Anthropogenic signature of global agricultural soil phosphorus

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Abstract

The global phosphorus (P) cycle has been dramatically altered by human activities through the use of mineral P fertilizers, often referred to anthropogenic P. The application of mineral P fertilizers on agricultural soils has driven the planet beyond its safe operating space but the extent to which the global P cycle relies on anthropogenic P has never been quantified. To fill this gap, we developed a model that simulates, at the country scale, the evolution of agricultural soil available P by distinguishing anthropogenic vs. natural P pools, and by accounting for farming practices, crop-livestock recycling loop, and agricultural trade, over the 1950-2017 period. At the global scale we found that the anthropogenic signature of soil available P was 45% ± 8% in 2017. The national anthropogenic signatures varied according to the cumulative mineral P fertilizer use as well as to the soil P availability in 1950. Despite having different historical trajectories, Western Europe, North America and Asia displayed similar reliance on anthropogenic P, close to 60% in 2017. Conversely, African soil anthropogenic signature remained around 30%. Trade did not modify the simulated signatures. Overall, our results unravel the strong reliance of our soil fertility and food production systems on anthropogenic P resources.

Main text

The Anthropocene is characterised by the profound, anthropogenic disturbance of the global biogeochemical cycles, which has led to drastic consequences for soil fertility, ocean and river quality, greenhouse gas emissions and biodiversity losses (Steffen et al., 2015). In particular, the global phosphorus (P) cycle has been altered in an unprecedented way, both in time and in space (Elser and Bennett, 2011). At the core of these modifications stand phosphate rocks and their use in agriculture, mainly as mineral P fertilizers but also, although at a lesser extent, as mineral feed for livestock animals. Both mineral P fertilizers and mineral P feed are derived from industrial treatments – to increase the P solubility of phosphate rocks – and are hereafter referred to as anthropogenic P.

The massive use of mineral P fertilizers combined with a sharp rise in livestock animals number – and resulting manure production – have globally increased cropland soil available P, although with some variations across world regions (Ringeval et al., 2017). The changes in croplands soil P fertility have been triggered by both local and global modifications in the P cycle. At the local scale, P has been added to agricultural soils through massive application of mineral P fertilizers –sometimes far above crop P uptake – resulting in increasing soil P legacy, together with some P deficits in some regions (MacDonald et al., 2011). Large amounts of P have also been transferred from grasslands to

croplands through the production, transport and soil application of animal manure (Sattari et al., 2016), a phenomenon much increased by the massive rise in global livestock population (Bouwman et al., 2011). At the global scale, P has been displaced through the extraction and international trade of phosphate rocks (Nesme et al., 2018). Highly concentrated in a few places such as Morocco and Western Sahara, phosphate rocks have been redistributed, yet unevenly, to the rest of the world, thus reshaping the spatial distribution of soil P. More recently, the trade of agricultural products has also contributed to displace large quantities of P worldwide (Lun et al., 2021). Indirectly this traded P has fertilized, at least partly, the agricultural soils of importing countries through the application of animal and human manure - derived from traded feed and food - on cropland and grassland soils.

This alteration of the global P cycle has had several, positive and negative consequences. On one hand, crop yields have increased as a result of an overall rise in soil P fertility, thereby helping to achieve food security objectives (Mueller et al., 2012). On the other hand, our agriculture has become dependent on phosphate rocks, an alarming situation due to the progressive exhaustion of the easily accessible, remaining reserves of this fossil ressource (Cordell et al., 2009; Reijnders, 2014). Although it will take centuries for phosphate rock reserves to be depleted, short-term challenges are likely to arise in the next years to decades (Scholz and Wellmer, 2018). The prices of mineral P fertilizers are likely to increase in most world regions and potential geopolitical conflicts might break out, thus putting at risk the resilience of our global food systems (Barbieri et al., 2022; Cordell et al., 2009; Van Vuuren et al., 2010).

Those alterations of the global P cycle and of soil P fertility following anthropogenic P supply have already been highlighted by recent synthesis studies (Chen and Graedel, 2016; Elser and Bennett, 2011; Lu and Tian, 2016). However, the specific and cumulated contribution of anthropogenic P supply to P fertility of global agricultural soils has never been estimated. Providing such estimate is key to assess both the anthropogenic disturbance of the global P cycle but also to estimate the past and current reliance of agricultural soils and food production to the finite phosphate rock resources. Here we address this knowledge gap by estimating the anthropogenic signature of available P in agricultural soils at the global scale. We defined the anthropogenic signature of soil available P as the ratio of anthropogenic over total soil available P, where total P refers to the sum of non-anthropogenic P (considered as natural P) and anthropogenic P.

To achieve this objective, we developed a model that simulates for each country and with a yearly time step, the evolution of agricultural soil P stocks (in both cropland and grassland soils) from 1950 to 2017, accounting for (i) direct P fertilizer inputs to soils but also (ii) P recycling from the crop-livestock cycling loop and (iii) geographic displacement of P through the trade of agricultural

products (Figure 1). We modelled soil available P as the interaction between a labile P pool (LP) and a stable P pool (SP), which is commonly done in the literature (Le Noë et al., 2020; Sattari et al., 2012; Zhang et al., 2017). The labile P pool referred to soil P directly available for plant uptake while the stable P pool acted as a slow-release buffer that could replenish the labile P pool following crop P uptake. Each pool was further broken down into a natural (Nat) vs. an anthropogenic (Ant) compartment. This allowed us to track the behaviour of P inputs according to their anthropic vs natural origin in a similar way as isotopic labelling approaches (Sebilo et al., 2013). For each country, the size of the four P pools (LP_{Nat}, LP_{Ant}, SP_{Nat}, SP_{Ant}) were simulated following the size of the previous years, soil P inputs and outputs, and soil P exchanges between labile and stable P pools (see Method).

In our model, P harvested from cropland and grassland soils ($P_{harvest,Tot}$) was computed as a function of the size of the labile P pool (Equation 2). The two parameters involved in the relation between P harvest and the size of the labile P pool were calibrated for each country. Similarly, and in an attempt to take into account the soil P dynamic specificities of each country, we calibrated the P exchange function between stable and labile soil P pools (Equation 3). Both calibrations were conducted independently for each country and were performed so that the model outputs could reproduce both P harvest time-series (from FAOSTAT) over the 1950-2017 period and the size of the labile P pool at year 2005 (derived from the global modelling approach of (Ringeval et al., 2017) or from observations for the few countries where data were available) (see Method).

