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Trade-Off Strategy for Usage of Phosphorus Fertilizer in Calcareous Soil-Grown Winter Wheat: Yield, Phosphorus Use Efficiency, and Zinc Nutrition Response

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Abstract: Although phosphorus (P) fertilizer application is widely used to improve production, irrational P application has a negative impact on the zinc (Zn) nutrition of cereal crops. Previous researchers observed and confirmed that P application decreases grain Zn concentrations and bioavailability in cereal crops. However, it remains unclear whether different P fertilizer types can alleviate the antagonism of P and Zn in the soil and grain and, thus, enhance the Zn nutritional level of cereal crops while maintaining production. Thus, a completely randomized pot experiment was conducted on winter wheat grown in two calcareous soils (lime concretion black soil and fluvo-aquic soil). Five P fertilizer types (single superphosphate, diammonium phosphate, fused calcium–magnesium phosphate, triple superphosphate, and ammonium polyphosphate, abbreviated, respectively, as SSP, DAP, FMP, TSP, and APP) were applied to each soil compared to no P fertilizer (CK). Plant and topsoil samples were collected during the flowering and maturity stages of winter wheat, and biomass, Zn concentrations in each organ, and grain phytic acid concentrations were analyzed. Grain yield was not affected by the application of different P fertilizer types to lime concretion black soil, while it was significantly increased by the application of TSP and APP to fluvo-aquic soil. The application of DAP and APP effectively promoted soil available Zn concentrations in both calcareous soils. In lime concretion black soil, the application of FMP significantly increased Zn remobilization to grains, while the application of DAP increased post-anthesis Zn uptake, thereby increasing grain Zn concentrations and its bioavailability. In fluvo-aquic soil, post-anthesis Zn remobilization and uptake were significantly increased by the application of TSP and APP, finally achieving higher grain Zn concentrations and Zn harvest index and effectively promoting grain Zn bioavailability. In conclusion, the rational application of DAP to wheat grown in lime concretion black soil and of TSP or APP to fluvo-aquic soil can achieve superior grain Zn nutrition quality while concurrently retaining high production and high P use efficiency, reducing micronutrient deficiency and further contributing to green agricultural development and human health.

Keywords: calcareous soil; P fertilizer types; soil available Zn; phytic acid; grain Zn concentration; Zn bioavailability

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1. Introduction

Zinc (Zn) is an indispensable micronutrient for plant and human health. It participates in the composition and activation of a variety of human enzymes, protein synthesis, cell growth, division, and proliferation, and hormone metabolism, promoting growth and development, improving taste, enhancing immunity, maintaining skin health, and protecting the eyes, among other aspects [1]. Human Zn nutrition is mainly obtained through staple dietary foods, especially in developing countries or other underdeveloped areas [2,3]. Wheat, one of the major grain crops in China, accounts for approximately 20% of the total sown area of grain crops, with a production of 137 million tons [4]. However, 51% of cultivated land is Zn-deficient, with Zn deficiency in calcareous soils being particularly prominent and, in turn, leading to 85.8% of wheat grain samples in major wheat-growing areas of China failing to meet the recommended Zn nutritional values (40–60 mg kg^{-1}) [5–7]. Therefore, it is very important to improve the grain Zn concentration of wheat for human health.

In modern intensive production, phosphorus (P) fertilizer plays an active role in the continuous improvement of crop yields. Many studies have shown that P fertilizer application can significantly increase wheat grain yield [8–10]. It is worth noting that P fertilizer, while increasing yield, also has a significant role in regulating processes such as the synthesis and accumulation of Zn and phytate in grains [11]. In both short-term and long-term experiments, cereal grain Zn concentrations decreased in a negative linear or linear plateau with increasing P fertilizer application rates [12–14]. It can be seen that optimizing P fertilizer dosage is an effective measure for grain Zn biofortification, but it is not clear how different P fertilizer types affect grain Zn nutrition due to the different physicochemical properties and application effects of the fertilizers. Therefore, clarifying the effects of different types of P fertilizers on grain yield and Zn concentrations can provide a theoretical basis for achieving three times as high a yield, P use efficiency (PUE), and Zn biofortification in wheat cropping systems.

Soil available Zn and crop Zn nutrition are different under different P fertilizers and soil types [8,13]. It has also been shown that compared with no P application, the application of diammonium phosphate (DAP) and ammonium polyphosphate (APP) can significantly increase the soil available Zn concentration in desert gray soil up to 33.3% and 72.6%, respectively [15]. In yellow loam soil and fluvo-aquic soil, the application of single superphosphate (SSP) significantly increased the available Zn concentration in rhizosphere soil and non-rhizosphere soil by 10.6% and 27.5%, respectively, compared with no P application [16,17]. The application of DAP had no significant effect on the soil available Zn concentration in black soil [18], whereas the results of a 37-year locational experiment conducted on brown soil showed that compared with no P application, the application of SSP reduced the soil available Zn concentration by up to 5.2% [19]. Meanwhile, there was no significant difference in the soil available Zn concentration between long-term triple superphosphate (TSP) treatment and no P application in calcareous soils [13]. Likewise, the results of studies on the effect of P fertilizer on wheat grain Zn concentrations are also complex and inconsistent. According to a global meta-analysis of wheat grain Zn concentrations, P application reduced grain Zn concentrations by 16.6%, and with a P increase rate of 10 kg P ha^{-1}, the corresponding decrease was 0.1 mg kg^{-1} [11,20]. The results of a long-term field trial on calcareous soil in North China showed that grain Zn concentrations had a negative linear plateau relationship with increases in the P application rate, and when the P application rate was less than 100 kg P ha^{-1}, the grain Zn concentration decreased by 3.77 mg kg^{-1} for every 10 kg P ha^{-1} increase in the P application rate [21]. Meanwhile, high soil Olsen-P and low DTPA-Zn concentrations were the main soil factors limiting the Zn nutrition of wheat in major wheat-producing areas of Iran and Serbia [22]. Studies have shown that the application of P fertilizer not only significantly reduced grain Zn concentrations by 33.0–39.1%, but also increased phytic acid (PA) by 11.1–25.4% and the PA/Zn molar ratio by 77.9–105.8%, resulting in a decrease in Zn bioavailability, which is not conducive to human health in terms of Zn nutrition [23]. These findings imply the implementation of rational strategies for managing P and maintaining optimal soil nutrient levels, particularly available P and Zn, is critical for enhancing crop Zn nutrition.

