

1 **Production and application of manure nitrogen and phosphorus**
2 **in the United States since 1860**

3
4 Zihao Bian¹, Hanqin Tian¹, Qichun Yang², Rongting Xu¹, Shufen Pan¹, Bowen Zhang³

5
6 ¹International Center for Climate and Global Change Research, School of Forestry and Wildlife
7 Sciences, Auburn University, Auburn, AL 36849, USA

8 ²Department of Infrastructure Engineering, The University of Melbourne, Parkville 3010,
9 Australia

10 ³Department of Environment, Geology, and Natural Resources, Ball State University, Muncie, IN
11 47306 USA

12
13 *Correspondence to:* Hanqin Tian (tianhan@auburn.edu)

26 **Abstract:** Livestock manure nitrogen (N) and phosphorus (P) play an important role in
27 biogeochemical cycling. Accurate estimation of manure nutrient is important for assessing
28 regional nutrient balance, greenhouse gas emission, and water environmental risk. Currently,
29 spatially explicit manure nutrient datasets over century-long period are scarce in the United
30 States (U.S.). Here, we developed four datasets of annual animal manure N and P production and
31 application in the contiguous U.S. at a 30 arc-second resolution over the period of 1860-2017.
32 The dataset combined multiple data sources including county-level inventory data, as well as
33 high-resolution livestock and crop maps. The total production of manure N and P increased from
34 1.4 Tg N yr⁻¹ and 0.3 Tg P yr⁻¹ in 1860 to 7.4 Tg N yr⁻¹ and 2.3 Tg P yr⁻¹ in 2017, respectively.
35 The increasing manure nutrient production was associated with increased livestock numbers
36 before the 1980s and enhanced livestock weights after the 1980s. The manure application
37 amount was primarily dominated by production and its spatial pattern was impacted by the
38 nutrient demand of crops. The intense-application region mainly enlarged from the Midwest
39 toward the Southern U.S., and became more concentrated in numerous hot spots after the 1980s.
40 The South Atlantic-Gulf and Mid-Atlantic basins were exposed to high environmental risks due
41 to the enrichment of manure nutrient production and application from the 1970s to the period of
42 2000-2017. Our long-term manure N and P datasets provide detailed information for national and
43 regional assessments of nutrient budgets. Additionally, the datasets can serve as the input data for
44 ecosystem and hydrological models to examine biogeochemical cycles in terrestrial and aquatic
45 ecosystems. Datasets are available at <https://doi.org/10.1594/PANGAEA.919937> (Bian et al.,
46 2020).

47 **Keywords:** Manure; Nutrient; Nitrogen; Phosphorus; Production; Application

48 **1 Introduction**

49 Animal manure, as a fertility package, is a traditional source of nutrients and can provide
50 abundant nitrogen (N), phosphorus (P), and potassium for cropland and pasture. Animal manure
51 nutrients circulate widely in the Soil-Plant-Animal system, and are highly involved in global
52 nutrient cycling (Bouwman et al., 2013; Sheldrick et al., 2003). Although synthetic fertilizer has
53 been widely used since the mid-20th century, livestock excreta is still the major nutrient source in
54 agricultural soils, accounting for approximately 18% and 28% of the total N and P inputs to
55 global cropland, respectively (Sheldrick et al., 2003; Zhang et al., 2020). Moreover, the total
56 global animal manure N and P production has exceeded global fertilizer use (Bouwman et al.,
57 2009). Therefore, the efficient recycling of manure can potentially meet the growing nutrient
58 demand of crops. The circular nutrient source provided by manure enables nations to sustain
59 their agricultural production with less reliance on imported fertilizer, especially mineral P
60 fertilizer (Koppelaar and Weikard, 2013; Powers et al., 2019). Different from N which can be
61 fixed from the atmosphere through microbial symbiosis with plants and the Haber-Bosch
62 process, P is a rock-derived nutrient and there is no biological or atmospheric source for P. The
63 limited and unevenly distributed P-rich rocks can threaten food security and have raised concerns
64 in many resource-limited countries, including the United States (U.S.) (Amundson et al., 2015).
65 Enhanced recovery nutrients from manure can not only increase agricultural dependence, but
66 may also reduce nutrient losses out of the Soil-Plant-Animal system. Additionally, the
67 improvement of livestock operations in recent decades also facilitated the recoverability and
68 utilization of animal manure (Kellogg et al., 2000).

69 Although the application of manure and fertilizer enhanced crop production, excessive nutrient
70 might leave the Soil-Plant-Animal system through the biogeochemical flow and potentially

71 contaminate the environment if not properly managed (Mueller and Lassaletta, 2020; Zanon et
72 al., 2019). Specifically, agricultural land is a sink for anthropogenic N and P inputs (e.g.
73 synthetic fertilizer, manure, atmospheric deposition), and simultaneously acts as N and P sources
74 for aquatic systems as well as a N source for atmosphere (Bouwman et al., 2013; Elser and
75 Bennett, 2011; Schlesinger and Bernhardt, 2013). The major N gaseous loss from fertilizer use
76 and animal excreta includes the emissions of ammonia (NH₃), nitrous oxide (N₂O), and nitric
77 oxide. NH₃ can react with other air pollutants and form aerosols to reduce visibility and threaten
78 human health (Bouwman et al., 2002; Xu et al., 2018), and N₂O is one of the most important
79 greenhouse gasses (Davidson, 2009). N₂O emission from animal manure is one of the major
80 contributors to global anthropogenic N₂O emissions (Tian et al., 2020). Additionally, large
81 fractions of the N and P applied to cropland lost through leaching, erosion, and surface runoff
82 and are transported into rivers toward lakes and coastal oceans (Smith et al., 1998; Van Drecht et
83 al., 2005). Excess N and P could dramatically impair freshwater and coastal ecosystems, causing
84 eutrophication, hypoxia, and fish-killing (Garnier et al., 2015; Smith et al., 2007). Oxygen-
85 depleted marine coastal “dead zones” associated with nutrient-stimulated algal blooms continue
86 to expand. For example, the northern Gulf of Mexico is one of the largest dead zones in the
87 world and the hypoxic area often exceeds 15,600 km² in midsummer (1968-2016) (Del Giudice
88 et al., 2019).

89 Considering the importance of manure nutrient on crop production, greenhouse gas emission,
90 and water pollution, it is vital to have a better understanding of livestock manure nutrient
91 production and application at national or even global scales (Potter et al., 2010; Sheldrick et al.,
92 2002; Tian et al., 2016). Quantized and spatialized manure nutrient data can help stakeholders
93 find a local recyclable nutrient source or make strategies to minimize N and P losses. Currently,

94 most studies only provided county-level manure nutrient production data in the U.S., with short
95 periods (Kellogg et al., 2000; Ruddy et al., 2006). Nevertheless, terrestrial biosphere models
96 usually require spatially explicit manure nutrient input data to simulate the anthropogenic effect
97 on biogeochemical cycle since the preindustrial period (Tian et al., 2019). Studies focusing on
98 soil nutrient storage change and legacy soil nutrient also need long-time series manure nutrient
99 data (MacDonald et al., 2012; Rowe et al., 2016). Moreover, previous studies usually assumed
100 that nutrient excretion per animal is constant over time when quantifying nutrient production
101 based on livestock number, which may lead to uncertainties (Zhang et al., 2020). Geographically
102 explicit manure nutrient application in cropland (excluding pasture), as the direct nutrient input
103 for the soil-crop system, hasn't been specifically estimated across the U.S. In this study, our
104 objectives are to (1) develop grid-level manure N and P production datasets in the U.S. based on
105 county-level livestock populations, dynamic livestock weight over time, and high-resolution
106 livestock distribution maps; (2) develop grid-level manure N and P application in cropland
107 datasets by integrating manure nutrient production and nutrient demand of crops; (3) investigate
108 the spatiotemporal patterns of manure nutrient production and application based on these
109 datasets, and (4) further identify regions with a high risk of excessive nutrient loading. The four
110 datasets display the masses of manure N and P per area in each 30×30 arc-second grid-cell
111 during 1860-2017. The datasets can be used to drive ecosystem, land surface, and hydrological
112 models to simulate manure-induced greenhouse gas emissions and nutrient loadings.

113 **2 Methods**

114 Datasets of manure N and P production and application were developed by incorporating
115 multiple datasets (Table 1). The geographically explicit manure N and P production data were
116 first calculated based on county-level livestock populations, dynamic livestock weights, and

117 livestock distribution maps. Then the crop nutrient demand maps were developed by merging
118 cropland distribution maps with crop-specific harvest area and nutrient assimilative capacities.
119 Finally, the spatially explicit manure N and P application data were estimated by incorporating
120 county-level manure production, recoverability factors, cropland fraction, and cropland nutrient
121 demand maps. To facilitate studying the impact of manure nutrients on water quality, we further
122 analyzed the average annual manure production and application in four decades (the 1860s,
123 1930s, 1970s, and 2010-2017) across the major 18 basins (Fig 1).

124 2.1 Manure nutrient production

125 Manure nutrient production refers to the animal excretion in this study. The county-level manure
126 N and P production during 1930-2017 were calculated based on the livestock population, animal
127 body weight, and nutrient excretion rates according to the method (Eq. 1) proposed by Puckett et
128 al. (1998).

129 County-level manure nutrient production was calculated as follows:

$$130 \quad Pro_{x,c} = \sum_{i=1}^n Pop_{i,c} \cdot W_i \cdot Er_{x,i} \cdot Days \quad (1)$$

131 where $Pro_{x,c}$ is the annual manure nutrient x (N or P) production in county c (kg N/P yr⁻¹); i is
132 animal type; $Pop_{i,c}$ is the county-level animal population (head); W_i is the annual average live
133 body weights of animal (kg); $Er_{x,i}$ represents the excreted manure nutrients rate per unit weight
134 of animal (kg N/P kg⁻¹ day⁻¹) (Table S1); $Days$ is the number of days in the life cycle of animal
135 within a year.