In addition to soil P pools, we also modelled the anthropogenic signature of all P fluxes incoming to or outgoing from agricultural soils (Figure 1). We assumed that P harvest had the same anthropogenic signature of that of the soil labile P pool in which plant uptake occurred. Similarly, we assumed that imported food and feed products had the same P signature as that of the soil labile P pool of the countries where they came from. Because P losses from soils may apply to both labile and stable P pools indifferently, we assumed that the signature of P losses was similar to that of the overall soil P pools. The anthropogenic P signature of animal manure was assumed equal to that of animal intake. By construction and because they are entirely processed from the chemical industry, an anthropogenic signature of 1 was attributed to both mineral P fertilizers applied to soils and to mineral feed supplements given to livestock (Figure 1).

We initialized the size of the P pools by assuming that, although some mineral P fertilizers were sometimes applied to agricultural soils in the first half of the 20th century (Bouwman et al., 2011; Grigg, 1987; Ringeval et al., 2014), anthropogenic soil P pools were negligible before 1950. The sizes of the labile and stable natural P pools were estimated based on the observed crop and forage P

harvest in 1950 and by assuming an equilibrium between the two soil P pools (see Methods). Hereafter, we comment the signature of the soil labile P pool both in 2017 and throughout the 1950-2017 period.

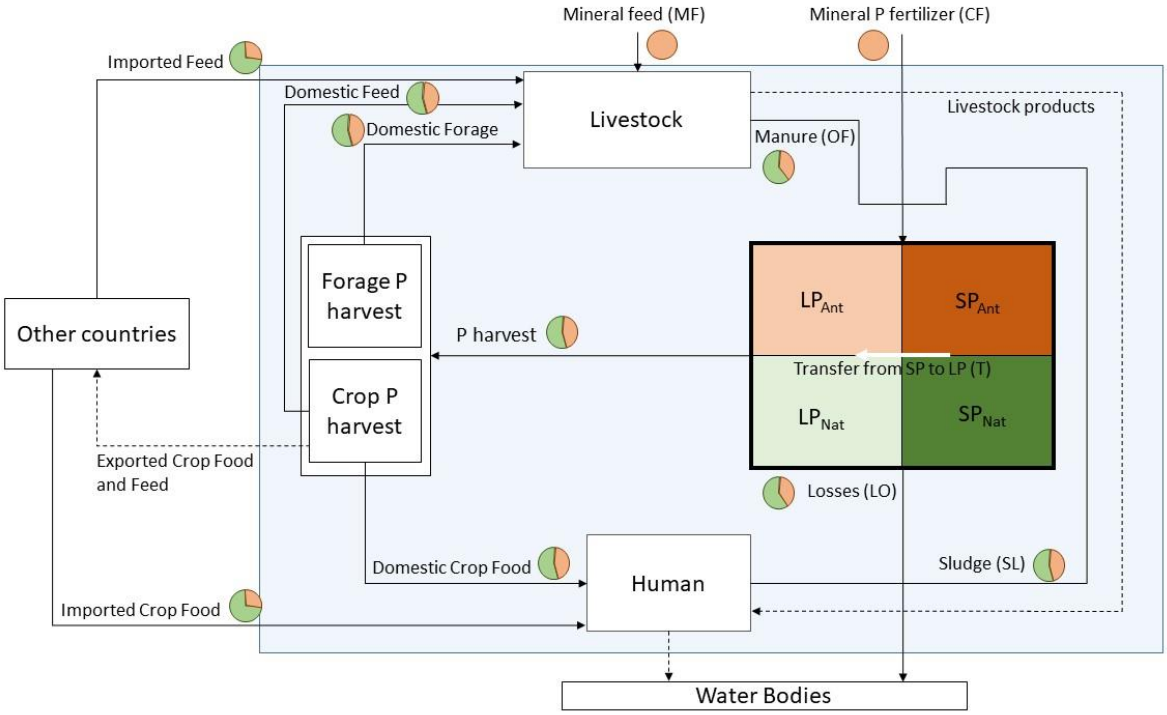

Figure 1 - Structure of the model for a given country, with specific focus on soil P compartments. LP and SP refer to the labile and stable soil P pools, respectively. The anthropogenic vs. natural soil P pools are represented in orange and green respectively. The fluxes in solid lines were explicitly simulated. Conversely, those in dotted lines were not modelled because they do not modify the anthropogenic signature of P pools. As illustrated by the pie charts, each flux was subdivided into anthropogenic (orange) vs. natural (green) components. The blue box refers to a given country that interacts with the rest of the world (other countries) through the trade of food and feed products.

Despite large spatial variabilities, the current global anthropogenic signature of the soil P is high

