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THE AMOUNT AND COMPOSITION OF RAIN FALLING AT ROTHAMSTED.

(BASED ON ANALYSES MADE BY THE LATE NORMAN H. J. MILLER.)

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THE composition of rainwater was a matter of serious interest to the agricultural chemists of the last generation. It was a side issue of the great controversy on the source of nitrogen for plant life, which occupied much time and energy between 1840 and 1870. The main issue was whether plants could or could not assimilate free nitrogen from the air; this was settled in the case of non-leguminous crops by the investigations of Lawes, Gilbert and Pugh in 1861[1]. The side issue was whether the nitrate and ammonia necessary for vegetation would need to be supplied by fertilisers or whether the natural stores in the rain and the air would suffice.

Liebig had stated[2]:

"If the mineral elements, phosphates, etc., be duly supplied, the plant will obtain a sufficient supply of ammonia from the atmosphere": and again:

"If the soil be suitable, if it contains a sufficient quantity of alkalis, phosphates and sulphates, nothing will be wanting. The plants will derive their ammonia from the atmosphere as they do carbonic acid[3]."

Liebig clearly supposed that there was a considerable amount of nitrogen in the rain, and while he does not seem to have committed himself to any figure in his earlier writings, he published in 1863[4] an estimate of 24 lb. of nitrogen per acre per annum.

Lawes and Gilbert did not accept this position. They showed by field experiments that the crop yield is proportional to the ammonia

[1] *Phil. Trans.*, 1861, Part II, 431.

[2] *Letters on Chemistry*, 1851, 3rd ed., 519. In this Letter, the 34th, Liebig sets out his views with characteristic clearness.

[3] *Farmers' Magazine*, 1847, **16**, 511.

[4] *Natural Laws of Husbandry*, 1863, 290.

supplied in the manure[1]. Further, they analysed the rain falling at Rothamsted, and their results indicated that it supplied only about 5 lb. per acre of ammonia[2], a quantity far below the 50 lb. of nitrogen needed by a 32 bushel crop of wheat. Their analytical procedure was extremely laborious, the Nessler test not then having been devised; in some experiments it even involved the distillation of over two hundred-weights of rain, and evaporation of the distillate with sulphuric acid. Lawes and Gilbert had not been the first in the field; they acknowledged their indebtedness to Boussingault who, working on his experimental farm at Bechelbronn in Alsace, with simpler methods and much smaller quantities of rain, had obtained results very similar to their own[3].

Lawes and Gilbert were unable to make satisfactory determinations of the nitric nitrogen in the rain although they recognised its presence: further analyses were therefore made in the two following years, 1855 and 1856, but the work was not done in the Rothamsted laboratories, Gilbert being too fully occupied with the field plots; it was carried out by J. T. Way, then a promising young agricultural chemist. His figures also were far lower than the crop requirements, being in 1855 only 6·5 lb. per acre, and in 1856 8·0 lb. per acre for the sum of ammonia[4] and nitrate. The regular determinations were then discontinued, but numbers of occasional analyses were made, first by Edward Frankland and afterwards by Warington. These results were quite consistent with the measurements of 1855 and 1856, and lent no support to Liebig's view.

The subject would perhaps have been allowed to drop, but for the circumstance that Lawes and Gilbert in 1870 erected the famous drain gauges at Rothamsted and proceeded to make determinations of the amount of nitrate and ammonia percolating through them. This necessitated systematic analyses of the whole of the rainfall and of the drainage water over a period of years. Lawes and Gilbert were fortunate in the men to whom they entrusted the undertaking. The analytical work was done between 1877 and 1885 by the late Robert Warington, and from January, 1888, to 1916 by the late N. H. J. Miller, while the

[1] For their side of this controversy see J. B. Lawes and J. H. Gilbert, "On Agricultural Chemistry," *Journ. Roy. Agric. Soc.*, 1851, **12**, 1.

[2] *British Assoc. Reports*, 1854. As will be shown later on, even this figure is twice what it should be.

[3] Now that our French friends have recovered Bechelbronn we hope it will be found possible to commemorate in some adequate manner the important work carried out on this farm by Boussingault in laying the foundations of modern agricultural chemistry.

[4] *Journ. Roy. Agric. Soc.*, 1856, **17**, 123, 618.

collection of samples at the gauges was done by E. Grey, who happily is still continuing the same work. Warington and Miller differed widely in personal characteristics but they were equally reliable analysts and equally undeterred by the monotony of the routine involved in the systematic work. Miller indeed made it his life-work, and only his sudden and unexpected death in January, 1917, prevented the completion of an important monograph which he had in preparation on the composition of rain and drainage waters.

Miller's results show that the data of 1855 and 1856, which had been wholly inadequate to give any support to Liebig's view, were probably in excess of the actual facts; the ammoniacal nitrogen apparently having been over estimated and given as 6 or 7 lb. per acre instead of 2·5 lb. to 3 lb.; during Miller's period the sum of the ammoniacal and nitric nitrogen was only 4 lb. per acre,—an amount still less adequate to supply the needs of crops.

Miller published his first paper in 1905[1], but since then a further ten years' results have accumulated which it is proposed to discuss here. The work has now been modified: no useful agricultural purpose would be served by continuing it in its original form, and its interest now lies in its relationship to atmospheric pollution. For the Rothamsted rain is collected in a part of the country which is fairly free from sources of impurity, and it may be taken as typical of "pure" rain: it thus affords a basis for estimating the amount of pollution. From this point of view there is the possibility of a useful continuation of the work.

The amount and distribution of the rainfall at Rothamsted.

The rainfall records at Rothamsted extend continuously over a period of 66 years, the results being

	Sept.	Oct.	Nov.	Dec.	Jan.	Feb.	
Average for 60 years (Sept. 1853–Aug. 1912)	2·37	3·18	2·58	2·47	2·34	1·81	
Average for 28 years (Sept. 1888–Aug. 1916)	1·87	3·23	2·64	3·05	2·15	2·02	
	March	**April**	**May**	**June**	**July**	**Aug.**	**Total**
Average for 60 years (Sept. 1853–Aug. 1912)	1·92	1·84	2·19	2·45	2·50	2·69	28·34
Average for 28 years (Sept. 1888–Aug. 1916)	2·41	1·69	1·99	2·53	2·41	2·83	28·82

The average for the year over the whole period is 28·3 inches. It will be shown later, however (p. 324), that the rainfall tends to increase:

[1] This *Journal*, 1905, **1**, 280–303.

for the last 14 years the average for the year has been 29·98 inches, and but for three abnormally dry years (1905–6, 1908–9, 1913–14) would have been well over 30 inches. Rothamsted lies in the rather narrow wet strip that runs in a north-easterly direction across the eastern counties, and separates the dry region, including the Thames Valley, South Middlesex, East Berkshire and East Oxfordshire on the south, from another dry tract comprising Bedfordshire, Cambridgeshire, Huntingdon, East Northamptonshire, etc., on the north.

The distribution calls for little comment. The four wettest months are July, August, October and November, and the four driest are February[1], March, April and June: allowing for the varying number of days, April is the driest month of all; this is usual over one-half the area of the British Isles[2]. Further details are given in Table 1, p. 331.

The amount and distribution of nitrogen compounds in the rain.

The amounts of ammoniacal and nitric nitrogen present in the rain are given in Table 2 (p. 332): in lbs. per acre they are

	Sept.	Oct.	Nov.	Dec.	Jan.	Feb.
Nitrogen as Ammonia (NH_3)	0·217	0·235	0·219	0·223	0·182	0·159
Nitrogen as Nitrate...	0·103	0·161	0·111	0·115	0·087	0·088
Sum ...	0·320	0·396	0·330	0·338	0·269	0·247
Rainfall ...	1·87	3·23	2·64	3·05	2·15	2·02

	March	April	May	June	July	Aug.	Total
Nitrogen as Ammonia (NH_3)	0·205	0·201	0·229	0·245	0·249	0·280	2·644
Nitrogen as Nitrate...	0·098	0·094	0·114	0·114	0·111	0·131	1·327
Sum ...	0·303	0·295	0·343	0·359	0·360	0·411	3·971
Rainfall ...	2·41	1·69	1·99	2·53	2·41	2·83	28·82

The yearly fluctuations in ammoniacal nitrogen are shown in Fig. 1: they are seen to follow the rainfall very closely with only four exceptions: the year 1893–4 when the ammonia fell although the rainfall rose, and 1895–6, 1901–2 and 1908–9, when the ammonia rose although there was less rain. Fig. 2 shows the fluctuations month by month; these also follow the rainfall. Tables 3 and 4 (pp. 333–4) give fuller details.

[1] February is one of the driest months in the whole year so far as actual rainfall is concerned, but there is a popular tradition which is still carried on by popular writers, and which no amount of statistical data is able to break, that it is a wet month. Probably the reason for the tradition is the old proverb "February fill dyke."

[2] H. R. Mill and C. Salter, *J. Roy. Met. Soc.*, 1915, **41**, 14.

