

MARC: Monitoring and Assessment Research Centre
GEMS: Global Environmental Monitoring System



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A PRELIMINARY EVALUATION OF WMO-UNEP PRECIPITATION CHEMISTRY DATA

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Technical Report

Prepared by:

**GLOBAL ENVIRONMENTAL MONITORING SYSTEM and
MONITORING AND ASSESSMENT RESEARCH CENTRE
Chelsea College, University of London**

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**A preliminary evaluation of WMO-UNEP
precipitation chemistry data**

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A Technical Report (1980)

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CONTENTS

Introduction	1
Quality of data	2
Area distribution of the annual mean averaged for 1972–76	3
Europe	4
North America	6
Nitrates in precipitation	7
Conclusions	7
Figure 1 Annual mean concentration of sulphate in precipitation, Europe, 1972–76	9
Figure 2 Annual mean concentration of sulphate in precipitation, Europe, 1954–59	10
Figure 3 Mean concentration of sulphate in precipitation, North America, 1972–76	11
Figure 4 Excess of sulphate in precipitation, North America, 1955–56	12
Figure 5 Mean concentration of nitrate in precipitation, Europe, 1972–76	13
Figure 6 Mean concentration of nitrate in precipitation, North America, 1972–76	14
Table 1 Monthly means of sulphate in precipitation	
Table 2 Annual means of sulphate in precipitation, Europe, 1972–76	
Table 3 Annual means of sulphate in precipitation, North America, 1972–76	
Table 4 Annual means of nitrates in precipitation, Europe, 1972–76	
Table 5 Annual means of nitrates in precipitation, North America, 1972–76	

Introduction

In systematic studies of atmospheric chemistry, background levels of the chemical composition of the atmosphere are often evaluated from measurements of various constituents in precipitation. International networks for studies of atmospheric chemistry, involving measurements of the chemical content in precipitation, were established as early as the mid-fifties, mainly in countries in the north and west of Europe under the auspices of Professor C. Rossby at the University of Stockholm, and at about the same time in the United States under Professor C. Junge at Cambridge, Massachusetts.

However, it was not until the mid-sixties that scientists became more generally concerned with the possibility that the chemical composition of the atmosphere might be changed through man's activities and that measurements of various constituents in the atmosphere might be important for recognizing and understanding changes in the global environment. Awareness of the special problem of increasing CO₂ in the atmosphere existed before this, however, and atmospheric CO₂ has been monitored at the Mauna Loa Observatory in Hawaii since 1958.

Interest in man's impact on the atmosphere through the burning of fossil fuels gradually led to increasing international awareness that efforts must be made to monitor the composition of the atmosphere on a continuing basis. Hence in 1970 the World Meteorological Organization (WMO) launched its still ongoing project to establish a global network of stations to monitor atmospheric constituents at the background level – Background Air Pollution Monitoring Network (BAPMoN). This project was supported by the United Nations Environment Programme (UNEP) from 1974 onwards as an important component of the Global Environmental Monitoring System (GEMS). This network includes three types of stations:

- (a) "regional stations" located in areas not influenced by short-term fluctuations in pollution which provide data on precipitation chemistry, atmospheric particulates and turbidity (at present there are about 100 such stations around the globe);
- (b) "baseline stations" located far away from pollution sources which measure, with more sophisticated means and in very clean air, the same variables as the regional stations but, in addition, CO₂ and various pollutants in air;

- (c) regional stations with expanded programmes which make similar measurements as baseline stations and are most often located on continents.

Data from this network have been collected since 1972 under the auspices of the Environmental Protection Agency (EPA) and the National Oceanic and Atmospheric Administration (NOAA) in the U.S.A. and have been published by them for the period 1972–76 in co-operation with WMO and UNEP*.

In this paper a preliminary evaluation is made of some of the precipitation chemistry data available from the network for the period 1972–76. It is expected that a more thorough analysis of the quality of the data and the existing trends will be arranged through WMO in the near future.

It should be kept in mind that during the period 1972–76, which was when the network was being built up, the only stations operational with regard to precipitation chemistry were in Europe and North America; stations in other continents gradually became operational only towards the end of the period. The evaluation of the situation is therefore limited to the European and the North American continents.

Quality of data 1972–76

Considerable deficiencies in quality were expected during the early years of operation, when no intercalibration of methodology had yet been undertaken and analytical standards had not yet been uniformly developed. The first intercalibration in fact took place in 1976.

In discussing the quality of data one should perhaps first consider whether the sampling technique applied leads to obtaining absolute truth about precipitation chemistry. However, no such studies have so far been made. Our discussion, therefore, can deal only with the relative reliability of the data, i.e. to what extent they render a consistent and reasonable statistical sample.

Another question of importance became evident in evaluating the statistical quality of the data published in the joint publication. As indicated in the text of each annual issue, there has been some

*Global Monitoring of the Environment for Selected Atmospheric Constituents. National Climatic Centre, Federal Building, Asheville, N.C. 28801, U.S.A.

confusion as to what extent sulphate, ammonium and nitrate content in precipitation has been reported in elemental units or in values converted to sulphur and nitrogen through multiplying the original data by 0.33, 0.78 and 0.23 respectively. As the data publications do not give a clear picture whether such conversion has taken place or not, the published data are difficult to compare and hence peculiarities occurred in the first calculations. However, through inquiries in the countries concerned, the matter was clarified and it became clear that most of the published data were not converted but published in elemental units, e.g. sulphate and nitrate.

