Supporting Information

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Text S1. Analytical characterization of the categories defined in Harpole et al. (2011).

First, we defined the function $f$ as $f: x \rightarrow \min(1, x)$. By definition, the N and P limitations encountered in the control experiment, $R_N$ and $R_P$, were defined as $R_N = f\left(\frac{S_N}{D_N}\right)$ and $R_P = f\left(\frac{S_P}{D_P}\right)$. As explained in the Main Text (Fig. 1 and Eq.3-6), the different experiments ($E_1$=control, $E_2$, $E_3$, $E_4$) were defined by:

$E_1$: $R_N(E_1) = f\left(\frac{S_N}{D_N}\right)$ and $R_P(E_1) = f\left(\frac{S_P}{D_P}\right)$. $R_N(E_1)$ and $R_P(E_1)$ are called $R_N$ and $R_P$ (as in the Main Text).

$E_2$: $R_N(E_2) = f\left(\frac{S_N + A_N}{D_N}\right)$ and $R_P(E_2) = f\left(\frac{S_P}{D_P}\right)$

$E_3$: $R_N(E_3) = f\left(\frac{S_N}{D_N}\right)$ and $R_P(E_3) = f\left(\frac{S_P + A_P}{D_P}\right)$

$E_4$: $R_N(E_4) = f\left(\frac{S_N + A_N}{D_N}\right)$ and $R_P(E_4) = f\left(\frac{S_P + A_P}{D_P}\right)$.

In the following sub-sections (Text S1.1 and Text S1.2), for each formalism (MH or LM) respectively, we combined the above experiment definitions with Eq.9-11 (Main Text) to express $\Delta \text{pro}_{+P}$, $\Delta \text{pro}_{+N}$ and $\Delta \text{pro}_{+NP}$ as functions of the N and P limitations encountered in the different experiments (i.e. $R_N(E_i)$ and $R_P(E_i)$ with $i$ in [1,4]).

Because each category of Harpole et al. (2011) could be defined as function of i) the character null or non-null of $\Delta \text{pro}_{+N}$ and $\Delta \text{pro}_{+P}$ and ii) the relationship between $\Delta \text{pro}_{+NP}$ and ($\Delta \text{pro}_{+N}+\Delta \text{pro}_{+P}$) (column 4 of Table 1), we then characterized each category in terms of the N and P limitations encountered in the different experiments (and if possible, only with the nutrient limitations in the control, i.e. $R_N$ and $R_P$).

Preamble

In the following, $A_N$ and $A_P$ are positive and non-null. By construction,

$$f\left(\frac{S_X + A_X}{D_X}\right) \geq f\left(\frac{S_X}{D_X}\right) \quad \text{(Eq.S1)}$$

with $X=$N or P. Equality is only possible when $f\left(\frac{S_X}{D_X}\right)=1$, i.e. $R_X=1$. Otherwise,

$$f\left(\frac{S_X + A_X}{D_X}\right) > f\left(\frac{S_X}{D_X}\right).$$

Thus, we have the two equivalences below:

$$[f\left(\frac{S_X + A_X}{D_X}\right) > f\left(\frac{S_X}{D_X}\right)] \Rightarrow [R_X < 1] \quad \text{(Ev.S1)}$$

$$[f\left(\frac{S_X + A_X}{D_X}\right) = f\left(\frac{S_X}{D_X}\right)] \Rightarrow [R_X = 1] \quad \text{(Ev.S2)}$$

Note that the above equivalences imply that $f\left(\frac{S_X + A_X}{D_X}\right)$ is necessarily non-null.
Text S1.1. MH formalism

With MH formalism (i.e. with Eq.7), Eq.9-11 become:

$$\Delta \text{ pro}_{+N} = f(\frac{S_N+A_N}{D_N}) \cdot f(\frac{S_p}{D_p}) - f(\frac{S_N}{D_N}) \cdot f(\frac{S_p}{D_p})$$ (Eq.S2)

$$\Delta \text{ pro}_{+p} = f(\frac{S_N}{D_N}) \cdot f(\frac{S_p+A_p}{D_p}) - f(\frac{S_N}{D_N}) \cdot f(\frac{S_p}{D_p})$$ (Eq.S3)

$$\Delta \text{ pro}_{+NP} = f(\frac{S_N+A_N}{D_N}) \cdot f(\frac{S_p+A_p}{D_p}) - f(\frac{S_N}{D_N}) \cdot f(\frac{S_p}{D_p})$$ (Eq.S4).

Thanks to Eq.S2, we find that $\Delta \text{ pro}_{+N}=0$ is only possible if either $f(\frac{S_p}{D_p})=0$ or $f(\frac{S_N+A_N}{D_N})=f(\frac{S_N}{D_N})$. The first condition corresponds to $R_p=0$, the 2nd one to $R_N=1$ (see Ev.S2). Thus,

$$(\Delta \text{ pro}_{+N}=0) \\iff (R_p=0 \text{ or } R_N=1) \quad (\text{Ev.S3})$$

And similarly,

$$(\Delta \text{ pro}_{+p}=0) \\iff (R_N=0 \text{ or } R_p=1) \quad (\text{Ev.S4}).$$

Also we have to note that $R_N$ or $R_p$ equal to 1 have some implications. From Ev.S2, we have $[R_N=1] \\Rightarrow [f(\frac{S_N+A_N}{D_N})=f(\frac{S_N}{D_N})=1]$. In that case, Eq.S2-S4 becomes $\Delta \text{ pro}_{+N}=0$, $\Delta \text{ pro}_{+p}=f(\frac{S_p+A_p}{D_p})-f(\frac{S_p}{D_p})$ and $\Delta \text{ pro}_{+NP}=f(\frac{S_p+A_p}{D_p})-f(\frac{S_p}{D_p})$. Thus, we get

$$\Delta \text{ pro}_{+NP}=\Delta \text{ pro}_{+N}+\Delta \text{ pro}_{+p}.$$ Thus,

$$(R_N=1) \Rightarrow (\Delta \text{ pro}_{+NP}=\Delta \text{ pro}_{+N}+\Delta \text{ pro}_{+p}).$$ Similarly,

$$(R_p=1) \Rightarrow (\Delta \text{ pro}_{+NP}=\Delta \text{ pro}_{+N}+\Delta \text{ pro}_{+p}).$$ Further, we can even demonstrate (Text S2) that:

$$(R_p=1 \text{ or } R_N=1) \iff (\Delta \text{ pro}_{+NP}=\Delta \text{ pro}_{+N}+\Delta \text{ pro}_{+p}) \quad (\text{Ev.S5}).$$

The use of Ev.S3-5 and the definition of the different categories (column 4 in Table 1) allow us to characterize each category in terms of value for $R_N$ and $R_p$. This is demonstrated for category A below for instance, and for all categories in Text S8. Category A is characterized by:

$$\Delta \text{ pro}_{+N}=0 \quad (x)$$

$$\Delta \text{ pro}_{+p}=0 \quad (y)$$

$$\Delta \text{ pro}_{+NP} \geq \Delta \text{ pro}_{+N} + \Delta \text{ pro}_{+p} \quad (z)$$

According to Ev.S3, $(x) \iff (R_p=0 \text{ or } R_N=1)$. $R_N$ cannot be equal to 1 otherwise $\Delta \text{ pro}_{+NP}=\Delta \text{ pro}_{+N}+\Delta \text{ pro}_{+p}$. Similarly, $(y) \iff (R_N=0 \text{ or } R_p=1)$ and $R_p$ cannot be equal to 1 otherwise $\Delta \text{ pro}_{+NP}=\Delta \text{ pro}_{+N}+\Delta \text{ pro}_{+p}$. The only combination possible is: $R_p=R_N=0$.

