Isotopic signatures

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Abstract

Isotopes record human influence on the Earth System, providing evidence for the Anthropocene. Lead and sulfur isotopes detail pollution histories going back millennia. Carbon and nitrogen isotopes show substantial change since the Industrial Revolution, and especially since the 1950s, related to increased fossil fuel consumption and fertilizer production. Boron isotopes record ocean acidification related to CO₂ emissions. Radioisotopes, e.g. plutonium, have been dispersed across the Earth since the 1950s due to nuclear weapons testing and can be used as stratigraphic markers for the time that carbon and nitrogen isotopes, and non-isotope proxies, show increased human influence on the Earth System.

Introduction

The Anthropocene is the period during which human activity has had a discernible impact on the functioning of the Earth System, causing changes outside of natural variability (Steffen et al., 2007). There is intense debate around whether or not to define the Anthropocene as a new geological time interval, and if so from what date. The concept of the Anthropocene is no longer restricted to Earth Science and has far-reaching cultural and political implications. Humans have influenced the functioning of different spheres of the Earth System: the atmosphere (e.g. increasing greenhouse gas concentrations that have contributed to global warming), the biosphere (e.g. increasing...
extinction rates and changes in biogeochemical cycles), the cryosphere (e.g. anthropogenic global warming contributing to the melting of ice sheets in some parts of the world), the hydrosphere (e.g. ocean acidification) and the lithosphere (e.g. draining of aquifers leading to earthquakes). These spheres are connected, for example human-forced changes in the composition of the atmosphere have led to changes in the hydrosphere (ocean acidification) and biosphere (associated impacts on coral ecosystems), so the Earth System as a whole can be said to be influenced by human actions. In this chapter, we focus on how isotope geochemistry can trace some of this human influence.

Isotopes are different forms of an element: they have the same number of protons but a different number of neutrons. Stable isotopes will not decay into other isotopes, whereas radioactive isotopes will. For example, carbon-12 has 6 protons and 6 neutrons and carbon-13 has 6 protons and 7 neutrons and both are stable isotopes, whereas carbon-14 (a radioactive isotope) has 6 protons and 8 neutrons and will decay (Figure 1). Isotope geochemistry offers a useful contribution to the Anthropocene debate because anthropogenic impacts on the Earth, such as heavy metal pollution and carbon emissions, often have isotope signatures that are distinct from those that would be expected if natural processes were leading to the same impacts. Isotopes occur in, for example, marine sediments, lake sediments and polar ice. By looking at how isotope ratios change from the top of a sediment or ice core to the bottom, it is possible to reconstruct the intensity of human impacts on the Earth System from the present day back through time. Whilst isotopes do not record every aspect of the Anthropocene, they can contribute important information to the debate. Where they come in particularly useful is in taking our knowledge back further in time than our observations extend, putting human impacts into long-term context, to show when different human
influences on the Earth System began across the planet, and the extent to which the various activities are causing variability outside of natural ranges.

Some of the most commonly used isotope ratios are in abundant elements such as carbon and nitrogen. The carbon isotope ratio ($\delta^{13}C$) is the ratio of carbon-13 to carbon-12; the nitrogen isotope ratio ($\delta^{15}N$) is the ratio of nitrogen-15 to nitrogen-14; the sulfur isotope ratio ($\delta^{34}S$) is the ratio of sulfur-34 to sulfur-32; and the boron isotope ratio ($\delta^{11}B$) is the ratio of boron-11 to boron-10. Delta ($\delta$) values are given in per mil ($‰$, parts per thousand) relative to the international scales of VPDB, AIR, VCDT and (usually) SRM 951 respectively. Lead isotopes are measured relative to a variety of standards, for example SRM 981. $\delta^{14}C$ is the per mil deviation of the carbon-14 to carbon-12 ratio relative to the oxalic acid standard, corrected for fractionation. The abundance of radioisotopes (radioactive isotopes) such as cesium-137 and plutonium-239 are measured in becquerel (Bq) units, with one Bq representing one decay per second. For more details on notation and standardization, see Dean et al. (2014) and references therein.

