Supplements for Gridded dataset of nitrogen and phosphorus point sources from wastewater in Germany (1950–2019)

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S1 NUTS-1 regions of Germany

Table S1 provides a list of the 16 regions at Nomenclature of Territorial Units for Statistics 1 (NUTS-1) level in Germany, including information on whether the regions belonged to the former West or East Germany. The NUTS-1 regions can be visualised in Figure S1. The NUTS-1 regions correspond to the federal states of Germany.

Table S1. The 16 Nomenclature of Territorial Units for Statistics level 1 (NUTS-1) regions (federal states) of Germany, including information on whether the regions belonged to the former West or East Germany.

NUTS-1 region name	Abbreviations used in this study	Former West or East Germany
Schleswig-Holstein	SH	West
Hamburg	HH	West
Lower Saxony	LS	West
Bremen	HB	West
North Rhine-Westphalia	NW	West
Hesse	HE	West
Rhineland-Palatinate	RP	West
Baden-Württemberg	BW	West
Bavaria	BV	West
Saarland	SL	West
Berlin	BE	West and East
Brandenburg	BB	East
Mecklenburg-Vorpommern	MV	East
Saxony	SN	East
Saxony-Anhalt	ST	East
Thuringia	TH	East

Note: the spatial location of the NUTS-1 regions can be visualised in Figure S1

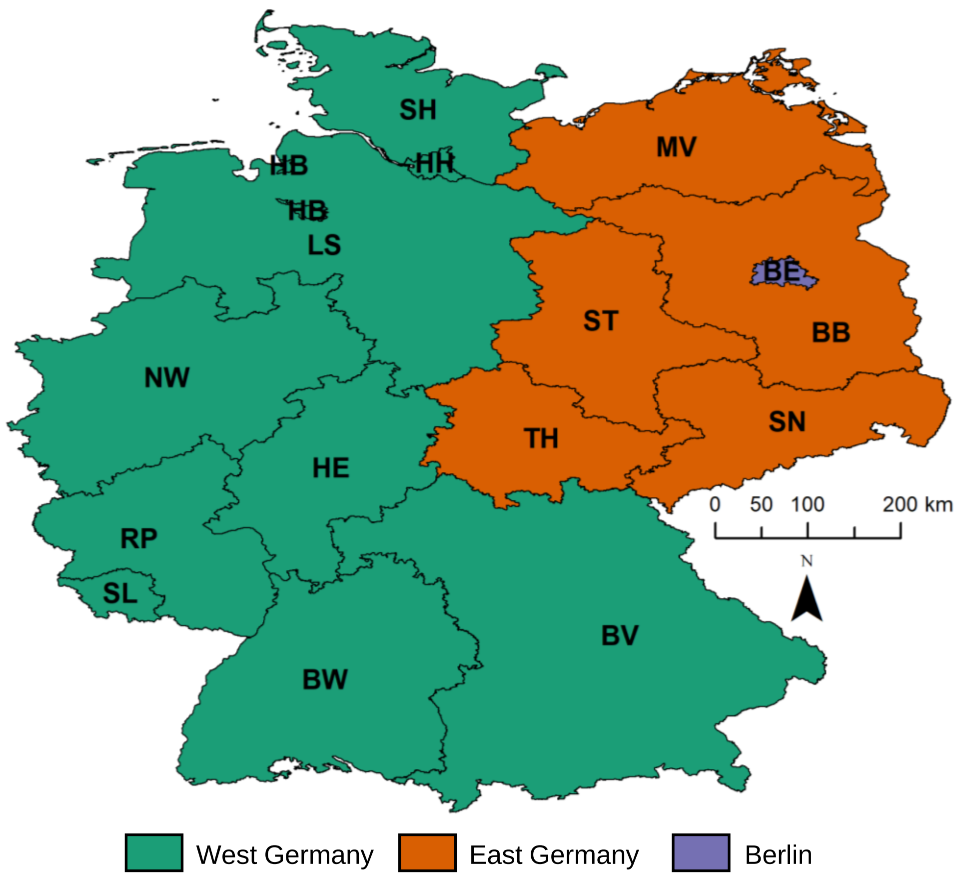


Figure S1. Map of the 16 Nomenclature of Territorial Units for Statistics level 1 (NUTS-1) regions (federal states) of Germany. Data source: the NUTS-1 map comes from the German Federal Agency for Cartography and Geodesy (BKG, 2020) ©GeoBasis-DE/BKG dl-de/by-2-0 license (https://www.govdata.de/dl-de/by-2-0). The map is shown in the WGS84 system in the figure.

S2 Other fates of N and P gross emissions to wastewater beyond point sources

This section describes the N and P gross emissions to wastewater that follow other pathways than point sources to surface water that can visualized in Figure 1 in the main text. Since the equations to calculate the N and P point sources are the same, we denote as $Nutri$ any of the two nutrients N and P. We also use the notation X to refer to the total ($X = all$), urban ($X = urb$) and rural ($X = rur$) components of the emissions.

First, part of the N and P gross domestic emissions are not collected in the sewer system nor treated in wastewater treatment plants (WWTPs). We estimate these emissions at time *t* and for the i-th NUTS1 region, that we denote as $J^{Nutri}_{noSewerWwtp,X}(t,i)$ ($kg\ yr^{-1}$), as follows:

$$J^{Nutri}_{noSewerWwtp,X}(t,i) = J^{Nutri}_{gross,dom,X}(t,i)(1 - T_{sewer-wwtp,X}(t,i)) \tag{S1}$$

where $J^{Nutri}_{gross,dom,X}$ ($kg\ yr^{-1}$) are the gross domestic emissions of N ($Nutri = N$) and P ($Nutri = P$), and $T_{sewer-wwtp,X}(t,i)$ ($-$) is the fraction of population connected to the sewer system or to WWTPs (defined in Eq. 9 in the main text), which includes the transportation of wastewater from cesspits to WWTPs. We do not track the fate of the emissions of Equation S1 that could be a diffuse source or a point source to surface water depending on the wastewater handling method used, as documented for example in MUGV (2010) for the contemporary period in Germany.

Second, part of the emissions that are collected in the sewer system or treated in WWTPs are lost during wastewater collection and transport. This includes losses in the sewer system (Morée et al., 2013), as well as losses during transportation of the wastewater collected in cesspits to WWTPs. These emissions are calculated differently for the urban and rural components, since industrial emissions occur in urban areas only, and are expressed as follows:

$$\begin{aligned} J^{Nutri}_{transportLoss,urb}(t,i) &= f^{N,P}_{loss,transport}(J^{Nutri}_{gross,dom,urb}(t,i)T_{sewer-wwtp,urb}(t,i) + J^{Nutri}_{gross,ind,urb}(t,i)) \\ J^{Nutri}_{transportLoss,rur}(t,i) &= f^{N,P}_{loss,transport}J^{Nutri}_{gross,dom,rur}(t,i)T_{sewer-wwtp,rur}(t,i) \end{aligned} \tag{S2}$$

where $J^{Nutri}_{transportLoss,X}(t,i)$ ($kg\ yr^{-1}$) are the N and P emissions lost during wastewater collection and transport, respectively, $J^{Nutri}_{gross,dom,X}$ ($kg\ yr^{-1}$) and $J^{Nutri}_{gross,ind,X}$ ($kg\ yr^{-1}$) are the N and P gross domestic and industrial/commercial emissions, respectively, and $f^{N,P}_{loss,transport}$ ($-$) is the fraction of N and P emissions lost during wastewater collection and transport. In Eq S2, we use the fraction of population connected to the sewer system or to WWTPs ($T_{sewer-wwtp,X}$) for the domestic emissions only, while all industrial/commercial emissions are collected in the sewer system or treated in WWTPs. We do not track the fate of the emissions of Eq. S2 that could in particular be leaked to soils, undergo biological degradation, settle in sewers, or volatilise, as reported in Morée et al. (2013).

Third, part of the emissions that are collected in the sewer system can be applied to agricultural soils in sewage farms and are thus a diffuse source to surface water. In our study, this only regards the state of Berlin, for which sewage farms are documented for instance in Lottermoser (2012). These emissions are calculated differently for the urban and rural components, since industrial emissions occur in the urban component only, and are expressed as follows:

$$J^{Nutri}_{farm,urb}(t,i) = \left(1 - f^{N,P}_{loss,transport}\right)\left(J^{Nutri}_{gross,dom,rur}(t,i)T_{farm,urb}(t,i) + J^{Nutri}_{gross,ind}(t,i)\frac{T_{farm,urb}(t,i)}{T_{sewer-wwtp,urb}(t,i)}\right)$$
$$J^{Nutri}_{farm,rur}(t,i) = \left(1 - f^{N,P}_{loss,transport}\right)J^{Nutri}_{gross,dom,rur}(t,i)T_{farm,rur}(t,i) \quad \text{(S3)}$$

where $J^{Nutri}_{farm,X}(t,i)$ $(kg\ yr^{-1})$ are the N and P emissions applied to agricultural soils in sewage farms, and $T_{farm,X}(t,i)$ $(-)$ is the fraction of population connected to sewage farms. In Eq. S3 the fate of the industrial/commercial gross emissions is assessed by normalizing the population connection to sewage farms by $T_{sewer-wwtp,urb}(t,i)$, which ensures that all industrial/commercial gross emissions are collected in the sewer system or treated in WWTPs. In this study, we do not track the possible retention, degradation and transport of the emissions of Eq. S3 from soils to surface waters.

Fourth, through treatment in WWTPs, part of the N and P emissions are removed from the wastewater according to the efficiency of N and P removal. We calculate these emissions, that we denote as $J^{Nutri}_{wwtpRem,X}(t,i)$ $(kg\ yr^{-1})$, as follows:

$$J^{Nutri}_{wwtpRem,X}(t,i) = J^{Nutri}_{wwtpIn,X}(t,i) - J^{Nutri}_{ps,wwtpOut,X}(t,i) \quad \text{(S4)}$$

where $J^{Nutri}_{wwtpIn,X}(t,i)$ $(kg\ yr^{-1})$ are the WWTP incoming N and P emissions, and $J^{Nutri}_{ps,wwtpOut,X}(t,i)$ $(kg\ yr^{-1})$ are the WWTP outgoing N and P emissions. We do not track the fate of the emissions of Eq. S4 which could in particular be lost to the atmosphere through gaseous forms or end up as sewage sludge that can for instance be applied as agriculture fertilizer or be combusted for energy purposes (Roskosch and Heidecke, 2018).

S3 Determination of the ranges of the parameters of N and P point sources model

Table S2 describes the parameters of the N and P point sources model and the ranges used in this study and it provides a summary of the methodology used to determine the parameter ranges. The determination of specific parameters are detailed in the following subsection, namely the fraction of protein supply wasted at the distribution and consumption level (f_{waste}^{pro}; Sect. S3.1), the efficiencies of N and P removal for the different types of wastewater treatment (eff_1^N, eff_1^P, $eff_{23^{noN}}^N$, $eff_{23^{noP}}^P$, $eff_{3^N}^N$, and $eff_{3^P}^P$; Sect. S3.2), the fractions of industrial/commercial to domestic N and P emissions ($f_{ind:dom,1950}^N$, $f_{ind:dom,1950}^P$, and $f_{ind:dom,2000}^N$, $f_{ind:dom,2000}^P$; Sect. S3.3), and the industrial/commercial detergent parameters ($f_{ind:dom,1950-2012}^{DD,P-PO_4}$, $f_{ind:dom,1950-2015}^{D,P-PO(OH)_2}$, and $LD_{ind,cap,1991-2019}^{P-PO_4}$; Sect. S3.4).

S3.1 Fraction of protein supply wasted at the distribution and consumption level

Protein supply data do not exclude food waste at the distribution (retail) and consumption (domestic) levels. From FAO and SIK (2011) and Noleppa and Cartsburg (2015) (see Table S3), we collect food waste estimates for ten main commodity groups, namely cereals, meat, milk, fish and sea food, eggs, vegetables, fruits, oil crops, starchy roots and pulses. The study of FAO and SIK (2011) reports values for Europe around the year 2007 and the study of Noleppa and Cartsburg (2015) reports values for Germany around the year 2012. We also extract the protein supply data for each of these ten commodities from FAO (2021, 2022) and we verify that they represent more than 90 % of the total protein supply, as shown in Fig. S2. We assess the total protein waste as the average of the food waste over the ten commodities weighted by their supply amounts.

Specifically, for each commodity, we derive lower and upper estimates of the fraction of food waste at the distribution and consumption level combining the values from FAO and SIK (2011) and Noleppa and Cartsburg (2015), as reported in the last column of Table S3. We then assess the total protein waste as the average of the food waste over the ten commodities weighted by their supply amounts for both the lower and upper estimates and for the period 1950–2019 to consider the temporal variation in protein supply data. Our calculations show limited variations in the estimated lower and upper bound of the protein waste between the years, namely 0.139–0.154 considering the lower food waste estimates and 0.212–0.228 considering the upper food waste estimates. We select an uncertainty range of 0.14–0.23 for the total protein waste $f_{waste}^{protein}$ (-), as reported in Table S2.

We note that Morée et al. (2013) assessed the protein waste as a simple average of the values for the different commodities provided in FAO and SIK (2011), thus neglecting the fact that the different commodities contribute differently to the total protein supply. For Europe, they found a value of the 0.048 for the food waste at the distribution level and 0.134 of the consumption level. This results in a total value of the protein waste of around 0.18 ($1-(1-0.048)(1-0.134) \approx 0.18$) for the recent period, which is within our uncertainty range (0.14–0.23). Our uncertainty range also includes the value reported in IPCC (2019, Table 6.a new) for Europe of 0.15 (protein consumed as a fraction of protein supply is 0.85). In addition, Morée et al. (2013) assumed changes in the fraction of food waste over time for the period 1900-2000, while here we neglect such temporal differences because of a lack of data for the period before 2007–2012.

Table S2. Description of the model parameters, ranges used in this study, and explanations and references for the determination of the ranges

Parameter	Description	Unit	Lower value	Upper value	Note and references for parameter range
f^{pro}_{waste}	Fraction of protein supply wasted at the distribution and consumption level	(-)	0.14	0.23	The range was determines using food waste values for ten main food commodities from FAO and SIK (2011) and Noleppa and Cartsburg (2015) and the protein supply originating from these ten commodities from FAO (2021, 2022) (see details in Sect. S3.1).
f^{N}_{pro}	N content in protein	(kg N kg^{-1})	0.16	0.18	The range includes the values reported in Morée et al. (2013) and Mariotti et al. (2008) (0.16 and 0.18, respectively).
$f^{N:P}_{intake}$	N:P ratio for human intake	(kg N $(kgP)^{-1}$)	9.5	11.5	The range includes the values reported in Morée et al. (2013) that are 10, 10.6, and 11.1.
$f^{N,P}_{loss,hum}$	Fraction of human N and P intake lost via sweat, hair and blood	(-)	0.02	0.04	The range includes the value reported in Morée et al. (2013) that is 0.03.
$f^{N,P}_{loss,transport}$	Fraction of N and P emissions lost during wastewater collection and transport (losses in the sewer system, and in cesspits and during transportation of wastewater collected in cesspits to WWTPs)	(-)	0.05	0.15	The range includes the values reported in Morée et al. (2013) that is 0.1.
eff^{N}_{1}	Efficiency of N removal for primary treatment	(-)	0.1	0.25	The ranges were determined from the values reported in Van Drecht et al. (2009), Morée et al. (2013), IPCC (2019), Eurostat (2021), and Vigiak et al. (2020) (see details in Sect. S3.2).
eff^{N}_{23noN}	Efficiency of N removal for secondary and tertiary treatment without targeted denitrification	(-)	0.35	0.6	
eff^{N}_{3N}	Efficiency of N removal for tertiary treatment with targeted denitrification	(-)	0.7	0.95	
eff^{P}_{1}	Efficiency of P removal for primary treatment	(-)	0.1	0.3	
eff^{P}_{23noP}	Efficiency of P removal for secondary and tertiary treatment without targeted P removal	(-)	0.45	0.65	
eff^{P}_{3P}	Efficiency of P removal for tertiary treatment with targeted P removal	(-)	0.8	0.98	
$f^{N}_{ind:dom,1950}$	Fraction of industrial/commercial (indirect release) to domestic human (physiological) N gross emissions in 1950	(-)	0.35	0.75	The ranges were determined from values reported in Morée et al. (2013), Vigiak et al. (2020), and IPCC (2019) (see details in Sect. S3.3).
$f^{P}_{ind:dom,1950}$	Fraction of industrial/commercial (indirect release) to domestic human (physiological) gross P emissions in 1950	(-)	0.35	0.75	
$f^{N}_{ind:dom,2000}$	Fraction of industrial/commercial (indirect release) to domestic human (physiological) gross N emissions for the period 2000–2019	(-)	0.05	0.35	
$f^{P}_{ind:dom,2000}$	Fraction of industrial/commercial (indirect release) to domestic human (physiological) gross P emissions for the period 2000–2019	(-)	0.05	0.35	
$f^{DD,P-PO_4}_{ind:dom,1950-2012}$	Fraction of industrial/commercial (indirect release) to domestic dishwasher detergent phosphate use for the period 1950-2012	(-)	0.15	0.35	The ranges were determined from data provided by IHO and IKW (see details in Sect. S3.4).
$f^{D,P-PO(OH)_2}_{ind:dom,1950-2015}$	Fraction of industrial/commercial (indirect release) to domestic detergent phosphonate use for the period 1950-2015	(-)	0.15	0.5	
$LD^{P-PO_4}_{ind,cap,1991-2019}$	Industrial/commercial (indirect release)laundry booster phosphate P use per capita for the period 1991–2019	(kg P $capita^{-1}\,yr^{-1}$)	0.005	0.020	

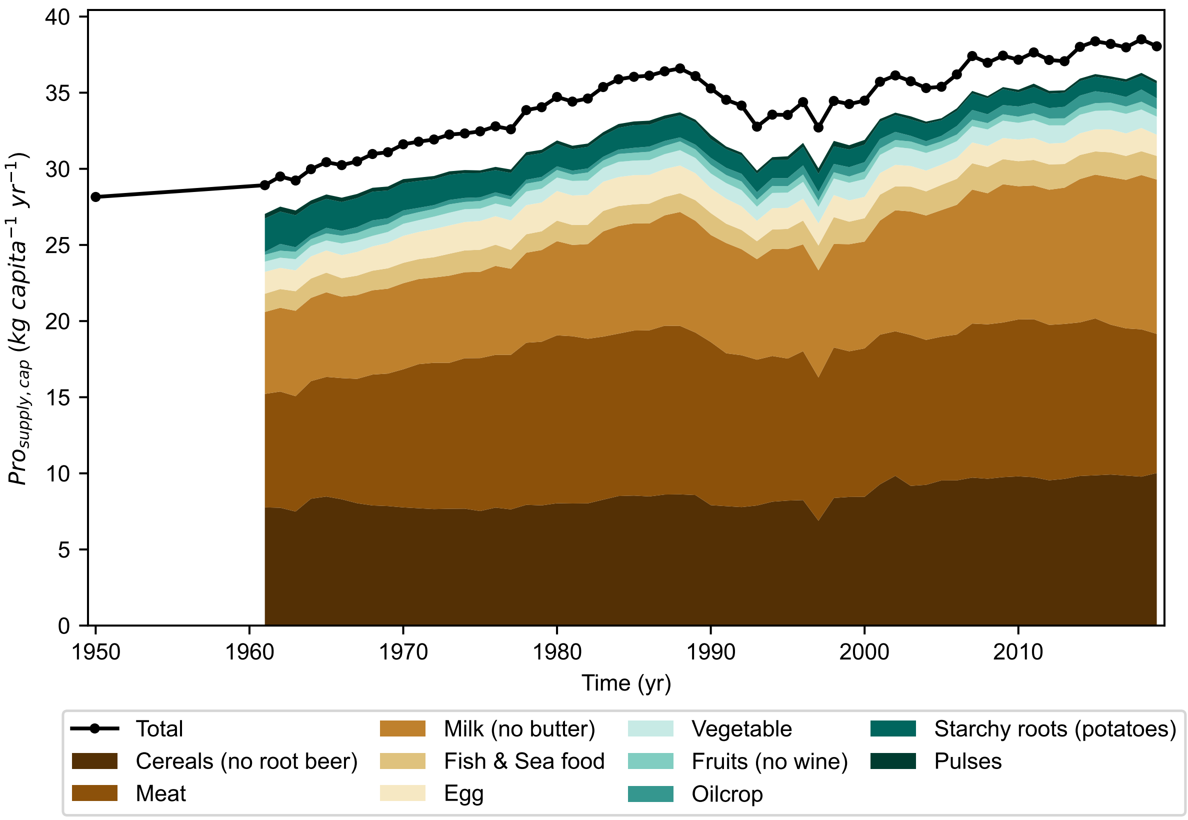


Figure S2. Annual protein supply ($Pro_{NUTS0,cap}$) totals and for ten commodity groups for Germany for the period 1950–2019. The values were taken from ©FAO (2021, 2022) over the period 1961–2019 and the 1950 value was estimated in this study as the average value over West and East Germany provided by ©FAO (1951) (License for FAO data: CC BY-NC-SA 3.0 IGO). For the total value (black), dots indicate years for which FAO data are available. The values for the other years were filled using linear interpolation. For the commodity groups, data are not available before 1961. The ten commodity groups reported account for more than 90 % of the total protein supply. These commodities were used to estimate the fraction of protein wasted at the distribution (retail) and consumption (domestic) levels.

Table S3. Food waste at the distribution (retail) level ($f^{food}_{waste,distr}$ (-)) and at the consumption (domestic) level ($f^{food}_{waste,cons}$ (-)) reported in FAO and SIK (2011) (©FAO) and Noleppa and Cartsburg (2015), and ranges selected in this study for these quantities and the total food waste $f^{food}_{waste,tot}$ (-))

Commodity	FAO and SIK (2011)		Noleppa and Cartsburg (2015)		Range used in this study		
	$f^{food}_{waste,distr}$	$f^{food}_{waste,cons}$	$f^{food}_{waste,distr}$	$f^{food}_{waste,cons}$	$f^{food}_{waste,distr}$	$f^{food}_{waste,cons}$	$f^{food}_{waste,tot}$ [a]
Cereals	0.02	0.25	0.015–0.114	0.23	0.015–0.114	0.230–0.250	**0.242–0.336**
Meat	0.04	0.11	0.015–0.04	0.16	0.015–0.04	0.11–0.16	**0.123–0.194**
Milk (no butter)	0.005	0.07	0.005–0.026	0.04	0.005–0.026	0.07–0.14	**0.075–0.162**
Fish and sea food	0.09	0.11	n.a.	n.a.	0.09–0.09	0.11–0.11	**0.19–0.19**
Eggs	n.a.	n.a.	0.005–0.026	0.16	0.005–0.026	0.16–0.16	**0.164–0.182**
Vegetables	0.10	0.19	0.05–0.10	0.26-0.29	0.05–0.10	0.19–0.29	**0.231–0.361**
Fruits (excluding wine)	0.10	0.19	0.05–0.10	0.24–0.29	0.05–0.10	0.19–0.29	**0.231–0.361**
Oilcrops	0.01	0.04	0.01	0.15	0.01–0-01	0.04–0.15	**0.05–0.159**
Starchy roots (potatoes)	0.07	0.17	0.07	0.26	0.07–0.07	0.17–0.26	**0.228–0.312**
Pulses	0.01	0.04	0.028	0.19	0.01-0.028	0.04-0.19	**0.05–0.213**

Notes: [a] $f^{food}_{waste,tot} = 1 - (1 - f^{food}_{waste,distr})(1 - f^{food}_{waste,dom})$
n.a.: not available

S3.2 Efficiency of N and P removal for the different types of wastewater treatment

We define the ranges for the wastewater treatment removal efficiency parameters based on the values reported in previous studies, that are reported in Tables S4–S5. We adopt the values of N and P removal efficiencies for tertiary treatment with targeted N and P removal, respectively, from Eurostat (2021) as lower bound. Regarding the other studies, we discard some values that we consider as being outliers (highlighed in red in Tables S4–S5), as further described in the following.

The lower values of the N removal efficiency for tertiary treatment with targeted denitrification in Kristensen et al. (2004) (as cited in Van Drecht et al. (2009)) appears to be very low (0.45) compared to the other studies and comparable to the efficiency of secondary and tertiary treatment without targeted denitrification. We hypothesize that this value may refer to tertiary treatment without targeted P removal and therefore that there are inconsistencies in the definition of the treatment types between Kristensen et al. (2004) and our study. This lower bound of 0.45 is also reported in IPCC (2019), which builds on the study of Kristensen et al. (2004). In addition, the upper value of the efficiency of P removal for secondary and tertiary treatment without targeted P removal in Kristensen et al. (2004) (0.9) also appears to be an outlier compared to the other studies and it is comparable to the efficiency of tertiary treatment with targeted P removal.

Vigiak et al. (2020) derived ranges of treatment efficiencies from the Waterbase-UWWTD database v6 (EEA, 2023). Many of these values are outliers as can be seen from Tables S4–S5. This can be due to the fact that data in the Waterbase-UWWTD database have large uncertainties. Vigiak et al. (2020) did not adopt these values for their estimation of N and P point sources, but they rather used typical values from the literature, that are reported in Tables S4–S5.

Table S4. Ranges from previous studies and selected in this study for the efficiency of N removal for primary treatment (eff_1^N), secondary and tertiary treatment without targeted denitrification ($eff_{23^{noN}}^N$), and tertiary treatment with targeted denitrification ($eff_{3^N}^N$).

Reference	eff_1^N		$eff_{23^{noN}}^N$		$eff_{3^N}^N$	
	Lower Value	Upper Value	Lower Value	Upper Value	Lower Value	Upper Value
Van Drecht et al. (2009), Morée et al. (2013)	0.1	0.1	0.35	0.35	0.8	0.8
Kristensen et al. (2004) as cited in Van Drecht et al. (2009)	0.2	0.25	0.36	0.55	**0.45**	0.83
Vigiak et al. (2020)[a]	0.25	0.25	0.55	0.55	0.8	0.8
Vigiak et al. (2020)[b]	0.19	**0.51**	0.39	**0.71**	0.7	0.9
IPCC (2019)	**0.05**	0.2	0.35	0.55	**0.45**	0.85
Eurostat (2021)	n.a.	n.a.	n.a.	n.a.	0.7	n.a.
This study	0.1	0.25	0.35	0.6	0.7	0.95

Values highlighted in red and bold are considered as outliers and are not used to define the parameter ranges in this study.

[a] Treatment removal efficiencies adopted in Vigiak et al. (2020).

[b] Interquartal range of the removal efficiencies in WWTPs estimated in Vigiak et al. (2020) from the Waterbase-UWWTD database v6 (EEA, 2023).

Table S5. Range of values from previous studies and selected in this study for the efficiency of P removal for primary treatment (eff_1^P), secondary and tertiary treatment without targeted P removal (eff_{23noP}^P), and tertiary treatment with targeted P removal (eff_{3P}^P).

Reference	eff_1^P		eff_{23noP}^P		eff_{3P}^P	
	Lower Value	Upper Value	Lower Value	Upper Value	Lower Value	Upper Value
Van Drecht et al. (2009), Morée et al. (2013)	0.1	0.1	0.45	0.45	0.9	0.9
Kristensen et al. (2004) as cited in Van Drecht et al. (2009)	0.28	0.3	0.51	**0.9**	0.88	0.95
Vigiak et al. (2020)[a]	0.3	0.3	0.6	0.6	0.9	0.9
Vigiak et al. (2020)[b]	0.1	**0.5**	0.5	**0.71**	**0.75**	0.92
Eurostat (2021)	n.a.	n.a.	n.a.	n.a.	0.8	n.a.
This study	0.1	0.3	0.45	0.65	0.8	0.98

Values highlighted in red and bold are considered as outliers and are not used the define the parameter ranges in this study.

[a] Treatment removal efficiencies adopted in Vigiak et al. (2020).

[b] Interquartal range of the removal efficiencies in WWTPs estimated in Vigiak et al. (2020) from the Waterbase-UWWTD database v6 (EEA, 2023).

S3.3 Fraction of industrial/commercial to domestic N and P emissions (indirect release)

We only consider industrial indirect release (e.g. breweries, factories producing strawboard, potato flour, food, paper, textiles, oil/grease/candles, and tanneries; Morée et al., 2013) and commercial indirect release (e.g. grocery stores, butchers; IPCC, 2006, 2019) that are collected in the sewer system and treated in public facilities. Similar to previous studies (Morée et al., 2013; Vigiak et al., 2020; IPCC, 2006, 2019), industrial/commercial gross N and P emissions are estimated as a fraction of domestic physiological N and P emissions. IPCC (2006, 2019) and Vigiak et al. (2020) report a similar fraction of industrial/commercial to domestic wastewater emissions for the recent period, namely a global default value of 0.25 in IPCC (2006, 2019) for N emissions for the period after 2006, and an average value across Europe of 0.23 in Vigiak et al. (2020) for waste emissions expressed in Population Equivalent (PE, i.e. amount of waste equal to 60 g per day of Biochemical Oxygen Demand) around the year 2014. Morée et al. (2013) suggest a lower global average value around the year 2000 equal to 0.15 for N and P emissions, but they explicitly refer to industrial release only (and not commercial release). They also consider that only 70 % of these industrial emissions are treated in WWTPs, while the remaining 30 % ends up in stabilization pounds or is lost via volatilization. Therefore, from Morée et al. (2013) the fraction of industrial to domestic N and P emissions that are treated in WWTPs is equal to 0.15 x 0.7 = 0.105. They also consider temporal variations in the ratio of industrial to domestic N and P emissions, with a global average equal to 1.4 (2 x 0.7 = 1.4) in 1900 and (0.5 x 0.7 = 0.35) in 1960. They assume linear temporal development between the years 1900 and 1960 and between the years 1960 and 2000.

In this study, we consider time varying fractions of industrial/commercial to domestic gross N and P emissions and similar to Morée et al. (2013) we consider a linear temporal development between the years 1950 and 2000 and a constant value between the years 2000 and 2019, since the values reported in IPCC (2006, 2019) and Vigiak et al. (2020) suggests that there were limited temporal changes during this time period. We define four parameters to estimate industrial/commercial gross emissions, namely the fraction of industrial/commercial to domestic N and P gross emissions in 1950 (denoted as $f^{N}_{ind:dom,1950}$ and $f^{P}_{ind:dom,1950}$, respectively) and in 2000 (denoted as $f^{N}_{ind:dom,2000}$ and $f^{P}_{ind:dom,2000}$, respectively). We consider that these parameters account for losses in N and P emissions prior to their collection in the sewer system and treatment in WWTPs (e.g. losses in stabilization pounds). We define an uncertainty range of **0.05—0.35** for $f^{N}_{ind:dom,2000}$ and $f^{P}_{ind:dom,2000}$ that includes the estimates of IPCC (2006, 2019), Morée et al. (2013), and Vigiak et al. (2020). Regarding $f^{N}_{ind:dom,1950}$ and $f^{P}_{ind:dom,1950}$, we define a wide uncertainty range of **0.35–0.75**, since these coefficients are highly uncertain. This interval includes the value from Morée et al. (2013) that is equal to 0.525 (calculated using linear interpolation between the values in 1900 and 1960). The ratio defined in this section only refer to human (physiological) N and P emissions. We refer to Section S3.4 for an explanation on the estimation of industrial/commercial detergent use.

S3.4 Industrial/commercial detergent parameters

We account for the industrial/commercial detergent P use in businesses and institution such as bars, restaurants, canteens, hotels, bakeries, butcher shops, schools, hospitals, retirement homes (see details on industrial/commercial detergents in Mehlhart et al., 2021). The laundry detergent phosphate use data before 1991 and 1992 for West Germany and East Germany, respectively already include both the domestic and industrial/commercial parts (Sect. S5.1). To estimate the other industrial/commercial detergent P components, to our knowledge, the only existing data come from the German Industry Association for Hygiene and Surface Protection for Industrial and Institutional Applications (IHO) and are provided at NUTS-0 level around the year 2008 for phosphonate (23.06.2010 IHO personal communication, as cited in Groß et al., 2012) and around the year 2015 for both phosphate (laundry and dishwasher detergent) and phosphonate (28.07.2017 IHO personal communication, as cited in Mehlhart et al., 2021).

We estimate laundry detergent (LD) phosphate after 1991 and 1992 for West Germany and East Germany, respectively, using a constant rate of industrial/commercial LD phosphate P use per capita, denoted as $LD^{P-PO_4}_{ind,cap,1991-2019}$ $(kg\ P\ cap^{-1}\ yr^{-1})$. We consider that industrial/commercial LD phosphate only comes from laundry boosters, while normal industrial/commercial LD is phosphate-free (Mehlhart et al., 2021). We use IHO data of industrial/commercial laundry booster use that are provided in 2015 at NUTS-0 level (Mehlhart et al., 2021). By dividing the IHO total amount by the NUTS-0 level population count in 2015, we find that LD booster phosphate consumption is around 0.011 $kg\ P\ capita^{-1}\ yr^{-1}$. We account for the uncertainty in this estimate by varying $LD^{P-PO_4}_{ind,cap,1991-2019}$ within the uncertainty range of **0.005–0.020** $kg\ P\ capita^{-1}\ yr^{-1}$.

In absence of further information, we assume that industrial/commercial dishwasher detergent (DD) phosphate and detergent phosphonate use followed the same temporal developments as domestic detergent use. We assess industrial/commercial detergent use from domestic use using a fraction of industrial/commercial to domestic DD phosphate use for the period 1950-2012, denoted as $f^{DD,P-PO_4}_{ind:dom,1950-2012}$ *(-)*, and a fraction of industrial/commercial to domestic detergent phosphonate use for the period 1950-2015, denoted as $f^{D,P-PO(OH)_2}_{ind:dom,1950-2015}$ *(-)*. These two fractions are estimated from IHO data (industrial/commercial use) and IKW data (domestic use). We also consider the European Union Detergent Regulation (2012) that led to a reduction in DD phosphate use (as can be seen in Figure S3.a after 2012) and a consequent increase in phosphonate detergent use (as can be seen in Figure S3.b after 2015) for domestic detergent only. This regulation does not apply to industrial/commercial DD for which we can consider that the composition was unchanged (Mehlhart et al., 2021). Therefore for the most recent years (after 2012 for DD phosphate and after 2015 and detergent phosphonate), we consider that industrial/commercial DD phosphate and detergent phosphonate are constant.

IHO data of industrial/commercial DD phosphate use are provided in 2015 only at NUTS-0 level (Figure S3.a). To calculate the fraction of industrial/commercial to domestic DD phosphate use, we do not use the domestic DD phosphate use per capita in 2015 but in 2012. This is because the 2015 value is affected by the European Union Detergent Regulation (2012). From Figure S3.a we see that the domestic DD phosphate use per capita was relatively stable in the period 2007–2012 and therefore we can consider that the 2012 value is representative of the DD phosphate use per capita before the implementation of the 2012 EU regulation. Therefore, we consider that the composition of industrial/commercial DD in 2015 was similar to that of domestic DD in 2012. We obtain a ratio of industrial/commercial to domestic DD phosphate equal to around 0.26. To account for uncertainty, we vary $f_{pro:dom}^{DD,P-PO_4}$ in the uncertainty range **0.15–0.35**.

IHO data of industrial/commercial detergent phosphonate use are provided in 2008 and in 2015 at NUTS-0 level (Figure S3.b). We find that the fraction of industrial/commercial to domestic detergent phosphonate use varies substantially between 2008 (0.21) and 2015 (0.43), which could be due to either a large inter-annual variability in detergent use or uncertainty in the data. To account for uncertainty, we vary $f_{pro:dom}^{D,P-PO(OH)_2}$ in the uncertainty range **0.15–0.50**.

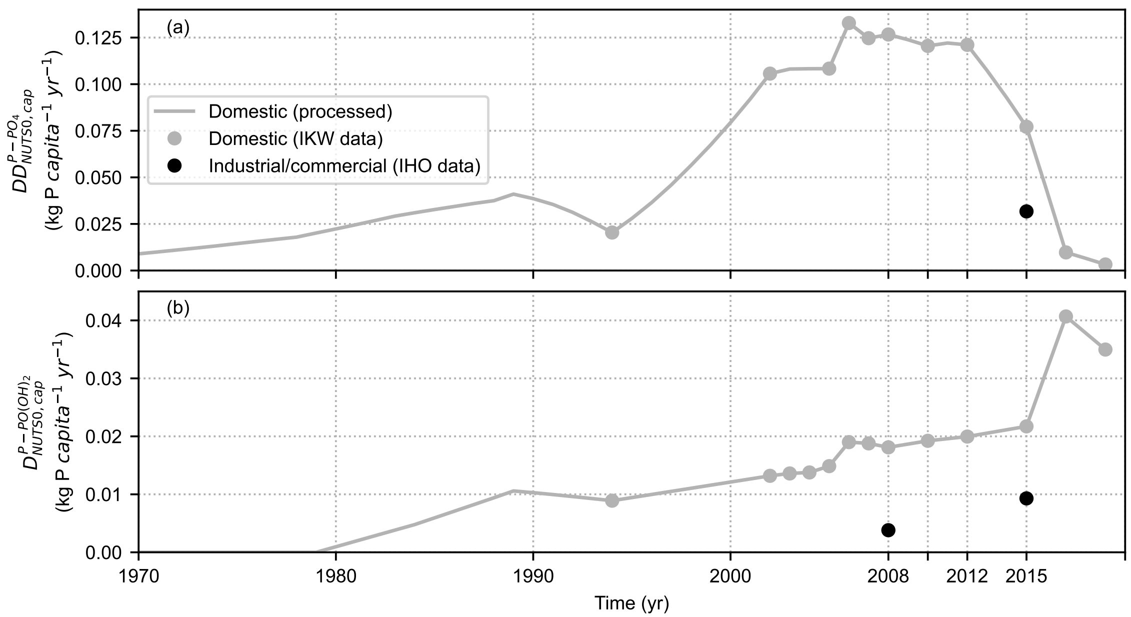


Figure S3. Domestic and industrial/commercial dishwasher detergent data at NUTS-0 level, namely (a) dishwasher detergent phosphate ($DD^{P-PO_4}_{NUTS0,cap}$) and (b) detergent phosphonate ($D^{P-PO(OH)_2}_{NUTS0,cap}$). The circles represent raw detergent data that were divided by the NUTS-0 population counts to obtain the amount per capita. The raw data of detergent domestic use come from the German Cosmetic, Toiletry, Perfumery and Detergent (IKW, 2005, 2011, 2017, 2019). The raw data of detergent industrial/commercial use come from the German Industry Association for Hygiene and Surface Protection for Industrial and Institutional Applications and are provided only around the year 2008 (23.06.2010 IHO personal communication, as cited in Groß et al., 2012) and around the year 2015 (28.07.2017 IHO personal communication, as cited in Mehlhart et al., 2021). The grey lines represent the domestic detergent final data processed in this study. Source of the data: this figure builds on data from ©IKW that are under a license that does not allow commercial use (https://www.ikw.org/impressum).

S4 Population data

S4.1 Processing of the population statistical data at NUTS-3 level accounting for the NUTS-3 region reforms

Using statistical data provided by the Statistical Office of Germany (SO-DE, 2022b), we infer population at NUTS-3 level for the period 1995–2019 with respect to the 2020 NUTS-3 classification that includes 401 regions (BKG, 2020). For each year, statistical data are reported with respect to the NUTS-3 classification in 2020. Therefore, we need to account for the NUTS-3 region reforms of (1) 2007 in Saxony-Anhalt, (2) 2008 in Saxony, (3) 2011 in Mecklenburg-Western Pomerania, and (4) for the formation of the new NUTS-3 region of Göttingen in Lower Saxony in 2016. For six NUTS-3 regions located in Saxony-Anhalt (Dessau-Roßlau, Anhalt-Bitterfeld, Jerichower, Wittenberg, Salzlandkreise and Harz) and two NUTS-3 regions located in Mecklenburg-Western Pomerania (Mecklenburgische Seenplatte and Vorpommern-Greifswald), the population counts cannot be directly retrieved from the statistical data. The NUTS-3 region reforms led to the division of three former NUTS-3 regions (Anhalt-Zerbst, Aschersleben and Demmin) to form these new eight NUTS-3 regions (see details in Table S6). We reconstruct the population counts before 2011 for the two NUTS-3 regions of Mecklenburg-Western Pomerania considering that the temporal variations were the same in these two NUTS-3 regions, while ensuring that the sum of the population counts over these two NUTS-3 regions is consistent with the statistical data. Similarly, we reconstruct the population counts before 2007 for the six NUTS-3 regions that belong to Saxony-Anhalt.

Table S6. Formation of the new NUTS-3 regions of Dessau-Roßlau, Anhalt-Bitterfeld, Jerichower, Wittenberg, Salzlandkreis, Harz, Mecklenburgische Seenplatte and Vorpommern-Greifswald

New NUTS-3 region	State	Year of formation	Former NUTS-3 regions[a]
Dessau-Roßlau	Saxony-Anhalt	2007	Dessau, **Anhalt-Zerbst**
Anhalt-Bitterfeld	Saxony-Anhalt	2007	Bitterfeld, Köthen,**Anhalt-Zerbst**
Jerichower	Saxony-Anhalt	2007	Jerichower, **Anhalt-Zerbst**
Wittenberg	Saxony-Anhalt	2007	Wittenberg, **Anhalt-Zerbst**
Salzlandkreis	Saxony-Anhalt	2007	Bernburg, Schönebeck, **Aschersleben-Staßfurt**
Harz	Saxony-Anhalt	2007	Halberstadt, Quedlinburg, Wernigerode, **Aschersleben-Staßfurt**
Mecklenburgische Seenplatte	Mecklenburg-Western Pomerania	2011	Neubrandenburg, Mecklenburg-Strelitz, Müritz, **Demmin**
Vorpommern-Greifswald	Mecklenburg-Western Pomerania	2011	Greifswald, Ostvorpommern, Uecker-Randow, **Demmin**

[a] The territories of the former NUTS-3 regions that appear in bold were divided during the NUTS-3 region reforms.

S4.2 Adjustment of the HYDE grid-level total population counts to match the statistical data

We utilise the gridded History Database of the Global Environment dataset (HYDE; Klein Goldewijk et al., 2017) that provides total, urban, and rural population counts for the period 1950–2017 at a spatial resolution of 5'. We derive the gridded total population counts (sum of urban and rural population) by adjusting the HYDE data to match annual statistical data at NUTS-1 level available for 1950–2019 and at NUTS-3 level available for 1995–2019. We downscale the HYDE data to our target resolution of 0.015625°using nearest neighbour resampling. We interpolate the HYDE data linearly to fill the values for missing years in the period 1950–2017, and assume the same values in 2018 and 2019 as in 2017. The HYDE dataset provides three population estimates at grid level corresponding to a baseline, upper, and lower scenario. Only the magnitude of the total population varies between the three scenarios, while the gridded fractions of urban and rural population to total population are constant across scenarios. After adjustment, we obtain only one total population estimate at grid level, as we do not detect appreciable differences between the adjusted estimates of the three scenarios. In the following, we provide further details on the adjustment procedure.

We adjust the resampled and interpolated HYDE data of total population so that they are consistent with the statistical data at NUTS-1 and NUTS-3 level. For the k_{j_i}-th grid cell that belongs to the j_i-th NUTS-3 region which is within the i-th NUTS-1 region, and for year *t*, we apply to the gridded original HYDE total population count denoted as $Pop_{grid,HYDE}(t,k_{j_i})$ two temporally-varying multiplier coefficients, namely a coefficient defined at the NUTS-3 level and denoted as $f_{pop,NUTS3}(t,j_i)$ (-), and a coefficient defined at the NUTS-1 level denoted as $f_{pop,NUTS1}(t,i)$ (-):

$$Pop_{grid,HYDE,adj}(t,k_{j_i}) = Pop_{grid,HYDE}(t,k_{j_i}) f_{pop,NUTS3}(t,j_i) f_{pop,NUTS1}(t,i) \tag{S5}$$

where $Pop_{grid,HYDE,adj}(t,k_{j_i})$ (-) is the adjusted HYDE total population count at grid level. The NUTS-3 level multiplier coefficient $f_{pop,NUTS3}(t,j_i)$ is defined as:

$$f_{pop,NUTS3}(t,j_i) = \frac{Pop_{NUTS3,stat}(t,j_i)}{\sum_{k_{j_i} \in E_{grid,j_i}} Pop_{grid,HYDE}(t,k_{j_i})} \quad for\ t \geq 1995 \tag{S6}$$

$$f_{pop,NUTS3}(t,j_i) = f_{pop,NUTS3}(1995,j_i) \quad for\ t < 1995 \tag{S7}$$

where $Pop_{NUTS3,stat}(t,j_i)$ is the NUTS-3 level total population count from statistical data and E_{grid,j_i} is the ensemble of grid cells that belong to the j_i-th NUTS-3 region which is within the i-th NUTS-1 region. From Equation S6, the NUTS-3 level multiplier is equal to the ratio of the statistical data at NUTS-3 level to the HYDE data at NUTS-3 level. It ensures that the adjusted HYDE data are consistent with the NUTS-3 level statistical data that are available for the period 1995–2019. Equation S7 ensures the continuity of the adjusted HYDE data between the period 1995–2019 and the period 1950–1994 where no NUTS-3 level statistical information is available. We thereby assume that before 1995, the relative mismatch between the NUTS-3 level statistical data and HYDE data was the same as the one in 1995.

The NUTS-1 level multiplier coefficient of Equation S5 $f_{pop,NUTS1}(t,i)$ is defined as:

$$f_{pop,NUTS1}(t,i) = 1 \quad for\ t \geq 1995 \tag{S8}$$

$$f_{pop,NUTS1}(t,i) = \frac{Pop_{NUTS1,stat}(t,i)}{\sum_{j_i \in E_{NUTS3,i}} \sum_{k_{j_i} \in E_{grid,j_i}} Pop_{grid,HYDE}(t,k_{j_i}) f_{pop,NUTS3}(t,j_i)} \quad for\ t < 1995 \tag{S9}$$

where $Pop_{NUTS1,stat}(t,i)$ is the NUTS-1 level total population count from statistical data and $E_{NUTS3,i}$ is the ensemble of NUTS-3 regions that belong to the *i*-th NUTS-1 region. Equation S8 ensures that, for the period 1995–2019, no further correction is applied to the HYDE data beyond the NUTS-3 level multiplier (defined in Equation S6). From Equation S9, the NUTS-1 level multiplier is equal to the ratio of the statistical data at NUTS-1 level to the HYDE data (adjusted using the NUTS-3 level multiplier) at NUTS-1 level. It ensures that the adjusted HYDE data are consistent with the NUTS-1 level statistical data that are available for the period 1950–1994.

We apply the multiplier coefficients of Eq. S5 to adjust the gridded total, urban and rural population. The original gridded HYDE data (baseline scenario) of urban and rural population averaged over three different decades (1970–1979, 1990–1999, and 2010–2019) are shown in Fig S4, while the adjusted data are shown in Figure S5 and S6. The relative differences between the maps of Fig S4 and S5, that are reported in Figure S7, are lower than 10 % for more than 75 % of the grid cells, and are higher than 100 % for less than 1 % of the grid cells.

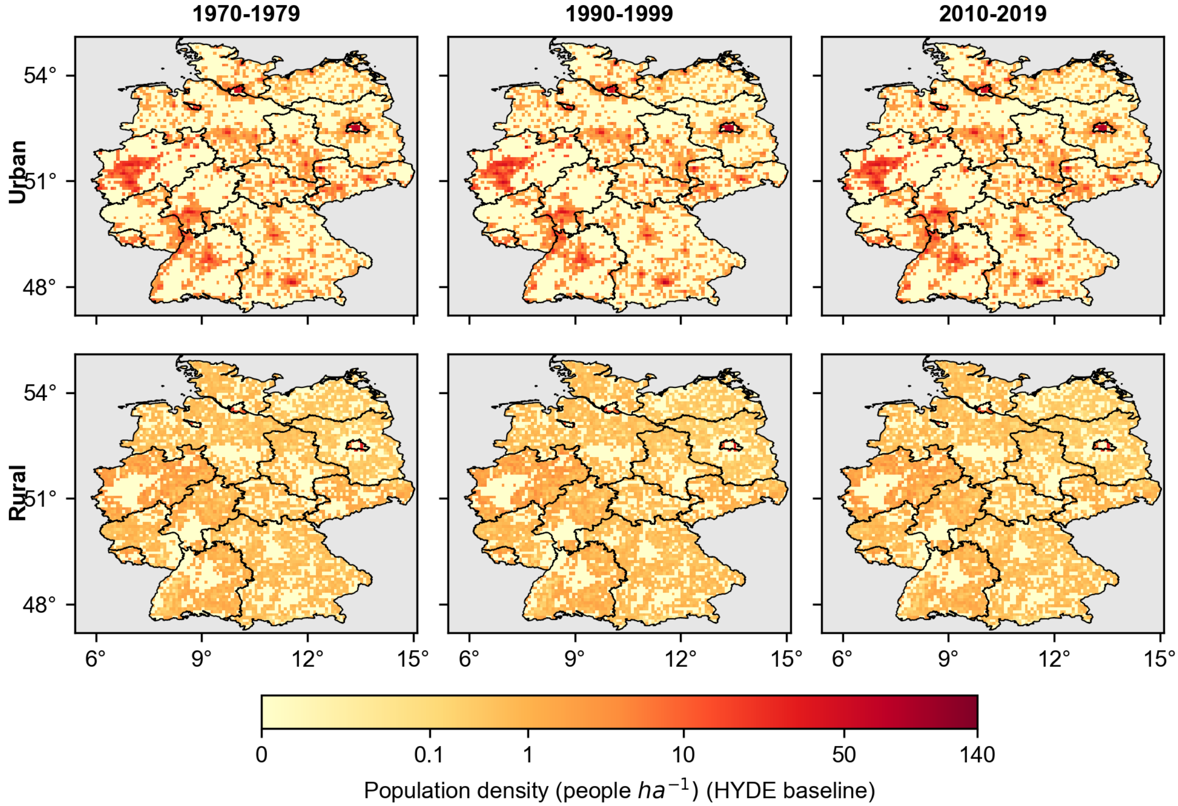


Figure S4. Gridded urban and rural population density from the original HYDE data (baseline scenario), averaged over three different decades (1970–1979, 1990–1999, and 2010–2019). The figure also reports the boundaries of the 16 NUTS-1 units of Germany. Data source: the HYDE data (Klein Goldewijk et al., 2017, 2022) are under a CC BY 4.0 license. The NUTS-1 map comes from the German Federal Agency for Cartography and Geodesy (BKG, 2020) ©GeoBasis-DE/BKG dl-de/by-2-0 license (https://www.govdata.de/dl-de/by-2-0). The map is shown in the WGS84 system in the figure.

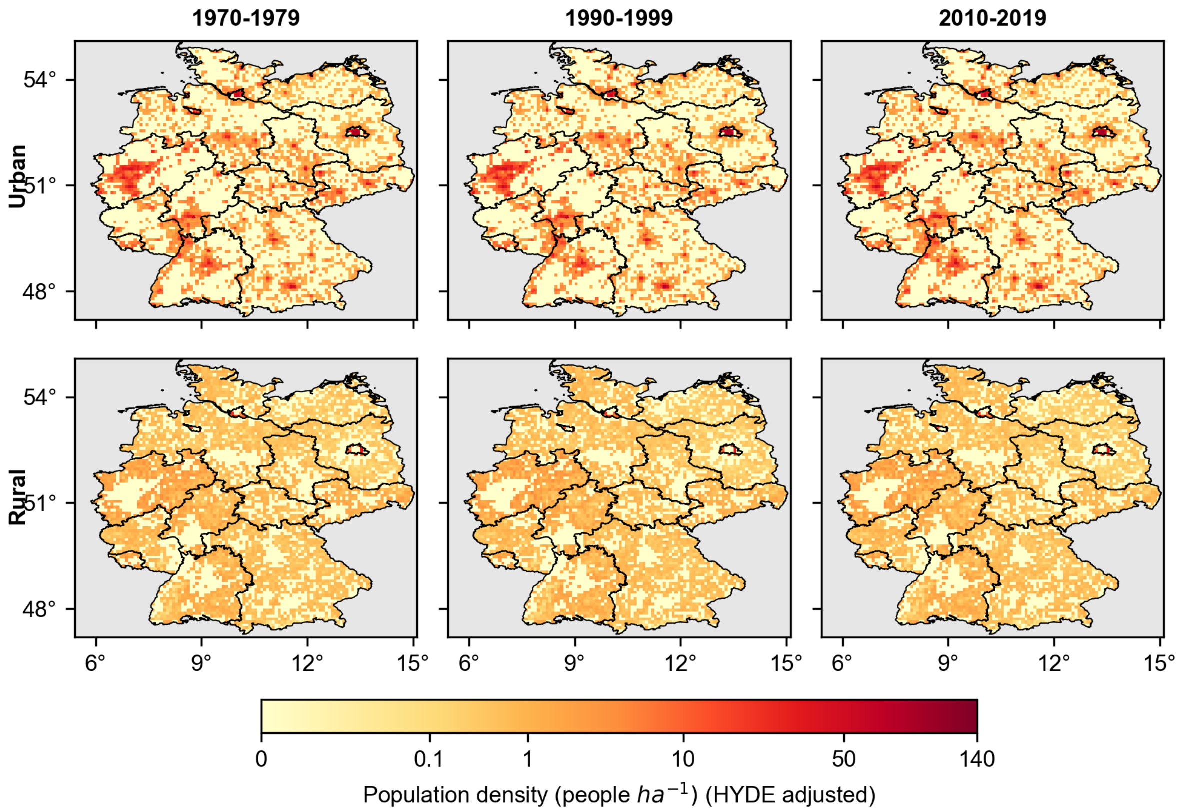


Figure S5. Gridded urban and rural population density from the adjusted HYDE data averaged over three different decades (1970–1979, 1990–1999, and 2010–2019). We used these data to downscale the point sources emissions from NUTS-1 to grid level. The figure also reports the boundaries of the 16 NUTS-1 units of Germany. Data source: the data shown in this figure were derived by adjusting the HYDE data (Klein Goldewijk et al., 2017, 2022), that are under a CC BY 4.0 license, using statistical population data provided by the German Statistical Office (details in Table 2 in the main text). The NUTS-1 map comes from the German Federal Agency for Cartography and Geodesy (BKG, 2020) ©GeoBasis-DE/BKG dl-de/by-2-0 license (https://www.govdata.de/dl-de/by-2-0). The map is shown in the WGS84 system in the figure.

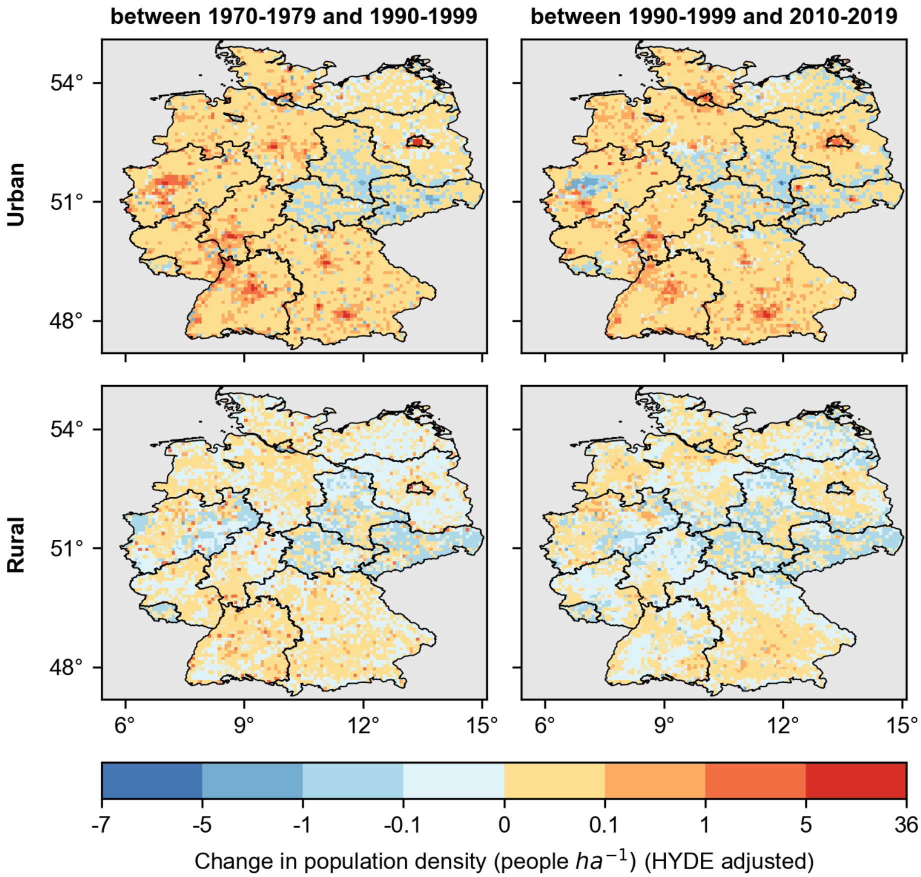


Figure S6. Absolute changes in gridded urban and rural population density from the HYDE data averaged between the decade 1970–1979 and 1990–1999 (left panels), and between 1990–1999 and 2010–2019 (right panels). Actual values of the population from the adjusted HYDE data are shown in Fig. S5. The figure also reports the boundaries of the 16 NUTS-1 units of Germany. The NUTS-1 map comes from the German Federal Agency for Cartography and Geodesy (BKG, 2020) ©GeoBasis-DE/BKG dl-de/by-2-0 license (https://www.govdata.de/dl-de/by-2-0). The map is shown in the WGS84 system in the figure.

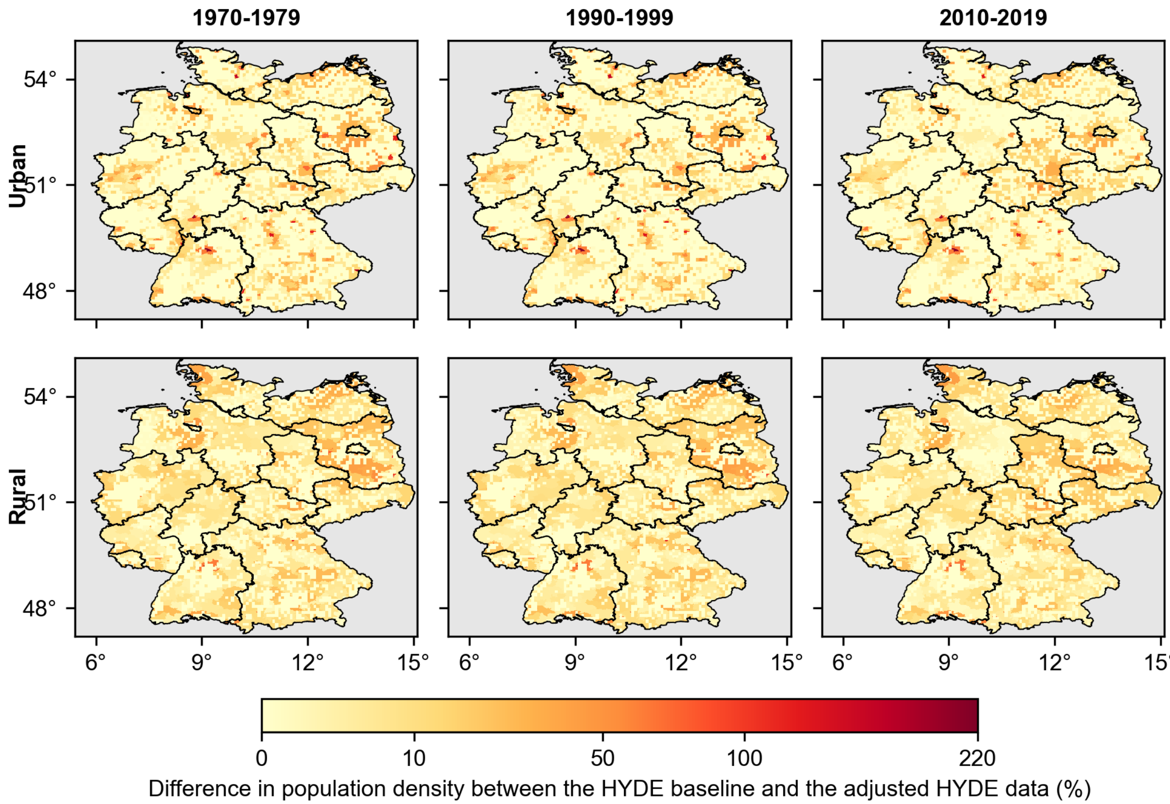


Figure S7. Relative differences in gridded urban and rural population density between the original HYDE data (baseline scenario, Fig. S4) and the adjusted HYDE data (Fig. S5), averaged over three decades (1970–1979, 1990–1999, and 2010–2019). The relative differences were calculated as $100\frac{|Pop_{grid,HYDE} - Pop_{grid,HYDE,adj}|}{Pop_{grid,HYDE}}$, where $Pop_{grid,HYDE}$ is the original HYDE population data averaged over a given decade (baseline scenario) and $Pop_{grid,HYDE,adj}$ is the adjusted HYDE population data averaged over a given decade. The relative differences are lower than 10 % for more than 75 % of the grid cells, and are higher than 100 % for less than 1 % of the grid cells for all six maps reported in the figure. The figure also reports the boundaries of the 16 NUTS-1 units of Germany. The NUTS-1 map comes from the German Federal Agency for Cartography and Geodesy (BKG, 2020) ©GeoBasis-DE/BKG dl-de/by-2-0 license (https://www.govdata.de/dl-de/by-2-0). The map is shown in the WGS84 system in the figure.

S4.3 Comparison between the population counts from the HYDE data (before correction) and statistical data

We compare the total population counts from the original HYDE population data (before adjustment) and the adjusted HYDE data (data adjusted in this study to match the statistical data) at NUTS-0 level (Table S7, Fig. S8), NUTS-1 level (Table S7, Fig. S9) and NUTS-3 level (Fig. S10). The comparison is performed over the period 1950–2017 at NUTS-0 and NUTS-1 level, and 1995–2017 at NUTS-3 level, since NUTS-3 level statistical data are not available before 1995.

At NUTS-0 and NUTS-1 level, the difference between original and adjusted HYDE data are generally contained with relative biases in the range ± 0.07 and correlation coefficient higher than 0.8, (Table S7, Fig. S8, and Fig. S9). However, they can be large for some NUTS-1 regions such as Bremen, for which the relative bias is around -0.22 and correlation coefficient around 0.26, Table S7.

At NUTS-3 level, the relative bias between the average population from the original and adjusted ata HYDE is higher than 0.1 or lower than -0.1 for as much as 42 % of the NUTS-3 regions and is higher than 0.2 or lower than -0.2 for 23 % of the NUTS-3 regions (see Figure S10). Therefore, the differences are generally larger at NUTS-3 level compared to NUTS-1 level.

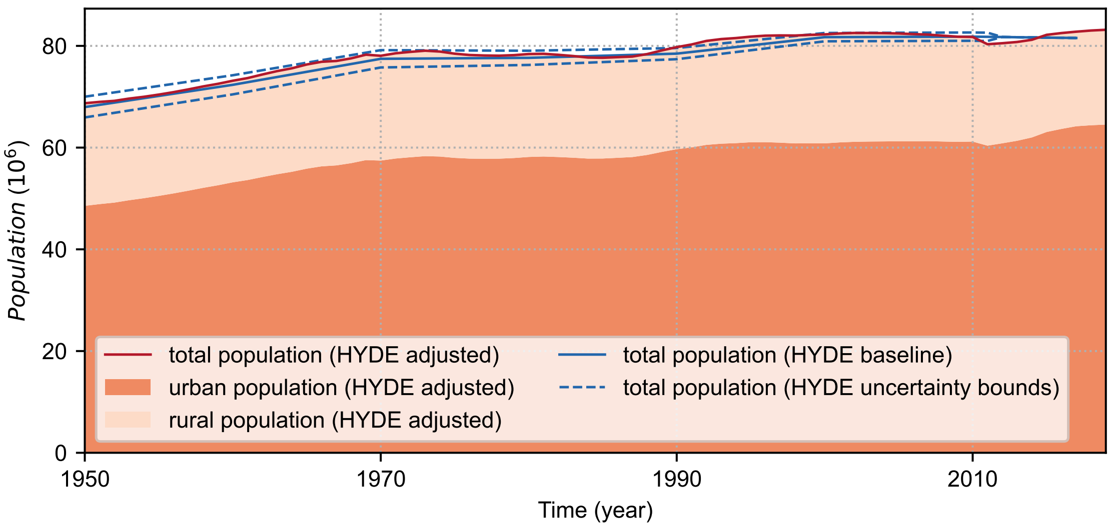


Figure S8. Population counts estimated at NUTS-0 level from the original HYDE data for the period 1950–2017 and from the HYDE data that were adjusted to match statistical data for the period 1950–2019. The original HYDE data include a baseline scenario and uncertainty bounds (lower and upper scenarios). Regarding the adjusted HYDE data, the figure shows the total, urban and rural population counts that were used to derive the N and P point sources data in this study. Data source: HYDE data (Klein Goldewijk et al., 2017, 2022) are under a CC BY 4.0 license. Statistical population data are from the German Statistical Office details in Table 2 in the main text.

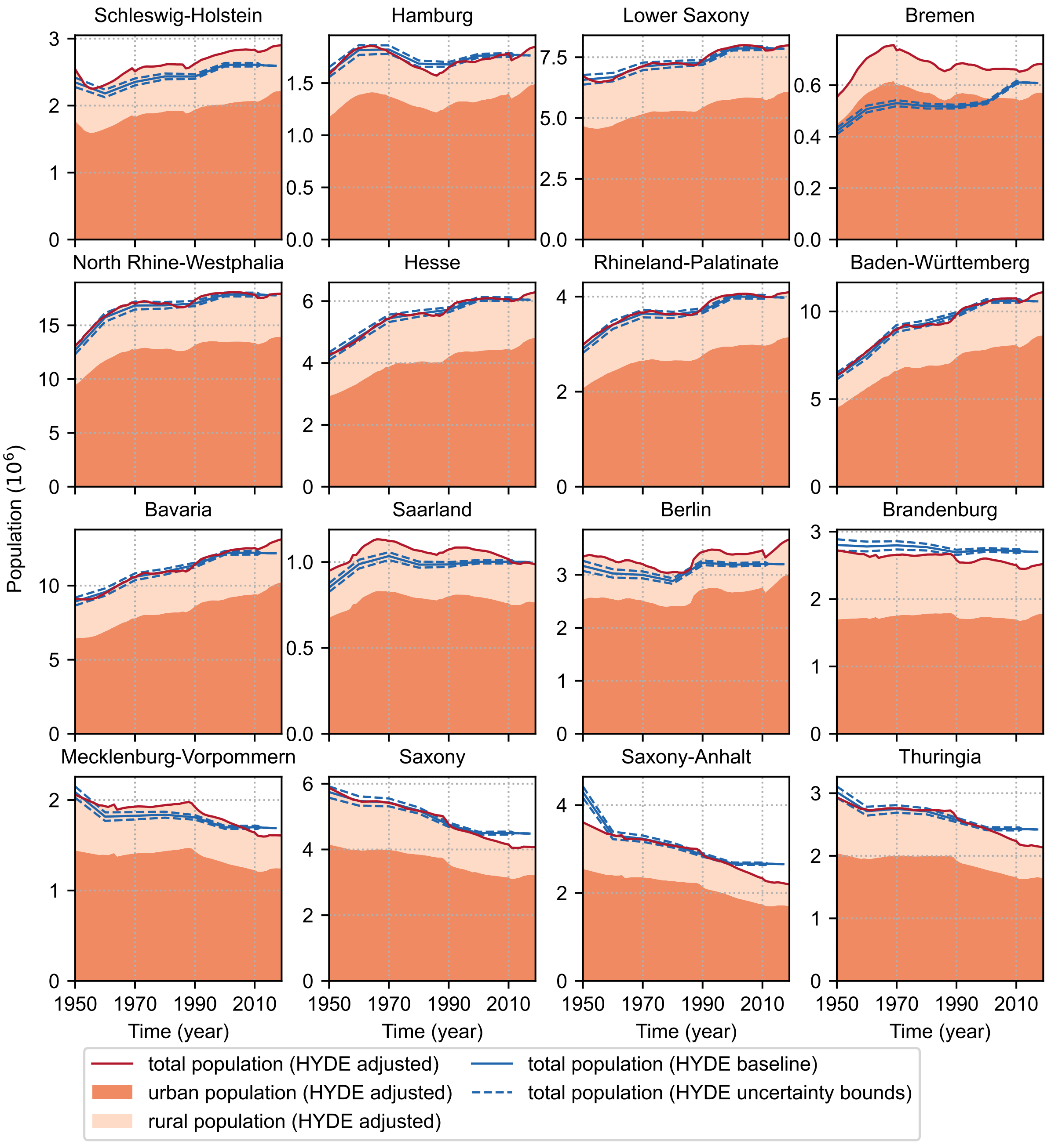


Figure S9. Population count estimated at NUTS-1 level from the original HYDE data for the period 1950–2017 and from the HYDE data that were adjusted to match statistical data for the period 1950–2019. The original HYDE data include a baseline scenario and uncertainty bounds (lower and upper scenarios). Regarding the adjusted HYDE data, the figure shows the total, urban and rural population counts that were used to derive the N and P point sources data in this study. Data source: HYDE data (Klein Goldewijk et al., 2017, 2022) are under a CC BY 4.0 license. Statistical population data are from the German Statistical Office details in Table 2 in the main text.

Table S7. Relative bias $rbias^{pop}$ and correlation coefficient ρ^{pop} between the population estimates from the original and adjusted HYDE dataset at NUTS-1 and NUTS-0 level for the period 1950–2017.

State	$rbias^{pop}$ (-) [a]			ρ^{pop} (-)		
	baseline	lower	upper	baseline	lower	upper
Schleswig-Holstein	-0.07	-0.08	-0.05	0.97	0.97	0.96
Hamburg	0.01	-0.01	0.03	0.91	0.86	0.91
Lower Saxony	-0.0	-0.02	0.01	0.99	0.98	0.99
Bremen	-0.22	-0.23	-0.21	0.25	0.22	0.29
North Rhine-Westphalia	-0.01	-0.03	0.0	0.99	0.98	0.99
Hesse	-0.0	-0.02	0.01	0.99	0.99	0.99
Rhineland-Palatinate	-0.01	-0.03	0.0	0.99	0.99	0.99
Baden-Württemberg	-0.0	-0.02	0.01	0.99	0.99	0.99
Bavaria	-0.01	-0.02	0.01	0.99	0.99	0.99
Saarland	-0.07	-0.08	-0.05	0.72	0.61	0.82
Berlin	-0.06	-0.08	-0.05	0.91	0.89	0.9
Brandenburg	0.05	0.04	0.07	0.78	0.46	0.84
Mecklenburg-Vorpommern	-0.03	-0.05	-0.01	0.86	0.83	0.87
Saxony	0.01	-0.0	0.03	0.98	0.97	0.98
Saxony-Anhalt	0.05	0.03	0.06	0.89	0.88	0.9
Thuringia	0.01	-0.0	0.03	0.92	0.91	0.93
Germany	-0.01	-0.02	0.01	0.99	0.98	0.99

[a] $rbias^{pop}$ is calculated as $\frac{\overline{Pop_{original}} - \overline{Pop_{adjusted}}}{\overline{Pop_{adjusted}}}$, where $\overline{Pop_{original}}$ is the temporal average of the total population count from the original HYDE dataset and $\overline{Pop_{adjusted}}$ is the temporal average of the total population count from the adjusted HYDE data that matches the statistical data at NUTS-0 and NUTS-1 level. The Values are reported for the three scenarios of the original HYDE dataset (baseline, lower and upper), the adjusted HYDE dataset has only one estimate.

