

1 Title:

2 **Long-term accumulation and transport of anthropogenic phosphorus in three river basins**

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24 Abstract:

25           Global food production crucially depends on phosphorus (P). In agricultural and urban  
26 landscapes much P is anthropogenic, entering through trade. Here we present a long-term, large-  
27 scale analysis of the dynamics of P entering and leaving soils and aquatic systems via a  
28 combination of trade, fluvial transport, and waste transport. We then report net annual P inputs,  
29 and the P mass accumulated over several decades, for three large river basins. Our analyses  
30 reveal rapid historical P accumulation for two mixed agricultural-urban landscapes (Thames  
31 Basin, UK, Yangtze Basin, China), and one rural agricultural landscape (Maumee Basin, USA).  
32 We also show that human modes of P transport involving trade and waste massively dominate  
33 over fluvial transport in these large basins, and we illustrate linkages between fluvial P dynamics  
34 and infrastructure such as wastewater treatment and dams. For Thames and Maumee Basins,  
35 recently there was modest P depletion/drawdown of the P pool accumulated in prior decades,  
36 whereas Yangtze Basin has consistently and rapidly accumulated P since 1980. These first  
37 estimates of the magnitude of long-term, large-scale P accumulation in contrasting settings  
38 illustrate the scope of management challenges surrounding the storage, fate, exploitation, and  
39 reactivation of legacy P that is currently present in the Earth's critical zone.

40

41           Phosphorus (P) is a key requirement for food production. Over the past 75 years,  
42 agricultural demand has increased the rate of global P mobilization four-fold<sup>1-3</sup>. Inefficiencies  
43 and large losses of P occur at many points in food production, and the great majority of P  
44 fertilizer originates in mines<sup>4,5</sup>, raising concerns about long-term supplies of affordable fertilizer  
45<sup>6,7</sup>. Fluvial transport of P from agricultural land, and release of P-rich animal and human wastes  
46 into the environment, have degraded lakes, rivers, reservoirs, and coastal waters with excess P,

47 causing costly damages<sup>8,9</sup>. These widespread inefficiencies in human P use have been  
48 characterized as a wholesale disruption of the global P cycle<sup>6</sup> that for ages has supported  
49 biological productivity through efficient recycling of P.

50 P inputs to agriculture initially increase soil fertility and crop yields, but continued P  
51 application in excess of plant uptake increases the risk of P loss from land to water bodies.  
52 Following storage in soils and aquatic sediments, the associated time lags for P mobilization and  
53 transport can last years to decades<sup>10-12</sup>. This relates to the notion that streams and rivers have a  
54 chemical memory of the past<sup>13,14</sup>, and legacies that delay recovery from water quality  
55 impairment. To date there have been few long-term studies of the landscape-level storage,  
56 transport, and fate of P accumulated in human-dominated basins (but see<sup>8,12,15-17</sup>), although has  
57 been much research on P in large basins over shorter time frames<sup>18</sup>. Similarly, there have been  
58 few direct comparisons of fluvial vs. human modes of P transport at broad scales (but see<sup>19</sup>).  
59 Rather, much research on P has involved studies of relatively short-term processes at the plot  
60 scale or within individual ecosystem types. This reflects the long-standing problem that changes  
61 in landscape-level P storage and legacy P are very difficult to measure directly. To address these  
62 needs, we synthesized diverse agronomic, urban, and river data sets, and examined the long-term  
63 dynamics of P accumulation in three large river basins using a difference approach. In advance  
64 of our calculations for long-term P accumulation, we also examined the dynamics of component  
65 P flows involving trade, fluvial transport, and waste transport (food waste disposal, sewer  
66 infrastructure) which have not been frequently juxtaposed over the long-term at large scales.

