

1 Title:

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

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23 Global food production depends on phosphorus. Phosphorus is broadly applied as
24 fertilizer but excess phosphorus contributes to eutrophication of surface water bodies and coastal
25 ecosystems¹. Here we present an analysis of phosphorus fluxes in three large river basins using
26 published data on fertilizer, harvested crops, sewage, food waste and river fluxes^{2,3,4}. Our
27 analyses reveal that the magnitude of phosphorus accumulation has varied greatly over the past
28 30-70 years in mixed agricultural-urban landscapes of the Thames Basin, UK, Yangtze Basin,
29 China, and the rural Maumee Basin, USA. Fluxes of phosphorus in fertilizer, crop harvest, food
30 waste, and sewage dominate over the river fluxes. Since the late 1990s, net exports from the
31 Thames and Maumee Basins have exceeded inputs, suggesting net mobilization of the
32 phosphorus pool accumulated in prior decades. In contrast, the Yangtze Basin has consistently
33 accumulated phosphorus since 1980. Infrastructure modifications such as sewage treatment and
34 dams may explain more recent declines in total phosphorus fluxes from the Thames and Yangtze
35 Rivers^{3,4}. We conclude that human-dominated river basins may undergo a prolonged but finite
36 accumulation phase when phosphorus inputs exceed agricultural demand, and this accumulated
37 phosphorus may continue to mobilize long after inputs decline.

38 Over the past 75 years, agricultural demand has increased the rate of global P
39 mobilization four-fold^{5,6,7}. Inefficiencies and large P losses occur at many points in food
40 production, and the majority of P fertilizer originates in mines^{8,9}, raising concerns about long-
41 term supplies of affordable fertilizer^{10,11}. Fluvial P transport from agricultural land, and release
42 of P-rich animal and human wastes into the environment, have degraded lakes, rivers, reservoirs,
43 and coastal waters with excess P, causing costly damages^{1,12}. These widespread inefficiencies in
44 human P use have been characterized as a wholesale disruption of the global P cycle¹⁰ that for
45 ages has supported biological productivity through efficient P recycling.

46 P inputs to agriculture initially increase soil fertility and crop yields, but continued P
47 application in excess of plant uptake increases the risk of P loss from land to water bodies.
48 Following storage in soils and aquatic sediments, the associated time lags for P mobilization and
49 transport can last years to decades¹³⁻¹⁵. This relates to the notion that streams and rivers have a
50 chemical memory of the past^{16,17}, and legacies that delay recovery from water quality
51 impairment. To date there have been few long-term studies of the landscape-level storage,
52 transport, and fate of P accumulated in human-dominated basins (but see^{12, 15,18-20}), although
53 there has been much research on P in large basins over shorter time frames²¹. Similarly, there
54 have been few direct comparisons of fluvial versus human modes of P transport at broad scales
55 (but see²²). Rather, much P research has involved studies of relatively short-term processes at
56 the plot scale or within individual ecosystems, reflecting the long-standing problem that changes
57 in landscape-level P storage and legacy P are difficult to measure directly. To address these
58 needs, we synthesized diverse agronomic, urban, and river data sets, and examined the long-term
59 dynamics of P accumulation in three large river basins using a difference approach. In advance
60 of our calculations for long-term P accumulation, we examined the dynamics of component P
61 flows involving trade, fluvial transport, and waste transport (food waste disposal, sewage
62 infrastructure) which have not been frequently juxtaposed over the long-term at large scales.

63 Our synthesis of long-term P fluxes involves: cropland-dominated Maumee River basin,
64 USA, tributary to Lake Erie, southernmost of the Laurentian Great Lakes; mixed agricultural-
65 urban Thames River basin, UK, which drains parts of the London metropolitan area *en route* to
66 the North Sea; Yangtze River basin, the largest in China, which has undergone rapid population
67 growth and economic development. To conceptualize these broadscale P dynamics, Haygarth et
68 al.⁴ recently proposed that human-dominated catchments consist of an accumulation phase, when

69 P gradually builds up, and a depletion phase (Fig. S1, Supplementary Information), when P
70 inputs decline and mobilization of accumulated P becomes an increasingly important
71 consideration. Here we test this accumulation-depletion framework, posing three questions: 1)
72 Which P fluxes drive the long-term dynamics in human-dominated river basins? 2) How do gross
73 P inputs and outputs, and net P inputs, change over the long-term? 3) How can understanding of
74 long-term accumulation inform management of P trajectories, regionally, nationally, and
75 internationally? The Maumee, Thames, and Yangtze Basins differ substantially in terms of socio-
76 economic history and physiographic features but are linked by common interests of water
77 security, food security, and resource management that transcend geopolitical hierarchies and
78 provide lessons about P.

