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Article

Consequences of 33 Years of Plastic Film Mulching and Nitrogen Fertilization on Maize Growth and Soil Quality

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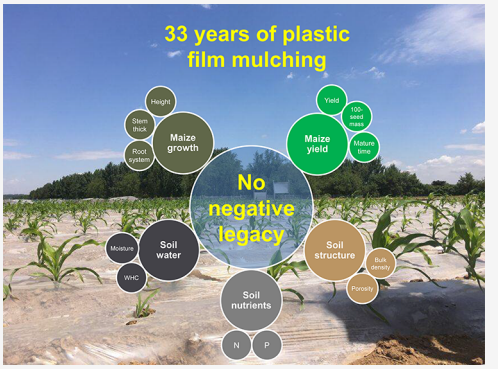
ABSTRACT: Plastic film mulching and urea nitrogen fertilization are widely used in agricultural ecosystems, but both their long-term use may leave a negative legacy on crop growth, due to deleterious effects of plastic and microplastic accumulation and acidification in soil, respectively. Here, we stopped covering soil with a plastic film in an experimental site that was previously covered for 33 years and compared soil properties and subsequent maize growth and yield between plots that were previously and never covered with the plastic film. Soil moisture was about 5−16% higher at the previously mulched plot than at the never-mulched plot, but NO_3^- content was lower for the former when with fertilization. Maize growth and yield were generally similar between previously and never-mulched plots. Maize had an earlier dough stage (6−10 days) in previously mulched compared to never-mulched plots. Although plastic film mulching did add substantial amounts of film residues and microplastic accumulation into soils, it did not leave a net negative legacy (given the positive effects of the mulching practice in the first place) for soil quality and subsequent maize growth and yield, at least as an initial effect in our experiment. Long-term urea fertilization resulted in a pH decrease of about 1 unit, which bring a temporary maize P deficiency occurring in early stages of growth. Our data add long-term information on this important form of plastic pollution in agricultural systems.



KEYWORDS: *plastic pollution, microplastic, legacy effect, soil health, crop performance*

1. INTRODUCTION

Plastics, a group of artificially synthesized compounds, are now ubiquitous on the earth even in remote places such as near the top of Mount Everest.[1] In recent decades, plastic pollution has attracted great attention due to its potential ecological and environmental implications on a global scale.[2] Consequently, plastic pollution was listed as one of the top 10 global environmental problems in 2014 by the United Nations Environment Program.[3] Due to the widespread use of plastic mulch, shed plastic film, and biosolids,[4−6] croplands have been identified as a major reservoir of plastic debris.[7] Plastic pollution in croplands has the potential to threaten long-term food security, as it possibly negatively impacts soil health and crop yield.[8,9]

Polyethylene (PE) plastic film mulching (PFM) is widely used in global agricultural ecosystems to improve crop yield because it increases soil temperature and moisture.[10−14] A recent meta-analysis showed that PFM increased crop yields by 24% on average.[15] However, increased adoption and time of soil contact results in greater soil accumulation of plastic residues because plastic films often cannot be completely removed, especially the thin films (i.e., 5−8 μm thick) used in countries such as China.[9,16,17] Our recent study showed that macro-residues of the plastic film (diameter >5 mm) were as high as 360 kg ha^{-1} and film-derived microplastics (<5 mm) exceeded 8000 items per kg soil in the 0−10 cm layer after 32 years of PFM.[18] Accordingly, long-term PFM is expected to leave a negative legacy for crop growth and yield.

In the previous literature, numerous studies explored the effects of the plastic residual film or PE microplastic addition on soil quality and crop performance. Excessive residual plastic (>360 kg hm^{-2}) accumulated in soil could decrease pore connectivity and porosity,[19] thus affecting the movement of nutrients and water in the soil.[20] The germination of cotton seeds would be compromised by the residual film when above 200 kg hm^{-2},[21] and the development of maize roots were influenced when above 150 kg hm^{-2}.[22] Hu et al.[22] found that maize yield was decreased by 15−18% and 23−25%, when adding plastic film residues into the tillage layer at levels of 300 and 600 kg ha^{-1}, respectively. A meta-analysis showed a reduction of yield by 3% for cotton but little effect on potato

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and maize at 100 kg ha^{-1} of residual film addition, as estimated through regression relationships between yield and soil residual film amount.[8] Negative[23,24] and no[25−27] impacts of PE microplastic on crop performance effect have both been reported for different types of crops, such as maize. However, all those previous studies were based on the artificial addition of fresh plastic residual film or microplastic into soils, which may not fully reflect reality. The reason is that plastic film in the field passes through a complex fragmentation and degradation process, which requires appreciable time. To our knowledge, there is no evaluation of the legacy of long-term PFM on subsequent crop growth and yield.

Our study evaluated the legacy effects of 33 years of PFM on soil properties, maize growth, and yield in a continuous PE plastic (non-biodegradable) film mulching and urea fertilization experiment initiated in 1987. To investigate the legacy effect, previous mulching plots were not covered with the PE film in 2021 and never-mulched plots served as a control. Maize above- and below-ground growth indices and soil basic physical and chemical properties were measured at different maize growth stages. Our aim was to test the hypothesis that long-term PFM leaves a net negative legacy on maize growth and yield, due to deleterious effects of plastic and microplastic accumulation in soil outweighing any positive legacy effects of the mulching practice. We also expect that long-term nitrogen (N) fertilization with urea would have a negative effect on maize growth, due to soil acidification and its induced plant phosphorus limitation. These results will test the sustainability of long-term agricultural management in croplands.