67 Our synthesis of long-term P fluxes involves: cropland-dominated Maumee River basin,  
68 USA, tributary to Lake Erie, southernmost of the Laurentian Great Lakes; mixed agricultural-  
69 urban Thames River basin, UK, which drains parts of the London metropolitan area en route to

70 the North Sea; Yangtze River basin, the largest in China, which has undergone rapid population  
71 growth and economic development. To conceptualize these broadscale P dynamics, Haygarth et  
72 al.<sup>21</sup> recently proposed that human-dominated catchments consist of an accumulation phase,  
73 when P gradually builds up, and a depletion phase (Fig. S1, Supplementary Information), when P  
74 inputs decline and mobilization of accumulated P becomes an increasingly important  
75 consideration. Here we test this accumulation-depletion framework, posing three questions: 1)  
76 Which P fluxes drive the long-term dynamics in human-dominated river basins? 2) How do gross  
77 P inputs and outputs, and net P inputs, change over the long-term? 3) How can understanding of  
78 long-term accumulation inform management of P trajectories, regionally, nationally, and  
79 internationally? The Maumee, Thames, and Yangtze Basins differ substantially in terms of socio-  
80 economic history and physiographic features but are linked by common interests of water  
81 security, food security, and resource management that transcend geopolitical hierarchies and  
82 provide lessons about P.

83 Biogeochemical studies of watersheds and landscapes commonly focus on fluvial fluxes  
84 but, in the Anthropocene, the P cycle has become increasingly dominated by human fluxes via  
85 trade of fertilizer and food as well as management of food waste and sewage. Our analysis  
86 provides new evidence that, indeed, human P fluxes massively dominate over the fluvial fluxes,  
87 even for large basins. In the agricultural Maumee Basin, both annual fertilizer P import and  
88 food/feed P export exceeded fluvial P export by 5- to 20-fold (Fig. 1), depending on the year. In  
89 Thames Basin, between World War II (1940) and 1980, fertilizer P import averaged >15-fold  
90 higher than river P export; food/feed P export from farms >7-fold higher; foodwaste P to  
91 landfills >4-fold higher; and P input from sewerage to treatment works (sewage production) >2-  
92 fold higher. Likewise, even during the era of highest sewage P effluent and highest river P export

93 in Thames Basin (1970-1990), mean fertilizer P import, food/feed P export from farms, total  
94 sewage production, and food waste P to landfills were 11, 8.0, 4.0, and 3.3 kilotons (kt) per year,  
95 respectively, compared to only 1.9 kt yr<sup>-1</sup> for river P export. These results for Maumee and  
96 Thames Basins suggest the changes in global fluxes of P since pre-industrial times may rival or  
97 exceed the changes in the global fluxes of N and C that have been reported<sup>1, 21</sup>. These major  
98 human alterations to the global P cycle are compatible with previous findings for heavier  
99 elements<sup>22</sup>, whose pre-industrial cycles in the biosphere were controlled mainly by rock  
100 weathering but now are being mobilized more rapidly from the crust via mining.

101 In the Yangtze River, dissolved P export increased by 10-fold between 1970 and 2010  
102 but our calculations indicate a 44% decline in river total P export between 1970 and 2010  
103 ( $p < 0.001$ , Fig. S5). This reflects a long-term decline in particulate P export that is likely linked to  
104 lower suspended sediment following the construction of large dams<sup>23</sup>, possibly combined with  
105 improvements in sewage treatment. Nonetheless, like Maumee and Thames, total P transport in  
106 Yangtze River was dwarfed by annual fertilizer P application, which increased by more than 10-  
107 fold over this period of record. We suggest the dominance of human P fluxes over fluvial fluxes  
108 extends to many other agricultural and urban basins of the world.

109 The highly agricultural Maumee Basin is the primary source of P to Lake Erie, where the  
110 return of major algae blooms in summer 2014 resulted in the shutdown of the drinking water  
111 supply to Toledo, Ohio<sup>24</sup>. Prior to 1990, and as previously shown by Baker and Richards<sup>25</sup>,  
112 gross P input greatly exceeded gross output (Fig. 2), consistent with expectations for P  
113 accumulation (Fig. S1). Since the late 1990s, gross P input and output have converged towards a  
114 common value between 15 and 20 kt yr<sup>-1</sup>. Our analyses reveal that interannual variations in gross  
115 P input and output in the 1990s and 2000s had only a minor influence on the >200 kt pool of P

116 that accumulated mostly during the 1970s and 1980s (Fig. 3). While annual P output has  
117 exceeded input for certain years (1997-1998, 2006, 2009), our calculations up to 2010 indicate  
118 there has not yet been meaningful P depletion.

