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THE INPUT OF ANTHROPOGENIC AIRBORNE NITROGEN TO THE MEDITERRANEAN SEA THROUGH ITS WATERSHED

by

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PREFACE

The study is dedicated to the Mediterranean Sea and its watershed pollution with nitrogen compounds of atmospheric origin entering the sea through direct deposition and with riverine and groundwater runoff. This work was carried out by the Meteorological Synthesizing Centre-East (MSC-E) of the Cooperative Programme for the Monitoring and Evaluation of Long-range Transmission of Air Pollutants in Europe (EMEP) of the UN ECE Convention on Long-range Transboundary Air Pollution under a contract with and at the request of the World Meteorological Organization (WMO) which is a cooperating agency for the MED POL programme being responsible for coordination of the MED POL activities in monitoring, modelling and assessment of airborne pollution. Financial support for the study was provided from the Mediterranean Trust Fund.

The study report consists of 5 chapters:

1. Introduction prepared by ISSP RAS (Institute of Soil Science and Photosynthesis, Russian Academy of Sciences) and MSC-E.

2. Short description of the Mediterranean watershed (ISSP RAS).

3. Nitrogen deposition on the Mediterranean Sea and its watershed (MSC-E).

4. Modelling of deposition nitrogen runoff (ISSP RAS).

5. The comparison of atmospheric nitrogen deposition runoff with direct deposition on the Mediterranean Sea (ISSP RAS and MSC-E).

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SUMMARY

Calculations of the deposition of airborne anthropogenic nitrogen compounds on the Mediterranean Sea and its watershed basin (without Nile catchment area) for 1992 was carried out by a MSC-E model. Computed values of nitrogen deposition amount to 1097 kt of N for this part of the Mediterranean watershed and to 1084 kt of N for the Mediterranean Sea. Estimates of nitrogen deposition on the Nile watershed were taken from published results of global modelling. It was found to be equal to 560 ± 200 kt of N per year.

On the basis of nitrogen deposition data the calculations of the deposited nitrogen runoff to the Mediterranean Sea via rivers and groundwater were made. An assessment of this input was made by the method including calculation of deposition nitrogen retention in terrestrial and freshwater ecosystems on the scale of EMEP or ordinary longitude/latitude (LoLa) grid cells. It was demonstrated that the main source of the data uncertainty is related to nitrogen runoff and retention in different land cover types. It was found that leached fraction of deposited airborne nitrogen was distributed over the Mediterranean watershed basin rather uneven and varied from 0 up to 47%. Maximum values were for mountain areas of the northern part of the drainage basin where the highest precipitations coincide with the mountain relief. Input of airborne nitrogen from the Nile watershed was found to be practically negligible due to the low or absent runoff and very low precipitation in the lower part of the river and intensive uptake of nitrogen in its biogeochemical turnover in subtropical and tropical regions belonging to the upper Nile flow.

According to ISSP model estimates the total riverine runoff of deposited atmospheric nitrogen to the Mediterranean Sea amounts to 99 kt of N. On the basis of published data the value of ground water nitrogen runoff was estimated as much as 2-9 kt of N. Consequently the total runoff of airborne nitrogen from the watershed is estimated as 105±4 kt or about 6% of the atmospheric anthropogenic nitrogen deposited on the Mediterranean Sea watershed.

Budget of the total nitrogen input to the Mediterranean Sea from land-based sources was considered. It consists of the input from rivers and coastal zone (800-1200 kt N, including the runoff of airborne nitrogen from the watershed 105±4 kt) and direct deposition on the Mediterranean Sea surface (1084 kt of N).

Thus, the value of the airborne nitrogen deposition runoff (- 100 kt of N) is equal to about 5% of the total nitrogen load to the Mediterranean Sea from land-based sources (~2 Mt of N) or about 10% of the anthropogenic nitrogen deposition on the Mediterranean Sea surface (- 1 Mt of N).

1. INTRODUCTION

The nitrogen input into the Mediterranean Sea aquatorium occurs due to river runoff, direct urban and industrial waste runoff and due to nitrogen deposition directly into the sea waters. The first type of the given inputs is also connected with atmospheric deposition of nitrogen but it is very difficult to distinguish the role of atmospheric deposition nitrogen in biogeochemical balance of this element, especially that part of nitrogen which reaches the Mediterranean Sea aquatorium being deposited on its drainage basin. 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. [*EC EGAP 9/4b/1,* 1992; *Bashkin et al*, 1995; *Bashkin et al*, 1996; *Nixon et al*, 1996; *Galloway et al*, 1996; *Prospero et al*, 1996].

As already noted in [UNEP, 89, p.10]:

"The Mediterranean Sea is well-adapted to avoid excessive eutrophication. It loses deep water, relatively rich in mineralized or recycled nutrients, and receives surface Atlantic water, in which most nutrients have been used before entering Gibraltar. The situation is exactly the opposite of that in the Baltic, where ecological mechanisms tend to recycle and accumulate large amounts of nutrients."

Self-purification of the Mediterranean Sea is sufficiently high and oxygen deficiency is usually observed in the vicinity of local sources of eutrophication more frequently involving industrial and municipal discharges. The distribution of these sources along the Mediterranean Sea is uneven. Their maximum fell within its north-western part and the Adriatic and the minimum - its south-eastern part.

"Waste loads of domestic sewage, industrial discharges and agricultural run-off are probably the major contributions polluting the Mediterranean Sea. The uneven distribution of runoff and precipitation along the northern coasts of the Mediterranean Sea, combined with the northern concentrations of population and industrial activity, contributes a waste load of pollutants to Mediterranean waters that is confined first to the northern coasts and then is spread and recirculated through the natural processes of advection and convection."

The nitrogen loading with river runoff, direct urban and industrial discharge to the sea in total amount to 800-1200 kt/yr [*UNEP*, 1989]. According to MSC-E calculations, the nitrogen deposition on the Mediterranean Sea surface from the atmosphere is about 1000 kt [*Erdman et al.*, 1994] being close to those characterizing the riverine loading. But until now there is no quantitative assessment of the role of nitrogen that being deposed on the Mediterranean Sea watershed enters the Mediterranean Sea with riverine discharge and direct coastal surface runoff. Even the most recent and comprehensive assessment of atmospheric deposition of nitrogen on the North Atlantic Ocean and its watershed (Prospero et al., 1996) does not give the quantitative assessment of the N atmospheric deposition amount in its riverine fluxes to the ocean and seas from the watersheds.

Furthermore, assuming that the nitrogen deposition is the only source of this element in natural landscape, the background loads of nitrogen to riverine fluxes might be assessed to represent the conditions in the watershed not affected by human activity but to find such watersheds which meet these conditions is almost impossible. Thus, the best estimate of background loads could be made for small, scarcely populated catchments with low human activity and this gives an opportunity to compare computed values with experimental monitoring of N runoff. However, contrary to the Baltic Sea drainage basin, there are only a few such investigations in the Mediterranean Sea watershed.

Thus in accordance with the contract between the World Meteorological Organization and the Meteorological Synthesizing Center-East for 1995/1996 FY, the report is aimed at the calculation

of nitrogen deposition from the atmosphere on the watershed basin of the Mediterranean Sea, the assessment of the input of airborne nitrogen to the sea through surface and underground runoffs from the watershed, and the comparison of this input with direct deposition of nitrogen to the Mediterranean Sea surface on an annual basis.

2. BRIEF DESCRIPTION OF THE MEDITERRANEAN SEA DRAINAGE AREA

The Mediterranean drainage basin is a vast area conjugating the territory with the surface runoff to the Mediterranean Sea including the Nile watershed. The total area of this drainage basin is 4,666,400 sq.km (1,796,400 sq.km without the Nile watershed which is equal to 2,870,000 sq.km). The sea surface area covers 2,505,000 sq.km.

For the sake of convenience the Mediterranean watershed is divided into watershed proper (without the Nile watershed) hereinafter referred to as WSH1 and the Nile watershed hereinafter referred to as WHS2 (Figures 2.1 and 2.2).

The following countries belong to the proper Mediterranean drainage basin (WSH1) having the direct coastal line: Spain, France, Italy, the former Yugoslavia*, Albania, Greece, Turkey, Syria, Lebanon, Israel, Jordan, Egypt, Libya, Tunis, Algeria, Morocco; a part of Switzerland is also included to the Mediterranean basin but without a coastal line.

The biggest part of the Mediterranean drainage basin (WSH1) is situated in the evergreen dry forests and bushes of subtropical climate. The natural unity reflects the predominance of Mediterranean climate types, vegetation, soil, water regime of rivers, and agricultural land use types as well as regional peculiarities of relief and humidity which determine the variety of landscape types.

A characteristic climate feature is a predominance of dry hot summer and nonfrozen wet winter. The humidity and precipitation decrease from north to south and from west to east (from 2000-3000 mm up to <100 mm). The permanent snow cover is absent in the northern part up to the 300 m asl, in southern ones - up to 900 m. In the majority of terrestrial ecosystems of the Mediterranean Sea drainage basin the evaporation exceeds the precipitation quantity: the difference is deviated from 0-400 mm up to 1600-2400 mm in the central part of the Nile basin. The negative precipitation-evaporation balance is also predominant for the aquatorium of the Mediterranean Sea where the precipitation values are 100 mm in the south-east part and 1000 mm in the north part.

In the whole area of this drainage basin the rivers with rain and snow-rain (in the mountains) type of water inflow are predominant. The maximum runoff is in autumn-winter and spring periods; this runoff decreases significantly during the summer time. The river water is used widely for irrigation. The values of full river runoff decrease from 600-1000 mm in the northern mountain regions up to 10-15 mm in southern desert areas. The runoff increase is also connected with up welling and carst regions. There are two groups of water balance and runoff. The undersurface runoff is more typical for the southern part of the Balkan mountains and Asia Minor (25-40 and up to 50%) whereas in the northern Africa the undersurface runoff does not exceed 20-25% and these values are related to very high contrast of precipitation during the seasons.

There are more than 70 rivers in the drainage basin of the Mediterranean Sea (Table 2.1).

The predominant part of the given area is presented by the Mediterranean soil region where the following soils are typical:

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* Note: The nitrogen deposition data used in this report are for 1991. For that time, the data exist only for the whole former Yugoslavia but not for individual states which have been formed on its territory.

- brown soils (cambisols) of dry forests and bushes having neutral pH values in the upper part of soil profile, saturated by basic cations, humus content up to 4-5% and sufficient nutrient content;
- brown forest soils are placed in the more humid north conditions with predominance of mountain subtypes, the main parameters of which are related to parent materials;
- gray-brown soils under xerophytic bushes and grasses having low depth, high carbonate contents with some spots of solonchaks in relief depressions (mainly placed in Spain);
- heavy montmorillonitic soils widespread at flat ancient alluvial plains of Pyrenees, Balkans and Apennines are used widely for irrigated rice plantations.

On the whole, the area of the Mediterranean soil region is up to 50% in agricultural land use.

In the east Asian part of this basin as well as in north mountain regions of Algeria and Morocco the mountain subtypes of above mentioned soils are widespread and up to 30% of them are used in agriculture. The soils of flat desert steppes of the south coast of the Mediterranean Sea basin were formed on limestone deposition with carbonate materials from the soil surface, these soil have low humus content and reddish colour. The relief depressions are under solonchaks. These areas are predominantly used for pastures.

The Nile river drainage basin (WSH2) consists of the valley and delta with characteristic features of irrigated ancient soils, with alluvial-meadow and alluvial-swamp soils and marshes as well as the tropical rain forests and savannas predominant in the upper part of the Nile watershed.

3. NITROGEN DEPOSITION ON THE MEDITERRANEAN SEA (MDT) AND ITS WATERSHED (WSH)

3.1 Introductory comments

In this chapter deposition of nitrogen compounds on the Mediterranean Sea (MDT) and its watershed (WSH) are considered.

The assessment of nitrogen deposition on MDT has already been made by MSC-E. The Report [*Erdman et al*, 1994] provides estimates of total depositions of nitrogen compounds in 1991 along with separate quantification of wet and dry depositions, deposition seasonal variations, deposition distribution with MDT subbasins and countries-sources. The total deposition of nitrogen compounds on MDT in 1991 [*Erdman et al*, 1994] was estimated as 1067 kt. In present study the deposition for 1992 was estimated as 1084 kt.

Some estimates of nitrogen depositions on MDT within the geographical scope of the EMEP grid (see Figure 3.1) are also presented in the work of *Barret et al* (1995). They are 606 and 607 kt of nitrogen in 1991 and 1992 respectively but the calculation region of EMEP does not cover the eastern and south-eastern parts of MDT.

Integral estimates of nitrogen deposition directly on WSH had not been made or at least published before preparation of the present report. As it was already mentioned in Chapter 2, WSH1 is a part of the MDT drainage basin without the Nile watershed. Some ideas on the nitrogen load on WSH1 can be inferred from assessments of depositions on the Mediterranean countries in 1991 provided by MSC-E [*Erdman et al*, 1994] and MSC-W [*Barret et al*, 1995] and presented in Table 3.1. It is evident that nitrogen deposition on the Mediterranean countries is nearly twice as high as on MDT. Since WSH1 area is only a part of that of the Mediterranean countries then deposition on it is of the same order of magnitude as depositions on MDT. Total deposition on WHS2 had not been estimated. There are also nitrogen deposition maps of the region in question compiled within the framework of global models ([*Rodhe et al*, 1995], [*Dentener&Crutzen*, 1994], [*Gallardo&Rodhe*, 1995]).

TABLE 2.1: *Mediterranean watershed area and flows (updated 1987)*** [UNEP, 1987]*

	Area (1000 km^2)		Theoretical resources (km^3/year)				Stable resources (km^3/year)	
			Total					
	Country	Med. region	Inc.neighbouring country inflows	Internal	Surface runoff	Underground runoff	Low flow = low water mark	Underground flow of coastal watertables
SPAIN	505	180	109.9	109.9	89.4	20.4	17.0	3-4
Med.region			28.3	28.2	19.8	8.4	6.3	1.6
FRANCE	550	130	185.0	170.0	70.0	100.0	85.0	1.1
Med.region			74.0	62.0	31.0	31.0	35.0	0.2
ITALY	300	300	187.0	185.0	155.0	30.0	18.5	12.0
Med.region								
MALTA	0.3	0.3	0.03	0.03		0.03	0.007	0.023
YUGOSLAVIA	256	80	265.0	150.0	130.0	20		
Med.region			77.5	77.5	62.0	15.5	6.5	5.0
ALBANIA	29	29	21.3	10.0	**	**	6.5	
GREECE	132	132	62.9	49.4	37.4	12.0	7.0	2.5
TURKEY	780	195	181.0	166.0	156.4	9.4	**	**
Med.region			77.0	70.0	**	**	14.5	1.1
CYPRUS	9.3	9.3	0.9	0.9	0.6	0.3	0.2	0.01
SYRIA	185	22	35.4	7.6	3.1	4.5	14.0	0.2
Med.region			4.4	3.4	1.3	2.1	2.1	0.2
LEBANON	10.4	9.8	4.8	4.8	1.3	3.5	2.1	0.9
Med.region			4.0	4.0	1.0	3.0	1.9	0.9
ISRAEL	21	12	1.0	1.0	0.2	0.8	0.2	0.08
Med.region								
EGYPT	1000	144	57.5*	1.0	0.5	0.5	55.5	0.3
Med.region			3	0.8	0.3	0.5	55.5	0.3
LIBYA	1760	250	0.6	0.6	0.1	0.5	0.1	0.1
Med.region			0.6	0.6	0.1	0.5	0.1	0.1
TUNISIA	164	90	4.35	3.75	2.13	1.62	1.0	0.7
Med.region			3.0	2.5	1.8	0.8	0.8	0.7
ALGERIA	2380	133	16.0	15.7	13.6	3.3	**	**
Med.region			13.2	13.2	12.0	1.2	1.8	0.7
MOROCCO	710	80	29.0	29.0	20.0	9.0	2.5	0.9
Med.region			4.0	4.0	3.0	1.0	0.6	0.2
MED.TOTAL	8792	1796.4	**	497.0	427 4	**	157.5	25.4

(*) Regular flow Former natural flow = 86 km^3/year; former low-water flow = 24 km^3/your
(**) Data are absent
(***) Source: *Blue Plan*, 1987: UNEP,87. Environmental data on Mediterranean Basin (natural environments and resources) Provisional version, Blue Plan, Sophia Antipolis

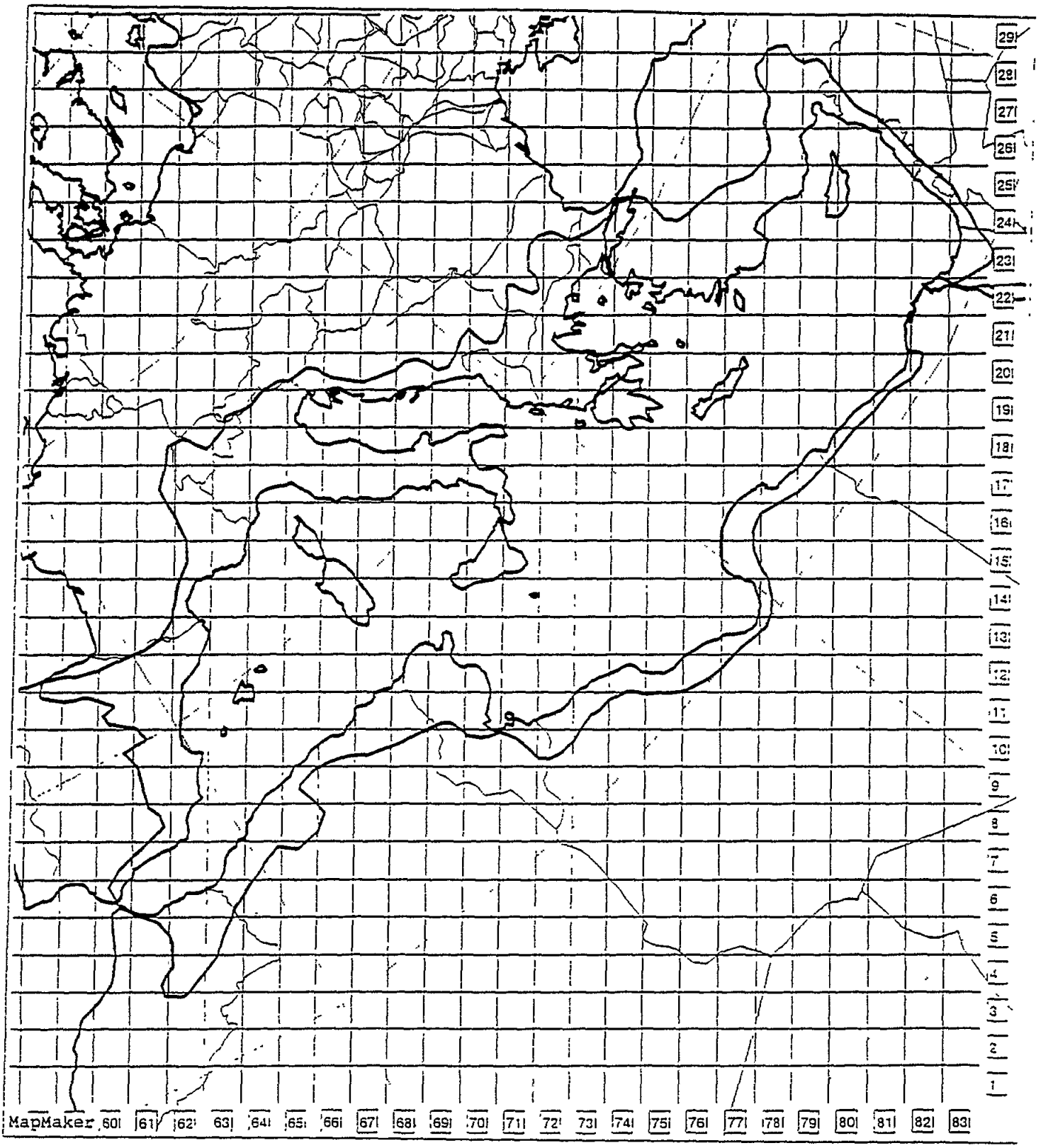

Figure 2.1: The Mediterranean Sea and its watershed (The Nile watershed is not included)

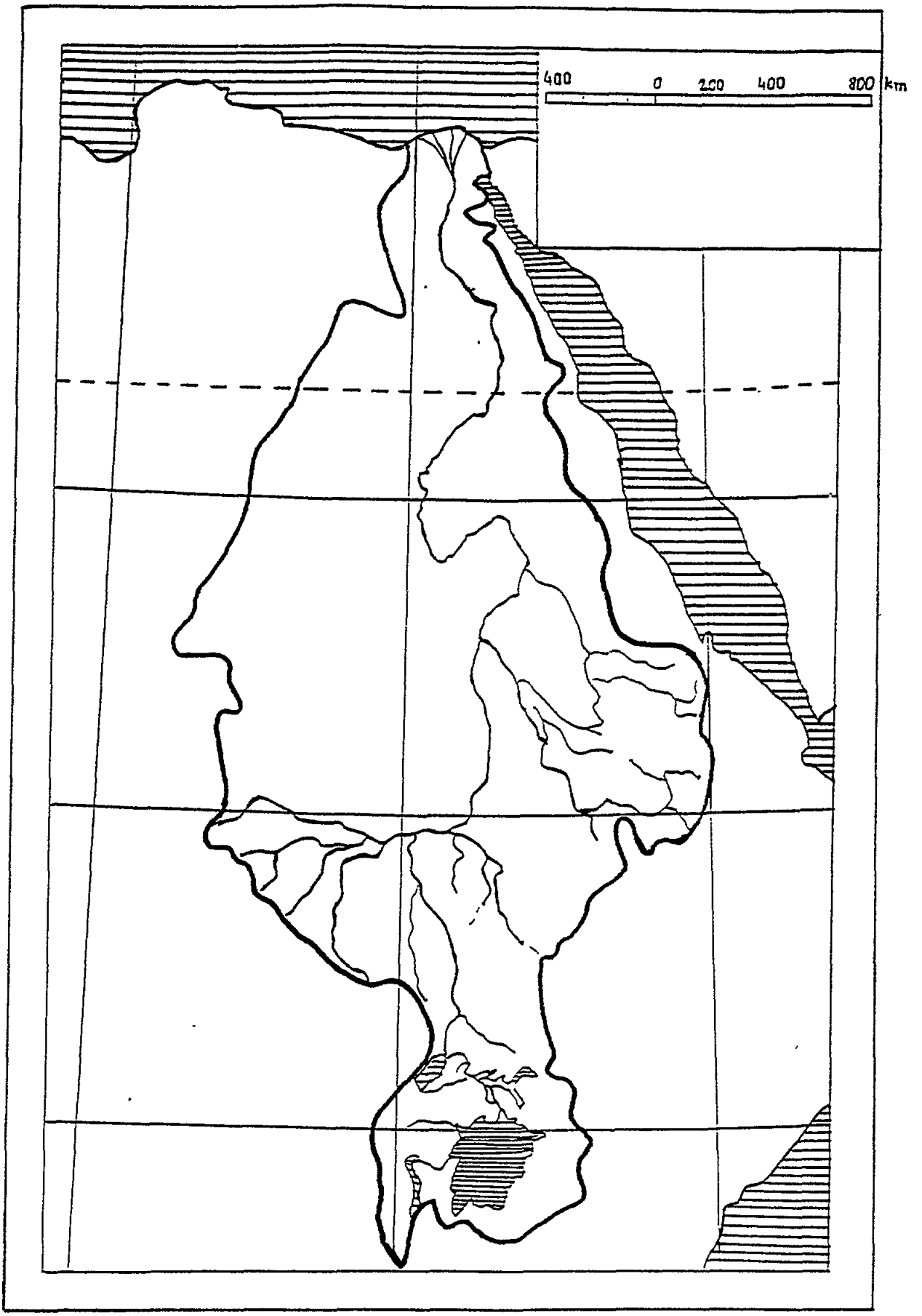

Figure 2.2: The Watershed of the Nile

TABLE 3.1: *Calculated depositions of nitrogen compounds on the Mediterranean Sea and Mediterranean countries in 1991. Unit = 1000 t N/year*

	Area (1000 km^2)		Depositions					
	Country	Med. region	MSC-E data			MSC-W data		
			NO_x	NH_x	N_{tot}	NO_x	NH_x	N_{tot}
SPAIN	505	180	104	124	228	119	174	293
FRANCE	550	130	353	484	837	250	379	629
ITALY	300	300	173	150	323	164	214	378
YUGOSLAVIA**	256	80	158	159	317	118	157	275
ALBANIA	29	29	14	13	27	11	18	29
GREECE	132	132	55	37	92	31	50	81
TURKEY	780	195	105	275	380	76	185	261
SYRIA	185	22	8	30	38			
LEBANON	10	10	1	1	2			
ISRAEL*	21	12	1	1	2			
EGYPT*	1000	144	13	13	26			
LIBYA*	1760	250	37	26	63			
TUNISIA*	164	90	20	23	43			
ALGERIA*	2380	133	44	59	103			
MOROCCO*	710	80	1	2	33			
MED.SEA			648	419	1067	407	199	606

* within the calculation area

** all countries located on the territory of the former Yugoslavia

Hence this chapter is dedicated to:

1. Calculations of deposition fields of nitrogen compounds for MDT and WSH1 and total deposition on these regions.
2. Estimation of total nitrogen deposition on the Nile watershed using deposition fields calculated by global models.

