



*Supplement of*

## **Improved representation of phosphorus exchange on soil mineral surfaces reduces estimates of phosphorus limitation in temperate forest ecosystems**

**Lin Yu et al.**

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## S1 Additional figures and tables

**Table S1: The normalized root mean square ratio ( $K_{nrmsr}$ ) between simulated and observed soil properties along soil profiles of four models at five study sites (Fig. 2).**

Soil properties	Site	4pool	siLang	dbLang	Control
<b>SOC</b>	BBR	0.32	0.32	0.32	0.32
	MTF	0.59	0.58	0.59	0.59
	VES	0.56	0.54	0.56	0.56
	COM	0.47	0.49	0.47	0.47
	LUE	0.59	0.58	0.63	0.61
<b>SOM CN ratio</b>	BBR	0.9	0.90	0.90	0.90
	MTF	0.95	0.95	0.95	0.95
	VES	0.93	0.93	0.93	0.93
	COM	0.85	0.84	0.85	0.85
	LUE	0.84	0.84	0.85	0.86
<b>SOM CP ratio</b>	BBR	0.54	0.54	0.54	0.54
	MTF	0.58	0.58	0.58	0.58
	VES	0.74	0.74	0.74	0.74
	COM	0.76	0.73	0.76	0.76
	LUE	0.61	0.61	0.63	0.67
<b>Bulk density</b>	BBR	0.69	0.69	0.69	0.69
	MTF	0.76	0.76	0.76	0.76
	VES	0.79	0.79	0.79	0.79
	COM	0.66	0.61	0.66	0.66
	LUE	0.81	0.81	0.82	0.84
<b>SOP</b>	BBR	0.33	0.33	0.33	0.33
	MTF	0.39	0.40	0.39	0.40
	VES	0.46	0.46	0.46	0.46
	COM	0.49	0.52	0.49	0.49
	LUE	0.49	0.50	0.45	0.42
<b>SIP</b>	BBR	0.77	0.78	0.77	0.79
	MTF	0.85	0.78	0.85	0.81
	VES	0.78	0.72	0.81	0.78
	COM	0.76	0.68	0.80	0.82
	LUE	0.81	0.81	0.81	0.77

<b>Labile Pi</b>	BBR	0.26	0.48	0.57	0.74
	MTF	0.29	0.47	0.68	0.71
	VES	0.3	0.51	0.60	0.56
	COM	0.18	0.37	0.36	0.76
	LUE	0.28	0.65	0.41	0.68
<b>Sorbed Pi</b>	BBR	NA	0.69	0.56	0.82
	MTF	NA	0.62	0.71	0.79
	VES	NA	0.55	0.78	0.72
	COM	NA	0.39	0.63	0.82
	LUE	NA	0.34	0.40	0.71
<b>Lab-to-Exchangeable P</b>	BBR	NA	0.47	0.62	0.80
	MTF	NA	0.46	0.70	0.67
	VES	NA	0.54	0.71	0.47
	COM	NA	0.35	0.37	0.72
	LUE	NA	0.55	0.62	0.61

**Table S2 Parameters for sensitivity analysis**

<b>Symbol</b>	<b>Description</b>	<b>Value</b>	<b>Unit</b>	<b>Reference</b>
$v_{max,PO_4}$	Maximum plant P uptake rate	0.044	$\mu\text{mol P/mol C/s}$	Thum et al. 2019
$x_{SOM_{fast}^{N:P}}$	N:P ratio of fast SOM pool	31	mol/mol	Thum et al. 2019
$x_{SOM_{slow}^{N:P}}$	N:P ratio of slow SOM pool	11.07	mol/mol	Thum et al. 2019
$\mu_P$	Microbial phosphorus-use efficiency	0.8	mol/mol	Thum et al. 2019
$\tau_{fast}$	Turnover time of fast SOM pool	2	years	Thum et al. 2019
$\tau_{slow}$	Turnover time of slow SOM pool	100	years	Thum et al. 2019
$\tau_{biomin}$	Phosphorus biomineralization turnover time	5	years	Thum et al. 2019
$k_{weath}$	Weathering rate constant of mineral soil	8.16208	$10^{-8} \mu\text{mol P/m}^3/\text{s}$	Yang et al. 2014
$k_{ocl}$	Occlusion coefficient of sorbed $PO_4$	3.86	$10^{-13} \text{s}^{-1}$	Yang et al. 2014
$k_{ads}$	$PO_4$ (ab)sorption rate from $P_{lab}$ to $P_{sorb}$	651.852	$\mu\text{mol P/kg soil/s}$	Yang et al. 2014

