Reviewer 1

Overall comment: This study reconstructs and analyzes long-term phosphorus (P) surplus in Europe from 1850 to 2019 at a high spatial resolution, providing a comprehensive dataset that includes 48 P surplus estimates and accounts for various uncertainties. The findings reveal that P surplus across the EU-27 has tripled over 170 years, highlighting the need for targeted management strategies. Additionally, the study identifies big variation in P surplus estimates by comparing its database with major previously published P databases and highlights areas for improvement. This research contributes to the improvement of quantification of historical and spatial P budgets in Europe by considering varying methods, including more budget terms, extending the temporal span of current studies, increasing resolution, and comparing various datasets. However, a few comments regarding the calculation details and discussions are provided for the authors' consideration.

Reply: We thank the Reviewer for their positive comments to the manuscript. We reply below to the points raised by the Reviewer.

1.1 — Section 2.1:

- A more detailed definition of each rate term needs to be provided wherever a rate appears. Since the rates are always expressed in $kg\ ha^{-1}\ yr^{-1}$ in this study, is the area being referred to the grid area or a specific land type area? Only part of the rate terms in this manuscript indicate which area is used as the denominator.

  Reply: Thank you for this valuable feedback. In response to the suggestion, we have thoroughly reviewed and updated the manuscript to provide clearer definitions of all rate terms. Specifically, we have clarified the land use area associated with each rate term to ensure a consistent and precise interpretation of phosphorus (P) surplus values and associated components. Throughout the manuscript, we now explicitly state the specific land use areas (such as grid physical area, agricultural area, cropland, or pasture) that correspond to the units presented.

  The rates are now expressed as follows:

  - $kg\ ha^{-1}\ yr^{-1}$ of physical area for grid-level estimates,
  - $kg\ ha^{-1}\ yr^{-1}$ of agricultural area for agricultural land,
  - $kg\ ha^{-1}\ yr^{-1}$ of cropland area for croplands,
  - $kg\ ha^{-1}\ yr^{-1}$ of pasture area for permanent pastures, and
  - $kg\ ha^{-1}\ yr^{-1}$ of grassland area for grassland (both temporary and permanent pastures).

  For instance, in line 9 of the revised manuscript, we have updated the phrasing to explicitly refer to "physical area" as follows: *"Specifically, the total P surplus across the EU-27 has tripled over 170 years, from 1.19 (±0.28) $kg\ ha^{-1}$ of physical area in 1850 to around 2.48 (±0.97) $kg\ ha^{-1}$ of physical area yr^{-1} in recent years"*. This updated phrasing ensures that readers are aware of the specific land use area considered in the corresponding rate calculations. We have made similar adjustments throughout the manuscript wherever needed to ensure that the respective land type (physical area, cropland, pasture, grassland, or agricultural area) is clearly specified.

- Since more definitions of land types and other budget terms are provided in the following sections, I suggest that the authors move Section 2.1 to the end of Section 2.

Reply: Thank you for your valuable feedback regarding the structure of Section 2. We understand your suggestion to move Section 2.1, but we believe that the current organization is structured in a way to retain the overall clarity and coherence of the manuscript. To this end, our intention was to provide an initial overview of phosphorus (P) surplus, presenting both agricultural and non-agricultural contexts, to establish a basic foundation for the subsequent detailed definitions and methodologies. We believe that this approach allows readers to grasp the broader significance of P surplus before delving into the specific details of land use classifications and P budget components.

By outlining the P surplus early in the manuscript, we guide readers through a step-wise progression, starting with the general concept of P surplus and then moving into more specific discussions about land use types and detailed methodological steps. This structure, in our view, ensures a more intuitive understanding of the complex processes and terms discussed in the manuscript.

To take this point into consideration and to better clarify this structure, we added an introductory texts to the paragraph starting at line 105 in section 2 of the revised manuscript, in which the flow of information is explained as follows:

"In the following sections, we first outline the definition of P surplus in both agricultural and non-agricultural soils. Next, we present a summary of the methodology used to reconstruct the land use types, including agricultural land, namely cropland and pasture, and non-agricultural land, including non-vegetated areas, semi-natural vegetation, forest, and urban areas. Crop-specific harvested areas for non-fodder and fodder crops are also defined. We refer to Batool et al. (2022) for detailed methodologies. Finally, we describe the steps employed to reconstruct P inputs, including fertilizer, manure, atmospheric deposition, and chemical weathering, and P outputs, focusing on P removal from cropland and pastures."

1.2 — Section 2.3:

- Line 215: Do you mean the IFA report published in 2017 instead of 2013 that contains 2014-2015 data?

Reply: Thank you for pointing this out this typing error. We have used the IFA report published in 2017 and therefore updated the reference in line 274 of the revised manuscript.

- Section 2.3.3: Not all terms in each equation are clearly defined in the text after the equation, in equations 15-23. For example, what is P2O5fercrops in Equation 15, what is nu and i in equations 22-23? It may seem redundant since some of the terms are discussed before the equations, but it would be easier for readers who are not familiar with these terms if each term's definition is clearly listed after the equations. Or, since this manuscript includes many parameters, it would be very helpful to include a table at the end listing all parameters' names, definitions, units, and/or calculation methods.

Reply:

Thank you for the insightful suggestion regarding the clarity of the terms in the equations. Based on your suggestion, we have reviewed and updated the manuscript to ensure that each term in equations 15–23 (now 27-35 in the revised manuscript) (and others) is clearly defined in the text following the equations.

Further, we have incorporated a **detailed table, Appendix Table A1 in the revised manuscript** that lists all variables, including their description and units, as suggested by the reviewer. We have also attached the Table R1 below for your quick reference. We believe these adjustments will clarify the manuscript and aid readers in following the notations.

Table R1: Table of variables used in the P surplus calculation and related sections, listing each variable, its description, and units. The variables cover multiple components of the phosphorus budget, including inputs, outputs, and surplus calculations across different land-use categories.

Variable	Description	Unit
General Variables		
P	Phosphorus	-
N	Nitrogen	-
i	Grid cell index	-
y	Year index	-
u	Country index	-
c	Crop index	-
l	Livestock category index	-
n_u	Number of grid cells in country u	-
n_c	Number of grid cells for each crop category c	-
n_l	Number of grid cells for each livestock category l	-
P Surplus		
$Surp_{\text{soil}}$	Total P surplus	$kg\ ha^{-1}$ of physical area yr^{-1}
$Surp_{\text{agri}}$	P surplus in agricultural areas (cropland and pasture)	$kg\ ha^{-1}$ of physical area yr^{-1}
$Surp_{\text{NonAgri}}$	P surplus in non-agricultural areas	$kg\ ha^{-1}$ of physical area yr^{-1}
$Surp_{\text{cr}}$	P surplus in cropland	$kg\ ha^{-1}$ of physical area yr^{-1}
$Surp_{\text{past}}$	P surplus in pasture	$kg\ ha^{-1}$ of physical area yr^{-1}
Inp_{cr}	Total P inputs in cropland	$kg\ ha^{-1}$ of physical area yr^{-1}
Out_{cr}	Total P removal from cropland	$kg\ ha^{-1}$ of physical area yr^{-1}
$FERT_{\text{cr}}$	Fertilizer P inputs in cropland	$kg\ ha^{-1}$ of physical area yr^{-1}
MAN_{cr}	Manure P inputs in cropland	$kg\ ha^{-1}$ of physical area yr^{-1}
Dep_{cr}	Atmospheric P deposition in cropland	$kg\ ha^{-1}$ of physical area yr^{-1}
CW_{cr}	P inputs from chemical weathering in cropland	$kg\ ha^{-1}$ of physical area yr^{-1}
Rem_{cr}	P removal by crop uptake from cropland	$kg\ ha^{-1}$ of physical area yr^{-1}
Inp_{past}	Total P inputs in pasture	$kg\ ha^{-1}$ of physical area yr^{-1}
Out_{past}	Total P removal from pasture	$kg\ ha^{-1}$ of physical area yr^{-1}
$FERT_{\text{past}}$	Fertilizer P inputs in pasture	$kg\ ha^{-1}$ of physical area yr^{-1}
MAN_{past}	Manure P inputs in pasture	$kg\ ha^{-1}$ of physical area yr^{-1}
Dep_{past}	Atmospheric P deposition in pasture	$kg\ ha^{-1}$ of physical area yr^{-1}
CW_{past}	P inputs from chemical weathering in pasture	$kg\ ha^{-1}$ of physical area yr^{-1}
Rem_{past}	P removal by grass uptake in pasture	$kg\ ha^{-1}$ of physical area yr^{-1}
$Surp_{\text{For}}$	P surplus in forest areas	$kg\ ha^{-1}$ of physical area yr^{-1}
$Surp_{\text{NatVeg}}$	P surplus in semi-natural vegetation areas	$kg\ ha^{-1}$ of physical area yr^{-1}
$Surp_{\text{NonVeg}}$	P surplus in non-vegetated areas	$kg\ ha^{-1}$ of physical area yr^{-1}
$Surp_{\text{Urban}}$	P surplus in urban areas	$kg\ ha^{-1}$ of physical area yr^{-1}
DEP_{For}	Atmospheric P deposition in forest areas	$kg\ ha^{-1}$ of physical area yr^{-1}
CW_{For}	P inputs from chemical weathering in forest areas	$kg\ ha^{-1}$ of physical area yr^{-1}
DEP_{NatVeg}	Atmospheric P deposition in semi-natural vegetation areas	$kg\ ha^{-1}$ of physical area yr^{-1}
CW_{NatVeg}	P inputs from chemical weathering in semi-natural vegetation areas	$kg\ ha^{-1}$ of physical area yr^{-1}
DEP_{NonVeg}	Atmospheric P deposition in non-vegetated areas	$kg\ ha^{-1}$ of physical area yr^{-1}
CW_{NonVeg}	P inputs from chemical weathering in non-vegetated areas	$kg\ ha^{-1}$ of physical area yr^{-1}
DEP_{Urban}	Atmospheric P deposition in urban areas	$kg\ ha^{-1}$ of physical area yr^{-1}
Land use		
$R_{\text{HYDE-cr}}$	Ratios represent the relative change (temporal variability) in cropland over time, normalized to the year 2000	-
$A_{\text{HYDE-cr}}$	Cropland area from HYDE	ha
$R_{\text{HYDE-past}}$	Ratios represent the relative change (temporal variability) in pasture area over time, normalized to the year 2000	-
A_{cr}	Cropland area in our study	ha
$A_{\text{Ramankutty-cr}}$	Cropland area from Ramankutty et al. (2008)	ha
A_{past}	Pasture area in our study	ha
$A_{\text{Ramankutty-past}}$	Pasture area from Ramankutty et al. (2008)	ha
$R_{A_{\text{cr}}}$	Ratios between cropland area from FAOSTAT and cropland area calculated in our study	-
$A_{FAO-\text{cr}}$	Cropland area from FAOSTAT	ha

