



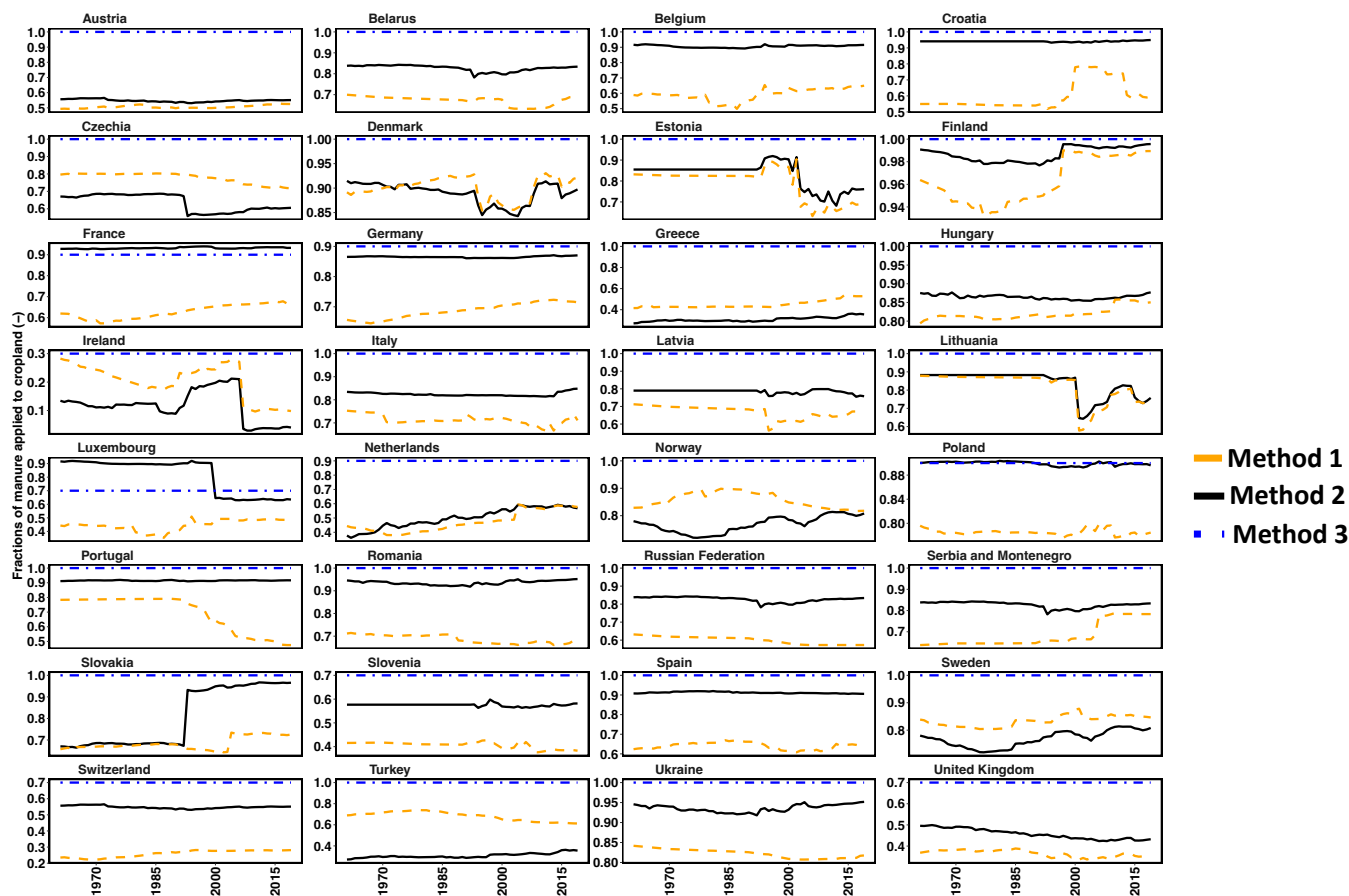
*Supplement of*

## **Century-long reconstruction of gridded phosphorus surplus across Europe (1850–2019)**

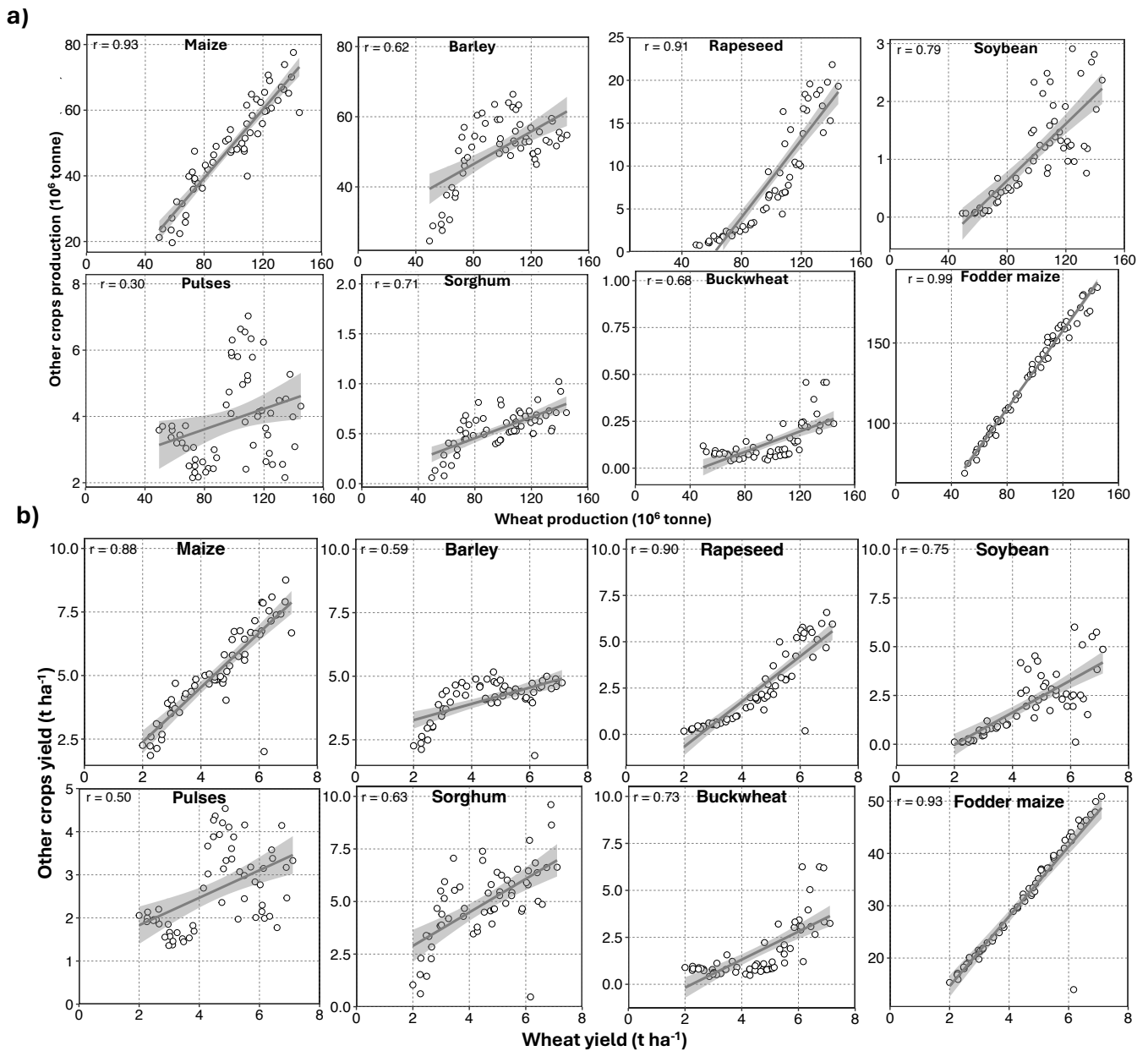
**Masooma Batool et al.**

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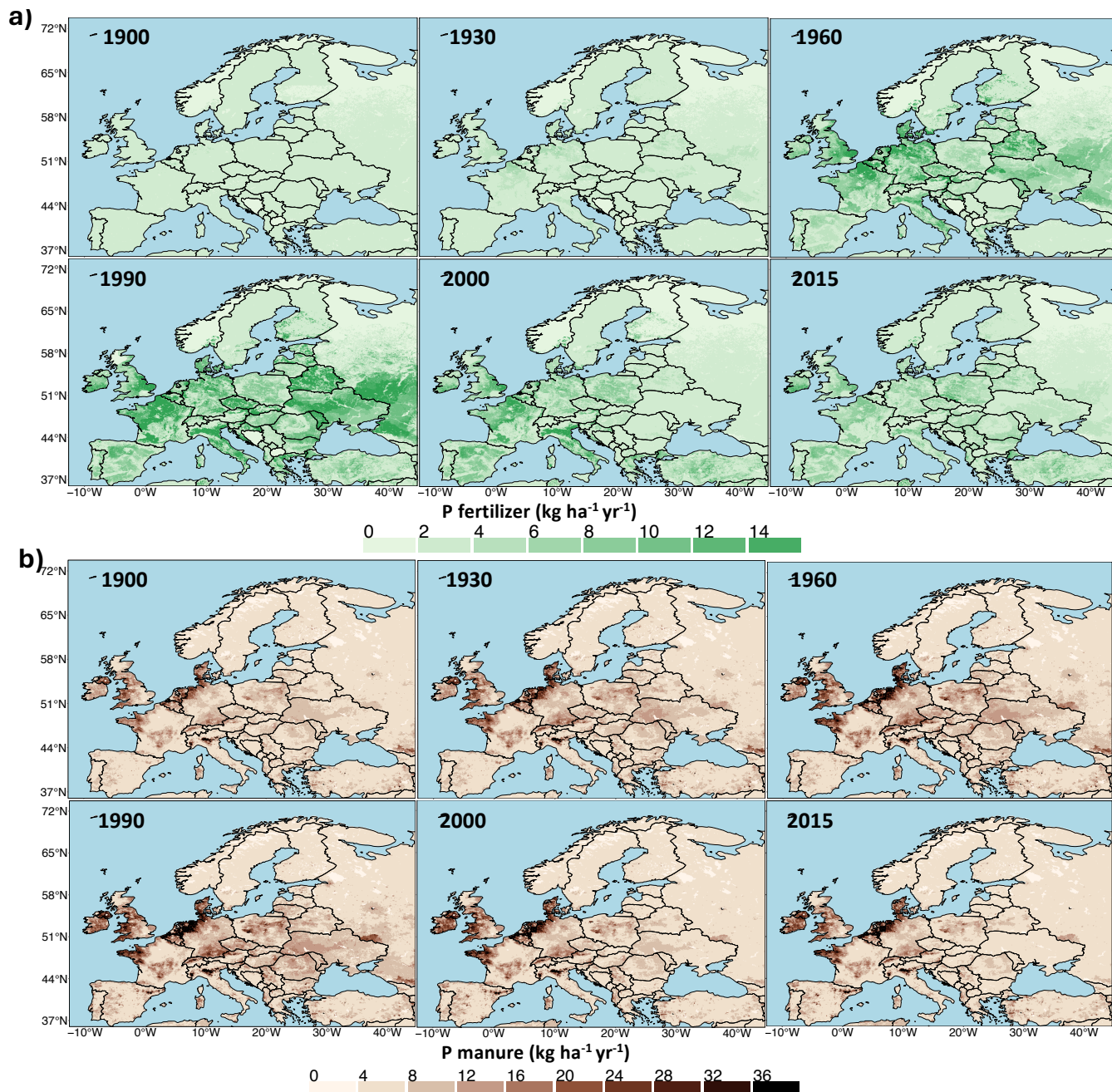
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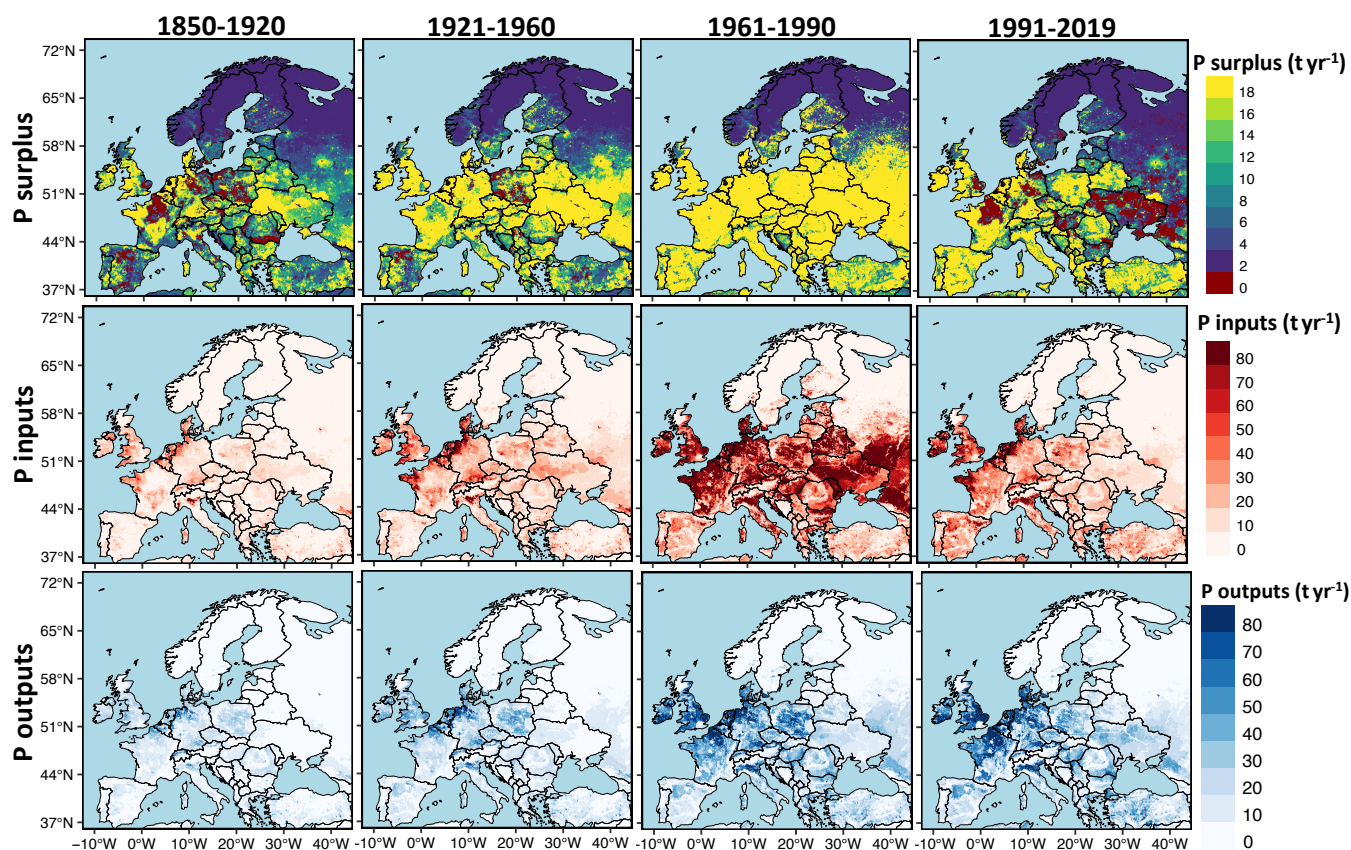
**Figure S1.** Fractions of manure distribution to cropland based on different methods utilized in this study. Method 1 represents the fraction of distribution of animal manure to cropland based on the equal distribution rates for cropland and pasture within each grid cell. Method 2 shows the fraction 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. (2024).



