EXPERIENTIA MAIORUM

The Discovery of Atmospheric Condensation Nuclei by Paul-Jean Coulier in 1875

A historical note

By F. VERZÁR*

Nobody will doubt the great value of JOHN AITKEN's[1–4] work of several decades on atmospheric condensation nuclei as the cause of haze, fog, and clouds, but all the basic observations were made without AITKEN's knowledge several years before by PAUL-JEAN COULIER, professeur au Val de Grâce, the military medical and pharmaceutical school of Paris.

In the Journal de Pharmacie et de Chimie (Ser. IV, *22*, 165, September 1875), appeared a paper entitled *Note sur une nouvelle propriété de l'air* par M. COULIER[5], professeur au Val de Grâce.

The Figure shows the first page.

COULIER describes a simple device for demonstrating the fact, which was already known at the time, that a sudden decrease in pressure in a closed volume of air, saturated with water vapour, leads to the formation of fog. Originally he used a 3-meter-long tube of zinc closed at both ends with a glass plate. The air inside was compressed first. It was then sufficient to open a stop-cock to decrease the pressure, and at this moment a fog appeared which was so thick that a candle flame could no longer be seen through the tube. (This certainly sounds like the model of our modern nucleus counters, but – of course – without photocells and electronics.)

A second device was also described. A flask with 3 stop-cocks was used. One was connected with a rubber balloon. The air was sucked in and, by compression of the balloon, was brought under higher pressure which could then be released at will, and then a fog appeared.

He even observed the fog droplets with a simple magnifying glass and saw that they are of different size and mobility. He also started counting the nuclei number per cm^3. He found that the air of Paris contains many such dust particles – as he called them – which he supposed to be responsible for the 'bad air' of the big city.

He then makes the interesting observation that after several decompressions of the same volume of air, it becomes 'inactive'. Oxygen and carbon dioxide do not play a part in this. With outside air, the reaction (fog formation) appears again.

Through a cotton-wool filter, the particles of the air can be filtered away and such air then becomes 'inactive', that is no fog is produced by sudden decrease in pressure.

These invisible particles sediment when the air stands. They are also retained in the respiratory tract (bronchi). Tobacco smoke makes air 'extraordinarily active'. An alcohol flame or a benzol flame, or a Bunsen burner, produces very much 'activity', and these particles are retained by cotton wool filters. Since the latter become black, he concludes that the dust-producing particles are very small, unburned particles of carbon. A calculation showed him that as little as $^{7}/_{100\,000}$ mg 'activate' the air.

***Note sur une nouvelle propriété de l'air;* par M. COULIER, professeur au Val-de-Grâce.**

On sait que lorsqu'une certaine quantité d'air saturé de vapeur d'eau est raréfiée brusquement, une partie de cette vapeur se précipite sous forme de brouillard, par suite de l'abaissement de température. — Pour rendre cette expérience plus visible, j'ai fait construire un large tube en zinc, de 3 mètres environ de longueur, et terminé par des glaces. En introduisant un peu d'eau dans cet appareil et en comprimant légèrement l'air qu'il contient, il suffit d'ouvrir un robinet placé sur le côté du tube pour que la décompression se produise. A ce moment un nuage se forme, et il est assez opaque pour qu'on ne puisse plus voir les contours de la flamme d'une bougie.

En répétant cette expérience à plusieurs jours d'intervalle, je m'aperçus qu'elle était capricieuse, et que souvent elle ne réussissait pas. C'est pour rechercher les causes de cet insuccès que j'ai entrepris les expériences dont je vais dire quelques mots.