136 Data of livestock and poultry population were derived from the U.S. Department of Agriculture
137 (USDA) census reports from 1930 to 2017 at 4- or 5-year intervals. Eleven livestock and poultry
138 categories were considered in this study, including beef cows, milk cows, heifers, steers, hogs,

139 sheep, horses, chickens, pullets, broilers, and turkeys. Livestock population data for the recent
140 five census reports (1997–2017) can be directly collected from the USDA Census Data Query
141 Tool. Livestock population data before 1997 were collected from Cornell Institute for Social and
142 Economic Research Data Archive (1949–1992), or manually digitalized from the USDA reports
143 (1930–1945). More details of data collection, methods of dealing with missing data can be found
144 in Yang et al. (2016). Annual average live weights of livestock and poultry, including cattle,
145 hogs, sheep, broilers, chickens, and turkeys, were derived from the USDA Economic Research
146 Service. We developed annual manure nutrient production by assuming a linear change between
147 every two census years.

148 Global Livestock Impact Mapping System (GLIMS) provided gridded livestock population
149 maps at a resolution of 30 arc-second (<https://livestock.geo-wiki.org/home-2/>). These maps were
150 developed according to statistical relationships between livestock inventory data and multiple
151 environmental variables, including climate, land cover, human activities (Robinson et al., 2014).
152 Combining with the GLIMS data, we spatially allocated manure nutrient production within each
153 county. The grid-level manure nutrient production was first calculated based on the GLIMS data,
154 and the total quantity of manure nutrient production in each county was obtained by calculating
155 the sum of productions in all grid-cells within each county. Then, we calculated ratios of USDA-
156 based county-level manure nutrient production to GIMS-based county-level data, and these ratios
157 were used to adjust grid-cell values within each county. After this step, the developed grid-level
158 products were in line with USDA-based annual county-level data in total quantities, with the
159 spatial pattern inside each county inherited from the GLIMS-based manure nutrient production
160 data (Eq. 2).

161
$$Pro_{x,j} = GPro_{x,j} \cdot \frac{Pro_{x,c}}{GPro_{x,c}} \quad (2)$$

162 where $Pro_{x,j}$ is manure nutrient production in grid-cell j (kg N/P km⁻² yr⁻¹); $GPro_{x,j}$ is manure
163 nutrient production in grid-cell j calculated based on the GLIMS livestock data (kg N/P km⁻² yr⁻¹); $GPro_{x,c}$
164 is the GLIMS-based manure nutrient production at county c where grid cell j is
165 located (kg N/P yr⁻¹).

166 To generate grid-level manure production from 1860 to 1930, we obtained manure production
167 change rates (1860-1930) from the dataset developed by Holland et al. (2005) and applied them
168 on the grid-level manure nutrient production in 1930. Holland et al. (2005) provided global
169 annual manure N production data from 1860 to 1960. In order to combine this dataset with the
170 U.S. manure nutrient production data, we assumed manure production changes in the U.S. were
171 consistent with the global trend and manure N:P ratio was constant during 1860-1930 (Zhang et
172 al., 2017).

173 2.2 Manure nutrient application

174 Manure nutrient application data were developed by allocating the county-level recoverable
175 manure nutrient according to the grid-level manure nutrient demand of crops. The recoverable
176 manure nutrient represents the proportion of manure nutrients that could reasonably be expected
177 to be collected from the confinement facility and later be applied to the land (Kellogg et al.,
178 2000). The recoverable manure nutrient was applied to cropland and pastureland according to
179 their demands. We calculated recoverable manure nutrient amounts by adjusting the county-level
180 manure production with recoverability factors provided by the Nutrient Use Geographic
181 Information System (NuGIS, <http://nugis.ipni.net/>). The nutrient demand was estimated
182 according to the assimilative capacity, the maximum amount of manure nutrient application

183 without building up nutrient level in the soil over time (Kellogg et al., 2000). We obtained the
 184 proportion of recoverable manure nutrient that can be applied to cropland by combining the
 185 assimilative capacities and areas of cropland and pastureland. The areas of cropland and
 186 pastureland during 1860-2016 were derived from the HYDE 3.2 (Klein Goldewijk et al., 2017).
 187 The above-mentioned processes are represented by the following equations:

$$188 \quad APP_{x,c} = Pro_{x,c} \cdot Rf_{x,c} \cdot f_{x,crop,c} \quad (3)$$

$$189 \quad f_{x,crop,c} = \frac{A_{crop,c} \cdot S_{x,crop}}{A_{crop,c} \cdot S_{x,crop} + A_{past,c} \cdot S_{x,past}} \quad (4)$$

190 where, $APP_{x,c}$ is the recoverable animal manure nutrient available for application on cropland in
 191 county c (kg N/P yr⁻¹); $Rf_{x,c}$ is the manure nutrient recoverability rate (the recoverability rates
 192 are unitless and county-specific with average values 0.19 for N and 0.35 for P), and $f_{x,crop,c}$
 193 refers to the fraction of manure nutrient that is available for cropland (unitless); $A_{crop,c}$ and
 194 $A_{past,c}$ represent the annual area of cropland and pasture in each county (km²), respectively,
 195 while $S_{x,crop}$ and $S_{x,past}$ represent the average assimilative capacities of cropland and
 196 pastureland (kg N/P km⁻²), respectively (Table S2).

197 To spatialize county-level recoverable animal manure nutrient to gridded maps, we first
 198 developed annual grid-level crop nutrient demands data as the base maps (Eq. 5). Nutrient
 199 demands of crops were estimated by combining the assimilative capacities, harvested areas and
 200 yields of 13 crops (maize, soybeans, sorghum, cotton, barley, wheat, oats, rye, rice, peanuts,
 201 sugar beets, tobacco, and potatoes). The grid-level average assimilative capacity of cropland was
 202 calculated based on crop-specific yield and harvested area maps in 2000 provided by Monfreda
 203 et al. (2008). Next, this map of cropland assimilative capacity was integrated with dynamic
 204 cropland fraction data (Yu and Lu, 2018) to obtain annual nutrient demand maps from 1860 to

205 2016. Original cropland fraction data was at a resolution of 1 km in the projected coordinate
 206 system which approximates 30 arc-second resolution in the geographic coordinate. We resampled
 207 the cropland fraction maps into the resolution of 30 arc-second to match the manure nutrient
 208 production data.

209

$$210 \quad Dem_{x,j} = \sum_{k=1}^m Y_{j,k} S_{x,k} \cdot Den_j \quad (5)$$

211 where, $Dem_{x,j}$ is the crop demand for manure nutrient in grid j (kg N/P km⁻² yr⁻¹); $Y_{j,k}$ is the
 212 yield of crop k (ton per area of cropland) and $S_{x,k}$ is the manure nutrient assimilative of crop k
 213 (kg N/P per ton product) (Table S3); Den_j represents the cropland density in each grid (unitless).

214 The downscaling of county-level recoverable manure nutrient data into grid maps was similar to
 215 the method used in developing manure nutrient production data (Eq. 2). The grid values on
 216 manure nutrient demand maps were adjusted to match annual county-level recoverable manure
 217 nutrient data (Eq. 6). The manure nutrient application data from 1860 to 2017 were developed
 218 through above-mentioned processes, however, several variables and parameters in these
 219 processes were not available through the whole study period (e.g., manure recoverability rate,
 220 crop yield). Therefore, we assumed these variables or parameters did not change before or after
 221 the data-available period (Kellogg et al., 2000; Puckett et al., 1998).

$$222 \quad APP_{x,j} = Dem_{x,j} \cdot \frac{APP_{x,c}}{Dem_{x,c}} \quad (6)$$

223 where, $APP_{x,j}$ is the manure nutrient application in grid j (kg N/P km⁻² yr⁻¹) and $Dem_{x,c}$
 224 refers to the demand for manure nutrient in county c where grid j is located (kg N/P yr⁻¹).

225 **3 Results**

226 **3.1 Temporal and spatial patterns of manure nutrient production**

227 We estimate that the total manure N and P production increased from 1.4 Tg N yr⁻¹ and 0.3 Tg P
228 yr⁻¹ in 1860 to 7.4 Tg N yr⁻¹ and 2.3 Tg P yr⁻¹ in 2017, respectively (Fig 2). The manure N and P
229 production reached the first peak in 1975 (6.1 Tg N yr⁻¹ and 1.8 Tg P yr⁻¹), and slightly declined
230 thereafter, then regrew since 1987 with the second peaks occurring in 2007 (N) and 2017 (P),
231 respectively. The slight decrease in manure nutrient production between 2008 and 2012 may be
232 associated with the financial crisis and the low demand for livestock products. The total manure
233 N and P production increased 5-fold and 7-fold during 1860-2017, with the increasing rates of
234 0.03 Tg N yr⁻² and 0.006 Tg P yr⁻² during 1860-1930, 0.05 Tg N yr⁻² and 0.02 Tg P yr⁻² during
235 1930-2017 (p<0.01), respectively. The N:P ratio in total manure production changed from 4.33 in
236 1930 to 3.25 in 2017. The decrease in the N:P ratio in total manure production was related to the
237 change in the structure of animal population. For example, the proportion of beef cows and
238 broilers (N:P ratio in excretion: 3.0-3.2) increased while that of milk cows and horses (N:P ratio
239 in excretion: 5.5-6.7) decreased over the study period.