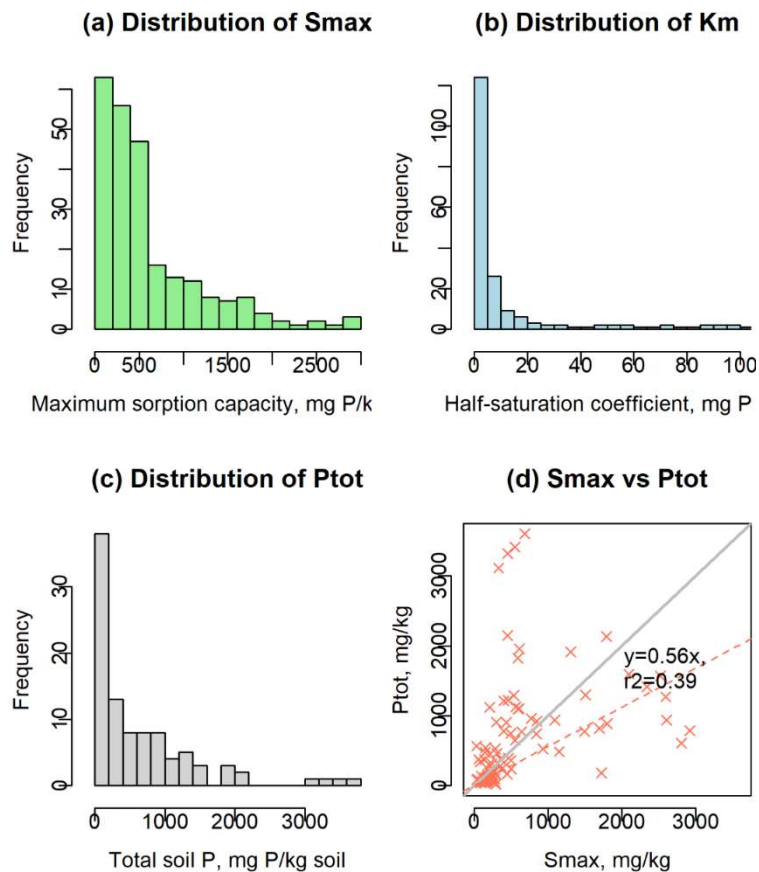
$k_{des}$	PO <sub>4</sub> desorption rate from P <sub>sorb</sub> to P <sub>lab</sub>	733	μmol P/kg soil/s	Yang et al. 2014
$K_{m,PO_4}^{pH}$	Correction coefficient of pH on Langmuir Km	0.4	-	this study
$Q_{max,PO_4}^{Al/Fe}$	Phosphate sorption capacity of Al/Fe oxides	9.134	mmol P/kg clay	this study
$Q_{max,PO_4}^{fs}$	Phosphate sorption capacity of fine soil	9.134	mmol P/kg fine soil	this study
$Q_{max,PO_4}^{sand}$	Phosphate sorption capacity of sand	4.567	mmol P/kg sand	this study
$Q_{max,PO_4}^{OM}$	Phosphate sorption capacity of organic matter	4.567	mmol P/kg OM	this study

**Table S3. The weighted mean values of partial correlation coefficient against all the output variables (Fig. 5) for the 16 selected parameters (Table S2) in the LHS sensitivity runs. Overall importance of parameters is measured by first calculating the RPCC for each output variable and then calculating the mean of the absolute RPCC values across selected outputs in Fig.5, weighted by the uncertainty contribution of these model outputs**

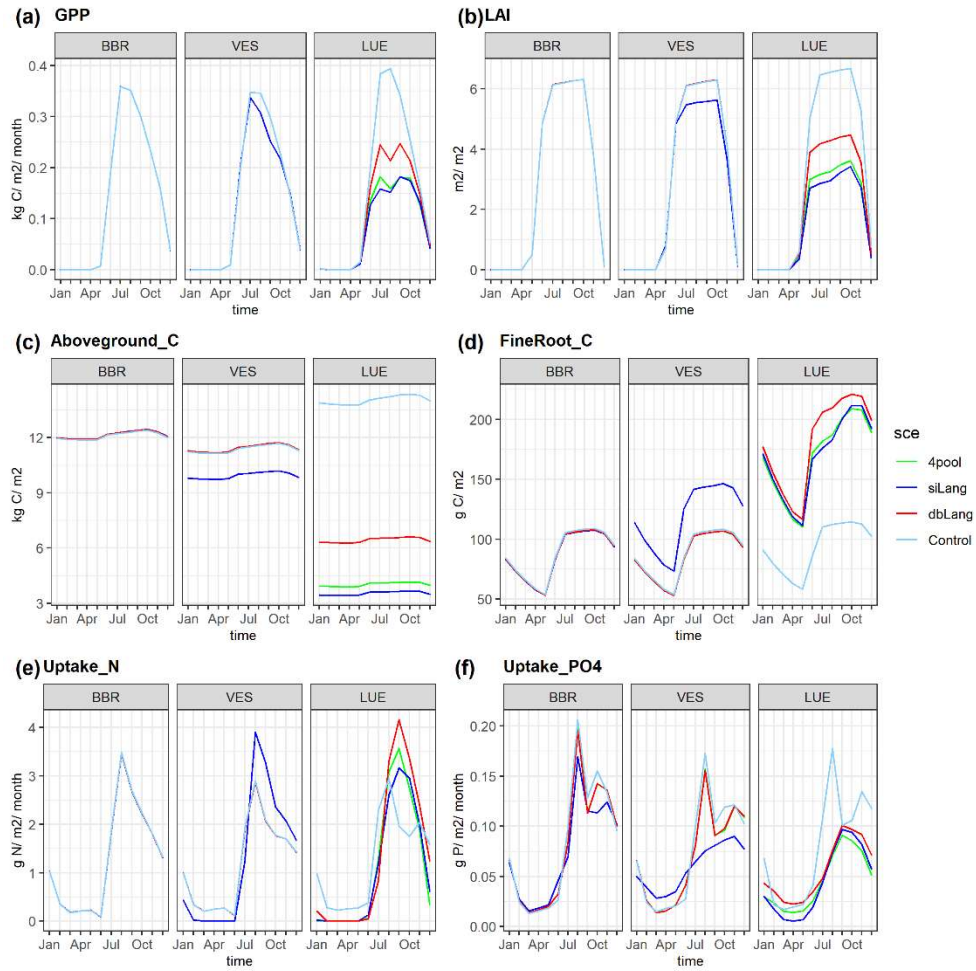
Models, sites & Parameters	dbLang		siLang	
	COM	MTF	COM	MTF
$\tau_{slow}$	0.89	0.97	0.82	0.78
$\tau_{fast}$	0.55	0.61	0.34	0.35
$x_{SOM_{slow}}^{N:P}$	0.63	0.34	0.62	0.32
$k_{des}$	0.03	0.03	0.61	0.54
$k_{ads}$	0.02	0.02	0.90	0.86
$v_{max,PO_4}$	0.31	0.17	0.50	0.39
$x_{SOM_{fast}}^{N:P}$	0.28	0.22	0.26	0.26
$Q_{max,PO_4}^{sand}$	0.14	0.16	0.21	0.19
$Q_{max,PO_4}^{fs}$	0.20	0.26	0.56	0.28
$Q_{max,PO_4}^{Al/Fe}$	0.32	0.32	0.05	0.18
$Q_{max,PO_4}^{OM}$	0.14	0.12	0.10	0.18

