Is there an isotopic signature of the Anthropocene?

Jonathan R Dean, Melanie J Leng and Anson W Mackay

The Anthropocene Review 2014 1: 276 originally published online 15 July 2014
DOI: 10.1177/2053019614541631

Review

THE ANTHROPOCENE REVIEW

The Anthropocene Review
2014, Vol. 1(3) 276–287

DOI: 10.1177/2053019614541631
anr.sagepub.com
SAGE

Is there an isotopic signature of the Anthropocene?

Jonathan R Dean,[1] Melanie J Leng[1,2] and Anson W Mackay[3]

Abstract

We consider whether the Anthropocene is recorded in the isotope geochemistry of the atmosphere, sediments, plants and ice cores, and the time frame during which any changes are recorded, presenting examples from the literature. Carbon and nitrogen isotope ratios have become more depleted since the 19th century, with the rate of change accelerating after ~AD 1950, linked to increased emissions from fossil fuel consumption and increased production of fertiliser. Lead isotope ratios demonstrate human pollution histories several millennia into the past, while sulphur isotopes can be used to trace the sources of acid rain. Radioisotopes have been detectable across the planet since the 1950s because of atmospheric nuclear bomb tests and can be used as a stratigraphic marker. We find there is isotopic evidence of widespread human impact on the global environment, but different isotopes have registered changes at different times and at different rates.

Keywords

Anthropocene, carbon, human impact, isotopes, lead, nitrogen, radioisotopes, Suess effect, sulphur

Introduction

The Anthropocene, the term used informally to denote the current interval where humans have become a dominant force of global environmental change (Crutzen, 2002; Crutzen and Stoermer, 2000), is contentious. There is no doubt that humanity has left its mark on the planet. For example, humans now transport more soil and rock around the surface of the Earth than natural processes do (Wilkinson, 2005), CO_2 levels have risen dramatically to the highest levels seen in at least 800,000 years (Keeling et al., 2005; updated: http://scrippsco2.ucsd.edu/data/in_situ_co2/monthly_mlo.csv; Lüthi et al., 2008) and humanity is implicated in causing rates of species extinctions to increase well beyond background levels (Barnosky et al., 2011). Consequently, a working group of the

[1]British Geological Survey, UK
[2]University of Nottingham, UK
[3]University College London, UK

Corresponding author:
Jonathan R Dean, NERC Isotope Geosciences Facilities, British Geological Survey, Nottingham NG12 5GG, UK.
Email: jond@bgs.ac.uk

International Commission on Stratigraphy is set to present its preliminary findings in 2016 on whether the Anthropocene is distinctive and enduring enough to be defined as a new epoch and if so where the Holocene–Anthropocene boundary should be set (Foley et al., 2013; Gale and Hoare, 2012; Vince, 2011; Zalasiewicz et al., 2011). Ruddiman (2003, 2013) and Ruddiman et al. (2011, 2014) have argued the Anthropocene started in the early to mid Holocene, when they suggest land clearance and agriculture initiated changes in the composition of the atmosphere. Crutzen and Stoermer (2000), Crutzen (2002) and Steffen et al. (2011) have suggested a later date, in the late 18th or early 19th centuries, associated with the Industrial Revolution in Northern Europe. Alternatively, a 'Great Acceleration' in human impacts on the global environment has been suggested to have occurred ~AD 1950 (Steffen et al., 2007) and it has been proposed the Anthropocene could be defined as starting around this time (Zalasiewicz et al., 2014).

There is an urgent need to understand the impact humans have had on the global environment and when changes occurred. This review concentrates on wide-scale anthropogenic impact as recorded by isotope data from natural archives. Isotopes are different types of an element: they have the same number of protons but a different number of neutrons (e.g. Hoefs, 2009; Sharp, 2007). The ratio of one isotope of an element to another can vary through time depending on a host of environmental factors, meaning changes in isotope ratios can be used to reconstruct changes in, for example, climate, pollution and the composition of the atmosphere. In this review, we have selected the isotopes that previous studies have highlighted as important in tracking human impacts on the global environment. We show how isotopes record heavy metal contamination linked to technological innovations from Greek and Roman times onwards (lead isotopes), late-Holocene forest clearance and widespread fossil fuel burning since the onset of the Industrial Revolution (carbon isotopes), increased production and use of artificial fertilisers (nitrogen isotopes), acid rain (sulphur isotopes) and atmospheric nuclear weapons testing (caesium and plutonium isotopes). We consider how isotopes could contribute to the debate on where to set the Holocene–Anthropocene boundary.

Notation and standardisation of stable isotope data are summarised in Sharp (2007) and Hoefs (2009). $\delta^{13}C$ represents the ratio of $^{13}C/^{12}C$ and $\delta^{15}N$ the ratio of $^{15}N/^{14}N$ and are given in per mil (‰) relative to VPDB and AIR respectively. $\delta^{34}S$ represents the ratio of $^{34}S/^{32}S$ and is given in ‰ relative to VCDT. Lead isotopes are measured against a variety of standards as reviewed in Komárek et al. (2008). The abundance of ^{14}C ($\Delta^{14}C$) in a sample is given in ‰ relative to NIST oxalic acid activity corrected for decay (Stuiver and Polach, 1977). The abundance of radioisotopes such as ^{137}Cs and $^{239,240}Pu$ are measured in becquerel (Bq), with one Bq representing one decay per second (L'Annunziata, 2012).