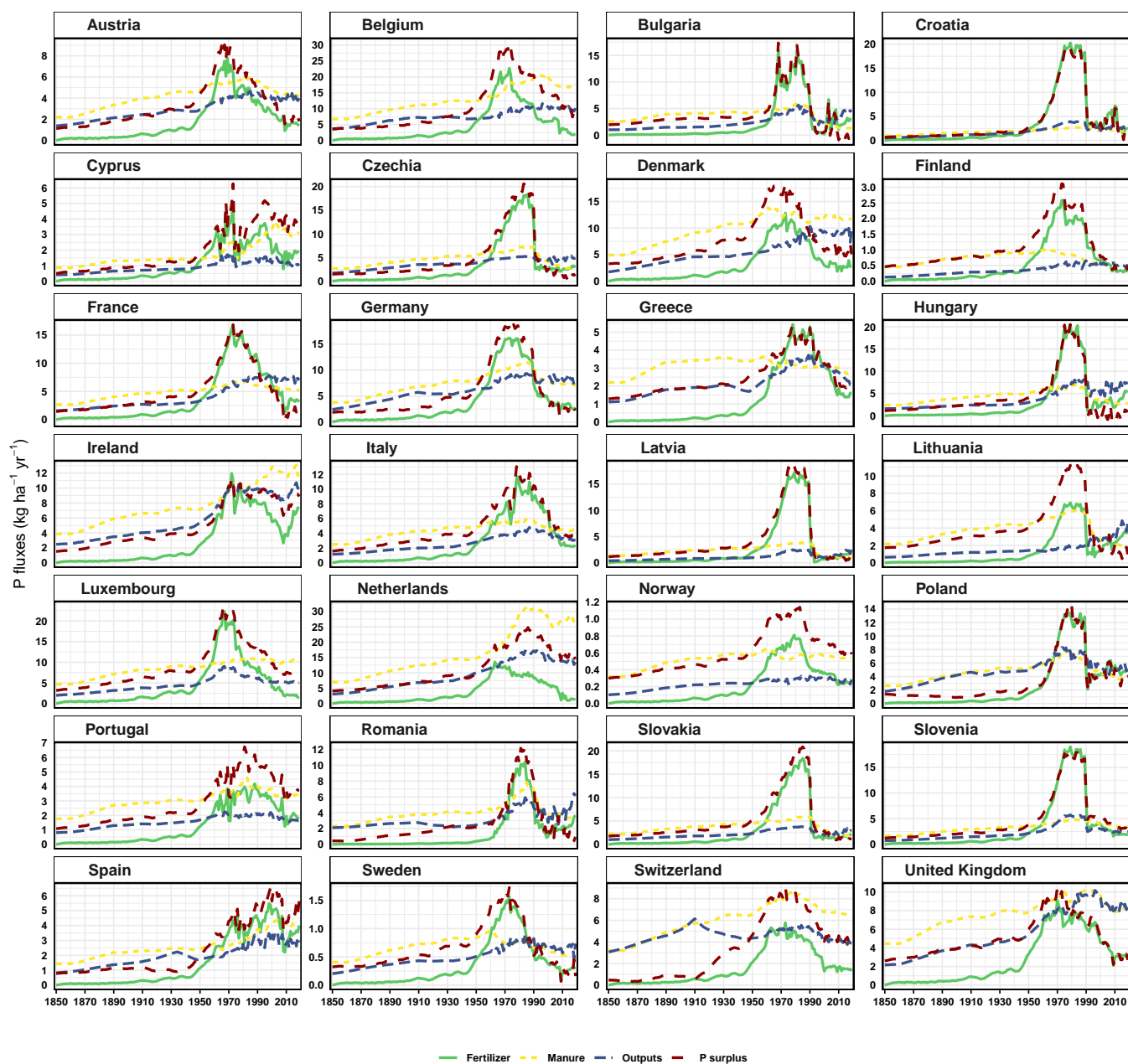
**Figure S2.** Scatter plots illustrating the relationship between wheat production and the production of various crops (panel a, in  $10^6 t$ ) and between wheat yield and the yield of various crops (panel b, in  $t\ ha^{-1}$ ) across the European Union (EU28) from 1961 to 2019. Each circle represents a single year, illustrating the temporal relationship between wheat and other crops. The correlation coefficient ( $r$ ) is annotated in each panel, highlighting the degree of alignment in temporal dynamics. The eight crops shown—maize, barley, rapeseed, soybean, pulses, sorghum, buckwheat, and fodder maize—were selected based on their wide cultivation across the EU28 and/or their relatively high P content.



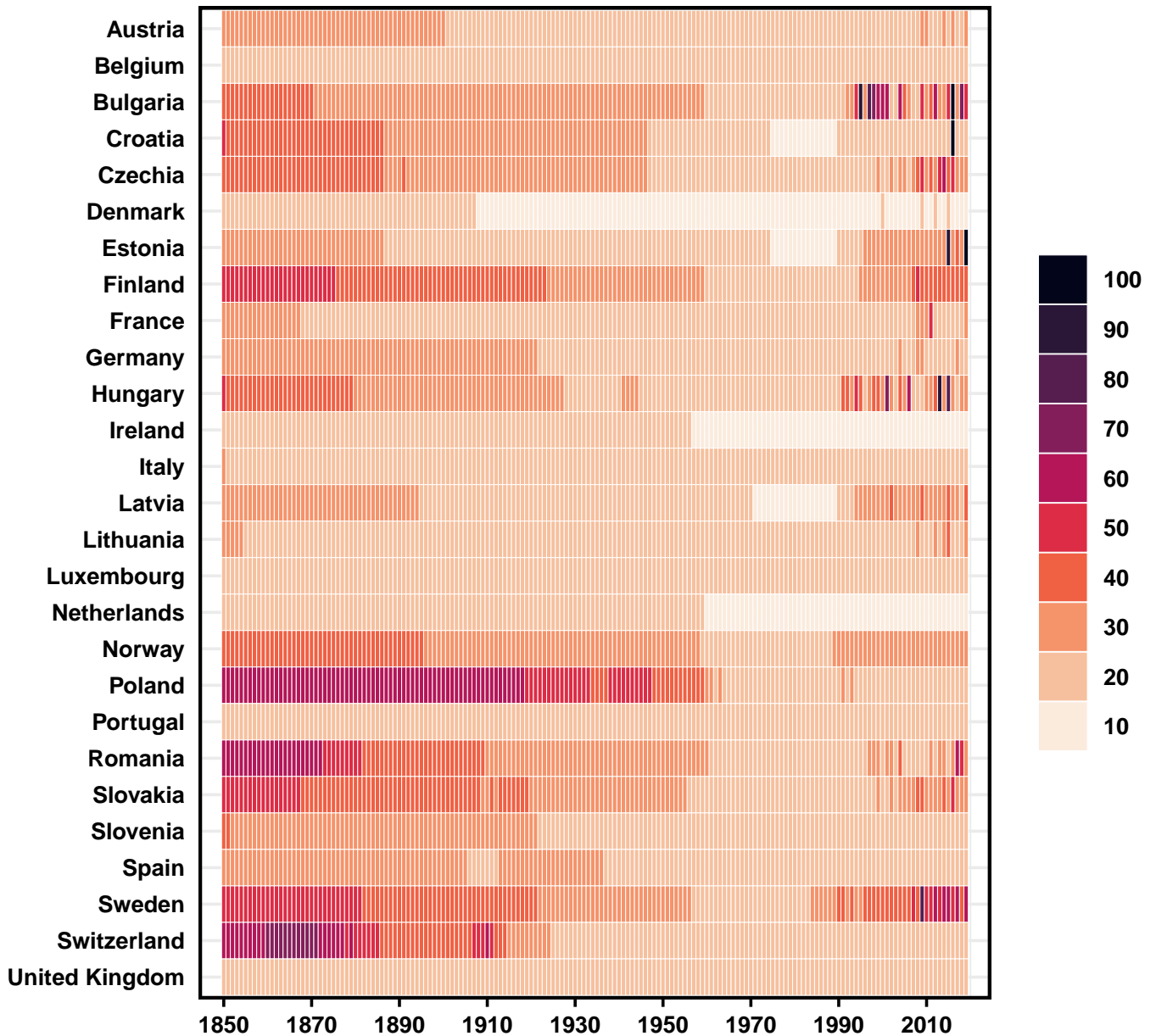
**Figure S3.** Snapshots of P inputs from mineral fertilizer (a) and animal manure (b) ( $\text{kg ha}^{-1}$  of physical area  $\text{yr}^{-1}$ ) across Europe. The figure shows the annual spatial variation in P inputs from mineral fertilizer and animal manure given estimates for the selected years.



