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1 **Isotopic signatures**

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3 Jonathan R. Dean[a,b*], Melanie J. Leng[a,b], Anson W. Mackay[c]

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5 *[a]NERC Isotope Geosciences Facilities, British Geological Survey, Nottingham, UK*

6 *[b]Centre for Environmental Geochemistry, School of Geography, University of*

7 *Nottingham, UK*

8 *[c]Environmental Change Research Centre, Department of Geography, UCL, UK*

9

10 *Corresponding author, jond@bgs.ac.uk

11 Melanie Leng: mjl@bgs.ac.uk

12 Anson Mackay: a.mackay@ucl.ac.uk

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15   plutonium, radioisotope, Suess effect, sulfur

16

17   **Abstract**

18

19   Isotopes record human influence on the Earth System, providing evidence for the

20   Anthropocene. Lead and sulfur isotopes detail pollution histories going back millennia.

21   Carbon and nitrogen isotopes show substantial change since the Industrial Revolution,

22   and especially since the 1950s, related to increased fossil fuel consumption and fertilizer

23   production. Boron isotopes record ocean acidification related to CO_2 emissions.

24   Radioisotopes, e.g. plutonium, have been dispersed across the Earth since the 1950s due

25   to nuclear weapons testing and can be used as stratigraphic markers for the time that

26   carbon and nitrogen isotopes, and non-isotope proxies, show increased human influence

27   on the Earth System.

28

29   **Introduction**

30

31   The Anthropocene is the period during which human activity has had a discernible

32   impact on the functioning of the Earth System, causing changes outside of natural

33   variability (Steffen et al., 2007). There is intense debate around whether or not to define

34   the Anthropocene as a new geological time interval, and if so from what date. The

35   concept of the Anthropocene is no longer restricted to Earth Science and has far-

36   reaching cultural and political implications. Humans have influenced the functioning of

37   different spheres of the Earth System: the atmosphere (e.g. increasing greenhouse gas

38   concentrations that have contributed to global warming), the biosphere (e.g. increasing

39   extinction rates and changes in biogeochemical cycles), the cryosphere (e.g.

40   anthropogenic global warming contributing to the melting of ice sheets in some parts of

41   the world), the hydrosphere (e.g. ocean acidification) and the lithosphere (e.g. draining

42   of aquifers leading to earthquakes). These spheres are connected, for example human-

43   forced changes in the composition of the atmosphere have led to changes in the

44   hydrosphere (ocean acidification) and biosphere (associated impacts on coral

45   ecosystems), so the Earth System as a whole can be said to be influenced by human

46   actions. In this chapter, we focus on how isotope geochemistry can trace some of this

47   human influence.

48

49   Isotopes are different forms of an element: they have the same number of protons but a

50   different number of neutrons. Stable isotopes will not decay into other isotopes, whereas

51   radioactive isotopes will. For example, carbon-12 has 6 protons and 6 neutrons and

52   carbon-13 has 6 protons and 7 neutrons and both are stable isotopes, whereas carbon-14

53   (a radioactive isotope) has 6 protons and 8 neutrons and will decay (Figure 1). Isotope

54   geochemistry offers a useful contribution to the Anthropocene debate because

55   anthropogenic impacts on the Earth, such as heavy metal pollution and carbon

56   emissions, often have isotope signatures that are distinct from those that would be

57   expected if natural processes were leading to the same impacts. Isotopes occur in, for

58   example, marine sediments, lake sediments and polar ice. By looking at how isotope

59   ratios change from the top of a sediment or ice core to the bottom, it is possible to

60   reconstruct the intensity of human impacts on the Earth System from the present day

61   back through time. Whilst isotopes do not record every aspect of the Anthropocene,

62   they can contribute important information to the debate. Where they come in

63   particularly useful is in taking our knowledge back further in time than our observations

64   extend, putting human impacts into long-term context, to show when different human

65   influences on the Earth System began across the planet, and the extent to which the

66   various activities are causing variability outside of natural ranges.

