

Increased nitrogen enrichment and shifted patterns in the world's grassland: 1860-2016

Rongting Xu¹, Hanqin Tian^{1,2}, Shufen Pan¹, Shree R. S. Dangal^{3,1}, Jian Chen^{4,1}, Jinfeng Chang⁵, Yonglong Lu², Ute Maria Skiba⁶, [Francesco N. Tubiello](#)⁷ and Bowen Zhang^{1#}

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

²Research Center for Eco-Environmental Sciences, State Key Laboratory of Urban and Regional Ecology, Chinese Academy of Sciences, Beijing 100085, China

³Woods Hole Research Center, Falmouth, Massachusetts 02540, USA

⁴Department of Computer Science and Software Engineering, Samuel Ginn College of Engineering, Auburn University, Auburn, AL 36849, USA

⁵Laboratoire des Sciences du Climat et de l'Environnement, LSCE, 91191 Gif sur Yvette, France

⁶Centre for Ecology and Hydrology, Bush Estate, Penicuik EH26 0QB, United Kingdom;

⁷[Statistics Division, Food and Agricultural Organization of the United Nations, Via Terme di Caracalla, Rome 00153 Italy](#)

[#][Present Address: Department of Natural Resources and Environmental Management, Ball State University, 2000 W. University Ave., Muncie, IN 47306, USA](#)

Correspondence to: Dr. Hanqin Tian (tianhan@auburn.edu)

1 **Abstract**

2 Production and application to soils of manure excreta from livestock production significantly
3 perturb the global nutrient balance and result in significant greenhouse gas emissions that warm
4 the earth's climate. Despite much attention paid to synthetic nitrogen (N) fertilizer and manure N
5 applications to croplands, spatially-explicit, continuous time-series datasets of manure and
6 fertilizer N inputs on pastures and rangelands are lacking. We developed three global gridded
7 datasets at a resolution of $0.5^\circ \times 0.5^\circ$ for the period 1860–2016 (i.e., annual manure N deposition
8 (by grazing animals) rate, synthetic N fertilizer and N manure application rates), by combining
9 annual and 5-arc minute spatial data on pastures and rangelands with country-level statistics on
10 livestock manure, mineral and chemical fertilizers, and land use information for cropland and
11 permanent meadows and pastures. Based on the new data products, we estimated that total N
12 inputs, sum of manure N deposition, manure and fertilizer N application to pastures and
13 rangelands increased globally from 15 to 101 Tg N yr⁻¹ during 1860–2016. In particular during
14 the period 2000-2016, livestock manure N deposition accounted for 83% of the total N inputs,
15 whereas manure and fertilizer N application accounted 9% and 8%, respectively. At the regional
16 scale, hotspots of manure N deposition remained largely similar during the period 1860–2016
17 (i.e., southern Asia, Africa, and South America), however hotspots of manure and fertilizer N
18 application shifted from Europe to southern Asia in the early 21st century. The new three global
19 datasets contribute to fill previous data gaps of global and regional N inputs in pastures and
20 rangelands, improving the abilities of ecosystem and earth system models to investigate the
21 global impacts of N enrichment due to agriculture, in terms of associated greenhouse gas
22 emissions and environmental sustainability issues. Datasets are available at
23 <https://doi.pangaea.de/10.1594/PANGAEA.892940>.

24 **Keywords:** nitrogen, manure, fertilizers, pasture, rangeland, FAOSTAT

25 **1 Introduction**

26 Livestock production has increased substantially in response to growing meat consumption
27 across the globe in the past century (Bouwman et al., 2013; Dangal et al., 2017). Agriculture
28 occupies 37% of Earth's ice-free land surface for use as cropland and permanent meadows and
29 pastures (Tubiello, 2018). Land used by livestock for permanent meadows and pastures is the
30 largest component, using 25% of the total land earth surface (FAOSTAT, 2018) to generate
31 33%–50% of world total agricultural GDP (Herrero et al., 2013). While livestock is a major
32 source of income for more than 1.3 billion people, it is also a major user of crop and freshwater
33 resources (Dangal et al., 2017; Herrero et al., 2013). Overall, livestock production plays a major
34 role as driver of global change in land use and nutrient cycles (Havlík et al., 2014; Herrero et al.,
35 2013; Zhang et al., 2017). There is a growing recognition that livestock production is linked to
36 increasing global greenhouse gas (GHGs) and ammonia emissions (Tian et al., 2016; Tubiello et
37 al., 2018; Xu et al., 2018). Unsustainable practices, especially in intensive systems, may lead to
38 severe pollution of aquatic systems and soil degradation locally, regional and globally, in
39 particular through nitrate leaching to water bodies (Dangal et al., 2017; Davis et al., 2015;
40 Fowler et al., 2013; Yang et al., 2016). Growing global demand for livestock products has
41 increased grain production for feed in many regions, and has become a global driver of fertilizers
42 trends, through increase in manure availability and synthetic fertilizer N use (FAOSTAT, 2018).

43 Livestock production systems therefore play an important role in global nutrient cycles. For
44 example, nitrogen excretion from livestock increased from 21 Tg N yr⁻¹ in 1860 to 123 Tg N yr⁻¹
45 in 2016 (FAOSTAT, 2018; Zhang et al., 2017). Livestock contribute roughly two-thirds of non-
46 CO₂ GHG emissions from agriculture (Smith et al., 2014), with roughly an equal share of CH₄
47 and N₂O emissions (Dangal et al., 2017; Tubiello et al., 2013). Importantly, about 45% of total

48 anthropogenic N₂O emissions are linked to manure deposited through grazing and manure
49 applied to croplands or left on pasture (Davidson, 2009; FAOSTAT, 2018). Globally, emissions
50 from manure N applied to soils or left on pastures has increased from 0.44 to 0.88 GtCO₂eq yr⁻¹
51 during 1961–2010 (FAOSTAT, 2018). [Increased meat and dairy products consumption](#)
52 worldwide was a major driver behind the documented increase in cattle herds globally
53 (FAOSTAT, 2018), and thus a major cause in the observed atmospheric increase of N₂O and
54 CH₄ over the past several decades (Bai et al., 2018; Bouwman et al., 2013; Dangal et al., 2017;
55 Tubiello, 2018).

56 While the availability of national-level statistics is a fundamental component of our knowledge
57 base, environmental problems related to nitrogen pollution or emissions are best tackled at local
58 scale and often require finer, geo-spatial information, for example to assess proximity to water
59 bodies and thus pollution risks. In particular, a number of studies have focused on downscaling
60 existing national information to develop geospatially explicit regional and global datasets of
61 nitrogen fertilizer and livestock manure production and use, to better understand their feedback
62 on the climate system. Several datasets of N fertilizer use were used in this study, in particular
63 the FAOSTAT annual, country-specific statistics on mineral and chemical fertilizers and
64 livestock manure over the period 1961-2016 (FAOSTAT, 2018), as well as specific geospatially-
65 downscaled products (e.g., Bouwman et al., 2005; Lu and Tian, 2017; Mueller et al., 2012;
66 Nishina et al., 2017; Potter et al., 2010; Sheldrick et al., 2002). Further, global manure
67 production datasets were developed in different studies to achieve various research goals
68 (Bouwman et al., 2009; Bouwman et al., 2013; Holland et al.; Potter et al., 2010; Zhang et al.,
69 2017). Although datasets of manure application in croplands are increasingly available, there is
70 considerable uncertainty in the estimation of total manure application and their spatial

71 distribution across different studies (Gerber et al., 2016; Herrero et al., 2013; Liu et al., 2010;
72 Zhang et al., 2017).

73 Although previous studies have provided spatially explicit datasets of N inputs in the form of
74 mineral or chemical and manure N in cropland systems, the spatially explicit datasets on N
75 inputs in grassland systems are still missing (Hauglustaine, 2016; Lassaletta et al., 2014; Stehfest
76 and Bouwman, 2006). By grassland systems we mean the FAO livestock land use definition, i.e.,
77 land used as permanent meadows and pastures (FAOSTAT, 2018). The same may also be
78 referred to in the literature as ‘pastures and rangelands’. We note that ‘grassland’ is in fact a land
79 cover definition. In order to avoid the confusion often made in the literature between land cover
80 and land use terminology, we will adopt FAO land use terminology of ‘permanent meadows and
81 pastures,’ to which the various national regional and global land use statistics cited in this work
82 refer. Furthermore, using results from the HYDE 3.2 dataset (Klein Goldewijk, 2017), we may
83 split the FAO land use category into ‘pastures’ and ‘rangelands’, to highlight differences
84 between managed intensive and unmanaged extensive systems, as needed. To enhance our
85 understanding of the role of livestock on the global GHG balance and nutrient budgets (e.g.,
86 ammonia emissions, nitrate leaching), global biogeochemistry models require spatially explicit
87 estimates of N inputs. In this study, we developed datasets for major sources of N inputs in
88 agriculture (i.e., manure and fertilizer application and manure deposition on permanent meadows
89 and pastures), using the recently published FAOSTAT statistics on manure N use in agriculture
90 (FAOSTAT, 2018). The latter are estimates based on IPCC Tier 1 methodology, i.e., they rely on
91 default coefficients prescribing, among other variables, N excretion rates by animal type and
92 region, as well as regional compositions of manure management systems (FAOSTAT, 2018).

93 Through combining the land-use dataset HYDE 3.2, FAOSTAT fertilizers N statistics, and
94 gridded manure production data in Zhang et al. (2017), we developed three annual global
95 datasets at a spatial resolution of $0.5^\circ \times 0.5^\circ$, as follows: 1) manure N application rates to
96 pastures (1860–2016); 2) synthetic N fertilizer application rates to pastures (1961–2016); and 3)
97 manure deposition rates by grazing livestock, to rangelands and pastures (1860–2016). We
98 quantified regional variations in N inputs, identified hotspots of N inputs from different N
99 sources from livestock, and discussed their uncertainty. These datasets are developed for global
100 model simulation studies in model inter-comparison projects (e.g., NMIP; Tian et al., 2018a;
101 Tian et al., 2018b), and will be updated annually based on regular annual updates of FAO
102 fertilizers and land use statistics and other sources of data such as global land use data products.

