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RESEARCH ARTICLE

Assessing long term effects of compost fertilization on soil fertility and nitrogen mineralization rate

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Abstract

Background: Fertilization with organic waste compost can close the nutrient cycles between urban and rural environments. However, its effect on yield and soil fertility must be investigated.

Aim: This study investigated the long-term effect of compost on soil nutrient and potentially toxic elements (PTEs) concentration, nutrient budgets, and nitrogen (N) mineralization and efficiency.

Methods: After 21 years of annual compost application (100/400 kg N ha^{-1} year^{-1} [100BC/400BC]) alone and combined with mineral fertilization, soil was analyzed for pH, organic carbon (SOC), nutrient (total N and P, N_{min}, extractable CAL-P, CAL-K, and Mg), and PTE (Cu, Ni, Zn) concentrations. Yields were recorded and nutrient/PTE budgets and apparent net mineralization (ANM, only 2019) were calculated.

Results: N efficiency was the highest in maize and for mineral fertilization. Compost application led to lower N efficiencies, but increased ANM, SOC, pH, and soil N, and surpluses of N, P, and all PTEs. Higher PTE concentrations were only found in 400BC for Cu. Nutrient budgets correlated with soil nutrient concentration. A surplus of 16.1 kg P ha^{-1} year^{-1} and 19.5 kg K ha^{-1} year^{-1} resulted in 1 mg kg^{-1} increase in CAL-P and CAL-K over 21 years.

Conclusion: Compost application supplies nutrients to crops with a minor risk of soil-accumulation of PTEs. However, the nutrient stoichiometry provided by compost does not match crop offtakes causing imbalances. Synchronization of compost N mineralization and plant N demand does not match and limits the yield effect. In winter wheat only 65–70% of N mineralization occurred during the growth period.

KEYWORDS
nitrogen efficiency, nutrient budget, nutrient recycling, phosphorus, potassium, potentially toxic elements, soil organic carbon

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1 | INTRODUCTION

Fertilizers from recycled organic waste potentially contribute to soil fertility by improving the physical, chemical, and biological properties of soils (e.g., aggregate stability) and by reducing the need for mined or synthetically produced fertilizers, for example, derived from limited rock phosphate resources (Diacono & Montemurro, 2011) or through the Haber-Bosch process. In Germany, the main fraction of separately collected organic municipal solid waste is composted (BMEL, 2015). Undergoing different metabolic pathways leading to a degradation of the organic matter, mainly of the easily degradable compounds, the composts are a stabilized and sanitized material that can be applied on agricultural fields as organic fertilizer and as a soil amendment (Cesaro et al., 2019). As organic material is already degraded during the composting process, compost has a high content of recalcitrant organic matter (Fabrizio et al., 2009) and consequently shows a higher humus reproduction potential compared to many other organic fertilizers. It can therefore increase soil organic carbon content (SOC) efficiently (Gómez-Muñoz et al., 2017; Peltre et al., 2017). Compost application is further associated with an increase of soil microbial biomass, microbial activity, and enzyme activity (Bellino et al., 2015; Emmerling et al., 2010; Ros et al., 2006), which are characteristics associated with “healthy” soils.

Nevertheless, fertilization with compost as the main source is related to some difficulties. As organic waste, compost is a multielement fertilizer that contains considerable amounts of phosphorous (P) and potassium (K) (Bartl et al., 2002), the application of compost according to calculated nitrogen (N) demand increases the risk of P and K oversupply (Möller, 2018). Compost N has a rather low immediate N availability after application of only 5–15% of total N applied (Gutser et al., 2005; Möller & Schultheiß, 2014). However, a low short-term N effect leads to a N accumulation in the soil and consequently to a residual long-term N effect over time resulting in approximately 30% N availability (Gutser et al., 2005). Mineralization of accumulated soil organic N determines the amount of plant available N in the long term for organic waste application (Amlinger et al., 2003). There are different methods for determining N mineralization from composts. Mineralization can be determined either through incubation experiments or by determining crop N uptake in pot or field experiments (Hartl & Erhart, 2005). Compared to the estimation of mineralization in incubation experiments, the preferable approach is through determination of crop N uptake in field experiments.

When composts are applied in the long term, the issue of the accumulation of potentially toxic elements (PTE) such as zinc (Zn), nickel (Ni), and copper (Cu), among others, arises. While plant uptake of PTEs is not measured frequently when applying compost on a long-term basis, their accumulation in soil is a risk often associated with compost fertilization (Amlinger et al., 2003; Smith, 2009; Weissengruber et al., 2018).

Many reports about the short-term effects of compost application are available in previously published literature. However, assessments of the long-term effects of compost application are rare. This study provides an overview of yield, nutrient budget, and soil development in an ongoing, long-term field experiment established in 1998. The aim of the experiment was to investigate the long-term effects of different compost fertilization treatments on crop yield, the accumulation of potentially toxic elements, soil-nutrient availability, and the location-specific gross mineralization of nitrogen from spring until harvest, with the latter for one experimental year only. Further, we tested the potential for (partly) substituting mineral N fertilizer with compost. Hence, the following hypotheses were tested: (1) The long-term fertilizer N efficiency, calculated as the percentage of input recovered in the plant offtake, of a moderate mineral fertilization is much higher than with composts, resulting in higher overall dry matter (DM) yields. (2) Long-term application of municipal solid waste compost results in a higher net N mineralization due to an increase in soil N compared to common mineral fertilizer application. (3) The application of compost results in nutrient imbalances when not combined in an appropriate manner with other nutrient sources. (4) Long-term application of municipal solid waste compost leads to higher soil nitrogen, phosphorus, organic carbon, pH, and PTE (Zn, Ni, Cu) concentrations compared to mineral nitrogen fertilization.

2 | MATERIALS AND METHODS

2.1 | Experimental setup

The experimental site on the research station Heidfeldhof (University of Hohenheim) is located in Stuttgart, South Germany (48°42’56 N 9°11’37 E). The station is located 500 m asl, the mean annual temperature is 9.7°C and mean annual precipitation 735 mm. The climate is classified as warm temperate, fully humid with warm summers according to the Köppen–Geiger climate classification (Kottek et al. 2006). The soil is a loess-derived Haplic Luvisol (30% clay, 68% silt, and 2% sand). The soil depth is estimated to be 1.2 m. Soil pH is close to neutral and ranges between 6.0 and 7.5. Soil organic carbon is estimated to have been approximately 1% prior to the start of the experiment. The experiment was established in 1997 and has eight treatments with four replicates (Table 1).

There are two levels of compost application rates (100 and 400 kg compost N ha^{-1} year^{-1} [100C, 400C]), a combination of compost and calcium ammonium nitrate (CAN) as mineral N fertilizer (100C+CAN), a control treatment without fertilizer application (CTRL), a mineral fertilizer treatment (CAN), and a “depletion treatment” with 5 years of 400 kg N ha^{-1} year^{-1} compost application (between 1998 and 2003) followed by no further fertilization since 2003 (DEP). The 400 kg compost-N ha^{-1} year^{-1} treatment (400C) exceeds the German application limit of 510 kg N ha^{-1} in 3 years established in the fertilizer ordinance (DüV; BMELV, 2017) and was established to simulate a longer time span of compost application in an accelerated approach. The compost was applied before sowing (in autumn before winter wheat or in spring before spring barley and maize) and immediately incorporated into the soil, while the mineral fertilization was applied during the growing period in a split application approach. After 12 years (in 2009), the experimental design was altered to consider pH-

TABLE 1 Treatments in the long-term organic waste compost experiment with the amount of calcium ammonium nitrate (CAN) and the amount of compost applied annually and the abbreviated treatment names. CAN applications were adjusted to soil mineral nitorgen (N_{min}) after soil sampling in spring

Treatment	Abbreviation	CAN (kg N ha^{-1} year^{-1})	Compost (kg N ha–1 year^{-1})
Control	CTRL	–	–
Control + lime	CTRL+L	–	–
Depletion	DEP	–	400 until 2003
100 kg compost N	100C	–	100
100 kg compost N + CAN	100C+CAN	80	100
400 kg compost N	400C	–	400
CAN	CAN	180	–
CAN + lime	CAN+L	180	–

effects of compost application. CTRL and CAN plots were divided, and lime was applied on one half (CTRL+L, CAN+L) to allow for the differentiation of pH effects and soil nutrient effects. The experiment was designed as a split plot in a randomized block design with four blocks and 24 main plots of size 10 × 5 m. Additionally, the CTRL and CAN plots (size 5 × 5 m) were split in 2009 to allow for the liming treatment (CAN+L, CTRL+L). The ongoing crop rotation on the site is silage maize (*Zea mays* L.)–winter wheat (*Triticum aestivum* L.)–spring barley (*Hordeum vulgare* L.). Maize was harvested as a whole crop, while only grains were harvested for winter wheat and spring barley. The straw was left on the field. Tillage was usually performed at a depth between 25 and 30 cm with a cultivator or a plow and crops were not irrigated.

Winter wheat was cultivated during the vegetation period of 2019 in the 23rd year of the experiment. Compost was applied and incorporated 2 weeks before sowing in autumn. CAN with 27% N as mineral N fertilizer was applied according to the regional N fertilization recommendation and adjusted by the soil mineral N content (N_{min}) analysis in spring (21 March 2019). For the two CAN treatments fertilizer applications were split: in the CAN treatment 70 kg N ha^{-1} at tillering, 70 kg N ha^{-1} at stem elongation, 40 kg N ha^{-1} at ear emergence. For 100C+CAN additionally to the 100 kg compost N ha^{-1} in autumn, 50 kg N ha^{-1} at stem elongation and 30 kg N ha^{-1} at booting were applied as mineral fertilizer. The compost used in the experiment was organic household waste (90%) and garden waste (10%) compost from a regional composting plant. Compost properties were analyzed regularly throughout the experiment (Table 2).

