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NUTRIENT FLUXES FROM DOMESTIC WASTEWATER: A NATIONAL-SCALE HISTORICAL PERSPECTIVE FOR THE UK 1800-2010

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Abstract

Nutrient emissions in human waste and wastewater effluent fluxes from domestic sources are quantified for the UK over the period 1800-2010 based on population data from UK Census returns. The most important drivers of change have been the introduction of the water closet (flush toilet) along with population growth, urbanisation, connection to sewer, improvements in wastewater treatment and use of phosphorus in detergents. In 1800, the population of the UK was about 12 million and estimated emissions in human waste were 37 kt N, 6.2 kt P and 205 kt organic C per year. This would have been recycled to land with little or no sewage going directly to rivers or coastal waters. By 1900, population had increased to 35.6 million and some 145 kt N were emitted in human waste but, with only the major urban areas connected to sewers, only about 19 kt N were discharged in sewage effluent. With the use of phosphorus in detergents, estimated phosphorus emissions peaked at around 63.5 kt P/year in the 1980s, with about 28 kt P/year being discharged in sewage effluent. By 2010, population had increased to 63 million with estimated emissions of 263 kt N, 43.6 kt P and 1460 kt organic C per year, and an estimated effluent flux of 104kt N, 14.8 kt P and 63 kt organic C. Despite improvements in wastewater treatment, current levels of nutrient fluxes in sewage effluent are substantially higher than those in the early 20th century.

Keywords

nutrients, human waste, sewage effluent, history of sewerage, wastewater treatment, nitrogen, phosphorus

Introduction

Nutrient enrichment has important consequences for freshwater as well as coastal and marine ecosystems. Domestic wastewater is an important source of nutrients in both global and national biogeochemical cycles (Bouwman et al., 2005; Van Drecht et al., 2009; Morée et al., 2013). Indeed, it is estimated that, at the end of the 20th century, urban wastewater contributed some 7.7 Tg/year nitrogen (N) and 1.0 Tg/year phosphorus (P) to the world's oceans (Morée et al., 2013). This is a significant flux, making up about 6% of the total N flux and 50% total P flux (Seitzinger et al., 2010). The impact of domestic wastewater on riverine ecosystems has been well documented (e.g. Jarvie, et al. 2006; Neal et al., 2010). It is of particular concern in relation to eutrophication as nutrient fluxes which, with the exception of storm-water overflows, tend to be constant through the year and, therefore, enter rivers under low flow conditions when dilution is minimal. This results in high nutrient concentrations which can lead to algal blooms and reductions in available oxygen.

National-scale data on the quality and volume of sewage effluent are generally only available for recent decades. Yet, human-induced eutrophication has been reported in Europe and elsewhere since the early 20th century (de Jonge et al., 2002; Lewitus et al., 2012; May et al., 2012). The lack of historical data makes establishing reference conditions for surface waters, especially in and downstream of well-populated areas, very difficult. Reconstruction of past inputs to surface water, including those from land clearance and agriculture, would enable us to identify the main changes leading to early eutrophication and the factors responsible for increasing nutrient enrichment. It also sets the context for current nutrient levels in rivers and lakes (Sharpley et al., 2013).

Over the last two centuries, there have been huge changes in population, urbanization, diet, detergent phosphorus, sewer infrastructure and wastewater treatment. These have all had an impact not only on the total nutrient loading from sewage but also on its spatial distribution and on the different forms of nutrients and nutrient ratios (Seitzinger et al., 2010). Piecing together a detailed quantitative picture of historical nutrient loading from wastewater using sparse data and anecdotal evidence is a huge task. Previous studies have either focused at the detailed catchment scale (Weber et al., 2006; Behrendt et al., 2008; Gadegast et al., 2012) or provided a global assessment (Bouwman et al., 2005; Van Drecht et al., 2009; Morée et al., 2013). This paper seeks to quantify the nutrient fluxes in wastewater effluent from domestic sources in the UK for the period 1800-2010 using available evidence to develop reasonable assumptions for simulating the most important changes over time.

By way of introduction, the history of sewage in the UK (Cannon, 1912; Sellers, 1997; Cooper, 2001; Halliday, 2009) is summarised in Table 1. Prior to the invention and rapid spread of the water closet (WC), or flush toilet, in the 1830s, domestic waste was simply collected locally and used on the land. Indeed, with the rapid expansion of cities in the early 19th century, there was quite an industry of collecting "night soil" and selling it as fertilizer in the surrounding countryside (Cannon, 1912). With the advent of the WC, water use, and hence the liquid content of waste, increased enormously (Strang, 1859). Coupled with the import of guano from South America, this meant that it was no longer cost effective to transport human waste for use as fertiliser. The effect of this in major cities was that cess pools overflowed into the streets and raw sewage ran into water courses. In London, this culminated in the "Great Stink" of 1858 (Halliday, 2009) while outside London there were serious attempts to discourage the WC to avoid similar levels of pollution. In response, the late 19th century saw the building of the main sewer systems in all the major cities of the UK.

Table 1 Timeline for sewage and its treatment

1800-1830	population growth and move to cities; cesspools and middens; "night soil men" take waste to countryside for use as fertiliser
1830-1860	Water Closet (WC), or flush toilet, introduced in 1810; increased very rapidly after 1830 but discouraged in some northern cities; water use increased by factor of up to ten (Strang, 1859); collapse of fertiliser market due to cheap imports; raw sewage flowed directly to river; in London The Great Stink of 1858 (Halliday, 2009)
1860-1890	main interceptor sewers built; no treatment just moved waste downstream
1890-1940	primary sewage treatment implemented; first activated sludge plant in Davyhulme, Manchester 1914; >30% rural households had no piped water or sewerage (Kinnersley, 1988); first septic tank introduced to the UK in 1890.
1948	introduction of phosphorus in detergents.
1951	Rivers Prevention of Pollution Act (consents for new discharges)
1940-1970	secondary sewage treatment implemented more generally
1973	Water Act set up the 10 Regional Water Authorities in England and Wales; only about 50% effluents compliant with BOD and SS consents (Annual Reports of Regional Water Authorities 1974)
1974	Control of Pollution Act – public registration of discharge consents and results of monitoring
1989	Privatisation of the Water Industry in England and Wales; major new investment in infrastructure began.
1995	Environment Act – set up the Environment Agency in England and Wales and the Scottish Environment Protection Agency (SEPA) in Scotland as regulatory bodies
1998	EU Urban Wastewater Treatment Directive: secondary treatment mandatory for WWTWs > 15000 population equivalents (PE)
2005	EU Urban Waste Water Treatment Directive: secondary treatment mandatory for WWTWs > 2000 PE; tertiary treatment for designated sensitive areas.

Initially, the new sewers simply meant that raw sewage was moved downstream, away from the main city centres. Sewage treatment processes, beginning with primary settlement and biological filters were implemented widely between 1895 and 1920 to treat sewage from the growing towns (Cooper, 2001). The use of activated sludge was piloted at Davyhulme sewage works in 1914 (http://www.engineering-timelines.com/timelines.asp) and the period 1914 to 1965 saw the widespread use of this secondary treatment process across the UK. Since then improvements in sewage treatment have largely been a response to legislative drivers which established the framework for current standards of treatment and environmental protection, as well as providing the opportunity for greater investment (http://www.ofwat.gov.uk/wp-content/uploads/2015/11/rpt_com_devwatindust270106.pdf; Kinnersley, 1988; Defra, 2012).

In this study, annual estimates of the different forms of nitrogen, phosphorus and organic carbon from sewage sources in the UK 1800-2010 have been derived at a 5km grid resolution from population data. As far as possible, available evidence has been used to develop a quantitative representation of the key historical drivers of change. In the case of sewage treatment, there is a lack of detailed historical evidence at the national scale and we have therefore chosen to represent this as a series of assumed step changes, which make clear the impact of sewage treatment, rather than attempting to formulate a more realistic spatial and temporal trend. The results form one of the inputs to a large-scale historical integrated modelling study of the UK which includes nutrients from agricultural and other sources as well as nutrient transformations and losses in both river and lake processes.

Methods Overview

The overall approach is based on population data from UK Census returns and a series of multiplying factors derived from the literature and available evidence (Figure 1). These factors describe the nutrient emissions per person, the proportion of the population connected to sewer and the proportion of nutrients retained during wastewater treatment.