$R_{A_{\text{past}}}$	Ratios between pasture area from FAOSTAT and pasture area calculated in our study	-
$A_{FAO-\text{past}}$	Pasture area from FAOSTAT	ha
$A_{\text{cr}}^{\text{cor}}$	Corrected cropland area (i.e. harmonized at country level with FAOSTAT country level cropland data) in our study	ha
$A_{\text{past}}^{\text{cor}}$	Corrected pasture area (i.e. harmonized at country level with FAOSTAT country level pasture area data) in our study	ha
$A_{\text{agri}}^{\text{cor}}$	Corrected agriculture area in our study	ha
Crop specific harvested area		
R_A	Ratio between crop-specific harvested areas calculated in our study and crop-specific harvested areas from FAOSTAT	-
$A_{crops_{FAO}}$	Crop-specific harvested areas from FAOSTAT	ha
A_{crops}	Crop-specific harvested areas in our study	ha
$A_{\text{crops}}^{\text{cor}}$	Corrected crop-specific harvested areas in our study	ha
Mineral Fertilizer		
$Pfer_{Missing_{East_{EU}}}$	Total fertilizer application for the missing countries and time period from FAOSTAT	$kg\ yr^{-1}$
$Pfer_{East_{EU}}$	Total fertilizer application for Eastern European countries (Czechoslovakia, Hungary and Bulgaria) with available data in FAOSTAT	$kg\ yr^{-1}$
$Pfer_{soil}$	Total P fertilizer application	$kg\ yr^{-1}$
$Pfer_{crops}$	P fertilizer application for different non-fodder crop types	$kg\ yr^{-1}$
$P_2O_5fer_{crops}$	P fertilizer application in the form of phosphate for different non-fodder crop types from IFA	$kg\ yr^{-1}$
$Pfer_{crops_{Rate}}$	Fertilizer application rates for different non-fodder crop types	$kg\ ha^{-1}$ of crop harvested areas yr^{-1}
$A_{crops_{FAO}}$	Crop-specific harvested area for different non-fodder crop types from FAOSTAT	ha
$Pfer_{grass_{Rate}}$	Fertilizer application rates for grassland	$kg\ ha^{-1}$ of grassland areas yr^{-1}
$Pfer_{grass}$	P fertilizer application for grassland (covering temporary and permanent grasslands and pastures for silage, hay, and grazing) from IFA	$kg\ yr^{-1}$
$A_{grass_{FAO}}$	Grassland area (including both permanent pastures and temporary grassland) from FAOSTAT	ha
$Pfer_{past}$	P fertilizer application for permanent pasture	$kg\ yr^{-1}$
$Pfer_{fodder}$	P fertilizer application for fodder crops	$kg\ yr^{-1}$
$Pfer^{\text{cor}}_{\text{crops}_{Rate}}$	Adjusted/corrected fertilizer application rates for crops	$kg\ ha^{-1}$ of crop harvested areas yr^{-1}
$Pfer^{\text{cor}}_{\text{grass}_{Rate}}$	Adjusted/corrected fertilizer application rates for grassland	$kg\ ha^{-1}$ of grassland areas yr^{-1}
Animal Manure		
R_{man}	Ratio of animal manure production between 1850–1889 and the base year 1890	-
man	Animal manure production for a given country	$kg\ yr^{-1}$
L	Estimated livestock number for each livestock category in a country	$head$
$Wratio$	Weighted livestock ratio per grid cell	-
$P_{man}(l)$	Total P manure production for each livestock category	$kg\ head^{-1}\ yr^{-1}$
P_{coeff}	P manure excretion coefficient for each livestock category	$kg\ head^{-1}\ yr^{-1}$
$R_{P_{man}}$	Country-level correction factor (as ratios) for P manure produced to match with FAOSTAT country-level estimates	-
$P_{man_{FAO}}$	Total P manure production for each livestock category derived from FAOSTAT	$kg\ head^{-1}\ yr^{-1}$
P_{man}^{cor}	Adjusted/corrected P manure produced in our study to match with FAOSTAT country-level estimates	$kg\ head^{-1}\ yr^{-1}$
P_{man}	Total P manure production	$kg\ yr^{-1}$
$R_{Nman_{treat}/prod}$	Country-level ratios of treated N manure to total excreted N manure	-
$Nman_{treat}$	Treated N manure	$kg\ yr^{-1}$
$Nman_{prod}$	Total N manure produced	$kg\ yr^{-1}$
$R_{Nman_{left}/prod}$	Country-level ratios of N manure left on pasture to total excreted N manure	-
$Nman_{left}$	N manure left on pasture	$kg\ yr^{-1}$
Man_{treat}	Treated manure	$kg\ yr^{-1}$
Man_{left}	Manure left on pasture	$kg\ yr^{-1}$
Man_{cr}	Manure applied to cropland	$kg\ ha^{-1}$ of physical area yr^{-1}
$Prop_{A_{\text{cr}}}$	Proportion of cropland area in the total agricultural area in a grid cell	-
A_{grid}	Area of grid cell	ha
$Man_{app_{past}}$	Manure applied to pasture areas	$kg\ ha^{-1}$ of physical area yr^{-1}
$Prop_{A_{\text{past}}}$	Proportion of pasture area in the total agricultural area in a grid cell	-

Man_{past}	Total manure applied to pasture (sum of manure applied to pasture and manure left on pasture)	$kg\ ha^{-1}$ of physical area yr^{-1}
Chemical Weathering		
CW_{Rate}	Rates of P release from chemical weathering	$kg\ ha^{-1}$ of physical area yr^{-1}
A_{For}	Forest area	ha
A_{NatVeg}	Semi-vegetation area	ha
P Removal		
P_{content}	Crop-specific P content of each crop type	$kg\ t^{-1}$
Pro_{crops}	Crop production	$t\ yr^{-1}$
$Pro_{\text{crops}_{Monfreda}}$	Crop production derived from Monfreda et al. (2008)	$t\ yr^{-1}$
$Rem^{coeff}_{\text{past}}$	Coefficient for P removal in pasture	-

– Line 230: The fertilizer application rates here are defined as total fertilizer use/harvest area. I was wondering if the authors use the same definition for all fertilizer application rates discussed in the study, such as the data from Batool et al. (2022) mentioned later, and in equations 18-20. It seems that the fertilizer application rates are defined as total fertilizer use/land use area in equations 18-20. However, land use areas may not correspond to harvest areas.

Reply: Thank you for this remark. We appreciate the opportunity to clarify our approach to defining and applying fertilizer application rates for different land-use types in our study. We consistently used two distinct methods for the application rates based on the land-use type:

- **Non-Fodder Crops:** For non-fodder crops, fertilizer application rates ($Pfer_{crops_{Rate}}$) ($kg\ ha^{-1}$ of crop harvested areas yr^{-1}) are calculated as total fertilizer use ($Pfer_{crops}$) ($kg\ yr^{-1}$) divided by the corresponding harvested area (A_{crops}) (ha) for each crop type (c) and country (u) for the year 2015 (y_{2015}). This approach ensures that the fertilizer rate corresponds specifically to the harvested area of each non-fodder crop, as shown in equation 28 of the revised manuscript (equation R1 here):

$$Pfer_{crops_{Rate}}(u,c,y_{2015}) = \frac{Pfer_{crops}(u,c,y_{2015})}{A_{crops_{FAO}}(u,c,y_{2015})} \tag{R1}$$

- **Grasslands (temporary and permanent):** For grasslands, which include both temporary grassland (represented as one of the six fodder crop categories) and permanent pasture, we calculated a single fertilizer application rate ($Pfer_{grass_{Rate}}$) ($kg\ ha^{-1}$ of grassland areas yr^{-1}) by combining IFA fertilizer usage data ($Pfer_{grass}$) ($kg\ yr^{-1}$) with the corresponding grassland area ($A_{grass_{FAO}}$ (ha). In this case, the total grassland area reflects both temporary and permanent pasture lands used for grazing and forage production in equation 29 of the revised manuscript (equation R2 here):

$$Pfer_{grass_{Rate}}(u,y_{2015}) = \frac{Pfer_{grass}(u,y_{2015})}{A_{grass_{FAO}}(u,y_{2015})} \tag{R2}$$

- **Application in equations 18–20 (equations 30–32 in Revised Manuscript):** In equation 18 (equation 30 in revised manuscript and equation (R3) here), we applied the country-level fertilizer rates of non-fodder crops ($Pfer_{\text{crops}}$) ($kg\ ha^{-1}$ of crop harvested areas yr^{-1}) to gridded harvested areas for non-fodder crops ($A^{\text{cor}}_{\text{crops}}$) ($ha$) for each year to derive fertilizer application amounts ($Pfer_{\text{crops}}$) ($kg\ yr^{-1}$) per grid cell for each crop.

$$Pfer_{\text{crops}}(i,c,y_{1850,-,2019}) = Pfer_{crops_{Rate}}(u,c,y_{2015}) \times A^{\text{cor}}_{\text{crops}}(i,c,y1850-2019) \tag{R3}$$