In most earlier presentations of precipitation chemistry data, the data have also been given in elemental units. In order to obtain overall comparability not only from country to country but also with earlier presentations of data, it was decided to recalculate those data which had been converted into sulphur and nitrogen so that in this paper all data are given as sulphate and nitrate. In particular, published data from Canada for the years 1973–76 and from the U.S.A. for 1975 and 1976 have been recalculated into elemental units.

In order to get a feel for the general statistical quality of the sulphate results, data expressed in $\text{mg SO}_4^{2-} \ell^{-1}$ were first analysed for 20 stations which had been in operation for more than three years during the period 1972–76. Table 1 gives the maximum and minimum monthly values of sulphate in precipitation observed in winter (October–March) and summer (April–September) together with their mean and standard deviations. Nine stations are from Europe, seven from the U.S.A. and four from Canada.

The first conclusion to be drawn from a look at Table 1 is that maximum values of sulphate content are higher in the U.S.A. than in Europe and Canada. This suggests that single events with high sulphate yield are more common in the U.S.A. than in Europe and cause considerable dispersion in the distribution of values around the mean. This fact is also revealed through the higher values of standard deviations relative to the mean obtained at the stations in the United States compared with those in Canada and Europe.

Area distribution of the annual mean averaged for 1972–76

The annual mean concentrations of sulphate in precipitation from

the European and North American stations based on available monthly values are given in Tables 2 and 3 respectively. Values have been put in parenthesis when they seem to be unreasonable. Overall annual mean figures for the period 1972–76 are given in the last column of the tables calculated from the yearly values available. Some of these are therefore based upon five years while others may be based on only two or even one year. In this preliminary evaluation no attempt has been made to interpolate values from years from which data are not available. Obviously the mean values given in the column are therefore not exactly comparable.

In Tables 2 and 3, two annual values are given for some stations. The reason for this is related to the problem of sea spray. As is well known, stations situated on the coast or on islands are greatly influenced by sea spray. In order to make the values from such stations comparable with those from stations over the continents from the point of view of anthropogenic sulphate content, they have to be reduced by the sulphate amount stemming from the sea-salt. It has long been customary (Junge 1960) to calculate the sea-salt sulphate concentration which is to be subtracted from the excess values of coastal stations by multiplying the values of chloride concentration from the same stations by 0.046. This was done in our case also. We have limited ourselves to carrying out this correction for stations reasonably close to the coast, because at stations further inland the correction is negligible for practical purposes. As seen from the two values presented for coastal stations, the correction may, however, be considerable at coastal or island stations.

The annual mean concentration of sulphate in precipitation expressed in $\text{mg SO}_4^{2-} \text{ l}^{-1}$ as given in Tables 2 and 3 has been presented on maps of Europe and North America in Figures 1 and 3.

Europe

With regard to Europe, it should be noted that only 25 stations could be plotted for the whole area. This is a relatively small number for accurate mapping but the general picture is so well known from maps previously published (to which further reference is made below) that it was possible to draw a reasonable map for the situation in the years 1972–76. Some data for part of the same period were also

obtained from stations participating in the Organisation for Economic Co-operation and Development (OECD) study on long-range transport of air pollutants in western Europe. They were used to check the concentrations in areas of the map where data were scarce. Some BAPMoN data from France for the year 1977 have also been included to check the geographical distribution.

Some data showing very high content of sulphate are doubtful. Values from the Faroe Islands are obviously too high, probably due to the stronger influence of sea spray than is obtained from the reduction by use of the average chlorine content. Likewise it is obvious that the values from Neuglobsow in G.D.R. are too high to reflect real background conditions. The station seems to be influenced too much by local pollution from nearby Berlin.

It seems to be possible to conclude from Figure 1 that existing data from the WMO/UNEP network give a reasonable, consistent and coherent picture of the distribution of sulphate in precipitation over Europe. The mean background sulphate level gradually increases from around $0.5 \text{ mg } \ell^{-1}$ in the north of Scandinavia to around $5.0 \text{ mg } \ell^{-1}$ in the maximum area in central Europe, which extends from the south of England over northern France, Germany and Poland to part of the Soviet Union. Moving south from this area, values again decrease and remain around $1.5 \text{ mg } \ell^{-1}$ as far south as northern Italy. Obviously the situation is somewhat uncertain in parts of south-western and eastern Europe due to lack of data but there is no reason to question the general picture.

It should be emphasized that the maps give the content of sulphate in precipitation and not the deposition of sulphate. Because the aim of BAPMoN is to use precipitation chemistry as an indication of the existence of pollutants in the atmosphere, only the observed data have been used here. No attempt is made to estimate deposition, since this would involve a number of assumptions that cannot be discussed in this presentation.

It is interesting to try to compare the pattern observed over Europe with earlier attempts to study the chemical content of precipitation. In 1963 de Bary and Junge published data on sulphate in precipitation over Europe for the period 1954–59. The maps given in that paper are based on data from a much larger number of stations than are

available in the WMO network and refer separately to summer and winter. In order to obtain a comparable picture, Junge's original data as presented in their paper are used as far as possible for the same or nearby stations as the ones used in Figure 1.

