Appendix A

1 Data and Methodss

1.1 Study area and time

Urban districts were also counted as counties, which can be recognized in county names by the ending “city”. The Songling district and Jiagedaqiqu district were not included in Heilongjiang because their land rights belong to Inner Mongolia. Due to changes in administrative divisions, some counties were split or merged with other counties during the period of interest. As a result, some counties might be annexed, bigger or smaller in some years. The counties that were annexed were: Heilongjiang province: Acheng city (2006-2015), Hulan county (2004-2015), Shuangcheng city (2014, 2015) and Dedu county (1996-2015); Jilin province: Shuangyang county (1995-2015), Linjiang city (1990, 1991), Jiangyuan county (1990-1994, 2006-2015), Songyuan city (1990, 1991), Fuyu county (1992-1994); Liaoning province: Pulandian city (2010-2015). There were some exceptions in the statistics for facilitating the presentation of results: Mohe city (in Heilongjiang province) included Mohe county, Xinlin district and Huzhong district; Baishan city (aliased as hunjiang city in Jilin province) included Baishan city, Linjiang city and Jiangyuan county; Songyuan city (in Jilin province) included Songyuan city and Fuyu county; Panjin city (in Liaoning province) included Panjin city and Panshan county.

1.2 NUE data and covariates

The pre-processing of the initial data set before computing NUE indicators and building models were as follows: (a) filling missing data and covariates in absent years by consulting statistical yearbook/published literature, supplementing them by mean values of adjacent years (if the absent data only occurred in a few years), or by using data of neighbor counties/cities that they belong to (there was one exception: the fruit yield in Hulan county during 1990-2003, which was supplemented by Haerbin city data). For covariates, we used the poly2nb function of the spdep package (v1.1-5) to fill gaps: local downslope curvature, downslope curvature, land surface elevation, surface temperature at daytime, enhanced vegetation index in January & February and September & October, and topographic position index in Changhai county; the amount of phosphorus (by Olsen method) in Changbaichaoxianzu county, Changhai county, Ji’an city, Liaoyuan city and Suifenhe city; pH (in H2O) in Anda city, Antu county, Boli county, Changbaichaoxianzu county, Changhai county, Changling county, Dongning county, Hegang city, Helong city, Huadian city, Huanrenmanzuzizhi county, Huinan county, Jiamusi city, Ji’an city, Jiaohe county, Jingyu county, Liaoyuan city, Linjiang city, Liuhe county, Longjing city, Meihekou city, Qianan county, Qitaihe city, Shangzhi city, Suangyashan city, Suifenhe city, Tonghua city, Tonghua county, Tumen city, Wangqing county, Wushong county, Yanji city, Yanshou county and

Zhaozhou county; (b) calculating indicators which cannot be obtained directly using other available data or parameters. Detailed equations are given in Liu et al. (2020) and required parameters (N ratio of compound fertilizer, amount of N content in excretion and urine in humans and livestock, returning field rate of manure, cake ratio, amount of N content in different cake crops, returning field rate of cake fertilizer, the ratio of straw to grain, amount of N content in different straw and returning field rate of straw) used to calculate NUE indicators were presented in He et al. (2018); (c) data calibration: the final step of pre-processing and directly determining the quality of the database and accuracy of the results. For flagged suspicious values (i.e., outliers: 10 times larger or smaller than that of adjacent years), we double-checked the reference data and compared it with other available data (e.g., the economic yield must be equal to the total production divided by the planting area of the crop).

2 Results

2.1 Spatial and temporal variations of NUE indicators from 1990 to 2015

Spatial and temporal variations of PFP_N During the first period (1990-1995), PFP_N was very low in 32 counties (central and east Jilin province, southeast and northwest Liaoning province); low in 48 counties (within Jilin and Liaoning provinces); moderate in 37 counties (within Heilongjiang province, west Jilin province and central Liaoning province); high in 34 counties and very high in 32 counties (mostly in Heilongjiang province) (Fig. 2-B). A similar spatial pattern was found in other study periods between 1996 and 2015 (Fig. 2-C–F).