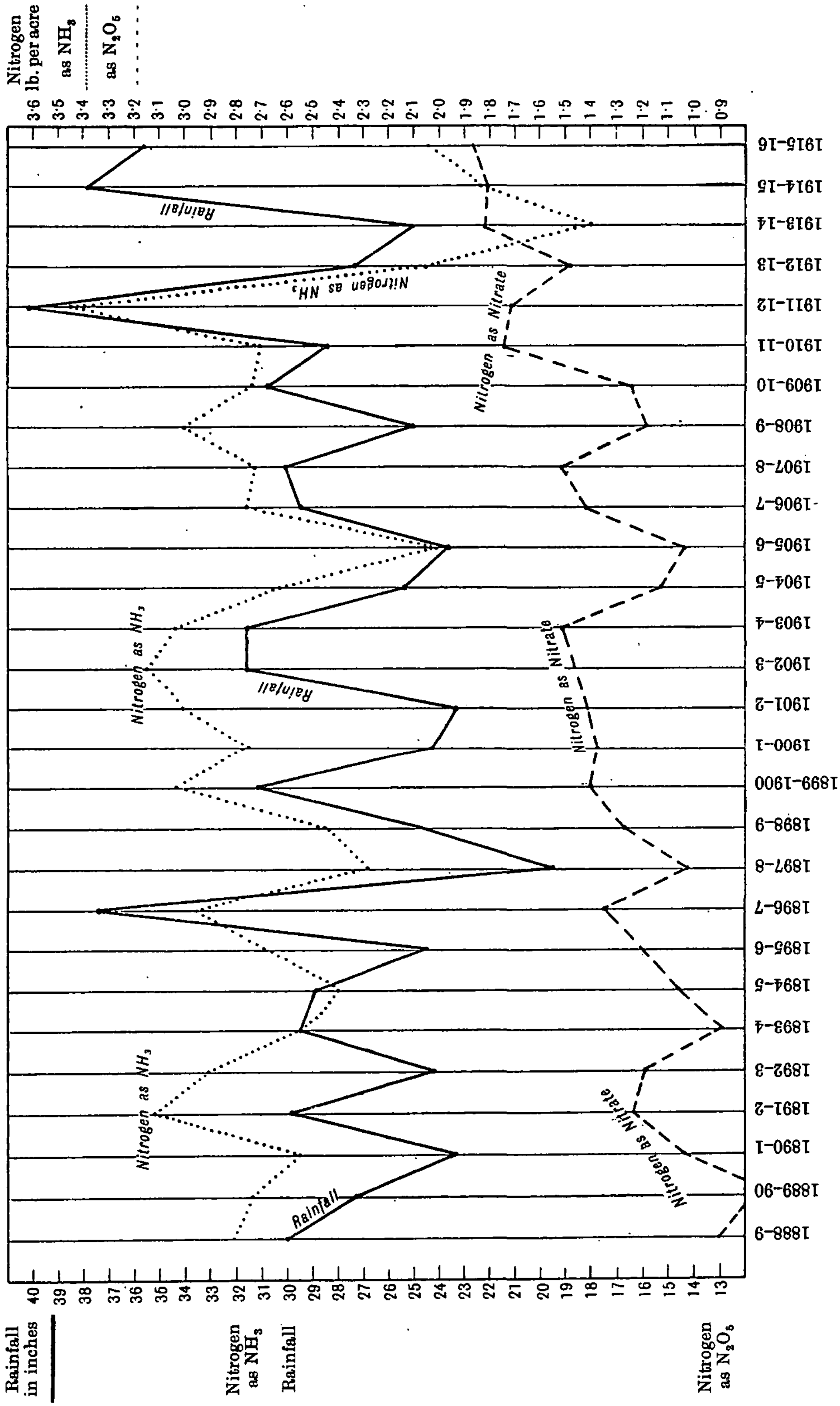


Fig. 1. Annual fluctuations in amounts of ammoniacal and nitric nitrogen in rainwater, 1888-1916.

Fig. 2, however, brings out the interesting point that the ammonia content of the rain is highest during May, June, July and August, and lowest during January, February, March and April.

The significance of this close relationship between rainfall and ammonia content is discussed later.

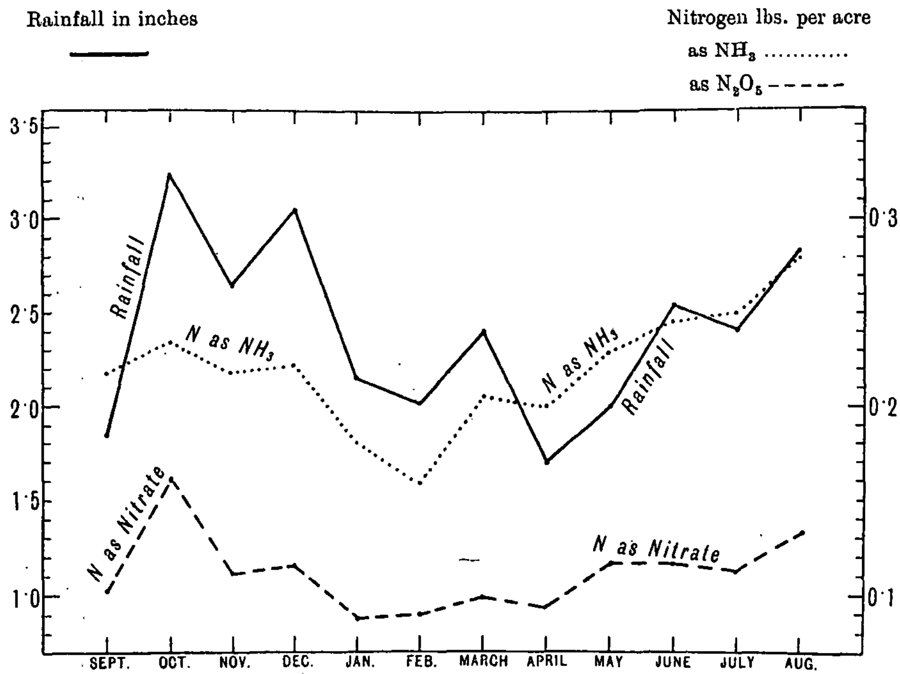


Fig. 2. Monthly fluctuations in amounts of ammoniacal and nitric nitrogen in rainwater, Sept. 1, 1888–Aug. 31, 1916.

The amount of nitrate in the rain.

The quantity of nitrate in the rain is shown in Tables 2, 3 and 5 (pp. 332–4); over the whole period it is half that of ammonia and amounts on an average to about one-tenth of a pound per acre per month (0·11 lb. per acre to be exact), but it varies less than the ammonia. In the Southern Hemisphere the proportions are reversed: at Lincoln, New Zealand, the ammonia is much smaller in amount than the nitrate[1], and at Melbourne the nitrate is high in amount and fluctuates[2]; in the Tropics also there is a considerable excess of nitrate over ammonia[3].

[1] G. Gray, *Proc. Austral. Assoc.*, Sydney, 1888.

[2] V. G. Anderson, *Journ. Roy. Meteorol. Soc.*, 1915, **41**, 99.

[3] J. B. Harrison and J. Williams, *Journ. Amer. Chem. Soc.*, 1897, **19**, 1.

Anderson[1] has argued that the values for nitric nitrogen are a function of the weather type which they may therefore help to characterise.

At Rothamsted the rain falling during the months May, June, July, August and October is richer in nitrate than that falling in January, February, March and April. This is similar to the distribution of ammonia. Examination of the curves of Fig. 1 shows that the nitric nitrogen fluctuated with the rainfall in much the same way as the ammoniacal nitrogen until 1910, but since then there has been no simple relationship. The monthly fluctuations shown in Fig. 2, also follow the rainfall and the ammonia.

For most of the period the relationship between ammoniacal and nitric nitrogen has been close and was almost exactly 2 : 1. Since 1912, however, the nitric nitrogen has increased so that on the four year average the nitric nitrogen is equal to the ammoniacal nitrogen, instead of being only half as much. This is not the result of a general rise throughout the whole of the year, but of a few exceptionally high values in the months February, March, July, August and October. During this period it happened that the rainfall in February and March has been unusually high; there was also one very wet July.

Ammoniacal and nitric nitrogen in rain: four year periods.

Four years' period {Sept. / Oct.	1888–1892	1892–1896	1896–1900	1900–1904	1904–1908	1908–1912	1912–1916
Nitric nitrogen ...	0·98	1·08	1·26	1·43	1·28	1·48	1·75
Ammoniacal nitrogen	2·82	2·63	2·68	2·98	2·54	2·97	1·84
Sum ...	3·80	3·71	3·94	4·41	3·82	4·45	3·59
Ratio: $\frac{\text{Ammoniacal}}{\text{Nitric}}$ nitrogen	3 : 1	5 : 2	2 : 1	2 : 1	2 : 1	2 : 1	1 : 1

The increase is presumably due to some artificial cause, though it is not easy to say what. During the period 1886 to 1908, which saw an enormous expansion of London into the Home Counties and a pushing out both of industrial concerns and of private residences, there was only a small rise in nitrate and none in ammonia. It is possible that the changes in stoves and gas-burners, which had become marked by 1912, and which have reduced the frequency of London fogs, have also tended to increase the nitrous fumes rather than the ammonia in the atmosphere.

The rain coming from the Atlantic contains much less nitrate than that falling in inland stations (see p. 317).