119 Unlike Maumee Basin, Thames Basin includes a substantial human population including  
120 parts of the London metropolitan area. Nevertheless, akin to Maumee, gross P input to Thames  
121 Basin greatly exceeded output until the 1990s, demonstrating a prolonged phase of P  
122 accumulation. Since the late 1990s, gross annual P outputs from Thames Basin have slightly  
123 exceeded the inputs. During the 2000s, Thames River P export declined by 86 % ( $p=0.001$ ) in  
124 association with a reduced flux from sewage treatment to river, reflecting higher sewage  
125 treatment efficiency motivated partly by the European Union Water Framework Directive. Over  
126 the same recent period, fertilizer P import declined by 26% ( $p<0.001$ ) while food/feed P export  
127 increased by 22% ( $p=0.044$ ). Thus, our calculations indicate Thames Basin shifted to modest  
128 depletion around 1998, following a long-term decline in fertilizer P import that began around  
129 1960 (Fig. 1 and 3).

130 In contrast to the slowing rates of P accumulation in Maumee and Thames Basins, the  
131 available P data for Yangtze reveal a consistent phase of rapid P accumulation, especially since  
132 1980. We were unable to determine Yangtze Basin sewage inputs ( $P_{sewage,in}$ ) or exports of food  
133 and feed ( $P_{food/feed,out}$ ) needed in Eq. 5 (Supplementary Information), so we did not estimate gross  
134 P input and output for this basin. Nevertheless, we provide estimates of net P input based on the  
135 assumption of  $P_{sewage,in} = P_{food/feed,out}$ . Our calculations reveal that Yangtze Basin, one of Earth's  
136 largest, was accumulating legacy P at a remarkable rate of  $1.7 \text{ Tg yr}^{-1}$  ( $1700 \text{ kt yr}^{-1}$ ) in 2010 (Fig.  
137 3). On an areal basis, Yangtze Basin net annual P input of  $940 \text{ kg km}^{-2} \text{ yr}^{-1}$  in 2010 approaches  
138 the maximum historical rate of P accumulation in Maumee Basin ( $1300 \text{ kg km}^{-2} \text{ yr}^{-1}$  in 1981) and

139 exceeds the maximum historical rate of Thames Basin ( $820 \text{ kg km}^{-2} \text{ yr}^{-1}$  in 1950). This annual  
140 rate of accumulation is also equivalent to about 8% of the global rate of P production from  
141 phosphate rock, or 43% of the national rate of P production by China <sup>2</sup>, suggesting that Yangtze  
142 Basin alone accounts for 17% of the annual P increment of  $10 \text{ Tg yr}^{-1}$  that has been reported for  
143 erodible soils globally <sup>8, 12</sup>. Like Maumee and Thames Basins, much accumulated P in Yangtze  
144 Basin occurs in arable upland soils <sup>26</sup> and eventually could be delivered to water bodies, adding  
145 to the more immediate effects of population change, dam construction, and sewage treatment on  
146 dissolved or particulate P transport by rivers globally <sup>27</sup>. Research is still needed to understand  
147 how interactions between land use change and climate variability affect the mobilization of  
148 legacy P from soils as well as from river channels, reservoirs, floodplains, wetlands, and natural  
149 lakes occurring within hydrologic networks.

150         Here we have demonstrated that large-scale assessments of landscape P storage and  
151 dynamics may be achieved by difference, as previously shown in global analyses of P <sup>8, 12</sup>. This  
152 approach provides a means for estimating the mass of legacy anthropogenic P that is currently  
153 present in the Earth's critical zone, and may inform efforts to exploit it <sup>4</sup>. Contributing challenges  
154 to the direct measurement of change in P storage are that soil P is notoriously heterogeneous in  
155 space and with soil depth, while historical soil sampling efforts have rarely targeted the entire  
156 landscape P pool. Thus, while P flux data are often lacking during early stages of P accumulation  
157 even in intensively monitored basins such as Maumee, there are pathways for long-term analysis  
158 through linkages between the P cycle and documented human activities.

