

1 **Title: A bat guano deposit in Jamaica recorded agricultural changes and metal exposure**
2 **over the last > 4,300 years**

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25 **Abstract:**

26 Bats are excellent ecological indicators because they are long-lived, globally distributed,
27 and show predictable responses to environmental stressors. Unaltered bat guano deposits,
28 although rare, can serve as environmental archives to reveal changes in dietary patterns over
29 millennial time scales. We inferred changes in agricultural and industrial practices using a
30 continuous 4,300-year-old bat guano deposit from Jamaica. Cadmium, mercury, lead, and zinc
31 increased during the Industrial Revolution, (which began in ca. 1760), a period characterized by
32 elevated emissions of metals to the atmosphere. Beginning in the early 20th century, decreases in
33 ²⁰⁶Pb/²⁰⁷Pb isotopes tracked the history of leaded gasoline use. Metal concentrations in the bat
34 guano deposit exceeded those recorded in two nearby lake sediment cores from Jamaica. Carbon,
35 nitrogen, and sulfur stable isotope profiles in bat guano tracked the agricultural history of
36 Jamaica, specifically the introduction of nitrogen fertilizers, sugarcane, and possibly fungicides.
37 Bat populations are under stress globally, and such intact guano deposits provide potentially
38 critical information on long-term changes in their food source and exposure to metals.

39

40 **Keywords:** carbon, nitrogen, lead, mercury, cadmium, isotopes

41 **1 Introduction**

42 Present across all continents except Antarctica, there are ~1,300 species of bats that
43 comprise ~20% of all mammal taxa (Fenton and Simmons 2014). Bats occupy a range of trophic
44 levels, and serve as pollinators, natural insecticidal agents, and seed dispersers (Kunz et al.
45 2011). Furthermore, at peak lactation, bats can consume over 100% of their body mass in a
46 single night (Kunz et al. 2011). Bats may be exposed to high concentrations of metals, some of
47 which passes in their guano (Afonso et al. 2016). Caves in which they roost can house large
48 guano deposits that may provide long-term records of bat exposure to metals. If intact deposits
49 can be located, radiometric dating methods can be combined with stratigraphic elemental
50 analyses to infer long-term changes in metal exposure.

51

52 *1.1 Using bat guano deposits to infer bat exposure to metals*

53 Many anthropogenic activities contribute to the concentration of metals in the environment
54 (Nriagu and Pacyna 1988). In particular, the onset of the Industrial Revolution in the late 1700s
55 and the introduction of leaded gasoline in the early 20th century contributed to atmospheric metal
56 concentrations in aerosols and soils (Nriagu 1990). Atmospherically released metals can enter
57 food webs, resulting in elevated concentrations of metals in bats (Pikula et al. 2010). Detectable
58 concentrations of metals were recovered previously from bat guano deposits. For example, lead
59 and copper concentrations in a 30,000-year-old bat guano deposit in the Philippines reached peak
60 concentrations of ~10 and >1000 ppm, respectively (Bird et al. 2007). Similarly, copper
61 concentrations in a 13,000-year-old bat guano deposit from Borneo reached ~8,000 ppm, with
62 lower concentrations of other metals such as chromium, vanadium, and zinc (Wurster et al.
63 2017). Copper, lead, and zinc profiles were also reported for a ~3,000-year-old bat guano deposit

64 in Romania (Onac et al. 2015). Given the concentrations to which metals can be detected in bat
65 guano deposits, and the well-recorded release of metals to the atmosphere from industrial
66 activity, it is interesting that metal concentrations in the aforementioned bat guano deposits did
67 not fluctuate coeval with industrial practices.

68 Lake sediment cores are commonly used to infer past environmental conditions and have
69 been employed to document increases in the metal concentrations that resulted from past human
70 activities (e.g.: Cooke et al. 2007; Cooke and Abbott 2008; Laird et al. 2014). For example, 11th
71 century metallurgy and 16th century silver mining in the Andes left a legacy of higher
72 concentrations of zinc and copper, and lead in Peruvian lake sediments, respectively (Cooke et
73 al. 2007). 20th century deposits are commonly enriched in metals, owing to high levels of
74 industrial activity (Cooke and Abbott 2008; Engstrom et al. 2014).

75

76 *1.2 Tracking past agricultural changes using bat guano deposits*

77 Several studies examined stratigraphic variations of $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ in bat guano deposits that
78 ranged in age from mid-20th century to 32,000 yr BP (Wurster et al. 2007; Wurster et al. 2017;
79 Widga and Colburn 2015; Bird et al. 2007; Mizutani et al. 1992). $\delta^{13}\text{C}$ values in bat guano have
80 been correlated to precipitation and photosynthetic pathway type, where higher values associated
81 with greater summer precipitation and C4 grasses (Wurster et al. 2007). Accordingly, $\delta^{13}\text{C}$
82 values in bat guano deposits were lowest during periods of drought (Wurster et al. 2017; Onac et
83 al. 2014). $\delta^{13}\text{C}$ values in guano have also been used to track bat dietary preferences, with lower
84 $\delta^{13}\text{C}$ values indicative of a diet dominated by C3 vegetation (Widga and Colburn 2015). A fall in
85 $\delta^{13}\text{C}$ values in a bat guano deposit was also correlated to agricultural practices: historically
86 elevated $\delta^{13}\text{C}$ values (C4 plants) corresponded to a period of deforestation and lower present-day

87 $\delta^{13}\text{C}$ values (C3 plants) corresponded to forests (Bird et al. 2007). $\delta^{15}\text{N}$ values may be more
88 susceptible to diagenesis, and as a result, it is difficult to associate $\delta^{15}\text{N}$ profiles with past climate
89 conditions (Wurster et al. 2017; Bird et al. 2007). Despite difficulties associated with
90 interpretation of nitrogen stable isotopes, greater $\delta^{15}\text{N}$ values in bat guano deposits have been
91 associated with insectivorous bats feeding at higher trophic level (Bird et al. 2007). $\delta^{15}\text{N}$ was
92 also used to identify the dietary proportions of aquatic and terrestrial vegetation in a bat guano
93 deposit (Ahmad and Davies 2017).

94

95 *1.3 Objectives*

96 Bat guano was widely mined for gunpowder and fertilizer, such that long sequences of bat
97 guano deposits are now scarce. Consequently, bat guano is rarely used as an archive for
98 paleoenvironmental inference. We present results from one of only a few intact guano deposits
99 remaining in Jamaica, the Home Away from Home (HOM) cave. We set out to analyze a
100 continuous record of bat guano in a core from the cave for stable and radiogenic isotopes and
101 metals. Specifically, our aim was to gain insights into past agricultural practices (using $\delta^{15}\text{N}$,
102 $\delta^{13}\text{C}$, and $\delta^{34}\text{S}$) and industrial activity (using cadmium, lead, mercury, and zinc) in this part of
103 Jamaica over the past four millennia. Lake sediment cores are frequently used to infer past
104 environmental conditions, and we compared the history recorded in the bat guano deposit to
105 histories from two, nearby, Jamaican lake sediment cores. Our guano analyses revealed a history
106 of bat community exposure to contaminants, stemming from a range of anthropogenic activities
107 over the last ~4,300 years.