67

68   <Insert Figure 1 here>

69

70   Some of the most commonly used isotope ratios are in abundant elements such as

71   carbon and nitrogen. The carbon isotope ratio (δ^{13}C) is the ratio of carbon-13 to carbon-

72   12; the nitrogen isotope ratio (δ^{15}N) is the ratio of nitrogen-15 to nitrogen-14; the sulfur

73   isotope ratio (δ^{34}S) is the ratio of sulfur-34 to sulfur-32; and the boron isotope ratio

74   (δ^{11}B) is the ratio of boron-11 to boron-10. Delta (δ) values are given in per mil (‰,

75   parts per thousand) relative to the international scales of VPDB, AIR, VCDT and

76   (usually) SRM 951 respectively. Lead isotopes are measured relative to a variety of

77   standards, for example SRM 981. Δ^{14}C is the per mil deviation of the carbon-14 to

78   carbon-12 ratio relative to the oxalic acid standard, corrected for fractionation. The

79   abundance of radioisotopes (radioactive isotopes) such as cesium-137 and plutonium-

80   239 are measured in becquerel (Bq) units, with one Bq representing one decay per

81   second. For more details on notation and standardization, see Dean et al. (2014) and

82   references therein.

83

84   Here we discuss the isotope evidence for the global impact of humans on several Earth

85   systems: (1) contamination of the environment by lead through our history of mineral

86   exploitation particularly over the past few millennia, and by sulfur from fossil fuel

87   burning especially since the 1950s; (2) alteration of the global nitrogen cycle due to

88   fossil fuel burning and the production of artificial fertilizers, especially since the 1950s;

89   (3) carbon isotopes tracking greenhouse gas emissions from fossil fuel combustion, the

90   industrialization of agricultural practices and extensive forest clearance, leading to

91   global warming since the Industrial Revolution but especially since the 1950s; and (4)

92   absorption of atmospheric carbon by the oceans, which has led to ocean acidification,

93   especially since the 1950s, at what likely amounts to the greatest rate for ~50 million

94   years. We also demonstrate how radioisotopes could be useful as stratigraphic markers

95   to define the beginning of the Anthropocene in the geological record (e.g. carbon-14,

96   cesium-137 and plutonium-239).

97

98   **Isotope evidence for the Anthropocene**

99

100   *Records of mineral exploitation*

101

102   Humans have been mining and using heavy metals for thousands of years, and the

103   subsequent liberation of heavy metal particles into the atmosphere means that the

104   pollution can spread thousands of kilometers from the source. Lead was one of the first

105   metals to be mined by humans. Different lead ores in different parts of the world can

106   have different lead isotope ratios, so the lead isotope ratio can potentially be used to

107   pinpoint the origin of lead pollution found at a particular location. For example, isotopes

108   have shown that between ~150 BC and AD 50, the majority of the lead particles

109   deposited on Greenland ice came from Roman lead mines in southern Spain. Nowadays,

110   lead isotopes show that a large proportion of the lead pollution in Greenland comes

111   from China. In addition, trends in $^{206}Pb/^{207}Pb$ have been used to trace pollution changes

112   through time. For example, in Sweden, the background (i.e. natural) $^{206}Pb/^{207}Pb$ is

113   around 1.5, while atmospheric lead pollution derived from smelting, leaded petrol and

114   burning of coal has a $^{206}Pb/^{207}Pb$ value of ~1.2 (Renberg et al., 2002). The particles of

115   lead released by these natural and human sources are deposited in lake sediments, and

116   by looking at how the lead isotope ratio changed back through time, the history of

117   anthropogenic lead pollution in Sweden has been reconstructed. There was a decline in

118   the ^{206}Pb/^{207}Pb ratio in Roman times (to ~1.46), indicating increased pollution from

119   anthropogenic activities, then an increase to higher values in the Dark Ages ~AD 500-

120   800 (~1.50) when lead mining and smelting activities must have decreased. Minimum

