Distribution of sulfur and chlorine over Europe

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ABSTRACT

Maps showing the mean concentration of sulfar and chlorine in air and precipitation over north western Europe in summer and winter have been constructed. Exess $SO_4 - S$ has been computed on the basis of the assumption that chloride is a conservative property of sea spray particles and that there are no other sources for chloride than sea spray. Finally, maps showing the ratio of concentration air/precipitation for sulfur and chlorine are presented.

In air pollution studies it is of certain interest to know the background concentration of some constituents in a larger area. For northern and western Europe such information can be obtained by an evaluation of the data from the Europan chemical network, published in this Journal over the past years (see e.g. EGNER and ERIKSSON, 1955). In the course of such evaluation several maps were prepared which may be of more general interest for chemical climatology and which are therefore presented here together with a brief discussion.

The maps reproduced here are concerned with the average *concentration* of sulfur and chlorine in air and precipitation, separate for summer and winter. The number of monthly average data available for the various stations differs widely since the network was extended over the years from Scandinavia to west and central Europe. The data used in the maps are given in Tables 1 and 2 and the stations are plotted in Fig. 1. For chlorine in precipitation the data could be supplemented by those from the IGY network in Czechoslovakia (MACKU *et al.*, 1959). The designation of the stations is the same as in the original papers to which we refer for more details.

For calculation of the excess sulfur in precipitation it was necessary that the monthly average values used refer to the same period for both sulfur and chlorine. For this reason we omitted some values for which the data were not complete.

For easier comparison we drew all isolines in the maps in the same almost logarithmic scale.

Isolines: 0.8, 1.3, 2.0, 3.0, 5.0, 8.0, 13;
Ratio of subsequent
isolines: 1.62, 1.54, 1.50, 1.67, 1.60, 1.62.

For the present purpose "summer" is composed of the months May to October and "winter" of the months November to April. In a few cases the figures in the map had to be disregarded for drawing the isolines; these figures are underlined.

We will discuss briefly the various maps and try to explain some of the salient features.

1. Sulfur in air (Figs. 2 and 3)

Sulfur in air is composed of the gases SO_2 and H_2S and of sulfate in aerosol particles. For most areas the sulfate-sulfur amounts to about 10 % of the gaseous sulfur, so that for all practical purposes the maps represent the sulfur in gaseous form. It is not known to what extent H_2S is included in the data given in the network.

The highest concentrations of S in air are observed over the highly populated and industrialized areas of Great Britain and the western and central continent indicating that anthropogenic sources of SO_2 are predominant for major parts of Europe. The concentrations during winter are about twice as high as during the summer as result of both the higher consumption of sulfur containing fuels and the higher frequency of stable inversion layers over land in the cold season. During inversion periods the SO_2 emitted is accumulated for many days in shallow layers near the ground,

TABLE 1. *Summer average concentration of sulfur and chlorine in air and precipitation computed from the data published in Tellus 1954–1959.*

Station	Air				Precipitation					Ratio air/Precipitation	
	SO_2-S $\mu g/m^3$	Number of months	Cl_2 $\mu g/m^3$	Number of months	$SO_4^{=}-S$ mg/l Total	$SO_4^{=}-S$ mg/l Excess	Cl^- mg/l	Amount of precipitation mm	Number of months	$\frac{SO_2-S}{SO_4^{=}-S}$ $\frac{\mu g/m^3}{mg/l}$	$\frac{Cl_2}{Cl^-}$ $\frac{\mu g/m^3}{mg/l}$
Ri	1.8	11	5.9	11	0.38	0.33	1.05	59.2	24	4.7	5.6
Ki	1.7	21	3.2	19	0.45	0.44	0.34	46.5	26	3.8	9.4
Ar	2.0	20	1.5	19	0.47	0.46	0.36	44.5	25	4.3	4.2
Öj	1.7	25	1.9	25	0.72	0.70	0.52	46.3	26	2.4	3.6
Rö	2.7	23	2.9	23	0.71	0.69	0.35	52.9	25	3.8	8.3
Öf	2.3	23	2.8	25	0.77	0.75	0.39	51.0	26	3.0	7.2
Br	2.5	18	1.9	20	0.55	0.54	0.19	52.0	21	4.5	10.0
ÅF	3.3	17	2.0	16	0.42	0.40	0.43	69.4	26	7.8	4.6
ÅH	2.2	5	2.2	5	0.50	0.48	0.39	56.3	16	4.4	5.6
Fö	—	—	—	—	0.76	0.74	0.33	50.9	10	—	—
Sv	—	—	—	—	0.53	0.52	0.23	65.6	22	—	—
Rä	3.1	23	2.8	24	0.60	0.58	0.34	48.7	25	5.2	8.2
Åm	2.5	22	3.0	22	0.60	0.58	0.26	63.5	25	4.2	11.5
Sa	3.9	26	2.6	26	0.70	0.69	0.31	62.5	26	5.6	8.4
Ul	2.9	25	1.7	25	0.76	0.74	0.41	55.7	26	3.8	4.1
Er	3.3	25	2.1	25	0.79	0.76	0.52	56.4	25	4.2	4.0
St	3.3	26	2.0	26	0.87	0.85	0.43	53.4	25	3.8	4.7
Fo	2.8	20	2.9	20	0.65	0.63	0.38	70.2	26	4.0	7.6
Kv 7	6.5	19	3.0	18	1.17	1.14	0.50	57.4	26	5.6	6.0
VK	6.2	6	3.1	6	1.04	1.01	0.55	53.8	21	6.0	5.6
La	3.1	16	4.7	16	1.01	0.94	1.41	52.6	25	3.1	3.3
Bo	2.8	16	10.8	16	0.95	0.78	3.73	63.3	25	2.9	2.9
Vi	5.9	16	114.0	15	2.46	1.39	23.03	50.8	21	2.4	4.9
Fa	2.5	17	6.6	17	0.90	0.87	0.59	51.0	26	2.8	11.2
Fl	2.9	24	5.2	24	0.86	0.83	0.79	65.2	26	3.4	6.6
Am	—	—	—	—	0.81	0.74	1.45	81.5	26	—	—
Fi	—	—	—	—	0.72	0.71	1.68	91.2	6	—	—
Si	—	—	—	—	0.96	0.95	2.33	96.6	18	—	—
Pl	3.3	23	4.5	23	1.08	0 94	3 01	62.4	26	3.1	1.5
Sö	—	—	—	—	1.20	1.16	0.82	61.3	23	—	—
Sm	2.3	26	2.3	26	1.00	0.97	0.49	49.1	25	2.3	4.7
Sy	2.2	16	6.1	15	0.83	0.80	0.79	51.0	25	2.7	7.7
BH	1.9	17	1.6	17	0.98	0.92	1.26	45.9	24	1.9	1.3
Sk	4.3	13	2.1	13	1.19	1.11	1.73	49.8	25	3.5	1.2
Al	7.4	18	3.4	18	1.37	1.30	1.59	52.4	25	5.4	2.1
Hi	—	—	—	—	1.12	1.05	1.69	43.2	26	—	—
Ta	1.7	9	2.4	9	0.73	0.61	2.50	32.1	11	2.3	1.0
Yt	—	—	—	—	0.38	0.27	2.25	53.6	14	—	—
Gj	—	—	—	—	0.40	0.26	2.83	86.2	14	—	—
Fn	—	—	—	—	0.64	0.63	0.35	55.2	14	—	—
Få	—	—	—	—	0.40	0.37	0.56	51.8	9	—	—
Vå	3.7	2.0	2.4	19	0.86	0.84	0.50	34.2	26	4.3	4.8
Tr	—	—	—	—	0.57	0.56	0.20	79.1	14	—	—
Ke	—	—	—	—	0.59	0.58	0.22	67.4	14	—	—
Sd	—	—	—	—	1.02	0.93	2.07	88.5	13	—	—
Da	—	—	—	—	0.57	0.56	0.31	72.3	13	—	—
Ås	4.6	16	3.3	16	0.78	0.75	0.69	68.8	26	5.9	4.8
Li	5.4	26	43.9	26	1.56	0.85	15.29	94.6	25	3.5	2.9
So	1.6	13	1.4	13	0.66	0.64	0.60	45.0	13	2.4	2.3
Ka	6.3	24	4.2	24	0.93	0.89	0.84	48 7	25	6 8	5.0
Ku	3.9	21	4.9	20	1.27	1.25	0.50	52.1	25	3.1	9.8
Jy	4.1	24	1.9	23	0.64	0.57	1.32	57.0	25	6.4	1.4
Pu	8.6	13	1.2	13	1.69	1.66	0.72	58.6	13	5.1	1.7