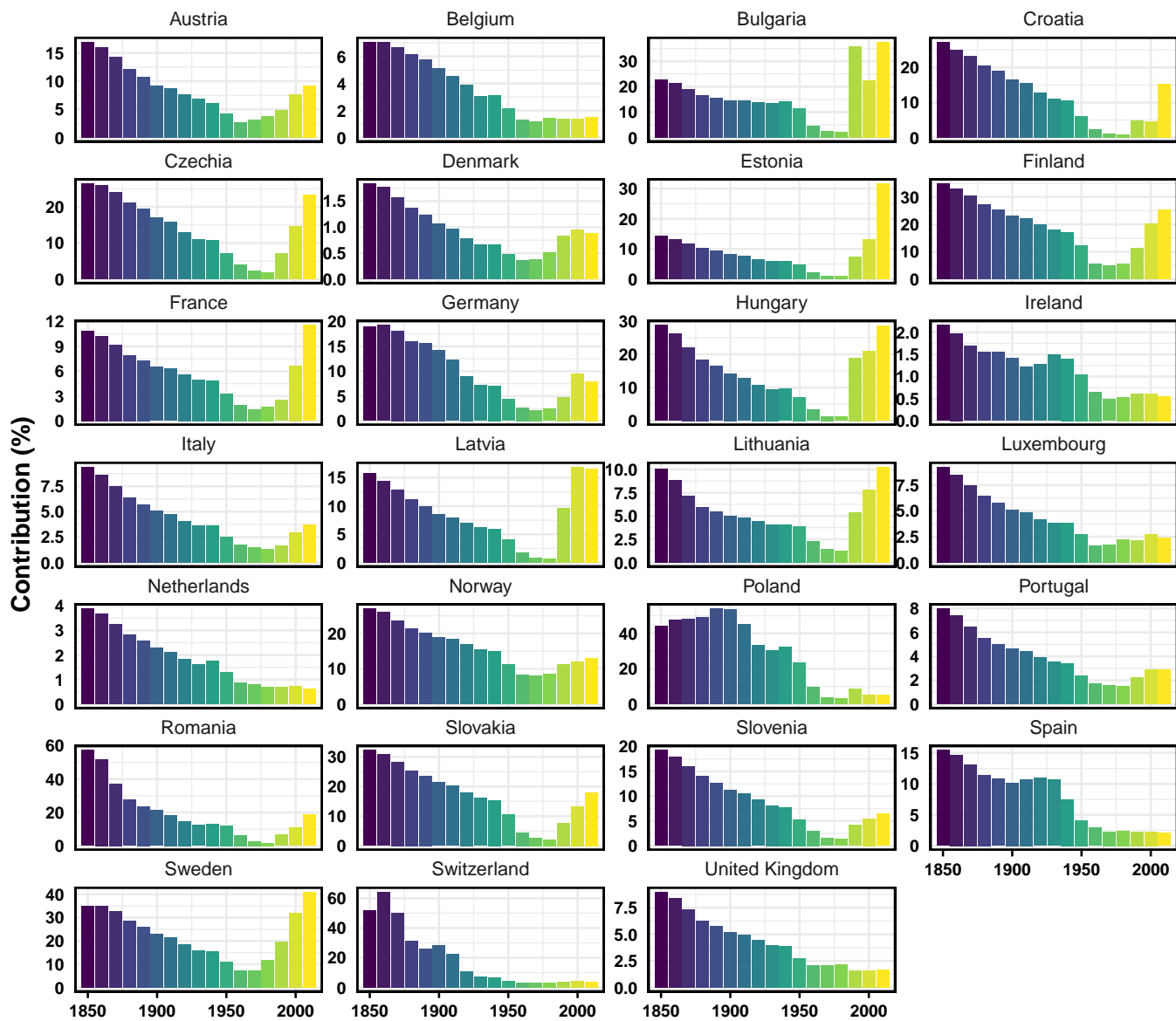
**Figure S4.** Cumulative total P surplus, P inputs, and P outputs over four historical periods across Europe. The first row shows the accumulated phosphorus (P) surplus, the second row displays P inputs, and the third row illustrates P outputs across Europe for four distinct periods, which we term as 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 S5.** Time series of phosphorus (P) inputs from fertilizer and manure, P outputs, and P surplus ( $kg\ ha^{-1}\ yr^{-1}$ ) 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 patterns provide insight into how various factors may have influenced P surplus dynamics over time.