79 Biogeochemical studies of watersheds and landscapes commonly focus on fluvial fluxes
80 but, in the Anthropocene, the P cycle has become increasingly dominated by human fluxes via
81 fertilizer and food trade as well as food waste and sewage management. Our analysis provides
82 new evidence that, indeed, human P fluxes massively dominate over the fluvial fluxes, even for
83 large basins. In the agricultural Maumee Basin, annual fertilizer P import and food/feed P export
84 exceeded fluvial P export by 5- to 20-fold (Fig. 1), depending on the year. In Thames Basin,
85 between World War II (1940) and 1980, fertilizer P import averaged >15-fold higher than river P
86 export; food/feed P export from farms >7-fold higher; food waste P to landfills >4-fold higher;
87 and P input from sewage to treatment works >2-fold higher. Likewise, even during the era of
88 highest sewage P effluent and highest river P export in Thames Basin (1970-1990), mean
89 fertilizer P import, food/feed P export from farms, total sewage production, and food waste P to
90 landfills were 11, 8.0, 4.0, and 3.3 kilotons (kt) per year, respectively, compared to only 1.9 kt yr⁻¹
91 for river P export. These results for Maumee and Thames Basins suggest the changes in global

92 P fluxes since pre-industrial times may rival or exceed the changes in the global N and C fluxes
93 that have been reported^{5,23}. These major human alterations to the global P cycle are compatible
94 with previous findings for heavier elements²⁴, whose pre-industrial cycles were controlled
95 mainly by rock weathering, but are now mobilized from the crust through mining.

96 In the Yangtze River, dissolved P export increased by 10-fold between 1970 and 2010
97 but our calculations indicate a 44% decline in river total P export between 1970 and 2010
98 ($p < 0.001$, Fig. S5). This reflects a long-term decline in particulate P export that is likely linked to
99 lower suspended sediment following the construction of large dams³, possibly combined with
100 improvements in sewage treatment. Nonetheless, like Maumee and Thames, total P transport in
101 Yangtze River was dwarfed by annual fertilizer P application, which increased by more than 10-
102 fold over this period of record. We suggest the dominance of human P fluxes over fluvial fluxes
103 extends to many other agricultural and urban basins of the world.

104 The highly agricultural Maumee Basin is the primary P source to Lake Erie, where major
105 algae blooms returned in summer 2014, causing the shutdown of drinking water supplies to
106 Toledo, Ohio²⁵. Prior to 1990, and as previously shown², gross P input exceeded gross output
107 (Fig. 2), consistent with expectations for P accumulation (Fig. S1). Since the late 1990s, gross P
108 input and output have converged towards a common value between 15 and 20 kt yr⁻¹. Our
109 analyses reveal that interannual variations in gross P input and output in the 1990s and 2000s had
110 only minor influence on the >200 kt pool of P that accumulated mostly during the 1970s and
111 1980s (Fig. 3). While annual P output has exceeded input for certain years (1997-1998, 2006,
112 2009), our calculations up to 2010 indicate there has not yet been meaningful P depletion.

113 Unlike Maumee Basin, the Thames Basin has a substantial urban population including
114 parts of Greater London. Nevertheless, akin to Maumee, gross P input to Thames Basin greatly

115 exceeded output until the 1990s, demonstrating a prolonged phase of P accumulation. Since the
116 late 1990s, gross annual P outputs from Thames Basin have slightly exceeded the inputs. During
117 the 2000s, Thames River P export declined by 86 % ($p=0.001$) in association with a reduced flux
118 from sewage treatment to river, reflecting higher sewage treatment efficiency motivated partly
119 by the European Union's Urban Waste Water Directive. Over the same recent period, fertilizer P
120 import declined 26% ($p<0.001$) while food/feed P export increased 22% ($p=0.044$). Thus, our
121 calculations indicate Thames Basin shifted to modest depletion around 1998, following a long-
122 term decline in fertilizer P import that began around 1960 (Fig. 1 and 3).