159         Concerns about excess P, its mobilization, and the lack of robust P recycling pathways <sup>5, 6</sup>  
160 are growing worldwide. These kinds of long-term portraits of P storage, mobilization, and  
161 legacies are needed to help understand the true causes and consequences of P transport. We

162 suggest an important role for new technologies and land practices that specifically target legacy  
163 P in terms of storage, fate, exploitation/recovery, and reactivation to more plant-available forms  
164 <sup>16</sup>. While our analysis has focused on a few major P-consuming nations <sup>5</sup>, the need for robust P  
165 recycling pathways extends to developing nations, especially those where mineral P is scarce <sup>28</sup>.  
166 In regions of intense P surplus <sup>29</sup>, managed drawdown of excess soil P represents an increasingly  
167 viable option. As demonstrated by the return of algae blooms to Lake Erie <sup>24, 30</sup>, P dynamics are  
168 complex, requiring vigilance to incorporate both new and historical information into adaptive  
169 management. Improved understanding of long-term time lags for transport <sup>10</sup>, and more timely  
170 updates to spatially- and temporally-explicit data sets on traded goods and wastes containing P,  
171 may help identify strategies that sustain food production while protecting water quality.

172

## 173 Methods

174 We used both published and new data on major P fluxes across the boundaries of the  
175 landscape P pool (soils+aquatic systems), as well as within-basin P transfers. Methods for the net  
176 annual P input calculations were informed by known properties of each basin, including  
177 physiographic setting, human population, and size (Table S1). A summary of the sources of P  
178 flux data and calculations is provided in Table S2. The time series for each P flux, and net annual  
179 P inputs, are provided in Table S3 (Maumee), Table S4 (Thames), and Table S5 (Yangtze), and  
180 we used discrete time in annual intervals. Three linked reasons for our focus on Maumee,  
181 Thames, and Yangtze Basins are: 1) each basin has major human influences that may relate to  
182 the long-term P dynamics; 2) there have been major management, monitoring, and research  
183 efforts in these basins for several decades, leading to the P data sets that provide a unique  
184 opportunity to reconstruct the long-term net P inputs to soils and aquatic systems; 3) the basins



185 differ substantially in terms of socio-economic history and physiographic features but are linked  
186 by common interests of water security, food security, and resource management.

187 We define the basin-level net annual P input ( $P_{net}$ , mass per year) as

$$188 \quad P_{net} = P_{in} - P_{out} \quad (1)$$

189 where  $P_{in}$  is gross annual input and  $P_{out}$  is gross annual output to/from the landscape P pool. In  
190 our conceptualization, human systems such as markets, waste treatment facilities, and landfills  
191 are not components of the landscape P pool, but still may greatly influence it through exchange.  
192 Note that the calculations of  $P_{net}$ ,  $P_{in}$ , and  $P_{out}$  were not merely the summation of the simple  
193 component fluxes plotted in Fig. 1, which includes internal transfers within the basin. Rather, the  
194 net/gross calculations required more thorough book-keeping of new/exogenous P inputs and  
195 permanent outputs across the basin boundaries, not double-counting of the same P mass moved  
196 internally. Gross inputs from equation 1 may be broken down further as

$$197 \quad P_{in} = P_{fert,in} + P_{sewage,in} + P_{precip} \quad (2)$$

198 where  $P_{precip}$  is atmospheric P input from precipitation,  $P_{fert,in}$  is gross mineral fertilizer P import  
199 via trade, and  $P_{sewage,in}$  is the subset of sewage P production that originates from imported  
200 products (food + household cleaners) and enters the environment either as effluent from sewage  
201 treatment or as biosolids/sludge waste applied to soils. The new landscape P input represented by  
202  $P_{sewage,in}$  is not to be confused with total sewage P production plotted in Fig 1. Rather, total  
203 sewage P production contains internally produced food P already accounted as fertilizer input.  
204  $P_{precip}$  in agricultural basins is often small relative to fertilizer use, as evidenced by Maumee  
205 River Basin, where  $P_{precip}$  was reported to be 0.2 kt per yr<sup>25</sup>, or <1% of mean fertilizer P import  
206 over our period of record. Equation 2 simplifies to

$$207 \quad P_{in} = P_{fert,in} + P_{sewage,in} \quad (3)$$

208 under the assumption of  $P_{precip}=0$ . The outputs may be broken down further as