2. MATERIALS AND METHODS

2.1. Study Site and Experiment Design. The experimental field site was a long-term PE film mulching (colorless and transparent, 8 μm thick) and fertilization station (built in 1987) at Shenyang Agriculture University (41°49′N, 123°34′E) in Shenyang, Liaoning Province, China. This site has a temperate continental monsoon climate, with a mean annual temperature of 7.9 °C and average annual rainfall of about 705 mm. The soil is a brown earth according to Chinese Soil Taxonomy (a Haplic-Udic Alfisol according to US Soil Taxonomy). The experiment was arranged in a split-plot design with two levels of PFM (with and without) as main plots and two levels of N fertilizer as subplots that produces a combination of four treatments with three plot replicates by treatment. The fertilizer levels included (i) zero N fertilizer (N_0) and (ii) 135 kg N ha^{-1} year^{-1} application (N_{135}). Each plot had an area of 69 m^2. The N fertilizer was urea powder, applied as basal fertilizer in spring. The crop type is monoculture maize (*Zea may* L.) with a conventional tillage system (a 20 cm-depth rotational till by a rotary cultivator in fall and ridging by a ridge plow in spring). A detailed description of agricultural operations at this field can be seen in Ding et al.[13]

In order to investigate the legacy effect of previous PFM, two ridges (5 × 2 m^2) were randomly selected within previous PFM plots to cease covering with plastic film in 2021: this is referred to as previous PFM (PrevPFM). Plots that never possessed PFM were set as the control, i.e., never-PFM plots (NeverPFM). Soil properties and maize growth at the N_0 and N_{135} plots under previous and never-PFM treatments (called N_0-PrevPFM, N_{135}-PrevPFM, N_0-NeverPFM, and N_{135}-NeverPFM, respectively) were measured during the growing season in 2021.

2.2. Sampling and Measurements. Soil moisture, plant height, and stem diameter were measured every 7 days from June to July, every 14 days from July to August, and every 21 days from August to September in 2021. Soil moisture (cm^3/cm^3) was measured for the surface horizon (0-10 cm) using a moisture probe (Trime-Pico 64/32, IMKO GmbH, Ettlingen, Germany). Three plants were randomly selected from each plot. Plant height was measured from the base to the tip with steel tape, and stem diameter, defined as the middle diameter of the second aboveground section, was measured with a vernier caliper.

Leaf pigments, above- and below-ground biomass, root morphological properties, root phosphorus concentration, and associated phosphatase activity were measured at the sixth leaf stage (V6, the key period from vegetative to reproductive growth, about 48 days after seeding), the tasseling stage (VT, the period when the plant reaches its full height and begins to shed its pollen, about 90 days after seeding), and physiological maturity stage (R6, about 149 days after seeding). The sampling dates for each of the three stages occurred when more than 80% of the plants were in that respective stage. Chlorophyll and flavonoid contents were measured for the third fully expanded mature leaf from top to bottom for a selected plant at 9:00−11:30 in the morning using a Dualex Scientific (Force-A, Orsay, France) portable meter. Two plants were randomly sampled from each plot and then divided into above- and below-ground tissues by cutting the first section of the stem with a sickle. Plant tissues were oven dried at 60 °C to constant weight. Within each plot, two plants were randomly sampled by excavating the soil adjacent to the main trunk up to a radius of 15 cm and a depth of 40 cm and collecting all scattered roots. Roots were washed with tap water to remove soil and then rinsed with ultrapure water 3−5 times. Roots from a single plant were cut into parts and measured using a root scanner (EPSON Expression 11000XL) and an image analyzer (the WinRHIZO software, Regent Instr., QC, Canada) for root morphology, including total root length, total surface area, total volume. Scanned roots were dried to a constant mass at 60 °C and then weighed. Dry roots were ground and passed through a 0.25 mm sieve and then digested with a combination of H_2SO_4 and H_2O_2 (8:5) to determine root phosphorus concentrations.[28] The remaining root was used to determine root-associated phosphatase activity (APase).[29]

Soil samples were collected at the 0−20 cm layer for the measurements of pH, plant-available soil phosphorus (Olsen-P), soil acid phosphatase (AcP), ammonium nitrogen (NH_4^+-N) and nitrate nitrogen (NO_3^--N) contents, bulk density, total porosity, and water holding capacity at corresponding crop stages. Three soil cores were randomly sampled using an auger (4 cm in diameter) and then composited for each plot. Soil samples were passed through a 2 mm sieve to remove plant debris and gravel. One part was air dried to determine soil pH and Olsen-P, and the field-moist soil was used to determine soil acid phosphatase (AcP), NH_4^+-N, and NO_3^--N (values were expressed on a dry weight basis). Soil pH was measured by a glass electrode in a 1:2.5 soil/distilled water suspension after shaking. Olsen-P concentration was measured after extraction with 0.5 M $NaHCO_3$ according to the colorimetric method.[28] Soil NH_4^+-N and NO_3^--N were extracted with 10 mM $CaCl_2$ (soil/water = 1:10) and measured using a continuous flow analyzer (Bran-Luebbe AA3, Germany). Soil bulk density, total soil porosity, and soil water holding capacity

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Environ. Sci. Technol. 2023, 57, 9174−9183

