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ORIGINAL ARTICLE

Urban waste fertilizer: effects on yield, nutrient dynamics, and potentially toxic element accumulation

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Abstract Recycling nutrients contained in urban wastes to agriculture is essential in a circular economy. This study simultaneously compares different recycled fertilizers (household waste compost, sewage sludge, human urine) with mineral fertilization and animal manures. Tested were their long-term effects on yield, nutrient budgets, potentially toxic element (PTE) accumulation, and nitrogen (N)/carbon (C) cycle (among others N efficiency, N losses, soil C). Therefore, data from a long-term field trial and predictions from the soil–plant-atmosphere model *Daisy* were evaluated. Based on trial data, human urine performed similar to the mineral fertilization for yield, N efficiency (mineral fertilizer equivalent (MFE) = 81%), and nutrient budget, while sewage sludge and compost were comparable to animal manures in terms of having lower yields, N efficiencies (MFE 70% and 19% respectively) and higher nutrient imbalances, especially P and S surpluses. Compost and sewage sludge applications resulted in net PTE inputs. Yet, plant uptake and soil accumulation seemed neglectable. Model outputs predicted N losses of 34–55% of supplied N. Losses were highest for compost, followed by deep litter, manure, sewage sludge, human urine, mineral fertilization, and slurry. Nitrate leaching was the main loss pathway (14–41% of N input). Within the compost and straw-rich manure fertilization, about 25% of applied N was stored in the soil which was accompanied by an increase in soil C. The study suggests substitution of established fertilizers with recycled ones is feasible. Thereby each fertilizer has advantages and disadvantages and thus should be utilized according to its strength or in mixtures.

Keywords Nitrogen efficiency · Nitrogen losses · Soil–plant-atmosphere modeling · Sewage sludge · Household waste compost · Human urine

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Introduction

Due to the expected upcoming resource scarcity, recycling and transitioning to a circular economy has become a primary objective not only in society, but also in the agricultural sector. One step towards achieving this goal could be the use of urban waste products as fertilizers on agricultural lands. It would help to close the nutrient cycle between urban and rural areas and reduce the need for non-renewable fertilizers such as rock phosphate and mineral N fertilizers, whose production is a major source of greenhouse gases in the agricultural sector (Safa et al. 2011).

The principle of recycling is already deeply rooted within the organic farming philosophy (Vogt 2000). Additionally, there is a strong need for suitable nutrient inputs in organic farming (Reimer et al. 2024). Recent developments, such as the farm to fork strategy, which aims at 25% organic managed land in the EU by 2030 (European Commission 2020), will further increase the need of suitable fertilizer for organic farming in the future. This has brought new attention to recycling urban wastes as fertilizers in organic farming (Løes et al. 2017; Möller et al. 2018; Milestad et al. 2020). Assessing the suitability of recycled urban waste fertilizers as substitutes for mineral fertilizers in conventional farming and for animal manures from conventional sources in organic farming could promote the growth of the organic sector. This approach could also replace contentious inputs like rock phosphate and conventionally sourced animal manures.

Significant amounts of nutrients can be recycled from urban waste streams. Zoboli et al. (2016), for example, estimated that recycling urban waste materials could substitute 70% of Austria's fossil P imports. The biggest sources of recycled nutrients are sewage sludge, slaughterhouse wastes, food wastes and food industry byproducts or wastes, and organic household wastes (Möller et al. 2018). Furthermore, solid urban waste materials like compost and sewage sludge lead to higher soil C contents (Peltre et al. 2015, 2017). Increased organic matter inputs are associated with many agronomic valuable soil traits, such as higher water infiltration, higher aggregate stability, better workability of the soil and increased pH levels in the soil (Weber et al. 2007; Singh and Agrawal 2008; Obriot et al. 2016). They can also increase soil microbial mass and thereby enhance nutrient absorption and use efficiency and control soil borne pathogens (Litterick et al. 2004; Obriot et al. 2016; Peng et al. 2017; Farzadfar et al. 2021; Vermeiren et al. 2021).

Despite the large potential of recycled fertilizers from urban waste, the current use is limited. The reasons are manifold. In many areas there is a lack of infrastructure to collect waste materials for recycling (Ott and Rechberger 2012). Additionally, concerns about contamination with potentially toxic elements (PTEs) like copper (Cu), cadmium (Cd), and zinc (Zn) (Løes et al. 2017) prevent several urban wastes from being permitted in organic farming, making farmers reluctant to use them (Reimer et al. 2024). Potentially toxic elements, especially Cd, pose a risk to human health (Åkesson et al. 2014) and are a key factor in evaluating recycled fertilizers, leading conventional farmers to opt for mineral fertilizers instead. Yet, the reputation of recycled fertilizers is worse than they actually are. There have been many technical improvements in sewage sludge plants and legislative directories that lowered the contamination load (Olofsson et al. 2012). A recent assessment of risks to human health and to the soil ecosystem, showed that land application of contemporary Danish sewage sludge entails similar risks overall as pig slurry (Magid et al. 2020). Finally, the nutrient availability and homogeneity can be a problem for recycled fertilizers (Milestad et al. 2020; Sailer et al. 2021) and due to their organic character, nutrient content and release can vary (Gutser et al. 2005; Sailer et al. 2021).