Comparing the situation in the fifties with the seventies reveals the following interesting facts (see Figures 1 and 2):

- (a) there is hardly any increase in sulphate content from the fifties to the seventies in northernmost Scandinavia;
- (b) in southern Scandinavia the increase is of the order of 50–100 per cent i.e. from 1.0 to 1.5–2.0 mg $\text{SO}_4^{2-} \ell^{-1}$;
- (c) in the maximum area in central Europe there is an increase of 100 per cent i.e. from about 2.3 to 5.0 mg $\text{SO}_4^{2-} \ell^{-1}$;
- (d) in south-central Europe the increase is also around 100 per cent i.e. from about 1.0 to 2.0 mg $\text{SO}_4^{2-} \ell^{-1}$;
- (e) the mean value for Europe as a whole, based upon the data used from de Bary and Junge, is 1.28 compared with 1.88 mg $\text{SO}_4^{2-} \ell^{-1}$ for the BAPMoN data, indicating an average increase for the whole area of about 50 per cent.

Interestingly enough, the increase in emission of sulphur over Europe from 1960 to 1972 has been estimated to be of the same order of magnitude (Field 1976).

The 1972–76 values can also be compared with other studies made during the seventies in Europe. Granat (1978) has recently presented a picture based upon data from the same network that de Bary and Junge used in 1963, and in comparing the areas of Europe which are common to our map, one finds reasonably good agreement.

North America

The general distribution of sulphate in the atmosphere over the North American continent is presented in Figure 3. The area of maximum concentration of sulphate in precipitation is located in the north-east of the U.S.A. and eastern Canada with values reaching about the same as in the maximum area of Europe, namely around 5 mg $\text{SO}_4^{2-} \ell^{-1}$. The minimum values are found in the north-west of the U.S.A. and in Canada of around 0.5 mg $\text{SO}_4^{2-} \ell^{-1}$.

It is worth comparing the distribution of sulphate in the atmosphere

over the United States with that obtained by Junge (1960) for the same area in 1955–56 reproduced in Figure 4. There is no doubt that the area of highest concentration of sulphate in the seventies was the same as in the fifties, located over the north and east of the United States and the east of Canada, around the Great Lakes. The values obtained by Junge in the fifties are not very much lower than those shown by the BAPMoN data for the period 1972–76. This seems to indicate that the rate of increase of sulphate in the atmosphere since the fifties in the United States has been somewhat slower than in Europe, notwithstanding the fact that the levels in the fifties were about double those in Europe.

Nitrates in precipitation

In addition to the above preliminary but somewhat detailed study of sulphate in precipitation, a first attempt has also been made to analyse the nitrate situation. Although the quality of the data has not yet been studied, maps have been prepared of the distribution of annual means of nitrates over Europe and North America, as has been done for the sulphates. In doing so, it is assumed that the quality of the nitrate data (see Tables 4 and 5) would be similar to that for the sulphates. This has had to be left for later confirmation. In the maximum area of Europe (see Figure 5) which is located similarly to that for sulphates, values up to $3.30 \text{ mg NO}_3 \ell^{-1}$ are found. In the minimum area, the values are about $0.25 \text{ mg NO}_3 \ell^{-1}$. In North America (see Figure 6) the maximum area of more than $2.5 \text{ mg NO}_3 \ell^{-1}$ covers the Great Plains in both the United States and Canada, stretching all the way to the east coast. It is possible that this area of high concentration is partly due to intense use of nitrate fertilizers required by the large-scale agricultural activities in the Great Plains. The values over the United States are more evenly distributed than over Europe but, surprisingly, the values are often higher in Europe than in the United States.

Conclusions

One may conclude from this preliminary evaluation that the precipitation chemistry data from the first years of sampling in the BAPMoN provide a reasonable and acceptable general picture of

the distribution of sulphates and nitrates over Europe and North America. It is also possible to conclude that sulphates in precipitation over Europe have increased since the fifties by 50–100 per cent. The smaller rate of increase over North America may be due, at least in part, to the effect of emission control measures introduced during the seventies.

It is hoped that a more detailed analysis of the data, including the years after 1976, will reveal that both the amount and quality of data have improved since the early days of the network. Presumably the increased emphasis on intercalibration of methods of analysis that now exists within the project will contribute to a gradual improvement in the quality of the data.

Acknowledgements

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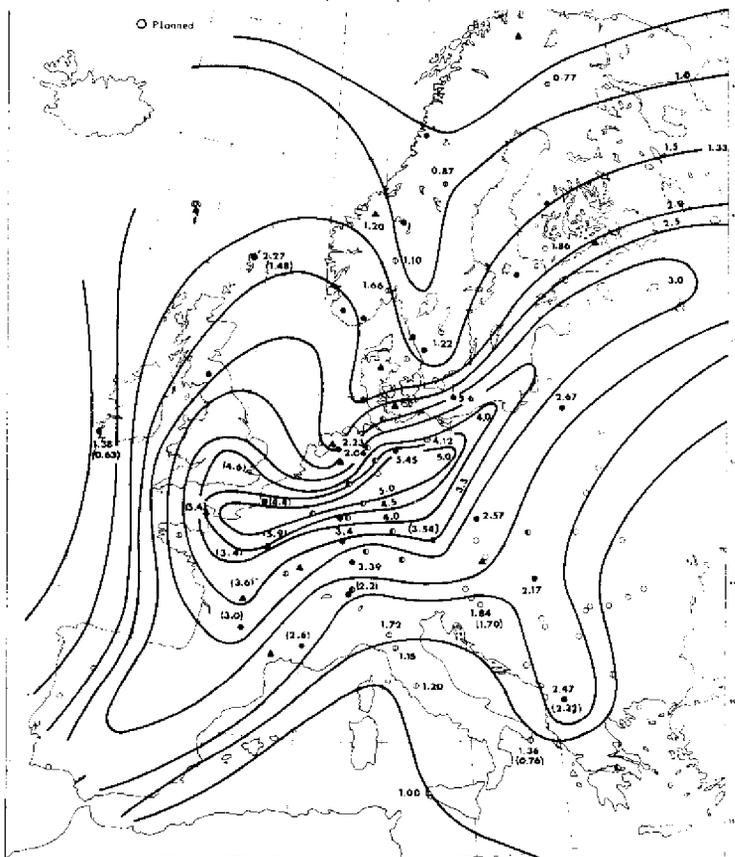


