

Spatiotemporal dynamics of soil phosphorus and crop uptake in global cropland during the twentieth century

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Abstract. Phosphorus (P) plays a vital role in global crop production and food security. In this study, we
15 investigate the changes in soil P pool inventories calibrated from historical countrywide crop P uptake, using a 0.5 by 0.5 degree spatially explicit model for the period 1900-2010. Globally, the total P pool per hectare increased rapidly between 1900 and 2010 in soils of Europe (+31%), South America (+2%), North America (+15%), Asia (+17%) and Oceania (+17%), while it has been stable in Africa. Simulated crop P
20 uptake is influenced by both soil properties (available P and the P retention potential) and crop characteristics (maximum uptake). Until 1950, P fertilizer application had a negligible influence on crop uptake, but recently it has become a driving factor for food production in industrialized countries and a number of transition countries like Brazil, Korea and China. This comprehensive and spatially explicit model can be used to assess how long surplus P-fertilization is needed or how long depletions of built-up surplus P can continue without affecting crop yield.

25 **Keywords:** cropland; dynamic; fertilizer; global; manure; modelling; phosphorus; soil reserves

1. Introduction

The current world population of 7.3 billion is expected to reach 9.7 billion in 2050 (UN, 2016), likely triggering a greater demand for food and resources. Moreover, increasing prosperity will lead to further shifts in human diets towards more meat and milk consumption, particularly in developing countries
30 (Alexandratos and Bruinsma, 2012). Livestock products require more nutrients for production compared to crops and thus may induce additional nutrient demands (Bouwman et al., 2013).

Phosphorus (P) is one of the major limiting nutrients in agriculture (Koning et al., 2008), which unlike nitrogen, cannot be fixed from the atmosphere by living organisms or industrial processes. In early
35 agricultural systems, P was supplied to soils by recycling animal manure, crushed animal bones, human excreta, city waste and ash (Beaton, 2006). Since the industrial revolution, however, soil P enrichment has been increasingly dominated by non-renewable P resources such as guano (accumulated seabird droppings) and phosphate rock. Presently, phosphate rock provides the P for producing 90% of global P
40 fertilizer use, which was 18.8 Tg P (Tg = teragram; 1 Tg = 10^{12} g = 1 million metric tons) in 2010 (PotashCorp, 2016). Yet, this resource is rapidly dwindling with estimates for phosphate rock availability ranging from peak P production in 2033 with subsequent rapid decline (Cordell et al., 2009) to half of the resources being used by 2100 (Van Vuuren et al., 2010) or complete exhaustion within the next 300-400 years (Van Kauwenbergh, 2010).

45 The availability of P for plant roots is determined by the concentration of phosphate ions in the soil solution and the ability of the soil to replenish them after plant uptake (Syers et al., 2008). The replenishing of phosphate ions depends on soil characteristics such as mineralogy, soil reaction and degree of weathering (Fairhurst et al., 1999). Soil materials rich in soluble alumina or iron, a high calcium activity, or clay minerals like kaolinite, react with P to form insoluble compounds inaccessible to plant
50 roots (Brady, 1990). Any surplus P application over crop uptake and losses by runoff and erosion accumulates in the soil as "residual P" which can be available for crop uptake for many years depending on the soil characteristics and management (Syers et al., 2008; Batjes, 2011). As long as adequate P is present in the readily available pools to maintain a critical threshold P concentration in the soil solution,

55 good crop yields can be maintained (Syers et al., 2008). Annual P inputs from fertilizer should compensate plant P uptake and erosion loss. However, when the amount of readily available P is below this critical threshold level, the rate of P release from residual P is insufficient to sustain optimal crop yields. Improving our mechanistic understanding of soil P dynamics locally and globally is important for evaluating broad-scale food security and improving sustainable P management.

60 This paper presents a global, spatially explicit analysis of soil P dynamics in global crop production systems using a soil model with two P pools (Wolf et al., 1987; Janssen et al., 1987), which was later presented as the "Dynamic Phosphorus Pool Simulator" (DPPS) (Sattari et al., 2012). DPPS describes the impact of long term P application on the transfers between a stable and a labile soil P pool as well as crop P uptake, accounting for weathering, deposition and erosion (Figure 1). The original model was developed
65 for the field scale (Wolf et al., 1987; Janssen et al., 1987) and has recently been applied to simulate the impact of soil residual P on crop uptake and future demand for fertilizer P on the continental scale (Sattari et al., 2012). Compared with previous studies, our model has been improved to include spatially explicit calculations, land use change, dynamic transfer between different soil P pools, initialization of the P pools with global observations, dynamic P loss by runoff, dynamic calculation of crop P uptake and time-variant
70 maximum uptake parameter (Table 1). Since build-up of residual P is a local process but with global consequences, the aim of this paper is to ultimately simulate the soil P dynamics, soil P fertility and crop P uptake for global cropland from the beginning of the 20th century up until the year 2010 at a 0.5 x 0.5 degree spatial scale.

2. Material and methods

75 2.1. Model description

The spatially explicit DDPS model presented here describes the P dynamics in croplands as a function of two soil P pools, a labile soil P pool (LP , kg P ha^{-1}) and a stable soil P pool (SP , kg P ha^{-1}) with an annual temporal scale and a spatial resolution of 0.5 by 0.5 degree (Figure 1). LP represents all forms of P that can directly be taken up by plant roots, comprising both organic and inorganic P; SP represents forms of

80 P bound to soil minerals and organic matter that are not directly available to plants (Table 2). P is transferred in both directions between the two pools.

Our model considers natural P inputs to the soil, i.e. weathering (*weathering*, kg P ha⁻¹yr⁻¹), litter (*litter*, kg P ha⁻¹yr⁻¹) and atmospheric deposition (*deposition*, kg P ha⁻¹yr⁻¹); and anthropogenic P inputs, including application of mineral P fertilizer (*fertilizer*, kg P ha⁻¹yr⁻¹) and animal manure (*manure*, kg P ha⁻¹yr⁻¹). P outflows from the soil system include the withdrawal of P in harvested crops (*uptake*, kg P ha⁻¹yr⁻¹) and runoff (*runoff*, kg P ha⁻¹yr⁻¹) (Figure 1; Table 2).

The thickness of the topsoil is assumed constant at 30 cm; soil loss by runoff is replaced at the bottom of the topsoil by fresh subsoil material from below 30 cm (*fresh_soil*, kg P ha⁻¹yr⁻¹). The proportion of *LP* and *SP* in *fresh_soil* is based on the pools according to the soil P inventory depicted in Yang et al. (2010). The P input from *litter* (kg P ha⁻¹yr⁻¹) remaining after crop harvest is returned to the soil and becomes part of the *LP* pool. For natural ecosystems, assuming there is no anthropogenic fertilizer application, the uptake equals the inputs from *litter*, *deposition* and *weathering*. In contrast, P *uptake* in agricultural soils is calculated explicitly (see below).

P from atmospheric deposition obtained from the Model of Atmospheric Transport and Chemistry (MATCH) is assumed to be a direct input for the SP pool only, since mineral aerosols (dust) are the dominant source of atmospheric P (~ 82%) (Mahowald et al., 2008) and are not readily available for plant uptake. P from weathering in global cropland is assumed to amount to 1.6 Tg yr⁻¹ (Liu et al., 2008). This value is used to calculate the weathering fraction of the soil P in apatite (*fr_weathering* = 0.001747) from the global natural soil P inventory by Yang et al. (2013), and assumed to be available directly for plant uptake.

105 The global spatially explicit P runoff is calculated following the approach of Beusen et al. (2015) who distinguished losses from recent nutrient applications in the form of fertilizer, manure or organic matter (Hart et al., 2004), and a “memory” effect related to long-term historical changes in soil nutrient

inventories (McDowell and Sharpley, 2001; Tarkalson and Mikkelsen, 2004). The memory effect is based on Cerdan et al. (2010) using slope, soil texture and land cover type to estimate country aggregated soil-loss rates for arable land, grassland and natural vegetation. The P content of the soil loss (*runoff_LP* and *runoff_SP*) is based on the *LP* and *SP* of the soil in the grid cell considered at that moment in time.