$K_{m,PO_4}^{PH}$	0.24	0.23	0.21	0.14
$k_{weath}$	0.25	0.13	0.31	0.10
$\tau_{biomin}$	0.03	0.02	0.02	0.01
$k_{ocl}$	0.04	0.04	0.03	0.03
$\mu_P$	0.02	0.01	0.01	0.01

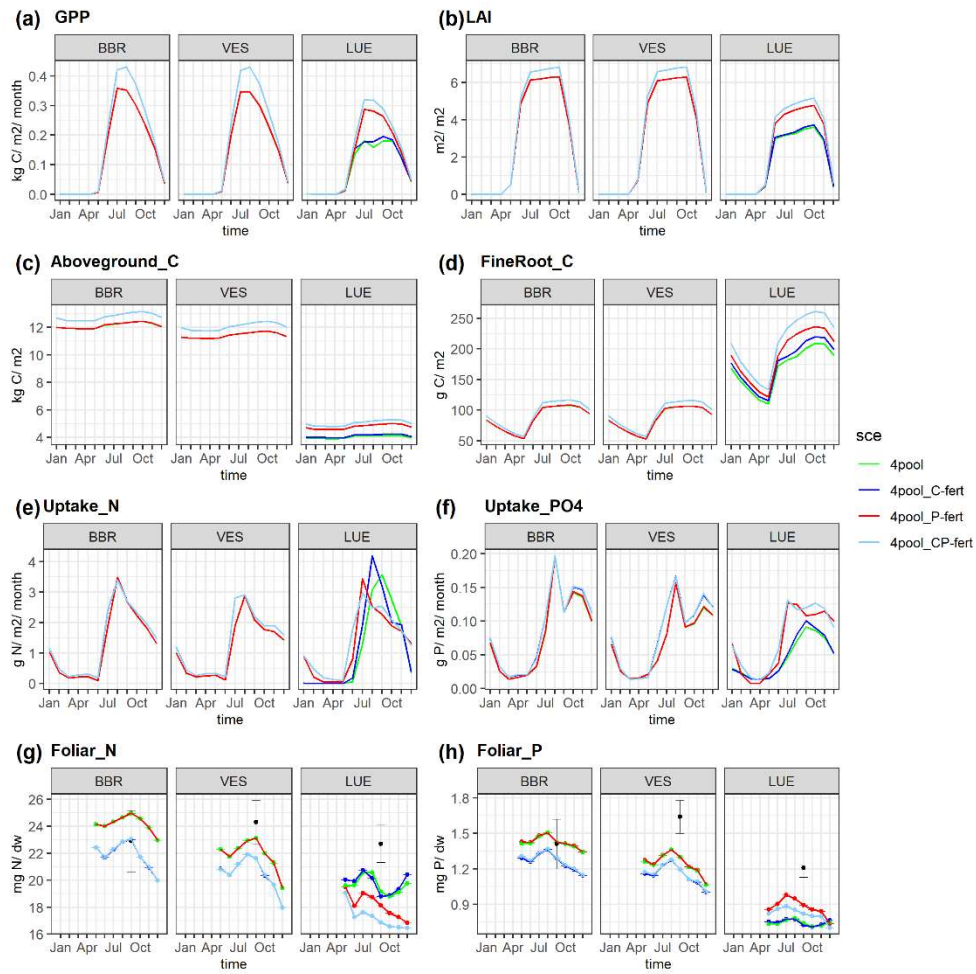
Figure S1: Reported Smax, Km, and Ptot in batch experiment data. The plot is produced based on 258 data inputs from [1-27]. The processing details are given in Section “A1 Processing of the reviewed data”.



