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Increased nitrogen enrichment and shifted patterns in the world's grassland: 1860-2014

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Abstract

As the largest sector on the Earth's land surface, the livestock system is not only a major contributor to global manure excreta that perturbs the global nutrient balance, but also a major emitter of greenhouse gases that warms the climate. Much attention has been paid to nitrogen (N) fertilizer and manure N applications to global croplands, however, there is still a lack of spatially-explicit estimates of continuous time-series datasets of manure and fertilizer N inputs in global grasslands. In this study, we therefore developed three global gridded datasets at a resolution of 0.5° × 0.5° for the period 1860–2014 (i.e., annual manure N deposition rate, synthetic N fertilizer use rate, and manure N application rate) by combining annual and 5-arc minute spatial data on pasture and rangeland with country-level manure and synthetic fertilizer N data from the Food and Agricultural Organization database (FAOSTAT). We found that total N inputs, sum of manure N deposition, manure and fertilizer N application to global grassland systems increased from 15.5 to 103.8 Tg N yr^{-1} during 1860–2014. Manure N deposition accounted for 83.7% of the total N inputs, whereas manure and fertilizer N application accounted for 7.7% and 8.6%, respectively, during 2000–2014. At the regional scale, hotspots of manure N deposition remained the same during 1860–2014 (i.e., southern Asia, Africa, and South America), but hotspots of manure and fertilizer N application have shifted from Europe to southern Asia in the early 21st century. These three datasets could fill data gaps of N inputs in global and regional grasslands and serve as input drivers for ecosystem and biogeochemistry models to investigate the impacts of N enrichment on the global grassland system, greenhouse gas emissions, and environmental sustainability. Datasets available at https://doi.pangaea.de/10.1594/PANGAEA.892940.

Keywords: nitrogen, manure, synthetic nitrogen fertilizer, global grassland systems

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1 Introduction

Livestock production has increased substantially in response to growing meat consumption across the globe in the past century (Bouwman et al., 2013). The livestock sector, which predominantly covers grassland systems, occupies 30% of Earth's ice-free land surface and accounts for 33%–50% of the world's agricultural gross domestic products (Herrero et al., 2013). While the livestock sector is a major source of income for more than 1.3 billion people globally, it is also a major user of crop products and freshwater resources (Dangal et al., 2017; Herrero et al., 2013). Thus, the expansion of this sector plays a significant role in land-use changes and global nutrient cycles (Havlík et al., 2014; Herrero et al., 2013). There is a growing recognition that the livestock sector has caused increasing greenhouse gases (GHGs) and ammonia emissions, severe pollution of aquatic systems, and degradation of soil properties at the site-, regional-, and global-scale (Dangal et al., 2017; Davidson, 2009; Davis et al., 2015; Oenema et al., 2005; Tian et al., 2016; Valin et al., 2013). The growing demand for livestock products has strengthened the reliance on grains for feed and has led to an increase in manure and fertilizer N use in croplands and consequently increased GHG emissions and other air-polluting gases (e.g., ammonia, nitric oxide) (Bouwman et al., 2013; Davis et al., 2015; Xu et al., 2018).

Livestock systems play an important role in global nutrient cycles. For example, nitrogen production from animal manure has increased from 21.4 Tg N yr^{-1} in 1860 to 131.0 Tg N yr^{-1} in 2014 (FAOSTAT, 2016; Zhang et al., 2017). Livestock contribute 80% to all non-CO_2 GHG emissions from the agricultural sector, among which CH_4 emissions account for 47%–54% (Dangal et al., 2017; Tubiello et al., 2013). In addition, manure deposited through grazing and manure applied to croplands and pasture account for ~45% of total anthropogenic N_2O emissions (Davidson, 2009). Globally emissions from manure N applied to soils or left on pastures has increased from 0.44 to 0.88 $GtCO_2eq\ yr^{-1}$ during 1961–2010 (Tubiello et al., 2013), and our desire for meat plays an important part in the atmospheric increase of N_2O and CH_4 (Bouwman et al., 2013; Dangal et al., 2017).

A number of studies have focused on developing regional and global datasets of nitrogen fertilizer and livestock manure production to understand their feedback on the climate system. The datasets of N fertilizer use were provided and annually updated by previous studies (Bouwman et al., 2005; FAOSTAT, 2016; Lu and Tian, 2017; Mueller et al., 2012; Nishina et al., 2017; Potter et al., 2010; Sheldrick et al., 2002). Further, global manure production datasets were developed in different studies to achieve various research goals (Bouwman et al., 2009; Bouwman et al., 2013; Holland et al.; Potter et al., 2010; Zhang et al., 2017). In addition, although datasets of manure application in croplands are becoming increasingly available, there is considerable uncertainty in the estimation of total manure application and their spatial distribution across different studies (Gerber et al., 2016; Herrero et al., 2013; Liu et al., 2010; Zhang et al., 2017).

Although previous studies have provided spatially explicit datasets of N inputs in the form of mineral and manure N in cropland systems, the spatially explicit datasets on N inputs in grassland systems are still missing (Hauglustaine, 2016; Lassaletta et al., 2014; Stehfest and Bouwman, 2006). The FAOSTAT database provides country-level manure and fertilizer N amounts on grasslands; however, directly using such datasets by process-based global biogeochemistry models may not fully investigate spatial pattern changes within countries. To enhance our understanding of the role of grassland systems on the overall global GHG balance, global biogeochemistry models require spatially explicit estimates of N inputs. In this study, according to grassland N inputs datasets provided by FAOSTAT (2016), we developed datasets for major sources of N inputs in grassland systems (manure and fertilizer application on pastures, and manure deposition on grasslands).

Through combining the land-use dataset HYDE 3.2 (Klein Goldewijk, 2017) and country-level manure and fertilizer database (FAOSTAT, 2016), we developed three gridded time-series datasets at a resolution of 0.5° × 0.5° for (1) manure N application to pastures (1860–2014), (2) N fertilizer application to pastures (1961–2014), and (3) manure deposition by grazing livestock to rangelands and pastures (1860–2014). We quantified regional variations in N inputs, identified hotspots of N inputs from different N sources in grassland systems, and discussed their uncertainty. These datasets are developed for global model simulation studies in model inter-comparison projects (e.g., NMIP; Tian et al., 2018), and will be updated annually based on the newest released country-level database in FAOSTAT and land-use datasets.

