MEDITERRANEAN ACTION PLAN
MED POL

UNEP

UNITED NATIONS ENVIRONMENT PROGRAMME

WORLD METEOROLOGICAL ORGANIZATION

ASSESSMENT OF AIRBORNE POLLUTION OF THE MEDITERRANEAN SEA BY SULPHUR AND NITROGEN COMPOUNDS AND HEAVY METALS IN 1991

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This volume is the eighty-fifth issue of the Mediterranean Action Plan Technical Report Series.

This series will collect and disseminate selected scientific reports obtained through the implementation of the various MAP components· Pollution Monitoring and Research Programme (MED POL), Blue Plan, Priority Actions Programme, Specially Protected Areas, Regional Marine Pollution Emergency Response Centre for the Mediterranean Sea, Environment Remote Sensing and Protection of Historic Sites.

Ce volume constitue le quatre-vingt cinquième numéro de la série des Rapports techniques du Plan d'action pour la Méditerranée

Cette série permettra de rassembler et de diffuser certains des rapports scientifiques établis dans le cadre de la mise en oeuvre des diverses composantes du PAM. Programme de surveillance continue et de recherche en matière de pollution (MED POL), Plan Bleu, Programme d'actions prioritaires, Aires spécialement protégées, Centre régional méditerranéen pour l'intervention d'urgence contre la pollution marine accidentelle, Centre méditerranéen de télédétection et Protection des sites historiques.

PREFACE

The United Nations Environment Programme (UNEP) convened an Intergovernmental Meeting on the Protection of the Mediterranean (Barcelona), 28 January - 4 February 1975), which was attended by representatives of 16 States bordering on the Mediterranean Sea. The meeting discussed the various measures necessary for the prevention and control of pollution of the Mediterranean Sea, and concluded by adopting an Action Plan consisting of three substantive components:

- Integrated planning of the development and management of the resources of the Mediterranean Basin (management component);

- Co-ordinated programme for research, monitoring and exchange of information and assessment of the state of pollution and of protection measures (assessment component);

- Framework convention and related protocols with their technical annexes for the protection of the Mediterranean environment (legal component)

All components of the Action Plan are interdependent and provide a framework for comprehensive action to promote both the protection and the continued development of the Mediterranean ecoregion. No component is an end in itself. The Action Plan is intended to assist the Mediterranean Governments in formulating their national policies related to the continuous development and protection of the Mediterranean area and to improve their ability to identify various options for alternative patterns of development and to make choices and appropriate allocations of resources.

The Co-ordinated Mediterranean Research and Monitoring Programme (MED POL) was approved as the assessment (scientific/technical component of the Action Plan.

The general objectives of its pilot phase (MED POL - Phase I), which evolved through a series of expert and intergovernmental meetings, were:

- to formulate and carry out a co-ordinated pollution monitoring and research programme taking into account the goals of the Mediterranean Action Plan and the capabilities of the Mediterranean research centres to participate in it;

- to assist national research centres in developing their capabilities to participate in the programme,

- to analyse the sources, amounts, levels, pathways, trends and effects of pollutants relevant to the Mediterranean Sea;

- to provide the scientific/technical information needed by the Governments of the Mediterranean States and the EEC for the negotiation and implementation of the Convention for the Protection of the Mediterranean Sea against Pollution and its related protocols,

- to build up consistent time-series of data on the sources, pathways, levels and effects of pollutants in the Mediterranean Sea and thus to contribute to the scientific knowledge of the Mediterranean Sea.

MED POL - Phase I initially consisted of seven pilot projects and baseline studies on the monitoring of oil, petroleum hydrocarbons and microbial pollution in sea water, heavy metals and chlorinated hydrocarbons in marine organisms, populations, communities and ecosystems In addition, four related projects were also included to broaden the scope of the programme or to provide ancillary support

Based on the recommendations made at various expert and intergovernmental meetings, a draft Long-term (1981-1990) Programme for pollution monitoring and Research in the Mediterranean (MED POL-Phase II) was formulated by the Secretariat of the Barcelona Convention (UNEP), in co- operation with the United Nations Agencies which were responsible for the technical implementation of MED POL-Phase I, and it was formally approved by the Second Meeting of the Contracting Parties of the Mediterranean Sea against pollution and its related protocols and Intergovernmental Review Meeting of Mediterranean Coastal States of the Action Plan held in Cannes, 2-7 March 1981.

The general long-term objectives of MED POL-Phase II were to further the goals of the Barcelona Convention by assisting the Parties to prevent, abate and combat pollution of the Mediterranean Sea area and to protect and enhance the marine environment of the area. The specific objectives were designed to provide, on a continuous basis, the Parties to the Barcelona Convention and its related protocols with:

- information required for the implementation of the Convention and the protocols,
- indicators and evaluation of the effectiveness of the pollution prevention measures taken under the Convention and the protocols;
- scientific information which may lead to eventual revisions and amendments of the relevant provisions of the Convention and the protocols and for the formulation of additional protocols;
- information which could be used in formulating environmentally sound national, bilateral and multilateral management decisions essential for the continuous socio- economic development of the Mediterranean region on a sustainable basis;
- periodic assessment of the state of pollution of the Mediterranean Sea.

The monitoring of, and research on, pollutants affecting the Mediterranean marine environment reflects primarily the immediate and long-term requirements of the Barcelona Convention and its protocols, but also takes into account factors needed for the understanding of the relationship between the socio-economic development of the region and the pollution of the Mediterranean Sea.

Individual and collective training is provided for scientists and technicians in techniques (methods) required for their effective participation in monitoring and research envisaged in the framework of MED POL - Phase II This assistance is in the form of fellowships, experts, workshops, seminars, grants for attendance to meetings, etc., and covers training in analytical and sampling techniques, data processing, interpretation of results and various research topics.

As in MED POL-Phase I, the overall co-ordination and guidance for MED POL-Phase II is provided by UNEP as the secretariat of the Mediterranean Action Plan (MAP). Co- operating specialized United Nations Agencies (FAO, UNESCO, WHO, WMO, IAEA, IOC) are responsible for the technical implementation and day-to-day co-ordination of the work of national centres participating in monitoring and research.

The eighty-fifth volume of the MAP Technical Reports Series presents an assessment of airborne pollution of the Mediterranean Sea by sulphur and nitrogen compounds and heavy metals in 1991.

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List of countries notations

Emitters	Notations
Albania	AL
Austria	AT
Belgium	BE
Bulgaria	BG
Czech Slovak	CS
Denmark	DK
Finland	FI
France	FR
Germany	GE
Greece	GR
Hungary	HU
Iceland	IS
Ireland	IE
Italy	IT
Luxembourg	LU
Netherlands	NL
Sweden & Norway	SK
Poland	PL
Portugal	PT
Romania	RO
Spain	ES
Switzerland	CH
Turkey	TR
United Kingdom	GB
Yugoslavia*	YU
Russia,Turkmenistan, Kazakhstan,Uzbekistan	RF
The Ukraine	UR
Moldova	MOL
Armenia,Georgia,Azerbaijan	ZKV
Belarus	BR
Estonia,Latvia,Lithuania	PBL
Morocco	MOR
Algeria	ALG
Tunisia	TUN
Libya	LIB
Cyprus	CYP
Syria	SYR
Lebanon	LEB
Israel	ISR
Egypt	EGP
Jordan	JOR
Other countries**	OC

* - All countries located on the territory of the former Yugoslavia

** - All countries covered by the calculation grid but not indicated in a given table (or figure)

1. EXECUTIVE SUMMARY

The present report prepared by the Meteorological Synthesizing Centre - East (MSC-E), Moscow of the ECE Co-operative Programme for the Monitoring and Evaluation of Long-range Transmission of Air Pollutants in Europe (EMEP) is dedicated to model evaluations of the Mediterranean Sea pollution by the airborne compounds of sulphur and nitrogen and by heavy metals. The Mediterranean Sea was considered both as the whole and as 10 separate sub-basins. In addition, the airborne pollution of neighbouring seas and countries was also considered.

Chapters 2-4 deal with the introduction of the problem, with the goals of the assessment, and a brief description of the MED POL Airborne Pollution Monitoring and Modelling Programme and related activities.

The basic elements of the modelling were considered in Chapters 5-7 including meteorological data for 1991, available emission data, principal processes of the atmospheric pollution transport and the description of calculation model used.

While compiling the complete meteodata set for 1991 a part of lacking meteoelements, such as data on precipitation in numerous points, were additionally modelled in MSC-E using the objective analysis method. The atmospheric mixing layer height was calculated as well. A detailed description and comparison of the meteoelements with the climatic ones is also presented.

Certain anomalies in the atmospheric circulation in 1991 were observed. In particular, the weather character was determined by the anticyclonic (not cyclonic) type of circulation during 50 days in winter of 1991.

Besides five principal atmospheric circulation patterns A, B, C, D and E (see Section 5.2), in the Mediterranean two special patterns were singled out in 1991. These are new types Ew (general duration 45 and 15 days in winter and autumn respectively) and El (32 days in summer). Their concise description is given in Section 5.2.

Emission data available within the EMEP grid were used for the present calculations. They include emission data for 1991 for sulphur and nitrogen compounds. The latest complete data set for heavy metals dated to 1982. The emission data for the countries and grid units within the calculation region located outside the EMEP grid were evaluated by MSC-E (see Chapter 6).

While modelling atmospheric processes, affecting the airborne pollutants, a compromise between a detailed description of these processes and necessary simplifications and generalizations was applied (see Chapter 7). Therefore, in order to simulate sulphur and nitrogen chemical transformations, the reactions, in which the above mentioned compounds are measured on the routine basis, were chosen along with volatile organic compounds (VOC) and ozone (see Section 7.1).

The non-linear dependence between the wash-out factors (coefficients), pollutants concentrations and precipitation amount was taken into account (see Section 7.2.2).

Newly introduced equivalent mixing layer height was calculated. Its value can be either higher or lower than that of Planetary Boundary Layer (PBL), depending on the meteorological situation. When PBL values were not available they were calculated (see Section 7.2.1).

For the simulation of heavy metals dry deposition, some mean value of dry deposition velocity was used for the whole mass inside a grid unit, instead of breaking the mass into several fractions. These averaged velocity was recalculated at each step (see Section 7.2.1.2).

While modelling horizontal transport the Eulerian scheme of calculations included a Lagrangian scheme element - a variable time step minimizing pseudodiffusion (see Section 7.3).

Chapters 8-10 contain the results of modelling of air pollution of the Mediterranean Sea and adjacent regions by sulphur, nitrogen and heavy metals.

In 1991 the deposition of sulphur compounds on the sea surface amounted to 1393 kt (as S), which is 7% of the total emission within the calculation region. Annual deposition density of these compounds was about 0.6 g S/m^2 per year; it is two times lower than the mean density in Europe or in the Baltic Sea. The main contribution to the airborne pollution of the Sea by sulphur compounds was made by Italy (30% of the total), Spain (17%), Greece (8%), Bulgaria (8%) and former Yugoslavia (7%). The input of the Mediterranean countries was 1046 kt. These countries produced 23% of the total emission within the grid and are responsible for 75% of the total deposition on the Sea. For remote countries, these figures are 77% and 25% respectively. Thus 1 kg of the total emissions in the Mediterranean countries results in the same deposition on the sea, as 10 kg of the total emissions in remote countries.

The mostly polluted region of the sea appears to be its northern part, except for the north-Levantin, especially the Adriatic, and the Aegean Seas. This is connected with the existence of zones, influenced by the main countries-emitters.

The adjacent seas (the Marmara, the Black and the Azov Seas) received 410 kt S in 1991. Together with the input of S to the Mediterranean Sea it makes up 9% of the total sulphur emission within the calculation area.

The Mediterranean countries received 2925 kt S, which is about 15% of the total emission within this calculated region. The bulk of the deposition was spread over the sea watershed, where from a part of it reaches the sea with the riverine run-off, increasing the input of originally airborne pollution to marine waters.

In the earlier works of the MSC-E it was shown, that the annual variations of S deposition on the Mediterranean are insignificant, the seasonal ones (even if averaged over 5 years) are great, and the presented report confirms this conclusion. The distinct minimum of depositions is observed in June, maximum - in December. The maximum exceeds the minimum three times.

The maps of the total S deposition and mean concentrations are given in Appendices A (annual) and C (seasonal), and maps of depositions from the main countries - emitters are in Appendix B.

The same Appendices contain similar data for the nitrogen compounds and heavy metals.

Depositions of sulphur compounds practically do not affect the content of these elements in the marine water (for the Mediterranean Sea in particular, since its salinity is sufficiently high contrary, for example, to the Baltic Sea) but nitrogen compounds entering the water can affect the marine biota causing eutrophication.

In 1991 the input of bound nitrogen (oxidised and reduced nitrogen in total) amounted to 1067 kt as N, which corresponds to 7% of its total emission. The deposition density of bound nitrogen over the Mediterranean Sea was 0.46 g N/m^2 per year. Similar to sulphur compounds it was two times lower than the deposition density over Europe and the Baltic Sea.

The main input to the pollution by bound nitrogen was made by Italy (29%), Greece (11%), France (11%), Spain (10%), Turkey (5%) and Germany (5%).

Germany being a remote country gives a good example of how the atmospheric

residence time affect a relative amount of pollutants entering the Mediterranean Sea. The deposition of "long-lived" oxidised nitrogen from Germany on the Sea exceeds 4% of its emission whilst similar value for "short-lived" reduced nitrogen is less than 1%.

The contribution to deposition from adjacent countries (recalculated to nitrogen) was the following: oxidised nitrogen - 397.6 kt, reduced nitrogen - 341.6 kt, bound nitrogen - 739.2 kt. The contributions of adjacent countries to the total deposition on the Mediterranean Sea was 61%, 82% and 69% respectively.

Thus the fraction of remote countries in total deposition of "long-lived" oxidised nitrogen (39%) is about 2 times bigger than similar fraction (18%) of "short-lived" reduced nitrogen and remote countries with emission amounting to about 75% of the total emission within the calculated region make a contribution to the total deposition of different nitrogen and sulphur compounds within the limits of 20%-40%.

Therefore, 1 kg of the Mediterranean countries emissions gives the same amount of deposition as 7 kg of the remote countries emissions, referring to bound nitrogen. The same values for oxidised and reduced nitrogen are 1:4.5 and 1:12.

The mostly polluted by bound nitrogen regions of the Mediterranean are: the Adriatic, the Tyrrhenian and the Aegean Seas. Similar to sulphur compounds, the reasons for that are a characteristic atmospheric transport pattern and presence of so-called "impact zones". These are areas constantly affected by neighbouring powerful emitters.

The adjacent seas together with the deposition on the Mediterranean Sea received about 1382 kt of bound nitrogen which made up 9% of total emission.

Seasonal variations of bound nitrogen depositions are considerably less than that of sulphur depositions, the ratio of the maximum and the minimum values do not exceed 1.5.

The depositions on the Mediterranean countries were estimated to be about 2500 kt of bound nitrogen, it is 16% of its total emission. This value is the upper limit of the bound nitrogen deposition on the Mediterranean watershed, considerable part of which is being washed with river flows into coastal waters of the Sea, increasing the pollution.

The deposition of heavy metals on the Mediterranean Sea in 1991 was about 7400 t of lead (Pb), 2500 t of zinc (Zn), 70 t of cadmium (Cd) and 200 t of arsenic (As), which make up 8%, 6%, 6%, 4% of their total emission within calculation grid correspondingly. The mean deposition densities of the metals over the sea surface were 3 mg Pb/m^2, 1 mg Zn/m^2, 30 ug Cd/m^2 and 80 ug As/m^2 per year, which are 2-3 times less than the densities over Europe.

The main countries-pollutants of the Sea were Italy and Spain for all metals, France - for Pb only, Bulgaria and Yugoslavia for Zn, Cd, As.

The Mediterranean countries with the emission not more than 32% of the total within the grid contributed from 64% for Cd to 85% for Pb to the total deposition on the Sea, while the remote countries with not less than 68% of the total emission gave 15%-36% of deposition. Therefore, 1 kg of Pb emission in the Mediterranean countries gives the same effect in deposition on the Sea as 13 kg emitted in remote countries. Similar ratio for Cd makes up 1:4.

The most polluted regions of the Mediterranean are: the Adriatic Sea - for all metals, the Tyrrhenian Sea - for Pb, the Aegean Sea - for Cd and As, the north - eastern part - for Zn, that can be due to the prevailing atmospheric transport pattern, emission distribution and "impact zones" location relative to the main countries-emitters.

The adjacent seas (the Marmara, the Azov and the Black Seas) received about 2% of the total emission of each of the metals in 1991. It made up summarily with the Mediterranean Sea 5.5% of As, 10% of Pb, and 8% both for Cd and Zn emission deposited

on the sea surfaces.

The Mediterranean countries received about 16%-17% of Pb, Zn and Cd emissions and about 10% of As emission as depositions. These figures are the upper estimate of the heavy metals depositions on the Mediterranean watershed, where from a part of them is being washed out to the sea with riverine run-off, increasing the pollution.

Four arbitrary points were chosen to follow seasonal variations of heavy metals deposition and concentrations over the sea: the Barcelona region (P1), northern Corsica (P2), eastern Crete (P3) and the Tel Aviv area (P4) (see Figure 3.3).

It was shown that the summer minimum of deposition was distinctly seen for all points, except for P3 and the concentration variations were considerable for all points.

Chapter 11 is dedicated to the comparison between the calculation results and measurement data. There is an appreciable deficiency of reliable measurements for sulphur and nitrogen in the Mediterranean. For heavy metals, the information is almost absent for 1991 except some data from the former Yugoslavia. Comparison for S and N compounds was made on the basis of six year calculation and measurement data sets (1987-1991). Calculation for 1991 and measurements for various years were taken for heavy metals. The results obtained show a good agreement of the calculated and measured values for S, N and Pb. The compliance for Zn, Cd, and As with measurement data available is worse. The comparison for heavy metals should not be considered as absolutely correct, because the data sets compared were for different years.

Chapter 12 contains a discussion of the calculation results for all the compounds taken together. Comparison of the MSC-E model results with the results of other authors and comparison of the pollution of the Mediterranean Sea via rivers and via the atmosphere was also made. Conclusions drawn from the present work and recommendations for the future research are given at the end of Chapter 12.

II. INTRODUCTION

The Mediterranean Sea has rather good selfpurification properties due to its rather big depth and appreciable water exchange with the Atlantic ocean [1]. Nevertheless the anthropogenic load on the Sea is increasing and this fact may lead to ecological disbalance.

There are two principal ways for the pollutants to reach the sea basin - with the air flows and with riverine runoff. According to [2.4] the atmospheric input of nitrogen to deposition on the Mediterranean is approximately equal to that via rivers. However, much of the river-borne nitrogen never reaches the open sea. A large fraction of it is being deposited in coastal and shelf sediments by biological transfer and some is re-emitted back to the atmosphere. It should be noted, that a part of the riverine nitrogen being a potential thread to coastal ecosystem eutrophication, arises from the atmospheric nitrogen deposition on the watershed of the Mediterranean. For the open sea, transport via the atmosphere is practically the only pathway of nitrogen.

Almost the same can by said referring to heavy metals. A comparison of atmospheric and riverine inputs of metals in particulate forms, (in which these substances are considered to be usually found in the environment), to the oceans as a whole, reveals that rivers are the dominant pathway. However, as for the open sea stretches, the atmospheric pathway is likely to dominate since much of the riverine input of particles to the sea is removed to the sediments in near-shore areas.

For dissolved heavy metals especially for Zn, Cd and Pb, the transport through the air is the major source and exceeds by many times the riverine input to the sea. For example, according to [3] a ratio between the atmospheric and riverine input to the western Mediterranean for lead equals to about 6.2.

Estimations provided by [4] indicate that about 98% of lead which eventually dissolves in sea water enter the global ocean via the atmosphere and that the most of dissolved cadmium and zinc are also primarily came from the atmosphere.

This is also the case in the Mediterranean where the atmosphere is a significant pathway for the transport and deposition of sulphur and nitrogen compounds, and heavy metals from the land sources into the marine environment and the atmospheric input dominates the riverine input for many species [5].

Evaluation of transport and deposition of the atmospheric pollutants can be made by means of measurements and computer simulation of these processes. Combination of both methods is necessary for a calibration of model parameters and verification of calculation results. Good examples of an experimental survey are the works accomplished for the Baltic Sea [6] with the usage of balance method. Unfortunately the Mediterranean region is far more poorly provided with the measurement data. There are only three EMEP stations located in the region with the representative data sets for sulphur and nitrogen compounds for several years. But even these stations are not located directly on the shoreline, so the data sets from the seasurface are practically absent.

The situation with heavy metal measurements is even worse. Fragmentary data sets for different years for the Mediterranean Sea are available in the works [7, 8, 9, 10,] and measurement data on Cd, Pb and Zn in precipitation in 1991 are available for three stations in the former Yugoslavia via [11]. More regular information was found in [6,12,14] from stations, located in the Baltic region. All the data are used for the model verification (see Chapter 11). More experimental data could be useful for the Mediterranean Sea region, especially continuous sets for long periods of time, because concentrations of some elements (Pb, for instance) can change more than 100 times during two days [13].

The second way to assess air pollution transport and deposition is a computer modelling of the processes.

Such a work appears to be the most important for the Mediterranean region, where the measurement network is not wide spread. Within the framework of EMEP the schemes of transboundary air pollutions transport in the scale of Europe and adjacent seas were developed with the results, well corresponding to the reliable measurement data from the continental and, especially the EMEP Baltic stations.

Among the works, dealing with heavy metals and published recently, surveys of T. Bartnicky et al. [14] and Y.A.van Jaarsveld [15], which are focused on continental Europe and the work of T. Alcamo et al.[16] considering trace metals input to the seas, adjacent to Europe, could be mentioned.

Among the works, where some regions of Mediterranean were considered, a trajectory analysis of pollutants transport, regardless of particular compounds [13,17] and almost the same, but for cadmium transport [18] should also be mentioned.

Therefore. to assess the transport. chemical transformation and deposition of pollutants in the Mediterranean (where only sparse experimental data are available), the integrated investigation of the area, covering the whole sea surface, adjacent seas (the Black and the Azov Seas), neighbouring countries and the sea watershed for the long period of time (not less than a year) is obviously necessary.

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III. SCOPE AND PURPOSES

MSC-E has gained an appreciable experience in the simulation of the long-range pollution transport within the scale of Europe and adjacent seas.

This report is dedicated to modelling of air pollution in the Mediterranean region with sulphur and nitrogen compounds and heavy metals in 1991. The objective is the determination of spatial and temporal distribution of the pollution over the Mediterranean Sea as well as contributions of individual countries - emitters.

The scope of work carried out for preparing the present report could be summarized as follows:

1. The whole region of the Mediterranean Sea including its eastern part not covered with the EMEP grid is considered. The region of calculations was formed by the shift of the EMEP grid by 5 squares (150x150 km^2) to the north-east (see Figure 3.1). For the comparison Figure 3.2 gives the EMEP calculation region.
2. The Mediterranean Sea is considered both as a whole and divided into 10 individual subbasins (see Figure 3.3). Table 3.1 slightly modified is taken from [1] and gives the names of these subbasins, and their notations. Among adjacent seas the Black Sea and the Seas of Marmara and of Azov are distinguished.
3. In order to make calculations for the whole region it was necessary to expand meteorological data base. The calculation of lacking meteoparameters was made by means of algorithm developed in MSC-E using real meteodata.
4. The available emission data within the geographical scope of the EMEP grid was also complemented. Estimates of emissions for countries of the North Africa and Middle East within the region of calculations both as national totals and distribution with the grid units were made by MSC-E.
5. Meteorological data set for 1991 in the calculated region were analysed and compared with climatic data.
6. Spatial and temporal distribution of pollution was calculated and its peculiarities were analysed in view of the emission and meteorological fields.
7. Total depositions on the Mediterranean Sea and its subbassins were calculated together with contributions of different countries. The input of the North African and Middle East countries were estimated for the first time for this calculated region.
8. Depositions on the Mediterranean countries including those of North Africa and Middle East were calculated.
9. Depositions on adjacent seas (the Marmara, the Black Sea and the Sea of Azov) were also calculated.
10. Total annual deposition, mean annual concentrations in air and precipitations were calculated and presented as maps.
11. Similar maps were made for seasons.
12. A comparison of calculated and measured data was made for the Mediterranean Sea and for the whole calculation region.
13. Provisional air pollution estimates were made for the Mediterranean watershed, where from a part of pollutants enters the sea with riverine runoff.
14. A comparison of the pollution input to the sea from the atmosphere and riverine runoff was made.
15. Recommendations for the refinement of the obtained estimates were set forth.

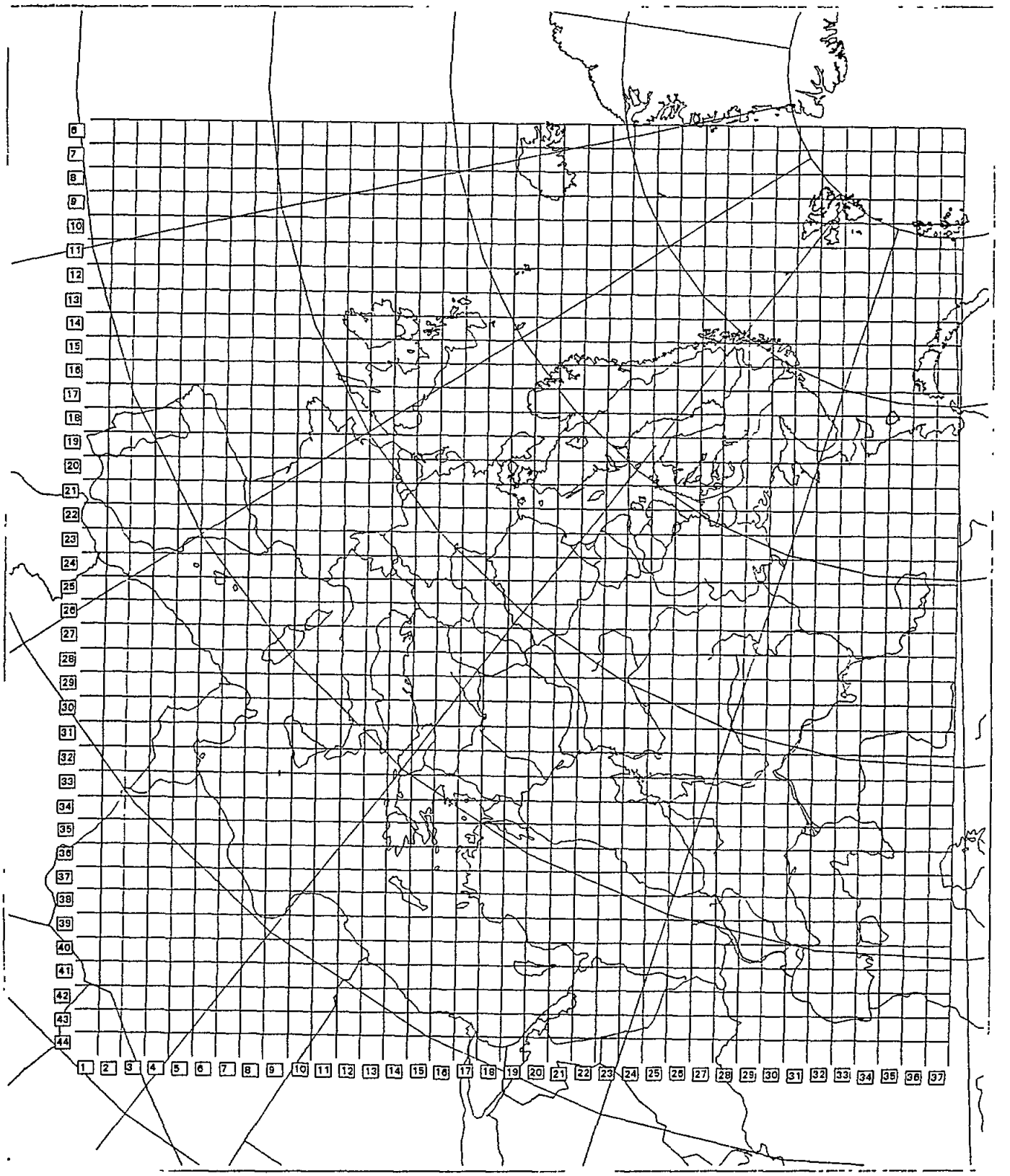

Figure 3.1. The region covered by calculations.

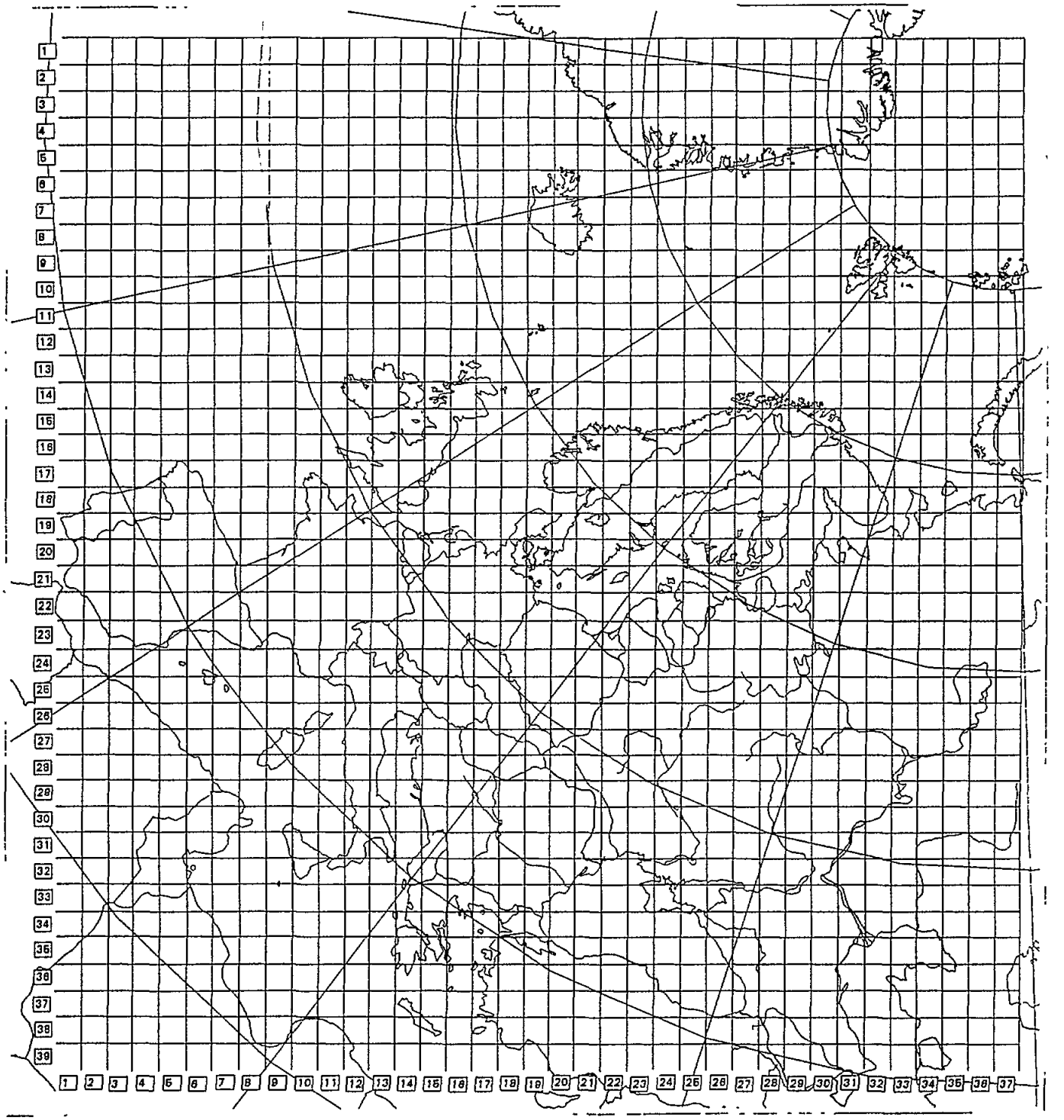

Figure 3.2. The EMEP grid. All previous calculations of MSC-E were made within the geographical scope of this grid.

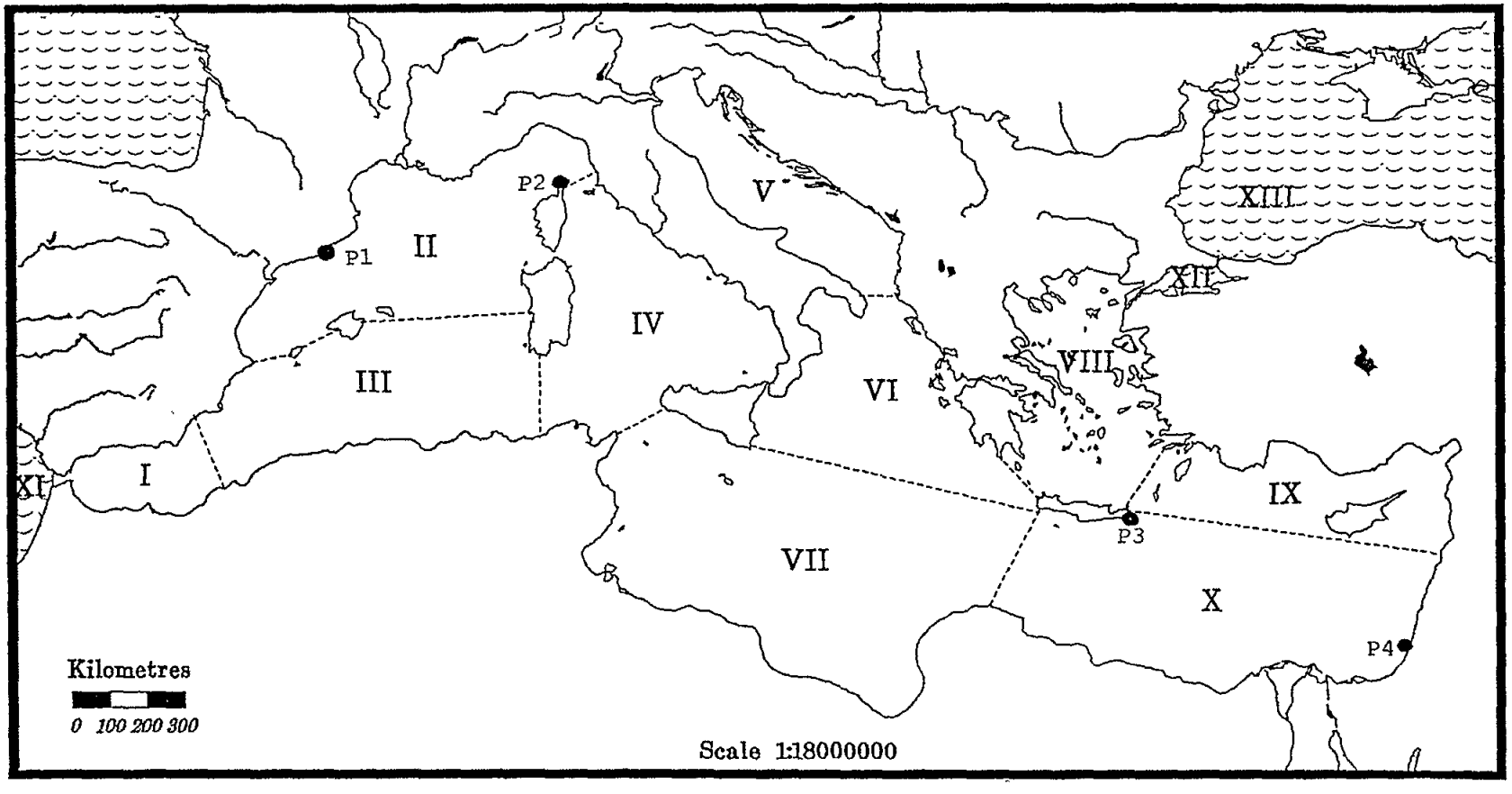

Fig.3.3. The Mediterranean Sea subbasins.

P1-P4 - Points chosen for the evaluation of seasonal variations of heavy metal deposition and concentrations

Table 3.1. Regional entities of the Mediterranean Sea

A. MEDITERRANEAN PROPER		
Number on the map	Notation	Regional sea
I	MT1	Alboran
II	MT2	North-Western
III	MT3	South-Western
IV	MT4	Tyrrhenian
V	MT5	Adriatic
VI	MT6	Ionian
VII	MT7	Central
VIII	MT8	Aegean
IX	MT9	North-Levantin
X	MT10	South-Levantin
B. ADJACENT AREAS		
XII	MAR	Sea of Marmara
XIII	BLC	Black Sea
	AZS	Sea of Azov

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IV THE MED POL AIRBORNE POLLUTION MONITORING AND MODELLING PROGRAMME

An important role in initiating airborne pollution studies in the Mediterranean belongs to the report on "Atmospheric Transport of Contaminants into the Mediterranean Sea" [1] prepared by the WMO-led GESAMP* Working Group on the Interchange of Pollutants between the Atmosphere and the Oceans. The report reviewed the knowledge on the levels of atmospheric contaminants over the Mediterranean, emissions, climatology pertinent to atmospheric transport and deposition. Despite a limited data base existing for the region it was possible to conclude that levels of atmospheric contamination over the Mediterranean were comparable to those over other European regional seas and atmospheric transport of some contaminants was at least comparable in magnitude to their riverine inputs onto the Mediterranean.

In November 1987 an airborne pollution monitoring and modelling programme was prepared during the WMO/UNEP Workshop on Airborne Pollution of the Mediterranean Sea held in Belgrade [2]. The Scientific and Technical Committee for Med POL agreed in May 1988 that this programme should be initiated within the framework of national MED POL monitoring programmes in as many countries as possible. It was also envisaged that financial support to the national monitoring programmes for purchasing sampling or analytical equipment could be provided subject to agreements signed by the National Co-ordinators for MED POL and the UNEP Co-ordinating Unit for the Mediterranean Action Plan located in Athens. This programme is also a part of the WMO Global Atmosphere Watch (GAW) and many meteorological services in the Mediterranean countries play an important role in implementing the programme.

The major goals of the programme [2] were identified as follows: to evaluate the importance of the atmospheric transport and deposition of land-based contaminants to coastal and open Mediterranean waters, to assess the airborne contamination levels of potentially harmful substances, to identify sources and source areas for these atmospheric contaminants, and to develop predictive models for assessing airborne pollution load.

For routine monitoring, the programme recommended by the Belgrade workshop includes measurements of the following parameters: in precipitation - pH, conductivity, SO_4, NH_4, NO_3, Na, K, Mg, Ca, Cl, Cd, Pb, Cu and Zn; in air - Cd, Pb and total particulate matter, and surface ozone. The emphasis on the heavy metals was made on the basis of previous research. At a later stage the monitoring programme should be complemented by measurements of radionuclides (^{137}Cs and transuranic elements) and organic species (PCBs, DDTs, HCHs, PAHs and other hydrocarbons). The Second WMO/UNEP Workshop on Airborne Pollution of the Mediterranean Sea held in Monaco in April 1991 [3] recommended that the priority measurement parameters should also include nutrients (N and P compounds) which may cause eutrophication, and black carbon as a good inert tracer for validating model calculations.

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* IMO/FAO/UNESCO/WMO/IAEA/UN/UNEP Joint Group of Experts on the Scientific Aspects of Marine Pollution

An expert meeting on the quality of measurements for the MED POL airborne pollution monitoring held in Ankara in 1994 [4] proposed that different measurement protocols be applied depending on the goals of the monitoring. Four such goals could be: assessment of total deposition of pollutants (primarily heavy metals) into the sea on monthly basis; assessment of long-term trends in concentrations of pollutants in air; assessment of wet versus dry depositions used in modelling, and assessment of wet and dry deposition and identification of the origin of the particulate matter. The corresponding measurement protocols are given in Table 4.1.

At present more than fifteen monitoring stations in the Mediterranean region (see Figure 4.1) conduct measurements which contribute to the MED POL airborne pollution monitoring programme. These measurements as a rule are still irregular and not continuous as required, but the first data have recently started entering the MED POL data centre and they are used for the assessment of deposition of some pollutants over the Mediterranean and its specific regions. Valuable contributions to the understanding of long-range transport and deposition of pollutants to the Mediterranean Sea were also made by a number of national research conducted at monitoring stations (e.g. in Corsica, Cap Ferrat, Mallorca) and during research cruises.

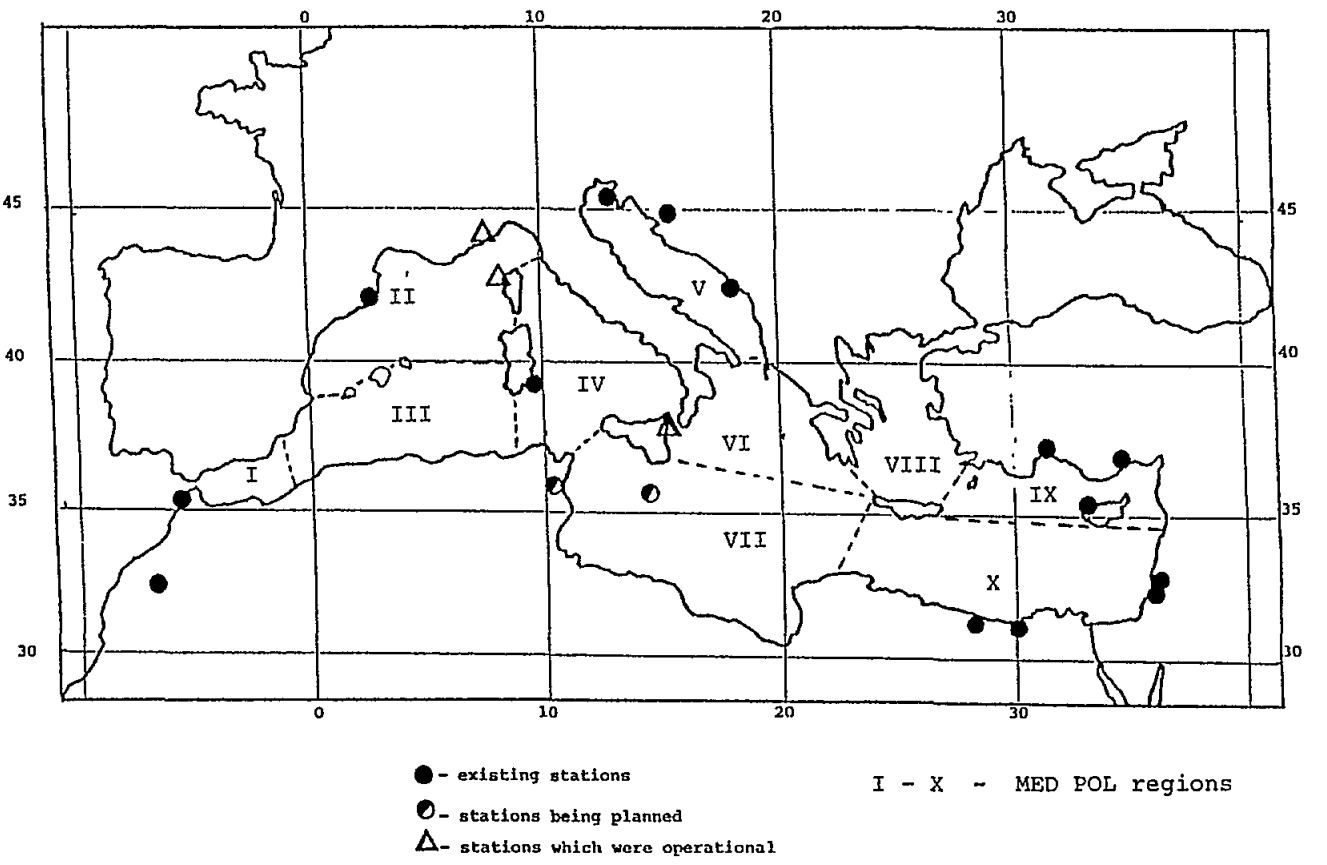

Figure 4.1. Locations of airborne pollution monitoring stations in the Mediterranean

Table 4.1. Levels of information required and associated sampling parameters in monitoring programmes [4]

Information Required	Approach	Sampling Duration‡	Sampling Frequency†	Supplementary Data Needed	Parameters of Interest	Sampler Type	Filter Type
Assessment of deposition on monthly basis	Bulk deposition sampling	2-weeks	Continuous	Local met. data	Cd, Pb,Cu, Zn, Al, Na	Bottle and funnel sampler	None
Assessment of long-term trends in concentration over long periods	Aerosol measurements	1-week (or for 3 days every week)	Continuous	Local met data	Cd, Pb, Cu, Zn, Al, Na	Hi-Vol sampler at reduced flow rate	GFF¶
Assessment of wet vs dry deposition	Simultaneous wet-only and bulk sampling, or wet and dry sampling ¶	2-weeks	Continuous	Local met data	Cd, Pb, Cu, Zn, Al, Na	Wet-only + Bottle and funnel, or wet and dry sampler	None
Assessment of wet and dry deposition and identification of the origin of the particulate matter	Simultaneous wet-only + bulk (or wet and dry) deposition and aerosol sampling	aerosols 1-3 days (preferably 1-day) wet only 1-day or event Bulk 1-week	Continuous	Size separated concentrations Back trajectories	Cd, Pb, Cu, Zn, Al, Na id. (plus major ions) id.	Wet-only + Bottle and funnel (or wet and dry) sampler Hi-Vol sampler	W-41¶

‡Sampling duration, refers to the time intervals in which samplers remain operational
¶Glass fibre filter
¶Dry and wet sampling refers to separate collection of wet-only and dry deposition samples

†Sampling frequency refers to the frequency of sampling in one year
¶Whatman-41 cellulose filter

The available measurement data and model calculations based on meteorological information have shown that a significant part of pollutants entering the Mediterranean Sea is transported via the atmosphere from both coastal and remote land-based sources. The evidence of the long-range transport of cadmium to the Mediterranean was confirmed by the calculations of 36-hours forward and backward trajectories correspondingly from some major cadmium sources in Europe and to some receptor points in the Mediterranean [3]. This study was conducted as a MED POL research project co-ordinated by WMO and supported from the Mediterranean Trust Fund. It was shown, for example, that about 32% of trajectories from Salzburg, Austria in winter reach the Mediterranean.

For the purposes of the MED POL airborne pollution monitoring programme, a detailed meteorological and climatological data review for the model assessment of atmospheric transport and deposition of pollutants in the Mediterranean basin was prepared under another research project in 1989 [5].

The measurement data on concentrations of some trace elements in air and precipitation over the Mediterranean show that the concentration levels are comparable to those over other European regional seas and much higher than over the open ocean areas (see Table 4.2.) [6]. Continuous measurements of aerosol compositions carried out in Corsica [3] allowed to reveal that there is a seasonal pattern for the elements of continental origin, either natural (e.g. Al, Si) or anthropogenic (e.g. S, Pb). This pattern is inversely related to the frequency and amount of rainfall such that the highest concentrations are observed between May and October. It was also shown that concentration changes of two orders of magnitude occurred on a daily time scale and were connected with precipitation patterns. This factor is of particular importance for developing sampling protocols.

The available data on concentrations of compounds or elements other than heavy metals are more scarce and limited to the western area. Concentrations of some synthetic organics (HCB, HCHs, DDTs and PCBs) in air over the Mediterranean are given in Table 4.3 [6]. One can see that these concentrations are close to those over the Baltic Sea and higher than over the North Atlantic. Concentration of the total aromatic hydrocarbons measured in Monaco ranged from 6 to 147 ng/m^3 (average 40.5) [3].

The assessment of the total deposition of trace elements to the north-western Mediterranean [7] confirms that the atmospheric input of these substances into the Mediterranean Sea is significant and for some heavy metals it represents 4-20% of their total anthropogenic emissions in Europe (see Table 4.4). It is worth noting that for the global ocean the atmospheric inputs of Fe, P, Cu, Ni and As are approximately equal to their inputs from rivers (see Table 4.5), and for Zn, Cd and Pb (soluble forms) the atmospheric inputs exceed by many times the riverine inputs of these elements. For the organochlorine compounds the calculated atmospheric input into the north western Mediterranean Sea is given in Table 4.6. As it was estimated in [7] for the global ocean the atmospheric inputs of these substances constitute 85-99% of their total (atmospheric + riverine) inputs.

There are only a few studies available on the quantities of petroleum hydrocarbons entering the marine environment through atmospheric fall-out. However, it can be expected that considerable quantities arrive into the Mediterranean Sea via the atmosphere since many countries in the basin are heavily industrialized and consequently burn large amounts of oil.

Table 4.2. Concentrations of some trace elements in air and precipitation over various regions

Region	Pb	Cd	Zn	P	Cu
AIR	*ng/m³*				
W. Medit. (cruises)	39	1.9	26	-	2.5
Messina, It.	153	0.19	-	-	-
Corsica	22	0.86	26	13.7	3.2
Cap Ferrat, France	34.2	0.29	26.4	-	4.58
Monaco	76.5	0.64	-	-	-
Beni-Mellal, Morocco	10	0.3	63	-	4.7
Cent. Medit.	8.8	0.04	11.3	-	-
N. North Sea	23	0.3	24	-	7
S. North Sea	83	2	80	-	10
Baltic Sea	10-60	0.1-0.5	23.6-250	-	-
Bermuda	3.3	0.2	-	-	-
Samoa	0.02	0.002	-	-	-
Precipitation	*ug/l*				
Cap Ferrat, France	4.6	0.18	8.6	-	2.15
Medit. Sea	6-12	-	-	-	-
Tangier, Morocco	12	0	13	-	2.5
N.North Sea	4-11	0.27-0.7	13-15	-	2.3
S. North Sea	10-29	0.5-9.5	26-490	-	2.5-77
Baltic Sea	5.8	-	16-38	-	-
Bermuda	0.77	0.006	-	-	-
Samoa	0.007	0	-	-	-

Table 4.3. Concentrations of synthetic organics (hexachlorobenzene, hexachlorocyclohexanes,dichlordiphenyl trichlormethyl, methanes and polychlorinated biphenyls) in air over various regions

Region	HCB	HCHs	DDTs	PCBs
AIR		*pg/m³*		
Mediterranean	100-200	288	22	540
Aspvreten, Sweden	67	897	15	341
Baltic Sea	291	553	-	-
North Sea	125	130	-	-
North Atlantic	115	362	11	74
Samoa	55	34	1.9	11

Table 4.4. Total atmospheric deposition of trace elements to the north-western Mediterranean (GESAMP, 1989)

	Al	Cd	Cu	Fe	Mn	P	Pb	Si	V	Zn	As
Total Deposition 10^9 g yr^{-1}	435	0.5	2.1	360	11	16	14.5	1535	12.5	17	0.5
% of European man-made emissions	-	19	11	-	-	-	12	-	4	21	7

Table 4.5. Global deposition of metals to the ocean (10^9g yr^{-1}) (GESAMP, 1989)

	Pb	Cd	Cu	Ni	Zn	As	Fe	P
Atmospheric input								
Dissolved	79	1 9-3.3	14-45	8-11	33-173	2.3-5.0	$3.2x10^3$	315
Particulate	9	0.4-0.7	2-7	14-17	11-55	1.3-2 9	$28x10^3$	630
Riverine input								
Dissolved	2	0.3	10	11	6	10	$1.1x10^3$	
								300
Particulate	1600	15	1500	1400	3900	80	$110x10^3$	

Table 4.6. Calculated total atmospheric inputs (in 10^6 g yr^{-1}) of organic pollutants to the North Sea, the Baltic Sea and the Mediterranean Sea

	HCHs	*DDTs*	*PCBs*	*Dieldrin*	*Chlordane*	*HCB*
North Sea	28	0.9	3.7	1.1	0.1	0.5
Baltic Sea	20.5	0.7	2.8	0.9	0 1	0.4
NW Mediterranean Sea	9 9	0.3	1.7	0.6	0 05	0 2

In a report on the state of pollution of the Mediterranean Sea by petroleum hydrocarbons it was estimated that 35,000 tons of these hydrocarbons (5.5% of the total input) enter the sea via the atmosphere [8].

On the basis of available measurement results and model calculations it can be concluded that the atmospheric input of many pollutants is significant in comparison with their riverine inputs. These conclusions are important not only from the scientific point of view, but also for the development of strategies, policies and legal instruments to protect the environment of the Mediterranean region. This means that to the control of airborne pollution of the sea should be given the same attention as has been the case of the control of pollution through rivers, direct effluent discharges or waste dumping.

The first step in this direction was made in October 1991 when the Contracting Parties to the Barcelona Convention adopted an annex to the Protocol on the Protection of the Mediterranean Sea against Pollution from Land-Based Sources which defined the conditions under which this Protocol should apply to pollution through the atmosphere. These conditions are the following:

- the discharge substance is or could be transported to the Protocol Area under prevailing meteorological conditions;

- the input of the substance into the Protocol Area or its subregions is hazardous for the environment in relation to the quantities of the same substance reaching the Area by other means.

To continue the work on airborne pollution assessment and pollution control the following actions were recommended for 1994-1995: compilation of information on existing legislative measures regarding the control of emissions of harmful substances into the atmosphere from various groups of sources; initiation of collection and dissemination of information on existing air pollution control technologies (starting with heavy metals and acidifying compounds); identification and categorization of the most important groups of emission sources (starting with heavy metals) and preparation of general recommendations for control; preparation of guidelines for inventory of emissions of other important pollution (e.g. organic species); reconsideration of the most important problems with regard to airborne pollution of the Mediterranean Sea and preparation of a plan for future actions.

As far as the future research and monitoring activities are concerned, the Workshop in Monaco recommended in 1991 that they should focus on the following: establishment of new monitoring stations in countries where they do not yet exist; improvement and standardization of measurement techniques; monitoring and assessment of atmospheric deposition of nutrients; improvement of data quality by means of conducting intercalibration and intercomparison exercises, and conducting of training courses and workshops to ensure the proper qualification of the monitoring station personnel and to improve data utilization.

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V. GEOGRAPHY AND CLIMATOLOGY OF THE MEDITERRANEAN REGION

5.1. Geographical characteristics

This section as well as Table 5.1. contains information collected and published within the Mediterranean Action Plan (MAP) [1].

5.1.1. Geographical location.

The Mediterranean Sea lies (46°N, 30°N, 6°W and 36°E) between Europe, Asia and Africa and without the Black Sea covers about 2.5 million km^2, with an average depth of about 1.5 km and a volume of 3.7 million km^3.

About 30% of the surface area and 50% of the total volume is contained between a 2 and 3 km depth contour. In contrast, the area with a shallower than 200 m contour constitutes more than 20% of the total area of the Mediterranean Sea but contains less than 1.5% of the total volume [5]).

The Mediterranean Sea consists of series of interacting parts and adjacent seas, with two major basins, western and eastern. In the western Mediterranean (about 0.85 million km^2) such parts are the Alboran Sea, the Algero-Provençal basin, the Ligurian Sea and the Tyrrhenian Sea. In the eastern Mediterranean (about 1.65 million km^2) such parts are the Adriatic Sea, the Ionian Sea, the Aegean Sea and the Levant (Figure 3.3).

The Mediterranean Sea is connected with (and separated from) Atlantic by the Strait of Gibraltar (15 km wide and 290 m deep), with the Sea of Marmara by the Dardanelles (between 450 m and 7.4 km wide and 55 m deep) and with the Red Sea by the Suez Canal (120 m wide and 12 m deep).

The maximum length of the Mediterranean Sea from Gibraltar to Syria is about 3,800 km and the maximum distance in the north-south direction from France to Algeria about 900 km, yet one is never further than 370 km from the coast, and most often, considerably less, with more than half of the Mediterranean Sea being less than 100 km from coast.

5.1.2. Circulation and water balance of the Mediterranean Sea.

The surface current system of the Mediterranean shows a migration of Atlantic water, with salinity slightly above 36, towards the east with numerous spin-off eddies along the way [6]. The annual thermal changes of surface waters are very large and control the density of surface waters and the basic characteristics of the annual biocycle. There is no surface return system from the east to the west.

The return of Mediterranean water is by way of Levantine intermediate water and Mediterranean deep water flowing from east to west and spilling over the sill of the Gibraltar into the deep Atlantic.

The estimated turnover time for Mediterranean waters is 80 years. The basic nature of the Mediterranean circulation system contains components of strong vertical convections which determine the distribution of salinity and produce vertical recycling of nutrients and other dissolved substances [6].

The Mediterranean Sea has a deficient hydrological balance, with loss through evaporation exceeding the input of water through run-off and precipitation. This deficiency is mainly compensated by the flow of Atlantic surface waters through the Strait of Gibraltar.

On the input side of the water balance are the net inflow through the Strait of Gibraltar, the net inflow through the Dardanelles, river run-off and precipitation. On the

negative side of the balance there is evaporation.

The precipitation and run-off contribution to the water balance is exceeded by evaporation by about 1 m yr-1. Since the Gibraltar constitutes the only access for the renewal of sea water, lost through evaporation, the increasing salinity of the Mediterranean water is due to the evaporation of the lower salinity water introduced from the Atlantic [6].

Table 5.1. Mediterranean watershed area and flows (updated 1987) [1].

	Area (1000 km²)		Theoretical resources (km³/year)				Stable resources (km³/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 Med. region	505	180	109.9 28.3	109.9 28.2	89.4 19.8	20.4 8.4	17.0 6.3	3-4 1.6
FRANCE Med. region	550	130	185.0 74.0	170.0 62.0	70.0 31.0	100.0 31.0	85.0 35.0	1.1 0.2
ITALY Med. region	300	300	187.0	185.0	155.0	30.0	18.5	12.0
MALTA	0.3	0.3	0.03	0.03		0.03	0.007	0.023
YUGOSLAVIA Med. region	256	80	265.0 77.5	150.0 77.5	130.0 62.0	20 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 Med. region	780	195	181.0 77.0	166.0 70.0	156.4 **	9.4 **	** 14.5	** 1.1
CYPRUS	9.3	9.3	0.9	0.9	0.6	0.3	0.2	0.01
SYRIA Med. region	185	22	35.4 4.4	7.6 3.4	3.1 1.3	4.5 2.1	14.0 2.1	0.2 0.2
LEBANON Med. region	10.4	9.8	4.8 4.0	4.8 4.0	1.3 1.0	3.5 3.0	2.1 1.9	0.9 0.9
ISRAEL Med. region	21	12	1.0	1.0	0.2	0.8	0.2	0.08
EGYPT Med. region	1000	144	57.5* .3	1.0 0.8	0.5 0.3	0.5 0.5	55.5 55.5	0.3 0.3
LIBYA Med. region	1760	250	0.6 0.6	0.6 0.6	0.1 0.1	0.5 0.5	0.1 0.1	0.1 0.1
TUNISIA Med. region	164	90	4.35 3.0	3.75 2.5	2.13 1.8	1.62 0.8	1.0 0.8	0.7 0.7
ALGERIA Med. region	2380	133	16.0 13.2	15.7 13.2	13.6 12.0	3.3 1.2	** 1.8	** 0.7
MOROCCO Med. region	780	80	29.0 4.0	29.0 4.0	20.0 3.0	9.0 1.0	2.5 0.6	0.9 0.2
MED. TOTAL	8792	1796.4	**	497.0	427.4	**	157.5	25.4

(*) Regular flow Former natural flow = 86 km³/year, former low-water flow = 24 km³/year

(**) Data are absent

5.2. Review of the atmospheric circulation peculiarities in the Mediterranean region in 1991.

The MSC-E model requires following meteorological parameters: baric topography at the levels of 1000 hPa and 850 hPa, windspeed at the same levels. surface temperature, cloudiness characteristics (amount), precipitation amount, mixing layer height.

These characteristics were estimated on the real meteodata basis in the process of the objective analysis developed in the MSC-E (see Chapter 7), or on the basis of the short-term forecast, when there was a shortage of the meteodata available. In particular, the mixing layer height was always calculated (see Chapter 7).

Basic climatological features of the region are described in [3]. There is sufficient bibliography in the same work. In the present chapter peculiarities of the atmospheric circulation and meteodata in general in 1991 are analysed.

5.2.1. Characteristics of synoptic situations.

For the description of evolution of baric situations in the Mediterranean, classification by P.Hess and H.Brezowsky developed in 1969 [2] and later modified is traditionally used.

The classification is based, generally, upon location of anticyclones and its impact on development and localization of principal systems of lower pressure - cyclones and depressions in the region limited by the central Atlantic (40°W) in the west, by the Ural mountains in the east (60°E), by the Polar circle in the north (about 67°N) and by the southern limit of Sahara desert in the south (10°S).

P.Hess and H.Brezowsky singled out five main types of the atmospheric circulation: A, B, C, D and E. The following descriptions as well as Figures 5.1-5.5 are taken from [3].

Type A: Blocking anticyclone or ridge stays over the north-eastern Atlantic and the British Isles. To the east from the block in the mid-troposphere (level 500 hPa) quasistationary depression exists with axis directed across the central Mediterranean. Existence of this depression leads to cyclogenesis and deepening of already existing cyclones in the region of the Genoa Bay. Then cyclones begin gradually move eastward (Figure 5.1).

Type B: Anticyclone stays over northern Europe. Low pressure system covers the most area of the Mediterranean (Figure 5.2).

Type C: The western type. Deep depression or series of depressions prevail in the mid-altitude over Europe. Western winds prevail over the Mediterranean (Figure 5.3).

Type D: The eastern type. Anticyclone stays over central and southern Europe, resulting in eastern winds over the most of the Mediterranean. Low pressure system stays over northern Europe (Figure 5.4).

Type E: The anticyclonic type. Anticyclone or ridge pressure system stays over the most of the Mediterranean, which leads generally to prevailing of low western winds in the north, eastern winds in the south and northern winds in the east of the region (Figure 5.5).

The types A,B and C are characteristic of the Mediterranean winter. The types D and E are the most frequent in summer. They prevail almost continuously in July and August, but from November to April occur very seldom.

For classification of synoptic situations in 1991, daily synoptic maps at 00h Greenwich time produced at the Meteorological Centre of Russian Federation were studied. As a result, periods of the circulation types (A-E) were estimated together with their duration for every month (Table 5.2). The same was done for all four seasons (Table 5.3).

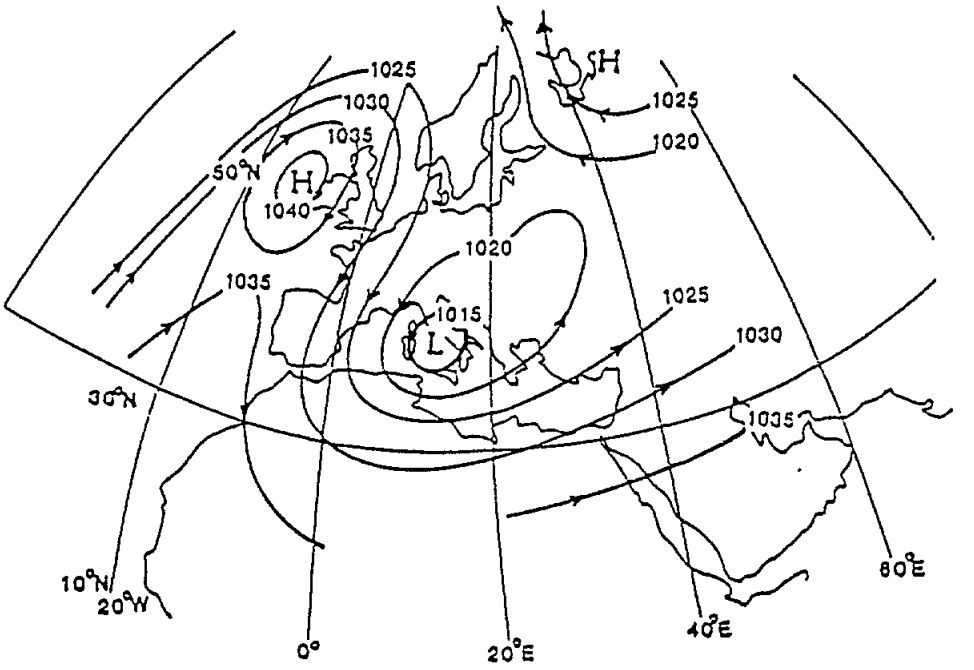

Fig.5.1. The atmospheric circulation type A

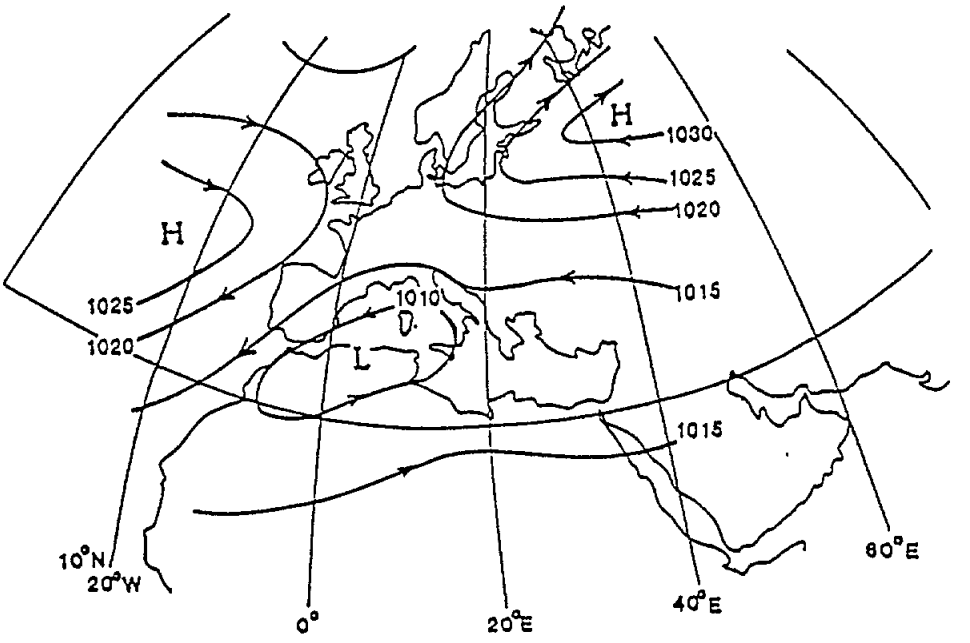

Fig.5.2. The atmospheric circulation type B

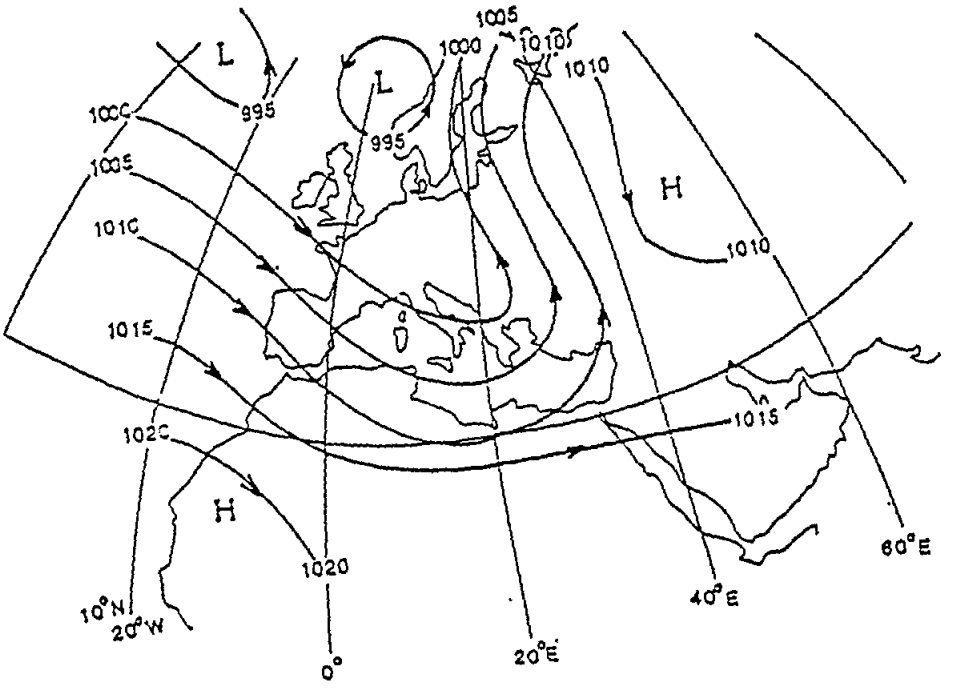

Fig.5.3. The atmospheric circulation type C

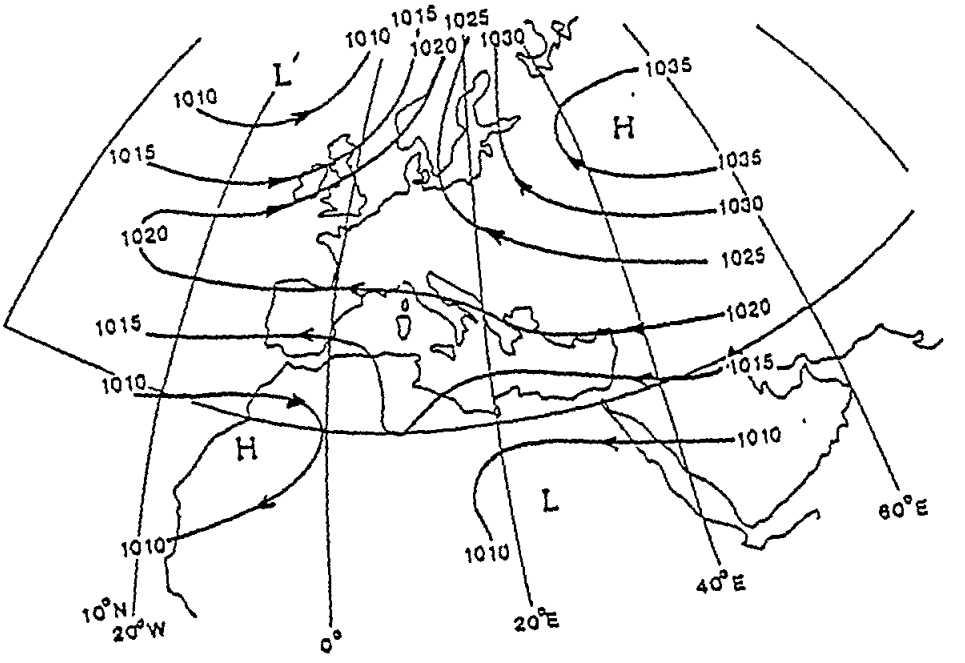

Fig.5.4. The atmospheric circulation type D

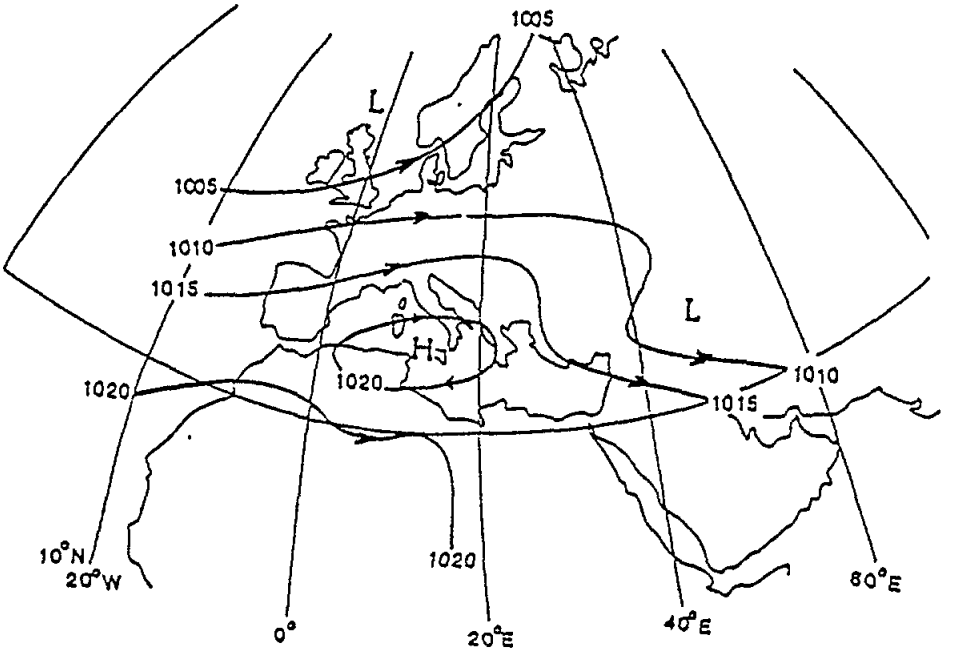

Fig.5.5. The atmospheric circulation type E

Table 5.2. The types of the atmospheric circulation in the Mediterranean in 1991 and its distribution with months (in days).

Month / type	J	F	M	A	M	J	J	A	S	O	N	D
A	3	4	2			3				5		
B		13	22	20	14					12	16	3
C	5		4	5					5			3
Ew	15	7								5	10	23
D				4								
E	5				16	21	14	20	18	5		
E1						6	16	10	4			
Restructuring	3	4	3	1	1	-	1	1	3	4	4	2

Table 5.3. The types of the atmospheric circulation in the Mediterranean in 1991 and their distribution with seasons (in days).

	Type							
Season	A	B	C	Ew	D	E	E1	Restructuring
Winter	7	16	8	45	-	5	-	9
Spring	2	56	9	-	4	16	-	5
Summer	3	-	-	-	-	55	32	2
Autumn	5	28	5	15	-	23	4	11

The study shows that the atmospheric circulation in the Mediterranean in 1991 had a number of anomalies.

Let us consider them seasonally. The winter types A, B, C with a vast area of low pressure over the Mediterranean sea basin stay for 31 days in winter. But the prevailing one was a new type of circulation, which resembles the summer type E and therefore called Ew (E-winter) (Figure 5.6).

For the type Ew a strong blocking anticyclone located over the whole Europe with the center in Austria and Hungary is typical. Low pressure occurs only in the north and in the east of the region, where fast Atlantic cyclones move along the edge of anticyclone bringing short time periods of precipitation. Clear sunny weather with low north-eastern winds stays over the Mediterranean. The Ew type stayed for 45 days in winter of 1991, including 23 days in December. For 5 days in January the summer type E occurred, when a ridge of the high pressure system (a ridge of the Azores anticyclone) came to the Mediterranean. It took 9 days for restructuring one type of weather to another. Consequently, the atmospheric circulation in the Mediterranean in winter of 1991 can be considered as abnormal. For 50 days out of 90 weather character was defined by anticyclonic baric systems, instead of cyclonic ones, which influenced greatly windfield, cloudiness and precipitation patterns of a given season.

The winter type B circulation was observed for 56 days out of 90 in spring of 1991. From March to May the number of days with this type of weather was decreasing (Table 5.2). The types A and C occurred only in March and April. The summer types E(16 days) and D(4 days) appeared in May. The circulation restructuring took 5 days. Therefore

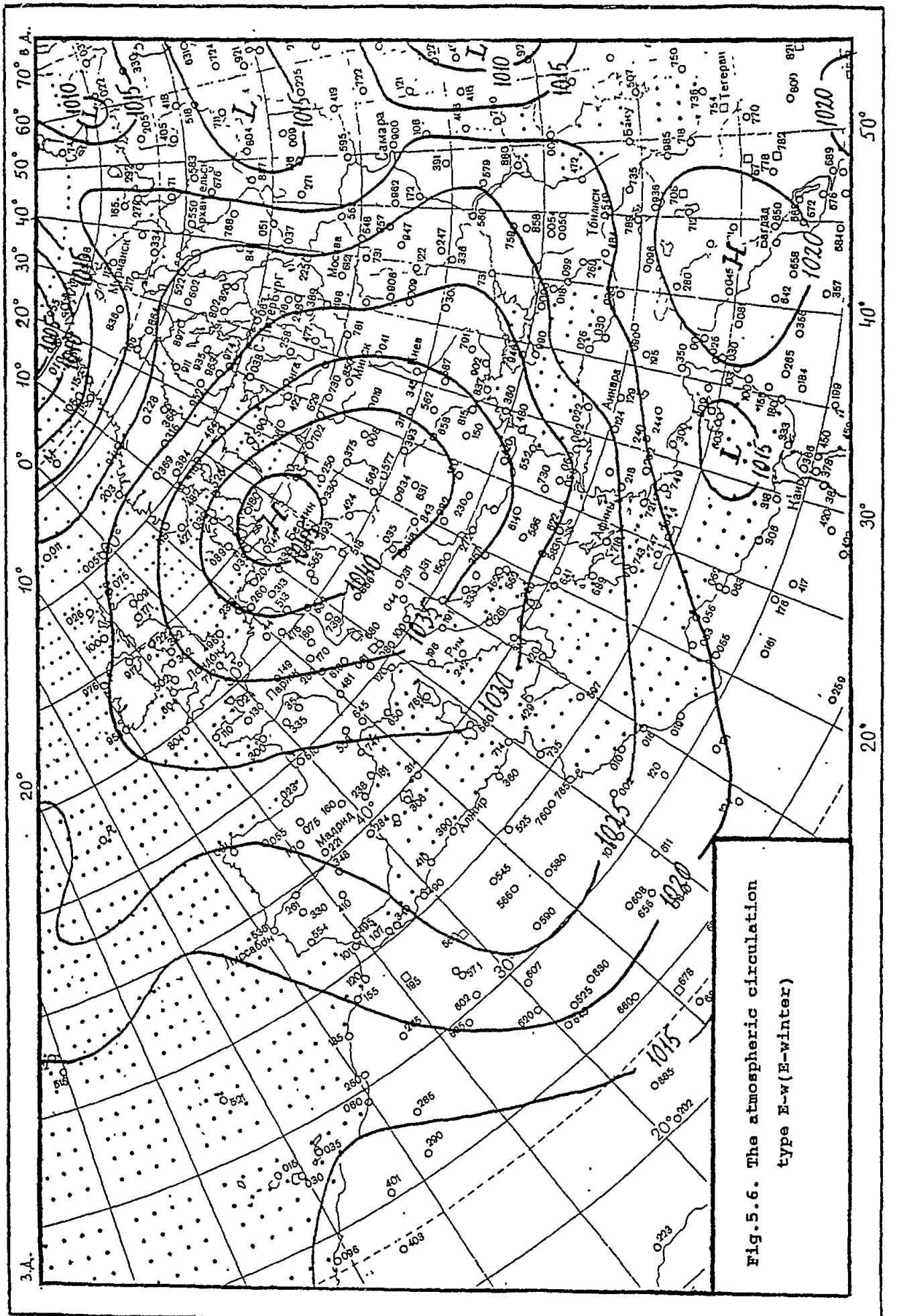
Fig.5.6. The atmospheric circulation
type E-w(E-winter)

in spring of 1991, 67 days of cloudy, rainy cyclonic weather and 20 days of typically anticyclonic summer warm weather with clear sky were observed.

In summer of 1991 the type E prevailed for 55 days out of 92, when anticyclone or high pressure ridge stayed over the Mediterranean. It caused weak northern winds in the north, eastern winds in the south and northern winds in the east. For 3 days in June the winter type A was observed when a deep cyclone sat in with centre over the Tyrrhenian Sea and a strong anticyclone - over the eastern Atlantic. The restructuring took 2 days. The remaining thirty two days (including 16 in July) were characterized by similar synoptic situations which could not be identified with the above mentioned types. The baric field of this new type slightly resembled the type E, moved to the north, thus the northern Mediterranean was under low pressure system (Figure 5.7). Therefore the new type was called El(E-low).

The type El was characterized by a strong blocking anticyclone over north Europe with the center over north-western Russia. Between this blocking anticyclone and the Azores anticyclone cold cyclones moved to the Mediterranean from the North Atlantic bringing into the northern part of the region low pressure areas with strong winds quickly changing their direction with each new passing cyclone. It caused a type of weather with massive frontal cloudiness and precipitation amount exceeding monthly average untypical of summer. In the southern part of the Mediterranean the influence of Atlantic cyclones were noticeably soften by the Azores anticyclone . The areas of high pressure broke off recurrently the Azores anticyclone and moved eastward between 30° and 40° N.

In autumn the atmospheric processes in the region were unstable, particularly in September and October (Tables 5.2, 5.3). All the types of circulation except for D were observed. The reconstruction of the atmospheric circulation takes 11 days in total, which is the longest period compared to other seasons of the year.

In September the summer type E still prevailed (18 days). During this period a high pressure ridge of the Azores anticyclone stayed over the Mediterranean. Other 12 days the region was under low pressure systems (the types El and C). In October high pressure systems were observed during 10 days (the types E and EW) and low pressure systems - during 17 days (the A and B); in November - 10 days (the type Ew) and 16 days (the type B) correspondingly. Thus, number of days with clear, dry weather were approximately equal to those with cloudy and rainy weather in the autumn of 1991.

As a conclusion one can note, that the atmospheric circulation in 1991 over the Mediterranean was close to a normal one only in autumn. In winter dry anticyclonic weather prevailed over wet cyclonic one, in spring situation was quite opposite with low pressure weather types prevailing and, finally, in summer cold cyclones from the north-western Atlantic occurred more frequently then ever. All these peculiarities can be found in mean monthly surface pressure fields (Appendix D, Figures 1-12).

5.2.2. Surface wind pattern and surface pressure.

For the analysis of surface wind patterns in the Mediterranean in 1991 mean monthly maps at 00h Greenwich time were studied (Appendix D, Figures 1-12). The climatic data were taken from [3, Figures 7-18].

Directions and speeds of surface winds greatly differed from the climatic ones in winter of 1991. This is in a consequence of the fact that in December and January the region was influenced by the southern periphery of a strong blocking anticyclone with a centre over central Europe. Therefore the north-eastern and eastern winds prevailed instead of the western and north-western ones. Wind speed was lower than usually. The exceptions were

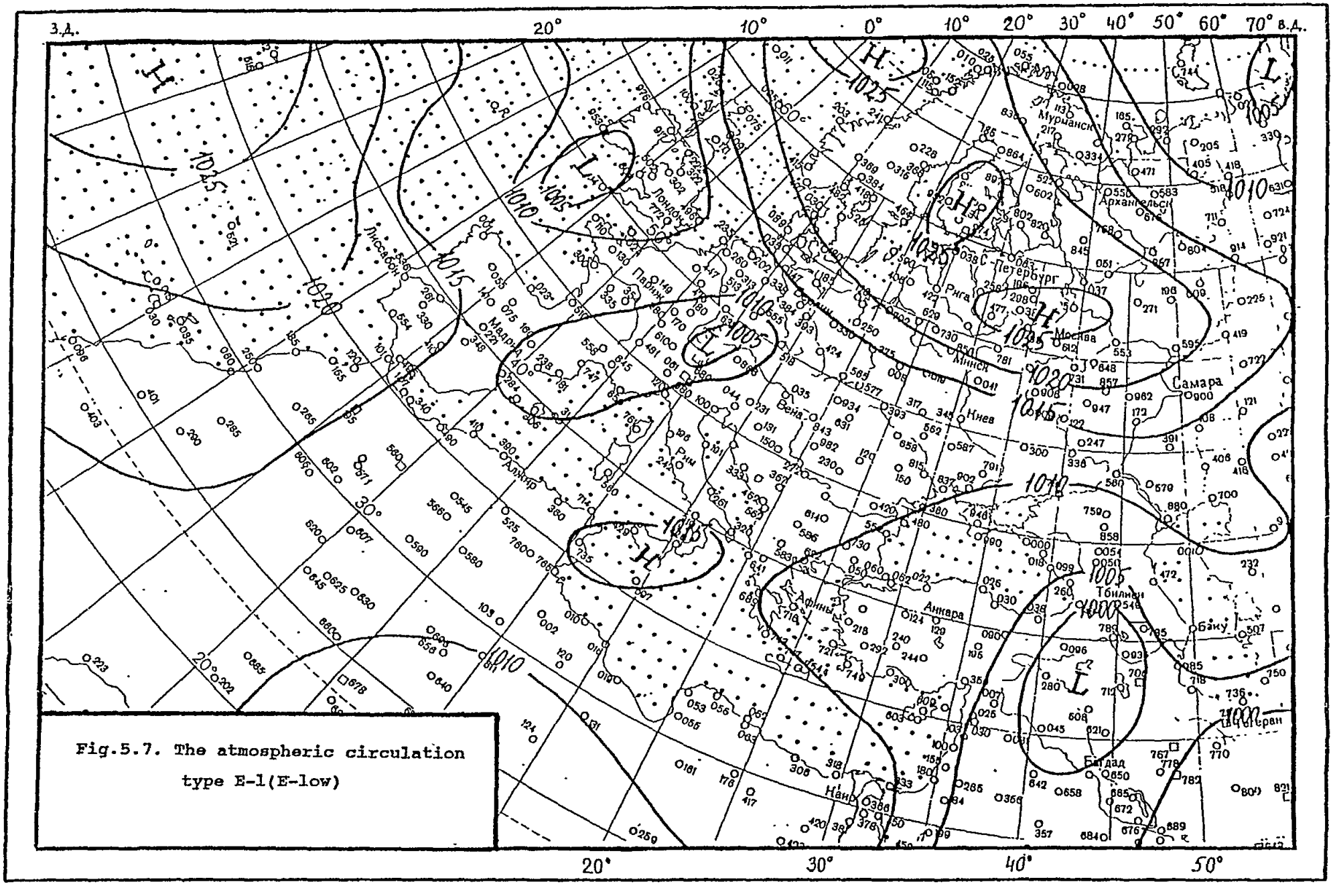

Fig.5.7. The atmospheric circulation type E-l(E-low)

the eastern part of the region near the Syrian coast with the western winds and the western part near the Morocco coast with the southern wind in December. In February, while the type B and A circulations prevailed, the windfield pattern was practically normal.

In spring the windfield was also close to normal. In February, while the type B and A circulations prevailed, the windfield pattern was practically normal.

In spring the windfield was close to normal one too. In March the whole region was under the winds of the western quarter, and only Peloponnesus and the Aegean Sea were mostly under the southern winds, the region to the north from the Pyrenees Peninsula was under the eastern winds.

The windfield pattern, in general, remained the same in April, but wind speed was lower. In May both the summer type E and the winter type B were observed, each of them took 50% of the month. So the windfield in May was very complicated, different from the climatic one. The eastern winds prevailed over the Mediterranean, excluding the Asia Minor, Pyrenees and Apennines Peninsulas and adjacent seas, where the western winds were prevailing.

The summer windfield pattern had a number of peculiarities. In June, during the type E circulation, the pattern practically did not differ from the one with the western winds over the north Mediterranean, the eastern winds over the south and the northern winds over the east. Wind speed seldom exceeded 2.5 m/sec.

In July, when a ridge of the Azores anticyclone was shifted to the west for 16 days, and the northern part of the Mediterranean was under the North Atlantic cyclones influence, a disordered windfield pattern with frequent changes of directions and speeds took place. Wind speed exceeded monthly average by 3-7 m/sec. The rest of July (14 days) the wind field remained normal.

Thus, the July wind pattern was mainly of the eastern direction over northern Sahara and the northern Mediterranean, of the northern direction over the east, the centre and the far west of the region and of the southern direction - over the Balearic Islands.

In August, directions and speeds of surface winds did not differ much from those of the climatic and June winds.

In autumn the windfield pattern was very different in every three months. The type E prevailed in September, therefore the windfield was similar to June and August ones, but windspeeds were even lower. That also corresponded very well to the climatic winds pattern. In October frequent restructurings of circulation occurred with changes of windfields, but after monthly averaging these fluctuations disappeared. It resulted in mostly eastern winds over the southern Mediterranean. Over the rest of the region winds were so weak, that the directions could not be distinguished. That poorly corresponds to the climatic windfield pattern.

The winter type B set up in November with characteristic windfield. The north-western and the western winds prevailed, which conformed to the yearly average situation, and only during 10-day period of the Ew type of circulation the windfield was oriented to the eastern direction.

As a conclusion it can be noted, that the character of the windfield pattern in 1991 differed considerably from that of climatic either when the atmospheric circulation was anomalous (December, January, July), or when restructuring was frequent (May, October).

5.2.3. Windfield pattern at the level of 850 hPa.

Maps of baric topography AZ 850 hPa of the Northern Hemisphere at 00h Greenwich time with wind direction vectors marked on them were studied (Appendix D,

Figures 13-24). The climatic data were taken from [3, Figures 19-30].

The greatest discrepancies of the real wind at the level of 850 hPa from the normal wind in 1991 were observed basically in months with anomalous circulation.

In December winds were eastern over the Gibraltar region and northern over the rest of the Mediterranean. Only over Syria and Asia Minor winds were mostly southern. Normally, the north-western winds with the maximum intensity over the west of African coast were observed this month during the observation period.

In January winds remained northern over the eastern part of the region and turned western over the western Mediterranean.

In February, March and April the windfield patterns were very similar to the climatic ones, except for the central Mediterranean which was under the influence of winds of the southern quarter in March.

Steady western fluxes predominated over the whole region in May, even over the north-western coast of the Sea, where eastern winds usually prevailed this time of year. The exception was the region adjacent to Pyrenees with the northern winds instead of the southern ones.

The western winds prevailed over the Mediterranean in June, and the north-western winds over the Syrian coast. In July the windfield was slightly different from the multiannual average, except for the Gibraltar region with the south-western winds instead of the south-eastern ones. The windfield almost alike the climatic one remained in August either.

The only peculiarity in the windfield pattern was observed in September and October, which was the south-western winds instead of south-eastern in the west of the region in September, and the north-western ones in October. Steady fluxes of the western quarter predominated over the Mediterranean in November. But the maximum windspeed was observed over the Pyrenees, Alps and the Tyrrhenian Sea, not over the northern coast of Africa as usual.

It should be noted, that the mean monthly values of wind speed were practically the same as the climatic ones throughout the year. Generally speaking, variations of the real wind compared with the climatic wind at the level 850 hPa were much less than variations of the surface wind.

5.2.4. Cloudiness over the Mediterranean in 1991.

The most remarkable feature of the Mediterranean climate is almost complete absence of cloudiness in summer. In winter mean cloud amount is about 5 over the whole water basin. From November to February the cloud amount is 5-6 over the central Mediterranean including Italy, Greece and the Aegean Sea, and 3-4 over the eastern Spain and Egypt.

In spring cloudiness gradually retreats to the north, therefore cloud amount of 5 is being observed only over the north of Italy and Greece in May. In summer over the north of Italy the maximum cloud amount exceeds 4, while the rest of the Mediterranean is almost cloudless. In September cloudiness starts to increase, but insignificantly, until October. In October cloud amount is 5 over Italy and Sicily, but still no clouds are observed over eastern Spain and Egypt. In November cloudiness is increased to the amount of winter one.

It is known that cloudiness has daily variations. This is the most characteristic of the Mediterranean winter. Unfortunately, cloudiness monitoring over the water surface is being carried out exclusively from ships. So the observations are irregular and therefore not very precise, especially averaged over the month.

Ship observations from February till December, analysed at the Hydrometeorologic Centre of Russian Federation, were studied for investigation of cloudiness fields over the Mediterranean in 1991 (Appendix D, Figures 25-35). Data for January 1991 could not be obtained. All the observations inside 5-degree gridcell were averaged over the month and this value was given to the point with coordinates calculated as the mean arithmetic of coordinates of ships provided the data in this gridcell. The climatic data on cloudiness were taken from [3, Figures 39-49].

In February two maxima of cloudiness were observed. The first one was over Sicily, Corsica and southern Italy and the second - over the south of Greece. Their location corresponded well to the climatic field. Cloud amount was 5-6, similar to the climatic data.

In March the cloudiness decreased over the eastern part of the Mediterranean (to the east of 15°E) and increased over the western part. It was caused by the fact, that while the circulation type B prevailed over the region, the western part was affected by a stationary low pressure system and the eastern part - by a ridge of the Siberian anticyclone.

In April the geographical pattern of cloud field was close to the climatic one, but the general background was slightly higher than ever, though lower than that in March. The maximum cloudiness was still observed over the western part of the sea (Algerian coast) and in the far east of the region.

In May the amount of cloudiness was still gradually decreasing everywhere, except for the south-eastern part of the region, where it even exceeded 5.5. Thus the region was under the influence of a general depression, which could be clearly seen on the map of the May surface pressure (Appendix D, Figure 5).

Cloudiness over the rest of the Mediterranean was similar to the normal one.

In June the cloudiness field was similar to the normal one either. Average amount of cloud did not exceed 3.5-4. The only exception was the southern coast of France, where cloud amount made up 5.5-6. It could be explained firstly by the fact that during 3 days cloudiness of 8-10 sat in with a deep depression while the circulation type A occurred, and, secondly that the majority of observations were carried out exactly during these 3 days.

July was almost cloudless over the southern part of the Mediterranean - no more than 1.5-3. But over the north-eastern Mediterranean the cloud amount exceeded the normal one by 0.5-1.5 on the average. It resulted from the fact that during 16 days in July circulation of the El type was observed. Enhanced cloudiness followed the way of north Atlantic cyclones to quasi-stationary depression over Saudi Arabia.

In August the cloudiness pattern was practically normal.

In September the amount of cloudiness increased by 0.5-1.0 over the central and the western parts of the region comparing with August and climatic ones. It was caused by a deep depression while the type C circulation prevailed during 5 days. Over the rest of the Mediterranean the amount of cloudiness was normal.

In October the cloud amount exceeded the climatic ones everywhere especially over Sicily and Algerian coast. Cloud amount was up to 6.3 over these regions. The reason for that was frequent restructurings while the winter types of circulation prevailed.

The general background of cloudiness was similar to the climatic one in November everywhere, except for the north-eastern part of the region. It was caused by the south-western periphery of a blocking anticyclone with the type Ew circulation during 10 days.

A blocking anticyclone (type Ew) was located over the western part of the Mediterranean in December. The result was that the cloud amount was less than usual by 1-2. But in the east, where a low pressure system stayed, mean monthly cloud amount exceeded normal ones by 0.5-1.5.

Therefore in 1991 seasonal variations according to climatic ones were clearly seen. They were disturbed only in the months with abnormal atmospheric circulation.

5.2.5. Mean monthly amount and geographical distribution of precipitation.

Another remarkable feature of the Mediterranean climate is presence of two distinguished seasons - a "wet" one - from late autumn till early spring, and a "dry" summer season. This is not true only for the northern part of the Adriatic and the Ligurian Seas where frequent heavy summer showers bring up to 25% of annual precipitation. In the northern Mediterranean summer rains makes up 10% of a yearly amount, down south from 40°N - no more than 5%. In the south-eastern Mediterranean there is practically no precipitation at all in the summer season.

As the beginning of the "wet" season depends on gradual movement of the polar air masses down south, the "wet" season starts in the northern Mediterranean in October, and in November in the region to the south from the Gibraltar latitude. Further southward the maximum of precipitation is observed in December and somewhere even in January.

To analyse the precipitation fields in the Mediterranean, maps of mean monthly precipitation amounts compiled at the Hydrometeorological Centre of Russian Federation were used (Appendix D, Figures 36-47). Unfortunately, the quality of the used data is rather poor. Moreover, the discrete nature of precipitation fields rules out any possibility of interpolation between points of observations. But still, such interpolation in some cases appears to be the only way to obtain information in the regions where the meteorological observations are irregular.

The climatic data for precipitation pattern were taken from meteorological maps [4], because in the work [3] there were mean monthly precipitation fields only for January and June.

In January 1991 the geographical distribution of precipitation differed from the climatic one. The principle difference was that the northern limit of the main precipitation zone, fixed along the isoline 50 mm/month was shifted to the south by 3-5° of latitude, which made the zone itself more narrow. It was caused by a stationary blocking anticyclone over the Mediterranean for 15 days (type Ew). During this period of time, there was deficiency of precipitation everywhere, except for the eastern part of the region. Rains occurred occasionally, when cyclonic waves passed by over the central Mediterranean, bringing in damp air masses from the Atlantic.

Other 5 days, when a ridge of the Azores anticyclone stayed over the western and the central Mediterranean, together with clear, dry weather, in the east of the region - over the Aegean Sea, Asia Minor and the Syrian coast heavy rainfalls were observed.

The biggest amount of rainfall occurred during 11 days, when a low pressure system prevailed over the Mediterranean with the types of circulation A and C. During this period the zone of rainfall was localized according to the climatic standard and rainfall intensity exceeded a normal one. As a result, monthly amount of precipitation observed was similar to the climatic one.

In February the precipitation field slightly differed from climatic, due to prevailing circulation of winter types A and B. The maximum of rainfall over the Libyan coast could be singled out. This zone originated at the end of deep cyclones passing along the eastern periphery of a blocking anticyclone over eastern Atlantic during 4 days with the circulation type A. Over the Libyan coast, in the southern part of a deep high-altitude depression adjoining the block from the east, cyclones became quasistationary, which resulted in heavier rainfalls than ever.

The circulation type B prevailed in the Mediterranean in March. A Siberian anticyclone was very strong this month, and its western periphery reached the Adriatic coast. Thus, while in the western and the central parts of the region the geographical distribution and the amount of precipitation were close to the normal ones. In the eastern part of western Greece especially, deficiency of precipitation was observed.

In April the precipitation field was determined by the types B and C in their classic form and practically did not differ from the monthly average.

In the first half of May the winter type B prevailed in the region, in the second half - the summer type E. As a result, a ridge of an Azores anticyclone was observed even in the field of mean monthly surface pressure in May (Appendix D, Figure 5) over the southern, western and north-western part of the Mediterranean. Within this ridge zone, the geographical distribution and amounts of precipitation were close to normal ones. The eastern and the north-western part of the region were under vast depression. Precipitation amounts in some places exceeded the climatic values by 50-100 mm.

In June the summer type E circulations were not observed at all or were insignificant. During 3 days only, when a deep depression with an axis along 5°E (type A) meridian intruded into the central part of the region, heavy rainfalls were observed over the Balearic Islands and northern Italy particularly. In general, precipitation field was close to the climatic one.

The atmospheric circulation in July was defined by both type E (14 days) and type El (16 days). Therefore a belt of heavier rainfalls was formed along the path of the North Atlantic cyclones from Britain to the northern coast of the Asia Minor Peninsula while the type El prevailed. There were practically no rainfalls over the rest of the region.

In August the precipitation pattern corresponded to the climatic one completely.

The anticyclone type E still prevailed in September but during 5 days, during the type C circulation, a deep depression was formed over the center of the region along with cloudy, rainy weather, resulting in monthly precipitation amount higher than usually by 25-50 mm.

October was characterized by prevailing of the winter circulation types B and A. Therefore the amount of precipitation was higher than normal by 20-30 mm all over the Mediterranean. It should be noted, there were no observation data over the south of the region.

The winter type B still prevailed in November, resulting in higher precipitation amounts over the central Mediterranean by 20-30 mm. But the type Ew stayed in during 10 days. The centre of a blocking anticyclone was shifted by 15-20° to the east, and there was a deficiency of precipitation in the north-eastern part of the region.

In December the Mediterranean, excluding the eastern part, was under a strong blocking anticyclone (type Ew). Rainfalls were practically absent. Rainfalls occurred during 6 days only with the circulation types B and C, but the monthly amount appeared to be by 30-50 mm lower comparing to the monthly average. On the other hand, in the east of the region, under deep altitude depression, active cyclones caused the amount of precipitation 4-6 times higher than ever. Therefore, in December, both geographical distribution and monthly amount were quite different from the climatic ones.

Thus, the analysis of synoptic situations, surface and 850 hPa windfield patterns, cloudiness and precipitation fields allowed to sort out the principle types of the atmospheric circulation over the Mediterranean in 1991 and to estimate their duration. It turned out that there were periods of time when all the above mentioned meteoelements were close to the climatic ones. On the other hand, serious anomalies of these meteoelements took place during

some months. In particular, December, January, May, July and October should be singled out. The character and possible reasons for these anomalies are analysed in details in this chapter. More detailed analysis of the atmospheric circulation parameters and the estimation of their impact on the air pollution transport requires special methods.

5.2.6. The main atmospheric transport patterns

Monthly averaged wind speed components are hardly representative in respect to transport directions. For example, while a cyclone moves along certain trajectory line (Figure 5.8), intensive air masses exchange is going in both directions across the line, especially that the cycloids, along which air pollutants are being transported across that line almost perpendicularly. After monthly averaging of the wind components one will get, as a rule, almost complete absence of the transport. "Averaged wind" trajectory will be stretched along the line. Therefore, assessment of mutual transport will be greatly underestimated, because the transport according to "the averaged wind" will take place along the line, not across it.

Climatologically, transport can be characterized with a set of direct and reverse trajectories. The results of monthly averaged 5-10 day reverse trajectories for 10 years for the western part of the Mediterranean and similar data for Israel for 5 years are given in the work [3].

It is shown [3, Figures 32-35], that climatologically characteristic directions of transport are: for the western point - primarily from W to NW, for the eastern point (except for December) from NW to N.

We did not calculate the trajectories, but the analysis of depositions from separate countries for the calculated year (see Chapters 8-10) shows, that the climatological direction of transport remained the same in 1991 (see also Appendix B).

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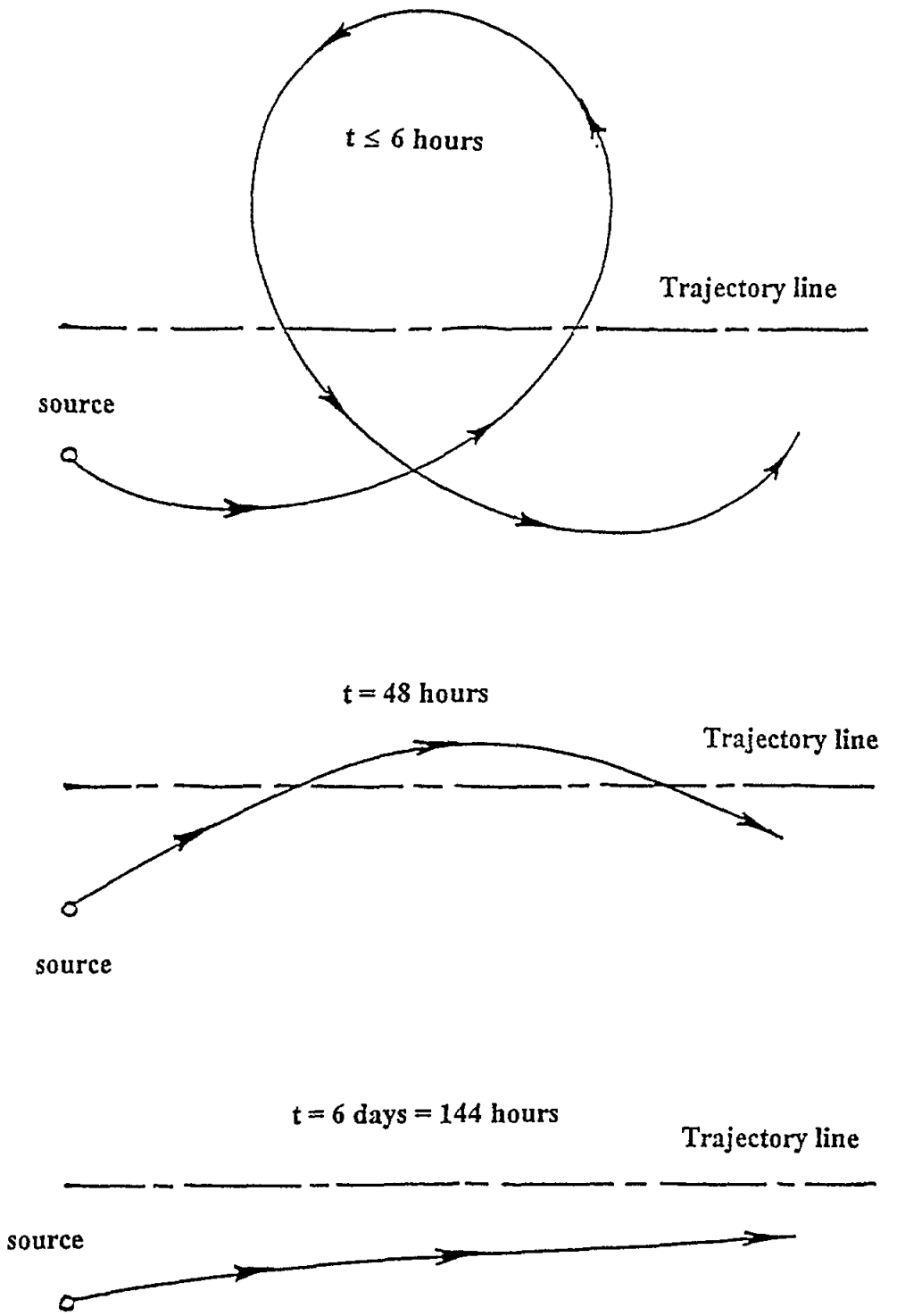

Fig. 5.8. Calculated trajectories of a particle (plume) moving along some trajectory line in a cyclone for different periods of averaging.

VI. EMISSION

Pollution of the atmosphere by sulphur and nitrogen oxides, ammonia and heavy metals results from the human activities such as mining, different kinds of industry, agriculture, transportation and waste incineration.

6.1. Main sources of emission

6.1.1. 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.

According to the CORINAIR inventory [1], about 66% of the total SO_2 is emitted by power plants and non-industrial (domestic) combustion, 19% by industrial combustion, 7% by oil refineries, 3% from road transport and the remaining 5% from various production processes, many of which also include some use of fossil fuel.

About 55% of SO_2 emission in the CORINAIR inventory 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. Second important industrial source of the SO_2 emission are both ferrous and non-ferrous metallurgies.

6.1.2. 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 a less important, yet 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 inventory indicated that 93% of the total NO_x emission is emitted from fuel combustion (including 54% from road transport, 24% from power plants and 6% from non-industrial combustion), 2% from oil refineries and 5% from production processes (the latter 2 categories being also dependent on the use of fuel to some extent).

6.1.3. Ammonia

According to CORINAIR inventory, 80% of the atmospheric ammonia (NH_3) emission are produced from the decomposition of livestock wastes from domestic animals. About 10% of NH_3 atmospheric input take place due to nitric fertilizers application. The rest of NH_3 emission originates from other sources, mostly from industrial nitric fertilizers production.

6.1.4. Heavy metals

Most heavy metals are toxic to plants and animals, but the toxicity depends on the concentration. Many metals play a key role in different life processes, and a small but continuous supply is necessary for most living organisms.

All heavy metals occur naturally at various concentration levels in air, soil, water and living organisms. Since the beginning of industrialization the concentration levels of many metals in nature increased considerably. Metal-containing particulate matters are emitted from different kinds of industries and subject to long-range transport via air before their effects could be recognized in the ecosystems. However, a significant part of the atmospheric heavy metal pollution results from burning of coal and oil, which was increasing

continuously in the XXth century. During the last decades the contribution from waste incineration and automobile traffic also increased the heavy metals emissions.

Air pollution by heavy metals from local and distant sources impacts the environment both as dry and wet deposition. Large particles deposit close to emission sources, while smaller particles in the form of aerosols can be transported hundreds of kilometers in the atmosphere.

Four metals were chosen for their transboundary transport assessment - As, Pb, Cd, Zn in the present report. The atmospheric pollution by these species takes place primarily due to the emission from non-ferrous metallurgy enterprises and fossil fuel combustion at power plants [2]. These powerful, as a rule, point sources are relatively few in number but they are of very significant importance in transboundary air pollution. The Table 6.1 shows the distribution of heavy metals emissions from various categories of sources [2].

Table 6.1. Atmospheric emissions of heavy metals from various categories of sources, in %

HEAVY METALS	POWER PLANTS	WASTE INCINERATION	FERROUS METALLURGY	MON-FERROUS METALLURGY	ROAD TRAFFIC	OTHER
Zn	5	6	23	57	-	9
As	7	8	4	74	-	7
Cd	13	15	5	65	-	2
Pb	2	2	4	15	76	1

In another study [3] the following sources and effects of heavy metal pollution are also mentioned:

A. Arsenic

Arsenic is emitted to the atmosphere mainly from coal combustion and more locally from mining industry. Minor emission sources are glass industries and the use of arsenic in pesticides and wood preservatives. Arsenic is moderately toxic to plants, but highly toxic and cancerogenic to mammals.

B. Cadmium

Cadmium is emitted to the environment as a result of the use of phosphate fertilizers in agricultural areas and through the emission from waste incineration and combustion of fossil fuels. Other emission sources are mining and metal industry especially as a by-product of zinc refinement. Cadmium is moderately toxic to all organisms, but is a cumulative poison. In mammals, cadmium is accumulated in kidneys and liver. Since acceptable daily intake is very low for humans, a restrictive legislation now controls the use of cadmium.

C. Lead

Lead is a local pollutant near mines and industries and along roads originating from the combustion of leaded gasoline. Long-range atmospheric transport from densely populated areas of Europe plays an important role for the background concentrations in remote areas. Lead is very toxic to all plants, but is usually tightly bound in the organic top soil layers. It is moderately toxic to mammals, where it acts as a cumulative poison; children

seem to be especially vulnerable.

D. Zinc

Zinc is mainly emitted by the metal industry. Zinc is essential to all organisms as a constituent of many metalloenzymes and of several proteins and zinc deficiency may in some cases be a problem. At higher concentration it is moderately toxic to plants, but only slightly toxic to mammals.

6.2. Emission data

Data on sulphur and nitrogen oxides and ammonia emission from considered countries for 1991 are given in Table 6.2. The same table contains the abbreviations used for countries names, groups of countries and individual water basins. The emission data for Europe used in the calculations were based on official information submitted to the Secretariat of the UN Economic Commission for Europe (ECE).

Emission estimations made by MSC-W/Co-ordinating Chemical Centre (CCC) [4] when official data are absent are taken into square brackets.

Emission estimations made by MSC-E are taken into parentheses.

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 MSC-W/CCC for the north Africa (Algeria, Tunis, Libya) [4]. 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 [5].

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 [5,6,7,8].

Emission from the sea area is taken from international shipping [4].

Figures 6.1-6.3 give maps of sulphur, and oxidized and reduced nitrogen emission intensities for each square of the calculated region, Figure 6.4 shows the emission seasonal variations in conventional units.

Slightly altered data on the national totals for the heavy metals emission of the countries located in European part of the EMEP grid were taken from [2], the same is true for the gridded emission for the same territory. Data on the national totals are given in Table 6.3, Figures 6.5-6.8 and show the gridded values, used in the calculations.

Assessments of the national totals and gridded emissions for North African and Middle East countries were made at MSC-E. These figures are taken into parentheses in Table 6.4. Estimates of heavy metals emissions per capita and population in the countries with similar economic development level were used for the calculations.

All the above mentioned estimations of the heavy metal emissions are referred to 1982, because the only complete set of information for all four considered metals was found in [2] for this particular year.

It must be noted that the heavy metal emissions for As, Cd and Zn are presumably rather underestimated. Only sufficiently powerful point sources, amounting to 1424 in Europe [2] were taken into consideration. Therefore a number of activities, such as open mining or phosphate fertilizers application, greatly contributing in some areas to the pollution of the atmosphere by As and Cd [3] were not quantitatively assessed. With regard to the Zn emission, in work [9] the total emissions of western and central European countries are estimated to be nearly 2 times greater than that in work [2], and this difference reaches the

Table 6.2

Sulphur and nitrogen compounds emission in 1991.

EMITTERS	NOTATIONS	Emission, kt per year: Sulphur dioxide as S	Emission, kt per year: Nitrogen as N	Emission, kt per year: Ammonia as N
Albania	AL	(55)	(12)	[25]
Austria	AT	49	64	[70]
Belgium	BE	207	96	[91]
Bulgaria	BG	633	47	[104]
Czech, Slovak Rep.	CS	1222	300	[167]
Denmark	DK	92	86	99
Finland	FI	130	88	35
France	FR	685	551	[636]
Germany	GE	2870	983	1062
Greece	GR	250	227	[64]
Hungary	HU	505	72	140
Iceland	IS	3	4	[2]
Ireland	IE	95	40	[138]
Italy	IT	1090	536	[338]
Luxemburg	LU	8	6	[5]
Netherlands	NL	101	167	193
Sweden and Norway	SK	109	193	81
Poland	PL	1498	367	418
Portugal	PT	102	37	[58]
Romania	RO	900	[119]	[305]
Spain	ES	1158	255	[282]
Switzerland	CH	31	53	50
Turkey	TR	(200)	(61)	(395)
United Kingdom	GB	1887	831	(385)
Yugoslavia**	YU	740	128	[174]
Russia*,Turkmenistan, Kazakhstan,Uzbekistan	RF	2233	800	[1386]
The Ukraine	UR	1391	334	[702]
Moldova	MOL	82	15	[40]
Armenia, Georgia, Azerbaijan	ZKV	119	71	[111]
Belarus	BR	281	79	[189]
Estonia,Latvia,Lithuania	PBL	196	47	[135]
Morocco*	MOR	[16]	(2)	[3]
Algeria*	ALG	[120]	[15]	(82)
Tunisia*	TUN	[60]	[6]	(38)
Libya*	LIB	[60]	[7]	(34)
Malta	MAL	(2)	(1)	(1)
Cyprus	CYP	(5)	(2)	(2)
Syria	SYR	(80)	(9)	(58)
Lebanon	LEB	(25)	(3)	(9)
Israel*	ISR	(60)	(24)	(16)
Egypt*	EGP	(125)	(15)	(33)
Jordan*	JOR	(15)	(6)	(8)
North Sea, English Channel (La Manche)	NOS	[87]	[58]	
Baltic Sea	BLT	[36]	[24]	
Strait of Gibraltar	GBR	[6]	[4]	
Atlantic Ocean	ATL	[149]	[96]	
Total		19638	6941	8164

* - within the calculation area

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

Table 6.3 **Heavy metals emission in 1982 (tons/year).**

Emitters	Notations	As	Pb	Cd	Zn
Albania	AL	16.5	170	1.10	37.0
Austria	AT	25.0	1031	5.80	175.0
Belgium	BE	85.0	2090	13.00	697.0
Bulgaria	BG	150.0	1500	64.90	1724.0
Czech Slovak	CS	94.0	1376	24.25	756.0
Denmark	DK	9.5	573	5.90	120.0
Finland	FI	106.0	996	7.95	211.5
France	FR	147.0	8654	36.20	3311.5
Germany	GE	398.0	7691	120.40	4538.0
Greece	GR	14.0	1104	3.60	175.5
Hungary	HU	18.5	657	4.35	203.0
Iceland	IS	0.1	37	0	1.4
Ireland	IE	5.0	438	0.90	43.5
Italy	IT	96.5	8576	35.85	1949.0
Luxembourg	LU	3.0	165	0.60	93.0
Netherlands	NL	34.5	2124	5.50	307.0
Sweden & Norway	SK	226.0	1728	18.60	504.9
Poland	PL	596.0	3086	180.45	4023.0
Portugal	PT	11.5	398	2.00	100.5
Romania	RO	114.5	1327	42.60	645.0
Spain	ES	268.0	4215	134.85	3982.5
Switzerland	CH	4.5	451	1.40	64.0
Turkey	TR	(39.6)	(2220)	(12.32)	(611.6)
United Kingdom	GB	119.5	8508	30.75	2296.0
Yugoslavia**	YU	271.5	1950	85.60	1804.5
Russia*,Turkmenistan, Kazakhstan,Uzbekistan	RF	1784.3	18273	186.45	7566.4
The Ukraine	UR	130.8	6825	50.66	2150.4
Moldova	MOL	3.5	473	1.06	12.0
Armenia,Georgia, Azerbaijan	ZKV	231.7	2596	71.12	2774.6
Belarus	BR	16.7	1324	4.05	66.6
Estonia,Latvia, Lithuania	PBL	11.8	1014	3.05	92.0
Morocco	MOR	(2.0)	(80)	(0.50)	(27.2)
Algeria	ALG	(15.7)	(1149)	(3.95)	(204.0)
Tunisia*	TUN	(7.1)	(338)	(1.80)	(90.6)
Libya*	LIB	(3.9)	(399)	(0.79)	(47.0)
Cyprus	CYP	(0.7)	(26)	(0.20)	(9.0)
Syria	SYR	(11.3)	(565)	(2.70)	(136.0)
Lebanon	LEB	(3.0)	(325)	(0.70)	(35.0)
Israel*	ISR	(4.0)	(440)	(1.00)	(62.0)
Egypt*	EGP	(17.5)	(832)	(4.37)	(144.8)
Jordan*	JOR	(1.7)	(75)	(0.58)	(26.1)
Total		5054.4	95799	1171.85	41818.4

* - within the calculation area
** - all countries located on the territory of the former Yugoslavia

factor 10 for some countries.

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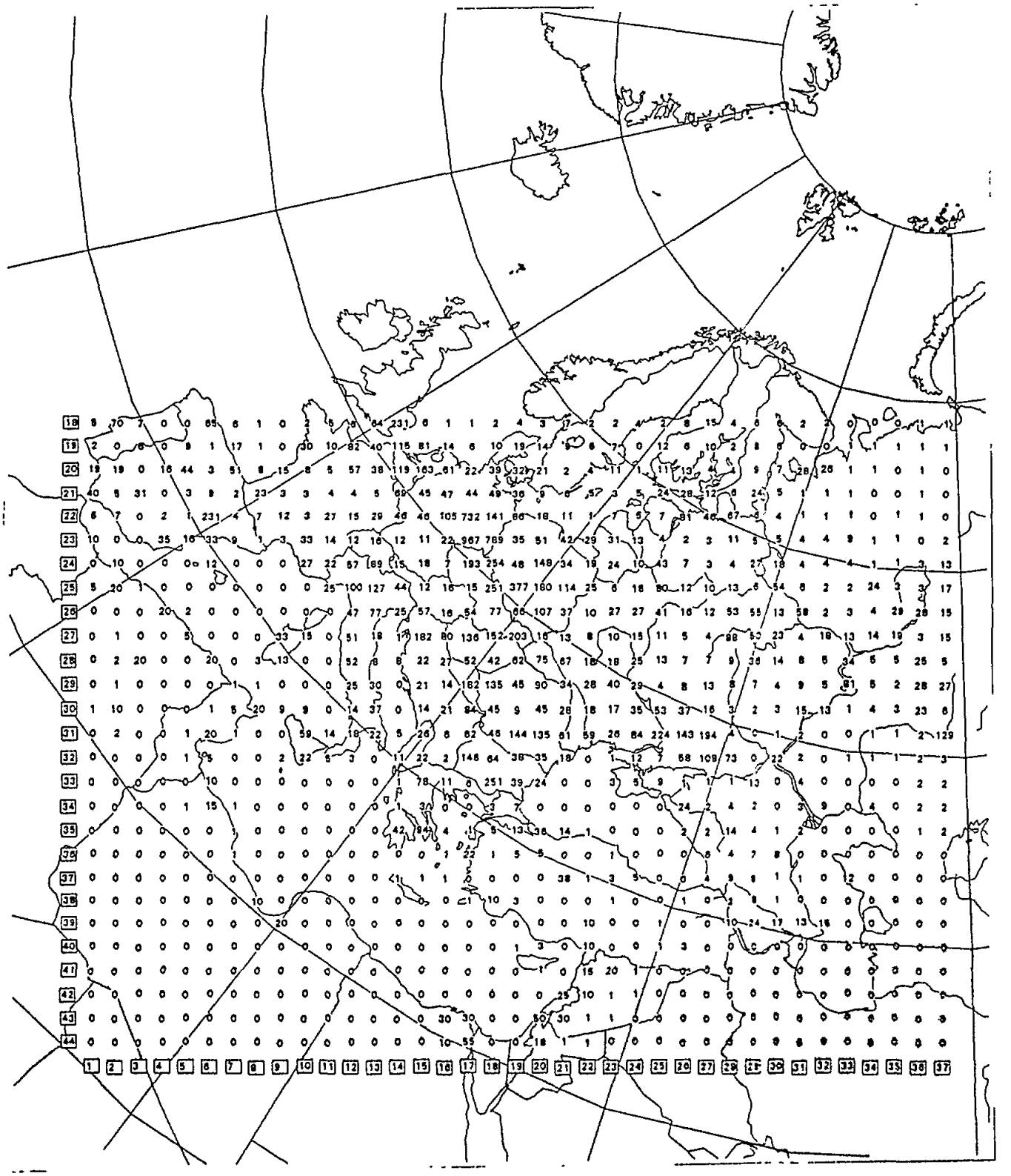


Fig.6.1 Sulphur emission intensity in grid squares of the calculation region *(1000 t S/year)*.

