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JAN INGEN HOUSZ AND JOSEPH PRIESTLEY CONTROVERSY ON CARBON DIOXIDE ABSORPTION

RICHARD FOREGGER, M.D.

CONSIDERABLE scientific interest during the latter part of the eighteenth century was directed toward the production and properties of various types of gases. Among those actively engaged in this research were Joseph Priestley and Jan Ingen Housz.

The number of gases known to chemists when Priestley began his work were three: carbon dioxide, inflammable air, and atmospheric air. He increased the number by ten, which in modern terms are hydrogen chloride, nitrogen, oxygen, nitric oxide, nitrous oxide, nitrogen peroxide, ammonia, sulfur dioxide, hydrogen sulfide, and silicon tetrafluoride (1–4). Of these, nitrogen was isolated and identified independently by Rutherford, and oxygen by Scheele. The majority, but not all, of Priestley's researches in chemistry were recorded in the three volumes of "Experiments and Observations on Different Kinds of Air," followed by a sequel in three volumes, "Experiments and Observations Relating to Various Branches of Natural Philosophy with a continuation of the Observations on Air," published successively between 1774 and 1786. For convenience Priestley referred to these as if they formed six volumes in continuation under the same title which he abbreviated to "E. and O." Priestley's major interest was theology, but for a time be taught classical languages and belles lettres, and for six years (1773–1779) he was librarian and literary companion to the Earl of Shelburne at whose home in London and at Bowood near Calne he made many of his major discoveries of gases.

In 1772 Priestley published a long paper entitled "Observations on Different Kinds of Air" (5) which marks an epoch in the history of science. This paper describes new methods of collecting and manipulating gases; the discovery of two new gases, nitric oxide and hydrogen chloride; a method of utilizing nitric oxide for measuring the goodness of air; and a method of impregnating water with carbon di-

Accepted for publication February 14, 1956. Dr. Foregger is Associate Professor of Anesthesiology, Marquette University School of Medicine, Milwaukee, Wisconsin.

oxide. He showed that in air exposed over water, one-fifth disappears in processes of combustion, respiration, and putrefaction and that living green plants sometimes restore air vitiated by these processes. It was not by chance that he sought for some cause for the restoration of air and found it in living, green vegetation. As early as 1767 he had tried to restore air rendered mephitic in respiration or combustion by passing electric current through it (6). He realized that this was a great problem in the world economy.

The publication of "Observations on Different Kinds of Air," soon followed by the first volume of his book, "Experiments and Observations on Different Kinds of Air" (1774), made him justly famous. The paper and the book had great influence upon contemporary chemists, among them Fontana, Cavendish, Landriani, Lavoisier, and Cavallo. Ingen Housz says in his preface to "Experiments on Vegetables" (1779):

> When I first found in the works of that excellent philosopher and inventive genius, the Reverend Dr. Priestley, his important discovery, that plants wonderfully thrive in putrid air; and that the vegetation of a plant could correct air fouled by the burning of a candle, and restore it again to its former purity and fitness for supporting flame, and for the respiration of animals; I was struck with admiration. . . .

Jan Ingen Housz * was born at Breda, Holland, on December 8, 1730, according to Wiesner, whose biography of Ingen Housz provided most of the material for this summary. His mother died when he was a year old. His father, Arnoldus Ingen Housz, whose occupation is uncertain, was sufficiently well-to-do to be able to grant his son Jan a finishing education, extending far beyond the usual measure, at the Universities of Leuven and Leiden (9, 10). He remained at his father's house in Breda and studied Latin and Greek at school up to the age of 16. Following this he went to Leuven and studied medicine. He graduated as a doctor of medicine at the age of 22 and went subsequently to Leiden to study physics, in which subject this university was outstanding at that time. In 1757 he returned to Breda where he practiced medicine for eight years. Some time after the death of his father in 1764, he went to England on the invitation of Sir John Pringle, famous military doctor and personal physician to the King of England, who had been a friend of the Ingen Housz family for twenty years (11, 12). The friendship with Pringle assured him of being accepted into the medical and scientific circles of London. He went to Edinburgh for a short time and studied under Cullen. Upon his return to London he studied children's diseases, especially inoculation for