Figure 1 Annual mean concentration of sulphate in precipitation ($\text{mg SO}_4^{2-} \text{ l}^{-1}$) 1972–76 (BAPMoN data)

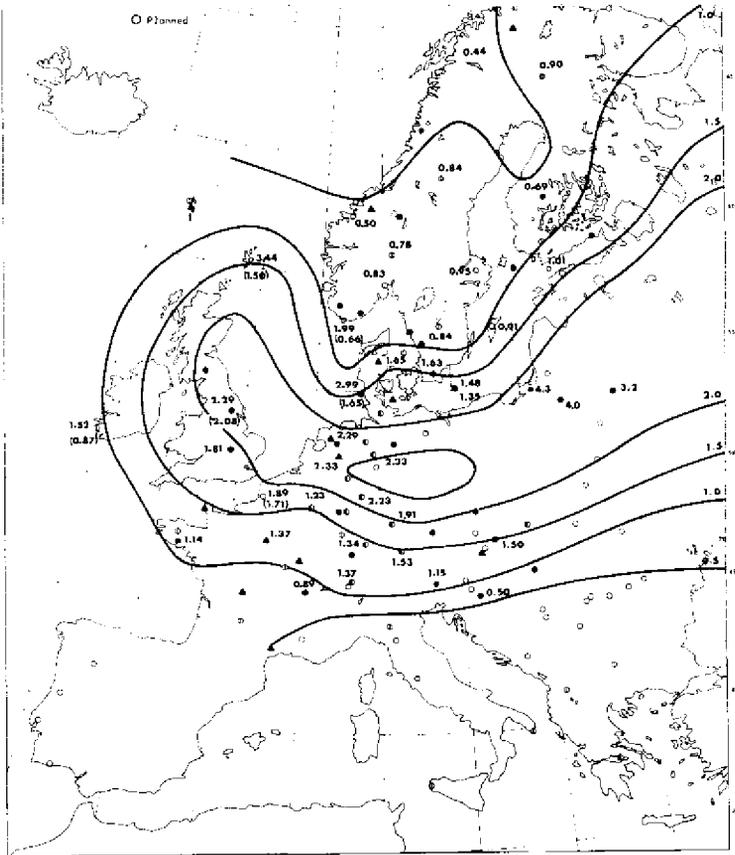


Figure 2 Annual mean concentration of sulphate in precipitation ($\text{mg SO}_4^{2-} \text{l}^{-1}$) 1954–59 (de Bary and Junge 1963)

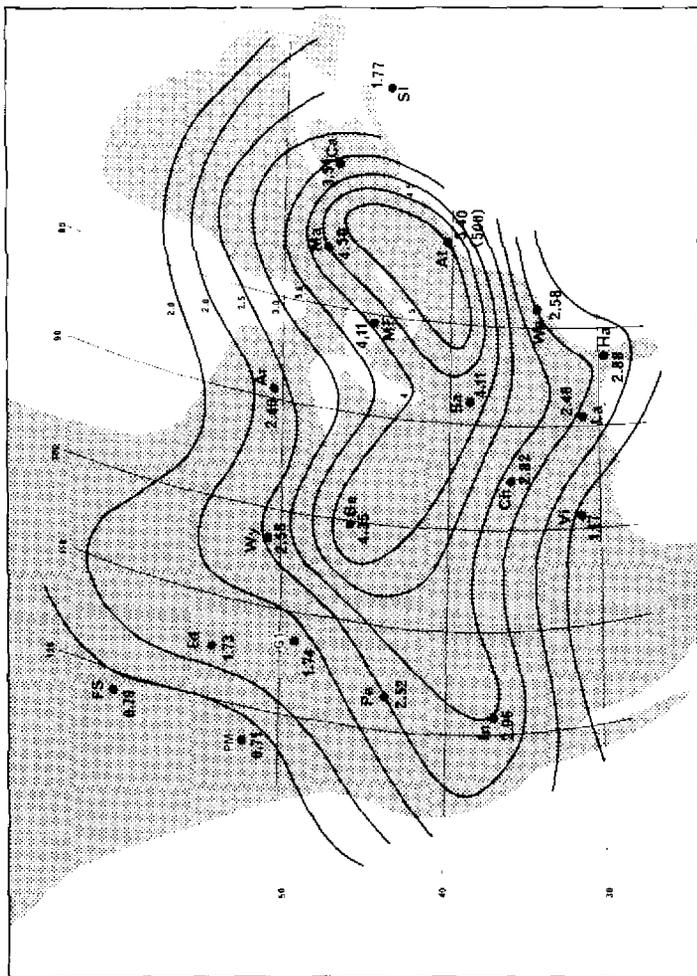


Figure 3 Mean concentration of sulphate in precipitation ($\text{mg SO}_4^{2-} \text{l}^{-1}$) 1972-76 (BAPMoN data)