In equation 19-20 (equations 31-32 in revised manuscript and equations (R4–R5) here), we applied the country-level fertilizer rates of grassland to both fodder crop harvested areas and permanent pasture areas. Specifically, for permanent pasture and fodder crops, the fertilizer application rate ($Pfer_{grass_{Rate}}$ (($kg\ ha^{-1}$ of grassland areas yr^{-1}))) was multiplied by the gridded pasture area and harvested area of each fodder crop

($A^{\text{cor}}_{\text{past}}$ and A_{fodder} in ha, respectively) to estimate the fertilizer application amount for pasture and fodder crops ($Pfer_{\text{past}}$ and $Pfer_{\text{fodder}}$, in $kg\ yr^{-1}$), respectively).

$$Pfer_{\text{past}}(i, y_{1850-2019}) = Pfer_{grass_{Rate}}(u, y_{2015}) \times A^{\text{cor}}_{\text{past}}(i, y_{1850-2019}) \quad \text{(R4)}$$

$$Pfer_{\text{fodder}}(i, c, y_{1850-2019}) = Pfer_{grass_{Rate}}(u, y_{2015}) \times A_{fodder}(i, c, y_{1850-2019}) \quad \text{(R5)}$$

We realize that these distinctions may not have been sufficiently emphasized in the original text. To clarity this, we have updated the manuscript to specify that fertilizer rates are based on crop-specific harvested areas. For clarity, we have revised the text in section 2.3.3 at line 290 of the revised manuscript:

We calculated fertilizer application rates by combining IFA fertilizer usage data with FAOSTAT's crop-specific harvested area ($A_{crops_{FAO}}$ (ha)) and grassland area ($A_{grass_{FAO}}$ (ha)). Grassland areas encompass temporary grasslands (represented as one of the six fodder crop categories) as well as permanent pastures, with a single fertilizer application rate applied consistently across all grassland uses for grazing and forage production.

For further clarification, we have added the following text in line 303:

Further, it is important to note that fertilizer application rates for non-fodder crops are based on fertilizer use per unit of corresponding harvested area to represent crop-specific fertilizer inputs. For grassland, encompassing both temporary and permanent pastures, the fertilizer application rate is calculated using total grassland area, and ensuring consistent application of this rate across all relevant grassland areas.

We hope that the above clarifications address potential confusion and ensure transparency regarding how fertilizer application rates are defined and applied to different land-use categories in our analysis.

– Equations 37-38: If the unit of Pman is $kg\ yr^{-1}$, why are the units of Mantreat and Manleft (=Pman*ratio) $kg\ ha^{-1}\ yr^{-1}$? Were any details omitted from the equations? Is the gridded area or the cropland+pasture area used as the denominator to calculate the application rate? It seems that you used the grid's physical area as the denominator based on Line 365. However, based on your Equations 41-42, it seems that the country total cropland+pasture area is used as the denominator (since you assume that "the entire amount of treated P manure is applied to soil" according to line 355 and "equal distribution rates for cropland and pasture within each grid cell" in Line 380)?

Reply: We appreciate your careful review of equations 37-38 (now 51-52 of the revised manuscript) and your comments regarding the units of P_{man}, Man_{treat}, and Man_{left}. Upon further inspection, we acknowledge that there was an inconsistency in how these units were represented. In the original equations, Man_{treat} and Man_{left} were incorrectly expressed in $kg\ ha^{-1}\ yr^{-1}$ rather than $kg\ yr^{-1}$.

In the revised version of the manuscript, we have updated this by correcting the text in section 2.3.8 at line 451-460. To clarify, the manure applied to cropland (Man_{cr}) ($kg\ ha^{-1}$ of physical area yr^{-1}) is calculated by dividing the total treated manure (Man_{treat}) ($kg\ yr^{-1}$) by the grid's physical area (A_{grid}) (ha) and multiplying it by the proportion of cropland within the grid cell ($Prop_{A_{\text{cr}}}$) (-), as outlined in equation R6 (equation 53 of the revised manuscript). Similarly, the manure designated for pasture application ($Man_{app_{past}}$) ($kg\ ha^{-1}$ of physical area yr^{-1}) is determined by dividing the treated manure (Man_{treat}) ($kg\ yr^{-1}$) by the grid's physical area (A_{grid})(ha) and multiplying it by the proportion of pasture area within the grid cell ($Prop_{A_{\text{past}}}$) (-), as indicated in equation R7 (equation 54 of the revised manuscript). Finally, the total manure allocated to pastures (Man_{past}) ($kg\ ha^{-1}$ of physical area yr^{-1}) is calculated by summing the manure applied to pastures ($Man_{app_{past}}$) ($kg\ ha^{-1}$ of physical area yr^{-1}) and the manure left on pastures by grazing animals (Man_{left}) ($kg\ yr^{-1}$) normalized by the grid's physical area (A_{grid}) (ha) as shown in equation R8 (equation 55 of the revised manuscript).

$$Man_{cr}(i, y_{1850-2019}) = \frac{Man_{treat}(i, y_{1850-2019}) \times Prop_{A_{\mathrm{cr}}}(i, y_{1850-2019})}{A_{\mathrm{grid}}(i)}, \tag{R6}$$

$$Man_{app_{past}}(i, y_{1850-2019}) = \frac{Man_{treat}(i, y_{1850-2019}) \times Prop_{A_{\mathrm{past}}}(i, y_{1850-2019})}{A_{\mathrm{grid}}(i)}, \tag{R7}$$

$$Man_{past}(i, y_{1850-2019}) = Man_{app_{past}}(i, y_{1850-2019}) + \frac{Man_{left}(i, y_{1850-2019})}{A_{\mathrm{grid}}(i)}. \tag{R8}$$

These adjustments maintain consistency in the units and accurately reflect the distribution of treated manure between cropland and pasture. We hope these clarifications resolve the confusion, and we have also included further explanations in line 451-460 for each variable and equation 53-55 in the revised manuscript.

Thank you again for pointing this out, and we have addressed the issue in both the equations and the corresponding text in the revised manuscript.

– Lines 430-435: You may want to add explanations for CW, C, and A after the equations or refer to a table listing all parameters. Do the area parameters C and A correspond only to the areas that are both of a specific land use type and have chemical weathering?

Reply: Thank you for highlighting this point. Upon revising the manuscript, we recognized potential confusion with the area parameters, particularly with the notation for corrected/normalized areas. Previously, we used C_A to denote corrected or normalized areas (i.e., our gridded cropland and pasture areas corrected at the country level to align with FAOSTAT's country-level estimates). To improve clarity, we have now revised this notation to $A_{\mathrm{cr}}^{\mathrm{cor}}$ throughout the manuscript (for instance, in equations 20-21 and subsequent equations), ensuring that A **consistently represents area terms**, and the "cor" subscript indicates correction.

To clarify on the parameters specifically asked in your comments:

– **CW** refers to the P inputs from chemical weathering, expressed in $kg\ ha^{-1}$ of physical area yr^{-1}.

– **A** represents the gridded area for each specific land-use type (cropland, pasture, forest, natural vegetation, and grid area), denoted as $A_{\mathrm{cr}}^{\mathrm{cor}}$, $A_{\mathrm{past}}^{\mathrm{cor}}$, A_{For}, A_{NatVeg}, and A_{grid} (in ha), respectively. These parameters represents the area of a particular land-use type within each grid cell or the grid physical area.

In response to your feedback on line 430-435 (now 525-528 in the revised manuscript), we have added explanations for CW and A directly following the equations in the revised manuscript to make them clearer. Furthermore, we have revised equations 44-48 (now 56-59 in the revised manuscript, shows as equations R9- R12 below) to normalize these values using the grid's physical area, providing P inputs in units of $kg\ ha^{-1}$ of physical area yr^{-1}, which accurately reflects spatial estimates.

The updated equations (56-59) and subsequent text in lines 530-534 now read as follows:

$$CW_{cr}(i, y_{1850-2019}) = \frac{A_{\mathit{cr}}^{\mathit{cor}}(i, y_{1850-2019}) \times CW_{Rate}(i)}{A_{\mathit{grid}}(i)} \tag{R9}$$

$$CW_{past}(i, y_{1850-2019}) = \frac{A_{\mathit{past}}^{\mathit{cor}}(i, y_{1850-2019}) \times CW_{Rate}(i)}{A_{\mathit{grid}}(i)} \tag{R10}$$

$$CW_{For}(i, y_{1850-2019}) = \frac{A_{\mathit{For}}(i, y_{1850-2019}) \times CW_{Rate}(i)}{A_{\mathit{grid}}(i)} \tag{R11}$$

$$CW_{NatVeg}(i, y_{1850-2019}) = \frac{A_{\mathit{NatVeg}}(i, y_{1850-2019}) \times CW_{Rate}(i)}{A_{\mathit{grid}}(i)} \tag{R12}$$

where CW_{cr}, CW_{past}, CW_{For}, and CW_{NatVeg} refer to P inputs from chemical weathering for areas covered by cropland, pasture, forest, and natural vegetation, respectively, in $kg\ ha^{-1}$ of physical area yr^{-1}; A_{cr}^{cor}, A_{past}^{cor}, A_{For}, A_{NatVeg}, and A_{grid} represent the gridded areas of cropland, pasture, forest, natural vegetation, and the grid's physical area, respectively, in ha; and CW_{Rate} denotes the P release rate from chemical weathering, based on lithological data, in $kg\ ha^{-1}$ of physical area yr^{-1}.

To further assist the reader, we have included all variables in the Appendix table A1 in the revised manuscript, where each variable, its description, and its unit are listed.

1.3 — Sections 2.4.1-2.4.2:

– It seems that Section 2.4.2 would be better placed before Section 2.4.1.?

Reply: Thank you for your suggestion regarding the order of Sections 2.4.1 and 2.4.2. We understand the logic behind presenting 'Crop Production' first, as it forms the basis for calculating P removal from pastures. However, we have deliberately structured the manuscript to first describe P removal from pasture, which utilizes crop production data, followed by the detailed description of how we constructed the crop production dataset. This flow ensures that the methodological steps are clearly outlined and remain logically connected. We hope the reviewer now recognizes our viewpoint regarding this structural decision, which supports the overall clarity of the presentation.

