Supporting Information

Supporting Text

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Supporting text

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

First, we defined the function f as f: $x \to min(1,x)$. By definition, the N and P limitations encountered in the control experiment, R_N and R_P, were defined as $R_N = f(\frac{S_N}{D_N})$ and $R_P = f(\frac{S_P}{D_P})$. 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: $R_N(E_1) = f(\frac{S_N}{D_N})$ and $R_P(E_1) = f(\frac{S_P}{D_P})$. $R_N(E_1)$ and $R_P(E_1)$ are called R_N and R_P (as in the Main Text).

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

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

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

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 Δpro_{+P}, Δpro_{+N} and Δpro_{+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 Δpro_{+N} and Δpro_{+P} and ii) the relationship between Δpro_{+NP} and (Δpro_{+N}+Δpro_{+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,

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

with X=N or P. Equality is only possible when $f(\frac{S_X}{D_X}) = 1$, i.e. R_X=1. Otherwise, $f(\frac{S_X + A_X}{D_X}) > f(\frac{S_X}{D_X})$. Thus, we have the two equivalences below:

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

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

Note that the above equivalences imply that $f(\frac{S_X + A_X}{D_X})$ is necessarily non-null.

Text S1.1. MH formalism

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

$$\Delta pro_{+N}=f\left(\frac{S_N+A_N}{D_N}\right).f\left(\frac{S_P}{D_P}\right)-f\left(\frac{S_N}{D_N}\right).f\left(\frac{S_P}{D_P}\right) \quad \text{(Eq.S2)}$$

$$\Delta pro_{+P}=f\left(\frac{S_N}{D_N}\right).f\left(\frac{S_P+A_P}{D_P}\right)-f\left(\frac{S_N}{D_N}\right).f\left(\frac{S_P}{D_P}\right) \quad \text{(Eq.S3)}$$

$$\Delta pro_{+NP}=f\left(\frac{S_N+A_N}{D_N}\right).f\left(\frac{S_P+A_P}{D_P}\right)-f\left(\frac{S_N}{D_N}\right).f\left(\frac{S_P}{D_P}\right) \quad \text{(Eq.S4).}$$

Thanks to Eq.S2, we find that $\Delta pro_{+N}=0$ is only possible if either $f\left(\frac{S_P}{D_P}\right)=0$ or $f\left(\frac{S_N+A_N}{D_N}\right)=f\left(\frac{S_N}{D_N}\right)$. The first condition corresponds to R_P=0, the 2nd one to R_N=1 (see Ev.S2). Thus,

$$(\Delta pro_{+N}=0)\Leftrightarrow(R_P=0 \text{ or } R_N=1) \quad \text{(Ev.S3)}$$

And similarly,

$$(\Delta pro_{+P}=0)\Leftrightarrow(R_N=0 \text{ or } R_P=1) \quad \text{(Ev.S4).}$$

Also we have to note that R_N or R_P equal to 1 have some implications. From Ev.S2, we have $[R_N=1]\Leftrightarrow[f\left(\frac{S_N+A_N}{D_N}\right)=f\left(\frac{S_N}{D_N}\right)=1]$. In that case, Eq.S2-S4 becomes $\Delta pro_{+N}=0$, $\Delta pro_{+P}=f\left(\frac{S_P+A_P}{D_P}\right)-f\left(\frac{S_P}{D_P}\right)$ and $\Delta pro_{+NP}=f\left(\frac{S_P+A_P}{D_P}\right)-f\left(\frac{S_P}{D_P}\right)$. Thus, we get $\Delta pro_{+NP}=\Delta pro_{+N}+\Delta pro_{+P}$. Thus, $(R_N=1)\Rightarrow(\Delta pro_{+NP}=\Delta pro_{+N}+\Delta pro_{+P})$.
Similarly, $(R_P=1)\Rightarrow(\Delta pro_{+NP}=\Delta pro_{+N}+\Delta pro_{+P})$. Further, we can even demonstrate (Text S2) that:

$$(R_P=1 \text{ or } R_N=1)\Leftrightarrow(\Delta pro_{+NP}=\Delta pro_{+N}+\Delta pro_{+P}) \quad \text{(Ev.S5).}$$

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:

$\Delta pro_{+N}=0$ (x)
$\Delta pro_{+P}=0$ (y)
$\Delta pro_{+NP}>\Delta pro_{+N}+\Delta pro_{+P}$ (z)

According to Ev.S3, $(x)\Leftrightarrow(R_P=0 \text{ or } R_N=1)$. R_N cannot be equal to 1 otherwise $\Delta pro_{+NP}=\Delta pro_{+N}+\Delta pro_{+P}$. Similarly, $(y)\Leftrightarrow(R_N=0 \text{ or } R_P=1)$ and R_P cannot be equal to 1 otherwise $\Delta pro_{+NP}=\Delta pro_{+N}+\Delta pro_{+P}$. 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 $(\Delta pro_{+NP}) \leq (\Delta pro_{+N} + \Delta pro_{+P})$. With MH formalism, because of Ev.S3, $(\Delta pro_{+N} \neq 0)$ 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 $(R_P = 1 \text{ or } R_N = 1) \Leftrightarrow (\Delta pro_{+NP} = \Delta pro_{+N} + \Delta pro_{+P})$ (Ev.S5), it means that $(\Delta pro_{+NP}) > (\Delta pro_{+N} + \Delta pro_{+P})$ 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), Eq9-11 become:

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

$$\Delta pro_{+P} = min[f(\frac{S_N}{D_N}), f(\frac{S_P + A_P}{D_P})] - min[f(\frac{S_N}{D_N}), f(\frac{S_P}{D_P})] \quad \text{(Eq.S6)}$$

$$\Delta pro_{+NP} = min[f(\frac{S_N + A_N}{D_N}), f(\frac{S_P + A_P}{D_P})] - min[f(\frac{S_N}{D_N}), f(\frac{S_P}{D_P})] \quad \text{(Eq.S7)}$$