Values possible for $R_p$ and $R_N$ for a given ecosystem and their implication on the category defined by Harpole et al. (2011) are summarized in Table S4. Table S4 was used to build the column 5 of Table 1.
In Harpole et al. (2011), category B encompasses different cases: sub-additive, additive and super-additive. Sub-additive and additive cases are not synergistic, i.e. they are characterized by \((\Delta \text{pro}_{NP}) \leq (\Delta \text{pro}_{N} + \Delta \text{pro}_{P})\). With MH formalism, because of Ev.S3, \((\Delta \text{pro}_{NP} \neq 0)\) as found in category B implies that \((R_p \neq 0 \text{ and } R_N \neq 1)\). Similarly, \((R_N \neq 0 \text{ and } R_P \neq 1)\). Because \((R_p = 1 \text{ or } R_N = 1) \iff (\Delta \text{pro}_{NP} = \Delta \text{pro}_{N} + \Delta \text{pro}_{P})\) (Ev.S5), it means that \((\Delta \text{pro}_{NP}) > (\Delta \text{pro}_{N} + \Delta \text{pro}_{P})\) necessarily happens in category B. Thus, only super-additive cases can be considered in category B with the MH formalism.

**Text S1.2. LM formalism**

With LM formalism (i.e. with Eq.8), Eq.9-11 become:

\[
\Delta \text{pro}_{N} = \min[f\left(\frac{S_N + A_N}{D_N}\right), f\left(\frac{S_P}{D_P}\right)] - \min[f\left(\frac{S_N}{D_N}\right), f\left(\frac{S_P}{D_P}\right)] \quad \text{(Eq.6)}
\]

\[
\Delta \text{pro}_{P} = \min[f\left(\frac{S_N}{D_N}\right), f\left(\frac{S_P + A_P}{D_P}\right)] - \min[f\left(\frac{S_N}{D_N}\right), f\left(\frac{S_P}{D_P}\right)] \quad \text{(Eq.6)}
\]

\[
\Delta \text{pro}_{NP} = \min[f\left(\frac{S_N + A_N}{D_N}\right), f\left(\frac{S_P + A_P}{D_P}\right)] - \min[f\left(\frac{S_N}{D_N}\right), f\left(\frac{S_P}{D_P}\right)] \quad \text{(Eq.7)}
\]

The above equations can be solved if we know how the different ratios involved \((S_N\frac{D_N}{D_P}, \frac{S_P}{D_P}, \frac{S_N + A_N}{D_N}, \frac{S_P + A_P}{D_P})\) and 1 are ranked. In the following, we define the conditions in terms of \(R_N\) and \(R_P\) encountered in the different experiments that are required to be in each category defined by Harpole et al. (2011).

In both categories C and E, the ecosystem is N-limited in the control \((E_1)\): adding N leads to an increase in the productivity (from \(E_1\) to \(E_2\)). Because adding P does not change the productivity, the ecosystem in \(E_1\) is not P-limited. In fact, except in some very specific cases, the ecosystem is mono nutrient-limited with the LM formalism. As the ecosystem is N limited in the control \((E_1)\), we have \(f\left(\frac{S_N}{D_N}\right) < f\left(\frac{S_P}{D_P}\right)\). Because of (Eq.S1), we also have:

\[
f\left(\frac{S_N}{D_N}\right) < f\left(\frac{S_P}{D_P}\right) \leq f\left(\frac{S_P + A_P}{D_P}\right) \quad \text{(Eq.8)}
\]

Eq.S5-S7 becomes:

\[
\Delta \text{pro}_{N} = \min[f\left(\frac{S_N + A_N}{D_N}\right), f\left(\frac{S_P}{D_P}\right)] - f\left(\frac{S_N}{D_N}\right) \quad \text{(Eq.9)}
\]

\[
\Delta \text{pro}_{P} = f\left(\frac{S_N}{D_N}\right) - f\left(\frac{S_N}{D_N}\right) = 0 \quad \text{(Eq.10)}
\]

\[
\Delta \text{pro}_{NP} = \min[f\left(\frac{S_N + A_N}{D_N}\right), f\left(\frac{S_P + A_P}{D_P}\right)] - f\left(\frac{S_N}{D_N}\right) \quad \text{(Eq.11)}
\]

Eq.S10 means that adding P does not modify the N-limitation and \(E_3\) is also N-limited. To go further and to distinguish categories C and E, we have to consider the different cases of nutrient limitation in \(E_2\).
If $E_2$ is N only-limited, we have $f\left(\frac{S_N+A_N}{D_N}\right)<f\left(\frac{S_P}{D_P}\right)$. Because of Eq. S1 applied to N, we get $f\left(\frac{S_N+A_N}{D_N}\right)<f\left(\frac{S_P}{D_P}\right)\leq f\left(\frac{S_P+A_P}{D_P}\right)$. Eq. S9 & S11 becomes $\Delta \text{pro}_N=f\left(\frac{S_N+A_N}{D_N}\right)-f\left(\frac{S_N}{D_N}\right)$ and $\Delta \text{pro}_{NP}=f\left(\frac{S_N+A_N}{D_N}\right)-f\left(\frac{S_N}{D_N}\right)$. Thus, $\Delta \text{pro}_{NP}=\Delta \text{pro}_N+\Delta \text{pro}_P$. We are in the case corresponding to category E.

If $E_2$ is only P-limited, we have:

$$f\left(\frac{S_P}{D_P}\right)<f\left(\frac{S_N+A_N}{D_N}\right) \quad (\text{Eq. S12})$$

and Eq. S9 becomes $\Delta \text{pro}_N=f\left(\frac{S_P}{D_P}\right)-f\left(\frac{S_N}{D_N}\right)$. Because $E_2$ is P-limited, we also have $f\left(\frac{S_P}{D_P}\right)<1$ and thus, following Eq. S1, we have:

$$f\left(\frac{S_P}{D_P}\right)<f\left(\frac{S_P+A_P}{D_P}\right) \quad (\text{Eq. S13}).$$

To compute $\Delta \text{pro}_{NP}$, we have to consider the different limitations that could occur in $E_4$. If $E_4$ is only P-limited, we have $f\left(\frac{S_P+A_P}{D_P}\right)<f\left(\frac{S_N+A_N}{D_N}\right)$. If $E_4$ is only N-limited, we have $f\left(\frac{S_N+A_N}{D_N}\right)<f\left(\frac{S_P+A_P}{D_P}\right)$. If $E_4$ is N and P limited or non-limited, we have $f\left(\frac{S_N+A_N}{D_N}\right)=f\left(\frac{S_P+A_P}{D_P}\right)$. In all cases, we can use Eq. S12 or Eq. S13 to show that $\min\left[f\left(\frac{S_N+A_N}{D_N}\right), f\left(\frac{S_P+A_P}{D_P}\right)\right]>f\left(\frac{S_P}{D_P}\right)$. Thus, $\Delta \text{pro}_{NP}>f\left(\frac{S_P}{D_P}\right)-f\left(\frac{S_N}{D_N}\right)$, i.e. $\Delta \text{pro}_{NP} > \Delta \text{pro}_N + \Delta \text{pro}_P$. We are in the case corresponding to category C.