The yearly changes of the *LP* and *SP* pools (kg P ha⁻¹yr⁻¹) are calculated as follows:

$$\frac{\partial_{LP}}{\partial_t} = F_{sp2lp} - F_{lp2sp} + weathering + fertilizer + manure + freshsoil_{LP} + litter - runoff_LP - uptake \quad (1)$$

$$\frac{\partial_{SP}}{\partial_t} = F_{lp2sp} - F_{sp2lp} + deposition + freshsoil_{SP} - runoff_SP \quad (2)$$

Where *F_{lp2sp}* and *F_{sp2lp}* (kg P ha⁻¹yr⁻¹) are the fluxes between *LP* and *SP* and *SP* to *LP*, respectively:

$$F_{lp2sp} = \frac{LP}{\mu_{LS}} \quad (3)$$

$$F_{sp2lp} = \frac{SP}{\mu_{SL}} \quad (4)$$

where the variables μ_{LS} and μ_{SL} are transfer times (years) between *LP* to *SP* and *SP* to *LP*, respectively. The transfer time μ_{SL} from *LP* to *SP* is set to 5 years based on the original DPPS (Janssen et al., 1987). The transfer time from *SP* to *LP* (μ_{SL}) is calculated for every grid cell based on the mass balance of *LP* for natural ecosystems using Equation (1) and assuming steady state:

$$\mu_{SL} = \frac{SP}{LP \times \frac{1}{\mu_{LS}} + deposition} \quad (5)$$

We selected the mass balance of *LP* to calculate μ_{SL} (instead of that for *SP*) because it yields a value that matches the value obtained by Wolf et al. (1987) based on experimental data.

130 The amount of P that can be accessed by plant roots (*availableP*, kg P ha⁻¹yr⁻¹) is calculated as a fraction of *LP* (*fr_mobile* × *LP*). The P uptake by crops was calculated using Michaelis-Menten kinetics (Michaelis and Menten, 1913; for elaboration for nutrient uptake see Nijland et al., 2008) as follows:

$$uptake = \frac{max_uptake \times availableP}{\left(\frac{c \times max_uptake}{init_recovery} + availableP\right)} \quad (6)$$

135 Where *max_uptake* (kg P ha⁻¹yr⁻¹) is the maximum P uptake, and *init_recovery* is the initial recovery fraction (no dimension), which is the initial slope of the P response curve presented (Batjes, 2011) for all soil types distinguished in the legend of the FAO-Unesco soil map of the world (FAO-Unesco, 1974); *c* is a constant to obtain the *availableP* for which uptake is 0.5 times *max_uptake* (no dimension; *c* = 0.5). The calculation of the area-weighted value of *init_recovery* for each grid cell is based on the sub-grid distribution of soil classes.

140 *Max_uptake* is a time dependent variable, considering the development of technology in crop production. A first condition is that *max_uptake* never decreases. A second condition is that *max_uptake* has a maximum value of 100 kg P ha⁻¹ yr⁻¹, which exceeds the present-day highest value for all countries of the world based on our data (section 2.2). We make a split in 1961, the year in which the FAO data series start (FAO, 2016a, b). Prior to 1961 *max_uptake* is the minimum of twice the uptake calculated from crop
145 production data for 1960, and the sum of P inputs from fertilizers and manure; from 1961 onwards, it is the uptake calculated from crop production data times a factor of 2, or the previous *max_uptake* if that is larger.

150 In grid cells where cropland expansion takes place, the initial conditions of virgin soil (without fertilizer history) are assigned to the new, additional area in the year considered. For land abandonment (arable land to natural land) we assume that it takes 30 years for abandoned land to revert to natural conditions, and in this period the P in *litter* increases and *uptake* decreases linearly with time from zero to the natural flux.

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2.2. Data used

We use the spatially explicit data on P fertilizer use and animal manure spreading, land use, crop production and cropland areas from the database used in the IMAGE-Global Nutrient Model (Beusen et al., 2016). In this database, for the period 1900-1960, the gridded inventories of P inputs from a recent
160 study (Bouwman et al., 2013) were used for the calculation of the soil P pools and crop uptake. This dataset was based on various sources for land use (Klein Goldewijk et al., 2011; Klein Goldewijk et al., 2010), livestock for 1900-1960 (Mitchell, 1998, 1993a, b), animal and crop production and fertilizer use for 1930-1950 (FAO, 1951) and fertilizer use prior to 1930 (Cressy Morrison, 1937), which were spatially distributed with IMAGE-GNM as described by Bouwman et al. (2013). Due to the lack of data during
165 this time period, uptake estimates are more uncertain than those for the years since 1961 to 2010 based on FAO statistics (see below).

Crop P uptake and fertilizer inputs per hectare are spatially homogeneous for all grid cells with cropland within each country. The total production of P from animal manure was estimated from country livestock
170 data for non-dairy and dairy cattle, pigs, poultry, sheep and goats, and the P content in the manure (Table S2). The amount of manure available for spreading in agricultural land excludes droppings in grassland, manure used as fuel or building material or manure otherwise ending outside the agricultural system (e.g. in lagoons) (Beusen et al., 2016).

175 IMAGE-GNM distinguishes two crop systems, i.e. (1) crops in mixed systems, in which there is a linkage between crop and livestock production through manure (from animals to crops) and feed (from crops to animals) exchanges, and (2) crops in pastoral systems, in which crop and livestock production are separate systems. While P fertilizers are the same in all cropland within a country, P inputs from animal manure are different in mixed and pastoral systems.

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For the period 1960 to 2010, simulated crop P uptake is validated against P in harvested crop production data covering all (>120) annual and perennial food, feed and fodder crops and fruits from FAO statistics (FAO, 2016a, b) and are aggregated into 34 crop groups distinguished in recent FAO studies

(Alexandratos and Bruinsma, 2012). The historical P uptake per hectare is calculated from the crop yields from the above-mentioned FAO statistics for 34 crop groups and P content for each crop group, as listed in Table S3.

We present the data and results for a number of individual countries (China, United States, South Africa and France), and world regions. The definition of the regions is provided in Table S1.

190 **2.3. Initialization and calibration**

To initialize the model in 1900, we assume that the P pools are in equilibrium. We furthermore assume that the sum of *LP* and *SP* equals the total P (*TP*) in natural systems proposed by Yang et al. (2013), representing the pre-industrial conditions. Model simulations are run until 2010 for the entire globe using the data stipulated in Section 2.2. The model is then calibrated by varying *fr_mobile* to achieve a good fit with the crop uptake data for each country using an iterative procedure to minimize the difference between model result and the uptake based on crop production data (see 2.2) by modifying *fr_mobile* (see equation 6). First, the country-specific error (kg P ha⁻¹yr⁻¹) in the modelled P uptake is calculated:

$$error = \frac{\sum_1^n (uptake_{model} - uptake_{data})}{n} \quad (7)$$

where *n* is the number of data points between 1960 and 2010. We compare every second year, so *n* = 26. *fr_mobile* is limited to the range [0.01, 0.7]. The calibration is considered successful when the absolute value of the error is less than 0.4. For countries where the calibration is not successful, we apply the regional (Table S1) average area-weighted *fr_mobile*. Once the optimal *fr_mobile* has been estimated, we use the normalized root mean square error (*NRMSE*) to compare simulated and measured P uptake (see S1).

205 **2.4. Sensitivity analysis**

The model sensitivity is investigated using Latin Hypercube Sampling (LHS), with uncertainty ranges for 12 model parameters (Table 3) and expressed as the standardized regression coefficient (*SRC*), to show the influence of model parameters to model output. We focused on the sensitivity of model parameters to

210 modelled *LP* and *SP* size, and crop P uptake for two years (1950 and 2000). A detailed description of the approach for the sensitivity analysis and the results is in Section S2 in the Supplement (S2).

3. Results

3.1. P inputs and uptake

215 The global P inputs (including mineral fertilizer and manure) have increased from 2.0 Tg P in 1900 to 23.0 Tg P in 2010 with large variations across different regions and countries of the world. Between 1900 and 2010, the global use of mineral fertilizer in croplands increased from 0.4 Tg to 15.8 Tg yr⁻¹ and the use of manure from 1.6 Tg to 7.2 Tg yr⁻¹. At the global scale, about half of the applied P in 2010 was taken up by harvested crops (modelled 12 Tg P yr⁻¹, 7.3 kg P ha⁻¹yr⁻¹; data 13.8 Tg P yr⁻¹, 8 kg P ha⁻¹yr⁻¹). All world regions and countries show similar patterns before 1950, i.e. very low P input levels and crop P uptakes that do not differ much from the inputs.

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The annual P inputs in Western Europe increased from 0.4 Tg P in 1900 to 2 Tg P in 1980 and then decreased to 0.9 Tg P in 2010, which translates into rates of 24 kg P ha⁻¹yr⁻¹ in 1980, followed by a gradual decrease to 11 kg P ha⁻¹yr⁻¹ in 2010 (Figure 2a). Crop uptake started to rapidly increase in Western Europe from 4 kg P ha⁻¹yr⁻¹ in 1950 to 13 kg P ha⁻¹yr⁻¹ in 2010; in 2010, the crop P uptake exceeded the P fertilizer and manure application (Figure 2a), and two levels of crop uptake have been achieved in different years at the same application rate (Figure 4a and 4j, Western Europe and France). This indicates that since the 1980s, P application has been reduced while uptake continues to increase due to the supply of residual soil P (Figure 5a). The model results also show this hysteresis, although DPPS underestimates the P uptake in the most recent years (Figure 2a).