The above equations can be solved if we know how the different ratios involved ($\frac{S_N}{D_N}$, $\frac{S_P}{D_P}$, $\frac{S_N + A_N}{D_N}$, $\frac{S_P + A_P}{D_P}$) 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 $f(\frac{S_N}{D_N}) < f(\frac{S_P}{D_P})$. Because of (Eq.S1) , we also have:

$$f(\frac{S_N}{D_N}) < f(\frac{S_P}{D_P}) \leq f(\frac{S_P + A_P}{D_P}) \quad \text{(Eq.S8).}$$

Eq.S5-S7 becomes:

$$\Delta pro_{+N} = min[f(\frac{S_N + A_N}{D_N}), f(\frac{S_P}{D_P})] - f(\frac{S_N}{D_N}) \quad \text{(Eq.S9)}$$

$$\Delta pro_{+P} = f(\frac{S_N}{D_N}) - f(\frac{S_N}{D_N}) = 0 \quad \text{(Eq.S10)}$$

$$\Delta pro_{+NP} = min[f(\frac{S_N + A_N}{D_N}), f(\frac{S_P + A_P}{D_P})] - f(\frac{S_N}{D_N}) \quad \text{(Eq.S11)}$$

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 $f(\frac{S_N+A_N}{D_N})<f(\frac{S_P}{D_P})$. Because of Eq.S1 applied to N, we get $f(\frac{S_N+A_N}{D_N})<f(\frac{S_P}{D_P})\leq f(\frac{S_P+A_P}{D_P})$. Eq.S9&S11 becomes $\Delta pro_{+N}=f(\frac{S_N+A_N}{D_N})-f(\frac{S_N}{D_N})$ and $\Delta pro_{+NP}=f(\frac{S_N+A_N}{D_N})-f(\frac{S_N}{D_N})$. Thus, $\Delta pro_{+NP}=\Delta pro_{+N}+\Delta pro_{+P}$. We are in the case corresponding to category E.

If E_2 is only P-limited, we have:

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

and Eq.S9 becomes $\Delta pro_{+N}=f(\frac{S_P}{D_P})-f(\frac{S_N}{D_N})$. Because E_2 is P-limited, we also have $f(\frac{S_P}{D_P})<1$ and thus, following Ev.S1, we have:

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

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

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

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

Thus, Eq.S9 becomes e.g. $\Delta pro_{+N}=f(\frac{S_P}{D_P})-f(\frac{S_N}{D_N})$ and Eq.S11 becomes $\Delta pro_{+NP}=min[f(\frac{S_P}{D_P}),f(\frac{S_P+A_P}{D_P})]-f(\frac{S_N}{D_N})$. Because of Eq.S1, the latter equation is equivalent to $\Delta pro_{+NP}=f(\frac{S_P}{D_P})-f(\frac{S_N}{D_N})$ and thus, $\Delta pro_{+NP}=\Delta pro_{+N}+\Delta pro_{+P}$. 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, $f(\frac{S_P+A_P}{D_P})>f(\frac{S_N+A_N}{D_N})$: E_4 is necessarily N-limited.

If E_2 is not limited, we have $f(\frac{S_P}{D_P})=f(\frac{S_N+A_N}{D_N})=1$. In that case, because of Eq.S1, we also have $f(\frac{S_P+A_P}{D_P})=1$ (it means that E_4 is not limited). Eq.S9&S11 becomes $\Delta pro_{+N}=1-f(\frac{S_N}{D_N})$ and $\Delta pro_{+NP}=1-f(\frac{S_N}{D_N})$, respectively. Thus, $\Delta pro_{+NP}=\Delta pro_{+N}+\Delta pro_{+P}$. 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 <=> [$R_N<R_P$ and $R_P(E_2)<R_N(E_2)$]
category E <=> [$R_N<R_P$ and $R_P(E_2)\geq R_N(E_2)$]

The same reasoning applies to categories D (E_1 P-limited and E_3 N-limited) and F (E_1 P-limited and E_3 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: $R_P=R_N\neq1$.
Category G is characterized by E_1 neither N-limited nor P-limited, thus, $R_P=R_N=1$.

Category B is defined by $\Delta pro_{+N}\neq0$. Thus, E_1 is N-limited, i.e. $R_N\leq R_P$. Because $\Delta pro_{+N}\neq0$, 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. $R_P=R_N\neq1$. This implies that Eq.S5 could be written as follow:

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

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

$$\Delta pro_{+N}=f(\frac{S_N}{D_N})-f(\frac{S_N}{D_N}) \quad \text{(because } R_N\neq1 \text{ and Ev.S1).}$$

Thus, $\Delta pro_{+N}=0$ 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 $(R_P=1 \text{ or } R_N=1) \Leftrightarrow (\Delta\, pro_{+NP} = \Delta\, pro_{+N} + \Delta\, pro_{+P})$ **with the MH formalism**

First we demonstrate that $(R_P=1 \text{ or } R_N=1) \Rightarrow (\Delta\, pro_{+NP} = \Delta\, pro_{+N} + \Delta\, pro_{+P})$ by contradiction (*reductio ad absurdum*):

By using definition of $\Delta\, pro_{+NP}$, $\Delta\, pro_{+N}$ and $\Delta\, pro_{+P}$ and after simplification, $\Delta\, pro_{+NP} > \Delta\, pro_{+N} + \Delta\, pro_{+P}$ implies that $[f(\frac{S_N + A_N}{D_N}) - f(\frac{S_N}{D_N})].[f(\frac{S_P + A_P}{D_P}) - f(\frac{S_P}{D_P})] > 0$.