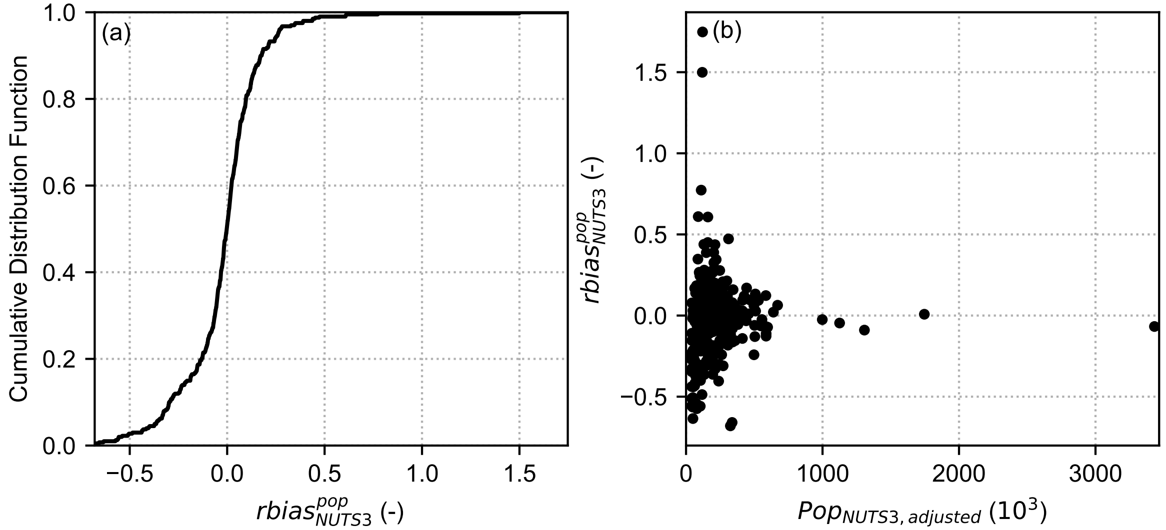


Figure S10. Relative bias ($rbias^{pop}_{NUTS3}$) between the population estimates of the original and adjusted HYDE data at NUTS-3 level for the period 1995–2017 calculated for the 401 NUTS-3 regions and for the baseline scenario of the HYDE dataset. (a) Cumulative distribution function of $rbias^{pop}_{NUTS3}$ across the 401 NUTS-3 regions. (b) $rbias^{pop}_{NUTS3}$ against the total population from the NUTS-3-level adjusted HYDE data that matches the statistical data ($Pop_{NUTS3,adjusted}$). Similar results are obtained when considering the lower and upper scenario of the original HYDE dataset (not shown).

Notes: $rbias^{pop}_{NUTS3}$ is calculated as $\frac{\overline{Pop^{tot}_{NUTS3,original}} - \overline{Pop_{NUTS3,adjusted}}}{\overline{Pop_{NUTS3,adjusted}}}$, where $\overline{Pop_{NUTS3,original}}$ is the temporal average over 1995–2017 of the NUTS-3-level total population count from the original HYDE dataset and $\overline{Pop_{NUTS3,adjusted}}$ is the temporal average over 1995–2017 of the NUTS-3-level total population count from the adjusted HYDE dataset. Data source: HYDE data (Klein Goldewijk et al., 2017, 2022) are under a CC BY 4.0 license. Statistical population data are from the German Statistical Office (details in Table 2 in the main text)

S5 Detergent data

S5.1 Laundry detergent (LD) phosphate data

Figure S11 shows the raw and processed LD phosphate data.

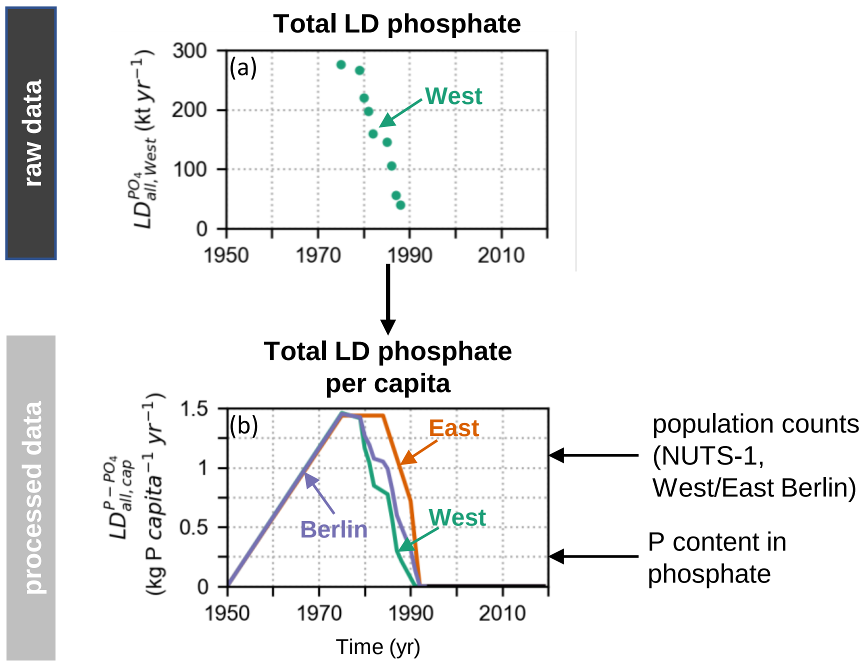


Figure S11. Raw (a) and processed (b) laundry detergent (LD) phosphate data for the domestic and industrial/commercial sectors. Data source: (a) Raw data (total LD phosphate use, $LD_{all}^{PO_4}$) come from the BMU (1989) and are provided for West Germany only for the period 1975–1989. (b) data of total LD phosphate P use per capita ($LD_{all,cap}^{PO_4-P}$) were processed for West, East and Berlin for the period 1950–1990, using the BMU data, NUTS-1 level population data from the German Statistical Office and population data for West/East Berlin from the Statistical Office of Berlin (details on population data sources in Table 2 in the main text).

S5.2 Domestic dishwasher detergent (DD) phosphate data

S5.2.1 Workflow for domestic dishwasher detergent (DD) phosphate reconstruction

Figure S12 shows the workflow for the construction of domestic DD phosphate data. Raw data are shown in Figure S12.a-c, namely domestic DD phosphate use ($DD_{dom,NUTS0}^{PO_4}$) provided by the German Cosmetic, Toiletry, Perfumery and Detergent Association IKW (IKW, 2005, 2011, 2017, 2019) at NUTS-0 level for the period 1994-2019 (Figure S12.a), and dishwasher ownership data for West and East Germany for the period 1962–2018 (N_{house}^{dish}, Figure S12.b) and at NUTS-1 level in 2008 (N_{house}^{dish}, Figure S12.c) provided by the Statistical Office of Germany (SO-DE, Fachserie 15 Heft 1). We disaggregate the dishwasher ownership totals for West and East Germany using the 2008 NUTS-1 level data (Figure S12.e, detailed explanations on the disaggregation are reported in Section S5.2.2). We then sequentially assess the domestic DD phosphate P use per household owning a dishwasher ($DD^{PO_4-P}_{dom,house}$) at NUTS-0 level (Figure S12.d) and the domestic DD phosphate P use ($DD^{PO_4-P}_{dom}$) at NUTS-1 level (Figure S12.f) to then derive the domestic DD phosphate P use per capita ($DD^{PO_4-P}_{dom,cap}$) at NUTS-1 level (final processed data, Figure S12.g).

S5.2.2 Household dishwasher ownership at NUTS-1 level

We use data of the number of private households owning at least one dishwasher (Equipment level) provided by the German Statistical Office (SO-DE, Fachserie 15 Heft 1 in 1962, 1978, 1983 and 1988 for West Germany, every five years in the period 1993-2018 for West and East Germany (data are reported in Figure S12.b), and in 2008 for the 16 German NUTS-1 regions as well as for West and East Berlin (data are reported in Figure S12.c). These data come from the extrapolation of the interview results of household samples. Depending on the years, this sample comprises between 50,000 and 69,000 households, which represents between 0.15 % and 0.25 % of the total number of German households. In the data up to 2008, West Germany includes West Berlin and East Germany includes East Berlin. In the data after 2008 (2013 and 2018), West Germany does not include West Berlin and East Germany includes the whole state of Berlin.

We consider that households started to be equipped with dishwashers in 1963 in West Germany, since the percentage of households having a dishwasher is around 0 % in 1962 and 2 % in 1969. For the years 1969 and 1973, only the percentage of households having a dishwasher is given. Since not information is available regarding the number of households for these two years, we do not use the data for these years. In addition, we consider that households started to be equipped with dishwashers in 1991 in East Germany following the German reunification, since the percentage of households having a dishwasher is only 2.7 % in 1993.

We use the 2008 NUTS-1 level data to disaggregate the totals for West and East Germany, assuming that all NUTS-1 regions that belong to West and East Germany, respectively, followed the same temporal dynamics. Specifically, we disaggregate the data of the number of households having a dishwasher for West Germany for the years for which a dishwasher survey is available (1978, 1983, 1988, 1993, 1998, 2003, 2008, 2013 and 2018) based on the 2008 NUTS-1 level data. Similarly, we disaggregate the data of the number of households having a dishwasher for East Germany for the years 1993, 1998, 2003, 2008, 2013 and 2018, based on the 2008 NUTS-1 level data. The dynamics until 2008 for the state of Berlin are derived for West and East Berlin separately. After 2008, we set the dynamics for the whole state of Berlin to the dynamics for East Germany, since the data do not separate West and East Berlin any more. In addition, the percentage of households having a dishwasher in 2008 is very similar in West and East Berlin (54.5 % and 53.0 % respectively), which suggests that it is sensible to assume a spatially uniform temporal development after 2008 across the whole state of Berlin. The specific equation used to calculate the number of households having a dishwasher, for a year *t* in the period before 2008 in which a dishwasher survey is available and in the i_w-th NUTS-1 region included in the data of West Germany ($N^{dish}_{house,calc}(t,i_w)$) is as follows:

$$N^{dish}_{house,calc}(t,i_w) = N^{dish}_{house,data}(2008,i_w)\frac{N^{dish}_{house,West,data}(t)}{\sum_{ii_w \in E^{1950-2008}_{West}} N^{dish}_{house,data}(2008,ii_w)} for\ t \leq 2008 \tag{S10}$$

where $N^{dish}_{house,West,data}(t)$ is the number of households having an dishwasher in West Germany in year t from the survey data, $N^{dish}_{house,data}(t, i_w)$ is the number of households having a dishwasher in year t and in the i_w-th NUTS-1 region included in the survey data of West Germany from the data, and $E^{1950-2008}_{West}$ is the ensemble of NUTS-1 regions included in the data of West Germany until 2008 (i.e. Schleswig Holstein, Hamburg, Lower Saxony, Bremen, North Rhine-Westphalia, Hesse, Rhineland-Palatinate, Baden-Württemberg, Bavaria, Saarland, and **West Berlin**).

Likewise, for the period 1950-2008, we calculate the number of households having a dishwasher in year t in which a dishwasher survey is available and the i_e-th NUTS-1 region included in the data of East Germany ($N^{dish}_{house,calc}(t, i_e)$) as follows:

$$N^{dish}_{house,calc}(t, i_e) = N^{dish}_{house,data}(2008, i_e) \frac{N^{dish}_{house,East,data}(t)}{\sum_{ii_e \in E^{1950-2008}_{East}} N^{dish}_{house,data}(2008, ii_e)} for\ t \leq 2008 \quad \text{(S11)}$$

where $N^{dish}_{house,East,data}(t)$ is the number of households having an dishwasher in East Germany in year t from the data, $N^{dish}_{house,calc}(t, i_e)$ is the number of households having a dishwasher in year t and in the i_e-th NUTS-1 region included in the data of East Germany, and $E^{1950-2008}_{East}$ is the ensemble of NUTS-1 regions included in the data of East Germany until 2008 (i.e. Brandenburg, Mecklenburg-Vorpommern, Saxony, Saxony-Anhalt, Thuringia, and **East Berlin**).

For the survey years 2013 and 2018, we calculate the number of households having a dishwasher for the j_w-th NUTS-1 region included in the data of West Germany ($N^{dish}_{house,calc}(t, j_w)$) as follows:

$$N^{dish}_{house,calc}(t, j_w) = N^{dish}_{house,calc}(2008, j_w) \frac{N^{dish}_{house,West,data}(t)}{\sum_{jj_w \in E^{2013-2018}_{West}} N^{dish}_{house,calc}(2008, jj_w)} for\ t \geq 2013 \quad \text{(S12)}$$

where $E^{2013-2018}_{West}$ is the ensemble of NUTS-1 regions included in the data of West Germany after 2008 (i.e. Schleswig Holstein, Hamburg, Lower Saxony, Bremen, North Rhine-Westphalia, Hesse, Rhineland-Palatinate, Baden-Württemberg, Bavaria, and Saarland, **excluding West Berlin**).

Likewise, for the survey years 2013 and 2018, we calculate the number of households having a dishwasher for the j_e-th NUTS-1 region included in the data of East Germany ($N^{dish}_{house,calc}(t, j_e)$) as follows:

$$N^{dish}_{house,calc}(t, j_e) = N^{dish}_{house,calc}(2008, j_e) \frac{N^{dish}_{house,East,data}(t)}{\sum_{jj_e \in E^{2013-2018}_{East}} N^{dish}_{house,calc}(2008, jj_e)} for\ t \geq 2013 \quad \text{(S13)}$$

where $E^{2013-2018}_{East}$ is the ensemble of NUTS-1 regions included in the data of East Germany after 2008 (i.e. Brandenburg, Mecklenburg-Vorpommern, Saxony, Saxony-Anhalt, Thuringia, and **Berlin - including East and West Berlin**).

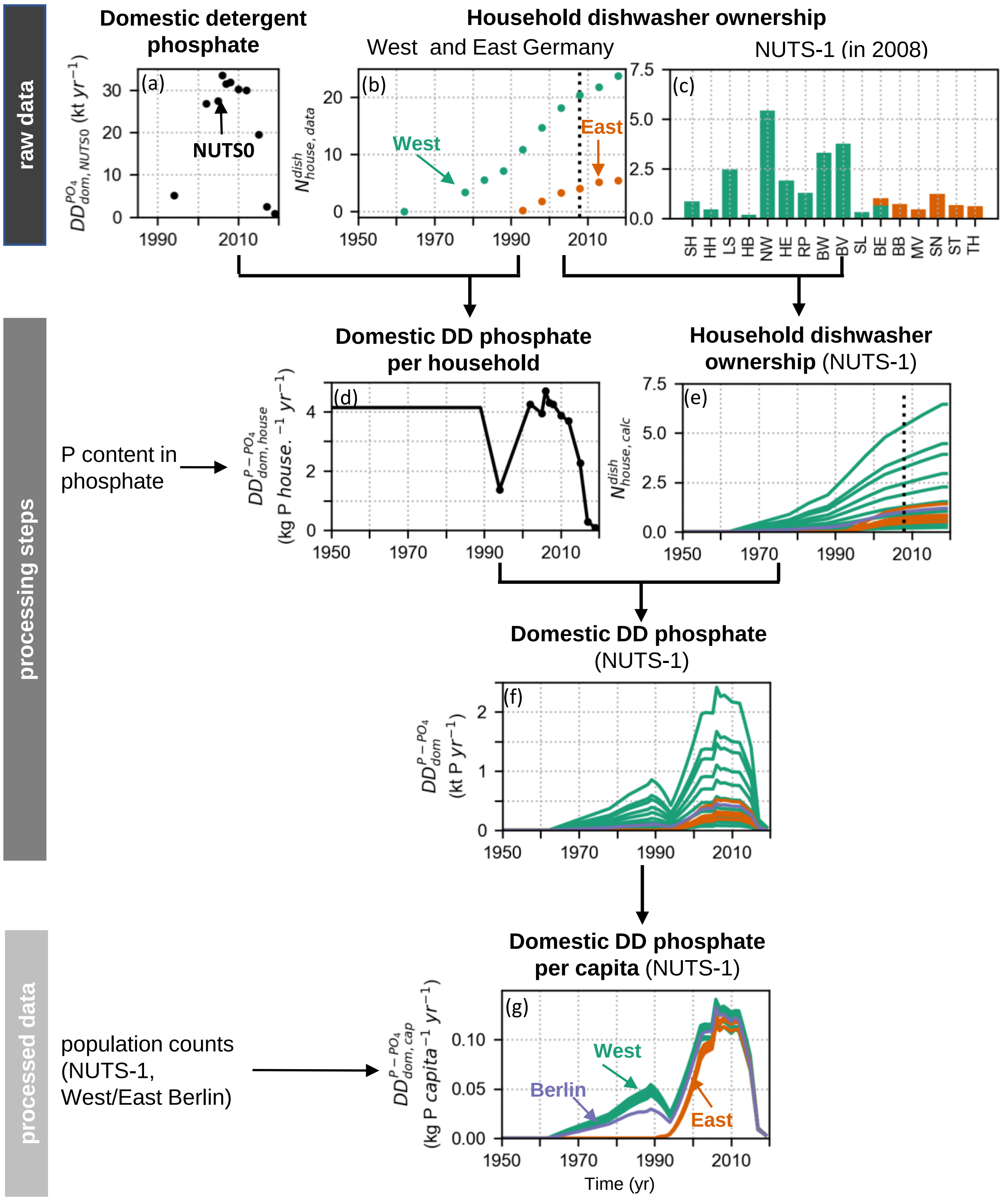
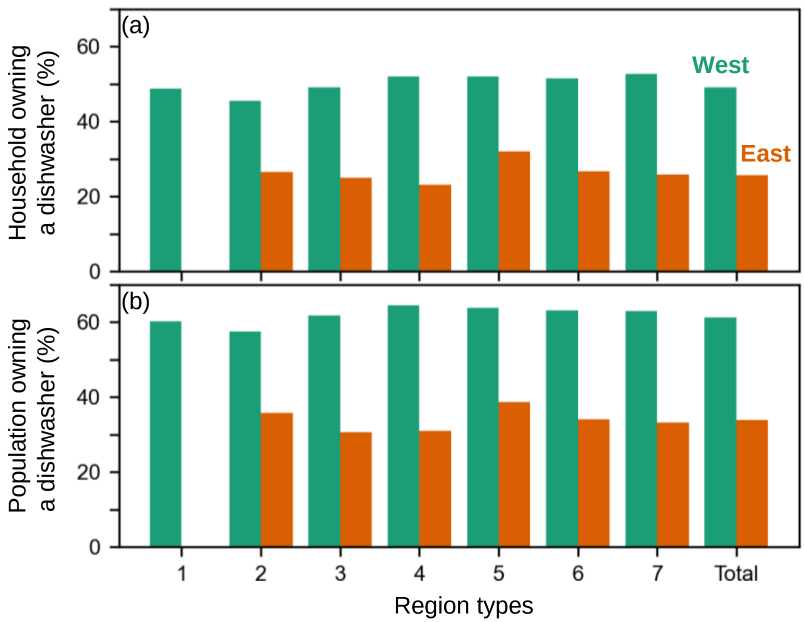


Figure S12. Workflow for the construction of the domestic dishwasher detergent (DD) phosphate data. Data source: (a) detergent data come from ©IKW (2005, 2011, 2017, 2019) (license that does not allow commercial use at https://www.ikw.org/impressum). (b-c) Household data come from the German Statistical Office (SO-DE Fachserie 15 Heft 1). Data in (d-g) build on the data in (a-c).

Equations (S10–S11) ensure that the sum of the calculated NUTS-1 level values in 2008 matches the data for West and East Germany. In fact, we find a 1.5 % and 2.8 % difference between the totals for West and East Germany, respectively, assessed from the NUTS-1 level data in 2008 and the data for West and East Germany (that are directly provided by the Statistical Office). This is likely due to roundoff errors in the provided data.

We then fill the values between the survey years in the period 1950–2019 for the 16 NUTS-1 regions using temporal linear interpolation and we set the 2019 value equal to the 2018 value. The resulting data are shown in Fig. S12.e.

S5.2.3 Household dishwasher ownership in different urban-rural region types



1. High-density agglomeration areas
2. Agglomeration areas with outstanding centers
3. Urbanized areas of higher density
4. Urbanized areas of medium density with major centers of population
5. Urbanized areas of medium density without major centers of population
6. Higher density rural areas
7. Lower density rural areas

Figure S13. Percentage of households (a) and population (b) owning a dishwasher in 1998 in seven region types with different degree of urbanization in West and East German, and aggregated values across the seven region types (total values). We note that they are no regions of type 1 (high-density agglomeration area) in East Germany. Data source: (a) detergent data come from ©IKW (2005, 2011, 2017, 2019) (license that does not allow commercial use at https://www.ikw.org/impressum). (b) . Data in (d-g) build on the data in (a-c).

From household dishwasher ownership data provided for seven region types having different degrees of urbanisation for the year 1998 (German Statistical Office, SO-DE, Fachserie 15 Heft 1), we infer that the degree of urbanisation has a limited impact on the percentage of households owning a dishwasher. In fact, from Figure S13.a, we see that the percentage of households owning a dishwasher varies within a limited range across the seven region types, namely in the range 45–53 % for West Germany and in the range 23–32 % for East Germany.

We also infer that the degree of urbanisation has a limited impact on the percentage of population owning a dishwasher. The population owning a dishwasher is not provided in the data but only the number of households owning a dishwasher. We estimate the population owning a dishwasher from the number of households owning a dishwasher which is provided for five size classes, namely households of one, two, three, four and five or more people. We consider that households in the class with five or more people have 5.3 people, which is the average number of people provided for this class for the years 2008, 2013 and 2018 (SO-DE, Fachserie 15 Heft 1). The resulting percentages of population that belong to a household owning a dishwasher are provided in Figure S13.b, and vary within a limited range across the seven region types, namely in the range 57–65 % for West Germany and in the range 30–39 % for East Germany.

S5.3 Domestic phosphonate detergent data

Figure S14 shows the raw and processed phosphonate detergent data.

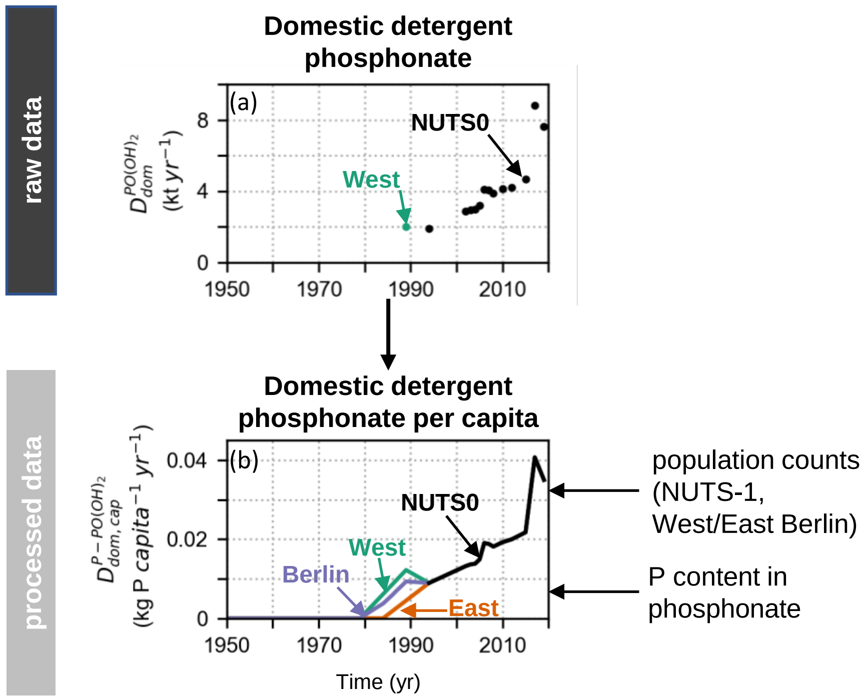


Figure S14. Raw (a) and processed (b) detergent phosphonate data for the domestic sector. From (a), we see that phosphonate detergent use is equal to 1900 $t\ yr^{-1}$ in 1994, around 3000 $t\ yr^{-1}$ in the period 2002-2005, around 4000 $t\ yr^{-1}$ in the period 2006-2015 and around 7000-8000 $t\ yr^{-1}$ in the period 2017-2019, which represents almost ten times the household detergent phosphate amount in 2019 (equal to 829 $t\ yr^{-1}$). In fact, in the 1990s, phosphonate was added in relation to the decrease in EDTA use in detergents after 1991, and ultimately to the production of EDTA-free detergents (IKW, 2019). Phosphonate increased from 2006 are a consequence of the increase in cold-water laundry detergent (IKW, 2009) and after 2015 of the decrease in the phosphate content in dishwasher detergents (IKW, 2019, 2021).(a) Raw data (domestic detergent phosphonate, $DD_{dom}^{PO(OH)_2}$) come from ©IKW (2005, 2011, 2017, 2019) (license that does not allow commercial use at https://www.ikw.org/impressum) and are provided for West Germany in 1989 and for Germany at NUTS-0 level for the period 1994-2019. (b) processed data for West, East and Berlin (domestic detergent phosphonate P per capita, $D_{dom,cap}^{PO(OH)_2-P}$) that build on the IKW data in (a), NUTS-1 level population data from the German Statistical Office and population data for West/East Berlin from the Statistical Office of Berlin (details on population data sources in Table 2 in the main text).

S6 Data of population connection to sewer and wastewater treatment at NUTS-1 level

Table S10 provides an overview of the population connection data availability at NUTS-1 level, while details on data sources and availability are reported in Tables S11–S26 (Sect. S6.1.1) for connection to the sewer system and total wastewater treatment, and Tables S27–S43 (Sect. S6.1.2) for connection to wastewater treatment for the different treatment types. The data mostly come from the Statistical Office of Germany and the different federal states. The notations are defined in Tables S8-S9 and the data can be visualised in Fig. S16–S19.

Tables S27–S43 document the type of data that we used to estimate the connection of the resident population of each NUTS-1 region to the different treatment types. Three categories of data were used, namely (1) **connection of the resident population** of each NUTS-1 region to the different treatment types, (2) **connection of the population treated** in each NUTS-1 region to the different treatment types, and (3) **volume of wastewater treated** in each NUTS-1 region following the different treatment types. When data belong the category (1) above, no further processing was needed and the raw data were directly adopted. For data of category (2), which regards most of the data, we consider that the same relative proportions of the different treatment types apply to the resident population. Exceptions are the states of Berlin and Brandenburg, since a large part of the population of Berlin is connected to WWTPs located in Brandenburg (35–65 % of the population over the period 1991–2019). Therefore, for Berlin, we estimate separately the population connection to WWTPs located in Berlin and Brandenburg, as detailed in Tables S37–S38 and Figure S19. From data of category (3), we derive temporal changes for the population connection to the different treatment types, assuming that the temporal changes in the fraction of population connected and in the volume treated are similar.

Table S8. Notations for the population connection variables used in Tables S10–S43 and in Figures S16–S20

Variable	Description	Unit
$sewer_{all}(t,i)$	fraction of resident population of NUTS-1 region *i* connected to the sewer system in year *t*	(-)
$wwtp_{tot,all}(t,i)$	fraction of resident population of NUTS-1 region *i* connected to wastewater treatment via the sewer system and cesspits in year *t* ($wwtp_{sewer,all}(t,i) + wwtp_{cesspit,all}(t,i)$), including foreign and industrial WWTPs over the period 1998–2019	(-)
$wwtp_{sewer,all}(t,i)$	fraction of resident population of NUTS-1 region *i* connected to the sewer system and to public wastewater treatment in year *t*, including foreign and industrial WWTPs for which data are provided over the period 1998–2019	(-)
$wwtp_{sewer,all}^{ind-for}(t,i)$	fraction of resident population of NUTS-1 region *i* connected to industrial and foreign WWTPs via the sewer system in year *t*	(-)
$wwtp_{cesspit,all}(t,i)$	fraction of resident population of NUTS-1 region *i* whose wastewater is collected in cesspit and transported to WWTPs in year *t*	(-)
$NoCon_{all}(t,i)$	fraction of resident population that is not connected to the sewer system nor to WWTPs	(-)
$T0_{all}(t,i)$	fraction of resident population of NUTS-1 region *i* connected to the sewer system but not to wastewater treatment in year *t*	(-)
$TFarm_{all}(t,i)$	fraction of resident population of NUTS-1 region *i* connected to the sewer system and sewage farms in year *t*	(-)
$T1_{all}(t,i)$	fraction of resident population of NUTS-1 region *i* connected to primary (mechanical) wastewater treatment in year *t*	(-)
$T2_{all}(t,i)$	fraction of resident population of NUTS-1 region *i* connected to secondary (biological) wastewater treatment in year *t*	(-)
$T3_{all}(t,i)$	fraction of resident population of NUTS-1 region *i* connected to tertiary (advanced) wastewater treatment in year *t*	(-)
$T23_{all}^{noN}(t,i)$	fraction of resident population of NUTS-1 region *i* connected to secondary or tertiary wastewater treatment without targeted denitrification in year *t*	(-)
$T23_{all}^{noP}(t,i)$	fraction of resident population of NUTS-1 region *i* connected to secondary or tertiary wastewater treatment without targeted P removal in year *t*	(-)
$T3_{all}^{N}(t,i)$	fraction of resident population of NUTS-1 region *i* connected to tertiary wastewater treatment with targeted denitrification in year *t*	(-)
$T3_{all}^{P}(t,i)$	fraction of resident population of NUTS-1 region *i* connected to tertiary wastewater treatment with targeted P removal in year *t*	(-)

Table S9. Notations for the total population connection variables for Berlin (BE) used in Tables S37–S38.

Variable	Description	Unit
$wwtp^{BE}_{sewer,all}(t, BE)$	fraction of resident population of Berlin connected to wastewater treatment (WWTPs **located in Berlin**) in year *t*	(-)
$wwtp^{BB}_{sewer,all}(t, BE)$	fraction of resident population of Berlin connected to wastewater treatment (WWTPs **located in Brandenburg**) in year *t*	(-)
$T1^{BE}_{all}(t, BE)$	fraction of resident population of Berlin connected to primary (mechanical) wastewater treatment **in Berlin** in year *t*	(-)
$T1^{BB}_{all}(t, BE)$	fraction of resident population of Berlin connected to primary (mechanical) wastewater treatment **in Brandenburg** in year *t*	(-)
$T2^{BE}_{all}(t, BE)$	fraction of resident population of Berlin connected to secondary (biological) wastewater treatment **in Berlin** in year *t*	(-)
$T2^{BB}_{all}(t, BE)$	fraction of resident population Berlin connected to secondary (biological) wastewater treatment **in Brandenburg** in year *t*	(-)
$T3^{BE}_{all}(t, BE)$	fraction of resident population of Berlin connected to tertiary wastewater treatment **in Berlin** in year *t*	(-)
$T3^{BB}_{all}(t, BE)$	fraction of resident population of Berlin connected to tertiary wastewater treatment **in Brandenburg** in year *t*	(-)
$T23^{noN,BE}_{all}(t, BE)$	fraction of resident population of Berlin connected to secondary or tertiary wastewater treatment without targeted denitrification **in Berlin** in year *t*	(-)
$T23^{noN,BB}_{all}(t, BE)$	fraction of resident population of Berlin connected to secondary or tertiary wastewater treatment without targeted denitrification **in Brandenburg** in year *t*	(-)
$T23^{noP,BE}_{all}(t, BE)$	fraction of resident population of Berlin connected to secondary or tertiary wastewater treatment without targeted P removal **in Berlin** in year *t*	(-)
$T23^{noP,BB}_{all}(t, BE)$	fraction of resident population of Berlin connected to secondary or tertiary wastewater treatment without targeted P removal **in Brandenburg** in year *t*	(-)
$T3^{N,BE}_{all}(t, BE)$	fraction of resident population of Berlin connected to tertiary wastewater treatment with targeted denitrification **in Berlin** in year *t*	(-)
$T3^{N,BB}_{all}(t, BE)$	fraction of resident population of Berlin connected to tertiary wastewater treatment with targeted denitrification **in Brandenburg** in year *t*	(-)
$T3^{P,BE}_{all}(t, BE)$	fraction of resident population of Berlin connected to tertiary wastewater treatment with targeted P removal **in Berlin** in year *t*	(-)
$T3^{P,BB}_{all}(t, BE)$	fraction of resident population of Berlin connected to tertiary wastewater treatment with targeted P removal **in Brandenburg** in year *t*	(-)

S6.1 Availability and sources of the population connection data

Table S10. First and last years for which data of total population connection are available for the different NUTS-1 regions.

NUTS-1 region (i)	$sewer_{all}(t,i)$	$wwtp_{sewer,all}$ (t,i)	$wwtp^{ind-for}_{sewer,all}$ (t,i)	$wwtp_{cesspit,all}$ (t,i)	$T1_{all}(t,i)$	$T2_{all}(t,i)$; $T3_{all}(t,i)$	$T23^{noN}_{all}(t,i)$; $T3^{N}_{all}(t,i)$	$T23^{noP}_{all}(t,i)$; $T3^{P}_{all}(t,i)$
SH	1975–2019	1975–2019	1998–2019	n.a.	1975–2019	1975–2019	1987–2019	1987–2019
HH	1975–2019	1975–2019	1998–2019	n.a.	1975–2019	1975–2019	1975–2019	1975–2019
LS	1975–2019	1948–2019	1998–2019	2013–2019	1975–2019	1975–2019	1987–2019	1987–2019
HB	1975–2019	1975–2019	1998–2019	n.a.	1975–2019	1975–2019	1975–2019	1975–2019
NW	1969–2019	1969–2019	1998–2019	n.a.	1969–2019	1975–2016	1987–2019	1987–2019
HE	1975–2019	1975–2019	1998–2019	n.a.	1975–2019	1975–2019	1987–2019	1987–2019
RP	1963–2019	1963–2019	1998–2019	2001–2019	1963–2019	1975–2019	1991–2019	1987–2019
BW	1963–2019	1963–2019	1998–2019	n.a.	1975–2019	1955–2019[a]	1955–2019[a]	1955–2019[a]
BV	1975–2019	1947–2019	1998–2019	2013–2016	1975–2016	1975–2016	1987–2016	1987–2016
SL	1975–2019	1975–2019	1998–2019	n.a.	1975–2019	1975–2019	1975–2019	1975–2019
BE	1975–2019[b]	1975–2019[b]	1998–2019	n.a.	1991–2019[c]	1991–2019[c]	1991–2019[c]	1991–2019[c]
BB	1991–2019	1991–2019	1998–2019	1998–2013	1991–2019	1991–2019	1991–2019	1991–2019
MV	1991–2019	1991–2019	1998–2019	1998–2016	1991–2016	1991–2016	1995–2016	1991–2016
SN	1991–2019	1990–2019	1991–2019	n.a.	1991–2019	1991–2019	1995–2019	1995–2019
ST	1991–2019	1991–2019	1998–2019	2013–2019	1991–2019	1991–2019	1991–2019	1991–2019
TH	1991–2019	1991–2019	1998–2019	n.a.	1991–2019	1991–2019	1991–2019	1991–2019

The variables are defined in Table S8. Details on data availability are reported in Tables S11–S43. The abbreviations for the NUTS-1 regions are defined in Table S1.
n.a.: not available.
[a] Only information on $T3_{all}(t,i)$, $T3^{N}_{all}(t,i)$, and $T3^{P}_{all}(t,i)$ is available.
[b] Data for the period 1975–1990 for BE is provided for West BE only. Given that $sewer_{all}(t,BE)$ and $wwtp_{sewer_all}(t,BE)$ take similar values for West and East Berlin in 1991 (0.98 and 0.95, respectively; SO-DE, 1995), we assume that these two quantities follow the same temporal dynamics in West and East Berlin before 1991. We estimate the values for total Berlin before 1991 by applying the temporal changes of West Berlin to the values for total Berlin provided in 1991.
[c] The connection of the resident population of Berlin to the different treatment types is estimated for the population connected to WWTPs located in Berlin and Brandenburg, as detailed in Tables S37–S38.

S6.1.1 Availability and sources of the data of total population connection to sewer and wastewater treatment at NUTS-1 level

Table S11. Source of the data of total resident population connection to sewer and wastewater treatment in **Schleswig-Holstein (SH)**

t	$sewer_{all}(t, SH)$	$wwtp_{sewer,all}(t, SH)$	$wwtp^{ind-for}_{sewer,all}(t, SH)$	$wwtp_{cesspit,all}(t, SH)$
1975	SO-WDE (1979)	SO-WDE (1979)	n.a.	n.a.
1979	SO-WDE (1983)	SO-WDE (1983)	n.a.	n.a.
1983	SO-WDE (1986), SO-SH (2001)	SO-WDE (1986), SO-SH (2001)	n.a.	n.a.
1987	SO-WDE (1990), SO-SH (2001)	SO-WDE (1990), SO-SH (2001)	n.a.	n.a.
1991	SO-DE (1995), SO-SH (2001)	SO-DE (1995), SO-SH (2001)	n.a.	n.a.
1995	SO-DE (1998), SO-SH (2001)	SO-DE (1998), SO-SH (2001)	n.a.	n.a.
1998	SO-DE (2001), SO-SH (2001)	SO-DE (2001), SO-SH (2001)	SO-DE (2001)[a]	n.a.
2001	SO-DE (2003), SO-SH (2003)	SO-DE (2003), SO-SH (2003)	SO-DE (2003)	n.a.
2004	SO-DE (2006), SO-HH&SH (2006b)	SO-DE (2006), SO-HH&SH (2006b)	SO-DE (2006)	n.a.
2007	SO-DE (2009), SO-HH&SH (2009b)	SO-DE (2009), SO-HH&SH (2009b)	SO-DE (2009)	n.a.
2010	SO-DE (2013), SO-HH&SH (2012b)	SO-DE (2013), SO-HH&SH (2012b)	SO-DE (2013)	n.a.
2013	SO-DE (2015), SO-HH&SH (2015b)	SO-DE (2015), SO-HH&SH (2015b)	SO-DE (2015)	n.a.
2016	SO-DE (2018), SO-HH&SH (2018b)	SO-DE (2018), SO-HH&SH (2018b)	SO-DE (2018)	n.a.
2019	SO-DE (2022), SO-HH&SH (2021b)	SO-DE (2022), SO-HH&SH (2021b)	SO-DE (2022)	n.a.

Notes: The variables are defined in Table S8.

SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany (data link:
https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694;
https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);
SO-SH: Statistical Office of Schleswig-Holstein; SO-HH&SH: Statistical Office of Hamburg and Schleswig-Holstein (data link:
https://www.statistischebibliothek.de/mir/receive/SHSerie_mods_00000083).

n.a.: not available.

[a] population connected to industrial WWTPs only.

Table S12. Source of the data of total resident population connection to sewer and wastewater treatment in **Hamburg (HH)**

t	$sewer_{all}(t, HH)$	$wwtp_{sewer,all}(t, HH)$	$wwtp^{ind-for}_{sewer,all}(t, HH)$	$wwtp_{cesspit,all}(t, HH)$
1975	SO-WDE (1979)	SO-WDE (1979)	n.a.	n.a.
1979	SO-WDE (1983)	SO-WDE (1983)	n.a.	n.a.
1983	SO-WDE (1986)	SO-WDE (1986)	n.a.	n.a.
1987	SO-WDE (1990)	SO-WDE (1990)	n.a.	n.a.
1991	SO-DE (1995)	SO-DE (1995)	n.a.	n.a.
1995	SO-DE (1998)	SO-DE (1998)	n.a.	n.a.
1998	SO-DE (2001)	SO-DE (2001)	SO-DE (2001)[a]	n.a.
2001	n.a.[b]	n.a.[b]	SO-DE (2003)	n.a.
2004	SO-DE (2006), SO-HH&SH (2006a)	SO-DE (2006), SO-HH&SH (2006a)	SO-DE (2006)	n.a.
2007	SO-DE (2009), SO-HH&SH (2009a)	SO-DE (2009), SO-HH&SH (2009a)	SO-DE (2009)	n.a.
2010	SO-DE (2013), SO-HH&SH (2012a)	SO-DE (2013), SO-HH&SH (2012a)	SO-DE (2013)	n.a.
2013	SO-DE (2015), SO-HH&SH (2015a)	SO-DE (2015), SO-HH&SH (2015a)	SO-DE (2015)	n.a.
2016	SO-DE (2018), SO-HH&SH (2018a)	SO-DE (2018), SO-HH&SH (2018a)	SO-DE (2018)	n.a.
2019	SO-DE (2022), SO-HH&SH (2021a)	SO-DE (2022), SO-HH&SH (2021a)	SO-DE (2022)	n.a.

Notes: The variables are defined in Table S8.

SO-WDE: Statistical office of West Germany; SO-DE: Statistical office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-HH&SH: statistical office of Hamburg-Schleswig-Holstein (https://www.statistischebibliothek.de/mir/receive/HHSerie_mods_00000126).

n.a.: not available

[a] population connected to industrial WWTPs only.

[b] Data of $sewer_{all}$ and $wwtp_{sewer,all}$ are available in SO-DE (2003) but we do not use them as they seem to be erroneous. In fact, while $sewer_{all}$ and $wwtp_{sewer,all}$ are both equal to 98.47 % in 1998 and to 98.9 % in 2004, the data for 2001 (in SO-DE, 2003) indicate that they are both equal to 100 %.

Table S13. Source of the data of total resident population connection to sewer and wastewater treatment in **Lower Saxony (LS)**

t	$sewer_{all}(t, LS)$	$wwtp_{sewer,all}(t, LS)$	$wwtp^{ind-for}_{sewer,all}(t, LS)$	$wwtp_{cesspit,all}(t, LS)$
1948	n.a.	NLWKM (2021, Fig. 1)	n.a.	n.a.
1970	n.a.	NLWKM (2021, Fig. 1)	n.a.	n.a.
1975	SO-WDE (1979)	SO-WDE (1979)	n.a.	n.a.
1979	SO-WDE (1983)	SO-WDE (1983)	n.a.	n.a.
1983	SO-WDE (1986)	SO-WDE (1986)	n.a.	n.a.
1987	SO-WDE (1990)	SO-WDE (1990)	n.a.	n.a.
1991	SO-DE (1995)	SO-DE (1995)	n.a.	n.a.
1995	SO-DE (1998)	SO-DE (1998)	n.a.	n.a.
1998	SO-DE (2001)	SO-DE (2001)	SO-DE (2001)[a]	n.a.
2001	SO-DE (2003), SO-LS (2003)	SO-DE (2003), SO-LS (2003)	SO-DE (2003)	n.a.
2004	SO-DE (2006), SO-LS (2009)	SO-DE (2006), SO-LS (2009)	SO-DE (2006)	n.a.
2007	SO-DE (2009), SO-LS (2010)	SO-DE (2009), SO-LS (2010)	SO-DE (2009)	n.a.
2010	SO-DE (2013), SO-LS (2014)	SO-DE (2013), SO-LS (2014)	SO-DE (2013)	n.a.
2013	SO-DE (2015), SO-LS (2019a)	SO-DE (2015), SO-LS (2019a)	SO-DE (2015)	SO-LS (2019a)
2016	SO-DE (2018), SO-LS (2019b)	SO-DE (2018), SO-LS (2019b)	SO-DE (2018)	SO-LS (2019b)
2019	SO-DE (2022), SO-LS (2022)	SO-DE (2022), SO-LS (2022)	SO-DE (2022)	SO-LS (2022)

Notes: The variables are defined in Table S8.

SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-LS: Statistical Office of Lower Saxony (https://www.statistischebibliothek.de/mir/receive/NISerie_mods_00000189).

n.a.: not available

[a] population connected to industrial WWTPs only.

Table S14. Source of the data of total resident population connection to sewer and wastewater treatment in **Bremen (HB)**

t	$sewer_{all}(t, HB)$	$wwtp_{sewer,all}(t, HB)$	$wwtp^{ind-for}_{sewer,all}(t, HB)$	$wwtp_{cesspit,all}(t, HB)$
1975	SO-WDE (1979)	SO-WDE (1979)	n.a.	n.a.
1979	SO-WDE (1983)	SO-WDE (1983)	n.a.	n.a.
1983	SO-WDE (1986)	SO-WDE (1986)	n.a.	n.a.
1987	SO-WDE (1990)	SO-WDE (1990)	n.a.	n.a.
1991	SO-DE (1995)	SO-DE (1995)	n.a.	n.a.
1995	SO-DE (1998)	SO-DE (1998)	n.a.	n.a.
1998	SO-DE (2001)	SO-DE (2001)	SO-DE (2001)[a]	n.a.
2001	SO-DE (2003)	SO-DE (2003)	SO-DE (2003)	n.a.
2004	SO-DE (2006)	SO-DE (2006)	SO-DE (2006)	n.a.
2007	SO-DE (2009)	SO-DE (2009)	SO-DE (2009)	n.a.
2010	SO-DE (2013)	SO-DE (2013)	SO-DE (2013)	n.a.
2013	SO-DE (2015)	SO-DE (2015)	SO-DE (2015)	n.a.
2016	SO-DE (2018)	SO-DE (2018)	SO-DE (2018)	n.a.
2019	SO-DE (2022)	SO-DE (2022)	SO-DE (2022)	n.a.

Notes: The variables are defined in Table S8.

SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B).

n.a.: not available.

[a] population connected to industrial WWTPs only.

Table S15. Source of the data of total resident population connection to sewer and wastewater treatment in **North Rhine-Westphalia (NW)**

Year	$sewer_{all}(t, NW)$	$wwtp_{sewer,all}(t, NW)$	$wwtp_{sewer,all}^{ind-for}(t, NW)$	$wwtp_{cesspit,all}(t, NW)$
1969	SO-NW (2001)	SO-NW (2001)	n.a.	n.a.
1975	SO-WDE (1979), SO-NW (2001)	SO-WDE (1979), SO-NW (2001)	n.a.	n.a.
1979	SO-WDE (1983), SO-NW (2001)	SO-WDE (1983)	n.a.	n.a.
1983	SO-WDE (1986), SO-NW (2001)	SO-WDE (1986), SO-NW (2001)	n.a.	n.a.
1987	SO-WDE (1990), SO-NW (2001)	SO-WDE (1990), SO-NW (2001)	n.a.	n.a.
1991	SO-DE (1995), SO-NW (2001)	SO-DE (1995), SO-NW (2001)	n.a.	n.a.
1995	SO-DE (1998), SO-NW (2001)	SO-DE (1998), SO-NW (2001)	n.a.	n.a.
1998	SO-DE (2001), SO-NW (2001)	SO-DE (2001), SO-NW (2001)	SO-DE (2001)[a]	n.a.
2001	SO-DE (2003), SO-NW (2003)	SO-DE (2003), SO-NW (2003)	SO-DE (2003)	n.a.
2004	SO-DE (2006), SO-NW (2006)	SO-DE (2006), SO-NW (2006)	SO-DE (2006)	n.a.
2007	SO-DE (2009), SO-NW (2009)	SO-DE (2009), SO-NW (2009)	SO-DE (2009)	n.a.
2010	SO-DE (2013), SO-NW (2013)	SO-DE (2013), SO-NW (2013)	SO-DE (2013)	n.a.
2013	SO-DE (2015), SO-NW (2016)	SO-DE (2015), SO-NW (2016)	SO-DE (2015)	n.a.
2016	SO-DE (2018), SO-NW (2018)	SO-DE (2018), SO-NW (2018)	SO-DE (2018)	n.a.
2019	SO-DE (2022)	SO-DE (2022)	SO-DE (2022)	n.a.

Notes: The variables are defined in Table S8.
SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany
(https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694;
https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);
SO-NW: Statistical Office of North Rhine-Westphalia (https://www.statistischebibliothek.de/mir/receive/NWSerie_mods_00000171)
n.a.: not available.
[a] population connected to industrial WWTPs only.

Table S16. Source of the data of total resident population connection to sewer and wastewater treatment in **Hesse (HE)**

Year	$sewer_{all}(t, HE)$	$wwtp_{sewer,all}(t, HE)$	$wwtp^{ind-for}_{sewer,all}(t, HE)$	$wwtp_{cesspit,all}(t, HE)$
1975	SO-WDE (1979)	SO-WDE (1979)	n.a.	n.a.
1979	SO-WDE (1983)	SO-WDE (1983)	n.a.	n.a.
1983	SO-WDE (1986)	SO-WDE (1986)	n.a.	n.a.
1987	SO-WDE (1990)	SO-WDE (1990)	n.a.	n.a.
1991	SO-DE (1995)	SO-DE (1995)	n.a.	n.a.
1995	SO-DE (1998)	SO-DE (1998)	n.a.	n.a.
1998	SO-DE (2001)	SO-DE (2001)	SO-DE (2001)[a]	n.a.
2001	SO-DE (2003), SO-HE (2016)	SO-DE (2003), SO-HE (2016)	SO-DE (2003)	n.a.
2004	SO-DE (2006)	SO-DE (2006)	SO-DE (2006)	n.a.
2007	SO-DE (2009), SO-HE (2009)	SO-DE (2009), SO-HE (2009)	SO-DE (2009)	n.a.
2010	SO-DE (2013), SO-HE (2012)	SO-DE (2013), SO-HE (2012)	SO-DE (2013)	n.a.
2013	SO-DE (2015), SO-HE (2015)	SO-DE (2015), SO-HE (2015)	SO-DE (2015)	n.a.
2016	SO-DE (2018), SO-HE (2018)	SO-DE (2018), SO-HE (2018)	SO-DE (2018)	n.a.
2019	SO-DE (2022), SO-HE (2022)	SO-DE (2022), SO-HE (2022)	SO-DE (2022)	n.a.

Notes: The variables are defined in Table S8.

SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-HE: Statistical Office of Hesse (https://www.statistischebibliothek.de/mir/receive/HESerie_mods_00000228).

n.a.: not available

[a] population connected to industrial WWTPs only.

Table S17. Source of the data of total resident population connection to sewer and wastewater treatment in **Rhineland-Palatinate (RP)**

t	$sewer_{all}(t, RP)$	$wwtp_{sewer,all}(t, RP)$	$wwtp^{ind-for}_{sewer,all}(t, RP)$	$wwtp_{cesspit,all}(t, RP)$
1963	SO-RP (2006)	SO-RP (2006)	n.a.	n.a.
1969	SO-RP (2006)	SO-RP (2006)	n.a.	n.a.
1975	SO-WDE (1979), SO-RP (2006)	SO-WDE (1979), SO-RP (2006)	n.a.	n.a.
1979	SO-WDE (1983), SO-RP (2006)	SO-WDE (1983), SO-RP (2006)	n.a.	n.a.
1983	SO-WDE (1986), SO-RP (2006)	SO-WDE (1986), SO-RP (2006)	n.a.	n.a.
1987	SO-WDE (1990), SO-RP (2006)	SO-WDE (1990), SO-RP (2006)	n.a.	n.a.
1991	SO-DE (1995), SO-RP (2006)	SO-DE (1995), SO-RP (2006)	n.a.	n.a.
1995	SO-DE (1998), SO-RP (2006)	SO-DE (1998), SO-RP (2006)	n.a.	n.a.
1998	SO-DE (2001), SO-RP (2006)	SO-DE (2001), SO-RP (2006)	SO-DE (2001)[a]	n.a.
2001	SO-DE (2003), SO-RP (2003)	SO-DE (2003), SO-RP (2003)	SO-DE (2003)	SO-RP (2003)
2004	SO-DE (2006), SO-RP (2006)	SO-DE (2006), SO-RP (2006)	SO-DE (2006)	SO-RP (2006)
2007	SO-DE (2009), SO-RP (2009)	SO-DE (2009), SO-RP (2009)	SO-DE (2009)	SO-RP (2019)
2010	SO-DE (2013), SO-RP (2012)[b]	SO-DE (2013), SO-RP (2012)[b]	SO-DE (2013)	SO-RP (2013)
2013	SO-DE (2015), SO-RP (2015)	SO-DE (2015), SO-RP (2015)	SO-DE (2015)	SO-RP (2015)
2016	SO-DE (2018), SO-RP (2018)	SO-DE (2018), SO-RP (2018)	SO-DE (2018)	SO-RP (2018)
2019	SO-DE (2022), SO-RP (2022)	SO-DE (2022), SO-RP (2022)	SO-DE (2022)	SO-RP (2022)

Notes: The variables are defined in Table S8.

SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany

(https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694;

https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-RP: Statistical Office of Rhineland-Palatinate https://www.statistischebibliothek.de/mir/receive/RPSerie_mods_00001054.

n.a.: not available.

[a] population connected to industrial WWTPs only.

[b] We adopt the data provided by SO-RP (2012). The data for the year 2010 present small differences between SO-DE (2013) and SO-RP (2012) (0.0036 different for $sewer_{all}$ and 0.0073 difference for $wwtp_{sewer,all}$). It seems that $sewer_{all}$ provided by SO-DE (2013) includes the population connected to WWTPs through cesspits, while the differences in the value of $wwtp_{sewer,all}$ is not clear.

Table S18. Source of the data of total resident population connection to sewer and wastewater treatment in **Baden-Württemberg (BW)**

t	$sewer_{all}(t, BW)$	$wwtp_{sewer,all}(t, BW)$	$wwtp^{ind-for}_{sewer,all}(t, BW)$	$wwtp_{cesspit,all}(t, BW)$
1963	LUBW and MUKEBW (2021, Fig. 1)	LUBW and MUKEBW (2021, Fig. 1)	n.a.	n.a.
1969	LUBW and MUKEBW (2021, Fig. 1)	LUBW and MUKEBW (2021, Fig. 1)	n.a.	n.a.
1975	SO-WDE (1979)	SO-WDE (1979)	n.a.	n.a.
1979	SO-WDE (1983)	SO-WDE (1983)	n.a.	n.a.
1983	SO-WDE (1986)	SO-WDE (1986)	n.a.	n.a.
1987	SO-WDE (1990)	SO-WDE (1990)	n.a.	n.a.
1991	SO-DE (1995)	SO-DE (1995)	n.a.	n.a.
1995	SO-DE (1998)	SO-DE (1998)	n.a.	n.a.
1998	SO-DE (2001)	SO-DE (2001)	SO-DE (2001)[a]	n.a.
2001	SO-DE (2003)	SO-DE (2003)	SO-DE (2003)	n.a.
2004	SO-DE (2006)	SO-DE (2006)	SO-DE (2006)	n.a.
2007	SO-DE (2009)	SO-DE (2009)	SO-DE (2009)	n.a.
2010	SO-DE (2013)	SO-DE (2013)	SO-DE (2013)	n.a.
2013	SO-DE (2015)	SO-DE (2015)	SO-DE (2015)	n.a.
2016	SO-DE (2018)	SO-DE (2018)	SO-DE (2018)	n.a.
2019	SO-DE (2022)	SO-DE (2022)	SO-DE (2022)	n.a.

Notes: The variables are defined in Table S8.

SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany

(https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694;

https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B).

n.a.: not available.

[a] population connected to industrial WWTPs only.

Table S19. Source of the data of total resident population connection to sewer and wastewater treatment in **Bavaria (BV)**

t	$sewer_{all}(t, BV)$	$wwtp_{sewer,all}(t, BV)$	$wwtp^{ind-for}_{sewer,all}(t, BV)$	$wwtp_{cesspit,all}(t, BV)$
1947	n.a.	Schleypen (2017)	n.a.	n.a.
1975	SO-WDE (1979)	SO-WDE (1979)	n.a.	n.a.
1979	SO-WDE (1983), SO-BV (2001)	SO-WDE (1983), SO-BV (2001)	n.a.	n.a.
1983	SO-WDE (1986), SO-BV (2001)	SO-WDE (1986), SO-BV (2001)	n.a.	n.a.
1987	SO-WDE (1990), SO-BV (2001)	SO-WDE (1990), SO-BV (2001)	n.a.	n.a.
1991	SO-DE (1995) , SO-BV (2001)	SO-DE (1995), SO-BV (2001)	n.a.	n.a.
1995	SO-DE (1998), SO-BV (2001)	SO-DE (1998), SO-BV (2001)	n.a.	n.a.
1998	SO-DE (2001), SO-BV (2001)	SO-DE (2001), SO-BV (2001)	SO-DE (2001)[a]	n.a.
2001	SO-DE (2003), SO-BV (2003)	SO-DE (2003), SO-BV (2003)	SO-DE (2003)	n.a.
2004	SO-DE (2006), SO-BV (2006)	SO-DE (2006), SO-BV (2006)	SO-DE (2006)	n.a.
2007	SO-DE (2009), SO-BV (2009)	SO-DE (2009), SO-BV (2009)	SO-DE (2009)	n.a.
2010	SO-DE (2013), SO-BV (2012)	SO-DE (2013), SO-BV (2012)	SO-DE (2013)	n.a.
2013	SO-DE (2015), SO-BV (2015)	SO-DE (2015), SO-BV (2015)	SO-DE (2015)	SO-BV (2015)
2016	SO-DE (2018), SO-BV (2020)	SO-DE (2018), SO-BV (2020)	SO-DE (2018)	SO-BV (2020)
2019	SO-DE (2022)	SO-DE (2022)	SO-DE (2022)	n.a.

Notes: The variables are defined in Table S8.

SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany

(https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694;

https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-BV: Statistical Office of Bavaria (https://www.statistischebibliothek.de/mir/receive/BYSerie_mods_00000808).

n.a.: not available

[a] population connected to industrial WWTPs only.

Table S20. Source of the data of total resident population connection to sewer and wastewater treatment in **Saarland (SL)**

t	$sewer_{all}(t, SL)$	$wwtp_{sewer,all}(t, SL)$	$wwtp^{ind-for}_{sewer,all}(t, SL)$	$wwtp_{cesspit,all}(t, SL)$
1975	SO-WDE (1979)	SO-WDE (1979)	n.a.	n.a.
1979	SO-WDE (1983)	SO-WDE (1983)	n.a.	n.a.
1983	SO-WDE (1986)	SO-WDE (1986)	n.a.	n.a.
1987	SO-WDE (1990)	SO-WDE (1990)	n.a.	n.a.
1991	SO-DE (1995)	SO-DE (1995)	n.a.	n.a.
1995	SO-DE (1998)	SO-DE (1998)	n.a.	n.a.
1998	SO-DE (2001)	SO-DE (2001)	SO-DE (2001)[a]	n.a.
2001	SO-DE (2003)	SO-DE (2003)	SO-DE (2003)	n.a.
2004	SO-DE (2006)	SO-DE (2006)	SO-DE (2006)	n.a.
2007	SO-DE (2009)	SO-DE (2009)	SO-DE (2009)	n.a.
2010	SO-DE (2013)	SO-DE (2013)	SO-DE (2013)	n.a.
2013	SO-DE (2015)	SO-DE (2015)	SO-DE (2015)	n.a.
2016	SO-DE (2018)	SO-DE (2018)	SO-DE (2018)	n.a.
2019	SO-DE (2022)	SO-DE (2022)	SO-DE (2022)	n.a.

Notes: The variables are defined in Table S8.

SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany

(https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694;

https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B).

n.a.: not available

[a] population connected to industrial WWTPs only.

Table S21. Source of the data of total resident population connection to sewer and wastewater treatment in **Berlin (BE)**

t	$sewer_{all}(t, BE)$	$wwtp_{sewer,all}(t, BE)$	$wwtp^{ind-for}_{sewer,all}(t, BE)$	$wwtp_{cesspit,all}$ (t,BE)[b]	$wwtp^{BE}_{tot,all}(t, BE)$, $wwtp^{BB}_{tot,all}(t, BE)$
1975	SO-WDE (1979)[a]	SO-WDE (1979)[a]	n.a.	n.a.	n.a.[b]
1979	SO-WDE (1983)[a]	SO-WDE (1983)[a]	n.a.	n.a.	n.a.[b]
1983	SO-WDE (1986)[a]	SO-WDE (1986)[a]	n.a.	n.a.	n.a.[b]
1987	SO-WDE (1990)[a]	SO-WDE (1990)[a]	n.a.	n.a.	n.a.[b]
1991	SO-DE (1995)	SO-DE (1995)	n.a.	n.a.	SO-DE (1995)
1995	SO-DE (1998)	SO-DE (1998)	n.a.	n.a.	SO-DE (1998)
1998	SO-DE (2001)	SO-DE (2001)	SO-DE (2001)	n.a.	SO-DE (2001)
2001	SO-DE (2003)	SO-DE (2003)	SO-DE (2003)	n.a.	SO-DE (2003)
2004	SO-DE (2006)	SO-DE (2006)	SO-DE (2006)	n.a.	SO-DE (2006)
2007	SO-DE (2009), SO-BE&BB (2010b)	SO-DE (2009)	SO-DE (2009)	n.a	SO-DE (2009)
2010	SO-DE (2013), SO-BE&BB (2013b)	n.a.[d]	SO-DE (2013)	n.a.	SO-BE&BB (2013b)[c]
2013	SO-DE (2015), SO-BE&BB (2017b)	SO-DE (2015), SO-BE&BB (2017b)	SO-DE (2015)	n.a.	SO-BE&BB (2017b)
2016	SO-DE (2018), SO-BE&BB (2019b)	SO-DE (2018), SO-BE&BB (2019b)	SO-DE (2018)	n.a.	SO-BE&BB (2019b)
2019	SO-DE (2022), SO-BE&BB (2022b)	SO-DE (2022), SO-BE&BB (2022b)	SO-DE (2022)	n.a.	SO-BE&BB (2022b)

Notes: The variables are defined in Table S8–S9.

SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-BE&BB: Statistical Office of Berlin and Brandenburg (https://www.statistischebibliothek.de/mir/receive/BBSerie_mods_00000564).

n.a.: not available.

[a] Data for the period 1975–1990 for BE is provided for West BE only. Given that $sewer(t, BE)$ and $wwtp_{sewer_all}(t, BE)$ take similar values for West and East Berlin in 1991 (0.98 and 0.95, respectively; SO-DE, 1995), we assume that these two quantities follow the same temporal dynamics in West and East Berlin before 1991. We estimate the values for total Berlin before 1991 by applying the temporal changes of West Berlin to the values for total Berlin provided in 1991.

[b] In absence of data of population connection to WWTPs located in BE and BB before 1991, we assume that the relative value of $wwtp^{BE}_{tot,all}(t, BE)$ and $wwtp^{BB}_{tot,all}(t, BE)$ is the same before 1991 as in 1991.

[c] We assume that $wwtp_{sewer,all}$ is equal to $sewer_{all}$ in 2010 as in previous and following years.

Table S22. Source of the data of total resident population connection to sewer and wastewater treatment in **Brandenburg (BB)**

t	$sewer_{all}(t, BB)$	$wwtp_{sewer,all}(t, BB)$	$wwtp^{ind-for}_{sewer,all}(t, BB)$	$wwtp_{cesspit,all}(t, BB)^a$
1991	SO-DE (1995), SO-BB (1995)	SO-DE (1995), SO-BB (1995)	n.a.	n.a.
1995	SO-DE (1998), SO-BB (1998)	SO-DE (1998), SO-BB (1998)	n.a.	n.a.
1998	SO-DE (2001), SO-BB (2002)	SO-DE (2001), SO-BB (2002)	SO-DE (2001), SO-BB (2002)[b]	SO-BB (2000)
2001	SO-DE (2003), SO-BB (2004)	SO-DE (2003), SO-BB (2004)	SO-DE (2003)	SO-BB (2004)
2004	SO-DE (2006), SO-BE&BB (2007)	SO-DE (2006), SO-BE&BB (2007)	SO-DE (2006)	SO-BE&BB (2007)
2007	SO-DE (2009), SO-BE&BB (2010a)	SO-DE (2009), SO-BE&BB (2010a)	SO-DE (2009)	SO-BE&BB (2010a)
2010	SO-DE (2013), SO-BE&BB (2013a)[c]	SO-DE (2013), SO-BE&BB (2013a)[c]	SO-DE (2013)	SO-BE&BB (2013a)
2013	SO-DE (2015), SO-BE&BB (2017a)	SO-DE (2015), SO-BE&BB (2017a)	SO-DE (2015)	SO-BE&BB (2017a)
2016	SO-DE (2018), SO-BE&BB (2019a)	SO-DE (2018), SO-BE&BB (2019a)	SO-DE (2018)	n.a.
2019	SO-DE (2022), SO-BE&BB (2022a)	SO-DE (2022), SO-BE&BB (2022a)	SO-DE (2022)	n.a.

Notes: The variables are defined in Table S8.

SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-BB: Statistical Office of Brandenburg; SO-BE&BB: Statistical Office of Berlin and Brandenburg (https://www.statistischebibliothek.de/mir/receive/BBSerie_mods_00000562).

n.a.: not available

[a] $wwtp_{cesspit,all}$ is not directly given but estimated from other data. In 2016 and 2019, no specific information can be recovered regarding the population connected to WWTPs through cesspits. We assume that all population connected to cesspits is also connected to WWTPs, as for the period 2001–2013 more than 98 % of the population connected to cesspits is also connected to WWTPs.

[b] population connected to industrial WWTPs is provided in SO-DE (2001) and population connected to WWTPs located in Poland in SO-BB (2002).

[c] $sewer_{all}$ and $wwtp_{sewer,all}$ in SO-BE&BB (2013) seems to include the population treated in WWTPs through cesspits. Therefore, we adopt the data from SO-DE (2013).

Table S23. Source of the data of total resident population connection to sewer and wastewater treatment in **Mecklenburg-Vorpommern (MV)**

t	$sewer_{all}(t, MV)$	$wwtp_{sewer,all}(t, MV)$	$wwtp^{ind-for}_{sewer,all}(t, MV)$	$wwtp_{cesspit,all}(t, MV)^a$
1991	SO-DE (1995), SO-MV (2013)	SO-DE (1995), SO-MV (2013)	n.a.	n.a.
1995	SO-DE (1998), SO-MV (2013)	SO-DE (1998), SO-MV (2013)	n.a.	n.a.
1998	SO-DE (2001), SO-MV (2013)	SO-DE (2001), SO-MV (2013)	SO-DE (2001)[b]	SO-MV (2019)
2001	SO-DE (2003), SO-MV (2013)	SO-DE (2003), SO-MV (2013)	SO-DE (2003)	SO-MV (2019)
2004	SO-DE (2006), SO-MV (2013)	SO-DE (2006), SO-MV (2013)	SO-DE (2006)	SO-MV (2019)
2007	SO-DE (2009), SO-MV (2013)	SO-DE (2009), SO-MV (2013)	SO-DE (2009)	SO-MV (2019)
2010	SO-DE (2013), SO-MV (2013)	SO-DE (2013), SO-MV (2013)	SO-DE (2013)	SO-MV (2019)
2013	SO-DE (2015), SO-MV (2016)	SO-DE (2015), SO-MV (2016)	SO-DE (2015)	SO-MV (2019)
2016	SO-DE (2018), SO-MV (2019)	SO-DE (2018), SO-MV (2019)	SO-DE (2018)	SO-MV (2019)
2019	SO-DE (2022)	SO-DE (2022)	SO-DE (2022)	n.a.

Notes: The variables are defined in Table S8.

SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-MV: Statistical Office of Mecklenburg-Vorpommern (https://www.statistischebibliothek.de/mir/receive/MVSerie_mods_00000334). n.a.: not available

[a] $wwtp^{public}_{cesspit}$ is not directly given but estimated from other data that are provided, i.e. by calcuting the difference between the total population connection to WWTPs through both sewer and cesspits provided by SO-MV (2019) and the connection to WWTPs through sewer ($wwtp_{sewer,all}$) provided in other statistical reports as indicated in the table.

[b] population connected to industrial WWTPs only.

Table S24. Source of the data of total resident population connection to sewer and wastewater treatment in **Saxony (SN)**

t	$sewer_{all}(t, SN)$	$wwtp_{sewer,all}(t, SN)$	$wwtp_{sewer,all}^{ind-for}(t, SN)$	$wwtp_{cesspit,all}(t, SN)^a$
1990	n.a.	SMUL and LfUG (2004)	n.a.	n.a.
1991	SO-DE (1995), SO-SN (2003)	SO-DE (1995), SO-SN (2003)	SO-SN (2003)	n.a.
1995	SO-DE (1998), SO-SN (2003)	SO-DE (1998), SO-SN (2003)	SO-SN (2003)	n.a.
1998	SO-DE (2001), SO-SN (2003)	SO-DE (2001), SO-SN (2003)	SO-DE (2001), SO-SN (2003)[a]	n.a.
2001	SO-DE (2003), SO-SN (2003)	SO-DE (2003), SO-SN (2003)	SO-DE (2003), SO-SN (2003)	n.a.
2004	SO-DE (2006), SO-SN (2006)	SO-DE (2006), SO-SN (2006)	SO-DE (2006), SO-SN (2006)	n.a.
2007	SO-DE (2009), SO-SN (2009)	SO-DE (2009), SO-SN (2009)	SO-DE (2009), SO-SN (2009)	n.a.
2010	SO-DE (2013), SO-SN (2011)	SO-DE (2013), SO-SN (2011)	SO-DE (2013), SO-SN (2011)	n.a.
2013	SO-DE (2015), SO-SN (2015)	SO-DE (2015), SO-SN (2015)	SO-DE (2015), SO-SN (2015)	n.a.
2016	SO-DE (2018), SO-SN (2018)	SO-DE (2018), SO-SN (2018)	SO-DE (2018), SO-SN (2018)	n.a.
2019	SO-DE (2022), SO-SN (2021)	SO-DE (2022), SO-SN (2021)	SO-DE (2022), SO-SN (2021)	n.a.

Notes: The variables are defined in Table S8.

SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-SN: Statistical Office of Saxony (https://www.statistischebibliothek.de/mir/receive/SNSerie_mods_00001093).

n.a.: not available

[a] population connected to industrial WWTPs only.

Table S25. Source of the data of total resident population connection to sewer and wastewater treatment in **Saxony-Anhalt (ST)**

t	$sewer_{all}(t, ST)$	$wwtp_{sewer,all}(t, ST)$	$wwtp^{ind-for}_{sewer,all}(t, ST)$	$wwtp_{cesspit,all}(t, ST)$
1991	SO-DE (1995)	SO-DE (1995)	n.a.	n.a.
1995	SO-DE (1998), SO-ST (2010)	SO-DE (1998), SO-ST (2010)	n.a.	n.a.
1998	SO-DE (2001), SO-ST (2010)	SO-DE (2001), SO-ST (2010)	SO-DE (2001)[a]	n.a.
2001	SO-DE (2003), SO-ST (2010)	SO-DE (2003), SO-ST (2010)	SO-DE (2003)	n.a.
2004	SO-DE (2006), SO-ST (2010)	SO-DE (2006), SO-ST (2010)	SO-DE (2006)	n.a.
2007	SO-DE (2009), SO-ST (2010)	SO-DE (2009), SO-ST (2010)	SO-DE (2009)	n.a.
2010	SO-DE (2013), SO-ST (2012)[b]	SO-DE (2013), SO-ST (2012)[b]	SO-DE (2013)	n.a.
2013	SO-DE (2015), SO-ST (2016)	SO-DE (2015), SO-ST (2016)	SO-DE (2015)	SO-ST (2016)
2016	SO-DE (2018), SO-ST (2018)	SO-DE (2018), SO-ST (2018)	SO-DE (2018)	SO-ST (2018)
2019	SO-DE (2022), SO-ST (2022)	SO-DE (2022), SO-ST (2022)	SO-DE (2022)	SO-ST (2022)

Notes: The variables are defined in Table S8.

SO-DE: Statistical office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-ST: statistical office of Saxony-Anhalt (https://www.statistischebibliothek.de/mir/receive/STSerie_mods_00000324).

n.a.: not available.

[a] population connected to industrial WWTPs only.

[b] The data for the year 2010 present small differences between SO-DE (2013) and SO-ST (2012) (0.28 % different for $sewer_{all}$ and for $wwtp_{sewer,all}$). We adopt the values in SO-ST (2012).

Table S26. Source of the data of total resident population connection to sewer and wastewater treatment in **Thuringia (TH)**

t	$sewer_{all}(t, TH)$	$wwtp_{sewer,all}(t, TH)$	$wwtp^{ind-for}_{sewer,all}(t, TH)$	$wwtp_{cesspit,all}(t, TH)$
1991	SO-DE (1995), SO-TH (2006)	SO-DE (1995), SO-TH (2006)	n.a.	n.a.
1995	SO-DE (1998), SO-TH (2006)	SO-DE (1998), SO-TH (2006)	n.a.	n.a.
1998	SO-DE (2001), SO-TH (2006)	SO-DE (2001), SO-TH (2006)	SO-DE (2001)[a]	n.a.
2001	SO-DE (2003), SO-TH (2006)	SO-DE (2003), SO-TH (2006)	SO-DE (2003)	n.a.
2004	SO-DE (2006), SO-TH (2006)	SO-DE (2006), SO-TH (2006)	SO-DE (2006)	n.a.
2007	SO-DE (2009), SO-TH (2009)	SO-DE (2009), SO-TH (2009)	SO-DE (2009)	n.a.
2010	SO-DE (2013), SO-TH (2012)	SO-DE (2013), SO-TH (2012)	SO-DE (2013)	n.a.
2013	SO-DE (2015), SO-TH (2015)	SO-DE (2015), SO-TH (2015)	SO-DE (2015)	n.a.
2016	SO-DE (2018), SO-TH (2019)	SO-DE (2018), SO-TH (2019)	SO-DE (2018)	n.a.
2019	SO-DE (2022), SO-TH (2022)	SO-DE (2022), SO-TH (2022)	SO-DE (2022)	n.a.

Notes: The variables are defined in Table S8.

SO-DE: Statistical office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-TH: statistical office of Thuringia (https://www.statistischebibliothek.de/mir/receive/THSerie_mods_00000176).

n.a.: not available.

[a] population connected to industrial WWTPs only.

