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Optimizing nitrogen input for sustainable tea production: an integrated assessment of soil aggregate stability, base-cation retention and quality improvement in an 8-year field study

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KEYWORDS

Exchangeable base cations, nitrogen application, soil aggregates, soil carbon, soil nitrogen, tea quality, tea yield

HIGHLIGHTS

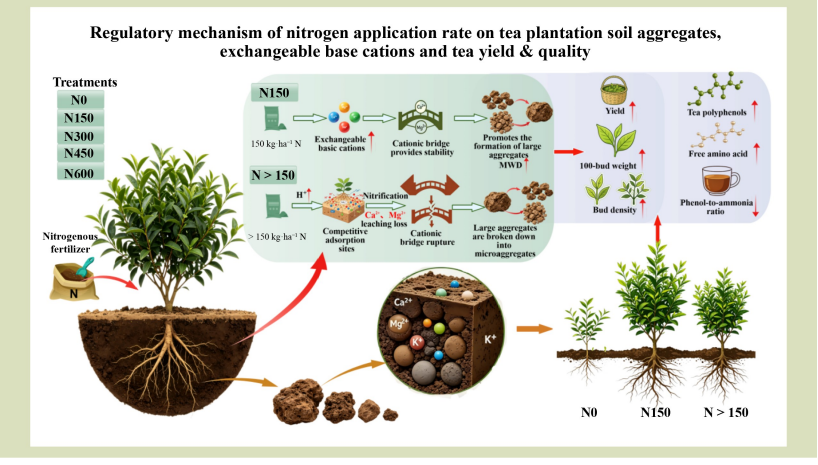
- Low N (150 kg·ha^{-1}) maximized tea yield and quality.
- Low N increased macroaggregates and mean weight diameter.
- N of > 150 kg·ha^{-1} depleted exchangeable Ca^{2+} and Mg^{2+} in aggregates.
- Cation bridging loss disintegrated large aggregates under excessive N (> 150 kg·ha^{-1} N).
- Aggregate-scale base cations better indicate soil fertility decline.

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GRAPHICAL ABSTRACT



ABSTRACT

Excessive nitrogen application in tea plantations can accelerate soil acidification and the loss of exchangeable base cations. However, how these changes are partitioned among soil aggregate fractions and linked to tea productivity remains unclear. An 8-year field experiment with five N application rates (0, 150, 300, 450 and 600 kg·ha^{-1}) was conducted in a tea plantation with red soil, initial pH 4.2 and cv. Tieguanyin in Anxi County, Fujian Province. Soil aggregate composition and stability were determined by wet sieving to assess the effects of N rates on aggregate stability, exchangeable base cations and nutrient contents, and tea yield and quality. N application significantly affected the stability of soil aggregates. The low N treatment (150 kg·ha^{-1} N) gave the highest mean weight diameter (MWD), indicating the

greatest aggregate stability. The results demonstrated that N application rates have nonlinear effects on soil properties and tea yield. During this 8-year-long experimental study, the low-nitrogen treatment maintained higher net concentrations of exchangeable alkali metal cations, indicating that its enhanced cation retention capacity is sustainable rather than a transient pulsed effect occurring after fertilizer application. Additionally, this treatment significantly increased the proportion of macroaggregates and the MWD content, thereby optimizing the soil physical structure. This structural improvement, together with enhanced nutrient availability and moderated soil acidification, was associated with the peak yield and quality observed under this treatment. In contrast, excessive N application severely depleted exchangeable Ca^{2+} and Mg^{2+} through intensified nitrification and H^+ competition, disrupted cationic bridging, and disintegrated large aggregates into smaller fractions, consequently compromising soil stability. These findings confirm that low-nitrogen represents the optimal fertilizer application strategy. Mechanistic analysis indicates that the improved tea quality with addition of 150 kg·ha^{-1} N is associated with increased levels of alkali-hydrolyzable-N, and exchangeable Ca and Mg within aggregates, which are positively correlated with soil pH. Therefore, to improve soil quality while enhancing tea yield and quality, it is recommended that N fertilizer input be maintained at a low rate (150 kg·ha^{-1}) in tea plantations in southeastern Fujian under the conditions of this study.

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1 Introduction

Beverages made with tea are widely consumed worldwide because of its distinctive flavor and rich nutritional and bioactive composition. As a long-established Chinese export commodity, tea also has substantial economic value[1]. Tea is a perennial evergreen woody crop with a relatively high demand for nitrogen, partly because repeated harvesting removes large quantities of N-rich young shoots from the plantation[2]. In practice, however, increasing N inputs to sustain high yields often results in low fertilizer-use efficiency[3]. Only about 25%–50% of applied N is assimilated by tea plants, while substantial amounts may be immobilized in soil or lost through leaching, erosion and volatilization[4,5]. In Anxi County, N surplus has been reported in a large proportion of tea plantations and excessive N application has been associated with nutrient losses and declining tea productivity[6].

In Anxi County, excessive N application, particularly with urea, is an important driver of soil acidification and can adversely affect tea yield and quality[7]. Exchangeable base cations are key indicators of soil fertility because K^+, Ca^{2+} and Mg^{2+} contribute directly to plant nutrient supply[8]. Nitrogen application can alter the retention and mobility of these cations through several coupled processes. Ammonium can compete with base cations for exchange sites whereas nitrification generates nitrate and protons; NO_3^- can promote accompanying cation leaching and H^+ further acidifies the soil[9]. Additionally, NO_3^- generated during the nitrification of NH_4^+ combines with exchangeable base cations and facilitates their leaching, while the concurrently produced H^+ contributes to soil acidification. Therefore, excessive N application leads to leaching of NO_3^- and exchangeable base cations, resulting in rapid accumulation of adsorbed H^+ on soil particle surfaces and continuous reduction of adsorbed exchangeable base cations such as K^+, Ca^{2+}, and Mg^{2+}. This negatively impacts tea plant growth and tea production.