2.2 | Measurements and calculations

A soil inventory was carried out in 2019 in the 23rd year of the experiment. In the following, the procedure for the sampling campaign is described.

TABLE 2 Properties of the compost (pH (10^{-2} M $CaCl_2$), dry matter concentration (DM), organic matter (OM), carbon concentration (C), total N (N), P, K, Mg, Fe, Cu, Mn, Zn and Ni concentration) used in the long-term compost experiment as mean of the last five years (2015–2019) with standard deviation (SD). (CAL = calcium acetate lactate, CAT = CAlciumchlorid and DTPA chealats)

	Method	Units	Mean	SD
pH	10^{-2} M $CaCl_2$		8.5	± 0.3
DM		%	71.2	± 1.3
OM		% DM	49.6	± 1.5
C	CN analyzer	% DM	29.1	± 1.2
C:N			13	± 0
N	CN analyzer	% DM	2.2	± 0.1
P	CAL-extraction	% DM	0.33	± 0.03
K	CAL-extraction	% DM	1.3	± 0.2
Fe	CAT-method	mg kg^{-1} DM	8211	± 0
Cu	CAT-method	mg kg^{-1} DM	49	± 13
Mn	CAT-method	mg kg^{-1} DM	277	± 0
Zn	CAT-method	mg kg^{-1} DM	156	± 6
Ni	CAT-method	mg kg^{-1} DM	14	± 1

Soil samples were taken for each layer separately at 0–30, 30–60, and 60–90 cm depth at the beginning of the vegetation period and analyzed for pH, mineral N (NH_4^+ and NO_3^-), extractable amounts of P, K, Mg, and total amounts of P, Cu, Zn, and Ni. NH_4^+ and NO_3^- was extracted using a 0.0125 M calcium chloride ($CaCl_2$) solution (VDLUFA, 2016). NH_4^+ and NO_3^- concentrations in the extracts were measured photometrically (CFA Evolution II, Alliance Instruments). Soil pH was measured according to VDLUFA (2016) in 0.01 M $CaCl_2$. C_t and N_t were analyzed using a CN analyzer (Vario MAX CN, Elementar). A subsample of six soil samples was also tested on inorganic carbon. Since none was found, it is assumed that the measured carbon is of organic origin. Soil extractable P (CAL-P) and K (CAL-K) were extracted with a calcium acetate lactate (CAL) solution (0.05 M calcium-acetate, 0.05 M calcium lactate, 0.05 M acetic acid, VDLUFA, 2016). CAL-K concentration was measured directly using a flame photometer (Elex 6361, Eppendorf). Phosphorus was determined colorimetrically (ammonium-vanadate / ascorbic acid) through spectrophotometry at 540 nm (Hitachi U-2900 Double-Beam UV-Visible Spectrophotometer). Magnesium was extracted with a 0.0125 M $CaCl_2$ solution and analyzed using an atomic absorption spectrometer at 285.2 nm (VDLUFA, 2016). Soil P_t, Cu_t, Ni_t, and Zn_t were analyzed after microwave pressure digestion with aqua regia solution (32% hydrochloric acid, 65% nitric acid, 3:2) (VDLUFA, 2011). The digested samples were analyzed through ICP-OES.

Yield data were recorded for each year, except for 2010 and 2014 due to a strong hail or mice damage. In addition, grain and straw N, P, and K concentration as well as the concentration of PTEs (Cd, Cu, Zn, and Ni) were measured regularly. For determination of N uptake, 0.33 m^2 of the crop was harvested three times in each plot, and grain as well as straw yield were measured. All plant material was dried at 60°C and above ground biomass and grain mass were determined. Dried grain and straw samples were milled and analyzed for N concentration (CN analyzer). In 2019, soil samples were taken a second time after harvest at the same location and analyzed for N_{min} (see above). The N mineralization was calculated as apparent net mineralization (ANM), which is the difference between recovered N and supplied N (Equation 1; Fink & Scharpf 2000). Recovered N is the sum of N in the harvested grain (N_G), N in the crop residues ($N_{R\ OUT}$) and soil N_{min} at harvest time ($N_{min\ END}$). Supplied N is the sum of soil N_{min} in fall before crop sowing ($N_{min\ START}$) and N in the applied mineral fertilizer (N_F):

$$\begin{aligned} \mathrm{ANM} &= N_{\mathrm{recovered}} - N_{\mathrm{supplied}} \\ &= \left(N_G + N_{\mathrm{ROUT}+}N_{\mathrm{min}\,\mathrm{END}}\right) - \left(N_F + N_{\mathrm{min}\,\mathrm{START}}\right). \end{aligned} \tag{1}$$

To investigate the N efficiency in the long term, measured data of N application and N offtake throughout the trial were used to calculate the N efficiency by dividing N offtake by N input (Equation 2).

$$\mathrm{N\ efficiency}\ \ (\%) = \frac{\mathrm{N\ offtake}}{\mathrm{N\ input}} \times 100\%. \tag{2}$$

Based on the recorded data, input-output budgets for N, P, K, Cd, Cu, Zn, and Ni were calculated including the entire experimental period of 21 years. As inputs the nutrients and PTE provided by the fertilizer were considered, and as outputs the crop offtake by harvested biomass. Crop offtake was calculated by multiplying the recorded yield data with the measured nutrient and PTE concentration. When measurements were not available (2004, 2010, 2011, 2014) the mean for the crop and treatment of the other years was used. The treatments with liming were not included into the calculations of N efficiency and nutrient budgets since they only were established in 2009.

2.3 | Statistical analysis

Statistical analysis was performed using the R programming environment for statistical computing (R Core Team, 2018). A linear mixed effect model with main plot as the random effect was set up and checked for variance homogeneity and normal distribution. An analysis of variance was performed, and means were compared with a Tukey-test at a significance level of $\alpha = 0.05$.

3 | RESULTS

3.1 | Yield and element concentration in the biomass

The yield effect of the treatments covers 2003 (after establishing the DEP treatment) up to 2019. The average yield of each treatment was set relative to the average yield of the mineral fertilized treatment. The average fresh matter yield for the mineral fertilizer treatment was 35400 kg ha^{-1} for maize, 7560 kg ha^{-1} for winter wheat, and 4260 kg ha^{-1} for spring barley. The effects on relative yields of treatment, crop, year of application, and the interaction of treatment:crop and treatment:year were analyzed in a linear model (Table S1). Since there was a significant interaction between the crop and the treatment, the effect of the treatments on the relative yields of crops are shown separately. Yet, the unfertilized CTRL treatment resulted in lowest yields across all crops (Figure 1a; Table S2). In maize, the 400C and 100C+CAN treatment resulted in yields higher than mineral fertilization alone. On average, the relative yield of the 400C treatment was 27% higher compared to the CAN treatment. The treatments 100C and DEP resulted in similar relative yields as in the CAN treatment, even though there was no fertilizer application for 15 years in the DEP treatment. In spring barley, the yield level almost followed the fertilizer N application rates (400C > CAN > 100C + CAN > 100C > DEP > CTRL). In winter wheat, the pattern was similar with the exception that the DEP treatment resulted in higher relative yields than the 100C. The yields of 400C and CAN were significantly higher than all other treatments for both crops, with the exception that the yield of 100C+CAN was not significantly different from CAN and 400C in spring barley.

Significant changes in yields over time (2003–2019) could only be observed for the DEP treatment, where the yields decreased over time (Table S1, Figure S1). However, in the first 5 years (1997–2003) of the trial, a significant increase in relative yields in the 400C treatment was observed (Figure S2).

The N concentration of the harvested product was significantly affected by the treatment, crop species, application duration as well as the interactions treatment:crop species and treatment:application duration (Table S1). Especially maize and winter wheat showed differences in measured N concentration while there were few differences for spring barley (Figure 1b). In winter wheat, the mineral fertilization (CAN and 100C+CAN) showed significantly higher concentrations than the compost treatments, while in maize, 400C, 100C+CAN, and even the DEP treatment resulted in similar or even higher N concentrations compared to CAN. Throughout the experimental years, the N concentration of the harvested product decreased as a general trend. However, the decrease was dependent on treatment and crop species (Figure S3)

For N offtake, the linear model revealed, similar to its two components, that the three main influences, treatment, crop, and application duration, but also the interactions treatment:application duration and treatment:crop were significant (Table S1). Therefore, the treatment effect was analyzed again for each crop separately (Figure 1c). The results of the statistical analysis are very similar to the overall results across all crops, besides for winter wheat. In winter wheat, no significant difference of N offtakes among the treatments 400C and CAN and 100C+CAN was observed. The effect of the duration of the experiment was also dependent on crop and treatment, but a general trend revealed a decrease of N offtakes (Figure S4).