At the aggregated provincial level, the PFP_N decreased dramatically from 60 kg kg^{-1} in 1990 to 41 kg kg^{-1} in 2003 (with a peak of 52 kg kg^{-1} in 1999), and then was stable until 2015 (with a small peak around 50 kg kg^{-1} yr^{-1} during 2011-2012) in Heilongjiang, with an average annual growth rate of -1.4% (average PFP_N of 47 kg kg^{-1} yr^{-1}); Jilin had a moderate reduction from 32 kg kg^{-1} in 1990 to 22 kg kg^{-1} in 2000, followed by a fluctuation phase around 25 kg kg^{-1} yr^{-1}, and then stayed stable at around 30 kg kg^{-1} yr^{-1} from 2011 to 2015, with an average annual growth rate of -0.3% (average PFP_N of 27 kg kg^{-1} yr^{-1}); PFP_N in Liaoning went down marginally from 33 kg kg^{-1} to 27 kg kg^{-1} in 1994, and then rose gradually to 35 kg kg^{-1} in 2015, with an average annual growth rate of 0.2% (average PFP_N of 34 kg kg^{-1} yr^{-1}) (Fig. 3-A).

Spatial and temporal variations of PNB_N From 1990 to 1995, the PNB_N was very low in 41 counties (eastern Jilin, eastern and southern Liaoning); low in 62 counties (eastern Jilin and northern Liaoning); moderate in 34 counties (southern Heilongjiang, eastern and northern Jilin, northern Liaoning); high in 28 counties and very high in 18 counties (northern and central Heilongjiang) (Fig. 5-B). A similar spatial pattern was found over the entire study period (Fig. 5-C–F).

At the aggregated provincial level, the PNB_N in Heilongjiang decreased considerably from 1.22 kg kg^{-1} in 1990 to 0.85 kg kg^{-1} in 2003, thereafter increasing up to 1.12 kg kg^{-1} in 2012, and then kept steady around 1.00 kg kg^{-1} yr^{-1}, with an average annual growth rate of -0.9% (average PNB_N of 1.02 kg kg^{-1} yr^{-1}). Jilin had a significant reduction of PNB_N from 0.64 kg kg^{-1} in 1990 to 0.40 kg kg^{-1} in 2000, and then first fluctuated around 0.50 kg kg^{-1} yr^{-1} and later around 0.55 kg kg^{-1} yr^{-1} during 2010-2015, with an average annual growth rate of -0.5% (average PNB_N of 0.52 kg kg^{-1} yr^{-1}). The PNB_N in Liaoning changed from 0.51 to 0.40 kg kg^{-1} yr^{-1} during 1990-2015, with an average annual growth rate of -1.0% (average PNB_N of 0.44 kg kg^{-1} yr^{-1}).

2.2 **Prediction models and performance evaluation**

SMLR model Pearson's correlation coefficients were calculated to quantify the pairwise relationships between the NUE indicators and explanatory variables (Appendix G). PFP_N had a significantly negative correlation with EVI in September & October, EVI in January & February, annual average daily minimum temperature, planting area index of cereal and orchard and local upslope curvature; and no significant correlation with the gross output value of agriculture (agricultural GDP) and exchangeable acidity. PFP_N had a positive correlation with nine other explanatory variables (Appendix G-A). PNB_N was significantly negatively correlated with EVI in November & December, coarse fragments (> 2 mm), planting area index of cereal and melons, agricultural GDP and Calcisols, while it was significantly positively correlated with silt content, exchangeable acidity, total phosphorus, total carbon, pH (in KCl), soil organic carbon content, available soil water capacity (with FC=pF 2.3) and planting area index of beans. Only the planting area index of tubers had no significant correlation with PNB_N (Appendix G-B).

The initial stepwise regression model for PFP_N included 27 explanatory variables, which explained 51% of the variance of PFP_N (Appendix C). Planting area index of fiber, melons and tubers, saturated water content, soil organic carbon density, ferralsols, acrisols, cambisols, cation exchange capacity (CEC) and rural population were dropped from the regression model in the VIF step to remove multicollinearity. This reduced the complexity of the model to 17 explanatory variables. The statistical significance was carried out by the confint function, and all variables were significant. For PNB_N, the initial stepwise regression model included 27 explanatory variables, which explained 71% of the variance of PNB_N (Appendix C). EVI in January & February and in May & June, planting area index of oil, the amount of water-soluble phosphorus, cambisols, gypsisols, kastanozems, histosols cumulative, available soil water capacity until wilting point and vapor pressure were dropped from the regression model in the VIF step to remove multicollinearity. This reduced the complexity of the model to 15 explanatory variables. Then statistical significance was also carried out by confint function and all variables met the requirements.