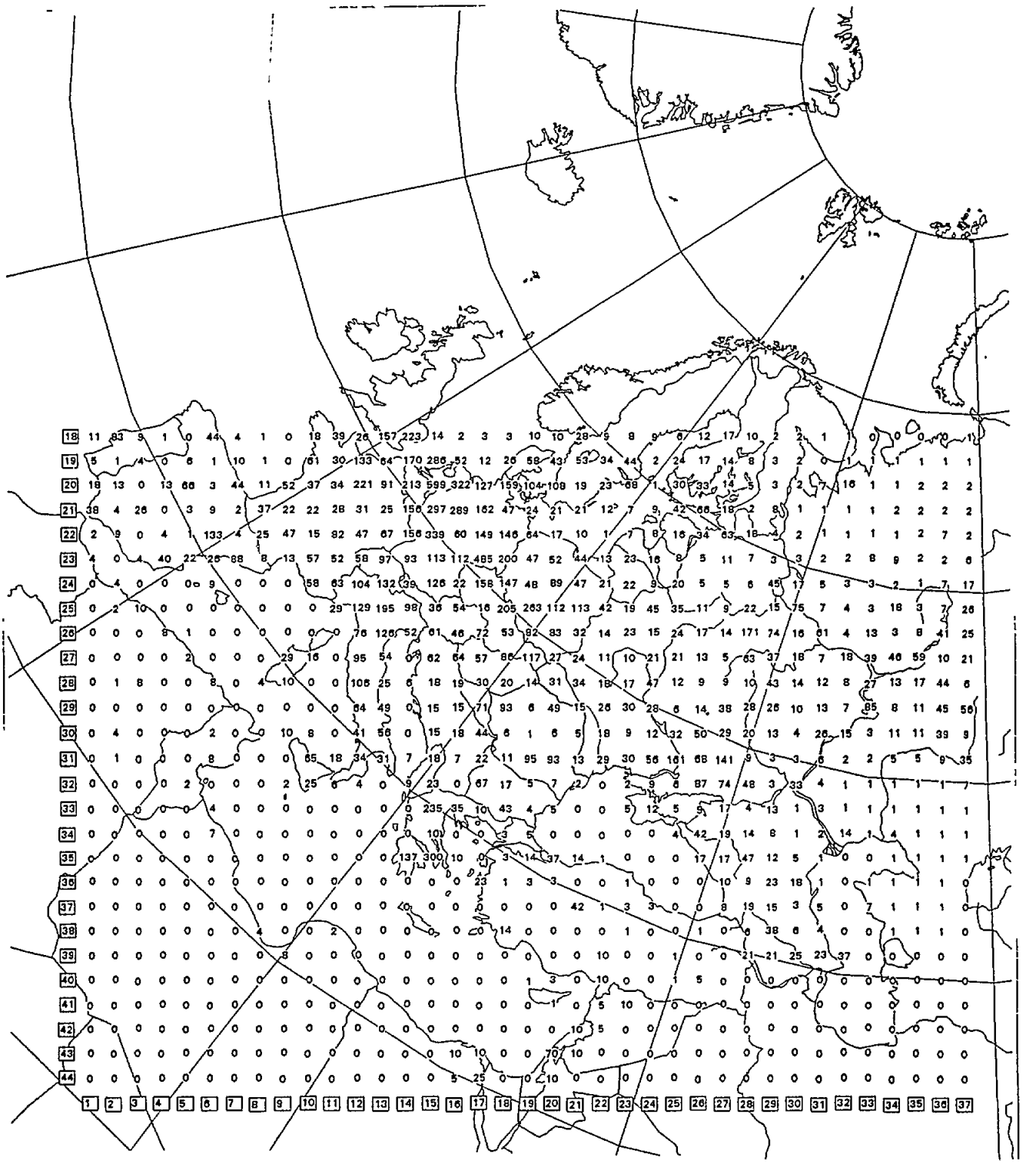


Fig.6.2 Oxidised nitrogen emission intensity in grid squares of the calculation region *(1000 t NO_2/year)*.

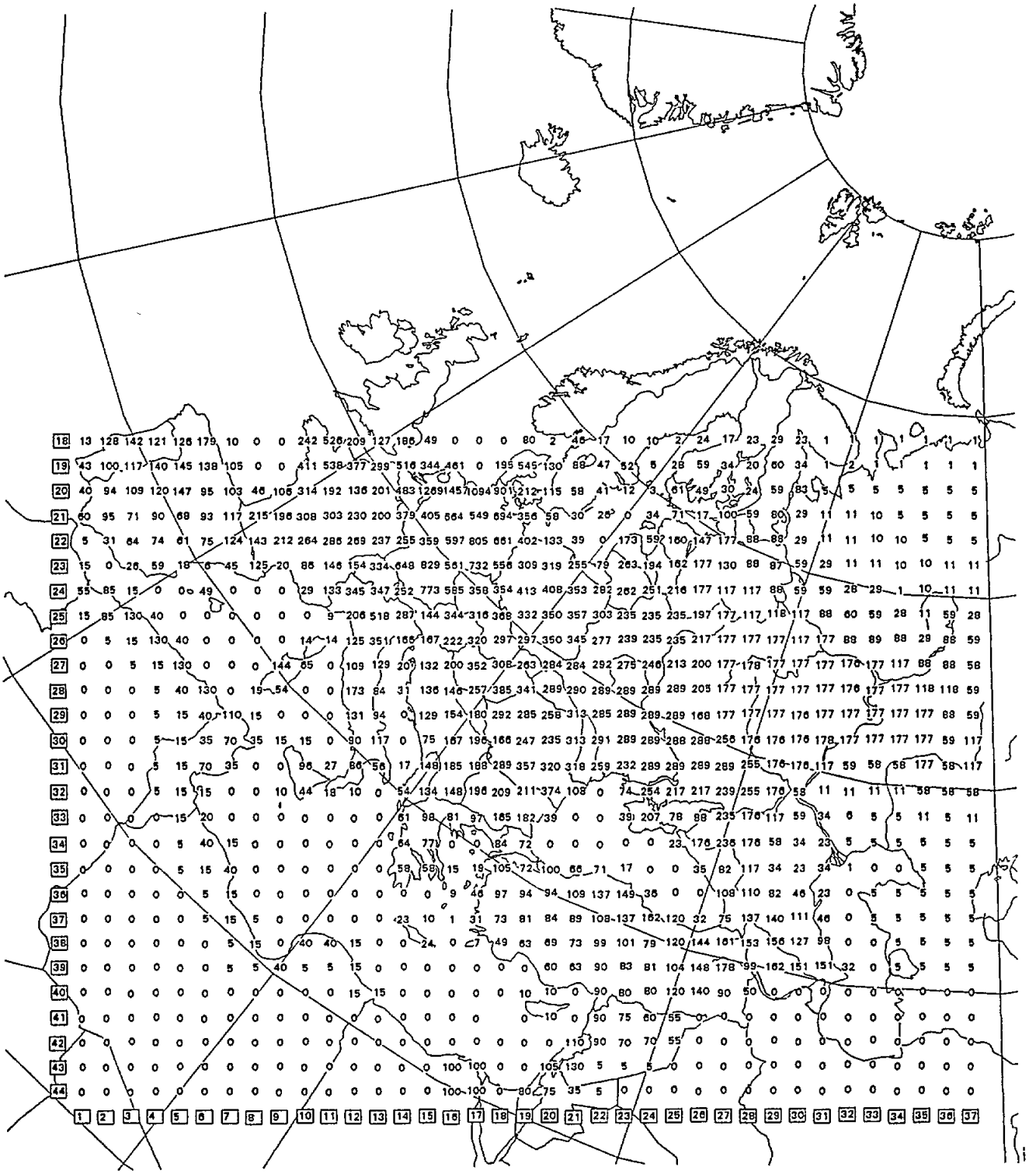


Fig.6.3 Reduced nitrogen emission intensity in grid squares of the calculation region *(100 t NH_3/year)*.

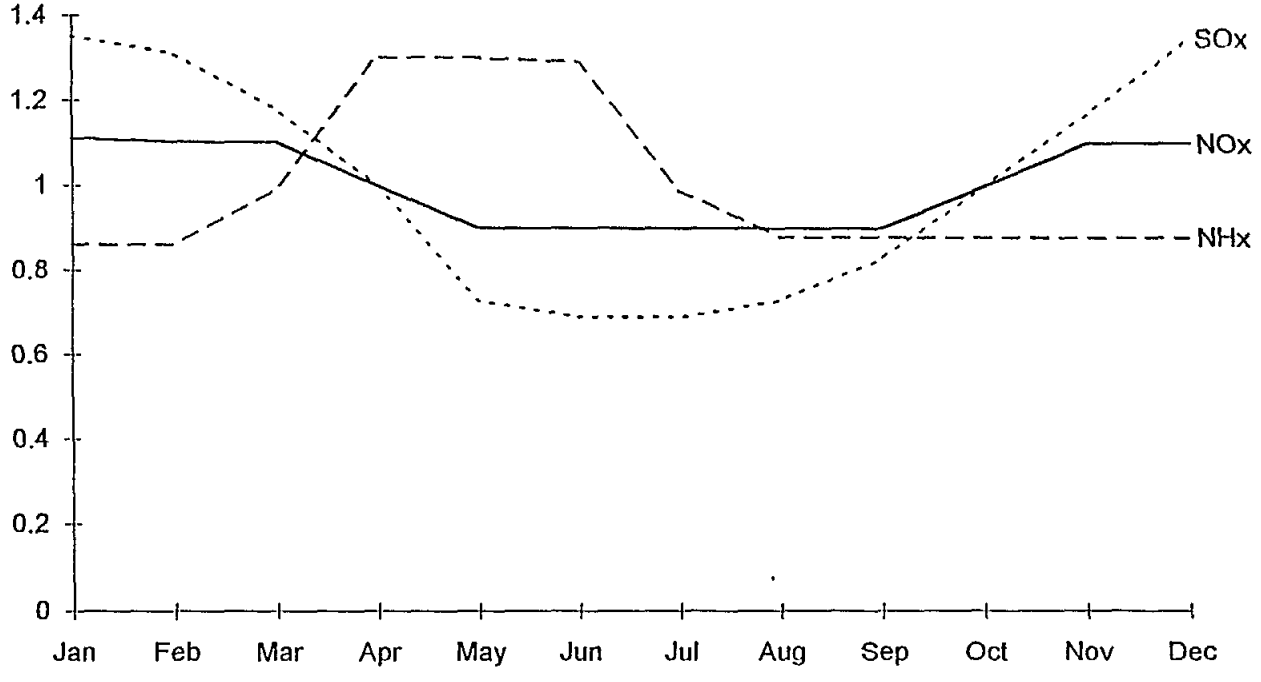


Fig.6.4 Seasonal SO_x, NO_x, NH_x emission variations for all countries (in per unit).

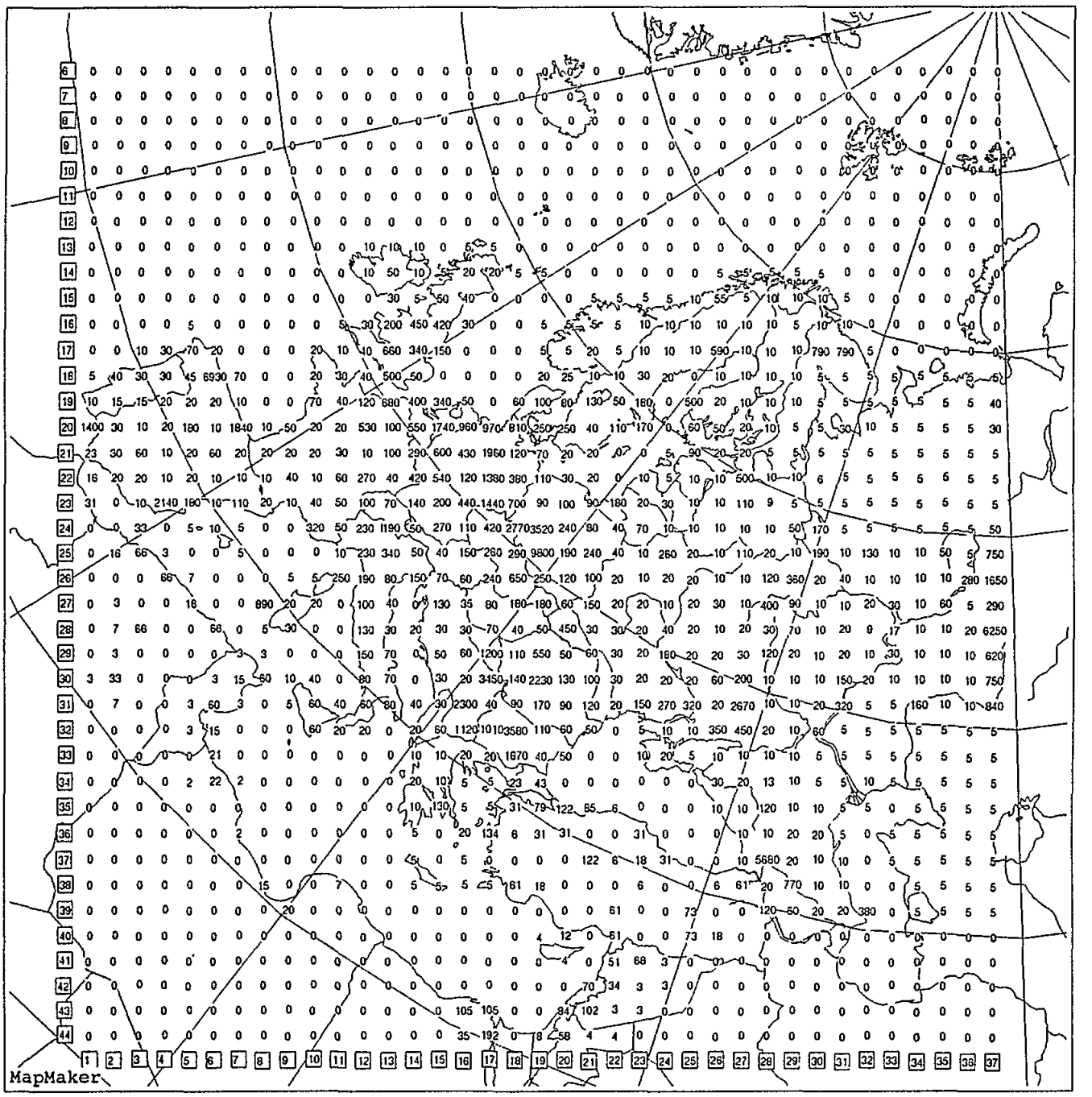


Fig. 6.5. Cadmium emission intensity in grid squares of the calculation region (10 kg Cd/year).

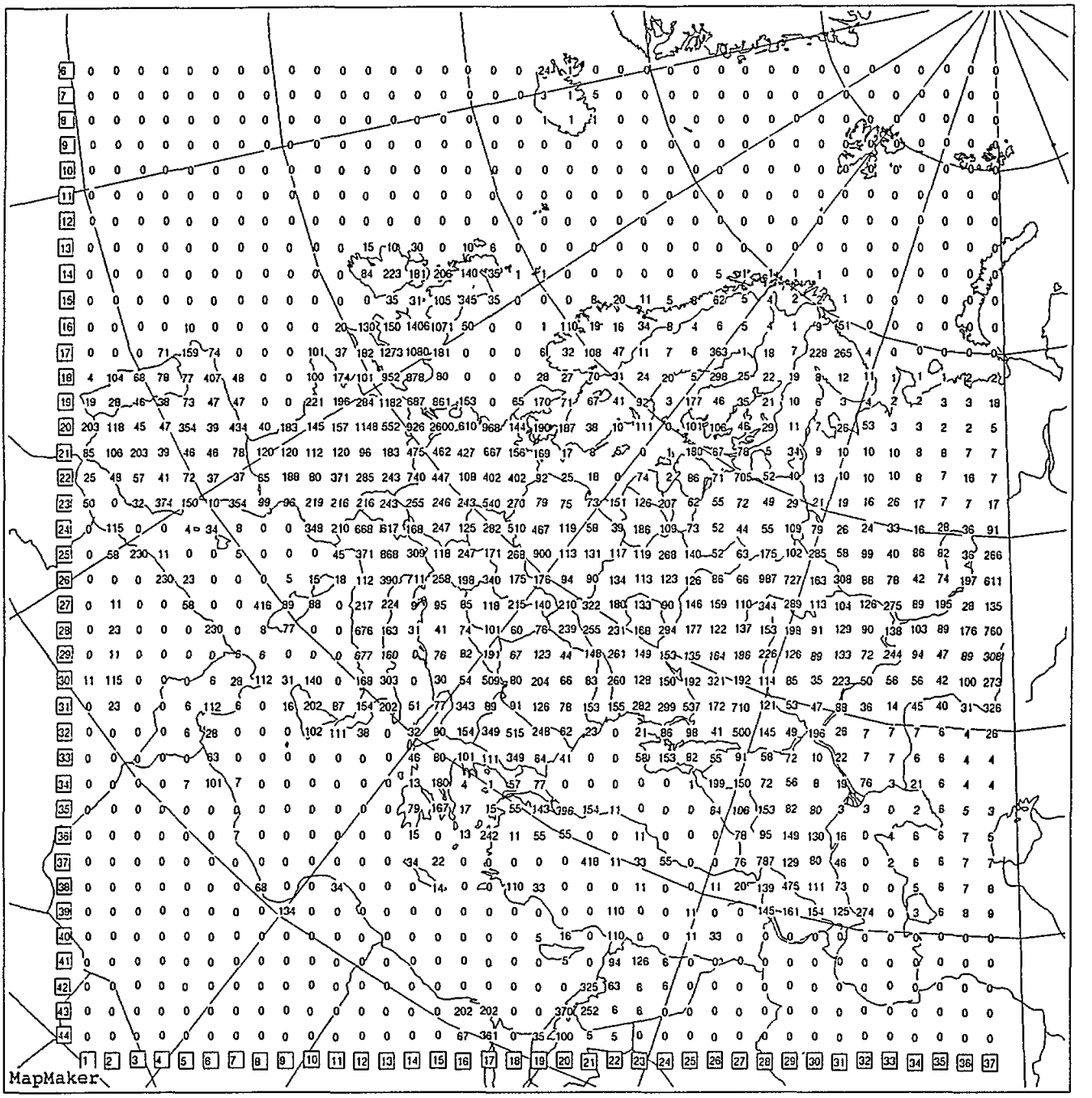


Fig. 6.6. Lead emission intensity in grid squares of the calculation region (1000 kg Pb/year).

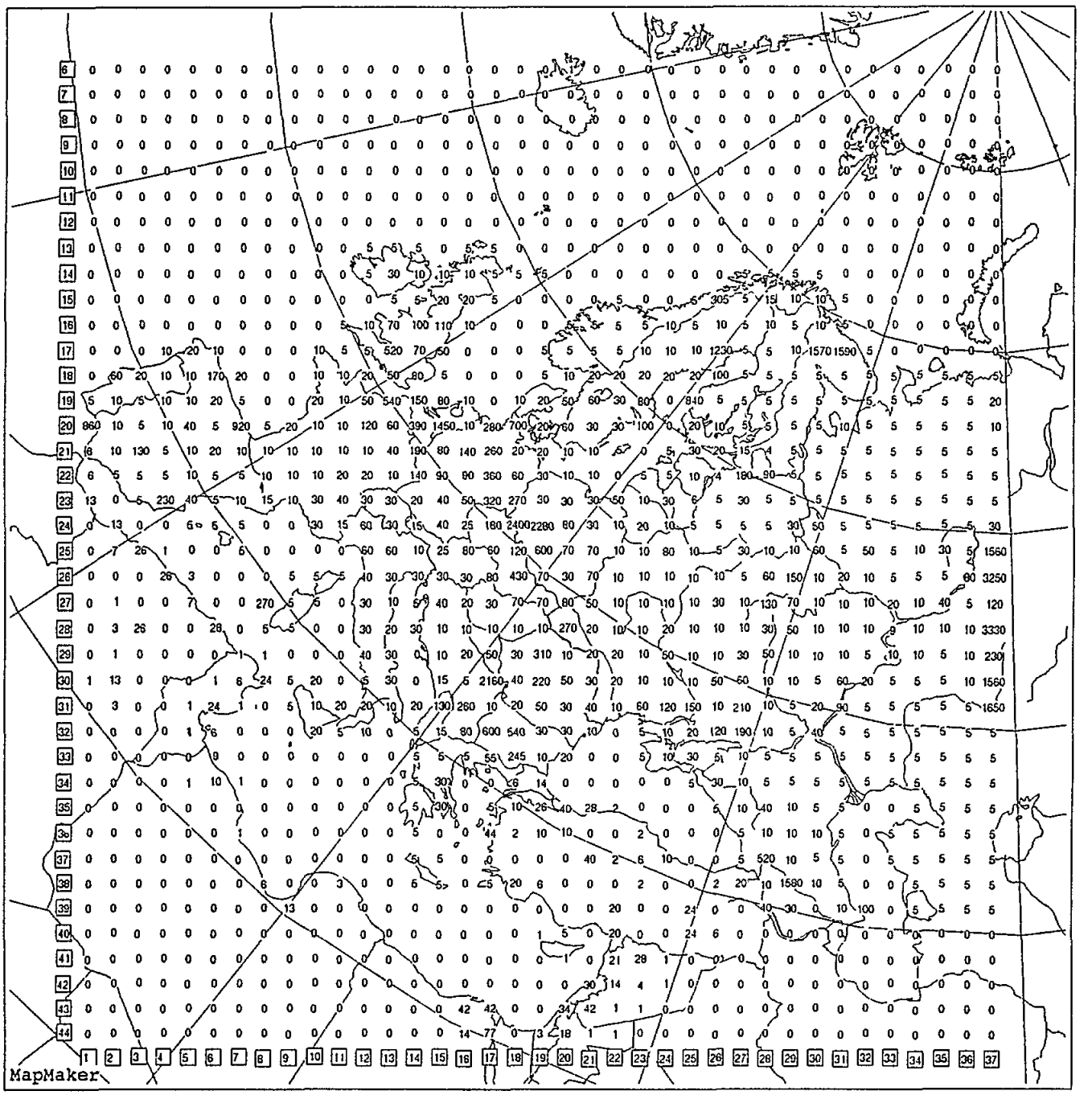


Fig. 6.7. Arsenic emission intensity in grid squares of the calculation region (100 kg As/year).

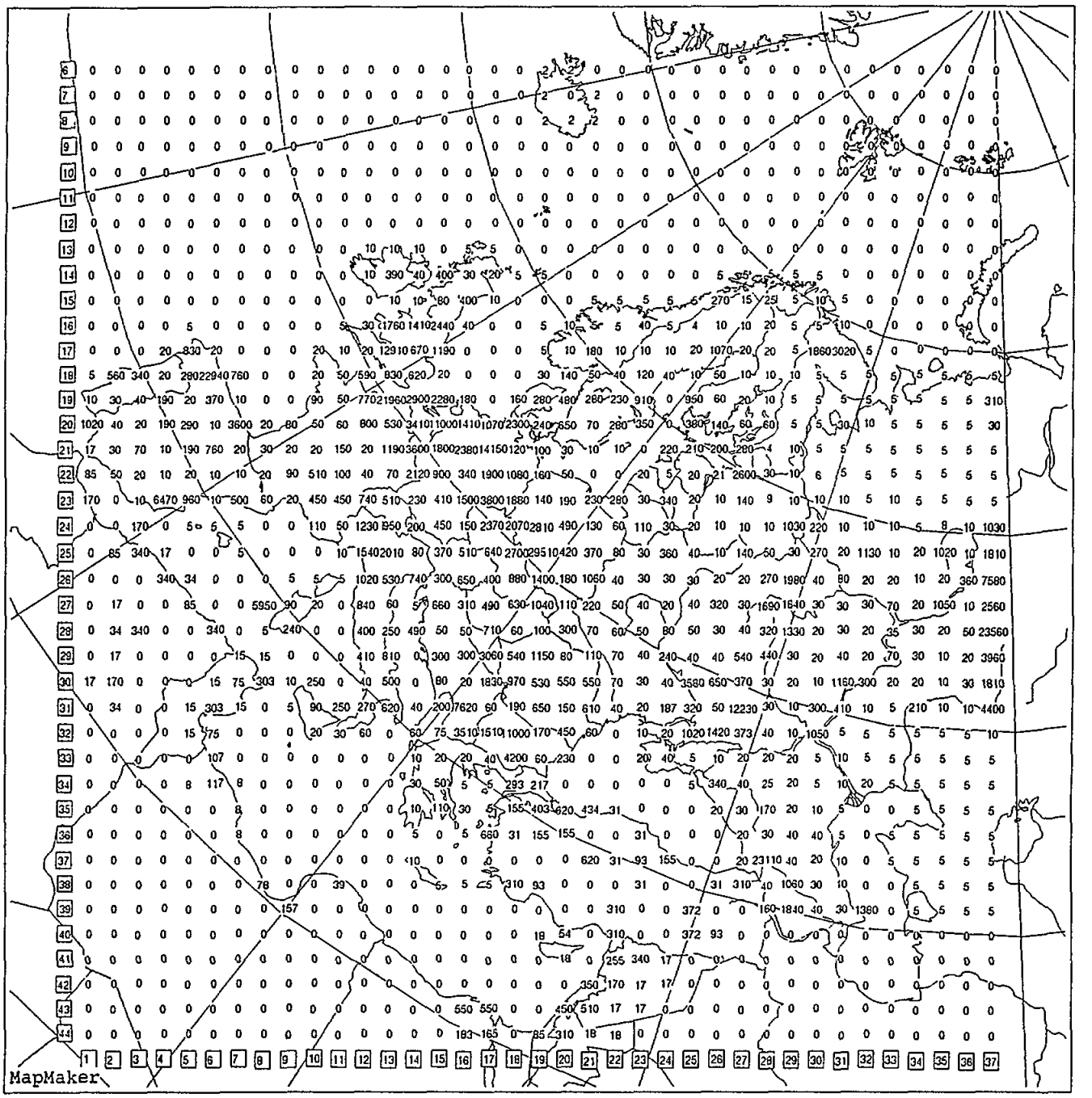


Fig. 6.8. Zinc emission intensity in grid squares of the calculation region (100 kg Zn/year).

VII. MODELLING OF ATMOSPHERIC POLLUTANT TRANSPORT AND DEPOSITION

The anthropogenic pollutants emitted into the air are transported downwind (advection) and dispersed along the horizontal (horizontal diffusion) and vertical (vertical diffusion). The pollution can be captured by precipitations and accumulated by clouds and washed out with them. In addition pollutants deposit due to gravitation (large particles with sizes more than a micron) and are trapped by the underlying surface (dry deposition) as well as they undergo chemical transformations. The latter is of a particular importance since initially inert species can be transformed into harmful substances or can provoke their formation. For example, NO (inert gas from the biological point of view) being transformed into nitric acid and nitrites in combination with volatile compounds may produce high ozone concentrations in the lower troposphere inflicting an adverse impact on biota. At the same time other NO_x entering the stratosphere destroy the stratospheric ozone layer required for the protection of biota from ultraviolet radiation.

7.1. Chemical and phase transformations

7.1.1. Physical-chemical transformation of sulphur oxides, nitrogen compounds and ozone.

Sulphur and nitrogen compounds entering various reactions undergo not only chemical transformations but also transfer from one phase to another (gas-aerosol) [1]. During these transformations changes in impacts on natural and other objects as well as in parameters of the process itself take place.

The development of a comprehensive model for chemical transformation for sulphur, nitrogen, peroxylacetylnitrate (PAN) and ozone is connected with a number of principal problems. A great number of intermediate reactions and reagents involved in the transformation of these substances and high reaction rates result firstly in the fact that reaction parameters and concentrations of many involved species (especially free radicals) are badly known and secondly more or less "complete" transformation schemes, being calculated with the help of equations, require so much computer time that cannot be afforded for routine calculations even if super computers were available.

Therefore a model chemical scheme should meet the following requirements:

a) As far as possible intermediate reaction products are excluded from the scheme. Substances which concentrations are measured and information on which is an objective of modelling are considered.

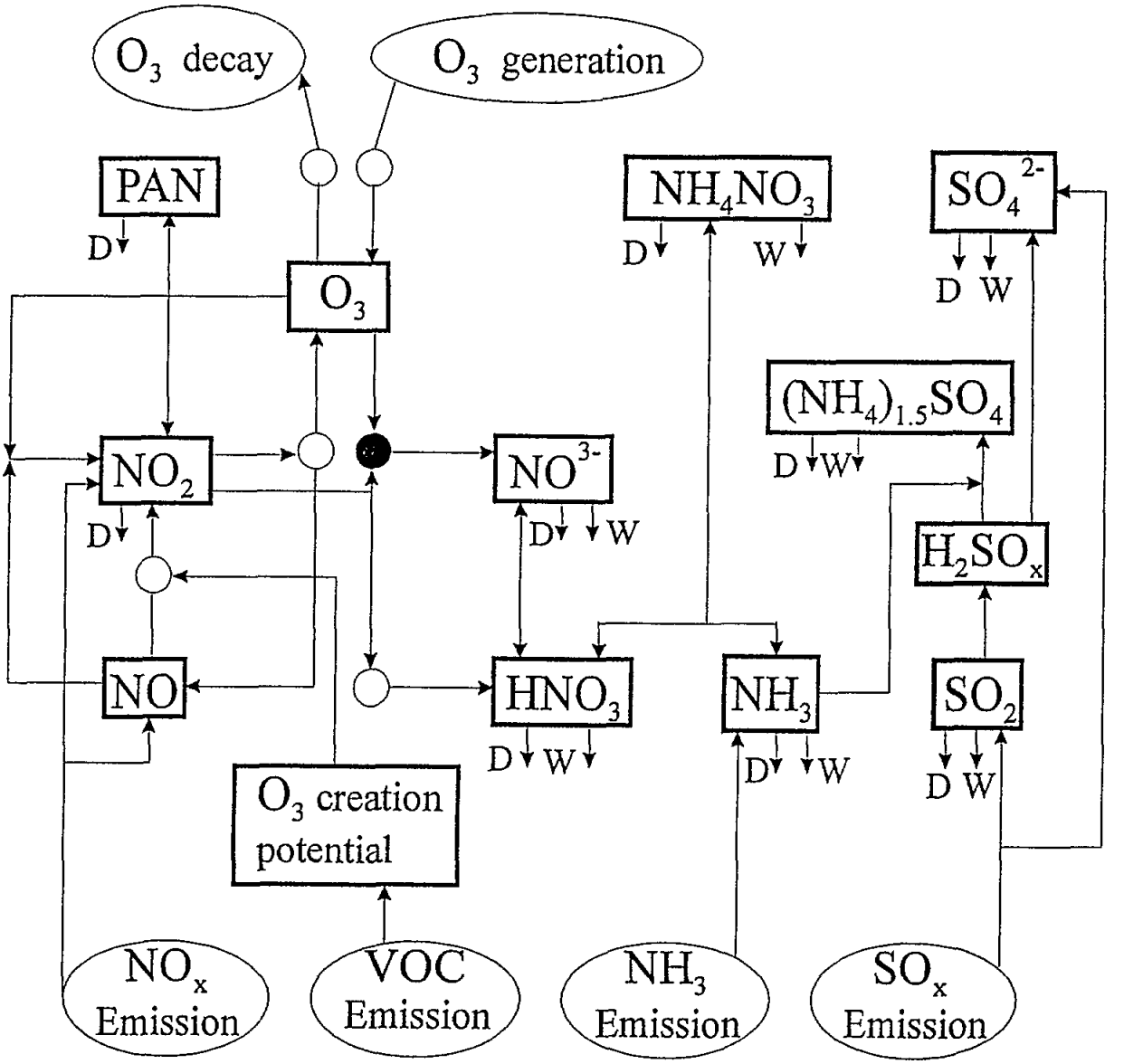
b) Chemical transformations of nitrogen oxides are in close connection with ozone concentrations and affect them greatly. Therefore concentrations of ozone and nitrogen oxides are calculated together.

c) Reactions occurring parallel (or chains of reactions) are reduced to a summarized reaction. In sequential reactions the slowest reactions are singled out.

d) In groups of reversible reactions the time of equilibrium establishment is determined by the fastest reactions. In these groups the equilibrium is calculated at each time step.

For the chemical scheme development the result and gained experience of investigations [2-11] were used. More detailed description of the scheme is given in [12].

A set of equations corresponding to chemical transformations shown in Figure 7.1 is written for an atmospheric column with unit bottom area of 150x150 km^2 (calculated grid cell). The upper limit is not prescribed - the mixing layer may be different for different types of pollutants.



○ - Photochemical reaction

● - Dark reaction

D - Dry deposition

W - Wet deposition

$(NH_4)_{1.5}SO_4$ is $(NH_4)_2SO_4 + NH_4HSO_4$

Fig.7.1. Model chemical scheme

The emission of SO_2, sulphates, NO, NO_2, NH_3 and volatile organic compounds (VOC) and chemical transformations depending on solar radiation, temperature and humidity take place in the column. Pollutants are removed from the column due to deposition and transport across lateral sides (advection) and inside the column the pollutants are redistributed along the vertical by turbulent diffusion. Calculations of these processes are made individually aside from those of chemical transformations therefore they are not taken into account in equations of the chemical scheme. The assumed notations are presented below:

[C] - concentration (mass in a calculated grid) of substance C;

$K_1,\ldots,K_{14}$ - reaction constants (see Table 7.1);

d(C) - concentration increment at a unit time step:

q(C)-emission of substances given in brackets ($NO_x=NO+NO_2$, $SO_x=SO_2+SO_4^=$, VOC−volatile organic compounds),

a and b - fractions of NO and SO_2 in the emission;

$g(O_3)$ - background rate of tropospheric ozone formation;

$(NH_4)_{1.5}SO_4$ - mixture in equal fractions of $NH_4HSO_4+(NH_4)_2SO_4$;

NO_3^- - substances incorporating ion NO_3^- in addition to HNO_3 and NH_4NO_3;

$SO_4^=$ - all substances containing ion $SO_4^=$;

R - free radicals (ozone potential).

Equations of the chemical transformation scheme of nitrogen oxides, sulphur oxides, reduced nitrogen, ozone, peroxylacetylnitrate (PAN) at the presence of volatile hydrocarbons (VOC) (coefficient and parameter values - see Table 7.1):

$$d(NO)=-K_1[O_3][NO]+K_2[NO_2]-K_3[NO][R]+aq(NO_X);$$
$$d(NO_2)=-d(NO)-d(PAN)-K_4[NO_2]-K_7[O_3][NO_2]+q(NO_X);$$
$$d(O_3)=-K_{13}[O_3]-K_1[O_3][NO]-K_7[O_3][NO_2]+K_2[NO_2]+g(O_3);$$
$$d(HNO_3)=K_4[NO_2]-K_9[HNO_3]+K_8[NO_3^-]-d(NH_4NO_3);$$
$$d(NO_3^-)=K_7[O_3][NO_2]+K_9[HNO_3]-K_8[NO_3^-];$$
$$d(NH_3)=-1.5d((NH_4)_{1.5}SO_4)-d(NH_4NO_3)+q(NH_3);$$
$$d(PAN)=K_5[R][NO_2]-K_6[PAN];$$
$$d(SO_2)=-K_{10}[SO_2]+bq(SO_X),$$
$$d(SO_4^=)=-d(SO_2)+q(SO_X);$$
$$d((NH_4)_{1.5}SO_4)=\text{Minimum}([NH_3]/1.5;K_{10}[SO_2]);$$
$$d[NH_4NO_3]=([NH_3]+[HNO_3])/2-(([NH_3]-[HNO_3])^2+4K_{11})^{1/2}/2$$
$$\text{at } ([NH_3]+[NH_4NO_3])([HNO_3]+[NH_4NO_3])>K_{11};$$
$$=-[NH_4NO_3]$$
$$\text{at } ([NH_3]+[NH_4NO_3])([HNO_3]+[NH_4NO_3])\le K_{11};$$
$$d(R)=-K_3[NO][R]-K_5[R][NO_2]+K_6[PAN]-K_{12}[R]+q(VOC).$$

Many reactions depend on the availability, level and spectrum of solar radiation. The latter is determined by the atmosphere state, latitude, local time and season and cloud cover density. Latitude and time determine the solar zenith angle Z. It is usually assumed that reaction rates vary proportionally to the function:

$$F(Z)=\exp(-r/\cos Z)$$

where r - constant characteristic of a given reaction. For the majority of reactions r value is

known to be approximately within the range of 0.5-2.5. In modelling relative (dimensionless) values of radiation are used. They are calculated by the 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 reference tables; M - number of a month of the year; T - local time in hours.

The radiation level at latitude 35° at equinox, at noon is taken to be S=1 (it means, that $0 \leq S \leq 1.2$). It is supposed that radiation decreases proportionally to cloud cover thickness (density). The final parameterization of the chemical scheme is specified on the basis of statistical analysis provided by the EMEP network data given in [13].

Table 7.1. Coefficients and parameters of chemical transformation scheme

Coefficient or parameter	Unit	Value or formulae
K_1	$m^3/(\mu mole * hour)$	15.24+0.068T
K_2	1/hour	12S
K_3	$m^3/(\mu mole * hour)$	$1100S^2+0.25$
K_4	1/hour	$0.08S^2$
K_5	$m^3/(\mu mole * hour)$	$12S^2$
K_6	1/hour	$28.6 * 10^{17}\exp[-12530/T]$
K_7	$m^3/(\mu mole * hour)$	$5.625 * 10^{-4}(T-228.7)$
K_8	1/hour	0.015
K_9	1/hour	0.06
K_{10}	1/hour	138500exp[-4517/T]
K_{11}	$[\mu mole/(m^2 * hour)]^2$	***
K_{12}	1/hour	0.03
K_{13}	1/hour	0.005+0.2S
$g(O_3)$	$\mu mole/(m^3 * hour)$	$0.044S^2+0.21S+0.03$
a,b	relative units	0.95

*** $K_{11} = 1.12 * 10^{34} * Ho * [\exp(-24220/T)](298/T)^6$ [1]
where Ho = (100-H)/38 at $H \geq 62$ and Ho = 1 at $H < 62$;
dependence K_{11} (T,H) is taken from data [14].
Notations:
T - temperature, in K°,
S - relative insolation (see text),
H - relative humidity, in %

7.1.2. Heavy metals

Heavy metals are emitted into the atmosphere, as a rule in the form of stable chemical compounds. Since the life time of such metal containing aerosol particles is several hours or days, metals can be considered chemically completely inert additive. In any case, the effective toxic properties of the metal under possible chemical transformations ought to be considered invariable.

7.2. Deposition processes and their modelling.

7.2.1. Vertical diffusion and dry deposition and their account in the calculation model.

The consideration of dry deposition is particularly important for the Mediterranean region which has a dry season when the input of airborne pollution into the water basin and adjacent counties is determined mainly by this process.

The dry deposition quantity depends on pollutant concentrations in the near surface (water) layer $C(z=0,t)$, vertical diffusion coefficient K_z, boundary layer height H_b and dry deposition velocity V_d (z - vertical co-ordinate, t - time).

It is known that dry deposition velocity V_d is determined as a coefficient in the equation

$$\frac{d}{dt}m(t)=-V_d C(0,t), \tag{7.1}$$

connecting concentration $C(z=0,t)$ in surface (near water) layer and pollution amount $m(t)$ in an air column

$$m(t)=\int_0^\infty C(z,t)dz$$

For modelling we introduce an equivalent mixing layer $H(t)$

$$H(t)=\frac{m(t)}{C(0,t)} \tag{7.2}$$

for which the equation from papers [16,17] is used:

$$\frac{dH}{dt}=\begin{cases}2V_d+\dfrac{2K_z}{\pi H} & H<H_b \\ \dfrac{0.2Kz}{\pi H} & H\geq H_b\end{cases} \tag{7.3}$$

Hence for calculation of

$$C(0,t)=\frac{m(t)}{H(t)}$$

a closed system of differential equations (7.1) and (7.3) relative to $H(t)$ and $m(t)$ is obtained.

Figure 7.2 shows dependencies of H_b on the temperature and underlying surface type used in the model when data on the boundary layer height are not available in the meteorological information [40].

For the coefficient of vertical turbulent diffusion the model uses a dependence of K_z on temperature:

$$K_z = 2.2[m^2/s]\exp(0.14T[^0C])$$

This dependence was obtained by regression analysis of K_z budget estimates for different temperatures [15].

The dry deposition velocity V_d for gases is determined by their physical - chemical properties and by properties of the underlying surface and for aerosols - mainly by particle sizes (see Section 7.2.1.1 and 7.2.1.2).

7.2.1.1. Sulphur and nitrogen compounds.

Since aerosol phase of sulphur and nitrogen compounds is composed of particles with diameters less than 0.1 μm, then liquid and gaseous phases of these substances can be considered a "weightless pollutant".

For the estimations of V_d of gases and aerosols with particle sizes less than 0.1 μm we used data available in the literature [18] and the information of two types. First, the direct measurements carried out in Poland as a contribution to EMEP [19]. The data provided rather comprehensive picture of the underlying surface impact on dry deposition velocity. In particular these data indicate that the extent of surface moistening strongly affects the variations of V_d for SO_2 But these data are valid only for positive temperatures. Second, the budget estimates of V_d for the whole EMEP territory [15] were used for the present calculations. It was shown that mean V_d is abruptly changed at the temperature about 0^0C, 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_{dn} and function types $K(T,P,F)$ are given in the table:

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

1. $K=1$ at $T<-2°C$ and any P and F
   $K=1$ at $T>-2°C$ $P=0$ and $F=0$
   $K=2$ at $T\geq 2°C$ $F=1$ or $1mm/h>P>0$
   $K=3$ at $T\geq -2°C$ $F=2$ or $P\geq 1mm/h$
2. $K=1$ at $T<-2°C$ and any P and F
   $K=1$ at $T>-2°C$ $F=0$ and $P=0$
   $K=0.5$ at $T\geq -2°C$ $F=1$ or $1mm/h>P>0$
   $K=0$ at $T\geq -2°C$ $F=2$ or $P\geq 1mm/h$

7.2.1.2. Heavy metals

In dry deposition of polydispersed smoke particles two processes are important sedimentation (gravitational deposition) and trapping of particles by the underlying surface. The first process is important for particles with sizes more than 1 μm. At the same time particles with sizes above 0.1 μm cannot be considered as Brownian ones. Therefore in case of particles with sizes 0.1-1 μm relative minimum of dry deposition velocity is observed. In the first approximation the dependence of integral deposition velocity on particle sizes is shown in Figure 7.3. The curve is plotted on the basis of data from [20,21]. This figure also shows sedimentation velocities obtained in [21] for different ranges of particle sizes D.

Figure 7.4. is a histogram showing particle distribution with sizes characteristic of a thermal power station as well as of other enterprises. The effect of gas treatment with the help of usual techniques was considered in this histogram [22].

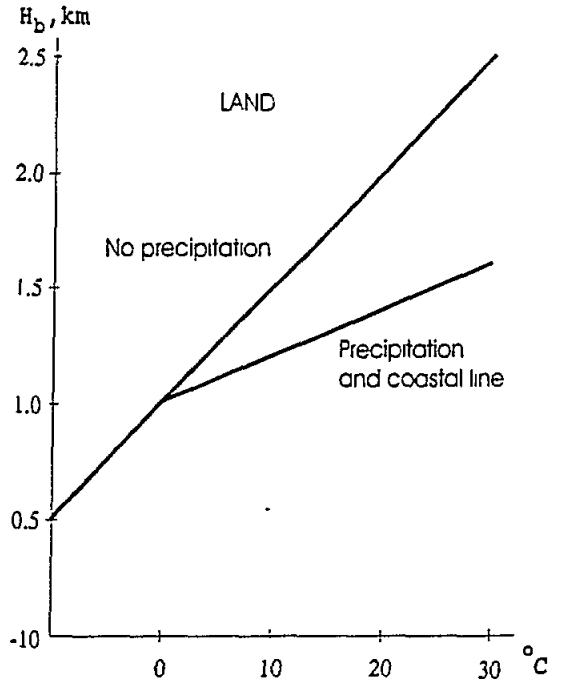
In order to make estimation of dry deposition, data similar to those of Figures 7.3 and 7.4 may be used. Each particle size range (fraction) is assigned with appropriate mean deposition velocity and depositions from all the fractions are summed. This approach gives rise to a problem of selection of the D ranges and computer resource consumption.

Quite different approach is possible when a certain velocity can be prescribed to all particles for every time t. This velocity $\overline{V_d(t)}$ is mathematical expectation of V_d(t), i.e.:

$$\overline{V_d}=\int_0^\infty V_d m(V_d)dV_d \Big/ \int_0^\infty m(V_d)dV_d \qquad (7.4)$$

where $m(V_d)dV_d$ - particle mass with deposition velocities between V_d and V_d+dV_d.

For the model calculation of $V_d(t)$ equation from [16] has been introduced:



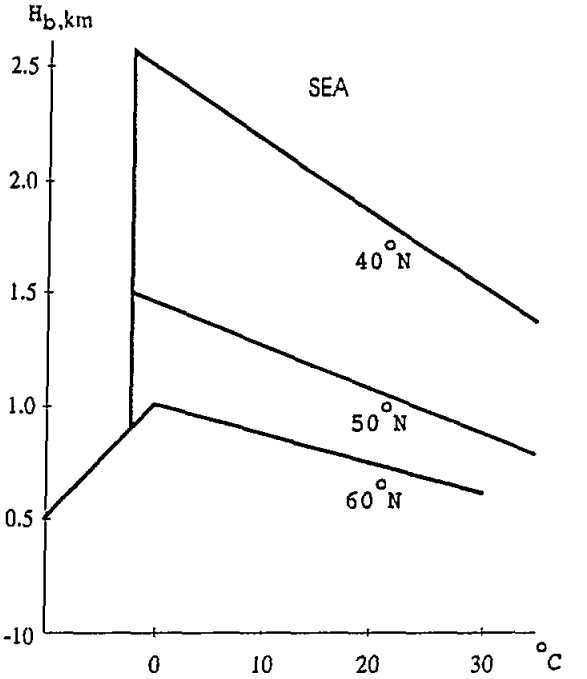


Fig.7.2. The assumed boundary layer height versus surface temperature.

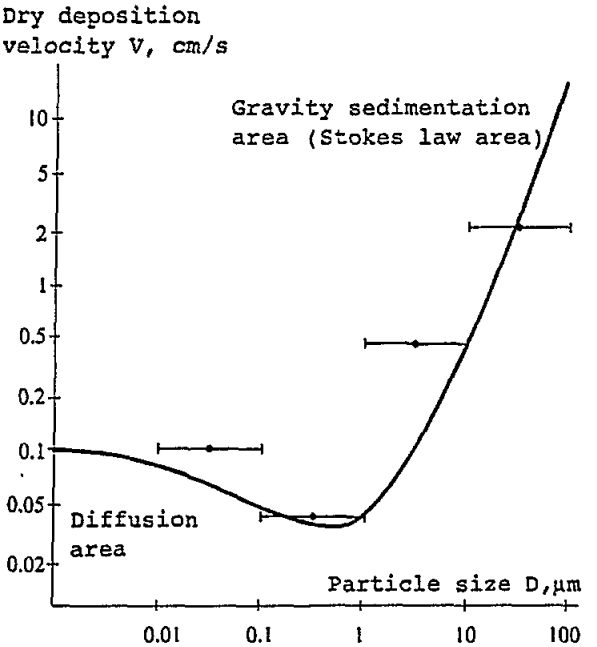


Fig.7.3. Particle dry deposition velocity V versus particle size and observed values of V for the Baltic Sea.

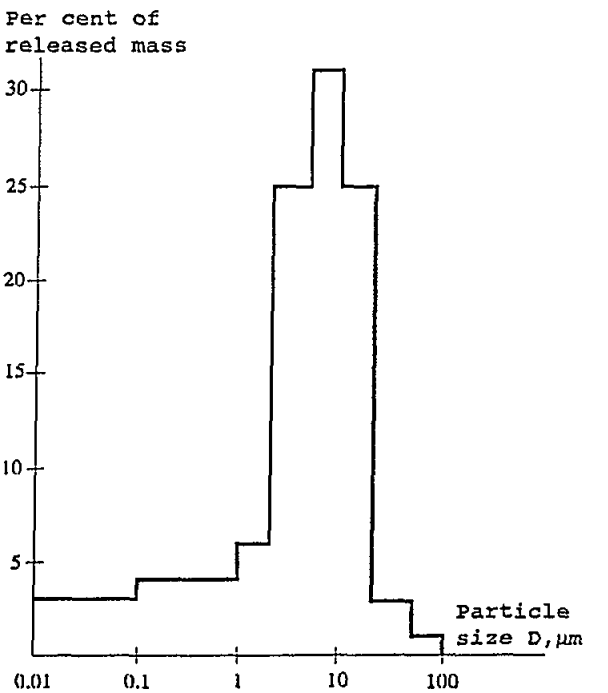


Fig.7.4. Typical histogram of distribution of particle size logarithms for the emission of the power stations operating on coal. Other plants (for example, metallurgical ones) have similar distributions.

$$\frac{d\overline{V_d}(t)}{dt} = -\frac{\sigma^2}{\overline{V_d^2}(O)} V_d^2(t)/H(t) \qquad (7.5)$$

where σ^2- variance of $V_d(0)$.

7.2.2. Wet deposition

The process of pollution scavenging from the air and its accumulation in clouds is one of the main processes of the atmosphere cleaning with simultaneous pollution of the underlying surface. A critical characteristic of it is the Langmuir washout coefficient (Λ) connecting pollution amount $m(t)$ in an air column with its scavenging velocity:

$$\frac{dm}{dt} = -\Lambda m \qquad (7.6)$$

If surface saturation of drops (flake of snow) in subcloud layer may be neglected in the process of scavenging then as it is shown in paper [23] Λ is determined as:

$$\Lambda = \beta S_0 I / v_p \qquad (7.7)$$

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

The relative surface of precipitation elements S_0/v_p (and consequently p_0) strongly depends on precipitation intensity and on its type. If we consider data presented in [24] then S_0/v_p for rain is:

$$S_0/v_p \cong 6(I^2 + I_0^2)/I^2 D \qquad (7.8)$$

where D - maximum effective diameter of a drop ($D \cong 1.5$-2 mm) and $I_0 \cong 0.8$ mm/h. For the snow relative surface varies slighter depending on precipitation intensity:

$$S_0/v_p \cong 6(I+I_0)/ID \qquad (7.9)$$

where $D \cong 5$ mm and $I_0 \cong 1$ mm/h and most probably it is connected with the formation of the near-surface air layer around snow flakes.

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

According to Brownian motion theory of Einstein on the one hand and Langmuir theory on the other it is possible to accept the following approximation:

$$\beta \cong \beta_0 [\sqrt{D_0/(D+D_0)} + D^2 p/(D^2 p + D_R^2 \rho_R)] \qquad (7.10)$$

where β_0 - efficiency factor at the "ideal" contact of precipitation elements and pollutant: ($\beta_0 \cong 0.25$ for rain and $\beta_0 =$ =0.5-1 for snow); D_0 - doubled radius of Van der Vaals for gaseous molecules ($D_0 \cong 10^{-3}$ μm); D - characteristic size (diameter) of washed out particles;

ρ- density of trapped particles; $D_R \cong 3$ μm - particle diameter corresponding to trapping efficiency 0.5 (at $(D \gg D_0)$ [21], $\rho_R \cong$ 1 g/cm^3 - particle density.

In (7.10) the first component in square brackets describes diffusive (Brownian) trapping, the second component - inertial trapping. Minimum $\beta = 0.08$ corresponds to value:

$$D_{min} \cong (D_0 D_R^4)^{1/5} = 0.6\,\mu m \qquad (7.11)$$

Dependence of β on D is shown in Figure 7.5. This figure also shows the dependence of β on D for precipitation intensity 1 mm/h calculated by (7.10) and (7.11) in comparison with estimations given in [21] and particle size spectrum over the continent according to [25]. It can be assumed that the spectrum shape is predetermined by the minimum on curve β (D).

Now consider the situation when $D \cong D_0$ (gases and aerosols with $D < 0.01$ μm) and the saturation effect characteristic of such substance. It is quite possible that this effect is true for larger particles but no experimental data are available to check this hypothesis.

Non-linear character of washout processes relative to concentrations of washed out pollutants and not only to the precipitation intensity, became clear to investigators at the beginning of the 80's when sufficiently representative measurement data became available [13]. Later a number of authors made investigations of kinds of these non-linearities [26-29]

An analysis and interpretation of these measurement data and development of a non-linear submodel of washout on their basis for the first time have been made by MSC-E [23].

The dependence of washed out substance concentration in precipitations-p on its air concentration-a is the main characteristics of the washout process.

An analysis of voluminous experimental data accumulated in the course of the EMEP implementation [13] made it possible to determine that dependence $p(a)$ for sulphur and nitrogen oxides has saturation at some $p = p_0$. At constant precipitation intensity these dependencies can be described by relationship:

$$p = p_0[1 - exp(-a/a_0)] \qquad (7.12)$$

where p_0 and a_0 - parameters depending on the precipitation intensity and pollutant type.

Figure 7.6 gives curves (7.12) for summer and winter halves of the year for sulphur and nitrogen oxides.

At first the $p(a)$ curves growth is 10^3-10^4 times greater than the equilibrium one corresponding to Henry's law for sulphates and nitrates. At the same time the saturation of the $p(a)$ curves takes place at concentration p_0 which is 10^3-10^4 times lower than the saturation concentrations for the same compounds.

In [23] on the basis of data [30] it was shown that p_0 and a_0 values in (7.12) correspond to the saturation of precipitation element surfaces compactly packed by molecules (or ions) of trapped substance and $p_0 \cong 0.1$ μmole/l and $a_0 \cong 0.1$ μmole/m^3. However, until now it remains unknown how certain pollutants could "help" or impede scavenging of the others.

Actually the air concentration is a function of z and time therefore more complete description of the washout process is a set of partial differential equations [23]. 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 in this part (Figure 7.7). Secondly, downgoing air flows entrain pollutants from upper to lower layers. Thirdly, since gas concentrations in falling drops are much higher than equilibrium ones (for example, SO_2) the part 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 [30].

The usage of a full model [23] for practical calculations leads to unjustified sophistication of computations. In order to develop an approximate model we use estimate of the pollution amount Δm_w scavenged by precipitations for Δt time period from the unit area:

$$\Delta m_w = m p_0 I \Delta t / (m + p_0 I \Delta t + m_s) \qquad (7.13)$$

where m_s is a value characterizing the maximum trapping capability of precipitations. The value of m_s is about 0.1 μmole/m^2 for rain as well as for snow.

The problem of rain drops saturation for pollutants producing cations in solution (for example, NH_4^+ or metals) is remained open. In the discussed model effects connected with saturation (in case of cations) are not considered, but dependence S_w / v_p on precipitation intensity and its type is taken into account.

7.3. Modelling of horizontal transport

In order to consider the non-linearity of physical-chemical processes and wind variations along the vertical it is necessary to apply calculation schemes for horizontal transport and diffusion which operate with total masses in grid cells. For calculations of the long-range transport the schemes with a small pseudodiffusion and adequately describing impulsive impacts on pollution concentrations (precipitations, point sources) are required. The most advanced Eulerian schemes do not meet these requirements [31,32].

In this context MSC-E has developed combined advection schemes. Practically they may be applied to any computational scheme of vertical diffusion and dry deposition.

A scheme with variable time step is used in these calculations. The scheme provides high calculation speed and is used mainly in long-term current calculations of the pollution transport from continuous sources, since it has some uncertainty in the estimate of pollution arrival within several hours.

"Lagrangian" and "Eulerian" steps of transport are completely separated. At the "Eulerian" step the mass and first moments exchange between cells are calculated. At the "Lagrangian" step we determine the moment of a cell mass transport to a neighbouring cell and the pollution input and scavenging process.

In order to carry out the Lagrangian phase of computations virtual trajectories coming from the cell mass center are calculated. The mass center prescribes only the initial position of the virtual point but the mass center shift does not affect the virtual point in future. The mass transport to an adjacent cell takes place when the virtual point (the trajectory end) is transmitted to a cell-receiver. Since the co-ordinates of the virtual point are the transported mass center the intercell transport velocity is exactly the wind speed, as it is in Lagrangian models.

Masses entering a cell are summed and the co-ordinates of their common center are calculated under the condition of mass center conservativity.

The scheme equations do not have terms describing horizontal diffusion of

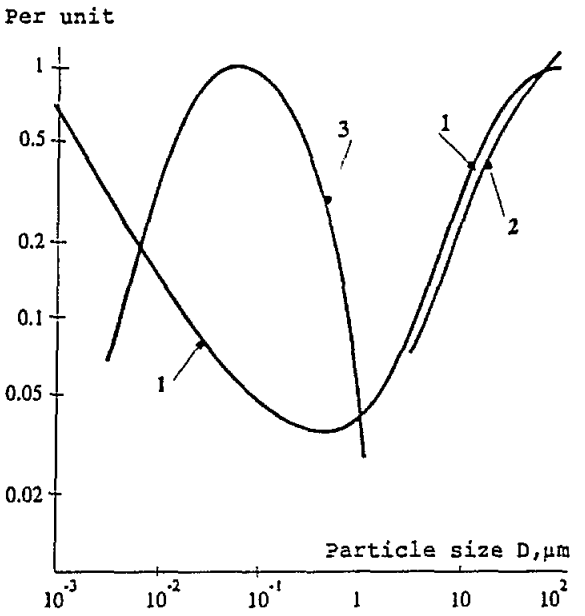


Fig.7.5. Relative efficiency of wash out versus particle sizes (1 - calculated from formula (7.10), 2 - according to Chamberlain's results [21]) and aerosol size spectrum over continents (according to Junge [25]).

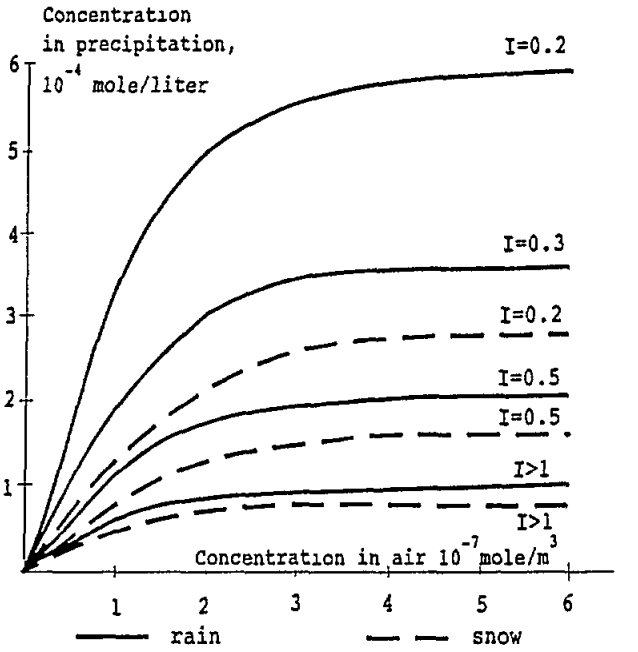


Fig.7.6. Sulphur and nitrogen oxide concentrations versus their concentrations in air for different precipitation intensities I (mm/h)

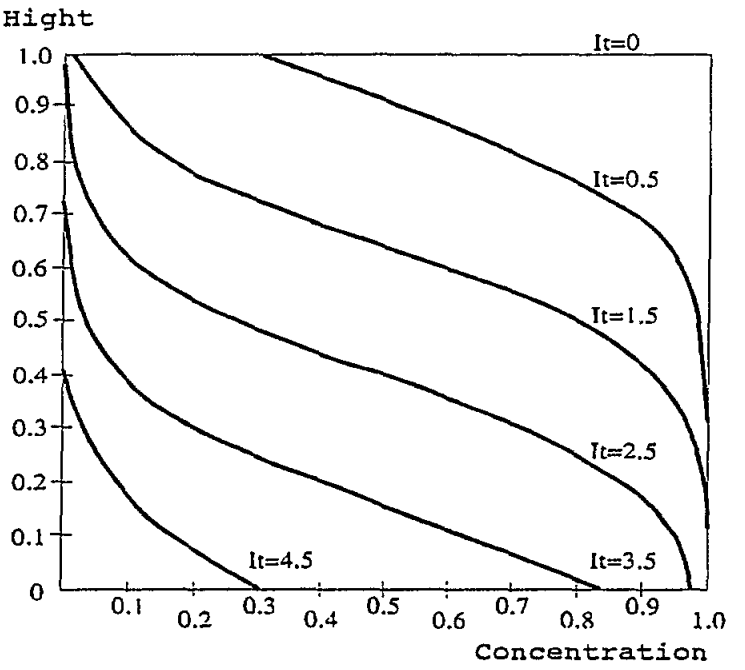


Fig.7.7. Height distributions of pollution concentration (relative unit) for different precipitation amount It (mm).

pollution. It is explained by the fact that at the calculated distances (cells 150x150 km^2 and the transport at the scale of continents) the horizontal diffusion is mainly concerned with wind variation in strength and direction at different heights [33]. At other scales the calculation of horizontal diffusion can be introduced, for example, proportionally to the distance covered by pollution according to known empirical dependencies. In this case the calculation of horizontal dispersion should be made only when the mass is transferred from cell to cell.

The scheme fixes the outflow of pollution outside the grid limits. If some mass reaches one of four grid limits it is assumed that it has left it forever. Total mass crossing the grid limit is calculated for each of them. If a mass crossed a grid angle it is added to the mass crossing the nearest limit in counter clock-wise direction. The model fixes pollution amounts leaving the grid in each direction.

If a mass m in a cell appeared to be smaller than the threshold value m_{min} then calculations for this cell at the $(i+1)$-th step are not made (certainly besides masses coming from other cells). Mass m is added to a value A which is used for the evaluation of unidentified deposition over the whole grid. The value m_{min} is chosen in such a way that it corresponds to concentration obviously smaller than the background (for sulphur and nitrogen compounds this concentration is $\cong 0.001$ μmole/m^3). Thereby the mass balance for the calculated period is provided.

The scheme has a number of useful properties:

a) it admits practically any description of vertical diffusion and dry deposition;

b) it operates with total mass in cells like Eulerian model and makes it possible to simulate non-linear effects (chemical transformation and precipitation scavenging);

c) like Lagrangian models it has no pseudodiffusion and it allows us to select model factors for horizontal diffusion fitting from the experimental data;

d) it provides high computation speed;

e) it is absolutely robust to both variations of coefficients and impulse impacts (emission sources, precipitations).

Figures 7.8-7.10 show the solution of three test problems by the scheme. It is demonstrated that there is no psuedodiffusion and the advection velocity is kept exactly as it would be if the classical Lagrangian scheme is used.

7.4. Structure, calculation arrangement, input and output data, main features of the MSC-E model.

The modelling is carried out in a separated region with dimensions 39x37=1443 cells (see Figure 3.1). The grid size is 150x150km^2 at the latitude 60°. Each cell is simultaneously an emitter and receiver. The adopted structure of modelling is shown in Figure 7.11. The input data do not involve measurements (but meteoinformation). Measurement data are used only for model verification for long periods of time (usually a month or a year).

Figure 7.12 shows the computational scheme in each grid cell at each time step equal to 1 hour.

The initial amount of pollution is calculated from the results of a previous step of calculations in the cell itself and in neighbouring cells (advection-transport along the horizontal) (see Section 7.3). This pollution is complemented by the emission amount in the cell and chemical transformation is calculated allowing for insolation (ultraviolet radiation), temperature and humidity (see Section 7.1). Then the vertical redistribution of pollution is

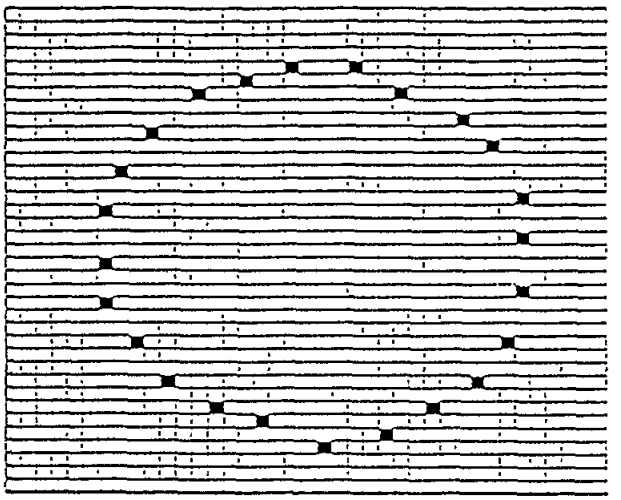


Fig.7.8. Sequential location (in every 6 hours) of isolated puff in circular vortex (angular velocity 60° per day).

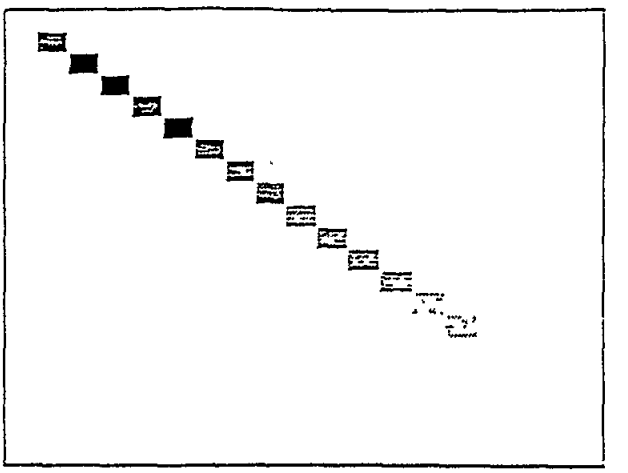


Fig.7.9. The 2-days plume from continuous source; both wind components are equal to 0.3 of grid size per hour.

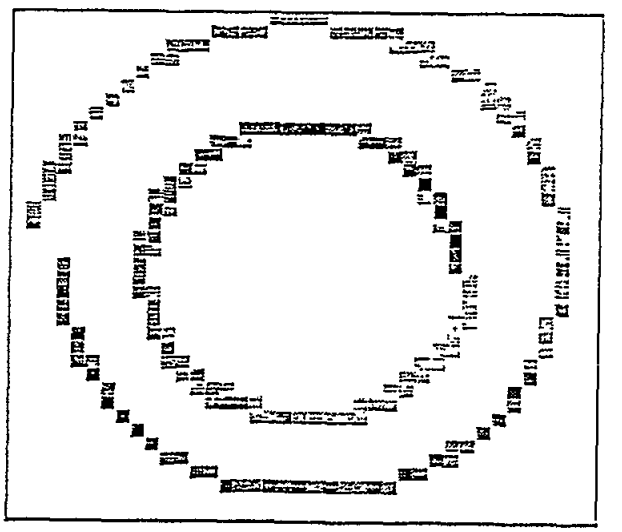


Fig.7.10. The 6-days plumes from two point continuous sources in circular vortex (angular velocity 60° per day).

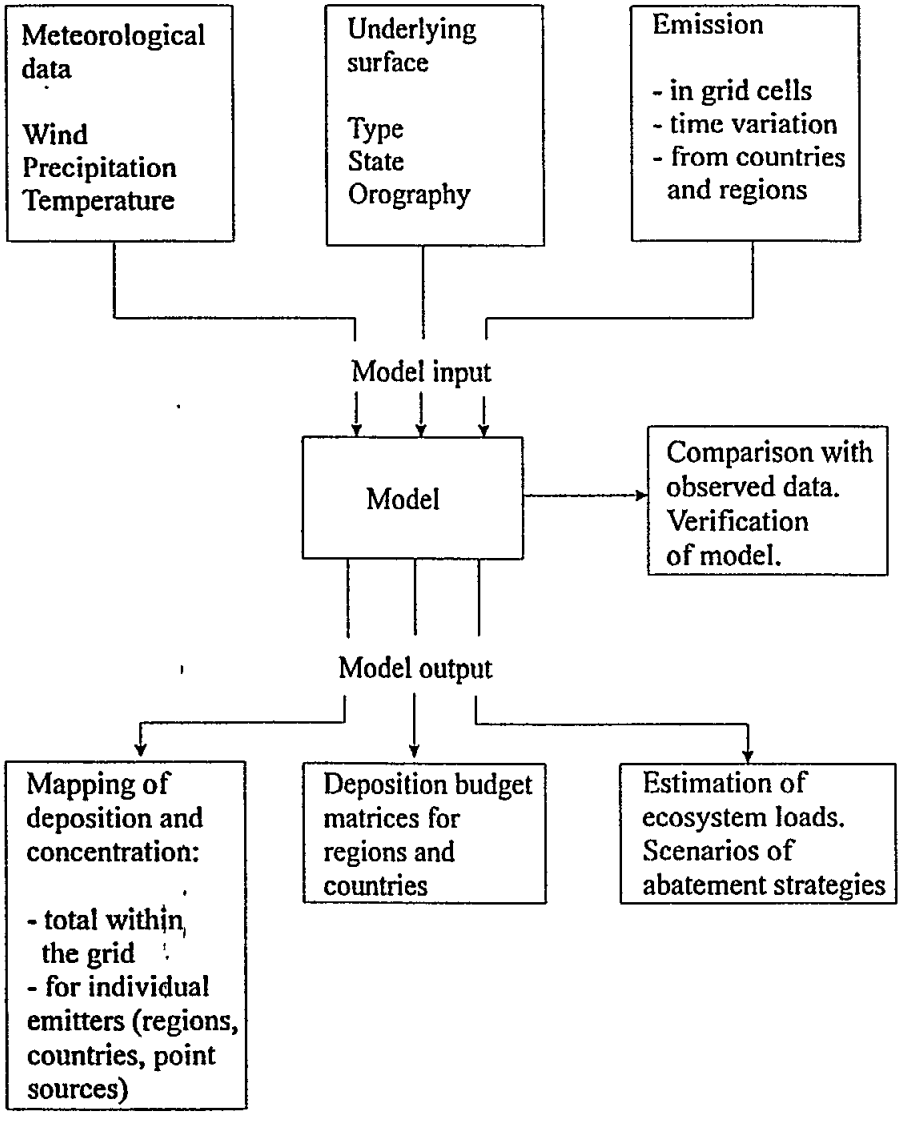


Fig.7.11. Scheme of modelling

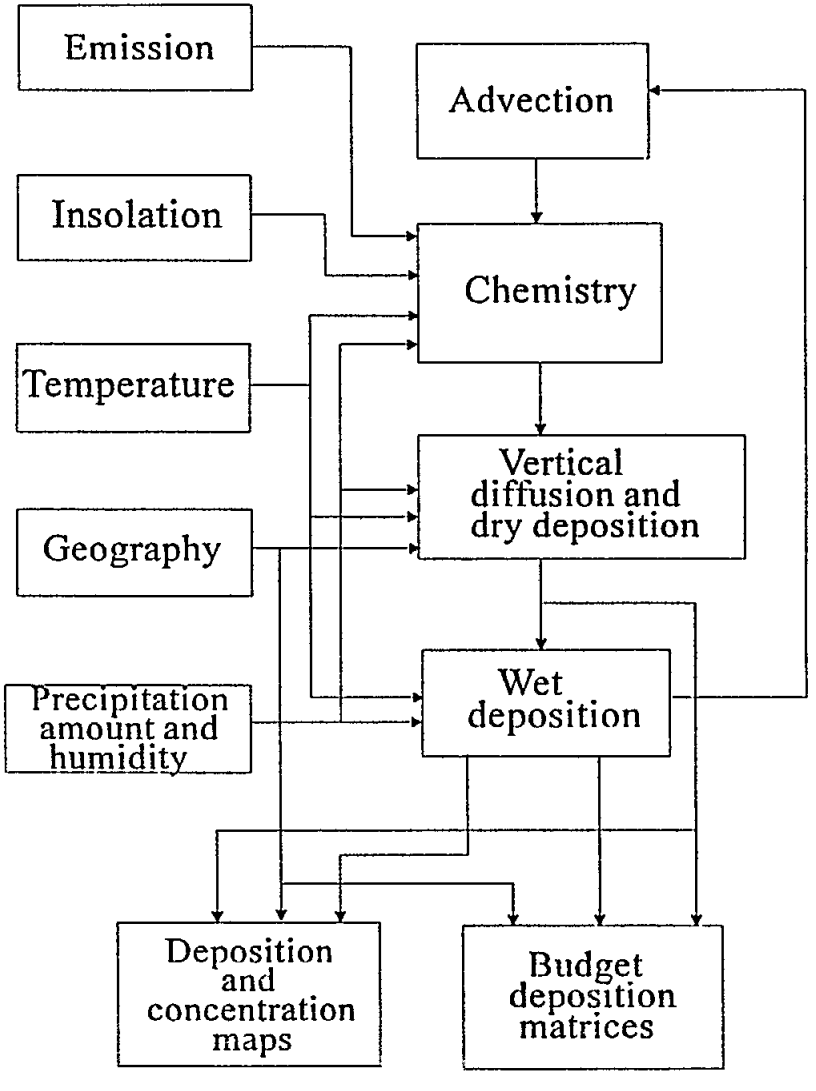


Fig.7.12. Calculation scheme

estimated accounting for the boundary layer height and the dry deposition is calculated (see Section 7.2.1). If there is precipitation, wet deposition is also calculated (see Section 7.2.2). Data on depositions and concentrations are stored for compiling the maps. Simultaneously initial data are aggregated for intercountry (or interregion) budget matrices of depositions. After that horizontal advection of pollution in the cell is calculated (see Section 7.3). All the cells are sequentially addressed within one time step. As a result initial data for the next step are prepared.

Input

Emission in the grid cell with indication of a country (region) or specific source.

In addition to emission amount it is important to have information on its temporal irregularity for the whole period being simulated by the model. The procedure of emission irregularity estimation for sulphur was developed in MSC-E in 1979-1984 [34,35]. Similar procedure was used for oxidised and reduced nitrogen. Figure 6.4 shows the plots of seasonal sulphur and nitrogen emission irregularities accepted for Europe. We believe that the emissions of volatile organic compounds have seasonal variation similar to oxidized nitrogen, that heavy metals and polyaromatic substance variations are close to those of sulphur oxides and that organochlorine emission - to ammonia.

Meteoinformation (mean values in grid cells for 6-hour periods)

- orthogonal wind components at 850 and 1000 hPa obtained from data on the geopotential and wind;
- precipitation (humidity) and their type determined from the temperature;
- temperature near the ground surface.

Real network meteorological data processed with a special objective analysis are used [36]. In cases when there is no information (oceans, deserts and so on) methods of short-term forecast are used. At present the algorithm described in [36] is improved in view of the grid expansion from the European area to the border of Northern Hemisphere regions.

Geographical information:

- underlying surface type (land, water surface, coastal line etc.);
- state of the surface estimated with allowance for meteorological information (dry, wet, etc.);
- geographical coordinates of the grid cells for the determination of height of the position of the Sun;
- identification of grid cells or their parts with countries and regions.

Output

The output data contain:

1. Maps of mean monthly and mean annual concentrations of air pollution by considered pollutants.
2. Maps of maximum ozone concentrations or other substances (if there is a necessity).
3. Maps of mean monthly and mean annual concentrations of scavenged pollutants.
4. Maps of depositions (total and for individual emitters).
5. Tables-matrices of country-by-country (region-by-region) deposition. These matrices also contain depositions on regions with negligible emissions (Arctic, seas etc.) as well as amounts and directions of the pollutant transport outside the calculated grid.

The verification procedures include a detailed regression analysis made with special methods. These techniques have been developed in view of analysed data peculiarities

[37-39]. The results of a comparison of calculated and measured data are given in Chapter XI.

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VIII. TRANSPORT AND DEPOSITION OF SULPHUR COMPOUNDS ON THE MEDITERRANEAN SEA.

8.1. Deposition on the Mediterranean Sea.

In 1991 the deposition of sulphur compounds amounted to 1393 kt (recalculated for sulphur). It is 7% of their total emissions within the calculation grid. Annual deposition density of these compounds is about 0.6 g S/m^2 year, it is two times lower than the mean density in Europe or in the Baltic Sea.

The deposition map of sulphur compounds for 1991 is given in Figure 8.1, maps of their dry and wet depositions, and surface concentrations of SO_2 and SO_4 as well as SO_4 concentrations in precipitation are given in Figures A1,A6 and A8, Appendix A.

Table 8.1 shows sulphur depositions from countries-emitters covered by the calculated grid on the Mediterranean Sea (column MDT), its subbasins (columns MT1-MT10) and adjacent seas (columns MAR, BLC, AZS). Line OC (other countries) in Table 8.1 and similar one in Tables 9.1 and 9.2 shows total deposition from countries within the calculated region, contributions of each is negligible. Figure 8.2 gives per cent contributions of countries to total deposition of sulphur and nitrogen compounds.

As it is seen from Table 8.1 and Figure 8.2, Italy, Spain (30% and 17% of total deposition), Greece, Bulgaria and the former Yugoslavia (8%,8% and 7% of total deposition respectively) make the main contribution to sulphur deposition on the Mediterranean Sea. Maps of deposition caused by these countries (within the calculation region) are given in Figures B1, B2 and B3, Appendix B.

It is worth noting that the input of Mediterranean countries is 1046 kt S, i.e. 23% of the total emissions of these countries and 75% of total deposition on the sea.

Thus the input of remote countries (not Mediterranean countries within the calculation region) to the sulphur deposition is 25%, including Bulgaria - 8%, Romania - 4% and Germany - 3%.

8.2. Deposition on the Mediterranean Sea subbasins.

Figure 8.3 gives the distribution of deposition of sulphur and nitrogen compounds on the Mediterranean Sea subbasins. It is evident that the maximum deposition of sulphur compounds is the subbasin MT2 (north-western part) and minimum - in MT1 (Alboran).

From Figure 8.4 presenting the distribution of deposition density of sulphur and nitrogen compounds with subbasins and the map of deposition (Figure 8.1) it is clear that the highest density of sulphur deposition is observed in the subbasins MT5 (the Adriatic) and MT8 (the Aegean Sea). This deposition density is three times higher in the Adriatic Sea and two times higher in the Aegean Sea compared with the mean value for the Mediterranean Sea.

The minimum deposition density (two times less than the mean one) is observed in the subbasins MT10, MT9 (northern and southern Levantin) and MT7 (central part).

Thus the deposition density decreases from north to south that corresponds to the emission distribution within the calculation region, to the main atmospheric transport directions and to annual precipitation amount distribution within the Mediterranean basin.

Figures 8.5a, 8.5b, 8.6a and 8.6b show deposition and deposition densities on the Mediterranean subbasins from countries making the main contribution to the sea pollution.

Figures 8.5a, 8.5b, 8.6a and 8.6b, Table 8.1 and maps B1-B2, Appendix B, indicate that the main countries-emitters have their own "impact-zones". For example, Spanish sources affect the western part of the sea (subbasins MT1-MT3), Italy - the central

Table 8.1

Sulphur deposition on the Mediterranean Sea, its subbasins and adjacent seas in 1991 from some countries-emitters (1000 t S per year) .

	MT1	MT2	MT3	MT4	MT5	MT6	MT7	MT8	MT9	MT10	MAR	BLC	AZS	MDT
AL	0	0	0	1	4	5	2	3	0	1	0	1	0	16
BG	0	1	1	4	7	10	10	57	5	14	5	43	2	109
CS	0	5	2	4	0	0	0	0	0	0				11
FR	0	26	9	10	3	2	3	0	0	0	0	1	0	53
GE	0	12	4	5	7	2	2	2	0	1	0	15	1	35
GR	0	0	0	1	2	12	13	56	5	18	0	4	0	107
HU	0	3	1	3	7	2	2	3	1	1	1	11	1	23
IT	0	76	20	109	87	59	44	13	2	6	0	5	0	416
PL	0	2	0	2	5	1	1	3	1	1	1	12	1	16
PT	1	1	1	0	0	0	0	0	0	0	0	1	0	3
RO	0	2	1	3	5	6	5	19	2	7	4	80	5	50
ES	14	117	67	25	5	4	7	1	0	0	0	0	0	240
TR	0	0	0	0	0	0	1	13	10	10	3	21	1	34
GB	0	2	1	2	0	0	1	0	0	0	0	0	0	6
YU	0	7	2	14	37	11	8	10	1	2	1	14	2	92
RF	0	0	0	0	1	0	0	1	0	0	0	16	5	2
UR	0	1	0	1	2	2	2	10	1	3	2	72	32	22
MOR	3	0	1	0	0	0	0	0	0	0	0	0	0	4
ALG	4	4	12	2	0	0	1	0	0	0	0	0	0	23
TUN	0	0	2	10	1	2	11	0	0	0	0	0	0	26
LIB	0	0	0	1	0	2	9	1	0	2	0	0	0	15
SYR	0	0	0	0	0	0	0	0	1	2	0	1	0	3
LEB	0	0	0	0	0	0	0	0	1	3	0	0	0	4
ISR	0	0	0	0	0	0	0	0	0	8	0	0	0	8
EGP	0	0	0	0	0	0	0	0	1	11	0	0	0	12
OC*	3	6	6	4	11	6	7	8	5	7	2	42	2	63
Tot.	25	265	130	201	184	126	129	200	36	97	19	339	52	1393

OC* - other countries

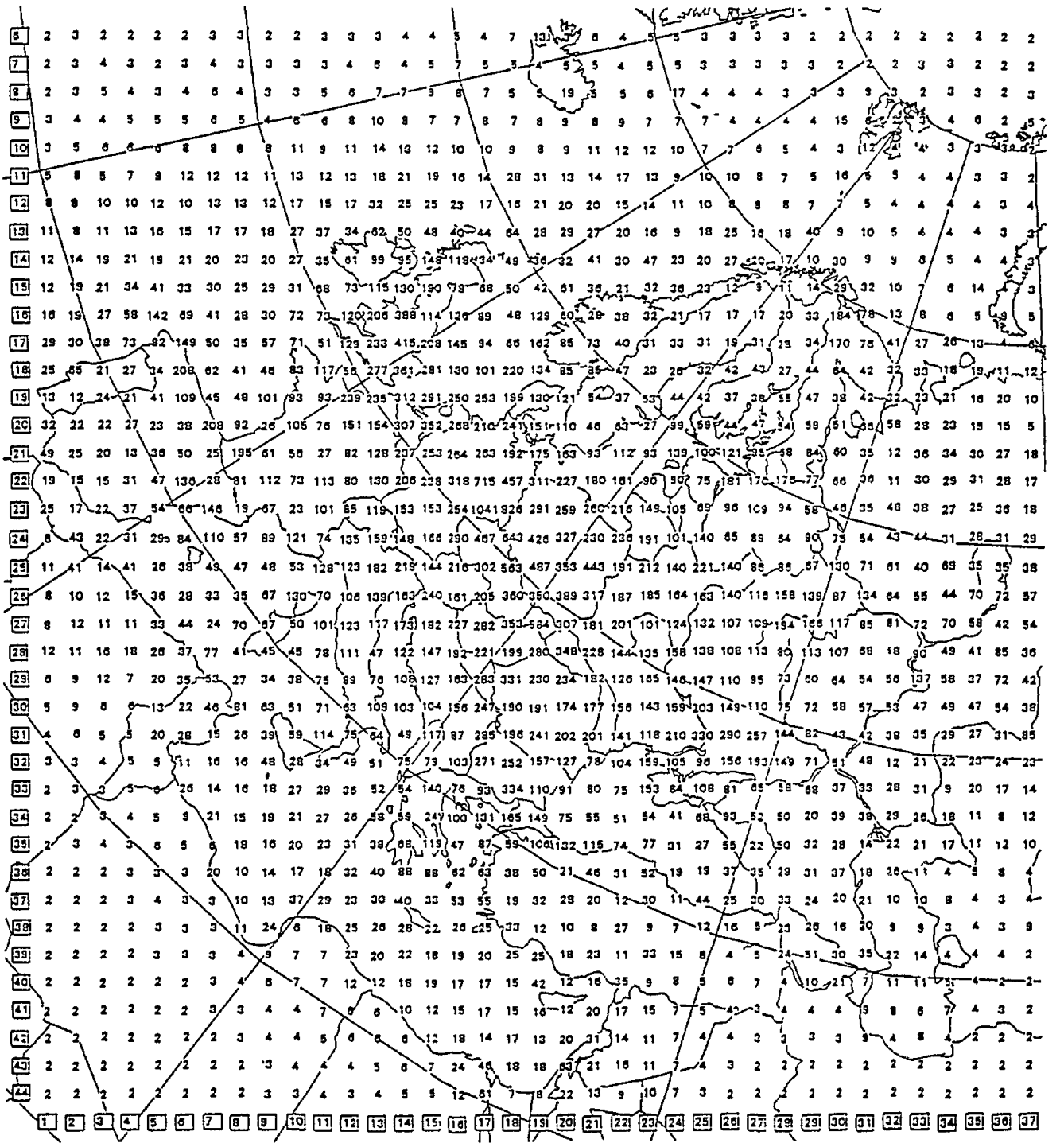


Figure 8.1. Total deposition of sulphur compounds in 1991 (10 mg S/m^2 year).

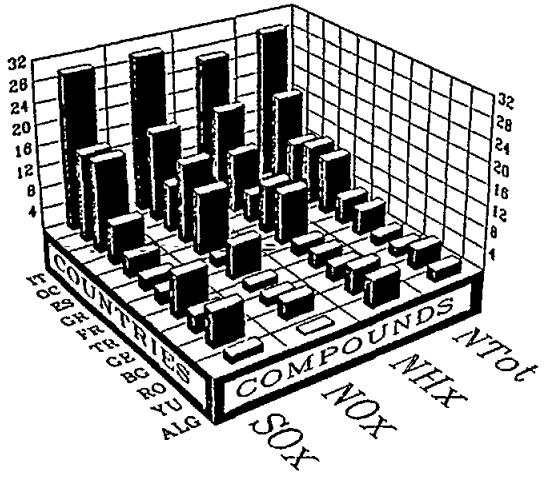


Fig.8.2 Contributions of countries to the total deposition
over the Mediterranean Sea for SOx,NOx,NHx and NTot, in per cent

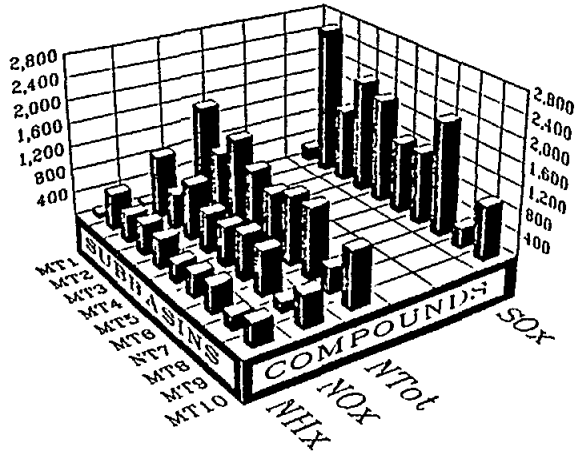


Fig.8.3 Depositions of SOx,NOx,NHx and NTot over subbasins
UNIT=100 tons S(N) for S(N) compounds Notations see Table 3 1

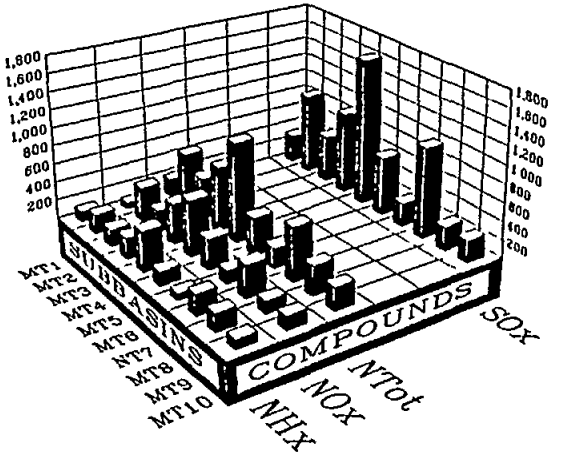


Fig.8.4 SOx,NOx,NHx,NTot deposition density over subbasins
UNIT= mg S/m2 yr (mg N/m2 yr) for S(N) compounds

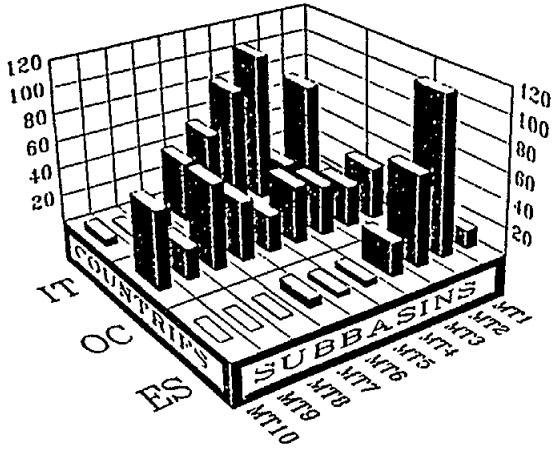


Fig.8.5a SOx deposition (from countries on subbasins)
UNIT=1000 tons S per year

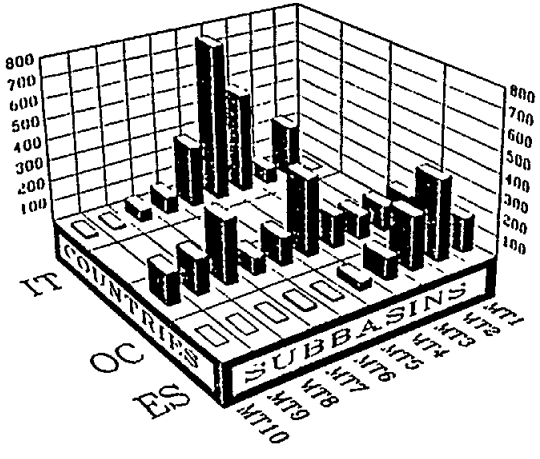


Fig.8.6a SOx deposition density (from countries over subbasins)
UNIT=mg S/m2 year

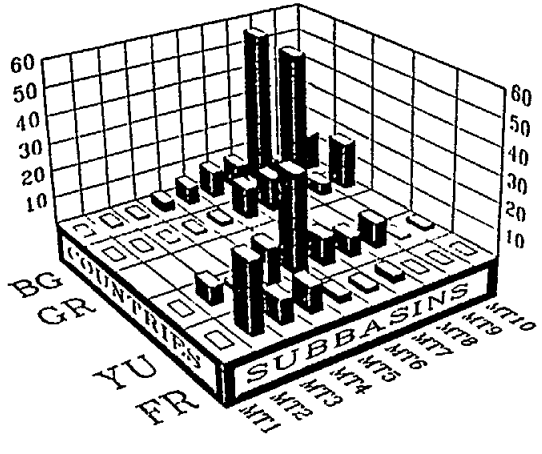


Fig.8.5b SOx depositon (from countries on subbasins)
UNIT=1000 tons S per year

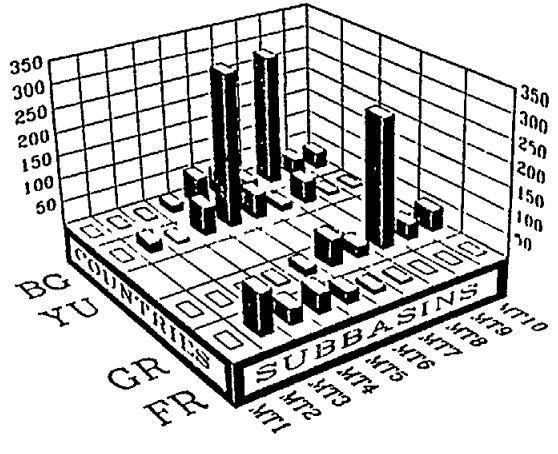


Fig.8.6b SOx deposition density (from countries over subbasins)
UNIT=mg S/m2 year

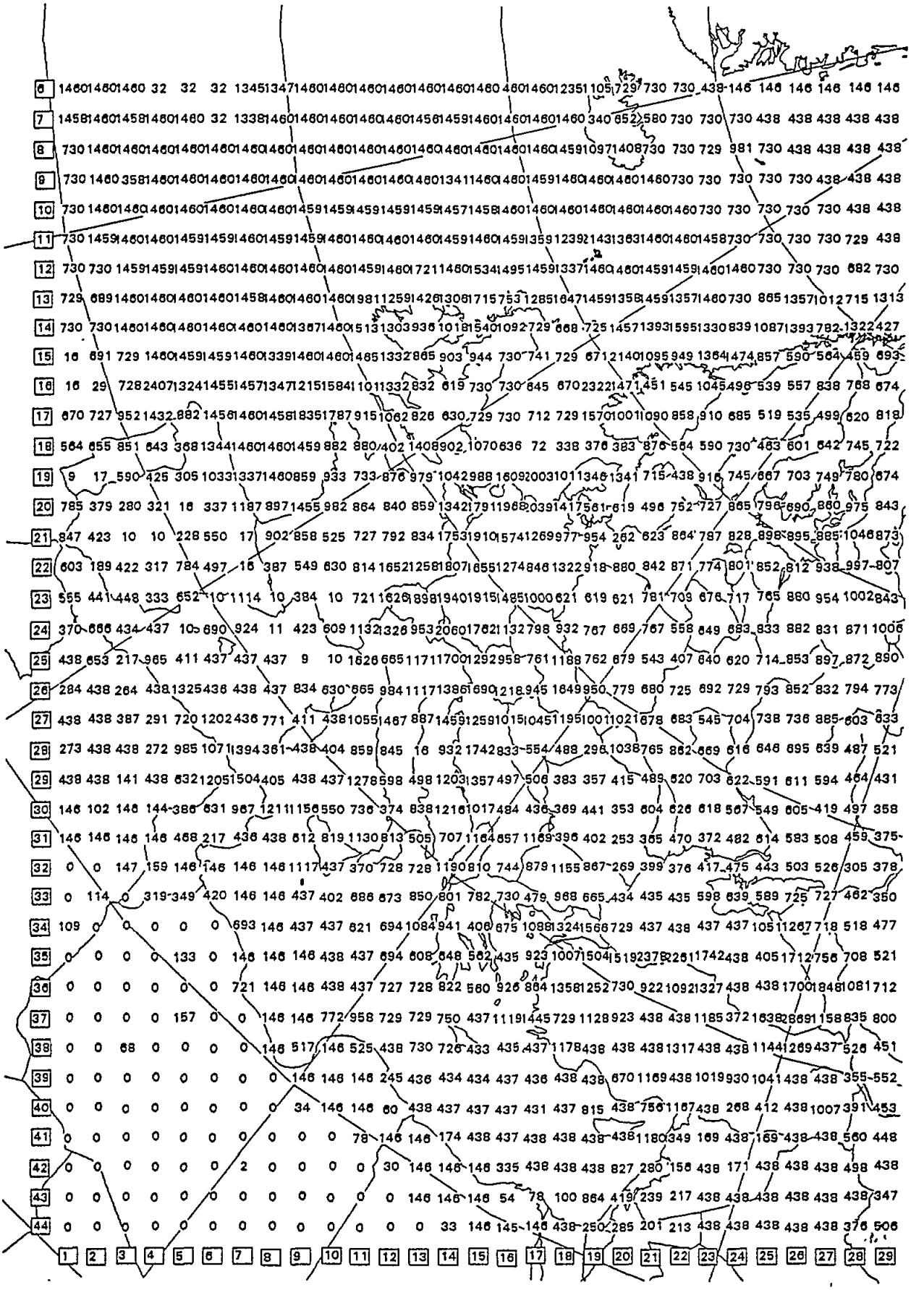


Fig.8.7. Precipitation intensity (mm/year) in grid squares in 1991.

part (subbasins MT4-MT7), and Greece, Bulgaria and Turkey - the eastern part (MT8-MT10).

Annual precipitation amount decreases eastward and southward. The map of precipitation amount for 1991 used in calculations for grid squares is given in Figure 8.7.

8.3 Sulphur deposition on the adjacent countries and seas

Sulphur deposition on adjacent seas is given in Table 8.1 (columns MAR, BLC, AZS) and on adjacent countries - in Table 9.3.

In 1991 the Marmara, the Black and the Azov Seas received in total 410 kt of sulphur compounds (recalculated as S) that approximately corresponds to 2% of total emissions of these compounds within the calculated region.

Thus on the whole in 1991 the southern European seas: the Mediterranean, the Sea of Marmara, the Black Sea and the Sea of Azov received 1803 kt of sulphur compounds (as S) that corresponds approximately to 9% of their total emissions within the calculation region.

In 1991 the adjacent countries received 2925 kt of sulphur compounds (as S) that corresponds to 14.8% of their emissions within the calculation region.

It is worth noting that the total deposition value of sulphur compounds obtained for adjacent countries (about 15% of the emissions within the calculation region) is a rough estimation of the deposition on the Mediterranean Sea watershed, where from a part of these compounds with river flows is transported to the coastal region, most sensitive to anthropogenic impact. Thus, due to the input to the marine environment of pollutants deposited on the sea watershed, the contribution of airborne pollution to the total pollution of the sea will be greater than the pollution resulted only from deposition directly on the sea.

8.4 Seasonal variations of sulphur deposition.

In MSC-E report [1] prepared in 1992 an analysis of dynamics of sulphur deposition on the Mediterranean Sea for the period of 1987-91 was made and it was shown that annual variations of deposition were small but their seasonal variations were significant.

The conclusions made on the character of seasonal variations are confirmed by the results obtained in this study. As seen from Figure 9.8 seasonal variations of deposition of sulphur compounds are rather significant in 1991. It is explained by both seasonal emission variations (Figure 6.4) and different atmospheric transport patterns and precipitation distribution for different months. Monthly precipitation amounts for January, April, July and October 1991 are given in Figures C1 and C2, Appendix C.

The deposition of sulphur compounds has a distinct minimum in June and maximum - in December that correlates with seasonal variations of sulphur emissions. The maximum exceeds the minimum more than 3 times.

Table 9.4 contains values of monthly deposition and deposition densities of sulphur compounds in the Mediterranean Sea (column MDT), in its subbasins (columns MT1-MT10) and in adjacent seas (columns MAR, BLC and AZS). Figure 9.9 gives monthly deposition distributions of sulphur compounds in subbasins. The irregularity of deposition distribution with months in individual subbasins is approximately of the same order magnitude as those observed in the Mediterranean Sea on the whole.

Maps of total depositions of sulphur and nitrogen compounds, dry and wet depositions, concentrations of various sulphur and nitrogen compounds in air and precipitation for January, April, July and October 1991 are given in Figures C3-C42, Appendix C.

References

1. Dedkova L., L. Erdman, M. Galperin, S. Grigoryan and M. Sophiev. Assessment of airborne sulphur and nitrogen pollution of the Mediterranean and Black Seas from European countries, 1987-1991. EMEP/MSC-E report 4/92, November 1992.

IX. TRANSPORT AND DEPOSITION OF NITROGEN COMPOUNDS ON THE MEDITERRANEAN SEA

9.1 Deposition on the Mediterranean Sea.

Depositions of sulphur compounds practically do not affect the content of these elements in the marine water (for the Mediterranean Sea in particular, since its salinity is sufficiently high, contrary, for example, to the Baltic Sea) but nitrogen compounds entering the water can affect more intensively than sulphur the marine biota, causing eutrophication.

In 1991 the input of oxidized nitrogen (recalculated as N) amounted to 648 kt, reduced nitrogen-419 kt. This corresponds respectively to 9.3% and 5.1% of their total emissions within the calculation grid. Thus bound (reduced plus oxidized) nitrogen deposition is 1067 kt or 7,1% of its emission.

The deposition density of nitrogen compounds over the Mediterranean Sea on the average is as follows: about 0.28 g N/m^2 year for oxidized and 0.18 g N/m^2 year for reduced nitrogen. The deposition density of the bound nitrogen is 0,46 g N/m^2 year. Similar to sulphur compounds these values are two times lower than mean values for Europe and the Baltic Sea.

Maps of oxidized and reduced nitrogen depositions are given in Figures 9.1 and 9.2, and of bound nitrogen - in Figure 9.3. Maps of dry and wet depositions of these compounds, and of concentrations of NO_2, NO_3+HNO_3 and NH_4 in air and of NO_3 and NH_4 in precipitation - in Figures A2-A5, A7, A8, Appendix A.

Tables 9.1 and 9.2 similar to Table 8.1 give deposition values of oxidized and reduced nitrogen from countries located within the calculation area. As it is seen from Tables 9.1 and 9.2 and Figure 8.2, the main contribution to the total pollution of the Mediterranean Sea by oxidized nitrogen is made by Italy (31%), Greece (15%), France (12%), Spain (9%), Germany (7%) and the former Yugoslavia (3%), by reduced nitrogen - Italy (27%), Spain (12%), Turkey (10%), France (9%), Greece (6%) and the former Yugoslavia (5%).

Italy (29%), Greece (11%), France (11%), Spain (10%), Turkey (5%) and Germany (5%) make the main input to the pollution by bound nitrogen. Maps of deposition of these compounds are given in Figures B4-B12, Appendix B.

Germany, being a remote country, gives a good example of how the atmospheric residence time of pollutants affects a relative amount of these pollutants entering the Mediterranean Sea. The deposition of "long-lived" oxidized nitrogen from Germany on the sea exceeds 4% of its emission whilst similar value for "short-lived" reduced nitrogen is less than 1%.

The contribution to deposition on the Mediterranean Sea from adjacent countries (recalculated as N) is the following: oxidized nitrogen - 397.6 kt, reduced nitrogen - 341.6 kt, bound nitrogen - 739.2 kt. These depositions correspond to 21%, 16% and 19% of total emissions from these countries. The fraction of adjacent countries in total deposition on the Mediterranean Sea is 61%, 82% and 69% respectively.

Thus, the fraction of remote countries in total deposition of "long-lived" oxidized nitrogen (39%) is about 2 times greater than similar fraction (18%) of "short-lived" reduced nitrogen and remote countries with emission amounting to about 75% of the total emission within the calculation region make a contribution to the total deposition of various nitrogen compounds within the range of 20%-40%.

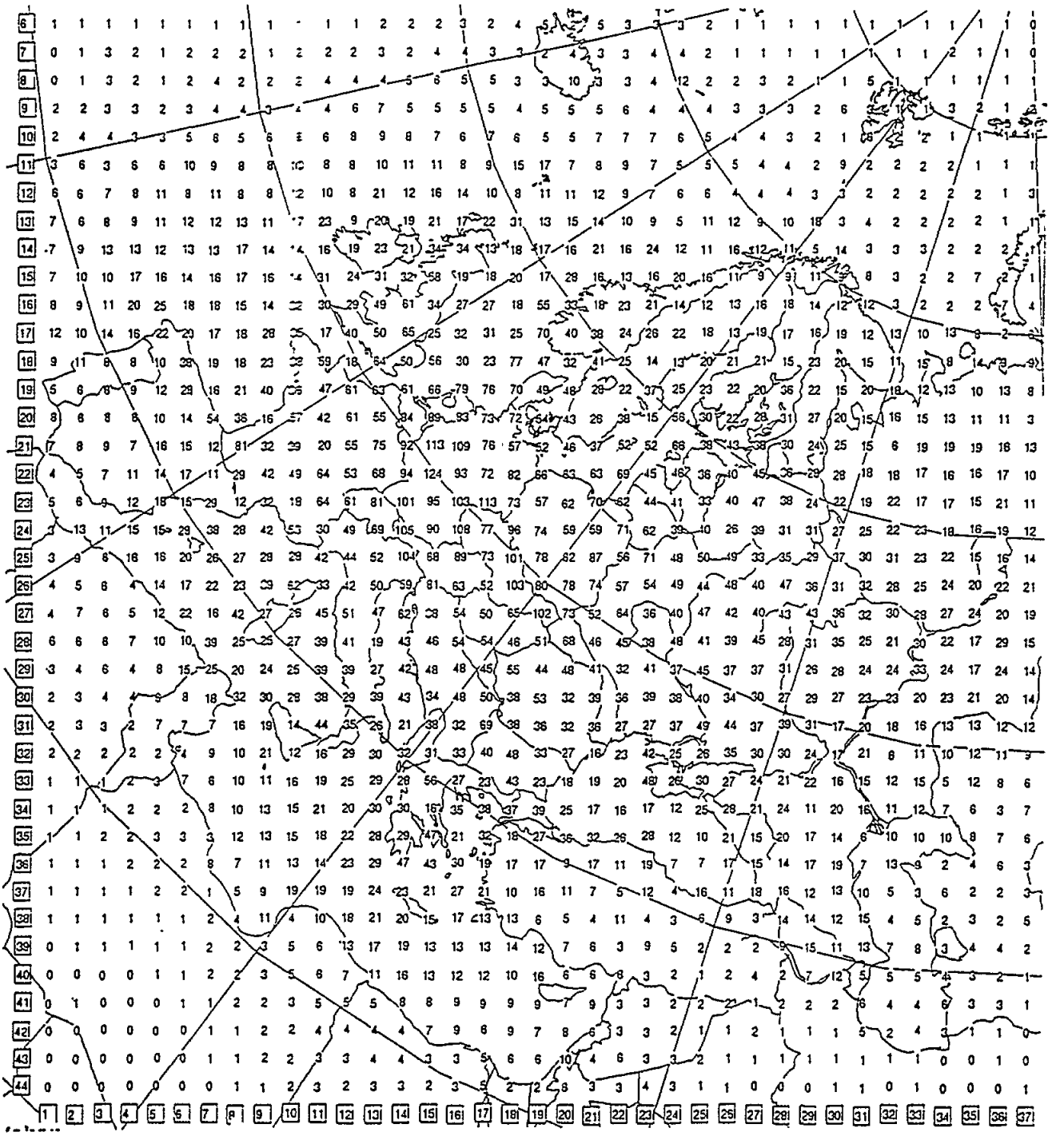


Fig. 9.1. Total deposition of oxidised nitrogen in 1991 (10 mg/m^2).

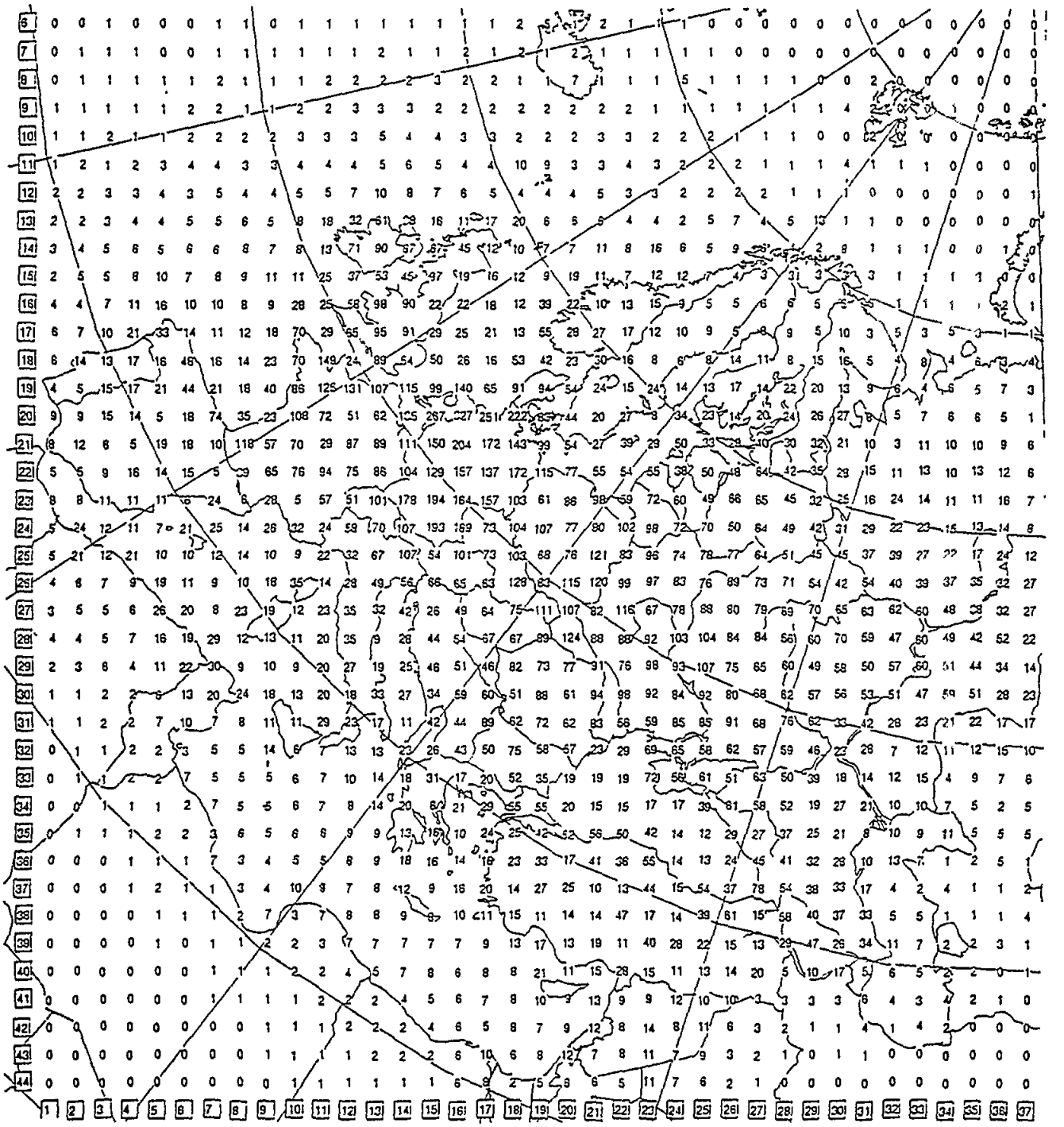


Fig. 9.2. Total deposition of reduced nitrogen in 1991 (10 mg/m^2).

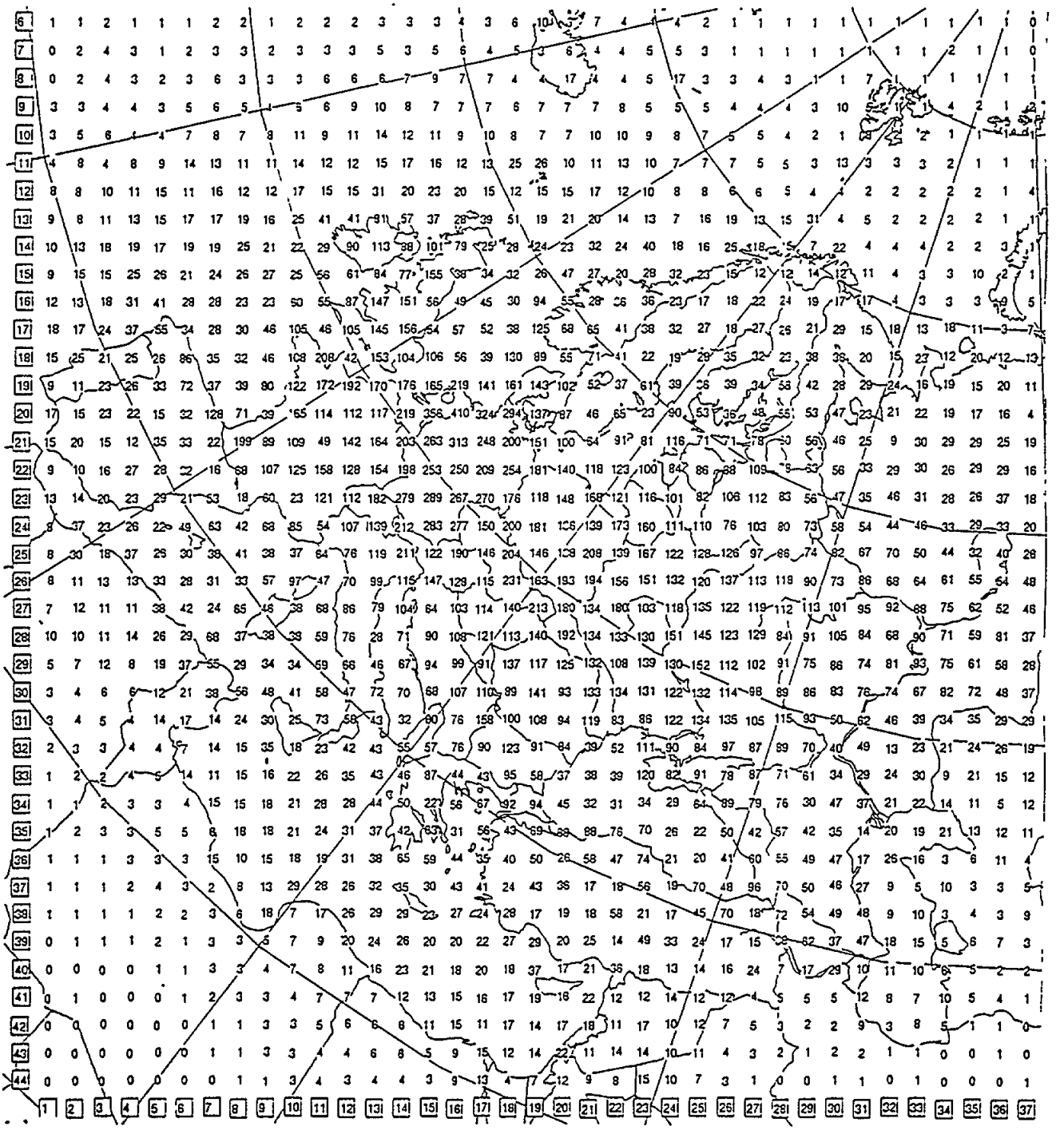


Fig. 9.3. Total deposition of bound nitrogen in 1991 (10 mg/m^2).

Table 9.1

Oxidised nitrogen deposition on the Mediterranean Sea, its subbasins and adjacent seas in 1991 from countries-emitters (100 t N per year).

	MT1	MT2	MT3	MT4	MT5	MT6	MT7	MT8	MT9	MT10	MAR	BLC	AZS	MDT
AL	0	0	0	2	6	9	6	9	1	4	0	3	0	37
AT	0	12	4	12	18	7	5	4	1	2	0	6	1	65
BE	0	9	5	4	2	1	2	1	0	0	0	2	0	24
BG	0	1	0	4	6	10	11	41	6	18	3	28	2	97
CS	0	25	9	23	28	14	11	18	4	7	2	35	3	139
FR	8	238	151	177	57	48	69	0	0	5	0	14	1	768
GE	2	136	60	80	63	28	28	0	12	7	1	54	4	422
GR	0	1	1	13	19	113	180	336	68	254	5	52	3	985
HU	0	5	2	6	15	9	6	10	2	4	1	24	2	59
IT	0	280	102	442	341	323	317	108	20	63	2	41	3	1996
NL	0	11	7	4	3	1	2	2	0	1	0	7	0	31
PL	0	9	2	14	21	14	21	14	9	19	2	54	6	123
PT	5	7	10	3	0	0	1	0	0	0	0	0	0	26
RO	0	2	1	5	8	15	14	40	7	20	6	102	8	112
ES	28	229	190	87	17	15	26	4	0	1	0	1	0	597
CH	0	28	12	20	13	5	7	1	0	1	0	2	0	87
TR	0	0	0	0	0	3	5	30	33	54	5	44	2	125
GB	3	39	27	24	6	7	12	4	1	1	1	16	1	124
YU	0	19	7	33	52	39	26	30	5	11	2	34	3	222
RF	0	1	0	1	4	4	4	13	7	12	3	128	30	46
UR	0	2	1	4	6	10	10	39	9	20	7	195	48	101
MOL	0	0	0	0	0	1	1	3	0	2	1	20	2	7
ZKV	0	0	0	0	0	0	0	2	5	5	0	47	3	12
MOR	2	0	1	0	0	0	0	0	0	0	0	0	0	3
ALG	3	6	13	4	1	1	2	0	0	0	0	0	0	30
TUN	0	1	3	6	1	3	7	0	0	0	0	0	0	21
LIB	0	0	0	1	0	2	9	1	0	2	0	0	0	15
ISR	0	0	0	0	0	0	0	0	0	15	0	0	0	15
EGP	0	0	0	0	0	0	0	0	2	13	0	0	0	15
OC*	11	35	27	28	14	15	14	16	3	13	2	55	8	176
Total	62	1096	635	997	701	697	796	754	188	554	43	964	130	6480

OC* - other countries

Table 9.2

Reduced nitrogen deposition on the Mediterranean Sea, its subbasins and adjacent seas in 1991 from countries-emitters (100 t N per year).

	MT1	MT2	MT3	MT4	MT5	MT6	MT7	MT8	MT9	MT10	MAR	BLC	AZ	MDT
AL	0	0	0	2	17	19	7	13	1	4	0	2	0	63
AT	0	4	1	4	11	2	2	1	0	1	0	3	1	26
BG	0	1	1	4	7	11	12	59	7	21	9	74	4	123
CS	0	3	1	4	9	2	2	3	1	2	0	10	1	27
FR	3	163	77	79	25	17	25	5	0	1	0	4	0	395
GE	0	33	14	16	25	5	6	3	1	1	0	31	2	104
GR	0	0	0	1	4	33	26	103	16	52	2	13	1	235
HU	0	5	3	5	15	5	5	9	2	4	2	27	3	53
IT	0	166	60	271	289	147	120	38	7	25	0	17	0	1123
PL	0	2	1	2	7	2	1	6	1	3	1	25	2	27
PT	6	5	7	2	0	0	0	0	0	0	0	0	0	20
RO	0	4	3	5	8	13	13	45	7	24	12	246	16	122
ES	49	191	156	55	11	8	14	1	0	1	0	0	0	486
CH	0	12	4	5	6	1	1	0	0	0	0	1	0	29
TR	0	0	0	0	0	3	8	88	182	140	25	432	8	421
YU	0	10	3	21	70	30	23	33	4	10	2	31	3	204
RF	0	0	0	0	1	2	1	5	2	3	1	166	71	14
UR	0	2	1	2	4	6	7	33	4	12	9	404	158	71
MOL	0	0	0	0	1	1	1	4	0	2	1	41	3	9
ZKV	0	0	0	0	0	0	0	1	1	1	0	77	3	3
ALG	32	30	126	17	2	2	7	0	0	0	0	0	0	216
TUN	0	3	19	44	4	12	48	1	0	1	0	1	0	132
LIB	0	0	1	3	1	9	53	5	2	19	0	0	0	93
ISR	0	0	0	0	0	0	0	0	0	17	0	0	0	17
EGP	0	0	0	0	0	0	0	1	3	27	0	1	0	31
OC*	8	18	15	13	6	8	23	11	16	33	3	48	6	151
Tot.	98	652	493	555	523	338	405	468	257	404	67	1654	282	4193

OC* - other countries

9.2. Deposition on the Mediterranean Sea subbassins.

Figure 8.3 shows that maximum amount of all nitrogen compounds deposits on the subbasin MT2 (north-western part) and minimum - on the subbasin MT1 (Alboran).

However, the maximum deposition densities of all nitrogen depositions (Figure 8.4) are in the subbasin MT5 (the Adriatic Sea), where the maximum densities exceed 2 times the mean ones over the Mediterranean Sea. Higher than mean densities are observed in the Tyrrhenian and the Aegean Seas.

The minimum densities are observed in the Alboran and the subbasins MT7 and MT10. They are about two times lower than corresponding mean value for the Mediterranean Sea. Thus, the deposition density of nitrogen compounds decreases from north to south.

Figures 9.4a, 9.4b, 9.5a, 9.5b. 9.6a, 9.6b and 9.7a, 9.7b show deposition and deposition densities of oxidized and reduced nitrogen in the Mediterranean Sea subbasins from countries making the greatest input to its pollution.

Figures 9.4a, b-9.7a,b, Tables 9.1 and 9.2, as well as maps B4-B12, Appendix B, show that countries-emitters have their own "impact zones". For example, Spain mainly affects western part of the sea (subbasins MT1-MT3), Italy - central part (subbasins MT4-MT7) and north-western part (subbasin MT2), Greece - the Aegean Sea and the South-Levantin, Turkey - the North-Levantin.

The distribution of deposition of both sulphur and nitrogen compounds over the subbasins is nearly the same because of similar distributions of their emissions and the same atmospheric transport patterns and annual precipitation amounts used in calculations.

9.3. Nitrogen deposition on the adjacent countries and seas.

Nitrogen deposition on adjacent seas is given in Tables 9.1 and 9.2 (columns MAR, BLC, AZS) and on adjacent countries - in Table 9.3.

In 1991 the Marmara, the Black and the Azov Seas received in total 314.6 kt of bound nitrogen compounds (recalculated as N) that approximately corresponds to 2% of total emission of these compounds within the calculation region.

In 1991 the Mediterranean Sea, the Sea of Marmara, the Black Sea and the Sea of Azov on the whole received 1382 kt of bound nitrogen compounds (as N) that corresponds approximately to 9% of the their emission within the calculation region.

In 1991 the adjacent countries received 2484 kt of bound nitrogen compounds (as N) that corresponds to 16.4% of their emissions within the calculation region.

It is worth noting that the total deposition value of nitrogen compounds obtained for adjacent countries (about 16% of the emissions within the calculation region) is a rough estimation of the deposition on the Mediterranean Sea watershed, where from a part of these compounds with river flows is transported to the coastal region, most sensitive to anthropogenic impact. Thus, due to the input to the marine environment of pollutants deposited on the sea watershed, the contribution of airborne pollution to the total pollution of the sea will be greater than the pollution resulted only from deposition directly on the sea.

For example, rough estimates made at MSC-E [2] showed that from 1500 kt of nitrogen deposited in 1991 over the Baltic Sea watershed, from 160 to 300 kt of this N (10-20%) might enter the sea through the riverine runoff in addition to 300 kt of N deposited directly to the sea surface. The total load of N entering the Baltic Sea with rivers and direct discharges from urban and industrial areas was 662 kt [3].