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Agriculture in Western Europe is much more intensive than in the United States (Van Grinsven et al., 2015). P inputs in North America peaked in 1980 with a total of 2.9 Tg yr⁻¹. Application rates increased from 1 kg P ha⁻¹ in 1900 to 12 kg P ha⁻¹yr⁻¹ in 1980 and 10 kg P ha⁻¹yr⁻¹ in 2010, and crop P uptake lagged behind P inputs for around 20 years but inputs and uptake are now at a similar level (Figure 5b). The trend

235 is visible in United States data, and DPPS results agree well with the uptake based on production data in both cases (Figure 2h and 2b).

In contrast to the other continents, the input level in Africa is much lower, with annual application rates ranging from 1 kg P ha⁻¹yr⁻¹ in 1900 to 5 kg P ha⁻¹yr⁻¹ in 2010. Along with the slowly increasing application rates, African crop uptake is also increasing at a low rate with no accumulation of soil P (Figure 5c). DPPS results are in good agreement with the production-based uptake for Africa (Figure 2c and 3c).

The annual P application in Asia increased more than 12-fold from 0.9 Tg yr⁻¹ in 1900 to 12.4 Tg yr⁻¹ in 2010, and manure P more than doubled from 0.8 Tg yr⁻¹ to 3.5 Tg yr⁻¹ from 1960 to 2010. The P application rates of Asia rose dramatically from low values in 1900 to 22 kg P ha⁻¹ yr⁻¹ in 2010. Asia is currently in the phase of rapidly increasing inputs, with crop P uptake also increasing but at a slower rate (Figure 2d). DPPS results are in good agreement with the uptake data for this continent (Figure 2d and 3d).

250 In South America, the annual P inputs increased from 5 to 23 kg P ha⁻¹yr⁻¹ between 1900 and 2010 and the annual P uptake also increased from 4 to 13 kg P ha⁻¹yr⁻¹ for the same period of time. Although DPPS underestimates the P uptake in recent years, the simulations agree with crop production data for most years (Figure 2e and 3e).

255 In Oceania, annual P application varied between 10 and 13 kg P ha⁻¹yr⁻¹ in recent decades. Oceania shows low uptake rates relative to inputs over the whole period 1900-2010, and the simulated results are in good agreement with the data (Figure 2f and 3f). This indicates that the cumulative inputs of P fertilizer and manure exceed the crop P uptake.

260 The DPPS model adequately simulates P uptake by crops in different regions and countries (Figure 2). The modelled P uptake values match well the historical records (Figure 3) as shown by the *NRMSE* of 19% (26 regions, 26 year, 676 points). At the country scale, simulations and observations of annual P

uptake for every second year (1960-2010) agree well ($NRMSE = 45\%$, 201 countries, 26 years, 5226 points). The $NRMSE$ of the country-level simulation exceeds that of the aggregated results for world regions, which is caused by the underestimation of peak values by DPPS. However, for the global and regional scale we consider the DPPS results acceptable ($< 50\%$ for global scale).

3.3. Soil P budgets

The soil P budget is defined as the biogeochemical soil P budget, which is the difference between soil P inputs (mineral fertilizer, manure and deposition) and outputs (uptake and runoff) (Figure 5 and 6a).

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The soil budgets (Figure 5) show the difference between the industrialized countries that are currently in an equilibrium or depletion phase with regard to soil P, transition countries that are in an accumulation phase (e.g. China, India and Brazil) and countries with a low input and productivity crop production system (e.g. Africa). The soil budgets also show how important runoff losses are for the budgets and for determining whether P is accumulating (Figure 6a).

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We also calculated the agronomic soil P budget, which is the difference between crop P uptake and inputs (the sum of fertilizer and manure) (Figure S1c). The crop P uptake and soil P budget have a large spatial heterogeneity. China, India, and Brazil are among the countries with the highest P input rates in 2010 (Figure S1a). China, USA, Eastern Africa and Western Europe are the areas with highest P uptake (Figure S1b). However, South Africa (like Brazil), China, India and central America are regions with large P surpluses. Western Europe, Western Russia and Western Africa currently show P deficits (Figure 6a).

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3.4. Soil P pools

The spatial distribution of the changes in the LP , SP and TP ($TP = LP + SP$) pools were generated yearly from 1900 until 2010. In the initial condition (1900), soils in wet tropical climates generally had low P contents as a result of prolonged weathering (Yang and Post, 2011) (Figure S2). Nevertheless, with the advent of intensive agricultural activities during the 20th century and up until 2010, TP has drastically

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increased along the western coast of the Americas, central and western India, southwest part of Saudi Arabia and Ethiopia (Figure 6c).

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The total P content per hectare increased rapidly between 1900 and 2010 in soils of Europe (31% relative to 1900), North America (15%), Asia (17%) and Oceania (17%), with a small increase in South America (2%), while total P content has been stable in Africa (Figure 7c). This increase primarily occurred in the decades after 1950.

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The labile P content per hectare of Western Europe was relatively stable from 1900 to 1930 (260 to 272 kg P ha⁻¹), then increased from 1940 to 1970 (314 to 370 kg P ha⁻¹) and decreased from 1980 to 2010 (387 to 350 kg P ha⁻¹), due to the decreasing P fertilizer application rates in recent years. *LP* in Asia and South America has been increasing, especially since 1980. In Africa, *LP* has remained consistently stable, while more recently it has become stable in Oceania and North America (Figure 7a).

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It is apparent that some regions had a more favourable starting position in terms of soil P fertility in 1900. For example, soil P contents (both total and labile P) in Western Europe were much higher than in all the other world regions. With the large surpluses in the period 1960-1980 the soil P reserves in Western Europe even increased, and started to decrease in the last decades to a level almost equal to that in China. However, China started from much lower soil P levels, and has been accumulating rapidly in a relatively short time period. Other regions like North America started at lower values than Western Europe, and surpluses have not been as large as in Western Europe, and the soil labile and total P slowly increased. This may explain why in North America, P input rates have decreased but to levels that are higher than those in Western Europe.

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4. Discussion

Sattari et al. (2012) applied the DPPS model to reproduce historical continental crop P uptake (1965-2007) and estimate P requirements for crop production in 2050 using fixed μ_{LS} and μ_{SL} . Our longer simulation time (1900-2010), as well as spatially explicit calculations (0.5 by 0.5 degree resolution)

315 allows assessing long-term changes in soil P pools taking into account local heterogeneity (Table 1).
Further improvements include the consideration of yearly land use changes, spatially explicit soil
properties and P contents, erosion and runoff, the dynamic calculation of transfer time between the soil P
pools and initialization of different soil P pools with global data. A final improvement is the calculation
of crop uptake using Michaelis-Menten kinetics, whereby the maximum uptake development reflects
320 technology changes such as improved crop varieties (Table 1).

The reason for P uptake underestimation in regions like Western Europe in recent years is that the model
is calibrated using *fr_mobile* over the whole period 1961-2010. Hence, the results for the middle of this
period are very close to observed uptake values, but in the early and late part of the period 1961-2010,
325 the model may deviate from the observations. We have deliberately chosen for a fixed value of *fr_mobile*.
In theory, *fr_mobile* could be estimated by using the characteristics of the different fertilizers used, but
such data is unavailable at our gridded scale. Furthermore, estimating a time-varying parameter describing
P availability would be extremely complex and falls outside the scope of this study.

330 The global and country-specific results for P cycling in croplands of our study agree well with estimates
from recent literature (Table 5). Large P surpluses in China, India, Western Europe in our study agree
with MacDonald et al. (2011), as well as P deficits in Argentina, the central parts of the United States and
Eastern Europe (especially Kazakhstan). For most provinces in China, especially in eastern China,
estimated P surpluses of $> 10 \text{ kg P ha}^{-1}\text{yr}^{-1}$ are similar to both Shen et al. (2011) and MacDonald et al.
335 (2011), who reported $> 10 \text{ kg P ha}^{-1}\text{yr}^{-1}$ and $> 13 \text{ kg P ha}^{-1}\text{yr}^{-1}$, respectively (Table 5). Unfortunately, it is
not possible to compare our results for the changes in soil P pools, since all the studies presented in Table
5 have no inventories of changes in soil P reserves.

In the early 20th century, the P uptake rates are only marginally smaller than the low P input rates,
340 implying that P was balanced in crop production systems although producing low yields. P inputs have
steadily increased globally since the 1950s, allowing for an increase in crop production, but ultimately
leading to a decrease in the efficiency of P application and an increase in the soil P reserves. Both our

study and those of Sattari et al. (2012; 2016) show that since 1980s, P application rates have been reduced in much of Europe while uptake continued to increase due to the supply of residual soil P.