If $E_2$ is both N and P-limited, we have:

$$f\left(\frac{S_P}{D_P}\right)=f\left(\frac{S_N+A_N}{D_N}\right) \quad (\text{Eq. S14}).$$

Thus, Eq. S9 becomes e.g. $\Delta \text{pro}_N=f\left(\frac{S_P}{D_P}\right)-f\left(\frac{S_N}{D_N}\right)$ and Eq. S11 becomes $\Delta \text{pro}_{NP}=\min\left[f\left(\frac{S_P}{D_P}\right), f\left(\frac{S_P+A_P}{D_P}\right)\right]-f\left(\frac{S_N}{D_N}\right)$. Because of Eq. S1, the latter equation is equivalent to $\Delta \text{pro}_{NP}=f\left(\frac{S_P}{D_P}\right)-f\left(\frac{S_N}{D_N}\right)$ and thus, $\Delta \text{pro}_{NP}=\Delta \text{pro}_N + \Delta \text{pro}_P$. We are in the case corresponding to category E. Note also that, because there is a P limitation in $E_2$, Eq. S13 also applies here. And thus, $f\left(\frac{S_P+A_P}{D_P}\right)>f\left(\frac{S_N+A_N}{D_N}\right)$: $E_4$ is necessarily N-limited.
If $E_2$ is not limited, we have $f\left(\frac{S_P}{D_P}\right)=f\left(\frac{S_N+A_N}{D_N}\right)=1$. In that case, because of Eq.S1, we also have $f\left(\frac{S_P+A_P}{D_P}\right)=1$ (it means that $E_1$ is not limited). Eq.S9&S11 becomes

$$\Delta \text{pro}_{+N}=1-f\left(\frac{S_N}{D_N}\right)\quad \text{and} \quad \Delta \text{pro}_{+NP}=1-f\left(\frac{S_N}{D_N}\right),$$

respectively. Thus,

$$\Delta \text{pro}_{+NP}=\Delta \text{pro}_{+N}+\Delta \text{pro}_{+P}. \quad \text{We are in the case corresponding to category E.}$$

To summarize, category C corresponds to: $E_1$ N-limited and $E_2$ P-limited, i.e. the addition of N alone (+N) switches the ecosystem from N-limitation to P-limitation. Category E corresponds to $E_1$ N-limited and $E_2$ either N-limited or NP-limited or not limited at all.

Expressed with equations, we have:

- category C $\iff [R_P<R_N \quad \text{and} \quad R_P(E_2)<R_N(E_2)]$
- category E $\iff [R_N<R_P \quad \text{and} \quad R_P(E_2)\geq R_N(E_2)]$

The same reasoning applies to categories D (E1 P-limited and E3 N-limited) and F (E1 P-limited and E3 either P-limited or NP-limited or not limited at all).

Category A is characterized by $E_1$ both limited by N and P, thus: $R_P=R_N\neq 1$.

Category G is characterized by $E_1$ neither N-limited nor P-limited, thus, $R_P=R_N=1$.

Category B is defined by $\Delta \text{pro}_{+N} \neq 0$. Thus, $E_1$ is N-limited, i.e. $R_N\leq R_P$. Because $\Delta \text{pro}_{+N} \neq 0$, $E_1$ is also P-limited and we have $R_P\leq R_N$. Thus, $R_P=R_N$. Because $E_1$ is limited, the ratios are different from 1, i.e. $R_P=R_N\neq 1$. This implies that Eq.S5 could be written as follow:

$$\Delta \text{pro}_{+N}=\min[f\left(\frac{S_N+A_N}{D_N}\right),f\left(\frac{S_P}{D_P}\right)]-\min[f\left(\frac{S_N}{D_N}\right),f\left(\frac{S_P}{D_P}\right)] \quad (\text{Eq.S5})$$

$$\Delta \text{pro}_{+N}=\min[f\left(\frac{S_N+A_N}{D_N}\right),f\left(\frac{S_N}{D_N}\right)]-f\left(\frac{S_N}{D_N}\right) \quad (\text{because } R_P=R_N)$$

$$\Delta \text{pro}_{+N}=f\left(\frac{S_N}{D_N}\right)-f\left(\frac{S_N}{D_N}\right) \quad (\text{because } R_N\neq 1 \text{ and Ev.S1}).$$

Thus, $\Delta \text{pro}_{+N}=0$ which is contrary to the definition of category B: category B cannot occur with LM.

The above results are summarized in Table 1.
Text S2. Demonstration of \( (R_p=1 \; \text{or} \; R_N=1) \Rightarrow (\Delta \text{pro}_{+NP}=\Delta \text{pro}_{+N}+\Delta \text{pro}_{+P}) \) with the MH formalism

First we demonstrate that \( (R_p=1 \; \text{or} \; R_N=1) \Rightarrow (\Delta \text{pro}_{+NP}=\Delta \text{pro}_{+N}+\Delta \text{pro}_{+P}) \) by contradiction (\textit{reductio ad absurdum}):

By using definition of \( \Delta \text{pro}_{+NP} \), \( \Delta \text{pro}_{+N} \) and \( \Delta \text{pro}_{+P} \) and after simplification,

\[ \Delta \text{pro}_{+NP}>\Delta \text{pro}_{+N}+\Delta \text{pro}_{+P} \] implies that

\[ f\left(\frac{S_N+A_N}{D_N}\right)-f\left(\frac{S_N}{D_N}\right) \cdot [f\left(\frac{S_P+A_P}{D_P}\right)-f\left(\frac{S_P}{D_P}\right)]>0 \] .

Thus, because \( [f\left(\frac{S_N+A_N}{D_N}\right)-f\left(\frac{S_N}{D_N}\right)] \) and \( [f\left(\frac{S_P+A_P}{D_P}\right)-f\left(\frac{S_P}{D_P}\right)] \) cannot be negative,

\[ \Delta \text{pro}_{+NP}>\Delta \text{pro}_{+N}+\Delta \text{pro}_{+P} \] implies that

\[ \begin{cases} f\left(\frac{S_N+A_N}{D_N}\right)-f\left(\frac{S_N}{D_N}\right)>0 \\ f\left(\frac{S_P+A_P}{D_P}\right)-f\left(\frac{S_P}{D_P}\right)>0 \end{cases} \] .

Because Ev.S1, it means that \( (\Delta \text{pro}_{+NP}>\Delta \text{pro}_{+N}+\Delta \text{pro}_{+P}) \Rightarrow R_N \neq 1 \)

\[ R_p \neq 1 \] .