* "In his works which appeared in print, the style of his name is Ingen-Housz, as also in the translations of his writings authorized by him. The name, however, appears in various ways according to the country and the correspondent." (7). "Ingen Housz never used the hyphen and the present living members of the family do not use the hyphen either" (8).

smallpox. His skill in smallpox inoculation led to his appointment, on the recommendation of the King of England, as court physician to the Austrian monarchs at Vienna, at which place he arrived in May, 1768. Here he successfully inoculated two of the children of the Empress Marie Theresa, and a daughter of Josef II. Early in 1769, the Empress sent him to Florence to inoculate her son, the Grand Duke of Tuscany, and on this occasion, he met and discussed scientific matters with the Abbé Felice Fontana (13, 14, 15). In this same year (May 25, 1769) he was elected fellow of the Royal Society of London (16) and became a member in March, 1771. In 1770–71 he visited Switzerland, Paris, Holland and England.

On this visit to England in 1771 he went on a two-week trip with Benjamin Franklin, at which time they called on Priestley at Leeds (17, 18). Ingen Housz spent almost all of this year in England often in the company of Sir John Pringle (19). In 1772 he traveled again to Florence to inoculate the children of the Grand Duke. In the summer of 1777, he was in Paris with Fontana, at which time they made examinations on the air extracted from water by heat (20, 21). In November, 1777, he was in Amsterdam where he observed Aeneae and Cuthberson perform experiments on a new inflammable air (ethylene). He arrived in London early in January, 1778 (22).

In three months during the summer of 1779 Ingen Housz performed the more than 500 experiments which form the basis of his famous book on the relation between air and plants, "Experiments on Vegetables" (1779). These experiments and the book were written as he relates at his "home in the country at Southall-Green, 10 miles from London" and at his home in London "next to the garden of the hotel named Carlton-House" (23). Before returning to Vienna, he visited again with Benjamin Franklin and Felice Fontana at Paris, remaining there from January to July, 1780 (7, 17). He stayed in Vienna for eight years before returning to England in 1789. For the rest of his life he remained in England, for during the last ten years the political situation on the Continent and his health were such that he never felt equal to attempting the journey back. His wife stayed in Vienna. He lived in London but visited often at Bowood, the estate of the Earl of Shelburne (the first Marquess of Lansdowne) near Calne. This was the same place where Priestley had been librarian. Ingen Housz died on September 7, 1799 and was buried at Calne (24).

Ingen Housz is known primarily for his work on the gas exchange in plants. He showed that oxygen evolution from green plants is most vigorous in sunlight and ceases in darkness. Green leaves in the dark, and roots, flowers, fruits, and the like vitiate the atmosphere by emitting a toxic gas. Ingen Housz was a scientific investigator of considerable stature. His cleverly conceived and well-executed experiments are a landmark in the development of the problem of the interaction of plants with the atmosphere (25). He also, however, made studies in the fields of electricity and magnetism, on the explosibility of ether and oxygen, on the composition of the air over the sea compared to that over the land, on the explosiveness of gunpowder, and on the heat conductivity of metals (26, 27).

Ingen Housz acknowledged his debt to Priestley, from whom he took much inspiration. The two men were personally known to each other and both were friends of Benjamin Franklin which relationship subsequently became significant.

Fontana Method of Carbon Dioxide Absorption

The controversy between Ingen Housz and Priestley regarding carbon dioxide absorption by limewater has been mentioned in an interesting article by Waters (28). It began with the publication by Ingen Housz in the preface to his "Experiments on Vegetables" (1779) where he says (page 45):

When this book was entirely printed, and nothing but the latter end of the preface unfinished, I was informed by my friend, the Abbé Fontana, that he discovered a few days ago a new method of procuring to a sick person the benefit of breathing any quantity of dephlogisticated air at a cheap rate.

Abbé Fontana found that an animal breathing in either common or dephlogisticated air renders it unfit for respiration by communicating to it a considerable portion of fixed air, which is generated in our body, and thrown out by the lungs as excrementitious. This fixed air is easily absorbed by shaking it in common water, but infinitely more readily by the contact with quick-lime water.