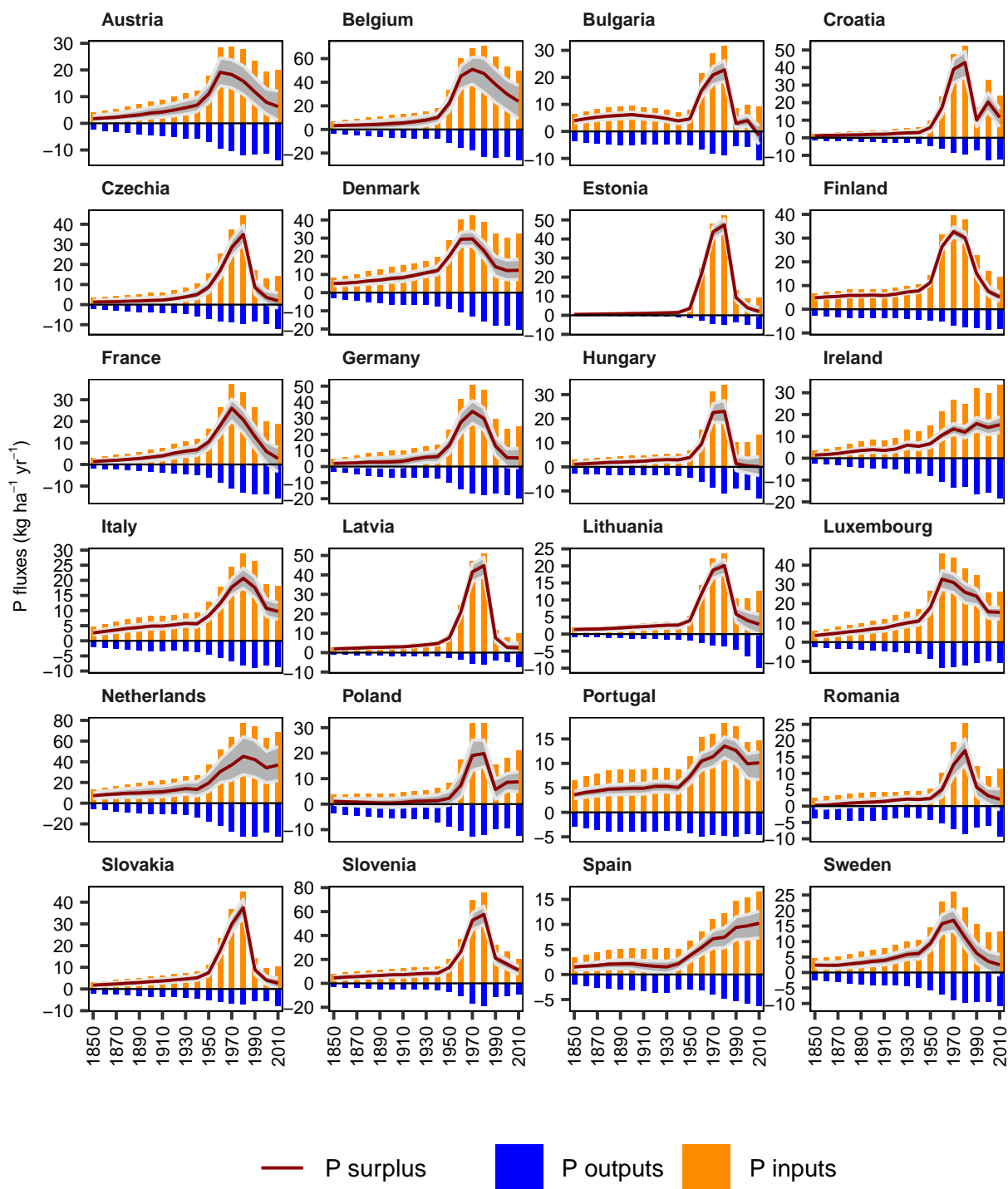


**Figure S6.** Heat map showing the temporal evolution of the contribution (%) of non-agricultural P surplus to the total P surplus across different European countries from 1850-2019. The figure highlights the annual variation in the proportion of non-agricultural P surplus to the total P surplus (averaged from 48 P surplus estimates) across different European countries, illustrating the evolving role of non-agricultural sources in European P dynamics over time.

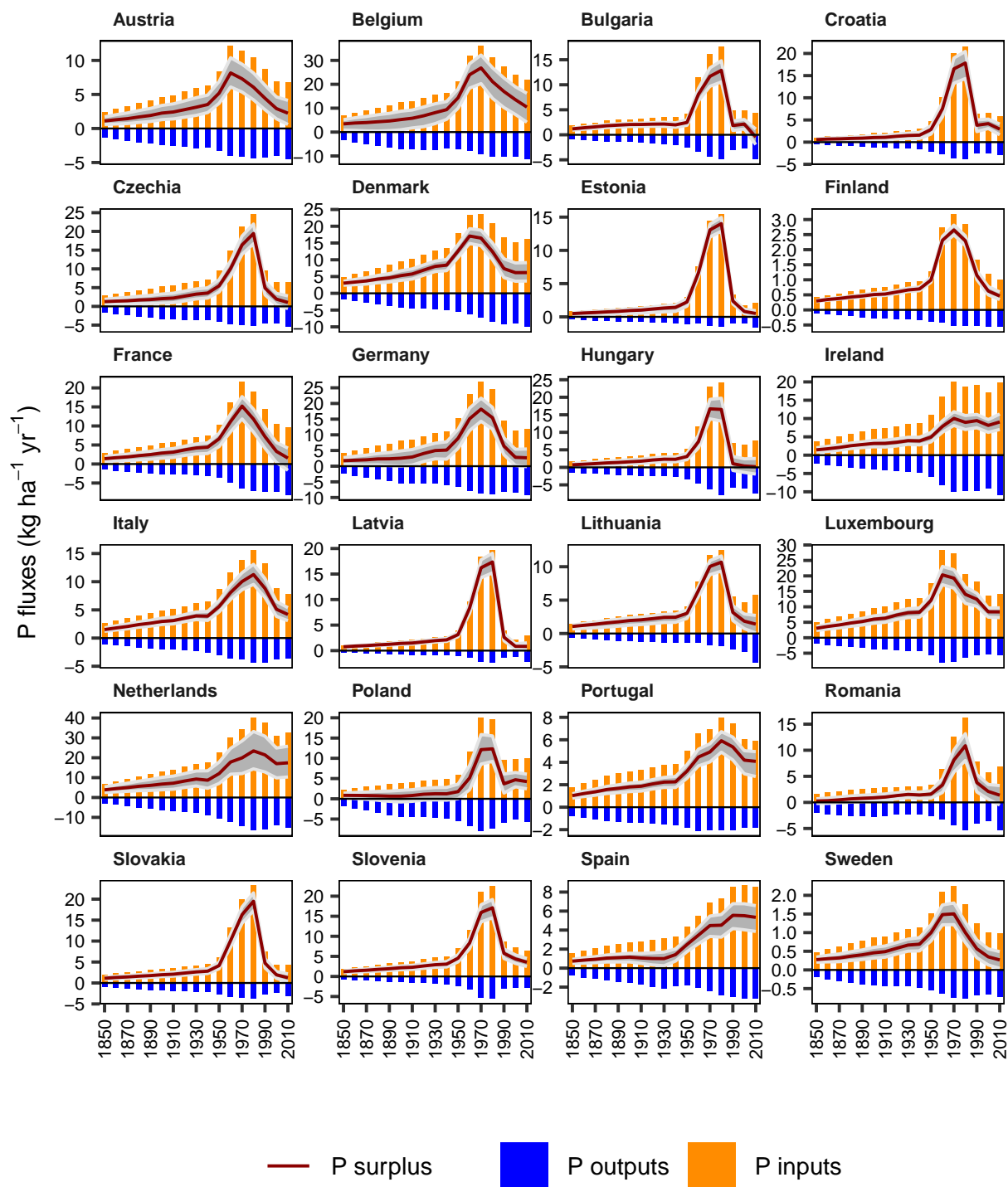


**Figure S7.** Decadal contribution of non-agricultural P surplus to the total P surplus across different European countries.