3 Discussion

3.1 Spatial and temporal variations of NUE indicators

The results indicated that the PFP_N in the counties of Heilongjiang is higher than those counties of Jilin and Liaoning. Some counties, such as Tongyu county in Jilin and Fuxinmengguzuzizhi county in Liaoning tend to obtain lower yield with higher N input, which indicates high N loss and environmental threats. Zhao et al. (2016) indicated that ecological intensification improved PFP_N from 50 kg kg^{-1} yr^{-1} to 66 kg kg^{-1} yr^{-1} in Liufangzi, Jilin province. But they only considered the influence of chemical N application on PFP_N in most cases, especially for a specific field study, where chemical fertilizer was the major N input. Therefore, their PFP_N may be overestimated compared with the actual practice. In this study, we considered the N input from both chemical N and from organic sources. This is more meaningful for analysis at the regional scale, while also more organic fertilizer was applied along with the encouragement by Chinese government for agricultural sustainability. Despite that, only 57% of counties in 2015 met the requirement of well-managed systems (PFP_N > 60 kg kg^{-1} yr^{-1}, Dobermann 2007), when considering chemical fertilizer as N input. The corresponding critical PFP_N in this study should be 35 kg kg^{-1} yr^{-1} when we considered both chemical and organic N inputs. In summary, there is still a huge potential to improve PFP_N in China. PFP_N in most of counties decrease during the study period, only increasing in Huma county of Heilongjiang, Beipiao city of Liaoning and Antu county of Jilin. The different PFPN trends over time could not be explained and would require more detailed information than was available.

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Appendix B Description of covariates and their sources.

Classification[1)]	Covariates	Abbreviation[2)]	Description	Resolution	Unit	Source
Crop	Enhanced vegetation index	EVI_JanFeb; EVI_MarApr; EVI_MayJun; EVI_JulAug; EVI_SepOct; EVI_NovDec	Long-term annual mean monthly (Every two month) MODIS Enhanced Vegetation Index (EVI). Derived using a stack of MOD13Q1 EVI images.	250		SoilGrids (covariates)
	Crop types	See notes	The proportion of the planting area for crop types (10 kinds of crop type) in total planting area	County	%	Municipal Yearbook (1991-2016)
Topography	Land surface elevation	DEM	DEM based on 100 m resolution from EarthEnv-DEM90. No additional post-processing has been applied to remove vegetation and filter noise.	100	meter	SoilGrids (covariates)
	Terrain slope	SLPt	Terrain slope based on DEMMRG5 derived in SAGA GIS and expressed in radians.	250	radians	SoilGrids (covariates)
	Topographic position index	TPI	Topographic Position Index is the difference to the mean calculation (residual analysis) proposed by Wilson & Gallant (2000).	250	meter	SoilGrids (covariates)
	Downslope curvature	CUVd	Downslope Curvature based on DEMMRG5 derived in SAGA GIS.	250	meter	SoilGrids (covariates)
	Local upslope curvature	CUVlu	Local upslope Curvature based on DEMMRG5 derived in SAGA GIS.	250	meter	SoilGrids (covariates)
	Local downslope curvature	CUVld	Local downslope Curvature based on DEMMRG5 derived in SAGA GIS.	250	meter	SoilGrids (covariates)
Soil	Organic carbon stock	SOCs	Organic carbon stock in 0.3 m	250	tonnes ha^{-1}	SoilGrids
	Organic carbon content	SOCc	Organic carbon content in 0.3 m	250	$g\ kg^{-1}$	SoilGrids
	Organic carbon density	SOCd	Organic carbon density in 0.3 m	250	$kg\ m^{-3}$	SoilGrids
	Histosols cumulative probability	HP	Histosols cumulative probability in 2 m	250	%	SoilGrids
	pH in H_2O	PHw	pH in H_2O in 0.3 m	250		SoilGrids
	pH in KCl	PHk	pH in KCl in 0.3 m	250		SoilGrids