$$209 \quad P_{out} = P_{food/feed,out} + P_{river} \quad (4)$$

210  $P_{food/feed,out}$  is gross P export via food/feed trade and waste transport to landfills, and  $P_{river}$  is P  
211 exported via fluvial transport. Note that un-mined rock-P is not a part of the landscape pool in  
212 our conceptualization, so there is no need to include an export term for fertilizer P. Substituting  
213 equations 3 and 4 into equation 1 gives

$$214 \quad P_{net} = P_{fert,in} + P_{sewage,in} - P_{food/feed,out} - P_{river} \quad (5)$$

215 and we used equation 5 as the central basis for constructing time series of net annual P input.  
216 Accumulated P stores were quantified by taking the cumulative sum of the  $P_{net}$  (t) time series,  
217 across years.

218

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291

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297

298 Author Contributions

299 S.M.P. led the writing of the paper, compiled the data, and analyzed the data. Key P data sets  
300 were contributed by H.P.J., N.J.K.H., F.W., T.W.B., and J.S. All authors participated in the  
301 interpretation of results and the writing and editing process.

302 Figure legends

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304 Figure 1. Component P fluxes used in calculating the net annual P inputs for the three river  
305 basins (Maumee R. USA, Thames R. UK, Yangtze R. China).

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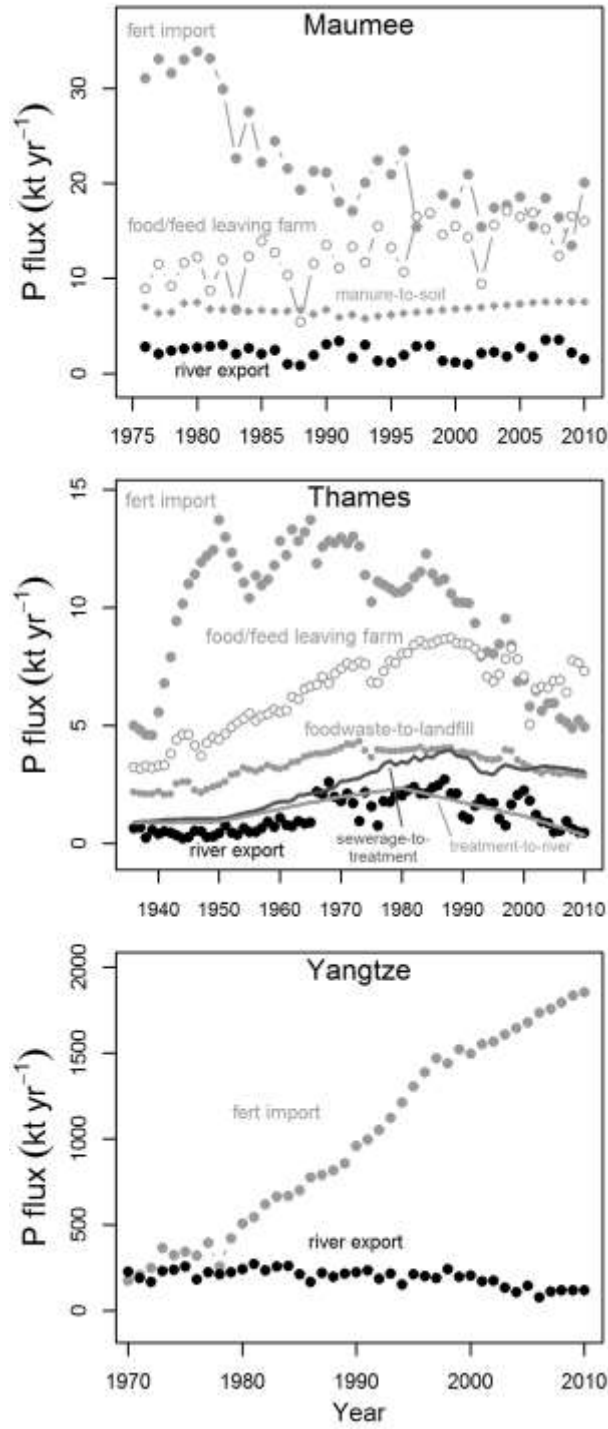
307 Figure 2. Gross P inputs and outputs to/from the landscape P pool (soils + aquatic systems) of  
308 Maumee and Thames Basins. Gross P input includes fertilizer import, and for Thames only,  
309 detergent import. Gross P output includes river export, food/feed exported from the basin via  
310 trade, and for Thames only, disposal of foodwaste to landfill and disposal of sewage biosolids to  
311 landfill, sea, or incinerator.