He fills one of the large receivers of an air-pump, which are very wide at their upper extremity, half full of dephlogisticated air extracted from nitre, so that it may contain about 500 cubic-inches of this air, which will serve for breathing during half an hour. The manner of drawing this air out of the receiver, is either by thrusting a bended glass tube under the receiver (when this is floating in water, in which it is supported by its peculiar bulky form), reaching into the air itself, and keeping the other extremity in the mouth; thus drawing this air in the lungs, and breathing it out by the same tube. This air returning from the lungs is infected by fixed air, which being immediately absorbed by the contact with lime-water, the dephlogisticated air is restored very near to its former purity. Instead of the bended tube just mentioned, the top of the receiver may be made as the neck of a bottle, and a tube may be fixed to it, having a cock to open and shut the passage as required.

We consume, by each inspiration, about 30 cubic-inches of air; and thus, allowing 15 inspirations for a minute, we consume each minute 450 cubic-inches of air. The Abbé Fontana found, that the dephlogisticated air being, after each respiration, purified again by the lime-water, will remain good about thirty times as long as it would when breathed in the ordinary way; and that thus the quantity of dephlogisticated air necessary for one minute will now serve for breathing during half an hour, and thus the expences will be thirty times less.

We consume, in the space of 24 hours, about 21,600 cubic-inches of air. One pound of nitre yields by heat about 12,000 cubic-inches of dephlogisticated air, and thus it yields more air than any patient could consume by breathing this beneficial air the whole day (for we must allow at least 12 hours in the 24 for sleeping and necessary occupations), because this quantity will serve thirty times longer by the method explained, than in the ordinary way. It follows by this, that the expences required for breathing a whole day dephlogisticated air cannot amount to one shilling.

I have only just time enough to announce this happy discovery to the publick; whose great utility will, I trust, soon be found in the curing of inflammatory and putrid diseases, &c. in which too great a quantity of inflammable principle is let loose in our blood.

An investigation of the literature up to the present time has not revealed a written communication from Fontana to Ingen Housz in 1779 regarding this method of carbon dioxide absorption by limewater. Both Fontana and Ingen Housz were in London in 1779. Fontana says:

I was in London in 1779 when I communicated to my very respectable friend, Ingen Housz, the results of my various experiments on the respiration of animals, of which he has already referred to in his wonderful work entitled "Experiments and Observations upon Vegetables" (29).

Cavallo (30) who lived in London at this time says:

Now Mr. Fontana has found (as is related by Dr. Ingen Housz, and as I remember myself to have heard him say) that an animal confined in a quantity of dephlogisticated air will live about thirty times longer in it, when the receiver, containing the dephlogisticated air *stands in limewater,* than when it stands in common water, because the lime-water absorbs the fixed air [CO_2] very readily (31).

From the above we conclude that Fontana's communication to Ingen Housz regarding a method of carbon dioxide absorption by limewater probably was verbal.

PRIESTLEY'S CRITICISM

Priestley criticized Ingen Housz' book "Experiments on Vegetables" extensively in volume V of his "Experiments and Observations on Different Kinds of Air" (1781), much of the criticism being directed toward Ingen Housz' observations on the role of green plants in purifying the atmosphere. Fixed air (CO_2) absorption by limewater occupies only a few pages. In commenting on this subject he says (page 158):

My friend Dr. Ingen-Housz has announced what he thought to be a very valuable discovery of the Abbé Fontana's with respect to the breathing of dephlogisticated air; and had there been no mistake in the business, it would have been a discovery of the very first magnitude. It is a method of making dephlogisticated air serve thirty times longer for respiration than when it is breathed in the common way, so that a pound of nitre would yield dephlogisticated air sufficient for the respiration of a man a whole day. . . .