The results obtained are used as the input for calculations of the airborne nitrogen run-off from WSH to MDT.

3.2 The process of airborne nitrogen transport. Peculiarities of the Mediterranean region

Nitrogen compounds are emitted to the atmosphere as NO_x (predominantly as NO) and as NH_x (actually as ammonia). Fuel combustion in industry and transport is the main source of NO_x emission. As to NH_x its main source is agricultural activity (livestock husbandry and the application of fertilizers) and fertilizer production. Burning of biomass is a considerable source in the tropics. Thunderstorms (for NO_x) and soil and vegetations (for both species) are natural sources of emissions.

The fate of nitrogen in the atmosphere depends on the initial height of its emission. In general the higher a pollutant is emitted the longer its transport can be. Normally the industrial emission sources are high (up to several hundreds of metres), and the transport emission sources are low. Ammonia emission sources from agricultural activity are low and sources of fertilizer plants can be high. Sulphur is also involved in the cycle of ammonia chemical transformation and more than a half of sulphur emission sources are high.

In the spatial distribution of nitrogen emissions the peculiarity of the MDT location plays an important role: the Mediterranean Sea is situated in a "trough" between two sufficiently powerful regions-emitters - central European and central African ones. The first region is characterized by highly developed industries and agriculture, the second one - by the activity of micro-organisms in tropical rain forests (only NO_x) and biomass burning (NO_x and NH_x) ([*Dentener &Crutzen*, 1994], [*Penner et al*, 1991]). Such distribution of sources in many respects determines the gradient of nitrogen deposition.

Nitrogen is transported by air flows. Absolute values and directions of transport velocities are controlling factors here. One of the most important peculiarities of the Mediterranean region is noted in [*Dayan et al*, 1989] "The topography behind the coastline of the Mediterranean is complex; it provides both barriers to and channels for air flow that bring at times extremely different air masses to the region. Strong winds, which are funnelled through gaps in the mountain ranges that surround the Mediterranean Basin, are among the best known meteorological features of the region: (1) **the north-westerly mistral** through the Alps-Pyrenees gap; (2) **the north-easterly bora** through the Trieste gap; (3) **the easterly levanter** and the **westerly vendaval** through the Strait of Gibraltar; and (4) **the warm south-easterly to south-westerly scirocco, ghibli, or Khamsin from Africa."**

In two thirds of cases the air masses reach the western part of MDT from the northern and western directions and the eastern part - from the north-eastern and north-western directions (data are obtained on the basis of a trajectory analysis at the level of 1000-850 hPa [*Dayan et al*, 1989, Fig. 31]).

In winter months for both eastern and western parts of MDT the initial points of reverse trajectories are located closer to the west and during the rest of the year - closer to the north [*Dayan et al*, 1989, Figs. 32 and 33]. The mean velocity of the atmospheric transport up to two days is 10 m/s and more for the predominating direction and it is two times less for less probable directions [*Dayan et al*, 1989, Figs.34 and 35].

In the course of transport the pollution is diffused in both horizontal and vertical directions. The velocity of vertical diffusion to a great extent is connected with the atmospheric stability in the point considered. In general the atmosphere above MDT is rather stable and the mixing layer depth is about 1000 m. This estimate is reasonable for the atmosphere above large water mass and it is confirmed by measurements of more than 100 radiosounding stations [*Dayan et al*, 1989, p.3]. However, the interaction of the large-scale atmospheric flows with peculiar orography of the MDT coastline should affect both horizontal transport and vertical mixing which should have been taken into account in the simulation in the scale of hundreds of kilometers.

The transported pollutants are scavenged from the atmosphere due to capture by the underlying surface (dry deposition) and due to wet deposition, i.e. scavenging by precipitation and cloud moisture. The Mediterranean region is characterized by a prolonged dry season. It is also true for desert and semidesert regions of North Africa.

A climatic pattern of the annual precipitation variation has a zonal character with distinct minimum (<100 mm/year) at 15^0-30^0 N with a gradual increase of precipitations to the north and south. In the vicinity of the equator the precipitation norm amounts to 1000-2000 mm/year and in the northern coast - 500-1000 mm/year. Therefore the share of dry deposition in nitrogen total deposition in the MDT region is greater than on the average for Europe and in particular for the European north-western part.

During the transport nitrogen compounds are involved in a number of chemical transformations with the participation of sulphur compounds, VOC`s and ozone. The chemical transformation scheme and basic reactions characteristic of the long-range transport of nitrogen compounds used in the MSC-E model are given in Annex A dedicated to the description of the MSC-E model. In comparison with average European conditions the MDT region is characterized by enhanced insolation, high temperatures and reduced humidity. These factors affect a number of reaction rates.

All these features are reflected in transport models of pollution in particular of nitrogen compounds. Individual characteristics of the process are treated in different ways depending on models, their specific designation and even personal inclination of modellers.

For the evaluation of depositions on MDT and WSH1 special calculations were made using the MSC-E regional model (the version of 1996). The evaluation of deposition on WSH2 were based on calculation results obtained by the global model MOGUNTIA ([*Rodhe et al*, 1995], [*Dentener&Crutzen*, 1994], [*Gallardo&Rodhe*, 1995]). Calculations with the MSC-E model are made for 1992, the MOGUNTIA assessments are climatic. Necessary information on the models used, input data and calculation results are discussed.

3.3 The MSC-E model. General description. Information fluxes

The MSC-E model developed in the period of 1991-96 was used for the assessment of depositions on MDT and WSH1. It is an Eulerian one-layer model with allowance for vertical distribution. It operates with real 6-hour meteorological information. The model is described in Annex A. The input data involve geographical, meteorological and emission information. Measurement data (if any) are used only for the evaluation of the quality of results.

3.3.1 Geographical data

The region covered with calculations of deposition on MDT and WSH1 is shown in Figure 3.1. It is produced by the shift of the standard EMEP grid which is also shown in the figure. The grid size is 150 x 150 km^2 at latitude 60^0 and it is reduced with the latitude on account of the scale distortion in the polar stereographic projection. The shift of the calculation region relative to the EMEP grid comprises 5 squares along both co-ordinates and it is directed from the pole. The choice of the calculation region was influenced by the wish to cover more completely the area of MDT and WSH1 and therewith to take into account the availability of meteorological and emission data.

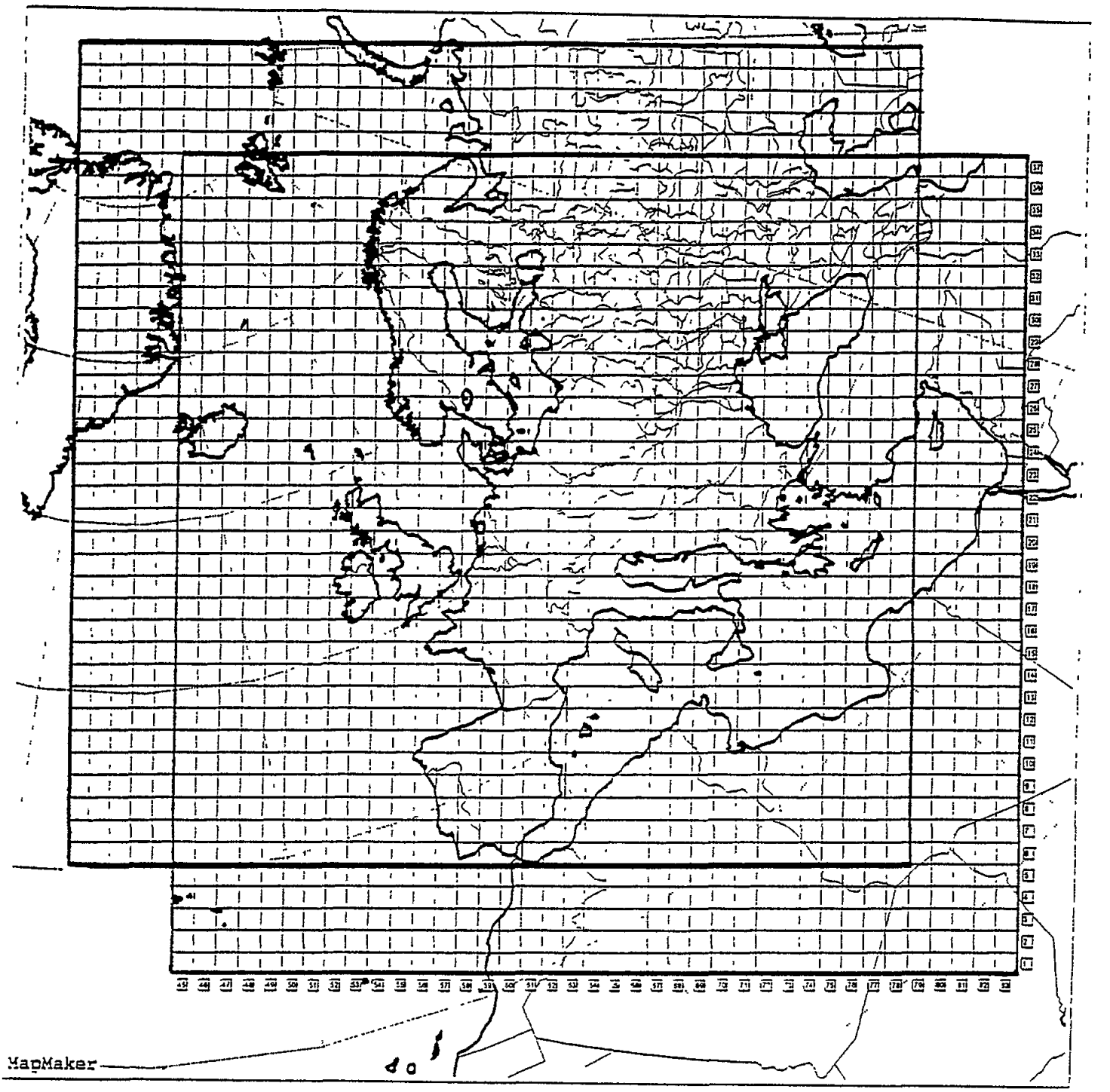

Figure 3.1: The calculation region (lower right rectangle) and the EMEP region (upper left rectangle)

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).

3.3.2 Meteorological information

Meteorological information used (1992) includes:

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

The averaging interval is 6 hours, spatial resolution - 150150 km^2. The data are prepared by the 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 the Russian Hydrometeorological Centre the models estimate the state of the underlying surface and other physical parameters important for the calculations.

It should be noted that for the MDT region especially for its southern part there is a deficiency of observational meteorological data in particular on precipitations. "The information about seasonal precipitation fields over the Mediterranean including rain frequencies and rates, is not sufficient" [UNEP, 92, Para 4.4.4]. This fact should be taken into account in the assessment of uncertainties of the obtained results.

3.3.3 Emission data

The bulk of anthropogenic pollution by sulphur compounds, nitrogen oxides and ammonia comes from various sectors of industry, transport and agriculture.

Nitrogen oxides

Nitrogen oxides (NO_x) usually include two pollutants - nitrogen dioxide (NO_2) and nitric oxide (NO). In comparison with SO_2, NO_x is less important, although significant contributor to acid deposition. NO_2 is significant in terms of impact on human health. In addition, NO_2 and NO are precursors of ozone (O_3), a greenhouse gas.

Similar to SO_2, NO_x emissions are strongly dependent on fossil fuel combustion. The CORINAIR 85 inventory indicated that 93% of the total NO_x emission in Europe are emitted from fuel combustion (including 54% from road transport, 24% from power plants and 6% from non-industrial combustion). The remained categories (oil refineries and production processes) to some extent are also dependent on the use of fuel. According to CORINAIR 90 (1995), 57% of NO_x emission are accounted for road transport and other mobile sources and 39% - for stationary ones (industrial and non-industrial fuel combustion and power stations).

Ammonia

According to the CORINAIR 85 inventory, 80% of the atmospheric ammonia (NH_3) emission are produced from the decomposition of wastes from domestic animals. About 10% of NH_3 atmospheric input is due to nitrogen fertilizers application. The rest of NH_3 emission originates from other sources, mostly from industrial nitrogen fertilizers production. From CORINAIR 90 the share of agriculture as a whole amounts to 92%.

It should be noted that in the African part of the calculation region biomass burning makes a certain input to the emission of the listed species. *Rodhe et al*, (1995, Table 1) pointed out that the fraction of biomass burning is about 20% of global NO_x emission. In the spatial distribution of NO_x emission originated from biomass burning the maximum is observed in tropical regions of Africa (alongside tropical regions of South and Central America, Australia and south-eastern Asia [*Penner et al*, 1991]). In calculations of deposition on WSH1 this sector of emission is not included.

Sulphur dioxide

Sulphur dioxide (SO_2) was one of the first pollutants to be inventoried due to its impact on human health and input to acid deposition. SO_2 is one of the air pollutants the emission of which is easy to quantify since it results from fossil fuel combustion and hence depends on the sulphur content in the fuels. Here sulphur compounds are considered because of their importance for chemical transportations of reduced nitrogen.

According to the CORINAIR 85 inventory [*CORINAIR* 85], about 66% of the total SO_2 in Europe are emitted by power plants and non-industrial (domestic) combustion, 19% by industrial combustion and the remaining 15% from oil refineries, road transport and various production processes, many of which also include some use of fossil fuel. According to CORINAIR 90 (1995) power plants and domestic fuel combustion emit 65% SO_2 and 25% - industry.

As to the source height then again from CORINAIR 85 about 55% of SO_2 emission were produced by large power plants. These power plants are relatively few in number: there were 1446 point sources inside the EMEP grid with thermal heat input greater than 50 MW. These plants are usually considered as a source with emission at >100m height. According to CORINAIR 90, high sources are responsible for more than 50% of SO_2 emission as well.

Emission data for 1992 used in calculations within the EMEP grid were provided by MSC-W of EMEP in 1993. They were updated with allowance for official total emissions in 1995 [UN Economic Commission for Europe, document EB.AIR/R.92/Add.1]. The latest official data were also published, for example, in [*Barret et al*, 1995]. For the Commonwealth of Independent States (CIS) countries some expert estimates were used.

Emission estimations for sulphur and nitrogen oxides for the Mediterranean countries within the limits of the calculated region but outside the EMEP grid were based on assessments made by EMEP/MSC-W/CCC for the north Africa (Algeria, Tunis, Libya) [*Sandnes&Styve*, 1992]. Specific values of the above mentioned compounds per capita for these north African countries were used for the emission estimations in the region of calculation allowing also for the industrial development level and population [*UNEP*, 89].

The main source of ammonia input to the atmosphere in the majority of countries is cattle breeding. NH_3 emission estimations were made on the basis of available data on quantity of domestic cattle and poultry in each country and averaged emission factors for each species [*Annual reference data*, 1990; *Buisman et al*, 1987; *Van der Most & Veland*, 1992; *Thomas & Erisman*, 1990].

Emission from the sea area is taken from international shipping [*Sandnes & Styve*, 1992].

Data on sulphur and nitrogen oxides and ammonia emissions from the considered countries for 1992 are given in Table 3.2. The same table contains the codes used for countries names, groups of countries and individual water basins.

Figures B1 and B2 (Annex B) give maps of oxidised and reduced nitrogen emission intensities for each square of the calculated region. Figure 3.2 shows the emission seasonal variations in conventional units.

3.3.4 Output data

The model output data are presented as maps of nitrogen deposition separately wet, dry and total and as budget matrices of depositions of countries (regions) -emitters to countries (regions)-receivers.

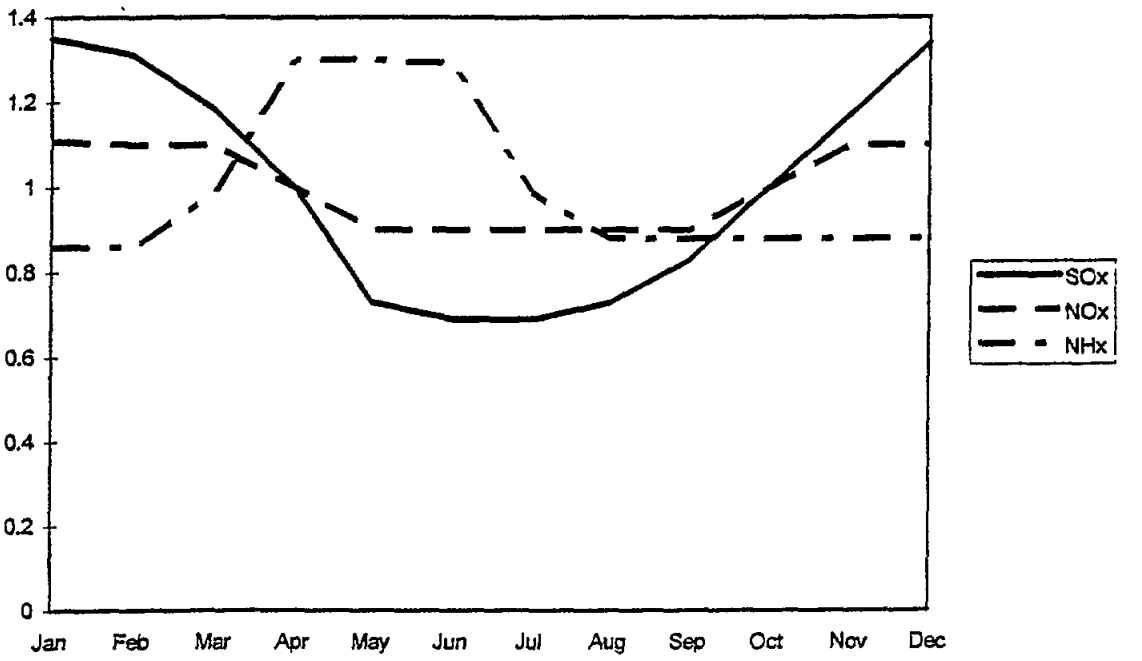

Figure 3.2: Seasonal SO_x, NO_x, and NH_x emission variations for all countries (in per unit)

3.4 Depositions on the Mediterranean Sea and its watershed

3.4.1 Deposition on the Mediterranean Sea

In 1992 the total nitrogen deposition on MDT from sources located within the calculation region (the Nile WHS excluded) amounted to 1084 kt N of which 660 kt N as NO_x and 424 kt N as NH_3. The map of deposition density of total nitrogen is given in Figure 3.3. Wet, dry and total depositions of oxidised, reduced and total nitrogen are presented in Figures B.3-B.10, Annex B.

As it is seen from Figure 3.3 total depositions mainly decrease from north to south. It is also evident from Table 3.3 which shows a meridianal view of the general pattern of total and wet depositions of total nitrogen.

TABLE 3.2: *Emissions of sulphur compounds, oxidised and reduced nitrogen in 1992. Unit=100 tonnes*

Notation	Country or region	Oxidised sulphur as SO_2	Oxidised nitrogen as NO_2	Reduced nitrogen as NH_3
alg	Algeria*	2400	490	270
egp	Egypt*	2500	500	400
lib	Libya*	1280	242	111
mor	Morocco*	320	60	35
tun	Tunisia	1120	208	146
isr	Israel*	1200	800	200
jor	Jordan*	300	50	100
leb	Lebanon	500	100	110
syr	Syria	1600	300	710
al	Albania	1201	299	295
ba	Bosnia and Herzegovina	4800	540	362
bg	Bulgaria	11198	2600	2299
hr	Croatia	1049	501	270
cyp	Cyprus	450	130	30
fr	France	12378	15991	6228
gr	Greece	5106	3058	778
it	Italy	15367	20616	3837
fym	Macedonia, FYR of	100	20	177
mal	Malta	0	0	0
pt	Portugal	3459	2480	929
ro	Romania	5595	4426	3707
sl	Slovenia	1880	510	269
es	Spain	23160	12568	3239
tr	Turkey	3820	1910	4758
yu	Yugoslavia	3960	489	991
gb@ie**		36615	28746	5083
cer**		45176	41847	10628
ner**		4920	11612	2781
eer**		119918	52778	30783
atl@bas**		5481	6042	0
med	The Mediterranean Sea	0	0	0
Total		316853	209913	79526

* Within the calculation area

** gb@ie = United Kingdom + Ireland
cer = Austria, Belgium, Germany, Luxembourg, Netherlands and Switzerland
ner = Denmark, Finland, Norway, Sweden and Iceland
eer = All countries located on the territory of former Soviet Union, Poland, Slovakia, Czech republic and Hungary
atl@bas=Atlantic Ocean and the Baltic Sea

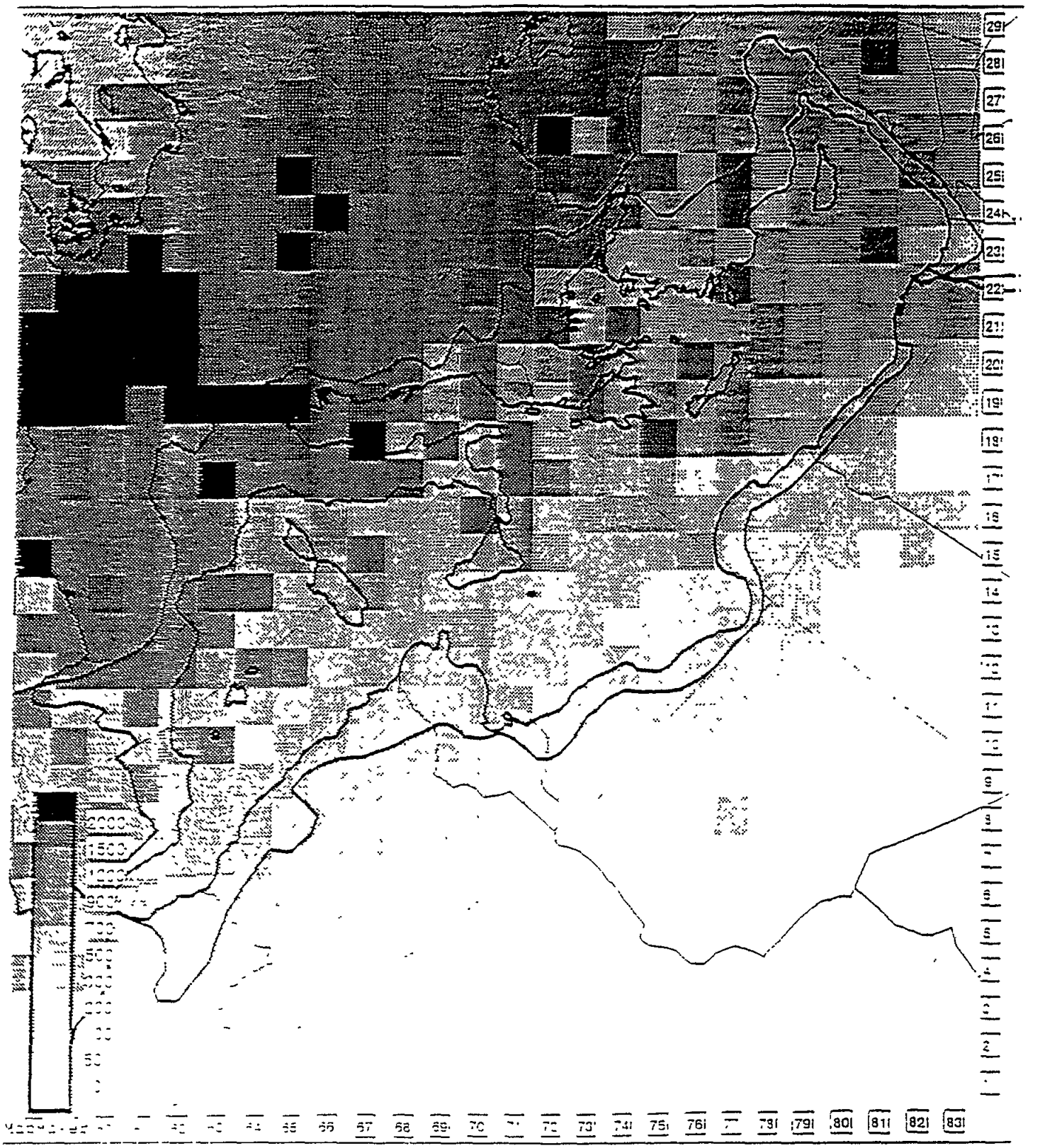

Figure 3.3 Distribution of the total nitrogen deposition on the Mediterranean in 1992.
Unit = mg N m^{-2} year

The reduction of depositions from north to south is resulted from the locations of main sources - to the north of MDT and by the prevailing transport direction from north-west to south-east and by the decrease of precipitation amount in the region considered from north to south. Table 3.3 also shows that the share of wet deposition of total deposition is depleted with the decrease of precipitation amount as approaching the south. Thereby the share of dry depositions is growing.

