1 Mercury pollution in the lake sediments and catchment soils of anthropogenically-disturbed sites
2 across England

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8

9 **Abstract**

10 Sediment cores and soil samples were taken from nine lakes and their catchments across England
11 with varying degrees of direct human disturbance. Mercury (Hg) analysis demonstrated a range of
12 impacts, many from local sources, resulting from differing historical and contemporary site usage
13 and management. Lakes located in industrially important areas showed clear evidence for early Hg
14 pollution with concentrations in sediments reaching 400 - 1600 ng g^{-1} prior to the mid-19th century.
15 Control of inputs resulting from local management practices and a greater than 90% reduction in UK
16 Hg emissions since 1970 were reflected by reduced Hg pollution in some lakes. However, having
17 been a sink for Hg deposition for centuries, polluted catchment soils are now the major Hg source
18 for most lakes and consequently recovery from reduced Hg deposition is being delayed.

19

20 Capsule abstract

21 Urban and suburban lake sediments across England reveal mercury pollution by local industry and
22 urbanisation, while catchment inputs have become the major source of Hg for most lakes.

23

24 **Keywords:** Urban and suburban lakes; Mercury; Catchment inputs; Lake restoration.

25

26 **1. Introduction**

27   Although the emission of mercury from anthropogenic sources has a very long history (Hylander
28 and Meili, 2003), the Industrial Revolution, principally through massive consumption of coal, greatly
29 elevated Hg atmospheric emission. Globally, a rapid increase in Hg release into the environment
30 occurred from the 1850s onwards, not only through coal burning, but also via other anthropogenic
31 processes such as ore refining and, more recently, waste incineration (Hylander and Meili, 2003;
32 Nriagu and Pacyna, 1988). It is estimated that Hg emissions to the atmosphere have increased by
33 around three times over this period (Lindberg et al., 2007).

34 Following release to the atmosphere, Hg returns to the Earth's surfaces by both wet and dry
35 depositional processes and lake sediments and their catchment soils are two important sinks. Soil,
36 which acts as both sink and source of Hg and other trace metals, is an important reservoir (Gillis and

37   Miller, 2000; Yang et al., 2001) and contaminated catchment soils can have a long-term impact on
38   freshwater ecosystems (Yang et al., 2002; Yang, 2010) with transfer via soil erosion being an
39   important process (Rothwell et al., 2005; 2007; Rose et al., 2012). However, erosion itself is
40   controlled by many factors including catchment slope, vegetation coverage, catchment hydrology
41   and climate (Morgan et al., 1998; Dearing, 1991), while land-use and other direct human
42   disturbance in catchments can also enhance soil movement (Bakker et al., 2008). Therefore, these
43   processes potentially have a large influence on the rate and scale of Hg transfer to aquatic systems
44   and can be recorded in sediment archives.

45   Lake sediments have been widely used to reconstruct pollution histories, but most published
46   Hg records are derived from remote lakes, where atmospheric deposition is the sole input for
47   anthropogenic Hg, and these can provide information on depositional trends over relatively large
48   regions (e.g. Bindler et al., 2001; Fitzgerald et al., 2005; Yang et al., 2010). However, Hg sources for
49   lakes that have been directly disturbed by human activities, e.g. lakes in urban or suburban areas,
50   are more complicated (Bookman et al., 2008; Yang, 2010; Van Metre, 2012; Li et al., 2013), but their
51   records may be equally important, especially considering the recent, and predicted, expansion of
52   urban areas across the globe. Cooper et al. (2007) summarise three reasons why the study of
53   human-dominated systems is important. First, because anthropogenic influences are so dominant
54   they need to be included in ecological modelling. Second, including these influences in a realistic way
55   raises the chances of solving existed problems, and third, ecosystem studies within human-
56   dominated environments are relatively sparse and hence knowledge of them is limited. Data from
57   this study addresses the second and third of these points and additionally provides data for future,
58   spatially-resolved Hg modelling.

59   The first increase in Hg contamination above background in many remote lake sediment
60   records is typically observed in the 1850s and 1860s (Johansson, 1985; Engstrom and Swain, 1997;
61   Fitzgerald et al., 2005), and records from relatively remote areas across the UK show a similar trend
62   (Yang and Rose, 2003). However, the UK was one of the pioneering nations in the Industrial
63   Revolution and so industrialisation, and the environmental consequences of it, including urban
64   expansion and population change, began much earlier than the mid-19th century. For example, rapid
65   industrial expansion occurred in Birmingham from the 1770s due to the application of the steam
66   engine driven by coal combustion (Skipp, 1997) while in Manchester coal consumption for domestic
67   use was already considerable in the early-17th century and expanded in the late-18th and early-19th
68   centuries with the rapid development of the cotton industry (Mosley, 2001). Since then, many lakes
69   and their catchments, especially in urban and suburban settings, have been disturbed to varying
70   degrees, mobilising industrial Hg deposition from catchments to lakes (e.g. Yang, 2010) but how
71   reliably these enhanced Hg inputs are recorded in lake sediments is not well known.

72   This study uses lake sediment records and catchment soil analyses from variably disturbed,
73   urban to rural lakes across England to explore how local inputs of Hg have affected these systems
74   and how faithfully the sediment archive records these changes. We also assess the extent to which
75   these lake systems have responded to the dramatic (>90%) reduction in UK Hg emissions since 1970
76   (NAEI, 2014) or whether this has been negated by an increased transfer of contamination from
77   disturbed catchments. The responses of these lakes provide a useful reference for other regions,
78   especially where urban and industrial development continues to expand.

79

80 **2. Sites and methods**

81 2.1. Study sites

82   Open Air Laboratories (OPAL), is a community-driven research programme in England
83 (Davies et al., 2011) designed to promote a greater understanding of the state of the natural
84 environment. As part of this, a national water survey was developed to encourage the public to
85 explore local lakes and ponds. A second strand involving detailed monitoring at a lake in each of nine
86 designated regions of England was also undertaken (Turner et al., 2013). These lakes were selected
87 by regionally based OPAL Community Scientists and represented sites where there was a particular
88 local interest. The monitoring programme provided these groups with useful information and they
89 were able participate in, and support, the research. As a consequence, the selected lakes were a
90 diverse group including urban lakes and ponds as well as those in more rural settings. These lakes
91 have been subject to a variety of local disturbances illustrative of the range of histories, impacts and
92 multiple stressors that have determined their current status. The locations of these lakes are shown
93 in Figure 1 while basic geographical information is provided in Table 1. A brief description of the
94 nature of the sites is provided in Supplementary Information (SI).