Here Priestley quotes the statement by Ingen Housz in his preface regarding Fontana's method of carbon dioxide absorption. Priestley in commenting on this says (pp. 159–160):

This language supposes that the Abbé had not only *reasoned* upon the case, but that he had also *verified* his reasoning by actual experiment; because it is said that he *found* it to be so. On the contrary, I can neither find any such thing in fact, nor the least colour for the expectation of it in reasoning; there being no advantage whatever in breathing dephlogisticated air in the manner that Dr. Ingen Housz describes. And his hypothesis concerning the nature of the injury that is done to air by respiration is manifestly erroneous. For the precipitation that is made of fixed air is nothing more than a *circumstance* attending the respiration of common or dephlogisticated air, the proper effect of that animal process being, as I think I have fully demonstrated, the *phlogistication* of the air; and therefore, though the precipitated fixed air be absorbed ever so readily, the remaining air will be but very little the better for it. For if we were to mix much more than that proportion of fixed air with the air that we breathe, we should not perceive it to be at all inconvenient to us.

It was but reasonable, however, that the assertion of so eminent a philosopher, and the assertion of a fact, should be tried by fact.

PRIESTLEY'S EXPERIMENT

Priestley performed an interesting experiment to test the comparative effectiveness of limewater and common water. Although his interpretation of the results was based upon his application of the phlogiston theory to the respiration process, a general analysis of the experiment in terms of present day knowledge shows the care with which the experiment was carried out.

Priestley does not give the weight of his mice, but elsewhere (page 79) he speaks of a "middle sized mouse" weighing 6 pennyweight, 3 grains (9.52 Gm.). He says (p. 162), "Both the mice were kept pretty warm." Elsewhere (p. 369), he speaks of keeping mice near the fire where the temperature was 80–90 F. If we assign a temperature of 85 F. (29.4 C.), this would be within the area of thermal neutrality for the mouse (32).

He placed two mice of nearly equal size in similar glass jars each containing 5 oz. (142 cc.) of oxygen, one standing in limewater and the other in common water, for two and one half hours. The oxygen as prepared by Priestley was probably not pure but this does not alter the following discussion. As the glass jars were placed over water at an assumed temperature of 29.4 C., the atmospheres contained about 4 per cent water vapor in equilibrium with the water phase (33). Over limewater this 142.0 cc. of atmosphere was reduced in the proportion of 9 to 5¼, or 82.8 cc. of atmosphere remained. Over water this 142.0 cc. of atmosphere was reduced in the proportion of 9 to 6¾, or 106.5 cc. of atmosphere remained. If we assume that the reduction of atmosphere was based on the utilization of oxygen in respiration and the subsequent absorption by the water and the limewater of the carbon dioxide produced, 23.7 cc. more carbon dioxide was absorbed by the limewater than the water.

No allowance has been made for an increase in the temperature of the confined air as a result of the presence of the mouse. The original temperature (29.4 C.) assigned is an approximation and the jars were placed over water and limewater, which would stabilize the temperature of the air in the jars and reduce any increase to a minimum (34). The amount of nitrogen in the body of the mouse with 10 per cent fat (35) is about 0.13 cc., and in the lungs and airways about 0.16 cc., making a total of somewhat less than 0.3 cc., which for the purposes of these calculations will merely be mentioned (36, 37).

It is possible to calculate the oxygen consumption of his mouse from Priestley's experiments. According to the experiment where the mouse is placed in a jar over limewater, 82.8 cc. of atmosphere remained at the end of the experiment. This reduction from 142.0 cc. means that 59.2 cc. of oxygen was used by the 9.5 Gm. mouse during two and one half hours. Converting this to standard form, 2.5 cc. /Gm./hr. of oxygen compares favorably with the figures given by modern workers (32, 38). The utilization of 59.2 cc. of oxygen by the mouse would produce 47.4 cc. of carbon dioxide, based on a respiratory quotient of 0.8. This amount of carbon dioxide would be absorbed by 100 cc. of saturated limewater at 29 C.

Priestley's experiment had demonstrated that CO_2 was absorbed by limewater, but the fact did not fit in with his theory of respiration. Priestley believed that respiration resulted in the giving out of phlogiston from the body into the atmosphere. Carbon dioxide was not a product of respiration, but the action of the exhaled phlogiston on common air separated carbon dioxide from the common air. And so, after making his experiment, he said:

> The precipitation that is made of fixed air [CO_2] is nothing more than a *circumstance* attending the respiration of common or dephlogisticated air [O_2] . . . and therefore, though the precipitated fixed air be absorbed ever so readily, the remaining air will be but very little the better for it.