Soil aggregates, as the fundamental units of soil structure, serve as critically important components of soil granular formation[10]. Stable aggregates can reduce soil erosion, conserve water and retain nutrients, serving as the material

foundation for forming good soil structure. They influence the dynamic changes of soil quality and soil health[11]. The response of soil aggregates varies under different nitrogen conditions. N application significantly increases the content of macroaggregates as well as the mean weight and diameter of aggregates, whereas high nitrogen has an inhibitory effect[12]. Excessive N application accelerates the mineralization of organic matter, leading to decreased soil organic matter content. This subsequently inhibits the formation of macroaggregates and reduces the stability of soil aggregates[13]. Based on differences in their formation processes, soil aggregates can be classified into microaggregates (< 0.25 mm) and macroaggregates (> 0.25 mm). The formation mechanisms and functions of aggregates vary with their particle size[14]. Exchangeable cations, including K^+, Na^+, Ca^{2+}, and Mg^{2+}, can participate in cation bridging between negatively charged organic functional groups and mineral surfaces, thereby contributing to the formation and stabilization of smaller aggregates[15,16]. These units can subsequently be bound into larger aggregates by roots, fungal hyphae, and plant- or microbially derived polysaccharides[17]. Given that aggregate fractions differ in their formation pathways, pore structure and nutrient-retention capacity, the abundance and distribution of exchangeable base cations within aggregates may be critical determinants of aggregate stability and soil fertility[18].

The leaching and redistribution of exchangeable base cations under excessive N application have been widely documented. However, most studies have assessed these cations in bulk soil, potentially obscuring heterogeneity among aggregate-size fractions. Such heterogeneity may be particularly important because aggregate fractions differ in their roles in nutrient protection, root-soil interactions and structural stability. The stability of soil aggregates is closely linked to cation exchange capacity and the retention of exchangeable base cations, while the abundance and distribution of these cations can, in turn, constrain aggregate stability and soil fertility[19]. Therefore, investigating the response and changes of exchangeable base cations to different N application rates at the scale of soil aggregates with varying particle sizes can provide insights into the impact of excessive nitrogen fertilizer on soil fertility in tea plantations, and reveal the underlying mechanisms behind the decline in tea yield and quality caused by excessive fertilizer application. To address this gap, this study employed an aggregate grading methodology instead of relying on traditional whole-soil analysis. This experimental design enabled us to elucidate the differential responses of exchangeable basic ions to N application across aggregates of varying particle sizes and to assess whether basic ion depletion at the aggregate scale provides a more sensitive indicator of soil fertility decline than measurements taken from whole-soil samples. Specifically, the present study aims to investigate (1) the effects of different N application rates on the distribution and stability of soil aggregates in tea plantations; (2) the concentration of exchangeable base cations in aggregates under varying N application levels; and (3) the relationships between exchangeable base cations and the tea yield and quality resulting from nitrogen fertilizer application.

We hypothesized that the response of tea productivity to N application is non-linear and reflects the balance between two opposing processes. At low-to-moderate N inputs, improved nutrient supply and greater organic inputs may favor macroaggregate formation and a more stable soil physical environment. In contrast, excessive N supply is expected to intensify soil acidification and deplete exchangeable base cations, particularly Ca^{2+} and Mg^{2+}, thereby weakening cation-mediated aggregation and reducing structural stability.

2 Materials and methods

2.1 Experimental site

The long-term positioning experiment was initiated in 2017 at a tea plantation (23°15′ N, 118°14′ E) in Anxi County, Quanzhou City, Fujian Province, China. The site experiences a subtropical monsoon climate, with an annual average temperature of 22.4 °C, total annual sunshine duration of 2030 h, annual rainfall of 1600 mm, and an annual frost-free period of 350 days. The soil type is classified as red soil. At the beginning of the experiment, the soil properties were as follows: pH 4.2, TAN content 104 $mg \cdot kg^{-1}$, available phosphorus content 54.7 $mg \cdot kg^{-1}$, available potassium content 92.3 $mg \cdot kg^{-1}$, exchangeable calcium content 109 $mg \cdot kg^{-1}$ and exchangeable magnesium content 14.7 $mg \cdot kg^{-1}$. The tested tea cultivar was 12-year-old Tieguanyin, cultivated under no-till conditions with a planting density set at 80 cm row spacing and 35 cm plant spacing.

2.2 Experimental design

The positioning experiment included five treatments with

different N application rates: (1) N0, 0 kg·ha^{-1} N; (2) N150, 150 kg·ha^{-1} N; (3) N300, 300 kg·ha^{-1} N; (4) N450, 450 kg·ha^{-1} N; and (5) N600, 600 kg·ha^{-1} N. The N fertilizer is ordinary urea, the phosphorus fertilizer is calcium superphosphate (44 kg·ha^{-1} P), and the potassium fertilizer is potassium sulfate (104 kg·ha^{-1} K). Based on the growth cycle and picking rounds of tea plants, fertilizers were applied three times per year: 30% as base fertilizer (late December), 20% as spring tea topdressing (mid-March) and 50% as autumn tea topdressing (late August). Each time, fertilizers were evenly broadcast below the canopy along the vertical edge. The experiment was laid out as a randomized block design with each treatment replicated four times, resulting in a total of 20 plots, each covering an area of 20 m^2.

2.3 Measurement items and methods

2.3.1 Tea yield and quality measurement

Tea samples were collected in May 2023. Four 0.1 m^2 sampling points were randomly established in each plot, and the number of tea buds within 0.1 m^2 was collected and calculated according to oolong tea harvesting standards. Free amino acids, tea polyphenols, and water extract contents were quantified in accordance with the respective Chinese national standards, using ninhydrin colorimetry, the furanol colorimetric method, and the gravimetric method respectively.

2.3.2 Soil sample collection and aggregate sieving

Soil samples were collected using a three-point sampling method: at each treatment plot, points were randomly selected about 10 cm below the tea tree roots beneath the canopy between rows, avoiding fertilizer application trenches and root systems. Stainless steel root drills were used to collect intact soil samples from two depths (0–20 and 20–40 cm) at each location, with this procedure repeated three times. The soils from all three points within the same layer were mixed to obtain the final soil sample for that treatment. Approximately 50 g of the collected intact soil was immediately stored in a 4 °C refrigerator for determination of ammonium nitrogen (NH_4^+-N) and nitrate nitrogen (NO_3^--N); another portion was air-dried and subjected to aggregate sieving[20].

