THE AMOUNTS OF NITROGEN AS AMMONIA AND AS NITRIC ACID, AND OF CHLORINE IN THE RAIN-WATER COLLECTED AT ROTHAMSTED.

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At the time of the commencement of the Rothamsted experiments very little was known as to the amounts of combined nitrogen and other substances present in rain-water.

Marggraf (74) found nitric acid, chlorine and lime in rain-water collected during the winter of 1749–50, and, subsequently, in snow. These analyses, which are probably the earliest[1], were followed by those of Bergman (15) who detected the same substances both in rain and snow[2]; whilst somewhat later de Saussure (99) noticed the presence of ammonia in the atmosphere. Between 1820 and 1825 several analyses of rain-water were made. Hermbstaedt (54) detected organic matter, Mulder (84) and Zimmermann (125) found chlorine and several other substances in rain-water collected at Utrecht and near Giessen; whilst von Liebig found sometimes nitric acid and invariably ammonia in a number of samples which had been collected by Zimmermann. Brandes (24) determined the amount of total solids and detected the presence of chlorine, sulphuric acid, ammonia and other substances. By this time chlorine had already been determined by Dalton (37) in rain-water collected in Manchester. The first determinations of ammonia seem to be those of Payen (47) in 1845–6. From that date to 1855

[1] Evelyn, in his *Philosophical Discourse of Earth* (London, 1676), speaks of rains and dews as being "impregnated...with Celestial Nitre" (p. 98), and of "nitrous spirits descending with their baulmy pearls" (p. 174). This does not of course indicate that nitrates had actually been detected in rain, as the terms nitrous air &c. were applied at this period—the time of Mayow's and Hooke's experiments on combustion—to what is now known as oxygen.

[2] "*Nivales* aliquantulum calcis salitae cum exiguis acidi nitri vestigiis in sinu fovent... *Pluviales* iisdem plerumque materiis inquinantur, sed majori dosi" (*Opuscula*, I. 84).

numerous analyses of rain-water were made, chiefly in France, and although many of the determinations were then, and some even much later, made without reference to the magnitude of the rainfall, some, as for instance those of Boussingault and Barral, were made in measured amounts of the total fall, so that it became possible for the first time to estimate how much of the constituents determined was supplied to a given area of land in a given period.

During the last fifty years analyses of the rain have been made in most European countries and in many other parts of the world. With few exceptions, however, the analyses extend over very short periods and partly for this reason the results are frequently conflicting.

The earliest analyses of Rothamsted rain were made in 1853–4, when the nitrogen in the form of ammonia was determined (63). Further determinations, both of ammoniacal and nitric nitrogen, were made by Way in 1855 and 1856 (121). These earlier results have already been fully discussed elsewhere (64), but to make the present record complete they are given in Appendix Tables III and IV, which contain the whole of the results relating to nitrogen obtained up to the end of the harvest year 1904–1905.

The determinations of ammonia in monthly samples of the rain were recommenced in 1877, and were continued with some interruptions until December, 1885. Since January, 1888, however, ammonia has been regularly determined each month.

Nitric nitrogen has been determined uninterruptedly since September, 1886; for some months by Schloesing's method and, subsequently, by Williams' zinc-copper couple method[1]. Chlorine has been determined each month since June, 1877 (Appendix Table V); and sulphuric acid, in mixed samples, representing winter and summer rain, from 1881 to 1887 (65 and 117).

Besides the monthly samples of rain, a large number of single samples have been analysed at Rothamsted (64), and in addition about 80 samples were analysed by the late Sir Edward Frankland (44).

NITROGEN AS AMMONIA AND AS NITRATES (AND NITRITES) IN ROTHAMSTED RAIN.

The average amounts of nitrogen in the forms of ammonia and nitric (and nitrous) acid in the Rothamsted rainfall during the thirteen harvest years, 1888–9 to 1900–1, is 3·84 lbs. per acre per

[1] *Trans. Chem. Soc.* 1881, 39, 100. For the minutiae of this and other methods employed at Rothamsted in the analysis of rain-water see Warington (118).

annum, the relative amounts of ammoniacal and nitric nitrogen being 70 and 30 per cent. of the total. The greatest amount of nitrogen in these forms during the above period was 4·43 lbs. (1899–1900), and the lowest amount 3·31 lbs. The variations from year to year (see Table 1)

TABLE 1.

NITROGEN AS AMMONIA AND NITRIC ACID IN RAIN-WATER COLLECTED AT ROTHAMSTED.

September 1 to August 31	Rainfall	Nitrogen						
		Per million		Per acre			% of Total	
		as NH_3	as N_2O_5	as NH_3	as N_2O_5	Total	as NH_3	as N_2O_5
	inches			lb.	lb.	lb.		
1853–4*	29·01	0·79	—	5·20	—	—	—	—
1855**	29·17	0·88	0·11	5·82	0·72	6·54	89·0	11·0
1856**	27·22	1·18	0·12	7·28	0·76	8·04	90·5	9·5
1881–2	32·31	0·33	—	2·443	—	—	—	—
1882–3	34·71	0·34	—	2·665	—	—	—	—
1883–4	25·77	0·40	—	2·334	—	—	—	—
1884–5	26·78	0·37	—	2·240	—	—	—	—
1886–7	23·61	—	0·138	—	0·736	—	—	—
1887–8	30·50	—	0·116	—	0·803	—	—	—
1888–9	30·09	0·412	0·134	2·806	0·911	3·717	75·5	24·5
1889–90	27·43	0·445	0·119	2·762	0·741	3·503	78·8	21·2
1890–1	23·41	0·483	0·195	2·560	1·033	3·593	71·2	28·8
1891–2	29·68	0·466	0·185	3·130	1·240	4·370	71·6	28·4
1892–3	28·94	0·535	0·217	2·914	1·185	4·099	71·1	28·9
1893–4	29·55	0·381	0·131	2·549	0·878	3·427	74·4	25·6
1894–5	28·94	0·364	0·161	2·397	1·068	3·465	69·2	30·8
1895–6	24·37	0·484	0·216	2·670	1·193	3·863	69·1	30·9
1896–7	37·24	0·350	0·160	2·947	1·346	4·293	68·6	31·4
1897–8	19·51	0·516	0·234	2·279	1·032	3·311	68·8	31·2
1898–9	24·69	0·443	0·228	2·474	1·276	3·750	66·0	34·0
1899–1900	31·02	0·431	0·200	3·028	1·401	4·429	68·4	31·6
1900–1	24·30	0·498	0·250	2·737	1·375	4·112	66·6	33·4