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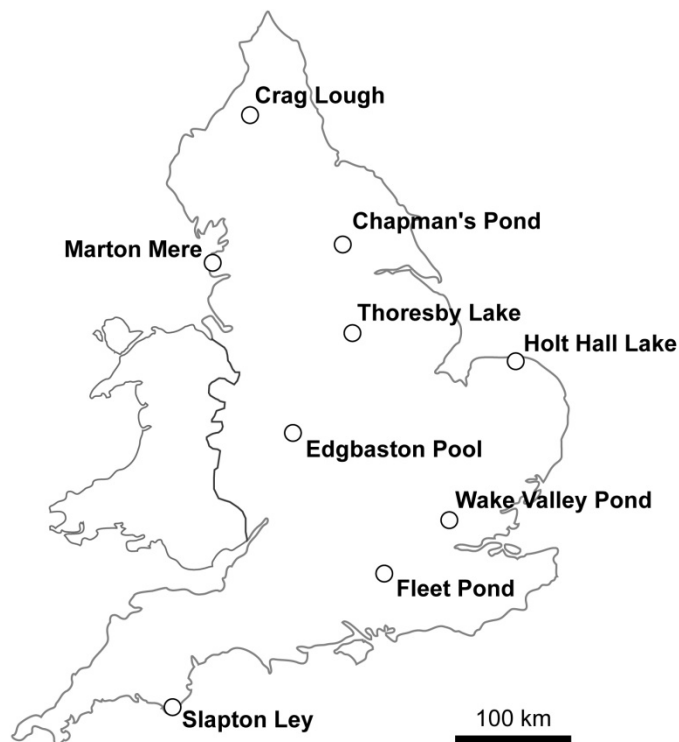
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98 Figure 1. Location of study sites in England.

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100

101   Table 1. Lake and catchment information

Site name [code]	Latitude	Longitude	Altitude (m a. s. l)	Lake Water area (ha)	Max depth (m)	Catchment Area (ha)
Chapman's Pond [CHAP]	53.93423	-1.12023	16	0.6	4.5	< 10
Crag Lough [CRAZ]	55.005853	-2.365318	244	10.1	2.0	182
Edgbaston Pool [EDGB]	52.453901	-1.920802	127	7.2	2.5	644
Holt Hall Lake [HOLTU]	52.915543	1.087271	47	0.7	1.0	65
Marton Mere [MARM]	53.809337	-2.999984	7	10.8	4.5	1015
Fleet Pond [PFLE]	51.287473	-0.825006	68	26.4	1.0	1230
Slapton Ley [SLT]	50.283843	-3.650164	3	65.9	2.5	1774
Thoresby Lake [THOP]	53.226564	-1.058643	37	30.4	3.5	9615
Wake Valley Pond [WAKE]	51.669496	0.053029	96	1.0	4.0	32

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103

104   2.2. Sediment and soil sampling

105   A sediment core was collected from the deepest part of each lake in 2008 using a wide-diameter
106   piston corer. These cores were sliced at 1-cm intervals. Two surface soil (0-5 cm) samples and one
107   soil profile were collected from representative areas of each lake catchment in 2011 (except Crag
108   Lough and Marton Mere, where only the soil profile was collected) using a standard soil auger. Soil
109   samples were collected as 5 cm vertical sections. Standard lithostratigraphic analyses (bulk wet
110   density, water content and loss-on-ignition at 550°C) (Dean, 1974) were undertaken on each
111   sediment sample. All sediment and soil samples were freeze-dried prior to chemical analysis.

112   2.3. Sample analyses and calculation of Hg fluxes in the sediments

113   Sediment cores were radiometrically-dated using ^{210}Pb, ^{137}Cs and [241]Am (see Supplementary
114   Information) and chronologies calculated using the constant rate of supply (CRS) model (Appleby,
115   2001). For Hg analysis, 0.2 g of dried sample was weighed into a 50 mL polypropylene DigiTUBE (SCP
116   Science). 8 mL aqua regia were added to each tube and gradually heated on a hotplate to 100 °C to
117   avoid violent reaction. After reaching 100 °C, the sample was digested for another 1.5 h and then
118   allowed to cool. The digested solution was diluted to 50 mL using distilled deionised water. Standard
119   reference stream sediment (GBW07305; certified Hg value 100 ± 10 ng g^{-1}; our measured mean
120   value 100.3 ng g^{-1}; RSD 4.5 ng g^{-1}; N=15) and sample blanks were digested with every 20 samples.

121 Mercury concentrations in digested solutions were measured by ASP cold vapour-atomic
122 fluorescence spectrometry following reduction with $SnCl_2$. Standard solutions and quality control
123 blanks were measured after every five samples to monitor measurement stability. Geochemical
124 element concentrations were measured using X-ray fluorescence (XRF) spectrometry techniques
125 described in Supplementary Information. Mercury fluxes in the sediments were derived from Hg
126 concentrations and sedimentation rates calculated from the radiometric chronologies.

127 2.4. Anthropogenic Hg calculation

128   In a sediment sample, the natural contribution of an element is given by LR_s/LR_b, where LR_s
129 and LR_b are the concentrations of a selected lithogenic reference element in the sample and the
130 background (no anthropogenic impact), respectively. Therefore, the natural contribution to Hg
131 concentration in a sample (Hg_{bc}) is:
132
133 $Hg_{bc} = (LR_s/LR_b) \times Hg_b$ (1)
134
135 where Hg_b is the Hg concentration in background sediments. The anthropogenic concentration (Hg_{ac})
136 is therefore:
137
138 $Hg_{ac} = Hg_t - (LR_s/LR_b) \times Hg_b$ (2)
139
140 where Hg_t is the total Hg concentration in the sample. Anthropogenic Hg flux to the sediments (Hg_{af})
141 is:
142
143 $Hg_{af} = Hg_{ac} \times SR$ (3)
144
145 where SR is sedimentation rate. This method is simpler than calculations of metal enrichment factors
146 but provides the same results (cf: Perry et al., 2006; Boes et al., 2011; Hermanns and Biester 2013).
147 In most sediment cores, temporal changes in Ti provide a better representation of lithogenic inputs
148 than other elements (cf. Yang and Smyntek, 2014), and so was chosen for the natural contribution
149 calculations.
150
151   Some cores such as THOP and WAKE show high Hg contamination even in basal samples and so
152 cannot be used for natural Hg contribution calculations. Therefore, as Hg and Ti concentrations in
153 uncontaminated deep soils are close to those in uncontaminated sediments, especially in urban sites
154 with high sedimentation rates, these were used to determine the natural Hg contribution, although
155 it is possible that Hg may be transported deeper into soils by complexation with dissolved organic
156 carbon (DOC) (Schuster et al., 2008).
157