For soil aggregate grading, 250 g of undisturbed soil sample was placed on the top layer of a nested sieve set with apertures of 2, 1 and 0.25 mm. Using an aggregate analyzer at a frequency of 40 times per minute for 20 min, the aggregates on each sieve were rinsed into aluminum boxes to obtain four fractions: macroaggregates (> 2 mm), medium aggregates (1–2 mm), microaggregates (0.25–1 mm), and silt+clay fractions (< 0.25 mm)[21]. The aggregates from each fraction were oven-dried at 80 °C and weighed to calculate the mass proportion of each aggregate class. To compare the stability of soil aggregates, the mean weight diameter (MWD) was estimated using Kemper's method as:

$$\mathrm{MWD} = \sum_{i=1}^{4} X_i \times M_i \quad (1)$$

where, X_i is the mean diameter of the i-th aggregate fraction and M_i is the mass percentage of the i-th aggregate fraction.

2.3.3 Soil physicochemical properties analysis

The physicochemical properties of soil aggregates at different levels were determined following the methods outlined by Bao[22]. Soil pH was measured using the potentiometric method (with a soil-to-water ratio of 2.5:1). Soil TAN was determined using the alkaline hydrolysis diffusion method. Soil ammonium and nitrate were extracted with KCl and measured using a continuous flow injection autoanalyzer. Soil exchangeable base cations (ex-K^+, ex-Na^+, ex-Ca^{2+}, ex-Mg^{2+}) were extracted with CH_3COONH_4 and determined by ICP-OES.

2.4 Data analysis

All data in this study were analyzed using Excel 2019 and IBM SPSS 22, with multiple comparisons assessed for significance using Duncan's test. Based on relevant agronomic theories concerning nitrogen cycling, leaching of exchangeable alkaline cations and tea crop productivity, a covariance-based structural equation model was implemented using the R package "lavaan" to quantify the direct, indirect and total effects of each factor.

3 Results and analysis

3.1 Effects of N application rates on soil aggregate composition and stability

As shown in Table 1, tea plantation soil aggregates were predominantly composed of microaggregates (0.25–1 mm), with average mass proportions of 44.7% in the surface layer

Table 1 Soil aggregate mass percentage and mean weight diameter under different nitrogen application rates

Soil depth (cm)	Treatment	Soil aggregates mass percentage (%)				MWD (mm)
		> 2 mm	1–2 mm	0.25–1 mm	< 0.25 mm	
0–20	N0	15.6 ± 3.11c	12.0 ± 1.92a	48.8 ± 5.14a	23.6 ± 3.96ab	1.0 ± 0.02b
	N150	22.6 ± 2.47a	17.5 ± 5.10a	47.4 ± 3.79a	12.5 ± 3.25c	1.2 ± 0.02a
	N300	20.0 ± 0.62ab	16.7 ± 5.69a	45.1 ± 3.22ab	18.3 ± 2.96bc	1.1 ± 0.06a
	N450	19.1 ± 1.38abc	13.7 ± 1.64a	39.5 ± 3.54b	27.7 ± 4.96a	1.0 ± 0.05b
	N600	17.2 ± 1.36bc	13.3 ± 1.93a	42.5 ± 2.65ab	27.0 ± 3.5a	1.0 ± 0.04b
20–40	N0	16.8 ± 1.15ab	15.6 ± 1.14b	44.2 ± 2.85a	23.5 ± 4.02a	0.7 ± 0.02a
	N150	13.7 ± 10.17ab	12.7 ± 2.65b	45.3 ± 11.97a	28.3 ± 9.43a	0.7 ± 0.08a
	N300	14.9 ± 9.89ab	14.8 ± 8.14b	42.3 ± 9.63a	28.0 ± 7.36a	0.7 ± 0.04a
	N450	18.7 ± 5.66a	12.0 ± 1.17b	38.3 ± 6.57a	31.0 ± 2.03a	0.6 ± 0.06a
	N600	3.7 ± 0.09b	25.5 ± 3.56a	33.3 ± 2.4a	37.5 ± 1.02b	0.7 ± 0.04a

Note: Means within columns followed by the same letter are not significantly different at the 0.05 level between treatments.

(0–20 cm) and 40.7% in the subsurface layer (20–40 cm). N application significantly affected the stability of surface-soil aggregates. Compared to the N0 treatment, the mass percentage of macroaggregates in the surface soil increased significantly by 44.5%, 28.1%, 22.4%, and 10.4% under the N150 to N600 treatments, respectively. Meanwhile, the proportion of microaggregates decreased significantly by 47.0% and 22.7% under the N150 and N300 treatments. The MWD of surface soil had an initial increase followed by a decrease with rising N application rates, reaching its maximum value under the N150 treatment, which represented an 18.3% increase compared to the N0 treatment. In the subsurface soil layer, the proportion of medium aggregates under the N600 treatment increased significantly compared to other treatments, whereas the proportions of macroaggregates and microaggregates decreased significantly. Apart from this, the differences in soil aggregate mass percentages and MWD among other treatments were not significant. N application rates did not affect the stability of aggregates in the subsurface soil layer.

3.2 Effects of N application on total alkali-hydrolyzable-N and total carbon content in soil aggregates

Total nitrogen (TAN) and total carbon (TC) were predominantly enriched in macroaggregates and medium aggregates. The TAN and TC contents in soil microaggregates were significantly lower than those in other aggregate fractions (Figs. 1 and 2). The TAN and TC contents in aggregates of all size classes within both surface and subsurface layers increased significantly with rising N application rates. In surface soil, the macroaggregates showed the greatest increase in TAN content, with N150 to N600 treatments rising by 43.1%, 45.8%, 61.4% and 89.4% compared to N0. Conversely, microaggregates had the highest increase in TC content, with N150 to N600 treatments increasing by 40.4%, 56.3%, 69.8% and 94.7% relative to N0. In the subsurface soil layer, macroaggregates had the most pronounced increases in both TAN and TC content. Compared to the N0 treatment, the TAN content under N150 to N600 treatments increased by 28.1%, 41.2%, 54.8% and 75.3%, respectively, whereas the TC content rose by 27.4%, 61.6%, 67.8% and 82.4%, respectively.