[1] V. G. Anderson, *Journ. Roy. Meteorol. Soc.*, 1915, **41**, 99.

Nitrites in rainwater.

The amount of nitrite in the Rothamsted rain has not been separately estimated: it is included in the nitrate figure. The rain from the Hebrides was frequently tested during the winter of 1911–12 and more frequently gave negative than positive results. Rain collected on land, however, usually contains nitrites and a long series of determinations made before the War at Prince Troubetzkoy's Experiment Station at Ploty in South Russia showed a considerable excess of nitrites in winter compared with summer rains.

The total nitrogen in rainwater.

No further determinations of organic nitrogen have been made beyond those recorded in Dr Miller's paper. The figure there given is 1·35 lb. per acre per annum; assuming this to be still correct on an average of the whole period the rain contained

2·64 lb. ammoniacal nitrogen,
1·33 ,, nitric nitrogen,
1·35 ,, organic nitrogen.

The ratios are almost exactly

2 parts ammoniacal nitrogen,
1 part nitric nitrogen,
1 part organic nitrogen.

But, as already pointed out, the proportions between ammoniacal and nitric nitrogen have fallen recently.

The sources of the combined nitrogen in rain.

Sources of ammonia. Certain indications as to the source of the ammoniacal nitric nitrogen in the air are afforded by their close relationship with the amount of rain: when the rainfall is high the ammonia is high, and conversely when the rainfall is low the ammonia is low also. There appear to be two possible sources: the ammonia may be brought into the district with the rain, or it may be already present in the atmosphere and dissolved out by the rain: in the latter case a constant renewal of the atmospheric supply has to be assumed, otherwise the first rain would wash out all that was present, leaving nothing to be removed later.

If the ammonia came with the rain the whole phenomena would be readily explicable: the more the rain the greater the amount of ammonia.

Since the larger part of the rain comes from the Atlantic it would follow that most of the ammonia would come from there also.

This view was held by Boussingault and developed by Schloesing[1] in an important series of papers. It has been subjected to a critical examination at Rothamsted.

Arrangements were made for the systematic collection of samples of rain at various stations (usually lighthouses) in the Hebrides and in Iceland, remote from atmospheric pollution. These samples were then sent to Rothamsted for analysis.

The results were[2]

		Nitrogen				
		Per million		Lbs. per acre per annum		
	Rainfall Inches	As Ammonia Average	As Nitrates Average	As Ammonia	As Nitrates	Sum
Rothamsted	28·04	0·437	0·202	2·774	1·251	4·025
Laudale, Ardgour ...	88·80	0·138	0·063	2·784	1·260	4·044
Barrahead, Berneray ...	35·28	0·145	0·138	1·164	1·104	2·268
Shilay Monach Islands, N. Uist.	48·36	0·115	0·054	1·260	0·588	1·848
Butt of Lewis, Stornoway	41·19	0·039	0·033	0·361	0·305	0·666
Vifilsstadir, Iceland ...	38·34	0·091	0·030	0·802	0·263	1·065

Much less ammonia was found than at inland stations and it was not always certain that the sample collected was as pure as the rain. In spite of serious efforts there was considerable difficulty at the lighthouses in keeping the rain gauges free from bird droppings, and it is possible that some of the ammonia came from adventitious contamination.

No analyses, so far as we know, have been made of rainwater collected on the Atlantic itself. One of us (E. J. R.) made several attempts to secure good samples when crossing and recrossing in 1909 and again in 1912, but without success. But even if the atmosphere over the Atlantic is not entirely free from ammonia, it certainly contains considerably less than that over the land. We therefore cannot agree with Schloesing that the bulk of the ammonia comes from the sea, though possibly part of it does.

It seems necessary therefore to suppose that some, if not all, of the ammonia is derived from the atmosphere. Owing to its high solubility

[1] *Compt. Rend.*, 1875, **80**, 175. "Sur l'ammoniaque de l'atmosphère." *Ibid.*, **81**, 81 and 1252; 1876, **82**, 747, 846 and 969.

[2] N. H. J. Miller, *Journ. Scottish Meteorol. Soc.*, 1913, iii, **16**, 141.

the ammonia may be expected to dissolve freely, in which case constant renewal would be necessary in order to account for the fact that high rainfall brings down more ammonia than low rainfall.

This constant renewal necessitates contact with a source of ammonia which, moreover, gives up its ammonia uniformly so that a definite equilibrium is attained. As soon as a shower of rain has fallen and removed the ammonia from the atmosphere a further supply must be drawn from the source sufficient to restore the disturbed equilibrium. The next shower removes some of this, but again the equilibrium is restored. On this assumption the total quantity brought down by the rain in any year would depend on the amount of the rainfall, but the quantity per inch of rain would show less variation.

Three possible sources have been considered: the sea, which has already been discussed, the soil, and the atmosphere over large towns and cities.

The soil seems quite a likely source: changes are constantly occurring there with formation of ammonia which is then transformed to nitrate. The amount of ammoniacal nitrogen existing as such at any time is at least 5 lb. per acre in the top 9 in., and its rate of diffusion into the atmosphere might be fairly rapid.

Hall and Miller[1] endeavoured to obtain information as to this possibility by exposing shallow dishes of sulphuric acid, some close to the ground and others four-feet above it, and then determining the ammonia absorbed by the end of each month: the experiment lasted for two years. The amounts absorbed corresponded only to 0·99 and 1·28 lb. per acre in the respective years,—much less than those recorded by previous observers, perhaps because of the efficiency with which dust and insects were excluded: the lower dishes, however, did not usually contain more ammonia than the upper ones, excepting when sulphate of ammonia was applied to the soil. No conclusions could be drawn as to whether the soil normally gives up ammonia to the atmosphere, or whether it absorbs ammonia from the atmosphere. From the circumstance that soil gives up ammonia when dressed with sulphate of ammonia it seems legitimate to infer that some ammonia is continuously being evolved.

The seasonal fluctuations in the amount of ammonia in the rain are quite consistent with the view that the ammonia comes from the soil. The amount of ammoniacal nitrogen in the rain is lowest in the four months, January, February, March and April, when biochemical

[1] This *Journal*, 1911, **4**, 46.

activity in the soil is at a minimum; and highest in June, July, August and October, when biochemical activity in the soil is at a maximum.

The other source with which the atmosphere may be in equilibrium is the atmosphere of cities, where the amount of ammonia is markedly greater than in the country. The amount of ammonia brought down by the rain of certain towns is as follows, the Rothamsted and Malvern figures for the same periods being given for purposes of comparison:

Nitrogen as ammonia in lbs. per acre per annum[1].

Average of four years, 1914–18	Summer	Winter	Total
London (Embankment Gardens)	5·22	5·04	10·26
Newcastle-on-Tyne	4·92	6·06	10·98
Malvern	1·14	0·48	1·62
Rothamsted (1912–1916) ...	1·03	0·81	1·84

If the whole of the ammonia were carried down in the rain there would of course be none to travel out into the country, but as pollution is constantly going on there may be an excess during the intervening dry periods. It is not certain, however, that atmospheric pollution would be sufficient by itself to account for all the ammonia present in the rain. The value obtained at Rothamsted is not likely to be higher, indeed it is probably lower, than the average over Great Britain: assuming only 2·6 lb. per acre the total amount of ammonia brought down on the 56,000,000 acres of Great Britain would be 65,000 tons of combined nitrogen, equivalent to 325,000 tons of sulphate of ammonia per annum[2]. The total coal consumption of the country is approximately 200,000,000 tons per annum: if all this were handled at gas works or in coke ovens it would yield some 2,000,000 tons of sulphate of ammonia, assuming 22·7 lb. sulphate of ammonia were obtained from each ton of coal. But the open grate is far less efficient as a producer of ammonia than the gas retort, and nothing approaching this quantity is likely to arise. Of the amount actually formed no less than 400,000 tons is collected from gas and recovery plants, and a further large quantity is absorbed in soot. The amount discharged into the atmosphere is the difference between these quantities and the total production: it is impossible to make a satisfactory estimate, but there seems hardly enough left to furnish even the 325,000 tons of sulphate of ammonia,

[1] Taken from reports of Advisory Committee on Atmospheric Pollution.

[2] This figure is rather interesting: it is nearly 900 tons per day, and is considerably more than the whole of the artificial nitrogenous fertilisers used by farmers in the United Kingdom.

which, as we have seen, is probably a minimum value for the total brought down in the rain.

There is a further difficulty in the assumption that atmospheric pollution is the main source of the ammoniacal nitrogen of country air. Atmospheric pollution of cities is as bad in winter as in summer if not worse, so that if it were the source of most of the ammonia in country rain we should expect at least as much in winter as in summer: in point of fact there is less. And, moreover, so far as Rothamsted is concerned, the only town of any size to the south-west is Reading, but as this is more than forty miles away, it is hardly likely to serve as a sufficient reservoir.

The formation of nitrates or nitrous fumes in the air is commonly attributed to the major electrical discharges,—thunderstorms, etc. Berthelot has shown that the silent electric discharge causes a production of nitric acid from moist nitrogen and oxygen[1]. There is also the possibility that the electrical stresses in the atmosphere may have some effect; Chree[2] estimates these at about 200 to 300 volts per metre at ground level, and much more at higher levels: at the top of tall trees there may be 5000 volts per metre. The potential gradient required to make a spark pass in ordinary air at normal pressure is of the order of 30,000 volts per centimetre.