Changes in the global carbon cycle

Human activity has altered the concentration and isotopic composition of the gases in the atmosphere. Rises in atmospheric methane (CH_4) and carbon dioxide (CO_2) are captured in gas bubbles in ice cores (e.g. MacFarling Meure et al., 2006; Rubino et al., 2013) ~5000 years ago and ~8000 years ago, respectively. Ruddiman (2003, 2013) and Ruddiman et al. (2011, 2014) have argued these increases were caused by humans, and this has led to the Early Anthropogenic Hypothesis, which argues anthropogenic effects on global climate began millennia ago and had it not been for human-induced greenhouse gas increases leading to global warming the climate would have cooled substantially during recent millennia. A key part of their argument involves using carbon isotopes to trace the origins of these increases in CH_4 and CO_2 to wetland expansion, linked to rice production, and to widespread forest clearance. $\delta^{13}C$ of atmospheric CH_4 ($\delta^{13}CH_4$) from ice core bubbles

from the late Holocene have values ~−47‰ to −49‰ (Ferretti et al., 2005; Mischler et al., 2009). While some argue that these low values of $\delta^{13}CH_4$ could be explained by increased delivery of depleted (more negative) carbon from natural wetlands (e.g. Schmidt et al., 2004), Ruddiman et al. (2011) contend this would have been unlikely because of the drying in the late Holocene of northern monsoonal regions and the cooling of boreal regions, which would have reduced, not increased, CH_4 emissions of natural wetlands. Rather, they suggest that $\delta^{13}CH_4$ data could be explained by human emissions, with the observed mean of −48‰ satisfied by emissions from rice paddies (−63‰) and livestock (−60‰) and anthropogenic burning of grasses (−25‰). In terms of CO_2, Elsig et al. (2009) argue that the very small decrease in the $\delta^{13}C$ of atmospheric CO_2 ($\delta^{13}CO_2$) in the mid to late Holocene (before the Industrial Revolution), as atmospheric CO_2 concentrations were rising, would limit the net terrestrial contribution to atmospheric CO_2 during the last 7000 years to only ~5 ppm. Instead, there could have been large releases of CO_2 from the oceans (Broecker et al., 1999; Ridgwell et al., 2003). However, Ruddiman et al. (2011) argue that Elsig et al. (2009) underestimate carbon burial in boreal peat, and if burial in peat over the last 7000 years was greater than Elsig et al. (2009) calculated then it would require far greater anthropogenic emissions, via forest clearance, to balance the $\delta^{13}CO_2$ budget. The complexities of the carbon cycle mean the debate vis-à-vis the relative importance of human versus natural sources and sinks of carbon is complicated, and many researchers (e.g. Steffen et al., 2011) dismiss the plausibility of the Early Anthropogenic Hypothesis, but it is clear carbon isotopes are a key part of this debate.

As recorded in direct measurements from the atmosphere, in gas bubbles trapped in ice cores and in natural archives including tree rings (February and Stock, 1999; Stuiver and Quay, 1981), corals (Nozaki et al., 1978; Swart et al., 2010), foraminifera (Al-Rousan et al., 2004; Black et al., 2011) and marine molluscs (Butler et al., 2009), there has been a more substantial change in the $\delta^{13}CO_2$ of the atmosphere since the 19th century, with the trend to lower values through the 19th century accelerating after ~AD 1950 (Figure 1), at the time of increased fossil fuel consumption that followed the Second World War (Steffen et al., 2007). The changes in $\delta^{13}C$ are of a different magnitude and the absolute values are different in tree rings, corals, foraminifera and direct measurements of the atmosphere or of gas bubbles in ice. This is because as organisms use carbon during growth, they preferentially take up one isotope over another, causing a change in the $\delta^{13}C$ from the source, a process known as fractionation (e.g. Hoefs, 2009; Sharp, 2007). However, assuming this fractionation is constant through time, it is still possible to track changes in the composition of the atmosphere using tree rings, corals and foraminifera. The classic graph from Mauna Loa shows $\delta^{13}CO_2$ declining (−7.6‰ in 1980 to −8.3‰ in 2011) as CO_2 concentrations in the atmosphere have risen (316 ppm in 1959 to 396 ppm in 2013) (Figure 2) (Keeling et al., 2005; updated: http://scripps-sco2.ucsd.edu/data/in_situ_co2/monthly_mlo.csv). There was also a decline in the amount of ^{14}C in atmospheric CO_2 ($\Delta^{14}CO_2$) in the first half of the 20th century (Levin et al., 2010; Stuiver and Quay, 1981), before the trend was interrupted in the 1950s and 1960s, followed by a decline again to the present day (Levin et al., 2013). These declines in $\delta^{13}CO_2$ and $\Delta^{14}CO_2$ (called the Suess Effect; Keeling, 1979; Suess, 1955) are linked to the burning of fossil fuels. Fossil fuels, such as the vast coal deposits of the Carboniferous period, are composed of the organic remains of organisms (mainly plants) that lived millions of years ago. Plants preferentially take up ^{12}C over ^{13}C so have low $\delta^{13}C$ (e.g. Farquhar et al., 1989), with most oil deposits having values of −32‰ to −21‰ and coal deposits −26‰ to −23‰ (Sharp, 2007). Consequently, CO_2 from fossil fuels contains on average 2% less ^{13}C per mole than atmospheric CO_2 (Keeling, 1979). Extraction and burning of these fossil fuel reserves releases this ^{12}C-enriched carbon back into the atmosphere, leading to a decline in $\delta^{13}CO_2$. Old carbon from fossil fuels is also virtually free of ^{14}C (Keeling, 1979), since the time between being deposited in the fossil record and burning is many thousands of half-lives