* March to February. ** January to December.

probably depend, in part, on the distribution of the rainfall, but seem to have no very definite relation to the total annual amount of rain. The lowest amount of nitrogen found during this period was, however, coincident with the lowest rainfall (19·51 inches in 1897–8). The large amounts of nitrogen found in the rain collected in the years 1853–6, are evidently mainly due to the ammonia results being far too high. As has been pointed out elsewhere (64), the methods employed at that time left a good deal to be desired.

The amounts of nitrogen in the monthly samples of rain depend partly on the temperature and partly on the amount of rain. The smallest amount of nitrogen is found in February, being coincident with the lowest rainfall, whilst the largest amount occurs in August, when, in addition to an increased temperature, there is also a large amount of rain only exceeded in October.

TABLE 2.

AVERAGE MONTHLY AMOUNTS OF NITROGEN AS AMMONIA AND NITRIC ACID IN ROTHAMSTED RAIN.

1888-9 to 1900-1	Rainfall	Nitrogen						
		Per million		Per acre			°/₀ of Total	
		as NH_3	as N_2O_5	as NH_3	as N_2O_5	Total	as NH_3	as N_2O_5
	inches			lb.	lb.	lb.		
September	2·10	0·518	0·202	0·246	0·096	0·342	71·9	28·1
October............	3·23	0·327	0·155	0·239	0·113	0·352	67·9	32·1
November.........	2·83	0·386	0·164	0·247	0·105	0·352	70·2	29·8
December.........	2·51	0·384	0·176	0·218	0·100	0·318	68·6	31·4
January	1·99	0·402	0·162	0·181	0·073	0·254	71·3	28·7
February	1·79	0·398	0·200	0·161	0·081	0·242	66·5	33·5
March	1·97	0·420	0·215	0·187	0·096	0·283	66·1	33·9
April	1·57	0·540	0·222	0·192	0·079	0·271	70·8	29·2
May	2·00	0·504	0·203	0·228	0·092	0·320	71·3	28·7
June	1·79	0·543	0·190	0·220	0·077	0·297	74·1	25·9
July	2·63	0·482	0·178	0·287	0·106	0·393	73·0	27·0
August	2·84	0·476	0·171	0·306	0·110	0·416	73·6	26·4
Sept.—Dec.	10·67	0·394	0·171	0·950	0·414	1·364	69·6	30·4
Jan.—April	7·32	0·435	0·199	0·721	0·329	1·050	68·7	31·3
May—August ...	9·26	0·497	0·184	1 041	0·385	1·426	73·0	27·0
April—Sept.......	12·93	0·506	0·191	1·479	0·560	2·039	72·5	27·5
Oct.—March ...	14·32	0·381	0·175	1·233	0·568	1·801	68·5	31·5
Whole year	27·25	0·440	0·183	2·712	1·128	3·840	70·6	29·4

Of greater interest is the comparison of the summer with the winter rain. The rainfall itself of the two seasons does not differ materially in quantity, although the winter months show a slight excess over the summer. Reference to Table 2 will show that whilst in the winter the total nitrogen amounts to 1·80 lbs. per acre, the summer rain contains 2·04 lbs., the excess being entirely due to an increased production of ammonia, since the nitric nitrogen remains constant. Of the total nitrogen the summer rain contains 72·5 per cent. in the form of ammonia and 27·5 per cent. in the form of nitrates (and nitrites); the winter rain contains 68·5 per cent. and 31·5 per cent. respectively. In other words, to 1 part of nitric nitrogen there are 2·55 parts of ammoniacal nitrogen in the summer, and only 2·15 parts in the winter months.

The influence of the amount of the rainfall on the composition of the rain is illustrated by the results given in Table 3, which shows the average amounts of nitrogen in the two forms in monthly samples of rain below 1 inch, and above 4 inches respectively during the last thirteen years. It happens that in both cases there were exactly the same number of samples (eighteen).

TABLE 3.

NITROGEN AS AMMONIA AND NITRIC ACID IN MONTHLY RAINFALLS.

	Rainfall below 1 inch			Rainfall above 4 inches		
	Rainfall	N. per million		Rainfall	N. per million	
		as Ammonia	as Nitrates		as Ammonia	as Nitrates
	inches			inches		
Min. ...	0·09	6·236	2·161	4·03	0·314	0·140
Max. ...	0·96	0·686	0·285	8·08	0·224	0·090
Mean ...	0·65	0·965	0·442	4·92	0·278	0·124

As was to be expected from the relatively greater variations in the amounts of rainfall below 1 inch, the amounts of nitrogen also differ widely; the average amount of nitrogen per million in the low rainfall is, however, more than twice as high as in the higher rainfall.

Comparing the amount of total nitrogen in the Rothamsted rain with the amounts found, during the past forty years, in various parts of the world (Table 4), the majority of the results are a good deal higher than those just described. In a few cases, notably Paris and Copenhagen, this may no doubt be attributed to the rain having been collected in, or near large towns. It was to be hoped that the considerable number of results now brought together would enable some kind of grouping to be made; that, for instance, the total nitrogen would show, at any rate in extreme cases, some relation to the rainfall, or would be influenced by proximity to, or distance from, the sea. It is evident, however, that whilst the total nitrogen may vary enormously under apparently quite similar climatic conditions, differences of climate are not necessarily coincident with great variation in the composition of the rain. The rain, for instance, collected at Manhattan, which is about 700 miles from the Gulf of Mexico and more than 1,100 miles from the Pacific, closely resembles the Rothamsted rain. Then again, the 102 inches of rain which fall in British Guiana do not supply to the soil more, but rather less, nitrogen than the 27 inches at Rothamsted.