103 **2 Methods**

104 **2.1 Land Use Categories**

105 The concepts of grassland, pastures and meadows span several international land cover and land
106 use statistical definitions, specifically those used by FAO (FAOSTAT, 2018). [In this paper, we](#)
107 [follow the relevant FAO land use definition of ‘permanent meadows and pastures,’ considering](#)
108 [our focus on livestock production. Importantly, complete country, regional and global statistics](#)
109 [available from FAO refer to this land use category.](#) This land use definition is roughly equivalent
110 to the one adopted by the academic community engaged in global biogeochemical modeling, for
111 which ‘grassland systems’ are thought of as land cover/land use areas dominated by herbaceous
112 and shrub vegetation, including savannas (Africa, South America and India), steppes (Eurasia),
113 prairies (North America), shrub-dominated areas (Africa), meadows and pastures (United
114 Kingdom and Ireland) and tundra (Breymer, 1990; White et al., 2000).

115 For mineral and chemical fertilizers, we further split the FAO definition using HYDE 3.2, into
116 ‘pastures’ and ‘rangelands,’ the former representing land use areas managed to support high
117 stocking densities of grass production for hay/silage, whereas the latter represents unmanaged
118 and grazed at low stocking densities. Although FAOSTAT land-use statistics cover in principle
119 these two sub-categories of land use, data coverage needed is insufficient for the consistent
120 global mapping needed herein. The spatial distribution map of pastures and rangelands provided
121 by HYDE are nonetheless based on and normalized to FAOSTAT land use statistics,
122 complemented by additional information (Klein Goldewijk, 2017). To investigate N inputs from
123 livestock at a regional level, the global landmass was disaggregated into seven regions: North
124 America, South America, Africa, Europe, southern Asia (i.e., west, south, east, central and
125 southeast Asia), northern Asia, and Oceania (Fig. S1).

126 2.2 Global synthetic fertilizer N application on pastures

127 We obtained national-level datasets of “Agricultural use of mineral or chemical fertilizers” from
128 the FAOSTAT (2018) ‘Fertilizers by Nutrient’ domain, over the time series 1961–2016. The
129 FAOSTAT statistics of agricultural use include use for both agriculture and forestry, as well as
130 use in aquaculture. Furthermore, agricultural use includes both cropland and permanent
131 meadows and pastures. We assumed that fertilizers use for forestry and aquaculture was zero, as
132 well as fertilizers applications on rangelands. Subsequently, we estimated N application rates to
133 pastures by using the ratio of pasture to cropland N use total published by Lassaletta et al. (2014).
134 We finally spatialized the pasture N data using HYDE 3.2, obtaining gridded maps of synthetic
135 fertilizers N application rates on pastures in each grid cell area, over the period 1961–2016 (Fig.
136 1). We assumed even application rates within each country. [Although gridded livestock density
137 maps were available from FAO, these are currently fixed for specific time periods, mainly 2010,](#)

138 so that we deemed their use not particularly relevant to improve estimates for the 1961-2016
139 time series considered herein. Improved live density map products from FAO will considerably
140 improve our work and reduce uncertainty, and will be used when available.

141 2.3 Global manure N application to pastures

142 We obtained country-level datasets of “manure applied to soils” from the FAOSTAT (2018)
143 ‘Livestock Manure’ domain for the period 1961–2016 (FAO, 2018). Following IPCC guidelines,
144 the data in this domain do not consider N leaching during treatment (FAOSTAT, 2018).
145 Furthermore, the FAOSTAT data do not separate manure application to cropland and pastures
146 and data of manure N application rates to pastures are currently not available. We therefore
147 assumed that manure N application rates in pastures and croplands were the same, considering
148 that the overall uncertainty in the input manure N data would not justify further assumptions at
149 this stage of knowledge. Improved FAO statistics on both use and application rates will be used
150 when available to improve this current work. Through combining land-use data HYDE 3.2, we
151 calculated the total cropland and pasture areas within each country where manure application
152 amount was larger than zero. We then computed mean manure N application rates on pastures,
153 annually over the period 1961–2016 (Fig. 2).

154 We calculated the national-level ratio of manure application to production ($R_{a2p_{y,j}}$) by
155 combining gridded manure production data in Zhang et al. (2017) and the grid cell area. To
156 spatialize the national-level manure N application amounts to gridded maps of application rates
157 in each grid area, we multiplied the $R_{a2p_{y,j}}$ in grids where pasture areas were larger than zero
158 with the time-series gridded spatial distribution maps of manure production rate in Zhang et al.

159 (2017) during 1961–2014 and based on the spatial distributions of global pastures in land-use
160 data HYDE 3.2 (Klein Goldewijk et al., 2017).

161 The above-mentioned processes are represented by following equations:

$$162 \quad R_{a2p_{y,j}} = \frac{T_{Mapp_{y,j}}}{\sum_{g=1}^{g=n \text{ in country } j} (R_{Mprod_{y,g}} \times A_g)} \quad (1)$$

163 where year is from 1961 to 2016, and country number is 165. $R_{a2p_{y,j}}$ is the ratio (unitless) of
164 manure application to production in the year y and country j . $T_{Mapp_{y,j}}$ is the national total manure
165 application amount (kg N yr⁻¹) derived from the FAO database for each year. A_g is the area of
166 each grid (km²).

$$167 \quad R_{Mapp_{y,g}} = R_{a2p_{y,j}} \times R_{Mprod_{y,g}} \quad (2)$$

168 where $R_{Mapp_{y,g}}$ is the gridded manure application rate (kg N km⁻² yr⁻¹) in year y and country j .

169 As the national-level manure application amount was not available during 1860–1960, we
170 assumed that $R_{a2p_{y,j}}$ is the same as for 1961. Combining with the gridded spatial maps of
171 manure production rates in Zhang et al. (2017), we generated the datasets of spatialized manure
172 application rates to global pastures during 1860–1960.

173 Finally, we calculated manure application amounts in each country by combining $R_{Mapp_{y,g}}$ and
174 grid areas to compare with national-level deposition amounts from the FAOSTAT database
175 during 1961–2016. As we calculated national-level manure application amounts during
176 1860–1960 using $R_{a2p_{y,j}}$ in 1961, these data served as national total manure N application
177 amounts to adjust $R_{Mapp_{y,g}}$ during 1860–1960.

178 The adjustment procedure is represented in the following equations:

179
$$CT_{Mapp_{y,j}} = \sum_{g=1}^{g=n \text{ in country } j} (R_{Mapp_{y,g}} \times A_g) \quad (3)$$

180 where year is from 1860–2016. $CT_{Mapp_{y,j}}$ (kg N yr⁻¹) is the calculated national-level manure
 181 application amounts in the year y and country j . If $CT_{Mapp_{y,j}}$ is less or more than $T_{Mapp_{y,j}}$, an
 182 adjustment is needed to keep calculated national total amounts consistent with amounts from the
 183 FAOSTAT database. In this case, $CT_{Mapp_{y,j}}$ is less than $T_{Mapp_{y,j}}$ using Eq. 3, thus an adjustment
 184 is needed, using the following equations:

185
$$R_{a_{y,j}} = \frac{T_{Mapp_{y,j}}}{CT_{Mapp_{y,j}}} \quad (4)$$

186 where $R_{a_{y,j}}$ is the regulation ratio (unitless) in the year y and country j .

187
$$R_{Mapp_{y,g(r)}} = R_{Mapp_{y,g}} \times R_{a_{y,j}} \quad (5)$$

188 where $R_{Mapp_{y,g(r)}}$ is real gridded manure application rate (kg N km⁻² yr⁻¹) in the year y and
 189 country j .

190 2.4 Global manure N deposition on pastures and rangelands

191 To develop global distribution maps of manure N deposition [by grazing animals](#), we first
 192 obtained country-level statistics of “manure left on pasture” over the period 1961–2016 from the
 193 FAOSTAT (2018) ‘Livestock manure’ domain of FAOSTAT agri-environmental indicators
 194 (FAO, 2018). We then obtained the national-level ratio of manure deposition to production
 195 ($R_{d2p_{y,j}}$) by combining country-level FAOSTAT datasets of “Manure left on pasture” and
 196 gridded total manure production datasets based on Zhang et al. (2017). Then, we used spatial
 197 distributions of global permanent meadows and pastures, including pastures and rangelands,
 198 based on HYDE 3.2 grassland data (Klein Goldewijk, 2017) and gridded maps of deposition

199 rates, to spatialize the national-level manure N deposition at the global scale. For example, we
 200 multiplied the $R_{d2p_{y,j}}$ ratio in grids within which the pastures and rangelands area was larger
 201 than zero, with the time-series gridded spatial distribution maps of manure production rates in
 202 Zhang et al. (2017) during 1961–2014 (Fig. 2).

203 The above-mentioned processes are represented by the following equations:

$$204 \quad R_{d2p_{y,j}} = \frac{T_{Mdep_{y,j}}}{\sum_{g=1}^{g=n \text{ in country } j} (R_{Mprod_{y,g}} \times A_g)} \quad (6)$$

205 where year (y) is from 1961 to 2016 and country number (j) is 157. $R_{d2p_{y,j}}$ is the ratio (unitless)
 206 of manure deposition to production in the year y and country j . $T_{Mdep_{y,j}}$ is national total manure
 207 deposition amount (kg N yr⁻¹) derived from the FAOSTAT database for each year. $R_{Mprod_{y,g}}$ is
 208 the gridded manure N production rate (kg N km⁻² yr⁻¹) in the year y and grid g .

$$209 \quad R_{Mdep_{y,g}} = R_{d2p_{y,j}} \times R_{Mprod_{y,g}} \quad (7)$$

210 where $R_{Mdep_{y,g}}$ is the gridded manure deposition rate (kg N km⁻² yr⁻¹) in the year y and country j .