Current research on Zn nutritional fortification of wheat grain is focused on the application of Zn fertilizers, methods or rates of Zn application and the impact of nitrogen and

P fertilizer rates on Zn nutrients [6,24–27]. The application of different P fertilizer types to Zn-deficient or potential Zn-deficient calcareous soils on Zn nutrients for winter wheat cultivation has been rarely investigated, despite the importance of achieving high crop yields and nutrient use efficiency through tailored fertilizer management [10,28]. In this study, we conducted a pot experiment with winter wheat grown on two calcareous soils to elucidate the impact of various P fertilizer types on soil available Zn concentrations and shoot Zn nutrition. Our analysis encompassed wheat yield, soil available Zn, shoot Zn accumulation in wheat organs, grain Zn concentrations, and Zn bioavailability. This research aims to optimize both productivity and utilization efficiency of P fertilizers while providing a theoretical foundation for enhancing the nutritional quality of winter wheat grain.

2. Materials and Methods

2.1. Experimental Design

A winter wheat (*Triticum aestivum* L., cv. Fanmai 8) pot experiment started in October 2020 and was conducted within the plant nutrition observation field (117°25′ E, 31°87′ N) at Anhui Agricultural University. Two calcareous soils (lime concretion black soil and fluvo-aquic soil) with a 0–20 cm soil layer were extracted from the North Comprehensive Experimental Station of Anhui Agricultural University (117°09′ E, 33°68′ N) and Bengbu, Anhui Province, China (117°38′ E, 32°96′ N). Each soil was air-dried, sieved, and thoroughly mixed before the pot experiment. The soil physicochemical properties of the lime concretion black soil and fluvo-aquic soil before the experiment began were as follows: 25.43 and 11.65 g kg^{-1} of organic matter (treated using the potassium dichromate external heating method), 1.41 and 0.65 g kg^{-1} of total nitrogen (micro Kjeldahl method), 5.57 and 4.94 mg kg^{-1} of available P (0.5 mol L^{-1} $NaHCO_3$), 140.7 and 101.3 mg kg^{-1} of available K (1 mol L^{-1} NH_4OAc), 0.93 and 0.96 mg kg^{-1} of DTPA-Zn, (DTPA solution method) and soil pH values of 7.83 and 8.04 (distilled water without CO_2), respectively.

This completely randomized design experiment consisted of five P fertilizer treatments, namely, single superphosphate (SSP), diammonium phosphate (DAP), fused calcium magnesium phosphate (FMP), triple superphosphate (TSP), and ammonium polyphosphate (APP) treatments, which were compared to a no P fertilizer (CK) treatment, with six replications for each treatment. Each pot (height: 17 cm; diameter: 21 cm) contained 3.5 kg of air-dried soil, and the dosages of nitrogen (N), P, and potassium (K) were 0.40 g N kg^{-1}, 0.30 g P kg^{-1}, and 0.13 g K kg^{-1} of air-dried soil, respectively, of which 50% of the N fertilizer dosage and all of the P and K fertilizers were applied at one time as basal fertilizer before the sowing of the winter wheat, and the remaining 50% of the N fertilizers were applied at the jointing stage along with watering. Seeds of uniform size and full morphology were selected, soaked in a 10% H_2O_2 solution for 30 min to kill the surface bacteria, and then rinsed with deionized water. Fifteen seeds were sown in each pot, and thinning was performed when three leaves and eight plants were retained in each pot. The positions of the treatments were randomly arranged throughout the whole winter wheat growth period, and the cultivation of winter wheat during each growth period was consistent with the high-yield cultivation techniques used by local farmers.

2.2. Sampling and Nutrient Analysis

At the anthesis (GS 62) and maturity (GS 92) stages, the aboveground parts of wheat were collected. Three pots with uniform growth were randomly selected for each treatment, and the aboveground plant samples were collected in a mesh bag and brought back to the laboratory to be quickly rinsed three times with tap water and ultrapure water, respectively; also at the laboratory, the aboveground organs were divided into stem-leaves and ears at anthesis and into stem-leaves, glumes, and grains at maturity. Samples of each part (50–80 g) were packed into kraft paper sacks and placed in an oven preheated to 105 °C for 30 min to curb physiological metabolism and enzyme activity, and then lowered to 75 °C to dry the samples to a constant weight. The samples were then weighed and crushed with a stainless-steel grinder for later analysis. All aboveground organ samples were digested using the HNO_3-H_2O_2 method in a high-performance microwave digestion system (ETHOS UP, Milestone, Modena, Italy), and the Zn levels in the digestion solution were measured using an inductively coupled plasma optical emission spectrometer (ICP-OES, iCAP 7000, ThermoFisher, Boston, MA, USA). The phytic acid concentration of the grains was determined via bipyridine spectrophotometry [29,30] using a UV–visible spectrophotometer (Cary 3500, Agilent, Santa Clare, CA, USA).

Soil samples were collected in conjunction with shoot sample collection. The wheat root system was removed intact, and soil samples were collected to avoid residual roots. All soil samples were ground after natural air drying, passed through 20- and 100-mesh sieves, and sealed. The soil available Zn (DTPA-Zn) concentration was determined using a DTPA-$CaCl_2$-TEA system and analyzed using a flame graphite furnace atomic absorption spectrometer (PinAAcle900T, PerkinElmer, Boston, MA, USA) [31].

2.3. Data Calculations and Statistical Analysis

The calculations listed below refer to Kutman et al. (2011), Xue et al. (2012), and Hamnér et al. (2017) [32–34].

PUE (%) = [(shoot P accumulation under P fertilizer − shoot P accumulation under no P fertilizer)/P application rate] × 100 (1)

Zn accumulation (mg pot^{-1}) = shoot Zn concentration × shoot biomass (2)

Zn remobilization (mg pot^{-1}) = shoot Zn accumulation at anthesis − straw Zn accumulation at maturity (3)

Post-anthesis Zn uptake (mg pot^{-1}) = shoot Zn accumulation at maturity − shoot Zn accumulation at anthesis (4)

Zn harvest index (%) = grain Zn accumulation/shoot Zn accumulation at maturity × 100 (5)

Phytic/Zn molar ratio = (grain phytic concentration/phytic molecular weight)/(grain Zn concentration/Zn molecular weight) × 1000 (6)

In addition to the PA/Zn molar ratio, a three-variable Zn uptake model (TAZ, total daily absorbed Zn, mg Zn d^{-1}) was used to evaluate grain Zn bioavailability [35].