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Over the last couple of decades, P management practices has varied widely throughout the globe, leading to different P input rates, P pool sizes, and crop uptake rates (Figure 6,7) For example, agricultural lands in Africa have increased by more than 40% in the past 4 decades, while both the labile and stable P pools have slightly decreased due to negative budgets. This indicates that in Africa there has been a net soil P depletion in cropland, which may be caused in spite of the expansion of arable land due to the low P application rates.

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In contrast, P surpluses in industrialized countries have been decreasing steadily from high values in the 1970s and 1980s towards a small deficit in recent years; a hysteresis effect of uptake is observed, with equal inputs resulting in low (1970s) and high (2010) P uptake, due to a gradual increase of labile pool over the whole period.

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The large P surpluses in the industrialized countries in the 1970s and 1980s represent a legacy for future productive capacity. The present situation in many industrialized countries shows that the P fertilizer inputs and P surpluses can be reduced considerably and the PUE can be increased to high levels without a yield penalty. This phenomenon of the legacy of residual soil P due to large P surpluses, such as during the 1970s and 1980s in Western and Eastern Europe, the Russian Federation and the USA, has also been recognized before (Sattari et al., 2012). The data for the industrialized countries show that this soil P depletion can continue for many years without yield declines. However, the inputs have been reduced to low levels and surpluses turned into deficits, and the current model can eventually be used to investigate how long this depletion can continue without a yield penalty by P limitation.

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China and India have shown large surpluses of P in particular since the 1990s, and are currently building up large soil P reserves, but heterogeneously distributed with negative budgets in some parts. In 2010, the residual soil P in China and India is 18 and 17 kg P ha⁻¹ yr⁻¹, respectively, compared to 1 kg P ha⁻¹ yr⁻¹ in

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the USA. The total accumulated residual P per hectare of cropland in China and India exceeds that in Western Europe, Russian Federation and the USA, suggesting that it may be possible to reduce P application rates without affecting crop uptake.

375 There are many factors that drive the spatiotemporal distribution of soil phosphorus in global cropland, including the natural soil biogeochemical background, soil properties, farming practices (mainly the increasing input of anthropogenic P fertilizer application), land use and land cover change (arable land expansion or abandonment and changes in crop species composition), P losses through soil erosion, soil temperature and soil water content, soil microbial ecosystem and crop production. Ringeval et al. (2017) 380 qualified the contribution of these different factors to the global soil P distribution and found that soil biogeochemical background and farming practices are the most important drivers for P uptake. This agrees with our focus on these factors as the biggest controllers for P distribution in cropland soils. The P content of soils under natural vegetation and the CaCO₃ content, pH, clay, moisture and water content, determines the capacity for P sorption, retention, and soil weathering. For example, in arid and semi-arid 385 regions the low soil water content hinders P diffusion and plant root growth, ultimately limiting crop P uptake. While this chemical background is highly heterogeneous, our gridded approach and model initialization capture some of these differences in global soils by distributing LP and SP according to natural soils of Yang et al. (2013). Farming practices, especially dramatically increasing P fertilizer and manure application rates have an important impact on the spatiotemporal changes of soil P and crop 390 uptake (Figure 2 and 7)

The contribution of expanding cropland areas and crop yields (P uptake per hectare) to the production increase since 1960 is about 18% for the former and 82% for the latter. Nevertheless, there are likely regional differences with developing countries having a greater proportion of agricultural areal expansion 395 and industrialized countries more improvements in nutrient use efficiency (Figure 4). This explains why for example in Africa, the labile and total P pools per hectare have been relatively constant, while uptake has exceeded inputs during the whole period 1900-2010.

5. Concluding remarks

This study presents a spatially explicit model-based inventory of global soil P stocks and crop uptake for the period of 1900-2010, which represents the years in the Anthropocene when human activities accelerated the global agricultural P cycle by more than a factor of 10. The spatially explicit DPPS model enables to match local soil P pools with crop P uptake for the past century with a high resolution (0.5 by 0.5 degree) while accounting for finer scale heterogeneity by including within grid cell land use change. Compared with previous non-spatial DPPS application by Sattari et al. (2012), our research has improved DPPS in calculating the spatial variability of the soil P pool changes within countries by accounting for soil characteristics and dynamic parameters, calculating the process-based uptake rather than fixed value, considering the yearly land use change, initializing different soil P pools with global data and modelling P losses by runoff instead of fixed numbers.

Global P inputs (including mineral fertilizer and manure) have increased from 2.0 Tg P yr⁻¹ in 1900 to 23.0 Tg P yr⁻¹ in 2010 with large variation across different regions and countries of the world. At the global scale, about half of the applied P in 2010 was taken up by harvested crops (modelled annual rates of 12 Tg P yr⁻¹, 7.3 kg P ha⁻¹; data 13.8 Tg P yr⁻¹, 8 kg P ha⁻¹). All world regions and countries show similar patterns before 1950, i.e. very low P input levels and crop P uptake that are not very much different from inputs. However, after 1950, regions and countries show different soil P inputs, crop P uptake, soil P budgets and pool changes, which indicate the spatial and temporal variability of soil P condition.

According to the model sensitivity analysis, the maximum uptake level (*max_uptake*, reflecting the technology level), the initial labile pool size (*LP_yang*), the calibration coefficient which mimics the direct P availability within the labile pool (*fr_mobile*) and the slope of the P input response curve at the origin (*init_recovery*) have an important influence on crop P uptake. This means that the simulated crop P uptake is influenced by both soil properties and crop characteristics.

The DPPS model can be used to assess the long-term changes in the soil P status, an important indicator of soil fertility, future soil productivity and food security. Via its gridded approach, DPPS can ultimately

help to improve our mechanistic understanding of P cycle in global cropland at local and global scale and our ability to evaluate broad-scale food security and make sustainable P management policies.

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Table 1. Comparison between non-spatial DPPS (Sattari et al., 2012) and the spatially explicit DPPS model.

Property	Non-spatial DPPS model (Sattari et al., 2012)	Spatially explicit model
Spatial Resolution	Continental	0.5 degree
Land use changes	No	Considering arable land expansion
Variable budget of soil P	Fixed	Recalculated every year
Initialization of soil pools	No	Initialized the soil pools for the pre-industrial period
Crop P uptake	Fixed fraction	Michaelis-Menten kinetics
P flow (μLS and μSL)	Fixed number	Calculated for every grid cell
Erosion and runoff	Fixed number	Changed with the pool size
Soil properties	One class in continental level	Grid-based resolution

Table 2. Model parameters of DPPS.

Parameter	Description (unit)	Method/Value/Source
<i>fr_weathering</i>	Fraction of apatite in soils that is released during weathering (no dimension)	0.001747
<i>weathering</i>	The annual amount of P that becomes available from the parent material (kg P ha ⁻¹ yr ⁻¹)	<i>fr_weathering</i> multiplied by the soil P present as apatite according to Yang et al. (2010)
<i>deposition</i>	The annual amount of P from atmospheric deposition (kg P ha ⁻¹ yr ⁻¹)	Modelled gridded P deposition from Mahowald et al. (2008)
<i>fertilizer</i>	Fertilizer P input (kg P ha ⁻¹ yr ⁻¹)	Beusen et al. (2016)
<i>manure</i>	Manure P input (kg P ha ⁻¹ yr ⁻¹)	Beusen et al. (2016)
<i>LP</i>	The LP pool, calculated for every grid every year (kg P ha ⁻¹)	Equation. 1
<i>SP</i>	The SP pool, calculated for every grid every year (kg P ha ⁻¹)	Equation. 2
<i>LP_yang</i>	The initial labile pool size	Yang et al. (2010)
<i>SP_yang</i>	The initial stable pool size	Yang et al. (2010)
<i>max_uptake</i>	The maximum amount of uptake by plants from <i>availableP</i> (kg P ha ⁻¹ yr ⁻¹)	Changes with time. The upper limit of <i>max_uptake</i> is 100
<i>init_recovery</i>	The initial recovery fraction, which is the slope of the P response curve at zero $Available_p$, specific per soil type (no dimension)	Batjes (2011)
<i>fr_mobile</i>	The calibration coefficient which mimics the direct P availability within the labile pool	$availableP = fr_mobile \times LP$
<i>availableP</i>	The amount of P available to plant roots (kg P ha ⁻¹ yr ⁻¹)	$fr_mobile \times LP$
<i>litter</i>	The P content in crop litter that returns to the soil (kg P ha ⁻¹ yr ⁻¹)	Soils under natural vegetation: see text Arable soil: 0
μ_{LS}	Transfer time for LP to SP (years)	5 (Sattari et al., 2012)
μ_{SL}	Transfer time for SP to LP (years)	Equation. 5
<i>uptake</i>	Crop P uptake (kg P ha ⁻¹ yr ⁻¹).	Soils under natural vegetation: the inputs from <i>litter</i> , <i>deposition</i> and <i>weathering</i> Arable soil: Equation. 6
<i>agri_2_natural_years</i>	The years for abandoned land to revert to natural conditions	30

Table 3. The parameters included in the sensitivity analysis.