Focusing solely on the long-term effects after 21 years of trial duration (winter wheat cropping season in 2019), the highest yields were achieved with the mineral fertilizer treatments (9.9 Mg ha^{-1} for CAN+L and 9.4 Mg ha^{-1} for CAN) (Figure 2). The yield of the 400C treatment was 17% lower, but with 8.2 Mg ha^{-1} the 400C treatment did not differ significantly from the CAN treatment. The 100C treatment

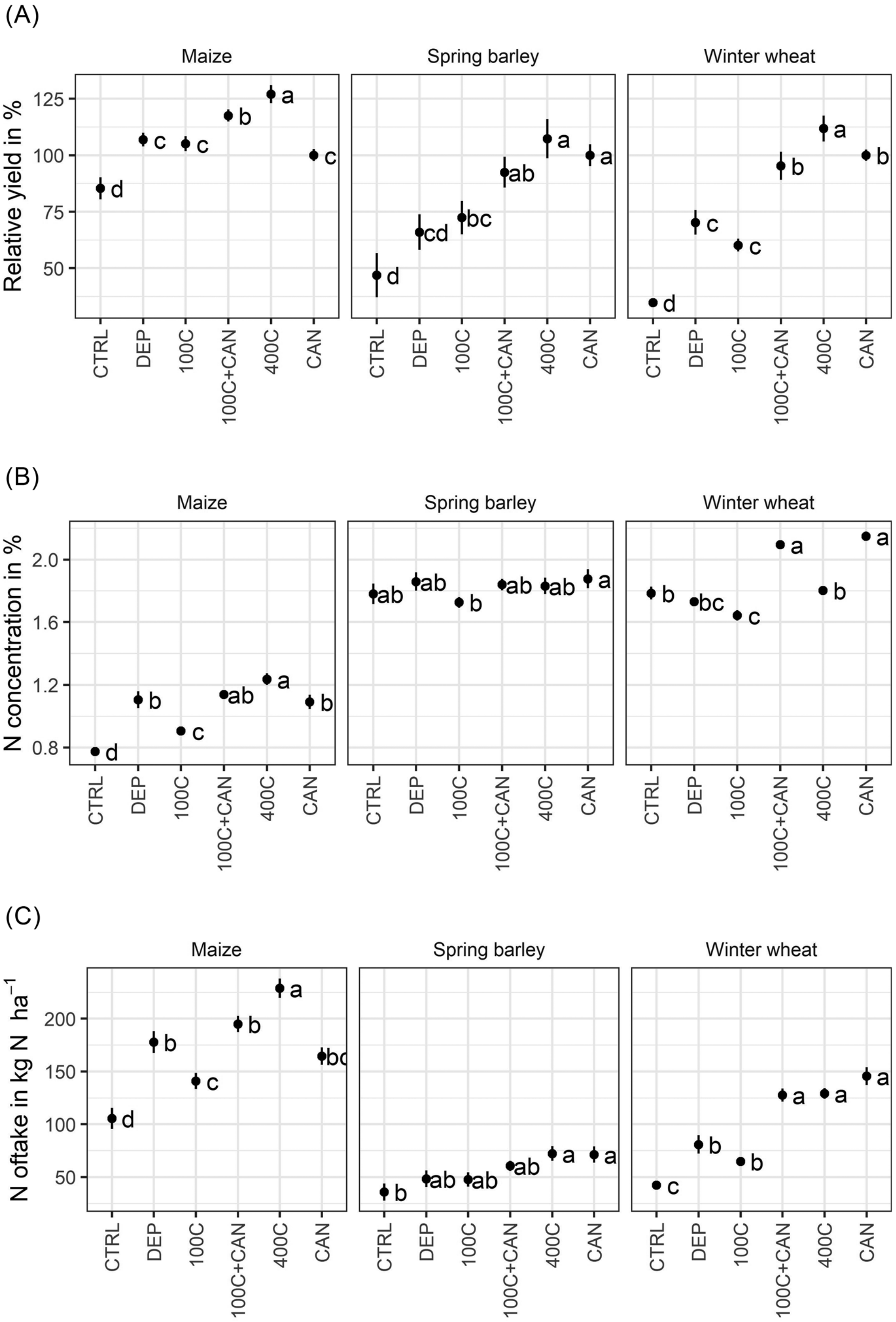

FIGURE 1 (a) Mean yield of maize ($n = 144$), spring barley ($n = 72$) and winter wheat ($n = 96$) from 2003–2019 relative to the mean yield of maize (35.40 Mg FW ha^{-1}), spring barley (4.26 Mg FW ha^{-1}) and winter wheat (8.10 Mg fresh matter ha^{-1}) of the CAN treatment. (b) N concentration in percentage of the harvested product (whole crop for maize, grain for spring barley and winter wheat). Dots represent the mean and the lines the standard deviation. Letters indicate significant differences within treatments ($\alpha = 0.05$ Tukey-test). For abbreviations, see Table 1.

showed yields 50% lower compared to mineral fertilization. Combining compost and mineral fertilizer (100C+CAN) resulted in lower yields (60%) than the application of equivalent amounts of N as mineral fertilizer (CAN+L). The additional mineral fertilization in the combination treatment (100C+CAN) did not result in significant higher yields compared to the 100C treatment, which received only compost. Omitting any fertilizer resulted in the lowest yields (2.6 and 2.7 Mg ha^{-1}). Yield of the DEP treatment (3.7 Mg ha^{-1}) with compost application 16 years ago was in between the yield of the controls and the 100C treatment.

Available data on the nutrient composition of harvested biomass indicate no clear treatment effects on the K concentration. Regarding the P concentration, a significant interaction between the treatment

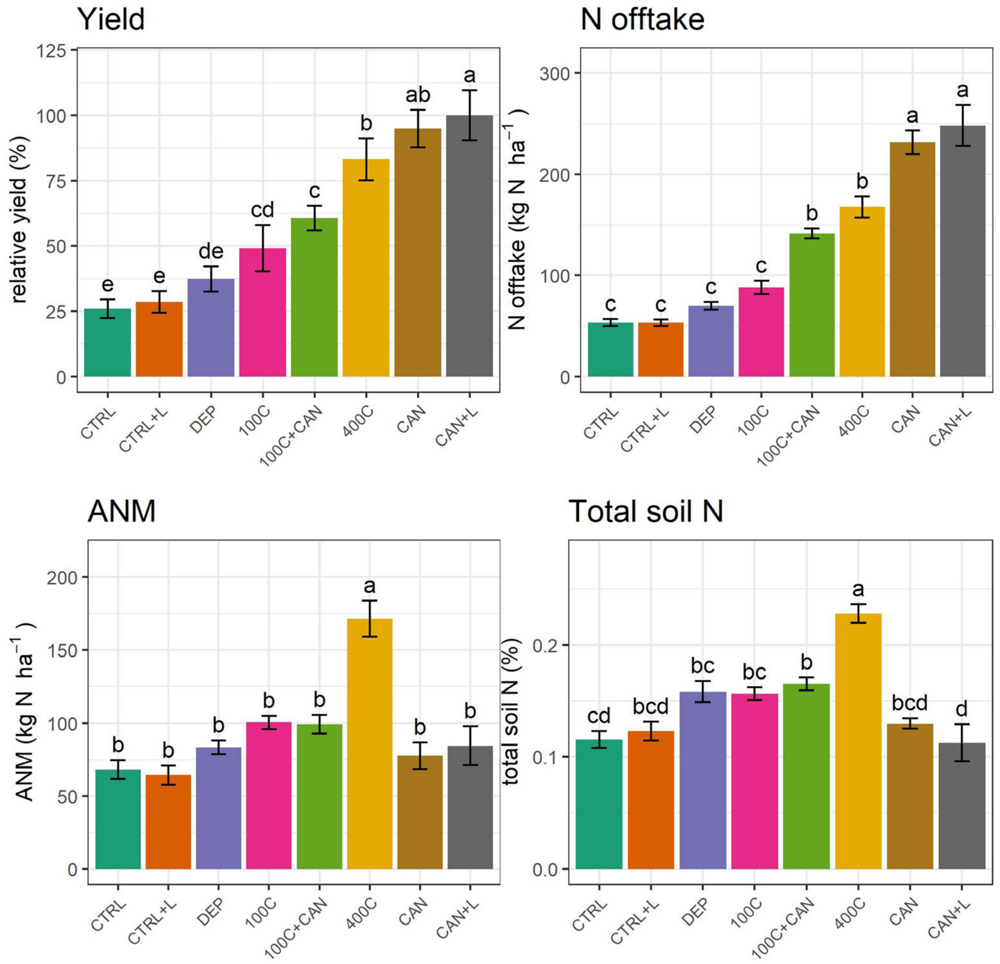

FIGURE 2 Wheat yield (%) relative to the yield of the CAN+L treatment (9.9 Mg ha^{-1}), wheat N uptake (kg N ha^{-1}), apparent N mineralization (kg N ha^{-1}) and soil N concentration (%) in 2019. Bars show the mean and lines the standard deviation. Letters indicate significant differences among treatments ($\alpha = 0.05$ Tukey-test). For abbreviations, see Table 1. ANM = apparent nitrogen mineralization

and the crop was detected. In winter wheat and spring barley, no effect of treatment was detected while for maize higher P concentration was observed for DEP than for the CTRL treatment (Table S3).

Available data on PTE concentration in the harvested products also indicate only a minor effect of treatment. For copper (Cu) and cadmium (Cd) no effect of treatment was detected. For zinc (Zn) significant effects of the three main factors treatment, crop, and year but also for the interactions between treatment and year as well as the interaction year and crop were found. When comparing the measured Zn concentration for each year, there is no clear effect that indicates that compost application increases the Zn concentration (Table S4). However, it needs to be noted that measurements were only available for some of the cropping years.