158 **3.   Results**

159 3.1. Sediment Chronologies and sedimentation rates

160   The unsupported ^{210}Pb profiles of all nine sediment cores show non-monotonic features, i.e.
161 unsupported ^{210}Pb activities do not show an exponential decline with depth in individual cores. This
162 implies changes in sedimentation rates at all sites. Equilibrium depths of total ^{210}Pb with the

163 supported ^{210}Pb activity (corresponding to the last c. 150 years sedimentation period) in the
164 sediment cores vary (Table 2). In general, they are considerably deeper than those in sediments
165 taken from remote regions (cf., Appleby, 2000; Appleby, 2004; Yang et al., 2010) indicating higher
166 sediment accumulation rates. Most of the cores have well-resolved 1963 ^{137}Cs peaks derived from
167 the maximum fallout from nuclear weapons testing. This indicates that the sediments in these sites
168 have been relatively unmixed since at least the 1960s, and that these cores might provide historically
169 reliable records of pollutant inputs. However, the 1963 ^{137}Cs peaks in the cores from Holt Hall
170 (HOLTU), Slapton Ley (SLT) and Thoresby Lake (THOP) are relatively poor, although still distinct,
171 suggesting that these sediments may have been subjected to limited mixing.

172

173 Table 2. Radiometric dating features for the sediment cores.

Site name [code]	Equilibrium depth (cm) of total and supported ^{210}Pb	^{210}Pb flux (Bq m^{-2} yr^{-1}) to the core location	Sedimentation rate range (g cm^{-2} yr^{-1})	^{137}Cs 1963 peak Depth (cm)
Chapman's Pond [CHAP]	38±3	115±10	0.04-0.79	30±2
Crag Lough [CRAZ]	105±5	117±7	0.03-1.86	13.5±1
Edgbaston Pool [EDGB]	53±3	240±12	0.02-0.17	27.5±1.5
Holt Hall Lake [HOLTU]	64±4	176±11	0.04-1.18	42±2
Marton Mere [MARM]	63±4	193±20	0.02-0.40	45.5±2
Fleet Pond [PFLE]	45±3	177±13	0.03-0.16	21±1.5
Slapton Ley [SLT]	24±2	82±5	0.006-0.05	15.5±1
Thoresby Lake [THOP]	75±4	211±51	0.10-0.50	28±2
Wake Valley Pond [WAKE]	57±3	224±16	0.03-0.24	30.5±2

174

175

176 Sedimentation rates cover a large range (Figure S1) from 0.006 g cm^{-2} yr^{-1} before 1900 in
177 Slapton Ley (SLT) to 1.86 g cm^{-2} yr^{-1} in the 1950s at Crag Lough (CRAZ). As the organic content and
178 geochemical element concentrations were constant at Crag Lough through this brief period of
179 extraordinarily high sediment accumulation, it appears that the sediment source did not change.
180 This 'event' is probably due to a rock-fall from the cliffs above the lake causing a redistribution of
181 contemporary sediments to the coring site (Turner et al., 2013).

182 In general, changes in sedimentation rates within individual cores are not smooth, and probably
183 reflect human disturbance in the catchments (Figure S1). For example, the sedimentation rate in the
184 core from Edgbaston Pool (EDGB) reached 0.17 g cm^{-2} yr^{-1} before the 1900s, implying enhanced
185 catchment input, followed by declining rates through the 20th century. Similarly, there are sharp

186 peaks in sedimentation rate in Chapman's Pond (CHAP) in the early-1980s and at Marton Mere
187 (MARM) in the 1950s probably derived from waste disposal at these sites.

188 3.2. Hg in soils and sediments

189   Mercury concentrations in catchment soils vary from site to site, but also show considerable
190 within-site variation (Figure S2) with differences in Hg concentrations in the surface soils varying by a
191 factor of 2 to 10. Mercury concentrations in the top 0-15 cm are almost all higher than the median
192 concentration (95 ng g-1) for UK rural topsoils (0-15 cm) sampled between 1998 and 2008 (Tipping
193 et al., 2011), indicating a higher level of contamination. The exception is Marton Mere, where the
194 surface soils have lower Hg concentrations, but are higher at depth, probably indicating burial of
195 contaminated soils during an earlier period of catchment disturbance.

196   Sediment records show clear increases in Hg concentrations at, or before, the 1850s. In
197 Edgbaston Pool and Marton Mere, Hg concentrations exceeded the consensus Probable effects
198 Concentration (PEC) for freshwater sediments (1060 ng g^{-1}) (MacDonald et al., 2000) before the
199 1850s, the Hg concentration at which adverse biological effects on benthic aquatic organisms are
200 commonly seen (e.g. US National Oceanic and Atmospheric Administration, 1999; CCME, 1999;
201 MacDonald et al., 2000). The industrialisation of the city of Birmingham, in which Edgbaston Pool lies,
202 started much earlier than some other regions of England (Allen, 2001) as it was active in the
203 Industrial Revolution before the mid-19th century (Stobart and Raven, 2005). Economic expansion
204 started in the 16th century (Allen, 2011) and by 1650 Birmingham had a population of around 5000,
205 with iron-working named as the employment for one in every six households (Skipp, 1997). Industry
206 expanded rapidly from the 1770s due to application of the steam engine driven by coal combustion,
207 a source of Hg emissions. The city's population also expanded through this period from
208 approximately 23,000 in 1731 to 170,000 in 1831 (Skipp, 1997), also due to this economic expansion
209 and development of industrialisation (Allen, 2001).