S6.1.2 Availability and sources of the data used to estimate the connection to wastewater treatment for the different treatment types at NUTS-1 level

Table S27. Description and sources of the data used to estimate the connection of the **total resident population of Schleswig-Holstein (SH)** to wastewater treatment for the different treatment types

t	$T1_{all}(t,SH)$	$T2_{all}(t,SH); T3_{all}(t,SH)$	$T23^{noN}_{all}(t,SH),; T3^{N}_{all}(t,SH)$	$T23^{noP}_{all}(t,SH); T3^{P}_{all}(t,SH)$
1975	population treated in SH SO-WDE (1979)	population treated in SH SO-WDE (1979)	n.a.	n.a.
1979	population treated in SH SO-WDE (1983)	population treated in SH SO-WDE (1983)	n.a.	n.a.
1983	population treated in SH SO-WDE (1986)	population treated in SH SO-WDE (1986)	n.a.	n.a.
1987	**resident population** of SH SO-DE (1990)	population treated in SH SO-DE (1990)	population treated in SH SO-DE (1990)	population treated in SH[a] SO-DE (1990)
1991	**resident population** of SH SO-DE (1995)	population treated in SH SO-DE (1995)	population treated in SH SO-DE (1995)	population treated in SH[a] SO-DE (1995)
1995	**resident population** of SH SO-DE (1998)	population treated in SH SO-DE (1998)	% population treated in SH SO-DE (1998)	population treated in SH[a] SO-DE (1998)
1998	population treated in SH SO-DE (2001), SO-SH (2001)	population treated in SH SO-DE (2001), SO-SH (2001)	population treated in SH SO-DE (2001), SO-SH (2001)	population treated in SH SO-DE (2001), SO-SH (2001)
2001	population treated in SH SO-DE (2003), SO-SH (2003)	population treated in SH SO-DE (2003), SO-SH (2003)	population treated in SH SO-DE (2003), SO-SH (2003)	population treated in SH SO-DE (2003), SO-SH (2003)
2004	population treated in SH SO-DE (2006), SO-HH&SH (2006b)	population treated in SH SO-DE (2006), SO-HH&SH (2006b)	population treated in SH SO-DE (2006), SO-HH&SH (2006b)	population treated in SH SO-DE (2006), SO-HH&SH (2006b)
2007	population treated in SH SO-DE (2009), SO-HH&SH (2009b)	population treated in SH SO-DE (2009), SO-HH&SH (2009b)	population treated in SH SO-DE (2009), SO-HH&SH (2009b)	population treated in SH SO-DE (2009), SO-HH&SH (2009b)
2010	population treated in SH SO-HH&SH (2012b)	population treated in SH SO-HH&SH (2012b)	population treated in SH SO-HH&SH (2012b)	population treated in SH SO-HH&SH (2012b)
2013	n.a.[b]	n.a.[b]	n.a.[b]	n.a.[b]
2016	population treated in SH SO-HH&SH (2018b)	population treated in SH SO-HH&SH (2018b)	population treated in SH SO-HH&SH (2018b)	population treated in SH SO-HH&SH (2018b)
2019	population treated in SH SO-HH&SH (2021b)	population treated in SH SO-HH&SH (2021b)	population treated in SH SO-HH&SH (2021b)	population treated in SH SO-HH&SH (2021b)

Notes: The variables are defined in Table S8.

SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany (data link: https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-SH: Statistical Office of Schleswig-Holstein; SO-HH&SH: Statistical Office of Hamburg and Schleswig-Holstein (data link: https://www.statistischebibliothek.de/mir/receive/SHSerie_mods_00000083).

n.a.: not available

[a] In 1987, 1991, and 1995, $T3^{P}_{all}$ is provided for different technologies of P removal (several of them could be used in combination) but the total P removal is not specified. We use the largest value across all technologies, which is therefore probably not the actual value but a lower bound estimate.

[b] data are provided in SO-HH&SH (2015), but we do not use them, as they seem to have uncertainties.

Table S28. Description and sources of the data used to estimate the connection of the **total resident population of Hamburg (HH)** to wastewater treatment for the different treatment types

t	$T1_{all}(t, HH)$	$T2_{all}(t, HH); T3_{all}(t, HH)$	$T23_{all}^{noN}(t, HH); T3_{all}^{N}(t, HH)$	$T23_{all}^{noP}(t, HH); T3_{all}^{P}(t, HH)$
1975	population treated in HH SO-WDE (1979)	population treated in HH SO-WDE (1979)	population treated in HH[a] SO-WDE (1979)	population treated in HH[a] SO-WDE (1979)
1979	population treated in HH SO-WDE (1983)	population treated in HH SO-WDE (1983)	population treated in HH[a] SO-WDE (1983)	population treated in HH[a] SO-WDE (1983)
1983	population treated in HH SO-WDE (1986)	% population treated in HH SO-WDE (1986)	population treated in HH[a] SO-WDE (1986)	population treated in HH[a] SO-WDE (1986)
1987	**resident population** of HH SO-DE (1990)	population treated in HH SO-DE (1990)	population treated in HH SO-DE (1990)	population treated in HH SO-DE (1990)
1991	**resident population** of HH SO-DE (1995)	population treated in HH SO-DE (1995)	population treated in HH SO-DE (1995)	population treated in HH[b] SO-DE (1995)
1995	**resident population** of HH SO-DE (1998)	population treated in HH SO-DE (1998)	population treated in HH SO-DE (1998)	population treated in HH[b] SO-DE (1998)
1998	population treated in HH SO-DE (2001)	population treated in HH SO-DE (2001)	population treated in HH SO-DE (2001)	population treated in HH SO-DE (2001)
2001	population treated in HH SO-DE (2003)	population treated in HH SO-DE (2003)	population treated in HH SO-DE (2003)	population treated in HH SO-DE (2003)
2004	population treated in HH SO-DE (2006), SO-HH&SH (2006a)	population treated in HH SO-DE (2006), SO-HH&SH (2006a)	population treated in HH SO-DE (2006)	population treated in HH SO-DE (2006)
2007	population treated in HH SO-DE (2009), SO-HH&SH (2009a)	population treated in HH SO-DE (2009), SO-HH&SH (2009a)	population treated in HH SO-DE (2009)	population treated in HH SO-DE (2009)
2010	population treated in HH SO-HH&SH (2012a)	population treated in HH SO-HH&SH (2012a)	n.a.[c]	n.a.[c]
2013	population treated in HH SO-HH&SH (2015a)	population treated in HH SO-HH&SH (2015a)	population treated in HH SO-HH&SH (2015)a	population treated in HH SO-HH&SH (2015a)
2016	population treated in HH SO-HH&SH (2018a)	population treated in HH SO-HH&SH (2018a)	population treated in HH SO-HH&SH (2018a)	population treated in HH SO-HH&SH (2018a)
2019	population treated in SH SO-HH&SH (2021a)	population treated in HH SO-HH&SH (2021a)	population treated in HH SO-HH&SH (2021a)	population treated in HH SO-HH&SH (2021a)

Notes: The variables are defined in Table S8.
SO-WDE: Statistical office of West Germany; SO-DE: Statistical office of Germany
(https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694;
https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);
SO-HH&SH: statistical office of Hamburg-Schleswig-Holstein (https://www.statistischebibliothek.de/mir/receive/HHSerie_mods_00000126).
n.a.: not available.
[a] $T3_{all}$ is equal to zero in 1975, 1979, and 1983, and therefore $T3_{all}^{N}$ and $T3_{all}^{P}$ as well.
[b] In 1991, and 1995, $T3_{all}^{P}$ is provided for different technologies of P removal (several of them could be used in combination) but the total P removal is not specified. We use the largest value across all technologies, which is therefore probably not the actual value but a lower bound estimate.
[c] $T3_{all}^{N}$ and $T3_{all}^{P}$ are not provided in 2010. We assume that they are equal to total connection to WWTP as in 2004, 2007 and after 2010.

Table S29. Description and sources of the data used to estimate the connection of the **total resident population of Lower Saxony (LS)** to wastewater treatment for the different treatment types

t	$T1_{all}(t, LS)$	$T2_{all}(t, LS); T3_{all}(t, LS)$	$T23_{all}^{noN}(t, LS); T3_{all}^{N}(t, LS)$	$T23_{all}^{noP}(t, LS); T3_{all}^{P}(t, LS)$
1975	population treated in LS SO-WDE (1979)	population treated in LS SO-WDE (1979)	n.a.	n.a.
1979	population treated in LS SO-WDE (1983)	population treated in LS SO-WDE (1983)	n.a.	n.a.
1983	population treated in LS SO-WDE (1986)	population treated in LS SO-WDE (1986)	n.a.	n.a.
1987	**resident population** of LS SO-DE (1990)	population treated in LS SO-DE (1990)	population treated in LS SO-DE (1990)	population treated in LS[a] SO-DE (1990)
1991	**resident population** of LS SO-DE (1995)	population treated in LS SO-DE (1995)	population treated in LS SO-DE (1995)	population treated in LS[a] SO-DE (1995)
1995	**resident population** of LS SO-DE (1998)	population treated in LS SO-DE (1998)	population treated in LS SO-DE (1998)	population treated in LS[a] SO-DE (1998)
1998	population treated in LS SO-DE (2001)	SO-DE (2001)	population treated in LS SO-DE (2001)	population treated in LS SO-DE (2001)
2001	population treated in LS SO-DE (2003)	population treated in LS SO-DE (2003)	population treated in LS SO-DE (2003)	population treated in LS SO-DE (2003)
2004	population treated in LS SO-DE (2006)	population treated in LS SO-DE (2006)	population treated in LS SO-DE (2006)	population treated in LS SO-DE (2006)
2007	population treated in LS SO-DE (2009)	population treated in LS SO-DE (2009)	population treated in LS SO-DE (2009)	population treated in LS SO-DE (2009)
2010	**water treated** in LS SO-DE (2013)	**water treated** in LS SO-DE (2013)	**water treated** in LS SO-DE (2013)	**water treated** in LS SO-DE (2013)
2013	**water treated** in LS SO-DE (2015)	**water treated** in LS SO-DE (2015)	**water treated** in LS SO-DE (2015)	**water treated** in LS SO-DE (2015)
2016	**water treated** in LS SO-DE (2018)	**water treated** in LS SO-DE (2018)	**water treated** in LS SO-DE (2018)	**water treated** in LS SO-DE (2018)
2019	**water treated** in LS SO-DE (2022)	**water treated** in LS SO-DE (2022)	**water treated** in LS SO-DE (2022)	**water treated** in LS SO-DE (2022)

Notes: n.a.: not available

The variables are defined in Table S8.

SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany

(https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694;

https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B).

[a] In 1987, 1991, and 1995, $T3_{all}^{P}$ is provided for different technologies of P removal (several of them could be used in combination) but the total P removal is not specified. We use the largest value across all technologies, which is therefore probably not the actual value but a lower bound estimate.

Table S30. Description and sources of the data used to estimate the connection of the **total resident population of Bremen (HB)** to wastewater treatment for the different treatment types

t	$T1_{all}(t, HB)$	$T2_{all}(t, HB); T3_{all}(t, HB)$	$T23^{noN}_{all}(t, HB); T3^{N}_{all}(t, HB)$	$T23^{noP}_{all}(t, HB); T3^{P}_{all}(t, HB)$
1975	population treated in HB SO-WDE (1979)	population treated in HB SO-WDE (1979)	population treated in HB[a] SO-WDE (1979)	population treated in HB[a] SO-WDE (1979)
1979	population treated in HB SO-WDE (1983)	population treated in HB SO-WDE (1983)	population treated in HB[a] SO-WDE (1983)	population treated in HB[a] SO-WDE (1983)
1983	population treated in HB SO-WDE (1986)	n.a.[b]	n.a.[b]	n.a.[b]
1987	**resident population** of HB SO-DE (1990)	population treated in HB SO-DE (1990)	population treated in HB SO-DE (1990)	population treated in HB SO-DE (1990)
1991	**resident population** of HB SO-DE (1995)	population treated in HB SO-DE (1995)	population treated in HB SO-DE (1995)	population treated in HB[c] SO-DE (1995)
1995	**resident population** of HB SO-DE (1998)	population treated in HB SO-DE (1998)	population treated in HB SO-DE (1998)	population treated in HB[c] SO-DE (1998)
1998	population treated in HB SO-DE (2001)	population treated in HB SO-DE (2001)	population treated in HB SO-DE (2001)	population treated in HB SO-DE (2001)
2001	population treated in HB SO-DE (2003)	population treated in HB SO-DE (2003)	population treated in HB SO-DE (2003)	population treated in HB SO-DE (2003)
2004	population treated in HB SO-DE (2006)	population treated in HB SO-DE (2006)	population treated in HB SO-DE (2006)	population treated in HB SO-DE (2006)
2007	population treated in HB SO-DE (2009)	population treated in HB SO-DE (2009)	population treated in HB SO-DE (2009)	population treated in HB SO-DE (2009)
2010	**water treated** in HB SO-DE (2013)	**water treated** in HB SO-DE (2013)	**water treated** in HB SO-DE (2013)	**water treated** in HB SO-DE (2013)
2013	**water treated** in HB SO-DE (2015)	**water treated** in HB SO-DE (2015)	**water treated** in HB SO-DE (2015)	**water treated** in HB SO-DE (2015)
2016	**water treated** in HB SO-DE (2018)	**water treated** in HB SO-DE (2018)	**water treated** in LHB SO-DE (2018)	**water treated** in HB SO-DE (2018)
2019	**water treated** in HB SO-DE (2022)	**water treated** in HB SO-DE (2022)	**water treated** in HB SO-DE (2022)	**water treated** in HB SO-DE (2022)

Notes: The variables are defined in Table S8.
SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany
(https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694;
https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B).
n.a.: not available.
[a] $T3_{all}$ is equal to zero in 1975 and 1979, and therefore $T3^{N}_{all}$ and $T3^{P}_{all}$ as well.
[b] $T3_{all}$ is provided in 1983 (SO-WDE, 1986), but we discard this value which seems implausible. In fact, while $T3$ is equal to 0 % in 1975, 1979, and 1987, it is equal to 18.10 % in 1983. We instead assume that $T3_{all}$, $T3^{N}_{all}$ and $T3^{P}_{all}$ are equal to 0 % in 1983.
[c] In 1991, and 1995, $T3^{P}_{all}$ is provided for different technologies of P removal (several of them could be used in combination) but the total P removal is not specified. We use the largest value across all technologies, which is therefore probably not the actual value but a lower bound estimate.

Table S31. Description and sources of the data used to estimate the connection of the **total resident population of North Rhine-Westphalia (NW)** to wastewater treatment for the different treatment types

t	$T1_{all}(t, NW)$	$T2_{all}(t, NW); T3_{all}(t, NW)$	$T23^{noN}_{all}(t, NW)$; $T3^{N}_{all}(t, NW)$	$T23^{noP}_{all}(t, NW)$; $T3^{P}_{all}(t, NW)$
1969	**resident population** of NW SO-NW (2001)	n.a.	n.a.	n.a.
1975	**resident population** of NW SO-NW (2001)	population treated in NW SO-WDE (1979)	n.a.	n.a.
1979	n.a.[a]	population treated in NW SO-WDE (1983)	n.a.	n.a.
1983	**resident population** of NW SO-NW (2001)	population treated in NW SO-WDE (1986)	n.a.	n.a.
1987	**resident population** of NW SO-DE (1990), SO-NW (2001)	population treated in NW SO-WDE (1990)	population treated in NW SO-DE (1990)	population treated in NW[b] SO-DE (1990)
1991	**resident population** of NW SO-DE (1995), SO-NW (2001)	population treated in NW SO-DE (1995)	population treated in NW SO-DE (1995)	population treated in NW[b] SO-DE (1995)
1995	**resident population** of NW SO-DE (1998), SO-NW (2001)	population treated in NW SO-DE (1998)	population treated in NW SO-DE (1998)	population treated in NW[b] SO-DE (1998)
1998	**resident population** of NW SO-NW (2001)	population treated in NW SO-DE (2001), SO-NW (2001)	population treated in NW SO-DE (2001)	population treated in NW SO-DE (2001)
2001	**resident population** of NW SO-NW (2003)	population treated in NW SO-DE (2003), SO-NW (2003)	population treated in NW SO-DE (2003)	population treated in NW SO-DE (2003)
2004	**resident population** of NW SO-NW (2006)	population treated in NW SO-DE (2006), SO-NW (2006)	population treated in NW SO-DE (2006)	population treated in NW SO-DE (2006)
2007	**resident population** of NW SO-NW (2009)	population treated in NW SO-DE (2009), SO-NW (2009)	population treated in NW SO-DE (2009)	population treated in NW SO-DE (2009)
2010	**resident population** of NW SO-NW (2013)	population treated in NW SO-NW (2013)	**water treated** in NW SO-DE (2013)	**water treated** in NW SO-DE (2013)
2013	**resident population** of NW SO-NW (2016)	population treated in NW SO-NW (2016)	**water treated** in NW SO-DE (2015)	**water treated** in NW SO-DE (2015)
2016	**resident population** of NW SO-NW (2018)	population treated in NW SO-NW (2018)	**water treated** in NW SO-DE (2018)	**water treated** in NW SO-DE (2018)
2019	**water treated**[c] in NW SO-DE (2022)	n.a.[d]	**water treated** in NW SO-DE (2022)	**water treated** in NW SO-DE (2022)

Notes: The variables are defined in Table S8.
SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany
(https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694;
https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);
SO-NW: Statistical Office of North Rhine-Westphalia (https://www.statistischebibliothek.de/mir/receive/NWSerie_mods_00000171)
n.a.: not available.
[a] $T1_{all}$ data with respect to the percentage of population treated in NW is provided in 1979 (WSO-DE, 1983). We do not use this data for consistency with the data type for the other years which refers to the percentage of resident population of NW. [b] In 1987, 1991, and 1995, $T3^{P}_{all}$ is provided for different technologies of P removal (several of them could be used in combination) but the total P removal is not specified. We use the largest value across all technologies, which is therefore probably not the actual value but a lower bound estimate. [c] In 2016, the volume of wastewater treated with primary treatment is close to 0, similar to previous years. Therefore, we assume that $T1_{all}$ is 0 in 2019. [d] In 2019, $T3_{all}$ is provided in SO-DE (2022) in terms of percentage of treated water. For consistency with the data type used in the previous year (percentage of population treated in NW) we do not use the 2019 data but assume that $T3_{all}$ in 2019 is equal to the 2016 value.

Table S32. Description and sources of the data used to estimate the connection of the **total resident population of Hesse (HE)** to wastewater treatment for the different treatment types

t	$T1_{all}(t, HE)$	$T2_{all}(t, HE); T3_{all}(t, HE)$	$T23^{noN}_{all}(t, HE); T3^{N}_{all}(t, HE)$	$T23^{noP}_{all}(t, HE); T3^{P}_{all}(t, HE)$
1975	population treated in HE SO-WDE (1979)	population treated in HE SO-WDE (1979)	n.a.	n.a.
1979	population treated in HE SO-WDE (1983)	population treated in HE SO-WDE (1983)	n.a.	n.a.
1983	population treated in HE SO-WDE (1986)	population treated in HE SO-WDE (1986)	n.a.	n.a.
1987	**resident population** of HE SO-DE (1990)	population treated in HE SO-DE (1990)	population treated in HE SO-DE (1990)	population treated in HE[a] SO-DE (1990)
1991	**resident population** of HE SO-DE (1995)	population treated in HE SO-DE (1995)	population treated in HE SO-DE (1995)	population treated in HE[a] SO-DE (1995)
1995	**resident population** of HE SO-DE (1998)	population treated in HE SO-DE (1998)	population treated in HE SO-DE (1998)	population treated in HE[a] SO-DE (1998)
1998	population treated in HE SO-DE (2001)	population treated in HE SO-DE (2001)	population treated in HE SO-DE (2001)	population treated in HE SO-DE (2001)
2001	population treated in HE SO-DE (2003)	population treated in HE SO-DE (2003)	population treated in HE SO-DE (2003)	population treated in HE SO-DE (2003)
2004	population treated in HE SO-DE (2006)	population treated in HE SO-DE (2006)	population treated in HE SO-DE (2006)	population treated in HE SO-DE (2006)
2007	population treated in HE SO-DE (2009)	population treated in HE SO-DE (2009)	population treated in HE SO-DE (2009)	population treated in HE SO-DE (2009)
2010	**water treated** in HE SO-DE (2013)	**water treated** in HE SO-DE (2013)	**water treated** in HE SO-DE (2013)	**water treated** in HE SO-DE (2013)
2013	**water treated** in HE SO-DE (2015)	**water treated** in HE SO-DE (2015)	**water treated** in HE SO-DE (2015)	**water treated** in HE SO-DE (2015)
2016	**water treated** in HE SO-DE (2018)	**water treated** in HE SO-DE (2018)	n.a.[b]	**water treated** in HE SO-DE (2018)
2019	**water treated** in HE SO-DE (2022)	**water treated** in HE SO-DE (2022)	**water treated** in HE SO-DE (2022)	**water treated** in HE SO-DE (2022)

Notes: The variables are defined in Table S8.

SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

n.a.: not available

[a] In 1987, 1991, and 1995, $T3^{P}_{all}$ is provided for different technologies of P removal (several of them could be used in combination) but the total P removal is not specified. We use the largest value across all technologies, which is therefore probably not the actual value but a lower bound estimate.

[b] Data of the volume of water treated with $T3^{N}_{all}$ is provided in SO-DE (2018). However we did not use this data, since we consider that it leads to an unrealistic drop in $T3^{N}_{all}$ (93.73 %), while $T3^{N}_{all}$ is estimated to be equal to 96.70 % and 95.49 % in 2013 and 2019, respectively.

Table S33. Description and sources of the data used to estimate the connection of the **total resident population of Rhineland-Palatinate (RP)** to wastewater treatment for the different treatment types

Year	$T1_{all}(t,RP)$	$T2_{all}(t,RP); T3_{all}(t,RP)$	$T23^{noN}_{all}(t,RP); T3^{N}_{all}(t,RP)$	$T23^{noP}_{all}(t,RP); T3^{P}_{all}(t,RP)$
1963	**resident population** of RP SO-RP (2006)	n.a.	n.a.	n.a.
1969	**resident population** of RP SO-RP (2006)	n.a.	n.a.	n.a.
1975	**resident population** of RP SO-RP (2006)	population treated in RP SO-WDE (1979)	n.a.	n.a.
1979	**resident population** of RP SO-RP (2006)	population treated in RP SO-WDE (1983)	n.a.	n.a.
1983	**resident population** of RP SO-RP (2006)	population treated in RP SO-WDE (1986)	n.a.	n.a.
1987	**resident population** of RP SO-DE (1990), SO-RP (2006)	n.a.	n.a.	population treated in RP[a] SO-DE (1990)
1991	**resident population** of RP SO-DE (1995), SO-RP (2006)	population treated in RP SO-DE (1995)	population treated of RP SO-DE (1995)	population treated in RP[a] SO-DE (1995)
1995	**resident population** of RP SO-DE (1998), SO-RP (2006)	population treated in RP SO-DE (1998)	population treated in RP SO-DE (1998)	population treated in RP[a] SO-DE (1998)
1998	**resident population** of RP SO-RP (2006)	**resident population** of RP SO-RP (2006)	**resident population** of RP SO-RP (2006)	**resident population** of RP SO-RP (2006)
2001	**resident population** of RP SO-RP (2003)	**resident population** of RP[b] SO-RP (2003)	**resident population** of RP[b] SO-RP (2003)	**resident population** of RP[b] SO-RP (2003)
2004	**resident population** of RP SO-RP (2006)	**resident population** of RP[b] SO-RP (2006)	**resident population** of RP[b] SO-RP (2006)	**resident population** of RP[b] SO–RP (2006)
2007	**resident population** of RP SO-RP (2009)	**resident population** of RP[b] SO-RP (2009)	**resident population** of RP[b] SO-RP (2009)	**resident population** of RP[b] SO-RP (2009)
2010	**resident population** of RP SO-RP (2012)	**resident population** of RP[b] SO-RP (2013)	**resident population** of RP[b] SO-RP (2013)	**resident population** of RP[b] SO-RP (2013)
2013	**resident population** of RP SO-RP (2015)	**resident population** of RP[b] SO-RP (2015)	**resident population** of RP[b] SO-RP (2015)	**resident population** of RP[b] SO-RP (2015)
2016	**resident population** of RP SO-RP (2018)	**resident population** of RP[b] SO-RP (2018)	**resident population** of RP[b] SO-RP (2018)	**resident population** of RP[b] SO-RP (2018)
2019	**resident population** of RP[b] SO-RP (2022)	**resident population** of RP SO-RP (2022)	**resident population** of RP[b] SO-RP (2022)	**resident population** of RP[b] SO-RP (2022)

Notes: The variables are defined in Table S8.
SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany
(https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694;
https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);
SO-RP: Statistical Office of Rhineland-Palatinate https://www.statistischebibliothek.de/mir/receive/RPSerie_mods_00001054.
n.a.: not available.
[a] In 1987, 1991, and 1995, $T3^{P}_{all}$ is provided for different technologies of P removal (several of them could be used in combination) but the total P removal is not specified. We use the largest value across all technologies, which is therefore probably not the actual value but a lower bound estimate.
[b] The data provided in the statistical reports refers to the population connected to wastewater treatment through the sewer system. To estimate population connected to each treatment type through cesspit as well, we assumed the same proportion of N and P removal with respect to total secondary treatment as for the population connected to sewer, since cesspits receive at least secondary treatment.

Table S34. Description and sources of the data used to estimate the connection of the **total resident population of Baden-Württemberg (BW)** to wastewater treatment for the different treatment types

t	$T1_{all}(t, BW)$	$T2_{all}(t, BW); T3_{all}(t, BW)$	$T23^{noN}_{all}(t, BW)$; $T3^{N}_{all}(t, BW)$	$T23^{noP}_{all}(t, BW); T3^{P}_{all}(t, BW)$
1955	n.a.	no WWTPs with $T3^a$ LUBW and MUKEBW (2021, Fig. 8)	no WWTPs with $T3^a$ LUBW and MUKEBW (2021, Fig. 8)	no WWTPs with $T3^a$ LUBW and MUKEBW (2021, Fig. 8)
1975	population treated in BW SO-WDE (1979)	population treated in BW SO-WDE (1979)	almost np WWTPs with ${T3_N}^a$ LUBW and MUKEBW (2021, Fig. 8)	almost no WWTPs with ${T3_P}^a$ LUBW and MUKEBW (2021, Fig. 8)
1979	population treated in BW SO-WDE (1983)	population treated in BW SO-WDE (1983)	almost no WWTPs with ${T3_N}^a$ LUBW and MUKEBW (2021, Fig. 8)	almost no WWTPs with ${T3_P}^a$ LUBW and MUKEBW (2021, Fig. 8)
1983	population treated in BW SO-WDE (1986)	population treated in BW SO-WDE (1986)	n.a.	n.a.
1987	**resident population** of BW SO-DE (1990)	population treated in BW SO-DE (1990)	population treated in BW SO-DE (1990)	population treated in BW[b] SO-DE (1990)
1991	**resident population** of BW SO-DE (1995)	population treated in BW SO-DE (1995)	population treated in BW SO-DE (1995)	population treated in BW[b] SO-DE (1995)
1995	**resident population** of BW SO-DE (1998)	population treated in BW SO-DE (1998)	population treated in BW SO-DE (1998)	population treated in BW[b] SO-DE (1998)
1998	population treated in BW SO-DE (2001)	population treated in BW SO-DE (2001)	population treated in BW SO-DE (2001)	population treated in BW SO-DE (2001)
2001	population treated in BW SO-DE (2003)	population treated in BW SO-DE (2003)	population treated in BW SO-DE (2003)	population treated in BW SO-DE (2003)
2004	population treated in BW SO-DE (2006)	population treated in BW SO-DE (2006)	population treated in BW SO-DE (2006)	population treated in BW SO-DE (2006)
2007	population treated in BW SO-DE (2009)	population treated in BW SO-DE (2009)	population treated in BW SO-DE (2009)	population treated in BW SO-DE (2009)
2010	**water treated** in BW SO-DE (2013)	**water treated** in BW SO-DE (2013)	**water treated** in BW SO-DE (2013)	**water treated** in BW SO-DE (2013)
2013	**water treated** in BW SO-DE (2015)	**water treated** in BW SO-DE (2015)	**water treated** in BW SO-DE (2015)	**water treated** in BW SO-DE (2015)
2016	**water treated** in BW SO-DE (2018)	**water treated** in BW SO-DE (2018)	**water treated** in BW SO-DE (2018)	**water treated** in BW SO-DE (2018)
2019	**water treated** in BW SO-DE (2022)	**water treated** in BW SO-DE (2022)	**water treated** in BW SO-DE (2022)	**water treated** in BW SO-DE (2022)

Notes: The variables are defined in Table S8.
SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany
(https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694;
https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B).
n.a.: not available.

[a] We consider that $T3^N_{all}$ and $T3^P_{all}$ started in 1980 (it was negligible before 1980), and that $T3_{all}$ started in 1956 (LUBW and MUKEBW, 2021, Fig. 8).

[b] In 1987, 1991, and 1995, $T3^P_{all}$ is provided for different technologies of P removal (several of them could be used in combination) but the total P removal is not specified. We use the largest value across all technologies, which is therefore probably not the actual value but a lower bound estimate.

Table S35. Description and sources of the data used to estimate the connection of the **total resident population of Bavaria (BV)** to wastewater treatment for the different treatment types

t	$T1_{all}(t, BV)$	$T2_{all}(t, BV); T3_{all}(t, BV)$	$T23^{noN}_{all}(t, BV); T3^{N}_{all}(t, BV)$	$T23^{noP}_{all}(t, BV); T3^{P}_{all}(t, BV)$
1975	population treated in BV SO-WDE (1979)	population treated in BV SO-WDE (1979)	n.a.	n.a.
1979	population treated in BV SO-WDE (1983)	population treated in BV SO-WDE (1983)	n.a.	n.a.
1983	population treated in BV SO-WDE (1986)	population treated in BV SO-WDE (1986)	n.a.	n.a.
1987	**resident population** of BV SO-DE (1990)	population treated in BV SO-DE (1990)	population treated in BV SO-DE (1990)	population treated in BV[a] SO-DE (1990)
1991	**resident population** of BV SO-DE (1995)	population treated in BV SO-DE (1995)	population treated in BV SO-DE (1995)	population treated in BV[a] SO-DE (1995)
1995	**resident population** of BV SO-DE (1998)	population treated in BV SO-DE (1998)	population treated in BV SO-DE (1998)	population treated in BV[a] SO-DE (1998)
1998	population treated in BV SO-DE (2001), SO-BV (2001)	population treated in BV SO-DE (2001), SO-BV (2001)	population treated in BV SO-DE (2001), SO-BV (2001)	population treated in BV SO-DE (2001), SO-BV (2001)
2001	population treated in BV SO-DE (2003), SO-BV (2003)	population treated in BV SO-DE (2003), SO-BV (2003)	population treated in BV SO-DE (2003), SO-BV (2003)	population treated in BV SO-DE (2003), SO-BV (2003)
2004	population treated in BV SO-DE (2006), SO-BV (2006)	population treated in BV SO-DE (2006), SO-BV (2006)	population treated in BV SO-DE (2006), SO-BV (2006)	population treated in BV SO-DE (2006), SO-BV (2006)
2007	population treated in BV SO-DE (2009), SO-BV (2009)	population treated in BV SO-DE (2009), SO-BV (2009)	population treated in BV SO-DE (2009), SO-BV (2009)	population treated in BV SO-DE (2009), SO-BV (2009)
2010	population treated in BV SO-BV (2012)	population treated in BV SO-BV (2012)	population treated in BV SO-BV (2012)	population treated in BV SO-BV (2012)
2013	population treated in BV SO-BV (2015)	population treated in BV SO-BV (2015)	population treated in BV SO-BV (2015)	population treated in BV SO-BV (2015)
2016	population treated in BV SO-BV (2020)	population treated in BV SO-BV (2020)	population treated in BV SO-BV (2020)	population treated in BV SO-BV (2020)
2019	n.a.[b]	n.a.[b]	n.a.[b]	n.a.[b]

Notes: The variables are defined in Table S8.

SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-BV: Statistical Office of Bavaria (https://www.statistischebibliothek.de/mir/receive/BYSerie_mods_00000808).

n.a.: not available.

[a] In 1987, 1991, and 1995, $T3^{P}_{all}$ is provided for different technologies of P removal (several of them could be used in combination) but the total P removal is not specified. We use the largest value across all technologies, which is therefore probably not the actual value but a lower bound estimate.

[b] The percentage of water treated in BV is available for all treatment type in SO-DE (2022). However we do not use these data because they are not consistent with the type of data used in the other years (percentage of population treated in BV). Instead, we use in 2019 the same value as in 2016.

Table S36. Description and sources of the data used to estimate the connection of the **total resident population of Saarland (SL)** to wastewater treatment for the different treatment types

t	$T1_{all}(t, SL)$	$T2_{all}(t, SL); T3_{all}(t, SL)$	$T23^{noN}_{all}(t, SL); T3^{N}_{all}(t, SL)$	$T23^{noP}_{all}(t, SL); T3^{P}_{all}(t, SL)$
1975	population treated in SL SO-WDE (1979)	population treated in SL SO-WDE (1979)	population treated in SL[a] SO-WDE (1979)	population treated in SL[a] SO-WDE (1979)
1979	population treated in SL SO-WDE (1983)	population treated in SL SO-WDE (1983)	population treated in SL[a] SO-WDE (1983)	population treated in SL[a] SO-WDE (1983)
1983	population treated in SL SO-WDE (1986)	population treated in SL SO-WDE (1986)	population treated in SL[a] SO-WDE (1986)	population treated in SL[a] SO-WDE (1986)
1987	**resident population** of SL SO-DE (1990)	n.a.	n.a.	n.a.
1991	**resident population** of SL SO-DE (1995)	population treated in SL SO-DE (1995)	population treated in SL SO-DE (1995)	n.a.
1995	**resident population** of SL SO-DE (1998)	population treated in SL SO-DE (1998)	population treated in SL SO-DE (1998)	population treated in SL[b] SO-DE (1998)
1998	population treated in SL SO-DE (2001)	population treated in SL SO-DE (2001)	population treated in SL SO-DE (2001)	population treated in SL SO-DE (2001)
2001	population treated in SL SO-DE (2003)	population treated in SL SO-DE (2003)	population treated in SL SO-DE (2003)	n.a.[c]
2004	population treated in SL SO-DE (2006)	population treated in SL SO-DE (2006)	population treated in SL SO-DE (2006)	population treated in SL SO-DE (2006)
2007	population treated in SL SO-DE (2009)	population treated in SL SO-DE (2009)	population treated in SL SO-DE (2009)	population treated in SL SO-DE (2009)
2010	**water treated** in SL SO-DE (2013)	**water treated** in SL SO-DE (2013)	**water treated** in SL SO-DE (2013)	**water treated** in SL SO-DE (2013)
2013	**water treated** in SL SO-DE (2015)	**water treated** in SL SO-DE (2015)	**water treated** in SL SO-DE (2015)	**water treated** in SL SO-DE (2015)
2016	**water treated** in SL SO-DE (2018)	**water treated** in SL SO-DE (2018)	n.a.[d] SO-DE (2018)	**water treated** in SL SO-DE (2018)
2019	**water treated** in SL SO-DE (2022)	**water treated** in SL SO-DE (2022)	**water treated** in SL SO-DE (2022)	**water treated** in SL SO-DE (2022)

Notes: The variables are defined in Table S8.
SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany
(https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694;
https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B).
n.a.: not available.
[a] $T3_{all}$ is equal to zero in 1979 and 1983 and therefore $T3^{N}_{all}$ and $T3^{P}_{all}$ as well. We also assume that $T3^{N}_{all}$ and $T3^{P}_{all}$ are equal to 0 before 1979 (although $T3_{all}$ is not equal to 0 % but to 8.08 % in 1975 from the data).
[b] In 1995, $T3^{P}_{all}$ is provided for different technologies of P removal (several of them could be used in combination) but the total P removal is not specified. We use the largest value across all technologies, which is therefore probably not the actual value but a lower bound estimate.
[c] We do not use the data provided in SO-DE (2003) because we consider it as being erroneous: $T3^{P}_{all}$ is equal to 51.45 % in 1998 and 74.42 % in 2004, while it is 36.81 % in 2001 from SO-DE (2003). [d] $T3^{N}_{all}$ is provided in SO-DE (2018). However, we discard this data because we consider that it is not plausible. While we estimate that $T3^{N}_{all}$ is equal to 87.11 % and 89.73 % in 2013 and 2019, respectively, it drops to 76.04 % in 2016, when using the data on the volume of wastewater treated to derive temporal changes on the fraction of population connected.

Table S37. Description and sources of the data used to estimate the connection of the **total resident population of Berlin (BE)** to wastewater treatment in **Berlin** for the different treatment types

t	$T1_{all}^{BE}(t, BE)$	$T2_{all}^{BE}(t, BE); T3_{all}^{BE}(t, BE)$	$T23_{all}^{noN,BE}(t, BE)$; $T3_{all}^{N,BE}(t, BE)$	$T23_{all}^{noP,BE}(t, BE)$; $T3_{all}^{P,BE}(t, BE)$
1991	**resident population** of BE SO-DE (1995)[a]	population treated in BE SO-DE (1995)	population treated in BE SO-DE (1995)	population treated in BE SO-DE (1995)[b]
1995	**resident population** of BE SO-DE (1998)[a]	population treated in BE SO-DE (1998)	n.a.[c]	population treated in BE SO-DE (1998)[b]
1998	population treated in BE SO-DE (2001)	population treated in BE SO-DE (2001)	population treated in BE SO-DE (2001)	population treated in BE SO-DE (2001)
2001	population treated in BE SO-DE (2003)	population treated in BE SO-DE (2003)	population treated in BE SO-DE (2003)	population treated in BE SO-DE (2003)
2004	population treated in BE SO-DE (2006)	population treated in BE SO-DE (2006)	population treated in BE SO-DE (2006)	population treated in BE SO-DE (2006)
2007	population treated in BE SO-DE (2009), SO-BE&BB (2010b)	population treated in BE SO-DE (2009), SO-BE&BB (2010b)	population treated in BE SO-DE (2009), SO-BE&BB (2010b)	population treated in BE SO-DE (2009), SO-BE&BB (2010b)
2010	population treated in BE SO-BE&BB (2013b)	population treated in BE SO-BE&BB (2013b)	population treated in BE SO-BE&BB (2013b)	population treated in BE SO-BE&BB (2013b)
2013	population treated in BE SO-BE&BB (2017b)	population treated in BE SO-BE&BB (2017b)	population treated in BE SO-BE&BB (2017b)	population treated in BE SO-BE&BB (2017b)
2016	population treated in BE SO-BE&BB (2019b)	population treated in BE SO-BE&BB (2019b)	population treated in BE SO-BE&BB (2019b)	population treated in BE SO-BE&BB (2019b)
2019	population treated in BE SO-BE&BB (2022b)	population treated in BE SO-BE&BB (2022b)	population treated in BE SO-BE&BB (2022b)	population treated in BE SO-BE&BB (2022b)

Notes: The variables are defined in Table S9.

SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-BE&BB: Statistical Office of Berlin and Brandenburg (https://www.statistischebibliothek.de/mir/receive/BBSerie_mods_00000564).

[a] The total population connection to primary treatment for Berlin ($T1_{all}(t, BE)$) is equal to 0 in 1991 and 1995, which applies to both the population treated in Berlin ($T1_{all}^{BE}(t, BE)$) and in Brandenburg ($T1_{all}^{BB}(t, BE)$).

[b] In 1991 and 1995, $T3_{all}^{P}$ is provided for different technologies of P removal (several of them could be used in combination) but the total P removal is not specified. We use the largest value across all technologies, which is therefore probably not the actual value but a lower bound estimate.

[c] Data of $T3_{all}^{N,BE}(t, BE)$ is provided in SO-DE (1998) but we discard it since it is implausibly high compare to the 1991 and 1998 values. While we estimate that $T3_{all}^{N,BE}(t, BE)$ is equal to 36.47 % in 1991 and 35.2 % in 1998, data suggest that it is equal to 44.72 % in 1995.

Table S38. Description and sources of the data used to estimate the connection of the **total resident population of Berlin (BE)** to wastewater treatment in **Brandenburg** for the different treatment types

t	$T1_{all}^{BB}(t, BE)$	$T2_{all}^{BB}(t, BE); T3_{all}^{BB}(t, BE)$	$T23_{all}^{noN,BB}(t, BE)$; $T3_{all}^{N,BB}(t, BE)$	$T23_{all}^{noP,BB}(t, BE)$; $T3_{all}^{P,BB}(t, BE)$
1991	**resident population** of BE SO-DE (1995)[a]	population treated in BB SO-DE (1995)[d]	population treated in BB SO-DE (1995)[d]	population treated in BB SO-DE (1995)[b,d]
1995	**resident population** of BE SO-DE (1998)[a]	population treated in BB SO-DE (1998)[d]	n.a.[f]	population treated in BB SO-DE (1998)[b,d]
1998	**resident population** of BE SO-DE (1995,1998)[c]	population treated in BB SO-DE (2001), SO-BB (2002)[d]	population treated in BB SO-DE (2001), SO-BB (2002)[d]	population treated in BB SO-DE (2001), SO-BB (2002)[d]
2001	**resident population** of BE SO-DE (1995,1998)[c]	population treated in BB SO-DE (2003), SO-BB (2004)[d]	population treated in BB SO-DE (2003), SO-BB (2004)[d]	population treated in BB SO-DE (2003), SO-BB (2004)[d]
2004	**resident population** of BE SO-DE (1995,1998)[c]	population treated in BE SO-DE (2006)[e]	population treated in BE SO-DE (2006)[e]	population treated in BE SO-DE (2006)[e]
2007	**resident population** of BE SO-DE (1995,1998)[c]	population treated in BE SO-DE (2009), SO-BE&BB (2010b)[e]	population treated in BE SO-DE (2009), SO-BE&BB (2010b)[e]	population treated in BE SO-DE (2009), SO-BE&BB (2010b)[e]
2010	**resident population** of BE SO-DE (1995,1998)[c]	population treated in BE SO-BE&BB (2013b)[e]	population treated in BE SO-BE&BB (2013b)[e]	population treated in BE SO-BE&BB (2013b)[e]
2013	**resident population** of BE SO-DE (1995,1998)[c]	population treated in BE SO-BE&BB (2017b) [e]	population treated in BE SO-BE&BB (2017b)[e]	population treated in BE SO-BE&BB (2017b)[e]
2016	**resident population** of BE SO-DE (1995,1998)[c]	population treated in BE SO-BE&BB (2019b)[e]	population treated in BE SO-BE&BB (2019b)[e]	population treated in BE SO-BE&BB (2019b)[e]
2019	**resident population** of BE SO-DE (1995,1998)[c]	population treated in BE SO-BE&BB (2022b)[e]	population treated in BE SO-BE&BB (2022b)[e]	population treated in BE SO-BE&BB (2022b)[e]

Notes: The variables are defined in Table S9.

SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-BB: Statistical Office of Brandenburg; SO-BE&BB: Statistical Office of Berlin and Brandenburg (https://www.statistischebibliothek.de/mir/receive/BBSerie_mods_00000562).

[a] The total population connection to primary treatment for Berlin ($T1_{all}(t, BE)$) is equal to 0 in 1991 and 1995, which applies to both the population treated in Berlin ($T1_{all}^{BE}(t, BE)$) and in Brandenburg ($T1_{all}^{BB}(t, BE)$).

[b] In 1991 and 1995, $T3_{all}^{P}$ is provided for different technologies of P removal (several of them could be used in combination) but the total P removal is not specified. We use the largest value across all technologies, which is therefore probably not the actual value but a lower bound estimate.

[c] We assume that $T1_{all}^{BB}(t, BE)$ is equal to 0 over the period 1998–2019, as in 1991 and 1995.

[d] For the period 1991–2001, we estimate the proportions of $T2_{all}^{BB}(t, BE)$, $T3_{all}^{BB}(t, BE)$, $T23_{all}^{noN,BB}(t, BE)$, $T3_{all}^{N,BB}(t, BE)$, $T23_{all}^{noP,BB}(t, BE)$, and $T3_{all}^{P,BB}(t, BE)$ with respect to total biological treatment (i.e. $T2_{all}^{BB}(t, BE) + T3_{all}^{BB}(t, BE)$) from the data of total population treated in Brandenburg. This is used to estimate the connection variables for the population of Berlin treated in Brandenburg.

[e] For the period 2004-2019, we assume that all the wastewater of the population resident in Berlin treated in Brandenburg undergo biological treatment with N and P removal, as it is the case for the resident population of Berlin treated in Berlin (Table S37).

[f] In 1995, $T3_{all}^{N,BB}(t, BE)$ can be estimated from the data of SO-DE (1998) as explained in note [d] above, but we discard the value since it is implausibly high compare to the 1991 and 1998 values. While we estimate that $T3_{all}^{N,BB}(t, BE)$ is equal to 3.1 % in 1991 and 27.2 % in 1998, data suggest that it is equal to 36.03 % in 1995.

Table S39. Description and sources of the data used to estimate the connection of the **total resident population of Brandenburg (BB)** to wastewater treatment for the different treatment types

t	$T1_{all}(t, BB)$	$T2_{all}(t, BB); T3_{all}(t, BB)$	$T23^{noN}_{all}(t, BB); T3^{N}_{all}(t, BB)$	$T23^{noP}_{all}(t, BB); T3^{P}_{all}(t, BB)$
1991	**resident population** of BB SO-DE (1995)	population treated in BB SO-DE (1995)[a]	population treated in BB SO-DE (1995)[a]	population treated in BB SO-DE (1995)[a,b]
1995	**resident population** of BB SO-DE (1998)	population treated in BB SO-DE (1998)[a]	n.a.[c]	population treated in BB SO-DE (1998)[a,b]
1998	population treated in BB SO-DE (2001), SO-BB (2002)[a]	population treated in BB SO-DE (2001), SO-BB (2002)[a]	population treated in BB SO-DE (2001), SO-BB (2002)[a]	population treated in BB SO-DE (2001), SO-BB (2002)[a]
2001	population treated in BB SO-DE (2003), SO-BB (2004)[a]	population treated in BB SO-DE (2003), SO-BB (2004)[a]	population treated in BB SO-DE (2003), SO-BB (2004)[a]	population treated in BB SO-DE (2003), SO-BB (2004)[a]
2004	population treated in BB SO-DE (2006), SO-BB (2007)[a]	population treated in BB SO-DE (2006), SO-BB (2007)[a]	population treated in BB SO-DE (2006), SO-BB (2007)[a]	population treated in BB SO-DE (2006), SO-BB (2007)[a]
2007	population treated in BB SO-DE (2009), SO-BE&BB (2010a)[a]	population treated in BB SO-DE (2009), SO-BE&BB (2010a)[a]	population treated in BB SO-DE (2009), SO-BE&BB (2010a)[a]	population treated in BB SO-DE (2009), SO-BE&BB (2010a)[a]
2010	population treated in BB SO-BE&BB (2013a)[a]	population treated in BB SO-BE&BB (2013a)[a]	population treated in BB SO-BE&BB (2013a)[a]	population treated in BB SO-BE&BB (2013a)[a]
2013	population treated in BB SO-BE&BB (2017a)[a]	population treated in BB SO-BE&BB (2017a)[a]	population treated in BB SO-BE&BB (2017a)[a]	population treated in BB SO-BE&BB (2017a)[a]
2016	population treated in BB SO-BE&BB (2019a)[a]	population treated in BB SO-BE&BB (2019a)[a]	population treated in BB SO-BE&BB (2019a)[a]	population treated in BB SO-BE&BB (2019a)[a]
2019	population treated in BB SO-BE&BB (2022a)[a]	population treated in BB SO-BE&BB (2022a)[a]	population treated in BB SO-BE&BB (2022a)[a]	population treated in BB SO-BE&BB (2022a)[a]

Notes: The variables are defined in Table S8.

SO-WDE: Statistical Office of West Germany; SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-BB: Statistical Office of Brandenburg; SO-BE&BB: Statistical Office of Berlin and Brandenburg (https://www.statistischebibliothek.de/mir/receive/BBSerie_mods_00000562).

[a] To estimate the connection of the resident population of Brandenburg (BB) to wastewater treatment for the different treatment types, we remove the population of Berlin treated in Brandenburg ($T3^{N,BB}_{all}(t, BE)$, estimated as explained in Table S38) from the data of population treated in WWTPs located in BB.

[b] In 1991 and 1995, $T3^{P}_{all}$ is provided for different technologies of P removal (several of them could be used in combination) but the total P removal is not specified. We use the largest value across all technologies, which is therefore probably not the actual value but a lower bound estimate.

[c] Since we discard the value of $T3^{N,BB}_{all}(t, BE)$ in 1995 (as explained in Table S38), we cannot estimate $T3^{N}_{all}(t, BB)$ in 1995.

Table S40. Description and sources of the data used to estimate the connection of the **total resident population of Mecklenburg-Vorpommern (MV)** to wastewater treatment for the different treatment types

t	$T1_{all}(t, MV)$	$T2_{all}(t, MV); T3_{all}(t, MV)$	$T23_{all}^{noN}(t, MV)$; $T3_{all}^{N}(t, MV)$	$T23_{all}^{noP}(t, MV); T3_{all}^{P}(t, MV)$
1991	**resident population** of MV SO-DE (1995)	population treated in MV SO-DE (1995)	n.a.	population treated in MV[a] SO-DE (1995)
1995	**resident population** of MV SO-DE (1998)	population treated in MV SO-DE (1998)	population treated in MV SO-DE (1998)	population treated in MV[a] SO-DE (1998)
1998	population treated in MV SO-DE (2001)	population treated in MV SO-DE (2001)	population treated in MV SO-DE (2001)	population treated in MV SO-DE (2001)
2001	population treated in MV SO-DE (2003)	population treated in MV SO-DE (2003)	population treated in MV SO-DE (2003)	population treated in MV SO-DE (2003)
2004	population treated in MV SO-DE (2006)	population treated in MV SO-DE (2006)	population treated in MV SO-DE (2006)	population treated in MV SO-DE (2006)
2007	population treated in MV SO-DE (2009)	population treated in MV SO-DE (2009)	population treated in MV SO-DE (2009)	population treated in MV SO-DE (2009)
2010	population treated in MV SO-MV (2013)	population treated in MV SO-MV (2013)	population treated in MV SO-MV (2013)	population treated in MV SO-MV (2013)
2013	population treated in MV SO-MV (2016)	population treated in MV SO-MV (2016)	population treated in MV SO-MV (2016)	population treated in MV SO-MV (2016)
2016	population treated in MV SO-MV (2019)	population treated in MV SO-MV (2019)	population treated in MV SO-MV (2019)	population treated in MV SO-MV (2019)
2019	n.a.[b]	n.a.[b]	n.a.[b]	n.a.[b]

Notes: The variables are defined in Table S8.

SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-MV: Statistical Office of Mecklenburg-Vorpommern (https://www.statistischebibliothek.de/mir/receive/MVSerie_mods_00000334).

n.a.: not available.

[a] In 1991 and 1995, $T3_{all}^{P}$ is provided for different technologies of P removal (several of them could be used in combination) but the total P removal is not specified. We use the largest value across all technologies, which is therefore probably not the actual value but a lower bound estimate.

[b] The percentage of water treated in MV is available for all treatment type in SO-DE (2022). However we do not use these data because they are not consistent with the type of data used in the other years (percentage of population treated in MV). Instead, we use in 2019 the same value as in 2016.

Table S41. Description and sources of the data used to estimate the connection of the **total resident population of Saxony (SN)** to wastewater treatment for the different treatment types

t	$T1_{all}(t, SN)$	$T2_{all}(t, SN); T3_{all}(t, SN)$	$T23^{noN}_{all}(t, SN); T3^{N}_{all}(t, SN)$	$T23^{noP}_{all}(t, SN); T3^{P}_{all}(t, SN)$
1991	**resident population** of SN SO-DE (1995)	population treated in SN SO-DE (1995)	n.a.	n.a.
1995	**resident population** of SN SO-DE (1998)	population treated in SN SO-DE (1998)	population treated in SN SO-DE (1998)	population treated in SN[a] SO-DE (1998)
1998	population treated in SN SO-DE (2001)	population treated in SN SO-DE (2001)	population treated in SN SO-DE (2001)	population treated in SN SO-DE (2001)
2001	population treated in SN SO-DE (2003), SO-SN (2003)	population treated in SN SO-DE (2003), SO-SN (2003)	population treated in SN SO-DE (2003), SO-SN (2003)	population treated in SN SO-DE (2003), SO-SN (2003)
2004	population treated in SN SO-DE (2006), SO-SN (2006)	population treated in SN SO-DE (2006), SO-SN (2006)	population treated in SN SO-DE (2006), SO-SN (2006)	population treated in SN SO-DE (2006), SO-SN (2006)
2007	population treated in SN SO-DE (2009), SO-SN (2009)	population treated in SN SO-DE (2009), SO-SN (2009)	population treated in SN SO-DE (2009), SO-SN (2009)	population treated in SN SO-DE (2009), SO-SN (2009)
2010	population treated in SN SO-SN (2011)	population treated in SN SO-SN (2011)	population treated in SN SO-SN (2011)	population treated in SN SO-SN (2011)
2013	population treated in SN SO-SN (2015)	population treated in SN SO-SN (2015)	population treated in SN SO-SN (2015)	population treated in SN SO-SN (2015)
2016	population treated in SN SO-SN (2018)	population treated in SN SO-SN (2018)	population treated in SN SO-SN (2018)	population treated in SN SO-SN (2018)
2019	population treated in SN SO-SN (2021)	population treated in SN SO-SN (2021)	population treated in SN SO-SN (2021)	population treated in SN SO-SN (2021)

Notes: The variables are defined in Table S8.
SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);
SO-SN: Statistical Office of Saxony (https://www.statistischebibliothek.de/mir/receive/SNSerie_mods_00001093).
n.a.: not available.
[a] In 1995, $T3^{P}_{all}$ is provided for different technologies of P removal (several of them could be used in combination) but the total P removal is not specified. We use the largest value across all technologies, which is therefore probably not the actual value but a lower bound estimate.

Table S42. Description and sources of the data used to estimate the connection of the **total resident population of Saxony-Anhalt (ST)** to wastewater treatment for the different treatment types

t	$T1_{all}(t,ST)$	$T2_{all}(t,ST); T3_{all}(t,ST)$	$T23^{noN}_{all}(t,ST); T3^{N}_{all}(t,ST)$	$T23^{noP}_{all}(t,ST); T3^{P}_{all}(t,ST)$
1991	**resident population** of ST	population treated in ST	population treated in ST [a]	population treated in ST[b]
	SO-DE (1995)	SO-DE (1995)	SO-DE (1995)	SO-DE (1995)
1995	**resident population** of ST	population treated in ST	population treated in ST	population treated in ST[b]
	SO-DE (1998), SO-ST (2010)	SO-DE (1998)	SO-DE (1998)	SO-DE (1998)
1998	population treated in ST	population treated in ST	population treated in ST	population treated in ST
	SO-DE (2001), SO-ST (2010)	SO-DE (2001)	SO-DE (2001)	SO-DE (2001)
2001	population treated in ST	population treated in ST	population treated in ST	population treated in ST
	SO-DE (2003)[c]	SO-DE (2003)	SO-DE (2003)	SO-DE (2003)
2004	population treated in ST	population treated in ST	population treated in ST	population treated in ST
	SO-DE (2006), SO-ST (2010)	SO-DE (2006)	SO-DE (2006)	SO-DE (2006)
2007	population treated in ST	population treated in ST	population treated in ST	population treated in ST
	SO-DE (2009), SO-ST (2010)	SO-DE (2009), SO-ST (2010)	SO-DE (2009), SO-ST (2010)	SO-DE (2009), SO-ST (2010)
2010	population treated in ST	population treated in ST	population treated in ST	population treated in ST
	SO-ST (2012)	SO-ST (2012)	SO-ST (2012)	SO-ST (2012)
2013	population treated in ST	population treated in ST	population treated in ST	population treated in ST
	SO-ST (2016)	SO-ST (2016)	SO-ST (2016)	SO-ST (2016)
2016	population treated in ST	population treated in ST	population treated in ST	population treated in ST
	SO-ST (2018)	SO-ST (2018)	SO-ST (2018)	SO-ST (2018)
2019	population treated in SN	population treated in ST	population treated in ST	population treated in ST
	SO-ST (2022)	SO-ST (2022)	SO-ST (2022)	SO-ST (2022)

Notes: The variables are defined in Table S8.

SO-DE: Statistical office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-ST: statistical office of Saxony-Anhalt (https://www.statistischebibliothek.de/mir/receive/STSerie_mods_00000324).

n.a.: not available.

[a] connection to WWTPs with nitrification is 0. We assume the same for $T3^{N}_{all}$, since nitrification is the first treatment step in WWTPs with targeted N removal.

[b] In 1991 and 1995, $T3^{P}_{all}$ is provided for different technologies of P removal (several of them could be used in combination) but the total P removal is not specified. We use the largest value across all technologies, which is therefore probably not the actual value but a lower bound estimate.

[c] In 2001, the value of $T1_{all}$ is also reported in SO-ST (2010) but we consider that it is erroneous.

Table S43. Description and sources of the data used to estimate the connection of the **total resident population of Thuringia (TH)** to wastewater treatment for the different treatment types

t	$T1_{all}(t,TH)$	$T2_{all}(t,TH); T3_{all}(t,TH)$	$T23^{noN}_{all}(t,TH); T3^{N}_{all}(t,TH)$	$T23^{noP}_{all}(t,TH); T3^{P}_{all}(t,TH)$
1991	**resident population** of TH	population treated in TH	population treated in TH[a]	population treated in TH[a]
	SO-DE (1995)	SO-DE (1995)	SO-DE (1995)	SO-DE (1995)
1995	**resident population** of TH	population treated in TH	population treated in TH	population treated in TH[b]
	SO-DE (1998)	SO-DE (1998)	SO-DE (1998)	SO-DE (1998)
1998	population treated in TH	population treated in TH	population treated in TH	population treated in TH
	SO-DE (2001), SO-TH (2006)	SO-DE (2001)	SO-DE (2001)	SO-DE (2001)
2001	population treated in TH	population treated in TH	population treated in TH	population treated in TH
	SO-DE (2003), SO-TH (2006)	SO-DE (2003)	SO-DE (2003)	SO-DE (2003)
2004	population treated in TH	population treated in TH	population treated in TH	population treated in TH
	SO-DE (2006), SO-TH (2006)	SO-DE (2006)	SO-DE (2006)	SO-DE (2006)
2007	population treated in TH[c]	population treated in TH	population treated in TH	population treated in TH
	SO-DE (2009), SO-TH (2009)	SO-DE (2009)	SO-DE (2009)	SO-DE (2009)
2010	population treated in TH	**water treated** in TH	**water treated** in TH	**water treated** in TH
	SO-TH (2012)	SO-DE (2013)	SO-DE (2013)	SO-DE (2013)
2013	population treated in TH	**water treated** in TH	**water treated** in TH	**water treated** in TH
	SO-TH (2015)	SO-DE (2015)	SO-DE (2015)	SO-DE (2015)
2016	population treated in TH	**water treated** in TH	**water treated** in TH	**water treated** in TH
	SO-TH (2019)	SO-DE (2018)	SO-DE (2018)	SO-DE (2018)
2019	population treated in TH	**water treated** in TH	**water treated** in TH	**water treated** in TH
	SO-TH (2022)	SO-DE (2022)	SO-DE (2022)	SO-DE (2022)

Notes: The variables are defined in Table S9.

SO-DE: Statistical office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-TH: statistical office of Thuringia (https://www.statistischebibliothek.de/mir/receive/THSerie_mods_00000176).

n.a.: not available.

[a] $T3$ is equal to zero in 1991 and therefore $T3^N$ and $T3^P$ as well.

[b] In 1991 and 1995, $T3^P_{all}$ is provided for different technologies of P removal (several of them could be used in combination) but the total P removal is not specified. We use the largest value across all technologies, which is therefore probably not the actual value but a lower bound estimate.

[c] In 2007, there are small differences between the data in SO-DE (2009) and SO-TH (2009) which leads to a population connection to primary treatment of 0 % and 0.42 % respectively. We adopt the data provided in SO-TH (2009) (0.42 %).

S6.2 Visualisation of the population connection data

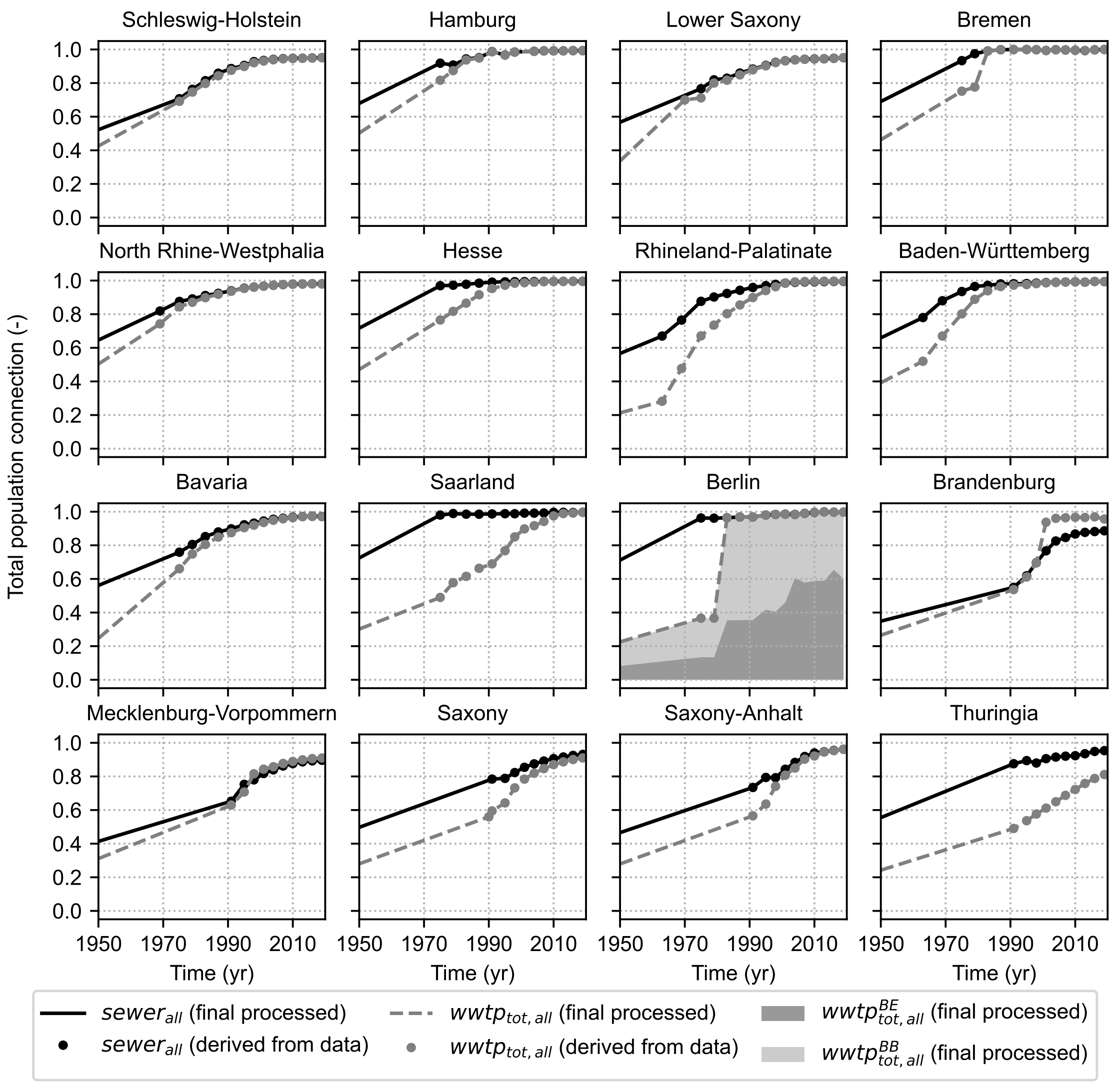


Figure S15. Data of total resident population connected to the sewer system ($sewer_{all}$) and to wastewater treatment ($wwtp_{tot,all}$), including public WWTPs, industrial and foreign WWTPs when data are available (Figure S16) and connection to WWTPs via cesspits when data are available (Figure S17), for each NUTS-1 region. The dots depict the estimates that are derived from the data are explained in Tables S11–S26. The continuous and dashed lines depict the final processed data obtained after performing linear interpolation between the years with available data (dots) and reconstructing the time series before the first years with data assuming that wastewater treatment starts in 1910 and sewer connection in 1880. For Berlin, we process the population connection to wastewater treatment for both the population connected to WWTPs located in Berlin ($wwtp^{BE}_{tot,all}$) and located in Brandenburg ($wwtp^{BB}_{tot,all}$). Source of the data: the data shown in this figure build on statistical data that mostly come from the Statistical Office of Germany and the different federal states, as well as additional sources, as detailed in Tables S11–S26.

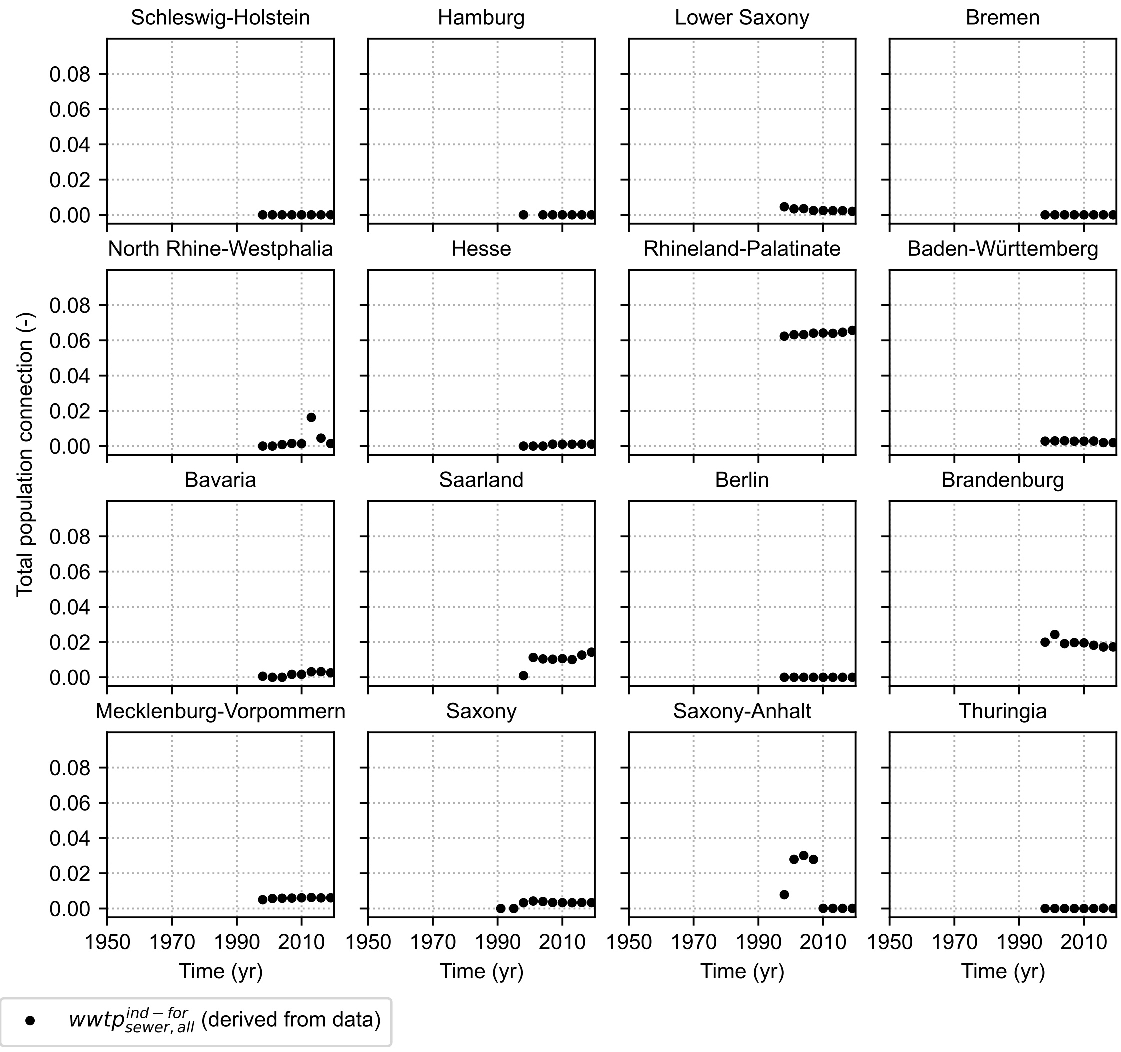


Figure S16. Data of resident population connected to industrial and foreign WWTPs for each NUTS-1 region ($wwtp_{sewer}^{ind-for}$). Data of population connection to industrial and foreign WWTPs are explicitly provided for the period 1998–2019, while before it is not explicitly reported whether WWTP connection data also account for industrial and foreign WWTPs in addition to public WWTPs. From the figure, we see that the percentage of population connected to industrial and foreign WWTPs is limited, since it is always lower than 3 %, apart from the state of Rhineland-Palatinate where it is around 6.5 %. Source of the data: the data shown in this figure build on statistical data that mostly come from the Statistical Office of Germany and the different federal states, as well as additional sources, as detailed in Tables S11–S26.

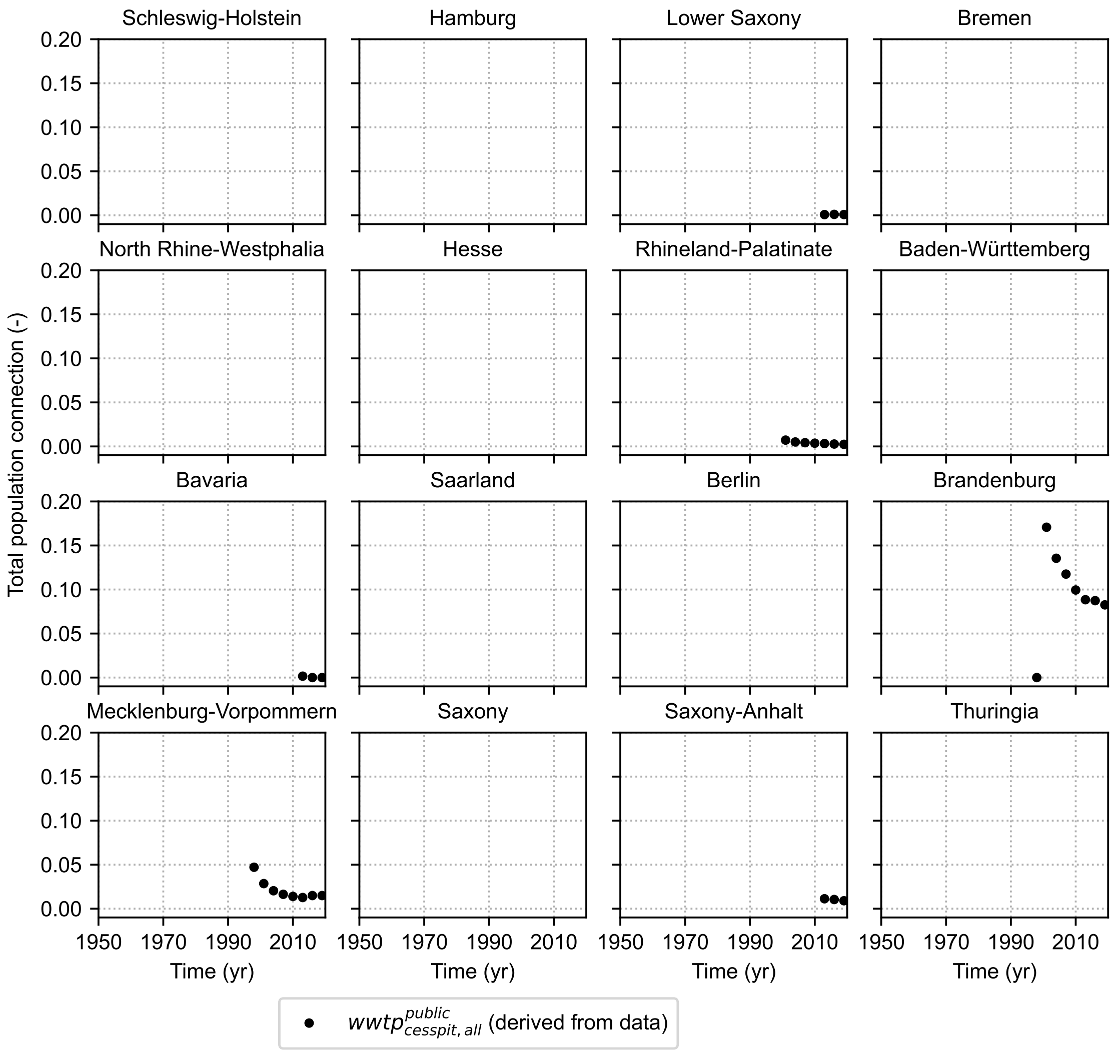


Figure S17. Data of resident population connected to WWTPs via cesspits for each NUTS-1 region ($wwtp_{cesspit}$). Source of the data: the data shown in this figure build on statistical data that mostly come from the Statistical Office of Germany and the different federal states, as well as additional sources, as detailed in Tables S11–S26. As explained in these tables, $wwtp_{cesspit}$ is not always directly provided in the statistical reports, but is sometimes estimated in this study.