3.2 | Nutrient and PTE budgets

Based on the measured yield data and measured nutrient and PTE concentrations in the harvested products as well as in the fertilizers, nutrient budgets for N, P, K, and the PTEs Cd, Cu, Ni, and Zn were calculated (Table 3). The accelerated treatment (400C) was established to assess long-term effects on soil. Since four times the usual amount of compost was applied, the resulting budgets are not applicable to practical farming conditions. Therefore, the accelerated treatment is excluded from further discussion. The moderate compost treatments (100C, 100C+CAN) resulted in surpluses of N and P, and PTEs while the other treatments (CAN, CTRL, DEP) resulted in a deficit of all nutrients. For N, the 100C+CAN treatment shows higher surpluses than 100C since the added N from mineral N fertilization was lower than the higher N offtake due to higher yields. However, the additional N from the mineralization resulted in slightly lower budgets in 100C+CAN for all other nutrients and PTEs, even though the differences were not significant. Sole mineral N fertilization resulted in a slightly negative N budget, as well as the highest deficits for all other nutrients and PTEs. The control and DEP treatment do not differ for the nutrients, even though there were high compost applications in the first 5 years. Yet, the high inputs of PTEs in the first 5 years, still resulted in an average surplus of PTEs after 18 years of no application. The high variance for

TABLE 3 Effects of long-term mineral nitrogen fertilizer and compost application on the nutrient (N, P, K) budgets (kg ha^{-1} $year^{-1}$) and budgets of potentially toxic elements (Cd, Cu, Ni, Zn) (g ha^{-1} $year^{-1}$) as treatment means with standard deviation (std) from 1997–2019. Letters indicate significant differences among the treatments ($\alpha = 0.05$ Tukey-test). For abbreviations, see Table 1

	N			P			K		
Treatment	**Mean**	**std**		**Mean**	**std**		**Mean**	**std**	
DEP	−67	197	d	−8	41	c	−54	178	c
CTRL	−66	50	d	−13	11	cd	−58	77	c
100C	17	49	c	5	10	b	−3	83	b
100C+CAN	60	102	b	1	10	b	−14	87	b
400C	297	65	a	64	18	a	189	114	a
CAN	−6	45	c	−20	9	d	−78	79	c

	Cd			Cu			Ni			Zn		
Treatment	**Mean**	**std**		**Mean**	**std**		**Mean**	**std**		**Mean**	**Std**	
DEP	0.9	3.3	b	188	557	c	55	145	b	436	1,610	b
CTRL	−0.3	0.4	c	−26	26	d	−3	5	c	−120	94	c
100C	1.5	0.7	b	292	50	b	68	17	b	693	179	b
100C+CAN	1.4	0.7	b	276	56	bc	67	16	b	654	173	b
400C	7	2.3	a	1,263	164	a	284	63	a	3,169	578	a
CAN	−0.4	0.5	c	−40	25	d	−4	5	c	−173	78	c

the DEP treatment was related to the high inputs in the first 5 years, followed by a period without any application.

3.3 | Long-term nitrogen efficiency

The long-term nitrogen efficiency, as the percentage of N offtake from N input, was calculated for all treatments with annual N application (100C, 100C+CAN, 400C, CAN) for each year and cumulative over the whole trial period for all treatments (besides CTRL). Over the whole trial period, the nitrogen efficiency was highest for CAN with 105%, which implies a higher N uptake than N input. Second highest was found for the DEP (86%). For the compost treatments, the N efficiency decreased with increasing N application rate from 81% for 100C, to 67% for 100C+CAN and 34% for 400C. The N efficiency depended on treatment and crop, however there are significant interactions between treatment and crop, as well as duration of application (Table S5). Therefore, the combination of crop and treatment with each other were compared and the effect of application duration for each treatment:crop subset was checked (Figure 3). The model was able to explain more than half of the variance within the data (adjusted R^2 = 0.63). Maize showed the highest N efficiency compared to spring barley and winter wheat. For maize only the 400C treatment resulted in a significantly lower nitrogen efficiency, while in winter wheat and spring barley CAN showed a significantly higher N efficiency than the compost treatments. With respect to changes over time of application, higher values with duration of application were only observed in maize with the 400C and 100C+CAN treatment. Contrastingly, 100C+CAN treatment for spring barley and the CAN treatment for winter wheat resulted in a significant decrease over time.

3.4 | N mineralization dynamics after long-term application

Data obtained in the year 2019 after 21 years of compost application indicated that the 400C treatment had the highest soil N_t concentration (0.23%; Figure 2) in the upper soil layer (0–30 cm). Similarly, the compost application in the 100C+CAN treatment showed higher soil N_t concentration (0.16%) than the control (0.12%). The treatments DEP and 100C did not significantly differ from the control, but from the CAN+L treatment, which was the treatment with the lowest soil N_t concentration (0.11%).

A similar pattern was observed for the apparent N mineralization (ANM (Figure 2). In fact, the ANM correlated strongly (Pearson's R of 0.79, $p < 0.001$) with the total N content in the soil. With an amount of 171 kg N mineralized ha^{-1}, the 400C treatment had significantly the highest mineralization. The N supply level was more than doubled compared to mineral fertilization. The other treatments did not differ significantly from each other due to a high variance. However, a pattern can be observed. The treatments with the lower compost application rate (100C and 100C+CAN) resulted in slightly higher ANM compared to the control and CAN treatments (approx. 25 kg N ha^{-1} higher). The treatments CTRL and CTRL+L without fertilizer application had the lowest mineralization rate, which however added up to about 65 kg N ha^{-1}.

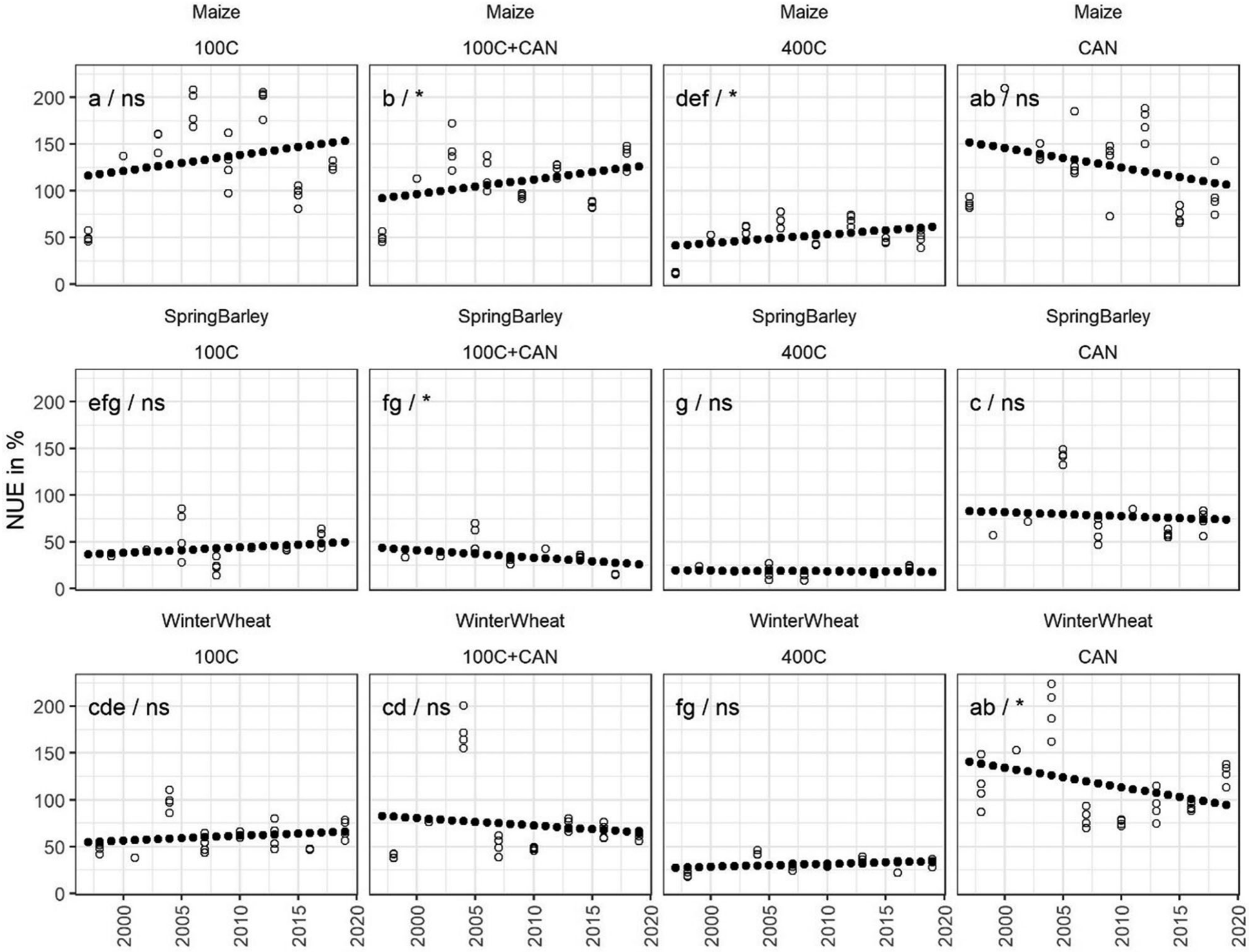

FIGURE 3 Nitrogen efficiency (N offtake / N input × 100) by crop and treatment over duration of the experiment. Letters represent significant differences between crop and treatment combinations, stars show if the year effect is significant within the crop- treatment combination (n.s.: $p > 0.05$, *: $p < 0.05$, **: $p < 0.01$, ***: $p < 0.001$). The adjusted r^2 was 63% for the full model. Hollow dots represent measured values, while filled dots represent the model prediction. For abbreviations, see Table 1.