210   Marton Mere is also located in an area of early factory-based industrialisation and urbanisation
211 and was greatly affected by historical coal burning and by waste from local metal production and
212 manufacturing entering the drainage system to which it was connected (Stobart, 2004). Therefore,
213 the start of the Hg increase in the lake may reflect regional industrial development of factory-based
214 industry, trade and transport systems produced by a locally integrated economy between 1700 and
215 1760 (Stobart, 2004). Hg concentrations in the sediments of Thoresby Lake show little temporal
216 pattern but remain high (approximately 500 ng g^{-1}) throughout, even before the 1850s. This
217 concentration is around 20 times higher than the Hg concentration in the deep soils taken from the
218 catchment. Coal production has a long history around the site with seams from the Thoresby Colliery
219 extending below the site (see Supplementary Information). The catchment area of Thoresby Lake is
220 316 times its lake area, high Hg concentrations in the pre-1850 sediments and significantly elevated
221 catchment background concentrations suggest contamination possibly derived from early industrial
222 activity in the area.

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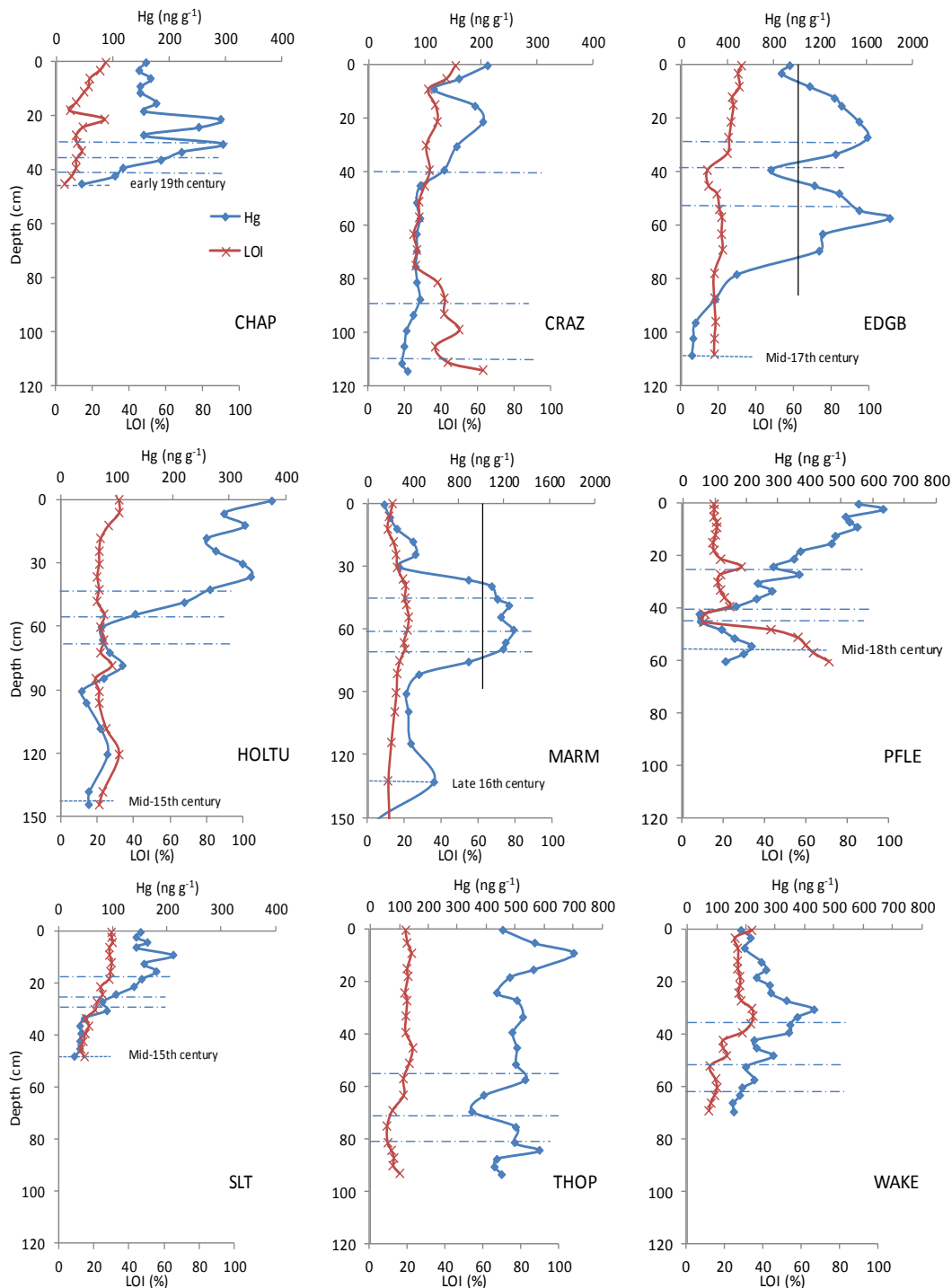
Figure 2. Mercury concentration profiles and loss-on-ignition (LOI) profiles for the lake sediment
cores plotted on depths. Note differences in depth and concentration scales. Horizontal dashed
lines (—·—) correspond to 1850, 1900, and 1950. Dates prior to the ^{210}Pb dated sections were
extrapolated and are for guidance only (see text). Vertical lines indicate consensus Probable Effect
concentration (PEC).

The differing historical influences to which the lakes have been subjected result in a variety
of temporal patterns in both the Hg concentration and flux profiles (Figures 2 and 3) since the 1850s.
Chapman's Pond was impacted by waste disposal from the early-1970s to the early-1980s resulting

8

in high sedimentation rates and a period of very high Hg fluxes (exceeding 1200 μg m^{-2} yr^{-1}) (Figure 3). Sediment organic content and geochemical element (e.g. Zr, Ti, Ca, K) concentrations also show sharp concentration changes in the 1980s (see Turner et al., 2013) indicating a change in sediment sources, and could be due to material brought from outside of the catchment for disposal. Since waste disposal stopped, Hg concentrations in the sediments have reduced to a constant value. In Marton Mere, wetlands adjoining the lake were gradually drained through to 1850 (Clarke, 1969), and also subsequently used for disposal of urban waste. This stopped in 1972 after a reform of lake management in the 1960s, resulting in a sharp decline in sediment Hg concentrations from approximately 1000 ng g^{-1} to 400 ng g^{-1}. This implies that the material disposed at the site may have contained high levels of Hg. Nearly all the sediment Hg in Marton Mere is from anthropogenic sources (Figure 3) and this has remained the dominant source over the last ca. 140 years. Anthropogenic Hg follows trends to sedimentation rates, although relative scales have varied suggesting changes to Hg concentrations in inputs.