3.3 Effects of N application on soil pH, and ammonium- and nitrate-nitrogen contents

As shown in Fig. 3, soil pH decreased significantly with increasing N application rates. In the surface soil, the pH under the N150 treatment showed no significant difference compared to N0, but was significantly higher than that under N300 to N600 treatments in both surface and subsurface soil layers. Also, increased N application rates significantly elevated nitrate and NH_4^+-N content in both surface and subsurface soil layers. Specifically, compared to the N0 treatment, NO_3^--N content in the surface soil under N150 to N600 treatments increased by 59%, 126%, 343% and 614%, respectively, whereas in the

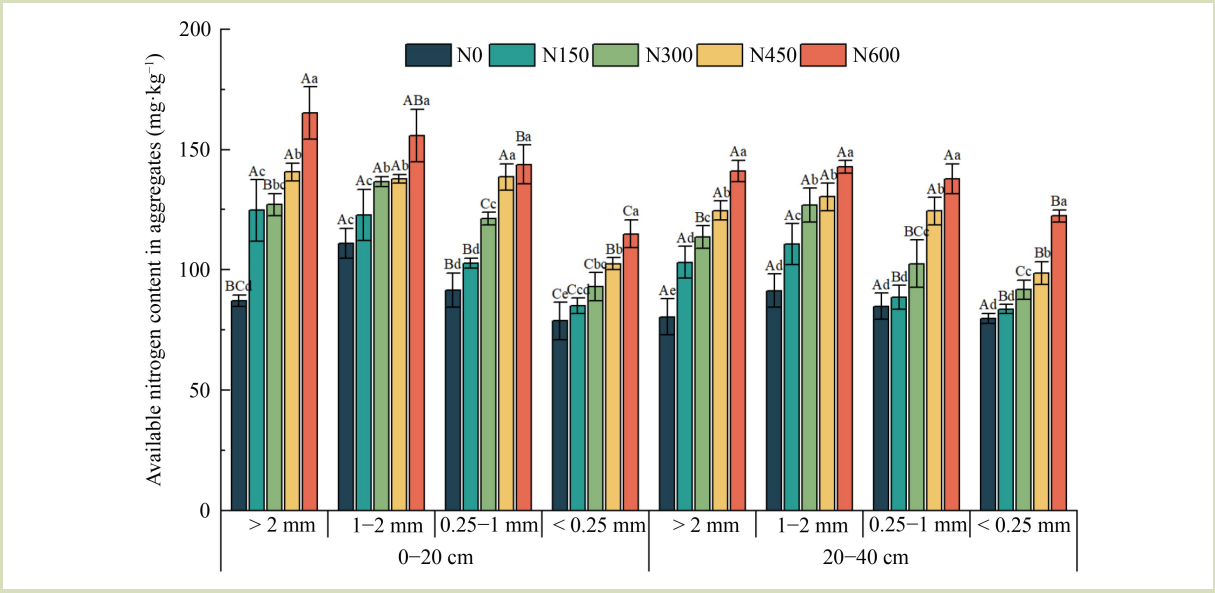


Fig. 1 Content of available nitrogen in soil aggregates under different nitrogen application rates. Means with the same uppercase letters are not significantly different at the 0.05 level between soil fractions and the same lowercase letters are not significantly different at the 0.05 level between treatments.

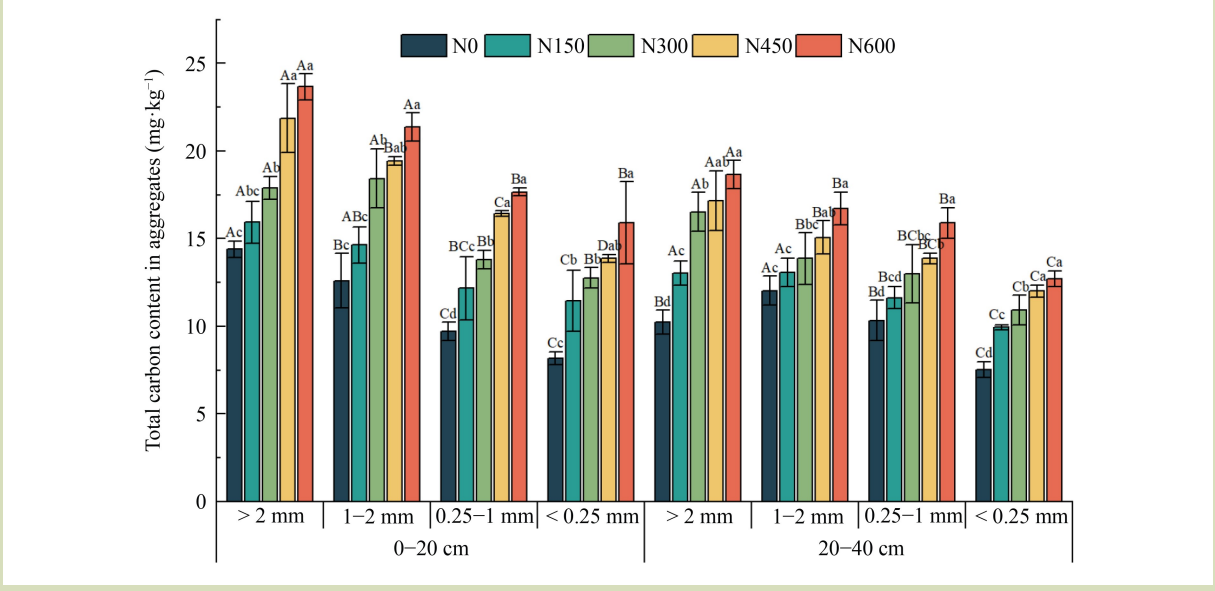


Fig. 2 Total carbon content of soil aggregates under different nitrogen application rates. Means with the same uppercase letters are not significantly different at the 0.05 level between soil fractions and the same lowercase letters are not significantly different at the 0.05 level between treatments.

subsurface soil it rose by 39%, 147%, 423% and 726%. Similarly, NH_4^+-N content increased by 81%, 105%, 173% and 254% in the surface soil, and by 52%, 102%, 200% and 280% in the subsurface soil under the same treatments.