211 Finally, we calculated the manure deposition amount for each country through combining
 212 $R_{Mdep_{y,g}}$ and grid area to compare with the national-level deposition amounts from the
 213 FAOSTAT database, using the following equation:

$$214 \quad CT_{Mdep_{y,j}} = \sum_{g=1}^{g=n \text{ in country } j} (R_{Mdep_{y,g}} \times A_g) \quad (8)$$

215 where $CT_{Mdep_{y,j}}$ (kg N yr⁻¹) is the calculated national-level manure deposition amount in the
 216 year y and country j . If $CT_{Mdep_{y,j}}$ is less or more than $T_{Mdep_{y,j}}$, an adjustment was made to keep
 217 calculated national total amounts consistent with those from the FAOSTAT database. In this case,
 218 $CT_{Mdep_{y,j}}$ is roughly equal to $T_{Mdep_{y,j}}$ using Eq. 8, thus no adjustment was needed.

219 Since the national-level manure deposition amounts are not available during 1860–1960, we
220 assumed that $R_{d2py,j}$ is the same as that in 1961. Combining the gridded spatial maps of manure
221 production rates in Zhang et al. (2017), we generated datasets of spatialized manure deposition
222 rates on permanent meadows and pastures globally, for the period 1860–1960.

223 **3 Results**

224 **3.1 Synthetic fertilizer N application to pastures, 1961–2016**

225 The FAO data, combined with the geospatial analysis in this work, show that the total amount of
226 synthetic N fertilizer applied to pastures increased from 0.04 to 8.7 Tg N yr⁻¹ during 1961–2016
227 at an average rate of ~0.18 Tg N yr⁻¹ ($R^2 = 0.98$) per year (Fig. 3a). Synthetic N fertilizer
228 application rates showed rapid increases across the globe, with large spatial variations during the
229 study period (Figs. 4b-c). The global average application rate on pastures was 0.07 kg N ha⁻¹ yr⁻¹
230 in 1961 and reached 10.9 kg N ha⁻¹ yr⁻¹ in 2016 (increased ~154 folds) (Table 1).

231 In the 1960s, Europe (0.2 Tg N yr⁻¹) was the largest contributor (67.8%) to the total global N
232 fertilizer use, followed by North America (0.06 Tg N yr⁻¹, 21.8%) and southern Asia (0.03 Tg N
233 yr⁻¹, 9.9%) (Fig. 5a). The remaining regions accounted for less than 1% of the total N fertilizer
234 application. During 1961–2016, southern Asia showed a continuous increase of N fertilizer
235 consumption and became the largest contributor (3.4 Tg N yr⁻¹, 45%) between 2000 and 2016.
236 In contrast, Europe’s synthetic N fertilizer use and contribution to the global total decreased
237 since the 1980s (Fig. 5a). This is a well-known trend, linked to EU-wide policy directives aimed
238 at minimizing N pollution (Tubiello, 2018). During 2000–2016, Europe applied 2.1 Tg N yr⁻¹,
239 which accounted for 27% of the total global N fertilizer use on pastures. There was a slight
240 increase in the contribution from North America, and the synthetic fertilizer N use amount

241 increased by 1.6 Tg N yr⁻¹. The remaining regions accounted for roughly 7% of the total N
242 fertilizer application on pastures.

243 The average synthetic N application rate in Oceania, North America, and southern Asia showed a
244 rapid increase over the period 1961–2016 (Fig. 5d). Africa and northern Asia showed a slight
245 increase in average N fertilizer application rates during the study period. Europe exhibited a
246 rapid increase of N fertilizer application rates since 1961, then decreased after 2000, and then
247 started to increase in recent five years (Fig. S3).

248 We identified the top five countries (India, United States, China, France, and Germany) with
249 highest fertilizer N application to pastures in 2016. These countries consumed 49% to 58% of the
250 total N fertilizer from 1961 to 2016. India (1.5 Tg N yr⁻¹) and the United States (1.5 Tg N yr⁻¹)
251 were the two largest contributors in 2016, at an increasing rate of 45 Gg N yr⁻¹ (R² = 0.98) per
252 year during 1980–2016 and 32 Gg N yr⁻¹ (R² = 0.99) per year during 1961–2016, respectively.
253 China consumed 1.4 Tg N yr⁻¹ in 2016 at an increasing rate of 34 Gg N yr⁻¹ (R² = 0.96) per year
254 during 1977–2016 while there was only a slight increase during 1961–1976. In contrast,
255 fertilizer N use in France peaked in 1999 (0.8 Tg N yr⁻¹), then showed a rapid decrease until
256 2016 (0.5 Tg N yr⁻¹). Similarly, in Germany, it peaked in 1988 (0.8 Tg N yr⁻¹), and showed a
257 continuous decrease until 2016 (0.3 Tg N yr⁻¹).

258 3.2 Manure N application to pastures, 1860–2016

259 Our results showed that the annual manure N application rates on pastures increased from 1.4 to
260 8.6 Tg N yr⁻¹ during 1860–2016 (Fig. 3a). Manure N application rates showed rapid increases
261 across the globe and exhibited large spatial variations, shifting the regional use from North
262 America and Europe to Asia, during the study period (Figs. 4d-f). The global average manure

263 application rate was 5.3 kg N ha⁻¹ yr⁻¹ in the 1860s and roughly doubled by 2016 (10.7 kg N ha⁻¹
264 yr⁻¹) (Table 1).

265 From the regional perspective (Fig. 5b), in the 1860s Europe (0.8 Tg N yr⁻¹) was the largest
266 contributor and accounted for 53%, while southern Asia (0.25 Tg N yr⁻¹) accounted for 17% of
267 the global total manure N application on pastures. South and North America shared the same
268 proportion (13%), whereas the remaining regions only shared 4%. Conversely during 2000–2016,
269 manure N application on pastures in southern Asia (2.9 Tg N yr⁻¹) was tenfold higher than that in
270 the 1860s and accounted for 36% of the global total, surpassing Europe, which accounted for 28%
271 of the global total. Manure N application amounts in North America and South America
272 increased, but with different magnitudes. During 2000–2016, North America accounted for 11%,
273 while South America accounted for 17% of the global total. In the remaining regions, significant
274 increases of annual manure N application on pastures also occurred, but their contributions to the
275 global total changed only slightly (8%) compared to the 1860s.

276 The regional average manure N application rate was increasing in southern Asia and Africa
277 during 1860–2016 (Fig. S3b). South America, Oceania, and North America exhibited a rapid
278 decreasing trend of manure N application rates from the 1860s to the 1960s and showed
279 continuous increases afterward until 2016 (Figs. 5e, S3b), which was associated with the
280 substantial expansion of pasture areas (Table S2). Europe exhibited a rapid increase of manure N
281 application rates since the 1860s, then decreased after the 1980s (Figs. 5e).

282 In 2016, the top five countries with largest manure N applications on pastures were China,
283 United States, Brazil, Russia, and France. Manure N application in these countries contributed 43%
284 to 52% of global total use from 1961 to 2016. China (2.5 Tg N yr⁻¹) alone accounted for 30% in
285 2016 at an increasing rate of 42 Gg N yr⁻¹ (R² = 0.98) per year during 1961–2016. Manure N use

286 on pastures in Brazil and the United States was roughly the same (0.7 Tg N yr^{-1}) in 2016. Both
287 countries showed a slower increasing trend (Brazil: 7 Gg N yr^{-1} per year and United States: 3 Gg
288 N yr^{-1} per year) during 1961–2016. In contrast, Russian manure N application peaked in 1989
289 (0.7 Tg N yr^{-1}), then showed a rapid decrease until 2016 (0.3 Tg N yr^{-1}). Similarly, in France, it
290 peaked in 1979 ($0.45 \text{ Tg N yr}^{-1}$), then showed a continuous decrease until 2016 ($0.28 \text{ Tg N yr}^{-1}$).

291 3.3 Manure N deposition on pastures and rangelands, 1860–2016

292 Our data show that the total amounts of manure N deposited on pastures and rangelands
293 increased from 14 to 84 Tg N yr^{-1} during 1860–2016 (Fig. 3b). Manure N deposition rates
294 increased steeply across the globe, but exhibited large spatial variations during the study period
295 (Fig. 4g-i). The increase was much larger in the eastern world (typically China and India) and
296 South America compared to the western world. The global average manure deposition rate was
297 $11 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ in 1860 and reached $25 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ in 2016 (Table 1).

298 At the regional scale (Fig. 5c), in the 1860s southern Asia was the region with the largest manure
299 N deposition on pastures and rangelands (4.4 Tg N yr^{-1} ; 30% of total manure N deposition
300 amounts), followed by Africa (2.8 Tg N yr^{-1} ; 19%) and South America (2.4 Tg N yr^{-1} ; 16%).
301 Manure N deposition in the remaining regions was estimated to be 5.1 Tg N yr^{-1} , contributing 35%
302 to the total manure N deposition amount. During 2000–2016, southern Asia, Africa, and South
303 America were still the three largest contributors: 27 Tg N yr^{-1} accounted for 34%, 20 Tg N yr^{-1}
304 accounted for 26%, and 15 Tg N yr^{-1} accounted for 20% of the global manure N deposition on
305 pastures and rangelands, respectively. The remaining regions (Oceania, North America, and
306 Europe) contributed to 20% of the global total during 2000–2016. Europe and Oceania saw an
307 increase in manure N deposition amounts from 1860 to 1960, but since 1980 there was a

308 significant decrease, partly explained by the onset of N pollution regulation. Manure N
309 deposition amounts in North America increased during 1860–1980, but changed slightly since
310 1960.