Thus, because $[f(\frac{S_N + A_N}{D_N}) - f(\frac{S_N}{D_N})]$ and $[f(\frac{S_P + A_P}{D_P}) - f(\frac{S_P}{D_P})]$ cannot be negative, $\Delta\, pro_{+NP} > \Delta\, pro_{+N} + \Delta\, pro_{+P}$ implies that $\begin{cases} f(\frac{S_N + A_N}{D_N}) - f(\frac{S_N}{D_N}) > 0 \\ f(\frac{S_P + A_P}{D_P}) - f(\frac{S_P}{D_P}) > 0 \end{cases}$. Because Ev.S1, it means that $(\Delta\, pro_{+NP} > \Delta\, pro_{+N} + \Delta\, pro_{+P}) \Rightarrow \begin{cases} R_N \neq 1 \\ R_P \neq 1 \end{cases}$.

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

$$(R_P=1 \text{ or } R_N=1) \Leftrightarrow (\Delta\, pro_{+NP} = \Delta\, pro_{+N} + \Delta\, pro_{+P}) \quad .$$

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:

$$HI = \frac{C_{grain}}{C_{shoot}} \quad (1)$$

$$PHI = \frac{P_{grain}}{P_{shoot}} \quad (2)$$

$$RSR = \frac{C_{root}}{C_{shoot}} \quad (3)$$

$$P_{\%,i} = \frac{P_i}{C_i} \quad (4)$$

$$Y = C_{grain} \quad (5)$$

where C_i: the carbon content of organ i (in gC), *Pi* : 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: $D_P = P_{shoot} + P_{root}$.

By using (2), (4) (applied to i=grain) and (5), we have: $P_{shoot} = P_{\%,grain} \cdot \frac{Y}{PHI}$.

By using (4) (applied to i=root) and (3), we get: $P_{root} = P_{\%,root} . RSR . C_{shoot}$, then: $P_{root} = P_{\%,root} . RSR . \frac{Y}{HI}$ by using (1) and (5).

Finally, we get: $D_P = P_{\%,grain} \cdot \frac{Y}{PHI} + P_{\%,root} . RSR . \frac{Y}{HI}$. Similarly for N, we found: $D_N = N_{\%,grain} \cdot \frac{Y}{NHI} + N_{\%,root} . RSR . \frac{Y}{HI}$. 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 $P_{\%,grain}$, $P_{\%,root}$, $N_{\%,grain}$, $N_{\%,root}$, *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:

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

where *m* is the month, *Δz* 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), *ρ* is a dimensionless ratio of soil cylinder to root radius, and *G(ρ)* is a dimensionless geometric function related to uptake by diffusion only. P_{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). *Δz* 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:

$$L_{rv}(m)=\frac{C_{root}(m)*SRL}{\Delta z}$$

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

C_{root} is computed as follows:

$$C_{root}(m)=\frac{C_{root;LPJmL}(m)}{C_{root;LPJmL;max}} . \frac{RSR}{HI} . Y_{pot}$$

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

Text S6. Spatial distribution of R_N, R_P, R_{NP}

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±0.00; R_P=0.62±0.01) and wheat (R_N=0.49±0.00; R_P=0.73±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 β 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:

$\Delta\, pro_{+N} = 0$ (x)

$\Delta\, pro_{+P} = 0$ (y)

$\Delta\, pro_{+NP} > \Delta\, pro_{+N} + \Delta\, pro_{+P}$ (z).

Thanks to Ev.S3, $(x) \Leftrightarrow (R_P = 0 \text{ or } R_N = 1)$. R_N cannot be equal to 1 otherwise $\Delta\, pro_{+NP} = \Delta\, pro_{+N} + \Delta\, pro_{+P}$ (Ev.S5).

Similarly, $(y) \Leftrightarrow (R_N = 0 \text{ or } R_P = 1)$ and R_P cannot be equal to 1 otherwise $\Delta\, pro_{+NP} = \Delta\, pro_{+N} + \Delta\, pro_{+P}$.

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

Category C is characterized by:

$\Delta\, pro_{+N} \neq 0$ (x)

$\Delta\, pro_{+P} = 0$ (y)

$\Delta\, pro_{+NP} > \Delta\, pro_{+N} + \Delta\, pro_{+P}$ (z).

$(y) \Leftrightarrow (R_N = 0 \text{ or } R_P = 1)$. 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 $\Delta\, pro_{+N} = 0$ 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:

$\Delta\, pro_{+N} \neq 0$ (x)

$\Delta\, pro_{+P} = 0$ (y)

$\Delta\, pro_{+NP} = \Delta\, pro_{+N} + \Delta\, pro_{+P}$ (z).

$(z) \Leftrightarrow (R_N = 1 \text{ or } R_P = 1)$. R_N cannot be equal to 1 otherwise, $\Delta\, pro_{+N} = 0$. 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:

$\Delta\, pro_{+N} = 0$ (x)

$\Delta\, pro_{+P} = 0$ (y)

$\Delta\, pro_{+NP} = \Delta\, pro_{+N} + \Delta\, pro_{+P}$ (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:

$\Delta\, pro_{+N} \neq 0$ (x)

$\Delta\, pro_{+P} \neq 0$ (y)

$\Delta\, pro_{+NP} > \Delta\, pro_{+N} + \Delta\, pro_{+P}$ (z).

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

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Supporting Tables

Table S1. Description and computation of the different terms used in Eq.1-2 of the Main Text.

