# **Title:** Insights on nitrogen and phosphorus co-limitation in global croplands from theoretical and modelling fertilization experiments

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**Running title:** Nitrogen and phosphorus co-limitation in global croplands

**Abstract:**

Crossed fertilization additions are a common tool to assess nutrient interaction in a given ecosystem. Such fertilization experiments lead to the definition of nutrient interaction categories: e.g. simultaneous co-limitation, single resource response, etc~~.~~ ~~(Harpole et al., 2011)~~. While these categories are commonly used in literature, what each category implies ~~However, the implications of such categories~~ in terms of formalism of nutrient interaction ~~modeling are not~~ remains unclear. To this end, we developed a theoretical analysis of nitrogen (N) and phosphorus (P) fertilization experiments based on the computation of ratios between plant demand and soil supply for each nutrient. The theoretical analysis is developed following two mathematical formalisms of interaction: Liebig's law of minimum (LM) and multiple limitation hypothesis (MH). As results of the theoretical framework, we defined what values of the limitation ~~the values of the limitation the corresponding between most Harpole categories and~~ by each nutrient when considered alone in the control experiment (i.e. without additional nutrient supply) are required to make an ecosystem in a given category. We clarified which categories are compatible with each interaction formalism assumed (LM or MH). In particular, ~~W~~we showed that synergistic co-limitation could occur even using ~~Liebig's~~LM formalism under certain conditions, ~~as a function of the amount of N and P added in fertilization experiments.~~ e.g. if the ecosystem is N-limited in the control and if the amount of N added in the fertilization experiment is enough to switch the ecosystem into P-limitation. We then applied our framework with global maps of soil supply and plant demand for croplands to achieve their potential yield. This allowed us to estimate the global occurrence of each limitation category, for each of the possible interaction formalism. We found that a true co-limitation could affect a large proportion of the global crop area (e.g. ~42% for maize) if ~~multiple limitation hypothesis~~MH is assumed.

~~Our work clarifies the conditions required to achieve N and P co-limitation as function of the interaction formalism.~~ Combined with compilation of field trials in cropland, our study would improve our understanding of nutrient limitation in cropland at the global scale.

**Keywords:** nutrient limitation, nutrient interaction, nitrogen, phosphorus, cropland

## 1. Introduction

In global assessments of crop ecosystem productivity limitations by nutrients, nitrogen (N) and phosphorus (P) are sometimes considered independently (Peñuelas et al., 2013); or they are considered together but without focusing on how the interaction modulates the limitation (Mueller et al., 2012). N and P cycles interact strongly with different processes that are key to this coupling (Achat et al., 2016). The most commonly studied interaction is related to the limitation of plant growth by nutrients: an increase in organ biomass (mainly composed of carbon, C) requires a given amount of both N and P, to respect stoichiometrical constraints. The interaction between ~~carbon~~C and nutrients is usually represented by C:nutrient ratios for each organ. Plant growth is assumed to be limited when the demand for nutrients, estimated from C:nutrient ratios and C available for potential growth, is not satisfied by the supply of nutrient taken up by the plant. Due to incomplete knowledge about the mechanisms at the basis of the interaction and how these mechanisms are combined when integrating spatial scales (plant organ, individual, community, ecosystem) (Ågren et al., 2012; Davidson and Howarth, 2007; Sistla and Schimel, 2012; de Wit, 1992), the characterization of multiple element limitation remains an open scientific question. Two formalisms are generally used: Liebig's law of the minimum (LM) or the multiple limitation hypothesis (MH). In LM, plants are assumed to be limited by a single nutrient at a time, while in MH, it is assumed that plants adjust their growth patterns and thus they are co-limited by multiple nutrients simultaneously (Ågren et al., 2012). The MH formalism thus assumes that plants will mine the least available nutrient by using other resources. For instance, plants or groups of species growing in an ecosystem with a P-poor soil will invest C and N in the root system (and potentially to fungal mycorrhizae that form symbioses with plant roots (Ryan and

Graham, 2018)) to access more P (Davidson and Howarth, 2007). Both formalisms could be considered as macro-properties that reflect the same plant adjustments processes but, depending on the conditions, those adjustments may lead to an emerging behaviour that verifies one or the other formalism (Ågren et al., 2012). The further the supply of an essential nutrient deviates from a conceptual and theoretical optimum stoichiometry of plants, the more plants will follow the LM formalism (Ågren et al., 2012). LM is commonly assumed in many studies and is for instance used in most large-scale models dealing with multiple nutrient limitations (Barros et al., 2004; Goll et al., 2012; Mueller et al., 2012).

One way to assess the current nutrient limitation empirically is to provide single applications of +N, +P and +NP and to measure the increase in ecosystem productivity as compared to a control trial without any application. Such experiments are usually called fertilization experiments. By definition (Harpole et al., 2011), there is a *true* NP co-limitation when the ecosystem is observed to respond to combined N and P addition only, or to both N and P when added separately. The different categories of nutrient limitation are summarized in Harpole et al. (2011) and in Table 1.~~ Fertilization experiments are common in natural ecosystems and meta-analysis of these experiments have provided a global picture of nutrient limitation in natural ecosystems (Augusto et al., 2017; Elser et al., 2007; Harpole et al., 2011). Results from recent meta-analyses have shown that a true co-limitation is found in 28-42% of the studies (Augusto et al., 2017; Harpole et al., 2011). This challenges the view that plants are generally limited by one nutrient at a time (i.e. LM). However, w~~ While~~ch~~ these categories~~ of nutrient limitation~~ are commonly used in literature, what each category implies in terms of formalism of ~~consistent with each~~ nutrient interaction~~ formalism (LM and MH)~~ remains unclear~~is not clear~~. In particular, we aim here to understand which categories are prevented and which ones are more or less promoted by the interaction formalism assumed. To this end, we provided a theoretical framework of N and P fertilization experiments based on the computation of ratios between plant demand and soil supply for each of the two nutrients. The theoretical analysis is developed for two mathematical formalisms of interaction (LM or MH). This allowed us to define, for each formalism, the corresponding between Harpole categories and the values of the limitation by each nutrient when considered alone. Then, we analytically investigated how the choice of formalism modifies the NP limitation.

Finally, we applied our framework to the case of nutrient limitations in croplands. The justification of this choice is twofold: first, nutrient limitation is a key question in croplands at the global scale. For instance, (MacDonald et al., 2011) showed that 30% of cropland are characterized by negative soil P budget. Mueller et al. (2012) showed that ~70% of the cropland where potential yield is not achieved could close their yield gap by increasing nutrient inputs. Second, ~~For croplands,~~ experiments with **single** and crossed N and P fertilizer applications ~~of fertilizer~~ (as defining “fertilization experiments”) are not so~~as~~ common in croplands~~as those for natural ecosystems~~. This prevents us from having a global picture of N and P limitation based solely on observations, contrary to what was done in natural ecosystems (Elser et al. 2007, Harpole et al. 2011). Indeed, in cropland, fertilization experiments are usually characterized by one single addition for N (e.g. Di Paolo and Rinaldi, 2008; Salvagiotti et al., 2008) while for P, the same amount of fertilizer is applied each year for decades in so-called ~~, On the other hand and it is usually difficult to retrieve the application level before the experiment (e.g. Deguchi et al., 2017; Restelatto et al., 2017), which prevents an accurate definition of the control in these cases.~~long-term field experiments with crops responding both to the annual supply of fertilizer and to the cumulative effect on soil P availability~~are common in croplands~~

(~~especially for P,~~ e.g. Bai et al., 2013). This makes difficult to decipher the contribution of each nutrient. Moreover, in such experiments, many P treatments are tested and ~~but~~ for a given treatment,~~;~~ the same amount of fertilizer is applied each year for decades, which makes the limitation in the long-term trial somehow non-representative to nutrient limitation happening in the surrounding fields~~precludes analysis of current limitation~~. ~~Crossed fertilization additions are difficult to decipher from multi-nutrients and repeated fertilizer applications, as usually performed in croplands. To our knowledge, no meta-analysis of NP limitation in cropland field trials exists, which prevents us from having a global picture of N and P limitation based solely on observations.~~ When a single application is the focus of a study, it is usually difficult to retrieve the application level before the experiment (e.g. Deguchi et al., 2017; Restelatto et al., 2017), which prevents an accurate definition of the control in these cases. Thus, our analysis, based on a theoretical analysis, is particularly adapted to investigate nutrient limitations in cropland. We applied our framework on global spatially explicit computations of soil supply and plant demand of N and P for croplands to achieve their potential yield, in order to assess the occurrence of co-limitation in croplands for each interaction formalism.~~we provided a theoretical framework of N and P fertilization experiments based on the computation of ratios between plant demand and soil supply for each of the two nutrients. The theoretical analysis is developed for two mathematical formalisms of interaction (LM or MH). This allowed us to define the corresponding between Harpole categories and the values of the limitation by each nutrient when considered alone. We analytically investigated how the choice of formalism modifies the NP limitation.~~ Potential yield is here defined as the yield achieved without limitations of water and nutrients and without pest/diseases.