108

109 **2 Materials and methods**

110 2.1 Study sites

111 The bat guano core was collected from Home Away from Home (HOM) cave, in Trelawny
112 Parish, Jamaica. The exact location of the cave is kept confidential for conservation reasons, but
113 inquiries from qualified researchers can be sent to the Jamaican Caves Organization. The cave is
114 well protected from local disturbances, as it is accessible only with technical climbing gear.
115 There is no evidence of human activity beyond the cave entrance. The cave temperature ranges
116 between 25 and 30°C and humidity approaches 100% during the rainy season. The HOM core
117 was taken from the deepest deposit within the cave, located approximately 30 m beneath the
118 entrance. An estimated 5,000 bats currently occupy HOM cave. Five bat species were identified
119 using a combination of sonar (Song-Meter SM2BAT), mist netting, and a taxonomic key created
120 by Dr. M. Brock Fenton and students: insectivorous *Pteronotus parnelli*, *Macrotus waterhousii*,
121 and *Mormoops blainvillii*, nectivorous *Glossophaga soricina*, and frugivorous *Artibeus*
122 *jamaicensis*. The HOM core was collected by excavation at the apex of the tallest guano deposit.
123 10 x10-cm cross-sections of bat guano were collected at 1-cm intervals to a depth of 129 cm.
124 Samples were stored in Whirl Pak® bags at -20°C until analysis.

125 Sediment cores were collected from Crystal Lake (18° 27' 39.71'' N, 77 22' 47.63'' W)
126 and Wakefield Lake (18° 24' 47.30" N, 77° 43' 42.95" W) using a 3-inch Glew and Smol (Glew
127 and Smol 2016) deep water corer. Cores were sectioned into 0.5-cm intervals, that were
128 refrigerated at 4°C and then freeze-dried for analysis. Crystal Lake is located 512 m from the
129 ocean and 33 km east of HOM cave. It is relatively free from human activity, with the exception
130 of local fishing and a highway to the north. Wakefield Lake is located 9.5 km northwest of HOM
131 cave, within a town that floods frequently and is thought to have existed only since the 1800s.

132

133 2.2 ²¹⁰Pb and ¹⁴C dating

134 ²¹⁰Pb dating and ¹³⁷Cs measurements on guano from the HOM deposit were accomplished
135 using an Ortec High Purity Germanium Gamma Spectrometer (Oak Ridge, TN, USA) at the
136 University of Ottawa. Measurements on samples from the lake cores were run on gamma
137 detectors at Queen's University. Certified Reference Materials, obtained from International
138 Atomic Energy Association (Vienna, Austria), were used for efficiency corrections, and results
139 were analyzed using ScienTissiME (Barry's Bay, ON, Canada). ¹³⁷Cs measurements were made
140 to confirm ²¹⁰Pb dates calculated using the CRS (constant rate of supply) model. ²¹⁴Pb activity,
141 which was used as a proxy for supported ²¹⁰Pb activity, remained relatively low and constant
142 throughout each profile. In the bat guano deposit, an average background value of 56.5 Bq kg⁻¹
143 was used to construct the dating profile. In the Crystal and Wakefield lake sediment cores, ²¹⁰Pb
144 approached background at 16.25 cm and 32.25 cm, respectively (Figure S3). ²¹⁴Pb activity was
145 averaged in the lake sediment cores; activity was set to a value of 37.11 Bq kg⁻¹ for Crystal Lake
146 and 47.49 Bq kg⁻¹ for Wakefield Lake.

147 Lipid-treated (lipids removed using dichloromethane and methanol) and un-treated samples
148 from three selected depths in the guano profile (14.5, 56.5, and 85.5 cm) were ¹⁴C-dated by
149 accelerator mass spectrometry at the Poznań Radiocarbon Laboratory in Poland. Because ¹⁴C
150 dates from the paired lipid-treated and untreated samples were similar (Table S1), ¹⁴C dates were
151 run on untreated samples at other depths in the profile. Radiocarbon dates were calculated using
152 OxCal 2013 and age calibrations were conducted using OxCal 4.1.5 software.

153

154 2.3 Metals

155 Metals were extracted by digesting freeze-dried guano in HNO₃ and H₂O₂ in a DigiPREP

156 Jr. (SCP Science, Baie D'Urfé, QC, Canada) in accordance with USEPA Method 3050b. Samples
157 were analyzed for total metals using an Agilent inductively coupled plasma mass spectrometer
158 (ICP-MS) (Santa Clara, CA, USA). Reference material was NIST 8704 Buffalo River Sediment.
159 Method detection limits are shown in Table S2. The standard error of triplicates in the lake
160 sediment samples ranged between 0.02 and 5.82 $\mu\text{g kg}^{-1}$ for each metal (Table S3). Samples
161 were re-analyzed for lead isotopes by ICP-MS; the standard error of triplicates in the lake
162 sediment samples ranged between 0.001 and 0.097. Approximately 10 mg of freeze-dried bat
163 guano was analyzed for total mercury on a NIC Mercury Analyzer 3000 (Bryan, TX, USA),
164 using USEPA method 7473. Reference material was NRC MESS-3 Marine Sediment 91+/-9 ng
165 g^{-1} total mercury; the method detection limit was 0.2 ng g^{-1} . In addition to normalizing the total
166 metal concentrations to organic carbon, the metal concentrations were also normalized to
167 titanium concentrations to account for natural weathering and ensure that any fluctuations in the
168 metal profiles were reflective of anthropogenic activity. This standardization accounts for the
169 natural variations in metal flux within the guano deposit as some total metals are immobilized
170 within titanium lattices (Last and Smol 2001).

171

172 *2.4 Carbon, nitrogen, and sulfur isotopes*

173 Percent carbon, nitrogen, and sulfur were analyzed using a Micro Cube elemental analyzer
174 at the Ján Veizer Stable Isotope Laboratory (formerly G.G. Hatch SIL), located at the University
175 of Ottawa. TOC was determined by analyzing samples after they were acid desiccated using
176 HCl. Organic $\delta^{13}\text{C}$ (‰ V-PDB), hereafter, referred to as $\delta^{13}\text{C}$, and $\delta^{15}\text{N}$ (‰ air) were analyzed
177 using an elemental analyzer interfaced to an isotope ratio mass spectrometer (IRMS). Samples
178 were normalized to the following internal standards: C-51 Nicotiamide ($\delta^{15}\text{N}$: +0.07 ‰, $\delta^{13}\text{C}$: -

179 22.95 ‰), C-52 ammonium sulphate and sucrose ($\delta^{15}\text{N}$: +16.58 ‰, $\delta^{13}\text{C}$: -11.94 ‰), and C-54
180 caffeine ($\delta^{15}\text{N}$: -16.61 ‰, $\delta^{13}\text{C}$: -34.46 ‰). The blind standard was C-55 glutamic acid ($\delta^{15}\text{N}$: -
181 3.98 ‰, $\delta^{13}\text{C}$: -28.53 ‰). Results are reported in delta notation (δ) where $\delta = ((R_x -$
182 $R_{\text{std}})/R_{\text{std}}) * 1000$; R = ratio of the abundance of the heavy to light isotope, x = sample, and std
183 = standard. $\delta^{15}\text{N}$ values were calibrated to the following international standards: IAEA-N1 (+0.4
184 ‰), IAEA-N2 (+20.3 ‰), USGS-40 (-4.52 ‰), and USGS-41 (+47.57 ‰). $\delta^{13}\text{C}$ values were
185 calibrated to the following international standards: IAEA-CH-6 (-10.4 ‰), NBS-22 (-29.91 ‰),
186 USGS-40 (-26.24 ‰), and USGS-41 (+37.76 ‰). Analytical precision was ± 0.2 ‰ using
187 glutamic acid. $\delta^{34}\text{S}$ (‰ VCDT) was analyzed using a Vario Micro Cube elemental analyzer
188 (Langensfeld, Germany) interfaced *via* a conflo IV to a Thermo Finnigan Delta XP IRMS
189 (Bremen, Germany).