240 The spatial pattern of animal manure N and P production showed the similar change over the
241 study period (Fig 3). The distribution maps showed that the Midwestern U.S. (e.g., Iowa,
242 Missouri, and Illinois) was the core region (> 300 kg N km⁻² yr⁻¹ or 100 kg P km⁻² yr⁻¹) of manure
243 N (P) production in 1860. From 1860 to 1930, the high manure nutrient production region (> 600
244 kg N km⁻² yr⁻¹ or 200 kg P km⁻² yr⁻¹) mainly enlarged outwards from the Midwest. Between 1930
245 and 1980, manure N (P) production not only intensified in the Midwest but also in the Southern
246 U.S. (e.g., Texas, Georgia, and North Carolina). After 1980, manure N (P) production became
247 more concentrated in many hot spots (> 6000 kg N km⁻² yr⁻¹ or 3000 kg P km⁻² yr⁻¹), especially

248 in the southeastern U.S. Meanwhile, part of regions around these hot pots experienced a decline
249 in manure production.

250 According to the change rates of manure nutrient production from 1860 through 2017 (Fig.4),
251 several growth poles (change rates $> 20 \text{ kg N km}^{-2} \text{ yr}^{-2}$ or $5 \text{ kg P km}^{-2} \text{ yr}^{-2}$, $p<0.01$) located in
252 Iowa, Arkansas, California, Alabama, Pennsylvania were identified. The belt (change rates > 5
253 $\text{kg N km}^{-2} \text{ yr}^{-2}$ or $1 \text{ kg P km}^{-2} \text{ yr}^{-2}$, $p<0.01$) from Minnesota to Texas, as well as scattered areas
254 along the east and west coasts, were the primary contributors to the increase in manure N (P)
255 production. Aside from the huge increase in the Midwest and Southeast, decreasing trends were
256 exhibited in some regions, particularly the northeastern border of the U.S.

257 3.2 Comparison of manure nutrient demand and production

258 We assumed that the capacity of crops to assimilate nutrients was equal to manure nutrient
259 demand. From 1860 to 1930, the manure N (P) demand of cropland intensified and enlarged
260 inside the Corn Belt region (e.g. Iowa, Illinois, Minnesota, Nebraska, North Dakota, and South
261 Dakota), as well as the Southern U.S. (e.g., Texas, Oklahoma, Arkansas, Mississippi, Alabama,
262 Georgia, and Tennessee) (Fig 5). After 1930, change in spatial pattern of manure nutrient demand
263 was dominated by the abandonment of cropland, and the magnitude of demand slightly
264 decreased, especially after 1980. Compared to the spatial patterns of manure production and
265 demand (Figs 3 and 5), it is worth noting that the high manure production and demand regions
266 overlapped in the Midwest and Southeastern U.S., but a large deficit (demand higher than
267 production) existed along the Lower Mississippi River Valley.

268 3.3 Temporal and spatial patterns of manure nutrient application in cropland

269 Animal manure N (P) application amount is primarily dominated by production and its spatial
270 pattern is impacted by demand. The overall manure application to production ratios were 0.15

271 and 0.23 for N and P, respectively. Driven by cropland expansion and enhanced manure
272 production, total manure N and P application in croplands increased 9-fold and 10-fold since
273 1860, reaching 1.3 Tg N yr⁻¹ and 0.6 Tg P yr⁻¹ in 2017 (Fig 6). The N:P ratio in manure
274 application decreased from 2.62 to 2.32 during 1930-2017. The substantial increase of manure N
275 and P application mainly happened in two periods: 1924-1970 (increase rates: 0.009 Tg N yr⁻²,
276 and 0.005 Tg P yr⁻², $p < 0.01$) and 1987-2017 (increase rates: 0.01 Tg N yr⁻² and 0.005 Tg P yr⁻²,
277 $p < 0.01$). The variations of total application and production quantities didn't follow the same
278 trajectory. For example, from 1975 to 1987, when manure N production decreased, the total
279 manure application still remained stable. The application to production ratios reached the first
280 peak in 1891 (N: 0.14, P: 0.25) followed by a decrease until 1945 (N: 0.13, P: 0.20), and then
281 resumed the increasing trend through 2017 (N: 0.18, P: 0.25).

282 The spatial shift of manure application, similar to manure nutrient demand, gradually expanded
283 inside the Corn Belt and toward the Southern U.S. (Fig 7). The expansion of manure application
284 region primarily occurred during 1860-1930, induced by cropland expansion. After 1930, the
285 changed spatial patterns of manure N (P) application were characterized by intensified
286 application in the Midwest and multiple hot spots ($> 2000 \text{ kg N km}^{-2} \text{ yr}^{-1}$ or $1000 \text{ kg P km}^{-2} \text{ yr}^{-1}$).
287 The spatial distribution of hot spots on application maps was similar to that on manure nutrient
288 production maps. In 2017, high manure nutrient application regions ($> 500 \text{ kg N km}^{-2} \text{ yr}^{-1}$ or 200
289 $\text{kg P km}^{-2} \text{ yr}^{-1}$) mainly distributed in the Midwestern U.S., Southern U.S., Mid-Atlantic (e.g.,
290 Pennsylvania, Maryland, and Virginia), and California, where abundant recoverable manure
291 nutrients were applied in the local cropland to meet the high nutrient demand of crops. A quite
292 low manure nutrient application rate ($< 100 \text{ kg N km}^{-2} \text{ yr}^{-1}$ or $50 \text{ kg P km}^{-2} \text{ yr}^{-1}$) was observed in

293 regions with less cropland demand (e.g., Southwestern U.S.) and low manure production (e.g.,
294 Lower Mississippi River Valley).

295 **3.4 Manure production and application across the major river basins**

296 From the 1860s to the 1970s, all basins exhibit increased manure nutrient production (Figs 8a
297 and 11b). However, from the 1970s to the 2010s, the manure N and P production decreased in
298 New England and Missouri basins, while a dramatic increase was shown in the South Atlantic-
299 Gulf, Mid-Atlantic, and Arkansas-White-Red basins. Manure application demonstrated a similar
300 pattern across different basins (Figs 8c and 8d), but it increased from the 1970s to the 2010s in
301 most basins except the two basins (the New England and Souris-Red-Rainy) in the northern
302 regions. In the 1970s, the Missouri basin was the largest source contributing ~20% N (P) of the
303 total manure production, while the Upper Mississippi basin had the highest manure N (P)
304 application in cropland accounting for 19% N and 24% P of the total manure N (P) application.
305 During 2011-2017, however, the dominant regions of manure nutrient production and application
306 were shifted to the South Atlantic-Gulf basin which accounted for the largest single share (18%
307 N and 19% P of the total N (P) production, 24% N and 21% P of the total N (P) application). The
308 uneven distribution of manure application intensified during 1860-2017, demonstrated by the
309 standard deviation of manure N and P application across all basins consistently increasing from
310 0.013 Tg N yr⁻¹ and 0.005 Tg P yr⁻¹ in the 1860s to 0.081 Tg N yr⁻¹ and 0.038 Tg P yr⁻¹ during
311 2010-2017.

312 **4 Discussion**

313 **4.1 Comparison with previous investigations**

314 Within this study, we compared **manure nutrients production data** with other four datasets from
315 Food and Agriculture Organization Corporate Statistical Database (FAOSTAT, 2019), NuGIS,

316 Kellogg et al. (2000) and Yang et al. (2016). FAOSTAT provides total manure N production at
317 the national level from 1961 to 2017, while the other three datasets provide county-level manure
318 N and P production data. The estimated manure N (P) production from this study was lower than
319 the other two datasets (FAOSTAT and Yang et al.) before 1982, and started to become the highest
320 dataset after 2003 (Fig 9). During 1982-2007, the estimation from this study is very close to
321 other estimations developed at the county-level. The average total manure N (P) production over
322 1987-1997 was 6.02 Tg N yr⁻¹ (1.79 Tg P yr⁻¹), 6.75 Tg N yr⁻¹, 5.96 Tg N yr⁻¹ (1.75 Tg P yr⁻¹),
323 5.64 Tg N yr⁻¹ (1.67 Tg P yr⁻¹), and 6.01 Tg N yr⁻¹ (1.86 Tg P yr⁻¹), respectively, for this study,
324 FAOSTAT, NuGIS, Kellogg et al., and Yang et al. The differences between different datasets
325 were derived from calculation methods, chosen livestock types and numbers, as well as
326 parameters, such as animal-specific excreted manure nutrient rates and the number of days in the
327 life cycle. In terms of changing trends, manure N and P production were relatively stable after
328 the 1960s in FAOSTAT, Kellogg et al., and Yang et al., while the NuGIS data increased slightly
329 between 1987 and 2007 and then decreased sharply after 2010. In contrast, our results showed an
330 increasing trend after the 1980s due to the consideration of the increased animal body sizes.

331 In the previous four datasets, temporal changes in manure N (P) production are driven by animal
332 numbers. It is worth noting that manure N (P) production can still increase despite the
333 stabilization of livestock numbers in recent years. Driven by the advanced technology, livestock
334 live weight and size consistently increased, which may enhance the manure nutrient excretion
335 rate of each animal (Lassaletta et al., 2014; Sheldrick et al., 2003; Thornton, 2010). We
336 compared manure nutrient production calculated with constant average weights and with
337 dynamic weights of livestock. The results showed that manure production with dynamic weights
338 increased dramatically after the 1990s (Fig.10). Enhanced livestock weights contributed 59% and

339 54% of the increase in manure N and P production, respectively, from 1987 to 2017 when the
340 differences between the two total production data reached 0.98 Tg N yr⁻¹ and 0.31 Tg P yr⁻¹.
341 It is difficult to compare our dataset of manure N (P) application in soils with previous studies
342 since these datasets provided reference values with various definitions and were generated based
343 on different statistical methods. For example, FAOSTAT provided annual data of “Manure
344 applied to soils” in the U.S., whereas this dataset was developed based on the assumption that all
345 treated manure, net of losses (e.g., NH₃ volatilization, N leaching, and runoff), is applied to soils
346 following the method in the 2006 IPCC guidelines (Eggleston et al., 2006). Kellogg et al. (2000)
347 and NuGIS both estimated recoverable manure nutrients by multiplying confined livestock units,
348 recoverability factors, and nutrients per ton of manure after losses. All three datasets do not
349 separate manure application to cropland and pastureland. This study developed manure nutrient
350 application data in cropland by applying the method of recoverability factor in combination with
351 the cropland nutrient assimilative capacity. Compared to the other three datasets, our data
352 subtracted the proportion of manure application on pastureland and considered the impact of the
353 change in cropland area, which can lead to relatively low data values.