Here we discuss the isotope evidence for the global impact of humans on several Earth systems: (1) contamination of the environment by lead through our history of mineral exploitation particularly over the past few millennia, and by sulfur from fossil fuel burning especially since the 1950s; (2) alteration of the global nitrogen cycle due to fossil fuel burning and the production of artificial fertilizers, especially since the 1950s; (3) carbon isotopes tracking greenhouse gas emissions from fossil fuel combustion, the industrialization of agricultural practices and extensive forest clearance, leading to
global warming since the Industrial Revolution but especially since the 1950s; and (4) absorption of atmospheric carbon by the oceans, which has led to ocean acidification, especially since the 1950s, at what likely amounts to the greatest rate for ~50 million years. We also demonstrate how radioisotopes could be useful as stratigraphic markers to define the beginning of the Anthropocene in the geological record (e.g. carbon-14, cesium-137 and plutonium-239).

**Isotope evidence for the Anthropocene**

*Records of mineral exploitation*

Humans have been mining and using heavy metals for thousands of years, and the subsequent liberation of heavy metal particles into the atmosphere means that the pollution can spread thousands of kilometers from the source. Lead was one of the first metals to be mined by humans. Different lead ores in different parts of the world can have different lead isotope ratios, so the lead isotope ratio can potentially be used to pinpoint the origin of lead pollution found at a particular location. For example, isotopes have shown that between ~150 BC and AD 50, the majority of the lead particles deposited on Greenland ice came from Roman lead mines in southern Spain. Nowadays, lead isotopes show that a large proportion of the lead pollution in Greenland comes from China. In addition, trends in $^{206}\text{Pb}/^{207}\text{Pb}$ have been used to trace pollution changes through time. For example, in Sweden, the background (i.e. natural) $^{206}\text{Pb}/^{207}\text{Pb}$ is around 1.5, while atmospheric lead pollution derived from smelting, leaded petrol and burning of coal has a $^{206}\text{Pb}/^{207}\text{Pb}$ value of ~1.2 (Renberg et al., 2002). The particles of lead released by these natural and human sources are deposited in lake sediments, and by looking at how the lead isotope ratio changed back through time, the history of
anthropogenic lead pollution in Sweden has been reconstructed. There was a decline in the $^{206}\text{Pb}/^{207}\text{Pb}$ ratio in Roman times (to ~1.46), indicating increased pollution from anthropogenic activities, then an increase to higher values in the Dark Ages ~AD 500-800 (~1.50) when lead mining and smelting activities must have decreased. Minimum values (~1.22) were reached in the 1970s when leaded petrol consumption peaked (Figure 2) (Renberg et al., 2002). In recent times, with the phasing out of leaded petrol in Europe, there has been an increase in $^{206}\text{Pb}/^{207}\text{Pb}$ as anthropogenic sources of lead pollution have diminished (currently ~1.28). The fact that particles of lead have been liberated into the atmosphere by humans, and then carried hundreds and thousands of kilometers from source to pollute Sweden and Greenland, could be used to support the contention of Ruddiman et al. (2011) that human influences on the environment were occurring millennia before the Industrial Revolution. However, although the anthropogenic lead signal may be near hemispheric in Roman times, localized heavy metal pollution in a certain parts of the world might not be considered to constitute a change in the functioning of the Earth System.

In addition to lead, sulfur isotopes can be used to track pollution because, as with lead isotopes, natural (e.g. volcanic eruptions and forest fires) and anthropogenic (e.g. burning of coal in power stations) sources of sulfur often have different isotope ratios: anthropogenic sources of sulfur emissions into the atmosphere are often low in $\delta^{34}\text{S}$ compared to natural sources. $\delta^{34}\text{S}$ of sulfate in rainwaters can therefore be used to track changes in anthropogenic sulfur emissions and acid rain through time. In the second half of the twentieth century, there was a particular need to understand the sources and changes in sulfur emissions from power stations as concern over acid rain gained
prominence in Europe and North America. Starting with the Convention on Long-Range Transboundary Air Pollution, signed by European and North American countries from 1979 onwards, a series of agreements have led to large decreases in SO₂ levels in the European Union, as traced by increases in δ²⁸S in rain at some monitoring stations (Dean et al. (2014) and references therein). As with lead pollution, the fact that sulfur emissions have started to decline across much of the planet shows that some anthropogenic impacts on the environment have already peaked, although as with lead pollution, sulfur pollution is far from being the most compelling indicator of the Anthropocene.