It will, however, be convenient to consider separately the rain falling in temperate and in tropical countries.

In non-tropical rain the relation of ammoniacal to nitric nitrogen varies less, perhaps, than might be expected from the differences in the amounts of total nitrogen, and the average of all the results recorded in the table (p. 286) is very similar to that found at Rothamsted. With the one exception of New Zealand, the ammonia is greatly in excess of the nitric acid. In the rain of Lincoln, New Zealand, the relation is reversed, the nitric nitrogen being in excess, as in the case of some tropical rains. This resemblance is, however, only partial, since the relation of ammonia nitrogen to nitric nitrogen in New Zealand rain is not due to a high amount of nitrates, but to an unusually small amount of ammonia. This is possibly due to the prevalence of sea-winds, as it has been pointed out by Anderlind (2) that the results obtained by Merino (79) in the north-west of Spain show less ammonia when the wind is from the sea than during an overland wind. Heinrich obtained similar indications at Rostock (53). On the other hand Schloesing believes that the sea is the chief source of the ammonia present in the air.

Exceptionally high relations of ammonia to nitric acid occur in the case of Ploty (122) and Pretoria (113); whilst the rain collected at Tokio shows a decidedly low proportion of ammonia to nitric acid. The results obtained at the ten different places (excluding New Zealand) at

TABLE 4.

NITROGEN AS AMMONIA AND NITRIC ACID IN RAIN.

	Date	Rainfall	Nitrogen: Per million as NH_3	Nitrogen: Per million as N_2O_5	Nitrogen: Per acre per annum as NH_3	Nitrogen: Per acre per annum as N_2O_5	Nitrogen: Per acre per annum Total	Nitrogen: °/₀ of Total as NH_3	Nitrogen: °/₀ of Total as N_2O_5
		inches			lb.	lb.	lb.		
Rothamsted	1888–9, 1900–1	27·25	0·440	0·183	2·71	1·13	3·84	70·6	29·4
Copenhagen (114)	1880–85	21·95	1·97	0·473	9·27	2·21	11·48	80·8	19·2
Gembloux (93)	1889–91	27·23	1·143	0·345	7·07	2·14	9·21	76·8	23·2
Montsouris (67)	1876–1900	21·52	2·13	0·66	10·37	3·22	13·59	76·3	23·7
Mettray (66)	1877	29·90	0·409	—	2·77	—	—	—	—
Dahme (39)	1865	17·09	1·42	0·30	5·50	1·16	6·66	82·6	17·4
Ida-Marienhütte (25)	1865–70	22·65	—	—	—	—	9·92	—	—
Insterburg (39)	1864–6	25·67	0·65	0·38	3·90	2·25	6·15	63·1	36·9
Kuschen (39)	1864–6	14·78	0·48	0·16	1·63	0·55	2·18	75·0	25·0
Proskau (39)	1864–5	17·81	3·21	1·73	12·94	6·97	19·91	65·0	35·0
Regenwalde (39)	1864–7	22·72	2·08	0·62	10·69	3·28	13·97	77·0	23·0
Rostock (53)	1880–1	33·27	0·892	—	6·73	—	—	—	—
Florence (11)	1869–75	38·31	1·004	0·57	8·70	3·09	11·79	73·8	26·2
Vallombrosa (11)	1872–5	59·89	0·617	0·253	8·36	3·46	11·82	70·7	29·3
Scandicci (90)	1888–90	29·18	0·614	0·266	4·06	1·76	5·82	69·8	30·2
Catania (10)	1888–9	18·36	0·327	0·161	1·36	0·67	2·03	66·9	33·1
St Michele, Tirol (69)	1885–6	43·93	1·188	0·579	11·83	5·76	17·59	67·3	32·7
Libwerd, Bohemia	1877–8	24·41	1·3	0·61	7·18	3·37	10·55	68·1	31·9
Peček, ,,	1883-4 to 85-6	19·34	1·26	0·50	5·53	2·19	7·72	71·6	28·4
Ploty (122)	1900–3	17·49	0·854	0·061	3·38	0·24	3·62	93·3	6·7
Pretoria (113)	1904*	—	0·68	0·12	—	—	—	85·1	14·9
Tokio (59)	1883–4	52·67	—	0·093	—	1·11	} 2·88	61·6	38·4
,, ,,	1885	62·28	0·126	—	1·77	—			
New Zealand (51)	1884–8	29·70	0·076	0·169	0·50	1·13	1·63	30·7	69·3
Kansas (42)	1887–9	29·41	0·393	0·154	2·62	1·03	3·64	71·8	28·2
Mississippi (112)	1894–5	44·11	0·235	0·074	2·35	0·74	3·09	76·0	24·0

TROPICAL RAIN.

Calcutta	1891	46·01	0·172	0·115	1·79	1·20	2·99	59·7	40·3
Madras	1888–93	39·21	—	—	—	—	1·91	—	—
Ceylon (4)	1898–9	82·13	0·196	0·069	3·65	1·28	4·93	72·0	28·0
East Java (75)	1891	(47)	0·11	0·06	1·13	0·71	1·84	61·5	38·5
Mauritius (21)	1895	(70)	0·43	0·40	6·81	6·34	13·15	51·8	48·2
Réunion (88)	1886–7	(40)	—	0·69	—	6·24	—	—	—
Barbados (5)	1885–97	63·95	0·084	0·268	1·22	3·88	5·10	23·9	76·1
Venezuela (71 and 88)	1883–5	(40)	1·55	0·58	14·03	5·20	19·23	72·8	27·2
British Guiana† (52)	1890-1900	102·41	0·055	0·078	1·17	1·82	2·99	39·1	60·9
Campinas (30)	1890	—	0·99	—	—	—	—	—	—

* Feb. to June only. † In part unpublished. Communicated by Prof. J. B. Harrison.

which analyses have been continued for at least four years, give an average of 73·5 of nitrogen as ammonia to 26·5 nitric nitrogen.