354 **4.2 The impact of manure nutrient enrichment on coastal oceans**

355 Animal manure N (P) that is lost through surface runoff or leaching exacerbated eutrophication
356 and hypoxia in the aquatic system in the U.S. (Feyereisen et al., 2010; Williams et al., 2011).
357 During the expansion of manure production from the Midwest to the Southeastern coastline,
358 massive amounts of nutrients get more of a chance to be transported to the estuary. When rivers
359 transport nutrients from land to coastal oceans, nutrients could be removed or retained through
360 denitrification, plant and microbial uptake, organic matter burial in sediment, and sediment
361 sorption (Billen et al., 1991; Seitzinger et al., 2002). As the accumulated manure gets closer to

362 the coastline, manure nutrients that enter into rivers may be less likely to decrease during
363 transportation due to the short distance. Additionally, the risk of massive manure loss in
364 hurricane events increases under the background of enhanced Atlantic hurricane activities since
365 1995 (Saunders and Lea, 2008; Trenberth, 2005). Flooding rains and high winds may destroy
366 manure storage structures (e.g., pad, pond, lagoon, tank, and building), resulting in the direct
367 release of untreated manure into rivers (Tabachow et al., 2001).

368 The South Atlantic-Gulf and Mid-Atlantic basins are two critical coastal regions with the
369 enrichment of manure nutrient production and application from the 1970s to the 2010s due to
370 intensive livestock farming. The low recovery and reuse rate of animal manure N (P) can
371 potentially cause a significant amount of manure N and P exports from the basins into the Gulf of
372 Mexico and the Atlantic Ocean (Sheldrick et al., 2003). The Upper Mississippi, Missouri, and
373 Arkansas-White-Red sub-basins within the Mississippi River basin were the three largest sources
374 of manure production and were the dominant contributors to N and P loads into the Gulf of
375 Mexico (David et al., 2010; Jones et al., 2019). The Upper Mississippi and Missouri basins that
376 had the highest manure nutrient production and application in the 1970s and maintained the high
377 quantities until 2010, while manure N (P) production and application largely increased in the
378 Arkansas-White-Red basin during 2011-2017. The enhanced total manure production may
379 continually be responsible for the enriched loads of N and P that can lead to coastal water
380 pollution (Rabalais and Turner, 2019).

381 **4.3 Implication for manure nutrient management**

382 The structure of animal agriculture has shifted toward concentrated animal feeding operations
383 (CAFOs), which led to the increased numbers of animals in confinements (Kellogg et al., 2000).
384 Thus, manure production became increasingly concentrated in several regions with large

385 operations. Meanwhile, the decreased manure production in partial areas of the Midwestern and
386 Southern U.S was due to the disappearance of small family farms. On the other hand, the
387 enhanced animal weight caused the additional increase in manure production in operations with
388 plenty of confined animals. The unevenly intensified distribution of manure production may have
389 further exacerbated the imbalance of regional nutrient allocation. Currently, opportunities for
390 widespread manure application are limited because the transport of manure can be costly.
391 Furthermore, the long distance between livestock farms and cropland can bring difficulties to
392 practical operations (MacDonald, 2009). There remain gaps between manure production and
393 demand in some regions of the U.S. (e.g., the Lower Mississippi River Valley). In contrast,
394 manure collected from many farms cannot be properly used to fertilize crops. The unusable
395 manure is not only a waste of manure resources, but may also cause serious environmental
396 problems through nutrient losses into the atmosphere and aquatic systems.

397 The efficient recovery and processing of manure nutrients, the transportation of manure from
398 CAFOs to the specific crop area, and the utilization of manure as bioenergy can be important
399 pathways to control pollution caused by the uneven distribution of manure production (He et al.,
400 2016). The CAFOs facilitate the recovery of animal manure, which has created conditions for
401 large-scale utilization and management of manure. Because of the economies of scale, the cost of
402 transportation and management for per unit animal manure can be reduced, making the
403 utilization of manure more feasible. Establishing a direct link between CAFOs and specific crop
404 area ensures that animal manure production can be consumed in large quantities and thereby
405 improving economic efficiency. For the centralized management of animal manure, nutrient
406 losses during collection, storage, and application should be constrained or avoided, because a
407 small proportion of nutrient losses can even contaminate regional environment if manure nutrient

408 amounts are huge. Manure management systems with the integrated package of measures are
409 necessary for controlling nutrient losses from the feed–animal–manure–crop chain (Oenema et
410 al., 2007).

411 **4.4 Assumptions and Uncertainties**

412 Uncertainties in this study are primarily associated with data sources and methods that were
413 used. First, multiple data sources were used to develop the datasets of manure production and
414 application data; however, biases exist in these source data. For instance, the non-disclosure of
415 the livestock data in the USGS Census of Agriculture can cause the underestimate of manure
416 production in numerous counties (Yang et al., 2016). Second, the parameters in the calculation
417 model, e.g., excreted manure nutrient rates, could bring uncertainties in the estimation of animal
418 manure nutrient production and application. Third, various assumptions were made in this study
419 to extend the time series of data and spatialize data from the county-level to the grid-level. These
420 assumptions were established based on available data and experience, but uncertainties still
421 existed and influenced the accuracy of the dataset. The limitations and uncertainties of these
422 assumptions were further discussed and explained in the following part.

423 The livestock distribution maps from the GLIMS dataset were the reference of the spatial pattern
424 of manure nutrient production data within each county. The GLIMS data were developed by
425 establishing statistical relationships between livestock inventory data and multiple environmental
426 variables (e.g., climate, land cover, human activities), and using these relationships to predict
427 livestock distributions across the globe. We assumed livestock distribution within each county
428 was stable over the study period because the dynamic livestock maps were unavailable.
429 However, the environmental variables can change and induce the variation in livestock

430 distribution inside each county. The accuracy of this manure nutrient production dataset can be
431 improved once dynamic livestock maps are developed in the future.

432 The manure nutrient production before 1930 was generated based on change rates in global
433 manure N datasets from Holland et al. (2005). There is a period of overlap (1930-2004) between
434 this global dataset and the USDA census data. During 1930-2004, the average annual change
435 rates of manure N production were 1.08% in the global dataset and 1.01% in this study.
436 Therefore, the changes in estimated manure N production in the U.S. before 1930 might be
437 reasonable at a long-time scale. The ratio of N to P in animal manure varies among different
438 animal species and changes along with proportions of different animal populations over time.
439 From 1930 to 2017, the N:P ratio in the total manure production slightly decreased from 4.33 to
440 3.25. Due to the lack of manure P production data before 1930, we calculated manure P
441 production in this period according to manure N production and the constant N:P ratio in 1930. If
442 the N:P ratio kept decreasing before 1930, the total manure P production may be overestimated
443 during 1860-1929.

444 Changes in recoverability factors and crop yields over the study period were ignored due to lack
445 of data support and that may cause a bias in quantifying manure nutrient application. With the
446 development of livestock confinement facilities, the confinement and recoverability factors of
447 animal manure may increase in recent decades (Kellogg et al., 2000). Hence, manure application
448 can be overestimated before the 1980s and underestimated after the 2000s. The yields of
449 different crops may change at different speeds over the study period, and that can affect the
450 spatial patterns of manure nutrient demand of cropland as well as manure nutrient application.

451 In addition, the development of manure application data was based on two assumptions: (1) The
452 allocation of manure nutrient application within the county was proportional to crop nutrient

453 demands; (2) Manure is assumed to be applied in the county where it was produced. Manure
454 application is controlled by distance, cost, and operating practice of humans. Currently, the
455 specific locations of animal farms across the country are not available, thus it is difficult to
456 evaluate the influence of distance between farms and croplands. Due to the practical limits of
457 manure transportation (Buckwell and Nadeu, 2016; MacDonald, 2009), it is reasonable to
458 assume manure production and application happen within the same county on a large scale.
459 However, ignoring the impact of multiple factors on manure application within the county can
460 still result in biases in spatial distribution of manure application.

461 **5 Data availability**

462 The gridded datasets of manure N and P production and application in the contiguous U.S. are
463 available at <https://doi.org/10.1594/PANGAEA.919937> (Bian et al., 2020). A supplement is
464 added to provide information about manure demand and all parameters that used to develop the
465 datasets.

466 **6 Conclusion**

467 Manure nutrient production and application in the livestock-crop system substantially altered the
468 regional and global N and P cycle. In this study, we developed geographically explicit datasets of
469 animal manure N and P production and their application in cropland across the contiguous U.S.
470 from 1860 to 2017. The dataset indicated that both manure N and P production and application
471 significantly increased over the study period. Although livestock numbers became stable in
472 recent decades, manure nutrient production still increased due to the enhanced livestock body
473 weight after the 1980s. Enhanced livestock weights contributed 59% and 54% of the increase in
474 manure N and P production, respectively, from 1987 to 2017. Meanwhile, manure nutrient

475 production intensified and enlarged inside the Midwest and toward the Southern U.S. from 1980
476 to 2017, and became more concentrated in numerous hot spots. As manure nutrient application
477 also expanded toward the Southeastern coastline, massive amounts of nutrients get more of a
478 chance to be transported to the estuary. The enrichment of manure nutrient in the South Atlantic-
479 Gulf, Mid-Atlantic, and Mississippi River basins increased the risk of excessive nutrient loading
480 into the Gulf of Mexico and the Atlantic Ocean under extreme weather conditions (e.g.,
481 hurricane). Therefore, it is of great importance to effectively store, utilize, and transport animal
482 manure in order to reduce nutrient pollution and restore the environment.