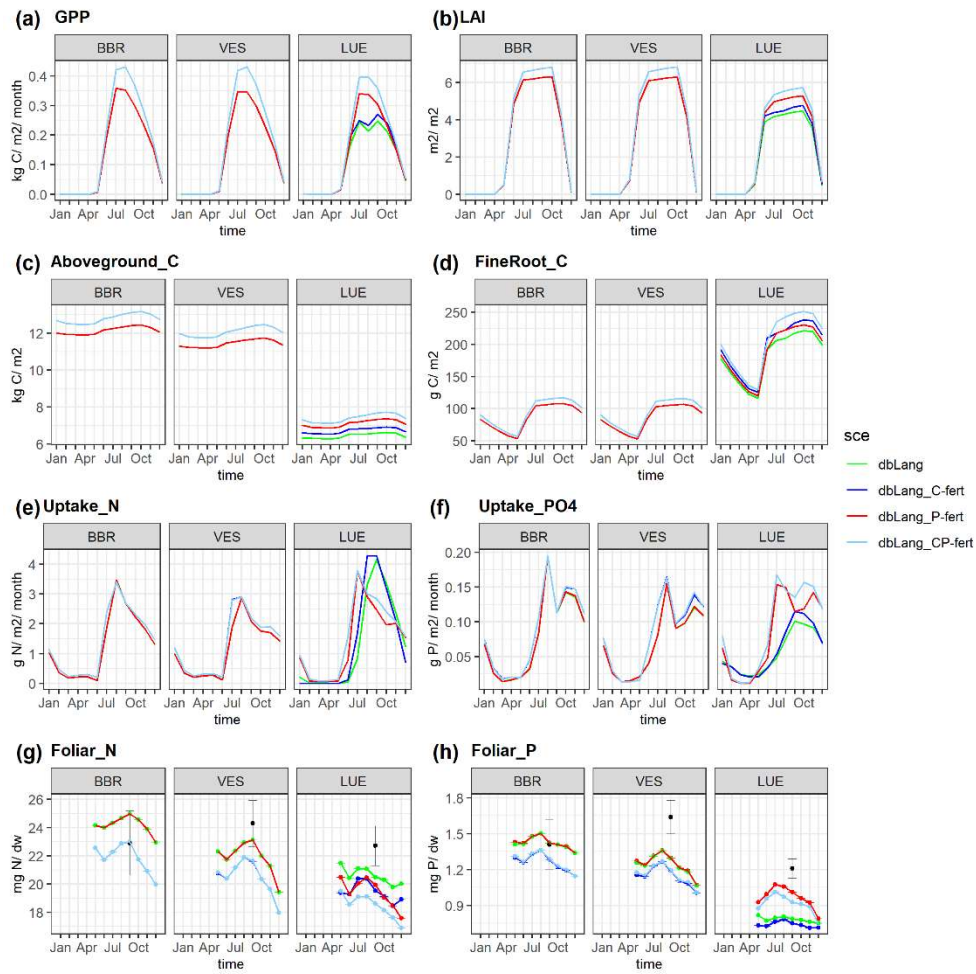
**Figure S2: Simulated GPP, LAI, aboveground C, fine root C, plant uptakes of N and P. The simulated values are the yearly average of the period 2006–2015. The lime line represents the Langmuir model with only four inorganic P pools (4pool), the blue line represents the single-surface Langmuir model (siLang), the red line represents the double-surface Langmuir model (dbLang), and the light sky-blue line represents the Control model.**



**Figure S3: 4pool model responses to C and P fertilisation. The simulated values are the yearly average of the period 2006–2015. The lime line represents the 4pool model, the blue line represents the CO<sub>2</sub> fertilization, the red line represents the P fertilization, and the light sky-blue line represents the C&P fertilization.**

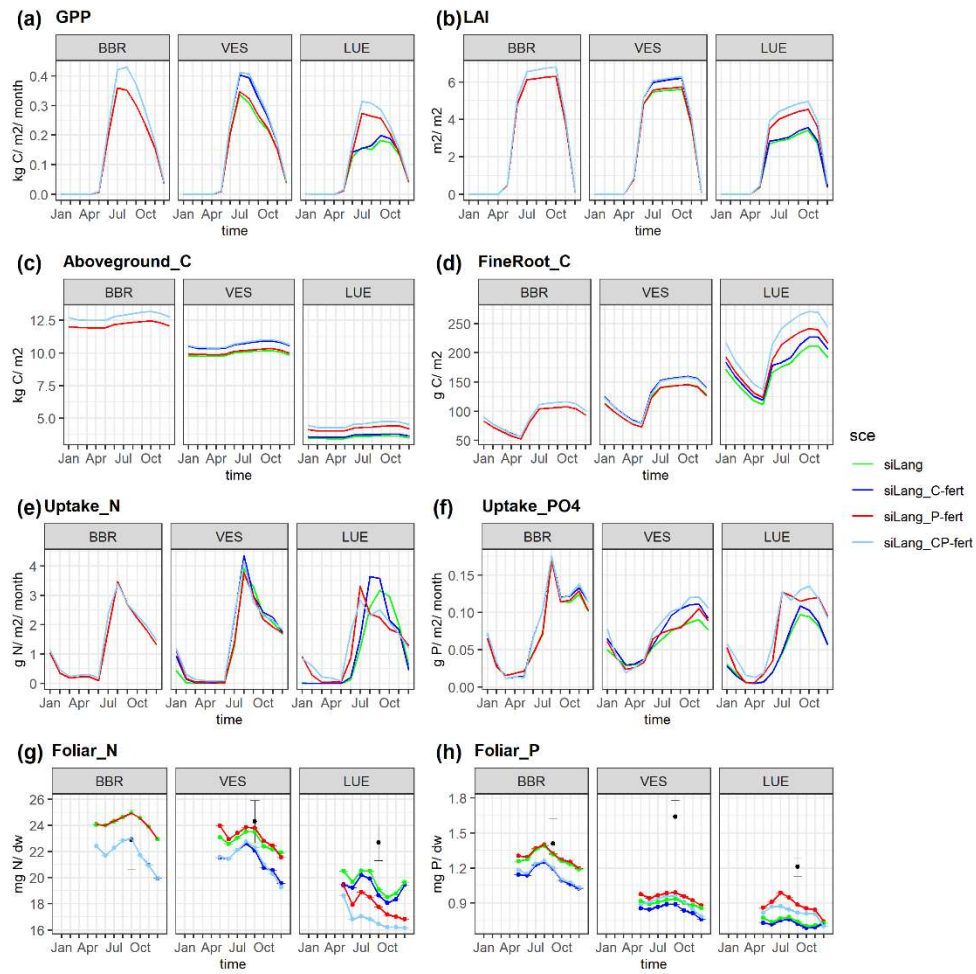


**Figure S4: dbLang model responses to C and P fertilization.** The simulated values are the yearly average of the period 2006–2015. The lime line represents the 4pool model, the blue line represents the CO<sub>2</sub> fertilization, the red line represents the P fertilization, and the light sky-blue line represents the C&P fertilization.