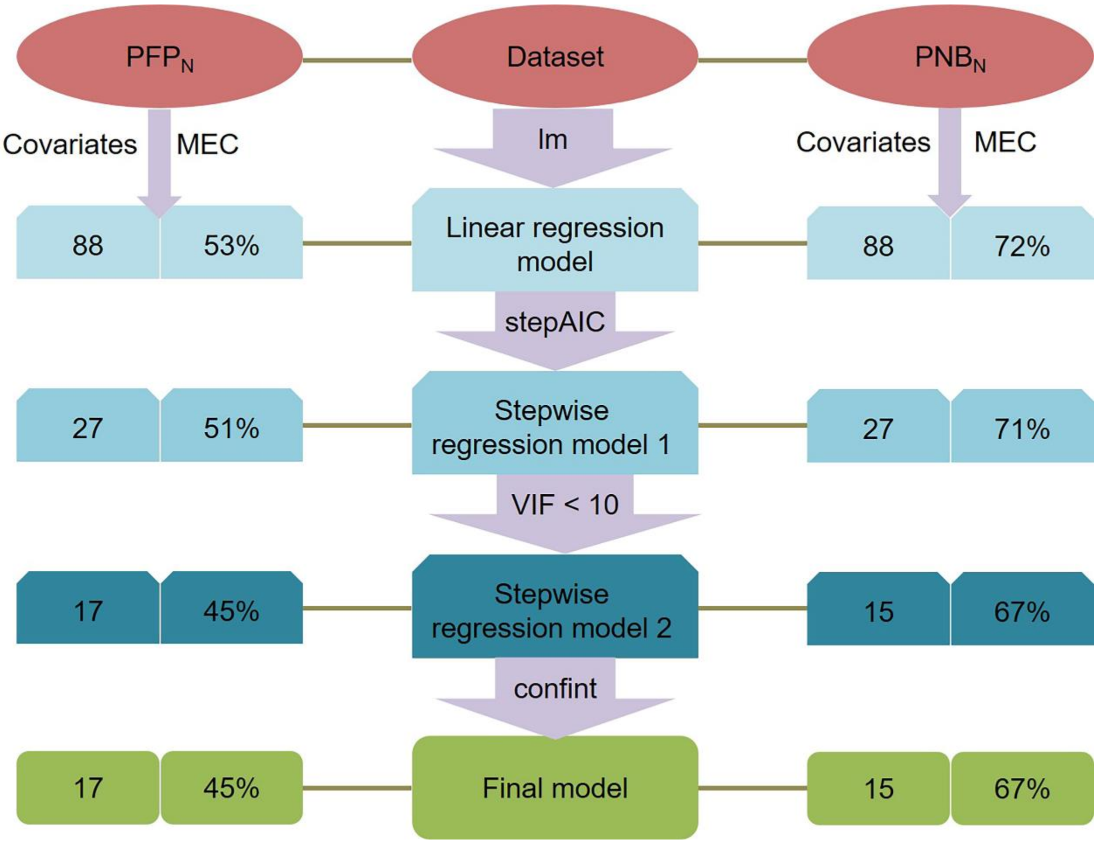
Sodic soil grade	SOD	Sodic soil grade in 2 m	250	grade	SoilGrids
Acid sub-soils grade	ACID	Acid sub-soils grade (0.15-0.6 m)	250	grade	SoilGrids
Coarse fragments volumetric	COA	Coarse fragments volumetric (>2mm) in 0.3 m	250	%	SoilGrids
Clay content	CLAY	Texture (clay content 0-2 micro meter) in 0.3 m	250	%	SoilGrids
Silt content	SILT	Texture (silt content 2-50 micro meter) in 0.3 m	250	%	SoilGrids
Sand content	SAND	Texture (sand content 50-2000 micro meter) in 0.3 m	250	%	SoilGrids
Bulk density	BD	Bulk density in 0.3 m	250	kg m^{-3}	SoilGrids
Absolute depth to bedrock	BED	Absolute depth to bedrock	250	cm	Shangguan et al, 2014
Soil types	SOIL	Soil types (17 kinds of soil type)	250	% or grade	SoilGrids
The amount of phosphorus by Olsen method	POL		1000	ppm of weight	Shangguan et al, 2014
The amount of water soluble phosphorus	PHO		1000	ppm of weight	Shangguan et al, 2014
Total carbon	TC		1000	% of weight	Shangguan et al, 2014
Total nitrogen	TN		1000	% of weight	Shangguan et al, 2014
Total phosphorus	TP		1000	% of weight	Shangguan et al, 2014
Total potassium	TK		1000	% of weight	Shangguan et al, 2014
Total sulfur	TS		1000	% of weight	Shangguan et al, 2014