Table 1 (*continued*)

Station	Air				Precipitation					Ratio air/Precipitation	
					$SO_4^{=}$ – S mg/l						
	SO_2 – S $\mu g/m^3$	Number of months	Cl_2 $\mu g/m^3$	Number of months	Total	Excess	Cl^- mg/l	Amount of precipitation mm	Number of months	$\frac{SO_2 - S}{SO_4^{=} - S}$ $\frac{\mu g/m^3}{mg/l}$	$\frac{Cl_2}{Cl^-}$ $\frac{\mu g/m^3}{mg/l}$
Tv	5.8	22	6.6	21	0.93	0.84	2.01	46.0	25	6.2	3.3
Rj	2.2	7	3.7	7	0.33	0.12	4.50	56.1	8	6.7	0.8
Vn	—	—	—	—	1.36	1.20	3.44	64.7	14	—	—
Gr	—	—	—	—	1.81	1.68	2.80	54.9	14	—	—
Öd	4.4	19	6.4	20	1.52	1.41	2.32	56.2	26	2.9	2.8
Bs	—	—	—	—	1.48	1.27	4.52	69.1	14	—	—
Ly	—	—	—	—	1.80	1.72	1.68	60.3	14	—	—
As	5.3	18	5.8	18	1.36	1.19	3.64	65.3	26	3.9	1.6
Vd	—	—	—	—	1.68	1.58	2.16	68.7	12	—	—
Bl	—	—	—	—	1.81	1.73	1.69	56.2	13	—	—
Ty	4.8	18	10.8	19	1.56	1.42	3.02	51.1	26	3.1	3.6
Hö	—	—	—	—	1.70	1.34	7.79	70.4	14	—	—
Ad	—	—	—	—	1.52	1.43	1.89	51.9	14	—	—
Lw	1.4	8	5.4	8	2.30	1.48	17.73	50.2	8	0.6	0.3
Sw	2.4	8	5.4	8	1.98	1.30	14.57	45.6	8	1.2	0.4
Ab	8.4	20	8.3	20	1.64	1.49	3.23	64.8	2	5.1	2.6
Ed	7.5	24	5.6	24	1.31	1.23	1.72	61.7	25	5.7	3.2
Es	4.1	9	3.6	9	1.39	1.29	2.02	140.2	9	2.9	1.8
Ag	10.9	7	5.6	7	1.64	1.54	2.05	52.7	8	6.6	2.7
Le	22.6	22	10.5	22	2.17	2.05	2.53	54.8	25	10.4	4.1
Ro	17.2	25	8.6	24	1.81	1.79	1.57	61.0	25	9.5	5.5
NA	6.5	24	4.7	24	1.42	1.26	3.46	73.3	26	4.6	1.4
Ca	4.9	8	5.2	8	1.72	1.30	9.24	60.5	8	2.8	0.6
Va	—	—	—	—	1.24	0.82	9.17	111.5	11	—	—
We	8.9	8	45.6	8	3.77	1.81	42.36	66.1	8	2.4	1.1
Sc	11.9	18	14.9	18	1.93	1.72	3.98	77.8	20	6.2	3.7
Bv	17.1	17	6.9	17	1.98	1.91	1.36	67.8	18	8.6	5.1
Bn	23.5	18	27.0	18	2.24	2.16	1.70	73.6	18	10.5	15.9
Au	16.0	22	11.8	22	1.64	1.61	0.80	72.0	23	9.7	14.8
Fe	8.2	22	4.0	22	2.32	2.30	0.43	85.7	25	3.5	9.3
Ba	9.0	25	5.7	26	0.89	0.87	0.39	101.9	26	10.1	14.6
Ho	15.6	17	10.8	17	1.28	1.26	0.53	101.8	17	12.2	20.4
Rm	8.0	12	4.9	12	1.42	1.39	0.81	77.8	12	5.6	6.1
Et	8.6	12	5.4	12	1.26	1.24	0.39	80.3	12	6.8	13.8
He	—	—	—	—	1.71	1.64	1.48	81.5	10	—	—
Rz	8.5	13	3.4	13	1.29	1.28	0.18	51.6	13	6.6	18.9
Wi	11.1	13	3.5	13	1.54	1.52	0.35	65.1	13	7.2	10.0
Lz	10.0	14	5.0	14	1.34	1.33	0.44	83.9	14	7.5	11.4
Kl	9.8	12	4.5	12	1.09	1.08	0.22	111.2	12	9.0	20.5
BL	14.3	14	9.5	14	1.06	1.03	0.72	116.7	14	13.5	13.2
Vl	—	—	11.6	2	1.59	1.20	8.38	73.0	1	—	1.4
DH	12.9	10	33.0	10	2.18	1.52	14.27	65.5	11	5.9	2.3
DB	12.1	20	11.8	20	1.86	1.74	2.79	72.7	20	6.5	4.2
W	10.4	11	15.1	12	2.42	1.48	2.46	83.9	12	4.3	6.1
SA	9.0	20	8.0	20	1.50	1.37	2.75	70.7	20	6.0	2.9
U	19.4	20	8.1	20	2.07	1.98	1.83	61.7	19	9.4	4.4
B	8.3	19	5.7	19	1.41	1.37	0.89	117.4	20	5.9	6.4
D	7.2	20	7.5	20	1.23	1.18	1.17	69.9	20	5.9	6.4
MH	11.1	11	6.3	10	1.21	1.67	0.94	51.8	10	9.1	6.7
Rs	5.9	10	9.8	10	0.66	0.51	3.34	64.8	10	8.9	2.9
LM	10.8	7	15.5	7	1.00	0.94	1.22	49.1	10	10.8	12.7
Lx	6.6	9	8.8	9	0.74	0.72	0.49	78.3	10	8.9	18.0
Bg	15.6	10	6.2	11	0.61	0.58	0.69	77.7	10	9.2	9.0
Ae	12.7	4	13.0	4	0.81	0.78	0.50	93.2	10	15.6	26.0