It has further been suggested by Soddy[3] that nitrous oxide may be formed by the action of the radium emanations always present in the lower portions of the atmosphere.

There are, however, other possible sources. Dust invariably contains nitrates, and in summer the atmosphere contains more dust than in the winter: the rain as we have seen also contains more nitrate. Gas flames and fires also produce nitrous fumes.

The close relationship between the amounts of ammoniacal nitrogen and of nitric nitrogen in the rain throws important light on the origin of the nitric compound. It must either be formed from the ammonia or come from the same source. It is possible, but not easy, to conceive of nitric oxides or acid compounds coming from the sea; it is not difficult to conceive of such compounds coming from the soil and from the air of cities. Formation from ammonia would present no serious difficulty: nitric compounds might arise from the oxidation of ammonia under the influence of the minor electrical disturbances and electrical stresses in the

[1] *Compt. Rend.*, 1906, **142**, 1367.
[2] *Journ. Roy. Meteorol. Soc.*, 1915, **41**, 121.
[3] *Chemical Soc. Reports*, 1911, **8**, 299.

atmosphere. It is known[1] that this oxidation proceeds under the action of the silent electric discharge, and it may well form part of the normal atmospheric phenomena.

In reviewing the evidence for and against these various possibilities one is driven to conclude that the ammonia in country rain probably arises from several sources and not from one only. The sea, the soil, and towns and cities may each contribute their share. Of these the soil appears to be the most important in view of the fact that the amount brought down by the rain is at a maximum in the summer months when biochemical activity in the soil is at its highest, and at a minimum in winter when biochemical activity in the soil is low. This does not amount to proof of the origin of the ammonia because the higher ammoniacal content of summer rain is affected by another factor: the rain during the summer months is probably on the whole formed at higher levels than that falling during winter months, and, having a greater distance to travel, it would wash out the ammonia from a larger quantity of air than is possible in the case of winter rain. A further effect is discussed on p. 328.

A certain amount of atmospheric ammonia may also come from the towns: the reality of this source is indicated by the fact that town rain-water is considerably richer in ammonia than country rainwater. This is unlikely to be the chief source, as the ammonia in town rain is high in winter, whereas we have seen it is then at its lowest in country rain.

The nitric nitrogen also probably arises from several sources. In the early years of the work the amount of nitric nitrogen recorded was only one-third that of the ammoniacal nitrogen, but for the greater part of the period of the observations it was one-half. Of late years there have been some exceptionally high values, causing the nitric nitrogen to become equal to the ammoniacal nitrogen. These changes indicate an artificial rather than a natural origin for part of the nitrate. But it cannot all arise in this way, for it is widely distributed over the world. Some of it may arise by direct combination of nitrogen and oxygen during lightning and other major electrical disturbances. For the greater part the close correlation between the amounts of nitric nitrogen and of ammoniacal nitrogen indicates either a common origin, or a formation of nitric compounds from ammonia by the minor electrical disturbances and electrical stresses normally occurring in the atmosphere, or in some other way.

[1] See W. G. Mixter, *Amer. Journ. Sci.*, 1898 [iv], **6**, 217–224.

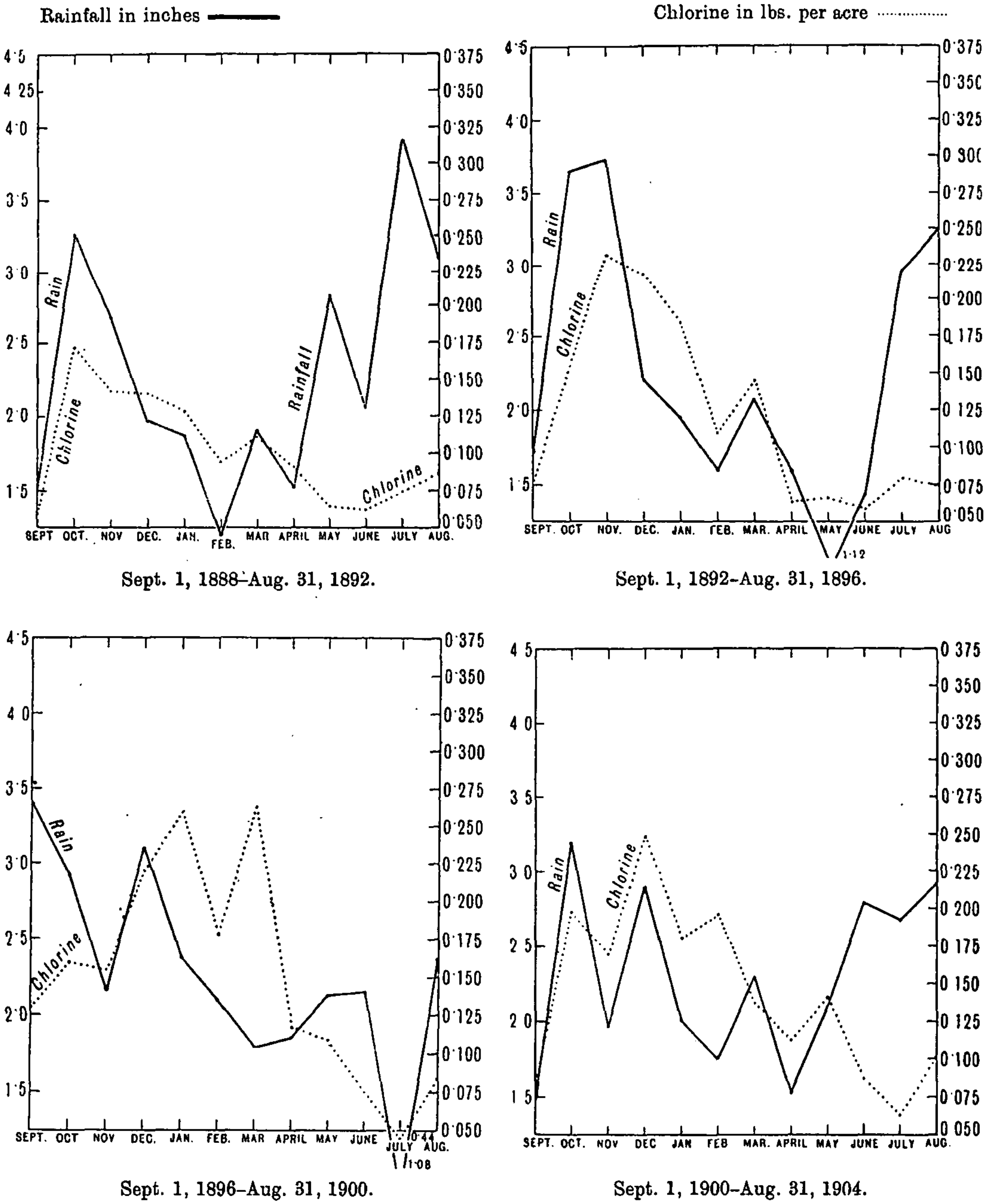
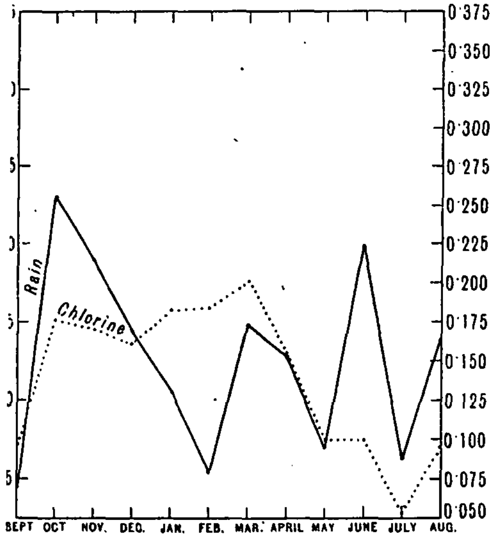
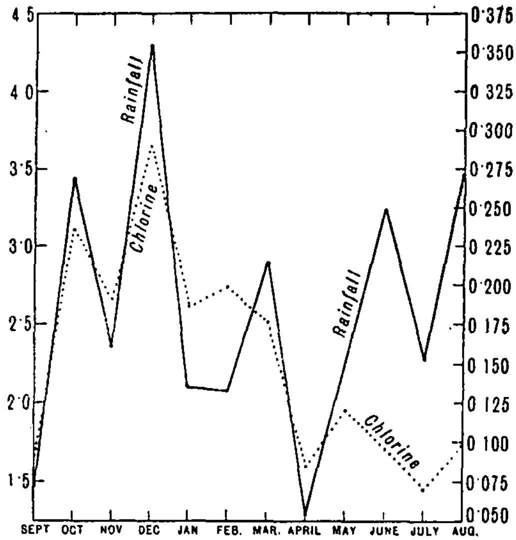


Fig. 3. Monthly fluctuations in amounts of chlorine in rainwater: four year periods between Sept. 1, 1888 and Aug. 31, 1916.