~~In this work, Finally, we applied our framework on global spatially explicit computations of soil supply and plant demand of N and P for croplands to achieve their potential yield, in order to assess the occurrence of co-limitation in croplands for each interaction formalism.~~

## 2. Theoretical framework

Based on a framework commonly used in global studies (Goll et al., 2012; Kvakić et al., 2018), we defined the limitation of a nutrient considered alone as the ratio of its soil supply (S) and the demand by the plant to achieve its potential biomass (D):

$$R_N = min(1, \frac{S_N}{D_N}) \quad \text{(Eq.1)}$$

$$R_P = min(1, \frac{S_P}{D_P}) \quad \text{(Eq.2)}$$

where $S_X$ and $D_X$ correspond to the supply and demand of the nutrient X, respectively (in kgX/ha/yr) with X is in {N,P}.

Crossed fertilization experiments are a common tool to assess nutrient limitation on a given site. They correspond to changes in nutrient supply in different combinations from the control ($E_1$): addition of N alone ($E_2$), P alone ($E_3$) or N and P together ($E_4$) (Fig. 1). Based on the above equations defining the limitations of N and P (Eqs 1 and 2, respectively), theses changes in nutrient supply translate into limitations of each nutrient for each experiment E as follows:

$$E_1: \quad R_N(E_1) = min(1, \frac{S_N}{D_N}) \text{ and } R_P(E_1) = min(1, \frac{S_P}{D_P}) \quad \text{(Eq.3)}$$

$$E_2: \quad R_N(E_2) = min(1, \frac{S_N + A_N}{D_N}) \text{ and } R_P(E_2) = min(1, \frac{S_P}{D_P}) \quad \text{(Eq.4)}$$

$$E_3: \quad R_N(E_3) = min(1, \frac{S_N}{D_N}) \text{ and } R_P(E_3) = min(1, \frac{S_P + A_P}{D_P}) \quad \text{(Eq.5)}$$

$$E_4: \quad R_N(E_4) = min(1, \frac{S_N + A_N}{D_N}) \text{ and } R_P(E_4) = min(1, \frac{S_P + A_P}{D_P}) \quad \text{(Eq.6)}$$

with $A_N$ and $A_P$ corresponding to the increase of N and P soil supply following addition of

N and P, respectively.

In the above framework, each nutrient is considered alone while the two nutrients interact. An ecosystem is thus defined by its NP limitation, called $R_{NP}$ in the following. Two formalisms of interaction have been here considered to compute $R_{NP}$ from $R_N$ and $R_P$: multiple limitation hypothesis (called MH in the following, Eq.7) or Liebig's law of the minimum (LM, Eq.8):

$$R_{NP\_MH}(E_i) = R_N(E_i) * R_P(E_i) \quad \text{(Eq.7)}$$

$$R_{NP\_LM}(E_i) = min(R_N(E_i), R_P(E_i)) \quad \text{(Eq.8)}$$

where $E_i$ is the experiment *i*. In MH, the limitations when the nutrients are considered independently ($R_N$ and $R_P$) are multiplied to compute the NP limitation while in LM, the smallest one is selected.

We analytically investigated to which extent the choice of the formalism has an effect on the value of $R_{NP}$ for a given ($R_N$, $R_P$) couple (Fig. 2a-c). We also investigated how the formalism modulates the increases in $R_N$ and $R_P$ required to alleviate the NP limitation (here represented by an increase in $R_{NP}$ to reach an arbitrary value of 0.75) (Fig. 2d-i). Such increases are called $\Delta R_N$ and $\Delta R_P$ in the following. $\Delta R_N$ and $\Delta R_P$ corresponds to the smallest increase in $R_N$ and $R_P$ required at the same time to make $R_{NP}$ equal to 0.75 (Fig. 3). We found that the largest differences in $R_{NP}$ between the LM and MH mathematical formulations are obtained for comparable $R_N$ and $R_P$ values ($R_N$~$R_P$) and both within [0.25-0.75] (Fig. 2c). In fact, x in [0.25-0.75] and x~y mathematically maximize the difference between x*y and min(x,y). In addition, the largest differences in $\Delta R_N$ between LM and MH occur for $R_N$<$R_P$ and both $R_N$ and $R_P$ lower than 0.75 (Fig. 2f). Symmetric results are obtained for $\Delta R_P$ (Fig. 2i).

In fertilization experiments, nutrient limitation is assessed by looking at the change in productivity (Δpro) according to the addition of P alone ($\Delta pro_{+P}$), N alone ($\Delta pro_{+N}$) or N and P together ($\Delta pro_{+NP}$). Δpro is here not expressed in absolute change but ~~expressed~~ relatively to the potential productivity (i.e. without any limitation). Harpole et al. (2011) defined different categories of limitation when considering the two nutrients in interaction. Each category is entirely defined by: i) the character null or non-null of $\Delta pro_{+N}$ and $\Delta pro_{+P}$ and ii) the relationship between $\Delta pro_{+NP}$ and ($\Delta pro_{+N}+\Delta pro_{+P}$) (column 3 of Table 1). Following Harpole et al. (2011), co-limitation exists when the increase in productivity following the addition of N and P together is strictly greater than the sum of increases in productivity when each nutrient is added alone (i.e. $\Delta pro_{+NP} > \Delta pro_{+N} + \Delta pro_{+P}$ ). Any co-limitation is defined as a synergistic relationship. A given co-limitation is in addition considered as true if the responses to +N and +P are either both equal to 0 (i.e. $\Delta pro_{+N}=0$ and $\Delta pro_{+P}=0$ , simultaneous co-limitation, category A in Table 1) or both non-null (i.e. $\Delta pro_{+N}\neq 0$ and $\Delta pro_{+P}\neq 0$ , independent co-limitation, category B).

Here, we assumed that the change in productivity following the addition of +N, +P or +NP is equal to the change in $R_{NP}$ following the nutrient addition, i.e.:

$$\Delta pro_{+N} = R_{NP}(E_3) - R_{NP}(E_1) \quad \text{(Eq.9)}$$

$$\Delta pro_{+P} = R_{NP}(E_2) - R_{NP}(E_1) \quad \text{(Eq.10)}$$

$$\Delta pro_{+NP} = R_{NP}(E_4) - R_{NP}(E_1) \quad \text{(Eq.11)}$$

where $E_i$ is the experiment *i* (Fig. 1). This is a key assumption in our approach based on two simplifications described in details in the following. First, ~~T~~through these equations, we assumed that the productivity of a given experiment is proportional to $R_{NP}$ and that the slope of this relationship is equal to 1. In fact, a slope equal to 1 is not necessary to develop the theoretical analysis described in Text S1. As mentioned before, Harpole categories are defined through i) the character null or non-null of $\Delta pro_{+N}$ and $\Delta pro_{+P}$ and ii) the relationship between $\Delta pro_{+NP}$ and ($\Delta pro_{+N}$+$\Delta pro_{+P}$). These definitions are true even if the productivity of each experiment (and thus the different Δpro) is divided by the same slope. We keep here a slope equal to 1 for the sake of simplicity. Second, Eq.9-11 also imply that the relationship of proportionality between the productivity and $R_{NP}$ is true for all values of $R_{NP}$, in the range [0-1]. In reality, the productivity vs. limitation relationship is very likely asymptotic (e.g. Bai et al., 2013). Here, we may approach this non-linearity by assuming a linear relationship for $R_{NP}$ in [0,*thresh*] where *thresh* is a given threshold; followed by a plateau for $R_{NP}$ in [*thresh*,1] (which corresponds to assume a slope of 0 for the productivity vs. limitation relationship). The corresponding between Harpole categories and the values of $R_N$ and $R_P$ found at the end of our theoretical analysis are still valid in this more general case, but on the restricted range of $R_N$ and $R_P$ values between 0 and *thresh* (in this case, the value 1 used as boundary for $R_N$ and $R_P$ in category definitions – columns 4 and 6 of Table 1 – should be replaced by *thresh*). For the sake of simplicity, we keep in the following 1 as the high boundary and came back on this point in the Discussion.