Symbol	Min	Default	Max	Type
μ_{LS}	3	5	7	Range
<i>fr_weathering</i>	0.0014	0.001747	0.0021	Range
<i>agri_2_natural_years</i>	10	30	50	Range
<i>max_uptake</i>	0.75	1	1.25	Multiplier
<i>fr_mobile</i>	0.75	1	1.25	Multiplier
<i>LP_yang</i>	0.75	1	1.25	Multiplier
<i>TP_yang</i>	0.75	1	1.25	Multiplier
<i>deposition</i>	0.75	1	1.25	Multiplier
<i>init_recovery</i>	0.75	1	1.25	Multiplier
<i>runoff</i>	0.75	1	1.25	Multiplier
<i>fertilizer</i>	0.75	1	1.25	Multiplier
<i>manure</i>	0.75	1	1.25	Multiplier

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Table 4. Standardized regression coefficient (*SRC*) representing the relative sensitivity of the soil *LP*, *SP* pools and crop P uptake to the variation of 12 model parameters. Columns values represent the global model results for the year 1950 and 2000.

Parameter	<i>LP</i>		<i>SP</i>		<i>uptake</i>	
	1950	2000	1950	2000	1950	2000
<i>μ_{LS}</i>	0.04	0.09	0.00	-0.01		0.05
<i>fr_weathering</i>	0.04	0.04	0.01	0.02		0.04
<i>agri_2_natural_years</i>						
<i>max_uptake</i>	-0.07	-0.12	-0.03	-0.05	0.85	0.47
<i>fr_mobile</i>	-0.03	-0.11	-0.01	-0.03	0.29	0.50
<i>LP_yang</i>	1.00	0.96	-0.09	-0.11	0.23	0.37
<i>TP_yang</i>	-0.02	-0.08	1.00	0.97	-	-0.03
<i>deposition</i>						
<i>init_recovery</i>	-0.02	-0.10	-0.01	-0.03	0.29	0.51
<i>runoff</i>				0.00		
<i>fertilizer</i>	0.06	0.24	0.02	0.10	0.03	0.18
<i>manure</i>	0.06	0.10	0.03	0.06	0.02	0.06

570 ^a Cells with no value represent insignificant *SRC* values; all cells with values have significant *SRC*, numbers with normal font indicate values $-0.2 < SRC < 0.2$; numbers with **bold** font indicate values < -0.2 and > 0.2 . The absolute value of an *SRC* value of 0.2 indicates that the parameter concerned has an influence of $0.2^2 = 0.04$ (4 %) on the model variable considered. We refer to this as an important effect on the model output variable considered. Positive *SRC* values implies that a higher parameter value yields a higher model output, while negative *SRC* values indicate an inverse relation between model output and parameter value. The blank cells are not significant. The full results with data for world regions and selected countries are in Table S4.

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Table 5. Comparison with other studies for global cropland.

Reference	P fertilizer application ^a (year)	Manure P Application ^a (Tg P yr ⁻¹)	Crop P uptake ^a	Agronomic soil P budget ^a
Sattari et al. (2012)		23.2 (2007)	11.5	11.7
MacDonald et al. (2011)	14.2 (2000)	9.6	12.3	11.5
Chen and Graedel (2016)	20.2 (2013)	6.5	12.4	14.3
Bouwman et al.(2009)	>13 (2000)	7.0	10.0	11.0
This study	13.6 (2000)	6.1	10.7	9.0
	15.8 (2010)	7.2	12.0	11.0

^a P fertilizer application is the application of mineral fertilizer; agronomic soil P budget is the difference between the sum of P mineral fertilizer and manure and crop P uptake (agronomic soil P budget = P fertilizer application + manure P application - crop P uptake).

Figure captions

585 Figure 1. Scheme of the DPPS model. The model includes two dynamic P pools, i.e. the labile pool (*LP*) and the stable pool (*SP*), comprising both organic and inorganic P. Five inputs of P to the system are defined: mineral fertilizer and manure, weathering, deposition, fresh soil and litter. μ_{LS} and μ_{SL} (years) denote the transfer time of P from *LP* to *SP* and from *SP* to *LP*, respectively. Modified from Sattari et al. (2012).

590 Figure 2. Trends of annual P application (including P from manure and fertilizer), historical P uptake and simulated P uptake in cropland for the period 1900-2010 in the continents and world regions : Western Europe (a) , North America (b) , Africa (c), Asia (d), South America (e) and Oceania (f), and the countries : China (g), United States (h), South Africa (i) and France (j). Open and closed dots refer to P application and observed P uptake rates from FAO validation data. Solid lines refer to simulated P uptake rates.
595 Western Europe includes Andorra, Austria, Belgium, Denmark, Faroe islands, Finland, France, Germany, Gibraltar, Greece, Holy See (Vatican City State), Iceland, Ireland, Italy, Liechtenstein, Luxembourg, Monaco, Netherlands, Norway, Portugal, San Marino, Spain, Svalbard and Jan Mayen, Sweden, Switzerland, United Kingdom. We can produce the results of every year, however, due to the space limitation, we only show the results of every ten years from 1900 to 2010.

600 Figure 3. Relationship between simulated and historical P uptake for individual regions, countries and the world presenting results for every two years from 1961 to 2010. Dashed line is 1:1.

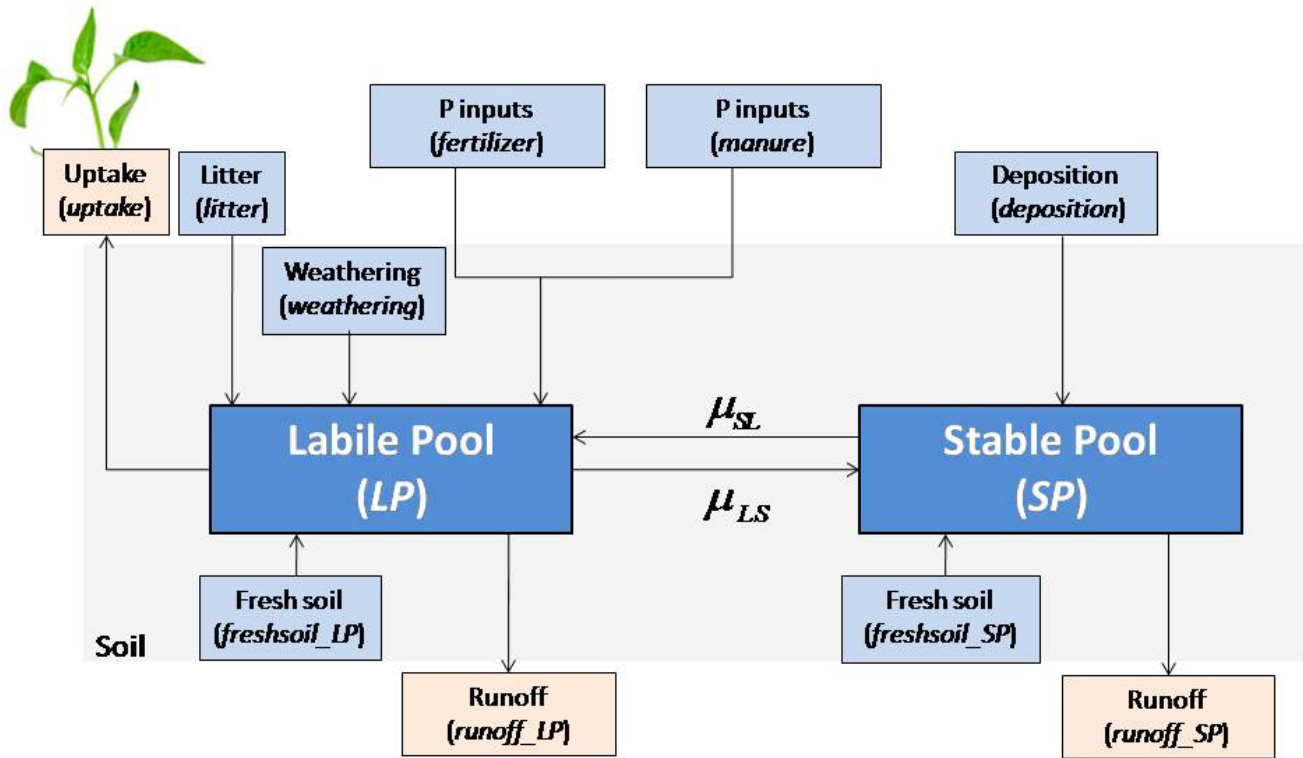
605 Figure 4. P uptake vs. P application between 1900 and 2010. There are two P uptake rates with the same amount of P application, which shows the contribution of residual P to the crop production in Western Europe (a) and France (j).