2 Methods

2.1 The categories of grasslands

Globally, grassland systems are defined as areas dominated by herbaceous and shrub vegetation, which includes savannas (Africa, South America and India), steppes (Eurasia), prairies (North America), shrub-dominated areas (Africa), meadows and pastures (United Kingdom and Ireland) and tundra (Breymeyer, 1990; White et al., 2000). In this study, we follow the definition of the FAO categories, and grasslands are pastures and rangelands. In general, pastures are managed to support high stocking densities of grass production for hay/silage, whereas rangelands are unmanaged and grazed at low stocking densities. The spatial distribution map of pastures and rangelands used were provided by the land-use dataset HYDE 3.2 (Klein Goldewijk, 2017). To investigate N inputs in regional grasslands, the global landmass was disaggregated into seven regions: North America, South America, Africa, Europe, southern Asia (i.e., west, south, east, central and southeast Asia), northern Asia, and Oceania (Fig. S1).

2.2 Fertilizer N application on global pastures

We obtained national-level datasets of "synthetic N fertilizer consumption" from FAOSTAT database during 1961–2014. However, this dataset included N fertilizer application amounts in both croplands and pastures. To separate N fertilizer application on pastures from croplands, we adopted the ratio of N use on pastures to total N amount in 151 countries from Lassaletta et al. (2014) even though the ratio in some countries is zero. Then, combining the land-use data (pastures) from HYDE 3.2, we spatialized the national-level N fertilizer application amounts to gridded maps of application rates in each grid cell area during 1961–2014 (Fig. 1). The application rate was the same within each country but different across countries because ratios of N use on pastures were available only at the country level.

2.3 Manure N deposition to global grasslands

To develop global distribution maps of manure deposition (aka manure left on pastures and rangelands), we first obtained country-level datasets of "manure left on pastures" from the FAOSTAT database during 1961–2014. We then obtained the national-level ratio of manure deposition to production ($R_{d2p_{y,j}}$) by combining country-level FAO datasets of "manure left on pastures" and gridded total manure production datasets based on Zhang et al. (2017). Then, we used spatial distributions of global grasslands including rangelands and pastures based on HYDE 3.2 (Klein Goldewijk, 2017) and gridded maps of deposition rates, to spatialize the national-level manure N deposition at the global scale. For example, we multiplied the $R_{d2p_{y,j}}$ ratio in grids within which the grassland (pastures and rangelands) area was larger than zero, with the time-

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series gridded spatial distribution maps of manure production rates in Zhang et al. (2017) during 1961–2014 (Fig. 2).

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

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

where year (y) is from 1961 to 2014 and country number (j) is 158. $R_{d2p_{y,j}}$ is the ratio (unitless) of manure deposition to production in the year y and country j. $T_{Mdep_{y,j}}$ is national total manure deposition amount (kg N yr^{-1}) derived from the FAO database for each year. $R_{Mprod_{y,g}}$ is the gridded manure N production rate (kg N km^{-2} yr^{-1}) in the year y and grid g. A_g is the area of each grid (km^2).

$$R_{Mdep_{y,g}} = R_{d2p_{y,j}} \times R_{Mprod_{y,g}} \quad (2)$$

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

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

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

where $CT_{Mdep_{y,j}}$ (kg N yr^{-1}) is the calculated national-level manure deposition amount in the 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 calculated national total amounts consistent with those from the FAOSTAT database. In this case, $CT_{Mdep_{y,j}}$ is roughly equal to $T_{Mdep_{y,j}}$ using Eq. 3, thus no adjustment was needed.

Since the national-level manure deposition amounts are not available during 1860–1960, we assumed that $R_{d2p_{y,j}}$ is the same as that in 1961. Combining the gridded spatial maps of manure production rates in Zhang et al. (2017), we generated datasets of spatialized manure deposition rates on global grasslands during 1860–1960.

2.4 Manure N application to global pastures

We obtained country-level datasets of "manure applied to soils" from the FAOSTAT database during 1961–2014. However, the FAOSTAT database did not provide the percentage of annual total manure application amount to pastures and data of manure N application rates to pastures are currently not available. We therefore assumed that manure N application rates in pastures and croplands were the same. Through combining land-use data HYDE 3.2, we calculated the total cropland and pasture areas within each country where manure application amount was larger than zero. We then extracted national-level manure N application amounts to pastures during 1961–2014 (Fig. 2).

We calculated the national-level ratio of manure application to production ($R_{a2p_{y,j}}$) by combining gridded manure production data in Zhang et al. (2017) and the grid cell area. To spatialize the national-level manure N application amounts to gridded maps of application rates in each grid area, we multiplied the $R_{a2p_{y,j}}$ in grids where pasture areas were larger than zero with the time-series gridded spatial distribution maps of manure production rate in Zhang et al. (2017) during 1961–2014 and based on the spatial distributions of global pastures in land-use data HYDE 3.2 (Klein Goldewijk et al., 2017).

The above-mentioned processes are represented by following equations:

$$R_{a2p_{y,j}} = \frac{T_{Mapp_{y,j}}}{\sum_{g=1}^{g=n\ in\ country\ j}(R_{Mprod_{y,g}} \times A_g)} \tag{4}$$

where year is from 1961 to 2014, and country number is 202. $R_{a2p_{y,j}}$ is the ratio (unitless) of manure application to production in the year *y* and country *j*. $T_{Mapp_{y,j}}$ is the national total manure application amount (kg N yr^{-1}) derived from the FAO database for each year.

$$R_{Mapp_{y,g}} = R_{a2p_{y,j}} \times R_{Mprod_{y,g}} \tag{5}$$

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

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

Finally, we calculated manure application amounts in each country by combining $R_{Mapp_{y,g}}$ and grid areas to compare with national-level deposition amounts from the FAOSTAT database during 1961–2014. As we calculated national-level manure application amounts during 1860–1960 using $R_{a2p_{y,j}}$ in 1961, these data served as national total manure N application amounts to adjust $R_{Mapp_{y,g}}$ during 1860–1960.

