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## Detection and attribution of nitrogen runoff trend in China's croplands<sup>☆</sup>



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## ABSTRACT

Reliable detection and attribution of changes in nitrogen (N) runoff from croplands are essential for designing efficient, sustainable N management strategies for future. Despite the recognition that excess N runoff poses a risk of aquatic eutrophication, large-scale, spatially detailed N runoff trends and their drivers remain poorly understood in China. Based on data comprising 535 site-years from 100 sites across China's croplands, we developed a data-driven upscaling model and a new simplified attribution approach to detect and attribute N runoff trends during the period of 1990–2012. Our results show that N runoff has increased by 46% for rice paddy fields and 31% for upland areas since 1990. However, we acknowledge that the upscaling model is subject to large uncertainties (20% and 40% as coefficient of variation of N runoff, respectively). At national scale, increased fertilizer application was identified as the most likely driver of the N runoff trend, while decreased irrigation levels offset to some extent the impact of fertilization increases. In southern China, the increasing trend of upland N runoff can be attributed to the growth in N runoff rates. Our results suggested that increased SOM led to the N runoff rate growth for uplands, but led to a decline for rice paddy fields. In combination, these results imply that improving management approaches for both N fertilizer use and irrigation is urgently required for mitigating agricultural N runoff in China.

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### 1. Introduction

Meeting food security targets while simultaneously reducing reactive nitrogen losses has drawn attention from both scientists and the public (Chen et al., 2014; Mueller et al., 2012; Tilman et al., 2011; Zhang et al., 2015). Large amounts of anthropogenic nitrogen (N) inputs have resulted in excess N being lost in runoff from croplands to water bodies and the atmosphere worldwide (Cui et al., 2014; Leip et al., 2011; Seitzinger et al., 2010). As one of the consequences, increased occurrences of aquatic eutrophication and

ecosystem degradation were observed, particularly in China and South Asia (Paerl et al., 2014). Reliable detection and attribution of cropland N runoff are crucial for policy makers and farmers to develop site-specific N management strategies (Cherry et al., 2008). Although cropland N runoff is substantial in China (e.g.,  $2.1 \pm 0.2$  or  $0.8 \text{ Tg N yr}^{-1}$  estimated by Gu et al. (2015) and Wang et al. (2014), respectively), large-scale, spatially detailed N runoff trends and its attribution remain poorly understood.

Cropland N runoff, defined as a generation process of N loss via surface runoff, depends on environmental conditions and agricultural management practices (Zhang et al., 2016). This complexity makes large-scale N runoff difficult to estimate using empirical models. Plot-scale N runoff flux data from croplands are also difficult to scale up into spatially detailed maps because of spatio-temporally varying results (Shen et al., 2012). Currently, an export

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### Acronyms and abbreviations

$R_{TN}$	Cropland N runoff
$R^0$	Background N runoff
$SOM$	Soil organic matter
$TN$	Soil total nitrogen
$N_{rate}$	Nitrogen (N) fertilizer application rate per unit sowing area
$x_k$	Environmental variables
$RR$	$R_{TN}$ per unit N fertilizer additions
pH	Soil pH value
Clay	Soil clay content
Temp	Mean air temperature
$W$	Sum of precipitation and irrigation within observation period
$CE$	Correction coefficient

coefficient approach has been widely used to estimate cropland N runoff (Hao et al., 2006; Liu et al., 2010; Velthof et al., 2009; Wang et al., 2014). For example, the first National Pollution Census Program of China (NPCP) provided a collection of N runoff flux coefficients for different geographical regions in China, determined by fitting cross-sectional site data to an export-coefficient model (Wang et al., 2014). Nevertheless, substantial evidence gathered from field observations indicates that linear and homogeneous models are rarely capable of capturing the spatial variability of N runoff at regional scale (Schaefer and Alber, 2007; Sobota et al., 2009; Hou et al., 2016). This highlights the difficulty of accurately predicting its future evolution as well as quantifying the impacts on aquatic ecosystems.