190

191 **3 Results and discussion**

192 *3.1 Dating an ancient bat guano deposit*

193 'Unsupported' ^{210}Pb reached radiogenic background at a depth of 14.5 cm in the HOM core
194 (see Methods), corresponding to ca. 1900 CE. We used ^{137}Cs to corroborate the ^{210}Pb dating
195 profile: ^{137}Cs was produced solely from nuclear weapons testing, and most lake sediments show
196 highest activity in 1963 CE, associated with the peak in such testing. ^{137}Cs activity peaked at 8.5
197 cm depth in the guano, corroborating that the depth corresponded to the early-1960s, as
198 determined by ^{210}Pb dating (Figure S1). Although ^{137}Cs dating is widely used in lake and peat
199 deposits, it has not been attempted previously in bat guano archives. Our study reveals that it is
200 possible to reconstruct the history of nuclear weapons testing using ^{137}Cs in bat guano and that it
201 can be used to validate ^{210}Pb dating models.

202 At 14.5 cm, there was a 580-year difference between the ^{210}Pb date (1900 CE) and the
203 calibrated ^{14}C date (1320 CE). The difference between the ^{210}Pb and ^{14}C dates may be attributed
204 to the dissolution of the limestone cave onto the bat guano deposit, which would contribute dead
205 carbon (carbon lacking ^{14}C) into the system and/or from the bat diet itself, where bats feeding on
206 aquatic insects can incorporate any reservoir effect from the environment into their system.
207 Consequently, we applied a 580-year reservoir effect correction to this bat guano deposit to
208 account for the different dates given by the ^{210}Pb and ^{14}C dating methods. A 2,000-year reservoir
209 effect correction was applied to a bat guano deposit from the Philippines, to account for the
210 deposition of weathered graphite from a marble cave (Bird et al. 2007). After accounting for the
211 reservoir effect in our guano deposit, the calibrated ^{14}C date on the oldest layer (128.5 cm) was
212 2307 BCE, indicating the HOM core spans the last ~4,300 years.

213

214 *3.2 History of metal exposure to bats*

215 The increase in lead concentrations in the HOM core coincided with the onset of the
216 Industrial Revolution ca. 1760 CE, a period of increased mining, coal combustion, and waste
217 incineration (Figure 1). (Clark 1979) reported lead concentrations ($61\text{-}65\ \mu\text{g g}^{-1}\ \text{dw}$) in guano
218 collected from bats living in proximity to a highway, suggesting that leaded gasoline was the
219 major contributor of lead to the bat's diet.

220 Lead isotope analyses, e.g. the $^{206}\text{Pb}/^{207}\text{Pb}$ ratio, were used to “fingerprint” the source of
221 additional lead (Komarek et al. 2008; Teutsch et al. 2001). The ratio decreased in the bat guano
222 deposit from 1.512 to 1.142 starting in the 1920s CE (Figure 1). Values approached those typical
223 of leaded gasoline from Australia (~1.04), Europe (1.115), Mexico (1.202-1.204), the United
224 States (1.04-1.39), and Canada (0.92-1.19) (Weiss et al. 1999; Erel et al. 1997; Sañudo-

225 Wilhelmy and Flegal 1994; Sturges and Barrie 1987). Similar $^{206}\text{Pb}/^{207}\text{Pb}$ values were observed
226 in the Jamaican lake sediments (Table S3, Figure 2, and Figure 3). Neither lake sediment core
227 was old enough to capture pre-industrial lead. Nevertheless, lead ratio values in the lake
228 sediments were more similar to values in 20th-century bat guano than to values in pre-industrial
229 guano (Figure 3). $^{208}\text{Pb}/^{206}\text{Pb}$ can identify the source of Pb used in leaded gasoline because
230 isotope ratios from North American ores (2.06) were generally lower than values in European
231 ores (2.145) (Teutsch et al. 2001). Accordingly, industrial-period $^{206}\text{Pb}/^{207}\text{Pb}$ (1.228) and
232 $^{208}\text{Pb}/^{206}\text{Pb}$ (1.994) values in the bat guano (Table S4) generally reflect leaded gasoline from
233 North America. Despite the absence of major roads within several kilometers of the HOM
234 deposit, the lead isotope profile suggests that the lead was carried into the foraging area of the
235 bats and transferred to them through their food web. The slight increase in the lead isotopic ratio
236 at the beginning of the 21st century may reflect a lagged response to removal of lead from
237 gasoline in the early-mid 1980s (Figure 1). Notably, the average crustal $^{206}\text{Pb}/^{207}\text{Pb}$ ratio (~1.2) is
238 similar to the ratio in pre-industrial guano (Weiss et al. 1999; Farmer et al. 2000). The
239 $^{208}\text{Pb}/^{207}\text{Pb}$ vs $^{206}\text{Pb}/^{207}\text{Pb}$ profile (Figure 3) further supported the shift from a natural to
240 anthropogenic lead source, as lower values of both ratios indicated anthropogenic origin
241 (Emmanuel and Erel 2002). $^{206}\text{Pb}/^{204}\text{Pb}$ can also distinguish between natural (18.5-19.5) and
242 anthropogenic (16-18.5) lead sources (Hansmann and Köppel 2000). Accordingly, $^{206}\text{Pb}/^{204}\text{Pb}$
243 decreased in industrial-era guano (Figure 1) and was typically < 18.5 in the lake sediments
244 (Table S3).

245 The range of mercury concentrations in this bat guano deposit (Table S4: 0.09-1.72 $\mu\text{g g}^{-1}$
246 dw OC) resembled those reported in fresh bat guano samples collected from historically
247 industrialized areas (0.12-0.26 $\mu\text{g g}^{-1}$ dw) and from rural environments (0.08-0.11 $\mu\text{g g}^{-1}$ dw)

248 (O'Shea et al. 2001). Mercury concentrations rose in this deposit at the onset of the Industrial
249 Revolution and the rise in mercury concentrations we recorded in guano from the 20th century is
250 consistent with the timing of an increase in Hg concentrations observed in North American lake
251 sediments (Engstrom et al. 2014). Interestingly, the slow rise in mercury beginning in ~1400
252 BCE coincided with the earliest record of mercury mining in Peru (Cooke et al. 2009). Mercury
253 amalgamation was introduced to Jamaica in ~1600 CE and may have contributed to the rise in
254 mercury observed within the HOM core. Notably, bat guano had higher mercury than other
255 mammalian faeces, such as that of otters (0.02-0.17 $\mu\text{g g}^{-1}$ dw (Ramos-Rosas et al. 2013)).
256 Elevated mercury concentrations were also observed in bat livers (14.0-151 $\mu\text{g g}^{-1}$ dw (Nam et al.
257 2012)), hair (0.04-145.27 $\mu\text{g g}^{-1}$ dw (Becker et al. 2018)), and brains (0.41-18.7 $\mu\text{g g}^{-1}$ dw (Nam
258 et al. 2012)).