$$\mathrm{TAZ} = 0.5 \times 65 \times 100 \times \left\{ \mathrm{A_{MAX}} + \mathrm{TDZ} + \mathrm{k_R} \times \left(1 + \frac{\mathrm{TDP}}{\mathrm{k_P}}\right) - \sqrt{\mathrm{A_{MAX}} + \mathrm{TDZ} + \mathrm{k_R} \times \left(1 + \frac{\mathrm{TDP}}{\mathrm{k_P}}\right) - 4 \times \mathrm{A_{MAX}} \times \mathrm{TDZ}} \right\}$$

The total daily absorbed Zn (TAZ, mg Zn d^{-1}) was estimated based on the daily dietary intake of PA (TDP, mmol PA d^{-1}) and Zn (TDZ, mmol Zn d^{-1}). A_{MAX}, maximum Zn absorption coefficient, 0.091; K_R, equilibrium dissociation constant of Zn receptor binding reaction, 0.680; K_P, equilibrium dissociation constant of Zn–PA receptor binding reaction, 0.033. Adult daily consumption of 300 g of grain was used as the sole source [36].

Data were organized in Excel 2019 (Microsoft, Ithaca, NY, USA) and statistical analyses were performed using SPSS 25.0 (IBM Corporation, Chicago, IL, USA). One-way analysis of variance (ANOVA) was performed to assess the effects of different P fertilizer types on grain yield, Zn concentrations, Zn accumulation, and grain phytic acid concentrations. The means were compared using Tukey's least significant difference test (LSDT) at a 0.05 confidence level. Figures were constructed using Origin 2021 (OriginLab Corporation, Northampton, MA, USA).

3. Results

3.1. Yield, Phosphorus Use Efficiency, and Grain Zn Concentration

In the lime concretion black soil, the winter wheat grain yield of FMP was slightly increased by 12.75 g pot^{-1} compared with no P fertilizer and other P fertilizer treatments, but the P fertilizer types did not have a significant effect on grain yield (Figure 1). In the fluvo-aquic soil, all the P fertilizer treatments significantly increased wheat yield, with the application of TSP and APP being significantly higher than in the other P fertilizer treatments, increasing yields by 40.3% and 35.2%, respectively, compared with the no P fertilizer application treatment. In the two calcareous soils, the PUE of the applied TSP and APP was significantly higher than that of other P fertilizer types, with PUE values of 6.96% and 8.81% in lime concretion black soil and 10.63% and 10.66% in fluvo-aquic soil, respectively.

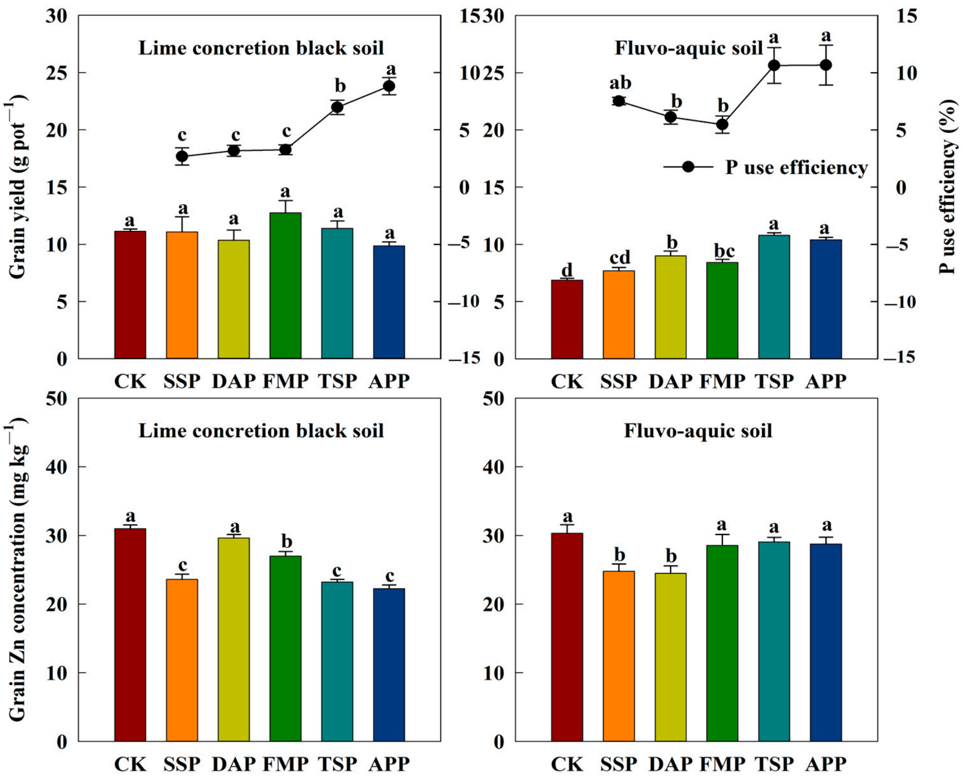

Figure 1. Grain yield, phosphorus use efficiency and grain Zn concentrations of winter wheat under different phosphorus fertilizer types in two calcareous soils. Different lowercase letters represent significantly different among phosphorus fertilizer types in the same soil type ($p < 0.05$). CK, SSP, DAP, FMP, TSP and APP are used to express no P fertilizer, single superphosphate, diammonium phosphate, fused calcium–magnesium phosphate, triple superphosphate and ammonium polyphosphate, respectively.

The application of P fertilizer reduced the grain Zn concentration of wheat grown in calcareous soil (Figure 1). The application of FMP, SSP, TSP, and APP to lime concretion black soil reduced grain Zn concentrations by 12.9%, 23.9%, 25.0%, and 28.2%, respectively, whereas the application of SSP and DAP to fluvo-aquic soil reduced grain Zn concentrations by 18.3% and 19.3%, respectively. There were no significant differences in grain Zn concentrations between the application of DAP to lime concretion black soil and that of FMP, TSP, and APP to fluvo-aquic soil, respectively, compared with the treatment without P fertilizer application.

3.2. Soil Available Zn Concentration

In the lime concretion black soil, there were no significant differences in soil available Zn concentrations among the treatments at the anthesis stage, whereas the soil available Zn concentration at the maturity stage was the highest in the APP treatment, significantly increasing by 30.2% and 15.0–43.8% compared with the no P fertilizer treatment and other P fertilizer types, respectively (Figure 2a,c). In the fluvo-aquic soil, the soil available Zn concentrations at the anthesis stage for the TSP treatment were significantly higher (26.0% and 20.2%) than those of the DAP and SSP treatments but not significantly different from those for the other P fertilizer types (Figure 2b). Soil available Zn concentrations at the maturity stage significantly increased by 23.9% for DAP compared with APP, but there was no significant difference between DAP and other P fertilizer types, except APP (Figure 2d).