Exchangeable aluminum	EXAL		1000	cmol kg^{-1}	Shangguan et al, 2014
Exchangeable calcium	EXCA		1000	cmol kg^{-1}	Shangguan et al, 2014
Exchangeable magnesium	EXMG		1000	cmol kg^{-1}	Shangguan et al, 2014
Exchangeable potassium	EXK		1000	cmol kg^{-1}	Shangguan et al, 2014
Exchangeable sodium	EXNA		1000	cmol kg^{-1}	Shangguan et al, 2014
Exchangeable acidity	EXH		1000	cmol kg^{-1}	Shangguan et al, 2014
Cation exchange capacity	CEC	Cation exchange capacity in 0.3 m	250	cmol kg^{-1}	SoilGrids
Electrical conductivity	EC		1000	ds m^{-1} or mmho cm^{-1}	Shangguan et al, 2014
Available soil water capacity with FC=pF 2.0	WATa1	Available soil water capacity with FC=pF 2.0 in 0.3 m	250	%	SoilGrids
Available soil water capacity with FC=pF 2.3	WATa2	Available soil water capacity with FC=pF 2.3 in 0.3 m	250	%	SoilGrids
Available soil water capacity with FC=pF 2.5	WATa3	Available soil water capacity with FC=pF 2.5 in 0.3 m	250	%	SoilGrids
Available soil water capacity (volumetric fraction) until wilting point	WATa	Available soil water capacity (volumetric fraction) until wilting point in 0.3 m	250	%	SoilGrids