121   values (~1.22) were reached in the 1970s when leaded petrol consumption peaked

122   (Figure 2) (Renberg et al., 2002). In recent times, with the phasing out of leaded petrol

123   in Europe, there has been an increase in ^{206}Pb/^{207}Pb as anthropogenic sources of lead

124   pollution have diminished (currently ~1.28). The fact that particles of lead have been

125   liberated into the atmosphere by humans, and then carried hundreds and thousands of

126   kilometers from source to pollute Sweden and Greenland, could be used to support the

127   contention of Ruddiman et al. (2011) that human influences on the environment were

128   occurring millennia before the Industrial Revolution. However, although the

129   anthropogenic lead signal may be near hemispheric in Roman times, localized heavy

130   metal pollution in a certain parts of the world might not be considered to constitute a

131   change in the functioning of the Earth System.

132

133   <Insert Figure 2 here>

134

135   In addition to lead, sulfur isotopes can be used to track pollution because, as with lead

136   isotopes, natural (e.g. volcanic eruptions and forest fires) and anthropogenic (e.g.

137   burning of coal in power stations) sources of sulfur often have different isotope ratios:

138   anthropogenic sources of sulfur emissions into the atmosphere are often low in δ^{34}S

139   compared to natural sources. δ^{34}S of sulfate in rainwaters can therefore be used to track

140   changes in anthropogenic sulfur emissions and acid rain through time. In the second

141   half of the twentieth century, there was a particular need to understand the sources and

142   changes in sulfur emissions from power stations as concern over acid rain gained

143   prominence in Europe and North America. Starting with the Convention on Long-

144   Range Transboundary Air Pollution, signed by European and North American countries

145   from 1979 onwards, a series of agreements have led to large decreases in SO_2 levels in

146   the European Union, as traced by increases in $\delta^{34}S$ in rain at some monitoring stations

147   (Dean et al. (2014) and references therein). As with lead pollution, the fact that sulfur

148   emissions have started to decline across much of the planet shows that some

149   anthropogenic impacts on the environment have already peaked, although as with lead

150   pollution, sulfur pollution is far from being the most compelling indicator of the

151   Anthropocene.

152

153   *Changes to the global nitrogen cycle*

154

155   In contrast to lead and sulfur, there has been a continued increase in anthropogenic

156   nitrogen emissions, showing how efforts to limit anthropogenic nitrogen emissions (e.g.

157   nitrous oxides and ammonia) have met with limited success. Fossil fuel emissions and

158   the artificial production of fertilizer have increased the amount of reactive nitrogen

159   (nitrogen compounds such as nitrogen oxides that support biological growth) in the

160   atmosphere, and this is thought to constitute the largest change to the global nitrogen

161   cycle for 2.5 billion years (Canfield et al., 2010). At present, humans fix more N_2 to

162   reactive nitrogen than natural processes do. In 2008 alone, the Haber-Bosch method of

163   producing fertilizer added 9.5×10^{12} mol of reactive nitrogen and fossil fuel burning

164   1.8×10^{12} mol to the Earth System (Canfield et al. (2010) and references therein). Crops

165   like wheat and maize do not use the nitrogen compounds from fertilizers efficiently, so

166   a lot of the reactive nitrogen is wasted and lost to the atmosphere or washed off fields,

167   leading to enhanced aquatic productivity and sometimes to eutrophication of rivers,

168   lakes and coastal waters. Isotopes allow these changes in the global nitrogen cycle to be

169   tracked. Recent declines in δ^{15}N of certain nitrogen compounds such as nitrates can be

170   observed in many geological archives. It is thought that this decline is the result of the

171   release of isotopically-depleted nitrogen from anthropogenic sources: anthropogenic

172   reactive nitrogen sources such as fertilized soils and fossil fuel emissions (Hastings et

173   al., 2009). More recently, however, other explanations for the decline in δ^{15}N in nitrates

174   in particular have been proposed such as changes in the type of fossil fuels being burnt

175   and changes in atmospheric acidity caused by the sulfate and nitrate emissions (e.g.

