

These differences agree with the ones measured in ordinary hydrogen at the level of 16 parts per billion. The authors used their results to estimate the fine-structure splitting (the $2P_{1/2}$ – $2P_{3/2}$ energy difference) in antihydrogen, with an uncertainty of 0.5%. This value is again in good agreement with the one for ordinary hydrogen.

In 2018, the ALPHA Collaboration measured the energy gap between the 1S and 2S states in antihydrogen¹¹ to one part in 10^{12} . In the current work, the authors combined this result with their measurement of the 1S– $2P_{1/2}$ energy difference to provide an estimate of the Lamb shift in antihydrogen. This value has an uncertainty of 11% (or 3.3%, when the fine-structure splitting in ordinary hydrogen is used in the analysis).

Over the past few years, high-precision laser spectroscopy of antihydrogen has become possible, and the ALPHA Collaboration has achieved spectacular progress. An examination of several transitions in antihydrogen would enable targeted tests of CPT symmetry, quantum electrodynamics and the standard model of particle physics. For example, a measurement of the Lamb shift with an uncertainty of less than one part in 10^4 would allow the antiproton charge radius to be determined¹². Moreover, improved measurements of the energy gap between magnetic substates in antihydrogen would provide detailed information about the magnetic structure of the antiproton¹³.

The laser used for spectroscopy in the current work will, in the future, be used for cooling of antihydrogen by inducing $1S$ – $2P_{1/2}$ and $1S$ – $2P_{3/2}$ transitions. Such cooling would greatly improve the achievable precision in all spectroscopy experiments on antihydrogen. In addition, ultracold antihydrogen can be used to study the effect of gravity on these atoms¹⁴. Cold antihydrogen thus promises many cool results.

Randolf Pohl is at the Institute of Physics, University of Mainz, 55128 Mainz, Germany. e-mail: pohl@uni-mainz.de

1. The ALPHA Collaboration. *Nature* **578**, 375–380 (2020).
2. Hagley, E. W. & Pipkin, F. M. *Phys. Rev. Lett.* **72**, 1172–1175 (1994).
3. Parthey, C. G. *et al.* *Phys. Rev. Lett.* **107**, 203001 (2011).
4. Bezginov, N. *et al.* *Science* **365**, 1007–1012 (2019).
5. Schwingenheuer, B. *et al.* *Phys. Rev. Lett.* **74**, 4376–4379 (1995).
6. Dehmelt, H., Mittleman, R., Van Dyck, R. S. Jr & Schwinberg, P. *Phys. Rev. Lett.* **83**, 4694–4696 (1999).
7. Hori, M. *et al.* *Nature* **475**, 484–488 (2011).
8. Smorra, C. *et al.* *Nature* **550**, 371–374 (2017).
9. Lamb, W. E. Jr & Retherford, R. C. *Phys. Rev.* **72**, 241–243 (1947).
10. Capra, A. & ALPHA Collaboration. *Hyperfine Interact.* **240**, 9 (2019).
11. Ahmadi, M. *et al.* *Nature* **557**, 71–75 (2018).
12. Crivelli, P., Cooke, D. & Heiss, M. W. *Phys. Rev. D* **94**, 052008 (2016).
13. Zemach, A. C. *Phys. Rev.* **104**, 1771–1781 (1956).
14. The ALPHA Collaboration & Charman, A. E. *Nature Commun.* **4**, 1785 (2013).

In retrospect

30 years of the iron hypothesis of ice ages

Heather Stoll

In 1990, an oceanographer who had never worked on climate science proposed that ice-age cooling has been amplified by increased concentrations of iron in the sea – and instigated an explosion of research.

Thirty years ago this month, John Martin proposed a solution to one of the biggest mysteries of Earth's climate system: how was nearly one-third of the carbon dioxide in the atmosphere (about 200 gigatonnes of carbon) drawn into the ocean as the planet entered the most recent ice age, then stored for tens of thousands of years, and released again as the ice sheets melted? These large natural cycles in atmospheric CO₂ levels (Fig. 1a) were revealed in 1987 by an analysis of ancient air bubbles trapped in the first long ice cores taken from the Antarctic ice sheet¹. Martin recognized that iron was a key ingredient that could have transformed the surface ocean during glacial times. His landmark iron hypothesis², published in *Paleoceanography*, described a feedback mechanism linking climatic changes to iron supply, ocean fertility and carbon storage in the deep ocean.

Two hundred gigatonnes is a lot of carbon to periodically withdraw from and release to the atmosphere. In the 1980s, a handful of models (see ref. 3, for example) had shown that an increase in biomass production in polar ocean regions was the most effective process for removing so much atmospheric carbon. Photosynthetic organisms in the surface ocean convert CO₂ from the atmosphere into biomass, much of which is subsequently broken down into CO₂ again by other organisms and returned to the atmosphere. But part of the biomass sinks into the deep ocean, which therefore effectively serves as a large storage reservoir of dissolved CO₂. This mechanism of CO₂ removal is called the biological pump.