259 Zinc concentrations in the HOM core (Table S4) generally exceeded those observed in
260 fresh bat guano (64-1,080 $\mu\text{g g}^{-1}$ dw (Zukal et al. 2015)). Furthermore, zinc bioaccumulates and
261 is readily transferred from the lithosphere to biota (Brimble et al. 2009), which explains the much
262 lower zinc concentrations observed in the Jamaican lake sediment cores (Table S3: 4.93-15.7 μg
263 g^{-1} dw). Anthropogenic emissions of zinc were reflected within this bat guano deposit. The
264 concentration of zinc peaked at 6,200 $\mu\text{g g}^{-1}$ total organic carbon (TOC) during the Industrial
265 Revolution, during which bauxite mining was introduced to Jamaica (Rainford and Richards
266 2008). Similarly, Norwegian Arctic lake sediment cores influenced by seabird guano showed an
267 increase in zinc concentrations as a result of the Industrial Revolution (196 $\mu\text{g g}^{-1}$ dw in pre-
268 industrial deposits to a peak of 434 $\mu\text{g g}^{-1}$ dw in 1996) (Evenset et al. 2007).

269 Previously reported cadmium concentrations in bat guano (0.03-8.5 $\mu\text{g g}^{-1}$ dw (Zukal et al.
270 2015)) and Jamaican lake sediments (Table S3: 0.03-0.28 $\mu\text{g g}^{-1}$ dw) were much lower than those

271 in the HOM core (Table S4). The concentration of cadmium in the bat guano deposit (median =
272 92.8 $\mu\text{g g}^{-1}$ dw) also exceeded that found in the hair, livers, and kidneys of bats (0.02-0.10 $\mu\text{g g}^{-1}$
273 dw) (Hernout et al. 2016). Elevated cadmium concentrations in the HOM core may reflect the
274 naturally high cadmium concentrations in soils from Jamaica (298-978 $\mu\text{g g}^{-1}$ dw) compared to
275 soils in other countries (Garrett et al. 2008). About 67% of Jamaica is covered by limestone,
276 which often contains phosphate-bearing minerals such as phosphorite, with cadmium
277 concentrations as high as 6,000 $\mu\text{g g}^{-1}$ dw (Garrett et al. 2008; Garrett et al. 2010; Draper 1986).
278 Moreover, soil collected above white limestone typically has 32 times more cadmium than
279 average Jamaican soil (Garrett et al. 2008). Consequently, bats at HOM cave are exposed to
280 naturally elevated cadmium concentrations in the soil as well as elevated cadmium in the white
281 limestone cave they inhabit. The cadmium rise in the HOM core was comparable to the 3-fold
282 increase in cadmium observed in Peruvian lake sediments, which was attributed to industrial
283 activity (Cooke and Abbott 2008).

284

285 *3.3 Stable isotopes and dietary trends*

286 The $\delta^{15}\text{N}$, $\delta^{13}\text{C}$, and $\delta^{34}\text{S}$ composition in this deposit reflects agricultural practices over the
287 past ~4,300 years. In this deposit, $\delta^{13}\text{C}$ averaged -26.5 ‰ until the 1670s CE, at which point,
288 $\delta^{13}\text{C}$ increased, reaching a maximum value of -22.09 ‰ at 2.5 cm (Figure 4). Typical C3 plants
289 have lower $\delta^{13}\text{C}$ values (-24 to -34 ‰) than C4 plants (-12 to -23 ‰) (Smith and Epstein 1971),
290 and thus the HOM core appears to have carried a C3 plant signal throughout with a slight C4
291 component. $\delta^{13}\text{C}$ in bat guano can fluctuate with the relative abundance of C4 grasses (Wurster et
292 al. 2007). Thus, although $\delta^{13}\text{C}$ values in the most recently deposited bat guano did not reach the
293 average $\delta^{13}\text{C}$ values found in C4 plants, the increase in $\delta^{13}\text{C}$ likely indicates a shift toward a

294 more C4 plant-based diet. The rise coincided with the agricultural history of Jamaica. The Taíno
295 people, who first arrived in Jamaica in ~650 CE, used maize, a C4 plant, which may have
296 contributed to the first gradual increase in $\delta^{13}\text{C}$ (Santos et al. 2013; Mickleburgh and Pagan-
297 Jimenez 2012). Christopher Columbus subsequently introduced sugarcane, another C4 plant, to
298 the Caribbean in the early 16th century (Found and Berbés-Blázquez 2012), which would have
299 contributed further to the rise in $\delta^{13}\text{C}$ observed in the HOM core. Notably, there is a sugarcane
300 plantation ~3 km north of HOM cave, thus foraging bats may consume insects supported by a
301 C4-influenced diet.

302 The median $\delta^{15}\text{N}$ value was +12.9 ‰ (range: +9.8 to +18.5 ‰) throughout the guano
303 deposit (Figure 4). $\delta^{15}\text{N}$ increased steadily in ca. 10th century CE, reached a value of +16.3 ‰ in
304 the 1870s CE, and then decreased to the lowest value of +9.8 ‰ by 2008 CE. Declining $\delta^{15}\text{N}$ in
305 post-industrial deposits may reflect NO_x emissions from coal combustion, as coal $\delta^{15}\text{N}$ values are
306 approximately 0 ‰ (Ding et al. 2018; Peterson and Fry 1987). $\delta^{15}\text{N}$ can also decrease with
307 increasing anthropogenic emissions from fossil fuel combustion and synthetic fertilizers (Freyer
308 1978). World-wide use of synthetic nitrogen fertilizers incorporate atmospheric nitrogen and thus
309 have lower $\delta^{15}\text{N}$ values (-1.7 to +3.9 ‰) (Vitoria et al. 2004). Lower $\delta^{15}\text{N}$ values observed post-
310 1870 CE may also be influenced by decomposition (Peterson and Fry 1987). The slow and
311 gradual rise in $\delta^{15}\text{N}$ pre-1870 CE may have resulted from increased ^{15}N from intensive manure
312 fertilization, which can increase $\delta^{15}\text{N}$ values to ~+7 ‰ (Bogaard et al. 2013). Manure-based
313 fertilizers were first used by Neolithic farmers (Bogaard et al. 2013), and elevated $\delta^{15}\text{N}$ values
314 have also been observed in manure-influenced soils from the Bronze Age (Simpson et al. 1999).
315 Consequently, consumption of plants and insects that were in contact with manure fertilizers and
316 subsequently, synthesized fertilizers, could account for the shifts in the $\delta^{15}\text{N}$ values in the HOM

317 core.