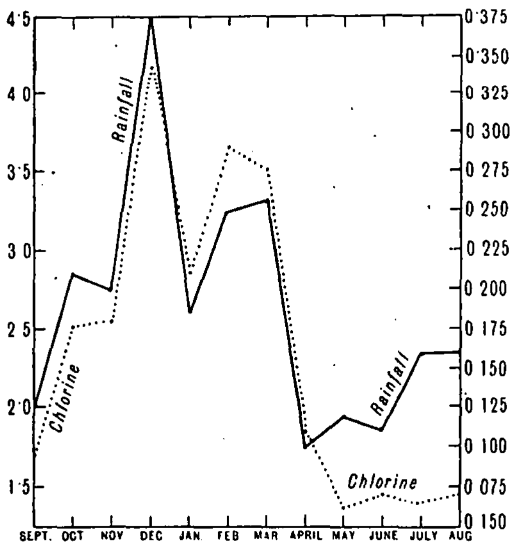
Rainfall in inches ——— Chlorine in lbs. per acre ....



Sept. 1, 1904–Aug. 31, 1908.



Sept. 1, 1908–Aug. 31, 1912.



Sept. 1, 1912–Aug. 31, 1916.

Fig. 3 (*cont.*).

The amount of chlorine in rainwater.

The chlorine is determined in the rainwater at Rothamsted because the values are wanted in connection with the drainage investigations. The data are given in Tables 6 and 7 (pp. 335–6): they throw interesting light on the origin of the rain.

The average amount of chlorine in the Rothamsted rainwater is 2·3 parts per million, bringing down 16 lb. per acre per annum, but the amount fluctuates considerably from year to year, the lowest being 10·3 lb. in 1889–90, and the highest 24·4 lb. in 1915–16. Most of this is brought down during the months September to April: a much smaller amount falls in the summer months. The fluctuations in the amount of chlorine carried down per acre closely follow the fluctuations in the amount of rainfall, especially in the months September to April: during the summer months, May to August, the rain contains so little chlorine that the fluctuations have less significance. This is well brought out by the curves (Fig. 3); only in two periods 1896–1900 and 1904–8, is there notable deviation from the close relationship in the winter months.

It has been usual to attribute the chlorine to sea spray blown over the land. This view is consistent with the facts, and would explain the large amount of chlorine in winter rain and the small amount in summer rain when gales are less common.

Another source exists, however. Some of the chlorine may come from fires: it is present in coal, which contains about 4 lb. per ton: it would be liberated during combustion in the gaseous form or as volatile chlorides. This source would be most in evidence during the winter.

The change in composition of Rothamsted rainwater.

Looking over the figures for successive four year periods given in Table 9 (p. 337) there seem to be signs of change in the composition of rain. The chlorine is increasing in amount, so also is the nitrate; the ammonia tends to fall, so that there is no steady alteration in the sum of the ammoniacal and nitric nitrogen. It is more easy to suggest possible explanations than to decide between them. The rainfall has increased since 1908, and this tends to increase the quantity of substances brought down per acre. The increase in rainfall has been especially marked in the winter months: we have no wind records at Rothamsted, but it is possible that the higher winter rainfall has been accompanied by heavier gales carrying more chlorine from sea spray. The increase in amount of chlorine is not accidental: it is equally manifest in the Cirencester data:

	Rothamsted		Cirencester (Kinch)[1]	
	Rain (inches per annum)	Chlorine (lbs. per acre per annum)	Rain (inches per annum)	Chlorine (lbs. per acre per annum)
1888–89 to 1891–92	27·65	12·25	27·99	14·53
1892–93 to 1895–96	27·95	14·35	26·14	16·36
1896–97 to 1899–1900	28·12	17·90	27·26	17·33
1900–01 to 1903–04	27·58	17·23	31·83	24·78
1904–05 to 1907–08	27·14	16·75	26·22	17·60
1908–09 to 1911–12	31·03	18·48	31·38	22·43
1912–13 and 1913–14	26·17	15·89	30·49	26·12

There appears, however, to have been some other factor concerned besides the increase in rainfall, as the concentration of chlorine in the rainwater has also increased: formerly the rain contained about 2–2½ parts of chlorine per million, now the amount varies from 2½–3 parts per million.

The increase in amount of nitric nitrogen is shown in Tables 5 and 9 and in Fig. 4: it is not as steady as that of chlorine and is intensified by a few abnormally high values especially in two dry months February and July, 1913, in the wet October, 1913, in the dry May, 1914, and in the wet August, 1916. It is, of course, impossible now to check these figures: we have gone carefully through the laboratory notes and can find nothing to indicate that they are unreliable. Even apart from these exceptional cases, however, there is a clear upward trend in the nitrate figures. Examination of the curves in detail shows that the tendency to increase is spread over the whole year and is not confined to any one season: formerly there was about 0·12 to 0·19 parts of nitric nitrogen per million of rain, now the figures are 0·2 to 0·3 parts.

Although the rise in nitric nitrogen is similar to that of chlorine the phenomena are not necessarily related. We have no grounds for supposing that sea spray contains nitrates in sufficient quantity to produce the observed effects. On the other hand if some of the chlorine comes from coal a relationship might be expected, as part of the nitrate may come from the same source.

The ammonia, on the other hand, has decreased in concentration: formerly there were usually 0·4 to 0·5 parts per million, now there are more usually 0·2 to 0·3 parts. This is shown in Tables 4 and 9 and in Fig. 4.

[1] Data collected from papers in *Agricultural Students' Gazette* and *Trans. Chem. Soc.*, 1887, **51**, 92 and 1900, **77**, 1271.