176   Geng et al., 2014). More research is required to establish better the causes of the δ^{15}N

177   decline.

178

179   Nevertheless, the clear trend in δ^{15}N records appears to be mostly the result of

180   anthropogenic activity. In nitrate (NO_3^-) in ice cores from Greenland (Hastings et al.,

181   2009) and in organic matter from remote lakes in North America and the Arctic (e.g.

182   Wolfe et al., 2013), there have been declines in δ^{15}N from ~AD 1850, but as with δ^{13}C

183   (next section) it is really after ~AD 1950 that the trend becomes pronounced (Figure 3).

184   In Sky Pond Lake in the US Rockies, the changes since the 1950s are without precedent

185   for at least the last 14,000 years (Wolfe et al., 2013). The decline in nitrogen isotopes

186   from ~AD 1850 was probably caused by the increase in the burning of fossil fuels, with

187   the acceleration ~AD 1950 caused by both artificial fertilizer production and fossil fuel

188   burning.

189

190   These records provide a clear example of how important isotope data are in

191   contextualizing human impacts on the Earth System. By using δ^{15}N, we can extend our

192   knowledge of changes in the nitrogen cycle back much further in time than we

193   otherwise would be able to do, to show that changes since the 1950s seem to be outside

194   that of natural variability.

195

196   <Insert Figure 3 here>

197

198   *Changes in the global carbon cycle*

199

200   There have also been substantial changes to the global carbon cycle because of human

201   activities. Small rises in atmospheric methane (CH_4) and carbon dioxide (CO_2) are seen

202   in gas bubbles in ice cores from ~8,000 years ago and have been linked to wetland

203   expansion related to rice production and to widespread forest clearance (Ruddiman et

204   al., 2011). Ruddiman et al. even argue that these changes might have delayed the onset

205   of the next glacial, which if true could constitute an anthropogenic impact on the

206   functioning of the Earth System. Carbon isotopes have been used to support this

207   hypothesis. $\delta^{13}C$ of atmospheric CH_4 ($\delta^{13}CH_4$) from bubbles within ice cores from the

208   late Holocene have values ~−48‰. Some have argued that such low values of $\delta^{13}CH_4$

209   could be accounted for by increased delivery of isotopically low carbon from natural

210   wetlands. However, Ruddiman et al. (2011) suggest this would not have been likely

211   because of the drying of northern monsoonal regions and the cooling of boreal regions

212   in the late Holocene, which would have meant reduced CH_4 emissions from natural

213   wetlands. Instead, they suggest that low $\delta^{13}CH_4$ could be explained by emissions from

214   rice paddies and livestock and anthropogenic burning of grasses. In terms of $\delta^{13}CO_2$,

215   Ruddiman et al. (2011) argue that carbon burial in boreal peat is underestimated and

216   therefore greater human emissions related to forest clearance are required to balance the

217    $\delta^{13}CO_2$ budget (see Dean et al. (2014) and references therein for more details). The

218   'Early Anthropogenic Hypothesis' of Ruddiman is contentious, with many people

219   arguing that human influence on the Earth was not outside of natural variability so far

220   back into the past. The carbon cycle is complex, and it needs to be better understood

221 (better accounting for the natural and anthropogenic drivers of carbon isotopes in carbon

222 dioxide and methane is key to this) in order to robustly test the hypothesis that human

223 actions thousands of years ago did indeed delay the next glacial.

224

225 What is not in doubt is that since the nineteenth century there has been a substantial

226 decline in atmospheric $\delta^{13}CO_2$ concomitant with the increase in atmospheric CO_2

227 concentrations, with the trend to lower values accelerating after ~AD 1950 (Figure 3).