The lowest N concentration in the harvested wheat was found in the CAN and control treatments, while all the compost treatments showed higher N concentrations, some of the differences were significant (Figure 2). The N offtakes were related to yields rather than to the ANM or soil N_t (Figure 2). In 2019, mineral fertilization (CAN and CAN+L) resulted in the significantly highest N offtake compared to all other treatments. Further, the 400C and 100C+CAN treatment showed significantly higher N offtakes compared to the unfertilized control treatments. Wheat N offtake in the 100C and the DEP treatment was much lower and not significantly higher than in the unfertilized control treatments. However, in wheat N offtake in the 100C treatment in comparison to the control treatment was approx. 35 kg N ha^{-1} higher and in the DEP, it was still 17 kg N ha^{-1} higher.

Soil N_{min} changed throughout the cropping season of winter wheat (autumn 2018–spring 2019) and the pattern depended on the treatments (Figure 4). In autumn 2018, especially the CAN+L showed a higher soil N_{min}, but also the CAN and the 400C treatment had a higher N_{min} than the other compost treatments and the CTRL. During winter, the soil N_{min} in the CAN+L treatment decreased more strongly resulting in similar spring N_{min} level as in the CAN and the 400C treatment. During the period between spring sampling until harvest, soil N_{min} for the CAN and CAN+L treatment decreased strongly on a similar level as in the CTRL treatments. In all other treatments, soil N_{min} at wheat harvest was higher than in spring. Consequently, the 400C treatment had the highest N_{min} level after harvest. Soil N_{min} in the other treatments with compost application did not differ to CTRL and CAN treatments.

3.5 | Effects on soil fertility and PTE accumulation

Most of the characteristics measured in the topsoil layer (0–30 cm depth) from sampling in the year 2019 were significantly different among treatments (Table 4). However, no significant effects of fertilizer and compost application on the soil properties were observed in the soil layers 30–60 and 60–90 cm, except for pH (Table S6). In general, treatments with compost application showed higher soil pH, SOC, total and extractable CAL-P, CAL-K, and Mg, as well as total Cu in the soil

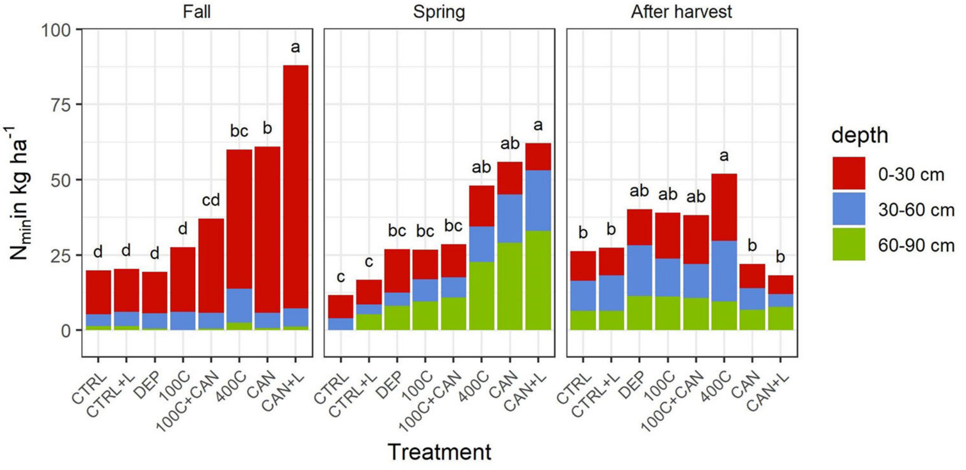

FIGURE 4 Soil mineral N (N_{min} in kg ha^{-1}) in 0–90 cm soil in fall 2018, spring and after harvest 2019. Different letters indicate significant differences among treatments ($\alpha = 0.05$ Tukey-test). For abbreviations, see Table 1.

TABLE 4 Soil pH, soil C_{org}, soil total phosphorus (P_t), CAL extractable phosphorus (CAL-P), share of CAL-P of total P, CAL extractable potassium (CAL-K) and $CaCl_2$-extractable magnesium (Ext. Mg), total copper (Cu_t), total zinc (Zn_t) and total nickel (Ni_t) concentration in 0–30 cm of soil in 2019. Letters indicate significant differences among treatments ($\alpha = 0.05$ Tukey-test). For abbreviations, consult Table 1

		CTRL	CTRL +L	DEP	100C	100C +CAN	400C	CAN	CAN +L
pH		6.4^{bc}	6.4^{bc}	6.4^{bc}	6.7^{b}	6.7^{b}	7.0^{a}	6.3^{cd}	6.5^{d}
SOC	%	1.0^{de}	1.0^{de}	1.3^{b}	1.3^{b}	1.4^{b}	2.0^{a}	1.1^{cd}	1.0^{e}
total N	%	0.12cd	0.12cd	0.16c	0.16c	0.17b	0.23a	0.13cd	0.11d
P_t	mg kg^{-1}	1018^{c}	1039^{c}	1250^{ac}	1262^{ac}	1269^{ac}	1489^{a}	1148^{c}	1315^{ab}
CAL P	mg kg^{-1}	72^{cd}	79^{cd}	97^{c}	105^{b}	91^{cd}	155^{a}	66^{d}	65^{d}
Share ext P/P_t	%	7.1	7.6	7.8	8.3	7.2	10.4	5.7	4.9
Ext. K	mg kg^{-1}	136^{cd}	140^{cd}	178^{c}	203^{b}	196^{b}	393^{a}	132^{d}	124^{d}
Ext. Mg	mg kg^{-1}	115^{ab}	109^{b}	117^{ab}	116^{ab}	124^{ab}	135^{a}	117^{ab}	106^{b}
Cu	mg kg^{-1}	28^{b}	31^{ab}	30^{ab}	29^{ab}	29^{ab}	33^{a}	30^{ab}	28^{b}
Zn	mg kg^{-1}	83^{a}	94^{a}	94^{a}	89^{a}	92^{a}	105^{a}	86^{a}	89^{a}
Ni	mg kg^{-1}	43^{a}	44^{a}	39^{a}	38^{a}	40^{a}	39^{a}	41^{a}	40^{a}

compared to CTRL and CAN, while levels of total soil Zn and Ni were not significantly different in the upper soil layer.

The highest pH was measured in the 400C treatment, followed by the 100C and the 100C+CAN treatments. The soil pH of treatments with lime application was similar to respective values of the control treatment as well as the fertilizer treatment.

The accelerated compost application rate also showed the highest SOC, which was doubled compared to the control and the mineral fertilized treatments. The 100C and 100C+CAN treatment had the second highest SOC concentrations, which was also significantly higher than the corresponding concentrations of the control and the mineral fertilized treatments. SOC concentration of the DEP treatment was also higher compared to CTRL and CAN. The treatments 100C, 100C+CAN, and DEP treatment showed SOC concentrations approx. 0.3% higher than the untreated as well as the mineral fertilizer control. In case of mineral fertilization, the additional lime application in the CAN+L treatment resulted in a lower SOC concentration than in the CAN treatment without lime.

The accelerated compost application showed significantly higher P_t concentration compared to the mineral fertilized treatments and the control treatments (Table 4). The control treatment with lime application had the second highest P_t concentration, which was significantly higher compared to the CTRL treatment without lime application. The other treatments with compost application (100C, 100C+CAN, DEP) showed an elevated P_t concentration compared to the control treatment without lime and the mineral fertilized treatments (CAN, CAN+L). However, the differences were not significant.

The treatment with the higher compost application rate showed the highest CAL-P concentration, approximately 50 mg kg^{-1} soil higher compared to the 100C treatment and almost 100 mg kg^{-1} soil higher than treatments with mineral fertilizer application (Table 4). The treatment 100C had the second highest CAL-P concentration followed by the DEP and the 100C+CAN treatment. However, differences to the mineral fertilized and the control treatments were not always significant. Mineral N fertilizer application resulted in the lowest CAL-P concentration. CAL-P concentrations of the control treatments were slightly higher than in the mineral fertilized treatments. Treatments with compost application showed higher CAL-P concentrations.

There were significant differences for CAL-K concentration in the upper soil layer (Table 4). The higher compost application rate resulted in the significantly highest CAL-K concentration. CAL-K concentration was tripled compared to the mineral fertilized and control treatments and doubled compared to the treatments with lower compost application rate and the depletion treatment. Input of 100 kg compost N ha^{-1} year^{-1} alone and in combination with mineral fertilizer resulted in the second highest CAL-K concentrations, which differed significantly from the unfertilized control and the mineral fertilized treatments. CAL-K concentration of the control treatments was slightly higher than that of treatments with mineral fertilization. The treatments with no compost application showed generally lower CAL-K concentrations.

Extractable Mg concentration was slightly higher with the higher compost application rate and differed significantly from both mineral fertilized treatments (Table 4). All the other treatments showed similar extractable Mg concentrations.

The treatments with compost application showed significantly higher soil Cu concentration but did not differ regarding the soil Ni and Zn concentration (Table 4). The highest soil Cu concentration was measured in the treatment with the higher compost application rate. However, it differed significantly only from the CTRL and the CAN+L treatments, which had the lowest Cu concentration. The other two treatments receiving compost (C100, 100C+CAN) did not differ in their Cu concentrations.

