Evaluation of historical global gaseous nitrogen emissions from croplands considering NH_4^+ and NO_3^- forming fertilizer species in global fertilizer dataset

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Abstract
We developed a new historical global N fertilizer map (half degree resolution) during 1961-2010 based on FAOSTAT and various global dataset. This new map incorporated the fraction of NH_4^+ (and NO_3^-) into N fertilizer inputs by utilizing fertilizer species information in FAOSTAT. In the data processing, we applied a statistical data imputation method for the missing data in FAOSTAT. The multiple imputation method enabled to fill gaps of the time-series data by the plausible values. In this study, we evaluated NH_3, NO, and N_2O emissions from agricultural soils with biogeochemical model "VISIT" using the developed map. During 1961-2010, synthetic fertilizer consumption increased from 15 Tg-N to 110 Tg-N at global. In this period, the global average fraction of NH_4^+ was about 80% to synthetic N fertilizer consumption. The most countries showed NH_4^+ based fertilizer are dominant, however, the ratio NH_4^+:NO_3^- in N fertilizer inputs shows clear differences among countries and periods. Considering the ratio NH_4^+:NO_3^- in N fertilizer inputs, the simulated NH_3 volatilization were generally reduced, compared to N fertilizer input dealt as only NH_4^+input assumption. On the other hand, NO and N_2O emissions shows both positive and negative impacts using the NH_4^+:NO_3^- fertilizer map. Our new map can be utilized and bring new insights in the global model studies for the assessment of historical terrestrial N cycling changes.

Key Words
Global N assessment, synthetic N fertilizer, FAOSTAT, NH_4^+ and NO_3^-, N gases emission

Introduction
Human activities have greatly disturbed terrestrial nitrogen cycling, especially since the industrial revolution. Synthetic nitrogen fertilizers and fossil fuel combustion are large sources of reactive nitrogen in terrestrial ecosystems. The use of synthetic nitrogen fertilizer rapidly increased after the birth of the Haber–Bosch process in the early 20th century. N fertilizer inputs into terrestrial ecosystems are further increasing in the 21st century with world population and economic growth. Recently, the reactive N loading from these sources on terrestrial ecosystems has been estimated at twice that of biogenic N fixation (Gruber et al., 2009). However, there are still large uncertainties in individual effects of NH_4^+ and NO_3^- in N fertilizer on terrestrial N cycling at global scale. The types of synthetic fertilizer is important factor for gaseous Nr release and leaching from crop- lands due to the different chemical characteristics and bioavailability for microbes and crops. For example, to the amount of NH_3 volatilization from N fertilized soils, whether ammonium (NH_4^+) (or NH_4^+-forming N fertilizers such as urea) or not is critical regulation factor to the Nr loss. Also, the types of fertilizer (e.g., nitrate salts, ammonium salts and urea, and anhydrous NH_4^+) determined the other N oxide gases emissions from fertilized soil (Bouwman, 1996). The early works by Matthews (1994) considered the types of N fertilizer in FAO statistics for global estimation of NH_4^+, however, as far, there are no available historical N fertilizer map considering types of N on a global basis.

In this study, to assess historical N impacts on a global scale, we constructed a new global spatial–temporal explicit N fertilizer input map for the years 1961–2010 and estimated the fractions of NH_4^+ and NO_3^- in N fertilizer inputs using FAOSTAT. With the N fertilizer map, we evaluated historical N gas exchanges from cropland soils in N cycling using a process-based ecosystem model called VISIT (Vegetation Integrative Simulator for Trace Gases) (Inatomi et al., 2010). In addition, we assessed the impacts of consideration of the ratio NH_4^+:NO_3^- in fertilizer input to gaseous N emissions.

Methods
Developing historical global N fertilizer map

Prior to the downscaling processes for global N fertilizer map, we applied the statistical data imputation to FAOSTAT data due to there existing many missing data especially in developing countries. For the data imputation, we used multiple data imputation method proposed by Honaker & King (2010), which was based on bootstrapping EMB algorithm with assumption of multivariate normal distribution. The statistics of various types of synthetic fertilizer consumption are available in FAOSTAT, which can be sorted by the content of NH_4^+ and NO_3^-, respectively. So, we converted total N fertilizer input to NH_4^+ and NO_3^- inputs from the fertilizer species composition. To downscaling the country by country N fertilizer consumptions data to the 0.5° x 0.5° grid-based map, we used historical land-use map in Earthstat (Rumankutty et al., 1999). Before the assignment of N fertilizer in each grid, we weighted the double cropping regions to be more N fertilizer input on to these regions. Using M3-Crops Data (Monfreda et al., 2008), we picked up the dominant cropping species in each grid cell. After that, we used Crop Calendar in SAGE dataset (Sacks et al., 2010) and determined schedule of N fertilizer input in each grid cell using dominant crop calendar. Base fertilizer was set to be 7 days before transplanting and second fertilizer to be 30 days after base fertilizer application.