The adjustment procedure is represented in the following equations:

$$CT_{Mapp_{y,j}} = \sum_{g=1}^{g=n\ in\ country\ j}(R_{Mapp_{y,g}} \times A_g) \tag{6}$$

where year is from 1860–2014. $CT_{Mapp_{y,j}}$ (kg N yr^{-1}) is the calculated national-level manure application amounts in the year *y* and country *j*. If $CT_{Mapp_{y,j}}$ is less or more than $T_{Mapp_{y,j}}$, an adjustment is needed to keep calculated national total amounts consistent with amounts from the FAOSTAT database. In this case, $CT_{Mdep_{y,j}}$ is less than $T_{Mdep_{y,j}}$ using Eq. 6, thus an adjustment is needed, using the following equations:

$$R_{a_{y,j}} = \frac{T_{Mapp_{y,j}}}{CT_{Mapp_{y,j}}} \quad (7)$$

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

$$R_{Mapp_{y,g\,(r)}} = R_{Mapp_{y,g}} \times R_{a_{y,j}} \quad (8)$$

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

3 Results

3.1 Fertilizer N application to pastures during 1961–2014

Our results showed that the total amount of N fertilizer applied to pastures increased from 0.05 to 8.50 Tg N yr^{-1} during 1961–2014 at an increasing rate of ~0.18 Tg N yr^{-1} (R^2 = 0.98) per year (Fig. 3a). Synthetic N fertilizer application rates showed rapid increases across the globe and exhibited large spatial variations during the study period (Fig. 4b-c). The global average application rate was 0.07 kg N ha^{-1} yr^{-1} in 1961 and reached 10.67 kg N ha^{-1} yr^{-1} in 2014 (increased ~151 folds) (Table 1).

In the 1960s, Europe (0.2 Tg N yr^{-1}) was the largest contributor (68.4%) to the total global N fertilizer use, followed by North America (0.06 Tg N yr^{-1}, 21.3%) and southern Asia (0.03 Tg N yr^{-1}, 10%) (Fig. 5a). The remaining regions accounted for less than 5% of the total N fertilizer application. During 1961–2014, southern Asia showed a continuous increase of N fertilizer consumption and became the largest contributor (3.3 Tg N yr^{-1}, 45%) between 2000 and 2014. In contrast, Europe's N fertilizer use and contribution to the global total had decreased since the 1980s (Fig. 5a). During 2000–2014, Europe applied 2 Tg N yr^{-1} which accounted for 28% of the total global N fertilizer use. There was a slight increase in the contribution from North America, and the fertilizer N use amount increased by 1.5 Tg N yr^{-1}. The remaining regions accounted for roughly 6% of the total N fertilizer application.

We identified the top 5 countries (India, United States, China, France, and Germany) with highest fertilizer N consumption in 2014. These countries consumed 48.7% to 61.5% of the total N fertilizer from 1961 to 2014. India (1.6 Tg N yr^{-1}) and the United States (1.5 Tg N yr^{-1}) were the two largest contributors in 2014, at an increasing rate of 46.7 Gg N yr^{-1} ($R^2 = 0.98$) per year during 1980–2014 and 31.1 Gg N yr^{-1} ($R^2 = 0.99$) per year during 1961–2014, respectively. China consumed 1.2 Tg N yr^{-1} in 2014 at an increasing rate of 32.4 Gg N yr^{-1} ($R^2 = 0.96$) per year during 1977–2014 while there was only a slight increase during 1961–1976. In contrast, fertilizer N use in France peaked in 1999 (0.76 Tg N yr^{-1}), then showed a rapid decrease until 2014 (0.52 Tg N yr^{-1}). Similarly, in Germany, it peaked in 1987 (0.76 Tg N yr^{-1}), and showed a continuous decrease until 2014 (0.36 Tg N yr^{-1}).

3.2 Manure N application during 1860–2014

Our results showed that the annual manure N use on global pastures increased from 1.4 to 8.5 Tg N yr^{-1} during 1860–2014 (Fig. 3a). Manure N application rates showed rapid increases across the globe and exhibited large spatial variations, shifting from the western to eastern world, during the study period (Fig. 4d-f). The global average manure application rate was 5.3 kg N ha^{-1} yr^{-1} in 1860 and roughly doubled by 2014 (10.72 kg N ha^{-1} yr^{-1}) (Table 1).

From the regional perspective (Fig. 5b), Europe (0.79 Tg N yr^{-1}) acted as the largest contributor and accounted for 51.6%, while southern Asia (0.26 Tg N yr^{-1}) accounted for 17.1% of the global total manure N application in the 1860s. South and North America shared the same proportion (12.6%), whereas the remaining regions only shared 5.5%. During 2000–2014, manure N application in southern Asia (3.0 Tg N yr^{-1}) was tenfold higher than that in the 1860s and accounted for 36.3% of global total manure N application. In contrast, Europe was surpassed by southern Asia and accounted for 27.6% of the global total manure N use. Manure N application amounts in North America and South America increased, but with different magnitudes. During 2000–2014, North America accounted for 11.2%, while South America accounted for 17.0% of the global total. In the remaining regions significant increases of annual manure N application amounts also occurred, but their contributions to global total manure N use changed only slightly (7.9%) compared to the 1860s.

In 2014, the top 5 countries with largest manure N application amounts were China, United States, Brazil, Russia, and France. Manure N application in these countries contributed 43.7% to 53.7% of global total use amounts from 1961 to 2014. China (2.6 Tg N yr^{-1}) alone accounted for 30.8% in 2014 at an increasing rate of 0.04 Tg N yr^{-1} ($R^2 = 0.98$) per year during 1961–2014. Manure N use in Brazil and the United States was 0.73 and 0.63 Tg N yr^{-1}, respectively, in 2014. Both countries showed a slower increasing trend (Brazil: 7.5 and United States: 2.8 Gg N yr^{-1} per year) during 1961–2014. In contrast, Russian manure N application peaked in 1990 (0.7 Tg N yr^{-1}), then showed a rapid decrease until 2014 (0.31 Tg N yr^{-1}). Similarly, in France, it peaked in 1978 (0.45 Tg N yr^{-1}), then showed a continuous decrease until 2014 (0.28 Tg N yr^{-1}).