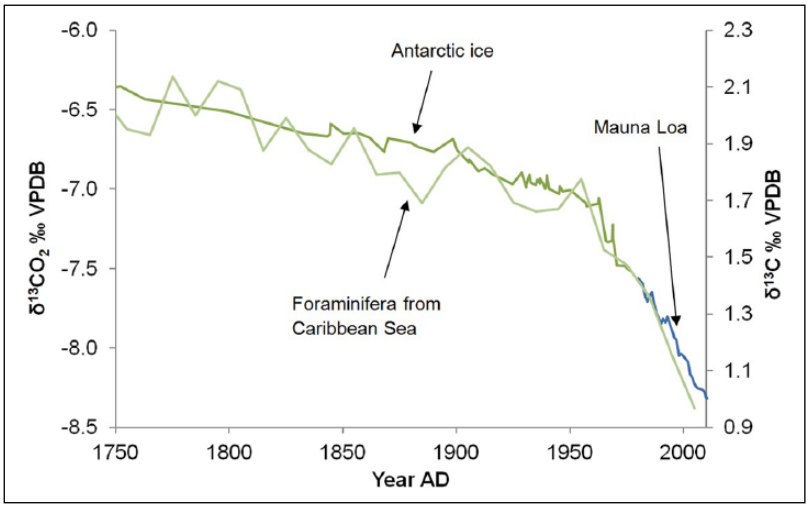

Figure 1. $\delta^{13}CO_2$ from Antarctic ice core record (Rubino et al., 2013), $\delta^{13}C$ record from foraminifera from the Caribbean Sea (Black et al., 2011) and $\delta^{13}CO_2$ from the Mauna Loa monitoring station (Keeling et al., 2005; updated: http://scrippsco2.ucsd.edu/data/in_situ_co2/monthly_mlo.csv). The former two records show a gradual depletion through the 19th century and an acceleration after ~AD 1950.

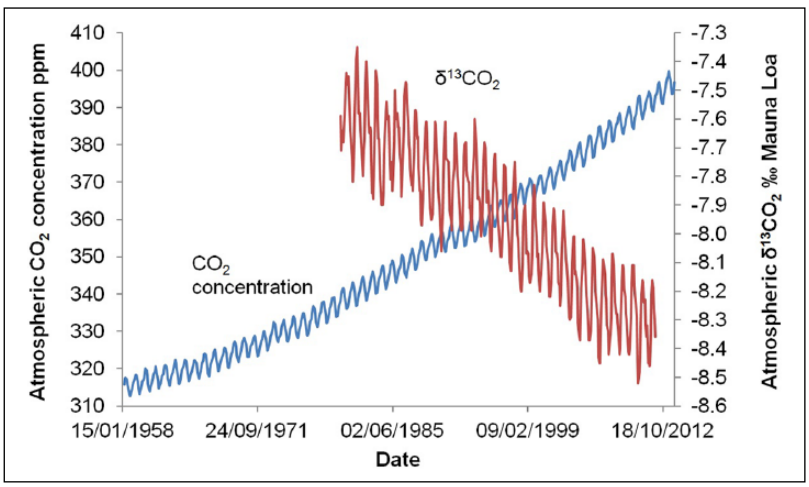

Figure 2. Monthly data from the Mauna Loa monitoring station (Keeling et al., 2005; updated: http://scrippsco2.ucsd.edu/data/in_situ_co2/monthly_mlo.csv) showing an increase in the concentration of CO_2 in the atmosphere from 1958 and a decline in $\delta^{13}CO_2$ from 1980 when monitoring of this began.

of ^{14}C, so the release of this old carbon will lead to a decline in $\Delta^{14}CO_2$ in the atmosphere. $\delta^{13}C$ changes in the atmosphere have been vital in allowing the Intergovernmental Panel on Climate Change (IPCC) to conclude there is a 'very high confidence' that the dominant cause of the observed increase in CO_2 concentrations in the atmosphere since the 19th century has been the human burning of fossil fuels (IPCC, 2013).

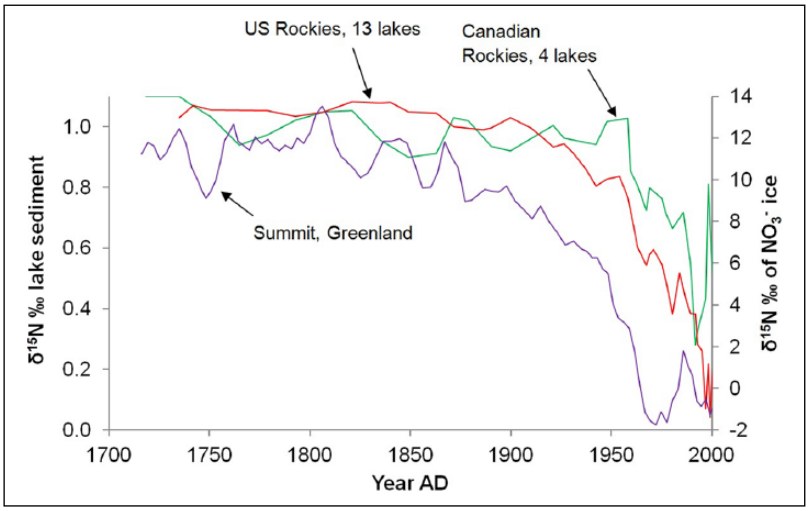

Figure 3. $\delta^{15}N$ from organic matter from lake sediments from the US and Canadian Rockies (three-point moving average) (Wolfe et al., 2013) and from nitrate in Greenland ice cores (Hastings et al., 2009). Depletion occurs after ~AD 1850, with an acceleration after ~AD 1950.