610 Figure 5. The total soil P inputs (fertilizer, manure and deposition), soil P outputs(crop uptake and runoff) and biogeochemical soil P budget (soil P input - soil P output) of different regions and countries. Soil P inputs are positive and outputs are negative.

615 Figure 6. (a) Global soil P budget in 2010. Soil P budget is the biogeochemical soil P budget, which is the difference between total soil P input (mineral fertilizer, manure and deposition) and total soil P output (uptake and runoff); the global distribution of (b) labile P (*LP*) and (c) total P in global agricultural land soils in 2010. Yearly grid maps for the period 1900-2010 can be viewed in supplementary movie S5.

Figure 7. Changes of soil *LP* (a) in different world regions and b) in different countries; and *TP* pools (c) for different world regions and (d) countries in arable land for the period 1900-2010.

620 Figure 8. Relative contribution of expansion of cropland area and crop P uptake in kg ha^{-1} for Western Europe, North America, Africa, Asia, South America, Oceania and Central Europe.



● Improvement of the spatially explicit DPPS model:

1. Spatially explicit calculation
2. Incorporation of land use change
3. Dynamic transfer between different soil P pools
4. Initialization P pools with global observation
5. Dynamic P loss by runoff
6. Dynamic calculation of crop P uptake
7. Time-variant maximum uptake parameter

Figure 1.

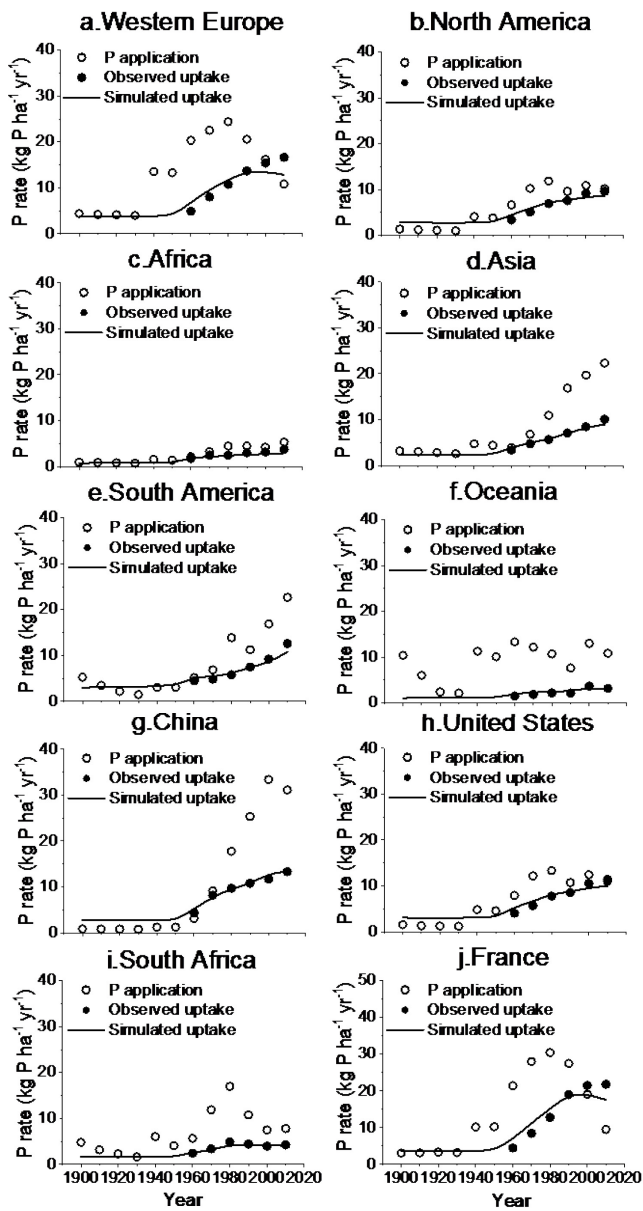
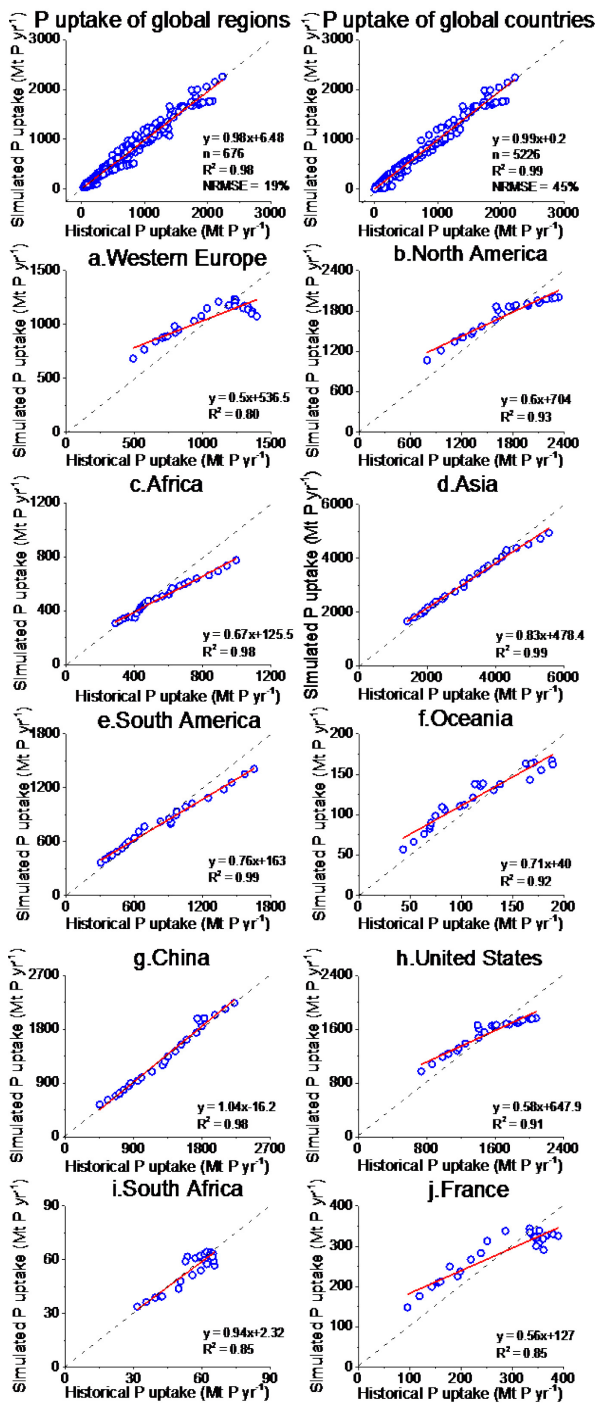


Figure 2.



635 Figure 3.

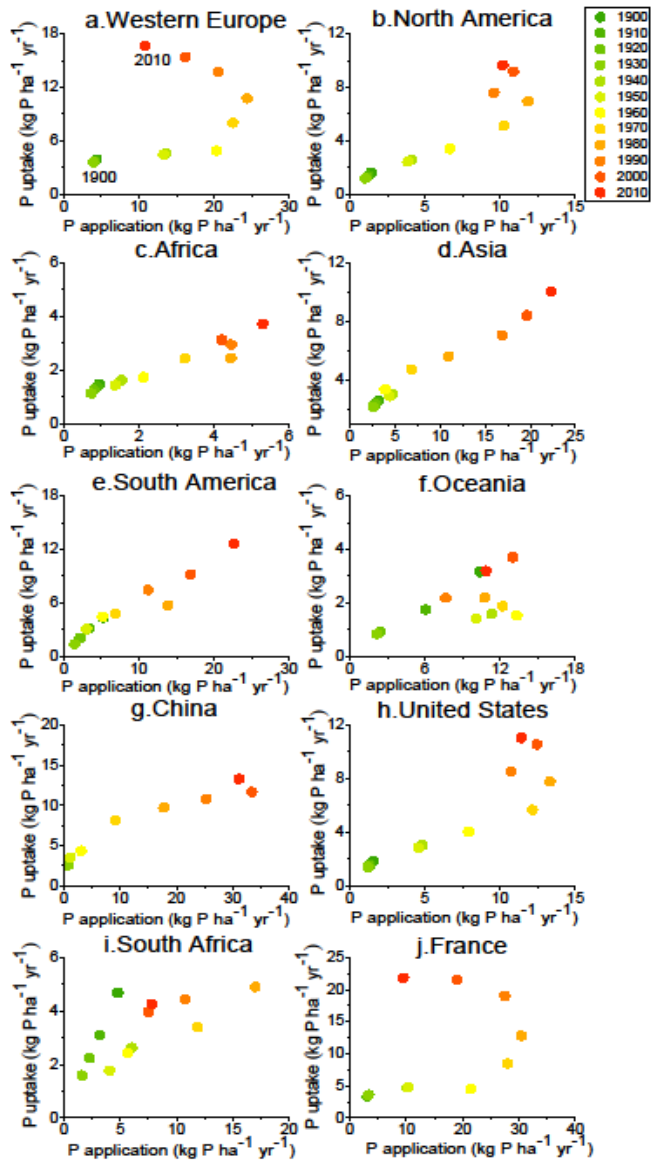


Figure 4.