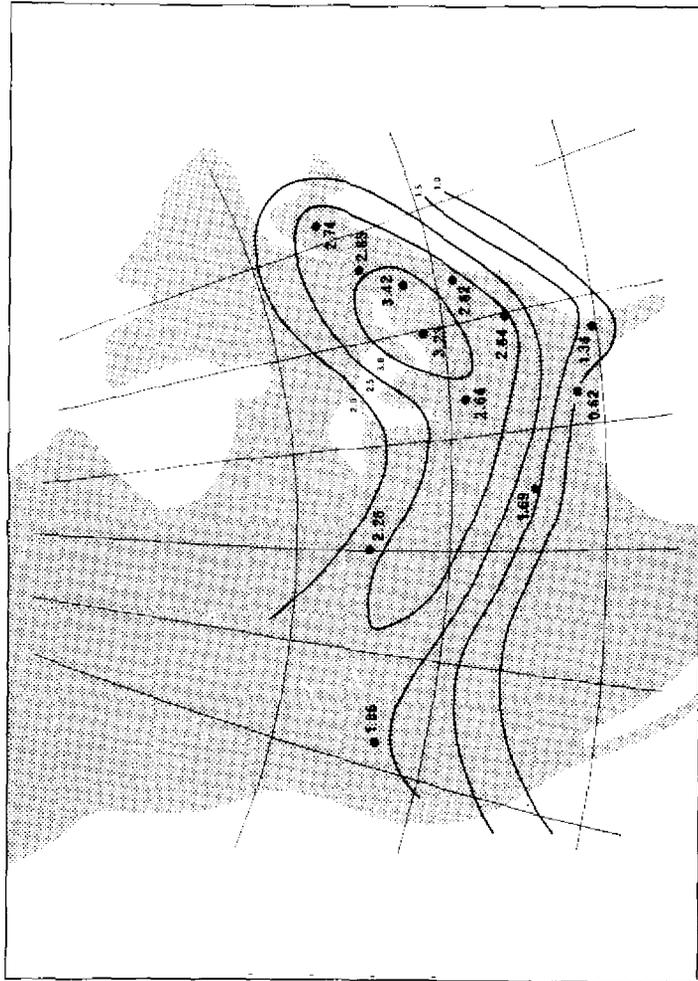


Figure 4 Excess of sulphate in precipitation (mg SO₄²⁻ l⁻¹) 1955-56 (according to Junge 1960)

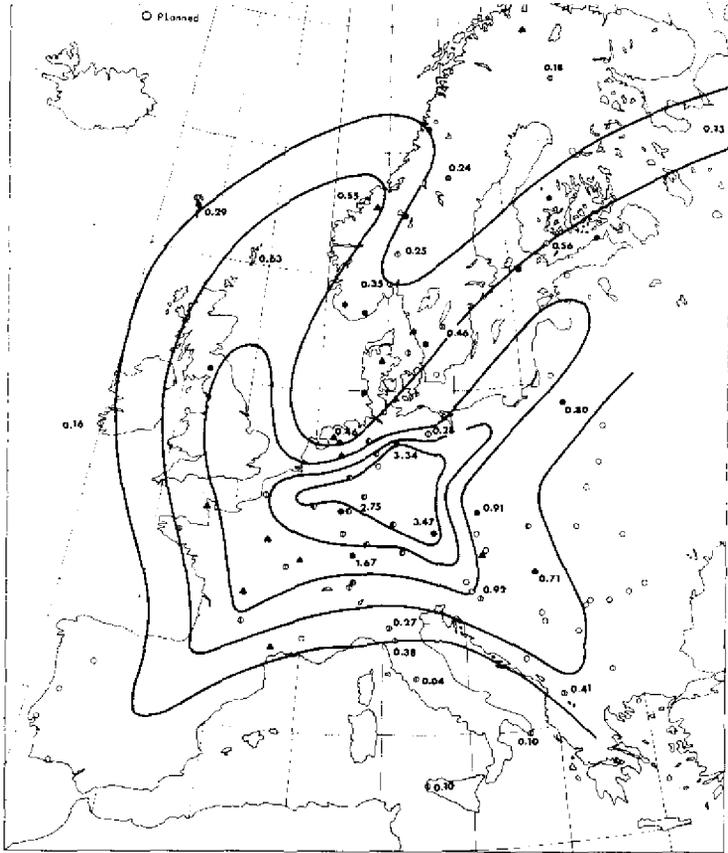


Figure 5 Mean concentration of nitrate in precipitation (mg NO₃ l⁻¹) 1972-76 (BAPMoN data)