Variable	S_P	D_P	S_N	D_N
Name	P supply	P demand	N supply	N demand
Unit	kgP/ha/yr	kgP/ha/yr	kgN/ha/yr	kgN/ha/yr
Computation	$S_P = P_{uptake} + \alpha . P_{fert} / CI$ with P_{uptake}: potential P root uptake, α: 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).	$D_P = Y_{pot} . (\frac{P_{\%,grain}}{PHI} + \frac{P_{\%,root} . RSR}{HI})$ with Y_{pot}: potential yield, PHI: P harvest index, HI: harvest index, RSR: root/shoot ratio and $P_{\%,grain}$ and $P_{\%,root}$: 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.	$D_N = Y_{pot} . (\frac{N_{\%,grain}}{NHI} + \frac{N_{\%,root} . RSR}{HI})$ with Y_{pot}: potential yield, NHI: N harvest index, HI: harvest index, RSR: root/shoot ratio and $N_{\%,grain}$ and $N_{\%,root}$: 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) - α: normal distribution with average=0.17 and CV=20%	Normal distribution for all parameters used (PHI, HI, RSR, $P_{\%,grain}$, $P_{\%,root}$) 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, $N_{\%,grain}$, $N_{\%,root}$) 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

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. *DM* used in the column "Unit" refers to Dry Matter. X_{organ} with X in {*C, N, P*} and *organ* in {*root, shoot, grain*} are in gX. γ is a converting factor equal to $0.45e^{+3}$ gC/kgDM.

Variable	Unit	Name	Definition	Maize	Wheat	Rice
RSR	[-]	Root/shoot ratio	$\frac{C_{root}}{C_{shoot}}$	0.16 (Amos & Walters, 2006)	0.15 (Williams et al., 2013)	0.15 ± 0.07 (Wissuwa & Ae, 2001)
HI	[-]	Harvest index	$\frac{C_{grain}}{C_{shoot}}$	0.53 (Hütsch & Schubert, 2017)	0.51 (Hütsch & Schubert, 2017)	0.51 ± 0.07 (Rose et al., 2010)
NHI	[-]	N harvest index	$\frac{N_{grain}}{N_{shoot}}$	0.66 ± 0.11 (Van Duivenboode n, 1992)	0.73 ± 0.03 (Górny & Garczyński, 2008)	0.61 ± 0.10 (Van Duivenbooden, 1992)
$N_{\%, grain}$	[gN/kgDM]	Grain N concentration	$\gamma.\frac{N_{grain}}{C_{grain}}$	15.5 ± 3.0 (Van Duivenboode n, 1992)	21.4 ± 4.8 (Van Duivenboode n, 1992)	11.7 (Ye et al., 2014)
$N_{\%, root}$	[gN/kgDM]	Root N concentration	$\gamma.\frac{N_{root}}{C_{root}}$	12.7 (Latshaw & Miller, 1924)	6.1 (Hocking, 1994)	13.4 ± 0.1 (Ye et al., 2014)
PHI	[-]	P harvest index	$\frac{P_{grain}}{P_{shoot}}$	0.67 ± 0.13 (Van Duivenboode n, 1992)	0.67 ± 0.07 (Górny & Garczyński, 2008)	0.61 ± 0.13 (Van Duivenbooden, 1992)
$P_{\%, grain}$	[gP/kgDM]	Grain P concentration	$\gamma.\frac{P_{grain}}{C_{grain}}$	2.90 ± 0.80 (Van Duivenboode n, 1992)	3.70 ± 0.80 (Van Duivenboode n, 1992)	3.58 ± 0.15 (Ye et al., 2014)
$P_{\%, root}$	[gP/kgDM]	Root P concentration	$\gamma.\frac{P_{root}}{C_{root}}$	1.20 (Latshaw & Miller, 1924)	1.01 (Hocking, 1994)	1.31 ± 0.21 (Ye et al., 2014)

Table S3. Global crop area and production provided by global datasets and considered in our study.

		Maize (Mha)	Wheat (Mha)	Rice (Mha)
Crop area [Mha]	Observed crop area (Ramankutty et al., 2008) = CROP	142	214	168
	Observed crop area considered in our study * = $CROP_f$ (n=number of grid-cells)	96 (n=11565)	158 (n=9891)	93 (n=6405)
Production [Mt]	CROP x Potential yield provided by Mueller et al. (2012)	1002	964	924
	$CROP_f$ x Potential yield provided by Mueller et al. (2012)	743	727	532

* 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.

			R_N		
			0	]0,1[	1
		Implications	$\Delta\ pro_{+P}=0$		$\Delta\ pro_{+N}=0$ and $\Delta\ pro_{+NP}=\Delta\ pro_{+N}+\Delta\ pro_{+P}$
R_P	0	$\Delta\ pro_{+N}=0$	Category A	Category D	Category F
	]0,1[		Category C	Category B	Category F
	1	$\Delta\ pro_{+P}=0$ and $\Delta\ pro_{+NP}=\Delta\ pro_{+N}+\Delta\ pro_{+P}$	Category E	Category E	Category G

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 %.