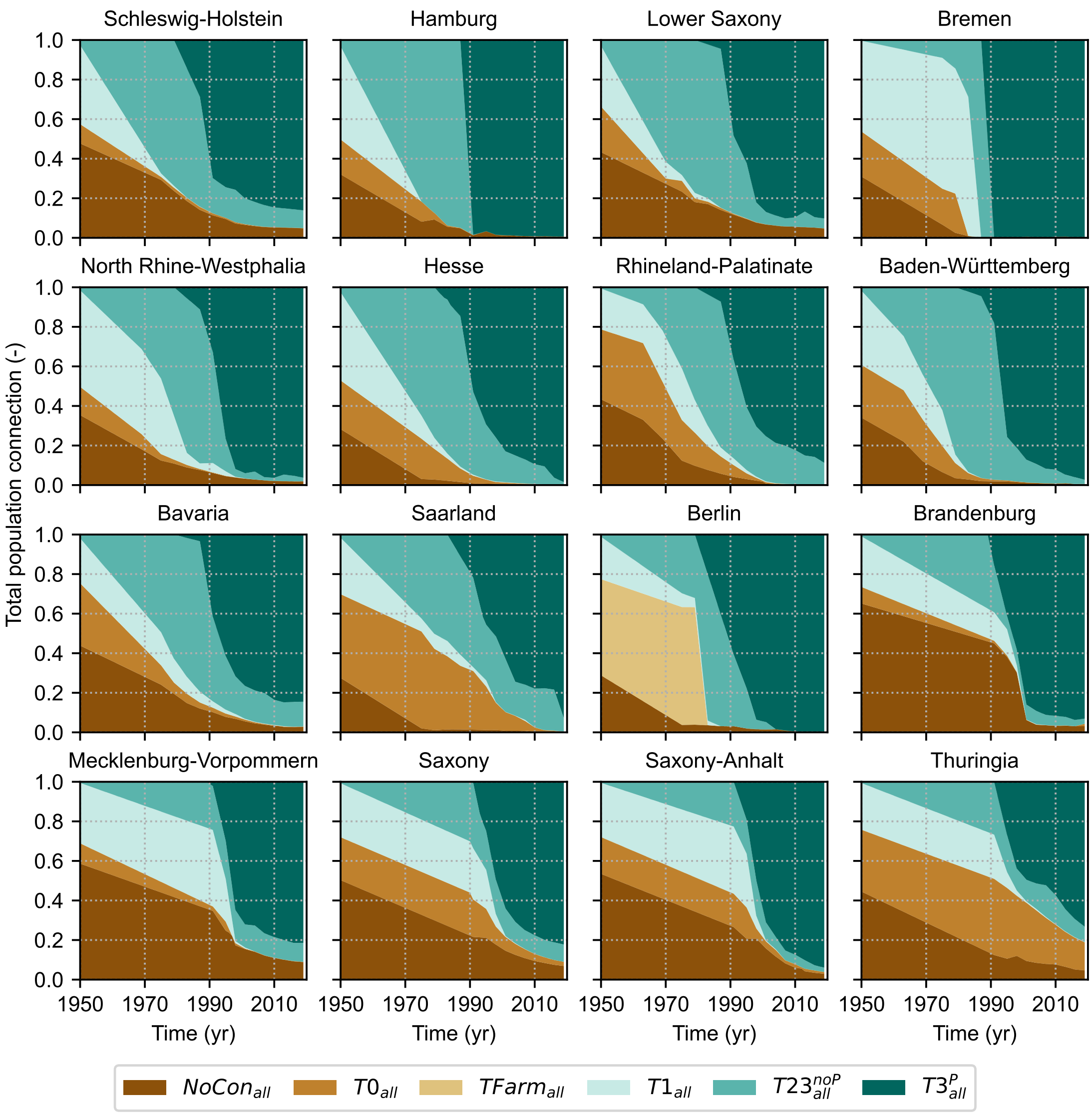


Figure S18. Processed fractions of total population connection at NUTS-1 level for the period 1950–2019, that were used to derive the N and P point sources data in this study. Specifically, the figure reports the fraction of total population that is not connected to the sewer system nor to WWTPs ($NoCon_{all}$) the fraction of total population connected to the sewer system but not to WWTPs ($T0_{all}$), the fraction of total population connected to sewage farms ($TFarm_{all}$), the fraction of total population connected to primary (mechanical) treatment ($T1_{all}$), the fractions of total population connected to secondary (biological) treatment and tertiary (advanced) treatment without targeted P removal ($T23_{all}^{noP}$), and the fraction of total population connected to tertiary treatment with targeted P removal ($T3_{all}^{P}$). Data source: the data shown in this figure were elaborated in this study building on data from the statistical offices of Germany and the different federal states, as well as additional sources, as detailed in Tables S11–S26.

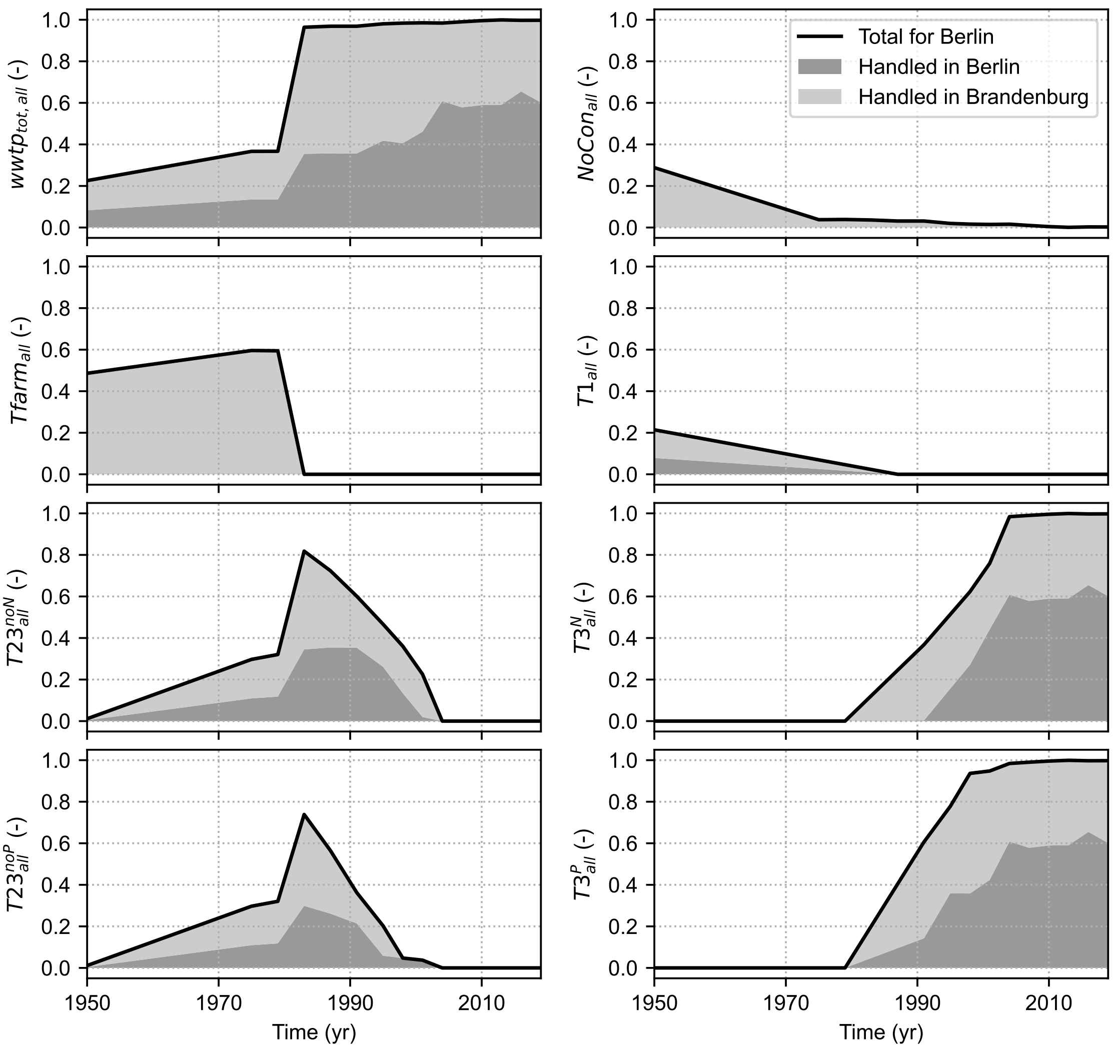


Figure S19. Processed data of connection of the total resident population of Berlin whose wastewater is handled in Berlin and in Brandenburg, namely fraction of total population connected to WWTP ($wwtp_{tot,all}$), fraction of total population not connected to the sewer system nor to WWTPs ($NoCon_{all}$), fraction of total population connected to sewage farms ($TFarm_{all}$), fraction of total population connected to primary treatment ($T1_{all}$), fraction of total population connected to secondary and tertiary treatment without targeted denitrification ($T23_{all}^{noN}$), fraction of total population connected to tertiary treatment with targeted denitrification ($T3_{all}^{N}$), fraction of total population connected to secondary and tertiary treatment without targeted P removal ($T23_{all}^{noP}$), and fraction of total population connected to tertiary treatment with targeted P removal ($T3_{all}^{P}$). We do not report the fraction of total population connected to the sewer system but not to WWTP ($T0_{all}$), since it is always equal to zero for Berlin. We consider that all wastewater that is collected in the sewer system but not treated in WWTPs is treated in sewage farms. Source of the data: the data shown in this figure were processed in this study, building on statistical data from the Statistical Office of Germany and from the federal states of Berlin and Brandenburg, as detailed in Table S21 and in Tables S37–S38.

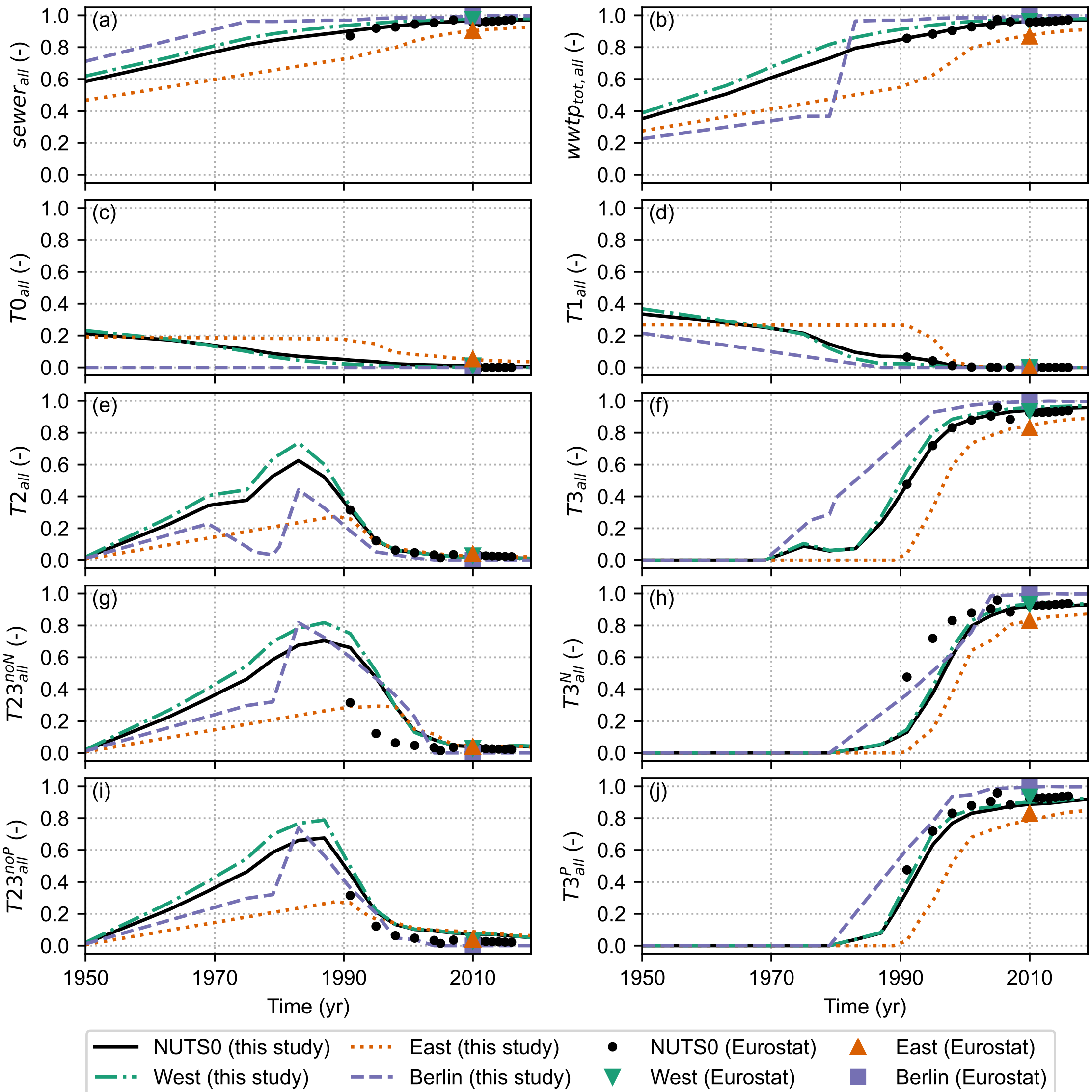


Figure S20. Data of total population connection to the sewer system and wastewater treatment processed in our study and provided by Eurostat (Eurostat, 2016, 2023) aggregated at NUTS-0 level, at the level of West Germany (excluding Berlin), at the level of East Germany (excluding Berlin) and for the state of Berlin. The notations are defined in Table S8. To construct the population connection to secondary treatment ($T2_{all}$ in panel e) and tertiary treatment ($T3_{all}$ in panel f) in this study, we fill the missing data for the early years assuming that tertiary treatment started in 1970 in West Germany as in Morée et al. (2013) and in 1990 in East Germany. We then perform linear interpolation until the first year with data. Connection to secondary treatment is calculated as the difference between 1 and the sum of the other population connection data. The Eurostat dataset only provides $T2_{all}$ and $T3_{all}$ and therefore the values provided are taken as $T23_{all}^{noN}$ and $T23^{noP}$ (panels g,h) and $T3_{all}^{N}$ and $T3_{all}^{P}$ (panels i,j), respectively. This leads to an underestimation of $T23^{noN}$ and $T23_{all}^{noP}$ and an overestimation of $T3_{all}^{N}$ and $T3_{all}^{P}$ compared to the data processed in this study as shown in panels (g,h,i,j). Source of the data: Eurostat data at NUTS-0 level are available from Eurostat (2023) and at NUTS-2 level from Eurostat (2016). The data from this study build on statistical data that come from the statistical offices of Germany and the different federal states, as well as additional sources, as detailed in Tables S11–S26.

S7 Estimation of urban and rural population connection to wastewater treatment at NUTS-1 level

S7.1 Connection of urban and rural population to wastewater treatment at NUTS-1 level for all NUTS-1 regions but Berlin

To estimate the fraction of both urban and rural population connection to the different types of treatment from the data that refer to the total population only (described in Sect. S6), we assume that the wastewater handling system is more advanced in urban areas. For year t and the i-th NUTS-1 region, we first estimate the urban and rural population connection to the type of treatment that has the highest efficiency of N and P removal, which is tertiary treatment with targeted nutrient (N and P) removal, denoted as $T3_X^{Nutri}(t,i)$ $(-)$, where X, can be urb (urban component), or rur (rural component). For this, we distribute the total population connected to this treatment type between the urban and rural population, considering that urban population has precedence over rural population as follows:

$$T3_{urb}^{Nutri}(t,i) = min\left(\frac{Pop_{all}(t,i)}{Pop_{urb}(t,i)} T3_{all}^{Nutri}(t,i), 1\right)$$

$$T3_{rur}^{Nutri}(t,i) = \frac{Pop_{all}(t,i) T3_{all}^{Nutri}(t,i) - Pop_{urb}(t,i) T3_{urb}^{Nutri}(t,i)}{Pop_{rur}(t,i)} \tag{S14}$$

where $Pop_X(t)$ is the total ($X = all$), urban ($X = urb$) and rural ($X = rur$) population count. From Eq. S14, part of the rural population can be connected to tertiary treatment with targeted nutrient removal only when the entire urban population is connected to this treatment type.

We then estimate the urban and rural population connection to the other types of treatment in descending order of technological advancement, starting with secondary and tertiary treatment without targeted N and P removal (denoted as $T23_X^{noNutri}(t,i)$ $(-)$), then primary treatment (denoted as $T1_X(t,i)$ $(-)$), treatment in sewage farms (denoted as $T_{farm,X}(t,i)$ $(-)$), and finally no treatment (denoted as $T0_X(t,i)$ $(-)$, as follows:

$$T23_{urb}^{noNutri}(t,i) = min\left(\frac{Pop_{all}(t,i)}{Pop_{urb}(t,i)} T23_{all}^{noNutri}(t,i), 1 - T3_{urb}^{Nutri}(t,i)\right)$$

$$T23_{rur}^{noNutri}(t,i) = \frac{Pop_{all}(t,i) T23_{all}^{noNutri}(t,i) - Pop_{urb}(t,i) T23_{urb}^{noNutri}(t,i)}{Pop_{rur}(t,i)} \tag{S15}$$

$$T1_{urb}(t,i) = min\left(\frac{Pop_{all}(t,i)}{Pop_{urb}(t,i)} T1_{all}(t,i), 1 - T3_{urb}^{Nutri}(t,i) - T23_{urb}^{noNutri}(t,i)\right)$$

$$T1_{rur}(t,i) = \frac{Pop_{all}(t,i) T1_{all}(t,i) - Pop_{urb}(t,i) T1_{urb}(t,i)}{Pop_{rur}(t,i)} \tag{S16}$$

$$T_{farm,urb}(t,i) = min\left(\frac{Pop_{all}(t,i)}{Pop_{urb}(t,i)} T_{farm,all}(t,i), 1 - T3_{urb}^{Nutri}(t,i) - T23_{urb}^{noNutri}(t,i) - T1_{urb}(t,i)\right)$$

$$T_{farm,rur}(t,i) = \frac{Pop_{all}(t,i) T_{farm,all}(t,i) - Pop_{urb}(t,i) T_{farm,urb}(t,i)}{Pop_{rur}(t,i)} \tag{S17}$$

$$T0_{urb}(t,i) = min\left(\frac{Pop_{all}(t,i)}{Pop_{urb}(t,i)}T0_{all}(t,i), 1 - T3_{urb}^{Nutri}(t,i) - T23_{urb}^{noNutri}(t,i) - T1_{urb}(t,i) - T_{farm,urb}(t,i)\right)$$

$$T0_{rur}(t,i) = \frac{Pop_{all}(t,i)T0_{all}(t,i) - Pop_{urb}(t,i)T0_{urb}(t,i)}{Pop_{rur}(t,i)} \tag{S18}$$

Equations S14–S18 ensure that the sum of the different population connection to the different types of treatment is always less than 1 for both the urban and rural population and that the sum of the urban and rural population connected to each type of treatment is equal to the total population connected to that type of treatment. We also note that, the sum of $T3_X^{Nutri}(t,i)$ and $T23_X^{noNutri}(t,i)$ takes the same value, whether $Nutri = N$ or $Nutri = P$, which is equal to the population connection to secondary and tertiary treatment. This is why Eq. S16–S18 can be written interchangeably using $Nutri = N$ or $Nutri = P$. The resulting fractions of urban and rural connected to the different treatment types can be visualized in Fig S21–S24.

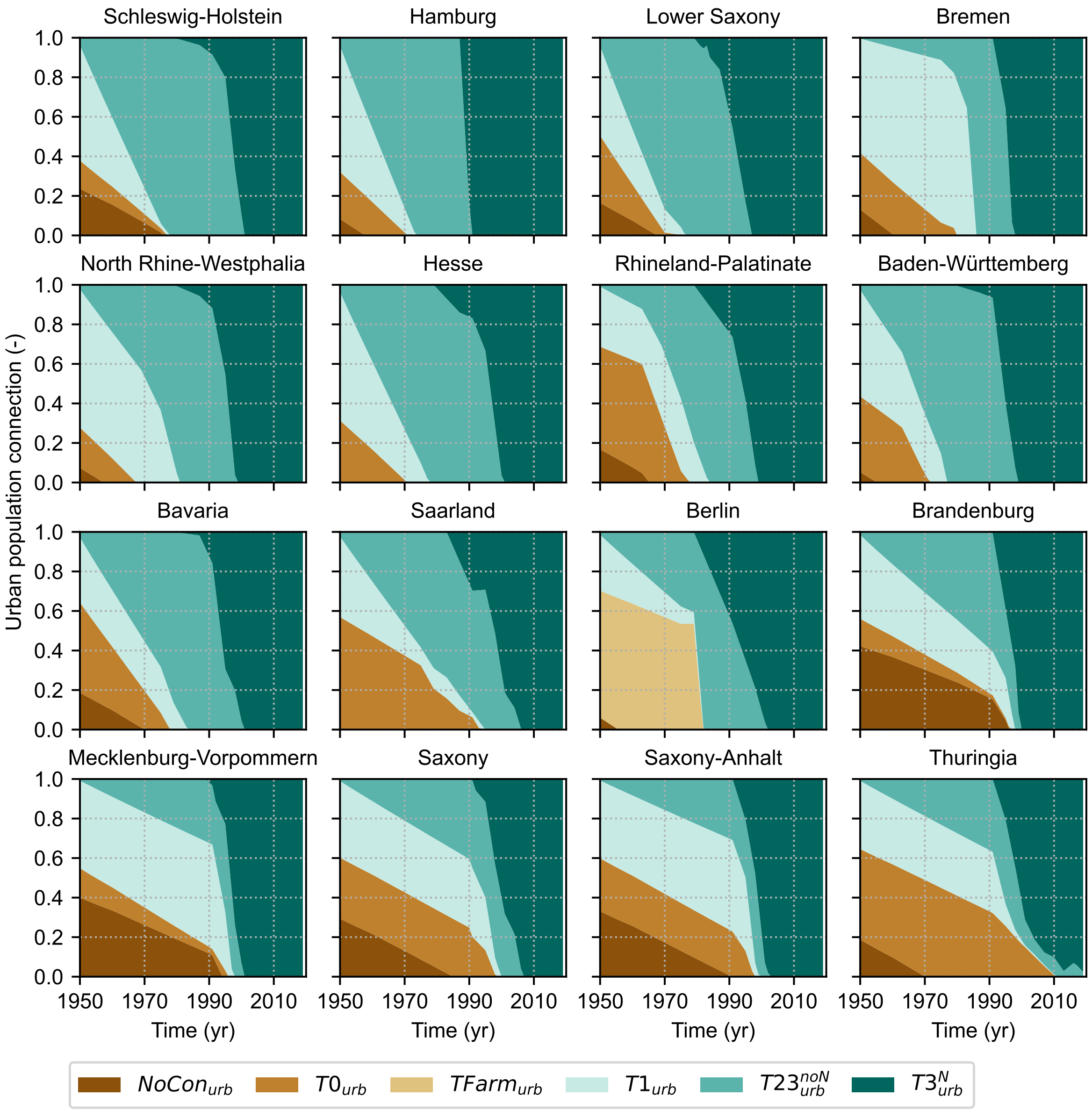


Figure S21. Estimated fractions of **urban** population connection at NUTS-1 level for the period 1950–2019, that were used to derive the N and P point sources data in this study. Specifically, the figure reports the fraction of urban population that is not connected to the sewer system nor to WWTPs ($NoCon_{urb}$) the fraction of urban population connected to the sewer system but not to WWTPs ($T0_{urb}$), the fraction of urban population connected to sewage farms ($TFarm_{urb}$), the fraction of urban population connected to primary (mechanical) treatment ($T1_{urb}$), the fractions of urban population connected to secondary (biological) treatment and tertiary (advanced) treatment without targeted N removal ($T23^{noN}_{urb}$), and the fraction of urban population connected to tertiary treatment with targeted N removal ($T3^{N}_{urb}$). Data source: the data shown in this figure were elaborated in this study building on total population connection data from the statistical offices of Germany and the different federal states, as well as additional sources, as detailed in Tables S11–S26. The connection data for the urban population were derived using Eq. S14–S18.

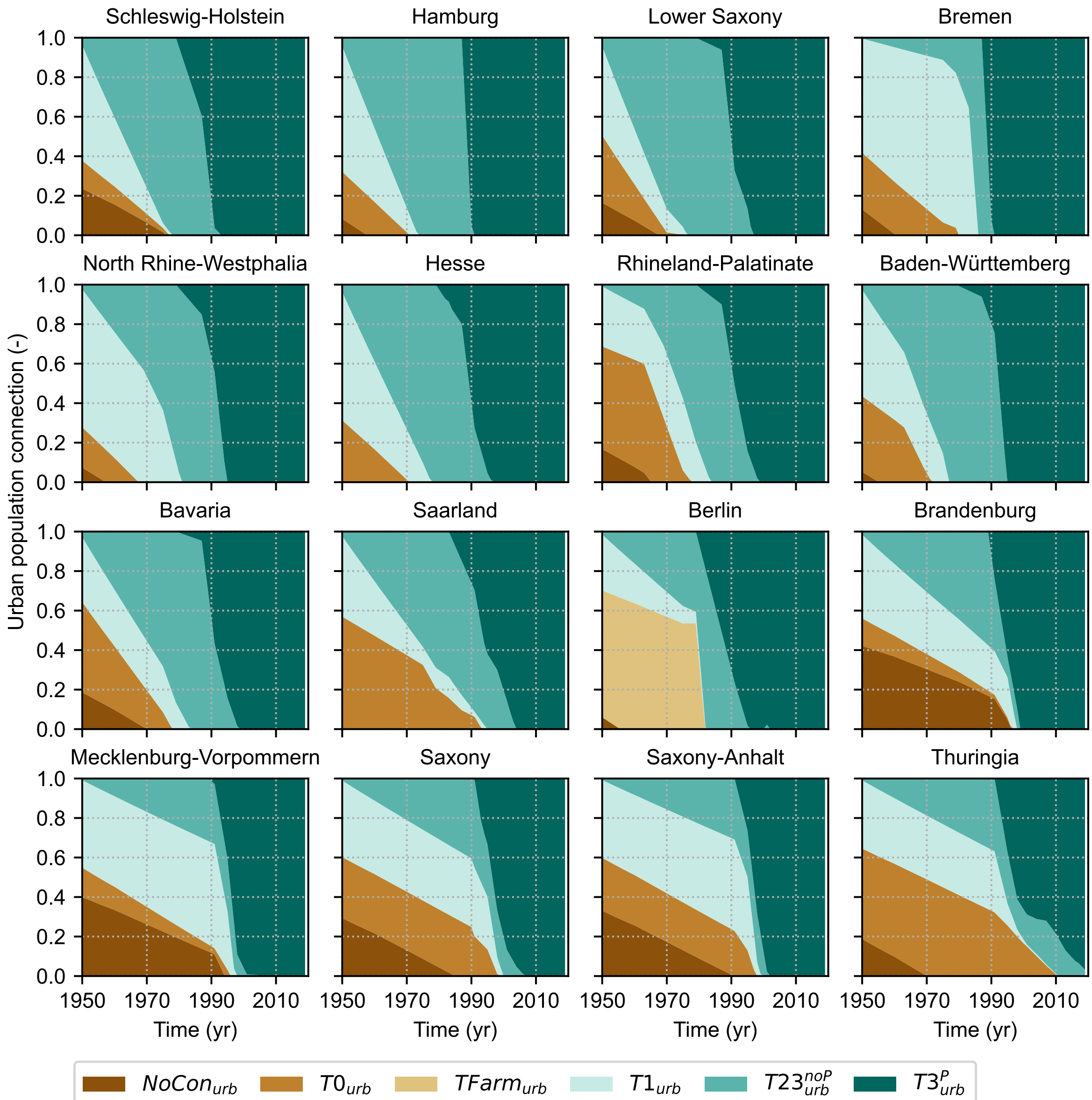


Figure S22. Estimated fractions of **urban** population connection at NUTS-1 level for the period 1950–2019, that were used to derive the N and P point sources data in this study. Specifically, the figure reports the fraction of urban population that is not connected to the sewer system nor to WWTPs ($NoCon_{urb}$) the fraction of urban population connected to the sewer system but not to WWTPs ($T0_{urb}$), the fraction of urban population connected to sewage farms ($TFarm_{urb}$), the fraction of urban population connected to primary (mechanical) treatment ($T1_{urb}$), the fractions of urban population connected to secondary (biological) treatment and tertiary (advanced) treatment without targeted P removal ($T23^{noP}_{urb}$), and the fraction of urban population connected to tertiary treatment with targeted P removal ($T3^{P}_{urb}$). Data source: the data shown in this figure were elaborated in this study building on total population connection data from the statistical offices of Germany and the different federal states, as well as additional sources, as detailed in Tables S11–S26. The connection data for the urban population were derived using Eq. S14–S18.

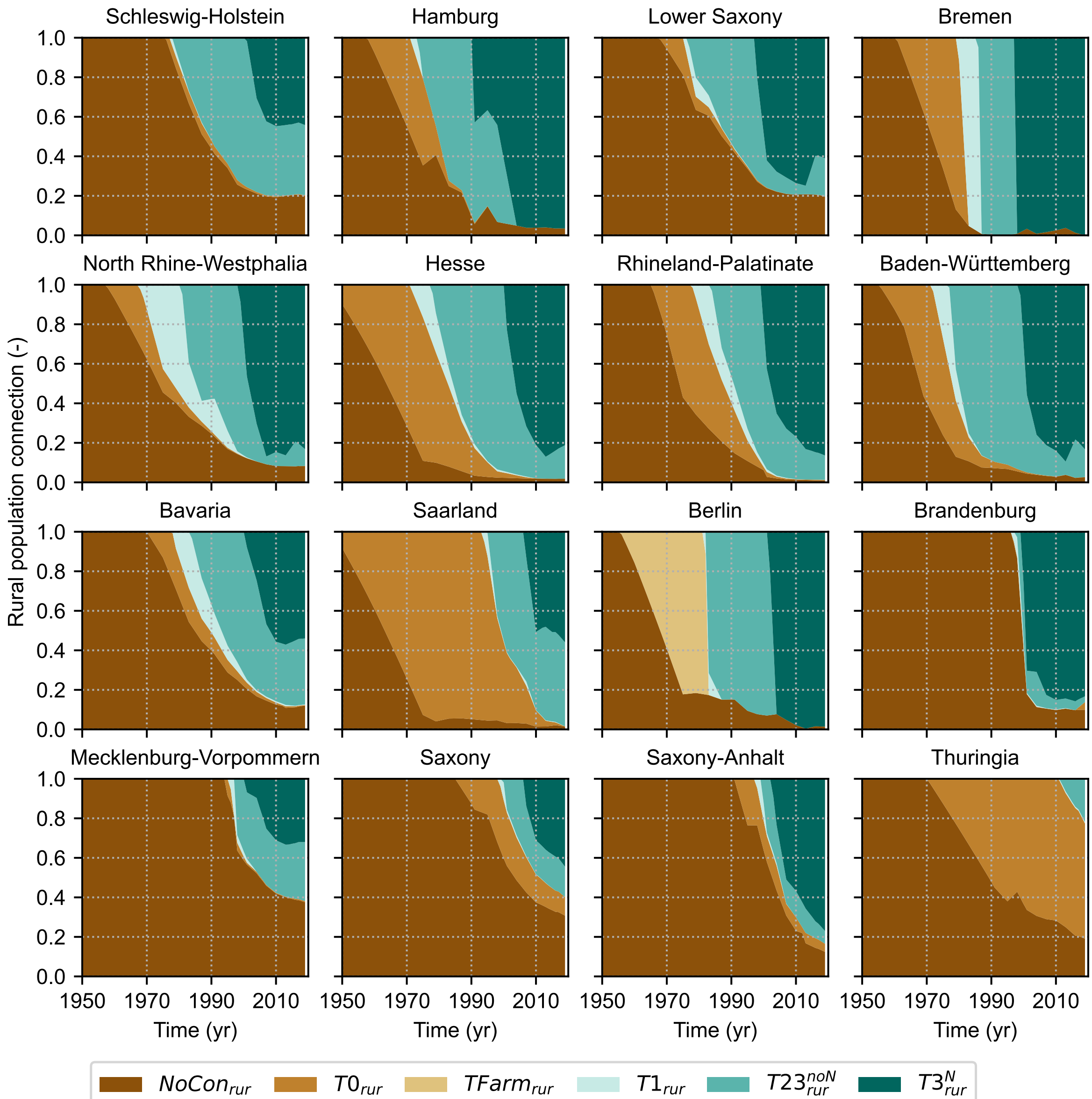


Figure S23. Estimated fractions of **rural** population connection at NUTS-1 level for the period 1950–2019, that were used to derive the N and P point sources data in this study. Specifically, the figure reports the fraction of rural population that is not connected to the sewer system nor to WWTPs ($NoCon_{rur}$) the fraction of rural population connected to the sewer system but not to WWTPs ($T0_{rur}$), the fraction of rural population connected to sewage farms ($TFarm_{rur}$), the fraction of rural population connected to primary (mechanical) treatment ($T1_{rur}$), the fractions of rural population connected to secondary (biological) treatment and tertiary (advanced) treatment without targeted N removal ($T23^{noN}_{rur}$), and the fraction of rural population connected to tertiary treatment with targeted N removal ($T3^{N}_{rur}$). Data source: the data shown in this figure were elaborated in this study building on total population connection data from the statistical offices of Germany and the different federal states, as well as additional sources, as detailed in Tables S11–S26. The connection data for the rural population were derived using Eq. S14–S18.

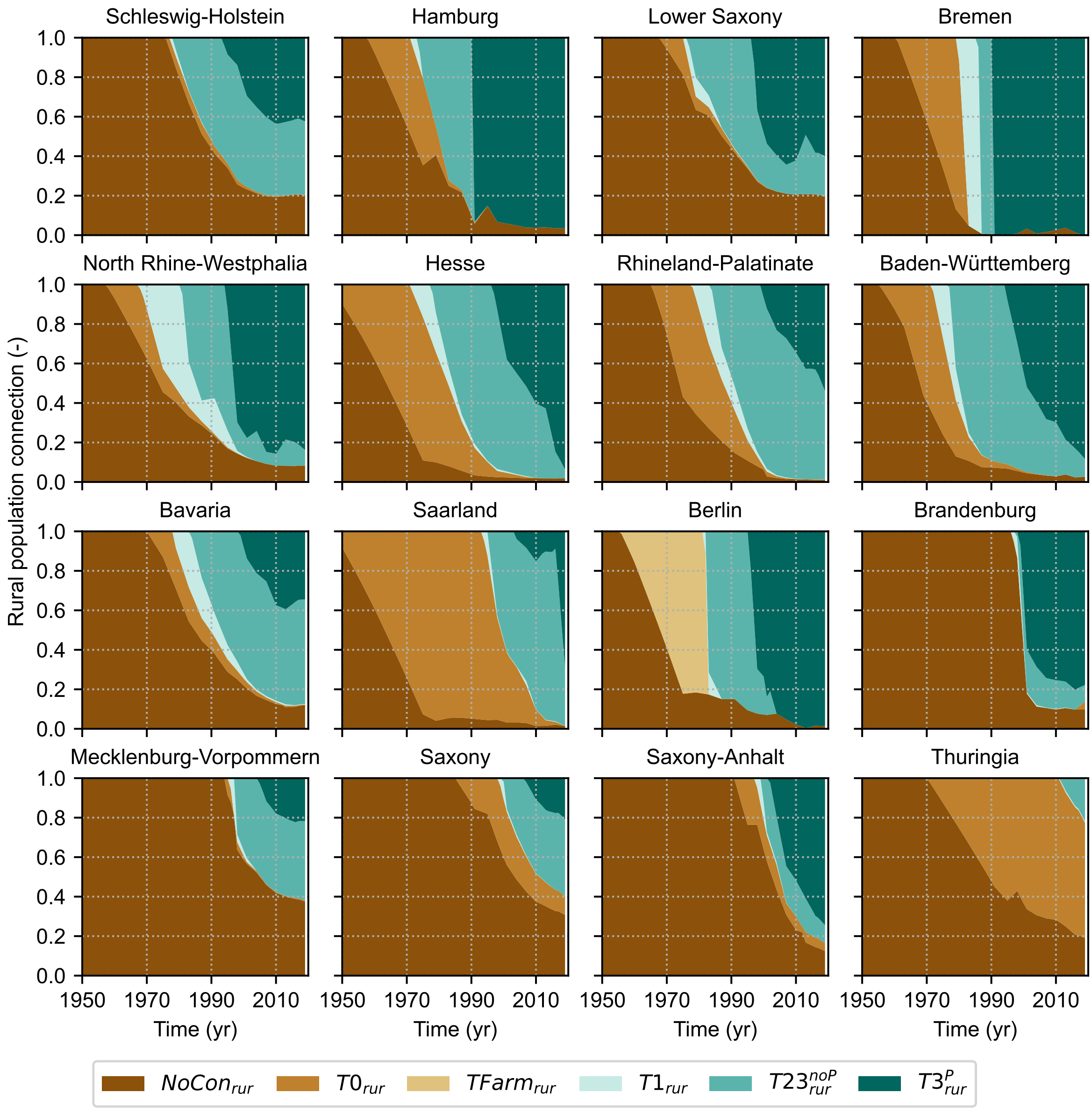


Figure S24. Estimated fractions of **rural** population connection at NUTS-1 level for the period 1950–2019, that were used to derive the N and P point sources data in this study. Specifically, the figure reports the fraction of rural population that is not connected to the sewer system nor to WWTPs ($NoCon_{rur}$) the fraction of rural population connected to the sewer system but not to WWTPs ($T0_{rur}$), the fraction of rural population connected to sewage farms ($TFarm_{rur}$), the fraction of rural population connected to primary (mechanical) treatment ($T1_{rur}$), the fractions of rural population connected to secondary (biological) treatment and tertiary (advanced) treatment without targeted P removal ($T23^{noP}_{rur}$), and the fraction of rural population connected to tertiary treatment with targeted P removal ($T3^{P}_{rur}$). Data source: the data shown in this figure were elaborated in this study building on total population connection data from the statistical offices of Germany and the different federal states, as well as additional sources, as detailed in Tables S11–S26. The connection data for the rural population were derived using Eq. S14–S18.

S7.2 Urban and rural population and their connection to wastewater treatment for the NUTS-1 region of Berlin

S7.2.1 Total population connection to tertiary and secondary treatment in Berlin

For the NUTS-1 region of Berlin, we distinguish four different categories of secondary/tertiary treatment, namely tertiary treatment with both targeted N and P removal, tertiary treatment with only targeted N removal, tertiary treatment with only targeted P removal, and secondary and tertiary treatment without both targeted N and P removal. We assume that tertiary WWTPs are more likely to perform treatment including both targeted N and P removal, or no targeted N and P removal at all, rather than performing treatment that removes specifically only one of the two nutrients (N or P). We then derive the corresponding equations for the total population of Berlin connected to secondary and tertiary treatment in Berlin ($j = BE$) or in Brandenburg ($j = BB$):

$$\begin{aligned}
&T3_{all}^{N-P,j}(t,BE) = min(T3_{all}^{N,j}(t,BE), T3_{all}^{P,j}(t,BE))\\
&T3_{all}^{N-noP,j}(t,BE) = max(T3_{all}^{N,j}(t,BE) - T3_{all}^{P,j}(t,BE), 0)\\
&T3_{all}^{noN-P,j}(t,BE) = max(T3_{all}^{P,j}(t,BE) - T3_{all}^{N,j}(t,BE), 0)\\
&T23_{all}^{noN-noP,j}(t,BE) = min(T23_{all}^{noN,j}(t,BE), T23_{all}^{noP,j}(t,BE))
\end{aligned} \tag{S19}$$

where $T3_{all}^{N-P,j}(t,BE)$ $(-)$, $T3_{all}^{N-noP,j}(t,BE)$ $(-)$, $T3_{all}^{noN-P,j}(t,BE)$ $(-)$, and $T23_{all}^{noN-noP,j}(t,BE)$ $(-)$ are the fractions of total population of Berlin whose wastewater is handled in Berlin ($j = BE$) or in Brandenburg ($j = BB$), that is connected to tertiary treatment with both targeted N and P removal, tertiary treatment with only targeted N removal, tertiary treatment with only targeted P removal, and secondary and tertiary treatment without both N and P removal, respectively. The variables defined in Equation S19 will be useful to disaggregate the urban and rural population connection estimates of Berlin (obtained as detailed in Sect. S7.2.2) between the parts of Berlin whose wastewater is handled in Berlin and Brandenburg (see Sect S7.2.3).

S7.2.2 Connection of the urban and rural population of Berlin

We assess the urban and rural population connection to each type of treatment for the entire Berlin from Eq. S16 for primary treatment, from Eq. S17 for sewage farms, and from Eq. S18 for no treatment. For tertiary treatment with both targeted N and P removal ($T3_X^{N-P}(t,BE)$ $(-)$), tertiary treatment with only targeted N removal ($T3_X^{N-noP}(t,BE)$ $(-)$), tertiary treatment with only targeted P removal ($T3_X^{noN-P}(t,BE)$ $(-)$), and secondary and tertiary treatment without both N and P removal $T23_X^{noN-noP}(t,BE)$ $(-)$), we use the following equations, that are similar to that of Sect. S7.1:

$$\begin{aligned}
&T3_{urb}^{N-P}(t,BE) = min\left(\frac{Pop_{all}(t,BE)}{Pop_{urb}(t,BE)} T3_{all}^{N-P}(t,BE), 1\right)\\
&T3_{rur}^{N-P}(t,BE) = \frac{Pop_{all}(t,BE) T3_{all}^{N-P}(t,BE) - Pop_{urb}(t,BE) T3_{urb}^{N-P}(t,BE)}{Pop_{rur}(t,BE)}
\end{aligned} \tag{S20}$$

$$T3^{N-noP}_{urb}(t,BE) = min\left(\frac{Pop_{all}(t,BE)}{Pop_{urb}(t,BE)}T3^{N-noP}_{all}(t,BE), 1 - T3^{N-P}_{urb}(t,BE)\right)$$
$$T3^{N-noP}_{rur}(t,BE) = \frac{Pop_{all}(t,BE)T3^{N-noP}_{all}(t,BE) - Pop_{urb}(t,BE)T3^{N-noP}_{urb}(t,BE)}{Pop_{rur}(t,BE)} \tag{S21}$$

$$T3^{noN-P}_{urb}(t,BE) = min\left(\frac{Pop_{all}(t,BE)}{Pop_{urb}(t,BE)}T3^{noN-P}_{all}(t,BE), 1 - T3^{N-P}_{urb}(t,BE) - T3^{N-noP}_{urb}(t,BE)\right)$$
$$T3^{noN-P}_{rur}(t,BE) = \frac{Pop_{all}(t,BE)T3^{noN-P}_{all}(t,BE) - Pop_{urb}(t,BE)T3^{noN-P}_{urb}(t,BE)}{Pop_{rur}(t,BE)} \tag{S22}$$

$$T23^{noN-noP}_{urb}(t,BE) = min\left(\frac{Pop_{all}(t,BE)}{Pop_{urb}(t,BE)}T3^{noN-noP}_{all}(t,BE), 1 - T3^{N-P}_{urb}(t,BE) - T3^{N-noP}_{urb}(t,BE) - T3^{noN-P}_{urb}(t,BE)\right)$$
$$T23^{noN-noP}_{rur}(t,BE) = \frac{Pop_{all}(t,BE)T3^{noN-noP}_{all}(t,BE) - Pop_{urb}(t,BE)T3^{noN-noP}_{urb}(t,BE)}{Pop_{rur}(t,BE)} \tag{S23}$$

Similar to Eq. S14, we first estimate the urban and rural population connection to the type of treatment that has the highest efficiency of N and P removal, which is tertiary treatment with both targeted N and P removal, considering that urban population has precedence over rural population (Eq. S20). We then estimate the connection to tertiary treatment with only targeted N removal (Eq. S21). This gives precedence to urban population connection to tertiary treatment with only targeted N removal over tertiary treatment with only targeted P removal. We assess the connection to secondary and tertiary treatment without both targeted N and P removal last, since it is the least advanced type of treatment (Eq. S23). Importantly, Eq. S20–S23 ensures that the following condition is satisfied:

$$\begin{aligned} T3^{N-P}_{X}(t,BE) + T3^{N-noP}_{X}(t,BE) + T3^{noN-P}_{urb}(t,BE) + T3^{noN-noP}_{X}(t,BE) &= T3^{N}_{X}(t,BE) + T23^{noN}_{X}(t,BE) \\ &= T3^{P}_{X}(t,BE) + T23^{noP}_{X}(t,BE) \end{aligned} \tag{S24}$$

S7.2.3 Disaggregation of the connection of the urban and rural population of Berlin between the parts of Berlin whose wastewater is handled in Berlin and in Brandenburg

In this section, we explain how we disaggregrate the estimates of rural and urban population for Berlin of Eq. S20–S23 between the parts of Berlin whose wastewater is handled in Berlin and in Brandenburg. For a given type of treatment, we distribute the urban and rural population of Berlin connected to WWTPs performing this type of treatment proportionally to the ratio of the total population connected to WWTPs performing this type of treatment located in Berlin (or Brandenburg) to the total population connected to WWTPs performing this type of treatment in both Berlin and Brandenburg as follows:

$$F^{j}_{X}(t,BE) = F_{X}(t,BE)\frac{F^{j}_{all}(t,BE)}{F_{all}(t,BE)} \tag{S25}$$

where $F_X^j(t,BE)$ $(-)$ is the fraction of the urban ($X = urb$) or rural ($X = rur$) population of Berlin whose wastewater is handled in Berlin ($j = BE$) or in Brandenburg ($j = BB$), and that is connected to the type of treatment F, that is, to no treatment ($F = T0$), sewage farm ($F = T_{farm}$), primary treatment ($F = T1$), secondary and tertiary treatment without both targeted N and P removal ($F = T23^{noN-noP}$), tertiary treatment with targeted N removal but without targeted P removal ($F = T3^{N-noP}$), tertiary treatment with targeted P removal but without targeted N removal ($F = T3^{noN-P}$), or tertiary treatment with both targeted N and P removal ($T3^{N-P}$); $F_X(t,BE)$ $(-)$ is the fraction of urban or rural population of Berlin that is connected to the type of treatment F (in both Berlin and Brandenburg); $F_{all}^j(t,BE)$ $(-)$ is the fraction of total population of Berlin whose wastewater is handled in Berlin ($j = BE$) or in Brandenburg ($j = BB$) that is connected to the type of treatment F; $F_{all}(t,BE)$ $(-)$ is the fraction of the total population of Berlin that is connected to the type of treatment F (in both Berlin and Brandenburg). Equation S25 ensures that the population of Berlin connected to a given treatment type ($F_X(t,BE)$) is equal to the sum of the population of Berlin connected to a given treatment type in Berlin and in Brandenburg ($F_X^{BE}(t,BE) + F_X^{BB}(t,BE)$) for urban ($X = urb$) and rural ($X = rur$) population.

The fraction of urban and rural population of Berlin whose wastewaster in handled in Berlin or in Brandenburg and that is connected to secondary treatment and tertiary treatment without targeted N removal ($T23_X^{noN,j}(t,BE)$ $(-)$), to secondary treatment and tertiary treatment without targeted P removal ($T23_X^{noP,j}(t,BE)$ $(-)$), to tertiary treatment with targeted N removal ($T3_X^{N,j}(t,BE)$ $(-)$), and to tertiary treatment with targeted P removal ($T3_X^{P,j}(t,BE)$ $(-)$), that are required to run the N and P point sources model, are then calculated from the variables of Eq. S25 as follows:

$$
\begin{aligned}
&T23_X^{noN,j}(t,BE) = T3_X^{noN-P,j}(t,BE) + T23_X^{noN-noP,j}(t,BE)\\
&T23_X^{noP,j}(t,BE) = T3_X^{N-noP,j}(t,BE) + T23_X^{noN-noP,j}(t,BE)\\
&T3_X^{N,j}(t,BE) = T3_X^{N-P,j}(t,BE) + T3_X^{N-noP,j}(t,BE)\\
&T3_X^{P,j}(t,BE) = T3_X^{N-P,j}(t,BE) + T3_X^{noN-P,j}(t,BE)
\end{aligned}
\tag{S26}
$$

We verify the consistency of Eq. S26, that is, the following condition is met:

$$
T3_X^{N,j}(t,BE) + T23_X^{noN,j}(t,BE) = T3_X^{P,j}(t,BE) + T23_X^{noP,j}(t,BE) \tag{S27}
$$

The resulting urban and rural population connection data for Berlin, including the parts whose wastewater is handled in Berlin and Brandenburg, are reported in Fig. S25–S26. From the population connection variables of Eq. S25 and S26, that refer to the urban and rural population of the entire Berlin, we can derive population connection variables that refer to the urban and rural population whose wastewater is handled in Berlin (Brandenburg) by applying the ratio of the urban population of the entire Berlin to the urban population whose wastewater in handled in Berlin (Brandenburg, respectively). The urban and rural population of Berlin whose wastewater is handled in Berlin and in Brandenburg are estimated as detailed in Sect. S7.2.4.

S7.2.4 Total, urban and rural population of Berlin whose wastewater is handled in Berlin and in Brandenburg

Since we consider that the part of the population of Berlin whose wastewater is handled in Brandenburg is all connected to WWTPs, we can estimate the total, urban and rural population of Berlin whose wastewater is handled in Brandenburg, denoted as $Pop_X^{BB}(t,BE)$ from the population of the entire Berlin, denoted as $Pop_X(t,BE)$, as follows:

$$Pop_X^{BB}(t,BE) = Pop_X(t,BE)(T3_X^{N,BB}(t,BE) + T23_X^{noN,BB}(t,BE) + T1_X^{BB}(t,BE)) \tag{S28}$$

The total, urban and rural population of Berlin whose wastewater is handled in Berlin (that can be not only connected to WWTPs, but also to sewage farms or that may not be handled in the public wastewater system), denoted as $Pop_X^{BE}(t,BE)$ is then estimated as the difference between the population of the entire Berlin and the population of Berlin whose wastewater is handled in Brandenburg as follows:

$$Pop_X^{BE}(t,BE) = Pop_X(t,BE) - Pop_X^{BB}(t,BE) \tag{S29}$$

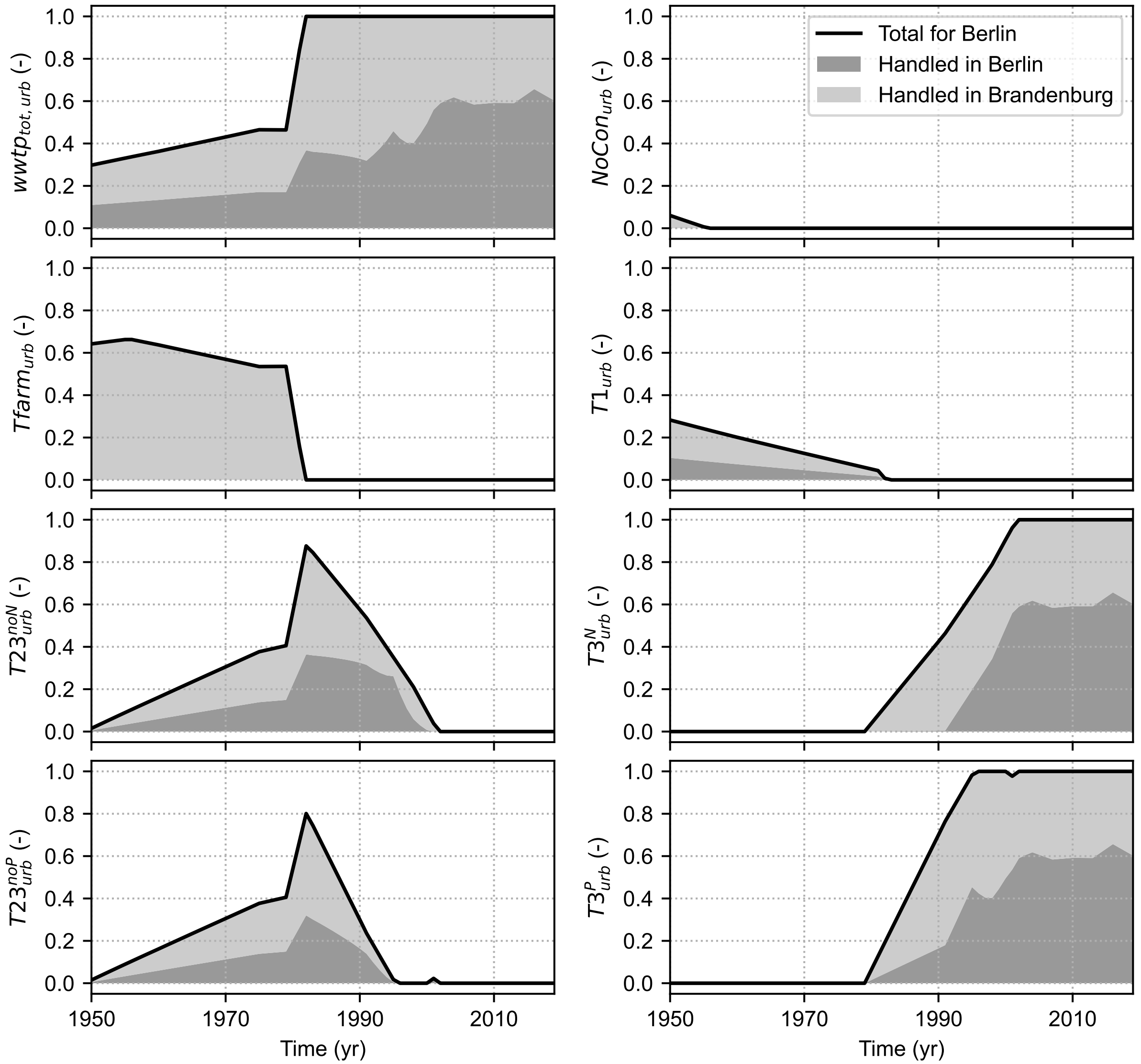


Figure S25. Processed connection data of the urban population of Berlin whose wastewater is handled in Berlin and in Brandenburg, namely fraction of urban population connected WWTP ($wwtp_{tot,urb}$), fraction of urban population not connected to the sewer system nor to WWTPs ($NoCon_{urb}$), fraction of urban population connected to sewage farms ($TFarm_{urb}$), fraction of urban population connected to primary treatment ($T1_{urb}$), fraction of urban population connected to secondary and tertiary treatment without targeted denitrification ($T23^{noN}_{urb}$), fraction of urban population connected to tertiary treatment with targeted denitrification ($T3^{N}_{urb}$), fraction of urban population connected to secondary and tertiary treatment without targeted P removal ($T23^{noP}_{urb}$), and fraction of urban population connected to tertiary treatment with targeted P removal ($T3^{P}_{urb}$). We do not report the fraction of urban population connected to the sewer system but not to WWTP ($T0_{urb}$), since it is always equal to zero for Berlin. We consider that all wastewater that is collected in the sewer system but not treated in WWTPs is treated in sewage farms. Source of the data: the data shown in this figure were processed in this study, building on total population connection data from the Statistical Office of Germany and from the federal states of Berlin and Brandenburg (see Table S21 and Tables S37–S38). The connection data for urban population were derived using as explained in Sect. S7.2.1-S7.2.3.

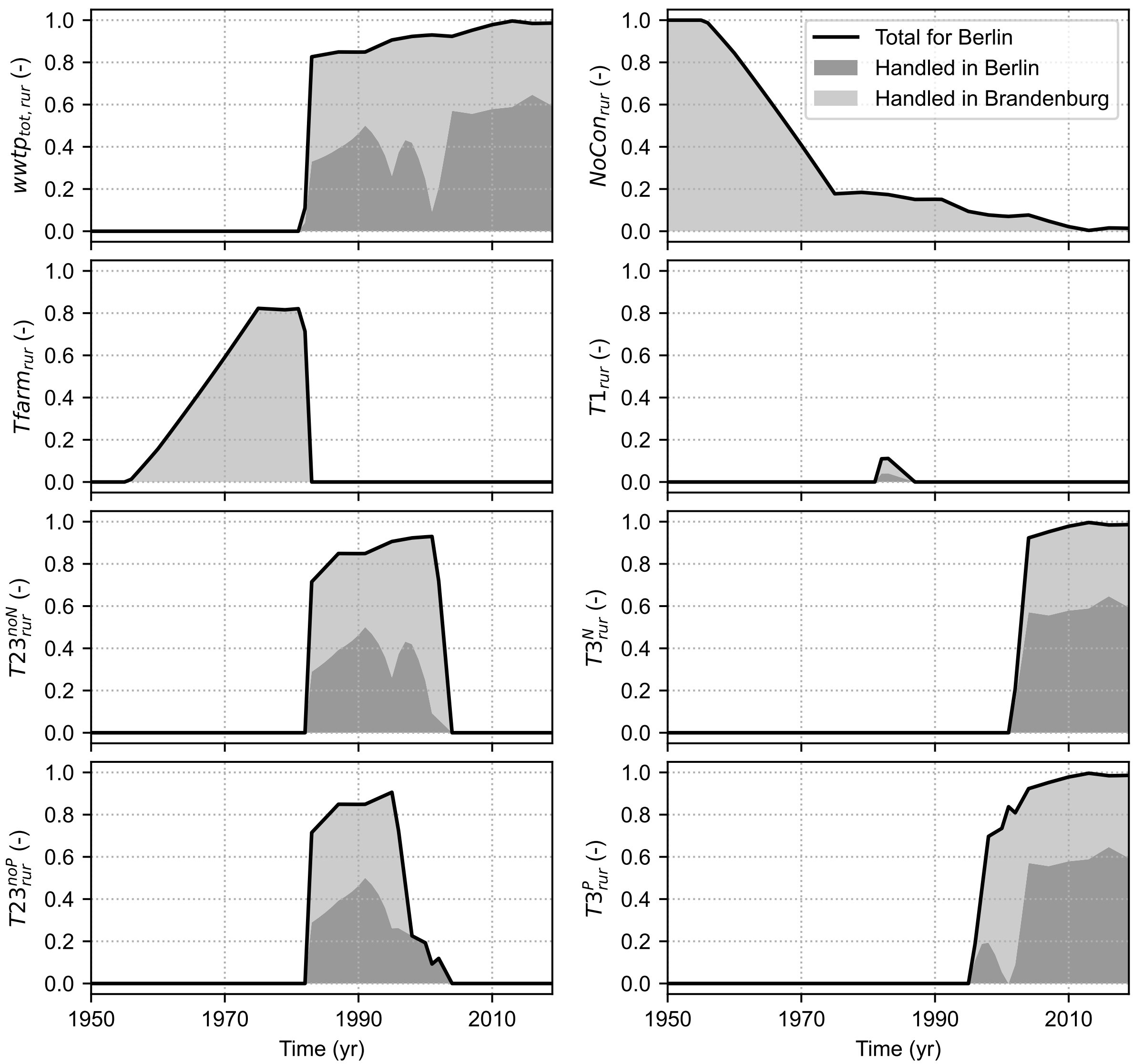


Figure S26. Processed connection data of the rural population of Berlin whose wastewater is handled in Berlin and in Brandenburg, namely fraction of rural population connected WWTP ($wwtp_{tot,rur}$), fraction of rural population not connected to the sewer system nor to WWTPs ($NoCon_{rur}$), fraction of rural population connected to sewage farms ($TFarm_{rur}$), fraction of rural population connected to primary treatment ($T1_{rur}$), fraction of rural population connected to secondary and tertiary treatment without targeted denitrification ($T23^{noN}_{rur}$), fraction of rural population connected to tertiary treatment with targeted denitrification ($T3^{N}_{rur}$), fraction of rural population connected to secondary and tertiary treatment without targeted P removal ($T23^{noP}_{rur}$), and fraction of rural population connected to tertiary treatment with targeted P removal ($T3^{P}_{rur}$). We do not report the fraction of rural population connected to the sewer system but not to WWTP ($T0_{rur}$), since it is always equal to zero for Berlin. We consider that all wastewater that is collected in the sewer system but not treated in WWTPs is treated in sewage farms. Source of the data: the data shown in this figure were processed in this study, building on total population connection data from the Statistical Office of Germany and from the federal states of Berlin and Brandenburg (see Table S21 and Tables S37–S38). The connection data for rural population were derived using as explained in Sect. S7.2.1-S7.2.3.

S8 Observations of WWTP incoming and outgoing N and P emissions at NUTS-1 level

We compile observational data of public WWTP incoming and outgoing N and P load at NUTS-1 level from different reports of the statistical offices of Germany and the federal states, as well as situation reports of the authorities of the German federal states. Tables S45 and S60 provide an overview of the availability of the data of WWTP incoming and outgoing N and P emissions at NUTS-1 level, respectively, while details on data sources and availability are reported in Tables S46–S59 for incoming N and P load and in Tables S61–S76 for outgoing N and P load. Table S44 defines the notations used.

The data in the situation reports sometimes refer to the WWTPs having a design capacity higher than 2000 population equivalent (PE; one PE being defined as the organic biodegradable load having a five–day biochemical oxygen demand - BOD5 of 60 g of oxygen per day, EC, 1991) only. Regarding incoming N and P loads, they are either directly provided in the reports or we estimate them based on the provided efficiency of treatment and outgoing N and P loads.

Some reports provide not only the value of the loads but also the total volumes of wastewater treated in WWTPs as well as the volumes of wastewater treated in WWTPs for which load measurements are taken. This allows us to extrapolate the value of the load for the WWTPs that are not accounted for in the measurements and to derive a lower bound estimate (measured value) and an upper bound estimate (extrapolated value) for the observational data. For this we assume a constant ratio of load to wastewater volume for the WWTPs with and without measurement. Tables S61–S76 and Table S46 indicate when, in addition to the observations, we could derive extrapolated values. We note that, for the NUTS-1 region of Schleswig-Holstein, for some years, extrapolated values and not observed values are directly provided by the state authorities (details in Table S61).

While P data always correspond to total P, N load data are provided either as inorganic N, which includes ammonium (NH4-N), nitrate (NO3-N), and nitrite (NO2-N), or as total N, which includes inorganic N and organic N species. When observations of outgoing total N load is not provided, we estimate the outgoing total N load from observed outgoing inorganic N load data. For this, we derive a ratio of total N to inorganic N, denoted as $f_{wwtpOut}^{N:N^{inorg}}$ $(-)$, for the NUTS-1 regions for which both data are provided over a common time period, which regards eight (half) NUTS-1 regions. $f_{wwtpOut}^{N:N^{inorg}}$ is calculated as the ratio of the average value of the total N load to the average value of the inorganic N load over the common time period. The ratios can be visualised in Fig S27, while the periods over which these ratios were calculated are reported in Fig. S28. We select a lower bound value of 1.05 and an upper bound value of 1.25 for $f_{wwtpOut}^{N:N^{inorg}}$(as shown in Fig S27). This range excludes outliers values calculated for North Rhine-Westphalia (NW) and Baden-Württemberg (BW) that could be due to the fact that the values of the total N and inorganic N load are not provided for the exact same years during the common time period of availability. In particular, the ratio for North Rhine-Westphalia is slightly below 1, which is not a physical value. We derive a lower and upper bound estimate of the total N load by applying the lower and upper bound values of $f_{wwtpOut}^{N:N^{inorg}}$ to the value of the observed inorganic N load (when observations of total N are not provided).

The data of incoming and outgoing N and P load can be visualised in Fig. S28–S31, including (1) the observations that are derived directly from the reports, (2) the extrapolated values of inorganic N and total P load, and (3) the estimates of total N that we derived from observations of inorganic N using the coefficient $f_{wwtpOut}^{N:N^{inorg}}$.

Table S44. Notations for the WWTP incoming and outgoing N and P emissions variables used in Tables S45–S76 and in Figures S28–S31.

Variable	Description	Unit
$J^{N}_{wwtpIn,all}$	Incoming total N load to WWTPs (all PE)	$(kg\,yr^{-1})$
$J^{N_{capa\geq2000}}_{wwtpIn,all}$	Incoming total N load to WWTPs with a design capacity higher or equal to 2000 PE	$(kg\,yr{-1})$
$J^{N_{inorg}}_{wwtpIn,all}$	Incoming inorganic N load to WWTPs (all PE)	$(kg\,yr^{-1})$
$J^{N_{inorg,capa\geq2000}}_{wwtpIn,all}$	Incoming inorganic N load to WWTPs with a design capacity higher or equal to 2000 PE	$(kg\,yr{-1})$
$J^{P}_{wwtpIn,all}$	Incoming total P load to WWTPs (all PE)	$(kg\,yr{-1})$
$J^{P_{capa\geq2000}}_{wwtpIn,all}$	Incoming total P load to WWTPs with a design capacity higher or equal to 2000 PE	$(kg\,yr{-1})$
$J^{N}_{ps,wwtpOut,all}$	Outgoing total N load from WWTPs (all PE)	$(kg\,yr^{-1})$
$J^{N_{capa\geq2000}}_{ps,wwtpOut,all}$	Outgoing total N load from WWTPs with a design capacity higher or equal to 2000 PE	$(kg\,yr^{-1})$
$J^{N_{inorg}}_{ps,wwtpOut,all}$	Outgoing inorganic N load from WWTPs (all PE)	$(kg\,yr^{-1})$
$J^{N_{inorg,capa\geq2000}}_{ps,wwtpOut,all}$	Outgoing inorganic N load from WWTPs with a design capacity higher or equal to 2000 PE	$(kg\,yr{-1})$
$J^{P}_{ps,wwtpOut,all}$	Outgoing total P load to WWTPs (all PE)	$(kg\,yr^{-1})$
$J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$	Outgoing total P load from WWTPs with a design capacity higher or equal to 2000 PE	$(kg\,yr^{-1})$

S8.1 Availability and sources of the data of WWTP incoming and outgoing N and P emissions

S8.1.1 Availability and sources of the data of WWTP incoming N and P emissions

Table S45. First and last years for which observational data of incoming and outgoing N and P emissions variables are available for the different NUTS-1 regions.

NUTS-1 region	$J^{N}_{wwtpIn,all}$	$J^{N_{capa\geq 2000}}_{wwtpIn,all}$	$J^{N_{inorg}}_{wwtpIn,all}$	$J^{N_{inorg,capa\geq 2000}}_{wwtpIn,all}$	$J^{P}_{wwtpIn,all}$	$J^{P_{capa\geq 2000}}_{wwtpIn,all}$
SH	n.a.	n.a.	n.a.	n.a.	2007–2020	n.a.
HH[a]	2004–2020	2004–2020	2004–2020	2004–2020	2004–2020	2004–2020
LS	n.a.	n.a.	n.a.	2004–2019	n.a.	2004–2019
HB[a]	2000–2008	2000-2008	2012–2020	2012–2020	2000–2020	2000–2020
NW	2004–2020	2020	n.a.	n.a.	2004–2020	2020
HE	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
RP	2012–2020	2012–2020	n.a.	n.a.	n.a.	n.a.
BW	2010–2020	2010–2020	n.a.	n.a.	2007–2020	2010–2020
BV	2009–2019	n.a.	n.a.	n.a.	2010–2019	n.a.
SL	n.a.	2000-2020	n.a.	n.a.	n.a.	2000-2020
BE[a]	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
BB	n.a.	n.a.	n.a.	n.a.	1991–1995	n.a.
MV	2010	n.a.	n.a.	n.a.	2010	n.a.
SN	n.a.	n.a.	n.a.	n.a.	2007–2019	n.a.
ST	n.a.	2008–2020	n.a.	1998–2006	n.a.	1998–2020
TH	2008–2019	2002–2006	n.a.	n.a.	2008–2019	2002–2006

Notes: The variables are defined in Table S44. Details on data availability are reported in Tables S46–S76.
n.a.: not available.
[a] There are only one WWTP located in Hamburg and in Berlin, and four WWTPs located in Bremen and all have a capacity higher than 2000 PE.

Table S46. Source of the data of WWTP incoming N and P loads in **Schleswig-Holstein (SH)**

t	$J^{N}_{wwtpIn,all}$	$J^{N_{capa\geq2000}}_{wwtpIn,all}$	$J^{N_{inorg}}_{wwtpIn,all}$	$J^{N_{inorg,capa\geq2000}}_{wwtpIn,all}$	$J^{P}_{wwtpIn,all}{}^{a}$	$J^{P_{capa\geq2000}}_{wwtpIn,all}$
2007	n.a.	n.a.	n.a.	n.a.	Efficiency (MUNL-SH, 2009)	n.a.
					$J^{P}_{ps,wwtpOut,all}$ (Table S61)	n.a.
2010	n.a.	n.a.	n.a.	n.a.	Efficiency (MUNL-SH, 2011)	n.a.
					$J^{P}_{ps,wwtpOut,all}$ (Table S61)	n.a.
2013	n.a.	n.a.	n.a.	n.a.	Efficiency (MELUL-SH, 2013, 2015)	n.a.
					$J^{P}_{ps,wwtpOut,all}$ (Table S61)	n.a.
2016	n.a.	n.a.	n.a.	n.a.	Efficiency (MELUL-SH, 2017)	n.a.
					$J^{P}_{ps,wwtpOut,all}$ (Table S61)	n.a.
2019	n.a.	n.a.	n.a.	n.a.	Efficiency (MELUND-SH, 2019, 2021b)	n.a.
					$J^{P}_{ps,wwtpOut,all}$ (Table S61)	n.a.
2020	n.a.	n.a.	n.a.	n.a.	Efficiency (MELUND-SH, 2021b)	n.a.
					$J^{P}_{ps,wwtpOut,all}$ (Table S61)	n.a.

Notes: The variables are defined in Table S44.

n.a.: not available.

[a] $J^{P}_{wwtpIn,all}$ is not directly provided but we calculate it from data of the efficiency of P removal and of $J^{P}_{ps,wwtpOut,all}$ (outgoing load, Table S61). For each year we derived a lower and upper bound of $J^{P}_{wwtpIn,all}$ using the observed and extrapolated values of $J^{P}_{ps,wwtpOut,all}$, respectively (see Table S61). In 2007, we use the value of the efficiency provided for 2008. In 2013 and 2019 for use the values of the efficiency provided for the previous and next years (2012/2014 and 2018/2020, respectively). We use the lower (higher) value of the efficiency to estimate the lower (upper) bound of $J^{P}_{wwtpIn,all}$.

Table S47. Source of the data of WWTP incoming N and P loads in **Hamburg (HH)**

t	$J^{N}_{wwtpIn,all}$	$J^{N_{capa\geq 2000}}_{wwtpIn,all}$	$J^{N_{inorg}}_{wwtpIn,all}$	$J^{N_{inorg,capa\geq 2000}}_{wwtpIn,all}$	$J^{P}_{wwtpIn,all}$	$J^{P_{capa\geq 2000}}_{wwtpIn,all}$
2004	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)
2005	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)
2006	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)
2009	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)
2010	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)
2011	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)
2012	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)
2013	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]	BUE-HH (2017)	BUE-HH (2017)
2014	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)
2015	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)
2016	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)
2017	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)
2018	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)
2019	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)
2020	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)

Notes: The variables are defined in Table S44. n.a.: not available.

Only one WWTPs is located in Hamburg, which as a capacity larger than 2000 PE.

[a] Data of incoming N load are provided in BUE-HH (2017) but we did not use them, since we discarded the data for the outgoing load (see Table S62).

Table S48. Source of the data of WWTP incoming N and P loads in **Lower Saxony (LS)**

t	$J^{N}_{wwtpIn,all}$	$J^{N_{capa\geq 2000}}_{wwtpIn,all}$	$J^{N_{inorg}}_{wwtpIn,all}$	$J^{N_{inorg,capa\geq 2000}\,a}_{wwtpIn,all}$	$J^{P}_{wwtpIn,all}$	$J^{P_{capa\geq 2000}\,a}_{wwtpIn,all}$
2004	n.a.	n.a.	n.a.	NLWKM (2005)	n.a.	NLWKM (2005)
2006	n.a.	n.a.	n.a.	NLWKM (2007)	n.a.	NLWKM (2007)
2008	n.a.	n.a.	n.a.	NLWKM (2009)	n.a.	NLWKM (2009)
2009	n.a.	n.a.	n.a.	NLWKM (2011)	n.a.	NLWKM (2011)
2011	n.a.	n.a.	n.a.	NLWKM (2013)	n.a.	NLWKM (2013)
2013	n.a.	n.a.	n.a.	NLWKM (2015)	n.a.	NLWKM (2015)
2015	n.a.	n.a.	n.a.	NLWKM (2017)	n.a.	NLWKM (2017)
2017	n.a.	n.a.	n.a.	NLWKM (2019)	n.a.	NLWKM (2019)
2019	n.a.	n.a.	n.a.	NLWKM (2021)	n.a.	NLWKM (2021)

Notes: The variables are defined in Table S44.

n.a.: not available.