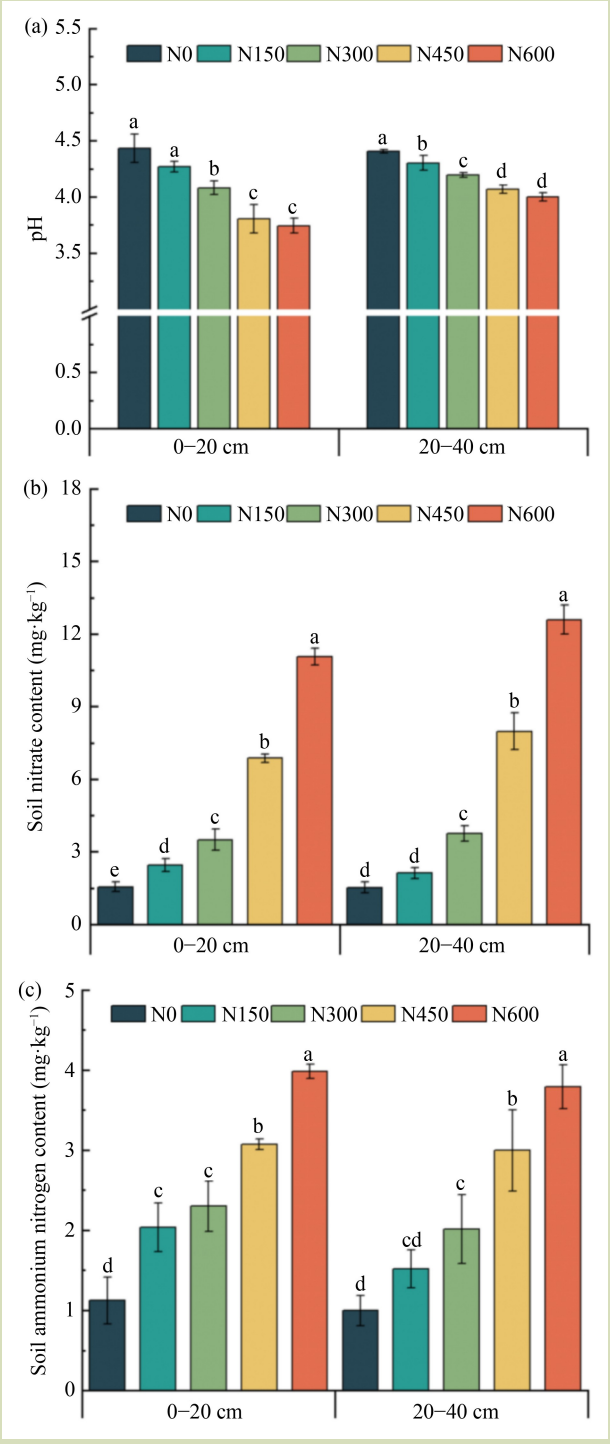


Fig. 3 pH (a), soil nitrate (b), and soil ammonium nitrogen (c) contents under different nitrogen application rates. Means with the same lowercase letters are not significantly different at the 0.05 level between treatments.

3.4 Effects of N application on exchangeable base-cation content in soil aggregates

As shown in Fig. 4, the content of exchangeable base cations (ex-K^+, ex-Na^+, ex-Ca^{2+}, ex-Mg^{2+}) in soil aggregates had the following trend: macroaggregates > medium aggregates > small aggregates > microaggregates. With increasing N application rate, the content of ex-K^+, ex-Ca^{2+} and ex-Mg^{2+} in aggregates of all size classes decreased consistently in both surface and subsurface soil layers. In surface soil, the content of ex-K^+, ex-Ca^{2+} and ex-Mg^{2+} in aggregates of all size classes under N0 and N150 treatments was significantly higher than that under N300 to N600 treatments. Under the N150 treatment, the content of ex-K^+, ex-Ca^{2+} and ex-Mg^{2+} in aggregates of all size classes was 1.3–1.7, 2.3–3.1 and 2.1–5.2 times higher than that under the N600 treatment, respectively, with the most pronounced differences observed in macroaggregates. ex-Na^+ content under the N0 treatment was significantly higher than that under other treatments only in medium aggregates and microaggregates. In the subsurface soil layer, the content of ex-K^+, ex-Ca^{2+} and ex-Mg^{2+} followed trends similar to those in the surface soil. However, in small aggregates and microaggregates, the ex-Na^+ content under the N150 treatment was significantly higher than that under other treatments, measuring 2.3 and 1.9 times that of the N600 treatment, respectively.

3.5 Effects of N application rates on tea yield and quality

Tea yield and quality had significant differences under varying N application rates (Tables 2 and 3). As N application increased, tea yield, bud density, and hundred-bud weight initially rose and then declined. The highest tea yield and optimal quality were observed under the N150 treatment. Compared to the N0 treatment, the N150 treatment showed increases of 37.5% in yield, 32.3% in bud density, and 44.1% in hundred-bud weight, whereas the N600 treatment had reductions of 6.7%, 12.3%, and 15.3% in these respective parameters.

The content of water extracts, tea polyphenols, and free amino acids in tea also had an initial increase followed by a decrease with rising N application rates. Among these, the N150 treatment gave the highest tea quality, with values 1.1, 1.2 and 1.3 times those of the N0 treatment, respectively. The phenol-ammonia ratio under the N150 and N300 treatments was significantly lower than that under other treatments, with reductions of 9.9% and 9.6%, respectively, compared to the N0 treatment.