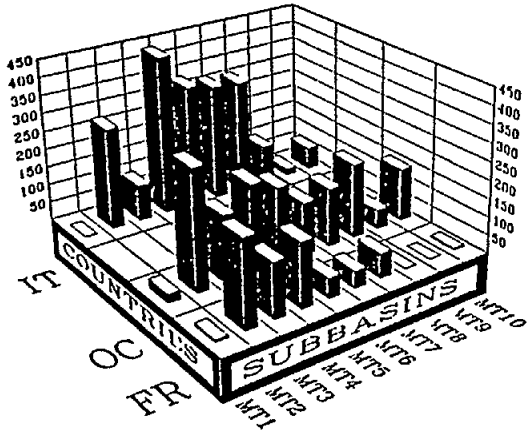


Fig.9.4a NOx deposition (from countries on subbasins)
UNIT=100 tons N per year

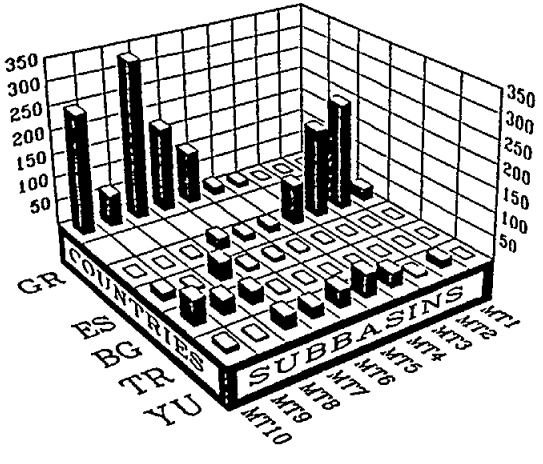


Fig. 9.4b NOx deposition (from countries on subbasins)
UNIT=100 tons N per year

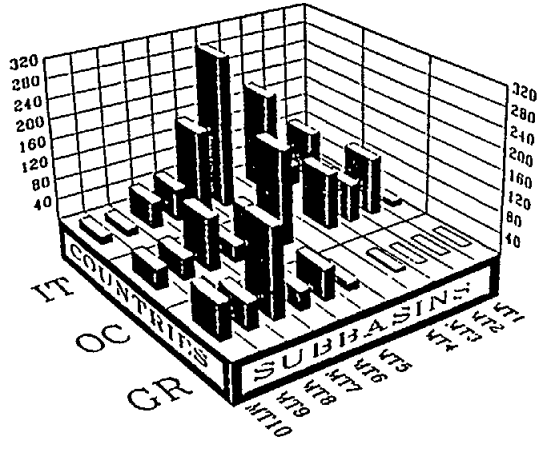


Fig.9.5a NOx deposition density (from countries over subbasins)
UNIT=mg N/m2 year

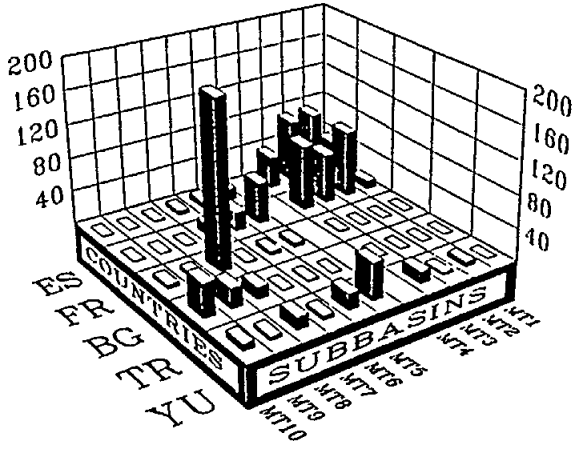


Fig.9.5b NOx deposition density (from countries over subbasins)
UNIT=mg N/m2 year

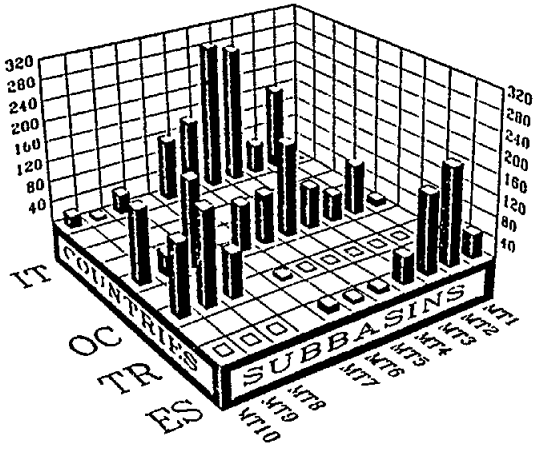


Fig.9.6a NHx deposition (from countries on subbasins)
UNIT=100 tons N per year

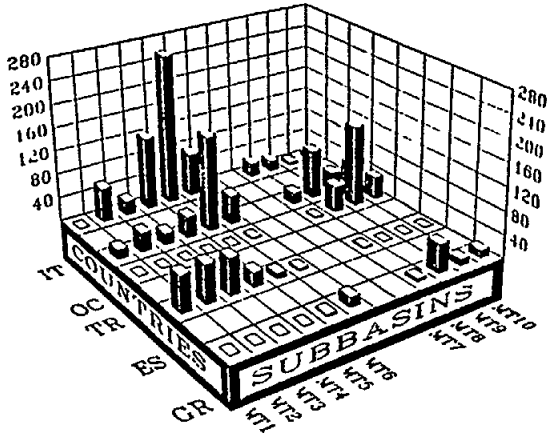


Fig.9.7a NOx deposition density (from countries over subbasins)
UNIT=mg N/m2 year

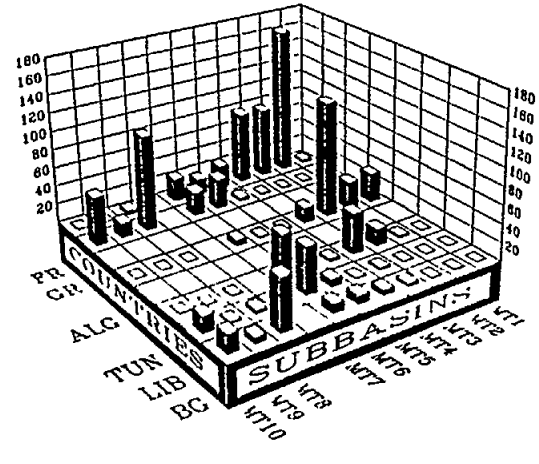


Fig.9.6b NHx deposition (from countries on subbasins)
UNIT=100 tons N per year

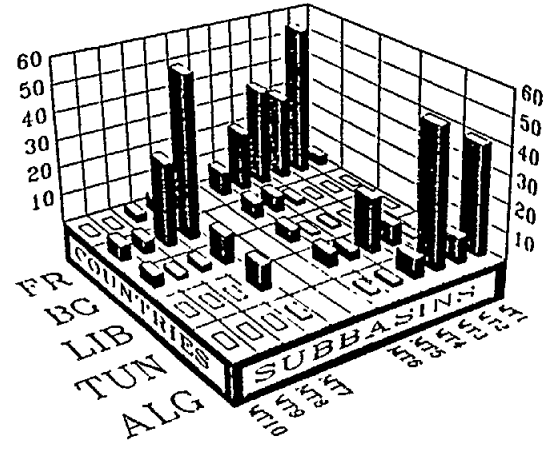


Fig.9.7b NOx deposition density (from countries over subbasins)
UNIT=mg N/m2 year

9.4. Seasonal variations of deposition.

In MSC-E report [1] prepared in 1992 an analysis of dynamics of nitrogen deposition on the Mediterranean Sea for the period of 1987-91 was made and it was shown that annual variations of deposition were small but their seasonal variations were significant.

The conclusion made on the character of seasonal variations are confirmed by the results obtained in this study. As seen from Figure 9.8 seasonal variations of deposition of all compounds in question are rather significant in 1991. It is explained by both seasonal emission variations (Figure 6.4) and different atmospheric transport patterns and precipitation distribution for different months. Monthly precipitation amounts for January, April, July and October 1991 are given in Figures C1 and C2, Appendix C.

The irregularity of nitrogen deposition is much smaller than of sulphur. The difference between the maximum and the minimum of monthly depositions of oxidized and reduced nitrogen is as much as 1.7 and of bound nitrogen - 1.5. Deposition variations from month to month have several maxima and minima. The overall maximum of deposition is observed in April-May when the reduced nitrogen emission is maximum.

Tables 9.4, 9.5 and 9.6 contain values of monthly deposition and deposition densities of sulphur compounds, oxidized and reduced nitrogen in the Mediterranean Sea (column MDT), in its subbasins (columns MT1-MT10) and in adjacent seas (columns MAR,BLC and AZS). Figures 9.9 and 9.10 give monthly deposition distributions of sulphur compounds and bound nitrogen in subbasins. The irregularity of deposition distribution with months in individual subbasins is approximately of the same order of magnitude as those observed in the Mediterranean Sea on the whole.

Maps of total depositions of sulphur and nitrogen compounds, dry and wet deposition, concentrations of various sulphur and nitrogen compounds in air and precipitations for January, April, July and October 1991 are given in Figures C3-C42, Appendix C.

References

1. Dedkova I., L. Erdman, M. Galperin, S. Grigoryan and M. Sophiev. Assessments of airborne sulphur and nitrogen pollution of the Mediterranean and Black Seas from European countries, 1987-1991. EMEP/MSC-E Report 4/92, Nov. 1992.
2. Erdman L., S. Subbotin, V. Bashkin, M. Kozlov and V. Komov. Airborne sulphur and nitrogen input to the Baltic Sea from its watershed. Provisional estimations. EMEP/MSC-E Report 7/93, May 1993.
3. HELCOM. Second Baltic Sea pollution load compilation. Baltic Sea Environment Proceedings, No. 45, 1993.

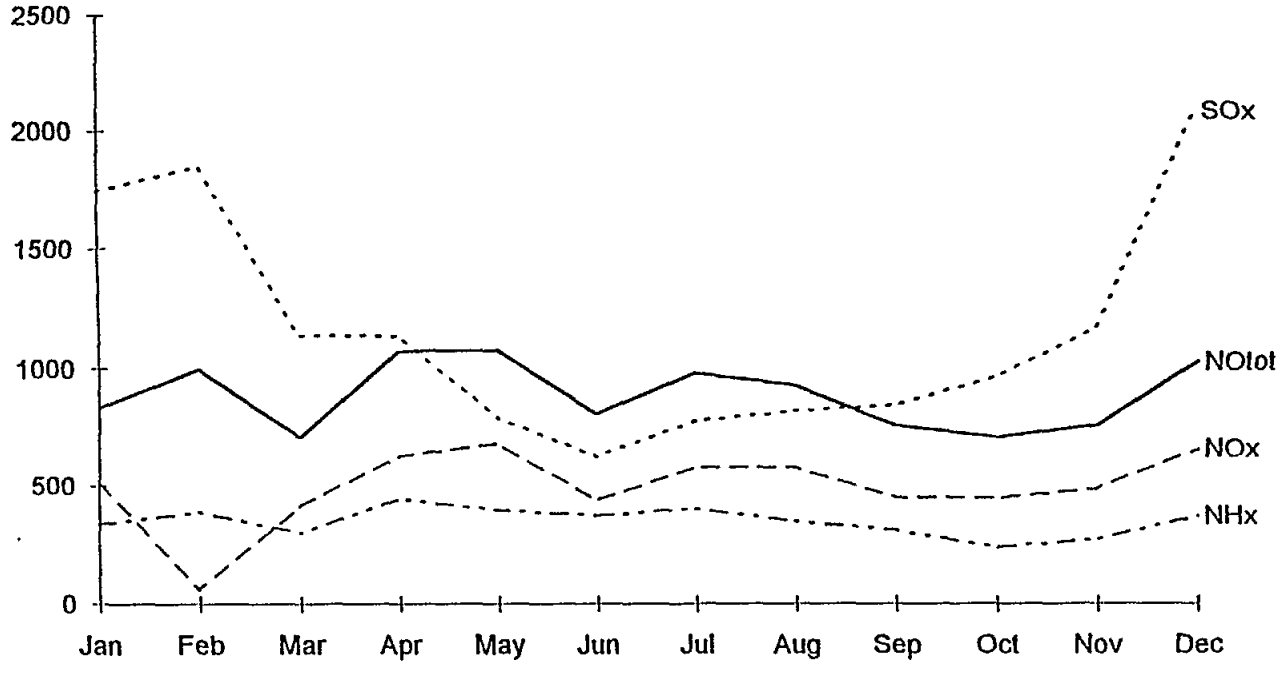


Fig.9.8. N and S compounds monthy deposition.
Unit: 100 tons per month as S or N.

Table 9.3

Deposition on the Mediterranean countries from all sources in 1991 (1000 t S per year for sulphur compounds, 1000 t N per year for nitrogen compunds).

	AL	FR	GR	IT	ES	TR	YU	MOR	ALG	TUN	LIB	SYR	LEB	ISR	EGP
NO_x	14	353	55	173	104	105	158	1	44	20	37	8	1	1	13
SO_x	39	741	154	425	370	316	570	6	113	44	63	31	4	4	45
NH_x	13	484	37	150	124	275	159	2	59	23	26	30	I	1	13
Ntot	27	837	92	323	228	380	317	3	103	43	63	38	2	2	26

Table 9.4

Monthly sulphur deposition (DEP) and deposition density (DNS) on the Mediterranean Sea, its subbasins and adjacent seas (100 t S per month for DEP, mg S/m^2, for DNS).

	MT1	MT2	MT3	MT4	MT5	MT6	MT7	MT8	MT9	MT10	MDT SUM	MAR	BCL	AZ
JAN														
DEP	28	369	162	226	222	153	146	290	43	99	1748	30	540	104
DNS	35	126	66	107	198	78	30	145	33	27	75	200	109	231
FEB														
DEP	32	415	207	310	220	129	152	245	38	82	1850	15	374	38
DNS	40	142	84	141	196	66	31	122	29	22	78	100	76	84
MAR														
DEP	31	222	104	136	110	89	89	179	54	124	1138	17	291	46
DNS	39	76	42	62	98	45	18	89	41	34	49	113	59	102
APR														
DEP	23	262	177	166	161	86	64	118	20	60	1137	7	179	43
DNS	29	90	72	75	144	44	13	59	15	16	49	47	36	96
MAY														
DEP	19	154	70	108	111	73	60	141	14	42	792	14	156	40
DNS	24	53	28	49	99	37	12	70	10	11	34	93	31	89
JUN														28
DEP	11	110	56	65	93	74	80	72	13	53	627	9	133	62
DNS	14	38	23	29	83	38	16	36	10	14	27	60	27	
JUL														
DEP	12	107	48	127	105	99	101	88	19	73	779	4	149	32
DNS	15	37	19	58	94	50	21	44	14	20	33	27	30	71
AUG														
DEP	7	144	57	96	92	129	119	89	16	73	822	6	174	33
DNS	9	49	23	44	82	66	24	44	12	20	35	40	35	73
SEP														
DEP	11	154	66	99	139	66	89	112	31	78	845	12	268	37
DNS	14	53	27	45	124	34	18	56	23	21	36	80	54	82
OCT														
DEP	26	196	73	141	132	82	74	138	27	75	964	18	222	25
DNS	32		30	64	118	42	15	69	21	20	41	20	45	56
NOV														
DEP	23	233	136	187	164	82	69	145	32	95	1166	14	254	35
DNS	29	67	55	85	146	42	14	72	25	26	50	93	51	78
DEC														
DEP	26	280	148	335	286	201	250	383	48	122	2079	42	647	62
DNS	32	96	60	152	255	103	51	191	37	33	89	280	131	138
TOT 1991 (100T)														
DEP	250	2650	1300	2010	1840	1260	1290	2000	360	970	13927	190	3390	520
DNS	312	907	526	914	1642	643	264	1000	277	264	597	1267	685	1156

Table 9.5

Monthly oxidised nitrogen deposition (DEP) and deposition density (DNS) on the Mediterranean Sea, its subbasins and adjacent seas (100 t N per month for DEP, mg N/m^2 for DNS).

	MT1	MT2	MT3	MT4	MT5	MT6	MT7	MT8	MT9	MT10	MDT SUM	MAR	BCL	AZ
JAN														
DEP	4	101	49	65	42	56	64	72	15	40	508	6	109	21
DNS	5	34	20	29	37	29	13	36	11	11	22	40	22	47
FEB														
DEP	5	126	72	116	66	46	69	66	11	33	610	3	72	7
DNS	6	43	29	53	58	23	14	33	8	9	26	20	14	16
MAR														
DEP	6	67	38	50	35	42	43	60	20	50	411	3	72	11
DNS	7	23	15	23	31	21	9	30	15	14	18	20	14	24
APR														
DEP	7	125	102	104	63	50	49	65	17	35	617	2	58	10
DNS	7	43	41	47	56	25	10	32	13	9	26	13	12	22
MAY														
DEP	11	143	76	108	66	69	60	84	19	47	683	4	68	10
DNS	14	49	31	49	59	35	12	42	15	13	29	27	14	22
JUN														
DEP	3	53	33	50	61	66	70	42	11	47	436	4	68	10
DNS	4	18	13	23	54	34	14	21	8	13	19	27	14	22
JUL														
DEP	4	77	37	111	61	94	94	49	15	55	579	2	71	10
DNS	5	26	15	50	54	48	19	24	11	15	26	13	14	22
AUG														
DEP	3	100	43	80	60	93	91	44	10	57	581	3	85	12
DNS	4	34	17	36	54	47	19	22	8	15	25	20	17	27
SEP														
DEP	2	68	36	53	67	40	54	56	16	58	450	4	87	11
DNS	2	23	15	24	60	20	11	28	12	16	19	27	18	24
OCT														
DEP	6	80	35	69	53	40	39	63	17	44	446	4	59	7
DNS	7	27	14	31	47	20	8	31	13	12	19	27	12	16
NOV														
DEP	6	73	52	89	58	37	48	62	23	40	488	3	69	8
DNS	7	25	21	40	52	19	10	31	18	11	21	20	14	18
DEC														
DEP	5	82	64	103	69	64	116	91	15	49	558	7	147	12
DNS	6	28	26	47	62	33	24	45	11	13	28	47	30	27
TOT 1991 (100T)														
DEP	62	1095	637	998	701	697	797	754	189	555	6485	45	965	129
DNS	77	375	258	454	626	356	163	377	145	151	278	300	195	304

Table 9.6

Monthly reduced nitrogen deposition (DEP) and deposition density (DNS) on the Mediterranean Sea, its subbasins and adjacent seas (100 t N per month for DEP, mg/Nm^2 for DNS).

	MT1	MT2	MT3	MT4	MT5	MT6	MT7	MT8	MT9	MT10	MDT SUM	MAR	BCL	AZ
JAN														
DEP	6	64	42	42	35	26	28	46	19	27	335	9	159	31
DNS	7	22	17	19	31	13	6	23	15	7	14	60	32	69
FEB														
DEP	7	83	54	66	42	22	30	42	18	22	386	4	109	13
DNS	9	28	22	30	37	11	6	21	14	6	16	27	22	29
MAR														
DEP	9	43	33	34	30	20	26	38	24	40	297	6	137	22
DNS	11	15	13	15	27	10	5	19	18	11	13	40	28	49
APR														
DEP	13	89	79	63	63	33	31	36	13	24	444	4	154	29
DNS	16	30	32	29	56	17	6	18	10	6	19	27	31	64
MAY														
DEP	15	69	49	53	53	32	35	52	13	23	394	6	140	35
DNS	19	24	20	24	47	16	7	26	10	6	17	40	28	78
JUN														
DEP	10	49	40	37	52	35	45	36	25	43	372	7	144	31
DNS	12	17	16	17	46	18	9	18	19	12	16	47	29	69
JUL														
DEP	9	44	31	60	41	42	47	45	30	57	401	3	133	24
DNS	11	15	13	27	37	21	10	22	23	14	17	20	27	53
AUG														
DEP	4	45	32	40	33	43	47	29	29	45	347	3	129	20
DNS	5	15	13	18	29	22	10	14	22	12	15	20	26	44
SEP														
DEP	5	40	33	33	48	19	30	31	30	41	310	5	141	22
DNS	6	14	13	15	43	10	6	15	23	11	28	33	28	49
OCT														
DEP	8	43	28	37	37	17	21	25	18	29	263	5	119	15
DNS	10	15	11	17	33	9	4	12	14	8	11	33	24	33
NOV														
DEP	6	41	37	42	38	18	19	24	18	27	270	5	102	17
DNS	7	14	15	19	34	9	4	12	14	7	12	33	21	38
DEC														
DEP	6	41	35	48	49	30	45	64	20	31	369	11	189	24
DNS	7	14	14	22	44	15	9	32	15	8	16	73	38	53
TOT 1991 (100T)														
DEP	98	651	493	555	521	337	404	468	257	404	4188	68	1656	283
DNS	122	223	200	252	465	172	83	234	198	110	179	453	334	628

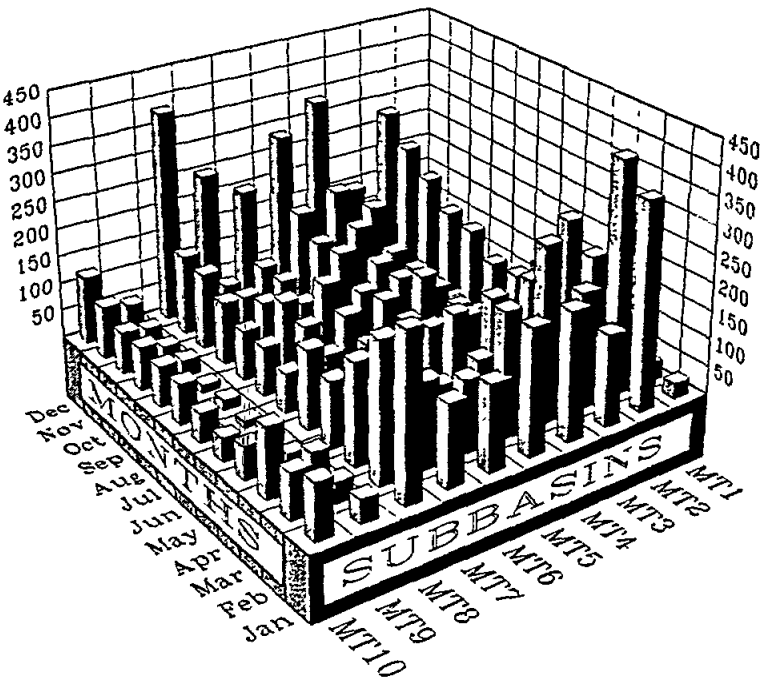


Fig.9.9 Sulphur monthly deposition on Mediterranean subbasins

Unit=100 tons per month as S

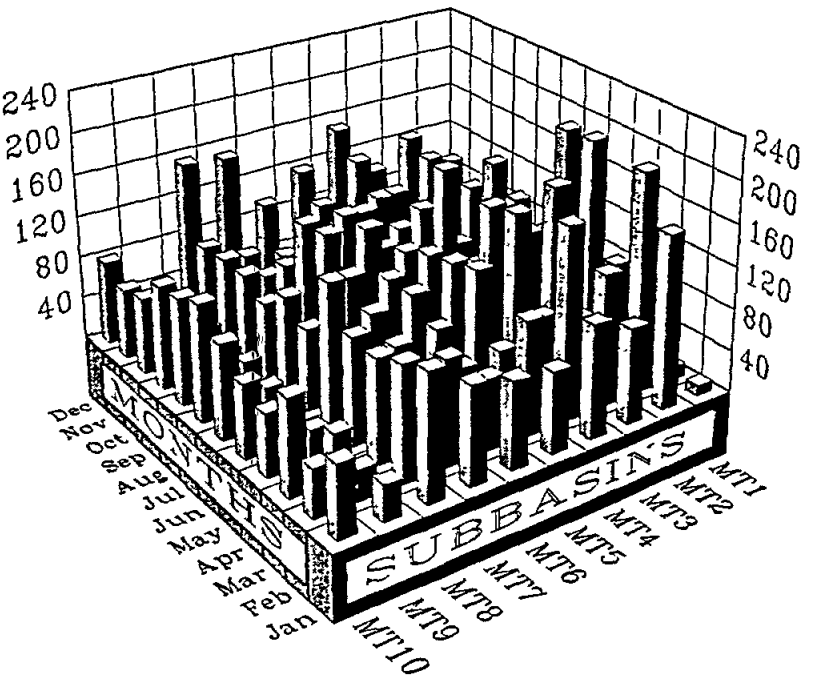


Fig 9.10 Nitrogen monthly deposition on Mediterranean subbasins

Unit=100 tons per month as N

X. TRANSPORT AND DEPOSITION OF HEAVY METALS ON THE MEDITERRANEAN SEA.

10.1 Lead.

10.1.1. Deposition on the Mediterranean Sea.

In 1991 the deposition of lead on the Mediterranean Sea amounted to 7404 t. It is 8% of its total emission within the calculation grid. Annual deposition density of lead is about 3 mg Pb/m^2 year, it is two times lower than mean density in Europe.

The deposition map of lead for 1991 is given in Figure 10.1. Maps of its dry deposition, and surface concentrations in air as well as in precipitation are given in Figures A9, A10, Appendix A.

Table 10.1 shows lead depositions from countries-emitters within the calculated grid on the Mediterranean Sea (column MDT), its subbasins (column MT1-MT10) and adjacent seas (columns MAR, BLC, AZS) and Figure 10.2 gives per cent contributions of countries to total deposition of heavy metals. Line OC (other countries) in Table 10.1 and similar one in Tables 10.3, 10.4 and 10.5 shows total deposition from countries within the calculated region, individual contributions of which are small.

As it is clear from Table 10.1 and Figure 10.2 Italy, Spain and France (40%, 11% and 10% of total deposition), Greece and Turkey (5% and 6% of total deposition respectively) make the main contribution to lead deposition on the Mediterranean Sea. Maps of deposition caused by these countries (within the calculation region) are given in Figures 13B-15B, Appendix B.

Note that the input of Mediterranean countries is 6278 t Pb, i.e. 20% of the total emission of these countries and 85% of total deposition on the sea.

Thus the input of remote countries to the lead deposition is about 15%, including Germany (2%), the Ukraine (2%) and Bulgaria (3%).

10.1.2. Deposition on the Mediterranean Sea subbassins.

Figure 10.3 gives distribution of heavy metals deposition over the Mediterranean Sea subbasins. It is evident that the maximum depositions of lead are in the subbasins MT2, MT3, MT4 and MT7 and minimum - in MT1. To a certain extent this is explained by the sizes of these subbasins.

From Figure 10.4 presenting deposition density distribution of heavy metals with subbasins and the map of lead deposition (Figure 10.1) it is clear that the greatest density of lead deposition is observed in subbasins MT5 and MT4. This deposition density is almost 3 times higher in the Adriatic Sea (MT5) and 1.5 times higher in the Tyrrhenian Sea (MT4) compared with the mean value for the Mediterranean Sea.

The minimum deposition density (almost two times less than mean one) is observed in subbasin MT1 (the Alboran Sea) and its subbassins MT7 and MT10.

Thus the deposition density is decreasing from north to south that corresponds to emission distribution within the calculation region, to main atmospheric transport routes and to annual precipitation amount distribution within the Mediterranean basin.

Figure 10.5 shows deposition of lead on the Mediterranean subbasins from countries making the main contribution to the sea pollution.

Figure 10.5, Tables 10.1 and 13B-15B (Appendix B) indicate that the main countries-emitters have their own "impact-zones" for lead. For example, Spanish sources affect the western part of the Sea (subbasins MT1-MT3). The same can be said about France (MT2-MT4 subbasins). Italy affects central part (MT4-MT7) giving 50-60 per cent of

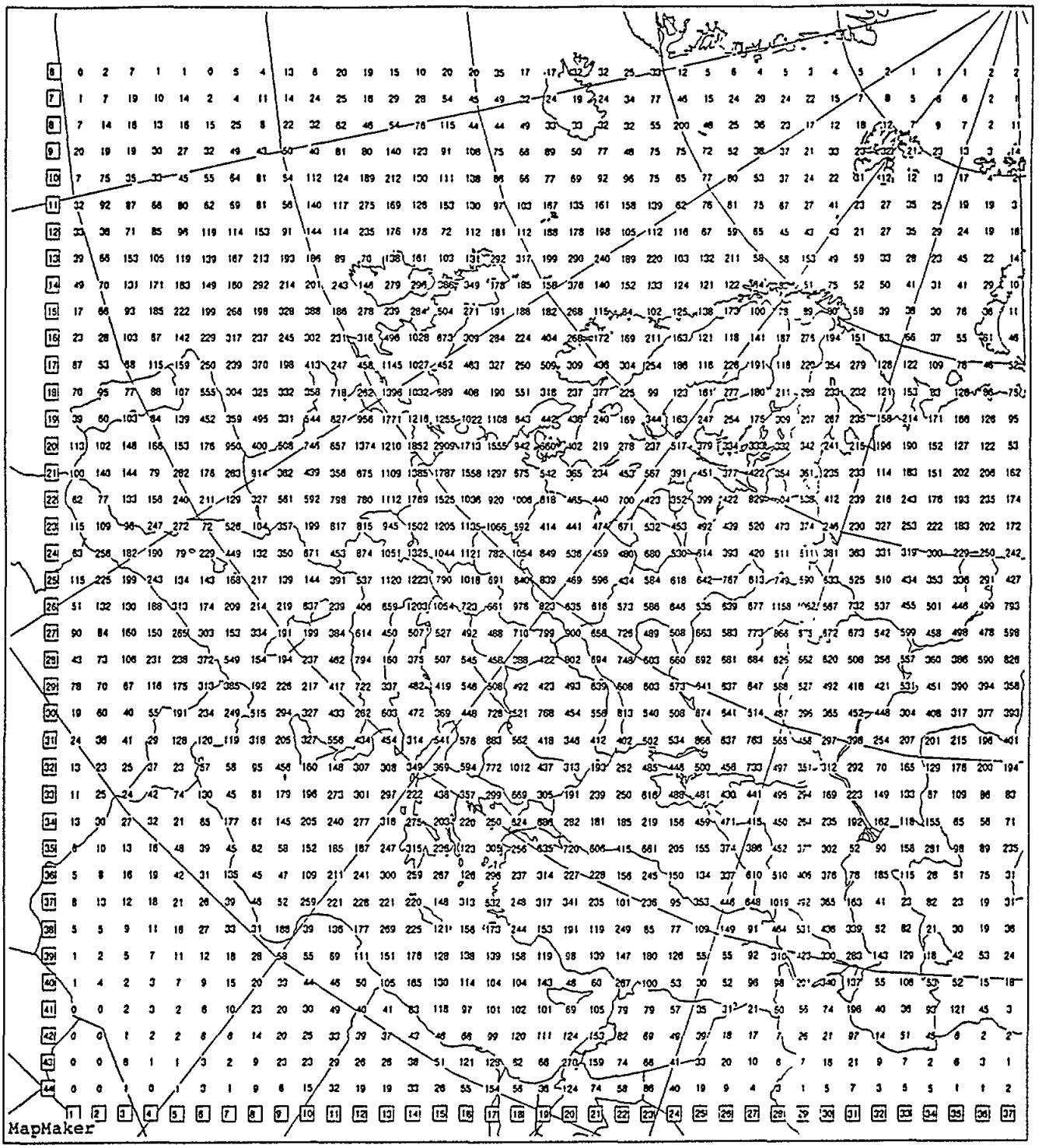


Fig.10.1. Total deposition of lead in 1991 *(10 μg Pb/m²/year).*

Table 10.1

Lead deposition on the Mediterranean Sea, its subbasins and adjacent seas in 1991 from some countries-emitters (1000 kg Pb per year)

	MT1	MT2	MT3	MT4	MT5	MT6	MT7	MT8	MT9	MT10	MAR	BLC	AZS	MDT	QA
AL	0	1	1	3	14	15	10	7	1	5	0	2	0	57	170
BG	0	3	6	6	13	28	29	103	14	41	10	74	6	243	1500
CS	0	5	2	5	9	5	4	5	1	3	1	15	1	39	1376
FR	12	247	203	152	61	31	70	6	4	5	1	17	0	791	8654
GE	0	40	33	22	21	7	9	6	3	5	1	28	1	146	7691
GR	0	1	1	4	12	50	59	124	23	100	4	27	1	374	1104
HU	0	3	2	4	9	5	4	5	1	2	1	14	1	35	657
IT	2	330	229	555	559	511	545	106	27	116	4	47	2	2980	8576
PL	0	5	1	5	13	8	6	6	3	6	1	30	2	53	3086
PT	4	5	8	2	0	1	1	0	0	0	0	0	0	21	398
RO	0	3	2	3	5	11	15	31	6	16	8	96	6	92	1327
ES	69	255	276	102	31	22	44	4	2	1	0	4	0	806	4215
TR	0	0	1	1	1	11	19	117	112	143	27	198	7	405	2220
GB	1	8	14	10	9	3	5	4	0	0	1	12	1	54	8508
YU	0	12	10	21	52	43	39	51	6	21	4	50	3	255	1950
RF	0	2	2	2	3	6	5	15	10	18	8	291	60	63	18273
UR	0	6	5	4	10	16	26	44	9	28	13	468	93	148	6825
MOR	10	1	3	1	0	0	0	0	0	0	0	0	0	15	80
ALG	35	28	110	23	6	6	14	1	0	0	0	0	0	223	1149
TUN	1	4	13	27	5	16	59	3	0	3	0	1	0	131	338
LIB	0	0	1	6	2	14	47	3	3	10	0	1	0	86	399
CYP	0	0	0	0	0	0	0	0	4	2	0	0	0	6	26
SYR	0	0	0	0	0	0	0	0	9	8	0	6	0	17	565
LEB	0	0	0	0	0	0	0	0	11	14	0	2	0	25	325
ISR	0	0	0	0	0	0	0	0	2	35	0	0	0	37	440
EGP	0	0	0	0	0	0	1	4	9	56	1	2	0	70	832
OC	1	40	26	31	36	14	22	24	14	24	6	329	24	232	15115
Total	135	999	949	989	871	823	1033	669	274	662	91	1714	208	7404	95799

Notations are given in tables 6.2 and 3.1

OC - other countries within calculation region

QA - emission of given country

Table 10.2

Deposition on the Mediterranean countries from all sources in 1991 (100 kg Zn, 10 kg Cd, 100 kg As, 1000 Pb per year)

	AL	FR	GR	IT	TR	ES	YU	MOR	ALG	TUN	LIB	SYR	LEB	ISR	EGP
Zn	746	20268	3226	6086	8910	10887	10005	94	3316	1555	1290	678	30	37	493
As	66	1242	281	406	830	619	1181	13	226	108	116	54	1	1	54
Cd	195	3779	1091	1542	2536	3731	3897	30	894	321	645	193	9	7	210
Pb	192	5146	648	2307	2149	1382	1933	27	958	402	473	220	15	19	207

Notations are given in tables 6.2 and 3.1

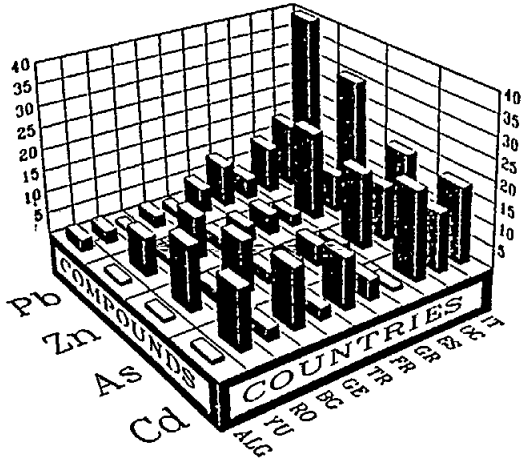


Fig.10.2 Contributions of countries to the total deposition
over the Mediterranean Sea for heavy metals,in per cent

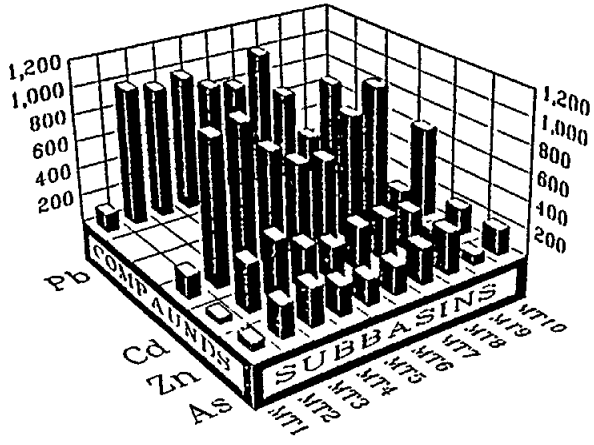


Fig.10.3 Depositions of heavy metals over subbasins
UNITS (per year): 10 kg Cd , 100 kg As , 1000 kg Pb and Zn

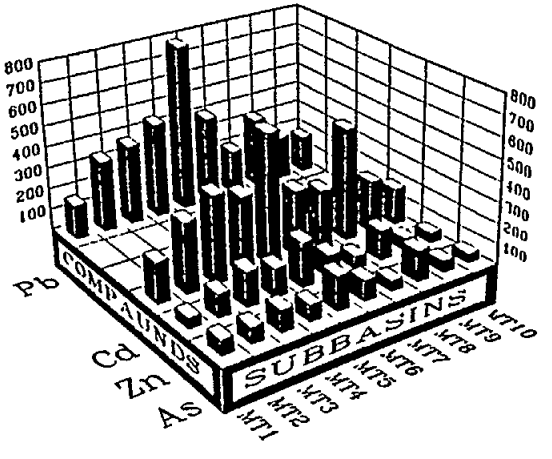


Fig. 10.4 Heavy metals deposition density over subbasins
UNITS(per y_ar). 0 1 ug Cd/m2, 1 ug As/m2, 10 ug Pb (Zn) / m2

deposition on it and in addition takes significant part in lead pollution of the other subbasins (excluding MT1 and MT9), giving 15-30 per cent of deposition.

Eastern subbasins (MT8-MT10) are polluted mainly by Turkey, Greece and Bulgaria.

10.1.3. Lead deposition on adjacent countries and seas.

Lead deposition on adjacent seas is given in Table 10.1 (columns MAR, BLC, AZS) and on adjacent countries - in Table 10.2 containing other heavy metals too.

In 1991 in total the Marmara, the Black and the Azov Seas received 2013 t of airborne lead that approximately corresponds to 2% of its total emission within the calculation region.

Thus on the whole in 1991 the area of regional seas: the Mediterranean, the Sea of Marmara, the Black Sea and the Sea of Azov received 9417 t of lead that corresponds approximately to 10% of its emission within the calculation region.

In 1991 the adjacent countries received 16078 t of lead that corresponds to 17% of its emission within the calculation region.

The total deposition value of 16078 t obtained for adjacent countries (about 17% of the emission within the calculation region) is an estimation of the deposition of lead on the Mediterranean Sea watershed, where from a part of it with river flows is transported to the coastal region, most sensitive to anthropogenic impact. Thus. due to the input to the marine environment of pollutants deposited on the sea watershed, the contribution of airborne pollution to the total pollution of the sea will be greater than the pollution resulted only from deposition directly on the sea.

10.1.4. Seasonal variations of lead pollution.

Seasonal variations of depositions and concentrations of pollutants depend upon a number of reasons. One can notice the dependence both on seasonal emission variation and the meteoelements fluctuations, such as characteristic airflows or mixing height layer in considered points. As indicated in the work [1, p.43], the factors of primary importance are presence or absence of precipitation. The data brought forward in the same work (observations made in 1985-86) show, that concentrations of Pb were 3-5 times higher in summer, than that in winter. The phenomenon is explained by the existence of wet and dry seasons. Impacts of local winds (breezes) and local orography [1, p.213] are of a great importance as well.

Four arbitrary points in the various regions of the Mediterranean have been chosen for the description of the calculation results: area of Barcelona (P1), northern Corsica (P2), eastern Crete (P3) and coastal waters in the area of Tel Aviv (P4)(Figure 3.3).

Seasonal variations of depositions and concentrations in air of Pb in the chosen grid units are shown in Figures 10.6 and 10.7 respectively. The maps for the same values for "the central" months of the seasons are presented in Figures C43-C46, Appendix C.

As it follows from Figure 10.6 the summer minimum of depositions in Barcelona, Corsica and Tel Aviv is observed during the period of the precipitation minimum.

Concentration fluctuations are insignificant in the area of Barcelona (Figure 10.7) where the ratio between the maximum and minimum values is 1.5.

As for the other points, these ratios vary from 2.5 (P4) to 4 (P2).

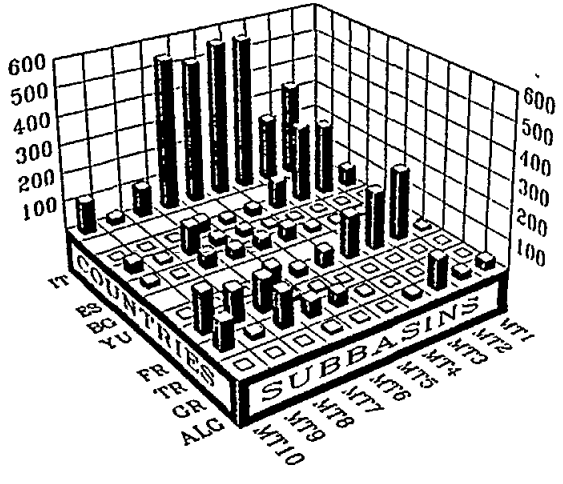


Fig.10.5 Pb deposition (from countries on subbasins)
UNIT=1000 kg Pb per year

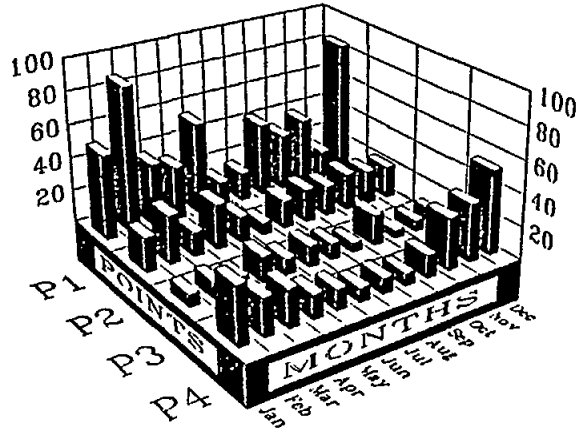


Fig 10.6 Lead monthly deposition in chosen points
Unit=10 ug/m2 P1-Barcelona,P2- Corsica,P3- Crete,P4-Tel Aviv

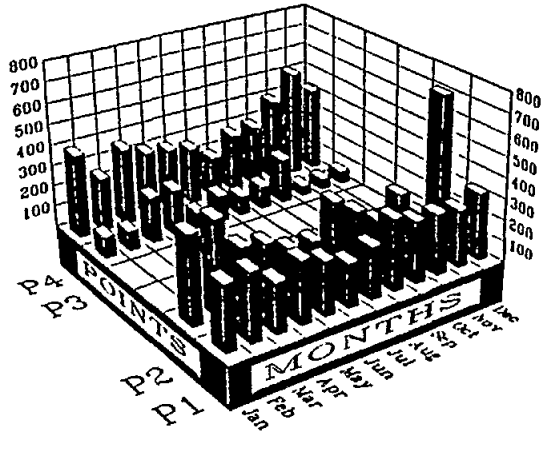


Fig 10.7 Lead monthly air concentrations in chosen points
UNIT=0 1 ng/m3 P1-Barcelona,P2- Corsica,P3-Crete,P4-Tel Aviv

10.2. Zinc.

10.2.1. Deposition on the Mediterranean Sea.

In 1991 the deposition of zinc on the Mediterranean Sea amounted to 2523 tons. It is 6% of its total emission within the calculation grid. Annual deposition density of zinc is about 1 mg Zn/m^2 year, it is nearly two times lower than mean density in Europe.

The total deposition map of zinc for 1991 is given in Figure 10.8, maps of dry depositions and surface concentrations of Zn in air as well as in precipitations are given in Figures A11 and A12, Appendix A.

Table 10.3 shows zinc depositions from countries-emitters within the calculated grid on the Mediterranean Sea (column MDT), its subbasins (columns MT1-MT10) and adjacent seas (columns MAR, BLC, AZS). Line OC (other countries) in Table 10.1 and similar one in Tables 10.3, 10.4 and 10.5 shows total deposition from countries within the calculated region, individual contributions of which are small.

As it is seen from Table 10.3 and Figure 10.2 Italy and Spain (30% and 21% of total deposition), Bulgaria, Yugoslavia and France (10%, 9% and 5% of total deposition respectively) make the main contribution to zinc deposition on the Mediterranean Sea. Maps of deposition caused by these countries within the calculation region are given in Figures B16-B18, Appendix B.

The input of the Mediterranean countries is 1902 t Zn, i.e. 15% of the total emission of these countries and 75% of total deposition on the sea.

Thus the input of remote countries to the zinc deposition is about 25%, including Bulgaria (10%), Poland (3%) and Germany (3%).

10.2.2. Deposition on the Mediterranean Sea subbassins.

As it follows from Figure 10.3 the maximum depositions of zinc are in the subbasins MT2, MT3 and MT7 and the minimum - in MT1.

From Figure 10.4 presenting the density distribution of heavy metals depositions over subbasins and the map of zinc deposition (Figure 10.8) it is clear that the greatest density of zinc deposition is observed in subbasins MT5 and MT3. This deposition density is nearly 3 times higher in the Adriatic Sea and in MT3 compared with the mean value for the Mediterranean Sea.

The minimum deposition density (almost two times less than mean one) is observed in subbasins MT1 and MT10.

Thus, the deposition density is decreasing from north to south too, that corresponds to the emission distribution within the calculation region, to main atmospheric transport routes and to annual precipitation amount distribution within the Mediterranean basin.

Figure 10.9 shows deposition on the Mediterranean subbasins from countries making the main contribution to the sea pollution.

Figure 10.9, Table 10.3 and maps B16-B18 (Appendix B) indicate that the main countries-emitters have their own "impact-zones" for zinc. For example, Spain sources affect the western part of the Sea (subbasins MT1-MT3), Italy - central part (subbasins MT4-MT7), and Bulgaria, Turkey and Yugoslavia - eastern part (MT8-MT10).

10.2.3. Zinc deposition on adjacent countries and seas.

Zinc deposition on adjacent seas is given in Table 10.3 (columns MAR, BLC, AZS) and on adjacent countries - in Table 10.2 containing other heavy metals too.

In 1991 in total the Marmara, the Black and the Azov Seas received 792 t of

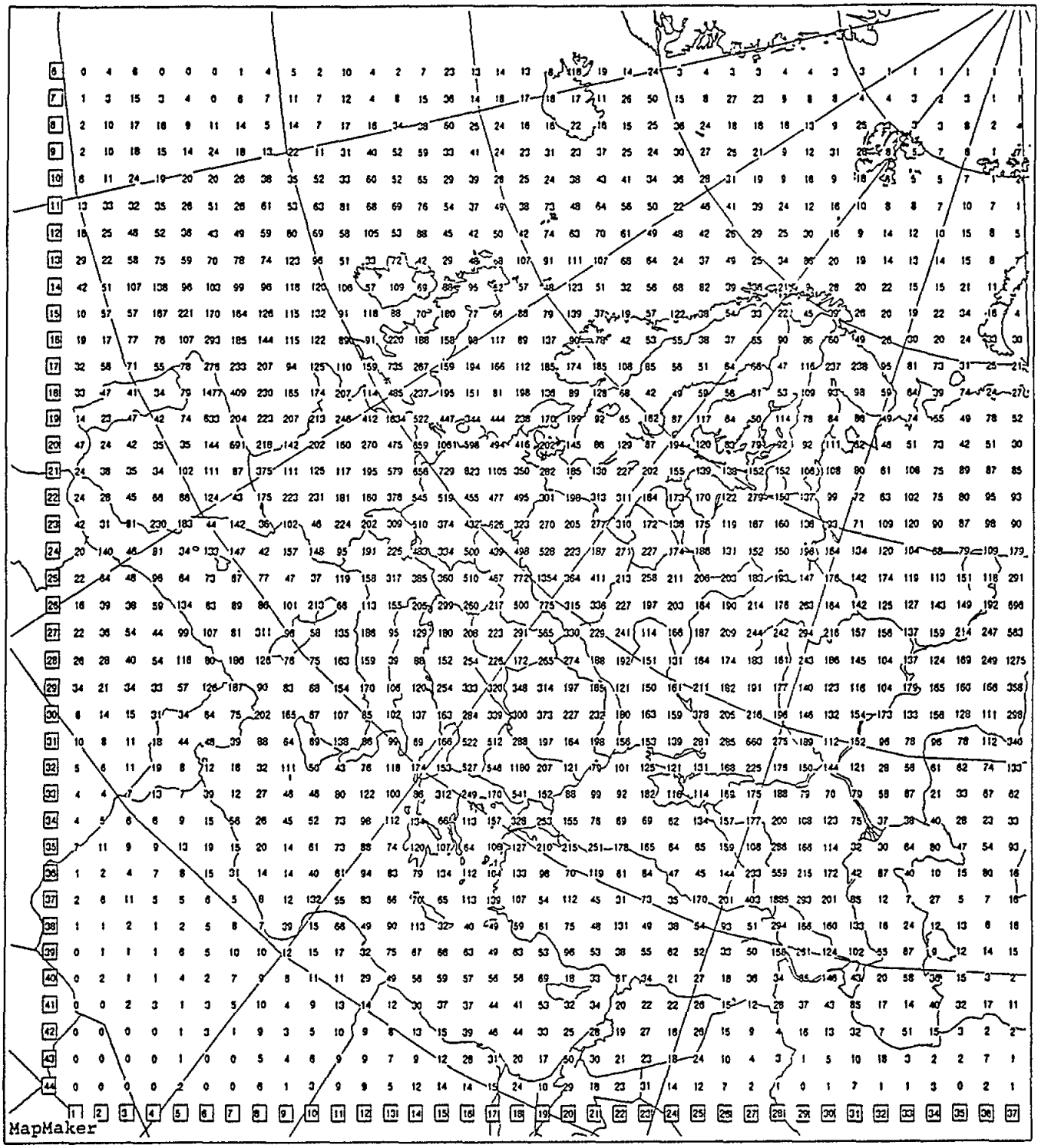


Fig.10.8. Total deposition of zinc in 1991 *(10 μg Zn/m^2/year).*

Table 10.3

Zinc deposition on the Mediterranean Sea, its subbasins and adjacent seas in 1991 from some countries-emitters (100 kg Zn per year)

	MT1	MT2	MT3	MT4	MT5	MT6	MT7	MT8	MT9	MT10	MAR	BLC	AZS	MDT	QA
AL	0	2	1	5	25	30	19	14	3	11	0	6	0	110	370
BG	0	67	24	58	135	271	380	1093	114	465	107	761	47	2607	17240
CS	0	26	12	27	47	22	30	21	5	11	6	76	4	201	7560
FR	24	378	362	260	79	81	153	12	5	11	1	57	3	1365	33115
GE	3	186	150	100	90	46	79	81	11	31	3	141	5	777	45380
GR	0	4	3	10	29	57	66	141	26	95	10	58	3	431	1755
HU	0	10	6	9	35	16	15	13	2	10	3	42	5	116	2030
IT	9	843	1348	1426	1047	941	1381	245	46	244	11	79	8	7530	19490
PL	0	41	23	51	200	85	86	117	64	32	14	399	16	699	40230
PT	12	11	22	6	1	3	3	0	0	0	0	0	0	58	1005
RO	0	15	14	13	25	58	60	153	19	74	33	470	31	431	6450
ES	311	1518	1899	767	187	162	290	36	17	23	1	12	1	5210	39825
TR	0	1	1	1	3	24	44	299	284	360	67	502	18	1017	6116
GB	2	33	51	30	5	14	21	15	4	1	1	64	1	176	22960
YU	0	87	51	175	454	343	337	536	58	176	39	481	41	2217	18045
RF	0	6	5	3	3	4	7	22	9	27	17	359	69	86	75664
UR	0	6	30	20	11	49	50	95	33	65	52	985	279	359	21507
MOR	34	4	10	2	0	0	0	0	0	0	0	0	0	50	272
ALG	51	45	186	33	10	12	23	3	0	1	0	0	0	364	2040
TUN	1	11	34	72	15	43	157	6	1	7	0	3	1	347	906
LIB	0	0	2	7	3	17	55	4	3	12	0	1	0	103	470
CYP	0	0	0	0	0	0	0	0	15	8	0	1	0	23	90
SYR	0	0	0	0	0	0	1	0	24	19	0	15	0	44	1360
LEB	0	0	0	0	0	0	0	0	12	15	0	2	0	27	350
ISR	0	0	0	0	0	0	0	0	3	47	0	0	0	50	620
EGP	0	0	0	0	0	1	3	7	13	108	1	4	0	132	1448
OC	3	69	67	60	54	39	48	103	104	150	33	2293	176	697	51886
Total	450	3363	4302	3135	2458	2318	3309	3016	875	2004	399	6812	708	25230	418184

Notations are given in tables 6.2 and 3.1

OC - other countries within calculation region

QA - emission of given country

airborne zinc that approximately corresponds to 2% of its total emission within the calculation region.

Thus on the whole in 1991 the area of regional seas: the Mediterranean, the Sea of Marmara, the Black Sea and the Sea of Azov received 3314 t of zinc that corresponds approximately to 8% of its emission within the calculation region.

In 1991 the adjacent countries received 6762 t of zinc that corresponds to 16% of its emission within the calculation region.

Note that total deposition value of 6762 t obtained for adjacent countries (about 16% of the emission within the calculated region) is an estimation of the deposition of zinc on the Mediterranean Sea watershed, where from a part of it with river flows is transported to the coastal region, the most sensitive to anthropogenic impact. Thus, due to the input to the marine environment of pollutants deposited on the sea watershed, the contribution of airborne pollution to the total pollution of the sea will be greater than the pollution resulted only from deposition directly on the sea.

10.2.4. Seasonal variations of zinc pollution.

Fluctuations of depositions and concentrations of Zn in the chosen points (see section 10.1.4) are shown in Figures 10.10 and 10.11. In the points P1 and P2 depositions in the winter and the spring seasons are higher than in summer and autumn. The distinguished minimum is obtained for P4 in summer. Seasonal fluctuations for the point P3 are slightly expressed. Generally speaking, variations of mean monthly depositions are considerable.

Mean monthly variations of zinc concentrations in air are considerably smaller. Difference by a factor of 2 between the maximum and the minimum concentrations is obtained for P1, by a factor of 2.5 - for P2 and P3, by a factor of 3 - for P4. Maps of seasonal depositions and concentrations in air for Zn are presented in Figures C47-C50, Appendix C.

10.3. Cadmium.

10.3.1. Deposition on the Mediterranean Sea.

In 1991 the deposition of cadmium on the Mediterranean Sea amounted to 73 tons. It is 6% of its total emission within the calculation grid. Annual deposition density of cadmium is about 30 μg Cd/m^2 year, it is about two times lower than mean density in Europe.

The deposition map of cadmium for 1991 is given in Figure 10.12, maps of dry depositions and surface concentrations of cadmium in air as well as in precipitations are given in Figures A13, A14, Appendix A.

Table 10.4 shows Cd depositions on the Mediterranean Sea from countries-emitters within the calculated grid (column MDT), its subbasins (column MT1-MT10) and adjacent seas (columns MAR, BLC, AZS) and Figure 10.2 gives per cent contributions of countries to total deposition of cadmium. Line OC (other countries) in Table 10.1 and similar one in Tables 10.3, 10.4 and 10.5 shows total deposition from countries within the calculated region, contributions of each is negligible.

It is seen from Table 10.4 and Figure 10.2 that Italy and Spain (18% and 21% of total deposition), Bulgaria and Yugoslavia (14% and 14% of total deposition respectively) make the main contribution to cadmium deposition on the Mediterranean Sea. Maps of deposition caused by these countries (within the calculation region) are given in Figures B19, B20, Appendix B.

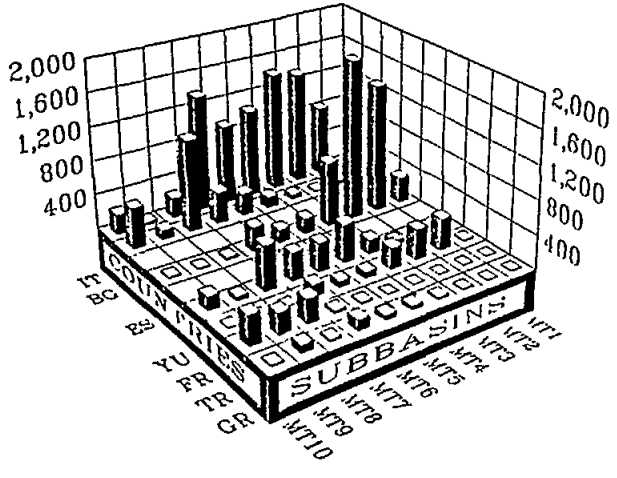


Fig.10.9 Zn deposition (from countries on subbasins)
UNIT=100 kg Zn per year

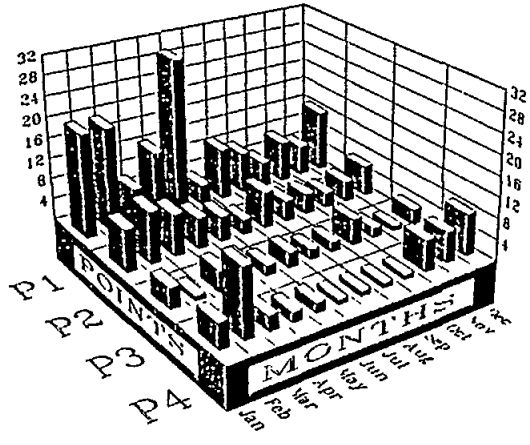


Fig.10.10 Zn monthly deposition in chosen points.
UNIT=10 ug/m2 P1-Barcelona,P2- Corsica,P3- Crete,P4-Tel Aviv

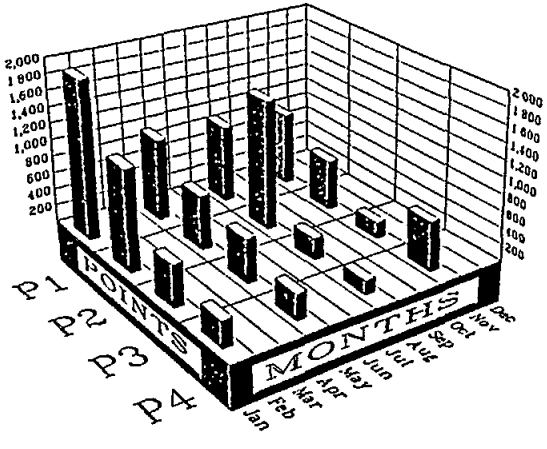


Fig.10.11 Zn monthly air concentrations in chosen points
UNIT=01 ng/m3 P1-Barcelona,P2- Corsica,P3- Crete,P4-Tel Aviv

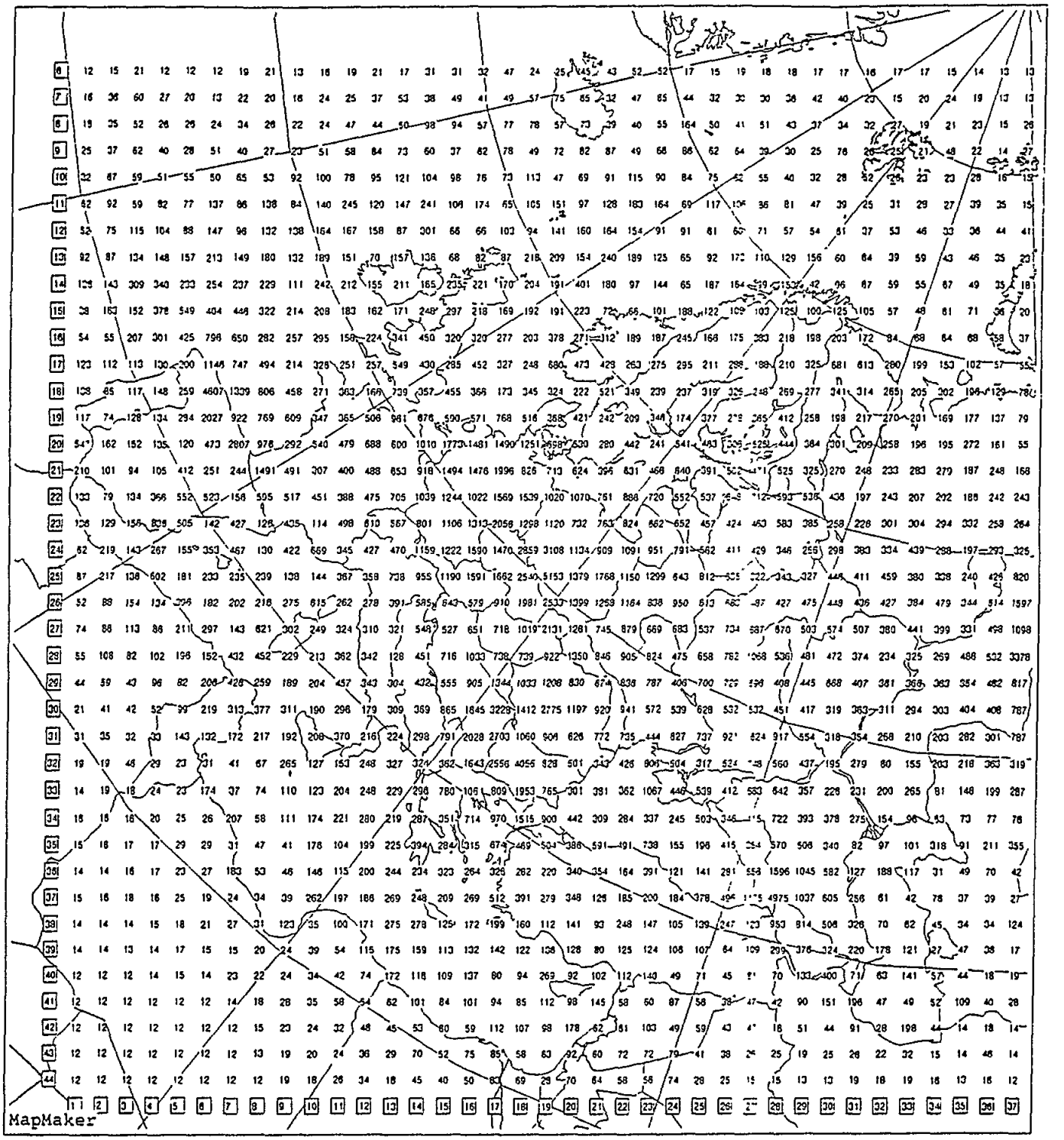


Fig.10.12. Total deposition of cadmium in 1991
(0.1 μg Cd/m²/year).

Table 10.4

Cadmium deposition on the Mediterranean Sea, its subbasins and adjacent seas in 1991 from some countries-emitters (10 kg Cd per year)

	MT1	MT2	MT3	MT4	MT5	MT6	MT7	MT8	MT9	MT10	MAR	BLC	AZS	MDT	QA
AL	0	0	0	0	3	3	1	1	0	1	0	0	0	9	110
BG	0	8	11	21	43	103	152	449	53	161	42	294	13	1043	6490
CS	0	7	4	7	16	9	8	7	3	4	2	21	1	67	2425
FR	1	117	68	91	29	22	30	3	1	4	0	11	0	366	3620
GE	2	50	36	40	41	12	17	9	4	8	1	36	2	220	12040
GR	0	0	0	0	0	5	9	31	4	17	0	2	0	66	360
HU	0	1	1	1	4	2	3	2	0	1	1	8	0	16	435
IT	1	153	217	272	189	183	225	40	9	38	1	15	1	1328	3585
PL	0	39	8	28	77	38	37	40	11	22	8	217	13	308	18045
PT	0	0	0	0	0	0	0	0	0	0	0	0	0	0	200
RO	0	11	3	7	14	32	34	84	12	44	13	297	23	254	4260
ES	130	499	575	182	41	28	69	6	0	4	1	7	0	1535	13485
TR	0	0	0	0	0	3	7	47	46	58	12	80	3	173	1232
GB	1	2	7	3	2	1	2	2	0	0	0	5	1	20	3075
YU	0	45	22	103	183	134	154	261	30	97	16	218	17	1045	8560
RF	0	1	1	0	1	2	3	5	1	4	2	54	14	20	18645
UR	0	2	5	4	3	5	9	20	8	15	9	227	70	80	5066
MOR	0	0	0	0	0	0	0	0	0	0	0	0	0	0	50
ALG	6	8	30	4	2	2	3	0	0	0	0	0	0	55	395
TUN	0	1	5	11	2	6	23	1	0	1	0	0	0	50	180
LIB	0	0	0	0	0	0	0	0	0	0	0	0	0	0	79
CYP	0	0	0	0	0	0	0	0	0	0	0	0	0	0	20
SYR	0	0	0	0	0	0	0	0	4	4	0	3	0	8	270
LEB	0	0	0	0	0	0	0	0	2	3	0	0	0	5	70
ISR	0	0	0	0	0	0	0	0	0	7	0	0	0	7	100
EGP	0	0	0	0	0	0	0	2	4	29	0	1	0	35	437
OC	19	63	73	58	28	63	135	64	60	139	9	609	53	721	13951
Total	160	1007	1066	832	688	653	921	1074	252	661	117	2105	211	7431	117185

Notations are given in tables 6.2 and 3.1

OC - other countries within calculation region

QA - emission of given country

The input of Mediterranean countries is 46,5 tons, i.e. 14% of the total emission of these countries and 64% of total deposition on the sea.

Thus the input of remote countries to the cadmium deposition is about 36%, including Bulgaria (14%), Poland (4%) and Germany (3%) and Romania (3%).

10.3.2. Deposition on the Mediterranean Sea subbassins.

Figure 10.3 gives distribution of heavy metals depositions over the Mediterranean Sea subbasins. As it is evident from Figure 10.3 the maximum depositions of cadmium are in the subbasins MT2, MT3 and MT8 and minimum - in the Alboran Sea.

From Figure 10.4 presenting deposition density distribution of heavy metals with subbasins and the map of cadmium deposition (Figure 10.12) it is clear that the greatest density of cadmium deposition is observed in the Adriatic and the Aegean Seas. This deposition density is about two times higher than the mean value for the Mediterranean Sea.

The minimum deposition density (1.5 times less than mean one) is observed in subbasin MT7 and MT10 and the Alboran Sea (MT1).

Thus the deposition density is decreasing from north to south that corresponds to emission distribution within the calculation region, to main atmospheric transport routes and to annual precipitation amount distribution within the Mediterranean basin.

Figure 10.13 shows deposition on the Mediterranean subbasins from countries making the main contribution to the sea pollution.

Figure 10.13, Table 10.4 and A13-A14 (Appendix A) indicate that the main countries-emitters have their own "impact-zones" for cadmium. For example, Spain sources affect western part of the Sea (subbasins MT1-MT3) and partly the Tyrrenian Sea (MT4), Italy - central part (subbasins MT4-MT7) and partly subbasins MT2 and MT3, and Bulgaria, Turkey and Yugoslavia - eastern part (MT8-MT10).

10.3.3. Cadmium deposition on adjacent countries and seas.

Cadmium and nitrogen deposition on adjacent seas is given in Table 10.4 (columns MAR, BLC, AZS) and on adjacent countries - in Table 10.2 containing other heavy metals too.

In 1991 in total the Marmara, the Black and the Azov Seas received 24 t of airborne cadmium that approximately corresponds to 2% of its total emission within the calculation region.

Thus on the whole in 1991 the area of regional seas: the Mediterranean, the Sea of Marmara, the Black Sea and the Sea of Azov received about 100 t of cadmium that corresponds approximately to 8% of its emission within the calculation region.

In 1991 the adjacent countries received about 190 t of cadmium that corresponds to 16% of its emission within the calculation region.

The total deposition value of 190 t obtained for adjacent countries (about 16% of the emission within the calculation region) is an estimation of the deposition of cadmium on the Mediterranean Sea watershed, where from a part of it with river flows is transported to the coastal region, most sensitive to anthropogenic impact. Thus, due to the input to the marine environment of pollutants deposited on the sea watershed, the contribution of airborne pollution to the total pollution of the sea will be greater than the pollution resulted only from deposition directly on the sea.

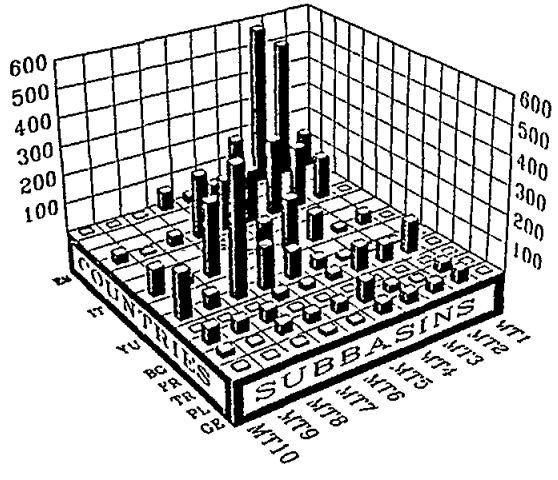


Fig.10.13 Cd deposition (from countries on subbasins)
UNIT=10 kg Cd per year

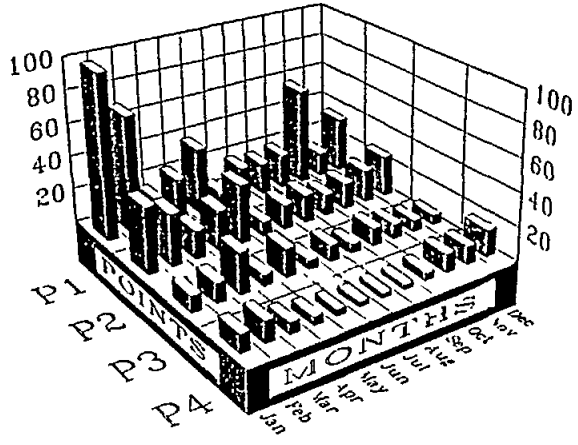


Fig 10.14 Cd monthly deposition in chosen points.
UNIT=0 1 ug/m2. P1-Barcelona,P2- Corsica,P3- Crete,P4-Tel Aviv

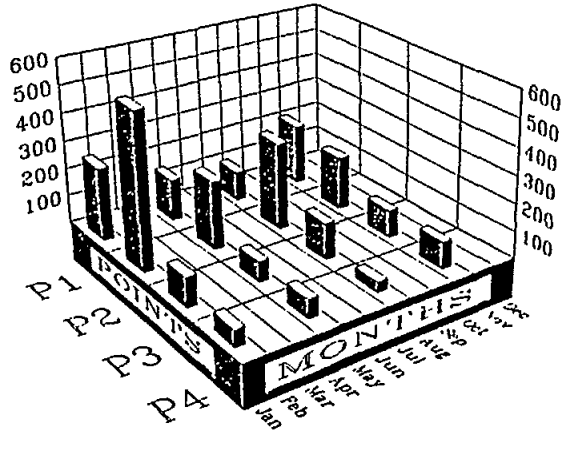


Fig 10.15 Cd monthly air concentrations in chosen points.
UNIT= 001 ng/m3 P1-Barcelona,P2- Corsica,P3- Crete,P4-Tel Aviv

10.3.4. Seasonal variations of cadmium pollution.

Seasonal data on depositions and concentrations of Cd in the air for the chosen points (see Chapter 10.1.4) are presented in Figures 10.14 and 10.15 respectively.

The distinct minimum of depositions for all four points is clearly seen in summer. Seasonal differences between the maximum and minimum depositions are considerable and make up not less than factor of 4 for all points.

Seasonal variations of Cd concentrations are insignificant. The ratios between maximum and minimum values are no more than 2.5 times for each point. Maps of seasonal depositions and concentrations of Cd are presented in Figures C51-C54, Appendix C.

10.4. Arsenic.

10.4.1. Deposition on the Mediterranean Sea.

In 1991 the deposition of arsenic on the Sea amounted to 200 tons. It is 4% of its total emission within the calculation grid. Annual deposition density of arsenic is about 80 μg As/m^2 year, it is about three times lower than mean density in Europe.

The map of arsenic deposition for 1991 is given in Figure 10.16, maps of dry depositions and surface concentrations of arsenic in air as well as concentrations in precipitations are given in Figures A15, A16, Appendix A.

Table 10.5 shows As depositions on the Mediterranean Sea from countries-emitters within the calculated grid (column MDT), its subbasins (column MT1-MT10) and adjacent seas (columns MAR, BLC, AZS) and Figure 10.2 gives per cent contributions of countries to total deposition of arsenic. Line OC (other countries) in Table 10.5 and similar one in Tables 10.3, 10.4 and 10.5 shows total deposition from countries within the calculation region, individual contributions of which are small.

It is seen from Table 10.5 and Figure 10.2 that Italy and Spain (19% and 18% of total deposition on the Mediterranean Sea), Bulgaria and Yugoslavia (15% and 12% of total deposition respectively) make the main contribution to arsenic deposition on the Mediterranean Sea. Maps of deposition caused by these countries (within the calculation region) are given in Figures B21, B22, Appendix B.

Note that the input of Mediterranean countries is 138 tons As, i.e. 15% of the total emission of these countries and 69% of total deposition on the sea.

Thus the input of remote countries to the arsenic deposition is about 31%, including Bulgaria (12%), Poland (5%), Germany and Romania (each 3%).

10.4.2. Deposition on the Mediterranean Sea subbassins.

Figure 10.3 gives distribution of heavy metals depositions over the Mediterranean Sea subbasins. It is evident from Figure 10.3 that the maximum deposition of arsenic is in the Aegean Sea (MT8) and subbasins MT3, MT7 and minimum - in subbasins MT1 and MT9.

From Figure 10.4 presenting the density distribution of arsenic depositions over subbasins and the map of deposition (Figure 10.16) it is clear that the maximum density of arsenic deposition is observed in the Adriatic (MT5) and the Aegean (MT8) Seas. This deposition density is about two times higher than the mean value for the Mediterranean Sea.

The minimum deposition density (1.5 times less than mean one) is observed in subbasins MT7 and MT10.

Thus the deposition density is decreasing mainly from north to south that corresponds to emission distribution within the calculation region, to main atmospheric transport routes and to annual precipitation amount distribution within the Mediterranean

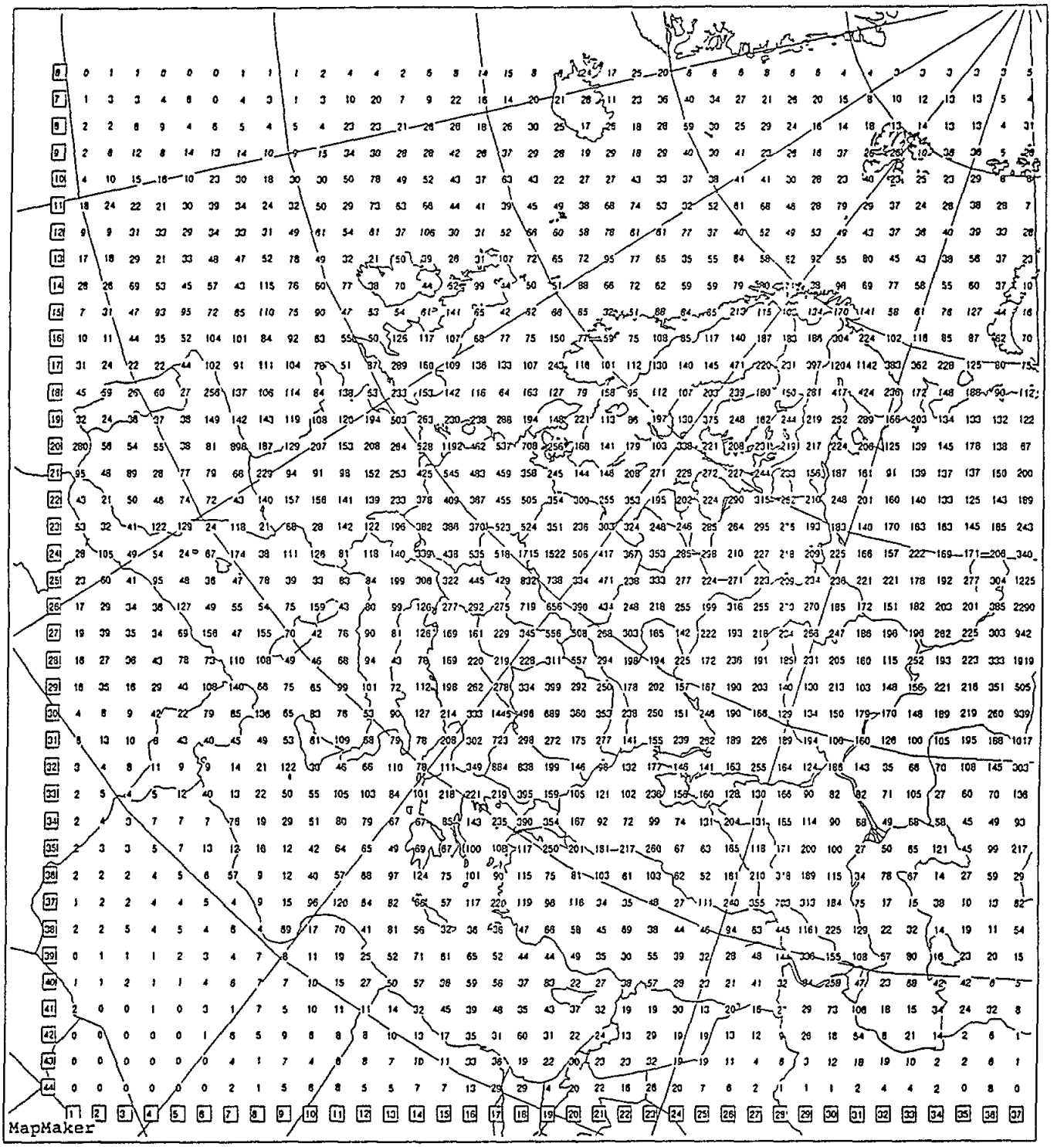


Fig.10.16. Total deposition of arsenic in 1991
(1 μg As/m²/year).

Table 10.5

Arsenic deposition on the Mediterranean Sea, its subbasins and adjacent seas in 1991 from some countries-emitters (100 kg As per year)

	MT1	MT2	MT3	MT4	MT5	MT6	MT7	MT8	MT9	MT10	MAR	BLC	AZS	MDT	QA
AL	0	1	1	2	12	9	7	5	1	4	0	3	0	42	165
BG	0	5	4	5	13	23	30	103	12	41	9	63	5	236	1500
CS	0	3	2	3	5	3	3	3	1	1	1	10	1	24	940
FR	1	29	24	19	6	6	9	1	0	1	0	3	0	96	1470
GE	1	14	8	9	10	3	5	6	1	3	1	9	0	60	3530
GR	0	0	0	0	1	6	8	17	4	17	0	2	0	53	140
HU	0	1	0	1	2	1	1	2	0	1	0	4	0	9	185
IT	1	39	61	74	44	57	73	13	3	12	0	4	0	377	965
PL	0	13	6	13	15	13	13	20	6	5	3	47	4	104	5960
PT	1	1	2	1	0	0	0	0	0	0	0	0	0	5	115
RO	0	4	1	2	4	9	9	23	5	12	5	73	5	69	1145
ES	48	97	129	44	14	11	18	2	0	1	0	1	0	364	2680
TR	0	0	0	0	0	2	3	19	18	23	4	32	1	65	396
GB	0	1	3	2	0	1	2	0	0	0	0	3	0	9	1195
YU	0	11	8	19	49	44	52	76	9	30	6	93	5	298	2715
RF	0	1	0	0	1	1	1	3	1	3	1	50	12	11	17843
UR	0	1	1	1	1	5	5	8	1	5	3	88	25	28	1308
MOR	3	0	1	0	0	0	0	0	0	0	0	0	0	4	20
ALG	4	4	14	3	1	1	2	0	0	0	0	0	0	29	157
TUN	0	1	2	5	1	4	12	1	0	1	0	0	0	27	71
LIB	0	0	0	1	0	1	4	0	0	1	0	0	0	7	39
CYP	0	0	0	0	0	0	0	0	1	0	0	0	0	1	7
SYR	0	0	0	0	0	0	0	0	2	2	0	1	0	4	113
LEB	0	0	0	0	0	0	0	0	1	1	0	0	0	2	30
ISR	0	0	0	0	0	0	0	0	0	3	0	0	0	3	40
EGP	0	0	0	0	0	0	0	1	2	12	0	0	0	15	175
OC	0	6	4	7	5	3	4	5	9	11	1	179	10	54	7640
Total	59	233	272	212	185	204	264	309	78	192	34	667	68	2008	50544

Notations are given in tables 6.2 and 3.1

OC - other countries within calculation region

QA - emission of given country

basin.

Figure 10.17 shows depositions on the Mediterranean subbasins from countries making the main contribution to the Sea pollution.

Figure 10.17, Table 10.5 and maps B21 and B22 indicate "impact-zones" of the main countries emitters. Spanish sources affect western part of the Sea (subbasins MT1-MT3) including the Tyrrhenian Sea (MT4), Italy - central part (subbasins MT4-MT7) complemented by subbasins MT2 and MT3 (mainly their western parts), Yugoslavia - subbasins MT5-MT8 and MT10, Bulgaria and Turkey - eastern part. Pollution from Yugoslavia (the biggest As emitter among the Mediterranean countries) spreads over central and eastern part of the Sea, giving not less than 10 per cent of deposition on each of these parts.

10.4.3. Arsenic deposition on adjacent countries and seas.

Arsenic deposition on adjacent seas is given in Table 10.5 (columns MAR, BLC, AZS) and on adjacent countries - in Table 10.2 containing other heavy metals too.

In 1991 in total the Marmara, the Black and the Azov Seas received 77 t of airborne arsenic that approximately corresponds to 1.5% of its total emission within the calculation region.

Thus on the whole in 1991 the area of regional seas: the Mediterranean, the Sea of Marmara, the Black Sea and the Sea of Azov received about 280 t of arsenic that corresponds approximately to 5.5% emission within the calculation region.

In 1991 the adjacent countries received about 520 t of arsenic that corresponds to 10% of its emission within the calculation region.

The total deposition value of 520 t obtained for adjacent countries (about 10% of the emission within the calculated region) is an estimation of the deposition of arsenic on the Mediterranean Sea watershed, where from a part of it with river flows is transported to the coastal region, most sensitive to anthropogenic impact. Thus, due to the input to the marine environment of pollutants deposited on the sea watershed, the contribution of airborne pollution to the total pollution of the sea will be greater than the pollution resulted only from deposition directly on the sea.

10.4.4. Seasonal variations of arsenic pollution.

Figures 10.18 and 10.19 present seasonal depositions and concentrations of As in air in the chosen points (see Section 10.1.4). The obvious summer minimum of monthly depositions is shown in Figure 10.18 for all points, except for Crete, that can be explained by the corresponding minimum of precipitation. Variations of the calculated values are great, the ratios between the maximum and the minimum values are not less than 5 times for all points. Fluctuations of the mean monthly concentrations are expressed more smoothly - the same ratios vary within factors 2-4.

The maps of seasonal depositions and concentrations of As in air are presented in Figures C55-C58, Appendix C.

References:

1. WMO/UNEP: Airborne pollution of the Mediterranean Sea. Report and proceedings of a WMO/UNEP Workshop. MAP Technical Reports Series No.31, UNEP, Athens, 1989.

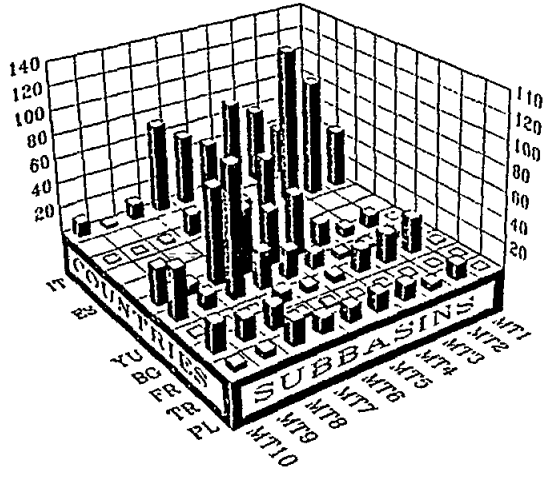


Fig.10.17 As deposition (from countries on subbasins)
UNIT=100 kg As per year

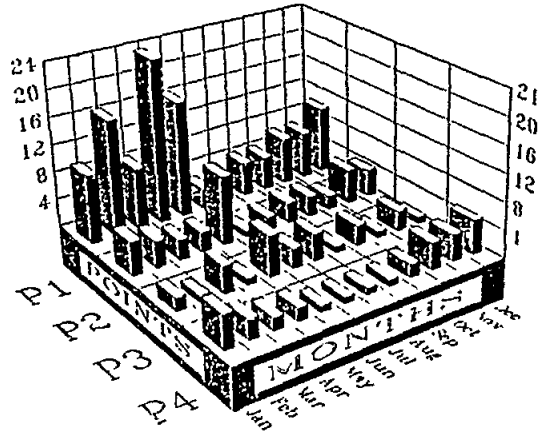


Fig 10.18 As monthly deposition in chosen points
UNIT=1 ug/m2. P1-Barcelona,P2- Corsica,P3- Crete,P4-Tel Aviv

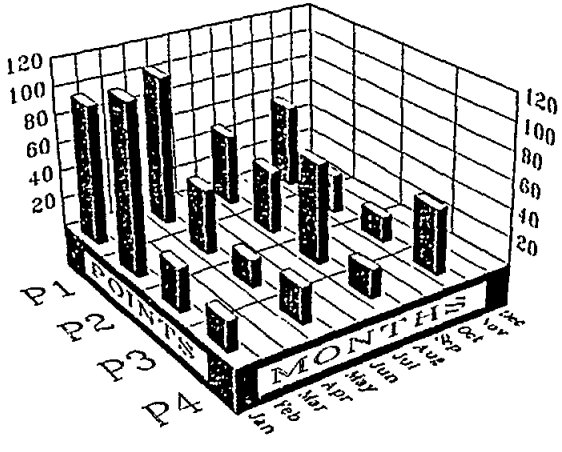


Fig 10.19 As monthly air concentrations in chosen points.
UNIT=0.01 ng/m3 P1-Barcelona,P2- Corsica,P3- Crete,P4-Tel Aviv

XI. COMPARISON OF CALCULATIONS AND MEASUREMENT DATA

11.1. Measurement data

Regular measurements of sulphur and nitrogen compounds are carried out at the EMEP network and data are available.

The comparison was made for the following components:

-SO_x: SO_2 concentrations in air, $SO_4^=$ concentrations in aerosol and precipitations;

-NO_x: NO_2 concentrations in air and in aerosol; $NO_3^- + HNO_3$ in aerosol and in precipitations;

-NH_x: $NH_3 + NH_4^+$ concentrations in aerosol and precipitations.

Since measurement data are of regular and long-term character, monthly average data up to 1991 were used for the comparison.

At the same time, sparse stations along the Mediterranean coastal line limit possibilities of the model calibration for this region. Only about 5 EMEP stations are located in the Mediterranean region. For comparison, in the Baltic region there are about 25 stations and about half of them belongs to the EMEP network. For that reason the following data were taken into consideration:

1. For general verification of the model data from all stations of the EMEP network with a completeness of daily measurements no less than 75% each month were taken (after eliminating the months with data completeness less than 6 months in a year).

2. For the evaluation of accuracy of the calculated results in the Mediterranean region, five-year data sets were taken for stations I4 (Ispra) in Northern Italy and YU2 in former Yugoslavia These two stations provided the most complete sets of measurements in the Mediterranean region.

Measurement data on heavy metals including ship measurements are rather scarce. The available data include mainly the measurement data from the regions of the North and Baltic Seas and data from measurement campaigns conducted in the Mediterranean in the late 1980s ([3] - [7]). The only data available for 1991 are the data on Pb and Zn concentrations in precipitation from three stations in the former Yugoslavia [10]. The following data were used for the comparison with the calculations:

- concentrations of heavy metals in air and precipitation (mean annual and available episodic measurements);
- total annual wet depositions of heavy metals.

11.2. Calculated data

For sulphur and nitrogen compounds calculations covering the period of 6 years (1987-92) were carried out by the MSC-E current model using a special grid (see Figure 3.1).

For heavy metals Pb, As, Zn, Cd provisional calculations were made by the MSC-E model described in Chapter VII using meteorological data for 1991. Only the calculated mean annual concentration data and total annual wet deposition data were used for the comparison with the measurement data.

The emission data base used is described in Chapter VI.

11.3. The comparison procedure

For the model verification a special procedure of the regression analysis and corresponding software allowing for basic statistical features of the model verification problem was developed at MSC-E [1].

Regression comparison was made for annual calculated and measured data. For sulphur and nitrogen compounds, a correspondence between a year of measurement and calculation was strictly kept. Calculations for heavy metals (total annual wet deposition and mean annual concentrations in air and precipitation) made for the 1991 meteorology were compared with measurement data of those years or periods for which they were available.

In some cases of the comparison the non-uniformly of measurement data base greatly distorted statistical properties of the regression point field.

For example, in case of the lead air concentration (Figure 11.12) the distortion was so significant that special procedure appeared to be unacceptable and the regression line was constructed by the standard least square method. In the rest of cases the violation of regression field properties was less important and CPEM (Choice Procedure of Effective Method - [1]) was used.

With regard to sulphur and nitrogen compounds, in addition to the regression procedure, time series of monthly concentrations for 6 years (1987-92) were used. A similar approach should be used for the development of air pollution control strategies since the multiannual variations of pollution fields strongly depend on multiannual variations of meteorological parameters [2].

11.4. The comparison results

The results of comparison between model results and measured monthly precipitation amounts are presented in Figure 11.1. The difference between the model results and observed precipitations is in a good agreement with current knowledge about precipitation field structure. Since the correlation distance for monthly averaged amount of precipitation is about 50-100 km the correlation between mean values in grid cell and point precipitation amount is about 0.5-0.7 [8]. At the same time the use for modelling of the climatological data (in absence of real meteorological information) results in the bias in regression and overestimation of mean values.

The results of comparison between the calculated and measured data for five years are presented as follows:

- for sulphur compounds - in Figure 11.2,
- for oxidized nitrogen compounds - in Figures 11.3, 11.4,
- for ammonia - in Figure 11.5.

Every point in the graph presents the annual calculated and measured values for each station.

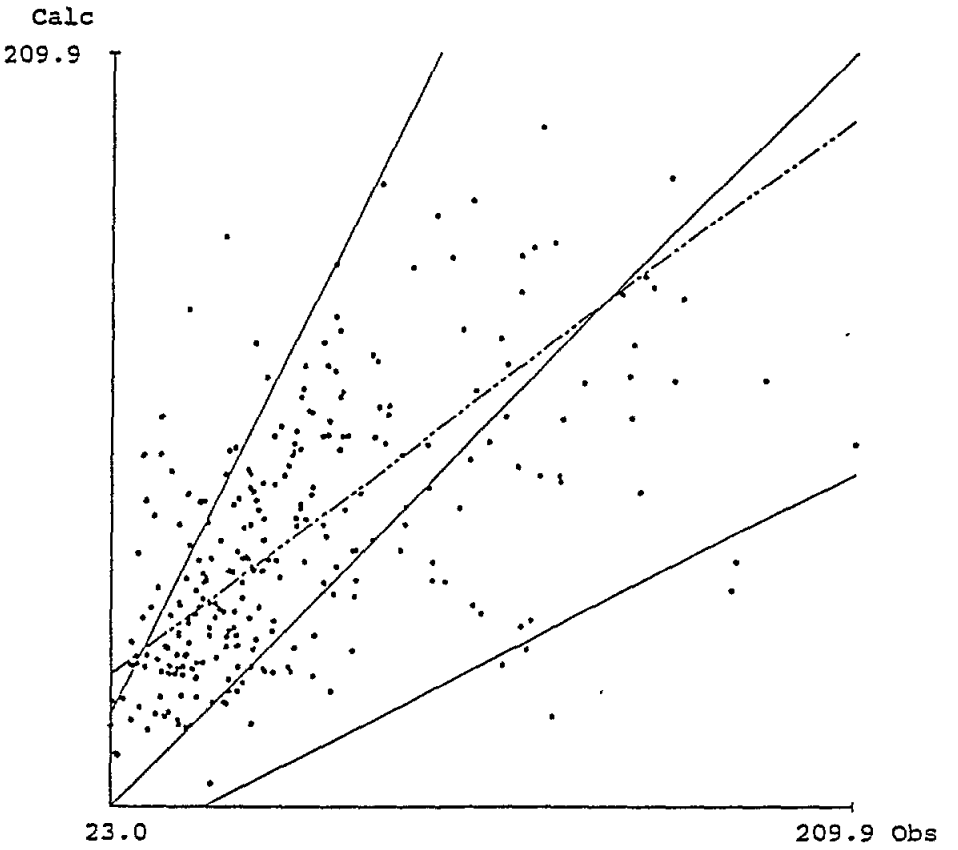
The six-year calculated data sets for the same compounds are presented for two EMEP stations: for I4 (Ispra) in Figures 11.6-11.9 and for YU2 (Putnijarka) in Figures 11.10, 11.11. Measurement data for these stations are available up to 1991.

On the whole the presented graphs show a satisfactory agreement between the calculated and measured data for both sulphur and nitrogen compounds over continental Europe.

In the region of the Mediterranean Sea (stations I4 and YU2) the agreement for sulphur remains satisfactory, but the calculation results for some nitrogen compounds are underestimated. This might be explained by underestimation of nitrogen emissions and a possible re-emission of N from the surface.

The results of comparison between the calculated and measurement data on annual wet depositions and mean concentrations in air and precipitation for Pb, Cd and Zn are given in Figures 11.12-11.14 and on mean concentrations in air and precipitation for As - in Figure 11.15.

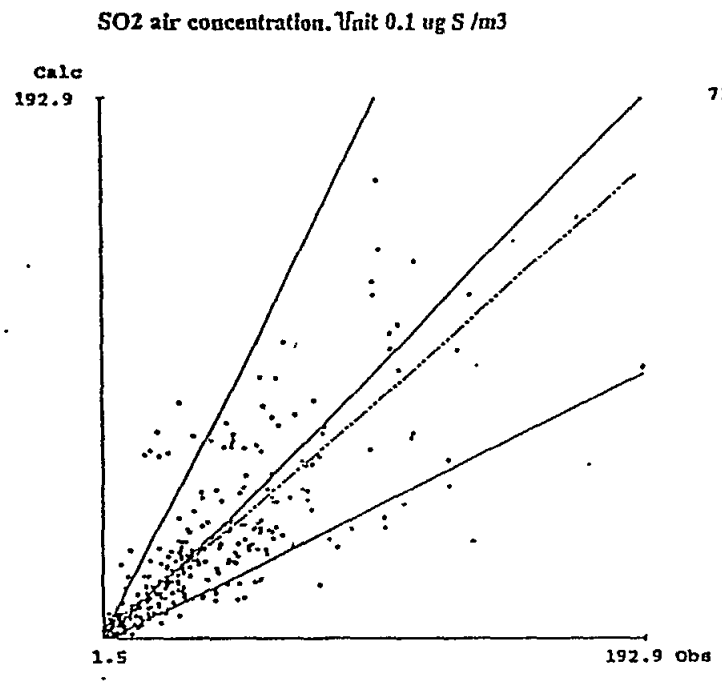
Mean monthly precipitation, mm

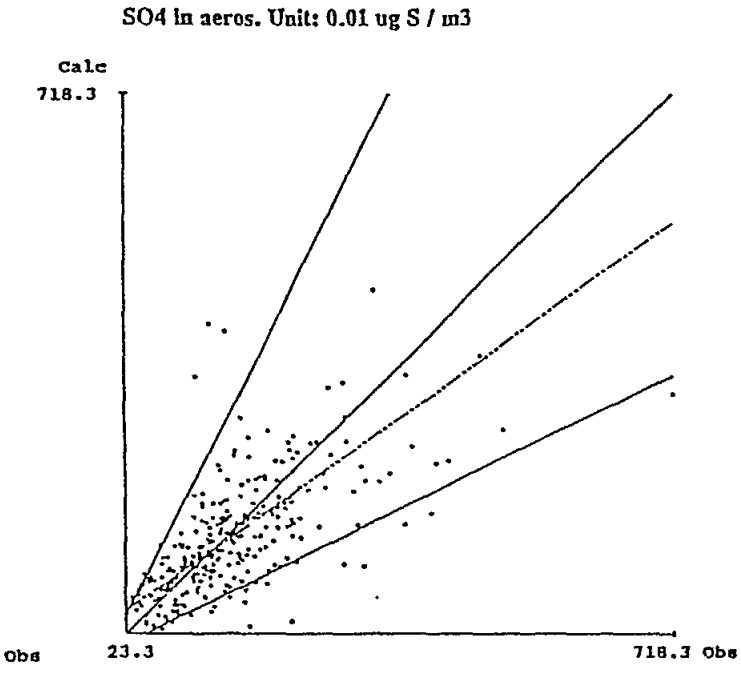


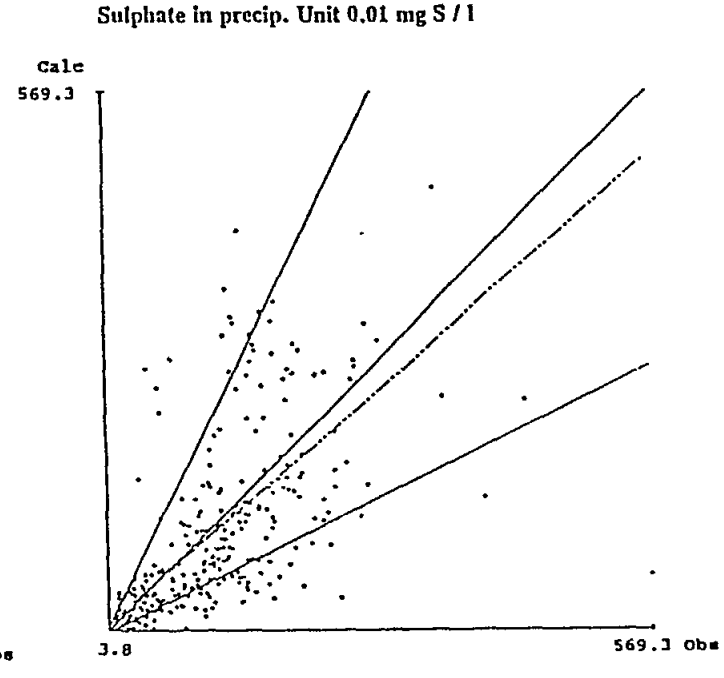
Regression coefficients:

Methods	A	B
CPEM	0.733	39.164
Number of points	266	
Mean Calculated	89.438	
Mean Observed	70.534	
Correlation	0.575	

Fig. 11.1. Comparison of mean monthly precipitation observed on EMEP stations and used in modelling







Regression coefficients:

Methods	A	B
CPEH	0.846	3.228
Number of points	299	
Mean Calculated	33.232	
Mean Observed	35.941	
Correlation	0.762	

Regression coefficients:

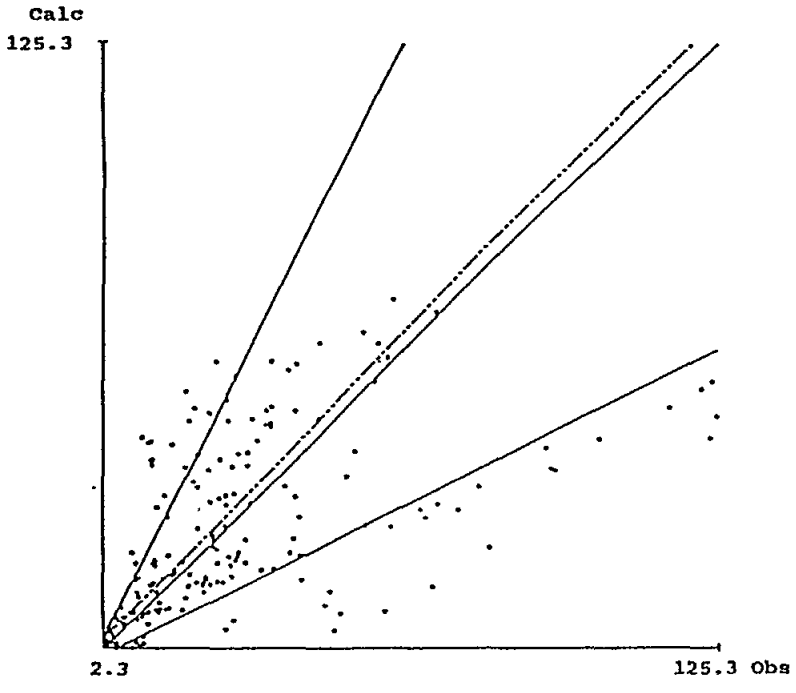
Methods	A	B
CPEH	0.717	37.028
Number of points	275	
Mean Calculated	148.820	
Mean Observed	159.279	
Correlation	0.647	

Regression coefficients:

Methods	A	B
CPEH	0.861	8.483
Number of points	257	
Mean Calculated	114.504	
Mean Observed	125.076	
Correlation	0.543	

Fig. 11.2. Comparison of sulphur compound annual concentrations observed on EMEP stations and calculated by MSC-E model for 1987-1991

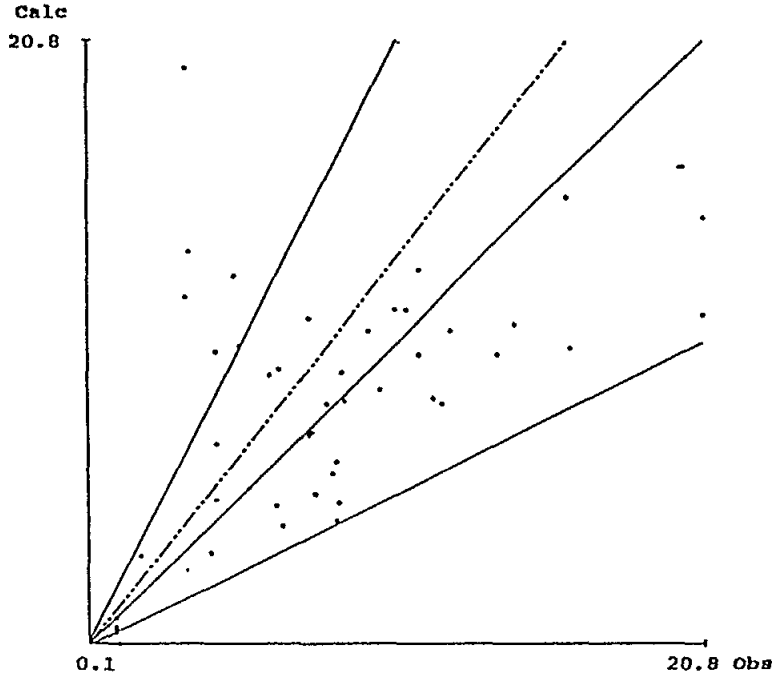
NO2 air concentration.Unit 0.1 ug N / m3



Regression coefficients:

Methods	A	B
CPEH	1.023	2.029
Number of points	175	
Mean Calculated	26.347	
Mean Observed	29.265	
Correlation	0.535	

NO3- in aerosol. Unit: 0.1 ug N / m3

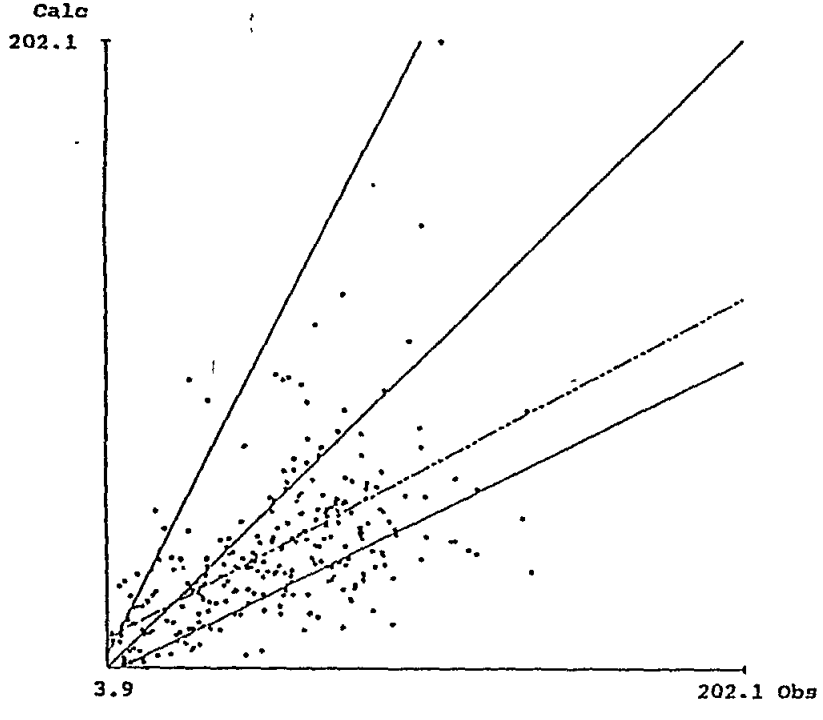


Regression coefficients:

Methods	A	B
CPEH	1.272	0.006
Number of points	49	
Mean Calculated	8.618	
Mean Observed	8.300	
Correlation	0.578	

Fig. 11.3. Comparison of NO_2 and nitrate annual concentrations in air observed on EMEP stations and calculated by MSC-E model for 1987-1991

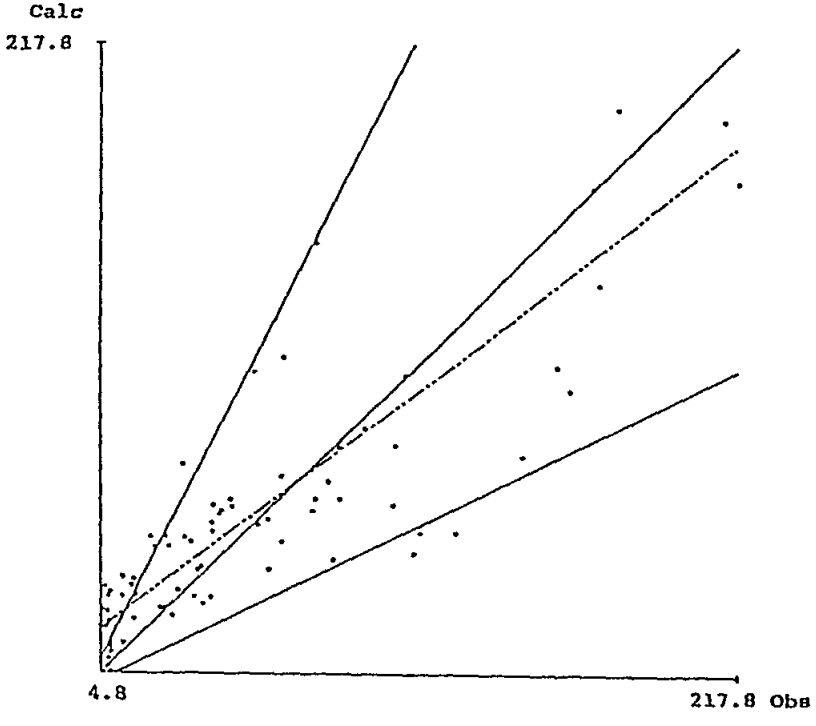
Nitrate in precip. Unit 0.01 mg N / l



Regression coefficients:

Methods	A	B
CPEM	0.543	11.018
Number of points	260	
Mean Calculated	40.955	
Mean Observed	56.236	
Correlation	0.535	

NO3-+HNO3 in air. Unit 0.01 ug N / m3

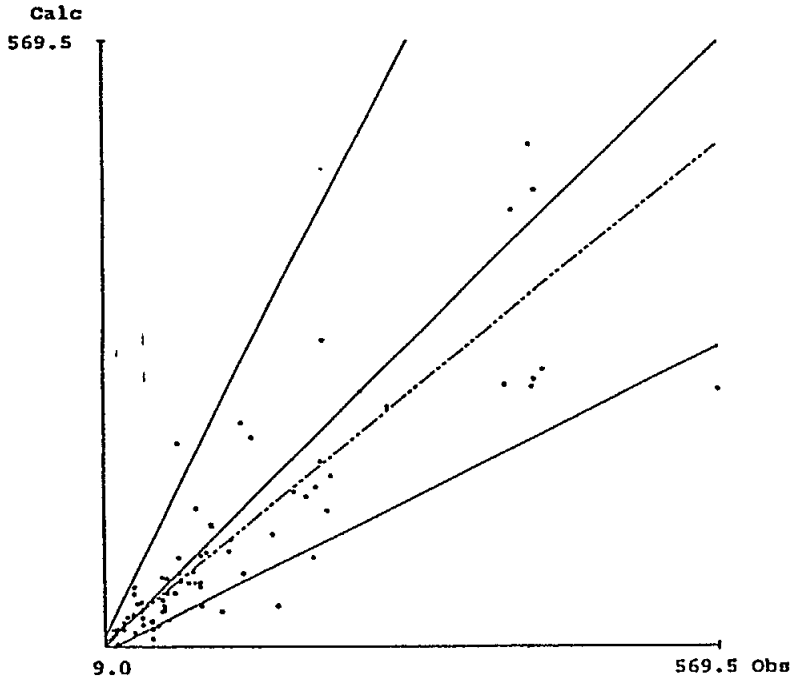


Regression coefficients:

Methods	A	B
CPEM	0.766	16.229
Number of points	74	
Mean Calculated	57.451	
Mean Observed	56.080	
Correlation	0.858	

Fig. 11.4. Comparison of oxidised nitrogen compound annual concentrations observed on EMEP stations and calculated by MSC-E model for 1987-1991

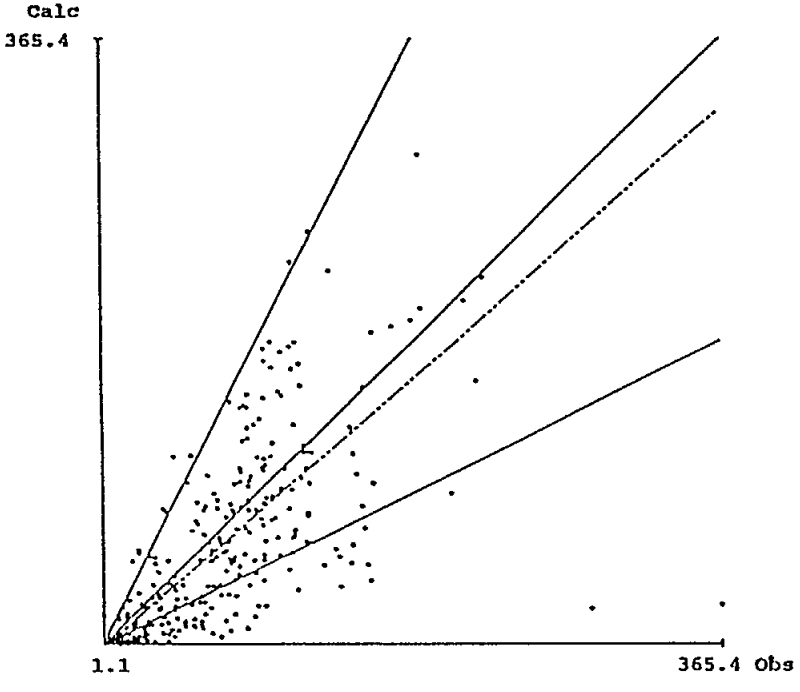
NH3+NH4- in air. Unit 0.01 ug N / m3



Regression coefficients:

Methods	A	B
CPEM	0.818	5.828
Number of points	74	
Mean Calculated	107.076	
Mean Observed	127.111	
Correlation	0.861	

Ammonia in precip. Unit 0.01 mg N / l



Regression coefficients:

Methods	A	B
CPEM	0.882	-1.728
Number of points	270	
Mean Calculated	63.248	
Mean Observed	74.324	
Correlation	0.610	

Fig. 11.5. Comparison of reduced nitrogen compound annual concentrations observed on EMEP stations and calculated by MSC-E model for 1987-1991

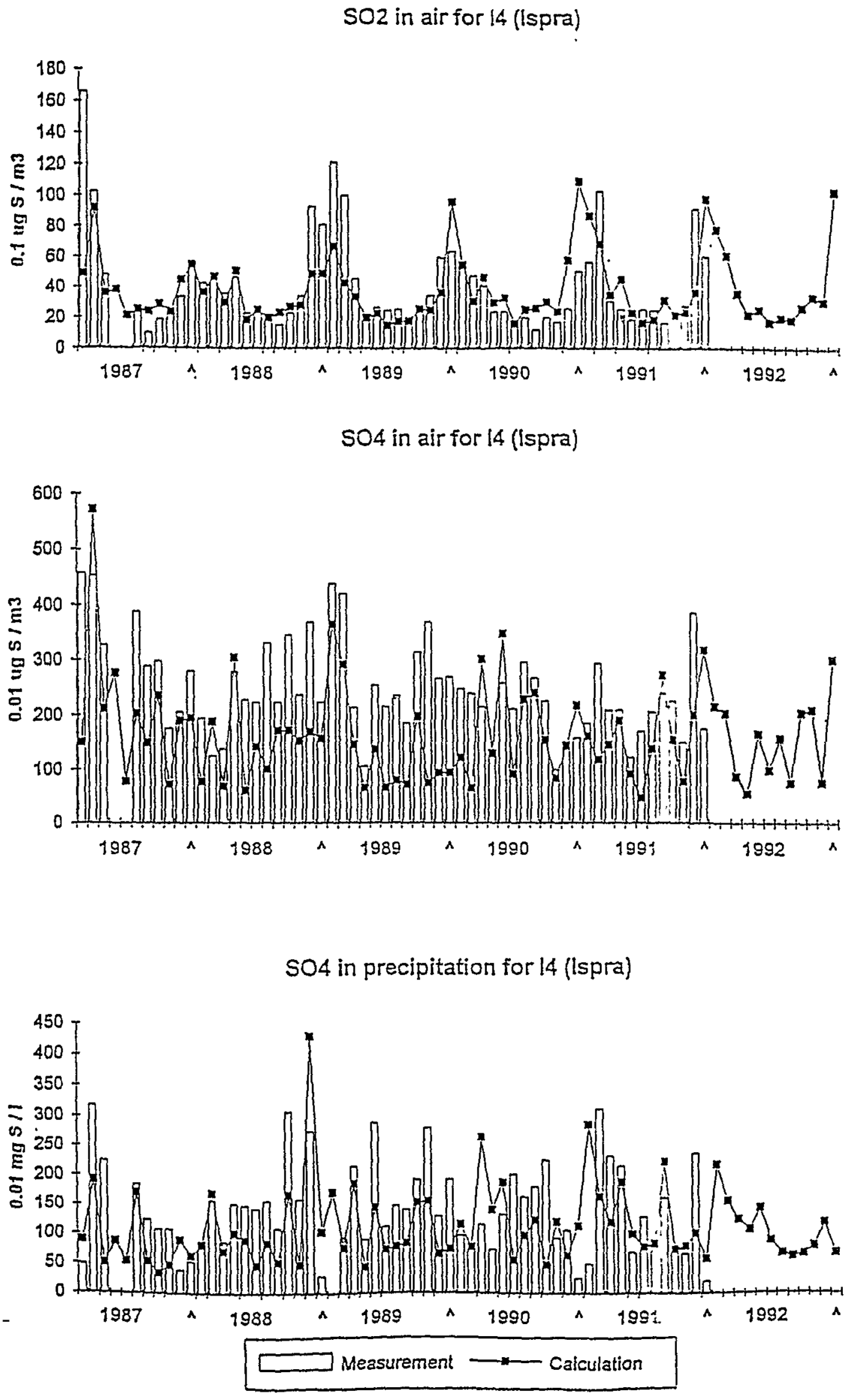


Fig. 11.6. *Comparison of model and measurement S compound concentrations for EMEP station I4 in North Italy*

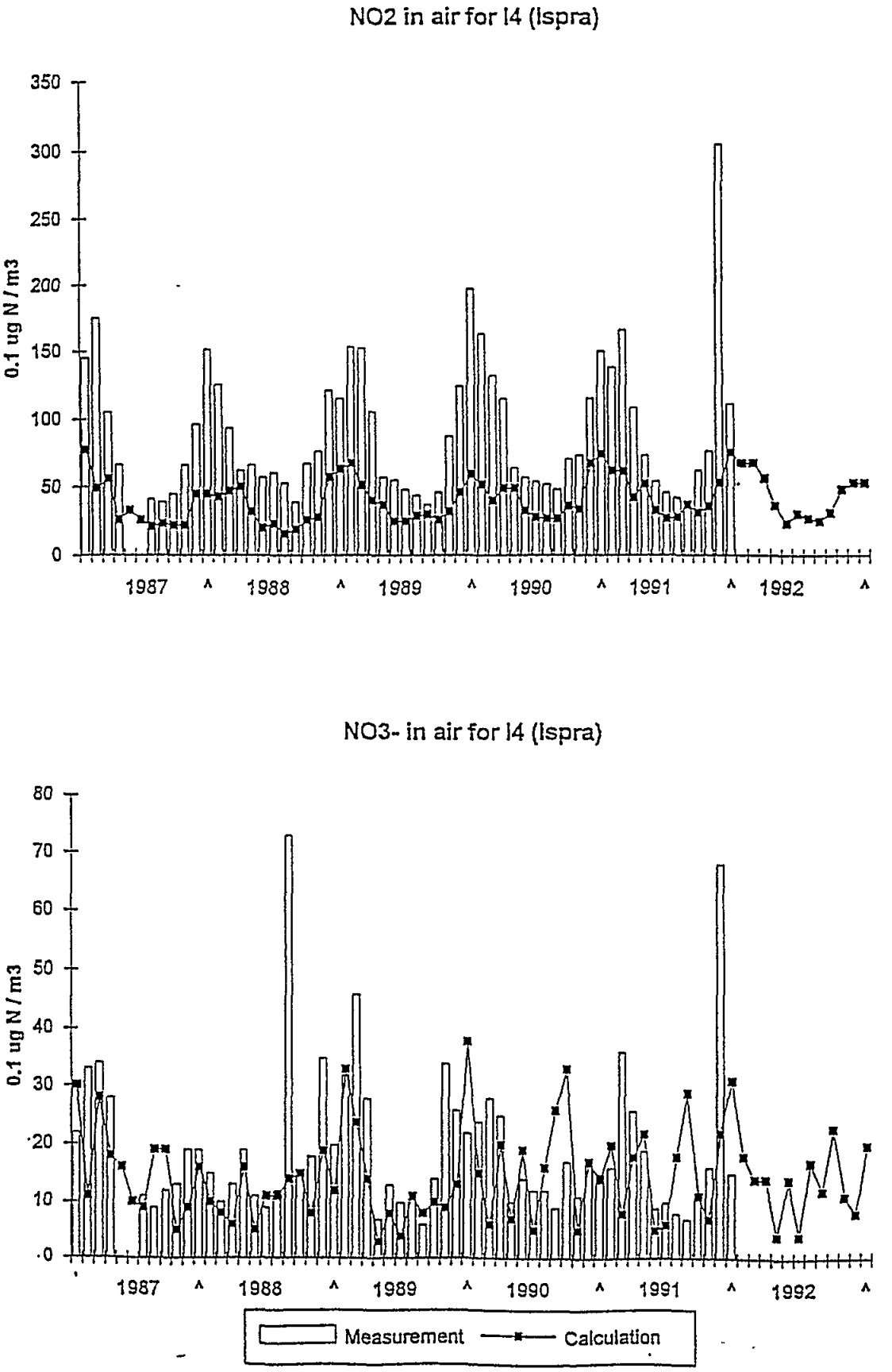


Fig. 11.7. Comparison of model and measurement NO_2 and NO_3^- concentrations in air for EMEP station I4 in North Italy

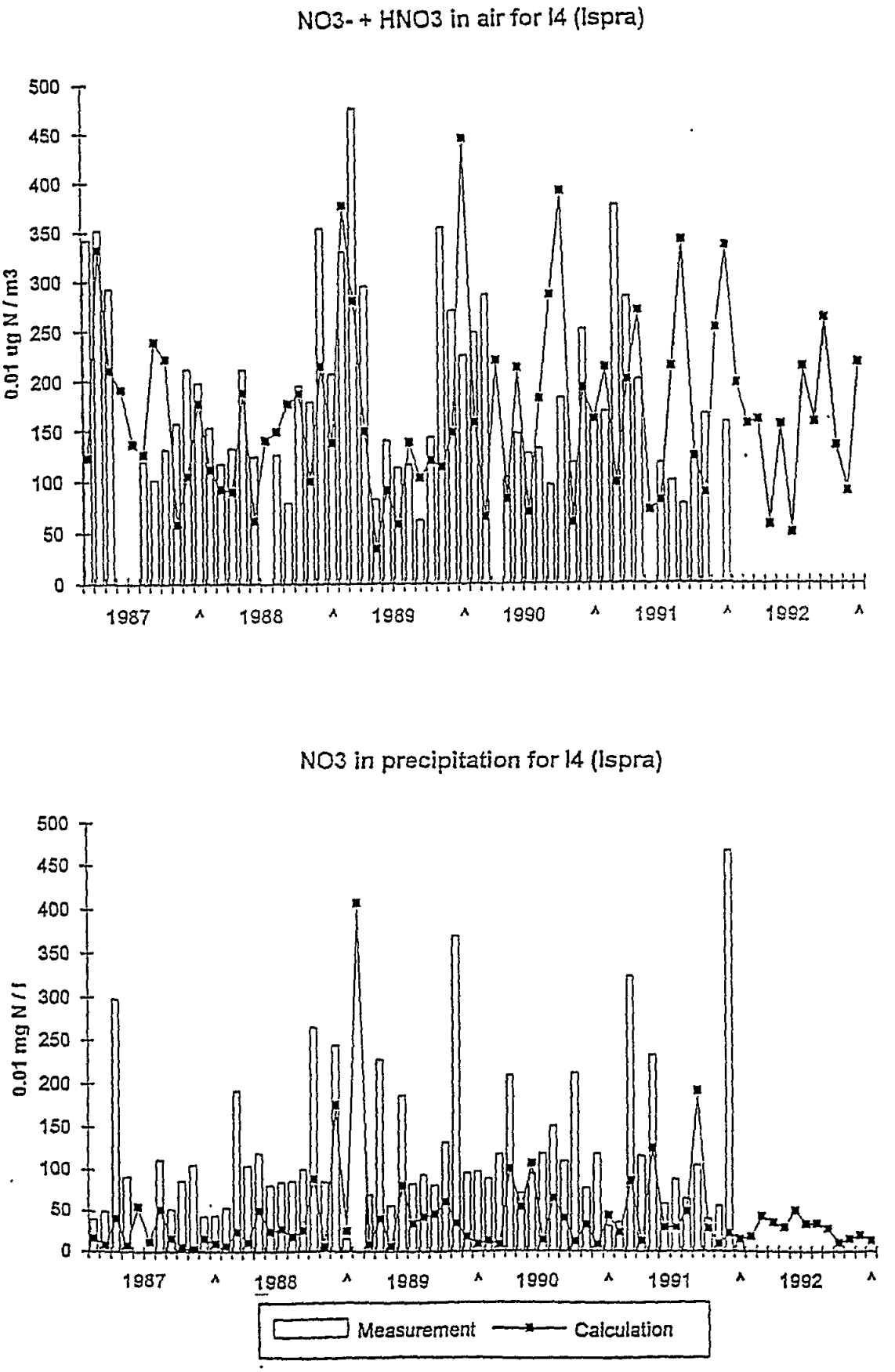


Fig. 11.8. Comparison of model and measurement NO_3^- + HNO_3 concentration in air and NO_3 in precipitation for EMEP station I4 in North Italy

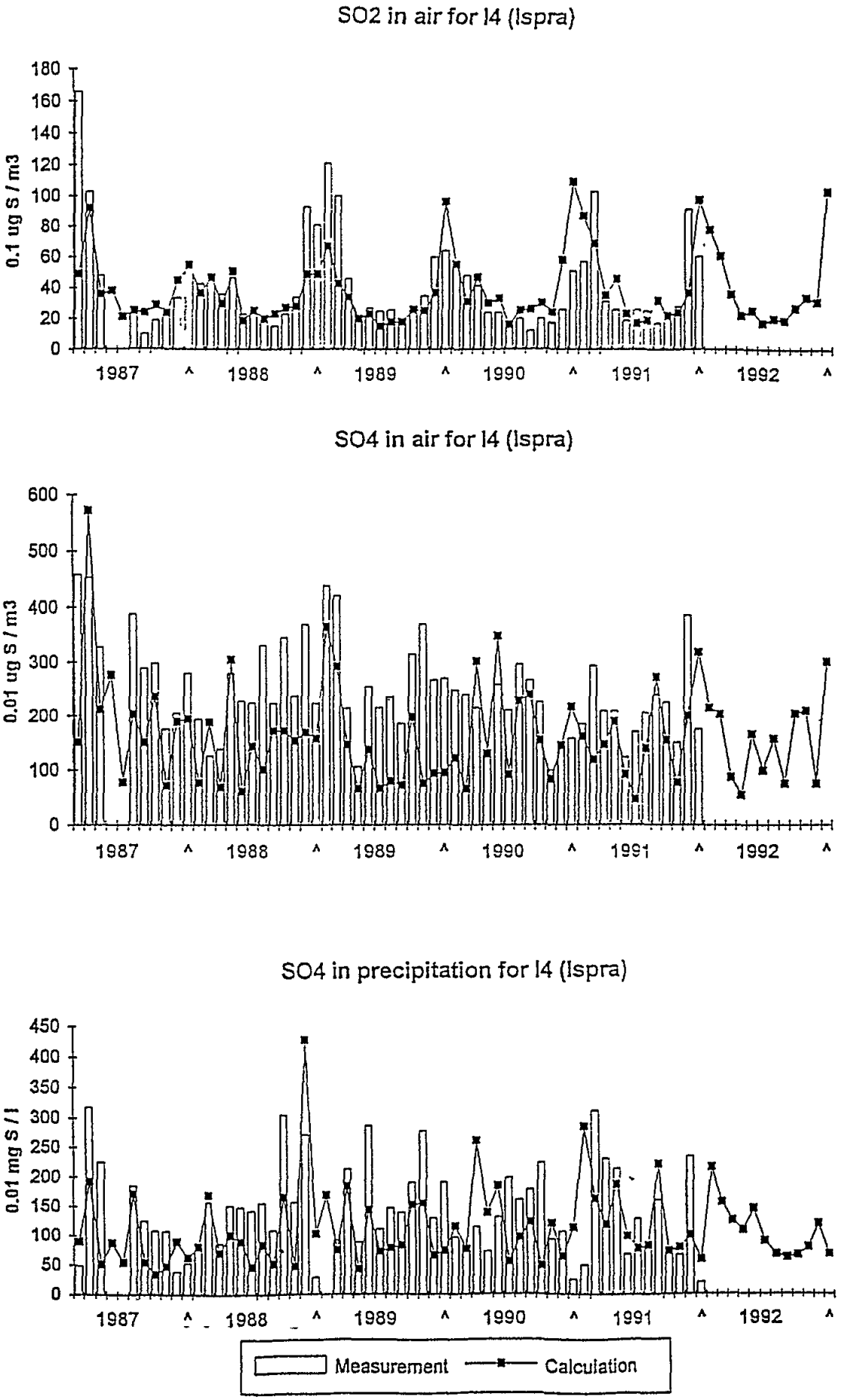


Fig. 11.9. Comparison of model and measurement data for EMEP *station I4 in North Italy*

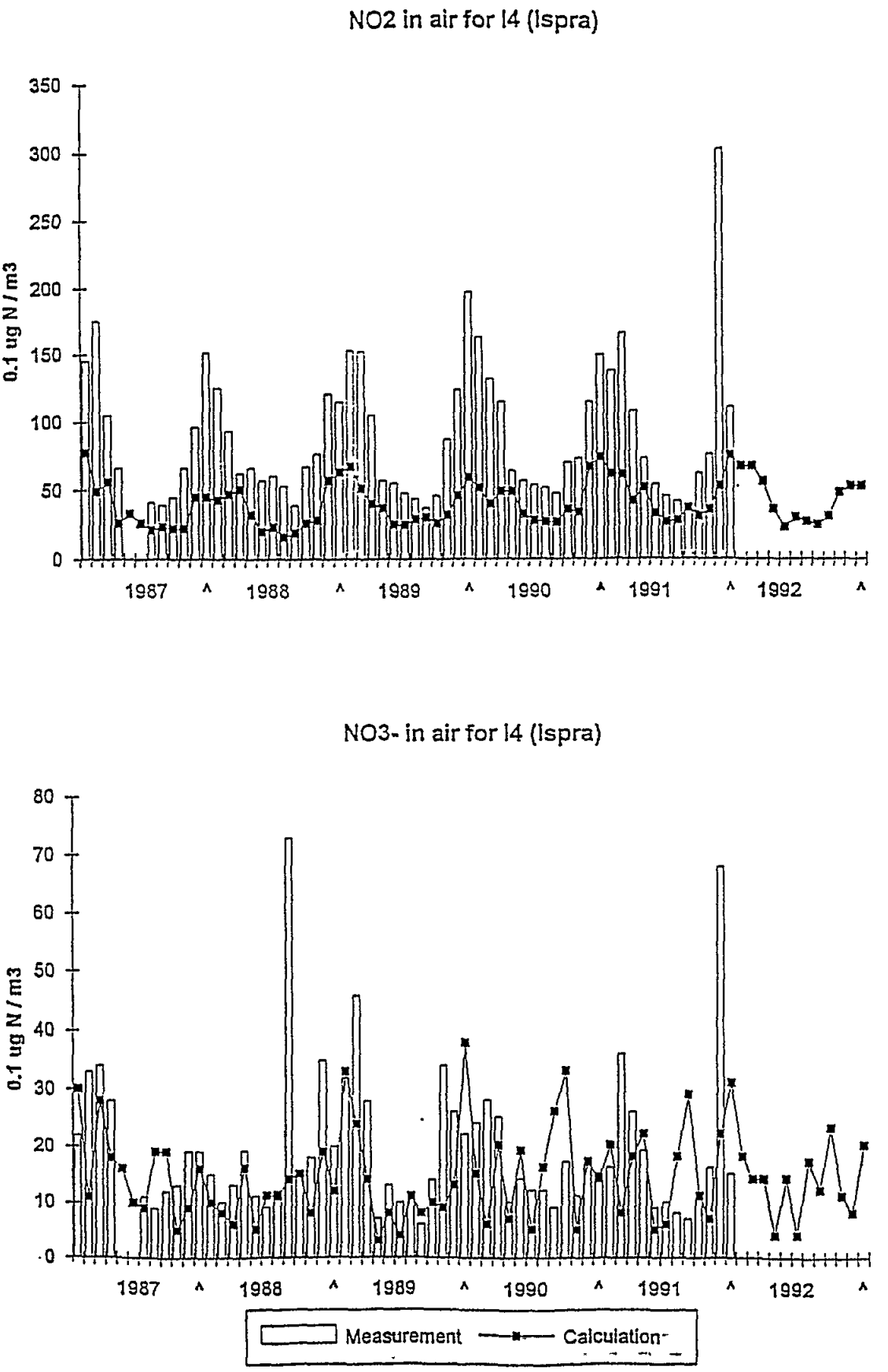


Fig. 11.10. Comparison of model and measurement S compound concentration for EMEP station YU2 closed to Adriatic coast in former Yugoslavia

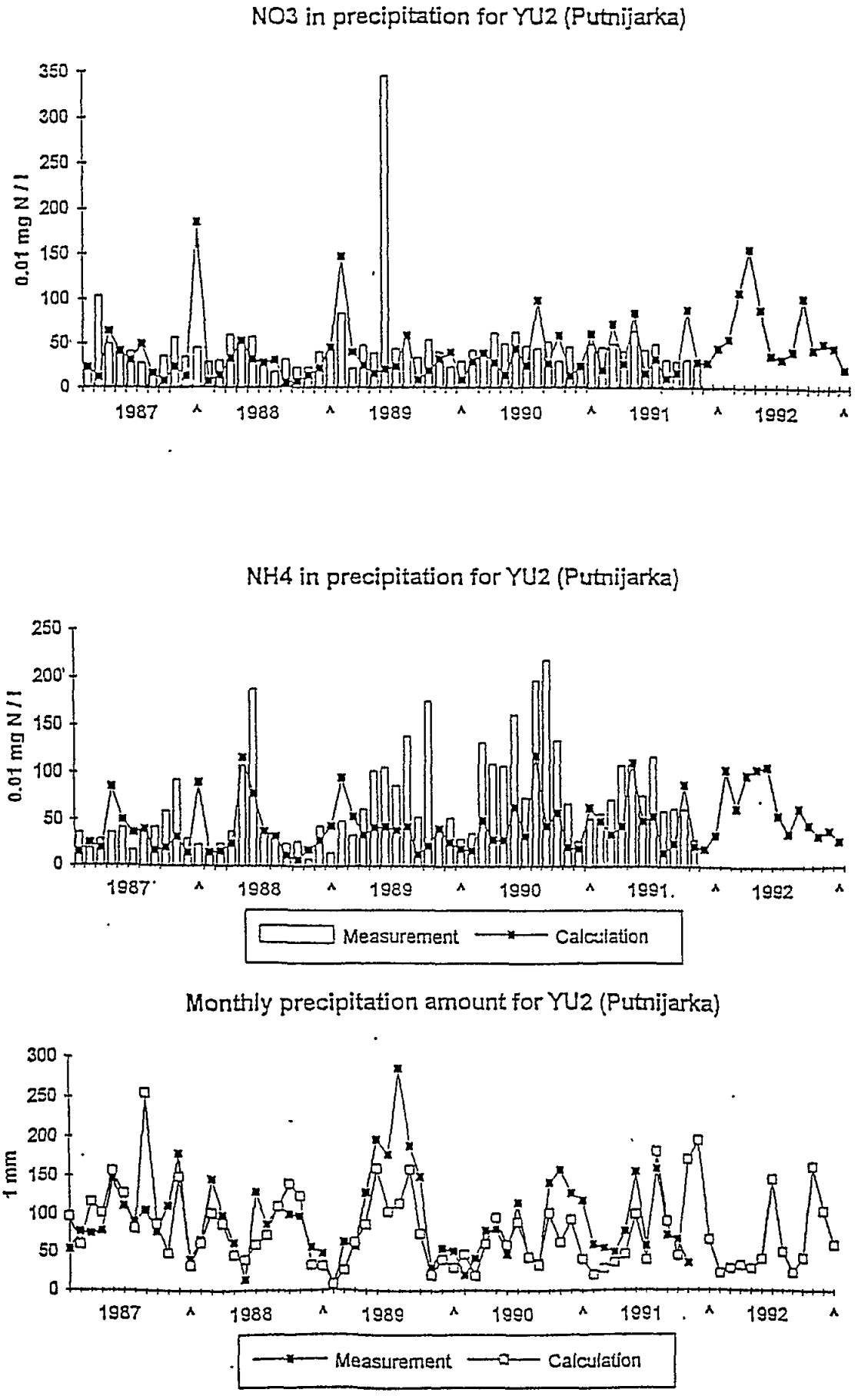
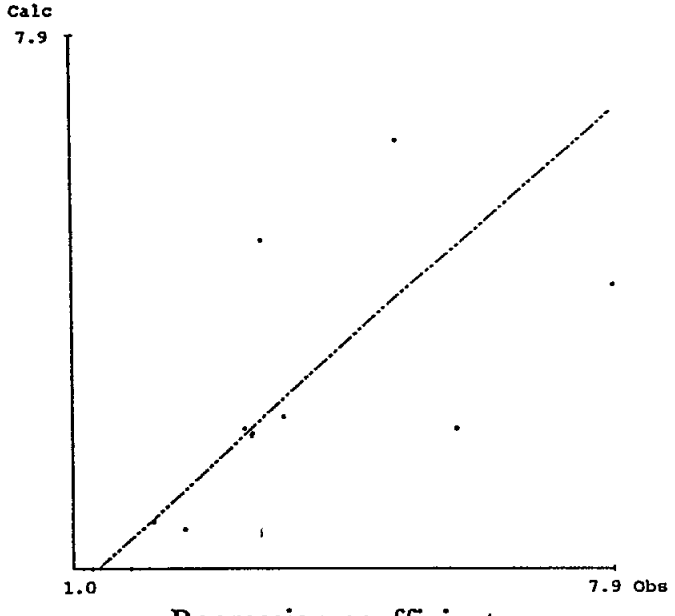


Fig. 11.11. Comparison of model and measurement N compound concentration and monthly precipitation amount for EMEP station YU2 close to Adriatic coast in former Yugoslavia

Pb wet deposition. *Unit*: 1mg Pb/m^2/year
(total annual)

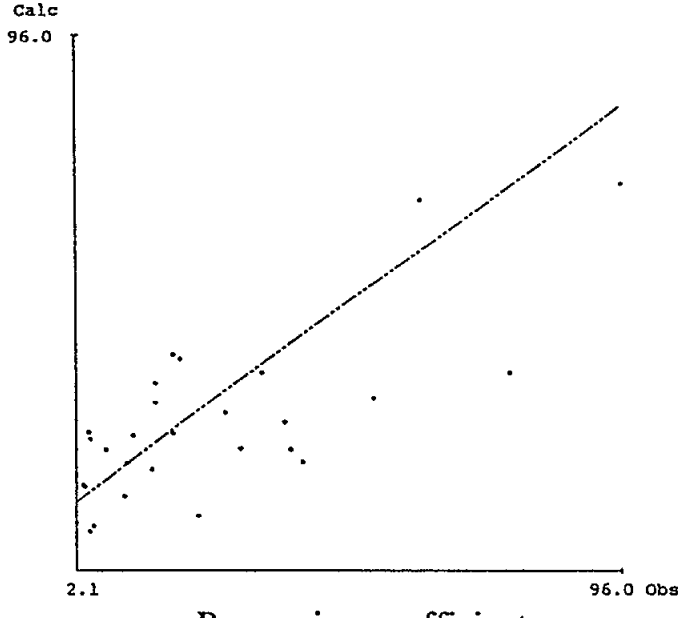


Regression coefficients:

Methods	A	B
CPEM	0.874	-0.157

Number of points	10
Mean Calculated	2.865
Mean Observed	3.556
Correlation	0.653

Pb air concentration. *Unit*: 1ng Pb/m^3
(mean annual)

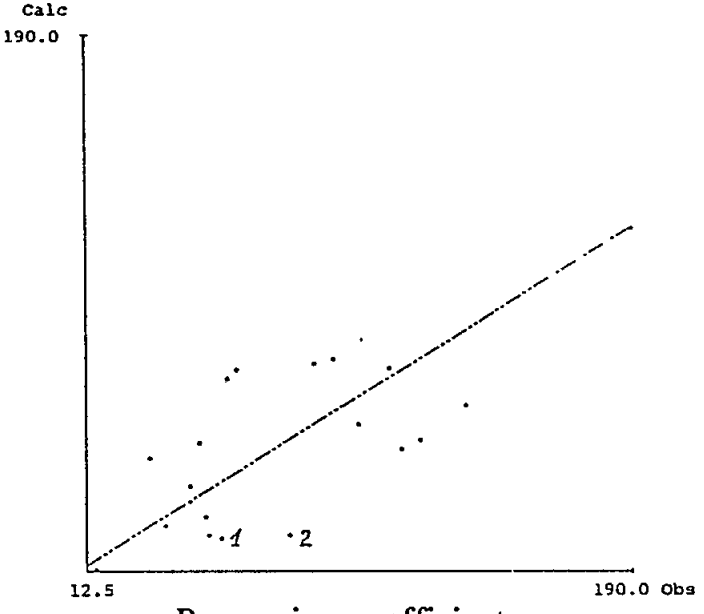


Regression coefficients:

Methods	A	B
MLS-Stand	0.238	20.172

Number of points	30
Mean Calculated	25.783
Mean Observed	23.563
Correlation	0.548

Pb in precipitation. *Unit*: 100 ng Pb/l
(mean annual)



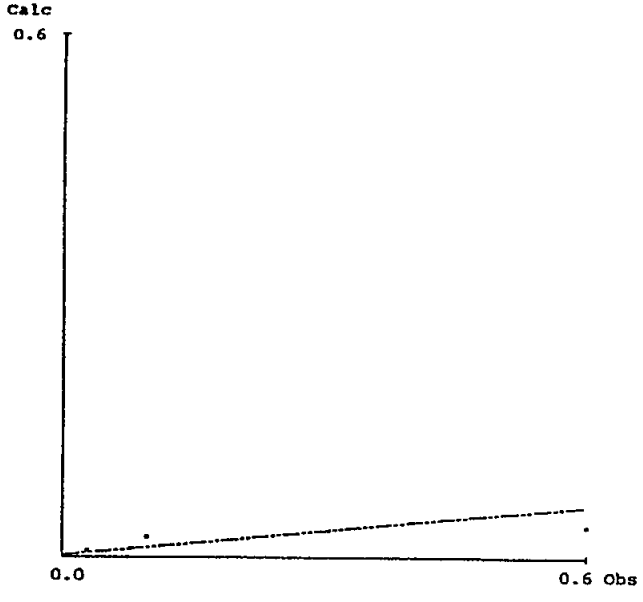
Regression coefficients:

Methods	A	B
CPEM	0.626	6.431

Number of points	21
Mean Calculated	51.548
Mean Observed	74.786
Correlation	0.746

Fig.11.12. Comparison of Pb annual calculated results with available measurement data. Annual mean data from Yugoslavian stations Ivan Sedlo and Herceg Novi in 1991 are indicated by points 2 and 1 correspondingly at the right graph.

