	25800	46900	–	72700
Ukraine	–	–	–	–	11000	–	11000
Slovak	–	–	–	–	8500	–	8500
Total	277000***	219100	413300	132100	554500	115050***	1733850***

* On a basis of PLC-2, 1993 [*Laane and Wilhelms,* 1993].
** Western Bays includes also the Sound and Kattegat.
*** Including Norwegian drainage area.
**** Gotland Island is not included.

For better understanding of the nitrogen transformation in terrestrial and freshwater ecosystems of various subregions, general information for the whole Baltic Sea basin was picking up. In accordance with PLC-2 [*Laane and Wilhelms*, 1993], a remarkably big part (60–70%) of the

area is under agricultural activity in Germany, Denmark and Poland, whereas this area in Estonia, Latvia and Lithuania is 30–50% and in Russia, Finland and Sweden less than 7-10% of drainage area. Forest, swamps and waterbodies, first of all lakes, of various sizes cover more than 65–90% in Finland, Russia, Sweden and Estonia. The urban area covers from 1.1% (Sweden) up to 15.0% (Denmark) of subregions.

3. GENERAL CONCEPTS OF CALCULATION

Taking into account the modern state-of-the-art of such investigations, two approaches can be used:

a) quantitative assessment of a fraction of atmospheric nitrogen deposition removed from terrestrial and freshwater ecosystems by surface and underground runoff and entered the Baltic Sea by rivers (direct);

b) quantitative assessment of a part of atmospheric nitrogen deposition entered the Baltic Sea by rivers as a difference between deposited nitrogen and its part retented in terrestrial and freshwater ecosystems (indirect).

Both direct and indirect approaches represent quantitative estimates of various links of the nitrogen biogeochemical cycle and its different trophical chains. These approaches are dealing with the conjugated links (retention v/v runoff), however, in reality the experimental data required for parametrization of models developed on a basis of the given approaches were picked up from fully different trials that allows us to consider these estimates to be rather independent. Furthermore, in order to harmonize the obtained results, their validation has been done using the third set of data related to a quantitative assessment of nitrogen leached from agrolandscapes. Each method has its own merits and drawbacks related firstly to the availability of experimental data and their uncertainty. Furthermore, these approaches seem to be interadditive. Consequently, it allows us to use all the suggested approaches for a comparative calculation of the relation of atmospheric deposition to riverine input of nitrogen to the Baltic Sea.

All 3 calculation methods used are SSMB (steady state mass balance) models.

4. MODEL STRUCTURE

The majority of existing experimental data on nitrogen transformation refer to soils, their main features, such as physical-chemical parameters, mechanical composition, water regime etc. as well as to land use types and nitrogen external input. Of course, these data should be used in the structure of models applied to calculations. It seems very important to use also experimental data on nitrogen retention (uptake, immobilization, denitrification etc.) during flowing in freshwater ecosystems taking into account their trophical stage and residence time. It is necessary to include in the model structure results on dynamic seasonal distribution of runoff values, straightly connected with wet and dry deposition of nitrogen, hydrothermic coefficients and temporal trends. For reducing the uncertainty of experimental results and expert estimations, the spatial and temporal resolution of model has to be determined very correctly, i.e. minimum taxon level must be chosen in order to provide the information for all model calculations. Accounting available information on the Baltic Sea basin, the resolution level was connected with LoLa-grid cells (2' longitude × 2' latitude), which were subdivided on elemental taxones in accordance with soils and land use types in accordance with GIS-data sets of the Baltic Sea drainage basin [*Langaas*, 1992].

Accordingly, the model structure consists of a nucleus which depends on methods of calculation (direct or indirect) and various DBs (atmospheric precipitation input of nitrogen compounds; soil and land use types; values of runoff coefficients; coefficients of nitrogen uptake, immobilization and denitrification; retention in terrestrial and freshwater ecosystems, hydrothermic coefficients, fertilizer application etc.).

The calculation algorithm of the model is shown in Fig. 1.

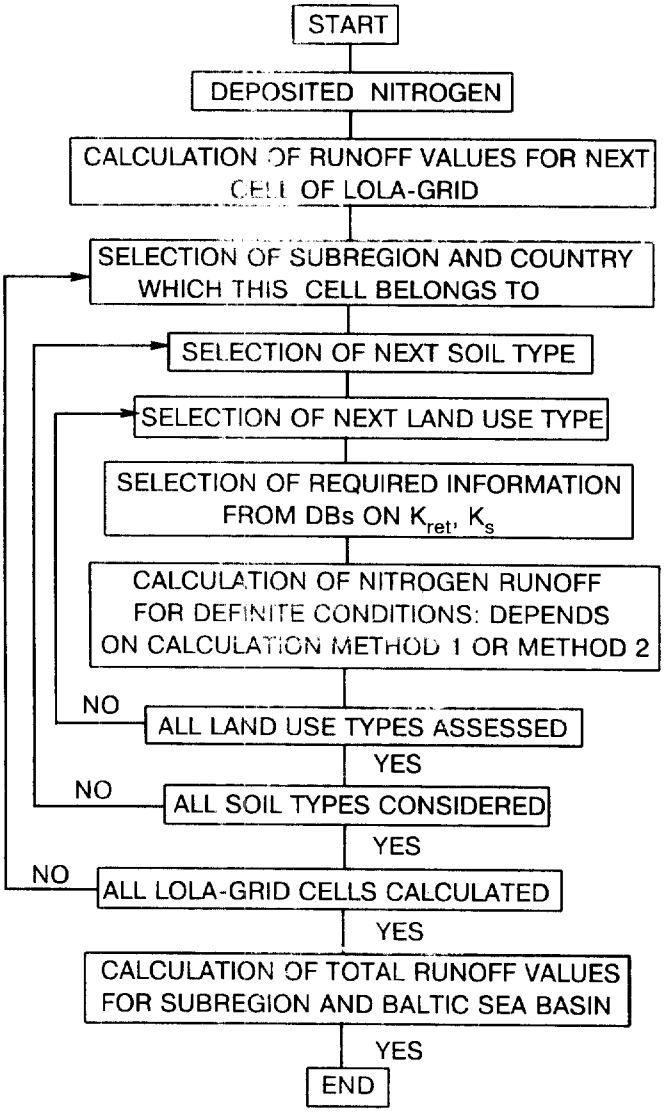

Fig. 1. Algorithm for calculation of deposition nitrogen runoff

A significant difference in model structure and algorithm of calculation is related to an assessment of values of nitrogen runoff: direct estimations in method 1 and indirect ones (as a difference between depositions and retentions) in method 2. Our previous calculations have been carried out on the basis of simplified method 2 [*Erdman et al.*, 1993].

All parameters of the biogeochemical nitrogen cycle included in models used in method 1 and method 2 were estimated and calculated independently on the basis of different data bases (for example, nitrogen water runoff v/v nitrogen plant uptake, both measured experimentally, etc.) giving an opportunity to consider both methods and obtained results as independent values.

Considering the structure of data bases related to nitrogen fertilizer application in HELCOM Contracting countries and obtained from national representatives on HELCOM request, a simplified algorithm has been used for running method 3.

5. CALCULATIONS OF NITROGEN DEPOSITION RUNOFF

5.1. Initial Information

The land cover types, types of soils and their properties are taken from GRID-Arendal DB [*Langaas,* 1992] as well as from soil maps [*Kovda*, 1973; *FAO-UNESCO,* 1989; *Roeschmann,*

1986; *Gerasimov and Ivanova*, 1959 etc.]. The scale of these maps varies from 1:1,000,000 up to 10,000,000. The land use map distinguishes 7 land use types: arable land, grassland for agricultural use, permanent crops, coniferous and mixed forest, broad-leaved forest, urban area and inland waters. It has been received from CCE/RIVM, the Netherlands, in 50 × 50 km² EMEP grid format [*LUGRID DB*, 1995; *Posch et al.*, 1995]. These maps have been used for quantification of various parameters of nitrogen runoff (see below).

5.2. Direct Calculations of Nitrogen Runoff (Method 1)

On the basis of above mentioned conceptual and technical approaches, the following model has been applied to an assessment of values of nitrogen input to the Baltic Sea from its drainage basin [*Bashkin et al.*, 1995]:

$$N_{leach} = \sum_{l=1}^{n} (K_{sl} \times N_{tdl}) \times K_{wl}, \quad (1)$$

where N_{leach} — deposition nitrogen riverine leaching into the Baltic Sea; l — land use types in every elemental soil taxon, accounting soil mechanical composition and slopes; K_{sl} — coefficient of nitrogen runoff in every land use type; N_{tdl} — nitrogen deposition in every land use type; K_{wl} — part of nitrogen passing through various water bodies (rivers, channels, lakes, swamps, ponds etc.) of every land use type.

5.2.1. Nitrogen runoff from terrestrial ecosystems

Coefficient of nitrogen runoff, K_{sl}, was computed for every 2' × 2' LoLa grid cell by following equation:

$$K_{sl} = K_{sr} \times K_{sb} \times C_t, \quad (2)$$

where K_{sr} — coefficients of runoff values for every land use type; K_{sb} — additional dimentionless coefficient accounting for slope and mechanical composition of soils; C_t — active temperature coefficient.

The corresponding values of K_{sr} are determined on the basis of experimental data obtained for every type of land use (forest, wetland, grassland, permanent crops, arable land including fodder crops, cereals etc.) by *G. Nazarov* [1981], *V. Kuznetsov* [1995], *V. K. Kusnecov and D. Balla* [1994] for Gulf of Finland and Gulf of Riga subbasins, *F. Welte and F. Timmermann* [1982], *M. Jansson et al.* [1994], *L. O. Nielsson and K. Wiklund* [1995] and *B. Kronvang et al.* [1995] for Baltic Proper and south-west (SOU) subregions as well as experimental data of other researchers [*Bashkin*, 1987; *Hinga et al.*, 1991; *Critical Loads...*, 1992; *Bashkin et al.*, 1993; *Khrissanov and Osipov*, 1993; *Erdman et al.*, 1993; *Acidification in Finland*, 1990; *De Vries and Jansen*, 1994; *De Vries et al.*, 1995] have been used for interpolation of K_{sl} values for those areas (firstly, the Bothnic Bay and Bothnic Sea as well as different ecosystems in other subbasins) where experimental results are scarce or absent (Table 2).

An additional coefficient has been applied (Table 3) in order to take into account the slopes and soil mechanical composition.

The values of nitrogen deposition in every land use type, N_{tdl}, were interpolated from our calculations of oxidised and reduced nitrogen depositions for every 150 × 150 km² EMEP grid cells (see Part I, RUN96).

Table 2

Average values and limits of deviations for runoff coefficients, K_{sr}, depending upon the land cover in various subbasin areas

Land cover	Subbasin					
	BOB	BOS	GUF	GUR	BAP	SOU
Town	0.60	0.65	0.63	0.65	0.70	0.75
	0.31–0.75	0.31–0.72	0.41–0.83	0.53–0.77	0.48–0.92	0.55–0.95
Forest	0.07	0.09	0.12	0.15	0.13	0.15
	0.01–0.24	0.01–0.21	0.05–0.37	0.05–0.52	0.03–0.60	0.05–0.65
Open lands	0.17	0.20	0.18	0.23	0.22	0.20
	0.05–0.37	0.03–0.43	0.04–0.58	0.05–0.60	0.07–0.57	0.07–0.43
Open waters	0.10	0.10	0.20	0.25	0.30	0.35
	0.03–0.22	0.05–0.22	0.07–0.43	0.05–0.39	0.03–0.97	0.05–0.93
Glaciers*	1.00	1.00	1.00	1.00	1.00	1.00

* No retention was assumed for glaciers.

Finally, the values of K_{sl} were calculated using also the figures of C_t, active temperature coefficient:

$$C_t = \frac{\text{period, } t > 5^\circ C}{\text{round year period}} \quad \text{for land use "Forest" and} \tag{3}$$

$$C_t = \frac{\text{period, } t > 10^\circ C}{\text{round year period}} \quad \text{for land use "Open land" and "Open water".} \tag{4}$$

The values of seasonal temperature coefficient, C_t, characterizing relative degree of intensity of various biogeochemical processes are presented to take into account seasonal deviations. An application of this coefficient shows that, for example, in comparison with summer season, the intensity of N uptake, immobilization and denitrification decreases up to 0.3–0.7 during autumn season, up to 0.2–0.7 in spring and during winter in northern subregion (BOB) these processes are stopped or decreased significantly up to 0.4–0.5 (in SOU subregion).

The calculation of these coefficients was based on various data: as it was mentioned above using sum of active temperatures (> 5°C for natural and > 10°C for agricultural vegetation), as well as continuation of unfrozen period, winter snow melting periods, duration of open water period etc. [*Lvovich,* 1972; *Resources...*, 1972; GRID DBs] was also taken into account.

It should be also taken into account that for land cover type "open water" (lakes and swamps with area more then 2' × 2' LoLa grid) the values of uptake for nitrogen entering directly from atmospheric deposition to the water body, W_s, were calculated using the following determination of the left part of equation 1: $K_s = W_s \times C_t$, where W_s is equal to 0.90 (BOB, BOS), 0.80 (GUF), 0.75 (GUR), 0.70 (BAP) and 0.65 (SOU) and C_t was calculated as this was mentioned above.

Table 3

Additional coefficients, K_{sb}, for land cover "Forest" and "Open land"*

FAO number	Soil index	FAO soil type	Slope, %		
			< 3	3–9	> 9
8	B:	Chromic Cambisols	0.85	1.05	1.25
9	B:	Dystric Cambisols	1.25	1.45	1.65
10	B:	Eutric Cambisols	0.95	1.05	1.15
12	B:	Gleyic Cambisols	0.98	1.08	1.18
15	B:	Vertic Cambisols	1.00	1.20	1.45
19	C:	Haplic Chernozems	1.00	1.20	1.40
20	C:	Calcic Chernozems	1.00	1.20	1.40
21	C:	Luvic Chernozems	1.10	1.30	1.50
22	D:	Podzoluvisols	0.88	1.08	1.28
23	D:	Dystric Podzoluvisols	1.28	1.48	1.68
24	D:	Eutric Podzoluvisols	0.88	1.08	1.28
25	D:	Gleyic Podzoluvisols	0.48	1.58	1.68
26	E:	Rendzinas	0.85	1.05	1.25
36	G:	Dystric Gleysols	0.45	0.55	0.65
37	G:	Eutric Gleysols	0.95	1.05	1.25
38	G:	Humic Gleysols	0.55	0.75	0.95
39	G:	Mollic Gleysols	0.98	1.00	1.18
43	H:	Calcaric Phaeozems	0.85	0.95	1.15
44	H:	Gleyic Phaeozems	0.80	1.00	1.20
45	H:	Haplic Phaeozems	0.85	0.95	1.15
47	R:	Lithosols	1.35	1.60	1.80
49	I:	Calcaric Fluvisols	1.00	1.20	1.40
51	I:	Eustric Fluvisols	1.10	1.30	1.50
52	I:	Thionic Fluvisols	0.30	0.45	0.60
59	L:	Chromic Luvisols	1.30	1.50	1.70
61	L:	Gleyic Luvisols	1.50	1.60	1.75
63	L:	Orthic Luvisols	1.25	1.45	1.65
68	M:	Orthic Greyzems	1.05	1.25	1.45
74	O:	Dystric Histosols	0.60	0.70	0.80
75	O:	Eutric Histosols	0.40	0.50	0.55
79	P:	Gleyic Podzols	0.50	0.60	0.65
80	P:	Humic Podzols	0.45	0.50	0.55
81	P:	Leptic Podzols	1.30	1.50	1.70
82	P:	Orthic Podzols	0.60	0.75	0.80
86	Q:	Cambic Arenosols	1.50	1.60	1.80
88	Q:	Luvic Arenosols	1.55	1.65	1.85
90	R:	Calcaric Regosols	1.50	1.70	1.90
109	W:	Eutric Planosols	0.60	0.70	0.80

* To the values of K_{sb} for Haplic Chernozems at slope < 3% is equal to 1.00.

5.2.2. Nitrogen retention in freshwater ecosystems

Depending on subregion, land use and soil types, hydrological and hydrochemical types of water bodies and especially their bottom sediments, the values of nitrogen retention (N uptake, N_{upt}; N immobolization, N_{im}; N denitrification, N_{de}) during transport by water flows (rivers, channels, lakes etc.), $K_{wret} = 1 - K_{wl}$ were suggested to be altered in limits of 0.03–0.85 accounting for different experimental results [*Jeffries and Semkin,* 1987; *Hinga et al.,* 1991; *Jansson et al.,* 1994; *Ahlgren et al.,* 1994; *Khrissanov and Osipov,* 1993; *Thies,* 1995 etc.].

Values of coefficients K_{wl} in various water sources have been calculated as following:

$$K_{wl} = (1 - K_{wret1}) \times (1 - K_{wret2}) \times (1 - K_{wret3}), \quad (5)$$

where K_{wret1} is retention in open water bodies such as lake and swamps; K_{wret2} is retention in river and channels; K_{wret3} is retention in large rivers.

The values of K_{wret} are dimentionless as a part of nitrogen entering the water body or per cent of the input whereas the values of retention may be expressed as absolute figures, for ex., mg m^{-2} yr^{-1} or kg m^{-2} yr^{-1} etc.

The minimum annual values of K_{wret} equal to 3% of N input to a water body were suggested for small rivers like those in southern Sweden (BAP subbasin) in accordance with experimental results of *M. Jansson* and his colleagues obtained in "WETLAND AND LAKES AS NITROGEN TRAPS" Project [*Jansson,* 1994]. Whereas, permanent retention of nitrogen in the sediments of the lakes was 14–33% and even more of the external loading [*Ahlgren et al.,* 1994] in the same regions. The analysis of input/output mass budgets for lakes carried out in the Ontario river basin showed that the ratio of output to total measured plus estimated input for the 1981/82 and 1982/83 water years for NH_4^+ varied from 0.2 to 0.9 (retention of 10–80%, mean 41%) and for NO_3^- from 0.7 to 0.9 (retention 10–30%, mean 21%) [*Jeffries and Semkin,* 1987].

Taking into account these results, the retention values for nitrogen species during their transport in water bodies were suggested to be equal to < 5–50% of the external loading in water ecosystems in the BAP, GUR, GUF, SOU subbasins where mineral soils are predominant and water bodies are of oligotrophic or mesotrophic types with residence time less than 1 year. The maximum values of K_{wret} up to 70–85% were suggested to be applied to the areas of the BOS and GUF subregions with predominant existence of peats and swamps having waters enriched with organic matter and residence time more than 1.5–2.0 years.

For freshwater ecosystems (lakes, swamps etc.), we assumed that the values of nitrogen retention include nitrogen uptake and immobilization ($N_{upt} + N_{im}$) as well as the values of denitrification. The latters were suggested to be equal 0.05–0.45 of the input, where the minimum values (< 0.1) were suggested to be used in the water bodies with sand bottom sediments and water resistance time less than 3 days, average values (0.1–0.4) for the most frequent cases with sandy loam and loam bottom sediments and water resistance time less than 1 year and the maximum values (> 0.4) for peats and swamps ecosystems with water resistance time more than 1 year [*Wyer and Hill,* 1984; *Bashkin,* 1987; *Jansson et al.,* 1994 etc.]. The type of bottom sediments were determined on the basis of land use and land cover data sets.