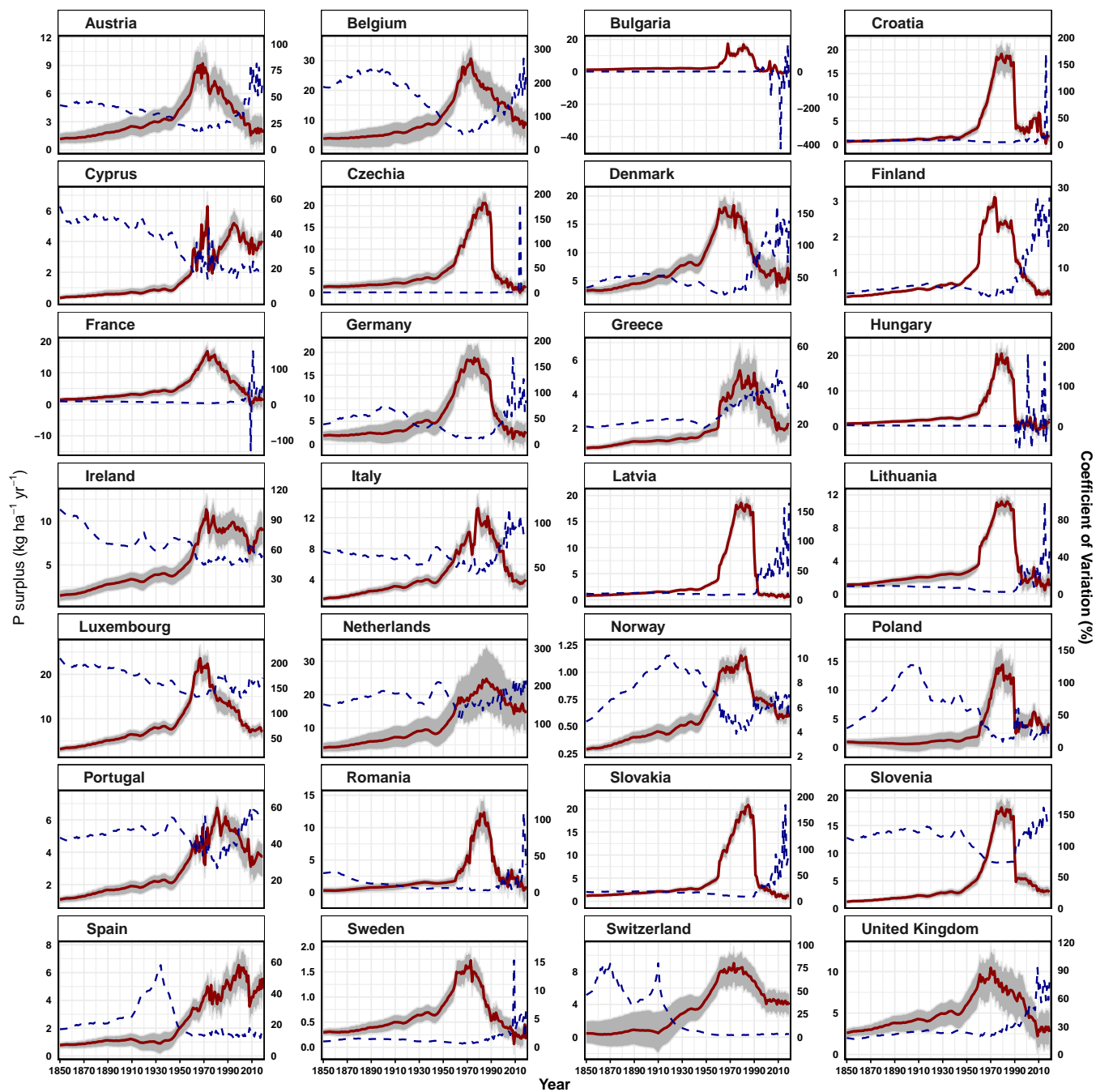




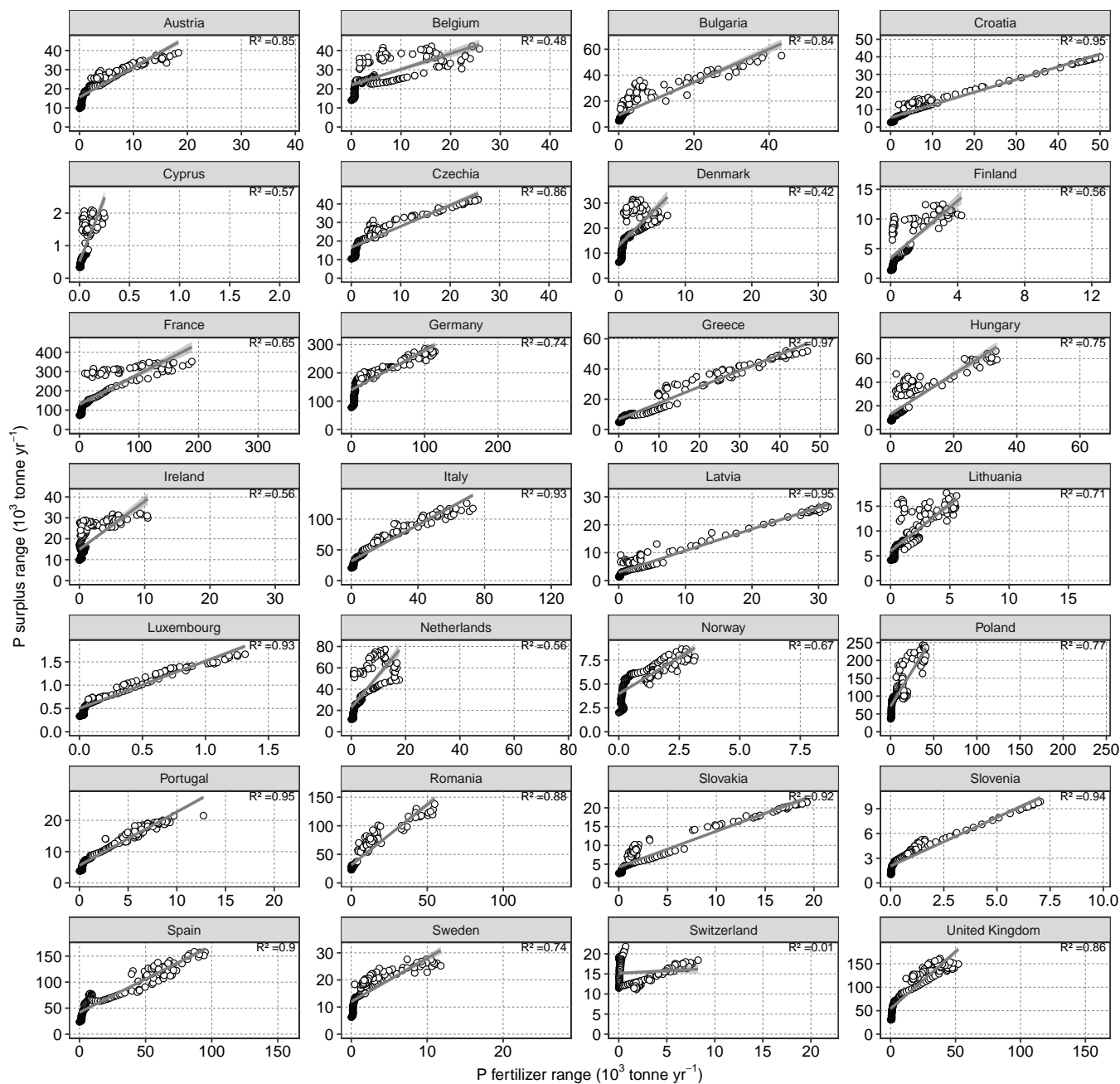
**Figure S8.** Decadal trajectory of agricultural P surplus ( $kg\ ha^{-1}$  of physical area  $yr^{-1}$ ) and its contributing components for different European countries from 1850 to 2019. Upward orange bars represent the average of 48 P inputs, while downward blue bars indicate the average of 48 P outputs, showing decadal means. The grey ribbon shows the range (min and max) of the 48 P surplus estimates, with the red line representing the average value for each decade.



