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# Supplement of

# Global and regional phosphorus budgets in agricultural systems and their implications for phosphorus-use efficiency

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# **Supporting Information**

# 1. Global P budgets in agricultural systems and their implications for phosphorus-

#### use efficiency

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- 4 Our study concentrated on global and regional P budgets in agriculture and their
- 5 phosphorus-use efficiency (PUE) in four main agricultural subsystems: cropland,
- 6 managed grassland (hereafter, pasture), livestock, and humans. The specific P pools in
- 7 this study were phosphate rock, phosphates produced from that rock, the atmosphere,
- 8 pasture, livestock, cropland, harvested crops, humans, and the environment. The inputs
- 9 and outputs for each P pool are described below.

# 1.1 Phosphate Rock

- As a non-renewable resource, phosphate rock is the main source for most of the P
- that humans use (Liu et al., 2008). Mined phosphate rock is processed into phosphates
- 13 (section 1.2) for subsequent use. Data for the annual quantities of mined phosphates
- 14 were obtained from the International Fertilizer Industry Association (IFA;
- 15 http://www.fertilizer.org/Statistics), which provided the data for global and regional
- levels (expressed as the P<sub>2</sub>O<sub>5</sub> equivalent). The annual amount of P that is mined can be
- calculated from the proportion of the P in the  $P_2O_5$  equivalent.

#### 1.2 Phosphates

- The P in phosphates is used in phosphate fertilizer, feed additives, detergents, and
- 20 other uses. The Food and Agriculture Organization (FAO;
- 21 http://www.fao.org/faostat/en/#home) provides the annual consumption of phosphate
- 22 fertilizer for each country. Some of the phosphate fertilizer is applied to pasture in some

countries (mainly in Europe), and FAO (2002) estimated the consumption of phosphate fertilizer by cropland and pasture in each country. Thus, from this information, we can estimate the amounts of phosphate fertilizer applied to cropland and pasture. In addition, 8% of the global mined phosphate is used in feed additives, and the remainder of the phosphate is used in detergents or other uses for humans (Ringeval *et al.*, 2014).

# 1.3 Atmosphere

The P input from the atmosphere refers to the atmospheric P deposition in cropland and pasture areas, whereas anthropogenic P outputs comes from the burning of crop residues and bioenergy within the agricultural system. All these inputs and outputs were estimated by using the PKU-FUEL model (Wang *et al.*, 2014, 2015). Wang *et al.* (2014) used the global 3D atmospheric transport model LMDz-INCA to simulate the transport and deposition of aerosols from different sources, with specific reference to the P concentration. The modeled P deposition maps agreed well with P deposition observed at 121 stations worldwide (Wang *et al.*, 2015).

# 1.4 Cropland and harvested crops

In addition to application of phosphate fertilizer and atmospheric P deposition, cropland P inputs also come from livestock manure and human excreta (as sewage sludge), as well as recycled crop residues. Cropland P outputs include P removals in harvested biomass (crops and crop residues) and P output by leaching or erosion.

#### 1.4.1 Cropland P inputs

The application of phosphate fertilizer and atmospheric deposition have been presented above. Manure P inputs from livestock and humans are described in sections

1.4.5 and 1.4.6, whereas P in recycled crop residues is presented in section 1.4.2.

# 1.4.2 Cropland P outputs

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Crops are the economically valuable outputs of cropland. FAO provided data for the 224 (Table SI-1) countries for the production of 178 crops (Table SI-2) that we included in our analysis. We grouped these crops into 13 types: wheat, rice, maize, other cereals, soybeans, palm oils, other oil crops, sugar crops, fibers, roots and tubers, vegetables, fruit, and other crops. The distributions of the harvested crops were also obtained from FAOSTAT, including crops that produced human food, livestock feed, industrial processing, wastes, and other uses. Feed crops flow directly into the livestock subsystem, and the remaining crops flow into the human subsystem. P in harvested crops can be estimated by multiplying their biomass production by their P content (COMIFER, 2007; USDA-NRCS, 2009; Waller, 2010). In addition, crop residues are also important for P removal from cropland. Half of the crop residues are returned to cropland, and 25% are used as livestock fodder (Liu et al., 2008). Of the remaining 25% of crop residues, some proportion is burned, and can be estimated by the PKU-FUEL model, and the remainder is transferred to the human subsystem. FAOSTAT provides the amount of crop residues that are recycled to cropland, so we can estimate P in these crop residues by multiplying the amount of residues by the corresponding P content. Based on the distribution of crop residues, we can estimate the P flows in harvested crop residues. The global P loss from leaching and runoff was estimated by Bouwman et al. (2013), who noted that these losses account for approximately 12.5% of the total P inputs in agricultural land. Thus, we used 12.5% of all P inputs in agricultural land to

- represent the leaching and runoff loss of P.
- 68 1.4.3 Cropland soil P budget
- The cropland soil P budget ( $\Delta P$ ) refers to the balance between all P inputs and all
- 70 Poutputs for cropland.
- 71 1.4.4 Pasture

- For pasture, the P inputs are from livestock manure, phosphate fertilizer, and 72 atmospheric deposition. Harvested grass is the economically valuable product removed 73 from pasture, but leaching also results in a P loss from pasture. The data on production 74 75 of grass as livestock feed was obtained from Herrero et al. (2013) and ORCHIDEE-GM (Chang et al., 2013, 2015). The P content of the grass was estimated at 0.19 to 76 0.56% of its biomass (Antikainen et al., 2005; COMIFER, 2007; USDA-NRCS, 2009; 77 78 Waller, 2010); we chose the midpoint of this range (0.38%) as the P content of grass in our study. Based on this P content, we estimated the P content in harvested grass. The 79 pasture P budget  $(\Delta P)$  was then estimated as the balance between its total P inputs and 80
- 82 1.4.5 Livestock

total P outputs.

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- The stock of P in livestock does not change substantially over time, so we assumed that livestock P inputs equaled livestock P outputs.
  - Livestock P outputs include P in livestock economic products (meat, eggs, and milk) and P in manure (Table SI-3). FAOSTAT provided the data for the production of meat, eggs, and milk for 16 types of animals. We used the P contents of meat, eggs, and milk from Grote *et al.* (2005) to estimate the P stocks in the livestock products.