318 Changes in dietary $\delta^{34}\text{S}$ are reflected in bat guano (Salvarina et al. 2013), so long-term
319 trends in dietary sulfur accumulation should also be traceable in the HOM core. The median $\delta^{34}\text{S}$
320 value was +18.0 ‰ (ranging from +9.7 to +22.4 ‰) (Figure 4). $\delta^{34}\text{S}$ was relatively constant pre-
321 1930 CE, and then decreased post-1930 CE, and reached a minimum of +9.7 ‰ in ca. 1990 CE.
322 The $\delta^{34}\text{S}$ values post-1930 CE began to approach sulfate $\delta^{34}\text{S}$ values associated with coal
323 combustion, which are typically $< +10$ ‰ (Guo et al. 2016). Therefore, the HOM core could be
324 tracking the increase in industrially sourced sulfur. The decrease in $\delta^{34}\text{S}$ could also be attributed
325 to the use of fertilizer. With the exception of superphosphate fertilizers, which display a wide
326 range of $\delta^{34}\text{S}$ values, from -4.2 to +17.2 ‰ (Chalk et al. 2017)), most fertilizers tend to have $\delta^{34}\text{S}$
327 values $< +12$ ‰ (Chalk et al. 2017). In particular, the prominent decrease in $\delta^{34}\text{S}$ in the early
328 1930s CE coincided with the introduction of a CuSO_4 fungicide, Bordeaux mixture, introduced to
329 Central America to combat plant pathogenic fungi in the genus *Cercospora* (Wardlaw 1941).
330 Bordeaux mixture typically contains 257-276 g kg^{-1} of copper (Arena et al. 2018), which may
331 account for the elevated copper concentrations in the HOM core (Table S4). Soil-enriched
332 copper has been observed in Bordeaux-sprayed areas, in one instance reaching 4,000 $\mu\text{g g}^{-1}$ dw
333 (Thrupp 1991). Copper is readily bound to organic matter (Couto et al. 2015), which may explain
334 the greater copper concentrations in the bat guano relative to the soil. In the HOM core, Cu/Ti
335 peaked in guano deposited in ca. 50 BCE (Figure 1), possibly the result of the mobility of copper
336 in the deposit. Bordeaux mixture has been seen to leach 40 cm down-core in lake sediments,
337 which can be attributed to the more mobile Cu^{2+} in the presence of dissolved organic matter
338 (Couto et al. 2015). Cu^{2+} is prevalent in soils ($> +43.5$ ‰) when the pH is closer to neutral (4 to
339 7) (Komarek et al. 2009). Fresh bat guano typically has pH values ranging from 5.1 to 8.0

340 (Shahack-Gross et al. 2004; Wurster et al. 2015); the pH then becomes more acidic over time,
341 ranging from 2.7 to 5.3 (Wurster et al. 2015). Consequently, the copper observed in the HOM
342 core may have leached downwards as Cu^{2+} until the pH increased to the point that it no longer
343 favoured the mobile Cu^{2+} form. The use of fertilizers and fungicides, as well as the increase in
344 anthropogenic activity in the 20th century, likely drove the decrease in $\delta^{34}\text{S}$ values observed in
345 the HOM core.

346

347 **4 Conclusions**

348 The HOM core provided a rare glimpse into the past ~4,300 years of tropical bat exposure
349 to multiple anthropogenic influences, most notably metal contamination and changing
350 agricultural practices. Bats that used this remote Jamaican cave were exposed to anthropogenic
351 metals, highlighting the importance of emission controls to reduce the risk of exposure to these
352 contaminants. Bat populations around the world are under stress and declining as a consequence
353 of human activities. Guano archives, although scarce, provide new insights into past diets and
354 contaminant exposure. As such, these deposits can be used to monitor the spatial and temporal
355 movement of contaminants that affect bats, humans, and other species.

356

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363

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365

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552

553 Figures for

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555 A bat guano deposit in Jamaica recorded agricultural changes and metal exposure over the last >
556 4,300 years

557

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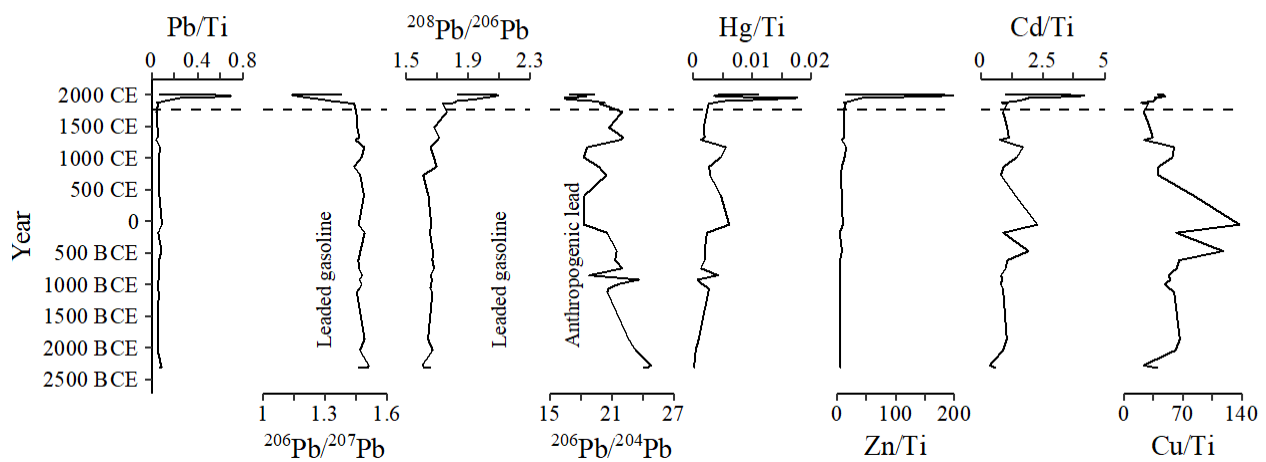
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569 **2 column fitting**