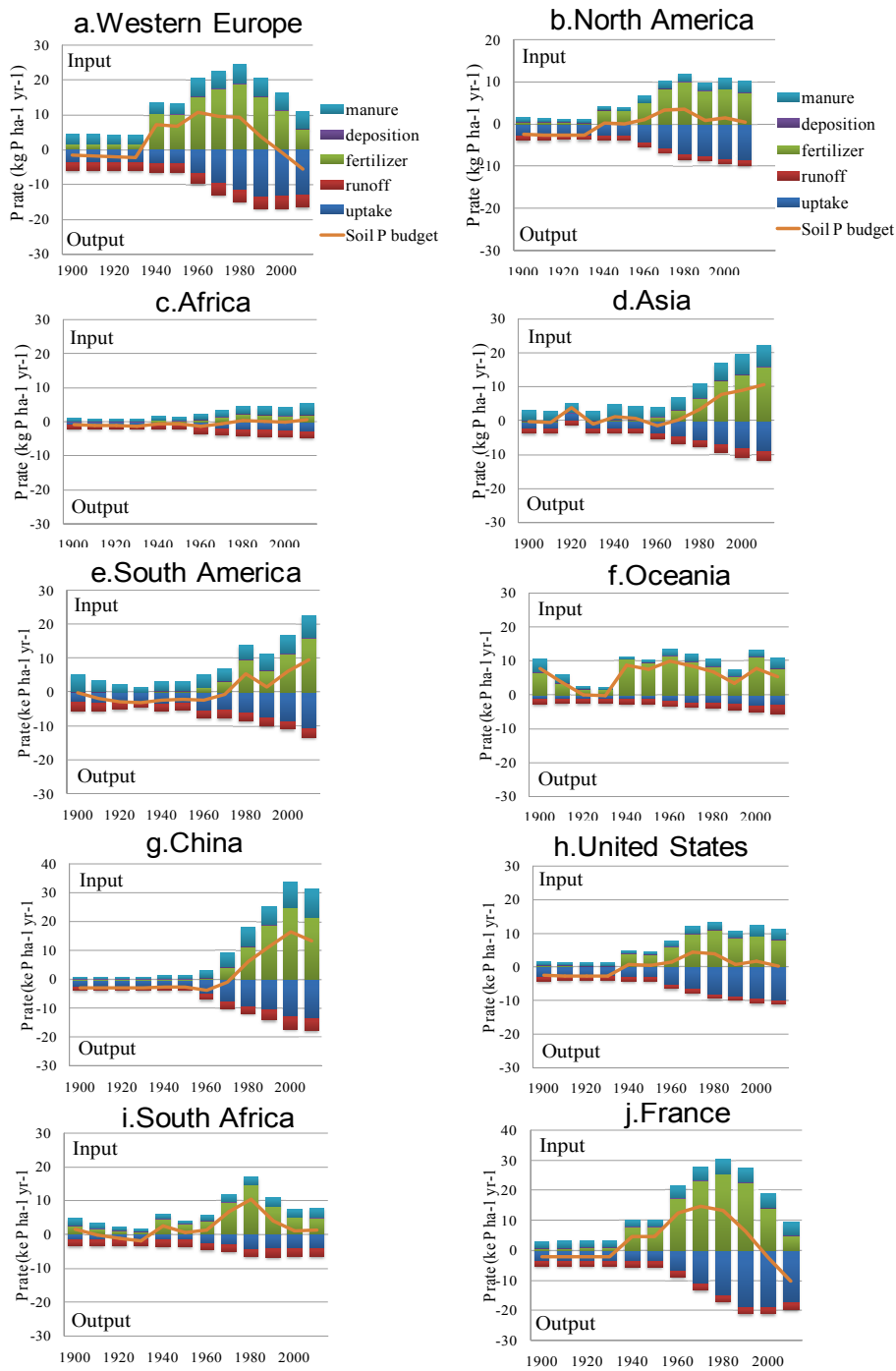
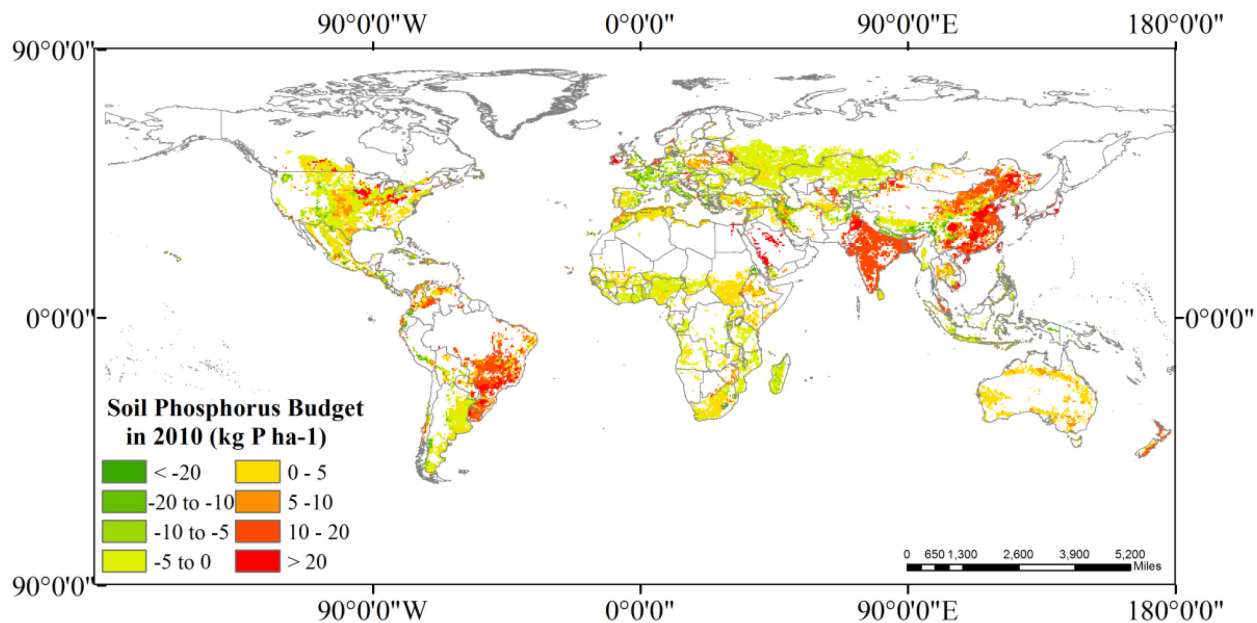
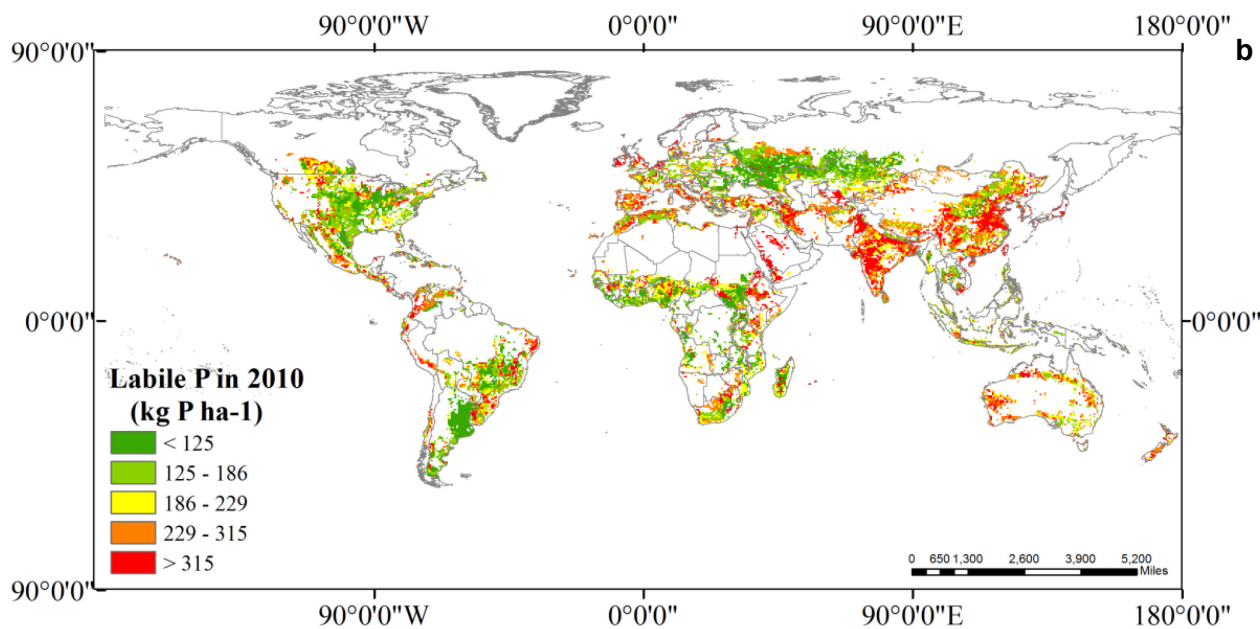


Figure 5.