1.4 — Section 3.1:

– Including a series of maps showing accumulated P surplus in the SI could be informative in illustrating how much P has accumulated or been lost in each area.

Reply: Thank you for this suggestion. In response, we have added figures R1(as Supplementary Figure S4). In figure R1, each panel illustrates the spatial distribution of P fluxes across Europe, normalized per year for the selected time intervals to allow spatial and temporal comparisons. Here, along with the P surplus, we also depict the accumulation of corresponding P-inputs and P-outputs. The first row presents accumulated P surplus, the second row illustrates P inputs, and the third row shows P outputs. The four columns correspond to four distinct periods: 1850–1920, 1921–1960, 1961–1990, and 1991–2019. These periods represent key phases in agricultural and environmental history, reflecting pre-modern agriculture, industrialization before the Green Revolution, the Green Revolution and synthetic fertilizer expansion, and the phase of environmental awareness and policy intervention. The graph illustrates key shifts in P flux dynamics, particularly the significant rise in P inputs after 1960 and the notable gap between inputs and outputs, which has driven the P surplus across Europe.

We also have incorporated this in line 618 under section 3.1 in the revised manuscript which reads as follows:

Furthermore, cumulative P fluxes, including P surplus, inputs, and outputs, are presented for four distinct time periods, which we term as following: (i) 1850–1920 (Pre-modern agriculture), (ii) 1921–1960 (Industrialization before the Green Revolution), (iii) 1961–1990 (Green Revolution and synthetic fertilizer expansion), and (iv) 1991–2019 (Environmental awareness and policy intervention phase) (Supplementary Figure S4). These plots revealed marked shift in P dynamics across Europe over time. During 1850-1920, P surplus was relatively low, averaging 8-10 $t\ yr^{-1}$ in much of the Central and Eastern Europe, with some Western Europe regions like France, the Netherlands, and Denmark exceeding 16 $t\ yr^{-1}$. Northern Europe typically showed much lower values of 2-4 $t\ yr^{-1}$. In the subsequent period (1921–1960), P inputs began to rise modestly, averaging 50-70 $t\ yr^{-1}$, driven by early industrialization and chemical fertilizer use, though P surplus remained moderate due to relatively high P outputs. The

Green Revolution period (1961–1990) saw a sharp increase in P inputs, exceeding 80 $t\ yr^{-1}$ in many regions due to agricultural intensification, resulting in substantial P surplus, with most areas surpassing 18 $t\ yr^{-1}$. In the most recent phase (1991–2019), P inputs declined steadily due to improved agricultural practices and environmental policies like the EU Nitrates Directive, while P outputs increased, narrowing P surplus. In some Western and Eastern Europe, P surplus even turned negative, reflecting P mining. These temporal and spatial trends highlight the importance of sustainable nutrient management practices and policies in reducing P surplus over time. Moving forward, strategies like reallocating nutrients inputs based on regional needs, improving the integration of crop and livestock systems could help to further optimize nutrient use efficiency. Such measures, coupled with continued monitoring of P indicators-P surplus and PUE- are essential to address P-related environmental challenges and promote sustainable agricultural practices (Zou et al., 2022).

1.5 — Figure 4:

– Are there any hypotheses that could explain the peaks in P surplus around 1980 and the subsequent decrease afterward?

Reply: Thank you for your question regarding the peaks in P surplus around 1980 and the subsequent decline. To provide clarity, we have added Figure R2 (corresponding to Supplementary Figure S5), which illustrates along with P surplus their inherent major components, i.e. P inputs from fertilizer and manure and P outputs across various European countries. In general, as shown in this figure, P surplus dynamics closely follow the temporal variability in mineral fertilizer inputs across most countries, with a notable peak around 1980. This peak might be linked to the Green Revolution, a period of agricultural intensification that likely drove a significant increase in synthetic fertilizer use, including phosphorus, to meet growing food demands (Müller-Karulis et al., 2024). The trend in P outputs—i.e., P harvested in crops and grasslands—also shows an increasing trend over time (Supplementary Figure R2), reflecting the need to meet growing crop demand. The interplay between these P inputs and outputs jointly determines the observed behavior in P surplus.

The subsequent decline in P surplus, as shown in Figure R2, could be associated with several factors. In Western Europe, policy changes such as the Nitrates Directive (91/676/EEC) European Commission (2000b), which limits manure application to 170 $kgN\ ha^{-1}$ and consequently restricts P inputs, along with the Water Framework Directive (Directive 2000/60/EC) (European Commission, 2000a) and the recent Farm to Fork Strategy (European Commission) as part of the EU Green Deal (European Commission, 2019) that restricts P inputs (manure and mineral fertilization) (Amery and Schoumans, 2014) may have encouraged more sustainable nutrient management practices, and likely reduced P applications and stabilizing P surplus (Ludemann et al., 2023). Further regional/country-specific legislation also plays a role since a few European countries, including the Netherlands, Ireland, Norway, and Sweden, have specific legislation limiting P applications (Bouraoui et al., 2011). In some cases, the decrease in P surplus began even earlier, as in Denmark and the UK, where P was not a major limiting factor for crop yield since soil P levels had likely reached sufficient levels for crop production without additional inputs (Bouraoui et al., 2011).

In Central and Eastern European countries, the collapse of the Soviet Union in the late 1980s and early 1990s and subsequent (agro-)economic restructuring may have led to a sharp reduction in P inputs, as noticed by the abrupt decline in fertilizer inputs (Figure R2) (Ludemann et al., 2023). This adjustment likely reflects economic changes that reshaped agricultural practices in the region (Csathó et al., 2007; Ludemann et al., 2023). Around this time, awareness was also increasing about the environmental consequences of unsustainable agricultural practices, which may have further motivated efforts to reduce P surplus (Cassou, 2018). For instance, in the late 1980s, the European Union introduced policies that shifted away from direct agricultural subsidies, instead offering incentives aligned with environmental goals (Cassou, 2018).

Overall, the distinct behavior observed across European countries likely reflect a combination of policy, economic pressures, and agricultural adaptations that have collectively influenced P surplus trends across the continent. Such factors are also reflected in a recent study by Zou et al. (2022) at a global scale, which attempts to differentiate and attribute the factors governing the dynamics of long-term P surplus evolution across different countries. Socioeconomic factors such as fertilizer affordability, the intensification of agricultural systems and changes in farm size are among others significant factors governing distinct P evolutions across European countries (Zou et al., 2022).

We would also like to emphasize that our current study is primarily focused on building a database, while in-depth analyses of evolving trends/patterns and underlying reasoning for these trends are left for future studies. Nevertheless, in response to the reviewer's suggestions, we have included brief information on this aspect in line 635-650 under section 3.1 in the revised manuscript:

The peak in P surplus observed around 1980 likely aligns with the intensified fertilizer use of the Green Revolution (Supplementary Figure R2). The subsequent decline in P surplus after 1990 reflects multiple factors, including policy shifts in Western Europe (e.g., Nitrate Directive (Directive 91/676/EEC) (European Commission, 2000b) and Water

Framework Directive (Directive 2000/60/EC) (European Commission, 2000a)), regional legislations that restricted P fertilization (Amery and Schoumans, 2014)), economic adjustments, and increased awareness of sustainable nutrient management (Ludemann et al., 2023; Senthilkumar et al., 2012; Cassou, 2018). Country-specific legislation also played a role, since a few European countries, including the Netherlands, Ireland, Norway, and Sweden, have specific legislation limiting P applications (Bouraoui et al., 2011). In some cases, the decrease in P surplus began even earlier, as in Denmark and the UK, where P was not a major limiting factor for crop yield since soil P levels had likely reached sufficient levels for crop production without additional inputs (Bouraoui et al., 2011). On the other side, in Central and Eastern European regions, the collapse of the Soviet Union and subsequent (agro-)economic restructuring may have led to reduced P inputs, as indicated by a sharp drop in fertilizer use (Csathó et al., 2007; Ludemann et al., 2023) (Supplementary Figure R2) and subsequently reflected in corresponding P surplus budgets. Such distinct P surplus patterns observed across Europe appear to have been shaped by these combined influences, and disentangling the different factors will require careful consideration in future studies. On a global scale, Zou et al. (2022) discussed the distinct roles of socioeconomic and environmental factors governing the dynamics of long-term P surplus evolution across different countries.

1.6 — Section 3.2: In addition, are there any reasons that can explain the varying uncertainty range over time based on your modeling methods? For example, could it be due to changes in crop portfolios over time or differences in data availability? Understanding these reasons could provide insights into improving the modeling in the future. The discussion can be included in a discussion section after the results.

Reply: Thank you for your insightful comments and for highlighting the importance of understanding temporal variability in uncertainty for our P surplus estimates. Indeed, we observe varying level of uncertainty, likely driven by shifts in crop portfolios, data availability, and evolving agricultural management practices (e.g., manure and mineral fertilizer applications) across different periods. To address these points, we conducted an analysis of P surplus uncertainty across various European countries from 1850 to 2019, as detailed below and supported by four supplementary figures.

To ascertain the evolving nature of uncertainty in P surplus over time, we calculated the coefficient of variation (CV, %) as the ratio of the standard deviation to the mean based on 48 P surplus estimates. Figure R3 (Supplementary Figure R3) illustrates CV for P surplus across various European countries, revealing trends in uncertainty that fluctuate across different time periods. In the early years (1850–1930), countries like Germany, France, and the United Kingdom show high CV values, largely due to the low magnitude of P surplus. From 1930 to 1990, there is a notable decrease in CV for most countries, including Germany and the Netherlands, which aligns with advances in agricultural practices during the Green Revolution. By 1990, CV values had stabilized around 20–30% in countries like France and Denmark. In recent decades, however, CV has modestly increased in some countries. For example, since 1990, Italy, Denmark, Slovakia, Slovenia, and some Eastern European countries have shown slight increases in CV.