483

484 **Author contributions**

485 HT designed and led this work. ZB is responsible for developing the datasets. QY provided the
486 county-level livestock dataset. RX proposed the methods in the study. SP and BZ analyzed the
487 results. All authors contributed to the writing of the manuscript.

488 **Competing interests**

489 The authors declare that they have no conflict of interest.

490 **Acknowledgments**

491 This study has been supported in part by National Science Foundation grant (1903722); National
492 Oceanic and Atmospheric Administration grants (NA16NOS4780204, NA16NOS4780207); the
493 National Aeronautics and Space Administration grants (NNX12AP84G, NNX14AO73G,
494 NNX10AU06G, NNX14AF93G), and OUC-AU Joint Center Program.

495 **References**

- 496 Amundson, R., Berhe, A. A., Hopmans, J. W., Olson, C., Sztein, A. E. and Sparks, D. L.: Soil and
497 human security in the 21st century, *Science*, 348(6235), 1261071–1261071,
498 doi:10.1126/science.1261071, 2015.
- 499 Bian, Z., Tian, H., Yang, Q., Xu, R., Pan, S. and Zhang, B.: Gridded datasets of animal manure
500 nitrogen and phosphorus production and application in the continental U.S. from 1860 to 2017,
501 [online] Available from: <https://doi.org/10.1594/PANGAEA.919937> (Accessed 10 July 2020),
502 2020.
- 503 Billen, G., Lancelot, C. and Meybeck, M.: N, P, and Si retention along the aquatic continuum
504 from land and ocean, in Dahlem workshop on ocean margin processes in global change, pp. 19–
505 44., 1991.
- 506 Bouwman, A. F., Boumans, L. J. M. and Batjes, N. H.: Estimation of global NH₃ volatilization
507 loss from synthetic fertilizers and animal manure applied to arable lands and grasslands, *Glob.*
508 *Biogeochem. Cycles*, 16(2), 8-1-8–14, 2002.
- 509 Bouwman, A. F., Beusen, A. H. and Billen, G.: Human alteration of the global nitrogen and
510 phosphorus soil balances for the period 1970–2050, *Glob. Biogeochem. Cycles*, 23(4), 2009.
- 511 Bouwman, L., Goldewijk, K. K., Van Der Hoek, K. W., Beusen, A. H. W., Van Vuuren, D. P.,
512 Willems, J., Rufino, M. C. and Stehfest, E.: Exploring global changes in nitrogen and
513 phosphorus cycles in agriculture induced by livestock production over the 1900–2050 period,
514 *Proc. Natl. Acad. Sci.*, 110(52), 20882–20887, doi:10.1073/pnas.1012878108, 2013.
- 515 Buckwell, A. and Nadeu, E.: Nutrient Recovery and Reuse (NRR) in European agriculture. A
516 review of the issues, opportunities, and actions, Bruss. RISE Found., 2016.
- 517 David, M. B., Drinkwater, L. E. and McIsaac, G. F.: Sources of nitrate yields in the Mississippi
518 River Basin, *J. Environ. Qual.*, 39(5), 1657–1667, 2010.
- 519 Davidson, E. A.: The contribution of manure and fertilizer nitrogen to atmospheric nitrous oxide
520 since 1860, *Nat. Geosci.*, 2(9), 659, 2009.
- 521 Del Giudice, D., Matli, V. R. R. and Obenour, D. R.: Bayesian mechanistic modeling
522 characterizes Gulf of Mexico hypoxia: 1968–2016 and future scenarios, *Ecol. Appl.*,
523 doi:10.1002/eap.2032, 2019.
- 524 Eggleston, S., Buendia, L., Miwa, K., Ngara, T. and Tanabe, K.: 2006 IPCC guidelines for
525 national greenhouse gas inventories, Institute for Global Environmental Strategies Hayama,
526 Japan., 2006.
- 527 Elser, J. and Bennett, E.: A broken biogeochemical cycle, *Nature*, 478(7367), 29–31,
528 doi:10.1038/478029a, 2011.

- 529 Feyereisen, G. W., Kleinman, P. J., Folmar, G. J., Saporito, L. S., Way, T. R., Church, C. D. and
530 Allen, A. L.: Effect of direct incorporation of poultry litter on phosphorus leaching from coastal
531 plain soils, *J. Soil Water Conserv.*, 65(4), 243–251, 2010.
- 532 Garnier, J., Lassaletta, L., Billen, G., Romero, E., Grizzetti, B., Némery, J., Le, T. P. Q.,
533 Pistocchi, C., Aissa-Grouz, N. and Luu, T. N. M.: Phosphorus budget in the water-agro-food
534 system at nested scales in two contrasted regions of the world (ASEAN-8 and EU-27), *Glob.*
535 *Biogeochem. Cycles*, 29(9), 1348–1368, 2015.
- 536 He, Z., Pagliari, P. H. and Waldrip, H. M.: Applied and environmental chemistry of animal
537 manure: A review, *Pedosphere*, 26(6), 779–816, 2016.
- 538 Holland, E. A., Lee-Taylor, J., Nevison, C. and Sulzman, J. M.: Global N Cycle: fluxes and N₂O
539 mixing ratios originating from human activity, ORNL DAAC, 2005.
- 540 Jones, C. S., Drake, C. W., Hruby, C. E., Schilling, K. E. and Wolter, C. F.: Livestock manure
541 driving stream nitrate, *Ambio*, 48(10), 1143–1153, 2019.
- 542 Kellogg, R. L., Lander, C. H., Moffitt, D. C. and Gollehon, N.: Manure nutrients relative to the
543 capacity of cropland and pastureland to assimilate nutrients, US Dep. Agric. Nat. Resour.
544 Conserv. Serv. Agric. Res. Serv., 2000.
- 545 Klein Goldewijk, K., Beusen, A., Doelman, J. and Stehfest, E.: Anthropogenic land use estimates
546 for the Holocene – HYDE 3.2, *Earth Syst. Sci. Data*, 9(2), 927–953,
547 doi:<https://doi.org/10.5194/essd-9-927-2017>, 2017.
- 548 Koppelaar, R. H. E. M. and Weikard, H. P.: Assessing phosphate rock depletion and phosphorus
549 recycling options, *Glob. Environ. Change*, 23(6), 1454–1466,
550 doi:[10.1016/j.gloenvcha.2013.09.002](https://doi.org/10.1016/j.gloenvcha.2013.09.002), 2013.
- 551 Lassaletta, L., Billen, G., Grizzetti, B., Anglade, J. and Garnier, J.: 50 year trends in nitrogen use
552 efficiency of world cropping systems: the relationship between yield and nitrogen input to
553 cropland, *Environ. Res. Lett.*, 9(10), 105011, doi:[10.1088/1748-9326/9/10/105011](https://doi.org/10.1088/1748-9326/9/10/105011), 2014.
- 554 MacDonald, G. K., Bennett, E. M. and Taranu, Z. E.: The influence of time, soil characteristics,
555 and land-use history on soil phosphorus legacies: a global meta-analysis, *Glob. Change Biol.*,
556 18(6), 1904–1917, doi:<https://doi.org/10.1111/j.1365-2486.2012.02653.x>, 2012.
- 557 MacDonald, J. M.: Manure use for fertilizer and for energy: report to congress, DIANE
558 Publishing., 2009.
- 559 Monfreda, C., Ramankutty, N. and Foley, J. A.: Farming the planet: 2. Geographic distribution of
560 crop areas, yields, physiological types, and net primary production in the year 2000, *Glob.*
561 *Biogeochem. Cycles*, 22(1), 2008.
- 562 Mueller, N. D. and Lassaletta, L.: Nitrogen challenges in global livestock systems, *Nat. Food*,
563 1(7), 400–401, doi:[10.1038/s43016-020-0117-7](https://doi.org/10.1038/s43016-020-0117-7), 2020.