Table 1 (*continued*)

Station	Air				Precipitation					Ratio air/Precipitation	
	SO_2-S	Number of	Cl_2	Number of	$SO_4^{=}-S$ mg/l		Cl^-	Amount of precipitation	Number of	$\frac{SO_2-S}{SO_4^{=}-S}$	$\frac{Cl_2}{Cl^-}$
	$\mu g/m^3$	months	$\mu g/m^3$	months	Total	Excess	mg/l	mm	months	$\frac{\mu g/m^3}{mg/l}$	$\frac{\mu g/m^3}{mg/l}$
Za	1.8	6	6.6	6	0.50	0.47	0.67	94.9	10	3.6	9.9
DA	—	—	—	—	1.32	1.23	1.97	124.4	8	—	—
Bt	—	—	—	—	1.44	0.86	12.5	107.0	8	—	—
Bi	—	—	—	—	0.91	0.81	2.21	83.1	7	—	—
Rl	—	—	—	—	1.18	0.87	6.67	108.5	8	—	—
Ma	—	—	—	—	1.32	0.72	12.96	116.6	7	—	—
Fj	—	—	—	—	1.56	1.32	5.33	71.4	5	—	—
ÖT	—	—	—	—	1.00	0.95	1.12	16.0	2	—	—
Cl	—	—	—	—	1.00	0.94	1.32	109.4	6	—	—

due to low rates of horizontal and vertical mixing. The maps suggest that the influence of the west and central European source areas apparently spreads as far as central Scandinavia and to some degree over the ocean around the British Isles. The stations in Iceland and the northern part of Scandinavia indicate a background level over the North Atlantic of about 1.5–2.0 in summer and 3–4 in winter, with a seasonal variation very similar to that of the polluted areas.

MEETHAM (1959) published a map on the SO_2 concentration over England for winter which shows more details than our map for this area

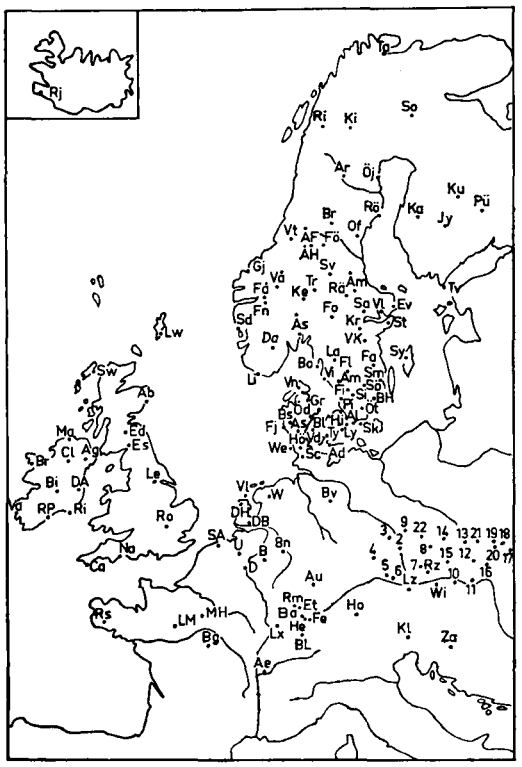

FIG. 1. Map of stations.

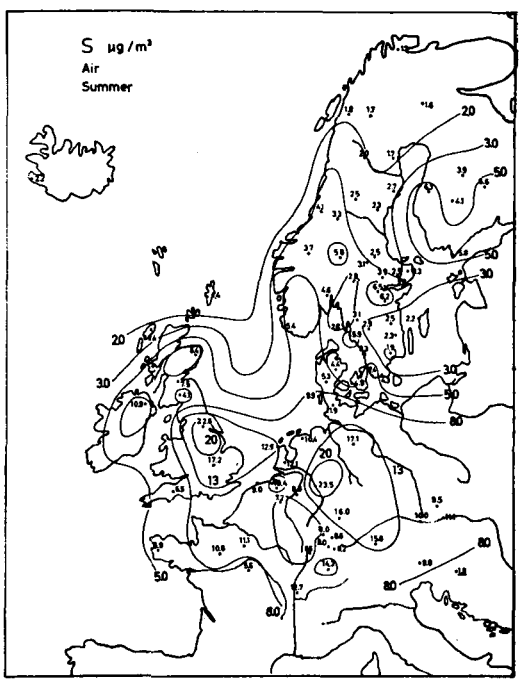

FIG. 2. Average concentration of SO_2-S in air for summer.

TABLE 2. *Winter average concentration of sulfur and chlorine in air and precipitation computed from the data published in Tellus 1954–59.*