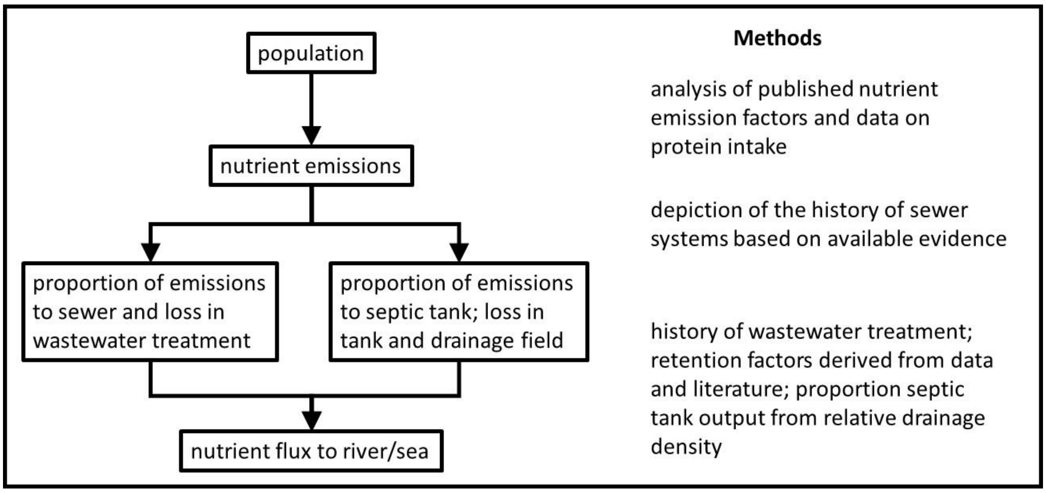

Figure 1 Summary of the approach taken and methods used

The standard calculation is given by (cf. Grizetti and Barraiou, 2006; Van Drecht et al., 2009; Williams et al., 2012)

$$NUT_{i,x,y} = P_{x,y}\, E_i\, C_{x,y} \sum_{j=0}^{4}\left[f_{j,x,y}\,(1 - T_{i,j})\right] \tag{1}$$

where $NUT_{i,x,y}$ is effluent flux to river/sea of nutrient i in grid square (x,y) in kg/year, $P_{x,y}$ is population in grid square (x,y), E_i is emission factor for nutrient i in kg/person/year, $C_{x,y}$ is proportion of population in grid square (x,y) connected to river/sea, $f_{j,x,y}$ is the fraction of effluent from grid square (x,y) treated by method j, $T_{i,j}$ is proportion of nutrient i retained in treatment j where j=0 indicates raw sewage, 1 primary treatment, 2 secondary treatment, 3 tertiary treatment and 4 phosphorus stripping.

A similar approach can be taken to estimate losses from septic tanks (e.g. Zhang et al., 2014):

$$NST_{i,x,y} = P_{x,y}\, E_i\, \left(1 - C_{x,y}\right)(1 - S_i)\, DD_{x,y}/DD_{max} \tag{2}$$

where $NST_{i,x,y}$ is flux from septic tanks to river/sea of nutrient i in grid square (x,y) in kg/year, S_i is proportion of nutrient i retained in septic tank settlement, $DD_{x,y}/DD_{max}$ is relative drainage density in grid square (x,y) scaled between 0 and 1.

Details of the methods and the data used are provided below. In overview, gridded population data were estimated from UK Census returns coupled with land cover mapping. Emission factors, or the amount of nutrients emitted per person, were derived from the literature and published data sources. Temporal variation in nitrogen emission factors was based on protein intake and phosphorus emission factors were based on the use of phosphorus in detergents. The historical development of the sewerage system was modelled using a logistic equation for the proportion of people connected to sewer as a function of population density, tied into published figures where available. Retention of nutrients for different methods of treatment was derived from data for wastewater treatment works (WWTWs) in England and Wales 1990-2005 and 1909-1912. Measured nutrient concentrations and dry weather flows (i.e. the measured or consented volume of effluent, not including storm discharges) were used to calculate annual effluent fluxes of nutrients for different treatment types. Representative factors for removal were then calculated as the proportion of the influent flux given by the population served and the relevant emission factor. Estimates for nutrient retention in septic tanks and for different nutrient species were based on typical compositions of wastewater. Drainage density was derived from the 1:50,000 river network of the UK and scaled to a maximum density of 2 km/km^2. This is used to represent both the proximity to a water course and the wetness of the drainage field.

Calculating nutrient emissions from human waste

Population estimates

Population estimates were derived from UK census returns and scaled to devolved authority level i.e. England/Wales/Scotland/Northern Ireland (Office of Nation Statistics (ONS), http://www.ons.gov.uk/). The population time series was constructed based on estimates for six chosen years: 1801, 1911, 1951, 1971, 1991 and 2011. These years were selected based on availability of census data and it is assumed that population changes between these years are linear. Great Britain (GB) census returns were downloaded from Vision of Britain (http://www.visionofbritain.org.uk/) at the parish level (1801, 1911 and 1951) and http://www.census.ac.uk/ at the Enumeration District (1981) and Output Area (OA) level (1991 and 2011). For 1801 and 1911, census returns were not available for all GB parishes and, for 1801 in particular, there were large areas of north-west England with missing data. Areas of missing data for each devolved authority were assumed to correspond to the deficit between the census data and the ONS population estimates. To model the population in 1971, enumeration district data from the 1981 census were used (due to availability at a higher resolution) and scaled to the 1971 devolved authority ONS population estimates. For 1991, the total population based on census returns was 10% lower than the official ONS county population estimates for this year; OAs were therefore scaled at the county level to match the ONS figures.

Spatial data relating to census returns from Northern Ireland (NI) were not available for 1801 and 1911 and the closest matches were taken from Clarkson et al. (1997), at the barony level for 1821 (to represent 1801) and at the Poor Law Union level for 1911. There were areas of missing data in the 1821 census returns, forcing the assumption that the proportion of persons in NI compared to Eire in the 1841 census (20.2 %) was the same as 1821. Similarly, in 1911, missing data for Poor Law Union areas were assumed to account for the 11.2% difference between Northern Ireland Statistics Research Agency (NISRA, http://www.nisra.gov.uk/) totals and the total of all Poor Law Union areas with data. NISRA small area population estimates for 2011 were used to provide the spatial distribution of the population; these were scaled to NISRA national population estimates for the years 1951, 1971 and 1991.

Census boundaries were not consistent throughout the population time series, therefore, population estimates were rasterized to a 1km grid resolution. For census areas that cover multiple 1km grid squares, land cover was used to distribute the population between grid squares. This was based on the current Land Cover Map 2007 (LCM2007: Morton et al., 2011) and the historical mapping of Dudley Stamp in the 1930s (Stamp, 1937). It was assumed that urban and suburban land cover types had higher population densities than other land cover types. Nominal weightings were assigned to each 1km square based on its land cover. For example, from examination of the available population data and the pattern of known urban/suburban/rural population densities, urban areas were assumed to be twice as densely populated as suburban areas and ten times denser than arable areas. Population estimates for the years 1801, 1901 and 1951 were distributed based on historical land cover, while later years are based on LCM2007. The 1km gridded population data for each time slice were then aggregated up to the 5km grid and linearly interpolated to provide a time series of 5km gridded population estimates for the whole of the UK.

Emission factors

Emission factors, the amount of nutrients emitted per person, have previously been estimated using a wide range of different methods based on either individuals, households or the quality of influent flows to WWTWs. In general, methods focused on households tend to underestimate emission factors due to the proportion of time spent outside the home while those focused on WWTWs may tend to overestimate emission factors due to receipt of industrial effluents which may include nutrients. Nearly all calculations of fluxes make use of measured concentrations and an average water usage figure which itself can be a large source of error (Friedler and Butler, 1996).

Nitrogen

Published nitrogen emission factors for the present day are in the range 3.94 to 7.67 kg/person/year with a median value of around 4.5 kg/person/year (Table 2). The high values quoted by Butler et al. (1995) are from the US where the expectation is for a high protein intake. By contrast, the very low values given by Siegrist et al. (1976) are also from the US but from a study of rural households. In this case, the flux per event is similar to that in other studies which implies that the low values simply reflect time spent away from home.

Table 2 Nutrient emission factors for human waste (excluding detergents)

Emission factor for total N kg/person/year	Emission factor for total P (excl. detergents) kg/person/year	Reference
	0.474 - 0.551	Jenkins and Lockett (1943)
	0.376 - 0.569	Devey and Harkness (1973)
1.51	0.20	Siegrist et al. (1976)
	0.438	Foy et al. (1995)
5.29, 6.12, 7.67	0.496, 0.548	quoted in Butler et al. (1995)
3.94		Johnes et al (1996)
4.56	0.548	Del Porto and Steinfeld (1999)
5.7 revised to 4.0	0.6	Tanner (2001)
4.38		Grizetti and Barraoui (2006)
4.82*		Gardner et al. (2013)

* derived from median concentrations of ammoniacal nitrogen and total oxidized nitrogen in sewage influent and assuming water use of 270 litres/person/day (median value of consented dry weather flow per population equivalent)

Nitrogen emission factors for the past can be estimated from available data on protein intake; nitrogen in urine is strongly related to protein intake with approximately 4 kg/person/year equating to a protein intake of about 70-80 g/person/day; nitrogen in faeces is about 0.5 kg/person/year (Drangert, 1998):

$$E_N = 0.0533\, prot + 0.5 \qquad (3)$$

where E_N is emission factor for nitrogen (kg/person/year) and *prot* is protein intake (g/person/day).

Historically, diet has varied considerably in the UK and a picture of changing protein intake has been compiled from Burnett (1989) for the period 1800-1940 (Table 3). The 19th century saw huge inequalities in terms of food consumption and quality of diet, with many agricultural workers having insufficient food intake. The period 1850 to 1914, in particular, saw alternating periods of affluence and great depression. Much of the period was characterised by less than adequate intake of food and poor diet; indeed actual starvation was not unknown amongst both the rural and urban poor. At the turn of the 19th century assessments by Booth in London and Rowntree in York were that about 30% population lived in poverty.

For the period 1940-2010, data on protein intake in the UK were downloaded from the Annual Reports of the National Food Survey Committee 1940-2000, the Expenditure and Food Survey 2001-02 to 2007 and Living Costs and Food Survey since 2008 (http://webarchive.nationalarchives.gov.uk/20130103014432/http://www.defra.gov.uk/statistics/foodfarm/food/familyfood/nationalfoodsurvey/). While statistics since 2000 are broken down by region and by rural/urban area, the spatial variation is much less than the historical trend and national statistics have been used throughout.