Recycled fertilizers are most often multi-nutrient fertilizers, and the nutrient concentration depends on the used waste material and treatment. Their nutrient stoichiometry does not always match the crop need, which can lead to an imbalance among nutrients (Möller 2018). Many recycled fertilizers such as composts have a lower N to phosphorus (P) ratio than plant offtake, which can result in an oversupply of P with its negative environmental impacts when applied based on the plant's N need (Zikeli et al. 2017; Reimer et al. 2023). Further, the nutrient release can be harder to predict since many nutrients are bound in organic forms and need to be mineralized before plant uptake (Gutser et al. 2005; Geisseler et al. 2021). Additionally, fertilizers with a wide C:N ratio might even cause net N immobilisation in the short-term (Reimer et al. 2025). This makes the synchronization of nutrient supply and N demand more challenging and generally results in a lower fertilization effect than mineral fertilization, especially for N (Pang and Letey 2000; Schröder 2014; Reimer et al. 2023). In the literature several results for MFE of recycled fertilizers can be found and they vary between 25% for compost up to 80% for digestates (Amlinger et al. 2003; Schröder 2014; Gómez-Muñoz et al. 2017; Möller 2018). For P and potassium (K), on the contrary, it is assumed that 100% of the nutrients will be taken up by the plant in the long term (Frossard et al. 2016; Schnug and Haneklaus 2016).

An inefficient use of recycled fertilizer N can lead to higher N losses to the environment in the form of nitrate leaching, nitrous oxide emissions and ammonia losses (Yoshida et al. 2016; Bruun et al. 2016; Gómez-Muñoz et al. 2017). The magnitude of these N losses is influenced by many factors, such as soil type, weather and climate, application timing, soil incorporation, crop demand, and the composition of the organic matter (Gerke et al. 1999; Cabrera et al. 2005; Cameron et al. 2013; Bruun et al. 2016). Measuring all the different pathways through which N can be lost from the system in a field trial is both challenging and costly. Soil–plant-atmosphere models can be a remedy to estimate environmental emissions in these situations (Heinen et al. 2020). Dynamic agro-ecosystem models take soil type, weather, climate, and crop rotation into account and predict the fate of N after fertilization (Bruun et al. 2006, 2016; Yoshida et al. 2016). *Daisy* is an example of such a model (Hansen et al. 2012). The *Daisy* model has performed well in previous model comparisons (Palosuo et al. 2011; Rötter et al. 2012; Kollas et al. 2015; Yin et al. 2017) and is a well-established model for predicting the fate of N and C in soil in diverse crop rotations after application of recycled fertilizers in the short-term (Yoshida et al. 2016; Bruun et al. 2016). Yet, as all soil–plant-atmosphere models it has limitations, such as not including changes in soil bulk density, pest and diseases or non-symbiotic N fixation (Yan et al. 2020). These limitations have to be considered when interpreting the model predictions. Additionally, to be able to check long term model predictions and ensure their validity, long-term field trials are indispensable (Smith et al. 1997). Many processes associated with the addition of organic fertilizers, like build-up of organic matter, PTEs, soil nutrients and yield effects, take a long time to develop and to reach their whole potential (Pang and Letey 2000; Macholdt et al. 2021). The CRUCIAL experiment is a field trial designed to investigate the long-term effect of recycled fertilizers on soil fertility, crop productivity and the risk of accumulation of contaminants (Magid 2006). Thus, it is ideal to be used in this study to investigate the long-term effect of recycled fertilizers in comparison to animal manures and mineral N fertilization.

To conclude, recycled fertilizers from urban wastes hold a high potential for use in agriculture. If recycled fertilizers are to substitute part of the need for mineral fertilizers in conventional farming, or meet nutrient demands and replace unwanted inputs in organic farming, they must be evaluated for their agronomic potential and environmental impact. Thus, this study aimed to compare different recycled fertilizers from urban waste streams (sewage sludge, household waste compost and human urine) with mineral N fertilization and cattle manures, to highlight their strengths and shortcomings. These recycled fertilizers were evaluated on multiple aspects to give a holistic assessment using data from a long-term field trial aided by the *Daisy* model simulations to investigate the N and C cycle in more detail. The study's objectives were, a) to determine if the *Daisy* model is an useful tool to simulate agronomic and environmental effect of long-term use of recycled fertilizers, b) to investigate the N crop supply from recycled fertilizers, c) to determine nutrient imbalances in form of surpluses of P and K as a result of the use of recycled fertilizers, d) assess the risk of soil PTE accumulation, e) to evaluate the N losses using model simulations, focusing on ammonia volatilization, N_2O emissions and N leaching as a result of long-term application of recycled fertilizers, and f) to determine changes in soil organic matter due to the long-term use of recycled fertilizers.