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

Supporting Figures

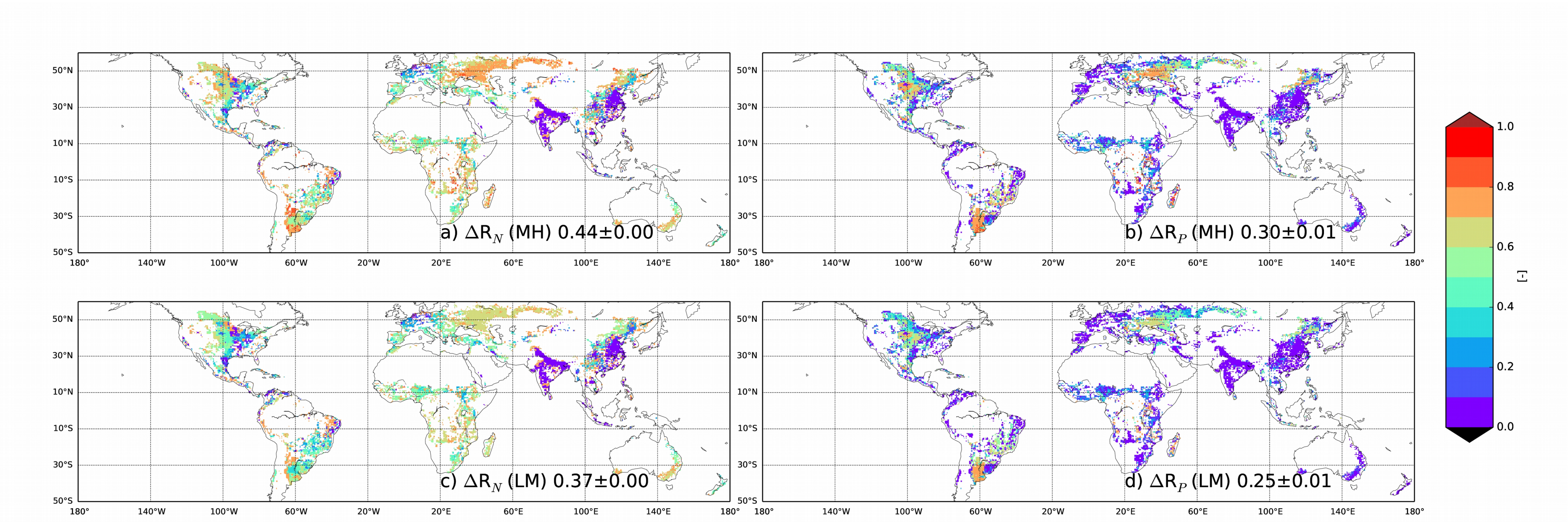

Figure S1. For maize, the spatial distribution of increase in R_N (left) and R_P (right) required at the same time to make R_{NP} equal to 0.75. The increases are computed with MH (top) and LM (bottom) formalisms. Global averaged values and one standard-deviation are provided in the right bottom corner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

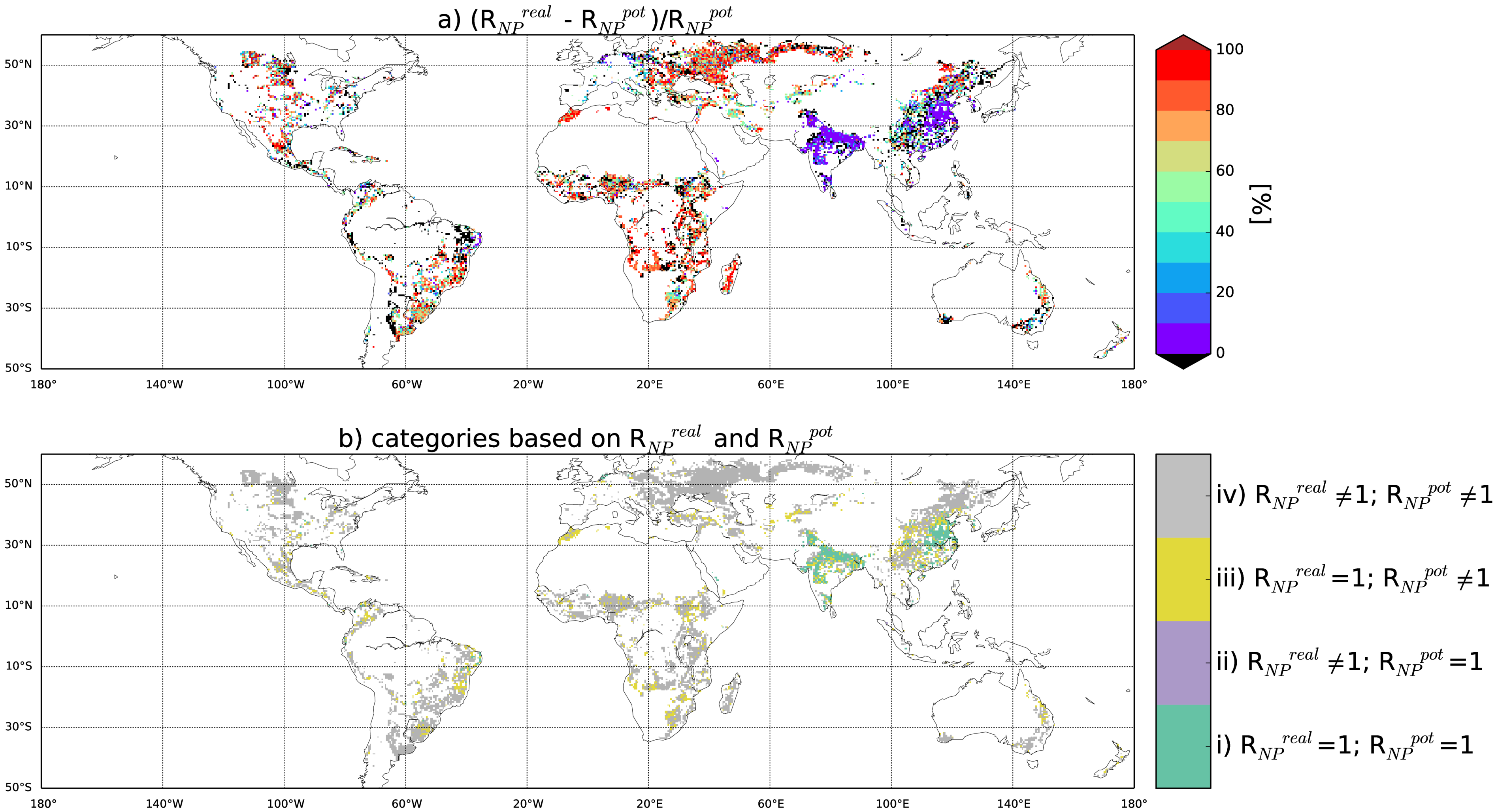
a) $(R_{NP}^{real} - R_{NP}^{pot})/R_{NP}^{pot}$
50°N
30°N
10°N
10°S
30°S
50°S
180°
140°W
100°W
60°W
20°W
20°E
60°E
100°E
140°E
180°
100
80
60
40
20
0
[%]
b) categories based on R_{NP}^{real} and R_{NP}^{pot}
iv) $R_{NP}^{real} \neq 1$; $R_{NP}^{pot} \neq 1$
iii) $R_{NP}^{real} = 1$; $R_{NP}^{pot} \neq 1$
ii) $R_{NP}^{real} \neq 1$; $R_{NP}^{pot} = 1$
i) $R_{NP}^{real} = 1$; $R_{NP}^{pot} = 1$