For example, mean median retention values during 1989–1993 based on mass balance studies for 37 intensively monitored Danish lakes was 41 ± 25 mg N m^{-2} yr^{-1}, corresponding to 57% ± 25% of the loads to the lakes. Retention in the river Gudena for 1990–1993 was equal to the average value of 54.9% (24.9–88.1%) of the nitrogen gross diffuse load [*Svendson et al.,* 1995].

Thus the calculation of K_{wret1} for land cover "open water" bodies such as lakes and swamps were made up for the integrated resolution 0.5 × 0.5° LoLa grid on the basis of the data presented in Table 4.

Table 4

The values of coefficients of nitrogen retention in lakes and swamps, K_{wret1}

Land use	% of water body area of the total area of 0.5 × 0.5° LoLa cell						
	< 5	5–10	11–20	21–30	31–40	41–50	> 50
Lakes	0.05	0.12	0.18	0.25	0.30	0.45	0.70
Swamps	0.10	0.19	0.35	0.47	0.55	0.68	0.80

Furthermore, for rivers and channels the calculation of K_{wret2} values has been made using the data set from Table 5 in accordance with the following equation:

$$K_{wret2} = N_{de} \times A/Nt_{leach}, \quad (6)$$

where N_{de} — denitrification rate, A — dimentionless coefficient, accounting river density, water flow rate, type of eutrophication process etc.; Nt_{leach} — nitrogen leaching from terrestrial ecosystems, equal to $K_{sl} \times Nt_{dl}$ (see equation 1).

Table 5

The values of parameters for calculating K_{wret2}

Parameters	Subbasin					
	BOB	BOS	GUF	GUR	BAP	SOU
N_{de}, kg km^{-2} yr^{-1}	80	95	120	190	200	240
A	0.4	0.4	0.75	0.60	0.54	0.4

Taking into account that different types of retention processes are more profound in large rivers in comparison with small and middle ones, the additional coefficient K_{wret3} was used for the Vistula and Oder rivers calculated on the basis of denitrification rates, areas of river channels and times of flowing: K_{wret3} for Vistula is equal to 0.25 and for Oder 0.32.

In accordance with Lithuanian data [*Paukstys,* 1993], from the area of the Baltic Proper, Gulf of Riga and Gulf of Finland subbasins which territories belong to Russia, Lithuania, Latvia, Estonia, Belarus and the Ukraine, 96% of precipitation water flows to the seawith riverine discharge and direct coastal surface runoff and only 4% — with groundwater discharge. Similar data were obtained by *R. Schwarze* [1987] for the drainage area of former East Germany. Extrapolating these data to the other part of the Baltic basin, it is possible to ignore the groundwater discharge and to take into account only surface runoff and riverine water discharge. It is also worthy to use Finnish data [*Lepisto and Seuna,* 1990] that show practically full discharge of subsurface and groundwaters to local erosion basis.

There exists a clear need for the development and application of dynamic models capable of predicting the response of soils and surface waters to coupled nitrogen and sulphur deposition scenarios. For example, the MAGIC-WAND model provides such a tool and offers wide

application within the UNECE Convention on Long Range Transboundary Air Pollution and in particular, within the critical load programme. The model describes the major dynamics and transformations of nitrogen within catchment soils and surface waters and couples this to the existing sulphur based model. Ammonium exchange within soils is not yet included to the model. The relatively simple structure of the model and simplicity of calibration provide the scope for regional application at large spatial scales [*Jenkins,* 1995].

However, there is no possibility to run this model and other similar models [see, e. g., *Kamari et al.,* 1989 etc.] due to lack of satisfactory DBs for the whole basin of the Baltic Sea and all calculations for applied model have been computed using steady-state mass-balance approaches.

5.3. Indirect Calculation of Nitrogen Runoff (Method 2)

On a basis of the 2nd approach, the following model has been used for indirect estimation of nitrogen runoff [*Bashkin et al.,* 1995]:

$$N_{leach} = \sum_{l=1}^{n} (N_{tdl} - N_{tretl}) \times K_{wl}; \quad (7)$$

where N_{tretl} — retention of deposition nitrogen in terrestrial ecosystems and other parameters are the same as in equation 1.

5.3.1. Nitrogen retention in terrestrial ecosystems

On the basis of existing approaches [*Calculation* ..., 1993] N retention in terrestrial ecosystems was calculated using following equation:

$$N_{tret} = (N_{upt} + N_{im} + N_{de}) \times C_t, \quad (8)$$

where N_{upt} — atmospheric deposition nitrogen uptake by vegetation; N_{im} — atmospheric deposition nitrogen immobilization; N_{de} — atmospheric deposition nitrogen denitrification; C_t — active temperature coefficient. The values of nitrogen uptake were determined by following equation:

$$N_{upt} = (K_1\ NO_x + K_2\ NH_3)_{wet} + (K_3\ NO_x + K_4\ NH_3)_{dry}, \quad (9)$$

where indices *wet* and *dry* mean wet and dry deposition, and values of $K_1 - K_4$ are given in Table 6 for various soil groups, their nitrogen mineralizing capacity (NMC), C:N ratio and different land use types. The values of nitrogen mineralizing capacity (NMC) were determined on the basis of experimental results and computer calculations [*Legg et al.,* 1971; *Jenkinson,* 1982; *Bashkin,* 1987; *Critical loads...,* 1992].

The values of nitrogen immobilization into soil organic matter were determined on the basis of following equation:

$$N_{im} = (K_5\ NO_x + K_6\ NH_3)_{wet} + (K_7\ NO_x + K_8\ NH_3)_{dry}, \quad (10)$$

The coefficients $K_5 - K_8$ are given in Table 7.

The values of Table 7 are determined on the basis of experimental results [*Bashkin,* 1987; *Kudeyarov,* 1989; *Critical Loads...,* 1992].

Table 6

Nitrogen retention in terrestrial ecosystems v/v soil properties

Soil	Land use	C : N	NMC kg N/ha	Nuptake			
				K_1	K_2	K_3	K_4
Peats	Swamps	> 30	70	0.20	0.15	0.15	0.10
Podzols	Forest	> 30	40	0.30	0.25	0.25	0.15
	Arable	< 10	25	0.30	0.15	0.20	0.10
	Pasture	15–25	50	0.35	0.20	0.20	0.15
Luvisols	Forest	> 30	45	0.35	0.25	0.25	0.20
	Arable	< 10	25	0.30	0.20	0.25	0.15
	Pasture	15–25	55	0.35	0.20	0.25	0.20
Rendzinas	Forest	15–25	90	0.40	0.30	0.25	0.20
	Arable	10–12	70	0.30	0.20	0.25	0.20
	Pasture	15–25	95	0.30	0.25	0.25	0.20
Fluvisols	Forest	> 30	70	0.35	0.25	0.30	0.20
	Arable	10–12	85	0.25	0.20	0.25	0.20
	Pasture	15–25	90	0.30	0.20	0.30	0.15
Cambisols	Forest	15–25	80	0.35	0.25	0.30	0.20
	Arable	< 10	90	0.30	0.20	0.25	0.20
	Pasture	10–15	100	0.35	0.25	0.30	0.20
Chernozems	Forest	15–25	100	0.35	0.25	0.30	0.25
	Arable	< 10	90	0.25	0.20	0.25	0.20
	Pasture	10–15	120	0.30	0.25	0.30	0.25

Table 7

Relationships between values of C : N ratio in upper humus layer and immobilization of atmospheric precipitation nitrogen

C : N	Wet deposition		Dry deposition	
	K_5/NO_x	K_6/NH_3	K_7/NO_x	K_8/NH_3
< 10	0.1	0.2	0.2	0.3
10–15	0.2	0.3	0.3	0.4
15–20	0.3	0.4	0.4	0.6
> 20	0.4	0.6	0.5	0.7

Values of nitrogen denitrification were determined by following equations:

$$N_{de} = K_9 N_{td}, \quad (11)$$

where

$$K_9 = K_{10} [(0.145NMC + 6.447)/NMC]. \quad (12)$$

The values of NMC are shown in Table 6 and the values of K_{10} were related to mechanical composition of organic and mineral soils: peats — 0.8; sand soils — 0.1; loam soils — 0.5; clay soils — 0.7. These values are similar to those used for the calculation of nitrogen critical loads [*Critical loads...*, 1992; *Calculation...*, 1993].

The computed results obtained for N_{tret} values for different land use and soil types were compared with those available in literature. The atmospheric N input in Europe and North America increased dramatically during recent decades due to emission of NO_x from combustion processes and of NH_3 from agricultural activities. The N deposition to forest ecosystems generally exceeded in the 1980ies 20 kg ha^{-1} yr^{-1} in the central part of Europe — even reached 100 kg ha^{-1} yr^{-1} in some impact areas [*Ivens et al.*, 1990; *Hauhs et al.*, 1989; *Gudrersen and Bashkin*, 1994]. Forest ecosystems may accumulate a considerable amount of N in biomass and soil organic matter, but there is a concept that forest ecosystems may be overloaded with N from atmospheric deposition leading to increasing leaching of nitrates and biodiversity change and to so-called "nitrogen saturation" occurrence.

The nitrogen saturation can be defined as a situation in which the supply of inorganic nitrogen exceeds the nutritional demand of biota and its operationally measured as increasing leaching of N below the rooting zone [*Aber et al.*, 1989]. The review of nitrogen saturation in forest ecosystems presented by *P. Gudrersen* and *V. Bashkin* [1994] shows that the values of nitrogen accumulation may be in the limits of 1–142 kg ha^{-1} yr^{-1} but the most statistically significant values for the forest ecosystems of the Central and Northwestern Europe were between 6–24 kg ha^{-1} yr^{-1}.

At present great attention is payed to an assessment of wetlands as intermediate ecosystems between terrestrial and water ones. Nitrogen retention capacity of different types of wetlands has been studied in project "WETLAND AND LAKES AS NITROGEN TRAPS" headed by *M. Jansson* at the beginning of the 1990s. The results demonstrated that N removal from wetlands depended mainly on denitrification. In addition to denitrification, sedimentation can be quantitatively important during water flooding periods. Large-scale establishment in the agricultural areas in southern Sweden with a great number of wetlands within the catchment may reduce the nitrogen transport by up to about 15%. In most cases, the result will be probably less. Furthermore, in addition to researches of nitrogen removal in wetlands, suitable for use in farmland areas, a study of the retention capacity of forest wetlands showed that N losses from forest (1–5 kg ha^{-1} yr^{-1}) are considerably lower than from arable lands [*Jansson et al.*, 1994]. In case of southern Sweden, where the forest ecosystems make up a major part of the catchments draining to the Baltic Sea coast, nitrogen derived from forest areas is as much as 50% of the total N input. The authors concluded that the N removal capacity of forest wetlands is more or less unknown but could be considerably high under the increasing input amounts of nitrogen [*Jacks et al.*, 1994].

The values of possible (potential) retention of N in different ecosystems depending upon the land cover and land use have been also compared with those suggested for the empirical N critical loads [*Bobbink and Roelofs*, 1995] and for natural N leaching [*Kuylenstierna and Chadwick*, 1992].

Resuming this part, one can conclude that the values of retention of atmospheric N deposition in different ecosystems obtained experimentally can be significant, up to 30–40 kg ha^{-1} yr^{-1} in agricultural and managed forest ecosystems, but both computed and experimental reasonable values were in the limits of 5–25 kg ha^{-1} yr^{-1}.

5.4. Fertilizer-Based Calculation of Nitrogen Runoff (Method 3)

The algorithm of calculations for method 3 is based on the following items:

1. Only experimental data have been used for calculation of nitrogen runoff depending on land use;
2. An assumption on the proportional runoff of deposition and fertilizer nitrogen from agricultural landscapes has been accepted;

3. No fertilizer application has been assumed for the land cover of "forest" type;
4. The resolution scale has been adopted at the country/subbasin level, because the initial data for fertilizer application were presented for the minimum scale of administrative districts.

Furthermore, the results of riverine input of nitrogen computed by methods 1 and 2 were compared with similar data calculated on the basis of DBs on mineral and fertilizer application as well as on nitrogen runoff from different land-use types experimentally obtained in various European countries and reviewed by *V. K. Kuznetsov* [*Kuznetsov, pers. comm.*, 1995; *Kusnecov und Balla*, 1994] and *L. M. Svendsen et al.*, 1995]. In accordance with these results, N runoff values from agricultural landscapes are of limits 1.48–30.8 (mean 10.2) kg ha^{-1} yr^{-1}, from urban areas — 0.12–10.0 (2.41) kg ha^{-1} yr^{-1}, from forested ones with nitrogen input less than 30 kg ha^{-1} yr^{-1} — 0.06–4.90 (1.88) kg ha^{-1} yr^{-1}.

When estimating the losses from fields, Finish specialists [*Knnuttila and Salo,* see *Svendsen et al.*, 1995] assumed that the loss from forests and the natural loss from presently cultivated land is 2.5 kg N ha^{-1} yr^{-1}. This assumption is based on the monitoring results from forested basins. The natural transport and the loss from forests were then subtracted from the total loss values for each basin. For the agricultural basins the specific loss from cultivated land was estimated to be from 18 to 20 kg N ha^{-1} yr^{-1}. After subtracting the background loss as well as industrial and municipal total river fluxes, in 4 Finish river basins the agricultural nitrogen loads varied from 11 to 22 kg ha^{-1} yr^{-1}. Taking into account uncertainties (many processes such as sedimentation, channel erosion, denitrification, uptake, immobilisation and other biological processes have not been considered), the net input of nitrogen to the Baltic Sea with coastal rivers can be both underestimated and overestimated. Nevertheless, an average value of 16 kg ha^{-1} yr^{-1} can be applied to Finnish agricultural areas.

The background load of nitrogen in Denmark was determined from measurements of the nutrient fluxes from 10 small non-agricultural catchments. They were subdivided into sandy and loamy catchments. The mean median figure for 9 to 10 natural watersheds during 1989–1993 was 2.0 ± 0.2 kg N ha^{-1} yr^{-1} but for one catchment of the Gudena river these values varied from 2.92 to 3.73 kg ha^{-1} yr^{-1}. The corresponding loss from agricultural fields was 21.50–25.36 kg ha^{-1} yr^{-1} in 1990–1991 [*Svendsen et al.*, 1995]. *B. Kronvang et al.* [1995] calculated that export coefficients for nitrogen were equal to 2.1–2.3 kg ha^{-1} yr^{-1} for Danish non-agricultural land and to 11.4–21.6 kg ha^{-1} yr^{-1} for agricultural land (1989–1990). For Danish area belonging to the Baltic Sea drainage basin, the values of 17.3 kg ha^{-1} yr^{-1} were calculated for cultivated land in 1989 and 2.3 kg ha^{-1} yr^{-1} for natural loamy areas.

The DBs on fertilizer application during 1987–1991 were formed on the basis of original data received from countries at HELCOM request.

6. RESULTS AND DISCUSSION

6.1. Computed Values of Atmospheric Deposition Nitrogen Runoff

Figures 2–6 and Tables 8–12 show the calculations of absolute and relative values of input of atmospheric nitrogen deposition with riverine runoff into the Baltic Sea basin from its drainage basin. The calculations were made by methods 1 and 2 for each 2' × 2' LoLa grid cell and grouped for each cell of EMEP 150 × 150 km^2 grid using RUN96 deposition values. The initial results for 2' × 2' LoLa grid cells were very similar for both applied methods. The results for EMEP (150 × 150 km^2) cells as well as total results for subbasin/country levels are shown only for method 1.

In dependence on subregion the part of atmospheric deposition nitrogen which entered the Baltic Sea basin changed from 5.1 up to 9.7%. Minimum values were obtained for northern subregions (BOB and BOS) with high forestation and the longest winter period. On the contrary, maximum values were obtained for subregion SOU and BAP with the lowest forestation and the highest degree of arable lands and mild climate. On the average, about 8% of atmospheric nitrogen deposition on the watershed area (1408 kt) entered the Baltic Sea during 1987–1991.

The comparison of these results with those computed by method 3 is shown in Table 13. One can see that the values for different countries and subbasins are of the same order of magnitude both for absolute figures of atmospheric riverine N input (kilotonnes) and relative ones (per cent).