Simulation settings

In this study, we evaluated N fertilizer and N deposition inputs on global terrestrial N cycling using ecosystem model "VISIT" (Inatomi et al, 2010). We considered N depositions onto croplands using ACCMIP dataset (Shindell et al., 2013). We used Harmonized land use map "LUHa v1.0" (Hurtt et al., 2011) for cropland area map during the simulation period. The simulation was conducted using climate reanalysis data from the U.S. National Centers for Environmental Prediction (NCEP) and the U.S. National Center for Atmospheric Research (NCAR). To assess the impact of the ratio NH_4^+ and NO_3^- in N gases emission, we used N fertilizer input in the simulation by two different ways; (i) all N fertilizer input dealt as NH_4^+, and (ii) considering the ratio NH_4^+:NO_3^- as in the product.

Results and Discussions

Using multiple imputation method, we were able to generate a filled dataset for national fertilizer consumption of all the countries in FAOSTAT database between 1961-2010. Also, this procedure enables to estimate the fraction of NH_4^+ (NO_3^-) in the total N fertilizer inputs with types of chemical fertilizer information. NH_4^+: NO_3^- among countries (Fig 1) and regions (Fig 2). Global mean of the fraction of NH_4^+ to N fertilizer input was about 80% in this period. North America consistently showed the lower fraction of NH_4^+ to N fertilizer than that of global average in this period. On the other hand, the fraction of NH_4^+ to N fertilizer in Latin America increased from 60% to global average by 2010.

Fig. 3 shows global NH_3, NO, and N_2O emissions during 1961-2001. In this period, NH_3 volatilization in East Asia (especially in China) considerably increased due to the expansion of cropland area and the intensification of N fertilizer rates (data not shown). Also, NO and N_2O emission increased accompanied with cropland area. However, N_2O emissions in East Europe seem to be reduced during 1961 to 2010.

Finally, we assessed how the ratio in N fertilizer input, land-use change, and climate affect historical gaseous N emission at global. In the most regions, the reduction of NH_4^+ input directly reduced NH_3 volatilization. For NO and N_2O, there were the both positive and negative effects on the emissions due to the different contributions of nitrification and denitrification processes.

Conclusions

The developed fertilizer map in this study is planned to publish as open data (Nishina et al., in prep). The ratio of NH_4^+ to NO_3^- in N fertilizer has not been considered in the previous studies, e.g., biogeochemical models for global N cycling studies. Although, in the fertilized soils, NH_4^+ and NO_3^- affect different biogeochemical processes (e.g., nitrification, denitrification), there were limited attention to the chemical forms in the N fertilizer dataset in the modelling studies. Thus, our map brings some new insights for the terrestrial N inputs and impacts via synthetic fertilizer from the view to NO_3^-.

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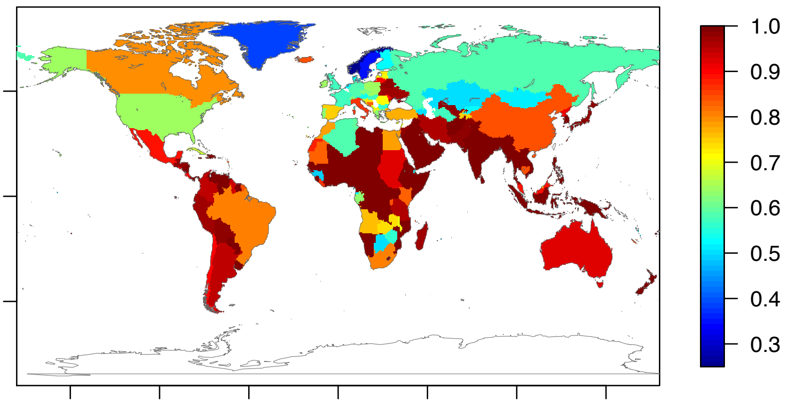

Figure 1. NH_4^+ fraction in national N fertilizer inputs at 2000.