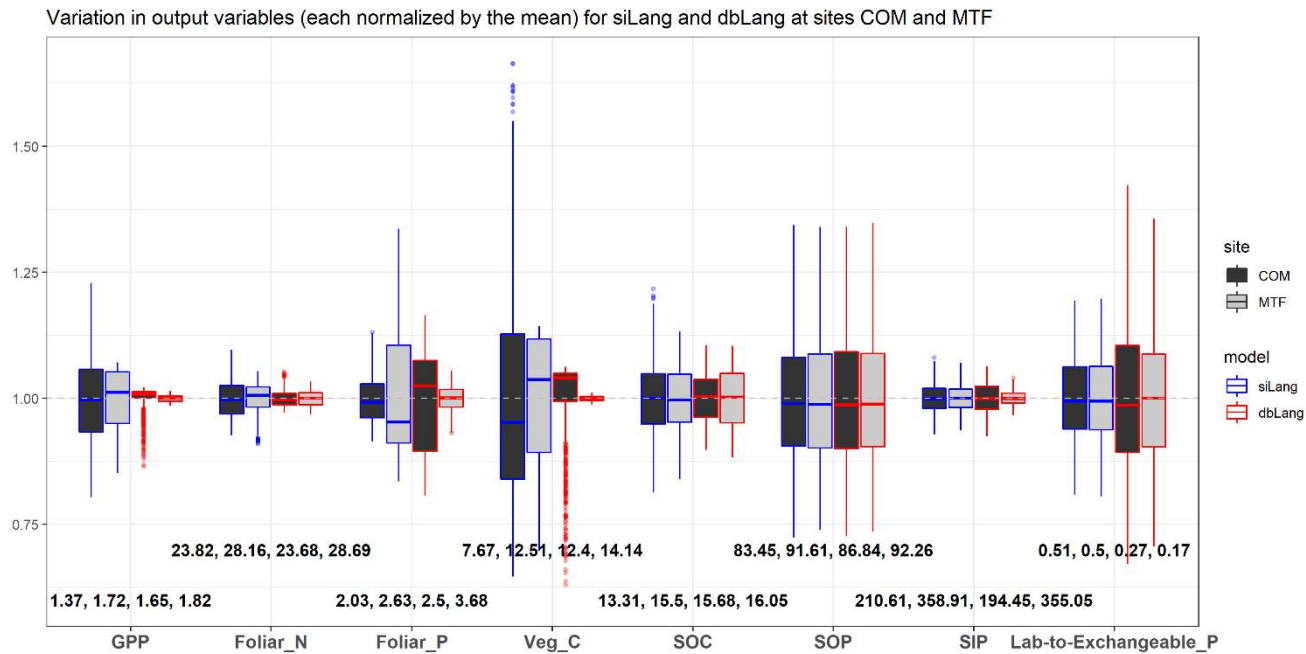




**Figure S5: siLang model responses to C and P fertilization. The simulated values are the yearly average of the period 2006–2015. The lime line represents the 4pool model, the blue line represents the CO<sub>2</sub> fertilization, the red line represents the P fertilization, and the light sky-blue line represents the C&P fertilization.**