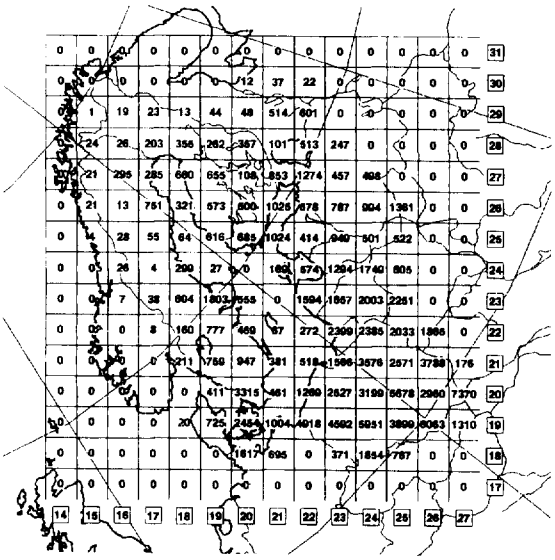

Fig. 2. Map of deposition nitrogen runoff values, t/yr/EMEP cell in 1987

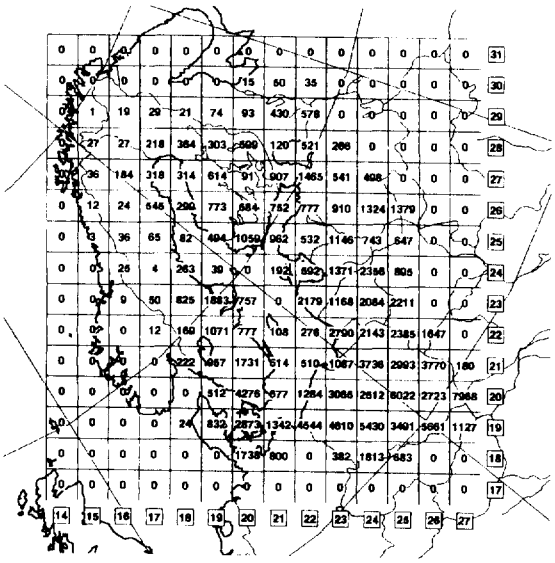

Fig. 3. Map of deposition nitrogen runoff values, t/yr/EMEP cell in 1988

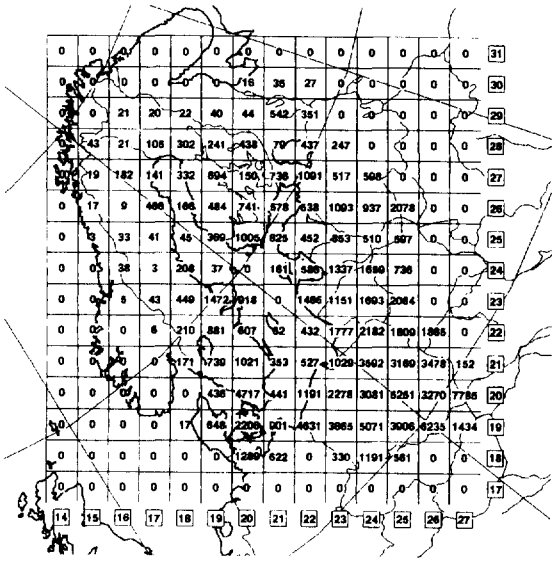

Fig. 4. Map of deposition nitrogen runoff values, t/yr/EMEP cell in 1989

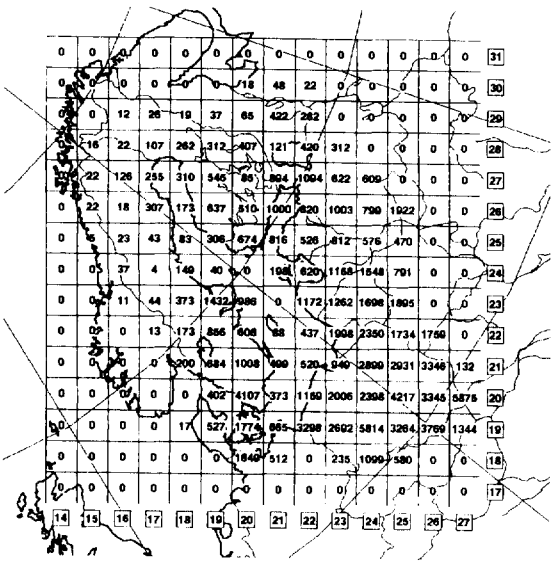

Fig. 5. Map of deposition nitrogen runoff values, t/yr/EMEP cell in 1990

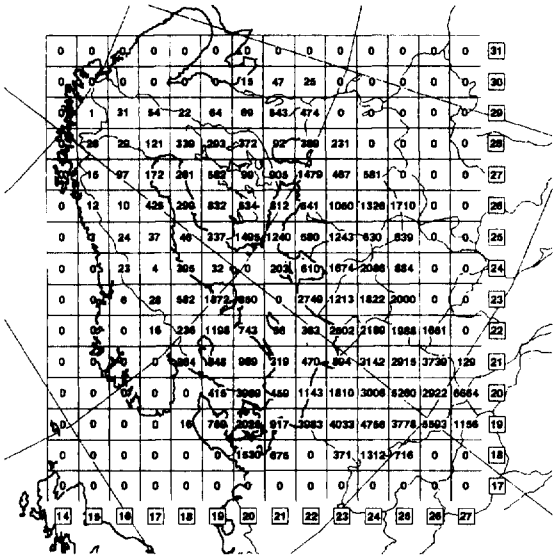

Fig. 6. Map of deposition nitrogen runoff values, t/yr/EMEP cell in 1991

Table 8

The cumulated values of nitrogen deposition and runoff for subbasin/country in 1987

W	C	S, km^2	N_{dep}, kt	N_{run}, kt	N_{run} : N_{dep}
SOU	DM	31500	41.3888	4.0292	0.0974
SOU	GER	11025	22.1020	2.3115	0.1046
SOU	SW	74250	48.5096	3.7305	0.0769
SOU			112.00	10.07	0.090
GUR	BEL	25875	18.1148	1.5337	0.0847
GUR	LAT	48600	40.2874	2.4815	0.0616
GUR	EST	17550	11.7677	0.6871	0.0584
GUR	RUS	22500	12.5415	1.6044	0.1279
GUR	LIT	16500	18.4857	1.6686	0.0902
GUR			101.20	7.97	0.078
GUF	LAT	3375	2.2489	0.2696	0.1199
GUF	EST	26325	16.6570	1.1090	0.0666
GUF	RUS	276075	143.5203	7.1472	0.0498
GUF	FIN	106650	47.8384	2.7983	0.0585
GUF			210.26	11.32	0.054
BOS	FIN	47925	18.5144	1.9168	0.1035
BOS	SW	180000	53.8256	2.6227	0.0487
BOS			72.34	3.82	0.053
BOB	FIN	146025	36.8316	2.2148	0.0601
BOB	SW	130950	25.5368	1.2538	0.0491
BOB			62.37	3.47	0.056
BAP	SLO	8550	20.0903	4.5573	0.2268
BAP	UKR	11025	17.6989	2.3442	0.1324
BAP	SW	84825	56.6957	4.2084	0.0742
BAP	DM	1125	1.8405	0.2146	0.1166
BAP	GER	12600	23.7578	3.5402	0.1490

Table 8 (continued)

W	C	S, km²	N_{dep}, kt	N_{run}, kt	$N_{run} : N_{dep}$
BAP	POL	311850	610.8435	58.2439	0.0953
BAP	LIT	48375	58.0619	3.0222	0.0521
BAP	LAT	12600	11.2217	1.2723	0.1134
BAP	EST	1125	0.9068	0.0673	0.0742
BAP	RUS	15075	23.4869	2.7799	0.1184
BAP	BEL	46800	53.0248	6.2970	0.1188
BAP			877.63	86.54	0.099
Total			1435.79	123.20	0.086

Table 9

The cumulated values of nitrogen deposition and runoff for subbasin/country in 1988

W	C	S, km²	N_{dep}, kt	N_{run}, kt	$N_{run} : N_{dep}$
SOU	DM	31500	49.5495	4.8114	0.0971
SOU	GER	11025	24.2251	2.5380	0.1048
SOU	SW	74250	62.8837	4.8935	0.0778
SOU			136.66	12.24	0.090
GUR	BEL	25875	24.6420	2.0752	0.0842
GUR	LAT	48600	45.2858	2.7197	0.0601
GUR	EST	17550	12.9411	0.7416	0.0573
GUR	RUS	22500	13.6890	1.7183	0.1255
GUR	LIT	16500	17.5441	1.5539	0.0886
GUR			114.10	8.80	0.077
GUF	LAT	3375	3.1028	0.3712	0.1196
GUF	EST	26325	18.5962	1.1980	0.0644
GUF	RUS	276075	167.2666	8.0529	0.0481
GUF	FIN	106650	47.7556	2.8090	0.0588
GUF			236.72	12.53	0.053
BOS	FIN	47925	21.8441	2.2650	0.1037
BOS	SW	180000	61.2792	2.9584	0.0483
BOS			83.12	5.22	0.053
BOB	FIN	146025	36.4660	2.0719	0.0568
BOB	SW	130950	24.6845	1.0376	0.0420
BOB			61.15	3.11	0.051
BAP	SLO	8550	19.7820	4.4811	0.2265
BAP	UKR	11025	17.2717	2.3211	0.1344
BAP	SW	84825	83.4968	6.0180	0.0721
BAP	DM	1125	2.1555	0.2513	0.1166
BAP	GER	12600	25.7814	3.6856	0.1430
BAP	POL	311850	599.3658	57.2019	0.0954
BAP	LIT	48375	59.2756	3.1309	0.0528
BAP	LAT	12600	14.8531	1.6623	0.1119
BAP	EST	1125	1.4614	0.1084	0.0742
BAP	RUS	15075	25.7400	3.0049	0.1167
BAP	BEL	46800	58.4456	6.6902	0.1145
BAP			907.62	88.56	0.097
Total			1538.77	130.46	0.085

Table 10

The cumulated values of nitrogen deposition and runoff for subbasin/country in 1989

W	C	S, km²	N_{dep}, kt	N_{run}, kt	$N_{run} : N_{dep}$
SOU	DM	31500	39.3975	3.8152	0.0968
SOU	GER	11025	21.0656	2.2046	0.1047
SOU	SW	74250	60.6773	4.5137	0.0744
SOU			121.14	10.53	0.087
GUR	BEL	25875	22.7205	1.9298	0.0849
GUR	LAT	48600	52.1942	3.1673	0.0607
GUR	EST	17550	14.6421	0.8417	0.0575
GUR	RUS	22500	15.9975	2.0386	0.1274
GUR	LIT	16500	18.1486	1.5626	0.0861
GUR			123.70	9.54	0.077
GUF	LAT	3375	2.8688	0.3440	0.1199
GUF	EST	26325	18.6034	1.2156	0.0653
GUF	RUS	276075	159.1515	7.9156	0.0497
GUF	FIN	106650	46.3774	2.7142	0.0585
GUF			227.00	12.19	0.054
BOS	FIN	47925	23.9083	2.5222	0.1055
BOS	SW	180000	50.2751	2.7390	0.0545
BOS			74.18	5.26	0.071
BOB	FIN	146025	37.2638	1.9766	0.0530
BOB	SW	130950	15.6708	0.6753	0.0431
BOB			52.93	2.65	0.050
BAP	SLO	8550	18.1449	4.1121	0.2266
BAP	UKR	11025	16.4057	2.1727	0.1324
BAP	SW	84825	69.7995	5.0422	0.0722
BAP	DM	1125	1.5188	0.1771	0.1166
BAP	GER	12600	20.2023	2.9712	0.1471
BAP	POL	311850	548.5086	52.0521	0.0949
BAP	LIT	48375	56.3888	2.9331	0.0520
BAP	LAT	12600	18.5674	2.0931	0.1127
BAP	EST	1125	0.7605	0.0564	0.0742
BAP	RUS	15075	23.7510	2.7914	0.1175
BAP	BEL	46800	52.8093	6.1083	0.1157
BAP			826.86	80.51	0.097
Total			1425.81	120.68	0.085

Table 11

The cumulated values of nitrogen deposition and runoff for subbasin/country in 1990

W	C	S, km^2	N_{dep}, kt	N_{run}, kt	$N_{run} : N_{dep}$
SOU	DM	31500	41.7116	4.0441	0.0970
SOU	GER	11025	18.2180	1.9109	0.1049
SOU	SW	74250	57.0478	4.7369	0.0830
SOU			116.98	10.69	0.091
GUR	BEL	25875	19.7111	1.6640	0.0844
GUR	LAT	48600	39.5361	2.4325	0.0615
GUR	EST	17550	11.2325	0.6462	0.0575
GUR	RUS	22500	17.3520	2.2800	0.1314
GUR	LIT	16500	15.8971	1.2171	0.0765
GUR			103.73	8.24	0.079
GUF	LAT	3375	2.1881	0.2620	0.1197
GUF	EST	26325	14.5712	0.9410	0.0646
GUF	RUS	276075	141.7986	6.9336	0.0489
GUF	FIN	106650	49.4435	2.5393	0.0514
GUF			208.00	10.68	0.051
BOS	FIN	47925	19.5667	2.0503	0.1048
BOS	SW	180000	46.0280	2.2833	0.0496
BOS			65.59	4.33	0.066
BOB	FIN	146025	28.8209	1.6069	0.0558
BOB	SW	130950	18.1494	0.7718	0.0425
BOB			46.97	2.38	0.051
BAP	SLO	8550	21.2823	4.8342	0.2271
BAP	UKR	11025	16.9394	2.2556	0.1332
BAP	SW	84825	63.1607	4.6116	0.0730
BAP	DM	1125	1.6549	0.1930	0.1166
BAP	GER	12600	21.9663	3.2899	0.1498
BAP	POL	311850	576.0225	54.8453	0.0952
BAP	LIT	48375	52.0661	2.6981	0.0518
BAP	LAT	12600	10.8990	1.2993	0.1192
BAP	EST	1125	0.8415	0.0624	0.0742
BAP	RUS	15075	18.4331	2.2092	0.1198
BAP	BEL	46800	50.4562	5.9920	0.1188
BAP			833.72	82.29	0.099
Total			1374.99	118.61	0.086

Table 12

The cumulated values of nitrogen deposition and runoff for subbasin/country in 1991

W	C	S, km²	N_{dep}, kt	N_{run}, kt	$N_{run} : N_{dep}$
SOU	DM	31500	35.1821	3.3714	0.0958
SOU	GER	11025	22.7981	2.3610	0.1036
SOU	SW	74250	52.7699	4.2683	0.0809
SOU			110.75	10.00	0.090
GUR	BEL	25875	19.0946	1.5652	0.0820
GUR	LAT	48600	37.1185	2.2392	0.0603
GUR	EST	17550	12.5003	0.7152	0.0572
GUR	RUS	22500	15.8445	2.0898	0.1319
GUR	LIT	16500	14.7219	1.2608	0.0856
GUR			99.28	7.87	0.079
GUF	LAT	3375	2.3366	0.2793	0.1195
GUF	EST	26325	16.8835	1.0993	0.0651
GUF	RUS	276075	147.1770	6.9027	0.0469
GUF	FIN	106650	44.8443	2.6759	0.0597
GUF			211.24	10.96	0.052
BOS	FIN	47925	15.3338	1.5547	0.1014
BOS	SW	180000	43.8793	2.1599	0.0492
BOS			59.21	3.71	0.063
BOB	FIN	146025	28.7114	1.6591	0.0578
BOB	SW	130950	17.8893	0.6997	0.039
BOB			46.60	2.36	0.051
BAP	SLO	8550	16.3346	3.7453	0.2293
BAP	UKR	11025	15.5714	2.0365	0.1308
BAP	SW	84825	62.5761	4.6004	0.0735
BAP	DM	1125	1.3309	0.1552	0.1166
BAP	GER	12600	15.8558	2.3658	0.1492
BAP	POL	311850	500.1183	46.9430	0.0939
BAP	LIT	48375	51.4499	2.6724	0.0519
BAP	LAT	12600	9.0243	1.0979	0.1217
BAP	EST	1125	1.1891	0.0882	0.0742
BAP	RUS	15075	20.4739	2.4473	0.1195
BAP	BEL	46800	48.2713	5.7035	0.1182
BAP			742.20	71.86	0.097
Total			1269.28	106.75	0.084

Table 13

Input of atmospheric nitrogen into the Baltic Sea basin from watershed areas of different countries in average for 1987–1991, kt/%

Country	M*	Subregions																	
		BOB			BAP			SOU			GUR			GUF			BOS		
		1	2	3	1	2	3	1	2	3	1	2	3	1	2	3	1	2	3
Russia	1	–	–	–	3	22	12	–	–	–	2	16	13	7	152	5	–	–	–
	2	–	–	–	2	22	9	–	–	–	3	16	18	7	152	5	–	–	–
	3	–	–	–	2	22	9	–	–	–	4	16	22	10	152	6	–	–	–
Belarus	1	–	–	–	6	53	11	–	–	–	2	21	9	–	–	–	–	–	–
	2	–	–	–	4	53	8	–	–	–	4	21	19	–	–	–	–	–	–
	3	–	–	–	7	53	13	–	–	–	5	21	22	–	–	–	–	–	–
Czech	1	–	–	–	4	19	21	–	–	–	–	–	–	–	–	–	–	–	–
Slovakia	2	–	–	–	3	19	16	–	–	–	–	–	–	–	–	–	–	–	–
	3	–	–	–	3	19	16	–	–	–	–	–	–	–	–	–	–	–	–
Finland	1	2	34	6	–	–	–	–	–	–	–	–	–	3	47	6	2	20	10
	2	3	34	9	–	–	–	–	–	–	–	–	–	4	47	9	2	20	10
	3	3	34	9	–	–	–	–	–	–	–	–	–	3	47	8	2	20	10
Estonia	1	–	–	–	0.1	1	7	–	–	–	1	13	6	1	17	7	–	–	–
	2	–	–	–	0.1	1	7	–	–	–	2	13	13	2	17	11	–	–	–
	3	–	–	–	0.1	1	7	–	–	–	1	13	10	2	17	10	–	–	–
Latvia	1	–	–	–	1	13	8	–	–	–	3	43	6	–	–	–	–	–	–
	2	–	–	–	1	13	8	–	–	–	4	43	9	–	–	–	–	–	–
	3	–	–	–	1	13	8	–	–	–	5	43	11	–	–	–	–	–	–
Lithuania	1	–	–	–	3	55	5	–	–	–	1	17	6	–	–	–	–	–	–
	2	–	–	–	3	55	5	–	–	–	1	17	6	–	–	–	–	–	–
	3	–	–	–	5	55	9	–	–	–	2	17	10	–	–	–	–	–	–
Poland	1	–	–	–	51	567	9	–	–	–	–	–	–	–	–	–	–	–	–
	2	–	–	–	54	567	9	–	–	–	–	–	–	–	–	–	–	–	–
	3	–	–	–	31	567	5	–	–	–	–	–	–	–	–	–	–	–	–
Germany	1	–	–	–	3	22	15	2	22	10	–	–	–	–	–	–	–	–	–
	2	–	–	–	2	22	11	3	22	13	–	–	–	–	–	–	–	–	–
	3	–	–	–	2	22	11	2	22	11	–	–	–	–	–	–	–	–	–
Denmark	1	–	–	–	0.2	2	11	4	41	9	–	–	–	–	–	–	–	–	–
	2	–	–	–	0.1	2	9	6	41	15	–	–	–	–	–	–	–	–	–
	3	–	–	–	0.1	2	7	3	41	7	–	–	–	–	–	–	–	–	–
Sweden	1	1	20	5	5	67	7	5	56	8	–	–	–	–	–	–	2	51	5
	2	2	20	10	7	65	10	8	56	14	–	–	–	–	–	–	4	51	8
	3	3	20	13	3	65	5	5	56	8	–	–	–	–	–	–	4	51	8
Ukraine	1	–	–	–	2	17	13	–	–	–	–	–	–	–	–	–	–	–	–
	2	–	–	–	1	17	8	–	–	–	–	–	–	–	–	–	–	–	–
	3	–	–	–	1	17	8	–	–	–	–	–	–	–	–	–	–	–	–
Total	1	3	54	6	78	838	9	11	119	9	9	110	8	11	216	5	4	71	6
Subregion	2	5	54	9	77	838	9	17	119	14	14	110	13	13	216	6	6	71	8
	3	6	54	11	55	838	7	10	119	8	17	110	16	15	216	7	6	71	8
Mean		5	54	9	70	838	8	13	119	11	13	110	12	13	216	6	6	71	8

Footnote: M*— method of calculation:
1 — nitrogen runoff, kt;
2 — nitrogen deposition, kt;
3 — nitrogen runoff, % from nitrogen deposition.

6.2. Comparison of Computed and Experimental Results for Forested Ecosystems

At present there is an agreement that the quantitative experimental assessment of the partition of atmospheric deposition nitrogen leached from the ecosystems is very difficult to make up due to many immobilization-mobilization processes in internal nitrogen biogeochemical cycle complicating the final values. However, in small forested catchments where the atmospheric N deposition is the only source of this nutrient, one can estimate the ratio between input and output of a given element [*Moldan and Cherny,* 1994]. Using so called "black box" method, the required values were estimated in small catchment experiments located in different places of Europe. The measurement of inputs and outputs of nitrogen in hydrologically and geologically well-defined drainage basins is a powerful tool for analysing the processes which determine the biogeochemical mass balance of this element [*Paches,* 1984; *Bashkin et al.,* 1984; *Bashkin,* 1987; *Kallio and Kauppi,* 1990; *Moldan and Cherny*, 1994 etc.].

Consequently, using this approach the element cycling has been quantified on a considerable number of sites during the last decade.