311 Oceania showed a continuously decreasing trend of average manure N deposition rates in
312 pastures and rangelands over the period 1860–2016. Manure N deposition rates in South
313 America decreased between 1860 and 1960 and then increased afterward until 2016 (Fig. S3c).
314 The significant contrast of changes in manure N deposition rates in Oceania and South America
315 between the 1860s and the 1960s is due to the substantial and rapid increase of grassland areas
316 (Tables S2, S3). Africa and southern Asia saw continuous increases in manure N deposition
317 rates from 1860 to 2016, whereas Europe and North America was found with decreasing
318 deposition rates since the 1980s (Figs. 5f, S3c).

319 In this study, we identified the top 10 countries (China, Brazil, India, Ethiopia PDR, United
320 States, Australia, Sudan (former), Pakistan, Argentina, and Nigeria) that together contributed to
321 48% of the global total manure N deposition on pastures and rangelands in 2016. Among these
322 countries, China (17%) and Brazil (21%) were the two largest contributors, with the similar
323 annual rate of increase of $\sim 125 \text{ Gg N yr}^{-1}$ ($R^2 = 0.99$) per year during 1961–2016. India was the
324 third largest contributor, however, at a small increasing rate of 63 Gg N yr^{-1} ($R^2 = 0.98$) per year
325 during 1961–2016. Annual manure N deposition in Ethiopia PDR was stable during 1961–2000,
326 but since then rapidly increased at a rate of 117 Gg N yr^{-1} ($R^2 = 0.96$) per year. The United States
327 showed a significant increase of annual manure N deposition on pastures and rangelands from
328 1961 to 1975 and then was stable after 1980. Australia showed a decreasing trend during
329 1990–2016 at a rate of 62 Gg N yr^{-1} ($R^2 = 0.92$) per year, whereas, in the former Sudan, Pakistan,
330 and Nigeria annual manure N deposition amounts to pastures and rangelands increased at an

331 annual average rate of 68 ($R^2 = 0.8$), 46 ($R^2 = 0.97$), 56 ($R^2 = 0.98$) Gg N yr⁻¹ per year,
332 respectively. There was no significant change in manure N deposition amounts in Argentina; the
333 annual from 1961 to 2016 was 2.6 Tg N yr⁻¹.

334 **4 Discussion**

335 4.1 Overview of global N inputs to pastures and rangelands

336 The global N cycle has been significantly perturbed by human activity since at least the
337 industrial revolution. Intense agricultural activities, such as synthetic N fertilizer production and
338 use, and intensive livestock production, were identified as major drivers to such change. In this
339 context, improving estimates of global anthropogenic N inputs to pastures and rangelands and
340 their consequences, including on N₂O emissions, is important (Galloway et al., 2008; Tian et al.,
341 2016; Xu et al., 2017). In this study, we generated global datasets of fertilizers N inputs from
342 livestock, both synthetic and from manure, during the period 1860–2016. Pastures and
343 rangelands have experienced substantial land expansion over the period of 1860–1998 (Klein
344 Goldewijk, 2017). The total amount of mineral and manure N applied to permanent meadows
345 and pastures increased by 573% over the study period, from 15 to 101 Tg N yr⁻¹ from 1860 to
346 2016. During 2000–2016, the global mineral N fertilizer application to agriculture was
347 significant, reaching 110 Tg N yr⁻¹ in 2016, while manure N production was 123 Tg N yr⁻¹ (FAO,
348 2018; FAOSTAT, 2018), resulting in a total input of 233 Tg N yr⁻¹. Our estimate of total N
349 inputs (synthetic N fertilizer: 7.5 Tg N yr⁻¹; manure N application: 8.2 Tg N yr⁻¹; manure N
350 deposition: 78.1 Tg N yr⁻¹) to permanent meadows and pastures (93.8 Tg N yr⁻¹) accounted for
351 45% of global total N production (manure: 114.2 Tg N yr⁻¹; synthetic N fertilizer: 96.4 Tg N yr⁻¹)
352 during 2000–2016.

353 4.2 Extension of FAO information

354 Our work extends the relevant FAO national-level statistics in order to provide input drivers for
355 process-based model simulations (e.g., N₂O-MIP, Tian et al., 2018a; Tian et al., 2018b). We
356 furthermore separated N application rates between pastures and cropland, based on previous
357 published work. We likewise extended information available in FAOSTAT by providing
358 spatialized manure N application rates to pastures and spatialized national-level manure N
359 deposition dataset from 1860 to 2016.

360 4.3 Comparison with other studies

361 We compared our datasets with other existing data sources (Table 2). Our estimate of world total
362 manure N use on pastures was 58% and 171% higher than that estimated by Stehfest &
363 Bouwman (2006) and Liu et al. (2010), respectively. However, our estimate was 39% and 87%
364 lower than estimates by Bouwman et al. (2002 and 2013, respectively). Critically, pasture area
365 data varied significantly across different studies. For example, Bowman et al. (2013) divided
366 grasslands into mixed and pastoral systems, and estimated grasslands area based on the country-
367 or regional-level grazing intensity (Table 2). In addition, synthetic fertilizers were applied to the
368 area of mixed agricultural systems (grassland and cropland) and manure N was assumed to be
369 applied to both mixed and pastoral systems. The HYDE 3.2 land use dataset divides the global
370 grazing area into intensively managed grasslands (pastures), and less intensive and unmanaged
371 grasslands (rangelands) (Klein Goldewijk et al., 2017). In this study, we rather assumed that all
372 manure N was applied to pastures, the latter estimated from the HYDE database (798 Mha).
373 Hence, pasture area defined in Bowman et al. (2013) was more than fourfold higher than the data

374 we used. Consequently, the spatial distribution and annual total N application differed
375 substantially compared with that in Bowman et al. (2013).

376 Similarly, the estimates of N fertilizer use in pastures showed large variations across studies
377 (Table 1). This study obtained country-level N fertilizer amounts applied to pastures from the
378 national-level ratios provided by Lassaletta et al. (2014) and total N amounts applied to soils
379 provided by FAOSTAT. Thus, the global N fertilizer amount in 2000 was consistent with that in
380 Lassaletta et al. (2014). Liu et al. (2010) assumed that 16% of fertilizer was applied to global
381 grasslands. Their estimate was roughly twice as high as this study (6.2 Tg N yr^{-1}) for the year
382 2000. The estimates by Bowman et al. (2002) and Stehfest & Bouwman (2006) were 31% and
383 50%, respectively, lower than our estimates in the corresponding years. Klein Goldewijk et al.
384 (2017) divided land used for grazing into more intensively used pastures, less intensively used or
385 unmanaged rangelands. In this study, we assumed N fertilizer was applied to all global pastures
386 and therefore the total area of intensively managed grassland was significantly different from the
387 area used in Bowman et al. (2002) and Chang et al. (2016).

388 4.4 Changes in N inputs hotspots

389 Overall, southern Asia ranks as a top hotspot of all sources of global N inputs in pastures and
390 rangelands during the past three decades, causing a major threat to environmental sustainability
391 and human health in this region. In the 1860's overall manure N production amounts were
392 similar in Asia and Europe (Zhang et al., 2017). However, manure N deposition was 2.4 times
393 higher than that in Europe, whereas manure N application was roughly three times lower than
394 that in Europe. During 2000–2016, southern Asia accounted for ~42% of global manure N
395 production. Consequently, manure N deposition and application amounts in southern Asia were
396 the highest compared to the rest of the regions between 2000–2016. These increases are due to

397 large increases in animal numbers (e.g., cattle, sheep and goats) since 1950 (Bouwman et al.,
398 2013; Dangal et al., 2017). For the rest of the regions, the increases of livestock numbers were
399 also found in South America and Africa since 1860, whereas livestock numbers in Europe and
400 North America showed a decreasing trend after 1980 (Dangal et al., 2017). Thus, besides
401 southern Asia, South America and Africa were hotspots for manure N deposition during
402 1860–2016, while manure N deposition amount decreased in Europe and North America since
403 the 1980s.

404 4.4.1 Shifting hotspots of N fertilizer application

405 European countries (e.g., Germany, United Kingdom, and Ireland) were identified as top
406 hotspots of global N fertilizer application in 1961 (Fig. 4b). However, these hotspots have shifted
407 from Western Europe towards southern Asia at the end of the 20th century (Fig. 4c). Southern
408 Asia was found with the highest N fertilizer application amounts between 2000 and 2016, most
409 concentrated in countries of East and South Asia (e.g., China and India). China and India
410 together applied 36% of global total N fertilizer to pastures and rangelands.

411 4.4.2 Shifting hotspots of manure N application

412 Manure application hotspots moved from European countries to southern Asia during the past
413 155 years. Between 1860 and 1999, Europe accounted for 50% of global total manure N
414 application to pastures and experienced a rapid growth of manure N application, peaked (3.5 Tg
415 N yr⁻¹) in 1986. In 1860, the highest applications were in the United Kingdom, France, and
416 Germany (Fig. 4d), but by 2016, the highest application was in the North China Plain (Fig. 4f).
417 China alone applied 29% of global total manure N during 2000–2016.

418 4.4.3 Shifted hotspots in manure N deposition

419 Southern Asia, as the hotspot of manure N deposition to pastures and rangelands, contributed 31%
420 of the global total amount during the past 157 years. Also, in Africa and South America
421 substantial increases of manure N deposition during 1860–2016 were observed. In the 1860s,
422 manure N deposition from southern Asia, Africa, and South America contributed to 65%,
423 whereas Europe accounted only for 12% of the global total manure N deposition. In 1860, the
424 highest deposition rates were observed for New Zealand, Australia, and Western Europe (Fig.
425 4g). In 2016, except for the above-mentioned regions, the highest deposition rates were in South
426 and West Asia, China, West and East Africa, and South America (Fig. 4i). During 2000–2016,
427 manure N deposition from southern Asia, Africa, and South America contributed to 80%, while
428 Europe accounted for 5% of the global total amount.