FAOSTAT estimated the amount of nitrogen in manure, as well as its distribution to pasture, to cropland as manure, and to the environment as waste. Thus, using P:N ratios for manure of different animals (MWPS-18, 1985; OECD Secretariat, 1991; Levington Agriculture, 1997; Sheldrick *et al.*, 2003; ASAE, 2005), we estimated the P flows in livestock manure.

Livestock P inputs include grass from pasture, crops, and crop residues from cropland, as well as feed additives from phosphate and processed feed from wasted human food. Therefore, the P in the feed processed from human food can be calculated by subtracting the other P inputs from the livestock total P outputs.

#### 1.4.6 Humans

As in the analysis for livestock, we assumed that total P inputs equal total P outputs for humans. All inputs have been described above. Human P outputs consist of food processed to provide livestock feed, transfer of excreta to cropland as sewage sludge, biomass combustion for energy, and wastes released into the environment. The total amount of P in human excreta can be estimated by multiplying the human population by the per capita annual amount of P in human excreta (Smil, 2000; Cordell *et al.*, 2009). Liu *et al.* (2008) reported that 30% of human excreta in urban areas and 70% of human excreta in rural areas is currently recycled to cropland. Thus, based on these proportions, we can calculate the amount of P in human manure that is contributed to cropland. The total amount of P released into the environment as waste can be estimated by subtracting the other human P outputs from the total human P inputs.

#### 1.4.7 Environment

- The P inputs to the environment include P leaching or runoff from agricultural soils and P flows into the environment as waste from humans. These values are described earlier in this section.
- For the methods described in the following sections, details of the components of the equations are presented in Table SI-4.

# 2. Annual P budgets of cropland and pasture soils

- Annual changes in soil P (the soil P budget,  $\Delta P$ ) are calculated as the differences
- between annual inputs and outputs:

$$119 \qquad \mathsf{D}P_{cropland} = (P_{\mathit{fer-crop}} + P_{\mathit{dep-crop}} + P_{\mathit{man-crop}} + P_{\mathit{slu-crop}} + P_{\mathit{res-rec}} + P_{\mathit{crop-seed}}) - (P_{\mathit{crop}} + P_{\mathit{crop-res}} + P_{\mathit{runoff-crop}})$$

120 (Eq. 1)

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$$\Delta P_{pasture} = (P_{fer-pas} + P_{dep-pas} + P_{man-pas}) - (P_{grass} + P_{unoff-pas})$$
 (Eq. 2)

- where  $\Delta P_{\text{cropland}}$  and  $\Delta P_{\text{pasture}}$  are annual soil budgets for cropland and pasture,
- respectively;  $P_{\text{fer-crop}}$  and  $P_{\text{fer-pas}}$  are the corresponding phosphate fertilizer applications;
- 124  $P_{\text{dep-crop}}$  and  $P_{\text{dep-pas}}$  are the corresponding atmospheric P deposition fluxes; and  $P_{\text{man-crop}}$
- and  $P_{\text{man-pas}}$  are the corresponding livestock manure fluxes applied to cropland and
- pasture.  $P_{\text{slu-crop}}$  is the input as human sewage sludge, which is only applied to cropland;
- 127  $P_{\text{res-rec}}$  is the input of P from recycled crop residues returned to cropland;  $P_{\text{crop-seed}}$  is the
- P in seeds;  $P_{\text{crop}}$  and  $P_{\text{crop-res}}$  are P removals in harvested crop biomass and crop residues,
- respectively;  $P_{\text{grass}}$  is the P removed in the intake of grass by animals; and  $P_{\text{runoff-crop}}$  and
- $P_{\text{runoff-pas}}$  are losses of dissolved and particulate P to bodies of water from cropland and
- pasture, respectively. All units for these fluxes are in kg P ha<sup>-1</sup> yr<sup>-1</sup> or Tg P yr<sup>-1</sup>,
- depending on the area or period they describe.

#### 3. Annual P budgets of cropland and pasture soils

We defined the annual labile P inputs ( $P_{labile-input}$ ) and stable P inputs ( $P_{stable-input}$ ) as:

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$$P_{labile-input} = 0.8 \hat{P}_{Inputs} \quad \text{(Eq. 3)}$$

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$$P_{stable-input} = 0.2 \cdot P_{Inputs} \quad \text{(Eq. 4)}$$

- where  $P_{\text{inputs}}$  represents the sum of all input fluxes in the first term on the right side of
- Eq. 1, excluding  $P_{\text{crop-seed}}$ . If  $P_{\text{labile-input}} \ge P_{\text{removal}}$ , with  $P_{\text{removal}}$  being the sum of
- removals as  $P_{\text{crop}}$ ,  $P_{\text{crop-res}}$ , and  $P_{\text{runoff-crop}}$  for cropland and the sum of removals as  $P_{\text{pasture}}$
- and  $P_{\text{runoff-pasture}}$  for pasture, the surplus labile P input is transferred to the stable P pool
- at the end of each year. Given the stable P losses by leaching or runoff ( $P_{\text{runoff}}$ ), the soil
- P export and the budget of the stable soil P pool are given by:

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$$P_{soil-exp} = P_{runoff}$$
 (Eq. 5)

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$$\Delta P_{stable} = P_{stable-input} + P_{labile-input} - P_{removal} - P_{runoff} \quad (Eq. 6)$$

- If  $P_{\text{labile-input}} < P_{\text{removal}}$ , there is no transfer of labile P into the stable P pool, but the
- extra P demand for crop biomass is satisfied by a transfer from the pool of stable P.
- In this case, the soil P balance includes P lost by leaching or runoff into bodies of
- water and the surplus labile P is incorporated in crop biomass from the stable P pool:

149 
$$P_{soil-exp} = P_{runoff} + (P_{removal} + P_{labile-input}) (Eq. 7)$$

Thus, the net annual soil-P budget can be estimated by:

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$$\Delta P = \Delta P_{stable} + P_{seed} \quad (Eq. 8)$$

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#### 4. Human and livestock P budgets

- We assumed that the stocks of P in living livestock, in livestock products, and in
- human bodies were constant over time. Total annual P inputs for humans and livestock

must then equal their P outputs each year, which defines the mass-balance equations for these two subsystems:

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$$P_{grass} + P_{feed-add} + P_{pro-feed} + P_{crop-feed} + P_{res-add} = P_{man} + P_{meat} + P_{egg} + P_{milk}$$
 (Eq. 9)

$$P_{crop-hum} + P_{res-add} + P_{meat} + P_{egg} + P_{milk} + P_{other} = P_{pro-feed} + P_{bioenergy} + P_{hum-env} + P_{slu-crop}$$

159 (Eq. 10)

where  $P_{\text{meat}}$ ,  $P_{\text{egg}}$ , and  $P_{\text{milk}}$  are the P fluxes associated with meat, eggs, and milk consumed by humans, respectively, and  $P_{\text{grass}}$ ,  $P_{\text{feed-add}}$ ,  $P_{\text{crop-feed}}$ ,  $P_{\text{res-feed}}$ , and  $P_{\text{pro-feed}}$  represent the ingestion of P by animals from grazed grass biomass, feed additives, crop biomass and residue feed, and feed from food not consumed by humans (see Fig. 1);  $P_{\text{man}}$  is the total flux of P rejected by animals in the form of manure delivered to both cropland and pasture;  $P_{\text{crop-hum}}$  defines the P input to humans from cropland, and represents the consumption of crop products;  $P_{\text{cropres-hum}}$  represents the P flux in crop residues used by humans to generate bioenergy;  $P_{\text{other}}$  represents the P input flux to humans directly from minerals (detergents and other non-fertilizer products);  $P_{\text{bioene}}$  is P lost from humans to the environment from the use of biofuels harvested from crops (thus, not including wood bioenergy);  $P_{\text{slu-crop}}$  is the input as human sewage sludge, which is only applied to cropland; and  $P_{\text{hum-env}}$  is the remainder of the P flux lost in human sewage, calculated as the amount that remains after accounting for the other terms.

#### 5. Phosphorus-use efficiencies

The PUE of cropland ( $\epsilon_{cropland}$ ), pasture ( $\epsilon_{pasture}$ ), and livestock ( $\epsilon_{livestock}$ ) is defined

176 as:

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$$\varepsilon_{cropland} = \frac{P_{crop}}{P_{fer-crop} + P_{man-crop} + P_{slu-crop} + P_{dep-crop}}$$
(Eq. 11)

178 
$$\varepsilon_{pasture} = \frac{P_{grass}}{P_{fer-pas} + P_{man-pas} + P_{dep-pas}} \quad \text{(Eq. 12)}$$

179 
$$\varepsilon_{livestock} = \frac{P_{meat} + P_{egg} + P_{milk}}{P_{grass} + P_{feed-add} + P_{crop-feed} + P_{res-feed} + P_{pro-feed}}$$
(Eq. 13)

- 180 We defined the PUE of human food ( $\epsilon_{food}$ ) as the ratio of P in human excreta to the total of all P inputs in human food. This represents an exception to our definition, since
- $\varepsilon_{food} = \frac{P_{excreta-hum}}{P_{crop-food} + P_{meat} + P_{egg} + P_{milk}}$ (Eq. 14)

human excreta have no economic value.

- Finally, we defined the P yield of livestock products per unit area of pasture  $(YP_{lp})$
- 185 pasture) as:

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$$YP_{lp-pasture} = \frac{P_{meat} + P_{egg} + P_{milk}}{A_{pasture}} \times \frac{P_{grass}}{P_{liv-input}} \quad \text{(Eq. 15)}$$

- where  $A_{\text{pasture}}$  is the area of pasture in a given region and  $P_{\text{liv-input}}$  refers to all livestock P inputs, including grazed grass from pasture, crops used as feed, and animal
- feed additives from phosphates given by humans to livestock.

#### 6. International trade dependency ratio

The P fluxes associated with the international trade of fertilizers, food, feed, and fiber commodities can also be associated with dependency ratios. The fertilizer import P-dependency ratio ( $F_{\text{fer}}$ ) is expressed as the ratio of P in imported fertilizers ( $P_{\text{fer-imp}}$ ) to P in all fertilizers ( $P_{\text{fer-con}}$ ) consumed by a country:

$$F_{fer} = \frac{P_{fer-imp}}{P_{fer-con}} \quad \text{(Eq. 16)}$$

The food import dependency ratio ( $F_{\text{food}}$ ) is expressed as the ratio of P in food imports ( $P_{\text{food-imp}}$ ) to P in all food consumed in one country:

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$$F_{food} = \frac{P_{food-imp}}{P_{food-pro} + P_{food-imp} - P_{food-exp}}$$
 (Eq. 17)

The proportion of imports ( $F_{total}$ ) is expressed as the ratio of total P imported as fertilizers and food to the total P in fertilizers and food:

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$$F_{total} = \frac{P_{fer-imp} + P_{food-imp}}{P_{fer-con} + P_{food-pro} + P_{food-imp} - P_{food-exp}}$$
(Eq. 18)

#### 7. Comparisons at a national level

We estimated the flows of P in traded food and fertilizer for the United States, for China, for Australia and France, and for Japan from in 2010 (Fig SI-1). We chose these countries because they were representative of the combinations of fertilizer exporter or importer with food exporter or importer. We then compared our results with those in previous reports.

#### 7.1 United States

The United States is an important exporter of food and phosphate fertilizer. Suh and Yee (2011) reported a net P export in food of 413 Gg P in 2007, which is slightly lower than the value of 435 Gg P in 2010 in our study. They estimated the net export of phosphate fertilizer as 1291 Gg P in 2007, which is slightly higher than the value of 1196 Gg P in 2010 in our study. The net export of food increased slightly from 2002 to 2010, whereas exports of phosphate fertilizer have decreased.

