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Short Communication

Legacy nitrogen fertilizer in a rice-wheat cropping system flows to crops more than the environment

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Rice-based cropping systems are prevalent in Asia, accounting for 16% of global nitrogen (N) fertilizer consumption and providing billions of calories [1]. Among these, the rice-wheat cropping system (RWCS) stands out as a critical contributor to global food security, covering an extensive area of 23.5 million hectares (mha) in Asia and sustaining 4.4 billion people [2]. Nevertheless, the substantial reliance on N fertilizer raises concerns about the long-term fate of N within this system. Negative environmental impacts, such as soil acidification, water eutrophication, and greenhouse gas emissions associated with the RWCS and other annual cropping systems, are often attributed to excessive N fertilizer inputs [3]. However, a significant portion of fertilizer N losses in annual cropping systems arises from the gradual release of N from the fertilizer-derived soil N pool [4].

Improving nitrogen use efficiency (NUE) promises both agronomic and ecological advantages [3]. However, the challenge of environmental N losses goes beyond mere fertilizer over-application; it's a systemic issue. Previous studies in dryland systems often posits that residual fertilizer N contributes significantly to environmental N losses [4]. Conversely, it can also become sequestered in the soil or taken up by future crop across disparate systems [5,6]. Paddy fields, characterized by deliberate flooding and puddling, exhibit increased soil organic matter (SOM) and clay content, resulting in a slower mineralization rate compared to well-aerated upland soils [7]. While global averages indicate that only 37% of applied ^{15}N-labeled fertilizer in paddy soils is harvested in the crop [8], the long-term destiny of the remaining N fertilizer remains mysterious, with its loss to the environment not being an inevitable outcome.

Residual fertilizer N, if retained in soils, can potentially provide a significant amount of N to subsequent crops. This suggests that the short-term assessments of NUE may not fully capture the complete extent how effectively crops utilize fertilizer N. Traditionally, research on N fertilizer recovery efficiency has focused primarily on single-season evaluations, with scant attention given to the absorption of residual ^{15}N by crops in subsequent growth periods [9]. Therefore, to accurately assess the fate of fertilizer N and to derive a precise estimation of NUE, the long-term residual effects of previously applied N should be fully considered. To date, however, no study has quantified the destiny of residual fertilizer N and obtain a precise measure of NUE, it is imperative to consider the long-term residual effects of previously applied N. Up to this point, however, no study has quantified the long-term fate of residual fertilizer N in paddy soils, as it is technically impractical to measure in-situ without causing significant disruption to the experimental environment. This knowledge gap results in substantial uncertainty regarding the exact proportions of residual fertilizer N that are either escape into the environment or get absorbed by crops in subsequent seasons, as well as the duration that residual fertilizer N persists within soil N pools. A potential methodology to quantify N fate in cropping system involves the use of ^{15}N-tracer, with the measurement of soil retention and crop

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uptake. In this study, we investigated the long-term fate of ^{15}N-labeled urea fertilizer were investigated in field lysimeters treated with ^{15}N-labeled urea applied at 100 and 300 kg N ha^{-1} (N100 and N300) during the initial season (in-season) followed by applications of unlabeled urea over 17 years under RWCS in eastern China. Further details can be found in the Supplementary Materials and Methods.

Most studies that have investigated the recovery efficiency (RE) of N fertilizer have traditionally focused solely on the season of application, referred to as in-season RE, with a global average of 42% [10]. However, the RE in subsequent seasons is typically minimal, averaging a mere 4.6% globally [10]. Our study, however, revealed a different pattern. Despite recording a relatively low in-season ^{15}N-labeled RE of 26.7% for N100 and 27.6% for N300, these values cumulatively increased to 43.0% and 38.6% (Fig. 1a), respectively over the ensuing 17 years. Remarkably, the recovery of residual fertilizer N by subsequent crops accounted for approximately one-third of the total RE during this 17-year period. This discovery suggests that NUE within RWCS is systematically underestimated, and that residual fertilizer N constitutes a significant source of for crops under long-term fertilization regimes.