- 564 Oenema, O., Oudendag, D. and Velthof, G. L.: Nutrient losses from manure management in the
565 European Union, *Livest. Sci.*, 112(3), 261–272, doi:10.1016/j.livsci.2007.09.007, 2007.
- 566 Potter, P., Ramankutty, N., Bennett, E. M. and Donner, S. D.: Characterizing the Spatial Patterns
567 of Global Fertilizer Application and Manure Production, *Earth Interact.*, 14(2), 1–22,
568 doi:10.1175/2009EI288.1, 2010.
- 569 Powers, S. M., Chowdhury, R. B., MacDonald, G. K., Metson, G. S., Beusen, A. H. W.,
570 Bouwman, A. F., Hampton, S. E., Mayer, B. K., McCrackin, M. L. and Vaccari, D. A.: Global
571 Opportunities to Increase Agricultural Independence Through Phosphorus Recycling, *Earths*
572 *Future*, 7(4), 370–383, doi:10.1029/2018EF001097, 2019.
- 573 Puckett, L., Hitt, K. and Alexander, R.: County-based estimates of nitrogen and phosphorus
574 content of animal manure in the United States for 1982, 1987, and 1992, US Geological Survey.,
575 1998.
- 576 Rabalais, N. N. and Turner, R. E.: Gulf of Mexico Hypoxia: Past, Present, and Future, *Limnol.*
577 *Oceanogr. Bull.*, 28(4), 117–124, doi:10.1002/lob.10351, 2019.
- 578 Robinson, T. P., Wint, G. R. W., Conchedda, G., Van Boeckel, T. P., Ercoli, V., Palamara, E.,
579 Cinardi, G., D’Aielli, L., Hay, S. I. and Gilbert, M.: Mapping the Global Distribution of
580 Livestock, edited by M. Baylis, *PLoS ONE*, 9(5), e96084, doi:10.1371/journal.pone.0096084,
581 2014.
- 582 Rowe, H., Withers, P. J. A., Baas, P., Chan, N. I., Doody, D., Holiman, J., Jacobs, B., Li, H.,
583 MacDonald, G. K., McDowell, R., Sharpley, A. N., Shen, J., Taheri, W., Wallenstein, M. and
584 Weintraub, M. N.: Integrating legacy soil phosphorus into sustainable nutrient management
585 strategies for future food, bioenergy and water security, *Nutr. Cycl. Agroecosystems*, 104(3),
586 393–412, doi:10.1007/s10705-015-9726-1, 2016.
- 587 Ruddy, B. C., Lorenz, D. L. and Mueller, D. K.: County-level estimates of nutrient inputs to the
588 land surface of the conterminous United States, 1982-2001, 2006.
- 589 Saunders, M. A. and Lea, A. S.: Large contribution of sea surface warming to recent increase in
590 Atlantic hurricane activity, *Nature*, 451(7178), 557–560, doi:10.1038/nature06422, 2008.
- 591 Schlesinger, W. H. and Bernhardt, E. S.: *Biogeochemistry: An Analysis of Global Change*,
592 Academic Press., 2013.
- 593 Seitzinger, S. P., Styles, R. V., Boyer, E. W., Alexander, R. B., Billen, G., Howarth, R. W., Mayer,
594 B. and Van Breemen, N.: Nitrogen retention in rivers: model development and application to
595 watersheds in the northeastern USA, in *The nitrogen cycle at regional to global scales*, pp. 199–
596 237, Springer., 2002.
- 597 Sheldrick, W., Syers, J. K. and Lingard, J.: Contribution of livestock excreta to nutrient balances,
598 *Nutr. Cycl. Agroecosystems*, 66(2), 119–131, 2003.

- 599 Sheldrick, W. F., Syers, J. K. and Lingard, J.: A conceptual model for conducting nutrient audits
600 at national, regional, and global scales, *Nutr. Cycl. Agroecosystems*, 62(1), 61–72, 2002.
- 601 Smith, D. R., Owens, P. R., Leytem, A. B. and Warnemuende, E. A.: Nutrient losses from manure
602 and fertilizer applications as impacted by time to first runoff event, *Environ. Pollut.*, 147(1),
603 131–137, 2007.
- 604 Smith, K. A., Chalmers, A. G., Chambers, B. J. and Christie, P.: Organic manure phosphorus
605 accumulation, mobility and management, *Soil Use Manag.*, 14(s4), 154–159, doi:10.1111/j.1475-
606 2743.1998.tb00634.x, 1998.
- 607 Tabachow, R. M., Peirce, J. J. and Essiger, C.: Hurricane-Loaded Soil, *J. Environ. Qual.*, 30(6),
608 1904–1910, doi:10.2134/jeq2001.1904, 2001.
- 609 Thornton, P. K.: Livestock production: recent trends, future prospects, *Philos. Trans. R. Soc. B*
610 *Biol. Sci.*, 365(1554), 2853–2867, doi:10.1098/rstb.2010.0134, 2010.
- 611 Tian, H., Lu, C., Ciais, P., Michalak, A. M., Canadell, J. G., Saikawa, E., Huntzinger, D. N.,
612 Gurney, K. R., Sitch, S. and Zhang, B.: The terrestrial biosphere as a net source of greenhouse
613 gases to the atmosphere, *Nature*, 531(7593), 225, 2016.
- 614 Tian, H., Yang, J., Xu, R., Lu, C., Canadell, J. G., Davidson, E. A., Jackson, R. B., Arneeth, A.,
615 Chang, J., Ciais, P., Gerber, S., Ito, A., Joos, F., Lienert, S., Messina, P., Olin, S., Pan, S., Peng,
616 C., Saikawa, E., Thompson, R. L., Vuichard, N., Winiwarter, W., Zaehle, S. and Zhang, B.:
617 Global soil nitrous oxide emissions since the preindustrial era estimated by an ensemble of
618 terrestrial biosphere models: Magnitude, attribution, and uncertainty, *Glob. Change Biol.*, 25(2),
619 640–659, doi:10.1111/gcb.14514, 2019.
- 620 Tian, H., Xu, R., Canadell, J. G., Thompson, R. L., Winiwarter, W., Suntharalingam, P.,
621 Davidson, E. A., Ciais, P., Jackson, R. B., Janssens-Maenhout, G., Prather, M. J., Regnier, P.,
622 Pan, N., Pan, S., Peters, G. P., Shi, H., Tubiello, F. N., Zaehle, S., Zhou, F., Arneeth, A., Battaglia,
623 G., Berthet, S., Bopp, L., Bouwman, A. F., Buitenhuis, E. T., Chang, J., Chipperfield, M. P.,
624 Dangal, S. R. S., Dlugokencky, E., Elkins, J. W., Eyre, B. D., Fu, B., Hall, B., Ito, A., Joos, F.,
625 Krummel, P. B., Landolfi, A., Laruelle, G. G., Lauerwald, R., Li, W., Lienert, S., Maavara, T.,
626 MacLeod, M., Millet, D. B., Olin, S., Patra, P. K., Prinn, R. G., Raymond, P. A., Ruiz, D. J., van
627 der Werf, G. R., Vuichard, N., Wang, J., Weiss, R. F., Wells, K. C., Wilson, C., Yang, J. and Yao,
628 Y.: A comprehensive quantification of global nitrous oxide sources and sinks, *Nature*, 586(7828),
629 248–256, doi:10.1038/s41586-020-2780-0, 2020.
- 630 Trenberth, K.: Uncertainty in Hurricanes and Global Warming, *Science*, 308(5729), 1753–1754,
631 doi:10.1126/science.1112551, 2005.
- 632 Van Drecht, G., Bouwman, A. F., Boyer, E. W., Green, P. and Siebert, S.: A comparison of global
633 spatial distributions of nitrogen inputs for nonpoint sources and effects on river nitrogen export:
634 GLOBAL NEWS-COMPARISON OF GLOBAL NITROGEN INPUTS, *Glob. Biogeochem.*
635 *Cycles*, 19(4), n/a-n/a, doi:10.1029/2005GB002454, 2005.

636 Williams, M. R., Feyereisen, G. W., Beegle, D. B., Shannon, R. D., Folmar, G. J. and Bryant, R.
637 B.: Manure application under winter conditions: Nutrient runoff and leaching losses, *Trans.*
638 *ASABE*, 54(3), 891–899, 2011.

639 Xu, R. T., Pan, S. F., Chen, J., Chen, G. S., Yang, J., Dangal, S. R. S., Shepard, J. P. and Tian, H.
640 Q.: Half-century ammonia emissions from agricultural systems in Southern Asia: Magnitude,
641 spatiotemporal patterns, and implications for human health, *GeoHealth*, 2(1), 40–53, 2018.

642 Yang, Q., Tian, H., Li, X., Ren, W., Zhang, B., Zhang, X. and Wolf, J.: Spatiotemporal patterns
643 of livestock manure nutrient production in the conterminous United States from 1930 to 2012,
644 *Sci. Total Environ.*, 541, 1592–1602, doi:10.1016/j.scitotenv.2015.10.044, 2016.

645 Yu, Z. and Lu, C.: Historical cropland expansion and abandonment in the continental U.S. during
646 1850 to 2016, *Glob. Ecol. Biogeogr.*, 27(3), 322–333, doi:10.1111/geb.12697, 2018.

647 Zanon, J. A., Favaretto, N., Goularte, G. D., Dieckow, J. and Barth, G.: Manure application at
648 long-term in no-till: Effects on runoff, sediment and nutrients losses in high rainfall events,
649 *Agric. Water Manag.*, 105908, 2019.

650 Zhang, B., Tian, H., Lu, C., Dangal, S. R. S., Yang, J. and Pan, S.: Global manure nitrogen
651 production and application in cropland during 1860–2014: a 5 arcmin gridded global dataset for
652 Earth system modeling, *Earth Syst. Sci. Data*, 9(2), 667–678, doi:10.5194/essd-9-667-2017,
653 2017.

654 Zhang, X., Davidson, E. A., Zou, T., Lassaletta, L., Quan, Z., Li, T. and Zhang, W.: Quantifying
655 Nutrient Budgets for Sustainable Nutrient Management, *Glob. Biogeochem. Cycles*, 34(3),
656 e2018GB006060, doi:10.1029/2018GB006060, 2020.