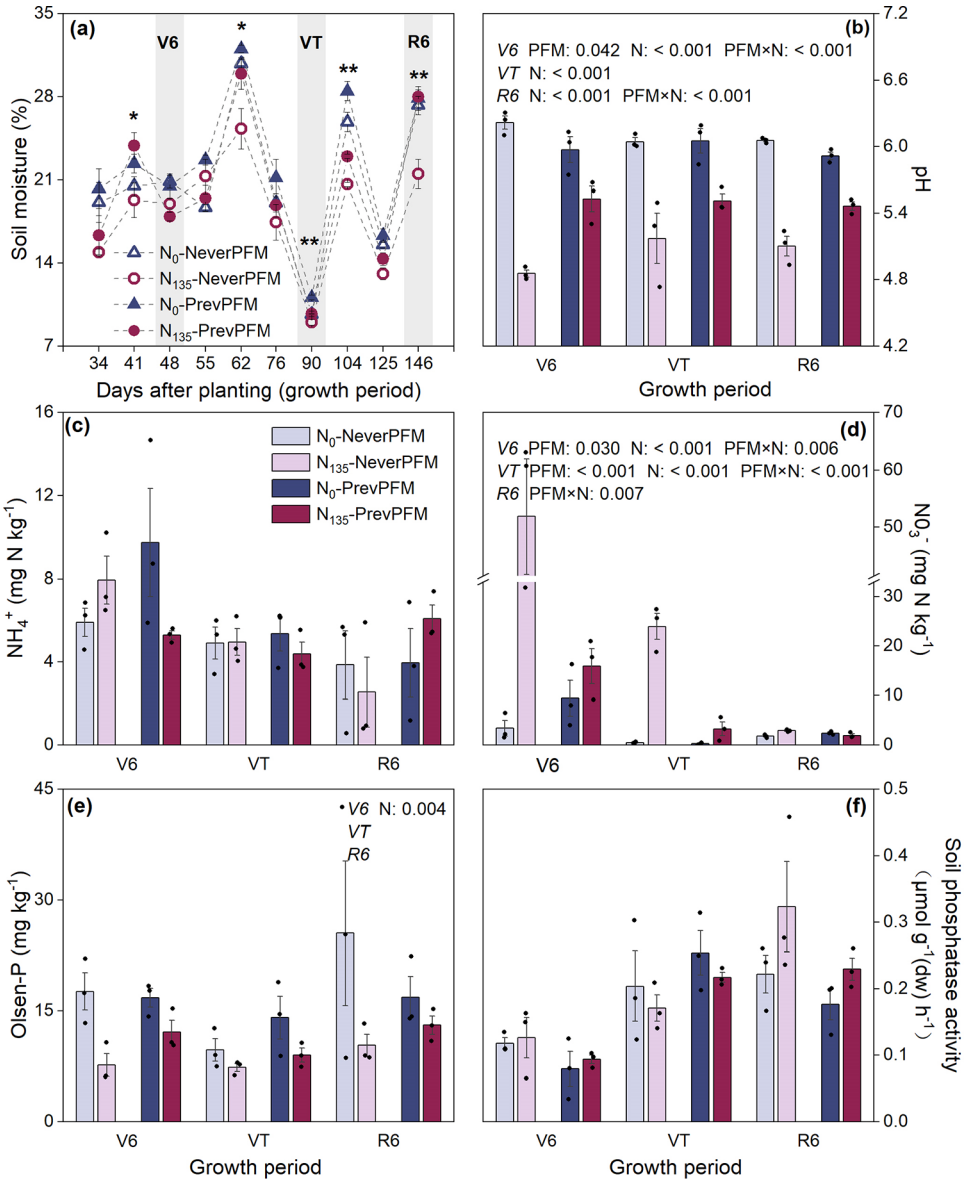

Figure 1. Soil moisture (a), pH (b), NH_4^+-N (c), NO_3^--N (d), Olsen-P (e), concentrations and phosphatase activity (f) during growth seasons. V6: sixth leaf stage, VT: tasseling stage, R6: physiological maturity stage. N_0: zero N fertilizer, N_{135}: 135 kg N ha^{-1} $year^{-1}$, PrevPFM: previous plastic film mulching, and NeverPFM: never plastic film mulching. Bars represent ±standard errors of the replicates (n = 3) and individual data points are shown as black opaque circles. The symbols "**" and "*" in panel (a) denote main effects of plastic film mulching from ANOVA results at $P < 0.01$ and $P < 0.05$, respectively. The decimals after the treatment acronyms "PFM", "N", or "PFM × N" represent the P values for main effects of plastic film mulching, N fertilization, and their interaction, respectively. Only P values less than 0.05 are shown in panels.

were determined according to the cutting-ring method in Chen.[30] After crop harvest in autumn, soil compaction was measured using a SC-900 soil compaction meter (Spectrum Technologies, Aurora, IL, USA). The conical head was pushed down at a constant speed and inserted into the soil with 45 cm depth, and data were automatically read and recorded.

Soil acid phosphatase and root-associated phosphatase activities were measured following the spectrophotometer method in Lin et al.[29] Briefly, 1 g fresh soil or 0.2 g fresh roots (<2 mm) were transferred into a centrifuge tube containing 50 mM acetate buffer (pH = 5.0). Then, 5 mM p-nitrophenyl phosphate (pNPP) was added to the centrifuge tube as the reaction substrate. The centrifuge tube was kept in the dark at 20 °C for 1 h, until stopping the reaction by adding 0.5 M NaOH and 0.5 M $CaCl_2$. Absorbance of p-nitrophenol (pNP) in the supernatant was then measured at 410 nm by an Unic-7200 Spectrophotometer (Shanghai, China). Four analytical replicates were used for each root sample, including a blank. For the blank, pNPP was added after NaOH and $CaCl_2$ stopped the reaction. The concentration of pNP is obtained by the standard curve between the configured pNP concentration and the absorbance value. Soil phosphatase activity is expressed by pNP produced in the above reaction divided by reaction time and dry weight. Root-associated phosphatase activity is expressed by pNP produced in the above reaction divided by reaction time and fresh weight.

Moreover, we observed and recorded the time when maize entered into the dough stage, which is defined as the time when starchy material within most kernels has dough-like consistency and accumulate almost 50% of the dry mass.[31] At