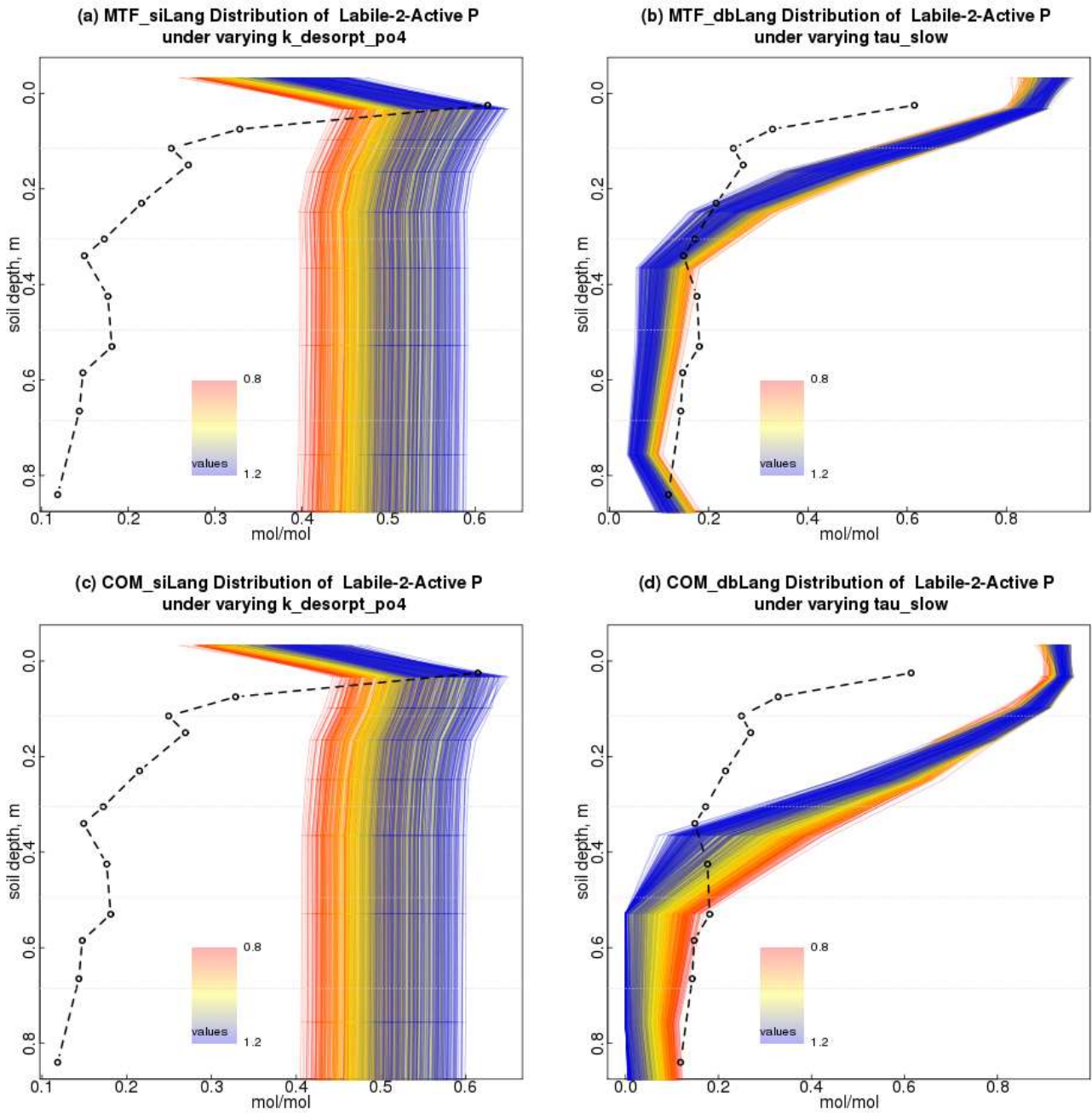


**Figure S6: Normalized output variations in the LHS sensitivity analysis of siLang and dbLang models at COM and MTF sites. The selected output variables include GPP (kg C/m<sup>2</sup>/yr), foliar N content (mg N/g d.w.), foliar P content (mg P/g d.w.), plant C (kg C/m<sup>2</sup>), SOC stock (kg C/m<sup>2</sup>), SOP and SIP stocks (g P/m<sup>2</sup>) and Lab-to-Exchangeable P ratio. All the calculations are performed based on data from the last 10 years of 1000 LHS simulations, and soil variables are based on the topmost 1m of soil. The numbers below bars are the mean values of the 1000 LHS runs, and bars are the distribution of all 1000 LHS runs normalized to the mean value.**



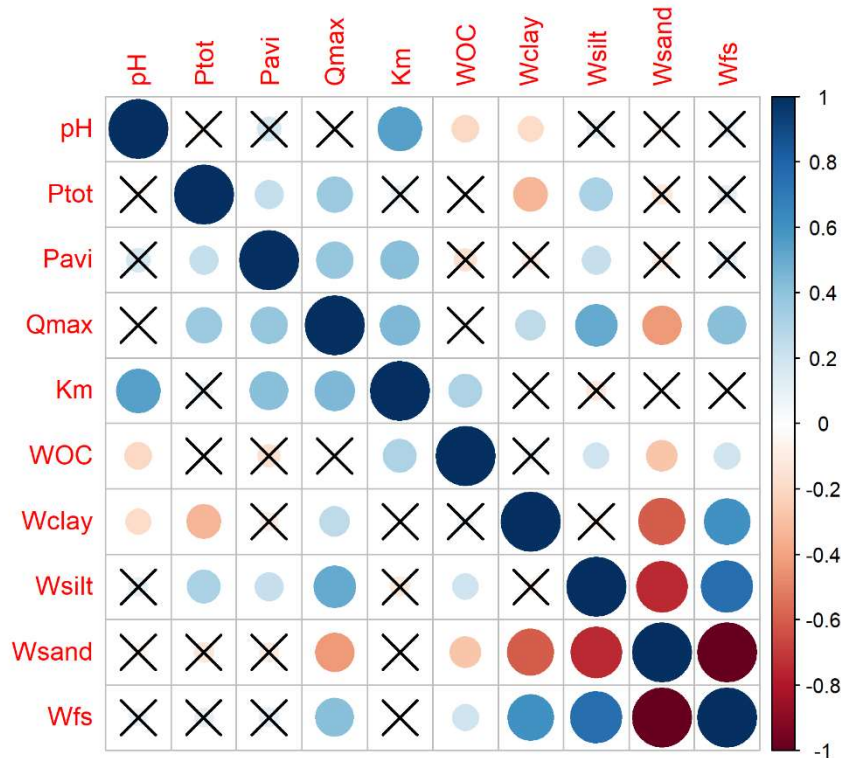
The simulated annual GPP was lower in **siLang** ( $1.37 \pm 0.12$  and  $1.72 \pm 0.12$  kg C/m<sup>2</sup>/yr for COM and MTF) than in **dbLang** ( $1.65 \pm 0.04$  and  $1.82 \pm 0.01$  kg C/m<sup>2</sup>/yr for COM and MTF), and **siLang** responded more strongly to changes of P cycling processes parameterization than **dbLang** (Figs. 5 and S6), reflecting a stronger P-control on plant productivity in the **siLang** model. This stronger P limitation also led to lower plant and soil C in **siLang** (plant C:  $7.67 \pm 1.58$  and  $12.51 \pm 1.55$  kg C/m<sup>2</sup> for COM and MTF; soil C:  $13.31 \pm 0.94$  and  $15.50 \pm 0.96$  kg C/m<sup>2</sup> for COM and MTF) than in **dbLang** (plant C:  $12.40 \pm 1.06$  and  $14.14 \pm 0.07$  kg C/m<sup>2</sup> for COM and MTF; soil C:  $15.78 \pm 0.76$  and  $16.04 \pm 0.95$  kg C/m<sup>2</sup> for COM and MTF).