[a] We aggregate the data over Lower Saxony, as the data are provided at the catchment level.

Table S49. Source of the data of WWTP incoming N and P loads in **Bremen (HB)**

t	$J^{N}_{wwtpIn,all}$	$J^{N_{capa\geq2000}}_{wwtpIn,all}$	$J^{N_{inorg}}_{wwtpIn,all}$	$J^{N_{inorg,capa\geq2000}}_{wwtpIn,all}$	$J^{P}_{wwtpIn,all}$	$J^{P_{capa\geq2000}}_{wwtpIn,all}$
2000	Efficiency, $J^{N}_{ps,wwtpOut,all}$ SBU-HB (2001)	Efficiency, $J^{N_{capa\geq2000}}_{ps,wwtpOut,all}$ SBU-HB (2001)	n.a.	n.a.	Efficiency, $J^{P}_{ps,wwtpOut,all}$ SBU-HB (2001)	Efficiency, $J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$ SBU-HB (2001)
2002	Efficiency, $J^{N}_{ps,wwtpOut,all}$ SBUV-HB (2001)	Efficiency, $J^{N_{capa\geq2000}}_{ps,wwtpOut,all}$ SBUV-HB (2001)	n.a.	n.a.	Efficiency, $J^{P}_{ps,wwtpOut,all}$ SBUV-HB (2001)	Efficiency, $J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$ SBUV-HB (2001)
2004	Efficiency, $J^{N}_{ps,wwtpOut,all}$ SBUV-HB (2003)	Efficiency, $J^{N_{capa\geq2000}}_{ps,wwtpOut,all}$ SBUV-HB (2003)	n.a.	n.a.	Efficiency, $J^{P}_{ps,wwtpOut,all}$ SBUV-HB (2003)	Efficiency, $J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$ SBUV-HB (2003)
2006	Efficiency, $J^{N}_{ps,wwtpOut,all}$ SBUV-HB (2007)	Efficiency, $J^{N_{capa\geq2000}}_{ps,wwtpOut,all}$ SBUV-HB (2007)	n.a.	n.a.	Efficiency, $J^{P}_{ps,wwtpOut,all}$ SBUV-HB (2007)	Efficiency, $J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$ SBUV-HB (2007)
2008	Efficiency, $J^{N}_{ps,wwtpOut,all}$ SUBVE-HB (2009)	Efficiency, $J^{N_{capa\geq2000}}_{ps,wwtpOut,all}$ SUBVE-HB (2009)	n.a.	n.a.	Efficiency, $J^{P}_{ps,wwtpOut,all}$ SUBVE-HB (2009)	Efficiency, $J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$ SUBVE-HB (2009)
2010	n.a.		n.a.	n.a.	Efficiency, $J^{P}_{ps,wwtpOut,all}$ SUBVE-HB (2011)	Efficiency, $J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$ SUBVE-HB (2011)
2012	n.a.	n.a.	Efficiency, $J^{N_{inorg}}_{ps,wwtpOut,all}$ SUBV-HB (2013)	Efficiency, $J^{N_{inorg,capa\geq2000}}_{ps,wwtpOut,all}$ SUBV-HB (2013)	Efficiency, $J^{P}_{ps,wwtpOut,all}$ SUBV-HB (2013)	Efficiency, $J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$ SUBV-HB (2013)
2014	n.a.	n.a.	Efficiency, $J^{N_{inorg}}_{ps,wwtpOut,all}$ SUBV-HB (2015)	Efficiency, $J^{N_{inorg,capa\geq2000}}_{ps,wwtpOut,all}$ SUBV-HB (2015)	Efficiency, $J^{P}_{ps,wwtpOut,all}$ SUBV-HB (2015)	Efficiency, $J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$ SUBV-HB (2015)
2016	n.a.	n.a.	Efficiency, $J^{N_{inorg}}_{ps,wwtpOut,all}$ SUBV-HB (2017)	Efficiency, $J^{N_{inorg,capa\geq2000}}_{ps,wwtpOut,all}$ SUBV-HB (2017)	Efficiency, $J^{P}_{ps,wwtpOut,all}$ SUBV-HB (2017)	Efficiency, $J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$ SUBV-HB (2017)
2018	n.a.	n.a.	Efficiency, $J^{N_{inorg}}_{ps,wwtpOut,all}$ SUBV-HB (2019)	Efficiency, $J^{N_{inorg,capa\geq2000}}_{ps,wwtpOut,all}$ SUBV-HB (2019)	Efficiency, $J^{P}_{ps,wwtpOut,all}$ SUBV-HB (2019)	Efficiency, $J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$ SUBV-HB (2019)
2020	n.a.	n.a.	Efficiency, $J^{N_{inorg}}_{ps,wwtpOut,all}$ SKUMSW-HB (2021)	Efficiency, $J^{N_{inorg,capa\geq2000}}_{ps,wwtpOut,all}$ SKUMSW-HB (2021)	Efficiency, $J^{P}_{ps,wwtpOut,all}$ SKUMSW-HB (2021)	Efficiency, $J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$ SKUMSW-HB (2021)

Notes: The variables are defined in Table S44.

n.a.: not available.

Only four WWTPs are located in Bremen and they all have a capacity which is larger than 2000 PE The data are provided separately for each of the four WWTPs, which we aggregate to obtain the total for Bremen.The incoming loads to WWTPs are not directly provided but we calculated them from data of the efficiency of N and P removal and of the outgoing loads (Table S64). We derive only one estimate of the incoming load from the provided data.

Table S50. Source of the data of WWTP incoming N and P loads in **North Rhine Westfalia (NW)**

t	${J_{wwtpIn,all}^{N}}^{a}$	$J_{wwtpIn,all}^{N_{capa\geq2000}}$	$J_{wwtpIn,all}^{N_{inorg}}$	$J_{wwtpIn,all}^{N_{inorg,capa\geq2000}}$	${J_{wwtpIn,all}^{P}}^{a}$	$J_{wwtpIn,all}^{P_{capa\geq2000}}$
2004	Efficiency (MUNLV-NW, 2006) $J_{ps,wwtpOut,all}^{N}$ (Table S65)	n.a.	n.a.	n.a.	Efficiency (MUNLV-NW, 2006) $J_{ps,wwtpOut,all}^{P}$ (Table S65)	n.a
2005	Efficiency (MUNLV-NW, 2006) $J_{ps,wwtpOut,all}^{N}$ (Table S65)	n.a.	n.a.	n.a.	Efficiency (MUNLV-NW, 2006) $J_{ps,wwtpOut,all}^{P}$ (Table S65)	n.a
2006	Efficiency (MUNLV-NW, 2006) $J_{ps,wwtpOut,all}^{N}$ (Table S65)	n.a.	n.a.	n.a.	Efficiency (MUNLV-NW, 2006) $J_{ps,wwtpOut,all}^{P}$ (Table S65)	n.a
2010	Efficiency (MUNLV-NW, 2020) $J_{ps,wwtpOut,all}^{N}$ (Table S65)	n.a.	n.a.	n.a.	Efficiency (MUNLV-NW, 2020) $J_{ps,wwtpOut,all}^{P}$ (Table S65)	n.a
2012	Efficiency (MUNLV-NW, 2012) $J_{ps,wwtpOut,all}^{N}$ (Table S65)	n.a.	n.a.	n.a.	Efficiency (MUNLV-NW, 2012) $J_{ps,wwtpOut,all}^{P}$ (Table S65)	n.a
2014	Efficiency (MUNLV-NW, 2020) $J_{ps,wwtpOut,all}^{N}$ (Table S65)	n.a.	n.a.	n.a.	Efficiency (MUNLV-NW, 2020) $J_{ps,wwtpOut,all}^{P}$ (Table S65)	n.a
2016	Efficiency (MUNLV-NW, 2020) $J_{ps,wwtpOut,all}^{N}$ (Table S65)	n.a.	n.a.	n.a.	Efficiency (MUNLV-NW, 2020) $J_{ps,wwtpOut,all}^{P}$ (Table S65)	n.a
2018	Efficiency (MUNLV-NW, 2020) $J_{ps,wwtpOut,all}^{N}$ (Table S65)	n.a.	n.a.	n.a.	Efficiency (MUNLV-NW, 2020) $J_{ps,wwtpOut,all}^{P}$ (Table S65)	n.a
2020	MUNLV-NW (2020)	MUNLV-NW (2020)	n.a.	n.a.	MUNLV-NW (2020)	MUNLV-NW (2020)

Notes: The variables are defined in Table S44.

[a] Apart from year 2020, $J_{wwtpIn,all}^{N}$ and $J_{wwtpIn,all}^{P}$ are not directly provided but are calculated from data of the efficiency of N and P removal, respectively, and of $J_{ps,wwtpOut,all}^{N}$ and $J_{ps,wwtpOut,all}^{P}$ (outgoing loads, Table S65). For each year we derived a lower and upper bound of $J_{wwtpIn,all}^{P}$ using the observed and extrapolated values of $J_{ps,wwtpOut,all}^{P}$, respectively, when available.

Table S51. Source of the data of WWTP incoming N and P loads in **Rhineland-Palatinate (RP)**

t	$J^{N}_{wwtpIn,all}$	$J^{N_{capa\geq2000}}_{wwtpIn,all}$	$J^{N_{inorg}}_{wwtpIn,all}$	$J^{N_{inorg,capa\geq2000}}_{wwtpIn,all}$	$J^{P}_{wwtpIn,all}$	$J^{P_{capa\geq2000}}_{wwtpIn,all}$
2012	LUWG-RP and MULEWF-RP (2013)	LUWG-RP and MULEWF-RP (2013)	n.a.	n.a.	n.a.[a]	n.a.[a]
2016	LU-RP and MUEEF-RP (2017)	LU-RP and MUEEF-RP (2017)	n.a.	n.a.	n.a.[a]	n.a.[a]
2018	LU-RP and MUEEF-RP (2019)	LU-RP and MUEEF-RP (2019)	n.a.	n.a.	n.a.[a]	n.a.[a]
2020	LU-RP and MKUEM-RP (2021)	LU-RP and MUEEF-RP (2021)	n.a.	n.a.	n.a.[a]	n.a.[a]

Notes: The variables are defined in Table S44.

n.a.: not available

[a] LUWG-RP and MULEWF-RP (2013), LU-RP and MUEEF-RP (2017, 2019) and LU-RP and MUEEF-RP (2021) provide values of $J^{P}_{wwtpIn,all}$ and $J^{P_{capa\geq2000}}_{wwtpIn,all}$ in 2012, 2016, 2018, and 2020. However, we discard these values since we also discard the values of outgoing load (see Table S67

Table S52. Source of the data of WWTP incoming N and P loads in **Baden-Württemberg (BW)**

t	$J^{N}_{wwtpIn,all}$	$J^{N_{capa\geq 2000}}_{wwtpIn,all}$	$J^{N_{inorg}}_{wwtpIn,all}$	$J^{N_{inorg,capa\geq 2000}}_{wwtpIn,all}$	$J^{P}_{wwtpIn,all}$	$J^{P_{capa\geq 2000}}_{wwtpIn,all}$
2007	n.a.	n.a.	n.a.	n.a.	Efficiency (LUBW and MUKEBW, 2021) $J^{P}_{wwtpIn,all}$ (Table S68)[a]	n.a.
2010	LUMNBW (2011)	LUMNBW (2011)	n.a.	n.a.	LUMNBW (2011)	LUMNBW (2011)
2012	LUMNBW and MUKEBW (2013)	LUMNBW and MUKEBW (2013)	n.a.	n.a.	LUMNBW and MUKEBW (2013)	LUMNBW and MUKEBW (2013)
2013	n.a.	n.a.	n.a.	n.a.	Efficiency (LUBW and MUKEBW, 2021) $J^{P}_{wwtpIn,all}$ (Table S68)[a]	n.a.
2015	LUMNBW and MUKEBW (2017)	LUMNBW and MUKEBW (2017)	n.a.	n.a.	LUMNBW and MUKEBW (2017)	LUMNBW and MUKEBW (2017)
2016	n.a.	n.a.	n.a.	n.a.	Efficiency (LUBW and MUKEBW, 2021) $J^{P}_{wwtpIn,all}$ (Table S68)[a]	n.a.
2018	LUBW and MUKEBW (2019)	LUBW and MUKEBW (2019)	n.a.	n.a.	LUBW and MUKEBW (2019)	LUBW and MUKEBW (2019)
2019	n.a.	n.a.	n.a.	n.a.	Efficiency (LUBW and MUKEBW, 2021) $J^{P}_{wwtpIn,all}$ (Table S68)[a]	n.a.
2020	LUBW and MUKEBW (2021)	LUBW and MUKEBW (2021)	n.a.	n.a.	LUBW and MUKEBW (2021)	LUBW and MUKEBW (2021)

Notes: The variables are defined in Table S44

n.a.: not available.

[a] In 2007, 2013, 2016, and 2019, $J^{P}_{wwtpIn,all}$ is not directly provided but we calculate it from data of the efficiency of P removal and of $J^{P}_{ps,wwtpOut,all}$ (outgoing load, Table S68). We derive an lower and upper bound of $J^{P}_{wwtpIn,all}$ using the observed and extrapolated values of $J^{P}_{ps,wwtpOut,all}$, respectively.

Table S53. Source of the data of WWTP incoming N and P loads in **Bavaria (BV)**

t	$J^{N}_{wwtpIn,all}$	$J^{N_{capa\geq2000}}_{wwtpIn,all}$	$J^{N_{inorg}}_{wwtpIn,all}$	$J^{N_{inorg,capa\geq2000}}_{wwtpIn,all}$	$J^{P}_{wwtpIn,all}$ [a]	$J^{P_{capa\geq2000}}_{wwtpIn,all}$
2009	Efficiency, $J^{N}_{wwtpIn,all}$ (LfU, 2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2010	Efficiency, $J^{N}_{wwtpIn,all}$ (LfU, 2021)	n.a.	n.a.	n.a.	Efficiency (LfU, 2021), $J^{P}_{wwtpIn,all}$ (Table S69)	n.a.
2011	Efficiency, $J^{N}_{wwtpIn,all}$ (LfU, 2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2012	Efficiency, $J^{N}_{wwtpIn,all}$ (LfU, 2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2013	Efficiency, $J^{N}_{wwtpIn,all}$ (LfU, 2021)	n.a.	n.a.	n.a.	Efficiency (LfU, 2021), $J^{P}_{wwtpIn,all}$ (Table S69)	n.a.
2014	Efficiency, $J^{N}_{wwtpIn,all}$ (LfU, 2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2015	Efficiency, $J^{N}_{wwtpIn,all}$ (LfU, 2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2016	Efficiency, $J^{N}_{wwtpIn,all}$ (LfU, 2021)	n.a.	n.a.	n.a.	Efficiency (LfU, 2021), $J^{P}_{wwtpIn,all}$ (Table S69)	n.a.
2017	Efficiency, $J^{N}_{wwtpIn,all}$ (LfU, 2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2018	Efficiency, $J^{N}_{wwtpIn,all}$ (LfU, 2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2019	Efficiency, $J^{N}_{wwtpIn,all}$ (LfU, 2021)	n.a.	n.a.	n.a.	Efficiency (LfU, 2021), $J^{P}_{wwtpIn,all}$ (Table S69)	n.a.

Notes: The variables are defined in Table S44.

n.a.: not available.

The incoming loads to WWTPs are not directly provided but we calculated them from data of the efficiency of N and P removal and of the outgoing loads.

[a] $J^{P}_{wwtpIn,all}$ is not directly provided but we calculate it from data of the efficiency of P removal and of $J^{P}_{ps,wwtpOut,all}$ (outgoing load, Table S69). We derive a lower and upper bound of $J^{P}_{wwtpIn,all}$ using the observed and extrapolated values of $J^{P}_{ps,wwtpOut,all}$.

Table S54. Source of the data of WWTP incoming N and P loads in **Saarland (SL)**

t	$J^{N}_{wwtpIn,all}$	$J^{N_{capa\geq 2000}}_{wwtpIn,all}$	$J^{N_{inorg}}_{wwtpIn,all}$	$J^{N_{inorg,capa\geq 2000}}_{wwtpIn,all}$	$J^{P}_{wwtpIn,all}$	$J^{P_{capa\geq 2000}}_{wwtpIn,all}$
2000	n.a.	MU-SL (2000)	n.a.	n.a.	n.a.	MU-SL (2000)
2002	n.a.	MU-SL (2002)	n.a.	n.a.	n.a.	MU-SL (2002)
2008	n.a.	MU-SL (2008)	n.a.	n.a.	n.a.	MU-SL (2008)
2012	n.a.	MUV-SL (2013)	n.a.	n.a.	n.a.	MUV-SL (2013)
2014	n.a.	MUV-SL (2014)	n.a.	n.a.	n.a.	MUV-SL (2014)
2016	n.a.	MUV-SL and LUA-SL (2017)	n.a.	n.a.	n.a.	MUV-SL and LUA-SL (2017)
2018	n.a.	MUV-SL and LUA-SL (2018)	n.a.	n.a.	n.a.	MUV-SL and LUA-SL (2018)
2020	n.a.	MUV-SL and LUA-SL (2021)	n.a.	n.a.	n.a.	MUV-SL and LUA-SL (2021)

Notes: The variables are defined in Table S44

n.a.: not available.

Table S55. Source of the data of WWTP incoming N and P loads in **Brandenburg (BB)**

t	$J^{N}_{wwtpIn,all}$	$J^{N_{capa\geq2000}}_{wwtpIn,all}$	$J^{N_{inorg}}_{wwtpIn,all}$	$J^{N_{inorg,capa\geq2000}}_{wwtpIn,all}$	$J^{P}_{wwtpIn,all}$	$J^{P_{capa\geq2000}}_{wwtpIn,all}$
1991	n.a.	n.a.	n.s.	n.a.	obs., extrapol. SO-BB (1995)	n.a
1995	n.a.	n.a.	n.a.	n.a.	obs., extrapol. SO-BB (1998)	n.a

Notes: The variables are defined in Table S44.

SO-BB: Statistical Office of Brandenburg; SO-BE&BB: Statistical Office of Berlin and Brandenburg (https://www.statistischebibliothek.de/mir/receive/BBSerie_mods_00000562).

n.a.: not available.

Table S56. Source of the data of WWTP incoming N and P loads in **Mecklenburg-Vorpommern (MV)**

t	$J^{N}_{wwtpIn,all}$	$J^{N_{capa\geq2000}}_{wwtpIn,all}$	$J^{N_{inorg}}_{wwtpIn,all}$	$J^{N_{inorg,capa\geq2000}}_{wwtpIn,all}$	$J^{P}_{wwtpIn,all}$	$J^{P_{capa\geq2000}}_{wwtpIn,all}$
2010	LUNG-MV (2011)	n.a.	n.a.	n.a.	LUNG-MV (2011)	n.a.

Notes: The variables are defined in Table S44.

n.a.: not available.

Table S57. Source of the data of WWTP incoming N and P loads in **Saxony (SN)**

t	J^{N}_{wwtpIn}	$J^{N_{capa \geq 2000}}_{wwtpIn,all}$	$J^{N_{inorg}}_{wwtpIn,all}$	$J^{N_{inorg,capa \geq 2000}}_{wwtpIn,all}$	$J^{P}_{wwtpIn,all}$	$J^{P_{capa \geq 2000}}_{wwtpIn,all}$
2007	n.a.	n.a.	n.a.	n.a.	Efficiency (SMUL and LfULG, 2008)	n.a.
					$J^{P}_{ps,wwtpOut,all}$ (Table S74)	
2010	n.a.	n.a.	n.a.	n.a.	Efficiency (SMUL and LfULG, 2011, 2013)	n.a.
					$J^{P}_{ps,wwtpOut,all}$ (Table S74)	
2013	n.a.	n.a.	n.a.	n.a.	Efficiency (SMUL and LfULG, 2015)	n.a.
					$J^{P}_{ps,wwtpOut,all}$ (Table S74)	
2016	n.a.	n.a.	n.a.	n.a.	Efficiency (SMUL and LfULG, 2017, 2019)	n.a.
					$J^{P}_{ps,wwtpOut,all}$ (Table S74)	
2019	n.a.	n.a.	n.a.	n.a.	Efficiency (SMUL and LfULG, 2021)	n.a.
					$J^{P}_{ps,wwtpOut,all}$ (Table S74)	

Notes: The variables are defined in Table S44.

n.a.: not available.

$J^{P}_{wwtpIn,all}$ is not directly provided but we calculate it from data of the efficiency of P removal and of $J^{P}_{ps,wwtpOut,all}$ (outgoing load, Table S74). We derive a lower and upper bound of $J^{P}_{wwtpIn,all}$ using the observed and extrapolated values of $J^{P}_{ps,wwtpOut,all}$, respectively. In 2010 and 2016, we use the values of the efficiency provided for the previous and next years (2009/2011 and 2015/2017, respectively). We use the lower (higher) value of the efficiency to estimate the lower (upper) bound of $J^{P}_{wwtpIn,all}$.

Table S58. Source of the data of WWTP incoming N and P loads in **Saxony-Anhalt (ST)**

t	$J^{N}_{wwtpIn,all}$	$J^{N_{capa \geq 2000}}_{wwtpIn,all}$	$J^{N_{inorg}}_{wwtpIn,all}$	$J^{N_{inorg,capa \geq 2000}}_{wwtpIn,all}$	$J^{P}_{wwtpIn,all}$	$J^{P_{capa \geq 2000}}_{wwtpIn,all}$
1998	n.a.	n.a.	n.a.	MRLU-ST (1999)	n.a.	MRLU-ST (1999)
2000	n.a.	n.a.	n.a.	MRLU-ST (2001)	n.a.	MRLU-ST (2001)
2002	n.a.	n.a.	n.a.	MLU-ST (2003)	n.a.	MLU-ST (2003)
2004	n.a.	n.a.	n.a.	MLU-ST (2005)	n.a.	MLU-ST (2005)
2006	n.a.	n.a.	n.a.	MLU-ST (2007)	n.a.	MLU-ST (2007)
2008	n.a.	MLU-ST (2009)	n.a.	n.a.	n.a.	MLU-ST (2009)
2010	n.a.	MLU-ST (2011)	n.a.	n.a.	n.a.	MLU-ST (2011)
2012	n.a.	MLU-ST (2013)	n.a.	n.a.	n.a.	MLU-ST (2013)
2014	n.a.	MLU-ST (2015)	n.a.	n.a.	n.a.	MLU-ST (2015)
2016	n.a.	MLU-ST (2017)	n.a.	n.a.	n.a.	MLU-ST (2017)
2020	n.a.	LU-ST (2021)	n.a.	n.a.	n.a.	LU-ST (2021)

Notes: The variables are defined in Table S44.

n.a.: not available.

Table S59. Source of the data of WWTP incoming N and P loads in **Thuringia (TH)**

t	$J^{N}_{wwtpIn,all}$	$J^{N_{capa\geq2000}}_{wwtpIn,all}$	$J^{N_{inorg}}_{wwtpIn,all}$	$J^{N_{inorg,capa\geq2000}}_{wwtpIn,all}$	$J^{P}_{wwtpIn,all}$	$J^{P_{capa\geq2000}}_{wwtpIn,all}$
2002	n.a.	TMLNU and TLUG (2003)	n.a.	n.a.	n.a.	TMLNU and TLUG (2003)
2004	n.a.	TMLNU and TLUG (2005)	n.a.	n.a.	n.a.	TMLNU and TLUG (2005)
2006	n.a.	TMLNU (2007)	n.a.	n.a.	n.a.	TMLNU (2007)
2008	TMLNU (2009)	n.a.	n.a.	n.a.	TMLNU (2009)	n.a.
2009	TMLFUN (2011)	n.a.	n.a.	n.a.	TMLFUN (2011)	n.a.
2011	TMLFUN (2013)	n.a.	n.a.	n.a.	TMLFUN (2013)	n.a.
2013	TMUEN (2015)	n.a.	n.a.	n.a.	TMUEN (2015)	n.a.
2015	TMUEN (2017)	n.a.	n.a.	n.a.	TMUEN (2017)	n.a.
2017	TMUEN (2019)	n.a.	n.a.	n.a.	TMUEN (2019)	n.a.
2019	TMUEN (2021)	n.a.	n.a.	n.a.	TMUEN (2021)	n.a.

Notes: The variables are defined in Table S44.

n.a.: not available.

S8.1.2 Availability and sources of the data of WWTP outgoing N and P emissions

Table S60. First and last years for which data of population connection are available for the different NUTS-1 regions.

NUTS-1 region	$J^{N}_{ps,wwtpOut,all}$	$J^{N_{capa\geq2000}}_{ps,wwtpOut,all}$	$J^{N_{inorg}}_{ps,wwtpOut,all}$	$J^{N_{inorg,capa\geq2000}}_{ps,wwtpOut,all}$	$J^{P}_{ps,wwtpOut,all}$	$J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$
SH	n.a.	n.a.	1991–2019	n.a.	1987–2020	n.a.
HH[a]	2004–2020	2004–2020	2004–2020	2004–2020	2004–2019	2004–2019
LS	n.a.	n.a.	2001–2019	2004–2019	2001–2019	2004–2019
HB[a]	1998–2010	1998–2010	2007–2020	2007–2020	1998–2020	1998–2020
NW	1991–2020	n.a.	2007–2019	n.a.	1991–2020	n.a.
HE	n.a.	n.a.	1996–2020	n.a.	1996–2020	
RP	2012–2020	2012–2020	2001–2019	n.a.	2001–2019	n.a.
BW	2010–2020	2010–2020	2007–2019	n.a.	2007–2020	2010–2020
BV	2000–2019	n.a.	2007–2019	n.a.	2007–2019	n.a.
SL	n.a.	2000–2020	2007–2019	n.a.	2007–2019	2000–2020
BE[a]	n.a.	n.a.	2007–2019	2007–2019	2007–2019	2007–2019
BB	n.a	n.a	1991–2019	n.a.	1991–2019	n.a.
MV	2010	n.a.	2007–2019	n.a.	2007–2019	n.a.
SN	n.a.	n.a.	2007–2019	n.a.	2007–2019	n.a.
ST	n.a.	2008–2020	2007–2019	1998–2006	2007–2019	1998–2020
TH	2008–2019	2002–2006	2007–2019	n.a.	2007–2019	2002–2006

Notes: The variables are defined in Table S44.

Details on data availability are reported in Tables S46–S76.

n.a.: not available.

[a] There are only one WWTP located in Hamburg and in Berlin, and four WWTPs located in Bremen. Since the load values include all these WWTPs, the values of the extrapolated load are the same as the values of the observed load.

Table S61. Source of the data of WWTP outgoing N and P loads in **Schleswig-Holstein (SH)**

t	$J^{N}_{ps,wwtpOut,all}$	$J^{N_{capa\geq2000}}_{ps,wwtpOut,all}$	$J^{N_{inorg}}_{ps,wwtpOut,all}$	$J^{N_{inorg,capa\geq2000}}_{ps,wwtpOut,all}$	$J^{P}_{ps,wwtpOut,all}$	$J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$
1987	n.a.	n.a.	n.a	n.a.	unspecified	n.a.
					MELUND-SH (2021a, Fig. 2.3)	
1991	n.a.	n.a	extrapol.[b]	n.a.	extrapol.[b]	n.a.
			MUNL SH (2005, Fig. 4)		MUNL-SH (2005, Fig. 4)	
1995	n.a.	n.a	extrapol.[b]	n.a.	extrapol.[b]	n.a.
			MUNL SH (2005, Fig. 4)		MUNL-SH (2005, Fig. 4)	
1998	n.a.	n.a	obs,extrapol.[a,b,c]	n.a.	obs,extrapol.[a,b]	n.a.
			SO-SH (2001), MUNL-SH (2005, Fig. 4)		SO-SH (2001), MUNL SH (2005, Fig. 4)	
2001	n.a.	n.a	obs, extrapol.[a,b]	n.a.	obs, extrapol.[a,b]	n.a.
			SO-SH (2003), MUNL-SH (2005, Fig. 4)		SO-SH (2003), MUNL SH (2005, Fig. 4)	
2004	n.a.	n.a	obs, extrapol.[a]	n.a.	obs, extrapol.[a]	n.a.
			SO-HH&SH (2006b)		SO-HH&SH (2006b)	
2007	n.a.	n.a	obs, extrapol.[a]	n.a.	obs, extrapol.[a]	n.a.
			SO-DE (2009), SO-HH&SH (2009b)		SO-DE (2009), SO-HH&SH (2009b)	
2010	n.a.	n.a	obs, extrapol.[a]	n.a.	obs, extrapol.[a]	n.a.
			SO-DE (2013), SO-HH&SH (2012b)		SO-DE (2013), SO-HH&SH (2012b)	
2013	n.a.	n.a	obs, extrapol.	n.a.	obs, extrapol.	n.a.
			SO-DE (2015), SO-HH&SH (2015b)		SO-DE (2015), SO-HH&SH (2015b)	
2016	n.a.	n.a	obs, extrapol.	n.a.	obs, extrapol.	n.a.
			SO-DE (2018), SO-HH&SH (2018b)		SO-DE (2018), SO-HH&SH (2018b)	
2019	n.a.	n.a	obs, extrapol.	n.a.	obs, extrapol.	n.a.
			SO-DE (2022), SO-HH&SH (2021b)		SO-DE (2022), SO-HH&SH (2021b)	
2020	n.a.	n.a.	n.a	n.a.	unspecified	n.a.
					MELUND-SH (2021a, 2021b)	

Notes: The variables are defined in Table S44

SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-SH: Statistical Office of Schleswig-Holstein; SO-HH&SH: Statistical Office of Hamburg and Schleswig-Holstein (data link: https://www.statistischebibliothek.de/mir/receive/SHSerie_mods_00000083).

n.a.: not available.

[a] Observed and extrapolated values are both provided in SO-SH (2001, 2003, 2006b, 2009, 2013).

[b] We assume that the values provided in MUNL-SH (2005) always correspond to extrapolated values, since for the years 1998 and 2001 they are consistent with the extrapolated values provided in SO-SH (2001) and SO-SH (2003).

[c] In SO-SH (2001), the observed value of $J^{N_{inorg}}_{ps,wwtpOut,all}$ is higher than the extrpolated value. We discard the observed value and we calculate it based on the volumes of wastewater treated provided.

Table S62. Source of the data of WWTP outgoing N and P loads in **Hamburg (HH)**

t	$J^{N}_{ps,wwtpOut,all}$	$J^{N_{capa\geq2000}}_{ps,wwtpOut,all}$	$J^{N_{inorg}}_{ps,wwtpOut,all}$	$J^{N_{inorg,capa\geq2000}}_{ps,wwtpOut,all}$	$J^{P}_{ps,wwtpOut,all}$	$J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$
2004	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)
2005	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)
2006	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)	BSU-HH (2006)
2007	n.a.	n.a.	SO-DE (2009)	SO-DE (2009)	SO-DE (2009)	SO-DE (2009)
2009	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	n.a.[b]	n.a.[b]
2010	BSU-HH (2013)	BSU-HH (2013)	SO-DE (2013)[a]	SO-DE (2013)[a]	SO-DE (2013)[b]	SO-DE (2013)[b]
2011	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	n.a.[b]	n.a.[b]
2012	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	BSU-HH (2013)	n.a.[b]	n.a.[b]
2013	n.a.[a]	n.a.[a]	SO-DE (2015)	SO-DE (2015)	SO-DE (2015)	SO-DE (2015)
			SO-HH&SH (2015a)[a]	SO-HH&SH (2015a)[a]	SO-HH&SH (2015a)[b]	SO-HH&SH (2015a)[b]
2014	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)	n.a.[b]	n.a.[b]
2015	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)	n.a.[b]	n.a.[b]
2016	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)	BUE-HH (2017)	SO-DE (2018)	SO-DE (2018)
			SO-DE (2018)	SO-DE (2018)	SO-HH&SH (2018a)[b]	SO-HH&SH (2018a)[b]
			SO-HH&SH (2018a)			
2017	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	n.a.[b]	n.a.[b]
2018	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	n.a.[b]	n.a.[b]
2019	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	SO-DE (2022)	SO-DE (2022)
			SO-DE (2022)	SO-DE (2022)	SO-HH&SH (2021a)[b]	SO-HH&SH (2021a)[b]
			SO-HH&SH (2021a)	SO-HH&SH (2021a)		
2020	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	BUKEA-HH (2021)	n.a.[b]	n.a.[b]

Notes: The variables are defined in Table S44

SO-DE: Statistical office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-HH&SH: statistical office of Hamburg-Schleswig-Holstein (https://www.statistischebibliothek.de/mir/receive/HHSerie_mods_00000126).

n.a.: not available.

There is only one WWTP located in Hamburg with a capacity higher than 2000 PE. This implies that the values of the extrapolated load are the same as the values of the observed load.

[a] Data of $J^{N_{inorg}}_{ps,wwtpOut,all}$ are provided in BUE-HH (2013,2017) for 2010 and 2013 and data of $J^{N}_{ps,wwtpOut,all}$ in BUE-HH (2017) for 2013, but we did not use them since they are inconsistent with the values provided in SO-DE (2013, 2015) and SO-HH&SH (2015).

[b] Values of $J^{P}_{ps,wwtpOut,all}$ are provided in BSU-HH (2013), BUE-HH (2017), and BUKEA-HH (2021) for the period 2009–2020 but we did not use these values, as they lack precision.

Table S63. Source of the data of WWTP outgoing N and P loads in **Lower Saxony (LS)**

t	$J^{N}_{ps,wwtpOut,all}$	$J^{N_{capa\geq 2000}}_{ps,wwtpOut,all}$	$J^{N_{inorg}}_{ps,wwtpOut,all}$	$J^{N_{inorg,capa\geq 2000}}_{ps,wwtpOut,all}$ [a]	$J^{P}_{ps,wwtpOut,all}$	$J^{P_{capa\geq 2000}}_{ps,wwtpOut,all}$ [a]
2001	n.a.	n.a.	obs., extrapol. SO-LS (2003)	n.a.	obs., extrapol. SO-LS (2003)	n.a.
2004	n.a.	n.a.	obs., extrapol. SO-LS (2009)	unspecified NLWKM (2005)	obs., extrapol. SO-LS (2009)	unspecified NLWKM (2005)
2006	n.a.	n.a.	n.a.	unspecified NLWKM (2007)	n.a.	unspecified NLWKM (2007)
2007	n.a.	n.a.	obs., extrapol. SO-DE (2009), SO-LS (2010)	n.a.	obs., extrapol. SO-DE (2009), SO-LS (2010)	n.a.
2008	n.a.	n.a.	n.a.	unspecified NLWKM (2009)	n.a.	unspecified NLWKM (2009)
2009	n.a.	n.a.	n.a.	unspecified NLWKM (2011)	n.a.	unspecified NLWKM (2011)
2010	n.a.	n.a.	obs., extrapol. SO-DE (2013), SO-LS (2014)	n.a.	obs., extrapol. SO-DE (2013), SO-LS (2014)	n.a.
2011	n.a.	n.a.	n.a.	unspecified NLWKM (2013)	n.a.	unspecified NLWKM (2013)
2013	n.a.	n.a.	obs., extrapol. SO-DE (2015), SO-LS (2019a)	unspecified NLWKM (2015)	obs., extrapol. SO-DE (2015), SO-LS (2019a)	unspecified NLWKM (2015)
2015	n.a.	n.a.	n.a.	unspecified NLWKM (2017)	n.a.	unspecified NLWKM (2017)
2016	n.a.	n.a.	obs., extrapol. SO-DE (2018), SO-LS (2019b)	n.a.	obs., extrapol. SO-DE (2018), SO-LS (2019b)	n.a.
2017	n.a.	n.a.	n.a.	unspecified NLWKM (2019)	n.a.	unspecified NLWKM (2019)
2019	n.a.	n.a.	obs., extrapol. SO-DE (2022), SO-LS (2022)	unspecified NLWKM (2021)	obs., extrapol. SO-DE (2022), SO-LS (2022)	unspecified NLWKM (2021)

Notes: The variables are defined in Table S44.
SO-DE: Statistical Office of Germany
(https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694;
https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B).
SO-LS: Statistical Office of Lower Saxony (https://www.statistischebibliothek.de/mir/receive/NISerie_mods_00000189).
n.a.: not available
unspecified: it is not specified whether the data correspond to observed and extrapolated values.
[a] We aggregate the data of $J^{P_{capa\geq 2000}}_{ps,wwtpOut,all}$ over Lower Saxony, as the data are provided at the catchment level.

Table S64. Source of the data of WWTP outgoing N and P loads in **Bremen (HB)**

t	$J^{N}_{ps,wwtpOut,all}$	$J^{N_{capa\geq2000}}_{ps,wwtpOut,all}$	$J^{N_{inorg}}_{ps,wwtpOut,all}$	$J^{N_{inorg,capa\geq2000}}_{ps,wwtpOut,all}$	$J^{P}_{ps,wwtpOut,all}$	$J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$
1998	SFGJSU-HB (1999)	SFGJSU-HB (1999)	n.a.	n.a.	SFGJSU-HB (1999)	SFGJSU-HB (1999)
2000	SBU-HB (2001)	SBU-HB (2001)	n.a.	n.a.	SBU-HB (2001)	SBU-HB (2001)
2002	SBUV-HB (2001)	SBUV-HB (2001)	n.a.	n.a.	SBUV-HB (2001)	SBUV-HB (2001)
2004	SBUV-HB (2003)	SBUV-HB (2003)	n.a.	n.a.	SBUV-HB (2003)	SBUV-HB (2003)
2006	SBUV-HB (2007)	SBUV-HB (2007)	n.a.	n.a.	SBUV-HB (2007)	SBUV-HB (2007)
2007	n.a.	n.a.	SO-DE (2009)	SO-DE (2009)	SO-DE (2009)	SO-DE (2009)
2008	SUBVE-HB (2009)	SUBVE-HB (2009)	n.a.	n.a.	SUBVE-HB (2009)	SUBVE-HB (2009)
2010	SUBVE-HB (2011)	SUBVE-HB (2011)	SO-DE (2013)	SO-DE (2013)	SO-DE (2013), SUBVE-HB (2011)	SO-DE (2013), SUBVE-HB (2011)
2012	n.a.	n.a.	SUBV-HB (2013)	SUBV-HB (2013)	SUBV-HB (2013)	SUBV-HB (2013)
2013	n.a.	n.a.	SO-DE (2015)	SO-DE (2015)	SO-DE (2015)	SO-DE (2015)
2014	n.a.	n.a.	SUBV-HB (2015)	SUBV-HB (2015)	SUBV-HB (2015)	SUBV-HB (2015)
2016	n.a.	n.a.	SO-DE (2018)[a]	SO-DE (2018)[a]	SO-DE (2018), SUBV-HB (2017)	SO-DE (2018), SUBV-HB (2017)
2018	n.a.	n.a.	SUBV-HB (2019)	SUBV-HB (2019)	SUBV-HB (2019)	SUBV-HB (2019)
2019	n.a.	n.a.	SO-DE (2020)	SO-DE (2020)	SO-DE (2020)	SO-DE (2020)
2020	n.a.	n.a.	SKUMSW-HB (2021)	SKUMSW-HB (2021)	SKUMSW-HB (2021)	SKUMSW-HB (2021)

Notes: The variables are defined in Table S44

SO-DE: Statistical office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B).

n.a.: not available.

Only four WWTPs are located in Bremen and they all have a capacity which is larger than 2000 PE. We only derive one estimate of the load (the observed and extrapolated values are equal).

[a] The values can be derived from both SO-DE (2018) (511 t yr^{-1}) and SUBV-HB (2017) (534 t yr^{-1}). The small differences may be due to roundoff errors and we adopt the value from SO-DE (2019).

Table S65. Source of the data of WWTP outgoing N and P loads in **North Rhine Westfalia (NW)**

t	$J^{N}_{ps,wwtpOut,all}$	$J^{N_{capa\geq2000}}_{ps,wwtpOut,all}$	$J^{N_{inorg}}_{ps,wwtpOut,all}$	$J^{N_{inorg,capa\geq2000}}_{ps,wwtpOut,all}$	$J^{P}_{ps,wwtpOut,all}$	$J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$
1991	unspecified	n.a.	n.a.	n.a.	unspecified	n.a
	MUNLV-NW (2012)				MUNLV-NW (2012)	
2004	unspecified	n.a.	n.a.	n.a.	unspecified	n.a
	MUNLV-NW (2006)				MUNLV-NW (2006)	
2005	unspecified	n.a.	n.a.	n.a.	unspecified	n.a
	MUNLV-NW (2006)				MUNLV-NW (2006)	
2006	unspecified	n.a.	n.a.	n.a.	unspecified	n.a
	MUNLV-NW (2006)				MUNLV-NW (2006)	
2007	n.a.	n.a.	obs.,extrapol.	n.a.	obs.,extrapol.	n.a
			SO-DE (2009)		SO-DE (2009)	
2008	unspecified	n.a.	n.a.	n.a.	unspecified	n.a
	MUNLV-NW (2012)				MUNLV-NW (2012)	
2010	unspecified	n.a.	obs.,extrapol.	n.a.	obs.,extrapol.	n.a
	MUNLV-NW (2012)		SO-DE (2013)		SO-DE (2013)[a]	
2012	unspecified	n.a.	n.a.	n.a.	unspecified	n.a
	MUNLV-NW (2012)				MUNLV-NW (2012)	
2013	n.a.	n.a.	obs.,extrapol.	n.a.	obs.,extrapol.	n.a
			SO-DE (2015)		SO-DE (2015)	
2014	unspecified	n.a.	n.a.	n.a.	unspecified	n.a
	MUNLV-NW (2020)				MUNLV-NW (2020)	
2016	unspecified	n.a.	obs.,extrapol.	n.a.	obs.,extrapol.	n.a
	MUNLV-NW (2020)		SO-DE (2019)		SO-DE (2019)[b]	
2018	unspecified	n.a.	n.a.	n.a.	unspecified	n.a
	MUNLV-NW (2020)				MUNLV-NW (2020)	
2019	n.a.	n.a.	obs.,extrapol.	n.a.	obs.,extrapol.	n.a
			SO-DE (2022)		SO-DE (2022)	
2020	unspecified	n.a.	n.a.	n.a.	unspecified	n.a
	MUNLV-NW (2020)				MUNLV-NW (2020)	

Notes: The variables are defined in Table S44

SO-DE: Statistical office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B).

n.a.: not available.

unspecified: it is not specified whether the data correspond to observed and extrapolated values.

[a] MUNLV-NW (2012) also provide a value for $J^{P}_{ps,wwtpOut,all}$ in 2010 equal to 1308 $t\ yr^{-1}$ (it is not specified whether the value is observed or extrapolated), but we adopt the values provided in SO-DE (2013) that are slightly different (observed value: 1325 $t\ yr^{-1}$; extrapolated value: 1352.4 $t\ yr^{-1}$).

[b] MUNLV-NW (2020) also provide a value for $J^{P}_{ps,wwtpOut,all}$ in 2010 equal to 1000 $t\ yr^{-1}$ (it is not specified whether the value is observed or extrapolated), but we adopt the values provided in SO-DE (2019) that are slightly different (observed value: 1035.4 tyr^{-1} ; extrapolated value: 1035.6 tyr^{-1}).

Table S66. Source of the data of WWTP outgoing N and P loads in **Hesse (HE)**

t	$J^{N}_{ps,wwtpOut,all}$	$J^{N capa\geq2000}_{ps,wwtpOut,all}$	$J^{N inorg}_{ps,wwtpOut,all}$	$J^{N inorg,capa\geq2000}_{ps,wwtpOut,all}$	$J^{P}_{ps,wwtpOut,all}$	$J^{P capa\geq2000}_{ps,wwtpOut,all}$
1996	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
1997	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
1998	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
1999	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
2000	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
2001	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
2002	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
2003	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
2004	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
2005	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
2006	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
2007	n.a.	n.a.	obs.,extrapol. SO-DE (2009)[a]	n.a.	obs.,extrapol. SO-DE (2009)[a]	n.a.
2008	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
2009	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
2010	n.a.	n.a.	obs.,extrapol. SO-DE (2013)[a]	n.a.	obs.,extrapol. SO-DE (2013)[a]	n.a.
2011	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
2012	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
2013	n.a.	n.a.	obs.,extrapol. SO-DE (2015)[a]	n.a.	obs.,extrapol. SO-DE (2015)[a]	n.a.
2014	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
2015	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
2016	n.a.	n.a.	obs.,extrapol. SO-DE (2018)[a]	n.a.	obs.,extrapol. SO-DE (2018)[a]	n.a.
2017	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
2018	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.
2019	n.a.	n.a.	obs.,extrapol. SO-DE (2022)[a]	n.a.	obs.,extrapol. SO-DE (2022)[a]	n.a.
2020	n.a.	n.a.	unspecified LNUG-HE (2021)	n.a.	unspecified LNUG-HE (2021)	n.a.

Notes: The variables are defined in Table S44

SO-DE: Statistical office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B).

n.a.: not available.

unspecified: it is not specified whether the data correspond to observed and extrapolated values.

[a] The values of the outgoing loads are also provided in LNUG-HE (2021), but we adopt the values provided in SO-DE (2009, 2013, 2015, 2018, 2022) because they have a higher precision.

Table S67. Source of the data of WWTP outgoing N and P loads in **Rhineland-Palatinate (RP)**

t	$J^{N}_{ps,wwtpOut,all}$	$J^{N_{capa\geq 2000}}_{ps,wwtpOut,all}$	$J^{N_{inorg}}_{ps,wwtpOut,all}$	$J^{N_{inorg,capa\geq 2000}}_{ps,wwtpOut,all}$	$J^{P}_{ps,wwtpOut,all}$	$J^{P_{capa\geq 2000}}_{ps,wwtpOut,all}$
2001	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol.	n.a.
			SO-RP (2003)		SO-RP (2003)	
2004	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol.	n.a.
			SO-RP (2006)		SO-RP (2006)	
2007	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol.	n.a.
			SO-DE (2009), SO-RP (2009)		SO-DE (2009), SO-RP (2009)	
2010	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol.	n.a.
			SO-DE (2013), SO-RP (2012)		SO-DE (2013), SO-RP (2012)	
2012	unspecified	unspecified	n.a.	n.a.	n.a.[a]	n.a.[a]
	LUWG-RP and MULEWF-RP (2013)	LUWG-RP and MULEWF-RP (2013)				
2013	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol.	n.a.
			SO-DE (2015), SO-RP (2015)		SO-DE (2015), SO-RP (2015)	
2016	unspecified	unspecified	obs., extrapol.	n.a.	obs., extrapol.[a]	n.a.[a]
	LU-RP and MUEEF-RP (2017)	LU-RP and MUEEF-RP (2017)	SO-DE (2018), SO-RP (2018)		SO-DE (2018), SO-RP (2018)	
2018	unspecified	unspecified	n.a.	n.a.	n.a.[a]	n.a.[a]
	LU-RP and MUEEF-RP (2019)	LU-RP and MUEEF-RP (2019)				
2019	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol.	n.a.
			SO-DE (2022), SO-RP (2022)		SO-DE (2022), SO-RP (2022)	
2020	unspecified	unspecified	n.a.	n.a.	n.a.[a]	n.a.[a]
	LU-RP and MUEEF-RP (2021)	LU-RP and MUEEF-RP (2021)				

Notes: The variables are defined in Table S44

SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-RP: Statistical Office of Rhineland-Palatinate https://www.statistischebibliothek.de/mir/receive/RPSerie_mods_00001054.

n.a.: not available.

unspecified: it is not specified whether the data correspond to observed and extrapolated values.

[a] LUWG-RP and MULEWF-RP (2013), LU-RP and MUEEF-RP (2017, 2019) and LU-RP and MUEEF-RP (2021) provide values of $J^{P}_{ps,wwtpOut,all}$ and $J^{P_{capa\geq 2000}}_{ps,wwtpOut,all}$ in 2012, 2016, 2018, and 2020. However, we discard these values because the values of $J^{P}_{ps,wwtpOut,all}$ (that vary between 261 and 372 t yr^{-1} over the period 2012–2016) appear to be lower with the values provided by SO-DE and SO-RP (that vary between 340 and 468 t yr^{-1} over the period 2010–2019).

Table S68. Source of the data of WWTP outgoing N and P loads in **Baden-Württemberg (BW)**

t	$J^{N}_{ps,wwtpOut,all}$	$J^{N_{capa\geq2000}}_{ps,wwtpOut,all}$	$J^{N_{inorg}}_{ps,wwtpOut,all}$	$J^{N_{inorg,capa\geq2000}}_{ps,wwtpOut,all}$	$J^{P}_{ps,wwtpOut,all}$	$J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$
2007	n.a.	n.a.	obs., extrapol. SO-DE (2009)	n.a.	obs., extrapol. SO-DE (2009)	n.a.
2010	unspecified LUMNBW (2011)	unspecified LUMNBW (2011)	obs., extrapol. SO-DE (2013)	n.a.	obs., extrapol. SO-DE (2013), LUMNBW (2011)	unspecified LUMNBW (2011)
2012	unspecified LUMNBW and MUKEBW (2013)	unspecified LUMNBW and MUKEBW (2013)	n.a.	n.a.	unspecified LUMNBW and MUKEBW (2013)	unspecified LUMNBW and MUKEBW (2013)
2013	n.a.	n.a.	obs., extrapol. SO-DE (2015)	n.a.	obs., extrapol. SO-DE (2015)	n.a.
2015	unspecified LUMNBW and MUKEBW (2017)	unspecified LUMNBW and MUKEBW (2017)	n.a.	n.a.	unspecified LUMNBW and MUKEBW (2017)	unspecified LUMNBW and MUKEBW (2017)
2016	n.a.	n.a.	obs., extrapol. SO-DE (2018)	n.a.	obs., extrapol. SO-DE (2018)	n.a.
2018	unspecified LUBW and MUKEBW (2019)	unspecified LUBW and MUKEBW (2019)	n.a.	n.a.	unspecified LUBW and MUKEBW (2019)	unspecified LUBW and MUKEBW (2019)
2019	n.a.	n.a.	obs., extrapol. SO-DE (2022)	n.a.	obs., extrapol. SO-DE (2022)	n.a.
2020	unspecified LUBW and MUKEBW (2021)	unspecified LUBW and MUKEBW (2021)	n.a.	n.a.	unspecified LUBW and MUKEBW (2021)	unspecified LUBW and MUKEBW (2021)

Notes: The variables are defined in Table S44

SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B).

n.a.: not available.

unspecified: it is not specified whether the data correspond to observed and extrapolated values.

Observed and extrapolated values are almost equal.

Table S69. Source of the data of WWTP outgoing N and P loads in **Bavaria (BV)**

t	$J^{N}_{ps,wwtpOut,all}$	$J^{N_{capa\geq2000}}_{ps,wwtpOut,all}$	$J^{N_{inorg}}_{ps,wwtpOut,all}$	$J^{N_{inorg,capa\geq2000}}_{ps,wwtpOut,all}$	$J^{P}_{ps,wwtpOut,all}$	$J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$
2000	not specified LfU, (2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2001	not specified LfU, (2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2002	not specified LfU, (2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2003	not specified LfU, (2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2004	not specified LfU, (2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2005	not specified LfU, (2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2006	not specified LfU, (2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2007	not specified LfU, (2021)	n.a.	obs., extrapol. SO-DE (2009)	n.a.	obs., extrapol. SO-DE (2009)	n.a.
2008	not specified LfU, (2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2009	not specified LfU, (2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2010	not specified LfU, (2021)	n.a.	obs., extrapol. SO-DE (2013)	n.a.	obs., extrapol. SO-DE (2013)	n.a.
2011	not specified LfU, (2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2012	not specified LfU, (2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2013	not specified LfU, (2021)	n.a.	obs., extrapol. SO-DE (2015), SO-BV (2015)	n.a.	obs., extrapol. SO-DE (2015), SO-BV (2015)	n.a.
2014	not specified LfU, (2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2015	not specified LfU, (2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2016	not specified LfU, (2021)	n.a.	obs., extrapol.[a] SO-BV (2020)	n.a.	obs., extrapol.[a] SO-BV (2020)	n.a.
2017	not specified LfU, (2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2018	not specified LfU, (2021)	n.a.	n.a.	n.a.	n.a.	n.a.
2019	not specified LfU, (2021)	n.a.	obs., extrapol. SO-DE (2022)	n.a.	obs., extrapol. SO-DE (2022)	n.a.

Notes: The variables are defined in Table S44

SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B);

SO-BV: Statistical Office of Bavaria (https://www.statistischebibliothek.de/mir/receive/BYSerie_mods_00000808).

n.a.: not available.

unspecified: it is not specified whether the data correspond to observed and extrapolated values.

[a] Data provided in SO-DE (2018) and SO-BV (2020) are not consistent. We adopt the values from SO-BV (2020), since it is the most recent report.

Table S70. Source of the data of WWTP outgoing N and P loads in **Saarland (SL)**

t	$J^{N}_{ps,wwtpOut,all}$	$J^{N_{capa\geq 2000}}_{ps,wwtpOut,all}$	$J^{N_{inorg}}_{ps,wwtpOut,all}$	$J^{N_{inorg,capa\geq 2000}}_{ps,wwtpOut,all}$	$J^{P}_{ps,wwtpOut,all}$	$J^{P_{capa\geq 2000}}_{ps,wwtpOut,all}$
2000	n.a.	unspecified	n.a.	n.a.	n.a.	unspecified
		MU-SL (2000)				MU-SL (2000)
2002	n.a.	unspecified	n.a.	n.a.	n.a.	unspecified
		MU-SL (2002)				MU-SL (2002)
2007	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol	n.a.
			SO-DE (2009)		SO-DE (2009)	
2008	n.a.	unspecified	n.a.	n.a.	n.a.	unspecified
		MU-SL (2008)				MU-SL (2008)
2010	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol	n.a.
			SO-DE (2013)		SO-DE (2013)	
2012	n.a.	unspecified	n.a.	n.a.	n.a.	unspecified
		MUV-SL (2013)				MUV-SL (2013)
2013	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol	n.a.
			SO-DE (2015)		SO-DE (2015)	
2014	n.a.	unspecified	n.a.	n.a.	n.a.	unspecified
		MUV-SL (2014)				MUV-SL (2014)
2016	n.a.	unspecified	obs., extrapol.	n.a.	obs., extrapol	unspecified
		MUV-SL and LUA-SL (2017)	SO-DE (2015)		SO-DE (2015)	MUV-SL and LUA-SL (2017)
2018	n.a.	unspecified	n.a.	n.a.	n.a.	unspecified
		MUV-SL and LUA-SL (2018)				MUV-SL and LUA-SL (2018)
2019	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol	n.a.
			SO-DE (2022)		SO-DE (2022)	
2020	n.a.	unspecified	n.a.	n.a.	n.a.	unspecified
		MUV-SL and LUA-SL (2021)				MUV-SL and LUA-SL (2021)

Notes: The variables are defined in Table S44

SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B).

n.a.: not available.

unspecified: it is not specified whether the data correspond to observed and extrapolated values.

Table S71. Source of the data of WWTP outgoing N and P loads in **Berlin (BE)**

t	$J^{N}_{ps,wwtpOut,all}$	$J^{N_{capa\geq 2000}}_{ps,wwtpOut,all}$	$J^{N_{inorg}}_{ps,wwtpOut,all}$	$J^{N_{inorg,capa\geq 2000}}_{ps,wwtpOut,all}$	$J^{P}_{ps,wwtpOut,all}$	$J^{P_{capa\geq 2000}}_{ps,wwtpOut,all}$
2007	n.a.	n.a.	SO-DE (2009)	SO-DE (2009)	SO-DE (2009)	SO-DE (2009)
2010	n.a.	n.a.	SO-DE (2013)	SO-DE (2013)	SO-DE (2013)	SO-DE (2013)
2013	n.a.	n.a.	SO-DE (2015)	SO-DE (2015)	SO-DE (2015)	SO-DE (2015)
2016	n.a.	n.a.	SO-DE (2018)	SO-DE (2018)	SO-DE (2018)	SO-DE (2018)
2019	n.a.	n.a.	SO-DE (2022)	SO-DE (2022)	SO-DE (2022)	SO-DE (2022)

Notes: The variables are defined in Table S44

SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B).

n.a.: not available.

Only one WWTPs is located in Berlin and it has a capacity which is larger than 2000 PE. Therefore, the observed and extrapolated values are equal.

Table S72. Source of the data of WWTP outgoing N and P loads in **Brandenburg (BB)**

t	$J^{N}_{ps,wwtpOut,all}$	$J^{N_{capa\geq2000}}_{ps,wwtpOut,all}$	$J^{N_{inorg}}_{ps,wwtpOut,all}$	$J^{N_{inorg,capa\geq2000}}_{ps,wwtpOut,all}$	$J^{P}_{ps,wwtpOut,all}$	$J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$
1991	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol.	n.a
			SO-BB (1995)		SO-BB (1995)	
1995	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol.	n.a
			SO-BB (1998)		SO-BB (1998)	
2007	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol.	n.a.
			SO-DE (2009)		SO-DE (2009)	
2010	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol.	n.a.
			SO-DE (2013)		SO-DE (2013)	
2013	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol.	n.a.
			SO-DE (2015)		SO-DE (2015)	
2016	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol.	n.a.
			SO-DE (2018)		SO-DE (2018)	
2019	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol.	n.a.
			SO-DE (2022)		SO-DE (2022)	

Notes: The variables are defined in Table S44

SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B).

SO-BB: Statistical Office of Brandenburg; (https://www.statistischebibliothek.de/mir/receive/BBSerie_mods_00000562).

n.a.: not available.

Table S73. Source of the data of WWTP outgoing N and P loads in **Mecklenburg-Vorpommern (MV)**

t	$J^{N}_{ps,wwtpOut,all}$	$J^{N_{capa\geq 2000}}_{ps,wwtpOut,all}$	$J^{N_{inorg}}_{ps,wwtpOut,all}$	$J^{N_{inorg,capa\geq 2000}}_{ps,wwtpOut,all}$	$J^{P}_{ps,wwtpOut,all}$	$J^{P_{capa\geq 2000}}_{ps,wwtpOut,all}$
2007	n.a.	n.a.	obs., extrapol. SO-DE (2009)	n.a.	obs., extrapol. SO-DE (2009)	n.a.
2010	unspecified LUNG-MV (2011)	n.a.	obs., extrapol. SO-DE (2013), SO-MV (2013)	n.a.	obs., extrapol. SO-DE (2013), SO-MV (2013), LUNG-MV (2011)	n.a.
2013	n.a.	n.a.	obs., extrapol. SO-DE (2015), SO-MV (2016)	n.a.	obs., extrapol. SO-DE (2015), SO-MV (2016)	n.a.
2016	n.a.	n.a.	obs., extrapol. SO-DE (2018), SO-MV (2019)	n.a.	obs., extrapol. SO-DE (2019), SO-MV (2019)	n.a.
2019	n.a.	n.a.	obs., extrapol. SO-DE (2022)	n.a.	obs., extrapol. SO-DE (2022)	n.a.

Notes: The variables are defined in Table S44

SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B).

SO-MV: Statistical Office of Mecklenburg-Vorpommern (https://www.statistischebibliothek.de/mir/receive/MVSerie_mods_00000334).

n.a.: not available. unspecified: it is not specified whether the data correspond to observed and extrapolated values.

Table S74. Source of the data of WWTP outgoing N and P loads in **Saxony (SN)**

t	$J^{N}_{ps,wwtpOut,all}$	$J^{N_{capa\geq2000}}_{ps,wwtpOut,all}$	$J^{N_{inorg}}_{ps,wwtpOut,all}$	$J^{N_{inorg,capa\geq2000}}_{ps,wwtpOut,all}$	$J^{P}_{ps,wwtpOut,all}$	$J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$
2007	n.a.	n.a.	obs., extrapol. SO-DE (2009)	n.a.	obs., extrapol SO-DE (2009)	n.a.
2010	n.a.	n.a.	obs., extrapol. SO-DE (2013)	n.a.	obs., extrapol SO-DE (2013)	n.a.
2013	n.a.	n.a.	obs., extrapol. SO-DE (2015)	n.a.	obs., extrapol SO-DE (2015)	n.a.
2016	n.a.	n.a.	obs., extrapol. SO-DE (2015)	n.a.	obs., extrapol SO-DE (2015)	n.a.
2019	n.a.	n.a.	obs., extrapol. SO-DE (2022)	n.a.	obs., extrapol SO-DE (2022)	n.a.

Notes: The variables are defined in Table S44

SO-DE: Statistical Office of Germany

(https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B).

n.a.: not available.

Table S75. Source of the data of WWTP outgoing N and P loads in **Saxony-Anhalt (ST)**

t	$J^{N}_{ps,wwtpOut,all}$	$J^{N_{capa\geq 2000}}_{ps,wwtpOut,all}$	$J^{N_{inorg}}_{ps,wwtpOut,all}$	$J^{N_{inorg,capa\geq 2000}}_{ps,wwtpOut,all}$	$J^{P}_{ps,wwtpOut,all}$	$J^{P_{capa\geq 2000}}_{ps,wwtpOut,all}$
1998	n.a.	n.a.	n.a.	unspecified	n.a.	unspecified
				MRLU-ST (1999)		MRLU-ST (1999)
2000	n.a.	n.a.	n.a.	unspecified	n.a.	unspecified
				MRLU-ST (2001)		MRLU-ST (2001)
2002	n.a.	n.a.	n.a.	unspecified	n.a.	unspecified
				MLU-ST (2003)		MLU-ST (2003)
2004	n.a.	n.a.	n.a.	unspecified	n.a.	unspecified
				MLU-ST (2005)		MLU-ST (2005)
2006	n.a.	n.a.	n.a.	unspecified	n.a.	unspecified
				MLU-ST (2007)		MLU-ST (2007)
2007	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol.	n.a.
			SO-DE (2009)		SO-DE (2009)	
2008	n.a.	unspecified.	n.a.	n.a.	n.a.	unspecified
		MLU-ST (2009)				MLU-ST (2009)
2010	n.a.	unspecified	obs., extrapol.	n.a.	obs., extrapol.	unspecified
		MLU-ST (2011)	SO-DE (2013)		SO-DE (2013)	MLU-ST (2011)
2012	n.a.	unspecified.	n.a.	n.a.	n.a.	unspecified
		MLU-ST (2013)				MLU-ST (2013)
2013	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol.	n.a.
			SO-DE (2015)		SO-DE (2015)	
2014	n.a.	unspecified.	n.a.	n.a.	n.a.	unspecified
		MLU-ST (2015)				MLU-ST (2015)
2016	n.a.	unspecified	obs., extrapol.	n.a.	obs., extrapol.	unspecified
		MLU-ST (2017)	SO-DE (2018)		SO-DE (2018)	MLU-ST (2017)
2019	n.a.	n.a.	obs., extrapol.	n.a.	obs., extrapol.	n.a.
			SO-DE (2022)		SO-DE (2022)	
2020	n.a.	unspecified.	n.a.	n.a.	n.a.	unspecified
		LU-ST (2021)				LU-ST (2021)

Notes: The variables are defined in Table S44

SO-DE: Statistical Office of Germany

(https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694;

https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B)

n.a.: not available.

unspecified: it is not specified whether the data correspond to observed and extrapolated values.

Table S76. Source of the data of WWTP outgoing N and P loads in **Thuringia (TH)**

t	$J^{N}_{ps,wwtpOut,all}$	$J^{N_{capa\geq2000}}_{ps,wwtpOut,all}$	$J^{N_{inorg}}_{ps,wwtpOut,all}$	$J^{N_{inorg,capa\geq2000}}_{ps,wwtpOut,all}$	$J^{P}_{ps,wwtpOut,all}$	$J^{P_{capa\geq2000}}_{ps,wwtpOut,all}$
2002	n.a.	unspecified TMLNU and TLUG (2003)	n.a.	n.a.	n.a.	unspecified TMLNU and TLUG (2003)
2004	n.a.	unspecified TMLNU and TLUG (2005)	n.a.	n.a.	n.a.	unspecified TMLNU and TLUG (2005)
2006	n.a.	unspecified TMLNU (2007)	n.a.	n.a.	n.a.	unspecified TMLNU (2007)
2007	n.a.	n.a.	obs., extrapol. SO-DE (2009)	n.a.	obs., extrapol SO-DE (2009)	n.a.
2008	unspecified TMLNU (2009)	n.a.	n.a.	n.a.	unspecified TMLNU (2009)	n.a.
2009	unspecified TMLFUN (2011)	n.a.	n.a.	n.a.	unspecified TMLFUN (2011)	n.a.
2010	n.a.	n.a.	obs., extrapol. SO-DE (2013)	n.a.	obs., extrapol SO-DE (2013)	n.a.
2011	unspecified TMLFUN (2013)	n.a.	n.a.	n.a.	unspecified TMLFUN (2013)	n.a.
2013	unspecified TMUEN (2015)	n.a.	obs., extrapol. SO-DE (2015)	n.a.	obs., extrapol. SO-DE (2015), TMUEN (2015)	n.a.
2015	unspecified TMUEN (2017)	n.a.	n.a.	n.a.	unspecified TMUEN (2017)	n.a.
2016	n.a.	n.a.	obs., extrapol. SO-DE (2018)	n.a.	obs., extrapol SO-DE (2018)	n.a.
2017	unspecified TMUEN (2019)	n.a.	n.a.	n.a.	unspecified TMUEN (2019)	n.a.
2019	unspecified TMUEN (2021)	n.a.	obs., extrapol. SO-DE (2022)	n.a.	obs., extrapol. SO-DE (2022), TMUEN (2021)	n.a.

Notes: The variables are defined in Table S44

SO-DE: Statistical Office of Germany (https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000203;jsessionid=430D199D96DF797451400D17CAD99694; https://www.statistischebibliothek.de/mir/receive/DESerie_mods_00000929;jsessionid=15ED32A1F6A7132F59E002EF6C17759B). .

n.a.: not available.

unspecified: it is not specified whether the data correspond to observed and extrapolated values.

S8.2 Visualisation of the raw and processed data of WWTP incoming and outgoing N and P emissions

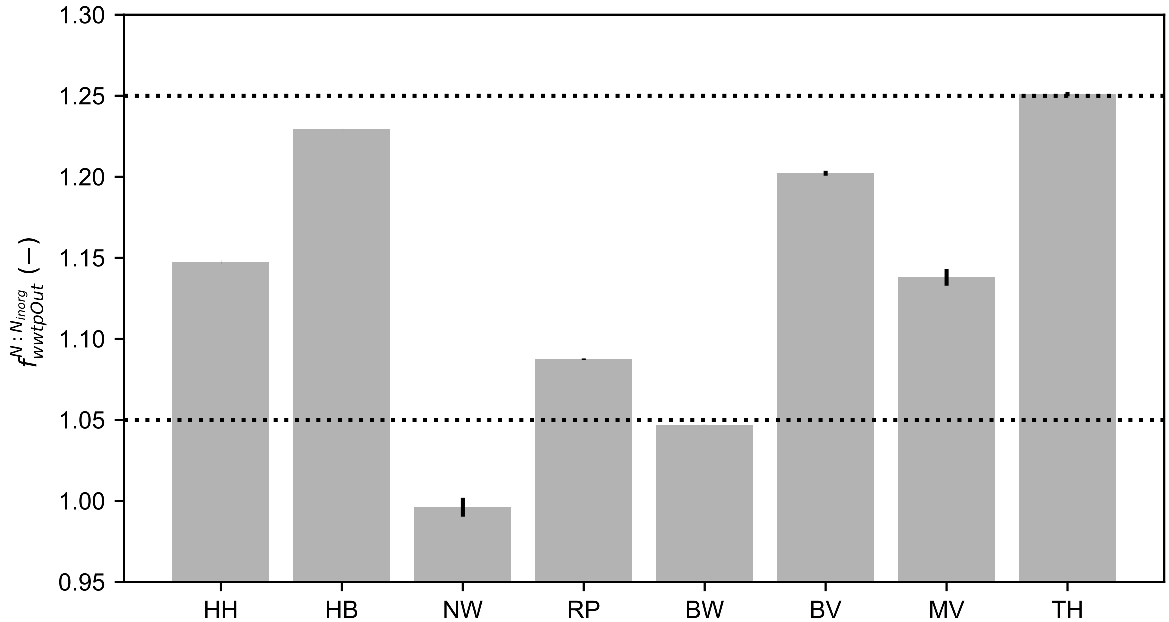


Figure S27. Ratio of total N to inorganic N in WWTP **outgoing** emissions $f_{wwtpOut}^{N:N_{inorg}}$ calculated for eight NUTS-1 regions for which both data of total N and inorganic N were available over a common period of availability. Specifically, the ratios were calculated over the period 2003–2020 for Hamburg (HH), 2006–2011 for Bremen (HB), 2006–2020 for North Rhine-Westphalia (NW), 2011–2020 for Rhineland-Palatinate (RP), 2009-2020 for Baden-Württemberg (BW), 2006–2020 for Bavaria (BV), 2009–2011 for Mecklenburg-Vorpommern (MV), and 2007–2020 for Thuringia (TH). These common period of availability as well as the N data that were used to calculate these ratios can be visualised in Fig. S28. The black error bar correspond to the value obtained using the lower bound estimate for inorganic N (observed value) and the upper bound estimate of for inorganic N (extrapolated value based on the volume of wastewater), while the grey bars indicate the average of the lower and upper bound values. The horizontal dashed black lines correspond to the lower bound (equal to 1.05) and upper bound (equal to 1.25) of the ratio that we selected to estimate total N from inorganic N for WWTP outgoing emissions (estimates of total N can be visualised in Fig. S28).

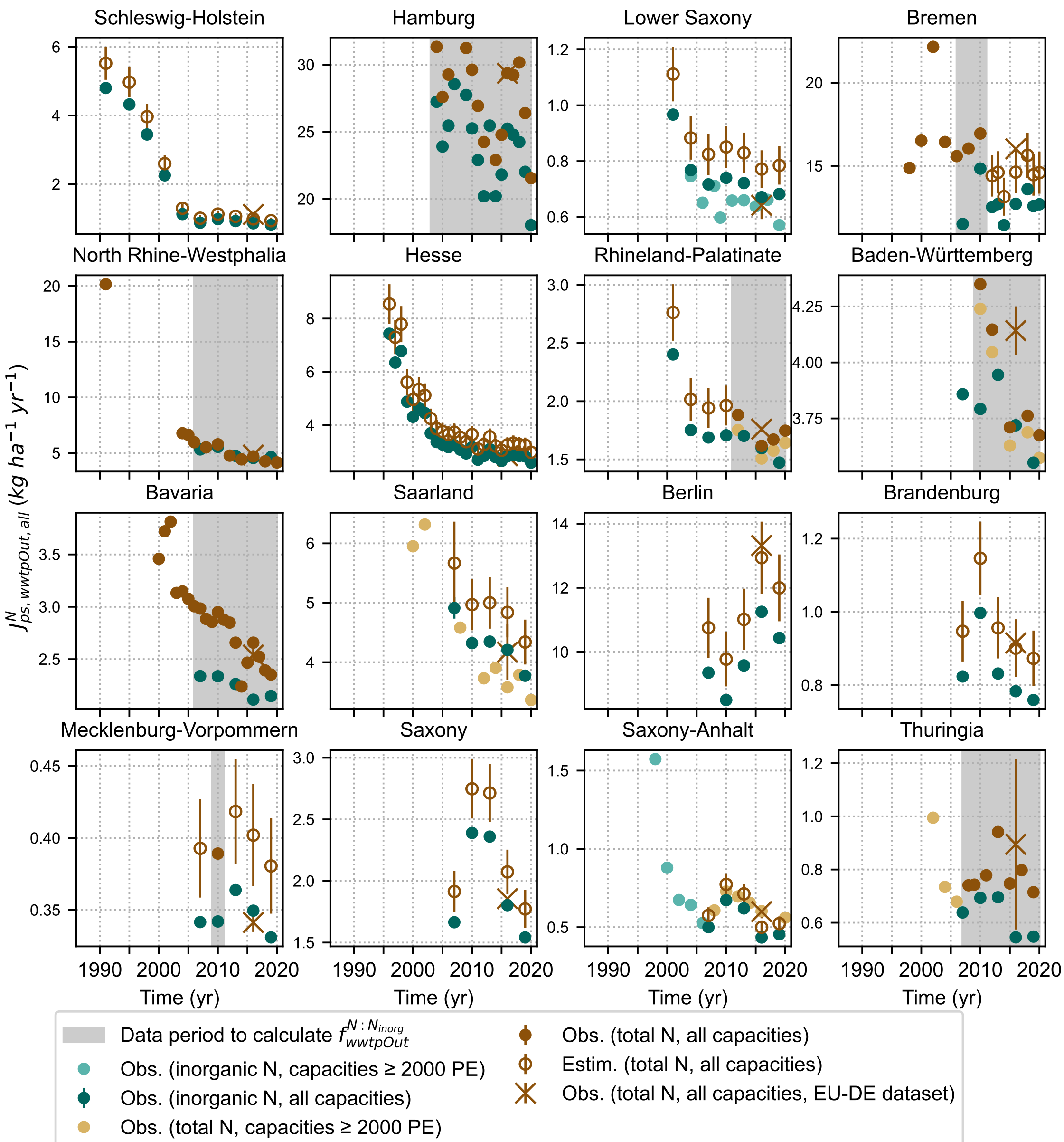


Figure S28. Observed WWTP **outgoing N** load at NUTS-1 level. Values come from (1) data available at NUTS-1 level from the sources described in Sect. S8.1.2 (filled circles), (2) estimates of total N calculated from inorganic N using a ratio reported in Fig. S27 (empty circles), (3) estimates of total N load in 2016 derived from observations available at the WWTP level from the EU and DE dataset as described in Sect. S9 (crosses). Some values are reported with error bars that depict a lower and upper bound estimates, while the markers (circles or crosses) represent the average of the lower and upper bound estimates. Uncertainty estimates come from (1) an extrapolation based on the volume of wastewater (filled circles), (2) the estimation of total N from inorganic N data (empty circles), and (3) the lower and upper estimates that we derived for the combined EU/DE dataset as explained in Sect. S9.2 (crosses). The grey shaded areas report the period over which the ratios of total N to inorganic N (shown in Fig. S27) were calculated. The load values shown in the figure are normalised by the area of each NUTS-1 region.

