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Exploring global nitrogen and phosphorus flows in urban wastes during the twentieth century

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[1] This paper presents a global model-based country-scale quantification of urban N and P mass flows from humans, animals, and industries and their waste N and P discharges to surface water and urban waste recycling in agriculture. Agricultural recycling was practiced commonly in early twentieth century Europe, Asia, and North America. During the twentieth century, global urban discharge to surface water increased ~3.5-fold to 7.7 Tg yr^{-1} for N and ~4.5-fold to 1.0 Tg yr^{-1} for P; the major part of this increase occurred between 1950 and 2000. Between 1900 and ~1940, industrial N and P flows dominated global surface water N and P loadings from urban areas; since ~1940, human wastes are the major source of urban nutrient discharge to both surface water and agricultural recycling. During the period 1900–2000, total global recycling of urban nutrients in agriculture increased from 0.4 to 0.6 Tg N yr^{-1} and from 0.07 to 0.08 Tg P yr^{-1}. A large number of factors (the major ones related to food consumption, urban population, sewer connection, and industrial emissions) contribute to the uncertainty of −18% to +42% for N and −21% to +45% for P around the calculated surface water loading estimate for 2000.

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1. Introduction

[2] Although urban areas cover only a fraction of the global land area, urban activities dramatically alter regional and global nitrogen (N) and phosphorus (P) cycles [*Svirejeva-Hopkins et al.*, 2011; *Van Drecht et al.*, 2009]. Urban wastewater is emitted as point sources with large nutrient loads from excreta from humans and animals, P-based detergents, and waste generated by industries [*Balmer and Hultman*, 1988; *Billen et al.*, 1999; *Billen et al.*, 2012; *Li et al.*, 2012; *Nyenje et al.*, 2010; *Quynh et al.*, 2005]. Since preindustrial times, river N export to the North Atlantic Ocean increased by a factor 2 to 20 [*Howarth et al.*, 1996] as a result of increasing diffuse agricultural [*Howarth et al.*, 1996; *Liu et al.*, 2012; *Némery et al.*, 2005; *Quynh et al.*, 2010] and urban point-source emissions [*Barles and Lestel*, 2007; *Billen et al.*, 1999; *Van Drecht et al.*, 2009].

[3] Reported effects of excessive nutrient discharge to aquatic ecosystems include eutrophication, oxygen depletion (hypoxia), fish kills, and harmful algal blooms (HABs). Human-induced eutrophication effects have been reported since the early twentieth century in Europe (Norway and England), South Korea, North America, and Mexico [*Lewitus et al.*, 2012; *Rabalais et al.*, 2010; *Zhang et al.*, 2010]. Paralytic shellfish poisoning, caused by some HAB species, has been reported as early as 1793 [*Hallegraeff*, 2003; *Lewitus et al.*, 2012]. Hypoxia events form a growing global problem since the late 1950s [*Rabalais et al.*, 2010; *Zhang et al.*, 2010]. N and P accumulation in urban soils and groundwater form an additional problem [*Kennedy et al.*, 2007; *Li et al.*, 2012; *Qiao et al.*, 2011].

[4] The environmental impacts of urban areas increased during the twentieth century due to rapid population growth, urbanization, and sewer construction. While during the twentieth century the global total population increased by a factor of almost 4, the urban population increased by a factor of close to 11 [*Klein Goldewijk et al.*, 2010] (SI 1). Since 2007, more than half of the global population lives in urban areas, and projections for 2050 suggest a growth of 70% [*UN*, 2006]. This development will inevitably contribute to increasing urban N and P loading of surface water.

[5] As opposed to diffuse agricultural nutrient emissions, nutrients can be partly removed from urban wastewaters if sewer connection and adequate treatment facilities are present [*Balmer and Hultman*, 1988; *Rabalais et al.*, 2010]. However, currently, only few countries have adequate treatment facilities [*UNEP*, 2010], and negative effects of surface water eutrophication increased during the last few decades, especially in densely populated areas [*Hallegraeff*, 2003; *Lewitus et al.*, 2012; *Zhang et al.*, 2010].

[6] The development of hypoxia often occurs after a threshold value of cumulative nutrient inputs is exceeded [*Rabalais et al.*, 2010; *Zhang et al.*, 2010]. It is therefore important to understand and quantify cumulative loading of surface water

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with N and P. Although the local history of urban waste generation and handling may be different, there are some general patterns. The recycling of urban waste as a source of nutrients in agriculture was common in many industrialized countries in the nineteenth century. Since the late nineteenth century, urban nutrients were increasingly discharged to surface water due to the successive introduction of water supply networks, water closets, and sewage systems. This transition reduced the availability of nutrients for recycling in agriculture [*Cordell et al.*, 2009; *Van Zon*, 1986] and caused surface water eutrophication and health problems due to the release of coliform bacteria [*Burian et al.*, 2000; *Heinonen-Tanski et al.*, 2010; *Hofstra et al.*, 2013]. Although the first modern treatment plants were constructed since ~1920, wastewater treatment plants became important only after the 1970s in Europe. Construction of treatment plants often lags the construction of sewage systems by several decennia in many industrialized and developing countries [*Van Drecht et al.*, 2009].

[7] The processes of agricultural and aquacultural (after 1970) intensification, urbanization, sewer construction and wastewater treatment have had different effects in river water; this is because each source has a typical signature in terms of nutrient forms and ratios [*Seitzinger et al.*, 2010], which is important for the development of eutrophication and the response of aquatic ecosystems [*Kemp et al.*, 2009]. To our knowledge, no quantification of historical global urban nutrient flows has been made so far. This paper presents a global, model-based country-scale inventory of urban nutrient flows in wastewater covering the full twentieth century. Since many aspects of urban nutrient cycles are poorly known, a thorough uncertainty analysis is included. Readers are cautioned against interpreting this paper as an exhaustive review or historical analysis. Instead, historical statistical data are combined with (often anecdotal) information from historical literature.