Methods and material

Field experiment

Experimental design

For this study, data were collected from the long-term field trial CRUCIAL (Magid 2006; López-Rayo et al. 2016; Gómez-Muñoz et al. 2017). The trial is located at an experimental farm operated by the University of Copenhagen in Denmark (55°40.0′N, 12°18.0′E). The soil is classified as a sandy loam with a clay content between 12 and 19% and a pH_{H2O} between 6.6 and 7.5 (Magid 2006). The experiment was established in autumn of 2002 and has been run in the same way since then. The trial is set up in a randomized block design with three blocks with 11 plots (each 33 m by 27 m). The eleven different treatments are composed of eight different organic fertilizer applications and three control fertilization schemes (Table 1). The organic fertilizer treatments were compost from household waste (CH), accelerated CH (CHA), sewage sludge (S), accelerated S (SA), human urine (HU), cattle slurry (CS), deep litter (DL, straw rich cattle manure), and an accelerated cattle manure (CMA) fertilization. The accelerated fertilization treatments (CMA, CHA and SA) received approximately three times the amount of the normal fertilization rate. The aim of the accelerated fertilizations was to simulate a longer-term effect of application of those fertilizers on soil characteristics. The control treatments were mineral fertilization and two unfertilized controls of which one received green manure as undersown grass clover (*Trifolium* sp. and *Lolium perenne*) most autumns (Supplementary Table S1). All fertilizers were applied in the amount of the recommended plant-available N based on Danish farming standards. The applied N amount depended on the crop, around 100 kg available N ha^{-1} (Table 1). The ratio of available N to total fertilizer N was calculated based on standard values for mineral N fertilizer replacement values as stated by the Danish Fertilizer Regulations (Anon 2013). The main properties of the fertilizers are listed in Table 1. Generally, the fertilizers were applied before sowing and incorporated into the soil by ploughing at a depth of 20–25 cm. Yet, NPK, HU, and CS fertilizers were applied in spring at the start of the growing season: they were used on growing crops for winter-sown crops and incorporated before sowing for spring-sown crops.

The crop rotation was dominated by spring cereals (barley, oats, and wheat), but also some winter crops were cultivated throughout the years (Supplementary Table S1). For cereal crops the straw was harvested together with the grain. Additionally, all plots were split in 2008, 2009 and 2010 into two subplots where half was organically managed without pesticides and the other half was managed conventionally. For a detailed description of the field experiment consult Magid (2006) and Gómez-Muñoz et al. (2017).

Measurements

In the field experiment, multiple measurements were conducted (Supplementary Table S2). Each year, the agronomic yield of grain and straw, or total biomass for silage crops, was recorded. Additionally, from 2002 to 2015, the N and C content of the harvested biomass was determined. Samples were oven-dried at 80 °C for 48 h, finely ground into powder, and analyzed for total C using a CNS Elemental Analyzer (Vario Micro Cube). Total N was quantified through Kjeldahl digestion followed by flow injection analysis (FIA).

During the same period, the dry matter N and C contents of the fertilizers were also assessed. The NH_4^+ concentration was measured using the FIAstar 5000 (FOSS Analytical, Denmark), while contaminant levels of PTEs were evaluated from 2002 to 2008. PTE contents were determined as described by López-Rayo et al. (2016). Soil samples were finely ground using an agate mill (Fritsch Pulverisette), and 250 mg of the ground soil was combined with 9 ml of 70% HNO_3 and 1 ml of 30% H_2O_2 and left to stand overnight. The following day, 2 ml of 30% HCl and 3 ml of 40% HF were added, and the mixture was digested using a microwave oven (Multiwave 3000, Anton Paar, GmbH, Graz, Austria). After digestion, samples were transferred to 50 ml volumetric flasks, diluted with 20 ml of 6% H_3BO_3, and Milli-Q water to a final volume of 50 ml. The samples were then analyzed using inductively coupled plasma-optical emission spectroscopy (ICP-OES, Optima 5300 DV, PerkinElmer).

Soil measurements were consistently taken throughout the study (Supplementary Table S2). Soil texture was analyzed in 2008 and 2011 (in only six plots). Soil C and N content was measured in 2001, 2002, 2006, 2007, 2009, 2011, 2013, 2019 (in a limited number of plots), and 2020. Soil bulk density was determined in 2011 and 2014. Mineral soil N was measured in 2002, 2003, and twice in 2004. Soil PTE levels were assessed in 2001, 2006, 2009, 2011, and 2013.

Table 1 Fertilization treatments applied and their main properties in the CRUCIAL trial in the period of 2002–2020. Given are average dry matter (DM%), total and ammonium nitrogen (N_{total}, NH_4^+-N), and carbon (C) content of the fertilizers