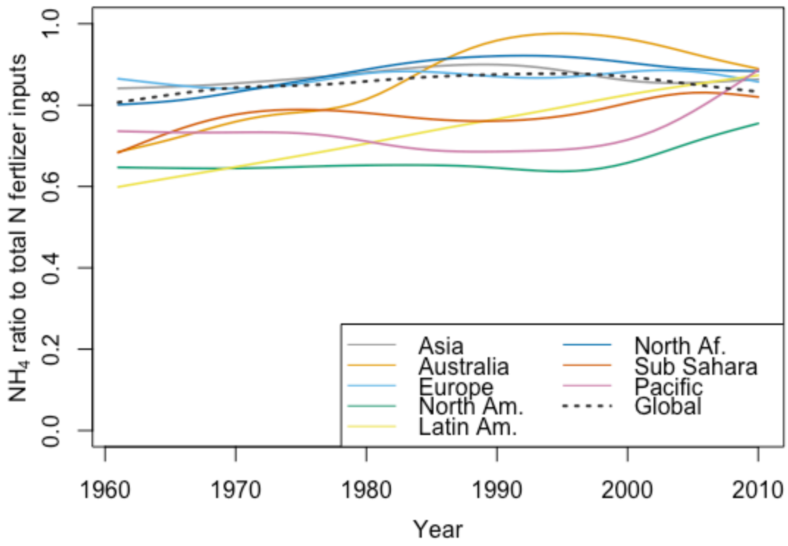

Figure 2. Regional NH_4^+ fraction in total N inputs during 1961-2010.

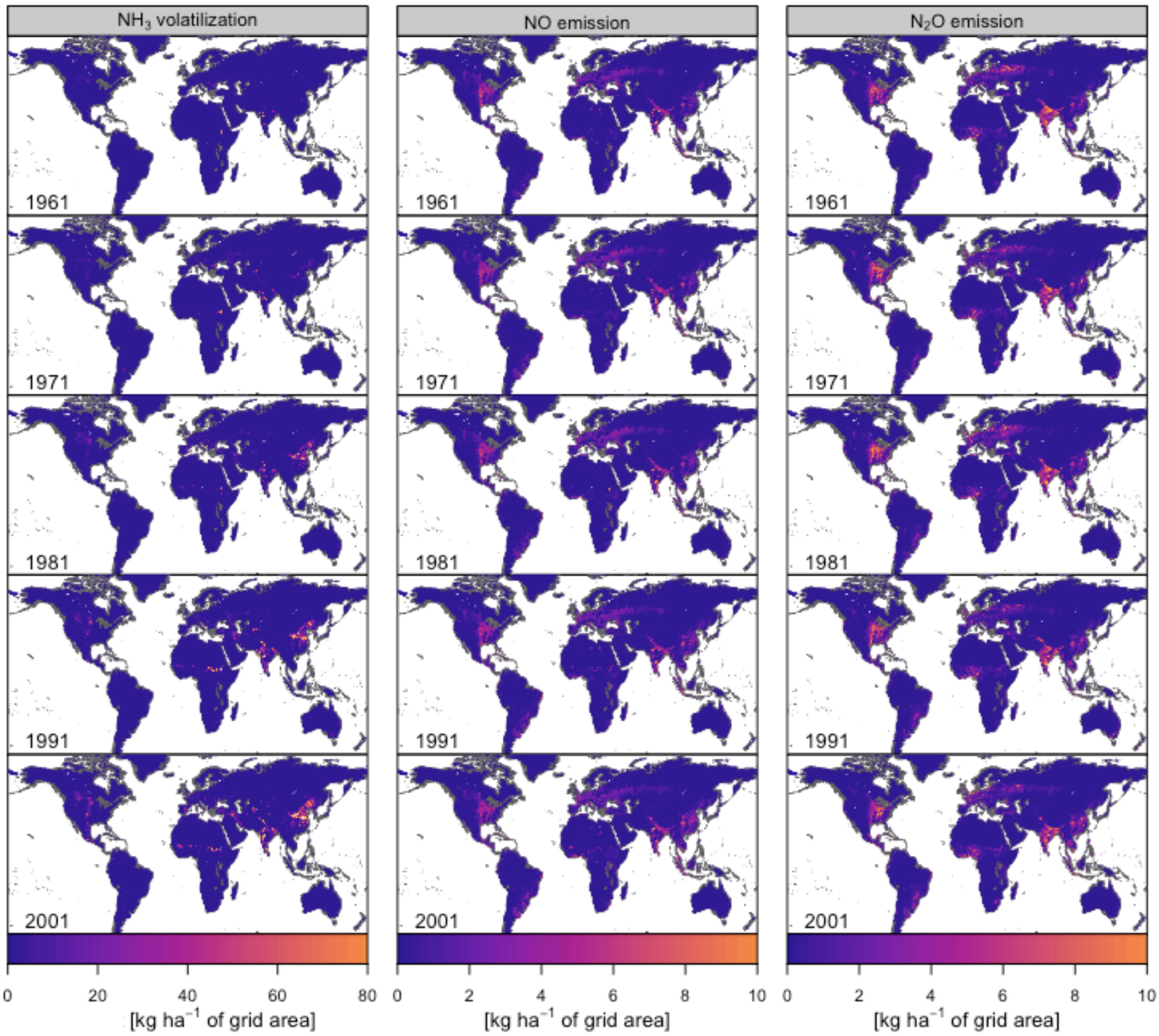

Figure 3. Historical changes in spatial distribution of NH_3 volatilization, NO, and N_2O emission.