Station	Air				Precipitation					Ratio air/Precipitation	
	SO_2-S $\mu g/m^3$	Number of months	Cl_2 $\mu g/m^3$	Number of months	$SO_4^=-S$ mg/l Total	$SO_4^=-S$ mg/l Excess	Cl^- mg/l	Amount of precipitation mm	Number of months	$\frac{SO_2-S}{SO_4^=-S}$ $\mu g/m^3$ / mg/l	$\frac{Cl_2}{Cl^-}$ $\mu g/m^3$ / mg/l
Ri	4.4	10	5.7	10	0.50	0.36	2.85	33.3	29	2.8	2.0
Ki	2.2	19	3.0	16	0.47	0.45	0.54	22.2	28	4.7	5.6
Ar	3.0	20	3.4	20	0.81	0.77	0.91	19.0	29	3.7	3.7
Öj	4.2	19	2.2	19	0.99	0.95	0.82	23.8	30	4.2	2.7
Rö	8.2	22	2.9	22	0.93	0.90	0.56	25.2	28	8.8	5.2
Of	4.8	17	5.0	22	0.97	0.88	1.92	22.6	30	4.9	2.6
Br	1.9	18	2.3	18	1.14	1.07	1.46	9.9	22	1.7	1.6
ÅF	6.2	16	4.0	16	0.58	0.52	1.28	33.5	26	10.7	3.1
ÅH	3.4	3	4.7	3	1.46	1.32	2.93	10.0	7	2.3	1.6
Fö	—	—	—	—	0.97	0.93	0.76	26.3	12	—	—
Sv	—	—	—	—	0.75	0.72	0.57	21.2	21	—	—
Rä	5.6	27	3.2	27	1.07	1.02	0.93	17.8	27	5.2	3.4
Åm	4.2	26	2.6	25	0.79	0.76	0.63	32.6	27	5.3	4.1
Sa	10.2	24	2.2	24	1.14	1.11	0.60	31.5	30	8.9	3.7
Ul	8.8	30	1.9	30	1.13	1.10	0.68	28.7	30	7.8	2.8
Er	5.2	23	1.4	23	1.21	1.16	1.04	25.1	29	4.3	1.4
St	7.2	30	2.2	30	1.21	1.18	0.69	26.5	30	6.0	3.2
Fo	3.4	25	2.4	25	0.86	0.83	0.58	34.8	28	4.0	4.1
Kv. 7	7.3	22	3.0	22	1.33	1.30	0.73	30.2	29	5.5	4.1
VK	10.0	9	3.6	9	1.05	1.00	1.01	30.8	23	9.5	3.6
La	5.9	20	5.8	20	1.28	1.17	2.04	25.6	29	4.6	2.4
Bo	5.1	17	7.0	18	1.03	1.01	5.80	42.7	28	5.0	1.2
Vi	13.2	23	82.9	23	4.15	3.89	56.80	26.3	24	3.2	14.6
Fa	4.4	21	3.8	22	0.87	0.84	0.66	41.5	28	5.1	5.8
Fl	4.5	29	4.8	29	0.82	0.76	1.42	39.4	30	5.5	3.4
Am	—	—	—	—	0.99	0.88	2.31	51.4	30	—	—
Fi	—	—	—	—	1.04	1.04	1.03	46.5	6	—	—
Si	—	—	—	—	0.96	0.87	1.90	76.7	18	—	—
Pl	8.4	30	5.0	30	1.54	1.58	4.75	38.9	30	5.5	1.1
Sö	—	—	—	—	1.42	1.33	1.92	32.2	24	—	—
Sm	6.5	29	2.7	29	1.17	1.12	1.19	28.3	29	5.6	2.3
Sy	4.1	18	9.2	18	0.99	0.93	1.37	33.9	30	4.1	6.7
BH	7.0	16	1.7	16	1.25	1.16	2.08	35.4	29	5.6	0.8
Sk	9.3	18	4.5	18	1.78	1.60	3.95	34.7	30	5.2	1.1
Al	15.6	23	6.4	23	1.90	1.74	3.59	38.1	29	8.2	1.8
Hi	—	—	—	—	1.46	1.32	3.11	26.2	26	—	—
Ta	3.4	8	6.7	8	1.25	0.64	13.14	21.5	10	2.7	0.5
Yt	—	—	—	—	0.57	0.17	8.57	49.6	18	—	—
Gj	—	—	—	—	0.60	0.18	9.10	91.7	18	—	—
Fn	—	—	—	—	0.54	0.48	1.26	59.8	17	—	—
Få	—	—	—	—	0.50	0.45	1.16	51.0	13	—	—
Vå	4.0	21	2.7	21	1.09	1.06	0.85	13.8	24	3.7	3.2
Tr	—	—	—	—	0.90	0.87	0.49	29.9	16	—	—
Ke	—	—	—	—	0.98	0.96	0.42	25.6	14	—	—
Sd	—	—	—	—	0.90	0.60	6.44	84.0	17	—	—
Da	—	—	—	—	0.51	0.46	0.97	48.0	18	—	—
Ås	8.4	25	4.8	25	0.88	0.83	1.30	44.1	28	9.6	3.7
Li	6.1	30	45.0	22	2.43	0.47	42.36	63.7	30	2.5	1.1
So	3.6	16	5.8	16	1.33	1.42	1.51	13.2	16	2.7	3.8
Ka	7.7	23	18.0	23	0.93	0.88	0.96	15.1	29	8.3	19.8
Ku	3.5	19	16.2	20	0.95	0.92	0.50	18.6	27	3.7	32.4
Jy	3.8	27	3.7	27	0.74	0.70	0.78	25.3	29	5.1	4.7
Pu	4.1	17	9.9	16	0.73	0.71	0.49	25.3	17	5.5	20.2

Table 2 (*continued*)

Station	Air				Precipitation					Ratio air/Precipitation	
					SO_4^{--} – S mg/l						
	SO_2 – S $\mu g/m^3$	Number of months	Cl_2 $\mu g/m^3$	Number of months	Total	Excess	Cl^- mg/l	Amount of precipitation mm	Number of months	$\frac{SO_2 - S}{SO_4^{--} - S}$ $\frac{\mu g/m^3}{mg/l}$	$\frac{Cl_2}{Cl^-}$ $\frac{\mu g/m^3}{mg/l}$
Tv	5.1	14	6.2	24	1.12	1.00	2.12	26.4	30	4.6	2.9
Rj	2.9	8	11.3	7	0.36	—	21.81	78.8	10	8.0	0.5
Vn	—	—	—	—	2.16	1.94	4.68	38.2	12	—	—
Gr	—	—	—	—	2.35	2.14	4.53	40.0	12	—	—
Öd	10.5	24	4.0	24	1.78	1.49	6.26	34.3	30	5.9	0.6
Bs	—	—	—	—	1.91	1.47	9.39	44.8	12	—	—
Ly	—	—	—	—	3.15	3.04	2.41	41.5	12	—	—
As	12.7	15	5.1	15	1.90	1.42	10.20	46.2	30	6.7	0.5
Vd	—	—	—	—	2.48	2.30	3.88	45.9	12	—	—
Bl	—	—	—	—	2.39	2.21	3.78	35.3	12	—	—
Ty	11.2	24	9.7	23	2.34	2.05	6.24	29.1	30	4.8	1.6
Hö	—	—	—	—	2.21	1.64	12.30	41.1	12	—	—
Ad	—	—	—	—	2.48	2.30	3.78	29.7	12	—	—
Lw	3.2	8	12.5	8	4.58	1.53	65.86	55.2	9	0.7	0.2
Sw	3.4	7	23.4	7	2.32	1.01	28.30	54.3	7	1.5	0.8
Ab	17.7	24	9.4	24	1.57	1.11	10.00	59.1	24	11.3	0.9
Ed	16.6	23	8.1	23	2.13	1.94	4.16	46.5	25	7.8	2.0
Es	9.4	12	4.9	12	1.55	1.36	4.18	100.3	12	6.1	1.2
Ag	18.3	6	8.8	5	1.96	1.66	6.40	39.4	5	9.3	1.4
Le	40.0	22	2.8	22	2.40	2.11	6.16	40.5	25	16.7	0.5
Ro	29.8	20	15.8	20	1.81	1.62	3.99	42.8	24	16.5	4.0
NA	16.2	24	11.9	24	1.74	1.38	7.81	95.6	23	9.3	1.5
Ca	6.1	12	5.4	12	3.14	1.71	31.06	86.9	12	1.9	0.20
Va	—	—	—	—	1.80	0.92	18.91	113.0	12	—	—
We	18.8	7	58.6	7	2.21	0.28	41.69	51.6	7	8.5	1.4
Sc	21.9	17	22.2	17	2.49	2.18	6.57	43.5	18	8.8	3.4
Bv	36.0	18	10.3	18	2.68	2.53	3.15	34.3	17	13.4	3.3
Bn	42.3	18	22.5	18	2.19	2.06	2.92	45.6	16	11.3	7.7
Au	30.1	24	9.1	24	2.18	2.12	1.29	50.7	22	13.8	7.1
Fe	7.0	17	4.8	17	1.37	1.34	0.74	64.7	23	5.1	6.5
Ba	20.3	23	11.5	23	1.00	0.97	0.64	68.1	24	20.3	18.0
Ho	26.4	18	8.6	18	1.77	1.73	0.71	48.1	18	14.9	12.1
Rm	7.9	11	3.9	9	1.26	1.21	1.06	40.3	12	6.3	3.7
Et	14.3	11	5.0	11	1.71	1.59	2.44	41.9	12	8.4	2.1
He	—	—	—	—	3.03	2.92	2.39	35.0	3	—	—
Rz	12.1	12	3.6	12	1.71	1.69	0.48	21.5	12	7.1	7.5
Wi	24.4	12	5.7	12	1.82	1.80	0.45	39.9	12	13.4	12.7
Lz	15.1	14	5.7	14	2.04	2.00	0.95	59.4	14	7.4	6.0
Kl	23.7	12	7.3	12	1.24	1.23	0.20	57.2	11	19.1	36.5
BL	18.4	15	11.5	15	1.81	1.74	1.61	50.7	15	10.2	7.1
Vl	—	—	12.6	2	0.75	—	83.29	45.5	2	—	0.2
DH	17.9	12	17.5	12	2.77	0.72	44.22	52.5	11	6.5	0.4
DB	25.4	19	13.7	20	2.37	2.21	3.54	47.2	17	10.7	3.9
W	16.3	11	15.6	11	2.16	2.00	3.35	55.3	11	7.5	4.7
SA	16.4	19	12.5	20	2.27	2.05	4.70	49.7	18	7.2	2.7
U	48.3	19	13.0	20	2.53	2.44	1.98	52.2	18	16.7	6.6
B	12.0	19	9.5	19	1.04	0.99	1.25	121.8	18	11.5	7.6
D	13.9	19	11.3	20	1.31	1.24	1.62	51.8	18	10.6	7.0
MH	31.9	12	20.5	12	1.53	1.44	1.87	50.4	11	20.8	11.0
Rs	7.8	10	13.2	12	1.61	0.36	5.55	95.5	11	4.8	2.4
LM	18.3	6	15.6	6	0.83	0.74	1.97	65.4	10	22.0	7.9
Lx	12.2	11	35.9	12	0.87	0.84	0.66	90.5	10	14.0	54.4
Bg	13.7	11	11.9	12	0.96	0.88	1.82	55.7	10	14.2	6.5
Ae	17.9	8	15.6	9	0.97	0.94	0.56	102.1	10	18.4	27.9