Because we have already shown in Text S1.1 that \( (R_N=1) \Rightarrow (\Delta \text{pro}_{+NP}=\Delta \text{pro}_{+N}+\Delta \text{pro}_{+P}) \) and \( (R_P=1) \Rightarrow (\Delta \text{pro}_{+NP}=\Delta \text{pro}_{+N}+\Delta \text{pro}_{+P}) \) , we finally get:

\( (R_p=1 \; \text{or} \; R_N=1) \Leftrightarrow (\Delta \text{pro}_{+NP}=\Delta \text{pro}_{+N}+\Delta \text{pro}_{+P}) \) .
Text S3. Computation of the nutrient demand ($D_N$ and $D_P$)

As an example, we focus here on the computation of the demand for P ($D_P$). The Harvest index ($HI$, dimensionless), P harvest index ($PHI$, dimensionless), root/shoot ratio ($RSR$, dimensionless), P concentration of a plant organ $i$ ($P_{\%i}$ in gP/gC) and yield ($Y$, in gC) are defined as follows:

$$HI = \frac{C_{\text{grain}}}{C_{\text{shoot}}}$$  \hspace{1cm} (1)

$$PHI = \frac{P_{\text{grain}}}{P_{\text{shoot}}}$$  \hspace{1cm} (2)

$$RSR = \frac{C_{\text{root}}}{C_{\text{shoot}}}$$  \hspace{1cm} (3)

$$P_{\%i} = \frac{P_i}{C_i}$$  \hspace{1cm} (4)

$$Y = C_{\text{grain}}$$  \hspace{1cm} (5)

where $C_i$: the carbon content of organ $i$ (in gC), $P_i$: the P content of organ $i$ (in gP), and the shoot is defined as (grain + leaf + stem).

We aimed to estimate the P demand, that is approached by the sum of P in the shoot and P in the root at maturity: $D_P = P_{\text{shoot}} + P_{\text{root}}$.

By using (2), (4) (applied to $i=\text{grain}$) and (5), we have:

$P_{\text{shoot}} = P_{\%\text{, grain}} \cdot \frac{Y}{PHI}$.

By using (4) (applied to $i=\text{root}$) and (3), we get:

$P_{\text{root}} = P_{\%\text{, root}} \cdot RSR \cdot C_{\text{shoot}}$, then:

$P_{\text{root}} = P_{\%\text{, root}} \cdot RSR \cdot \frac{Y}{HI}$ by using (1) and (5).

Finally, we get: $D_P = P_{\%\text{, grain}} \cdot \frac{Y}{PHI} + P_{\%\text{, root}} \cdot RSR \cdot \frac{Y}{HI}$. Similarly for N, we found: $D_N = N_{\%\text{, grain}} \cdot \frac{Y}{HI} + N_{\%\text{, root}} \cdot RSR \cdot \frac{Y}{HI}$. The potential yield ($Y_{pot}$) is used to compute the potential demands in N and P ($D_N$ and $D_P$, respectively). These equations correspond to Eq.5 given in the Main Text.

The spatially explicit potential yield ($Y_{pot}$) for maize, wheat and rice is provided by Mueller et al. (2012) while the constants $P_{\%\text{, grain}}$, $P_{\%\text{, root}}$, $N_{\%\text{, grain}}$, $N_{\%\text{, root}}$, RSR, PHI, NHI and HI are found in the literature (see Table S2).
Text S4. Computation of the potential P uptake

Following some assumptions (in particular, that P concentration at the root surface reaches zero and the uptake of P is the same as the rate at which it diffuses there (Kvakić et al., 2018; Mollier et al., 2008)), the potential P uptake is given by:

\[ P_{\text{uptake}} = \sum_{m=1}^{12} \pi \Delta z . L_{rv,i}(m) . D . \frac{\rho^2 - 1}{G(\rho)} . C_p \]  
(Eq.S15)

where \( m \) is the month, \( \Delta z \) is soil depth (cm), \( L_{rv} \) is the monthly root length density (cm/cm\(^3\)), \( D \) is the coefficient of P diffusion (cm\(^2\)/d), \( C_p \) is the mean concentration of orthophosphate ions in the soil solution in the top 0.0–0.3m (mgP/L), \( \rho \) is a dimensionless ratio of soil cylinder to root radius, and \( G(\rho) \) is a dimensionless geometric function related to uptake by diffusion only. \( P_{\text{uptake}} \) is then converted in kgP/ha/yr. \( C_p \) was derived from inorganic labile P provided by Ringeval et al. (2017). The inorganic labile P was winsorized to 0.01% to prevent outliers in the soil P distribution that bias the global average (see the distribution in Fig. S3). This results in the prescription of the value of 2 grid-cells for each simulation out of the 1000 replicates (see Text S5). \( \Delta z \) is equal to 30cm as considered in Ringeval et al. (2017). Further details and the values of some parameters can be found in Kvakić et al. (2018).

The monthly root length density is computed as follows:

\[ L_{rv}(m) = \frac{C_{\text{root}}(m) \times SRL}{\Delta z} \]

where \( C_{\text{root}} \) is the carbon (C) in root biomass, SRL is the specific root length (m/g), and \( \Delta z=30 \)cm. The following values were used for SRL: 74 (wheat), 100 (maize) and 146 m/g (rice) as used in Kvakić et al. (2018).

\( C_{\text{root}} \) is computed as follows:

\[ C_{\text{root}}(m) = \frac{C_{\text{root};LPJmL}(m)}{C_{\text{root};LPJmL;max}} \times \frac{RSR}{HI} \times Y_{\text{pot}} \]

where \( Y_{\text{pot}} \) is the potential yield provided by Mueller et al. (2012) (kgC/ha/yr), \( C_{\text{root};LPJmL} \) is the average monthly root biomass simulated by LPJmL (kgC/ha/yr) and \( C_{\text{root};LPJmL;max} \) is the yearly maximum of \( C_{\text{root};LPJmL} \) (kgC/ha/yr). HI and RSR are the harvest index and root/shoot ratio (dimensionless) and are described in Table S2. The ratio \( \frac{C_{\text{root};LPJmL}(m)}{C_{\text{root};LPJmL;max}} \) varies between 0 and 1 and allows the description of seasonality in root biomass.

LPJmL (von Bloh et al., 2018) is one the Global Gridded Crop Models (GGCM) participating in a recent intercomparison (Elliott et al., 2015). Because of the divergence in simulated potential yields between GGCMs and the mismatch between the GGCMs and potential yield given by Mueller et al. (2012) (used in particular in our approach to compute the nutrient demand) (not shown), we chose to keep only the seasonality simulated by one GGCM instead of using the simulated root biomass directly. This allows consistency between computation of nutrient (N and P) demands and the P supply. The LPJmL simulation used to provide \( C_{\text{root};LPJmL} \) and \( C_{\text{root};LPJmL;max} \). In the above equation was performed by assuming the absence of nutrient limitation (called “harm-suffN” in Müller et al. (2017) and “harmnon” in Elliott et al. (2015)) and irrigated conditions following the protocol of the GGCM intercomparison. LPJmL considered spring and winter wheat and here we used the most productive one if both were simulated in the same grid-cell.
Text S5. Global values and uncertainty