The available protein data were fitted with a smooth curve (Figure 2) and annual estimates of nitrogen emissions were calculated from equation (3). For the present-day, these estimates are broadly in agreement with the figures quoted in Table 2 from other sources.

Table 3 Data relating to historical consumption of protein (derived from Burnett, 1989)

Dates	**Sector**	**Location and other descriptors**	**Protein g/person/day**	**Reference**
1800-1834	agricultural labourers		49	Oddy, D.J. 1981 Diet in Britain during industrialisation. Leyden Colloquim The Standard of Living in Western Europe.
1841	skilled workers in employment	average Manchester Dukinfield	65 71 44-83	William Neild analysed in McKenzie, J.C. 1962 The composition and nutritional value of diets in Manchester and Dukinfield in 1841. Transactions of Lancashire and Cheshire Antiquarian Society, 72, 123-140.
1863	agricultural workers	minimum subsistence estimate average food intake	70	Dr Edward Smith First National Food inquiry on behalf of Medical Officer of Privy Council
1863	urban indoor workers	average	55	
		poorest	49	
1861	Lancashire cotton famine		84	
1862	Lancashire cotton famine		59	
1886-1902	Charles Booth survey (n=55)	London working class	61	Oddy, D.J. 1970 Working-class diets in nineteenth cenury Britain Economic History Review (2nd Series) XXIII (1,2 and 3), 319.
1901	Rowntree survey (n=20)	York - average for man in moderate work	57	
		poorest	42	
		middle class	96	
1936	Survey of 69 working class families	Newcastle	51-161	Annual Report of the Medical Officer of Health for the City and County of Newcastle upon Tyne on the Sanitary Condition of the City, 1936. Appendix A Astudy of the diet of 69 working-class families in Newcastle upon Tyne, 23 et seq
1930s	women		36	Oddy, D J. 1982 The health of the people in Baker, T. and Drake, M. (eds) Population and Society in Britain 1850-1980. page 129.
1930s	lowest group 1 (income <10s/wk)		63	Orr J. B. 1936 Food, Health and Income. Report on a Survey of Adequacy of Diet in Relation to Income. 18pp.

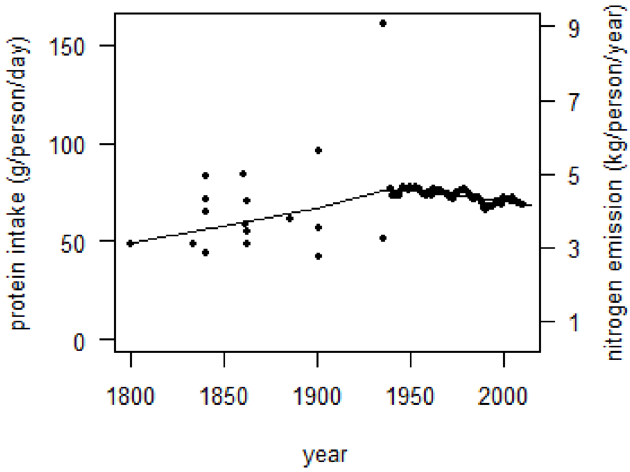

Figure 2 Available data for protein intake (points) fitted by a smooth curve using the lowess function in R; secondary scale giving the equivalent annual nitrogen emission factor derived by equation (3)

Phosphorus

Ignoring the very low value of 0.2 kgP/person/year (Siegrist et al., 1976), phosphorus emission factors for human waste fall in the range 0.376 to 0.569 kgP/person/year with a median value of 0.524 kgP/person/year (Table 2). While phosphorus in human sewage is dependent on the relative proportions of meat and vegetable protein in diet (Cordell et al., 2009), historical data relating to this are not readily available. Given that human waste is not the only source of phosphorus in wastewater, the emission factor for phosphorus was assumed to be constant over time.

A second important source of phosphorus in wastewater is from detergents. Both the use of detergents and their phosphorus content have changed substantially over time since detergents were first introduced in the late 1940s. Many countries in Western Europe and North America began to regulate the amount of phosphate in laundry detergents as early as the 1970s and it was virtually phased out in some countries using a mixture of voluntary agreements and legislation by 2000 (Litke, 1999; North et al., 2006). In the UK, there was a

40% reduction in detergent phosphorus between 1985 and 2000 (Glennie et al., 2002). More recent legislation has further restricted the amount of inorganic phosphate in laundry detergents in the UK, although there is currently no legislation for dishwasher detergents and these now comprise the larger proportion of the total phosphorus load (Richards et al., 2015).

Published values of emission factors for phosphorus from detergents have been derived using different methods: analysis of the detergent and phosphate content of wastewater influent to treatment works (Devey and Harkness, 1973; Foy et al., 1995); national estimates based on population and detergent use (e.g. Glennie et al., 2002); detailed measurements of household wastewaters (Almeida et al., 1999; Gilmour et al., 2008; Richards et al., 2015). The values derived by Devey and Harkness from analysis of wastewaters in the Birmingham area seem to be anomalously high and include different amounts of trade effluent. The remaining data were fitted with a smooth curve (Figure 3). Prior to 1975, there is some information on detergent use but the conversion of this to phosphorus emissions is problematic given the wide range of phosphorus content in washing powders and other products (cf. Devey and Harkness, 1973). Accordingly, we simply assumed an exponential increase in detergent P from near zero in 1950 to 0.525 kg/person/year in 1975, on the basis that initial uptake of new technology, in this case the washing machine, tends to be exponential in form.

When the phosphorus emission factors from detergents are combined with those from human waste, the total is consistent with published total phosphorus emissions of up to 1.16 kg/person/year (Johnes, 1996), 0.9125 kg/person/year (Grizetti and Baraaoui, 2006), 0.766 kg/person/year (Smith et al., 2005) and 0.69-0.89 kg/person/year derived from Gardner et al. (2013).

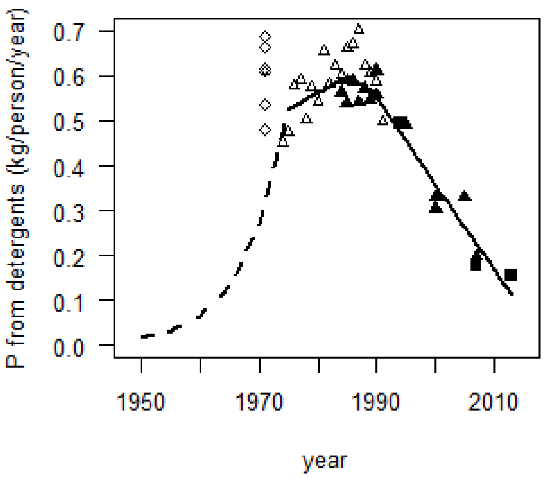

Figure 3 Emission factors for phosphorus from detergents. Data sources grouped by method: ◊ Devey and Harkness (1973); Δ Foy et al. (1995); ▲ Glennie et al. (2002), UKWIR (2008); Bouraoui et al. (2011); ■ Almeida et al. (1999), Gilmour et al. (2008), Richards et al. (2015). Smooth curve fitted to data post 1970 using the lowess function in R; dashed line shows hypothetical curve used to depict increasing use of washing machines and detergents.

A third source of phosphorus in domestic wastewater is from phosphate dosing by water companies to control the dissolution of lead pipes in older properties. This practice began in the 1980s as a response to drinking water legislation (Comber, 2011) and is widespread in the UK (CIWEM, 2011). It is estimated that around 6% total phosphorus load to WWTWs comes from dosing (Comber et al., 2013). With the reduction in P from detergents, this represents an important load in the context of modern-day sewage treatment. However, in the context of the historical estimates derived here, it represents only 0.042 kg/person/year which is very small compared to the range in published emission factors for human waste (0.376 to 0.569 kgP/person/year – see Table 2) plus the P from detergents (Figure 3). Thus, we have not explicitly included phosphate dosing in the total load.

Organic Carbon

Published data on carbon emission factors are very limited. Del Porto and Steinfeld (1999) give a figure of 10.95 kg/person/year. Organic carbon fluxes in sewage may also be estimated from other determinands such as chemical oxygen demand (COD). Chemical oxygen demand concentrations were converted to organic carbon (OC) concentrations assuming that [COD]/[OC] = 3.0. This is a rounded value, based on the expected stoichiometric ratio for carbohydrate (2.67), observed average values of 3.3 (Gardner et al., 2012) and 3.8 (Dubber & Gray, 2010), and a value of 3.0 recommended by www.wastewaterinfo.com. Fluxes were then derived using water usage figures to give estimates of 18.2 kg organic C/person/year for a typical raw municipal wastewater (Henze and Comeau, 2008; water usage 200 l/person/day) and 17.6 to 25.6 kg organic C/person/year (Gardner et al., 2013; interquartile range of influent to 16 WWTWs in the UK with average water usage 270 l/person/day).

Calculating nutrient flux from domestic wastewater to river/sea

In addition to calculating nutrient emissions, we also need to know the proportion which reaches the river system or coastal zone (Figure 1). This is a function of connection to sewer and retention during treatment. Here, we derive mathematical functions, representative factors for removal at treatment works and a set of simplified assumptions to portray the history timeline given in Table 1.