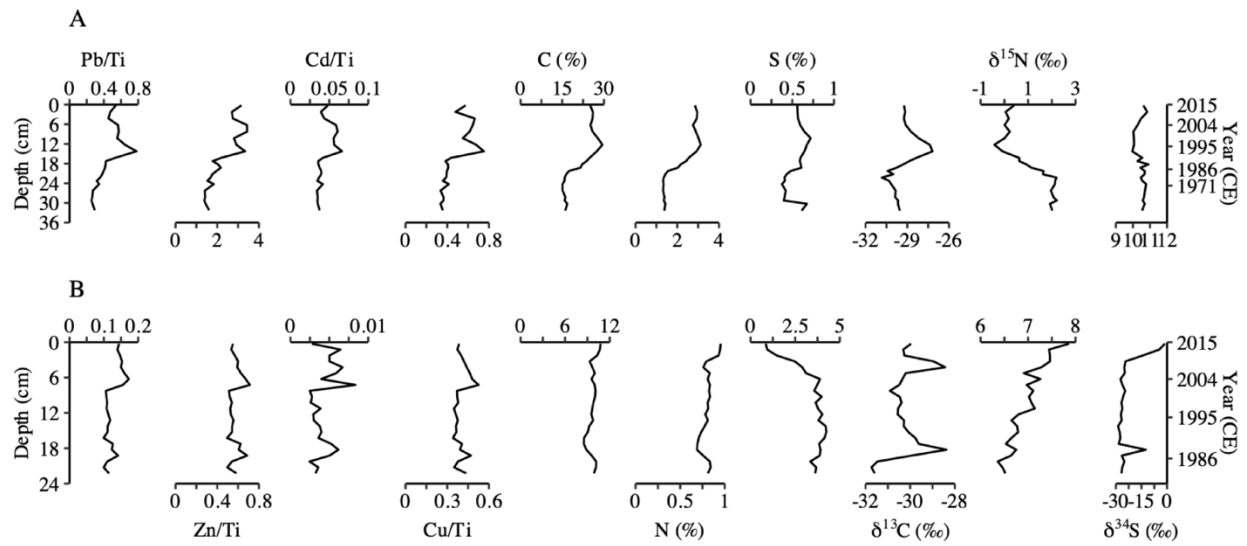


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572 Figure 1: Lead ratios and metal concentrations in the HOM core. Metal concentrations are
573 normalized to titanium in order to account for natural weathering. The dashed line separates bat
574 guano deposited before and after 1760 (the year of the start of the Industrial Revolution). ²¹⁰Pb
575 and ¹⁴C years are presented on the y-axis. The shaded area indicates the range of values reported
576 in leaded gasoline for ²⁰⁶Pb/²⁰⁷Pb (Sturges and Barrie 1987) and ²⁰⁸Pb/²⁰⁶Pb (Teutsch et al. 2001),
577 and in the case of ²⁰⁶Pb/²⁰⁴Pb, anthropogenic lead (Hansmann and Köppel 2000).

578 **2 column fitting**



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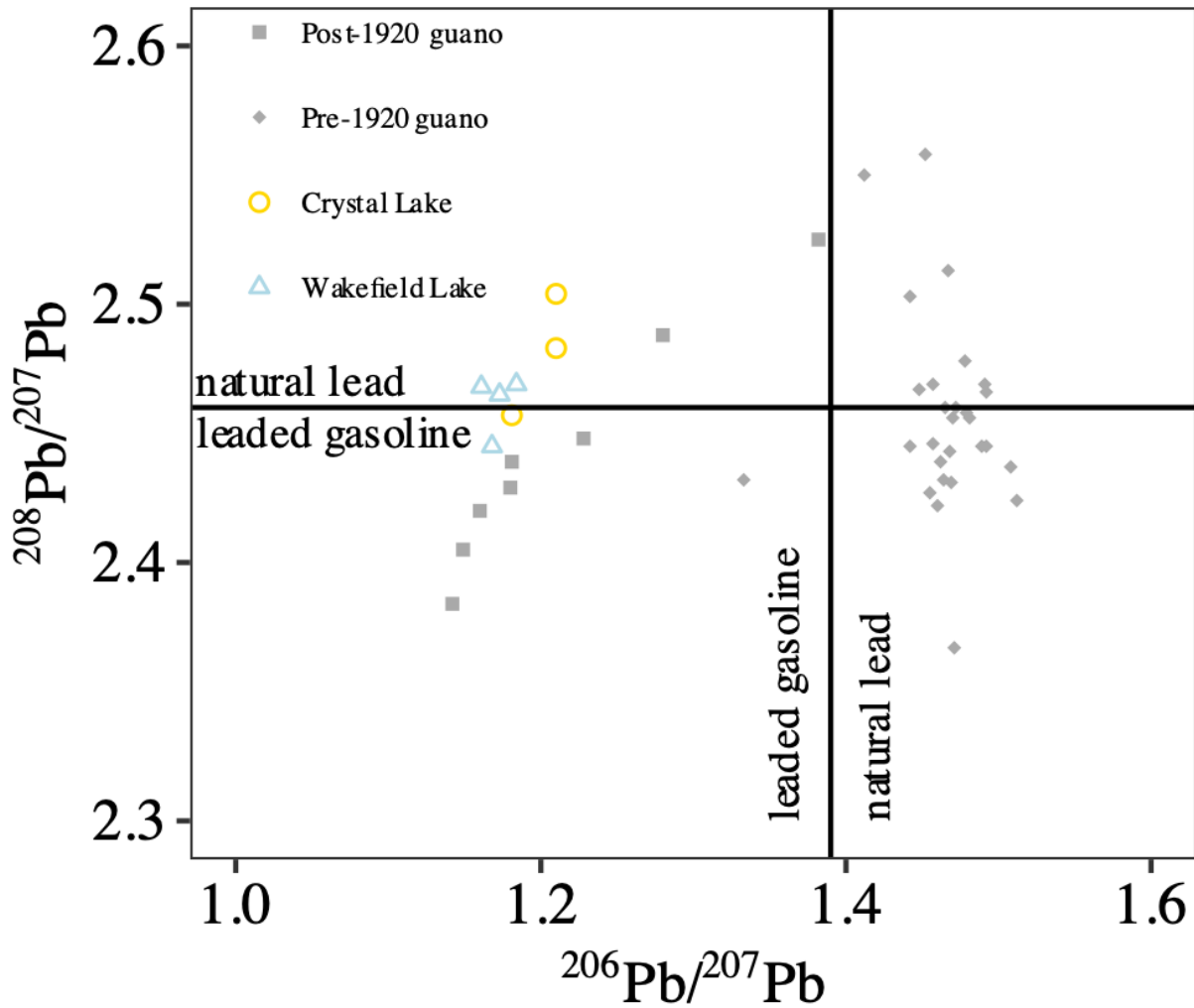
581 Figure 2: Metal, elemental, and stable isotope profiles in Wakefield Lake (panel A) and Crystal

582 Lake (panel B) sediment cores. Metal concentrations are normalized to titanium to account for

583 natural weathering. Select ²¹⁰Pb dates are presented on the right y-axis.

