Early Roots of the Carbon Cycle Concept

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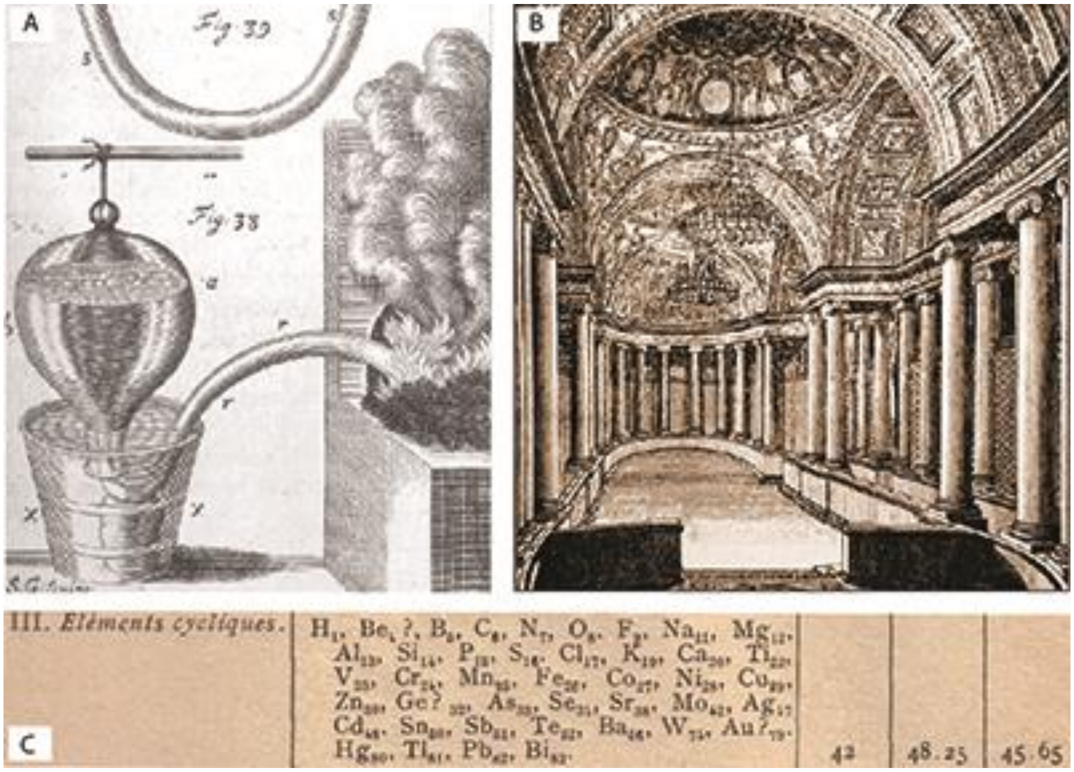
The understanding of how deep carbon processes influence the global carbon cycle is one of the major frontiers of current scientific exploration. However, at the end of the 18^{th} century, the major frontier was to understand how living organisms interact with atmospheric gases. In 1789, the ancient fascination of Democritus and Lucretius with cyclicity in the cosmos [1], revived in the Middle Ages [2], was still alive and well. Scottish physician and geologist James Hutton (1726-1797) had just published his *Theory of the Earth* [3] and was preparing the second edition of this foundational work [4], envisioning a key role for coal and life in sustaining his cyclic understanding [5] of the Earth *machine* [6,7]. Meanwhile, british chemist and philosopher Joseph Priestley (1733-1804) had been pioneering the study of air in relation to living organisms, but a chemical understanding of the underlying phenomena was still lacking [8]. Simultaneously, prominent French chemists Antoine Laurent Lavoisier (1743-1794) and Armand Séguin (1767-1835) [9,10] jointly endeavored to shed new light on the chemical process of animal respiration by making use of rigorous, observation-based chemical principles. They adopted a novel and simplified chemical terminology, including the *simple substances*[11] *oxygen* [12] and *carbon* [11], after the discovery of the compound nature of *carbonic acid* [13] (*carbonic acid* would only be proposed as a *ternary compound,* CO_2, in 1808 [13,14]). Repeating some of Priestley's experiments using inverted bell jars, a procedure originally invented in Britain earlier in the century [15] to collect gases (Fig. 1A), they showed that a guinea pig could not survive long (~75 minutes) when placed within the jar with air; the air soon became vitiated by carbonic acid and thus irrespirable [10]. By adding quicklime to the system and regularly *renovating* the jar with oxygen, they managed to sustain the animal for days [9].