Connection to sewer

To estimate the sewage flux to water at the national scale, the connection to sewer was represented on a 5km grid. It is assumed that prior to 1830 and the rapid spread in the use of the WC, there was no *direct* loss of nutrients from sewage to watercourses; waste would have been spread on the land and incorporated into soils, much as manure is today, and given the very low levels of nutrient application, losses would have been small. Between 1830 and 1911, it is assumed that only the major urban centres had a direct connection to water. This represents the flow of raw sewage initially through the streets, and then via the newly-built interceptor sewers, to nearby watercourses. Post 1911, it is assumed that the sewer network was gradually extended to other parts of the country as a function of population density. A logistic curve was used to describe the proportion of the population in a 5km grid square directly connected to sewer:

$$C_{x,y} = \frac{\exp(a + b\ log_{10}[P_{x,y}/1000])}{1 + \exp(a + b\ log_{10}[P_{x,y}/1000])} \qquad (4)$$

where $C_{x,y}$ is proportion of population directly connected to sewer in grid square (x,y), $P_{x,y}$ is population in grid square (x,y), a and b are constants.

The parameters of the curve (a and b) were allowed to change such that, over time, there was an exponential decrease in the population density (i.e. population per 5km grid square) at which 20% population in the grid square was connected, from 55,000 per grid square in 1911 to only 600 per grid square in 2011 (Figure 4). Thus, in 1911 only the major urban centres with a population density great than 60,000 per grid square are assumed to have a direct connection to river and the different curves depict how the connection to sewer increased over time from areas with high population density to those with low population density. The curve for 2011 is consistent with the data quoted in Anthony et al. (2008) assuming 2.4 people per household. In the intervening years, the curves are in agreement with other published figures; for example, the percentage of unconnected population in 1951 was 26% (http://www.visionofbritain.org.uk/atlas/data_map_page.jsp?data_theme=&data_year=1951&u_type=MOD_DIST&data_rate=R_HOUS_AMENITY_GEN_no_wc; Kinnersley, 1988) and in 1991 5% (Rural Development Commission, 1989; Anderson, 1992).

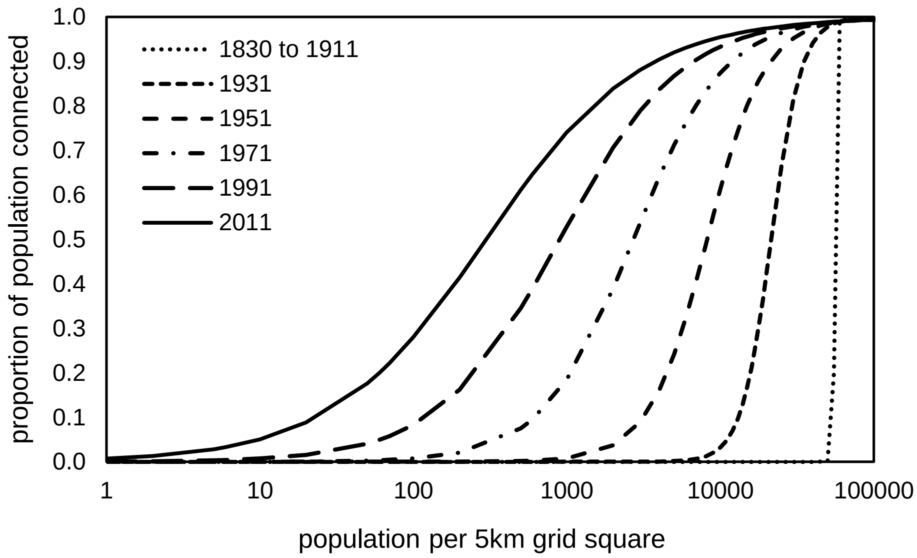

Figure 4 Estimated proportion of population in a 5km grid cell directly connected to sewer (equation 4) based on population density 1911-2011.

Nutrient retention factors in sewage treatment

The retention of nutrients during sewage treatment is specific to the design and operation of each individual WWTW. Specific historical information is very limited and so, for this national-scale study, we have derived average retention factors for each level of treatment. Data for WWTWs in England and Wales were available for the period 1990-2005. Influent loads were estimated using the resident population for each works (Keller et al., 2006; Williams et al., 2009) and the relevant annual emission factors for nitrogen and phosphorus per person given above. Effluent loads were derived from effluent water quality data (concentrations of nitrate-N, ammonium-N and orthophosphate) coupled with the consented dry weather flow provided for each works. Retention factors were derived from the average proportion of the calculated total influent load discharged. Works were grouped according to primary, secondary and tertiary treatment. Amounts of organic N, P and C were estimated from published averages (see below). There were insufficient effluent quality data for nitrate and ammonium from works with only primary treatment. However, data were available for nitrate species from 25 sewage works across England for the period 1909-1912 (HMSO,

1912) and these data were used to provide nitrogen retention factors for primary treatment. There were no phosphorus data for these early sewage works.

Nitrogen

Using the data for 1909-1912 as representative of primary treatment, the total effluent load was based on the mean measured concentrations of ammonium, nitrate and organic N multiplied by the consented dry weather flow given for each works. The data suggest that only 35% of the nitrogen load is discharged i.e. 65% is retained. The proportion of nitrate-N in the effluent has a median value of 0.32 but a large interquartile range of 0.08 to 0.42 suggesting that there is considerable variation in the efficiency of these early treatment works.

Data for 1990-2005 from between 330 and 667 WWTWs were used to provide representative nitrogen retention factors for secondary and tertiary treatment. There were no data for organic nitrogen but calculations for typical sewage compositions suggest that 95% of this will be retained (see below). Annual average values across all WWTWs suggested that, in secondary treatment, some 0.41 (median value over the 15 years analysed) of the nitrogen load is discharged as dissolved inorganic nitrogen (DIN). Of this 82% (interquartile range 81-83%) is nitrate-N. For tertiary treatment, 0.37 (median value over the 15 years analysed) of the nitrogen load is discharged as DIN of which 85% (interquartile range 83-87%) is nitrate-N.

Nitrogen retention factors based on these data are compared in Table 4 with the ranges quoted in the literature for European-wide assessments. The retention factors derived here for modern-day secondary and tertiary treatment show reasonable agreement with the ranges quoted by Kristensen et al. (2004), although they are higher than the values assumed by

Grizette and Bourai (2006). However, the retention factor of 58-73% for primary treatment derived from the 1909-1912 data is much higher than the quoted ranges. Examination of the 1909-1912 data suggests that, although there is wide variation in both the consented dry weather flow per person and the ratio of influent to effluent load, both have a relatively small inter-quartile range such that there is no obvious reason to doubt these results. The precise form of treatment in these early works is unknown. The published values in Table 4 are from reported national statistics from other European countries for present-day treatment and the basis of these statistics is also unknown. Some 25% (Henze and Comeau, 2008) of total nitrogen in human waste is in organic form and, based on calculations for typical sewage, it is estimated that 86% of organic nitrogen is retained in primary treatment. If published assessments do not take the influent organic nitrogen, much of it in solid form, into account, they will underestimate the amount of total N retained, such that retention of 20-48% may become 37-58%.

Table 4 Percentage retention on wastewater treatment

Treatment type	**% N retained on treatment**			**% P retained on treatment**			
	Grizette & Bourai (2006)	Kristensen et al. (2004)	derived in this study	Grizette & Bourai (2006)	Kristensen et al. (2004)	Gardner et al. (2013)	derived in this study
Primary (mechanical)	10-20	20-25	58-73[*]	10-30	28-30	-	12-35[c]
Primary (chemical)		20-48			86-92		
Secondary (biological)	20-40	36-55	51-69[a]	20-40	51-90	47-80[b]	53-63[c]
Tertiary	20-40	45-83	57-72[a]	85-97	88-95	70-89	54-67[c]
Advanced[#]	85-95						70-89[c]

[#] inclusion of nitrification/denitrification processes for N; inclusion of stripping for P
[*] inter-quartile range of 17 sewage works with water use 90-200 l/day in 1909-1912
[a] inter-quartile range of annual values 1990-2005 based on DIN concentrations
[b] inter-quartile range of published figures from 14 UK WWTWs
[c] inter-quartile range of annual values 1990-2005 based on orthophosphate concentrations

Phosphorus

Orthophosphate concentration data from between 771 and 2044 WWTWs 1990-2005 were used to derive retention factors for phosphorus using the same method as for nitrogen. There was no information on organic or particulate phosphorus. However, based on typical figures for raw and treated sewage, over 95% of these components is retained on treatment so it is assumed that the retention factors based on orthophosphate will approximate those for total phosphorus. Furthermore, there is a substantial contribution of inorganic detergent P in these years which means that the proportion of organic P will be much smaller than that in raw sewage (i.e. human waste). In addition to grouping works according to primary, secondary and tertiary treatment, data were available to identify those WWTWs where P-stripping had been introduced.

Annual average values across all WWTWs suggested that, taking median values over the 15 years of available data, 0.78 of the total phosphorus was discharged as orthophosphate in primary treatment, 0.42 in secondary, 0.35 in tertiary and 0.16 following P-stripping post 2000. These values were used to calculate representative retention factors for the different levels of treatment and these compare reasonably well with published values (Table 4) from European assessments and Gardner et al. (2013).