4 | DISCUSSIONS

4.1 | Effects on yield and N efficiency

The data of this study partly support the first hypothesis, which stated that long-term fertilizer N efficiency and hence DM yields of a moderate mineral fertilization are much higher than with composts application. The higher yields of the CAN treatments in comparison to the 400C treatment despite much lower overall supply of the major nutrients N, P, and K showed that the crop N supply is the major driving factor influencing crop yields in present experiments. Consequently, the long-term fertilizer N efficiency, calculated as the percentage of input recovered in the plant offtake, of a moderate mineral fertilization (CAN) is approximately 50% higher than the regular compost treatment across the three crops. However, the data also demonstrate that the effect of fertilization is highly dependent on the crop, as indicated by significant interactions between the factors fertilizer treatment and crop (Table S1). Differences in yield among mineral and compost application decreased in the order winter wheat–spring barley–silage maize. The low differences obtained with maize can be related to two factors: the steepness of the N production function and the overall growing pattern of maize. It is well known that the steepness of the N production functions of these crops follows this order (e.g., Heyn, 2018). The growing period of maize fits better with the mineralization pattern of compost due to a longer cropping season in summer and autumn. Furthermore, maize is sown in spring, which enables compost application in spring and direct soil incorporation before ploughing or seedbed preparation. Thus, the compost N is not subjected to leaching over winter as in comparison to winter wheat. Therefore, maize is better adapted to a compost fertilization than winter wheat or spring barley. However, the data on maize performance indicated that other factors than N have influenced growth of maize crops, as the regular compost application (100C) showed similar fresh matter yields as the CAN treatment (180 kg N), while the N concentration in the biomass and the resulting N offtakes were significantly higher in the CAN treatment (Figure 1a–c). Simultaneously, the 100C+CAN treatment (a total of 180 kg N) showed significantly higher yields and similar N concentrations as the CAN treatment. Data on soil CAL-P and CAL-K in the CAN treatment (Table 4) were higher than the optimal range recommended in Germany (Baumgärtel et al., 1999; VDLUFA, 2018). Additionally, the lack of differences of the P and K concentration in the harvested products indicates that other nutrients than P and K, provided by the compost application but not by CAN, are probably the limiting factors for the yield level in the CAN treatment (e.g., sulfur or alternative micronutrients). It seems that maize is more affected by these factors than the other two crops spring barley and winter wheat.

Contrary to the expectations, the relative yields of the compost treatments did not increase over the whole experiment duration, while the N efficiency only increased over time for the low compost application rates in maize (Figure 3). We assumed that with increasing time, the long-term N mineralization would increase overall N mineralization and the yields. However, there was an increase in yields for the high compost application within the first 5 years of the trial, which suggests that over the years a new equilibrium was achieved. Consequently, further high applications do not further enhance crop growth. The comparison of the N offtakes and N balances of the 400C and the DEP treatment provide some indications that the long-term effects of compost application on the N availability and productivity are in the range of 15–25% of the short-term effects. This mean that approx. 80% of the compost effects on N availability and productivity are achieved on a short-term perspective, while the longer lasting effects accounted for approx. 20% of the effects. These findings are in line with Pang and Letey (2000) who stated that organic fertilizers need only two or few more years to achieve their full potential due to carry over effects of unmineralized organic materials and increasing soil organic N and thereby increasing N mineralization potential. Gómez-Muñoz et al. (2017) on the other hand argued, that the N efficiency of carbon rich organic fertilizers decreases with time through substantial N immobilization due to a wide C/N ratio of the fertilizers. This should not have been the case in this study, since the compost used in the experiment was relatively mature, low in carbon, and thus rather stable. Therefore, we expect rather a slow N mineralization than an N immobilization.

Compost fertilization resulted in higher wheat yield in 2019 compared to the unfertilized controls. The observed effect agrees with other compost experiments and can be explained by the effect of compost on soil fertility, especially the increased N mineralization potential (Gómez-Muñoz et al., 2017; Lehtinen et al., 2017). However, compost fertilization did not result in crop yields as high as mineral fertilization, although a much larger N amount was applied in the 400C treatment. The 100C treatment received less N with fertilizers than the 100C+CAN and CAN treatments (180 kg N ha^{-1} corrected for mineral soil N in spring mineral soil N 150 kg N ha^{-1}, 120 kg N ha^{-1} CAN was applied, respectively). This explains the considerably lower yield, showing that the availability of N is a major driving factor of the observed yield differences among the treatments.

There are several indications that compost application resulted in a low overall N efficiency, in part due to a lack in the synchronization between plant N demand and soil N release. For example, spring soil mineral N in 2019 indicated that long-term compost application increased the soil mineral N in spring by approx. 10% of the yearly applied N amounts (Figure 4). The data on soil mineral N after harvest in 2019 also indicated that a similar share of the nitrogen is mineralized late in the season when cereals are not further able to take up soil N (Figure 4). A similar increase in soil mineral N was also observed in autumn of the year before. The higher protein concentration in winter wheat grains in 2019 in the compost treatments also provides strong indications that compost N was mineralized during summer rather late in the growing season (Figure 2). This analysis is supported by a study of Erhart et al. (2005). They analyzed yield components of cereals in compost treatments and compared them to mineral fertilizer treatments. They concluded that compost supplied sufficient N during early growth and after flowering, but not during the stem extension phase in spring when wheat N uptake is highest. Crops with extended growing periods in summer like maize or potentially late maturing potatoes can usually benefit more from compost fertilization (Lehtinen et al., 2017), as shown by the long-term N efficiency and yield effect in our study.

The timing issue is further underlined by the results of soil mineral N content sampling (N_{min}). The increased spring N_{min} soil levels in compost treatments indicated a higher mineralization potential compared to the control. Similarly, Erhart et al. (2005) and Tits et al. (2014) detected higher spring N_{min} soil levels after compost application. After sampling in spring during the main vegetation period, wheat takes up most of the N and N mineralization increases due to higher temperatures (Robertson & Groffman, 2007). Over the cropping period, all treatments except the mineral fertilized ones showed an increase in N_{min}, which means more net N mineralization occurred than N was taken up. That N was not taken up by wheat indicates that mineralization occurred during a time when wheat N demand declined due to the generally emerging senescence of the crop. The high mineral N in the soil is then prone to leaching over winter and will thus be prone to be lost from the system and may impact the environment. In contrast, the CAN treatments showed a lower level of N_{min} compared to spring, which shows in combination with the data on crop N offtakes that fertilizer N and mineralized N was efficiently taken up. The high values in autumn of 2018 can be explained by a low uptake of N in the 2018 cropping season due to a major drought phase.

Lehtinen et al. (2017) observed crop yields in compost treatments as high as in mineral fertilized treatments in maize, winter wheat, and peas. In the long-term experiment of Gómez-Muñoz et al. (2017) only the accelerated compost treatment (900 kg N ha^{-1} year^{-1}), but not the moderate compost rate (300 kg N ha^{-1} year^{-1}), had similar oat yields compared to the mineral fertilized treatment. The combination of organic and mineral fertilizer sources did not influence the overall N efficiency and mineralization pattern, as indicated by results in Figures 2 and 3. This contradicts findings of Bedada et al. (2016) and Lehtinen et al. (2017). They observed a more distinct increase in yields when composts were applied in combination with mineral N fertilizers compared to sole compost application. Bedada et al. (2016), however, investigated the effect on a soil with low nutrient supplies and Lehtinen et al. (2017) used higher applications of mineral N, which could explain the different results. Input of mineral N fertilizers can potentially decrease compost N immobilization and consequently improve net mineralization (Gutser et al., 2005). N efficiency, on the other hand, is lower for high applications of N. To conclude, the yield effect of compost is variable, but lower when compared to mineral fertilizer. The yield effect of compost application is increasing with increasing application rate and for more coarsely textured and nutrient depleted soils (Körschens et al., 2013). Thus, the loamy texture and the relatively fertile soil in this experiment may explain the lower yield effect of compost in this experiment compared to other studies.

4.2 | Nitrogen dynamics after long-term application

As stated in hypothesis two, 21 years of compost fertilization increases N mineralization by about 20 kg N ha^{-1} for 100C (approx. 20% of the applied compost-N) and 100C+CAN and by 90 kg N ha^{-1} for 400C (approx. 25% of the applied compost-N). Simultaneously, soil N_t concentration increased by about 0.03% for 100C and 100C+CAN and by 0.10% for 400C compared to the mineral fertilization. The comparison of N_t in 2008 and 2019 further suggests a build-up of N_t with application of 400 kg compost N. In accordance with our findings, Emmerling et al. (2010) and Lehtinen et al. (2017) detected higher N_t concentrations in the soil after 10 years of compost application compared to unfertilized and mineral fertilization treatments. Gomez-Muñoz et al. (2017) measured significantly higher N_t concentration in the soil for compost and accelerated compost treatments after 11 years of application. Compared to other organic wastes, compost has a larger fraction of recalcitrant organic N, which is not immediately plant available and accumulates in the soil (Peltre et al., 2017). Most of the nitrogen accumulation by compost application is bound to organic matter (Tits et al., 2014). The higher N_t concentration in the DEP treatment compared to control and mineral fertilizer treatments underlines the slow release of N from compost after application.

The increase of soil N_t concentration is closely linked to an increased ANM. Application of 400 kg compost N with an ANM of 172 kg N ha^{-1}

should have overall met the wheat N demand of 180 kg N ha^{-1}, while treatments with lower compost amount showed lower ANM. Present data indicate an increasing net mineralization with higher application rates (Figure 2). Probably the mineralization rates increased since larger compost application rates saturate the capacity of the soil to protect the supplied organic matter from decomposition, a process described by Six et al. (2002), resulting in higher overall N mineralization rates. However, a four times higher compost application rate did not even double ANM (100BC: 100 kg N ha^{-1} vs. 400BC: 172 kg N ha^{-1}). The relation is clearly not proportional, which underlines that high application rates of compost are not input-efficient regarding not only the N use efficiency, but also regarding the accumulation of organic matter in the soil.