Table 2 (*continued*)

Station	Air				Precipitation					Ratio air/Precipitation	
	SO_2-S $\mu g/m^3$	Number of months	Cl_2 $\mu g/m^3$	Number of months	$SO_4^{=}-S$ mg/l Total	$SO_4^{=}-S$ mg/l Excess	Cl^- mg/l	Amount of precipitation mm	Number of months	$\frac{SO_2-S}{SO_4^{=}-S}$ $\frac{\mu g/m^3}{mg/l}$	$\frac{Cl_2}{Cl^-}$ $\frac{\mu g/m^3}{mg/l}$
Za	14.0	5	4.2	4	0.52	0.50	0.51	95.0	7	26.9	8.2
DA	—	—	—	—	2.50	2.16	7.32	56.0	9	—	—
Bt	—	—	—	—	1.96	0.76	25.89	97.1	8	—	—
Bi	—	—	—	—	1.22	1.00	4.94	66.1	7	—	—
Rl	—	—	—	—	1.98	1.29	14.86	79.8	6	—	—
Ma	—	—	—	—	2.12	0.51	34.75	115.0	6	—	—
Fj	—	—	—	—	1.72	1.15	12.26	55.2	6	—	—
ÖT	—	—	—	—	0.92	0.86	1.33	39.4	5	—	—
Cl	—	—	—	—	1.24	1.03	4.59	77.6	6	—	—

but otherwise agrees well, so far as the few stations over this part of Europe enable a comparison. It should be pointed out that the stations of the European network are located in such a manner that local pollution is excluded to a large extent and that the data can be considered representative for larger areas. Polluted areas, if included in our maps, would appear as small circles of considerable higher concentrations.

2. Chlorine in air (Figs. 4 and 5)

Chlorine in air is composed of chloride in aerosols, primarily from the sea, and of a gaseous compound, supposedly Cl_2. Most Cl

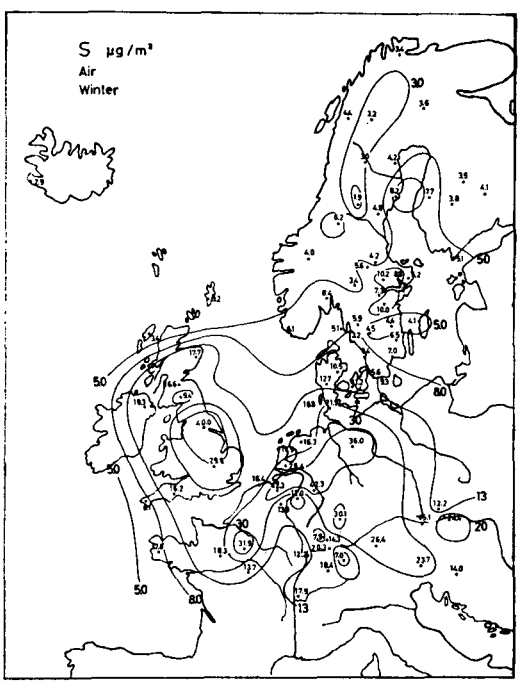

FIG. 3. Average concentration of SO_2-S in air for winter.

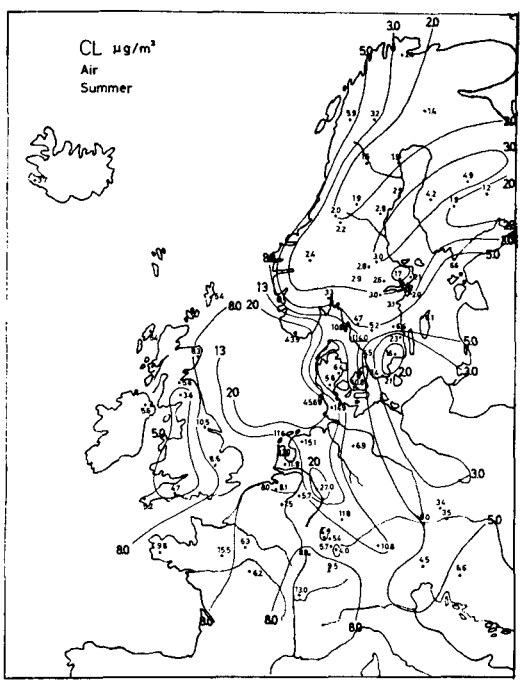

FIG. 4. Average concentration of Cl in air for summer.