As regards the tropics, the rain collected in British Guiana and Barbados contains a large excess of nitric over ammoniacal nitrogen, attributed by Harrison (52) to the prevalence of violent thunderstorms. In Mauritius, however, there is a slight excess of nitrogen in the form of ammonia; whilst the rain collected at Caracas, Venezuela, at Calcutta, and at Colombo contains a more or less considerable excess of ammoniacal nitrogen in relation to nitric nitrogen.

The only explanation of these differences seems to be the one offered in the case of New Zealand rain, but this leaves quite unaccounted for the discrepancies shown in the rain of Java and Mauritius and again in that of British Guiana and Venezuela.

The one conclusion which may safely be drawn is that tropical rain does not supply to the soil an essentially greater amount of nitrogen than the rain of temperate climates. The average for the seven tropical places in which both forms of nitrogen have been determined is 7·2 lbs. of nitrogen per acre per annum. And this includes the exceptional amounts found at Caracas and in Mauritius. When these abnormal results are omitted the average total nitrogen for tropical countries is only 3·58 lbs. per acre, with a high average rainfall of 68·3 inches.

NITROGEN AS NITROUS ACID IN RAIN.

The nitrogen in the form of nitrites has not been separately determined in the rain at Rothamsted, being included along with the nitrates, by the reduction method employed; and very few determinations seem to have been made elsewhere. Such results, however, as are available (Table 5) indicate that the amounts of nitrogen present in this form are usually insignificant.

The results of qualitative experiments made by Failyer and Willard with rain collected at Manhattan (42) showed that from December to March only 28 per cent. of the samples contained nitrous nitrogen, whilst from June to September 89 per cent. of the samples gave positive results. There is, however, no evidence to show whether the conditions of the summer months are favourable to the production or merely to the conservation of nitrites.

With regard to the amount of organic nitrogen in the rain-water, the only available analyses relating to Rothamsted are those of

Frankland who found from 0·03 to 0·66 per million in 69 samples. The average amount (0·19 per million) equalled rather more than one-third of the nitrogen present in the forms of ammonia and nitric acid (0·51 per million). The total nitrogen in the Rothamsted rain is, therefore, 3·84 + 1·35 lb., or about 5·2 lb. per acre per annum.

In rain-water collected in New Zealand, Gray found 0·45 lb. of albuminoid nitrogen per acre per annum.

TABLE 5.

NITROGEN AS NITROUS AND NITRIC ACIDS IN RAIN AND SNOW.

	Rainfall	Nitrogen							
		Per million						Per acre	
		as Nitrites			as Nitrates			as Nitrites	as Nitrates
		Min.	Max.	Mean	Min.	Max.	Mean		
	inches							lb.	lb.
St Chamas (31)......	(20)	0·0	0·316	0·133	0·001	0·716	0·162	(0·60)	(0·73)
,, (snow)...	—	—	—	0·250	—	—	—	—	—
Langres (31) ,, ...	—	—	—	0·270	—	—	0·881	—	—
Scandicci (90)	27·28	0·0	0·0168	0·0045	0·033	0·729	0·255	0·028	1·687
Catania (10)	18·36	0·0001	0·0027	0·0006	0·035	0·244	0·161	0·003	0·671
Ploty (122)............	17·49	0·0	0·025	0·011	0·002	0·178	0·050	0·043	0·197

In addition to the nitrogen brought to the soil by rain, Schloesing (104) has shown that soils may absorb as much as 47 kilos. of nitrogen per hectare per annum directly from the air. He found that the character of the soil has no great influence on the absorption, but that the presence of moisture acts favourably owing to the increased nitrification of the ammonia absorbed, and the resulting reduction of the tension of the ammonia in the soil. Employing 20 per cent. hydrochloric acid as the absorbent, Heinrich (53) found that the ammonia thus fixed amounted to 30·6 kilos. per hectare; but he considers that the absorption would probably be less over a large surface, and that the calculation per hectare from a small experiment is not altogether admissible. Müller (84) and Kellner (59), who both employed dilute sulphuric acid, obtained results indicating an absorption of only 12 and 13 kilos. respectively of nitrogen per hectare per annum. As these amounts include a certain quantity of ammonia—perhaps most of it—originally derived from the soil, some of which would, under ordinary conditions, be brought down by the rain, the actual gain due to direct absorption must be less than that indicated by experiments such as those referred to.

CHLORINE AND SULPHURIC ACID.

As already stated the chlorine in the monthly samples of rain has been determined since 1877–8, that is to say for 28 years. The annual amounts of chlorine per acre show great variations, which are to a great extent independent of the total yearly rainfall.

	Date	Rainfall	Chlorine	
			Per million	Per acre
		inches		lb.
Minimum rainfall......	1897–8	19·51	3·74	16·51
Maximum ,, ...	1878–9	41·05	1·69	15·73
Minimum chlorine ...	1889–90	27·43	1·66	10·32
Maximum ,, ...	1896–7	37·24	2·51	21·19

The maximum and minimum annual rainfalls show, therefore, no very material difference in the amounts of chlorine per acre (both are somewhat in excess of the average), and results may be obtained, quite independently of the total rainfall, which differ by 100 per cent., and are evidently mainly due, on the one hand to a deficiency, and on the other to an excess of rain during the winter months, the rainfall of which, as will presently be shown, contains more than twice as much chlorine per million as that of the summer months.

The amount of chlorine per million varies with considerable regularity from month to month. The lowest amount occurs in July, after which there is a rise until January, when the highest amount is reached. In February there is a drop, followed by a rise in March and then by a fall until July. The amounts per acre show similar variations but with somewhat less regularity.

TABLE 6.

AVERAGE AMOUNT OF CHLORINE IN ROTHAMSTED RAIN.