However, biomass production requires not only CO₂, but also other nutrients to build lipids, proteins and enzymes. Researchers were struggling to ascertain how the ocean's abundance of key nutrients, such as nitrates or phosphates, might have increased during glacial times to fuel a stronger biological pump.

Martin argued that iron is another nutrient that limits the biological pump. He suggested that the modern marine ecosystem

of the Southern Ocean around Antarctica is starved of iron, and therefore relatively low in biomass, despite having abundant nitrates and phosphates. But during glacial times, strong winds over cold, sparsely vegetated continents could have transported large amounts of iron-bearing dust into this ocean (Fig. 1b). Martin reasoned that this dust could have fertilized marine ecosystems and strengthened the biological pump, so that more carbon was transferred into the deep ocean, lowering atmospheric CO₂ levels.

Around the time of publication, evidence for high dust delivery during glacial periods had just emerged from studies of deep Antarctic ice cores⁴. But there were no reliable measurements of dissolved iron in the Southern Ocean that could confirm that its surface waters are iron-starved in modern times, or data supporting the proposal that delivery of iron-rich dust would make a difference to ocean productivity. It was clear, however, that large patches of the world's ocean had much lower quantities of biomass than would be expected on the basis of the concentrations of key nutrients such as nitrates and phosphates. But many researchers argued that this was due to natural overgrazing of algae by herbivores⁵.

The idea that modern algal growth is limited by iron availability had, in fact, been proposed⁶ in the 1930s, but had been incorrectly discounted by oceanographers – who had measured plenty of iron in seawater samples collected from the waters around their iron ships⁷. Martin was one of the first oceanographers to implement painstaking procedures to avoid the contamination of samples and to determine that iron concentrations in the north Pacific Ocean were extremely low⁷, certainly low enough to curtail biomass production.

Despite the initial scepticism that greeted the iron hypothesis, 12 separate experiments⁸ were carried out between 1993 and 2005 in which around 300–3,000 kilograms of dissolved iron were injected into small patches of the Southern Ocean, the equatorial Pacific

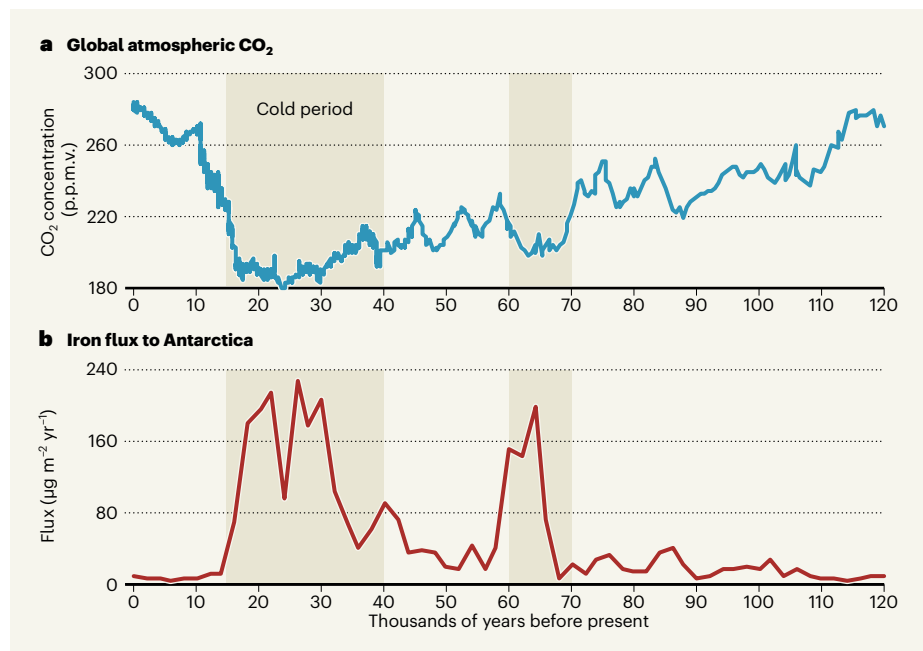


Figure 1 | The anti-correlated data that inspired the iron hypothesis. **a**, Measurements of air bubbles trapped in cores drilled from the Antarctic ice sheet show that atmospheric levels of carbon dioxide were significantly lower during the coldest periods (shaded regions) than during modern times (data from ref. 16; CO₂ concentrations are shown in parts per million by volume; p.p.m.v.). **b**, The ice-core records also reveal that more iron was transported to the Southern Ocean in wind-blown dust during the coldest periods than during warmer times (data from ref. 17; iron flux is measured in micrograms per square metre per year). In 1990, Martin² hypothesized that the increased levels of iron in the Southern Ocean during the coldest periods fertilized the growth of photosynthetic microorganisms in the surface Southern Ocean, which therefore produced more biomass from CO₂. This, in turn, would have increased the strength of the biological pump, a mechanism that sequesters some of the biomass (and the carbon within it) in the deep ocean. Martin proposed that the stronger biological pump explains why so much atmospheric CO₂ is drawn into the ocean during cold times.

Ocean and the north Pacific. The biomass of algae increased wherever iron was added, as biological production surged.