The potential value of the empirical information was illustrated by a compilation of N data from outgoing investigations in 10 coniferous forest sites over a pollution gradient in Europe [*Tietema and Beier*, 1995]. The continuation of these experimental trials' compilations was done under the project "Element Cycling and Output-fluxes in Forest Ecosystems in Europe" — ECOFEE and the results from a preliminary analysis of data on N cycling have been published [*Gundersen*, 1995a]. It was shown that the total inorganic N input ranged from 3 to 34 kg N ha^{-1} yr^{-1} in bulk deposition and from 2 to 64 kg ha^{-1} yr^{-1} in throughfall.

It was suggested that forest ecosystems would respond to permanent N input by increasing the internal cycling of this element, especially such links of biogeochemical cycling as biomass return, mineralization and litterfall fluxes would increase their capacity and turnover rate [*Aber et al.*, 1989]. The results of ECOFEE project testified the effect of N input on the litterfall flux. Of all the sites included in ECOFEE, 60% leached more than 5 kg N ha^{-1} yr^{-1}. [*Gundersen*, 1995b]. Elevated nitrate leaching appeared in inputs above 10–12 kg N ha^{-1} yr^{-1} and only young stands retained all inputs up to 30 kg N ha^{-1} yr^{-1}. Increased nitrate leaching with increasing stand age was also observed in Wales [*Emmett et al.*, 1995]. These authors remark that enhanced nitrate leaching losses (5–35 kg N ha^{-1} yr^{-1}) in excess of those expected from pristine sites (< 5 kg N ha^{-1} yr^{-1}) were observed from all stands greater than 25–30 years of age. Under the conditions of nitrogen saturation (i.e. the leaching > 5 kg N ha^{-1} yr^{-1}) the nitrate leaching was proportional to % N in forest floor horizons. Similarly, at annual inputs of less than about 10 kg N ha^{-1} yr^{-1} nearly all nitrogen is retained and outputs are very small [*Tietema et al.*, 1995].

Studying the biogeochemical fluxes in plantation forests on acid soils — the EXMAN project (Klosterhede, Denmark; Kootwijk and Harderwijk, the Netherlands; Hoglwald and Solling, Germany) *T.Cummins* and co-workers [1995] showed that nitrogen inputs were higher than nitrate-N outputs in five of the sites, and it is likely that these sites were accumulating nitrogen. At one site, Hoglwald, nitrogen output is considerably in excess of the current input, and this site probably has reached saturation and was leaking nitrogen. The same situation was observed in ECOFEE project, where four sites leached more nitrogen than input. It might be due to a disruption of the N cycle [*Gundersen and Bashkin*, 1994; *Gundersen,* 1995b]. Possible explanations could be the following. At one of these sites, leaching of 45 kg N ha^{-1} yr^{-1} was connected with an insect pest [*Pedersen*, 1993]. At the site with leaching 50 kg N ha^{-1} yr^{-1}, tree species are changed from beech to spruce seemed to cause a release of soil born N. Two Danish sites on calcareous soils with very high nitrification rates were leaching 75–85 kg N ha^{-1} yr^{-1}.

However, one should remember that differences from the input may be within the range of uncertainty of the flux estimates.

P. Gundersen [1995b] has suggested to cluster the sites leaching nitrates in two groups: one group of sites with inputs of 15–25 kg N ha^{-1} yr^{-1} that leached almost all the N input, and another group of sites with inputs of 40-60 kg N ha^{-1} yr^{-1} that leached only c. 50% of the input.

The main difference between these two groups appeared to be in the fraction of ammonium in the input. High inputs above 40 kg N ha^{-1} yr^{-1} were generated from high ammonium depositions. Deposition of oxidised nitrogen usually only contributed with 10–15 kg N ha^{-1} yr^{-1} of the input. High ammonium inputs to some extent could be retained by soil and vegetation, whereas the retention of nitrate seems to be low.

Similar results were obtained in study of the fluxes of N in bulk precipitation, throughfall and stemflow as well as input of nitrogen to the Ratanica forested catchment (South Poland), which is exposed to moderate anthropogenic pollution [*Szarek,* 1995]. It has been shown that the main part of inputting nitrogen was in the form of ammonium N causing the significant leaching of this element.

Thus the analysis of the input-output data allows to draw the conclusion that at inputs above 10–15 kg N ha^{-1} yr^{-1} elevated nitrate leaching can be found in some forest ecosystems. New catchment scale data from UN-ECE, Integrated Co-operative Programme (ICP) on Integrated Monitoring (IM) also confirmed the input threshold at c.10 kg N ha^{-1} yr^{-1} when nitrate leaching started to increase. A regional survey of throughfall deposition and nitrate concentrations in soil water on 60 forest sites in South Sweden showed a picture similar to that of whole European scale. Nitrate concentrations in soil water were close to zero at inputs below 15 kg N ha^{-1} yr^{-1} and elevated at inputs above 22 kg N ha^{-1} yr^{-1} [*Westling,* 1991].

However, the relationships between ecosystem characteristics and leaching are not conclusive. Ammonium dominated sites could often retain c. 50% of the N input even at high input levels, whereas more nitrate dominated sites at moderate inputs had low N retention [*Gundersen,* 1995b]. This emphasizes the mobility of the nitrate ion and indicates that capacity to retain nitrate in forests is relatively low. It was suggested that the internal N cycle would accelerate in an integrated response of leaching processes. Relatively close relation between input and output of N observed in experimental studies may lead to a conclusion that forest ecosystems can leach considerable amounts of nitrate even before they achieve the saturation stage in a biological cycle.

Relatively close relation between input and output was also emphasized in experiments with decreased N deposition at saturated sites. At such sites nitrate leaching decreased immediately after building a roof construction to remove the N input. The response of soil solution chemistry to reduced inputs of N (and S) was strong and fairly rapid [*Bredemeier et al.*, 1995]. The same conclusion is true for the case of reducing N input due to decreased nitrogen emission by the industrial complex in Novgorod, Russia [*Makarov and Kiseleva,* 1995]. In a short time it resulted in decreasing nitrification activity and in corresponding significant changes of soil solution composition and riverine N runoff.

Under the study during 1985–1987 carried out by *K. Kallio and L. Kauppi* [1990] and aimed at quantitative assessment of the ion budgets of 5 small forest basins in Finland belonging to the BOB, BOS, GUF and partly to BAP subbasins, it has been determined that the percentage of nitrogen output from such type of catchments was about of 5% in Myllypuro (BOB) experimental plot (our computed value for this area was 5.9%), of 3.5% in Kesseninpuro (GUF)

one (our computed data were 7.1%), of 8.8% in Huhtisuonoja (GUF) one (our data were 7.3%), of 20% and 15% (mean 17.5%) in Teeressuonoja and Yli-Knuutila experimental plots situated in south-west Finland nearby the BAP subbasin border (our computed data for this area were 8.2%). Thus, these values were of the same order of magnitude as those computed by both our methods for the corresponding areas (Table 14).

Table 14

Comparative assessment of experimental and computed nitrogen runoff values from different forest ecosystems of the Baltic Sea drainage basin (% from N deposition input)

EMEP co-ordinates		Computed N runoff	Experimental values		
X	Y	subbasin	values, %	runoff	reference
21	26	GUF	8.2	17.5	*Kallio and Kauppi*, 1990
21	27	GUF	7.3	8.8	*Kallio and Kauppi*, 1990
20	28	GUF	7.1	3.5	*Kallio and Kauppi*, 1990
18	28	BOB	5.9	5.0	*Kallio and Kauppi*, 1990
27	19	BAP	22–25	30.0	*Kinkor*, 1987
19	19	SOU	9.6	16.0	*Rasmussen*, 1987; *Tietema et al.*, 1995
17	26	BOB	9.0	7.1	*Lofgren and Kvarnas*, 1995
20	23	BOS	5.7	6.8	*Lofgren and Kvarnas*, 1995
20	21	BAP	5.3	7.0	*Lofgren and Kvarnas*, 1995
27	19	BAP	24.5	14.0	*Szarek*, 1995
19	19	SOU	9.6	6–174*	*Pedersen and Bille-Hansen*, 1995
23	27	GUF	3.9	5–90**	*Makarov and Kiseleva*, 1995
24	26	GUF	8.8	4–10	*Kusnecov and Balla*, 1994
22	19	BAP	17.5	25–75***	*Schaaf et al.*, 1995
20	20	SOU	14.9	12	*Nielsson and Wiklund*, 1995
24	26	GUF	8.8	15–32**	*Rovinsky et al.*, 1995
20	26	GUF	3.4	< 5	*Starr et al.*, 1995
20	28	GUF	7.2	< 5	*Starr et al.*, 1995
18	29	BOB	1.5	< 5	*Starr et al.*, 1995
21	19	BAP	10.8	13	*Behrendt*, 1995

* — *** See explanations in the text.

Under the study in the representative catchment in Crusne Mountains (Czech Republic), which was originally wholly forested and after tree-died and clear-cut was reforested by birch and mountain-ash, the mean annual fluxes of nitrogen were: deposition input — 16.2 kg ha^{-1} yr^{-1}, output with runoff — 5.5 kg ha^{-1} yr^{-1} that amounts to about 30% of leaching [*Kinkor*, 1987].

The study area "Forellenbach" is located at Bayerischer Wald National Park, a middle-mountain forest area in the eastern part of Bavaria close to the border to Czech Republic [*Beudert and Kennel*, 1995]. Mean annual nitrogen output in 1992 and 1993 was 6.4 kg/ha as nitrate nitrogen and less than 0.7 kg/ha as ammonia nitrogen. Compared to deposition input of at least 14 kg/ha (assumed additional dry deposition uptake in the canopy not considered)

an amount of at least 7 kg/ha is retained in the system and N retention is 52%. The simulation of nitrogen budget is more complex, but preliminary results indicate that tree growth (7 to 9 kg ha^{-1} yr^{-1}) possibly is not the only sink responsible for reducing of nitrogen flux from about 21 kg ha^{-1} yr^{-1} (55% nitrate and 45% ammonia nitrogen) in deposition to 6 to 8 kg ha^{-1} yr^{-1} in catchment runoff of "Grobe Ohe". The suggestion that nitrate reduction to gaseous nitrogen or N_2O could be responsible for the remained sink term should be further investigated, long-term accumulation in humus matter due to lowering C/N-ratios could be another possible sink of nitrogen.

Ion mass budget calculations were performed for three small, forested catchments in Sweden. The study sites are situated along a deposition gradient from high loads of sulphur and nitrogen in south-west Sweden to low deposition in the northern part of the country [*Lofgren and Kvarnas,* 1995]. The comparison of the given experimental calculations of N output with relevant computed data demonstrate suitable similarity (Table 14).

The inorganic nitrogen removed by stream transport is 6 or 7 percent of the wet deposition at all three catchments, irrespective of different levels of deposition in Sweden [*Bringmark,* 1995]. When organic nitrogen is included, the stream transport removes 18, 25 and 62% of the input at the different sites. Thus, sites with low deposition in the north have an output almost in balance with the input, while an effective accumulation is mainly observed at the southern sites. Comparisons with sites in Central Europe show that the capacity for accumulation will probably be exhausted when the amount of nitrogen deposited from the atmosphere is 800–1200 mg/m^2 [*Gundersen & Bashkin,* 1994].

Ion budget studies were also carried out in the middle of the 1980th at three sites in Denmark, all with 40–60 yr old Norway spruce plantations, but with different soil chemistry [*Rasmussen,* 1987; *Tietema et al.*, 1995]. The input/output budgets and the internal cycling of various cations and anions, including nitrate and ammonium ions, were determined by measurements of fluxes in bulk precipitation, throughfall and soil solutions in the A- and B-horizons. Two of these catchments belong to SOU subbasin of the Baltic Sea (Tange and Strodam) and one to the North Sea (Klosterhede). The values of N leaching were 16% for Klosterhede catchment with practical depressed nitrification processes (where changes were relatively low and slow due to the fact that this site was not saturated yet), about 70% for Tange one where the nitrogen input was connected with sea salt enrichment and the output of nitrogen exceeded sharply the input in Strodam where rich organic soil favoured to nitrification. Danish results give about 16% leaching values of atmospheric deposition nitrogen runoff from forest ecosystems that is close to the results computed for the same SOU subbasin area by our different methods of calculation (10–14%).

Hydrochemical observations according to which the balance of pollutants could be tested has been carried out over a small forested catchment area near Valday lake since 1974 [*Rovinsky et al.,* 1995].Annual concentrations of nitrogen oxides in precipitation are increased constantly during last decade and those of sulphur oxides are definitely decreased. Due to paucity of data for 1985-1990 one cannot follow changes from year to year. Therefore averaged five-year values were compared to determine deviations of mass balance components for the catchment as a whole and the comparison of the data was made for two periods: 1981–1985 and 1990–1994. The annual wet deposition input of nitrogen compounds onto the catchment amounts to 20–60 mg-eq/m^2 of ammonium nitrogen (NH_4^+-N) and 14–25 mg-eq/m^2 of nitrate nitrogen (NO_3^--N). Total output with running water is 5–20 mg-eq/m^2 of NH_4^+-N and 5–7 mg-eq/m^2 of NO_3^--N. The retention of deposited species in the catchment comprises 37–79% of nitrates and 61–85% of ammonium. The calculated data of deposition nitrogen runoff for EMEP cell 24/26 are 8.8%.

Most catchments lose nitrate because the nitrification (internal production of nitrate) exceeds the assimilation. High nitrogen deposition combined with catchment characteristics such as deep soils and gentle slopes (longer residence times) may increase the pools of soil N and enhance nitrification. On the contrary shallow soils and steep slopes (short residence times), which are characteristic of Norwegian catchments, may allow input N to pass quickly through a catchment into runoff. Acid deposition could also increase nitrate in runoff directly by elevating N input, or indirectly by impeding root uptake of N due to the toxic effects of soil acidity (low pH) and reactive aluminium [*Skjelkvale*, 1995]. Retention of N in a catchment is a measure of a quantity of incoming nitrogen retained in the catchment. The retention of nitrogen has been calculated in 4 calibrated catchments and 13 rivers situated in a large range of N-deposition and in 4 small lakes situated in south-western Norway, in the area with the highest N-deposition in Norway. In general the retention for most Norwegian catchments are very high (> 85%). Exceptions are three rivers in western Norway, and also four lakes in south-western Norway.

It was shown that the concentration and deposition of inorganic nitrogen in bulk precipitation at Finnish integrated monitoring (IM) catchments were very low compared to those experienced in Northern and Central Europe [*Starr et al.*, 1995]. The catchments also exhibit nitrogen retention rather than release. Estimates of mean annual input-output budget for the Valkea-Kotinen and Hietajarvi IM areas indicated that > 95% of the input of N was retained within the catchment [*Forsius et al.*, 1995].

Resuming this comparison of experimental and computed results one can see that the nitrogen cycle is complex as well as there are many uncertainties under survey of various features of ecosystems [*Posch et al.*, 1993; *Forsius & Kleemola*, 1995 etc.]. From this it appeared that most ecosystems are accumulating nitrogen and that there is different leakage. Increased biomass production has been observed throughout Europe, which may be related to the deposition of N. The question of whether there is a nitrogen problem was therefore posed. It was concluded that although there is a tremendous accumulation of N going on at present, dramatic effects (leaching) may occur when the systems become N saturated. It was pointed out that NO_3 runoff from lakes and N in throughfall have been shown to be positively related to N deposition. This led to a discussion about the need to group throughfall data by stand composition and characteristics in order to clarify the effects of N deposition [*Forsius & Kleemola*, 1995]. For those countries establishing new IM sites, it was recommended that the catchment should be carefully surveyed before deciding where to locate permanent plots. It was noted that calculating input-output balances are easier at the catchment-scale than at the plot-scale.

A similar comparison of catchment experimental and computed results of N runoff has been carried out for other sites (Table 14). With some exceptions logically explained (* — the highest value of 174% and 75% connected to extremely high nitrification activity; ** — high output connected with high input due to N emission during fertilizer plant operation; *** — calculations only for nitrate-N whereas nitrate and ammonium N were practically equal in depositions), the experimental and computed results of airborne nitrogen leaching are of reasonable agreement testifying to the applicability of the suggested models for assessing N runoff.

7. UNCERTAINTY ESTIMATES

7.1. Nitrogen Runoff from Terrestrial Ecosystems

Uncertainties connected with the assessment of nitrogen runoff from terrestrial ecosystems are subdivided into four main sets: (1) uncertainty associated with field measurements of nitrogen runoff in various subbasins of the Baltic Sea drainage area; (2) uncertainty associated with

different assumptions and simplifications in the methods applied; (3) uncertainty related to the interpolation procedures and (4) uncertainty caused by rescaling from small cells (2' × 2' LoLa grid) to big cells (150 × 150 km^2 EMEP grid). It was shown that the main source of uncertainty depended upon subbasin. On the basis of initial information sets and experimental field measurements three types of subregions were distinguished: a) the minimum uncertainty — the maximum number of field experimental comparison plots- Finnish part of GUF and BOS, Swedish part of BAP and BOS; b) the minimum uncertainty — the medium number of field experimental comparison plots — German and Danish parts of BAP and SOU; c) the medium uncertainty — the medium number of field experimental comparison plots — subregions of GUR subbasin, Polish, Russian and Belarus part of BAP; d) the maximum uncertainty — the minimum number of field experimental comparison plots — especially BOB and GUF subbasin.

7.2. Nitrogen Retention in Aquatic Ecosystems

Uncertainties connected with nitrogen retention in aquatic ecosystems are connected with the same reasons mentioned above and originated mainly from (1) uncertainty associated with various assumptions and simplifications and (2) uncertainty caused by spatial resolution and rescaling from small to big cells. Uncertainties of retention values in aquatic ecosystems depend on characteristics of the region considered.

7.3. Nitrogen Fertilizer Application

The uncertainty in regional scale of nitrogen fertilizer application that was used in method 3 is associated mainly with different resolution level of datapresented by HELCOM participating countries. The minimum uncertainty is related to Polish and Swedish data presented on the basis of district resolution for every year from 1987 to 1991, as well as to German and Danish ones estimated by national researchers [*Kronvang et al.*, 1995], the medium uncertainty is associated with data from Estonia (averaged data for the whole country for every year) and Russia (medium level of confidence for statistical estimates in general for a given period); the maximum uncertainty is related to other countries which have not submitted data sets on fertilizer application therefore these values have been extracted from FAO yearbooks of the corresponding periods.

7.4. Relation of Atmospheric Deposition to Riverine Input of Nitrogen

The uncertainty in regional scale for the values of relation of atmospheric nitrogen deposition to its riverine runoff strongly depends on the pollution pattern and on the land use in the area under study. This uncertainty is also related to the data of PLC-2 calculation of riverine nitrogen input to the Baltic Sea that have been indicated by authors [*Laane & Wilhelms*, 1993] as well as to the data of nitrogen atmospheric deposition presented in Part I of this report. For example, the values of riverine nitrogen assessed by PLC-2 did not include organic nitrogen for some countries such as Russia, Estonia, Latvia, Lithuania, Belarus and the Ukraine as well as on a basis of the PLC-2 results it is unclear whether (NO_3 + NO_2) − N was included in total nitrogen sum in Poland. Furthermore, German values of riverine input are too small in comparison with neighbouring Danish values [see table 4.38, p. 104, PLC-2 Report, *Laane & Wilhelms*, 1993].