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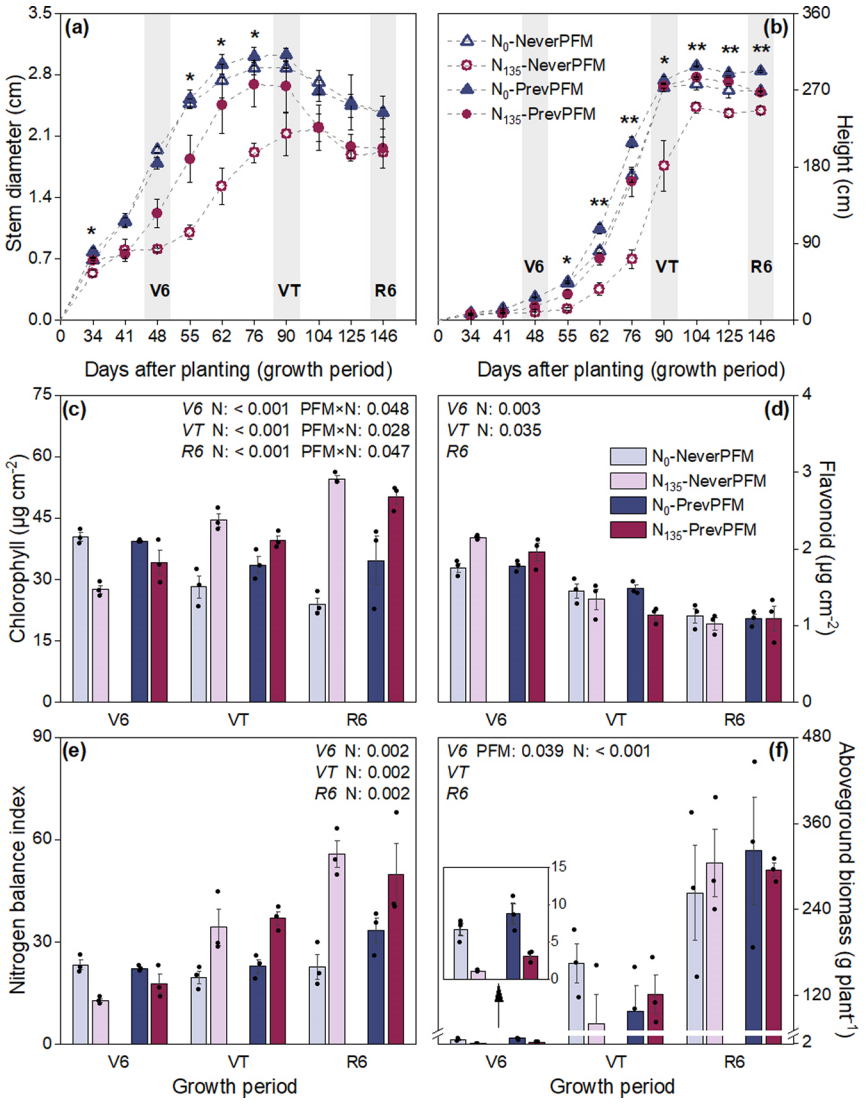

Figure 2. Maize aboveground parameters during various growth stages. Stem diameter (a), height (b), leaf chlorophyll (c), flavonoid (d), nitrogen balance index (e), and aboveground biomass (f). The nitrogen balance index was calculated by chlorophyll/flavonoid. V6: sixth leaf stage, VT: tasseling stage, and R6: physiological maturity stage. N_0: zero N fertilizer, N_{135}: 135 kg N ha^{-1} $year^{-1}$, PrevPFM: previous plastic film mulching, and NeverPFM: never plastic film mulching. Bars represent ±standard errors of the replicates (n = 3), and individual data points are shown as black opaque circles. The symbols "**" and "*" in panel (a) denote main effects of plastic film mulching from ANOVA results at $P < 0.01$ and $P < 0.05$, respectively. The decimals after the treatment acronyms "PFM", "N", or "PFM × N" represent the P values for main effects of plastic film mulching, N fertilization, and their interaction, respectively. Only P values less than 0.05 are shown in panels.

the physiological maturity stage, the yield was measured by randomly selecting four plants in the middle of each plot. The 100-seed dry weight (randomly chosen 100 maize seeds) and the length of the maize cob were recorded. Maize ears were dried at 60 °C to constant weight in an oven and then used to obtain the yield.

2.3. Statistical Analyses and Calculations. The effects of PFM (PrevPFM and NeverPFM, whole-plot factor), N fertilization (N_0 and N_{135}, subplot factor), and their interactions on soil and crop parameters were assessed by split-plot ANOVA at each sampling time. Normality of residuals and homogeneity of the variances of the residuals across groups were checked through the Shapiro−Wilk test and Levene's test, respectively.[32] When necessary, the data were logarithmically transformed. Pearson's correlation analyses were conducted between plant growth parameters and three soil parameters (i.e., pH, moisture, and Olsen-P concentrations) at the sixth leaf stage, tasseling stage, and physiological maturity stage, respectively.

To understand how the treatments (PrevPFM vs NeverPFM and N_0 vs N_{135}) influence total maize performance and their relations with soil properties, redundancy analysis (RDA) was conducted based on crop performance data (stem diameter, height, above- and below-ground biomasses, total root length, root surface area, chlorophyll, root P, and APase) and soil properties (pH, soil moisture, Olsen-P, bulk density, soil porosity, water holding capacity, and AcP). Monte Carlo permutations were used to test significance of relationships between selected soil factors and plant growth ($P < 0.05$), and we then tested the significance of the difference between each soil factor and plant growth through the envfit function in the vegan package. RDA was performed using R. 4.1.3. The other statistical analyses were conducted using SPSS version 22.0. All reported differences are significant at $P < 0.05$.