Unfortunately, Martin died mere months before the first of these experiments, and did not witness the ocean-scale confirmation of his hypothesis, nor the internationally coordinated campaign to measure iron geochemistry throughout the world's oceans⁹ – which confirmed iron limitation and revealed the intricate strategies used by marine ecosystems to acquire and recycle iron¹⁰.

Earth scientists also tried to test the iron hypothesis computationally using simple ocean models. They used the changes in the dust-accumulation rate recorded in ice cores as input to simulate changes in iron delivery to the Southern Ocean, and data from the experimental iron fertilizations to calculate how this iron could affect algal growth and the biological pump. Such models could reproduce the timing and magnitude of about half of the observed decrease in atmospheric CO₂ levels during glacial periods¹¹. Iron fertilization is therefore clearly an important process that causes atmospheric changes, but might not be the only one.

Finding data to prove that biological production had been higher during glacial

was a harder task – after all, the ecosystem during the most recent glacial period (about 20,000 years ago) is long dead. One possible solution was to extract cores from sediments piled on the sea floor, to see whether the mineral skeletons of algae accumulated faster during glacial times than in the modern era. However, the results were often ambiguous¹², for several reasons: many algae don't produce a preservable skeleton; numerous factors determine what proportion of biological remains is preserved on the sea floor; and the location of biological production changes through time as ocean fronts and sea-ice positions migrate.

Fortunately, Martin² and others¹³ had anticipated an alternative, global-scale test of the biological pump during glacial times. If more biomass reached the deep ocean during glacial, then deep-sea microorganisms would use up more oxygen as they consumed it, decreasing the concentration of oxygen in deep waters. Evidence of deep-ocean oxygen depletion would therefore be indicative of a strong biological pump.

Martin recognized that the presence of certain microfossils in glacial-age sediments meant that the deep ocean had not become completely devoid of oxygen during glacial.

But although this evidence crudely constrained estimates of the degree to which iron fertilization might have enhanced productivity during glacial, it could not be used to determine whether levels of deep-ocean oxygen were lower than during modern times. Since then, analysis of more-sensitive geochemical records indicates that the oxygen concentration in bottom waters did decrease during glacial times¹⁴. This provides the strongest confirmation yet of the large-scale accumulation of carbon in the deep ocean during glacial periods owing to a stronger biological pump.

Slower rates of mixing between the deep and shallow oceans could also have enhanced the biological pump during glacial. The latest generation of climate models in which the ocean and atmosphere are coupled can test the contribution of the multiple processes that could have resulted in a reduction in bottom-water oxygen levels. Such models indicate that mixing rates can account for only half of the observed deep-ocean storage of CO₂ during the glacial period, and that iron fertilization of the Southern Ocean is the major cause of the extra CO₂ storage observed¹⁵.

Martin concluded his paper by saying that iron availability “appears to have been a player” in strengthening the biological pump during glacial cycles, but that the size of its role remained to be determined. Thirty years later, the evidence convincingly shows that iron fertilization of the Southern Ocean was indeed a leading actor in this global-climate feedback.

Heather Stoll is in the Department of Earth Sciences, ETH Zurich, 8092 Zurich, Switzerland.
e-mail: heather.stoll@erdw.ethz.ch

- Barnola, J. M., Raynaud, D., Korotkevich, Y. S. & Lorius, C. *Nature* **329**, 408–414 (1987).
- Martin, J. H. *Paleoceanography* **5**, 1–13 (1990).
- Sarmiento, J. L., Toggweiler, J. R. & Najjar, R. *Phil. Trans. R. Soc. Lond. A* **325**, 3–21 (1988).
- De Angelis, M., Barkov, N. I. & Petrov, V. N. *Nature* **325**, 318–321 (1987).
- Cullen, J. J. *Limnol. Oceanogr.* **36**, 1578–1599 (1991).
- Hart, T. J. *Discov. Rep.* **VIII**, 1–268 (1934).
- Gordon, R. M., Martin, J. H. & Knauer, G. A. *Nature* **299**, 611–612 (1982).
- Boyd, P. W. *et al. Science* **315**, 612–617 (2007).
- Anderson, R. F. & Henderson, G. M. *Oceanography* **18**, 76–79 (2005).
- Tagliabue, A. *et al. Nature* **543**, 51–59 (2017).
- Watson, A. J., Bakker, D. C. E., Ridgwell, A. J., Boyd, P. W. & Law, C. S. *Nature* **407**, 730–733 (2000).
- Kohfeld, K. E., Le Quére, C., Harrison, S. P. & Anderson, R. F. *Science* **308**, 74–78 (2005).
- Boyle, E. A. *Nature* **331**, 55–56 (1988).
- Jaccard, S. L., Galbraith, E. D., Martínez-García, A. & Anderson, R. F. *Nature* **530**, 207–210 (2016).
- Yamamoto, A., Abe-Ouchi, A., Ohgaito, R., Ito, A. & Oka, A. *Clim. Past* **15**, 981–996 (2019).
- Bereiter, B. *et al. Geophys. Res. Lett.* **42**, 542–549 (2015).
- Wolff, E. W. *et al. Nature* **440**, 491–496 (2006).

This article was published online on 17 February 2020.