429 4.5 Limitations and uncertainties

430 This study attempts to provide an overall estimate of N inputs to global rangelands and pastures,
431 during the period 1860–2016. However, before these data are used in global models,
432 uncertainties of these datasets need to be addressed. First, the different definitions of grassland
433 systems used by the scientific community introduce uncertainties of the spatial patterns and
434 annual total amounts of N inputs. Chang et al. (2016) generated global maps of grassland
435 management intensity since 1901 based on modeled net primary production and the use of grass
436 biomass generated by Herrero et al. (2013). Their total grassland area substantially differed from
437 pasture area developed by HYDE 3.1 (Chang et al., 2016). In this study, we used HYDE 3.2 to
438 generate N inputs to global grasslands, [defined more appropriately by using the FAO land use](#)
439 [definition of ‘permanent meadows and pastures’](#). This dataset exactly followed the FAOSTAT
440 data during 1960–2015, and combined population density data to reconstruct land use prior to
441 1960. Pastures and rangelands defined in HYDE 3.2 were based on the intensity of human

442 management. Although Bouwman et al. (2013) indicated that grassland areas in their study were
443 also calculated based on the grazing intensity, their total area (pastures and rangelands) and
444 spatial patterns were obviously different from HYDE 3.2 (Table 2). Thus, a better understanding
445 of land use is vital to reduce the uncertainty of estimating N input rates and amounts in pastures
446 and rangelands.

447 Second, the FAOSTAT database provides country-level manure N applied to soils; however, this
448 dataset could not be directly applied to study N cycles on pastures since applications to cropland
449 and pasture soils are not differentiated. In this study, it remains large uncertainty that we
450 separated national-level manure N application on pastures simply based on pasture area over
451 total agricultural area (cropland, pastures and rangelands). In previous studies, Bouwman et al.
452 (2013) assumed that 50% and only 5% of the available manure was applied to grasslands in most
453 industrialized countries and in most developing countries, respectively. Liu et al. (2010)
454 allocated 34% of the national total solid manure to pastures in European countries and Canada,
455 13% of the national total manure to pastures in the United States, and 10% of the national total
456 manure to pastures in developing countries. Chang et al. (2016) assumed that manure N
457 application rate changes along with changes in the total ruminant stocking density. Moreover, the
458 spatialization process of N application rates might introduce large uncertainty. The spatial
459 pattern of gridded manure N application rates in our study are correlated with manure production
460 rates in Zhang et al. (2017). The assumptions and uncertainties mentioned in their study, such as
461 without considering livestock migration, might cause uncertainty of spatial distribution.

462 Third, studies used different data sources and made various assumptions of the annual amount of
463 fertilizer N applied on global pastures (Bouwman et al., 2002; Chang et al., 2016; Lassaletta et
464 al., 2014; Liu et al., 2009; Stehfest and Bouwman, 2006). Thus, there remains large uncertainty

465 of total N application on permanent meadows and pastures globally. Moreover, N fertilizer
466 application rates by crops were highly investigated and documented in previous studies. Hence,
467 N fertilizer application datasets were generated considering crop-specific fertilizer rates and
468 cropland area in each grid (Lu and Tian, 2017; Mueller et al., 2012; Nishina et al., 2017; Potter et
469 al., 2010). In reality, N fertilizer application on pastures of each country is not homogeneous. In
470 this study, we assumed that N fertilizer application rate in each country was constant, which
471 means fertilizer was applied evenly in each grid with pastures area larger than zero. Last, inside
472 each relevant land use cell pastures and rangelands may be characterized by different livestock
473 density and deposition rates, which is not considered in our current datasets. The final manure N
474 deposition would be highly affected by the proportion of each type of management in the cell.
475 Thus, it is necessary to consider these in the future research.

476 Furthermore, other human-induced sources of N inputs to pastures and rangelands were not
477 included in our study, which may underestimate total N received globally. For example,
478 biological N fixation was one of the major N sources in the terrestrial ecosystem in the absence
479 of human influence (Cleveland et al., 1999). Pastures and rangelands occupy 25% of the Earth's
480 ice-free land surface across different latitudes with divergent biological N fixation abilities. Plant
481 production in temperate grasslands is proximately limited by N supply due to little N via N
482 fixation; however, tropical savannah received a large amount of N through leguminous species
483 (Cleveland et al., 1999; Vitousek et al., 2013). An estimate of potential N fixation amount by
484 global grassland systems is $\sim 46.5 \text{ Tg N yr}^{-1}$, with a range of $26.6\text{--}66.5 \text{ Tg N yr}^{-1}$ (Cleveland et al.,
485 1999). Atmospheric N deposition is another major source of N input to permanent meadows and
486 pastures globally and increased from 2 to 14 Tg N yr^{-1} for the period 1860–2016 based on the

487 Chemistry–Climate Model Initiative N deposition fields (Eyring et al., 2013; Tian et al., 2018a;
488 Tian et al., 2018b).

489 **5 Data availability**

490 The $0.5^\circ \times 0.5^\circ$ gridded global datasets of manure nitrogen deposition, manure nitrogen
491 application, and nitrogen fertilizer application in grassland systems are available at
492 <https://doi.pangaea.de/10.1594/PANGAEA.892940> (Xu et al., 2018). Data are in ASCII format.
493 A supplemental file is added to the list of all other parameters used in this study to calculate
494 these three datasets in global grassland systems.

495 **6 Conclusion**

496 In the context of increasing livestock production, manure and fertilizer N inputs to permanent
497 meadows and pastures (pastures and rangelands areas) globally have increased rapidly since the
498 industrial revolution. However, datasets of global N inputs are still incomplete. This is the first
499 study that has attempted to consider major sources of anthropogenic N inputs in permanent
500 meadows and pastures and hence generated time-series gridded datasets of manure and fertilizer
501 N application rates, and manure deposition rate during 1860–2016. Our datasets indicated a rapid
502 increase of total N inputs to pastures and rangelands globally during this period, especially the
503 past half century. The hotspots of grassland N application shifted from European countries to
504 southern Asia, specifically China and India during 1860–2016, which indicated the spatial
505 transformation of environmental problems. In this study, we have obtained N data from various
506 sources to fill the data gap; however, large uncertainties still remain in our datasets (e.g., N
507 application rate within each country, annual manure application amounts). More information is
508 needed to improve these datasets in our further work.

509 **Acknowledgements:** This study has been supported by National Key R & D Program of China
510 (Grant Number: 2017YFA0604702, 2018YFA0606001), NOAA Grants (NA16NOS4780207,
511 NA16NOS4780204), National Science Foundation (1210360, 1243232), STS Program of the
512 Chinese Academy of Sciences (KFJ-STZ-ZDTP-010-05), SKLURE Grant (SKLURE2017-1-6).
513 We are grateful to FAO and its member countries for the collection, analysis and dissemination
514 of fertilizers and land use statistics. We thank Dr. Wilfried Winiwarter in International Institute
515 for Applied Systems Analysis for constructive comments that have helped improve this study.

516 **References**

- 517
- 518 Bai, Z., Lee, M. R., Ma, L., Ledgard, S., Oenema, O., Velthof, G. L., Ma, W., Guo, M., Zhao, Z., and Wei,
519 S.: Global environmental costs of China's thirst for milk, *Global Change Biology*, 24, 2198-2211,
520 2018.
- 521 Bouwman, A., Boumans, L., and Batjes, N.: Estimation of global NH₃ volatilization loss from synthetic
522 fertilizers and animal manure applied to arable lands and grasslands, *Global Biogeochemical Cycles*,
523 16, 2002.
- 524 Bouwman, A., Beusen, A. H., and Billen, G.: Human alteration of the global nitrogen and phosphorus soil
525 balances for the period 1970–2050, *Global Biogeochemical Cycles*, 23, 2009.
526 <https://doi.org/10.1029/2009GB003576>
- 527 Bouwman, A., Van Drecht, G., Knoop, J., Beusen, A., and Meinardi, C.: Exploring changes in river
528 nitrogen export to the world's oceans, *Global Biogeochemical Cycles*, 19, 2005.
529 <https://doi.org/10.1029/2004GB002314>
- 530 Bouwman, L., Goldewijk, K. K., Van Der Hoek, K. W., Beusen, A. H., Van Vuuren, D. P., Willems, J.,
531 Rufino, M. C., and Stehfest, E.: Exploring global changes in nitrogen and phosphorus cycles in
532 agriculture induced by livestock production over the 1900–2050 period, *Proceedings of the National*
533 *Academy of Sciences*, 110, 20882-20887, 2013.
- 534 Breymer, A.: Managed grasslands and ecological experience, *Ecosystems of the world*, 17, 335-350,
535 1990.
- 536 Chang, J., Ciais, P., Herrero, M., Havlik, P., Campioli, M., Zhang, X., Bai, Y., Viovy, N., Joiner, J.,
537 Wang, X., Peng, S., Yue, C., Piao, S., Wang, T., Hauglustaine, D. A., Soussana, J.-F., Peregón, A.,
538 Kosykh, N., and Mironycheva-Tokareva, N.: Combining livestock production information in a
539 process-based vegetation model to reconstruct the history of grassland management, *Biogeosciences*,
540 13, 3757-3776, 2016.
- 541 Cleveland, C. C., Townsend, A. R., Schimel, D. S., Fisher, H., Howarth, R. W., Hedin, L. O., Perakis, S.
542 S., Latty, E. F., Von Fischer, J. C., and Elseroad, A.: Global patterns of terrestrial biological nitrogen
543 (N₂) fixation in natural ecosystems, *Global Biogeochemical Cycles*, 13, 623-645, 1999.
- 544 Dangal, S. R., Tian, H., Zhang, B., Pan, S., Lu, C., and Yang, J.: Methane emission from global livestock
545 sector during 1890–2014: Magnitude, trends and spatiotemporal patterns, *Global Change Biology*, 23,
546 4147-4161, 2017.
- 547 Davidson, E. A.: The contribution of manure and fertilizer nitrogen to atmospheric nitrous oxide since
548 1860, *Nature Geoscience*, 2, 659-662, 2009.
- 549 Davis, K. F., Yu, K., Herrero, M., Havlik, P., Carr, J. A., and D'Odorico, P.: Historical trade-offs of
550 livestock's environmental impacts, *Environmental Research Letters*, 10, 125013, 2015.
551 DOI:10.1088/1748-9326/10/12/125013
- 552 Eyring, V., Lamarque, J.-F., Hess, P., Arfeuille, F., Bowman, K., Chipperfield, M. P., Duncan, B., Fiore,
553 A., Gettelman, A., and Giorgetta, M. A.: Overview of IGAC/SPARC Chemistry-Climate Model
554 Initiative (CCMI) community simulations in support of upcoming ozone and climate assessments,
555 *Sparc Newsletter*, 40, 48-66, 2013.
- 556 FAO, 2018. Nitrogen inputs to agricultural soils from livestock manure: New statistics. FAO Rome, Italy.
557 FAOSTAT (Food and Agriculture Organization Corporate Statistical Database): FAO online database,
558 available at: <http://www.fao.org/faostat/en/#data> (last access: July 2017), 2016. Land Use domain:
559 <http://www.fao.org/faostat/en/#data/RL>; fertilizers by nutrient:
560 <http://www.fao.org/faostat/en/#data/RFN>; livestock manure:
561 <http://www.fao.org/faostat/en/#data/EMN>.
- 562 Fowler, D., Coyle, M., Skiba, U., Sutton, M. A., Cape, J. N., Reis, S., Sheppard, L. J., Jenkins, A.,
563 Grizzetti, B., and Galloway, J. N.: The global nitrogen cycle in the twenty-first century, *Philosophical*
564 *Transactions of the Royal Society B: Biological Sciences*, 368, 2013. DOI: 10.1098/rstb.2013.0164