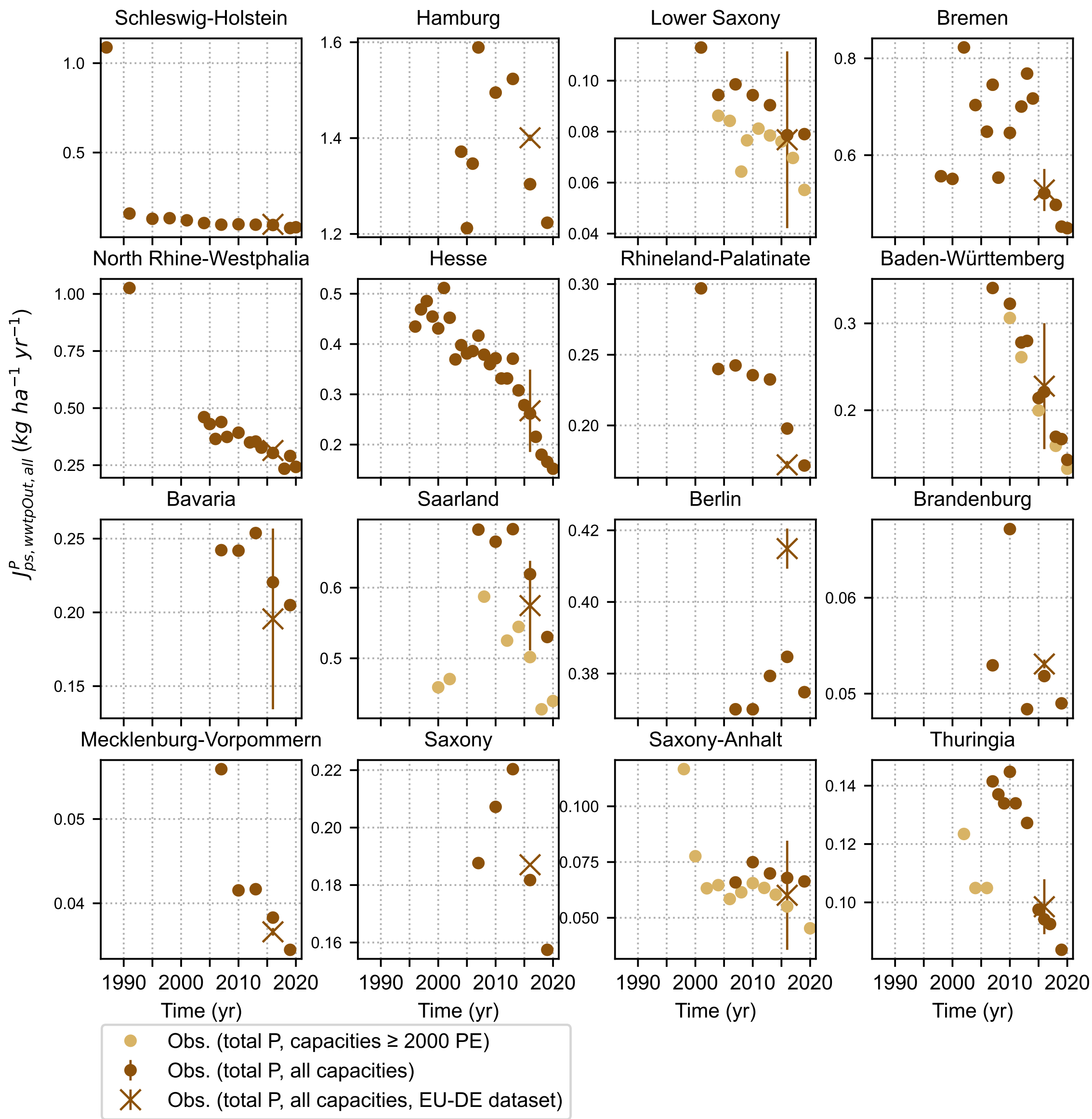


Figure S29. Observed WWTP **outgoing P** load at NUTS-1 level. Values come from (1) data available at NUTS-1 level from the sources described in Sect. S8.1.2 (filled circles), and (2) estimates of total P load in 2016 derived from observations available at the WWTP level in the EU and DE datasets as described in Sect. S9 (crosses). Some values are reported with error bars that depict a lower and upper bound estimates, while the markers (circles or crosses) represent the average of the lower and upper bound estimates. Uncertainty estimates come from (1) an extrapolation based on the volume of wastewater (filled circles), and (2) the lower and upper estimates that we derived for the combined EU/DE dataset as explained in Sect. S9.2 (crosses). The load values shown in the figure are normalised by the area of each NUTS-1 region.

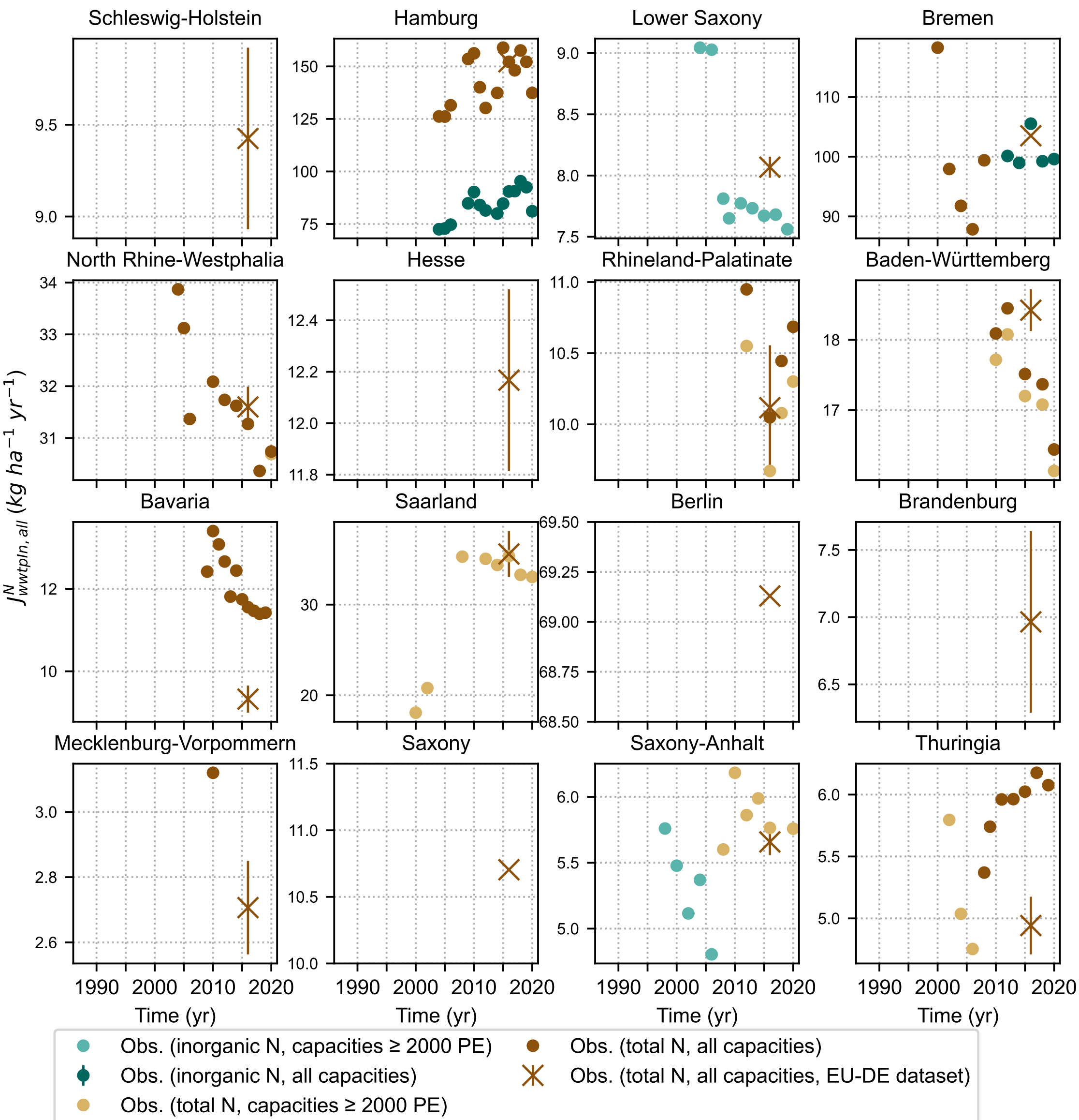


Figure S30. Observed WWTP **incoming N** load at NUTS-1 level. Values come from (1) data available at NUTS-1 level from the sources described in Sect. S8.1.2 (filled circles), and (2) estimates of total N load in 2016 derived from observations available at the WWTP level in the EU and DE datasets as described in Sect. S9 (crosses). Some values are reported with error bars that depict a lower and upper bound estimates, while the markers (circles or crosses) represent the average of the lower and upper bound estimates. Uncertainty estimates come from (1) an extrapolation based on the volume of wastewater (filled circles), and (2) the lower and upper estimates that we derived for the combined EU/DE dataset as explained in Sect. S9.2 (crosses). The load values shown in the figure are normalised by the area of each NUTS-1 region.

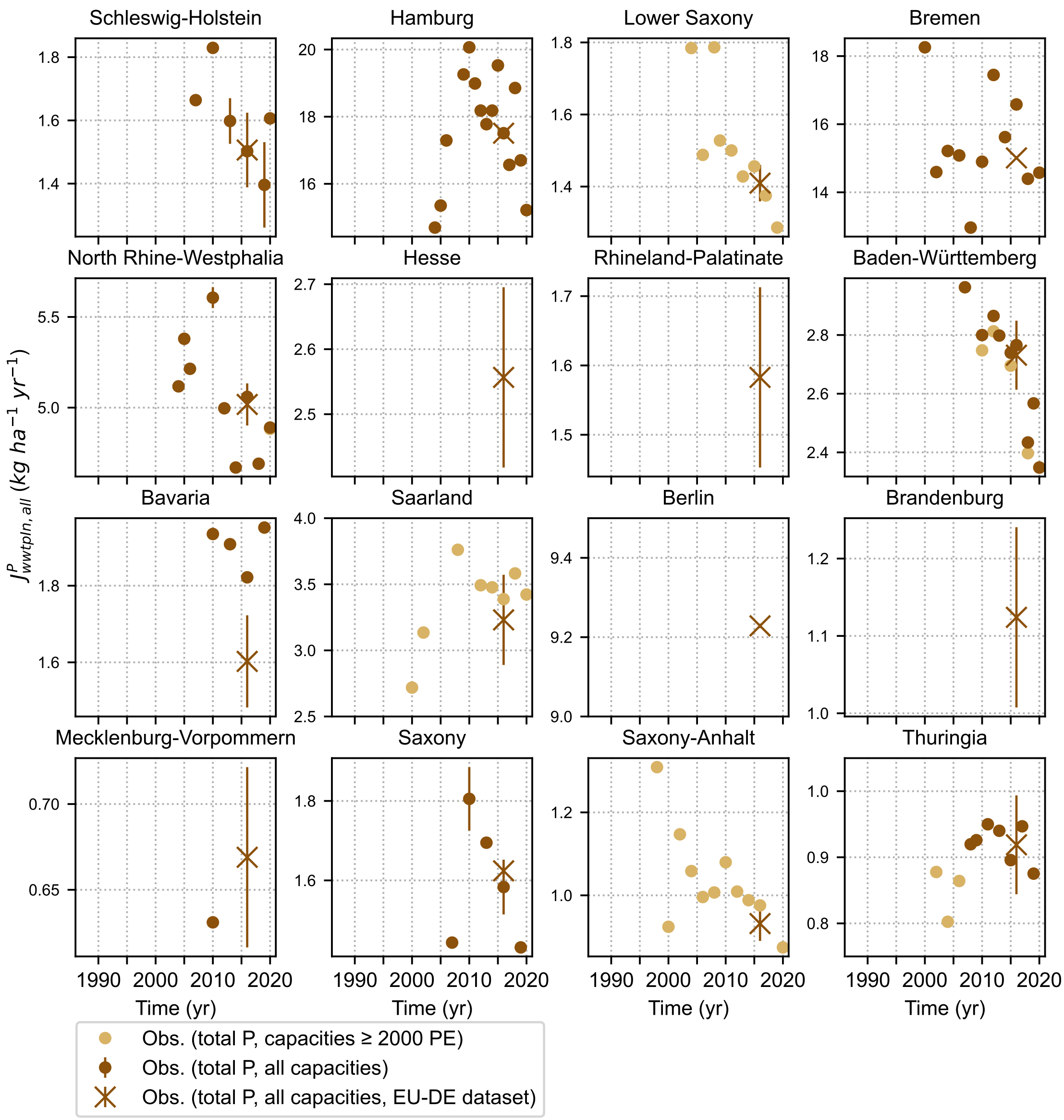


Figure S31. Observed WWTP **incoming P** load at NUTS-1 level. Values come from (1) data available at NUTS-1 level from the sources described in Sect. S8.1.2 (filled circles), and (2) estimates of total P load in 2016 derived from observations available at the WWTP level in the EU and DE datasets as described in Sect. S9 (crosses). Some values are reported with error bars that depict a lower and upper bound estimates, while the markers (circles or crosses) represent the average of the lower and upper bound estimates. Uncertainty estimates come from (1) an extrapolation based on the volume of wastewater (filled circles), and (2) the lower and upper estimates that we derived for the combined EU/DE dataset as explained in Sect. S9.2 (crosses). The load values shown in the figure are normalised by the area of each NUTS-1 region.

S9 Observations of WWTP incoming and outgoing N and P emissions at WWTP level

We construct a dataset of WWTPs incoming and outgoing N and P emissions around the year 2016 encompassing a total 9006 WWTPs that are either reported in the EU dataset (Waterbase versions 4 to 8 for reporting years 2010 to 2018; EEA, 2023) only, that is, 2645 WWTPs, or in the DE dataset (Büttner, 2020) only, that is, 4704 WWTPs, or in both datasets, that is, 1657 WWTPs. 92 additional WWTPs reported in the EU dataset are not considered, since they are indicated as not being active in 2016.

We combine the incoming and outgoing N and P load records provided in the EU and DE datasets. We derive a lower and upper bound estimate of the incoming and outgoing N and P loads for each WWTP. Our uncertainty estimates account for the uncertainties arising from (1) the discrepancies in the values of the load between the EU and DE dataset for the 1657 WWTPs that are reported in both datasets, (2) the low precision of the load values reported in the EU dataset, (3) our gap-filling procedure that we apply to estimate the value of the load when it is not reported, using either values from other years when available or an extrapolation approach based on the entering load expressed in PE (population equivalent).

In the following we provide details on the matching procedure we used to identify the 1657 WWTPs that are present in both the EU and DE datasets (Sect S9.1) and on the methodology used to estimate the lower and upper bound estimates of the load in our combined dataset (Sect S9.2).

S9.1 Matching procedure to identify WWTPs that are present in both the EU and DE datasets

Identifying the WWTPs that are present in both the EU and DE dataset is not straightforward, due to the inconsistencies and uncertainties in the datasets. For each WWTP in the EU dataset, we search for potentially matching WWTP in the DE dataset within a 10 km radius, which we consider to be large enough to account for reasonable uncertainties in the WWTPs coordinates. Uncertainties in the coordinates could arise in particular from coordinate conversions that must have been done between the coordinate systems used in the different NUTS-1 regions (typically Gauss-Krüger systems) and the coordinate system WGS84 used in the EU dataset and in this study.

We first filter out WWTPs that do not show similarities in their names. For the WWTPs having a similar name, we further check that the potentially matching WWTPs have at least one other characteristic which shows consistency in particular intrinsic properties such as the coordinates and the design capacity (which is expressed in PE). Our **first criterion** (denoted as $Crit_{dist}$) for assessing the consistency in the **coordinates** is that coordinates are consistent when the distance between the two WWTP records is less than 100 m. Our **second criterion** (denoted as $Crit_{capa}$) for assessing the consistency in the **design capacity** is that the design capacities are consistent when their relative difference is less than 25 %. For the latter criterion, we examine the value of the design capacity provided for all years in the EU dataset (2010, 2012, 2014, 2016, and 2018), that is, we consider the criterion as being satisfied when the value of the design capacity is consistent to that of the EU record for at least one of the reporting years. Since the design capacity can be missing from the record or can have large uncertainties, we define a **third criterion** (denoted as $Crit_{PE}$) for assessing the consistency in the **entering load expressed in PE**. This criterion is similar to that for the design capacity in that we consider that PE are consistent when their relative difference is less than 25 %. Unlike the name, coordinates and design capacities that are intrinsic properties of the WWTPs, their PE changes in time. Therefore we expect to see larger differences in the PE between the EU and DE datasets than in the name, coordinates and design capacities, due to possible inconsistencies in the reporting period between the two datasets. For a similar reason, we do not define criteria based on the value of the incoming and outgoing N and P loads because of their temporal variability. We consider the value of the load only when WWTPs that have a similar name but none of the three criteria based on the coordinates, design capacity and PE is satisfied.

Using the above-described procedure, we identify 1657 WWTPs that have a record in both the EU and DE datasets. 98.4 % of these WWTPs (1631 WWTPs) have the exact same value of the design capacity in the two datasets, while for only 50.7 % (838 WWTPs) the distance between the coordinates present in both datasets is less than 100 m ($Crit_{dist}$) . Although we observe some discrepancies between the coordinates reported in the EU and DE dataset, the distance calculated between the reported coordinates is lower than 1 km for 96 % of the WWTPs (see Fig S32). The uncertainty in the coordinates is thus mostly within to our target grid resolution of 0.01625°, which is on average 1.74 km x 1.09 km over Germany. Table S77 provides further details on the number of WWTPs that satisfy the different criteria.

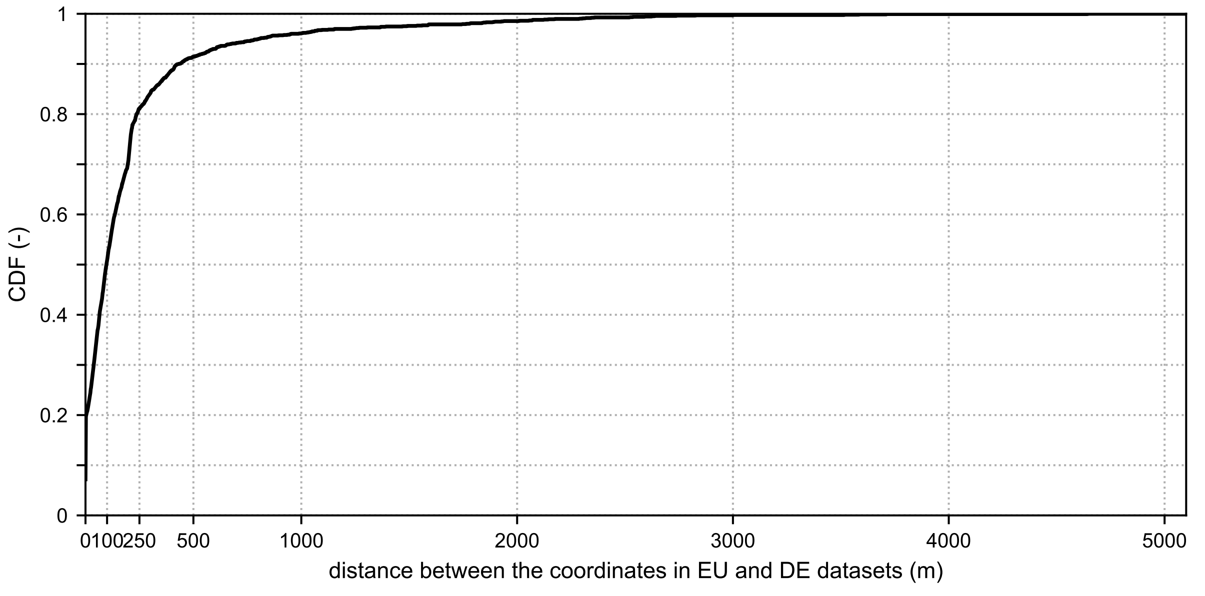


Figure S32. Cumulative Distribution Function (CDF) of the distance between the coordinates reported in the EU and DE datasets for the 1657 WWTPs that are present in both datasets. The distance is equal to 0m (i.e. the coordinates are identical in both datasets) for 7.2 % of the WWTPs, and is less or equal to 10 m, 100 m, 250 m, 500 m, and 1000 m, for 21.1 %, 50.6 %, 81 %, 91 %, and 96 % of the WWTPs, respectively. The maximum value of the distance is 5100 m.

Table S77. Matching criteria (distance, design capacity, and PE) for the 1657 WWTPs that are present in both the EU and DE datasets

		$Crit_{capa}$: yes			$Crit_{capa}$: no			$Crit_{capa}$: Capacity missing[a]			Total
		$Crit_{PE}$: satisfied	$Crit_{PE}$: not satisfied	$Crit_{PE}$: PE missing[a]	$Crit_{PE}$: satisfied	$Crit_{PE}$: not satisfied	$Crit_{PE}$: PE missing[a]	$Crit_{PE}$: satisfied	$Crit_{PE}$: not satisfied	$Crit_{PE}$: PE missing[a]	
$Crit_{dist}$	satisfied	768	65	0	2	2	0	0	0	1	838
	not satisfied	643	138	33	3	0	**1**[b]	0	0	**1**[b]	819
Total		1647			8			2			1657

Notes: The table reports the number of WWTPs that satisfy or not each of the three criteria: (1) $Crit_{dist}$: criterion for assessing the consistency in the coordinates, prescribing that coordinates are consistent when the distance between the two WWTP records is less than 100 m; (2) $Crit_{capa}$: criterion for assessing the consistency in the design capacity, prescribing that the design capacities are consistent when their relative difference is less than 25 %; (3) $Crit_{PE}$: criterion for assessing the consistency in the entering load expressed in PE prescribing, that the PE are consistent when their relative difference is less than 25 %.

[a] $Crit_{capa}$ and $Crit_{PE}$ cannot be assessed for some WWTPs, since the design capacity and the PE, respectively, are missing from the record in the EU or DE dataset.

[b] Two WWTPs do not satisfy any of the three criteria. For one of these two WWTPs, we verify that the incoming and outgoing N and P loads are consistent in the EU and DE record. For the other WWTP, no value of the design capacity, the PE, as well as incoming and outgoing N and P loads are provided in the DE dataset. Therefore considering that these WWTP records are matching or not does not have an impact on our final WWTP dataset.

S9.2 Estimation of the value (upper and lower bound)of the incoming and outgoing N and P load in 2016

We derive estimates of the N and P load around the year 2016, from the reported values in the EU dataset in 2016 (Waterbase v7) and from the reported values in the DE dataset that correspond to 2015 or 2016 depending on the WWTPs. We estimate a lower and upper bound for each WWTP w, that we denote as $\hat{Y}_{lb}(w)$ *($kg\ yr^{-1}$)* and $\hat{Y}_{ub}(w)$ *($kg\ yr^{-1}$)*, respectively, where Y is among $J^N_{wwtpIn,all}$ (incoming N load), $J^P_{wwtpIn,all}$ (incoming P load), $J^N_{ps,wwtpOut,all}$ (outgoing N load), $J^P_{ps,wwtpOut,all}$ (outgoing P load).

The precision of the load data in the EU dataset for the 2016 reporting year is coarse (1000 $kg\ yr^{-1}$), while the precision is much higher in the DE dataset (1 $kg\ yr^{-1}$). For this reason, for a WWTP w_{EU} that is reported in the EU dataset only, we set the 2016 lower and upper bound estimates by adding $\pm 500\ kg\ yr^{-1}$ to the 2016 reported value denoted as $Y^{EU}(2016, w_{EU})$ *($kg\ yr^{-1}$)*:

$$\begin{aligned}
\hat{Y}_{lb}(w_{EU}) &= max(Y^{EU}(2016, w_{EU}) - 500, 0) \\
\hat{Y}_{ub}(w_{EU}) &= Y^{EU}(2016, w_{EU}) + 500
\end{aligned} \tag{S30}$$

In addition, for 8 WWTPs reported in the EU dataset, it is indicated that they are inactive in 2016, while some load values are provided. Therefore one of the reported information is incorrect (either the inactive status or the load values). For these 8 WWTPS, we set the lower bound $\hat{Y}_{lb}(w_{EU})$, of Eq.S30 to 0, taking into account the fact that they may have been inactive in 2016.

For a WWTP w_{DE} that is reported in the DE dataset only, we set the 2016 lower and upper bound estimates to the 2015/2016 reported value (depending on the WWTPs, the value is provided for year 2015 or 2016) denoted as $Y^{DE}(2015/2016, w_{DE})$ $(kg\ yr^{-1})$:

$$\hat{Y}_{lb}(w_{EU}) = Y^{DE}(2015/2016, w_{DE})$$
$$\hat{Y}_{ub}(w_{EU}) = Y^{DE}(2015/2016, w_{DE}) \tag{S31}$$

For a WWTP $w_{EU\&DE}$ that is reported in the both the EU and DE datasets, we set the 2016 lower and upper bound estimates to the 2015/2016 value of the DE dataset when the values in the EU and DE dataset are consistent. Otherwise, our uncertainty estimates account for the discrepancies in the values between the two datasets. The specific equations are as follows:

$$when\ Y^{EU}(2016, w_{EU\&DE}) - 500 \leq Y^{DE}(2015/2016, w_{EU\&DE}) \leq Y^{EU}(2016, w_{EU\&DE}) + 500:$$
$$\hat{Y}_{lb}(w_{EU\&DE}) = Y^{DE}(2015/2016, w_{EU\&DE})$$
$$\hat{Y}_{ub}(w_{EU\&DE}) = Y^{DE}(2015/2016, w_{EU\&DE})$$
$$when\ Y^{DE}(2015/2016, w_{EU\&DE}) < X^{EU}(2016, w_{EU\&DE}) - 500$$
$$or\ Y^{DE}(2015/2016, w_{EU\&DE}) > Y^{EU}(2016, w_{EU\&DE}) + 500:$$
$$\hat{Y}_{lb}(w_{EU\&DE}) = min(Y^{DE}(2015/2016, w_{EU\&DE}), Y^{EU}(2016, w_{EU\&DE}) - 500)$$
$$\hat{Y}_{ub}(w_{EU\&DE}) = max(Y^{DE}(2015/2016, w_{EU\&DE}), Y^{EU}(2016, w_{EU\&DE}) + 500) \tag{S32}$$

For some WWTP, no load estimates can be derived using Eq. S30–S32 because no load values are reported in the EU and DE datasets. We fill these values, as explained in the following. First, we first try, for the WWTPs that are reported in the EU dataset (including the WWTPs that are reported in both datasets), to fill the gaps using values for the other years when available, as follows:

$$\hat{Y}_{lb}(w_{EU}) = min(X^{EU}(2010, w_{EU}), X^{EU}(2012, w_{EU}), X^{EU}(2014, w_{EU}) - 500, X^{EU}(2018, w_{EU}) - 500)$$
$$\hat{Y}_{ub}(w_{EU}) = max(X^{EU}(2010, w_{EU}), X^{EU}(2012, w_{EU}), X^{EU}(2014, w_{EU}) + 500, X^{EU}(2018, w_{EU}) + 500) \tag{S33}$$

Equation S33 accounts for the fact that the precision of the load data in the EU dataset changes across years. While the precision is high for years 2010 and 2012 (1 $kg\ yr^{-1}$) it is low for years 2014, 2016, and 2018 (1000 $kg\ yr^{-1}$). Therefore, in Eq. S33, we account for the uncertainty in the reported load in 2014 and 2018 by adding $\pm 500\ kg\ yr^{-1}$, similar to the reported load in 2016 in Eq. S30. Furthermore, the status of some WWTPs reported in the EU dataset is not clear, because these WWTPs are not present in the 2016 reporting, while no closing date prior to year 2016 or starting date later than year 2016 is provided. Therefore, we do not know whether these WWTPs were inactive in 2016, or whether they were omitted from the 2016 reporting. For these WWTPs that have an unknown status and that also have some load values in other years, we set the lower bound estimate ($\hat{Y}_{lb}(w_{EU})$) of Eq S33 to 0, to account for the fact that they may have been inactive in 2016. The estimation procedure of Eq. S33 cannot be applied to WWTPs that are reported in the DE dataset only, since only one year of data is included for each WWTP.

Second, for a WWTP w for which the load value is not reported in 2016 or in other years (and therefore the value of the load can not be estimated using Eq. S33), we extrapolate the value of the upper bound of the load, calculated for the WWTP that have a load record with Eq. S30–S33, based on the entering load expressed in PE (population equivalent). To account for the uncertainty related to the extrapolation procedure based on the PE of Eq. S34, we set the lower bound of the load to 0. The equations are as follows:

$$\hat{Y}_{lb}(w) = 0$$
$$\hat{Y}_{ub}(w) = \frac{\sum_{k \in E_{WWTP,data}} \hat{Y}_{ub}(k)}{\sum_{k \in E_{WWTP,value}} L_{PE}(k)} L_{PE}(w) \tag{S34}$$

where $L_{PE}(w)$ *(PE)* is the entering load expressed in PE for WWTP w, $E_{WWTP,data}$ is the ensemble of WWTP for which a value of the $\hat{Y}_{ub}(k)$ could be estimated using Eq. S30–S33 and for which a estimate of $L_{PE}(k)$ can be derived. When L_{PE} of Eq. S34 is provided in both the EU and DE datasets, we use the average value. When it is not provided for the year 2016, we either use the average value across the other years, if provided in the EU dataset, or we use the value of the design capacity (expressed in PE as well) in 2016 or its average value across the other years.

Our final dataset comprises values of the incoming and outgoing N and P loads for over 99 % of the 9006 WWTPs, while only for 39 WWTPs no values at all of the N and P loads can be estimated and for 16 WWTPs no value of the incoming N and P loads and the outgoing N loads can be derived, as shown in Fig S33. Moreover, although the number of WWTPs for which the values of the load have to be estimated based on PE (using Eq. S34) is large, in particular for the outgoing N load for the NUTS-1 region of Thuringia or more in general for the N and P incoming load for all NUTS-1 regions (light red color in Fig S33), the corresponding values of the loads (light red color in Fig S34) are actual very small compared to the values that are derived from data (dark blue color in Fig S34). This means that the missing value are for smaller WWTPs and that the impact of these missing value on the total load at NUTS-1 level is limited.

We also assess the width of the uncertainty interval (that is the difference between the upper and lower bound values, denoted as UI) at NUTS-1 level, as reported in Table S78. UI is in general relatively contained and represents less than 25 % of the average value of the upper and lower bound estimates. The uncertainty interval is however larger for the outgoing N load for the NUTS-1 region of Thuringia, where it is around 72 % of the average estimate. This is due to inconsistencies in the load record in the EU (for year 2016) and DE dataset (for year 2015) for one WWTP. This could be due large inter-annual variability in the load for this WWTP or errors in the records. We also verify that average load values in the combined EU/DE dataset (average over the lower and upper bound estimates) are consistent with the statistical data that we collected at NUTS-1 level (described in Sect. S8), with values of the relative differences always lower than 20 % (Table S78). The average, lower and upper bound values of the combined EU/DE datasets at NUTS-1 level are reported in Fig. S28–S31, which also shows the load data collected at NUTS-1 level from the statistical offices of Germany and the federal states and the authorities of the states.

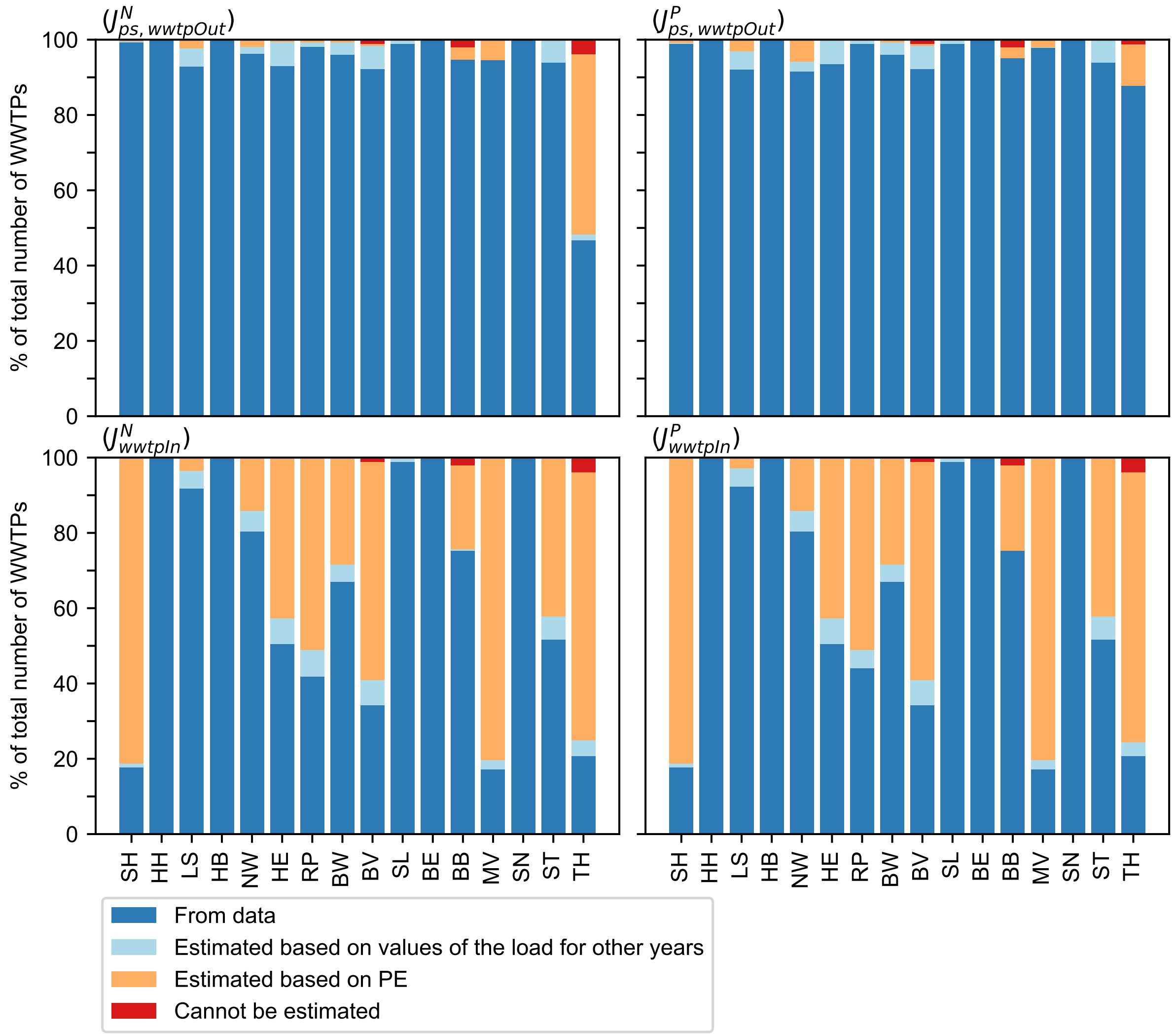


Figure S33. Distribution of the WWTPs in the combined EU/DE dataset according to the source of the load data at NUTS-1 level. Specifically, the figure shows the percentage of WWTPs in the combined dataset for which (1) the data provide the value of the load for the year 2016 (or 2015) either in the EU or in the DE dataset (dark blue; Eq. S30–S32), (2) the value of the load is filled based on values for the other years provided in the EU dataset (light blue; Eq. S33), (3) the value of the load is filled based on the value of the entering load expressed in population equivalent (PE; light red; Eq. S34), and (4) the value of the load cannot be estimated (dark red). The figure reports these percentages for the outgoing N load ($J^N_{ps,wwtpOut}$), the outgoing P load ($J^P_{ps,wwtpOut}$), the incoming N load (J^N_{wwtpIn}), and the incoming P load (J^P_{wwtpIn}), and for the 16 NUTS-1 regions of Germany that are defined in Table S1.

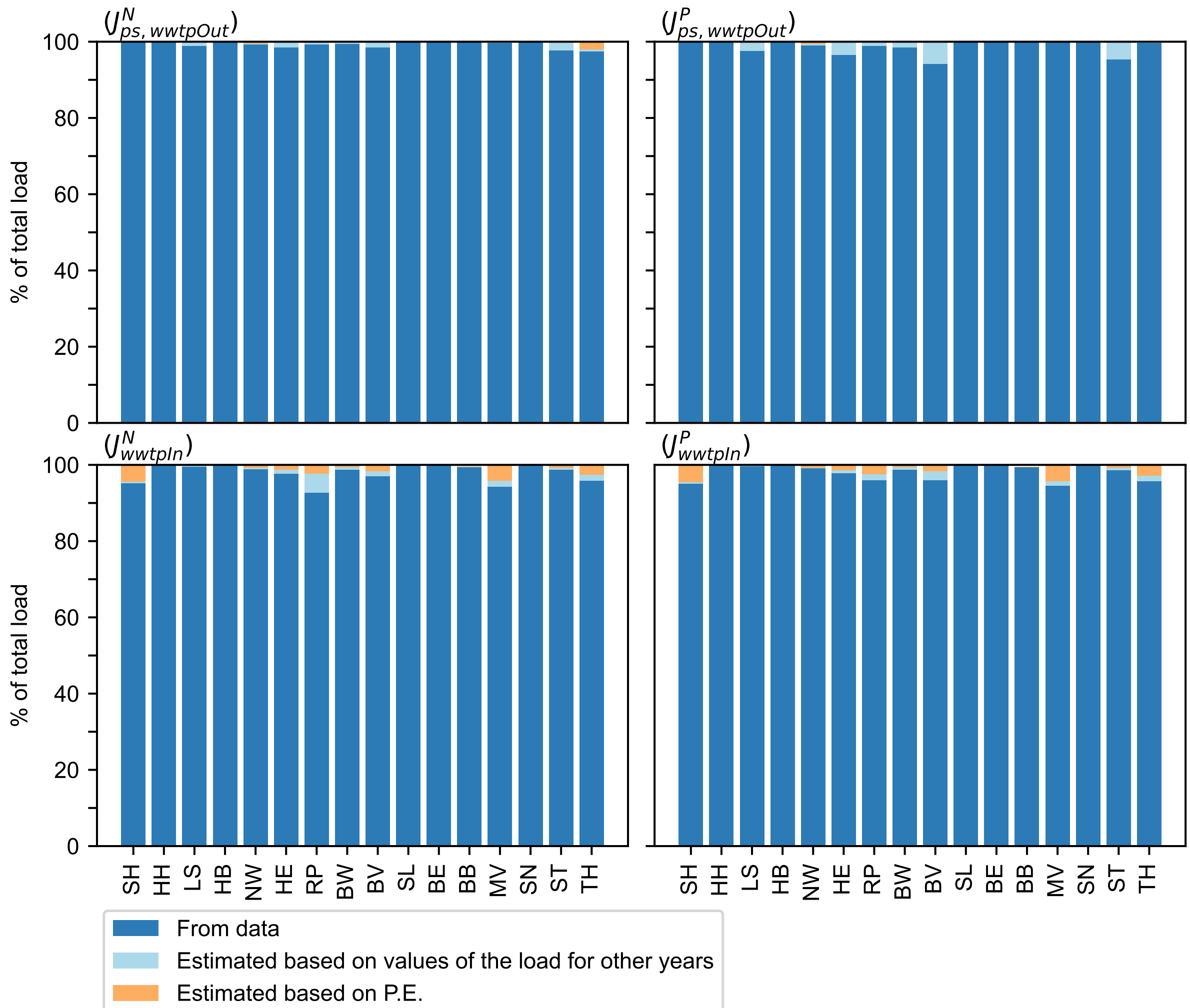


Figure S34. Distribution of the load in the combined EU/DE dataset according to the source of the load data at NUTS-1 level. Specifically, percentage of the load in the combined dataset that come from (1) data for the year 2016 (or 2015) provided either in the EU dataset or in the DE dataset (dark blue; Eq. S30–S32), (2) estimates based on values for the other years provided in the EU dataset (light blue; Eq. S33), (3) estimates based on the value of the entering load expressed in population equivalent (PE; light red; Eq. S34). The figure reports these percentages for the outgoing N load ($J^N_{ps,wwtpOut}$), the outgoing P load ($J^P_{ps,wwtpOut}$), the incoming N load (J^N_{wwtpIn}), and the incoming P load (J^P_{wwtpIn}), and for the 16 NUTS-1 regions of Germany that are defined in Table S1. The figure was drawn using the average of the upper and lower bound load estimates. For each NUTS-1 region, the values of the load that come from data account for at least 97.5 %, 94.2 %, 92.8 %, 94.5 % of the total value of $J^N_{ps,wwtpOut}$, $J^P_{ps,wwtpOut}$, J^N_{wwtpIn}, and J^P_{wwtpIn}, respectively.

Table S78. Statistics of the combined EU/DE dataset in 2016 at NUTS-1 level: width of the uncertainty interval (UI) and difference to the value in the NUTS-1 level dataset of Sect. S8 ($BIAS$).

NUTS-1	$J^N_{ps,wwtpOut,all}$		$J^P_{ps,wwtpOut,all}$		$J^N_{wwtpIn,all}$		$J^P_{wwtpIn,all}$	
region	UI (%)	$BIAS$ (%)	UI (%)	$BIAS$ (%)	UI (%)	$BIAS$ (%)	UI (%)	$BIAS$ (%)
Schleswig-Holstein	11.23	3.64	2.15	0.32	15.67	n.a.	10.5	0.0
Hamburg	0.05	0.0	0.1	0.33	0.08	0.0	0.01	0.0
Lower Saxony	15.47	8.13	9.03	0.2	7.19	n.a.	2.13	n.a.
Bremen	0.62	0.91	1.65	0.05	0.66	n.a.	0.1	9.47
North Rhine-Westphalia	1.4	2.71	0.23	0.23	4.62	1.05	2.48	0.82
Hesse	8.74	3.84	6.13	0.16	10.85	n.a.	5.8	n.a.
Rhineland-Palatinate	2.47	8.89	0.32	1.59	16.43	0.67	8.68	n.a.
Baden-Württemberg	5.21	n.a.	6.36	n.a.	8.62	n.a.	3.22	1.24
Bavaria	7.64	4.44	6.27	0.93	15.0	19.3	7.06	12.01
Saarland	22.3	5.12	2.21	0.92	21.12	n.a.	14.21	n.a.
Berlin	0.08	0.0	0.27	0.23	0.12	n.a.	0.02	n.a.
Brandenburg	3.18	0.0	0.17	0.14	20.73	n.a.	19.4	n.a.
Mecklenburg-Vorpommern	3.84	6.25	0.25	0.41	15.71	n.a.	10.59	n.a.
Saxony	0.0	1.78	0.0	0.26	0.0	n.a.	0.0	0.0
Saxony-Anhalt	14.13	11.24	8.14	1.55	8.96	n.a.	3.57	n.a.
Thuringia	71.62	n.a.	1.91	n.a.	16.28	n.a.	9.43	n.a.

Variables definition: $UI = \frac{\hat{Y}_{ub} - \hat{Y}_{lb}}{\frac{\hat{Y}_{ub}+\hat{Y}_{lb}}{2}} 100$

where $\hat{Y}_{lb}$ $(kg\ yr^{-1})$ and $\hat{Y}_{ub}$ $(kg\ yr^{-1})$ are the lower upper bound estimate in the combine EU/DE dataset, respectively.

$BIAS = max(\frac{\hat{Y}_{ub}+\hat{Y}_{lb}}{2} - Y^{NUTS-1}_{ub}, 0) + max(Y^{NUTS-1}_{lb} - \frac{\hat{Y}_{ub}+\hat{Y}_{lb}}{2}, 0)$

where $\hat{Y}^{NUTS-1}_{lb}$ $(kg\ yr^{-1})$ and $\hat{Y}^{NUTS-1}_{ub}$ $(kg\ yr^{-1})$ are the lower upper bound estimate in the NUTS-1 dataset of Sect. S8.

Notations: $J^N_{ps,wwtpOut,all}$ is the WWTP outgoing N load, $J^P_{ps,wwtpOut,all}$ is the WWTP outgoing P load, $J^N_{wwtpIn,all}$ is the WWTP incoming N load, and $J^P_{wwtpIn,all}$ is the WWTP incoming P load.

n.a.: no load estimates are available in the NUTS-1 dataset in 2016.

S10 N and P emissions realisations

S10.1 Evaluation of the point source estimates at NUTS-0 and NUTS-1 level

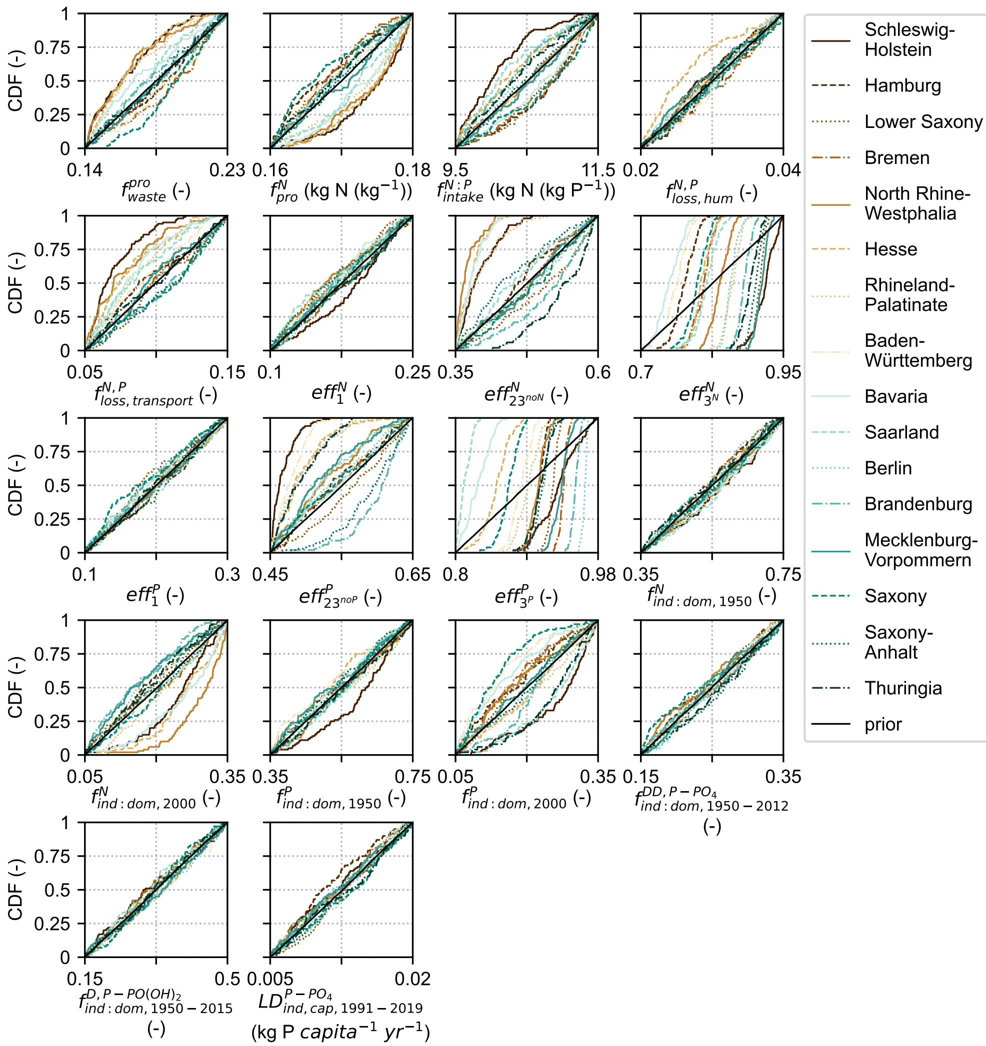


Figure S35. Cumulative distribution functions (CDFs) of the 18 point sources model parameters (defined in Table S2) at NUTS-1 level. Colored lines refer to the posterior CDFs resulting from the 100 posterior parameterisations, and grey lines refer to the prior CDFs in the original sample of size 100,000. The prior CDFs are all uniform and are identical for all NUTS-1 regions. Parameters for which the posterior distributions is different from the prior distribution are influential for the metrics used for parameter estimation (here root mean square error), for instance the efficiency of tertiary treatment with N and P removal (eff_3^N and eff_3^P). However, the fact that the distributions are the same (for instance the industrial/commercial detergent parameters ($f_{ind:dom,1950-2012}^{DD,P-PO_4}$, $f_{ind:dom,1950-2015}^{D,P-PO(OH)_2}$, and $LD_{ind,cap,1991-2019}^{P-PO_4}$) does not necessarily mean that the parameter is not influential, as it could have an impact through interactions with other parameters.

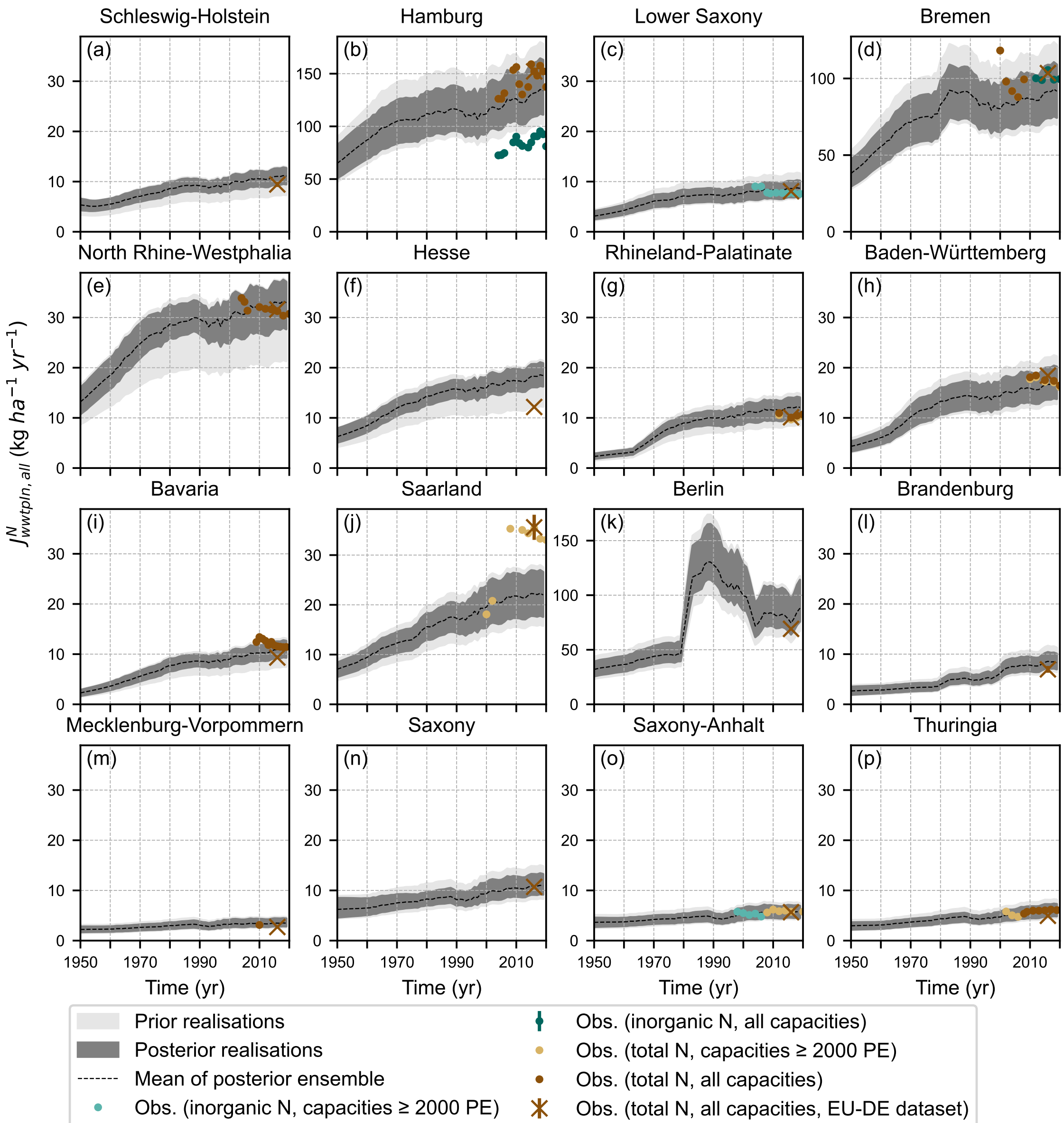


Figure S36. WWTP incoming N loads $J^N_{wwtpIn,all}$, including both the urban and rural components, from the prior and posterior model realisations (grey shaded areas and dashed lines), and observations (coloured circles and crosses) at NUTS-1 level for the period 1950–2019. The shaded areas delineate the minimum and maximum values of the 100,000 prior realisations (light grey) and the 100 posterior realisations (dark grey). Observational values are shown for both the ensemble of WWTPs having a design capacity higher than 2000 PE (light green and brown) and for all WWTPs (dark green and brown). Some observations are reported with error bars that depict the upper and lower bound estimates while the markers (circles/crosses) represent the average value. The crosses shows the data from the combined EU/DE dataset that were aggregated at NUTS-1 level. The scale on the y-axis is the same for all NUTS-1 regions but the city states (Hamburg, Bremen, and Berlin) for which the loads are much higher compared to the other NUTS-1 regions. Data source: observational data come from the statistical offices of Germany and the federal states and from the authorities of the federal states (circles), and the EU Waterbase dataset EEA (2023) and Büttner (2020) (crosses). The uncertainty intervals on the observations were processed in this study. Details on observational data sources and processing are in Sect. S8–S9.

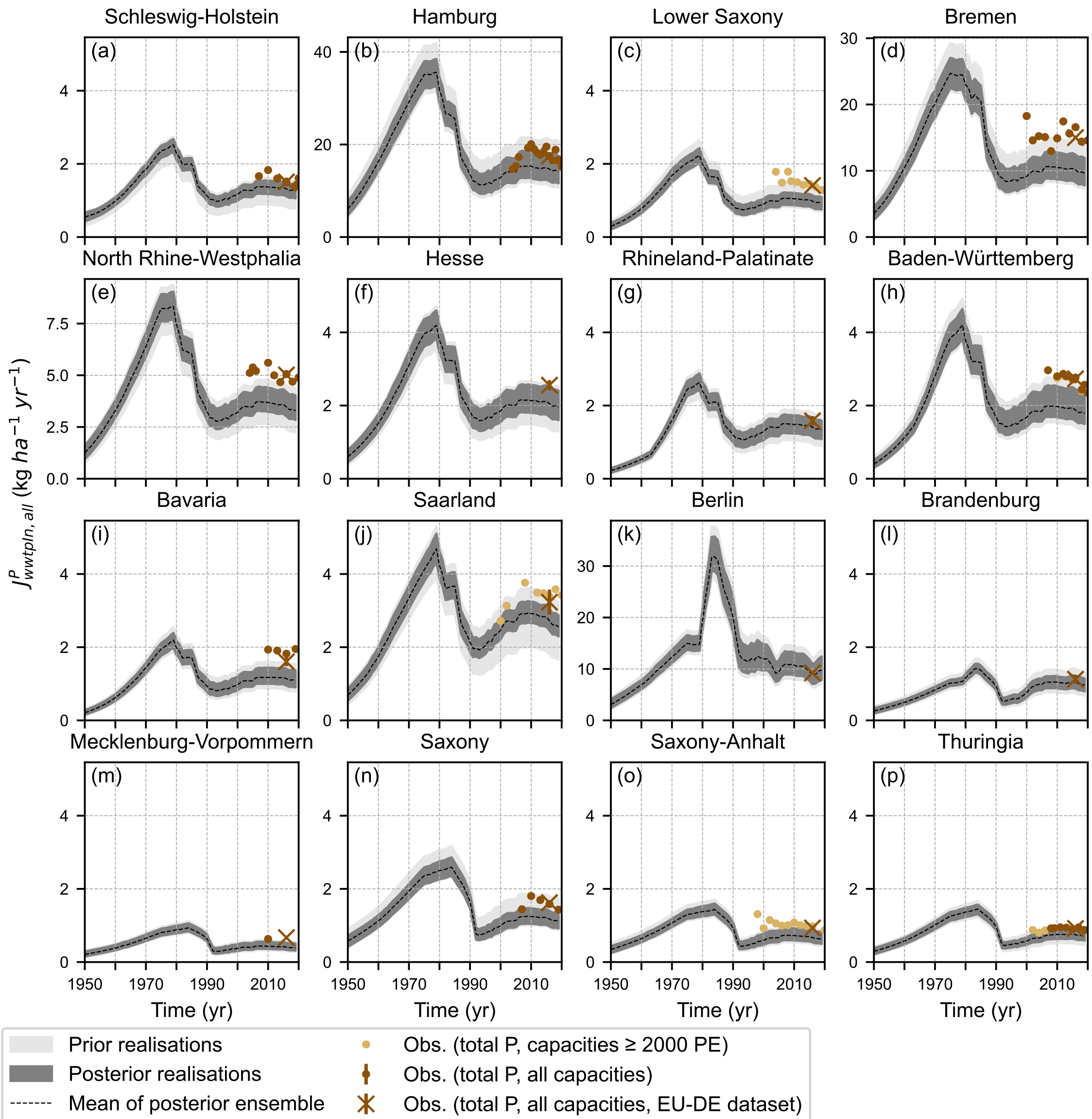


Figure S37. WWTP incoming P loads $J^P_{wwtpIn,all}$, including both the urban and rural components, from the prior and posterior model realisations (grey shaded areas and dashed lines), and observations (coloured circles and crosses) at NUTS-1 level for the period 1950–2019. The shaded areas delineate the minimum and maximum values of the 100,000 prior realisations (light grey) and the 100 posterior realisations (dark grey). Observational values are shown for both the ensemble of WWTPs having a design capacity higher than 2000 PE (light green and brown) and for all WWTPs (dark green and brown). Some observations are reported with error bars that depict the upper and lower bound estimates while the markers (circles/crosses) represent the average value. The crosses shows the data from the combined EU/DE dataset that were aggregated at NUTS-1 level. The scale on the y-axis is the same for all NUTS-1 regions but the city states (Hamburg, Bremen, and Berlin) for which the loads are much higher compared to the other NUTS-1 regions. Data source: observational data come from the statistical offices of Germany and the federal states and from the authorities of the federal states (circles), and the EU Waterbase dataset EEA (2023) and Büttner (2020) (crosses). The uncertainty intervals on the observations were processed in this study. Details on observational data sources and processing are in Sect. S8–S9.

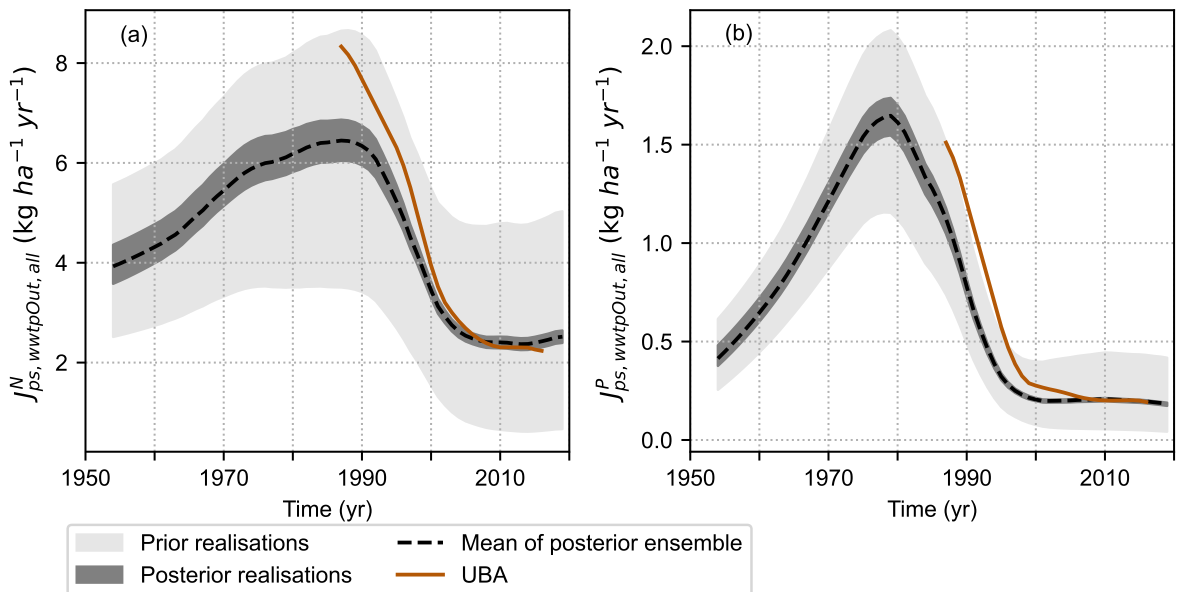


Figure S38. WWTP outgoing N and P loads ($J^{N}_{ps,wwtpOut,all}$ and $J^{P}_{ps,wwtpOut,all}$, respectively), including both the urban and rural components, from our prior and posterior model realisations (grey shaded areas and dashed black lines) and from the German Environmental Agency (UBA, 2020) (solid dark red line) at NUTS-0 level for the period 1950–2019. All fluxes are shown as 5-year backward moving averages (the value in 1954 corresponds to the average of the values ib 1950–1954). The shaded areas delineate the minimum and maximum values of the 100,000 prior realisations (light grey) and the 100 posterior realisations (dark grey).

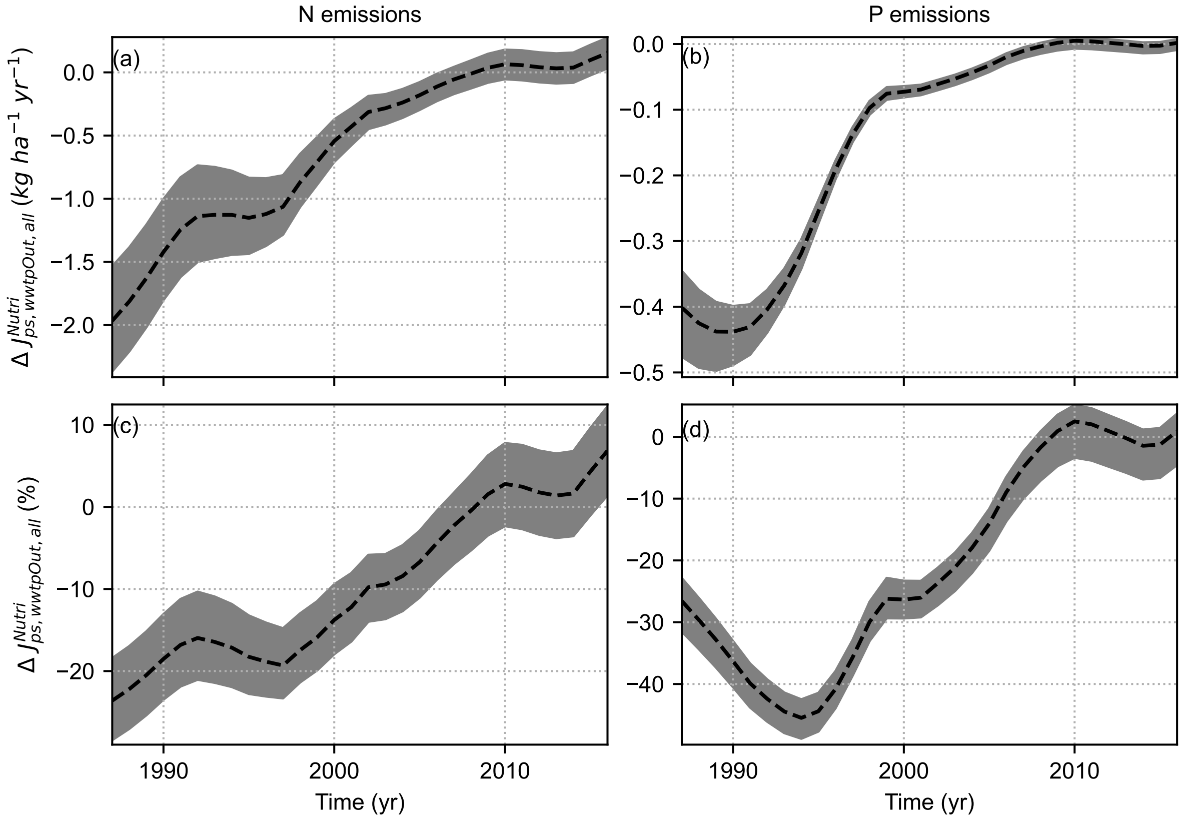


Figure S39. Difference in WWTP outgoing N ($Nutri = N$) and P ($Nutri = P$) P loads (denoted as $\Delta J^{Nutri}_{ps,wwtpOut,all}$) between thr 5-year backward moving average estimates from our posterior model realisations and from the German Environmental Agency (UBA, 2020) at NUTS-0 level for the period 1987–2016 for which both estimates are available (estimates are shown in Fig. S38). The shaded grey areas delineate the minimum and maximum values across the 100 posterior realisations and the black dashed lines represent the difference using the mean of the posterior realisations. A negative value indicates that our realisations are lower then the UBA value.

Notes: In panels (a-b), the differences are expressed in $kg\ ha^{-1}\ yr^{-1}$ and are calculated as follows:

$\Delta J^{Nutri}_{ps,wwtpOut,all} = J^{Nutri}_{ps,wwtpOut,all} - J^{Nutri}_{ps,wwtpOut,all,UBA}$

where $J^{Nutri}_{ps,wwtpOut,all,UBA}$ is the WWTP outgoing N ($Nutri = N$) and P ($Nutri = P$) emissions from the German Environmental Agency, and $J^{Nutri}_{ps,wwtpOut,all}$ is the WWTP outgoing N and P emissions from our posterior ensemble.

In panels (c-d) the differences are expressed in percentages and are calculated as follows:

$\Delta J^{Nutri}_{ps,wwtpOut,all} = \frac{J^{Nutri}_{ps,wwtpOut,all} - J^{Nutri}_{ps,wwtpOut,all,UBA}}{J^{Nutri}_{ps,wwtpOut,all,UBA}} 100$

Table S79. First and last years of the period with observations (calibration period) and the period without observations of WWTP outgoing total N load and WWTP outgoing total P load (from the NUTS-1 level dataset of Sect. S8.1.2) to estimate the model parameters for each NUTS-1 region.

NUTS-1 region	$J^N_{ps,wwtpOut,all}$				$J^P_{ps,wwtpOut,all}$			
	period without obs.		period with obs.		period without obs.		period with obs.	
	first year	last year	first year	last year	first year	last year	first year	last year
Schleswig-Holstein	1950	1990	1991	2019	1950	1986	1987	2019
Hamburg	1950	2003	2004	2019	1950	2003	2004	2019
Lower Saxony	1950	2000	2001	2019	1950	2000	2001	2019
Bremen	1950	1997	1998	2019	1950	1997	1998	2019
North Rhine-Westphalia	1950	1990	1991	2018	1950	1990	1991	2019
Hesse	1950	1995	1996	2019	1950	1995	1996	2019
Rhineland-Palatinate	1950	2000	2001	2018	1950	2000	2001	2019
Baden-Württemberg	1950	2009	2010	2018	1950	2006	2007	2019
Bavaria	1950	1999	2000	2019	1950	2006	2007	2019
Saarland	1950	2006	2007	2019	1950	2006	2007	2019
Berlin	1950	2006	2007	2019	1950	2006	2007	2019
Brandenburg	1950	1990	1991	2019	1950	1990	1991	2019
Mecklenburg-Vorpommern	1950	2006	2007	2019	1950	2006	2007	2019
Saxony	1950	2006	2007	2019	1950	2006	2007	2019
Saxony-Anhalt	1950	2006	2007	2019	1950	2006	2007	2019
Thuringia	1950	2007	2008	2019	1950	2006	2007	2019

Notations: $J^N_{ps,wwtpOut,all}$ is the WWTP outgoing N load; $J^P_{ps,wwtpOut,all}$ is the WWTP outgoing P load.

Table S80. Width of the uncertainty interval (UI) of the behavioural simulation ensemble for WWTP outgoing N and P load, calculated as the difference between the maximum and minimum values across the 100 behavioural realisations, averaged over the period with or without observations for parameter estimation for each NUTS-1 region.

NUTS-1 region	$J^N_{ps,wwtpOut,all}$				$J^P_{ps,wwtpOut,all}$			
	period without obs.		period with obs.		period without obs.		period with obs.	
	UI (%)	UI ($kg\,ha^{-1}\,yr^{-1}$)	UI (%)	UI ($kg\,ha^{-1}\,yr^{-1}$)	UI (%)	UI ($kg\,ha^{-1}\,yr^{-1}$)	UI (%)	UI ($kg\,ha^{-1}\,yr^{-1}$)
Schleswig-Holstein	37.233	1.66	43.58	0.858	24.322	0.183	65.818	0.099
Hamburg	51.651	28.713	6.711	1.869	41.172	4.31	6.706	0.093
Lower Saxony	58.161	1.676	13.333	0.112	48.856	0.261	9.31	0.009
Bremen	48.668	23.415	8.417	1.374	36.564	3.513	8.749	0.055
North Rhine-Westphalia	30.334	4.807	31.514	2.195	36.123	0.985	34.981	0.145
Hesse	27.307	1.969	22.75	0.918	35.638	0.415	25.718	0.092
Rhineland-Palatinate	40.16	1.509	17.124	0.322	33.757	0.226	12.44	0.029
Baden-Württemberg	48.482	2.774	8.112	0.323	34.38	0.343	9.37	0.024
Bavaria	38.049	1.35	11.355	0.325	37.908	0.209	8.963	0.021
Saarland	42.873	3.172	8.163	0.389	29.852	0.393	5.631	0.035
Berlin	49.158	15.128	13.665	1.565	39.505	2.015	10.646	0.039
Brandenburg	40.759	0.904	27.438	0.348	28.146	0.111	24.078	0.018
Mecklenburg-Vorpommern	47.2	0.866	6.39	0.025	41.483	0.143	10.311	0.004
Saxony	47.25	2.346	7.827	0.176	36.907	0.348	6.008	0.011
Saxony-Anhalt	50.274	1.454	11.156	0.069	40.098	0.209	10.19	0.007
Thuringia	48.894	1.168	14.728	0.115	30.797	0.154	11.83	0.013

The periods with and without observations are reported in Table S79. The width of the uncertainty interval CI is expressed % when it is normalised by the mean value over the behavioural realisations and then averaged over time (period with or without observations), or expressed in $kg\ ha^{-1}\ yr^{-1}$ when it is directly averaged over time without any normalisation.

Notations: $J^N_{ps,wwtpOut,all}$ is the WWTP outgoing N load; $J^P_{ps,wwtpOut,all}$ is the WWTP outgoing P load.

Table S81. Width of the uncertainty interval (UI) of the behavioural simulation ensemble for total N and P point sources, calculated as the difference between the maximum and minimum values across the 100 behavioural realisations, averaged over the period with or without observations for parameter estimation for each NUTS-1 region.