TABLE 3.3: *Three meridianal cross-sections of total and wet calculated depositions of total nitrogen in the Mediterranean within the limits of calculation region (mg N/m^2/year)**

Latitude	Depositions					
	0° E.Longitude		15° E.Longitude		30° E.Longitude	
	Total	Wet	Total	Wet	Total	Wet
	1026	766	1225	926	442	358
	1278	1051	1834	1625	702	581
	822	455	649	379	962	794
40°	567	379	438	188	367	294
	369	178	745	625	296	151
	405	359	639	581	383	325
	207	189	75	51	186	164
	105	86	123	113	189	166
	13	8	121	105	146	127
	12	8	13	0	186	5
30°	8	4	13	0	24	0
	6	3	7	0		
	1	0	4	0		
			1	0		
			1	0		
			1	0		
			1	0		
20°			1	0		

* Grid cells covering the Mediterranean Sea are shaded

Tables 3.4 and 3.5 give values of deposition of oxidised and reduced nitrogen on MDT and MDT countries. The main contribution to MDT pollution makes the MDT countries. The input of non-MDT countries is 25-27% of the MDT pollution by nitrogen compounds whereas their fraction in total emission of the calculation region is about 65% (i.e. $\sim^2/_3$ of the region emission give $\sim^1/_4$ of MDT pollution).

Integral values of the MDT pollution by NO_x and NH_x are approximately equal.

In the spatial distribution of depositions from individual countries-sources [*Erdman et al*, 1994] each country has its "impact zone". For instance Spain and France affect the western part of MDT, Italy - the central and eastern parts, Greece and Turkey - the eastern part.

Finally, deposition on MDT has seasonal variations with maximum in spring [*Erdman et al*, 1994].

TABLE 3.4: *Oxidised nitrogen deposition on the Mediterranean Sea and Mediterranean countries in 1992 in 100 kt N per year**

Rc/Em	alg	egp	lib	mor	tun	isr	jor	leb	syr	al	ba	bg	hr	cyp	fr
alg	65	0	2	2	6	0	0	0	0	0	0	1	0	0	43
egp	0	43	1	0	0	3	0	0	0	0	0	3	0	0	1
lib	0	2	16	0	2	0	0	0	0	1	1	6	1	0	7
mor	5	0	0	3	0	0	0	0	0	0	0	0	0	0	11
tun	3	0	3	0	11	0	0	0	0	0	0	0	0	0	12
isr	0	0	0	0	0	14	0	0	1	0	0	0	0	0	0
jor	0	0	0	0	0	32	3	1	2	0	0	0	0	1	1
leb	0	0	0	0	0	0	0	2	0	0	0	0	0	0	0
syr	0	1	0	0	0	10	0	5	29	0	0	3	0	4	1
al	0	0	0	0	0	0	0	0	0	8	2	3	1	0	2
ba	0	0	0	0	0	0	0	0	0	1	22	1	9	0	11
hr	0	0	0	0	0	0	0	0	0	0	10	1	20	0	25
fr	2	0	0	0	0	0	0	0	0	0	1	1	2	0	1589
gr	0	0	1	0	1	0	0	0	0	11	4	42	3	0	10
it	2	0	1	0	3	0	0	0	0	2	7	4	6	0	171
fym	0	0	0	0	0	0	0	0	0	3	1	13	1	0	1
sl	0	0	0	0	0	0	0	0	0	0	1	0	4	0	10
es	7	0	0	3	1	0	0	0	0	0	0	1	1	0	172
tr	0	2	0	0	0	2	0	1	6	4	5	79	3	3	16
yu	0	0	0	0	0	0	0	0	0	3	14	11	6	0	13
med	45	12	19	6	32	40	1	3	2	34	36	173	27	15	614

(continued)

Rc/Em	gr	it	fym	pt	ro	sl	es	tr	yu	gb@ie	cer	ner	eer	ind	sum
alg	2	54	0	15	1	0	124	1	0	12	30	1	4	4	379
egp	8	4	0	0	2	0	0	6	0	1	6	0	7	1	87
lib	26	27	0	0	4	1	2	4	1	6	15	0	9	4	138
mor	0	5	0	13	0	0	41	0	0	4	6	0	1	1	102
tun	1	32	0	1	1	1	13	0	0	4	9	1	4	0	96
isr	0	0	0	0	0	0	0	0	0	0	0	0	0	0	17
jor	1	1	0	0	0	0	0	2	0	0	2	0	2	0	50
leb	0	0	0	0	0	0	0	0	0	0	0	0	0	0	4
syr	2	2	0	0	4	0	0	17	0	2	4	1	13	1	100
al	5	41	0	0	3	1	0	1	1	2	11	0	8	0	89
ba	0	118	0	0	5	6	4	0	3	5	49	3	46	0	283
hr	0	163	0	0	3	11	3	0	1	8	65	3	43	0	359
fr	1	217	0	16	2	3	269	0	1	419	921	23	99	1	3667
gr	161	120	0	0	31	2	4	6	4	15	53	4	56	0	530
it	6	1547	0	4	4	13	61	1	3	25	229	6	70	1	2171
fym	7	17	0	0	6	0	0	0	2	1	5	0	11	0	69
sl	0	57	0	0	1	20	1	0	0	3	45	1	18	0	164
es	1	40	0	97	2	1	1112	0	1	82	171	4	33	1	1802
tr	53	68	0	0	79	3	4	232	5	25	79	12	286	2	973
yu	2	85	1	0	25	4	2	0	28	15	83	4	87	0	389
med	453	2360	1	54	149	23	940	130	23	170	668	30	472	6	6601

* See abbreviations in Table 3.2
Rc - countries-receivers
Em - countries-emitters

TABLE 3.5: *Reduced nitrogen deposition on the Mediterranean Sea and Mediterranean countries in 1992 in 100 kt N per year**

Rc/Em	alg	egp	lib	mor	tun	isr	jor	leb	syr	al	ba	bg	hr	cyp	fr
alg	95	0	2	4	14	0	0	0	0	0	1	2	1	0	34
egp	0	83	1	0	0	7	0	0	0	1	1	6	0	0	0
lib	0	4	20	0	2	0	0	0	0	3	1	11	1	0	3
mor	8	0	0	5	0	0	0	0	0	0	0	0	0	0	7
tun	4	0	2	0	30	0	0	0	0	0	0	1	0	0	9
isr	0	0	0	0	0	7	2	0	1	0	0	0	0	0	0
jor	0	1	0	0	0	17	13	4	12	0	0	1	0	1	1
leb	0	0	0	0	0	0	0	4	1	0	0	0	0	0	0
syr	0	1	0	0	0	3	3	16	201	0	0	4	0	2	0
al	0	0	0	0	0	0	0	0	0	40	3	6	1	0	1
ba	0	0	0	0	0	0	0	0	0	2	63	2	23	0	6
hr	0	0	0	0	0	0	0	0	0	1	25	4	38	0	16
fr	3	0	0	0	1	0	0	0	0	0	1	2	1	0	2507
gr	0	0	1	0	1	0	0	0	0	31	5	81	3	0	3
it	3	0	1	0	4	0	0	0	0	4	10	7	10	0	104
fym	0	0	0	0	0	0	0	0	0	15	2	31	1	0	0
sl	0	0	0	0	0	0	0	0	0	0	2	0	7	0	5
es	13	0	0	4	2	0	0	0	0	0	1	2	1	0	178
tr	0	5	1	0	0	2	0	1	58	9	6	155	3	2	7
yu	0	0	0	0	0	0	0	0	0	13	34	40	12	0	5
med	69	23	19	8	46	19	4	6	7	76	52	273	41	9	359

(continued)

Rc/Em	gr	it	fym	pt	ro	sl	es	tr	yu	gb@ie	cer	ner	eer	ind	sum
alg	1	26	0	10	3	0	77	3	1	1	9	0	5	8	297
egp	7	2	1	0	3	0	0	18	1	0	2	0	10	1	145
lib	14	11	1	0	7	1	2	15	3	2	5	0	10	8	125
mor	0	3	0	11	0	0	28	0	0	0	2	0	0	2	66
tun	1	15	0	1	1	1	7	0	0	0	4	0	3	1	80
isr	0	0	0	0	0	0	0	2	0	0	0	0	0	0	14
jor	1	0	0	0	0	0	0	11	0	0	1	0	4	0	67
leb	0	0	0	0	0	0	0	2	0	0	0	0	0	0	8
syr	2	1	0	0	3	0	0	184	1	0	1	0	12	1	437
al	7	16	5	0	4	0	0	2	10	0	4	0	9	0	109
ba	0	58	1	0	9	8	2	0	20	1	22	1	52	0	271
hr	0	86	0	0	6	23	1	1	9	2	43	1	63	0	320
fr	1	100	0	13	3	2	192	0	1	160	497	17	49	2	3552
gr	128	45	20	0	45	2	2	21	19	2	17	1	60	1	489
it	4	1072	2	3	5	16	34	2	14	2	148	3	48	1	1499
fym	16	7	25	0	10	0	0	1	20	0	2	0	19	0	149
sl	0	31	0	0	2	39	0	0	1	0	42	1	17	0	147
es	0	15	0	136	4	1	1104	1	2	17	67	3	19	2	1571
tr	42	24	7	0	125	3	2	1952	23	2	24	3	352	4	2812
yu	3	36	14	0	49	5	1	1	222	1	35	2	103	0	578
med	255	1044	27	36	202	23	468	379	98	17	248	9	409	11	4237

* See abbreviations in Table 3.2

Rc - countries-receivers

Em - countries-emitters

3.4.2 Deposition on the Mediterranean Sea watershed excluding the Nile basin

About 1097 kt of nitrogen compounds of which about 601 kt are oxidised and 496 kt are reduced nitrogen deposited on WSH1 in 1992. The pattern of deposition can be found in Figure.3.5 and Figures A.3-A.10 in Annex A. As it is seen from Figure 3.3 and Table 3.3, the northern coast is considerably more polluted than the southern one. The trend of reduction of total nitrogen pollution and the fraction of wet pollution in the direction from north to south remains. The reasons are discussed in section 3.4.1.

Of special note is a greater density of nitrogen deposition on the northern part of Algeria and Morocco (207 mg N/m^2/year for total deposition and 189 N/m^2/year for wet one) compared to the rest part of the African MDT coast (13 mg N/m^2/year for total deposition and zero for wet one) (see Table 3.3, distribution along 0^0 and 15^0 meridians). It is conditioned by enhanced precipitation over this region in which the orographic barrier (Atlas Mountains) changes the zonal character of precipitation distribution.

The spatial distribution of nitrogen deposition on WSH1 is well demonstrated by Table 3.6 containing deposition values on the MDT countries. The highest deposition falls on the MDT northern coast. In the deposition distribution on the MDT countries (Tables 4 and 5) "impact zones" are also detected. For example, Spain affects itself and France as well as Algeria and Morocco. Italy affects itself, France and the countries of the former Yugoslavia and to a less extent - countries of North Africa. Greece and Turkey inflict the impact on countries of the eastern part of the MDT region including countries of Middle East.

TABLE 3.6: *Depositions of nitrogen compounds on the Mediterranean Sea and Mediterranean countries in 1992 (MSC-E calculations), in kt N per year*

Notation	Country or region	NH_x	No_x	NTot
alg	Algeria*	30	40	70
egp	Egypt*	15	9	24
lib	Libya*	12	14	26
mor	Morocco*	7	10	17
tun	Tunisia	8	10	18
isr	Israel*	1	2	3
jor	Jordan*	7	5	12
leb	Lebanon	1	0	1
syr	Syria	44	10	54
al	Albania	11	9	20
ba	Bosnia and Herzegovina	27	28	55
hr	Croatia	32	36	68
fr	France	355	367	722
gr	Greece	49	53	102
it	Italy	150	217	367
fym	Macedonia, FYR of	15	7	22
sl	Slovenia	15	16	31
es	Spain	157	180	337
tr	Turkey	281	97	378
yu	Yugoslavia	58	39	97
med	Mediterranean Sea	424	660	1084

* Within the calculation area

The input of non-MDT countries to the pollution of the MDT countries is small for non-European countries-receivers: in most cases it is <10% of the total pollution of the country. For Spain and Italy this contribution is 15% or less. For France, Greece and Turkey this input is 40% and more for oxidised nitrogen and 20% and more for reduced nitrogen.

3.4.3 Deposition on the Nile watershed

The MSC-E model could not be applied to calculations of the nitrogen deposition on the Nile watershed because necessary input data were not available. Thus it was decided to use published results for this region obtained by global modelling. In doing so on the one hand the considerable uncertainties of results were realized but on the other hand, practically negligible runoff of airborne nitrogen from the Nile watershed reduces the importance of this area (see next chapter).

Data on wet deposition of oxidised nitrogen [*Gallardo&Rodhe*, 1995] and total deposition of reduced nitrogen [*Dentener&Crutzen*, 1994] obtained by the "Moguntia" model [*Zimmerman*, 1988] and cited in [*Rodhe et al*, 1995, fig.3 and 4] are used in the calculation of the nitrogen runoff. The deposition pattern shown in these figures is of a distinctive zonal character. The reduction of deposition density of oxidised and reduced nitrogen is observed from the equator to almost the Mediterranean coast.

Approximately 190 ± 60 kt N deposit on the Nile WSH as wet NO_x deposition and 370±140 kt N as NH_3 total deposition. Hence total deposition on the Nile WSH amounts to 560±200 kt of nitrogen compounds excluding NO_x dry deposition.

All integral estimates are summarized in Table 3.7 containing calculated values of nitrogen deposition on MDT and its watershed.

Table 3.7: Deposition of nitrogen compounds on the Mediterranean Sea and its watershed (kt N per year) in 1992

Receivers/Compounds	NO_x	NH_x	N Total
MED	660	424	1084
WSH1	601	496	1097
WSH2	190 ± 60*	370 ± 140	560 ± 200
WSH	790 ± 60	870 ± 140	1660 ± 200

MED The Mediterranean Sea
WSH The Mediterranean watershed
WSH1 The Mediterranean watershed without watershed of the Nile
WSH2 Watershed of the Nile
* Wet only

3.5 *Model-measurement comparison*

Plots of model-measurement comparison are presented in Figures B.11-B.19 of Annex B. The summary of the model-measurement comparison is presented in Table 3.8. The EMEP stations located in the Central and Southern Europe were considered in the current analysis. This subset of stations was split into four classes (according to [*EMEP/CCC*, report 3/95]) dedicated to the quality of the measurement data. The first class stations are of the highest quality. Some difficulties were connected with the aerosol concentrations of HNO_3&NO_3^- and NH_3&NH_4^+ where the information on the station data quality is absent. In this case the selection was based on the information about the quality of NO_2 in air and sulphate in aerosol data which seem to be most representative for the nitrogen aerosol compounds.

Comments on the Class of sites column

The notation "All" means that all stations are included into the consideration. The notation "1,2" or "3,4" means that only the stations of classes 1 and 2 or 3 and 4 were selected.

TABLE 3.8 *The summary of the results of model-measurement comparison*

Class of sites	Number of sites	Mean observed	Mean calculated	Correlation	Regression slope	Regression bias	Slope StDev
NO2 air concentrations, 0.1 μg N/m^3							
All	23	33.9	29.0	-0.197	-0.14	33.9	0.03
3,4	12	40.6	21.9	-0.283	-0.135	27.4	0.06
1,2	11	26.6	36.9	0.133	0.105	34.1	0.03
NO3+HNO3 air concentrations, 0.1 μg N/m^3							
All	6	82.2	110.0	0.881	1.084	21.0	0.07
Nitrates in precipitation, 0.1 mg N/l							
All	29	52.6	58.0	0.222	0.232	45.7	0.03
NH3+NH4 air concentrations, 0.1 μg N/m^3							
All	5	220.2	195.1	-0.427	-0.443	292.6	0.09
Ammonium in precipitation, 0.1 mg N/l							
All	26	71.3	52.4	0.491	0.437	21.2	0.04
3,4	18	73.8	51.5	0.521	0.7	1.0	0.36
1,2	8	65.9	54.3	0.5	0.221	39.686	0.1

Very small number of samples in Southern and Central Europe significantly reduce the precision of the analysis. Therefore only qualitative conclusions could be made. Some underestimation of the calculated NH_3 air concentrations versus measured ones has been found. This may be explained by the following reasons:

- re-emission of NH_3 is not included into the model,
- uncertainties in emission and meteorological data,
- natural emission could be of a considerable importance for Southern Europe [*Rodhe et al*, 1995], but it was not taken into the consideration during the calculations.

It should be noted that similar trend of the underestimation of NH_3 concentration in precipitation is demonstrated by several models (in particular by the acid model of EMEP/MSC-W) [*Tuovinen*, 1994, Fig. 5.4]. This probably means that either emission data or representation of physical and chemical features of ammonia compounds in the Southern Europe are to be improved.

4. MODELLING OF THE AIRBORNE NITROGEN RUNOFF

4.1 General ideas for calculation

The following approach was used in order to estimate the riverine input of airborne nitrogen to the Mediterranean Sea from its drainage basin: quantitative assessment of a part of atmospheric deposition nitrogen which is retained in terrestrial and freshwater ecosystems and calculating a part of nitrogen entered the Mediterranean Sea aquatorium by rivers as a difference between its deposition and retention. This approach represents the quantitative estimates of various links of nitrogen biogeochemical cycle and its different tropical chains [*Kuylenstierna and Chadwick*, 1992; *Bashkin et al*, 1995; *Gough et al*, 1995].

4.2 Model structure

The majority of existing experimental data on nitrogen transformation are related to specific soils, their main features, such as physical-chemical parameters, mechanical composition, relief position, water regime etc. as well as with land use/cover types and nitrogen uptake in terrestrial ecosystems depending upon the external input of this nutrient. So, these data have to be used in the structure of the model 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 tropical stage and residence time. For reducing an uncertainty of experimental results and expert estimates, the spatial and temporal resolution of the model has to be determined very correctly, i.e. minimal taxon level must be chosen in order to support by information all model calculations. Accounting to the available information on the Mediterranean Sea drainage basin, the resolution level was connected with LoLa-grid cells (1 grad.longitude x 1 grad.latitude), which were subdivided into elemental taxones in accordance with soils and land cover types of NASA data sets of the Mediterranean Sea drainage basin [*Rudolf and Schneider*, 1994; *Koster et al*, 1994; *DeFries and Townshend*, 1994].

Accordingly, the model structure consists of a nucleus and various data bases (DBs) (atmospheric precipitation input of nitrogen compounds; soil and land cover types; values of runoff coefficients; values of nitrogen uptake, immobilization and denitrification; retention in terrestrial and freshwater ecosystems etc.). The calculation algorithm is shown in Figure 4.1.

4.3 Calculations of the nitrogen runoff

4.3.1 Initial Information

The land cover types, types of soils and their properties (texture types and relief), and water regime are taken from NASA DB [*Rudolf and Schneider*, 1994; *Koster et al*, 1994; *DeFries and Townshend*, 1994]. C:N ratio of soils in Europe was taken from FAO-UNESCO maps, [*FAO-UNESCO*,1981] as well as a map of the World [*Kovda*, 1973].

4.3.2 Calculation algorithm

On the basis of above mentioned conceptual and technical approaches, the following algorithm has been applied to the assessment of values of nitrogen input to the Mediterranean Sea from its drainage basin [*Kuylenstierna and Chadwick*, 1992; *Bashkin et al.*, 1995]:

$$N_{leach} = 3\ (N_{td} - N_u)*(K_{tretl}*K_{tret2}*K_{tret3}*K_{tret4})*K_w,$$

where: N_{leach} - deposition nitrogen leaching into the rivers and then into the Mediterranean Sea; l - land cover types in every elemental soil taxon, accounting to soil texture and slopes; N_{td} - nitrogen deposition in every land cover type; N_u - nitrogen uptake rate in every terrestrial land cover type; K_{tret1}-K_{tret4} - coefficients of nitrogen retention in terrestrial ecosystems depending on soil features; K_w - coefficient of nitrogen retention in various water bodies (rivers, channels, lakes, swamps, ponds etc.) of every land cover type.

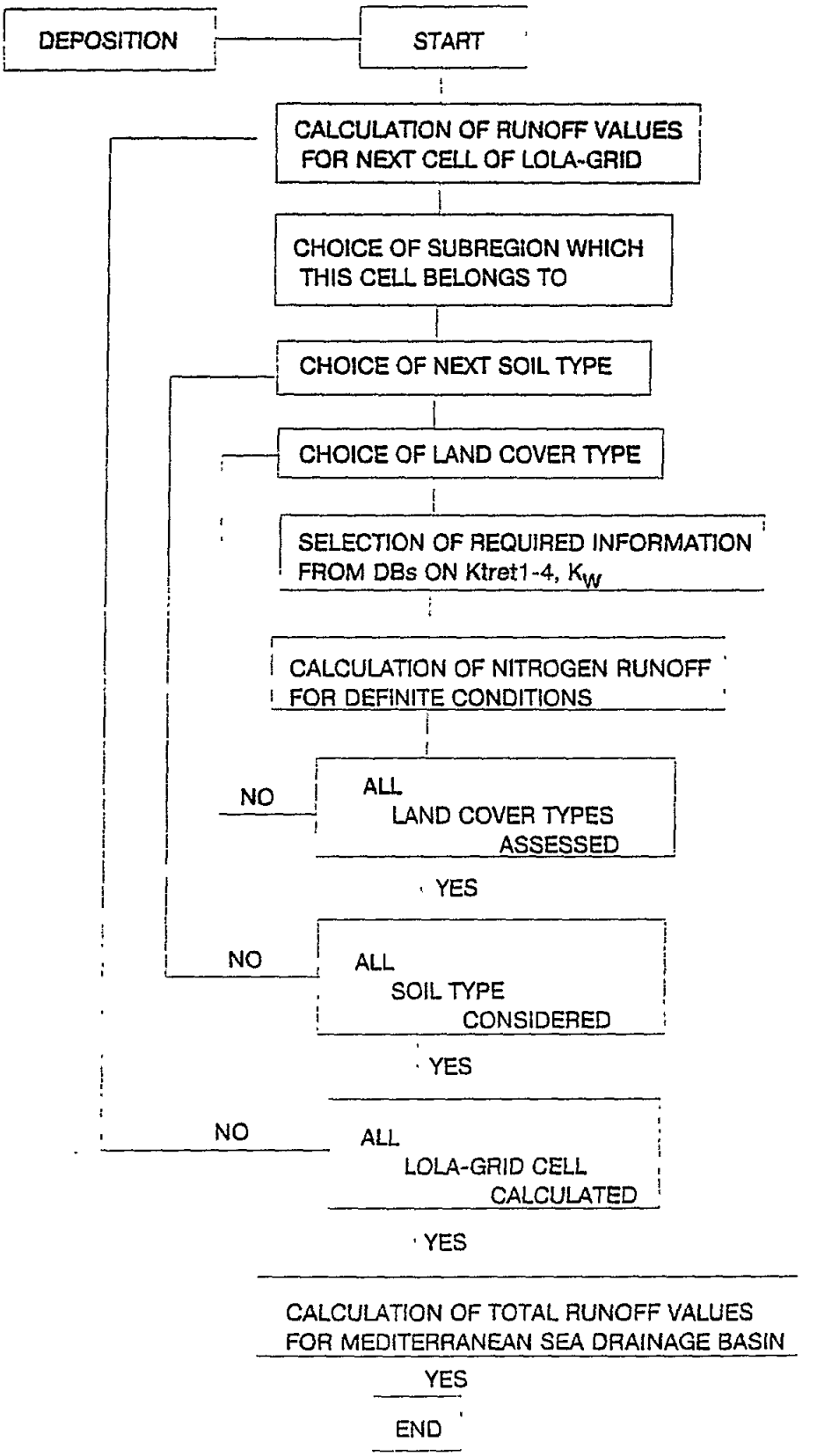

Figure 4.1 Algorithm for calculation of the riverine runoff of nitrogen

4.3.2.1 Nitrogen runoff from terrestrial ecosystems

Nitrogen as nitrate or ammonium is taken up by plants or used by the soil microfauna and bacteria. Nitrogen deposited as nitrate on to the soil could leach from the soil where deposition exceeds uptake and immobilization. Ammonium is less likely to leach as it is adsorbed by the cation exchange complex and may be held from leaching by certain "reaction" with clay minerals [*Kuylenstierna and Chadwick*, 1992).

The nitrogen removal from ecosystems is equal to the uptake and accumulation of nitrogen during the crop rotation period into the harvested parts of the plants. This uptake will be a function of plant species, site fertility and climatic variables as well as management regimes. The removal from the ecosystem will then depend on what part of the plant is removed: in many tree species, for example, over 90 per cent of the nitrogen in the standing crop is in the leaf material [*Kuylenstierna and Chadwick*, 1992].

In areas where deposition exceeds uptake and removal rates there is still often a very low rate of nitrate leaching which suggests another sink for nitrogen. Denitrification is one mechanism but this is unlikely to be high except in wet, waterlogged areas that are very scarce in the Mediterranean drainage basin. There is a very good evidence for nitrogen build up in soils under certain conditions. There is evidence that the accumulation rate of organic matter has increased in recent decades and it would seem to be temporarily immobilizing the increased nitrogen input in some ecosystems.