3.3 Manure N deposition during 1860–2014

Our results showed that the total amounts increased from 14.1 to 86.7 Tg N yr^{-1} during 1860–2014 (Fig. 3b). Manure N deposition rates increased steeply across the globe, but exhibited large spatial variations during the study period (Fig. 4g-i). The increase was much larger in the eastern world (typically China and India) and South America compared to the western world. The global average manure deposition rate was 11.27 kg N ha^{-1} yr^{-1} in 1860 and reached 26.3 kg N ha^{-1} yr^{-1} in 2014 (Table 1).

At the regional scale (Fig. 5c), southern Asia was the region with largest manure N deposition (4.4 Tg N yr^{-1}; 29.8% of total manure N deposition amounts), followed by Africa (2.8 Tg N yr^{-1}; 18.6%) and South America (2.4 Tg N yr^{-1}; 16.5%) in the 1860s. Manure N deposition in the remaining regions was estimated to be 5.2 Tg N yr^{-1}, contributing 35.1% to the total manure N deposition amount. During 2000–2014, southern Asia, Africa, and South America were still the three largest contributors: 27.6 Tg N yr^{-1} accounted for 34.5%, 21.2 Tg N yr^{-1} accounted for 26.4%, and 15.6 Tg N yr^{-1} accounted for 19.6% of the global manure N deposition, respectively. The remaining regions (Oceania, North America, and Europe) contributed to 19.5% of the global total during 2000–2014. Europe and Oceania experienced an increase in manure deposition amounts from 1860 to 1960, but since 1980 there was a significant decrease. Manure N deposition amounts in North America increased during 1860–1980, but changed slightly since 1960.

In this study, we identified the top 10 countries (China, Brazil, India, Ethiopia PDR, United States, Australia, Sudan (former), Pakistan, Argentina, and Nigeria) that together contributed to 52% of the global total manure N deposition to grasslands in 2014. Among these countries, China (20%) and Brazil (20%) were the two largest contributors, with the same annual rate of increase of 0.1 Tg N yr^{-1} (R^2 = 0.99) per year during 1961–2014. India was the third largest contributor, however, at a smaller increasing rate of 0.06 Tg N yr^{-1} (R^2 = 0.98) per year during 1961–2014. Annual manure N deposition in Ethiopia PDR was stable during 1961–2000, but since then rapidly increased at a rate of 0.1 Tg N yr^{-1} (R^2 = 0.96) per year. The United States showed a significant increase of annual manure N deposition from 1961 to 1975 and then was stable after 1980. Australia showed a decreasing trend during 1990–2014 at a rate of 0.06 Tg N yr^{-1} (R^2 = 0.91) per year, whereas, in the former Sudan, Pakistan, and Nigeria annual manure N deposition amounts to grasslands increased at an annual average rate of 0.07 (R^2 = 0.8), 0.05 (R^2 = 0.97), 0.06 (R^2 = 0.98) Tg N yr^{-1} per year, respectively. There was no significant change in manure N deposition amounts in Argentina; the annual from 1961 to 2014 was 2.58 Tg N yr^{-1}.

4 Discussion

4.1 Overview of global N inputs to grasslands

The global N cycle has been altered by human perturbation since the industrial revolution. Intense agricultural activities, such as land use change, N fertilizer input, and intensive livestock rearing play an important part in these changes. However, little attention has been paid to estimate global N inputs to grassland systems and consequences on N_2O emissions (Galloway et al., 2008; Tian et al., 2016; Xu et al., 2017). In this study, we have generated datasets of N input to grassland systems during 1860–2014. Pastures and rangelands have experienced substantial land expansion over the period of 1860–1998, thereafter this increase has slowed down (Klein Goldewijk, 2017). The total amount of mineral and manure N applied to grasslands had increased by 570%, from 15.5 to 103.8 Tg N yr^{-1} from 1860 to 2014. During 2000–2014, the global mineral N fertilizer application in agricultural systems was estimated at 95 Tg N yr^{-1} (FAOSTAT, 2016), and manure production was 122 Tg N yr^{-1} (Zhang et al., 2017), resulting in a total reactive N production of 217 Tg N yr^{-1}. Our estimate of total N inputs to grasslands (96 Tg N yr^{-1}) accounted for 44% of global total N applied to agricultural systems during 2000–2014.

4.2 Improvements beyond FAOSTAT datasets

The original national-level synthetic N fertilizer and manure application/deposition datasets were downloaded from the FAOSTAT database. Our work took a step further in order to provide input drivers for process-based model simulations (e.g., N_2O-MIP, Tian et al., 2018). FAOSTAT mainly provides data of synthetic N fertilizer application amounts for agricultural use. In our dataset we have separated rates of N application to pastures from other agricultural activities for each country for the period 1961 to 2014, based on the percentage provided by Lassaletta et al. (2014). FAOSTAT only provides a total amount of national-level manure N application to soils starting with the year 1961. Our study extended this dataset by providing spatialized manure N application rates to pastures and spatialized national-level manure N deposition dataset from 1860 to 2014.

4.3 Comparison with other studies

We compared our datasets with other existing data sources (Table 2). Our estimate of manure N use on global pastures was 58% and 171% higher than that estimated by Stehfest & Bouwman (2006) and Liu et al. (2010), respectively. However, our estimate was 39% and 87% lower than estimates by Bouwman et al. (2002 and 2013, respectively). Pasture areas varied significantly across different studies. For example, Bowman et al. (2013) divided grasslands into mixed and pastoral systems and their grassland area was calculated based on the country- or regional-level grazing intensity (Table 2). In addition, synthetic fertilizers were applied to the area of mixed agricultural systems (grassland and cropland) and manure N was assumed to be applied to both

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mixed and pastoral systems. The HYDE 3.2 land use dataset divides the global grazing area into intensively managed (pastures), less intensive and unmanaged grasslands (rangelands) (Klein Goldewijk et al., 2017). In this study, we assumed that all manure N was applied to pastures (798 Mha). Hence, pasture area defined in Bowman et al. (2013) was more than fourfold higher than the data we used. Consequently, the spatial distribution and annual total N application differed substantially compared with that in Bowman et al. (2013).