The uncertainty in our P surplus budget largely stems from the combined effects of differences in crop P content coefficients, manure databases, and application rates of mineral fertilizers and manures in pasture and croplands. These factors vary across both space and time, as reflected by the evolution of different crop types and their productivity, as well as changes in land cover areas over time, i.e., the areas covered by croplands and pastures. Ascertaining the individual contributions of different sources of uncertainty requires careful consideration, including analyses across different European regions and contexts, as well as their evolving nature over time. We have conducted a preliminary investigation to understand the contributing and driving factors, which provides an initial basis for future detailed analyses. To this end, we analyzed the uncertainty ranges (calculated as maximum minus minimum) of key components and examined their relationships with P surplus, as shown in Figures R3- R3). For most countries, differences in the mineral fertilizer inputs show the strongest association with P surplus uncertainty, with relatively high R^2 values across different European Countries. For example, Eastern and Central European countries like Germany, Slovakia, Slovenia, Latvia, and Poland, as well as Mediterranean countries such as Spain, Italy, and Portugal, where R^2 values range between 0.7 and 0.9 (Figure R4, Supplementary Figure R3). This indicates that fluctuations in fertilizer inputs are a significant contributor to P surplus uncertainty over time. On the other side, manure inputs display a more variable relationship with P surplus uncertainty, showing an overall relatively weaker associations than fertilizer in most countries (Figure R5). However, in livestock-intensive countries such as Ireland and the Netherlands, differences in manure inputs emerge as a major contributor to P surplus uncertainty, with R^2 values of 0.71 and 0.95, respectively, compared to 0.56 for association with fertilizer inputs in both countries. Another key contributor to the uncertainty in the P surplus budget is the variation in P outputs, reflected in P crop removal coefficients. This component showed moderate to strong associations with P surplus uncertainty in countries such as Germany, France, Spain, the UK, the Netherlands, and Italy,indicating that P-output variability also substantially impacts P surplus uncertainty, especially in areas with high agricultural productivity (Figure R6).

Our analysis focused on assessing the uncertainty in key factors contributing to P surplus, examining each factor individually. However, we acknowledge that multiple factors often interact in complex ways, influencing P surplus collectively. Addressing these complexities would require more detailed statistical approaches, such as those employed by (Zhang et al., 2021), which we leave for future consideration. To summarize, based on our analysis, we find that fertilizer inputs emerge as the primary driver of P surplus uncertainty. However, P outputs and manure inputs also play significant roles, with their influence varying according to regional agricultural practices. By highlighting key regions and components, our study provides a foundation for future efforts to improve the database and refine the understanding of P surplus dynamics. To reflect these aspects, we have revised the section 3.1 by adding the following texts in line 702-721:

To assess the uncertainty in P surplus estimates, we calculated the coefficient of variation (CV, %), defined as the ratio of the standard deviation to the mean across our 48 P surplus estimates. This analysis, shown in Supplementary Figure S10, offers insights into how relative uncertainty has evolved over time. The CV was highest in the early period (1850–1920) for many countries, including Germany and France and then declined significantly during the mid-20th century (1950–1990). However, in recent decades, relative uncertainty has increased again, especially in countries like Spain and Italy.

In addition, we examined the absolute uncertainty ranges (calculated as maximum minus minimum) of P surplus estimates for each year, comparing these against the ranges of key components, including fertilizer, manure, and P output (Supplementary Figures S11–S13). The results indicate that in central, eastern, and Mediterranean countries such as Germany, Spain, Italy, Slovakia, Slovenia, Poland, and Portugal, fertilizer input uncertainty aligns closely with P surplus uncertainty, identifying fertilizer as potentially a primary driver of variation in these regions (Supplementary Figure S11). In contrast, manure inputs show a more variable relationship with P surplus uncertainty across countries, with generally weaker associations than fertilizer. However, in livestock-intensive regions such as Ireland and the Netherlands, manure uncertainty strongly contributes to P surplus variation (Supplementary Figure S12). For P outputs, associations with P surplus uncertainty are moderate to strong in countries including Germany, France, Spain, the UK, the Netherlands, and Italy, suggesting that output variability also plays a role in P surplus uncertainty, especially in areas with high agricultural productivity (Supplementary Figure S13). Overall, fertilizer inputs emerge as the dominant factor influencing P surplus uncertainty, although the impact of P outputs and manure inputs also varies by region, reflecting distinct agricultural practices. These preliminary findings emphasize the substantial spatial and temporal variability in P surplus uncertainties and underscore the value of ensemble datasets in capturing comprehensive nutrient flows. Further statistical analyses would be required to investigate the factors controlling the uncertainties in P surplus in future studies.

Furthermore, we have included the following texts in line 931 under the section 5 for the future improvement section of the revised manuscript:

Improving the historical land-use and livestock data is essential for refining long-term phosphorus (P) surplus estimates. Currently, the dataset applies uniform land-use data throughout the study period, but incorporating detailed historical land-use reconstructions, such as changes in cropland expansion, pasture reduction, or urbanization, would enhance the spatial accuracy of P surplus calculations. Additionally, livestock and manure data from the mid-20th century introduce uncertainties, particularly in estimating manure production and distribution. A more detailed reconstruction of historical livestock numbers, along with records on manure management systems, would increase reliability. As part of this effort, promoting standardized data collection and reporting methods across European countries would further enhance data accuracy and consistency. Engaging with agricultural practitioners, policymakers, and environmental organizations could help refine data collection methods, ensure alignment with user needs, and expand the dataset's applicability for practical use. Further explorations of contributing parameters in priority areas could also help guide future updates.

We appreciate your valuable feedback, which prompted us to explore the contributing factors to the uncertainty of our P surplus estimates. Although our investigation has been preliminary in nature, we hope it provides insights into the roles of different factors contributing to P surplus uncertainty and highlights regions where future efforts can focus on further improving the presented database.

1.7 — Section 3.2:

– Some of the calculation details could be mentioned in the Methods section.

Reply: Thank you for your feedback. We have significantly revised the manuscript and hope it meets your expectations. Regarding the suggestion to include calculation details in the Methods section, it is unclear which specific calculations is referred to here. Please provide further guidance, and we will be happy to address them in the revised manuscript. However, we have now added explanations about land use and harvested area calculations, which were previously only referred to Batool et al. (2022), to provide more clarity and detail in the Methods section.

1.8 — Lines 645-650:

– The discussion starting here can be included in a new Discussion section. Since your database only includes a limited number of scenarios and your parameter values are borrowed from previous studies, it would be informative to include a brief discussion that lists the key limitations of your database and suggests ways to improve them in the future, instead of simply providing some limitation examples. Other limitations could include, for example, the temporal and spatial variation of parameter values that are not/partially included in the current database and the uncertainty in applying country-level data to a high-resolution map.

Reply: Thank you for your valuable suggestion regarding a more comprehensive discussion and limitations section. We have now added a dedicated *Potential use and limitations of the dataset* section to the revised manuscript (refers to section 4 of the revised manuscript). Therein, we describe key limitations in our approach, such as the temporal and spatial uncertainties before 1961, assumptions related to land-use distributions, fertilizer, and manure applications, and variability in P removal calculations. We also identify and outline potential avenues to improve the dataset in the future (see section 5 of the revised manuscript), including enhancing data collection methods, refining assumptions, and conducting more detailed sensitivity analyses.

1.9 — Technical corrections:

– Abstract & Introduction Line 15: It would be helpful to include a short note defining 'P surplus' (similar to the definitions you provided in Lines 30, 35 and 55) upon its first appearance in the abstract and introduction for readers who are unfamiliar with the term or have different definitions for it.

Reply: Thank you for this comment. We have added the definition of P surplus as the difference between P inputs and P outputs with the term P surplus when it first appears in the Abstract (at line 3) and in Introduction section (at line 19).

– Abstract line 5-10: Does the area in ha refer to the country's total land area or specifically to agricultural land?

Reply:

Thank you for this suggestion. We have updated the sentence in the Abstract to make the units clearer. In particular, we have added that the ha refer to the total physical area at EU27 level. In this context, we have changed the text in the lines 8-10 of the revised manuscript, which now reads as follows:

"Specifically, the total P surplus across the EU-27 has tripled over 170 years, from 1.19 (±0.28) $kg\ ha^{-1}$ of physical area in 1850 to around 2.48 (±0.97) $kg\ ha^{-1}$ of physical area yr^{-1} in recent years".

– Line 35: Provide the full name of FAOSTAT the first time it appears.

Reply: The full name of FAOSTAT has been added as "Food and Agriculture Organization Corporate Statistical Database" when it first appears in line 36 of the revised manuscript.

– Line 55: You may want to move 'phosphorus (P)' to the first time the letter 'P' appears in the manuscript. In addition, there are multiple places in the manuscript where the same abbreviations are explained repeatedly (e.g., 'phosphorus (P)' appears multiple times).

Reply: Thank you for this suggestion. We have revised the manuscript to use "P" repeatedly instead of "phosphorus (P)" wherever appropriate. In some instances, we retain the full structure of "phosphorus (P)", such as in table and figure captions, to emphasize it for quick reference to clarify what "P" stands for.

– Figure 1: I can guess what the arrows in different colors represent, but it would improve clarity if the authors also mention their meanings in the caption.

Reply: Thank you for your comment. We have revised the caption of Figure 1 to clarify the meaning of the colored arrows, which now explicitly correspond to different land use types. Specifically, we added the following line at the end of the caption in the revised manuscript:

"Arrow colors denote land use types: dark green (cropland), orange (pasture), light green (forest), yellow (semi-natural vegetation), brown (urban), and blue (other land uses such as bare rocks and water bodies)".