Application

Historical data on the level of sewage treatment are not generally available, although there are detailed histories of a few individual works and anecdotal evidence as to the efficiency of sewage treatment for particular time slices. Consequently, we have chosen to represent the predominant level of sewage treatment as a series of step changes. This has the advantage that the effect of changing population, connection to sewer and emission factors is readily separated out from evolving improvements in sewage treatment. In reality, there would have been a temporal variation in the level and effectiveness of treatment which would have varied spatially across the country as technology developed and as individual works became overloaded. There would have been stark differences between urban and rural areas, and between effluent discharged to river and that discharged to sea, which was often untreated even in the 1990s. The scenario we have chosen is based on the historical timeline (Table 1) and simply attempts to capture the major changes over the last two centuries. The assumptions made are as follows:

- 1800-1830 no sewage flux to river or sea
- 1830-1889 raw sewage flowed to river/sea mostly from urban areas
- 1890-1950 primary treatment of all sewage from connected population
- 1951-2000 secondary treatment of all sewage from connected population
- 2001 tertiary treatment applied in each grid square according to the national proportion of consented dry weather flow under tertiary treatment
- 2005 first tranche of P-stripping applied in each grid square according to the national proportion of consented dry weather flow with P-stripping
- 2010 second tranche of P-stripping applied in each grid square according to the national proportion of consented dry weather flow with P-stripping.

In addition to the above assumptions, one notable piece of evidence is that contained within the 1974 reports of the ten newly formed Water Authorities in England and Wales. At that time, many WWTWs were in poor repair, due to lack of investment, and were often overloaded beyond their capacity to treat effluent. Some 50% works were failing biological oxygen demand (BOD) and suspended solids consents. To represent this, it has been assumed that in 1974 the efficiency of works was only 90%; this assumption is consistent with a halving of ammonium concentrations in sewage effluent between 1974 and 1995. It was further assumed that performance declined in a linear fashion from 1951. A linear recovery to 100% efficiency in 2000 was then assumed to represent subsequent improvements in the performance of works following substantial investment since the Control of Pollution Act in 1974 and privatization of the water industry in England and Wales in 1989 (Table 1).

The retention factors derived above have also been simplified and adjusted where appropriate to give the proportions of influent N and P discharged (Table 5). In particular, it is assumed that 0.4 influent nitrogen is discharged as DIN in all treated effluent (cf. the similar values across all levels of treatment in Table 4) and that 0.67 influent phosphorus is discharged as dissolved inorganic phosphorus (DIP) following primary treatment i.e. similar to raw sewage. The reason for this latter assumption is that the proportion of DIP discharged following primary treatment given in Table 4 is derived from data for 1990-2005 when almost half the influent P was inorganic P from detergents. It is, therefore, likely to be an overestimate for years prior to the introduction of detergents. The proportion of the effluent DIN which is nitrate is also given in Table 5.

Table 5 Derived proportions of DIN, DIP and nitrate following sewage treatment

	relative to	**Raw**	**Primary**	**Secondary**	**Tertiary**	**P-stripping**
DIN	influent N	0.75	0.4	0.4	0.4	0.4
DIP	influent P	0.67	0.67	0.42	0.35	0.16
Nitrate	effluent DIN	0	0.3	0.9	0.9	0.9

We have also derived multipliers for calculating the flux of other species of nutrients: dissolved organic nitrogen (DON), dissolved organic phosphorus (DOP), dissolved organic carbon (DOC), particulate organic nitrogen (PON), particulate organic phosphorus (POP) and particulate organic carbon (POC), with particulates further subdivided into labile (L) and non-labile (NL) forms. Values for raw sewage were derived from figures reported in Henze and Comeau (2008) for a typical wastewater. Organic carbon losses and speciation across the different organic fractions of N and P were derived from COD in published compositions of treated sewage: HMSO (1912) for primary treatment and Gardner et al. (2012) for secondary treatment. It was further assumed that organic nutrients can be estimated from the effluent DIN (Table 6) which effectively means that these are realistically related to diet as well as to the level of treatment.

Table 6 Derived multipliers for calculating the flux of other nutrient species from effluent DIN following wastewater treatment

	Raw	**Primary**	**Secondary**	**Tertiary**	**P-stripping**
DON	0.133	0.041	0.029	0.029	0.029
PONL	0.133	0.041	0.005	0.005	0.005
PONNL	0.066	0.021	0.003	0.003	0.003
DOP	0.044	0.004	0.010	0.010	0.010
POPL	0.044	0.004	0.002	0.002	0.002
POPNL	0.022	0.002	0.001	0.001	0.001
DOC	2.212	0.175	0.487	0.487	0.487
POCL	2.212	0.175	0.090	0.090	0.090
POCNL	1.106	0.088	0.045	0.045	0.045

abbreviations: D dissolved; P particulate; O organic; I inorganic; L labile; NL non-labile.

Septic tanks

For completeness, we have also estimated fluxes to river/sea from septic tanks. Septic tanks were introduced in the 1890s. However, relatively few rural households had a flush toilet; even in the 1940s, 30% rural households had no running water (Kinnersley, 1988). It was, therefore, assumed that the proportion of the population not connected to sewer that had a flush toilet, and therefore a septic tank, could be estimated by:

$$p_{x,y} = \frac{\exp(-5.6 + 0.1\,Y)}{1 + \exp(-5.6 + 0.1\,Y)} \qquad (5)$$

where $p_{x,y}$ is the proportion of the population not connected to sewer in grid square (x,y) likely to have a septic tank and Y is the number of years since 1900. Thus, under these assumptions, over 99% population either had a septic tank or was connected to sewer by 1976 and 99.9% by 1995.

Based on Lowe et al. (2009), the dominant effect of a septic tank is to remove a proportion of the solid waste and convert dissolved organic nitrogen to ammonium. Total phosphorus concentration is little changed. It is, therefore, assumed that 0.84 total nitrogen, as ammonium, and all phosphorus is discharged from the tank. This is consistent with field measurements reported by Withers et al. (2011) for an English rural headwater. While issues of pipe misconnections and of septic tanks discharging directly to water courses should not be underestimated, most newer septic tanks discharge to a soak away or drainage field. We have, therefore, assumed that the proportion of discharged material that reaches a river is ~~assumed to be~~ dependent on the relative drainage density (i.e. length of river per unit area scaled using a maximum drainage density of 2km/km^2 to be between 0 and 1), which acts as a surrogate for proximity to a watercourse, soil type and the wetness of the drainage field. Multiplying the fractional losses by the relative drainage density across all UK grid squares means that the median export is 0.48 (interquartile range 0.31 to 0.64) for nitrogen and 0.56 (interquartile range 0.37 to 0.75) for phosphorus which is not inconsistent with the wide range of values quoted in the literature (Withers et al., 2011; Withers et al., 2012).

Estimating river fluxes to the sea

The above calculations were performed on a 5km grid for the whole of the UK. The gridded values were then accumulated through a representation of the river network at the 5km scale using the routing method implemented in Bell et al. (2007) to provide the total fluxes for downstream river reaches and outflows to the sea. In reality, riverine processes will change these totals and this is part of our wider study.

Results and Discussion

Historical results are presented in terms of both national totals and maps showing the spatial distribution of fluxes for historical time slices. An assessment of the approach using population grids against measured data for WWTWs in the recent period is also provided.

National figures 1800-2010

The national total nitrogen and phosphorus flux to river/sea for the UK (Figure 5) shows clearly that, for nitrogen and organic carbon, the main factors driving change have been the increase in population, from about 12 million in 1800 to 63 million in 2010, and the proportion of the population connected to sewer. In the case, of phosphorus, the use of P in detergents has also been a predominant driver.