Cd wet deposition. *Unit*: 1mg Cd/m^2/year
(total annual)

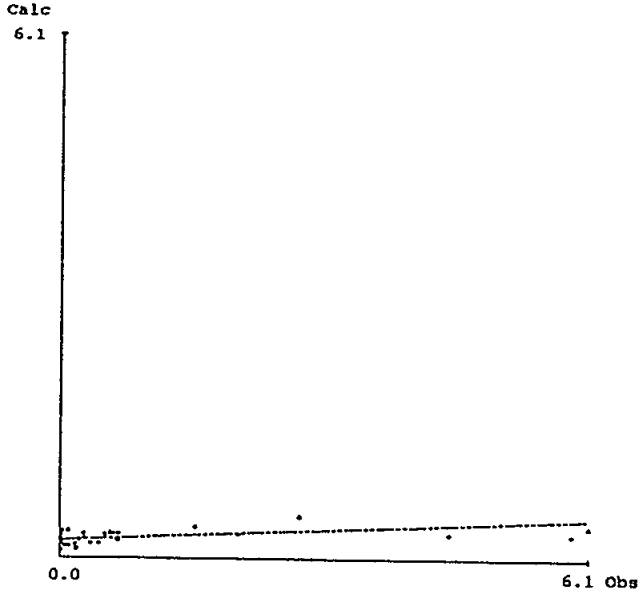


Regression coefficients:

Methods	A	B
CPEM	0.088	0.017

Number of points	5
Mean Calculated	0.028
Mean Observed	0.158
Correlation	0.848

Cd air concentration. *Unit*: 1ng Cd/m^3
(mean annual)

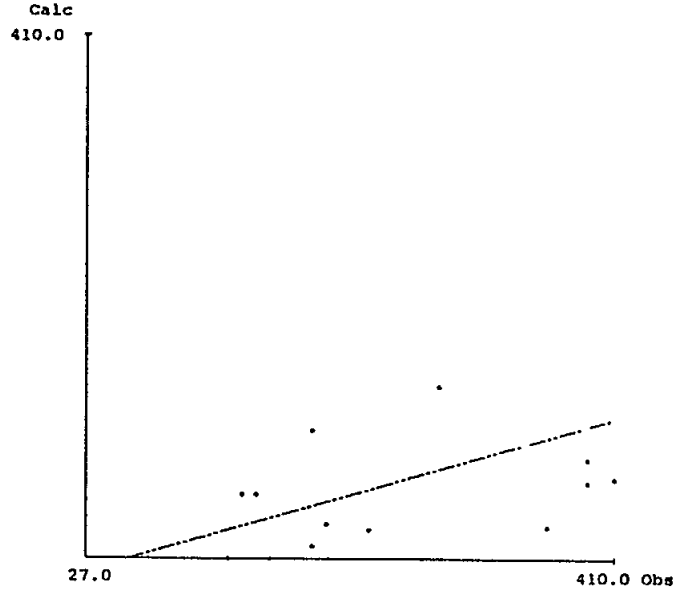


Regression coefficients:

Methods	A	B
CPEM	0.041	0.213

Number of points	30
Mean Calculated	0.249
Mean Observed	0.986
Correlation	0.504

Cd in precipitation. *Unit*: 1 ng Cd/l
(mean annual)



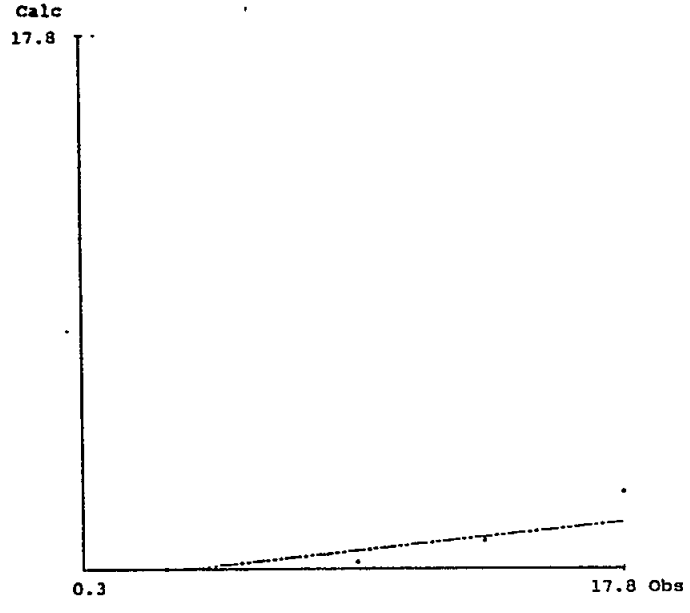
Regression coefficients:

Methods	A	B
CPEM	0.285	10.519

Number of points	13
Mean Calculated	69.453
Mean Observed	209.688
Correlation	0.468

Fig.11.13. Comparison of Cd annual calculated results with available measurement data.

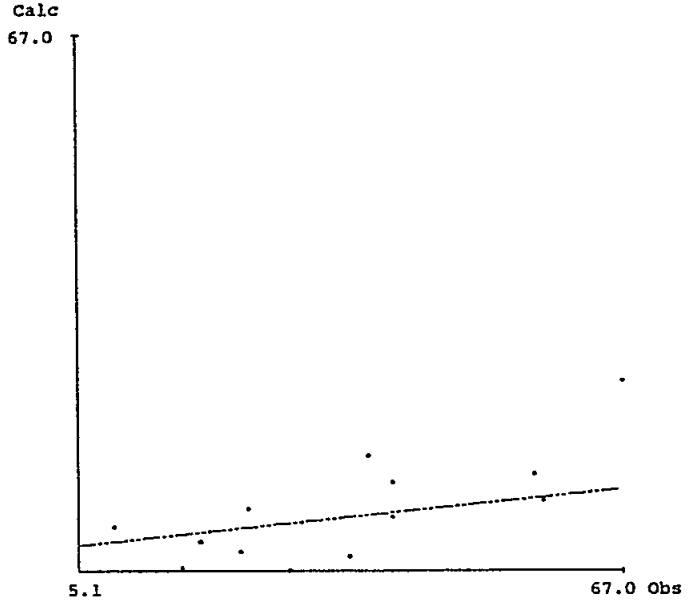
Zn wet deposition. *Unit*: 1mg Zn/m^2/year
(total annual)



Regression coefficients:

Methods	A	B
CPEM	0.102	-0.025
Number of points	5	
Mean Calculated	1.060	
Mean Observed	9.780	
Correlation	0.921	

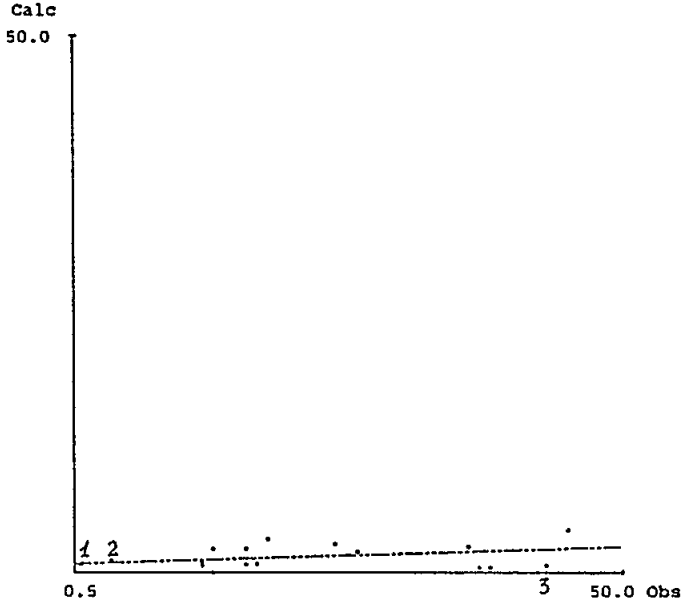
Zn air concentration. *Unit*: 1ng Zn/m^3
(mean annual)



Regression coefficients:

Methods	A	B
CPEM	0.102	7.426
Number of points	14	
Mean Calculated	11.631	
Mean Observed	34.271	
Correlation	0.745	

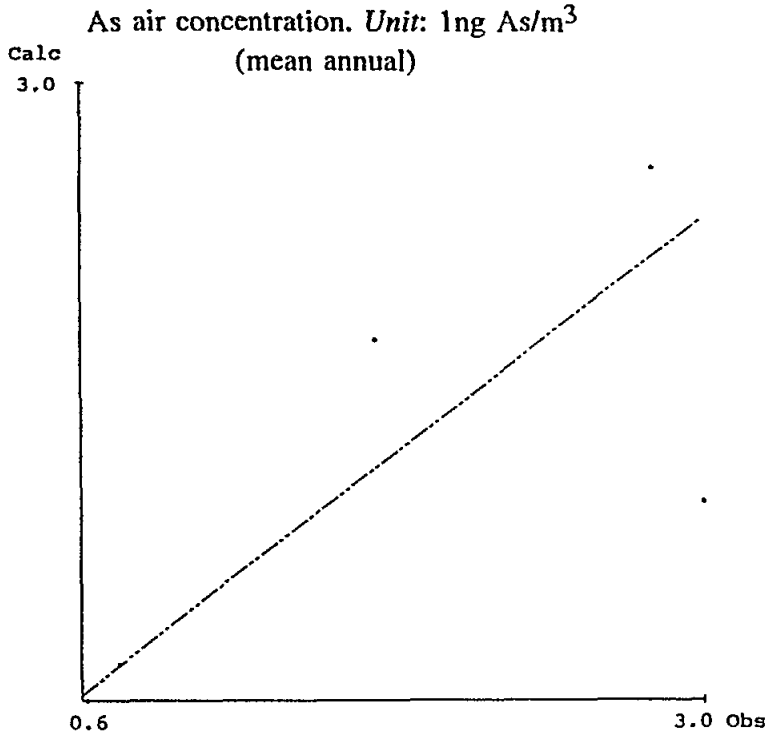
Zn in precipitation. *Unit*: 1 ug Zn/l
(mean annual)



Regression coefficients:

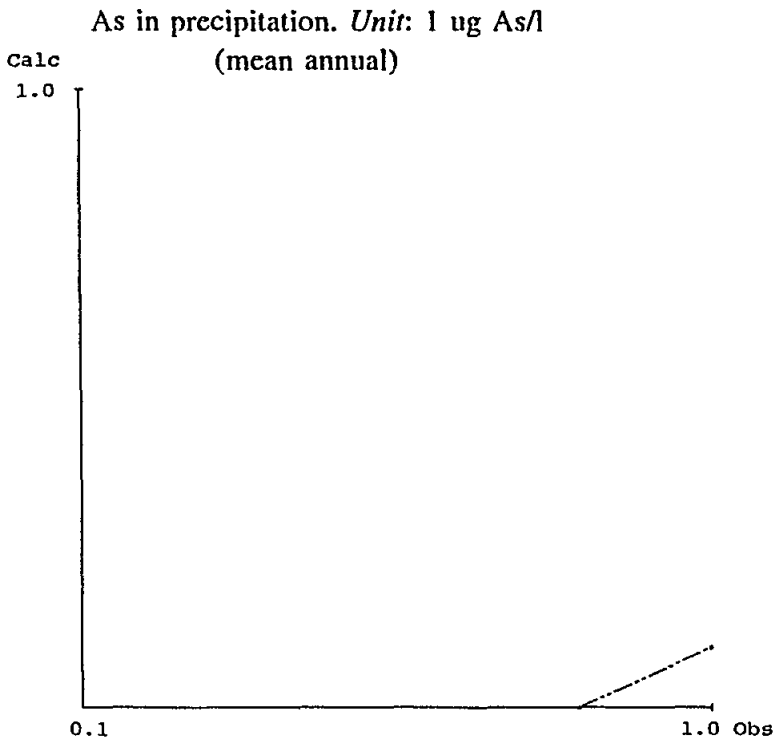
Methods	A	B
CPEM	0.031	1.213
Number of points	18	
Mean Calculated	1.801	
Mean Observed	23.381	
Correlation	0.093	

Fig.11.14. Comparison of Zn annual calculated results with available measurement data. Annual mean data from Yugoslavian stations Zavizan, Ivan Sedlo and Herceg Novi in 1991 are indicated by points 3, 2 and 1 correspondingly at the right graph.



Regression coefficients:

Methods	A	B
CPEM	0.765	0.150
Number of points	5	
Mean Calculated	1.434	
Mean Observed	1.754	
Correlation	0.733	



Regression coefficients:

Methods	A	B
CPEM	0.450	-0.310
Number of points	2	
Mean Calculated	0.095	
Mean Observed	0.900	
Correlation	1.000	

Fig.11.15. Comparison of As annual calculated results with available measurement data.

The results of comparison are as follows:

1. For Pb there is a good agreement between the model and measured data. Discrepancy between mean values does not exceed 20%. But the form of field is partially distorted that could be connected with irregular character of measurements and absence of compatibility for different sets of data.

2. Agreement for Cd is not so good. The most reliable data for Cd are the concentrations in precipitation and the majority of these measurements were carried out at the Baltic stations during 1986-1990. These measurements are of a regular character and this set of data agrees relatively well with the present model calculations. Total underestimation of mean values is within a factor of 3 which is confirmed by the same value of regression coefficient. That may be explained either by general underestimation of emissions, or by incorrect distribution of it within the calculation region.

3. The worst results of comparison are for Zn. Here the underestimation is about a factor of 10 and there is a disagreement between the regression line and the mean values ratio. One of the problems is the quality of measurement data. According to EGAP 10/4/2 (see [9]) the available measurement data for Zn wet deposition could not be considered as reliable.

4. For arsenic the main problem is the lack of measurement data. Presented graphs do not allow to draw any conclusions.

On the whole, the model - measurement comparison for heavy metals shows that the model results are acceptable as provisional assessments. But for Zn and As, as well as for Cd air concentrations, underestimation of all the parameters up to 10 times is observed. Regression slope of about 0.1 along with the good correlation (up to 0.92), make it possible to suppose that the underestimation of calculation results is partially due to the emission underestimation. This subject was discussed in Chapter 6 more thoroughly.

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XII. THE DISCUSSION ON THE MODEL CALCULATION RESULTS, CONCLUSIONS AND RECOMMENDATIONS

12.1. *Discussion of modelling results*

The prevailing transport over the Mediterranean Sea is from north-west and the prevailing winds mainly vary between western to northern [1]. As a rule it makes the situation unfavourable for the Mediterranean Sea in view of pollution transport since the main emission sources are located to the north of the Sea. In particular the northern Mediterranean countries including Turkey give 16-29% of total emissions of sulphur and nitrogen compounds and heavy metals within the calculation grid and the rest of Mediterranean countries - only 1-3%. Emissions of all the Mediterranean countries do not exceed 32% of total emissions within the calculation area (Figures 6.1-6.3, 6.5-6.8). Practically all other countries within the calculation grid, remote from the Mediterranean Sea, with integral emission not less than 68% of the total emission are also located to the north of the sea.

The existence of the prevailing atmospheric transport directions results in forming clearly defined "impact zones", constantly affected by powerful countries-emitters: Italy, Spain, France, Greece, Yugoslavia, Bulgaria and Turkey (see Figure 10.5 for instance). By the way, the annual maps of depositions from these countries reveal annually averaged transport directions (primarily from the west-northern sector) (see Appendix B). The share of the country emissions which could deposit on the sea depends on characteristic atmospheric transport patterns. These shares for As for various countries are for example, the following: Spain - 15%, Bulgaria - 15%, Israel - 8%, Syria - 4%, Turkey -12%, Albania -25%, Italy - 39%.

There is much common in the general pattern of the Mediterranean Sea pollution by sulphur and nitrogen compounds, and by heavy metals. That can be explained by the similarity of the atmospheric transport conditions for various pollutants and their emission distributions. In particular, having considered the depositions distribution with subbasins for Pb (Figure 10.4) and NH_3 (Figure 8.4), it becomes obvious that they are very similar.

The difference in pollution patterns of various compounds one can explain by the irregularities of their emission distribution, different types of sources and peculiarities of behaviour of pollutants in the atmosphere. Particularly, the emission of sulphur and nitrogen compounds as well as lead are diffused evenly across the calculation territory, but the other heavy metals are emitted mostly by a small number of powerful point sources.

In order to develop abatement strategies with the usage of calculation results it is necessary to take into account that the calculation period of one year is not sufficient since the calculation results are affected by peculiarities of meteorological situation (fluctuations of the transport relative to mean multiannual values).

The study carried out within the EMEP calculation area (Figure 3.1) demonstrated that the assessments independent on meteorological situations could be obtained only with the calculation data for 3-6 years period with their further averaging [10].

At the same time one should bear in mind that measurements and calculation results have principal unavoidable uncertainties (about 20%), connected with the singularity of measurements used for the model verification [10].

Results of heavy metals transport calculations should be considered as provisional for the following reasons:

1. The heavy metals behaviour in the atmosphere is a sophisticated problem which requires further investigation. Particularly, there is no commonly accepted opinion on their phase state in the atmosphere (in the work presented, the modelling was done upon the assumption, that heavy metals are adsorbed by aerosols).

2. The only complete emission data set available at the moment of calculations was for 1982, published in 1992 [2]. Lists of emission sources, taken into consideration in data sets and in publications available differ from each other both with types of sources and their priorities [4,5]. Therefore, the emission data set used, should not be considered as a complete one. The degree of uncertainty put into both the original data and additional estimates for the territories outside the EMEP grid, made by MSC-E, are considerable. For instance, the total emission of Zn for 1979 in western and central Europe in work [5] exceeds the one used in calculation two times Data taken from the work [2] show, that the emission estimates for individual countries made by various researchers, sometimes differ more than 10 times.

The accuracy of the calculation results of heavy metal emission assessments is evaluated by the authors within a factor of 3. Besides, the emission of lead should have been substantially reduced since 1982, due to a prohibition of leaded gasoline in European countries.

3. The measurement data available are very scarce, sometimes controversial (see, for example, [6,p.5,points 4.1-4.3]), and contain fragmentary parts for different years. Data for 1991 as well as for the Mediterranean are even more sparse. That is why, although presented in the report, the comparison with the measurements can not be considered sufficiently correct. In addition to the comparison with measurements, intercomparison with other calculation results available was also made. The results of our calculations together with the results received in [4] and [7] are presented in Table 12.1.

Since the calculations in these publications were made within slightly different grids (for instance the Mediterranean Sea is completely covered in the MSC-E work only), we rearranged our results in a way, as we would use the same grid and emission data as in each of the works [4] and [7]. For that purpose, the part of deposition on the territory of the Mediterranean, not covered by the grid in each of these works, being compared to, was subtracted from the total Mediterranean deposition. Then inputs of the countries of northern Africa, Middle East and Turkey were subtracted from the total, both for the Mediterranean and the Black Seas, because these countries had not been considered in [4,7]. After that the deposition was recalculated according to the ratio between the emission values used. For comparison with the work [4] the ratio between the total emissions was used since only this value was given in this work. The ratio for individual countries was used for comparison with the work [7], since practically the same data were used, except the data for Pb (which in [7] were updated for 1985).

The results of MSC-E recalculated as described above are marked (1)→(2) for comparison with work [7] and (1)→(3) for work [4].

As it follows from Table 12.1, the discrepancy between the corrected MSC-E results and the results of the papers [4] and [7] for the Mediterranean, do not exceed 20%. The difference is higher for the Black Sea and makes up 40% for Cd, comparing to paper [7]. It should be noted that the MSC-E evaluations for Cd and Pb are overestimated comparatively to the papers [4,7] and underestimated comparatively to the observation data.

As a whole, the results obtained allow us to assert, that the share of the total of heavy metals emission within the calculation grid, which deposit on the Mediterranean and Black Seas, is estimated with an error of not more than 40%.

There is a possibility to make a comparison between the atmospheric and the riverine inputs to the Mediterranean Sea. According to [8] inflow with riverine run-off for different kinds of pollutants are: 800-1,200 kt of N; 4,300-4,500 t of Pb and 21,000-29,000 t of Zn per year. Our calculation results show, that the deposition of airborne bound nitrogen (NO_x+NH_y) on the sea amounted to 1,000-1,100 kt (as N), that is practically equal to the riverine nitrogen. The estimate of the airborne Pb deposition on the Mediterranean Sea is

about 7,400 t, that is even more, than the estimate of the riverine input. As for Zn, it appears that the riverine input to the sea is much greater than the atmospheric one (2,500 t).

Table 12.1. Comparison of depositions on the Mediterranean and the Black Seas, obtained by various authors, and the emissions used. (*Unit = 1 t*)

		Metals			
SEAS	VARIANTS	As	Cd	Pb	Zn
MDT	(1)	201	73	7404	2523
	(2)	218	59	3550	2093
	(1)→(2)	170	61	4300	2030
	(3)		31	2000	
	(1)→(3)		35	2400	
BLC	(1)	67	21	1714	681
	(2)	58	12.5	1101	493
	(1)→(2)	63	20	1500	620
	(3)		7.5	520	
	(1)→(3)		11	650	
Total emission within calculation region	(1)	5054	1172	95×10^3	$42.0 * 10^3$
	(2)	4950	1143	85×10^3	$40.5 * 10^3$
	(3)		590	38×10^3	

Calculation results and emission used:

(1) - MSC-E,
(2) - [7],
(3) - [4].
The results of MSC-E recalculated for the direct comparison with other results in accordance with the emission used and the region of calculation:

(1)→(2) - (MSC-E)→[7],
(1)→(3) - (MSC-E)→[4].

12.2. Conclusions

1. The calculation of the Mediterranean region pollution by sulphur and nitrogen compounds and heavy metals has been carried out for 1991.

2. About 7% of sulphur and bound nitrogen emissions within the calculation region deposit on the Mediterranean Sea, and about 9% on the Mediterranean Sea, the Sea of Marmara, the Black Sea and the Sea of Azov in total.

The depositions of heavy metals on the Mediterranean Sea and on all southern European Seas vary from 4% (As) to 8% (Pb) and from 5% (As) to 10% (Pb) respectively

3. The adjacent countries received in 1991 about 15% of sulphur and bound nitrogen emission within the calculation region. The same values for heavy metals make up

from 10% (As) to 17% (Pb). These values roughly characterize the amount of deposition on the watershed of the Mediterranean Sea, part of this pollution with river runoff enters coastal waters, and thus the contribution of airborne pollution to the total pollution of the sea will be greater than the pollution resulted only from deposition directly on the sea.

4. The Mediterranean countries which emissions amount to 20-25% of the total sulphur and nitrogen emissions and about 18%-32% of the total heavy metal emissions within the calculation region are the main source of the Mediterranean Sea airborne pollution (60%-80% for sulphur and nitrogen compounds and 65%-85% for heavy metals).

The input of remote countries is 20%-40% for sulphur and nitrogen compounds and 15%-35% for heavy metals.

The input of adjacent countries located to the south and to the east of the Mediterranean Sea does not exceed 10% of the total contribution of all the Mediterranean countries.

5. Seasonal variations of deposition are significant for all the substances considered and they are explained by seasonal emission variations and the atmospheric transport conditions.

6. For some compounds (e.g. Pb, N) the airborne input is practically equal or even more than the riverine input.

12.3. Recommendations for future research

1. The meteorological data for 1991 considerably deviated from the climatic ones. Therefore the present estimates of airborne pollution may differ from the multiannual values. So it would be expedient to fulfill the calculation for three years at minimum, to obtain averaging results closer to the multiannual values and to make the trend analysis. Continuous calculations will be needed to provide information on current states of airborne pollution and to develop any air pollution abatement strategies.

2. Further improvement of the emission data base will allow to make refined recalculations of the deposition of Pb, As, Cd, and Zn on the Mediterranean region. At the same time it would be useful to make the calculations for other toxic compounds, such as Hg, Ni, Cr, V, Cu, benzo(a)pyren and chlororganic compounds.

3. In addition to simulation of the whole Mediterranean region it would be desirable to make the "local" modelling considering orography, local winds, types of underlying surface (which is the most important for the Adriatic Sea), and so on. The development of applied programmes for various regional or local situations could be useful as well.

4. The experience gained in the research of the Baltic Sea airborne pollution shows that an important source of pollution for the seas is their watershed [9]. Pollutants deposited on a watershed are being washed out to coastal waters with riverine run-off. It would be expedient to assess quantitatively the contribution of airborne pollution to the pollution of the Mediterranean Sea from its watershed.

5. In order to obtain accurate and comprehensive assessments of the airborne input to the pollution of the Mediterranean and the Black Seas it is desirable:

- to have more representative measurement data obtained from an expanded network of stations, to establish "marine stations" on islands in the Mediterranean Sea or to use data obtained from other stations located outside the EMEP grid;
- simultaneously, intercalibration of measurements at the stations would be necessary, as well as harmonization of the measurement and the calculation results.

6. The refinement of the emission data for countries, especially for the Mediterranean and those ones, located beyond the EMEP grid limits and the consideration

of the pollution from shipping are the task of primary importance in order to improve the accuracy of assessment of the Mediterranean and the Black Seas pollution by airborne compounds.

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APPENDIX A.

Maps of dry and wet depositions and annual concentrations in air and precipitation of sulphur and nitrogen compounds, and dry deposition and concentrations in air and precipitation of heavy metals in 1991.

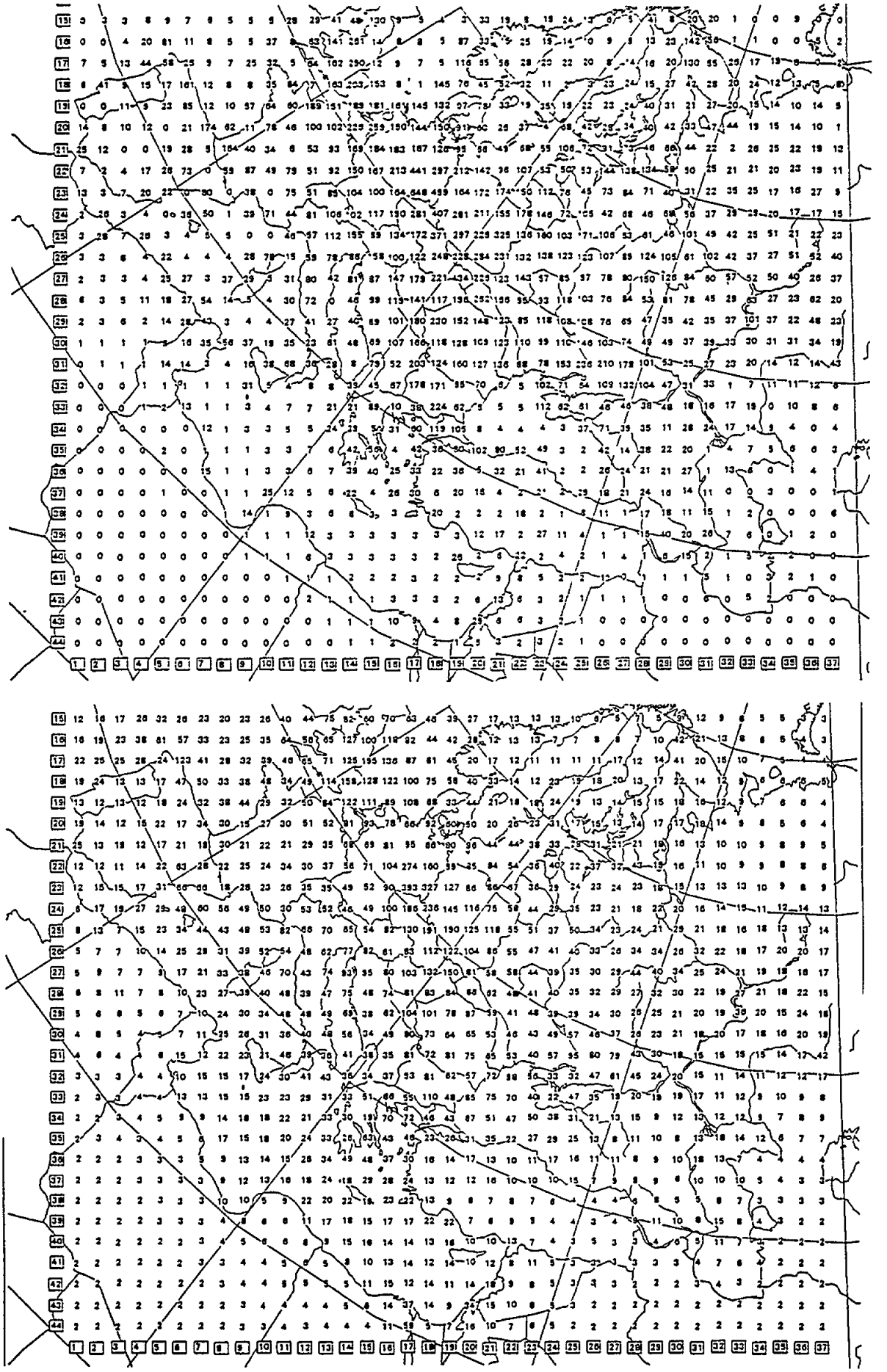


Fig.A1 Deposition of sulphur compounds (10 mg S/m^2/year) (upper - wet deposition, lower - dry deposition).

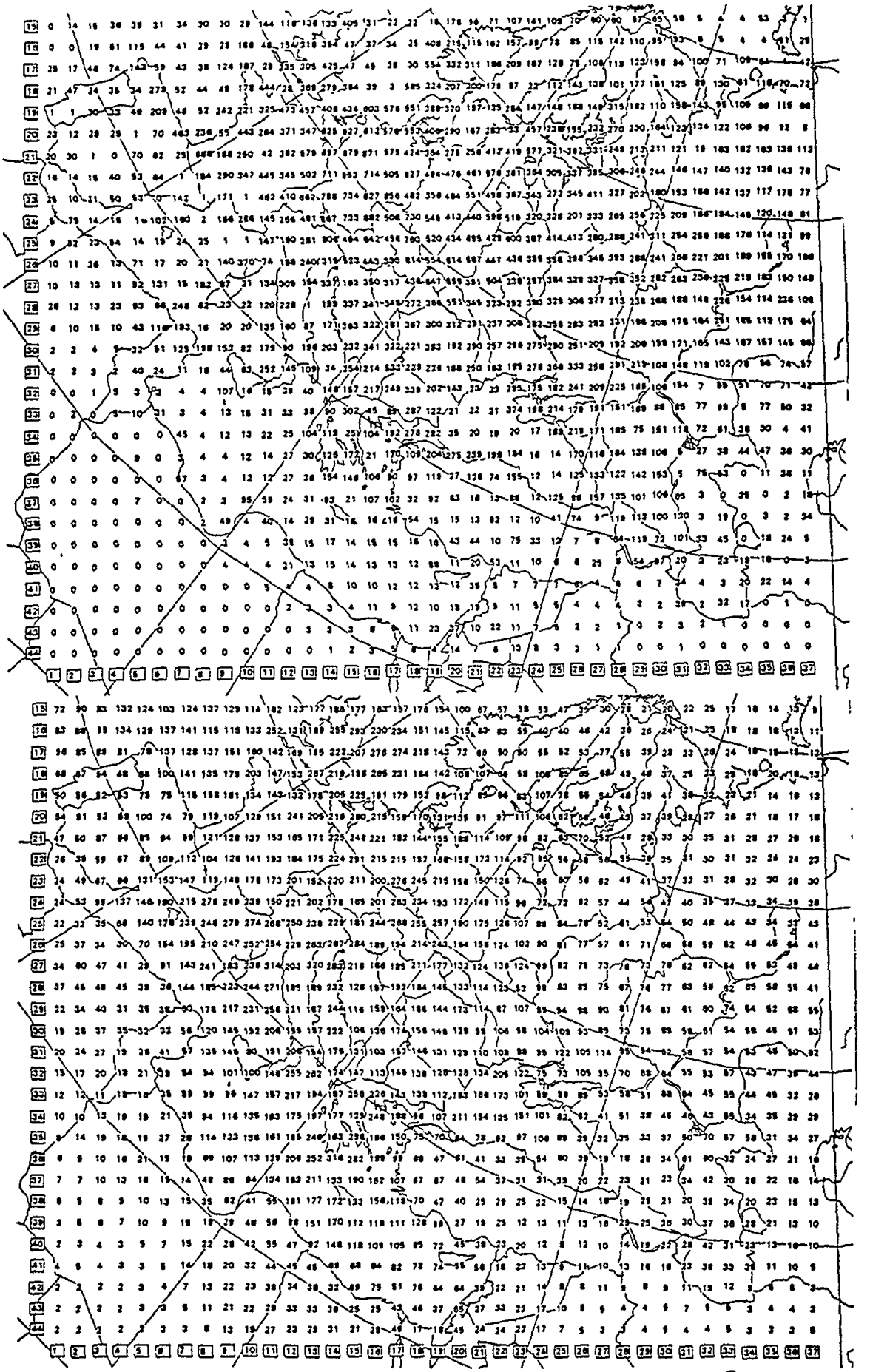


Fig. A2 Deposition of oxidised nitrogen (mg N/m^2 year) (upper - wet deposition, lower - dry deposition).

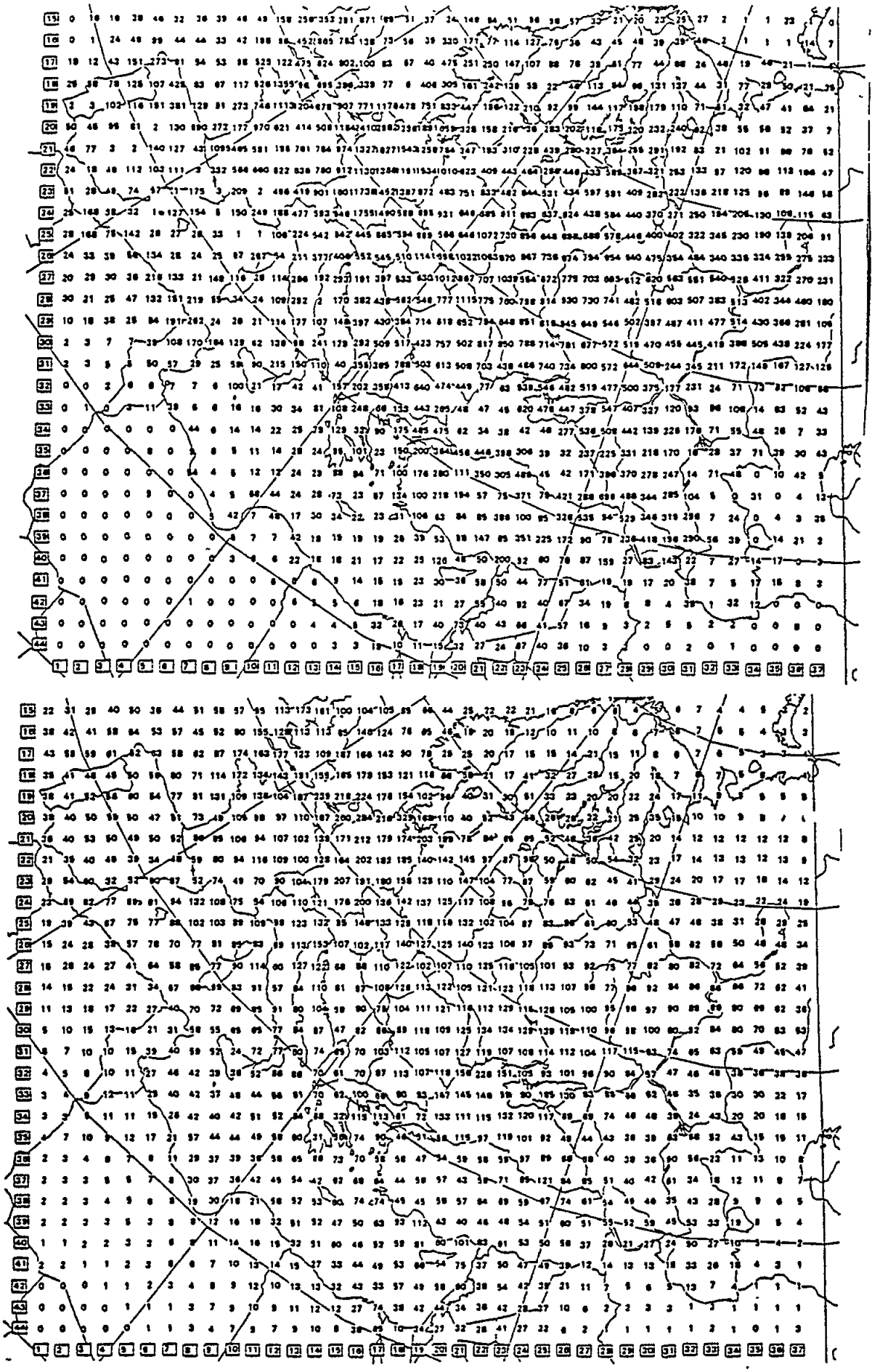


Fig. A3 Deposition of reduced nitrogen (mg N/m^2 year) (upper - wet deposition, lower - dry deposition).

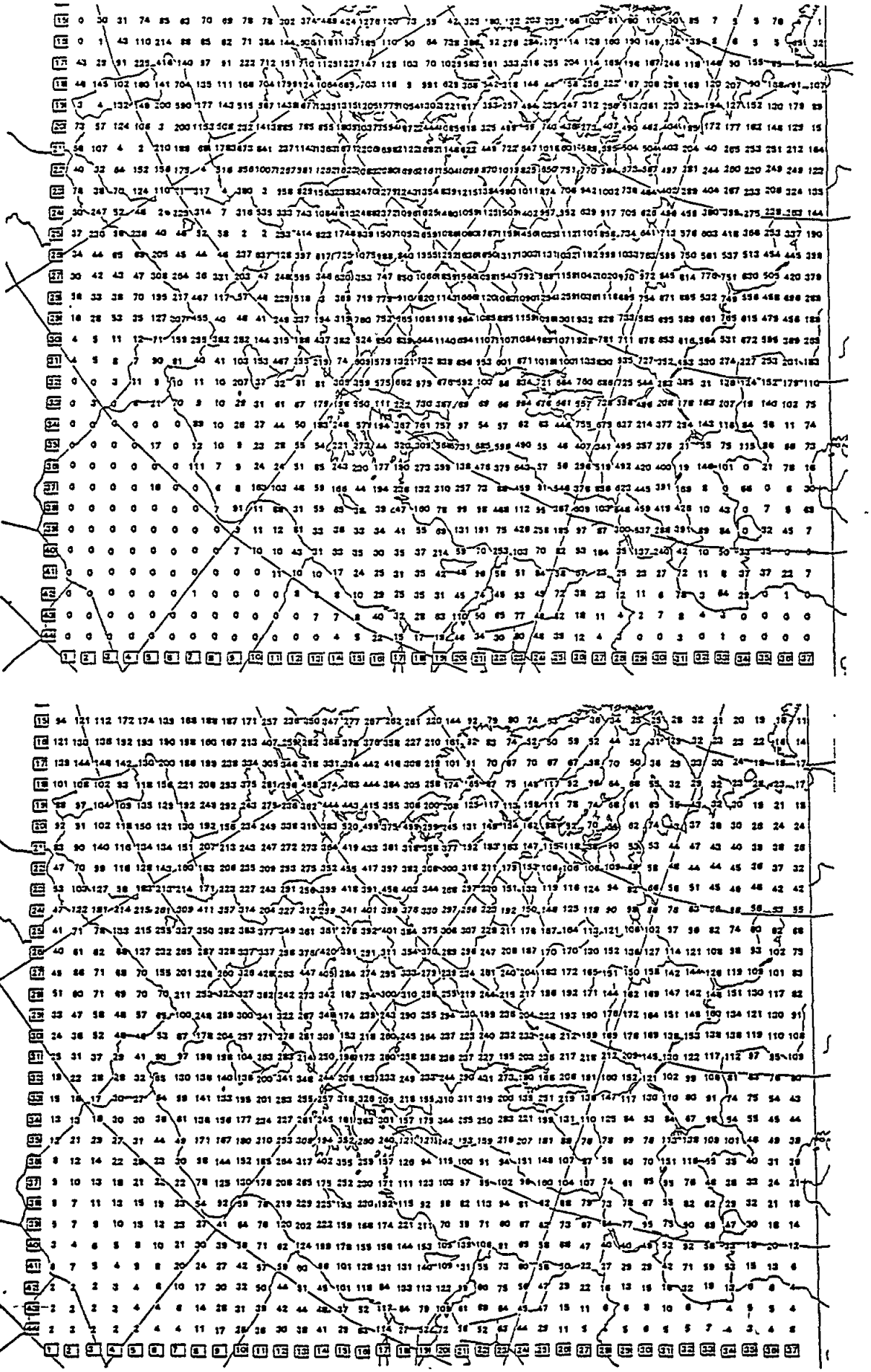


Fig.A4 Deposition of bound nitrogen (mg N/m^2/year)
(upper - wet deposition, lower - dry deposition).

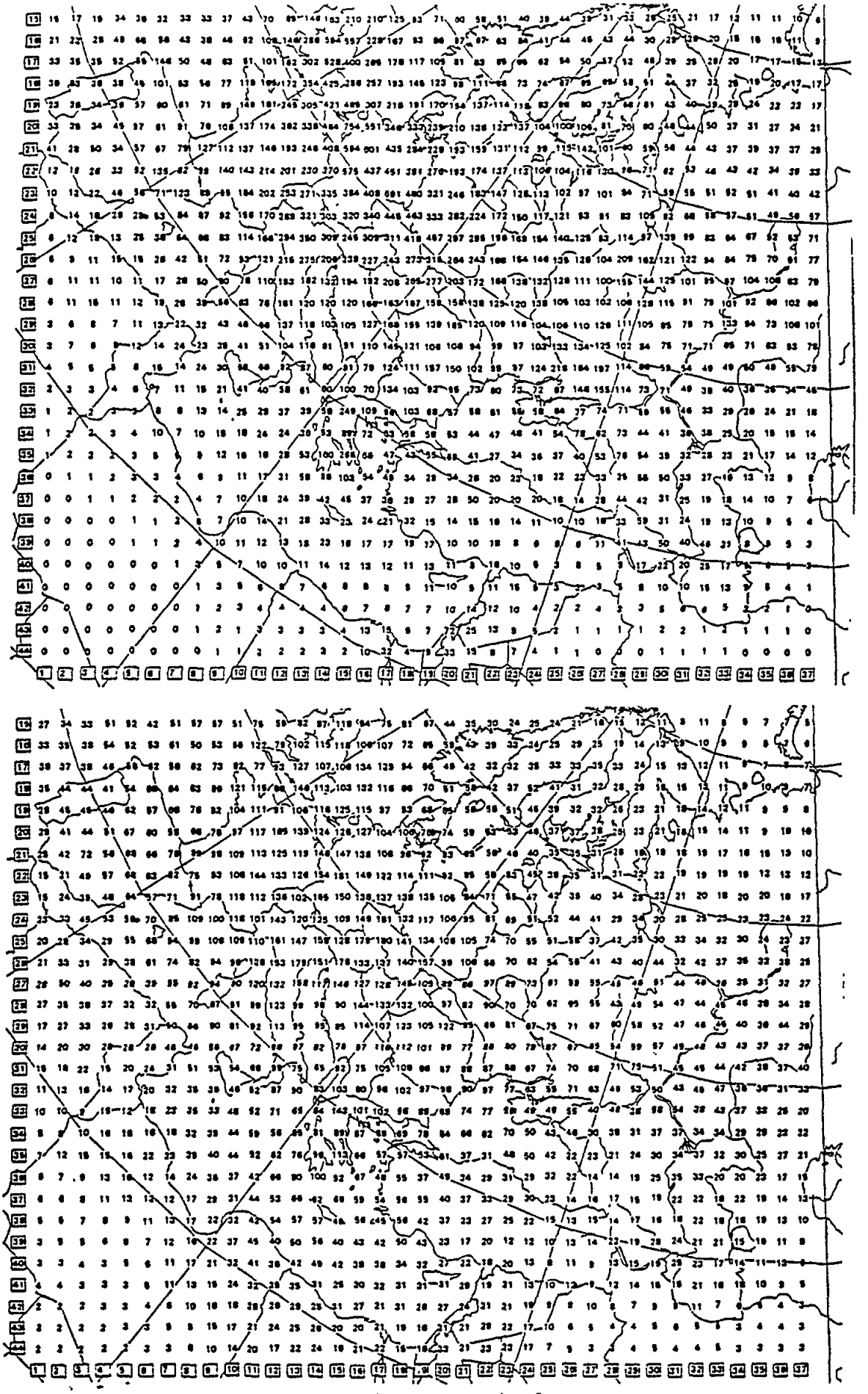


Fig. A5 Mean annual air concentrations (0.01 ug N/m^3) (upper - NO_2, lower - NO_3+HNO_3).

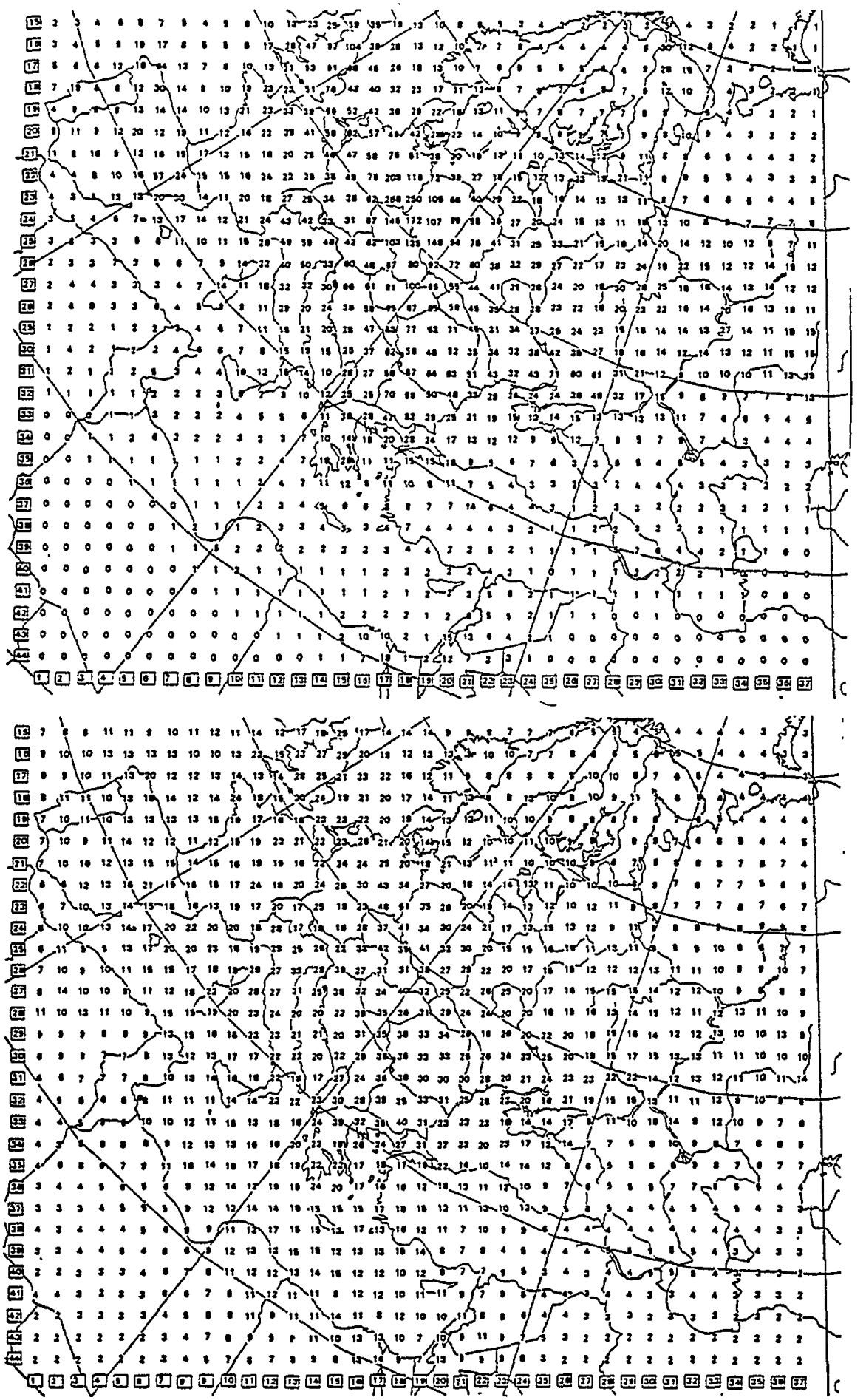


Fig. A6 Mean annual air concentrations (0.1 ug S/m^3) (upper - SO_2, lower - SO_4).

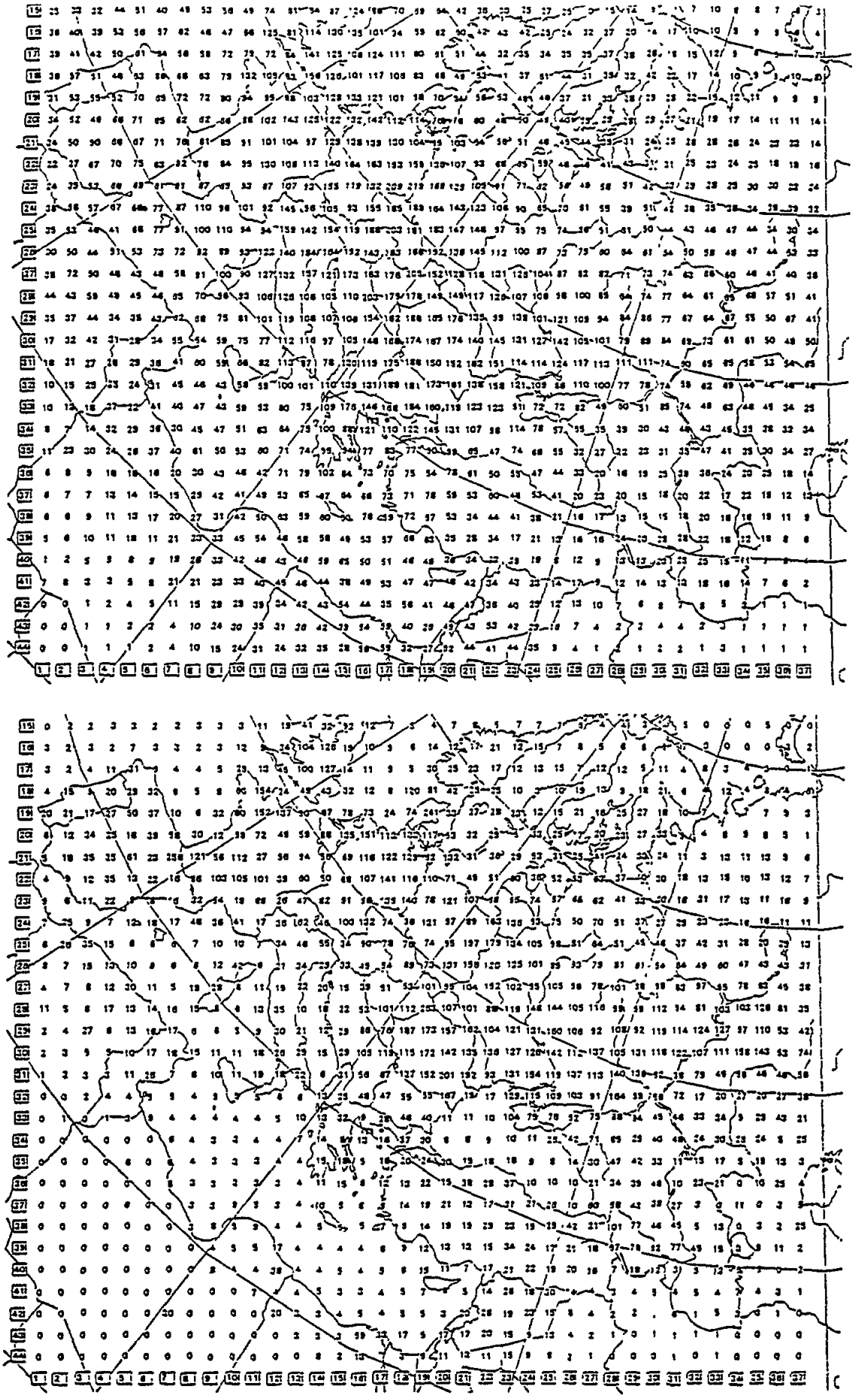


Fig. A7 Mean annual concentrations of NH4 in air (0.01 μg N/m^3) (upper) and in precipitation (0.01 mg N/l) (lower).

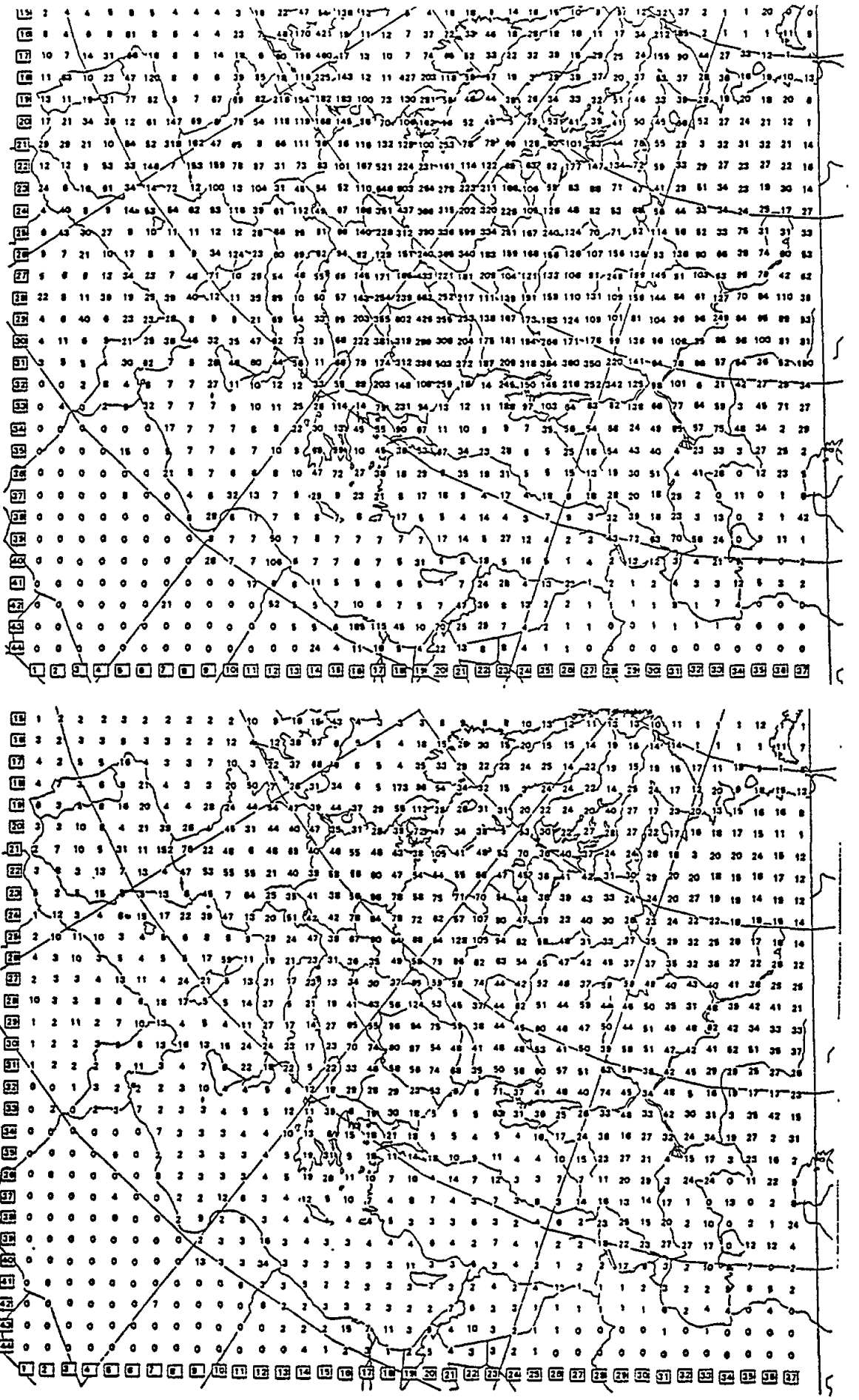


Fig.A8 Mean annual concentrations of SO_x (0.01 mg S/l) (upper) and NO_x (0.01 mg N/l) (lower) in precipitations.

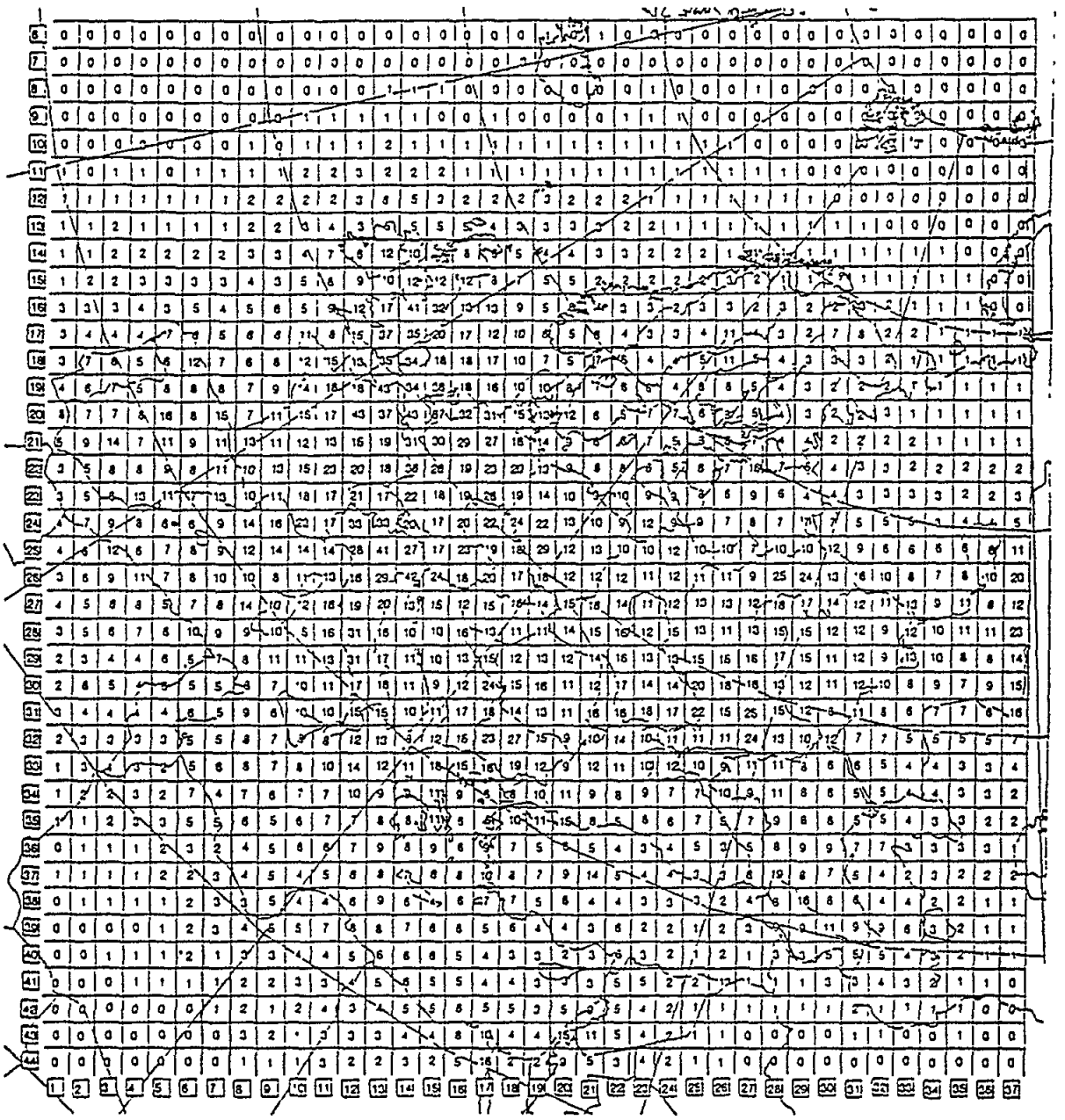
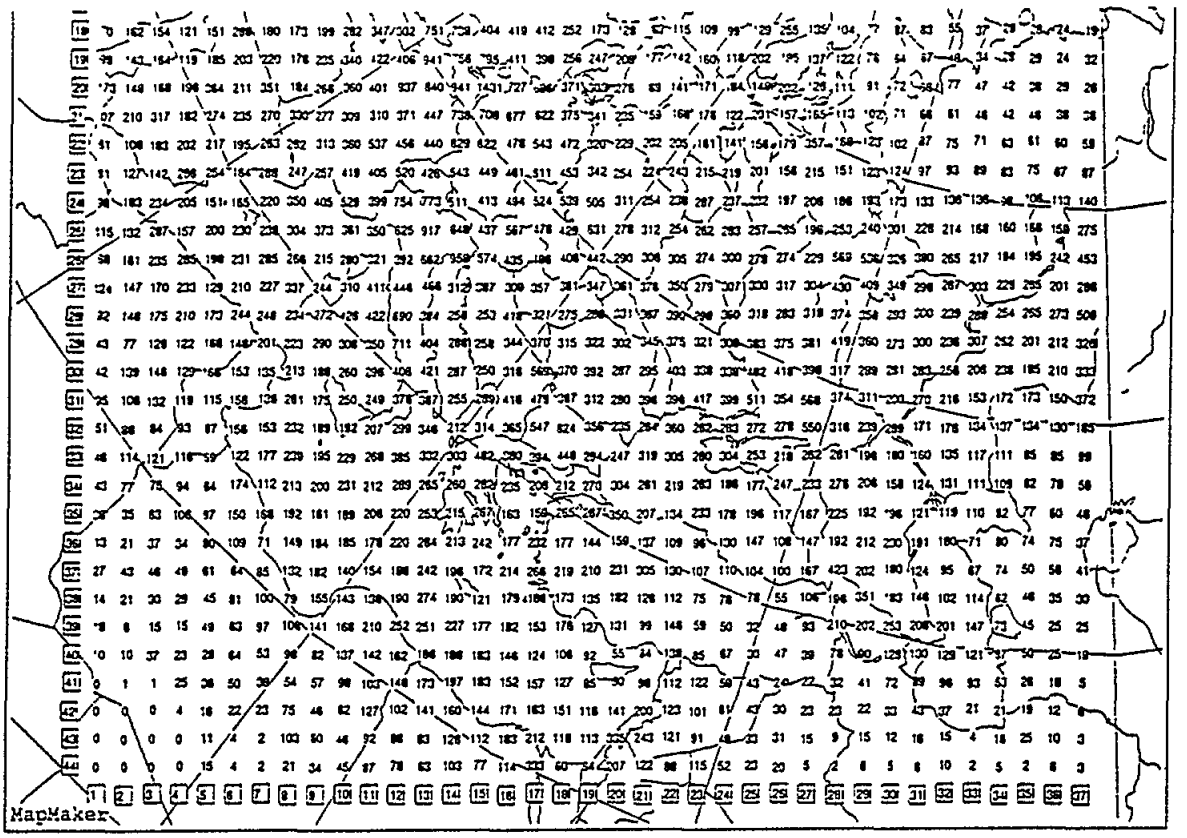


Fig. A9. Dry deposition of lead in 1991 (100 ug Pb/m^{2}year).



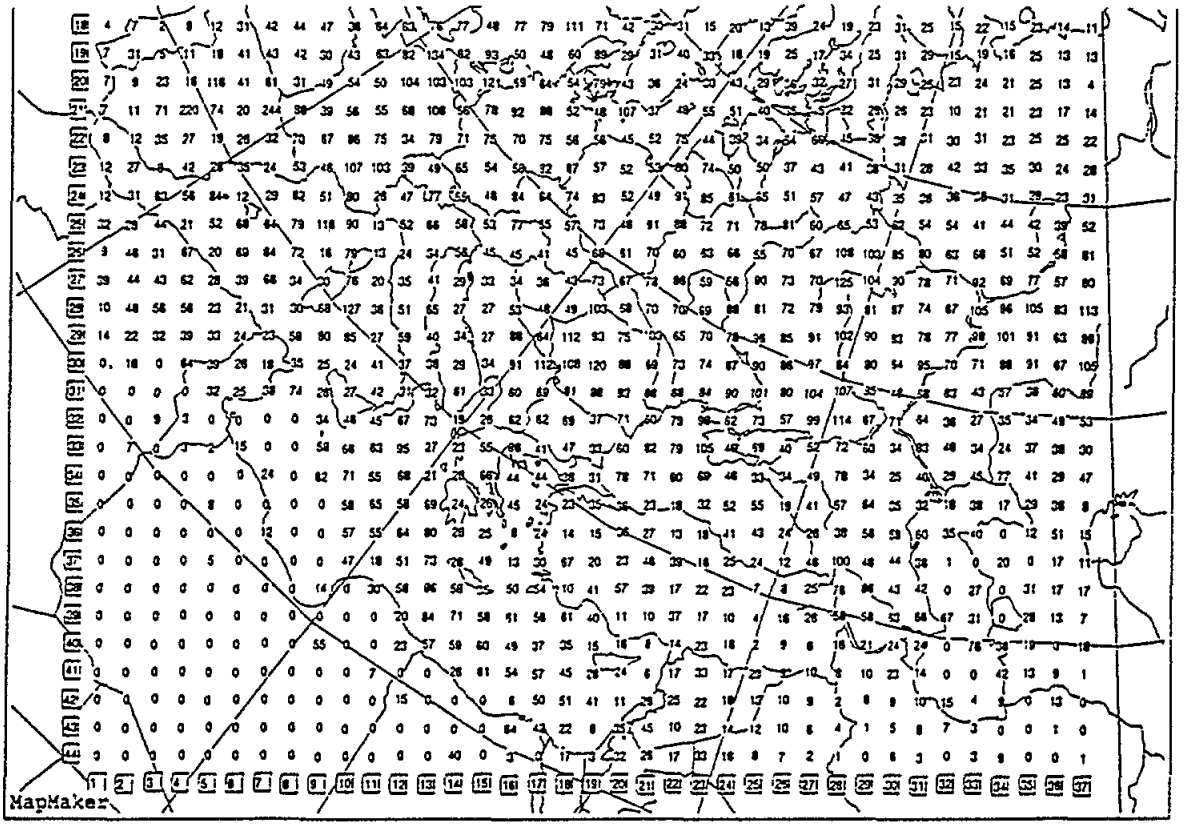


Fig.A10 Concentration of lead in air in 1991 (0.10 ng Pb/m^3), upper panel.

Concentration of lead in precipitation in 1991 (100 ng Pb/l), lower panel.

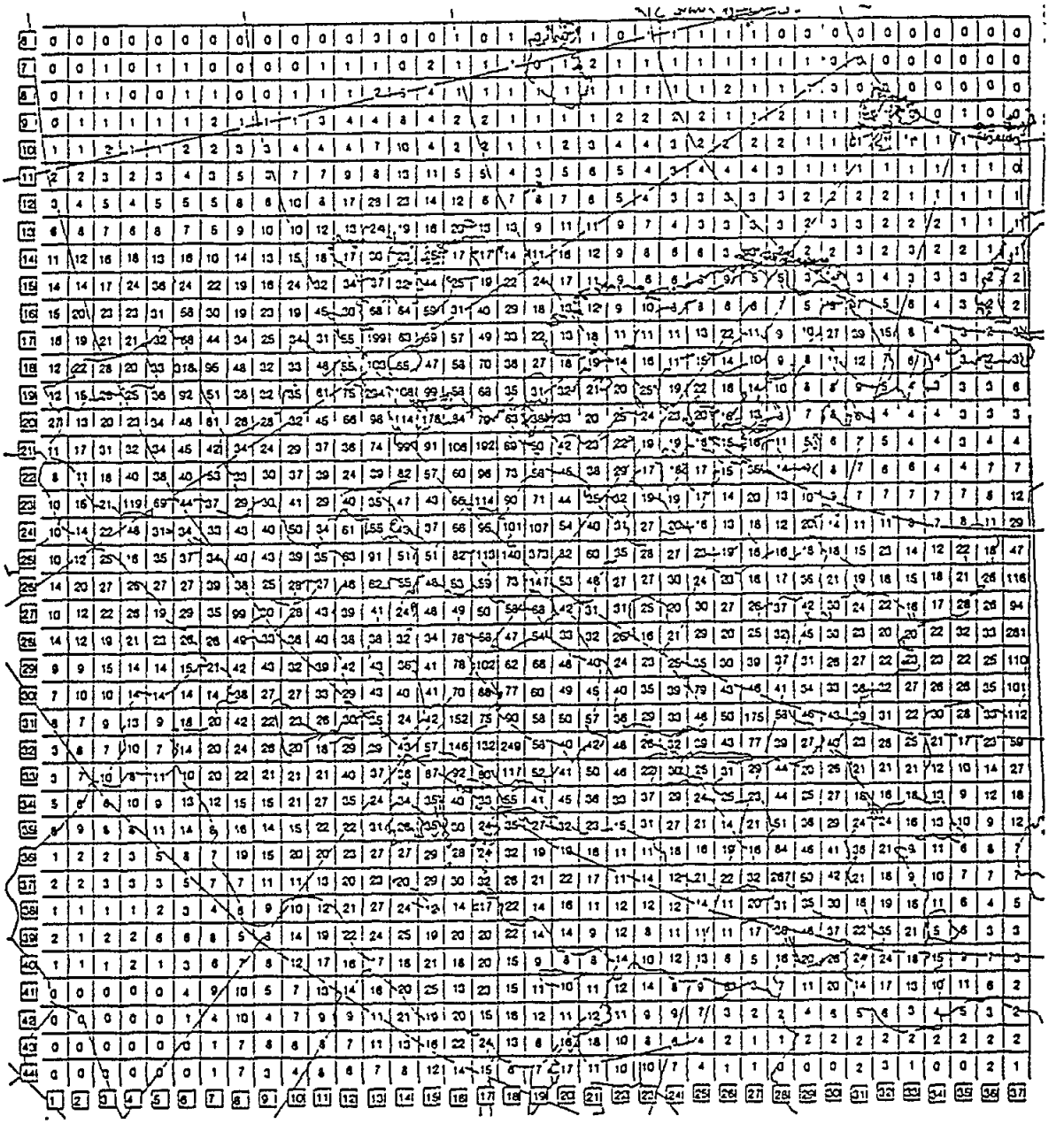
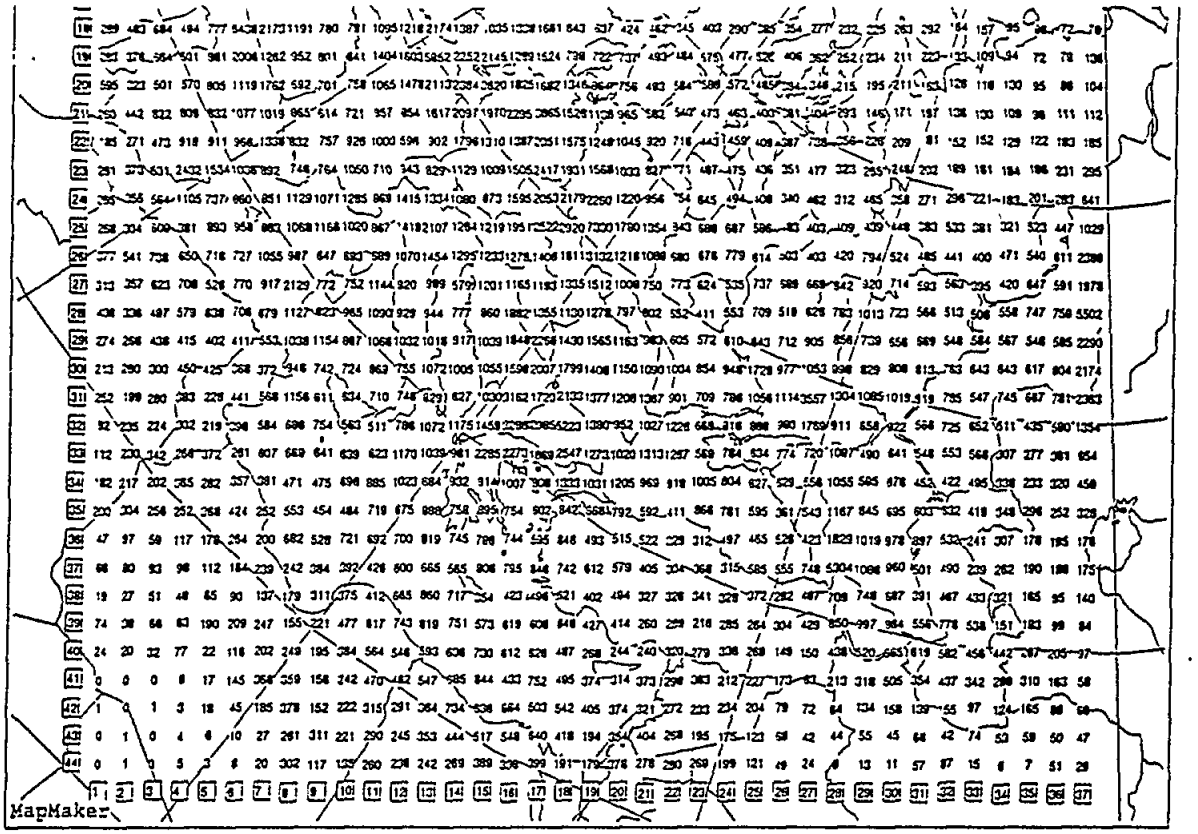


Fig. A11. Dry deposition of zinc in 1991 (10 ug Zn/m^{2}year).



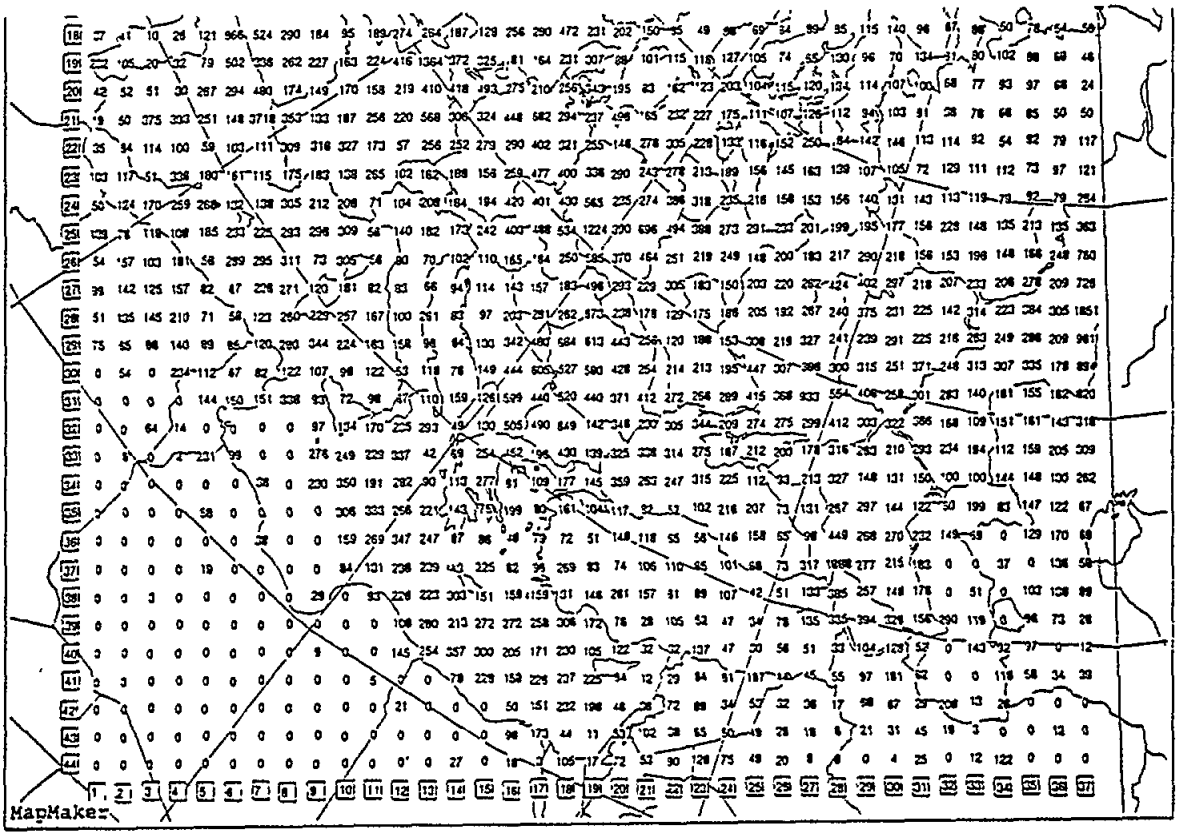


Fig.A12 Concentration of zinc in air in 1991 (0.01 ng Zn/m^3), upper panel.

Concentration of zinc in precipitation in 1991 (10 ng Zn/l), lower panel.

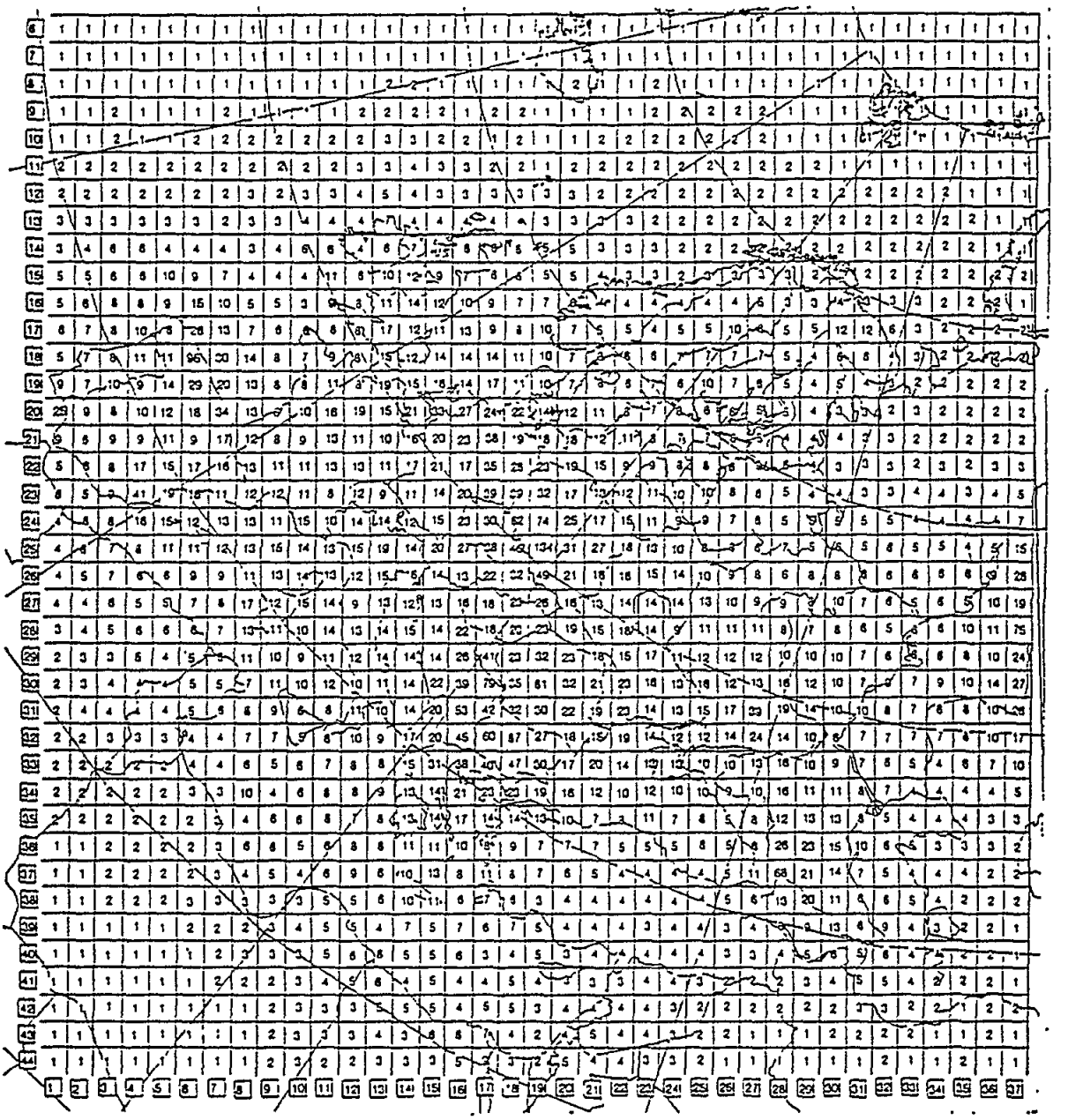
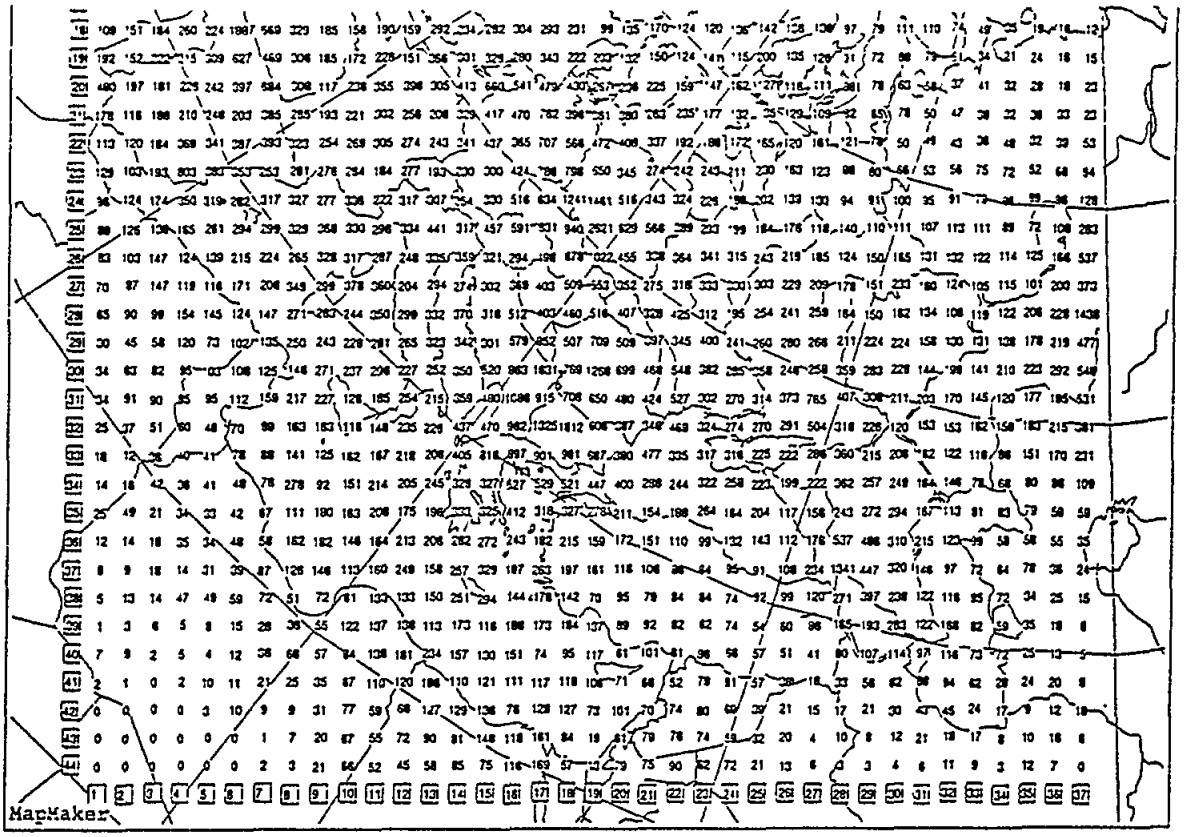


Fig. A13. Dry deposition of cadmium in 1991 (1 ug Cd/m^{2}year).



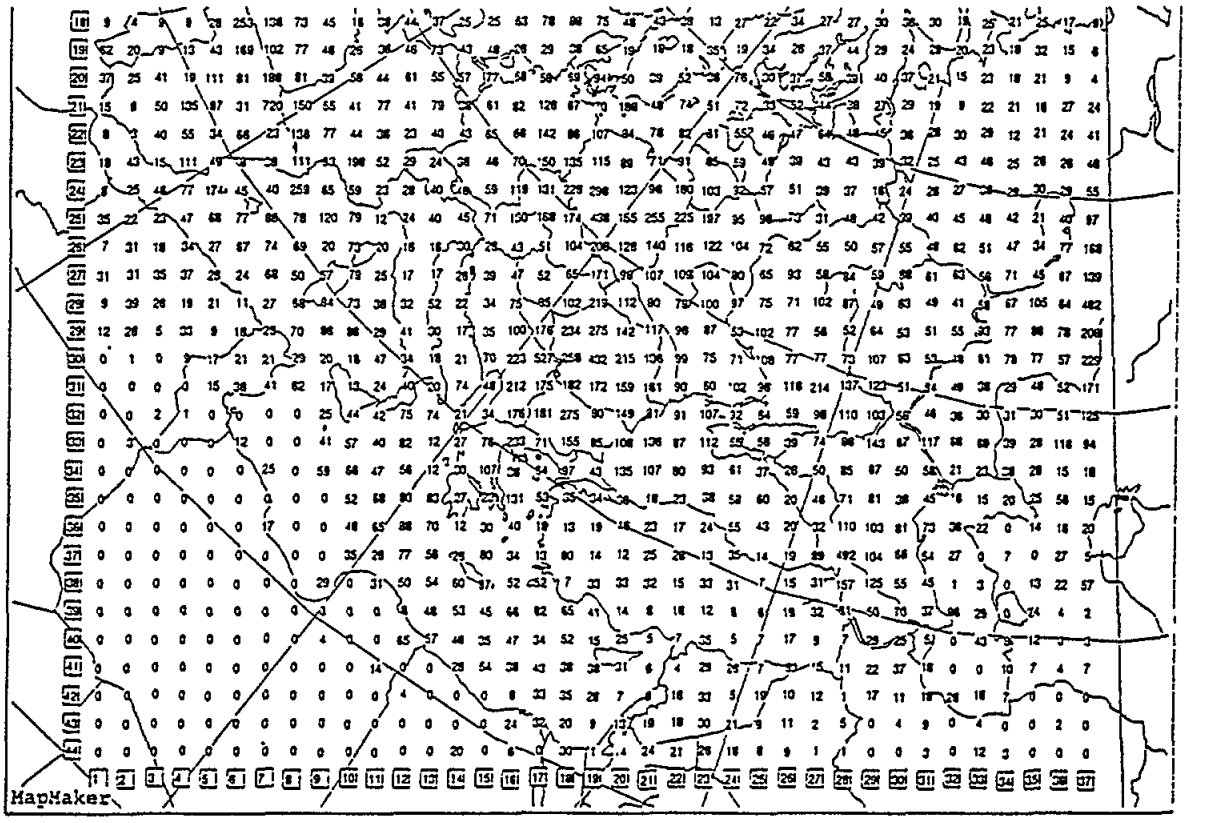


Fig.A14 Concentration of cadmium in air in 1991 (0.001 ng Cd/m^3), upper panel.

Concentration of camium in precipitation in 1991 (1 ng Cd/l), lower panel.

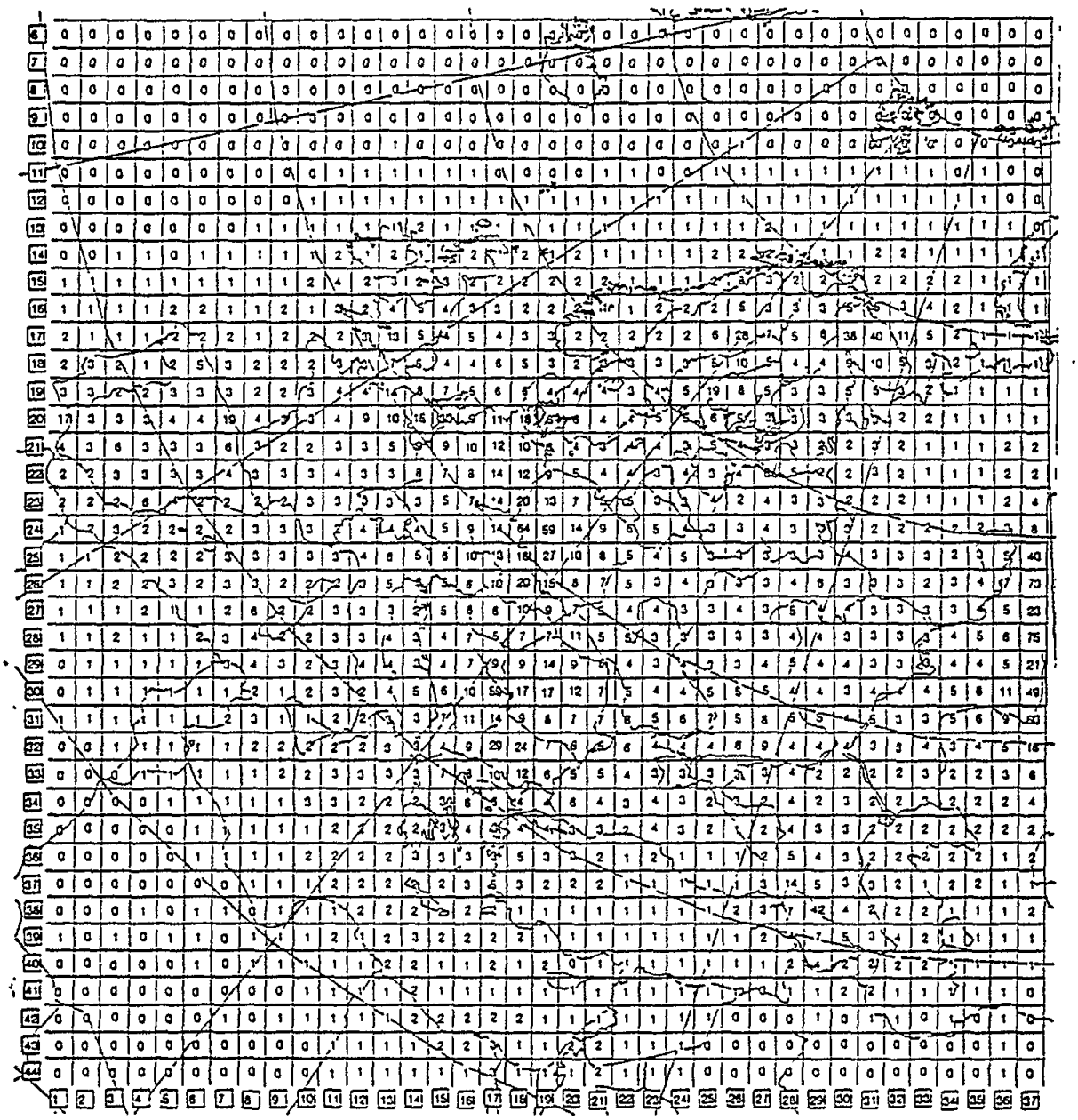
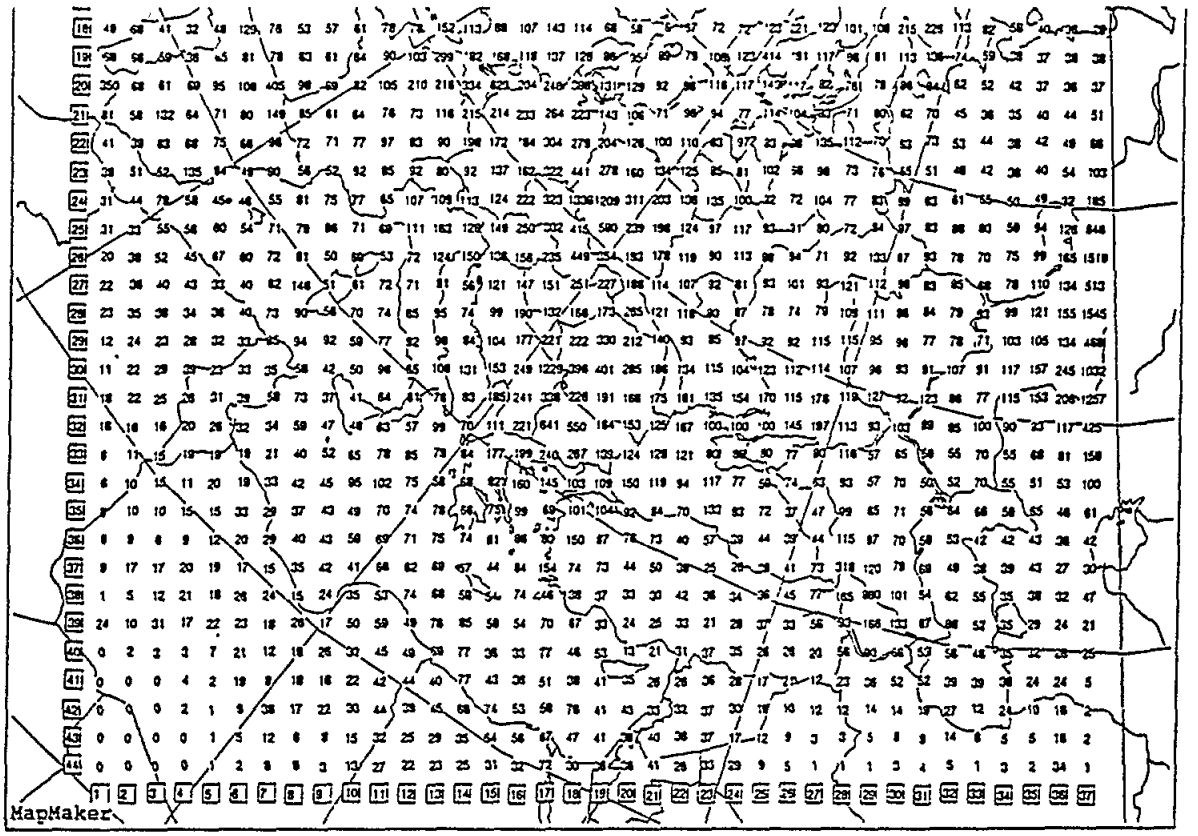


Fig. A15. Dry deposition of arsenic in 1991 (10 ug As/m^{2}year).



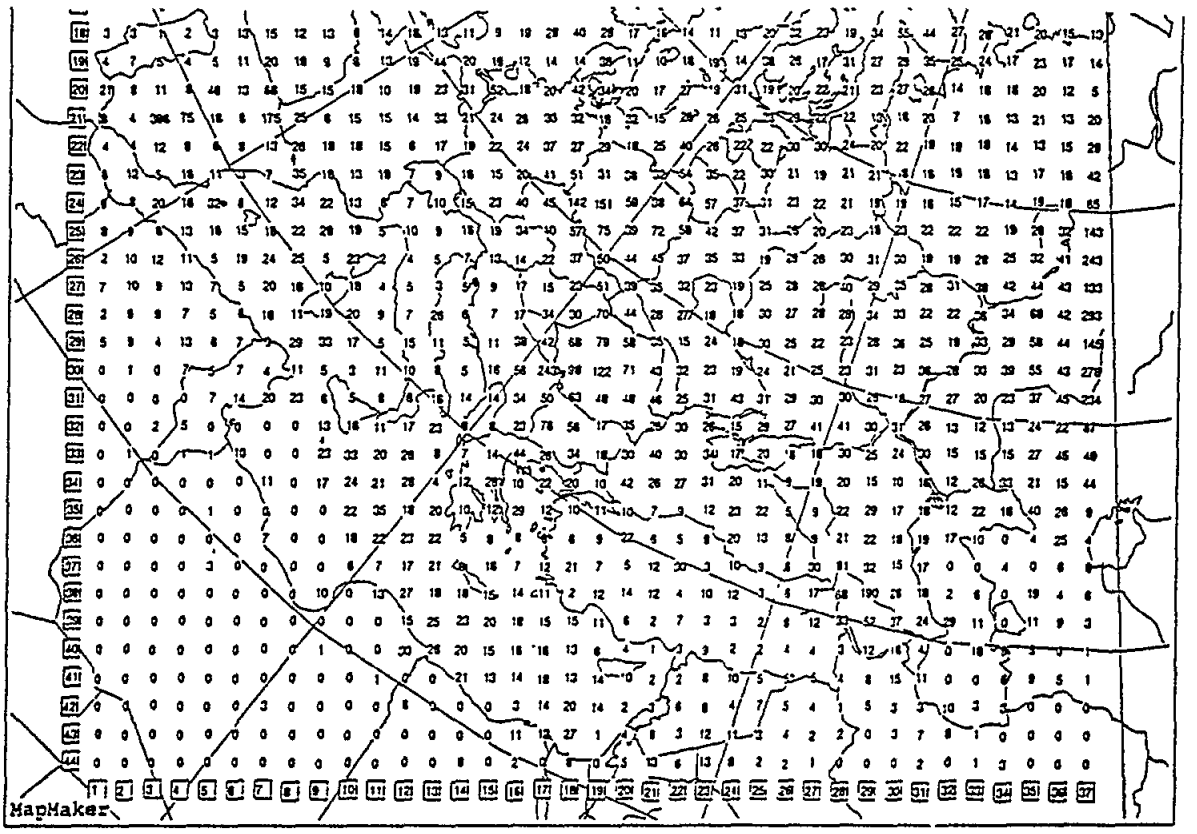


Fig.A16 Concentration of arsenic in air in 1991 (0.01 ng As/m^3), upper panel.

Concentration of arsenic in precipitation in 1991 (10 ng As/1), lower panel.

APPENDIX B.

Sulphur and nitrogen compounds and heavy metal deposition from some countries-emitters in 1991.

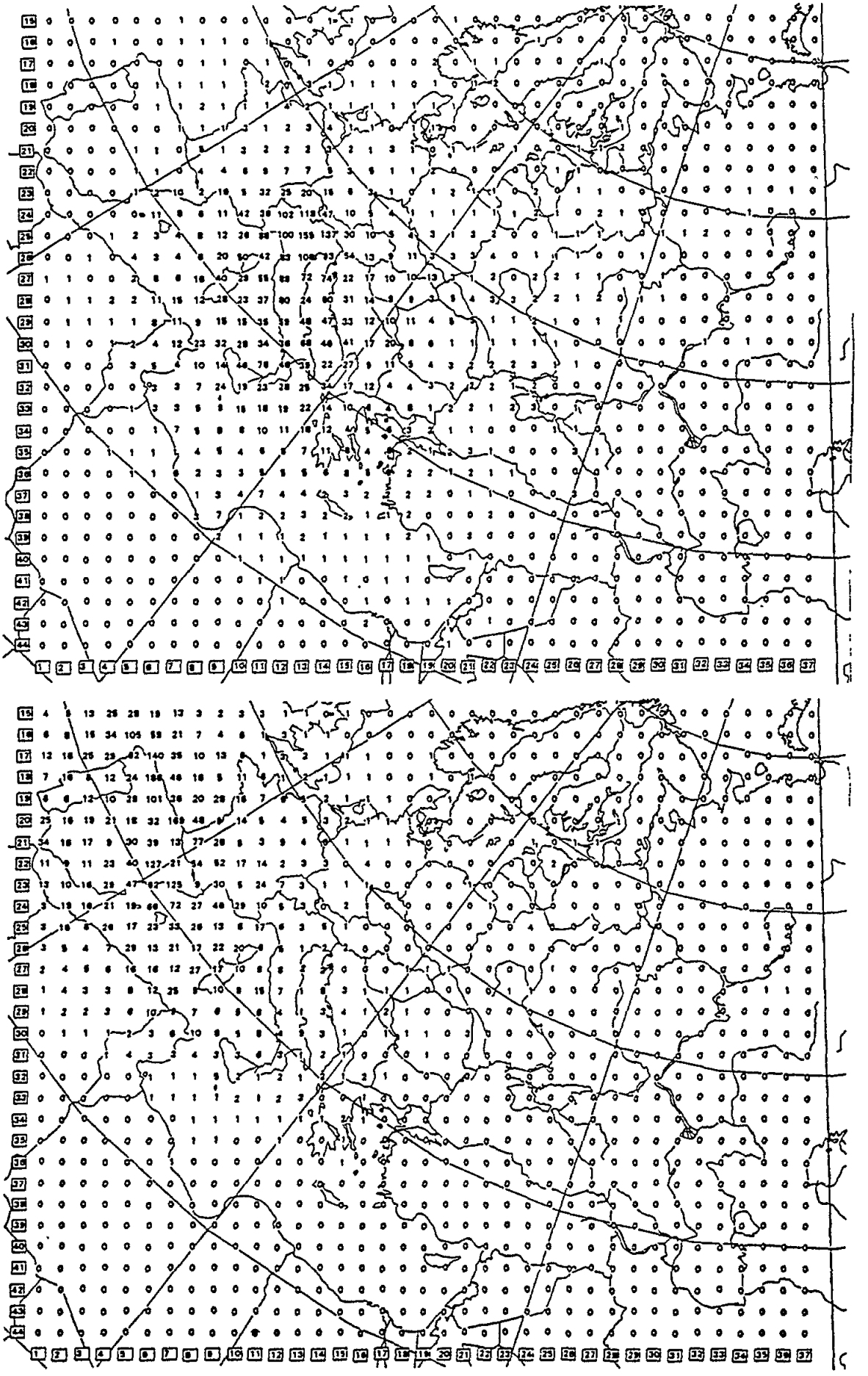


Fig. B1 Total deposition of sulphur compounds from Italy (upper) and from Spain (lower) (10 mg S/m^2 year).

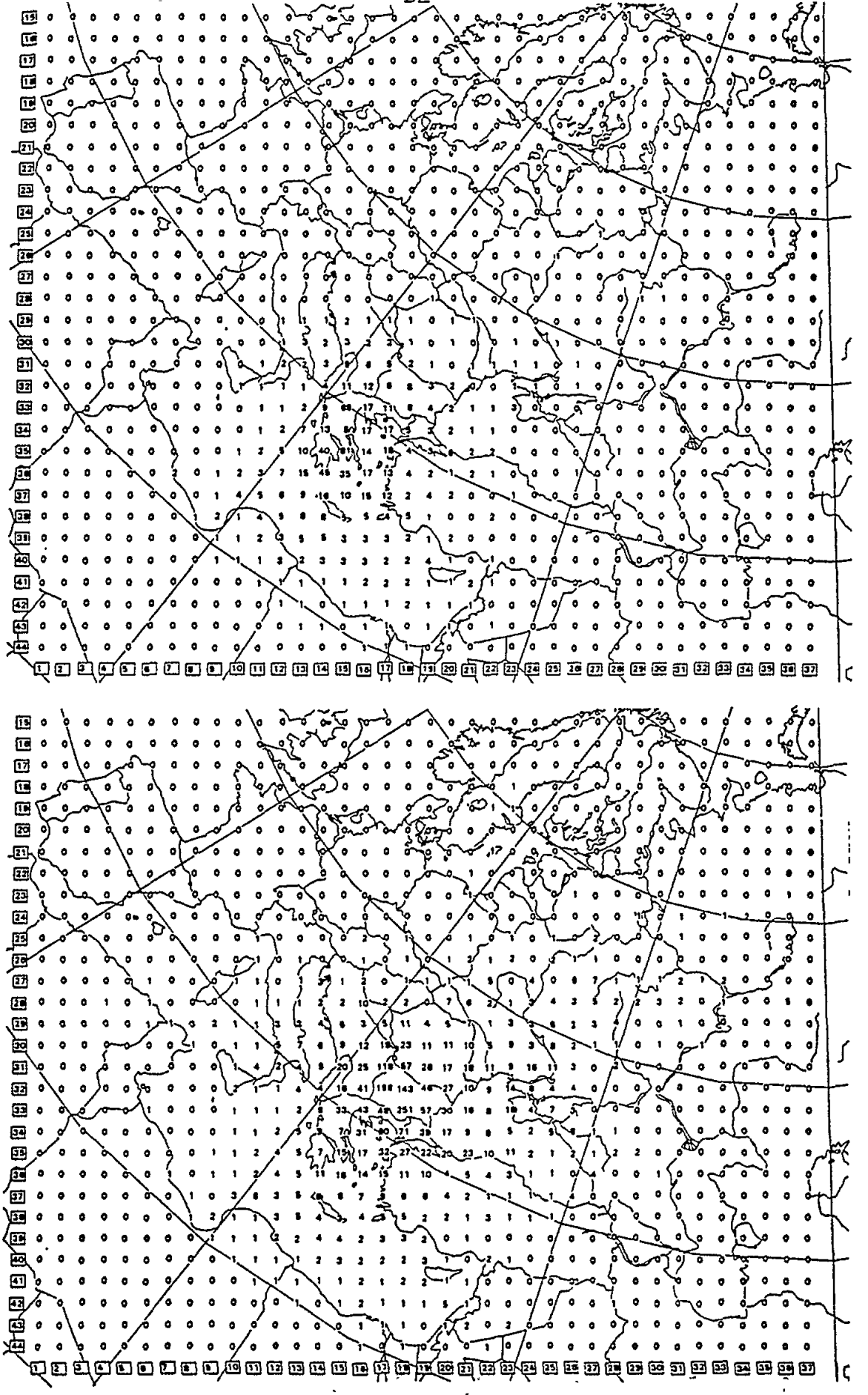


Fig. B2 Total deposition of sulphur compounds from Greece (upper) and Bulgaria (lower) (10 mg S/m^2 year).