Changes to the nitrogen cycle

There have also been changes in the global nitrogen cycle, with increases in the amount of reactive nitrogen (nitrogen compounds such as nitrogen oxides that support biological growth) in the atmosphere, thought to be mainly due to the burning of fossil fuels and the use of fertiliser in agriculture (Galloway et al., 2004; Jaegle et al., 2005). As with carbon isotopes and the carbon cycle, $\delta^{15}N$ can be used to track changes in the nitrogen cycle and identify the sources of the nitrogen released. Anthropogenic reactive nitrogen sources, especially fertilised soils (Park et al., 2012; Pérez et al., 2001), but also fossil fuel emissions (Felix et al., 2012), are generally thought to be depleted in $\delta^{15}N$ relative to natural sources (although they can have highly variable values and some have argued $\delta^{15}N$ from fossil fuel emissions is unlikely to be lower than that from natural sources; Sharp, 2007; Geng et al., 2014). In organic matter from remote lake sediments from across North America and the Arctic (Holmgren et al., 2010; Holtgrieve et al., 2011; Wolfe et al., 2013), and in nitrate (NO_3–) from ice cores from Greenland (Hastings et al., 2009), there have been declines in $\delta^{15}N$ from ~AD 1850 (Figure 3). (Again, note that as a result of fractionation, the values and magnitudes of change of $\delta^{15}N$ in lake organic matter and ice core NO_3– differ, but they both shown a decline at similar times.) $\delta^{15}N$ values in Greenland NO_3– declined from +10.6‰ in AD 1716 to +0.8‰ in AD 2005 (Hastings et al., 2009). The trend in $\delta^{15}N$ may be because of the increase in isotopically depleted nitrogen from anthropogenic sources (fossil fuel combustion and fertilisers) (Felix and Elliott, 2013; Hastings et al., 2009; Holtgrieve et al., 2011), although Geng et al. (2014) have argued that the decline may be due to an equilibrium shift in gas-particle partitioning of atmospheric NO_3– caused by increasing atmospheric acidity resulting from anthropogenic emissions of nitrogen and sulphur oxides.

As with $\delta^{13}C$, while there is a decline in $\delta^{15}N$ from the 19th century in many records, it is really after ~AD 1950 that the trend accelerates and becomes pronounced (Figure 3). The changes that have occurred in the last century in Sky Pond lake in the US Rockies, for example, are without

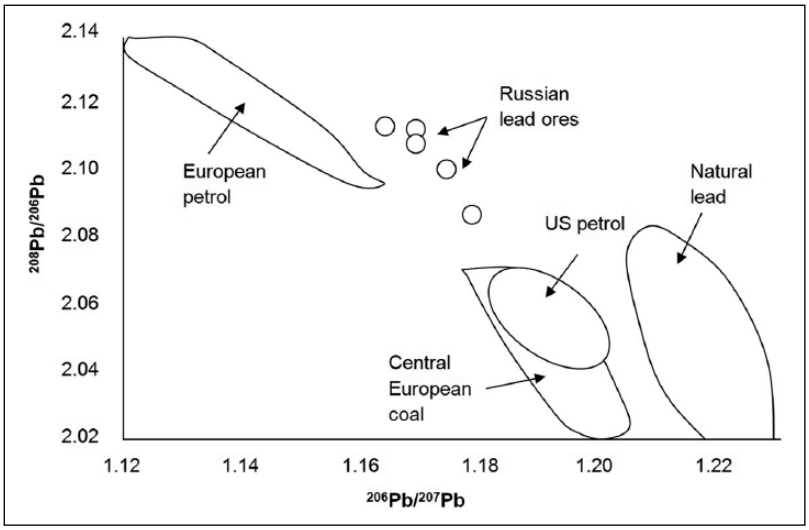

Figure 4. A $^{206}Pb/^{207}Pb$ versus $^{208}Pb/^{206}Pb$ plot showing the different isotopic compositions of selected lead sources. Modified from Komárek et al. (2008).

precedent in the 14,000 year record (Wolfe et al., 2013). Although, as we have demonstrated, the real drivers of the $\delta^{15}N$ trend are debated, it is probable that a combination of anthropogenic processes are causing this decline, so $\delta^{15}N$ is a useful tool in tracing human impacts on the global nitrogen cycle.

Tracing pollution

As well as causing changes in the carbon and nitrogen cycles, human activity has caused pollution by remobilising certain elements. This can be traced using isotopes.

Lead isotopes

For millennia, humans have been mining and smelting lead ores, which has released vast quantities of lead into the atmosphere, causing widespread airborne pollution (Adriano, 2001; Settle and Patterson, 1980). There is evidence for lead contamination in Greenland ice cores, carried there in the atmosphere as microparticles, for over 2000 years (e.g. Hong et al., 1994; Rosman et al., 1997). Since different lead ores have different lead isotope ratios, it is possible to pinpoint where the lead was being mined. Rosman et al. (1997) showed that between ~150 BC and AD 50, 70% of the lead seen in Greenland ice cores originated from southern Spain, and historical records show the Romans mined the area at this time. As well as different lead ores, lead isotope ratios can be used to distinguish between pollution from different industrial processes. Komárek et al. (2008), using $^{206}Pb/^{207}Pb$ versus $^{208}Pb/^{206}Pb$, were able to distinguish between lead emitted from vehicles in Europe and the USA, coal burning in central Europe and natural sources (Figure 4). More recently it has been shown that lead in Greenland ice is increasingly from Chinese sources (Bory et al., 2014).

Trends in lead isotope ratios (especially $^{206}Pb/^{207}Pb$) can also be used to track changes in pollution through time. For example, in Sweden, background $^{206}Pb/^{207}Pb$ is thought to be around 1.5, whereas atmospheric lead pollution derived from smelting, leaded petrol and burning of coal has a