NUTS-1 region	$J^N_{ps,tot,all}$				$J^P_{ps,tot,all}$			
	period without obs.		period with obs.		period without obs.		period with obs.	
	UI (%)	UI ($kg\,ha^{-1}\,yr^{-1}$)	UI (%)	UI ($kg\,ha^{-1}\,yr^{-1}$)	UI (%)	UI ($kg\,ha^{-1}\,yr^{-1}$)	UI (%)	UI ($kg\,ha^{-1}\,yr^{-1}$)
Schleswig-Holstein	36.191	1.795	43.154	0.858	23.102	0.197	64.685	0.1
Hamburg	48.386	31.592	6.711	1.869	36.829	4.637	6.706	0.093
Lower Saxony	55.179	1.855	13.306	0.112	45.438	0.285	9.299	0.009
Bremen	46.35	27.315	8.417	1.374	33.196	4.056	8.749	0.055
North Rhine-Westphalia	28.895	5.186	31.498	2.196	33.434	1.031	34.921	0.145
Hesse	24.816	2.317	22.613	0.919	30.306	0.477	25.457	0.093
Rhineland-Palatinate	34.5	2.168	16.993	0.322	28.258	0.31	12.365	0.029
Baden-Württemberg	44.048	3.15	8.109	0.323	30.506	0.405	9.365	0.024
Bavaria	36.765	1.8	11.27	0.326	32.273	0.238	8.875	0.021
Saarland	31.514	4.781	8.062	0.408	19.305	0.57	5.752	0.038
Berlin	49.158	15.128	13.665	1.565	39.505	2.015	10.646	0.039
Brandenburg	40.347	1.039	27.215	0.348	25.928	0.118	24.011	0.018
Mecklenburg-Vorpommern	45.324	0.979	6.282	0.025	39.509	0.158	10.221	0.004
Saxony	42.191	3.512	7.025	0.18	30.136	0.474	6.92	0.016
Saxony-Anhalt	45.077	1.994	9.499	0.065	34.656	0.274	10.239	0.008
Thuringia	41.783	2.345	16.407	0.312	27.233	0.292	18.093	0.046

The periods with and without observations are reported in Table S79. The width of the uncertainty interval CI is expressed % when it is normalised by the mean value over the behavioural realisations and then averaged over time (period with or without observations), or expressed in $kg\ ha^{-1}\ yr^{-1}$ when it is directly averaged over time without any normalisation.

Notations: $J^N_{ps,tot,all}$ is the total N point sources (including both the treated and untreated parts); $J^P_{ps,tot,all}$ is the total P point sources.

S10.2 Fate of N and P gross emissions at NUTS-0 and NUTS-1 level (1950–2019)

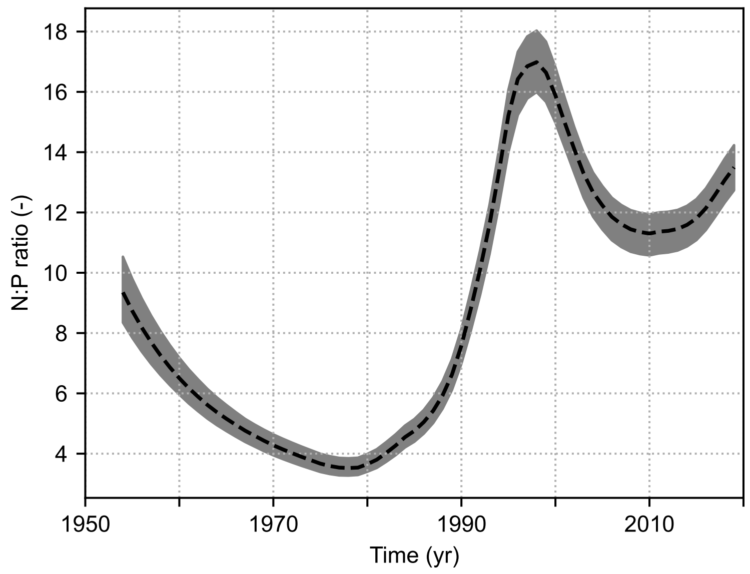


Figure S40. N:P ratio of the total point sources at NUTS-0 level calculated from the 5-year backward moving average of the N and P point sources over the period 1954–2019 (the value in 1954 corresponds to the ratio of the average N and P point sources over the period 1950–1954). The grey shaded delineate the minimum and maximum values of the 100 posterior realisations, while the dashed black line represents the mean of the posterior ensemble. The total point sources include the treated and untreated components.

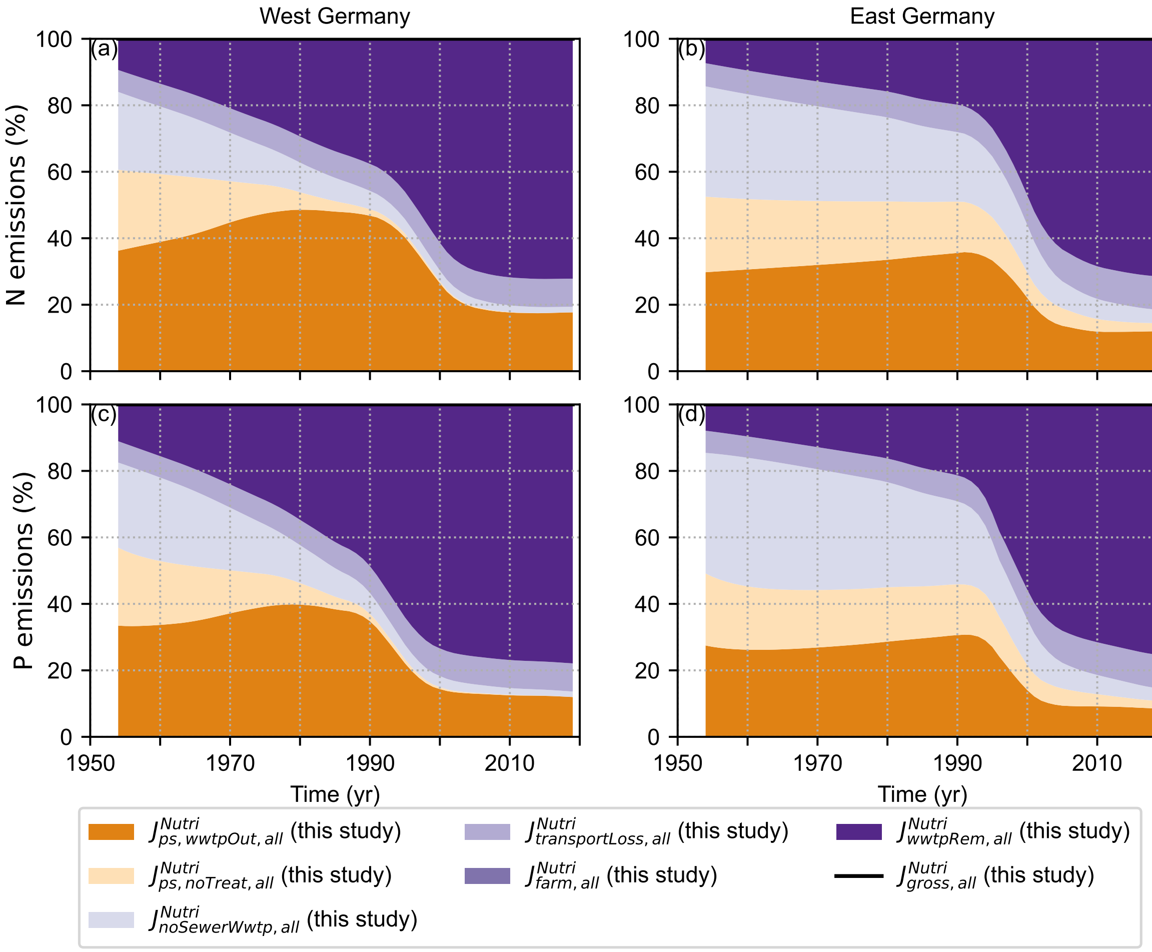


Figure S41. Fate of the N (panels a,b) and P (panels c,d) gross emissions (expressed in % of the gross emissions) averaged over the NUTS-1 regions of West (panels a,c) and East Germany (panels b,d) for the period 1954–2019 from the mean of our posterior realisations. All fluxes are shown as 5-year backward moving averages (the value in 1954 corresponds to the average of the values for 1950–1954). The pathways of the gross emissions are point sources (reddish colours) or other pathways (purplish colours). The data shown include both domestic and industrial emissions, and both urban and rural emissions. Notations: $J^{Nutri}_{gross,all}$ are the N ($Nutri = N$) and P ($Nutri = P$) gross emissions; $J^{Nutri}_{ps,wwtpOut,all}$ are the N and P WWTP outgoing emissions; $J^{Nutri}_{ps,noTreat,all}$ are the N and P emissions collected in the public sewer system that are not treated in WWTPs; $J^{Nutri}_{noSewerWwtp,all}$ are the N and P gross emissions that are not collected in the sewer system nor treated in WWTPs; $J^{Nutri}_{transportLoss,all}$ are the N and P emissions lost during wastewater collection and transport; $J^{Nutri}_{farm,all}$ are the N and P emissions applied to agricultural soils in sewage farms; $J^{Nutri}_{wwtpRem,all}$ are the N and P emissions that are removed during treatment in WWTPs.

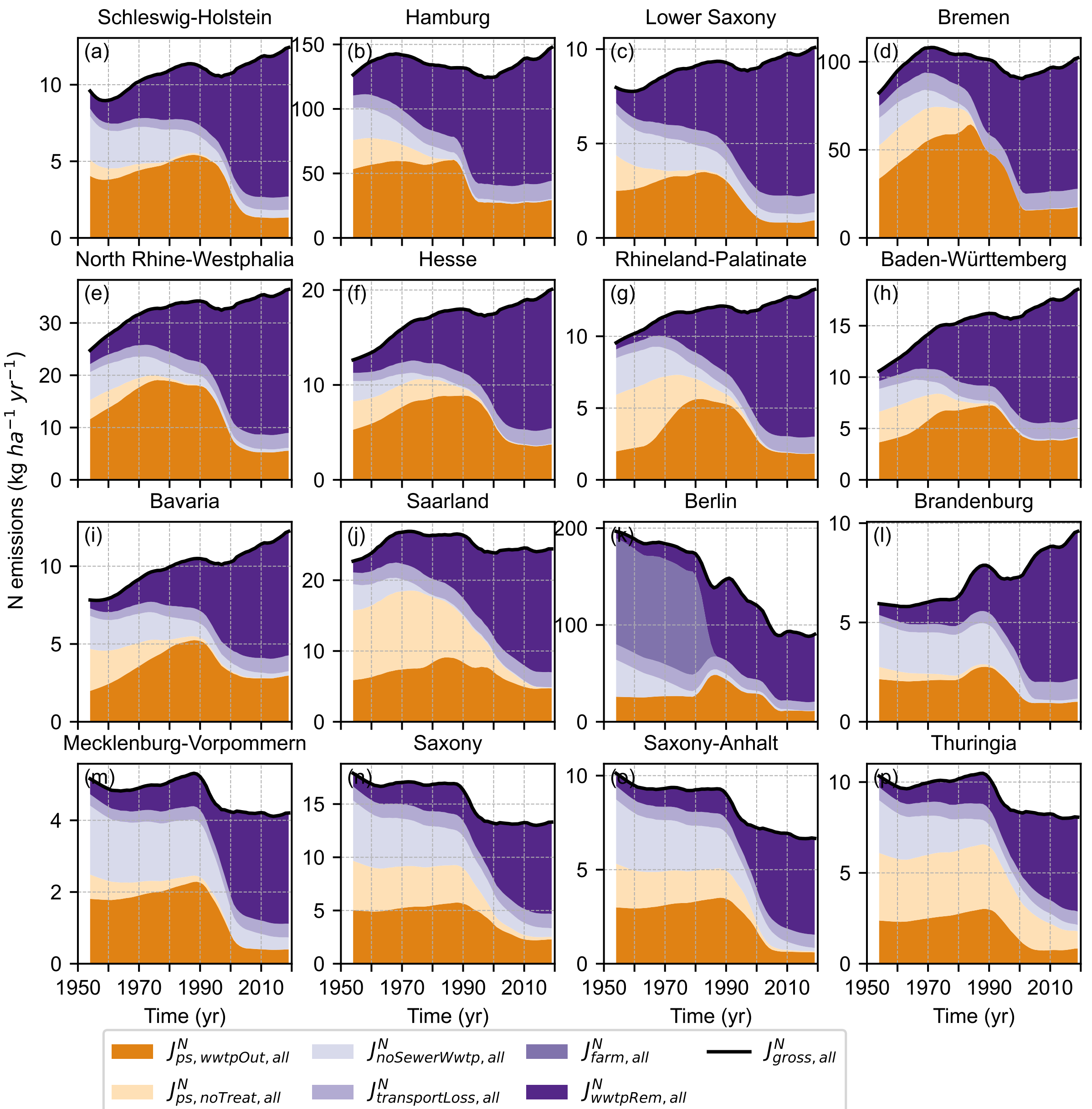


Figure S42. Fate of the N gross emissions at NUTS-1 level for the period 1954–2019 from the mean of our posterior realisations. All fluxes are shown are 5-year backward moving averages (the value in 1954 corresponds to the average of the values for 1950–1954). The pathways of gross emissions are point sources (reddish colours) or other pathways (purplish colours). The data shown include both domestic and industrial emissions, and both urban and rural emissions. See the definition of the notations in the caption of Fig.S41.

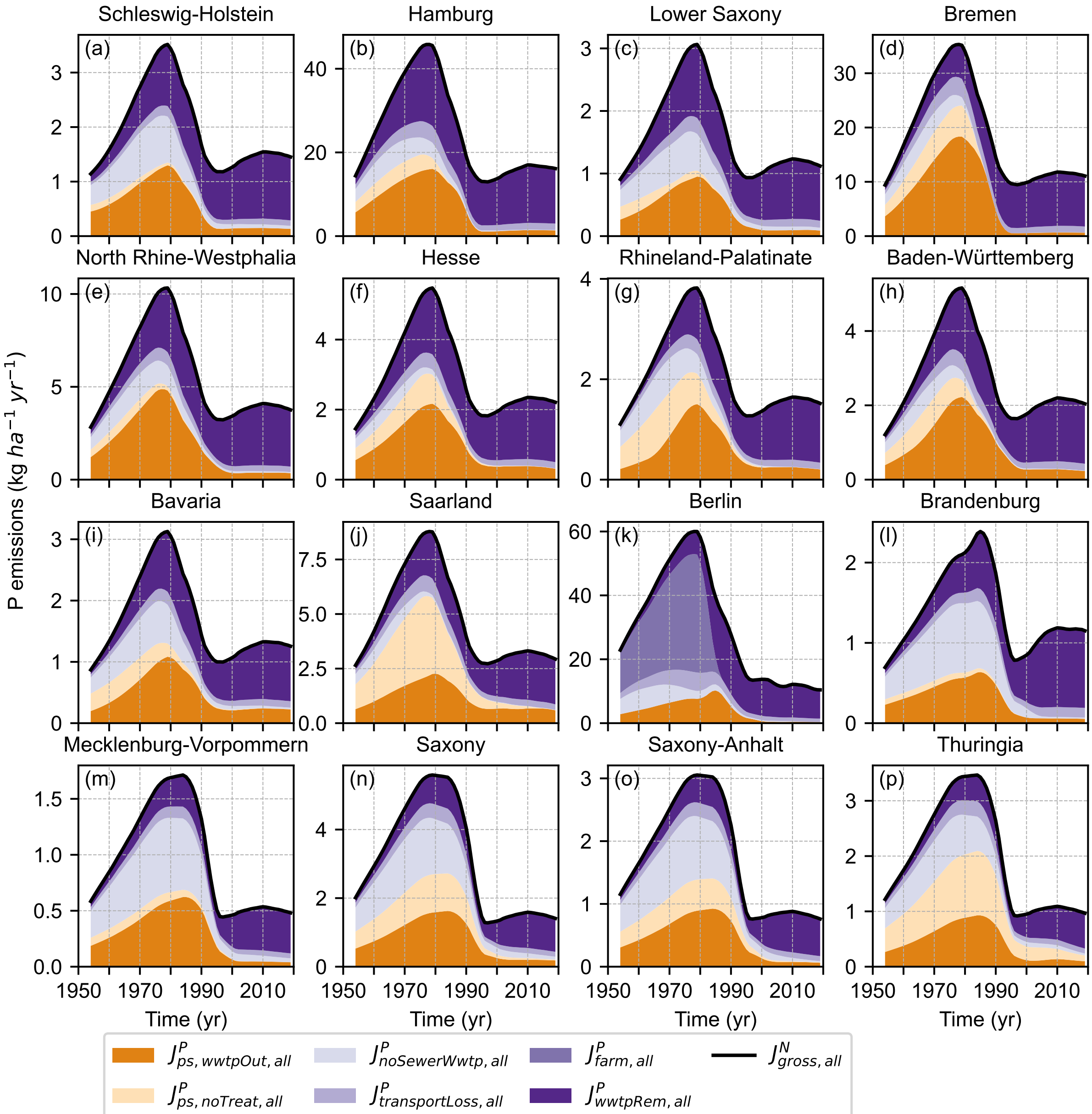


Figure S43. Fate of the P gross emissions at NUTS-1 level for the period 1954–2019 from the mean of our posterior realisations. All fluxes are shown are 5-year backward moving averages (the value in 1954 corresponds to the average of the values for 1950–1954). The pathways of gross emissions are point sources (reddish colours) or other pathways (purplish colours). The data shown include both domestic and industrial emissions, and both urban and rural emissions. See the definition of the notations in the caption of Fig. S41.

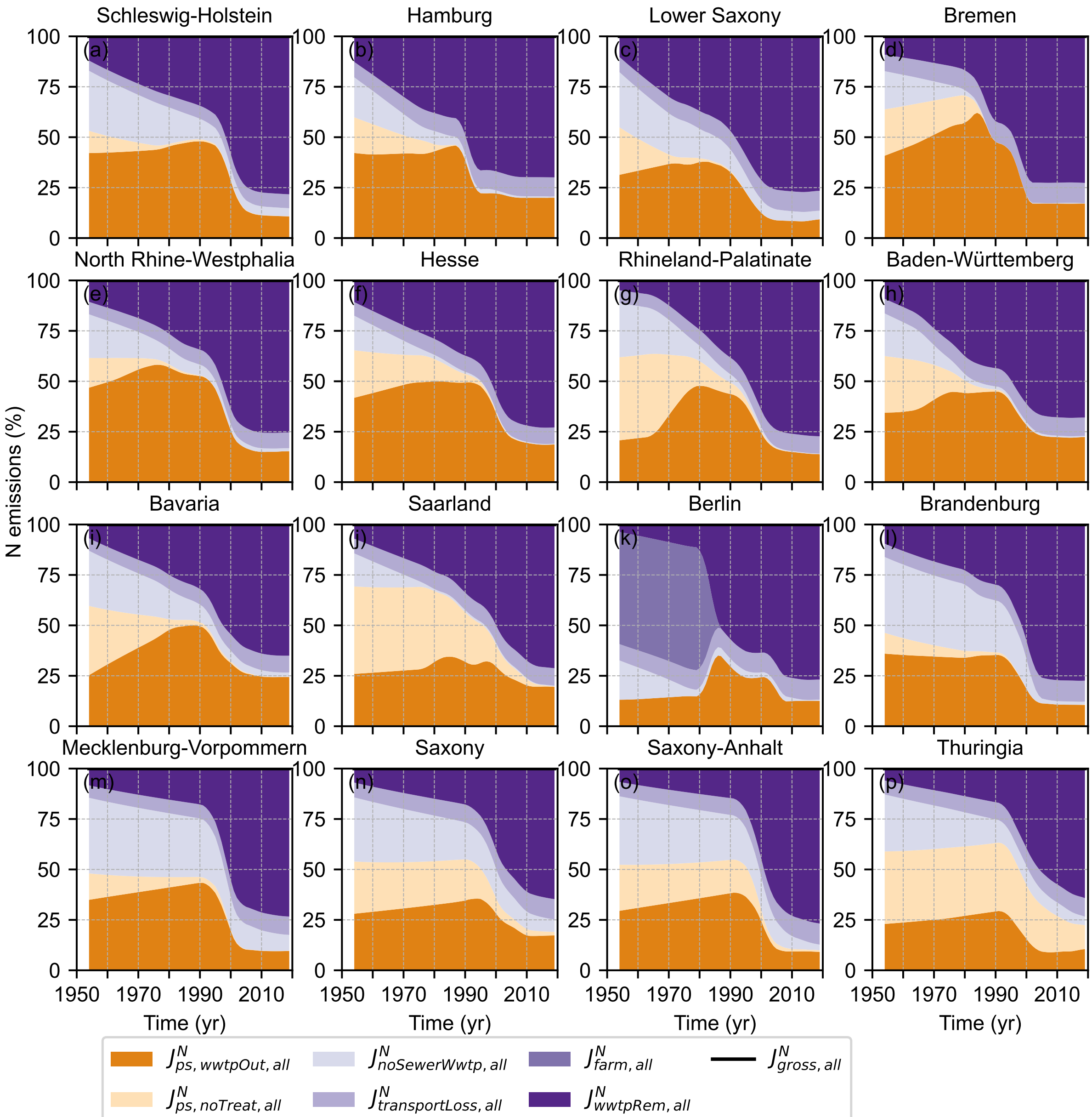


Figure S44. Fate of the N gross emissions (expressed in % of N gross emissions) at NUTS-1 level for the period 1954–2019 from the mean of our posterior realisations. All fluxes are shown are 5-year backward moving averages (the value in 1954 corresponds to the average of the values for 1950–1954). The fates of gross emissions are point sources (reddish colours) or other pathways (purplish colours). The data shown include both domestic and industrial emissions, and both urban and rural emissions. See the definition of the notations in the caption of Fig. S41.

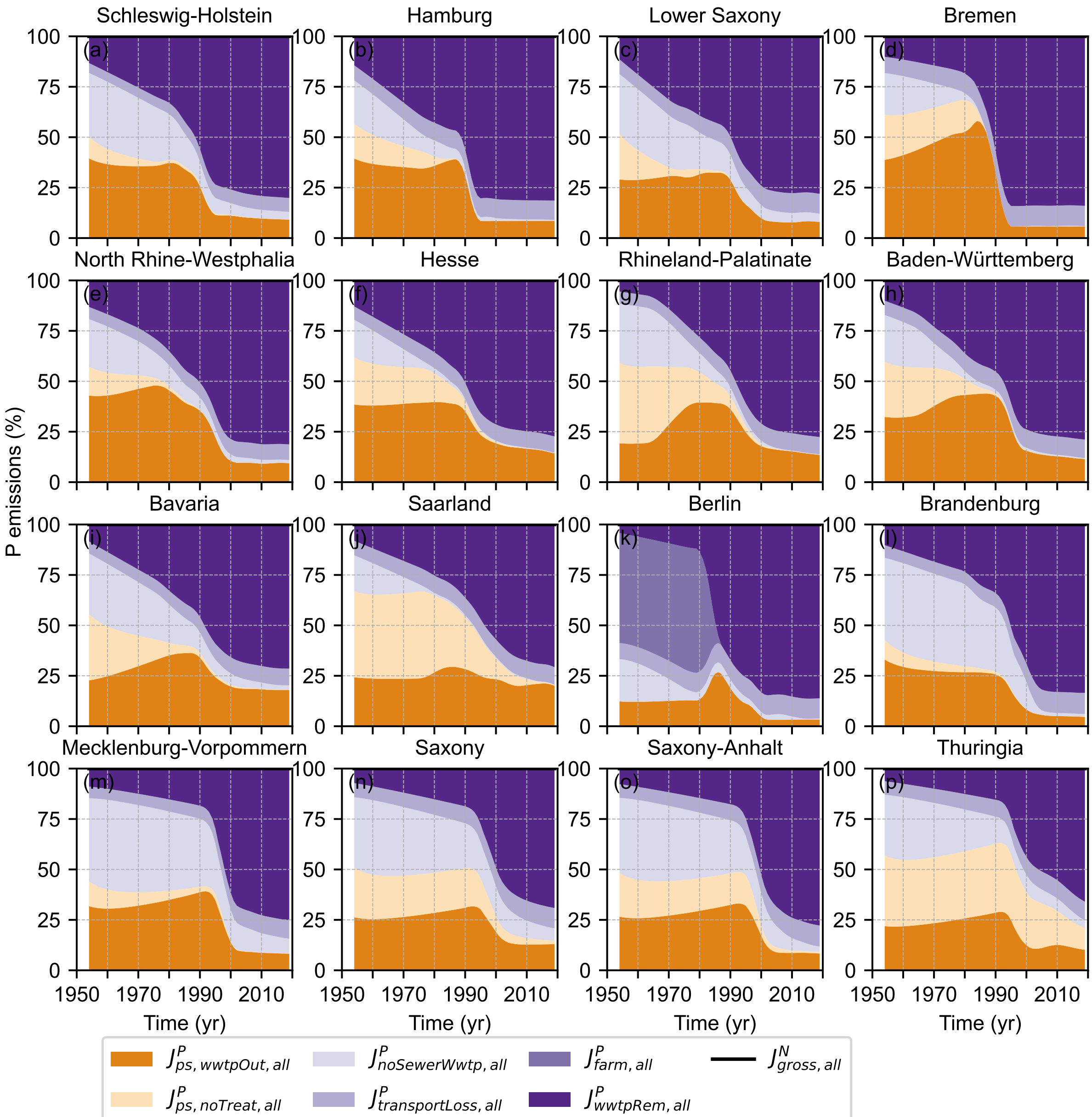


Figure S45. Fate of the P gross emissions (expressed in % of P gross emissions) at NUTS-1 level for the period 1954–2019 from the mean of our posterior realisations. All fluxes are shown are 5-year backward moving averages (the value in 1954 corresponds to the average of the values for 1950–1954). The fates of gross emissions are point sources (reddish colours) or other pathways (purplish colours). The data shown include both domestic and industrial emissions, and both urban and rural emissions. See the definition of the notations in the caption of Fig. S41.

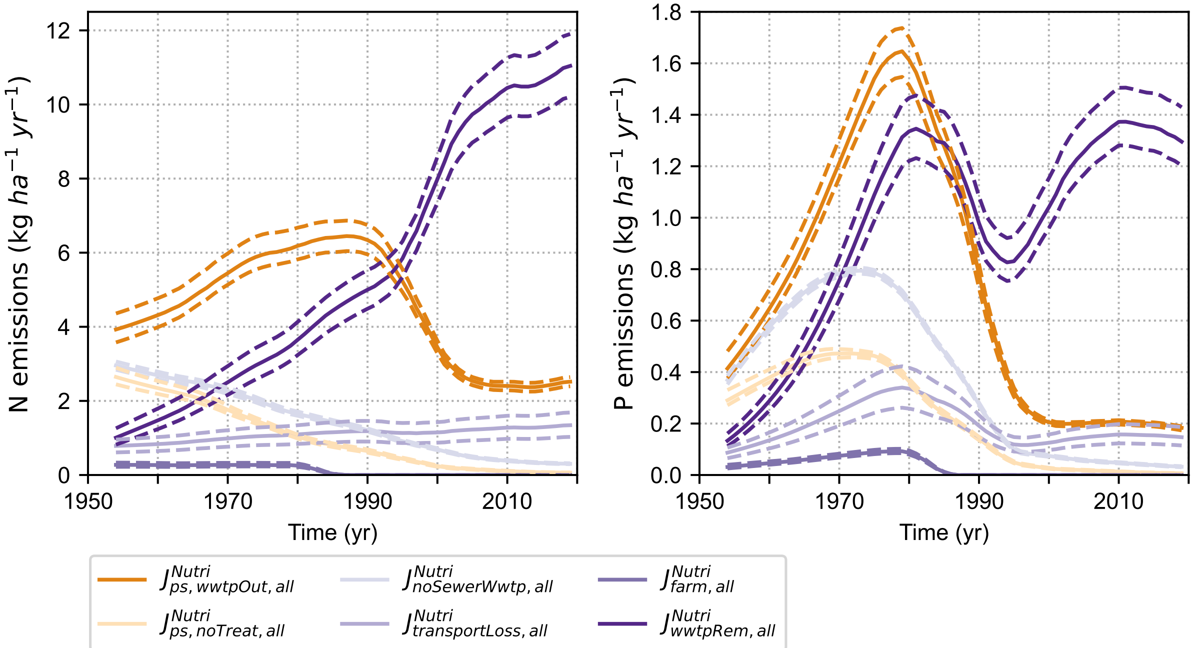


Figure S46. Fate of the N and P gross emissions at NUTS-0 level for the period 1954–2019 from our posterior realisation ensemble. The mean values across the 100 posterior realisations are shown as solid lines, and the minimum and maximum values as dashed lines. All fluxes are shown are 5-year backward moving averages (the value in 1954 corresponds to the average of the values for 1950–1954). Panel (a) represents N emissions and panel (b) P emissions. The pathways of gross emissions are point sources (reddish colours) or other pathways (purplish colours). The data shown include both domestic and industrial emissions, and both urban and rural emissions. We see that the uncertainty intervals of the different N and P emission pathways have limited overlap. This means that, on average at NUTS-0 level, the uncertainty has a limited impact on the relative contributions of the different N and P emission pathways. See the definition of the notations in the caption of Fig. S41.

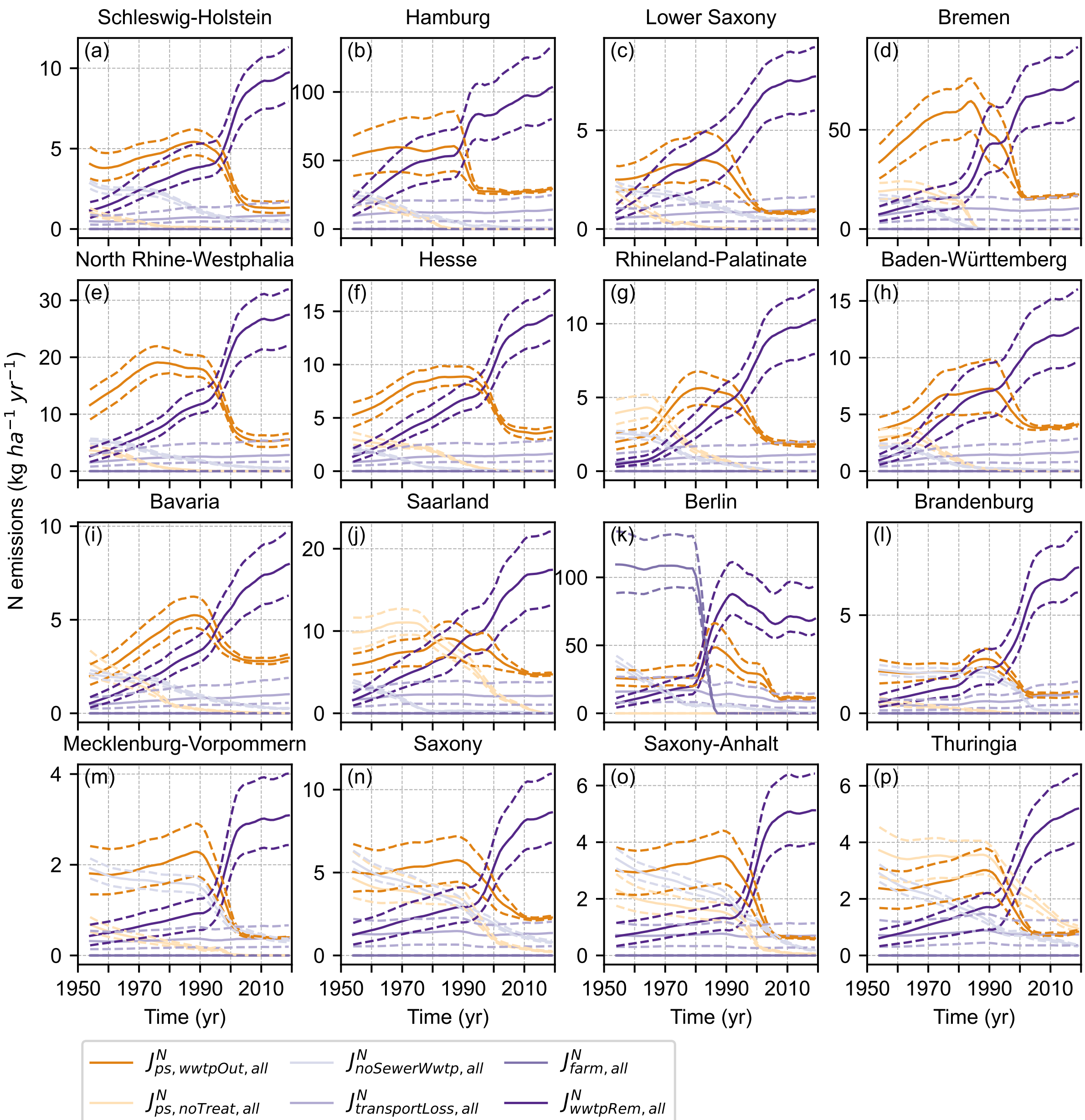


Figure S47. Fate of the N gross emissions at NUTS-1 level for the period 1954–2019 from our posterior realisation ensemble. The mean values across the 100 posterior realisations are shown as solid lines, and the minimum and maximum values as dashed lines. All fluxes are shown are 5-year backward moving averages (the value in 1954 corresponds to the average of the values for 1950–1954). The pathways of gross emissions are point sources (reddish colours) or other pathways (purplish colours). The data shown include both domestic and industrial emissions, and both urban and rural emissions. We see that, in some cases, the uncertainty intervals are wide and are overlapping. This means that the uncertainty overlap, impact to some degree the relative importance of the different N emission pathways. See the definition of the notations in the caption of Fig.S41.

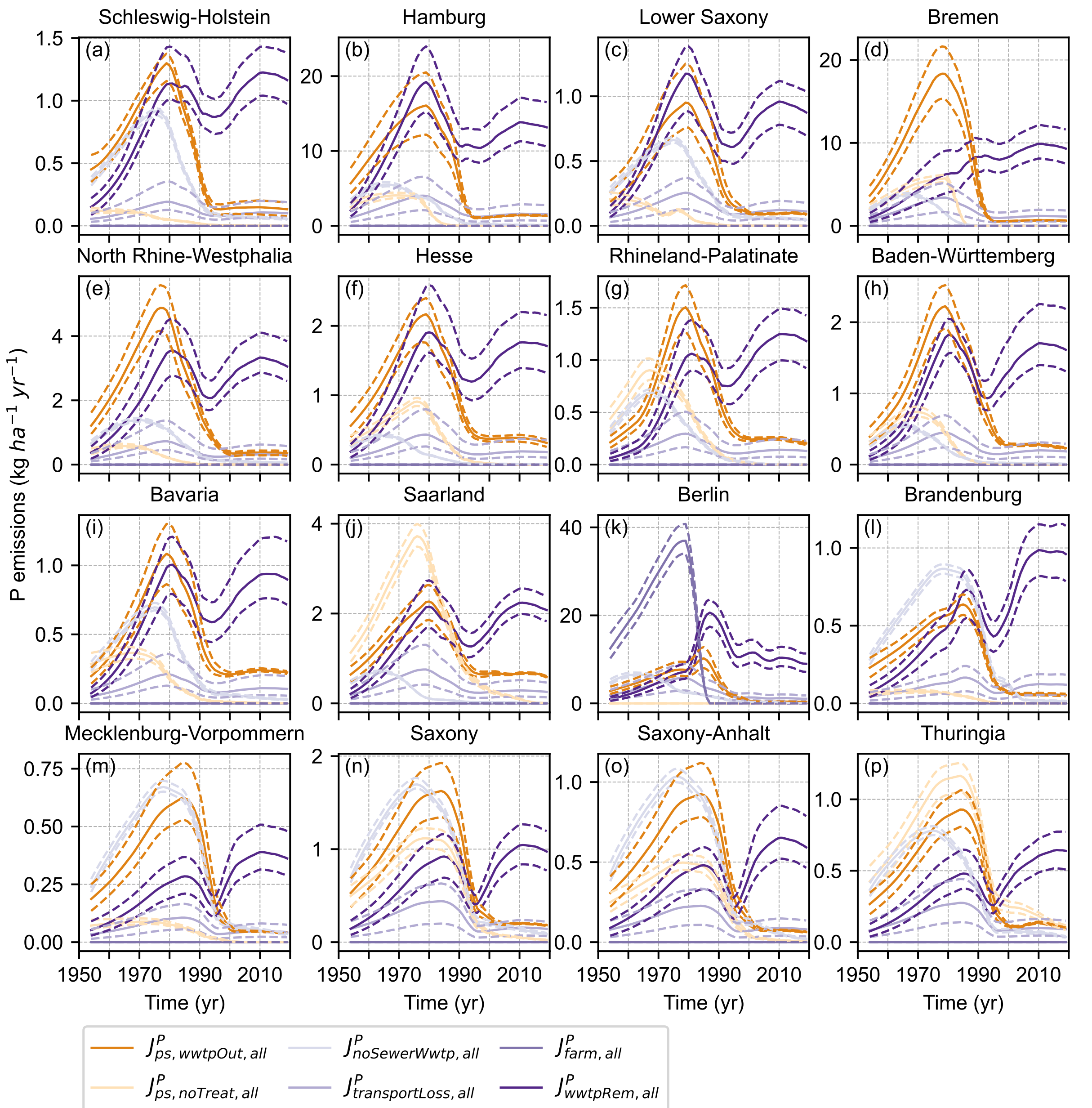


Figure S48. Fate of the P gross emissions at NUTS-1 level for the period 1954–2019 from our posterior realisation ensemble. The mean values across the 100 posterior realisations are shown as solid lines, and the minimum and maximum values as dashed lines. All fluxes are shown are 5-year backward moving averages (the value in 1954 corresponds to the average of the values for 1950–1954). The pathways of gross emissions are point sources (reddish colours) or other pathways (purplish colours). The data shown include both domestic and industrial emissions, and both urban and rural emissions. We see that, in some cases, the uncertainty intervals are wide and are overlapping. This means that the uncertainty overlap, impact to some degree the relative importance of the different P emission pathways. See the definition of the notations in the caption of Fig.S41.

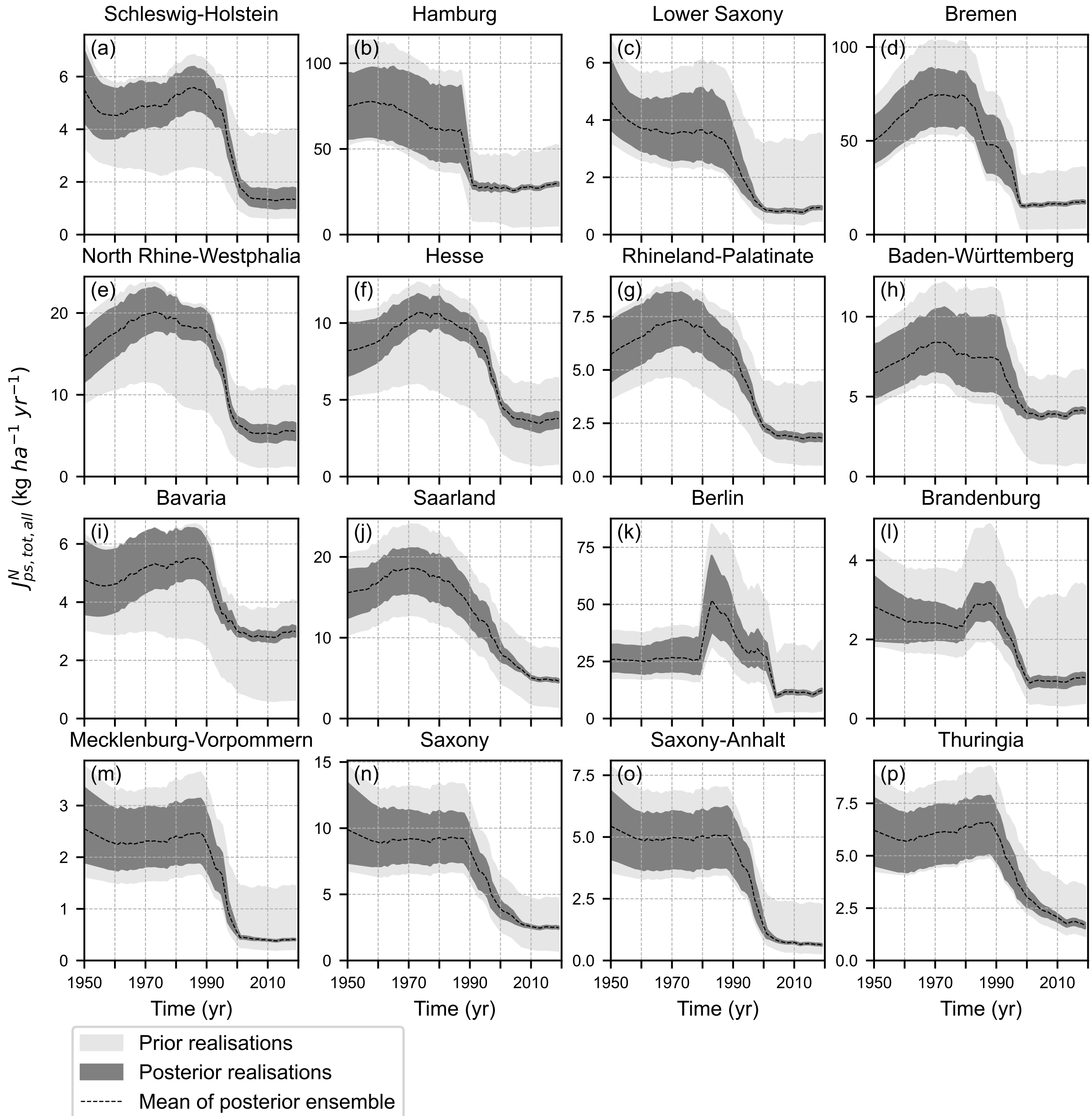


Figure S49. N point source load $J^N_{ps,tot,all}$, including both the urban and rural components, the domestic and industrial/commercial components, and the treated and not treated components ($J^N_{ps,wwtpOut,all}$ and $J^N_{ps,NoTreat,all}$, respectively), from the prior and posterior model realisations at NUTS-1 level for the period 1950–2019. The shaded areas delineate the minimum and maximum values of the 100,000 prior realisations (light grey) and the 100 posterior realisations (dark grey).

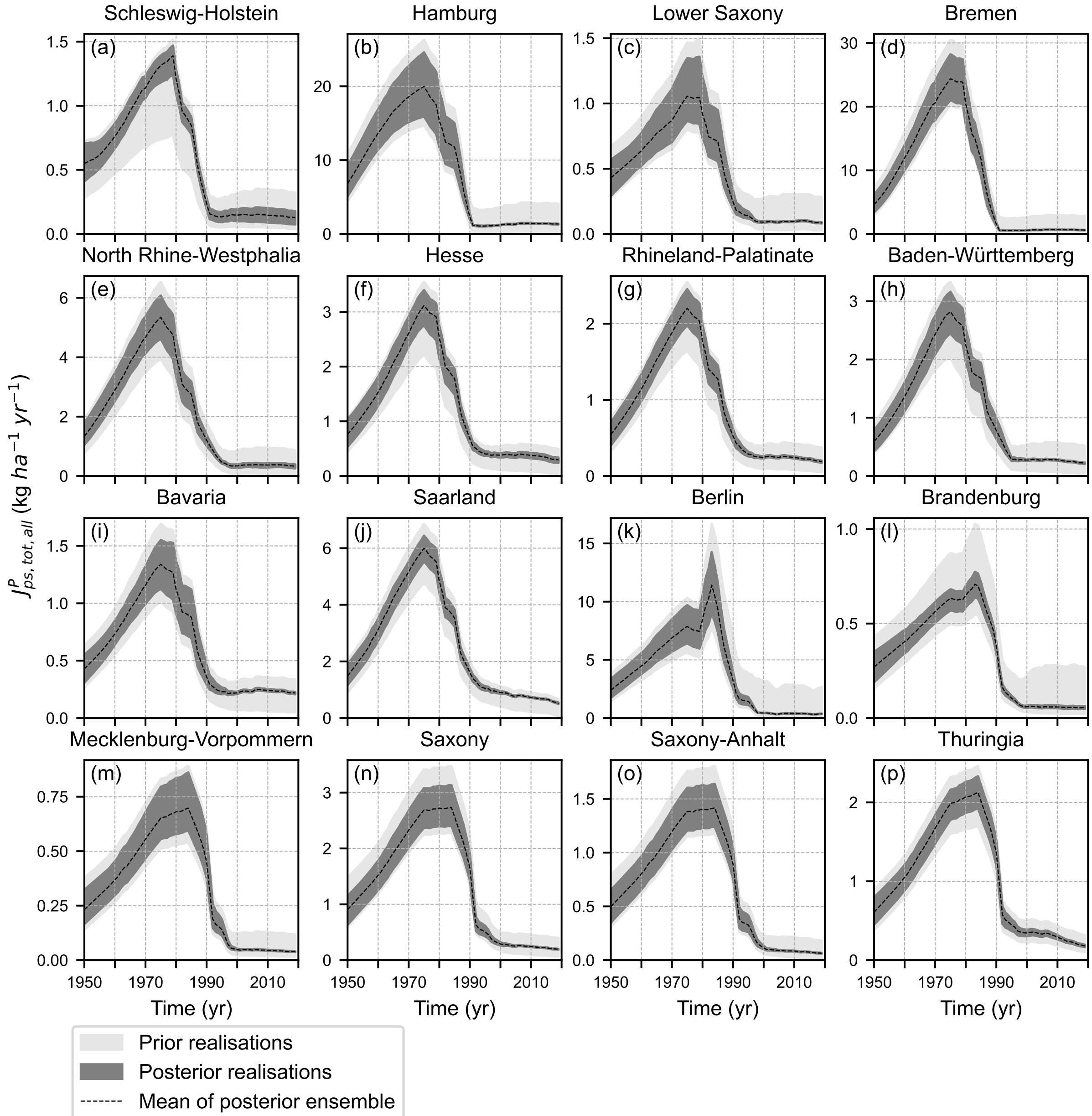


Figure S50. P point source load $J^P_{ps,tot,all}$, including both the urban and rural components, the domestic and industrial/commercial components, and the treated and not treated components ($J^P_{ps,wwtpOut,all}$ and $J^P_{ps,NoTreat,all}$, respectively), from the prior and posterior model realisations at NUTS-1 level for the period 1950–2019. The shaded areas delineate the minimum and maximum values of the 100,000 prior realisations (light grey) and the 100 posterior realisations (dark grey).

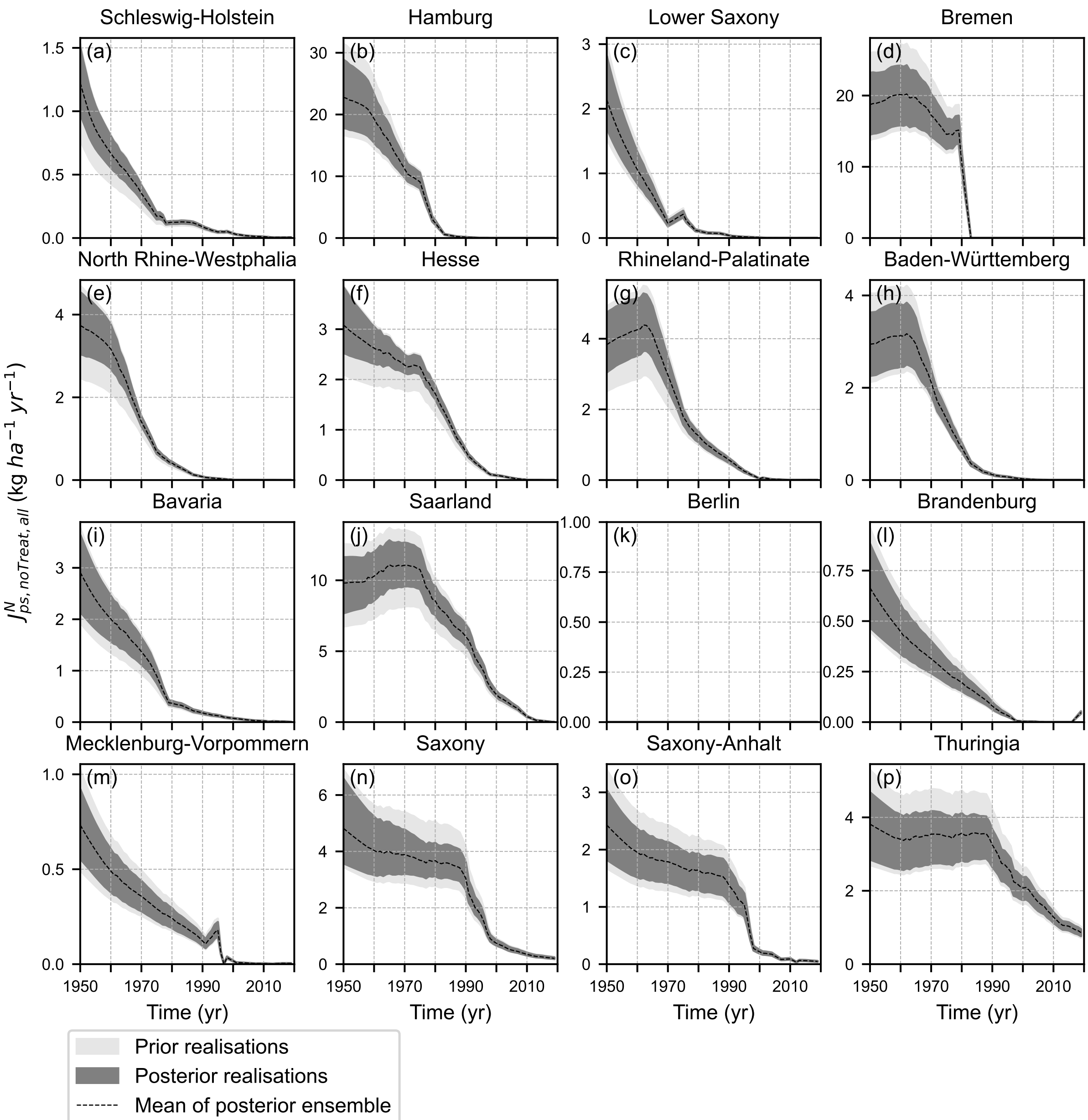


Figure S51. N point source emissions that are collected in the public sewer system but that are not treated in WWTPs $J^N_{ps,NoTreat,all}$, including both the urban and rural components, and the domestic and industrial/commercial components, from the prior and posterior model realisations at NUTS-1 level for the period 1950–2019. The shaded areas delineate the minimum and maximum values of the 100,000 prior realisations (light grey) and the 100 posterior realisations (dark grey).

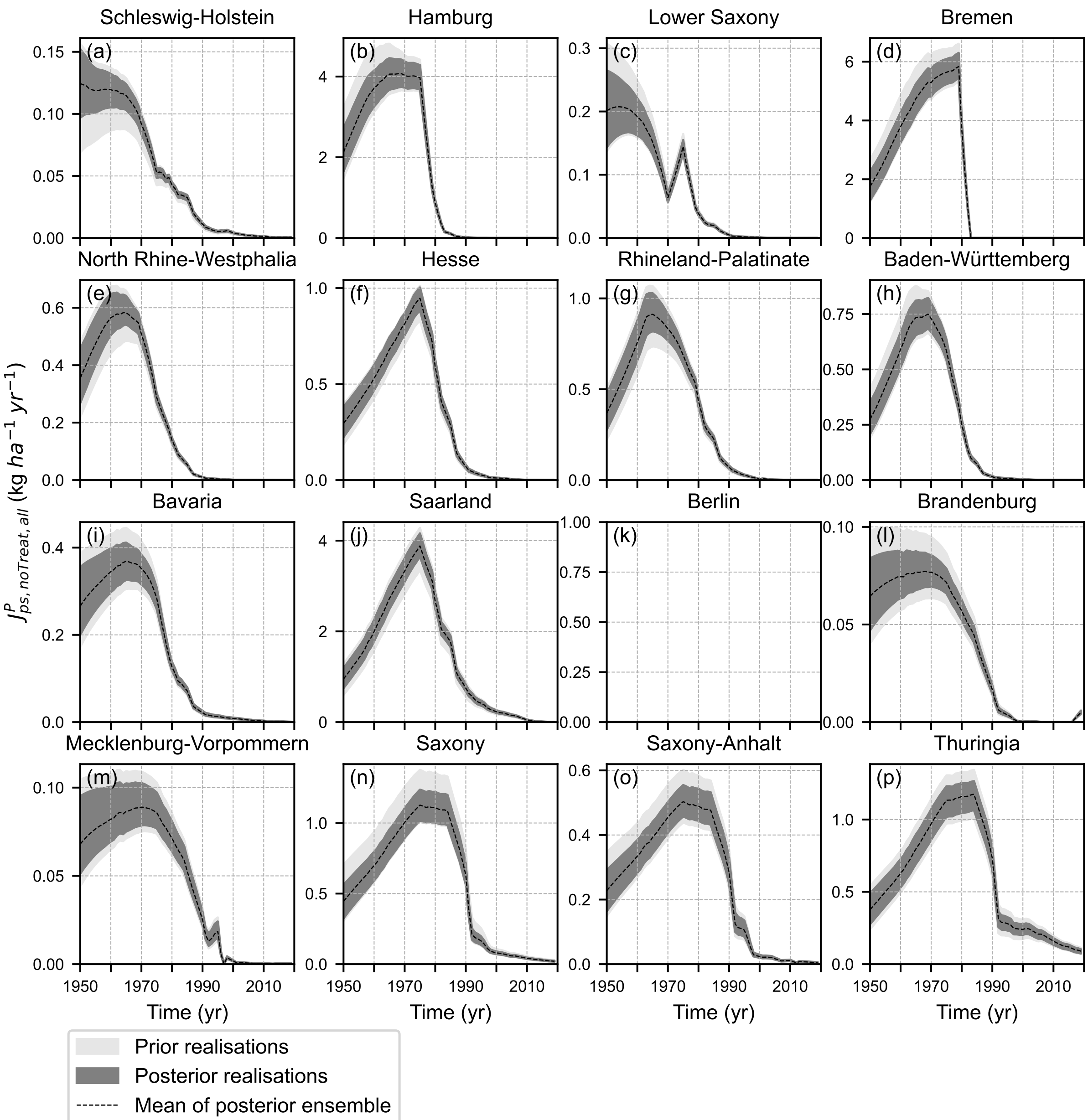


Figure S52. P point source emissions that are collected in the public sewer system but that are not treated in WWTPs $J^P_{ps,NoTreat,all}$, including both the urban and rural components, and the domestic and industrial/commercial components, from the prior and posterior model realisations at NUTS-1 level for the period 1950–2019. The shaded areas delineate the minimum and maximum values of the 100,000 prior realisations (light grey) and the 100 posterior realisations (dark grey).

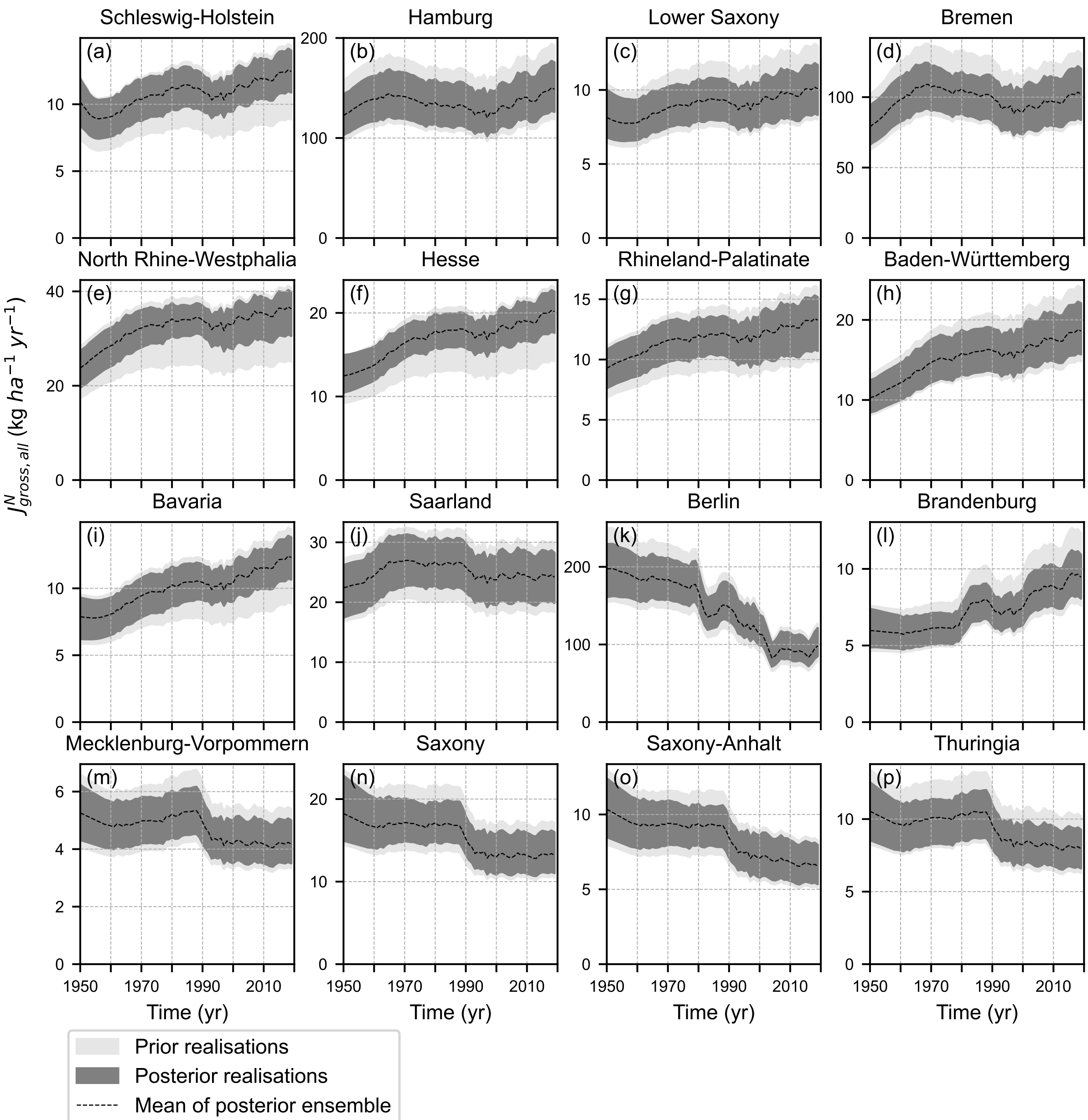


Figure S53. N gross emissions $J^{N}_{gross,all}$, including both the urban and rural components, and the domestic and industrial/commercial components, from the prior and posterior model realisations at NUTS-1 level for the period 1950–2019. The shaded areas delineate the minimum and maximum values of the 100,000 prior realisations (light grey) and the 100 posterior realisations (dark grey).

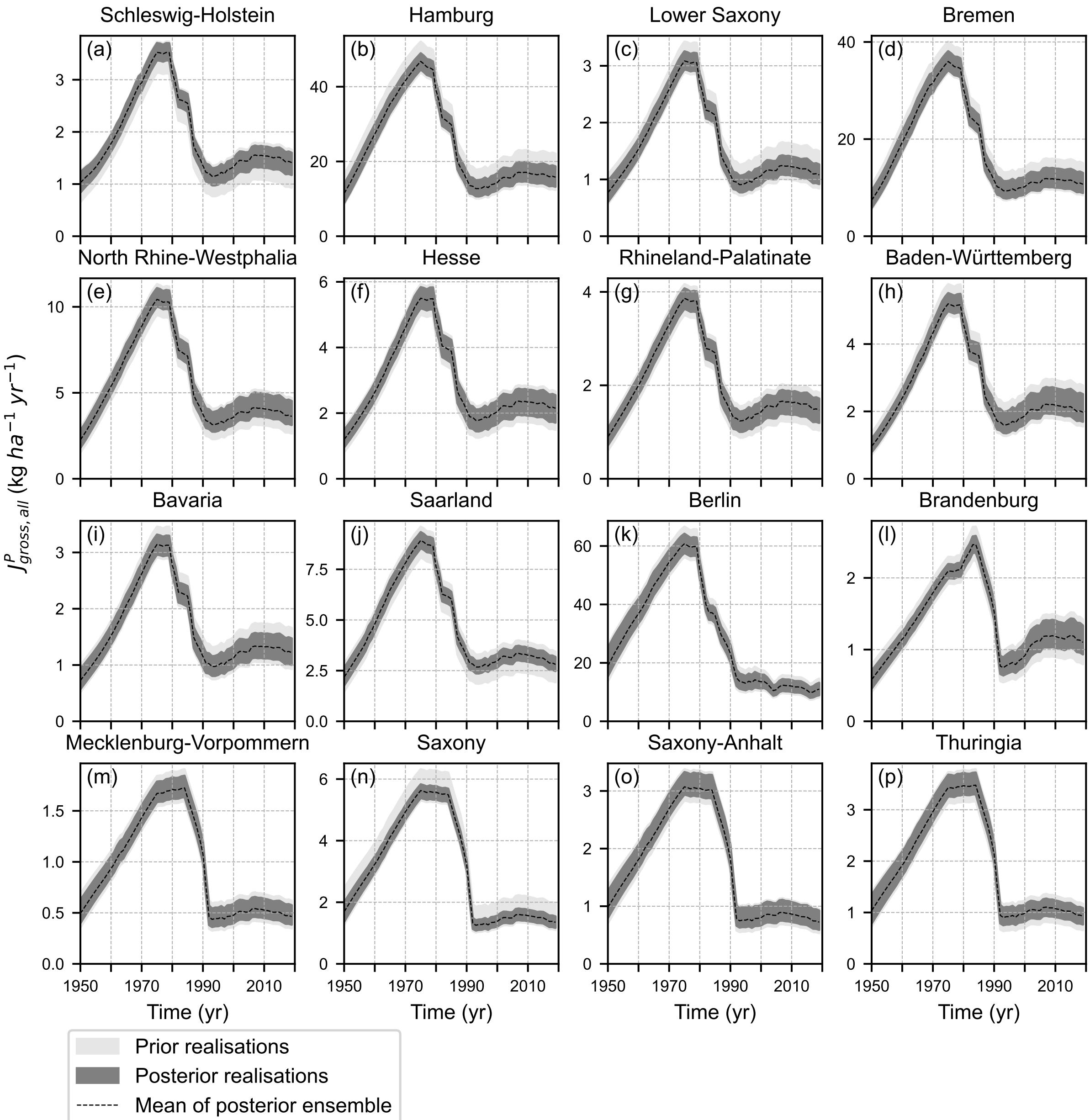


Figure S54. P gross emissions $J^{P}_{gross,all}$, including both the urban and rural components, and the domestic and industrial/commercial components, from the prior and posterior model realisations at NUTS-1 level for the period 1950–2019. The shaded areas delineate the minimum and maximum values of the 100,000 prior realisations (light grey) and the 100 posterior realisations (dark grey).

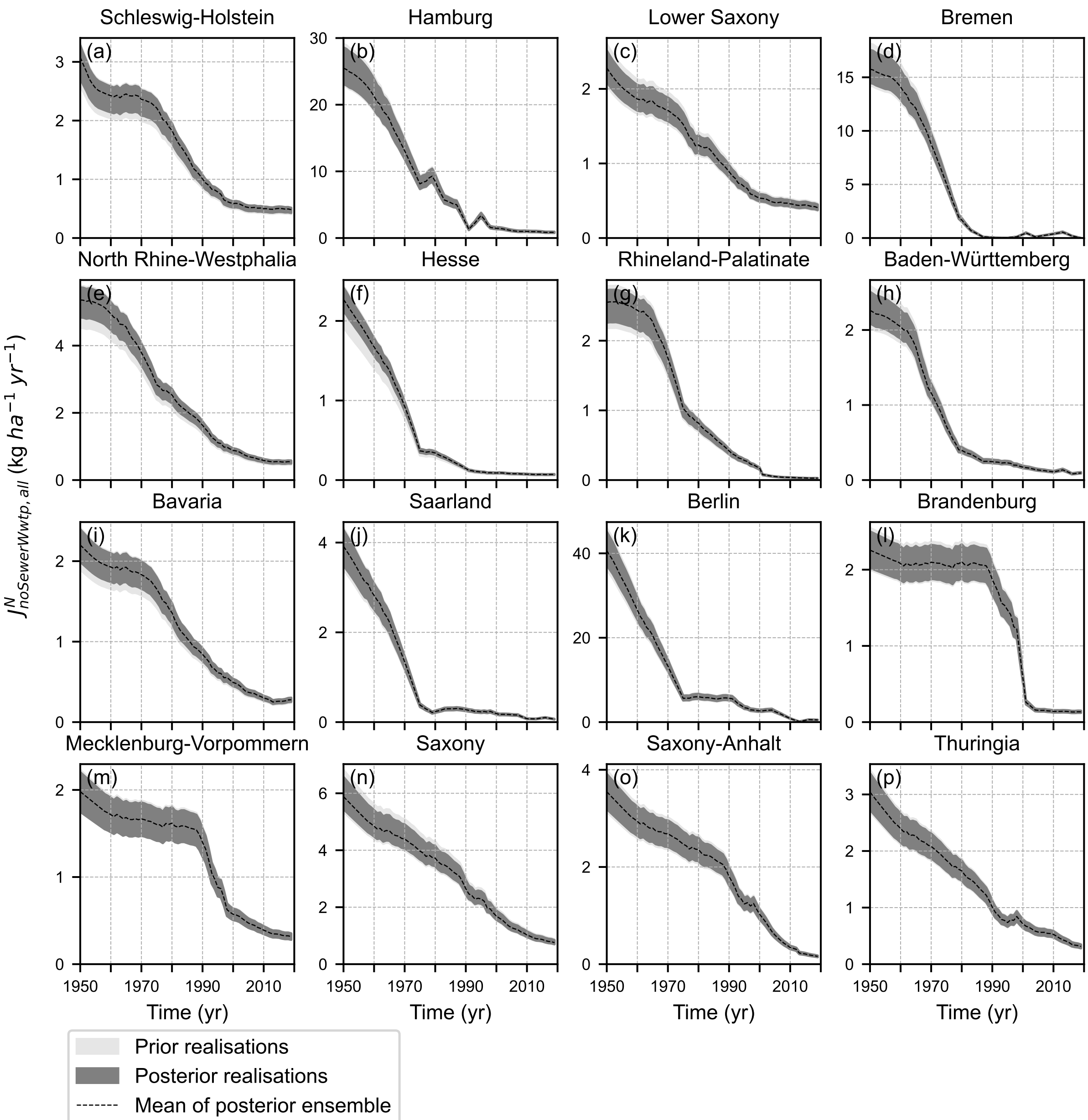


Figure S55. N gross emissions that are not collected in the sewer system nor treated in WWTPs $J^N_{noSewerWwtp,all}$, including both the urban and rural components, and the domestic and industrial/commercial components, from the prior and posterior model realisations at NUTS-1 level for the period 1950–2019. The shaded areas delineate the minimum and maximum values of the 100,000 prior realisations (light grey) and the 100 posterior realisations (dark grey).

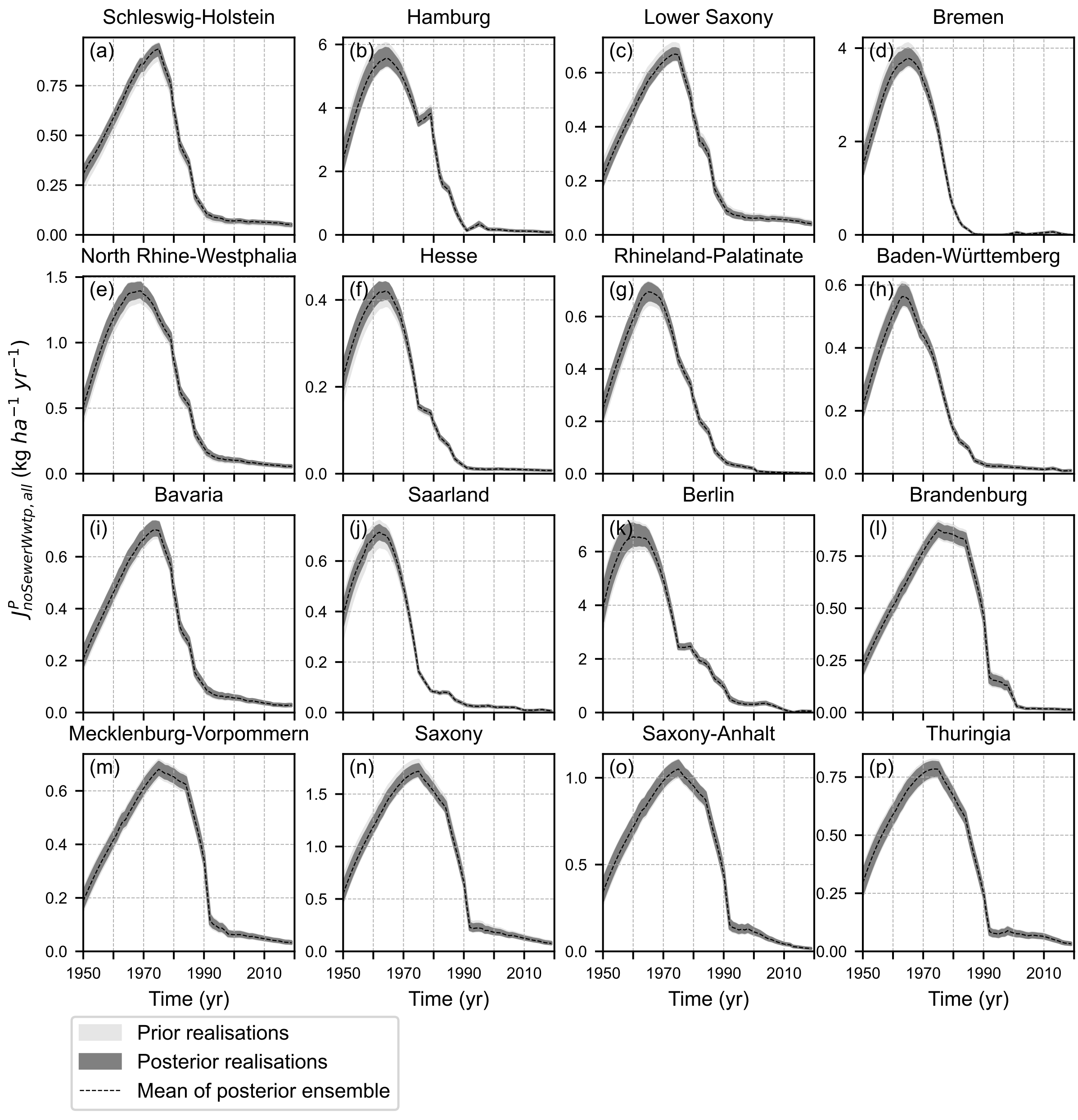


Figure S56. P gross emissions that are not collected in the sewer system nor treated in WWTPs $J^P_{noSewerWwtp,all}$, including both the urban and rural components, and the domestic and industrial/commercial components, from the prior and posterior model realisations at NUTS-1 level for the period 1950–2019. The shaded areas delineate the minimum and maximum values of the 100,000 prior realisations (light grey) and the 100 posterior realisations (dark grey).