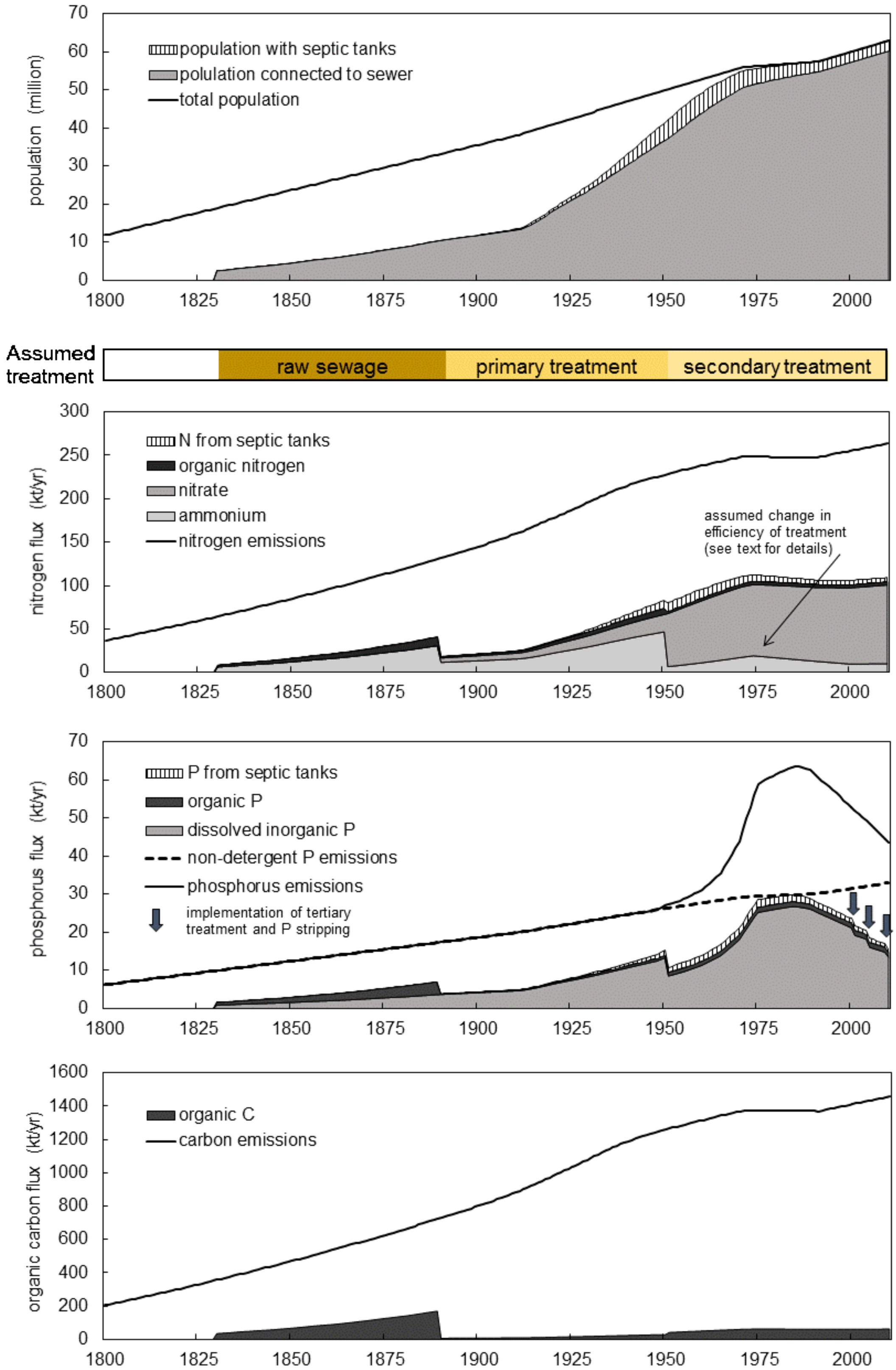

Figure 5 Estimated total nutrient flux in effluent from domestic wastewater for the UK 1800-2010 based on population (top graph). Total emissions of N, P and organic C shown by solid line; non-detergent P emissions by dashed line; amount going to river/sea by shaded area subdivided according to nutrient species. For septic tanks, the fluxes are ammonium N and dissolved inorganic P.

Up to the early 20th century, only large urban centres were connected to sewer, compared with 96% of today's population. Looking at the nitrogen fluxes, it is clear that sewage treatment goes some way to ameliorating the increase in fluxes as shown by the drop in nitrogen effluent flux in 1890 when all effluent is assumed to have primary treatment. Subsequently, the main change, however, is in the relative proportions of different nitrogen species, with raw and primary treated sewage dominated by ammonium and secondary treated sewage dominated by nitrate. The reduced efficiency in WWTWs in the mid 1970s is also clearly seen in the changing proportions of nitrate and ammonium.

With regard to phosphorus fluxes, the most important factor is the amount of detergent phosphorus emitted. This meant a peak in flux in the 1980s, with subsequent reductions being mainly due to reduced phosphorus content of detergents. The effect of tertiary treatment and P stripping at WWTWs is included but the effect of this is only apparent at the national scale in the small steps (highlighted by the arrows in Figure 5) seen in the P fluxes in 2000, 2005 and 2010. The impact of P-stripping is more appropriately seen at the local and regional scale when applied to individual WWTWs rather than as a proportion of the total effluent in each 5km grid square. At the national scale, phosphorus fluxes from septic tanks are small although, within rural areas, these can be major factors in local river water quality (e.g. Withers et al., 2011).

For organic carbon, the dominant changes prior to 1890 relate to the increase in urban population. Subsequent changes follow implementation of different methods of sewage treatment. Due to lack of data, these reflect the published values for typical compositions of treated sewage and the derived retention factors assumed in this paper. However, it is noteworthy that the highest fluxes of organic carbon were at the end of the 19th century when raw sewage was discharged into UK rivers. This also meant high levels of COD and BOD which led to low levels of oxygen and the loss of salmon and other species from UK rivers. Implementation of primary treatment reduced organic carbon fluxes considerably due to the fact that some 60% carbon in human waste is particulate compared to 17% N and 27% P (Henze and Comeau, 2008).

Changing nutrient ratios (Figure 6) are also important with respect to understanding the impact of increasing nutrient contamination and development of eutrophication. The main factors driving these changes are the method of sewage treatment and the impact of detergent phosphorus. Thus, C:N ratios have changed from just over 4 when raw sewage flowed into UK rivers to about 0.6 at the present day; the main change being at the end of the 19th century when primary treatment of sewage was introduced. The ratio of N:P has generally been about 5.6 up until 1950. Detergent P led to a minimum of around 3.6 in the 1980s but, through a combination of the reduction of P in detergents and improvements in sewage treatment, N:P ratios are now returning to levels not seen since the early 1950s.

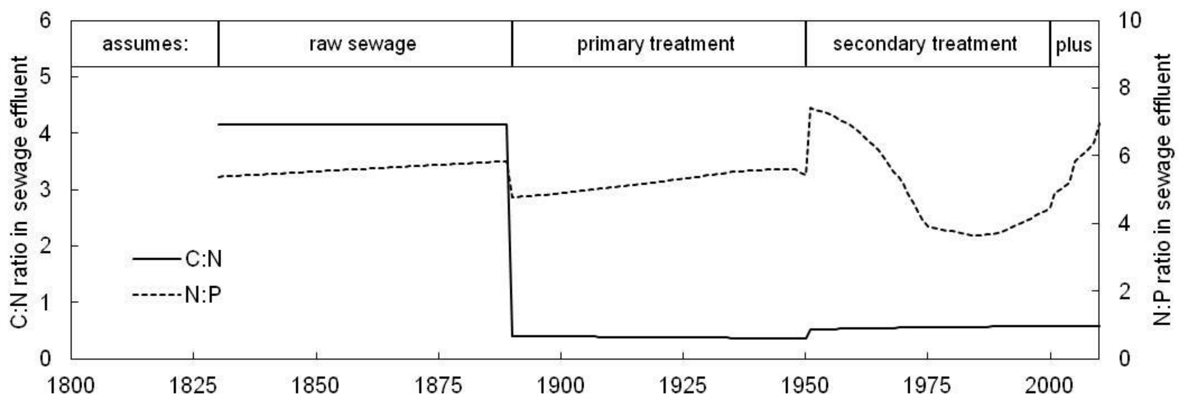

Figure 6 Estimated changes in C:N and N:P ratios in sewage fluxes to river/sea: zero flux prior to 1830; raw sewage 1830-1889; primary treatment 1890-1950; secondary treatment 1951-2000; tertiary treatment and P stripping for a proportion of effluent post 2000.

Spatial distribution of nutrient losses for historical time-slices

The 19th century saw not only large population growth but a major shift to urban areas with the ratio of rural to urban population decreasing from 80:20 in 1801 to 50:50 in 1851 and 20:80 in 1911 (Figure 7). Thus, not only was there a large increase in population but the spatial distribution shows a much more concentrated pattern by 1911. To some degree this same trend has continued and is seen in both the expansion in major centres of population and depopulation in the more remote areas of Scotland and, to a lesser extent, Northern England and Wales.

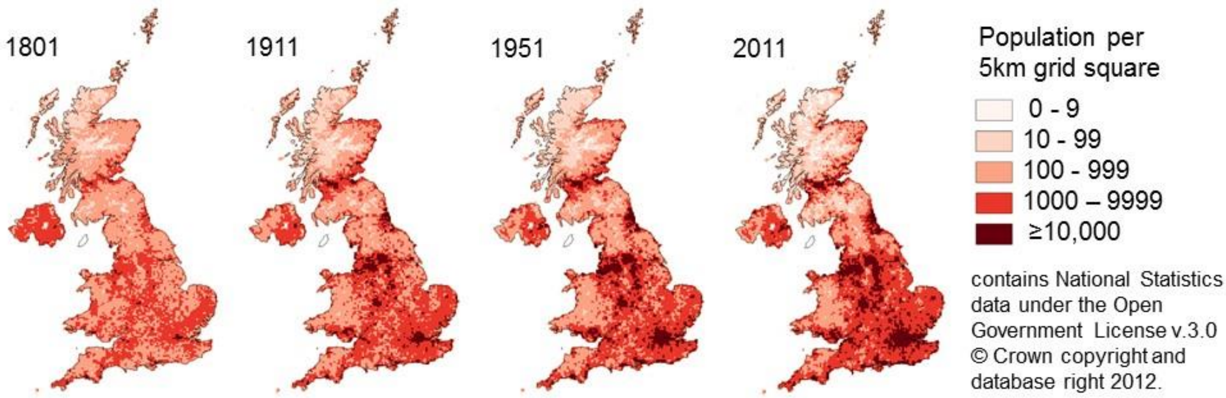

Figure 7 Population in each 5km grid square of the UK for selected years 1801-2011.

The spatial distribution of ammonium and nitrate effluent fluxes (Figure 8) reflects not only the population distribution and the increasing connection to sewer but also the change in treatment methods and their efficiency. In 1880 it was raw sewage which ran into water courses from the main urban centres and this is seen in the spatial distribution of ammonium fluxes. Primary treatment was assumed between 1890 and 1950. This converted some 30% of the ammonium to nitrate and a comparison of the maps for 1911 and 1950 essentially shows the increase in population and its connection to sewer. The maps for 1951 show the impact of secondary treatment with 90% ammonium converted to nitrate. The assumed loss of efficiency with lack of investment is seen in the maps for 1974, which show a higher flux of ammonium, with subsequent improvement to 2010.