We took an uncertainty associated with the supply and demand variables into account. To do this, we computed 1000 replicates for each variable by considering different sources of uncertainty (Table S1). This uncertainty was then propagated to $R_N$, $R_P$ and $R_{NP}$ and 1000 replicates were considered for each ratio. These replicates were used to compute an average and a standard-deviation for each grid-cell, and were plotted as 2D maps in the Main Text and Supporting Figures. Two values are given to provide information at the global scale: the average and the standard-deviation of the 1000 global averages. Each global average is computed by using the grid-cell crop area (Ramankutty et al., 2008) as weight.
At the global scale, the limitation by N is larger than that by P, when N and P are considered as independent, especially for maize ($R_N = 0.42 \pm 0.00$; $R_P = 0.62 \pm 0.01$) and wheat ($R_N = 0.49 \pm 0.00$; $R_P = 0.73 \pm 0.00$) (Table 2 of the Main Text). The spatial distributions of $R_N$ and $R_P$ are very different (Fig. S4 for maize), leading to all combinations possible (high N and P limitations, high N limitation and low P limitation, etc.), except the one with severe P limitation and no N limitation (the very few grid-cells in green in Fig. S5). Taking maize as an example, we found that: India and China are not severely limited by any of the nutrients (e.g. for China: $R_N = 0.61$; $R_P = 0.79$), the USA is moderately limited in both nutrients ($R_N = 0.43$; $R_P = 0.49$), Western Europe is more N- than P-limited (e.g. for Spain: $R_N = 0.24$; $R_P = 0.96$) and, the Western Russian Federation and Ukraine are severely limited in both N and P (e.g. for Ukraine: $R_N = 0.12$; $R_P = 0.15$) (Fig. S5). The uncertainty at the grid-cell scale, arising from the uncertainty in the datasets and equation parameters, is larger for P than for N (Fig. S4), which reflects the large uncertainty in the P supply (Table S5). Nevertheless, the uncertainty regarding global values remains small (Table 2).

When N and P are considered in interaction, we found that nutrient limitation is common with the exception of China, India and to a lesser extent, Western Europe and Eastern USA (Fig. S6). Consequently, the global supply/demand ratio $R_{NP}$ drops to ~0.30 (Table 2). Our study indicates that the interaction is a process that must be considered in the estimates of nutrient limitation. In our approach, regions with low NP limitations are restricted to China, India and to a lesser extent, Western Europe and Eastern USA. Some elements support these findings. Previous studies partly based on substance flow analysis show very positive current soil nutrient balances in China (Liu et al., 2010; Ma et al., 2010). Croplands of China, India, and the USA together account for ~65% of global N and P excess (West et al., 2014). We found that Western Europe is more N-limited than P-limited. Despite a decrease in soil P input following improvements in fertilization reasoning since 1970 in Western European countries (Senthilkumar et al., 2012), P accumulated in soils during the past decades can still be used by plants (Ringeval et al., 2014). This legacy effect does not exist for N, and N fertilisation rates are now increasingly limited by environmental regulations in many Western European countries (European Commission, 2018). N stress was found to occur in Spain and France in a study performed with EPIC (Fig. 7 of Balkovič et al. (2013)) and in Schils et al. (2018). We found that the USA is moderately limited in both nutrients with contrasting behaviour between the centre of the USA (low $R_{NP}$) and the East (high $R_{NP}$). Spatial heterogeneity has been underlined at the Mississippi watershed scale by Jacobson et al. (2011) where there are large inputs of P fertilizers in the Corn Belt. Some modeling difficulties related to the representation of soil P dynamics in our approach could also contribute to an overestimation of P limitation in the USA. American soils are mainly represented by Mollisols and Oxisols, which are characterized by a high P fixing capacity. While our approach takes into account P exchange between the soil solution and soil particles, it does so by considering the long-term equilibrium, which may be not relevant for the representation of fertilizer application onto high fixing capacity soils (Kvakić et al., 2018).
Text S7. Relationship between $R_{NP}$ and yield

The relationship between the yield gap ($Y_{real}/Y_{pot}$, with $Y_{real}$ and $Y_{pot}$ being the actual and potential yield, respectively) and $R_{NP}$ was assessed. This was done at country scale. Nutrient limitation when both N and P are considered ($R_{NP}$) is supposed to be closer to the actual nutrient limitation than the one considering only one nutrient (either N or P). That is why we restrict our analysis to the relationship between nutrient limitation and yield gap to $R_{NP}$. $Y_{real}/Y_{pot}$ was provided by Mueller et al. (2012). The $Y_{real}/Y_{pot}$ ratio is used as a measure of the yield gap and is a function of nutrient limitations, water limitation, pest and diseases, etc. Country values of $R_{NP}$ and $Y_{real}/Y_{pot}$ were computed by considering only grid-cells for which our analysis provides $R_{NP}$ values (Table S3) and by using crop-area (Ramankutty et al., 2008) as weight. A negative exponential model $g (g: x \rightarrow 1-\beta.exp(-x), with \beta the constant calibrated) was fit using ordinary least squares. The portion of variance in $Y_{real}/Y_{pot}$ explained by $R_{NP}$ was estimated with the coefficient of determination ($R^2$). We investigated how a third variable could modulate the relationship between the yield and $R_{NP}$. To do this, we divided all countries into 4 equal quarters based on the quartiles of this third variable ([minimum, Q1[, [Q1, Q2[, [Q2, Q3[ and [Q3, maximum], where Q1, Q2, Q3 are the quartiles of the third variable) and computed $R^2$ of $g$ for each quarter. We checked that the change in $R^2$ found when focusing on quarters is not explained by a reduction in the numbers of countries considered by using random country subsets. The variables chosen as the third variable are related to other limiting factors (irrigated fraction for the crop considered, or precipitation or pesticide use per agricultural area). The analysis was restricted to the country scale because most of these variables are available at that scale only. Irrigated fractions for each crop are given by MIRCA (Portmann et al., 2010), precipitation is provided by CRU (Mitchell & Jones, 2005) and pesticide use (and agricultural area used to compute the pesticide use per ha) is derived from FAOSTAT (FAO, 2018). All variables are representative of the year 2000.

At country-scale, the spatial variance of $Y_{real}/Y_{pot}$ explained by a negative exponential fit against $R_{NP}$ (measured with $R^2$) is small: 0.10 for maize (Fig. S7) and wheat (not shown) and 0.25 for rice (not shown), a crop that is usually grown with sufficient irrigation. A small $R^2$ could be explained by other factors limiting yield (e.g. insufficient water) whose spatial distribution might be different to that of $R_{NP}$. For maize, we found that overall, $R^2$ increases when it is computed on subsets of countries characterized by more homogeneous water conditions, approached here by the national crop area fraction irrigated (Fig. S7, panels b-e) or the amount of precipitation (Fig. S7, panels h-k). Only the quarter with largest fractions of irrigated maize (Fig. S7e) or with the lowest precipitation (Fig. S7h) has an $R^2$ lower than the $R^2$ computed for all countries (Fig. S7, first column). The increase in $R^2$ when sampling countries with homogeneous irrigation practices or precipitation is found (to a lesser extent) for wheat (not shown), but not for rice (not shown). We did not find any increase in $R^2$ when countries are segregated according to the amount of pesticides used per agricultural area (third row of Fig. S7 for maize).
Text S8. Characterization of each category defined in Harpole et al. (2011) in terms of values for $R_P$ and $R_N$ with the MH formalism

The category A is defined by:

\[ \Delta \text{pro}_N = 0 \] (x)
\[ \Delta \text{pro}_P = 0 \] (y)
\[ \Delta \text{pro}_{NP} > \Delta \text{pro}_N + \Delta \text{pro}_P \] (z).