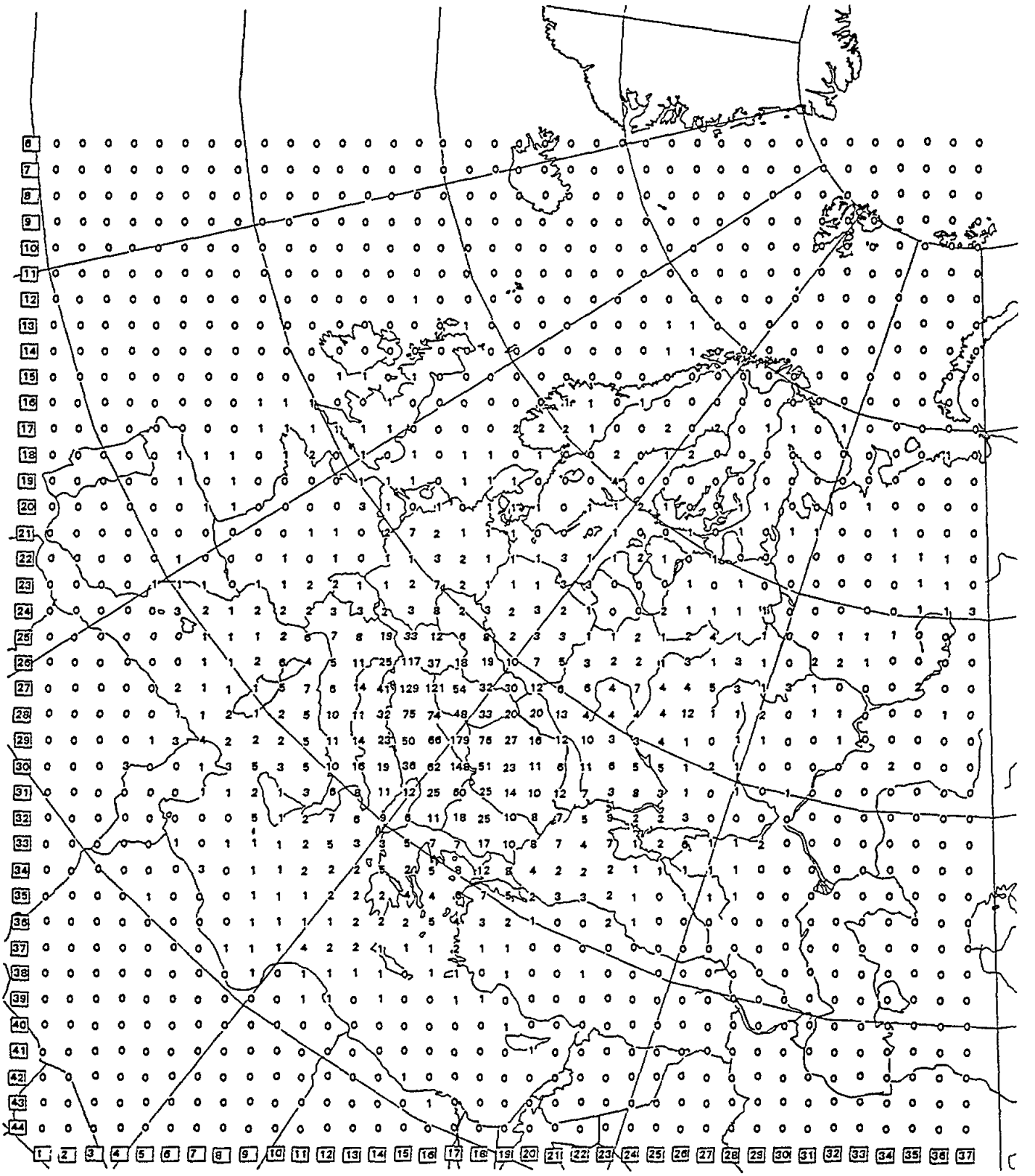


Fig.B3 Total deposition of sulphur compounds from former Yugoslavia (10 mg S/m^2/year).

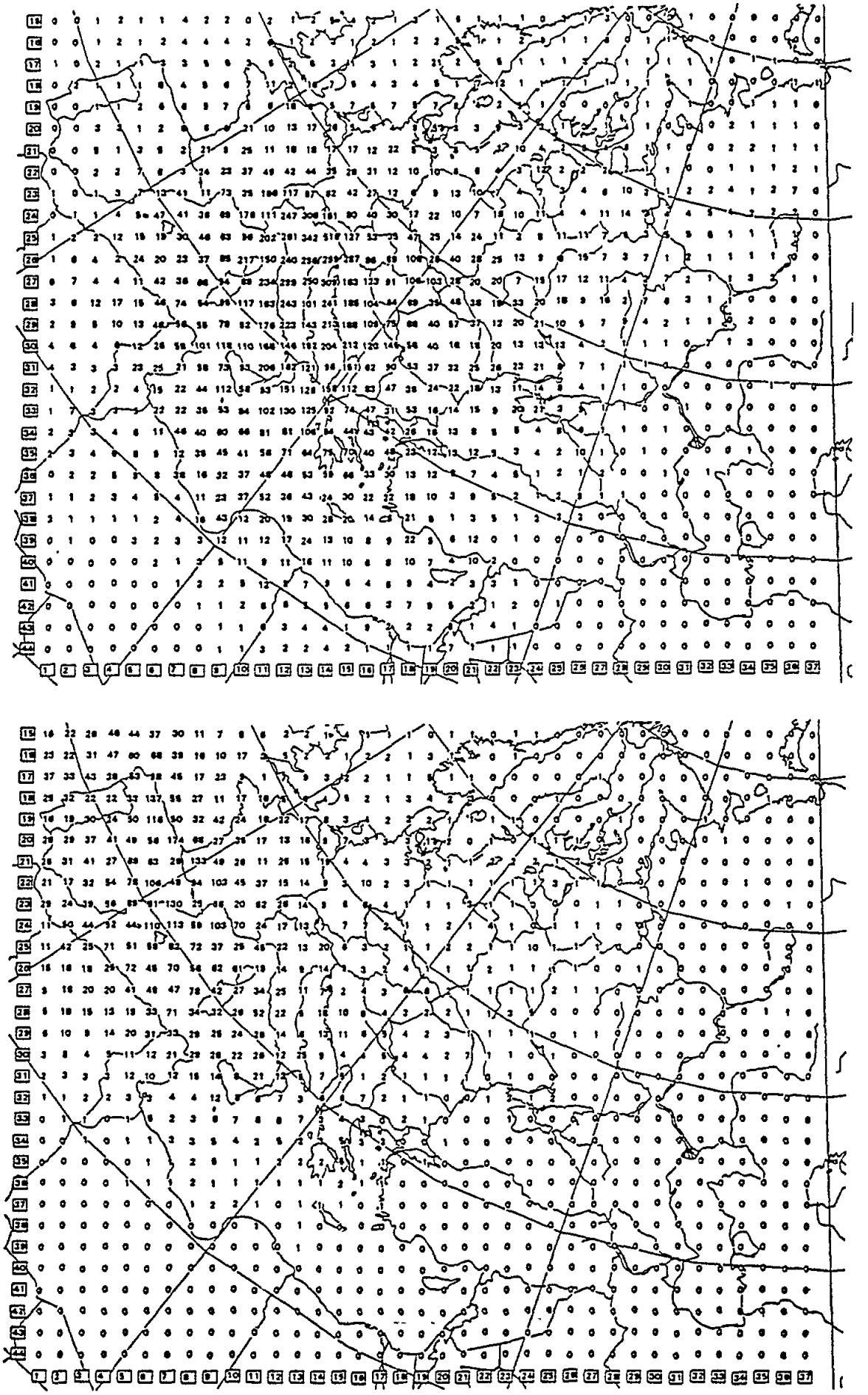


Fig.B4 Total deposition of oxidised nitrogen from Italy (upper) and Spain (lower) (1 mg N/m^2/year).

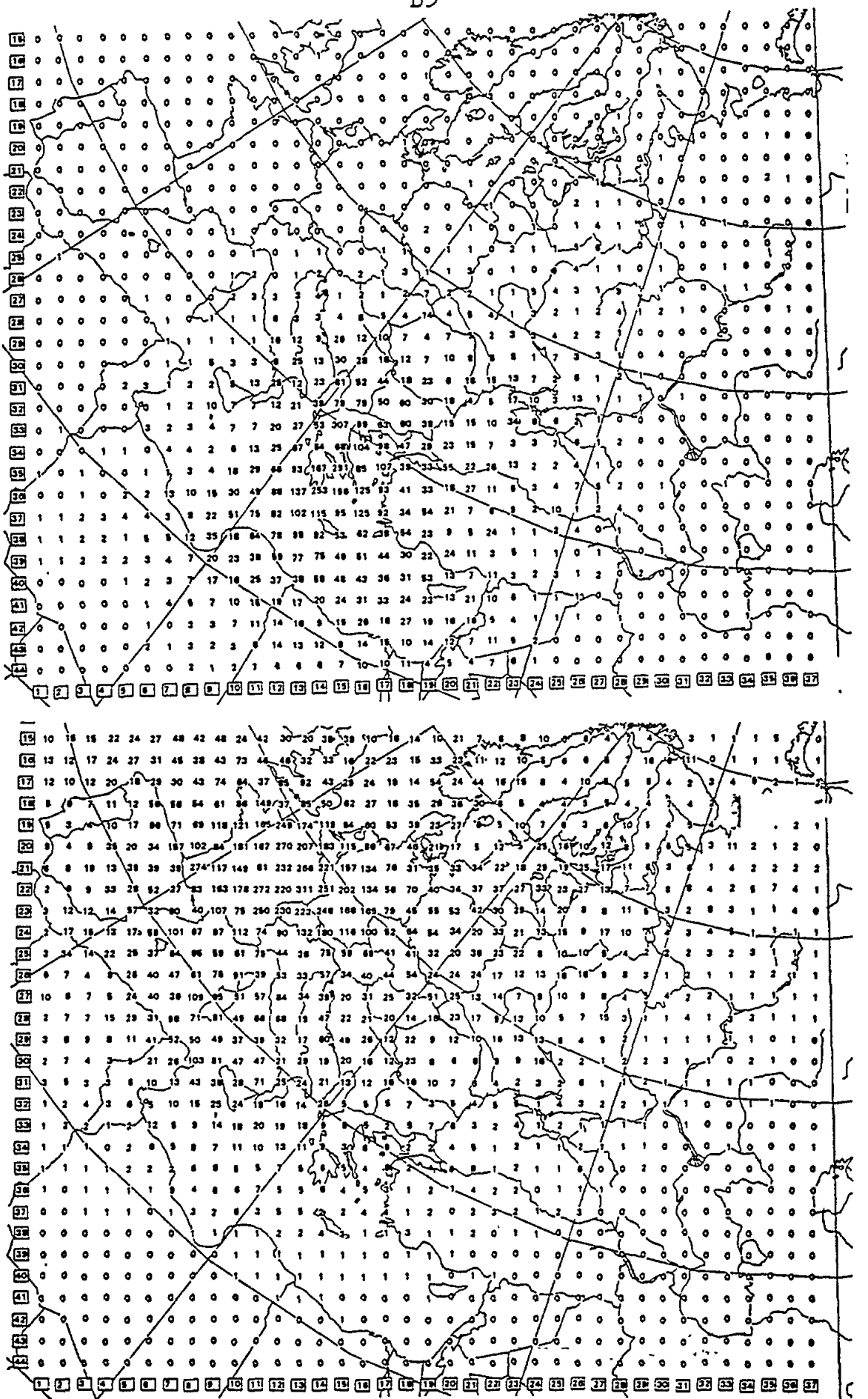


Fig. B5 Total deposition of oxidised nitrogen compounds from Greece (upper) and France (lower) (1 mg $N/m_2/year$).

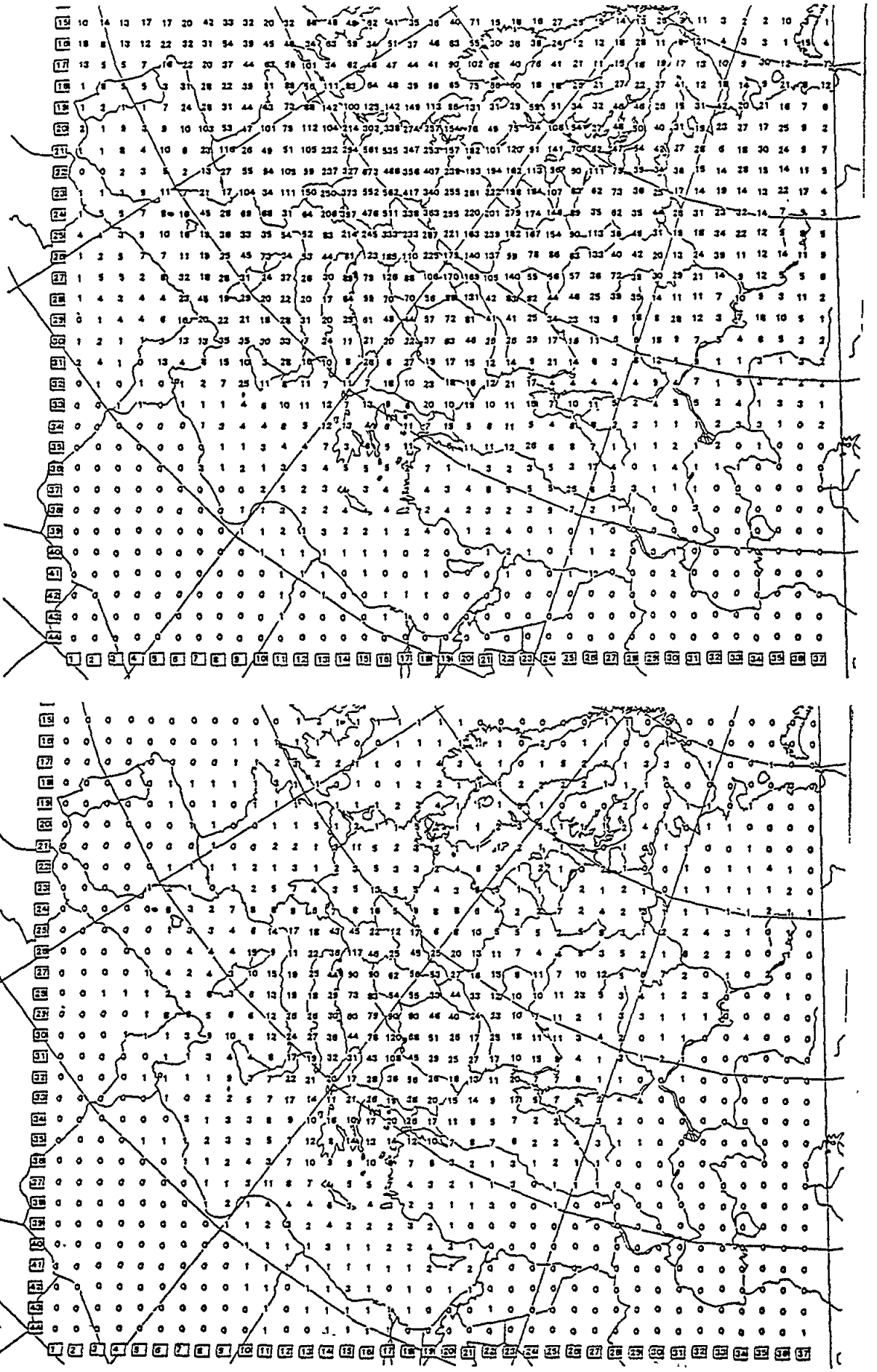


Fig.B6 Total deposition of oxidised nitrogen from Germany (upper) and former Yugoslavia (lower) (1 mg N/m^2/year).

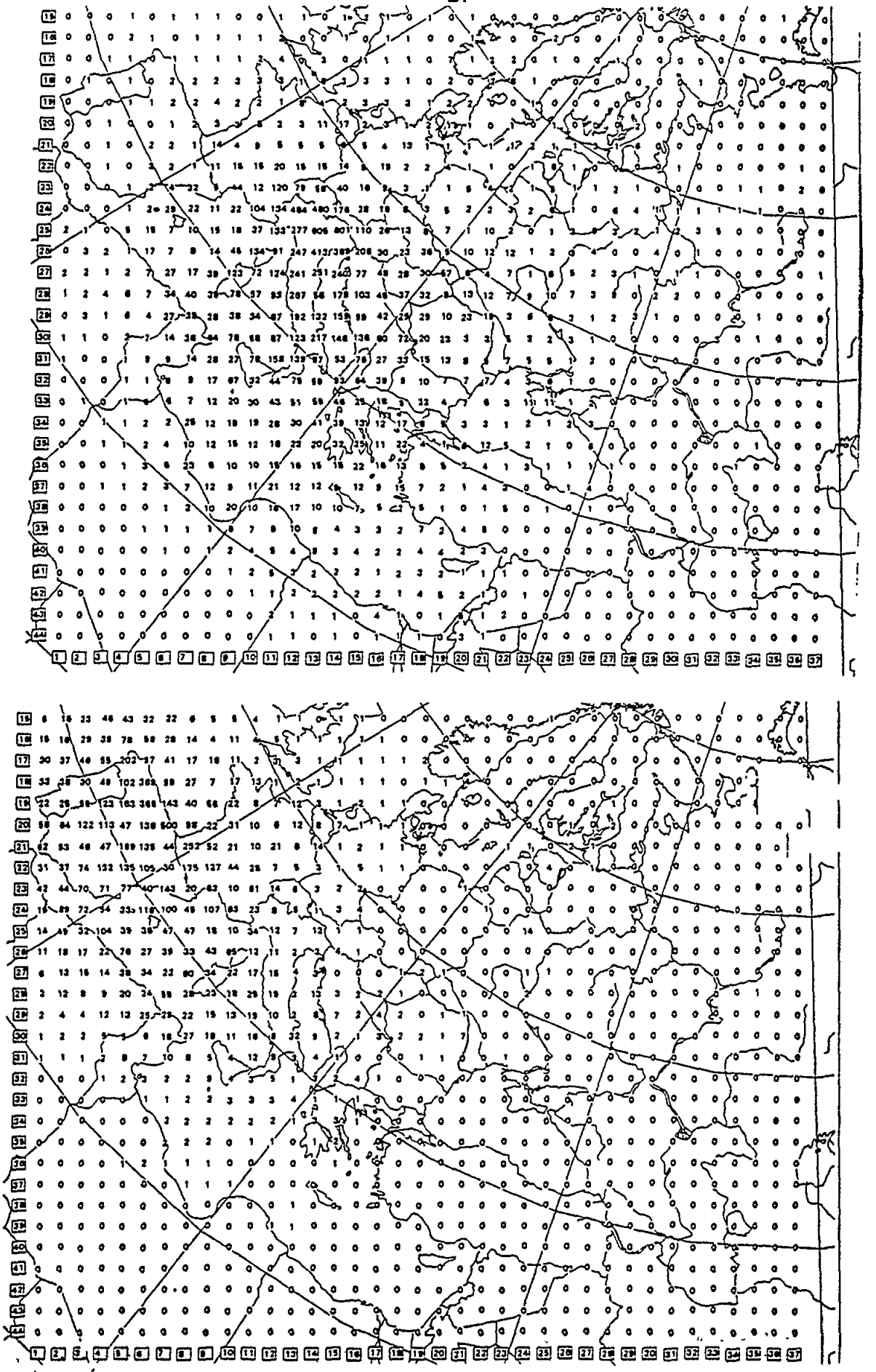


Fig. B7 Total deposition of reduced nitrogen from Italy (upper) and from Spain (lower) (1 mg N/m_2 year).

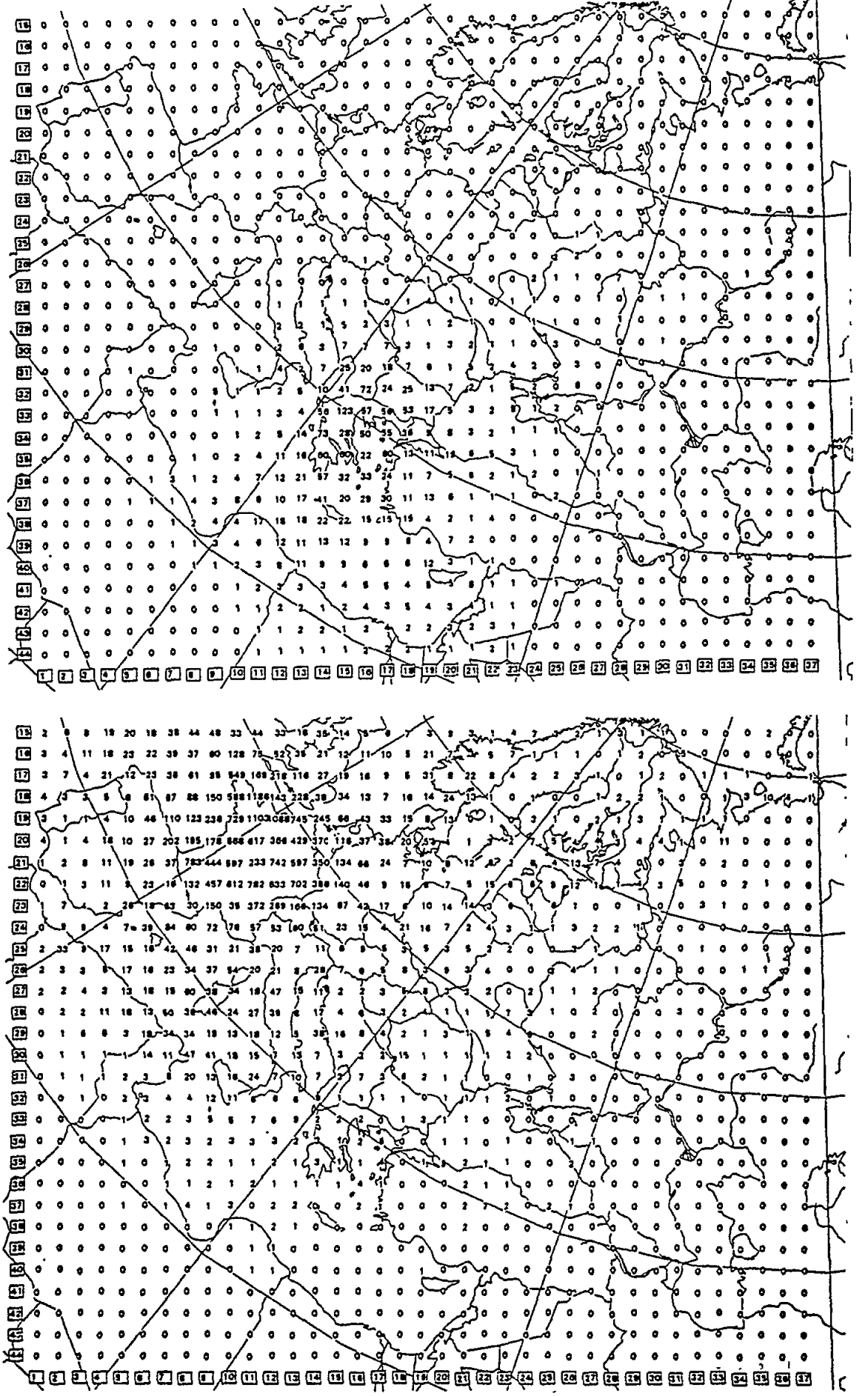


Fig. B8 Total deposition of reduced nitrogen from Greece (upper) and from France (lower) (1 mg N/m^2 year).

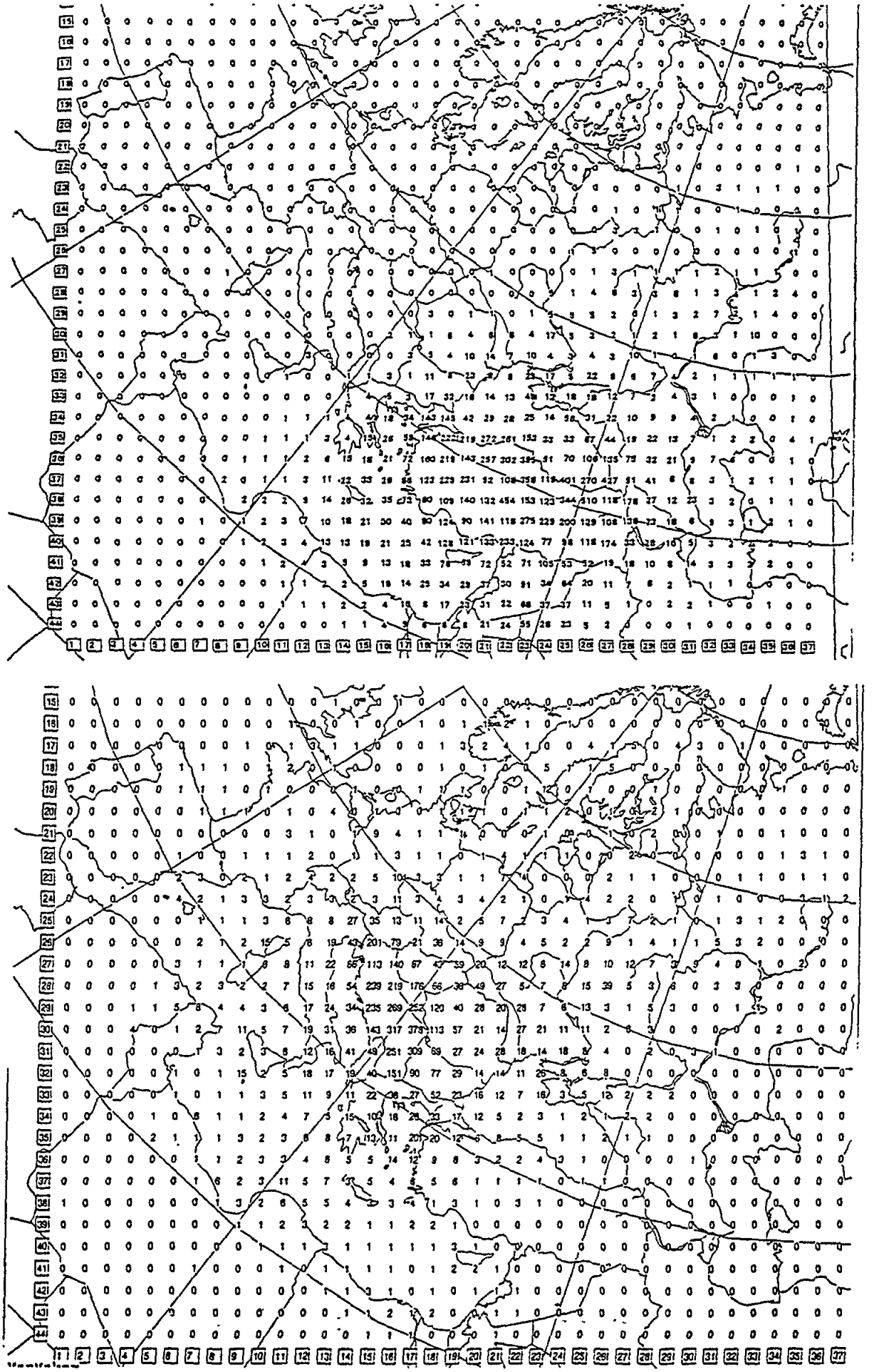


Fig.B9 Total deposition of reduced nitrogen from Turkey (upper) and former Yugoslavia (lower) (1 mg N/m^2/year).

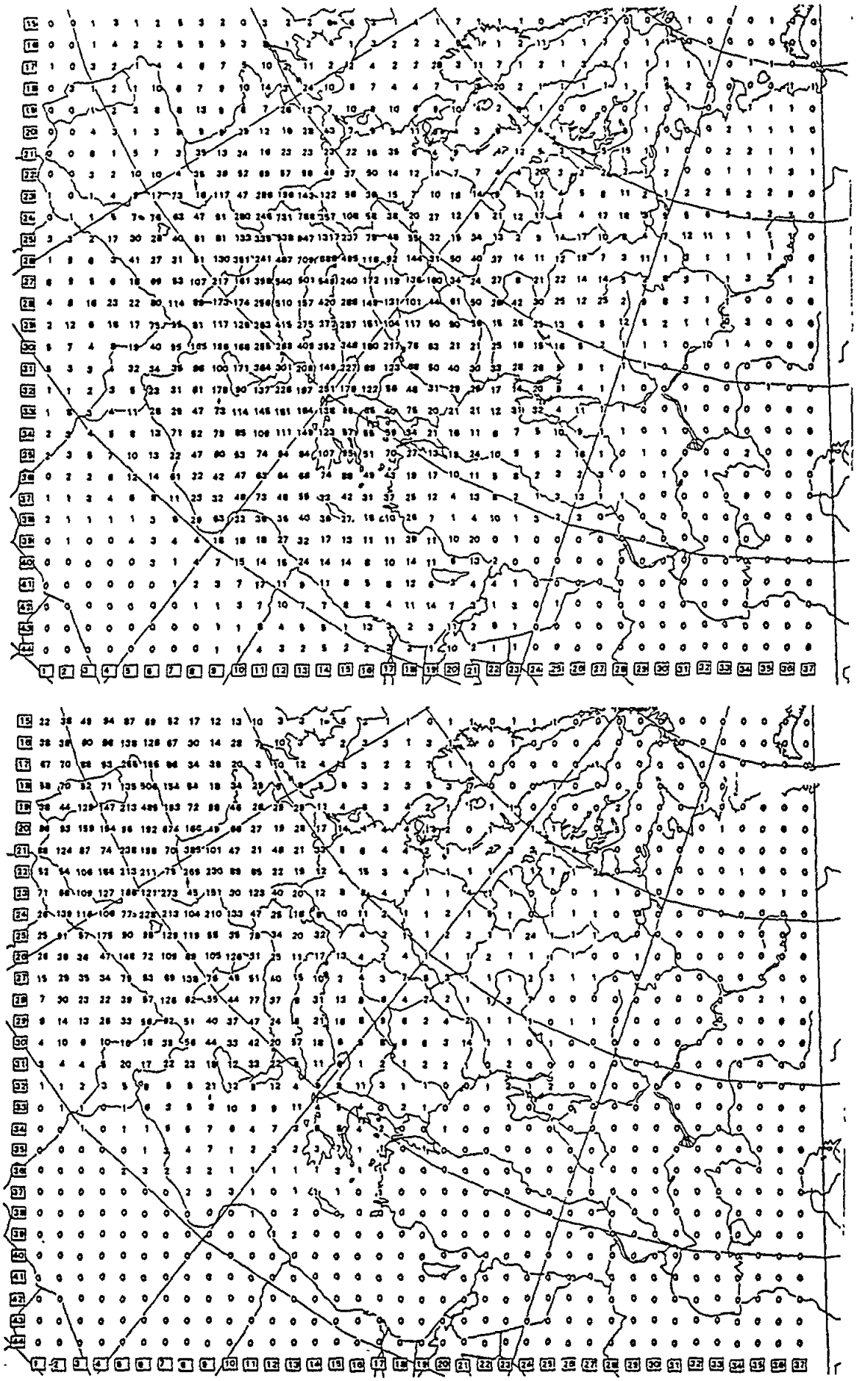


Fig. B10 Total deposition of bound nitrogen from Italy (upper) and from Spain (lower) (1 mg N/m^2 year).

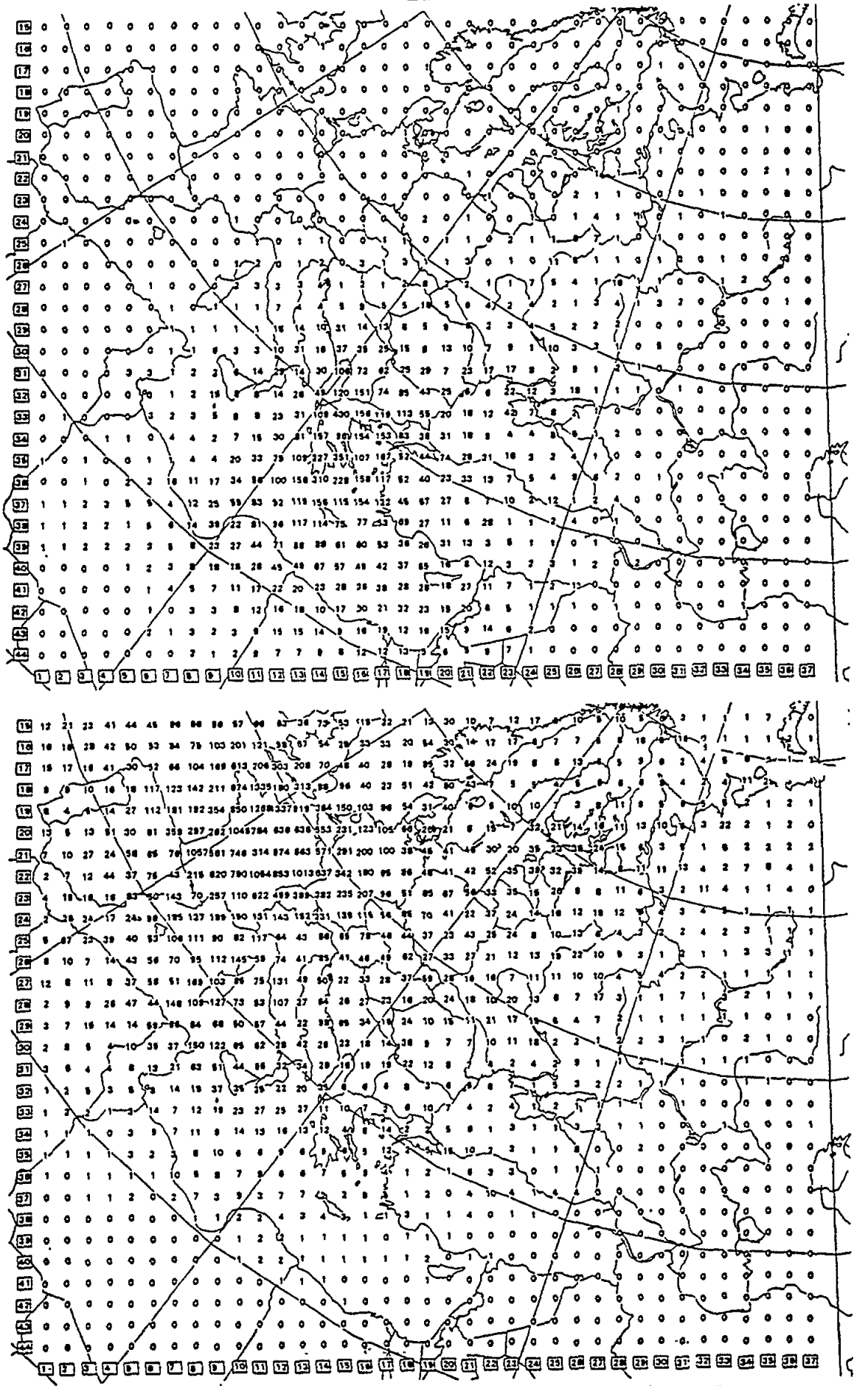


Fig. B11 Total deposition of bound nitrogen from Greece (upper) and from France (lower) (1 mg N/m^2 year).

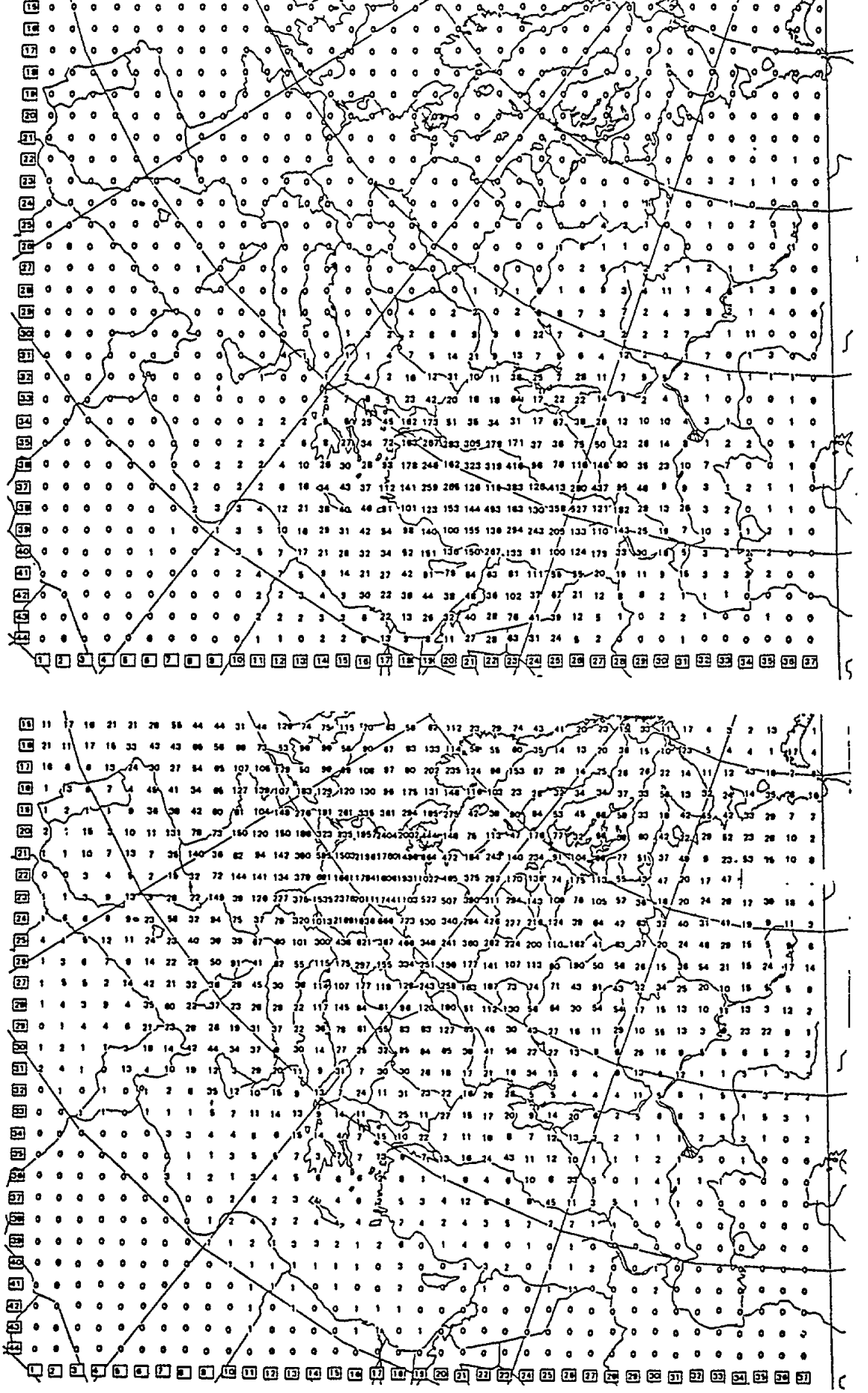


Fig. B12 Total deposition of bound nitrogen from Turkey (upper) and from Germany (lower) (1 mg N/m^2 year).

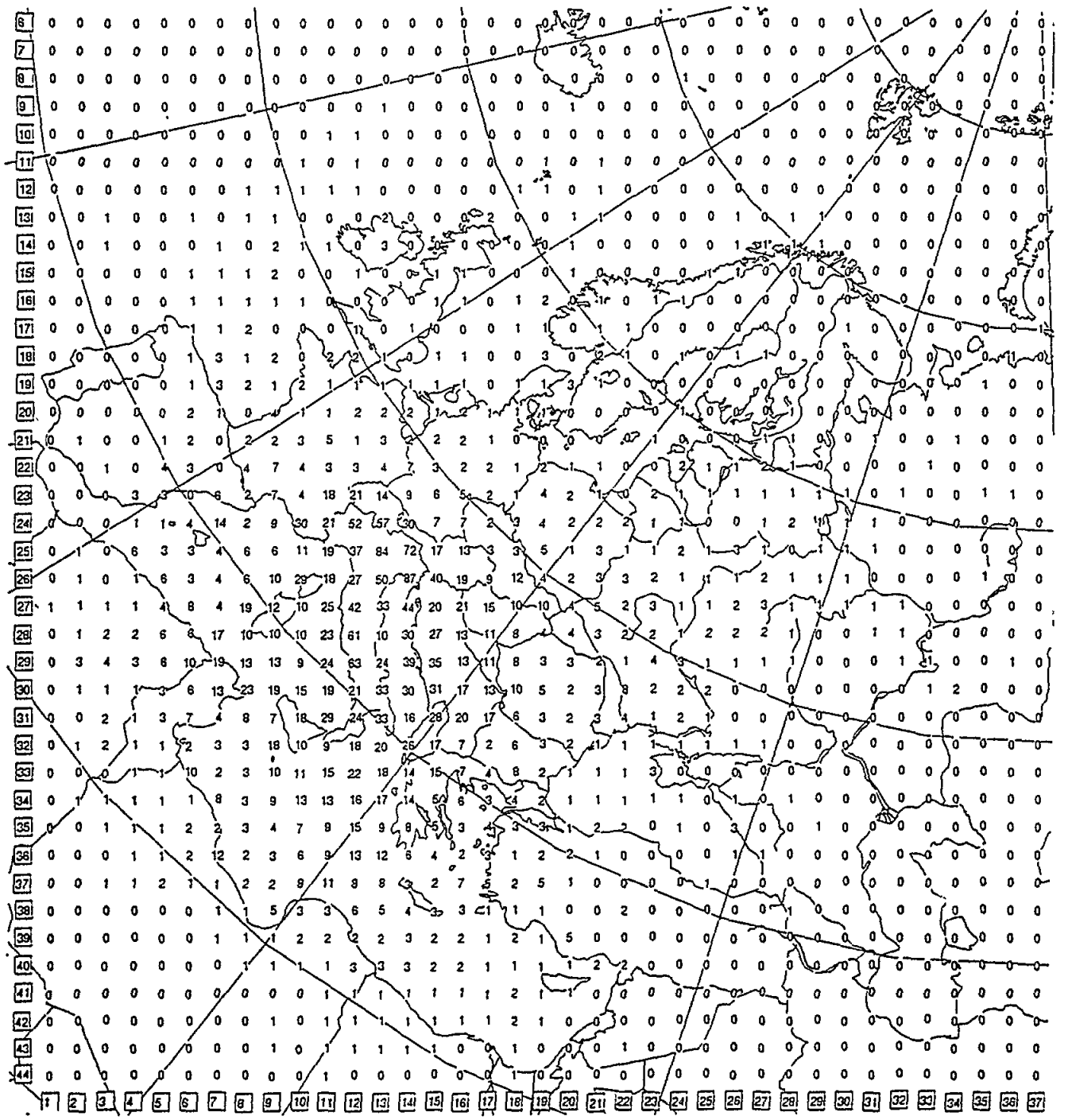


Fig. B13. Total deposition of lead from Italy in 1991 (100 μg Pb/m^2).

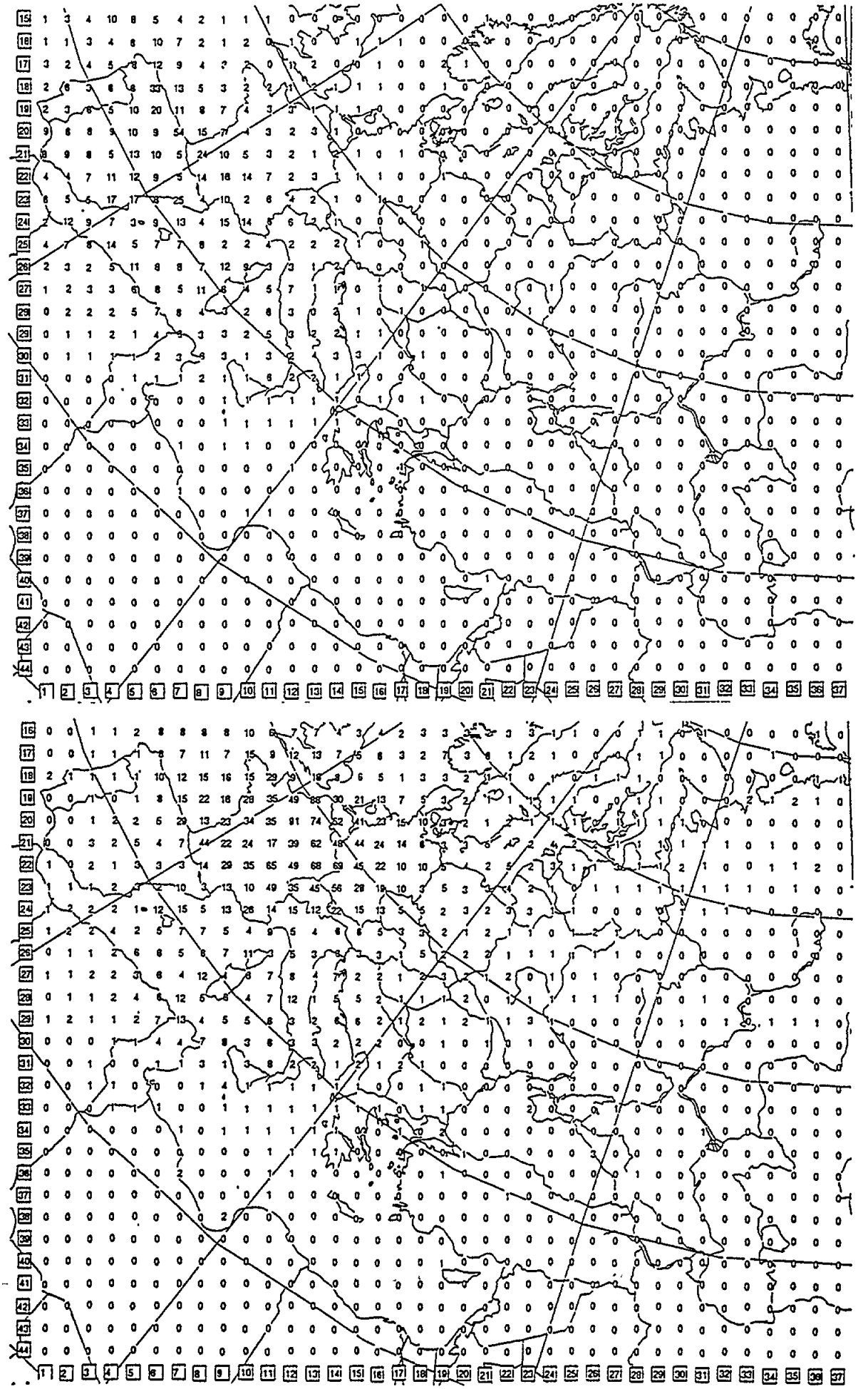


Fig. B14. Total depositions of lead from Spain (upper) and France (lower) in 1991 (100 μg Pb/m^2).

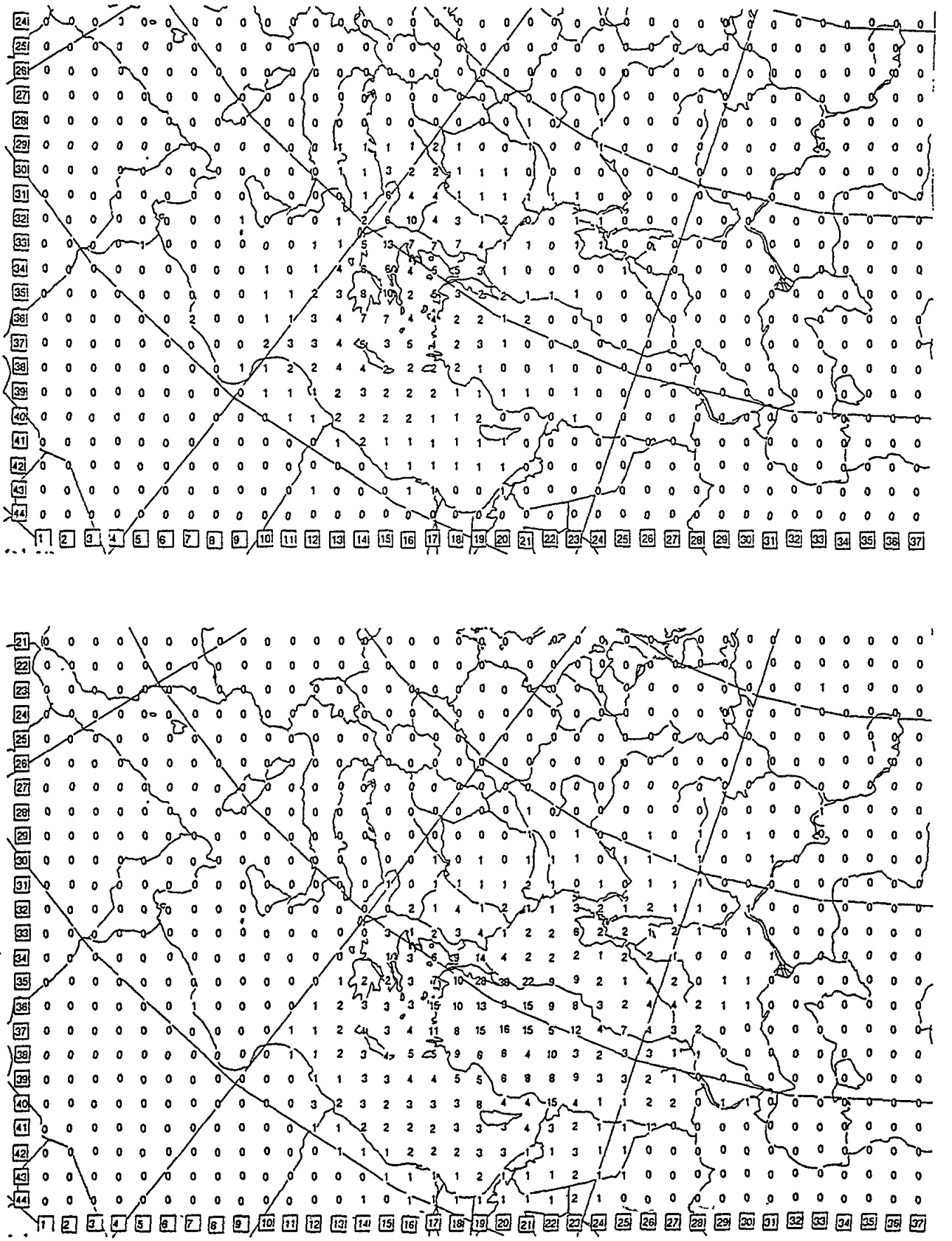


Fig. B15. Total depositions of lead from Greece (upper) and Turkey (lower) in 1991 (100 μg Pb/m^2).

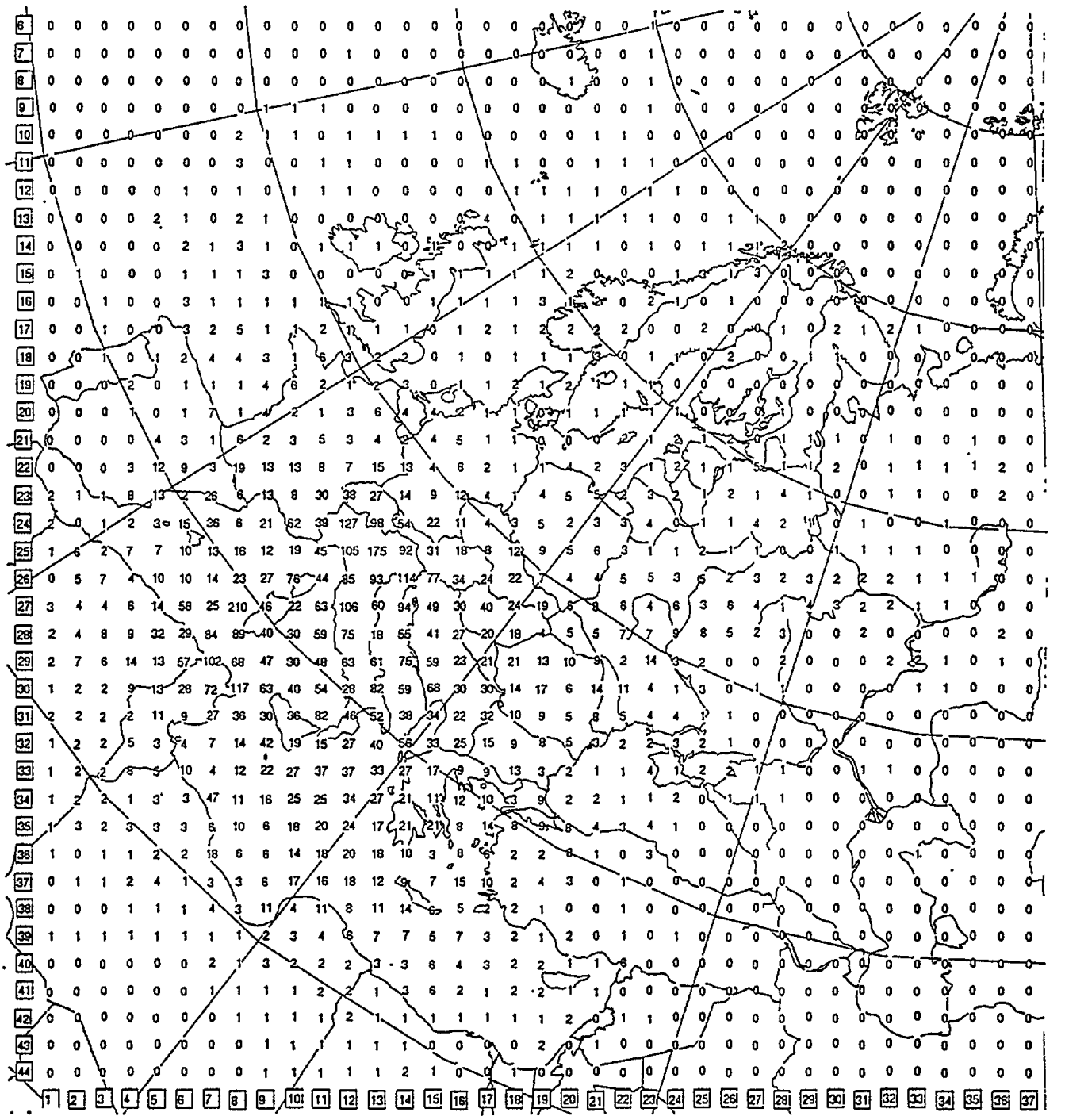


Fig. B16. Total deposition of zinc from Italy in 1991 (10 μg Zn/m^2).

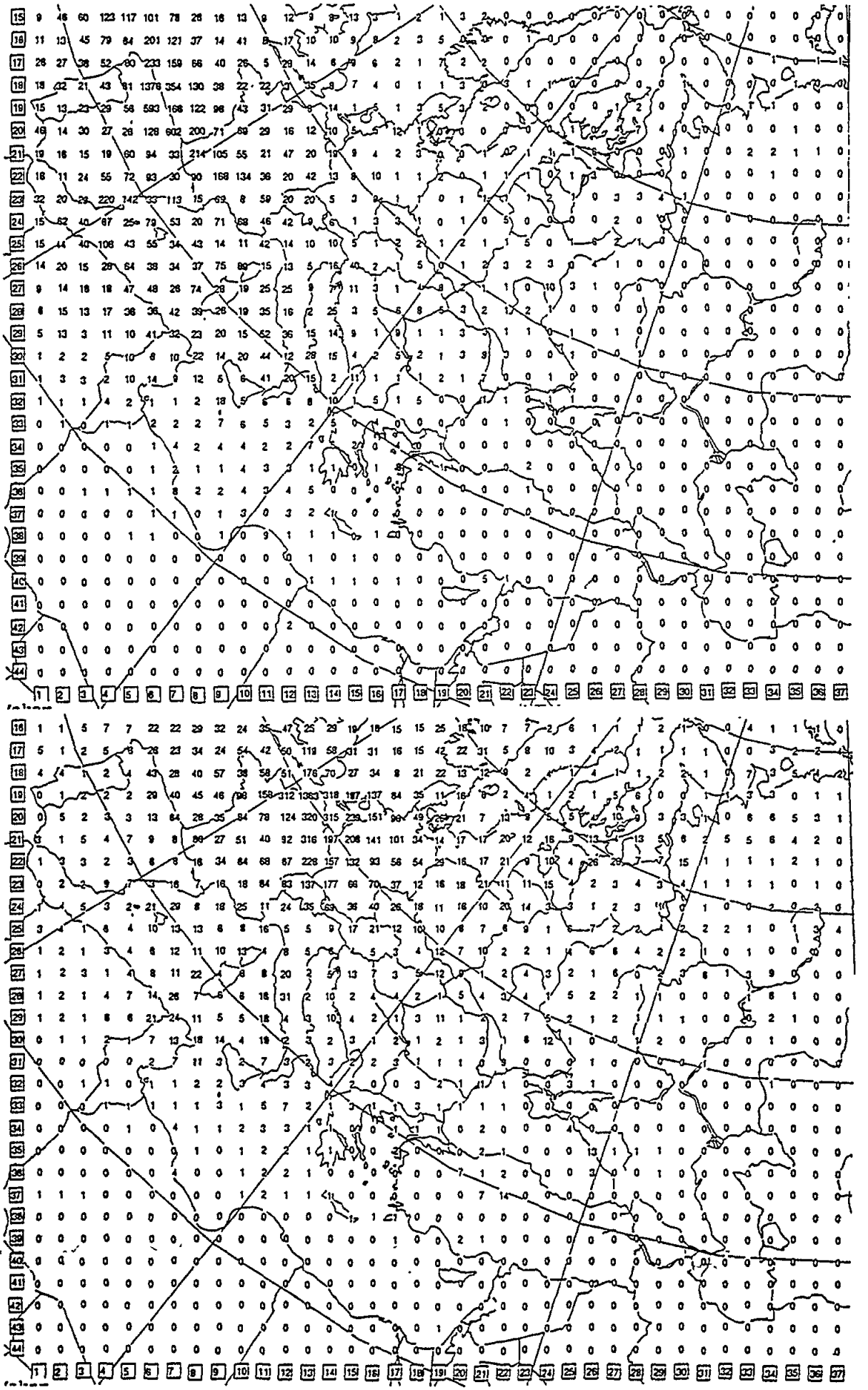


Fig. B17. Total depositions of zinc from Spain (upper) and France (lower) in 1991 (10 μg Zn/m^2).

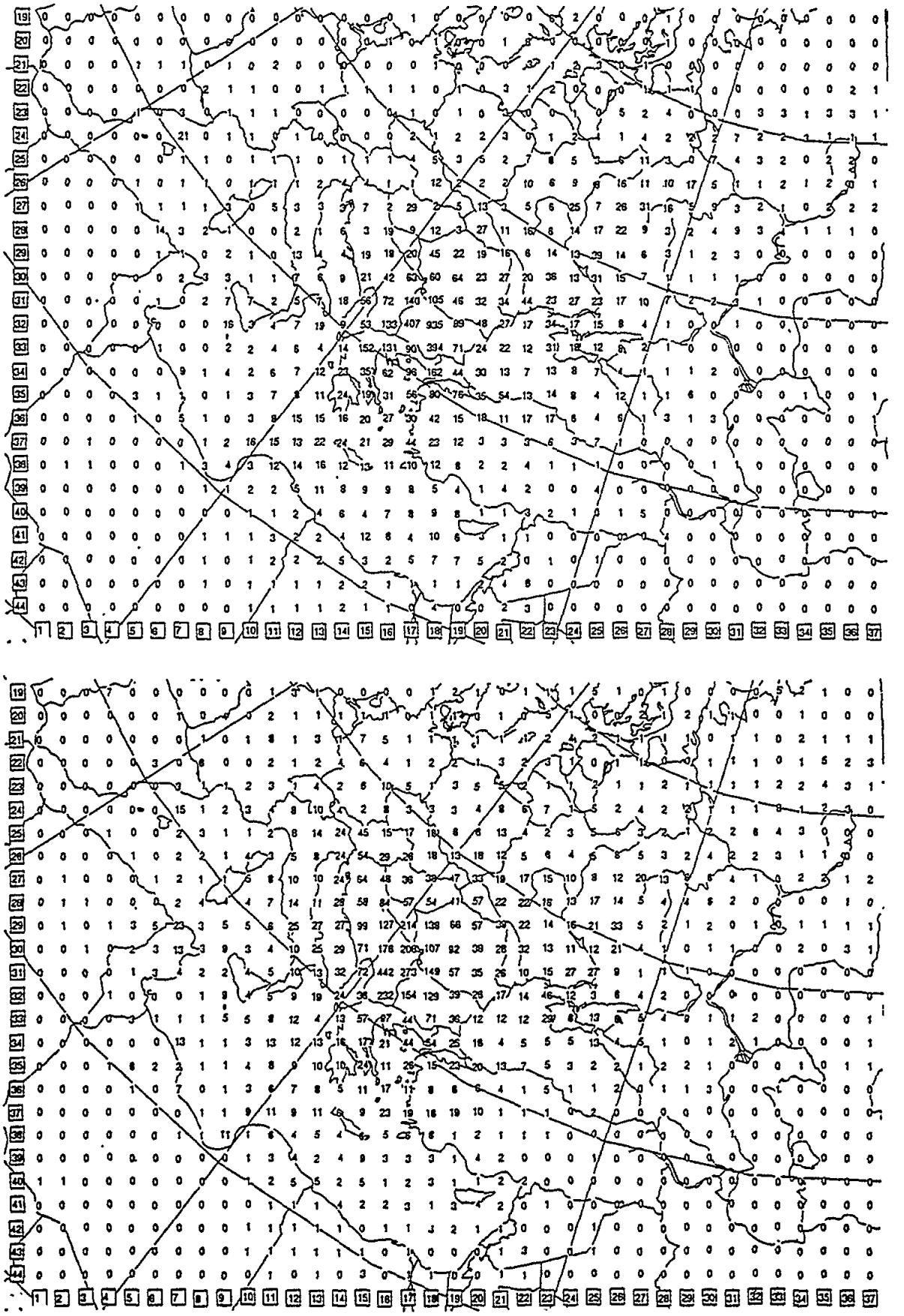


Fig. B18. Total depositions of zinc from Bulgaria (upper) and Yugoslavia (lower) in 1991 (10 μg Zn/m²).

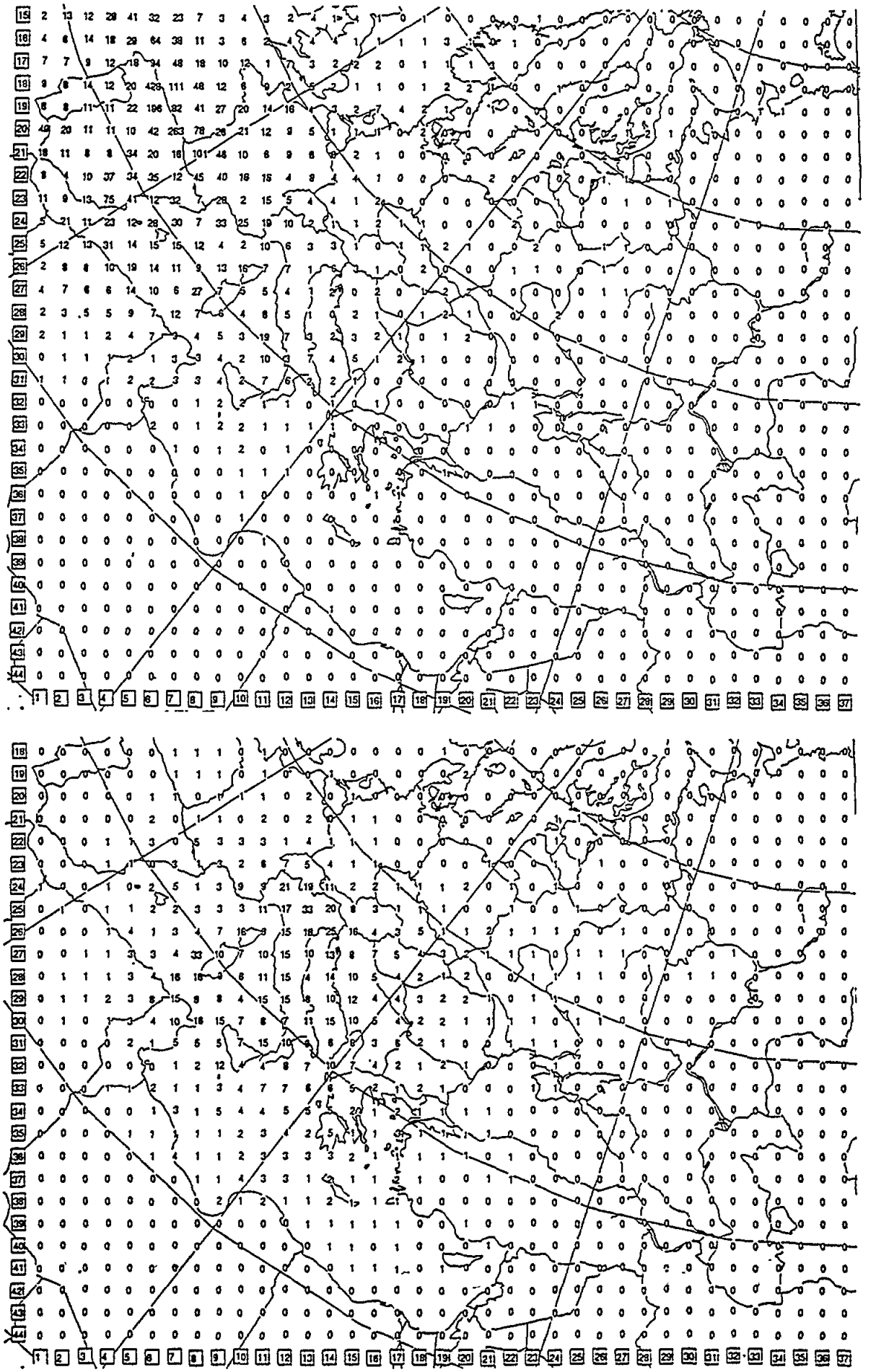


Fig. B19. Total depositions of cadmium from Spain (upper) and Italy (lower) in 1991 (1 μg Cd/m^2).

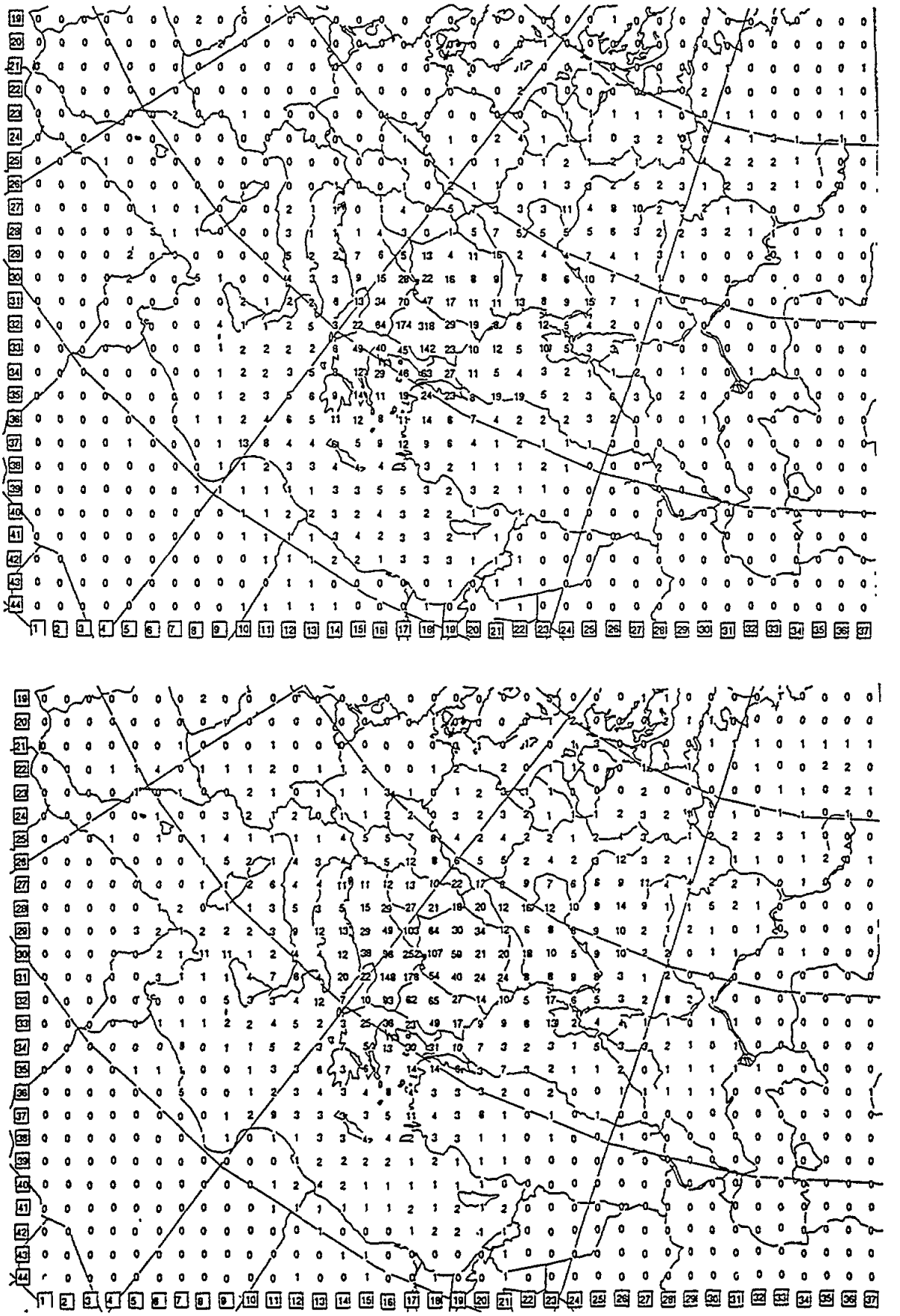


Fig. B20. Total depositions of cadmium from Bulgaria (upper) and Yugoslavia (lower) in 1991 (1 μg Cd/m^2).

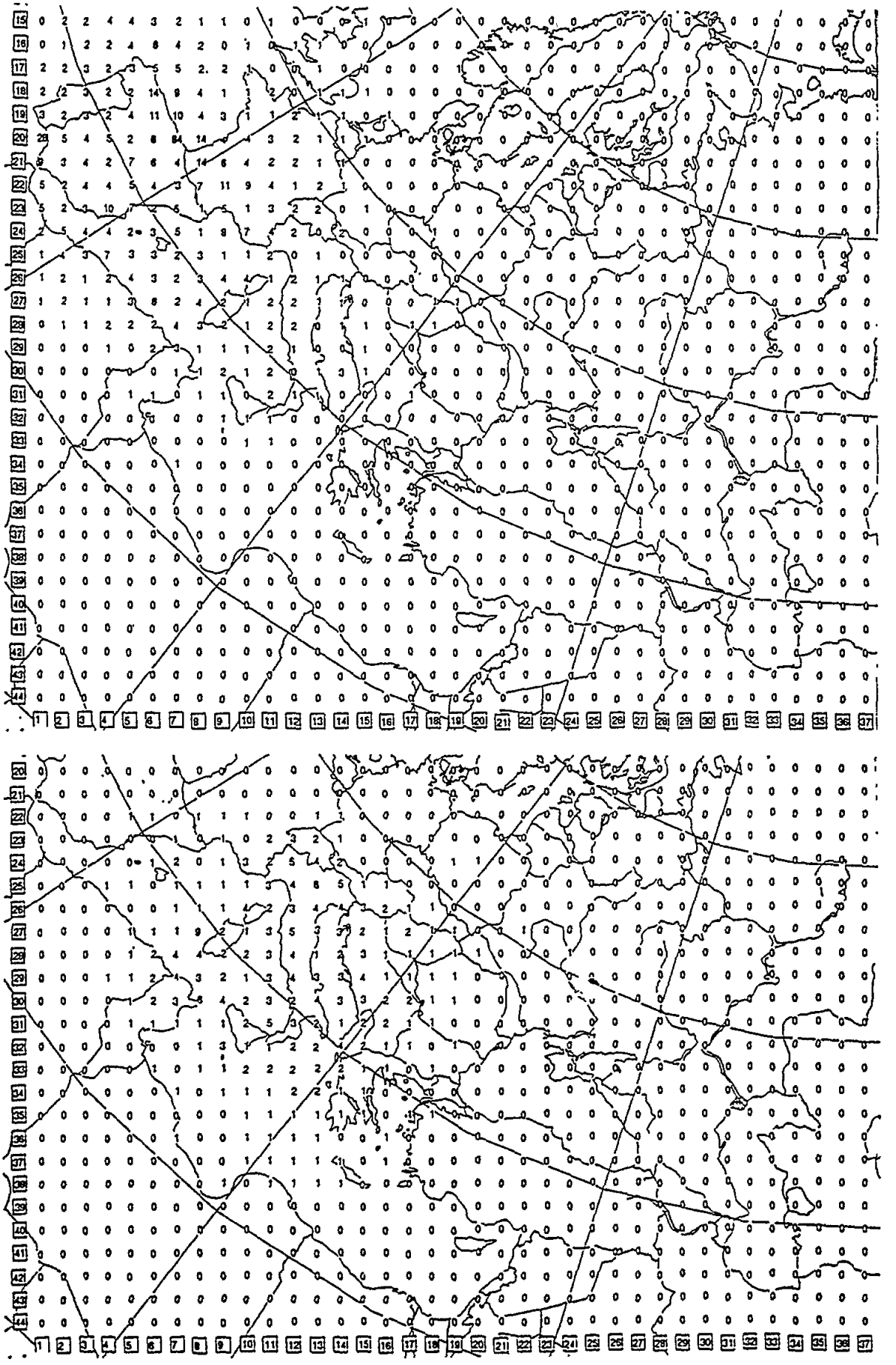


Fig. B21. Total depositions of arsenic from Spain (upper) and Italy (lower) in 1991 (10 ug As/m^2).

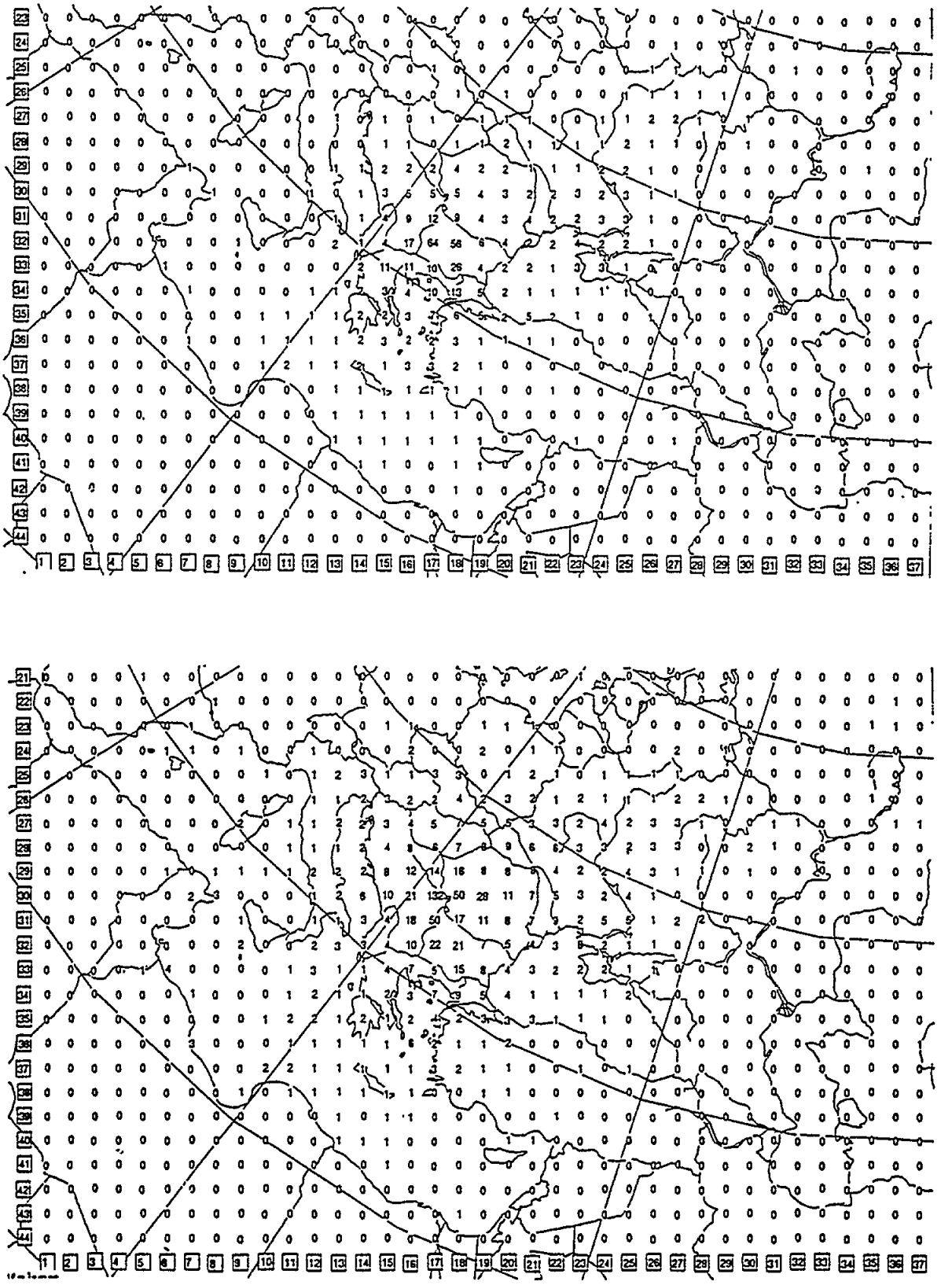


Fig. B22. Total depositions of arsenic from Bulgaria (upper) and Yugoslavia (lower) in 1991 (10 ug As/m^2).

APPENDIX C.

Maps of precipitations; total deposition and mean concentrations in air of sulphur and nitrogen compounds and heavy metals, and wet and dry depositions and mean concentrations in precipitation of sulphur and nitrogen compounds for January, April, July and October 1991.

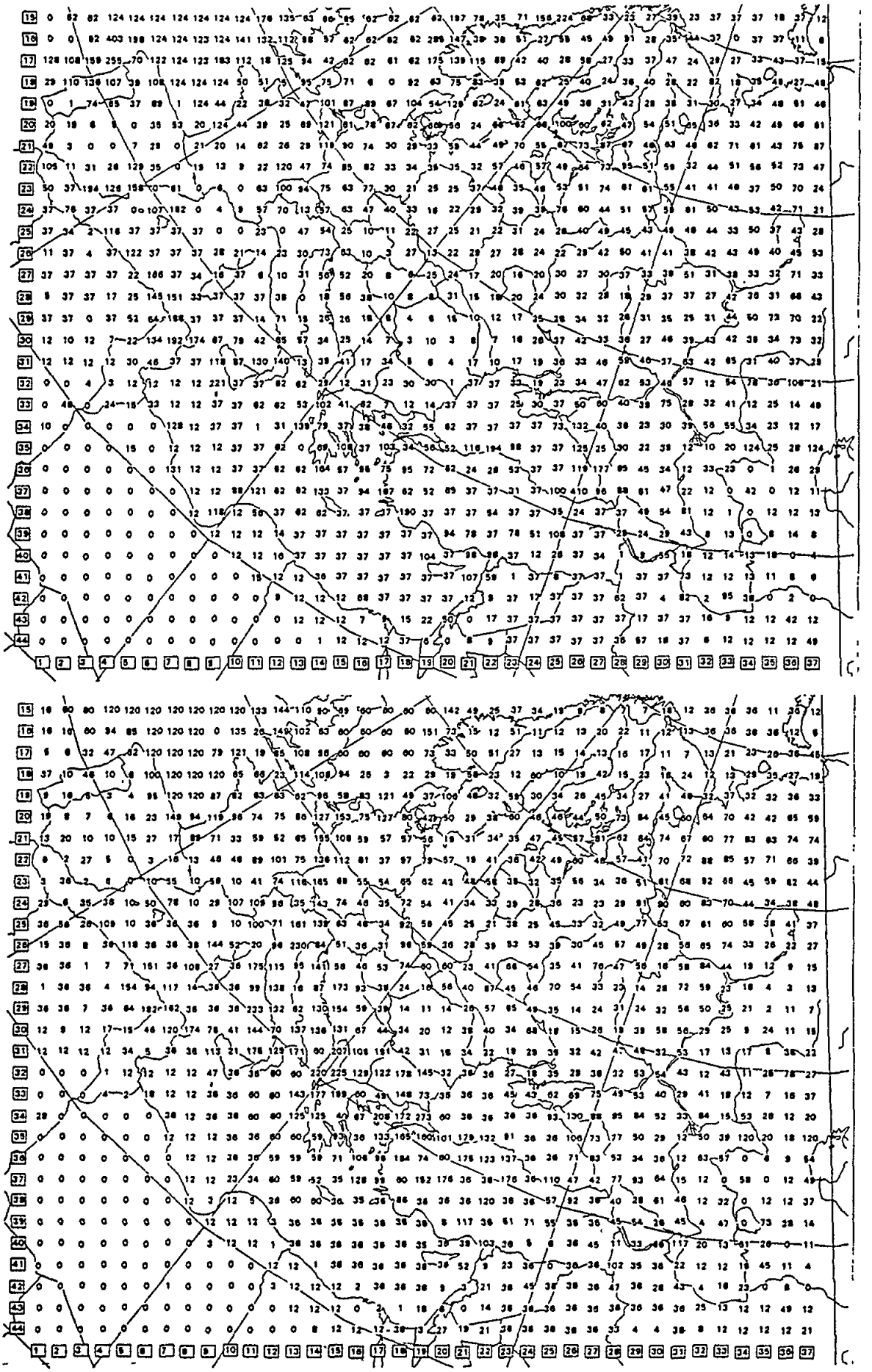


Fig.C1 Precipitation intensity (mm/month)
for January (upper) and April (lower).

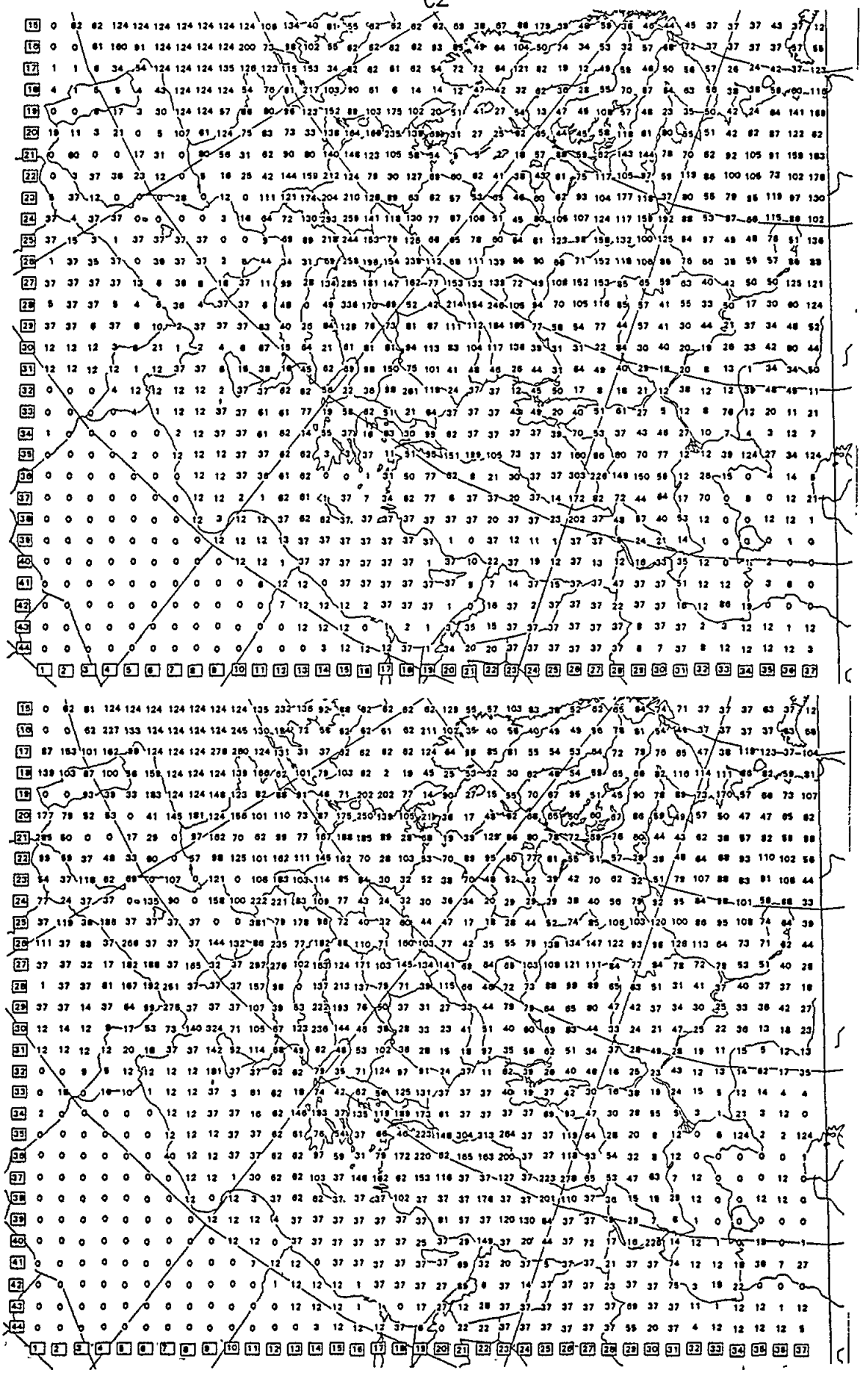


Fig.C2 Precipitation intensity (mm/month) for July (upper) and October (lower).

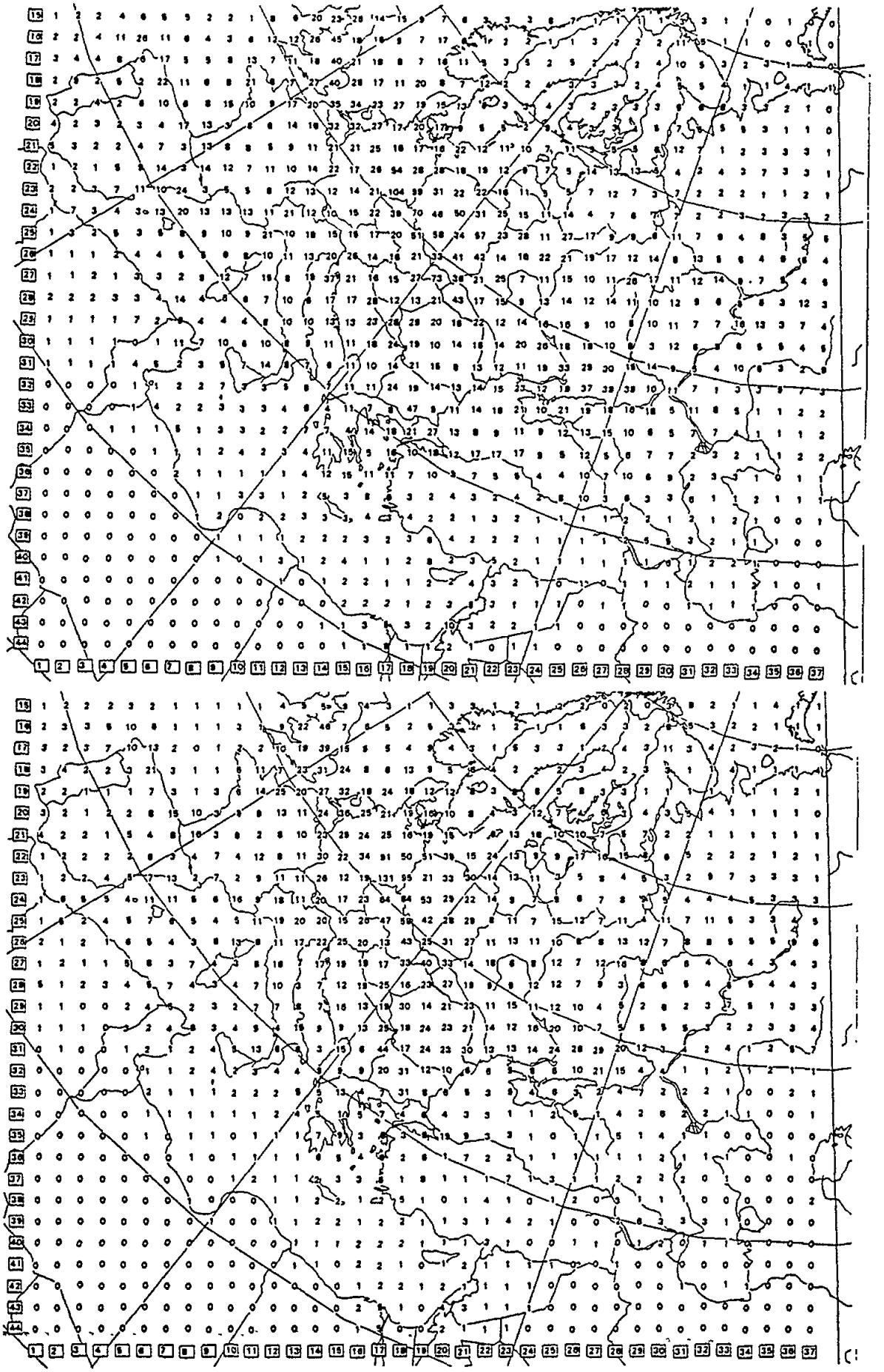


Fig.C3 Total deposition of sulphur compounds (10 mg S/m^2 month) for January (upper) and April (lower).

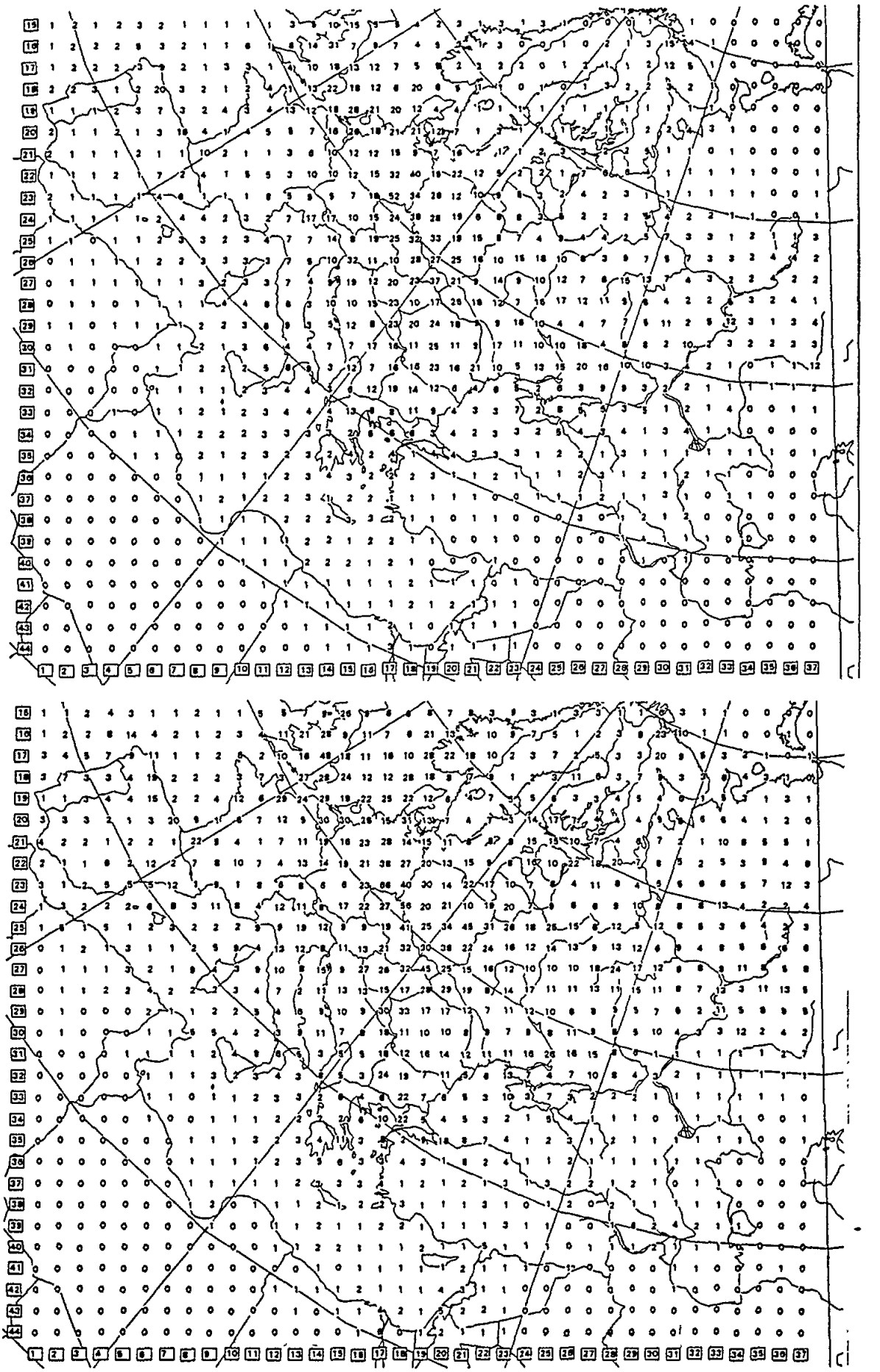


Fig.C4 Total deposition of sulphur compounds (10 mg S/m^2 month) for July (upper) and October (lower).

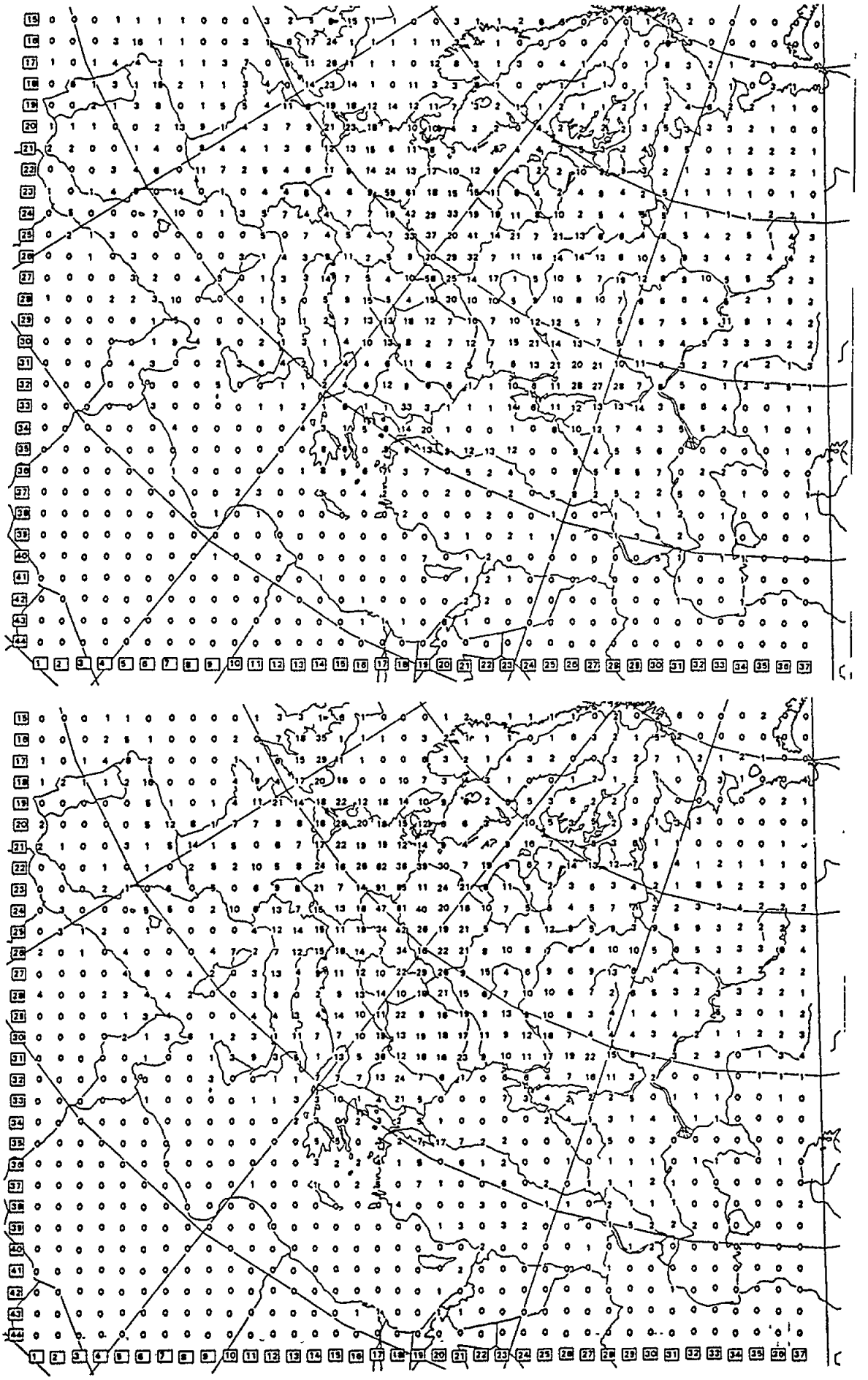


Fig.C5 Wet deposition of sulphur compounds (10 mg S/m^2 month) for January (upper) and April (lower).

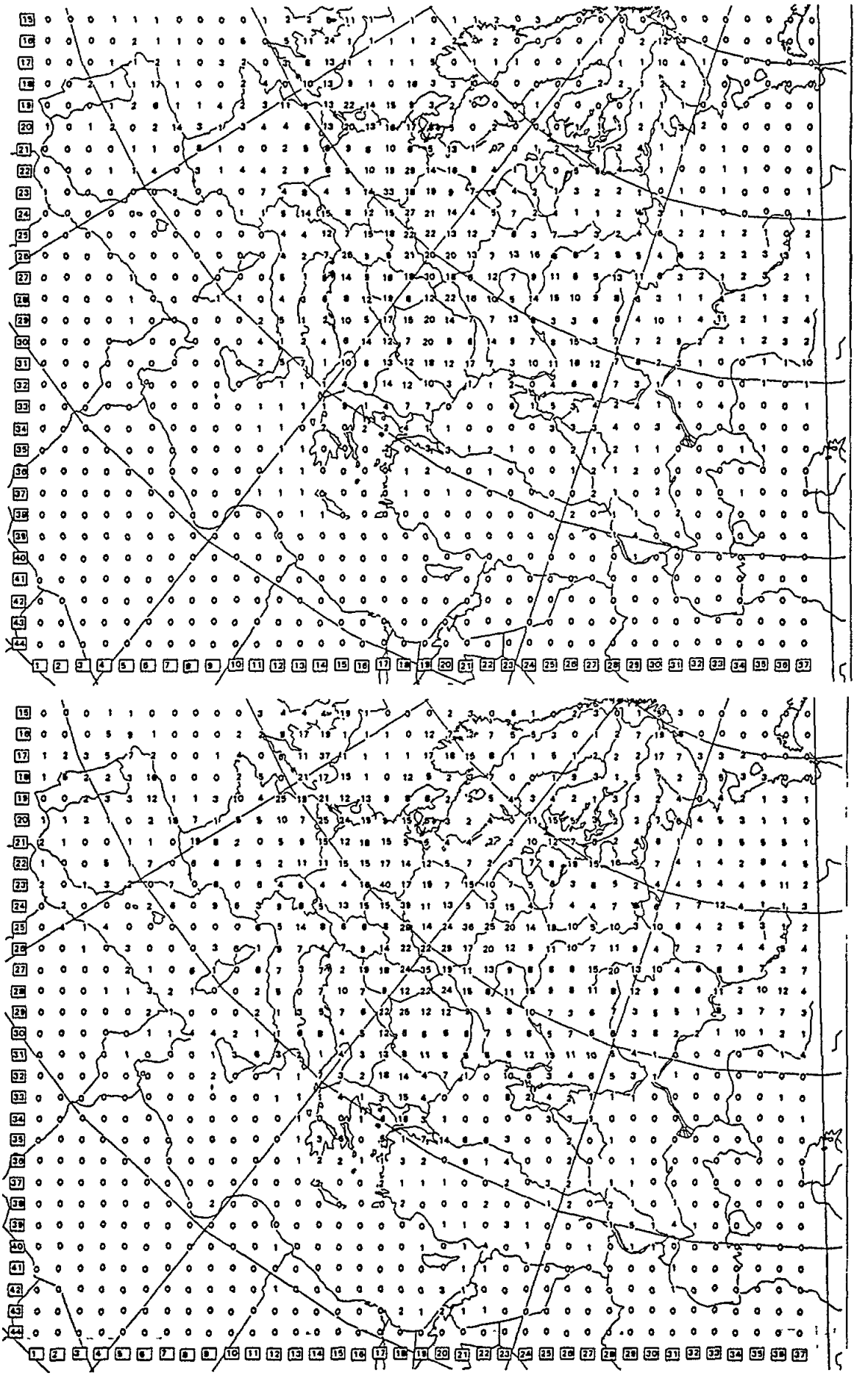


Fig.C6 Wet deposition of sulphur compounds (10 mg S/m^2 month) for July (upper) and October (lower).

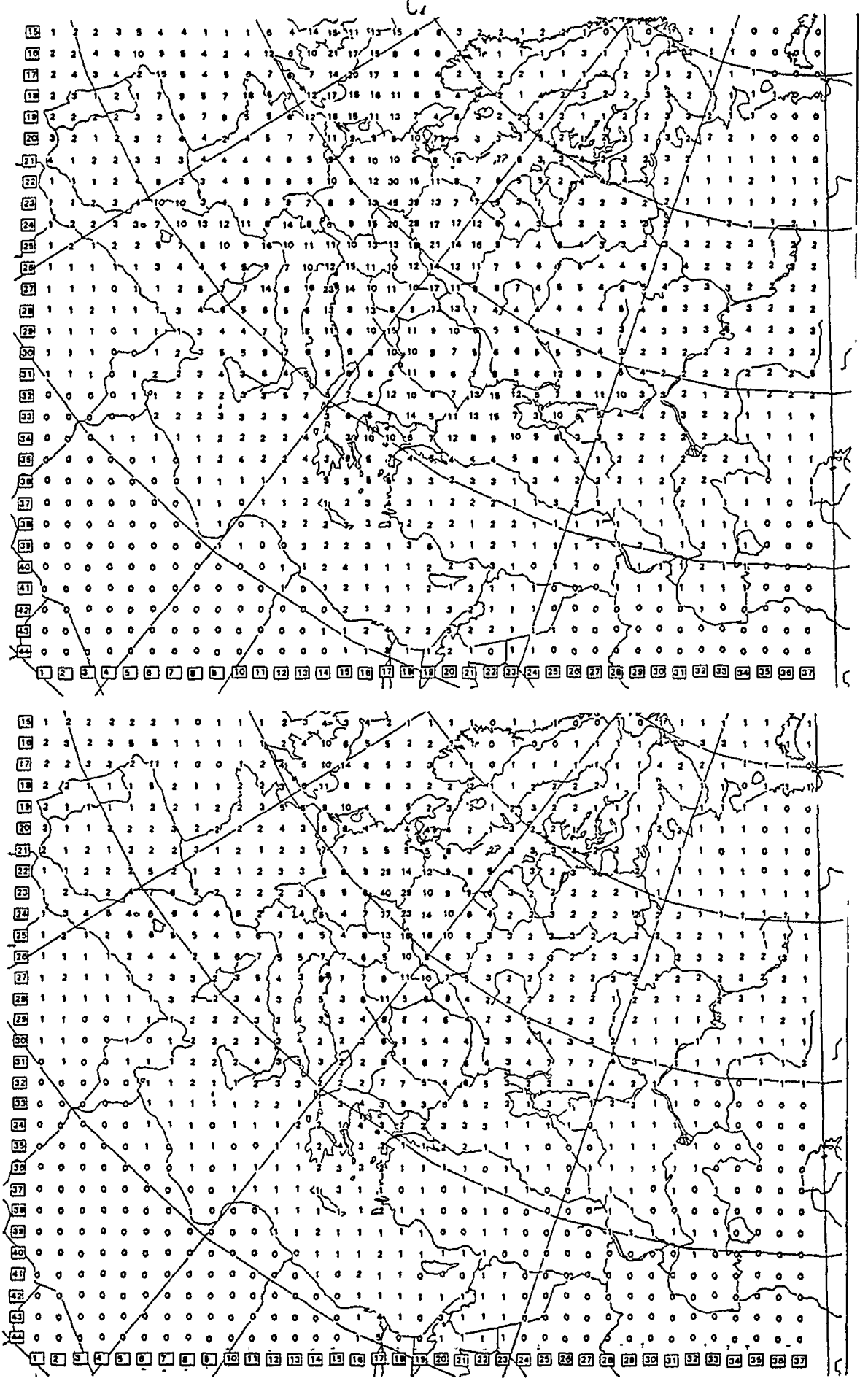


Fig.C7 Dry deposition of sulphur compounds (10 mg S/m^2 month) for January (upper) and April (lower).

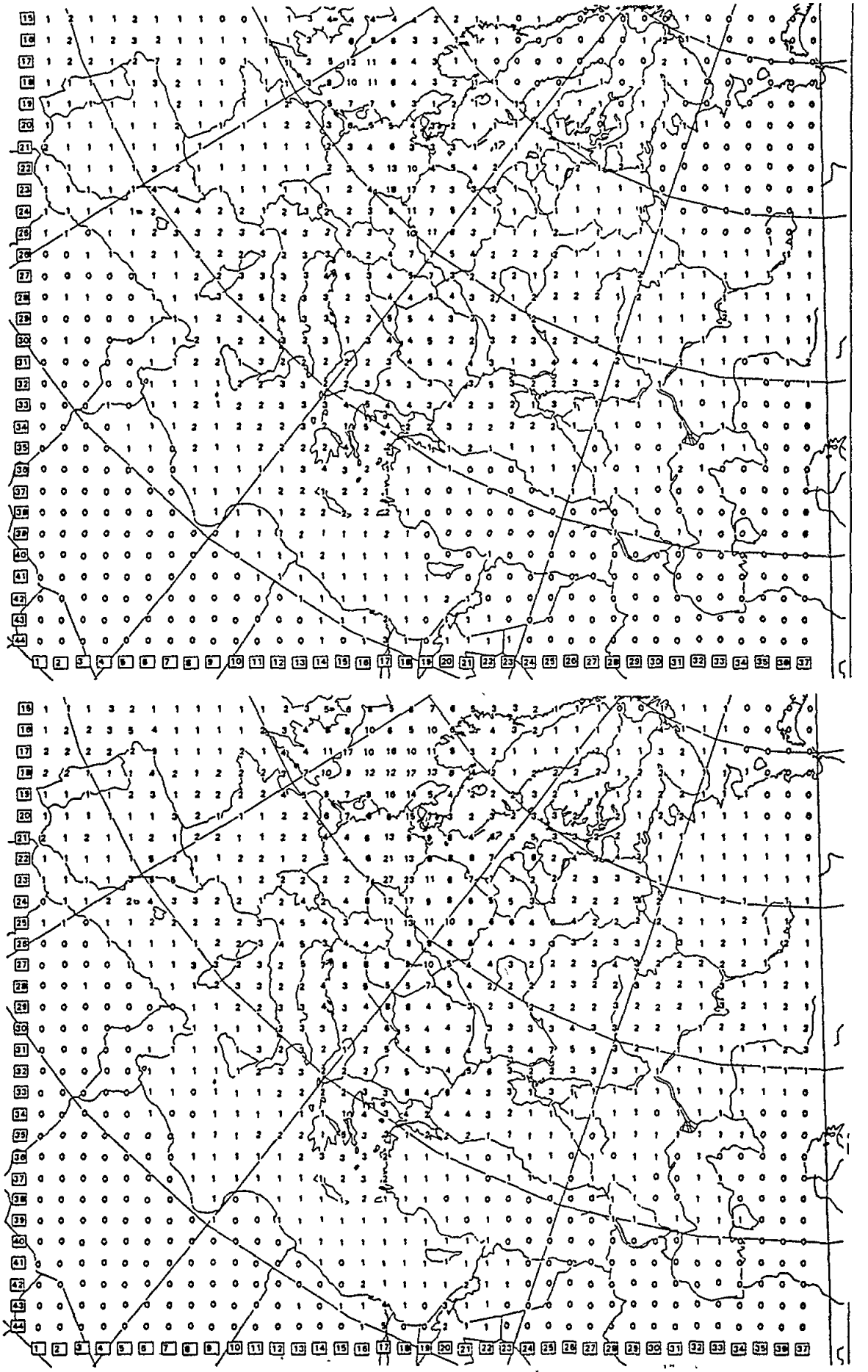


Fig.C8 Dry deposition of sulphur compounds (10 mg S/m^2 month) for July(upper) and October (lower).

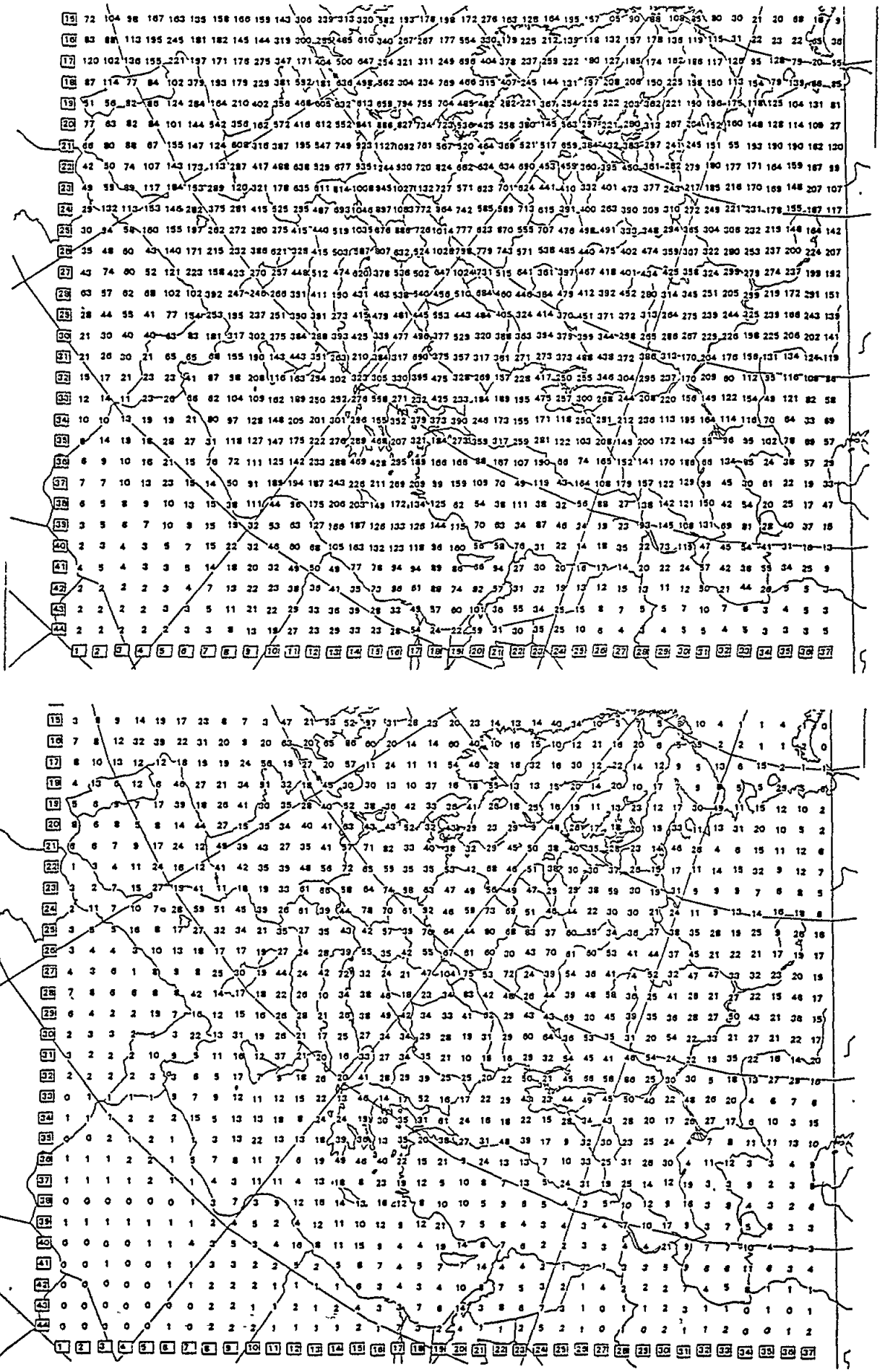


Fig.C9 Total deposition of oxidised nitrogen (mg N/m^2 month) for January (upper) and April (lower).

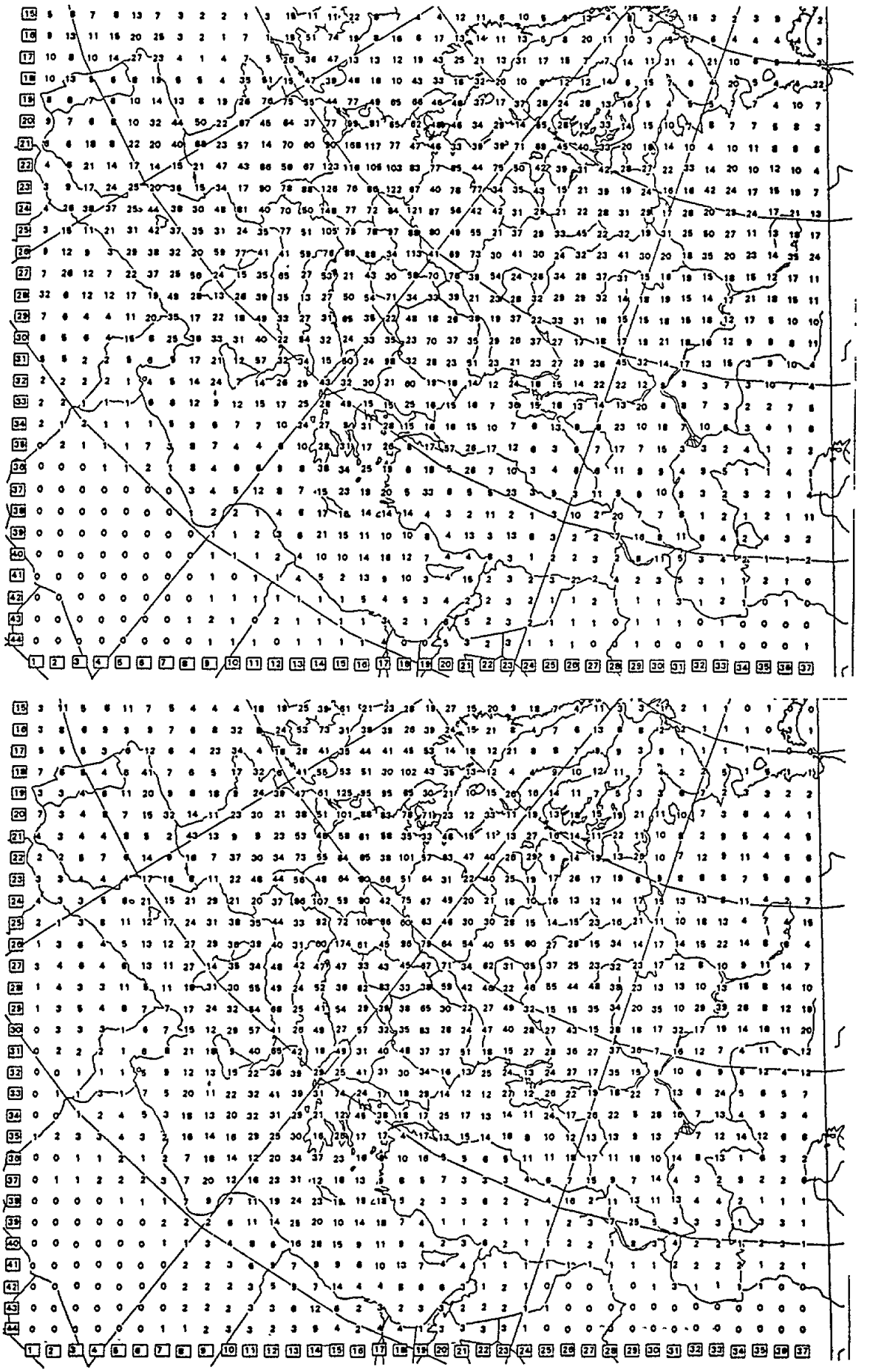


Fig.C10 Total deposition of oxidised nitrogen compounds (mg N/m^2 month) for July (upper) and October (lower).

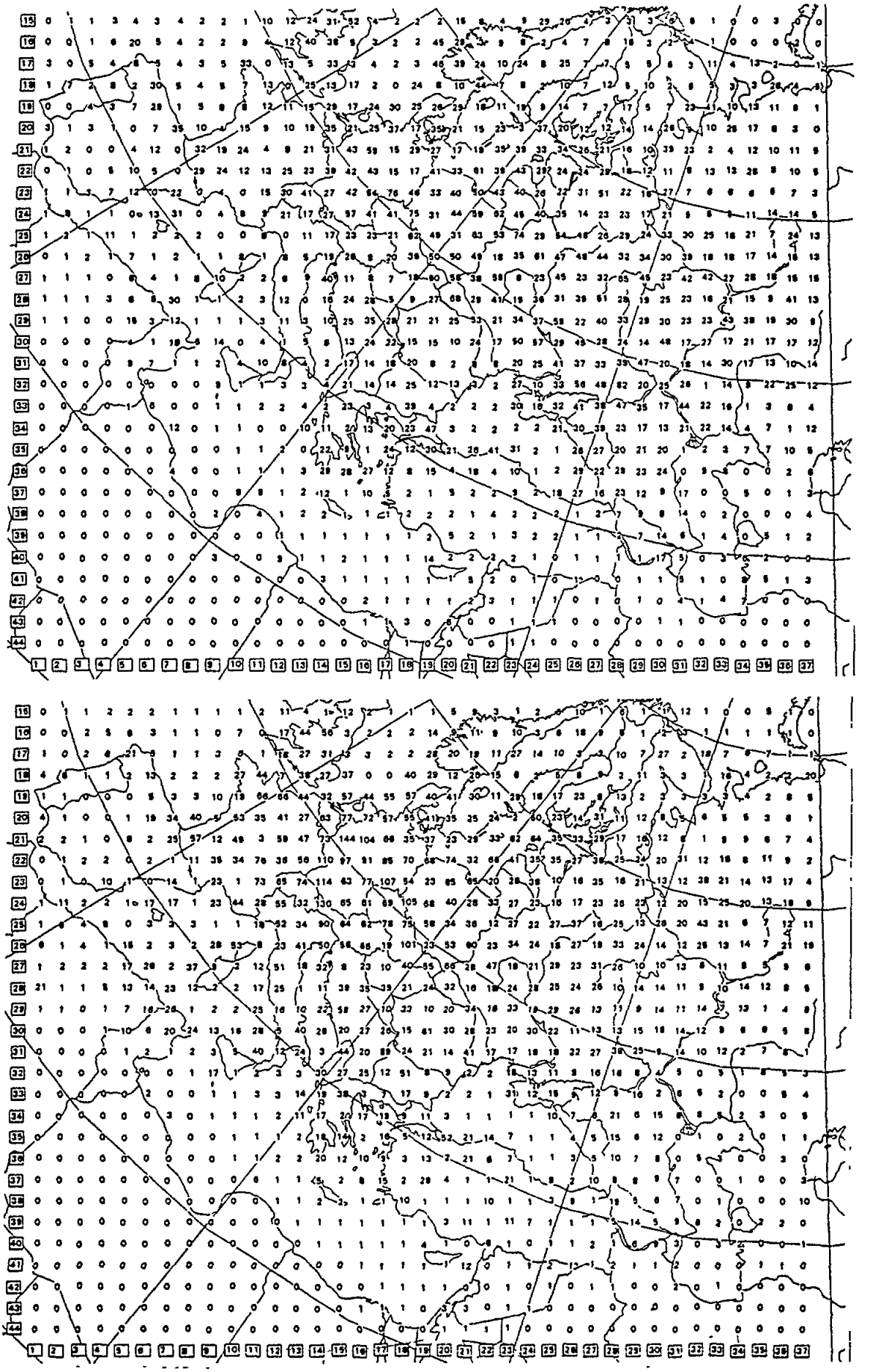


Fig.C11 Wet deposition of oxidised nitrogen compounds (mg N/m^2 month) for January (upper) and April (lower).