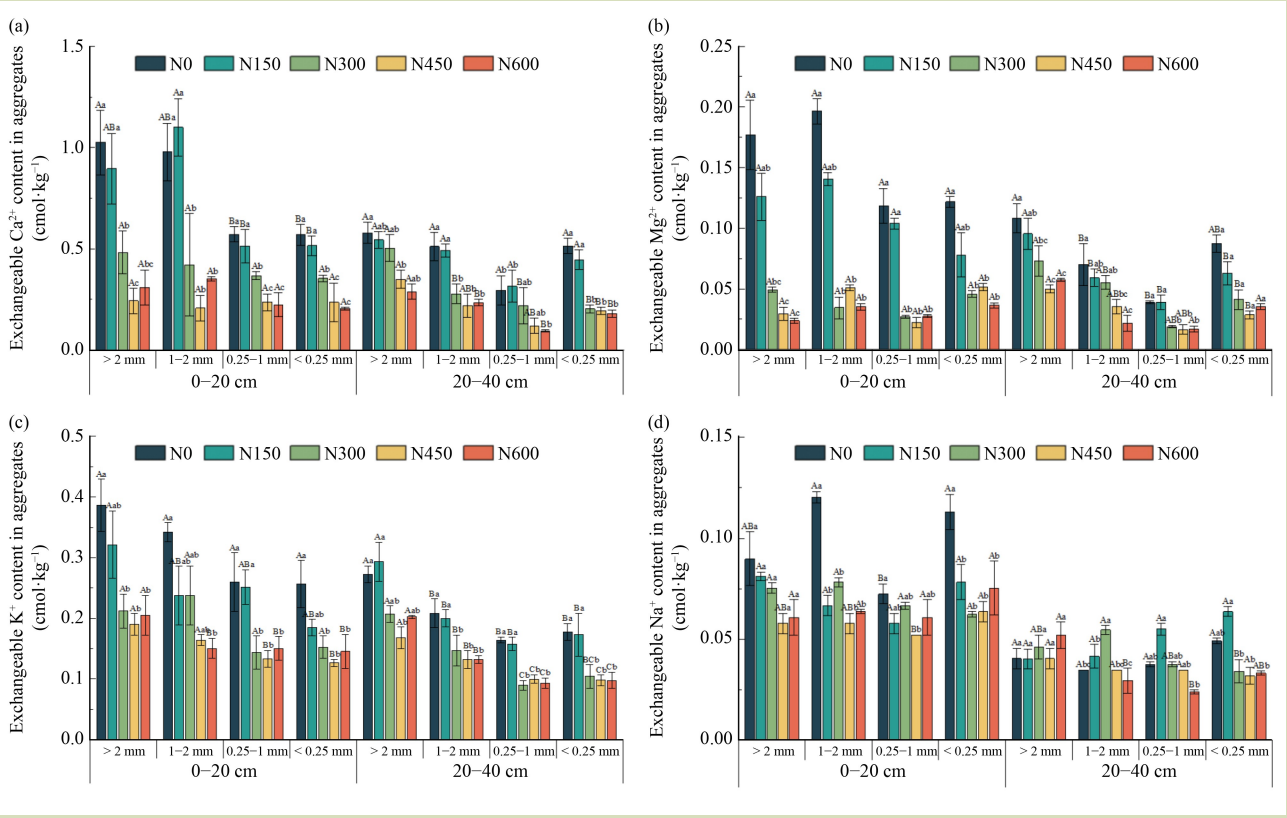


Fig. 4 Exchangeable Ca^{2+} (a), Mg^{2+} (b), K^{+} (c), and Na^{+} (d) content of soil aggregates under different nitrogen application rates. Means with the same uppercase letters are not significantly different at the 0.05 level between soil fractions and the same lowercase letters are not significantly different at the 0.05 level between treatments.

Table 2 Tea yield and agronomic characteristics under different nitrogen application rates

Treatment	Yield (t·ha^{-1})	Bud density (bud·m^{-2})	100-bud weight (g)
N0	10.6 ± 0.17d	92.0 ± 3.61c	137 ± 11.97c
N150	14.5 ± 0.51a	121 ± 6.43a	198 ± 1.81a
N300	12.6 ± 0.36b	103 ± 2.08b	156 ± 7.16b
N450	11.5 ± 0.41c	91.7 ± 3.06c	139 ± 2.99c
N600	9.9 ± 0.62d	80.7 ± 4.16d	116 ± 6.00d

Note: Means within columns followed by the same letter are not significantly different at the 0.05 level between treatments.

3.6 Relationship between exchangeable base cations content in soil aggregates and tea yield under excessive fertilizer application

To investigate the relationship between exchangeable base cations content and tea yield under excessive N application, correlation analysis was conducted on various indicators from N150 to N600 treatments, and path analysis was performed using covariance-based structural equation modeling. The results indicated significant positive correlations among yield, MWD, pH and the total base cation content (sum of

Table 3 Tea quality indexes under different nitrogen application rates

Treatment	Tea extract (%)	Tea polyphenol (mg·g^{-1})	Free amino acid (mg·g^{-1})	Phenol-ammonia ratio
N0	32.7 ± 2.15c	88.3 ± 7.04b	11.2 ± 0.77b	7.91 ± 0.33a
N150	39.3 ± 1.54a	101 ± 2.58a	14.2 ± 0.52a	7.13 ± 0.14b
N300	36.8 ± 0.92ab	96.8 ± 2.28ab	13.6 ± 0.41a	7.15 ± 0.09b
N450	34.7 ± 2.11bc	86.7 ± 1.15b	11.4 ± 0.6b	7.62 ± 0.3a
N600	34.5 ± 2.46bc	83.5 ± 4.46b	11.1 ± 0.42b	7.57 ± 0.19a

Note: Means within columns followed by the same letter are not significantly different at the 0.05 level between treatments.

exchangeable K^+, Na^+, Ca^{2+}, and Mg^{2+}) across all aggregate-size classes. These parameters showed significant negative correlations with NH_4^+-N and NO_3^--N content (Table 4).

As shown in Fig. 5, the path analysis explained 99% of the variation in tea yield. This exceptionally high coefficient of determination stems from the fact that this experiment was a nitrogen gradient positioning study with uniform cultivation management, and interfering variables such as climate, tea cultivar and moisture were kept consistent. Increased N application rate and soil NH_4^+-N content had direct positive effects on NO_3^--N content (path coefficients of 0.28 and 0.91, respectively). In contrast, increased NH_4^+-N and NO_3^--N content had direct negative effects on soil pH and macroaggregate TBC (path coefficients of –0.72 and –0.61, and –0.74 and –0.72, respectively). Soil pH directly influenced soil aggregate stability, macroaggregate TBC and tea yield (path coefficients of 0.84, 0.23 and 0.30, respectively). Macroaggregate TBC directly affected aggregate stability (path coefficient of 0.76) and indirectly improved tea yield (path coefficient of 0.24).