Figure S2 (previous page). The effect of using the acual yield (instead of potential yield) on the computed nutrient limitation. In the Main Text, the potential yield (Y_{pot}) is used to compute R_{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_{NP}) when computed with potential yield (called here R_{NP}^{pot}) as in the Main Text and when computed with Y_{real} (called here R_{NP}^{real}). Both Y_{pot} and Y_{real} are provided by Mueller et al. (2012). The figure shows the difference between R_{NP}^{pot} and R_{NP}^{real} (expressed in percentage of R_{NP}^{pot}, top) and some categories based on the values of R_{NP}^{pot} and R_{NP}^{real} (bottom). Only grid-cells with $Y_{real}<0.75*Y_{pot}$ are plotted in the two panels. The different categories used in the bottom panel can be interpreted as follows:

- Class i: $R_{NP}^{real}=1$ and $R_{NP}^{pot}=1$. The actual yield is not limited by NP but by other factors (because Y_{real} is smaller than Y_{pot}). Current NP supply would be sufficient to satisfy the demand if the limitation by these other factors was removed.
- Class ii: $R_{NP}^{real}\neq1$ and $R_{NP}^{pot}=1$. No grid-cell in this category. This is partly explained by the fact that R_{NP}^{real} is mostly greater than R_{NP}^{pot}.
- Class iii: $R_{NP}^{real}=1$ and $R_{NP}^{pot}\neq1$. The actual yield is not limited by NP but by other factors (because Y_{real} is smaller than Y_{pot}). Current NP supply would be insufficient to satisfy the demand if the limitation by these other factors was removed.
- Class iv: $R_{NP}^{real}\neq1$ and $R_{NP}^{pot}\neq1$. The actual yield is limited by NP and potentially by other factors.

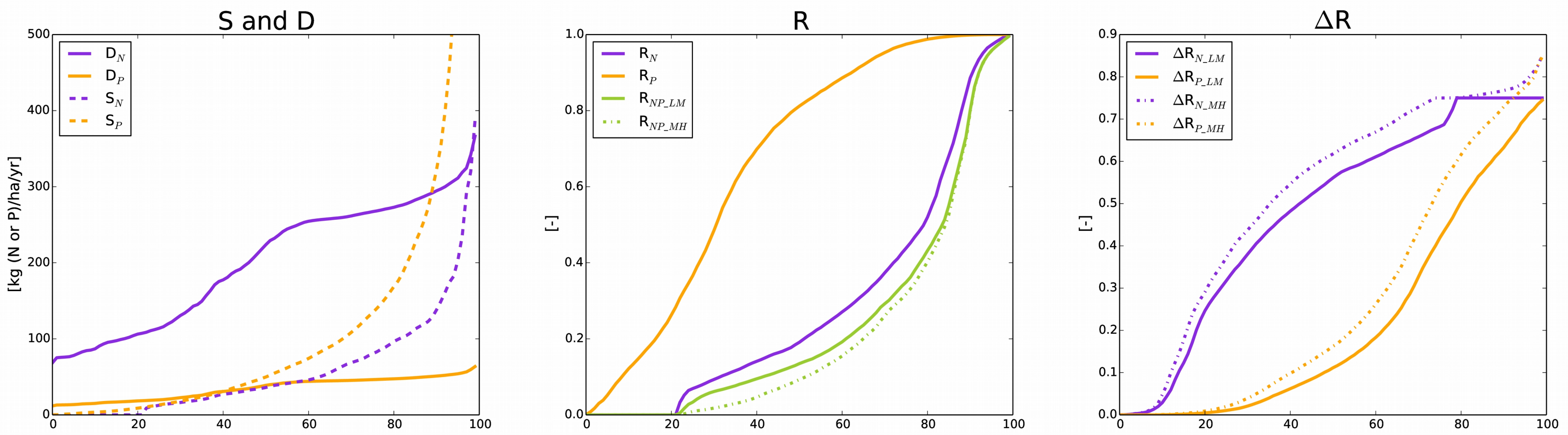

Figure S3. Grid-cell distribution in percentiles of different variables (S: supply, D: demand, R: supply/demand ratio, ΔR: increase in R required to make R_{NP}=0.75) for maize. Values plotted in this figure are not weighted by the cropland area of each grid-cell. 11565 grid-cells have been considered for maize in our approach (Table S3).