Changes to the global nitrogen cycle

In contrast to lead and sulfur, there has been a continued increase in anthropogenic nitrogen emissions, showing how efforts to limit anthropogenic nitrogen emissions (e.g. nitrous oxides and ammonia) have met with limited success. Fossil fuel emissions and the artificial production of fertilizer have increased the amount of reactive nitrogen (nitrogen compounds such as nitrogen oxides that support biological growth) in the atmosphere, and this is thought to constitute the largest change to the global nitrogen cycle for 2.5 billion years (Canfield et al., 2010). At present, humans fix more N₂ to reactive nitrogen than natural processes do. In 2008 alone, the Haber-Bosch method of producing fertilizer added 9.5×10¹² mol of reactive nitrogen and fossil fuel burning 1.8×10¹² mol to the Earth System (Canfield et al. (2010) and references therein). Crops like wheat and maize do not use the nitrogen compounds from fertilizers efficiently, so a lot of the reactive nitrogen is wasted and lost to the atmosphere or washed off fields, leading to enhanced aquatic productivity and sometimes to eutrophication of rivers, lakes and coastal waters. Isotopes allow these changes in the global nitrogen cycle to be
tracked. Recent declines in $\delta^{15}$N of certain nitrogen compounds such as nitrates can be observed in many geological archives. It is thought that this decline is the result of the release of isotopically-depleted nitrogen from anthropogenic sources: anthropogenic reactive nitrogen sources such as fertilized soils and fossil fuel emissions (Hastings et al., 2009). More recently, however, other explanations for the decline in $\delta^{15}$N in nitrates in particular have been proposed such as changes in the type of fossil fuels being burnt and changes in atmospheric acidity caused by the sulfate and nitrate emissions (e.g. Geng et al., 2014). More research is required to establish better the causes of the $\delta^{15}$N decline.

Nevertheless, the clear trend in $\delta^{15}$N records appears to be mostly the result of anthropogenic activity. In nitrate (NO$_3^-$) in ice cores from Greenland (Hastings et al., 2009) and in organic matter from remote lakes in North America and the Arctic (e.g. Wolfe et al., 2013), there have been declines in $\delta^{15}$N from ~AD 1850, but as with $\delta^{13}$C (next section) it is really after ~AD 1950 that the trend becomes pronounced (Figure 3). In Sky Pond Lake in the US Rockies, the changes since the 1950s are without precedent for at least the last 14,000 years (Wolfe et al., 2013). The decline in nitrogen isotopes from ~AD 1850 was probably caused by the increase in the burning of fossil fuels, with the acceleration ~AD 1950 caused by both artificial fertilizer production and fossil fuel burning.

These records provide a clear example of how important isotope data are in contextualizing human impacts on the Earth System. By using $\delta^{15}$N, we can extend our knowledge of changes in the nitrogen cycle back much further in time than we otherwise would be able to do, to show that changes since the 1950s seem to be outside that of natural variability.
Changes in the global carbon cycle

There have also been substantial changes to the global carbon cycle because of human activities. Small rises in atmospheric methane (CH$_4$) and carbon dioxide (CO$_2$) are seen in gas bubbles in ice cores from ~8,000 years ago and have been linked to wetland expansion related to rice production and to widespread forest clearance (Ruddiman et al., 2011). Ruddiman et al. even argue that these changes might have delayed the onset of the next glacial, which if true could constitute an anthropogenic impact on the functioning of the Earth System. Carbon isotopes have been used to support this hypothesis. $\delta^{13}$C of atmospheric CH$_4$ ($\delta^{13}$CH$_4$) from bubbles within ice cores from the late Holocene have values ~$-48\%$. Some have argued that such low values of $\delta^{13}$CH$_4$ could be accounted for by increased delivery of isotopically low carbon from natural wetlands. However, Ruddiman et al. (2011) suggest this would not have been likely because of the drying of northern monsoonal regions and the cooling of boreal regions in the late Holocene, which would have meant reduced CH$_4$ emissions from natural wetlands. Instead, they suggest that low $\delta^{13}$CH$_4$ could be explained by emissions from rice paddies and livestock and anthropogenic burning of grasses. In terms of $\delta^{13}$CO$_2$, Ruddiman et al. (2011) argue that carbon burial in boreal peat is underestimated and therefore greater human emissions related to forest clearance are required to balance the $\delta^{13}$CO$_2$ budget (see Dean et al. (2014) and references therein for more details). The ‘Early Anthropogenic Hypothesis’ of Ruddiman is contentious, with many people arguing that human influence on the Earth was not outside of natural variability so far back into the past. The carbon cycle is complex, and it needs to be better understood.
(better accounting for the natural and anthropogenic drivers of carbon isotopes in carbon dioxide and methane is key to this) in order to robustly test the hypothesis that human actions thousands of years ago did indeed delay the next glacial.