[8] Section 2 presents methods and data, followed by the results and discussion (section 3) and conclusions (section 4). Supporting information (SI) is available online.

2. Methods and Data

2.1. General Model Description

[9] Starting point of the country-scale calculations is the gross urban N and P flow, which is the sum of nutrients from human excreta and P-based detergents, animal excreta, and industrial wastes. For each of these sources, we consider three possible fates of N and P, i.e., (i) surface water, (ii) agricultural land and soil, and (iii) "other," including groundwater, nonagricultural soils, and atmosphere (Figures 1a, 1b, and 1c). A model was developed to quantify the N and P mass flows in the urban system for each year and country. Results are spatially distributed using population density maps (section 2.6). For the results and discussion in this paper, data are presented for 10 world regions (SI Figure 2). A list of all model input parameters with a description, value and units is provided in SI Table 1.

2.2. Human Waste

2.2.1. Gross Human N and P Flows

[10] Human urban gross N and P flows consist of N and P in human excreta and detergent P. N in human excreta was estimated based on protein consumption. FAO provides country data on retail stage per capita protein consumption for most countries for the period 1961-2000 [*FAO*, 2012] and estimates for the 1930s and 1950s for ~15% of all countries [*FAO*, 1949, 1951, 1955, 1957, 1958, 1963]. These data were supplemented with various data sources [*Boomgaard and Van Zanden*, 1990; *CBS*, 2001; *FAO/WHO*, 1965; *Grigg*, 1995; *Schmid Neset*, 2004; *Segura*, 2005; *State Statistical Bureau*, 1986; *USDA*, 2012] (SI 2). To create a full data set for the twentieth century, we assumed that periods without data could be linearly interpolated between the nearest years with data. Protein consumption in the first decades of the twentieth century was close to midcentury protein consumption for most countries with data [*Boomgaard and Van Zanden*, 1990; *Schmid Neset*, 2004; *Segura*, 2005]. For countries lacking early twentieth century data, we therefore assumed that protein intake in 1900 was equal to that in the nearest year with available data for the country considered. For countries with no data for any year, protein intake was assumed to be equal to the population-weighted average for the corresponding world region (SI Figure 4).

[11] The data collected represent consumption at the retail stage and need to be corrected for retail and household losses to obtain the actual protein consumption and excretion. Retail and household losses of protein were estimated at a regional scale based on *SIK/FAO* [2011], and values in 1900 were assumed to equal the lowest regional loss percentage of 2010 (SI 3). This agrees with literature for the U.K. [*Cathcart and Murray*, 1939] and the general pattern of increasing household and retail food waste losses as incomes grow [*SIK/FAO*, 2011]. The country average per capita protein consumption was directly applied to urban inhabitants, which is in agreement with observed small differences between rural and urban protein consumption [*Liu et al.*, 2009; *Liu*, 2005; *Qiao et al.*, 2011; *Shehu et al.*, 2010].

[12] We used an N content in protein of 0.16 [*Block and Bolling*, 1946]. P consumption was obtained from N consumption using a N:P ratio of 10:1 (mass basis) (SI 4). This is based on a near complete twentieth century N and P consumption data set for the United States [*USDA*, 2012] and supported by a rather fixed N:P ratio in dietary intakes surveys in the range of 10.6:1 (adults) [*Sette et al.*, 2011] to 11.1:1 (adolescent males) [*Turan et al.*, 2009] and other estimates [*Billen et al.*, 2009].

[13] Excretion occurs in the form of urine (80% of intake for N, 62% for P) and feces (17% for N, 35% for P); 3% of N and P intake is lost via sweat, hair, and blood [*Calloway and Margen*, 1971; *Chittenden*, 1904; *Kimura et al.*, 2005; *Langergraber and Muellegger*, 2005; *Oddoye and Margen*, 1979; *Schmid Neset et al.*, 2006; *Takahashi et al.*, 1985].

[14] P-based detergent use for the period 1970–2000 was taken from the study by *Van Drecht et al.* [2009]. Assuming that the use of P-based detergents started in 1950, for all countries a linear interpolation was made between 1950 and 1970. This is based on the start of large-scale introduction of laundry machines around the 1950s in many industrialized countries [*Billen et al.*, 1999; *Han et al.*, 2011; *Hukka et al.*, 2010].

2.2.2. Fate of Human N and P Waste

[15] All food losses and all losses through non-feces and non-urine pathways were assumed to end up in the "other" sink (Figure 1a). The fate of N and P in human excreta depends on the presence of a sewer connection (SI Figure 6); P-based detergents were assumed to be used exclusively by households with a sewer connection [*Van Drecht et al.*, 2009]. The