In Upper Holt Hall Lake, Hg concentrations increased rapidly from the 1900s (Figure 2) and may at least partially have been derived from inputs of domestic effluent and waste water from the surrounding area. These inputs are also likely to have increased the sedimentation rates as well as total and anthropogenic Hg fluxes, but this is not the case in all sites. The high sedimentation rate in the 1950s in Crag Lough possibly caused by a rock fall resulted in a brief period of very high Hg fluxes (800 μg m^{-2} yr^{-1}) (Figure 3) but does not indicate elevated contamination. Here, anthropogenic Hg fluxes were relatively constant before the 1940s while total Hg fluxes increased, following the same trend as sedimentation rate, indicating an increase in catchment inputs of less contaminated materials into the lake.

In recent decades, while some sites have shown constant Hg concentrations or a decline, others have shown an increase. Decreases may be due to improved catchment management or reduced atmospheric deposition. However, some decreases in Hg concentration may also be due to dilution by an increased sedimentation rate (e.g. WAKE core, see Figure S3), and in these cases concomitant Hg fluxes continue to follow the increasing trend in sediment accumulation (Figure 3). In Crag Lough, increasing trends in total and anthropogenic Hg concentrations and sediment fluxes over the last decade suggest an increase in atmospherically-contaminated soil input, possibly via an increase in catchment soil erosion or agricultural activity.

At Fleet Pond, there has been a rapid increase in Hg sediment concentrations since the 1970s (Figure 2), which is in direct contrast to the dramatic reduction in UK Hg emissions (emissions reduced from 63 tonnes yr^{-1} in 1970 to their current level of c. 6.3 tonnes yr^{-1}; NAEI, 2012). This increase in Hg concentration at Fleet Pond is therefore likely to be due to local contamination. Similar rates of change in sedimentation and Hg fluxes before 1970 (Figure 3), indicate catchment inputs. As Hg concentrations in the catchment soils are considerably lower than those in surface sediments, the high sediment Hg concentrations are likely to be derived from local, recently emitted Hg rather than Hg released from catchment soil storage. The delayed increase in sedimentation rates compared with the increase in Hg fluxes after 1970, also suggests a recent local Hg source.

In most sites, anthropogenic Hg has been the dominant source especially over the last hundred years, and changes in total and anthropogenic fluxes to the sediments are strongly linked with sedimentation rates (Figure 3).

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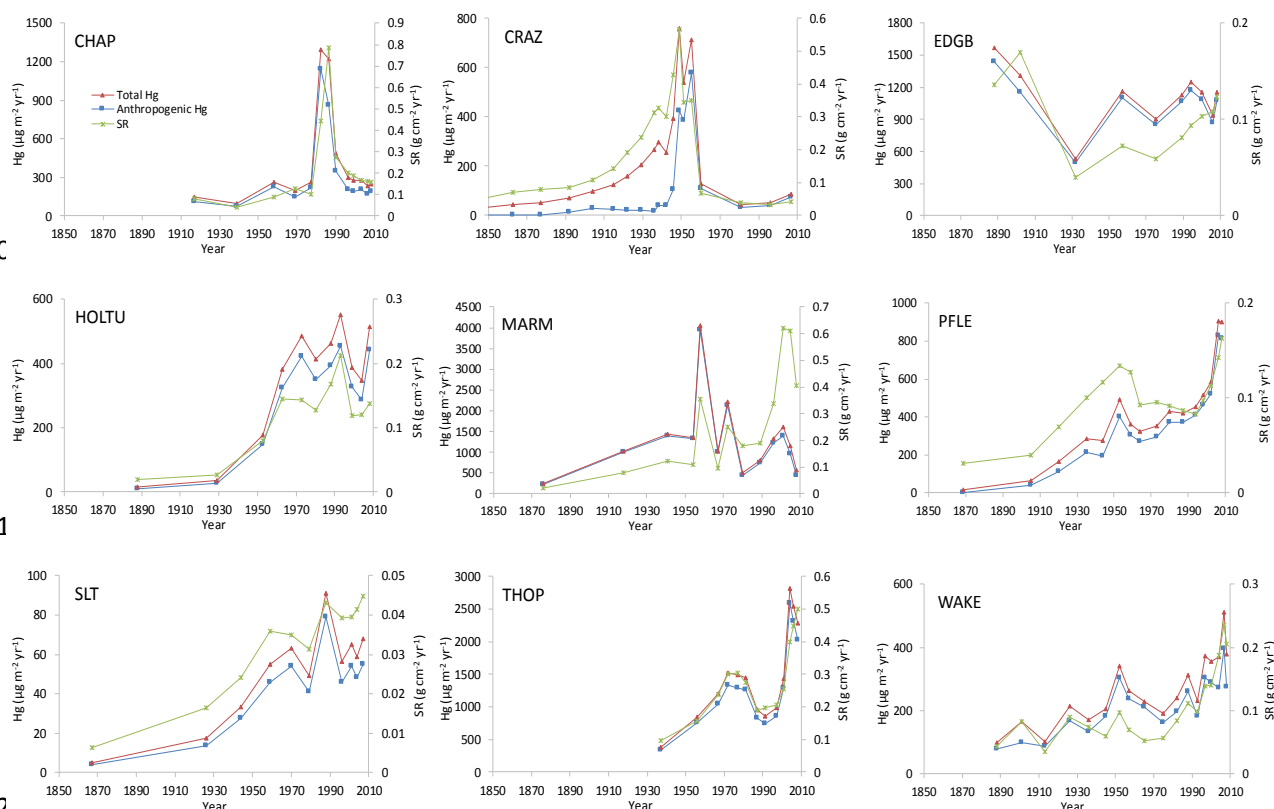
Figure 3. The total and anthropogenic Hg flux profiles and sedimentation rate changes for the sediment cores. Note the differences in the flux and rate scales.