Therefore, using all these estimates, the uncertainty of computed values characterizing the evaluation of the relation of atmospheric deposition to riverine runoff of nitrogen in various subregions of the Baltic Sea drainage basin can be as large as 10−50%, being on average 25% which is in agreement with Table 14.

8. THE ASSESSMENT OF THE RELATION OF ATMOSPHERIC DEPOSITION TO RIVERINE INPUT OF NITROGEN TO THE BALTIC SEA

The final aim of a given project is related to comparative assessment of values characterizing the nitrogen input to the Baltic Sea from its drainage basin and those showing other sources of nitrogen input via river runoff, direct discharge from urban and industrial territories. Correspondingly, Table 15 shows that the part of atmospheric nitrogen deposition input into the Baltic Sea in 1990 year was 118 kt/yr or 21% of total nitrogen riverine input value, which is equal to 568 kt/yr and 18% of total input of nitrogen with rivers, urban and industry (662 kt/yr) in accordance with PLC-2 Report No. 45 [*Laane and Wilhelms*, 1993]. However, these values vary significantly. Thus, in subbasins of the Bothnian Bay, the Bothnian Sea and the Gulf of Finland where the input of atmospheric deposition nitrogen does not exceed 10 kg ha^{-1} yr^{-1}, they were equal to 7–10% of total N riverine input. Exceptions were connected with results obtained for St. Petersburg region where nitrogen deposition rates were up to 15–20 kg ha^{-1} yr^{-1} but given figures were nivelized in total runoff sum from huge area of Gulf of Finland.

Under increasing rates of atmospheric pollution by nitrogen compounds in subbasin GUR and especially in Polish and German parts of subregion BAP, the contribution of atmospheric deposition nitrogen to its total riverine input enhances significantly up to 45% in BAP subbasin. In the western area of drainage basin (subbasin SOU) the nitrogen atmospheric contribution to the total sum of this element entering the Baltic Sea basin via rivers was equal to 10% (Table 15).

In the same time, the input of airborne nitrogen from the watershed area of the Baltic Sea, on the average being equal to 119 kt per year for 1987–1991 period was 39% of nitrogen pool that directly comes to the Baltic Sea basin with atmospheric deposition (298.8 kt per year) and 12% of total input of nitrogen to the Baltic Sea (directly to the sea, via riverine input and from coastal industrial and municipal inputs equal to 991 kt in 1990). However, Swedish compilation showed that the same values of total nitrogen input to the Baltic Sea (without N fixation) for 1992–1993 was equal to 1275 kt per year [*Enell and Fejes*, 1995]. In this case the part of our calculated values for nitrogen atmospheric deposition would be decreased to about 9%.

One can see that the comparison with PLC-2 values for riverine runoff [*Laane and Wilhelms*, 1993] shows confusing data for BAP subbasin, especially its Polish part where airborne nitrogen is equal totally to 45% of riverine input. However, as it was mentioned above there are some uncertainties in PLC-2 data and given results differed from the similar compilations [see *Enell and Fejes*, 1995]. As it was mentioned above, deposition data for BAP can be overestimated as well. Furthermore, the application of mineral fertilizers decreased sharply in Poland during 1990–1991 that accompanied by decreasing relative part of fertilizer nitrogen and increasing of deposition nitrogen part.

Furthermore, it is necessary to take into account possible deviations of our computed data which can achieve 25% for the whole watershed and up till now that is a matter of uncertainties and discrepancies in data as it was mentioned above.

Consequently, the value of atmosphere deposition riverine input of nitrogen must be undoubtedly taken into account considering the Baltic Sea pollution sources.

Table 15

Comparative assessment of total nitrogen input into the Baltic Sea in 1990 from various sources, kt/yr, method 1 data

Subregions	Sources of input*			Total	Part of atmospheric deposition		
	river	urban	industry		kt	1**	2***
BOB	35.0	1.6	1.6	38.2	2.38	6.2	6.8
BOS	50.7	2.5	4.2	57.4	4.33	7.5	8.5
GUF	109.6	30.0	0.9	140.4	10.68	7.6	9.7
GUR	80.0	5.0	0.3	85.3	8.24	9.7	10.3
BAP	182.1	24.7	2.4	209.3	82.29	39.2	45.2
SOU	110.2	18.2	2.7	131.2	10.69	8.1	9.7
Whole Baltic	567.7	82.0	12.1	661.9	118.61	17.9	20.9

Footnotes: * PLC-2 results [*Laane and Wilhelms*, 1993].
1** — per cent of total input.
2*** — per cent of riverine input.

The distribution of nitrogen load on the Baltic Sea in 1990 with different pathways and with different emission sources is shown in the figure 7a and 7b correspondingly.

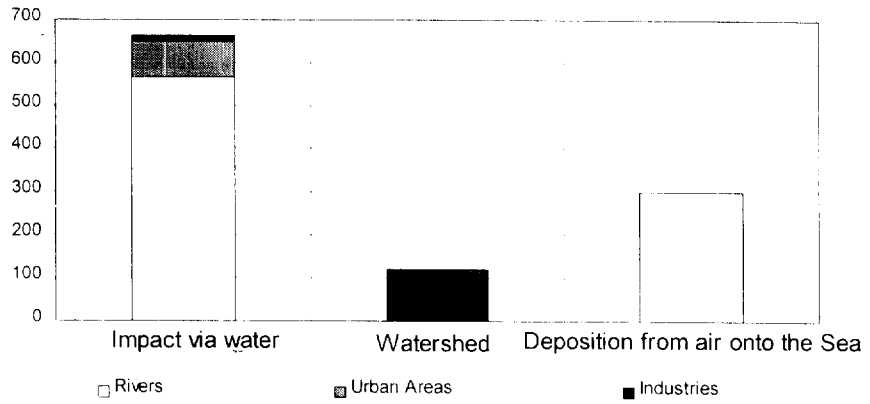

Fig. 7a. Input of nitrogen to the Baltic Sea in 1990 by different pathways (Unit: kt N/yr)

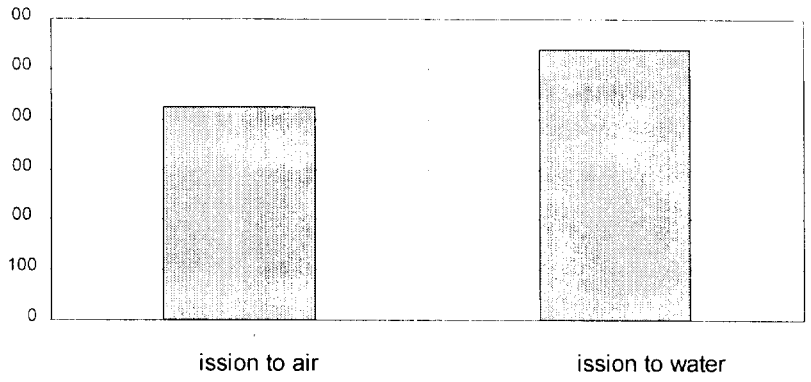

Fig. 7b. Input of nitrogen to the Baltic Sea in 1990 by different emission sources (Unit: kt N/yr)

9. CONCLUSIONS

1. Evaluation of the relation of atmospheric deposition to riverine input of nitrogen to the Baltic sea from its drainage basin has been carried out for the period of 1987–1991. The assessment has been done on the basis of non-linear leaching models using nitrogen runoff and retention coefficients for various terrestrial and freshwater ecosystems. Three various methods were used.

2. The results of three methods used were relatively similar and available experimental data are not contradictory with these results.

3. It was calculated that in various drainage subbasins of the Baltic Sea leaching of atmospheric deposition nitrogen varied within the limits of 5–10% of deposition on these subbasins being maximum in the southern and western parts and minimum one — in the northern part of the basin. The mean values during 1987–1991 period were equal to 120 kt/yr or 8% of the nitrogen deposition on the whole drainage area.

4. Among various sources of nitrogen input to the Baltic Sea, estimated by PLC-2, the contribution of airborne nitrogen deposited on the drainage basin area and entering via rivers the sea water is 21% (7–45% depending on subbasin) of total riverine input. The part of atmospheric deposition riverine nitrogen is equal to 12% of the total nitrogen input (riverine, industrial, municipal, deposited on the aquatorium) to the Baltic Sea in 1990.

5. To diminish the degree of existing uncertainties it is reasonable to make similar calculations for a longer period of time with higher resolution of depositions.

Acknowledgement

Authors wish to thank HELCOM for financial support of these researches, A. Duchnowska (Poland), A. Laane (Estonia), H. Herata (Germany), M. Henriksson (Sweden) for presenting requested national data on fertilizer application and S. Langaas (Sweden) for presenting GIS-data sets, T Rusanova (Institute of Soil Science and Photosynthesis RAS, Russia) for technical assistance.

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ANNEX A.
DESCRIPTION OF THE MSC-E ACID MODELS APPLIED TO THE BALTIC WATERSHED DEPOSITION CALCULATIONS

The MSC-E modelling technology includes several models based on the same key units: advection scheme, vertical profile description, meteorology and emission datapre-processing, unit for emitter-receptor calculations and control unit. Physical and chemical features of the pollutants are given in specific units, separately for each substance group.

The acid model includes a scheme of chemical transformations for more than 15 reactions (including some of the second order) and descriptions of dry and wet depositions of all considered pollutants.

All model units are subjects of changes and improvements. For this study operational acid model of 1992 and updated model of 1996 were used (hereinafter referred as M-92 applied to RUN92) and M-96. The differences between them are discussed further in relevant parts of the model description.

The description of main units of the M-92 can be found in [*Pressman et al.*, 1991; *Afinogenova et al.*, 1992; *Annual report*, 1993]. Later modifications are mentioned in [*Galperin et al.*, 1994; *Sofiev et al.*, 1994; *Galperin*[1] *et al.*, 1995; *Galperin*[2] *et al.*, 1995; *Sofiev et al.*, 1995].

THE MODEL ADVECTION SCHEME

On the whole the presented scheme is similar to numerical advection schemes [*Egan and Mahoney*, 1972; *Pepper and Long*, 1978] and to those of "particle-in-cell" [*Pedersen and Prahm*, 1974].

Let F(t) be a radius-vector of the mass in a cell (i, j). If there are no mass exchange between this cell and others then trajectory steps are realized (Fig. A1):

$$\vec{r}(t + \Delta t) = \vec{r}(t) + \vec{V}(t)\Delta t,$$

where Δt is the model time step, $\vec{V}$ is the wind.

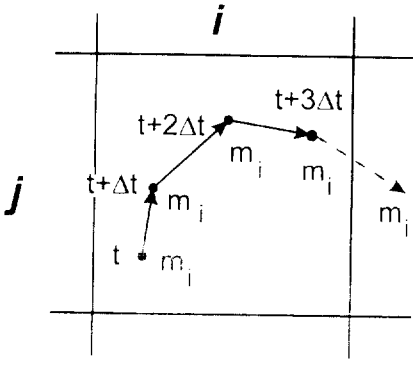

Fig. A1. Trajectory steps

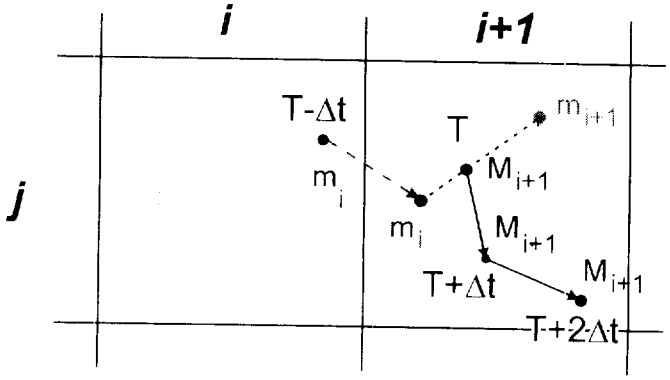

Fig. A2. Eulerian step

In case of some mass input to a cell at moment T (either because of advection, or because of emission) the Eulerian step is applied with the conservation of mass in a cell and its first moment (Fig. A2):

$$M_{i+1} = \sum m_i,$$

$$\vec{P}_{M_{i+1}} = \sum \vec{P}_{m_i},$$

where $\vec{P}_{m_i} = m\vec{r}_m$ is the first moment of the corresponding mass.

At each time step all grid cells are addressed sequentially and independently from each other. All masses come to a cell (as well as their first moments) are summed up and stored as initial conditions for the next time step.

The scheme errors are resulted from non-uniformity of mass distribution with grid cells (Gibbs effect). The suppression of the oscillations is realized by distribution of mass with Eulerian grid or by averaging over considerable time periods not less than one week.

VERTICAL DISTRIBUTION

Model M-92

The model uses an expression of equivalent mixing layer [*Pressman et al.*, 1991] H = M/C(0) where M is the pollution mass in a grid cell, C(0) is the surface concentration used for dry deposition calculations:

$$\Delta M = - \frac{MV_d\tau}{H},$$

where τ is the model time step and V_d is the dry deposition velocity.

Temporal variations of the mixing layer height are derived from the diffusion equation [*Pressman et al.*, 1989].

Model M-96

In this model the truncated gaussian profile of pollution distribution along the vertical C(Z) is used:

$$C(Z) = M\varphi(Z) / P(\mu / \sigma),$$

where

$$\varphi(Z) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left(\frac{-(Z-\mu)^2}{2\sigma^2} \right), \quad P(\mu / \sigma) = \int_0^{\infty} \varphi(Z)\,dZ = \int_{-\infty}^{\mu/\sigma} \varphi_0(\lambda)\,d\lambda$$

and $\varphi_0 = \dfrac{\exp(-\lambda^2/2)}{\sqrt{2\pi}}$.

Here $\varphi(Z)$ describes the shape of the vertical concentration profile, $P(\mu/\sigma)$ is the normalizing divisor, μ — co-ordinate of maximum concentration and σ — dispersion measure along the vertical.

The relationship of concentration distribution with height C(Z), mass center Z_0 and total mass in a cell M is represented by formulas:

$$Z_0 = \frac{1}{M}\int_0^\infty ZC(Z)\,dZ, \qquad M = \int_0^\infty C(Z)\,dZ.$$

The boundary condition near the surface is:

$$V_d\,C(0) = -K_z \left.\frac{\partial C(Z)}{\partial Z}\right|_{Z\to 0},$$

where V_d is the diffusional surface dry deposition velocity and K_z is the turbulence coefficient.

Under these conditions at known M(t) and $Z_0(t)$ distribution parameters of C(Z) may be calculated. Then physical-chemical processes in a cell at time step Δt are calculated alongside dry, gravitational and wet deposition, particle size spectrum variations, chemical transformations, etc. After these calculations new value $M(t+\Delta t)$ is known and a new value of $Z_0(t+\Delta t)$ is determined, which is the initial state for the next time step.

Vertical diffusion leads to the increase of σ:

$$d\sigma(t)/dt = K_z(\sigma, t)/\sigma.$$

The position of the mass centre is calculated from the following equation:

$$Z_0(t+\Delta t) - Z_0(t) = \frac{d\sigma}{dt}\left(\frac{\varphi_0(y)}{P(y)} + y\right)\Delta t,$$

where $y = \mu/\sigma$.

The interaction with advection is based on the moment conservation along the vertical during the mass exchange between horizontal cells:

$$M = \Sigma M_i, \qquad Z_0 M = \Sigma Z_{0i} M_i,$$

where M — total mass in a cell, M_i — masses appeared to be in a cell after time-step realization, Z_0, Z_{0i} — vertical co-ordinates of centres of gravity.

When a source is introduced mass $M = E(t)\Delta t$ is added. The vertical co-ordinate Z_0 for this mass is taken to be equal to the effective source height.

If layer-by-layer wind is available, distribution C(Z) is used for the evaluation of weighted advection velocity.

DRY AND WET DEPOSITION

Dry deposition is calculated in a common way:

$$Q(t + Dt) - Q(t) = V_d C(0) Dt.$$

It is assumed that V_d depends on the surface type and state, and V_d is taken at the height of roughness layer (i.e. for a level of a millimetre to several tens of centimeters [*Wieringa,* 1981]).

For the estimations of V_d of gases and aerosols with particle sizes less than 0.1 m we used data available in the literature [*Sehmel,* 1980] and direct measurements carried out in Poland as a contribution to EMEP [*Nowicki,* 1987]. The data provide rather comprehensive picture of the underlying surface impact on dry deposition velocity. In particular these data indicate that the extent of surface moistening inflicts drastic impact on V_d variation for SO_2. It was shown that mean V_d is abruptly changed at the temperature about 0°C, i. e., when water freezes. Therefore the following formula is used for V_d calculation:

$$V_d = V_{d0}\, K(T, P, F),$$

where V_{d0} — dry deposition velocity for dry surface; K(T, P, F) — factor depending on temperature T, precipitation amount P, and underlying surface type F (F = 2 for water, F = 1 for grid squares with mixed surface type, F = 0 for land). Values of V_{d0} and function types K(T, P, F) are given below:

Substance	V_{d0}, cm/s	K (T, P, F) type
SO_2, NH_3	0.3	1
Aerosols (SO_4^-, NH_4NO_3, etc.)	0.1	1
NO_2	0.1	2
PAN, R (VOC)	0.1	2
HNO_3	1.0	1

1. K = 1 at T < −2°C and any P and F
   K = 1 at T > −2°C P = 0 and F = 0
   K = 2 at T ≥ −2°C F = 1 or 1 mm/h > P > 0
   K = 3 at T ≥ −2°C F = 2 or P ≥ 1 mm/h

2. K = 1 at T < −2°C and any P and F
   K = 1 at T > −2°C F = 0 and P = 0
   K = 0.5 at T ≥ −2°C F = 1 or 1 mm/h > P > 0
   K = 0 at T ≥ −2°C F = 2 or P ≥ 1 mm/h

For smooth water surface and wet surface after rain and for quiet sea surface it is taken $V_d = 3V_{d0}$ [*Rodhe and Soderlund,* 1980].

Model M-96 considers the effect of V_d increase with wind intensification due to the effect of the surface roughness increase with wave creation and crashing.

In the first approximation this effect could be described by the Charnock formula

$$z_0 \in \frac{u_*^2}{g}$$

which leads to square dependence of the roughness on the wind speed.

This formula is suitable for moderate wind speeds (approximately up to 3 balls of the Bofort scale). In case of stronger winds the wave crashing process leads to further increase of the deposition velocity because the sedimentation to the sea surface is becoming a wet scavenging process.

There are few data suitable for the parametrization of the process. Some of them have been presented in the Ukrainian report "in-kind" [*Ukrainian contribution,* 1995].

In the model it is assumed that the dry deposition velocity is growing up in the square proportion to the wind velocity which results in some non-uniformity of dry deposition pattern. The same correction factor is used for all hydrophilic pollutants but the initial dry deposition velocity is different for different substances. The range of this factor is as wide as one decimal order.