312

313 Figure 3. Net annual P input and accumulation curves for landscape P pools (soils+aquatic  
314 systems) of three river basins (Maumee R. USA, Thames R. UK, Yangtze R. China).

315 Accumulated P is the cumulative sum of net annual P input over time.

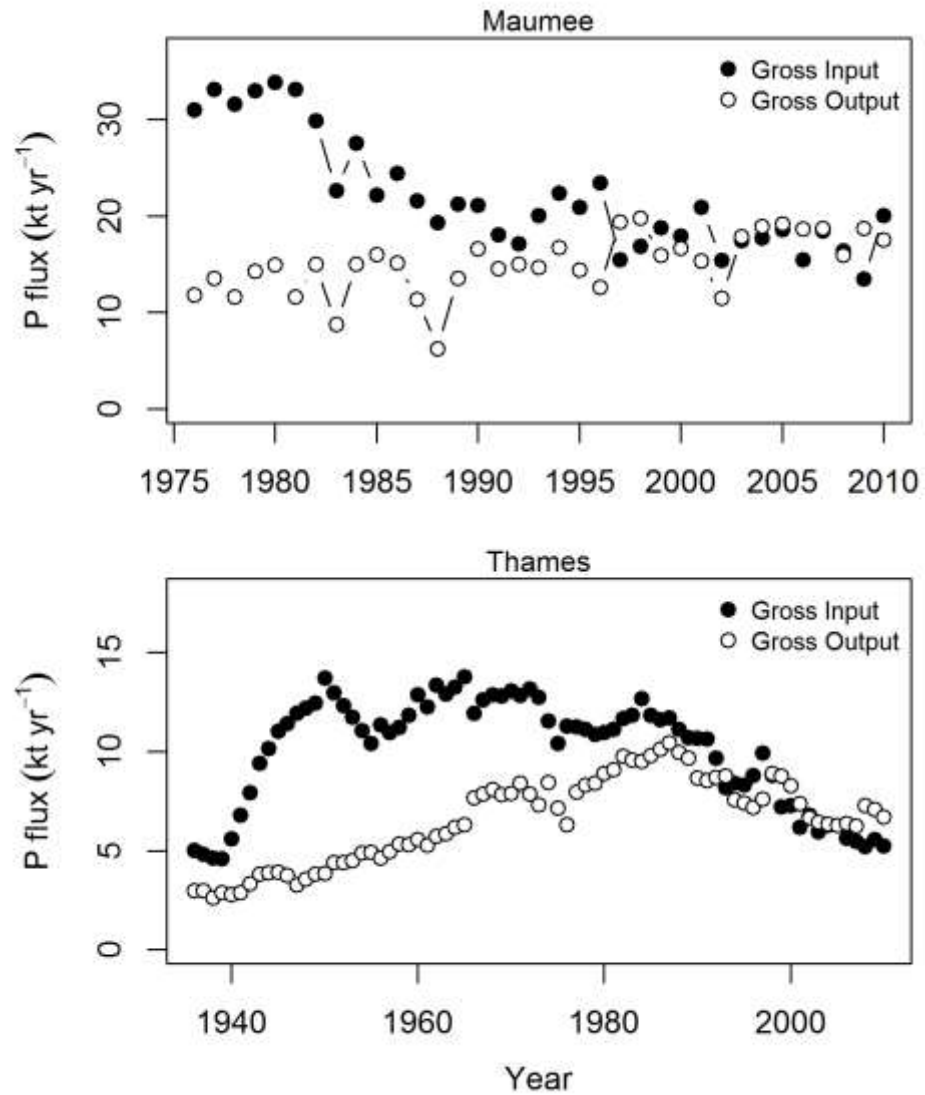
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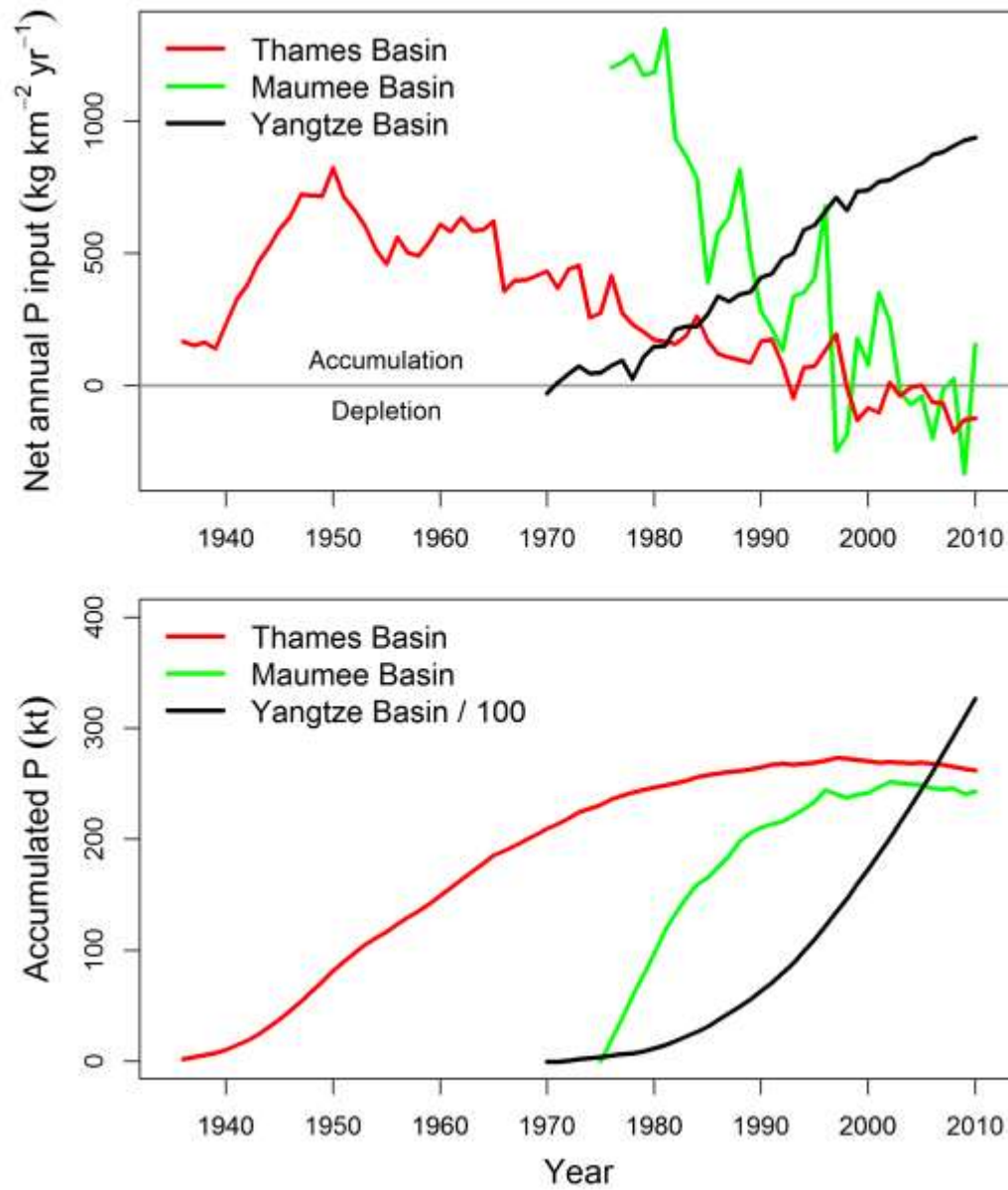
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329 landfill, sea, or incinerator.

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Figure 3. Net annual P input and P accumulation curves for the landscape P pools (soils+aquatic systems) of three river basins (Maumee R. USA, Thames R. UK, Yangtze R. China). Accumulated P is the cumulative sum of net annual P input over time.