123 In contrast to the slowing rates of P accumulation in Maumee and Thames Basins, the
124 available P data for Yangtze reveal a consistent phase of rapid P accumulation, especially since
125 1980. We were unable to determine Yangtze Basin sewage inputs ($P_{sewage,in}$) or exports of food
126 and feed ($P_{food/feed,out}$) needed in Eq. 5 (Supplementary Information), so we did not estimate gross
127 P input and output for this basin. Nevertheless, we provide estimates of net P input based on the
128 assumption of $P_{sewage,in} = P_{food/feed,out}$. Our calculations reveal that Yangtze Basin, one of Earth's
129 largest, was accumulating legacy P at a remarkable rate of 1.7 Tg yr^{-1} (1700 kt yr^{-1}) in 2010 (Fig.
130 3). On an areal basis, Yangtze Basin net annual P input of $940 \text{ kg km}^{-2} \text{ yr}^{-1}$ in 2010 approaches
131 the maximum historical rate of P accumulation in Maumee Basin ($1300 \text{ kg km}^{-2} \text{ yr}^{-1}$ in 1981) and
132 exceeds the maximum historical rate of Thames Basin ($820 \text{ kg km}^{-2} \text{ yr}^{-1}$ in 1950). This annual
133 rate of accumulation is also equivalent to about 8% of the global rate of P production from
134 phosphate rock, or 43% of the national rate of P production by China⁶, suggesting that Yangtze
135 Basin alone accounts for 17% of the annual P increment of 10 Tg yr^{-1} that has been reported for
136 erodible soils globally^{12,15}. Like Maumee and Thames Basins, much accumulated P in Yangtze
137 Basin occurs in arable upland soils²⁶ and eventually could be delivered to water bodies, adding to

138 the more immediate effects of population change, dam construction, and sewage treatment on
139 dissolved or particulate P transport by rivers globally²⁷. Research is still needed to understand
140 how interactions between land use change and climate variability affect the mobilization of
141 legacy P from soils as well as from river channels, reservoirs, floodplains, wetlands, and natural
142 lakes occurring within hydrologic networks.

143 Here we demonstrated that large-scale assessments of landscape P storage and dynamics
144 may be achieved by difference, as previously shown in global analyses of P^{12,15}. This approach
145 provides a means for estimating the mass of legacy anthropogenic P currently present in the
146 Earth's critical zone, and may inform efforts to exploit it⁸. Contributing challenges to the direct
147 measurement of change in P storage are that soil P is notoriously heterogeneous in space and
148 with soil depth, while historical soil sampling efforts rarely encompass the entire landscape P
149 pool. Thus, while P flux data are often lacking during early stages of P accumulation even in
150 intensively monitored basins such as Maumee, there are pathways for long-term analysis through
151 linkages between the P cycle and documented human activities.

152 Concerns about excess P, its mobilization, and the lack of robust P recycling pathways^{9,10}
153 are growing worldwide. These kinds of long-term portraits of P storage, mobilization, and
154 legacies are needed to help understand the true causes and consequences of P transport. We
155 suggest an important role for new technologies and land practices that specifically target legacy
156 P in terms of storage, fate, exploitation/recovery, and reactivation to more plant-available
157 forms¹⁹. While our analysis has focused on a few major P-consuming nations⁹, the need for
158 robust P recycling pathways extends to developing nations, especially those where mineral P is
159 scarce²⁸. In regions of intense P surplus²⁹, managed drawdown of excess soil P represents an
160 increasingly viable option. As demonstrated by the return of algae blooms to Lake Erie^{25,30}, P

161 dynamics are complex, requiring vigilance to incorporate both new and historical information
162 into adaptive management. Improved understanding of long-term time lags for transport¹³, and
163 more timely updates to spatially- and temporally-explicit data sets on traded goods and wastes
164 containing P, may help identify strategies that sustain food production while protecting water
165 quality.