For each interaction formalism (Eq.7 or 8), we translated the Harpole category definition (column 3 of Table 1) into conditions on $R_N(E_1)$ and $R_P(E_1)$ thanks to Eq.9-11. This means that we are able to define most Harpole categories in terms of limitation of each nutrient considered alone in the control experiment. $R_N(E_1)$ and $R_P(E_1)$ are respectively called $R_N$ and $R_P$ in the following. The equation manipulation is described in details in

Text S1 for both MH and LM.

We showed in particular that to belong to the category “independent co-limitation” (category B in Table 1) with MH formalism, an ecosystem has to be characterized by both $R_N$ and $R_P$ in (~~]~~0,1)~~[~~ (a parenthesis instead of a square ~~reversed~~ bracket used in an interval means here that the corresponding endpoint is excluded from the interval; e.g. R in [0,1) means 0≤R<1). All other categories (A, C-G) require at least one ratio equal to 0 or 1: e.g. serial limitation N (category C) requires $R_N$ equal to 0 and $R_P$ in (~~]~~0,1)~~[~~ (Table 1). Categories E,F,G are defined by $\Delta pro_{+NP} = \Delta pro_{+N} + \Delta pro_{+P}$ and we showed that this requires at least one ratio equal to 1 with MH formalism (Text S2).

We showed that the formalism LM cannot represent true co-limitation, except in the very specific category A (i.e. $R_P = R_N \neq 1$). We found that synergistic co-limitation alone (categories C and D) can occur with LM but to be in these categories, the amount of N (if the control is N limited) or P (if the control is P limited) added in the fertilization experiments should be large enough to remove the initial limitation.

Conclusions of this analysis are summarized in Table 1 (columns 4 for MH and column 6 for LM).

## 3. Quantifying co-limitation in croplands at the global scale

### 3.1. Computation of spatially explicit $R_N$ and $R_P$

We computed spatially explicit maps of $R_N$ and $R_P$ in croplands (0.5° latitude x 0.5° longitude) based on the computation of nutrient demand and soil supply. We then applied the above described theoretical framework on these $R_N$ and $R_P$ values to classify each grid-cell according to Harpole categories for the two interaction formalisms.

The computation of supply and demand maps used to estimate $R_N$ and $R_P$ are described below. To summarize, plant nutrient demand is based on nutrient harvest index data from the literature combined with spatially explicit distribution of crop potential yield ($Y_{pot}$) (Mueller et al., 2012). The soil N supply has been approached by using a soil N budget taking into account fertilizer (mineral and organic), atmospheric deposition, biological fixation, and losses by ammoniac volatilization (Bouwman et al., 2011a), while the soil P supply is assessed by a potential root uptake model that accounts for soil P diffusion and soil P legacy effects. Such supply and demand estimates are representative to a growing season time-scale. More details about the computation of each variable can be found in Table S1.

Following Kvakić et al. (2018), demands for N and P to reach potential yields ($D_N$ and $D_P$, in kg(N or P)/ha/yr) were derived from the combination of i) fixed parameters related to distribution of carbon (C) and nutrients between the different plant organs at maturity found in the literature and ii) spatially explicit potential yield ($Y_{pot}$, in kgC/ha/yr):

$$D_X = Y_{pot} \cdot \left( \frac{X_{\%,grain}}{XHI} + \frac{X_{\%,root} \cdot RSR}{HI} \right) \quad \text{(Eq.12).}$$

XHI corresponds to the nutrient harvest index (i.e. the ratio between the nutrient content of grain and the nutrient content of shoot, no unit), HI is the harvest index (i.e. the ratio between the carbon content of grain and the carbon content of shoot, no unit), RSR is the root/shoot ratio (no unit) and $X_{\%,grain}$ and $X_{\%,root}$ are nutrient concentrations (kg(N or P)/kgC) for grain and root, respectively. Kvakić et al. (2018) has shown that a XHI-based method provides similar demand estimates compared with others based on the nutrient concentration of all plant organs or QUEFTS (Sattari et al., 2014). The definition of the parameters used in Eq.12 (XHI, HI, RSR, $X_{\%,grain}$, $X_{\%,root}$) is based on nutrient and C biomass of different plant organs. These definitions as well as values used in the study are given in Table S2. Spatially constant values are here used. In particular, as the aim of our study is to assess nutrient limitation, we used organ concentrations derived from field experiments in stressed conditions in a multitude of climatic and socio-economic environments (Van Duivenbooden (1992) and Table S2). Details about the Eq.12 are given in Text S3.

The potential yield ($Y_{pot}$) is provided by Mueller et al. (2012) in tons of dry matter per hectare. In Mueller et al. (2012), the world grid-cells are divided into climate bins, defined by different combinations of growing degree days and amount of yearly precipitation; and within a climate bin, the potential yield characterizing this bin is defined as the area-weighted 95th percentile of the grid-cell observed yields.

The supply of P ($S_P$, in kgP/ha/yr) corresponds to the sum of a potential root uptake and a prescribed fraction (called $\alpha$) of the inorganic content of total P fertilizer applied in the year considered Kvakić et al. (2018). The potential root uptake is determined by soil P availability and monthly root length density, following some assumptions about P diffusion in soil (Text S4). Soil P availability is derived from the current global distribution of soil P, as in Kvakić et al. (2018). The global distribution of soil P was determined by combining information on farming practices, soil P dynamics, soil biogeochemical background, climate effect on soil P dynamics, etc., as well as the past variation of some of these drivers (Ringeval et al., 2017). Thus, we explicitly considered the soil P legacy effect, as it has been shown to be an important process (Ringeval et al., 2014; Sattari et al., 2012). Root characteristics parameters, root biomass at harvest (derived from $Y_{pot}$, RSR and HI) and seasonality in root biomass (computed by 0 and 1 and derived from simulations of one global gridded crop model, LPJmL (von Bloh et al., 2018)) were combined to compute monthly root length density (Text S4). As in Kvakić et al. (2018), we assumed that a fraction of the applied fertilizer P is directly available to the plant in the same growing season, thus bypassing the P diffusion pathway.

Following Bouwman et al. (2017), the supply of N is approached by the soil N input of the year considered (chemical and organic fertilizer, atmospheric deposition and fixation) minus losses corresponding to $NH_3$ volatilization. Datasets were provided by Bouwman et al. (2011). Except in few recent studies (e.g. ten Berge et al. (2019) that focuses on sub-Saharan Africa), N applied in previous years is generally neglected due to the higher lability of N than P. We follow the same assumption as it is common in global modelling approaches (Bouwman et al., 2017; Conant et al., 2013; Lassaletta et al., 2014). Mineralization of soil organic N was also neglected as under steady state conditions it is expected to be compensated by N immobilisation in soil microbial biomass. N leaching was also neglected as in Bouwman et al. (2011), it is assumed that N leaching concerns only the surplus of annual soil agronomic N budget and occurs at the end of the growing season.

We recognize that the use of constant parameters at the global scale in the computation of supply and demand is too simple (Sadras, 2006). This is in particular true with respect to plant adjustments to nutrient limitations (Colomb et al., 2007) which are susceptible to modify nutrient organ concentrations. Also, some agronomic management such as cultivar diversity across World regions is susceptible to modify parameters, in particular HI. However, both plant adjustments (Franklin et al., 2012) and the effect of cultivar diversity on allocation (Folberth et al., 2016) are difficult to represent at the global scale. Considering grid-cells independently in our uncertainty analysis (Text S5) made these parameters artificially vary in space.

Each term ($S_N$, $D_N$, $S_P$, $D_P$) is spatially explicit at half-degree resolution. An uncertainty related to each term has been considered (Text S5). Maize, rice and wheat are considered in this study (see the crop-dependent terms in Table S1) and the ratios computed are representative of the year 2000. Only grid-cells for which $R_P$ and $R_N$ could be computed are considered, which determines the crop area and the global crop production considered in our study (Table S3). In the Main Text, a specific focus is made on maize because it is the most widespread across latitudes. ~~Caveats of our approach are discussed in Section 4.~~

### 3.2. Effects of formalism choice on global NP limitation

Spatial distributions of $R_N$, $R_P$ as well as $R_{NP}$ computed with both formalisms are described and discussed in Text S6. The relationship between $R_{NP}$ and actual yield gap provided by statistical approaches ($Y_{real}/Y_{pot}$, with $Y_{real}$ being the actual yield) has been investigated at country scale (Text S7).