4. **Discussion**

4.1. Historical Hg trends

Organic content of the sediments is relatively high with loss-on-ignition reaching 30% or more in the surface sediments of half of the cores (Figure 2). Variation in Hg concentrations might be related with the organic content of the pre-1850 sediments in Slapton Ley, pre-1900 sediments in Holt Hall Lake and pre-1950 sediments in Wake Valley Pond, as they follow similar trends. However, in most cases, changes in Hg concentrations in the sediments have only a weak relationship with sediment organic content (Figure 2). High organic content in lake sediments may cause considerable redox changes (Davison, 1993). Rapid changes of Mn concentrations in the surface sediments of Crag Lough, Edgbaston Pond and Slapton Ley (Figure S4) suggest a redox effect, as Mn is sensitive to redox change. However, the Hg concentration profiles of Edgbaston Pond and Slapton Ley do not follow the Mn trends, and Hg changes in the surface sediments of Crag Lough are more likely due to catchment inputs and suggesting only limit impact of redox on Hg profiles. As the ^{137}Cs records suggest that sediment mixing is also limited, then Hg profiles of the cores are likely to reliably represent historical Hg trends.

301   In lake sediment cores taken from many remote regions around the world, the first
302   significant increase in Hg concentration is typically observed from the mid-19th century (Johansson,
303   1985; Verta et al., 1989; Engstrom and Swain, 1997; Lamborg et al., 2002; Fitzgerald et al., 2005;
304   Muir et al., 2009) and an increase in Hg concentration observed prior to this time can usually be
305   linked to small and local sources (e.g. Balogh et al., 1999; Bindler et al., 2001; Yang et al., 2010). Mid-
306   19th century increases are usually attributed to the escalation in Hg emissions resulting from coal
307   combustion during the Industrial Revolution (Hylander and Meili, 2003), and sediment and peat
308   cores taken from relatively remote areas across the UK reflect this (Yang and Rose, 2003, Farmer et
309   al., 2009). By contrast, the temporal records of Hg observed in this study are very different both
310   from each other and from the larger-scale pattern of historical deposition. Instead, they record the
311   local pollution history of disturbances and it is only with a more detailed knowledge of the lake
312   histories, their catchments and the immediate area that the Hg records can be unravelled.

313   The Industrial Revolution started from the mid-19th century at a global scale, but was not
314   the abrupt discontinuity that its name suggests, more the result of an economic expansion that
315   started in the 16th century (Allen, 2011). In Britain, especially in regions such as the Midlands and
316   north-west England, industrial development began much earlier and significant Hg concentration
317   increases prior to the mid-19th century record evidence for early environmental impact resulting
318   from it. In this study there are considerable increases in Hg concentrations in five of the nine lake
319   sediment cores before the 1850s (Chapman's Pond; Edgbaston Pool; Marton Mere; Slapton Ley;
320   Wake Valley Pond). While comparison of basal sediments with bottom soil samples in two others,
321   Fleet Pond and Thoresby Lake, indicate that Hg concentrations had already increased to around 200
322   and 500 ng g^{-1} respectively, by this time. The most contaminated sites during the mid-19th century
323   were Edgbaston Pool and Thoresby Lake in the Midlands and Marton Mere in north-west England,
324   with Hg concentrations reaching 1200, 585 and 1800 ng g^{-1}, respectively. At Edgbaston and Marton
325   Mere these mid-19th century concentrations exceed the sediment consensus-PEC for Hg,
326   highlighting the severity of contamination that had already occurred by this time. Away from the
327   Midlands and north-west, increases in pre-1850 Hg concentrations are substantially lower, in
328   agreement with the intensity of industrial activities at the time, and showing similar spatial trend of
329   industrial impacts on the lakes in the US (Van Metre, 2012).

330   After the mid-19th century, Hg concentrations continue to follow increasing trends at
331   Chapman's Pond, Slapton Ley and Wake Valley Pond, while at Holt Hall Pond and Fleet Pond, a new
332   phase of rapidly increasing contamination commenced. It is quite likely that local sources of Hg
333   contamination to all these sites resulted from a combination of historical factors including ignorance
334   of the impacts of waste inputs, lack of knowledge of the contamination burden within the waste,
335   and lack of a realistic and pragmatic alternative to waste disposal (e.g. Yang, 2010). Hence,
336   Chapman's Pond and Marton Mere were used to dump urban waste and Upper Holt Hall Lake
337   received local effluent and waste water. However, contamination was not only directly received by
338   release in the catchment but also via atmospheric deposition which was stored in both lake
339   sediments and catchment soils. Our data indicate substantial catchment soil contamination and that
340   these soils have been a source of Hg to the lakes to some degree.

341   4.2. Catchment Hg inputs to the lakes and sedimentation impacts

342   Increased awareness of the effects of metal contaminants in aquatic systems has resulted in
343   improved lake management and a reduction of direct release to surface waters at local scales
344   (Burniston et al., 2011). In this study, we observe this as a decline in Hg concentrations in Edgbaston
345   Pool, Marton Mere and Wake Valley Pond since the 1960s. At other sites however, the scale of Hg
346   contamination has not declined significantly despite a 90% reduction in UK Hg emissions from
347   anthropogenic sources since the 1970s (NAEI, 2012).

348   Previous studies at upland UK lakes (e.g. Yang et al., 2002; Rose et al., 2012) have
349   demonstrated that Hg stored in catchment soils could be a major source of contamination,
350   obscuring the signal of reduction in atmospheric Hg deposition in the sediment record. Despite its
351   potentially long atmospheric life-time (Lindberg et al., 2007) Hg deposition tends to be higher, closer
352   to emission sources (Chen et al., 2010; Chen et al., 2012; Van Metre, 2012; Drevnick et al, 2012) and
353   so we might expect catchment soils near urban lakes to exceed those in more remote locations. We
354   see that this is the case as urban catchment surface soil Hg concentrations exceed the median
355   concentration for UK rural topsoils (95 ng g^{-1}) (Tipping et al., 2011), except at Marton Mere. As
356   catchment soil erosion is an important source of lake sediment material (Dearing, 1991), the transfer
357   of Hg from catchment soils to lakes cannot be ignored, and will delay the response of the lake
358   system to the reduction in emissions and deposition of contaminants (Rose et al., 2012), including
359   Hg (Yang, 2015).