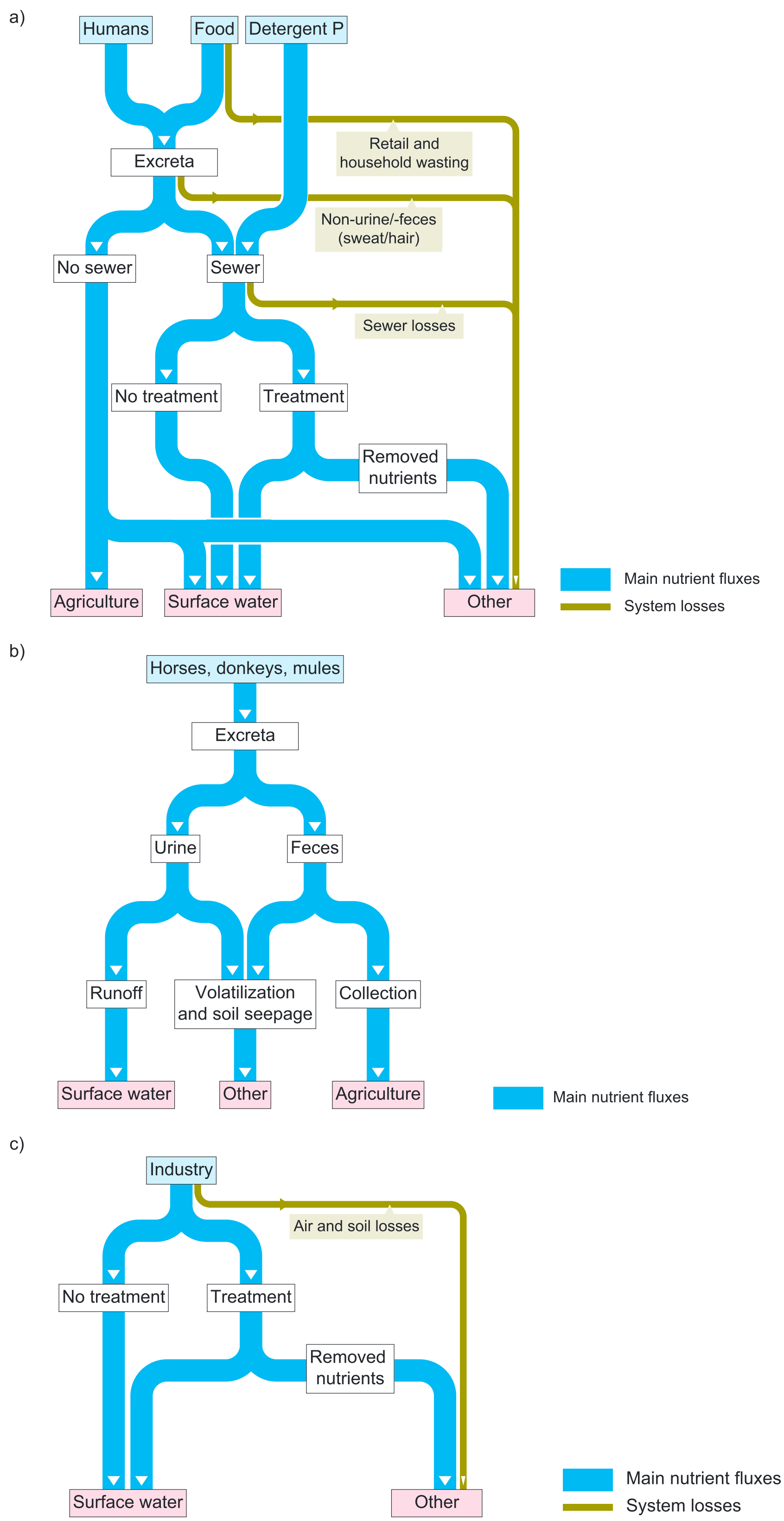

Figure 1. Flow diagram representing the model outline for N and P mass flows from (a) urban human excreta and P-based detergents; (b) horses, donkeys and mules; and (c) industries to surface water, agriculture, and "other."

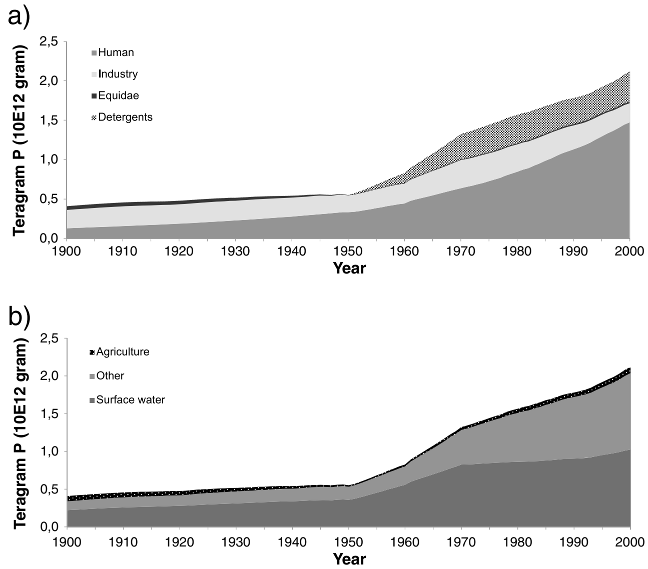

Figure 2. (a) Mass of the global gross P flow from human excreta, P-based detergents, animal excreta, and industries. (b) Mass of global P discharge after all corrections (Figures 1a, 1b, and 1c) to surface water, recycling in agriculture and "other." Unit: Tg P yr^{-1}.

fraction of inhabitants with a sewer connection often increased rapidly when cities started to construct sewer networks. Well-documented examples include the city of Wagga Wagga (Australia) (1930–1970 exponential increase) [*Burn et al.*, 1999], Helsinki (Finland) (1950–1980 steep increase) [*Laakkonen and Lehtonen*, 1999], and Paris (France) (32% connection in 1900 and 70% in 1914) [*Barles*, 2007; *Barles and Lestel*, 2007].

[16] For the period between 1970 and 2000, sewer connection data from *Van Drecht et al.* [2009] were used. Since quantitative global country data on sewer connection and timing of rapid sewer construction during the period 1900–1970 are scant, sewage systems in industrialized countries were assumed to be constructed on a large scale from the year 1870 onward. This agrees with the development in many cities worldwide [*Mokyr*, 1998] such as Paris [*Barles*, 2007; *Barles and Lestel*, 2007], London, San Francisco/United States [*Burian et al.*, 2000; *Schultz and McShane*, 1978; *Smith*, 2007], and multiple cities in the Netherlands [*Lohuizen*, 2006; *Van Zon*, 1986]. The separation between developing countries and industrialized countries was made based on the study by *Ott et al.* [2004]. For developing countries, we assumed that construction of sewage systems in cities began 50 years later (1920) based on historical estimates for sub-Saharan Africa [*Nilsson*, 2006; *Nyenje et al.*, 2010] and Egypt [*Roberts and Flaxman*, 1985]. For the periods 1870–1970 (industrialized) and 1920–1970 (developing), we assumed a linear increase in the fraction of the urban population connected to a sewer. At present, large variations in human sewer connection exist between countries [*WHO/UNICEF*, 2000], with lower (but increasing) degree of sewer connection in developing countries (SI Figure 6). The definition of urban population was extended in those cases where urban population was smaller than the number of people with a sewer connection to incorporate all sewer connected persons in the model.