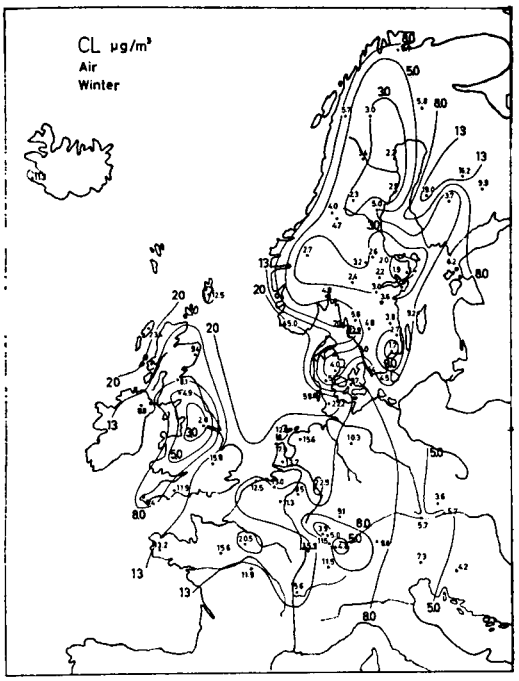

FIG. 5. Average concentration of Cl in air for winter.

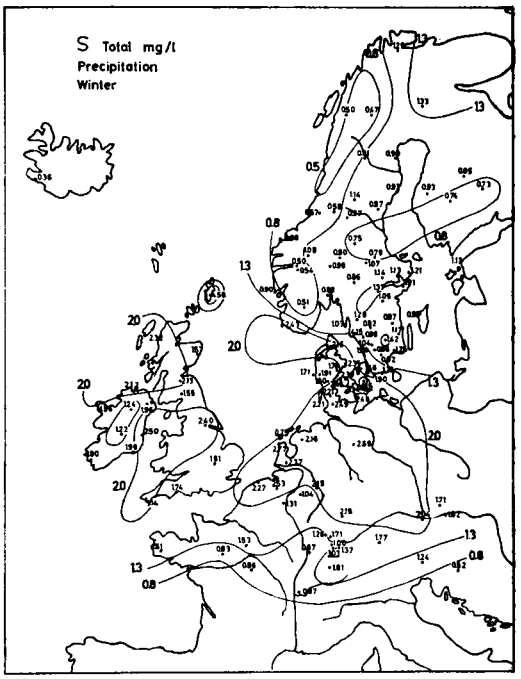

FIG. 7. Average concentration of total $SO_4 - S$ in precipitation for winter.

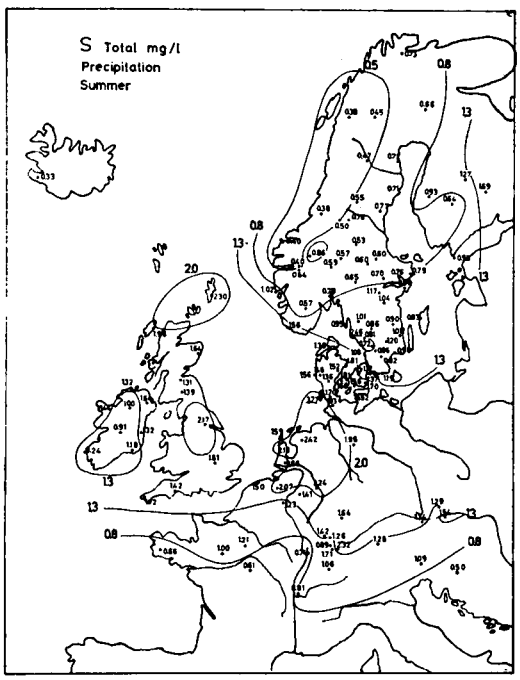

FIG. 6. Average concentration of total $SO_4 - S$ in precipitation for summer.

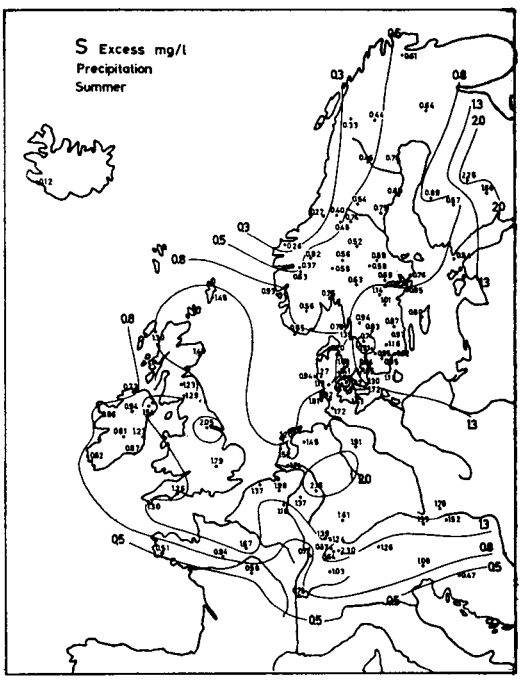

FIG. 8. Average concentration of excess $SO_4 - S$ in precipitation for summer.

comes from the sea as sea spray and accordingly the concentration drops with increasing distance from the sea. This decrease is rather pronounced over England and Scandinavia but is much slower over France and the adjacent parts of central Europe. This seems to indicate considerable anthropogenic production in these latter areas. The Cl concentration over Germany, the Benelux countries and most of France shows only a slight seasonal variation, if at all, suggesting that the continental and most likely industrial Cl sources in this area are different from the sulfur sources. Other areas of high Cl concentrations are located over Finland and close to the Alps, suggesting local sources in certain valleys.

3. Total sulfur in precipitation
(Figs. 6 and 7)

The concentration of total sulfate-sulfur in precipitation is more uniform over sea and land than that in air. The comparison with Figs. 2 and 3 may suggest that the removal efficiency by rain of sulfur compounds over the sea is larger but it should be considered that the periods of high concentrations of anthropogenic sulfur in continental air during winter time are connected with no or only little precipitation. We will discuss this in more detail in connection with Figs. 12 and 13. The highest concentrations in precipitation are found over the British Isles and in central Europe. The high values over the north western part of Scotland are remarkable and also the general tendency for lower concentrations over the southern part of the continent. In contrast to Figs. 2 and 3 the seasonal variation is only slight.