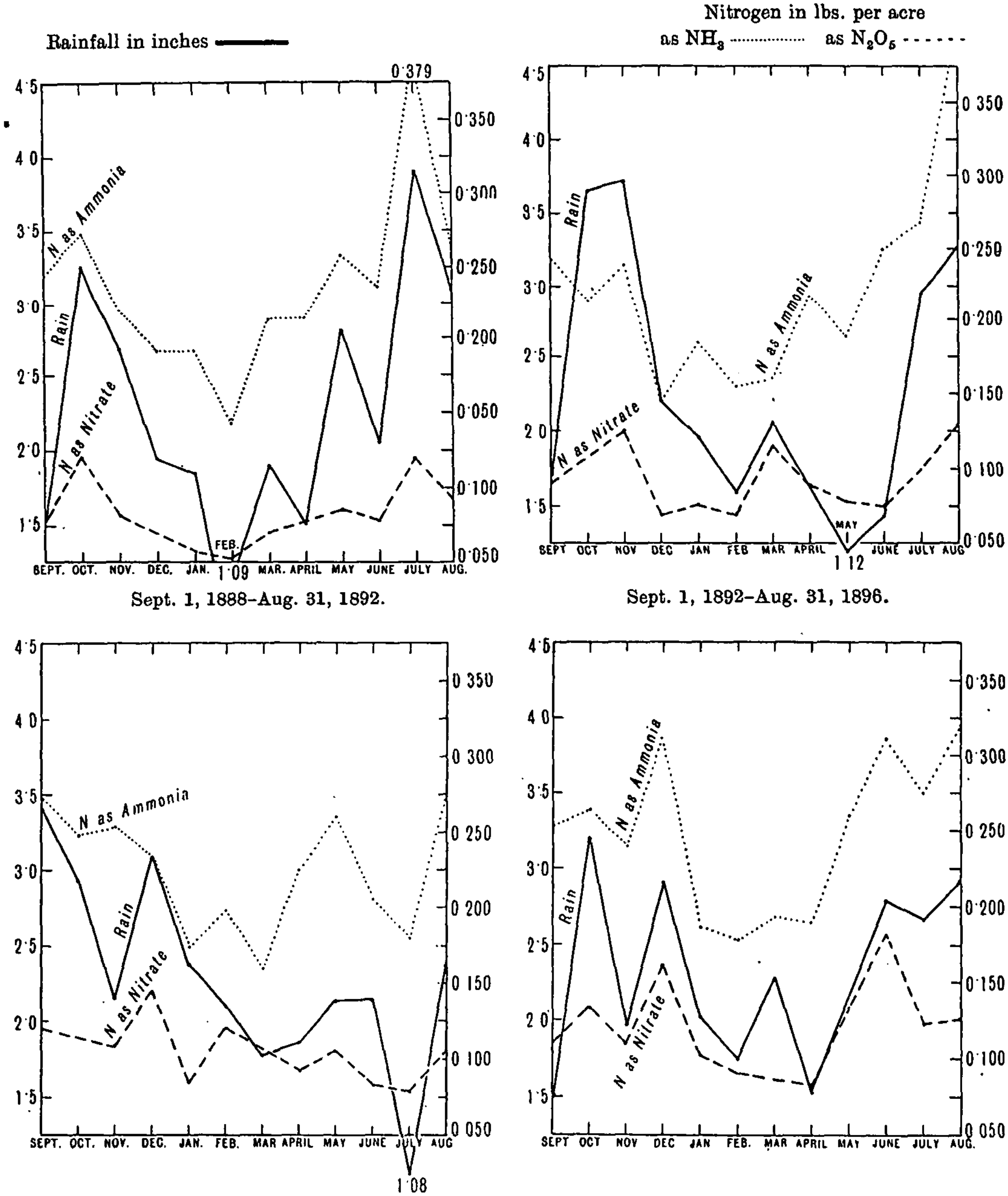
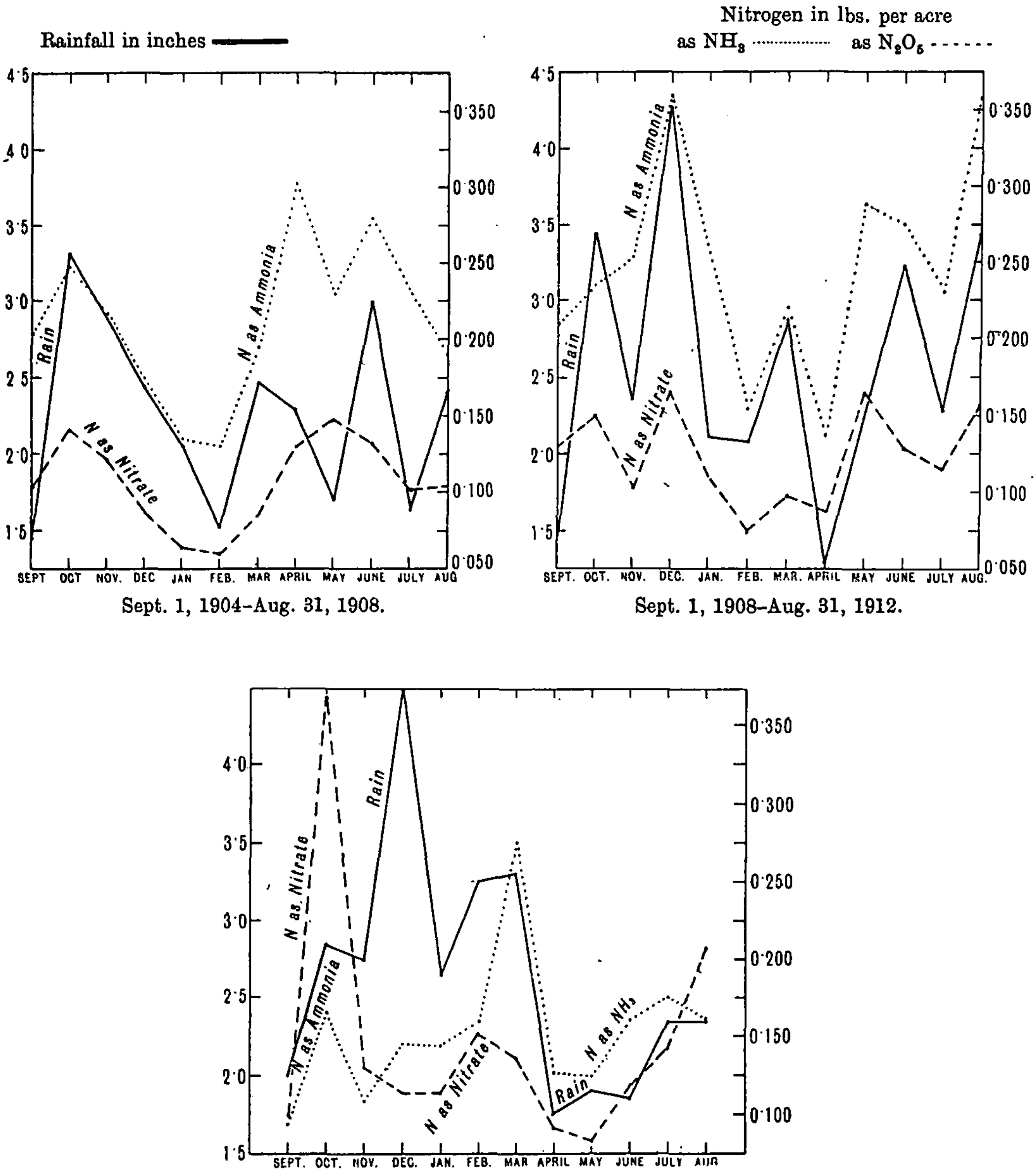


Fig. 4. Monthly fluctuations in amounts of nitric and ammoniacal nitrogen in rainwater: four year periods between Sept. 1, 1888 and Aug. 31, 1916.



Sept. 1, 1904–Aug. 31, 1908.

Sept. 1, 1908–Aug. 31, 1912.

Sept. 1, 1912–Aug. 31, 1916.

Fig. 4 (*cont.*).

The fall in ammonia and the simultaneous rise in nitrate may of course be wholly unconnected, but it suggests that a former source of ammonia now turns out nitrate instead, or that more nitrate is being produced from ammonia in the air than was previously the case. We have no data for examining the second possibility, but the first has some degree of probability: modern incandescent burners, gas and coal fires, may discharge more nitrous fumes and less ammonia into the atmosphere than the older types.

Dissolved oxygen in rainwater.

During the year 1915 estimations of dissolved oxygen in rain at Rothamsted were made[1] on rainfalls exceeding 0·30 inch. The great majority of the samples were found to be nearly saturated with oxygen but a few were considerably under saturation, notably those collected

Dissolved Oxygen in rainwater. Parts per Million

Date 1918	Temp. in °C. at time of collection	Bar. pressure mm.	Dittmar's figure for 760 mm.	Saturation corrected for pressure	Oxygen in sample	Percentage saturation (corrected)
			Summer Rain.			
July 11	15·5	745	10·2	9·8	9·7	99
,, 14	18·0	748	9·7	9·3	9·3	100
,, 19	20·5	750	9·2	8·8	8·7	99
,, 23	16·0	741	10·1	9·5	8·9	94
,, 26	15·5	749	10·2	9·9	9·1	92
Aug. 2	17·0	745	9·9	9·5	8·8	93
,, 25	18·0	749	9·7	9·3	8·5	91
				Mean	9·0	95 %
			Winter Rain.			
Nov. 29	9·5	752	11·6	11·2	10·6	95
Dec. 3	13·0	749	10·7	10·3	10·2	99
,, 10	11·0	750	11·2	10·8	10·6	98
,, 16	7·5	753	12·1	11·8	10·7	91
,, 19	7·0	733	12·2	11·7	11·8	101
,, 20	6·0	741	12·5	12·1	13·2	109
				Mean	11·2	99 %

in July and August. More recent determinations of summer and winter rains gave mean values of 95 and 99 per cent. respectively calculated on Dittmar's figures for distilled water saturated with air at the observed temperature and pressure. It seems possible that the greater height of

[1] This *Journal*, 1917, **8**, 331–337.

clouds in summer may account for the smaller oxygen content of rain, as condensation would occur at pressures much below normal. The velocity of the rain drops falling from greater heights may not allow time for equilibrium to be established with conditions at ground level.

The amounts of dissolved oxygen found were 9·0 parts per million in the summer, corresponding to 20·8 lb. per acre during the period May to August inclusive, and 11·2 parts per million in winter, corresponding to 26 lb. per acre during the four months November to February. Over the whole year the amount brought down was 66·4 lb. per acre. The results are shown in the table on p. 328.

The chemical characterisation of rainwater.

The above discussion shows that the rain falling at Rothamsted in winter differs chemically from that falling in summer: the values being

	Parts per million Four-month periods		Lbs. per acre Four-month periods	
	Winter (Nov.–Feb.)	Summer (May–Aug.)	Winter (Nov.–Feb.)	Summer (May–Aug.)
Ammoniacal nitrogen ...	0·35	0·45	0·78	1·00
Nitric nitrogen	0·18	0·21	0·40	0·47
Chlorine	3·38	1·38	7·50	3·08
Dissolved oxygen ...	11·2	9·0	26·0	20·8*

* In the whole year it is estimated that 66·4 lb. of dissolved oxygen is brought down.

The winter rainfall is richer in chlorine and oxygen but poorer in ammoniacal and nitric nitrogen than the summer rainfall.

The marked differences in the amounts of chlorine and ammonia suggest that winter rain may differ in origin from summer rain. The winter rain resembles Atlantic rain in its high chlorine and low ammonia content, suggesting that it is derived from the Atlantic. The summer rain, on the other hand, is characterised by a lower content of chlorine and higher proportion of ammonia which suggests that it arises from the soil by evaporation of water and condensation at higher altitudes than in the case of winter rain; this would also account for the difference in amount of dissolved oxygen.

SUMMARY.

The ammoniacal nitrogen in the Rothamsted rainwater amounts on an average to 0·405 part per million, corresponding to 2·64 lb. per acre per annum. The yearly fluctuations in lbs. per acre follow the rainfall fairly closely. The monthly fluctuations also move in the same direction as the rain, but the general level is highest during May, June,

July and August, and lowest during January, February, March and April (Tables 1, 2, 3, and Figs. 1 and 2).

The nitric nitrogen is on an average one-half the ammoniacal, viz., 1·33 lb. per acre per annum. The amounts fluctuated year by year and month by month in the same way as the ammoniacal nitrogen and the rainfall until 1910, since when there has been no simple relationship (Tables 1, 2, 3, and Figs. 1 and 2).