The effect of formalism choice on the global values of $R_{NP}$, $\Delta R_N$ and $\Delta R_P$ depends on the distribution of grid-cells in the ($R_N$, $R_P$) space (grey dots in Fig. 2c,f,i). Even though significant, the difference between LM and MH in global $R_{NP}$ is small (Table 2). This is explained by a small number of grid-cells (~2%) characterized by conditions that maximize the difference between the LM and MH mathematical formulations (i.e. comparable $R_N$~$R_P$ and both within [0.25-0.75], see above). Finally, ~55% of the grid-cells are characterized by a difference LM – MH which is smaller than the uncertainty computed with a given formalism (LM or MH).

The global averages of ΔR computed with MH ($\Delta R_N$=0.44±0.00, $\Delta R_P$=0.30±0.01) are larger than those computed with LM ($\Delta R_N$=0.37±0.00, $\Delta R_P$=0.25±0.01). Numbers provided correspond to averages and standard-deviations among 1000 replicates of global averaged $R_N$ and $R_P$. Replicates differ between them by considering uncertainties in parameters at the basis of the computation of supply and demand (Text S5). It was analytically shown (Fig. 2) that the LM – MH difference of $\Delta R_N$ is maximal for a combination of small $R_N$ and medium $R_P$, as encountered in the centre of the USA (Text S6 and Fig. S1). Large differences are also noticeable in regions with high limitations of both nutrients, such as the Western Russian Federation and Ukraine.

### 3.3. Occurrence of Harpole categories

We computed the occurrence of each Harpole category by using conditions ~~on~~in terms of $R_N$ and $R_P$, as described in Table 1. We checked that these occurrences are equal to the ~~values~~occurrences found when: ~~numerical~~modelling fertilization experiments are performed, $R_{NP}$ are computed for each experiment (Eq.7-8) and Eq.9-11 are then applied. The ~~I~~increase of N and P supply ($A_N$ and $A_P$) in fertilization experiments are here equal to 30kgN/ha/yr and 5kgP/ha/yr, respectively and are spatially homogeneous for all cropland around the World. While our theoretical framework was initially developed for productivity (Section 3.2), we applied it here to cropland yield, which is consistent with the assumption of fixed harvest index as described in Section 3.1.

With the formalism MH, we found that true co-limitation occurs in 41.7±0.6 % of the global crop area for maize, via independent co-limitation (category B in Table 1). This category is found in the USA, South America, the Western Russian Federation and Ukraine (Fig. 4a). As showed theoretically, to belong to that category a crop has to be characterized by both $R_N$ and $R_P$ in (~~]~~0,1)~~[~~. In our simulations, these conditions occur for ~42% of the maize crop area.

Synergistic co-limitation alone (categories C and D) occurs for 6.7±0.3 % of the global maize crop area and this is only explained by serial limitation N (category C, dark blue in Fig. 4a): no serial limitation P was found in our numerical application. This can be explained by the fact that $R_P$ (contrary to $R_N$) is never null in our simulations because of the soil P legacy taken into account in our approach (Ringeval et al., 2017). This also prevents simultaneous co-limitation (A) from being found. The occurrence of co-limitation at the global scale varies between crops (41.7±0.6 % for maize, 32.5±0.4 % for wheat and 18.7±0.8 % for rice, not shown). Except for few regions (e.g. India), grid-cells where the three crops are grown belong to the same limitation category for all crops (not shown): the difference in occurrence of co-limitation between crops is mainly explained by the crop-specific global distribution.

As theory shows (Text S1), the formalism LM cannot represent true co-limitation, except in the very specific category A (i.e. $R_P=R_N\neq1$), which is never encountered in our study (Fig. 4b and Table 1). We found that synergistic co-limitation alone (categories C and D) can occur in more than 15% of the global maize area with LM. However, this number is sensitive to the amount of N and P added in the fertilization experiments (called respectively $A_N$ and $A_P$ in Fig. 1). E.g. a cropland which is initially P-limited is classified in the category D if the amount of P added ($A_P$) is sufficient to remove the P limitation (i.e. the cropland becomes N limited); otherwise, it belongs to the category F (Table 1).

## 4. Discussion

~~Previous studies estimating the occurrence of co-limitation in natural ecosystems were based on fertilization experiments performed around the World and provide some insights into the best way to represent the NP interaction (Elser et al., 2007; Harpole et al., 2011). Studies reviewing such experiments (characterized by a single application of nutrient) are not available for cropland, so we chose another strategy by computing the occurrence of co-limitation for each interaction formalism.~~ Our work ~~also~~ clarifies the mathematical conditions in terms of supply/demand ratios required to place an ecosystem into a category of nutrient limitation, as defined by Harpole et al. (2011). In particular, we found that synergistic co-limitation can occur with Liebig's law of the minimum under certain conditions that are functions of the amount of N and P added in fertilization experiments, as already suggested by Ågren et al. (2012). While Liebig's law of the minimum is based on the limitation by a single nutrient at a time, it allows synergistic co-limitation to happen, which could be counter-intuitive. We found that, if multiple limitation hypothesis is the most appropriate way to represent nutrient interaction, co-limitation should occur for ~50% of the maize crop area (42% of true and synergistic co-limitation + 7% for synergistic alone co-limitation). The occurrence of true co-limitation in croplands would be of a similar magnitude to those reported for natural ecosystems (28% in ~~The percentage of true co-limitations found here is higher than those reported for natural ecosystems in~~ Harpole et al. (2011), 42% in ~~(28%) but similar to values found in~~ Augusto et al. (2017)). ~~(42%). However, these previous figures should be compared with caution as they were not obtained with the same methodology.~~

More investigations are needed to precise the real occurrence of co-limitation in croplands. To do so, a method based on observations, as performed for natural ecosystems (Harpole et al. 2011), is required. As mentioned in the Introduction, crossed single fertilizer application are not as common in croplands as in natural ecosystems. Or at least, they exist but are exploitable with difficulties. A huge work is required to select studies that deal with single application for both nutrients, that provide information about the previous applications (that determines the control), and that are characterized by a control that is representative to the region where the trial occurs. Once these studies have been selected, they can be compiled. If numerous enough, ~~It is also worth noting that change in the plant community can occur consecutively to fertilizer application  in natural ecosystems, which does not happen in cropland systems as they are mostly single crop. Experiments with single increase of fertilizer in cropland have to be selected and then compiled. Such meta-analysis~~they would tell us if co-limitation is really common in croplands, suggesting e.g. that farming practices tend to promote co-limitation. On the ~~contrary~~opposite, the absence of such co-limitation would suggest that human perturbation of nutrient cycles pushes the crop plant outside of its adaptation capacity. Contrary to what happens in natural ecosystems, change in the plant community cannot occur consecutively to fertilizer application in cropland systems as they are mostly single crop. Thus, co-limitation in cropland should be considered as reflecting plant adaptations, e.g.: plant can invest nutrient in excess to access the limiting nutrient. And an absence of co-limitation would suggest that plant cannot adapt to perturbations.

The occurrences of the different limitation categories that we provided are a function of the spatial distribution of $R_N$ and $R_P$, as posited by our theoretical framework. However, these maps are prone to uncertainty due to simplifications in our modelling approach. As mentioned in Section 3.1, some simplifications are related to the use of constant parameters at the global scale in the computation of supply and demand while plant adjustments and some farming practices are susceptible to modify them. Global changes are also very likely modifying yield and grain composition (e.g. Long et al., 2006; Müller et al., 2014) and this effect was not considered in our study which does not simulate temporal changes in nutrient limitation.