565 Galloway, J. N., Townsend, A. R., Erismann, J. W., Bekunda, M., Cai, Z., Freney, J. R., Martinelli, L. A.,
566 Seitzinger, S. P., and Sutton, M. A.: Transformation of the nitrogen cycle: recent trends, questions,
567 and potential solutions, *Science*, 320, 889-892, 2008.

568 Gerber, J. S., Carlson, K. M., Makowski, D., Mueller, N. D., Garcia de Cortazar-Atauri, I., Havlík, P.,
569 Herrero, M., Launay, M., O'Connell, C. S., and Smith, P.: Spatially explicit estimates of N₂O
570 emissions from croplands suggest climate mitigation opportunities from improved fertilizer
571 management, *Global change biology*, 22, 3383-3394, 2016.

572 Havlík, P., Valin, H., Herrero, M., Obersteiner, M., Schmid, E., Rufino, M. C., Mosnier, A., Thornton, P.
573 K., Böttcher, H., and Conant, R. T.: Climate change mitigation through livestock system transitions,
574 *Proceedings of the National Academy of Sciences*, 111, 3709-3714, 2014.

575 Herrero, M., Havlík, P., Valin, H., Notenbaert, A., Rufino, M. C., Thornton, P. K., Blümmel, M., Weiss,
576 F., Grace, D., and Obersteiner, M.: Biomass use, production, feed efficiencies, and greenhouse gas
577 emissions from global livestock systems, *Proceedings of the National Academy of Sciences*, 110,
578 20888-20893, 2013.

579 Holland, E., Lee-Taylor, J., Nevison, C., and Sulzman, J.: Global N Cycle: Fluxes and N₂O Mixing Ratios
580 Originating from Human Activity. Data Set (Oak Ridge National Laboratory Distributed Active
581 Archive Center, Oak Ridge, TN, 2005).

582 Klein Goldewijk, K., Beusen, A., Doelman, J., and Stehfest, E.: Anthropogenic land use estimates for the
583 Holocene–HYDE 3.2, *Earth System Science Data*, 9, 927-953, 2017.

584 Lassaletta, L., Billen, G., Grizzetti, B., Anglade, J., and Garnier, J.: 50 year trends in nitrogen use
585 efficiency of world cropping systems: the relationship between yield and nitrogen input to cropland,
586 *Environmental Research Letters*, 9, 105011, 2014. DOI:10.1088/1748-9326/9/10/105011

587 Liu, J., You, L., Amini, M., Obersteiner, M., Herrero, M., Zehnder, A. J., and Yang, H.: A high-resolution
588 assessment on global nitrogen flows in cropland, *Proceedings of the National Academy of Sciences*,
589 107, 8035-8040, 2010.

590 Lu, C. and Tian, H.: Global nitrogen and phosphorus fertilizer use for agriculture production in the past
591 half century: shifted hot spots and nutrient imbalance, *Earth System Science Data*, 9, 181-192, 2017.

592 Mueller, N. D., Gerber, J. S., Johnston, M., Ray, D. K., Ramankutty, N., and Foley, J. A.: Closing yield
593 gaps through nutrient and water management, *Nature*, 490, 254-257, 2012.

594 Nishina, K., Ito, A., Hanasaki, N., and Hayashi, S.: Reconstruction of spatially detailed global map of
595 NH₄⁺ and NO₃⁻ application in synthetic nitrogen fertilizer, *Earth System Science Data*, 9, 149-162,
596 2017.

597 Oenema, O., Wrage, N., Velthof, G. L., van Groenigen, J. W., Dolfing, J., and Kuikman, P. J.: Trends in
598 global nitrous oxide emissions from animal production systems, *Nutrient cycling in agroecosystems*,
599 72, 51-65, 2005.

600 Potter, P., Ramankutty, N., Bennett, E. M., and Donner, S. D.: Characterizing the spatial patterns of
601 global fertilizer application and manure production, *Earth Interactions*, 14, 1-22, 2010.

602 Sattari, S., Bouwman, A., Rodríguez, R. M., Beusen, A., and Van Ittersum, M.: Negative global
603 phosphorus budgets challenge sustainable intensification of grasslands, *Nature communications*, 7,
604 10696, 2016. DOI: 10.1038/ncomms10696

605 Sheldrick, W. F., Syers, J. K., and Lingard, J.: A conceptual model for conducting nutrient audits at
606 national, regional, and global scales, *Nutrient Cycling in Agroecosystems*, 62, 61-72, 2002.

607 Smith P., M. Bustamante, H. Ahammad, H. Clark, H. Dong, E.A. Elsiddig, H. Haberl, R. Harper, J.
608 House, M. Jafari, O. Masera, C. Mbow, N.H. Ravindranath, C.W. Rice, C. Robledo Abad, A.
609 Romanovskaya, F. Sperling, and F. Tubiello, 2014: Agriculture, Forestry and Other Land Use
610 (AFOLU). In: *Climate Change 2014: Mitigation of Climate Change. Contribution of Working Group
611 III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change* [Edenhofer, O.,
612 R. Pichs-Madruga, Y. Sokona, E. Farahani, S. Kadner, K. Seyboth, A. Adler, I. Baum, S. Brunner, P.
613 Eickemeier, B. Kriemann, J. Savolainen, S. Schlömer, C. von Stechow, T. Zwickel and J.C. Minx
614 (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

615 Stehfest, E. and Bouwman, L.: N₂O and NO emission from agricultural fields and soils under natural
616 vegetation: summarizing available measurement data and modeling of global annual emissions,
617 *Nutrient Cycling in Agroecosystems*, 74, 207-228, 2006.

618 Tian, H., Lu, C., Ciais, P., Michalak, A. M., Canadell, J. G., Saikawa, E., Huntzinger, D. N., Gurney, K.
619 R., Sitch, S., and Zhang, B., Yang, J., Bousquet, P., Bruhwiler, L., Chen, G., Dlugokencky, E.,
620 Friedlingstein, P., Melillo, J., Pan, S., Poulter, B., Prinn, R., Saunio, M., Schwalm, C. R., and Wofsy,
621 S. C.: The terrestrial biosphere as a net source of greenhouse gases to the atmosphere, *Nature*, 531,
622 225–228, 2016.

623 Tian, H., Yang, J., Lu, C., Xu, R., Canadell, J. G., Jackson, R., Arneeth, A., Chang, J., Chen, G., and Ciais,
624 P.: The global N₂O Model Intercomparison Project (NMIP), *Bulletin of the American Meteorological*
625 *Society*, 2018. <https://doi.org/10.1175/BAMS-D-17-0212.1>

626 Tian, H., Yang, J., Xu, R., Lu, C., Canadell, J. G., Davidson, E. A., Jackson, R. B., Arneeth, A., Chang, J.,
627 and Ciais, P.: Global soil nitrous oxide emissions since the pre-industrial era estimated by an
628 ensemble of Terrestrial Biosphere Models: Magnitude, attribution and uncertainty, *Global Change*
629 *Biology*, 2018. <https://doi.org/10.1111/gcb.14514>

630 Tubiello, F.N., 2018. GHG emissions due to agriculture. Encyclopedia of Food Systems, reference
631 module in food science, Elsevier. DOI: 10.1016/B978-0-08-100596-5.21996-3

632 Tubiello, F. N., Salvatore, M., Rossi, S., Ferrara, A., Fitton, N., and Smith, P.: The FAOSTAT database
633 of greenhouse gas emissions from agriculture, *Environmental Research Letters*, 8, 015009, 2013.
634 DOI:10.1088/1748-9326/8/1/015009

635 Valin, H., Havlik, P., Mosnier, A., Herrero, M., Schmid, E., and Obersteiner, M.: Agricultural
636 productivity and greenhouse gas emissions: trade-offs or synergies between mitigation and food
637 security?, *Environmental Research Letters*, 8, 035019, 2013. DOI:10.1088/1748-9326/8/3/035019

638 Vitousek, P. M., Menge, D. N., Reed, S. C., and Cleveland, C. C.: Biological nitrogen fixation: rates,
639 patterns and ecological controls in terrestrial ecosystems, *Philosophical Transactions of the Royal*
640 *Society of London B: Biological Sciences*, 368, 20130119, 2013. DOI: 10.1098/rstb.2013.0119

641 White, R. P., Murray, S., Rohweder, M., Prince, S., and Thompson, K.: *Grassland ecosystems*, World
642 Resources Institute Washington, DC, 2000.