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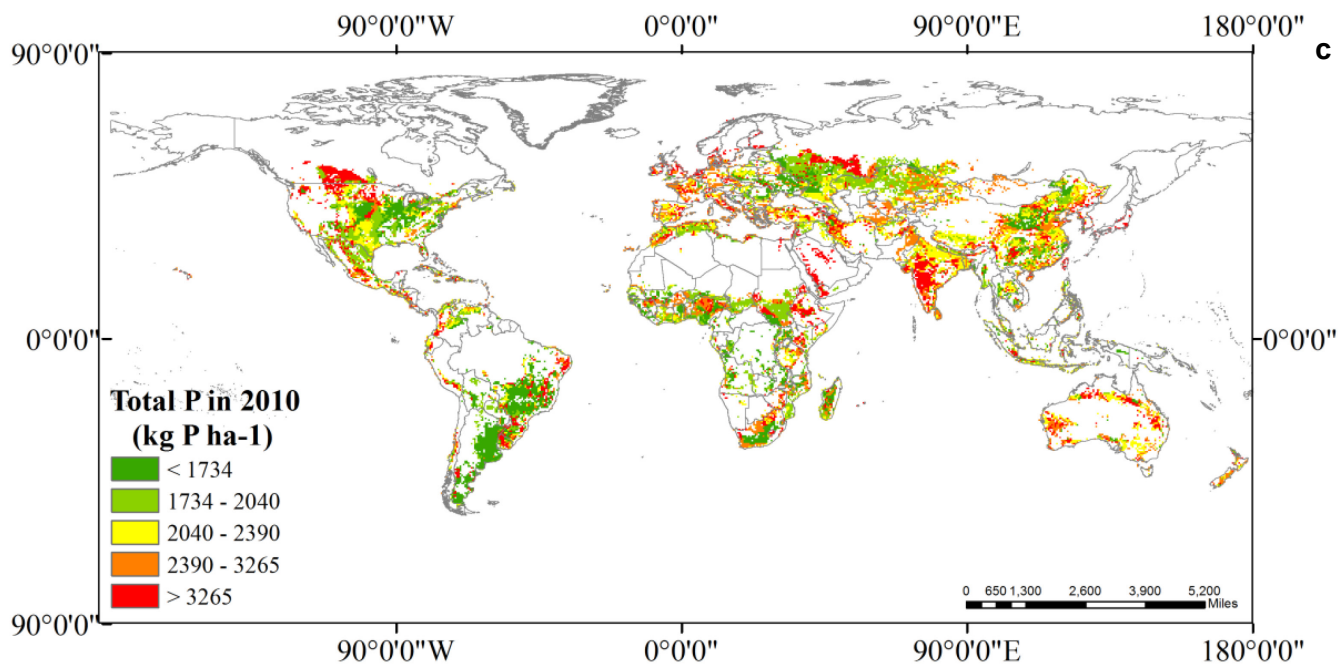
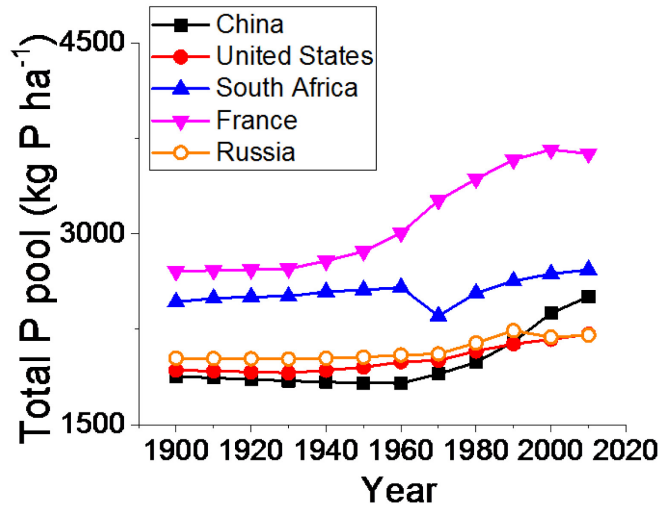
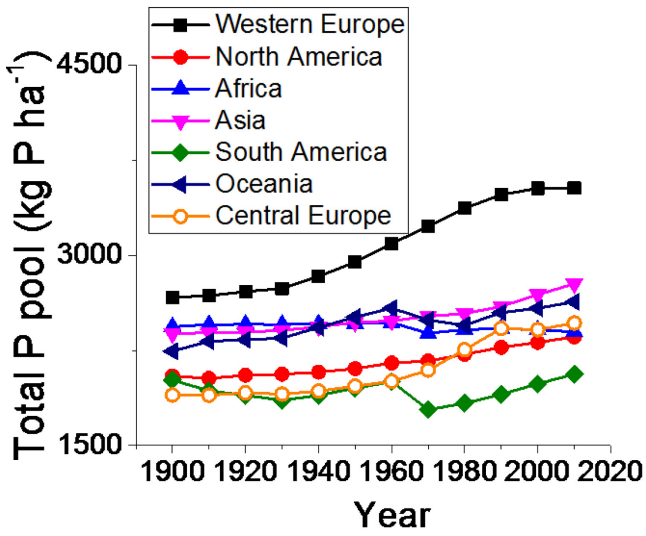
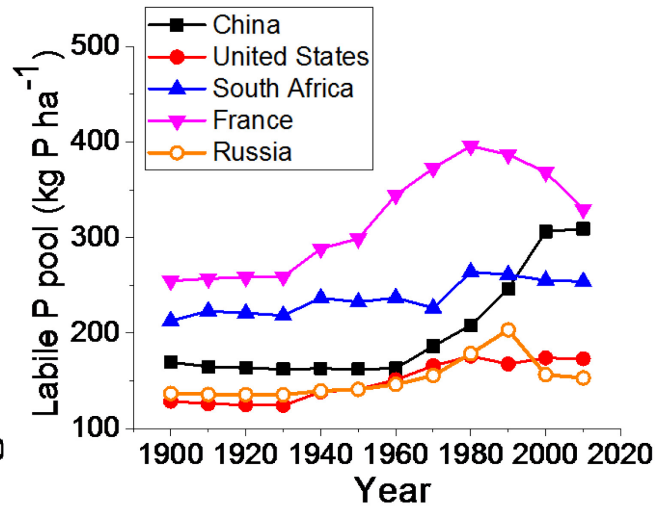
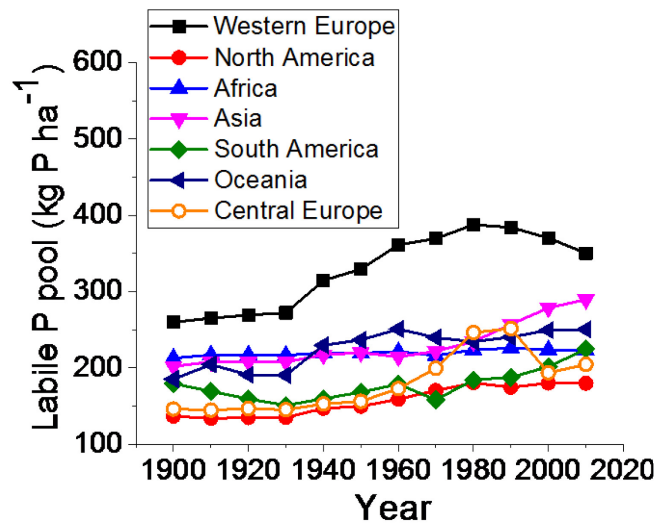


Figure 6.



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Figure 7.