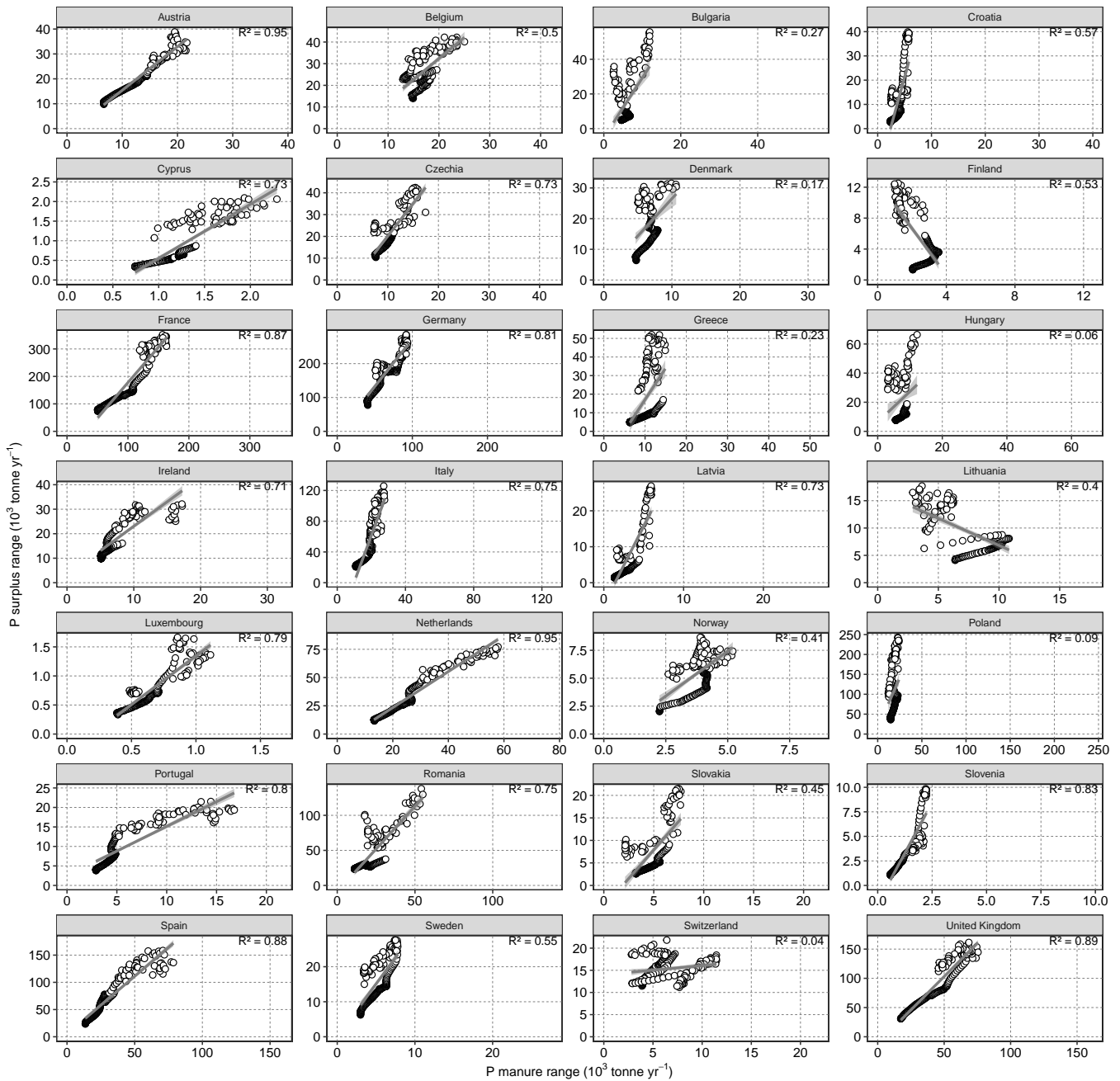
**Figure S9.** Decadal trajectory of total P surplus ( $kg\ ha^{-1}$  of physical area  $yr^{-1}$ ) and its contributing components for different European countries from 1850 to 2019. Upward orange bars represent the average of 48 P inputs, while downward blue bars indicate the average of 48 P outputs, showing decadal means. The grey ribbon shows the range (min and max) of the 48 P surplus estimates, with the red line representing the average value for each decade.



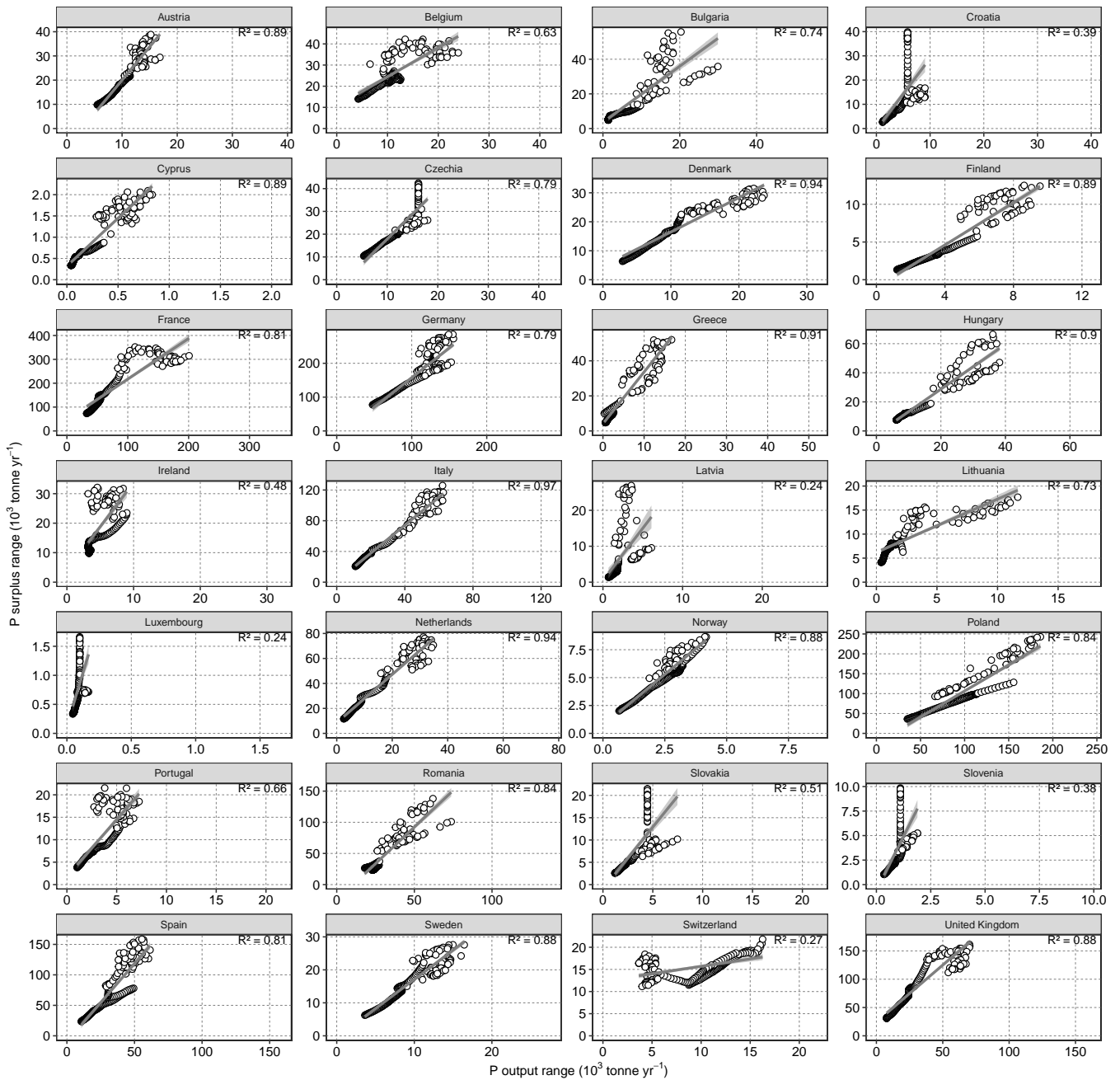
**Figure S10.** Mean and coefficient of variations (CV, %) of 48 total P surplus estimates across different European countries. The grey ribbon shows the range (min and max) of the 48 P surplus estimates, with the red line representing the average value. The CV is depicted by dashed lines.