1877-8 to 1900-1901	Rainfall (average 24 years) Inches		Chlorine			
			Per million		Per acre (lb.)	
September	2·46		1·62		0·90	
October	3·24	11·22	2·46	2·61	1·80	6·41
November	3·03		2·71		1·86	
December	2·50		3·27		1·85	
January	2·06		3·80		1·77	
February	2·03	7·79	3·05	3·16	1·40	5·57
March	1·79		3·56		1·44	
April	1·91		2·22		0·96	
May	2·16		1·76		0·86	
June	2·22	9·79	1·29	1·31	0·65	2·89
July	2·59		1·06		0·62	
August	2·79		1·20		0·76	
Summer (April—September)	14·13		1·49		4·75	
Winter (October—March)	14·65		3·05		10·12	
Whole year	28·78		2·28		14·87	

With regard to the relation between the quantity of the monthly rainfall and the amount of chlorine, the results (see Table 7) show a regular decrease in the chlorine as the rainfall increases, although not in the

TABLE 7.

AVERAGE AMOUNTS OF CHLORINE IN MONTHLY RAINFALLS GROUPED ACCORDING TO THE AMOUNT OF THE FALL.

	Whole Year			Winter			Summer		
	Average Rainfall	Chlorine Per million	Chlorine Per acre	Average Rainfall	Chlorine Per million	Chlorine Per acre	Average Rainfall	Chlorine Per million	Chlorine Per acre
	inches		lb.	inches		lb.	inches		lb.
Below 1 in.	0·72	4·11	0·67	0·74	4·95	0·83	0·67	3·23	0·49
1–2 inches	1·43	3·06	0·99	1·46	3·93	1·30	1·41	2·19	0·70
2–3 inches	2·51	2·34	1·33	2·52	3·28	1·87	2·46	1·53	0·85
3–4 inches	3·52	2·24	1·78	3·51	2·76	2·19	3·53	1·41	1·13
Above 4 in.	4·99	1·59	1·80	4·89	2·44	2·70	5·08	0·86	0·99

same proportion, the rate of decrease in the chlorine being much less than the rate of increase in the rain. In this respect the summer and winter rains show great difference. In the summer the monthly rainfalls of 4 inches or more (average 5·08 inches) contain about a quarter as much chlorine per million as is found in the rainfalls of less than 1 inch (average 0·67 inch). With corresponding winter rainfalls the higher amounts of rain contain half as much chlorine as the lower.

No recent determinations of sulphuric acid have been made in the rain at Rothamsted. In the following table is a summary of the results obtained in 1881–7, with the chlorine results, for the same period, for comparison.

TABLE 8.

SULPHURIC ACID AND CHLORINE IN ROTHAMSTED RAIN.

April 1881 to March 1887	Rainfall	Per million		Per acre		SO_3 to 1 Cl
		Cl	SO_3	Cl	SO_3	
	inches			lb.	lb.	
April to September ...	13·90	1·31	2·77	4·11	8·71	2·12
October to March	16·05	2·89	2·39	10·51	8·70	0·83
Whole year............	29·95	2·16	2·57	14·62	17·41	1·19

The similarity of the amounts of sulphuric acid in the summer and winter rain is very striking, especially in view of the great variation in the chlorine results. The slightly higher results obtained in the summer as compared with the winter accord with observations made by Russell in London, and support the view that the sulphuric acid is to a great extent an oxidised product of the decomposition of organic matters. Very little of it can be derived from the sea, since the relation of sulphuric acid to chlorine in sea-water is only 11 : 100; and Russell found that rain-water from Dartmoor collected during a strong sea-wind contained only a trace of sulphuric acid.

Compared with London rain the amount of sulphuric acid found at Rothamsted is small; but it is somewhat in excess of the amount found in New Zealand. Sestini found 5·02 per million, equivalent to 20·89 lb. per acre, in the rain collected at Catania.

TABLE 9.

CHLORINE AND SULPHURIC ACID IN RAIN.

	Date	Rainfall	Per million				Per acre		SO_3 to 1 Cl
			Chlorine			Sul-phuric acid	Chlorine	Sul-phuric acid	
			Min.	Max.	Mean				
		inches					lb.	lb.	
Rothamsted	1877–8 } 1900–1 }	28·78	0·40	20·10	2·28	2·57*	14·87	17·41*	1·19
Cirencester (60)	1874–1900	30·61	1·15	10·38	3·17	—	21·90	—	—
Scandicci (91)	1889–91	27·67	0·17	31·95	5·63	—	35·35	—	—
Perugia (14)	1886–7	33·96	1·38	40·28	3·15	—	24·22	—	—
Catania (10)	1888–9	18·36	1·47	7·36	5·48	5·02	22·79	20·89	0·92
La Guardia (79)	1892–3	56·42	7·1	71·9	31·2	—	399·5	—	—
New Zealand (51)	1884–8	29·70	2·6	36·4	8·83	2·22	59·44	14·94	0·25
Barbados (5)	1885–97	63·95	3·55	33·97	8·14	—	127·8	—	—
British Guiana (52)	1890–1900	102·41	(1·68)	(17·68)	5·04	—	116·88	—	—
Ceylon (4)	1898–9	82·13	—	—	9·72	—	180·63	—	—
Calcutta	1894	46·01	1·82	5·44	3·16	—	32·87	—	—
Madras	1888–93	39·21	—	—	4·08	—	36·27	—	—

* 1881–87.

All the very high chlorine results recorded in Table 9 are to be accounted for by proximity to the sea. In this connexion it may be mentioned that Frankland found as much as 218 parts of chlorine per million in rain collected at Land's End, at a height of about 100 feet above the sea.

It has been pointed out by Kinch (60) that the quantity of chlorine brought down by the rain at Rothamsted and at Cirencester is sufficient for the requirements of most crops. The same may be said as regards the sulphuric acid in the rain at Rothamsted.

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APPENDIX—TABLE I.

SUMMARY OF METEOROLOGICAL OBSERVATIONS AT ROTHAMSTED.