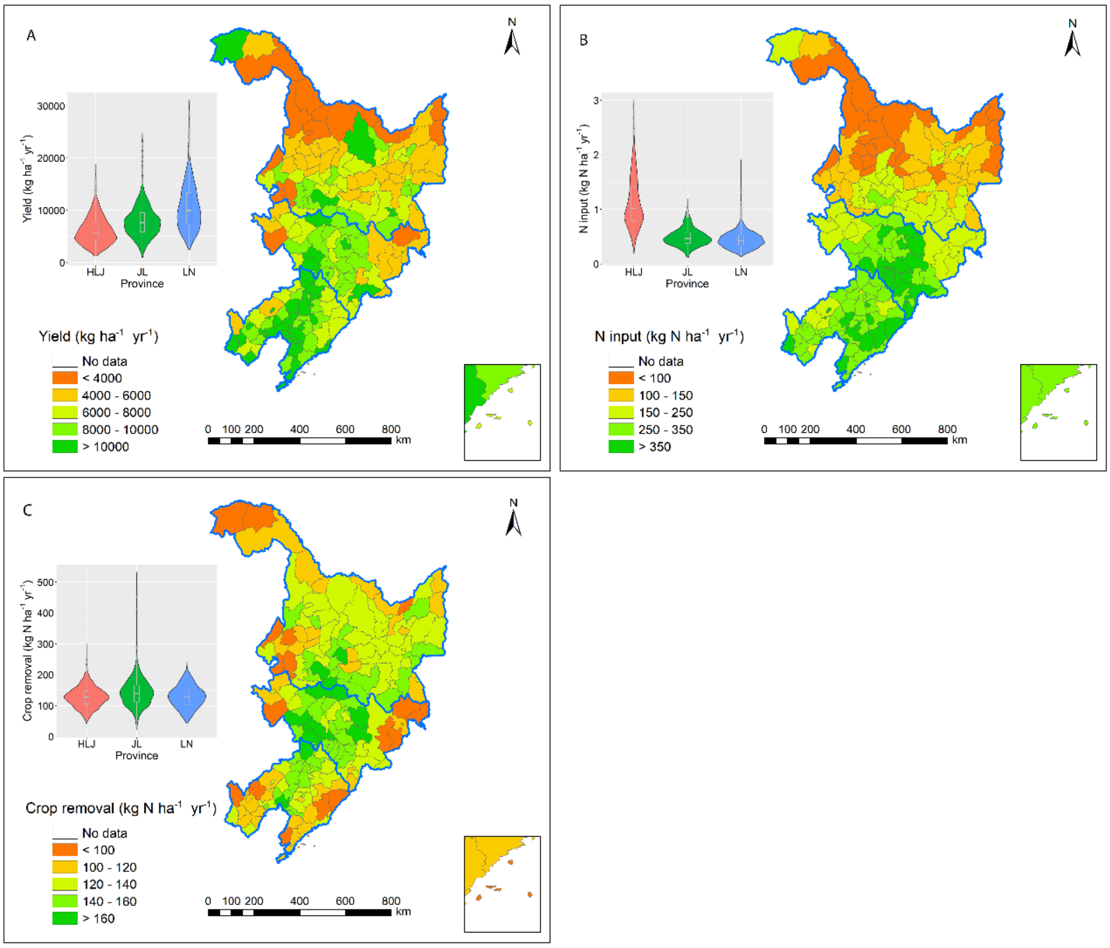
	Saturated water content (volumetric fraction)	WATs	Saturated water content (volumetric fraction) in 0.3 m	250	%	SoilGrids
Climate	Annual average surface temperature (daytime)	TMPd	Long-term averaged mean annual surface temperature (daytime) MODIS. Derived using a stack of MOD11A2 LST images.	1000	Kelvin	SoilGrids (covariates)
	Annual average surface temperature (nighttime)	TMPn	Long-term averaged mean annual surface temperature (nighttime) MODIS. Derived using a stack of MOD11A2 LST images.	1000	Kelvin	SoilGrids (covariates)
	Daily mean temperature	TMP		10000	℃	Climatic Research Unit
	Annual average daily maximum temperature	TMX		10000	℃	Climatic Research Unit
	Annual average daily minimum temperature	TMN		10000	℃	Climatic Research Unit
	Diurnal temperature range	DTR		10000	℃	Climatic Research Unit
	Cloud cover	CLD		10000	%	Climatic Research Unit
	Potential evapo-transpiration	PET		10000	Millimetres	Climatic Research Unit
	Vapour pressure	VAP		10000	Hecta-Pascals	Climatic Research Unit
	Wet day frequency	WET	Wet day frequency (rain days per year)	10000	Days	Climatic Research Unit
	Precipitation	PRE		10000	Millimetres	Climatic Research Unit
	Frost day frequency	FRS		10000	Days	Climatic Research Unit
Economy	Gross output value of agriculture	GDP		County	RMB ha^{-1}	Municipal Yearbook (1991-2016)
AMP	Rural population	POP	Rural population per planting area	County	person ha^{-1}	Municipal Yearbook (1991-2016)
	Available irrigation area index	IRRI	Available irrigation area per planting area		ha ha^{-1}	Municipal Yearbook (1991-2016)

[1] AMP: agricultural management practice.