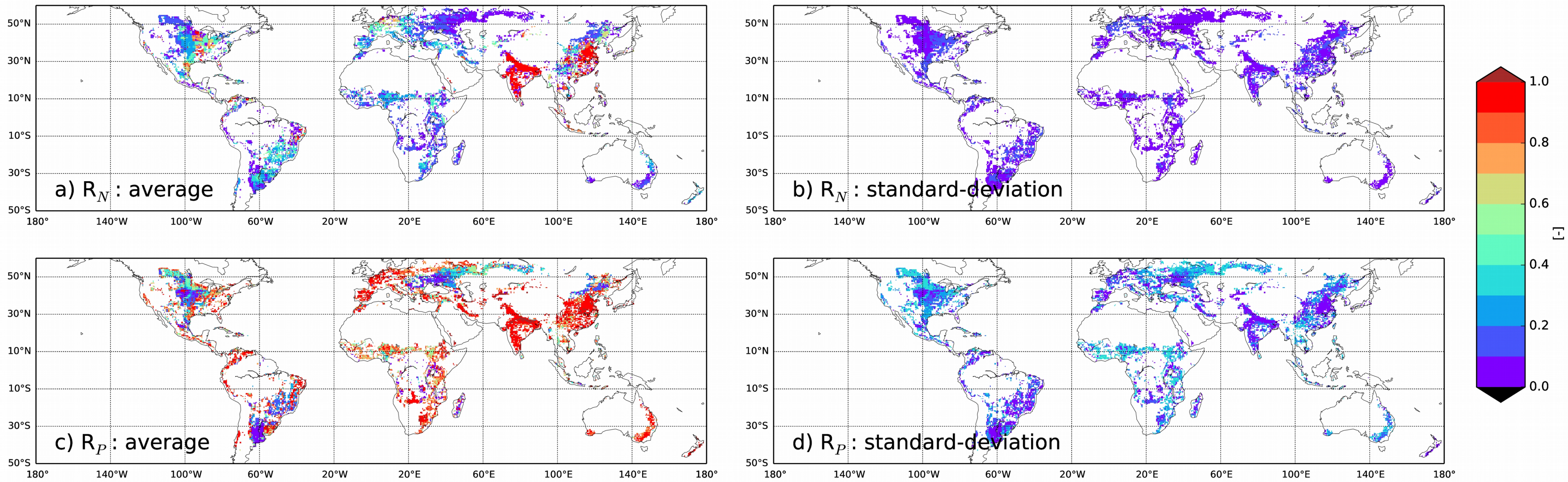

Figure S4. For maize, spatial distribution of R_N and R_P when N and P are considered as independent: average and standard-deviation of the 1000 replicates.

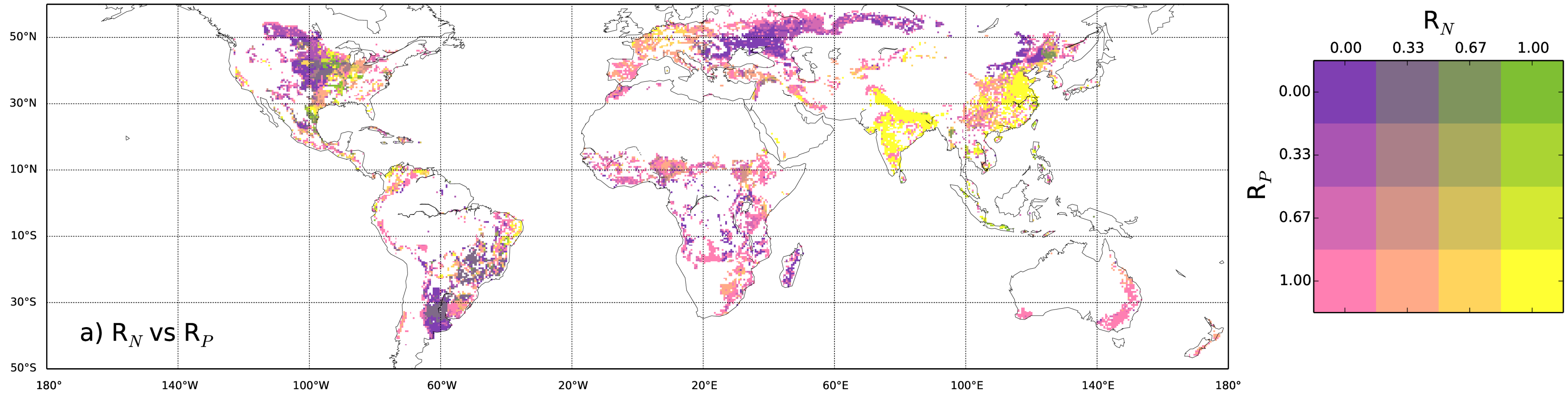

Figure S5. For maize, the spatial distribution of nutrient limitation when N and P are considered to be independent (bivariate plot of R_N and R_P).