[17] Sanitation coverage is generally much further developed in urban than in rural areas. For example, rural human excreta collected in (primitive) tanks are often dumped in surface water without treatment [*UNEP*, 2010], open defecation is still common in rural areas of the world such as in some African countries [*Erni et al.*, 2010], and open drainage systems discharge excreta from both animals and humans to surface water in many densely populated regions of the world. Estimated sewer leakage, biological degradation, nutrient particle settlement, and volatilization processes are 10% for both N and P [*Nyenje et al.*, 2010; *Rutsch*, 2006; *Wakida and Lerner*, 2005] (SI 5).

[18] In the model, three treatment classes were distinguished based on the study by *Van Drecht et al.* [2009], i.e., primary treatment (10% N and P removal), secondary treatment (35% N and 45% P removal), and tertiary treatment (80% N and 90% P removal). Treatment fractions were linearly interpolated to 1970 [*Van Drecht et al.*, 2009], assuming large scale primary treatment to start in 1920 [*Barles and Lestel*, 2007; *Gadegast et al.*, 2012; *Melosi*, 2000; *Shuval*, 1986] and secondary and tertiary treatment to start in 1950 [*Brosnan and O'Shea*, 1996; *Cooper*, 2001; *Liu et al.*, 2008; *Ogoshi et al.*, 2001; *Schmid Neset et al.*, 2010] (SI 6). All nutrients removed were assumed to flow to the "other" sink because of the large variety in the handling of treatment sludge [*Burian et al.*, 2000; *Van Zon*, 1986]. The remainder of nutrients (i.e. wastewater that is not treated at all or nutrients that are not removed) present in sewers was assumed to be discharged to surface water [*UNEP*, 2010; *Van Drecht et al.*, 2009; *Van Zon*, 1986] (Figure 1a).

[19] Possible fates for nonsewered human N and P are surface water discharge, agriculture, or "other." We assumed that 20% of N from human excreta from inhabitants lacking a sewage connection is lost as ammonia ("other") (SI 5) [*Huang et al.*, 2012; *Kimura et al.*, 2005]. The collection of human excreta from urban areas for use in agriculture was common in the early twentieth century, most substantially in Europe, Asia, and North America [*Barles*, 2007; *Liu et al.*, 2008; *Noort*, 1999; *Shuval*, 1986; *Van Zon*, 1986]. Only few

quantitative studies are available on recycling practices in the early twentieth century. A good example is Paris, which has been reported to reuse most of its waste in sewage farms in the early twentieth century [*Shuval*, 1986].

[20] Agricultural recycling was quantified for 1900 based on four recycling classes: none (0% recycling), low (10%), medium (40%), and high (70%) (SI 7 and SI Figure 8). Countries or regions where recycling was important in 1900 include West Europe [*Barles and Lestel*, 2007; *Svirejeva-Hopkins et al.*, 2011; *Van Zon*, 1986], China, India, and Japan [*Drechsel et al.*, 2010; *Shuval*, 1986; *WHO*, 1989; *Xue*, 1961]. Countries with no or negligible recycling include, among others, Islamic countries and most African countries, where religious taboos limit handling and recycling of human excreta [*Shuval*, 1986]. Medium-recycling regions include the United States [*Tzanakakis et al.*, 2006], Mexico, Central Europe, and South-East Asia [*Shuval*, 1986]. Low-recycling countries include those for which no literature on recycling was found and no taboos were expected but which are close to neighboring medium- or high-recycling countries (Canada, South America, and Russia).

[21] Over the course of the twentieth century, a decline in agricultural nutrient recycling resulted from (i) the introduction of the water closet in industrialized countries, thereby diluting wastewaters; (ii) the need for higher hygiene standards; (iii) the introduction of treatment facilities; and (iv) the application of cheap chemical fertilizers [*Billen et al.*, 1995; *Schmid Neset*, 2005; *Schmid Neset et al.*, 2010]. In the model, this development is mimicked by a linear decrease of the global fraction of recycled human waste from nonsewered populations between 1900 and 1950 to a value of 15% of the 1900 value. This trend is in agreement with qualitative information from various countries [*Angelakisa and Durhamb*, 2008; *Cooper*, 2001; *Mokyr*, 1998; *Schladweiler*, 2012; *Tzanakakis et al.*, 2006; *Van Zon*, 1986] and data for Sweden [*Schmid Neset*, 2005].

[22] In the last few decades of the twentieth century, a renewed interest developed in the recycling of urban wastes including human excreta [*IWMI and FAO*, 2001; *Shuval*, 1986] in both industrialized [*Angelakisa and Durhamb*, 2008; *Schmid Neset et al.*, 2010; *Tzanakakis et al.*, 2006] and developing countries [*Hayashi et al.*, 2012; *Hofstedt*, 2005]. Countries generally increased the amount of agricultural recycling to (i) reduce wastewater discharge, or use wastewater for irrigation in dry regions [*Ogoshi et al.*, 2001; *Olson*, 1987]; (ii) avoid landfill and regular treatment costs; and (iii) close nutrient cycles for sustainable nutrient use and food production [*Angelakisa and Durhamb*, 2008; *Bixio et al.*, 2005; *IWMI and FAO*, 2001; *Tzanakakis et al.*, 2006]. In the model, this trend was described for countries with no recycling in 1950 by simulating an increase of the fraction of nonsewered human excreta being recycled to 20% in 1990 [*Hayashi et al.*, 2012; *Hofstedt*, 2005; *Liu et al.*, 2008] and no change between 1950 and 1990 for all other countries. For all countries, between 1990 and 2000, a linear increase by 20% of the 1990 value was incorporated in the model to simulate the most recent increase in recycling [*IWMI and FAO*, 2001].