Le phénomène de la précipitation de la vapeur d'eau dans une atmosphère qu'on raréfie présente une particularité qui permet de l'étudier plus facilement. On peut, pour le produire, provoquer dans l'air saturé une compression momentanée qui ne dure qu'un instant très-court. Le brouillard se produit immédiatement au moment de la décompression. L'appareil suivant est fort commode pour ce genre d'expérience. Il se compose d'un flacon de 1 à 2 litres, à trois tubulures (A, fig. 1). La première, B, est munie d'un robinet et se termine par un tube en caoutchouc. La deuxième, C, est dépourvue de robinet,

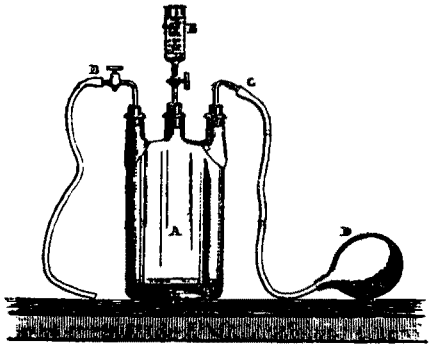

Fig. 1.

mais le tube en caoutchouc se termine par une poire, D.

He further showed that the number of nuclei changes with the weather, and he therefore proposed that in towns, and also at different altitudes, and during epidemics (!), the presence of these 'dust' particles should be measured and their quality studied.

Ozone produced great activity in his air samples, and this active principle was not retained by the cotton wool filters. This fact remained inexplicable and is today again under discussion (JUNGE[6]).

* Basel.

[1] J. AITKEN, Trans. R. Soc. Edinburgh, Dec. 20th (1880).

[2] J. AITKEN, Nature *23*, 195 (1881).

[3] J. AITKEN, Nature *23*, 384 (1881).

[4] J. AITKEN, *Collection of Scientific Papers* (University Press, Cambridge 1923).

[5] M. COULIER, J. Pharm. Chim. [4] *22*, 165, 254 (1875).

[6] C. E. JUNGE, Adv. Geophys. *4*, 1 (1958).

Of special interest is his remark that 'Sulphur was the most active fog-producing substance'. In recent years, GERHARD and JOHNSTONE[7] and JUNGE[6] have discussed the role of SO_2 in the atmosphere and its possible oxydation to sulphuric acid by ozone and by a photochemical reaction (VERZÁR[8,9]).

In the October number of the same journal where COULIER's[5] first paper appeared, followed a second additional note with a similar title, of $1^1/_2$ pages only. In this he was much perturbed whether his original interpretation, that the dust nuclei are mainly combustion products, was right. He now had made the following observations: The flask was filled with inactive (filtered) air. In the bottle was a platinum wire, which was brought to a red glow. The air then became very active. This and some more experiments with heated glass-tubes etc. made it uncertain whether his explanation was right that carbon particles are the nuclei on which water vapours condense. Here a new form of nuclei were produced which – he thought – were not products of burning organic matter.

It cannot be said that COULIER's papers were quite unknown; the German scientific weekly periodical 'Naturforscher' gave a lengthy review in Vol. *8* (1875), p. 400 to 401, and a second one on p. 453.

However, 5 years later, AITKEN[1] published in the Royal Society of Edinburgh on December 20th, 1880, his famous paper in which he showed that 'the dust is the germ on which fogs and clouds are the developed phenomena', as it was quoted in Nature *23*, December 30th (1880), p. 195 and p. 204.

This led to a series of critical 'Letters to the Editor' in Nature, by RUSSELL[10], PREECE[11], and mainly one by GRONEMAN[12] in Groningen (Holland). The latter pointed out the importance of COULIER's work on p. 337 as primary to that of AITKEN, but GRONEMAN was interested in the problem only from the point of view of his 'théorie cosmique de l'aurore polaire'.

AITKEN[3] answered in a letter to the Editor and fully acknowledged that he had not known of COULIER's work: 'I need not say that the information... was a most unexspected surprise. Nothing whatever seems to have been known in England about the results obtained ... There be no doubt that M. COULIER was the first to show the important part played by dust in the cloudy condensation of the vapour in air.' AITKEN thought, however, that COULIER's second paper, in which he showed that a heated glowing platinum wire also produced condensation nuclei, was the cause that no further interest had been taken in his work. AITKEN himself explained this experiment so that some dirt must have been present which burned on the wire, and thus no special explanation of the origin of the nuclei would be necessary.