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List of Figures

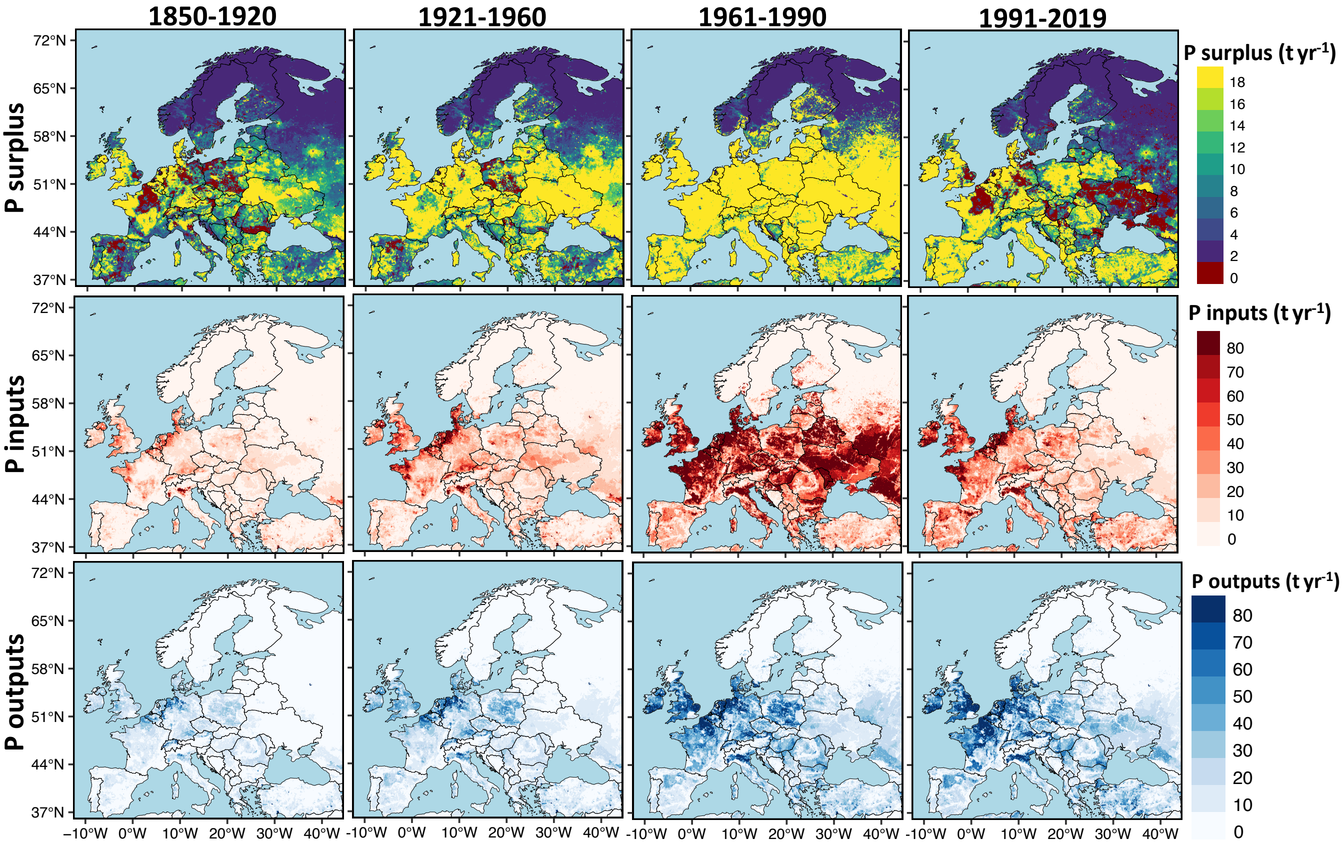

Figure R1. Cumulative total P surplus, P inputs, and P outputs over four historical periods across Europe. The first row shows the accumulated phosphorus (P) surplus, the second row displays P inputs, and the third row illustrates P outputs across Europe for four distinct periods, which we term as following: (i) 1850–1920 (Pre-modern agriculture), (ii) 1921–1960 (Industrialization before the Green Revolution), (iii) 1961–1990 (Green Revolution and expansion of synthetic fertilizers), and (iv) 1991–2019 (Environmental awareness and policy intervention phase). All values are normalized per year within each time period, with units in tonnes per year.

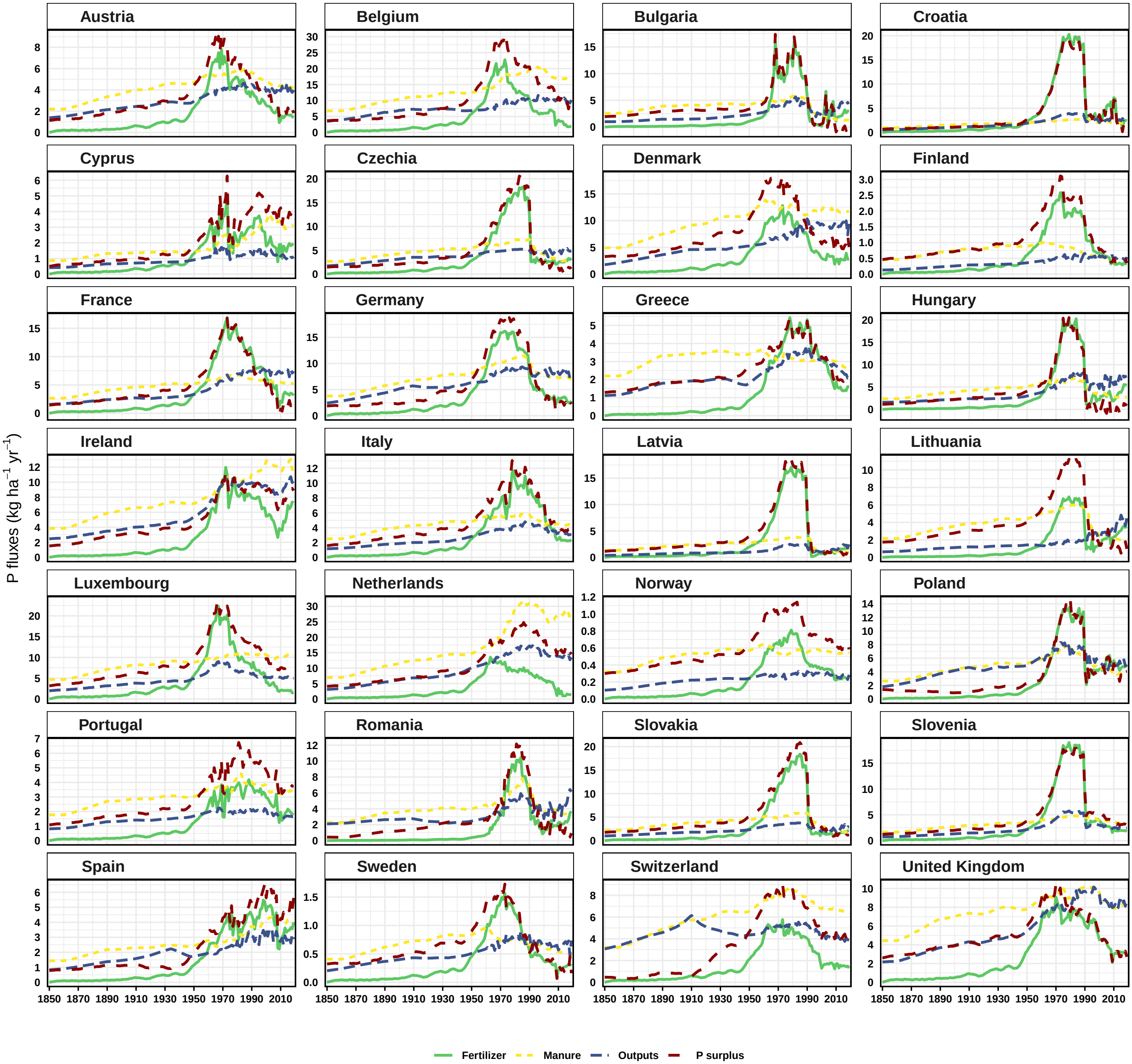

Figure R2. Time series of phosphorus (P) inputs from fertilizer and manure, P outputs, and P surplus ($kg\ ha^{-1}$ of physical area yr^{-1}) across various European countries from 1850 to 2019. This figure highlights changes in P fluxes over time, showing a peak in P surplus around 1980 followed by a decline after 1990 for most countries. These patterns illustrate the influence of agricultural intensification during the Green Revolution, as well as subsequent policy, economic, and environmental shifts in both Western and Eastern Europe. The red line represents the mean of 48 P surplus estimates, while green, yellow, and blue lines depict temporal dynamics for P inputs from fertilizer, manure, and P outputs, respectively. Together, these pattern provide insight into how various factors may have influenced P surplus dynamics over time.