As it was shown in [*Engelmann,* 1963; *Galperin,* 1989] the **washout coefficient** is:

$$\Lambda = \beta S_0 I / v_p,$$

where S_0 — surface area of precipitation element, v_p — its volume, I — precipitation intensity (for example, mm/h), β — trapping efficiency factor.

The value of β strongly depends on aprecipitation type (snow, rain) and on drop or flake sizes as well as on scavenged particle sizes.

In case of gases and particles smaller than 0.01 m (Brownian particles) aerodynamic bending around drops and flakes is not important. Drops and flakes are in permanent contact with renewed portions of gas, therefore characterizes the effective cross-section and equals to about 0.25 for rain and 1.0 — for snow. It is assumed that in clouds $\beta \cong 1$ always. For SO_2 and NO_x depends on saturation of precipitation elements [*Galperin,* 1989].

Actually the air concentration is afunction of z and time therefore more complete description of the washout process is a set of equations in terms of partial derivatives. During the washout process the vertical redistribution of pollution takes place due to three factors. Firstly, scavenging occurs "from top to bottom" since at the upper part of the subcloud layer precipitation elements are not saturated and scavenging is more intensive. Secondly, downward air flows entrain pollutants from upper to lower layers. Thirdly, since gas concentrations in falling drops are much higher than equilibrium ones (for example, for SO_2) the fraction of gases which was not oxidised while being deposited on the ground surface enters again the atmosphere. The occurrence of the latter two effects is confirmed by statistical data on the growth SO_2 and NO_2 surface concentration with the commencement of aprecipitation event [*Afinogenova et al.,* 1990].

The application of the full-scale model [*Galperin,* 1989] for practical calculations leads to unjustified sophistication of computations. In order to develop an approximate model we take advantage of an analytical solution obtained in [*Galperin,* 1989] and refining semiempirical formula:

$$\Lambda_s = \Lambda / (m/m_s + 1),$$

where m is the pollutant mass in air, $m_s \cong 0.1$ mmole $\cdot m^{-2}$ is the parameter of trapping capability of precipitation.

The dependence of relative surface of precipitation elements S_0/v_p on precipitation intensity and on its type is clearly expressed. If we consider data presented in [*Kelkar,* 1959] then for rain:

$$S_0/v_p \cong 6(I + I_0)/ID,$$

where D — maximum effective diameter of a drop ($D \cong 1.5 - 2$ mm) and $I_0 \cong 0.8$ mm/h. For snow the relative surface varies slighter depending on precipitation intensity. Most probably it is connected with the formation of the near-surface air layer around snow flakes. The problem of rain drops saturation for pollutants producing cations in solution (for example, NH_4^+) is remained open.

THE MODEL CHEMICAL SCHEME

The chemical scheme of M-96 is shown in Fig. A3. It differs from that of M-92 by the consideration of ozone creation potential as a result of VOC emission and by the consideration of strong acid formation in case of lack of ammonia. The scheme includes the following equations partly described in [*Pressman et al.,* 1991]:

$$d(NO) = - K_1[O_3]\,[NO] + K_2[NO_2] - K_3[NO]\,[R] + aq(NO_x);$$

$$d(NO_2) = - d(NO) - d(PAN) - K_4[NO_2] - K_7[O_3]\,[NO_2] + q(NO_x);$$

$$d(O_3) = - K_{13}[O_3] - K_1[O_3]\,[NO] - K_7[O_3]\,[NO_2] + K_2[NO_2] + g(O_3);$$

$$d(HNO_3) = K_4[NO_2] - K_9[HNO_3] + K_8[NO_3^-] - d(NH_4NO_3);$$

$$d(NO_3^-) = K_7[O_3]\,[NO_2] + K_9[HNO_3] - K_8[NO_3^-];$$

$$d(NH_3) = - 1.5d((NH_4)_{1.5}SO_4) - d(NH_4NO_3) + q(NH_3);$$

$$d(PAN) = K_5[R]\,[NO_2] - K_6[PAN];$$

$$d(SO_2) = - K_{10}[SO_2] + bq(SO_x);$$

$$d(SO_4^{2-}) = - d(SO_2) + q(SO_x);$$

$$d((NH_4)_{1.5}SO_4) = \text{Min}\,([NH_3]/1.5;\ [SO_4^{2-}] - [(NH_4)_{1.5}SO_4]);$$

$$d[NH_4NO_3] = ([NH_3] + [HNO_3])/2 - (([NH_3] - [HNO_3])^2 + 4K_{11})^{1/2}/2$$

$$\text{at } ([NH_3] + [NH_4NO_3])\,([HNO_3] + [NH_4NO_3]) > K_{11};$$

$$= - [NH_4NO_3] \text{ at } ([NH_3] + [NH_4NO_3])\,([HNO_3] + [NH_4NO_3]) \le K_{11};$$

$$d(R) = - K_3[NO]\,[R] - K_5[R]\,[NO_2] + K_6[PAN] - K_{12}[R] + K_{14}q(VOC).$$

Here: [C] is the concentration (mass in a calculated grid) of substance C; $K_1,\ldots,K_{14}$ are the reaction constants (see Table 1); d(C) is the concentration increment at a unit time step; q(C) is the emission of substances given in brackets ($NO_x = NO + NO_2$, $SO_x = SO_2 + SO_4^{2-}$ VOC

is the volatile organic compound), a and b — fractions of NO and SO_2 in the emission; $g(O_3)$ is the background rate of tropospheric ozone formation; $(NH_4)_{1.5}SO_4$ is the mixture in equal fractions of NH_4HSO_4 + $(NH_4)_2SO_4$; SO_4^{2-} is all substances containing ion SO_4^{2-} ; R is the free radicals (ozone creation potential).

Table 1

Coefficients and parameters of chemical transformation scheme

Coefficient or parameter	Unit	Value or formulas
K_1	$m^3/(\mu mole^{-1}\ h^{-1})$	15.24 + 0.068T
K_2	h^{-1}	12S
K_3	$m^3/(\mu mole^{-1}\ h^{-1})$	$1100S^2 + 0.25$
K_4	h^{-1}	$0.08S^2$
K_5	$m^3/(\mu mole^{-1}\ h^{-1})$	$12S^2$
K_6	h^{-1}	$28.6 \times 10^{17} \exp(-12530/T)$
K_7	$m^3/(\mu mole^{-1}\ h^{-1})$	$5.625 \times 10^{-4} \times (T - 228.7)$
K_8	h^{-1}	0.015
K_9	h^{-1}	0.06
K_{10}	h^{-1}	$138500 \exp(-4517/T)$
K_{11}	$\mu mole^2\ m^{-4}\ h^{-2}$	1.12×10^{34} [exp(−24220/T)] × × $(298/T)^{6.1}$ [× (100 − H)/38 if H > 62; × 1 if H < 62]
K_{12}	h^{-1}	0.03
K_{13}	h^{-1}	0.005 + 0.2S
K_{14}	relative units	
$g(O_3)$	$\mu mole^{-3}\ h^{-1}$	$0.044S^2 + 0.21S + 0.03$
a, b	relative units	0.95

Dependence K_{11} (T,H) is taken from [*Finlayson-Pitts and Pitts,* 1988], K_{14} are calculated using the data basis taken from [*Simpson,* 1992].

Notations: T — temperature in K°, S — relative insolation, H — relative humidity in %

Many reactions depend of solar radiation determined by the atmosphere state, latitude, local time, season and cloudiness. Latitude and time are determined by the solar zenith angle Z. It is usually assumed that reaction rates are varied proportionally to function [*Carter et al.,* 1976]:

$$S(Z) = \exp(-r/\cos Z),$$

where r — constant characteristic of a given reaction. For the majority of reactions r value is known to be approximately within the range of 0.5−2.5. In modelling relative (dimensionless) values of radiation are used. They are calculated by formula:

$$S = S_M(L) \cos(M\pi/6) + S_A(L) \cos(t\pi/12),$$

where $S_M(L)$ and $S_A(L)$ — functions of latitude L taken from tables, M — number of a month of the year, t — local time in hours. The radiation level at latitude 35° at equinox, at noon

is taken to be S = 1 (it means, that $0 \leq S \leq 1.2$). It is supposed that radiation is decreased proportionally to cloud cover density. The final parametrization of the chemical scheme is specified on the basis of statistical analysis provided by the EMEP network data given in EMEP/CCC reports 1980–1994.

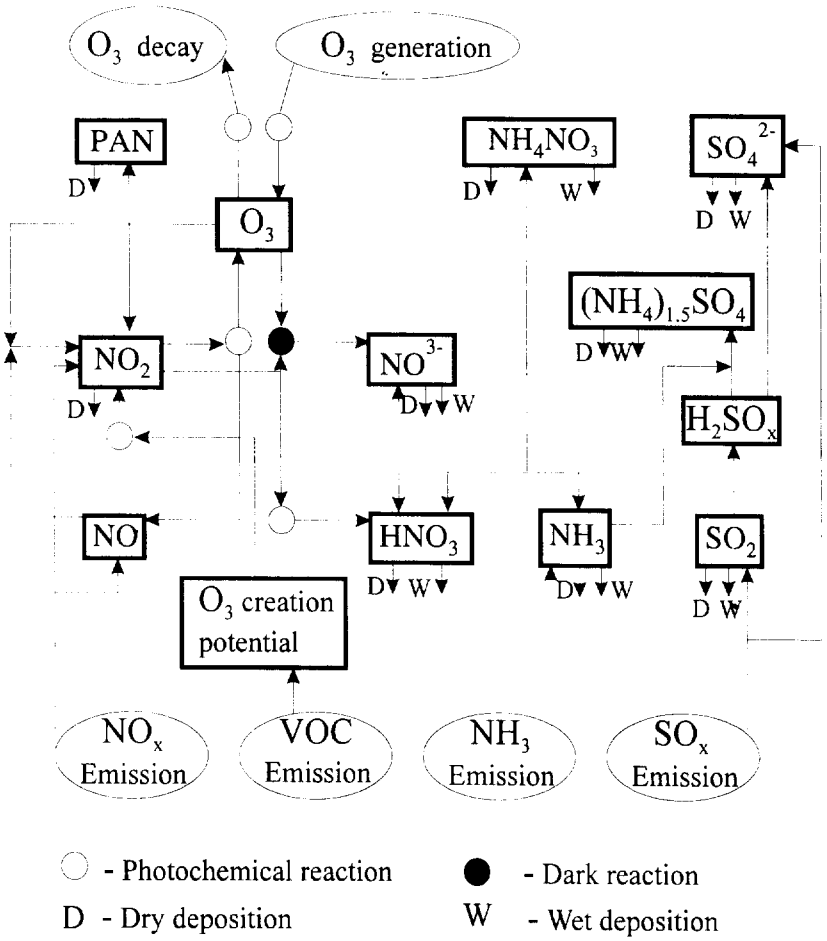

Fig. A3. Model chemical scheme

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ANNEX B.
MAPS OF THE EMISSIONS USED AND CALCULATION RESULTS

Contents

Figure B1. NO_2 (low) emission averaged for 1990, used for RUN96, unit = 100 tonnes

Figure B2. NO_2 (high) emission averaged for 1990, used for RUN96, unit = 100 tonnes

Figure B3. NH_3 emission averaged for 1990, used for RUN96, unit = 100 tonnes

Figure B4. Deposition of total nitrogen on the Baltic Sea region averaged for 1987–1991, RUN92, unit = mg N m^{-2} yr^{-1}

Figure B5. Deposition of total nitrogen on the Baltic Sea region averaged for 1987–1991, RUN96, unit = mg N m^{-2} yr^{-1}

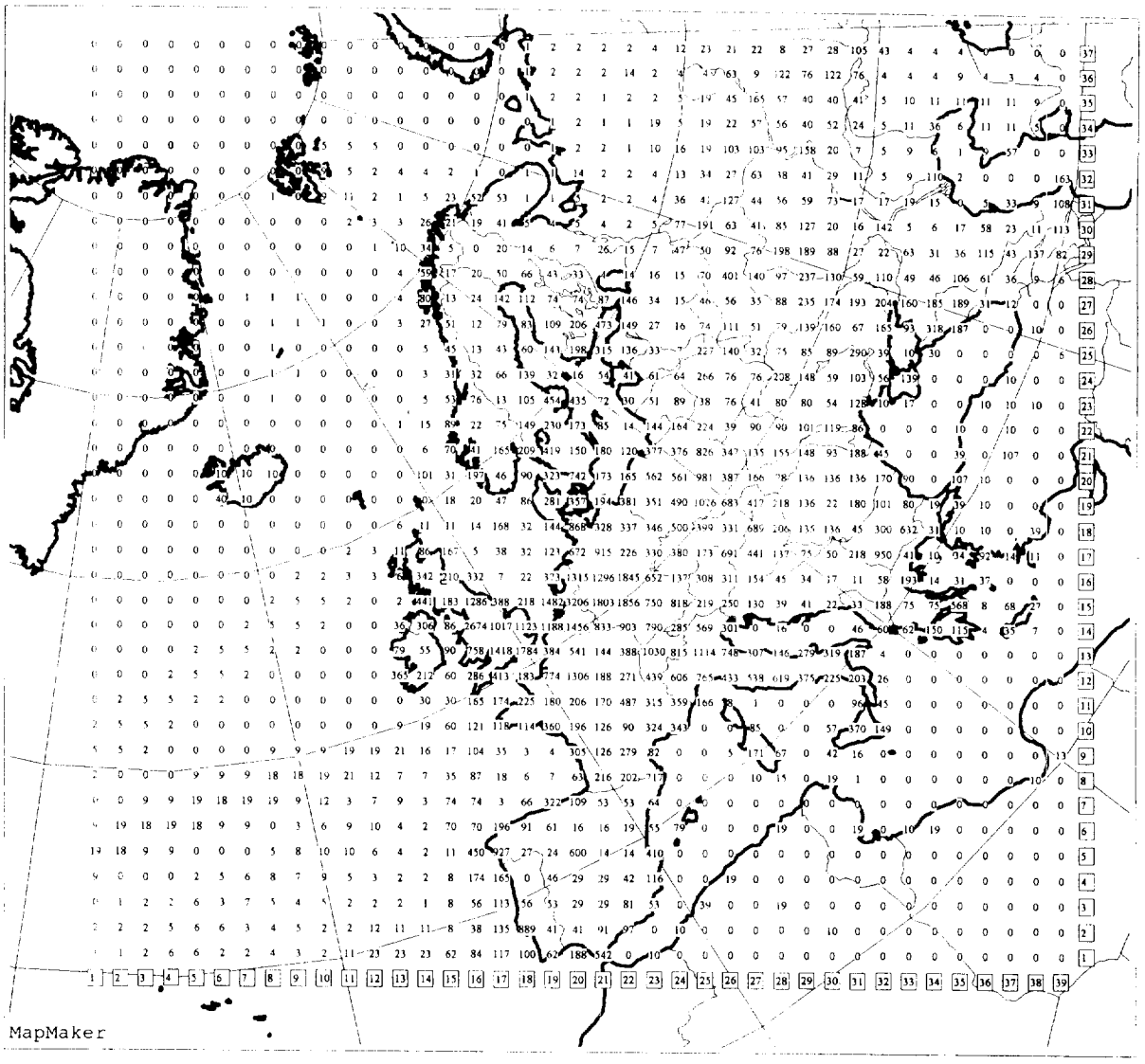


Figure B1. NO_2 (low) emission averaged for 1990, used for RUN96, unit = 100 tonnes

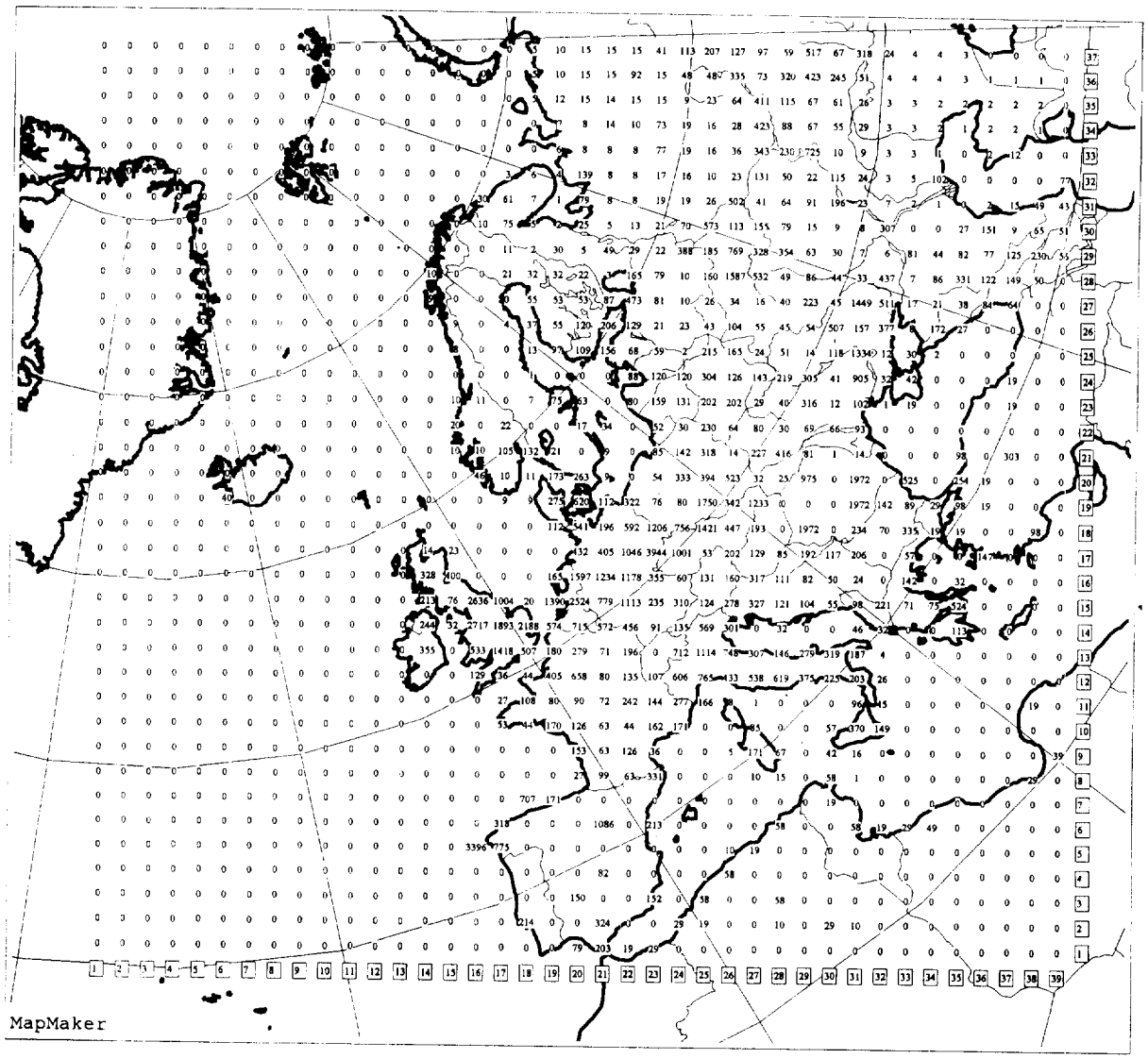


Figure B2. NO_2 (high) emission averaged for 1990, used for RUN96, unit = 100 tonnes