#### 7.2 China

Chinese food imports have been increasing due to population growth and dietary

changes in China, especially in recent years, and food security is therefore a potentially serious problem in China. The trade in phosphate fertilizer changed greatly during the study period. Before 2007, China depended strongly on imported phosphate fertilizer, with a decreasing trend. However, China became a large exporter of phosphate fertilizer after 2007, with the exports increasing thereafter. If this trend continues, a P scarcity could develop in China. Our results indicated that the soil P accumulation in wheat cropland (29.4 kg P ha yr<sup>-1</sup>) was higher than the national level of 25.42 kg P ha yr<sup>-1</sup>. However, the accumulation of P in rice and maize fields was lower than the national average (Ma *et al.*, 2011).

# 7.3 Australia and France

Australia and France both have relatively stable food exports, with about 100 Tg P yr<sup>-1</sup> stored in food. Both countries import phosphate fertilizer, but at decreasing rates. Both countries are main crop exporters. Senthilkumar *et al.* (2012) reported that France imported 113 Gg P yr<sup>-1</sup> in crops and feed and 318 Gg P yr<sup>-1</sup> of phosphate fertilizer from 2002 to 2006, compared with 26 Gg P yr<sup>-1</sup> in food and feed and 271.4 Gg P yr<sup>-1</sup> in fertilizer in our study. France also exported 133 Gg P yr<sup>-1</sup> in food and feed and 29.8 Gg P yr<sup>-1</sup> in phosphate fertilizer during the same period, compared with 122 and 25.4 Gg P yr<sup>-1</sup>, respectively, in our study. The main differences may be because we did not account for the international trade of grass feed. Cordell *et al.* (2013) reported that Australia imported a net amount of 115 Gg P yr<sup>-1</sup> of phosphate fertilizer and exported a net amount of 106 Gg yr<sup>-1</sup> in crops, compared with 102 and 45 Gg yr<sup>-1</sup>, respectively in our study.

#### 7.4 Japan

Japan depends strongly on imported food and phosphate fertilizer from other countries. P in the imported food remained steady at around 110 Gg P yr<sup>-1</sup> in Japan. Although most of the applied phosphate fertilizer was obtained from other countries, Japan's cropland PUE was low, leading to a serious problem of soil P accumulation in cropland. Because the government is aware of the problem, they have made an effort to increase cropland PUE, which increased from 15.7% in 1985 to 20.1% in 2005 (Mishima *et al.*, 2010). This is close to our result (20% in 2002 to 23% in 2010); thus, the net imports of phosphate fertilizer have decreased in Japan.

#### 8. Comparisons of PUE

We defined PUE as the ratio of the economic P outputs to the total P inputs. Because the economic output differs among commodities, PUE is unique to each commodity. Table SI-6 presents the values for cropland as a whole, by region and globally, and Table SI-7 presents the values for individual crops. For pasture, the harvest P output equals the total P output, and does not account for loss of soil P by leaching or runoff into bodies of water. For cropland, P in the harvested crops was defined as the economic P output, excluding the P in crop residues. For livestock, the economic P outputs only include P embodied in livestock products. However, some portion of the livestock manure is accounted for as P inputs to cropland and pasture. We calculated PUE as follows:

**Cropland total PUE**: the ratio of P outputs in harvested crops and crop residues to total P inputs into cropland

**Cropland PUE**: the ratio of P outputs in harvested crops to total P inputs into cropland (i.e., excluding crop residues)

Figure SI-2 presents the relationship between cropland total PUE and cropland PUE.

Livestock total PUE: the ratio of P outputs in livestock products and in the recycled manure transferred to cropland and pasture to the total P inputs into livestock

Livestock PUE: the ratio of P outputs in livestock products to the total P inputs into livestock (i.e., excluding recycled manure)

# 8.1 Cropland PUE

Cropland total PUE had a strong and significant linear relationship with cropland PUE (Fig. SI-2). Global cropland total PUE was estimated to be 0.76, which was 1.65 times the global cropland PUE (excluding crop residues) of 0.46. However, with different crop harvest index values, cropland total PUEs and PUEs (excluding residues) differed among the regions (Table SI-6). The ratio of cropland total PUE to cropland PUE was relatively high in Southern and Southeastern Asia, northern Africa, and North America, and was relatively low in the Caribbean and Central America and South America. The cropland PUE was 0.67 when the cropland soil P balance was neutral because parts of the P output (i.e., the residues) are not considered. There was more recycling of P than loss of P in surface runoff into bodies of water. Thus, cropland total PUE should be more than 1 when the cropland soil P balance is neutral.

Substantial differences in PUE and total PUE occurred among crops because of their different harvest indices, yields, and external Pinputs (Table SI-7). Oil palm, fiber,

fruits, and vegetable crops had very low total PUE, and therefore a low total PUE to PUE ratio (<1.2) because few of their P inputs flowed into their crop biomass. In contrast, the remaining crop types (excluding the "other" category) had high total PUE because more of their P was transferred into the crop and crop residues, leading to a high total PUE to PUE ratio (>1.3). Furthermore, P inputs did not meet the P demand for wheat and other cereals. However, due to their low harvest index, cereals produced a large amount of crop residues; hence, their PUE was much lower than their total PUE, especially for rice and maize. For the "other" category, there was no difference between PUE and total PUE because there was little production of residues.

Based on the available data, it was not possible to determine the source of the cropland P (i.e., manure or mineral fertilizer) that was lost into bodies of water. Thus, it is hard to define a PUE term that accounts for the impacts of different fertilizer types. Since this is an important problem for managing P inputs and outputs in agricultural ecosystems, further research will be necessary to clarify the relationships between cropland total PUE and PUE for different crops, and how they are affected by human and natural factors.