228 Since the 1950s, this has been recorded by direct measurements of atmospheric CO_2 at

229 sites such as the Mauna Loa monitoring station in Hawaii. Further back into the past,

230 the record is locked away as bubbles of CO_2 in ice cores or can be calculated using the

231 carbon in geological archives such as tree rings, corals and foraminifera in marine

232 sediments. The decline in $\delta^{13}CO_2$ and $\Delta^{14}CO_2$ is known as the Suess Effect and is

233 thought to be linked to the burning of fossil fuels with low $\delta^{13}C$ and $\Delta^{14}CO_2$. Fossil fuels

234 are made up of the remains of organisms, mostly plants, that lived millions of years ago.

235 When plants grow, they take up carbon from their surroundings, and plants

236 preferentially take up ^{12}C over ^{13}C, so have low $\delta^{13}C$, because the bonds of the lighter

237 isotope are more easily broken down to participate in organic reactions. As a result, CO_2

238 released from the burning of fossil fuels in power stations, vehicles, etc. contains on

239 average 2% less ^{13}C per mol than atmospheric CO_2, and thus its release back into the

240 atmosphere leads to a decline in atmospheric $\delta^{13}CO_2$. Fossil fuel carbon also has very

241 little ^{14}C (radioactive carbon) because the time between when the organisms died and

242 were laid down in sediments and when humans started to burn them is many thousands

243 of half-lives of ^{14}C. Figure 4 shows data from the Mauna Loa monitoring station, with

244 $\delta^{13}CO_2$ declining (–7.6‰ in 1980 to –8.4‰ in 2014) as CO_2 concentrations in the

245 atmosphere have risen (316 ppm in 1959 to 401 ppm in 2015) (Keeling et al., 2005;

246 updated). In terms of $\Delta^{14}CO_2$, there was a decline in the first half of the twentieth

247   century as fossil fuel emissions increased, followed by an increase in the 1950s and

248   1960s as a result of the vast amounts of neutrons produced by atmospheric

249   thermonuclear bomb testing that increased ^{14}C production in the atmosphere. There has

250   since been a decline again to the present day as a result of the reduction in atmospheric

251   nuclear tests and the continued increase in fossil fuel emissions. The $\delta^{13}C$ and $\Delta^{14}CO_2$

252   changes in the atmosphere have been key in enabling the Intergovernmental Panel on

253   Climate Change to state that there is a "very high confidence" that the main contributor

254   to the observed increase in CO_2 concentrations in the atmosphere since the nineteenth

255   century has been the human burning of fossil fuels, rather than any potential natural

256   causes. This CO_2 rise is believed to be responsible for a large proportion of recent

257   global warming.

258

259   <Insert Figure 4 here>

260

261   As well as causing changes in the functioning of the atmosphere, the release of CO_2

262   from anthropogenic activities has led to ocean acidification as large quantities of CO_2

263   have been absorbed from the atmosphere by the oceans (~30% of the total according to

264   estimates from the Intergovernmental Panel on Climate Change). Ocean acidification

265   has the potential to reduce coral calcification and threaten the survival of coral

266   ecosystems, and this could produce a distinctive event in the future fossil record. For the

267   last hundred years or so, pH records measured directly from seawater samples can be

268   used to assess the acidification trend, but to extend the record further back in time

269   proxies such as boron isotopes can be used. Calcifying organisms, such as foraminifera

270   and corals, incorporate the borate ion from seawater as they grow and $\delta^{11}B$ is pH

271   dependent. Since the Industrial Revolution, as CO_2 levels have risen, there has been a

272   decline in the pH of the oceans by about 0.1 (in what could be the fastest rate of ocean

273   chemistry change in the last 50 million years), charted by a decline in δ^{11}B. This is to be

274   expected, as δ^{11}B from ocean sediment cores has shown that there was ocean

275   acidification when atmospheric CO_2 concentrations rose during previous interglacials

276   (warm periods). This knowledge gained from boron isotopes with regards how ocean

277   acidification and atmospheric CO_2 concentrations were linked in the past allows us to

278   predict that ocean acidification will increase as CO_2 concentrations continue to rise.