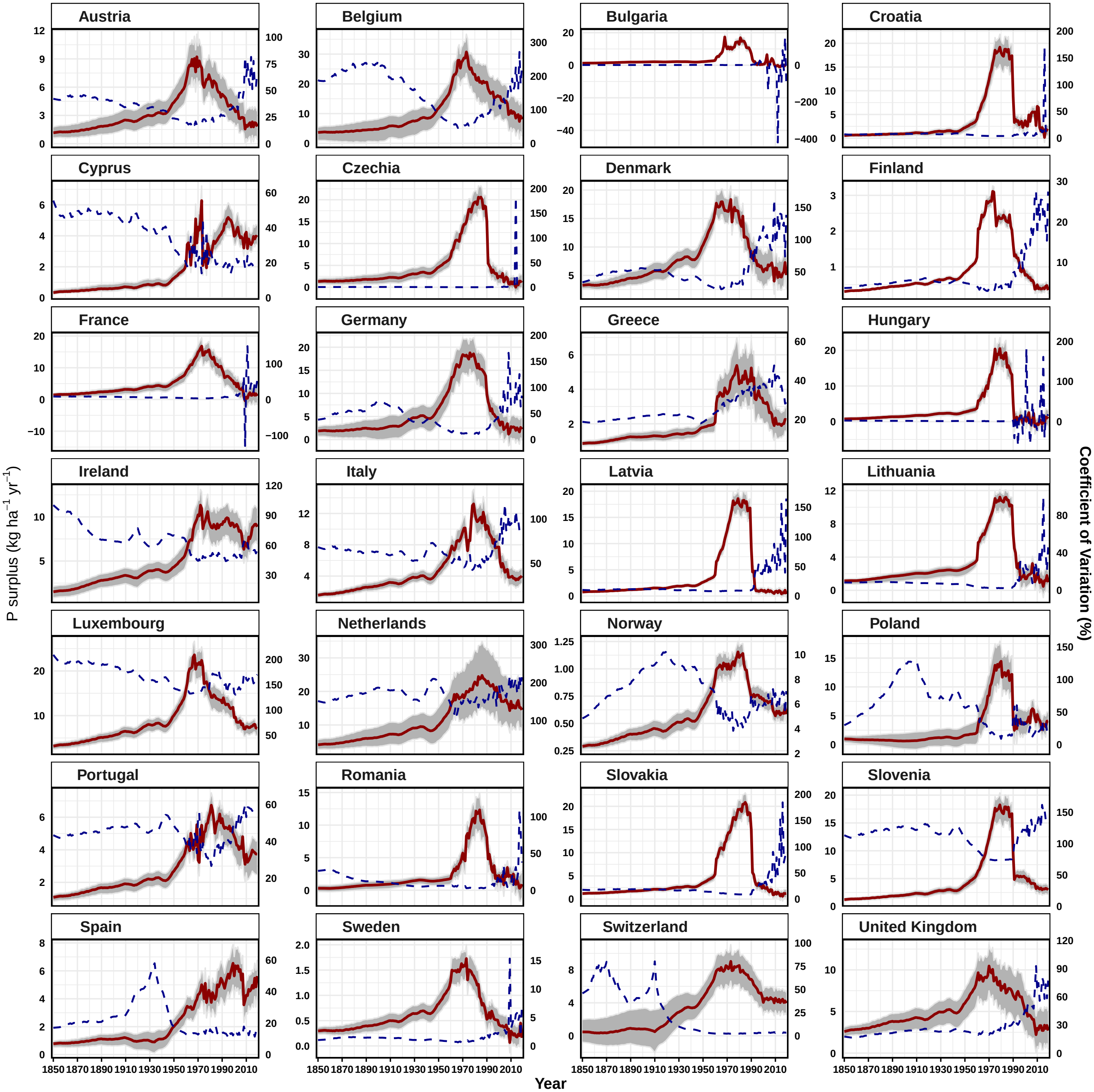

Figure R3. Mean and coefficient of variations (CV, %) of 48 total P surplus estimates across different European countries. The grey ribbon shows the range (min and max) of the 48 P surplus estimates, with the red line representing the average value. The CV is depicted by dashed lines.

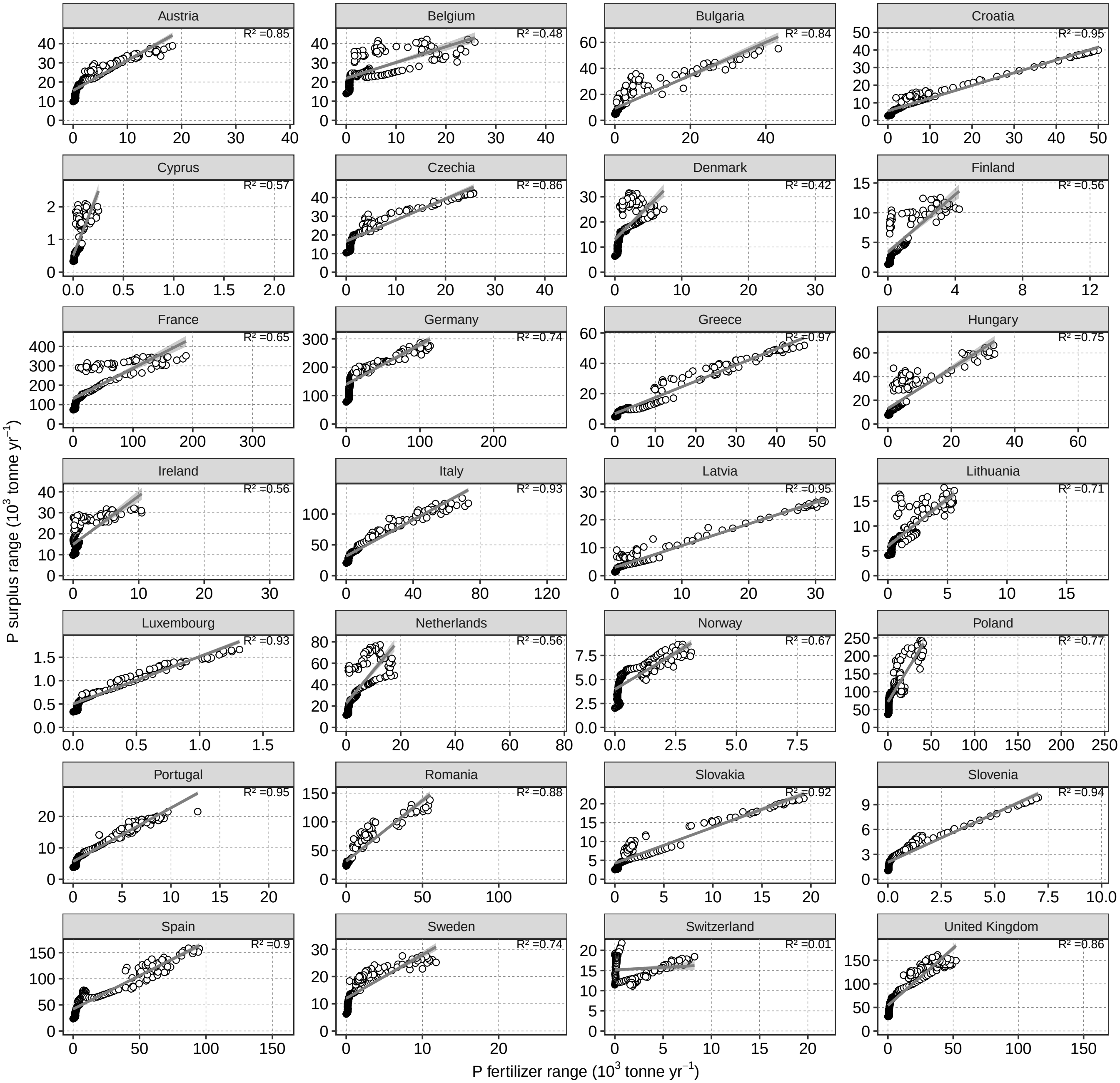

Figure R4. Scatter plot showing the relationship between uncertainty ranges (calculated as the maximum minus minimum) in P fertilizer applied to cropland (x-axis) and total P surplus (y-axis) across different European countries from 1850 to 2019. Each point represents the annual range of fertilizer inputs and corresponding P surplus for a specific country and year. The linear trend line for each country indicates the strength and direction of the association, with a steeper slope suggesting a greater influence of fertilizer input variability on P surplus uncertainty. This plot highlights the variability in P surplus associated with fertilizer input uncertainty across regions and underscores the role of fertilizer as a major contributor to P surplus fluctuations.

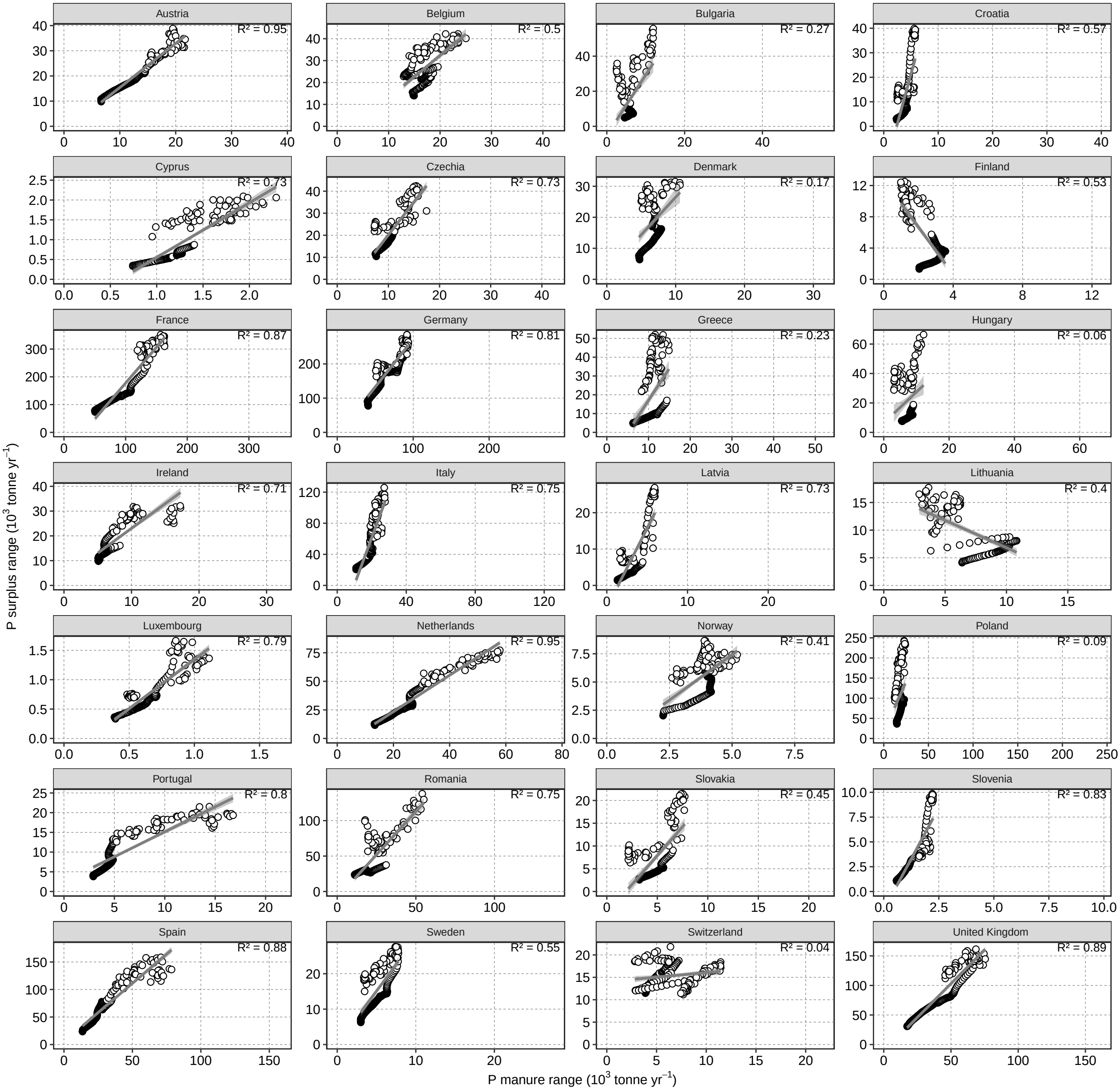

Figure R5. Scatter plot showing the relationship between uncertainty ranges (calculated as the maximum minus minimum) in P manure applied to cropland (x-axis) and total P surplus (y-axis) across different European countries from 1850 to 2019. Each point represents the annual range of manure inputs and corresponding P surplus for a specific country and year. The trend line for each country illustrates the association strength, with a steeper slope indicating a stronger influence of manure input variability on P surplus uncertainty. This plot highlights the variability in P surplus associated with fluctuations in manure inputs, particularly in regions with significant livestock production.