What is not in doubt is that since the nineteenth century there has been a substantial decline in atmospheric $\delta^{13}$CO$_2$ concomitant with the increase in atmospheric CO$_2$ concentrations, with the trend to lower values accelerating after ~AD 1950 (Figure 3). Since the 1950s, this has been recorded by direct measurements of atmospheric CO$_2$ at sites such as the Mauna Loa monitoring station in Hawaii. Further back into the past, the record is locked away as bubbles of CO$_2$ in ice cores or can be calculated using the carbon in geological archives such as tree rings, corals and foraminifera in marine sediments. The decline in $\delta^{13}$CO$_2$ and $\Delta^{14}$CO$_2$ is known as the Suess Effect and is thought to be linked to the burning of fossil fuels with low $\delta^{13}$C and $\Delta^{14}$CO$_2$. Fossil fuels are made up of the remains of organisms, mostly plants, that lived millions of years ago. When plants grow, they take up carbon from their surroundings, and plants preferentially take up $^{12}$C over $^{13}$C, so have low $\delta^{13}$C, because the bonds of the lighter isotope are more easily broken down to participate in organic reactions. As a result, CO$_2$ released from the burning of fossil fuels in power stations, vehicles, etc. contains on average 2% less $^{13}$C per mol than atmospheric CO$_2$, and thus its release back into the atmosphere leads to a decline in atmospheric $\delta^{13}$CO$_2$. Fossil fuel carbon also has very little $^{14}$C (radioactive carbon) because the time between when the organisms died and were laid down in sediments and when humans started to burn them is many thousands of half-lives of $^{14}$C. Figure 4 shows data from the Mauna Loa monitoring station, with $\delta^{13}$CO$_2$ declining (~7.6‰ in 1980 to ~8.4‰ in 2014) as CO$_2$ concentrations in the atmosphere have risen (316 ppm in 1959 to 401 ppm in 2015) (Keeling et al., 2005; updated). In terms of $\Delta^{14}$CO$_2$, there was a decline in the first half of the twentieth
century as fossil fuel emissions increased, followed by an increase in the 1950s and 1960s as a result of the vast amounts of neutrons produced by atmospheric thermonuclear bomb testing that increased $^{14}$C production in the atmosphere. There has since been a decline again to the present day as a result of the reduction in atmospheric nuclear tests and the continued increase in fossil fuel emissions. The $\delta^{13}$C and $\Delta^{14}$CO$_2$ changes in the atmosphere have been key in enabling the Intergovernmental Panel on Climate Change to state that there is a “very high confidence” that the main contributor to the observed increase in CO$_2$ concentrations in the atmosphere since the nineteenth century has been the human burning of fossil fuels, rather than any potential natural causes. This CO$_2$ rise is believed to be responsible for a large proportion of recent global warming.