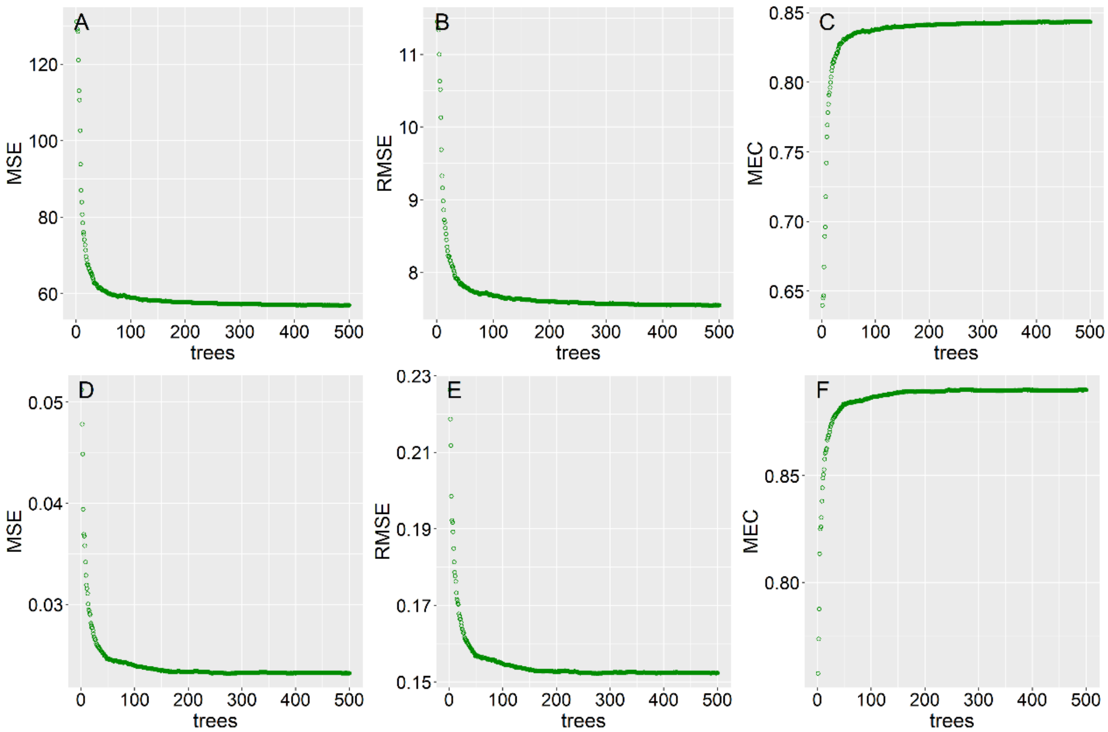
[2)] 17 soil types: Acrisols, ACR; Albeluvisols, ALB; Alisols, ALI; Andosols, AND; Arenosols, ARE; Calcisols, CAL; Cambisols, CAM; Chernozems, CHE; Cryosols, CRY; Durisols, DUR; Ferralsols, FER; Fluvisols, FLU; Gleysols, GLE; Gypsisols, GYP; Histosols, HIS; Kastanozems, KAS; Leptosols, LEP. 10 crop types: Cereal, CER; Beans, BEAN; Tubers, TUB; Oil, OIL; Sugar, SUG; Fiber, FIB; Tobacco, TOB; Vegetables, VEG; Orchards, ORC; Melons, MEL. Climatic Research Unit: (http://www.cru.uea.ac.uk/data); Shangguan, W., Dai, Y., Duan, Q., Liu, B., & Yuan, H. (2014). A global soil data set for earth system modeling. Journal of Advances in Modeling Earth Systems, 6(1), 249-263; SoilGrids. (https://soilgrids.org/).



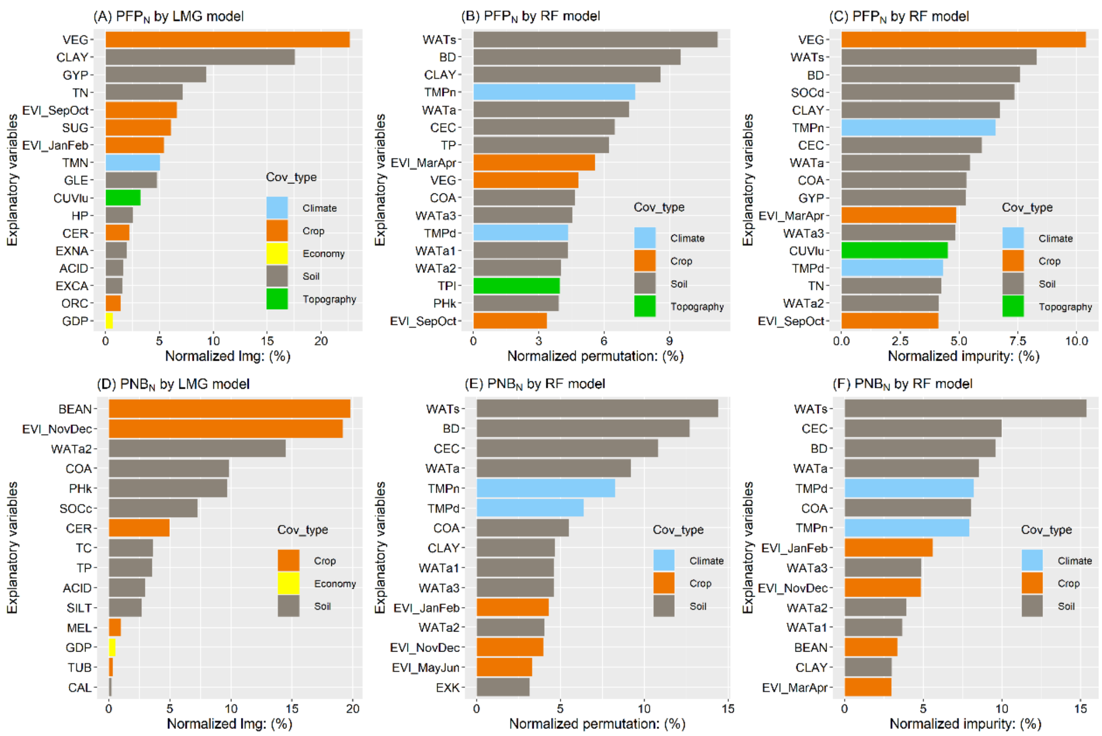
Appendix C The stepwise linear regression model building procedure. The R functions that were used for building the model are shown in the purple shaded arrows. MEC refers to the model efficiency coefficient of the regression models. VIF means variance inflation factor. The numbers under "covariates" represent the number of covariates in the model, and the percentage numbers under the MEC indicate how much MEC is explained by the model.