Reasons are adduced for supposing that the ammonia arises from several sources. The sea, the soil, and city pollution may all contribute. Neither the sea nor city pollution seems able to account for all the phenomena: the soil is indicated as an important source by the fact that the ammonia content is high during periods of high biochemical activity in the soil, and low during periods of low biochemical activity.

The close relationship between the amounts of ammoniacal and nitric nitrogen suggests either a common origin or the production of nitric compounds from ammonia.

The average amount of chlorine is 2·43 parts per million bringing down 16 lb. per acre per annum (Table 6). The fluctuations closely follow the rainfall both month by month and year by year, but the general level is much higher during the months September to April than during the summer months (Fig. 3). It seems probable that the chlorine comes from the sea, but some may come from fuel.

Since 1888, when the experiments began, to 1916, when they terminated, there has been a rise in the amounts of nitric nitrogen and of chlorine in the rain. (Tables 5, 7, 8 and 9; Figs. 3 and 4.) In the case of chlorine a parallel series of determinations made at Cirencester over the same period shows a similar rise (p. 325). There is no rise of ammonia but on the contrary a tendency to drop (Tables 4 and 9; Fig. 4): the sum of ammoniacal and nitric nitrogen shows little change over the period. This seems to suggest that a former source of ammonia is now turning out nitric acid: it is possible that modern gas burners and grates tend to the formation of nitric oxides rather than of ammonia.

Rain contains on an average 10 parts of dissolved oxygen per million, the amount being higher in winter than in summer: 66·4 lb. per acre per annum was brought down during the two years over which the determinations extended (p. 328).

The marked difference in composition between summer and winter rainfall suggests that these may differ in their origin. The winter rain resembles Atlantic rain in its high chlorine and low ammonia and nitrate content: the summer rain is characterised by low chlorine but high ammonia and nitrate content, suggesting that it arises by evaporation of water from the soil and condensation at higher altitudes than in the case of winter rain.

TABLE 1. *Rainfall at Rothamsted during harvest years, Sept. 1–Aug. 31.*

(For previous years see this *Journal*, 1905, **1**, 300.)

	Sept.	Oct.	Nov.	Dec.	Jan.	Feb.	March	April	May	June	July	August	Year
	inches	inches	inches	inches	inches	inches	inches	inches	inches	inches	inches	inches	inches
1905–6	2·25	1·67	3·23	1·10	4·09	2·33	1·64	0·80	1·38	3·61	0·42	1·25	23·77
1906–7	1·15	5·30	4·22	2·79	1·25	1·48	1·31	2·85	2·40	2·61	2·21	1·80	29·37
1907–8	0·78	4·89	2·44	3·40	1·58	1·34	3·40	3·28	1·89	1·68	2·43	3·01	30·12
1908–9	1·56	2·26	0·82	2·07	0·98	0·43	3·74	1·82	1·32	4·10	3·10	2·74	24·94
1909–10	1·94	5·19	1·25	3·42	2·18	3·78	1·24	1·58	2·16	2·81	2·00	3·38	30·93
1910–11	0·89	2·83	3·93	5·36	1·38	1·88	2·27	1·59	3·79	2·74	0·65	1·05	28·36
1911–12	1·52	3·47	3·44	6·26	3·89	2·21	4·29	0·17	1·47	3·28	3·35	6·53	39·88
1912–13	2·72	2·74	2·52	3·42	3·36	1·00	2·52	3·16	1·81	1·20	1·29	1·58	27·32
1913–14	1·50	3·49	2·94	0·87	0·88	3·45	4·43	1·15	1·42	1·72	1·57	1·59	25·01
1914–15	1·30	2·53	3·10	8·11	4·11	4·54	1·38	1·22	2·48	1·79	4·72	2·59	37·87
1915–16	2·49	2·60	2·38	5·56	2·24	3·97	4·92	1·43	1·97	2·71	1·77	3·58	35·62
Average for period Sept. 1, 1905–Aug. 31, 1916	1·65	3·36	2·75	3·85	2·36	2·40	2·83	1·73	2·01	2·57	2·14	2·64	30·29
Av. for whole period Sept. 1, 1888–Aug. 31, 1916	1·87	3·23	2·64	3·05	2·15	2·02	2·41	1·69	1·99	2·53	2·41	2·83	28·82

TABLE 2. *Nitrogen as ammonia and nitric acid in rainwater collected at Rothamsted.*

(For previous years see this *Journal*, 1905, **1**, 282.)

Harvest year September 1 to August 31	Rainfall in inches	Nitrogen: Per million As NH_3	Per million As N_2O_5	Per acre (lb.) As NH_3	Per acre (lb.) As N_2O_5	Per acre (lb.) Total	% of total As NH_3	% of total As N_2O_5
1901–2	23·26	0·571	0·267	3·006	1·407	4·413	68·1	31·9
1902–3	31·26	0·447	0·203	3·159	1·436	4·595	68·8	31·2
1903–4	31·50	0·424	0·214	3·026	1·523	4·549	66·5	33·5
1904–5	25·31	0·460	0·197	2·634	1·128	3·762	70·0	30·0
1905–6	23·77	0·373	0·194	2·007	1·045	3·052	65·8	34·2
1906–7	29·37	0·417	0·213	2·774	1·416	4·190	66·2	33·8
1907–8	30·12	0·404	0·222	2·752	1·516	4·268	64·5	35·5
1908–9	24·94	0·532	0·211	3·001	1·193	4·194	71·6	28·4
1909–10	30·93	0·393	0·179	2·753	1·254	4·007	68·7	31·3
1910–11	28·36	0·421	0·272	2·705	1·743	4·448	60·8	39·2
1911–12	39·88	0·381	0·190	3·436	1·718	5·154	66·7	33·3
1912–13	27·32	0·336	0·239	2·078	1·478	3·556	58·4	41·6
1913–14	25·01	0·246	0·323	1·395	1·827	3·222	43·3	56·7
1914–15	37·87	0·214	0·211	1·837	1·808	3·645	50·4	49·6
1915–16	35·62	0·253	0·232	2·037	1·874	3·911	52·1	47·9
Average for period Sept. 1, 1901–Aug. 31, 1916	29·63	0·384	0·222	2·573	1·491	4·064	63·3	36·7
Average for whole period Sept. 1, 1888–Aug. 31, 1916	28·82	0·405	0·203	2·644	1·327	3·971	66·6	33·4

The analytical determinations are carried out once a month on a sample made up from the daily collections. After each wet day a sample is taken from the gauge proportional in amount to the rainfall: one decigallon per inch of rain: this is kept in a Winchester quart bottle. At the end of the month the whole sample is measured and should correspond with the total rainfall.

The average figures quoted in the tables are true averages of rainfall and lbs. per acre but not of parts per million. In the latter case the figure shows the number of parts which when multiplied by the average rainfall gives the average number of lbs. per acre. This is the old Rothamsted convention and has been in use so long that change would be a serious matter. The figures are usually not widely different from the true averages, but no definite relationship exists between them.

TABLE 3. *Average monthly amounts of nitrogen as ammonia and nitric acid in rainwater collected at Rothamsted.*

Sept. 1, 1888 to Aug. 31, 1916	Rainfall in inches	Nitrogen: Per million As NH_3	Per million As N_2O_5	Per acre (lb.) As NH_3	Per acre (lb.) As N_2O_5	Per acre (lb.) Total	% of total As NH_3	% of total As N_2O_5
September	1·87	0·514	0·244	0·217	0·103	0·320	67·8	32·2
October	3·23	0·321	0·220	0·235	0·161	0·396	59·3	40·7
November	2·64	0·366	0·186	0·219	0·111	0·330	66·4	33·6
December	3·05	0·323	0·167	0·223	0·115	0·338	66·0	34·0
January	2·15	0·374	0·179	0·182	0·087	0·269	67·7	32·3
February	2·02	0·349	0·193	0·159	0·088	0·247	64·4	35·6
March	2·41	0·376	0·180	0·205	0·098	0·303	67·7	32·3
April	1·69	0·524	0·245	0·201	0·094	0·295	68·1	31·9
May	1·99	0·508	0·253	0·229	0·114	0·343	66·8	33·2
June	2·53	0·428	0·199	0·245	0·114	0·359	68·2	31·8
July	2·41	0·457	0·204	0·249	0·111	0·360	69·2	30·8
August	2·83	0·438	0·205	0·280	0·131	0·411	68·1	31·9
Sept.–Dec.	10·79	0·366	0·201	0·894	0·490	1·384	64·6	35·4
Jan.–April	8·27	0·399	0·196	0·747	0·367	1·114	67·1	32·9
May–August	9·76	0·454	0·213	1·003	0·470	1·473	68·1	31·9
April–Sept.	13·32	0·471	0·221	1·421	0·667	2·088	68·0	32·0
Oct.–March	15·50	0·348	0·188	1·223	0·660	1·883	64·9	35·1
Whole year	28·82	0·405	0·203	2·644	1·327	3·971	66·6	33·4

TABLE 4. *Nitrogen as ammonia in rainwater collected at Rothamsted in parts per million. Harvest years, Sept.* 1–*Aug.* 31.

(For previous results see this *Journal*, 1905, **1**, 301.)