Thanks to Ev.S3, \((x) \Leftrightarrow (R_P = 0 \text{ or } R_N = 1)\). $R_N$ cannot be equal to 1 otherwise

\[ \Delta \text{pro}_{NP} = \Delta \text{pro}_N + \Delta \text{pro}_P \] (Ev.S5).

Similarly, \((y) \Leftrightarrow (R_N = 0 \text{ or } R_P = 1)\) and $R_P$ cannot be equal to 1 otherwise

\[ \Delta \text{pro}_{NP} = \Delta \text{pro}_N + \Delta \text{pro}_P \].

The only combination possible is: $R_P = R_N = 0$.

Category C is characterized by:

\[ \Delta \text{pro}_N \neq 0 \] (x)
\[ \Delta \text{pro}_P = 0 \] (y)
\[ \Delta \text{pro}_{NP} > \Delta \text{pro}_N + \Delta \text{pro}_P \] (z).

\((y) \Leftrightarrow (R_N = 0 \text{ or } R_P = 1)\) and $R_P$ cannot be equal to 1 because of (z). Thus, $R_N = 0$. In addition, $R_P$ cannot be equal to 0 because it would imply $\Delta \text{pro}_N = 0$ thanks to Ev.S3. Thus, category C occurs if and only if: $R_N = 0$ and $R_P$ in $]0,1[$.

Similarly, the category D is characterized by: $R_P = 0$ and $R_N$ in $]0,1[$.

The category E is characterized by:

\[ \Delta \text{pro}_N \neq 0 \] (x)
\[ \Delta \text{pro}_P = 0 \] (y)
\[ \Delta \text{pro}_{NP} = \Delta \text{pro}_N + \Delta \text{pro}_P \] (z).

\((z) \Leftrightarrow (R_N = 1 \text{ or } R_P = 1)\). $R_N$ cannot be equal to 1 otherwise, $\Delta \text{pro}_N = 0$. Thus, the category E occurs if and only if: $R_P = 1$ and $R_N$ in $[0,1[$.

Similarly, the category F is characterized by: $R_N = 1$ and $R_P$ in $[0,1[$.

Category G is characterized by:

\[ \Delta \text{pro}_N = 0 \] (x)
\[ \Delta \text{pro}_P = 0 \] (y)
\[ \Delta \text{pro}_{NP} = \Delta \text{pro}_N + \Delta \text{pro}_P \] (z).

\((x) \Leftrightarrow (R_P = 0 \text{ or } R_N = 1)\) and \((y) \Leftrightarrow (R_N = 0 \text{ or } R_P = 1)\) and

\((z) \Leftrightarrow (R_P = 1 \text{ or } R_N = 1)\). The only combination allowed is: $R_P = R_N = 1$.

Category B is characterized by:

\[ \Delta \text{pro}_N \neq 0 \] (x)
\[ \Delta \text{pro}_P \neq 0 \] (y)
\[ \Delta \text{pro}_{NP} > \Delta \text{pro}_N + \Delta \text{pro}_P \] (z).

It occurs if and only if: both $R_P$ and $R_N$ are in $]0,1[$.
References


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### Supporting Tables

<table>
<thead>
<tr>
<th>Table 1</th>
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<td>Value C</td>
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<td>Value A</td>
<td>Value B</td>
<td>Value C</td>
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<td>Column B</td>
<td>Value D</td>
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Table S1. Description and computation of the different terms used in Eq.1-2 of the Main Text.

<table>
<thead>
<tr>
<th>Variable</th>
<th>( S_p )</th>
<th>( D_p )</th>
<th>( S_N )</th>
<th>( D_N )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Name</td>
<td>P supply</td>
<td>P demand</td>
<td>N supply</td>
<td>N demand</td>
</tr>
<tr>
<td>Unit</td>
<td>kgP/ha/yr</td>
<td>kgP/ha/yr</td>
<td>kgN/ha/yr</td>
<td>kgN/ha/yr</td>
</tr>
</tbody>
</table>

**Computation**

\( S_p = P_{uptake} + \alpha \cdot P_{fert} / CI \)

with \( P_{uptake} \): potential P root uptake, \( \alpha \): constant, \( P_{fert} \):
inorganic content of total P fertilizer applied the year considered, CI: crop harvest per year (Portmann et al., 2010).

The \( P_{uptake} \) computation accounts for the global distribution of soil P (Ringeval et al., 2017) and the diffusion of soil P to the root (see Text S4).

\( D_p = Y_{pot} \cdot \left( \frac{P_{\%\text{, grain}}}{\text{PHI}} + \frac{P_{\%\text{, root}} \cdot \text{RSR}}{\text{HI}} \right) \)

with \( Y_{pot} \): potential yield, PHI: P harvest index, HI: harvest index, RSR: root/shoot ratio and \( P_{\%\text{, grain}} \) and \( P_{\%\text{, root}} \): P concentration for grain and root, respectively.

See Text S3

\( S_N = N_{fix} + N_{dep} + N_{fert} + N_{man} - N_{vol} \)

Where \( N_{fix}, N_{dep}, N_{fert}, N_{man} \) are soil N input corresponding to fixation, deposition, chemical fertilizer and manure, respectively. \( N_{vol} \) corresponds to NH\(_3\) volatilization.

\( D_N = Y_{pot} \cdot \left( \frac{N_{\%\text{, grain}}}{\text{NHI}} + \frac{N_{\%\text{, root}} \cdot \text{RSR}}{\text{HI}} \right) \)

with \( Y_{pot} \): potential yield, NHI: N harvest index, HI: harvest index, RSR: root/shoot ratio and \( N_{\%\text{, grain}} \) and \( N_{\%\text{, root}} \): N concentration for grain and root, respectively.

See Text S3

**Consideration of the uncertainty: distribution**

How is one replicate (out of the 1000 replicates for \( S_p, D_p, S_N, D_N \)) chosen?

A combination of:

- \( P_{uptake} \): 1000 replicates given by Kvakić et al. (2018)
- \( P_{fert} \): 30 replicates given by Ringeval et al. (2017)
- \( \alpha \): normal distribution with average=0.17 and CV=20%

Normal distribution for all parameters used (PHI, HI, RSR, \( P_{\%\text{, grain}}, P_{\%\text{, root}} \)) with average and STD provided in the literature

A normal distribution with CV=20% is assumed (20% corresponds to the default value of uncertainty in Kvakić et al. (2018))

Normal distribution for all parameters used (NHI, HI, RSR, \( N_{\%\text{, grain}}, N_{\%\text{, root}} \)) with average and STD provided in the literature

**Crop dependence (wheat, maize, rice)?**

Yes, through:

- the potential root uptake that depends on root biomass (the soil P maps (Ringeval et al., 2017) are not crop-dependent)
- CI

Yes

No

Yes

**Reference for the computation**

(Kvakić et al., 2018) (Kvakić et al., 2018) (Bouwman et al., 2011) This study

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Table S2. Parameters used to estimate the N and P demands ($D_N$ and $D_P$, respectively). Values in the Table are representative of plant maturity and were taken from field experiments (rather than hydroponic experiments) if possible (this is still not the case for roots). Crop-specific values for N and P concentrations organs were derived from field experiments in stressed conditions focusing on the lower, linear part of the uptake-yield curve when nutrient use efficiency is maximal. Consequently, the nutrient demand estimates correspond to the minimum amount of nutrients required to achieve a certain grain yield. Mean values are shown with their standard error. If a standard error was not provided in the source material, a coefficient of variation of 20% was assumed.