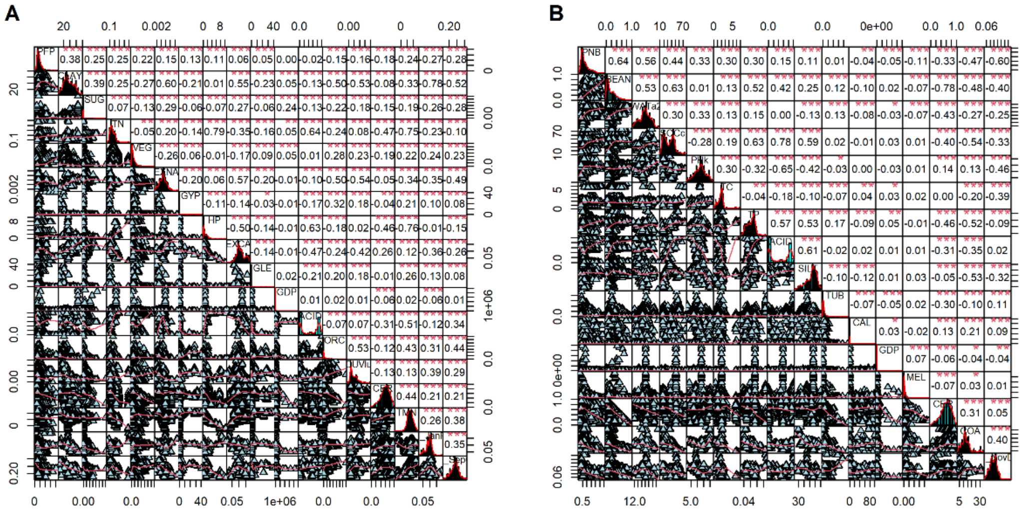
Appendix D Spatial distribution of average yield (A), nitrogen input (B) and crop removal (C) from 1990 to 2015.



Appendix E Cumulative MSE (A, D), RMSE (B, E) and MEC (C, F) (scatter plot: x axis- trees) in Random Forest models for the PFP_N and PNB_N, respectively.



Appendix F Relative importance of explanatory variables for the PFP$_N$: A) LMG method in in calc.relimp function; B) permutation method and C) impurity methods for RF model with ranger function; and for PNB$_N$: D) LMG method in calc.relimp function; E) permutation method and F) impurity methods for RF model with ranger function. Note: See Abbreviations (Appendix B) for explanations of the variable code names.



Appendix G Pairwise comparison of all explanatory variables used in the PFP_N (A) and PNB_N (B) regression models. The upper-right panel contains predicted pair-wise Pearson's correlations. The diagonal panel shows histograms and the lower-left panel shows scatterplots with a LOESS smoother added to aid visual interpretation. Top row and left columns show bivariate relations between yield and explanatory variables. Note: See Abbreviations (Appendix B) for explanation of the variable code names. *, *P-value*<0.05; **, *P-value*<0.01; ***, *P-value*<0.001.