After long-term application of organic fertilizers with low short-term N release, such as compost, Gutser et al. (2005) estimated on a long-term perspective an annual N effect of 31% of the regularly applied N amount. Results of presented study provides evidence that the N effect depends on the application rate. Correcting the ANM of the compost treatments for the ANM of the control treatment, the data indicated a higher share of applied N is mineralized in 100C (32%) than in 400C (26%). Yet, the values are in the range of long-term fertilizer efficiencies reported for urban composts of 20–40% (Amlinger et al., 2003; Krauss, 1997; Gutser & Claasen, 1994; Gutser et al., 2005). Furthermore, data about the effects of compost applications on apparent N mineralization and soil mineral N in spring, after harvest and in autumn indicate that 65–70% of this N is mineralized during the wheat growing cycle, and 30–35% after harvest in late summer and autumn. Approx. 10% of the long-term compost N is mineralized during winter before start of the plant growth in spring, another 10% during the growing period of wheat, and another 10% after ripening of the winter wheat crop.

4.3 | Effects on nutrient and PTE budgets

Compost is a multinutrient fertilizer. The stoichiometry of the main nutrients in compost, however, does not match the nutrient stoichiometry of plant offtakes, as shown by elemental budgets far away from zero in Table 3. This matches earlier statements (Tittarelli et al., 2017; Möller, 2018). A fertilization strategy that utilizes compost as the main N input therefore inevitably leads to an oversupply of other nutrients, especially P (Zikeli et al., 2017). The results of this study support hypothesis three, that compost application results in imbalances if not compensated with additional nutrient sources. Data presented in Table 3 indicate that the 100C+CAN treatment are nearly balanced in terms of P, but they showed some K deficits. Therefore, a K fertilizer free of P is needed to balance out this unbalance in the long term. Compost is often applied in order to meet N demand of crops. Also, in field experiments in most cases the compost application rates are determined by the amount of N. However, in an optimized fertilization scheme with compost, the overall compost application rates should be determined by the P inputs in relationship to the offtakes of the whole crop rotation. To meet the demand of other nutrients like N, K, or S additional sources free of P are required to achieve balanced systems. Furthermore, an optimized fertilization strategy in conventional farming would apply compost mainly to summer crops (in present case mainly maize), while mineral nitrogen should be applied mainly to winter crops with a steep N production function. Another approach to balance out the P surpluses by compost applications is the cropping of legumes.

Additionally, to the main nutrients, compost does also contain substances that in high amounts can be potentially toxic. The PTE investigated in this study are Cd, Cu, Ni, and Zn, which are often considered to be of highest importance in agroecosystems, yet they are also micronutrients, except Cd, and as such essential for plant growth (He et al., 2005). The plant uptake, however, is considerably lower than the supply by compost in present experiments, which results in a net surplus of PTEs. Nonetheless, the question remains, if any surplus of PTEs is equivalent to environmental pollution (see Section 4.4.) or increase in plant uptake and thus risk of entering the human food chain. The data from our experiment suggest that even though high surpluses of PTEs are applied, no clear increase in biomass concentrations can be observed especially for Cu and Cd. For Zn, the picture was less clear since a few increases of Zn in plant biomass were detected. Investigations by Al-Najar et al. (2016) suggest that Zn is mostly present in its bioavailable organic form while Cu is mostly in its unavailable inorganic form in soils amended with compost. This could explain the discrepancies between the effects of compost application on Cu and Zn uptake. This is further supported by López-Rayo et al. (2016), who investigated the effect of long-term compost application on the PTE uptake of plants and found only an increase in Zn uptake but not in Cu. Zn is an essential nutrient for humans and thus a small increase in plant content far below toxic levels could be seen as a beneficial side-effect of compost application.

4.4 | Effects on soil fertility and PTE accumulation

The fourth hypothesis could be partly supported by the data. Long-term compost application increased SOC, total soil N, CAL-P, and CAL-K, and pH in comparison to the controls, but only Cu out of all PTEs was increased in 400C. Thus, the assumptions made on soil Cu, Ni, Zn concentration in hypothesis four need to be rejected.

The effects of compost fertilization on soil pH (Table 4) are related to the contents of carbonic acids and bases of organic acids in composts. Further, approx. 8% of the C compounds of composts may be carbonate C (He et al., 1995). These properties of composts were also reported by Hargreaves et al. (2008) and Costello and Sullivan (2014). Further, composts are often amended with liming material during the composting process. The strong increase in pH is also supported by other long-term experiments (Emmerling et al., 2010; Glæsner et al., 2019; Lehtinen et al., 2017). The strong increase of soil pH in the treatment with large compost inputs indicates that these large inputs are in the long-term not only not sustainable in terms of their nutrient surpluses (N, P, K), but also in terms of their effect on soil pH. High increases in soil pH can potentially affect the availability of many nutrients sensitive to

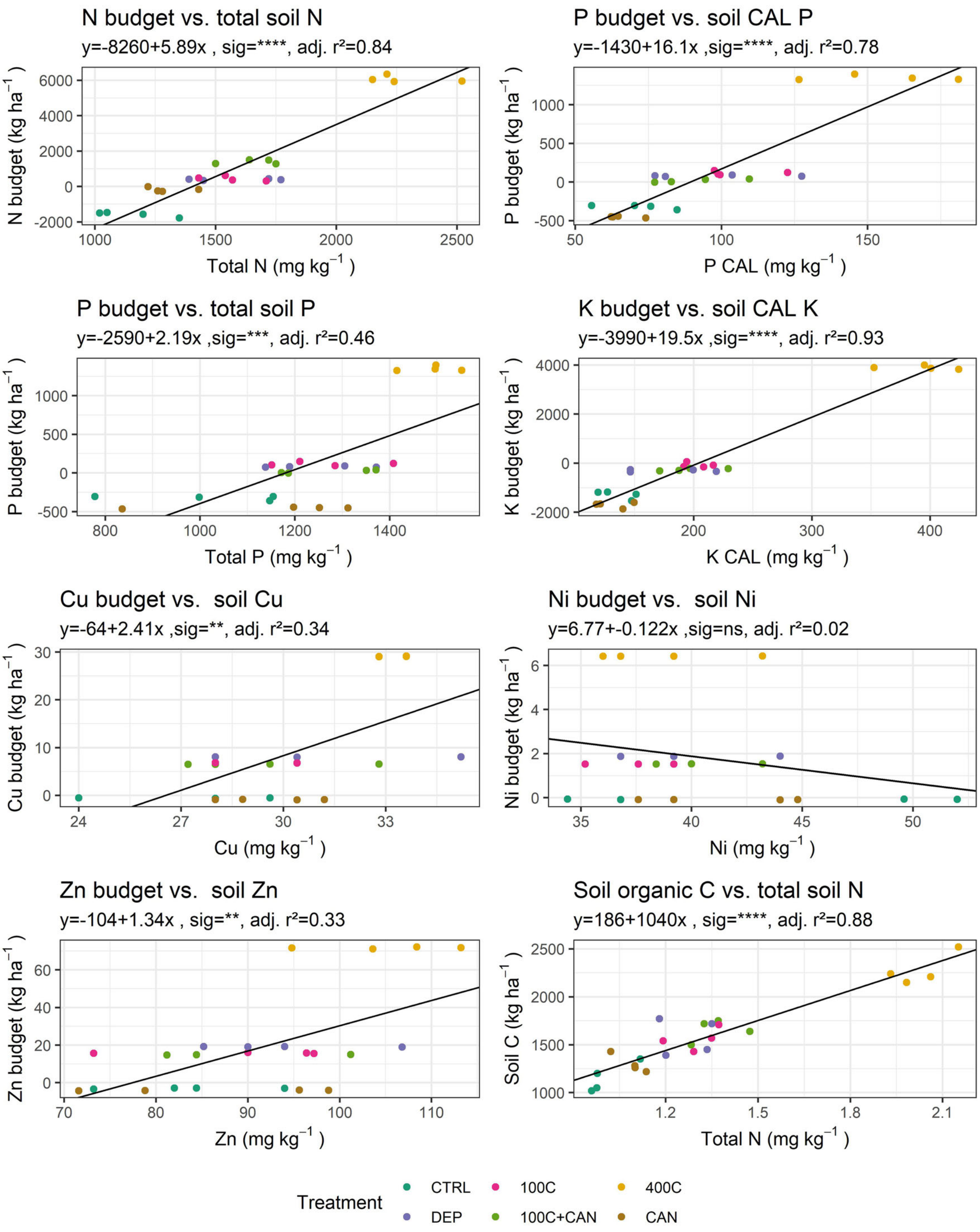

FIGURE 5 Relationship between cumulative field nutrient budget (for the period between 2003 and 2019) for nitrogen, phosphorus and potassium and the respective soil concentrations on nitrogen, total phosphorus, extractable phosphorus (CAL P) and potassium (CAL K), as well as for cumulative field budgets of the potentially toxic elements Cu, Ni and Zn. ($\alpha = 0.05$, $n = 24$, df $= 22$)

soil pH (e.g., P, micronutrients) (Blume et al., 2018). Yet, an increase in pH reduces the availability of PTEs in the soil in terms of transport or leaching and bioavailability (He et al., 2005).