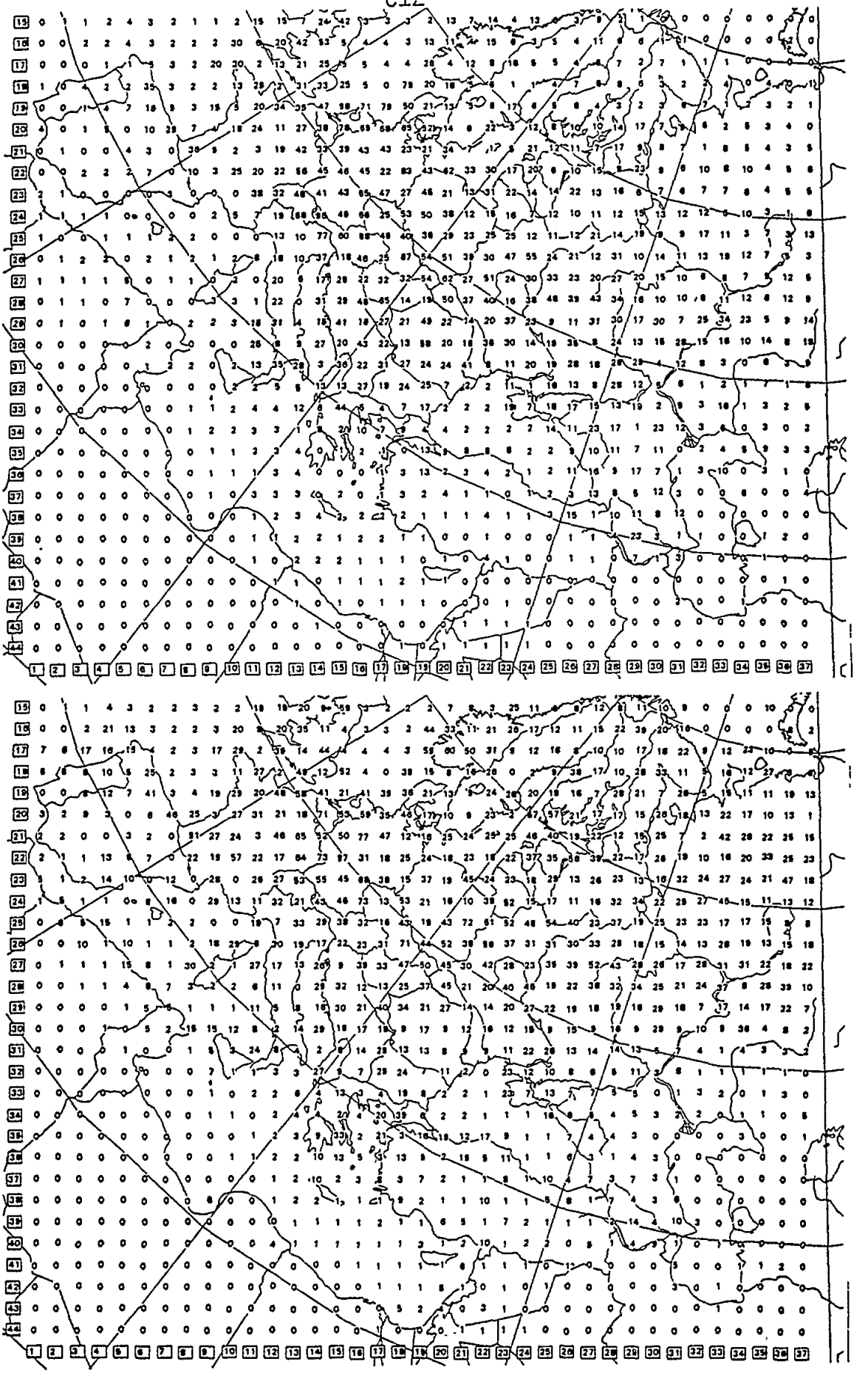


Fig.C12 Wet deposition of oxidised nitrogen compounds (mg N/m^2 month) for July (upper) and October (lower).

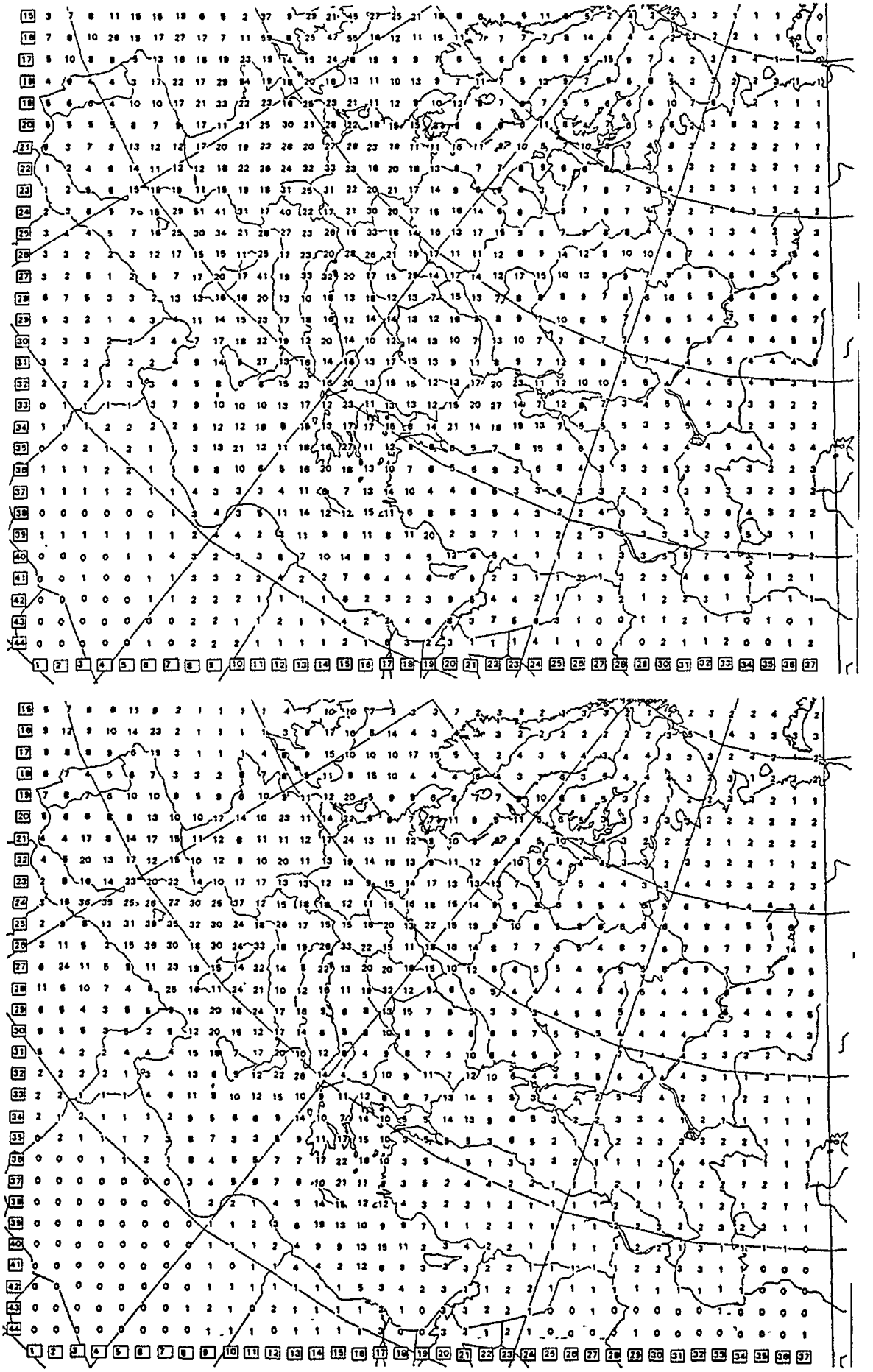


Fig.C13 Dry deposition of oxidised nitrogen compounds (mg N/m^2 month) for January (upper) and April (lower).

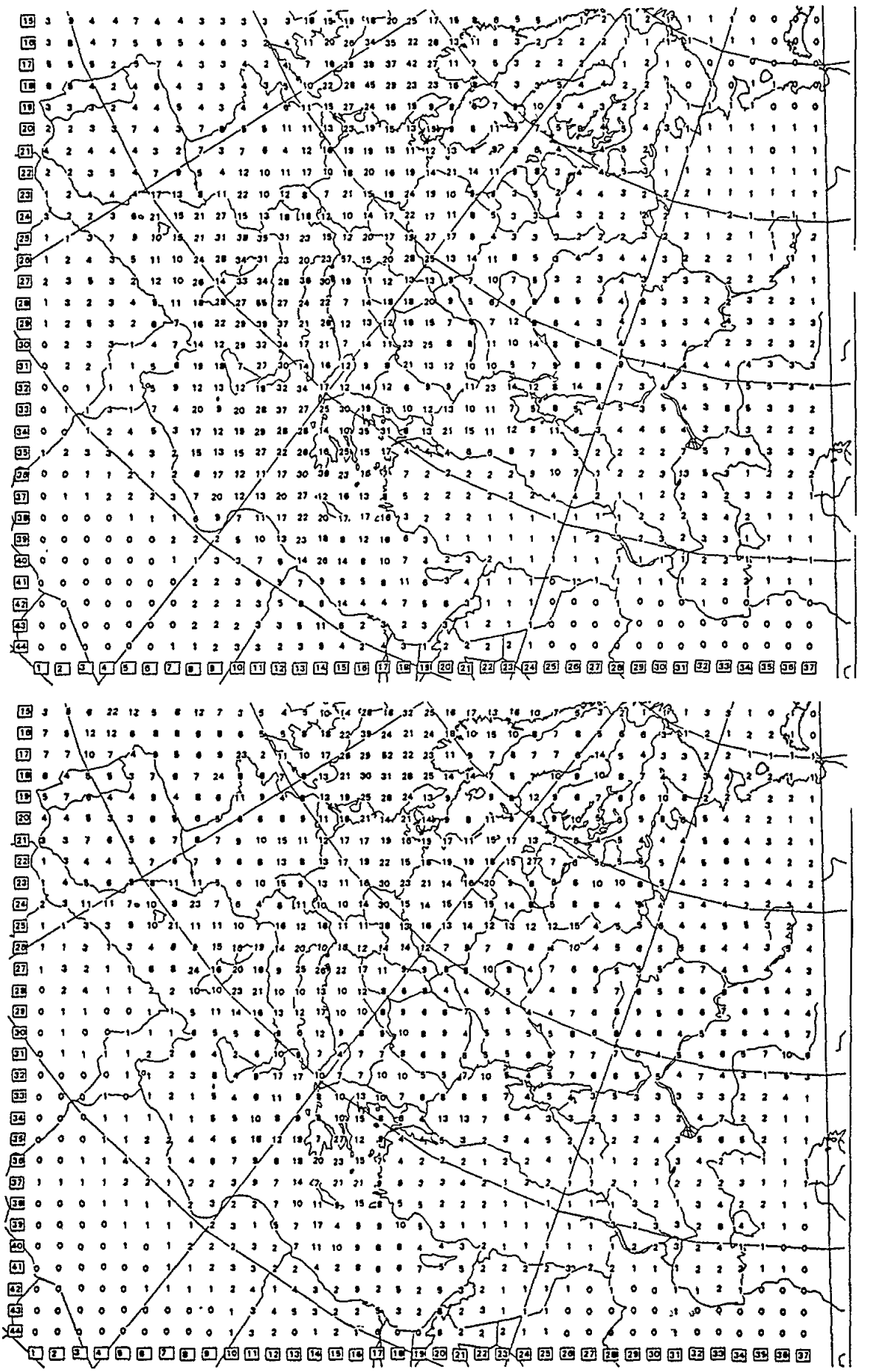


Fig.C14 Dry deposition of oxidised nitrogen compounds (mg N/m^2 month) for July (upper) and October (lower).

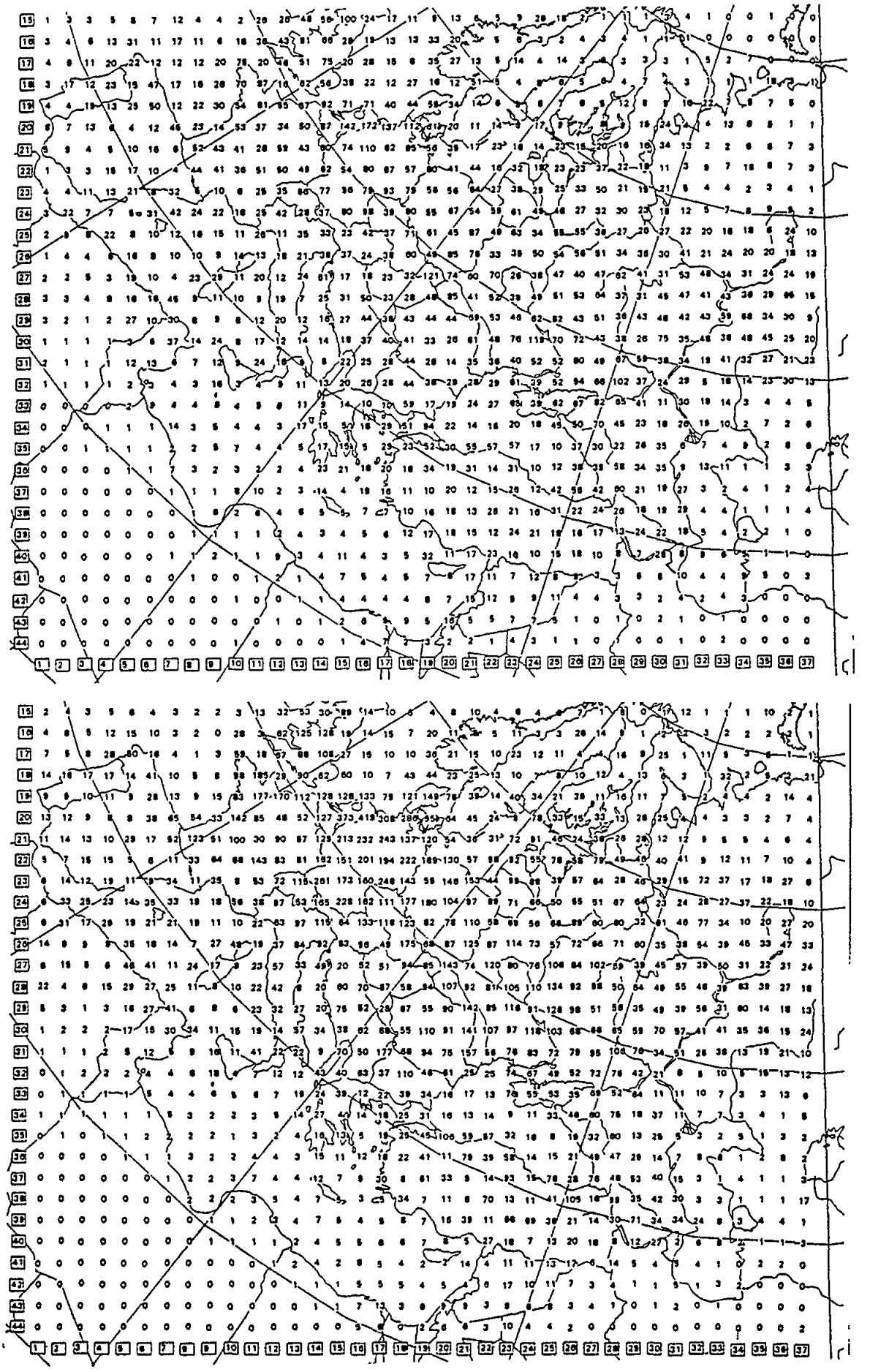


Fig.C15 Total deposition of reduced nitrogen compounds (mg N/m^2 month) for January (upper) and April (lower).

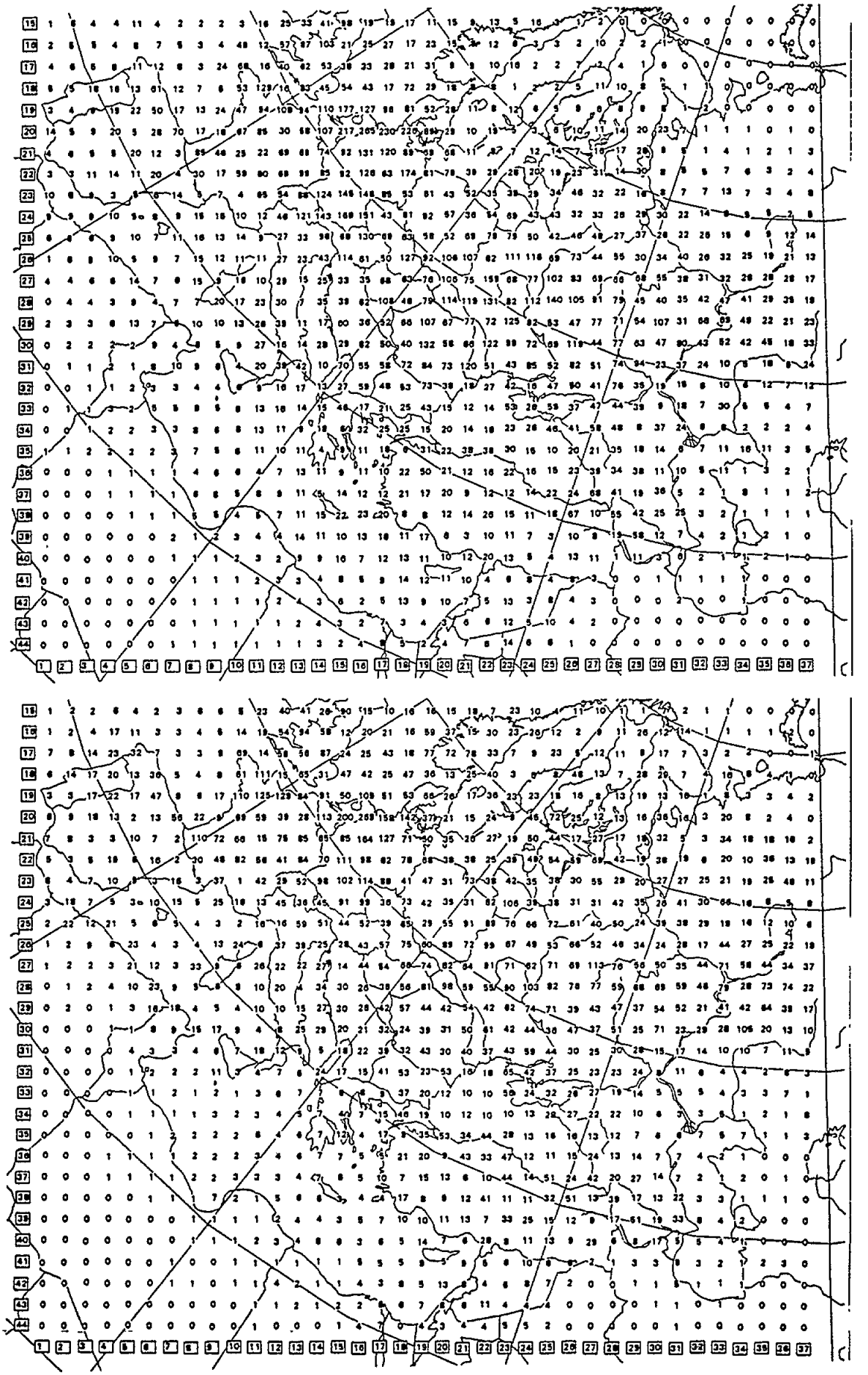


Fig.C16 Total deposition of reduced nitrogen compounds (mg N/m^2 month) for July (upper) and October (lower).

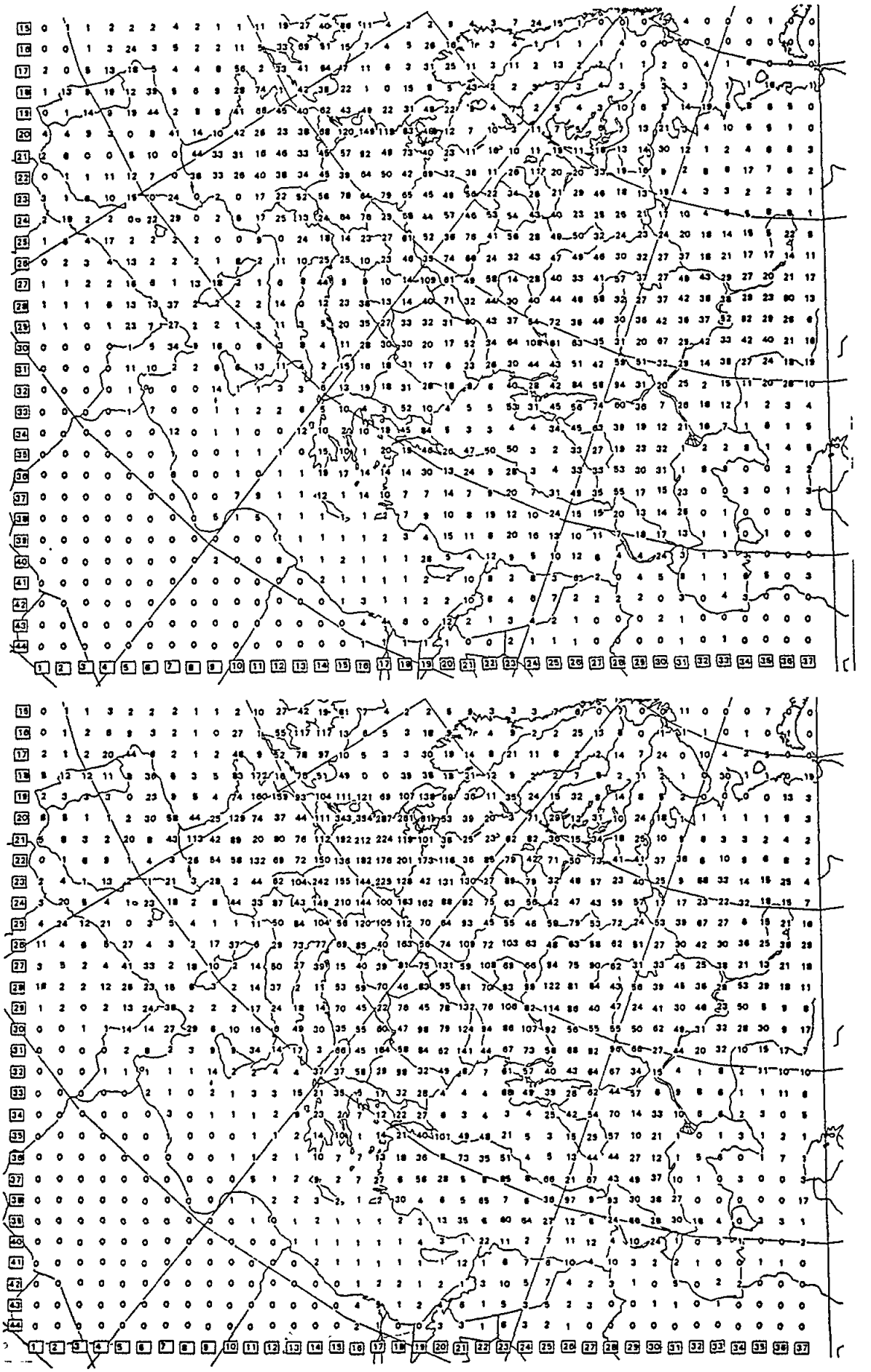


Fig.C17 Wet deposition of reduced nitrogen compounds (mg N/m^2 month) for January (upper) and April (lower).

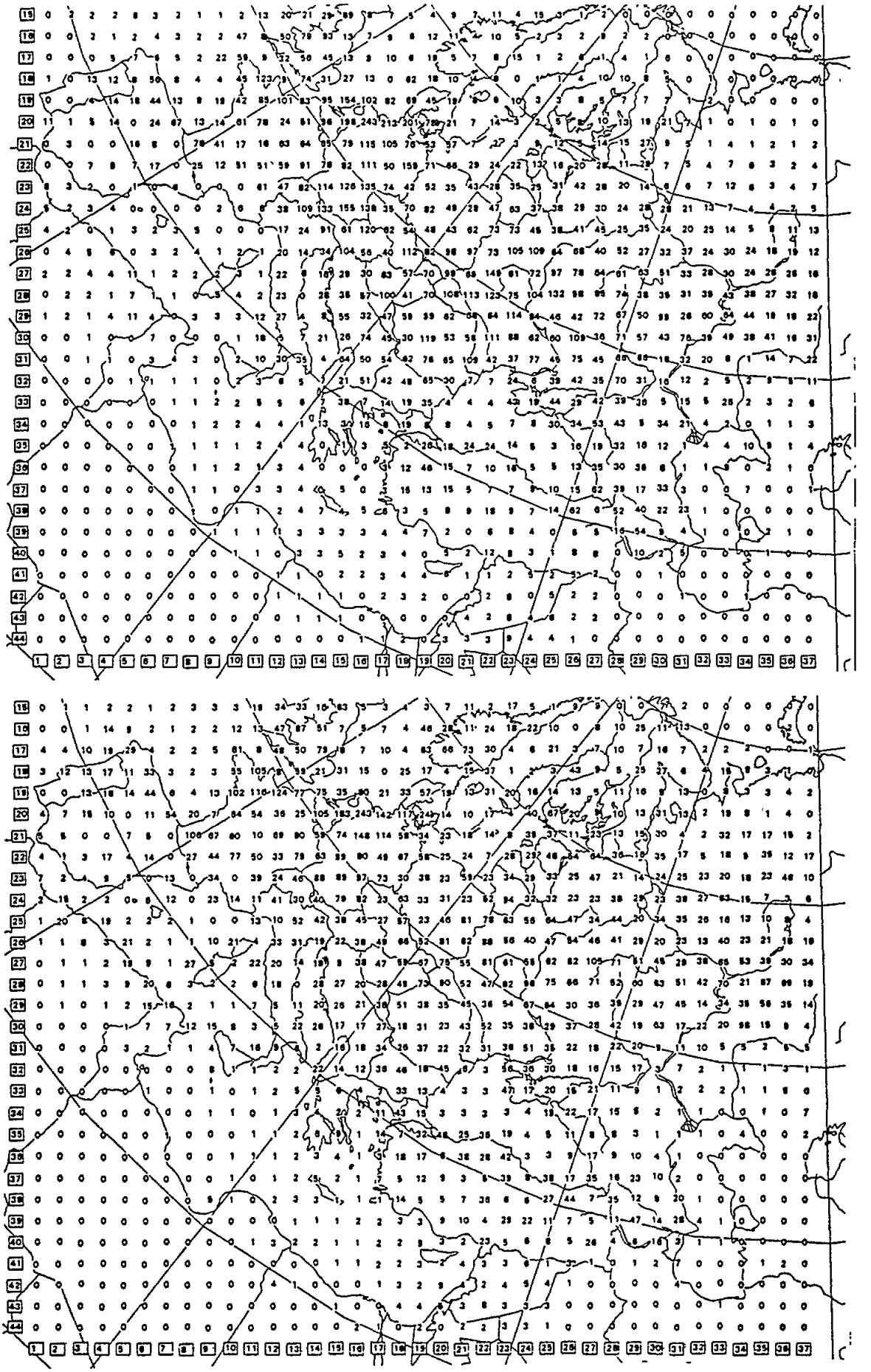


Fig.C18 Wet deposition of reduced nitrogen compounds (mg N/m^2 month) for July (upper) and October (lower).

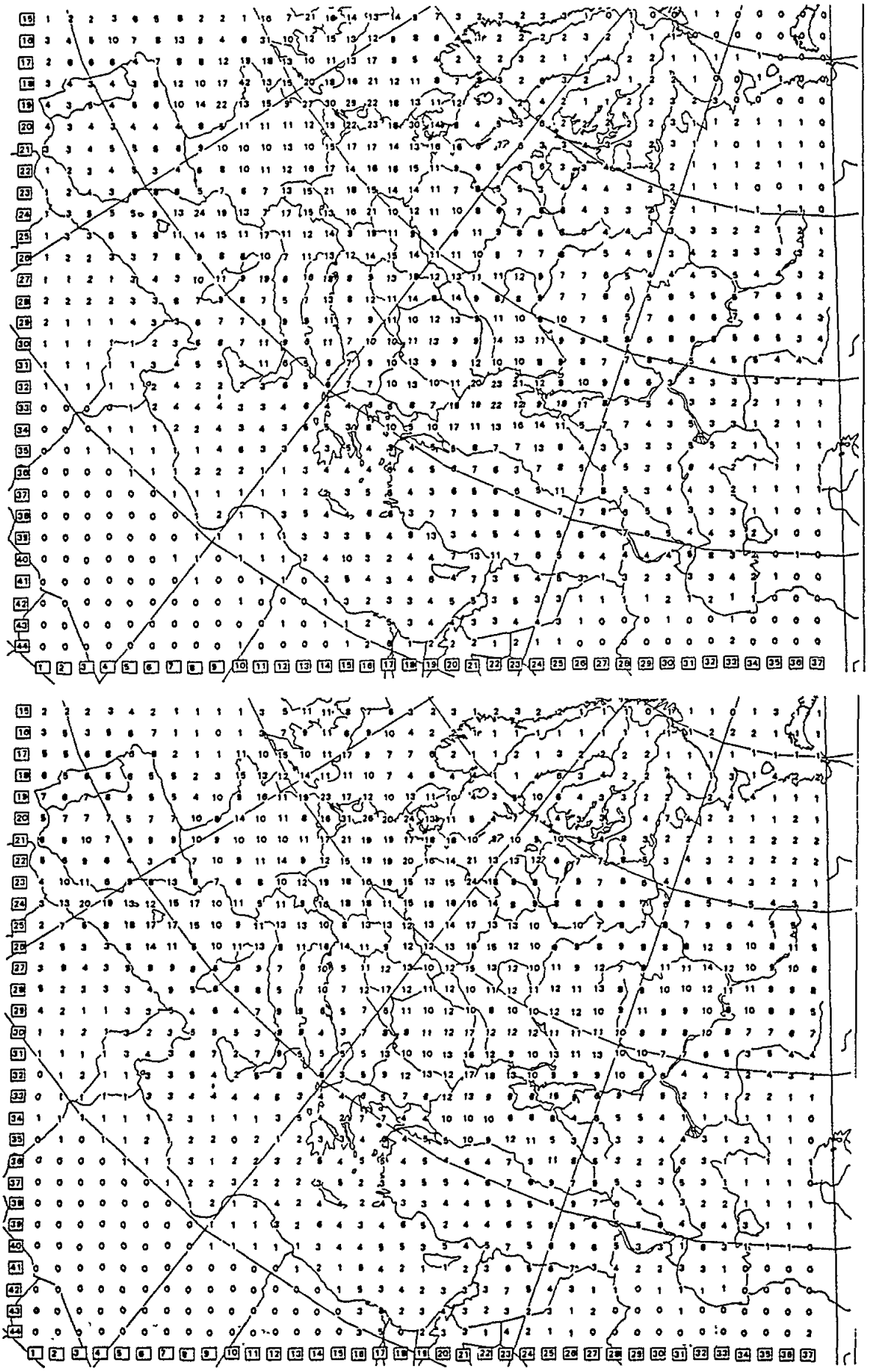


Fig.C19 Dry deposition of reduced nitrogen compounds (mg N/m^2 month) for January (upper) and April (lower).

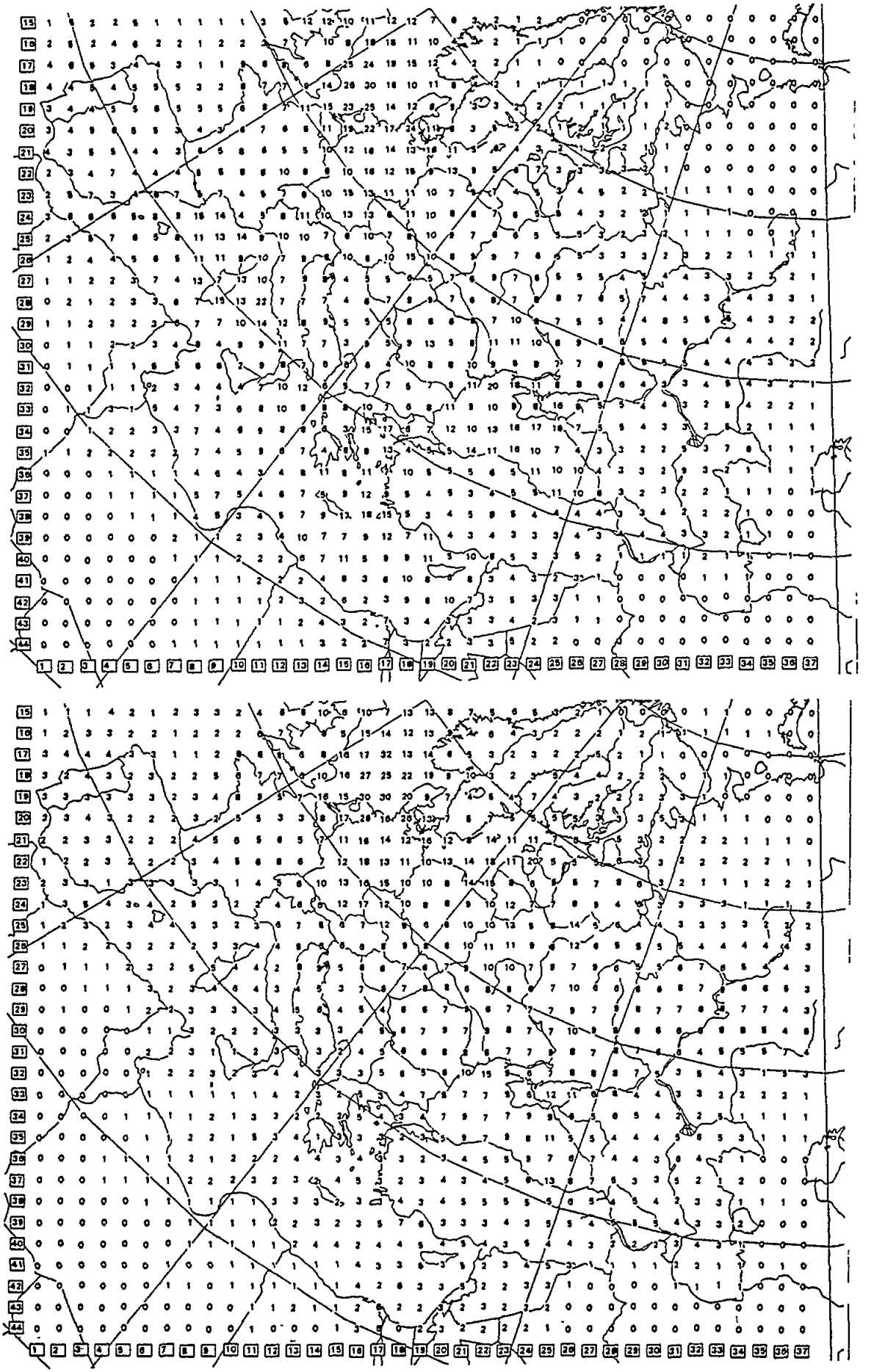


Fig.C20 Dry deposition of reduced nitrogen compounds (mg N/m^2 month) for July (upper) and October (lower).

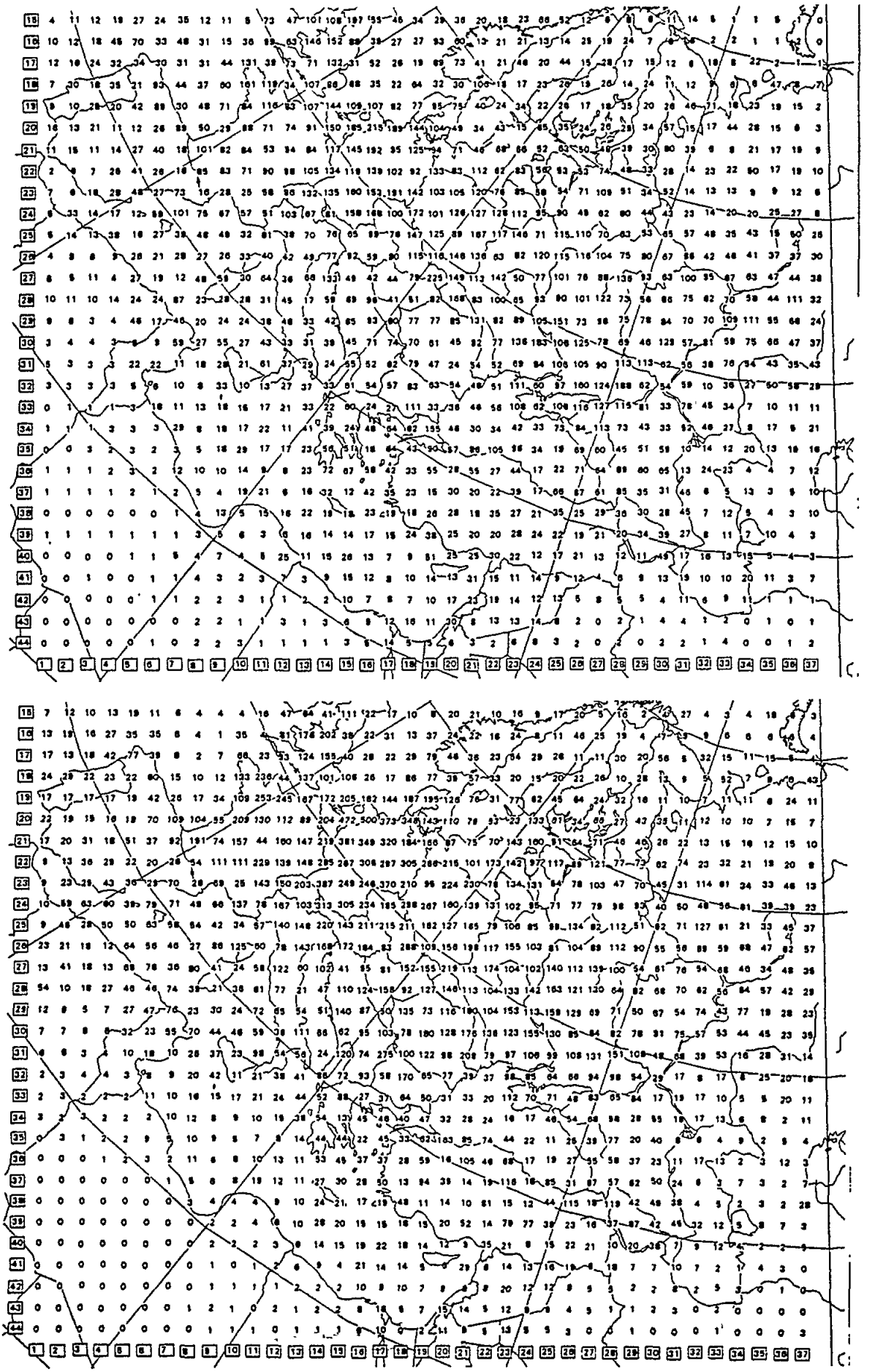


Fig.C21 Total deposition of bound nitrogen (mg N/m^2 month) for January (upper) and April (lower).

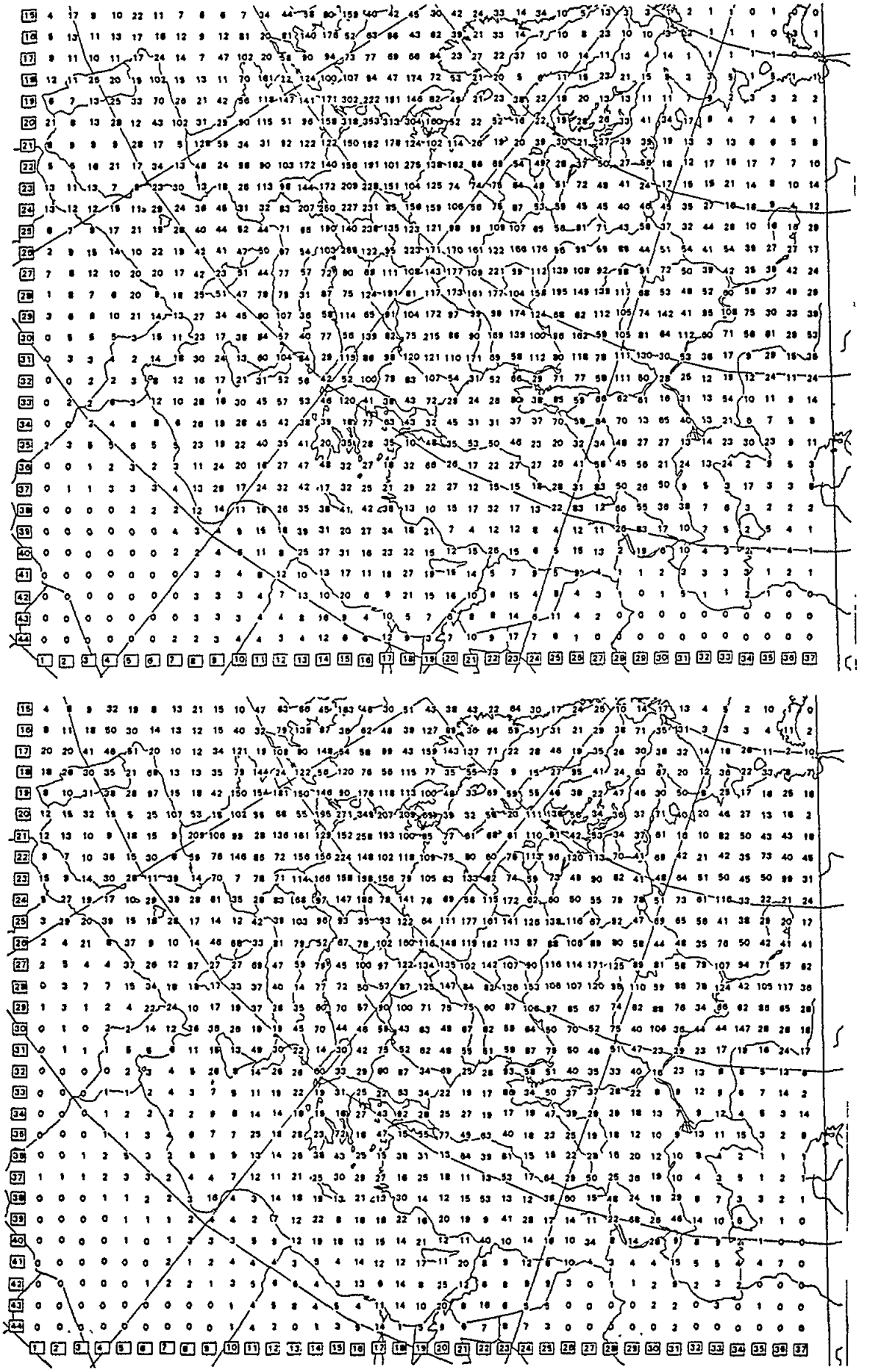


Fig.C22 Total deposition of bound nitrogen (mg N/m^2 month) for July (upper) and October (lower).

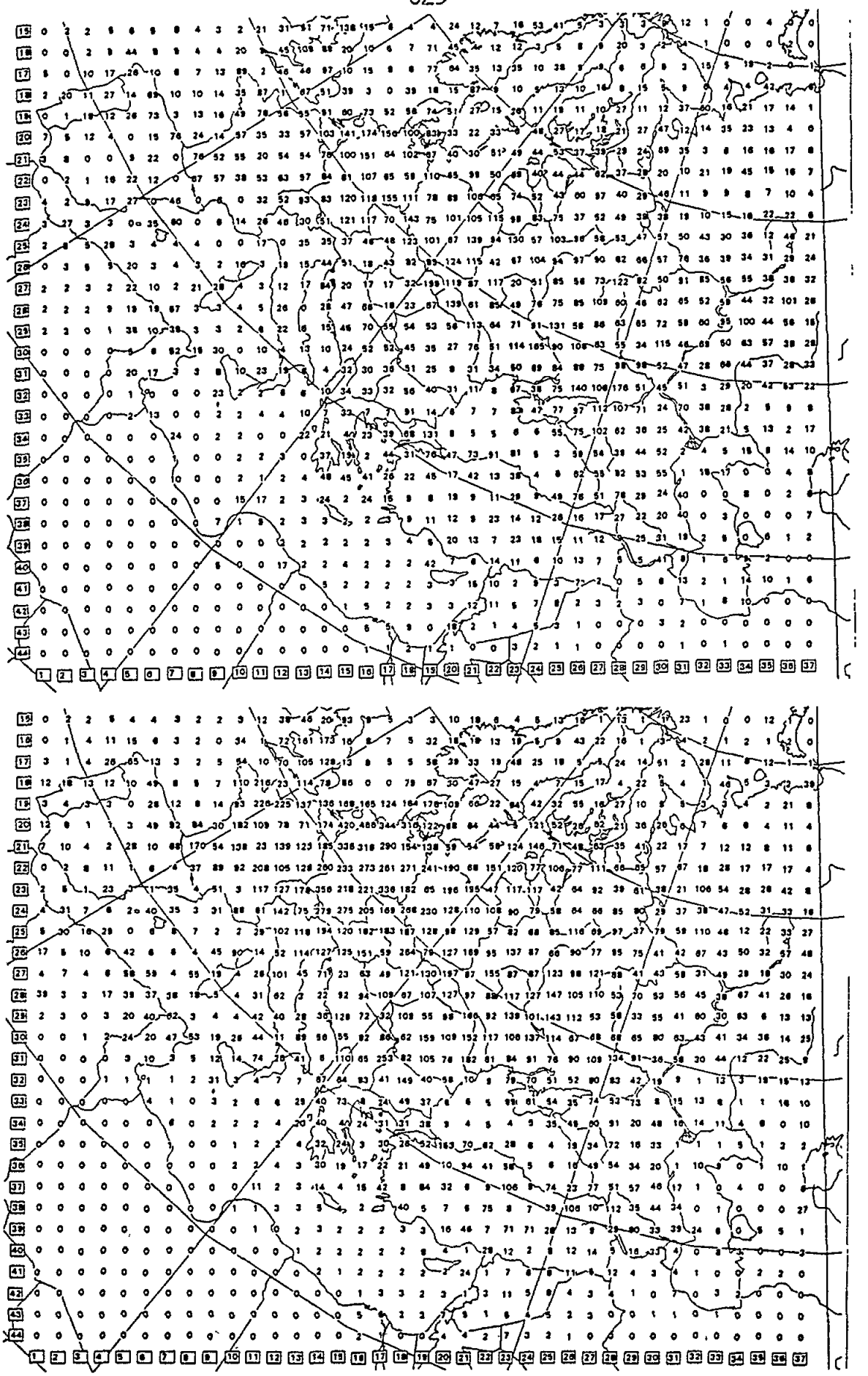
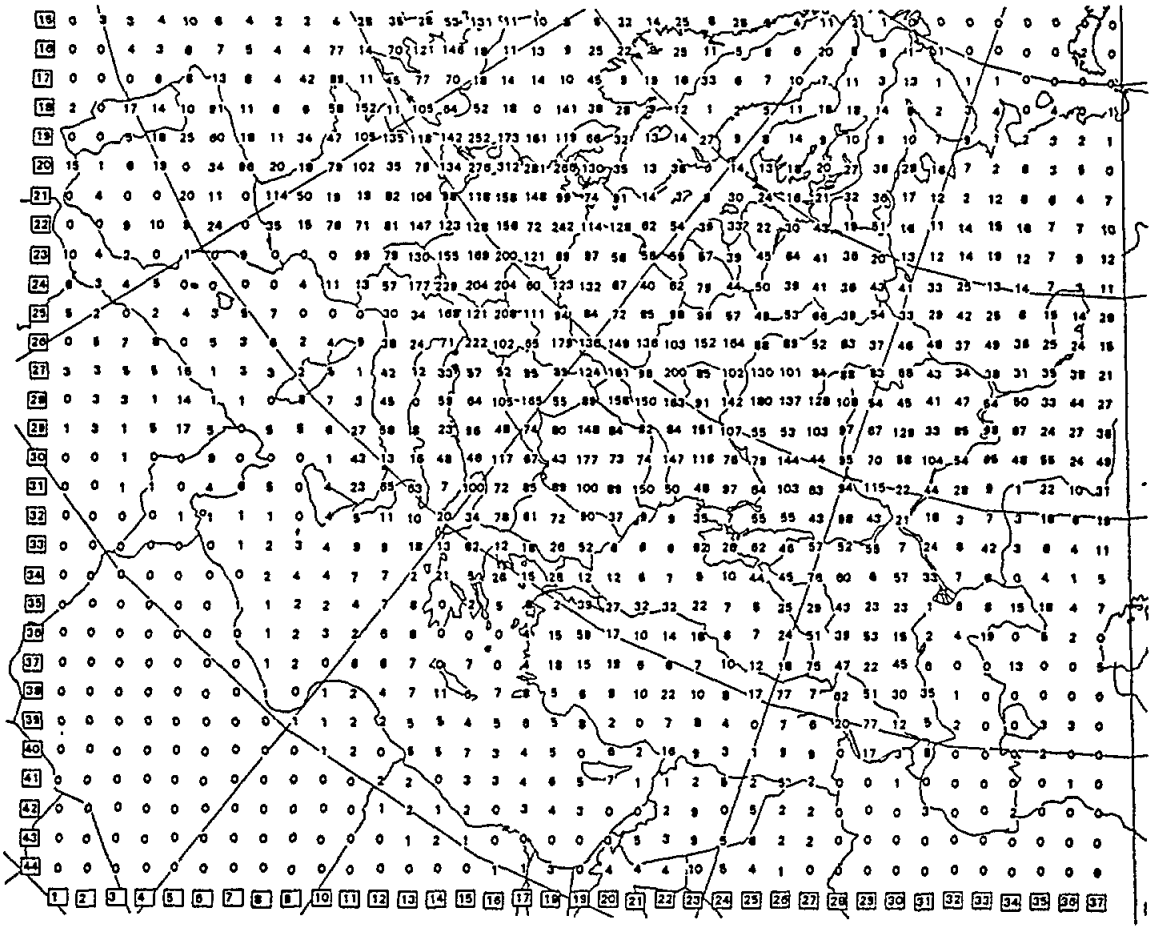


Fig.C23 Wet deposition of bound nitrogen (mg N/m^2 month) for January (upper) and April (lower).



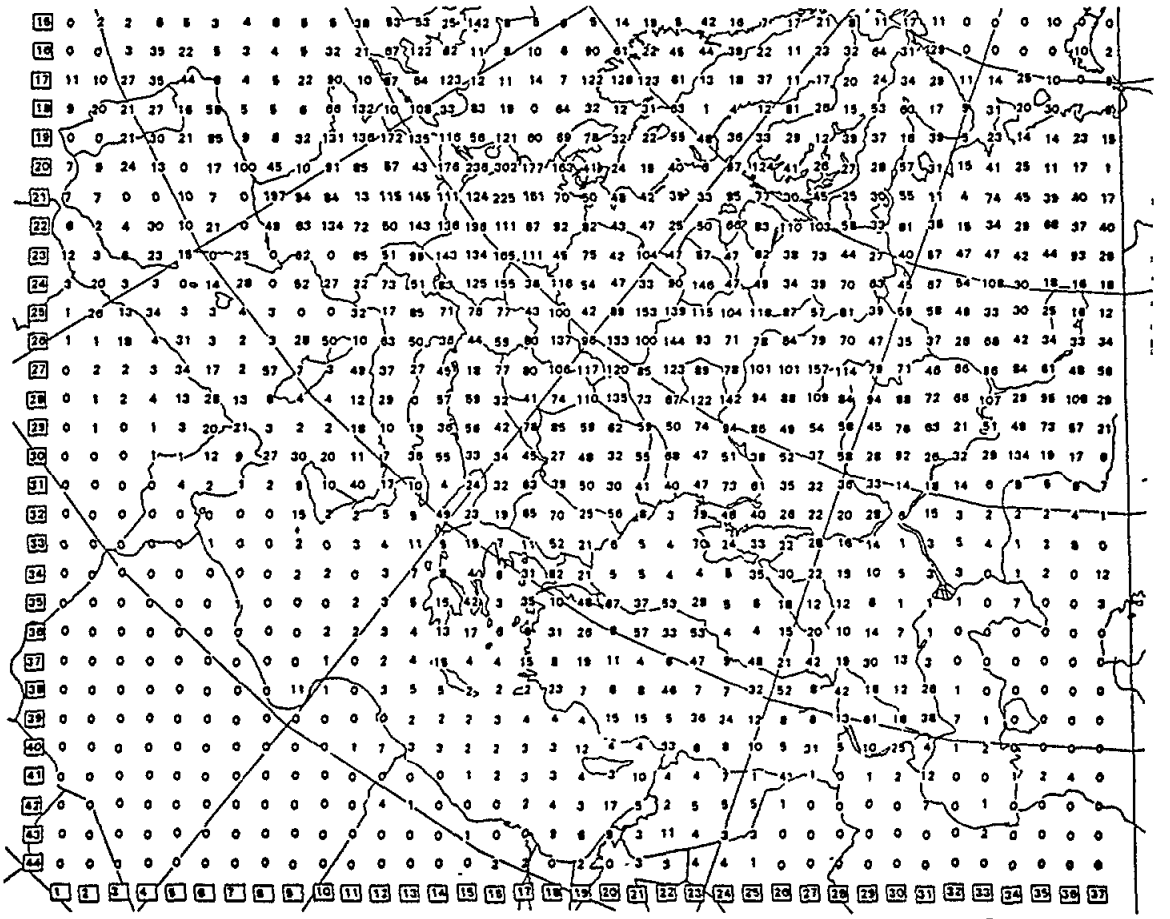


Fig.C24 Wet deposition of bound nitrogen (mg N/m^2 month) for July (upper) and October (lower).

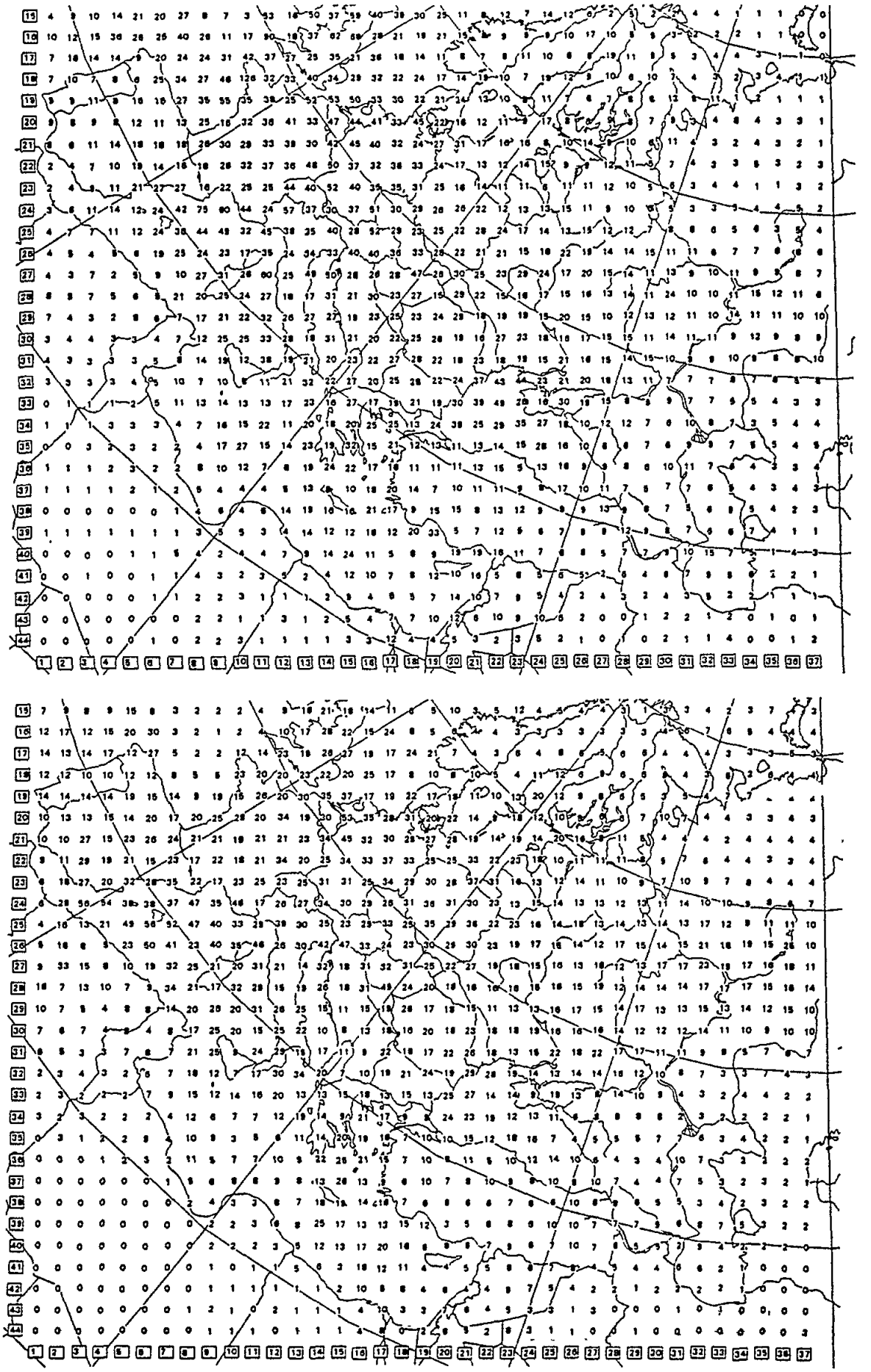


Fig.C25 Dry deposition of bound nitrogen (mg N/m^2 month) for January (upper) and April (lower).

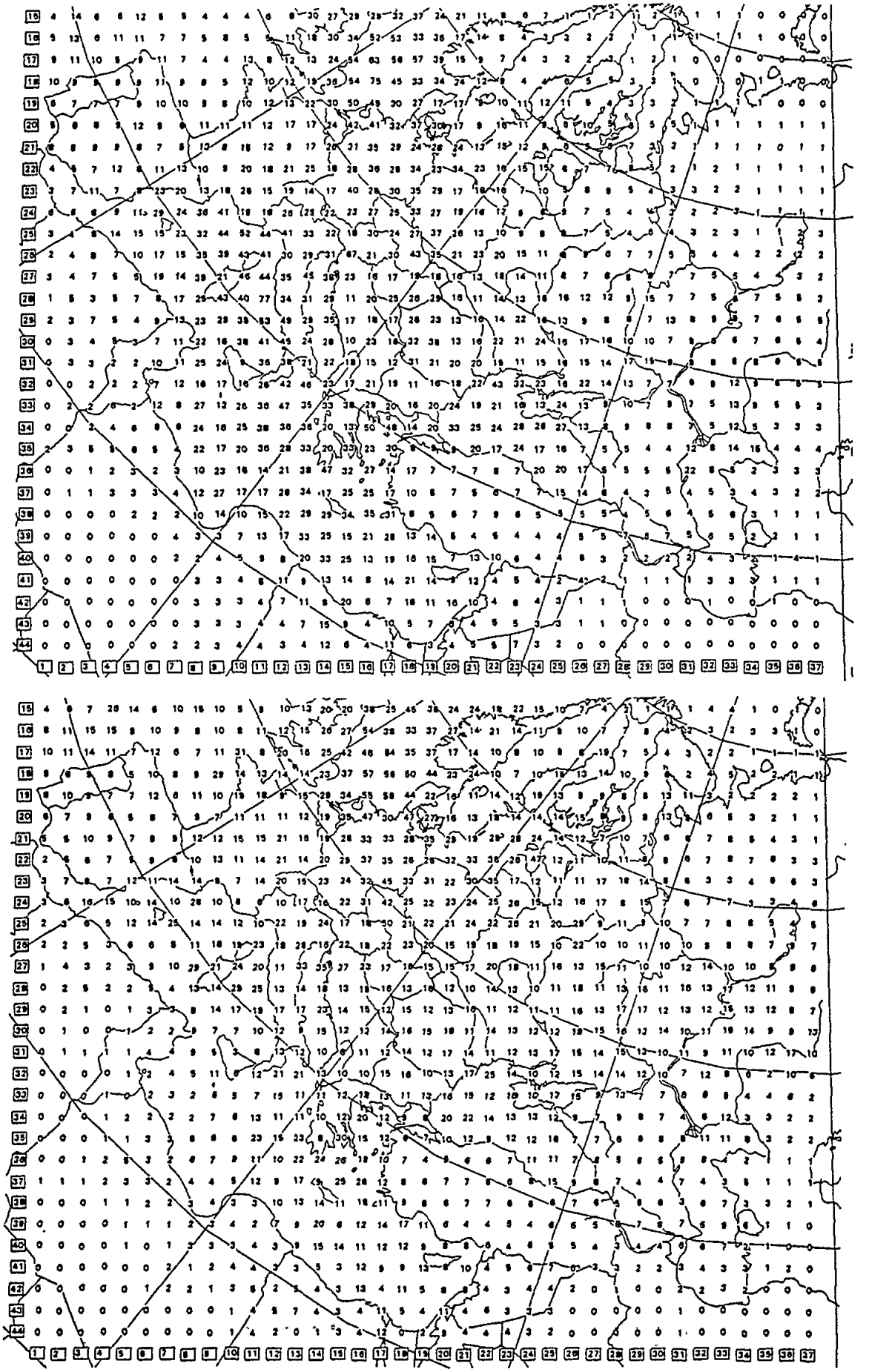


Fig.C26 Dry deposition of bound nitrogen (mg N/m^2 month) for July (upper) and October (lower).

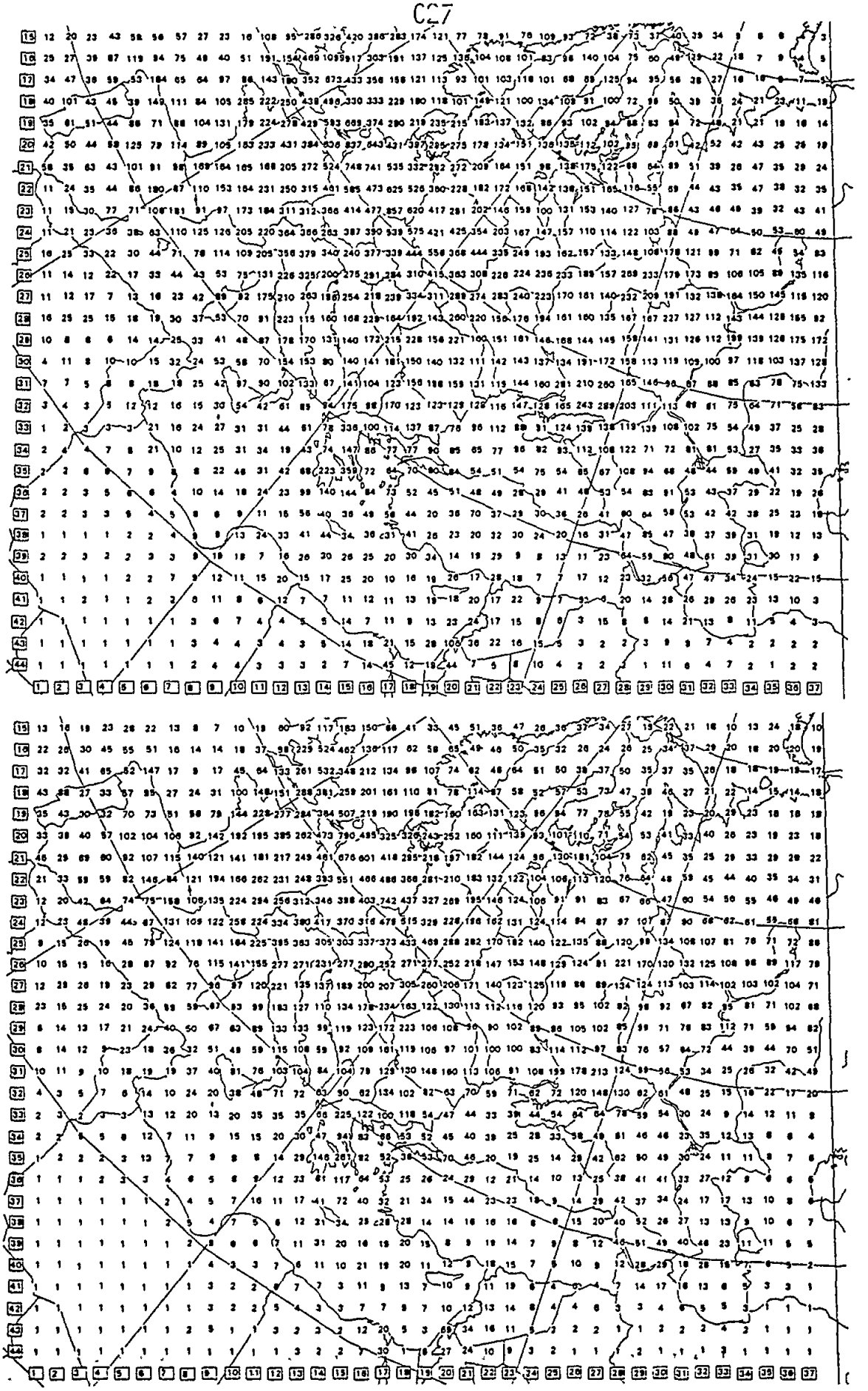


Fig.C27 Mean monthly NO_2 air concentrations (0.01 ug N/m^3) for January (upper) and April (lower).

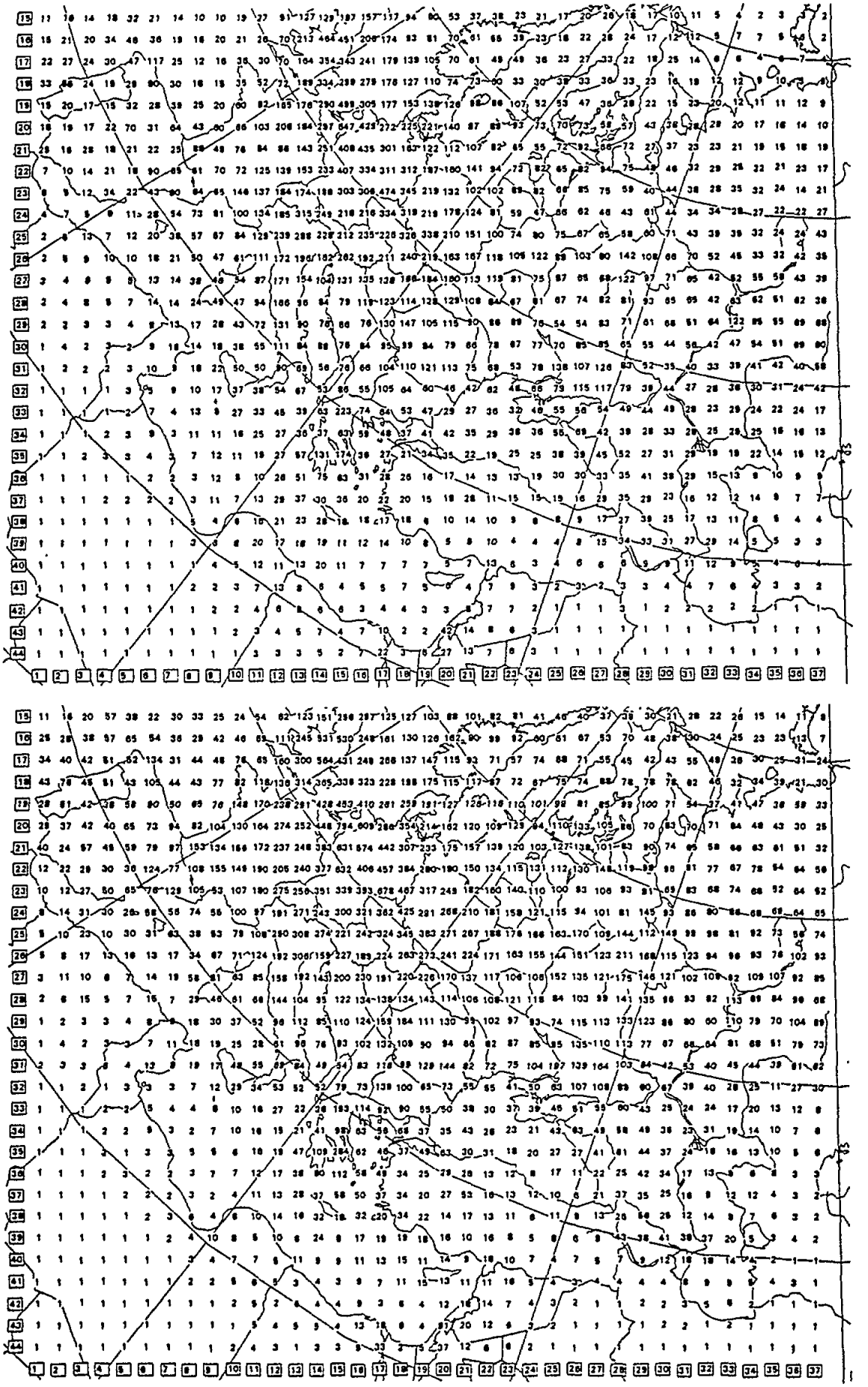


Fig.C28 Mean monthly NO_2 air concentrations (0.01 ug N/m^3) for July (upper) and October (lower).

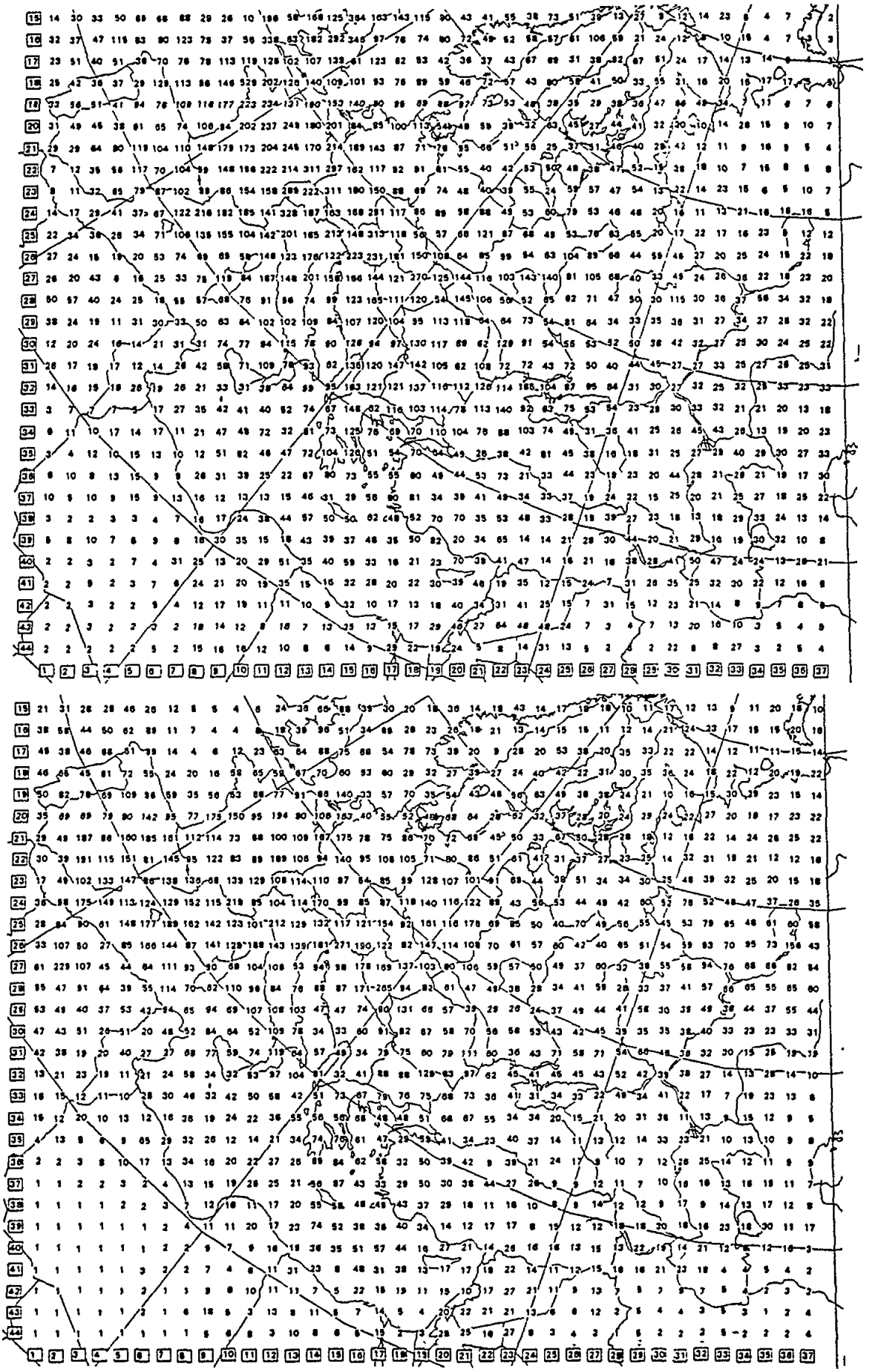


Fig.C29 Mean monthly HNO_3+NO_3 air concentrations (0.01 ug N/m^3) for January (upper) and April (lower).

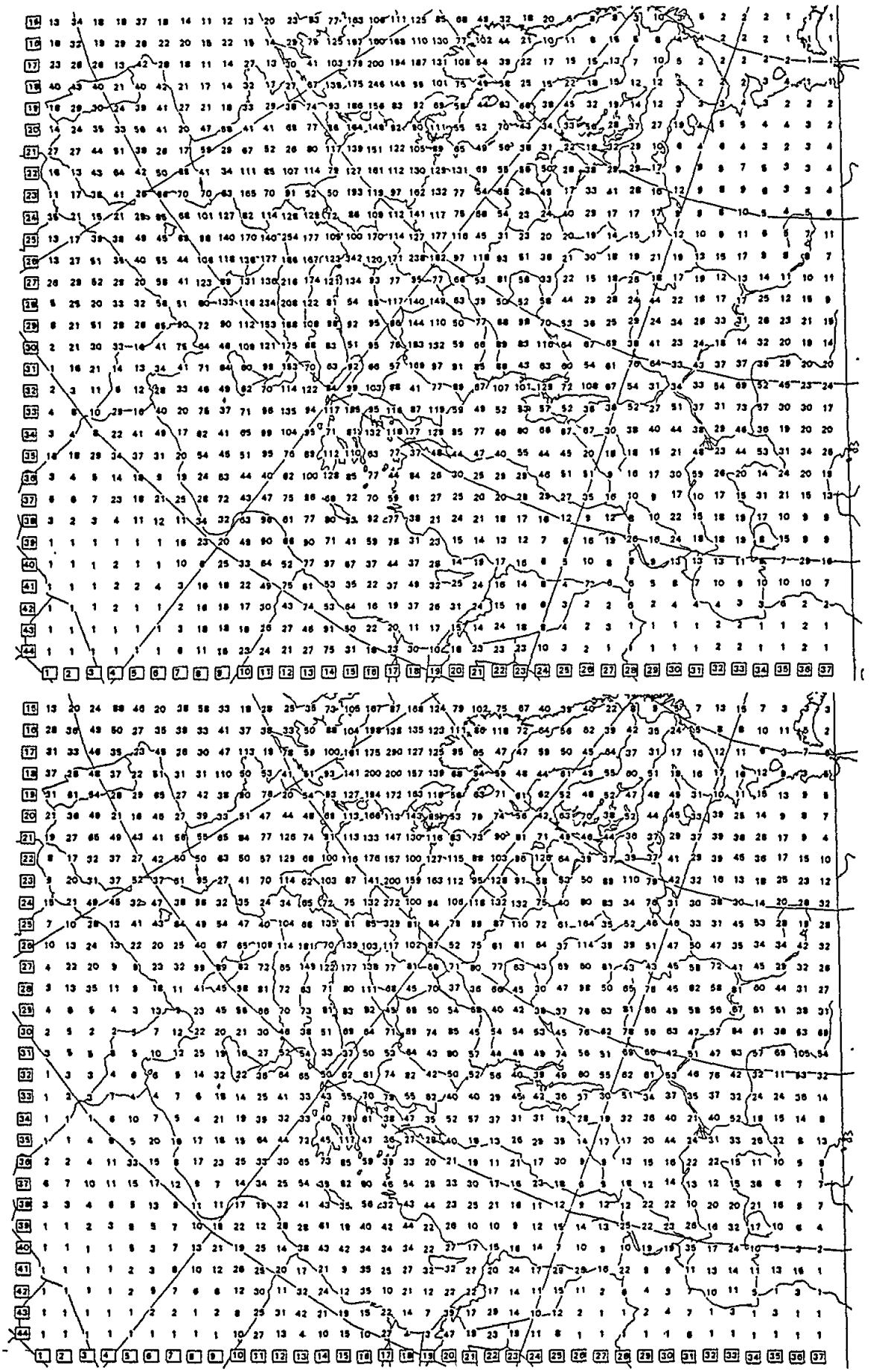


Fig.C30 Mean monthly HNO_3+NO_3 air concentrations (0.01 ug N/m^3) for July (upper) and October (lower).

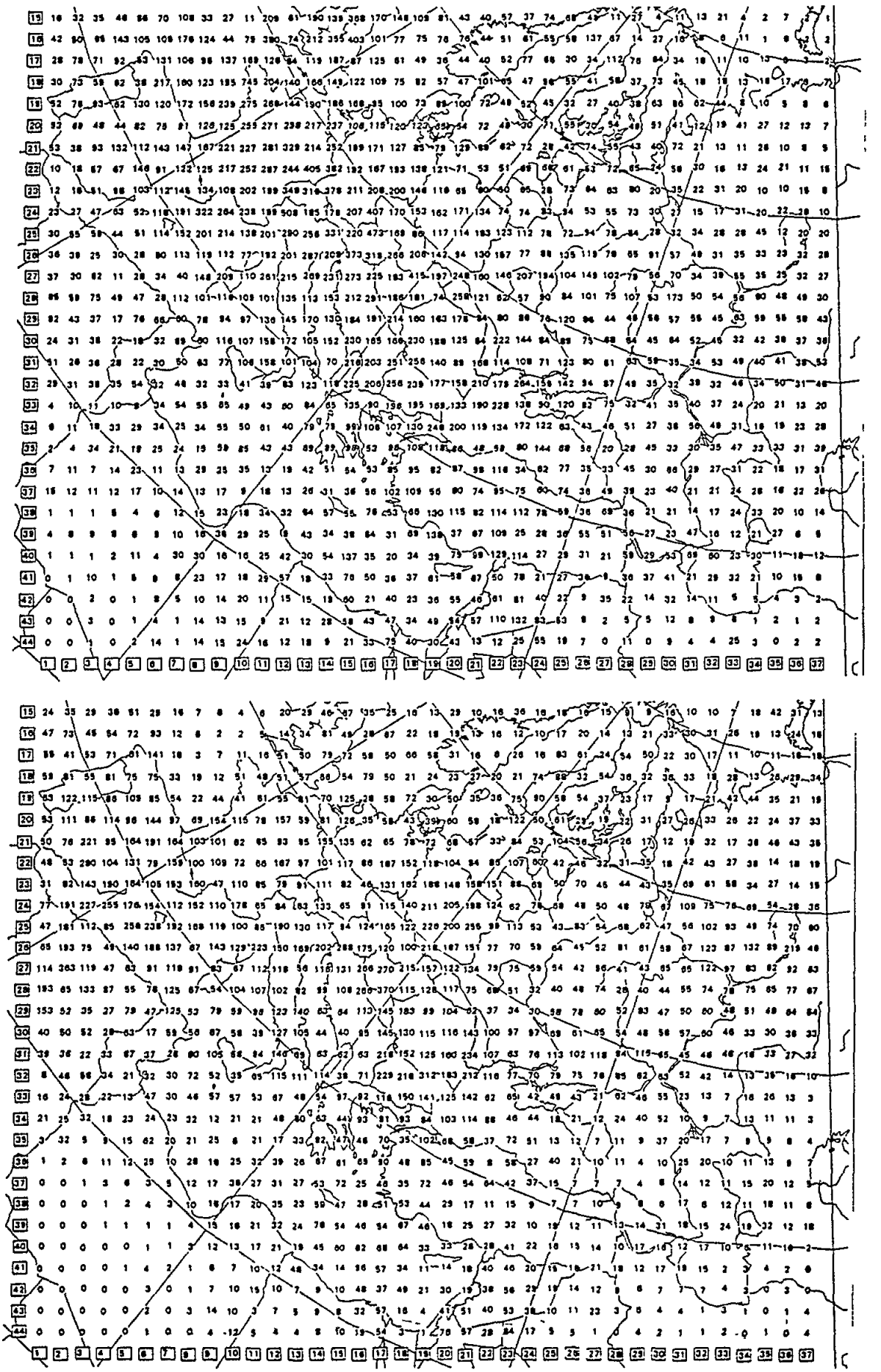


Fig.C31 Mean monthly NH_4 air concentrations (0.01 ug N/m^3) for January (upper) and April (lower).

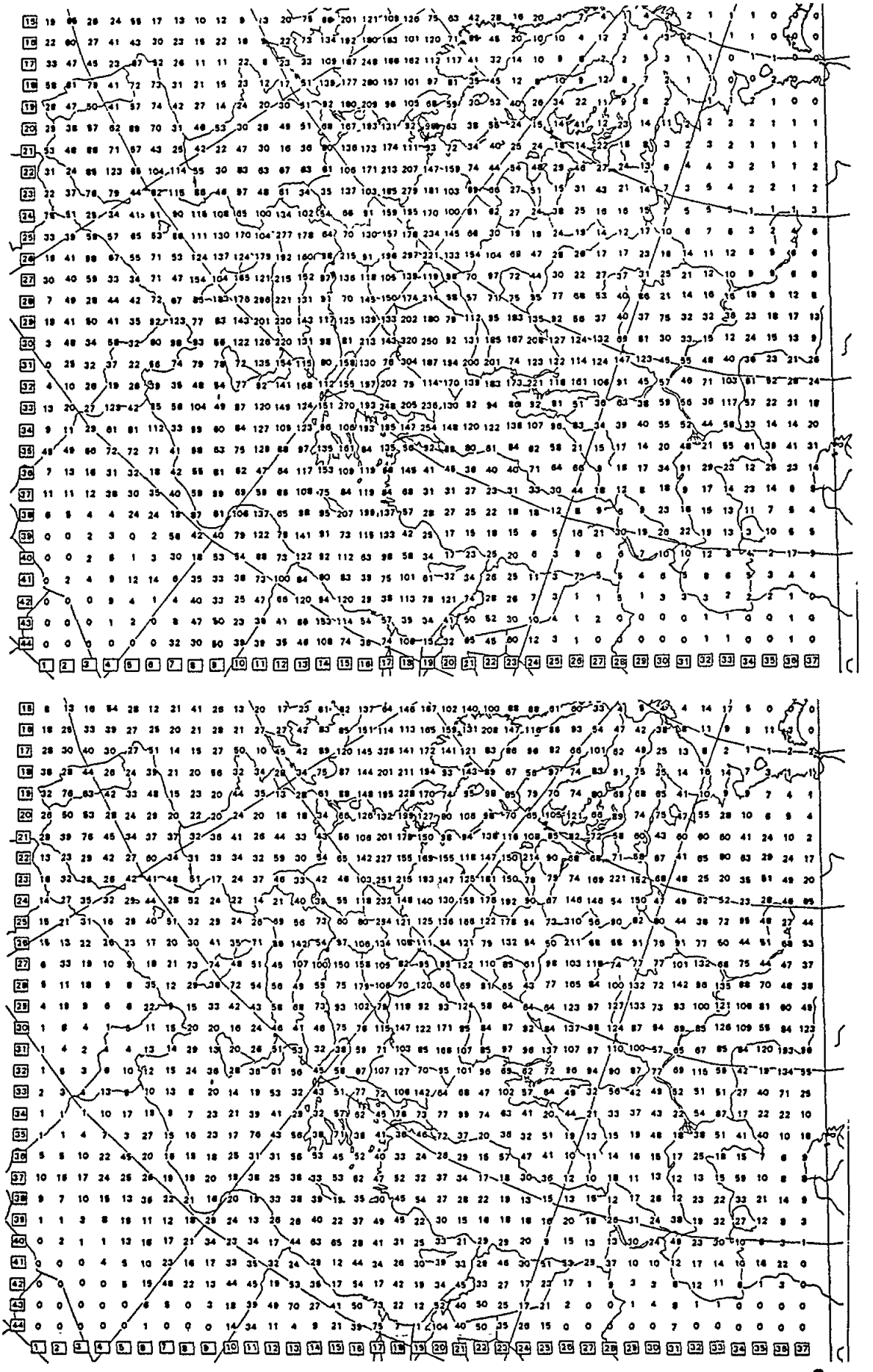


Fig.C32 Mean monthly NH_4 air concentrations (0.01 ug N/m^3) for July (upper) and October (lower).

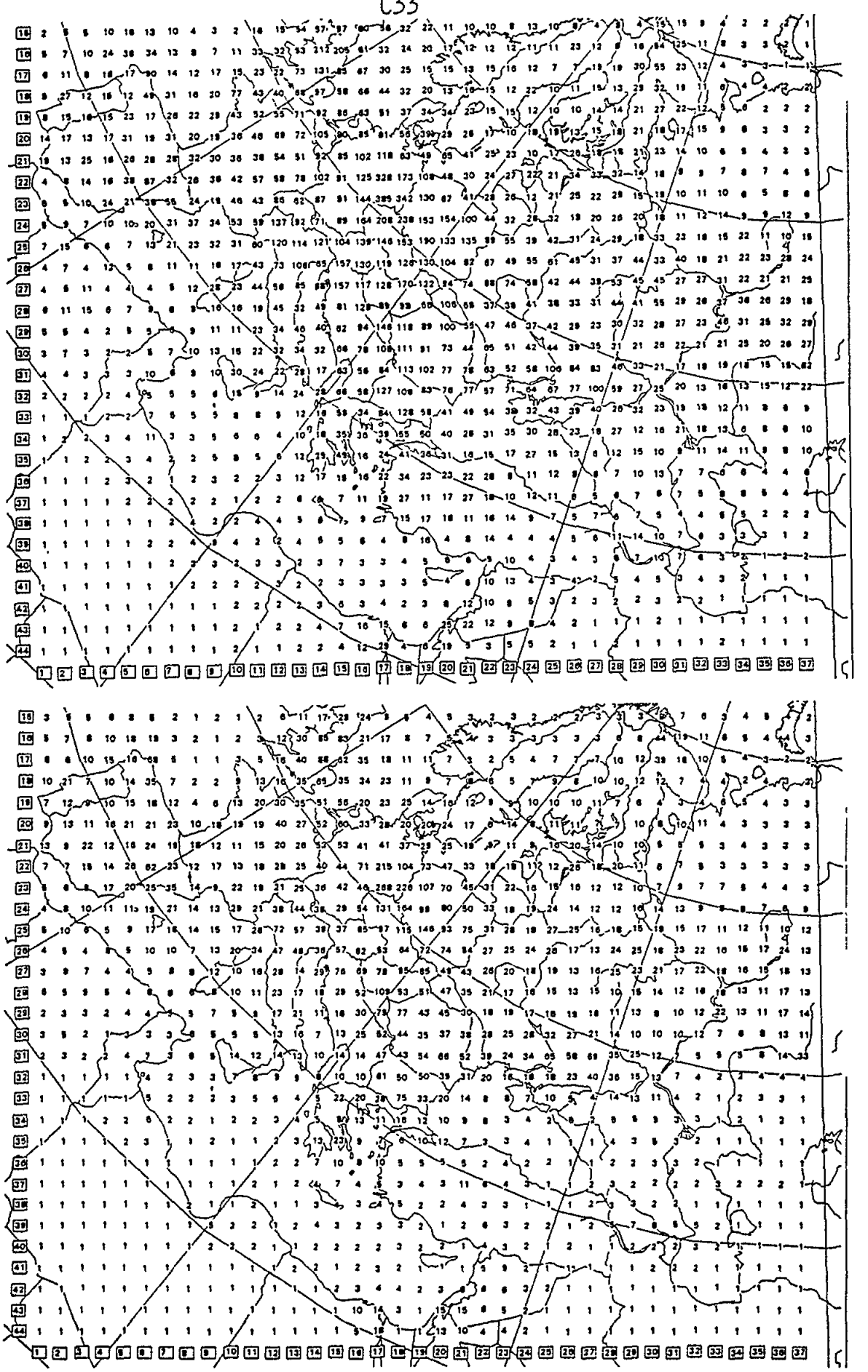


Fig.C33 Mean monthly SO_2 air concentrations (0.1 ug S/m^3) for January (upper) and April (lower).

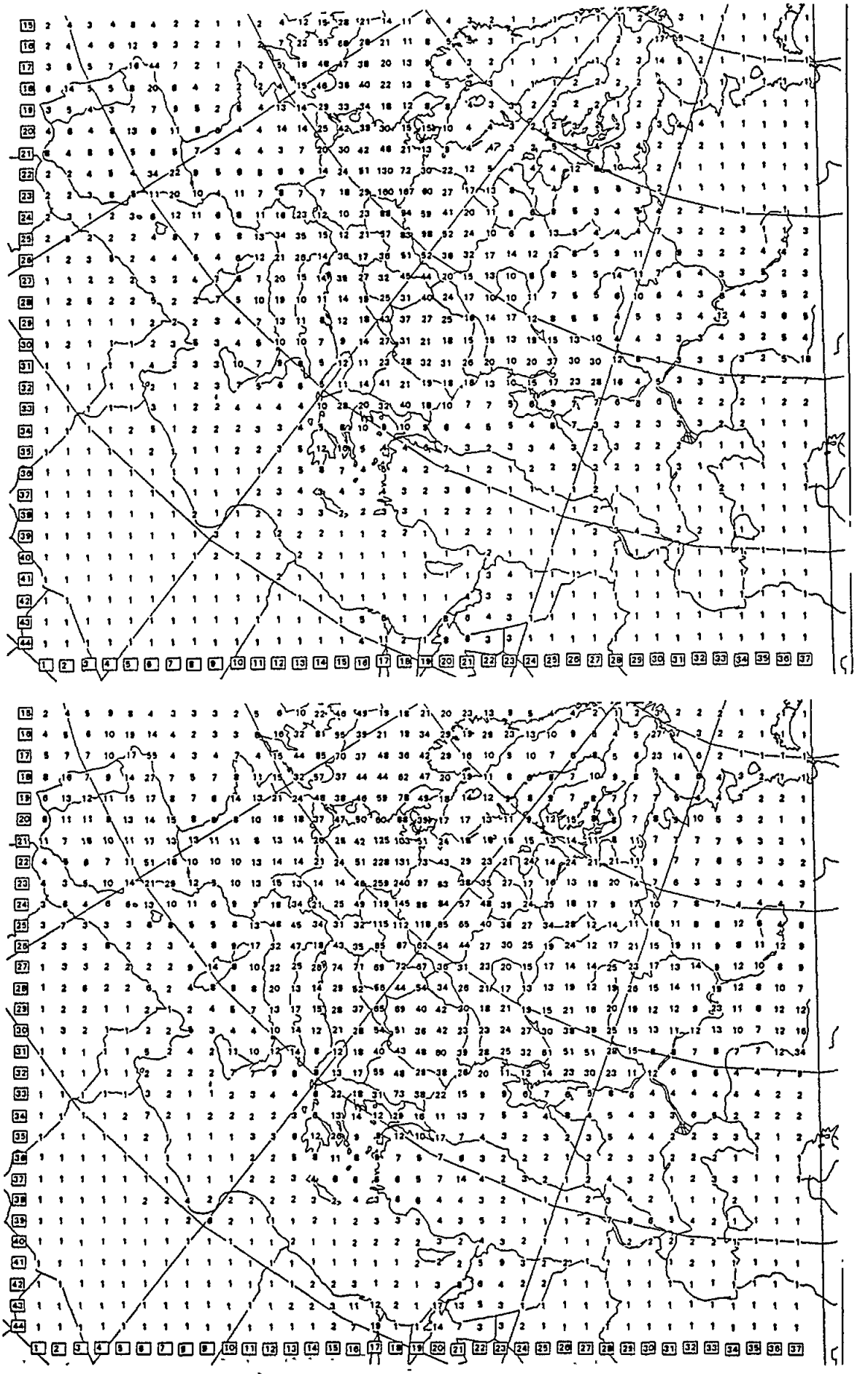


Fig.C34 Mean monthly SO_2 air concentrations (0.1 ug S/m^3) for July (upper) and October (lower).

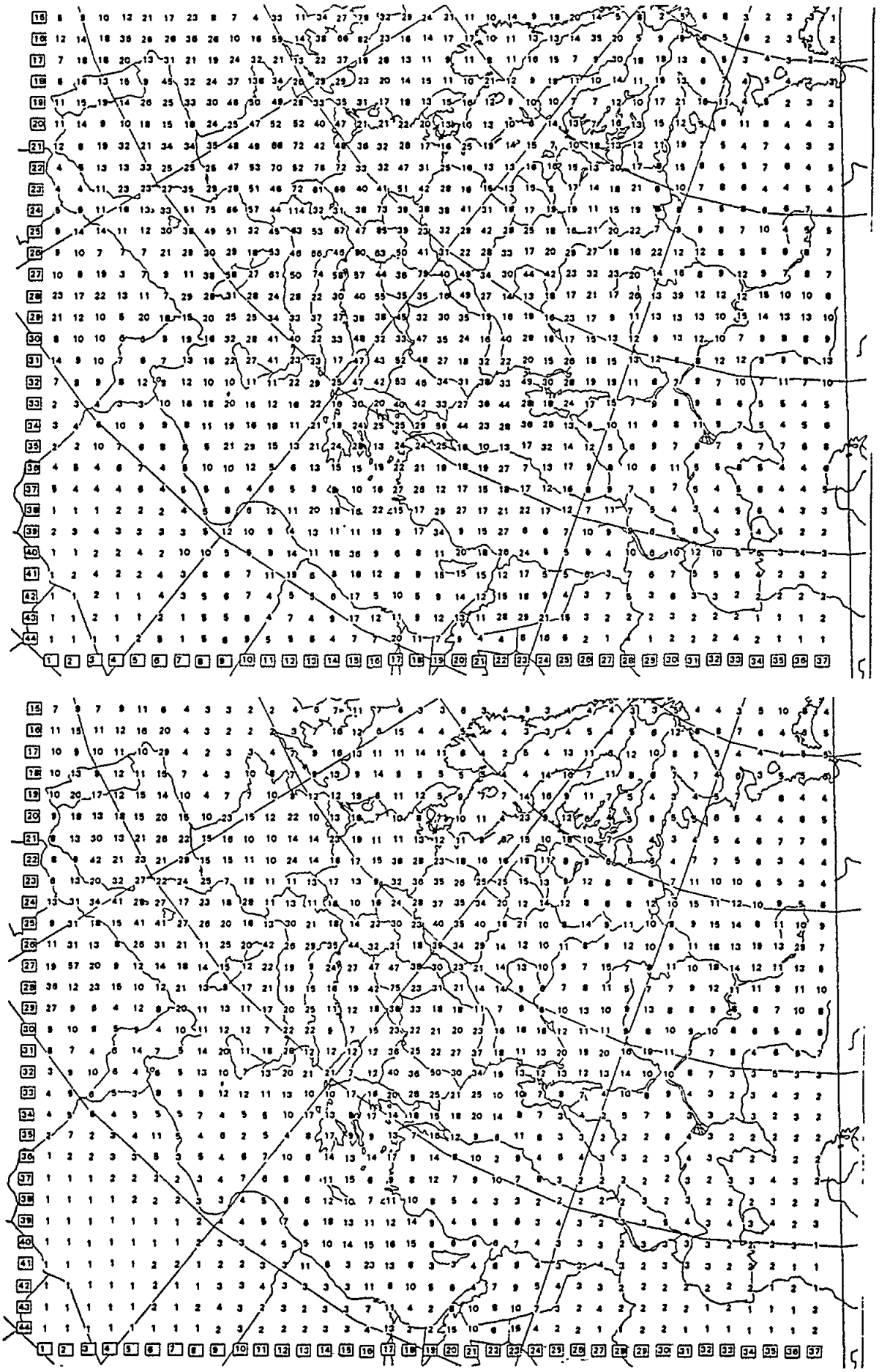


Fig.C35 Mean monthly SO_4 air concentrations (0.1 ug S/m^3) for January (upper) and April (lower).

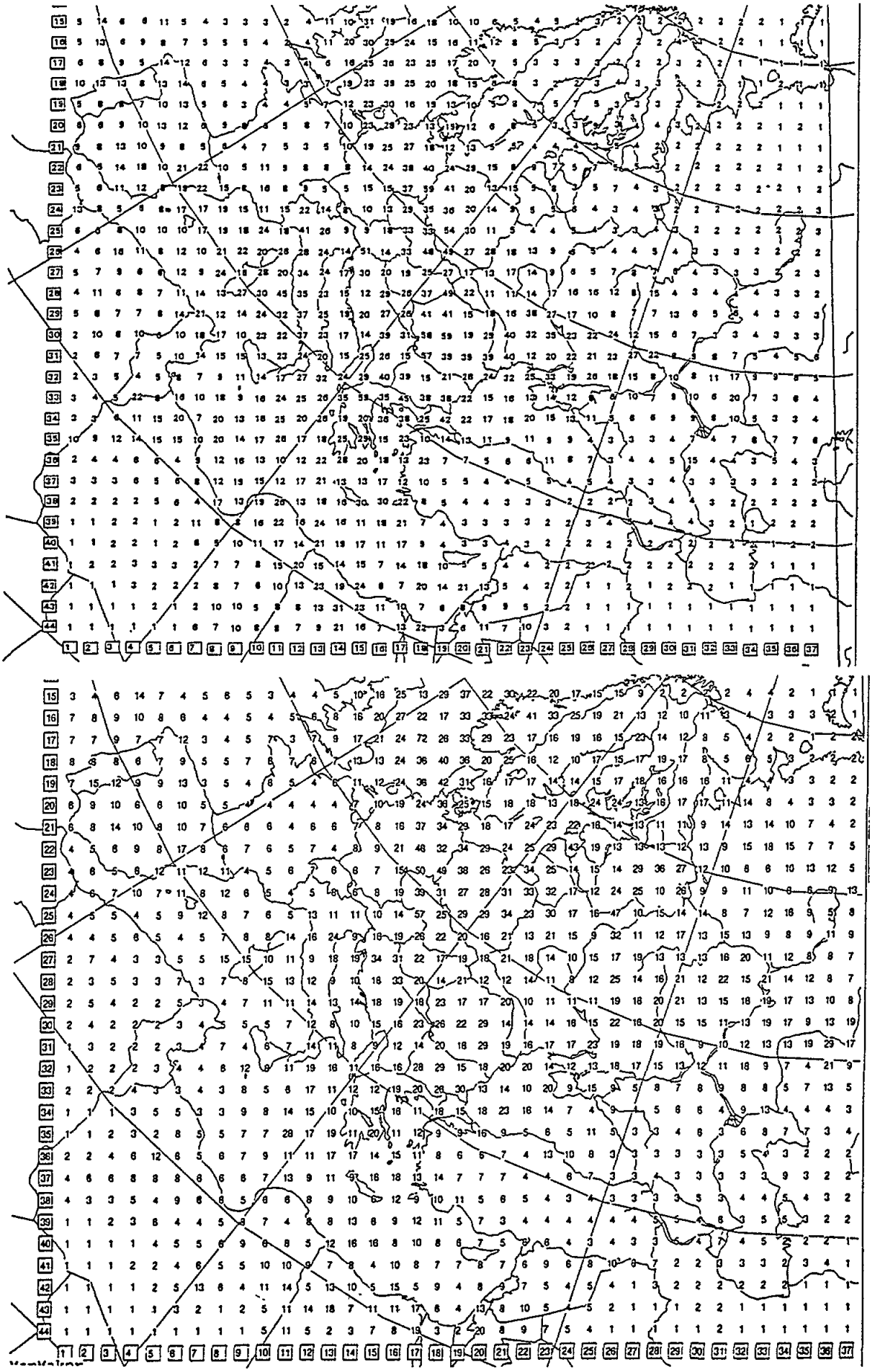


Fig.C36 Mean monthly SO_4 air concentrations (0.1 ug S/m^3) for July (upper) and October (lower).

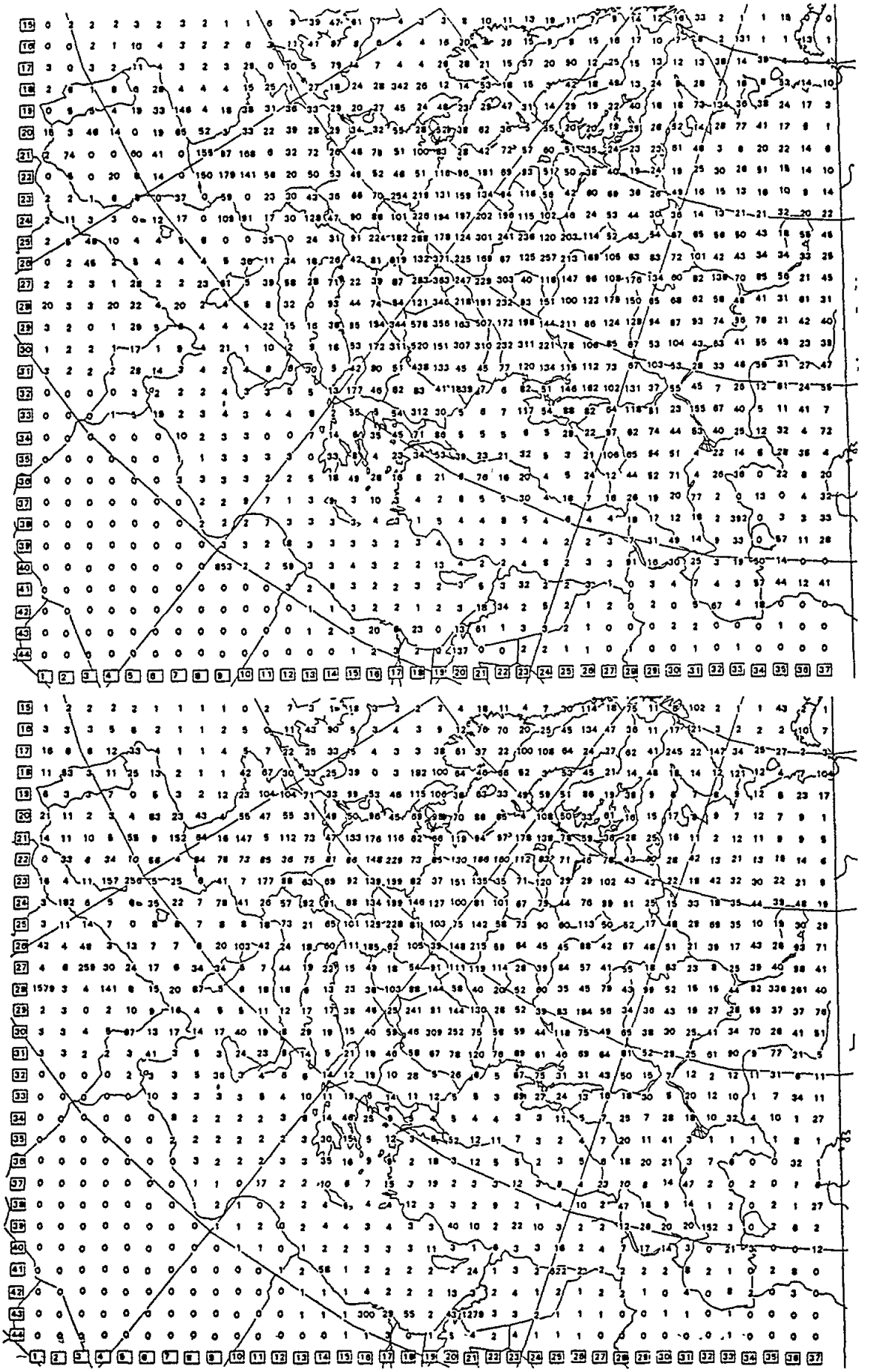


Fig.C37 Mean monthly nitrate concentrations in precipitation (0.01 mg N/l) for January (upper) and April (lower).

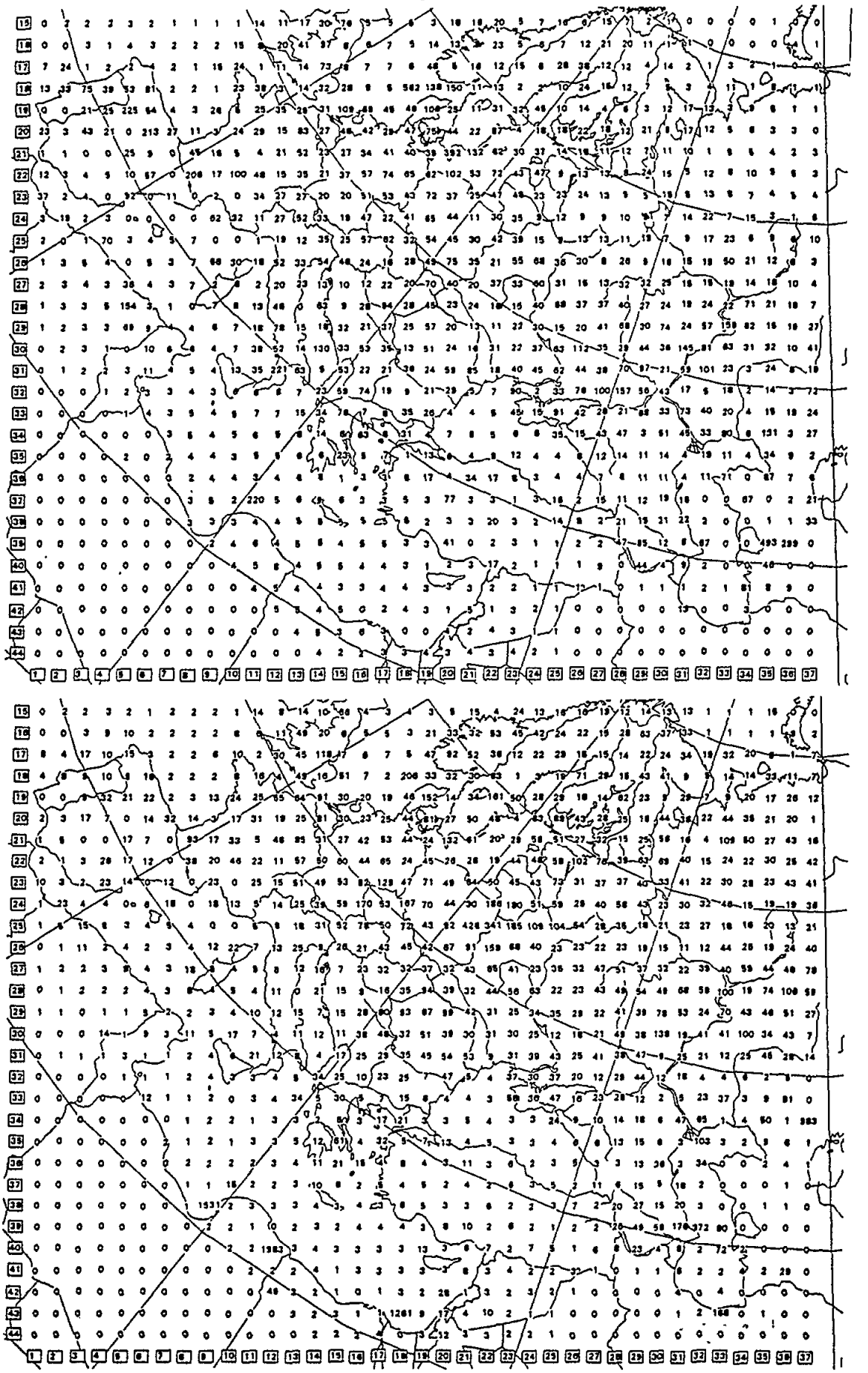


Fig.C38 Mean monthly nitrate concentrations in precipitation (0.01 mg N/l) for July (upper) and October (lower).

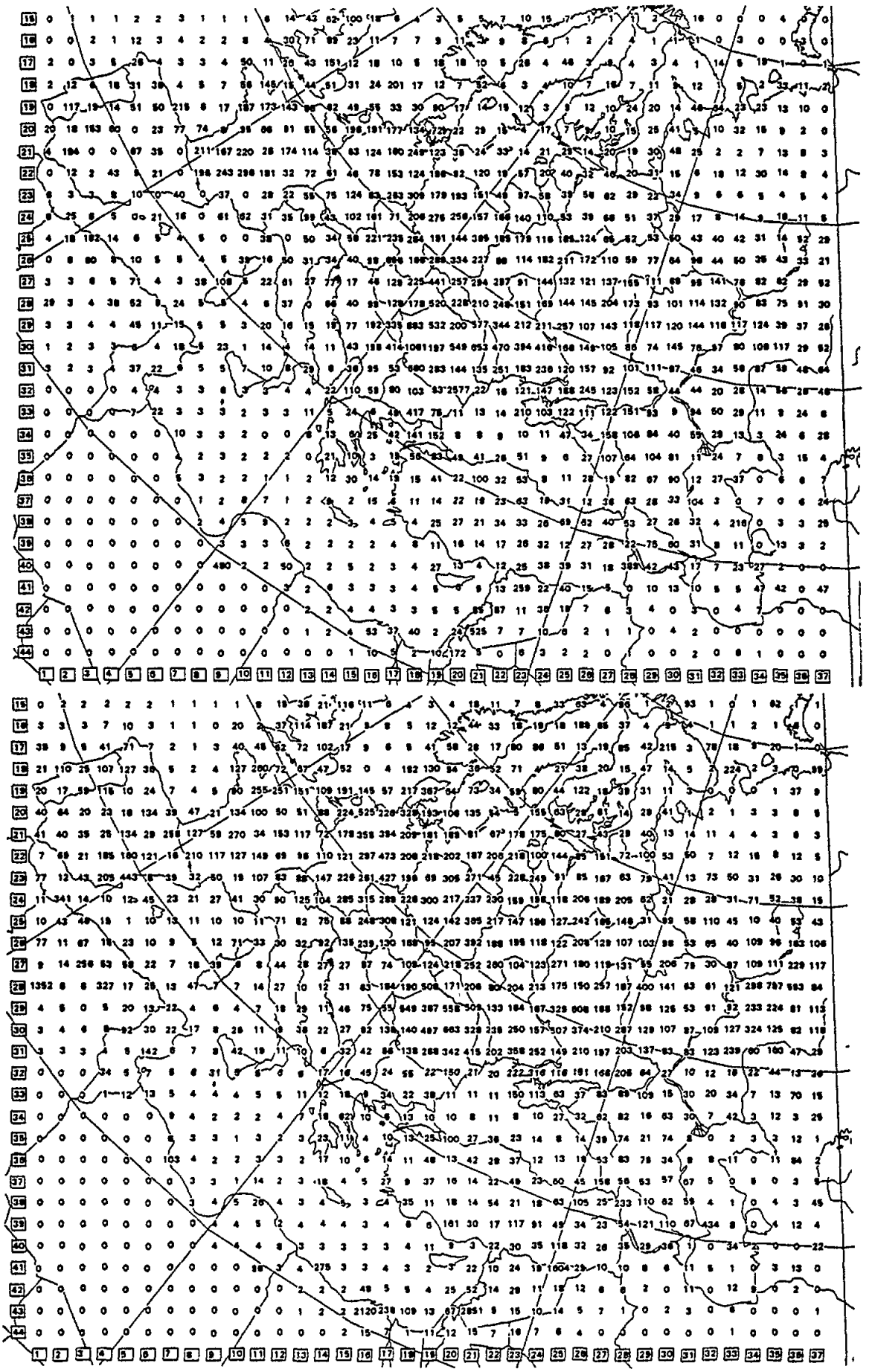


Fig.C39 Mean monthly ammonium concentrations in precipitation (0.01 mg N/l) for January (upper) and April (lower).

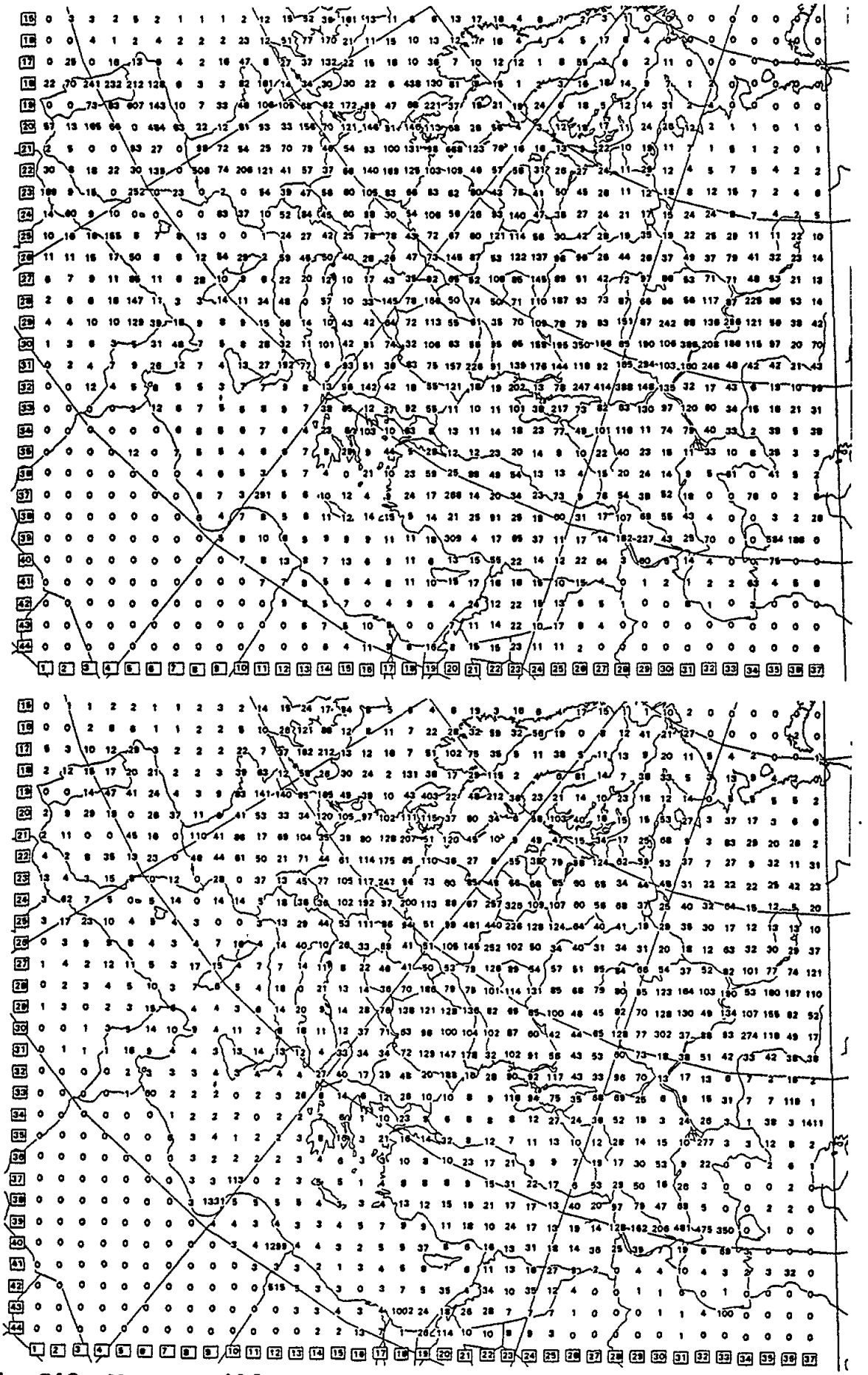


Fig.C40 Mean monthly ammonium concentrations in precipitation (0.01 mg N/l) for July (upper) and October (lower).

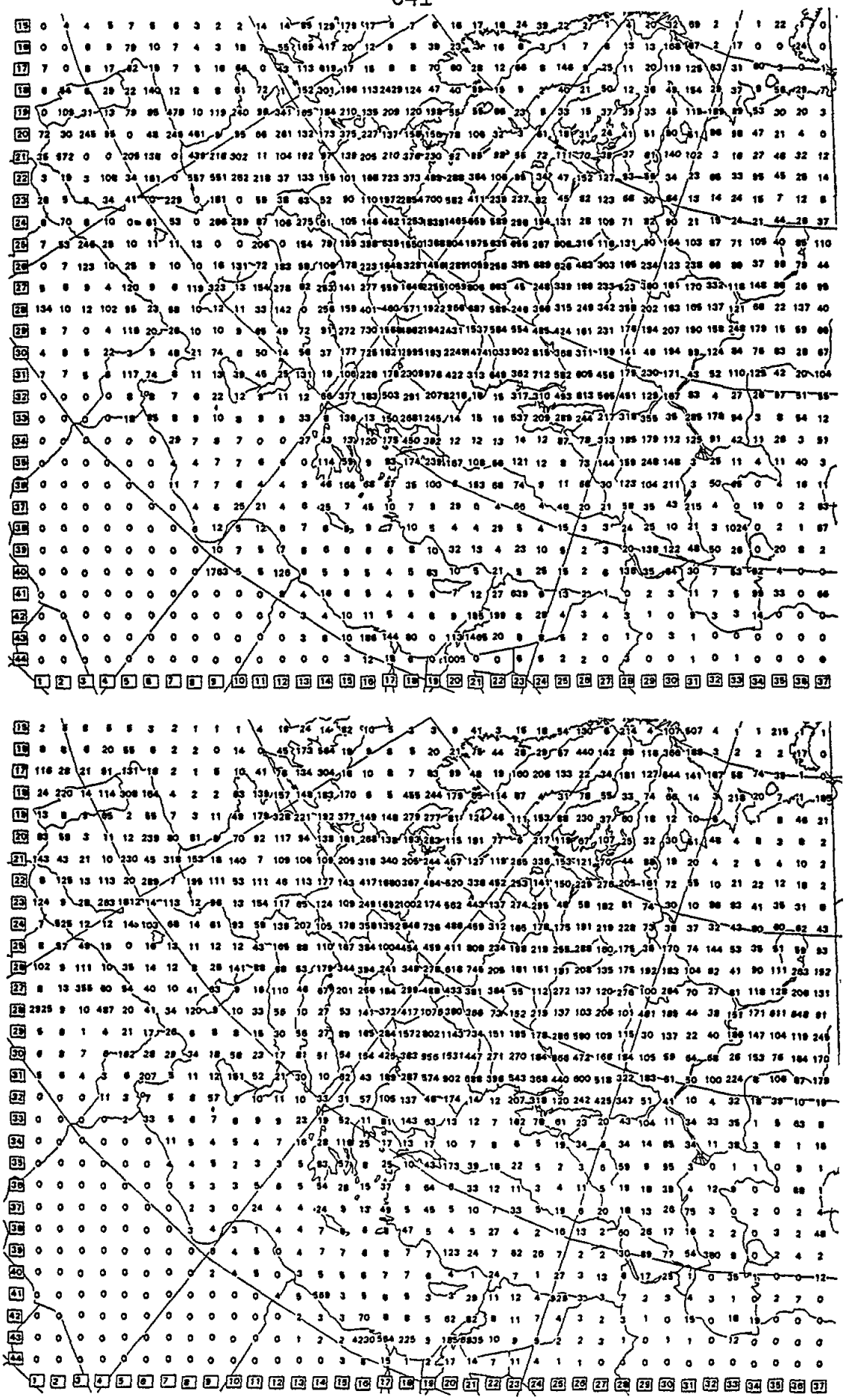


Fig.C41 Mean monthly sulphate concentrations in precipitation (0.01 mg S/l) for January (upper) and April (lower).

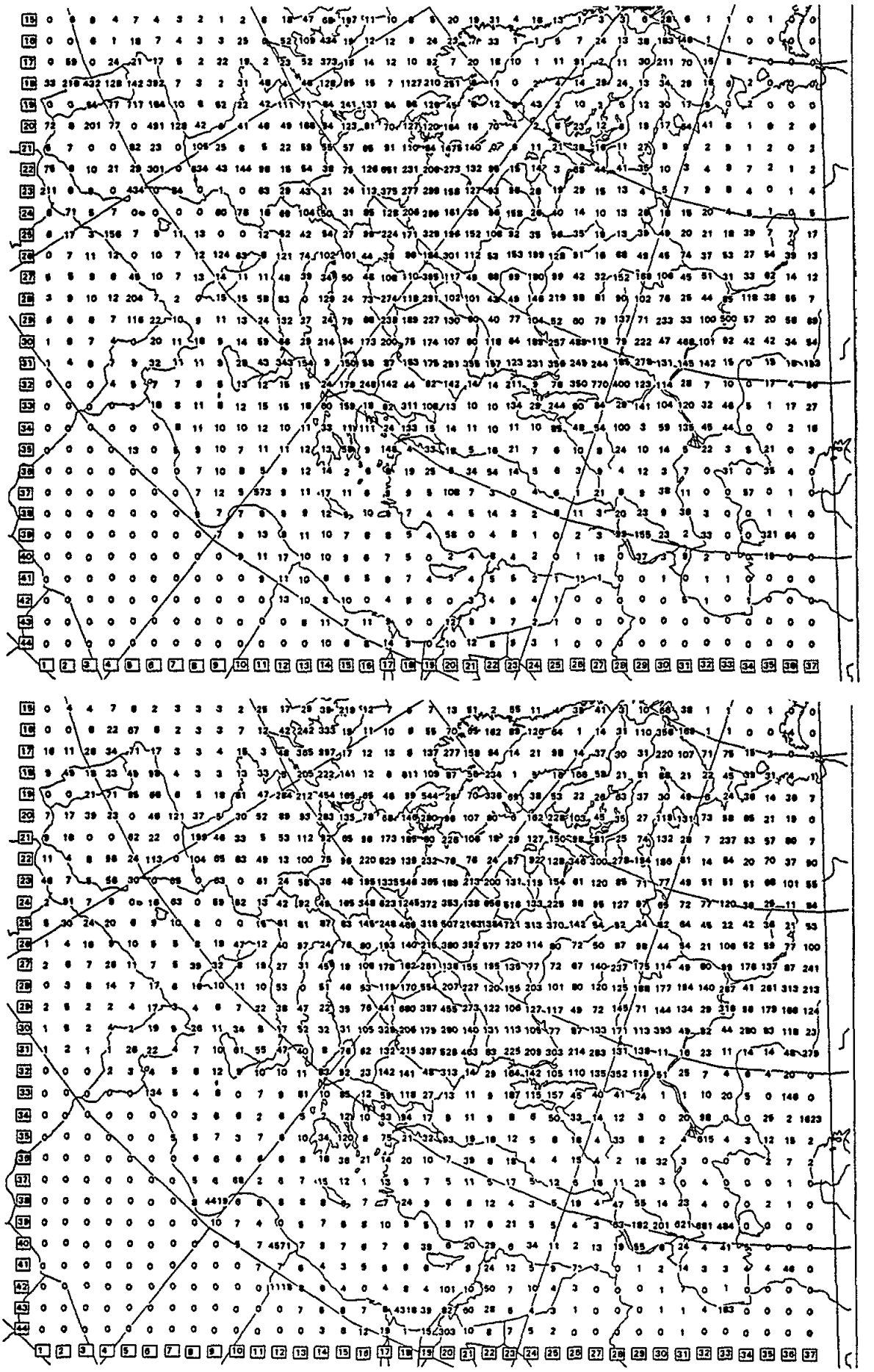


Fig.C42 Mean monthly sulphate concentrations in precipitation (0.01 mg S/l) for July (upper) and October (lower).