360   Change in sedimentation rate may be due to changes in productivity especially in the urban
361   lakes (Turner et al, 2013). However, this should not increase the amount of Hg in the lake. In all the
362   cores, total Hg fluxes follow the same trends as sedimentation rates (Figure 3). Their considerable
363   changes in relative level indicate that catchment inputs are the most important source for these
364   lakes. When anthropogenic Hg fluxes follow the same trends as total Hg fluxes, it suggests that
365   anthropogenic Hg is mainly from catchment inputs. This is the case in all cores except CRAZ, where
366   sedimentation rates and total Hg fluxes increase rapidly from the 1850s to the 1940s, while
367   anthropogenic Hg fluxes change only slowly, indicating an increase in less contaminated material
368   entering the lake.

369   The temporal distribution of some geochemical elements in the sediment cores suggests
370   that sedimentation processes have gone through dramatic changes in the past (Figure S3). For Ca,
371   concentrations have changed considerably in most cores and may be due to hydrological change
372   (Dean, 1999) (e.g. Slapton Ley), or changes within the catchment (e.g. Crag Lough) and these
373   processes could also affect Hg input to the sediments. However, direct human impact in urban or
374   suburban sites has played a more important role in Hg input to the lakes. For example, waste inputs
375   to Chapman's Pond changed sediment Ca and Zr concentrations, and increased Hg fluxes to the
376   sediments. Mercury concentrations reach or exceed consensus-PECs in two cores (Edgbaston Pool
377   and Marton Mere), and 500 ng g^{-1} at a further two (Thoresby Lake and Fleet Pond), all of which have
378   clear evidence for local human direct impacts. This shows similar results to increases in Hg loading
379   from local source discharge in other urban, suburban or industrial areas (e.g. Yang, 2010; Van Metre,
380   2012; Li et al., 2013; Drevnick et al., 2016). Furthermore, human activities that disturb catchment
381   soils such as agricultural and forestry processes raise the likelihood of increased Hg transfer from
382   contaminated soils, so catchment management practices are therefore also an important
383   consideration in controlling erosion and hence Hg burden in lakes (Lyons et al., 2006; Porvari et al.,
384   2009).

385   Mercury accumulation rates (fluxes) are derived from the product of Hg sediment
386 concentration and bulk sediment accumulation rate and hence an increase in one, or both, of these
387 parameters can lead to an observed Hg flux increase. If both Hg concentration and sedimentation
388 rate are high, then the combination can result in very high Hg fluxes. For example, the highest Hg
389 flux measurement in this study was in the 1950s at Marton Mere where a sedimentation rate of over
390 0.35 g cm^{-2} yr^{-1} occurred with a Hg concentration of 1142 ng g^{-1} to produce a Hg flux of over 4000 µg
391 m^{-2} yr^{-1}. Mercury fluxes reach or exceed 1000 µg m^{-2} yr^{-1} in four cores. Three (Edgbaston Pool,
392 Thoresby Lake and Marton Mere) are in the Midlands and north-west England, and here, the high Hg
393 fluxes are mainly derived from the high concentrations in the sediments. In Edgbaston Pool, Hg
394 concentrations are over 1000 ng g^{-1} in most sediments deposited since the beginning of the 19th
395 century, while sediments in Marton Mere have Hg concentrations over 1000 ng g^{-1} from the mid-
396 19th century to the 1960s. While the high fluxes at Thoresby Lake are also due to high Hg
397 concentrations, changes in flux through the core are driven by changing sediment accumulation rate
398 as concentrations remain relatively constant (Figure S3). The high Hg flux in the fourth site,
399 Chapmans Pond in York, was due to high sedimentation rates in the early-1980s (see Figure S3)
400 probably due to the impact of waste disposal at the site. In the last decade, Hg fluxes in five
401 sediment cores (Edgbaston Pool; Thoresby Lake; Marton Mere, Fleet Pond; Holt Hall Lake) have
402 remained above 500 µg m^{-2} yr^{-1}, while those in Chapman's Pond and Wake Valley Pond exceed 250
403 µg m^{-2} yr^{-1}, and those of Crag Lough and Slapton Ley are lower, at 87 and 68 µg m^{-2} yr^{-1}, respectively
404 (Figure 3). These are all considerably higher than the limited Hg depositional flux data available for
405 other sites in the UK for the same period (Table 3) (Yang et al., 2002; 2009).

406

407 Table 3. Hg sediment and soil concentrations and fluxes compared with Lochnagar (Yang et al, 2001)
408 and London sites (Yang et al, 2009). Italic numbers in parentheses are measured atmospheric Hg
409 deposition fluxes

Site	Soil surface Hg concentrations (ng g^{-1})	Sediment surface Hg concentrations (ng g^{-1})	Sediment fluxes (*atmospheric deposition fluxes*) (µg m^{-2} yr^{-1})
This study			
Chapman's Pond	237, 416, 806	159	249
Crag Lough	111	212	87
Edgbaston Pool	242, 386, 569	939	1155
Upper Holt Hall Lake	81, 100, 255	376	515
Marton Mere	65	141	570
Fleet Pond	181, 219, 306	556	903
Slapton Ley	101, 224, 459	152	68
Thoresby Lake	93, 257, 270	458	2287
Wake Valley Pond	161, 1438, 1551	185	379
Other UK data			
Lochnagar, Scotland	132-277 (n=10)	111- 228 (n=17)	17.6 (*35.9*)
Central London		100-1718 (mean 491, n=30)	(*15 – 52.5*)