	Rainfall, Average 53 years 1852–1905		Bright sunshine, Average 15 years 1890–1905		Temperature, Average 27 years 1878–1905		
	Total	Days with 0·01 inch	Total	Per cent.	Min.	Max.	Mean
	inches		hours		deg. F.	deg. F.	deg. F.
September............	2·49	13	160·5	43	47·5	64·1	55·8
October	3·18	18	107·0	33	41·1	54·9	48·0
November	2·62	16	58·8	23	36·7	48·3	42·5
December	2·33	16	41·8	18	32·4	42·9	37·7
January...............	2·36	16	51·6	21	31·4	41·7	36·5
February	1·80	13	72·8	27	32·5	43·9	38·2
March	1·84	13	117·5	32	33·5	48·3	40·9
April	1·86	13	160·1	39	37·0	54·2	45·6
May	2·19	13	195·2	40	42·1	60·2	51·1
June	2·38	12	196·3	40	48·3	66·4	57·4
July	2·59	13	217·4	44	51·8	70·0	60·9
August	2·67	14	199·0	44	51·1	68·4	59·8
Year............	28·31	170	1578·0	34	40·5	55·3	47·9

APPENDIX—TABLE II.

Rainfall at Rothamsted.

	Sept.	Oct.	Nov.	Dec.	Jan.	Feb.	March	April	May	June	July	Aug.	Year
	inches	inches	inches	inches	inches	inches	inches	inches	inches	inches	inches	inches	inches
1852–3	—	—	—	—	—	—	2·36	3·00	1·68	3·40	4·48	2·98	—
1853–4	2·01	3·66	2·05	0·41	2·03	0·95	0·51	0·50	4·38	0·76	1·05	2·82	21·13
1854–5	0·78	2·29	1·53	1·76	0·60	0·99	2·36	0·41	2·32	1·65	6·96	2·63	24·28
1855–6	1·55	5·50	2·47	1·72	2·78	1·35	1·00	2·61	4·71	1·91	1·48	2·65	29·73
1856–7	2·19	2·87	1·42	2·24	3·71	0·57	1·48	2·16	1·11	2·22	1·61	3·08	24·66
1877–8	1·53	1·95	5·16	2·28	1·75	1·80	0·98	4·09	4·97	2·51	0·65	4·98	32·65
1878–9	1·46	2·99	4·55	1·60	2·85	3·80	1·18	2·79	3·48	5·55	4·24	6·56	41·05
1879–80	3·13	0·82	0·81	0·82	0·55	2·90	1·13	2·16	0·74	1·97	5·26	1·07	21·36
1880–1	5·86	5·94	2·92	3·47	1·14	3·70	2·15	1·00	1·38	1·63	1·76	5·82	36·77
1881–2	2·17	3·05	3·47	4·38	1·57	2·02	1·57	3·92	2·07	3·93	2·09	2·07	32·31
1882–3	2·29	6·52	3·44	3·28	3·30	4·34	0·89	1·48	1·89	2·23	4·21	0·84	34·71
1883–4	3·99	2·49	3·52	1·16	2·56	1·42	1·66	1·79	0·64	2·50	2·44	1·60	25·77
1884–5	2·18	1·70	2·05	3·06	2·99	2·85	1·46	2·88	2·88	2·76	0·38	1·59	26·78
1885–6	4·39	4·82	3·77	1·33	3·44	0·61	1·59	1·96	4·24	1·23	2·42	1·22	31·02
1886–7	1·51	3·94	2·77	4·21	2·39	0·95	1·76	1·19	2·35	0·71	0·79	1·04	23·61
1887–8	3·11	1·69	3·41	1·66	0·94	1·03	3·13	2·14	1·28	4·87	3·86	3·38	30·50
1888–9	1·03	1·09	4·45	1·69	1·29	1·95	1·89	2·47	5·00	1·38	5·67	2·18	30·09
1889–90	2·44	3·62	1·21	1·46	2·94	0·82	2·78	1·31	1·38	2·40	4·56	2·51	27·43
1890–1	1·20	1·57	2·76	0·56	2·25	0·09	1·76	1·50	3·46	1·89	2·34	4·03	23·41
1891–2	1·39	6·76	2·25	4·13	1·01	1·48	1·22	0·79	1·40	2·56	3·00	3·69	29·68
1892–3	2·46	3·99	2·06	1·63	2·05	3·62	0·42	0·25	1·22	1·00	3·00	2·38	24·08
1893–4	1·14	4·46	2·92	2·63	2·38	1·96	2·19	1·71	2·07	2·01	2·40	3·68	29·55
1894–5	2·22	3·45	4·98	2·18	2·23	0·19	1·91	1·47	0·69	0·45	5·12	4·05	28·94
1895–6	1·06	2·69	4·96	2·34	1·12	0·59	3·75	0·95	0·48	2·25	1·27	2·91	24·37
1896–7	8·08	4·13	1·39	4·42	2·03	2·92	4·20	1·91	1·72	2·73	0·47	3·24	37·24
1897–8	2·44	0·96	1·05	3·50	0·80	1·10	1·06	1·44	2·89	1·61	1·45	1·21	19·51
1898–9	0·60	2·89	2·44	3·01	2·96	2·44	0·87	2·73	2·81	1·58	1·28	1·09	24·70
1899–1900	2·46	3·75	3·76	1·41	3·67	4·91	0·96	1·33	1·08	2·63	1·13	3·93	31·02
1900–1	0·84	2·60	2·60	3·65	1·18	1·26	2·57	2·51	1·81	0·84	2·44	2·00	24·30
1901–2	1·35	2·03	1·05	4·13	0·83	1·25	1·49	0·83	2·20	3·33	1·24	3·53	23·26
1902–3	1·05	1·88	1·95	1·39	2·55	1·06	3·47	1·53	2·22	6·12	4·08	3·96	31·26
1903–4	2·75	6·32	2·21	2·42	3·50	3·45	1·58	1·25	2·15	0·81	2·92	2·15	31·51
1904–5	1·59	1·37	1·67	2·48	1·34	0·95	3·57	2·22	1·13	4·05	1·47	3·46	25·30

APPENDIX—TABLE III.

Nitrogen as Ammonia in Rain-Water collected at Rothamsted in parts per Million.