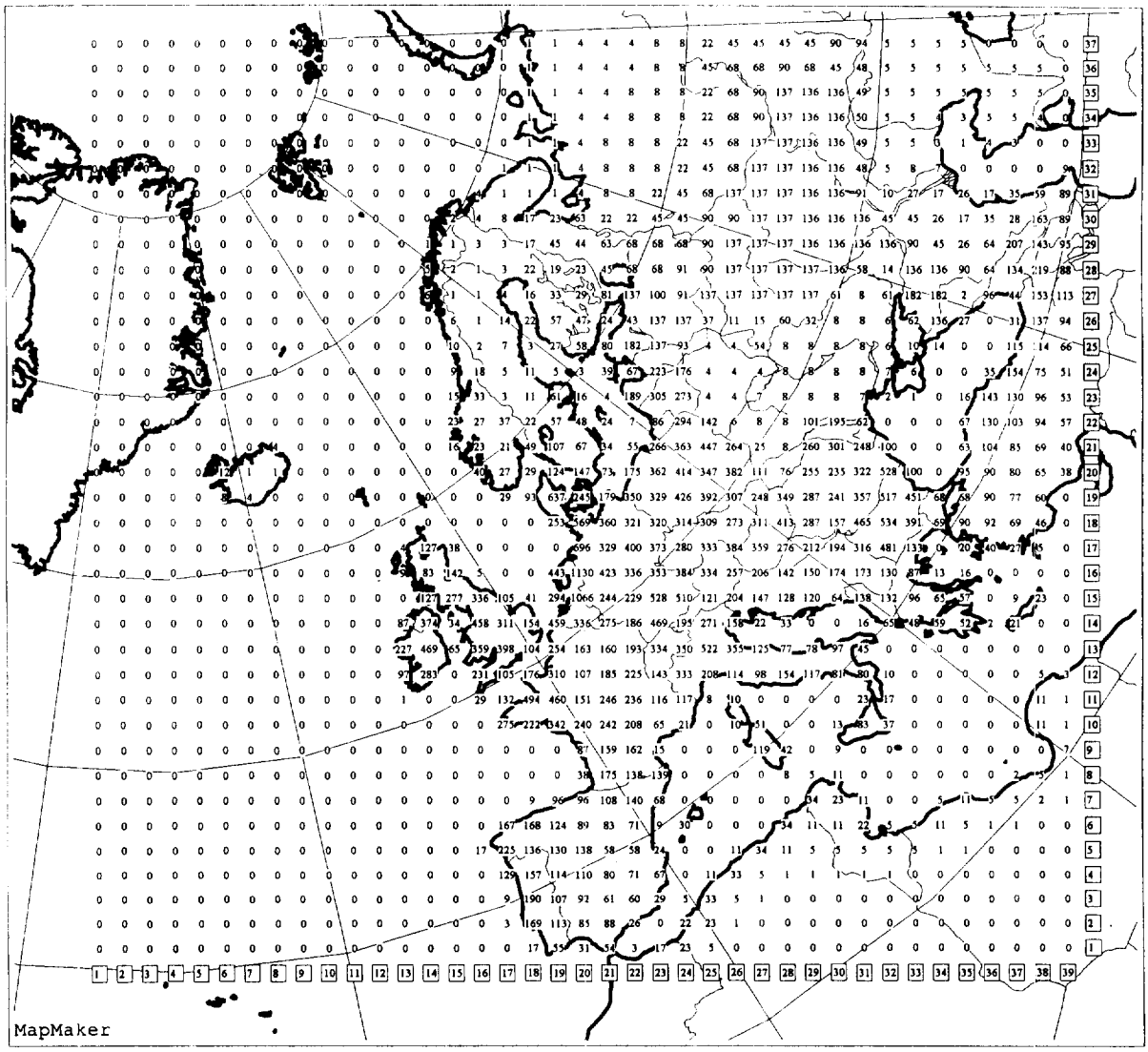


Figure B3. NH_3 emission averaged for 1987–1990, used for RUN96, unit = 100 tonnes

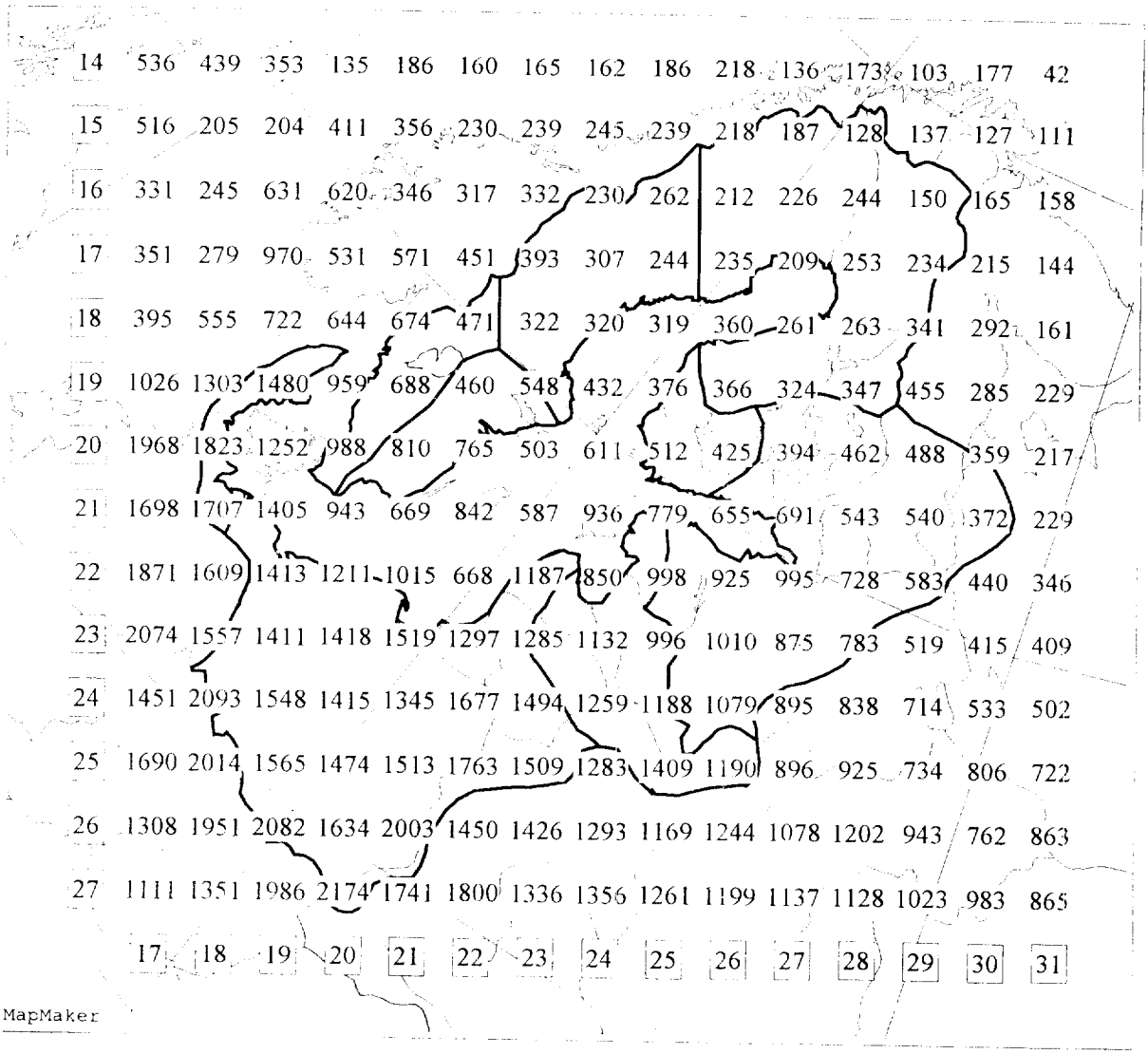


Figure B4. Deposition of total nitrogen on the Baltic Sea region averaged for 1987–1991, RUN92, unit = mg N m^{-2} yr^{-1}

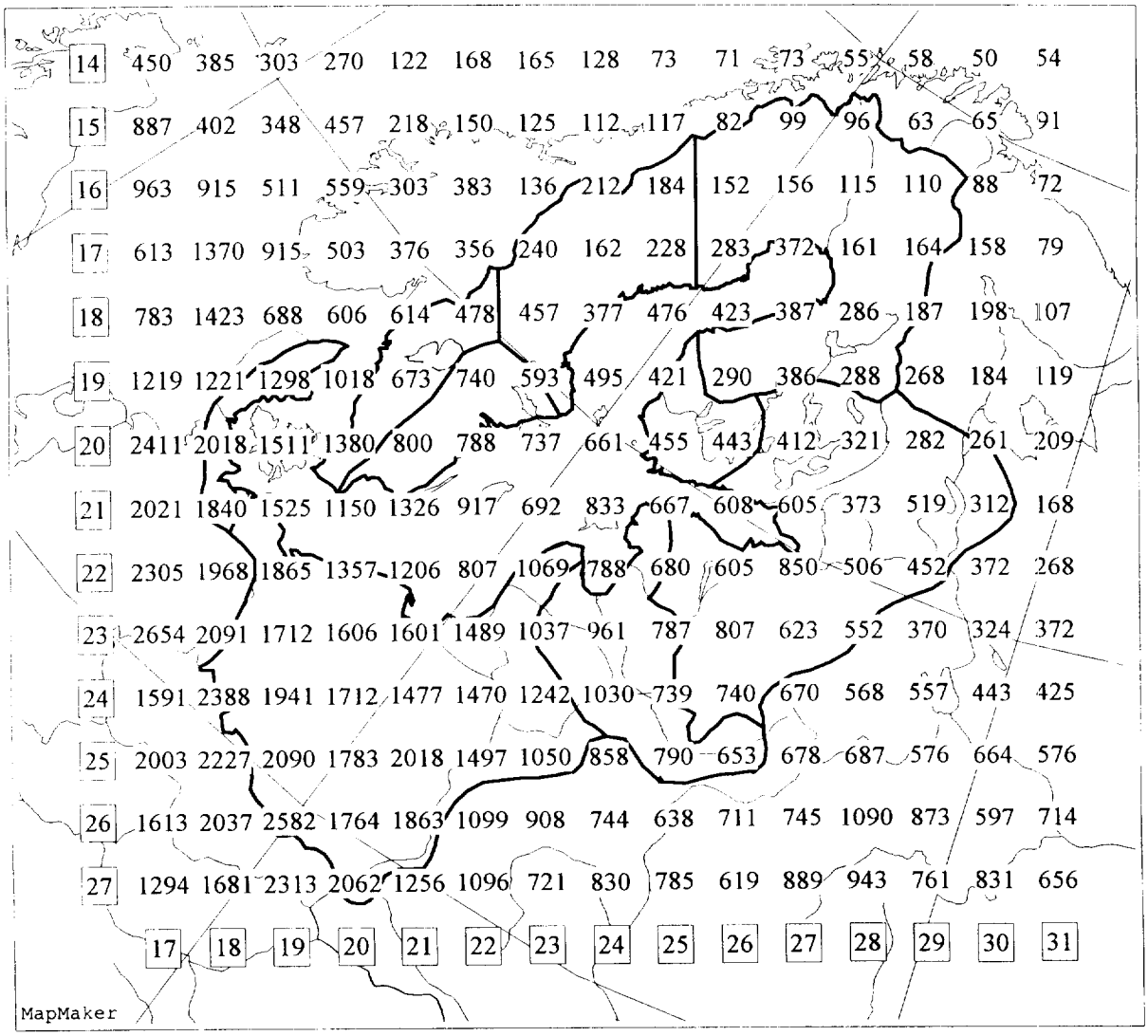


Figure B5. Deposition of total nitrogen on the Baltic Sea region averaged for 1987--1991, RUN96, unit = mg N m^{-2} yr^{-1}

ANNEX C.
RESULTS OF MODEL-MEASUREMENT AND MODEL-MODEL COMPARISON

Contents

Figure C1. Comparison of NO_x and NH_x mean wet deposition in 1987–1990 measured at EMEP stations and calculated in RUN92, unit = mg N/m² per annum.

Figure C2. Comparison of NO_x and NH_x mean wet deposition in 1987–1990 measured at EMEP stations and calculated in RUN96, unit = mg N/m² per annum.

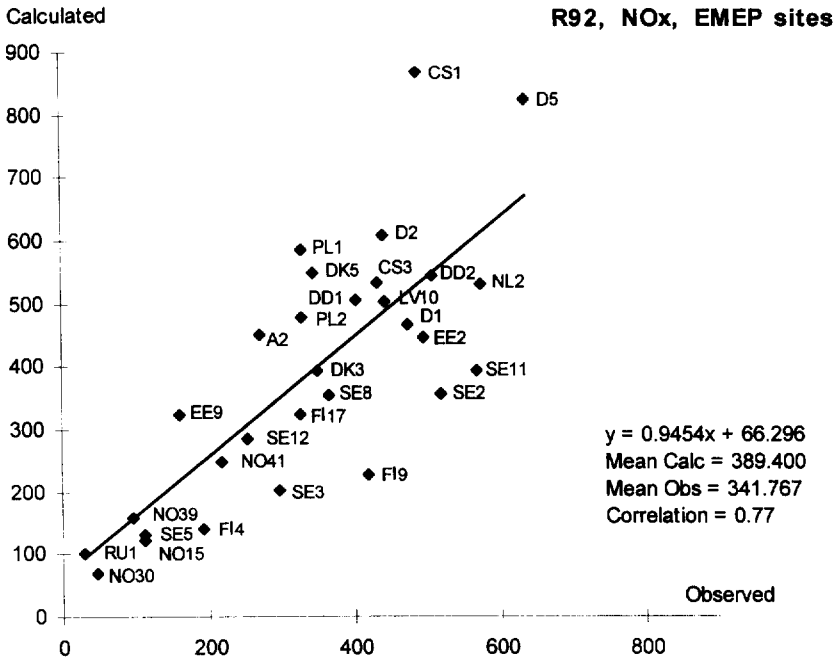
Figure C3. Comparison of N_{tot} mean wet deposition in 1987–1990 measured at EMEP and EGAP stations and calculated in RUN92, unit = mg N/m² per annum

Figure C4. Comparison of N_{tot} mean wet deposition in 1987–1990 measured at EMEP and EGAP stations and calculated in RUN96, unit = mg N/m² per annum

Figure C5. Comparison of RUN92 and RUN96 calculated results referred to grid squares with EMEP/EGAP stations (N_{tot} (NO_x + NH_x) mean annual wet deposition in 1987–1990), unit = mg N/m² per annum

Figure C6. Comparison of RUN92 and W95 calculated results referred to grid squares with EMEP/EGAP stations (N_{tot} (NO_x + NH_x) mean annual wet deposition in 1987–1990), unit = mg N/m² per annum

Figure C7. Comparison of RUN96 and W95 calculated results referred to grid squares with EMEP/EGAP stations (N_{tot} (NO_x + NH_x) mean annual wet deposition in 1987–1990), unit = mg N/m² per annum



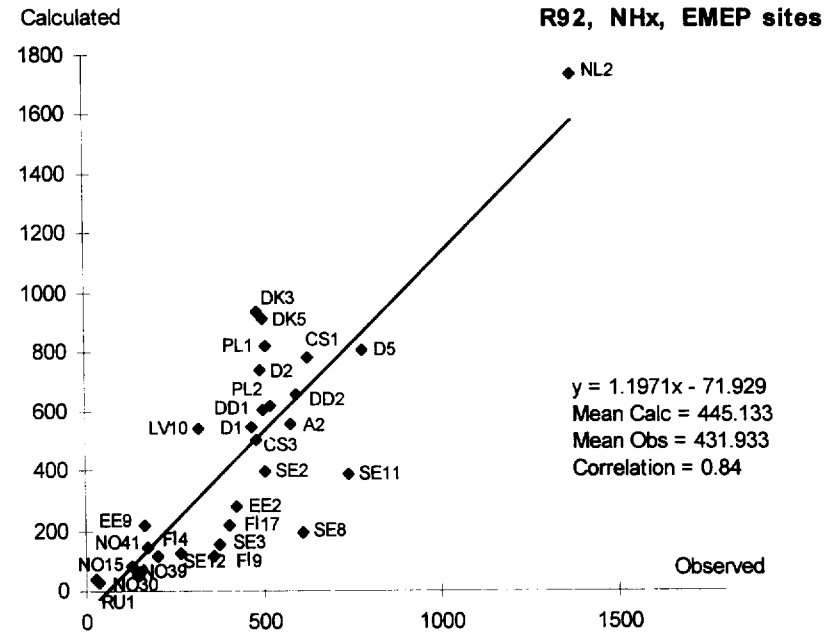
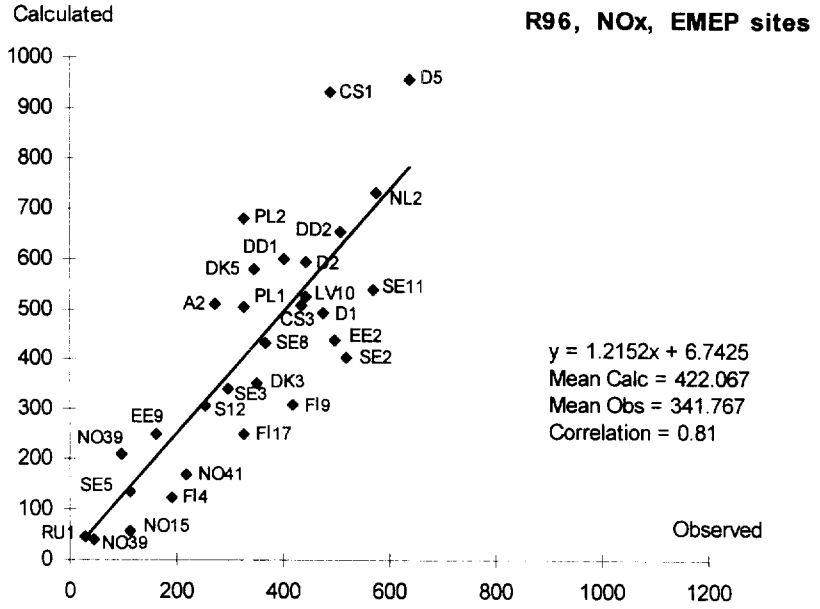


Figure C1. Comparison of NO_x and NH_x mean wet deposition in 1987–1990 measured at EMEP stations and calculated in RUN92, unit = mg N/m^2 per annum