Likewise ion leaching, the nitrate leaching rate is greater in soils with good drainage properties. In poorly drained and waterlogged soils less nitrate is leached down through the soil profile and so leaves the soil either by lateral flow or by denitrification. There are difficulties in quantifying denitrification and immobilization rates since there is a great degree of spatial heterogeneity in the denitrification and also uncertainty in measurements at specific sites [*Kuylenstierna and Chadwick*, 1992].

So, taking into account the nitrogen deposition field (see Chapter 3) it would seem that there is a deposition rate below which there is little nitrate leaching and above which nitrate leaches at different rates. This could be explained by the fact that nitrogen does not leach until uptake rate is exceeded and that, after this point has been reached, the leaching rate then depends on the degree of immobilization in the soil.

In order to quantify the values of nitrogen runoff from terrestrial ecosystems, it is necessary to estimate the uptake and removal of nitrogen and also the rate of immobilization and denitrification and use these values to determine the degree to which nitrogen deposited in ecosystems will leach.

Uptake and removal

Natural communities which are not harvested or managed will have only small overall net nitrogen removal rates that may be ignored. The uptake rates and removal at the harvest may be estimated from the amount of biomass harvested. The method used here is not to access such data which exist but require a huge data gathering effort for the Mediterranean drainage basin; it is to approximate the nitrogen removal rates for various vegetation types by using values given in the literature. Nitrogen deposition itself will cause plant growth to increase and nitrogen uptake rates to increase. Such degree of accuracy is not included in the given simple estimation of uptake and removal rates.

On the basis of NASA DB [*Rudolf and Schneider*, 1994; *Koster et al*, 1994; *DeFries and Townshend*, 1994] on vegetation and land cover and from a literature survey [*Kuylenstierna and Chadwick*, 1992] it was decided that the following land cover categories which could represent the variation in the nitrogen uptake are water/freshwater, forests, various grassland, shrubs, deserts

deserts and bare ground. Table 4.1 summarizes nitrogen uptake rate (removal rates) of these main categories of land cover estimated from the data in the literature review. The estimates in Table 4.1 are approximate due to variation from site to site within a land cover/vegetation category [*Bashkin*, 1987; *Kuylenstierna and Chadwick*, 1989; *Chadwick and Kuylenstierna*, 1990; 1991; *Bashkin et al*, 1993].

TABLE 4.1: *Approximative nitrogen uptake rates for various land cover type*

N	Land cover category [*Rudolf and Schneider*, 1994]	N uptake rate (Nu), mg/m^2/yr
0	water / fresh water	500
1	broadleaf evergreen forest	300
2	broadleaf deciduous forest and woodland	600
3	mixed coniferous and broadleaf deciduous forest and woodland	550
4	coniferous forest and woodland	450
5	high latitude deciduous forest and woodland	400
6/8	wooded grassland	500
7/6	grassland	450
8/6	wooded and grassland	500
9/7	shrubs and bare ground	100
15/9	grassland	250
11/6	desert, bare ground	70
12/9	cultivated	1500
14	wooded grassland	400

Note: according to Rudolf and Schneider (1994) the double indices such as 6/8 mean the combination of various land cover types

One important aspect of nitrogen uptake is that in the Mediterranean Sea drainage basin it is seasonal. In temperate regions the nitrogen uptake takes place during the growing season. In this region nitrogen uptake is lowest during the periods of drought.

Immobilization of N in accumulated organic matter

Leaching of soil nitrogen, not subject to uptake by plants and other organisms, is controlled by factors affecting the rate of incorporation and release of nitrate from organic matter (mineralization and nitrification), denitrification and solute leaching.

Mineralization and nitrification rates

The C:N ratio in the A-horizon (upper horizon) of soil affects mineralization rates [*Kriebitzch*, 1978; *Bashkin*, 1987; *Kudeyarov*, 1989; *Gundersen and Rasmussen*, 1988; *Kuylenstierna and Chadwick*, 1992]. One can conclude that soils with C:N ratios of 15-20 have intermediate leaching rates, and C:N ratios <15, high leaching rates. The C:N ratios considered typical for different soil types are derived from data accompanying the FAO-UNESCO Soil Map of the World (1981) and the relationship is shown in Table 4.2. These values would be valid at the time when the sample was taken and may change over time due to the nitrogen deposition. It is not possible here to take into account these dynamic changes and it is assumed that the error introduced by not incorporating the dynamic aspects of the C:N ratio change are smaller than the error in extrapolating the C:N data available to a wider area.

TABLE 4.2: *The degree to which site factors affect the leaching of available nitrogen. Numbers refer to the amount of available nitrogen that will be leached (Kuylenstierna and Chadwick, 1992 and Bashkin et al, 1996)*

Factor	Category	Proportion of the leached nitrogen
	K_{tret1}	
C:N ratio	<15	0.90
(A-horizon)	15-20	0.50
	>20	0.10
	K_{tret2}	
Drainage	Well drained	0.90
(texture	Medium drained	0.75
indices)	Poorly drained	0.50
	K_{tret3}	
Runoff:	>0.80	0.90
precipitation	0.21-0.80	0.50
ratio	0.05-0.20	0.10
	<0.05	0.05
	K_{tret4}	
Relief	31-40	0.95
(slope),%	21-30	0.75
	<20	0.50

Soil drainage, dependent on soil texture, is a factor which affects the rate and degree of solute (nitrate) leaching [*Haynes et al.*, 1986; *Kuylenstierna and Chadwick*, 1992). Denitrification rates will be higher in the poorly drained soils and waterlogged soils. Only 40 per cent of nitrate was found to leach from poorly drained clay soils compared to well drained sandy soils [*Haynes et al.*, 1986; *Kudeyarov*, 1989]. About 50 per cent of the excess nitrogen are considered to leach from poorly drained soils as compared with >90 per cent from well drained soils (Table 4.2). Soil drainage properties typical of different soil types have been collated from the NASA Soil DB [*Koster et al*, 1994).

Soil moisture also affects the nitrate leaching rates with dry soils leaching little nitrate compared with moist soils. Irrigation has been found to increase the nitrate leaching by more than 50 per cent [*Haynes et al.*, 1986; *Kuylenstierna and Chadwick*, 1992). The precipitation to potential evapotranspiration ratio (P:E) can be used as an indicator of soil moisture conditions [*Wilsie*, 1962] as well as runoff to precipitation ratio (R:P). Four categories of the runoff to precipitation ratio(R:P) are used to describe the moisture status of soils [*Rudolf and Schneider*, 1994]:

i.	R:P >0,80	very moist soils;
ii.	R:P = 0.21-0.80	moderately moist soils;
iii.	R:P = 0.05-0.20	dry soils;
iiii.	R:P <0.05	very dry soils.

The nitrogen leaching for the four categories of R:P ratio is shown in Table 4.2. Less than 5 per cent of excess nitrate is considered to leach from very dry soils compared to 50 per cent from moderately moist soils and 90 per cent from moist soils.

Steep topography increases the rate of leaching. Generalized (area weighted) slopes have been taken as a substitute for topography according to Koster et al. (1994). Regions with the slopes more than 30-40% were assumed to increase the leaching up to 95 per cent, whereas under flatty relief only 50 per cent of nitrogen could leach (Table 4.2).

So, Table 4.2 shows the categories of the main factors together with estimates of the degree to which the different categories affect nitrate leaching. The combined effect of the factors on the nitrogen leaching is considered to be multiplicative. For example, sites with a C:N ratio of >20 are considered to leach only 10 per cent of the available nitrate in the soil, the rest being immobilized. If this site also is poorly drained, then only 50 per cent of these 10 per cent could be leached (i.e., only 5 per cent of the available nitrogen will leach).

The analogous approach has been applied to the European part of the Mediterranean Sea drainage basin by J.Kuylenstierna and M.Chadwick (1992) to show the acidification influence of nitrogen.

The computed results obtained for the N_{tret} values in different land cover and soil types were compared with those existed in literature. The atmospheric N input in Europe and North America has increased dramatically during the last decades due to emission of NO_x from combustion processes and of NH_3 from agricultural activities. The nitrogen deposition to forest ecosystems generally exceeded in the 1980`s 20 kg/ha/yr in the central part of Europe and even reached 100 kg/ha/yr in some impact areas [*Ivens et al*, 1990; *Hauhs et al*, 1989; *Gundersen and Bashkin*, 1994). Forest ecosystems may accumulate considerable amount of N in biomass and soil organic matter, but there is an increasing evidence that forest ecosystems may be overloaded with N from atmospheric deposition that leads to increasing leaching of nitrates and so called "nitrogen saturation" can occur. The nitrogen saturation can be defined as a situation in which the supply of inorganic nitrogen exceeds the nutritional demand of biota and it becomes apparent through increasing leaching of N below the rooting zone [*Aber et al*., 1989]. The review of nitrogen saturation in forest ecosystems presented by P.Gundersen and V.Bashkin (1994) shows that the values of nitrogen accumulation may be within the limits of 1-142 kg/ha/yr but the most statistically significant values for the forest ecosystems of central and north-western Europe were between 6-24 kg/ha/yr. In south Europe these values are a little bit lower.

At present a great attention is given to an assessment of wetlands as intermediate ecosystems between terrestrial and water ones. The nitrogen retention capacity of different types of wetlands has been studied in the project "WETLAND AND LAKES AS NITROGEN TRAPS" led by M.Jansson in the beginning of the 1990`s. The results have demonstrated that N removal in wetlands depends 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 a catchment may reduce the nitrogen transport by up to about 15%. In most cases, the result will probably be less. Furthermore, in addition to studies 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/yr) 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 amounts 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 values 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; *Hornung*, 1995] and for natural N leaching [*Kuylenstierna and Chadwick*, 1992] which vary within the limits of 3-25 and 2-8 kg/ha/yr, correspondingly.

Resuming this part, one can conclude that the values of retention of atmospheric deposition N in different ecosystems obtained experimentally can be quite significant, up to 30-40 kg/ha/yr in agricultural and managed forest ecosystems, but both computed and experimental reasonable values were within the limits of 3-15 kg/ha/yr. These values are similar to those used for calculation of nitrogen critical loads [*Grennfelt and Thornelof,* 1992; *Downing et al*, 1993, *Posch et al*, 1995). For example, the critical loads of permissible nitrogen leaching for the southern regions of Switzerland are within the limits of 4-7 kg/ha/yr [*Ackerman*, 1995, *Posch et al*, 1995]. Similar values were found by J.Kuylestierna (1996) for southern Europe in general.

4.3.2.2 Nitrogen retention in freshwater ecosystems

The coefficient K_w, reflecting the ratio between output of N from the water system and its input to a given system has been applied.

The values of K_w in various water sources have been calculated as following:

$$K_w = (1 - K_{wret1})*(1 - K_{wret2})*(1 - K_{wret3}),$$

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_{wret1}- K_{wret3} are dimensionless as a part of nitrogen entering the water body or per cent from the input whereas values of retention may be expressed as absolute values, for example in mg/m^2/yr or kg/km^2/yr, etc.

The minimum annual values of retention coefficients K_{wret} equal to 5% of the N input to a water body for temporary rivers were assumed to be like those in northern Africa. Whereas, the permanent retention of nitrogen in the sediments of lakes can be of 14-33% and even more of the external loading [*Ahlgren et al*, 1994; *Mosello et al*, 1995]. The experiments from other regions give similar results. For example, the analysis of the input/output mass budgets for lakes 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 varied for NH_4 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].

Depending on subregion, land cover 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 immobilization, N_{im}; N denitrification, N_{de}) during transport by water flows (rivers, channels, lakes etc), were assumed to alter within the limits of 0.05-0.80 accounting for different experimental results [*Jeffries and Semkin*, 1987; *Hinga et al*,1991; *Jansson et al*, 1994; *Fleischer et al*, 1994; *Ahlgren et al*, 1994; *Khrissanov and Osipov*, 1993; *Thies*, 1995 et al).

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 denitrification. The latters were assumed to be equal to 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 residence time less than 3 days. Average values (0.1-0.4) were used for the most frequent cases with sandy loam and loam bottom sediments and water residence time less than 1 year and the maximum values (>0.4) for peats and swamps ecosystems with water residence time more than 1 year [*Wyer*, *Hill*, 1984; *Bashkin*, 1987; *Jansson*, 1994 etc]. The type of bottom sediments were determined on the basis of land use and land cover data sets.

So, the calculation of K_{wret1} for land cover "open water" bodies such as fresh and salt lakes and swamps has been made for the resolution 1x1^o LoLa grid on the basis of the data presented in Table 4.3.

TABLE 4.3: *The values of coefficients of nitrogen retention in water bodies, K_{wret1}*

Water bodies	% of the area under water bodies from the total area of 1×1° LoLa grid						
	<5	5-10	11-20	21-30	31-40	41-50	>50
SLAK	0.05	0.12	0.18	0.25	0.30	0.45	0.70
SWMP	0.10	0.19	0.35	0.47	0.55	0.68	0.80
SLTW	0.04	0.07	0.12	0.17	0.20	0.30	0.50
ILAK	0.20	0.25	0.30	0.45	0.55	0.70	0.80

SLAK - lakes; SWMP - swamps;
SLTW - salt water bodies; ILAK - temporary water bodies

Furthermore, for rivers and channels the calculation of K_{wret2} values has been made for every 1 x 1° LoLa grid cell using the data set from Table 4.4 in accordance with the following equation:

$$K_{wret2} = (N_{de} * A)/N_{trun},$$

where: N_{de} - denitrification values, mg/m²/a; A - dimensionless coefficient, accounting for river density, water flow rate, type of an eutrophication process etc.; N_{trun} - nitrogen leaching from terrestrial ecosystems, equal to $(N_{td} - N_u) * (K_{tret1} * K_{tret2} * K_{tret3} * K_{tret4})$, mg/m²/a.

TABLE 4.4: *The values of parameters for calculating K_{wret2}*

Parameter	Mediterranean Sea drainage subbasins									
	I	II	III	IV	V	VI	VII	VIII	IX	X
N_{de}, mg/m²/yr	200	150	110	130	150	140	100	160	170	135
A	0.04	0.02	0.01	0.15	0.20	0.30	0	0.30	0.25	0.30

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} has been applied to the Nile, Po, Moulonya and Ebro rivers equal to 0.70, 0.25, 0.35 and 0.30 correspondingly.

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 one such tool and offers wide application within the UN ECE Convention on Long Range Transboundary Air Pollution and in

application within the UN ECE Convention on Long Range Transboundary Air Pollution and in particular, within the critical loads 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. A relatively simple structure of the model and ease of calibration provide the scope for regional application on large spatial scales [*Jenkins*, 1995].

However, there is no possibility to run this model and other similar models [*J.Kamari et al,* 1989, etc.] due to lack of satisfactory data bases for the whole drainage basin of the Mediterranean Sea and all calculations for the applied model has been computed using steady state mass-balance approaches.

4.3.2.3 Underground runoff of nitrogen

At present there is little information regarding the leaching of atmospheric deposition nitrogen into underground waters in the Mediterranean Sea drainage basin. The available information is confined to nitrogen content in various underground waters in different parts of this basin [*Moratti and Chiarelli*, 1988 etc.] and to the total underground runoff [*UNEP*, 1987]. Using these data, only very preliminary calculations of the underground runoff of atmospheric deposition nitrogen can be carried out for WSH1.

The following assumptions have been used:

- the total underground runoff that is equal to more than 107 Gm^3 (UNEP, 1987) can be applied to calculation without subdivision into separate stratas;
- nitrogen content in the total underground runoff was assumed to be equal to 3 mg/L in the whole Mediterranean Sea drainage basin;
- the share of deposition that could penetrate into underground water and form the underground runoff is limited to 5 - 10%.

Using given assumptions two different methods of calculation were applied:

- calculation of the underground deposition nitrogen runoff as a part of the surface runoff;
- calculation of a deposition part in the total sum of nitrogen leaching with the underground runoff.

4.4. Results and discussion

4.4.1 Computed values of the atmospheric deposition nitrogen runoff

Figure 4.2 and Tables C.1 and C.2 ANNEX C show the calculated absolute and relative values of the input of atmospheric deposition nitrogen with riverine runoff into the Mediterranean Sea aquatorium from its drainage basin. The calculations have been made by the above mentioned method for every 11^o LoLa grid cell and grouped for every EMEP grid cell only for WSH1.

The deposition fields are presented in Chapter 3 and Annex B.

Depending on the EMEP or LoLa cell location, the share of the atmospheric deposition nitrogen which enter the Mediterranean Sea aquatorium varied from 0 to 47%, for both WSH1 and WSH2. The minimum values were found for Asia Minor, northern African areas and the Nile watershed with low/absent runoff and very low precipitation or with intensive uptake of nitrogen in its biogeochemical turnover in subtropical and tropical regions belonging to the upper Nile flow.

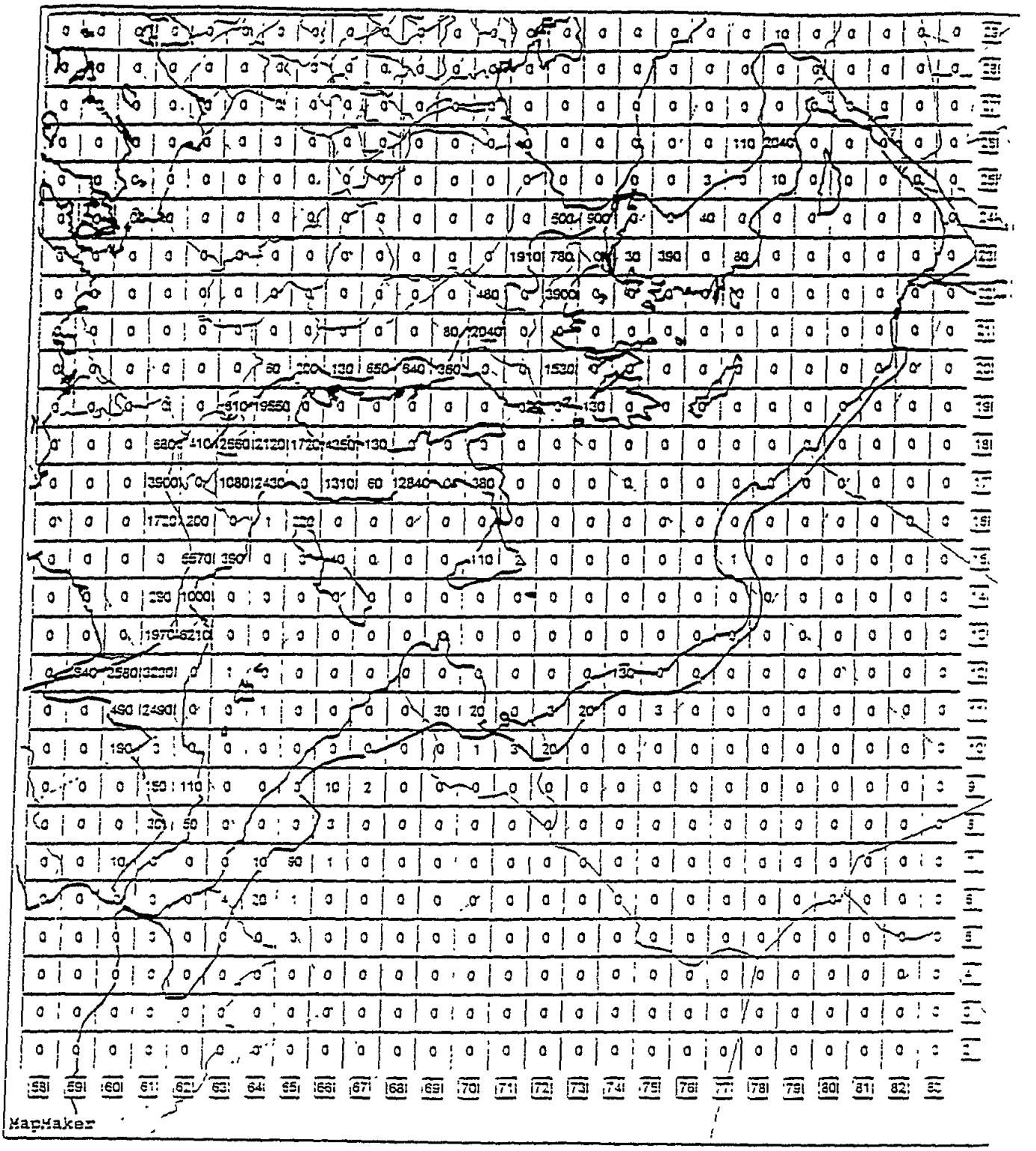

Figure 4.2: Map of riverine runoff of airborne nitrogen into the Mediterranean Sea in 1992.
Unit = t N/year

On the contrary, maximum values were found for mountain areas in the northern part of the drainage basin where the highest depositions coincide with the mountain relief. It should be noted that the maximum value of ratio between the airborne N runoff and direct deposition on the sea equal to 0.88 was found for the EMEP grid cell 68/17. It seems to be unrealistically high and apparently this value reflects an uncertainty connected with conversion from LoLa to EMEP cells. The value of 99 kt was calculated for the total WSH1 and the atmospheric deposition nitrogen runoff was almost negligible from the Nile watershed (WSH2). For the whole Mediterranean Sea drainage basin the average riverine nitrogen runoff (99kt) was about 6% of the total of atmospheric nitrogen deposited on the watershed area (1660± 200 kt) in 1992.

With regard to the underground nitrogen runoff directly into the Mediterranean Sea, these total values varied within the limits of 4-9 kt/yr (method 1, section 4.3.2.3) and 2-4 kt/yr (method 2) being on average 5.5 kt.

4.4.2 Comparison of computed and experimental results for various ecosystems

At present there is an agreement that the quantitative experimental assessment of the fraction of atmospheric deposition nitrogen leached from the ecosystems is very difficult to make due to many immobilization-mobilization processes in the internal nitrogen biogeochemical cycle complicating the final values. However, in small forested catchments where the N atmospheric deposition is the only source of this nutrient, one can estimate the ratio between input and output of the 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 [*Paces*, 1984; *Bashkin et al*, 1984; *Bashkin*, 1987; *Kallio and Kauppi*, 1990; *Moldan and Cherny*, 1994.].

Using this approach the nitrogen cycling was quantified for a considerable number of sites during the last decade. Some examples are given below just to show the atmospheric deposition nitrogen leaching parametrization in various regions of Europe.

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]. These experimental compilations have been continued during the project "Element Cycling and Output-Fluxes in Forest Ecosystems in Europe" - ECOFEE and the results from a preliminary analysis of the data on N cycling have been published [*Gundersen*, 1995]. It has been shown that the total inorganic N input ranged from 3 to 34 kg N/ha/yr in bulk precipitation and from 2 to 64 kg/ha/yr in throughfall.

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

Studying the biogeochemical fluxes in plantation forests on acid soils (Klosterhede, Denmark; Kootwijk and Harderwijk, the Netherlands; Hoglwald and Solling, Germany) T.Cummins and co-workers (1995) have shown that nitrogen inputs are higher than nitrate-N outputs in five of the soils, and it is likely that these sites are accumulating nitrogen. At one site, Hoglwald, the nitrogen output is considerably in excess of the current input, and this site may have reached saturation and be leaking nitrogen. The same situation has been observed in the ECOFEE project, where four sites leached more nitrogen than the input was. This might be due to a disruption of the N cycle [*Gundersen and Bashkin*, 1994; *Gundersen*, 1995]. The possible explanations could be the following. At one of these sites, the leaching of 45 kg N /ha/yr was connected with an invasion of insects resulted in defoliation of trees, decrease in the N uptake and increase in N leaching [*Pedersen*, 1993]. At the site with the leaching of 50 kg N /ha/yr, the tree species change from beech to spruce seemed to cause a release of the soil born N. Two Danish sites on calcareous soils with very high nitrification rates were leaching 75-85 kg N /ha/yr. However, one should bear in mind that differences from the input may be within the range of uncertainty of the flux estimates.

P.Gundersen (1995) has suggested to cluster the sites leaching nitrates in two groups: one group of sites with inputs of 15-25 kg N/ha/yr that leached almost all the N input, and another group of sites with inputs of 40-60 kg N /ha/yr that leached only about 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/yr were generated from high ammonium depositions. Deposition of oxidised nitrogen usually contributed only 10-15 kg N /ha/yr to the total input. High ammonium inputs could to some extent be retained by soil and vegetation, whereas the retention of nitrate seems to be low.

So, the analysis of the input-output data allows to draw the conclusion that at inputs above 10-15 kg N /ha/yr elevated nitrate leaching can be found in some forest ecosystems. New catchment scale data from the UN-ECE Integrated Cooperative Programme (ICP) on Integrated Monitoring (IM) also confirmed the input threshold at about 10 kg N /ha/yr where nitrate leaching started to increase [*EDC*, 1995].

However, the relationships between ecosystem characteristics and leaching are not conclusive. Ammonium dominated sites could often retain about 50% of the N input even at the high input levels, whereas nitrate dominated sites at moderate inputs had low N retention [*Gundersen*, 1995]. 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 to the leaching processes. A relative close relation between input and output of N observed from the experimental studies can lead to the conclusion that forest ecosystems may leach considerable amounts of nitrate even before they achieve the saturation stage in a biological meaning.