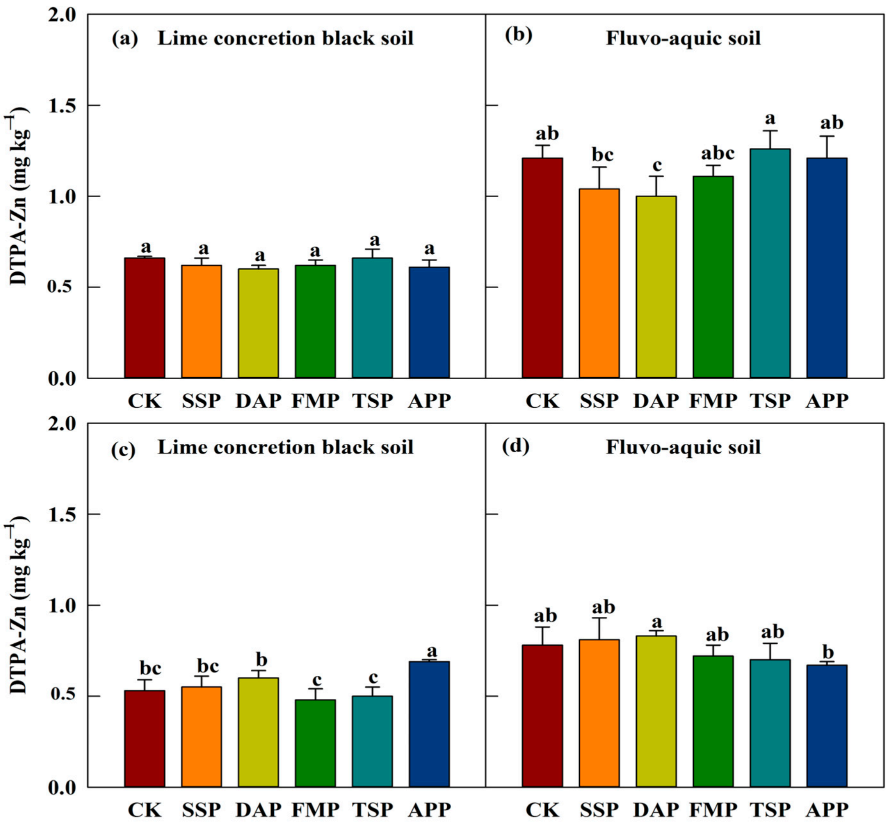

Figure 2. Soil available Zn (denoted as DTPA-Zn) at anthesis (**a**,**b**) and maturity stage (**c**,**d**) under different phosphorus fertilizer types in two calcareous soils. Different lowercase letters represent significantly different among phosphorus fertilizer types in the same soil type ($p < 0.05$). CK, SSP, DAP, FMP, TSP and APP are used to express no P fertilizer, single superphosphate, diammonium phosphate, fused calcium–magnesium phosphate, triple superphosphate and ammonium polyphosphate, respectively.

3.3. Aboveground Zn Accumulation at Anthesis

In the lime concretion black soil, the application of P fertilizer significantly reduced stem leaves, ear, and aboveground Zn accumulation (Figure 3a,c,e). Stem leaves and aboveground Zn accumulation under the FMP treatment were significantly higher than those obtained with other P fertilizer types, with increases of 33.3–52.8% and 16.0–31.8%, respectively (Figure 3a,e). Ear Zn accumulation under the application of TSP was 22.2–37.5% higher than that under the treatments using SSP, DAP, and FMP, but there was no significant difference with respect to APP (Figure 3c). In the fluvo-aquic soil, Zn accumulation in stem leaves under the APP treatment and ear Zn accumulation under the FMP treatment were significantly higher than those for other P fertilizer types, with increases of 18.8–35.7% and 30.0–62.5%, respectively (Figure 3b,d).