To determine if the air was improved by the absorption of the CO_2 by the limewater, he tested for the amount of dephlogisticated air remaining (oxygen) with the nitrous air test. When he found about the same proportion of oxygen over limewater as over water, he concluded that limewater had not increased the amount of oxygen present.

Priestley's book "E. and O.," volume V, was reviewed in the "Critical Review," 1781 (40). The anonymous reviewer says:

> The remainder of this section (XIV) is employed in correcting a most egregious blunder committed by Dr. Ingen-Housz. This is an office which we find him [Priestley] discharging more than once in the course of this volume; and we are apt to think, from our own experience, that if the delicacy of friendship had not restrained him, he might have informed the public of many other errors in that gentleman's [Ingen Housz'] work, which are too glaring to be overlooked by the most common experimentalist; in the present instance however, it is shown, that Dr. Ingen-Housz is wrong, in what the Doctor flatters

himself was a most important discovery, viz., that of making dephlogisticated air serve thirty times longer than when it is used in the common way. It is evident, at first sight, that Dr. Ingen-Housz falls there into Messrs. Kirwan and Cruickshanks' error, viz. that phlogistication changes common into fixed air; and that by passing air after it is breathed through lime-water, you deprive it of its noxious ingredients. The Doctor [Priestley] is not contented with the decisive facts he has enumerated in other cases in opposition to this opinion, but he makes an experiment in which two mice are thrown into convulsions by the frequent respiration of pure air notwithstanding the precautions prescribed by Dr. Ingen-Housz.

After Ingen Housz returned to Vienna he continued his research and the results of these experiments were prepared for publication in the French language. This manuscript was translated into German and published as "Vermischte Schriften" early in 1782. The translator, Niklas Karl Molitor, who helped Ingen Housz with many of the experiments, inserted a long section of his own in which he says (41):

How unfair Mr. Priestley acts throughout, is shown by the severe misgivings with which he takes to task the Abbé Fontana's method to allow patients to breathe in dephlogisticated air [O_2] advantageously. Mr. Ingen-Housz received it first, as one can read in his English edition, page 45, when the entire book was already printed and he wrote expressly that it was known for only a few days. Such a thing thus could be proved neither by the discoverer, nor sufficiently by the publisher, although Mr. Priestley seeks intentionally to cite it. It had all the impression of haste, and the good intention to originate a few trials with it should have excused it. In this regard Mr. Ingen-Housz reports on the same in this book and as I think more exactly and more painstakingly, as has been demanded by Mr. Priestley. The entire section IV of the first essay of this collection from page 43 to 74 is devoted to this, and, as one will see therein, it certainly convinces everyone, that it is no business of which the result should be determined in a few days.

Ingen Housz' Method of Breathing Oxygen and Absorbing CO_2

Ingen Housz' method of oxygen breathing and carbon dioxide absorption was first published in Holland (42). The work was done in Vienna, written in French, and transmitted in March to his friend, J. Van Breda at Delft, for translation. It was published in November, 1781. Figure 1 shows the method of breathing oxygen used by Ingen Housz in 1781. The caption reads: "Arrangement of the manner of breathing dephlogisticated [O_2] air whereby each expiration from the lungs would be purified of contamination by fixed air [CO_2]." Either water or limewater could be placed in the cylinder to absorb CO_2, but Ingen Housz preferred limewater. In this article he also reports experiments using birds placed in oxygen over mercury as a control. He then performed an experiment placing birds in oxygen over plain water, and once he cleared the confined atmosphere of carbon dioxide by means of limewater. He concluded from these researches that "water, and especially limewater, contribute much to purify the air."

These researches were published in German in 1782 (41). The text of the German and Dutch articles is essentially similar. Ingen Housz had originally intended that the French edition of his miscellaneous essays should appear before the German, but it did not appear until 1785, and consequently he was able to make changes and to enlarge the text. The 1785 collection of his essays was entitled "Nouvelle Experiences et Observations sur divers objets de physique" (Paris, 1785) and was dedicated to his friend, Benjamin Franklin under date of November 1, 1783.