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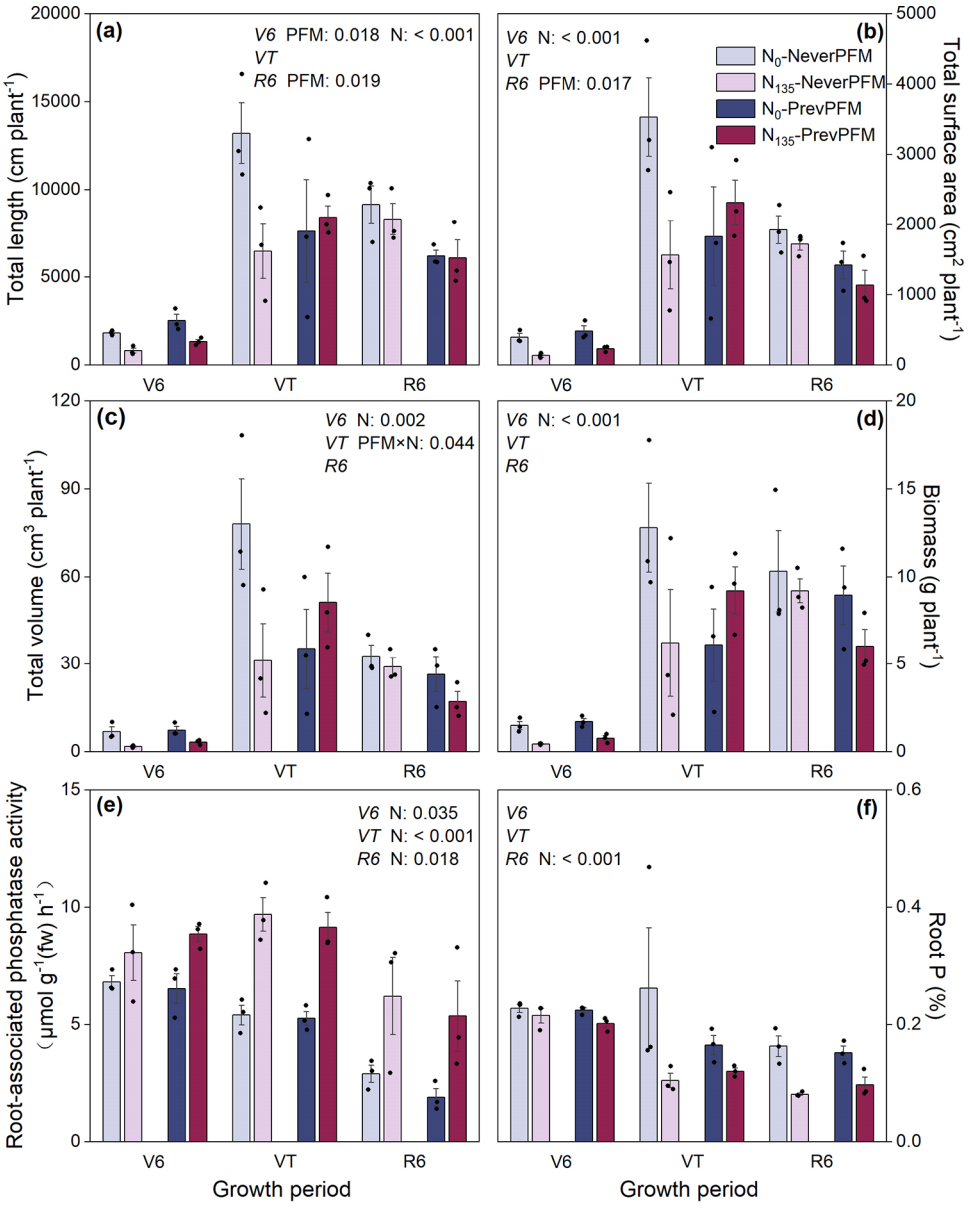

Figure 3. Maize belowground (root) parameters during various growth stages. Total length (a), total surface area (b), total root volume (c), biomass (d), root associated phosphatase activities (e), and P concentration (f). V6: sixth leaf stage, VT: tasseling stage, and R6: physiological maturity stage. N_0: zero N fertilizer, N_{135}: 135 kg N ha^{-1} $year^{-1}$, PrevPFM: previous plastic film mulching, and NeverPFM: never plastic film mulching. Bars represent ±standard errors of the replicates (n = 3), and individual data points are shown as black opaque circles. The decimals after the treatment acronyms "PFM", "N", or "PFM × N" represent the P values for the main effects of plastic film mulching and N fertilization or their interaction, respectively. Only P values less than 0.05 are shown in panels.

3. RESULTS

3.1. Soil Properties. Soil moisture was about 5−16% higher for previous PFM than for never-mulching (most P < 0.05, df = 1, Figure 1a and Table S1). In the N_{135} treatments, soil pH was higher in previously mulched plots than in never mulched plots (Figure 1b). Soil NH_4^+-N concentrations were similar between previous and never PFM (P > 0.05, df = 1, Figure 1c), but NO_3^--N concentrations were lower for previous PFM than never PFM at the sixth leaf stage and tasseling stage only for N_{135} treatment (PFM × N: P = 0.006, df = 1 and PFM × N: P < 0.001, df = 1, respectively, Figure 1d). Soil Olsen-P concentrations and phosphatase activity were both similar between previous and never PFM in all the growth stages (P > 0.05, df = 1, Figure 1e,f). Both NH_4^+-N and NO_3^--N concentrations were lower at tasseling and physiological maturity stages than at the sixth leaf stage (Figure 1c,d). Soil phosphatase activity was higher at tasseling and physiological maturity stages than at the sixth leaf stage (Figure 1f), although Olsen-P changed little across growth stages (Figure 1e).

Soil moisture was about 5−21% lower at N-fertilized plots than at non-fertilized plots for most of the growing season (Figure 1a and Table S1). Average soil pH was about 1 unit lower at N-fertilized plots than at non-fertilized plots across growth stages (P < 0.001, df = 1, Figure 1b). Soil NO_3^--N concentrations were higher with fertilizer addition at never-mulched plots but previously PFM plots showed no difference during the sixth leaf stage (PFM × N: P = 0.006, df = 1) and tasseling stage, PFM × N: P < 0.001, df = 1, Figure 1d), but the two fertilizer level plots always had similar NH_4^+-N (P >

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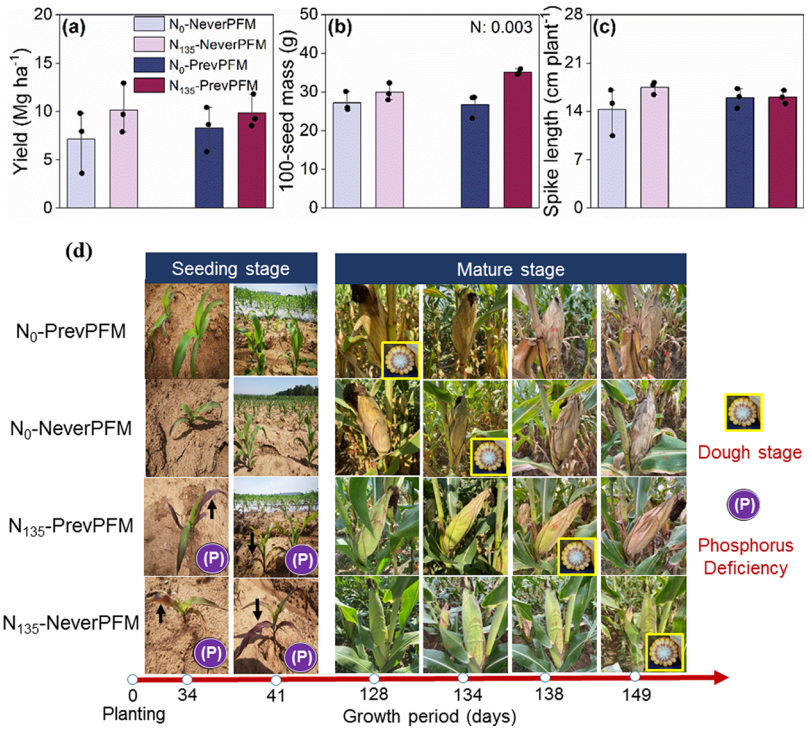

Figure 4. Maize yield (a), 100-seed mass (b), spike length (c), and growth process and maturation time (d) under the combined plastic film mulching and fertilization with urea-nitrogen (N) treatments. N_0: zero N fertilizer, N_{135}: 135 kg N ha^{-1} $year^{-1}$, PrevPFM: previous plastic film mulching, and NeverPFM: never plastic film mulching. Bars represent ±standard errors of the mean (n = 3) and individual data points are shown as black opaque circles. The decimals after the treatment acronyms "PFM", "N", or "PFM × N" represent the P values for the main effects of plastic film mulching and N fertilization, or their interaction, respectively. Only P values less than 0.05 are shown in panels.