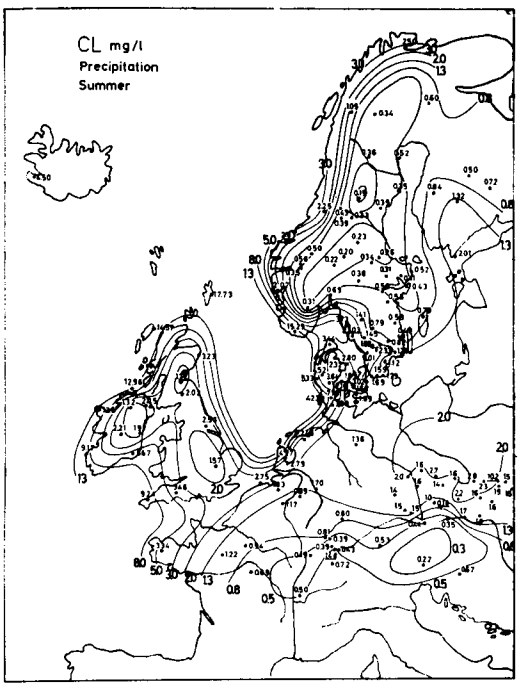

FIG. 10. Average concentration of Cl in precipitation for summer.

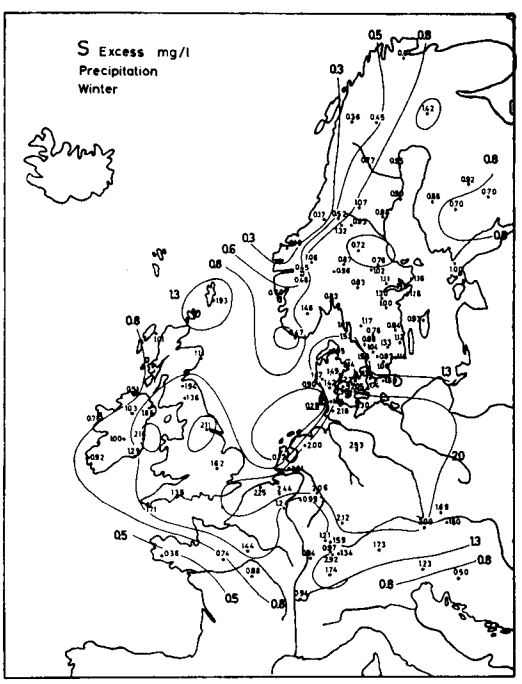

FIG. 9. Average concentration of excess $SO_4 - S$ in precipitation for winter.

4. Excess sulfur in precipitation
(Figs. 8 and 9)

It has become customary to compute as excess sulfur the sulfur concentration in rain due to sources other than sea spray by subtracting the amount of sea salt sulfate from the data in Figs. 6 and 7. The sea salt sulfate concentration is 0.0463 times the Cl concentration of Figs. 10 and 11. This procedure is based on the assumption that chloride is a conservative property of the sea spray particles and that there are no sources of chloride other than sea spray. Both these assumptions are not correct

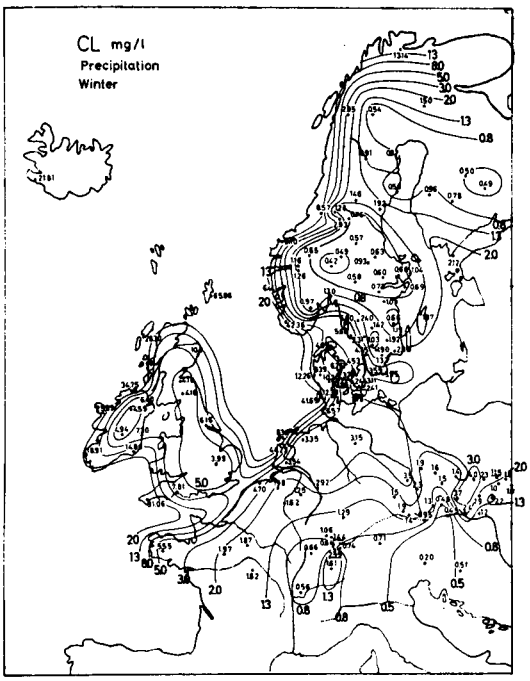

FIG. 11. Average concentration of Cl in precipitation for winter.

but the result can still be regarded as an approximation. As to be expected, Figs. 8 and 9 show some similarity in pattern with Figs. 2 and 3. However, the differences in concentration between polluted and unpolluted areas are less for excess S than for S in air. The ratio in concentration between central Europe and north Scandinavia, e.g., is about 4 for excess S and 10 for S in air. Also, excess S shows only slight seasonal variations. Both features are most likely due to high accumulations in surface air of anthropogenic SO_2 during periods of stable inversion layers and of little precipitation. Apparently the sulfur concentration in air masses which contribute to precipitation is on the average rather uniform over the year.

5. Chloride in precipitation
(Figs. 10 and 11)

Of all components, chloride in precipitation shows the greatest difference between sea and land, indicating again that the sea is the origin of most of the chloride. In coastal areas the winter concentrations are higher by factors of 2 to 3, due to the higher frequency of storms. These factors are somewhat higher than those for chlorine in air. The isolines of concentration run fairly parallel to the coast lines and the decrease in concentration is approximately logarithmic within the first few hundred kilometers inland from the coast. As in Figs. 4 and 5 the decrease is less pronounced over most of central Europe suggesting that the higher inland concentrations of 1.5 to 2 in summer and 2 to 3 in winter in this region are due to anthropogenic, most likely industrial sources. The data from Czechoslovakia fit nicely into the general pattern. There is again a general tendency of decreasing concentrations over southern continental Europe.

6. Ratio of concentration air/precipitation for sulfur and chlorine

The European chemical network offers a unique opportunity to compare the distribution in air and precipitation. Figs. 12 to 15 give the ratio for sulfur and chlorine. We like to start the discussion of these maps with some theoretical considerations about the relation between the concentration in air and in rain water.

The concentration of a substance in rain water is primarily the result of the processes within the clouds, i.e. by the processes of rainout. If c_c is the concentration of a substance in *cloud air*, expressed in $\mu g/m^3$, k_c the concentration in *cloud water* expressed in mg/liter, l the liquid water content of raining clouds and ε the net collection efficiency of all scavenging processes active in clouds it can be expected that

$$k_c = \varepsilon\, c_c/l. \qquad (1)$$

Data from various sources seem to indicate that this formula can be considered a first approximation, provided that both the values for k_c and c_c refer to *cloud air* (JUNGE, 1963). But usually k and c are measured near the earth surface as k_s and c_s. It must be expected that $k_s = \eta_k \cdot k_c$ with $\eta_k > 1$ since some matter is picked up below the clouds by washout mechanisms and because of evaporation of the falling raindrops. Similarly $c_s = \eta_c \cdot c_c$ with $\eta_c > 1$, since for substances with sources at the earth surface the concentration will normally decrease with altitude. With (1) we obtain for the concentration ratio air/precipitation

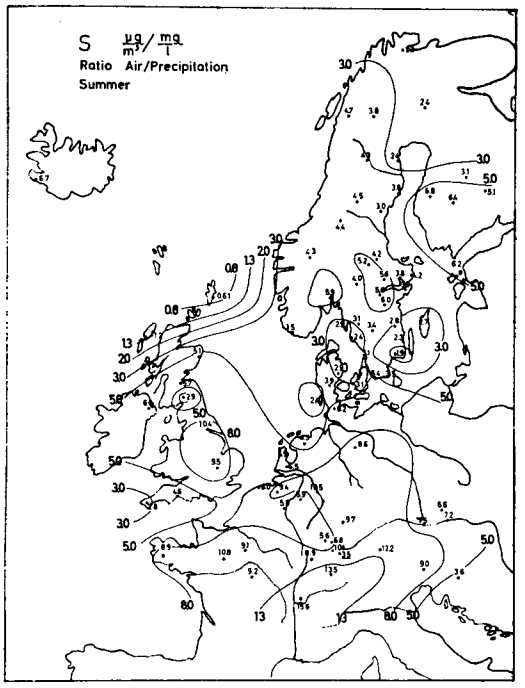

FIG. 12. Average concentration ratio air/precipitation of S for summer.