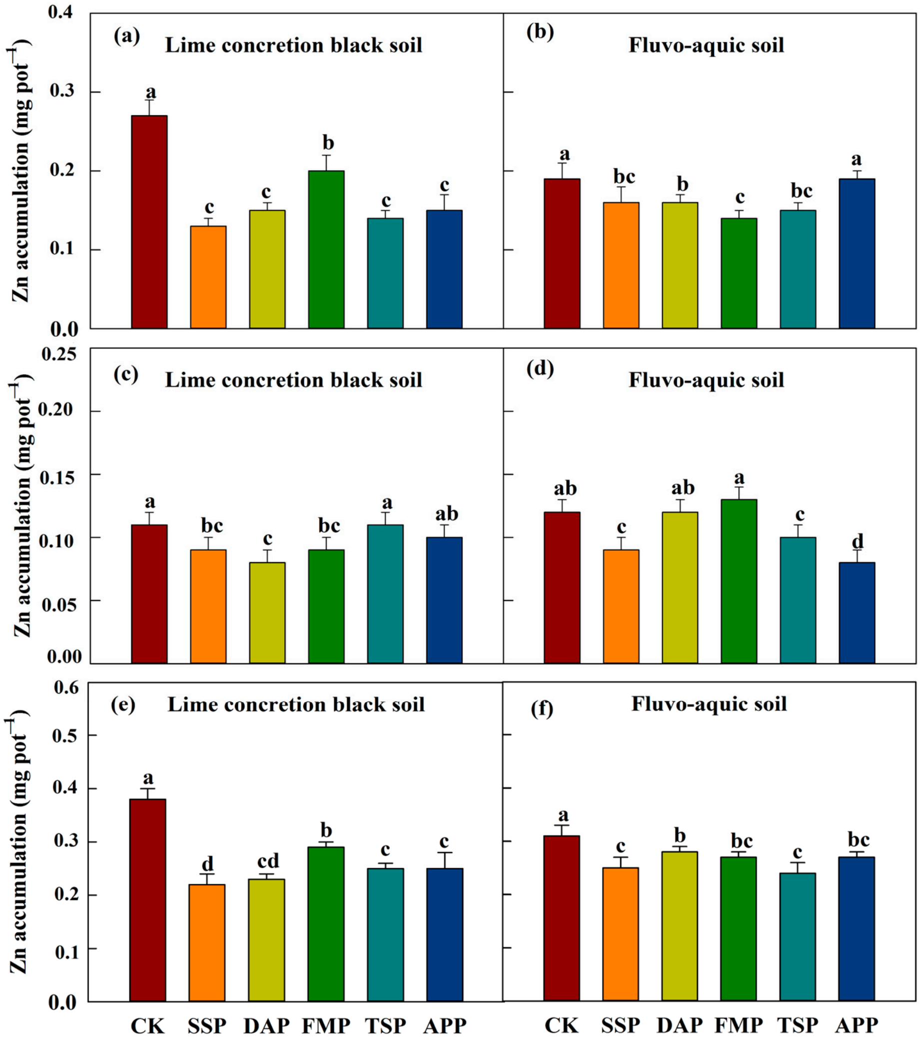

Figure 3. Stem leaf (**a**,**b**), ear (**c**,**d**) and aboveground (**e**,**f**) Zn accumulation at anthesis under different phosphorus fertilizer types in two calcareous soils. Different lowercase letters represent significantly different among phosphorus fertilizer types in the same soil type ($p < 0.05$). CK, SSP, DAP, FMP, TSP and APP are used to express no P fertilizer, single superphosphate, diammonium phosphate, fused calcium–magnesium phosphate, triple superphosphate and ammonium polyphosphate, respectively.

3.4. Aboveground Zn Accumulation at Maturity

In the lime concretion black soil, straw Zn accumulation was the highest under the DAP and APP treatments but was not significantly different from that under the TSP treatment (Figure 4a). Grain Zn accumulation was the highest in the DAP and FMP treatments; especially, it was significantly higher than that in the TSP, SSP, and APP treatment, being 42.9%, 50.0%, and 57.9% greater, respectively (Figure 4c). Aboveground Zn accumulation was the highest in the DAP treatment: it was significantly increased by 9.1–37.1% compared with that observed for other P fertilizer types (Figure 4e). For the fluvo-aquic soil, the application of SSP, DAP, and APP significantly increased straw Zn accumulation by 41.7–50.0%, but its results were not significantly different from those of TSP and FMP (Figure 4b). Grain Zn accumulation in the TSP treatment was significantly higher than that in the treatments using other P fertilizer types (except for the no P fertilizer application treatment), with an increase of 15.4–50.0% (Figure 4d). Aboveground Zn accumulation under the TSP and APP treatments was 13.2% and 10.3% higher than that under the SSP and DAP treatments, respectively, but not significantly different from that under the FMP treatment (Figure 4f).

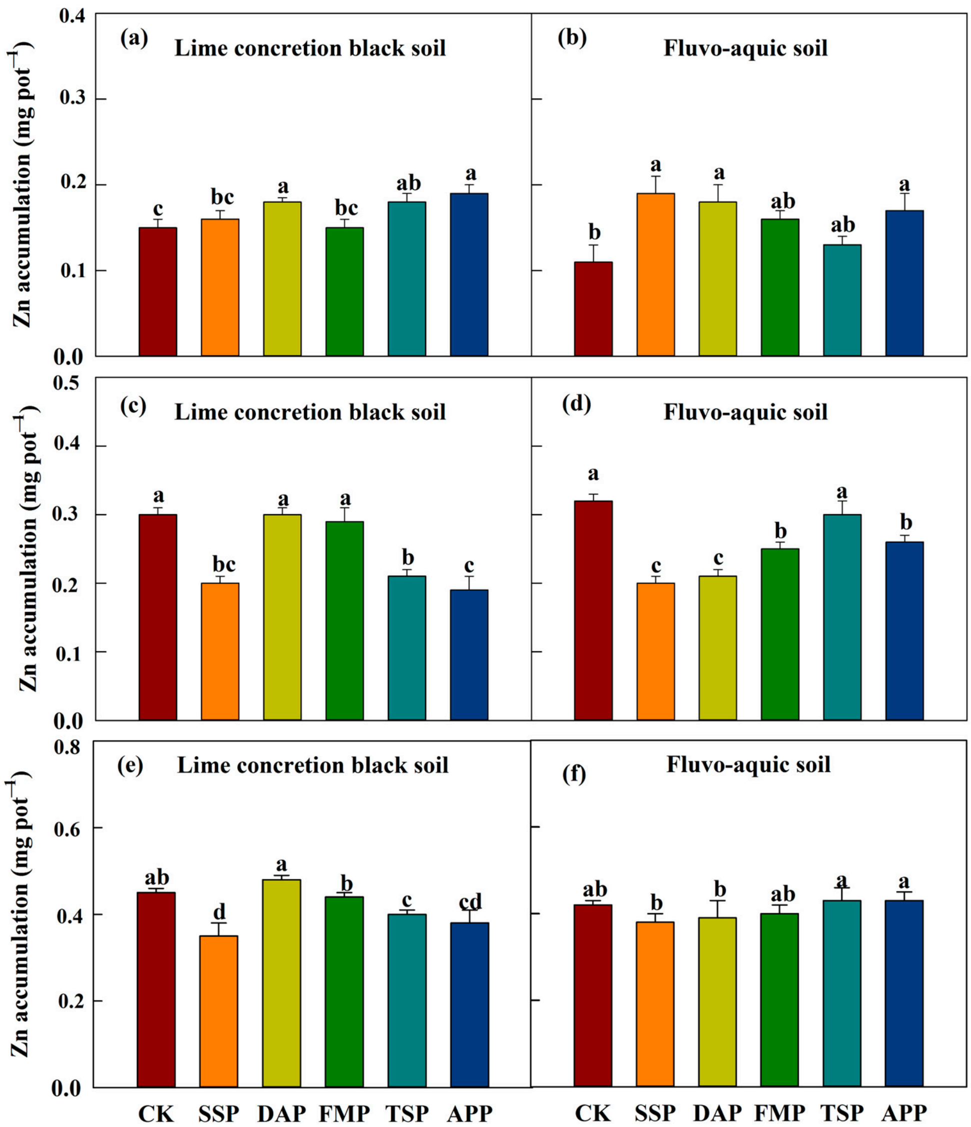

Figure 4. Straw (**a**,**b**), grain (**c**,**d**) and aboveground (**e**,**f**) Zn accumulation at maturity under different phosphorus fertilizer types in two calcareous soils. Different lowercase letters represent significantly different among phosphorus fertilizer types in the same soil type ($p < 0.05$). CK, SSP, DAP, FMP, TSP and APP are used to express no P fertilizer, single superphosphate, diammonium phosphate, fused calcium–magnesium phosphate, triple superphosphate and ammonium polyphosphate, respectively.