#### 8.2 Livestock PUE

Because livestock total PUE includes P in recycled manure, its global value (0.83) was far higher than the global livestock PUE (excluding manure) of 0.06 (Table SI-6). These two PUE parameters differed greatly among the regions due to differences in the mixture and quantity of different livestock species, different livestock husbandry methods, and different manure management methods. The yield of livestock products

was very low in African countries, resulting in low livestock PUE. However, almost all their manure was applied to agricultural land as an important P input, leading to much higher livestock total PUE (≥0.92) than in other regions. Therefore, it will be necessary for African countries to find ways to increase the economic value of livestock outputs while continuing to use manure efficiently to relieve the pressure on global sources of phosphates. In contrast, efficient livestock husbandry allowed a higher proportion of P inputs to flow into livestock products in Eastern Asia and Europe (≥9.9%) than in Africa (<2%). However, as the application of phosphate fertilizer increased, the proportion of livestock manure recycled into agricultural soils decreased. Consequently, livestock total PUE was relatively low in Eastern Asia and Europe; this represents a waste of the livestock manure resource and excessive application of phosphate fertilizer. Therefore, countries in Eastern Asia and Europe should look for ways to increase their use of livestock manure.

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- related emissions to the atmospheric phosphorus budget. *Nature Geoscience*, **8**,
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Academy of Sciences of the United States of America, 111, 2459-2463.

Table SI-1: Global regions and countries

Eastern and Southern Africa	Northern Africa	Western and Central Africa	Eastern As	ia	Southern and Southeastern Asia	Western and Central Asia	Oceania	Europe	North America	The Carib and Cer Ameri	ıtral	South America
Angola	Algeria	Benin	China		Bangladesh	Afghanistan	American	Albania	Canada	Anguilla		Argentina
Botswana	Burkina Faso	Burundi	Japan		Bhutan	Armenia	Samoa	Andorra	Greenland	Antigua	and	Bolivia
British Indian	Chad	Cameroon	Democratic		Brunei	Azerbaijan	Australia	Austria	Mexico	Barbuda		Brazil
Ocean	Djibouti	Cabo Verde	People's		Darussalam	Bahrain	Cook Islands	Belarus	Saint Pierre	Aruba		Chile
Territory	Egypt	Central	Republic	of	Cambodia	Cyprus	Fiji	Belgium	and	Bahamas		Colombia
Comoros	Eritrea	African	Korea		India	Georgia	French	Bosnia and	Miquelon	Barbados		Ecuador
Kenya	Ethiopia	Republic	Republic	of	Indonesia	Iran	Polynesia	Herzegovina	United	Belize		Falkland Islands
Lesotho	Libya	Congo	Korea		Laos	Iraq	Guam	Bulgaria	States	Bermuda		French Guiana
Madagascar	Mali	Democratic	Mongolia		Malaysia	Israel	Kiribati	Croatia		British	Virgin	Guyana
Malawi	Mauritania	Republic of			Maldives	Jordan	Marshall	Czech Republic		Islands		Paraguay
Mauritius	Morocco	the Congo			Myanmar	Kazakhstan	Islands	Denmark		Cayman Isl	ands	Peru
Mayotte	Niger	Côte d'Ivoire			Nepal	Kuwait	Nauru	Estonia		Costa Rica		South Georgia
Mozambique	Somalia	Equatorial			Pakistan	Kyrgyzstan	New Caledonia	Faroe Islands		Cuba		and the South
Namibia	Sudan	Guinea			Philippines	Lebanon	New Zealand	Finland		Dominica		Sandwich
Réunion	Tunisia	Gabon			Singapore	Oman	Niue	France		Dominican		islands
Seychelles	Western	Gambia			Sri Lanka	Qatar	Northern	Germany		Republic		Suriname
South Africa	Sahara	Ghana			Thailand	Saudi Arabia	Mariana Islands	Gibraltar		El Salvado	r	Uruguay
Swaziland		Guinea			Timor-Leste	Syrian Arab	Palau	Greece		Grenada		Venezuela
Tanzania		Guinea-			Vietnam	Republic	Papua New	Hungary		Guadeloup	e	
Uganda		Bissau				Tajikistan	Guinea	Iceland		Guatemala		
Zambia		Liberia				Turkey	Pitcairn Islands	Ireland		Haiti		

Zimbabwe	Nigeria	Turkmenista	Samoa	Italy	Honduras
2	Rwanda	n	Solomon	Latvia	Jamaica
	São Tome and	United Arab	Islands	Liechtenstein	Martinique
	Principe	Emirates	Tokelau	Lithuania	Montserrat
	Senegal	Uzbekistan	Tonga	Luxembourg	Netherlands
	Sierra Leone	Yemen	Tuvalu	Macedonia	Antilles
	Saint Helena		Vanuatu	Malta	Nicaragua
	Togo		Wallis and	Moldova	Panama
			Futuna Islands	Monaco	Puerto Rico
				Netherlands	Saint Kitts and
				Norway	Nevis
				Poland	Saint Lucia
				Portugal	Saint Vincent and
				Romania	the Grenadines
				Russia	Trinidad and
				San Marino	Tobago
				Serbia	Turks and Caicos
				Montenegro	Islands
				Slovakia	
				Slovenia	
				Spain	
				Sweden	
				Switzerland	
				Ukraine	
				United	
				Kingdom	