Figure S7: Responses of Labile-to-Active P fraction to changes in parameterization in siLang and dbLang models at COM and MTF sites



## 5 S2 Processing of the reviewed data

As most P species present in soil solution are negatively charged, the major P sorbents are those constituents that bear positive charges. These include hydroxyl (Fe and Al oxides), carboxyl (organic matter) or silanol (clays) groups [28]. Of the studies that we reviewed, the P sorption capacity of soils has been variously related to soil pH [18, 23, 29], mineralogy or clay content [30-32], organic complexes of Fe and Al [33], soil organic matter such as DOC, organic acids, DOP etc. [29, 34-37], calcium carbonate [8, 26], soil pedogenesis [32, 35], extractable Fe and Al oxides, hydroxides, and oxyhydroxides, and other soil properties. The correlation between soil properties and Langmuir parameters are demonstrated in the figure below,



**Figure S8: Correlation between reported soil properties and Langmuir parameters from reviewed literature. More details given below.**

- 15 To derive Figs. S1 and S8, the raw data from reference [1-27] were processed in the following steps:
1. Conversion of Langmuir coefficient from  $K_L$  to  $K_m$ . The Langmuir isotherm in the batch sorption experiments were reporting the Langmuir coefficient either as  $K_L$  in the unit of L/mg P, or as  $K_m$  in the unit of mg P/L. We unified the Langmuir coefficient to  $K_m$  by inverting  $1/K_L$ . To further generate the data for Table 1 and Fig. 2, we assumed that the soil water content for all experiments data are 330 L/m<sup>3</sup> soil and converted the unit from mg P/L to g P/m<sup>2</sup>.
  2. Selection of reported soil properties. The reviewed papers don't follow the same protocols in reporting the soil properties, apart from the two Langmuir isotherm parameters ( $S_{max}$  and  $K_m$ ), many studies also reported organic matter contents (OM, in percent), soil texture (clay, silt and sand), pH (measured in water or CaCl<sub>2</sub>/KCl), total soil P (Ptot,
- 20

mg P/ kg soil), available P (P<sub>av</sub>, mg P/kg soil). We selected the abovementioned variables (pH as water measured values) to understand their relationships with Langmuir parameters.

25 Conversion of OM and soil texture to weights. In order to derive a weight-based relationship between Langmuir parameters, the reported OM contents and soil texture were converted to OM and clay, silt, sand weights, assuming an OM density of 250 kg/m<sup>3</sup> and mineral soil bulk density of 1000 kg/m<sup>3</sup>. The correlation between the soil properties and Langmuir parameters are plotted in Fig. S7, where W<sub>OC</sub>, W<sub>clay</sub>, W<sub>silt</sub>, W<sub>sand</sub> are the OM, clay, silt, and sand weights [kg/m<sup>3</sup>], respectively, and W<sub>fs</sub> stands for weight of fine soil (clay plus silt).

### 30 S3 Double-surface Langmuir isotherm and parametrization

As both single- and double-surface Langmuir isotherm could be fitted against the same experiments data, the apparent maximum sorption capacity ( $S_{max}$ ) in Eq. 1 is the sum of sorption maxima of two sorption sites ( $S_{max,1}$  and  $S_{max,2}$ , Eqs. 3.1 and 3.2) and the apparent Langmuir coefficient ( $K_m$ ) in Eq. 1 could be derived mathematically from the Langmuir coefficients of two sorption sites ( $K_{m,1}$  and  $K_{m,2}$ , Eqs. 3.4 and 3.5).

35 Following the concept of double-surface Langmuir isotherm, we assume that the apparent maximum sorption capacity and Langmuir coefficient in all the batch sorption experiment are in fact a combined value of two (or more) sorption surfaces.

$$S = S_{max} \frac{P_{sol}}{K_m + P_{sol}} = S_{max,1} \frac{P_{sol}}{K_{m,1} + P_{sol}} + S_{max,2} \frac{P_{sol}}{K_{m,2} + P_{sol}}, \quad (S1)$$

Where the apparent maximum sorption capacity is the sum of sorption maxima of individual sorption surface,

$$S_{max} = S_{max,1} + S_{max,2} \quad (S2)$$

40 The apparent Langmuir coefficient is calculated in Eq.3d which is derived using the differential form of double-surface Langmuir isotherm,

$$\frac{dS}{dt} = \frac{S_{max}K_m}{(K_m + P_{sol})^2} \frac{dP_{sol}}{dt} = \frac{S_{max,1}K_{m,1}}{(K_{m,1} + P_{sol})^2} \frac{dP_{sol}}{dt} + \frac{S_{max,2}K_{m,2}}{(K_{m,2} + P_{sol})^2} \frac{dP_{sol}}{dt}, \text{ thus} \quad (S3)$$

$$K_m = \frac{\frac{S_{max}}{hl} - 2P_{sol} \pm \sqrt{\frac{S_{max}^2}{hl^2} - 4\frac{S_{max}P_{sol}}{hlp}}}{2}, \text{ where} \quad (S3.1)$$

$$hlp = \frac{S_{max,1}K_{m,1}}{(K_{m,1} + P_{lab})^2} + \frac{S_{max,2}K_{m,2}}{(K_{m,2} + P_{lab})^2} \quad (S3.2)$$