3.5. Post-Anthesis Zn Remobilization, Uptake, and Zn Harvest Index

In the lime concretion black soil, compared with the treatment in which no P fertilizer was applied, P application significantly reduced post-anthesis Zn remobilization and the Zn harvest index but increased post-anthesis Zn uptake (Figure 5a,c,e). Among the different P fertilizer types, post-anthesis Zn remobilization and uptake were highest in the FMP and DAP treatments, with increases of 50.0–64.3% and 40.0–48.0%, respectively, and there were no significant differences among the other P fertilizer types. The Zn harvest index was highest in the FMP treatment and was not significantly different from that in the DAP treatment. In the fluvo-aquic soil, post-anthesis Zn remobilization was still higher in the FMP treatment than in the treatments involving other P fertilizer types, but there were no significant differences between the FMP, TSP, and DAP treatments (Figure 5b). Post-anthesis Zn uptake was the highest in the TSP treatment, but there were no significant differences with respect to the APP treatment (Figure 5d). The Zn harvest index was highest in the TSP treatment (70.04%), followed by the FMP and APP treatments, with the SSP treatment yielding the lowest value (Figure 5f).

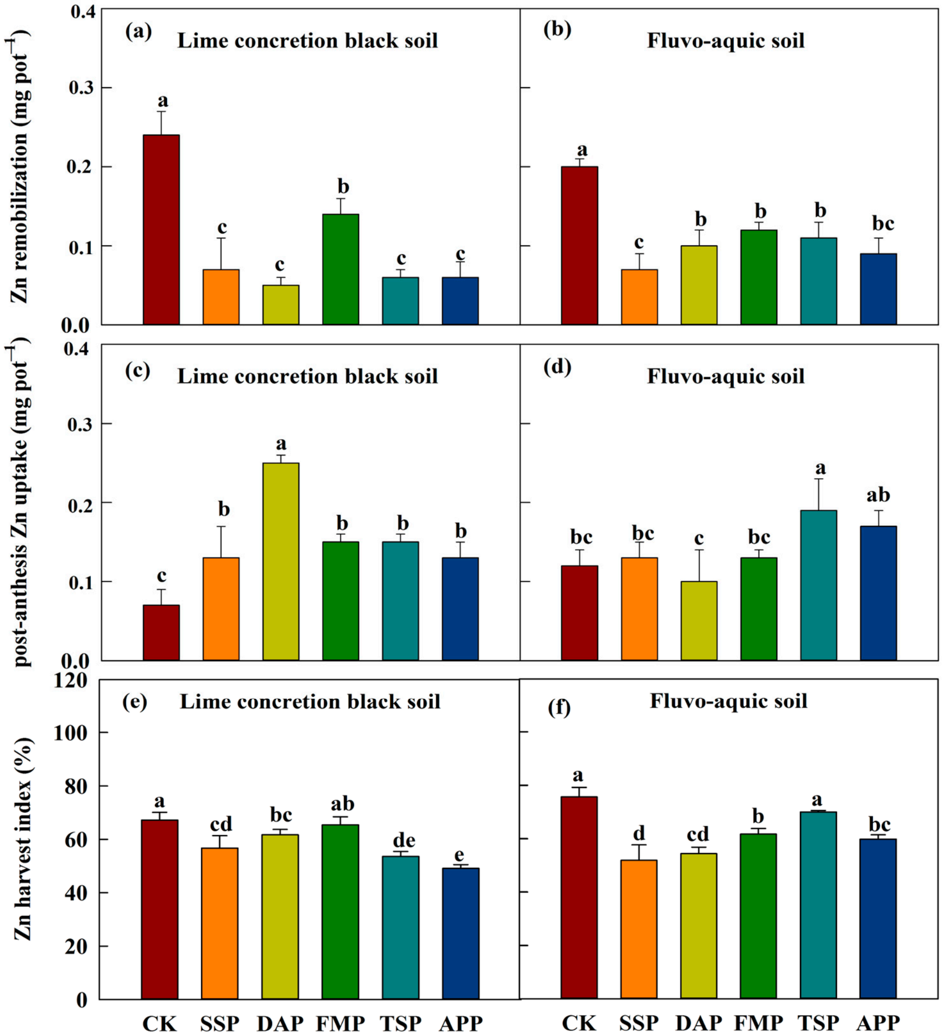

Figure 5. Zn remobilization (**a**,**b**), post-anthesis Zn uptake (**c**,**d**) and Zn harvest index (**e**,**f**) under different phosphorus fertilizer types in two calcareous soils. Different lowercase letters represent significantly different among phosphorus fertilizer types in the same soil type ($p < 0.05$). CK, SSP, DAP, FMP, TSP and APP are used to express no P fertilizer, single superphosphate, diammonium phosphate, fused calcium–magnesium phosphate, triple superphosphate and ammonium polyphosphate, respectively.

3.6. Grain Phytic Acid, PA/Zn Molar Ratio, and Zn Bioavailability

In both calcareous soils, P application significantly increased the PA concentrations and the PA/Zn molar ratios and decreased the Zn bioavailability of winter wheat compared with the treatment without P application (Figure 6). The PA concentrations and PA/Zn molar ratios include by treatment were not significantly different among the treatments with different P fertilizer types for the lime concretion black soil, although the DAP treatment had significantly higher grain Zn bioavailability, but some differences were observed in the fluvo-aquic soil. The PA concentrations and PA/Zn molar ratio under SSP were the highest. However, there were no significant differences between DAP, FMP, and TSP. The increases in PA concentrations and PA/Zn molar ratio under APP were the lowest, and this treatment induced higher Zn bioavailability.