4 Discussion

4.1 Effects of long-term N application on the distribution and stability of soil aggregates

Soil aggregates are fundamental structural units that regulate soil stability and nutrient retention[19]. Multivalent cations and humified organic matter contribute to microaggregate formation, whereas roots, fungal hyphae, and plant- and microbially- derived polysaccharides bind smaller units into macroaggregates[23]. Transformations among aggregate-size classes therefore influence nutrient cycling and the physical protection of soil organic matter[18]. Stable aggregation can also

Table 4 Correlation coefficients of soil indexes under different nitrogen application rates

	Yield	MWD	pH	NH_4^+-N	NO_3^--N	Lar-TBC	Med-TBC	Sma-TBC	Mic-TBC
Yield	1								
MWD	0.792**	1							
pH	0.924**	0.772**	1						
NH_4^+-N	–0.947**	–0.755**	–0.896**	1					
NO_3^--N	–0.940**	–0.783**	–0.889**	0.983**	1				
Lar-TBC	0.792**	0.690*	0.786**	–0.691*	–0.612*	1			
Med-TBC	0.835**	0.708**	0.833**	–0.684*	–0.634*	0.954**	1		
Sma-TBC	0.897**	0.732**	0.895**	–0.780**	–0.732**	0.949**	0.974**	1	
Mic-TBC	0.941**	0.730**	0.913**	–0.828**	–0.796**	0.892**	0.948**	0.980**	1

Note: MWD, mean weight diameter; NH_4^+-N, ammonium-N content; NO_3^--N, nitrate-N content; Lar-TBC, total base-cation content in macroaggregates; Med-TBC, total base-cation content in medium aggregates; Sma-TBC, total base-cation content in small aggregates; Mie-TBC, total base-cation content in microaggregates. * and ** indicate significant correlations at $P < 0.05$ and $P < 0.01$ levels, respectively (two-tailed test).

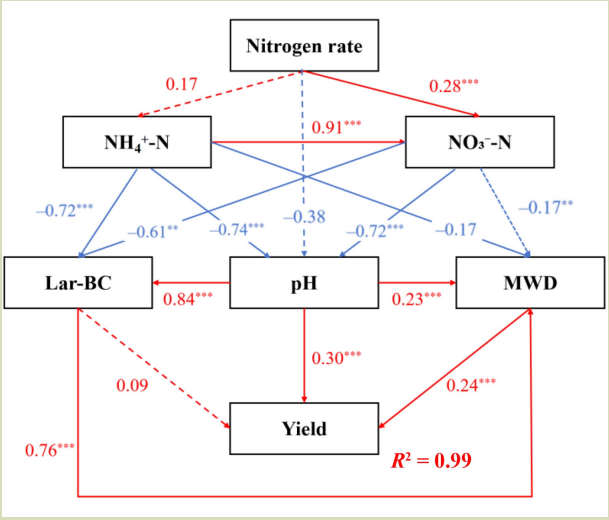


Fig. 5 Structural model of the effect of excessive nitrogen fertilizer application on exchangeable base cations content and tea yield. Solid lines indicate significant pathways, dashed lines indicate non-significant pathways; red arrows represent positive effects, blue arrows represent negative effects; values on lines are standardized path coefficients; * $P < 0.05$; ** $P < 0.01$; *** $P < 0.001$. Model goodness-of-fit statistics: χ^2/df = 1.24, RMSEA = 0.042, CFI = 0.978, TLI = 0.965, SRMR = 0.031. The coefficient of determination R^2 = 0.99 for tea yield means 99% of the total variation in tea yield can be explained by the constructed model.

reduce erosion and nutrient loss while enhancing the capacity of soil to retain applied fertilizers[15]. MWD and the proportion of water-stable aggregates > 0.25 mm are commonly used indicators of aggregate stability[24]. In the present study, surface-soil MWD responded non-linearly to N input and peaked at N150; when N application exceeded 150 kg·ha^{-1}·yr^{-1} N, both the proportion of >2 mm aggregates and MWD declined as N rate increased. This pattern supports previous observations that excessive N inputs can impair aggregate stability[12,17]. Moderate N addition may enhance organic inputs through greater root activity and litter turnover, thereby promoting the formation of larger aggregates[25]. In contrast, high N inputs can alter litter decomposition and accelerate organic matter mineralization, reducing the organic binding agents that stabilize macroaggregates[12,13].

4.2 Effects of long-term N application on C and N content in soil aggregates

Soil organic carbon is a central indicator of soil fertility and quality[14]. Research has indicated that nitrogen addition increases soil carbon input through two primary mechanisms: first, by enhancing plant biomass production, promoting growth and accumulating litter; and second, by stimulating the growth of microbial communities, improving CUE and NUE, and facilitating the accumulation of microbial residues[26], thereby elevating soil carbon content. The results of this study demonstrate that, compared to the no-nitrogen treatment, the TC content in aggregates of all size classes increased with rising N application rates. These findings align with the results of previous studies[12,14]. Our results also indicate that under N application, the TC content in aggregates increased with larger aggregate particle sizes (Fig. 2). A greater proportion of soil TC was enriched in macroaggregates (> 0.25 mm), which may be due to the fact that macroaggregates have critical sites for carbon sequestration in the soil and are important in soil carbon pool[26]. Plant-derived residues incorporated into macroaggregates can be transformed into particulate organic matter and subsequently protected through interactions with minerals and microbial products[27]. Previous work has similarly reported greater organic C concentrations in macroaggregates than in smaller fractions under long-term N input[25]. Thus, the preferential enrichment of TC in larger aggregates observed in the present study is consistent with the hierarchical organization of soil structure and the stronger physical protection afforded by larger aggregate units.

The distribution of TAN among aggregate fractions broadly paralleled that of TC (Figs 1 and 2). TAN increased significantly with N application across all aggregate-size classes relative to N0. This coupling is consistent with the close association between soil C and N pools[28], because a large proportion of soil N occurs in organic forms associated with amino compounds, proteins and humified organic matter[25]. Nitrogen addition may therefore increase both C and N inputs through enhanced plant production, litter return, and root-derived inputs. The greater TAN concentrations in larger aggregates are also consistent with their higher organic matter content and stronger capacity to retain organic substrates. Similar responses have been reported in other long-term N application studies, in which N enrichment increased N storage preferentially in larger aggregate fractions[27].