[23] The remainder of the nonsewered human excreta ends in (i) surface water, representing the processes of surface runoff of wastes and dumping of collected excreta in open water as reported for both historical and present times [*Noort*, 1999; *UNEP*, 2010; *Van Zon*, 1986], and (ii) in "other", representing leakage and seepage to soils, waste transport losses and losses through leaky cesspits, and tons and pit latrines [*Jacks et al.*, 1999; *Van Zon*, 1986] (Figure 1a).

2.3. Animal Waste

2.3.1. Gross Animal N and P Flows

[24] Animal traction was an important aspect of early twentieth century cities, and large livestock such as horses, donkeys, and mules (equidae) were kept in cities in many countries [*Barles*, 2007; *Mokyr*, 1998]. At present, many developing countries still have large urban animal stocks [*FAO*, 2001]. Here we only consider equidae used for animal traction because their excreta are generally dropped in streets and city stables and can potentially end in surface water or be collected for use in agriculture. Due to lack of data, we ignored other animals present in urban areas such as cows, dogs, cats, poultry, buffaloes, camels, sheep, pigs, and goats [*FAO*, 2001; *Schiere*, 2012] (SI 8.1).

[25] Nutrients in waste from equidae were calculated using historical country data on stocks of horses, donkeys, and mules for the twentieth century [*FAO*, 2008; *Mitchell*, 1993a, 1993b, 1998]. Data on urban equidae populations are not available at the country scale. To obtain the urban equidae stocks (N_A), for each country total stocks (N_T) were distributed over the human population using the urbanization fraction (U, based on world population data [*Klein Goldewijk et al.*, 2010]):

$$N_A = U \times N_T \times F \tag{1}$$

[26] The parameter F describes changes in animal densities over time. Urban equidae are absent in urban areas of industrialized countries after 1950 (linear reduction of F between 1 in 1900 and 0 in 1950) [*Billen et al.*, 2009; *Mokyr*, 1998; *Swaney et al.*, 2012]. In developing countries after 1950, equidae were assumed to be 10% of the calculated total equidae population ($U \times N_T$) [*FAO*, 2001] (linear reduction of F between 1 in 1900 and 0.1 for 1950–2000). To avoid unrealistic stocks of equidae for countries with a low urbanization fraction and large animal stocks, a maximum of one animal per 20 inhabitants was applied [*Barles*, 2007; *FAO*, 2001].

[27] Animal N excretion rates of 110 g d^{-1} for horses and 82 g d^{-1} for donkeys and mules are based on various sources [*Barles*, 2007; *Bouwman and Van der Hoek*, 1997; *Smil*, 1999]. P excretion was obtained from a N:P mass ratio of 7:1 based on the study by *Quynh et al.* [2005] and the composition of animal feed such as grass, hay, and straw [*CVB*, 1996] (SI 8.2). Nutrient fractions in urine (55% of N, 1% of P) and feces (45% of N, 99% of P) differ from typical values for humans [*Eckert et al.*, 2010; *Freeman et al.*, 1988; *Hintz and Schryver*, 1972; *Kohn et al.*, 2005; *Van Doorn et al.*, 2004].

2.3.2. Fate of Animal Excreta N and P

[28] Urine is assumed to be susceptible to runoff from streets (50%) and seepage into soils (50%). For N, 20% of the urine runoff is assumed to volatilize [*Bouwman et al.*, 1997]. Feces are assumed to be collected for use in agriculture (70%) [*Tarr*, 1975] or lost to the soil (30%) [*Barles*, 2007], with 5% volatilization loss of N in collected feces [*Bussink and Oenema*, 1998] (Figure 1b).

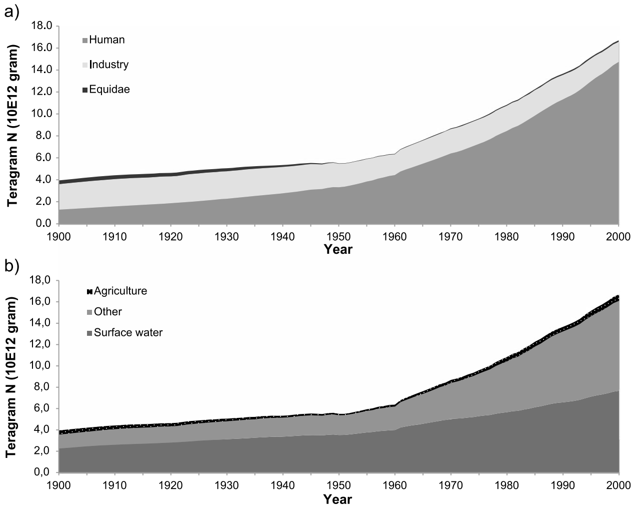

Figure 3. (a) Mass of the global gross N flow from human excreta, animal excreta, and industry. (b) Mass of global N discharge after all corrections (Figures 1a, 1b, and 1c) to surface water, recycling in agriculture, and "other." Unit: Tg N yr^{-1}.