Similarly, the estimates of N fertilizer use in grassland systems showed large variations across studies (Table 1). In this study, we obtained country-level N fertilizer amounts applied to grasslands from the national-level ratios provided by Lassaletta et al. (2014) and total N amounts applied to soils including grasslands and croplands provided by FAOSTAT. Thus, the global N fertilizer amount in 2000 was consistent with that in Lassaletta et al. (2014). Liu et al. (2010) assumed that 16% of fertilizer was applied to global grasslands. Their estimate was roughly twice as high as this study (6.3 Tg N yr^{-1}) for the year 2000. The estimates by Bowman et al. (2002) and Stehfest & Bouwman (2006) were 29% and 55%, respectively, lower than our estimates in the corresponding years. Klein Goldewijk et al. (2017) divided land used for grazing into more intensively used pastures, less intensively used or unmanaged rangelands. In this study, we assumed N fertilizer was applied to all global pastures and therefore the total area of intensively managed grassland was significantly different from the area used in Bowman et al. (2002) and Chang et al. (2016).

4.4 Changes in N inputs hotspots

Overall, southern Asia ranks as a top hotspot of all sources of global N inputs in grassland systems during the past three decades, causing a major threat to environmental sustainability and human health in this region. In the 1860's overall manure N production amounts were similar in

Asia and Europe (Zhang et al., 2017). However, manure N deposition was 2.4 times higher than that in Europe, whereas manure N application was three times lower than that in Europe. During 2000–2014, southern Asia accounted for ~42% of global manure N production. Consequently, manure N deposition and application amounts in southern Asia were the highest compared to the rest of the regions between 2000–2014. These increases are due to large increases in animal numbers (e.g., cattle, sheep and goats) since 1950 (Bouwman et al., 2013; Dangal et al., 2017). For the rest of the regions, the increases of livestock numbers were also found in South America and Africa since 1860, whereas livestock numbers in Europe and North America showed a decreasing trend after 1980 (Dangal et al., 2017). Thus, besides southern Asia, South America and Africa were hotspots for manure N deposition during 1860–2014, while manure N deposition amount decreased in Europe and North America since the 1980s.

4.4.1 Shifting hotspots of N fertilizer application

European countries (e.g., Germany, United Kingdom, and Ireland) were identified as top hotspots of global N fertilizer application in 1961 (Fig. 4b). However, these hotspots have shifted from Western Europe towards southern Asia at the end of the 20th century (Fig. 4c). Southern Asia was found with the highest N fertilizer application amounts between 2000 and 2014, most concentrated in countries of East and South Asia (e.g., China and India). China and India together applied 32.5% of global total N fertilizer to grasslands.

4.4.2 Shifting hotspots of manure N application

Manure application hotspots moved from European countries to southern Asia during the past 155 years. Between 1860 and 1999, Europe accounted for 50% of global total manure N application to grasslands and experienced a rapid growth of manure N application, peaked (3.5

Tg N yr^{-1}) in 1986. In 1860, the highest applications were in the United Kingdom, France, and Germany (Fig. 4d), but by 2014, the highest application was in the North China Plain (Fig. 4f). China alone applied 30% of global total manure N during 2000–2014.

4.4.3 Shifted hotspots in manure N deposition

Southern Asia, as the hotspot of manure N deposition to grasslands, contributed 31% of the global total amount during the past 155 years. Also, in Africa and South America substantial increases of manure N deposition during 1860–2014 were observed. In the 1860s, manure N deposition from southern Asia, Africa, and South America contributed to 65%, whereas Europe accounted only for 13% of the global total manure N deposition. In 1860, the highest deposition rates were observed for New Zealand, Australia, and Western Europe (Fig. 4g). In 2014, except for the above-mentioned regions, the highest deposition rates were in South and West Asia, China, West and East Africa, and South America (Fig. 4i). During 2000–2014, manure N deposition from southern Asia, Africa, and South America contributed to 80%, while Europe accounted for 5% of the global total amount.

4.5 Uncertainties

This study attempts to provide an overall estimate of N inputs to global rangelands and pastures, during the period 1860–2014. However, before these data are used in global models, uncertainties of these datasets need to be addressed. First, the different definitions of grassland systems used by the scientific community introduce uncertainties of the spatial patterns and annual total amounts of N inputs. Chang et al. (2016) generated global maps of grassland management intensity since 1901 based on modeled net primary production and the use of grass biomass generated by Herrero et al. (2013). Their total grassland area substantially differed from pasture area developed by HYDE 3.1 (Chang et al., 2016). In this study, we used HYDE 3.2 to generate N inputs to global grasslands. This dataset exactly followed the FAOSTAT data during 1960–2015, and combined population density data to reconstruct land use prior to 1960. Pastures and rangelands defined in HYDE 3.2 were based on the intensity of human management. Although Bouwman et al. (2013) indicated that grassland areas in their study were also calculated based on the grazing intensity, their total area (pastures and rangelands) and spatial patterns were obviously different from HYDE 3.2 (Table 2). Thus, a better understanding of land use is vital to reduce the uncertainty of estimating N input rates and amounts in grasslands. Second, the FAOSTAT database provides the country-level manure N applied to soils; however, this dataset could not be directly applied to study N cycles on pastures since all N were applied to both cropland and pasture soils. In this study, it remains large uncertainty that we separated national-level manure N application on pastures simply based on pasture area over total agricultural area (pastures and croplands). In previous studies, Bouwman et al. (2013) assumed that 50% and only 5% of the available manure was applied to grasslands in most industrialized countries and in most developing countries, respectively. Liu et al. (2010) allocated 34% of the national total solid manure to pastures in European countries and Canada, 13% of the national total manure to pastures in the United States, and 10% of the national total manure to pastures in developing countries. Chang et al. (2016) assumed that manure N application rate changes along with changes in the total ruminant stocking density. Moreover, the spatialization process of N application rates might introduce large uncertainty. The spatial pattern of gridded manure N application rates in our study are correlated with manure production rates in Zhang et al. (2017). The assumptions and uncertainties mentioned in their study, such as without considering livestock migration, might cause uncertainty of spatial distribution. Third, studies used different data sources and made various assumptions of the annual amount of fertilizer N applied on global pastures (Bouwman et al., 2002; Stehfest and Bouwman, 2006; Chang et al., 2016; Liu et al., 2009; Lassaletta et al., 2014). Thus, there remains large uncertainty of total N application on global grasslands. Moreover, N fertilizer application rates by crops were highly investigated and documented in previous studies. Hence, N fertilizer application datasets were generated considering crop-specific fertilizer rates and cropland area in each grid (Potter et al., 2010; Mueller et al., 2012; Nishina et al., 2017; Lu and Tian, 2017). In reality, N fertilizer application on pastures of each country is not even. In this study, we assumed that N fertilizer application rate in each country was constant, which means fertilizer was applied evenly in each grid with pastures area larger than zero.