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585 **1 column fitting**

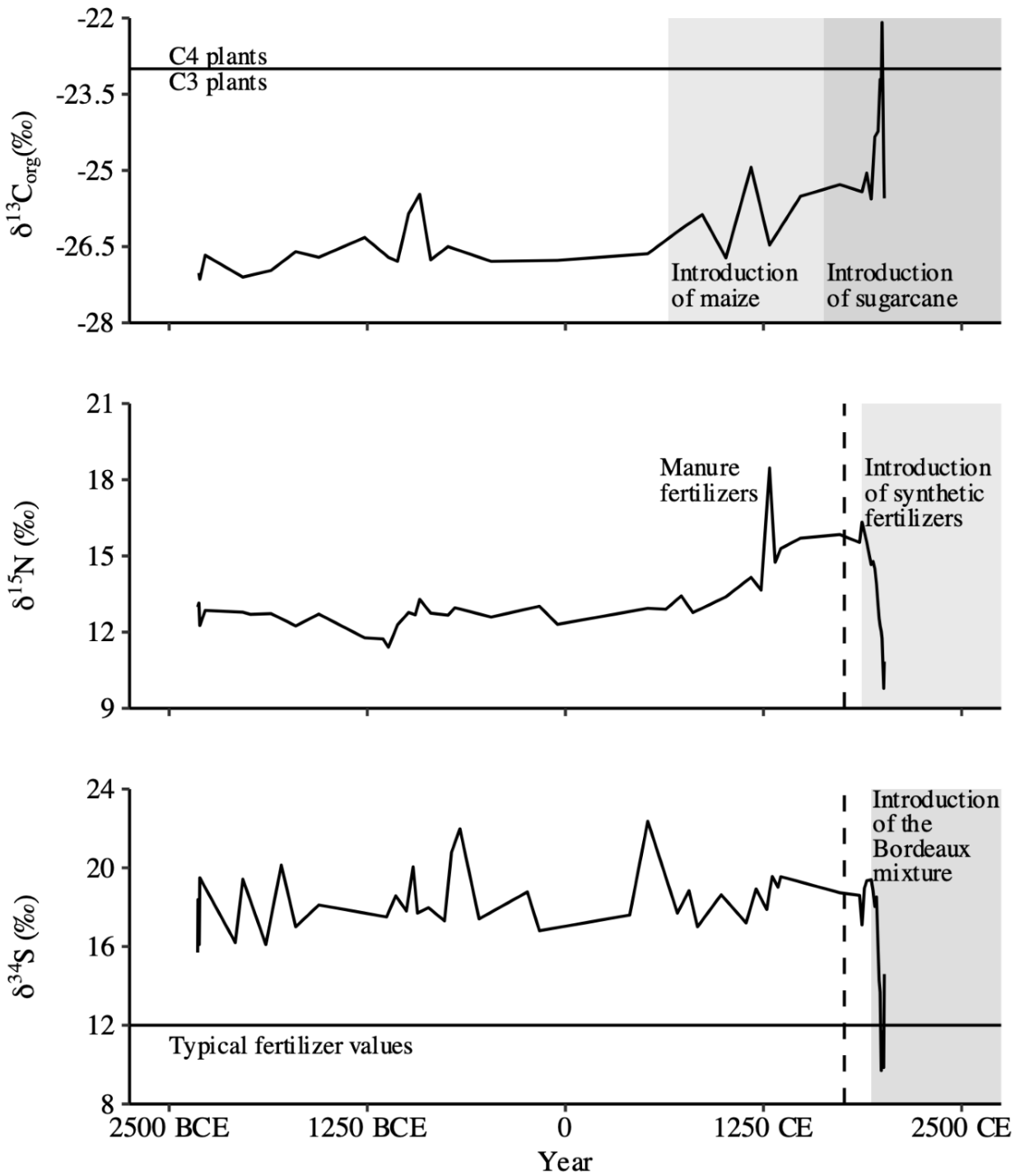


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588 Figure 3: Pb isotope ratios in the HOM core in pre-1920 deposited guano and post-1920
589 deposited guano. Pb ratios in lake sediments from Crystal Lake and Wakefield Lake are also
590 plotted. Lines divide the stable isotope values of naturally occurring lead from leaded gasoline
591 (Emmanuel and Erel 2002).

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593 **1 column fitting**



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Figure 4: Stable isotope profiles in the HOM deposit graphed along the ^{210}Pb and ^{14}C dates. The dashed line indicates the year 1760 [CE](#), the start of the Industrial Revolution. Org = organic.

599 **Supplementary Information for**

600
601 A bat guano deposit in Jamaica recorded agricultural changes and metal exposure over the last >
602 4,300 years

603
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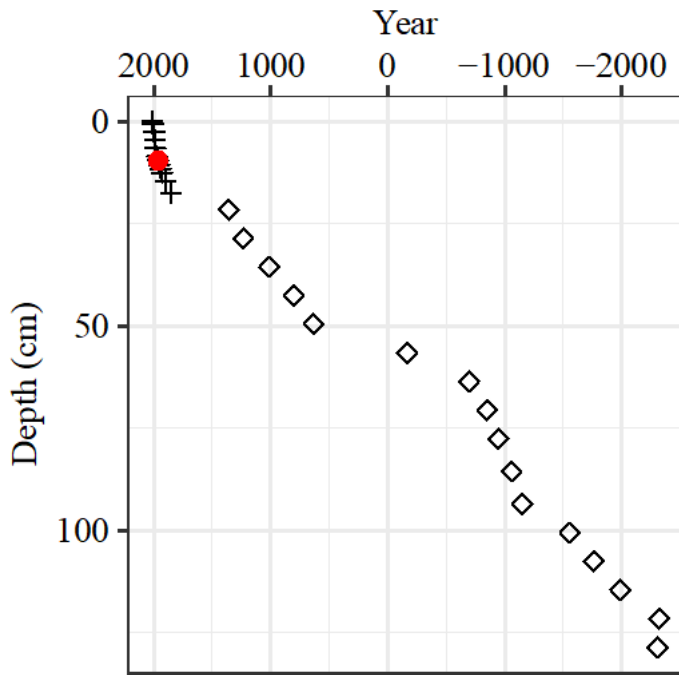
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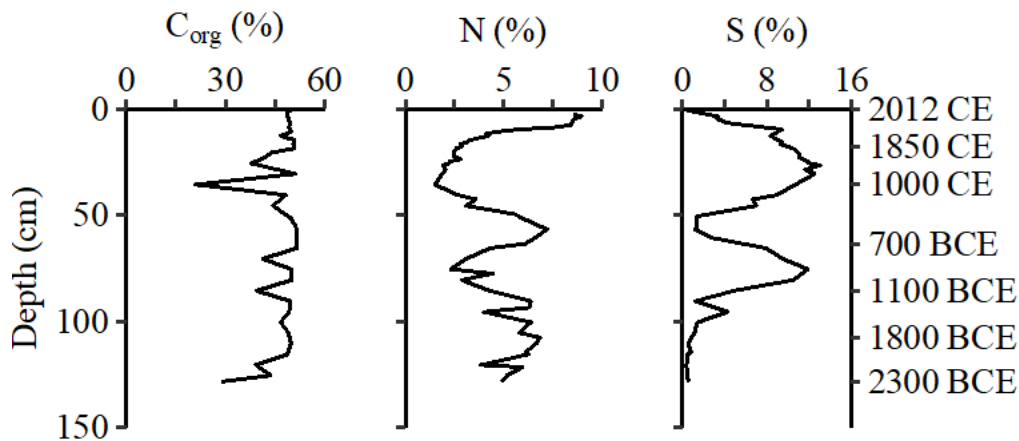
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618 Figure S1 to S3
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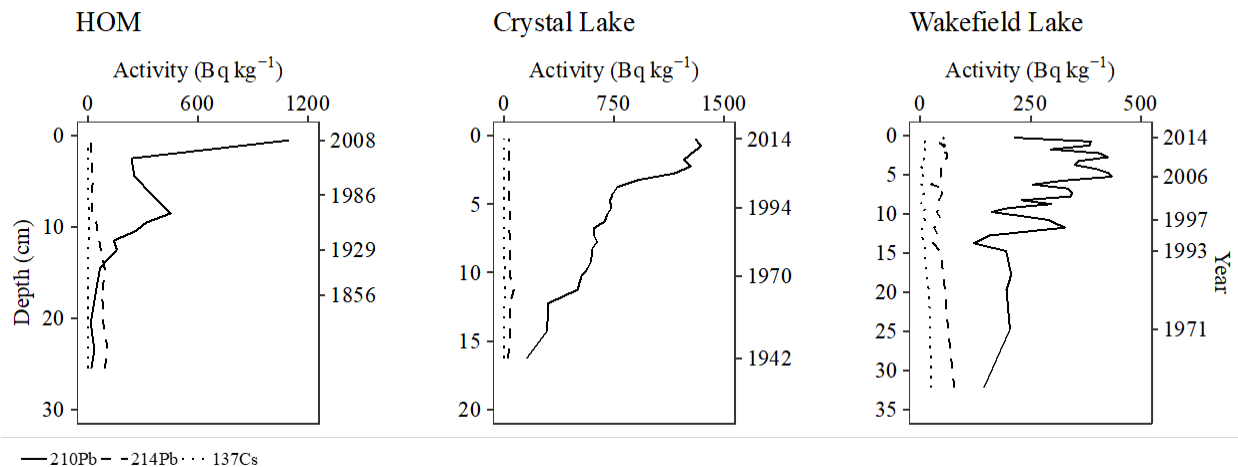
645 **1 column fitting**