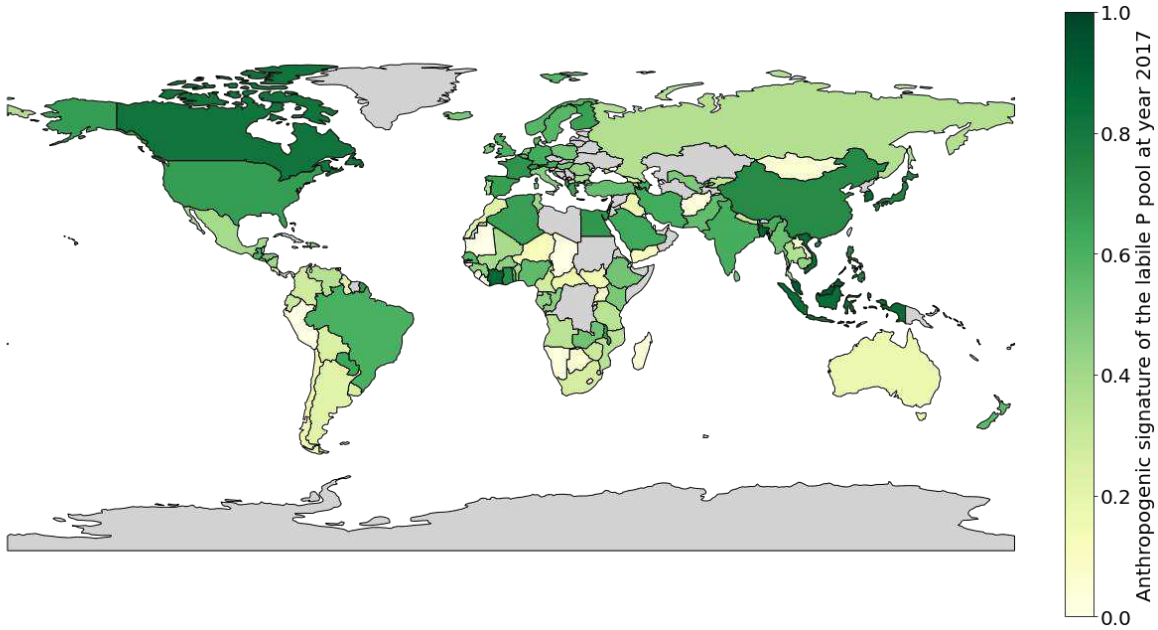

Figure 2 - Anthropogenic signature of the labile P pool of agricultural soils in 2017. Data are displayed per country. Countries coloured in grey were excluded from the calculation either because of missing data (as for Democratic Republic of Congo, Libya and Somalia among others) or because their calibration did not succeed (Cuba, Estonia, Jordan, Kazakhstan, Latvia, Lithuania, Tajikistan, Turkmenistan, and Ukraine).

Our results showed that the use of anthropogenic P - derived from phosphate rocks - greatly modified agricultural soil P fertility worldwide (Figure 2). In 2017, the global mean anthropogenic signature of the soil labile P pool was 45 ±8 %, suggesting that almost half of the global agricultural soil P fertility was derived from anthropic P supply. This result clearly mirrors the overall intensification growth path of the world agriculture, which has relied on the massive use of agricultural inputs and mineral fertilizers from the 1950s (Coomes et al., 2019).

Our findings also highlight large spatial variabilities of anthropogenic soil P signatures among world regions (Figure 2). North America, Western Europe and Asia displayed the highest anthropogenic signatures, with values of 69 ±8 %, 61 ± 8 % and 59 ± 6 %, respectively. Central and South America had an intermediate anthropogenic signature of 41 ±9 %, close to that of Eastern Europe (39 ±10 %). In Africa, the soil P anthropogenic signature remained moderate at 30 ±6 %. Finally, Oceania, which includes Australia and New Zealand, had the lowest anthropogenic signature with value of 18 ±11 %.

We also found that the anthropogenic signatures of soil P were in fact explained by two main factors: (i) the cumulative application of anthropogenic P - mostly as mineral P fertilizer - over the 1950-2017 period and (ii) the initial soil P status in 1950 – assimilated to natural P in our model (Section S8). As a result, the massive mineral P fertilization in North America, Western Europe and Asia since the mid-20th century translated into high anthropogenic signatures in those regions. Besides, high initial soil P pools in 1950 in Western Europe mechanistically ‘diluted’ the massive anthropogenic P supply to agricultural soils experienced by this region, which has been three times higher than in North

America and Asia (Table S8). This explains why despite very different cumulated mineral P fertilization (Figure S15), the three regions all displayed similar anthropogenic signatures in 2017. Finally, and contrary to the stable P pool, the anthropogenic signature of the labile P pool was dependant on a third and last driver: the P transfer between the soil P pools (Text S8), which underlines the need to account for soil P dynamics when simulating soil P availability and its changes over time.

Highly different trends in anthropogenic signature of soil P among countries over 1950-2017

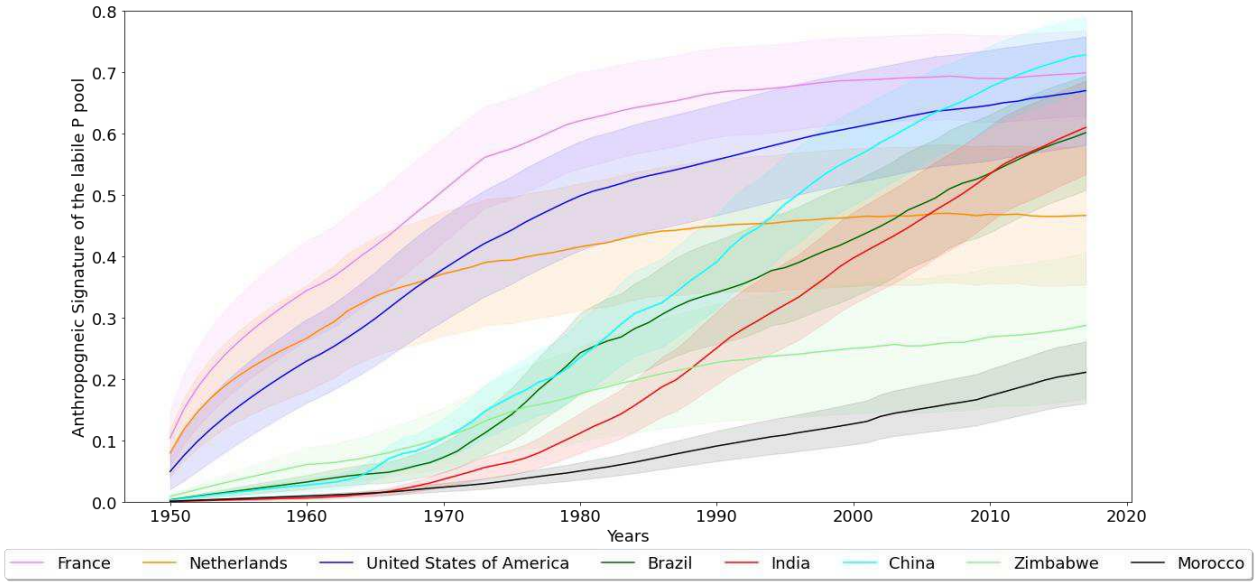

Figure 3 – Temporal evolution of the anthropogenic signature of the soil labile P pool for eight contrasting countries (France, the Netherlands, the United States of America, Brazil, India, China, Zimbabwe and Morocco) for the 1950-2017 period. Mean (solid line) and standard deviation (blurred zone) data are shown.