657

658

659

660

661

662

663

664

665

666

667

668

669

670

Table 1. Summary of data sources

Data variables	Time period	Resolution	Reference/source
Livestock numbers	1930-2017	County-level	USDA National Agricultural Statistics Service https://www.nass.usda.gov/index.php
Livestock weights	1921-2017	Country-level	USDA Economic Research Service database http://www.ers.usda.gov/
Livestock distribution	2007	30 arc-second	Global Livestock Impact Mapping System (GLIMS) (Robinson et al., 2014)
Manure recoverability rates	1987-2014	County-level	Nutrient Use Geographic Information System (NuGIS) http://nugis.ipni.net/
Crop harvested area and yield	2000	5 arc-min	(Monfreda et al., 2008)
Crop and pasture distributions	1860-2016	5 arc-min	History Database of the Global Environment (HYDE 3.2) (Klein Goldewijk et al., 2017)
Crop density	1850-2016	1×1 km	(Yu and Lu, 2018)

671

672

673

674

675

676

677

678



679

680 **Figure 1. Eighteen Hydrologic Units in the contiguous U.S. (Recreated from the U.S. hydrologic unit**
681 **map: <https://water.usgs.gov/GIS/regions.html>)**

682

683

684

685

686

687

688

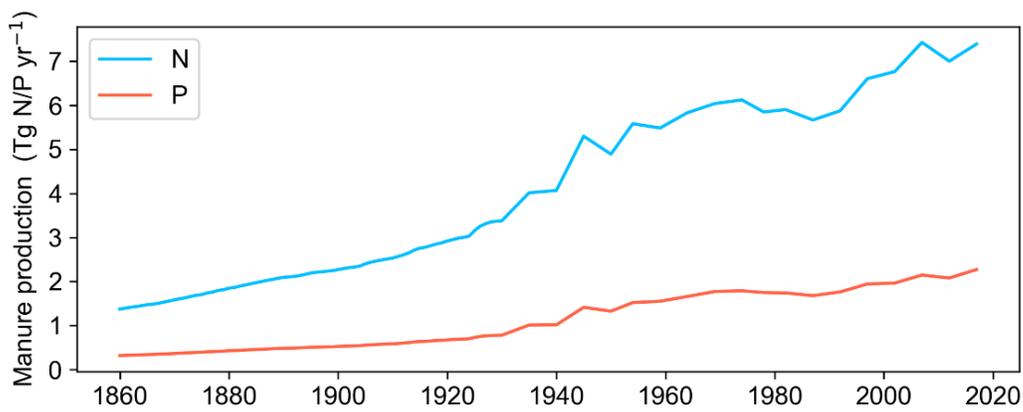
689

690

691

692

693



694

695

696

Figure 2. Trend and variation of total manure N and P production in the contiguous U.S from 1860 to 2017

697

698

699

700

701

702

703

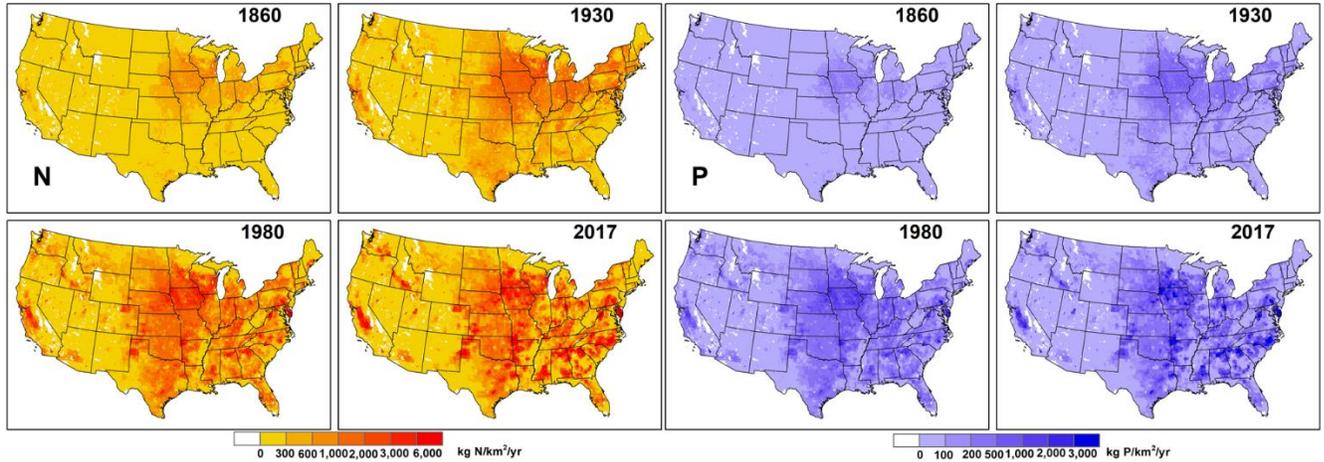
704

705

706

707

708



709

710

Figure 3. Spatial distribution of manure N and P production across the contiguous U.S. in 1860, 1930, 1980, and 2017. (Note: 1930 and 2017 were the earliest and latest years of available USDA census data, respectively, and 1980 was chosen as the year at the middle of these two years)

712

713

714

715

716

717

718

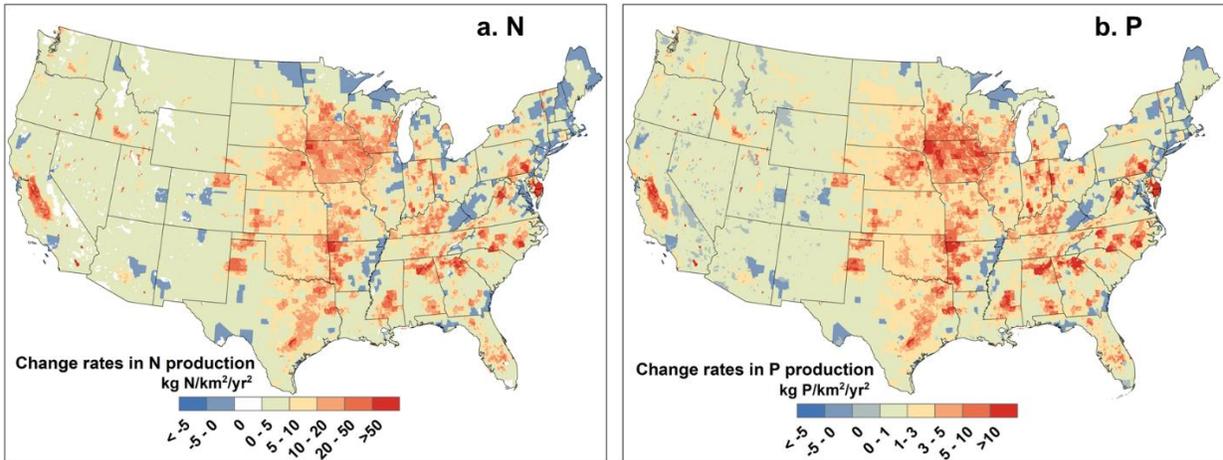
719

720

721

722

723



724

725 Figure 4. Change rates of manure (a) N and (b) P production across the contiguous U.S. during 1860-
 726 2017. (Note: the increasing rates were calculated based on the Mann-Kendall Test)

727

728

729

730

731

732

733

734

735

736

737

738

739

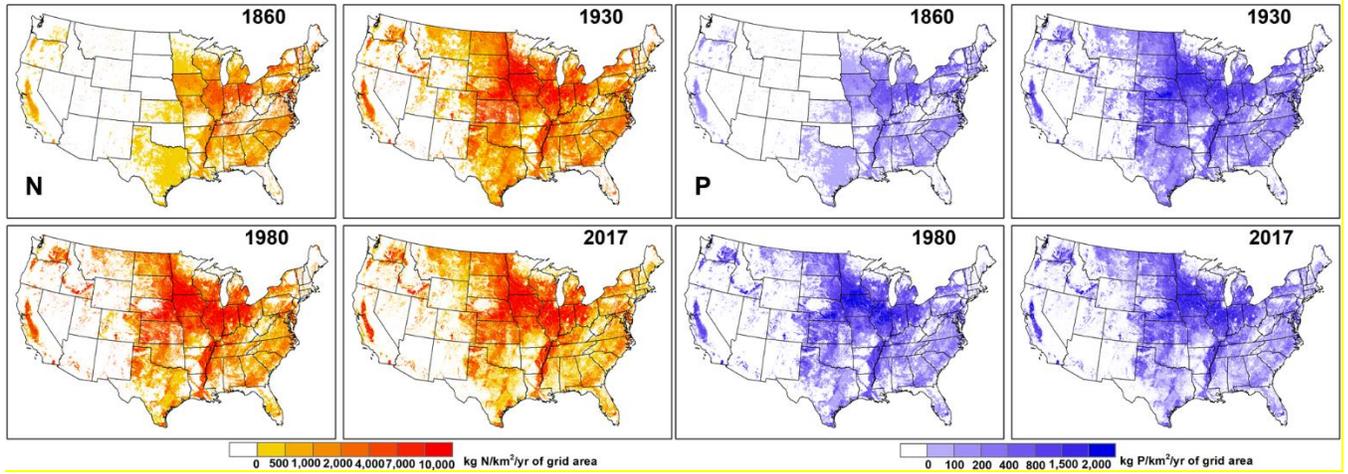
740

741

742

743

744



745

746

Figure 5. Spatial distribution of N and P demand of crops in 1860, 1930, 1980, and 2017.