	Sept.	Oct.	Nov.	Dec.	Jan.	Feb.	March	April	May	June	July	August	Year
1905–6	0·269	0·300	0·350	0·781	0·150	0·250	0·350	0·800	0·625	0·300	2·125	0·425	0·373
1906–7	0·883	0·263	0·319	0·204	0·325	0·452	0·525	0·650	0·525	0·525	0·500	0·350	0·417
1907–8	1·083	0·375	0·344	0·200	0·625	0·400	0·375	0·375	0·500	0·656	0·367	0·333	0·404
1908–9	0·650	0·321	0·800	0·500	0·781	1·281	0·470	0·450	0·766	0·400	0·425	0·719	0·532
1909–10	0·917	0·263	0·550	0·250	0·350	0·225	0·300	0·625	0·450	0·475	0·425	0·400	0·393
1910–11	0·525	0·475	0·225	0·250	0·650	0·257	0·375	0·338	0·625	0·313	0·875	1·300	0·421
1911–12	0·300	0·200	0·650	0·500	0·550	0·375	0·213	0·317	0·500	0·313	0·400	0·250	0·381
1912–13	0·183	0·188	0·213	0·188	0·338	0·525	0·775	0·400	0·300	0·425	0·500	0·263	0·336
1913–14	0·217	0·225	0·058	0·263	0·300	0·158	0·150	0·250	0·575	0·400	0·600	0·650	0·246
1914–15	0·319	0·375	0·300	0·117	0·200	0·150	0·300	0·288	0·288	0·425	0·188	0·188	0·214
1915–16	0·167	0·350	0·117	0·133	0·142	0·263	0·375	0·225	0·350	0·325	0·350	0·263	0·253
Averages:													
1905–1916	0·433	0·289	0·320	0·260	0·351	0·274	0·365	0·438	0·468	0·398	0·428	0·390	0·355
1888–1916	0·514	0·321	0·366	0·323	0·374	0·349	0·376	0·524	0·508	0·428	0·457	0·438	0·405

TABLE 5. *Nitrogen as nitrates in rainwater collected at Rothamsted in parts per million. Harvest years, Sept.* 1–*Aug.* 31.

(For previous results see this *Journal*, 1905, **1**, 302.)

	Sept.	Oct.	Nov.	Dec.	Jan.	Feb.	March	April	May	June	July	August	Year
1905–6	0·188	0·192	0·225	0·325	0·083	0·133	0·083	0·450	0·400	0·175	0·475	0·213	0·194
1906–7	0·325	0·133	0·131	0·184	0·225	0·150	0·250	0·275	0·400	0·250	0·263	0·175	0·213
1907–8	0·625	0·213	0·213	0·092	0·225	0·213	0·175	0·200	0·350	0·325	0·225	0·231	0·222
1908–9	0·400	0·221	0·375	0·175	0·219	0·525	0·142	0·158	0·263	0·175	0·142	0·263	0·211
1909–10	0·294	0·092	0·200	0·125	0·133	0·075	0·117	0·300	0·275	0·250	0·263	0·238	0·179
1910–11	0·600	0·325	0·175	0·200	0·281	0·183	0·250	0·450	0·375	0·142	0·313	0·450	0·272
1911–12	0·375	0·213	0·163	0·167	0·275	0·200	0·108	0·500	0·375	0·142	0·250	0·117	0·190
1912–13	0·139	0·213	0·142	0·117	0·183	0·850	0·250	0·300	0·175	0·263	0·525	0·350	0·239
1913–14	0·325	1·063	0·250	0·275	0·400	0·125	0·092	0·150	0·550	0·288	0·225	0·300	0·323
1914–15	0·350	0·450	0·213	0·100	0·142	0·175	0·283	0·263	0·200	0·375	0·238	0·213	0·211
1915–16	0·158	0·400	0·213	0·100	0·200	0·158	0·200	0·150	0·225	0·225	0·225	0·575	0·232
Averages:													
1905–1916	0·292	0·307	0·194	0·144	0·190	0·180	0·166	0·262	0·297	0·218	0·250	0·264	0·225
1888–1916	0·244	0·220	0·186	0·167	0·179	0·193	0·180	0·245	0·253	0·199	0·204	0·205	0·203

Table 6. *Average monthly amount of chlorine in rain falling at Rothamsted.*

Sept. 1, 1877 to Aug. 31, 1916	Rainfall Average 39 years Inches	Chlorine	
		Per million	Per acre (lb.)
September ...	2·15	1·87	0·91
October	3·24	2·56	1·88
November ...	2·82	2·86	1·82
December ...	2·89	3·27	2·14
January ...	2·14	3·75	1·82
February ...	2·10	3·63	1·72
March	2·16	3·43	1·68
April	1·82	2·50	1·03
May	2·10	1·93	0·92
June	2·55	1·29	0·74
July	2·47	1·09	0·61
August	2·78	1·29	0·81
April–September	13·86	1·60	5·02
October–March ...	15·35	3·18	11·06
Whole year ...	29·21	2·43	16·08

TABLE 7. *Chlorine in rainwater collected at Rothamsted in parts per million.*

(For previous results see this *Journal*, 1905, 1, 303.)

	Sept.	Oct.	Nov.	Dec.	Jan.	Feb.	March	April	May	June	July	August	Year
1905–6	2·96	3·00	3·12	3·36	2·04	3·48	4·35	5·75	3·45	1·05	3·71	1·85	2·78
1906–7	3·98	1·65	2·28	3·20	6·72	3·35	4·68	2·21	2·51	2·25	1·56	1·05	2·55
1907–8	2·85	2·93	3·00	3·00	7·13	6·24	2·70	2·71	1·88	1·14	1·32	2·43	2·91
1908–9	2·70	1·95	6·93	4·05	5·93	10·27	2·25	1·91	2·90	1·22	0·98	1·29	2·41
1909–10	2·88	3·12	6·60	2·16	3·60	4·23	2·25	4·22	2·34	1·05	1·82	1·29	2·76
1910–11	3·56	4·65	2·33	4·08	6·63	4·41	3·60	3·39	2·40	1·89	1·61	2·43	3·39
1911–12	1·97	2·67	3·11	2·22	2·60	2·91	2·72	6·42	2·28	1·14	1·38	1·08	2·13
1912–13	2·18	2·90	2·60	2·70	3·33	8·25	4·26	2·67	1·56	1·83	1·37	1·77	2·85
1913–14	2·75	2·22	2·07	6·06	6·82	3·50	2·19	2·91	3·57	1·56	1·29	1·41	2·50
1914–15	3·08	3·12	3·45	3·18	2·25	3·66	3·03	2·86	1·34	1·73	1·31	0·87	2·56
1915–16	1·17	2·94	3·57	3·69	4·88	3·68	4·91	2·82	1·77	1·59	0·98	1·40	3·03
Average for period 1905–1916	2·56	2·91	3·06	3·19	3·79	4·09	3·29	2·87	2·12	1·44	1·37	1·43	2·73
Average for whole period 1877–1916	1·87	2·56	2·86	3·27	3·75	3·63	3·43	2·50	1·93	1·29	1·09	1·29	2·43

TABLE 8. *Chlorine in rainwater collected at Rothamsted.*

Harvest year Sept. 1 to Aug. 31	Rainfall in inches	Parts per million	Lbs. per acre
1901–2	23·26	2·81	14·81
1902–3	31·26	2·52	17·84
1903–4	31·50	2·76	19·66
1904–5	25·31	2·66	15·24
1905–6	23·77	2·78	14·94
1906–7	29·37	2·55	16·95
1907–8	30·12	2·91	19·86
1908–9	24·94	2·41	13·62
1909–10	30·93	2·76	19·33
1910–11	*26·77	3·39	*20·55
1911–12	39·88	2·13	19·23
1912–13	27·32	2·85	17·61
1913–14	25·01	2·50	14·17
1914–15	37·87	2·56	21·90
1915–16	35·62	3·03	†24·40
Average for period 1901–1916	29·53	2·70	18·01
Av. for whole period 1877–1916	29·21.	2·43	16·08

* 11 months only: April figures not included.

† August value estimated.

TABLE 9. *Nitrogen as ammonia and nitric acid and chlorine in rainwater collected at Rothamsted in seven periods of four years, Sept.* 1, 1888 *to Aug.* 31, 1916.

September 1 to August 31	Rainfall in inches	Nitrogen: Lbs. per acre: As NH_3	Nitrogen: Lbs. per acre: As N_2O_5	Nitrogen: Lbs. per acre: Total	Nitrogen: % of total: As NH_3	Nitrogen: % of total: As N_2O_5	CHLORINE Lbs. per acre
1888–9 to 1891–2	27·65	2·815	0·981	3·796	74·2	25·8	12·25
1892–3 to 1895–6	27·95	2·633	1·081	3·714	70·9	29·1	14·35
1896–7 to 1899–1900	28·12	2·682	1·264	3·946	68·0	32·0	17·90
1900–1 to 1903–4	27·58	2·982	1·435	4·417	67·5	32·5	17·23
1904–5 to 1907–8	27·14	2·542	1·276	3·818	66·6	33·4	16·75
1908–9 to 1911–12	31·03	2·974	1·477	4·451	66·8	33·2	18·48
1912–13 to 1915–16	31·46	1·837	1·747	3·584	51·3	48·7	19·54

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