Today we see the situation differently: Only recently O'CONNOR *et al.*[13] have studied the nuclei produced from glowing metal wires and found that they have a radius of 1×10^{-6} cm; but their nature is still unknown today. Thus COULIER made an important new observation even in this second short note of 1875.

[7] E. R. GERHARD and H. F. JOHNSTONE, Ind. Eng. Chem. *47*, 972 (1955).

[8] F. VERZÁR and Y. KUNZ, Geofis. pur. appl. *38*, 215 (1957).

[9] F. VERZÁR, Geofis. pur. appl., in press (1959).

[10] R. RUSSELL, Nature *23*, 267 (1881).

[11] W. H. PREECE, Nature *23*, 194 (1881).

[12] M. H. J. H. GRONEMAN, Nature *23*, 337 (1881).

[13] T. C. O'CONNOR, W. P. F. SHARKEY, and C. O'BROILCHAIN, Geofis. pur. appl. *42*, 109 (1959).

It seems astonishing that COULIER did not continue this most promising completely new work. The explanation is that less than one year after his most important discoveries, in 1876, he left his chair of chemistry and accepted an administrative position as 'Inspecteur' in the army medical services[14]. Obviously he even did not take notice of AITKEN's work on his problems during the next 15 years.

PAUL-JEAN COULIER was born in Paris on August 31st, 1824. He was a pharmacist and became in 1853 professeur agrégé on the 'Ecole de médecine et de pharmacie militaire du Val de Grâce' in Paris and from 1858 professor of applied chemistry and hygiene.

From his 30 papers the earlier dealt with blood and viruses, the later with hygiene of soldiers, heating and ventilation[15]. This obviously lead him to study formation of fogs. After 1876, when he took up his new post, he was especially known for his fights for the liberty of medical opinion in military service. He died greatly honoured, on July 23rd, 1890.

Few know his name today, but he is mentioned in later reviews. (LANDSBERG[16], p. 160 and p. 172, MASON[17], GREEN, and LANE[18].)

COULIER should be remembered as the first who showed with ingeniously simple methods, and few words, the existence, the rôle and most of the basic facts of atmospheric condensation nuclei.

I have to thank Médecine-Colonel HASSENFORDER of the Musée du Val de Grâce, and the Bibliothèque centrale du service de santé, for the biographical data.

Zusammenfassung

1875 hat PAUL-JEAN COULIER, Professor für Chemie und Hygiene an der Medizinischen Militärschule Val de Grâce in Paris, mit einfachen Mitteln die Rolle der atmosphärischen Kondensationskerne bei der Nebelbildung entdeckt und viele wesentliche Tatsachen beschrieben. Erst 5 Jahre später erfolgte die erste Arbeit von AITKEN. Die Geschichte dieser Entdeckung und die Ursache dafür, dass sie nicht besser bekannt wurde, wird besprochen.

[14] CHARLES LAVANZALLE, *Notices Biographiques sur les anciens inspecteurs de l'armée* (Paris 1892), p. 35.

[15] A. BALLAND, *Travaux scientifiques des pharmaciens militaires français* (Edit. Asselin, Paris 1882), p. 26.

[16] H. E. LANDSBERG, Erg. Kosm. Phys. *3*, 155 (1938).

[17] B. J. MASON, *The Physics of Clouds* (Oxford 1957), p. 20.

[18] H. L. GREEN and W. R. LANE, *Particulate Clouds, Dusts, Smokes and Mists* (London 1957), p. 231.

COGITATIONES

Rauschzustände nach Pilzgenuss

Bei der Eroberung von Mexiko lernten die Spanier in den südlichen Provinzen eigenartige Kulthandlungen kennen, während derer sich die Teilnehmer durch den Genuss von Hutpilzen in Trance versetzten. Pater DE OLMOS berichtet schon um das Jahr 1543, dass junge indianische Männer und Töchter zur Erbauung der Götter in blumengeschmückten Tempeln tanzten, wobei der Satan sie einen berauschenden Pilz einnehmen liess, so dass sie ihre Sinne verloren und Visionen sahen.