30		rable 31-2. Crop categories and then 1 contents
Category	P content (% w/w)	Items
Wheat	0.38	wheat
Rice	0.25	rice
Maize	0.18 (0.09-0.27)	maize
Other	0.31 (0.29-0.34)	rye, oats, millet, sorghum, triticale, canary seeds, buckwheat, quinoa, fonio, popcorn,
cereals		mixed grains, cereals nes
Soybeans	0.60	soybeans
Oil palms	0.54	palm oil and kernels
Other oil	0.06 (0.04-0.08)	olives, sunflower seeds, sesame seeds, seed cotton, cottonseed, linseed, groundnuts with
crops	for oilseed rape,	shells, oilseed rape, coconuts, castor oil seeds, tung nuts, safflower seeds, mustard seeds,
	0.47 (0.32-0.62)	poppy seeds, oilseeds nes, melon seeds, hemp seeds, tallowtree seeds, karite nuts
	for others	(sheanuts), kapok fruit, jojoba seeds
Sugar	0.05 (0.04-0.06)	sugar beets, sugar cane, sugar crops nes
crops		
Fiber	0.67	seed cotton, cotton lint, other bast fibers, sisal, flax and tow fiber, fiber crops nes, jute,
		ramie, hemp tow waste, agave fibers nes, manila fiber (abaca), kapok fruit
Roots and	0.07 (0.04-0.09)	potatoes, cassava, taro (cocoyam), yams, sweet potatoes, yautia (cocoyam), roots and
tubers		tubers nes
Vegetables	0.06 (0.05-0.07)	cabbages and other brassicas, tomatoes, cauliflowers and broccoli, cucumbers and
		gherkins, dry onions, garlic, green peas, carrots and turnips, fresh vegetables nes,
		watermelons, other melons (including cantaloupes), spinach, pumpkins, squash and
		gourds, eggplants (aubergines), chili and green peppers, onions and green shallots, leeks
		and other alliaceous vegetables, green beans, leguminous vegetables nes, okra,
		mushrooms and truffles, artichokes, maize greens, asparagus, string beans, lettuce and
		chicory, cassava leaves
Fruit	0.02 (0.01-0.04)	Apples, pears, apricots, cherries, peaches and nectarines, plums and sloes, stone fruits
		nes, berries nes, grapes, tropical fresh fruit nes, fresh fruit nes, oranges, citrus fruit nes,
		figs, quinces, sour cherries, carobs, tangerines, mandarins, clementines, satsumas, lemons
		and limes, grapefruit (incl. pomelos), dates, bananas, pineapples, mangoes, mangosteens,
		guavas, strawberries, avocados, papayas, raspberries, currants, persimmons, kiwi fruit,
		gooseberries, plantains, cashewapple, blueberries, cranberries, pome fruits nes
Other	0.43 for pulses,	dry beans, dry peas, lentils, forage and silage (maize, grasses nes, alfalfa, clover,
crops	0.41 for nuts, 0.03	sorghum, green oilseeds, legumes, rye grass), forage products, vegetables and root
	for stimulants and	fodder, tobacco (unmanufactured), pulses nes, almonds with shells, walnuts with shells,
	spices, and 0.15 for	pistachios, nuts nes, anise, badian, fennel, coriander, broad beans, dry horse beans,
	others	vetches, chestnuts, hops, spices nes, chick peas, groundnuts with shells, beets for fodder,
		chilies and dry peppers, cocoa beans, coffee greens, lupins, tea, maté, peppermint, pigeon
		peas, natural rubber, Brazil nuts with shells, nutmeg mace, cardamoms, areca nuts,
		ginger, dry cow peas, bambara beans, kola nuts, hazelnuts with shells, pepper (Piper
		spp.), natural gums, cinnamon (canella), cloves, chicory roots, cabbage for fodder, teas
		nes, carrots for fodder, vanilla, dried pyrethrum, swedes for fodder, turnips for fodder

Table SI-3: P:N ratios in manure and the P contents of livestock and their products.

	Productio.			
Livestock and products	P:N ratio for livestock manure	P content of livestock and their		
		products (% w/w)		
Buffaloes	0.18 (0.13-0.24)	0.21		
Cattle, dairy	0.18 (0.13-0.24)	0.21		
Cattle, non-dairy	0.18 (0.13-0.24)	0.21		
Sheep	0.15 (0.09-0.23)	0.16		
Goats	0.15 (0.09-0.23)	0.16		
Swine, market	0.28 (0.23-0.35)	0.56		
Swine, breeding	0.28 (0.23-0.35)	0.56		
Chickens, layers	0.24 (0.13-0.35)	0.15		
Chickens, broilers	0.24 (0.13-0.35)	0.15		
Turkeys	0.25 (0.21-0.29)	0.15		
Horses	0.19 (0.18-0.21)	0.17		
Donkeys	0.19 (0.18-0.21)	0.17		
Mules	0.19 (0.18-0.22)	0.17		
Camels	0.19 (0.18-0.23)	0.17		
Ducks	0.25 (0.21-0.29)	0.15		
Llamas	0.19 (0.18-0.25)	0.17		
Eggs	-	0.26		
Milk	-	0.093		

#### Source:

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Table SI-4: Equations and data sources used in this study

Pool	Flow	Abbrev	Method	Period	Data Source	Parameters
		iation				
Phosphate	Phosphate acid	$P_{\mathrm{pa}}$	$P_{ m pa}=P_{ m P2O5}\% imes PA$	2002-2010	Phosphate acid production from IFA	P fraction of P in P <sub>2</sub> O <sub>5</sub>
	Fertilizer	$P_{ m fer}$	$P_{ ext{fer}} = P_{ ext{P2O5}}\%  imes Fer$	2002-2010	Fertilizer consumption from FAO	P fraction of P in P <sub>2</sub> O <sub>5</sub>
	Feed additives	$P_{ m feed}$	$P_{ ext{feed-add}} = 8\%  imes P_{ ext{pa}}$	2002-2010	_	_
		add				
	Detergent and other	$P_{ m det}$	$P_{ m det} = P_{ m pa}\!\!-\!\!P_{ m fer}\!\!-\!\!P_{ m feed ext{-}add}$	2002-2010	_	_
Atmosphere	Deposition to cropland	$P_{ m dep ext{-}crop}$	PKU-FUEL Model	2007	_	_
	Deposition to pasture	$P_{ m dep ext{-}}$	PKU-FUEL Model	2007	_	_
		grass				
	Cropland field burning	$P_{ m crop ext{-}bur}$	PKU-FUEL Model	2007	_	_
	Bioenergy emission	$P_{ m bioener}$	PKU-FUEL Model	2007	_	_
Cropland	Crop production	$P_{\mathrm{crop}}$	$P_{ ext{crop}} = Crop  imes P_{ ext{crop}} \%$	2002-2010	Crop production from FAO	P fraction of crops
	Crops as food	$P_{ m crop ext{-}}$	$P_{ ext{crop-food}} = Crop ext{-}Food  imes P_{ ext{crop}}\%$	2002-2010	Crop production as food from FAO	P fraction of crops
		food				
	Crops as feed	$P_{\mathrm{crop-}}$	$P_{ ext{crop-feed}} = Crop ext{-}Feed  imes P_{ ext{crop}}\%$	2002-2010	Crop production as feed from FAO	P fraction of crops
		feed				
	Crops as seed	$P_{ m crop ext{-}}$	$P_{ ext{crop-seed}} = Crop ext{-}Seed  imes P_{ ext{crop}}\%$	2002-2010	Crop production as seed from FAO	P fraction of crops
		seed				
	Crops as processing	$P_{ m crop ext{-}pro}$	$P_{\text{crop-pro}} = Crop\text{-}Processing \times P_{\text{crop}}\%$	2002-2010	Crop production as processing from	P fraction of crops
					FAO	
	Crops as waste	$P_{\mathrm{crop}}$	$P_{\text{crop-was}} = Crop\text{-}Waste \times P_{\text{crop}}\%$	2002-2010	Crop production as waste from FAO	P fraction of crops
		waste				
	Crops as other uses	$P_{ ext{crop-oth}}$	$P_{\text{crop-oth}} = Crop ext{-}Other\ Use  imes P_{ ext{crop}}\%$	2002-2010	Crop production as other use from FAO	P fraction of crops