2.4. Industrial Waste

2.4.1. Gross Industrial N and P Flows

[29] Important industries for urban nutrient flows are breweries, factories producing strawboard, potato flour, food (milk), paper, textiles, oil/grease/candles, and tanneries [*Billen et al.*, 1999; *Lohuizen*, 2006]. These industries were often situated at the (downstream) borders of urban areas [*Plaats*, 1902]. Due to scarcity of data, we estimated gross industrial nutrient flows as a fraction of gross human urban nutrient flows [*Billen et al.*, 2005; *Billen et al.*, 1999; *Liu*, 2005; *Luu et al.*, 2012; *Quynh et al.*, 2005]. Since the 1950s, detergent-producing or detergent-consuming industries add to P discharges. For both N and P, industrial waste nutrient emissions were assumed to equal 2 times the urban domestic emissions in 1900, 0.5 in 1960, and 0.15 times domestic emissions in 2000 using linear interpolation for the years in between. This trend is based on historical and recent industrial nutrient loadings to the environment [*Bixio et al.*, 2005; *Liu*, 2005; *Luu et al.*, 2012; *Quynh et al.*, 2005] (SI 9). For the early twentieth century, literature describing the situation in the Netherlands and the Seine, Scheldt, and Zenne river basins in Western Europe was used [*Billen et al.*, 2005; *Billen et al.*, 1995; *Billen et al.*, 1999; *Garnier et al.*, 2012], while for the late twentieth century, studies of the Red River Delta (Northern Vietnam) [*Luu et al.*, 2012], the Red River Basin (China and Vietnam) [*Quynh et al.*, 2005], the Nete river (Belgium), the Seine and Zenne river basins (Belgium and France) [*Billen et al.*, 1999], and Africa [*Nyenje et al.*, 2010] provided (semi)quantitative estimates of industrial nutrient loadings.

2.4.2. Fate of Industrial N and P Waste

[30] For both N and P, 30% of the gross industrial nutrient flows were assumed to end in wastewater stabilization ponds or be lost by ammonia volatilization, which is in agreement with values used by *Billen et al.* [1999]. For the complement (70%), the same types of treatment and nutrient removal efficiencies apply as those used for public wastewater treatment (Figure 1c). Since all industries were assumed to be close to surface water, treatment was assumed to be directly applied to the industry nutrient flows; no sewer connection was considered regarding industrial wastes (Figure 1c).

2.5. Sensitivity and Uncertainty Analyses

[31] Since many model parameters are uncertain and some are fraught with potential error, we first performed a sensitivity analysis to select the most important model parameters in terms of their effect on N and P discharge to surface water. This was done using Latin hypercube sampling (LHS) [*Saltelli et al.*, 2000], using default ranges prescribed for each parameter (SI Table 3). LHS can be used in combination with linear regression to quantify the uncertainty contributions of the input parameters to the model outputs. We did 1000 runs for 3 years (1900, 1950, and 2000), the two nutrients (N and P), model outputs for the three flows (discharge to surface water, agriculture recycling, and "other" nutrient flows) (3), all 46 input parameters, and 10 world regions. Standardized regression coefficients (SRCs) were computed for combinations of input parameter and the model output N and P discharge to surface water (SI Table 4) and N and P recycling in agricultural land (SI Table 5). We selected input parameters for which SRC resulted with values >0.2 or <-0.2 ($>4\%$ contribution to model output parameter) for at least one output parameter for the years 1900, 1950, and 2000 to analyze the model uncertainty with LHS (1000 runs). The uncertainty range for the selected input model parameters was determined with more detail than the sensitivity ranges based on literature and expert judgment (SI Table 6).

2.6. Spatial Allocation

[32] For allocating the N and P emissions, we count the people starting from the grid cells with highest population density [e.g., *Klein Goldewijk et al.*, 2010] within each country by means of ranking, up to the number of persons equals the number of people with a sewage connection. Areas with high population density most logically have the highest degree of connection to sewerage systems, and rural areas with low population density have the lowest degree of connection.

3. Results and Discussion

3.1. Global Scale

[33] During the twentieth century, several developments affected nutrient flows in urban areas. Global average per capita protein consumption was stable at about 70 g d^{-1} during the 1900–2000 period, but individual countries and regions showed different trends (SI 2). There were regional differences of ~30 g d^{-1} and differences between countries of up to 130 g d^{-1} (SI Figure 3). Furthermore, total global urban equidae stocks remain close to 120 million animals but vary considerably in the different world regions (SI Figure 9). P flows from laundry and dishwasher detergents increased from zero in 1950 to ~0.4 Tg P yr^{-1} in 2000 (Figure 2). Globally, urban industries were estimated to generate waste flows containing ~2.3 Tg N and in 1900, ~2.1 Tg N in 1950 and ~1.8 Tg N in 2000. Industrial waste P discharge is ~0.2 Tg P yr^{-1} for 1900, 1950, and 2000, with a peak of 0.35 Tg P yr^{-1} in 1973. Technological development and efficiency improvement in industry compensate the increasing industrial production, thus leading to relatively stable nutrient emissions (SI 9).