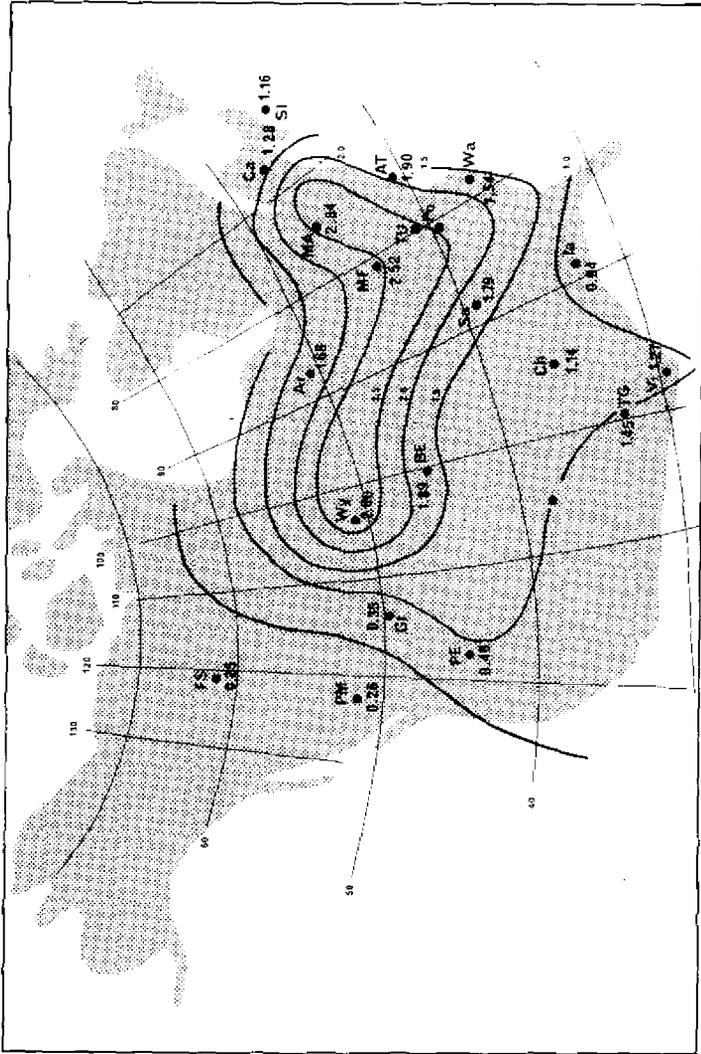


Table 2 Annual means of sulphate in precipitation [$\text{mg SO}_4^{2-} \text{ l}^{-1}$] in 1972-76

	Europe					Mean
	1972	1973	1974	1975	1976	
Souankylä, Finland		1.045	0.738	0.641	0.646	0.768
Jokioinen, Finland				2.186	1.527	1.856
Bredkälen, Sweden	0.688	0.805	0.882	1.959	—	0.869
Velen, Sweden	1.265	1.329	0.992	1.329	—	1.229
Kise, Norway				0.910	1.280	1.095
Ås, Norway				1.620	1.700	1.660
Birkenes, Norway				1.300	1.300	1.200
Faroe Islands, Denmark				(4.171)	(5.486)	(4.828)
" (reduced for sea spray)				(3.286)	4.524	(3.905)
Lerwick, U.K.	2.547	0.731	1.176	2.936	3.936	(2.271)
" (reduced for sea spray)	(1.562)			1.328	1.898	1.475
Valentia, Ireland	1.334	1.120	1.778	1.415	1.274	(1.884)
" (reduced for sea spray)	0.546	0.512	0.619	0.877	0.605	0.632
Witteveen, The Netherlands	2.414	1.904	2.296	2.284	—	(2.229)
" (reduced for sea spray)	2.172	1.616	2.103	2.153	(13.894)	2.011
Neulobow, G.D.R.				(10.851)	2.674	(12.373)
Suwaki, Poland				5.921	2.674	2.674
Langenlugg, F.R.G.		4.538	2.054	4.021	—	5.462
Brojackriegel, F.R.G.		2.354	3.229	1.580	—	3.544
Schaunsland, F.R.G.	2.434			1.873	—	2.399
Svatouch, Czechoslovakia				1.873	3.261	2.967
Kecskemet, Hungary				2.370	1.780	2.166
Puntjarika, Yugoslavia	1.200	2.525	1.990	2.370	2.085	(1.835)
" (reduced for sea spray)	1.038	1.924	2.094	1.870	2.061	1.704
Lazaropole, Yugoslavia				1.669	2.472	(2.472)
" (reduced for sea spray)				1.680	2.320	2.320
Monte Cimone, Italy				1.129	1.179	1.154
Parma, Italy				1.904	1.546	1.725
St. Maria di Leuca, Italy				1.590	1.138	(1.364)
" (reduced for sea spray)				0.953	0.573	0.763
Trapani, Italy				1.429	0.988	0.988
Viterbo, Italy				1.429	0.979	1.204
Siktivkar, U.S.S.R.	1.830	0.800	1.898	1.333	—	1.415

Values have been put in parenthesis when they seem to be unreasonable.