45 To estimate the sorption maximum and Langmuir coefficient of each sorption surface, we derived pedo-functions based on the reported soil properties from the batch sorption experiment data. As shown in Fig.S2, the maximum sorption capacity is positively correlated with clay and silt content, negatively correlated with sand content and does not show correlation with OM content; the Langmuir coefficient is strongly correlated with soil pH and OM content and not correlated with soil texture. Given the high uncertainty in the original experiment data, and the lack of extractable Al and Fe content that represent  
50 crystalline and non-crystalline Fe and Al oxides and organic complexes of Fe and Al, which are commonly considered the

main sorbents of P, we tested different combination of soil sorbents and finally used a four sorbents pedo-functions for  $S_{max}$  and  $K_m$ ,

$$S_{max} = S_{max,1} + S_{max,2} \quad (\text{Equation S2})$$

$$S_{max,1} = Q_{max,PO_4}^{Al/Fe} \cdot W_{clay} \cdot f_{Al/Fe} + Q_{max,PO_4}^{OM} \cdot W_{OM} \quad (\text{Equation S4.1})$$

$$55 \quad S_{max,2} = Q_{max,PO_4}^{fs} \cdot W_{clay+silt} \cdot f_{Al/Fe} + Q_{max,PO_4}^{sand} \cdot W_{sand} \quad (\text{Equation S4.2})$$

$$f_{Al/Fe} = \frac{(Fe_{ox} + Al_{ox})}{10 \text{ mmol/kg}} \quad (\text{Equation S4.3})$$

$$K_{m,1} = k_{ph} \cdot pH \cdot f(K_m^{OM}, K_m^{Al/Fe}) \quad (\text{Equation S4.4})$$

$$K_{m,2} = k_{ph} \cdot pH \cdot f(K_m^{fs}, K_m^{sand}) \quad (\text{Equation S4.5})$$

Where  $Q_{max,PO_4}^{Al/Fe}$  is the sorption capacity of crystalline and non-crystalline Fe and Al oxides normalized by the clay content;

60  $f_{Al/Fe}$  is the correction coefficient of oxalate-extractable Al and Fe, which is calculated in Eq. 4.3, assuming that most Al- & Fe- (hydro)oxides reside in clay and the rest in silt;  $k_{ph}$  is a unitless correction factor to account for the effect of pH, and  $K_{m,1}$  and  $K_{m,2}$  are calculated using Eqs.4.4 and 4.5.

**Table S4: Parameters for double-surface Langmuir isotherm**

Symbol	Description	Value	Unit
$K_{m,PO_4}^{pH}$	Correction coefficient of pH on Langmuir $K_m$	0.4	-
$Q_{max,PO_4}^{Al/Fe}$	Phosphate sorption capacity of Al/Fe oxides	9.134	mmol P/kg clay
$Q_{max,PO_4}^{fs}$	Phosphate sorption capacity of fine soil	9.134	mmol P/kg fine soil
$Q_{max,PO_4}^{sand}$	Phosphate sorption capacity of sand	4.567	mmol P/kg sand
$Q_{max,PO_4}^{OM}$	Phosphate sorption capacity of organic matter	4.567	mmol P/kg OM
$K_m^{Al/Fe}$	Langmuir coefficient of Al/Fe oxides	315.445	$\mu\text{mol P/L}$
$K_m^{fs}$	Langmuir coefficient of fine soil	2.176	mmol P/L
$K_m^{OM}$	Langmuir coefficient of soil organic matter	20	mmol P/L
$K_m^{sand}$	Langmuir coefficient of sand	20	mmol P/L

#### 65 S4 Deep soil inorganic P initialization

The initial inorganic P pools of soils deeper than 1m was calculated using the following equations:

$$P_{primary}^{frac} = \min\left(\frac{2*\sqrt{z}, 3.4}{3.5}\right) \quad (\text{Equation S5.1})$$

$$P_{exchangeable}^{frac} = \frac{0.8^{2*z}}{3.5} \quad (\text{Equation S5.2})$$

$$P_{ocl}^{frac} = \max\left(1 - P_{primary}^{frac} - P_{exchangeable}^{frac}, \frac{0.1}{3.5}\right) \quad (\text{Equation S5.3})$$

70 Where  $z$  is the soil depth in m, and  $P_{exchangeable}$  includes  $P_{sorb}$  and  $P_{lab}$  with a constant ratio of 9/8 at all depths. The total soil inorganic P contents for all the study sites (g P/m<sup>3</sup>, >1m depth) are given in Table S5.

depth (m)	BBR	MTF	VES	COM	LUE
1.14	124.1	12.9	31.0	18.8	7.6
1.58	133.9	15.0	32.8	18.3	5.8
2.20	100.0	16.7	40.7	23.4	5.9
3.06	126.0	16.9	41.9	25.0	6.8
4.27	122.2	17.5	41.1	21.1	8.3
5.95	145.0	15.1	48.4	21.9	8.4
8.22	90.0	15.6	54.0	15.1	8.5

**Table S5: The total soil inorganic P contents (g P/m<sup>3</sup>, >1m depth) for all study sites at specific depth at QUINCY initialization. The initial values of soil inorganic P contents at top 1 m soil were prescribed from the Hedley fractionation measurements.**

75

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