166

167 Methods

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

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

$$182 \quad P_{net} = P_{in} - P_{out} \quad (1)$$

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

$$191 \quad P_{in} = P_{fert,in} + P_{sewage,in} + P_{precip} \quad (2)$$

192 where P_{precip} is atmospheric P input from precipitation, $P_{fert,in}$ is gross mineral fertilizer P import,
 193 and $P_{sewage,in}$ is the subset of sewage P production that originates from imported products (food +
 194 household cleaners) and enters the environment either as effluent from sewage treatment or as
 195 biosolids/sludge waste applied to soils. The new landscape P input represented by $P_{sewage,in}$ is not
 196 to be confused with total sewage P production plotted in Fig 1. Rather, total sewage P production
 197 contains internally produced food P already accounted as fertilizer input. P_{precip} in agricultural
 198 basins is often small relative to fertilizer use, as evidenced by Maumee River Basin, where P_{precip}
 199 was reported to be 0.2 kt per yr², or <1% of mean fertilizer P import over our period of record.

200 Equation 2 simplifies to

$$201 \quad P_{in} = P_{fert,in} + P_{sewage,in} \quad (3)$$

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

$$203 \quad P_{out} = P_{food/feed,out} + P_{river} \quad (4)$$

204 $P_{food/feed,out}$ is gross P export via food/feed trade and waste transport to landfills, and P_{river} is P
 205 exported via fluvial transport. Note that un-mined rock-P is not a part of the landscape pool in

206 our conceptualization, so there is no need to include an export term for fertilizer P. Substituting
207 equations 3 and 4 into equation 1 gives

$$208 \quad P_{net} = P_{fert,in} + P_{sewage,in} - P_{food/feed,out} - P_{river} \quad (5)$$

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

212

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294 Author Contributions

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

298 Figure legends

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

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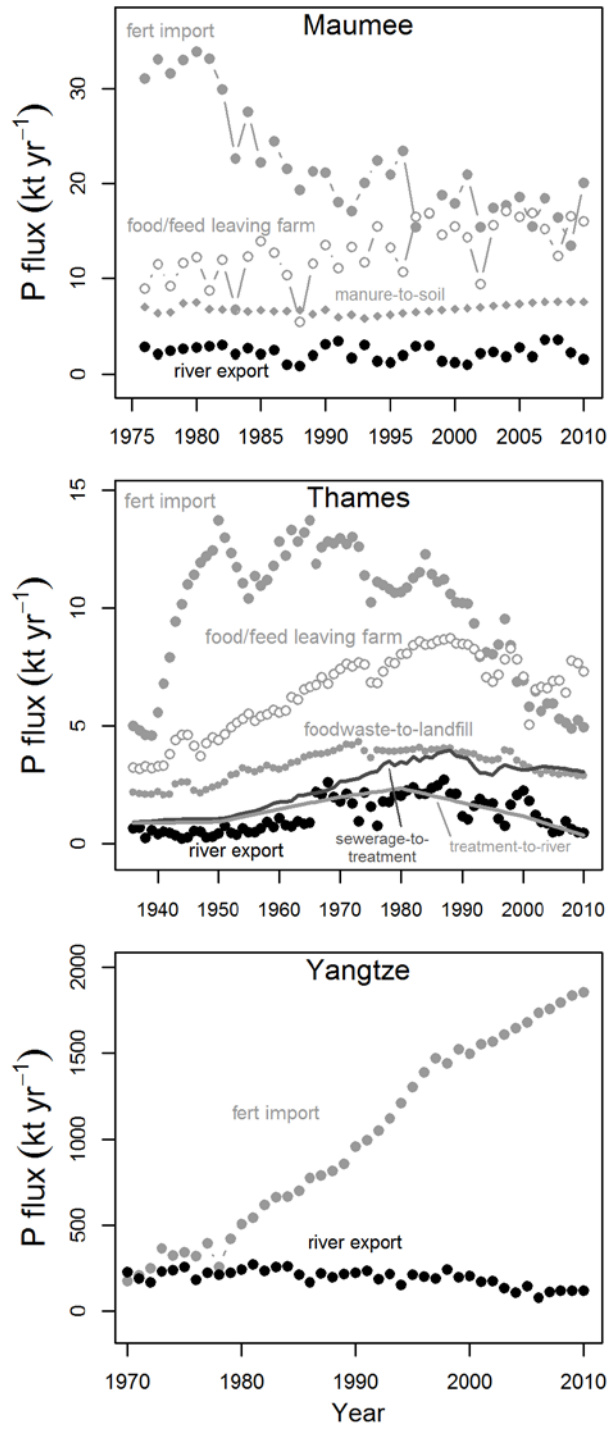
303 Figure 2. Gross P inputs and outputs to/from the landscape P pool (soils + aquatic systems) of
304 Maumee and Thames Basins. Gross P input includes fertilizer import, and for Thames only,
305 detergent import. Gross P output includes river export, food/feed exported from the basin via
306 trade,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