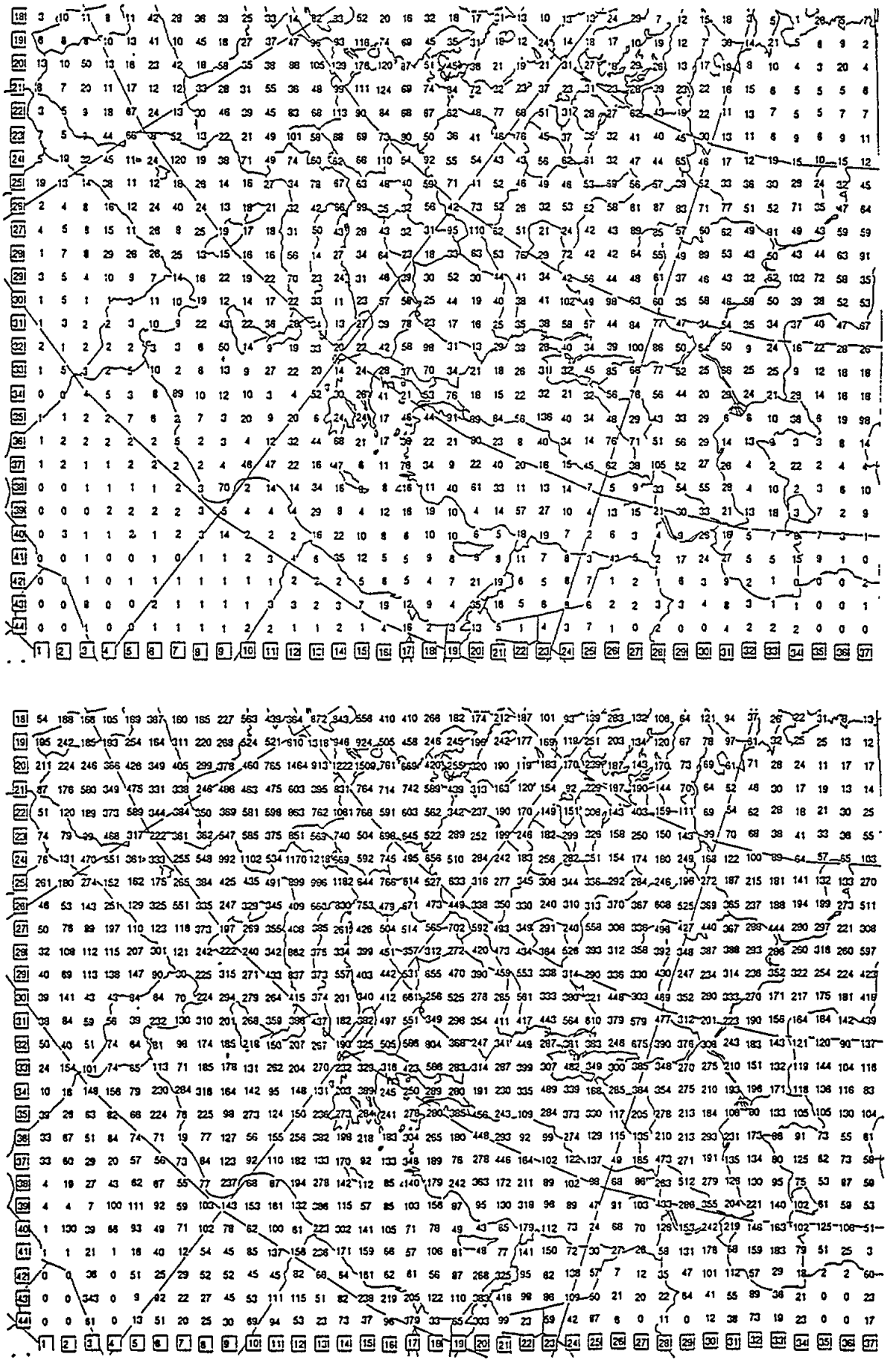


Fig. C43. Lead total deposition (10 ug Pb/m^2) - upper panel, concentration in air (0.1 ng Pb/m^3) - lower panel, January 1991.

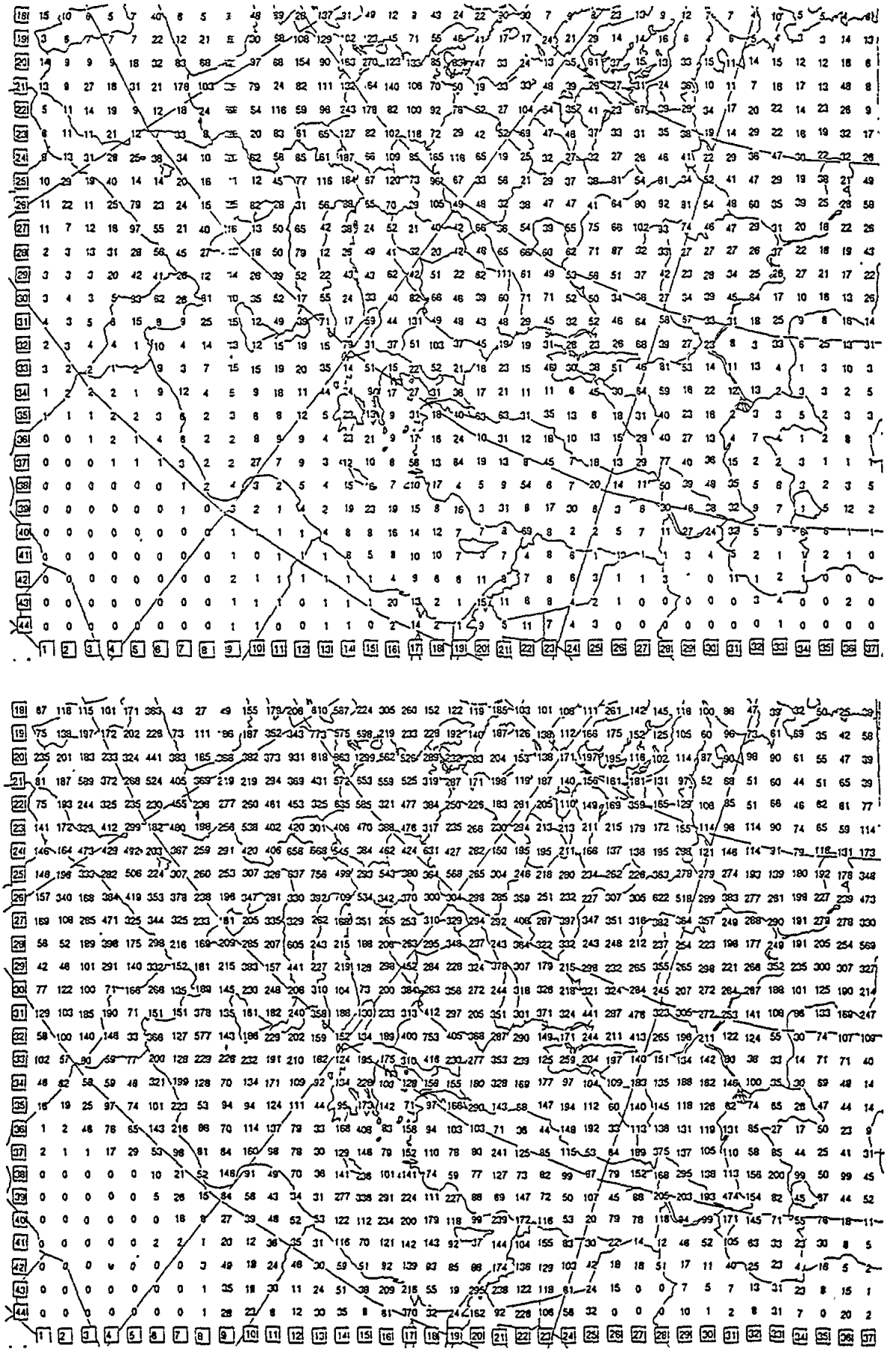


Fig. C44. Lead total deposition (10 ug Pb/m^2) - upper panel, concentration in air (0.1 ng Pb/m^3) - lower panel, April 1991.

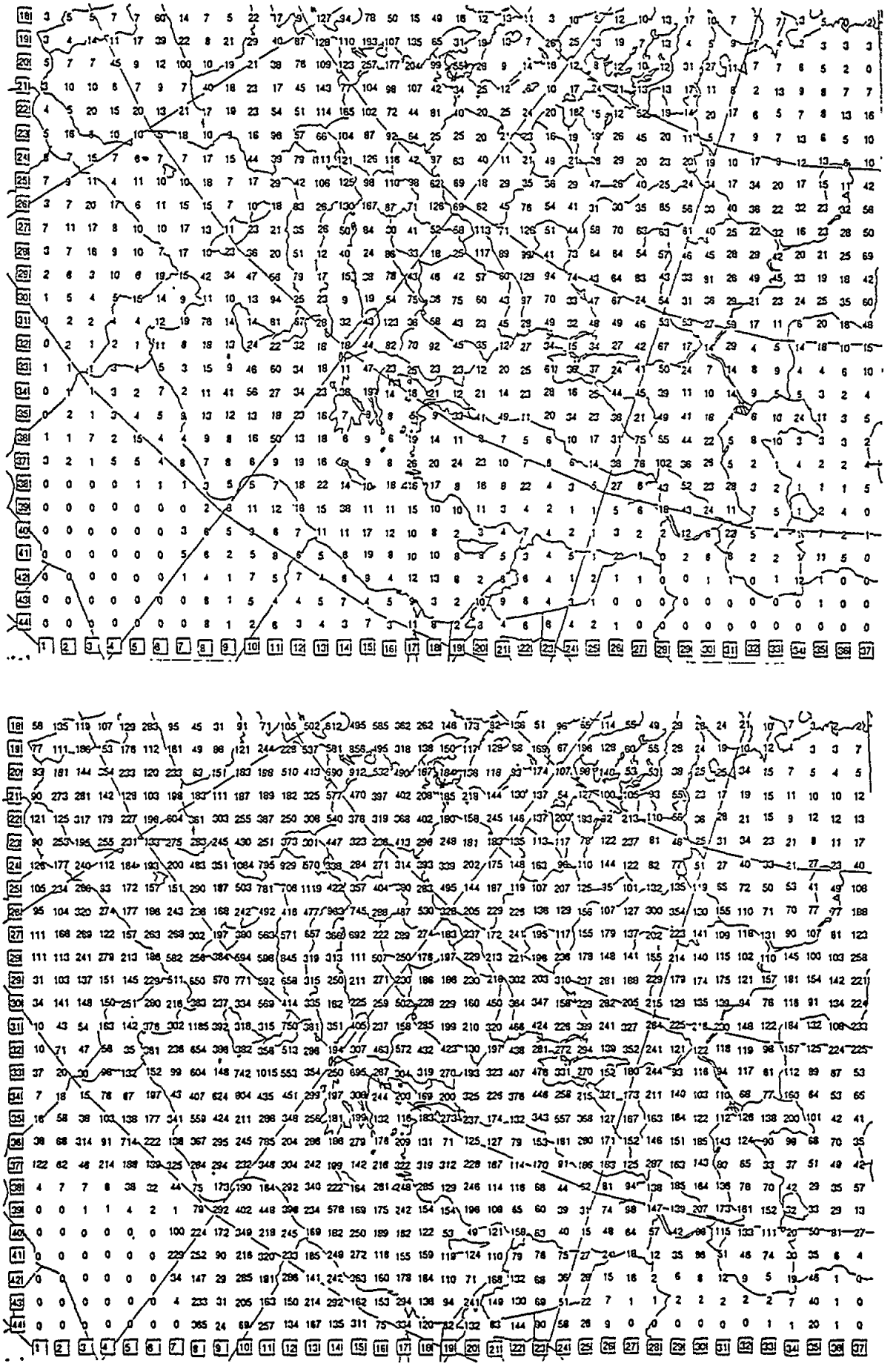


Fig. C45. Lead total deposition (10 ug Pb/m^2) - upper panel, concentration in air (0.1 ng Pb/m^3) - lower panel, July 1991.

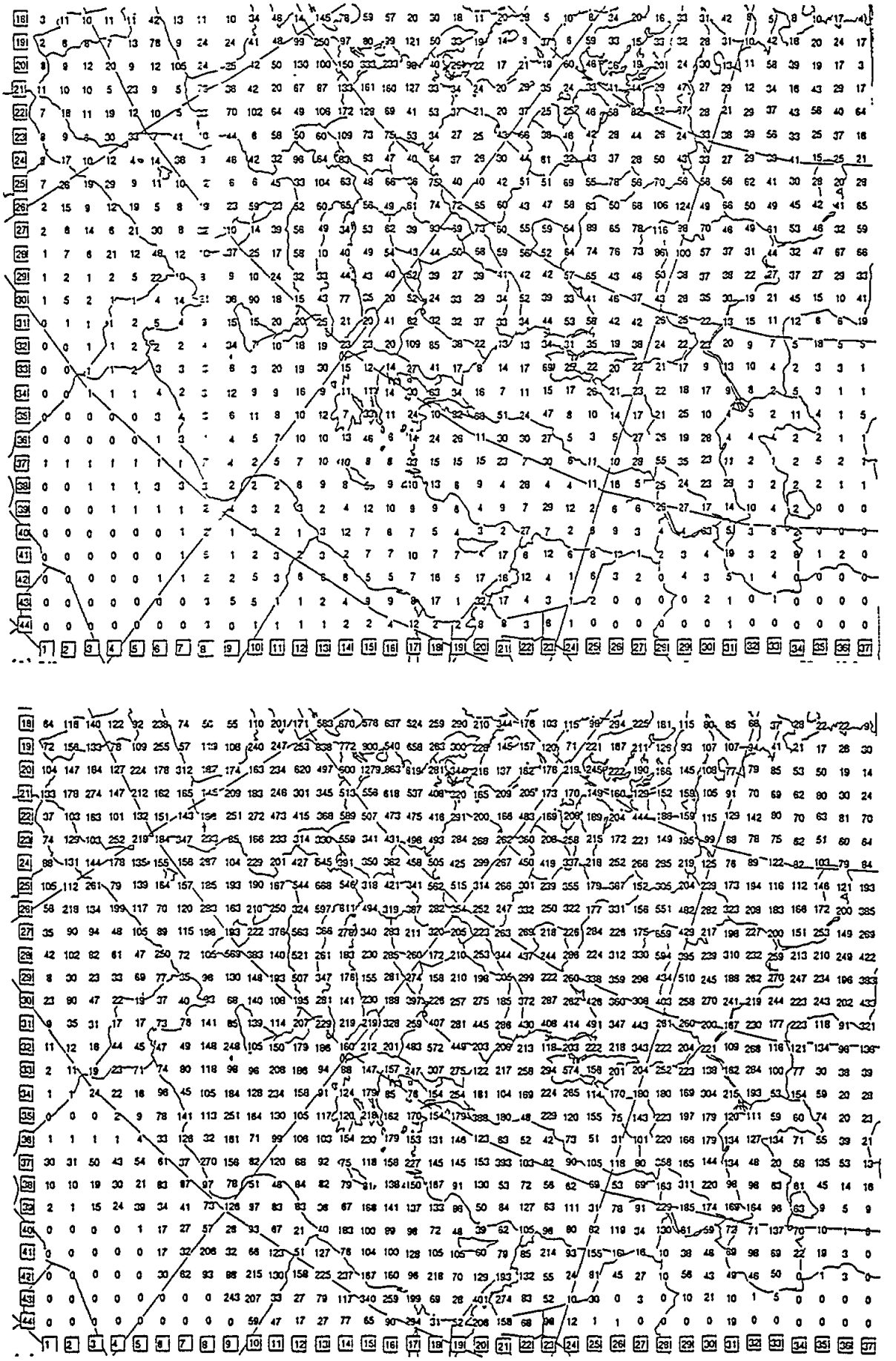


Fig. C46. Lead total deposition (10 ug Pb/m^2) - upper panel, concentration in air (0.1 ng Pb/m^3) - lower panel, October 1991.

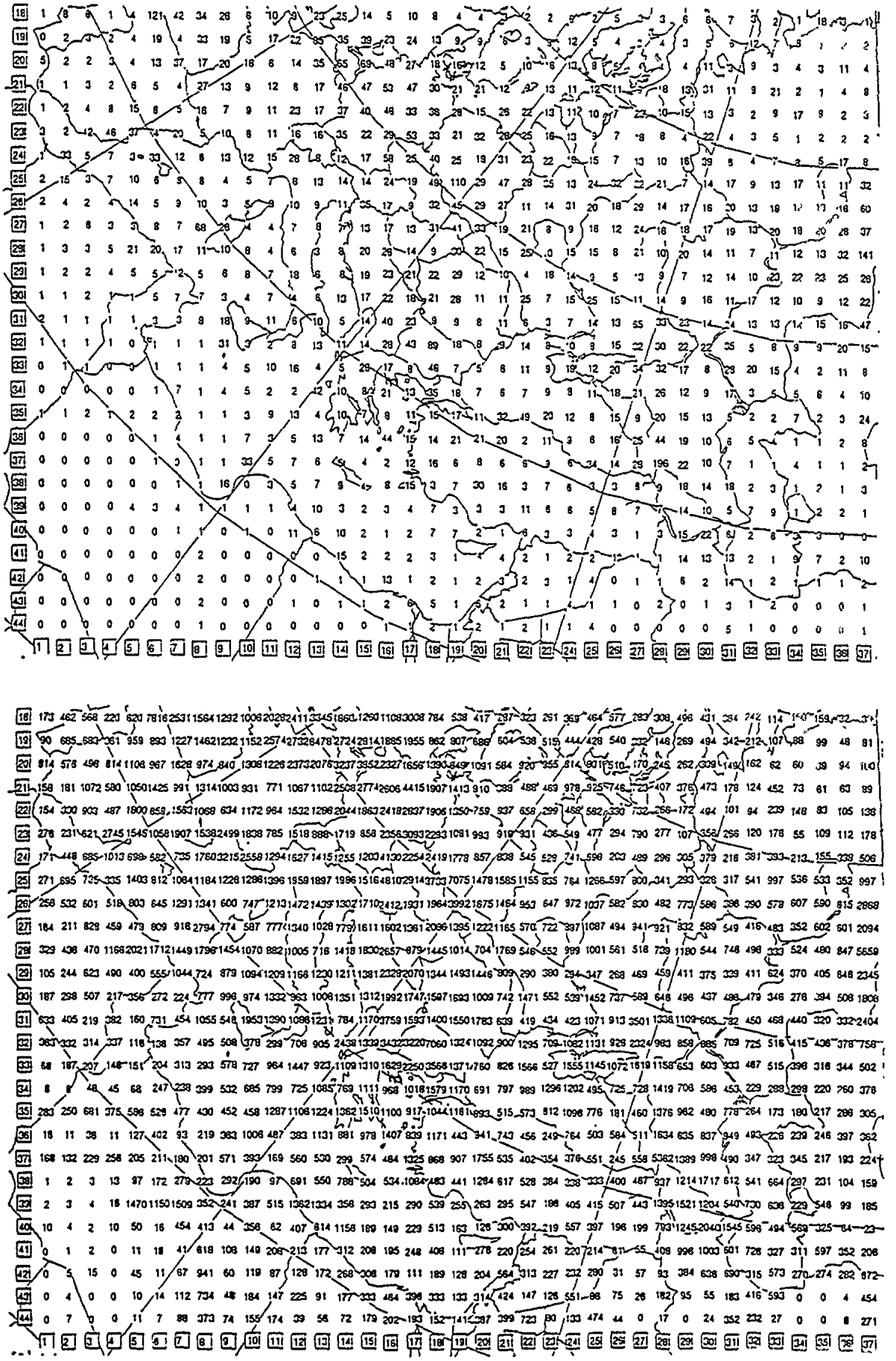


Fig. C47. Zinc total deposition (10 ug Zn/m^2) - upper panel, concentration in air (0.01 ng Zn/m^3) - lower panel, January 1991.

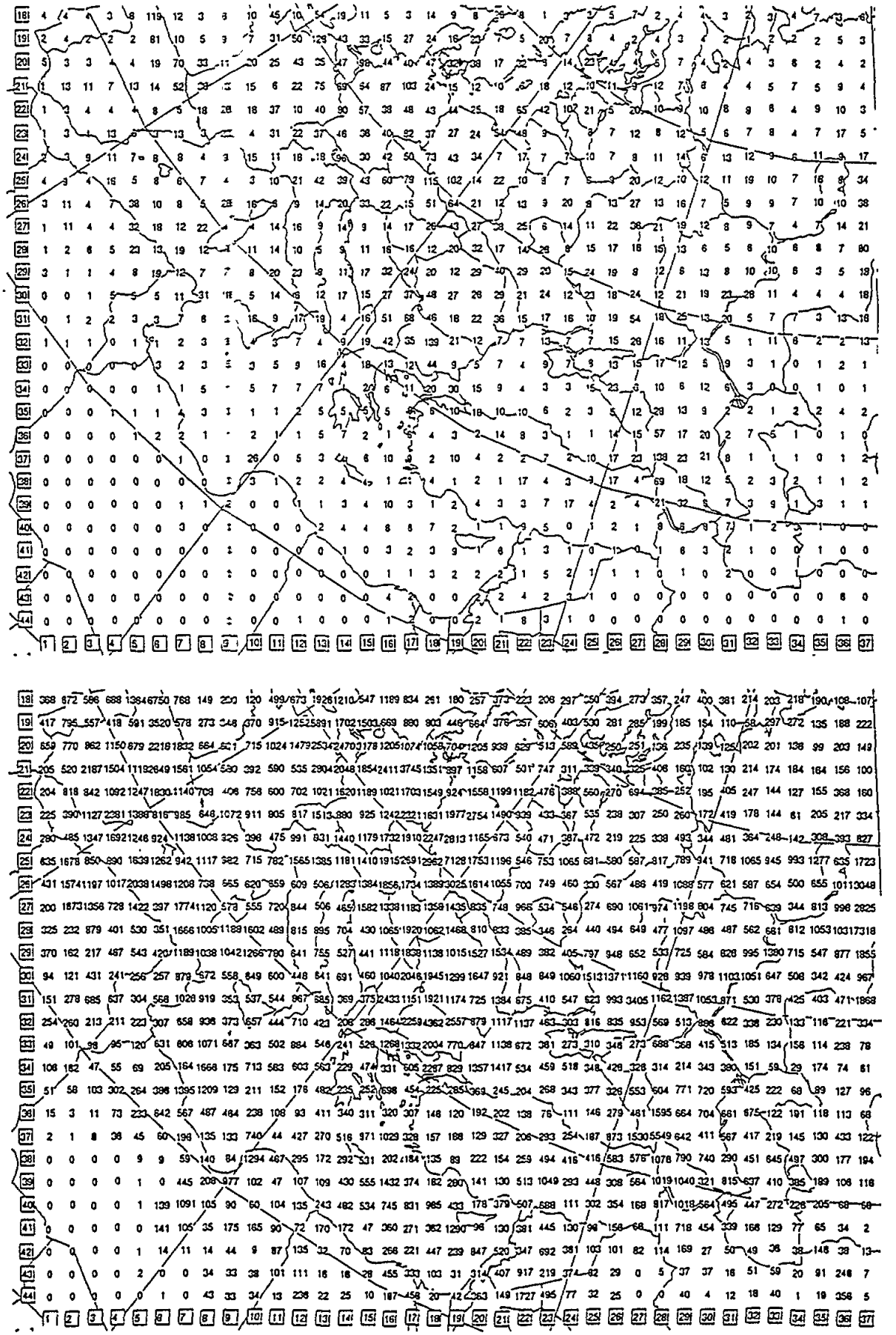


Fig. C48. Zinc total deposition (10 ug Zn/m^2) - upper panel, concentration in air (0.01 ng Zn/m^3) - lower panel, April 1991.

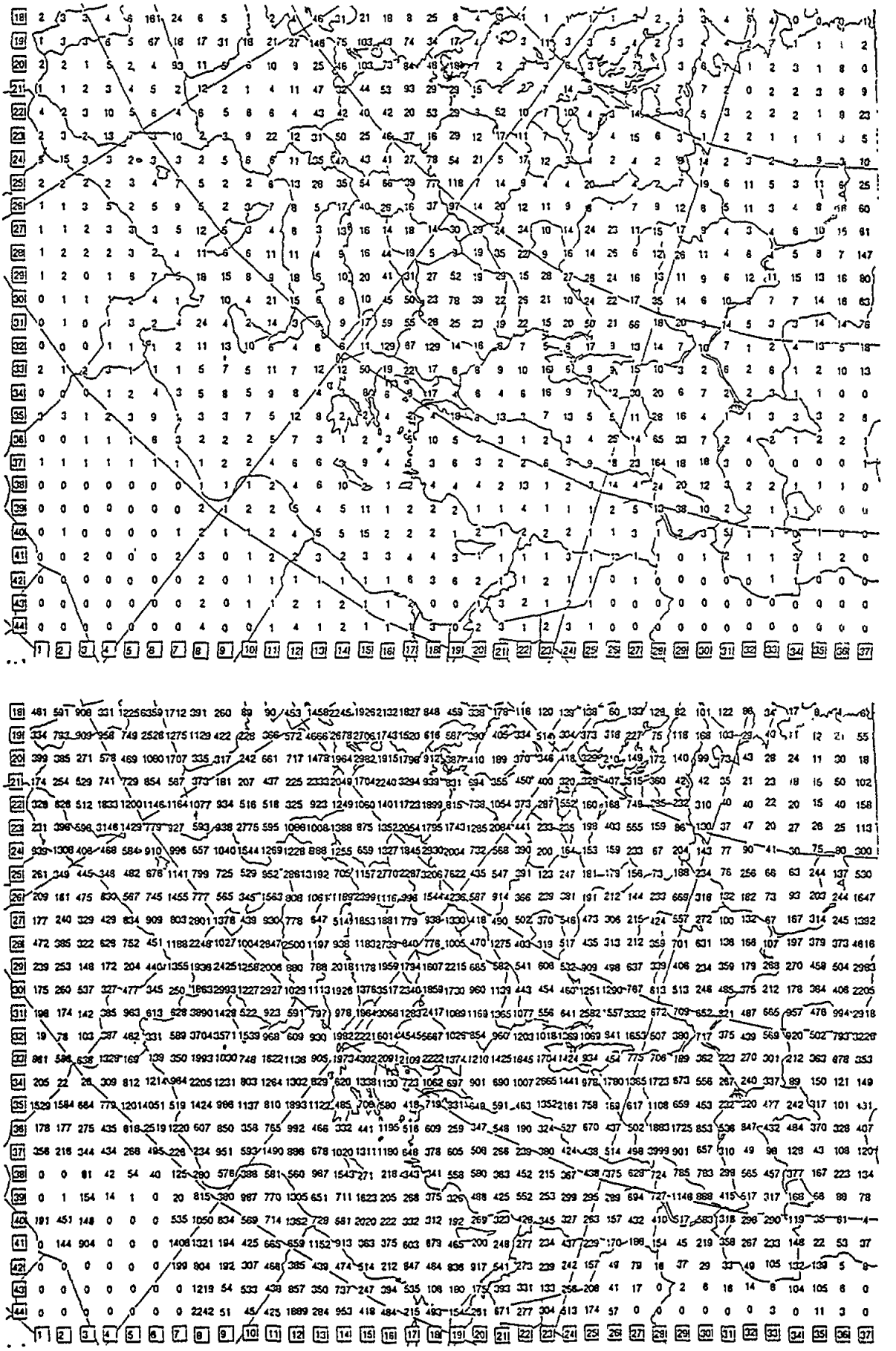


Fig. C49. Zinc total deposition (10 ug Zn/m^2) - upper panel, concentration in air (0.01 ng Zn/m^3) - lower panel, July 1991.

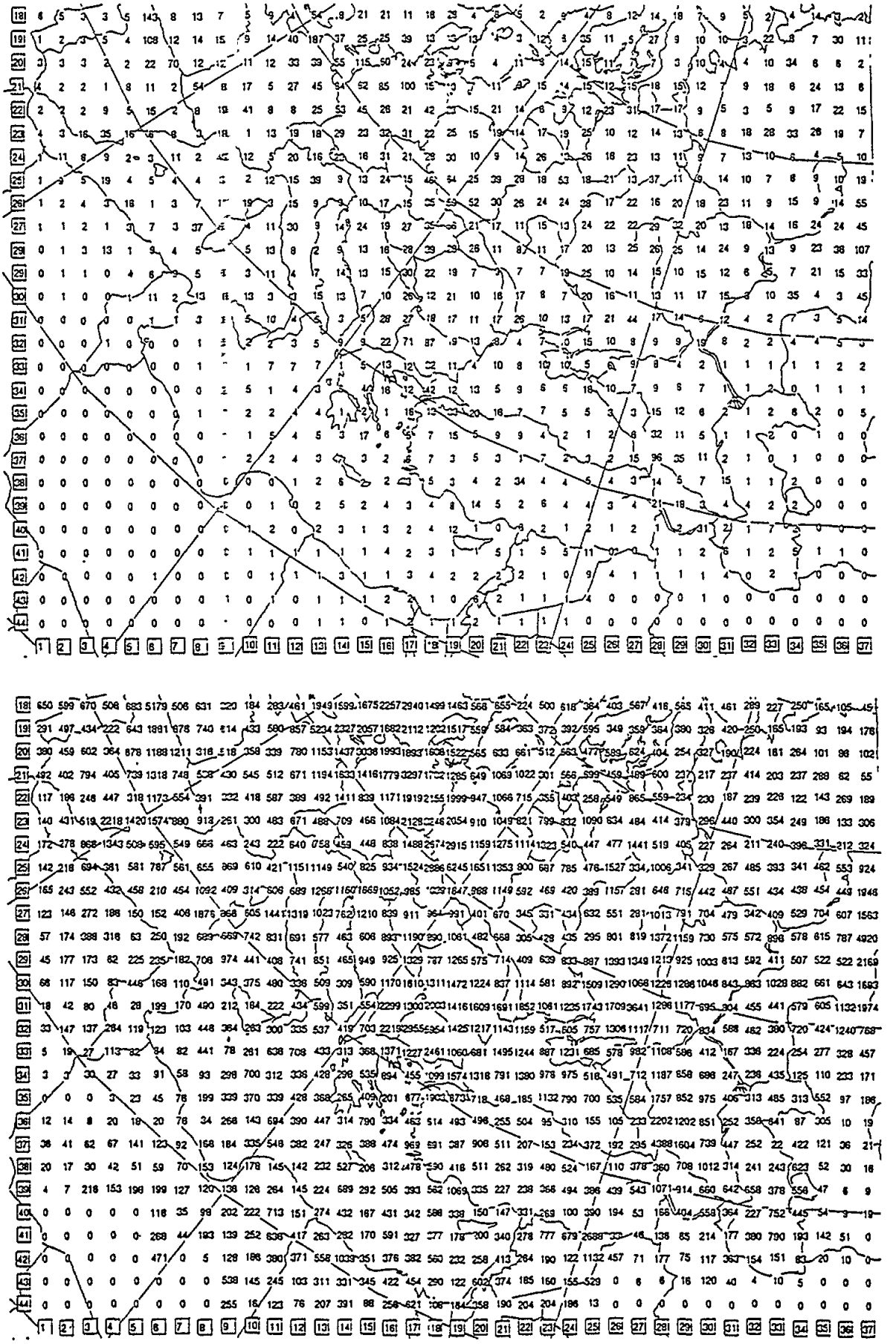


Fig. C50. Zinc total deposition (10 ug Zn/m^2) - upper panel, concentration in air (0.01 ng Zn/m^3) - lower panel, October 1991.

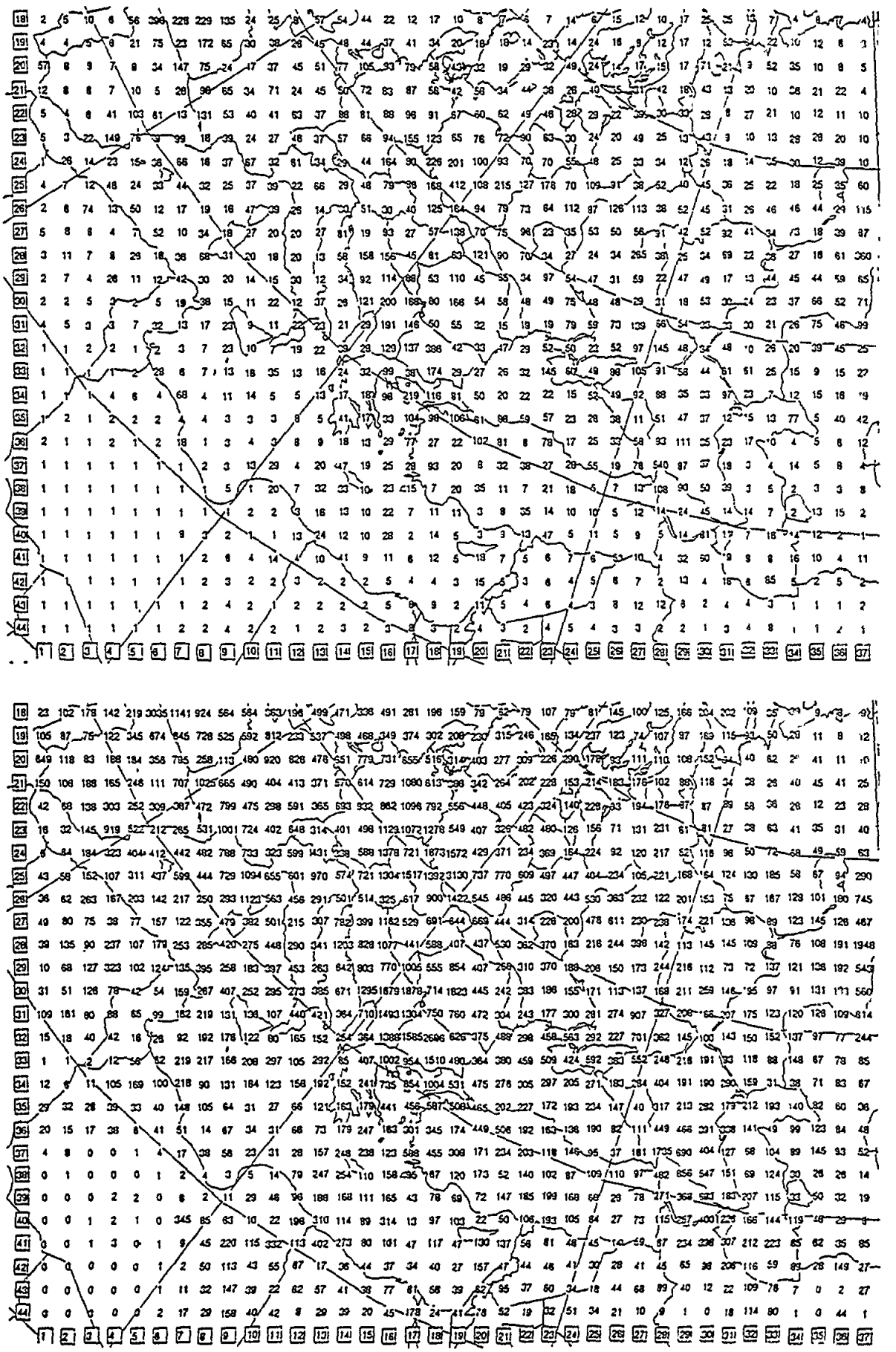


Fig. C51. Cadmium total deposition (0.1 ug Cd/m^2) - upper panel, concentration in air (0.001 ng Cd/m^3) - lower panel, January 1991.

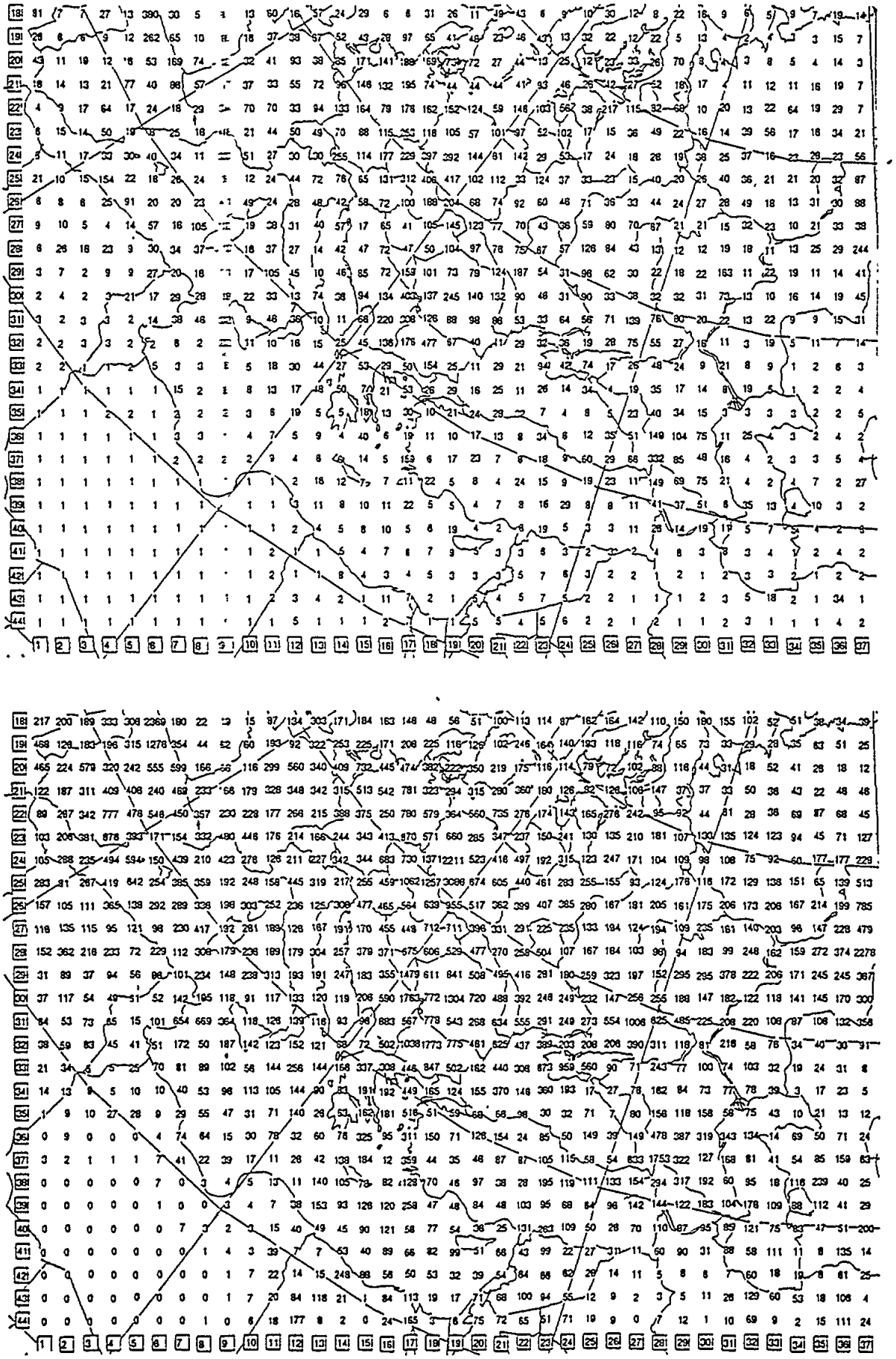


Fig. C52. Cadmium total deposition (0.1 ug Cd/m^2) - upper panel, concentration in air (0.001 ng Cd/m^3) - lower panel, April 1991.

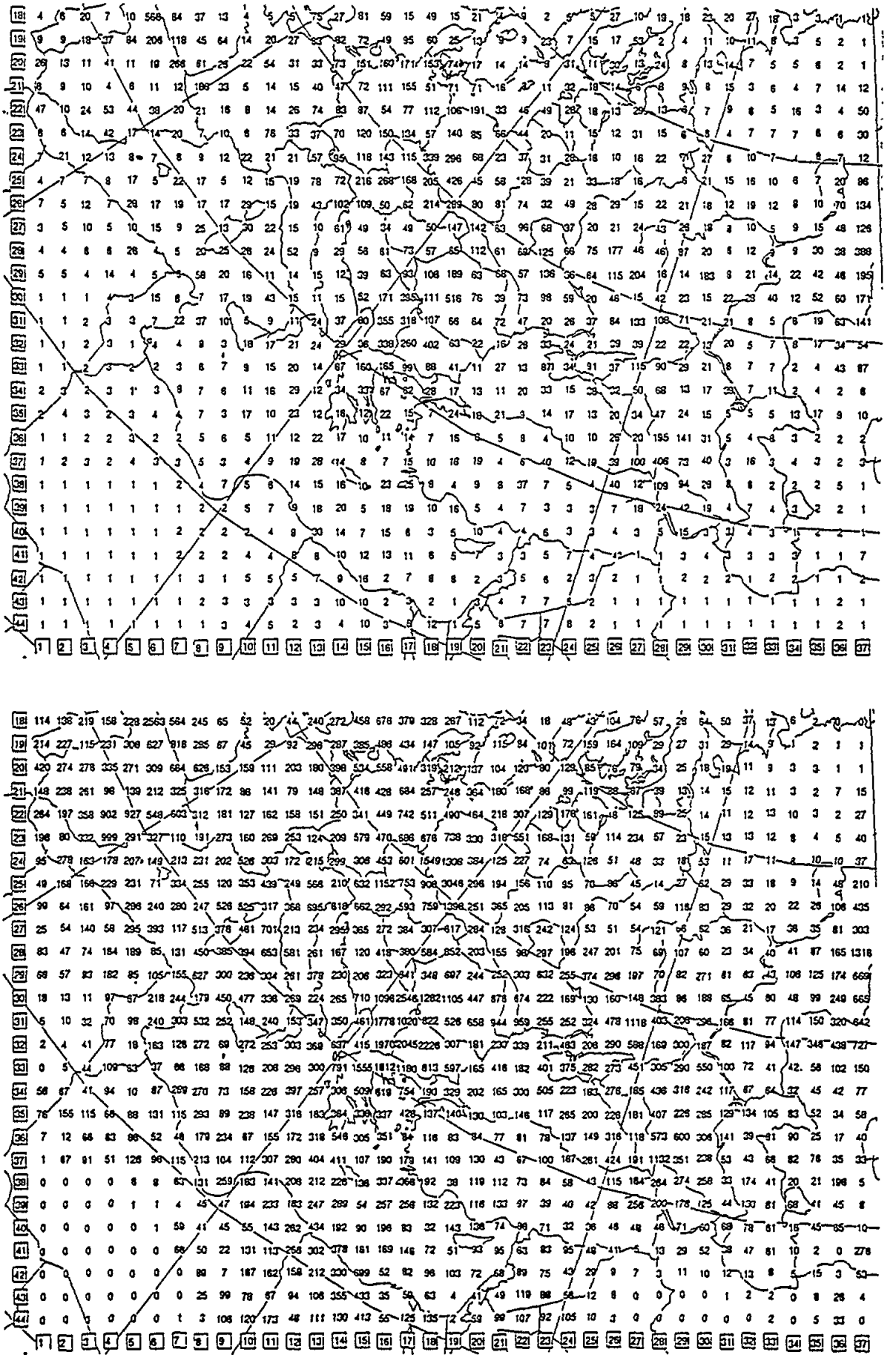


Fig. C53. Cadmium total deposition (0.1 ug Cd/m^2) - upper panel, concentration in air (0.001 ng Cd/m^3) - lower panel, July 1991.

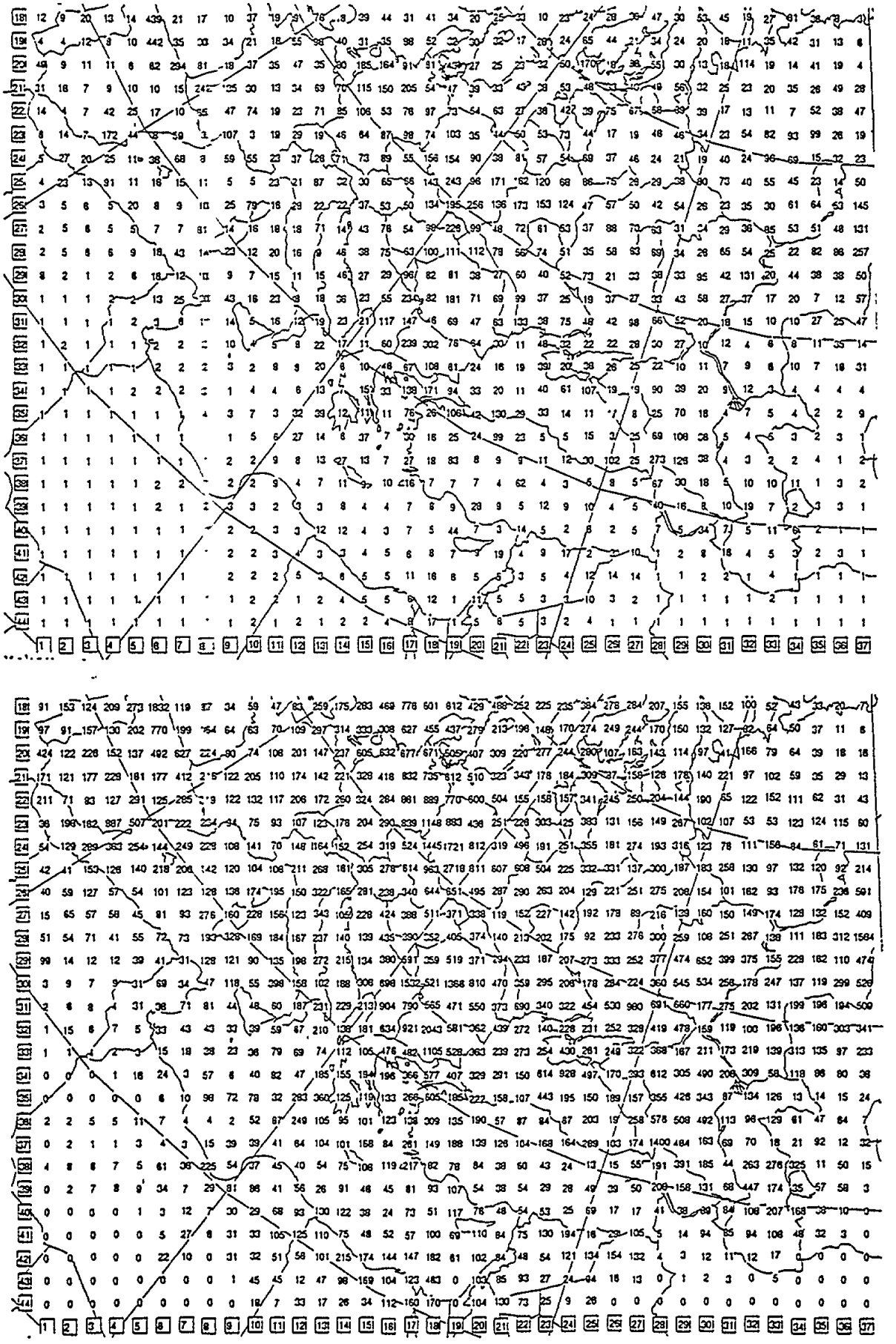


Fig. C54. Cadmium total deposition (0.1 ug Cd/m^2) - upper panel, concentration in air (0.001 ng Cd/m^3) - lower panel, October 1991.

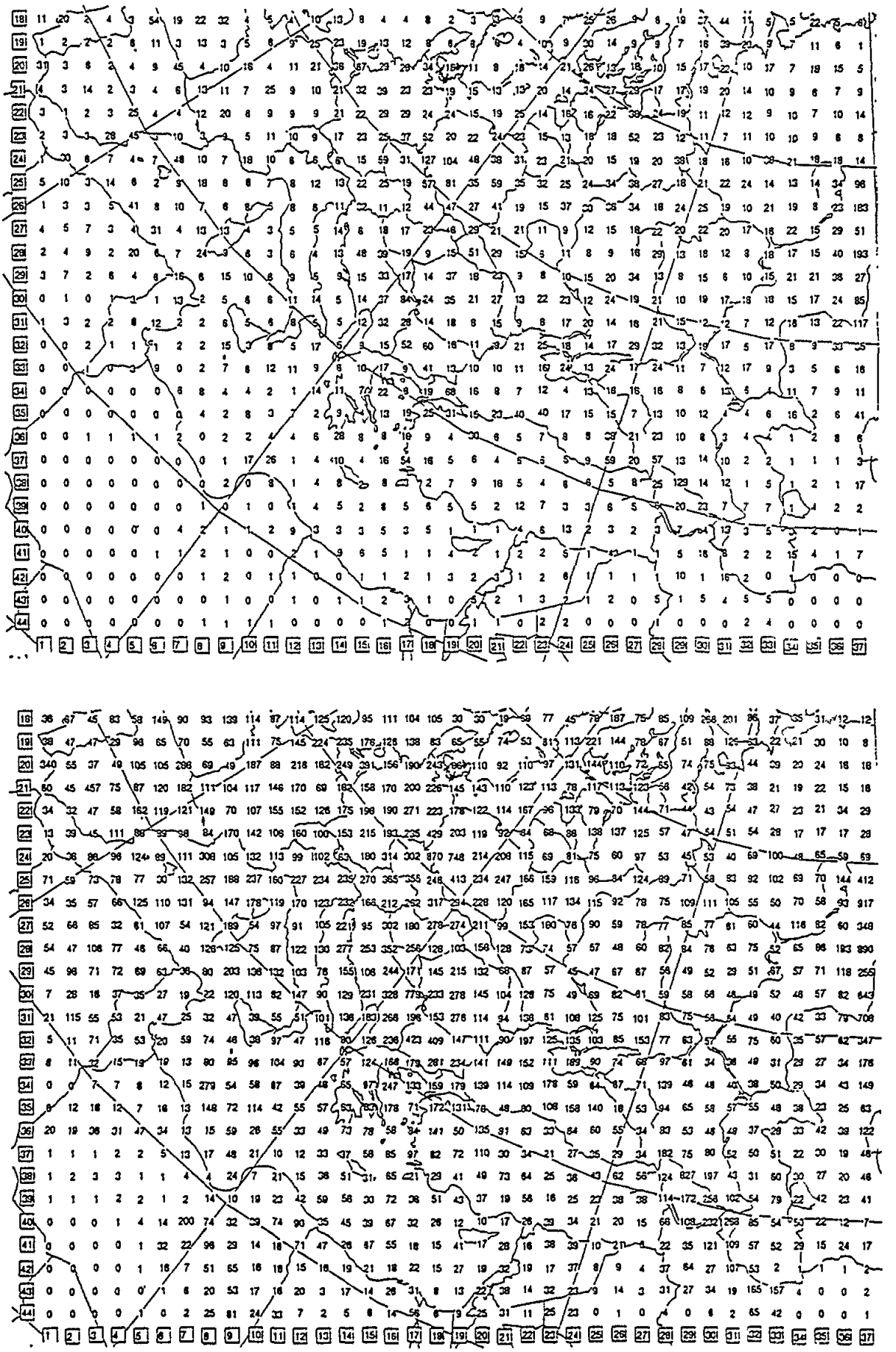


Fig. C55. Arsenic total deposition (1 ug As/m^2) - upper panel, concentration in air (0.01 ng As/m^3) - lower panel, January 1991.

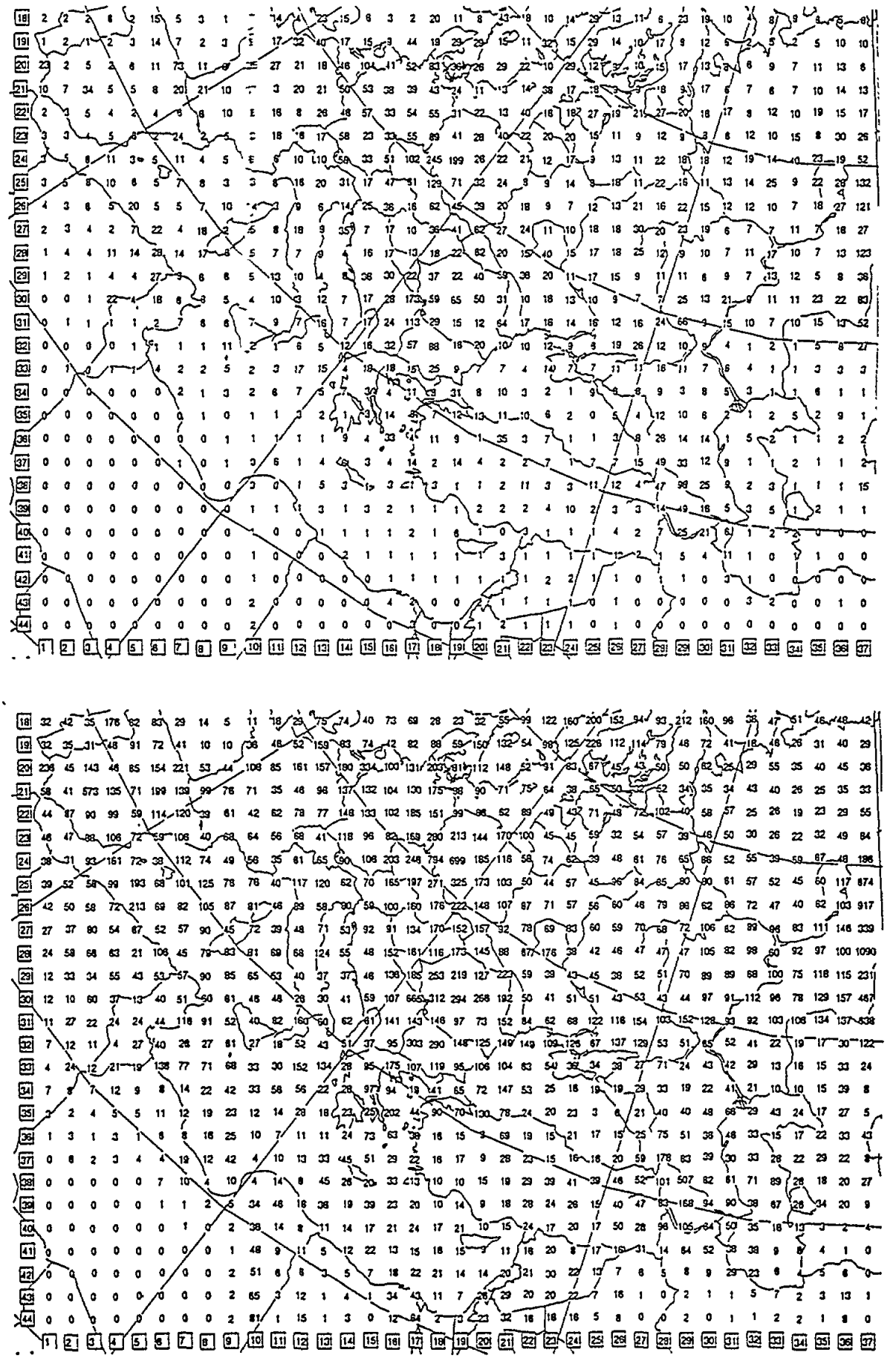


Fig. C56. Arsenic total deposition (1 ug As/m^2) - upper panel, concentration in air (0.01 ng As/m^3) - lower panel, April 1991.

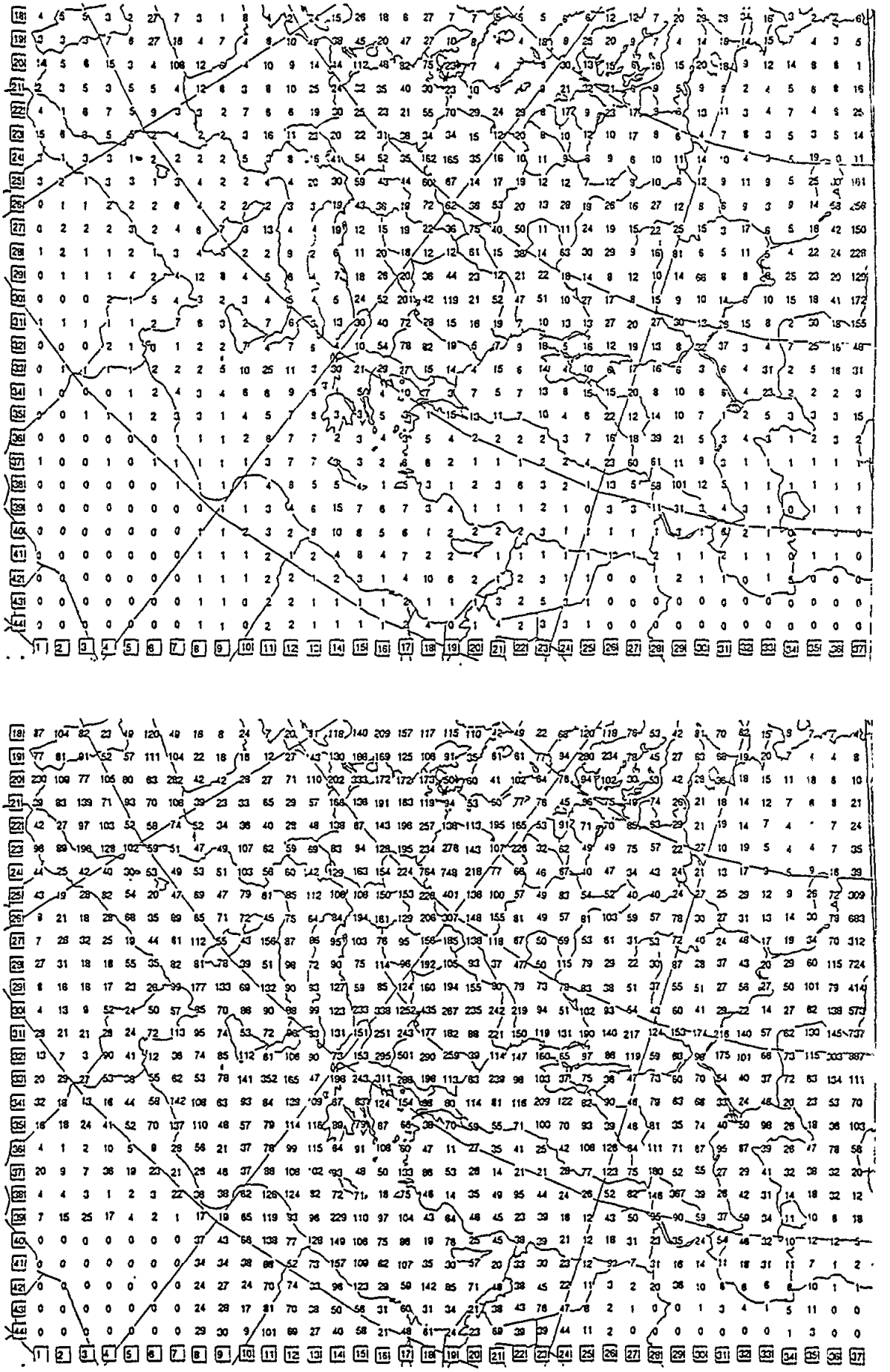


Fig. C57. Arsenic total deposition (1 ug As/m^2) - upper panel, concentration in air (0.01 ng As/m^3) - lower panel, July 1991.

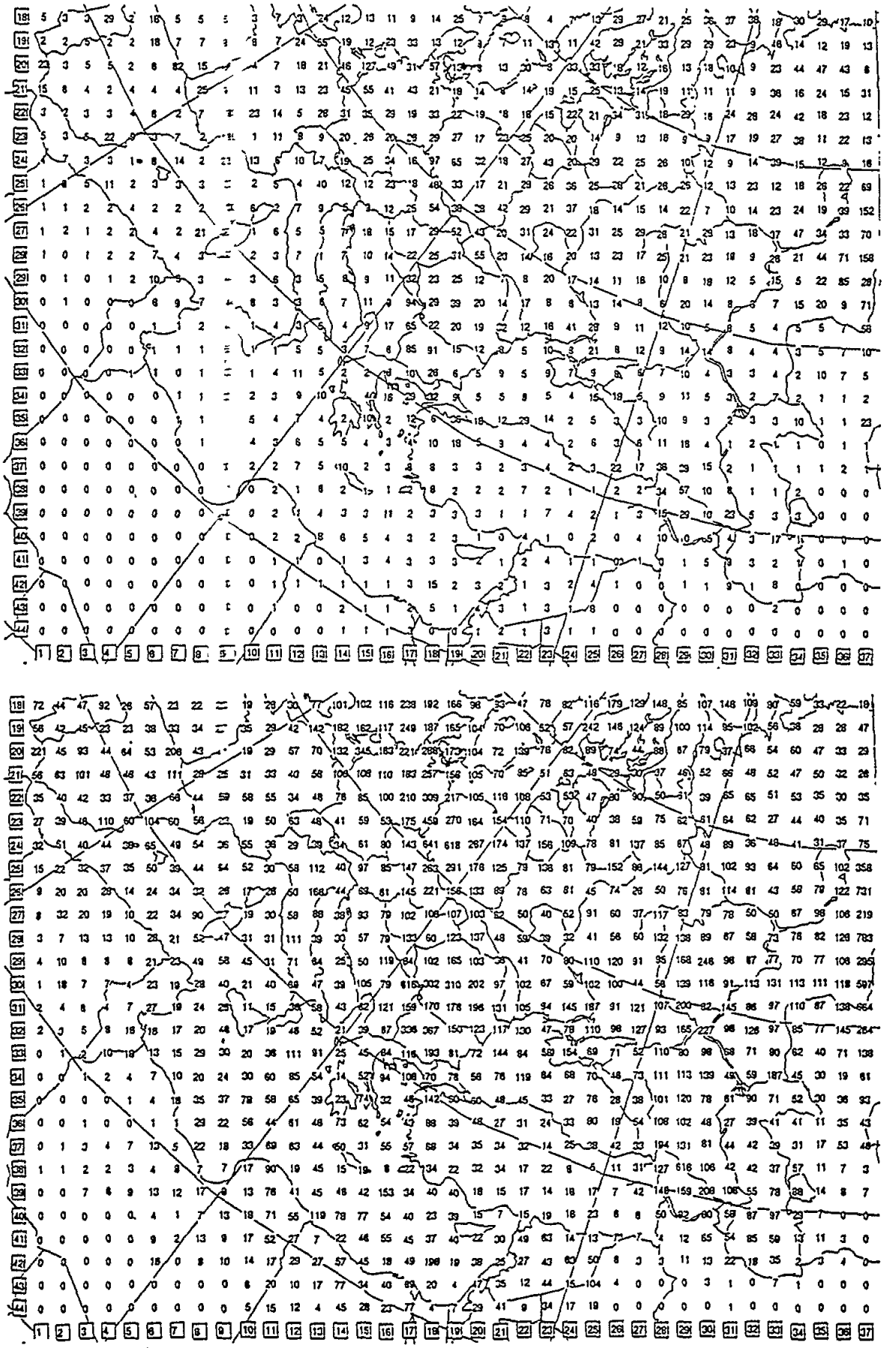


Fig. C58. Arsenic total deposition (1 ug As/m^2) - upper panel, concentration in air (0.01 ng As/m^3) - lower panel, October 1991.

APPENDIX D

Monthly maps of surface pressure fields (in hPa), baric topography at 850 hPa level (in 10m), cloudiness fields and precipitation amount (in mm).