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 647 Figure S1: ^{210}Pb (+), peak ^{137}Cs (red circle), and ^{14}C (squares) dates in the bat guano deposits. A
 648 580-year reservoir effect for radiocarbon was applied to the ^{14}C dates, based on the calculated
 649 difference between the ^{14}C date on material from the depth where the ^{210}Pb date was 1900 CE.
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 652 Figure S2: Percentage of organic carbon (C_{org}), nitrogen, and sulfur in the HOM deposit. Select
 653 estimated ^{210}Pb and ^{14}C dates are presented on the right y-axis.
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Figure S3: ^{210}Pb , ^{214}Pb , and ^{137}Cs activity in the HOM core, Crystal Lake, and Wakefield Lake. The constant rate of supply (CRS) model is used to infer year and is shown on the right y-axes. Table S1: ^{14}C dates for lipid-treated and untreated bulk guano samples.

Depth (cm)	Lipid treated	Untreated
14.5	675 ± 30 BP	745 ± 30 BP
56.5	2625 ± 35 BP	2570 ± 35 BP
85.5	3310 ± 35 BP	3415 ± 25 BP

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Table S2: Method detection limits (MDLs) for metals, presented in counts per second (CPS), calculated using 5 blanks.

Analyte	Standard deviation of the blanks (CPS)	Slope of calibration curve	MDL ($\mu\text{g kg}^{-1}$)
Cu	117543	14530	24.27
Zn	1918	2350	2.45
Cd	3.2	3381	0.00
Pb	588	26445	0.07
Ti	21	243	0.26

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Table S3: Metal concentrations, metal ratios, and Pb isotope ratios in the lake sediments. All values are in $\mu\text{g g}^{-1}$ dw. Sample size indicated by 'n'. min = minimum, max = maximum.

Crystal lake				
n = 23	Pb	Zn	Cd	Cu
Median	1.31	5.92	0.04	4.13
Min-max	1.00-2.01	4.93-7.73	0.03-0.09	3.21-5.39
Median (metal/Ti)	0.12	0.55	0.004	0.38
Min-max (metal/Ti)	0.10-0.17	0.49-0.71	0.002-0.008	0.34-0.53

n = 3	²⁰⁶ Pb/ ²⁰⁷ Pb	²⁰⁸ Pb/ ²⁰⁶ Pb	²⁰⁸ Pb/ ²⁰⁷ Pb	²⁰⁶ Pb/ ²⁰⁴ Pb
Median	1.210	2.069	2.483	18.21
Min-max	1.181-1.210	2.051-2.081	2.457-2.504	18.13-18.60

Wakefield lake

n = 22	Pb	Zn	Cd	Cu
Median	2.27	11.60	0.23	2.49
Min-max	1.92-3.18	9.30-15.68	0.17-0.28	1.97-3.58
Median (metal/Ti)	0.42	2.00	0.04	0.41
Min-max (metal/Ti)	0.26-0.78	1.38-3.44	0.03-0.07	0.33-0.75
n = 4	²⁰⁶ Pb/ ²⁰⁷ Pb	²⁰⁸ Pb/ ²⁰⁶ Pb	²⁰⁸ Pb/ ²⁰⁷ Pb	²⁰⁶ Pb/ ²⁰⁴ Pb
Median	1.171	2.098	2.467	18.13
Min-max	1.161-1.184	2.086-2.125	2.445-2.469	17.70-18.46

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Table S4: Metal concentrations, metal ratios, and Pb isotope ratios in the bat guano deposit. All values are in $\mu\text{g g}^{-1}$ dry unless identified with ‘*’, indicating mg g^{-1} . TOC = total organic carbon, min = minimum, max = maximum, CE = common era.

	Pb	Hg	Zn	Cd	Cu
Median ($\mu\text{g g}^{-1}$)	8.04	0.18	997	92.8	3.92*
Min-max ($\mu\text{g g}^{-1}$)	2.17-26.2	0.04-0.85	312-3,060	32.1-247	0.58-15.2*
Median ($\mu\text{g g}^{-1}$ TOC)	17.9	0.41	2050	213	10.7*
Min-max ($\mu\text{g g}^{-1}$ TOC)	4.27-81.7	0.09-1.72	627-6,190	65.6-503	1.18-32.7*
Median (metal/Ti)	0.06	0.002	8.37	1.04	45.8
Min-max (metal/Ti)	0.04-0.70	0-0.02	3.54-198	0.38-4.18	20.6-138
Pre-1850 CE median	0.06	0.002	5.44	0.92	54.3
Post-1850 CE median	0.25	0.004	43	1.95	39.4

	²⁰⁶ Pb/ ²⁰⁷ Pb	²⁰⁸ Pb/ ²⁰⁶ Pb	²⁰⁸ Pb/ ²⁰⁷ Pb	²⁰⁶ Pb/ ²⁰⁴ Pb
Median	1.461	1.678	2.446	20.52
Min-max	1.142-1.512	1.603-2.094	2.367-2.558	16.42-24.77
Pre-1850 CE median	1.469	1.664	2.446	21.45
Post-1850 CE median	1.228	1.994	2.439	18.09

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Table S5: Average and standard error of Wakefield Lake (depth of sample = 21.5 cm, n = 3) and Crystal Lake (depth of sample = 13.5 cm, n = 3).

Analyte	Wakefield Lake		Crystal Lake	
	Average ($\mu\text{g kg}^{-1}$)	Standard error ($\mu\text{g kg}^{-1}$)	Average ($\mu\text{g kg}^{-1}$)	Standard error ($\mu\text{g kg}^{-1}$)
Cu	23.0	0.83	43.2	0.87

Zn	105	2.94	65.1	1.89
Cd	2.31	0.09	0.35	0.02
Pb	22.7	0.62	13.8	0.33
Ti	60.7	1.65	124	5.82
²⁰⁶ Pb/ ²⁰⁷ Pb	1.172	0.001	1.194	0.007
²⁰⁸ Pb/ ²⁰⁶ Pb	2.113	0.005	2.086	0.007
²⁰⁸ Pb/ ²⁰⁷ Pb	2.476	0.005	2.489	0.006
²⁰⁶ Pb/ ²⁰⁴ Pb	18.37	0.097	18.05	0.089

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