747

748

749

750

751

752

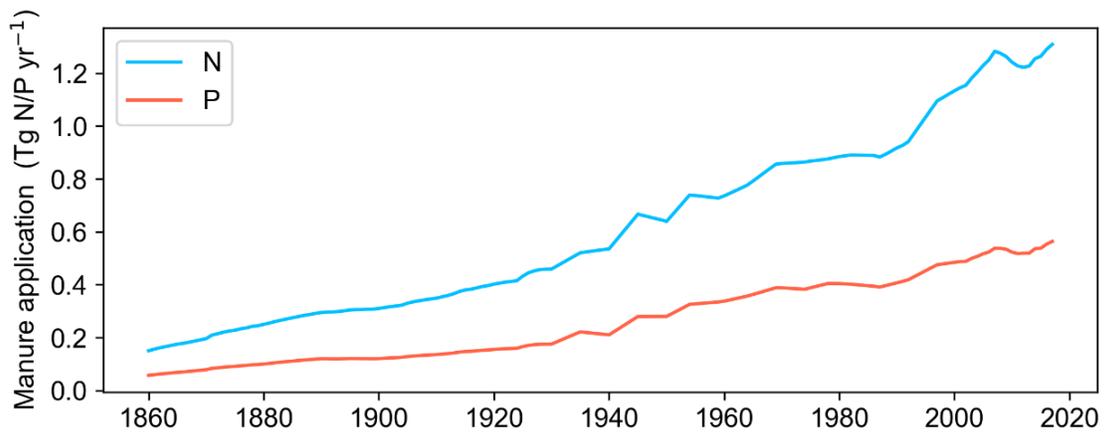
753

754

755

756

757



758

759 Figure 6. Trend and variations of total manure N and P application in the contiguous U.S. from 1860 to
 760 2017

761

762

763

764

765

766

767

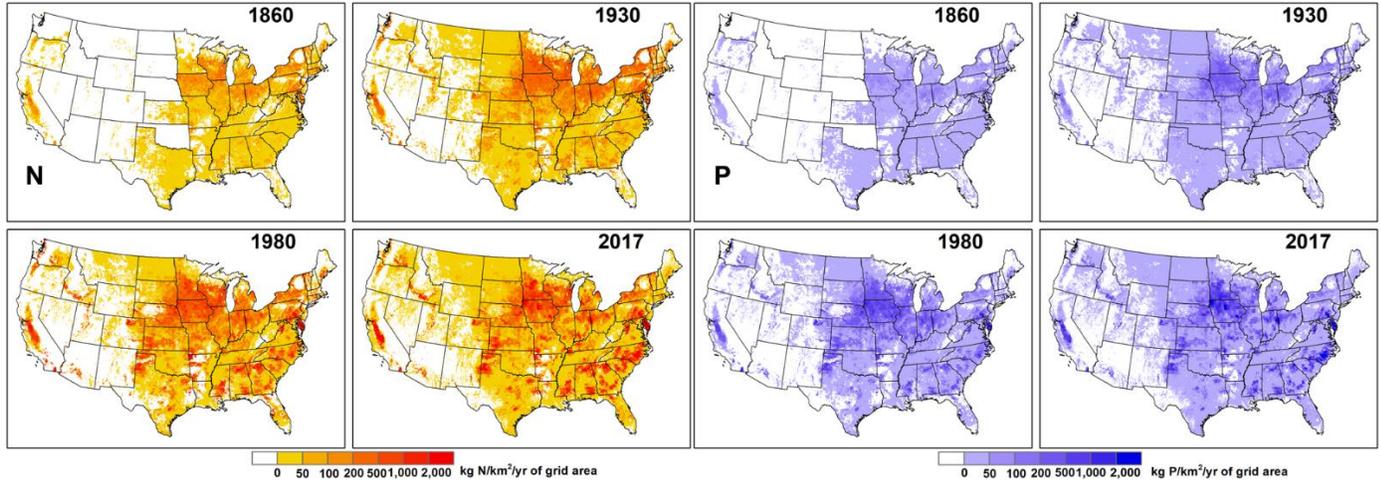
768

769

770

771

772



773

774

775 Figure 7. Spatial distributions of manure N and P application in the U.S. cropland in 1860, 1930, 1980,
 776 and 2017.

777

778

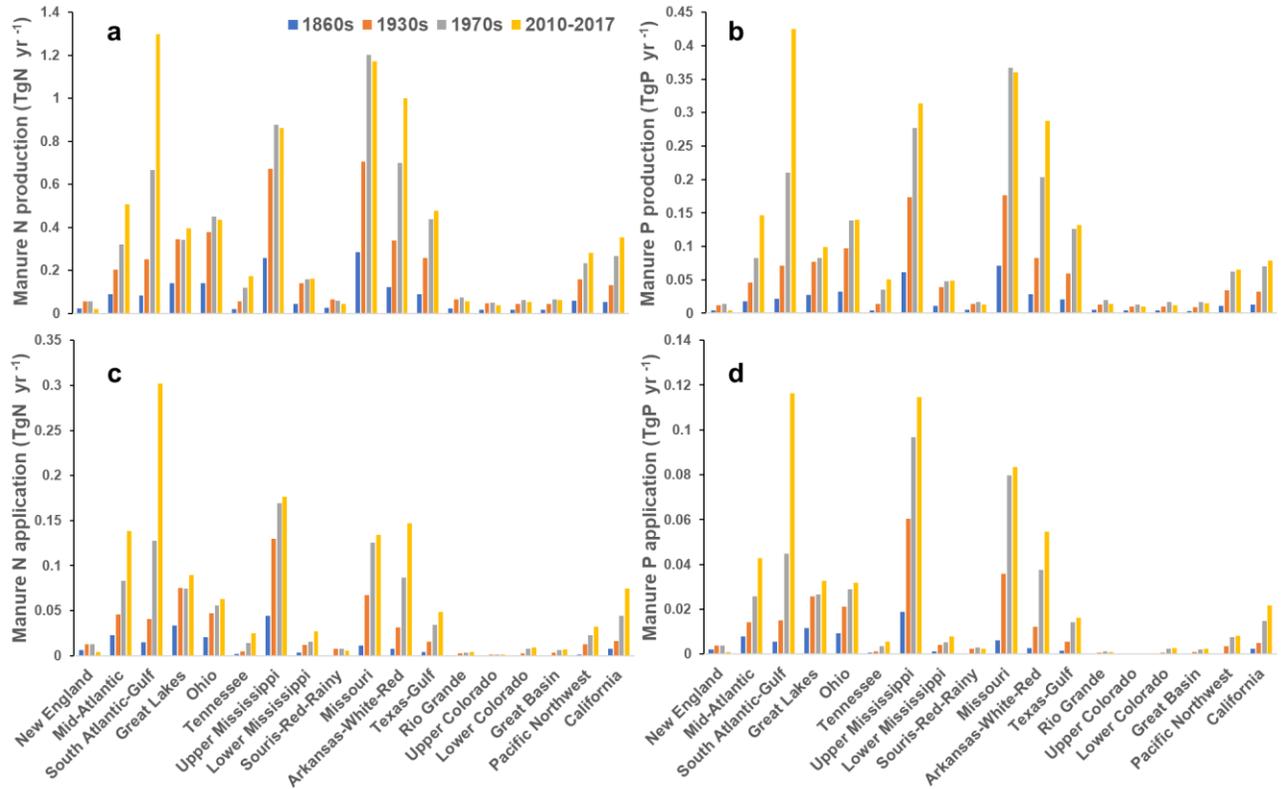
779

780

781

782

783



784

785 Figure 8. Average annual manure production (a. N, b. P) and application (c. N, d. P) across 18 major

786

basins in the 1860s, 1930s, 1970s, and 2010-2017.

787

788

789

790

791

792

793

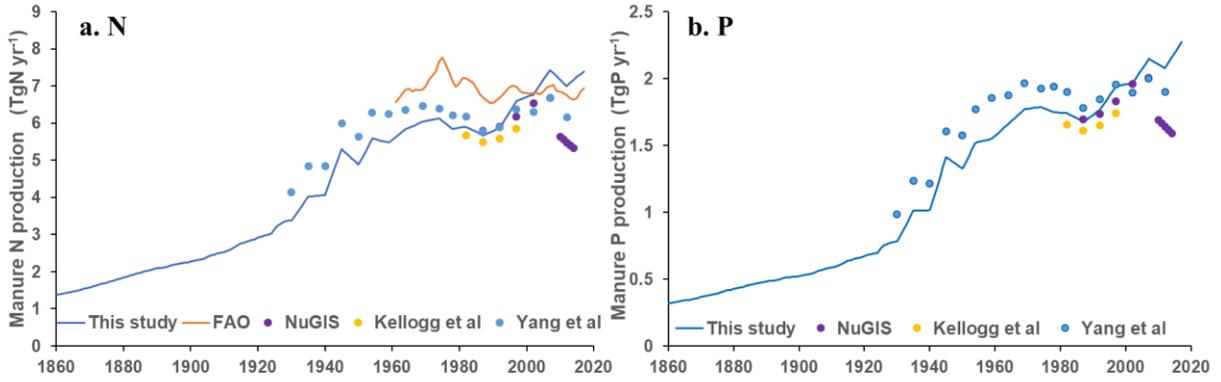
794

795

796

797

798



799

800 Figure 9. Comparison of manure nutrients production in this study with the four previous datasets.

801

802

803

804

805

806

807

808

809

810

811

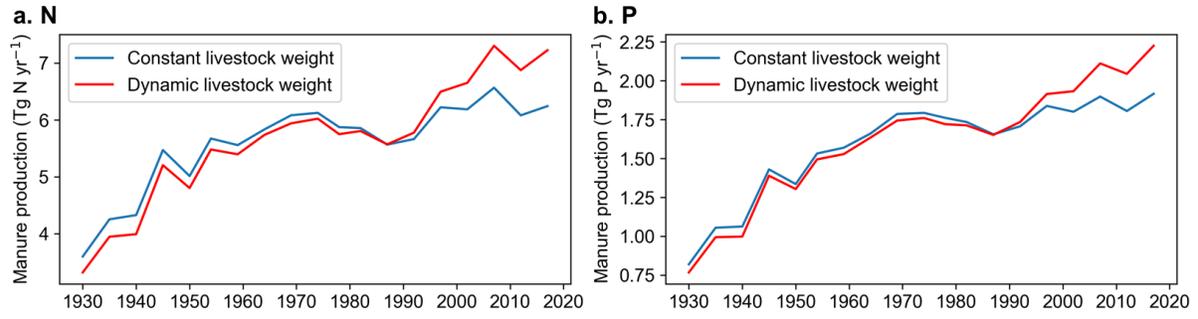
812

813

814

815

816



817

818

819

Figure 10. Comparison of manure N (P) production calculated based on dynamic weight of livestock and constant weight.

820