643 Xu, R., Tian, H., Lu, C., Pan, S., Chen, J., Yang, J., and Bowen, Z.: Preindustrial nitrous oxide emissions
644 from the land biosphere estimated by using a global biogeochemistry model, *Climate of the Past*, 13,
645 977-990, 2017.

646 Xu, R., Pan, S., Chen, J., Chen, G., Yang, J., Dangal, S., Shepard, J., and Tian, H.: Half-Century
647 Ammonia Emissions From Agricultural Systems in Southern Asia: Magnitude, Spatiotemporal
648 Patterns, and Implications for Human Health, *GeoHealth*, 2, 40-53, 2018.

649 Yang, Q., Tian, H., Li, X., Ren, W., Zhang, B., Zhang, X., Wolf J.: Spatiotemporal patterns of livestock
650 manure nutrient production in the conterminous United States from 1930 to 2012, *Science of the*
651 *Total Environment*, 541, 1592-1602, 2016.

652 Zhang, B., Tian, H., Lu, C., Dangal, S. R., Yang, J., and Pan, S.: Global manure nitrogen production and
653 application in cropland during 1860–2014: a 5 arcmin gridded global dataset for Earth system
654 modeling, *Earth System Science Data*, 9, 667-678, 2017.

655

656 **Table 1.** The N input rates, applied/deposited area, and total amounts in global pastures and rangelands in
 657 1860, 1961, 2000, and 2016 (1 km² = 100 ha).

	1860	1961	1980	2000	2016
Averaged N fertilizer application rate (kg N ha⁻¹ yr⁻¹)	NA	0.07	3.6	7.8	10.9
Total applied area (Mha)	NA	623.8	725	797.8	803.1
Total amounts (Tg N yr⁻¹)	NA	0.04	2.6	6.2	8.7
Average manure N application rate (kg N ha⁻¹ yr⁻¹)	5.3	8.1	9.8	9.5	10.7
Total applied area (Mha)	268.2	623.8	725	797.8	803.1
Total amounts (Tg N yr⁻¹)	1.4	5.0	7.1	7.6	8.6
Average manure N deposition rate (kg N ha⁻¹ yr⁻¹)	11.2	15.4	19.0	20.7	25.3
Total deposited area (Mha)	1250.1	3070.7	3194.2	3398.5	3295
Total amounts (Tg N yr⁻¹)	14.0	47.2	60.7	70.5	83.5

658

659 **Table 2.** Comparison of manure and fertilizer N application amounts between this study and published
 660 datasets.

	Bouwman et al., (2002)^a	Stehfest & Bouwman, 2006^β	Bouwman et al., 2013^γ	Chang et al., 2016^α	Liu et al., 2010^γ	Lassaletta et al., 2014^γ	This study^γ
Manure N application (Tg N yr⁻¹)	12.4	4.8	57.8	12.4	~2.8	NA	7.6
Applied area (Mha)	625	NA	3358 ^η	1231	NA	NA	798
N fertilizer application (Tg N yr⁻¹)	4.3	3.1	NA	3.1	12.9	6.5	6.2
Applied area (Mha)	103	NA	NA	39	NA	NA	798

661 ^α estimated in 1995.

662 ^β national-level fertilizer data for 1998. The total grassland area for N fertilizer and manure was 677 Mha.

663 ^γ estimated in 2000.

664 ^η the grassland area includes both mixed and patrol systems.

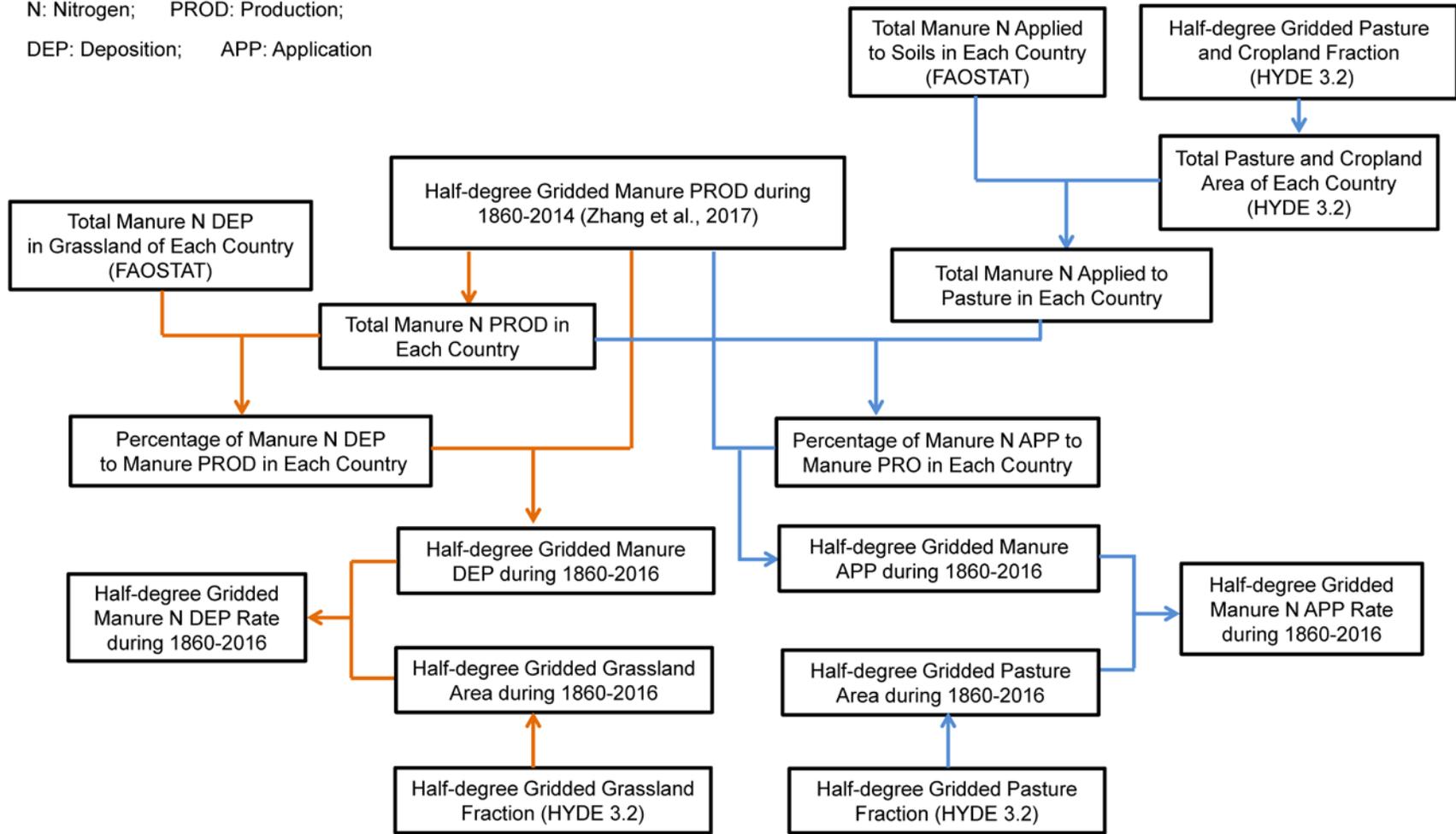
665

666

667 **Figure 1.** Diagram of the workflow for developing the database of global annual N fertilizer use
668 rate in pasture during the period 1961–2016.

669

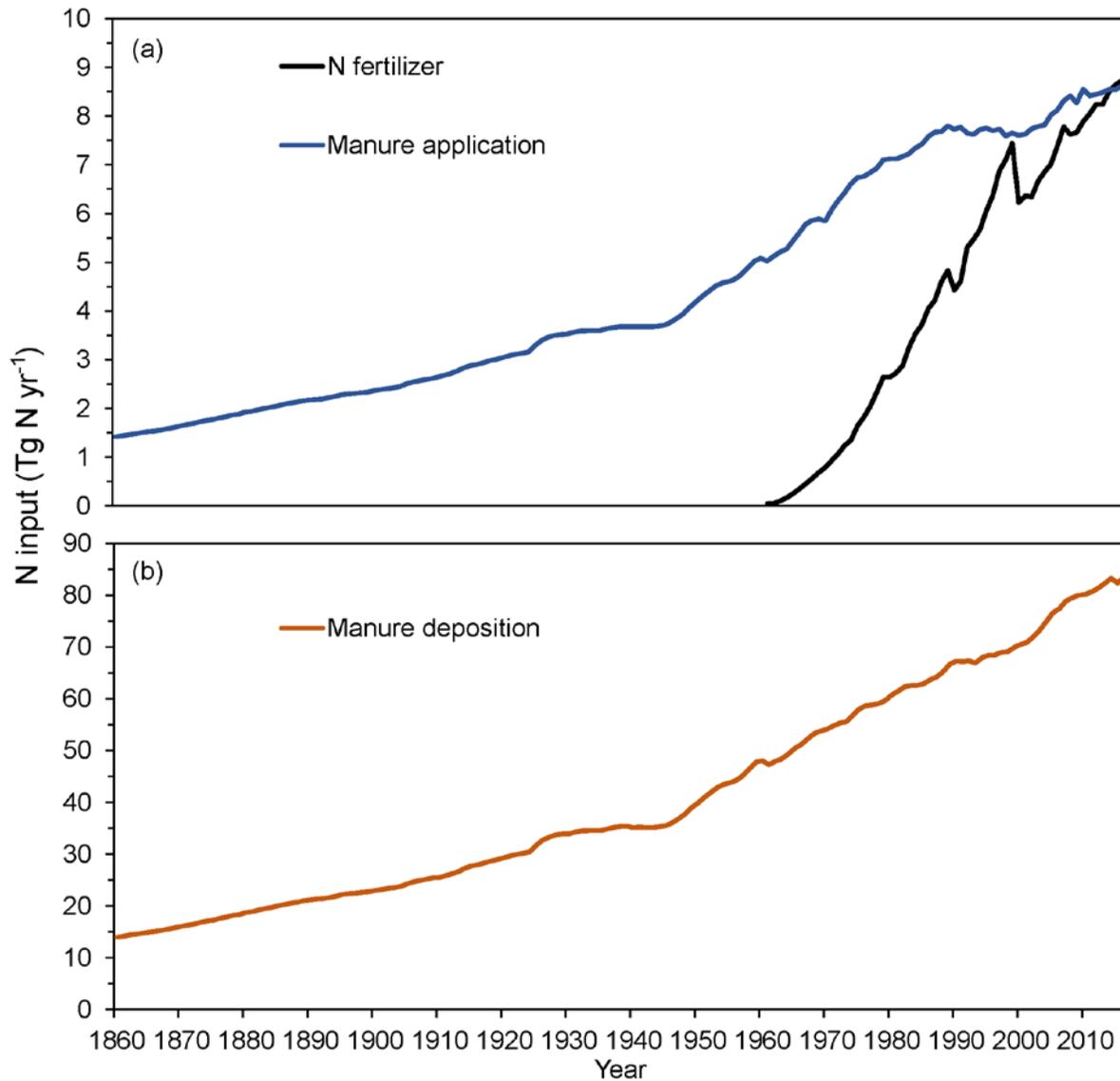
N: Nitrogen; PROD: Production;
 DEP: Deposition; APP: Application