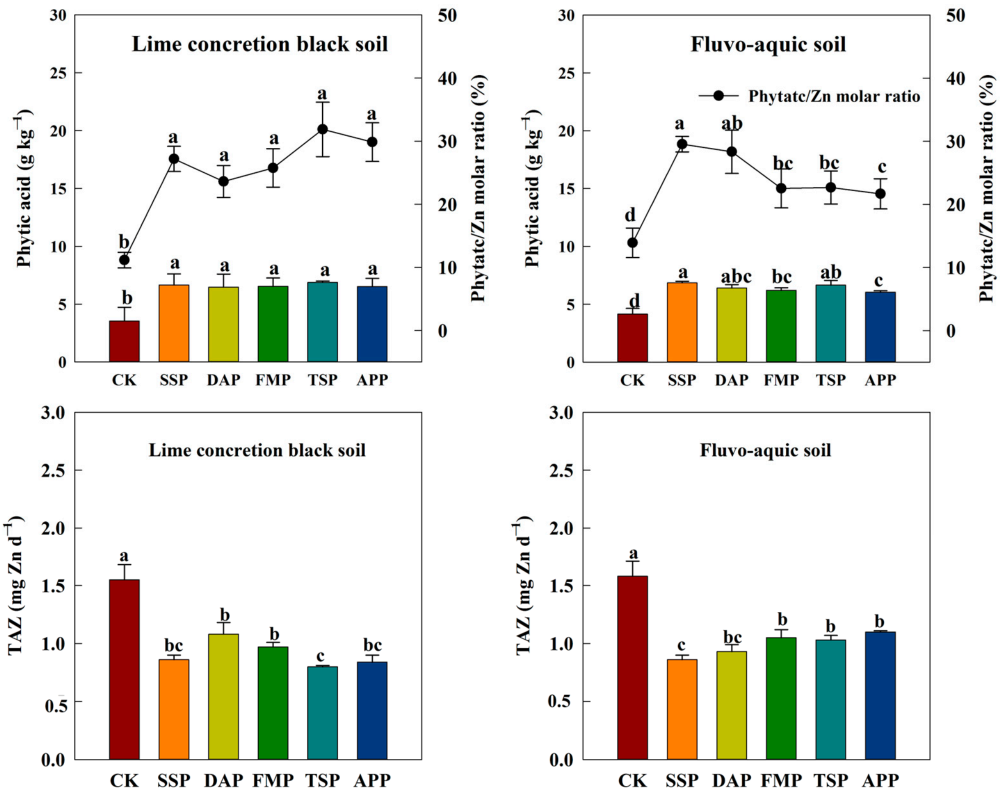

Figure 6. Grain phytic acid, phytate/Zn molar ratio and Zn bioavailability (TAZ) of winter wheat under different phosphorus fertilizer types in two calcareous soils. Different lowercase letters represent significantly different among phosphorus fertilizer types in the same soil type ($p < 0.05$). CK, SSP, DAP, FMP, TSP and APP are used to express no P fertilizer, single superphosphate, diammonium phosphate, fused calcium–magnesium phosphate, triple superphosphate and ammonium polyphosphate, respectively.

4. Discussion

4.1. Wheat Grain Yield Response to Different P Fertilizer Types

There have been many studies on the effects of P fertilizer application on crop yield, and most concur that the application of P fertilizer is an effective measure for improving crop yields [8–10,28]. This conclusion basically stems from the fact that P deficiency is common in most agricultural soils [8,28]. The results of this study are partly in agreement with previous results, as applications of TSP and APP significantly increased wheat grain yield in the fluvo-aquic soil compared to that induced by other P fertilizer types, while the applied P fertilizers did not influence wheat grain yield in the lime concretion black soil. Based on these results, DAP and FMP are not suitable for calcareous soils, and this is probably related to the characteristics of these P fertilizers. DAP and FMP are alkaline, while SSP, TSP, and APP are all acidic. For calcareous soils, alkaline P fertilizer (DAP and FMP) may promote P fixation and is not conducive to crop uptake, whereas acid P fertilizer is relatively weak in terms of chemical fixation and may be more conducive to crop uptake [10,37]. Although SSP is accompanied by a great deal of sulfuric acid in the production process and can be maintained in the fertilizer microdomain environment for a certain period of time in calcareous soil, the release of water-soluble phosphorus in SSP is relatively fast; therefore, the chemical fixation process of soil is relatively fast. As a high-concentration P source, APP is mainly composed of orthophosphorus, pyrophosphate, and polyphosphate. Because orthophosphorus can be directly absorbed by crops, pyrophosphate can also be absorbed by crops after a period of hydrolysis to orthophosphorus phosphate, and polyphosphate can compete with phosphate for adsorption and complex sites and will not be immediately fixed by calcium, iron, and aluminum ions in the soil [38,39]. In addition, APP's chelation of metal ions greatly reduces the degradation of available P in soil [40]; therefore, it yields efficient and stable results both as a short- and long-term P source.

4.2. Soil Available Zn Response to Different P Fertilizer Types

Phosphorus is an important component of a large number of compounds needed by crops, and the application of P fertilizer is an important agronomic measure for ensuring crop yields and quality. Nevertheless, some studies have suggested that the combination of soluble P ions and soil alkalinity after irrational P fertilizer application severely limits the Zn supply in agricultural soils [41]. Zn deficiencies in crop production are mainly due to the unreasonable application of P fertilizer, which inhibits Zn's diffusion in soil as well as its transport within plants [42,43]. Phosphorus and Zn are two plant nutrients known to have antagonistic effects on soil–plant systems. The precipitation reaction between P and Zn promotes the increase in Zn concentrations in ineffective states, such as residual–Zn, thereby reducing Zn availability in soil. It has also been suggested that there is some degree of P and Zn synergy in soil induced by the application of optimal P management practices [26,43,44]. Soil pH is another limiting factor, with high pH leading to reduced availability of micronutrients such as iron and Zn, thus affecting plant morphology and physiological parameters [45]. In this study, the application of different P fertilizer types had no significant effects on the available zinc concentration in lime concretion black soil, but the difference in fluvo-aquic soil might be different from that induced by different P fertilizer types, which altered the soil pH (not given) to different degrees, promoted the activation of Zn in the soil, and thus increased soil available Zn concentrations. A field experiment conducted in paddy soil showed that the application of acid P fertilizer could effectively promote the transformation of soil Zn into weak-acid-soluble and -reducible forms and, thus, increase the soil available Zn concentration by 44.1% compared with a treatment in which P was not applied [17]. The effect of P fertilizer application on soil available Zn is not only reflected in soil pH and the available P level itself but also in the different transformations of soil Zn fractions under different available P levels, in line with the analogous findings of Singh et al. (2021) [42]. Moreover, soil minerals are important components of soil's solid phase. Different soil types have different mineral compositions, which affect the ability of soil to supply P and Zn. The clay mineral composition of lime concretion black soil mainly consists of montmorillonite, which exhibits strong swelling and shrinkage, while the most abundant clay mineral in fluvo-aquic soil is hydromica, with high clay content, and the adsorption capacities of P and Zn are significantly different. Compared with fluvo-aquic soil, the expansion and shrinkage of montmorillonite are weaker, and there are fewer charges, so the adsorption capacity of P and Zn is weaker. Therefore, the application effects of different P fertilizer types on the two calcareous soils were different, especially regarding soil available P and DTPA-Zn. Accordingly, the reasonable application of P fertilizer can alleviate this problem to a certain degree and yield relatively high soil available Zn levels, thus ensuring the uptake and utilization of Zn by crops.