0.05, df = 1, Figure 1c). Soil Olsen-P concentrations were lower at N fertilized than at non-fertilized plot, especially at the sixth leaf stage (i.e., 17.21 mg kg^{-1} vs 9.89 mg kg^{-1}) (P = 0.004, df = 1, Figure 1e). Soil phosphatase activity did not differ between the contrastingly fertilized plots (P > 0.05, df = 1, Figure 1f).

3.2. Maize Above- and Below-Ground Parameters. Long-term PFM did not have a negative legacy for subsequent maize and even promoted maize growth in some cases. Stem diameter and height were generally greater for previous PFM than for never mulching across the whole growing season, especially at the N_{135} level (most P < 0.05 or P < 0.01, df = 1, Figure 2a,b). Correspondingly, aboveground biomass was larger for previous PFM than for never mulching, but these differences only occurred at the sixth leaf stage (P = 0.039, df = 1, Figure 2f) and disappeared at tasseling and maturity stages (P > 0.05, df = 1, Figure 2f). Both leaf chlorophyll and flavonoid concentrations and NBI were similar between previous and never PFM (P > 0.05, df = 1, Figure 2c–e). Total root length was 46% higher in previous PFM than in never mulching treatment at the sixth leaf stage (P = 0.018, df = 1, Figure 3a), but this trend was reversed at the physiological maturity stage (P = 0.019, df = 1). Similarly, the root total surface area was about 30% smaller for previous PFM than for never mulching only at the physiological maturity stage (P = 0.017, df = 1, Figure 3b). However, other root properties, i.e., total volume, biomass, root-associated phosphatase activity, and root P were all similar between previous PFM and never mulching (P > 0.05, df = 1, Figure 3c–f).

Long-term N fertilization inhibited maize growth, especially at the seedling stage. Specifically, stem diameter and height were much lower at N-fertilized plots than at non-fertilized plots during the whole growing season (Figure 2a,b). Correspondingly, aboveground biomass was much smaller at N-fertilized plots than at non-fertilized plots, but these differences only occurred at the sixth leaf stage (P < 0.001, df = 1, Figure 2f) and disappeared at tasseling and maturity stages (P > 0.05, df = 1). At the sixth leaf stage, plants from N-fertilized plots had lower chlorophyll concentrations and NBI but higher flavonoid contents in leaves than non-fertilized plots, especially for never PFM (P < 0.001, Figure 2c; P = 0.002 Figure 2e; and P = 0.003, Figure 2d respectively). By contrast, at tasseling and maturity stages, chlorophyll concentrations were higher in N-fertilized plots, especially for never PFM (P < 0.001, df = 1, Figure 2c). Roots generally followed similar trends to aboveground biomass in response to N fertilization. Root biomass, total root length, total surface area, and total volume were much smaller at N fertilized than at non-fertilized plots at the sixth leaf stage (all P < 0.01, df = 1, Figure 3a–d), but the difference disappeared at tasseling and maturity stages (P > 0.05). In response to Olsen-P deficiency induced by N fertilization (Figure 1e), root-associated phosphatase activities were about 20–100% higher at N-fertilized plots than at non-fertilized plots during the whole growing season (all P < 0.05 or P < 0.001, df = 1, Figure 3e). Accordingly, root P concentrations were lower at N fertilized plots, especially for the physiological maturity stage (P < 0.001, df = 1, Figure 3f).

3.3. Maize Yield and Maturation Time. Maize yields were similar between previous and never PFM (P > 0.05, df = 1, Figure 4a), and also yield parameters (100-seed mass and spike length) (P > 0.05, df = 1, Figure 4b,c). However, maize at previous PFM plots had an earlier dough stage (6–10 days) than those at never mulching plots (Figure 4d). Maize yield

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Environ. Sci. Technol. 2023, 57, 9174–9183