To preserve the integrity of the soil monolith lysimeters, residual measurements of ^{15}N-labeled fertilizer were confined to the top 20 cm soil throughout the observation period until the 17th year harvest. After the first season, 17.8% (N100) and 13.3% (N300) of the applied N fertilizer remained within this upper soil layer, gradually decreasing by ~0.6 percentage points annually in subsequent seasons (Fig. 1b). This reduction can be attributed to crop uptake and losses through gaseous or hydrological processes [5]. By the 17th year, the remaining N fertilizer in the 0–100 cm soil profile was between 10.5% (N100) and 5.73% (N300), with ~70% of it concentrated within the top 20 cm (Fig. 1c). This distribution indicates a sustained potential for crop utilization. Using decay functions (Fig. 1d–e; Table S1 online), we estimated the residence time of fertilizer N residence time in the soil to be merely 23–31 years for N100 and 23–28 years for N300 within the RWCS. This duration is significantly shorter than what was observed in the only other comparable study in a sugar beet-wheat rotation, where it ranged from 80 to 100 years [4].

In summary, after 17 years, 53.5% (N100) and 44.3% (N300) of the tracer fertilizer N were recovered in soil (0–100 cm) while the remaining 46.5% and 55.7%, respectively, were lost (Fig. 1a, c and f; Table S2 online). To comprehensively understand the fate of N during the growing season, we estimated residual N fertilizer in the 0–100 cm soil and root were estimated during the fertilizer application season (Table S3 online, SI Appendix). Our results showed that 28.3% (N100) and 21.9% (N300) of the tracer fertilizers remained within the RWCS, with only 3% remaining in the root system. Consequently, the total N fertilizer loss during the first season amounted to 45.0% (N100) and 50.5% (N300) (Fig. 2a). Assuming complete release of root residual N over the subsequent 33 seasons, the accumulated residual N fertilizer loss rate was only 1.5% and 5.2%, with over 91% of N losses occurring in the first season over the 17-year observation period (Fig. 2a).

Higher N rates were associated with increased residual N losses, exacerbating its environmental impact and reducing plant N availability. This observation aligns with previous research, which identified minimal soil residual N loss except in cases of high N