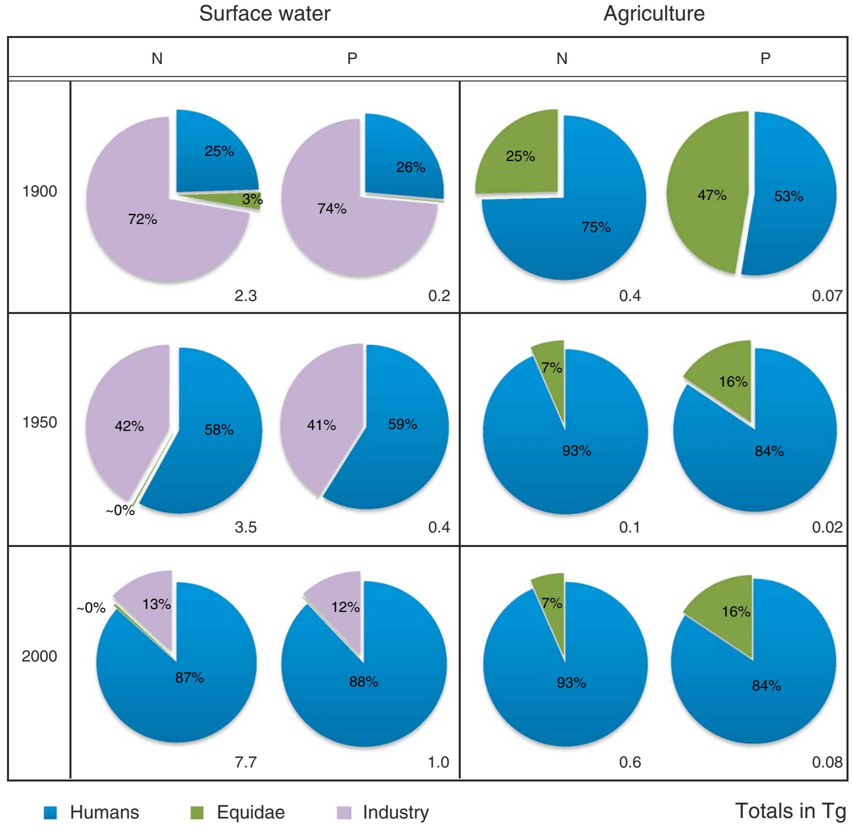

Figure 4. The relative contribution from urban human wastes (including human excreta and P-based detergents), urban animal excreta, and industrial wastes to total surface water N and P discharge and N and P recycling in agriculture for the years 1900, 1950, and 2000. Lower right values are totals in teragrams for that year.

[34] Global urban nutrient discharge to surface water increased from ~2.3 to ~7.7 Tg N yr^{-1} (factor of ~3.5) and from ~0.2 to ~1.0 Tg P yr^{-1} (factor of ~4.5) between 1900 and 2000, the majority of which occurred after 1950 (Figures 2 and 3). There has been a strong temporal variation of the contribution from humans, animals, and industries to total surface water N and P discharges and recycling of urban wastes in agriculture (Figure 4). The four major causes of variation are industries, recycling in agriculture, animal stocks, and rapid urbanization.

[35] The contribution of industries to total surface water N and P discharge decreased from 72% to 13% for N and from 74% to 12% for P between 1900 and 2000 (both sources contributed ~50% to total surface water loading just after 1940) (Figure 4).

[36] Global urban wastewater recycling in agriculture increased from ~0.4 to ~0.6 Tg yr^{-1} for N and from ~0.07 to ~0.08 Tg yr^{-1} for P, with a midcentury decline (Figures 2 and 3). In 1900, 47% of the total recycled P was estimated to come from equidae, which contrasts the 25% for N (Figure 4). Due to relatively constant global urban animal stocks and strong urban population growth, the relative contribution of equidae excreta to recycling in agriculture decreased to 16% for P and 7% for N in 2000.

[37] Human waste flow to all sinks was dominant since the mid-twentieth century due to urbanization. The prominent influence of human urban wastes in 2000 is illustrated by a global human contribution to global urban surface water nutrient discharge of 87% for N and 88% for P. Urban human waste is also important for agricultural recycling in 2000 (93% for N and 84% for P) (Figure 4).

[38] Globally, an estimated 4% of urban N and P flows were recycled in agriculture in 2000. Our estimate is lower than the ~10% of global human (urban and rural) excreta to aquaculture and agriculture [*Cordell et al.*, 2009] (which suggests high rural and aquacultural recycling) and the ~20% of global urban human waste P recycled in agriculture [*Liu et al.*, 2008]. For 1900, the estimated global urban N recycling in agriculture was 40% of the N input to agriculture from guano, nitrate salts, and other N compounds used as fertilizer (1.0 Tg N yr^{-1} [*Bouwman et al.*, 2011]). However, global fertilizer use in 2000 of 83 Tg N yr^{-1} and 14 Tg P yr^{-1} [*Bouwman et al.*, 2011] is over 100 times larger than global urban nutrient recycling in agriculture for 2000 for both N and P.

3.2. Regional Scale

[39] Total urban N and P discharges to surface water differ strongly between world regions (Figures 5a and 5b and SI Figures 10a, 10b, and 10c). Globally, total urban surface water discharges in Europe and North America were the largest for both N and P for almost all years and all sources.

[40] Urban surface water N discharge increased throughout most of the twentieth century for all regions (Figure 5a). Europe, North America, and North Asia account for the majority of global N discharges. In East Asia, the N discharge to surface water increased by a factor of ~8 between 1900 and

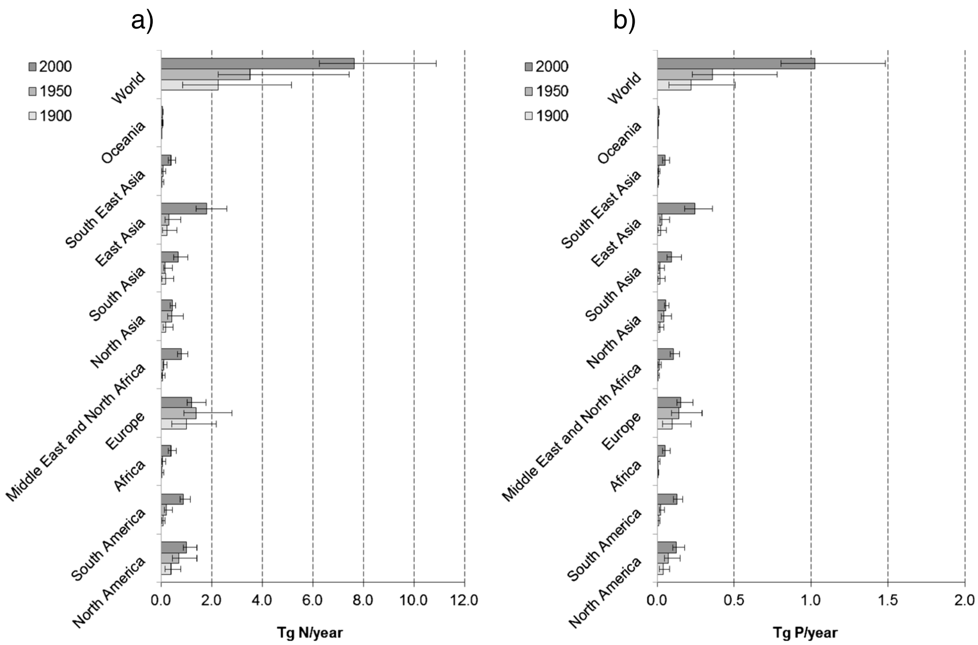

Figure 5. Total mass discharged to surface water from urban areas and uncertainty ranges (5% and 9% percentiles) for 10 world regions and globally for (a) N and (b) P. Unit: Tg of element yr^{-1}.