The relative close relation between input and output was also emphasized by 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 the 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 corresponding significant changes of soil solution composition and the riverine N runoff.

Under the study in the representative catchment in Crusne Hory Mountains (Czech Republic), which was originally wholly forested and after tree-died and clear-cut was reforested by birch and mountain-ash, mean annual fluxes of nitrogen were: deposition input - 16.2 kg/ha/yr, output with runoff - 5.5 kg/ha/yr that means 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 of Czech Republic [*Beudert and Kennel*, 1995]. The area of 0.69 km^2 ranges from 787 to 1292 m a.s.l., annual mean air temperature is 5.60C and mean annual precipitation amounts to 1319 mm (both in 945 m a.s.l.). Montane spruce and beech forest with silver fir are growing on rather poor brown earth soils derived from granite or gneiss and their periglacial solifluction covers. These solifluction covers together with underlying layers of intensively disintegrated rock detritus form a huge pore ground water reservoir which plays a major role in water cycling processes. Intensive measurements on element cycling are carried out at each one monitoring plot under spruce and beech stands at 815 m and 825 m a.s.l. The "Forellenbach" area is also a sub-catchment of the "Grobe Ohe" catchment (19.1 km^2) which is being monitored since 1979. 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 the deposition input of at least 14 kg/ha (assumable additional dry deposition uptake in the canopy is not considered) at least 7 kg/ha are retained in the system and the N retention was 52%. Simulation of the nitrogen budget is more complex, but preliminary results indicate that tree growth (7 to 9 kg/ha/yr) possibly is not the only sink responsible for reducing nitrogen flux from about 21 kg/ ha/yr (55% nitrate and 45% ammonia nitrogen) in deposition to 6 to 8 kg/ha/yr in catchment runoff of "Grobe Ohe". The suggestion that nitrate reduction to gaseous nitrogen or N_2O could be responsible for the remaining sink term needs further investigations, another possible sink of nitrogen could be long-term accumulation in humus matter due to lowering C/N-ratios.

From this comparison of experimental and computed results one can see that the nitrogen cycle is complex as well as there are many uncertainties in various features of ecosystems [*Posch et al*, 1993; *Forsius & Kleemola,* 1995 etc.]. From this it appeared that most ecosystems are accumulating nitrogen and that the leakages are quite different. 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 the 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 the N deposition [*Forsius & Kleemola*, 1995]. It was noted that calculating input-output balances are easier at the catchment-scale than at the plot-scale.

Water chemistry data on which all investigations in the AL:PE 1(Acidification of Mountain Lakes:Paleolimnology and Ecology) and AL:PE 2 (Remote Mountain Lakes as Indicators of Air Pollution and Climate Change) projects are based, are available for 28 lakes in UK, Italy, Norway, France (AL:PE 1) and in Norway, Ireland, Austria, Spain, Portugal, Poland, Slovakia, Slovenia and Russia (AL:PE 2). Nitrate and sulphate concentrations have different distribution patterns among the sites. Leaching of nitrogen was of a considerable importance to the acidification of lakes in the Italian Alps [*Mosello et al*, 1995].

On the basis of the given biogeochemical approaches the comparison of available catchment experimental and computed results of the N runoff has been carried out for various sites in the Mediterranean Sea drainage basin (Table 4.5). One can see that there are only a few such studies and the majority of them were carried out with lake watersheds [*Mosello et al*, 1995; *Camarero et al*, 1995]. Nevertheless, both experimental and computed values have the same order of magnitude.

Table 4.5: Comparative assessment of experimental and computed nitrogen runoff values from different ecosystem belonging to the Mediterranean Sea drainage basin (% from the N deposition input)

Computed N, runoff, mean		Experimental values		Reference
Country	kg/ha/yr	Lake*	kg/ha/yr	
Spain	2.19	Cimer	2.93	Mosello et al, 1995
Spain		Caldera	2.23	Mosello et al, 1995
Spain		Fero	17.86	Mosello et al, 1995
Spain		Aguillo	3.34	Mosello et al, 1995
Italy	2.39	Paione	2.23	Mosello et al, 1995
Italy		di Latte	13.39	Mosello et al, 1995
Italy		Lungo	8.92	Mosello et al, 1995

*: all lakes are situated in mountain regions

4.5 Uncertainty estimates

4.5.1 Nitrogen runoff from terrestrial ecosystems

The values of the nitrogen runoff from terrestrial ecosystems are subject to several sources of uncertainty. They are subdivided into four main sets: (1) uncertainty associated with the field measurements of the nitrogen retention and runoff in various subbasins of the Mediterranean 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 the conversion from 1x1° LoLa grid cell to 150x150 km^2 EMEP grid cells.

It has been shown that the main source of uncertainty in the given drainage basin is related to the insufficient data sets connected with multi-scale experimental studies of various links of nitrogen biogeochemical cycling.

Furthermore, the method described above considers only nitrogen leaching as the mechanism determining the runoff and calculated as a difference between deposition and retention.

The uptake rate implied by using the land cover/use could be improved by further data acquisition and this could make a considerable difference in the outcome. Perusal of the deposition rates leads to the conclusion that the deposition estimated by the EMEP West and East Synthesizing Centers could be an underestimate in certain parts of Europe, or at least small-scale spatial variations of deposition within EMEP squares are lost [*Kuylenstierna and Chadwick*, 1992; *Bajic and Duricic*, 1995; *Erisman et al*, 1995; *Bolloch and Guerzoni*, 1995; *Vidic*, 1995]. This means that in many regions the suggested uptake rates, even for tree species, are often higher than the deposition rates of nitrogen. The variation of nitrogen uptake over year is another important consideration. The uptake could be assigned to the part of the year when plants are growing. This could be compared with deposition over the months when plants are growing as these data exist. However, it is difficult to see how this could easily be incorporated into calculation of the nitrogen runoff.

The C:N ratio may be a rather rough parameter to be used for estimating the soil organic mineralization rates since there are other factors such as climate and vegetation type which may influence the rate of mineralization. However, the C:N ratio is correlated with both of these features. The knowledge of the immobilization rates may not warrant the complexity of the method suggested here in which case more rough estimation methods could be used.

The transmission from 1x1^{o} LoLa grid cells to 150x150 km^{2} EMEP ones is connected with losing some information because the NASA DBs are related to a center of a LoLa cell and during the conversion about 20% of existing and computed data could be lost.

4.5.2 Nitrogen retention in aquatic ecosystems

Uncertainties connected with the nitrogen retention in aquatic ecosystems are also subject to the similar sources as it was mentioned above and originate mainly from (1) uncertainty associated with various assumptions and simplifications and (2) uncertainty caused by spatial resolution and scale transition from LoLa to EMEP cells. Similar conclusions can be drawn regarding the nitrogen retention in aquatic ecosystems as this has been already done for terrestrial ecosystems. The majority of simplifications are connected with extrapolation of retention (including denitrification) data to the region where the experimental studies are scarce or absent, especially for the Nile watershed.

Therefore, using all of these estimates, the uncertainty of the computed values characterizing the atmospheric deposition riverine input of nitrogen into the Mediterranean Sea aquatorium from its drainage basin can be as large as 10-50%, being in average 30%.

5. COMPARISON OF DIRECT AND INDIRECT INPUTS OF AIRBORNE NITROGEN TO THE MEDITERRANEAN SEA

The final aim of the present study is related to a comparative assessment of values characterizing the nitrogen input to the Mediterranean Sea from its drainage basin (indirect) and those showing the nitrogen input to the sea surface (direct). Correspondingly, Table 5.1 shows that the atmospheric deposition nitrogen input into the Mediterranean Sea via rivers in 1992 was 99 kt/yr or about 6% of total nitrogen deposited on the whole drainage basin, which was about 1660 kt/yr in accordance with our results. As it was mentioned above, no atmospheric nitrogen runoff to the sea was found for the Nile watershed area (WSH2). If the whole Nile watershed area (about 2,870,000 sq.km) is excluded from the Mediterranean drainage basin, the riverine input of airborne nitrogen deposited on the watershed was 9.0% of the total N deposition (1097 kt/yr) on the Mediterranean watershed without the Nile basin.

With regard to the underground nitrogen runoff directly into the Mediterranean Sea, the preliminary total values varied within the limits of 2-9 kt/yr (6kt/yr on average) and the total sum of atmospheric deposition nitrogen entering the Mediterranean Sea with the riverine and underground runoff was about 105 kt (99 + 6 kt accordingly).

The MSC-E computed value of atmospheric nitrogen deposition on the Mediterranean Sea surface was 1084 kt in 1992. So, the part of airborne nitrogen entering the sea with the riverine runoff was 9.1% (or 9.7% including the underground runoff) from that deposited on the sea surface. The estimated total riverine pollution and background load of nitrogen was 800 kt (600-1000 kt). This means that in 1992 the part of atmospheric nitrogen, deposited on the Mediterranean Sea drainage basin and entered the sea with rivers was equal to 12.3% of the total riverine load from the watershed or 9.9% from total coastal (200 kt) and mean riverine loads.

With the total input of nitrogen into the Mediterranean Sea (including coastal and riverine inputs and direct deposition on the sea surface) being about 2084 kt (1884-2284 kt), the estimated part of the atmospheric deposition nitrogen entering the sea from the watershed with the rivers was 4,8% of the total N input (5.0% including the underground runoff).

Table 5.1: Summary data on depositions and inputs of nitrogen for the Mediterranean Sea and its watershed

Notations		Kt/yr	% of I_t	% of D_{WSH}	% of R_t (of R_r)	% of D_{MDT}
D_{MDT}	Atmospheric N deposition on the sea surface	1084	52			
D_{WSH}	Atmospheric N deposition on the watershed	1660 ± 200				
D_{WSH1}	- without the Nile basin	1097				
D_{WSH2}	- on the Nile basin	560 ± 200				
R_t	Total N runoff from the watershed	1000 ± 200	48			
R_r	- with rivers	800 ± 200	38.4			
R_c	- coastal runoff	200	9.6			
$I_t = R_t + D_{MDT}$	Total N input to the sea	2084				
R_{at}	Airborne N runoff from the watershed	105 ± 4	5.0	6.3	10.5	9.7
R_{ar}	- riverine runoff	99	4.8	6.0	(12.3)	9.1
R_{au}	- underground runoff	6 ± 4				
$D_{MDT} + R_{at}$	Total input of airborne N to the sea	1189	57			

As it was indicated in section 4.4.1 the input of the airborne nitrogen from the watershed in comparison with direct deposition on the sea surface is more important in the northern Mediterranean then in the southern and that should be taken into account in developing the airborne pollution control strategies.

For the Adriatic Sea the ratio of the airborne N input from the watershed to the direct deposition to the sea is about 25% (10% for the whole Mediterranean Sea and about 40% for the Baltic Sea), the shares of the airborne N runoffs in the total N inputs are consequently 10,5 and 12% for these seas. These ratios have been received from the following estimates: (i) direct depositions of nitrogen to the Adriatic, Mediterranean and Baltic seas in kt/yr - 122.4, 1084 and 300; (2) runoffs of airborne N to the seas - 30, 105 and 120 kt/yr, and (3) the riverine inputs - 182, 1000 and 650-850 kt/yr correspondingly. These estimates allow also to conclude that the runoffs of airborne N from the watersheds to the seas constitute correspondinly 9.8, 5 and 10.4-12.6% of the total N inputs to the seas. Thus the total shares of airborne N (direct depositions plus runoffs) from the total inputs would be 50, 57 and 36.5-44.2%.

6. CONCLUSIONS

(a) Calculations of the deposition of airborne anthropogenic nitrogen compounds on the Mediterranean Sea and its watershed (excluding the Nile watershed) for 1992 were carried out using MSC-E model. Estimates of the nitrogen deposition on the Nile watershed were taken from published results of global modelling.

(b) The computed nitrogen deposition amounts to 1097 kt of N for the Mediterranean watershed (excluding the Nile watershed) and to 1084 kt of N for the Mediterranean Sea surface. On the basis of published data the nitrogen deposition on the Nile watershed was estimated as much as 560 ± 200 kt of N (the computed value was 574 kt of N).

(c) Calculations of the atmospheric deposition nitrogen input to the Mediterranean Sea with the rivers from the drainage basin was carried out for 1992. The assessment was made on the basis of a non-linear steady state mass-balance model of ISSP RAS using values of the nitrogen runoff and retention in various terrestrial and freshwater ecosystems.

(d) The computed values of the atmospheric deposition nitrogen runoff to the sea was equal to 99 kt/yr or about 6% of the total N deposited on the Mediterranean Sea drainage basin which amounted to about 1660 kt in 1992.

(e) With regard to the underground nitrogen runoff directly into the Mediterranean Sea, these total values varied within the limits of 2-9 kt/yr being very preliminary due to insufficient data.

(f) The part of atmospheric deposition nitrogen entering the sea with the riverine runoff was 9.1% (9.7% including underground runoff) of that deposited on the Mediterranean Sea surface.

(g) Contribution of the airborne nitrogen deposited on the drainage basin area and entered the sea via rivers was 12.3% of the total N riverine input of 800 kt/yr (600-1000 kt/yr) as calculated for 1992 and about 6,3% of atmospheric N deposited on the watershed finally entered the sea through runoffs.

(h) With the total input of nitrogen into the Mediterranean Sea (including coastal and riverine inputs and direct deposition on the sea surface) being about 2084 kt (1884-2284 kt), the estimated part of atmospheric deposition nitrogen entering the sea from the watershed with the rivers was 4.8% (or 5% including the underground runoffs). Thus in the total N load to the Mediterranean Sea about 57% are of airborne origin (52% due to the direct deposition on the sea and 5% from the watershed). These values could be taken into consideration in different models and scenarios of nitrogen emission reduction in the Mediterranean Sea drainage basin.

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ANNEX A

Description of the MSC-E acid model used for calculations of depositions on the Mediterranean

The MSC-E modelling technology includes several models based on the same key units: advection scheme, vertical profile description, meteorology and emission data preprocessing, 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 chemistry 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 the subjects of changes and improvements. The previous version (Version 1992) was applied e.g. for preparing the report by *Erdman et al* (1994). The description of the model version and results of the model run were given in this report. For the present study the recent version of the model, Version-96 is used.

The description of main units of the Version-92 can be found also 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].

General comments

The model is ment for long-term (from 1 month to decades) calculations of airborne pollution transport using actual meteorological and emission data. Calculations can be made for a great number of sources, their groups or for single sources and episodes. Provision is made for operation following the emitter-receptor pattern with a number of point or area sources up to 200 and arbitrary number of receptors within the limits of the grid.

Model has the spatial resolution of 150 km at 60^0N for the EMEP grid (39 × 37 grid cells) (Figure 3.1).

The maximum grid contains 120 × 120 square cells and covers Northern Hemisphere to the north of 15^0. Maximum gridsize is 150 × 150 km^2. Grid cell size could be reduced down to 50 km × 50 km and then down to 25 km × 25 km. Thus, it could cover the EMEP area with the resolution 50 km × 50 km in case of the corresponding resolution of the emission data. The time step is changed automatically. Values of resolution are referred to 60^0 .

Geographical information includes geographical co-ordinates, surface type (sea, land, large fresh water basins, coastal line) and the height above the sea level. Sources are characterized by their location in grid cells. Thus 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 for current calculations (1987-1991) is prepared by the Russian Hydrometcentre and includes:

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

Averaging interval is 6 hours, spatial resolution used - 150×150 km^2. The data are prepared on the basis of actual network meteorological data processed with a special objective analysis [*Shapiro* (1981)]. In areas where the observational information is not available (oceans, deserts, etc.) a combination of extrapolation/interpolation algorithms with climatology data is used.

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

Emission data are separated into high (>100m) and low (<100m) emissions (not available in the previous model version). Annual emission and coefficients of temporal irregularities are specified as far as it is required.

For the regional calculations it is envisaged to perform preliminary calculations of concentrations for 3-14 days before the calculated time interval. If the model is used for calculations of an episode this stage is excluded.

If the concentration in a cell at some time is smaller than prescribed level, this cell is ignored and its pollution amount is added to the inattributable deposition which is uniformly distributed with the grid. Sums of the unattributable deposition from each emitter and for each receptor are incorporated into budget matrices. The amount of pollution gone outside the grid is also stored and incorporated into budget matrices.

The model advection scheme

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

Let $\vec{r}(t)$ 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 (Figure A. 1):

(A. 1) $$\vec{r}(t+\Delta t) = \vec{r}(t) + \vec{V}(t)\Delta t$$

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

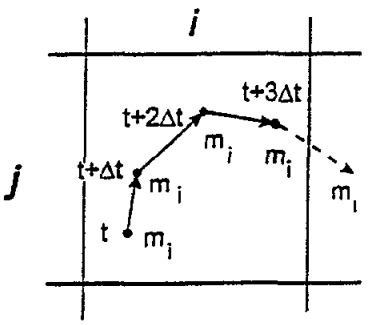

Figure A. 1 *Trajectory steps*

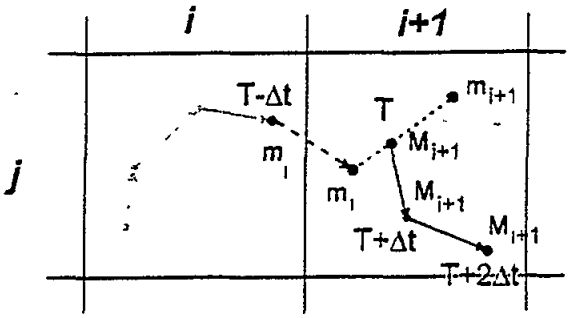

Figure A. 2 *Eulerian step*

In case of some mass input to a cell at moment *T* (either because of the advection, or because of emission) the Eulerian step is applied with the conservation of mass in a cell and its first moment (Figure A. 2):

(A. 2) $$M_{i+1} = \sum m_i$$

(A. 3) $$\vec{P}_{M_{i+1}} = \sum \vec{P}_{m_i}$$

where $\vec{P}_m = m\vec{r}_m$ 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 coming 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 the Eulerian grid or by averaging over considerable time periods not less than one week.

Vertical distribution

In the latest version of the model the truncated Gaussian profile of pollution distribution along the vertical *C(Z)* is used:

(A. 4) $$C(Z) = M\varphi\ (Z) / P(\mu / \sigma)$$

where:

(A. 5) $$\varphi(Z) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left(\frac{-(Z-\mu)^2}{2\sigma^2}\right), \qquad P(\mu / \sigma) = \int_0^{\infty} \varphi\ (Z) dZ = \int_{-\infty}^{\mu/\sigma} \varphi_0(\lambda) d\lambda$$

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

Here $\varphi\ (Z)$ describes the shape of the vertical concentration profile, $P(\mu / \sigma)$ is 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:

(A. 6) $$Z_0 = \frac{1}{M} \int_0^{\infty} ZC(Z) dZ\ , \qquad M = \int_0^{\infty} C(Z) dZ$$

The boundary condition near the surface is:

(A. 7) $$V_d\ C(0) = -K_z \frac{\partial\ C(Z)}{\partial\ Z}\bigg|_{Z \to 0}$$

where V_d is 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:

(A. 8) $$d\sigma\ (t) / dt = K_z(\sigma.t) / \sigma$$

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

(A. 9) $$Z_0(t+\Delta t) - Z_0(t) = \frac{d\sigma}{dt}\left(\frac{\varphi_0(y)}{P(y)} + y\right)\Delta t$$

where $y = \mu / \sigma$ . .

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

(A. 10) $$M = \sum M_i \ , \qquad Z_0 M = \sum Z_{0i} M_i$$

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 a 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 the weighted advection velocity.

Dry and wet deposition

Dry deposition is calculated in a common way:

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

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 um we used data available in the literature [*Sehmel* (1980)] and the 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 in the table:

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 $T<-2^oC$ and any P and F
   $K=1$ at $T>-2^oC$ $P=0$ and $F=0$
   $K=2$ at $T\geq 2^oC$ $F=1$ or 1mm/h$>P>0$
   $K=3$ at $T\geq -2^oC$ $F=2$ or $P\geq 1$mm/h

2. $K=1$ at $T<-2^oC$ and any P and F
   $K=1$ at $T>-2^oC$ $F=0$ and $P=0$
   $K=0.5$ at $T\geq -2^oC$ $F=1$or 1mm/h$>P>0$
   $K=0$ at $T\geq -2^oC$ $F=2$ or $P\geq 1$mm/h

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

The last version of model 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

(A. 11) $$z_0 \propto \frac{u_*^2}{g}$$

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 a few data suitable for the parametrisation of the process. Some of them have been presented in the Ukrainian report for EMEP [*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 by *Engelmann* (1963) and *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 a precipitation 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 um (Brownian particles) aerodynamic bending around drops and flakes is not important. Drops and flakes are in permanent contact with renewed portions of gas, therefore β characterizes the effective cross-section and equals to about 0.25 for rain and 1.0 - for snow. It is assumed that in clouds β≅1 always. For SO_2 and NO_x Λ depends on saturation of precipitation elements [Galperin (1989)].

Actually the air concentration is a function 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 when it starts precipitating [Afinogenova et al (1990)].

The usage of a full 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 by Galperin (1989) and a refining semiempirical formula:

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

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

The dependence of the relative surface of precipitation elements S_0/v_p on precipitation intensity and on its type is sharply expressed. If we consider data presented by Kelkar (1959), then for a rain:

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

where D - maximum effective diameter of a drop (D ≅ 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 the model version 1996 (M-96) is shown in Figure A.3. It differs from that of M-92 by the consideration of an ozone creation potential as a result of VOC emission and by the consideration of a strong acid formation in case of lack of ammonia. The scheme includes the following equations partly described by Pressman et al (1991):

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

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

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

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

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

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

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

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

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

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

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

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

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

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

Here: [C] is concentration (mass in a calculated grid) of substance C; $K_1,\ldots,K_{14}$ are reaction constants (see Table A.1); d(C) is concentration increment at a unit time step; q(C) is emission of substances given in brackets ($NO_x = NO + NO_2, SO_x = SO_2 + SO_4^{2-}$); VOC are volatile organic compounds, a and b - fractions of NO and SO_2 in the emission; $g(O_3)$ is background rate of tropospheric ozone formation; $(NH_4)_{1.5}SO_4$ is a mixture in equal fractions of $NH_4HSO_4 + (NH_4)_2SO_4$; SO_4^{2-} are all substances containing ion SO_4^{2-}; R are free radicals (ozone creation potential).

Dependence K_{11} (*T,H*) is taken from the work of *Finlayson-Pitts and Pitts* (1990), K_{14} are calculated on the data basis taken from [*Simpson* (1992)].

Notations: *T* - temperature in K°, *S* - relative insolation, *H* - relative humidity in %.