FIG. 1. Ingen Housz method of breathing oxygen and purifying the air of carbon dioxide used in 1781. The cylinder contained water but preferably limewater—"And in this way one can again free the dephlogisticated air after each breath of its aerial acid, for which use limewater is preferable to rain water." Reported in the Proceedings of the Batavian Society of Experimental Philosophy at Rotterdam (42). (Illustration taken from Ingen Housz' original work.)

The essay entitled "On the Nature of Dephlogisticated Air, the manner of obtaining it, and of making use of it for the care of the Sick" is of particular interest. Here he repeats the experiments with birds as in the Dutch and German essays, but in addition he reports the following experiments made on himself, using the apparatus shown in figure 2. Experiment 7 is a control breathing 500 cc. oxygen over plain water (20 inspirations and 20 expirations). Then he tested for CO_2 and found the gas in the reservoir contained 4 per cent CO_2. He repeated this experiment (experiment 8) under the same conditions except he substituted limewater "in order to be able to judge with precision if there is really some advantage of employing limewater in preference to pure water and if this advantage is sufficiently consider-

FIG. 2. Method of breathing oxygen and absorbing carbon dioxide used by Ingen Housz in 1783. "Several hours before using the dephlogisticated air one slakes in the water of the bucket a good piece of quicklime." Lacking exact knowledge of Fontana's method of proceeding. . . . "I devised a method which succeeded for me just as well." Nouvelle Researches, 1785. (Illustration taken from Ingen Housz' original work.)

able to make use of it." On testing the gas after breathing he found that it contained 2 per cent CO_2. He says:

One can conclude, I think, from these experiments, that there is some advantage of using limewater in preference to pure water, when one wishes to breath dephlogisticated air in the manner that I have just detailed, for sick persons; for the air used in experiment 8 only contained half as much fixed air [CO_2] as was contained in that of the seventh experiment; although as a matter of fact this advantage would be infinitely less than I had figured it beforehand.

The long preface by Molitor in "Vermischte Schriften" apparently written soon after reading Priestley's criticism of his "Experiments on Vegetables" (1779) was omitted from the French book, "Nouvelle Experiences" (1785) which contains a dedication to Benjamin Franklin. The following two letters explain why the long preface was omitted and serve to illustrate Ingen Housz' feelings toward Priestley:

Letter from Ingen Housz to Franklin:

Vienna, Apr. 24, 1782

. . . You will find a polemical dissertation at the beginning of the book which the translator asked me leave to place in it. I gave him leave for it under condition he should use no expression which could offend my friend Dr. Priestley. But he seems to me to have been rather heated on the subject. Dr. Priestley in my opinion has been in the wrong to attack me without the least provocation, finding at the same time nothing in my book worth his recommendation but that it gave him satisfaction of having found my assiduous attention on the subject of plants; at least smothering in polemic every article by which he could be instructed but in one article, which is that leaves of plants retain their life longer than he thought; but even this he says he might have learned from Mr. Bonnet (Priestley Vol. V, p. 29 and 30). I got letters from England by which I was informed that Mr. Priestley's volume V was so far derogatory to my book, that my whole doctrine seems overturned by it, and that a second edition of my book would not sell. The Critical Review of Septem. 1781, page 180, draws from Dr. Priestley's book a still more humiliating consequence in regard to my book. However I will not quar'l with him about it. I have faith so convincing that I can confound him and his whole manner of thinking about the manner in which the vegetables are subserviant to the animal creation (43).

Letter from Franklin to Ingen Housz:

Passy, 21 June, 1782

Dear Sir: I am sorry that any misunderstanding should arise between you and Dr.————————. The indiscretions of friends on both sides often occasion such misunderstandings. When they produce public altercations, the ignorant are diverted at the expense of the learned. I hope, therefore, that you will omit the polemic piece in your French edition, and take no public notice of the improper behavior of your friend, but go on with your excellent experiments, produce facts, improve science, and do good to mankind. Reputation will follow, and the little injustices of contemporary laborers will be forgotten; my example may encourage you, or else I should not mention it. You know that when my papers were first published, the Abbe Nollet, then high in reputation, attacked them in a book of letters. An answer was expected from me, but I made none to that book, nor to any other. They are now all neglected, and the truth seems to be established. You can always employ your time better than in polemics (44).

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