Fertilization		DM	N_{total} applied	N_{total}	NH_4^+-N	C	C/N	C	P	K	Mg	S	Cd	Cu	Zn
		%	kg N ha^{-1} year^{-1}	% DM	% DM	% DM		kg C ha^{-1} year^{-1}	kg P ha^{-1} year^{-1}	kg K ha^{-1} year^{-1}	kg Mg ha^{-1} year^{-1}	kg S ha^{-1} year^{-1}	g Cd ha^{-1} year^{-1}	g Cu ha^{-1} year^{-1}	g Zn ha^{-1} year^{-1}
CH	Composted household waste	66.2	388	1.95	0.04	23.7	12.1	4716	115	186	61.7	47.5	10.1	2668	5556
CHA	CH accelerated	66.2	1164	1.95	0.04	23.7	12.1	14,147	345	557	185	143	30.4	8004	16,668
S	Sludge	25.7	200	4.75	0.89	31.7	6.67	1335	161	10.7	26.4	54.3	14.3	1252	3167
SA	S accelerated	25	577	4.56	0.84	31	6.79	3923	482	32.3	79.3	163	43.0	3763	9518
HU	Human urine	0.45	150	48.5	39.4				0.050	0.185	0.0002	0.065	0.0003	0.059	0.139
DL	Deep litter cattle manure	34.1	308	1.84	0.04	34.3	18.7	5742	90.1	538	64.0	65.8	6.36	357	2838
CMA	Cattle manure accelerated	22	401	2.13	0.52	45.8	21.4	8622	122	330	84.1	66.2	4.33	486	2452
CS	Cattle slurry	7.25	114	2.77	1.23	43.6	15.8	1794	36.7	178	28.8	21.0	0.864	160	986
NPK	NPK fertilizer	100	108	20.7	9.1	–	–	–	–	–	–	–	–	–	–
U	Unfertilized														
GM	Green manure														

Further the applications of the main nutrients (N, P, K, Mg, S), C, and potentially toxic elements (Cd, Cu, Zn) are shown in kg ha^{-1} year^{-1}. Values are the mean over all application years

Daisy model description

The *Daisy* model is a one-dimensional mechanistic model that simulates water, N, C, and pesticides in the bioactive zone of the soil (Hansen et al. 1991, 2012). The model consists of a hydrology model (simulating soil temperature, evapotranspiration, and soil water transport with the Richard's equation), crop models (simulating crop N uptake, dry matter growth and phenology), a mineral N model (simulating nitrification, denitrification and transport of ammonium and nitrate), and a soil organic matter model (simulating mineralization of C and N). For a detailed description of the model see Hansen et al. (2012). The model is available online free of charge at https://daisy.ku.dk/.

Model setup

The *Daisy* model version 5.67 was used for the simulations. The weather data were taken from Taastrup weather station which is located 1.83 km away from the experimental site (Svane and Petersen 2021) and the daily values for precipitation, global radiation, and temperature from 1991 to 2020 were used.

The hydraulic soil parameters were estimated by using the RetC-Model with a van-Genuchten–Mualem estimation (Van Genuchten et al. 1992). For the estimation, the soil texture and bulk density as measured in the field trial was used. Soil columns were defined based on the soil profile in the field until a depth of 2 m (horizons: 0–25 cm Ap, 25–45 cm E, 45–85 cm Bt, and 85–200 cm BC) and maximal rooting depth was set to 1 m. The model outputs were reviewed for their hydraulic parameter descriptions, and the hydraulic conductivity of the aquitard was calibrated to ensure that 70% of the water percolates through the soil to the groundwater, as required for soil type JB5, according to the *Daisy* model manual (Styczen et al. 2004).

The partitioning of organic fertilizers C and N between added organic matter (AOM) pools, and the decomposition constants of AOM pools were calibrated for compost and sewage sludge based on previous incubation studies, while for CS, CMA and DL were initialized according to default parametrizations provided by the model. The compost calibration was based on results by Bruun et al. (2006) and the sewage sludge calibration on Bruun et al. (2016, dewatered and anaerobically digested sewage sludge). The HU fertilization was treated as a mineral fertilizer, due to the high mineral N and low dry matter and C content of the fertilizer. The fertilizer calibrations were checked by comparing simulated and measured values of soil C and total N. The built-in crop modules were used and calibrated to fit the measured yield data by adjusting the parameters for maximum photosynthesis efficiency and N efficiency as recommended in the standard set up recommendations for the *Daisy* model (Styczen et al. 2004; Styczen et al. 2025). Specifically, the *FM* parameter, which describes the maximum CO_2 assimilation rate, and the parameter for the maximum NH_4^+ and NO_3^- uptake per unit of root length (*MxNH4Up / MxNO3Up*). The calibrated crop and fertilizer modules were then used to simulate the different treatments according to the management records of the CRUCIAL trial.

The fit was evaluated by plotting observed with predicted values against each other and comparing them to the 1:1 line as suggested by Piñeiro et al. (2008) and by comparing the root mean squared error (RMSE), and mean absolute percentage error (MAPE), where the evaluation index of decision was the MAPE. The index of agreement (IA) as suggested by Willmott (1981) was used as a more general indication of model fit. The IA can range between 0 and 1, where higher values indicate a better model simulation.