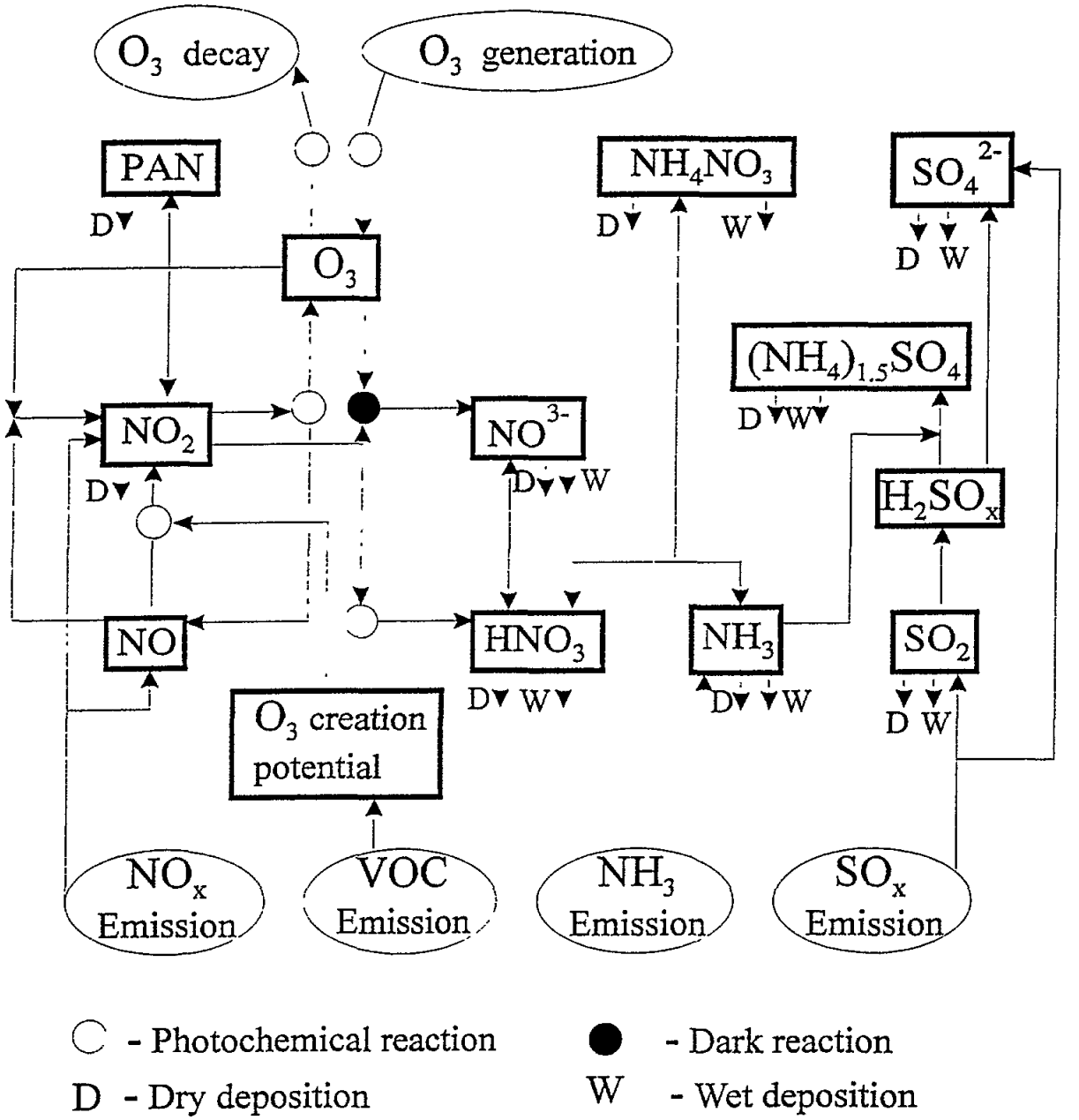

Figure A.3: *The chemical scheme of the Model*

Table A.1 *Coefficients and parameters of the chemical transformation scheme*

Coefficient or parameter	Unit	Value or formulae
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}	$28.6\times10^{17}\exp(-12530/T)$
K_7	$m^3/(\mu mole^{-1}h^{-1})$	$5.625\times10^{-4}\times(T-228.7)$
K_8	h^{-1}	0.015
K_9	h^{-1}	0.06
K_{10}	h^{-1}	$138500\exp(-4517/T)$
K_{11}	$\mu mole^2m^{-4}h^{-2}$)	$1.12\times10^{34}[\exp(-24220/T)]\times(298/T)^{6\,1}$ [$\times(100-H)/38$ if $H>62$; $\times1$ if $H<62$]
K_{12}	h^{-1}	0.03
K_{13}	h^{-1}	$0.005+0.2S$
K_{14}	relative units	
$g(O_3)$	$\mu molem^{-3}h^{-1}$)	$0.044S^2+0.21S+0.03$
a,b	relative units	0.95

Many reactions depend on the 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 vary proportionally to the following 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 the 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 $0\le S\le1.2$). It is supposed that the radiation decreases 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.

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ANNEX B

NITROGEN EMISSION MAPS.

NITROGEN DEPOSITION MAPS.

PLOTS OF MODEL-MEASUREMENT COMPARISON

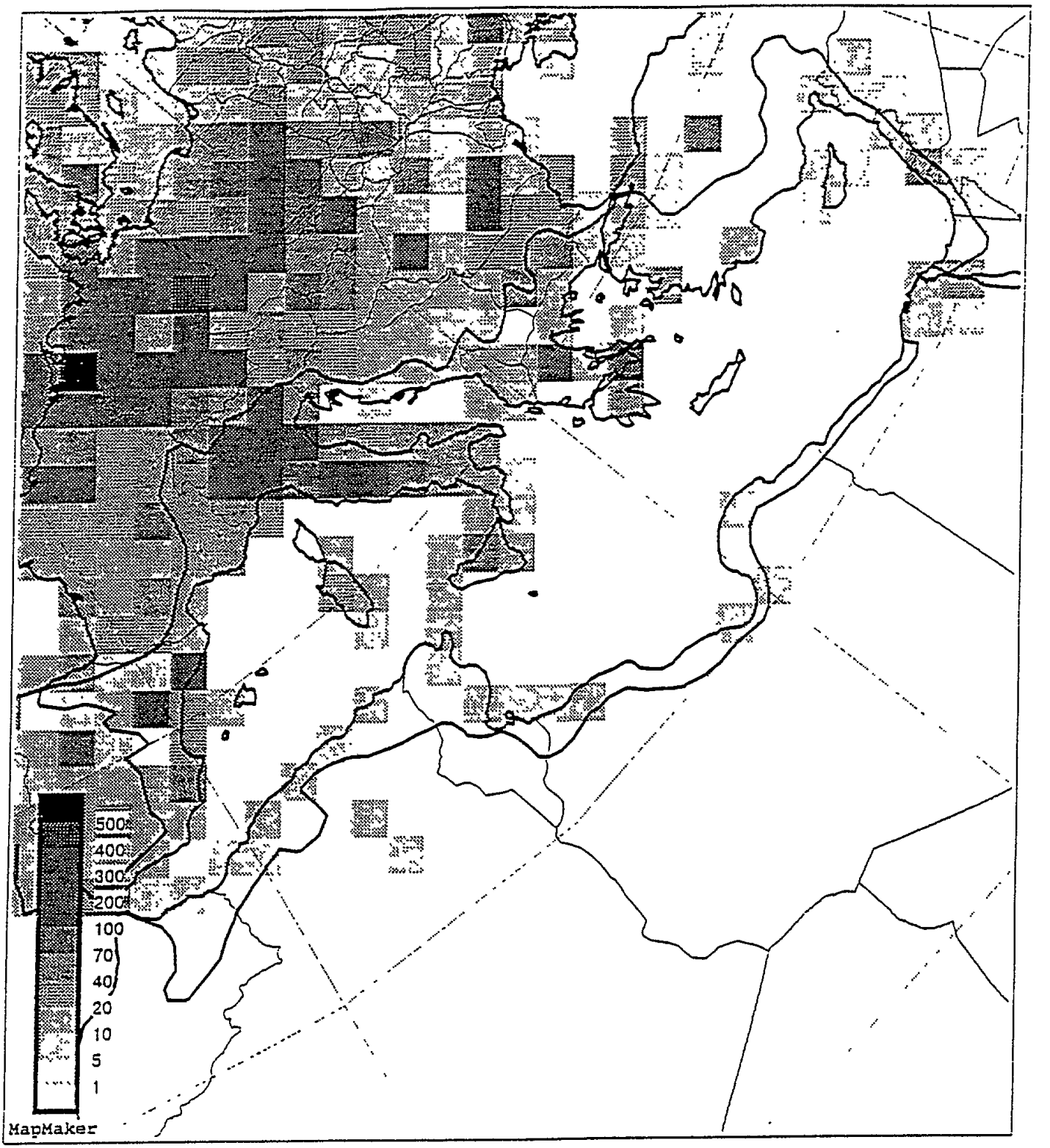

Fig.B1: Oxidised nitrogen emission intensity in grid squares of the calculation region in 1992
Unit = 1000 t NO_2/year

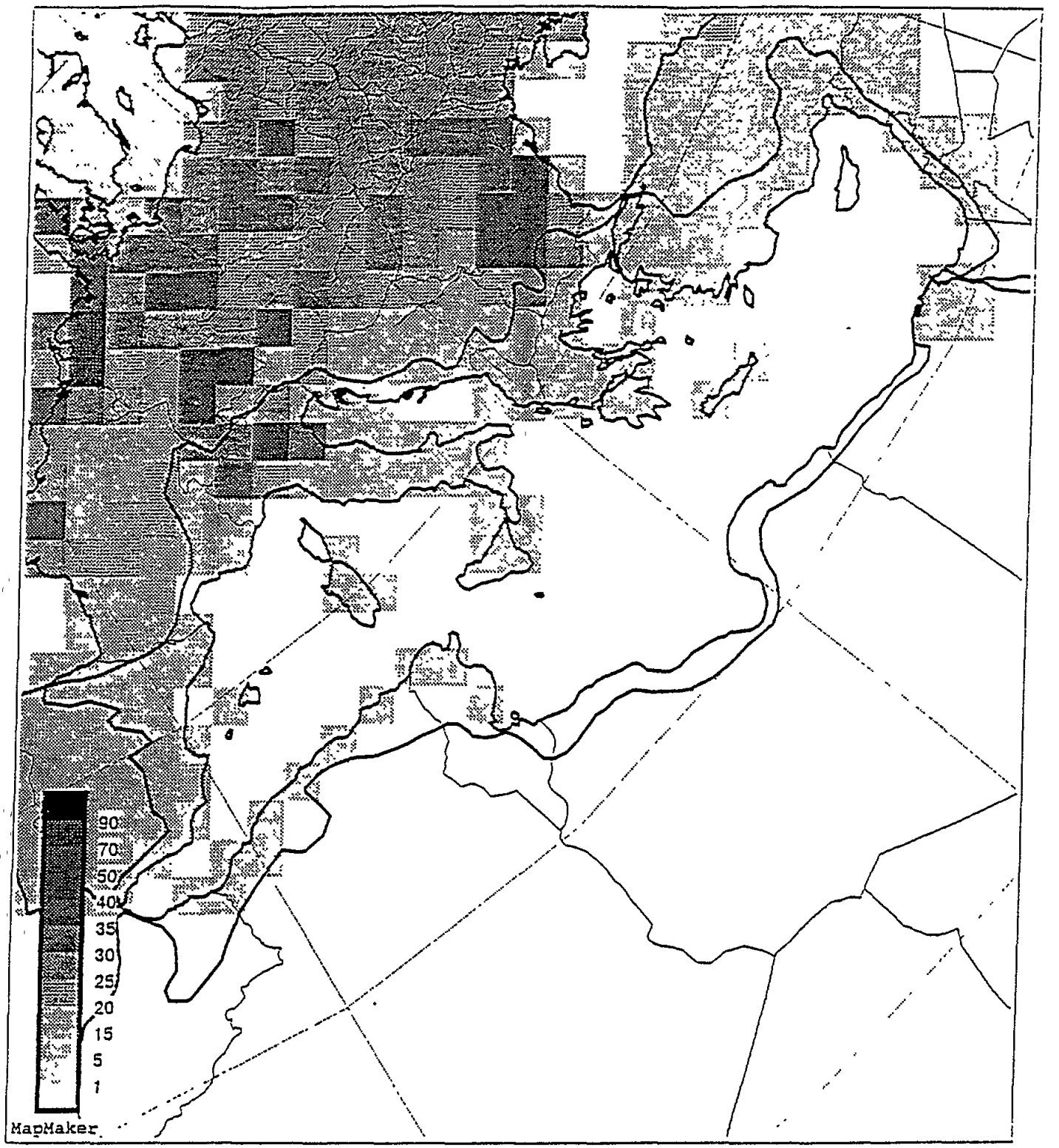

Fig.B2: Reduced nitrogen emission intensity in grid squares of the calculation region in 1992
Unit = 1000 t NH_3/year

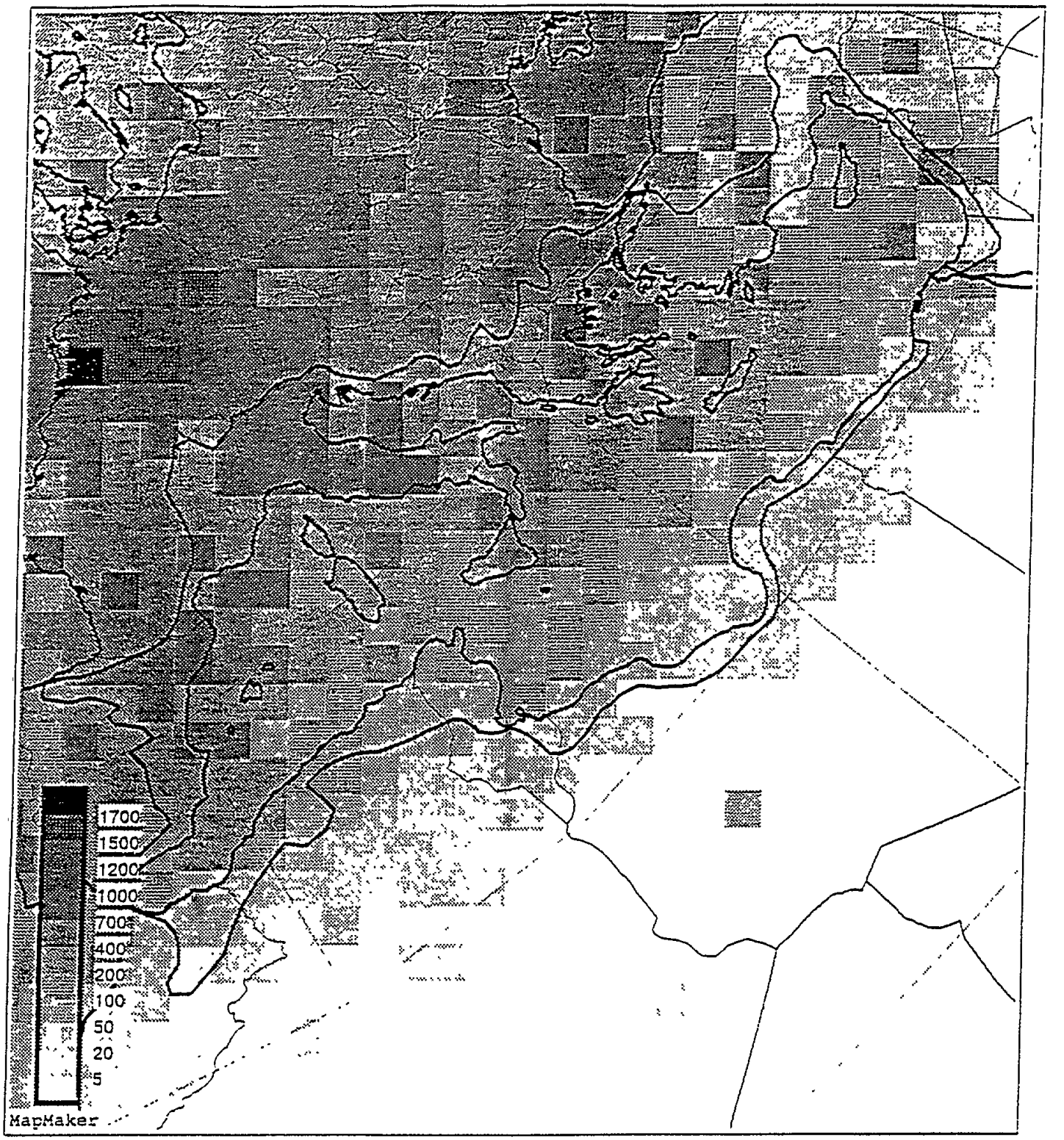

Fig.B3· Density of total deposition of oxidised nitrogen on the Mediterranean in 1992
Unit = 1 mg N/m²/year

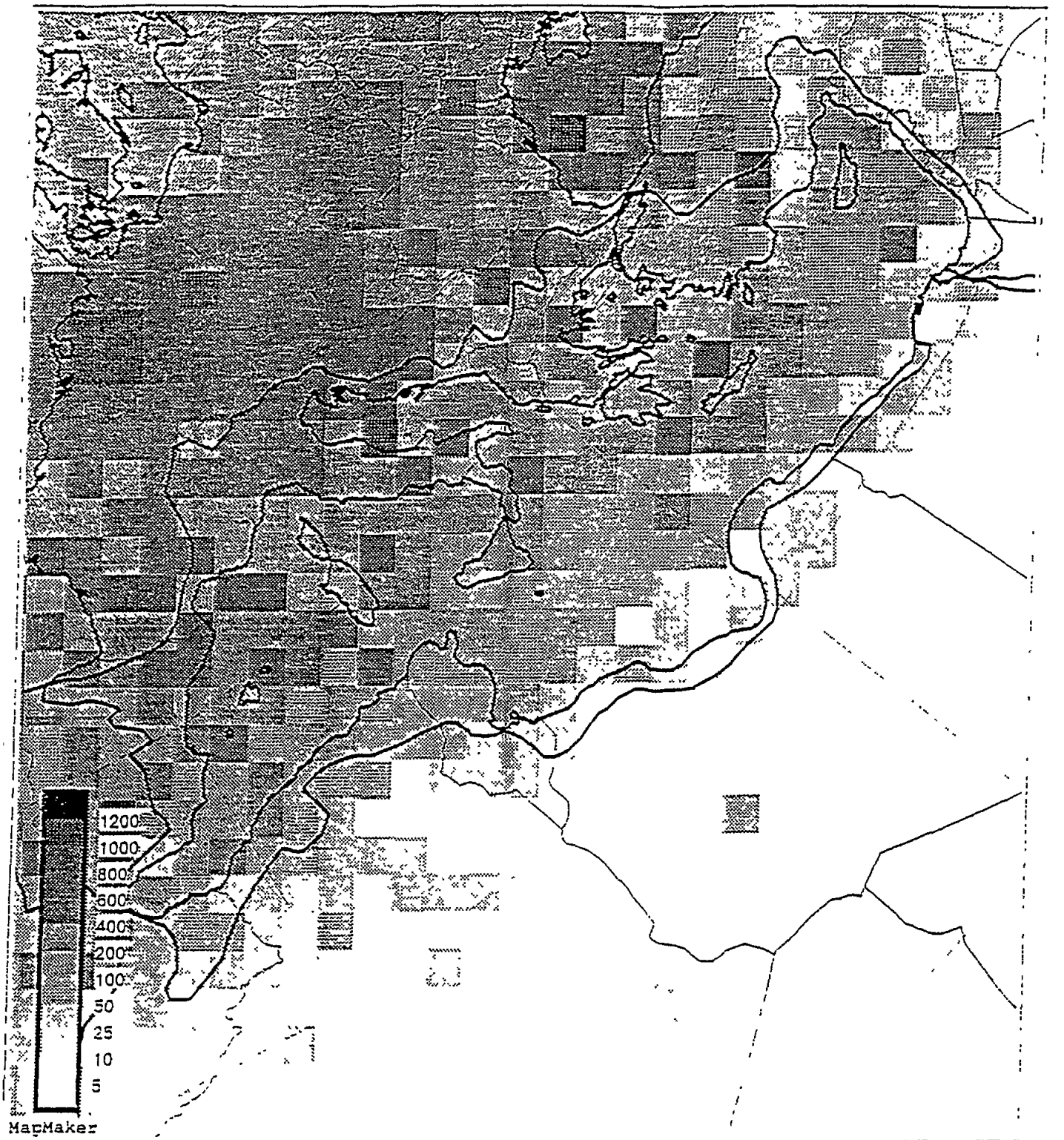

Fig.B4 Density of wet deposition of oxidised nitrogen on the Mediterranean in 1992
Unit = 1 mg N/m^2/year

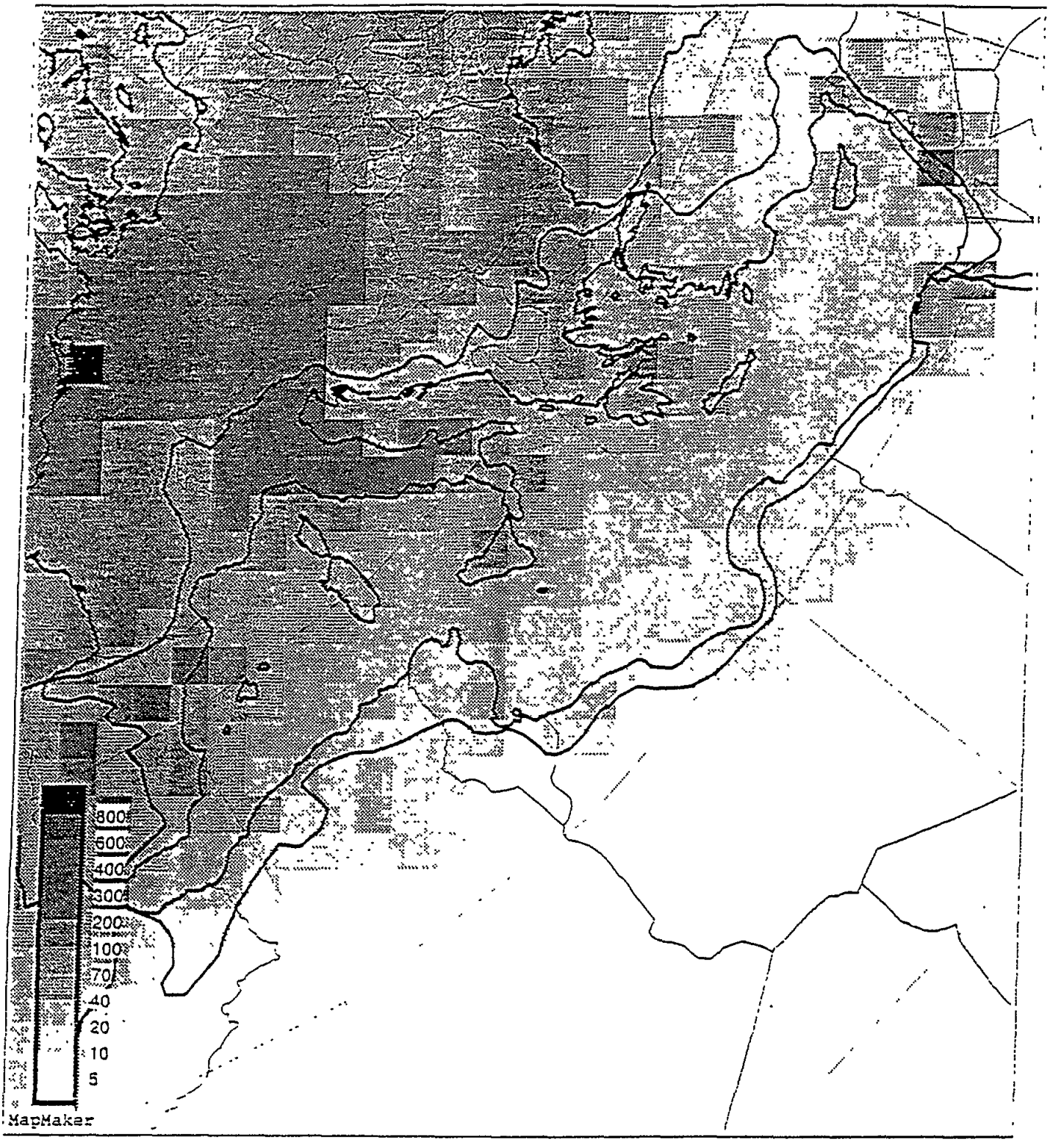

Fig.B5 Density of dry deposition of oxidised nitrogen on the Mediterranean in 1992
Unit = 1 mg N/m^2/year

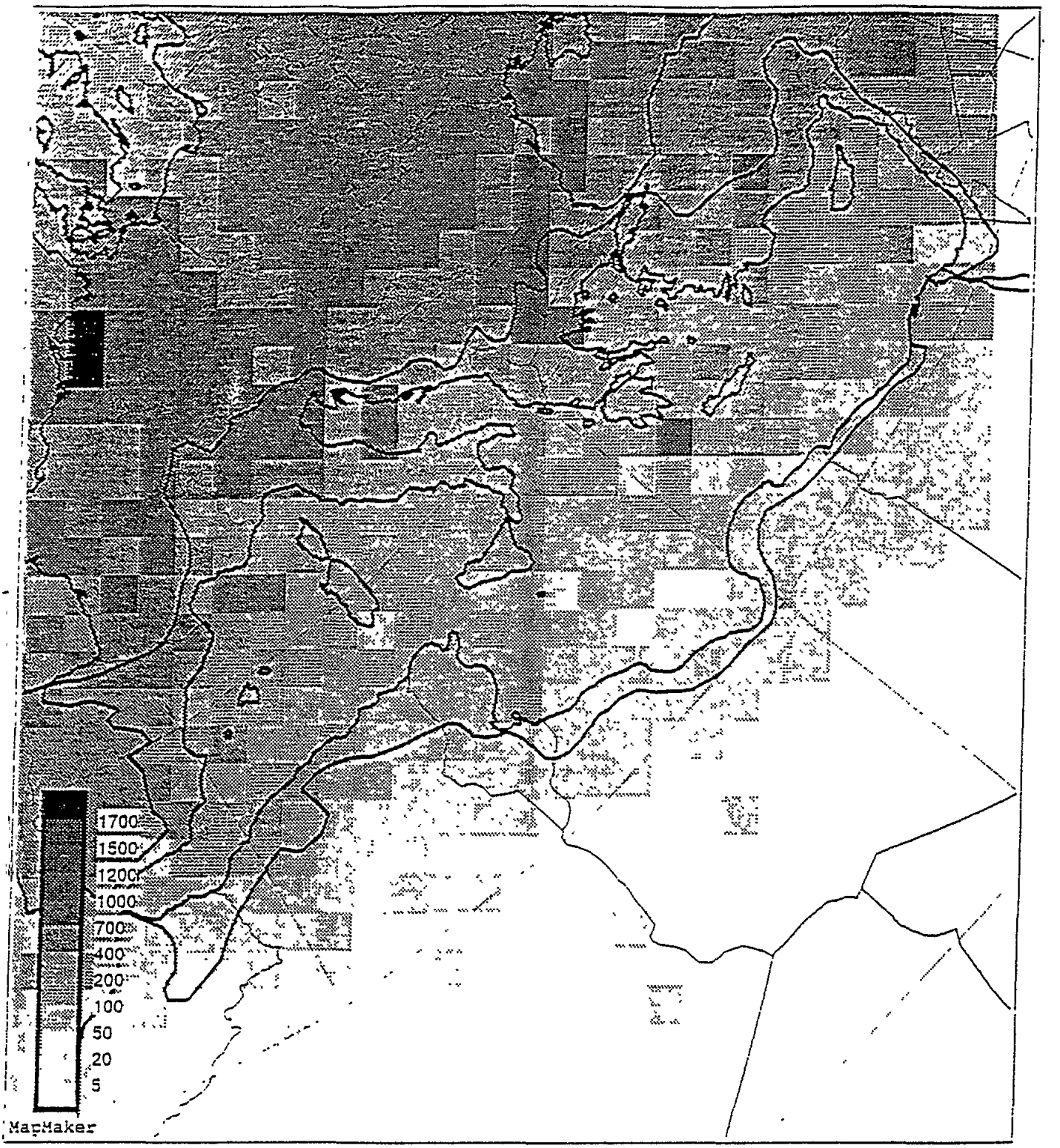

Fig.B6: Density of total deposition of reduced nitrogen on the Mediterranean in 1992
Unit = 1 mg N/m^2/year