	Sept.	Oct.	Nov.	Dec.	Jan.	Feb.	March	April	May	June	July	Aug.	Year
1852–3	—	—	—	—	—	—	1·19	0·67	1·10	1·05	0·77	0·69	—
1853–4	0·61	0·57	0·66	1·33	0·64	0·78	0·78	0·80	0·37	—	—	—	—
1854–5	—	—	—	—	1·08	1·22	1·01	1·45	0·94	1·59	0·72	0·94	—
1855–6	1·12	0·72	0·64	0·79	0·93	1·60	1·09	1·72	1·49	1·33	1·00	0·82	1·08
1856–7	1·42	0·71	0·94	0·94	—	—	—	—	—	—	—	—	—
1877–8	—	0·350	—	—	—	—	0·357	0·466	—	—	—	—	—
1878–9	0·576	—	—	—	0·219	0·298	0·638	0·617	0·470	0·384	—	—	—
1879–80	0·412	0·988	—	1·038	0·495	0·371	0·371	0·881	1·276	0·508	0·309	—	—
1880–1	0·160	0·165	—	0·162	0·659	0·467	—	0·604	0·631	0·412	0·618	0·178	—
1881–2	0·350	0·214	0·237	0·196	0·422	0·227	0·313	0·319	0·535	0·445	0·503	0·453	0·334
1882–3	0·401	0·254	0·137	0·360	0·213	0·199	0·856	0·576	0·412	0·391	0·432	0·725	0·339
1883–4	0·362	0·280	0·190	0·824	0·268	0·453	0·412	0·617	0·617	0·576	0·350	0·463	0·400
1884–5	0·275	0·412	0·361	0·230	0·319	0·213	0·453	0·535	0·350	0·350	1·071	0·638	0·370
1885–6	0·178	0·247	0·275	0·577	—	—	—	—	—	—	—	—	—
1887–8	—	—	—	0·337	—	0·538	0·315	0·600	0·256	0·500	0·388	0·288	—
1888–9	1·025	0·525	0·313	0·500	0·575	0·238	0·400	0·575	0·300	0·378	0·350	0·538	0·412
1889–90	0·625	0·338	0·625	0·413	0·363	0·625	0·288	0·600	0·625	0·463	0·363	0·425	0·445
1890–1	0·775	0·475	0·225	1·300	0·450	0·250	0·550	0·625	0·350	0·719	0·417	0·313	0·483
1891–2	0·594	0·300	0·495	0·291	0·562	0·656	1·000	0·800	0·700	0·425	0·700	0·300	0·466
1892–3	0·625	0·250	0·675	0·225	0·500	0·350	1·625		0·950	1·000	0·500	0·650	0·535
1893–4	0·550	0·200	0·233	0·250	0·350	0·375	0·213	0·625	0·458	0·625	0·400	0·583	0·381
1894–5	0·625	0·300	0·150	0·325	0·300	0·958	0·500	0·575	1·167	1·437	0·225	0·357	0·364
1895–6	0·700	0·325	0·287	0·350	0·650	0·950	0·350		0·800	0·675	0·900	0·675	0·484
1896–7	0·225	0·250	0·350	0·275	0·400	0·375	0·233	0·383	0·625	0·525	1·375	0·525	0·350
1897–8	0·400	0·688	0·813	0·225	0·781	0·525	0·475	0·600	0·640	0·529	0·600	0·550	0·516
1898–9	1·350	0·575	0·475	0·288	0·225	0·238	0·950	0·525	0·350	0·313	0·475	0·781	0·443
1899–1900	0·500	0·275	0·525	0·900	0·263	0·250	0·550	0·700	0·650	0·333	0·950	0·400	0·431
1900–1	0·800	0·288	0·625	0·675	0·575	0·400	0·275	0·263	0·375	0·575	0·750	0·525	0·498
1901–2	0·625	0·625	0·550	0·225	0·383	0·867	0·475	1·500	0·625	0·550	0·575	0·675	0·571
1902–3	1·500	0·525	0·675	0·475	0·400	0·500	0·312	0·450	0·541	0·417	0·275	0·313	0·447
1903–4	0·500	0·263	0·325	0·600	0·375	0·300	0·575	0·625	0·625	0·750	0·417	0·600	0·424
1904–5	0·719	0·475	0·313	0·400	0·263	0·600	0·250	0·750	0·900	0·350	0·821	0·350	0·460

APPENDIX—TABLE IV.

NITROGEN AS NITRATES IN RAIN-WATER COLLECTED AT ROTHAMSTED IN PARTS PER MILLION.