We found that the soil P anthropogenic signatures displayed highly variable historical trajectories among countries (Figure 3), thereby reflecting contrasting and region-specific histories of soil P fertilization (Figure S16-S17). More precisely, our results showed that the anthropogenic signature of industrialised Western countries such as France, the Netherlands and the USA rose sharply from the 1950s to the 1970s (Figure 3), as a result of the very early and massive application of mineral P fertilizers on agricultural soils in industrialised, Western countries. However, since the 1980s the anthropogenic signatures of France and the Netherlands have stabilized around values of 70 ± 7% and 47 ± 11% (in 2017), respectively. Conversely, the signature of the soil labile P pool in the USA was still raising in 2017. The observed stabilization in France and in the Netherlands resulted from smaller mineral P inputs over the last four decades, partly compensated by higher manure inputs with less effects on the anthropogenic signatures (Figure 4). Indeed, when imported feed are small compared to domestically produced feed – which is the general case –, manure simply recycles P from domestic

feed and fodder back to agricultural soils, without affecting the anthropogenic signature of agricultural soils.

Our results also showed that the soil P anthropogenic signatures of Brazil, India and China took off in the 1970s and 1980s up to values of 60 ± 9%, 61 ± 8% and 73 ± 6% in 2017, respectively (Figure 3). Those results reflect the Green Revolution and the sudden and sharp increase in mineral P fertilizer use that transformed agriculture and soil fertility of most Asian and South American countries from the 1970s (Figure S16). Interestingly, the signatures of those countries have become similar and even higher compared to those of Western European countries in 2017.

Finally, the soil P anthropogenic signatures of most African countries e.g. Morocco and Zimbabwe exhibited a late and slow take off, with values remaining low and close to 20-30% in 2017. This is mainly due to small applications of mineral P fertilizers, resulting on strong limitation of crop yields by soil P availability in those countries (Kvakíc et al., 2018).

Overall, these results illustrate the 'Great Divergence' (Pomeranz, 2000) that, from the 1950s, has seen industrialised countries such as Europe and the USA standing out from the rest of the world in term of growth and development, thanks among other things to the appropriation of overseas resources, such as phosphate rocks (Le Noë et al., 2020). This development gap is being filled by most Asian and South American countries, as illustrated in our estimated trends in anthropogenic signature for the specific case of P.

Unexpectedly, the trade of feed and food products had almost no effects on soil P anthropogenic signatures

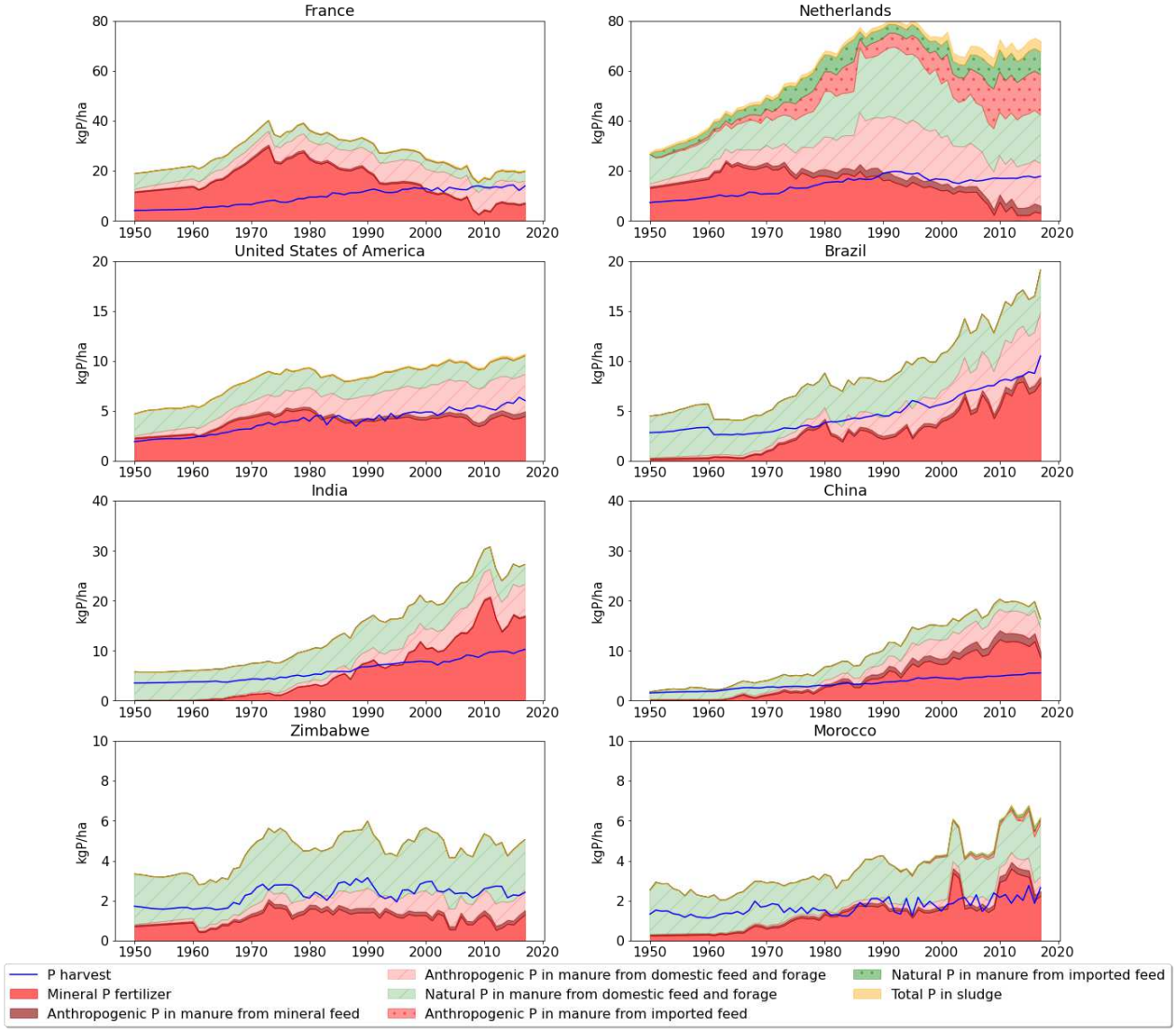

Figure 4 – Temporal evolution of P inputs to agricultural soils over the 1950-2017 period, with distinction between anthropic (in red) and natural (in green) origin of P fluxes. For comparison, P harvested from agricultural soils has also been displayed. For clarity, we did not show the anthropic vs. natural origin of P embedded in sludge (in yellow), although they have been considered in the model. Manure input was split in three categories: (i) manure that came from the consumption of domestically produced feed and forage (ii) manure that came from the consumption of imported feed and forage, and (iii) manure that came from the consumption of mineral P feed. Note that the Y-axis scale differs among countries.