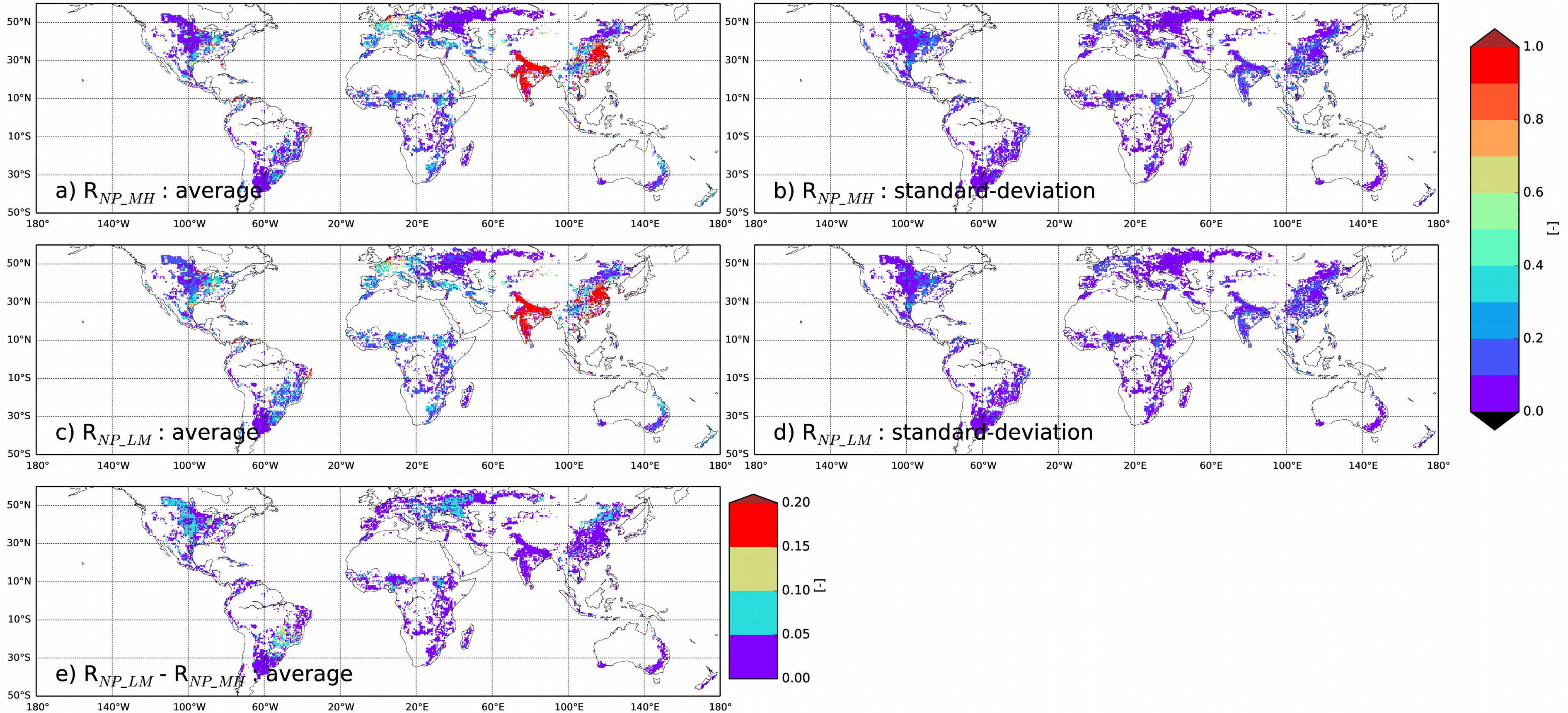

Figure S6. For maize, spatial distribution of R_{NP}: average and standard-deviation for both formalisms of interaction (a-b: MH; c-d: LM). The averaged difference of R_{NP} between LM and MH is also plotted (panel e).

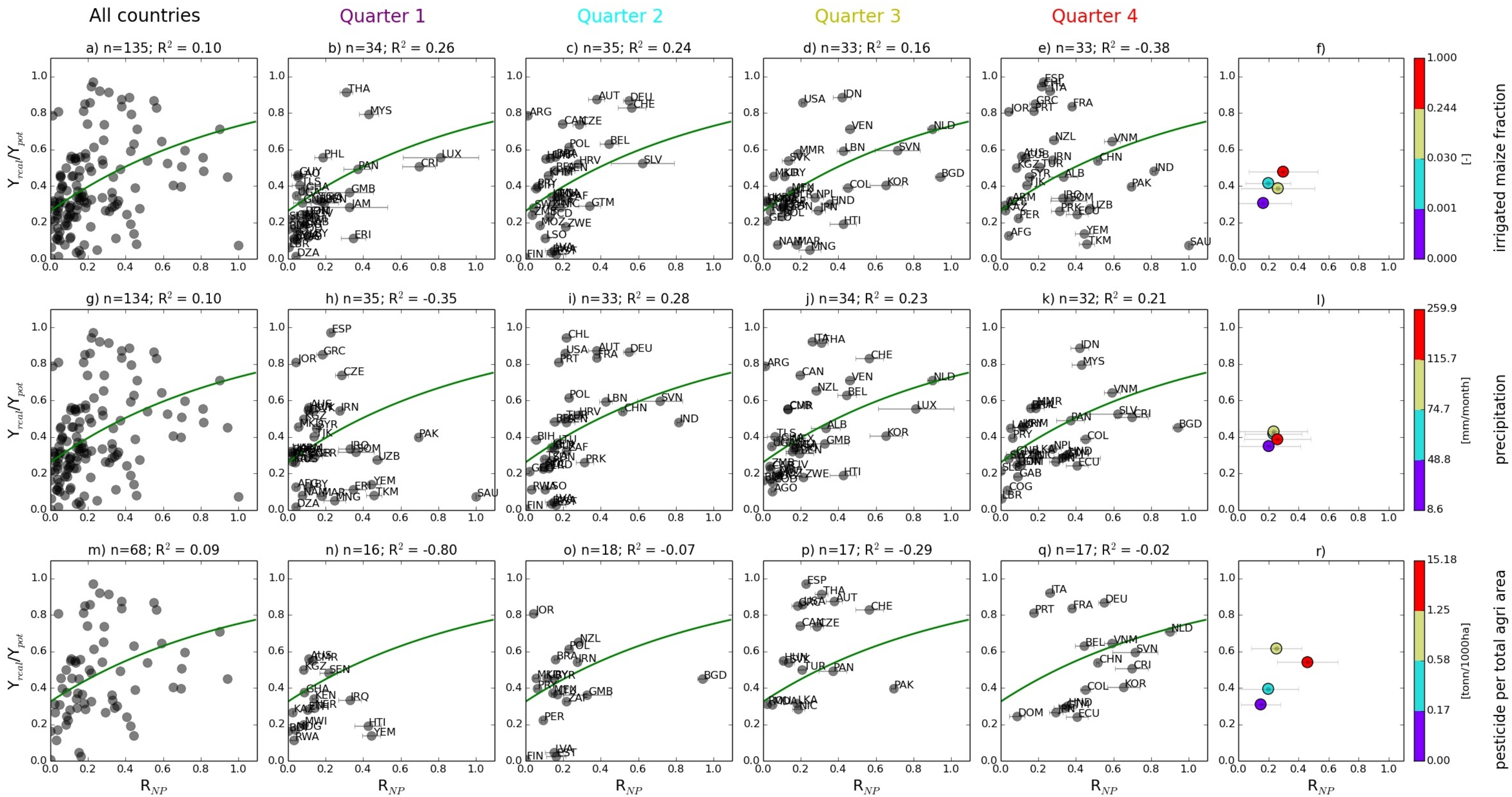

Figure S7. Scatterplots of the ratio Y_{real}/Y_{pot} provided by Mueller et al. (2012) vs. the simulated R_{NP} (here only computed by using the MH formalism for the purpose of simplicity) at the country scale for maize. Each dot corresponds to one country. In the extreme-left column, all countries 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* (*g: x* → *1-β.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.