~~First, we recognize that the use of constant parameters at the global scale in the computation of supply and demand is too simple (Sadras, 2006) in particular with respect to plant adjustments to nutrient limitations (Colomb et al., 2007) which are susceptible to modify nutrient organ concentrations. However, the aim of our study is to assess nutrient limitation and thus, we used organ concentrations derived from field experiments in stressed conditions (Van Duivenbooden (1992) and Table S2). Global changes are also very likely modifying yield and grain composition (e.g. Long et al., 2006; Müller et al., 2014) and this effect was not considered in our study which does not simulate temporal changes in nutrient limitation. Besides, it is worth nothing that considering grid-cells independently in our uncertainty analysis (Text S5) made these parameters artificially vary in space.~~

Other simplification is related to the ~~We~~ use of~~d~~ potential yield provided by statistical methods based on maximum attainable yield within climate bins (Mueller et al. 2012)~~,~~. ~~but s~~Such approaches have difficulty distinguishing irrigated and rainfed crops and thus, the here used $Y_{pot}$ could be in fact water-limited in some places (van Ittersum et al., 2013b). Updates to the statistical methodology are ongoing to improve the separation between water-limited and irrigated yield potential (Mueller, personal comm.). Alternative estimates of potential yield such as the ones simulated by Global Gridded Crop Models are very likely prone to huge uncertainties too (Müller et al. 2017).

~~We also did not consider some agronomic managements that are susceptible to modulate nutrient limitation. In particular, we did not consider cultivar diversity across World regions. Such diversity is susceptible to modify parameters (in particular, HI) which are considered constant in space in our approach. However, cultivar diversity is difficult to consider at the global scale and up to now, it was mainly investigated through spatial variability in phenological development (van Bussel et al., 2015). To a much lesser extent, the effect of cultivar diversity on allocation (e.g. through variability in harvest index, susceptible to modify the here compute nutrient demand) was taken into account (Folberth et al., 2016). Also, some effects of crop rotation on nutrient limitation were not considered in our study. E.g. crop rotation can modulate the soil P availability because of difference in the strategies developed for enhancing nutrient acquisition among crops (Redel et al., 2007) and this effect was neglected here. However, N fixation by leguminous that can be incorporated within the crop rotation with cereals was indirectly considered in our study: our computation of N supply was not a function of crop (Table S1) and thus, the N supply budget encompasses an term for N fixation by leguminous occurring in the same grid-cell as cereals (Bouwman et al., 2011).~~

In our approach, the limitation of potential yield is computed by considering current farming practices to derive the supply. Current practices could be influenced by other limiting factors: e.g. if a crop is water limited, farmers can adapt their practices and reduce their nutrient applications accordingly. Sensitivity tests where the demand would be derived from actual yield (instead of potential yield, as in the Main Text) could help in the determination of areas where other limiting factors might play a role (Fig. S2). The next step is to consider more limiting factors together.

Our theoretical analysis has also few caveats. In particular, we assumed a linear relationship between $R_{NP}$ and the productivity of each experiment (Eq. 9-11). As underlined in the method section, our conclusions are still valid if we assumed a linear relationship up to a value *thresh* if *thresh* replaces 1 in the definition categories given in Table 1. The value *thresh* is nevertheless theoretical because the calculated nutrient limitation ($R_N$, $R_P$, $R_{NP}$) has no physical meaning and is disconnected from physical measure of e.g. soil P content (Olsen P, etc.). The fact that the transition between linear and plateau regimes occurs for the same $R_{NP}$ (1 or *thresh*) globally should be an acceptable assumption as we took into account the spatial variation in soil properties to compute the soil nutrient supply.

In our analysis, we computed ~~Our computation of~~ $\Delta R_N$ and $\Delta R_P$, i.e. the increase in $R_N$ and $R_P$ required to increase $R_{NP}$ up to 0.75 and assessed how the choice of the interaction formalism has an effect on $\Delta R_N$ and $\Delta R_P$. ~~is based on the minimum “physiological” needs for plants. Behind the multiple limitation's mathematical formalism, an increase in $R_{NP}$ can be achieved for different combinations of increases in N and P (i.e. for different couples ($\Delta R_N$, $\Delta R_P$)): despite non-substitution at the molecular or cellular level (Sinclair & Park, 1993), one element can partly compensate for the other at the plant scale. Here, we considered only one couple (Fig. 3), while external variables such as the price or the ease of access to fertilizers will also influence the farmer's choice and could make him/her select another NP combination. This should be taken into account in future attempts to make link with scenarios of nutrient management and policy more straightforward.~~ $\Delta R_N$ and $\Delta R_P$ could be translated to increase in soil supply by considering nutrient demand in each grid-cell. However such change in supply cannot be easily translated into a change in fertilizer, since our supply estimates take into account some processes occurring after the fertilizer application: for P, we take into account the dynamics of P in soil (diffusion and root uptake) while for N, we allow for $NH_3$

volatilization. Our nutrient requirement calculation is driven solely by nutrient limitation, independently of yield gap, contrary to previous estimates based on: soil quality indicators (with no distinction between N and P) (Fischer et al., 2012; Pradhan et al., 2015), statistical relationships between fertilizer application and yield (Mueller et al., 2012) or “N uptake gaps” based on yield gap and minimal/maximal values of the physiological N efficiency in aboveground biomass derived from the QUEFTS model (ten Berge et al., 2019; Schils et al., 2018). More generally, our nutrient limitation is not straight connected to the yield gap because the actual yield is not used in our computation. It is interesting to note that our computation of $\Delta R_N$ and $\Delta R_P$ is based on the minimum “physiological” needs for plants. Behind the multiple limitation's mathematical formalism, an increase in $R_{NP}$ can be achieved for different combinations of increases in N and P (i.e. for different couples ($\Delta R_N$, $\Delta R_P$)): despite non-substitution at the molecular or cellular level (Sinclair & Park, 1993), one element can partly compensate for the other at the plant scale. Here, we considered only one couple (Fig. 3), while external variables such as the price or the ease of access to fertilizers will also influence the farmer's choice and could make him/her select another NP combination. This should be taken into account in future attempts to make link with scenarios of nutrient management and policy more straightforward.

~~Our theoretical analysis has also few caveats. In particular, we assumed a linear relationship between $R_{NP}$ and the productivity of each experiment (Eq. 9-11). As underlined in the method section, our conclusions are still valid if we assumed a linear relationship up to a value *thresh* if *thresh* replaces 1 in the definition categories given in Table 1. The value *thresh* is nevertheless theoretical because the calculated nutrient limitation ($R_N$, $R_P$, $R_{NP}$) has no physical meaning and is disconnected from physical measure of e.g. soil P content (Olsen P, etc.). The fact that the transition between linear~~

~~and plateau regimes occurs for the same $R_{NP}$ (1 or *thresh*) globally should be an acceptable assumption as we took into account the spatial variation in soil properties to compute the soil nutrient supply.~~

Two formalisms are usually used to characterize multiple element limitation: in Liebig's law of the minimum, plants are generally limited by one nutrient at a time, while plants are generally co-limited in a multiple limitation hypothesis. Our study reveals that the choice of the formalism has only a marginal effect on the estimate of current global NP limitation ($R_{NP}$) for the cereals considered. This result is explained by the fraction of grid-cells in our approach that is within the area of the $R_N$ vs $R_P$ space that maximizes the difference between the two formalisms. The formalism choice has a bigger effect on the increases in $R_N$ and $R_P$ required at the same time to alleviate the NP limitation. Because of very different theoretical founding principles behind each formalism, the use of one or other formalism leads to very different estimates of occurrence of co-limitation in cropland. As mentioned earlier, Liebig's law of minimum or multiple limitation hypothesis could be considered as macro-properties that reflect the same processes of plant adjustments but, depending on the context, plant adjustments lead to one or the other formalism (Ågren et al., 2012). It was also stipulated (Farrior et al., 2013) that plants can be limited by only one resource at a time, but that the integration of the different limitations in time makes the plants limited by several resources at the scale of the growing season. In our point of view, the use of mechanistic models, through the representation of dynamic allocation (e.g. through a cost-based approach (Franklin et al., 2012)) and floating C:nutrient ratios (Zaehle and Dalmonech, 2011) would allow the explicit consideration of some plant adjustments, preventing the need to choose between formalisms.