These fundamental observations, gathered in spatially limited and transiently closed vessels, had direct applications for the chemical functioning of public places at that time [10]. Poorly ventilated chambers comparable to their laboratory models were numerous: the *Salle des Machines* of the *Palais des Tuileries* in Paris (Fig. 1B), which had recently been hosting the French comedy, was one of them. Lavoisier showed that the amount of oxygen in the upper part of the room was diminished after a performance, whereas carbonic acid had accumulated. He then proposed that the relative stability of the composition of the air in the lower part of the theater was due to the limited but regular circulation of fresh air from outside [10]. In the absence of this process, and considering a ~640 m^3 crowded show hall, he calculated that at a respiration rate of *ca.* 141 L per hour per person the air in the room would become completely irrespirable after four and a half hours.

In 1792, two years before he would be beheaded (mainly due to his position as a General Farmer (*Fermier général*) in the economic administration of the *Ancien Régime*) Lavoisier reviewed contemporary work on the chemical process of plant nutrition and growth [16] (not yet understood as *photosynthesis*). This topic had attracted flourishing interest throughout Europe, and scientists, among them Priestley [8] and Lavoisier, envisioned that plants had probably been the *"means nature uses to maintain the respirability of air"* [16] at the Earth scale. Animal respiration and plant nutrition would thus act as counterbalancing processes, mimicking, at the larger scale, Lavoisier and Séguin's laboratory devices of adding quicklime to, and restoring the air in, the bell jar. This had direct consequences for the circulation of carbon on Earth. In his ultimate exposition, laid out in an autographed manuscript of 1793 dedicated to coal [17], Lavoisier introduced quantitative reasoning on the global transfer of carbon, estimating the amount of this *simple substance* in crustal carbonates:

"We can conceive what immense quantity of carbon is sequestered in the womb of the Earth since marbles, limestones, and calcareous earths contain about 3/10th and sometimes 1/3 of their weight in fixed air, and that this latter is composed for 28/100thof his weight of carbon; then, it is easy to conclude that the calcareous rocks contain 8 to 9 pounds of true carbon by quintal. […] We will not follow here the change in form that carbon takes by passing from the mineral to the plant and to the animal kingdoms. We would throw ourselves into chemical discussions that are outside of the scope of this article." [17]

Less than a year later he would be sentenced to death, and his manuscript was never published. Nevertheless, along with entailing a major revolution in chemical methodology [18,19], application of these concepts and methods to Earth and life processes motivated a vast amount of work, ultimately shedding more and more light on these basic natural phenomena. Other key contributors to the development of carbon *rotation* [20] or *cycle* [21] included N.T. De Saussure (1767-1845), J. Dalton (1766-1844), J.J. Berzelius (1779-1848), H. Davy (1778-1829), G. Bischof (1792-1870), J.B. Boussingault (1802-1887), J.J. Ebelmen (1814-1852), J. Liebig (1803-1873), V.M. Goldschmidt (1888-1947) and W. Vernadsky (1863-1945) [7,22]. The latter coined the term *cycles* in 1923 [21] as describing the *geochemical history* of *cyclic*, or *organogenic*, elements, including carbon (Fig. 1C).

In hindsight, the representations of the carbon *rotation* and *cycle* have emerged as byproducts of centuries of international efforts aimed at understanding the complex relationship between life and Earth processes in increasingly large but finite systems [7]. This, in essence, is still the major challenge of numerous current scientific efforts. Time passes, and however deep paradigm shifts affect our understanding of − and relation to − past achievements, we cannot respond with anything less than awe at the works and words of these early explorers who paved the ground on which we now boldly stand. For this reason alone, a grateful, retrospective look at this literature is worth a few minutes of our precious time.

- A reprint of the author's paper (ref 7) dealing with some aspects of this subject can be provided upon request by Matthieu Galvez (mgalvez@ciw.edu) or Jérôme Gaillardet (gaillardet@ipgp.fr). The latter I especially thank for a fruitful collaboration during the work leading to our joint contribution on this fascinating issue. The author warmly acknowledges the help provided by Katie Pratt and Shaun Hardy during the preparation of this column. Finally, I thank my colleagues S. Lobanov, C Shiffries, A. Karandikar, C. Glein, S. Shirey and G. Cody for their useful comments and for their interest.

**Caption: A. The original bell jar apparatus designed and drawn by Stephen Hales (ref 15) to collect gases during combustion experiments. Lavoisier and Séguin used a modified version of it to perform their own investigations on the respirability of air. B. View of the Salle des Machines at the Palais des Tuileries, or «Théâtre des Tuileries», in which Lavoisier performed a survey of the respirability and circulation of air in times of high attendance. Iconography from the first* *edition of the Architectonographie des théâtres de Paris, 1821, by A. Donnet and J. Orgiazzi. C. Portion of Table 3 of La géochimie by W. Vernadsky (edition of 1923), showing the classification of carbon, C6, among the «cyclic» elements.*

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