Mean: 1.76 mg l^{-1}

σ : 1.34

σ : 0.27

Table 3 Annual means of sulphate in precipitation ($\text{mg SO}_4 \text{ l}^{-1}$) 1972-76

	North America						Mean
	1972	1973	1974	1975	1976	1976	
Hawaii, Hawaii	—	—	1.086	2.376	1.683	1.710	
Pago Pago, American Samoa	—	—	—	1.329	1.377	1.350	
Glacier National Park, Montana	—	2.784	1.827	1.083	1.269	1.740	
Inyo, California	—	—	2.540	3.690	2.710	2.966	
Pendleton, Oregon	—	—	2.916	2.292	2.343	2.520	
Cherokee, Oklahoma	—	1.824	1.953	2.133	5.340	2.820	
Beeble, South Dakota	—	—	3.125	3.823	5.400	4.332	
Lauderdale, Mississippi	—	1.938	3.513	1.866	2.574	2.460	
Victoria, Texas	1.047	—	—	1.020	1.359	1.170	
Salem, Illinois	3.045	3.849	4.296	3.678	5.670	4.110	
Hardee, Florida	2.448	3.300	—	—	—	(2.880)	
Wake, North Carolina	0.509	2.781	2.904	3.060	2.634	2.580	
Macon, North Carolina	—	—	—	—	2.751	2.751	
Atlantic City, New Jersey	—	5.091	5.412	4.620	6.453	(5.400)	
" (reduced for sea spray)	—	4.741	5.190	4.158	6.030	5.029	
Caribou, Maine	—	3.828	3.078	4.038	3.069	3.510	
Mount Forest, Ontario	—	3.771	4.068	4.011	4.602	4.110	
Wynyard, Saskatchewan	—	—	3.156	1.500	3.036	2.580	
Maniwaki, Quebec	—	—	—	5.579	3.528	4.500	
Armstrong, Ontario	—	—	1.995	2.085	3.084	2.400	
Edson, Alberta	—	—	0.960	2.185	2.050	1.732	
Fort Simpson, N.W.T.	—	—	0.816	—	0.762	0.790	
Puntzi Mountain, B.C.	—	—	0.786	0.942	0.690	0.710	
Sable Island, Nova Scotia	—	—	—	3.267	5.703	(4.500)	
" (reduced for sea spray)	—	—	—	1.533	1.998	1.770	

Values have been put in parenthesis when they seem to be unreasonable.

Mean: 2.62 mg l^{-1}

σ : ± 1.62

σ' : ± 0.35

Table 4 Annual means of nitrates in precipitation ($\text{mg NO}_3 \text{ l}^{-1}$) 1972-76

	Europe					Mean
	1972	1973	1974	1975	1976	
Sodankylä, Finland	—	0.189	0.067	0.275	0.181	0.178
Jokioinen, Finland	—	—	—	0.647	0.468	0.560
Bredkälen, Sweden	—	0.150	0.207	0.348	—	0.237
Velen, Sweden	—	0.761	0.383	0.534	—	0.460
Kise, Norway	—	—	—	0.100	0.390	0.250
Ås, Norway	—	—	—	0.130	0.570	0.350
Birkenes, Norway	—	—	—	0.470	0.630	0.550
Faroe Islands, Denmark	—	—	0.720	0.134	0.304	0.286
Lerwick, U.K.	0.829	0.276	0.595	0.829	(9.276)	0.625
Valentia, Ireland	0.128	0.098	0.273	0.206	0.122	0.159
Witteveen, The Netherlands	0.427	0.423	0.441	0.554	—	0.461
Suwalki, Poland	—	—	—	—	0.803	0.803
Neuglobsow, G.D.R.	—	—	—	0.219	0.336	0.277
Langenbrugge, F.R.G.	—	—	2.254	3.168	4.592	3.338
Brotjacklriegel, F.R.G.	—	—	—	—	3.474	3.474
Schauinsland, F.R.G.	—	1.050	1.638	2.677	1.313	1.669
Svratouch, Czechoslovakia	—	—	1.193	11.231	0.794	0.906
Kecskemet, Hungary	—	—	0.350	1.163	0.622	0.712
Puntjarika, Yugoslavia	0.354	0.882	1.109	1.031	1.241	0.923
Lazaropole, Yugoslavia	—	—	—	—	0.407	0.407
Monte Cimone, Italy	—	—	—	0.276	0.477	0.376
Parma, Italy	—	—	—	0.126	0.405	0.265
St. Maria di Leuca, Italy	—	—	—	0.154	0.047	0.101
Trapani, Italy	—	—	—	—	0.099	0.099
Viterbo, Italy	—	—	—	0.046	0.040	0.043

Values have been put in parenthesis when they seem to be unreasonable.

Mean 0.66
± 0.87

Since, however, data for the North American continent are given as sulphate and most data for Europe are given as sulphur, no direct comparison between the values from the two continents can be made. In order to make such a comparison the North American data should be converted into sulphur by multiplying by 0.33. It then has to be concluded that the sulphur content of precipitation in areas of maximum concentration in Europe is two to three times as high as in areas of maximum concentration in North America. The reason for this difference could be either that in Central Europe some countries have still reported in sulphate or that the regional background air pollution stations in Europe are located closer to local sources and are thus more influenced by them than is the case on the North American continent. This latter hypothesis will, however, need to be confirmed by a special study.

The nitrate and nitrogen situation (Figures 5 and 6 in the paper) is parallel to that of sulphate and sulphur. This implies that the values in Figure 5 (North America) are in nitrate and those in Figure 6 in nitrogen. Consequently, in order to compare the data in those two figures the data from North America (Figure 6) should be multiplied by 0.23 to convert them to nitrogen. The difference between lower concentration in the USA (low) and Europe (high) is even more accentuated than is the case for sulphur.