Nutrient budget, nitrogen and carbon balance and nitrogen efficiency

The nutrient budgets were based on the measured field trial data and calculated as the difference between nutrient input and crop offtake. For N, P, K and the PTEs Cd, Cu, and Zn a simple budget was calculated, where only total nutrient content of the fertilizers and in the case of N biological N fixation (BNF, values taken from model output) from green manures are considered as inputs and crop offtakes as outputs. Since the straw was removed in this trial both straw and kernels are considered crop offtake. For the crop nutrient contents measured data or reference values from the literature were used (Grytsyuk et al. 2006; KTBL 2015; Weissengruber et al. 2018). For the nutrient and PTE contents of the fertilizers, measurements done within the CRUCIAL trial were used, and if there were no measurements for a specific year available, the average over the whole trial period (2002–2020) was used instead. In addition to the simple budgets based on the field trial data, a more comprehensive balance for N and C was calculated by the *Daisy* model and investigated. Given the focus on N loss pathways, the model output data was used to investigate the various ways N can be lost from the system, including leaching, volatilization, denitrification, surface runoff, and N_2O emissions. However, the *Daisy* model only estimates N_2O emissions from nitrification, while for denitrification it estimates the total N losses and does not distinguish between N_2 and N_2O. Therefore, the SimDen model by Vinther and Hansen (2004) was used to estimate the ratio of N_2O to total denitrification N losses (N_2O / (N_2+N_2O)). SimDen is a simple model that estimates the denitrification and N_2O emissions based on soil type, precipitation level and amount and kind of applied N. These parameters were adjusted for each fertilization and the annual average total N losses due to denitrification were multiplied by the ratio of N_2O to get the total N_2O emissions. The resulting ratios for each fertilization can be seen in Supplementary Table S3 and vary between 0.18 and 0.20.

To compare the different fertilizations for their agronomic value, the mineral fertilizer equivalent (MFE) was calculated, where data availability allowed it. The MFE shows the relation of the apparent N use efficiency (NUE) of an organic fertilizer compared to the NUE of mineral N fertilization and is calculated as follows:

$$MFE\,(\%) = \frac{NUE_{fertilizer}}{NUE_{NPK}} * 100\%$$

$$NUE(\%) = \frac{N_{uptake}(fertilized) - N_{uptake}(unfertilized)}{N_{applied}(fertilized)} * 100\%$$

Since measures of N uptake were not available for the whole trial period (no measurements after 2014), relative agronomic efficiency was used as an alternative measure. This metric is analogous to MFE but based on dry matter yield of the harvested product (grain yield or whole crop yield for silage crops) rather than N offtake and is calculated similarly as follows:

$$relative\ agronomic\ efficiency\,(\%) = \frac{agronomic\ efficiency_{fertilizer}}{agronomic\ efficiency_{NPK}} * 100\%$$

$$Agronomic\ efficiency(\%) = \frac{yield_{dry\,matter}(fertilized) - yield_{dry\,matter}(unfertilized)}{N_{applied}(fertilized)} * 100\%$$

Results for relative agronomic efficiency and MFE from the years 2004, 2009 and 2010 were discarded, since the unfertilized control resulted in higher yields than the mineral fertilized one. Furthermore, MFE and relative agronomic efficiency were not calculated for the GM treatment, due to the lack of fertilizer input and high uncertainties of biological N fixation.

Statistical analysis

All statistical analyses were done in R (R Core Team 2018) and the R code is available. To detect significant differences, linear models were used and done with the *lmerTest* and *agricolae* package. The significance level was set to 0.05. Inclusion of factors was handled with a bottom-up approach. Data visualization was performed with the *ggplot2* package..

Due to additional complexity of the statistical analysis and especially due to missing data points for some years, it was decided not to use a repeated measure approach. In detail, the relative dry matter yield and N grain concentration were analyzed using a linear model. The factors included in the model were the fertilization treatment, the year of application, the cropped culture, the block, and the interactions between fertilization treatment and year of application, as well as fertilization treatment and cropped culture. Relative agronomic efficiency and MFE were analyzed using a linear model that included the factors of fertilization, year of application, applied N amount, cropped culture, block, and the interactions of fertilization with year of application, applied N amount, and cropped culture. Since the nutrient and PTE budgets, as well as changes in soil C, were calculated as sums or averages over the entire trial period, they were analyzed using linear models that included only fertilization treatment and block as factors. For all variables significant main effects were analyzed using Tukey's multiple comparison tests. If significant interactions were found, the factors were also analyzed separately for each fertilization treatment. To analyze the trend of agronomic efficiency over the experiment's duration, we used the LOESS (Locally Estimated Scatterplot Smoothing) method. This non-parametric technique fits local regression models to subsets of the data, creating a smooth curve that represents the trend.

Results

Calibration of the *Daisy* model

The model calibrations of the fertilizer were evaluated and the overall fit for soil organic C (Supplementary Fig. S1) had a lower MAPE of 10.4% and a higher IA of 0.94 compared to soil total N (Supplementary Fig. S2), which had a MAPE of 12.4% and an IA of 0.81 (Supplementary Fig. S3). However, the fit differed depending on fertilization treatment (Supplementary Fig. S1). For soil organic C the MAPE was below 15% for all fertilization treatments except the accelerated compost fertilization, which had an MAPE of 25%. The model underestimated the long-term C accumulation for compost. The IA ranged from 0.18 to 0.93 for all fertilizations, but was lowest for the GM, HU and NPK fertilization (0.18, 0.29, 0.37). The simulated total amount of N also fitted well to the measured values. The MAPE was below 15% for all fertilizations besides CHA and ranged between 7 and 22% (Supplementary Fig. S2). The IA ranged from 0.24 (CS) to 0.82 (CMA). There seemed to be a general trend that with increasing time, the total N in the soil was underestimated by the model especially for the CH, CHA, S, SA and NPK fertilizations.