	Crop residues recycled	$P_{\mathrm{res-re}}t$	$P_{\text{res-ret}} = Residues\text{-}Return \times P_{\text{crop}}\%$	2002-2010	Crop residues production returned to	P fraction of crop residues
	to cropland				cropland from FAO	
	Total crop residues	$P_{\text{crop-res}}$	$P_{\text{crop-res}} = P_{\text{res-ret}} / 50\%$	2002-2010	_	-
	Crop residues as feed	$P_{ m res-feed}$	$P_{ ext{res-feed}} = P_{ ext{crop-res}}  imes 25\%$	2002-2010	_	-
	Crop residues to human	$P_{\mathrm{res-hum}}$	$P_{\text{res-hum}} = P_{\text{crop-res}} - P_{\text{res-ret}} - P_{\text{res-feed}} - P_{\text{crop-}}$	2002-2010	_	-
			bur			
	Cropland runoff	$P_{ m run ext{-}crop}$	$P_{\text{run-crop}} = 12.5\% \times (P_{\text{fer-crop}} + P_{\text{dep-crop}} +$	2002-2010	_	-
			$P_{ m livman-crop} + P_{ m man-hum} + P_{ m res-ret})$			
Pasture	Grass as feed	$P_{ m grass}$	ORCHIDEE Model	2002-2010	_	P fraction of grass and forage
	Pasture runoff	$P_{ m run}$ -	$P_{\text{run-crop}} = 12.5\% \times (P_{\text{fer-pas}} + P_{\text{dep-pas}} +$	2002-2010	_	-
		grass	$P_{ m livman-pas})$			
Livestock	Manure to cropland	$P_{ m manliv}$	$P_{ m manliv-crop} = N_{ m Manure-Crop} \ { m P}\%/N\%$	2002-2010	Livestock manure production to cropland	P fraction of livestock
		crop			from FAO	manure to CROPLAND
	Manure to pasture	$P_{ m manliv}$	$P_{ ext{manliv-pas}} = N_{ ext{Manure-grass}} \times P\%/N\%$	2002-2010	Livestock manure production to pasture	P fraction of livestock
		grass			from FAO	manure to pasture
	Manure as waste	$P_{man-}$	$P_{ ext{manliv-crop}} = N_{ ext{Manure-waste}}  imes P\%/N\%$	2002-2010	Livestock manure production as wastes	P fraction of livestock
		waste			from FAO	manure as waste
	Meat	$P_{ m meat}$	$P_{\mathrm{meat}} = Meat \times P_{\mathrm{meat}}\%$	2002-2010	Meat production from FAO	P fraction of meat
	Eggs	$P_{ m egg}$	$P_{ ext{egg}} = Eggs  imes P_{ ext{egg}}\%$	2002-2010	Egg production from FAO	P fraction of egg
	Milk	$P_{ m milk}$	$P_{ m milk} = Milk  imes P_{ m milk} \%$	2002-2010	Milk production from FAO	P fraction of milk
	Feed from human food	$P_{ m feed}$	$P_{\text{pro-feed}} = (P_{\text{meat}} + P_{\text{egg}} + P_{\text{milk}} + P_{\text{manliv-crop}})$	2002-2010	_	-
	waste		$+P_{ m manliv-gpas}+P_{ m man-pas})$			
			$-\left(P_{ ext{fee-add}}+P_{ ext{res-feed}}+P_{ ext{crop-feed}}+P_{ ext{grass}}+ ight.$			
			$P_{ m for})$			
Humans	Human excreta as	$P_{\mathrm{man-}}$	$P_{\mathrm{man-hum}} = Excreta\text{-}Human \times (70\% \times$	2002-2010	Rural and urban population from FAO	P fraction of human excreta,
	manure to crops	hum	$Population_{ m rural}$			human excreta production

 $+30\% \times Population_{urban}$ )

Human excreta as	$P_{ m exchum}$	$P_{\mathrm{exchum-waste}} = Excreta-Human \times (30\% \times$	2002–2010	_	_	
manure wasted	waste	$Population_{ m rural}$				
		$+70\% \times Population_{urban}$ )				
Waste from humans	$P_{ m waste-}$	$P_{\text{waste-hum}} = [(P_{\text{crop}} - P_{\text{crop-seed}}) + (P_{\text{meat}} + P_{\text{meat}})]$	2002–2010	_	-	
	hum	$P_{ m egg} + P_{ m milk}) + P_{ m det}] - P_{ m man-hum} - P_{ m bioener}$				

#### 413 Source:

- 414 1. IFA, http://www.fertilizer.org/Statistics
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- 425 9. MWPS-18 (1985) Livestock Waste Facilities Handbook. Midwest Plan Service, University of Missouri, Ames, IA, USA, 112 pp.
- 426 10. OECD Secretariat (1991) National Soil Surface Nutrient Balances, 1985 to 1995. Explanatory Notes. Table 2 Coefficients to convert livestock numbers into manure nitrogen quantities from
- ational sources. Organisation for Economic Cooperation and Development, Paris.
- 428 11. USDA-NRCS (2009) Crop Nutrient Tool: Nutrient Content of Crops. United States Department of Agriculture, Natural Resource Conservation Service, Washington.
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- 430 13. Wang R, Balkanski Y, Boucher O et al. (2015) Significant contribution of combustion-related emissions to the atmospheric phosphorus budget. Nature Geoscience, 8, 48-54.
- 431 14. Wang R, Tao S, Balkanski Y et al. (2014) Exposure to ambient black carbon derived from a unique inventory and high resolution model. Proceedings of the National Academy of Sciences
- 432 of the United States of America, 111, 2459-2463.