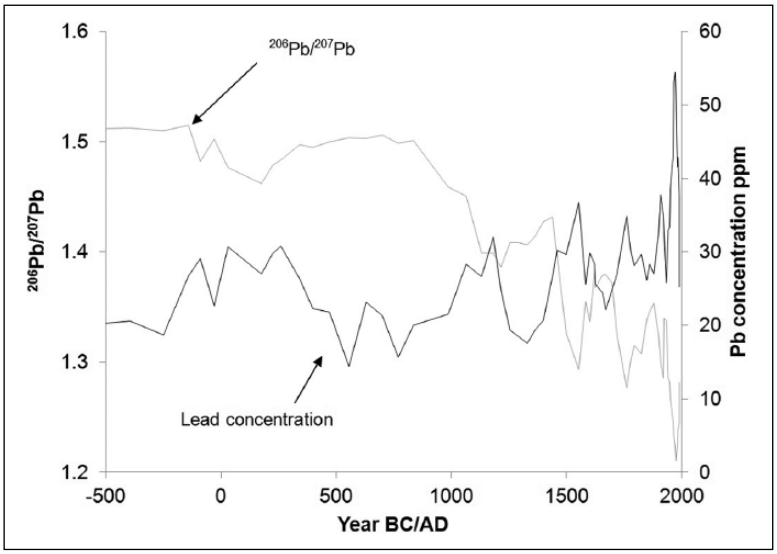

Figure 5. Trends in $^{206}Pb/^{207}Pb$ and lead concentrations from Lake Koltjärn in Sweden, with a depletion in the ratio taken to represent increased anthropogenic lead pollution (Renberg et al., 2002).

$^{206}Pb/^{207}Pb$ value of ~1.2 (Renberg et al., 2002). Lake sediments show there was a decline in the ratio in Roman times (to ~1.46), and then an increase to higher values in the Dark Ages ~AD 500–800 (~1.50) (Renberg et al., 2002). Minimum $^{206}Pb/^{207}Pb$ ratios (~1.22) were reached in the 1970s when leaded petrol consumption peaked (Figure 5). With the phasing out of leaded petrol in Europe there has been an increase in the ratio (currently ~1.28). The low $^{206}Pb/^{207}Pb$ in Roman times, related to lead mining, as seen in Greenland and Sweden, could be used to support the argument made by others (Certini and Scalenghe, 2011; Ellis et al., 2013; Ruddiman, 2003) using different proxies that substantial human impacts on the environment were occurring millennia before the Industrial Revolution.

Sulphur isotopes

Sulphur isotope ratios can be used to track fossil fuel burning and to trace the sources of pollution because, as with lead isotopes, natural and anthropogenic sources often have different isotope ratios (e.g. Krouse et al., 1984; Lim et al., 2014). Sulphur released into the atmosphere has the potential to cause acid rain. Concerns over widespread ecosystem damage resulting from acid rain first gained prominence in Europe in the late 1950s. Tracing the sources of sulphur pollution is particularly important given sulphur compounds produced and released into the atmosphere in one country can travel across borders and cause acid rain in another (Metcalfe and Derwent, 2005). Yu et al. (2007) demonstrated how the $\delta^{34}S$ of sulphate in meteoric waters from Chuncheon in South Korea vary from +2.6 to +7.5‰, which is significantly different from the $\delta^{34}S$ of sulphate from locally combusted coal (−4.5 to −0.7‰). This was taken to suggest that sulphur implicated in acid rain in that region was not the result of local pollution. A decline in emissions over time from brown coal power stations in eastern Germany has been recorded in an increase in $\delta^{34}S$ of rain in Wroclaw in Poland, demonstrating the effectiveness of measures taken to reduce acid rain resulting from anthropogenic emissions (Jędrysek, 2000). Indeed, global sulphur emissions are showing an

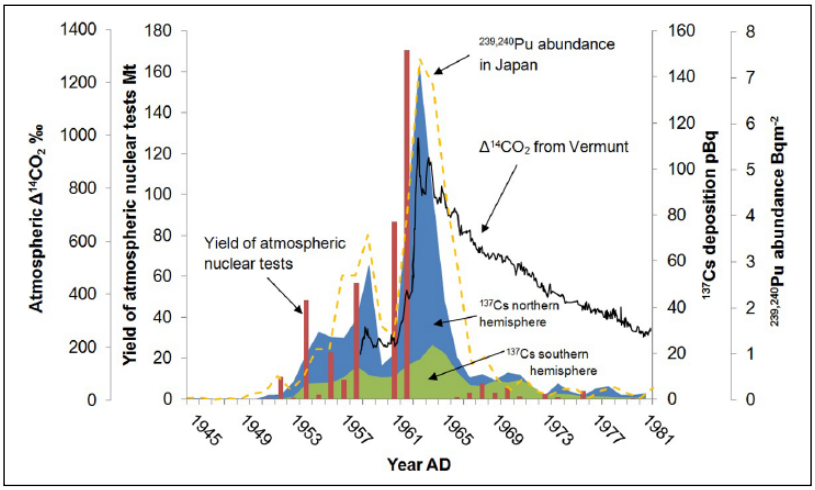

Figure 6. Yield of atmospheric nuclear tests per year shown by bars (UNSCEAR, 2000), ^{137}Cs deposition in Northern and Southern Hemispheres represented by areas (UNSCEAR, 2000), ^{239,240}Pu deposition in Japan shown by the dashed line (Hirose et al., 2000) and $\Delta^{14}CO_2$ measured at Vermunt, Austria shown by the solid line (Levin et al., 1985). The yield of atmospheric nuclear tests in the atmosphere peaked in 1962. $\Delta^{14}CO_2$ at Vermunt, ^{239,240}Pu in Japan and ^{137}Cs deposition in the Northern Hemisphere peaked in 1963 and ^{137}Cs in the Southern Hemisphere in 1964.

overall decline (Klimont et al., 2013). This demonstrates that some anthropogenic impacts on the environment, in this case acid rain linked to sulphur emissions as recorded by δ^{34}S, have peaked, at least in some parts of the world.