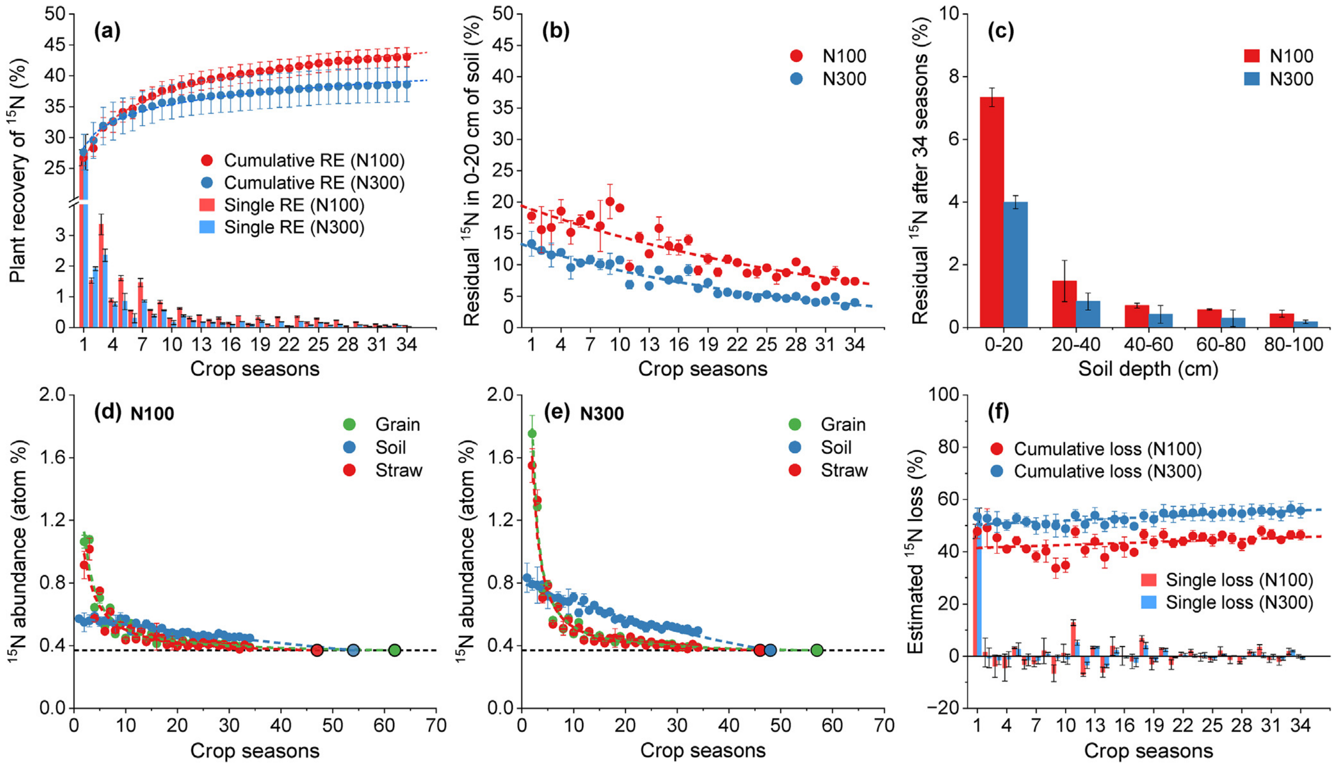

Fig. 1. (a) Cumulative recovery efficiency (RE) and single RE of fertilizer ^{15}N by the plant during 34 seasons. (b) The 0–20-cm residual rate of fertilizer ^{15}N in soil. (c) The distribution of soil residual fertilizer N in 0–100-cm depth lysimeters after 34 continuous cropping. (d, e) Decay functions fitted to ^{15}N values of soils (0–20 cm) and plants. The start time of the model in the grain and straw is the second crop season (2004W), representing the first season in which soil residual fertilizer N emerged. The models indicate that it will take circa 23 to 31 years (N100, d) and 23 to 28 years (N300, e) to achieve the soil background ^{15}N value of 0.371% (black dashed line). The parameters of the decay models are presented in Table S1 online. (f) Estimated fertilizer ^{15}N loss calculated by the balance method based on 0–100-cm soil N retention and crop uptake (a). Fertilizer ^{15}N retention in 0–100-cm soil estimated from the 0–20-cm soil N retention (b) and from residual fertilizer distribution in 0–100-cm depth (c, estimation details are in Supplementary Materials). Data are means of triplicate lysimeters. The colored dotted line is the trend line of the data, and the parameters of the trend equation are presented in Table S2 online. Error bars represent standard errors.

treatments [11]. Furthermore, measurements of reactive N loss indicated that significant losses primarily occurred in the first season, with cumulative losses to ammonia volatilization accounting for mere 0.1% of the residual N fertilizer, and nitrate leaching accounting for 0.21% (N100) and 0.54% (N300) over subsequent two years (Tables S4 and S5 online). These results indicate a lower environmental impact of residual fertilizer N in RWCS, with decline primarily from crop N uptake rather than losses, contrary to previous assumptions for dryland systems [4]. The management of paddy soils, which often undergoes dry-wet rotation, likely contributes to this discrepancy. Paddy soil characteristics developed from long-term rice cultivation are known to inhibit N leaching [7], and prolonged flooding also increases the dissimilatory nitrate reduction to ammonium (DNRA) rate, thereby enhancing N retention [12]. Moreover, the mineralization of residual N in paddy is slow and more compatible with crop N uptake.

However, it is a cause for concern that approximately half of the applied fertilizer N is rapidly lost during the growing season, posing risk to environmental safety and limiting both crop and soil N recovery. This, in turn, affects the productivity of the RWCS. To ensure long-term sustainability, it is imperative to optimize N management strategies, focusing on both in-season and residual NUE within RWCS.

Our study uncovers a regime of "low NUE, high N loss" for RWCS. Despite factoring in long-term residual effects, the cumulative tracer NUE remained low at 40%, with about half of fertilizer N lost to the environment in-season (Fig. 2a, Table S6 online). While in-season soil N retention was modest in RWCS, residual fertilizer N displayed a propensity remain within the system and was more effectively utilized by crops in subsequent seasons. As a response to these findings, we propose a two-step strategy for optimizing N management within RWCS (Fig. 2b):

1. Reducing in-season losses through in-field practices that enhance both crop and soil N recovery, in addition to implementing edge-of-field approaches to mitigate drainage losses.
2. Enhancing the availability of residual fertilizer N to subsequent crops, thereby improving overall NUE across the entire system.