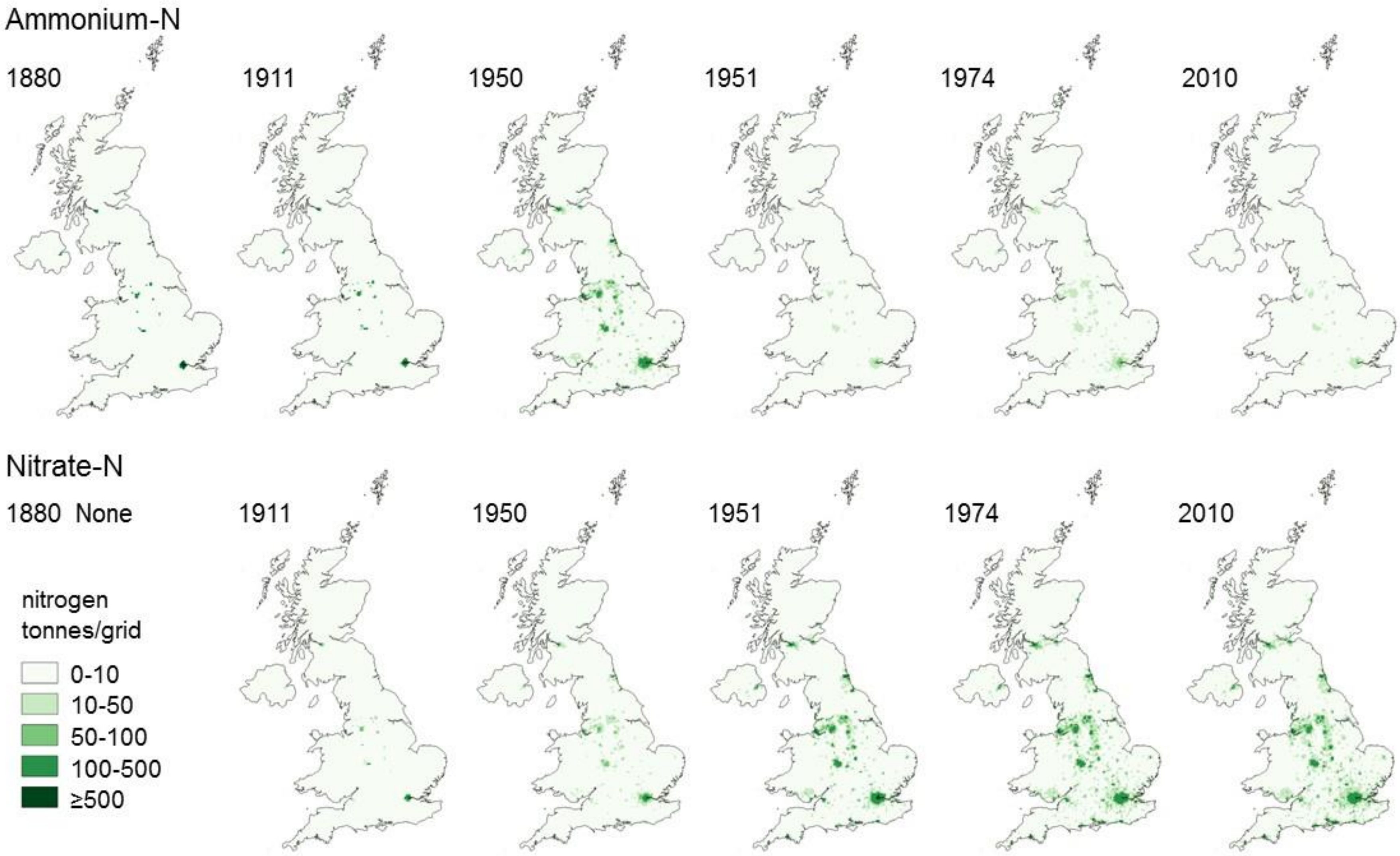

Figure 8 Estimated annual dissolved ammonium and nitrate fluxes in effluent from domestic wastewater for the UK in selected years, based on gridded population data at a 5km grid resolution.

The spatial distribution of sewage fluxes for dissolved inorganic phosphorus (Figure 9) again clearly shows the spatial concentration of fluxes due to the limited extent of sewer connections in 1911, population growth and improved sewer connections by 1951, the influence of detergent P in 1974 and the subsequent decreases in detergent P and improvements in sewage treatment by 2010.

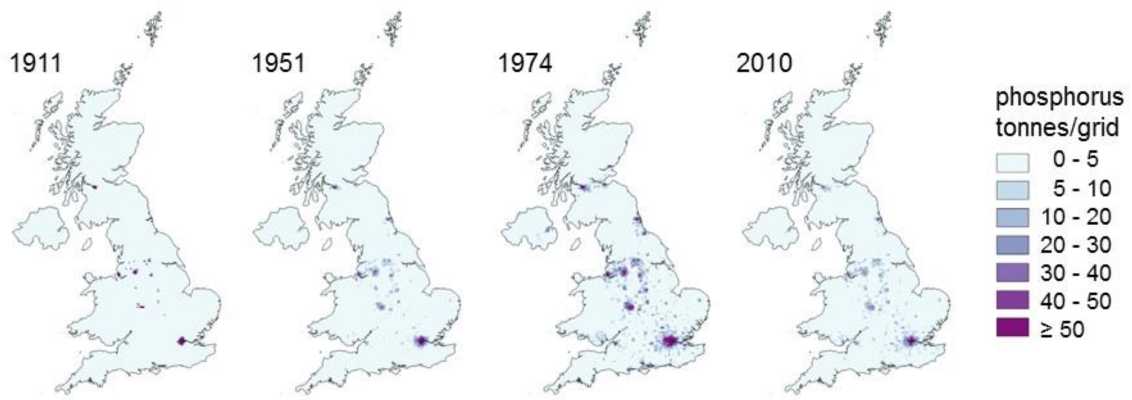

Figure 9 Estimated annual dissolved inorganic phosphorus fluxes in effluent from domestic wastewater for the UK in selected years, based on gridded population data at a 5km grid resolution.

Comparison of estimates from population to data from WWTWs for the present day

Deriving nutrient fluxes on a 5km grid based on population and assumptions relating to emission factors, sewer connection and retention in sewage treatment is the only realistic approach to reconstructing past nutrient fluxes from domestic wastewater. This is due to the lack of data, at the national scale, relating to the location of specific works, level of treatment, volume and quality of effluent. However, data do exist for the present day and it is worth comparing the two different estimates. This has been carried out for the gridded nitrate fluxes for the year 2000 as an example. Measured data for mean annual nitrate concentration and dry weather flows were used to calculate the annual flux from each WWTW. These were then aggregated at the 5km grid scale based on the location of sewage outfalls. Where water quality data were not available, these were infilled by the median nitrate concentration across all WWTWs; where dry weather flows were not available, these were infilled based on the population served multiplied by the average water use figure across all works. The simple comparison of grid square estimates shows that, at the 5km grid scale, agreement between the two methods is relatively low (Figure 10a). This is likely due to the fact that the outfalls of sewage works are not co-located with the local population – particularly in the case of large works serving major cities. Indeed, the main purpose of the original interceptor sewers in London was to move the sewage downstream and away from the city (Halliday, 2009). With the exception of large cities, it is expected that sewage works may have been more co-located with population in the past when there were more small treatment works e.g. figures for individual water authorities in England and Wales from 1974 suggest that some 92-94% works served populations less than 10,000 compared to an overall figure of 89% today.

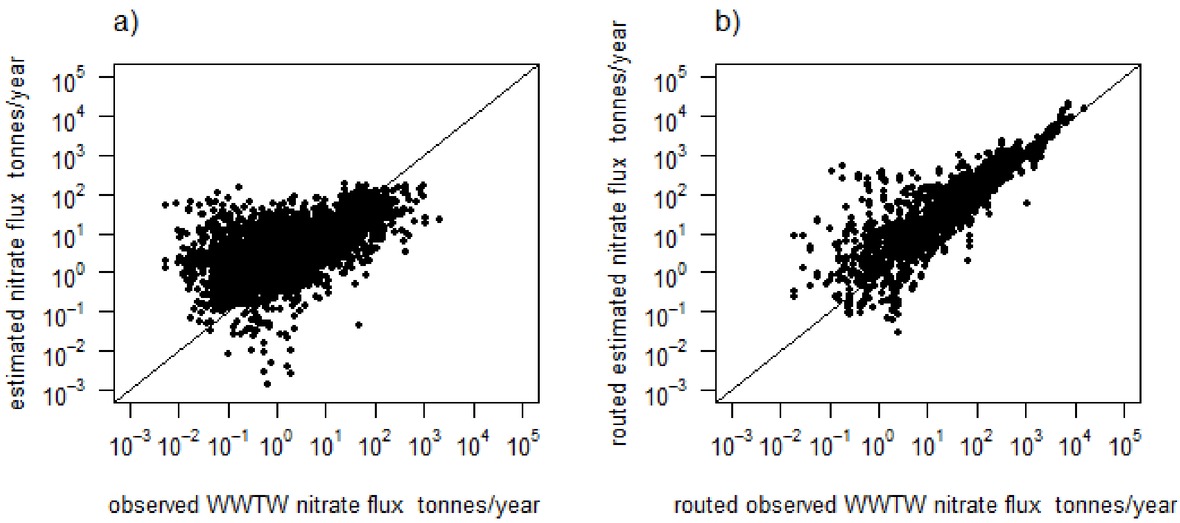

Figure 10 Comparison of methods for estimating nitrate fluxes from WWTWs in England and Wales for the year 2000: a) raw values at the 5km grid scale; b) cumulated values for river catchments with a minimum area of 100km^2

A fairer comparison of the estimated and measured sewage flux is, therefore, to look at the fluxes cumulated for downstream river reaches with larger catchment areas, rather than the individual gridded values. To this end, a 5km representation of the river network in the UK was used to cumulate, or route, the nutrient fluxes from source to sea. A comparison of the routed sewage fluxes for catchments with an area of over 100km^2 (Figure 10b) suggests that the problem of co-location is greatly reduced after routing and this provides some confidence in the method for estimating not only national totals but also catchment-based fluxes to the sea. The spatial distribution of the routed nitrate and dissolved inorganic phosphorus fluxes from domestic wastewater (Figure 11) shows clearly the accumulation of these fluxes down the major river systems of the UK. It also shows the relative importance of each of these rivers in terms of the nutrient flux from wastewater to the coastal zone. Modification of these fluxes through river and lake processes is being addressed as part of our wider integrated modelling study.