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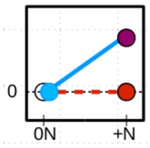
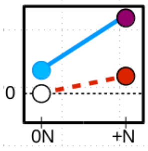
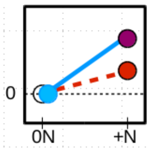
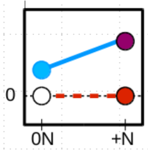
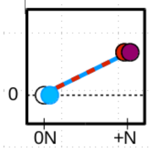
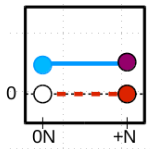
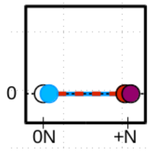
## Tables

**Table 1** (two pages). Nutrient limitation categories defined in Harpole et al. (2011) and occurrence for each crop in our modelling approach with MH formalism. $\Delta \text{pro}_{+X}$ is the change in productivity following the application of +X (with X=N, P or NP) in fertilization experiments. In the 1st column, the y-axis defines ecosystem productivity and the dots correspond to the different experiments (white: control, blue: after addition of P, red: after addition of N, magenta: after addition of NP). Each category is 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 ($\Delta \text{pro}_{+N}+\Delta \text{pro}_{+P}$) (3th column). Synergistic co-limitation means that $\Delta pro_{+NP} > \Delta pro_{+N} + \Delta pro_{+P}$ . The different categories (columns 1-3) are derived from Harpole et al. (2011) while category B is restricted here to the “super-additive case” (sub-additive or additive are neglected because they cannot happen in MH or LM, see Text S1).

| Long name of the category, letter used hereafter and figure | Co-limitation | Definition of the category | Analytic conditions required to be in that category for MH * | Occurrence for MH: % of global crop area (% of grid-cells) | Analytic conditions required to be in that category for LM | Occurrence for LM: % of global maize area (% of grid-cells) |
|---|---|---|---|---|---|---|
| Simultaneous~~Silmutaneous~~ co-limitation<br>A<br> | True and synergistic | $\Delta\, pro_{+N}=0$<br>$\Delta\, pro_{+P}=0$<br>$\Delta\, pro_{+NP}>\Delta\, pro_{+N}+\Delta\, pro_{+P}$ | $R_N=0$<br>and<br>$R_P=0$ | 0.0±0.0<br>(0.0±0.0) | $R_P=R_N\neq1$ | 0.0±0.0<br>(0.0±0.0) |
| Independent co-limitation; super-additive<br>B<br> | True and synergistic | $\Delta\, pro_{+N}\neq0$<br>$\Delta\, pro_{+P}\neq0$<br>$\Delta\, pro_{+NP}>\Delta\, pro_{+N}+\Delta\, pro_{+P}$ | $R_N$ in (~~]~~0,1)~~[~~<br>and<br>$R_P$ in ~~]~~(0,1)~~[~~ | 41.7±0.6<br>(35.9±0.3) | Cannot occur | 0.0±0.0<br>(0.0±0.0) |
| Serial limitation N<br>C<br> | Synergistic | $\Delta\, pro_{+N}\neq0$<br>$\Delta\, pro_{+P}=0$<br>$\Delta\, pro_{+NP}>\Delta\, pro_{+N}+\Delta\, pro_{+P}$ | $R_N=0$<br>and<br>$R_P$ in ~~]~~(0,1)~~[~~ | 6.7±0.3<br>(8.9±0.2) | $R_N<R_P$<br>and<br>$R_P(E_2)<R_N(E_2)$<br>** | 6.3±0.4<br>(9.4±0.2) |
| Serial limitation P<br>D<br> | Synergistic | $\Delta\, pro_{+N}=0$<br>$\Delta\, pro_{+P}\neq0$<br>$\Delta\, pro_{+NP}>\Delta\, pro_{+N}+\Delta\, pro_{+P}$ | $R_N$ in ~~]~~(0,1)~~[~~<br>and<br>$R_P=0$ | 0.0±0.0<br>(0.0±0.0) | $R_P<R_N$ and $R_N(E_3)<R_P(E_3)$ | 9.5±0.5<br>(10.6±0.3) |
| Single-resource response N<br>E<br> | No | $\Delta\, pro_{+N}\neq0$<br>$\Delta\, pro_{+P}=0$<br>$\Delta\, pro_{+NP}=\Delta\, pro_{+N}+\Delta\, pro_{+P}$ | $R_N$ in [0,1)~~[~~<br>and<br>$R_P=1$ | 37.1±0.6<br>(46.0±0.4) | $R_N<R_P$<br>and<br>$R_P(E_2)\geq R_N(E_2)$<br>*** | 48.4±0.8<br>(60.7±0.5) |
| Single-resource response P<br>F | No | $\Delta\, pro_{+N}=0$<br>$\Delta\, pro_{+P}\neq0$<br>$\Delta\, pro_{+NP}=\Delta\, pro_{+N}+\Delta\, pro_{+P}$ | $R_N=1$<br>and<br>$R_P$ in [0,1)~~[~~ | 2.7±0.3<br>(1.3±0.1) | $R_P<R_N$<br>and<br>$R_N(E_3)\geq R_P(E_3)$ | 24.0±0.9<br>(11.5±0.5) |

| | | | | | | |
|---|---|---|---|---|---|---|
|  | | | | | | |
| No response<br>G<br> | No | $\Delta\, pro_{+N}=0$<br>$\Delta\, pro_{+P}=0$<br>$\Delta\, pro_{+NP}=\Delta\, pro_{+N}+\Delta\, pro_{+P}$ | $R_N=1$<br>and<br>$R_P=1$ | 11.8±0.3<br>(7.8±0.1) | $R_N=1$<br>and<br>$R_P=1$ | 11.8±0.3<br>(7.8±0.1) |

** a parenthesis instead of a square bracket~~reversed bracket~~ used in an interval means that the corresponding endpoint is excluded from the interval; e.g. R in [0,1)~~[~~ means 0≤R<1.*

*** corresponds to “$E_1$: N-limited and $E_2$: P-limited”*

**** corresponds to “$E_1$: N-limited and ($E_2$: N-limited or NP-limited or not limited at all)”*

**Table 2.** Global values (± one standard-deviation) of the supply/demand ratio (R) for N, P or NP.

| Nutrient(s) | | N | P | NP (formalism LM) | NP (formalism MH) |
|---|---|---|---|---|---|
| Ratio (R) | Maize | 0.42 ± 0.00 | 0.62 ± 0.01 | 0.32 ± 0.00 | 0.29 ± 0.00 |
| | Wheat | 0.49 ± 0.00 | 0.73 ± 0.00 | 0.43 ± 0.00 | 0.40 ± 0.00 |
| | Rice | 0.70± 0.00 | 0.79 ± 0.01 | 0.60 ± 0.01 | 0.59 ± 0.01 |