Given that the trade of agricultural products displace annually 3Mt P.yr^{-1}, equivalent to 27% of the P traded through mineral P fertilizers (Nesme et al., 2018), we would have expected trade to substantially modify the soil P anthropogenic signature of large importers such as China or the Netherlands. Yet, we found that the trade of feed and food products had almost no impact on the anthropogenic signatures of the soil P pools. The reasons for that were twofold. First, for 94% of the countries studied, the soil P inputs derived from imported feed and food products (and applied to soils as animal manure and sewage sludge) contributed to less than 5% of the cumulative P inputs over the 1950-2017 period (Figure 4 for all countries except the Netherlands). As a result, for these countries, imported products – whatever their anthropogenic P signature – had almost no effect on soil P anthropogenic signatures. Second, for a handful of countries, despite the P embedded in

imported feed and food products represented an important, indirect P input to their soils, the anthropogenic signature of these imported products was generally close to that of their domestic agricultural soils (Figure S28), thus resulting in neutral effect on soil P anthropogenic signature (Figure S27).

We nevertheless found that, for some countries (such as the Netherlands, Saudi Arabia, Israel, Belgium, Egypt, Lebanon, Portugal and Switzerland), imported feed and food products represented an important input of anthropogenic P to agricultural soils. In these countries, P in animal manure and sewage sludge derived from imported feed and food products accounted for 10% to 34% of their total soil P inputs over the 2007-2017 period, with around half being from anthropic origin. More generally, we found that, at the global scale, 53% of the P embedded in traded products was of anthropic origin; a value amounting to 1.5 Mt $P.yr^{-1}$ or 8% of the global use of mineral P fertilizers (see Methods). Those results illustrate that food and feed trade displaces large amounts of anthropogenic P that need to be accounted for when assessing the reliance of our global food system to phosphate rock resources (Barbieri et al., 2022; Nanda et al., 2019).

Implications for sustainable phosphate rock management

Thanks to our large scale and time-dependant modelling approach, our results helped to picture both the geographical variations and temporal evolutions of the countries' reliance on anthropogenic P. Overall, we found that anthropogenic P has greatly contributed to build the current P fertility of most agricultural soils, the average global anthropogenic signature of agricultural soil available P being around 45%. We also found that the discrepancies in anthropogenic signatures among countries were explained by both an uneven use of mineral P fertilizers since the Second World War and inequalities in the inherited agricultural soil P stocks in 1950. In some cases, the use of mineral P fertilizers has even exacerbated the existing inequalities in soil available P in 1950. For example, Western Europe was the region that has fertilized the most with mineral P over the 1950-2017 period while its soil available P stocks in 1950, inherited from biogeochemical background and past fertilization practices, were also among the highest in the world (Ringeval et al., 2017). Our findings also illustrate that, on the other side of the gradient, many countries – especially in Africa – have very little benefited from phosphate rocks for sustaining their soil P fertility, the anthropogenic signature of their soils remaining below 30%. These results question the past distribution of phosphate rock resources, which has been rather based on regional economic and financial ability to buy fertilizers than on the search to maximise global food production (Langhans et al., 2022).

The high reliance we found of the global soil P fertility to anthropogenic resources raises concerns regarding the vulnerability of our agricultural systems to potential future P scarcity. Our results

229 indeed demonstrate that our agricultural systems have been built on the massive use of mineral P
230 fertilizers. Complemented by proxies on current P fertilizer use as well as on soil P stocks (Barbieri et
231 al., 2022), our result show that world regions are facing differentiated risks of P deficits. For instance,
232 Western Europe has high soil P stocks (Jones et al., 2020; Ringeval et al., 2017) but its high
233 anthropogenic P signature shows its strong reliance on mineral P fertilizers, suggesting some long-
234 term vulnerability (Le Noë et al., 2020). In contrast, countries such as India or Brazil are particularly at
235 risk as they use massive amounts of mineral P fertilizers to maintain high yields – as illustrated by the
236 recent increase in their soil P anthropogenic signatures – but, unlike European countries, their soil
237 available P stocks are small (Ringeval et al., 2017). It is worth noting that the anthropogenic signature
238 we simulated is rather conservative and will decrease only slightly even if mineral P fertilizer use is
239 drastically reduced. This is due to the fact that increasing P recycling on agricultural soils would affect
240 both anthropogenic and natural P fluxes in similar proportion to that of the soil P anthropogenic
241 signature. This makes our anthropogenic metric suitable to estimate the current reliance on
242 anthropogenic P but not appropriate to analyse how soil P fertility would be affected under future
243 farming scenarios.

244 Our work also offers a way to quantify the anthropogenic P content of agricultural food and feed
245 products by considering not only contemporary soil P inputs but also the inherited soil P legacy from
246 past anthropogenic P supply. Most studies estimating the P footprint of crop products have focused
247 on the annual P supply to soils under the considered crop (Barbieri et al., 2022; Lun et al., 2021;
248 Nesme et al., 2018). By labelling, the anthropic vs. natural origin of P in agricultural soils and in
249 agricultural products, our methodology helps to get more accurate estimates of P footprint that
250 account for past mineral P fertilization. Despite negligible effect of trade on soil P anthropogenic
251 signature, our results provide, for the first time, an estimate of the flux of anthropogenic P
252 exchanged between countries through agricultural trade. We found that this flux was equivalent to
253 around 8% of the global use of mineral P fertilizers.

254 To conclude, the quantification of the anthropogenic signature of agricultural soil P shed a new light
255 on our understanding of the Anthropocene, which we analysed through the spectrum of the global P
256 cycle. World regions have unequally benefited from the use of mineral P fertilizers, thus calling for
257 different desirable trajectories for the future. Where the signature is high there is a need to move
258 away from the massive use of anthropogenic P, thus helping to save phosphate rock resources for
259 regions that have little benefitted from these resources so far – and that are often facing high
260 undernourishment issues (Langhans et al., 2022). The distribution of the remaining phosphate rocks
261 reserves should thus be coordinated on a global scale in order to maximise world food production.