The effect of compost on soil total N was already discussed in Section 4.2. Still, data indicated a strong relation between soil organic carbon and soil total N (Figure 5). The SOC increase detected in the compost treatments is a well-known effect of compost application (Emmerling et al., 2010; Glæsner et al., 2019; Lehtinen et al., 2017). As per the findings by López-Rayo et al. (2016), higher amounts of compost (e.g., 400C) resulted in a non-linear increase in SOC, which is explained by higher C inputs. Higher SOC in DEP compared to the mineral fertilized and the control treatments even 18 years after the last application indicates that there was still a residual effect of compost application and underlines the fact that compost application contributes to a more stable fraction of organic matter (Peltre et al., 2017). Comparing our results to data from 2008 (Al Sharif, 2008) shows that in 2008 after 10 years of compost application the plateau of SOC accumulation was not yet reached. There was still an increase from 2008 to 2019. The lack of effects of mineral fertilizer N on the SOC in soil contradicts to some extent earlier findings (e.g., Kätterer et al., 2012; Poffenbarger et al., 2017; Powlson et al., 2011). This is probably related to the net mineralization of organic matter, which is indicated by a higher N offtake than N supply (Table 3).

Compost and fertilizer application affects CAL-P concentration as well as P_t concentration. Although differences were not significant in all cases, there is a tendency for higher P_t concentrations in the compost-fertilized treatments due to the P additions achieved by the compost. Glæsner et al. (2019) and López-Rayo et al. (2016) detected higher P_t for compost treatments as well. The higher concentrations of CAL-P compared to mineral fertilization for the 400C, 100C and DEP treatments is in line with literature findings (Brown & Cotton, 2011; Emmerling et al., 2010) and indicate that at least parts of the P provided by the compost is extractable and therefore to some extend plant available. An increase in soil P has been associated with a net surplus of P as we detected in the P budgets for 400C and 100C (Korsaeth, 2012; Reimer et al., 2020). Present data indicate that an annual net removal or addition of 16.1 kg P ha^{-1} resulted in change of 1 unit in mg P kg^{-1} soil of CAL-P in the long-term (Figure 5b). These values are higher than values between 8–12 kg P ha^{-1} of annual net removal or surplus to change the concentrations of soil CAL-P by about 1 mg per kg soil reported in literature (Knauer, 1968; Kerschberger & Schröter, 1996; Römer, 2009). The reasons for these large differences are not known, as the relationship depends probably to a large extend on the overall soil P level, characteristics of P source and a range of many interactions between P and many elements influencing their extractability. One of which is the pH level and the addition of large amounts of Mg and Ca, which are increased or added with the compost application and could influence the extractability of P. The relatively high amounts of net P flows needed to change the levels of CAL-P in present experiments might be related to the high fertility level and the high availability of sorption sites for phosphate in the soil. The effects of net nutrient removal or surplus on soil total P is less pronounced than the effect on CAL- P (2.19 kg P ha^{-1} per mg P_t kg^{-1} soil; Figure 5c), indicating that soil total P is less influenced by the fertilization management than the CAL-P.

The lower share of soil CAL-P to soil total P in the mineral N treatments (Table 4) are probably related to significantly higher P offtakes as it can be concluded from Table 3 when comparing these treatments to the control, meaning that mainly CAL-P was removed, without an equivalent P transfer from the less reactive soil fractions to the reactive P. Additionally the literature suggests, that compost application can lead to a mobilization of the adsorbed P pool in the soil due to the contained organic acids and silicates (Koski-Vähälä et al., 2001; Möller et al., 2018). Furthermore, in present experiments, liming within the CAN treatment might have induced some additional fixation of CAL-P, as previously reported by Curtin and Syers (2001). Other than for liming, the compost application in present experiments did not affect the share of CAL-P to total P. This contradicts in part former reports. Lehtinen et al. (2017) did not detect an increase of CAL-P for organic waste compost with application rates like in our experiment, also leading to large P surpluses. Similarly, Glæsner et al. (2019) measured a high total P content for compost-amended soils similar to sewage sludge and twofold higher than manure compost, but P extractability was lower in the compost treatments. P in compost-amended soils is mainly present in organic forms or associated to calcium (Glæsner et al., 2019; Jorgensen et al., 2010). Glæsner et al. (2019) proposed that the high C input with compost in combination with calcium input and the pH effect of compost lead to adsorption of P in the soil. Yet, these effects could be soil type dependent.

The higher CAL-K concentration in the soil for the three compost treatments corresponds with other experiments, as well as with the calculated K budgets. Baldantoni et al. (2016) detected higher CAL-K concentrations for 2 years within 7 years of compost application. Emmerling et al. (2010) detected increased CAL-K compared to the control after 10 years of compost for one of three crop rotations. Generally, household waste compost, like the compost used in present experiment, contains high amounts of K. Present data indicate that an annual net removal or addition of approx. 19.5 kg K ha^{-1} will result in change of 1 unit of CAL-K in mg K kg^{-1} soil in the long term (Figure 5b). This relation between K budget and soil CAL-K concentration, however, cannot be observed in farm inventories (Løes & Øgaard, 1997; Reimer et al., 2020).

Extractable Mg was only increased with the high compost application rate. Baldantoni et al. (2016) and Bartl et al. (2002) did not detect an increase in total soil Mg and extractable Mg in compost-amended treatments in their long-term experiments. Contrastingly, Weber et al. (2007) detected higher extractable Mg after compost application. The higher concentration of extractable Mg in the high compost application treatments is related to the Mg applied with compost, which resulted in an annual Mg input of 25 and 100 kg ha^{-1} for the 100C treatments and the 400C treatment, respectively. Simultaneously, extractable Mg concentrations also increased in the treatments without any Mg application when comparing the datasets of 2019 and 2008, an effect that cannot be explained by the dataset. Most of the findings in literature and the fact that there is only tendency towards higher Mg concentration in the normal rate compost treatment leads to the conclusion that the effects of composts on extractable Mg concentration are only a minor effect associated with compost application. Further, as mentioned above, the comparison between the 2 years must be done with caution. An analytical bias cannot fully be excluded.

Contrary to common concern, the study did not show any risk of soil contamination with PTEs when compost is applied in amounts that do not exceed the requirements of macro nutrients like P. The levels of soil Ni, Cu, and Zn concentration in all treatments did not exceed the soil precautionary values for loam soils according to German legislation (BBodSchV) (Table 4). The slight increase of soil Cu concentration in the accelerated compost treatment with an annual Cu load of above 1 kg Cu ha^{-1} simulating around 100 years of annually compost application, indicate that even after such long period of time no relevant negative influence on soil fertility is expected. This is in line with López-Rayo et al. (2016) who measured a higher soil Cu concentration only for the treatments with an accelerated compost application approach. Cambier et al (2019) also only observed an increase in EDTA-extractable copper compared to EDTA-extractable cadmium, zinc, and nickel since these are influenced by the compost induced change of soil pH and soil organic matter. On the other hand, Baldantoni et al. (2016) and Emmerling et al. (2010) also detected higher total Cu concentrations after lower compost application rates. This indicates that the effect of accumulation if Cu does not solely depend on the net surplus of Cu, but also on different environmental factors such as soil type and climate. Furthermore, it should be considered, that a weakness of any accelerated approach is that it does not account for the leaching losses expected for such a long period, in combination with the short-term over-liming effect, which reduces PTE leaching, as indicated by Kim et al. (2015). Therefore, under humid climatic conditions the PTE accumulation risk will be overestimated by the performed accelerated approach.

The lack of accumulation of Zn and Ni is partly supported by the literature (Baldantoni et al., 2016; Bartl et al., 2002). Yet, an increase in soil Zn with compost application is frequently reported (Bartl et al., 2002; Emmerling et al., 2010; López-Rayo et al., 2016). However, the Zn concentrations of the compost used in this experiment was lower than that of compost used in the experiments of López-Rayo et al. (2016) and Bartl et al. (2002), which could explain the differences in results. This stresses that to avoid PTE accumulation in the soil, the compost must be of high quality. In organic farming, for instance, compost quality is ensured by strict PTE thresholds set by European Union regulations (European Commission, 2021). The used compost in the experiment is in line with these regulations, which suggests that they are effective in preventing soil PTE contamination. In addition, it must be noted, that soil Zn concentrations at the experimental site was higher than the natural background concentration of noncontaminated soils (10–80 mg kg^{-1}) (Blume et al., 2010) which means that the site was already high in Zn.

5 | CONCLUSION

The results of this long-term experiment underline the fertilization effect of compost through the soil nutrient pool and suggest that compost can substitute mineral N, P, and K fertilizer to some extent. A major problem, however, is the synchronization of plant N demand and N supply through mineralization. In winter wheat about 35–30% of the total N mineralization occurs outside the cropping period. This issue varies depending on crop species and their growing season. Compost application with the goal of fulfilling the crop's N demand results in imbalances of nutrients, especially a surplus of P. Nutrient surpluses and deficits also effect the extractable soil nutrient content. Therefore, compost could be used to increase soils low in P as often found in organic agriculture. In soils with moderate or high CAL-P levels, compost application should be limited by the P input and combined with additional N sources. A combination of compost with mineral fertilizer can secure yields in all investigated crops and has the additional advantage of balancing out the P supply. Even though compost application is related to an input of PTEs (Cu, Zn, Ni), the use of high-quality compost with low PTE contamination prevents soil PTE accumulation and increased plant uptake. However, to optimize the use of compost as fertilizer and soil amendment further research and a continued long-term experiment is necessary to improve utilization from mineralized compost N to determine how to efficiently integrate compost into the crop rotation and substitute mineral N, for example, using early sowing of winter crops or cover cropping.

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DATA AVAILABILTY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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SUPPORTING INFORMATION

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