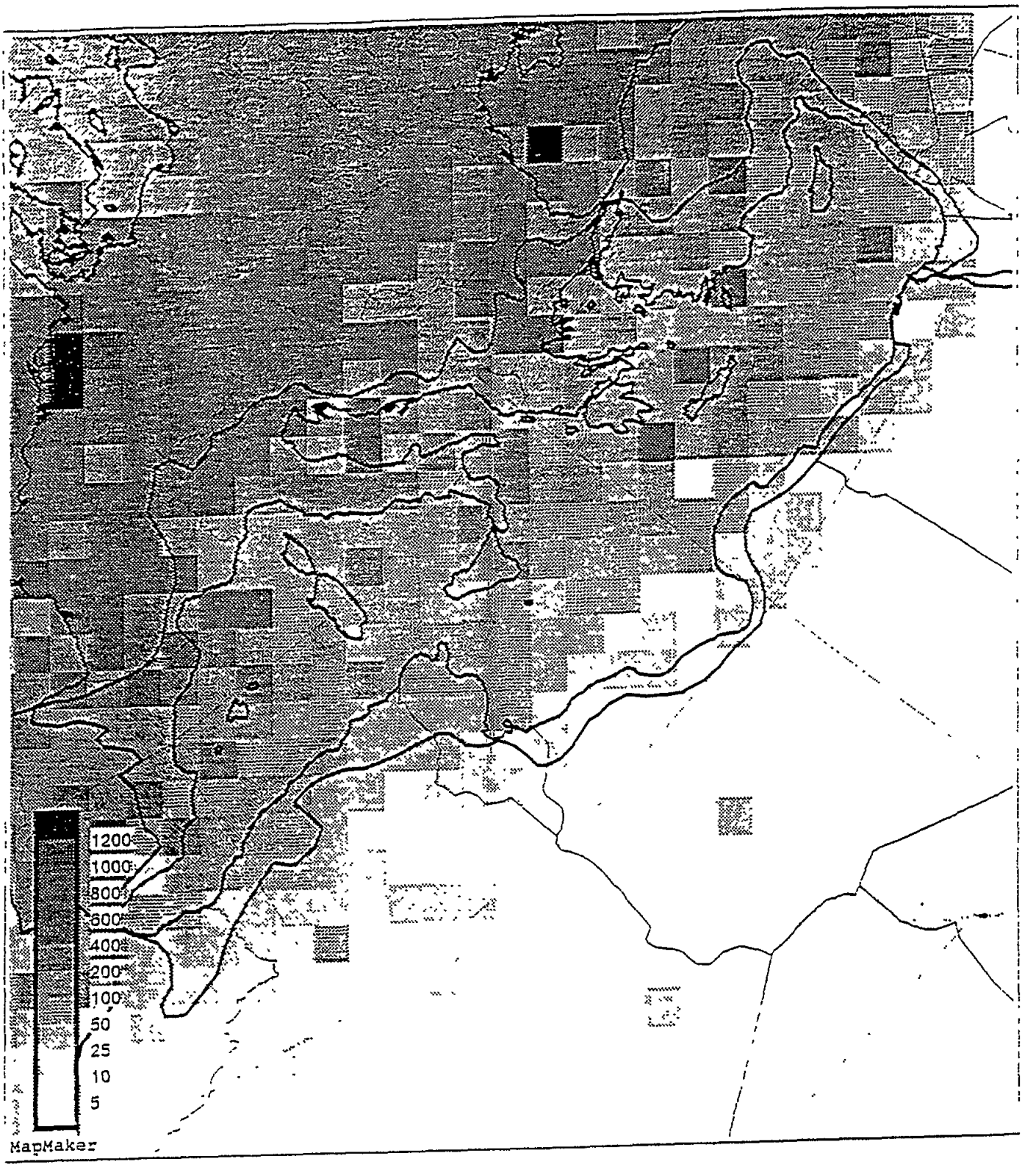

Fig.B7· Density of wet deposition of reduced nitrogen on the Mediterranean in 1992.
Unit = 1 mg N/m^2,year

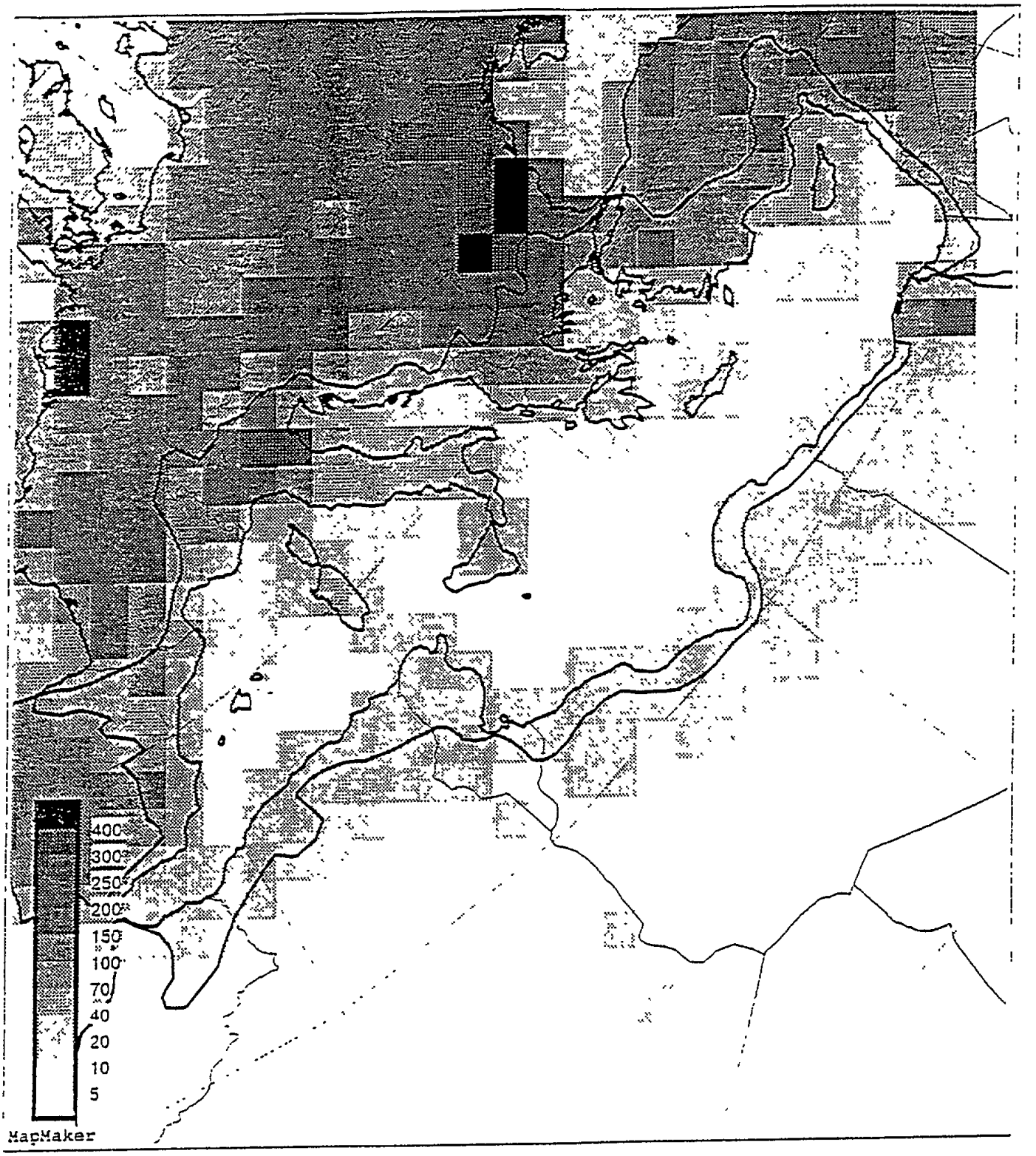

Fig.B8: Density of dry deposition of reduced nitrogen on the Mediterranean in 1992.
Unit = 1 mg N/m^2/year

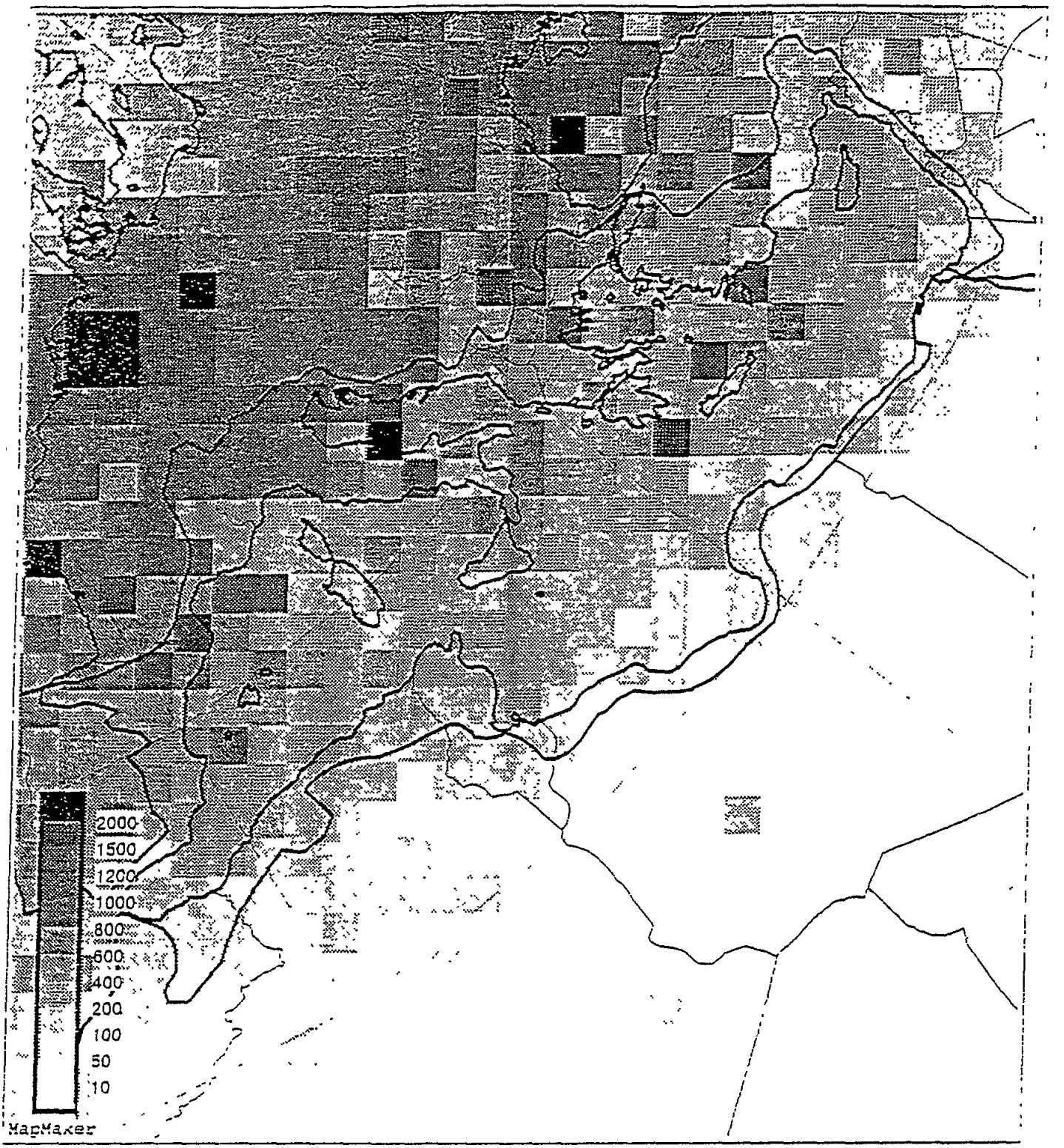

Fig.B9: Density of wet deposition of total nitrogen on the Mediterranean in 1992.
Unit = 1 mg N/m^2/year

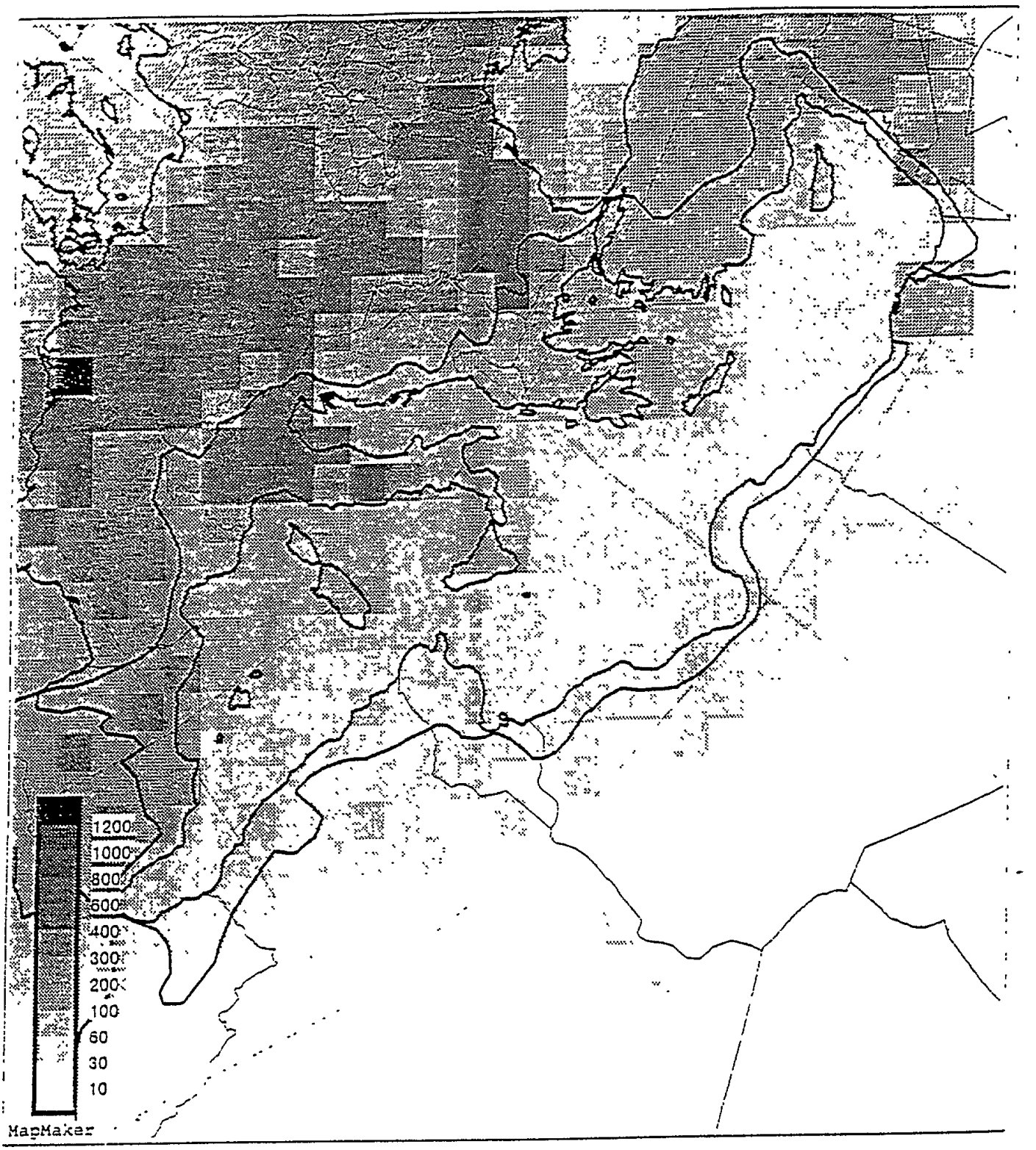

Fig.B10: Density of dry deposition of total nitrogen on the Mediterranean in 1992.
Unit = 1 mg N/m^2/year

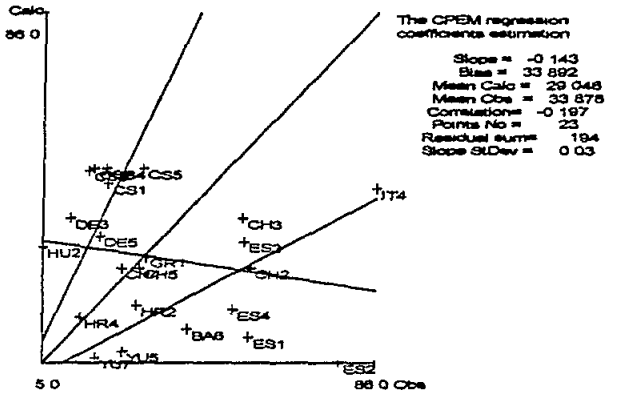

Fig.B11 *Comparison of model results with NO_2 air concentration measurements (all stations). Unit = 0.1 mg N/m^3.*

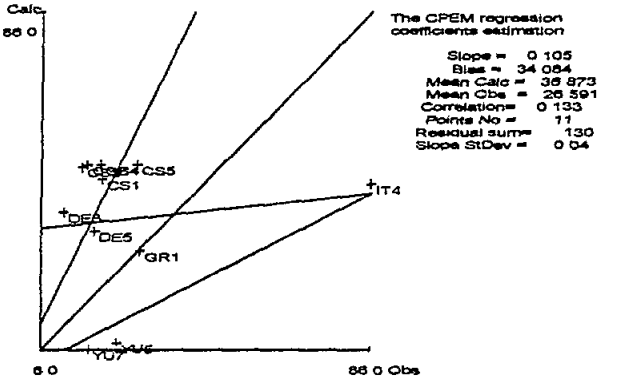

Fig.B12 *Comparison of model results with NO_2 air concentration measurements (stations of classes 1 and 2). Unit = 0.1 mg N/m^3.*

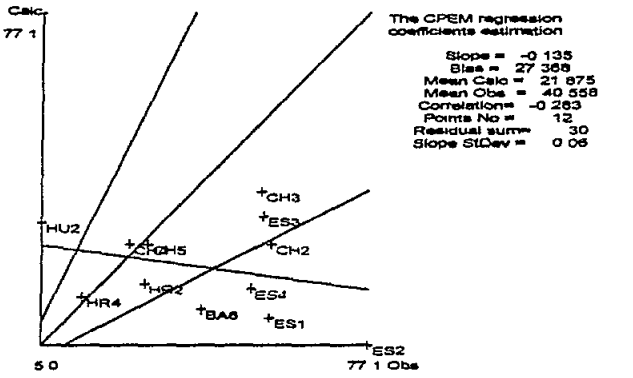

Fig.B13 *Comparison of model results with NO_2 air concentration measurements (stations of classes 3 and 4). Unit = 0.1 mg N/m^3.*

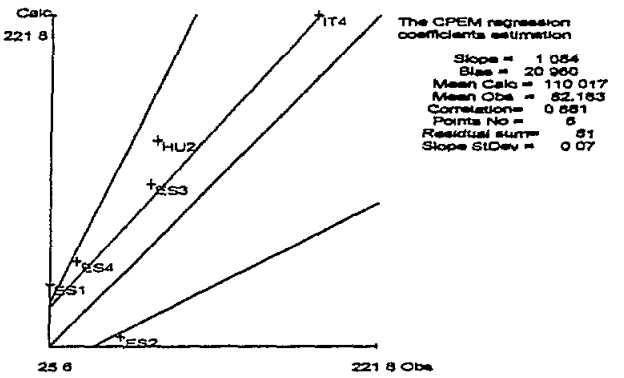

Fig.B14 *Comparison of model results with NO_3 + HNO_3 air concentration measurements (all stations). Unit = 0.1 mg N/m^3.*

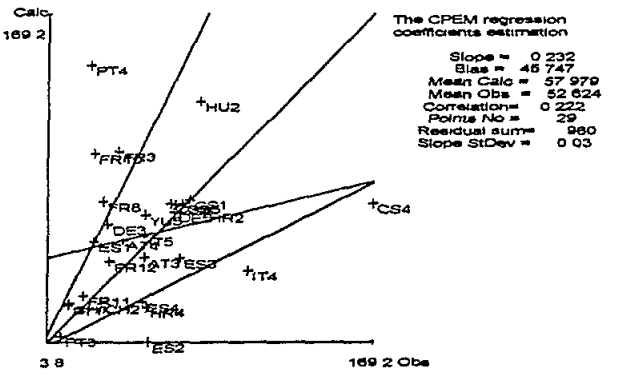

Fig.B15 *Comparison of model results with nitrates in precipitation measurements (all stations). Unit = 0.1 mg N/l.*

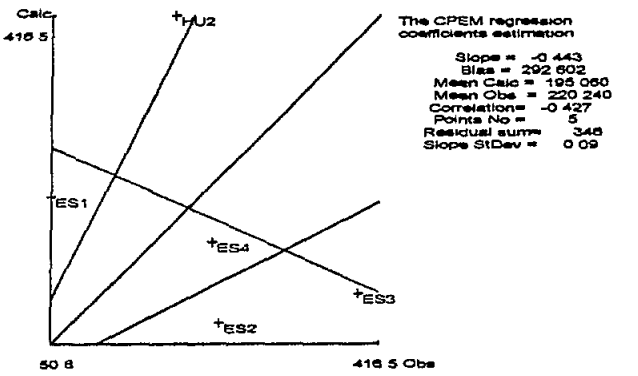

Fig.B16 *Comparison of model results with NH_3+NH_4 air concentration measurements (all stations). Unit = 0.1 mg N/m^3.*

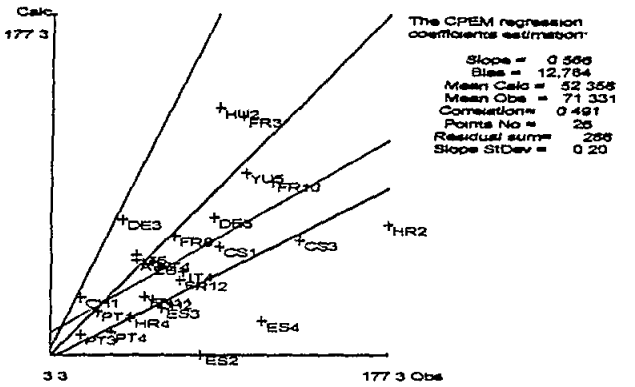

Fig.B17 *Comparison of model results with ammonium in precipitation measurements (all stations). Unit = 0.1 mg N/l.*

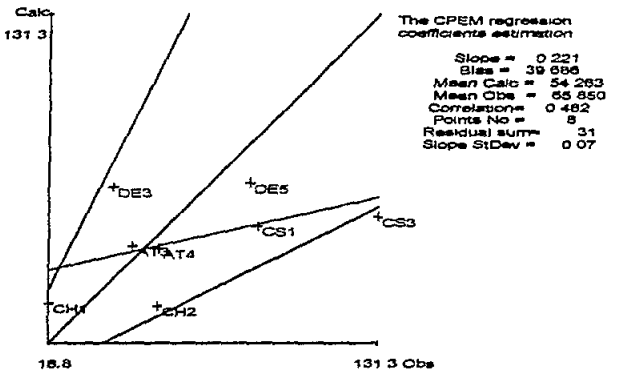

Fig.B18 *Comparison of model results with ammonium in precipitation measurements (stations of classes 1 and 2). Unit = 0.1 mg N/l.*

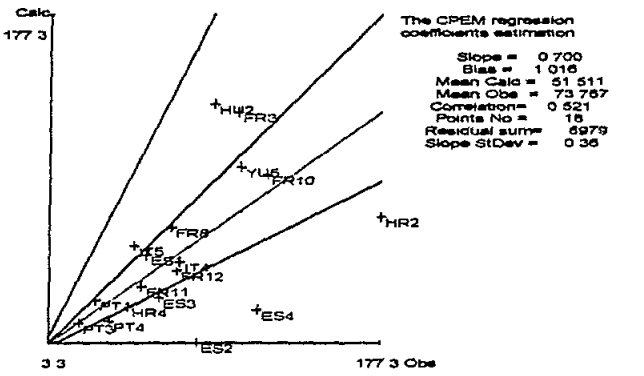

Fig.B19 *Comparison of model results with ammonium in precipitation measurements (stations of classes 3 and 4). Unit = 0.1 mg N/l.*