Radioisotopes

Some isotopes (e.g. ^{137}Cs, ^{239}Pu and ^{240}Pu) occur on Earth almost entirely because of their production and release into the atmosphere from nuclear reactors and especially atmospheric nuclear weapons testing. They provide a rather precise stratigraphic point in geological archives, with detectable levels first apparent ~AD 1952, and peak abundance ~AD 1963/1964 after a large number of atmospheric nuclear tests were carried out in AD 1962 before the Partial Nuclear Test Ban Treaty came into effect (Figure 6) (Hirose et al., 2000; United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 2000). ^{14}C is produced naturally in the atmosphere through the interaction of neutrons with nitrogen atoms, but as discussed above the burning of fossil fuels had been leading to a decline in $\Delta^{14}CO_2$ in the atmosphere. This trend was interrupted as neutrons released by atmospheric nuclear tests increased the production of ^{14}C in the atmosphere, with a peak at a similar time to the peaks in ^{137}Cs, ^{239}Pu and ^{240}Pu (Figure 6) (Graven et al., 2012; Levin and Kromer, 2004; Levin et al., 1985; Naegler and Levin, 2009), before a decline again to the present day, producing a time-dependent distribution pattern that is referred to as the 'bomb curve'. This is seen in archives such as tree rings (Hua et al., 2000) and corals (Roark et al., 2006).

Conclusion

Changes in isotope geochemistry demonstrate that humans are having an impact on the global environment. Different isotopes have recorded different anthropogenic impacts, and changes have

occurred at different times and different rates. $\delta^{13}C$ and $\Delta^{14}C$ show the input of fossil fuel-derived CO_2 into the atmosphere, $\delta^{15}N$ records reveal a change in the global nitrogen cycle, lead and sulphur isotopes are tracers of human pollution histories and radioisotopes record the point at which humans mastered nuclear weapons technology. Some of the isotopes that we use to demonstrate human impacts, especially carbon and nitrogen isotopes, could also be influenced in similar ways by natural processes. This complexity has led to the Early Anthropogenic Hypothesis debate. On the other hand, other isotopes, especially radioisotopes, but arguably also lead isotopes, show a clear human imprint: in the case of certain radioisotopes their occurrence is almost entirely due to human-induced nuclear reactions and in the case of lead isotopes the ratios are changed in ways unlikely to be due to natural processes.

As for whether isotopes can contribute to the debate on where to set the Holocene–Anthropocene boundary, we have shown there is a clear acceleration in the trend to lower $\delta^{13}C$ and $\delta^{15}N$ after ~AD 1950, at the time of the 'Great Acceleration' in human activities (Steffen et al., 2007), and a decade later there was a near synchronous, worldwide peak in radioisotopes related to atmospheric nuclear weapons testing that could be useful as a unique stratigraphic marker to define the boundary (Zalasiewicz et al., 2014). However, it has been argued that carbon isotopes show (smaller) changes in the global composition of the atmosphere hundreds to thousands of years before AD 1950 and other isotopes, such as lead, also show human impacts on the environment millennia ago. Therefore, while there *is* an isotopic signature of the Anthropocene, and isotope geochemistry can play a role in the decision of the International Commission on Stratigraphy regarding whether to define a new geological epoch, it is not clear from isotopes alone *where* to set the Holocene–Anthropocene boundary.

Acknowledgements

Frank Oldfield and Jan Zalasiewicz are thanked for their invitation to write this review and three anonymous reviewers are thanked for their comments that improved the manuscript. We are also grateful to David Black, Alexander Wolfe and Ingemar Renberg for providing us with data to use in our figures and to Elsevier for permission to reproduce Figure 4. This work is published with the permission of the Executive Director of the British Geological Survey.

Funding

This research received no specific grant from any funding agency in the public, commercial, or not-for-profit sectors.

References

Adriano DC (2001) *Trace Elements in the Terrestrial Environments: Biogeochemistry, Bioavailability, and Risks of Metals*. New York: Springer.

Al-Rousan S, Pätzold J, Al-Moghrabi S et al. (2004) Invasion of anthropogenic CO_2 recorded in planktonic foraminifera from the northern Gulf of Aqaba. *International Journal of Earth Sciences* 93: 1066–1076.

Barnosky AD, Matzke N, Tomiya S et al. (2011) Has the Earth's sixth mass extinction already arrived? *Nature* 471: 51–57.

Black D, Thunell R, Wejnert K et al. (2011) Carbon isotope composition of Caribbean Sea surface waters: Response to the uptake of anthropogenic CO_2. *Geophysical Research Letters* 38: L16609.

Bory AJM, Abouchami W, Galer SJG et al. (2014) A Chinese imprint in insoluble pollutants recently deposited in central Greenland as indicated by lead isotopes. *Environmental Science and Technology* 48: 1451–1457.

Broecker WS, Clark E, McCorckle DC et al. (1999) Evidence for a reduction in the carbonate ion content of the deep sea during the course of the Holocene. *Paleoceanography* 14: 744–752.

Butler PG, Scourse JD, Richardson CA et al. (2009) Continuous marine radiocarbon reservoir calibration and the ^{13}C Suess Effect in the Irish Sea: Results from the first multi-centennial shell-based marine master chronology. *Earth and Planetary Science Letters* 279: 230–241.

Certini G and Scalenghe R (2011) Anthropogenic soils are the golden spikes for the Anthropocene. *The Holocene* 21: 1269–1274.

Crutzen PJ (2002) Geology of mankind. *Nature* 415: 23.

Crutzen PJ and Stoermer EF (2000) The Anthropocene. *IGBP Newsletter* 41: 12.

Ellis EC, Kaplan JO, Fuller DQ et al. (2013) Used planet: A global history. *PNAS* 110: 7978–7985.

Elsig J, Schmitt K, Leuenberger D et al. (2009) Stable isotope constraints on Holocene carbon cycle changes from an Antarctic ice core. *Nature* 461: 507–510.

Farquhar GD, Ehleringer JR and Hubick KT (1989) Carbon isotope discrimination and photosynthesis. *Annual Review of Plant Physiology and Plant Molecular Biology* 40: 503–537.

February EC and Stock WD (1999) Declining trends in the $^{13}C/^{12}C$ ratio of atmospheric carbon dioxide from tree rings of South Africa. *Quaternary Research* 52: 229–236.