\[ \text{DM used in the column "Unit" refers to Dry Matter.} \]

\[ X_{\text{organ}} \text{ with } X \text{ in } \{ C, N, P \} \text{ and } \text{organ in } \{ \text{root, shoot, grain} \} \text{ are in } gX. \gamma \text{ is a converting factor equal to } 0.45e^{3} \text{gC/kgDM.} \]

<table>
<thead>
<tr>
<th>Variable</th>
<th>Unit</th>
<th>Name</th>
<th>Definition</th>
<th>Maize</th>
<th>Wheat</th>
<th>Rice</th>
</tr>
</thead>
<tbody>
<tr>
<td>RSR</td>
<td>[-]</td>
<td>Root/shoot ratio</td>
<td>$\frac{C_{\text{root}}}{C_{\text{shoot}}}$</td>
<td>0.16 (Amos &amp; Walters, 2006)</td>
<td>0.15 (Williams et al., 2013)</td>
<td>0.15 ± 0.07 (Wissuwa &amp; Ae, 2001)</td>
</tr>
<tr>
<td>HI</td>
<td>[-]</td>
<td>Harvest index</td>
<td>$\frac{C_{\text{grain}}}{C_{\text{shoot}}}$</td>
<td>0.53 (Hütsch &amp; Schubert, 2017)</td>
<td>0.51 (Hütsch &amp; Schubert, 2017)</td>
<td>0.51 ± 0.07 (Rose et al., 2010)</td>
</tr>
<tr>
<td>NHI</td>
<td>[-]</td>
<td>N harvest index</td>
<td>$\frac{N_{\text{grain}}}{N_{\text{shoot}}}$</td>
<td>0.66 ± 0.11 (Van Duivenbooden, 1992)</td>
<td>0.73 ± 0.03 (Górny &amp; Garczyński, 2008)</td>
<td>0.61 ± 0.10 (Van Duivenbooden, 1992)</td>
</tr>
<tr>
<td>$N_{%_{\text{grain}}}$</td>
<td>[gN/kgDM]</td>
<td>Grain N concentration $\gamma$</td>
<td>$\frac{N_{\text{grain}}}{C_{\text{grain}}}$</td>
<td>15.5 ± 3.0 (Van Duivenbooden, 1992)</td>
<td>21.4 ± 4.8 (Van Duivenbooden, 1992)</td>
<td>11.7 (Ye et al., 2014)</td>
</tr>
<tr>
<td>$N_{%_{\text{root}}}$</td>
<td>[gN/kgDM]</td>
<td>Root N concentration $\gamma$</td>
<td>$\frac{N_{\text{root}}}{C_{\text{root}}}$</td>
<td>12.7 (Latshaw &amp; Miller, 1924)</td>
<td>6.1 (Hocking, 1994)</td>
<td>13.4 ± 0.1 (Ye et al., 2014)</td>
</tr>
<tr>
<td>PHI</td>
<td>[-]</td>
<td>P harvest index</td>
<td>$\frac{P_{\text{grain}}}{P_{\text{shoot}}}$</td>
<td>0.67 ± 0.13 (Van Duivenbooden, 1992)</td>
<td>0.67 ± 0.07 (Górny &amp; Garczyński, 2008)</td>
<td>0.61 ± 0.13 (Van Duivenbooden, 1992)</td>
</tr>
<tr>
<td>$P_{%_{\text{grain}}}$</td>
<td>[gP/kgDM]</td>
<td>Grain P concentration $\gamma$</td>
<td>$\frac{P_{\text{grain}}}{C_{\text{grain}}}$</td>
<td>2.90 ± 0.80 (Van Duivenbooden, 1992)</td>
<td>3.70 ± 0.80 (Van Duivenbooden, 1992)</td>
<td>3.58 ± 0.15 (Ye et al., 2014)</td>
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<tr>
<td>$P_{%_{\text{root}}}$</td>
<td>[gP/kgDM]</td>
<td>Root P concentration $\gamma$</td>
<td>$\frac{P_{\text{root}}}{C_{\text{root}}}$</td>
<td>1.20 (Latshaw &amp; Miller, 1924)</td>
<td>1.01 (Hocking, 1994)</td>
<td>1.31 ± 0.21 (Ye et al., 2014)</td>
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<tr>
<td></td>
<td>Maize (Mha)</td>
<td>Wheat (Mha)</td>
<td>Rice (Mha)</td>
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<tr>
<td><strong>Crop area [Mha]</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Observed crop area (Ramankutty et al., 2008) = CROP</td>
<td>142</td>
<td>214</td>
<td>168</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Observed crop area considered in our study * = CROP$_f$ (n=number of grid-cells)</td>
<td>96 (n=11565)</td>
<td>158 (n=9891)</td>
<td>93 (n=6405)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Production [Mt]</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CROP x Potential yield provided by Mueller et al. (2012)</td>
<td>1002</td>
<td>964</td>
<td>924</td>
<td></td>
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<tr>
<td>CROP$_f$ x Potential yield provided by Mueller et al. (2012)</td>
<td>743</td>
<td>727</td>
<td>532</td>
<td></td>
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* the reduction from CROP to CROP$_f$ is totally explained by the exclusion of grid-cells without a soil P estimate in Ringeval et al. (2017), which prevents the computation of $S_P$ and $R_P$. In Ringeval et al. (2017), some grid-cells are not considered because of missing data about soil biogeochemical background (Yang et al., 2013).
**Table S4.** Values possible for $R_N$ and $R_P$ and the implications with MH formalism.

<table>
<thead>
<tr>
<th>$R_P$</th>
<th>$R_N$</th>
<th>Implications</th>
<th>Category A</th>
<th>Category B</th>
<th>Category C</th>
<th>Category D</th>
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<th>Category G</th>
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<tbody>
<tr>
<td>0</td>
<td>0</td>
<td>$\Delta pro_{+P} = 0$</td>
<td>Category A</td>
<td>Category D</td>
<td>Category C</td>
<td>Category B</td>
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<td>Category F</td>
<td>Category G</td>
</tr>
<tr>
<td>0</td>
<td>]0,1[</td>
<td>$\Delta pro_{+N} = 0$ and $\Delta pro_{+NP} = \Delta pro_{+NP}$</td>
<td>Category E</td>
<td>Category E</td>
<td>Category E</td>
<td>Category E</td>
<td>Category G</td>
<td></td>
<td></td>
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<tr>
<td>]0,1[</td>
<td>1</td>
<td>$\Delta pro_{+P} = 0$</td>
<td>Category B</td>
<td>Category E</td>
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<td>Category F</td>
<td></td>
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</tbody>
</table>
Table S5. For all crops, global values of supply (S), demand (D) and supply/demand ratio (R) for N and P when the two nutrients are considered as independent. For all variables (S, D, R), we computed a global average weighted by the crop area for each simulation out of the 1000 replicates (see Text S5). Average (AVG), standard-deviation (STD) and coefficient of variation (CV) of the 1000 global averages are given in the Table. AVG and STD are in kg(N or P)/ha/yr while CV is in %.