## Figures

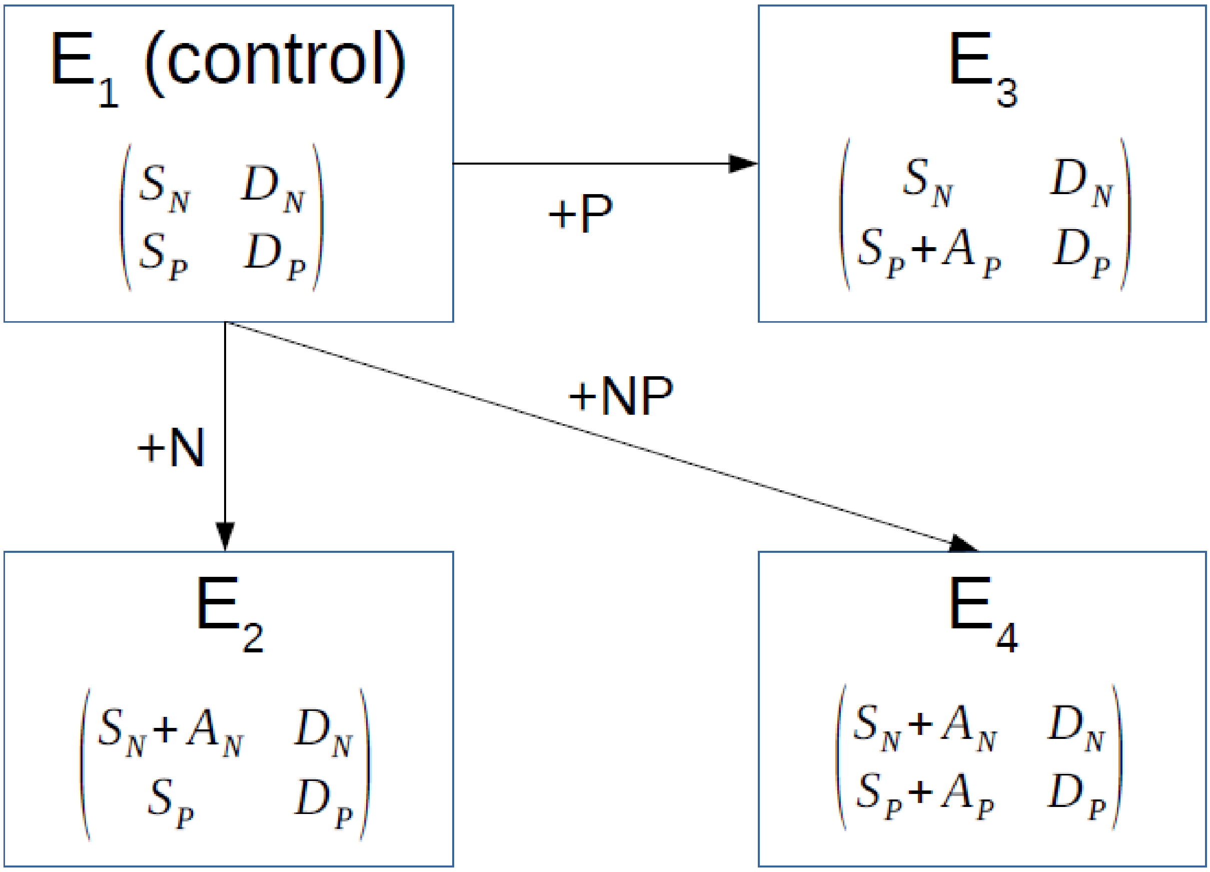


**Figure 1.** Fertilization experiments. The different experiments ($E_1$-$E_4$) vary as function of their supply of N ($S_N$ or $S_N+A_N$) and P ($S_P$ or $S_P+A_P$).

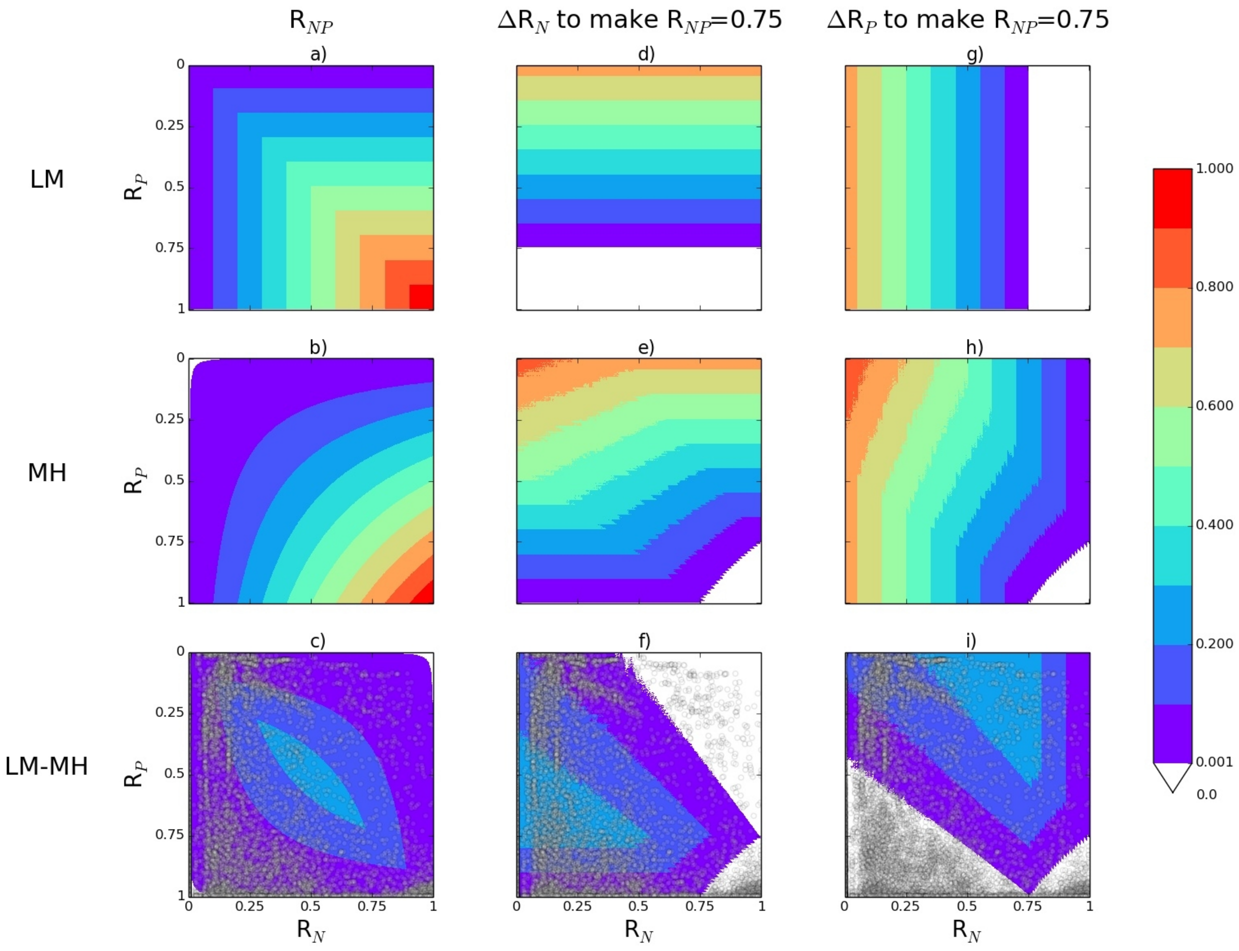


**Figure 2**. R$_{NP}$, ΔR$_N$ and ΔR$_P$ for any values of R$_P$ (x-axis) and R$_N$ (y-axis). Each variable is provided for the two formalisms (LM and MH) as well as for the difference LM-MH. Grey transparent dots in panels c,f,i correspond to all grid-cells considered for maize in our modelling approach.