279

280   In summary, changes in carbon isotopes record the human influence on the carbon cycle

281   caused by the release of vast quantities of carbon into the atmosphere because of fossil

282   fuel burning. Changes in boron isotopes record another influence of the rise in

283   atmospheric CO_2 concentrations: ocean acidification. As Figure 3 shows, changes in

284   $\delta^{13}CO_2$ and $\delta^{15}N$ are similar, with both showing trends towards lower values through the

285   nineteenth century but with an acceleration after the Second World War at the time of

286   the 'Great Acceleration' (Steffen et al., 2007, 2015) in human activity.

287

288   **A golden spike to define the start of the Anthropocene**

289

290   As well as providing evidence that humans have influenced the Earth System, and thus

291   contributing to the debate vis-à-vis whether the Anthropocene should be defined as an

292   official geological time interval and if so at what time, isotopes may also provide the

293   stratigraphic marker that could define the start of the Anthropocene. While we have

294   discussed the Anthropocene from an Earth System Science perspective, its formal

295   adoption by the geological community needs to be grounded in stratigraphy.

296   Zalasiewicz et al. (2015) have suggested that the start of the Anthropocene could be

297   defined by a Global Standard Stratigraphic Age: by assigning a point from the human

298   calendar at the time when human influences on the Earth System reached a significant

299 level. However, during the current Eon, the standard practice for defining geological

300 time intervals is the use of a golden spike (i.e. a Global Stratotype Section and Point:

301 GSSP). A GSSP is a single reference point found in the geological record at a specific

302 location on Earth that is the physical manifestation of a global change that marks the

303 beginning of the time interval (Waters et al., 2015). For example, the beginning of the

304 Paleogene 66 million years ago is defined by an iridium-enriched layer found in

305 sediments in Tunisia related to the meteorite impact that is believed to have contributed

306 to the extinction of the dinosaurs. The iridium layer is found around the world but the

307 reference point chosen as the GSSP is in Tunisia. There are a number of radioisotopes

308 that show either clear anthropogenic alteration to natural trends (carbon-14) or whose

309 occurrence on Earth is almost entirely the result of human actions (for example, cesium-

310 137 and plutonium-239). What is important to understand here is that whatever marker

311 in the geological record is chosen to set the GSSP, it does not necessarily have to itself

312 be an indicator of substantial human influence on the Earth System; it just has to be

313 something unique and easily identifiable that occurs around the time other isotope and

314 non-isotope proxies have been used to establish where the Anthropocene should start.

315 Cesium-137 and plutonium-239 first appear at detectable levels in the early 1950s,

316 especially after high yield atmospheric thermonuclear weapons testing (as opposed to

317 atomic bomb testing and use over Japan) began. Peak levels were reached in AD 1963/4

318 due to the large number of atmospheric nuclear tests that were carried out in AD 1962

319 before the Partial Nuclear Test Ban Treaty came into force (Figure 5). ^{14}C abundance

320 also starts to increase in the early 1950s; ^{14}C is produced naturally in the atmosphere

321 through the interaction of neutrons with nitrogen atoms, but the production rate was

322 increased by the release of additional neutrons by atmospheric nuclear tests.

323

324 <Insert Figure 5 here>

325

326   These radioisotopes were deposited in geological records, from tree rings to lake

327   sediments, across the Earth and thus provide a precise stratigraphic point in geological

328   archives at around the time of the 'Great Acceleration' in human activity when isotope

329   and non-isotope proxies show an acceleration in human impacts on the Earth System

330   (Steffen et al., 2007, 2015). Plutonium-239 is considered by many to be potentially the

331   most suitable to use as a GSSP since its long half-life (24,110 years) and low solubility

332   mean it will persist in the geological record for longer than many other radioisotopes

333   (Waters et al., 2015), being detectable even by current mass spectrometry for about

334   100,000 years into the future. Its first real appearance in the geological record in the

335   early 1950s around the start of the 'Great Acceleration' could serve as a GSSP.