Felix JD and Elliott EM (2013) The agricultural history of human–nitrogen interactions as recorded in ice core $\delta^{15}N\text{-}NO_3^-$. *Geophysical Research Letters* 40: 1642–1646.

Felix JD, Elliott EM and Shaw SL (2012) Nitrogen isotopic composition of coal-fired power plant NO_x: Influence of emission controls and implications for global emission inventories. *Environmental Science and Technology* 46: 3528–3535.

Ferretti DF, Miller JB, White JWC et al. (2005) Unexpected changes to the global methane budget over the last 2,000 years. *Science* 309: 1714–1717.

Foley SF, Gronenborn D, Andreae MO et al. (2013) The Palaeoanthropocene – The beginning of anthropogenic environmental change. *Anthropocene* 3: 83–88.

Gale SJ and Hoare PG (2012) The stratigraphic status of the Anthropocene. *The Holocene* 22: 1491–1495.

Galloway JN, Dentener FJ, Capone DG et al. (2004) Nitrogen cycles: Past, present, and future. *Biogeochemistry* 70:153–226.

Geng L, Alexander B, Cole-Dai J et al. (2014) Nitrogen isotopes in ice core nitrate linked to anthropogenic atmospheric acidity change. *PNAS*. Epub ahead of print. DOI: 10.1073/pnas.1319441111.

Graven HD, Gruber N, Key R et al. (2012) Changing controls on oceanic radiocarbon: New insights on shallow-to-deep ocean exchange and anthropogenic CO_2 uptake. *Journal of Geophysical Research Oceans* 117: C10005.

Hastings MG, Jarvis JC and Steig EJ (2009) Anthropogenic impacts on nitrogen isotopes of ice-core nitrate. *Science* 324: 1288.

Hirose K, Igarashi Y, Aoyama M et al. (2000) Long-term trends of plutonium fallout observed in Japan. In: Kudo A (ed.) *Plutonium in the Environment*. Amsterdam: Elsevier, pp. 251–266.

Hoefs J (2009) *Stable Isotope Geochemistry*. Berlin: Springer-Verlag.

Holmgren SU, Bigler C, Ingólfsson Ó et al. (2010) The Holocene–Anthropocene transition in lakes of western Spitsbergen Svalbard (Norwegian High Arctic): Climate change and nitrogen deposition. *Journal of Paleolimnology* 43: 393–412.

Holtgrieve GW, Schindler DE, Hobbs WO et al. (2011) A coherent signature of anthropogenic nitrogen deposition to remote watershed of the northern hemisphere. *Science* 334: 1545–1548.

Hong S, Candelone J, Patterson CC et al. (1994) Greenland ice evidence of hemispheric lead pollution two millennia ago by Greek and Roman civilisations. *Science* 265: 1841–1843.

Hua Q, Barbetti M, Jacobsen GE et al. (2000) Bomb radiocarbon in annual tree rings from Thailand and Australia. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* 173: 359–365.

Intergovernmental Panel on Climate Change (2013) *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge: Cambridge University Press.

Jaegle L, Steinberger L, Martin RV et al. (2005) Global partitioning of NO_x sources using satellite observations: Relative roles of fossil fuel combustion, biomass burning and soil emissions. *Faraday Discuss* 130: 407–423.

Jędrysek MO (2000) Oxygen and sulphur isotope dynamics in the $SO_{2}{}_{-}{}^{4}$ of an urban precipitation. *Water, Air and Soil Pollution* 117: 15–25.

Keeling CD (1979) The Suess Effect: ^{13}Carbon–^{14}Carbon interrelations. *Environment International* 2: 229–300.

Keeling CD, Piper SC, Bacastow RB et al. (2005) Atmospheric CO_2 and $^{13}CO_2$ exchange with the terrestrial biosphere and oceans from 1978 to 2000: Observations and carbon cycle implications. In: Ehleringer JR, Cerling TE and Dearing MD (eds) *A History of Atmospheric CO_2 and its effects on Plants, Animals, and Ecosystems*. New York: Springer Verlag, pp. 83–113.

Klimont Z, Smith SJ and Cofala J (2013) The last decade of global anthropogenic sulphur dioxide: 2000–2011 emissions. *Environmental Science Letters* 8: 014003.

Komárek M, Ettler V, Chrastný V et al. (2008) Lead isotopes in environmental sciences: A review. *Environment International* 34: 562–577.

Krouse HR, Legge AH and Brown HM (1984) Sulphur gas emissions in the boreal forest: The West Whitecourt case study. *Water, Air and Soil Pollution* 22: 321–347.

L'Annunziata MF (2012) Radiation physics and radionuclide decay. In: L'Annunziata MF (ed.) *Handbook of Radioactivity Analysis*. Oxford: Academic Press, pp. 2–148.

Levin I and Kromer B (2004) The tropospheric $^{14}CO_2$ level in mid-latitudes of the Northern Hemisphere (1959–2003). *Radiocarbon* 46: 1261–1272.

Levin I, Kromer B and Hammer S (2013) Atmospheric $\Delta^{14}CO_2$ trend in Western European background air from 2000 to 2012. *Tellus B* 65: 20092.

Levin I, Kromer B, Schoch-Fischer H et al. (1985) 25 years of tropospheric ^{14}C observations in central Europe. *Radiocarbon* 27: 1–19.

Levin I, Naegler T, Kromer B et al. (2010) Observations and modelling of the global distribution and longterm trend of atmospheric $^{14}CO_2$. *Tellus B* 62: 26–46.

Lim C, Jang J, Lee I et al. (2014) Sulfur isotope and chemical compositions of the wet precipitation in two major urban areas, Seoul and Busan, Korea. *Journal of Asian Earth Sciences* 79: 415–425.