Presently, the majority of N management strategies primarily emphasize the enhancement of in-season NUE and the reduction of N losses constituting "step 1" of our proposed strategy.

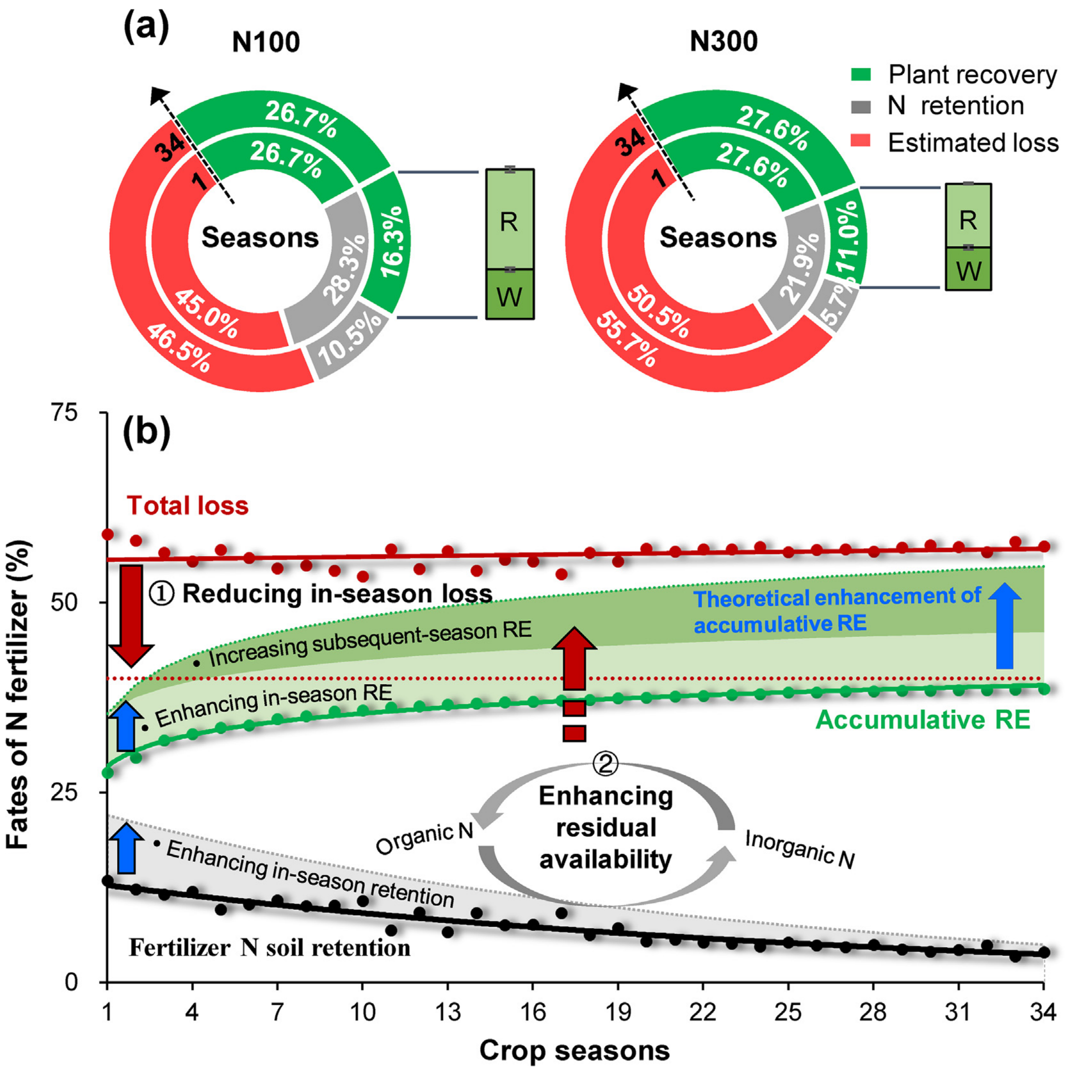

Fig. 2. (a) Plant N recovery efficiency, residual rate, and loss rate of after one season (inner circle) and 34 seasons (outer circle). Residual fertilizer N of the first season includes residual N in the soil (0–100 cm) and in the roots (Table S6 online, estimation details are in Supplementary Materials). The bars represent rice/wheat recovery in the 33 subsequent seasons, and R and W denote subsequent rice and wheat recovery, respectively. Data are means of triplicate lysimeters. (b) A conceptualized two-step strategy for optimal N fertilizer management of RWCS. The solid lines and dots re the actual long-term fate of N fertilizer in RWCS (data from N300); including cumulative N recovery efficiency (RE; green), 0–20-cm soil residual fertilizer N (black), and estimated cumulative N loss based on the balance methods (red). The shades show fertilizer RE (green) and soil residual fertilizer N (gray) enhancement. The darker green shade indicates potential residual fertilizer N availability increase.

Numerous N management practices have been developed, including 4Rs (right source, rate, time, placement) [13] and integrated process management practices (including controlled-release fertilizers, inhibitors, co-application with organics, etc.) [14]. These practices primarily target improving fertilizer NUE and mitigating N losses, with varying degrees of effectiveness [14]. It is worth noting that the acquisition of in-season N fertilizer by crops can be constrained by microbial competition and dynamic crop demands over long growth periods. Given the observed positive effects of residual fertilizer N in promoting subsequent crop uptake within the RWCS, there is an opportunity to maximize NUE by improving soil retention. This represents "step 2" in our proposed strategy – harnessing residual N. Specifically, this involves converting residual fertilizer N in the soil into forms readily available for plant uptake, as illustrated in Fig. 2b.

However, the implementation of step-2 in N management depends on specific soil conditions. When the native supply of soil N supply surpasses the ability to retain exogenous N, leading to a net depletion of soil N pool, the primary focus should be on maximizing in-season fertilizer N retention to replenish the soil N. This becomes the paramount N management strategy.