336

337   **Conclusion**

338

339   Isotopes have certainly recorded the mark that humans have had, and are having, on the

340   Earth System. Lead isotopes can trace human pollution, thousands of kilometers from

341   source, millennia ago, but it would be difficult to argue that this constitutes a change in

342   the functioning of the Earth System. Sulfur isotopes have recorded a more recent

343   pollution phenomenon, acid rain, but the impacts of acid rain are also localized. On the

344   other hand, boron isotopes provide a record of global ocean acidification, which is

345   leading to a substantial change in the functioning of the hydrosphere and marine

346   biosphere. Carbon and nitrogen isotopes show changes from the start of the Industrial

347   Revolution that become particularly pronounced around the time of the 'Great

348   Acceleration'. The change in the former is related to human influence on the carbon

349   cycle (release of carbon from fossil fuel burning, land use change and the

350   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.

Acknowledgments

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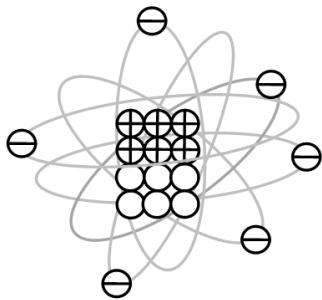
416

417   **Figure captions**

418

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

420   *but the same number of protons.*

421

422   *Figure 2. Trends in lead isotopes and concentrations from the sediments of Lake*

423   *Koltjärn in Sweden, with a decline in $^{206}Pb/^{207}Pb$ taken to indicate increased*

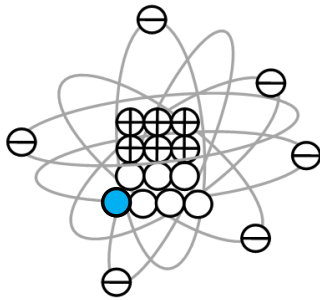424   *anthropogenic contribution to overall lead pollution (Renberg et al., 2002).*

425

426   ***Figure 3.*** *a: $\delta^{13}CO_2$ from bubbles in an Antarctic ice core record (Rubino et al., 2013)*

427   *and $\delta^{13}CO_2$ measured from the atmosphere at the Mauna Loa monitoring station in*

428   *Hawaii (annual average) (Keeling et al., 2005; updated). b: $\delta^{15}N$ from organic matter*

429   *from lake sediments from the US Rockies (3 point moving average, 13 lakes) (Wolfe et*

430   *al., 2013) and from nitrate in Greenland ice cores (Hastings et al., 2009). In both the*

431   *carbon and nitrogen isotope records, a trend to lower values occurs through the*

432   *nineteenth and early twentieth centuries, with an acceleration after ~AD 1950.*

433

434   ***Figure 4.*** *Monthly data from the Mauna Loa monitoring station in Hawaii (Keeling et*

435   *al., 2005; updated) showing an increase in atmospheric CO_2 concentrations from AD*

436   *1958-2015 and a decline in $\delta^{13}CO_2$ from AD 1980 (when monitoring of this began).*
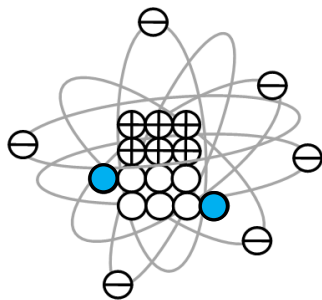
437

438   ***Figure 5.*** *Yields of atmospheric nuclear bomb tests per year shown by bars (UNSCEAR,*

439   *2000) and $^{239,240}Pu$ deposition in Japan (as recorded in water and dust samples) shown*

440   *by the dashed line (Hirose et al., 2000). The yield of atmospheric nuclear tests in the*

441   *atmosphere peaked in AD 1962. $^{239,240}Pu$ in Japan peaked the following year.*

Carbon-12   Carbon-13   Carbon-14

⊖ Electron
⊕ Proton
○ Neutron

a.

Antarctic ice

Mauna Loa

b.

Sediments from 13 lakes in
the US Rockies

Greenland ice