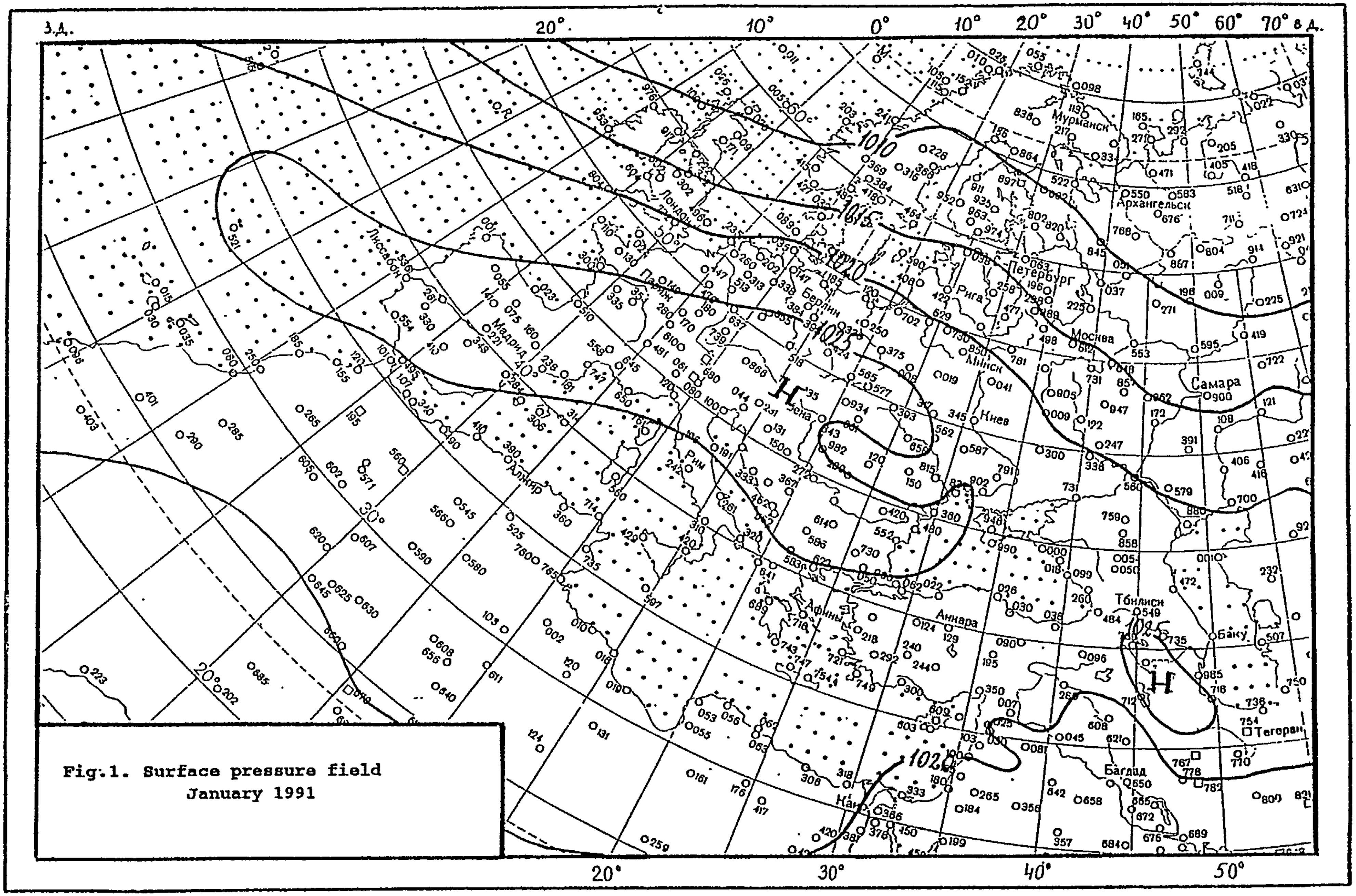


Fig.1. Surface pressure field
January 1991

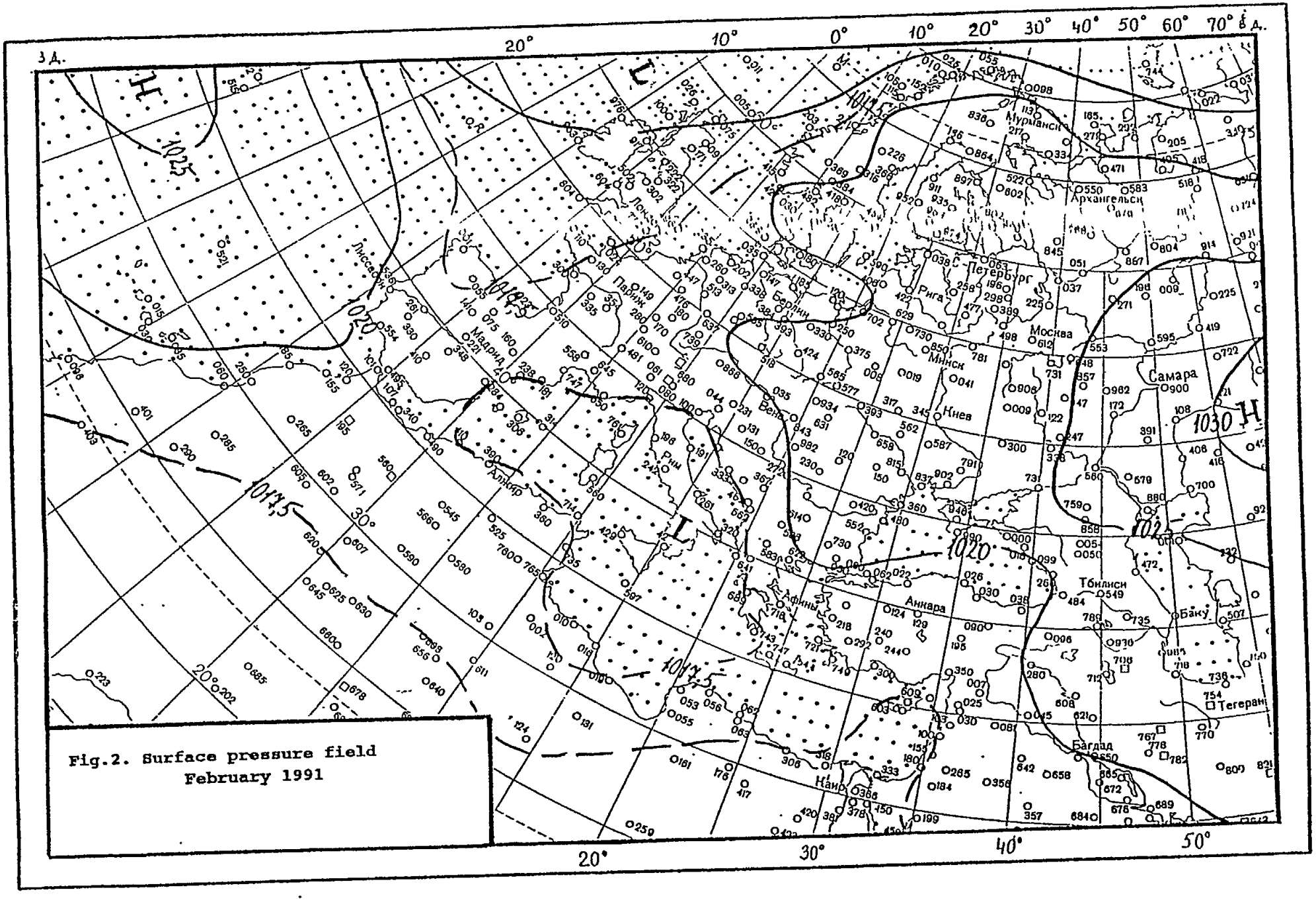


Fig.2. Surface pressure field
February 1991

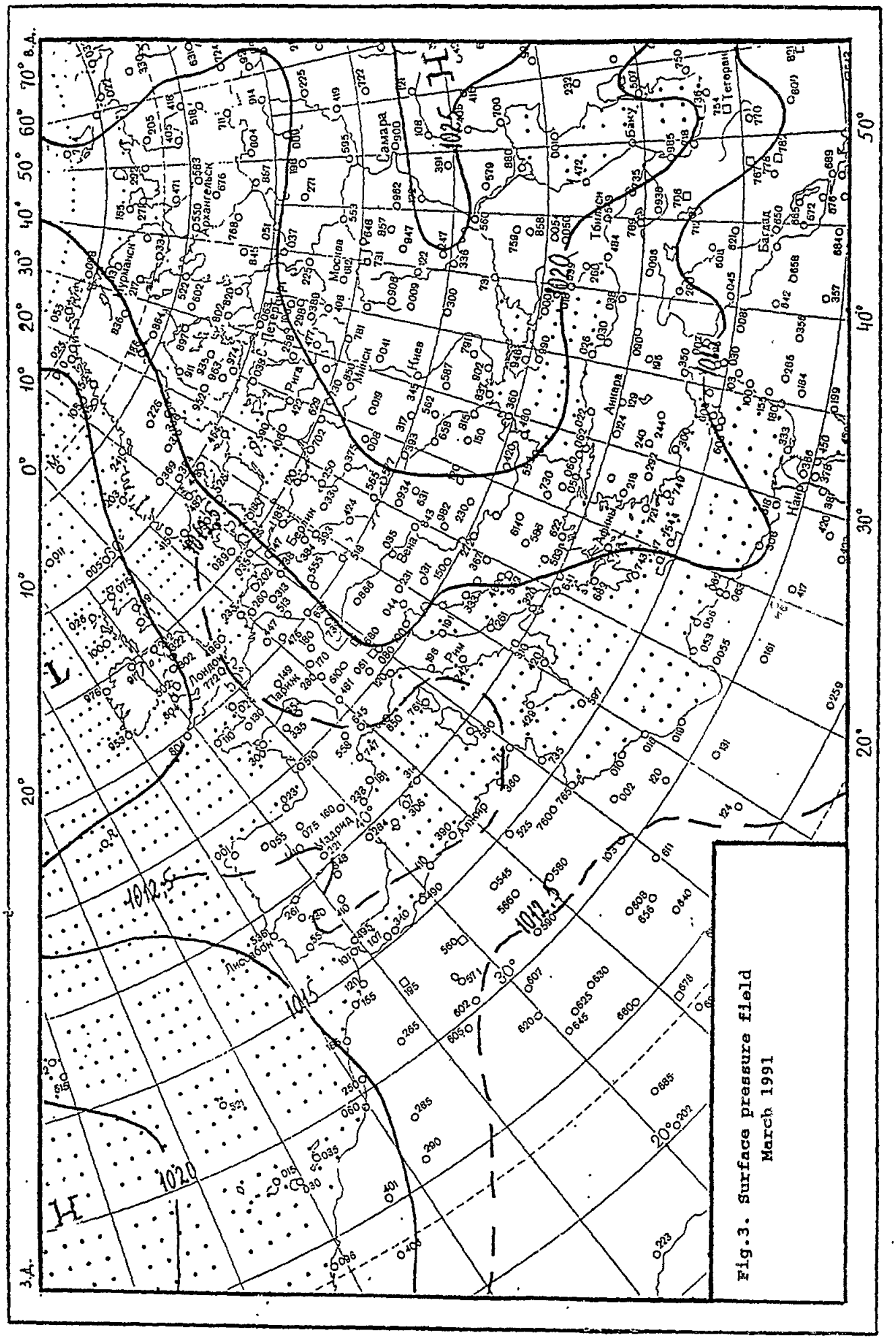


Fig.3. Surface pressure field
March 1991

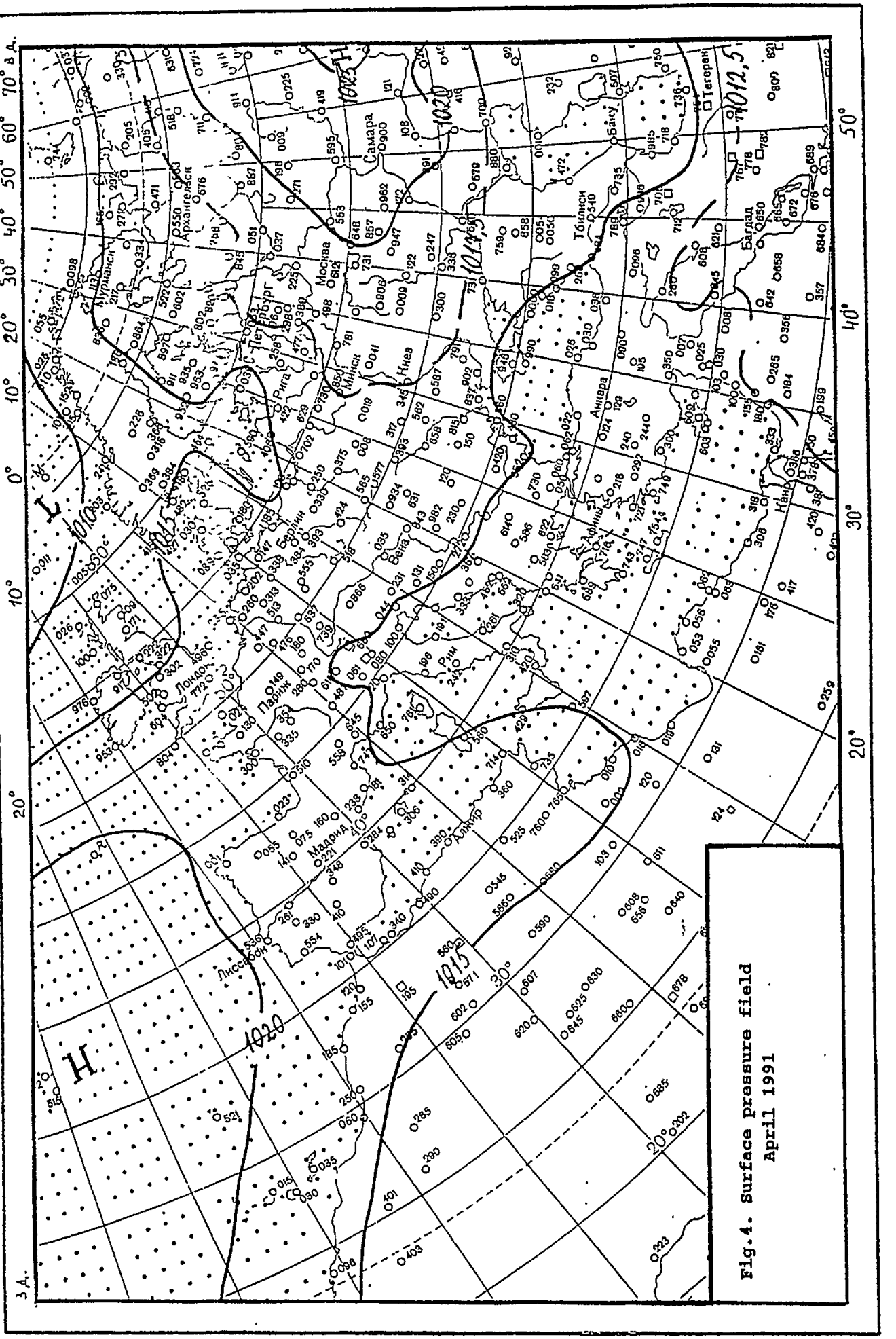


Fig.4. Surface pressure field
April 1991

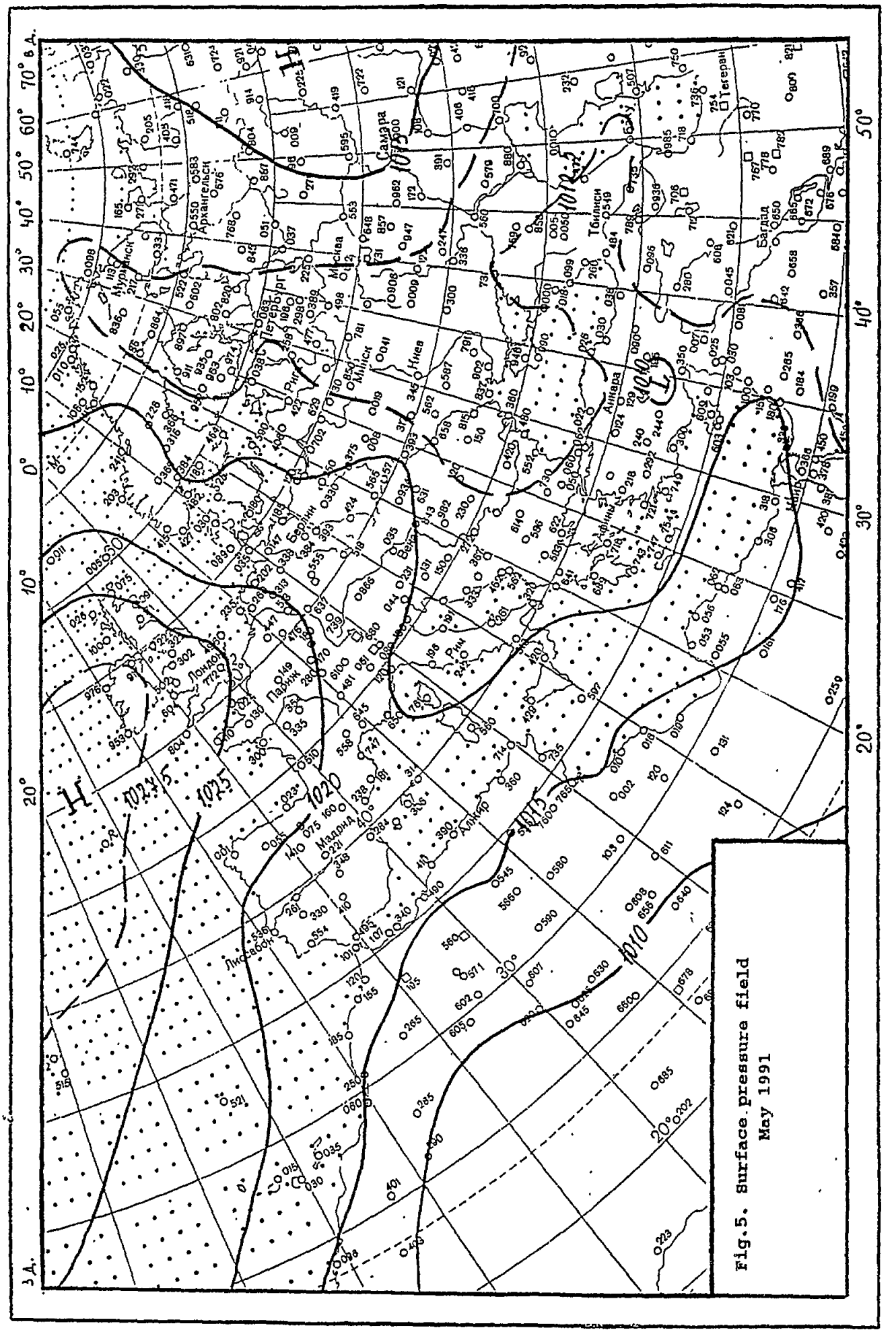


Fig.5. Surface pressure field
May 1991

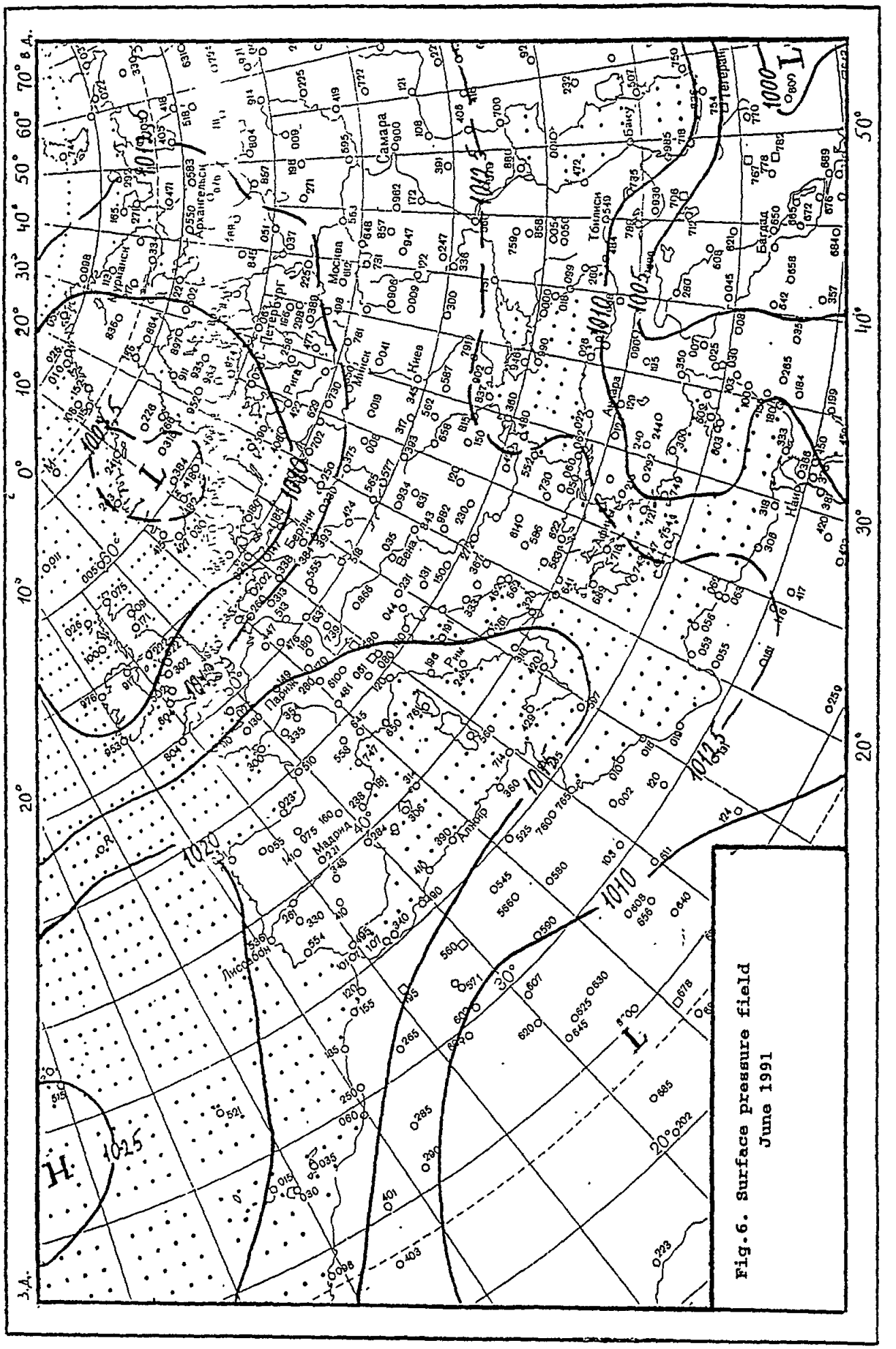


Fig.6. Surface pressure field
June 1991

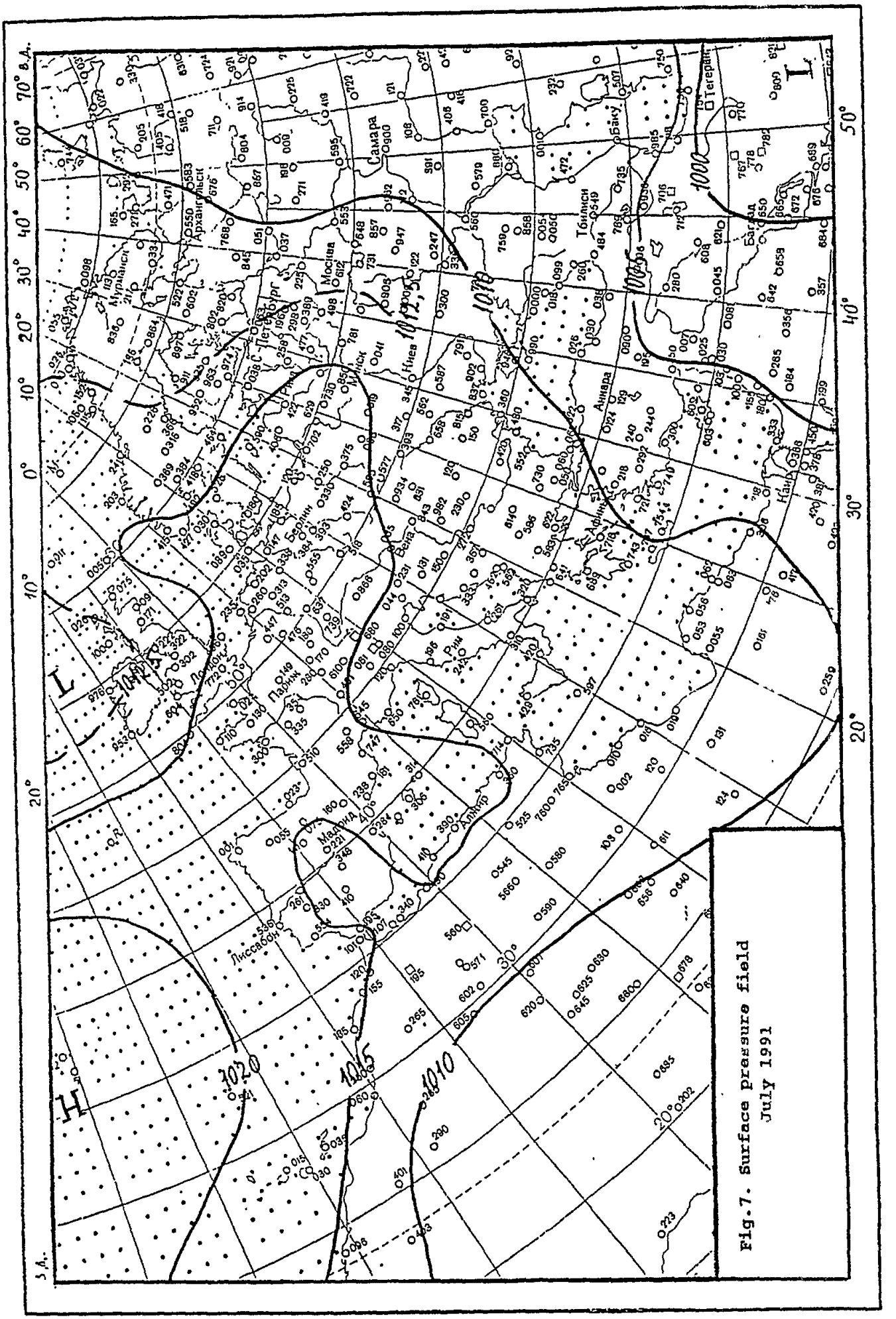


Fig.7. Surface pressure field
July 1991

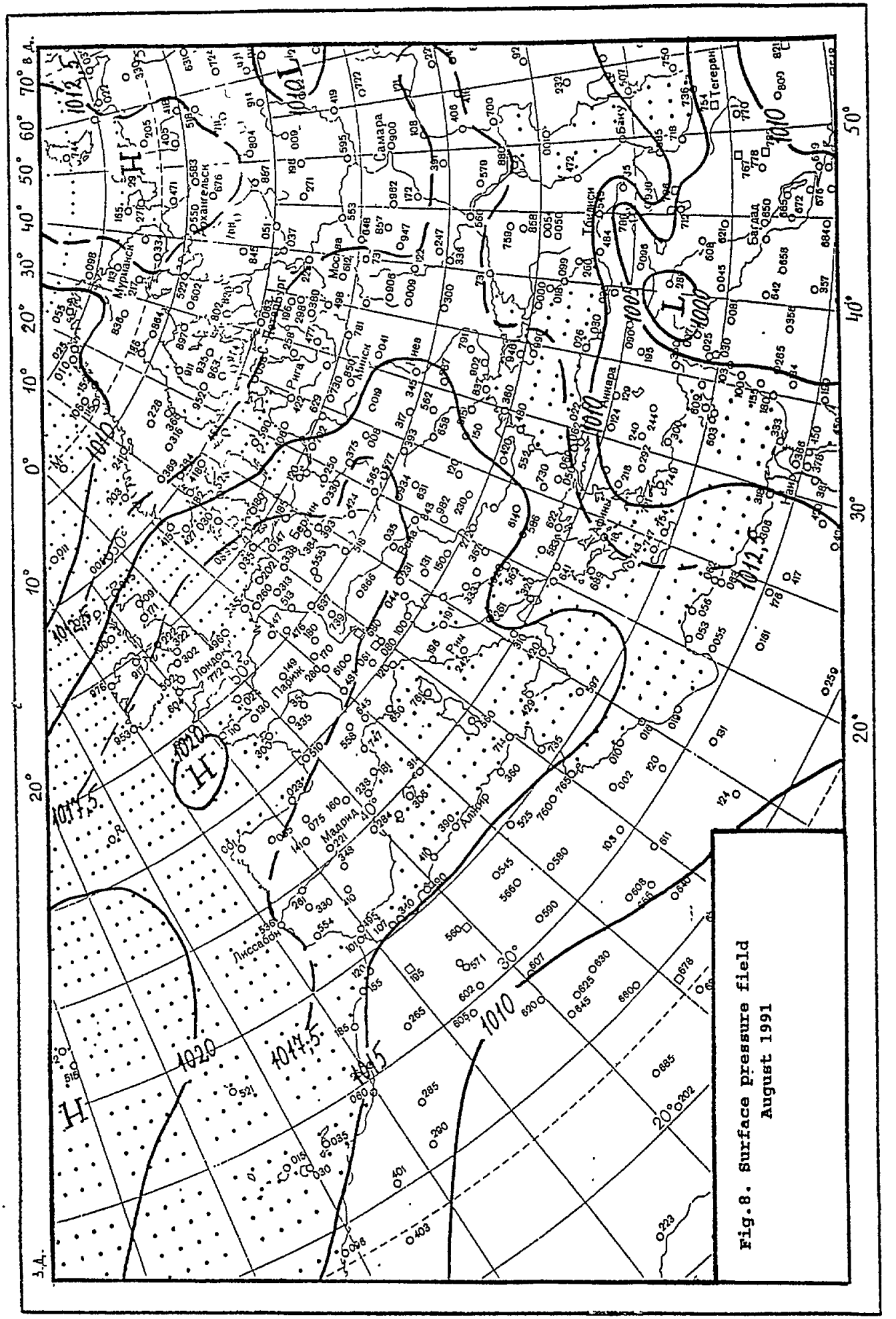


Fig.8. Surface pressure field
August 1991

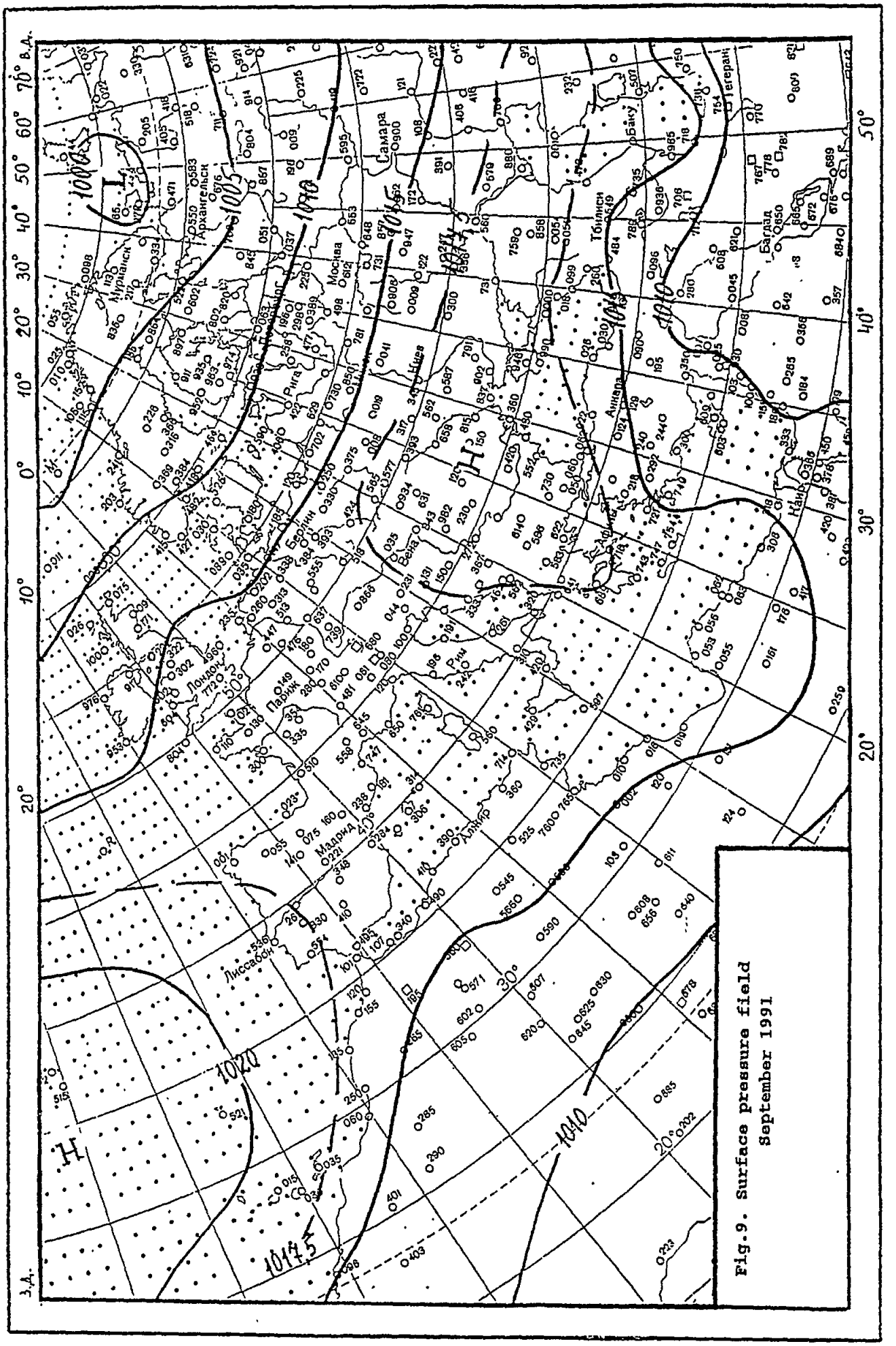


Fig.9. Surface pressure field
September 1991

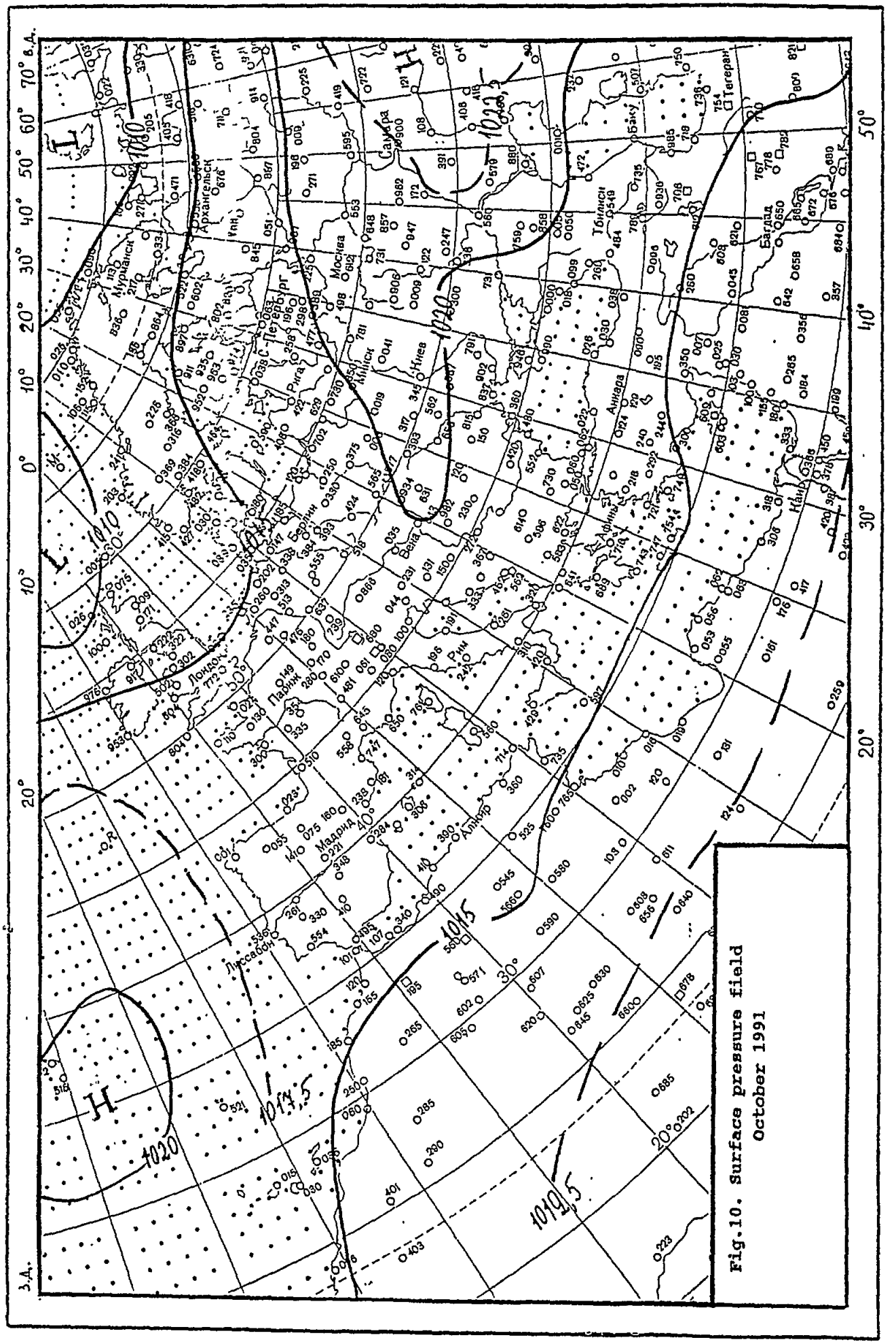


Fig.10. Surface pressure field
October 1991

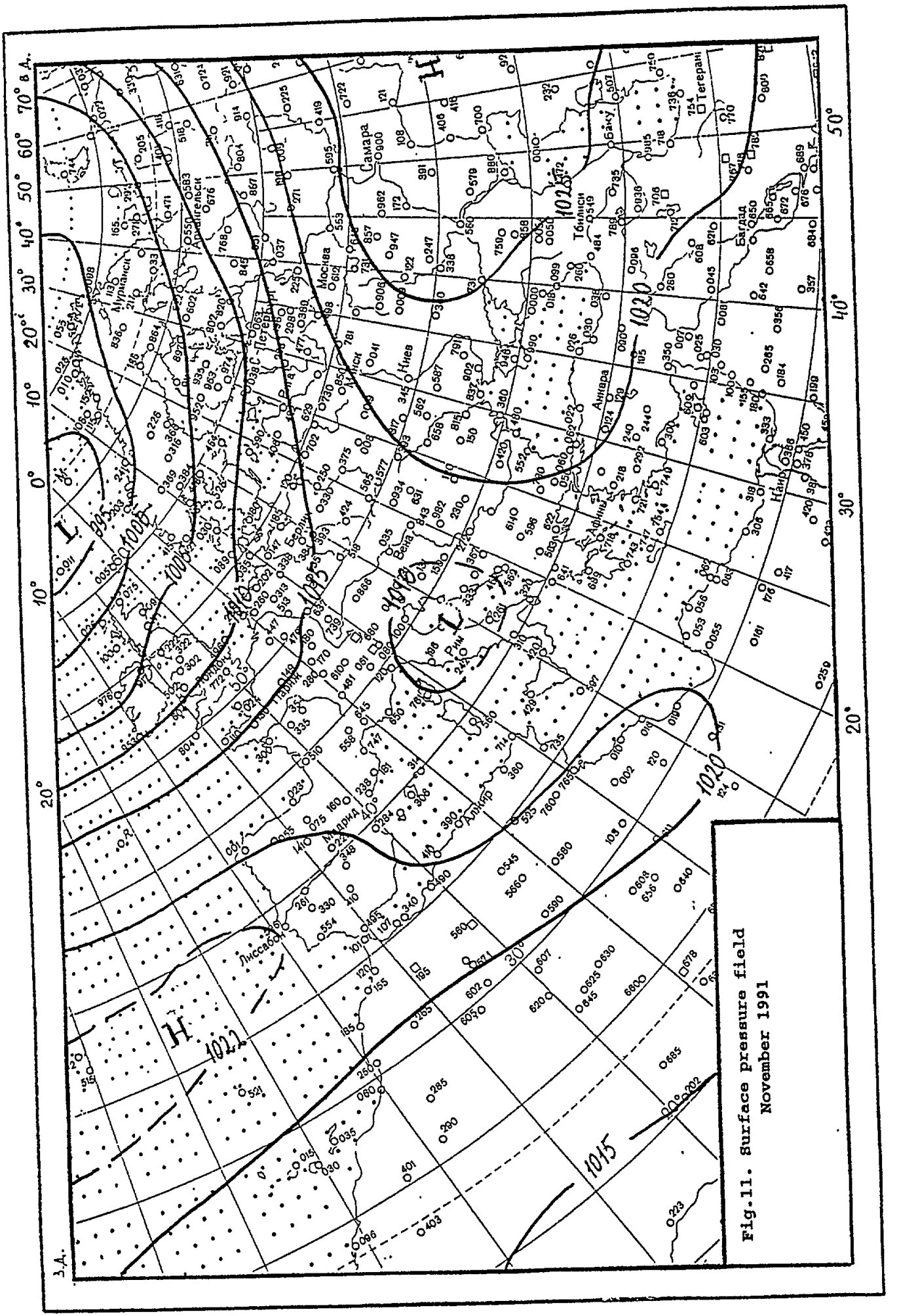


Fig.11. Surface pressure field
November 1991

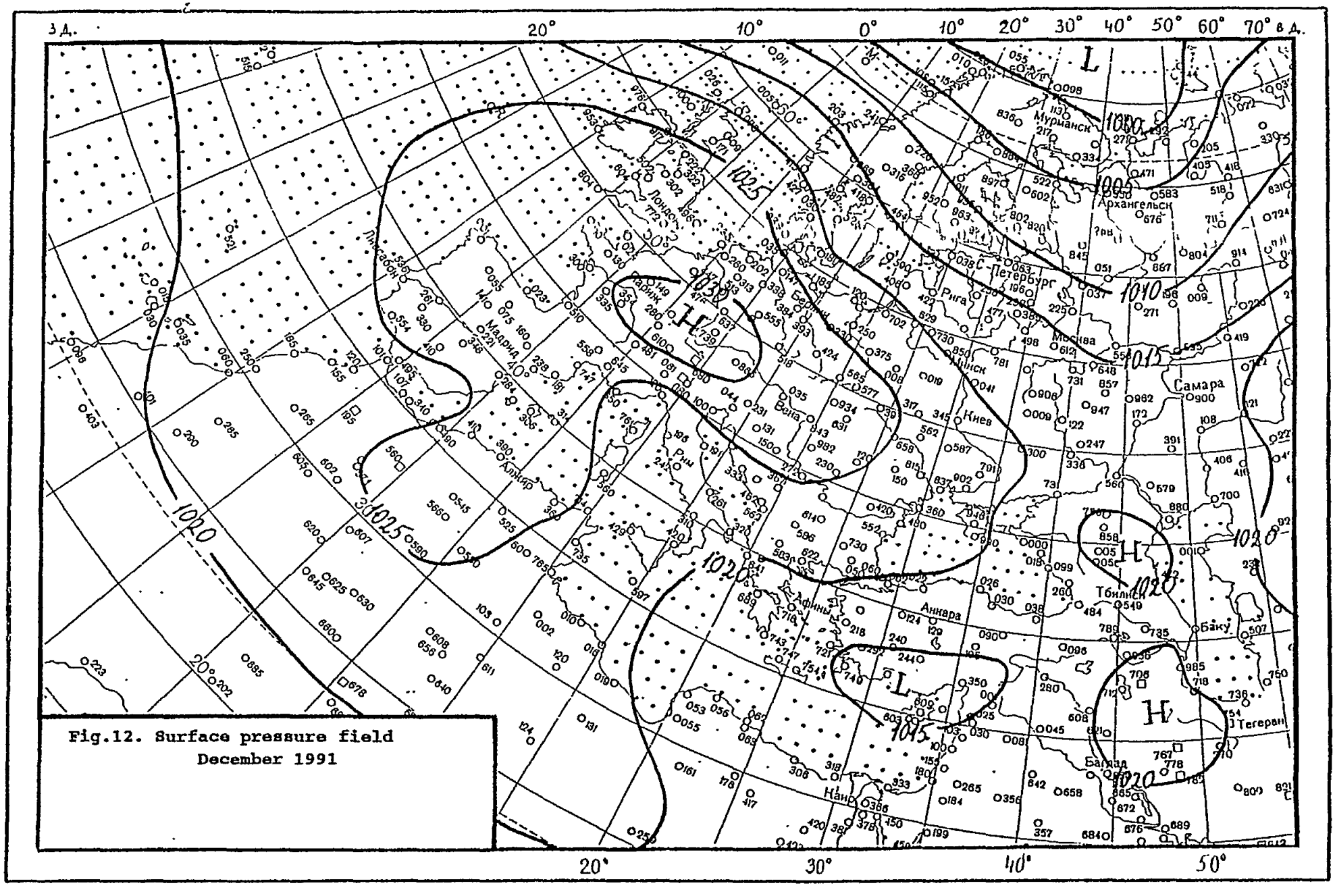


Fig.12. Surface pressure field
December 1991

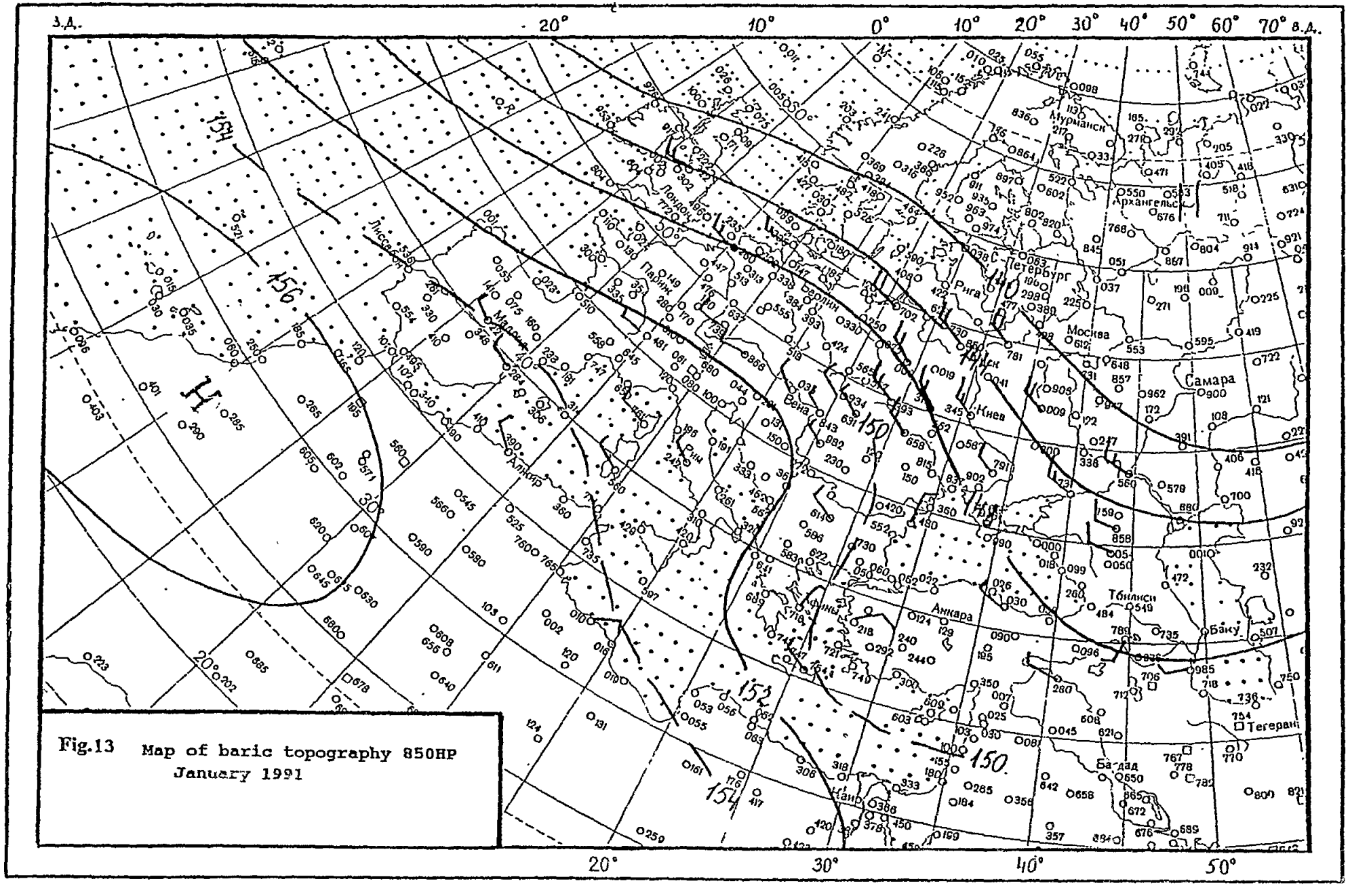


Fig.13 Map of baric topography 850HP
January 1991

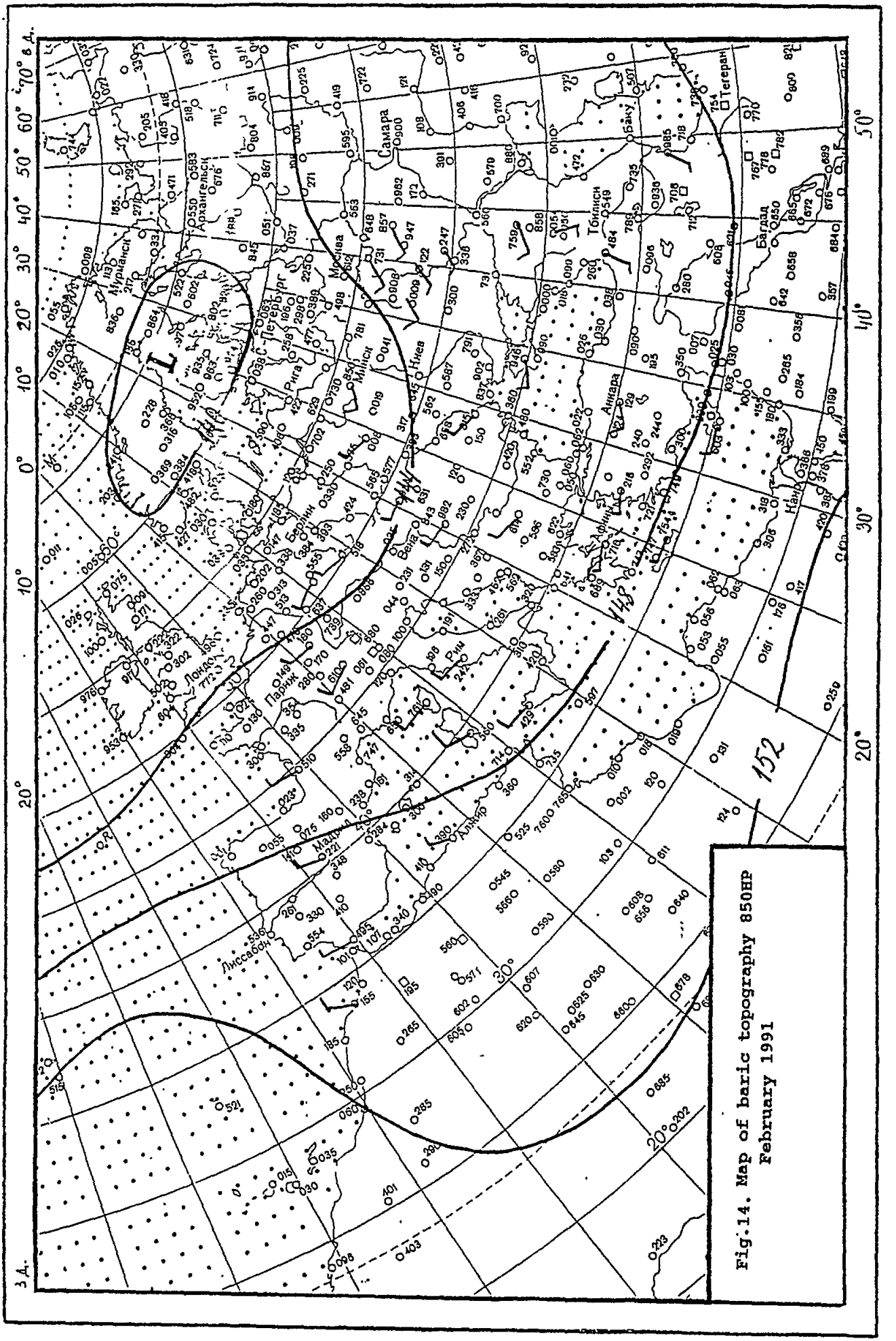


Fig.14. Map of baric topography 850HP
February 1991

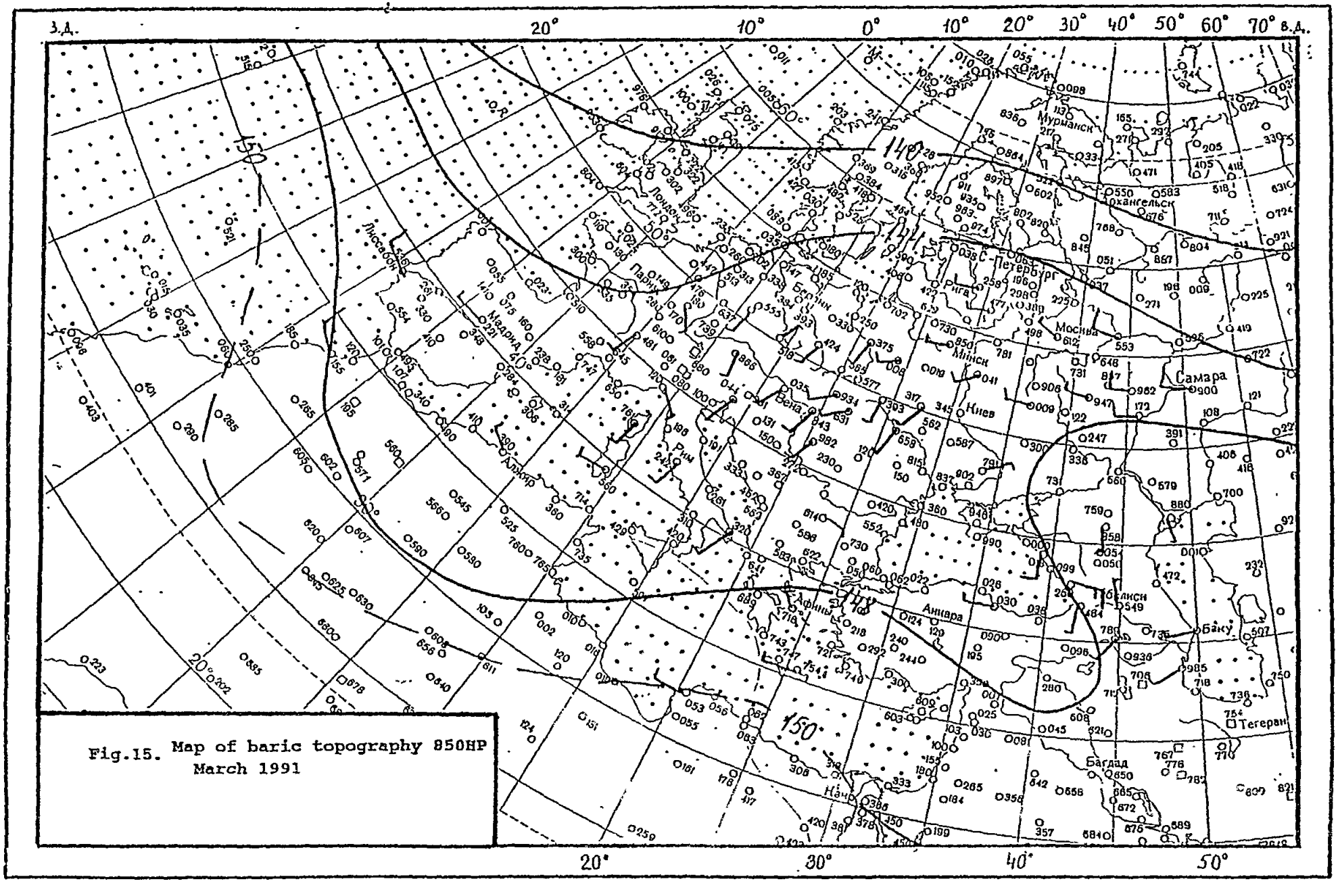


Fig.15. Map of baric topography 850HP
March 1991

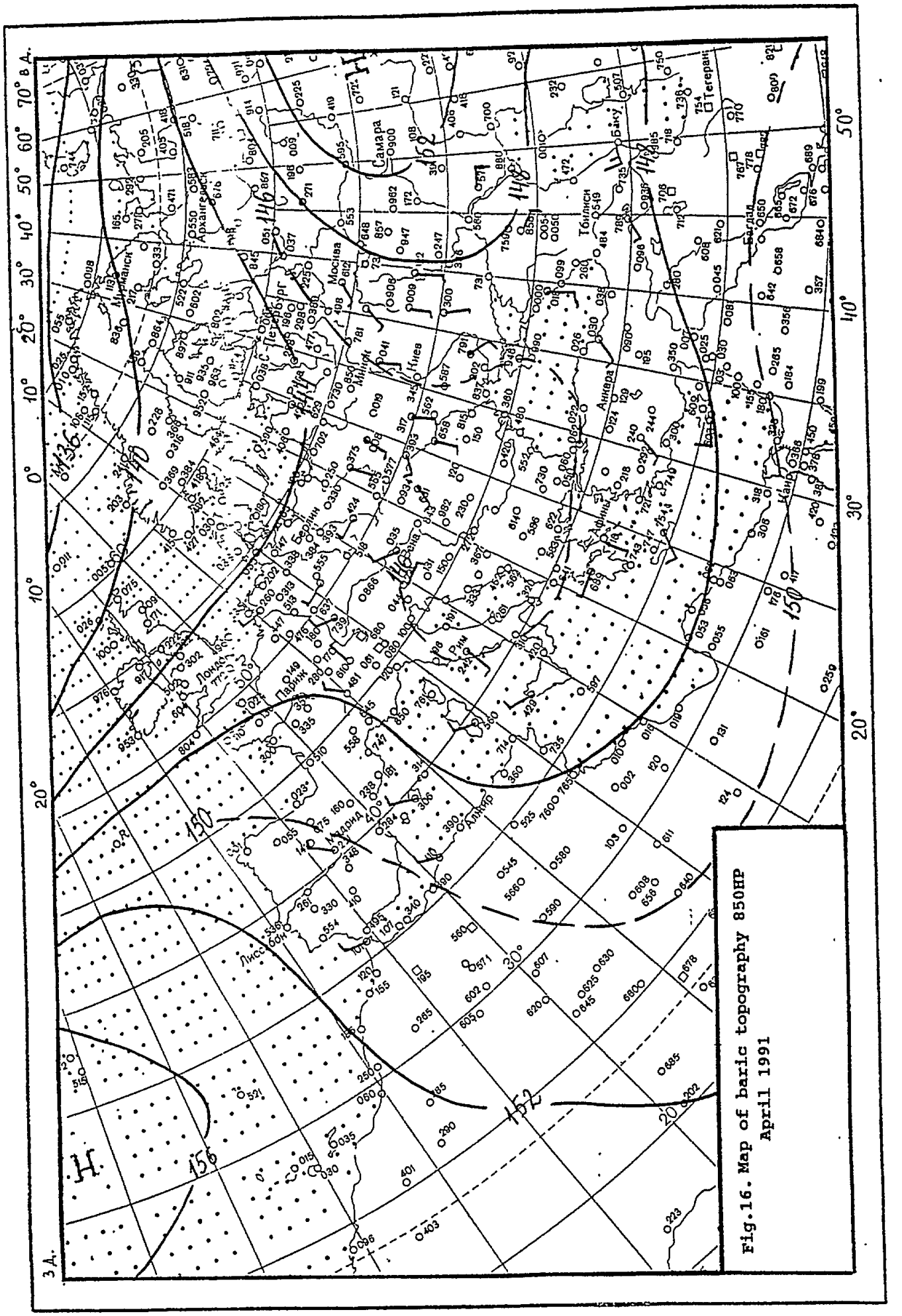


Fig.16. Map of baric topography 850HP
April 1991

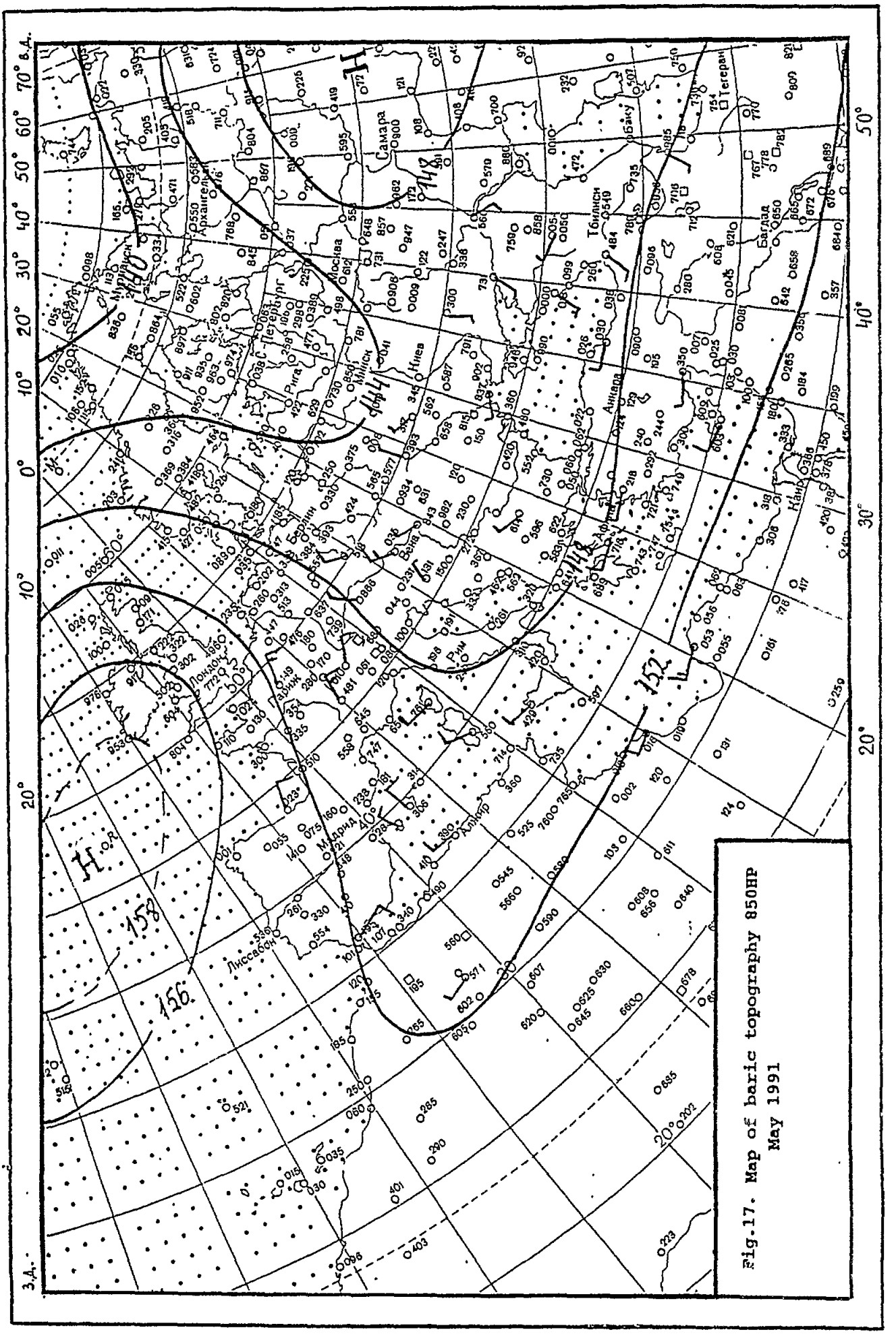


Fig.17. Map of baric topography 850HP
May 1991

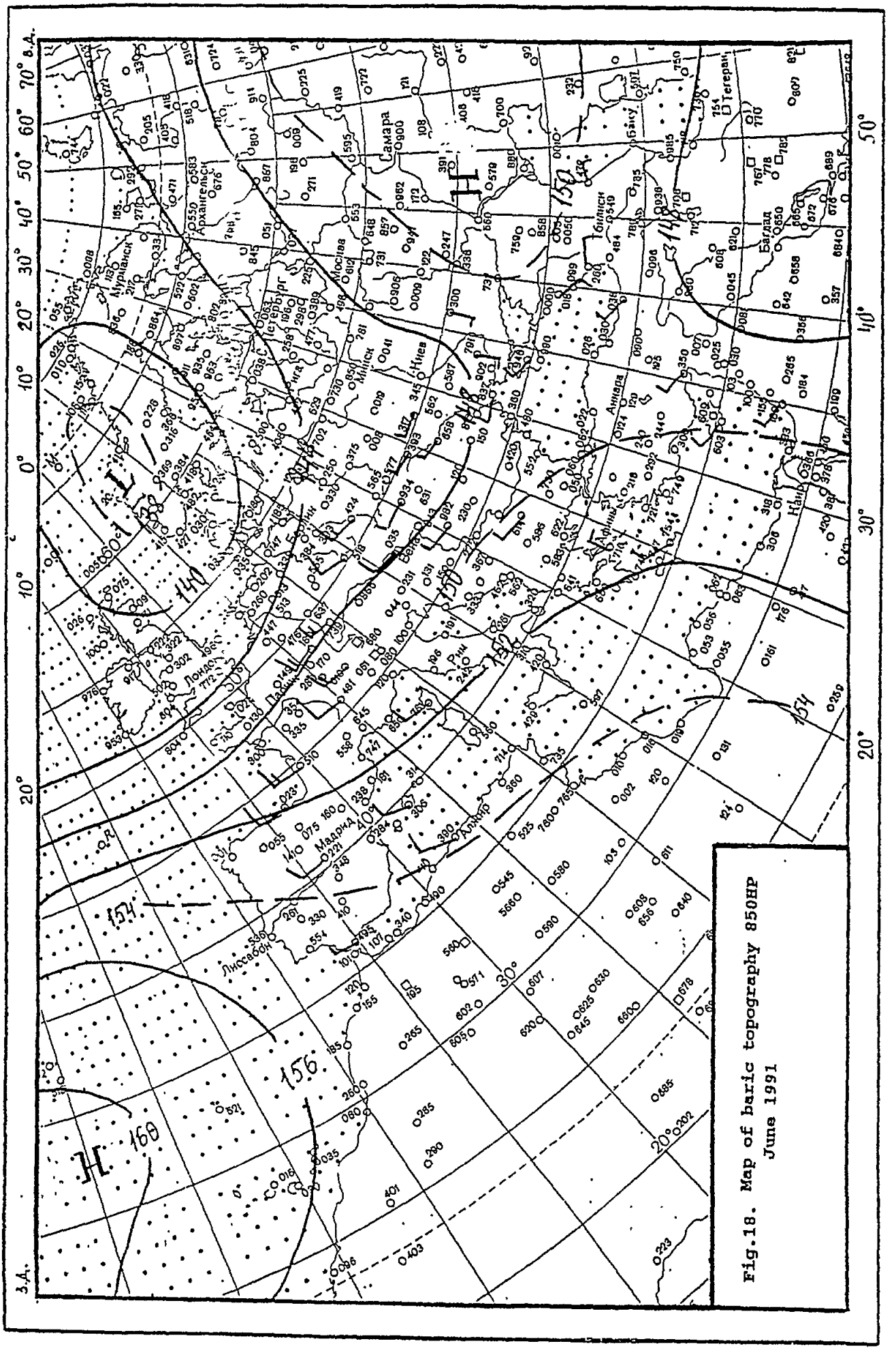


Fig.18. Map of baric topography 850HP
June 1991

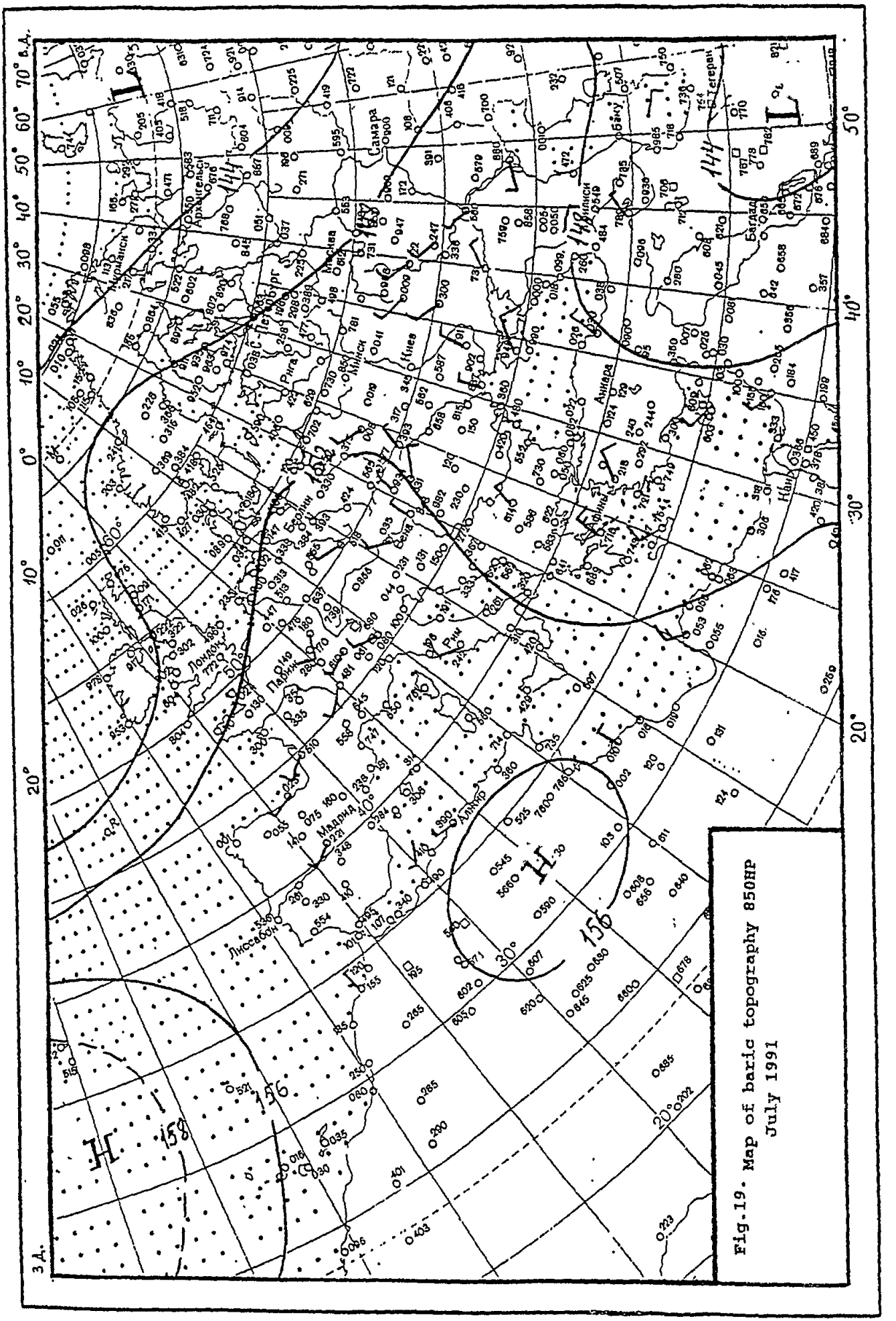


Fig.19. Map of baric topography 850HP
July 1991

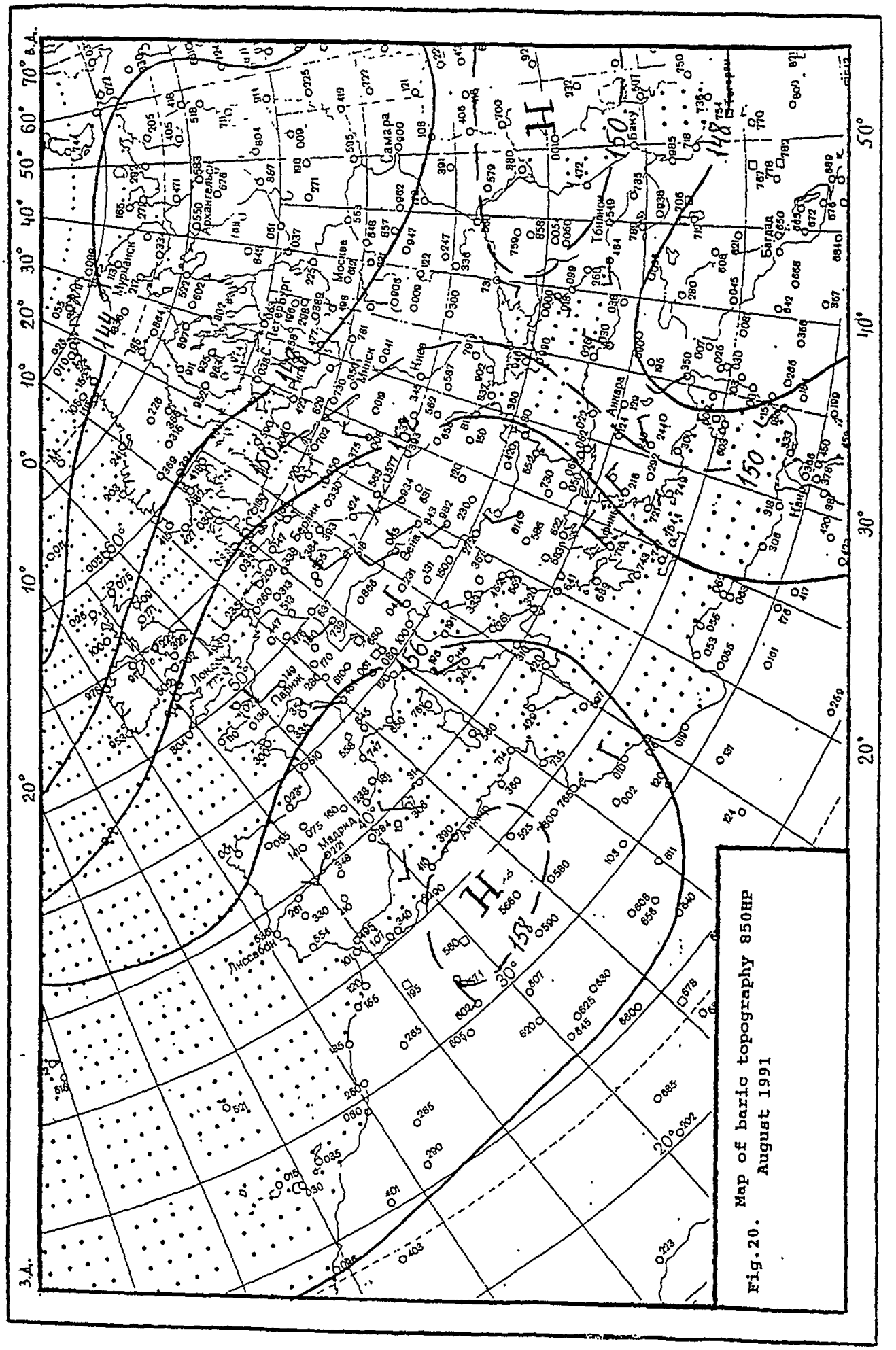


Fig.20. Map of baric topography 850HP
August 1991

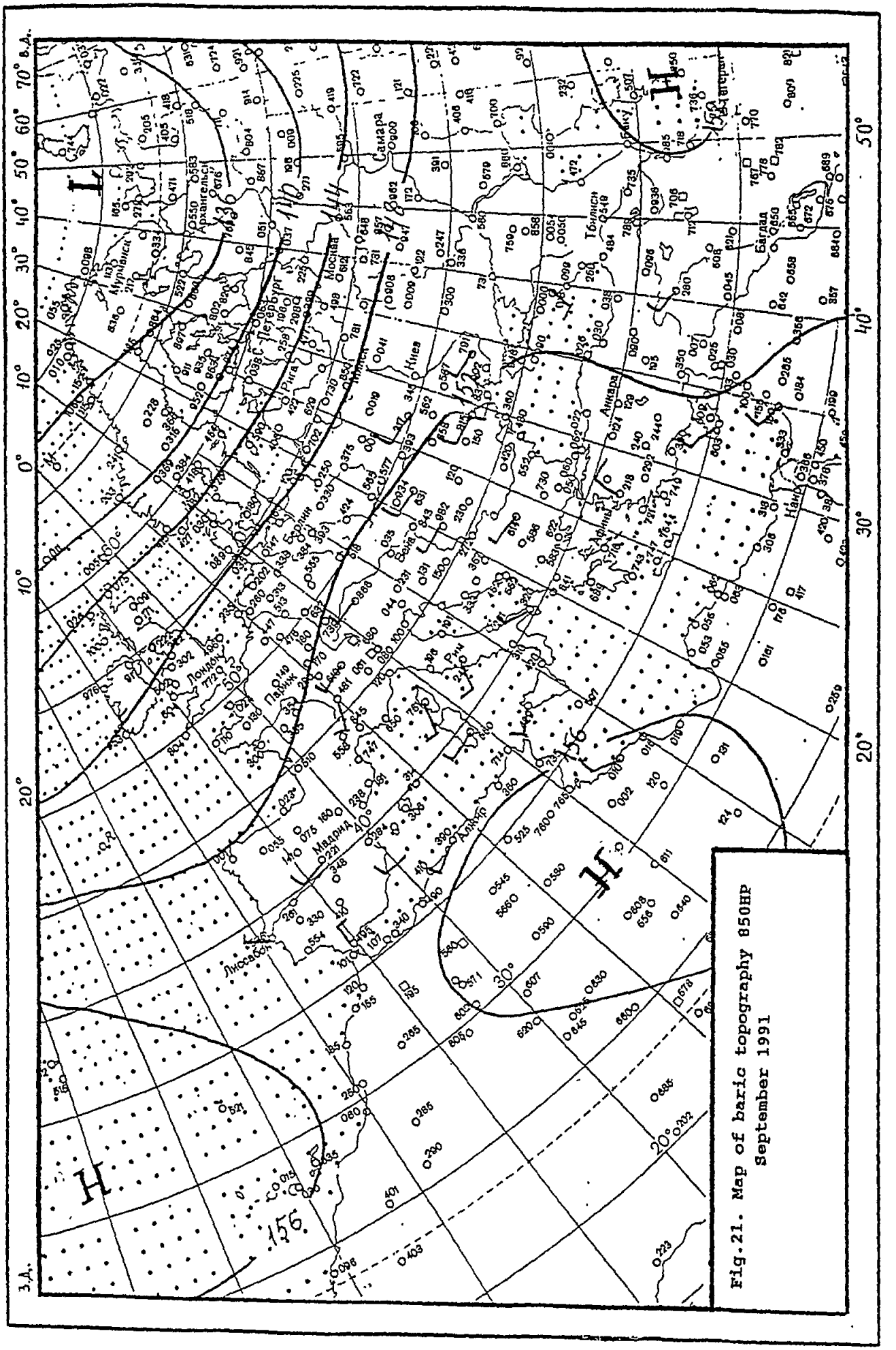


Fig.21. Map of baric topography 850HP
September 1991

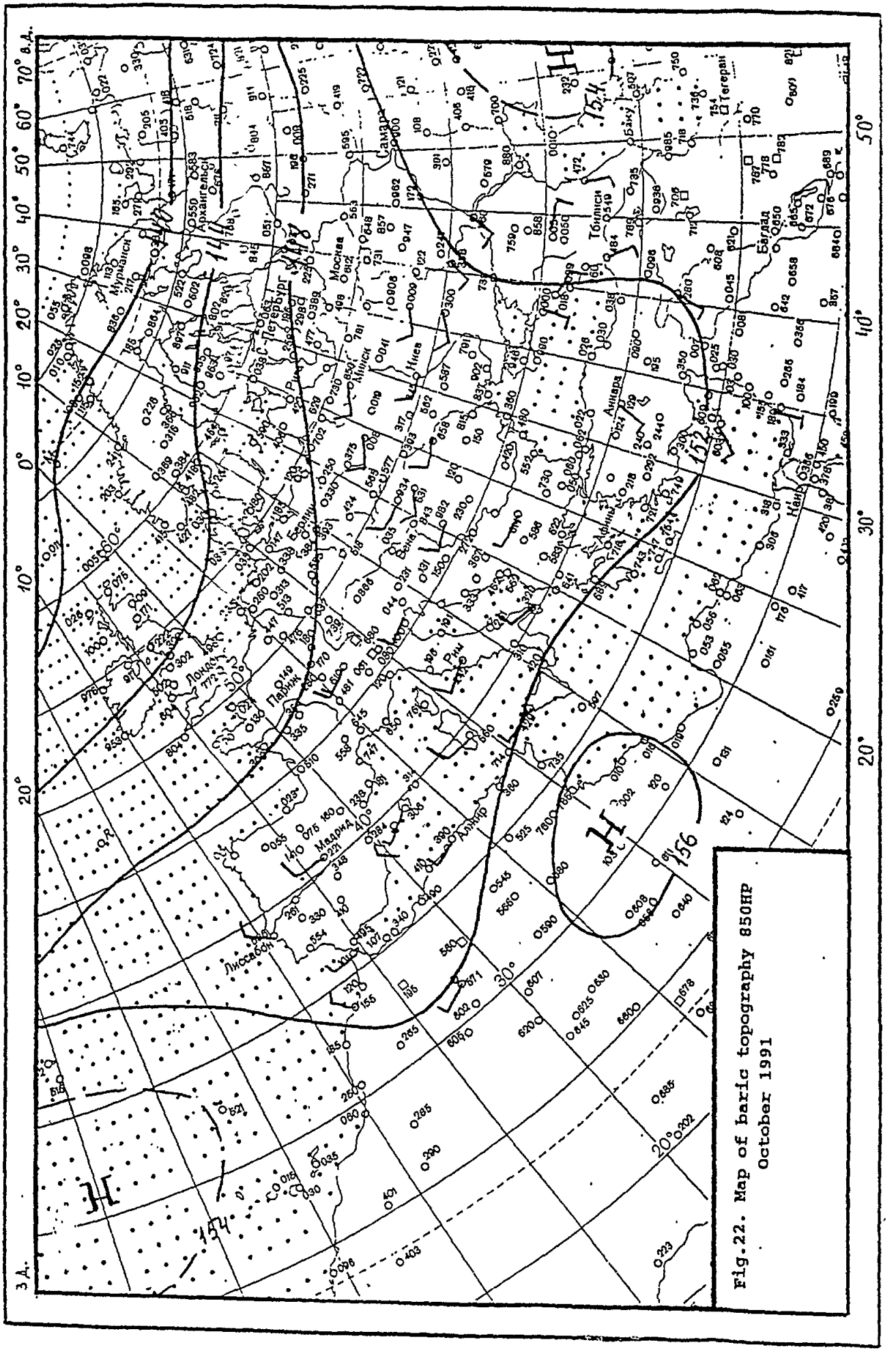


Fig.22. Map of baric topography 850HP
October 1991

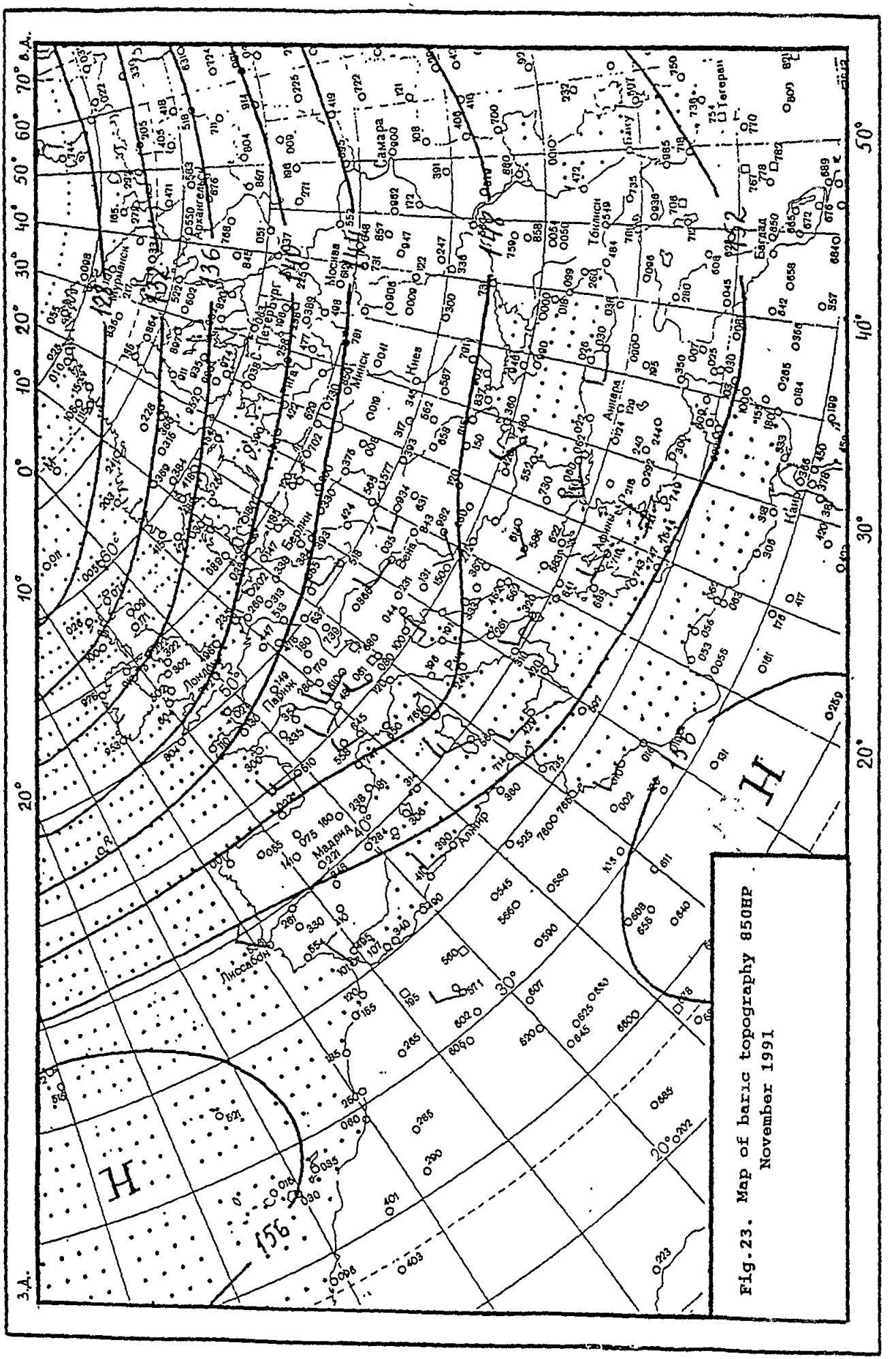


Fig.23. Map of baric topography 850HP
November 1991

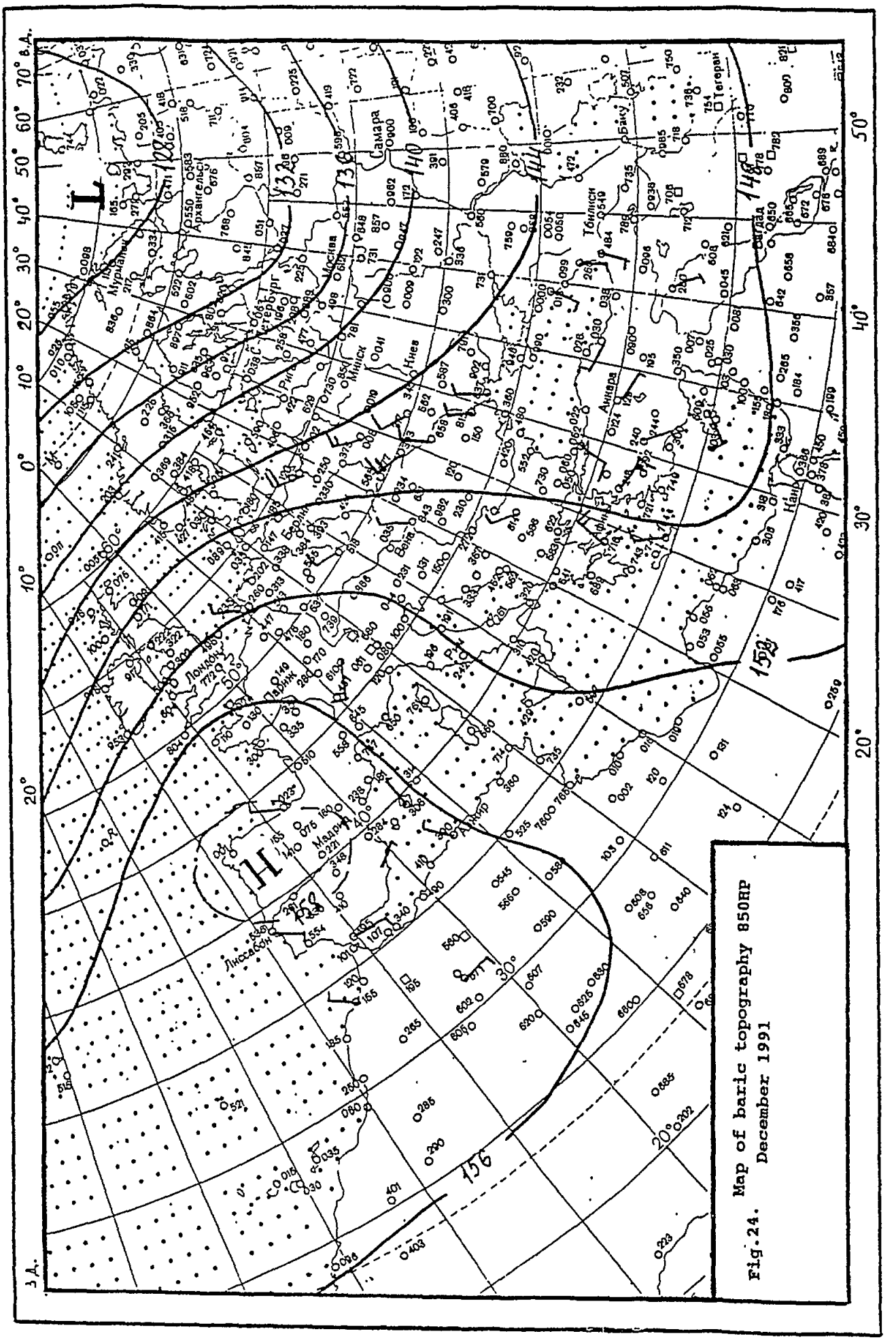


Fig.24.

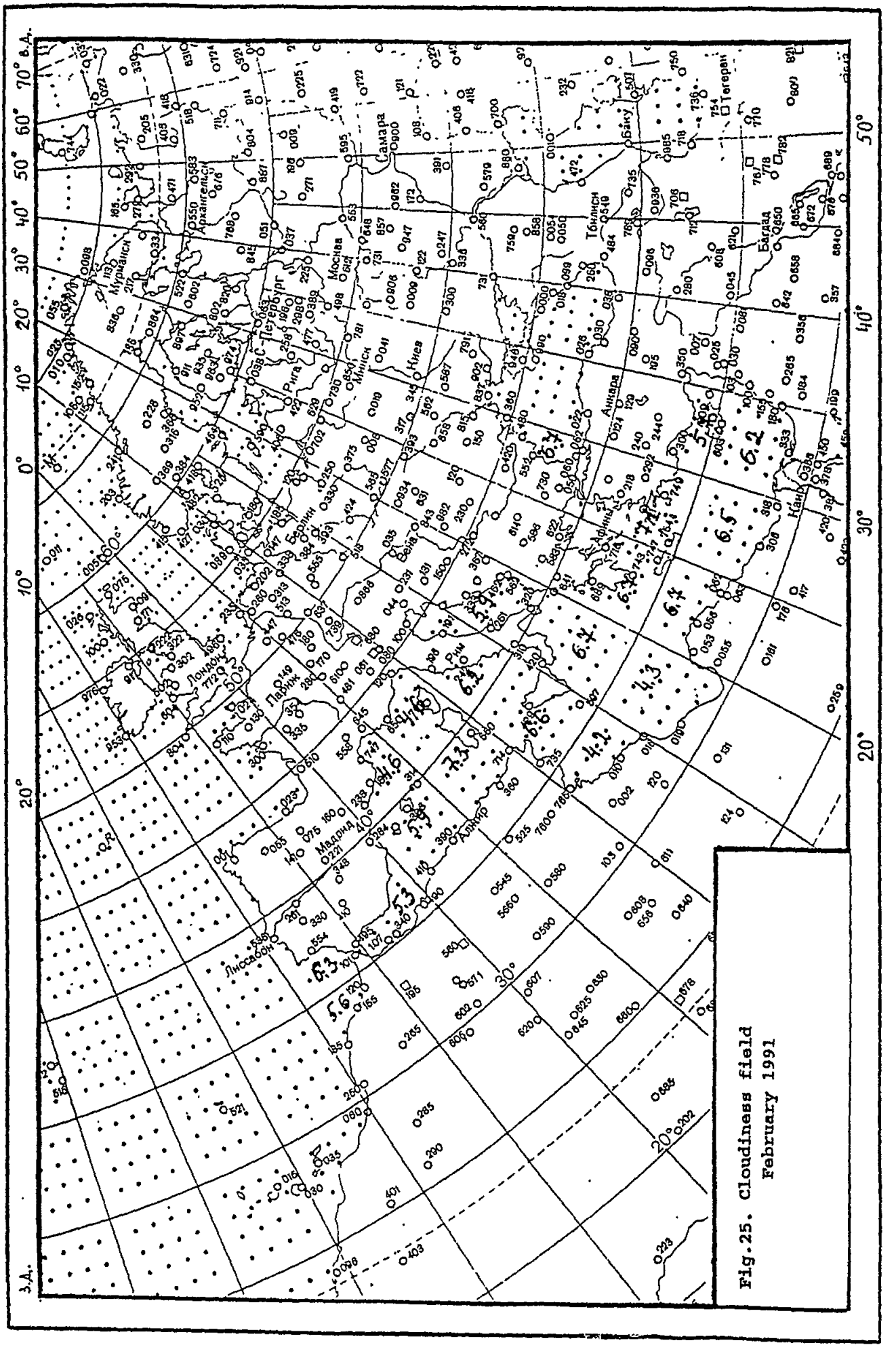


Fig.25. Cloudiness field
February 1991

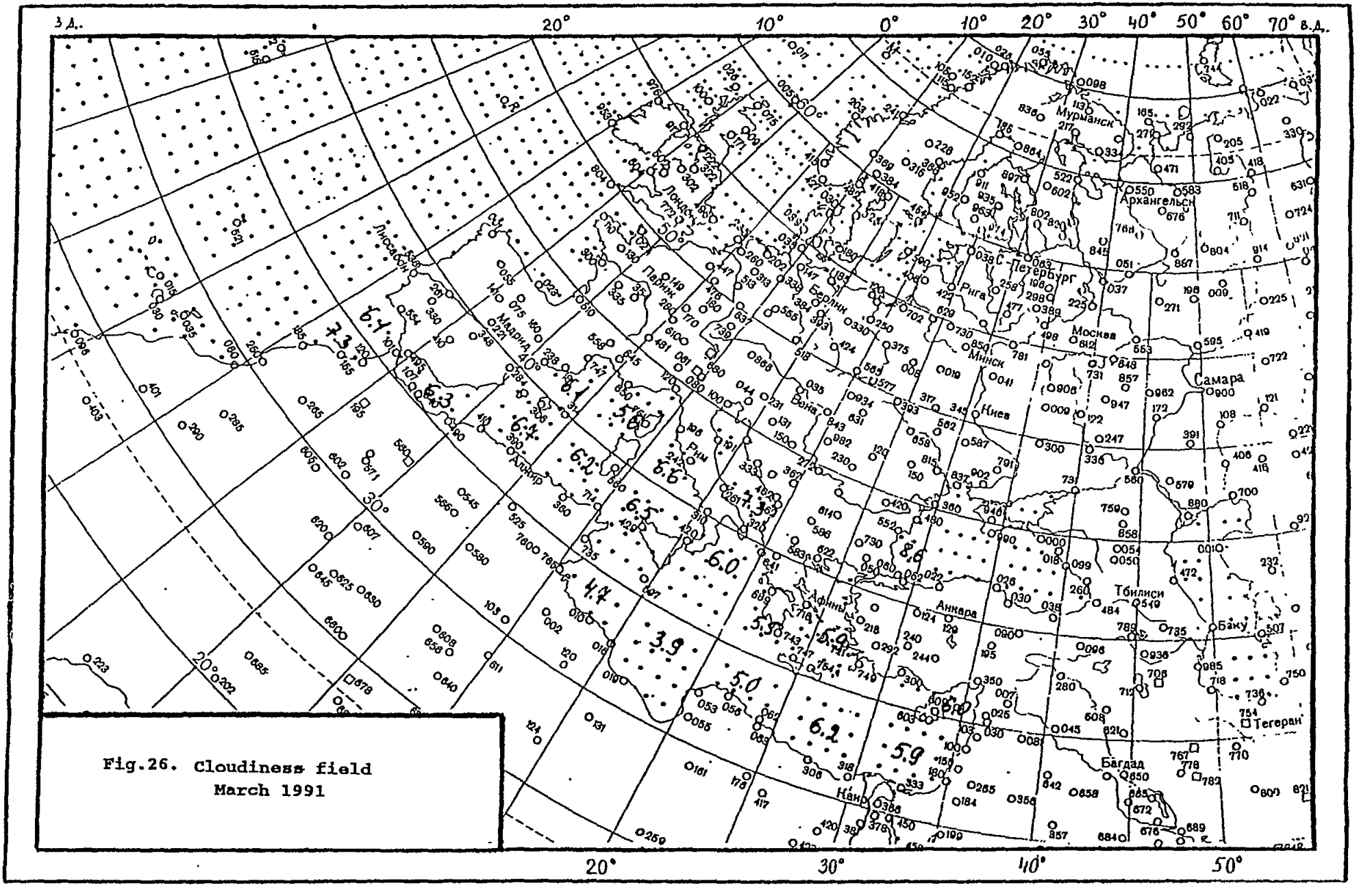


Fig.26. Cloudiness field
March 1991

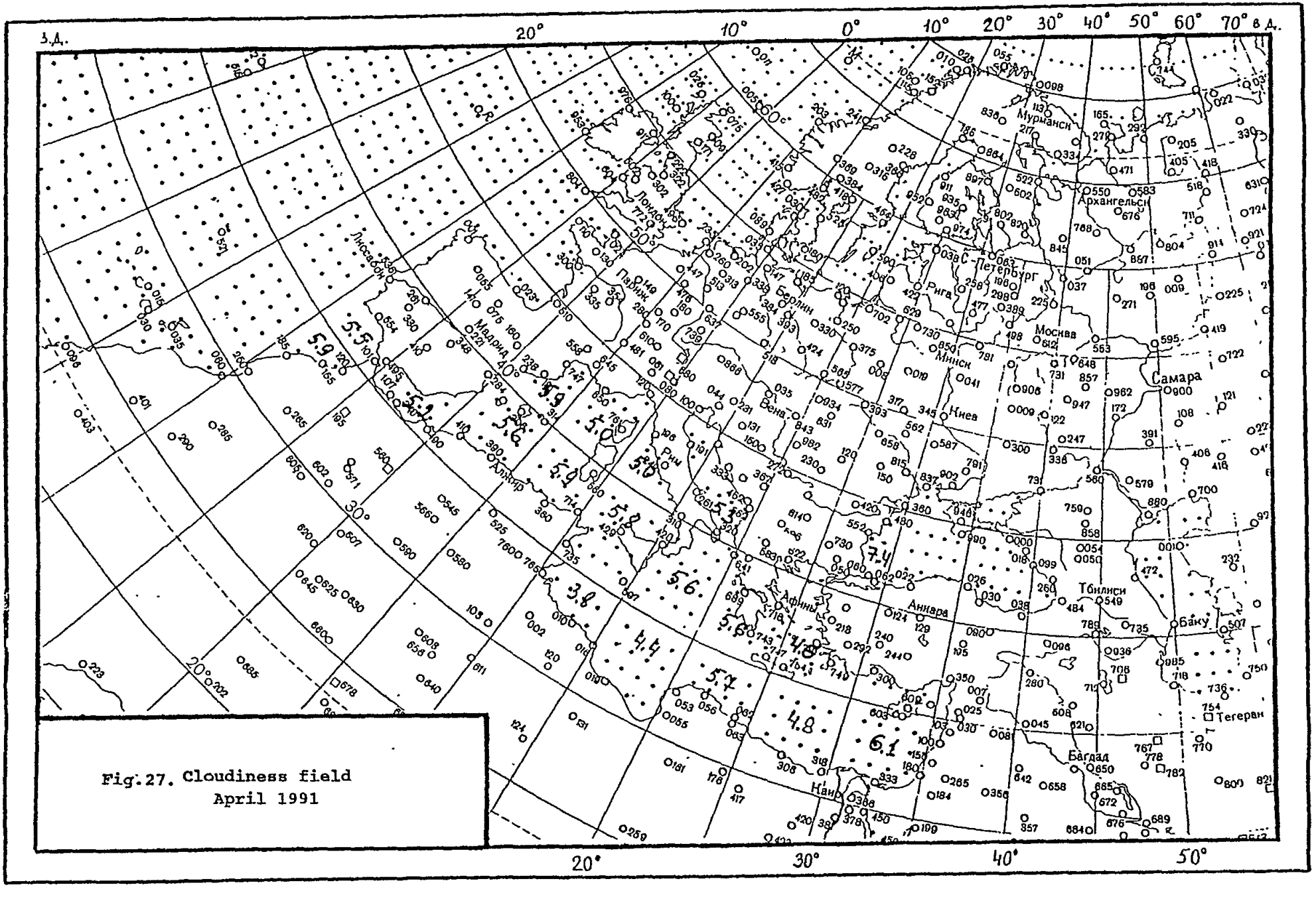


Fig.27. Cloudiness field
April 1991

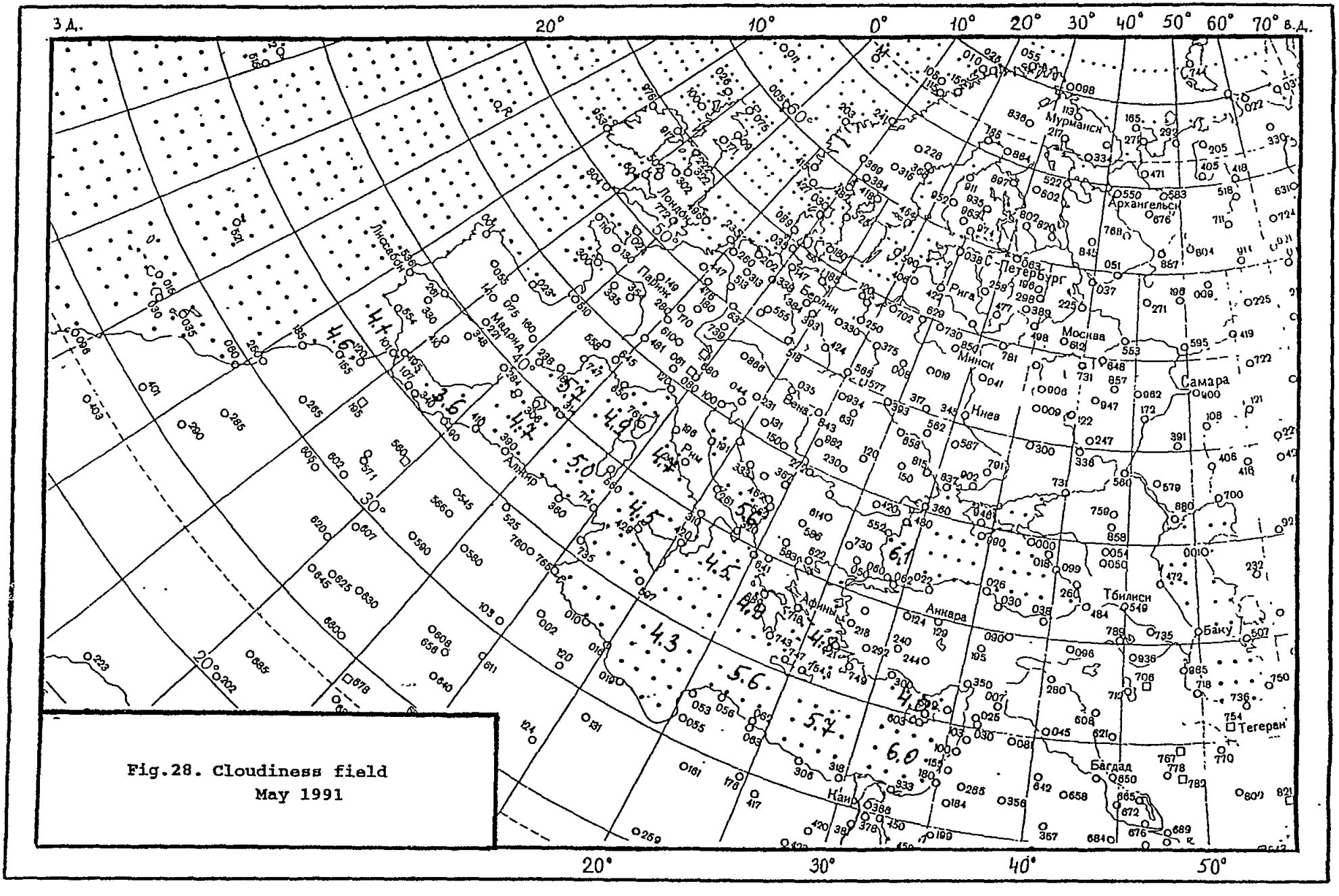


Fig.28. Cloudiness field
May 1991

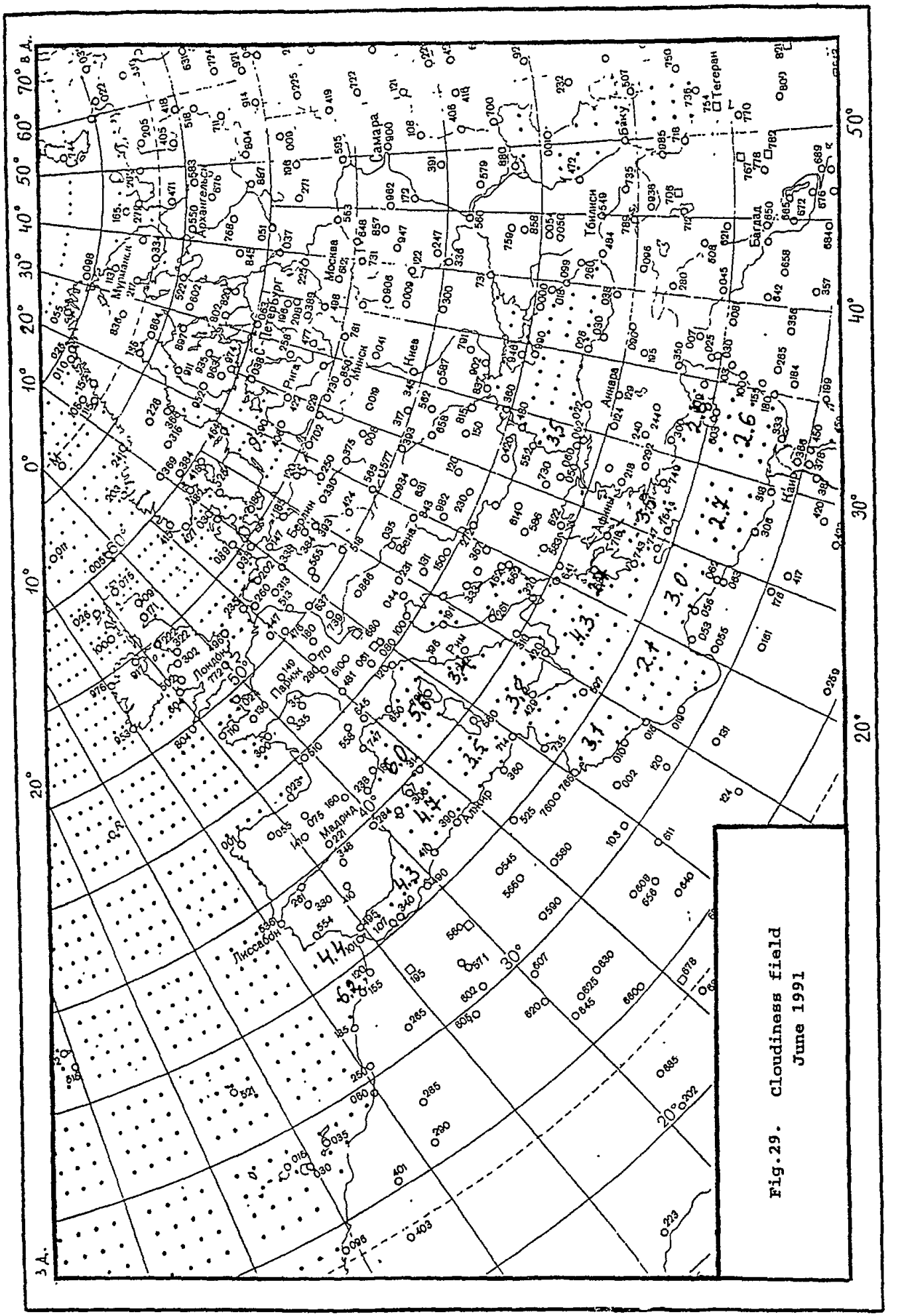


Fig.29. Cloudiness field
June 1991

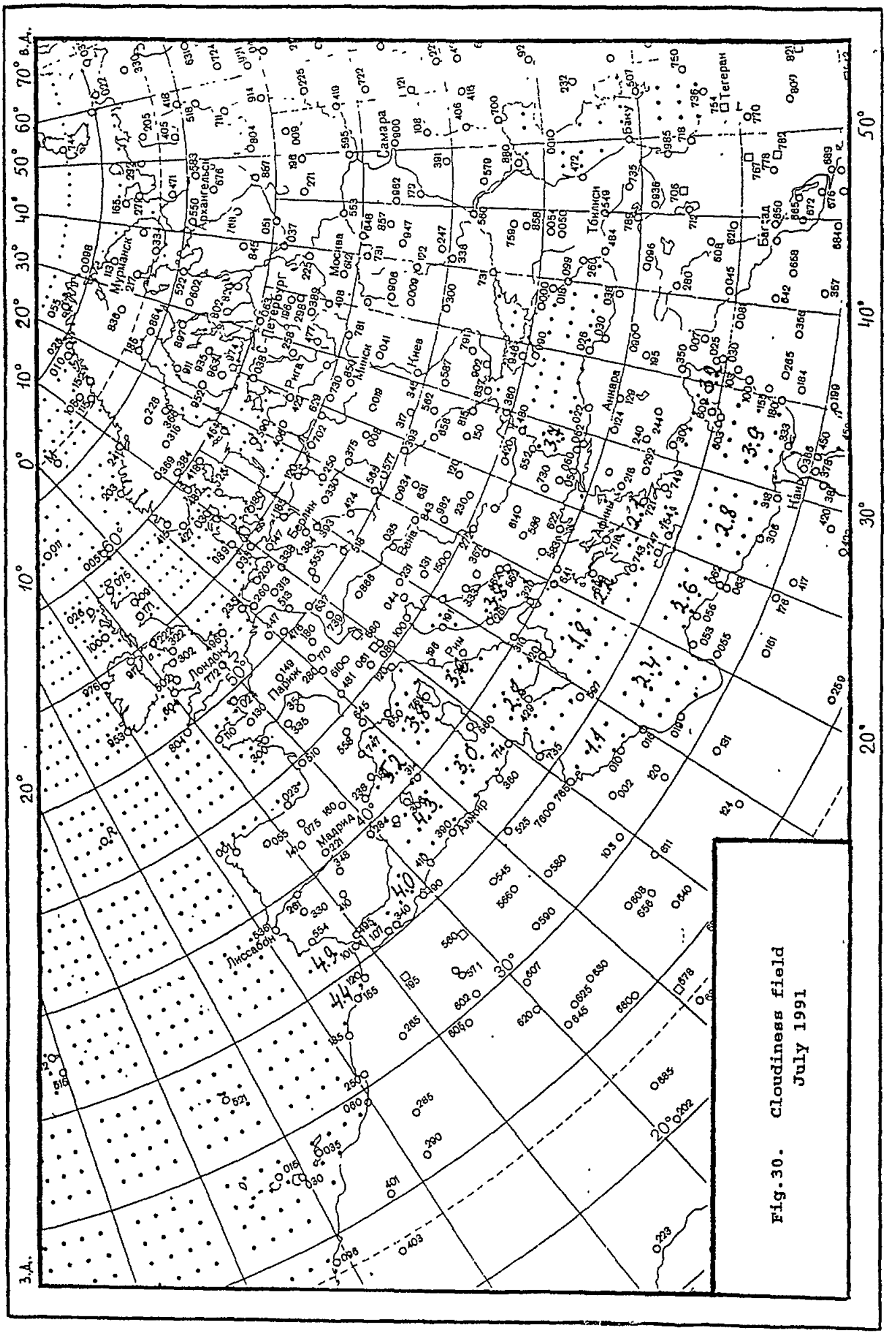
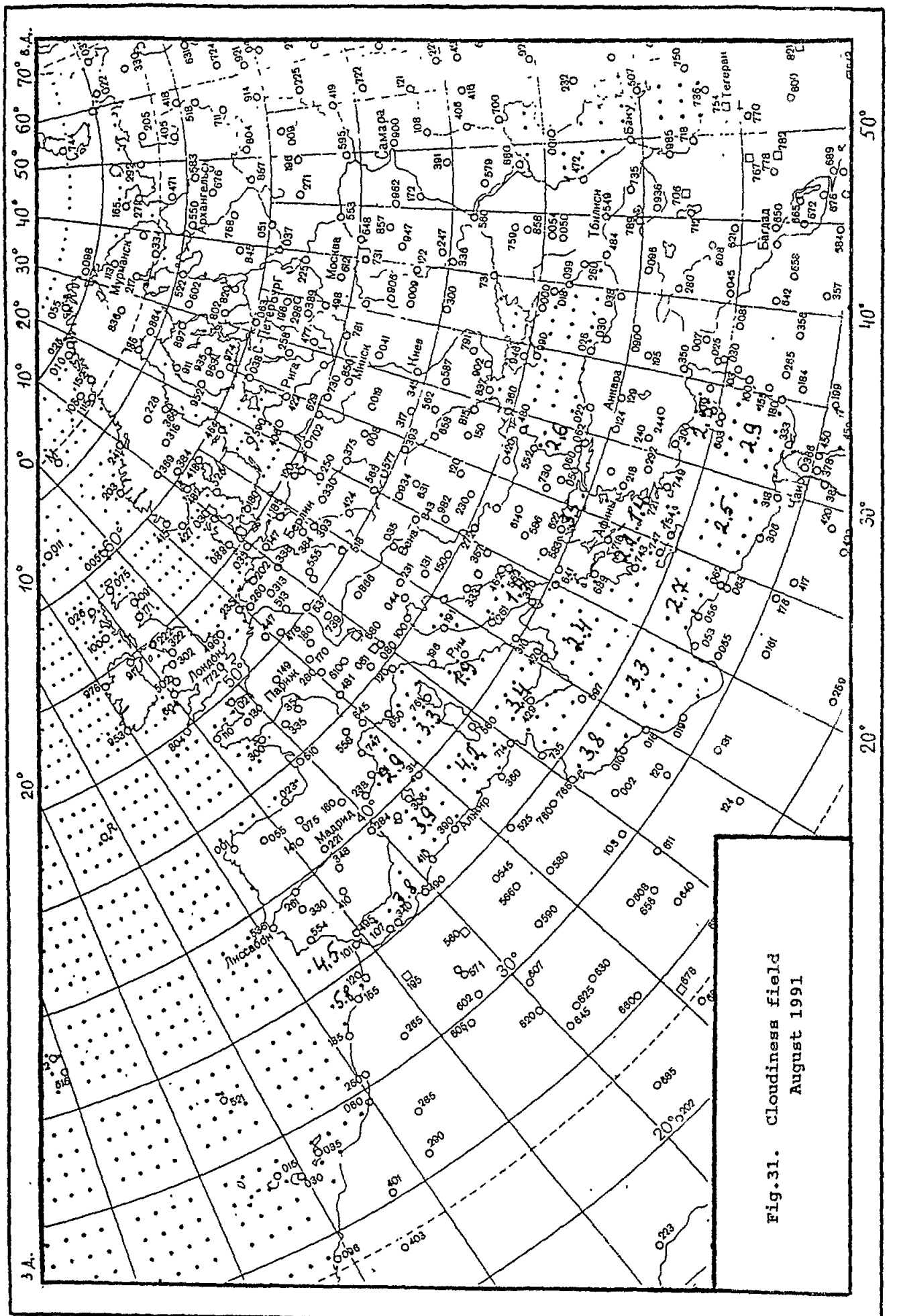


Fig.30. Cloudiness field
July 1991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August 1991

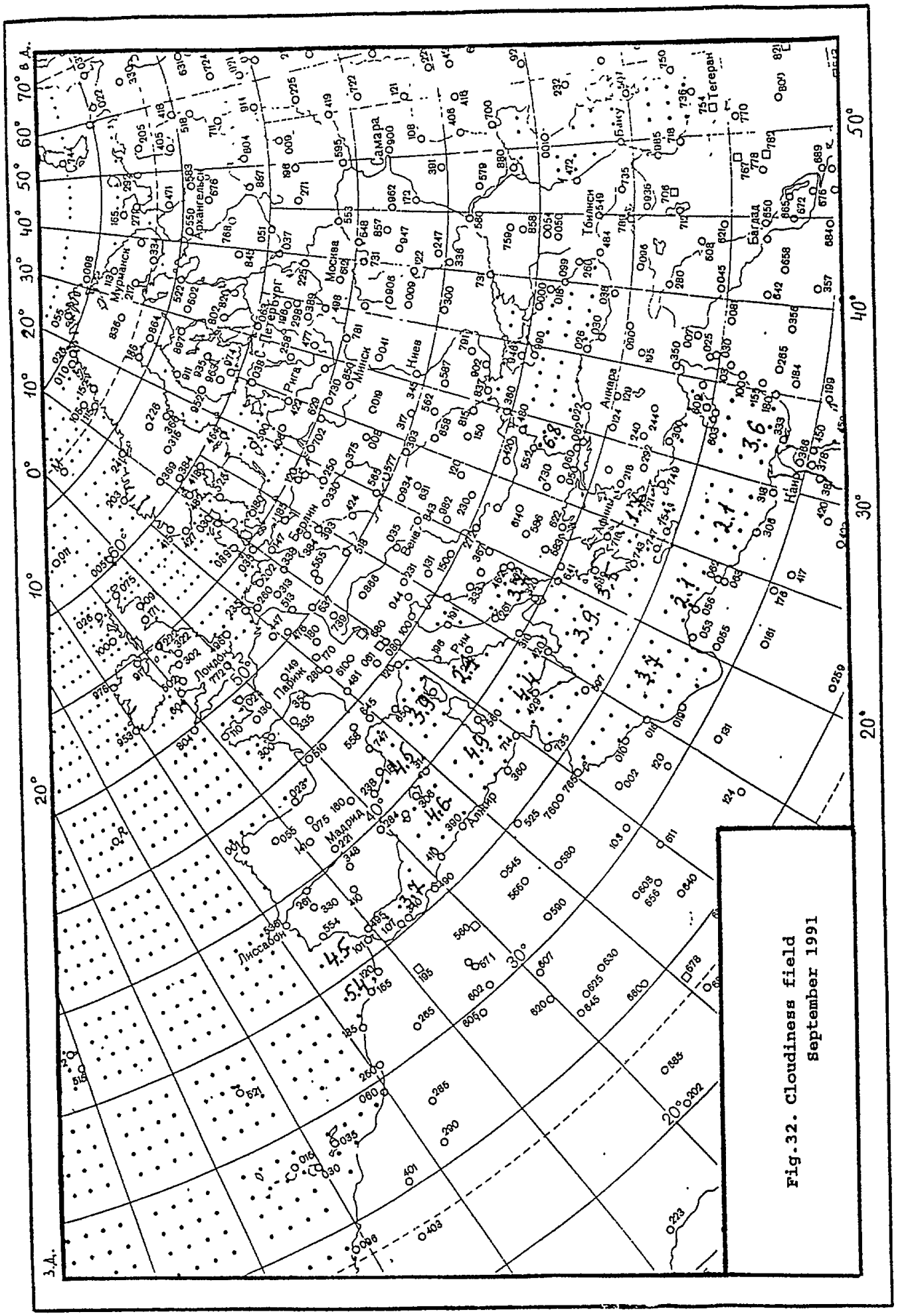


Fig.32. Cloudiness field
September 1991

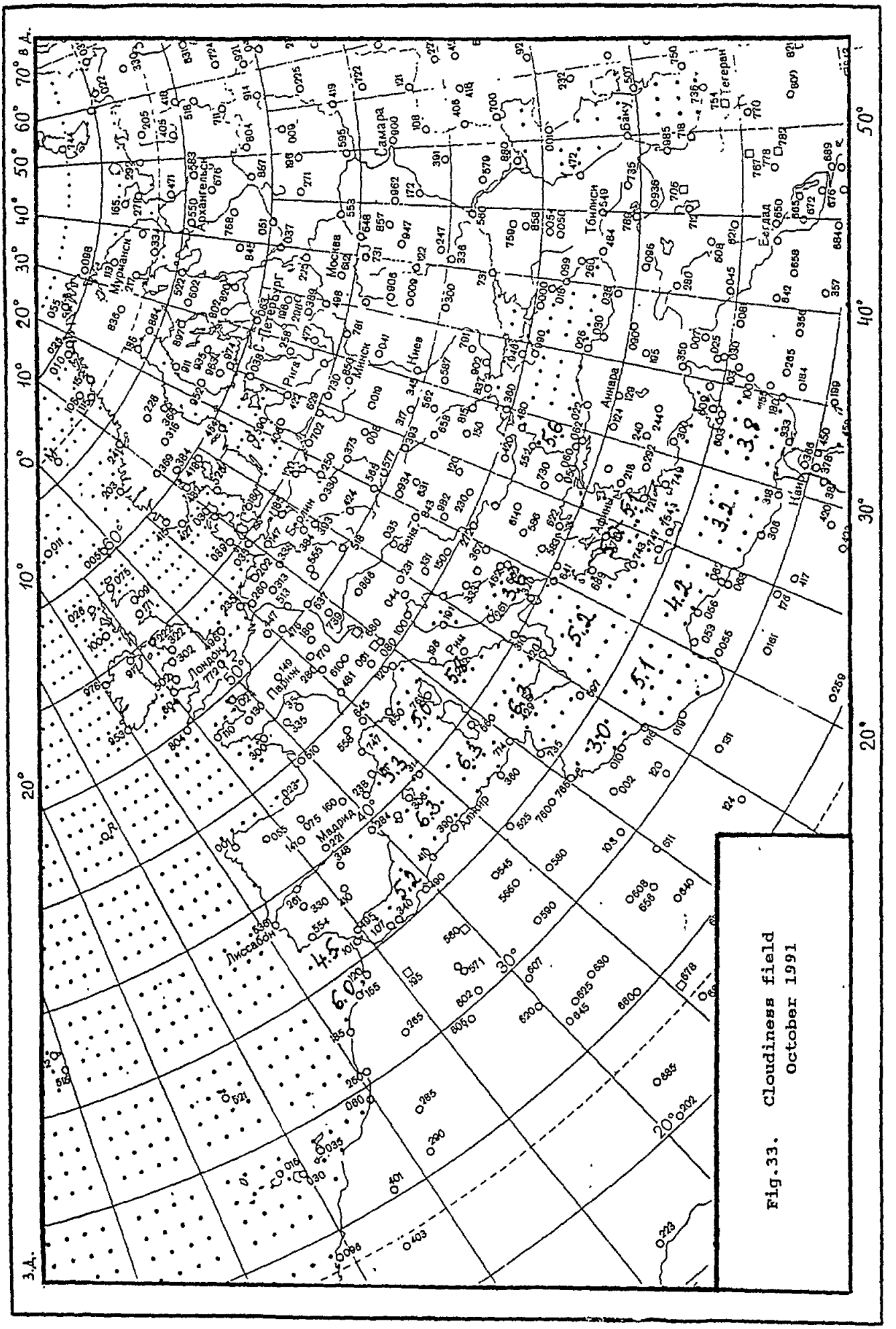


Fig.33. Cloudiness field
October 1991

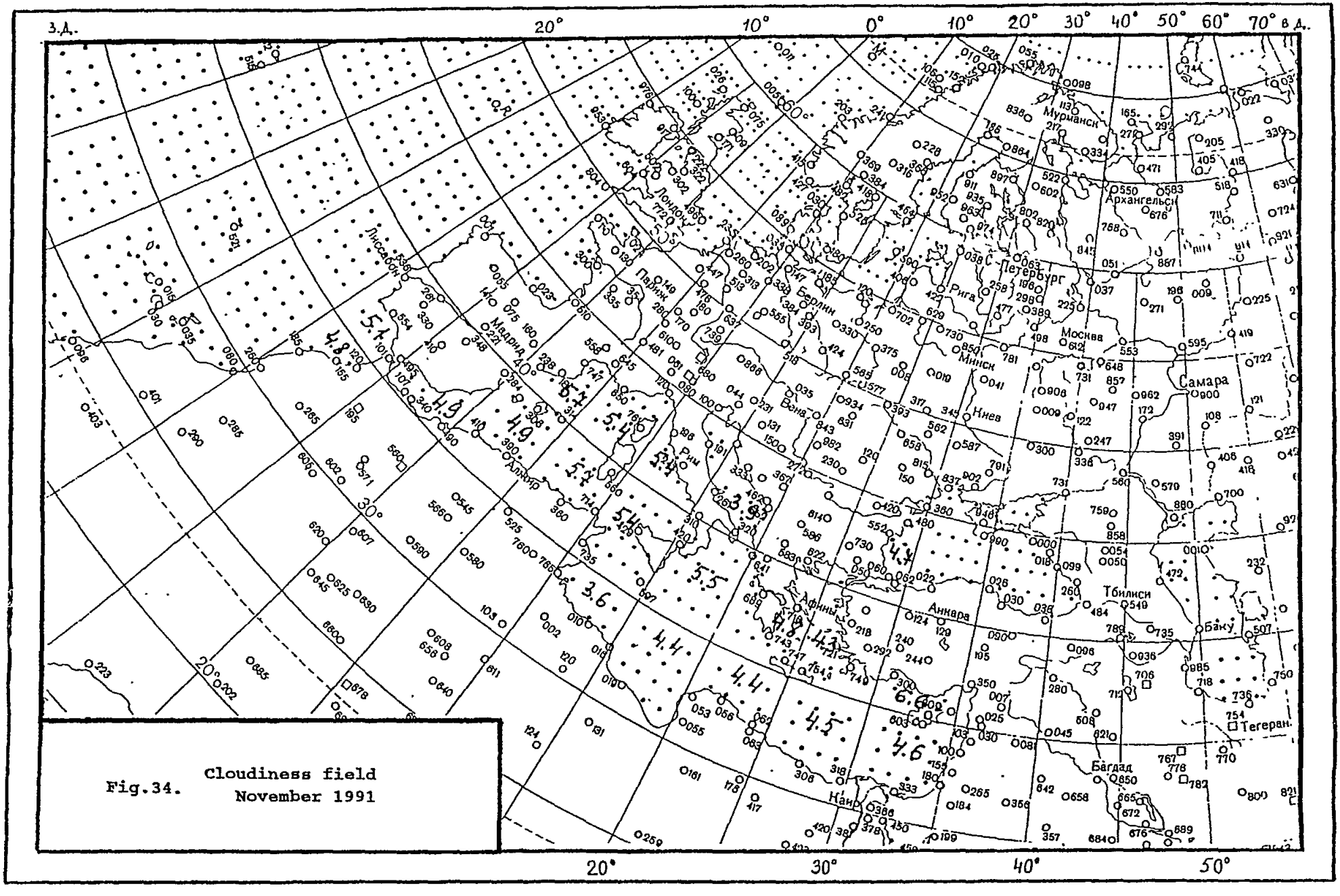


Fig.34. Cloudiness field
November 1991

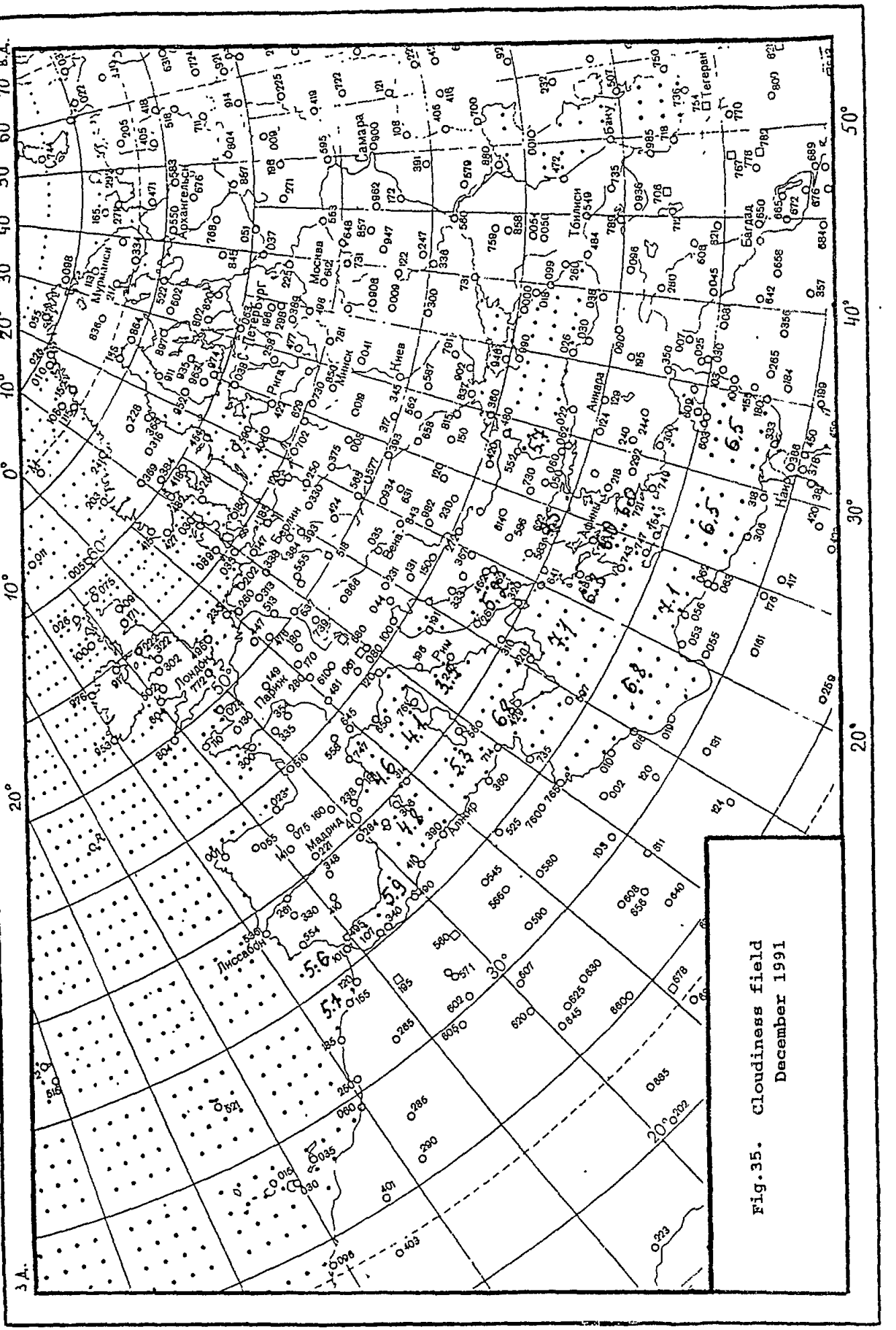


Fig.35. Cloudiness field
December 1991

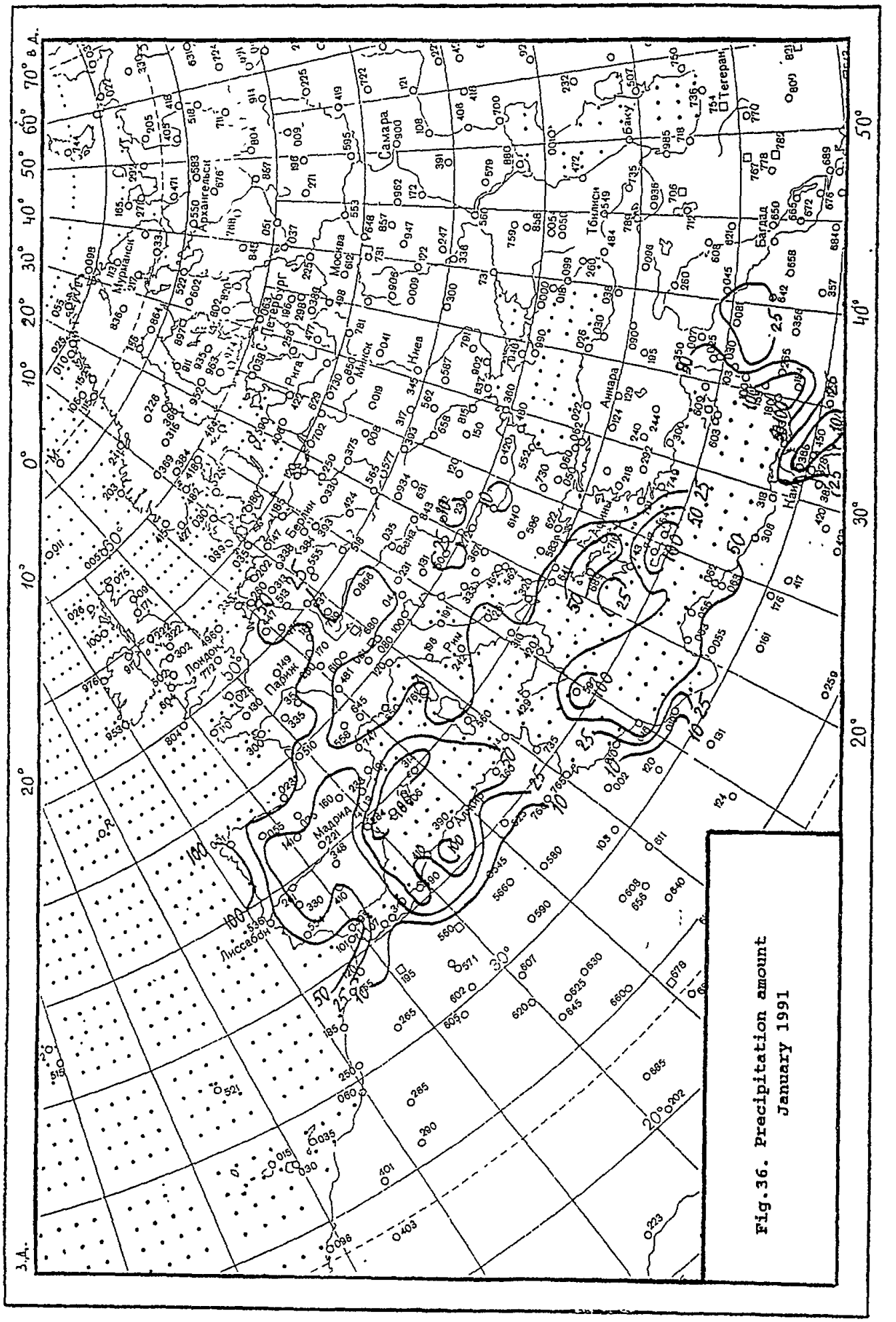


Fig.36. Precipitation amount
January 1991

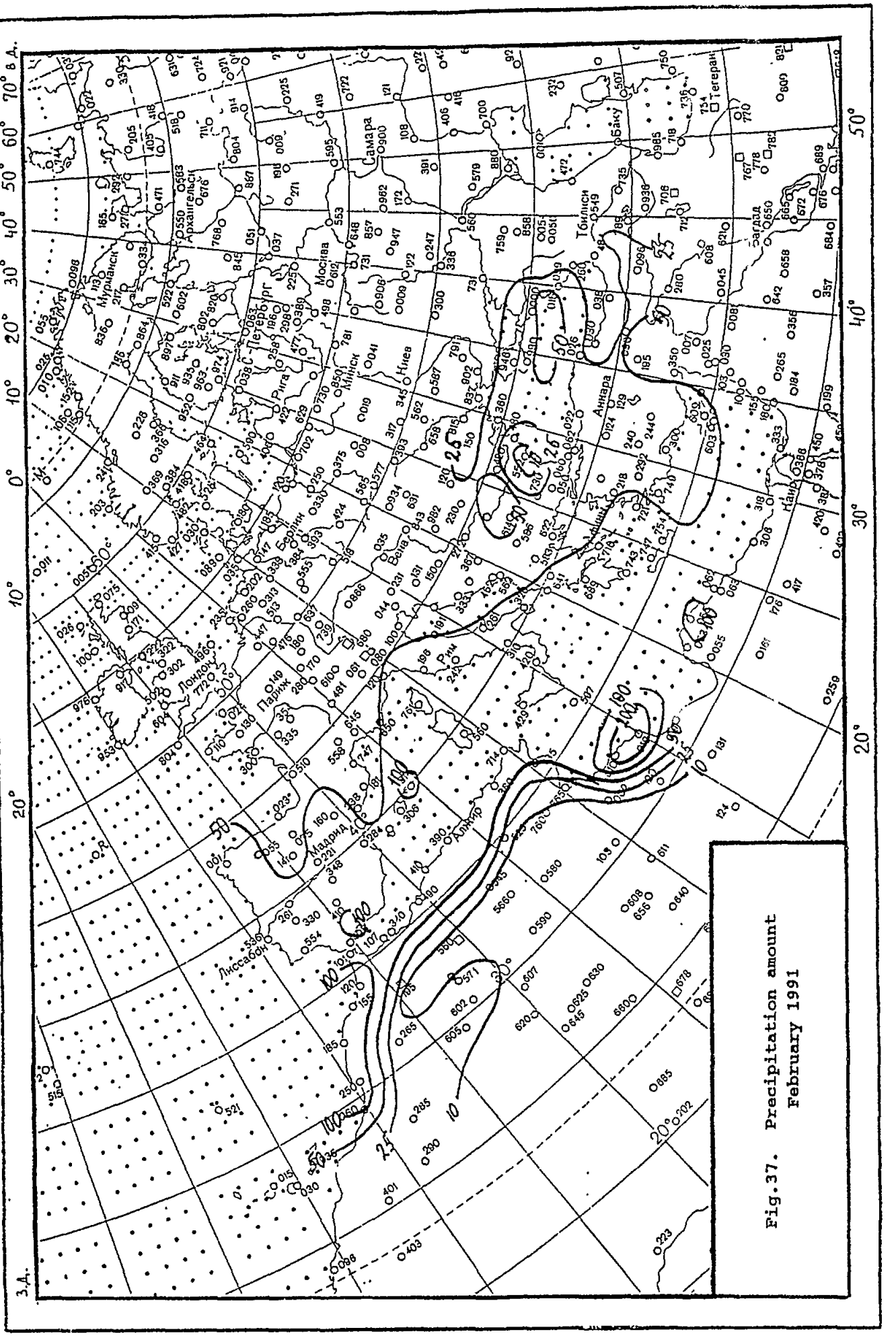


Fig.37. Precipitation amount
February 1991

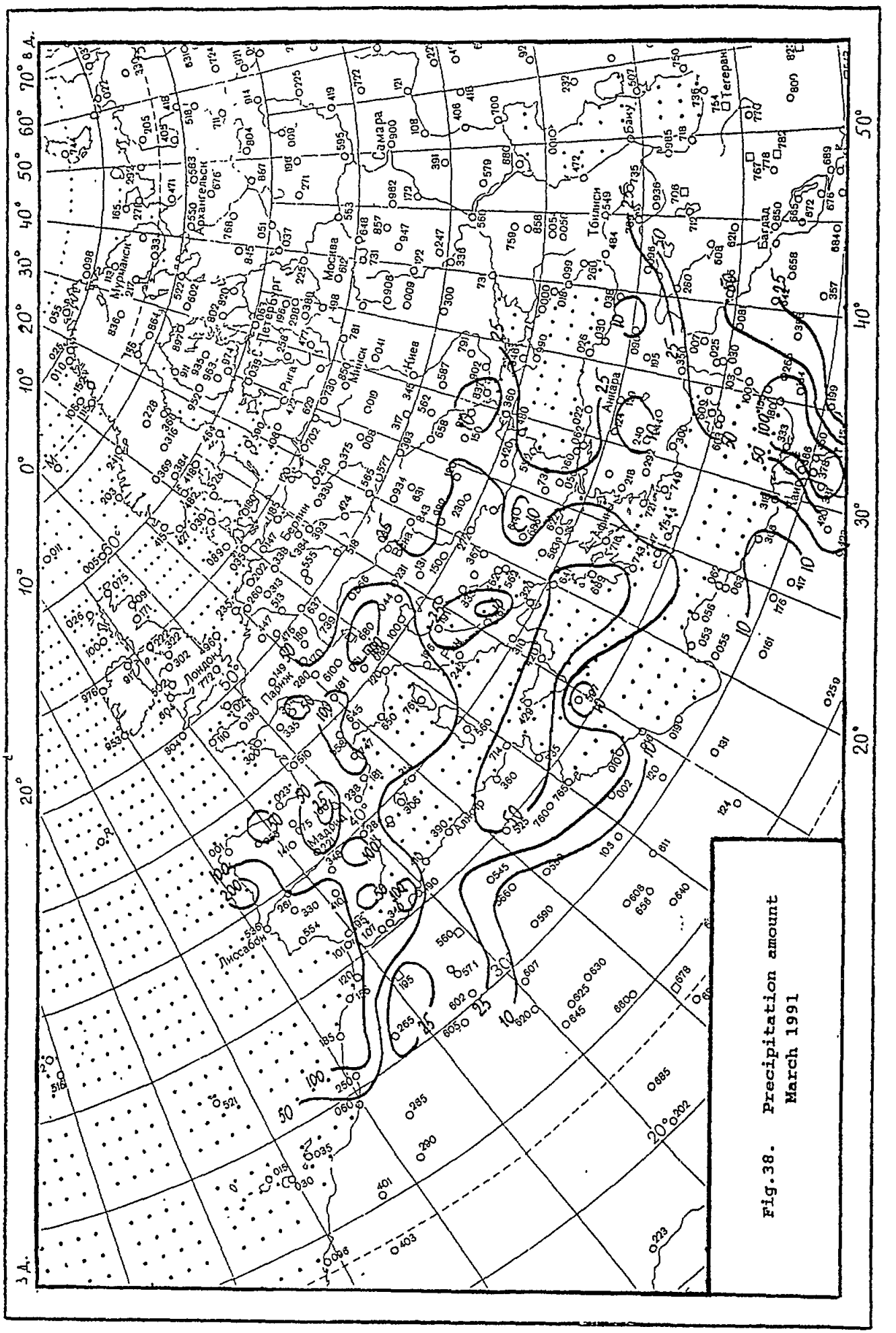


Fig.38. Precipitation amount
March 1991

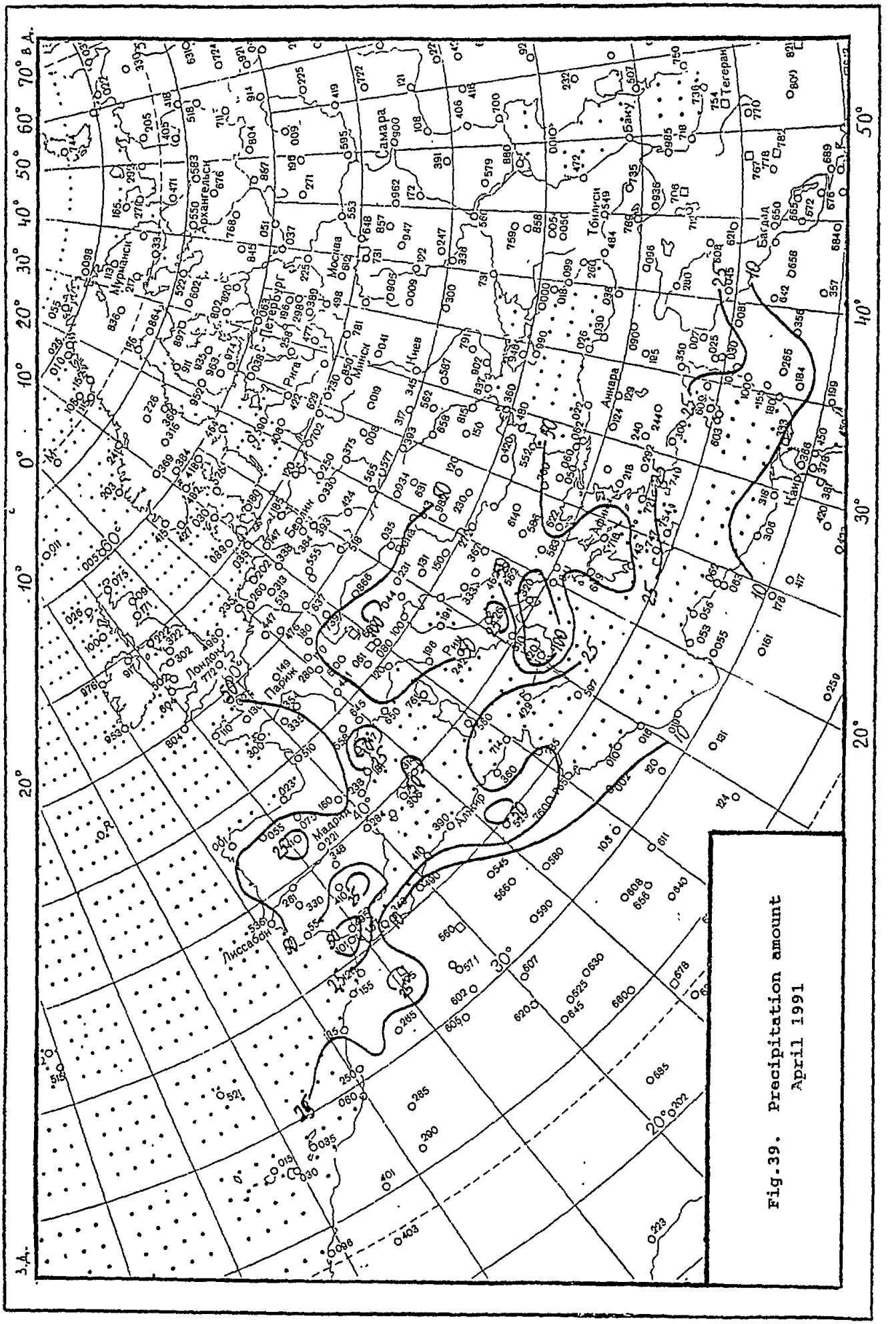


Fig.39. Precipitation amount
April 1991

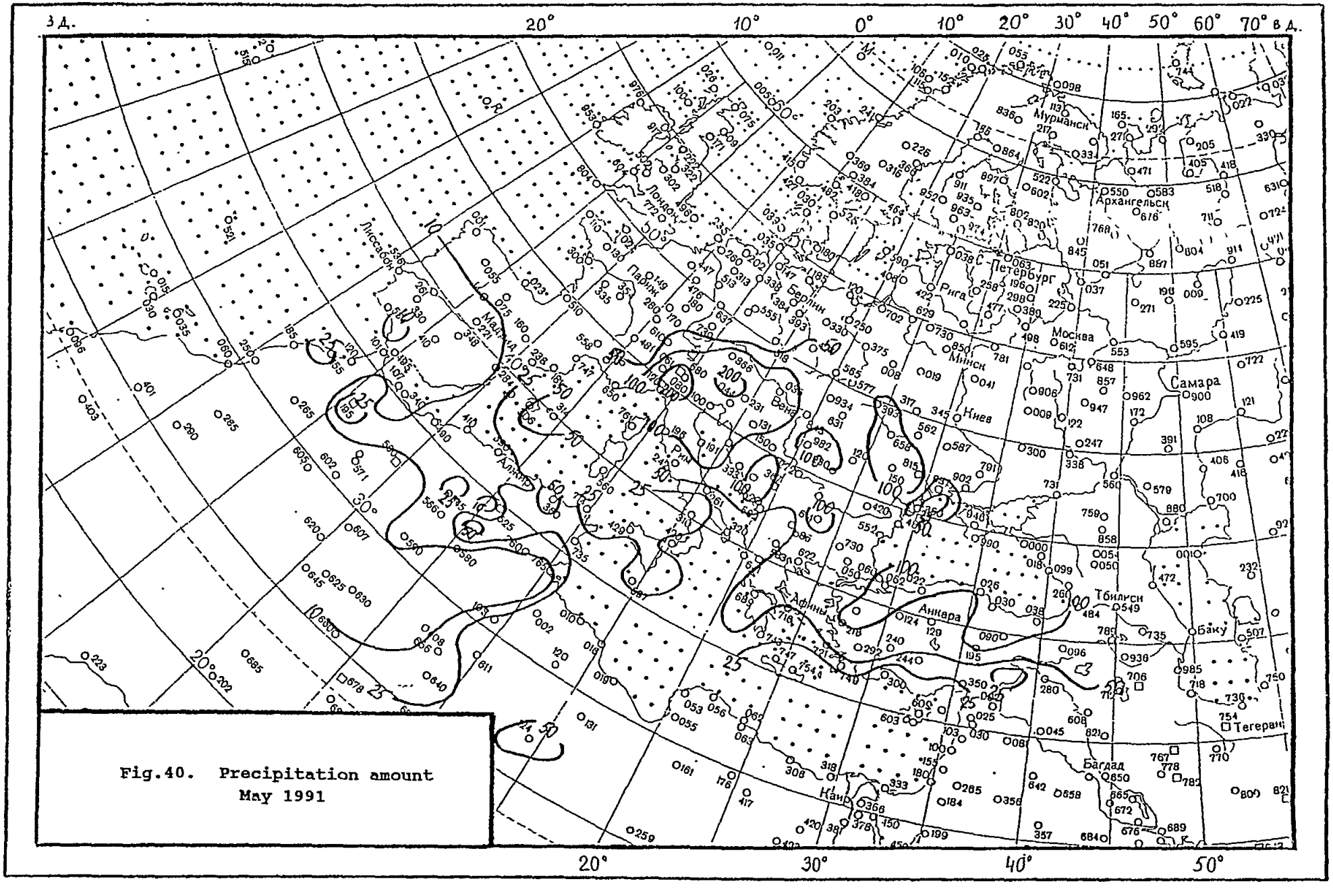


Fig.40. Precipitation amount
May 1991

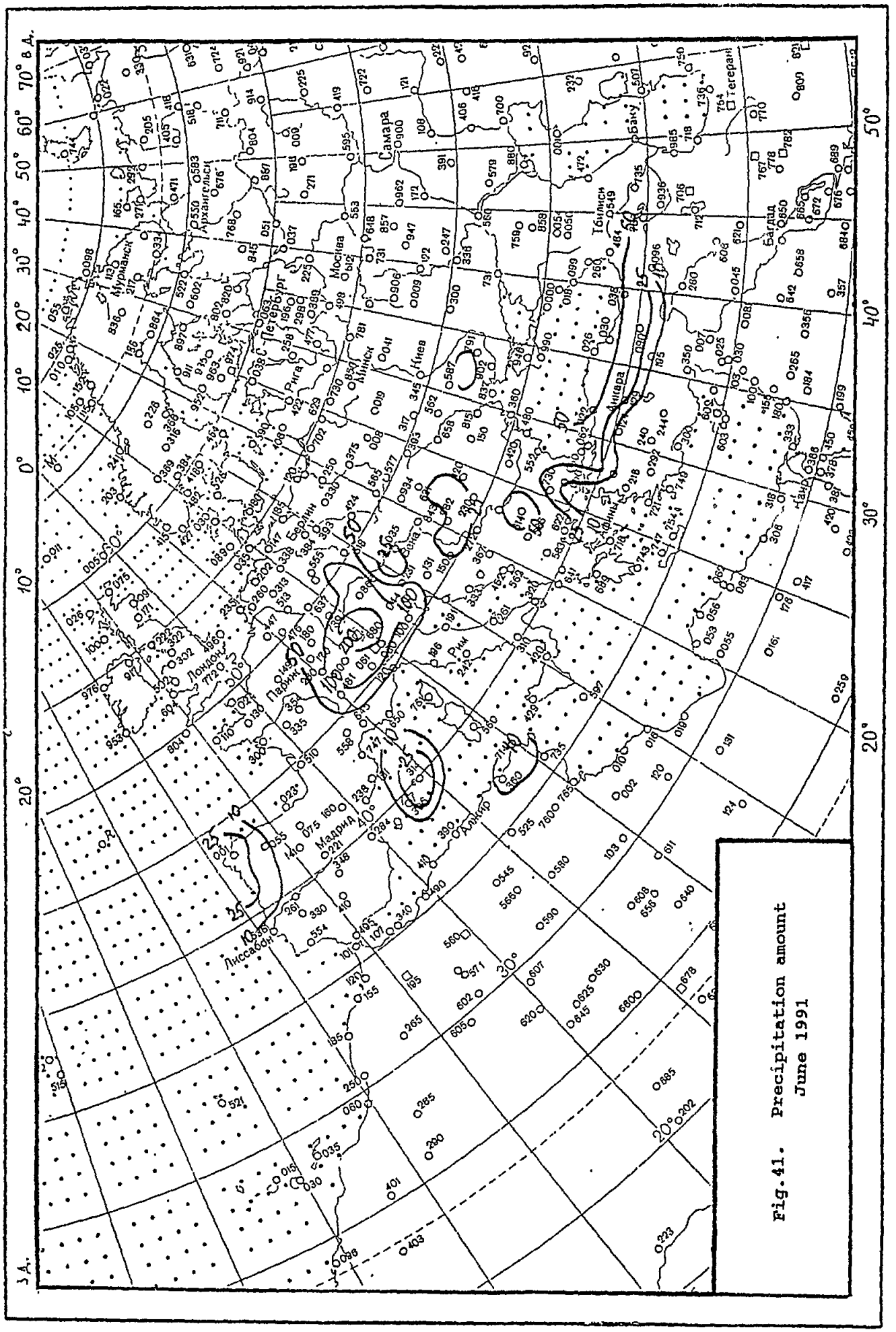


Fig.41. Precipitation amount
June 1991

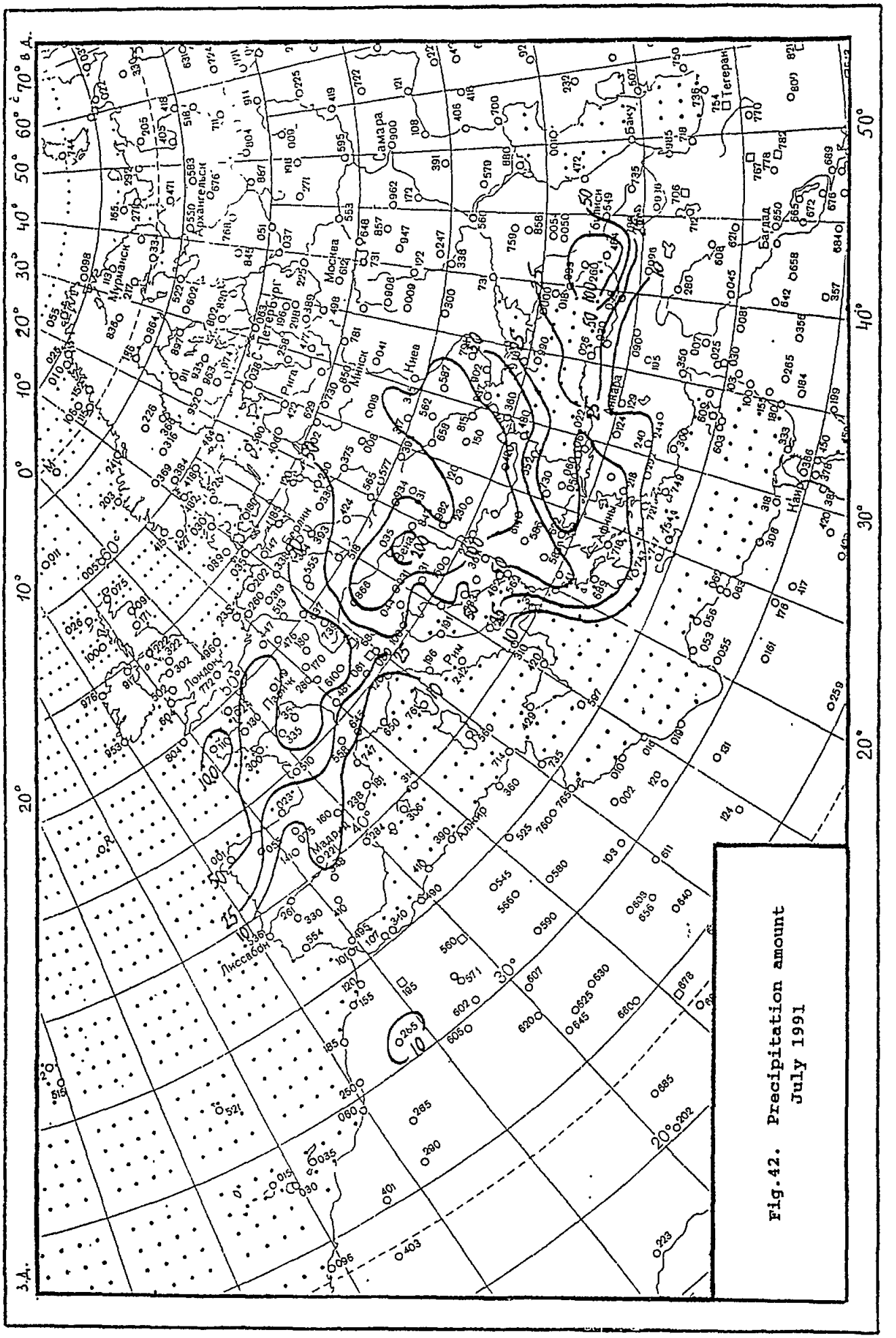
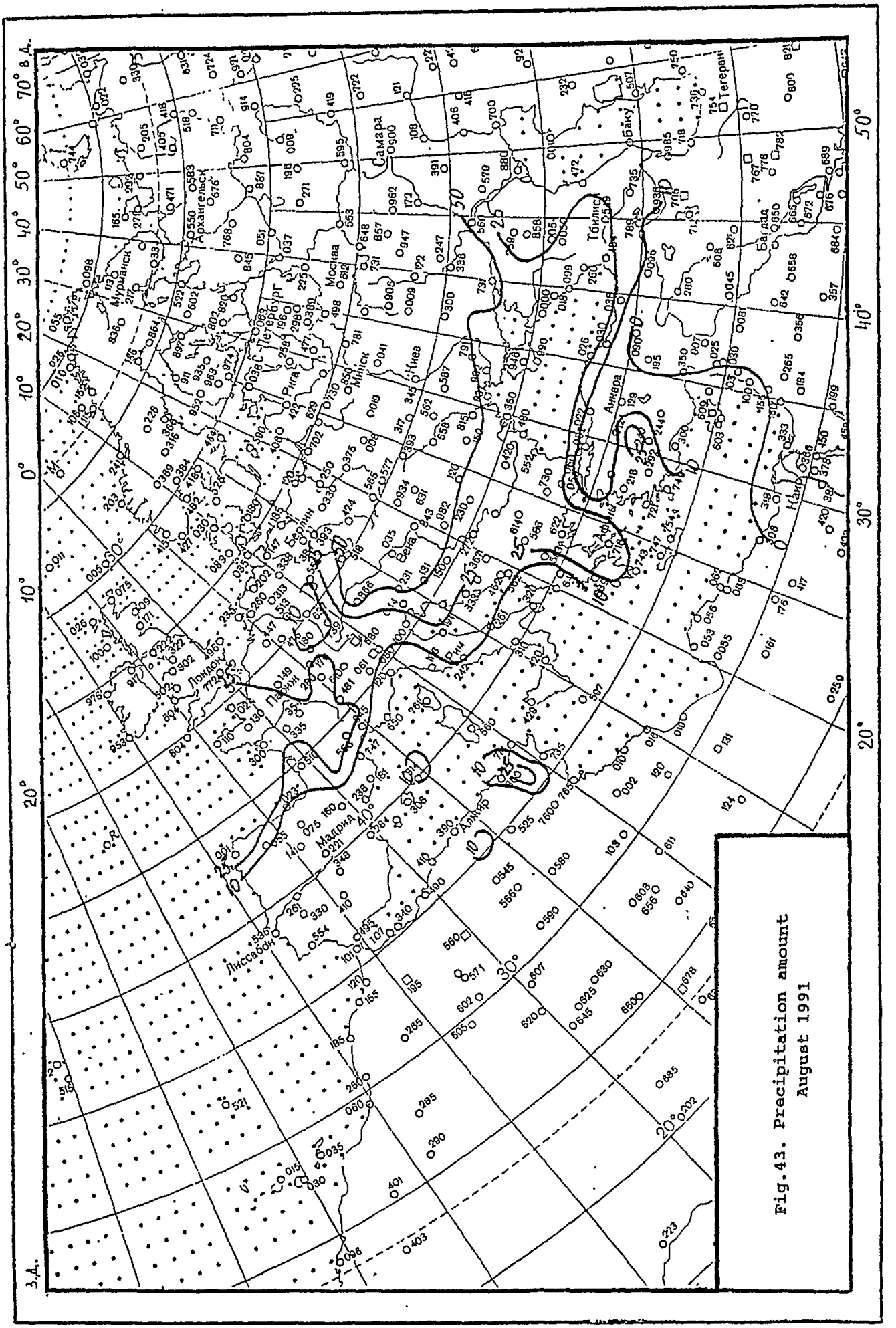


Fig.42. Precipitation amount
July 1991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August 1991

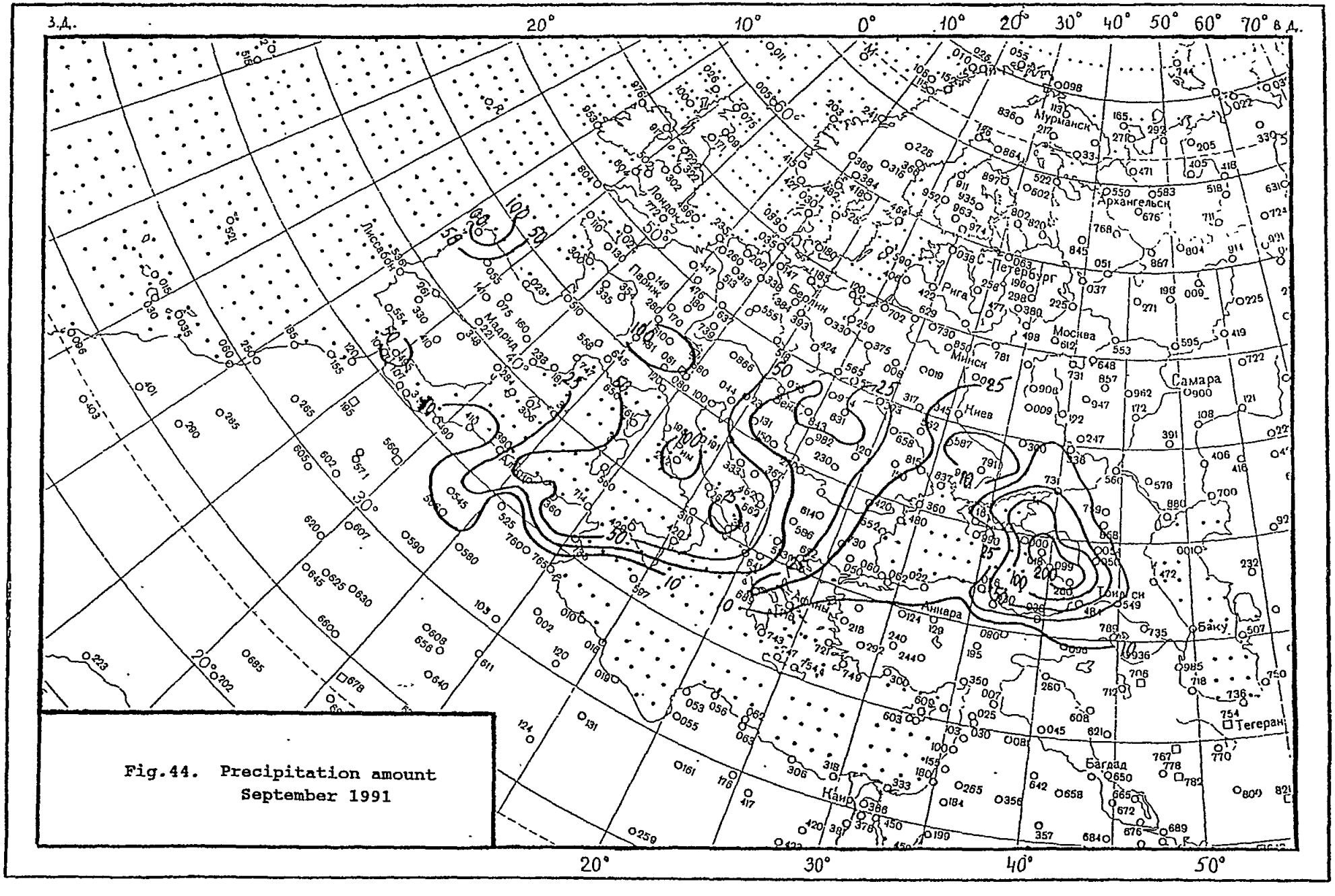
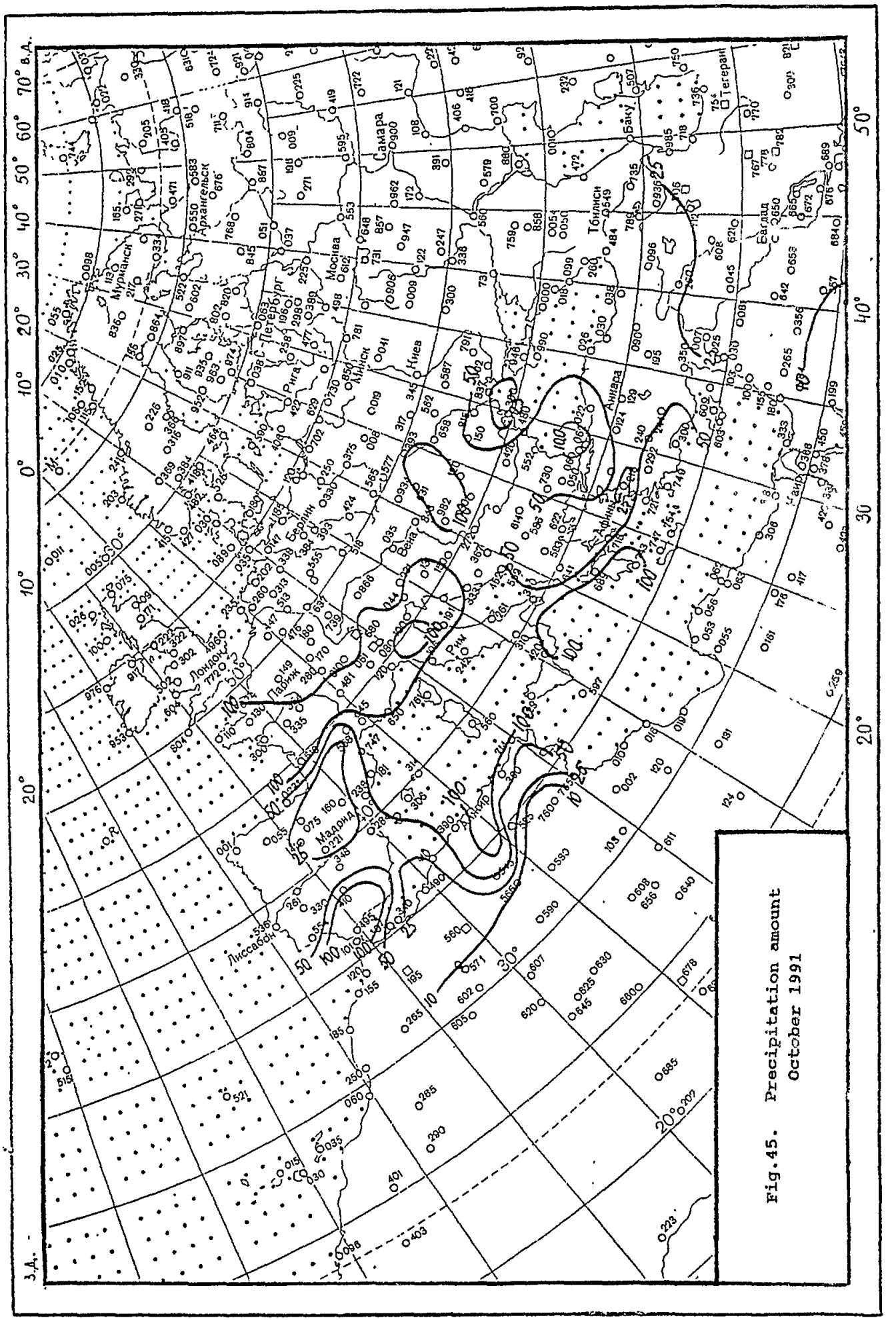


Fig.44. Precipitation amount
September 1991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October 1991

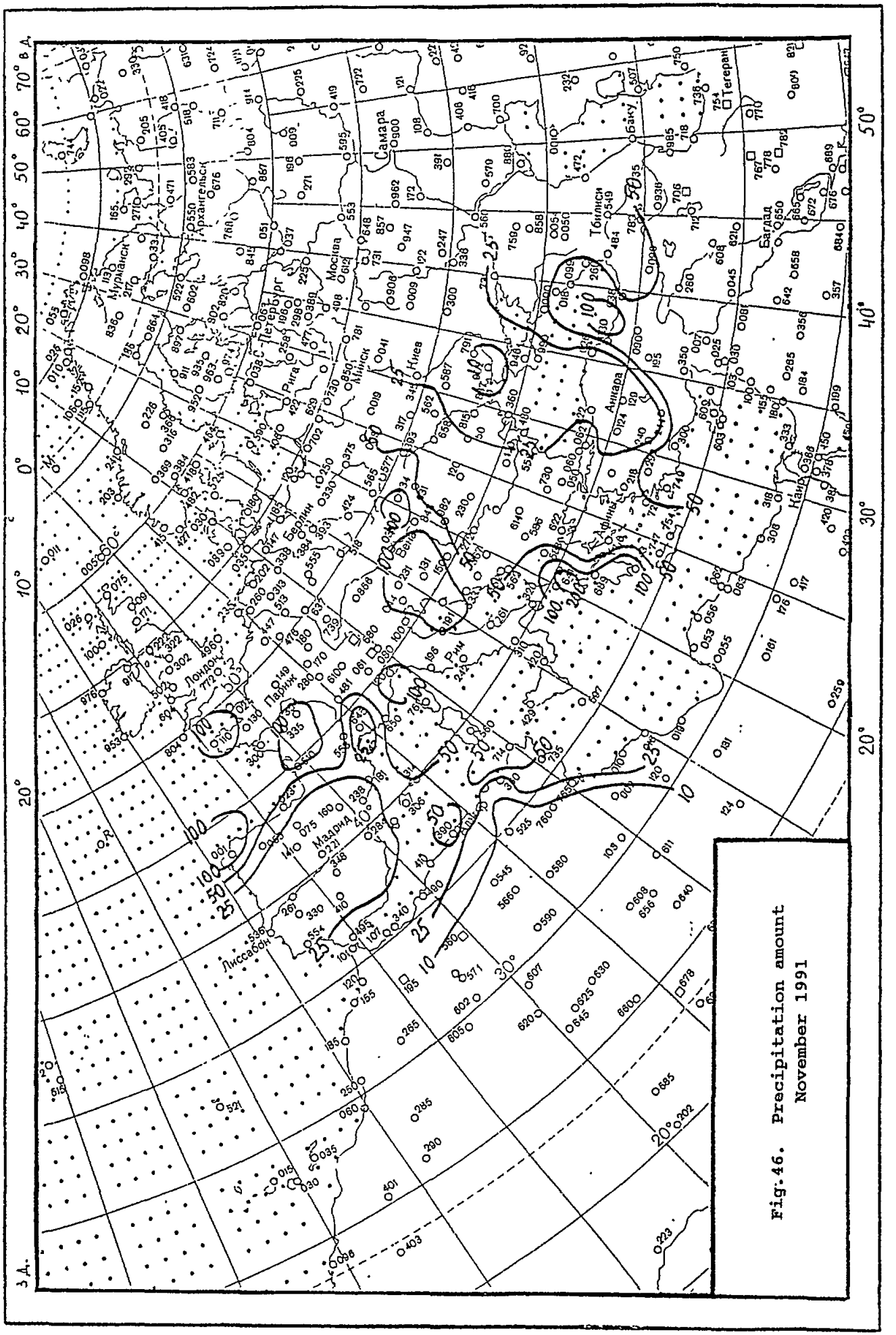
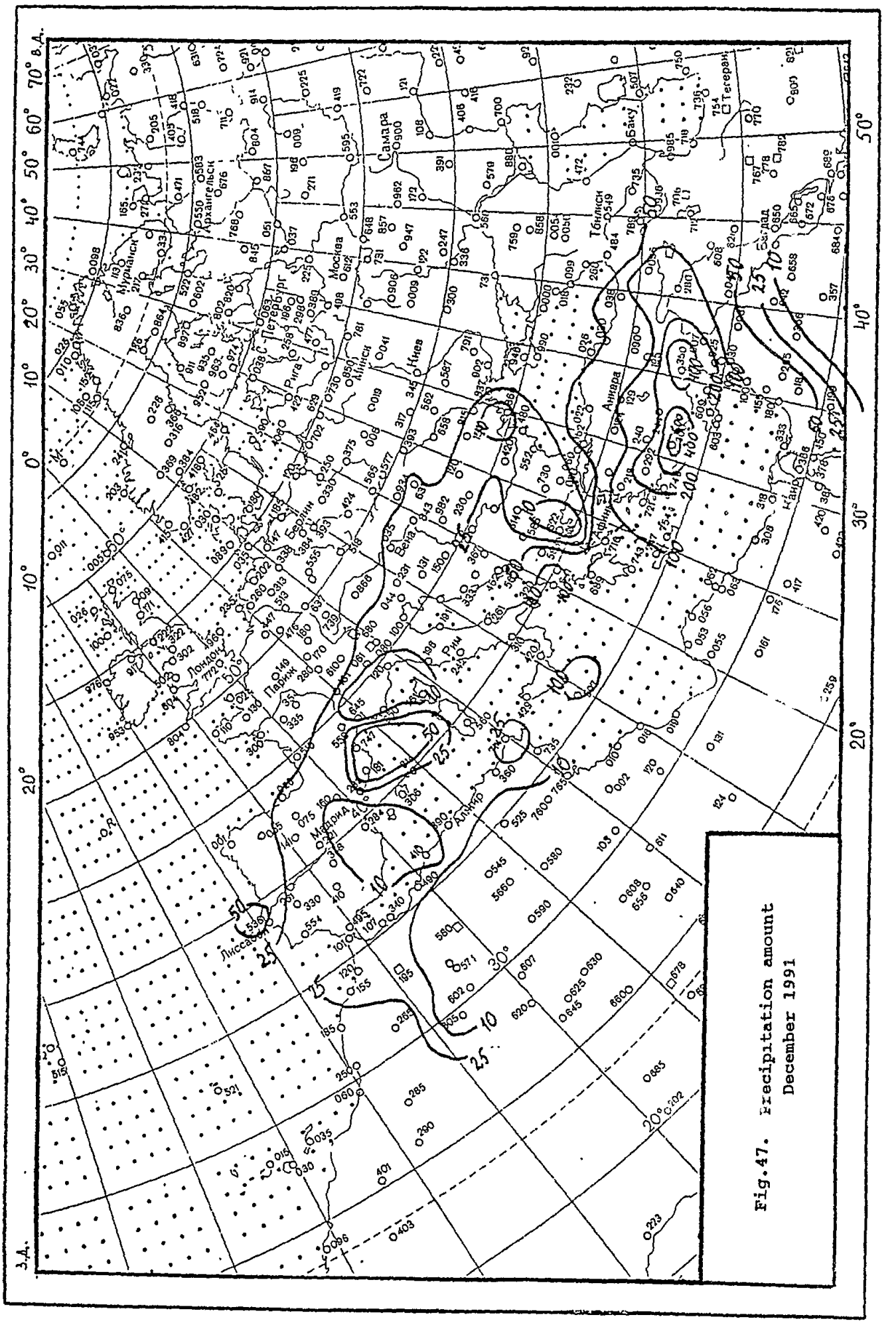


Fig.46. Precipitation amount
November 1991


З.А.
В.А.
70°
60°
50°
40°
30°
20°
10°
0°
Архангельск
Мурманск
Санкт-Петербург
Москва
Самара
Баку
Тбилиси
Тегеран
Рига
Минск
Киев
Берлин
Вена
Париж
Лондон
Рим
Мадрид
Лиссабон
Алжир
Анкара
Каир
Афины
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December 1991

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MTS 83. PNUE/UICN: **Les aires protégées en Méditerranée. Essai d'étude analytique de la législation pertinente.** MAP Technical Reports Series No. 83. PNUE, Centre d'activités régionales pour les aires spécialement protégées, Tunis, 1994 (55 pgs) (français seulement).

MTS 82. UNEP/IUCN: **Technical report on the State of Cetaceans in the Mediterranean.** MAP Technical Reports Series No. 82. UNEP, Regional Activity Centre for Specially Protected Areas, Tunis, 1994 (37 pgs.) (English).

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MTS 75. UNEP/WHO: **Development and Testing of Sampling and Analytical Techniques for Monitoring of Marine Pollutants (Activity A).** MAP Technical Reports Series No. 75. UNEP, Athens, 1993 (90 pgs.) (English).

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MTS 73. UNEP/FAO: **Final Reports on Research Projects Dealing with the Effects of Pollutants on Marine Communities and Organisms.** MAP Technical Reports Series No. 73. UNEP, Athens, 1993 (186 pgs.) (English and French). PNUE/FAO: **Rapports finaux sur les projets de recherche traitant des effets de polluants sur les communautés et les organismes marins.** MAP Technical Reports Series No. 73. UNEP, Athens, 1993 (186 pgs.).

MTS 72. UNEP: **Costs and Benefits of Measures for the Reduction of Degradation of the Environment from Land-based Sources of Pollution in Coastal Areas. A - Case Study of the Bay of Izmir. B - Case Study of the Island of Rhodes.** MAP Technical Reports Series No. 72. UNEP, Athens, 1993 (64 pgs.) (English).

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MTS 61. UNEP: **Integrated Planning and Management of the Mediterranean Coastal Zones. Documents produced in the first and second stage of the Priority Action (1985-1986).** MAP Technical Reports Series No. 61. UNEP, Priority Actions Programme, Regional Activity Centre, Split, 1991 (437 pgs.) (parts in English or French only). PNUE: **Planification intégrée et gestion des zones côtières méditerranéennes. Textes rédigés au cours de la première et de la deuxième phase de l'action prioritaire (1985-1986).** MAP Technical Reports Series No. 61. UNEP, Priority Actions Programme, Regional Activity Centre, Split, 1991 (437 pgs.) (parties en anglais ou français seulement).

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