Methods

Modelling framework.

Our model aims to simulate P fluxes within the agricultural system of each country, as well as P flows between countries through the trade of agricultural products (Figure 1). As a result, not only did we simulate soil P stocks and dynamics, but also the fluxes of P embedded in crop and livestock products as well as P fluxes in animal manure and sewage sludge.

The P dynamics in agricultural soils (including both cropland and grassland) were simulated by using a two-pool soil P model. Such models have been used in previous large scale studies and proved successful to reproduce observed P harvest over long-term periods (Le Noë et al., 2020; Sattari et al., 2012; Zhang et al., 2017). For the specific purpose of our study, we distinguished the natural vs. anthropic origin of each P flow and stock, following the methodology of a preliminary study conducted at the French scale (Ringeval et al., 2014). Soil P pools were expressed in $kgP.ha^{-1}$, while fluxes were all expressed in $kgP.ha^{-1}.yr^{-1}$. We assumed that for each country the 0-30 cm soil horizon was relevant to study the soil P dynamics following fertilizer inputs, plant uptake by roots and erosion losses.

For each country, we modelled the size of the soil P pools using the Equations below. We distinguished between labile (LP) and stable (SP) P pools and between anthropic (subscript "Ant") or natural (subscript "Nat") origin of P within each pool.

Equation 1a

$$LP_{Nat}(y+1) = LP_{Nat}(y) + \theta[OF_{Nat}(y) + SL_{Nat}(y)] - P_{harvest,Nat}(y) - LO_{LP,Nat}(y) + T_{Nat}(y)$$

Equation 1b

$$LP_{Ant}(y+1) = LP_{Ant}(y) + CF(y) + \theta[OF_{Ant}(y) + SL_{Ant}(y)] - P_{harvest,Ant}(y) - LO_{LP,Ant}(y) + T_{Ant}(y)$$

Equation 1c

$$SP_X(y+1) = SP_X(y) + (1-\theta)[OF_X(y) + SL_X(y)] - LO_{SP,X}(y) - T_X(y) \quad Where\ X \in \{Nat, Ant\}$$

Where *y* refers to the year considered, *X* stands for the sub-scripts *Nat* and *Ant,* and *SL*, *OF*, *CF* and *LO* correspond to soil P inputs as sewage sludge, animal manure, mineral P fertilizer and outputs as losses via erosion to water bodies, respectively. *Θ* refers to the bioavailable fraction of manure and sludge, which was assumed to equal 0.8 based on reported values from the literature (Ringeval et al., 2014; Sattari et al., 2012). *T* represents the net transfer of P from the stable to the labile P pool and captures soil P dynamics such as adsorption/desorption, precipitation/solubilisation and organisation/mineralisation processes. $P_{harvest}$ refers to the P in the plant biomass that is harvested

from both croplands and grasslands, both as grains for animal feed and human food consumption (either domestically consumed or exported to other countries) and as forage.

$P_{Harvest}$. For each country, $P_{harvest,Tot}$ of crop and forage products was annually simulated based on Equation 2.

Equation 2

$$P_{harvest,Tot}(y) = \beta\left(1 - e^{-\gamma.LP_{Tot}(y)}\right)$$

Where β (in kgP.ha^{-1}.yr^{-1}) refers to the maximum attainable yield without any P limitation, and γ (in ha.kgP^{-1}) depicts the ability of crops to extract soil P. $P_{harvest,Tot}$ and LP_{Tot} refer respectively to P exported from agricultural soils either for animal or human consumption (in kgP.ha^{-1}.yr^{-1}) and to the size of the labile P pool (kgP.ha^{-1}). The subscript "Tot" refers to the sum of both anthropogenic (Ant) and natural (Nat) P. A calibration procedure (see below) was performed to determine the values of β and γ for each country.

P transfers from the stable to the labile P pool. The net flux of P from the stable to the labile P pool was also explicitly simulated, as described in Equation 3.

Equation 3

$$T = \mu_{SPtoLP} SP - 0.2\,LP$$

Where μ_{SPtoLP} (in yr^{-1}) represents the percentage of P in the stable P pool that transfers annually to the labile P pool. The inverse of μ_{SPtoLP} can also be interpreted as a mean residence time of P in the stable P pool. For the labile P pool, a turnover rate of 0.2 yr^{-1} - corresponding to a mean residence time of 5 years - was used as proposed by the literature (Sattari et al., 2012). Similarly to β and γ, μ_{SPtoLP} was also calibrated for each country, thus counterbalancing the 0.2 fixed rate of transfer from labile to stable P pool.

Input data. Input data were collected for each individual country. We detail hereafter how data were collected or estimated. *Mineral P fertilizer (CF)* data were collected from FAOSTAT for the 1961-2017 period. Data for the 1950-1960 period were estimated from the spatially explicit dataset of (Lu and Tian, 2016) by aggregating values per country. Because data were only available for the years 1950 and 1960, we performed a linear interpolation to estimate the missing years. *Manure (OF)* data were estimated by considering 10 animal categories (cattle, poultry, asses, mules, goats, sheep, horses, buffaloes, pigs and rabbits) and by multiplying the number of heads (FAOSTAT; Mitchell, 1998a, 1998b, 1993) by species-dependent P excretion rates (Sheldrick et al., 2003). All the manure