Furthermore, other human-induced sources of N inputs to global grasslands were not included in our study, which may underestimate total N received by global grassland systems. For example, biological N fixation was one of the major N sources in the terrestrial ecosystem in the absence of human influence (Cleveland et al., 1999). Grassland system occupies 30% of the Earth's ice-free land surface across different latitudes with divergent biological N fixation abilities. Plant production in temperate grasslands is proximately limited by N supply due to little N via N fixation; however, tropical savannah received a large amount of N through leguminous species (Cleveland et al., 1999; Vitousek et al., 2013). An estimate of potential N fixation amount by global grassland systems is ~46.5 Tg N yr^{-1}, with a range of 26.6–66.5 Tg N yr^{-1} (Cleveland et al., 1999). Atmospheric N deposition is another major source of N input to global grassland system and increased from 2.0 to 13.7 Tg N yr^{-1} for the period 1860–2014 based on the Chemistry–Climate Model Initiative N deposition fields (Eyring et al., 2013; Tian et al., 2018).

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5 Data availability

The 0.5° × 0.5° gridded global datasets of manure nitrogen deposition, manure nitrogen application, and nitrogen fertilizer application in grassland systems are available at https://doi.pangaea.de/10.1594/PANGAEA.892940 (Xu et al., 2018). Data are in ASCII format. A supplemental file is added to the list of all other parameters used in this study to calculate these three datasets in global grassland systems.

6 Conclusion

In the context of increasing livestock production, manure and fertilizer N inputs to the global grassland system have increased rapidly since the industrial revolution. However, datasets of global N inputs in grasslands are still missing. This is the first study that has attempted to consider major sources of anthropogenic N inputs in grassland systems and hence generated time-series gridded datasets of manure and fertilizer N application rates, and manure deposition rate during 1860–2014. Our datasets indicated a rapid increase of total N inputs to global grasslands during this period, especially the past half century. The hotspots of grassland N application shifted from European countries to southern Asia, specifically China and India during 1860–2014, which indicated the spatial transformation of environmental problems. In this study, we have obtained N data from various sources to fill the data gap; however, large uncertainties still remain in our datasets (e.g., N application rate within each country, annual manure application amounts). More information is needed to improve these datasets in our further work.

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Table 1. The N input rates, applied/deposited area, and total amounts in global grasslands (pastures and rangelands) in 1860, 1961, 2000, and 2014 (1 km^2 = 100 ha).

	1860	1961	1980	2000	2014
Averaged N fertilizer application rate (kg N ha^{-1} yr^{-1})	NA	0.07	3.68	7.79	10.67
Total applied area (Mha)	NA	623.83	724.98	797.80	797.36
Total amounts (Tg N yr^{-1})	NA	0.05	2.67	6.21	8.50
Average manure N application rate (kg N ha^{-1} yr^{-1})	5.30	8.06	9.83	9.53	10.72
Total applied area (Mha)	268.21	623.83	725.0	797.80	797.36
Total amounts (Tg N yr^{-1})	1.42	5.03	7.13	7.61	8.55
Average manure N deposition rate (kg N ha^{-1} yr^{-1})	11.27	16.09	19.76	21.45	26.3
Total deposited area (Mha)	1250.13	3070.65	3194.23	3398.45	3300.05
Total amounts (Tg N yr^{-1})	14.09	49.40	63.13	72.90	86.78

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

	Bouwman et al., (2002)[α]	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	6.3
Applied area (Mha)	103	NA	NA	39	NA	NA	798

[α] estimated in 1995.

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

[γ] estimated in 2000.

[η] the grassland area includes both mixed and patrol systems.

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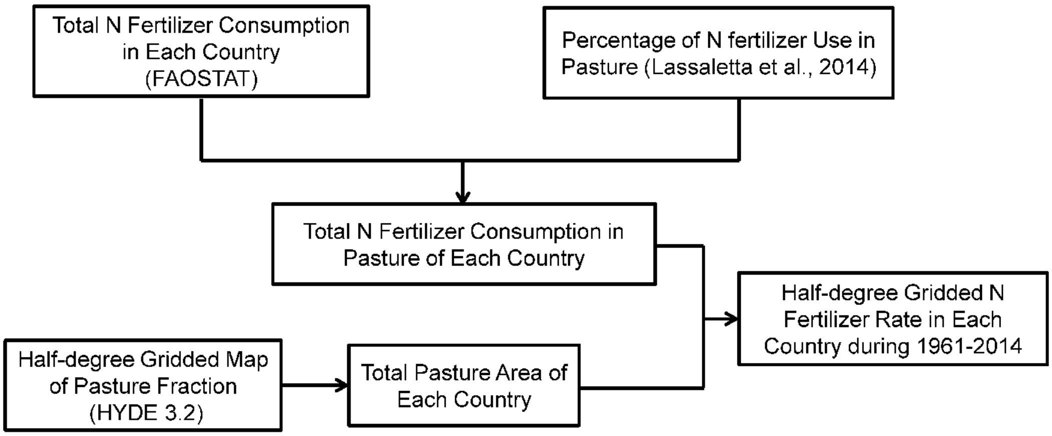

Figure 1. Diagram of the workflow for developing the database of global annual N fertilizer use rate in pasture during the period 1961–2014.