In summary the following corrigenda should be made to the figures and tables in MARC Report No. 22:

Figure 1: change title to read: Annual mean concentration of sulphur in precipitation ($\text{mg S } \ell^{-1}$) 1972-76. (BAPMoN data)

Figure 2: change title to read: Annual mean concentration of sulphur in precipitation ($\text{mg S } \ell^{-1}$) 1954-59. (Acc. de Bary and Junge 1963)

Figure 5: change title to read: Annual mean concentration of nitrogen in precipitation ($\text{mg N } \ell^{-1}$) 1972-76. (BAPMoN data)

Table 1: change title to read: Monthly means (1972-76) of sulphur in precipitation ($\text{mg S } \ell^{-1}$) for European stations and of sulphate in precipitation ($\text{mg SO}_4 = \ell^{-1}$) for North American stations.

Table 2: change title to read: Annual means of sulphur in precipitation ($\text{mg S } \ell^{-1}$) 1972-76.

Table 4: change title to read: Annual means of nitrogen in precipitation ($\text{mg N } \ell^{-1}$) 1972-76.

The discussion of the matter of units presented on pages 2 and 3 of the MARC Report No. 22 is now superseded by the above note.

Corrigendum to "A preliminary evaluation of WMO-UNEP precipitation chemistry data".

MARC Technical Report No. 22 by C. C. Wallén

Data from the UNEP/WMO BAPMoN network for the period 1972-76 were analysed in this paper. A serious difficulty, however, has been to clarify the extent to which the published values for sulphate and nitrate content of precipitation have been given as either sulphate and nitrate or as derived values of sulphur and nitrogen respectively (see discussion on pages 2 and 3 of the paper).

The BAPMoN manual requests WMO members to report in values converted to sulphur and nitrogen as of 1974. In spite of this, at the time the paper was published it was assumed that the majority of North American and European data for the period 1972-76 had been reported as sulphate and nitrate. In fact, it subsequently became clear that data from the United States of America for 1972 to 1974 had been reported as sulphate and in 1975 and 1976 as sulphur. Canadian data were first reported as sulphate but later the data for the whole period 1972-76 were converted to sulphur and reissued in the converted form.

Thus North American data were presented in the early publications as a mixture of sulphate and sulphur values. In order, therefore, to obtain a homogeneous data set it was necessary that they be expressed in the same units. In order to make possible a comparison with the data published for the USA by Junge (1960) which were given as sulphate, the decision was taken to present all North American data as sulphate (see Tables 1 and 3; Figures 3 and 4). No changes, therefore, are necessary to the conclusions drawn from the comparison between the North American situation in the 1950s and that of the 1970s.

The discussion in the paper of the European data, however, does need amendment. Initial comparison of the magnitudes of the European and North American data led to the conclusion that most of the European data had also been reported as sulphate. Subsequent inquiries made by WMO to European Member States after the MARC report had been published made it evident that most of the European data since 1972 had been reported as sulphur. One or two countries, however, did not make the switch from reporting in sulphate to sulphur until 1977.

Most of the European data given in Figure 1 and Tables 1 and 2 of the paper have, therefore, been reported as sulphur. Fortunately, it is now known that the European data taken from de Bary and Junge (1963) and presented in Figure 2 are also given as sulphur. Hence conclusions in the paper regarding changes over the European continent between the 1950s to the 1970s are still valid.

Table 5 Annual means of nitrates in precipitation ($\text{mg NO}_3 \text{e}^{-1}$) 1972-76

	North America					Mean
	1972	1973	1974	1975	1976	
Hawaii, Hawaii	—	—	0.560	—	—	(0.560)
Pago Pago, American Samoa	—	—	0.164	—	—	(0.164)
Glacier National Park, Montana	0.154	0.076	0.282	0.440	0.666	0.324
Pendleton, Oregon	0.376	0.290	0.967	1.188	1.712	0.907
Beeble, South Dakota	0.882	2.159	—	2.642	—	1.894
Cherokee, Oklahoma	0.841	—	0.842	1.181	1.698	1.141
Tom Green, Texas	—	1.176	—	1.357	—	1.267
Victoria, Texas	—	—	—	—	1.052	1.052
Salem, Illinois	1.458	1.198	1.996	1.820	2.480	1.790
Lauderdale, Mississippi	0.796	0.792	0.949	0.792	1.238	0.913
Hardee, Florida	3.900	—	—	—	—	(3.900)
Wake, North Carolina	—	0.590	1.477	1.865	1.418	1.340
Atlantic City, New Jersey	0.832	1.449	2.054	1.936	3.220	1.906
Caribou, Maine	—	—	1.277	1.435	1.132	1.281
Mount Forest, Ontario	—	1.065	2.780	2.040	4.081	2.418
Maniwaki, Quebec	—	—	—	2.340	3.340	2.840
Wynyard, Saskatchewan	—	—	2.585	2.156	3.557	2.766
Armstrong, Ontario	—	—	2.450	0.742	1.353	1.682
Puntzi Mountain, B.C.	—	—	0.093	0.293	0.355	0.249
Fort Simpson, N.W.T.	—	—	0.222	—	—	0.222
Sable Island, Nova Scotia	—	—	—	0.587	1.749	1.163

Values have been put in parenthesis when they seem to be unreasonable.

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- 21 *Lead in the soil environment* by D. H. Khan (in press)
- 22 *A preliminary evaluation of WHO-UNEP precipitation chemistry data* by C. C. Wallén

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