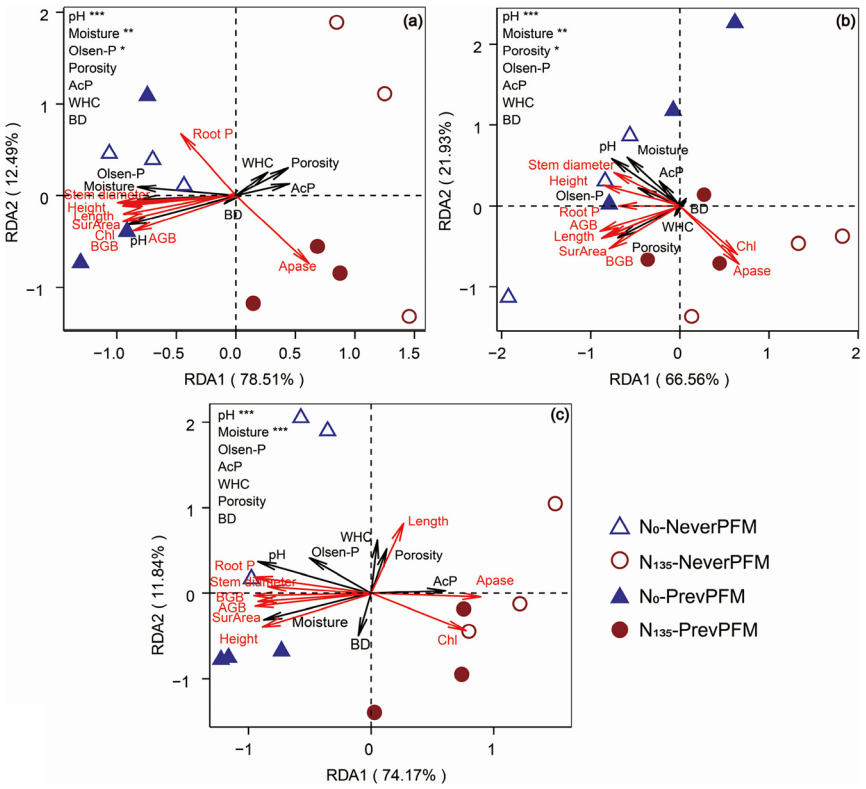

Figure 5. Redundancy analysis of plant growth impacted by soil properties at the sixth leaf stage (a), tasseling stage (b), and physiological maturity stage (c). Red and black arrows indicate plant growth parameters and soil properties, respectively. SurArea: total root surface area; AGB: aboveground biomass; BGB: belowground biomass; Chl: chlorophyll; APase: root-associated phosphatase activity; BD: soil bulk density; WHC: water holding capacity; and AcP: soil phosphatase activity. On top, the soil properties were fitted to the ordination plots using a 999 permutations test (P values). * $P < 0.05$, ** $P < 0.01$, and *** $P < 0.001$.

was similar between fertilized and non-fertilized plots ($P > 0.05$, Figure 4a). This was also the case for spike length, but 100-seed mass was larger at fertilized than at non-fertilized plots ($P = 0.003$, df = 1, Figure 4b). At the sixth leaf stage, plants at N-fertilized plots displayed symptoms of serious P deficiency, indicated by purple leaf and obvious growth inhibition, whereas plants at non-fertilized plots did not have these symptoms (Figure 4d). The symptoms at fertilized plots were a litter lighter for previous PFM than never PFM. Although P deficiency symptoms were no longer present at the tasseling stage and maturity stage (Figure 4d), the time of the dough stage was delayed at the fertilized plot for 10−15 days.

3.4. Influence of PFM and N Treatments on Total Maize Performance and Their Relations with Soil Properties. RDA results showed that axis 1 and axis 2 together explained 91%, 88%, and 86% of the variance between soil properties and maize performance at the sixth leaf stage, tasseling stage, and physiological maturity stage, respectively (Figure 5a−c). The groups of PrevPFM and NeverPFM generally clustered together, both for N_0 and N_{135} levels. By contrast, the groups N_{135} and N_0 were positioned at opposite ends of the first canonical axis, and the data points of N_0 stood generally in the positive direction of all the maize growth parameters (except for leaf chlorophyll content and root-associated phosphatase activity) during all growth stages. Soil pH and moisture were the two most important soil factors influencing maize performance during all growth stages and were positively correlated with most crop growth parameters. Soil Olsen-P content was also a key factor for maize growth at the sixth leaf stage but did not play an important role after this period.

4. DISCUSSION

4.1. Legacy Effects of Long-Term Plastic Film Mulching. In contrast to our hypothesis, 33 years of PFM did not appear to leave a net negative legacy on maize growth and yield, although root total length and surface area were inhibited at previously mulched plots at the physiological maturity stage (Figure 3a,b). This occurred despite the high levels of macro-plastic residues (diameter >5 mm) present at the mulched plots in surface soil: plastic film residues had accumulated to 360 kg ha^{-1} or 6796 pieces m^{-2}, of which about 80% were <4 cm^2 and 20% were 4−25 cm^2 in area.[18] Plastic film residue accumulations may reduce maize yield by inhibiting root growth and development.[15,22,33] Xie et al.[34] found that the yield of maize was only decreased when the residual film amount was above 720 kg ha^{-1}. Hu et al.[22] showed that maize yield was decreased by 15−18 and 23−25%, when adding plastic film residues at 300 and 600 kg ha^{-1}, respectively. Chen et al.[33] found that the threshold when maize yield started to decrease was 180 kg ha^{-1} plastic film residues. However, all these studies were conducted by artificially adding plastic film residues to soil, in which the plastic residue is fresh and does not experience a long-term aging process. Aged plastic residues may affect crop growth less than fresh residue because it is more brittle and easy to form holes and may thus not interfere with root growth as fresh ones. Fresh plastic film residues have high tensile strength and are thus difficult to be torn, due to containing high molecular weight polymers with

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Environ. Sci. Technol. 2023, 57, 9174−9183

high hydrophobicity and semi-crystalline structures.[35] Contrastingly, aged plastic film residues after ultraviolet radiation are easily fragmented, accompanied by the formation of cracks and cavities on the mulch film surface and an increase in the crystallinity and hydroxyl index.[36] Pflugmacher et al.[37] found that the adverse effects on the germination and seedling growth of *Lepidium sativum* were reduced as a function of the aging time applied to the polycarbonate. Accordingly, we did not observe a negative legacy on maize growth and yield though the amounts of plastic film residues are close to or exceed the calculated thresholds. Similarly, a recent meta-analysis did not observe a decrease in maize yield with increasing amounts of residual films and more than half of their collected data points even showed an increase in maize yield to plastic film residue.[8]

Apart from macro-residues of the plastic film, the accumulation of film-derived microplastic reached as high as 8318 particles per kg soil in the 0−10 cm layer, 436 particles per kg soil in the 80−100 cm layer, and a total of 3.7×10^6 particles m^{-2} soil in the 0−100 cm soil profile at our mulched plots.[18] In the literature, numerous studies reported that microplastic had caused inhibitory effects on crop growth (e.g., Qi et al.[25] and Colzi et al.[26]). However, the microplastic accumulation in our experiment site seems to have no net negative impact on maize growth and yield. The reason could be that PE film-derived microplastic is not as toxic as other types of microplastic.[38] Many studies did not observe negative impact of PE microplastic on plant growth but observed the negative impact of polyvinyl chloride or polylactic acid microplastic.[25−27] This may result from the minor effect of PE plastic on the soil structure and microbial activities, as compared to polyester and polyacrylic microplastics.[39] Nevertheless, several studies observed the negative impact of PE microplastic on maize growth in pots[23] and hydroponic conditions,[40] suggesting that this explanation needs to be further verified.