670

671 **Figure 2.** Diagram of the workflow for developing the database of global annual manure N use rate in pastures and manure N
 672 deposition rate in pastures and rangelands during the period 1860–2016.

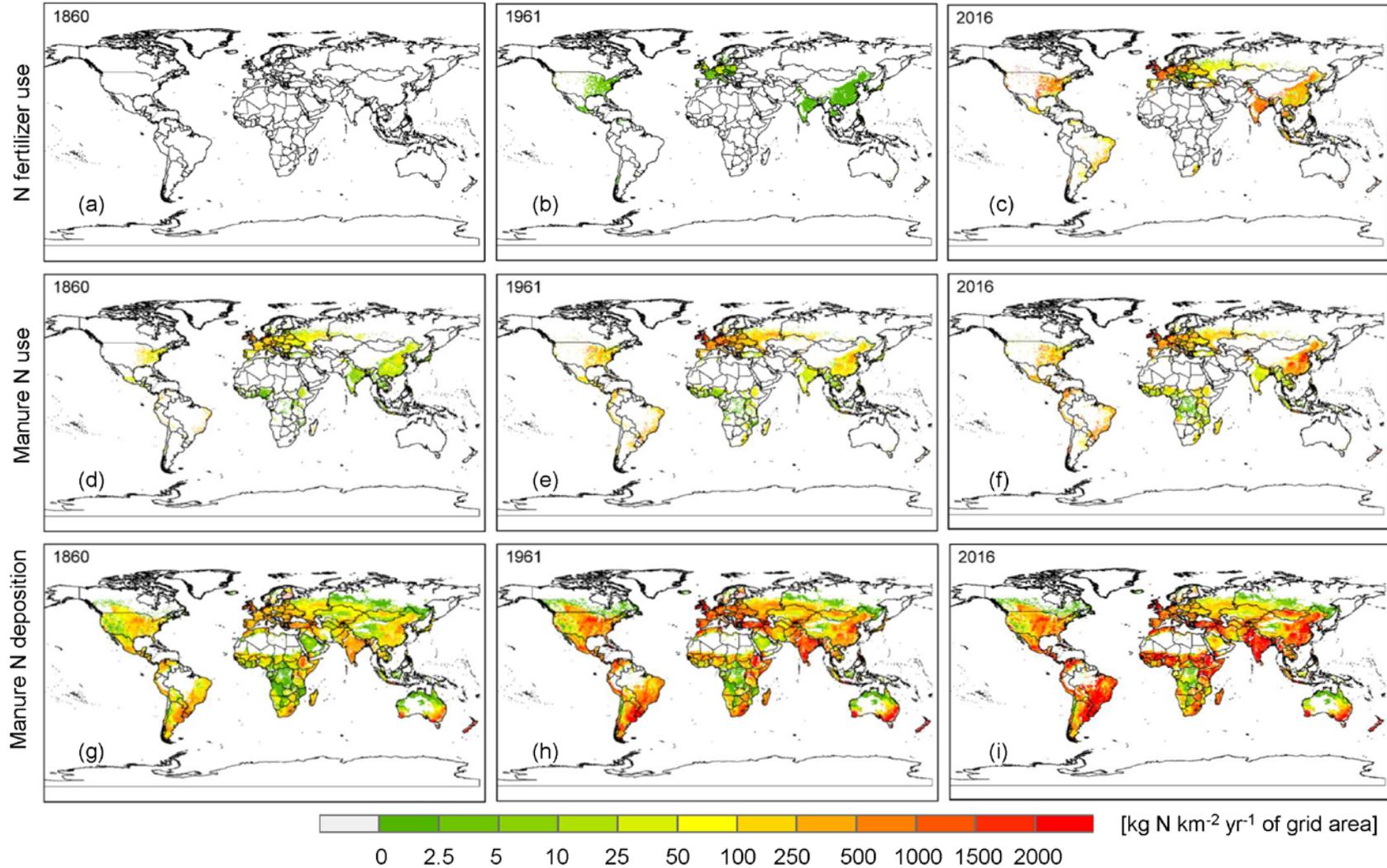
673



674

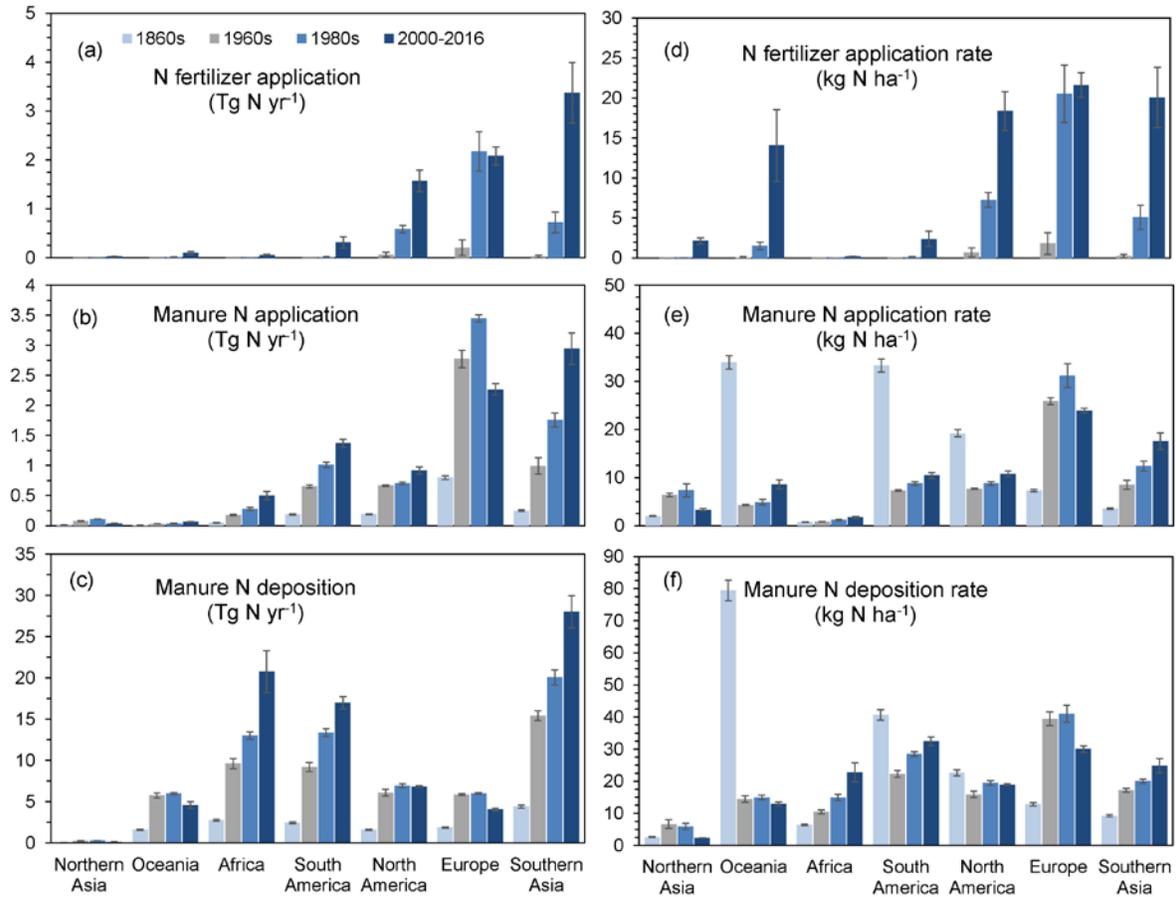
675 **Figure 3.** Temporal patterns of global manure N use, N fertilizer use, and manure deposition in
 676 grassland systems: (a) Manure N use and N fertilizer use on global pastures during 1860–2016
 677 and during 1961–2016, respectively; (b) Manure N deposition to global pastures and rangelands
 678 during 1860–2016.

679



680

681 **Figure 4.** Spatial patterns of N input rates in global pastures and rangelands in 1860, 1961, and 2016: (a)–(c) N fertilizer application
 682 rates; (d)–(f) manure N application rates; (g)–(i) manure N deposition rates.



683

684 **Figure 5.** Nitrogen fertilizer use (a) and rate (d), manure N use (b) and rate (e), and manure N
 685 deposition (c) and rate (f) at regional scales in 1860s, 1960s, 1980s, and 2000–2016. Error bars
 686 represent standard deviation within each decade.

687

688

689

690