While it is still challenging to attribute contributions of each individual driving factor (e.g., climate condition, agricultural management practices) to the cropland N runoff trend assessment, statistical correlation or regression analyses have been widely applied (Korsaeth and Eltun, 2000; Stalnacke et al., 2015) over past decades. However, this approach has two potential limitations. Firstly, statistical analyses of historical  $R_{TN}$  generally characterizes the related major drivers, thus includes the signals not only from the temporal trends, but also from inter-annual or decadal variability. Secondly, the use of statistical analysis generally assumes that the effects of drivers on N runoff are linear and independent of each other (Piao et al., 2015). However, a growing number of studies based on both data from field experiments and theoretical analyses indicated non-linear responses of N runoff to changes in driving factors as a consequence of complex interactions (Hou et al., 2016). Although these limitations in attribution analysis could be overcome through the application of process-based models (Hao et al., 2006; Abbaspour et al., 2015; Liu et al., 2016), core limitations of such simulation models are the large uncertainties arising from model structure and parameter choice. One way to separate the contribution of natural and anthropogenic controls is to use the Kaya Identity concept developed in economics (Raupach et al., 2007), which is adopted when studying climate change and hydrological science (Streimikiene and Balezentis, 2016; Wang et al., 2015b).

To quantify and attribute cropland N runoff trends during past decades, we analyzed the data in this study is based on an upscaling model following a new simple attribution approach. Synthesized field measurements were used for model calibration and cross-validation based on the Bayesian Recursive Regression Tree algorithm version 2.0 (BRRT v2, Zhou et al., 2015), utilizing high-

resolution gridded datasets including climate conditions, soil attributes, and agricultural management practices. First, we assessed inter-annual dynamics of cropland N runoff derived from the data-driven upscaling model to detect trends for the period from 1990 to 2012. Second, we compared the proportional change rate of each driver to upscaling results of  $R_{TN}$ , which allowed us to diagnose the contributions of different drivers. Finally, we discussed how each driver modulates the temporal trend of  $R_{TN}$  and the implications for site-specific N management.

## 2. Data and methodology

### 2.1. Dataset

Based on the National Pollution Census Program of China (NPCP) and datasets published by the scientific community, *in situ* measurements of N runoff and associated variables in each plot were collated from 100 experimental sites for both rice paddy and upland fields (i.e., as a flooded parcel of arable land used for growing rice and non-rice crops, respectively). Water samples were collected in the drainage outlet for each rainfall event in most of the measurements, where the runoff volume was consecutively measured within the observation period. N concentrations in water samples were analyzed using ultraviolet spectrophotometric methods, following the Standard Methods for the Examination of Water and Wastewater approach for China (SEPA, 2002). Precipitation within the observation period and soil properties (0–20 cm depth) at the beginning of the experiment were synchronously monitored. Missing values of soil properties or climatic factors at a few sites were supplemented with data from the China Soil Scientific Database (<http://vdb3.soil.csdb.cn/>) based on the corresponding soil type of the county or from the 0.1-degree China Meteorological Forcing Dataset (CMFD) v0106 (<http://data.cma.cn/>) depending on its geographic coordinates. Information on agricultural management practices including N fertilizer application rate, irrigation amount, fertilizer types, and crop types were recorded, including the timing of the application. The dataset comprised 535 site-years data (293 for upland and 242 for rice paddy fields) (Fig. 1a and Supplementary Data S1), and can be considered representative of most major cropping areas except northwestern China (Fig. 1).

### 2.2. Data-driven upscaling model

We developed an upscaling model which accounts for the effects of environmental conditions and agricultural management (Eq. (1)). Specifically, N fertilizer application rate ( $N_{rate}$ ) and environmental conditions ( $x_k$ ) are directly included as independent variables, whereas fertilizer application and crop types are considered as correction terms in the model:

$$R_{TNl} = RR_l(x_k) \cdot N_{rate} + R^0_l(x_k) + \varepsilon, \forall l = 1, 2, \dots, L, x_k \in \Omega_l \quad (1a)$$

where

$$RR_l = RR_l^*(x_k) \times CE_i(RR) \times CE_j(RR), \quad (1b)$$

$$R^0_l = R^{0,*}_l(x_k) \times CE_j(R^0), \quad (1c)$$

$$RR_l^*(x_k) = f(x_k) \cdot N_{rate} + g(x_k), R^{0,*}_l(x_k) = h(x_k), \quad (1d)$$

and  $i$  and  $j$  represent the index of fertilizer types and crop types, respectively;  $l$  and  $L$  are the index and number of piecewise functions.  $x_k$  is climatic condition or soil attribute. Observations (Fig. S1) of air temperature, water input and soil clay content can be used as

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