ANNEX C

TABLES OF ATMOSPHERIC DEPOSITION OF NITROGEN ON INDIVIDUAL EMEP AND LOLA GRID CELLS AND OF NITROGEN LEACHING FROM THESE CELLS

TABLE C1: *Atmospheric deposition of nitrogen on individual EMEP grid cells of the Mediterranean (Ntd) and leaching (runoff) of nitrogen from these cells (N leach)*

X	Y	N_{td}	N_{leach}		X	Y	N_{td}	N_{leach}	
EMEP grid		kt/yr	kt/yr	N_{leach}:N_{td} ratio	EMEP grid		kt/yr	kt/yr	N_{leach}:N_{td} ratio
58	11	3.07	0.00	0.00	64	8	6.26	0.00	0.00
59	11	2.93	0.00	0.00	64	9	1.25	0.00	0.00
59	12	6.43	0.64	0.10	64	11	0.41	1.00E-03	2.50E-03
60	6	0.88	0.00	0.00	64	12	0.88	0.00	0.00
60	7	1.23	0.01	0.01	64	16	1.55	1.90E-03	7.60E-04
60	10	1.98	0.19	0.10	64	17	17.58	2.43	0.14
60	11	6.37	0.49	0.08	64	18	36.33	2.12	0.06
60	12	15.97	2.58	0.16	64	19	41.18	19.55	0.47
61	5	0.35	0.00	0.00	64	20	1.37	0.06	0.04
61	6	0.38	0.00	0.00	65	6	0.12	1.70E-03	0.01
61	7	0.50	0.00	0.00	65	7	2.05	0.09	0.04
61	8	2.34	0.03	0.01	65	8	2.86	0 00	0.00
61	9	9.12	0.15	0.02	65	9	3.40	0.00	0.00
61	10	7.74	0.00	0.00	65	10	0.79	0.00	0.00
61	11	14.26	2.49	0.17	65	15	3.81	0.00	0.00
61	12	22.54	3.23	0.14	65	16	2.94	0.22	0.08
61	13	6.03	1.97	0.33	65	17	17.36	0.00	0.00
61	14	0.99	0.29	0.30	65	18	24.30	1.72	0.07
61	16	5.33	1.72	0.32	65	19	19.88	0.00	0.00
61	17	24.37	3.90	0.16	65	20	6.28	0.2	0.03
61	18	15.98	0.68	0.04	66	7	0.14	1.20E-03	0.01
62	4	0.43	0.00	0.00	66	8	0.76	3.90E-03	0.01
62	5	1.49	0.00	0.00	66	9	2.43	0.01	0.01
62	6	1.93	0.00	0.00	66	10	3.87	0.00	0.00
62	7	0.17	0.00	0.00	66	11	0.17	2.80E-04	1.60E-03
62	8	3.96	0.05	0.01	66	13	0.31	0.00	0.00
62	9	4.29	0.11	0.03	66	14	3.79	0.00	0.00
62	10	4.70	0.00	0.00	66	15	2.88	0.04	0.02
62	11	1.74	0.00	0.00	66	17	17.11	1.31	0.08
62	12	9.65	0.00	0.00	66	18	11.50	4.35	0.38
62	13	21.5	6.21	0.29	66	19	1.89	2.30E-04	1.20E-04
62	14	6.32	1.00	0.16	66	20	3.43	0.13	0.04
62	15	25.33	5.57	0.22	67	9	0.23	2.00E-03	0.01
62	16	15.69	0.2	0.01	67	10	1.21	0.00	0.00
62	17	24.47	0.00	0.00	67	11	2.18	0.00	0.00
62	18	1.30	0.41	0.31	67	12	0.36	0.00	0.00
63	4	0.14	0.00	0.00	67	13	0.19	0.00	0.00
63	5	1.15	0.00	0.00	67	14	2.63	0.00	0.00
63	6	0.99	4.80E-03	4.90E-03	67	17	17.17	0.06	3.80E-03
63	7	3.39	0.00	0.00	67	18	14.56	0.13	0.01
63	10	0.28	0.00	0.00	67	19	8.35	0.00	0.00
63	11	0.50	9.10E-05	1.80E-04	67	20	4.82	0.65	0.14
63	12	0.56	9.90E-04	1.80E-03	68	10	0.12	0.00	0.00
63	15	7.75	0.39	0.05	68	11	1.34	0.00	0.00
63	16	13.35	0.0C	0.00	68	12	2.06	0.00	0.00
63	17	37.41	1.08	0.03	68	13	0.60	0.00	0.00
63	18	27 19	2.66	0.1	68	17	14.63	12.84	0.88*
63	19	8.15	0.81	0.1	68	18	6.91	0.00	0.00
64	5	0.04	4.30E-04	0.01	68	20	15.53	0 64	0.04
64	6	1.38	0.02	0.01	69	11	1.37	0.03	0.02
64	7	3.25	0.0	4 50E-03	69	12	2.92	0 00	0.00

X	Y	N_{td}	N_{leach}		X	Y	N_{td}	N_{leach}	
EMEP grid		kt/yr	kt/yr	N_{leach}:N_{td} ratio	EMEP grid		kt/yr	kt/yr	N_{leach}:N_{td} ratio
69	13	0.91	0.00	0.00	75	11	0.19	3.90E-03	0.02
69	14	0.48	0.00	0.00	75	12	0.46	0.00	0.00
69	15	0.83	0.00	0.00	75	19	0.44	0.00	0.00
69	17	3.78	0.00	0.00	75	21	0.13	0.00	0.00
69	18	14.22	0.00	0.00	75	22	2.20	0.00	0.00
69	20	6.68	0.36	0.05	75	23	8.10	0.39	0.05
69	21	0.75	0.08	0.1	75	24	3.16	0.00	0.00
70	10	0.09	1.60E-03	0.02	76	11	0.14	0.00	0.00
70	11	1.15	0.02	0.02	76	12	0.45	0.00	0.00
70	12	1.16	0.00	0.00	76	13	0.07	0.00	0.00
70	14	0.09	0.00	0.00	76	19	0.94	0.00	0.00
70	15	5.30	0.11	0.02	76	20	1.06	0.00	0.00
70	16	3.13	0.00	0.00	76	22	1.66	0.00	0.00
70	17	3.65	0.38	0.1	76	23	10.72	0.00	0.00
70	18	5.18	0.00	0.00	76	24	5.23	0.04	0.01
70	19	0.63	0.00	0.00	76	25	0.31	3.20E-03	0.01
70	20	12.22	0.00	0.00	77	11	0.02	0.00	0.00
70	21	7.24	2.04	0.28	77	12	0.18	0.00	0.00
70	22	2.46	0.48	0.19	77	13	0.54	0.00	0.00
71	10	0.17	3.10E-03	0.02	77	14	0.02	0.00	0.00
71	11	0.81	0.00	0.00	77	15	0.52	1.00E-03	1.90E-03
71	15	3.37	2.50E-03	7.40E-04	77	16	2.45	0.00	0.00
71	16	1.12	0.00	0.00	77	17	0.61	0.00	0.00
71	17	0.71	0.00	0.00	77	19	0.36	0.00	0.00
71	19	3.86	0.00	0.00	77	20	2.68	0.00	0.00
71	20	8.36	0.00	0.00	77	22	2.08	0.00	0.00
71	21	17.87	0.00	0.00	77	23	3.62	0.08	0.02
71	22	18.63	0.00	0.00	77	24	9.73	0.00	0.00
71	23	15.44	1.91	0.12	77	25	15.22	0.00	0.00
72	10	0.43	0.02	0.05	77	26	2.51	0.11	0.04
72	11	0.32	3.30E-03	0.01	77	28	1.13	0.00	0.00
72	19	1.84	0.00	0.00	77	29	0.33	0.00	0.00
72	20	11.90	1.53	0.13	78	12	0.08	0.00	0.00
72	21	18.10	0.00	0.00	78	13	0.24	3.00E-04	1.20E-03
72	22	8.98	3.90	0.43	78	14	1.08	0.00	0.00
72	23	18.27	0.78	0.04	78	15	0.66	0.00	0.00
72	24	7.14	0.50	0.07	78	16	0.83	0.00	0.00
73	10	0.15	0.00	0.00	78	17	0.57	0.00	0.00
73	11	0.48	0.02	0.04	78	23	0.15	0.00	0.00
73	12	0.22	0.00	0.00	78	24	0.13	0.00	0.00
73	19	8.76	0.13	0.01	78	25	3.67	0.01	1.90E-03
73	20	10.40	0.00	0.00	78	26	6.03	2.04	0.34
73	23	7.93	0.00	0.00	78	27	3.99	0.00	0.00
73	24	7.51	0.90	0.12	78	28	5.14	0.00	0.00
74	10	0.05	0.00	0.00	78	29	0.76	0.01	0.01
74	11	0.22	0.00	0.00	79	13	0.02	0.00	0.00
74	12	1.17	0.13	0.11	79	14	0.04	0.00	0.00
74	19	5.92	0.00	0.00	79	15	0.13	0.00	0.00
74	20	5.35	0.00	0.00	79	16	0.80	0.00	0.00
74	21	1.47	0.00	0.00	79	17	0.37	0.00	0.00
74	22	1.04	0.00	0.00	79	18	0.92	0.00	0.00
74	23	3.07	0.03	0.01	79	24	0 82	0.00	0.00
75	10	0.01	0.00	0.00	79	25	0 73	0.00	0.00

X	Y	N_{td}	N_{leach}	
EMEP grid		kt/yr	kt/yr	N_{leach}:N_{td} ratio
79	26	0.51	0.00	0.00
79	27	2.62	0.00	0.00
79	28	0.29	0.00	0.00
80	16	0.04	0.00	0.00
80	17	0.23	0.00	0.00
80	18	1.77	0.00	0.00
80	19	0.19	0.00	0.00
80	24	0.37	0.00	0.00
80	25	1.07	0.00	0.00
80	27	0.79	0.00	0.00
81	17	0.02	0.00	0.00
81	18	0.25	0.00	0.00
81	19	1.95	0.00	0.00
81	20	0.23	0.00	0.00
81	25	0.27	0.00	0.00
81	26	0.70	0.00	0.00
82	18	0.03	0.00	0.00
82	19	0.68	0.00	0.00
82	23	0.02	0.00	0.00
82	25	3.39	0.00	0.00
83	23	0.30	0.00	0.00
83	24	1.67	0.00	0.00
83	25	0.62	0.00	0.00

- Explanation see in sections 4.4 and 4.3

- TABLE C2: *Atmospheric deposition of nitrogen on individual LoLa grid cells of the Nile watershed and leaching (runoff) of nitrogen from these cells (N leach)*

Lo ° grid	La °	N_{td} kt	N_{leach} kt	N_{leach}:N_{td} ratio
30	32	0.27	0.00	0.00
31	32	0.44	0.00	0.00
29	31	0.35	0.00	0.00
30	31	0.69	0.00	0.00
31	31	1.12	0.00	0.00
32	31	0.45	0.00	0.00
28	30	0.07	0.00	0.00
29	30	0.63	0.00	0.00
30	30	0.70	0.00	0.00
31	30	0.85	0.00	0.00
28	29	0.28	0.00	0.00
29	29	0.71	0.00	0.00
30	29	0.71	0.00	0.00
31	29	0.91	0.00	0.00
32	29	0.34	0.00	0.00
28	28	0.04	0.00	0.00
29	28	0.68	0.00	0.00
30	28	0.71	0.00	0.00
31	28	1.15	0.00	0.00
32	28	0.98	0.00	0.00
29	27	0.36	0.00	0.00
30	27	0.72	0.00	0.00
31	27	1.16	0.00	0.00
32	27	1.16	0.00	0.00
33	27	0.52	0.00	0.00
27	26	0.65	0.00	0.00
28	26	0.62	0.00	0.00
29	26	0.54	0.00	0.00
30	26	0.72	0.00	0.00
31	26	0.72	0.00	0.00
32	26	0.72	0.00	0.00
33	26	0.69	0.00	0.00
34	26	0.09	0.00	0.00
26	25	0.18	0.00	0.00
27	25	0.51	0.00	0.00
28	25	0.51	0.00	0.00
29	25	0.51	0.00	0.00
30	25	0.51	0.00	0.00
31	25	0.51	0.00	0.00
32	25	0.51	0.00	0.00
33	25	0.90	0.00	0.00
34	25	0.45	0.00	0.00
26	24	0.43	0.00	0.00
27	24	0.51	0.00	0.00
28	24	0.51	0.00	0.00
29	24	0.51	0.00	0.00
30	24	0.51	0.00	0.00
31	24	0.51	0.00	0.00
32	24	0.51	0.00	0.00
33	24	0.90	0.00	0.00
34	24	0.54	0.00	0.00
25	23	0.23	0.00	0.00
26	23	0.51	0.00	0.00
27	23	0.51	0.00	0.00
28	23	0.51	0.00	0.00
29	23	0.51	0.00	0.00
30	23	0.51	0.00	0.00
31	23	0.51	0.00	0.00
32	23	0.51	0.00	0.00
33	23	0.91	0.00	0.00
34	23	0.55	0.00	0.00
24	22	0.10	0.00	0.00
25	22	0.52	0.00	0.00
26	22	0.52	0.00	0.00
27	22	0.52	0.00	0.00
28	22	0.52	0.00	0.00
29	22	0.52	0.00	0.00
30	22	0.52	0.00	0.00
31	22	0.92	0.00	0.00
32	22	0.92	0.00	0.00
33	22	0.92	0.00	0.00
34	22	0.87	0.00	0.00
24	21	0.21	0.00	0.00
25	21	0.52	0.00	0.00
26	21	0.52	0.00	0.00
27	21	0.52	0.00	0.00
28	21	0.52	0.00	0.00
29	21	0.52	0.00	0.00
30	21	0.92	0.00	0.00
31	21	0.92	0.00	0.00
32	21	0.92	0.00	0.00
33	21	0.92	0.00	0.00
34	21	0.92	0.00	0.00
35	21	0.32	0.00	0.00
24	20	0.61	0.00	0.00
25	20	1.22	0.00	0.00
26	20	1.22	0.00	0.00
27	20	1.22	0.00	0.00
28	20	1.22	0.00	0.00
29	20	1.22	0.00	0.00
30	20	1.22	0.00	0.00
31	20	1.22	0.00	0.00
32	20	1.22	0.00	0.00
33	20	1.22	0.00	0.00
34	20	1.22	0.00	0.00
35	20	1.22	0.00	0.00
36	20	0.49	0.00	0.00
24	19	1.02	0.00	0.00
25	19	1.23	0.00	0.00
26	19	1.23	0.00	0.00
27	19	1.23	0.00	0.00
28	19	1.23	0.00	0.00
29	19	1.23	0.00	0.00
30	19	1.23	0.00	0.00

Lo° grid	La°	N_{td} kt	N_{leach} kt	N_{leach}:N_{td} ratio
31	19	1.23	0.00	0.00
32	19	1.23	0.00	0.00
33	19	1.23	0.00	0.00
34	19	1.23	0.00	0.00
35	19	1.23	0.00	0.00
36	19	0.86	0.00	0.00
23	18	0.74	0.00	0.00
24	18	1.23	0.00	0.00
25	18	1.23	0.00	0.00
26	18	1.23	0.00	0.00
27	18	1.23	0.00	0.00
28	18	1.23	0.00	0.00
29	18	1.23	0.00	0.00
30	18	1.23	0.00	0.00
31	18	1.23	0.00	0.00
32	18	1.23	0.00	0.00
33	18	1.23	0.00	0.00
34	18	1.23	0.00	0.00
35	18	1.23	0.00	0.00
36	18	0.62	0.00	0.00
22	17	0.31	0.00	0.00
23	17	1.24	0.00	0.00
24	17	1.24	0.00	0.00
25	17	1.24	0.00	0.00
26	17	1.24	0.00	0.00
27	17	1.24	0.00	0.00
28	17	1.24	0.00	0.00
29	17	1.24	0.00	0.00
30	17	1.24	0.00	0.00
31	17	1.24	0.00	0.00
32	17	1.24	0.00	0.00
33	17	1.24	0.00	0.00
34	17	1.24	0.00	0.00
35	17	1.24	0.00	0.00
36	17	0.68	0.00	0.00
22	16	1.00	0.00	0.00
23	16	1.25	0.00	0.00
24	16	1.25	0.00	0.00
25	16	1.25	0.00	0.00
26	16	1.25	0.00	0.00
27	16	1.25	0.00	0.00
28	16	1.25	0.00	0.00
29	16	1.25	0.00	0.00
30	16	1.25	0.00	0.00
31	16	1.25	0.00	0.00
32	16	1.25	0.00	0.00
33	16	1.25	0.00	0.00
34	16	1.25	0.00	0.00
35	16	1.25	0.00	0.00
36	16	1.12	0.00	0.00
37	16	0.69	0.00	0.00
38	16	0.56	0.00	0.00
24	15	1.61	0.00	0.00
25	15	2.15	0.00	0.00
26	15	2.15	0.00	0.00
27	15	2.15	0.00	0.00
28	15	2.15	0.00	0.00
29	15	2.15	0.00	0.00
30	15	2.15	0.00	0.00
31	15	2.15	0.00	0.00
32	15	2.15	0.00	0.00
33	15	2.15	0.00	0.00
34	15	2.15	0.00	0.00
35	15	2.15	0.00	0.00
36	15	2.15	0.00	0.00
37	15	2.15	0.00	0.00
38	15	2.15	0.00	0.00
39	15	1.29	0.00	0.00
24	14	1.29	0.00	0.00
25	14	2.16	0.00	0.00
26	14	2.16	0.00	0.00
27	14	2.16	0.00	0.00
28	14	2.16	0.00	0.00
29	14	2.16	0.00	0.00
30	14	2.16	0.00	0.00
31	14	2.16	0.00	0.00
32	14	2.16	0.00	0.00
33	14	2.16	0.00	0.00
34	14	2.16	0.00	0.00
35	14	2.16	0.00	0.00
36	14	2.16	0.00	0.00
37	14	2.16	0.00	0.00
38	14	2.16	0.00	0.00
39	14	1.83	0.00	0.00
23	13	0.11	0.00	0.00
24	13	2.16	0.00	0.00
25	13	2.16	0.00	0.00
26	13	2.16	0.00	0.00
27	13	2.16	0.00	0.00
28	13	2.16	0.00	0.00
29	13	2.16	0.00	0.00
30	13	2.16	0.00	0.00
31	13	2.16	0.00	0.00
32	13	2.16	0.00	0.00
33	13	2.16	0.00	0.00
34	13	2.16	0.00	0.00
35	13	2.16	0.00	0.00
36	13	2.16	0.00	0.00
37	13	2.16	0.00	0.00
38	13	2.16	0.00	0.00
39	13	1.73	0.00	0.00
24	12	2.02	0.00	0.00
25	12	2.17	0.00	0.00
26	12	2.17	0.00	0.00
27	12	2.17	0.00	0.00
28	12	2.17	0.00	0.00

Lo ° grid	La °	N_{td} kt	N_{leach} kt	N_{leach}:N_{td} ratio
29	12	2.17	0.00	0.00
30	12	2.17	0.00	0.00
31	12	2.17	0.00	0.00
32	12	2.17	0.00	0.00
33	12	2.17	0.00	0.00
34	12	2.17	0.00	0.00
35	12	2.17	0.00	0.00
36	12	2.17	0.00	0.00
37	12	2.17	0.00	0.00
38	12	2.17	0.00	0.00
39	12	1.96	0.00	0.00
24	11	1.81	0.00	0.00
25	11	2.18	0.00	0.00
26	11	2.18	0.00	0.00
27	11	2.18	0.00	0.00
28	11	2.18	0.00	0.00
29	11	2.18	0.00	0.00
30	11	2.18	0.00	0.00
31	11	2.18	0.00	0.00
32	11	2.18	0.00	0.00
33	11	2.18	0.00	0.00
34	11	2.18	0.00	0.00
35	11	2.18	0.00	0.00
36	11	2.18	0.00	0.00
37	11	2.18	0.00	0.00
38	11	2.18	0.00	0.00
39	11	1.96	0.00	0.00
23	10	1.09	0.00	0.00
24	10	2.60	0.00	0.00
25	10	2.73	0.00	0.00
26	10	2.73	0.00	0.00
27	10	2.73	0.00	0.00
28	10	2.73	0.00	0.00
29	10	2.73	0.00	0.00
30	10	2.73	0.00	0.00
31	10	2.73	0.00	0.00
32	10	2.73	0.00	0.00
33	10	2.73	0.00	0.00
34	10	2.73	0.00	0.00
35	10	2.73	0.00	0.00
36	10	2.60	0.00	0.00
37	10	2.46	0.00	0.00
38	10	2.40	0.00	0.00
39	10	1.09	0.00	0.00
23	9	0.19	0.00	0.00
24	9	0.96	0.00	0.00
25	9	2.60	0.00	0.00
26	9	2.74	0.00	0.00
27	9	2.74	0.00	0.00
28	9	2.74	0.00	0.00
29	9	2.74	0.00	0.00
30	9	2.74	0.00	0.00
31	9	2.74	0.00	0.00
32	9	2.74	0.00	0.00
33	9	2.74	0.00	0.00
34	9	2.74	0.00	0.00
35	9	2.74	0.00	0.00
36	9	2.74	0.00	0.00
37	9	0.27	0.00	0.00
25	8	1.51	0.00	0.00
26	8	2.75	0.00	0.00
27	8	2.75	0.00	0.00
28	8	2.75	0.00	0.00
29	8	2.75	0.00	0.00
30	8	2.75	0.00	0.00
31	8	2.75	0.00	0.00
32	8	2.75	0.00	0.00
33	8	2.75	0.00	0.00
34	8	2.75	0.00	0.00
35	8	2.66	0.00	0.00
36	8	0.69	0.00	0.00
26	7	1.51	0.00	0.00
27	7	2.75	0.00	0.00
28	7	2.75	0.00	0.00
29	7	2.75	0.00	0.00
30	7	2.75	0.00	0.00
31	7	2.75	0.00	0.00
32	7	2.75	0.00	0.00
33	7	2.75	0.00	0.00
34	7	2.75	0.00	0.00
35	7	1.79	0.00	0.00
27	6	1.79	0.00	0.00
28	6	2.76	0.00	0.00
29	6	2.76	0.00	0.00
30	6	2.76	0.00	0.00
31	6	2.76	0.00	0.00
32	6	2.76	0.00	0.00
33	6	2.76	0.00	0.00
34	6	2.76	0.00	0.00
35	6	1.16	0.00	0.00
27	5	0.37	0.00	0.00
28	5	1.10	0.00	0.00
29	5	1.47	0.00	0.00
30	5	2.76	0.00	0.00
31	5	3.68	0.00	0.00
32	5	2.76	0.00	0.00
33	5	2.76	0.00	0.00
34	5	0.97	0.00	0.00
31	4	3.50	0.00	0.00
32	4	2.76	0.00	0.00
33	4	2.76	0.00	0.00
34	4	1.24	0.00	0.00
31	3	2.77	0.00	0.00
32	3	2.77	0.00	0.00

Lo ° grid	La °	N_{td} kt	N_{leach} kt	N_{leach}:N_{td} ratio
33	3	2.77	0.00	0.00
34	3	2.35	0.00	0.00
29	2	0.14	0.00	0.00
30	2	1.80	0.00	0.00
31	2	2.77	0.00	0.00
32	2	2.77	0.00	0.00
33	2	2.77	0.00	0.00
34	2	2.49	0.00	0.00
35	2	0.42	0.00	0.00
29	1	1.52	0.00	0.00
30	1	2.77	0.00	0.00
31	1	2.77	0.00	0.00
32	1	2.77	0.00	0.00
33	1	2.77	0.00	0.00
34	1	2.77	0.00	0.00
35	1	0.97	0.00	0.00
29	0	2.49	0.00	0.00
30	0	2.77	0.00	0.00
31	0	2.77	0.00	0.00
32	0	2.77	0.00	0.00
33	0	2.77	0.00	0.00
34	0	2.77	0.00	0.00
35	0	1.80	0.00	0.00
29	-1	1.35	0.00	0.00
30	-1	2.46	0.00	0.00
31	-1	2.46	0.00	0.00
32	-1	2.46	0.00	0.00
33	-1	2.46	0.00	0.00
34	-1	2.46	0.00	0.00
35	-1	1.60	0.00	0.00
29	-2	1.23	0.00	0.00
30	-2	2.09	0.00	0.00
31	-2	2.21	0.00	0.00
32	-2	2.46	0.00	0.00
33	-2	2.46	0.00	0.00
34	-2	2.34	0.00	0.00
35	-2	0.86	0.00	0.00
29	-3	1.23	0.00	0.00
30	-3	0.86	0.00	0.00
31	-3	0.12	0.00	0.00
32	-3	1.11	0.00	0.00
33	-3	1.35	0.00	0.00
34	-3	0.25	0.00	0.00
29	-4	0.25	0.00	0.00
30	-4	0.25	0.00	0.00

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