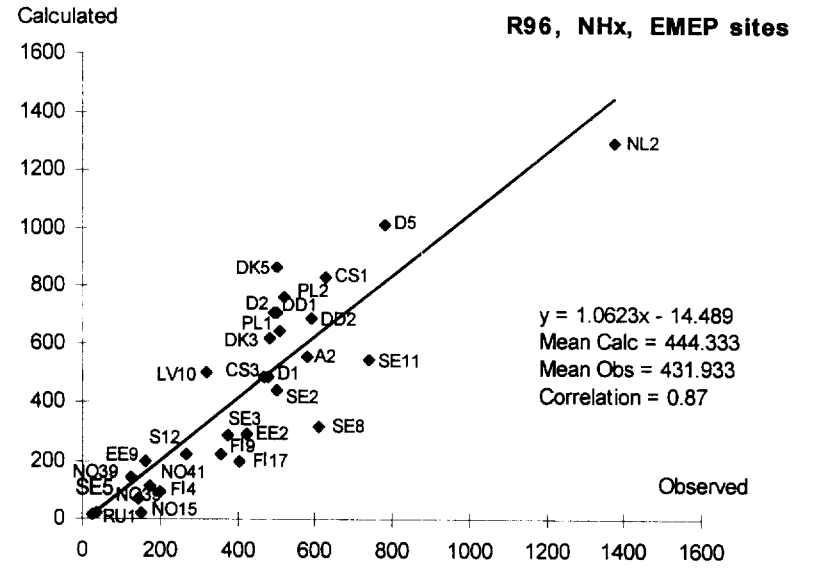
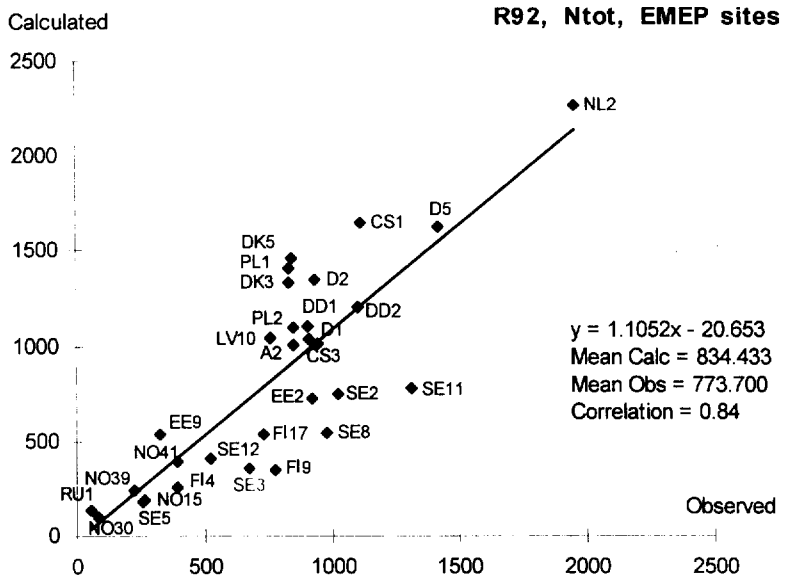


Figure C2. Comparison of NO_x and NH_x mean wet deposition in 1987–1990 measured at EMEP stations and calculated in RUN96, unit = mg N/m² per annum



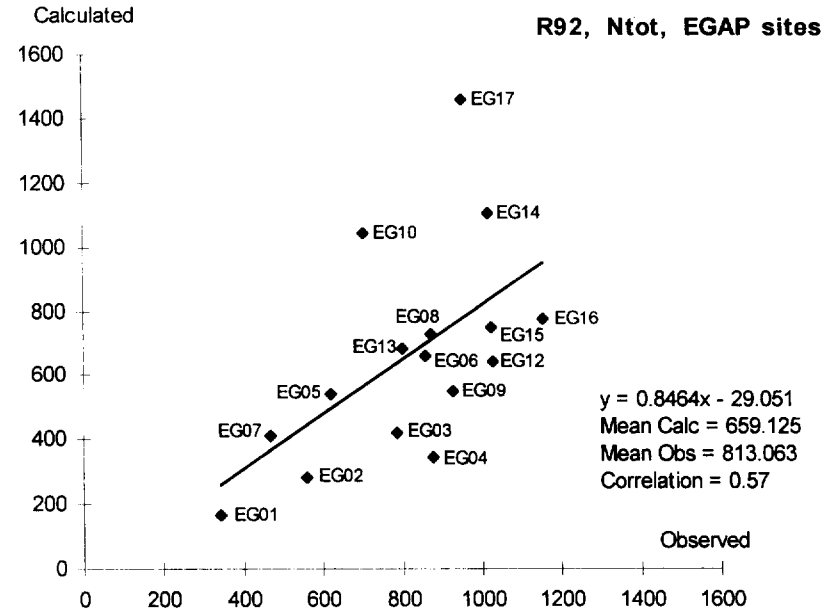
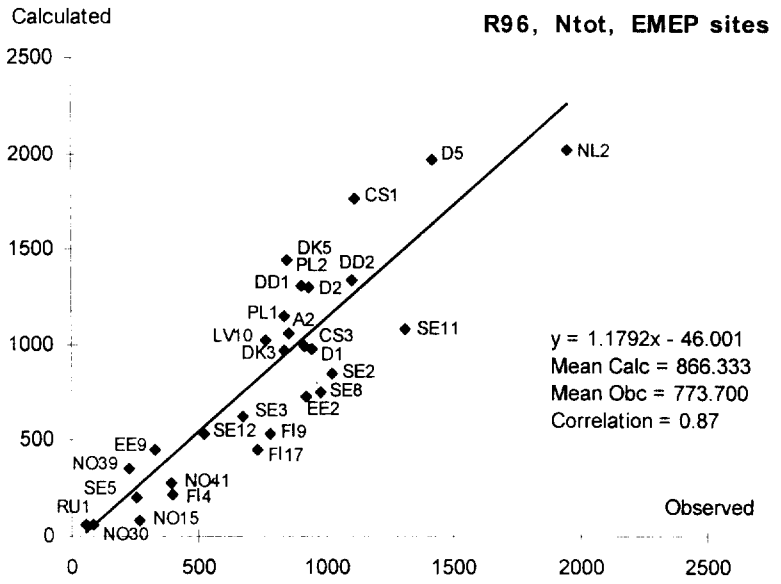


Figure C3. Comparison of N_{tot} mean wet deposition in 1987–1990 measured at EMEP and EGAP stations and calculated in RUN92, unit = mg N/m² per annum



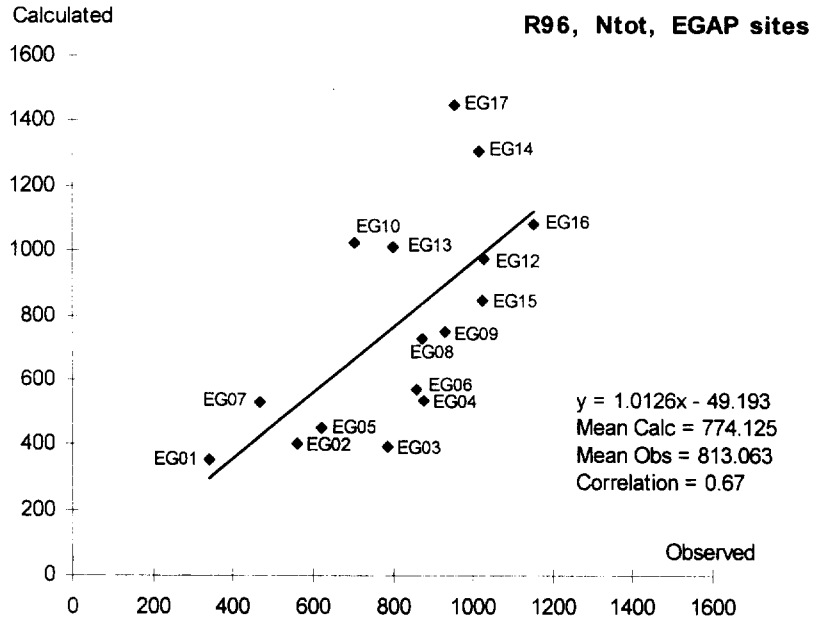
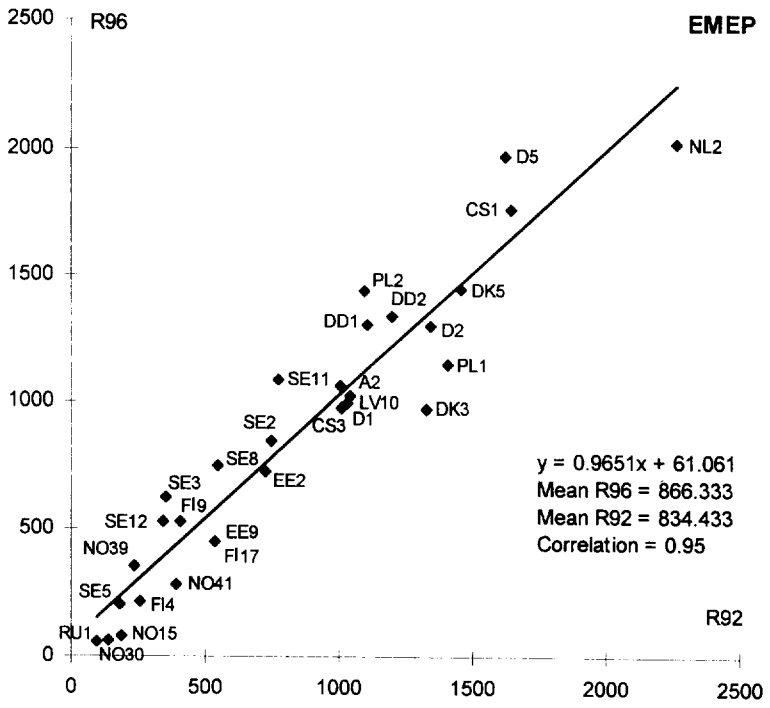


Figure C4. Comparison of N_{tot} mean wet deposition in 1987–1990 measured at EMEP and EGAP stations and calculated in RUN96, unit = mg N/m^2 per annum



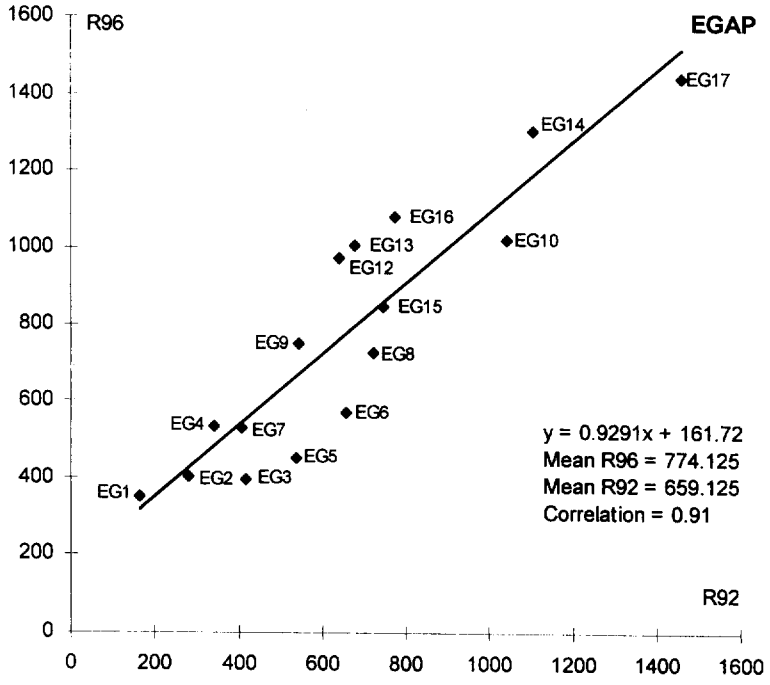


Figure C5. Comparison of RUN92 and RUN96 calculated results referred to grid squares with EMEP/EGAP stations (N_{tot} (NO_x + NH_x) mean annual wet deposition in 1987–1990), unit = mg N/m² per annum

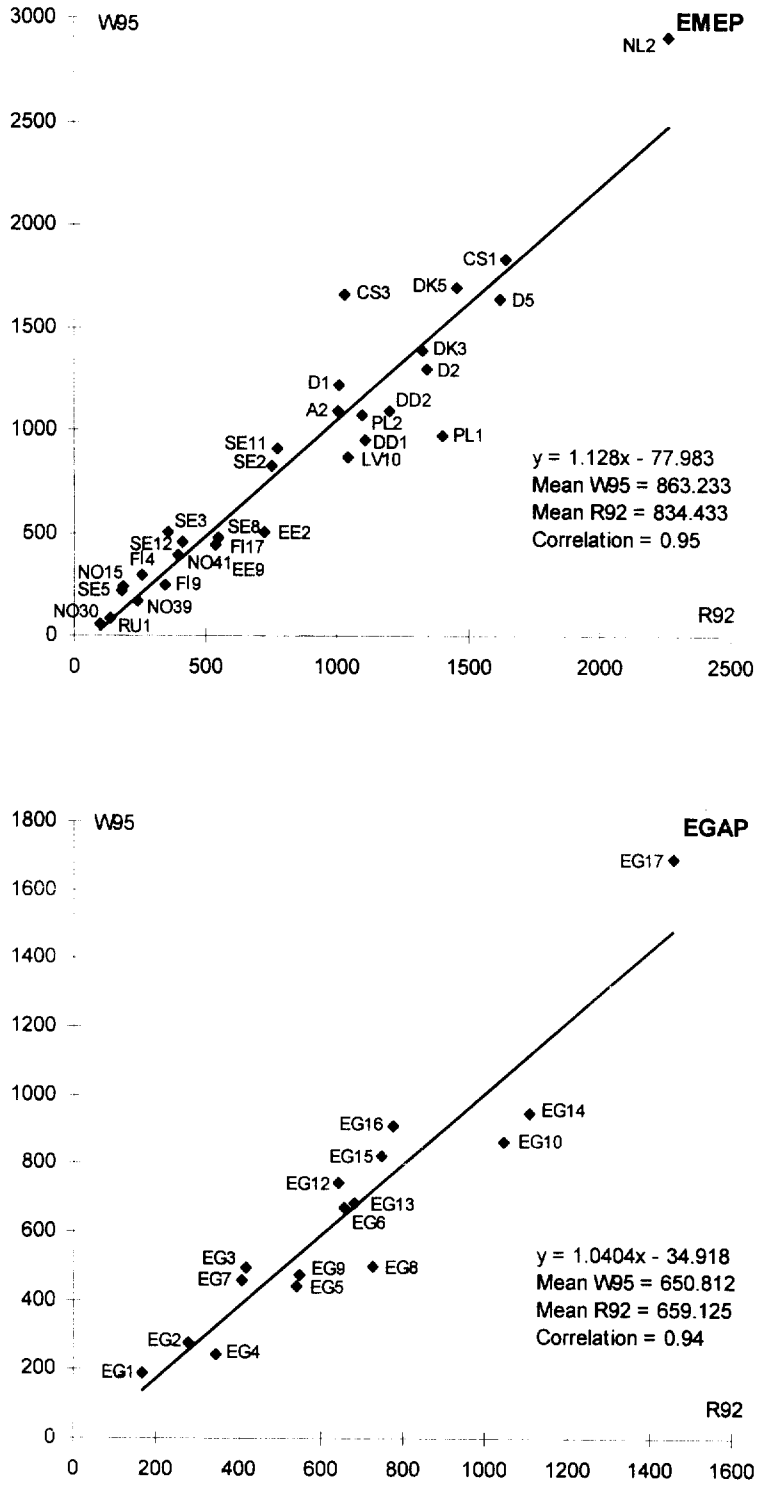


Figure C6. Comparison of RUN92 and W95 calculated results referred to grid squares with EMEP/EGAP stations (N_{tot} (NO_x + NH_x) mean annual wet deposition in 1987–1990), unit = mg N/m² per annum

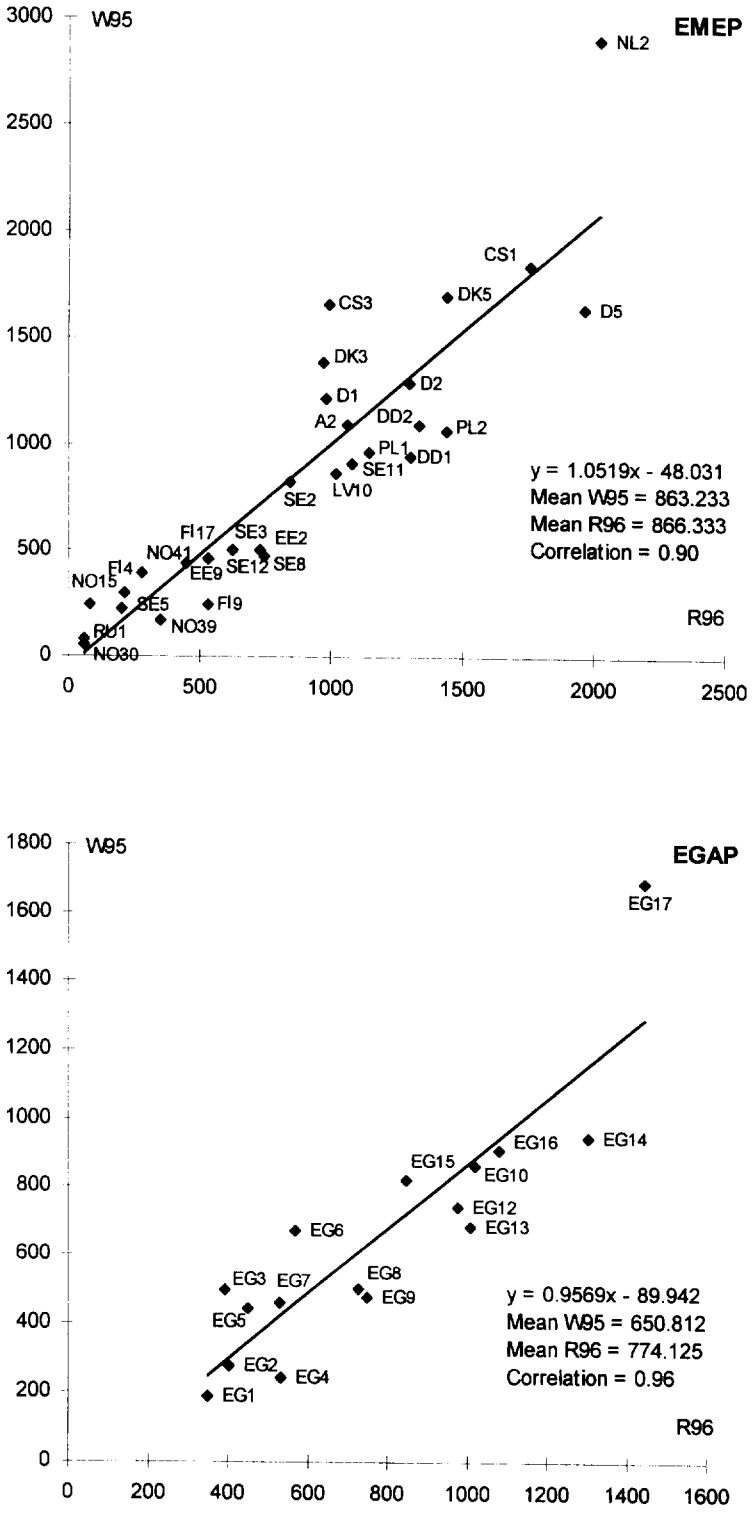


Figure C7. Comparison of RUN96 and W95 calculated results referred to grid squares with EMEP/EGAP stations (N_{tot} (NO_x + NH_x) mean annual wet deposition in 1987–1990), unit = mg N/m² per annum

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