4.3. Grain Zn Concentration and Bioavailability Response to Different P Fertilizer Types

Numerous studies have shown that due to P–Zn antagonism, P fertilizer application affects Zn uptake by crop roots and Zn translocation from vegetative organs to grains, thus significantly reducing Zn accumulation in aboveground organs and affecting grain Zn concentrations [26,27,46,47]. A field experiment conducted on calcareous soil showed that long-term P fertilizer application increased grain yield by 15%, but the grain Zn concentration decreased by 6.8–9.2 mg kg^{-1} per 100 kg P ha^{-1} application, with P application significantly reducing root mycorrhizal infection rates, thus affecting root Zn uptake [13,48]. In both pot and field experiments, the mycorrhizal infection rate significantly decreased with an increasing P application rate [48–50]. For each 1.0% decrease in the mycorrhizal infection rate, the Zn uptake in straw and grain decreased by 1.5 and 1.4 g ha^{-1}, respectively, and grain Zn concentrations decreased by 0.6 mg kg^{-1} [13]. In the present study, it was found that compared with no P application, the application of P fertilizer significantly affected grain Zn concentrations. The application of DAP to lime concretion black soil resulted in a higher grain Zn concentrations, whereas the grain Zn concentrations induced by applying the TSP, APP, and FMP treatments to fluvo-aquic soil were significantly higher than those brought about by the other P fertilizer treatments, the results for which were basically consistent with the trend of post-anthesis Zn remobilization and uptake. Grain Zn accumulation in cereal crops mainly occurs through two pathways: the remobilization of Zn accumulated in aboveground vegetative organs to the grains through the phloem and post-anthesis Zn uptake [32,51,52]. Although P fertilizer application significantly reduced the remobilization of Zn accumulated in the vegetative organs to the grains, post-anthesis Zn uptake was significantly higher after DAP application than after the other P treatments, a finding that is consistent with the higher grain Zn concentrations induced by the DAP treatment applied to lime concretion black soil. Moreover, the reason for this may also be that P application induced P-Zn antagonism in plants, which significantly affected the remobilization of Zn from the vegetative organs to the grains through phloem, but P application promoted the absorption of Zn during the grain-filling stage to a certain extent; and had a positive effect on the increase in the Zn concentration in grains [11,41,53]. From the differences and characteristics of the two tested soil types, it is clear that APP has a higher precipitation rate in soil than that of DAP, and when it is applied to soil, it has a stronger ability to reduce soil pH in the topsoil layer than DAP, and it is less affected by soil fixation and has a longer migration distance. Moreover, lime concretion black soil and fluvo-aquic soil have high proportions of calcium and magnesium ions and are weakly alkaline, while the pH of APP is close to neutral, and FMP is an acidic fertilizer, which has a buffer effect on soil pH after application. The adsorption of inorganic P and Zn by the large amount of calcium carbonate present in lime concretion black soil in which sand ginger was grown decreased with the decrease in pH value. In addition, P in APP is mainly polymerized, allowing it to chelate with calcium and Zn plasma in soil after application, reduce the adsorption and fixation of orthophosphate and Zn ions in soil, and increase the soil available P and Zn concentrations. Therefore, TSP application to fluvo-aquic soil increases the grain Zn concentration by directly promoting post-anthesis Zn uptake and an increase in the Zn harvest index. Research has proven that using rational P fertilizers for winter wheat can ensure high grain yields and root activity and maintain high Zn nutrition levels in aboveground organs [11,13]. The last two contribute to increasing Zn remobilization from vegetative organs to grains and post-anthesis Zn uptake by roots and help to ensure that wheat grains are rich in Zn, supporting the nutritional health of those who consume dietary grains.

Phytic acid is the main form of P present in grains, and PA-P accounts for approximately 60–80% of the total P in grains, whereas Zn ions easily combine with PA to form insoluble precipitates [23,54]. Numerous studies have shown that the application of P fertilizers significantly increases the PA concentration of crop grains, which, in turn, seriously affects the human body's absorption of Zn; moreover, the human body is unable to effectively absorb Zn nutrients under the conditions of an adequate dietary intake, leading to the persistence of Zn deficiency in people who consume cereals as a staple food [11,23,54,55]. Currently, the PA/Zn molar ratio is often used as an indicator for evaluating the bioavailability of Zn in grains, and when the PA/Zn molar ratio exceeds 15, phytate significantly decreases Zn absorption in the human body [56,57]. The results of the current study showed that after the application of P fertilizer to lime concretion black soil and fluvo-aquic soil, the PA/Zn molar ratio reached more than 15, and P fertilizer significantly reduced grain Zn bioavailability. However, under the influence of DAP on lime concretion black soil and of APP, FMP, and TSP on fluvo-aquic soil, the smaller the inhibition effect, the smaller the effect on grain Zn bioavailability. This finding is consistent with the variation in Zn concentrations in grain in the two calcareous soils treated with different P fertilizer types.

5. Conclusions

To address the problem of hidden hunger, especially Zn deficiency in wheat, the effects of P fertilizer types on wheat yield, grain Zn concentrations, and bioavailability in calcareous soil were quantified. In lime concretion black soil, although P fertilizer types had no significant effect on grain yield, DAP treatment significantly increased post-anthesis Zn uptake and the Zn harvest index, which not only increased Zn accumulation in grains but also increased grain Zn concentrations and Zn bioavailability. In fluvo-aquic soil, the application of TSP and APP not only significantly increased wheat yield but also led to higher post-anthesis Zn remobilization and uptake, Zn harvest index value, grain Zn concentrations, and bioavailability, alleviating the inhibitory effect of P application on plant Zn nutrition. Based on the analysis of yield, grain Zn concentrations, and bioavailability, DAP was determined to be the optimal P fertilizer type for application to lime concretion black soil, while TSP and APP were the most suitable for fluvo-aquic soil, constituting findings that could ensure food security and alleviate deficiencies in or improve human Zn nutrition and health in developing countries or regions where wheat is the main food source.

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