324 produced was assumed to reach agricultural soils, either on croplands or on grasslands. *Losses (LO)* of
325 P from soils were calculated by multiplying country-specific percentages of soil lost by erosion (Van
326 Oost et al., 2007) by the here-simulated P pool sizes, by assuming that the rate of losses affected
327 equally stable and labile soil P pools. *Sludge P (SL)* production was estimated by focussing on sewage
328 sludge coming from human food consumption only (thereby excluding P release from detergents).
329 We then estimated the fraction of P in sludge that would reach agricultural soils by multiplying each
330 national human P excretions by the fraction of sewage sludge that is treated and by the P removal
331 efficiency of treatments plants. The methodology and data used were derived from (Van Puijenbroek
332 et al., 2019). We assumed that *Mineral P feed (MF)* represented 5% of the global production of
333 phosphate rock (from FAOSTAT) annually (Cordell and White, 2014; Smil, 2000). The global use of
334 mineral P feed was then allocated to each country based on both their total number of livestock
335 heads and their consumption of mineral P fertilizer (see Section S2.5). *Harvested P* data were used to
336 calibrate our model. P harvest from grasslands was assumed to equal livestock forage P demand at
337 the country scale, assuming no trade of forage between countries. Livestock forage P demand was
338 estimated annually by multiplying livestock number of cattle, sheep and goats by their total energy
339 requirement (Barbieri et al., 2021), by the percentage of forage consumption in their diet (Herrero et
340 al., 2013), and by P concentration of forage. P harvest from croplands was estimated by considering
341 31 crop species (Table S5) and by collecting harvested data from FAOSTAT for the 1961-2017 period.
342 Crop yield data for the 1950-1960 period were estimated by assuming that crop-specific production
343 varied linearly with the country human population as in (Bouwman et al., 2011). Finally, crop P
344 harvest was estimated by multiplying crop yields by the species-dependent P concentration, derived
345 from (Comifer, 2009). The *trade of agricultural products* was considered by including 55 crop
346 products, representing more than 85% of the P traded through food and feed at the global scale
347 (Table S7). From 1986 to 2017, we extracted the data regarding the imported quantities of feed and
348 food products and the countries of origin from the FOATSAT Detailed Trade Matrix. From 1961 to
349 1985, the imported quantities were derived from the Food Balances matrix (FAOSTAT) and the
350 countries of origin were assumed to be roughly the same as those from the 1986-1991 period (see
351 Section S4). For feed, imported quantities and their origin for the 1950-1960 period were estimated
352 based on the information at year 1961 and scaled down based on the livestock density. For food, we
353 assumed that the traded quantities were negligible for the 1950-1960 period. Note that the
354 quantities and origin of imported food and feed products were only used to compute the
355 anthropogenic signatures of manure and sludge, respectively.

356 **Model initialisation.** To initiate the model, we had to determine the size of each soil P pool in 1950.
357 We hypothesised that before 1950 the application of mineral P fertilizer was negligible compared to

the use of other fertilizers, mainly animal manure. As a result, we considered that LP_{Ant} (1949)= SP_{Ant} (1949)=0. In 1950, the LP_{Ant} value was set equal to the inputs of mineral P fertilizer that year. The SP_{Ant} was null in 1950 because, by definition all mineral P fertilizers are applied to the labile P pool. The size of the LP_{Tot} in 1950 was calculated based on the P harvest that year, using Equation 2, possibly leading to significant effects of P harvest in 1950 on the initial soil P pool sizes. The size of LP_{Nat} was then calculated as LP_{Tot} minus LP_{Ant}. Finally, based on equilibrium between the labile and stable P pools we determined SP_{Nat} (Equation 3 with T=0).

Calibration procedure. For each country, an independent calibration procedure was performed to estimate the values of β, γ and μ_{SPtoLP} parameters. The general idea was to test many triplets of parameters, to select the ones that allow the simulations to match P harvest time-series available data (see Section S2.2) and labile P pool size available data in 2005 (see Section S2.7). The adequacy between the simulation outputs and the available data were quantified through calibration scores (Equations 4 and 5). Depending on their calibration scores, some triplets were selected to run the calculations and to compute an uncertainty related to the anthropogenic signatures. Values of calibrated parameters are presented and discussed in Section S6.2.

Calibration started first by choosing the range of values to be tested for each parameter (see Section S6.1). Note that for β, the ranges were country-dependent. Then, from all possible values we produced triplets (β_i, γ_i, μ_i) for which we ran the model and computed score 1 and score 2, as described in Equations (4,5). From 1500 to 8000 triplets were tested for each country.

Equation 4 – Score 1

$$score1 = \frac{1}{\overline{P_{harvest,obs}}} \sqrt{\frac{1}{N} \sum_{i=1950}^{1950+N} (P_{harvest,obs}(i) - P_{harvest,sim}(i))^2}$$

Equation 5 – Score 2

$$score2 = \left| \frac{LP_{obs}(2005) - LP_{sim}(2005)}{LP_{obs}(2005)} \right|$$

Where *N* refers to the number of years studied (N=67), $\overline{P_{harvest,obs}}$ to the mean value of P harvest available data for the time-period studied, $P_{harvest,obs}$ and $P_{harvest,sim}$ to the P harvest time-series available and simulated values, LP_{obs} and LP_{sim} to labile P pool size available and simulated value at year 2005. $P_{harvest,obs}$ was derived from international database (see Equation S3) and $P_{harvest,sim}$ was calculated with Equation 2.

Score 1 calculates the normalized root mean square error (nRMSE) between the simulated total P harvest and the P harvest available data over the 1950-2017 period. Score 2 computes the error between the total labile P pool simulated at year 2005 and the total labile P pool available data extracted either from (Ringeval et al., 2017) or from observations representative to country-scale values for the few countries where observed soil P values were available (see Section S6.3). In the main text, only results from the calibration performed with labile P pool available data from Ringeval et al. 2017 were presented.

Based on these scores, triplets were selected for each country. When possible, all triplets that led to score 1+ score 2 < 30% were selected and the corresponding countries were classified in category 1 (62% of countries). This category thus encompassed countries for which the model managed to reproduce available data on both historical P harvest and the size of the labile P pool in 2005. For some countries, no triplets were able to match the above condition. In this case and when possible we selected the triplets for which score 1 < 30%. The corresponding countries were classified in category 2 (29% of countries). For such countries, the parameters were thus only constrained by P harvest. Finally, when no triplets led to score 1 < 30%, the corresponding countries were classified in category 3 (7% of countries) and not further studied.

In an effort to calibrate the model with observed data on soil available P pool instead of (simulated) available values from (Ringeval et al., 2017), we collected data on soil available P measurements (whatever the chemical extraction used) conducted on agricultural soils (Section S2.7). We found enough data for 24 countries including European countries (Jones et al., 2020), the USA and Canada (IPNI, 2015), Tanzania (Hengl et al., 2017), Botswana and Yemen (Batjes, 2010). For these countries, we found that using the measured data to calibrate the model instead of the simulated ones did not significantly change the anthropogenic signatures of the soil P pools (see Section S6.3).