"Step-2" becomes applicable when the native soil N supply is either equal to or less than exogenous N retention, allowing net soil N accumulation. Continued net N accumulation may eventually lead to N saturation, increasing the risk of residual N loss. Consequently, controlling excess residual N through "step-2" becomes crucial in such complementing in-season management efforts. However, directly managing residual N can be challenging. Studies suggest that specific carbon inputs can enhance N retention by reestablishing the carbon and N cycles [15]. Yet, practices optimally promote long-term N retention remain limited across croplands [15]. Additionally, it is impractical to regulate residual N independently because distinguishing between recent, historical, and native soil N pools is challenging under field conditions. Instead, a more realistic approach may involve optimizing the turnover of soil organic matter (SOM) in N-surplus soils to regulate mineralizable N, including residual N, which is susceptible to loss (further details are discussed in the Supplementary Discussion).

Hence, in addition to in-season management, adopting a comprehensive approach that reduces cumulative N losses while improving cumulative NUE through enhanced residual N retention capacity and availability (as depicted in Fig. 2b) is crucial for sustainable practices that ensure both food security and environmental integrity.

Author Contributions

Xu Zhao, Xiaoyuan Yan and Guangxi Xing performed and supervised the research; Yingying Wang analyzed plant and soil samples; Yingying Wang and Xu Zhao presented the results. Xu Zhao and Yingying Wang wrote the original draft, Siyuan Cai, Jagdish K. Ladha, Michael J. Castellano and Longlong Xia revised the manuscript; Yingxin Xie and Zhengqin Xiong participated in the experiments. Baojing Gu, Zhengqin Xiong and Xiaoyuan Yan commented on the manuscript.

Conflict of interest

The authors declare no competing interests.

CRediT authorship contribution statement

Xu Zhao: Conceptualization, Funding acquisition, Methodology, Writing-original draft, Visualization, Supervision. **Yingying Wang:** Data curation, Writing-original draft, Investigation, Visualization. **Siyuan Cai:** Writing-review & editing, Visualization. **Jagdish K. Ladha:** Writing-review & editing. **Michael J. Castellano:** Writing-review & editing. **Longlong Xia:** Writing-review & editing. **Yingxin Xie:** Investigation. **Zhengqin Xiong:** Investigation, Writing-review & editing. **Baojing Gu:** Writing-review & editing. **Guangxi Xing:** Conceptualization, Funding acquisition. **Xiaoyuan Yan:** Conceptualization, Writing-review & editing.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scib.2024.02.027.

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