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

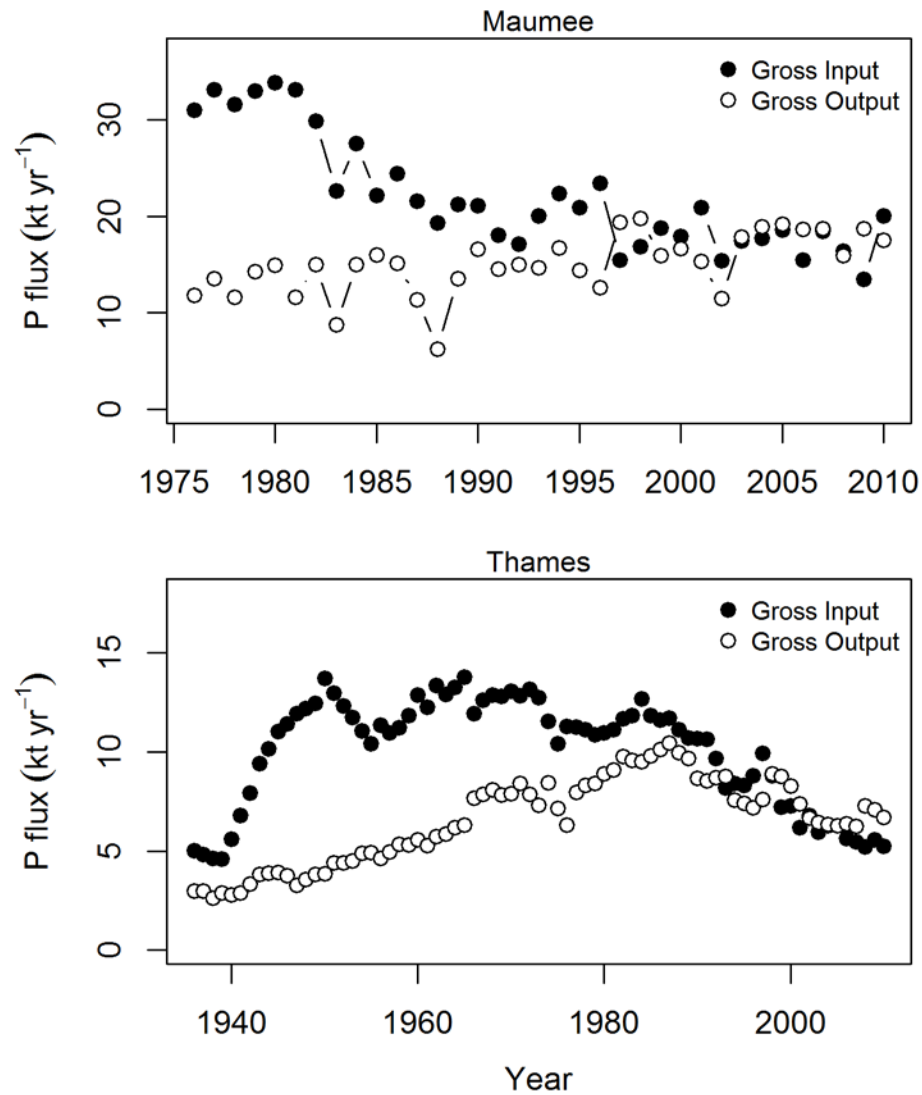
311 Accumulated P is the cumulative sum of net annual P input over time.