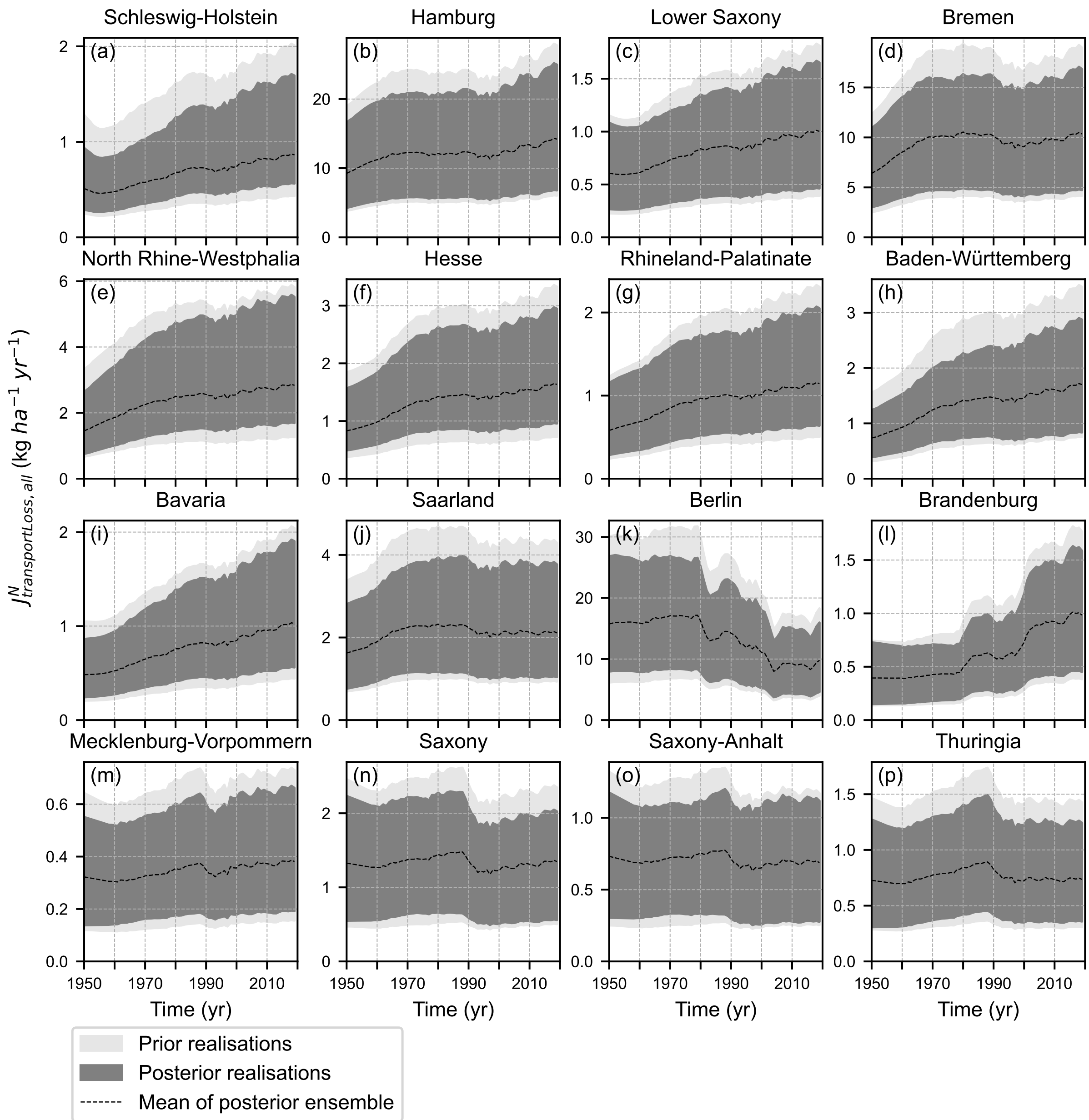


Figure S57. N emissions lost during wastewater collection and transport $J^N_{transportLoss,all}$, including both the urban and rural components, and the domestic and industrial/commercial components, from the prior and posterior model realisations at NUTS-1 level for the period 1950–2019. The shaded areas delineate the minimum and maximum values of the 100,000 prior realisations (light grey) and the 100 posterior realisations (dark grey).

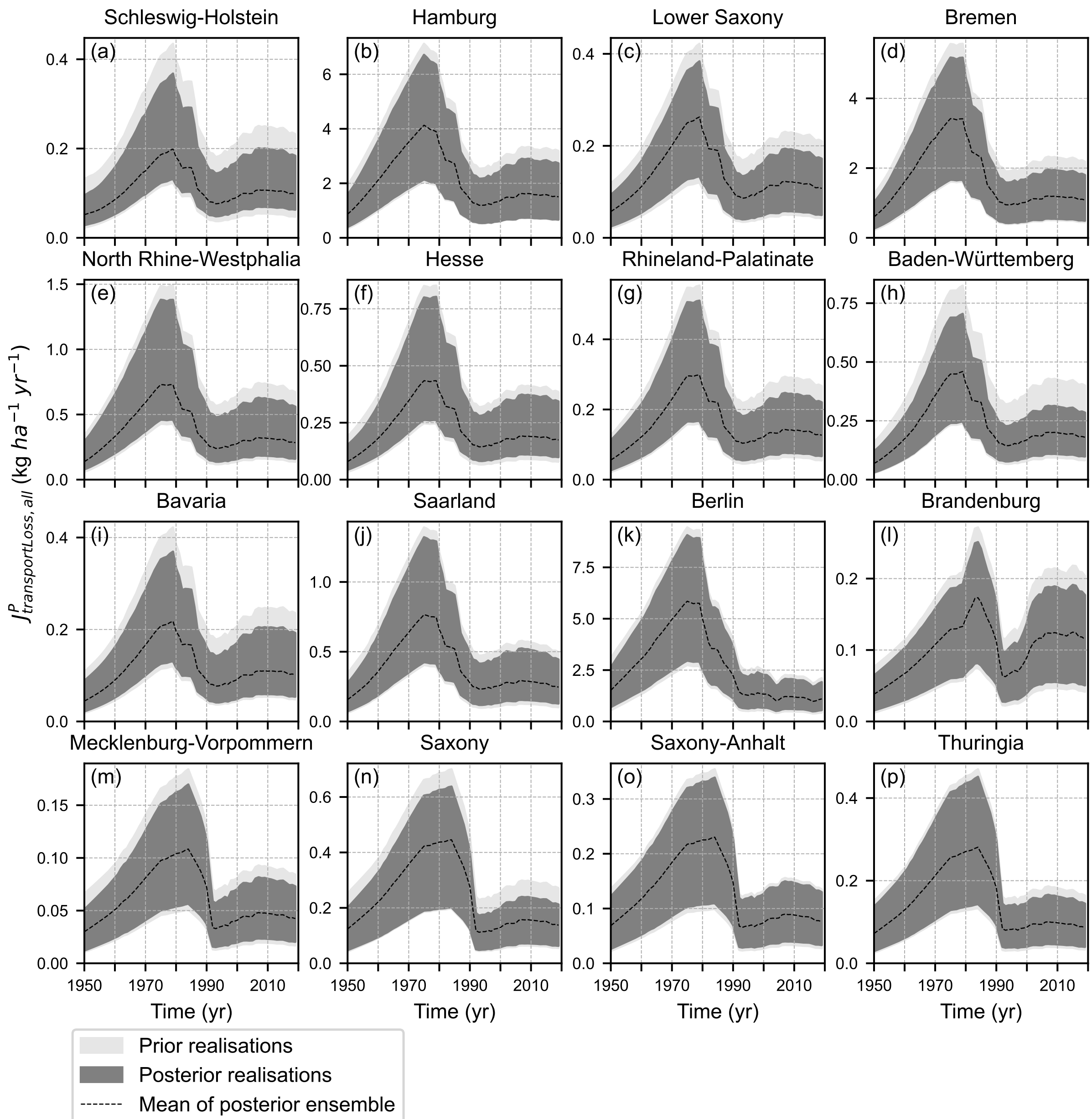


Figure S58. P emissions lost during wastewater collection and transport $J^P_{transportLoss,all}$, including both the urban and rural components, and the domestic and industrial/commercial components, from the prior and posterior model realisations at NUTS-1 level for the period 1950–2019. The shaded areas delineate the minimum and maximum values of the 100,000 prior realisations (light grey) and the 100 posterior realisations (dark grey).

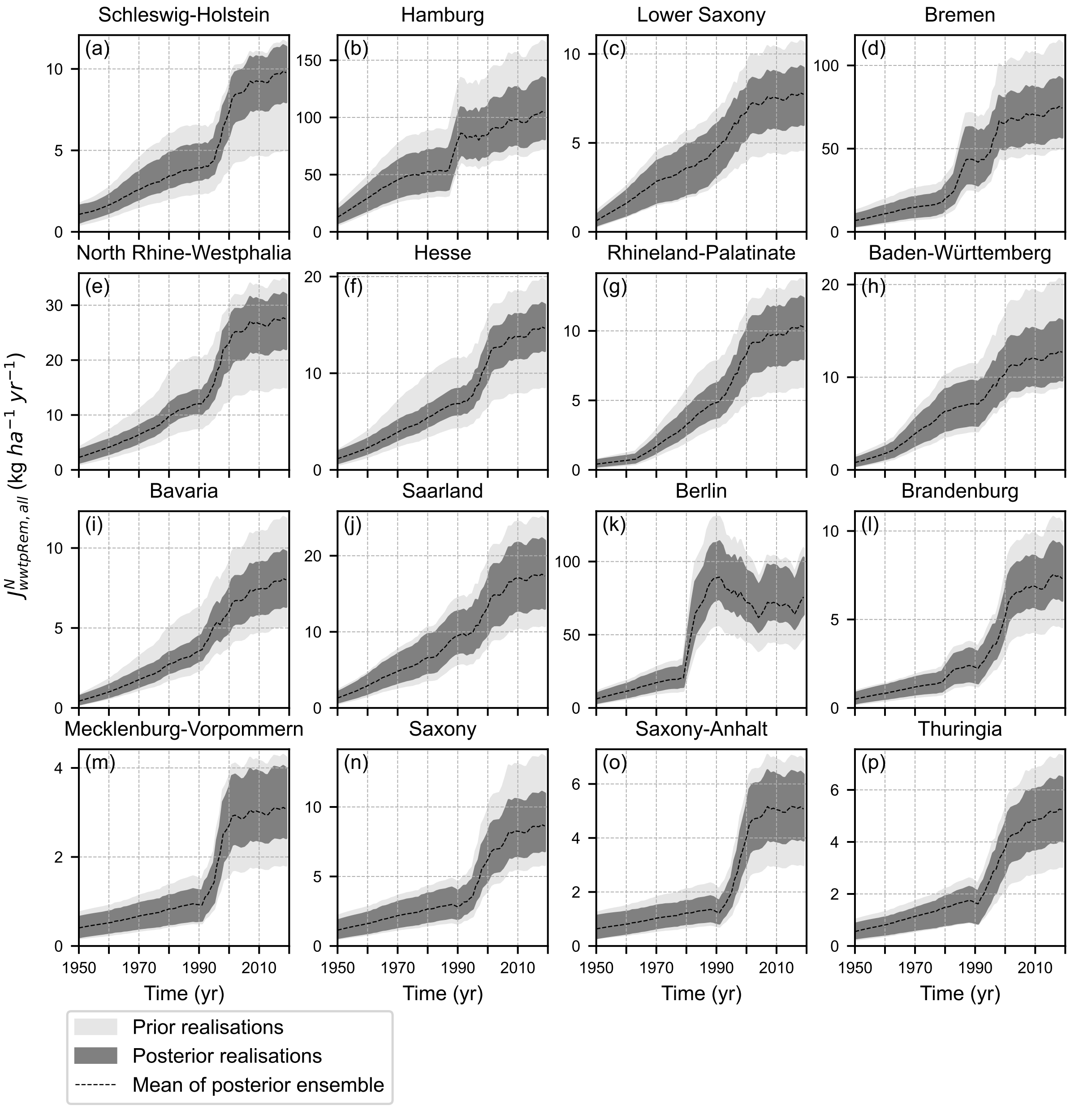


Figure S59. N emissions removed during treatment in WWTPs $J^N_{wwtpRem,all}$, including both the urban and rural components, and the domestic and industrial/commercial components, from the prior and posterior model realisations at NUTS-1 level for the period 1950–2019. The shaded areas delineate the minimum and maximum values of the 100,000 prior realisations (light grey) and the 100 posterior realisations (dark grey).

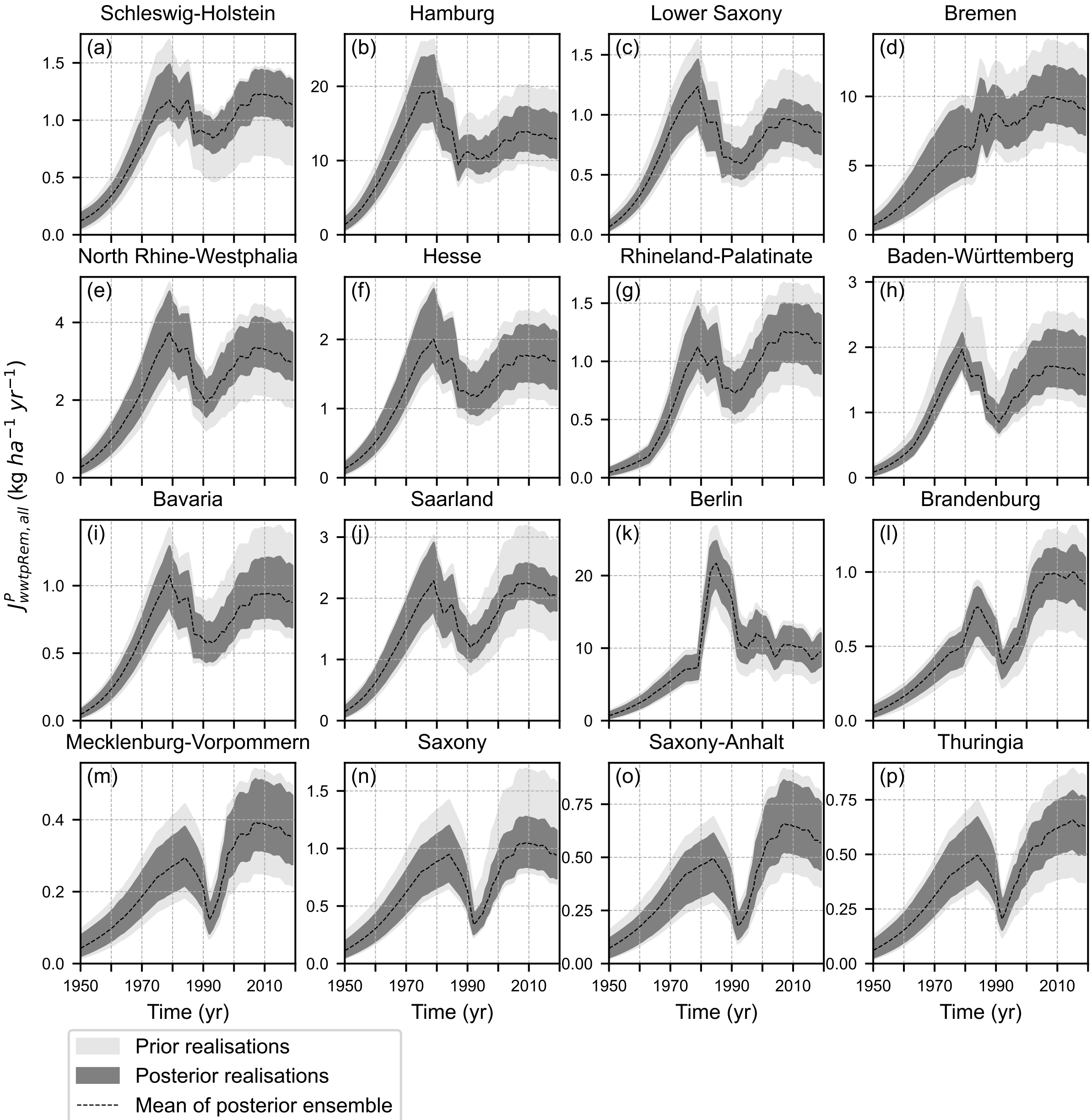


Figure S60. N emissions removed during treatment in WWTPs $J^P_{wwtpRem,all}$, including both the urban and rural components, and the domestic and industrial/commercial components, from the prior and posterior model realisations at NUTS-1 level for the period 1950–2019. The shaded areas delineate the minimum and maximum values of the 100,000 prior realisations (light grey) and the 100 posterior realisations (dark grey).

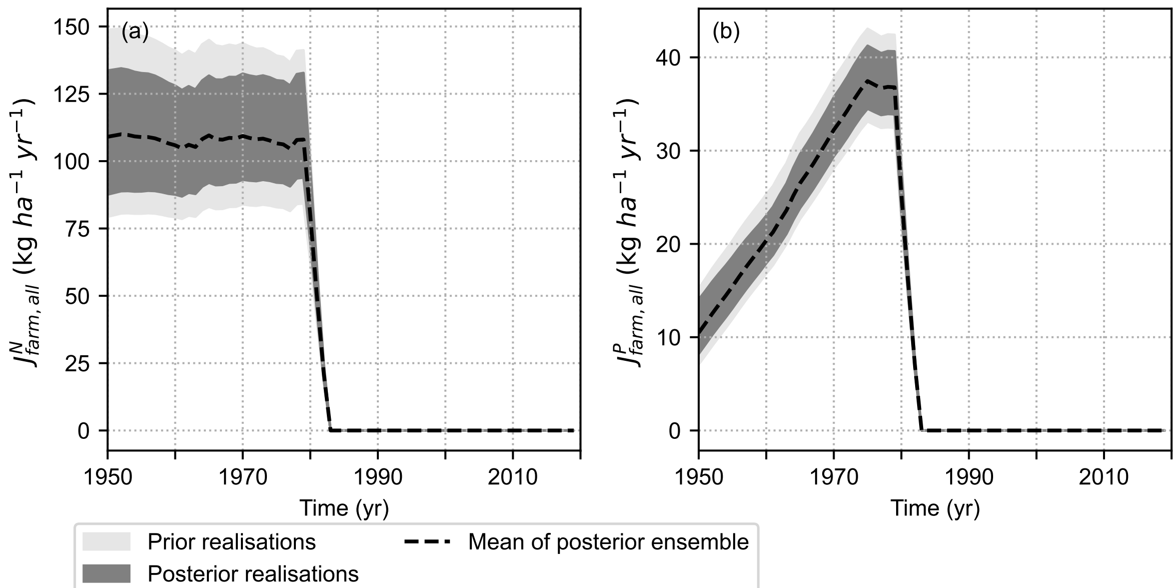


Figure S61. N and P emissions applied to agricultural soils in sewage farms ($J^N_{farm,all}$ and $J^P_{farm,all}$, respectively), including both the urban and rural components, and the domestic and industrial/commercial components, from the prior and posterior model realisations for Berlin for the period 1950–2019. The shaded areas delineate the minimum and maximum values of the 100,000 prior realisations (light grey) and the 100 posterior realisations (dark grey). We only consider sewage farms in the NUTS-1 region of Berlin.

S10.3 Gridded point sources estimates and their uncertainty due to the spatial disaggregation scheme

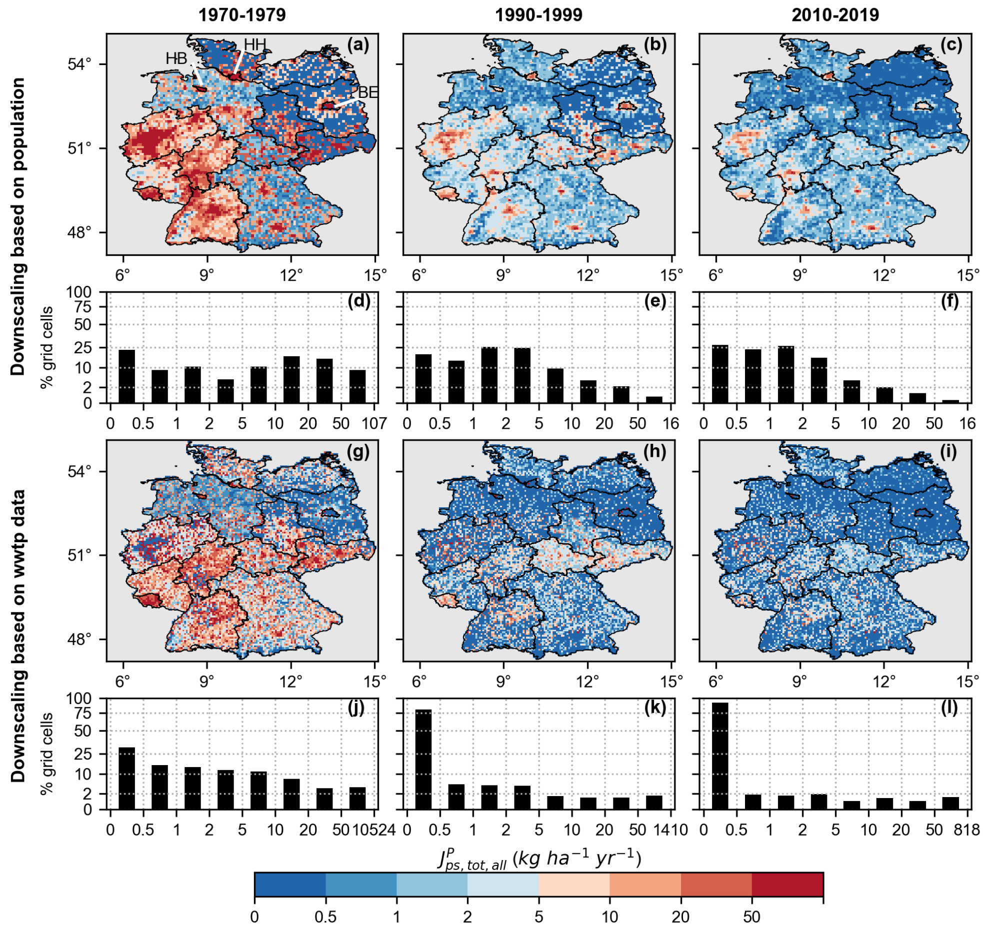


Figure S62. Gridded maps of total P point sources ($J^{P}_{ps,tot,all}$ in $kg\ ha^{-1}\ yr^{-1}$; panels a–c and g–i), including both the treated and untreated components, and distribution of the grid cell values of $J^{P}_{ps,tot,all}$ (panels d–f and j–l) obtained by downscaling the mean of the posterior realisations from NUTS-1 to grid level using population data (panels a–f) and WWTP data (panels g-l) for the treated component. The untreated components is always downscaled based on population data. The figure provides the average values over three different decades, that is, 1970–1979 (left panels), 1990–1999 (middle panels) and 2010–2019 (right panels). Panels (a–c) show the data at a resolution of 0.015625°, while panels (g–i) report aggregated values at a resolution of 0.0625°for depiction purpose (the maps at 0.015625°resolution are reported in Supplementary Fig S64). Panels (d–f and j–l) all refer to the distribution of the grid cells values at 0.015625°resolution. In all panels, the values of $J^{P}_{ps,tot,all}$ were grouped into the same eight classes. The last class of values (depicted in dark red colour in the maps) includes all values higher than 5 $kg\ ha^{-1}\ yr^{-1}$. The figure also reports the boundaries of the 16 NUTS-1 units of Germany and indicates the location of the city states that are Berlin (BE), Bremen (HB), Hamburg (HH), and North Rhine-Westphalia (NW). Data source: the NUTS-1 map comes from the German Federal Agency for Cartography and Geodesy (BKG, 2020) ©GeoBasis-DE/BKG dl-de/by-2-0 license (https://www.govdata.de/dl-de/by-2-0). The map is shown in the WGS84 system in the figure.

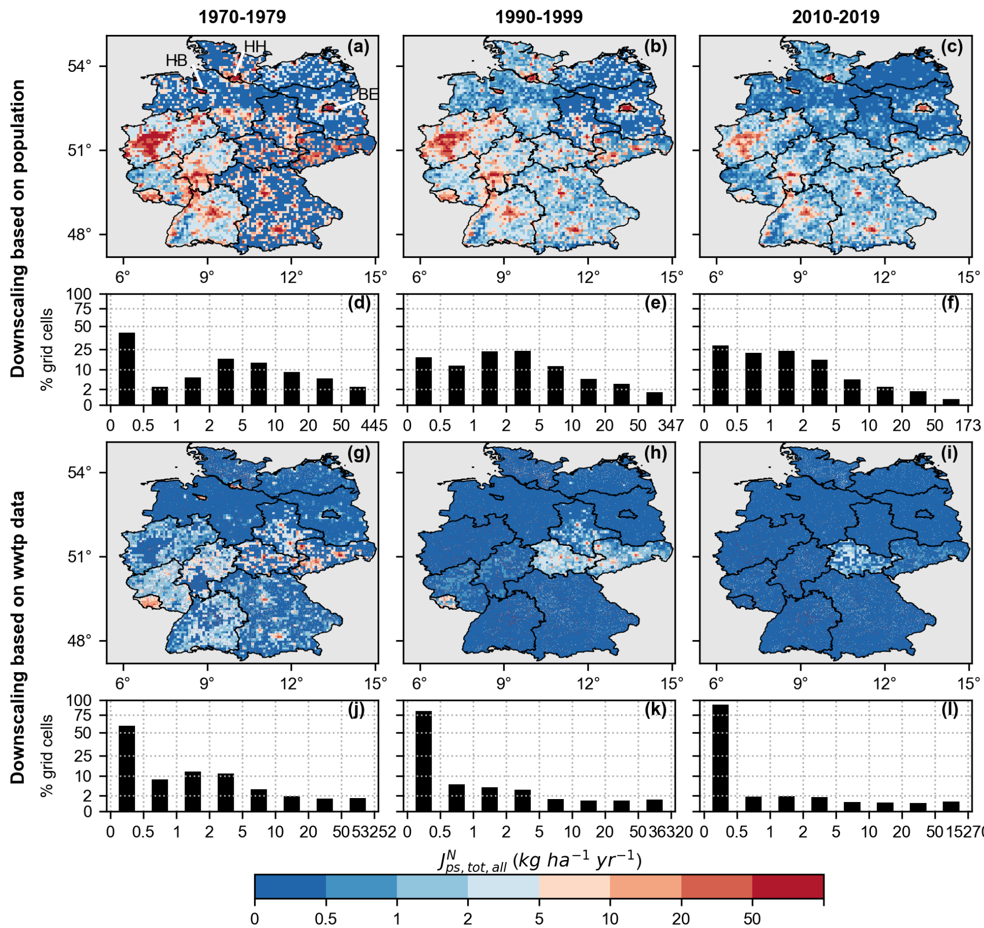


Figure S63. Gridded maps of total N point sources ($J^N_{ps,tot,all}$ in $kg\ ha^{-1}\ yr^{-1}$; panels a–c and g–i) at a resolution of 0.015625°, including both the treated and untreated components, and distribution of the grid cell values of $J^N_{ps,tot,all}$ (panels d–f and j–l) obtained by downscaling the mean of the posterior realisations from NUTS-1 to grid level using population data (panels a–f) and WWTP data (panels g-l) for the treated component. The untreated components is always downscaled based on population data. The figure provides the average values over three different decades, that is, 1970–1979 (left panels), 1990–1999 (middle panels) and 2010–2019 (right panels). In all panels, the values of $J^N_{ps,tot,all}$ were grouped into the same eight classes. The last class of values (depicted in dark red colour in the maps) includes all values higher than 50 $kg\ ha^{-1}\ yr^{-1}$. The figure also reports the boundaries of the 16 NUTS-1 units of Germany and indicates the location of the city states that are Berlin (BE), Bremen (HB), Hamburg (HH), and North Rhine-Westphalia (NW). Data source: the NUTS-1 map comes from the German Federal Agency for Cartography and Geodesy (BKG, 2020) ©GeoBasis-DE/BKG dl-de/by-2-0 license (https://www.govdata.de/dl-de/by-2-0). The map is shown in the WGS84 system in the figure.

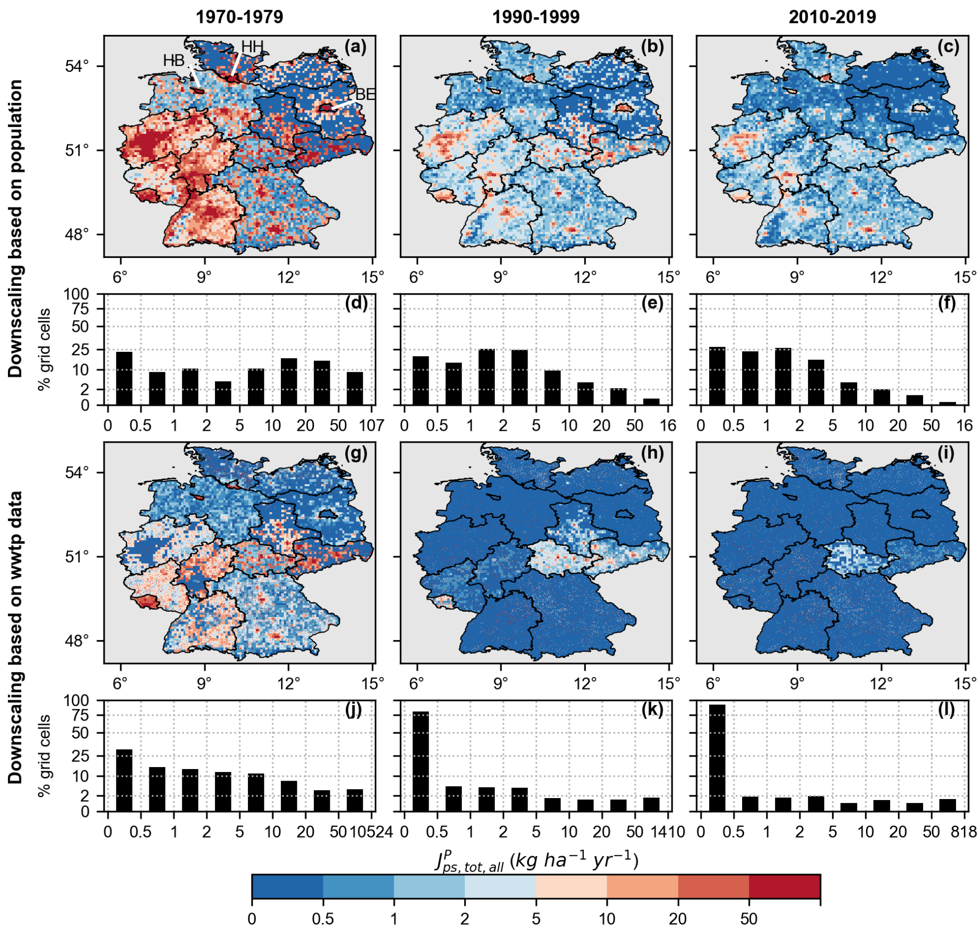


Figure S64. Gridded maps of total P point sources at a resolution of 0.015625° ($J^P_{ps,tot,all}$ in $kg\ ha^{-1}\ yr^{-1}$; panels a–c and g–i), including both the treated and untreated components, and distribution of the grid cell values of $J^P_{ps,tot,all}$ (panels d–f and j–l) obtained by downscaling the mean of the posterior realisations from NUTS-1 to grid level using population data (panels a–f) and WWTP data (panels g-l) for the treated component. The untreated components is always downscaled based on population data. The figure provides the average values over three different decades, that is, 1970–1979 (left panels), 1990–1999 (middle panels) and 2010–2019 (right panels). In all panels, the values of $J^P_{ps,tot,all}$ were grouped into the same eight classes. The last class of values (depicted in dark red colour in the maps) includes all values higher than 5 $kg\ ha^{-1}\ yr^{-1}$. The figure also reports the boundaries of the 16 NUTS-1 units of Germany and indicates the location of the city states that are Berlin (BE), Bremen (HB), Hamburg (HH), and North Rhine-Westphalia (NW). Data source: the NUTS-1 map comes from the German Federal Agency for Cartography and Geodesy (BKG, 2020) ©GeoBasis-DE/BKG dl-de/by-2-0 license (https://www.govdata.de/dl-de/by-2-0). The map is shown in the WGS84 system in the figure.

S10.4 Uncertainty in point source estimates at river basin level due to the spatial disaggregation scheme

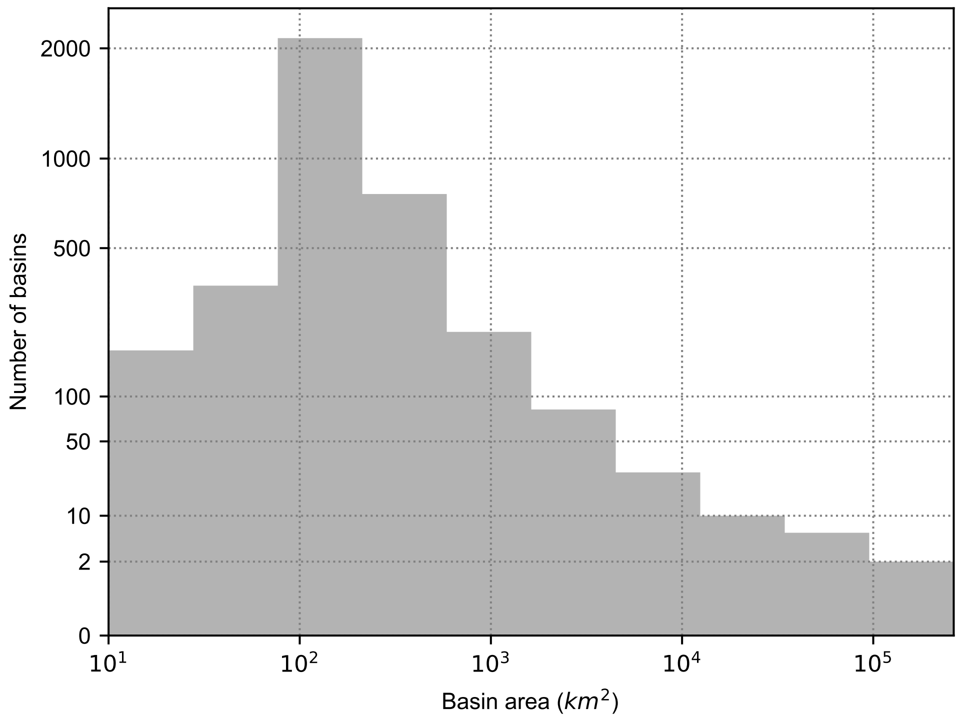


Figure S65. Histogram of the basin area for the 3778 selected HydroSHEDS basins. Data source: basin boundaries come from the HydroBASINS v1.c of the HydroSHEDS database (HydroSHEDS, 2014; Lehner and Grill, 2013) ©HydroSHEDS.

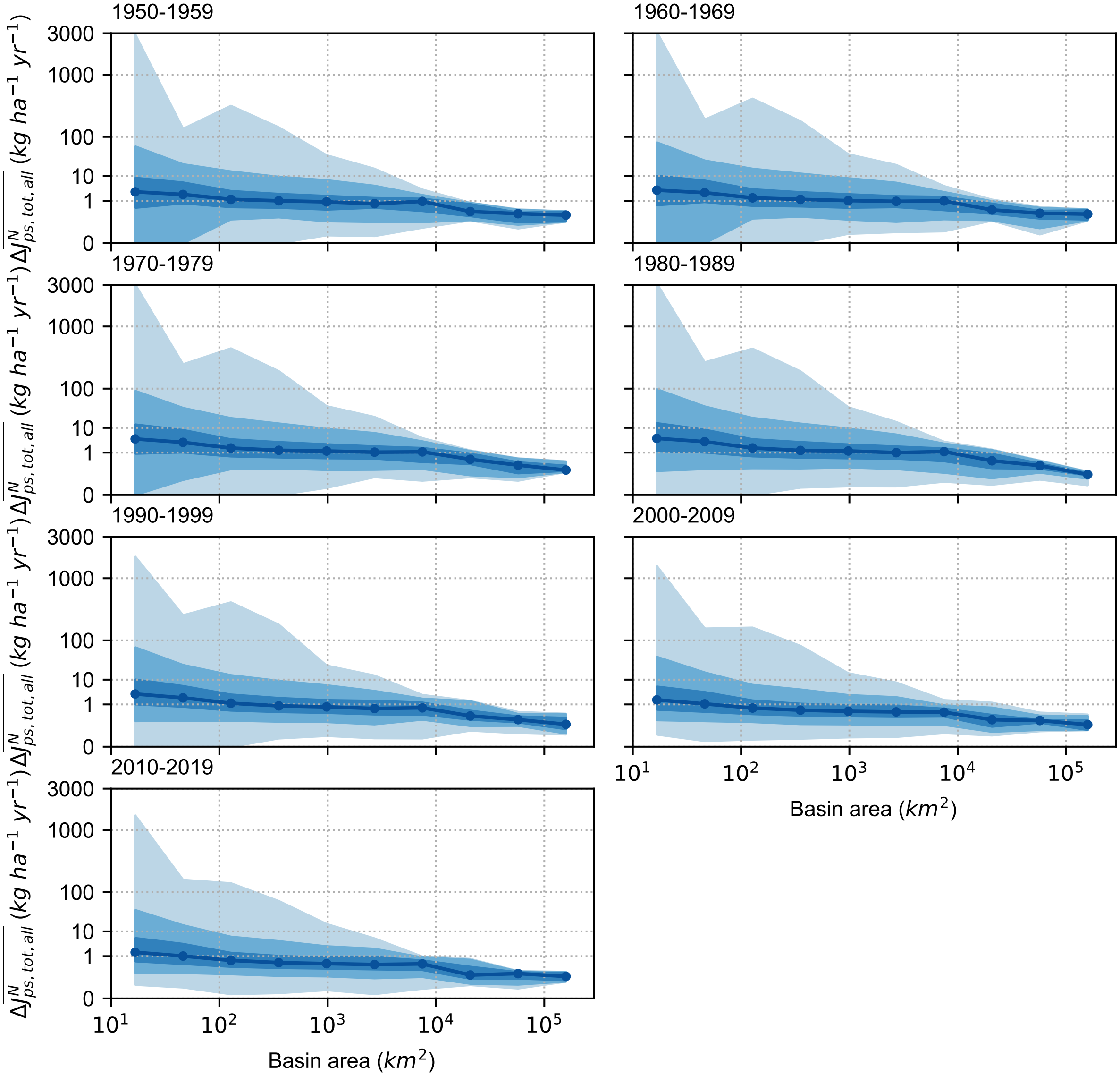


Figure S66. Distribution (median, Interquartile range (IQR), 90 % confidence interval (CI) and full range) of the absolute difference in the means of the posterior N point source realisations between the two downscaling approaches based on population and WWTP data ($\overline{\Delta J^N_{ps,tot,all}}$ *in* $kg\ ha^{-1}\ yr^{-1}$) against the basin area, that is grouped into ten classes whose centers are reported as dark blue circles along the median line for the 3778 HydroSHEDS basins. The distribution is reported for seven different decades, from 1950–1959 to 2010–2019. Data source: basin boundaries come from the HydroBASINS v1.c of the HydroSHEDS database (HydroSHEDS, 2014; Lehner and Grill, 2013) ©HydroSHEDS.

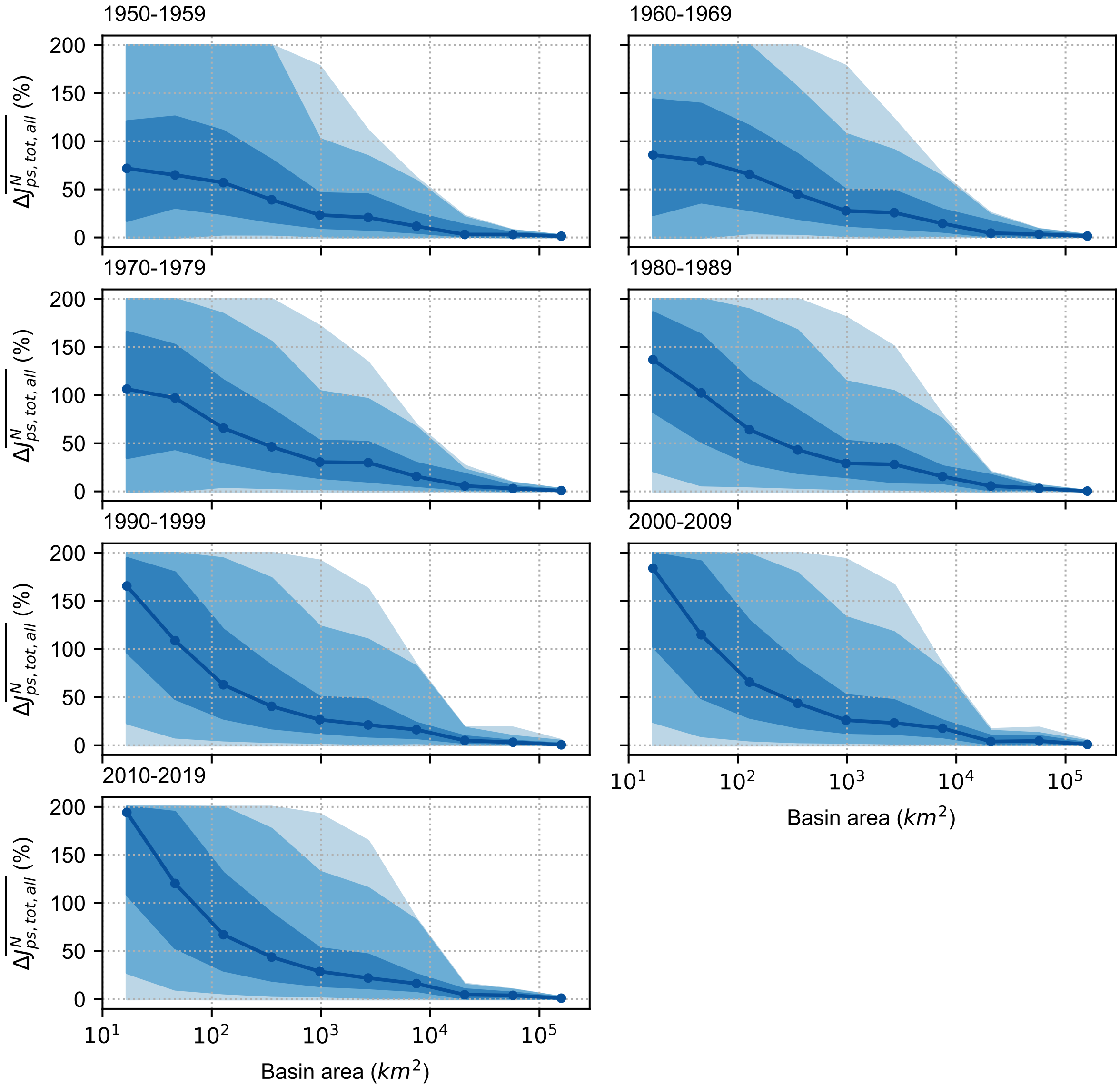


Figure S67. Distribution (median, Interquartile range (IQR), 90 % confidence interval (CI) and full range) of the relative difference in the means of the posterior N point source realisations between the two downscaling approaches based on population and WWTP data ($\Delta\overline{J^N_{ps,tot,all}}$ *in* %) against the basin area, that is grouped into ten classes whose centers are reported as dark blue circles along the median line for the 3778 HydroSHEDS basins. The distribution is reported for seven different decades, from 1950–1959 to 2010–2019. The relative difference is defined as follows: $\Delta\overline{J^N_{ps,tot,all}} = 100\frac{|J^N_{ps,tot,all,pop} - J^N_{ps,tot,all,wwtp}|}{\frac{J^N_{ps,tot,all,pop} + J^N_{ps,tot,all,wwtp}}{2}}$ where $J^N_{ps,tot,all,pop}$ is the total N point sources disaggregated using population data for the treated part and $J^N_{ps,tot,all,wwtp}$ is the total N point sources disaggregated using WWTP data for the treated part. Data source: basin boundaries come from the HydroBASINS v1.c of the HydroSHEDS database (HydroSHEDS, 2014; Lehner and Grill, 2013) ©HydroSHEDS.

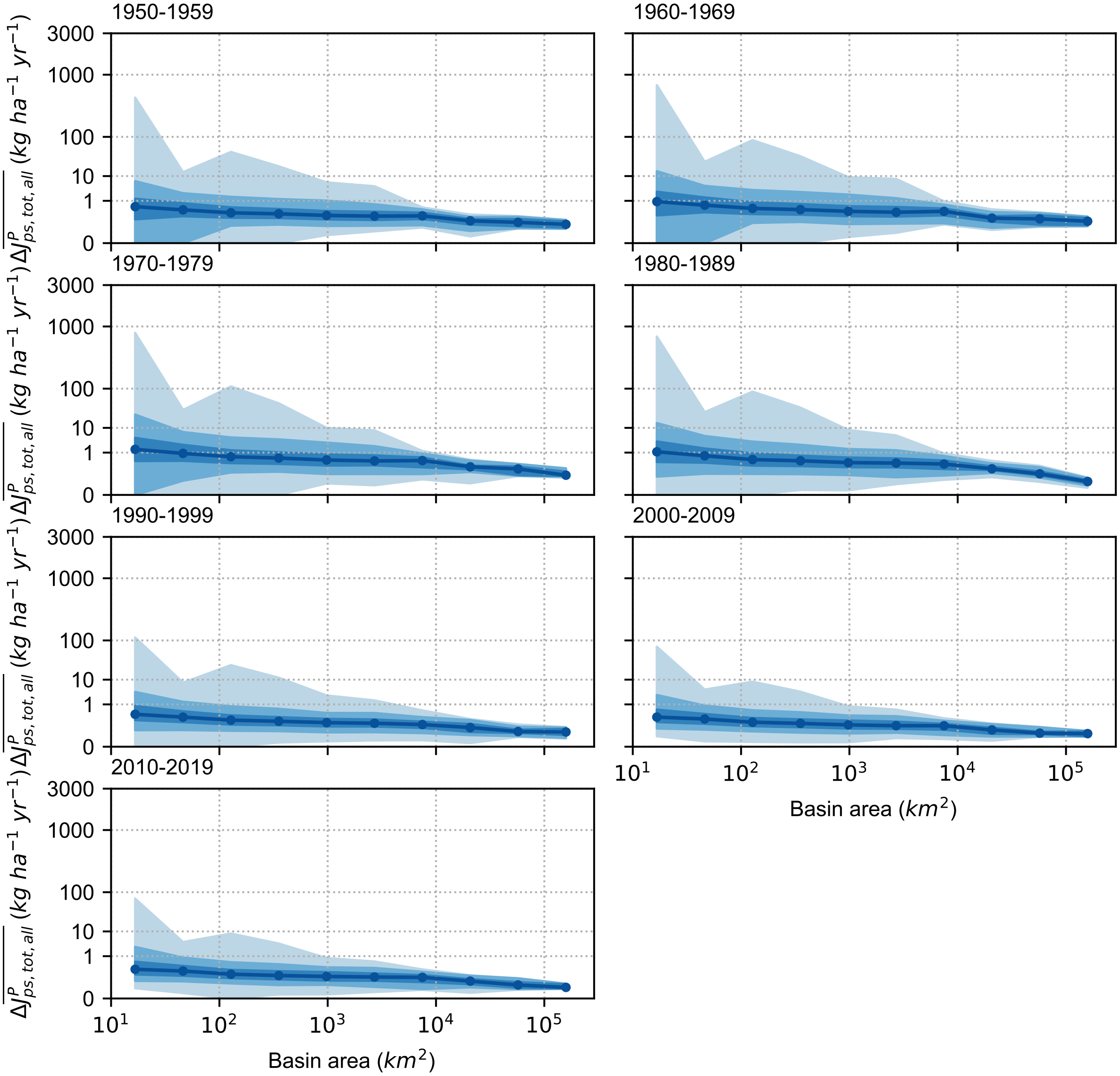


Figure S68. Distribution (median, Interquartile range (IQR), 90 % confidence interval (CI) and full range) of the absolute difference in the means of the posterior P point source realisations between the two downscaling approaches based on population and WWTP data ($\Delta\overline{J^P_{ps,tot,all}}$ *in* $kg\ ha^{-1}\ yr^{-1}$) against the basin area, that is grouped into ten classes whose centers are reported as dark blue circles along the median line for the 3778 HydroSHEDS basins. The distribution is reported for seven different decades, from 1950–1959 to 2010–2019. Data source: basin boundaries come from the HydroBASINS v1.c of the HydroSHEDS database (HydroSHEDS, 2014; Lehner and Grill, 2013) ©HydroSHEDS.

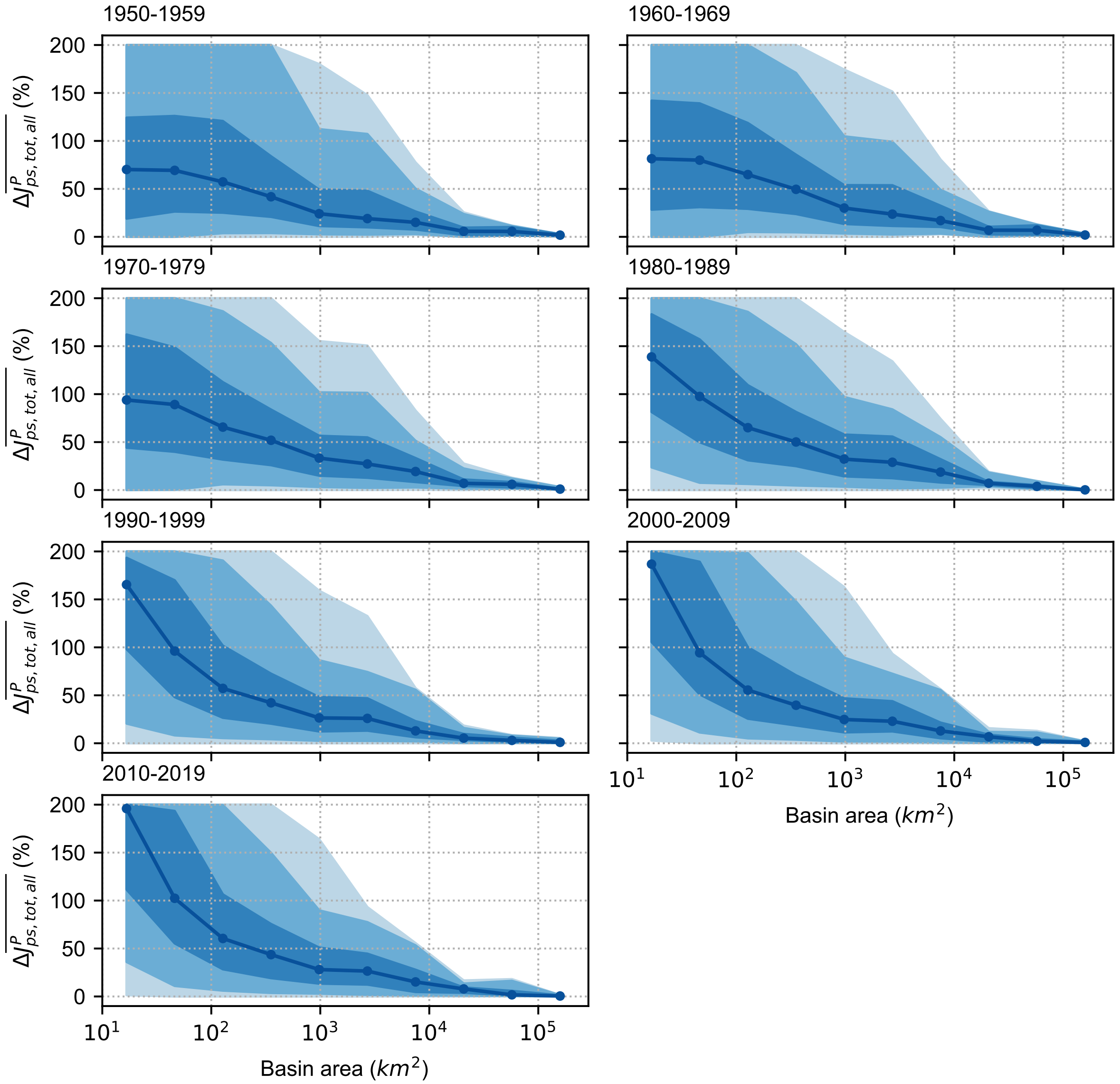


Figure S69. Distribution (median, Interquartile range (IQR), 90 % confidence interval (CI) and full range) of the relative difference in the means of the posterior P point source realisations between the two downscaling approaches based on population and WWTP data ($\Delta\overline{J^P_{ps,tot,all}}$ *in* %) against the basin area, that is grouped into ten classes whose centers are reported as dark blue circles along the median line for the 3778 HydroSHEDS basins. The distribution is reported for seven different decades, from 1950–1959 to 2010–2019. The relative difference is defined as follows: $\Delta\overline{J^P_{ps,tot,all}} = 100\frac{|J^P_{ps,tot,all,pop} - J^P_{ps,tot,all,wwtp}|}{\frac{J^P_{ps,tot,all,pop} + J^P_{ps,tot,all,wwtp}}{2}}$ where $J^P_{ps,tot,all,pop}$ is the total P point sources disaggregated using population data for the treated part and $J^P_{ps,tot,all,wwtp}$ is the total P point sources disaggregated using WWTP data for the treated part. Data source: basin boundaries come from the HydroBASINS v1.c of the HydroSHEDS database (HydroSHEDS, 2014; Lehner and Grill, 2013) ©HydroSHEDS.

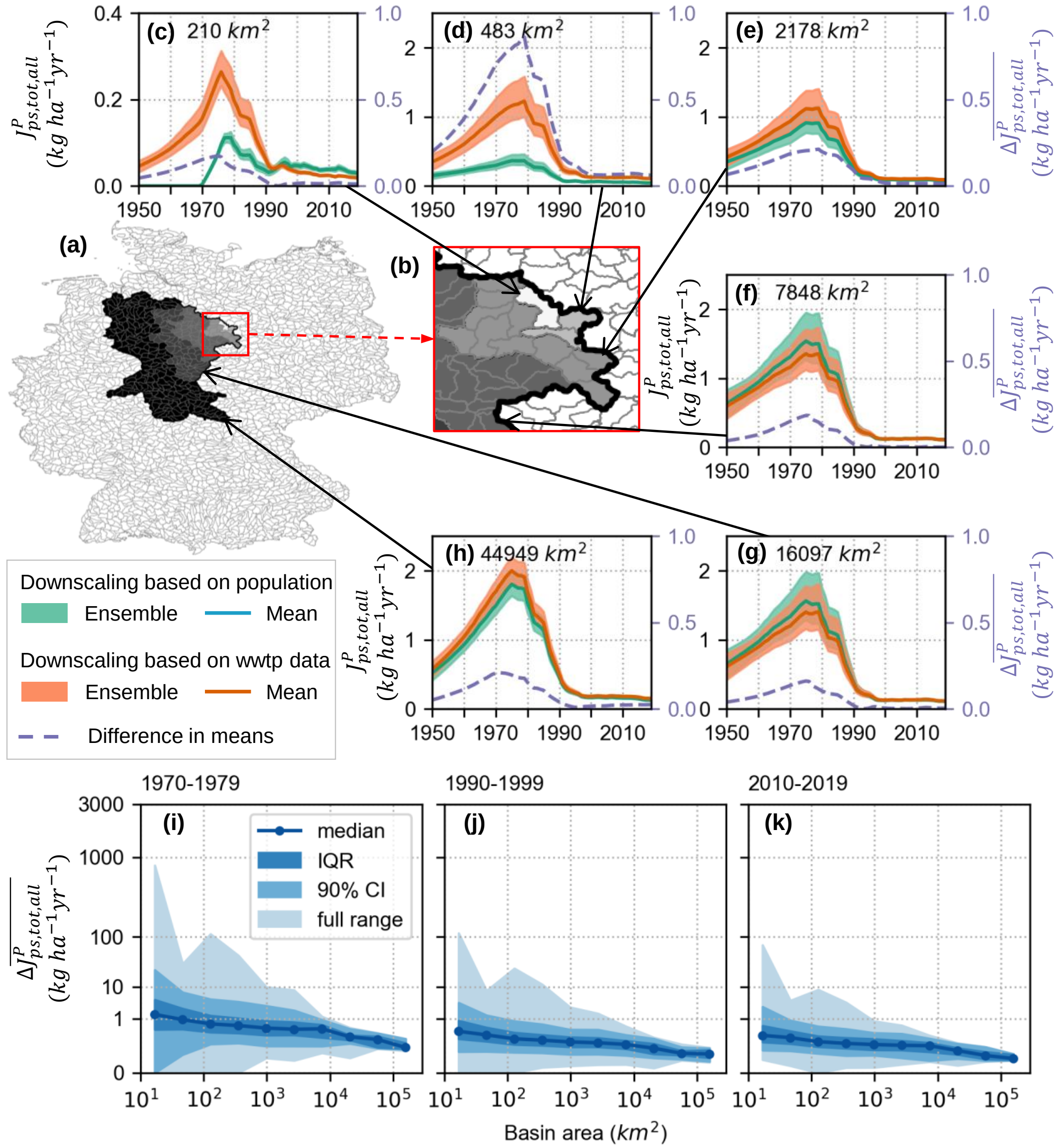


Figure S70. P point sources ($J^P_{ps,tot,all}$ *in kg* ha^{-1} yr^{-1}) and **absolute differences** in the mean of the posterior ensemble between the two downscaling approaches based on population and WWTP data ($\Delta\overline{J^P_{ps,tot,all}}$ *in kg* ha^{-1} yr^{-1}), aggregated at basins level for basins of the HydroSHEDS database. Panels (a-b) report the map of the 3778 HydroSHEDS basins selected over Germany, including six exemplary nested sub-basins of the Weser River Basin reported in grey shaded areas. Panels (c-h) represent the N point sources for the six sub-basins of the Weser River Basin. The light green and red shaded areas delineate the minimum and maximum values of the 100 posterior realisations for the downscaling approach based on population and WWTP data for the treated component, respectively. The solid dark green and red lines represent the mean of the posterior realisations, and the absolute difference between these two means is shown as dashed purple lines (secondary y-axis). Panels (i-k) report the distribution (median, Interquartile range (IQR), 90 % confidence interval (CI) and full range) of the difference in the means ($\Delta\overline{J^P_{ps,tot,all}}$) against the basin area, that is grouped into ten classes whose centers are reported as dark blue circles along the median line, for the 1970–1979 decade (panel i), 1990–1999 decade (panel j), and 2010–2019 decade (panel k). Data source: basin boundaries come from the HydroBASINS v1.c of the HydroSHEDS database (HydroSHEDS, 2014; Lehner and Grill, 2013) ©HydroSHEDS.

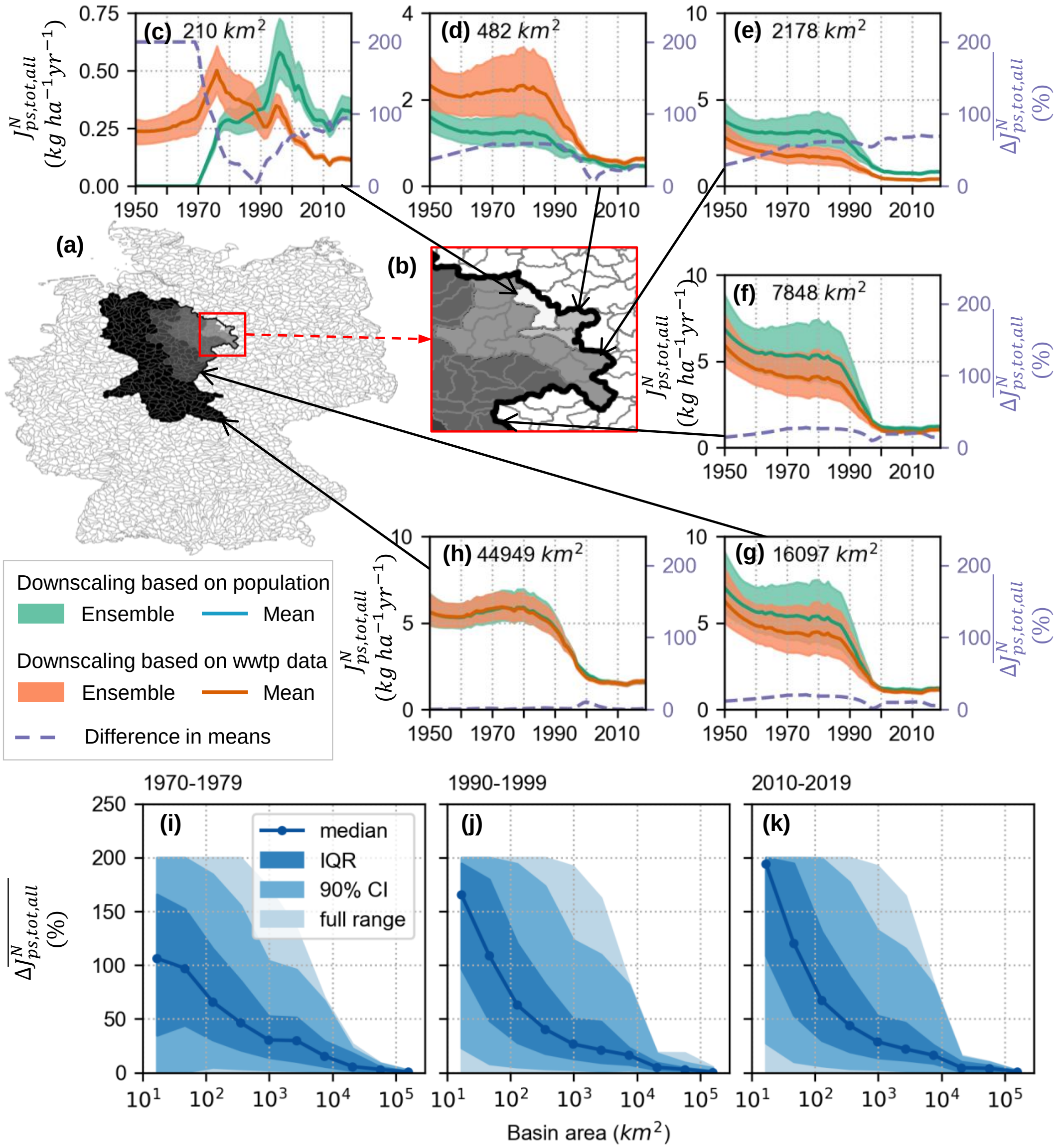


Figure S71. N point sources ($J^N_{ps,tot,all}$ *in kg ha*$^{-1}$ *yr*$^{-1}$) and **relative differences** in the mean of the posterior ensemble between the two downscaling approaches based on population and WWTP data ($\Delta\overline{J^N_{ps,tot,all}}$ *in* %), aggregated at basins level for basins of the HydroSHEDS database. Panels (a-b) report the map of the 3778 HydroSHEDS basins selected over Germany, including six exemplary nested sub-basins of the Weser River Basin reported in grey shaded areas. Panels (c-h) represent the N point sources for the six sub-basins of the Weser River Basin. The light green and red shaded areas delineate the minimum and maximum values of the 100 posterior realisations for the downscaling approach based on population and WWTP data, respectively, for the treated component. The solid dark green and red lines represent the mean of the posterior realisations, and the absolute difference between these two means is shown as dashed purple lines (secondary y-axis). Panels (i-k) report the distribution (median, Interquartile range (IQR), 90 % confidence interval (CI) and full range) of the relative difference in the means ($\Delta\overline{J^N_{ps,tot,all}}$) against the basin area, that is grouped into ten classes whose centers are reported as dark blue circles along the median line, for the 1970–1979 decade (panel i), 1990–1999 decade (panel j), and 2010–2019 decade (panel k). The relative difference is defined in the caption of Fig. S67. Data source: basin boundaries come from the HydroBASINS v1.c of the HydroSHEDS database (HydroSHEDS, 2014; Lehner and Grill, 2013) ©HydroSHEDS.

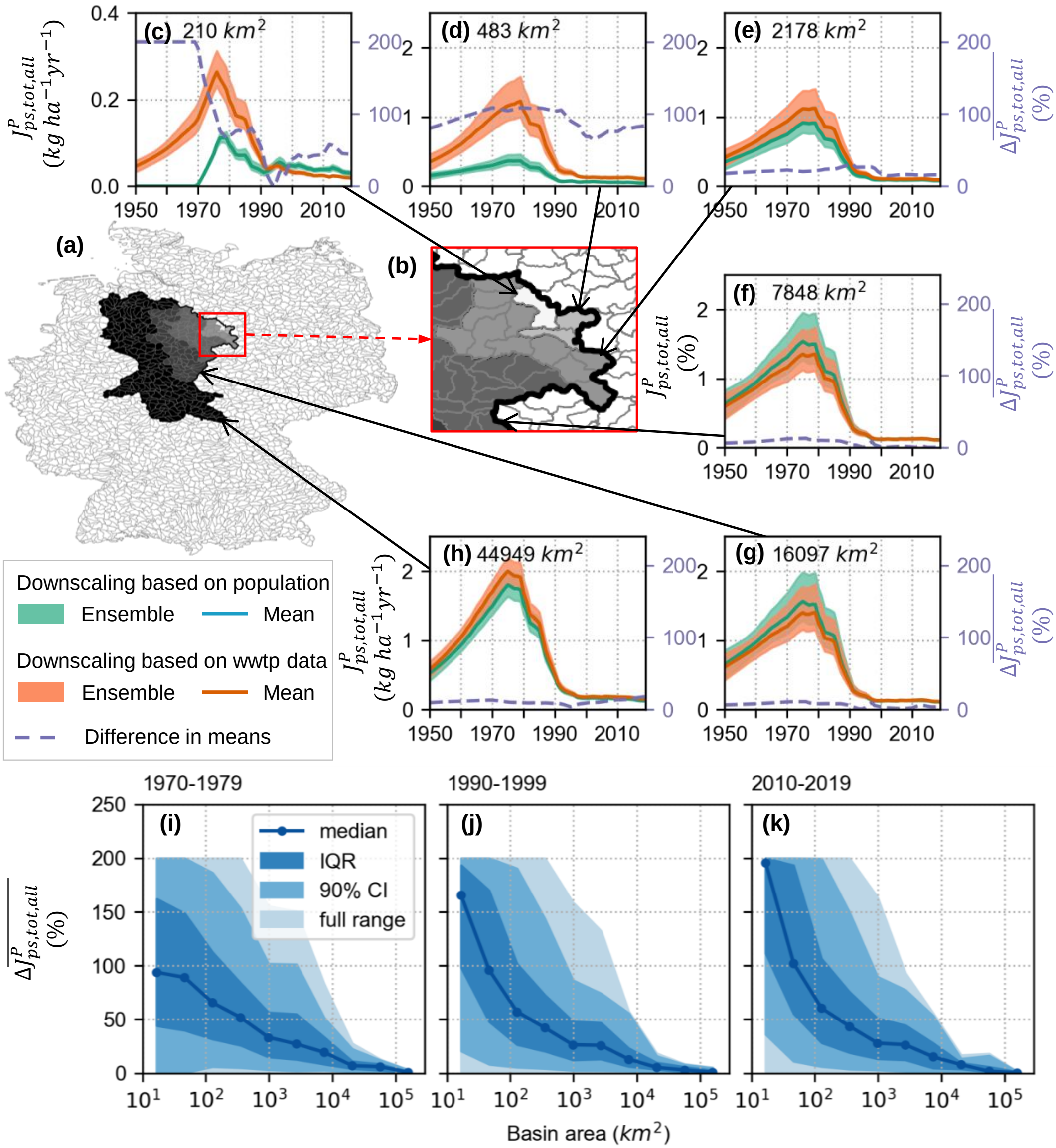


Figure S72. P point sources ($J^P_{ps,tot,all}$ *in* $kg\ ha^{-1}\ yr^{-1}$) and **relative differences** in the mean of the posterior ensemble between the two downscaling approaches based on population and WWTP data ($\overline{\Delta J^P_{ps,tot,all}}$ *in* %), aggregated at basins level for basins of the HydroSHEDS database. Panels (a-b) report the map of the 3778 HydroSHEDS basins selected over Germany, including six exemplary nested sub-basins of the Weser River Basin reported in grey shaded areas. Panels (c-h) represent the N point sources for the six sub-basins of the Weser River Basin. The light green and red shaded areas delineate the minimum and maximum values of the 100 posterior realisations for the downscaling approach based on population and WWTP data, respectively, for the treated component. The solid dark green and red lines represent the mean of the posterior realisations, and the absolute difference between these two means is shown as dashed purple lines (secondary y-axis). Panels (i-k) report the distribution (median, Interquartile range (IQR), 90 % confidence interval (CI) and full range) of the relative difference in the means ($\overline{\Delta J^P_{ps,tot,all}}$) against the basin area, that is grouped into ten classes whose centers are reported as dark blue circles along the median line, for the 1970–1979 decade (panel i), 1990–1999 decade (panel j), and 2010–2019 decade (panel k). The relative difference is defined in the caption of Fig. S69. Data source: basin boundaries come from the HydroBASINS v1.c of the HydroSHEDS database (HydroSHEDS, 2014; Lehner and Grill, 2013) ©HydroSHEDS.

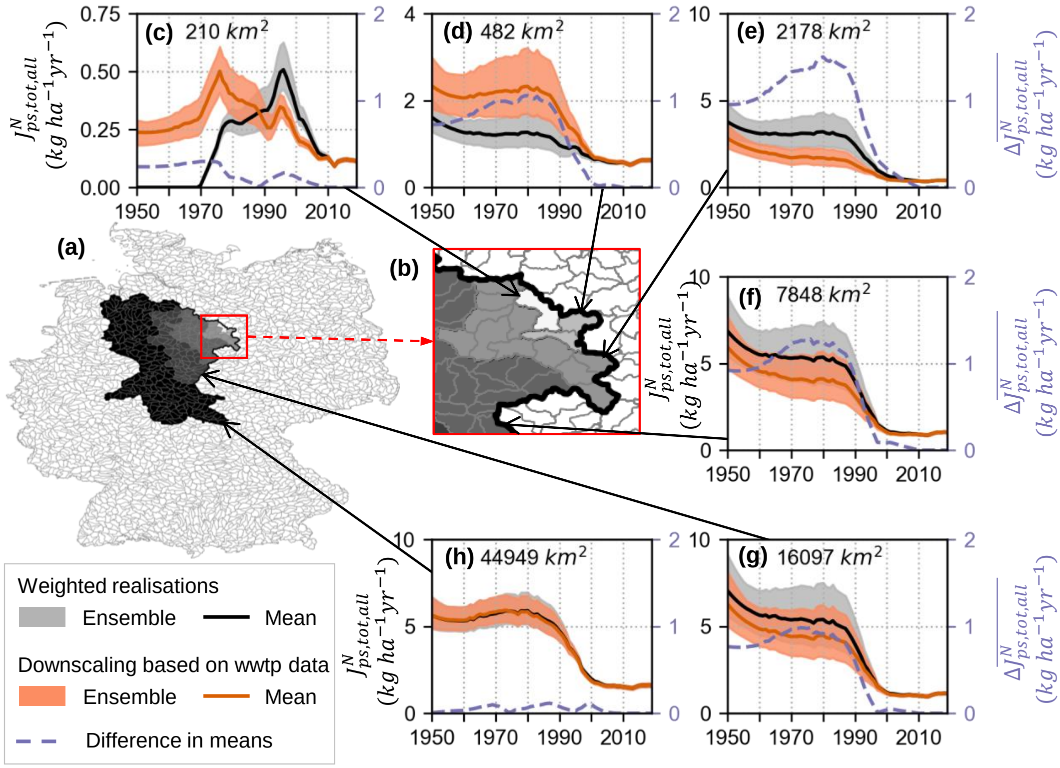


Figure S73. N point sources ($J^N_{ps,tot,all}$ *in kg ha*$^{-1}$ *yr*$^{-1}$) for two realisation ensembles and **absolute differences** in the mean of the two realisation ensembles ($\overline{\Delta J^N_{ps,tot,all}}$ *in kg ha*$^{-1}$ *yr*$^{-1}$), aggregated at basins level for basins of the HydroSHEDS database. Panels (a-b) report the map of the 3778 HydroSHEDS basins selected over Germany, including six exemplary nested sub-basins of the Weser River Basin reported in grey shaded areas. Panels (c-h) represent the N point sources for the six sub-basins of the Weser River Basin. The first realisation ensemble, shown in red colours, corresponds to point sources estimates downscaled based on WWTP data and is identical to the ensemble reported in Fig. 9 of the main paper in red colours. In the second realisation ensemble, shown in grey colours, each realisation corresponds to a weighted average of point sources estimated using one specific parameter set and the two different downscaling schemes based on WWTP and population data, that is, it corresponds to the average of the red and green realisations reported in Fig. 9 of the main paper. This weighted (grey) ensemble replace the (green) ensemble downscaled based on population data in Fig. 9 of the main paper. The weights are selected so to modify the estimates downscaled based on population data for the recent period, to be consistent with the estimates downscaled based on WWTP data that are more accurate. Specifically, here, for the period 2010–2019, the weight for the estimates based on WWTP data is 1, while it is 0 for the estimates based on population data, and conversely for the period 1950–1990 prior to the Urban Waste Water Treatment Directive EC (1991) that led to a large changes in wastewater treatment plants. For the intermediate period (1991–2009), the weights are linearly interpolated to ensure a smooth transition. The light red and grey shaded areas delineate the minimum and maximum values of the 100 posterior realisations for each ensemble. The solid dark red and black lines represent the mean of the posterior realisation ensembles, and the absolute difference between these two means is shown as dashed purple lines (secondary y-axis). We suggest that both the red and grey ensembles shown in this figure could be considered in water quality studies, instead of the red and green ensembles reported in Fig. 9 of the main paper, so to have a more reasonable uncertainty representation. The figure only illustrates a possible weighted scheme (grey ensemble), which could be adapted by the users of the dataset. Source: Basin boundaries come from the HydroBASINS v1.c of the HydroSHEDS database (HydroSHEDS, 2014; Lehner and Grill, 2013).

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