Lüthi D, Le Floch M, Bereiter B et al. (2008) High-resolution carbon dioxide concentration record 650,000–800,000 years before present. *Nature* 453: 379–382.

MacFarling Meure C, Etheridge D, Trudinger C et al. (2006) Law Dome CO_2, CH_4 and N_2O ice core records extended to 2,000 years BP. *Geophysical Research Letters* 33: L14810.

Metcalfe SE and Derwent D (2005) *Atmospheric Pollution and Environmental Change*. London: Hodder Arnold.

Mischler JA, Sowers TA, Alley RB et al. (2009) Carbon and hydrogen isotopic composition of methane over the last 1000 years. *Global Biogeochemical Cycles* 23: GB4024.

Naegler T and Levin I (2009) Observation-based global biospheric excess radiocarbon inventory 1963–2005. *Journal of Geophysical Research* 114: D17302.

Nozaki Y, Rye DM, Turekian KK et al. (1978) A 200 year record of carbon-13 and carbon-14 variations in a Bermuda coral. *Geophysical Research Letters* 5: 825–828.

Park S, Croteau P, Boering KA et al. (2012) Trends and seasonal cycles in the isotopic composition of nitrous oxide since 1940. *Nature Geoscience* 5: 261–265.

Pérez T, Trumbore SE, Tyler SC et al. (2001) Identifying the agricultural imprint on the global N_2O budget using stable isotopes. *Journal of Geophysical Research* 106: 9869–9878.

Renberg I, Brännvall M, Bindler R et al. (2002) Stable lead isotopes and lake sediments – A useful combination for the study of atmospheric lead pollution history. *The Science of the Total Environment* 292: 45–54.

Ridgwell AJ, Watson AJ, Maslin MA et al. (2003) Implications of coral reef buildup for the controls on atmospheric CO_2 since the Last Glacial Maximum. *Paleoceanography* 18: 1083.

Roark EB, Guilderson TP, Dunbar RB et al. (2006) Radiocarbon-based ages and growth rates of Hawaiian deep-sea corals. *Marine Ecology Progress Series* 327: 1–14.

Rosman KJR, Chisholm W, Hong S et al. (1997) Lead from Carthaginian and Roman Spanish mines isotopically identified in Greenland ice dated from 600 B.C. to 300 A.D. *Environmental Science and Technology* 31: 3413–3416.

Rubino M, Etheridge DM, Trudinger CM et al. (2013) A revised 1000 year atmospheric $\delta^{13}C$-CO_2 record from Law Dome and South Pole, Antarctica. *Journal of Geophysical Research: Atmospheres* 118: 8482–8499.

Ruddiman WF (2003) The anthropogenic greenhouse era began thousands of years ago. *Climatic Change* 61: 261–293.

Ruddiman WF (2013) The Anthropocene. *Annual Review of Earth and Planetary Sciences* 41: 45–68.

Ruddiman WF, Kutzbach JE and Vavrus SJ (2011) Can natural or anthropogenic explanations of late-Holocene CO_2 and CH_4 increases be falsified? *The Holocene* 21: 1–15.

Ruddiman WF, Vavrus S, Kutzbach J et al. (2014) Does pre-industrial warming double the anthropogenic total? *The Anthropocene Review*. Epub ahead of print. DOI: 10.1177/2053019614529263.

Schmidt GAD, Shindell DT and Harder S (2004) A note on the relationship between ice core methane concentrations and insolation. *Geophysical Research Letters* 31: L23206.

Settle D and Patterson CC (1980) Lead in Albacore: Guide to lead pollution in Americans. *Science* 201: 1167–1176.

Sharp Z (2007) *Stable Isotope Geochemistry*. New Jersey: Pearson.

Steffen W, Crutzen P and McNeil JR (2007) The Anthropocene: Are humans now overwhelming the great forces of nature? *Ambio* 36: 1317–1321.

Steffen W, Grinevald J, Crutzen P et al. (2011) The Anthropocene: Conceptual and historical perspectives. *Philosophical Transactions of the Royal Society* 369: 842–867.

Stuiver M and Polach HA (1977) Discussion: Reporting of ^{14}C data. *Radiocarbon* 19: 355–363.

Stuiver M and Quay P (1981) Atmospheric ^{14}C changes resulting from fossil fuel CO_2 release and cosmic ray flux variability. *Earth and Planetary Science Letters* 53: 349–362.

Suess HE (1955) Radiocarbon concentration in modern wood. *Science* 122: 415–417.

Swart PK, Greer L, Rosenheim BE et al. (2010) The ^{13}C Suess Effect in scleractinian corals mirror changes in the anthropogenic CO_2 inventory of the surface oceans. *Geophysical Research Letters* 37: L05604.

United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) (2000) *Sources and effects of ionizing radiation. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation to the General Assembly*. New York: United Nations.

Vince G (2011) An Epoch debate. *Science* 334: 32–37.

Wilkinson BH (2005) Humans as geologic agents: A deep-time perspective. *Geology* 33: 161–164.

Wolfe AP, Hobbs WO, Birks HH et al. (2013) Stratigraphic expressions of the Holocene–Anthropocene transition revealed in sediments from remote lakes. *Earth-Science Reviews* 116: 17–34.

Yu J, Park Y, Mielke RE et al. (2007) Sulfur and oxygen isotopic compositions of the dissolved sulphate in the meteoric water in Chuncheon, Korea. *Geosciences Journal* 11: 357–367.

Zalasiewicz J, Williams M, Haywood A et al. (2011) The Anthropocene: A new epoch of geological time? *Philosophical Transactions of the Royal Society A* 369: 835–841.

Zalasiewicz J, Williams M and Waters CN (2014) Can an Anthropocene Series be defined and recognised? *Geological Society, London, Special Publications*. Epub ahead of print. DOI: 10.1144/SP395.16.