	Sept.	Oct.	Nov.	Dec.	Jan.	Feb.	March	April	May	June	July	Aug	Year
1854–5	—	—	—	—	0·06	0·16	0·08	0·13	0·13	0·30	0·06	0·22	—
1855–6	0·08	0·13	0·07	0·06	0·09	0·07	0·13	0·07	0·10	0·17	0·13	0·13	0·10
1856–7	0·13	0·12	0·16	0·15	—	—	—	—	—	—	—	—	—
1886–7	0·270	0·089	0·064	0·048	0·172	—	0·247	0·106	0·162	0·284	0·204	0·385	0·138
1887–8	0·093	0·090	0·093	0·066	0·198	0·229	0·095	0·145	0·109	0·167	0·104	0·090	0·116
1888–9	0·253	0·173	0·096	0·155	0·190	0·095	0·136	0·230	0·100	0·184	0·120	0·096	0·134
1889–90	0·150	0·083	0·180	0·127	0·097	0·220	0·093	0·220	0·185	0·104	0·083	0·123	0·119
1890–1	0·330	0·213	0·120	0·773	0·120	2·160	0·200	0·200	0·130	0·246	0·200	0·140	0·195
1891–2	0·240	0·192	0·213	0·080	0·213	0·240	0·306	0·300	0·213	0·160	0·200	0·147	0·185
1892–3	0·187	0·120	0·420	0·098	0·288	0·147	0·640		0·347	0·340	0·173	0·187	0·217
1893–4	0·160	0·088	0·090	0·090	0·114	0·150	0·093	0·240	0·173	0·147	0·160	0·160	0·131
1894–5	0·267	0·128	0·072	0·140	0·088	0·667	0·420	0·260	0·500	0·380	0·100	0·120	0·161
1895–6	0·320	0·200	0·142	0·222	0·260	0·420	0·170		0·540	0·220	0·240	0·267	0·216
1896–7	0·090	0·080	0·170	0·200	0·130	0·290	0·210	0·147	0·187	0·173	0·460	0·150	0·160
1897–8	0·166	0·286	0·293	0·120	0·300	0·173	0·333	0·320	0·270	0·200	0·293	0·320	0·234
1898–9	0·720	0·240	0·251	0·120	0·130	0·150	0·427	0·227	0·180	0·285	0·267	0·460	0·228
1899–1900	0·227	0·190	0·200	0·620	0·160	0·150	0·290	0·213	0·240	0·093	0·360	0·128	0·200
1900–1	0·580	0·200	0·187	0·220	0·293	0·320	0·187	0·160	0·280	0·320	0·347	0·267	0·250
1901–2	0·267	0·251	0·270	0·200	0·160	0·336	0·176	0·320	0·206	0·500	0·274	0·200	0·267
1902–3	0·560	0·272	0·304	0·375	0·225	0·300	0·125	0·250	0·180	0·168	0·116	0·135	0·203
1903–4	0·183	0·133	0·263	0·288	0·213	0·133	0·213	0·313	0·450	0·317	0·175	0·275	0·214
1904–5	0·325	0·325	0·213	0·142	0·133	0·250	0·125	0·225	0·375	0·125	0·313	0·163	0·197

APPENDIX—TABLE V.

CHLORINE IN RAIN-WATER COLLECTED AT ROTHAMSTED IN PARTS PER MILLION.

	Sept.	Oct.	Nov.	Dec.	Jan.	Feb.	Mar.	April	May	June	July	Aug.	Year
1876-7	—	—	—	—	—	—	—	—	—	1·95	0·24	0·95	—
1877-8	1·73	3·40	1·97	1·96	2·91	0·50	4·00	0·55	0·91	1·48	4·31	1·16	1·62
1878-9	2·28	2·58	1·83	3·00	3·04	1·83	5·80	1·67	1·40	0·80	0·80	0·85	1·69
1879-80	1·05	2·65	9·38	5·75	3·20	3·20	2·90	1·73	3·43	2·47	0·64	1·30	2·25
1880-1	0·97	3·00	2·95	1·70	10·00	3·45	1·95	3·20	2·38	1·63	0·67	0·53	2·17
1881-2	0·75	4·20	2·70	1·80	1·70	2·50	3·75	2·60	1·60	0·80	1·60	1·68	2·13
1882-3	1·15	2·28	4·30	1·60	3·00	2·25	8·85	1·60	1·40	1·12	0·73	2·00	2·23
1883-4	1·90	2·50	2·30	8·80	3·60	5·75	1·65	1·65	3·70	0·80	0·62	0·90	2·43
1884-5	0·85	2·25	3·33	3·30	3·42	2·75	2·35	1·45	1·50	0·75	1·45	2·27	2·20
1885-6	1·00	1·90	1·57	3·43	2·58	2·25	3·60	2·77	0·93	0·98	0·50	1·45	1·73
1886-7	2·10	2·95	1·65	4·63	2·28	3·50	3·28	4·75	2·15	2·00	2·90	2·70	2·99
1887-8	2·70	3·35	2·95	2·73	4·90	11·05	3·40	2·20	2·50	0·75	1·33	1·03	2·46
1888-9	2·30	2·15	2·37	3·43	3·05	3·90	2·25	2·55	0·40	2·17	0·77	1·42	1·85
1889-90	1·30	1·83	2·40	2·10	3·05	3·95	1·60	2·93	1·15	1·15	0·43	1·20	1·66
1890-1	1·90	2·00	1·95	8·05	2·50	7·50	3·50	2·50	1·30	1·25	1·45	1·20	1·99
1891-2	2 00	2·71	2·75	2·75	4·08	3·45	4·20	2·60	2·40	1·20	1·00	1·13	2·31
1892-3	1·95	1·75	1·82	1·08	3·20	2·85	4·95		2·15	2·32	1·30	1·25	2·05
1893-4	3·04	1·68	4·44	6·80	4·62	2·95	2·87	2·05	2·50	1·20	1·00	0·50	2·71
1894-5	1·90	2·25	2·10	4·06	4·90	4·02	3·38	1·44	2·80	3·00	1·00	1·20	2·25
1895-6	1·40	2·04	2·80	4·20	3·18	3·86	2·22		4·10	1·89	1·94	1·13	2·42
1896-7	1·25	2·52	3·39	2·28	8·61	1·82	3·48	2·94	2·10	1·55	3·92	1·74	2·51
1897-8	1·70	4·20	4·98	3·81	3·59	4·47	20·10	2·30	2·25	2·04	1·25	1·88	3·74
1898-9	5·63	2·51	2·85	2·78	5·88	4·80	5·33	2·42	2·40	1·50	1·37	2·55	3·24
1899-1900	2·22	1·67	2·75	4·70	2·22	1·88	5·93	3·68	2·25	1·26	2·09	1·28	2·25
1900-1	3·69	2·34	3·83	3·45	7·13	3·96	1·43	2·70	5·03	2·90	1·40	1·43	3·02
1901-2	2·43	4·44	5·49	2·85	3·00	6·38	2·04	3·80	3·08	1·35	1·83	1·53	2·81
1902-3	3·05	3·56	4·58	5·49	4·02	5·48	3·00	3·90	1·74	1·02	0·80	1·68	2·52
1903-4	1·95	2·07	2·42	5·01	3·15	4·65	4·62	3·18	2·45	2·65	0·81	1·77	2·76
1904-5	1·98	2·25	1·92	2·33	3·14	11·60	3·68	3·54	2·90	1·49	0·94	1·50	2·66