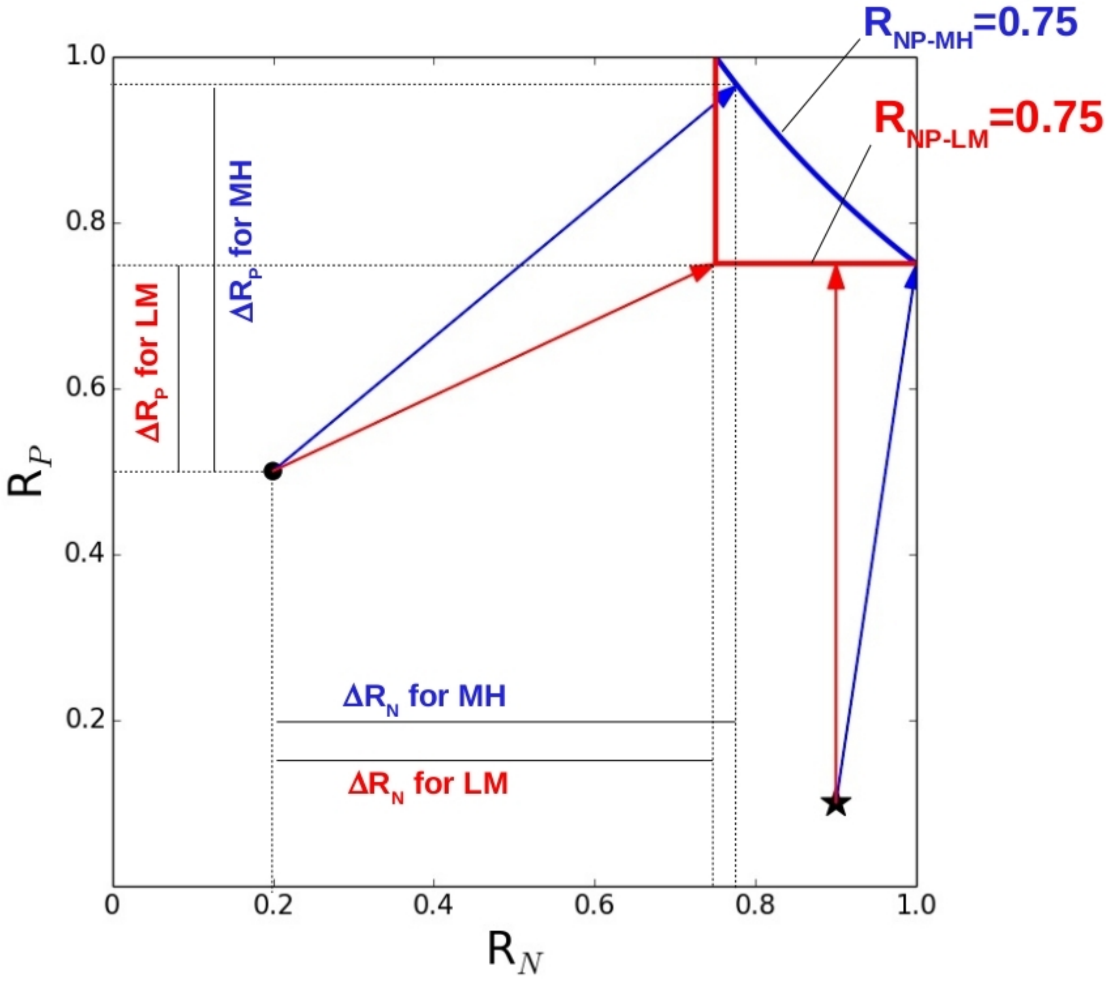


**Figure 3.** Schematic representation of the computation of ($\Delta R_N$, $\Delta R_P$). A given grid-cell is defined by its ($R_N$, $R_P$) in the plan characterized by the base $(\vec{R}_N, \vec{R}_P)$ . For a given grid-cell and a given formalism, we called $\vec{u}$ the shortest vector linking ($R_N$, $R_P$) and the curve (or segments) defining $R_{NP}$=0.75. We called x and y the compounds of $\vec{u}$ in the basis $(\vec{R}_N, \vec{R}_P)$ , i.e. $\vec{u}=\begin{pmatrix} x \\ y \end{pmatrix}$ . We defined $\Delta R_N = max(0, x)$ and $\Delta R_P = max(0, y)$ . In the above figure, two grid-cells are provided as an example: ($R_N$=0.2; $R_P$=0.5) for the black dot, and ($R_N$=0.9; $R_P$=0.1) for the black star. The formalism of interaction defines the ($R_N$,$R_P$) couples that make $R_{NP}$=0.75: the blue curve defines $R_{NP}$=0.75 for MH while the two orthogonal red segments define $R_{NP}$=0.75 for LM. $\vec{u}$ is provided for each grid-cell and each formalism (blue arrow for MH ; red arrow for LM). We explicitly plotted the $\Delta R_N$ and $\Delta R_P$ for the black dot and the two formalisms (solid black lines). Note that for the grid-cell symbolized by the black star, $\Delta R_N = 0$ for LM.

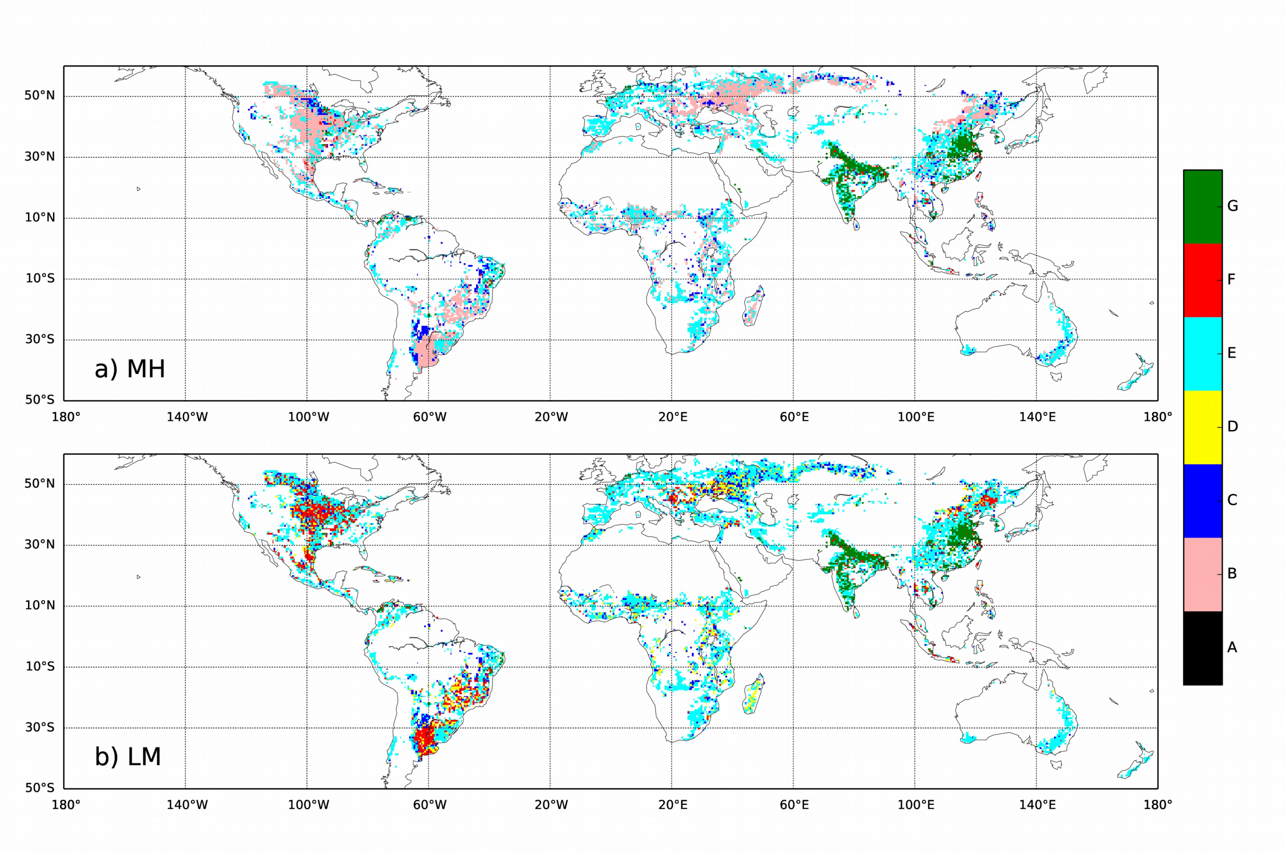


**Figure 4.** Spatial distribution of the categories defined in Table 1 and in Harpole et al. (2011) for MH (a) and LM (b) for maize. Category A corresponds to simultaneous co-limitation, category B to independent co-limitation (super-additive), categories C and D to serial limitation (N and P, respectively), categories E and F to single-resource response (N and P, respectively) and category G to no response. For LM, whether one grid-cell belongs either to category C (dark blue) or to category E (cyan) depends on the value of $A_N$. The same reasoning applies for categories D (yellow) and F (red) with $A_P$.

## Data and Code availability:

Files corresponding to supply and demand for N and P (variables called $S_N$, $S_P$, $D_N$, $D_P$ in the manuscript) are made available (Ringeval et al. ,2019) on the following link.
Computer scripts written by the authors to generate and manipulate files of supply and demand for N and P are available upon request to the corresponding author.

## Acknowledgements:

This research was supported by the "Institut National de la Recherche Agronomique" (INRA) and the Environnement et Agronomie (EA) division. We thank Lex Bouwman for providing variables at the basis of N supply computation. We thank Thomas A.M. Pugh, Frédéric Barraquand, Alain Mollier, Pascal Denoroy and Noémie Schaller for helpful discussion. Finally, we would like to thank Aldyth Nys for her assistance with the linguistic aspects of this paper. Modelling and analysis were performed in using Python (Python Software Foundation. Python Language Reference, version 2.7. Available at http://www.python.org).

## Author contribution:

B.R conceived the project; B.R. and M.K. performed the computations; B.R, S.P and L.A analysed the results; N.D.M, C.M, T.A.M.P and X.W. provided datasets and their expertise on these datasets; B.R, M.K, L.A, P.C, D.G, N.D.M, C.M, T.N, T.A.M.P, N.V, S.P wrote the article.

## Competing interests:

The authors declare no conflict of interest.

## Supporting Information

### Supporting Text

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

**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

**Text S3.** Computation of the nutrient demand ($D_N$ and $D_P$)

**Text S4.** Computation of the potential P uptake

**Text S5.** Global values and uncertainty

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

**Text S7.** Relationship between $R_{NP}$ and yield

**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

### Supporting Tables

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

**Table S2.** Parameters used to estimate the N and P demands ($D_N$ and $D_P$, respectively)

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

**Table S4.** Values possible for $R_N$ and $R_P$ and the implications with MH formalism

**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

### Supporting Figures

**Figure S1.** Spatial distribution of $\Delta R_N$ and $\Delta R_P$

**Figure S2**. The effect of using the real yield (instead of potential yield) on the computed nutrient limitation

**Figure S3.** Grid-cell distribution in percentiles of different variables

**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

**Figure S7.** Scatterplots of the ratio $Y_{real}/Y_{pot}$ provided by Mueller et al. (2012) vs. the simulated $R_{NP}$ at the country scale for maize