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Quantifying nitrogen leaching response to fertilizer additions in China's cropland☆

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A B S T R A C T

Agricultural soils account for more than 50% of nitrogen leaching (L_N) to groundwater in China. When excess levels of nitrogen accumulate in groundwater, it poses a risk of adverse health effects. Despite this recognition, estimation of L_N from cropland soils in a broad spatial scale is still quite uncertain in China. The uncertainty of L_N primarily stems from the shape of nitrogen leaching response to fertilizer additions (N_{rate}) and the role of environmental conditions. On the basis of 453 site-years at 51 sites across China, we explored the nonlinearity and variability of the response of L_N to N_{rate} and developed an empirical statistical model to determine how environmental factors regulate the rate of N leaching (LR). The result shows that L_N-N_{rate} relationship is convex for most crop types, and varies by local hydro-climates and soil organic carbon. Variability of air temperature explains a half (~52%) of the spatial variation of LR. The results of model calibration and validation indicate that incorporating this empirical knowledge into a predictive model could accurately capture the variation in leaching and produce a reasonable upscaling from site to country. The fertilizer-induced L_N in 2008 for China's cropland were 0.88 ± 0.23 TgN (1σ), significantly lower than the linear or uniform model, as assumed by Food and Agriculture Organization and MITERRA-EUROPE models. These results also imply that future policy to reduce N leaching from cropland needs to consider environmental variability rather than solely attempt to reduce N_{rate}.

1. Introduction

As a soluble and mobile contaminant, fertilizer nitrogen not used by crops is prone to leaching through the root zone into groundwater (Kramer et al., 2006; Wick et al., 2012). When excess levels of nitrate (NO_3^-) accumulate in groundwater, it poses a risk of methaemoglobinaemia in infants and risks of gastric and oesophageal cancers (Fewtrell, 2004; Nolan and Hitt, 2006; WHO, 2011). Nitrate in groundwater aquifers has represented a major environmental problem worldwide, particularly in Europe, USA, and South and East Asia (Nolan and Hitt, 2006; Wick et al., 2012; Gu et al., 2013; Rodríguez-Lado et al., 2013). In China, where groundwater provides 20% of the total water supply (Qiu, 2011; Zheng and Liu, 2013), the nitrate levels in more than 25% of samples taken in croplands during 2000–2009 exceeded the World Health

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Abbreviation

N_{rate}	Nitrogen (N) fertilizer application rate per unit sowing area
L_N	Amount of N leaching
LR	N leaching rate
ΔLR	Change in LR per unit of N_{rate}
L_0	Base N leaching
LR^0	Initial value of LR without the impact of fertilization
f_k	Environmental factors (soil attribute, hydro-climates, sampling Factors)
α_k	Coefficient for f_k in LR term
b	Intercept in LR term
β_k	Coefficient for f_k in LR^0 term
c	Intercept in LR^0 term
Temp	Average air temperature during the observation period
P+I	Sum of precipitation and irrigation during the observation period
SOC	Soil organic carbon
pH	Soil pH value
BD	Soil bulk density
Clay	Soil clay content
TN	Soil total nitrogen
Depth	Sampling depth
NPCP	National pollution census program

Organization (WHO) recommended maximum limit of 10 mg/L (NO_3—N; Gu et al., 2013). Such conditions indicate the likelihood of adverse health effects in nearby rural residents, particularly in north and northwest China (Liu et al., 2006). While fertilizer use accounts for more than 50% of the nitrogen leaching into groundwater in China (Gu et al., 2013), nitrogen loss from cropland is poorly understood (Zheng and Liu, 2013).

Nitrogen leaching depends on the flow rate of infiltrating water and the concentration of nitrogen in the root zone (Riley et al., 2001; Zheng and Bennett, 2002; Li et al., 2006). In the root zone, nitrogen concentrations are controlled by nitrogen additions minus nitrogen losses via ammonia (NH_3) volatilization, denitrification and crop uptake (Van Drecht et al., 2003; Keuskamp et al., 2012; Bouwman et al., 2013). Therefore, the suggested linear relationship between nitrogen application rate (N_{rate}, kgN·ha^{-1}) and fertilizer-induced nitrogen leaching (L_N, kgN·ha^{-1}) is inconsistent with the denitrification process within the root zone (IPCC, 2006; Amon-Armah et al., 2015), which in fact varies significantly with nitrogen input (Jahangir et al., 2012; Bouwman et al., 2013; Zhou et al., 2014).