As well as causing changes in the functioning of the atmosphere, the release of CO$_2$ from anthropogenic activities has led to ocean acidification as large quantities of CO$_2$ have been absorbed from the atmosphere by the oceans (~30% of the total according to estimates from the Intergovernmental Panel on Climate Change). Ocean acidification has the potential to reduce coral calcification and threaten the survival of coral ecosystems, and this could produce a distinctive event in the future fossil record. For the last hundred years or so, pH records measured directly from seawater samples can be used to assess the acidification trend, but to extend the record further back in time proxies such as boron isotopes can be used. Calcifying organisms, such as foraminifera and corals, incorporate the borate ion from seawater as they grow and $\delta^{11}$B is pH dependent. Since the Industrial Revolution, as CO$_2$ levels have risen, there has been a decline in the pH of the oceans by about 0.1 (in what could be the fastest rate of ocean
chemistry change in the last 50 million years), charted by a decline in $\delta^{11}B$. This is to be expected, as $\delta^{11}B$ from ocean sediment cores has shown that there was ocean acidification when atmospheric CO$_2$ concentrations rose during previous interglacials (warm periods). This knowledge gained from boron isotopes with regards how ocean acidification and atmospheric CO$_2$ concentrations were linked in the past allows us to predict that ocean acidification will increase as CO$_2$ concentrations continue to rise.

In summary, changes in carbon isotopes record the human influence on the carbon cycle caused by the release of vast quantities of carbon into the atmosphere because of fossil fuel burning. Changes in boron isotopes record another influence of the rise in atmospheric CO$_2$ concentrations: ocean acidification. As Figure 3 shows, changes in $\delta^{13}$CO$_2$ and $\delta^{15}$N are similar, with both showing trends towards lower values through the nineteenth century but with an acceleration after the Second World War at the time of the ‘Great Acceleration’ (Steffen et al., 2007, 2015) in human activity.

A golden spike to define the start of the Anthropocene

As well as providing evidence that humans have influenced the Earth System, and thus contributing to the debate vis-à-vis whether the Anthropocene should be defined as an official geological time interval and if so at what time, isotopes may also provide the stratigraphic marker that could define the start of the Anthropocene. While we have discussed the Anthropocene from an Earth System Science perspective, its formal adoption by the geological community needs to be grounded in stratigraphy. Zalasiewicz et al. (2015) have suggested that the start of the Anthropocene could be defined by a Global Standard Stratigraphic Age: by assigning a point from the human calendar at the time when human influences on the Earth System reached a significant
level. However, during the current Eon, the standard practice for defining geological
time intervals is the use of a golden spike (i.e. a Global Stratotype Section and Point:
GSSP). A GSSP is a single reference point found in the geological record at a specific
location on Earth that is the physical manifestation of a global change that marks the
beginning of the time interval (Waters et al., 2015). For example, the beginning of the
Paleogene 66 million years ago is defined by an iridium-enriched layer found in
sediments in Tunisia related to the meteorite impact that is believed to have contributed
to the extinction of the dinosaurs. The iridium layer is found around the world but the
reference point chosen as the GSSP is in Tunisia. There are a number of radioisotopes
that show either clear anthropogenic alteration to natural trends (carbon-14) or whose
occurrence on Earth is almost entirely the result of human actions (for example, cesium-
137 and plutonium-239). What is important to understand here is that whatever marker
in the geological record is chosen to set the GSSP, it does not necessarily have to itself
be an indicator of substantial human influence on the Earth System; it just has to be
something unique and easily identifiable that occurs around the time other isotope and
non-isotope proxies have been used to establish where the Anthropocene should start.
Cesium-137 and plutonium-239 first appear at detectable levels in the early 1950s,
especially after high yield atmospheric thermonuclear weapons testing (as opposed to
atomic bomb testing and use over Japan) began. Peak levels were reached in AD 1963/4
due to the large number of atmospheric nuclear tests that were carried out in AD 1962
before the Partial Nuclear Test Ban Treaty came into force (Figure 5). $^{14}$C abundance
also starts to increase in the early 1950s; $^{14}$C is produced naturally in the atmosphere
through the interaction of neutrons with nitrogen atoms, but the production rate was
increased by the release of additional neutrons by atmospheric nuclear tests.

<Insert Figure 5 here>
These radioisotopes were deposited in geological records, from tree rings to lake sediments, across the Earth and thus provide a precise stratigraphic point in geological archives at around the time of the ‘Great Acceleration’ in human activity when isotope and non-isotope proxies show an acceleration in human impacts on the Earth System (Steffen et al., 2007, 2015). Plutonium-239 is considered by many to be potentially the most suitable to use as a GSSP since its long half-life (24,110 years) and low solubility mean it will persist in the geological record for longer than many other radioisotopes (Waters et al., 2015), being detectable even by current mass spectrometry for about 100,000 years into the future. Its first real appearance in the geological record in the early 1950s around the start of the ‘Great Acceleration’ could serve as a GSSP.