<table>
<thead>
<tr>
<th></th>
<th>N</th>
<th>P</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maize</td>
<td></td>
<td></td>
</tr>
<tr>
<td>S</td>
<td>AVG = 100.38, STD = 0.65, CV = 1</td>
<td>AVG = 136.33, STD = 5.62, CV = 4</td>
</tr>
<tr>
<td>D</td>
<td>AVG = 219.20, STD = 1.16, CV = 1</td>
<td>AVG = 38.01, STD = 0.30, CV = 1</td>
</tr>
<tr>
<td>R</td>
<td>AVG = 0.42, STD = 0.00, CV = 1</td>
<td>AVG = 0.62, STD = 0.01, CV = 1</td>
</tr>
<tr>
<td>Wheat</td>
<td></td>
<td></td>
</tr>
<tr>
<td>S</td>
<td>AVG = 88.92, STD = 0.57, CV = 1</td>
<td>AVG = 242.48, STD = 7.97, CV = 3</td>
</tr>
<tr>
<td>D</td>
<td>AVG = 144.04, STD = 0.54, CV = 0</td>
<td>AVG = 27.18, STD = 0.11, CV = 0</td>
</tr>
<tr>
<td>R</td>
<td>AVG = 0.49, STD = 0.00, CV = 0</td>
<td>AVG = 0.73, STD = 0.00, CV = 1</td>
</tr>
<tr>
<td>Rice</td>
<td></td>
<td></td>
</tr>
<tr>
<td>S</td>
<td>AVG = 125.77, STD = 1.05, CV = 1</td>
<td>AVG = 97.09, STD = 2.88, CV = 3</td>
</tr>
<tr>
<td>D</td>
<td>AVG = 136.04, STD = 1.00, CV = 1</td>
<td>AVG = 37.66, STD = 0.32, CV = 1</td>
</tr>
<tr>
<td>R</td>
<td>AVG = 0.70, STD = 0.00, CV = 1</td>
<td>AVG = 0.79, STD = 0.01, CV = 1</td>
</tr>
</tbody>
</table>
Supporting Figures

Figure S1. For maize, the spatial distribution of increase in $R_N$ (left) and $R_P$ (right) required at the same time to make $R_{NP}$ equal to 0.75. The increases are computed with MH (top) and LM (bottom) formalisms. Global averaged values and one standard-deviation are provided in the right bottom corner of each panel.
a) \( \frac{R_{NP}^{real} - R_{NP}^{pot}}{R_{NP}^{pot}} \)

b) categories based on \( R_{NP}^{real} \) and \( R_{NP}^{pot} \)

- iv) \( R_{NP}^{real} \neq 1; R_{NP}^{pot} \neq 1 \)
- iii) \( R_{NP}^{real} = 1; R_{NP}^{pot} \neq 1 \)
- ii) \( R_{NP}^{real} \neq 1; R_{NP}^{pot} = 1 \)
- i) \( R_{NP}^{real} = 1; R_{NP}^{pot} = 1 \)

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Figure S2 (previous page). The effect of using the actual yield (instead of potential yield) on the computed nutrient limitation. In the Main Text, the potential yield (Y\text{pot}) is used to compute R\text{NP} (through Eq.5 for the N and P demands and Eq.S15 for the P uptake involved in the P supply computation). In this figure, we compared the NP limitation (R\text{NP}) when computed with potential yield (called here R\text{NP}\text{pot}) as in the Main Text and when computed with Y\text{real} (called here R\text{NP}\text{real}). Both Y\text{pot} and Y\text{real} are provided by Mueller et al. (2012). The figure shows the difference between R\text{NP}\text{pot} and R\text{NP}\text{real} (expressed in percentage of R\text{NP}\text{pot}, top) and some categories based on the values of R\text{NP}\text{pot} and R\text{NP}\text{real} (bottom). Only grid-cells with Y\text{real}<0.75*Y\text{pot} are plotted in the two panels. The different categories used in the bottom panel can be interpreted as follows:

- **Class i**: R\text{NP}\text{real}=1 and R\text{NP}\text{pot}=1. The actual yield is not limited by NP but by other factors (because Y\text{real} is smaller than Y\text{pot}). Current NP supply would be sufficient to satisfy the demand if the limitation by these other factors was removed.
- **Class ii**: R\text{NP}\text{real}≠1 and R\text{NP}\text{pot}=1. No grid-cell in this category. This is partly explained by the fact that R\text{NP}\text{real} is mostly greater than R\text{NP}\text{pot}.
- **Class iii**: R\text{NP}\text{real}=1 and R\text{NP}\text{pot}≠1. The actual yield is not limited by NP but by other factors (because Y\text{real} is smaller than Y\text{pot}). Current NP supply would be insufficient to satisfy the demand if the limitation by these other factors was removed.
- **Class iv**: R\text{NP}\text{real}≠1 and R\text{NP}\text{pot}≠1. The actual yield is limited by NP and potentially by other factors.
Figure S3. Grid-cell distribution in percentiles of different variables (S: supply, D: demand, R: supply/demand ratio, \( \Delta R \): increase in R required to make \( R_{NP} = 0.75 \)) for maize. Values plotted in this figure are not weighted by the cropland area of each grid-cell. 11565 grid-cells have been considered for maize in our approach (Table S3).
**Figure S4.** For maize, spatial distribution of $R_N$ and $R_P$ when N and P are considered as independent: average and standard-deviation of the 1000 replicates.
Figure S5. For maize, the spatial distribution of nutrient limitation when N and P are considered to be independent (bivariate plot of $R_N$ and $R_P$).
Figure S6. For maize, spatial distribution of $R_{NP}$: average and standard-deviation for both formalisms of interaction (a-b: MH; c-d: LM). The averaged difference of $R_{NP}$ between LM and MH is also plotted (panel e).
Figure S7. Scatterplots of the ratio $Y_{\text{real}}/Y_{\text{pot}}$ provided by Mueller et al. (2012) vs. the simulated $R_{NP}$ (here only computed by using the MH formalism for the purpose of simplicity) at the country scale for maize. Each dot corresponds to one country. In the extreme-left column, all countries are considered while columns 2-5 correspond to the different quarters based on the quartiles of a third variable (column 2: [minimum, Q1], column 3: [Q1, Q2], column 4: [Q2, Q3], column 5: [Q3, maximum] with Q1-3 the quartiles of a third
variable). The different rows correspond to different third variables used to distinguish the countries in quarters (top: irrigated fraction of maize, middle: precipitations, bottom: pesticide use per total agricultural area). The extreme-left panels vary among rows because we consider only countries for which data about the third variable is available. The green line corresponds to a negative exponential fit $g(x) = 1 - \beta \exp(-x)$. The fit is made for each extreme-left panels (a, g, m) and reported in columns 2-5 of the same row. Influence plots are provided in Figure S11. The number of countries considered (called n in the title of each panel), as well as the $R^2$ for the fit computed on all countries are given for each panel. The name of the country (ISO nomenclature) is given for each panel of columns 2-5. The extreme-right panels (l, f, r) provide the average of countries considered in each quarter and the boundaries of the colour palet are defined by (min, Q1, Q2, Q3, max). The error bars of panels b-e, h-k and n-q correspond to the standard-deviation arising from the 1000 simulations described in the Main Text. In panels f, l and r, the error bars correspond to the standard-deviation arising from the different countries within each quarter.