According to a growing number of field observations at specific sites, LRs (the rate of N leaching) also varies with environmental conditions (f_k), focusing on hydroclimatic factors or soil properties (Van Drecht et al., 2003; Velthof et al., 2009; Nolan and Hitt, 2006; Chen et al., 2014). First, the sensitivity of leaching rate (LR, %) to N_{rate} (defined as ΔLR, the first-order derivative of LR to N_{rate}) may be controlled by f_k. For example, Wick et al. (2012) observed that ΔLR values were systematically larger after previous cropping or when the amount of precipitation was greater than 281 mm·yr^{-1}. Second, LRs under a given N_{rate} may be regulated by levels of f_k that determine denitrification rates and the residence time of nitrate in the root zone (Van Drecht et al., 2003; Velthof et al., 2009). However, those results only reflect the effects of f_k under a limited range of the environmental factors. When upscaling to national or larger scale predictions of N leaching rate, unreasonable fluxes may occur widely for environments that are not captured by the small group of field experiments.

To test if it is the similar case in China, the synthesized N leaching measurements were used in this study to investigate the nonlinearity and variability of the response of L_N to N_{rate} for cropland across diverse environmental conditions. A new empirical model (PKU-NLEACH) calibrated by the Bayesian Recursive Regression Tree algorithm (BRRT v2) was also proposed to determine the shape of LRs related to controlling factors. The results were then compared with the models without the considerations of nonlinearity and variability, including those used in MITERRA-EUROPE model (Velthof et al., 2009) and FAO Assessment model for soil nutrient balance (de Willigen, 2000; Roy et al., 2003; Liu et al., 2010). In the end, this study assessed how f_k modulates the spatial variability of LR and discussed about the implications on regional budgets.

2. Data and methods

2.1. Field observations in China

Field observations of L_N, N_{rate}, and f_k were collected from sites of both upland and paddy soils from 50 peer-reviewed publications of field trials (80% for upland crops and 20% for paddy rice). Lysimeters were used to quantify the water flow and N movement through agricultural soils. Installation details were as described in Huang et al. (2011). Retailed flux measurements had to have at least two levels of N_{rate}, in addition to a zero-N control experiment. Three types of samples were excluded from the analysis: (i) measurements that included the use of such chemicals as nitrification inhibitors that are not widely applied in China, (ii) measurements with the duration not covering all split-applied fertilizations for a crop growing season, and (iii) measurements without zero-N controls. This reduced dataset was comprised of 453 site-years (397 for upland crops and 56 for paddy rice) at 51 sites observed from 1992 to 2013 (Fig. 1 and Data S1), which could produce 338 pairs of LR. However, only 388 site-years at 37 sites with at least three N-input levels could produce 96 pairs of ΔLR.

To interpret the nonlinearity of nitrogen leaching to N_{rate}, we analyzed the f_k (Table 1) known to affect the soil N cycling and transport in the root zone when available in field experiments. However, fertilization methods (e.g., broadcast, incorporation, injection, and deep placement) and tillage practices were not considered due to lack of information about in the surveyed literature. All site-years in the original studies were used and were averaged by replicates if necessary. Few of the missing climate and soil properties within observation periods were supplemented by the 1-km Harmonized World Soil Database (HWSD) v1.2 (http://www.iiasa.ac.at) and by the 0.1-degree China Meteorological Forcing Dataset (CMFD; Chen et al., 2011).

2.2. Statistical analysis

The L_N model assumed a quadratic form of nitrogen leaching change along a gradient of N_{rate}: $L_N = [\Delta LR \cdot N_{rate} + LR^0] \cdot N_{rate} + L^0$, because most of control experiments were determined to be well expressed ($R^2 > 0.9$ and $P < 0.01$ for 89% field experiment; Fig. S2). Before investigating the nonlinearity and variability of the response of L_N to N_{rate}, LR was calculated for each non-zero N_{rate} as a difference between nitrogen leaching (L) at N_{rate} and the corresponding control (L^0) divided by N_{rate} (unit: %), and ΔLR is calculated as the change in LRs per unit of incremental N_{rate} (unit: %·kgN^{-1}·ha). Their meanings were also graphically described in Fig. 2a and b. A Shapiro–Wilk test was then performed to check whether their