4.3 Effects of long-term N application on exchangeable base-cation content in soil aggregates

Previous work in tea-plantation soils has found that exchangeable base cations (ex-K^+, ex-Na^+, ex-Ca^{2+} and ex-

Mg^{2+}) are more concentrated in larger than in smaller aggregate fractions[29] Our results showed the same general pattern, with the highest base-cation concentrations occurring in the > 2 mm and 1–2 mm fractions. More importantly, once N application exceeded 150 kg·ha^{-1}·yr^{-1} N, exchangeable base-cation concentrations declined markedly across aggregate fractions, indicating that long-term N input altered not only bulk-soil chemistry but also the within-aggregate distribution of nutrient cations. This response is mechanistically consistent with acidification under long-term urea application[30]. Soil colloids retain cations through negatively charged surface sites whose abundance is related to mineral surfaces and organic functional groups[14,17]. During nitrification, NH_4^+ is converted to NO_3^- and H^+ is released; increasing H^+ activity can displace base cations from exchange sites, and NO_3^- can facilitate their downward leaching[29]. The progressive decline in ex-K^+, ex-Ca^{2+} and ex-Mg^{2+} from N150 to N600 therefore indicates substantial base-cation depletion under excessive N input. Across treatments, divalent Ca^{2+} and Mg^{2+} remained more abundant than monovalent K^+ and Na^+, consistent with the stronger electrostatic association of divalent cations with negatively charged soil surfaces[31].

The contents of four exchangeable cations ranked as ex-Ca^{2+} > ex-Mg^{2+} > ex-K^+ > ex-Na^+. Monovalent cations disperse soil aggregates, among which Na^+ acts as a powerful dispersant to destroy aggregate structure directly[32]. In contrast, Ca^{2+} and Mg^{2+} can promote flocculation and cation bridging, whereas K^+ contributes to plant nutrition and can also influence particle interactions. Thus, the preferential loss of Ca^{2+} and Mg^{2+} under high N input is potentially important not only for nutrient supply but also for maintaining aggregate cohesion. Excessive accumulation of Na^+ is undesirable for soil structure and crop productivity, whereas the retention of Ca^{2+} and Mg^{2+} is particularly important for sustaining the chemical and physical quality of acidic tea-garden soils[33].

During the 8-year experimental treatment period, the increased concentrations of exchangeable Ca^{2+} and Mg^{2+} under the N150 treatment condition indicate that the exchangeable cation pool was continuously restored. In contrast to other treatments, the persistent acidification weakened this buffering capacity, leading to a progressive and irreversible depletion of the exchangeable alkaline cation pool. Therefore, the elevated concentrations observed under the N150 condition do not represent a transient pulsed phenomenon, but rather reflect a steady-state condition achieved through improved soil structure and reduced acid stress.

4.4 Effects of long-term N application on tea yield and quality

The nitrogen-containing components in tea directly affect the aroma and taste of harvested tea[34]. Excessive nitrogen use reduces both tea quality and yield[35]. Tea polyphenols serve as an indicator of bitterness in tea, while free amino acids represent the sweet and mellow aroma, which are frequently used to evaluate tea quality. It was found that tea quality is negatively correlated with tea polyphenol content, and a lower phenol-ammonia ratio indicates higher tea quality[8]. The results of this study indicate that, compared to treatments without N fertilizer application, the phenol-ammonia ratio under N150 treatment gave the lowest value and the best tea quality (Table 3), which is consistent with previous research findings[36]. The decline in tea quality resulting from excessive N application may be attributed to the dilution effect caused by overly rapid tea plant growth[37]. An antagonistic relationship exists between secondary metabolites of carbon and nitrogen metabolism in tea leaves[38].The increased tea yield and quality under N150 can be attributed to a combination of factors, including enhanced aggregate stability, improved nutrient supply and reduced acidification stress.

4.5 Regulatory pathways of soil acidification under long-term N application

In this experiment, correlation analysis revealed a significant positive relationship between soil pH and exchangeable base cations, while a significant negative correlation was observed with exchangeable acidity, TAN and TC. Structural equation modeling further confirmed that exchangeable base cations had a direct positive effect on soil pH and a direct negative effect on TAN. These findings align with previous studies[39,40], indicating that nitrogen saturation may be a key driver of soil acidification[41]. This is primarily because nitrogen saturation depletes exchangeable base cations content in soil aggregates, reducing soil-buffering capacity and promoting a shift toward iron and aluminum dominance, thereby exacerbating soil acidification. This acid-driven salinity depletion directly disrupts the cationic bridge bonds that maintain the stability of macroaggregates[6]. Once these bridge ions are extensively displaced or leached, the cohesive strength between microaggregates weakens, leading to the disintegration of large aggregates into microaggregates (Table 1) and a significant decrease in MWD. The collapse of the aggregate structure means that even under high-nitrogen conditions, these nutrients are highly susceptible to mineralization and loss due

to the absence of physical protection provided by large aggregates, rendering them unavailable for effective utilization by tea plants.

5 Conclusions

This 8-year-long experimental study found that N150 treatment of tea plantation soil increased the content of exchangeable alkali metal cations, which was attributed to enhanced aggregate stability and reduced soil acidification. Under long-term low nitrogen treatment (N150), the content of soil macroaggregates increased, thereby enhancing soil stability. In contrast, high-nitrogen treatments (> N150) with all soil layers dominated by microaggregates (0.25–1 mm). Simultaneously, prolonged N application elevated the soil carbon and nitrogen content across aggregate fractions in both soil layers, while reducing soil pH, the total exchangeable base cations, and the concentrations of exchangeable calcium and magnesium ions. Soil TAN has a direct negative impact on the exchangeable base cations content of large aggregates and soil pH, and soil pH has a significant positive effect on tea yield. The saline ion content of large aggregates further positively influences tea yield by affecting aggregate stability. Under varying N fertilizer application rates, the tea quality and yield were highest under low nitrogen treatment (N150). Therefore, rational regulation of nitrogen fertilizer input can enhance soil stability and fertility, mitigate soil acidification, and ultimately improve both the quality and yield of tea.

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Compliance with ethics guidelines

Xinhao Rao, Xinge Shi, Aolin Wu, Shuhao Liu, Lijin Guo, Liangquan Wu, and Wenhao Yang declare that they have no conflicts of interest or financial conflicts to disclose. This article does not contain any studies on human or animal subjects performed by any of the authors.

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