On the contrary, 33 years of PFM even had a positive legacy for maize at the seedling stage, as maize aboveground biomass and root length were larger for previous PFM than for never mulching at the sixth leaf stage (Figures 2f and 3a). This may be driven by higher soil moisture for previous PFM than for never-mulching (Figure 1a and Table S1). The RDA result showed soil moisture was a key soil property controlling crop growth performance and was positively correlated with most growth parameters (Figures 5a and S1). Higher soil moisture was attributed to a higher degree of compaction of surface and subsurface soils for previous PFM than for never-PFM ($P < 0.05$, Figure S2), which slowed down water evaporation. Accordingly, we observed deeper tracks from tractors at previous PFM plots than at never-PFM plots when planting in the spring of 2021. This is supported by Sun and Ma[41] who observed film mulching promoted the movement of clay particles to the subsurface soil resulting in obvious deposition and cementation. The reason could be linked with the diurnal internal water cycle under the mulch, i.e., plastic mulch traps evaporative water, and condensed water drops underneath the mulch during the daytime can be returned to soil during the nighttime.[42] Frequent alternation of wet and dry changes the composition of soil particles, and more clay particles move and deposit with water, thereby blocking the pore space and increasing soil compaction.[41] Accordingly, we observed the lower soil porosity under the previous PFM than under the never PFM, although it only occurred at the sixth leaf stage ($P = 0.03$, Table S3). The positive legacy of previous PFM on maize growth did not occur at tasseling and maturity stages, suggesting that soil moisture was a limiting factor for maize growth only at the sixth leaf stage but not later stages.

In our study, we only measured maize morphology indices and a few physiological indices and demonstrated they are generally not influenced by previous long-term PFM. Future studies need to confirm whether maize microcosmic indices respond to long-term PFM through nutritional analysis of the produced corn, and transcriptomic, metabolomic, or proteomic analyses. Plastics can serve as carriers for other soil contaminants, such as residual metals[43] and pesticides.[44,45] It may also need to conduct crop phytotoxicity analyses (e.g., redox enzymes, lipid peroxidation), as well as soil, rhizosphere, and plant microbiome measurements in the future.

Plastic mulching is a management strategy that intentionally induces positive effects on soils; our study therefore could only assess the net effects resulting from these positive effects of the mulching practice (moisture legacy) and the presumed negative effects of plastic accumulation. Our data showed that, at least in the short term, there are no strong net negative effects on the parameters measured. Such negative effects may materialize in the future, as the positive effects subside with the absence of plastic cover, while negative effects become more apparent, for example, by increasing fragmentation of plastic to micro- or nanoplastic size. Future research should determine if there are delayed negative effects of plastic pollution that develop with time after ceasing plastic use, and also address whether the microplastics at this site have potential to become nanoplastics and impact organisms.

4.2. Impacts of Long-Term N Fertilization. In our experiment, 33 years of only N fertilization induced severe P limitation for maize growth, confirming our previous study.[13] Soil Olsen-P (available for plant) concentrations were lower at N-fertilized plots than non-fertilized plots (Figure 1e), indicating a decline of soil P supply capacity following N fertilization. Accordingly, maize root P concentrations were lower at fertilized plots (Figure 3f). To alleviate this situation, maize roots at fertilized plots secreted larger amounts of phosphatase compared to non-fertilized plots (Figure 3e). This is in line with previous studies that have shown that long-term application of N fertilizer exacerbated P deficiency.[29] Ultimately, long-term N applications reduce soil pH, which has a major impact on soil P solubility. Soil acidification following urea fertilization occurs due to the nitrification process.[46] This acidification then increases the solubility of iron and aluminum minerals,[47] which can decrease soil P availability through re-precipitation of P with free Fe^{3+} and Al^{3+} and also increase the ability of Fe and Al oxy-hydroxide minerals to strongly adsorb P by ligand exchange.[48] A 10 year N-fertilized grassland experiment also observed the increase of Al−P and Fe−P amounts with the decrease of pH.[49] In our study, although we did not measure Al−P and Fe−P, this mechanism is supported by the decrease of soil pH by about 1 unit (Figure 1b) and the increased DTPA-Fe (Table S4) following 32 years of N fertilization. This is likely occurring in our case because the pH dropped from above 6 to below 5.5, which is the pH zone in which P solubility dramatically decreases due to the increase in Al solubility.[48]

However, urea-induced P deficiency only inhibited maize growth at the sixth leaf stage (Figure 4). At this stage, maize leaves had lower chlorophyll concentration but higher flavonoid concentration at fertilized plots than at non-fertilized

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Environ. Sci. Technol. 2023, 57, 9174−9183

plots, also suggesting plant growth suffering from stress following fertilization (Figure 2c,d). In contrast, at the middle (tasseling stage) and late stages (physiological maturity stage), maize growth rates were greater at fertilized plots, indicated by its higher chlorophyll concentration at fertilized plots than at non-fertilized plots. Maize above- and below-ground biomass at fertilized plots eventually recovered to equal those at non-fertilized plots (Figures 2f and 3d). The seedling stage is the most vulnerable period when crops are sensitive to various environmental stresses.[50] At tasseling and maturity stages, maize may have multiple strategies to relieve P deficiency. For example, the difference of root-associated phosphatase between fertilized and non-fertilized plots (fertilized > non-fertilized) increased from the sixth leaf stage to tasseling and maturity stages (Figure 3e), suggesting that maize root at fertilized plots was stimulated to secrete phosphatase at later stages to increase P sources for uptake. In addition, the difference in root P content between fertilized and non-fertilized plots (fertilized < non-fertilized) increased from the sixth leaf stage to tasseling and maturity stages (Figure 3f), suggesting that maize at fertilized plots may have transferred large amounts of P from root to aboveground biomass at later stages.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.2c08878.

Additional method details for the measurements of root P, soil acid phosphatase, root-associated phosphatase, Olsen-P concentration, bulk density, total soil porosity, soil water holding capacity, detailed soil data for moisture, extractable Fe, compactness, and ANOVA and correlation test results (PDF)

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Notes

The authors declare no competing financial interest.

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