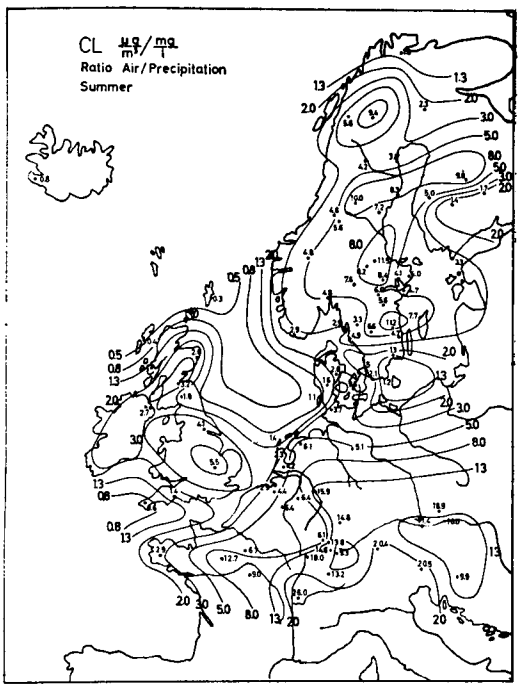

FIG. 14. Average concentration ratio air/precipitation of Cl for summer.

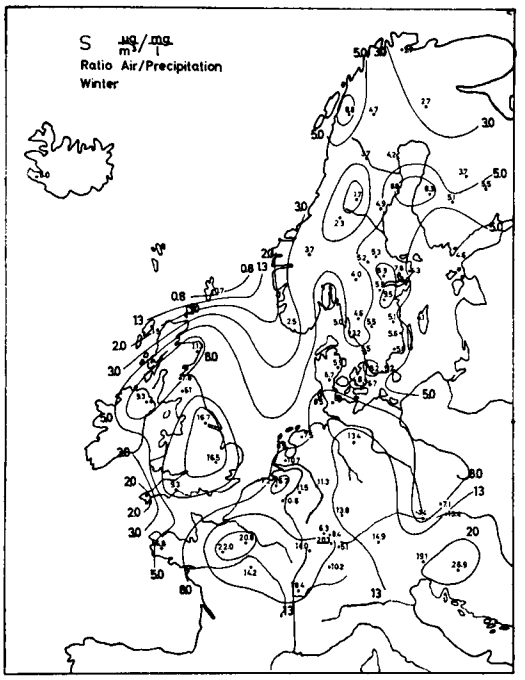

FIG. 13. Average concentration ratio air/precipitation of S for winter.

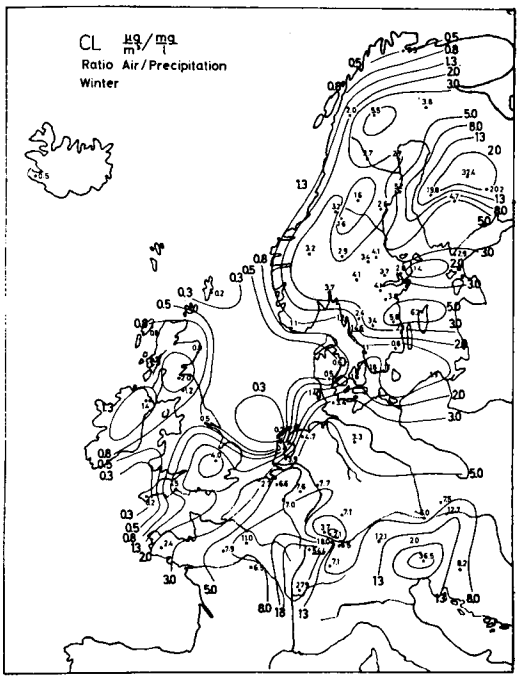

FIG. 15. Average concentration ratio air/precipitation of Cl for winter.

$$R = C_s/K_s = \frac{l}{\varepsilon}\frac{\eta_c}{\eta_k}. \quad (2)$$

Under normal conditions ε and l vary only within small ranges; ε between 0.5 and 1.0 and l between 1 and 2. Thus l/ε can be expected to vary between about 1 and 4. Variations of R beyond this range should then be due to variations of η_c/η_k. Values of $R < 1$ can be interpreted as being caused by relatively large η_k values, since η_c cannot be smaller than 1. Values of $R >$ about 4, however, can easily be explained by relatively large η_c values, i.e. by a rapid decrease of air concentration with altitude.

All these considerations about the ratio R are, of course, only valid, if on the average the values of c_s are the same for days with and without rain. Most likely the air concentrations will be larger for days without rain resulting in an increase of η_c and R.

The distribution of the ratio R for sulfur in Figs. 12 and 13 parallels roughly the distribution of sulfur in air, Figs. 2 and 3. The maximum values for R in the industrial areas of England and of the Continent are about 8 in summer and about twice as high in winter. This seasonal variation is about the same as for sulfur in air and seems to indicate the dominating role of η_c for the R values. Anticyclonic conditions with subsidence and the formation of stable inversion layers below the 800–900 level will result in high values of c_s particularly during the cold season. During such periods precipitation is light so that the monthly average rain concentrations are generally not much influenced by these accumulations of sulfur in surface air, resulting in high average η_c values. Some areas of higher R values with similar seasonal variations are also observed over Scandinavia, particularly over the southern part.

To some degree the distribution of R for Cl in Figs. 14 and 15 is similar to that for S. For large parts of the map R is between 1 and 4 as to be expected if the η values are not too far from unity. There are maxima over England, the Continent and also over Scandinavia with R values of 5–10. In most of these areas, however, the winter values are not higher than the summer values, in contrast to the sulfur data. Only in a few isolated spots, e.g. near the Alps and in Finland are the winter values of R higher indicating perhaps local production. Values higher than 4 during the warm season are difficult to explain by land sources. But it should be pointed out that the theoretical concept discussed here is only an approximation. At the present state of knowledge of the process of removal from the atmosphere of trace substances it is hardly possible to draw further conclusions. It is possible, e.g. that the l and ε values may differ systematically between land and sea or with season and that the range of variation of these parameters is somewhat larger than expected. Besides the scientific interest in the R values, maps of this kind may be useful—at least in certain areas—to derive average air concentration values in cases where only rainwater analyses are available and vice versa.

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