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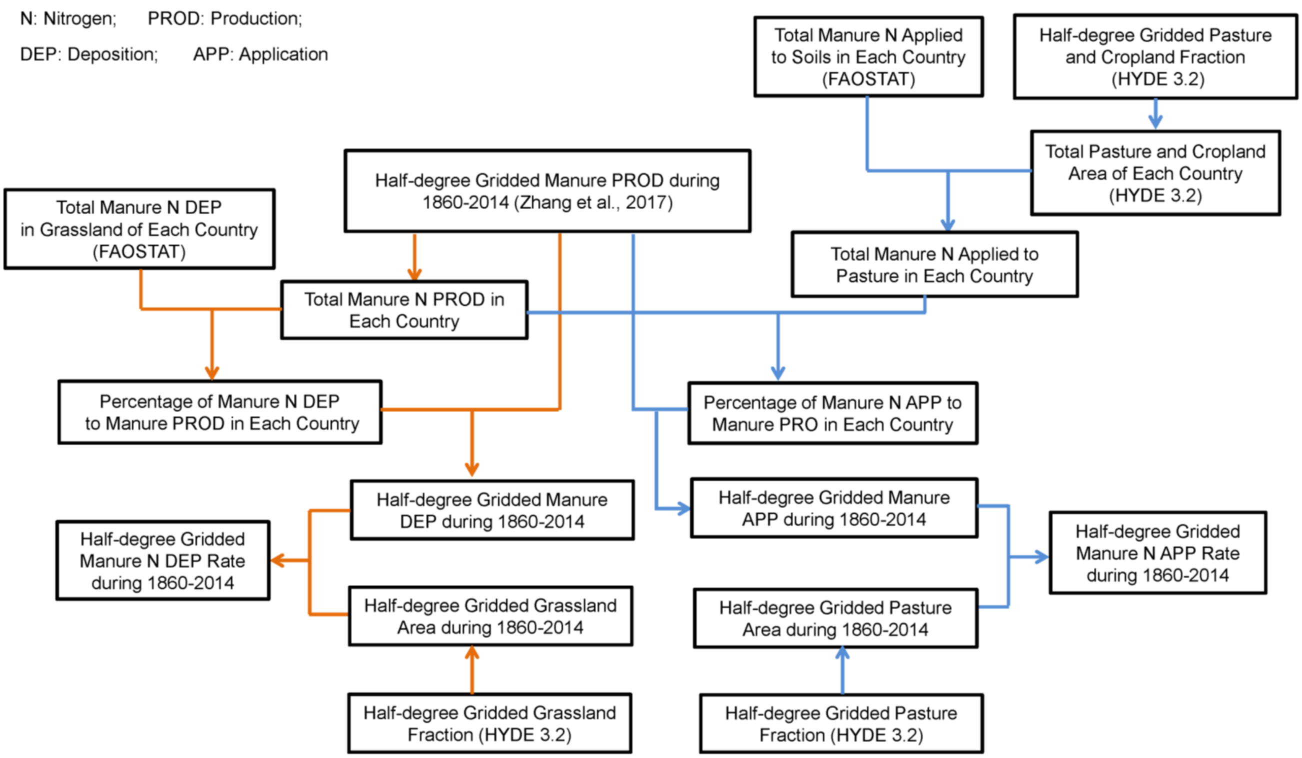

Figure 2. Diagram of the workflow for developing the database of global annual manure N use rate in pasture and manure N deposition rate in grassland during the period 1860–2014.

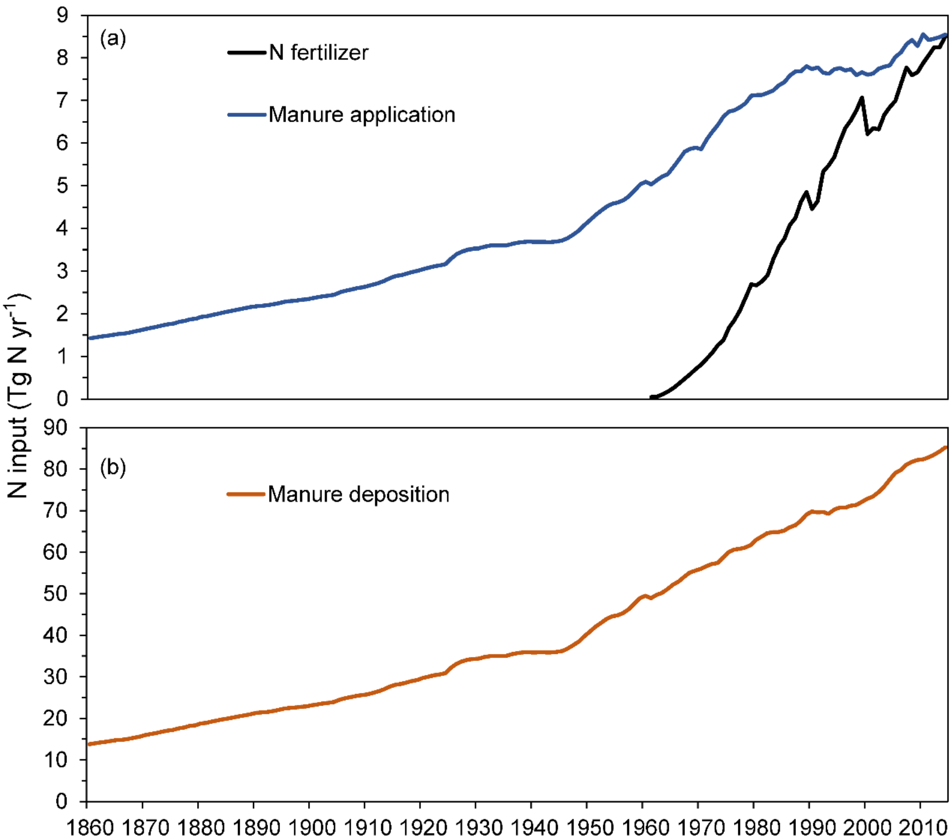

Figure 3. Temporal patterns of global manure N use, N fertilizer use, and manure deposition in grassland systems: (a) Manure N use and N fertilizer use on global pastures during 1860–2014 and during 1961–2014, respectively; (b) Manure N deposition to global grasslands during 1860–2014.