Data analysis. The outputs of our model were the anthropogenic signatures of the fluxes and soil P pools, as well as the size of the soil P pools. For each output, we computed a mean and a standard deviation value, reflecting uncertainties in the parametrization of the three calibrated parameters. The global and regional averages of the soil P anthropogenic signatures are mean values weighted by the agricultural area of each country. We also chose to present detailed results for eight contrasted countries (France, the Netherlands, the USA, Brazil, India, China, Zimbabwe and Morocco), to better appreciate the temporal evolutions in soil P inputs and in soil P anthropogenic signatures. Following the calibration procedure, these eight countries were all in category 1, which was a guarantee of good quality results. We only displayed results for the anthropogenic signature of the labile P pool, since that of the stable P pool were very similar.

Uncertainties and limitations. The uncertainties associated with our estimate of the anthropogenic signatures of the soil available P are numerous and come from both the model itself (structure and parameter values) and the input data used. Like all models, the one we used is imperfect and cannot represent the dynamics and mechanisms at play in a fully accurate way. However, because of the constraints inherent to global and time-dependant modelling, our model appeared as the most adequate. We yet recognize limitations in using an only two-pool soil P model. Indeed, this model cannot represent the full diversity of P forms and mechanisms at play in soils. More complex mechanistic models exist but they remain hard to parametrize and are still under development (Wang et al., 2022). Our yield response curve to soil P availability is also very simplistic as it considers changes in yields only as a function of changes in soil available P while other factors such as water supply or nitrogen availability should have been considered. Nevertheless, this type of P response curves has been widely used throughout the literature and has repeatedly demonstrated its ability to simulate the temporal evolutions of P harvest over time (Sattari et al., 2012; Zhang et al., 2017). In addition, a special feature of our work has been to calibrate β, γ and μ_{SPtoLP} for each country (see Section S6.1), thereby helping to capture country-specific conditions. The calibration of β (referring to the maximum attainable yield without any P limitation) allowed us to take into account other limiting factors that would be country specific. The calibration of μ_{SPtoLP} made it possible to take into account the soil-type specificities (even if a calibration at the country level is questionable because of the diversity of soil types within each country).

Regarding input data to the model, the uncertainties were also potentially large, although difficult to quantify. In particular, we recognise large uncertainties in our estimates of P harvest from grasslands, especially in some countries such as India (Fetzel et al., 2017). We explicitly chose to base our estimates of P harvest from grasslands on forage demand and not on forage production given the large uncertainty in Net Primary Productivity of grassland (Sun et al., 2021). Data on the trade of agricultural products were used to determine the anthropogenic signature of animal manure and sewage sludge, when animals and humans consumed imported products (see Section S3 and S4). For several reasons, we underestimated the global P flows that occurred through the trade of agricultural products. Indeed, we estimated that around 1MtP.yr^{-1} embedded in food and feed products were traded at the global scale, while other studies reported values around 3MtP.yr^{-1} (Nesme et al., 2018). Our underestimation of these flows comes from (i) the fact that we excluded several countries from our analysis (see above about the calibration procedure) and (ii) some revisions of data from the Detailed Trade Matrix – because those data did not always match with those from the Food Balances, we revised them downwards, notably about trade in soybean cakes. However, the impact of these errors on the anthropogenic signatures of the different countries are

likely to be small, given that the anthropogenic signatures are more sensitive to the signature of imported products rather than on their quantities. Finally, we hypothesized that all the animal manure produced was available for agricultural soil application, which is highly questionable. Other studies have used global average but those were too uncertain to be included in our estimate (Chen and Graedel, 2016). However, because manure application on agricultural soils is basically P recycling that conserves anthropogenic signature, the possible overestimation of manure application in our approach is likely to have small effects on the estimated anthropogenic soil P signatures.

For some countries, we recognise that we did not find any triplet of parameters that allowed the model to replicate the historical available data on soil P harvest. As a result, these countries (category 3) were excluded from our analyses. Conversely, the results we obtained for countries in category 1 were the most reliable. The countries in category 2 were not able to match the constraints on both P harvest and labile P pool size in 2005. For these countries, mainly located in Africa, we recognise strong uncertainties in our estimates of the size of soil P pools and thus in the estimated anthropogenic signatures. One of the reasons for the difficulties to match the two constraints comes from the methodology we used to estimate the size of the soil labile P pool in 1950. In our study, the latter was estimated from data on P harvest in 1950, while initial size of labile P pool in (Ringeval et al., 2017) – used to constraint the size of the labile P pools in our model- were derived from a dataset about soil biogeochemical background (Yang et al. 2013). However, although our uncertainties on the size of the stable and labile soil P pools are large (see Section S9), our final uncertainties on anthropogenic signatures remain reasonable (Figure S14). The difficulties in providing accurate estimates of soil P content at large spatial scales is often raised in the literature (Das et al. 2019), even though more and more studies are tackling this issue (He et al., 2021; Hengl et al., 2021; Ringeval et al., 2017).

Another limitation comes from neglecting mineral P fertilizer use before 1950. Although mineral P fertilizer use was small for most countries in 1950 (Bouwman et al., 2011), several European countries had already high fertilisation rates at that period. We nevertheless chose to start the simulations in 1950 because of the critical lack of accurate, global data before that date. A sensitivity test performed in Ringeval et al. (2014) – on which our methodology is based – has shown that our estimates are likely to remain robust. Running the model for 30 years with soil inputs and outputs related to the 1st year (~1950) before the transient run showed only a ~10% underestimation of the anthropogenic signature of agricultural soils.

Finally, our definition of anthropogenic P – which referred to the use of mineral P fertilizers and mineral P feed derived from the extraction of phosphate rocks – is questionable. Indeed, other

anthropogenic practices such as animal manure application, deforestation and P fertility transfers from grasslands to croplands have also modified the global P cycle. However, in a context of increasing scarcity of phosphate rock reserves, we found it more relevant to quantify the modification of the P cycle related solely to the use of this critical and highly processed resource.

Code and data availability

Data are available in the Supplementary Information file. The python code used for computing all calculations is available at https://data.inrae.fr/privateurl.xhtml?token=4ddb8501-c41d-4ad6-8a09-5c1ebb8289f9.

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Competing interests

The authors declare no competing interests.

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