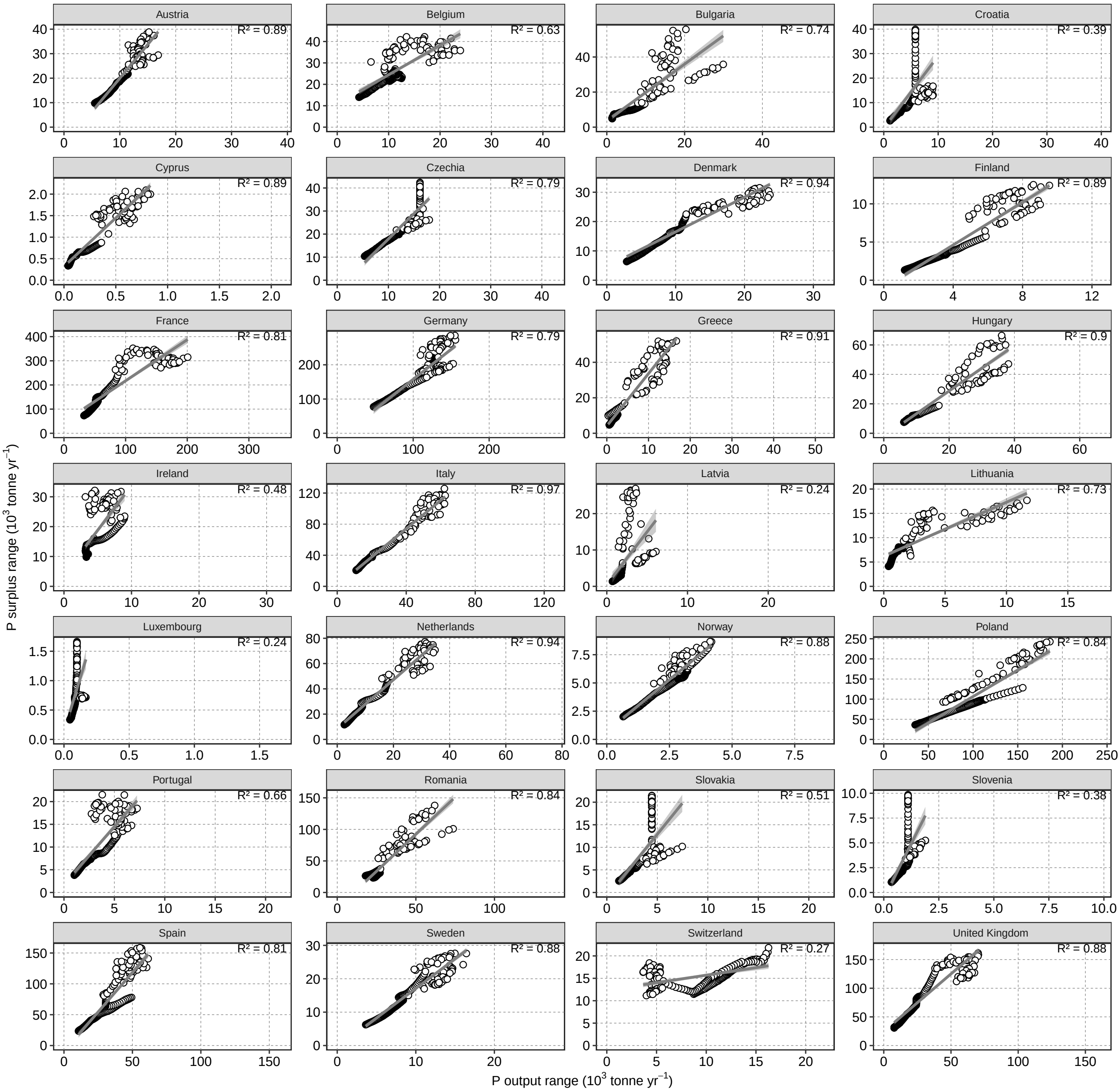

Figure R6. Scatter plot illustrating the relationship between uncertainty ranges calculated as the maximum minus minimum) in P outputs/P removal from cropland (x-axis) and P surplus (y-axis) across various European countries from 1850 to 2019. Each point represents the annual range of P outputs and the corresponding P surplus for a specific country and year. Trend lines are shown for each country, with steeper slopes suggesting a stronger influence of P output variability on P surplus uncertainty. This plot underscores the extent to which changes in P output (e.g., crop removal) contribute to fluctuations in P surplus, particularly in agriculturally intensive regions.

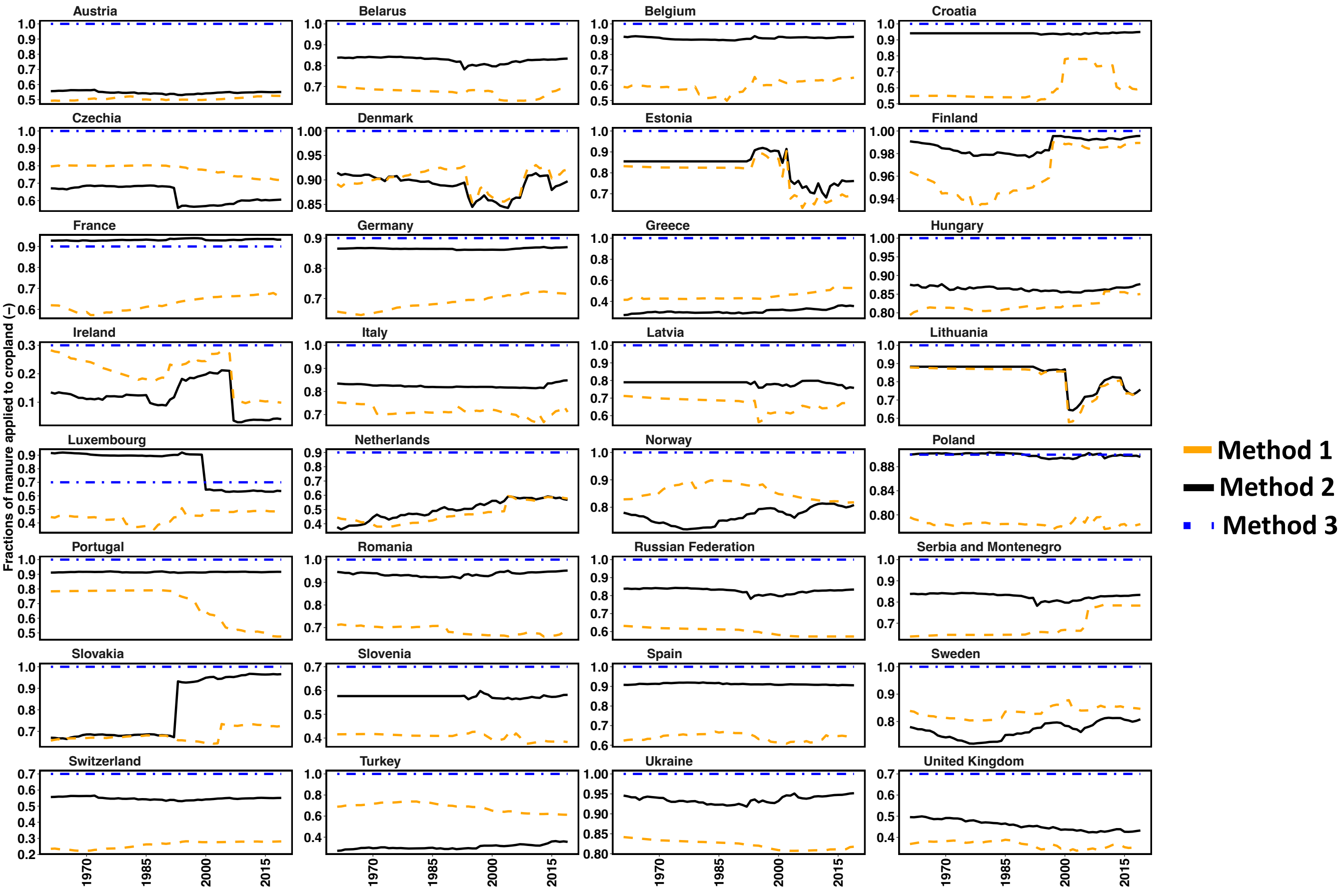

Figure R7. Fractions of manure distribution to cropland based on different methods utilized in this study. Method 1 represents the fraction of distribution of animal manure to cropland based on the equal distribution rates for cropland and pasture within each grid cell. Method 2 shows the fraction using the time-varying national proportions of nitrogen (N) manure applied to both cropland and pasture, as provided by Einarsson et al. (2021). Method 3 shows the manure distribution based on country-level data on manure application proportions to cropland and pasture, as reported by Ludemann et al. (2023).