2000, which primarily occurred between 1960 and 2000. Because of sewer connection and treatment improvements, Europe's surface water N discharge decreased after 1970 and after 1990 in North Asia due to the collapse of the Soviet Union.

[41] Urban P discharge to surface water increased only slowly in most world regions during the first half of the twentieth century and then rapidly increased since the 1950s due to the introduction of P-based laundry detergents (Figure 5b). After 1970 for North America, North Asia, and Europe, total P loadings to surface water decreased mainly due to wastewater treatment improvements and the use of P-free detergents.

[42] During the first half of the twentieth century, nutrient recycling in agriculture showed a decrease for all regions except East Asia (SI Figures 11a and 11b). After ~1970, total N and P recycling in agriculture increased again in most regions due to urban population growth and increasing interest in wastewater recycling in agriculture. Globally, our results indicate that East Asia had the highest total mass of nutrients recycled in agriculture since ~1930 for both N and P, with a strong effect of urban population increase. The increase is balanced somewhat by declining human excreta recycling in China since the 1980s [*Liu et al.*, 2008; *McGarry*, 1976].

[43] The increase in N and P discharge to surface water since the second half of the twentieth century was most rapid in Europe and North America (Figures 5a and 5b), which agrees with twentieth century measurements of N in the United States [*Laakkonen and Lehtonen*, 1999], N and P measurements in the Rhine [*Hallegraeff*, 2003] and modeling of twentieth century N loading to the Oder [*Gadegast et al.*, 2012].

3.3. Uncertainties

[44] The model results for N and P discharge to surface water, agriculture recycling, and "other" nutrient flows showed to be most sensitive (SRC >0.2 or <-0.2) to one or more of the following model parameters (SI Tables 4 and 5): (i) protein consumption, (ii) total population, (iii) urban population, (iv) sewer connection, (v) industry fraction (fraction of household nutrient waste), (vi) N fraction in protein, (vii) fraction of nonsewered waste to agriculture, (viii) maximum number of animals per inhabitant, (ix) P as a fraction of N in human consumption, (x) tertiary treatment connection, and (xi) industry nutrient losses (both N and P). For each of these parameters, uncertainty ranges were determined (SI Table 6).

[45] Historical estimates are generally more uncertain than recent ones (Figures 5a and 5b for surface water N and P and SI Figures 11a and 11b for agricultural recycling of N and P). Also, uncertainties estimated for the major contributing regions (Europe and East Asia) can be as large as total discharges for regions such as the Middle East and North Africa and North and South Asia. Especially for the year 2000, increased knowledge on the urban nutrient systems in East Asia would strongly improve global discharge estimates (Figure 5a). For the years 1900 and 1950, Europe is the most uncertain and largest N and P discharger to surface water worldwide. Further information on sensitivity and uncertainty estimates is provided in SI 10.

[46] Global surface water N discharge estimates range between 0.8 and 5.2 Tg N yr^{-1} in 1900, 2.3 and 7.4 Tg N yr^{-1} in 1950, and between 6.3 and 10.9 Tg N yr^{-1} in 2000 (Figure 5a). Global surface water P discharge estimates are in the range 0.08–0.5 Tg P yr^{-1} in 1900, 0.2–0.8 Tg P yr^{-1} in 1950, and 0.8–1.5 Tg P yr^{-1} in 2000 (Figure 5b).

[47] A number of nutrient sources were not included in the model such as (i) other solid household refuse and sewage as reported for Paris [*Barles and Lestel*, 2007] and (ii) urban livestock other than equidae [*FAO*, 2001] (SI 10.3). Furthermore, the modelled urban system (Figures 1a, 1b, and 1c) may not be correct for some regions due to lack of information on recycling of urban wastes in aquaculture, recycling of treated untreated wastewaters in agriculture, on-site sanitation systems, and incineration of waste (SI 10.3).

4. Conclusions

[48] Global urban nutrient flows from humans, animals, and industries increased by a factor of ~3.5 for N and by a factor of ~4.5 for P during the twentieth century; since 1950 urban nutrient flows are dominated by household N and P flows, i.e., human excreta and P-based detergent use. Up till about 1940, industries contributed most to total urban N and P surface water discharges. However, while global industrial nutrient emissions were fairly constant (as a result of simultaneous production increase and technological improvement), the relative contribution of industries decreased dramatically due to rapidly increasing household emissions (human population growth and urbanization). A large number of factors (the major ones related to food consumption, urban population, sewer connection and industrial emissions) contribute to the uncertainty of −18% to +42% for N and −21% to +45% for P around the standard surface water loading estimate of in 2000.

[49] The industrialized and urbanized regions of Europe and North America are the largest dischargers of N and P to surface water for almost all years. These regions are also the ones that show a major decrease in nutrient discharge in the late twentieth century, which is due to the construction and improvement of treatment facilities and the use of P-free detergents. Since urban populations will inevitably continue to increase globally, measures are required to counteract the adverse effects of nutrients in urban wastewater. As demonstrated in North America and Europe, wastewater treatment can effectively reduce surface water N and P discharges.

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