**Figure S11.** 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 S12.** 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 S13.** 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.

Table S1: 48 Combinations of P surplus

<b>P ID</b>	<b>Surplus</b>	<b>Fertilizer Type</b>	<b>Manure Type</b>	<b>Crop P Removal</b>	<b>Pasture P Removal</b>
1		Fertilizer 1	Manure 1	Crop P Removal 1	Pasture P Removal 1
2		Fertilizer 2	Manure 1	Crop P Removal 1	Pasture P Removal 1
3		Fertilizer 1	Manure 2	Crop P Removal 1	Pasture P Removal 1
4		Fertilizer 2	Manure 2	Crop P Removal 1	Pasture P Removal 1
5		Fertilizer 1	Manure 3	Crop P Removal 1	Pasture P Removal 1
6		Fertilizer 2	Manure 3	Crop P Removal 1	Pasture P Removal 1
7		Fertilizer 1	Manure 1	Crop P Removal 2	Pasture P Removal 1
8		Fertilizer 2	Manure 1	Crop P Removal 2	Pasture P Removal 1
9		Fertilizer 1	Manure 2	Crop P Removal 2	Pasture P Removal 1
10		Fertilizer 2	Manure 2	Crop P Removal 2	Pasture P Removal 1
11		Fertilizer 1	Manure 3	Crop P Removal 2	Pasture P Removal 1
12		Fertilizer 2	Manure 3	Crop P Removal 2	Pasture P Removal 1
13		Fertilizer 1	Manure 4	Crop P Removal 1	Pasture P Removal 1
14		Fertilizer 2	Manure 5	Crop P Removal 1	Pasture P Removal 1
15		Fertilizer 1	Manure 6	Crop P Removal 1	Pasture P Removal 1
16		Fertilizer 2	Manure 4	Crop P Removal 1	Pasture P Removal 1
17		Fertilizer 1	Manure 5	Crop P Removal 1	Pasture P Removal 1
18		Fertilizer 2	Manure 6	Crop P Removal 1	Pasture P Removal 1
19		Fertilizer 1	Manure 4	Crop P Removal 2	Pasture P Removal 1
20		Fertilizer 2	Manure 5	Crop P Removal 2	Pasture P Removal 1
21		Fertilizer 1	Manure 6	Crop P Removal 2	Pasture P Removal 1
22		Fertilizer 2	Manure 4	Crop P Removal 2	Pasture P Removal 1
23		Fertilizer 1	Manure 5	Crop P Removal 2	Pasture P Removal 1
24		Fertilizer 2	Manure 6	Crop P Removal 2	Pasture P Removal 1
25		Fertilizer 1	Manure 1	Crop P Removal 1	Pasture P Removal 2
26		Fertilizer 2	Manure 1	Crop P Removal 1	Pasture P Removal 2
27		Fertilizer 1	Manure 2	Crop P Removal 1	Pasture P Removal 2
28		Fertilizer 2	Manure 2	Crop P Removal 1	Pasture P Removal 2
29		Fertilizer 1	Manure 3	Crop P Removal 1	Pasture P Removal 2
30		Fertilizer 2	Manure 3	Crop P Removal 1	Pasture P Removal 2
31		Fertilizer 1	Manure 1	Crop P Removal 2	Pasture P Removal 2
32		Fertilizer 2	Manure 1	Crop P Removal 2	Pasture P Removal 2
33		Fertilizer 1	Manure 2	Crop P Removal 2	Pasture P Removal 2
34		Fertilizer 2	Manure 2	Crop P Removal 2	Pasture P Removal 2
35		Fertilizer 1	Manure 3	Crop P Removal 2	Pasture P Removal 2
36		Fertilizer 2	Manure 3	Crop P Removal 2	Pasture P Removal 2
37		Fertilizer 1	Manure 4	Crop P Removal 1	Pasture P Removal 2
38		Fertilizer 2	Manure 5	Crop P Removal 1	Pasture P Removal 2
39		Fertilizer 1	Manure 6	Crop P Removal 1	Pasture P Removal 2
40		Fertilizer 2	Manure 4	Crop P Removal 1	Pasture P Removal 2
41		Fertilizer 1	Manure 5	Crop P Removal 1	Pasture P Removal 2
42		Fertilizer 2	Manure 6	Crop P Removal 1	Pasture P Removal 2
43		Fertilizer 1	Manure 4	Crop P Removal 2	Pasture P Removal 2
44		Fertilizer 2	Manure 5	Crop P Removal 2	Pasture P Removal 2
45		Fertilizer 1	Manure 6	Crop P Removal 2	Pasture P Removal 2
46		Fertilizer 2	Manure 4	Crop P Removal 2	Pasture P Removal 2
47		Fertilizer 1	Manure 5	Crop P Removal 2	Pasture P Removal 2
48		Fertilizer 2	Manure 6	Crop P Removal 2	Pasture P Removal 2

**Table S2.** Descriptions of Methods for Fertilizer, Manure, Crop P Removal, and Pasture P Removal

<b>Component</b>	<b>Method Description</b>
<b>Fertilizer Type</b>	<b>Fertilizer 1:</b> Distributed based on cropland and pasture area fractions within each grid cell, following Xu et al. (2019).
	<b>Fertilizer 2:</b> Distributed based on country-level data on fertilizer application proportions to cropland and pasture, as reported by Ludemann et al. (2024).
<b>Manure Type</b>	<b>Manure 1:</b> FAOSTAT-based, distributed based on cropland and pasture area fractions within each grid cell, following Xu et al. (2019).
	<b>Manure 2:</b> FAOSTAT-based, distributed based on country-level data on manure application proportions to cropland and pasture, as reported by Ludemann et al. (2024).
	<b>Manure 3:</b> FAOSTAT-based, distributed based on time-varying national proportions of nitrogen (N) manure applied to cropland and pasture, as provided by Einarsson et al. (2021).
	<b>Manure 4:</b> Einarsson et al. (2021)-based, distributed based on cropland and pasture area fractions within each grid cell, following Xu et al. (2019).
	<b>Manure 5:</b> Einarsson et al. (2021)-based, distributed based on country-level data on manure application proportions to cropland and pasture, as reported by Ludemann et al. (2024).
	<b>Manure 6:</b> Einarsson et al. (2021)-based, distributed based on time-varying national proportions of nitrogen (N) manure applied to cropland and pasture, as provided by Einarsson et al. (2021).
<b>Crop P Removal</b>	<b>Crop P Removal 1:</b> Minimum P removal from crop content.
	<b>Crop P Removal 2:</b> Maximum P removal from crop content.
<b>Pasture P Removal</b>	<b>Pasture P Removal 1:</b> P removal rate of 0.6.
	<b>Pasture P Removal 2:</b> Regional-specific P removal rates (0.4 for Eastern EU, 0.5 for Western EU).



## References

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- Xu, R., Tian, H., Pan, S., Dangal, S. R., Chen, J., Chang, J., Lu, Y., Skiba, U. M., Tubiello, F. N., and Zhang, B.: Increased nitrogen enrichment and shifted patterns in the world's grassland: 1860–2016, *Earth System Science Data*, 11, 175–187, 2019.