The *Daisy* model was able to predict the dry matter grain yield, grain N yield, and grain N concentration to a certain extent (Supplementary Fig. S4). The statistical indexes for model fit varied considerably ranging from 17 to 54% MAPE and IA ranging from 0.57 to 0.93 with grain N yield showing the worst and grain N concentration the best fit. There were differences between how well the individual crop models were able to predict the measured variables. The best fit was achieved by the spring barley model, while the spring oilseed rape model predictions were less precise. However, these shortcomings were accepted for crops that were not used repeatedly in the crop rotation. A detailed table with RSME, MAPE, and IA of fit for all variables can be found in the Supplementary Table S4.

Yield effect

The yield level at the site was moderate with spring cereal averages for each year varying between 3 and 5 Mg ha^{-1} and the annual average for the mineral fertilization as a reference varied between 3 and 7 Mg ha^{-1} (Supplementary Table S5). The different fertilizations resulted in different dry matter yields throughout the experiment, even though the trial was set up to give the same amount of plant available N for each fertilizer, excluding the accelerated fertilization treatments (Fig. 1). The results of the linear model analysis revealed that the relative dry matter yield in relation to the annual average over all treatments was not only dependent on the fertilization, but also the years of application as well as the crop and their interaction with the fertilization (Supplementary Table S6). Overall, the highest yields were found for mineral fertilization (NPK), HU, and the accelerated fertilizations (CHA, SA, CMA), which had the highest N application rates (Fig. 1). The recycled bulky fertilizers (CH and S) showed lower yields, comparable to that of cattle manures. The unfertilized controls (U and GM) had the lowest yield with only 60% and 71% of the annual average respectively. Therefore, the highest yield effect per kg supplied N was achieved with NPK fertilizer (yield increase of 0.58% per kg N compared to U). Cattle slurry and HU achieved similar yield increases (0.37 and 0.39% per kg N compared to U). Intermediate values were found for SA (0.24% per kg N), the lowest relative yield increase was found for DL (0.15% per kg N) and CH (0.12% per kg N).

Due to the significant interaction between fertilization and years of application as well as cultivated crop (Supplementary Table S6), the influence of years of application and cultivated crop were tested on subsets for each fertilization separately. The relative yield only significantly increased with years of application for CHA, SA, and CMA while the yield decreased for both unfertilized controls (U and GM). The cultivated crops showed different yield responses for the CHA, SA, HU, NPK, GM and U (Supplementary Fig. S5). The yield due to CHA and SA fertilization was higher for the winter-sown crops, which was the opposite for the GM.

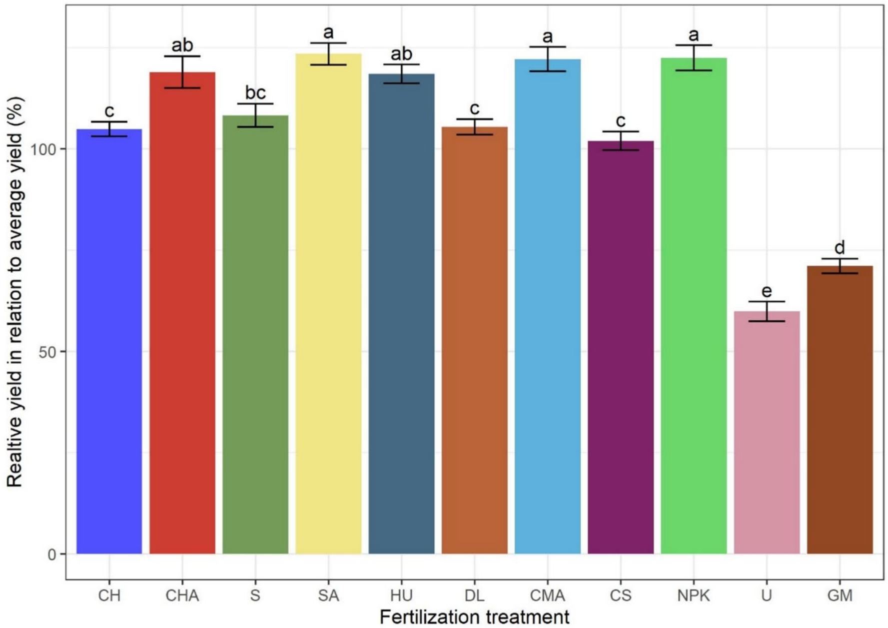

Fig. 1 Overall yield effect of the different fertilization treatments as the relative yield in relation to the annual average over all fertilizations as measured in the CRUCIAL field trial. Bars represent the annual average and error bars the standard error. For the treatment abbreviations consult Table 1

The effect of the fertilization on the measured grain N concentrations was less pronounced than the effect on yields. Still, there were some significant differences (Supplementary Table S6). The CHA and SA fertilization showed slightly higher values, while the unfertilized control (U) resulted in the lowest grain N concentration (Supplementary Fig. S6). The crop type had a large influence. The crop N concentration decreased with increasing application time, but this effect could be due to the kind of crops cultivated in the trial. Spring oilseed rape, which has a higher N concentration, was cropped in the beginning of the trial, while toward the end, mostly spring cereals were grown. The interactions between fertilization and the other two factors, years of application and cultivated crop, were not significant (Supplementary Table S6).