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



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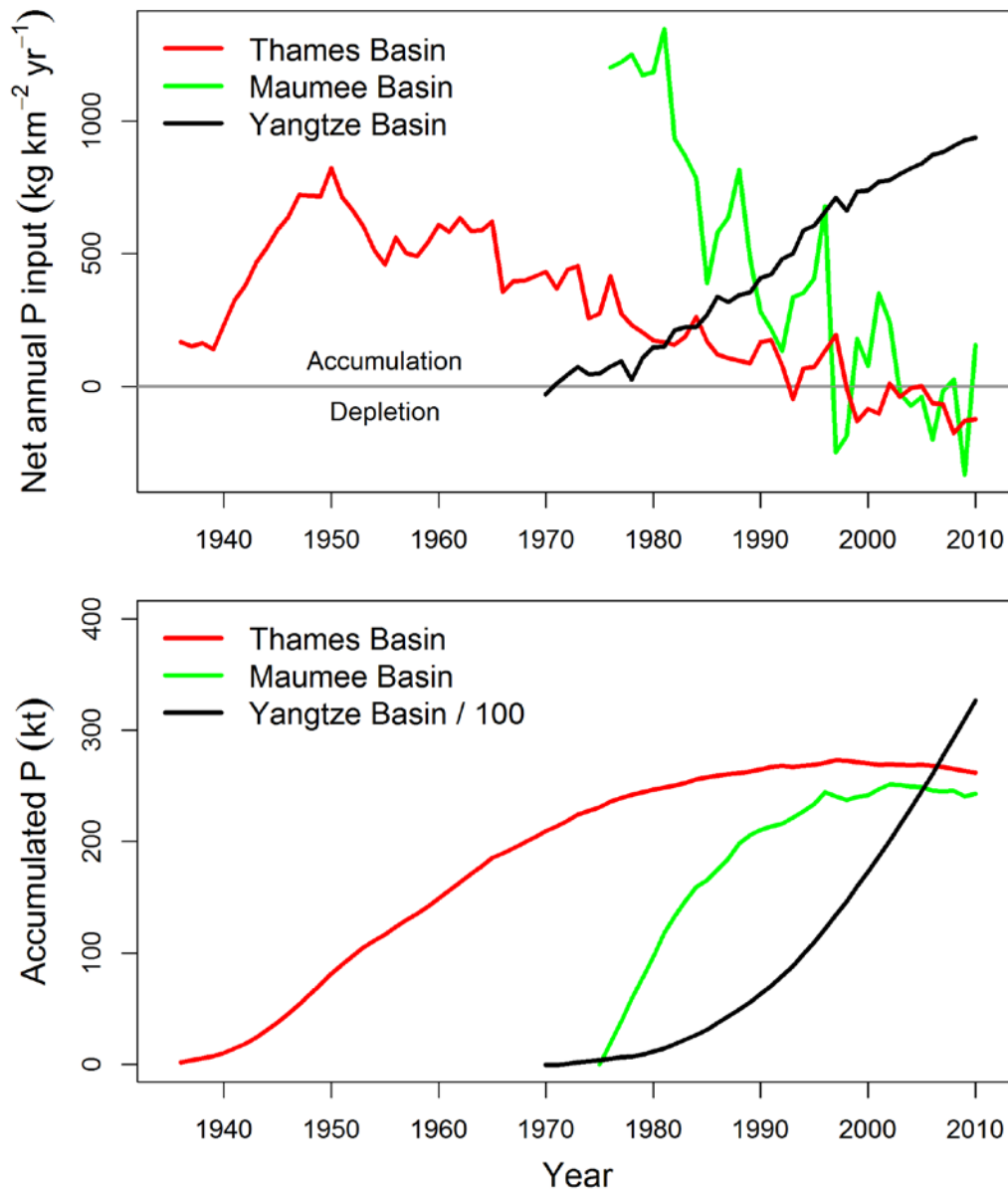
322 Maumee and Thames Basins. Gross P input includes fertilizer import, and for Thames only,

323 detergent import. Gross P output includes river export, food/feed exported from the basin via

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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.