Table SI-5: Ranges of cropland P fluxes used in the uncertainty analysis and for comparison with earlier studies.

		Total	output (Tg P	yr <sup>-1</sup> )			
Fertilizer inputs	Livestock manure to cropland	Human sewage sludge to cropland	Recycled crop residues to cropland	Deposition	Harvested crops	Harvested crop residues	Leachin g or runoff
13.7–15.0	6.0-8.0	1.3–1.5	1.0-3.5	0.6–1.0	8.2-12.3	3.8-6.7	3.2-4.0

Note: For Research, the global flows and budget was in the year 2000.

Source: 1. Liu et al., 2008; 2. Smil, 2000; 3. Cordell et al., 2009; 4. MacDonald et al., 2011; 5. Bouwman et al., 2009; 6. Bouwman et al., 2011.

Table SI-6: Cropland total PUE and PUE (excluding residues for cropland and manure for livestock) at the global and regional levels.

	Cropland			Livestock			
	PUE	Total PUE	Soil P balance	Total	Livestock	PUE	Livestock total PUE
				PUE:			
				PUE			
				ratio			
World	0.46	0.76	4.68	1.65	0.06		0.83
Eastern and Southern Africa	0.80	1.26	-1.03	1.58	0.02		0.92
Northern Africa	0.84	1.48	-1.48	1.76	0.02		0.95
Western and Central Africa	1.51	2.28	-2.72	1.51	0.01		0.92
Eastern Asia	0.27	0.44	23.45	1.63	0.08		0.81
Southern and Southeastern Asia	0.43	0.77	4.14	1.79	0.05		0.78
Western and Central Asia	0.64	1.09	0.22	1.70	0.04		0.69
Oceania	0.31	0.51	5.17	1.65	0.04		0.93
Europe	0.54	0.88	2.78	1.63	0.09		0.78
North America	0.57	0.99	1.46	1.74	0.08		0.89
Caribbean and Central America	0.53	0.69	3.79	1.30	0.03		0.78
South America	0.63	0.88	2.25	1.40	0.03		0.84

Table SI-7: Cropland total PUE and cropland PUE (excluding residues) for different crops

		Cropland tota PUE	Croplandl PUE	Cropland PUE: cropland total PUE ratio
Cereals	Wheat	0.55	1.06	1.93
	Rice	0.33	0.90	2.73
	Maize	0.36	0.90	2.50
	Other cereals	0.70	1.43	2.04
Oil crops	Soybean	0.73	0.96	1.32
	Oil palm	0.24	0.24	1.0
	Other oil crops	0.60	0.60	1.0
Sug	gar crops	0.83	0.83	1.0
	Fiber	0.19	0.19	1.0
Roots	and tubers	0.54	0.69	1.28
	Fruits	0.10	0.10	1.0
Vegetables		0.25	0.28	1.12
Otl	ner crops	0.52	0.52	1.0

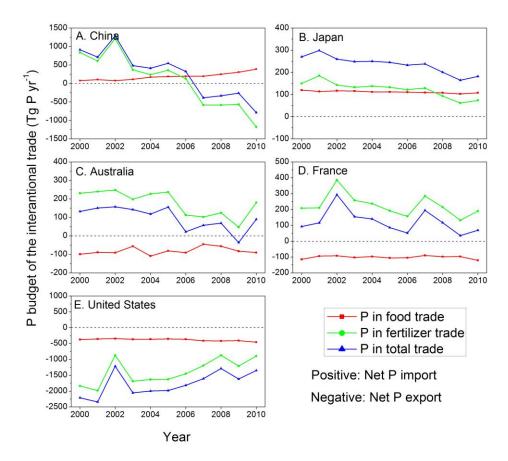


Figure SI-1: Flows of P in international trade for (A) China, (B) Japan, (C) Australia, (D) France, and (E) the United States from 2000 to 2010. Positive values represent net imports; negative values represent net exports.

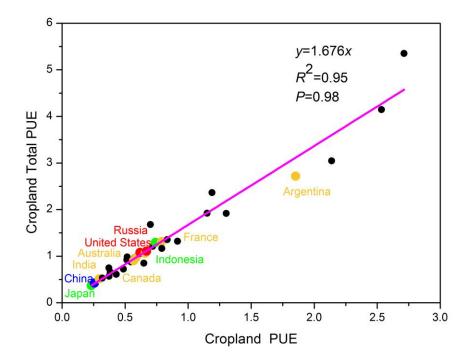


Figure SI-2: The relationship between cropland total PUE (harvested crops + residues) and cropland PUE (harvested crops, excluding residues) for 35 large countries.

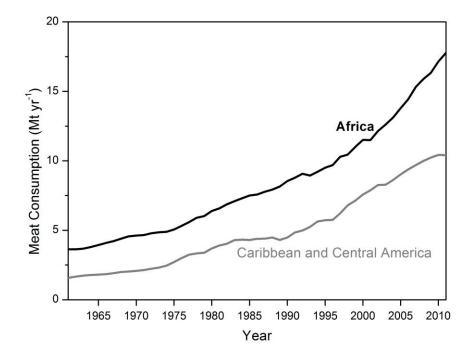


Figure SI-3: Changes in meat consumption in Africa and in the Caribbean and Central America region between 1961 and 2011