Mineral fertilizer equivalent and relative agronomical efficiency

The absolute values and the pattern of the fertilization effect on the MFE, which is based on the N yield, and relative agronomic efficiency, which is based on dry matter yield, were similar even though measurement periods and measurements differed (Table 2). The only differences could be seen for S and SA where MFE was higher compared to relative agronomic efficiency. None of the recycled fertilizers outperformed the mineral fertilization (all MFE < 100%). The highest MFEs were observed for CS and HU, the fertilizers with the lowest organic matter fraction and the lowest C/N ratio (Table 1). For the more C rich fertilizers (CH, S, DL, CMA), S showed significantly higher values than the others, while the accelerated fertilizer treatments (especially CHA) showed lower values for MFE and relative agronomic efficiency.

Besides the fertilization effect, the relative agronomic efficiency was also significantly influenced by the years of application, the amount of applied N, the crop, and their interactions (Supplementary Table S7). There were a few differences among crops with oats showing the highest relative agronomic efficiency (55%) followed by winter barley (49%) and finally winter wheat, spring barley and spring oilseed rape (45–41%).

Since the interactions between factors were also significant (Supplementary Table S7), the effects of years of application and applied N amount were separately analyzed for each fertilization and indicated that the amount of N applied showed significant effects for all fertilizations besides CHA, while the duration of the experiment showed only significant influences for CHA, S, HU, and CS. Yet, for the CHA, HU and CS, there were significant interactions between the applied N amount and the duration

Table 2 Means for MFE (mineral fertilizer equivalent in %) and relative agronomic efficiency (in %) as affected by fertilization based on field trial data

Fertilization	Estimate	SE	N	Min	Max	Q25	Q50	Q75	
Fertilization effect on MFE (%)									
CH	20.05	1.58	27	8.95	47.04	14.45	18.27	25.08	de
CHA	10.82	0.89	27	−2.69	16.95	9.58	11.57	14.08	e
S	64.89	5.74	27	5.80	121.79	45.68	64.96	85.87	c
SA	35.11	2.51	27	6.00	56.13	26.63	37.13	41.13	d
HU	84.23	5.77	27	31.17	171.21	67.42	82.02	94.57	b
DL	30.24	2.64	27	10.03	59.16	18.52	32.49	38.11	d
CMA	35.22	2.95	24	10.93	58.87	25.81	34.32	46.82	d
CS	88.21	9.92	27	5.47	247.62	54.97	83.09	105.48	ab
NPK	100.00	3.76	27	53.17	135.04	91.12	102.31	110.38	a
Fertilization effect on relative agronomic efficiency based on dry matter yields of crops (%)									
CH	24.63	1.59	45	5.21	48.37	15.57	25.44	31.66	d
CHA	10.24	0.83	45	−4.25	26.31	6.85	10.30	12.74	e
S	54.84	4.73	45	−19.46	146.00	35.59	54.17	69.92	c
SA	22.35	1.66	45	2.45	66.33	14.29	21.63	28.08	de
HU	70.44	4.16	45	26.14	143.90	48.51	68.17	85.65	b
DL	32.49	1.86	45	8.53	68.21	23.61	31.63	36.96	d
CMA	30.90	2.15	42	7.57	73.77	22.31	28.71	39.15	d
CS	78.42	7.12	45	−9.90	234.46	49.57	74.76	94.96	b
NPK	100.00	4.38	45	−19.11	207.56	91.73	101.00	106.51	a

The table includes estimates (Estimate), standard error (SE), sample sizes (N), minimum (Min) and maximum (Max) values, and quartiles (Q25, Q50, Q75). Letters indicate significant differences between fertilization treatments (alpha = 0.05). For the fertilization abbreviations consult Table 1

of the experiment, which hinders the interpretation of the factors on their own. Thus, the only fertilization, which had a pure duration effect is S, where the relative agronomic efficiency decreased with longer duration of the experiment. However, data provided in Supplementary Fig. S7, indicated an increase of efficiency during the first five years for most organic fertilizers, except for CH which showed a decrease.

Budgets of nutrients and potentially toxic elements

For the nutrient budgets based on the CRUCIAL field trial, the accelerated fertilizations (CHA/SA/CMA) were excluded, because applying approximately three times the realistic values led to substantial nutrient budget surpluses (e.g., approx. 1000/500/250 kg N ha^{-1} year^{-1} respectively). The variation between the fertilizations is mainly due to large differences in the amounts of inputs rather than due to differences in outputs (Supplementary Table S9), while the variation within each fertilization can be attributed to yearly variation. In general, fertilization with a more C rich fertilizer (CHA, S, DL) resulted in the highest surplus of nutrients, due to the high application rates. The sole mineral N fertilization (NPK) resulted in a balanced budget for N and deficits for all other nutrients (Fig. 2). The HU fertilization performed very similarly to the NPK fertilization. The unfertilized controls (U and GM) resulted in deficits for all nutrients. The pattern for the PTEs is similar, the organic fertilizers had surpluses while the others had negative budgets. The CH fertilization had the highest surplus for all PTEs, with the largest one for Cu.