410

411   High Hg fluxes can also be derived from sediment focussing (where sediments from littoral
412   areas are resuspended and transported to deeper areas where cores are usually taken), the scale of
413   which can be assessed by comparing unsupported ^{210}Pb fluxes in the sediment cores with those from
414   modelled atmospheric ^{210}Pb deposition (Appleby, 2001) . For the lakes in this study, sediment [210]Pb
415   fluxes are only 1-3 times higher than depositional fluxes suggesting that the contribution of
416   focussing to high sediment Hg fluxes is limited. Sediment focussing corrected Hg accumulation rates
417   for the 2000s have a relatively good linear relationship with the ratio of catchment area to lake area
418   in these sites (Figure 4). The intercept of regression line (107.64 $\mu g\ m^{-2}\ yr^{-1}$, see Figure 4, the 2000s)
419   which should provide an indication of atmospheric depositional Hg flux in the sampling region may
420   not be correct as some lakes (e.g. Fleet Pond, see SI) have received local, direct or point source
421   inputs during this time, which are likely to increase the intercept level. The considerably higher
422   predicted depositional flux than that for monitored Hg fluxes in Central London (Table 3) might also
423   show evidence for direct, local anthropogenic Hg inputs. The proportion of atmospheric Hg that has
424   been transported from catchments to the lakes in the 2000s has changed considerably when
425   compared with the 1850s, 1900s, and 1950s (Figure 4), suggesting that these sites cannot be used to
426   reconstruct the history of atmospheric Hg deposition (Swain et al 1992, Yang 2015). The poor linear
427   relationship of Hg accumulation rates with the catchment to lake ratios in the 1850s, 1900s, and
428   1950s (Figure 4) imply complexity of Hg sources, and that atmospheric deposition is not the major
429   source of Hg during the time.

430
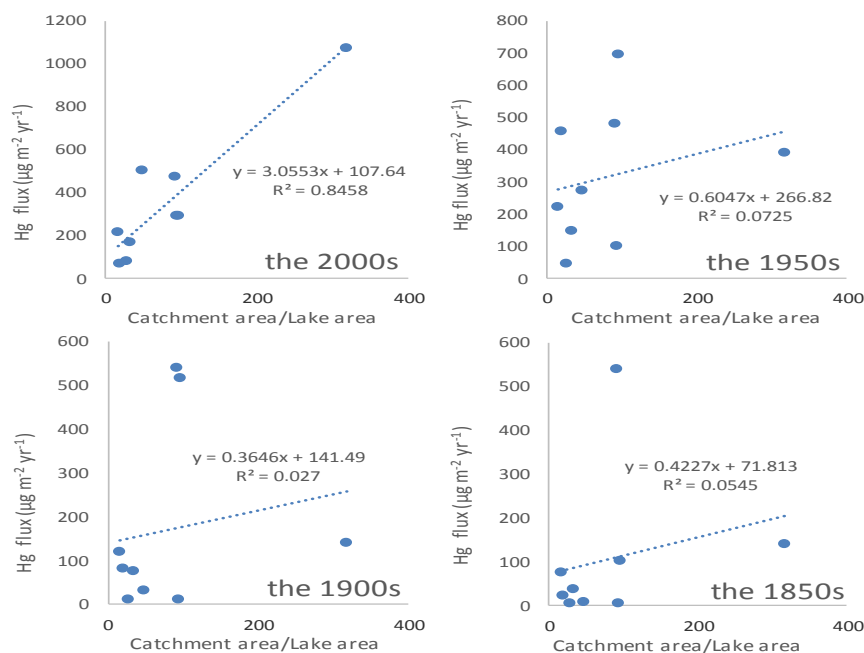
431

432

433   Figure 4. Sediment focussing corrected Hg accumulation rates as linear functions of the ratio of
434   terrestrial catchment area to lake area in the 1850s, 1900s, 1950s and the 2000s.

435

436   Parallel changes in sedimentation rates and Hg fluxes (Figure 3) and high soil and sediment
437   Hg concentrations and sediment Hg fluxes (Table 3) indicate the importance of catchment sources
438   for Hg inputs to a range of aquatic environments. However, it is likely that this will only become

439 further exacerbated in future decades as Hg emissions and deposition remain low and
440 remobilisation of Hg stored within catchment soils remains elevated or increases as a result of
441 climate-enhanced soil erosion (e.g. Rose et al., 2012). This has two consequences. First, inputs of Hg
442 and other contaminants previously stored in catchment soils are transferred to aquatic systems at
443 an increased rate and, second, sediment accumulation rates increase. In general, sedimentation
444 rates at all these sites have increased within the ^{210}Pb dated period (excepting major changes from
445 high magnitude, low frequency events such as at Crag Lough) and are in agreement with the pattern
446 observed for many lake types across Europe (Rose et al., 2011). As described above, the combination
447 of increased Hg inputs and elevated sedimentation rates results in significantly increased Hg fluxes
448 and therefore these could remain high for some considerable time to come.

449

450 5. **Conclusions**

451 The use of lake sediment archives from urban and sub-urban lakes has been very limited,
452 until recently, although there is now an increasing ecological and societal interest in them. While
453 they are less likely to reveal the internationally important patterns of contaminant pathways that
454 are commonly identified from remote lakes, they can faithfully record local disturbances and provide
455 an historical archive of local environmental change, if the sediments have not been too disturbed.
456 They may, therefore, be an under-used resource that, with local knowledge, can provide useful
457 temporal data on contaminant inputs.

458 The sediment records from most of the lakes in this study, particular those in urban areas,
459 show considerable Hg contamination earlier than the mid-19th century probably derived from local
460 industrial and urban development. The data show that at a number of these lakes, Hg
461 concentrations exceeded sediment quality guidelines for the protection of benthic aquatic
462 organisms, before the mid-19th century, and remained high for a considerable period.

463 While UK Hg emissions have declined by over 90% since the 1970s, this has not brought a
464 significant recovery in Hg concentrations and inputs to most of these lakes and mercury fluxes
465 continue to increase in more than half during the period. While this is partially due to local direct
466 inputs and urban development, our data show that this is also due to the reservoir of Hg stored in
467 catchment soils as a result of contamination by centuries of atmospheric Hg deposition and direct
468 release. Hence, Hg inputs to the sediments are mainly derived from catchment inputs reflected by
469 the changes in sedimentation rates. Heavily contaminated soils usually exist at the soil surface but
470 may be buried by catchment disturbance. The remobilisation of this contaminated soil, and the
471 transfer of Hg to aquatic systems, increases both Hg inputs and sediment accumulation rates. Given
472 the massive store of Hg within many catchments and the impact of climate change likely to enhance
473 catchment soil erosion, Hg transfer and inputs to aquatic systems from these sources are likely to
474 remain elevated for many decades to come.

475

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483

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