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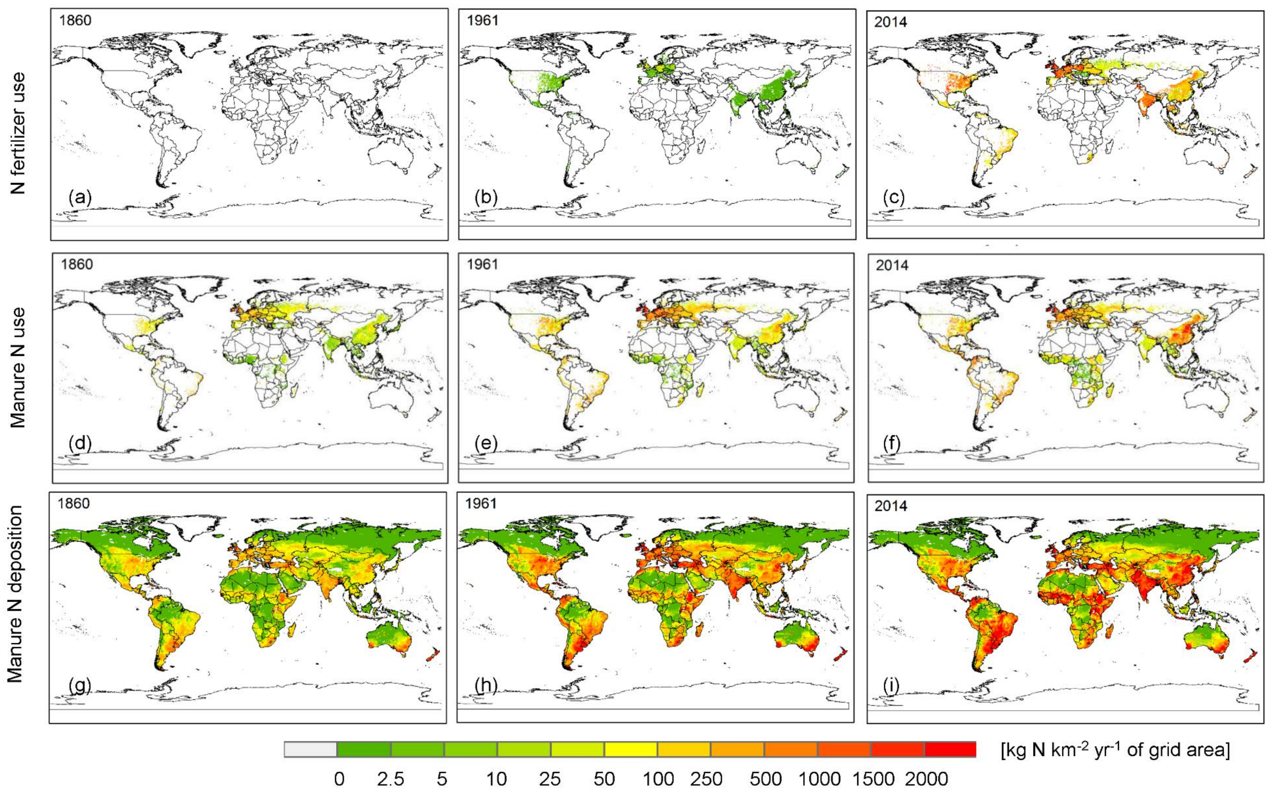

Figure 4. Spatial patterns of N input rates in global grassland systems in 1860, 1961, and 2014: (a)–(c) N fertilizer application rates; (d)–(f) manure N application rates; (g)–(i) manure N deposition rates.

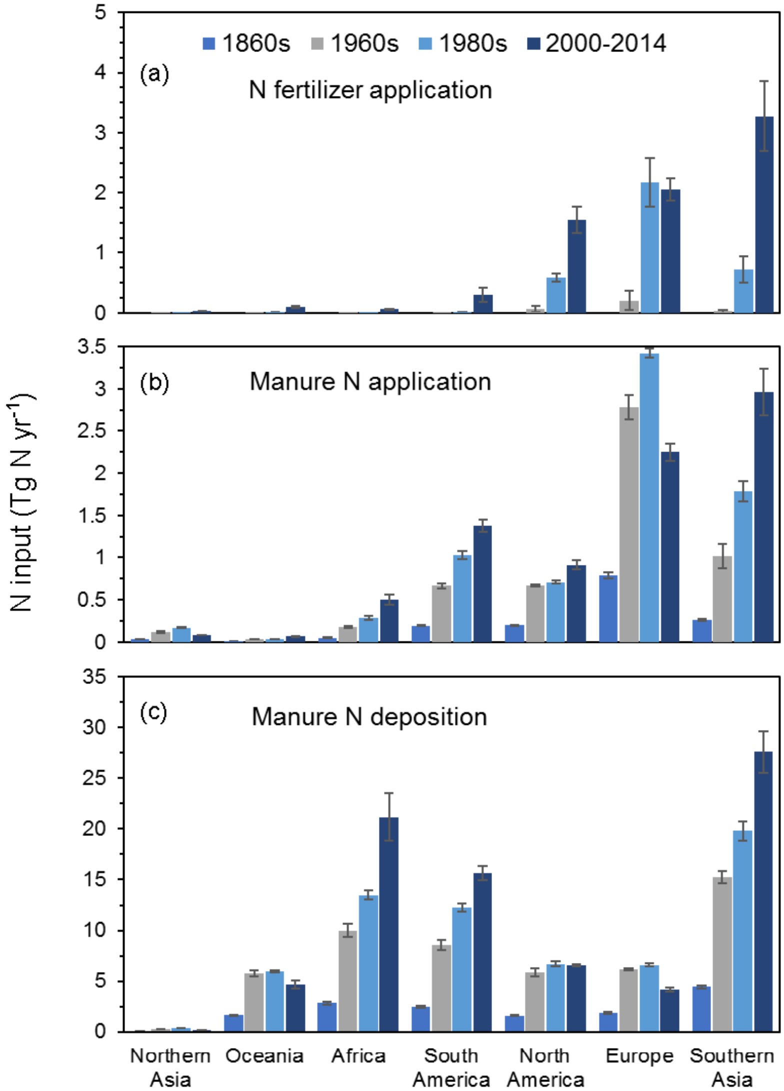

Figure 5. Nitrogen fertilizer use, manure N use, and manure N deposition at regional scales in 1860s, 1960s, 1980s, and 2000–2014. Error bars represent standard deviation within each decade.