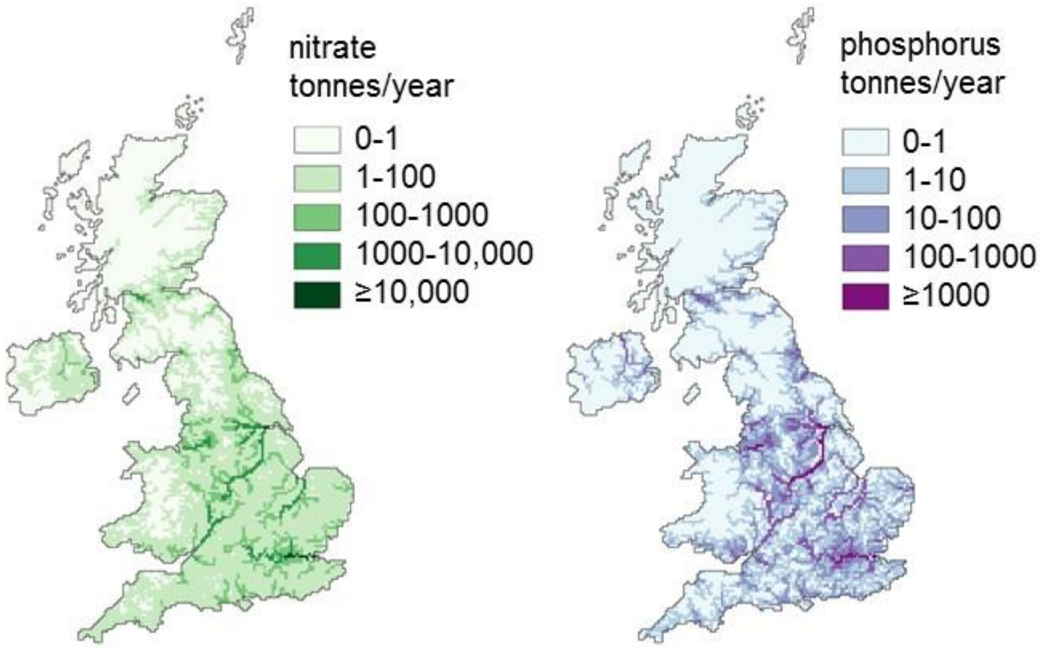

Figure 11 Nutrient fluxes of nitrate and dissolved inorganic phosphorus in effluent from domestic wastewater for the year 2000 as routed through a 5km representation of the river system. Estimates are based on population and the representative emission and treatment factors derived in this study; the hydrological boundary for catchments in Northern Ireland is shown.

Uncertainty in the estimates

Inevitably, the reconstruction of historical estimates of both nutrient emissions and effluent fluxes is hugely uncertain. The figures presented here are based on readily available evidence and are simply designed to provide a historical perspective on current nutrient emissions from human waste and the resulting effluent fluxes to rivers and coastal waters. Given the different data sources and levels of information, a formal analysis of uncertainty is not possible but some idea of the error in the estimates can be provided. This also indicates how additional data could usefully improve the estimated values.

At the national scale, looking at equations (1) and (2), we can regard the population derived from the national census as highly accurate. However, interpolation between the census dates used will bring in some uncertainty; this is particularly the case for the period 1801-1911. Table 7 summarizes the uncertainty in the emission factors and the proportion of nutrients retained on treatment, indicating additional assumptions and omissions. The focus is on the estimates for inorganic nitrogen and phosphorus; estimates for organic carbon and other nutrient species are based on average compositions of raw and treated effluent and should, therefore, be treated with caution. Combining the uncertainty in both emission and retention factors, the overall uncertainty in the estimates will be of the order of 20-30%. Clearly, the further back in time, the higher is the degree of uncertainty. For example, we have simply assumed plausible functions for the population connected to sewer and for the increase in P emissions from detergents between 1948 and 1975. Furthermore, due to a lack of national-scale information, we also chose to take a scenario approach to the implemented level of sewage treatment. This means that we have not taken into account the actual proportion of effluent treatment using different methods, with the exception of the implementation of tertiary treatment and phosphorus stripping post 2000. Prior to this, it is likely that the scenario approach underestimates the effluent flux between 1950 and 2000 as, for example, in the mid 1990s about 17% sewage effluent was discharged through marine outfalls with little or no treatment. Conversely, the effluent flux in the period 1914-1950 may be overestimated as it is over this period that new activated sludge treatment plants were brought into operation but the very high nitrogen retention factors, found for primary treatment from the 1909-1912 data for sewage works and used over this period, may counter this.

Table 7 Summary of uncertainty in the estimates

Factor	**Known ranges of uncertainty**[#]	**Unknown errors, assumptions and omissions**
Emission factor for N	Interquartile range of published figures imply error of -12 to +16%	Unknown error in conversion of protein to N (equation 3) Unknown error in dietary protein statistics (Figure 2)
Emission factor for P	Interquartile range of published figures imply error of -10 to +5%; -11 to +15% in detergent P	Effect of diet omitted: a vegetarian diet may halve P emissions (Cordell et al., 2009) Phosphate dosing since 1980 omitted: may underestimate current P emissions by 6% (Comber et al., 2013)
Proportion of N retained in treatment	Primary: -3 to +23% Secondary: ±15% Tertiary: -5 to +20%	Scenario approach to treatment level and efficiency; does not account for mix of treatment levels. Retention factor for primary treatment is from 1909-1912 data.
Proportion of P retained in treatment	Primary: -63% to 6% Secondary: ±8% Tertiary: -16% to 3% P stripping: -16% to 6%	Scenario approach to treatment level and efficiency; does not account for mix of treatment levels

[#] based on quartile values expressed as a percentage of the median or value used (Table 5)

Uncertainty also increases as the resolution of the estimates is reduced. This was brought out in Figure 10 in relation to the 5km grid scale and accumulated fluxes for catchments ≥ 100 km^2. The issue here is largely the lack of co-location between population and sewage outfalls. However, at these finer scales, and particularly in the earlier period, there is also some uncertainty in the spatial distribution of the population as census data are only available at the parish level. Furthermore, emission factors, level of sewage treatment and its efficiency have all been applied as national-scale constants and, therefore, do not represent specific local conditions.

Conclusions

Quantitative reconstruction of past nutrient fluxes relies on trying to piece together sparse data and anecdotal evidence in order to provide sensible scenarios of change. Many of the techniques described here for quantifying these changes will be generally applicable elsewhere. Based on the assumptions made in this paper and UK population census data, the main conclusions with regard to historical nutrient fluxes from domestic wastewater are:

- In 1800 estimated emissions in human waste were around 37 kt N, 6.2 kt P and 205 kt organic C per year. This would have been recycled to the land with little or no sewage going directly to rivers, lakes or coastal waters.
- The most important drivers of change since 1800 have been the introduction of the WC, or flush toilet, along with population growth, urbanization, connection to sewer, improvements in wastewater treatment and the use of phosphorus in detergents.
- In 1900, an estimated 145 kt N were emitted in human waste but only about 19 kt N were discharged in sewage effluent. Of this, about 27% was nitrate, 63% ammonium and 10% organic nitrogen. By 2000, estimated emissions had risen to 255 kt N, with 97 kt N being discharged, of which 90% was nitrate.
- In the case of phosphorus, an estimated 18.6 kt P were emitted in human waste in 1900 but only 4.3 kt P were discharged in effluent, of which 96% was dissolved inorganic phosphorus. Estimated phosphorus emissions peaked at around 63.5 kt P/year in the 1980s due to large increases in the population connected to sewer and the use of P in detergents. Some 28 kt P/year were estimated to be discharged in sewage effluent. By

2010, following the reduction of phosphates in detergents and improvements in sewage treatment, estimated emissions were down to 43.6 kt P/year with 14.8 kt P/year being discharged in effluent.

- For organic carbon, the highest effluent fluxes (estimated to be ca. 170 kt organic C/year) were towards the end of the 19^{th} century when raw sewage was discharged directly into UK rivers and seas. The introduction of primary sewage treatment led to a dramatic reduction in organic carbon fluxes to an estimated 8 kt C/year in 1900. Estimated organic carbon fluxes have subsequently increased with the increasing population connected to sewer and, assuming typical compositions for treated wastewater, the estimated effluent flux for 2010 is 62 kt organic C /year.

Acknowledgements

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The gridded population estimates (1801-1951) created for use here are based on data provided through www.VisionofBritain.org.uk and uses historical material which has been re-districted by the Linking Censuses through Time system, created as part of ESRC Award H507255151, by Danny Dorling, David Martin and Richard Mitchell.

The gridded population estimates (1971-2011) created for use here are based on data provided with the support of the ESRC and JISC and uses boundary material which is copyright of the Crown and the ED-LINE Consortium data provided with the support of the ESRC and JISC and uses boundary material which is copyright of the Crown and the ED-LINE Consortium.

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