Conclusion

Isotopes have certainly recorded the mark that humans have had, and are having, on the Earth System. Lead isotopes can trace human pollution, thousands of kilometers from source, millennia ago, but it would be difficult to argue that this constitutes a change in the functioning of the Earth System. Sulfur isotopes have recorded a more recent pollution phenomenon, acid rain, but the impacts of acid rain are also localized. On the other hand, boron isotopes provide a record of global ocean acidification, which is leading to a substantial change in the functioning of the hydrosphere and marine biosphere. Carbon and nitrogen isotopes show changes from the start of the Industrial Revolution that become particularly pronounced around the time of the ‘Great Acceleration’. The change in the former is related to human influence on the carbon cycle (release of carbon from fossil fuel burning, land use change and the industrialization of farming), and the change in the latter shows what is believed to be
the largest change in the global nitrogen cycle for 2.5 billion years, likely to be related
to fossil fuel burning and the artificial production of fertilizers. The appearance of
radioisotopes, especially plutonium-239, released by atmospheric nuclear weapons
testing around the start of the ‘Great Acceleration’, could be used as a stratigraphic
marker for the beginning of the Anthropocene. The search for sections around the world
that could be designated as the GSSP, using plutonium-239 or other proxies, should
now begin in earnest.

Overall, we have shown how isotopes have recorded important anthropogenic impacts
on different parts of the Earth System, and in some cases these impacts constitute
changes in the functioning of the Earth System. The carbon and nitrogen isotope records
in particular could be used to support the argument of Steffen et al. (2015) that it is only
beyond ~AD 1950 that there is clear evidence for fundamental shifts in the state and
functioning of the Earth System, that are beyond the range of variability of the
Holocene and driven by human activities.

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**Figure captions**

**Figure 1.** The three isotopes of carbon, showing differences in the number of neutrons but the same number of protons.

**Figure 2.** Trends in lead isotopes and concentrations from the sediments of Lake Koltjärn in Sweden, with a decline in $^{206}\text{Pb}/^{207}\text{Pb}$ taken to indicate increased anthropogenic contribution to overall lead pollution (Renberg et al., 2002).
Figure 3. a: $\delta^{13}CO_2$ from bubbles in an Antarctic ice core record (Rubino et al., 2013) and $\delta^{13}CO_2$ measured from the atmosphere at the Mauna Loa monitoring station in Hawaii (annual average) (Keeling et al., 2005; updated). b: $\delta^{15}N$ from organic matter from lake sediments from the US Rockies (3 point moving average, 13 lakes) (Wolfe et al., 2013) and from nitrate in Greenland ice cores (Hastings et al., 2009). In both the carbon and nitrogen isotope records, a trend to lower values occurs through the nineteenth and early twentieth centuries, with an acceleration after ~AD 1950.

Figure 4. Monthly data from the Mauna Loa monitoring station in Hawaii (Keeling et al., 2005; updated) showing an increase in atmospheric CO$_2$ concentrations from AD 1958-2015 and a decline in $\delta^{13}CO_2$ from AD 1980 (when monitoring of this began).

Figure 5. Yields of atmospheric nuclear bomb tests per year shown by bars (UNSCEAR, 2000) and $^{239,240}Pu$ deposition in Japan (as recorded in water and dust samples) shown by the dashed line (Hirose et al., 2000). The yield of atmospheric nuclear tests in the atmosphere peaked in AD 1962. $^{239,240}Pu$ in Japan peaked the following year.
Carbon-12

Carbon-13

Carbon-14

- Electron
- Proton
- Neutron
Sediments from 13 lakes in the US Rockies

Greenland ice

Mauna Loa

Antarctic ice

δ¹³C₂ ‰ VPDB

δ¹⁵N ‰ NO₃⁻ ice

Year AD
Atmospheric $\delta^{13}$CO$_2$ concentration ppm

Year AD


Atmospheric $\delta^{13}$CO$_2$ %

Total annual yields of nuclear tests

\[ ^{239,240}\text{Pu abundance} \]