Nitrogen flows in the cropping system

Concerning the high N surpluses as described in Sect. 3.4, it is important to investigate where the surplus N ended up as there could be possible environmental effects. The *Daisy* model output for N in the soil layer from 0 to 200 cm shows that the highest absolute N losses were assessed for the organic fertilizations (Fig. 3, Supplementary Table S10), which increased with increasing N inputs. The unfertilized controls (U and GM) showed the lowest values for N losses, while moderate losses of approximately 70 kg N ha^{-1} year^{-1} were found for NPK and HU. The proportion of N losses from the total N application (Table 3) had a narrow range of 34% to 55%, except for the U control where the proportion of lost

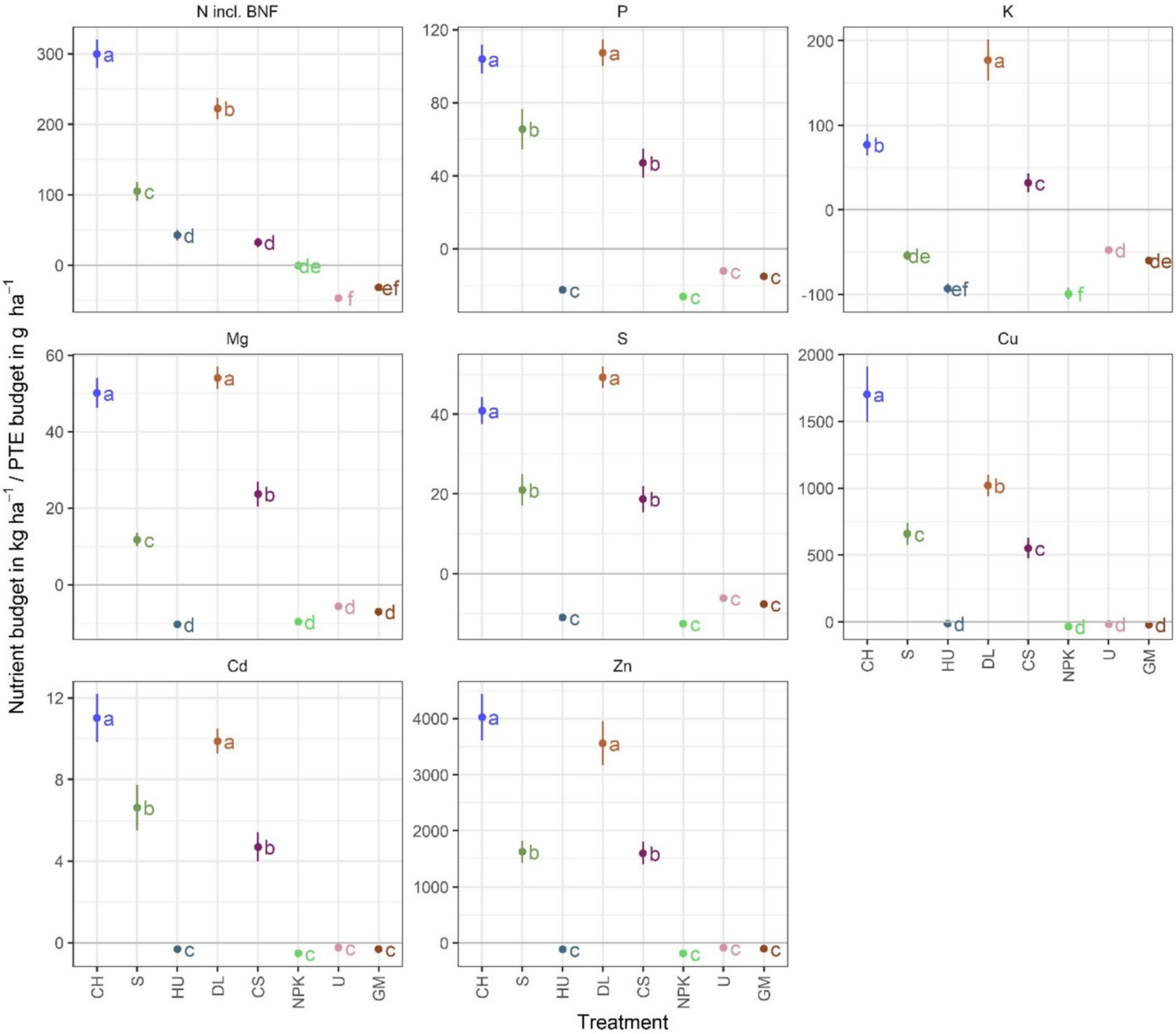

Fig. 2 Measured input–output budgets for the main nutrients in kg ha^{-1} year^{-1} (for N including biological nitrogen fixation (BNF based on model output), P, K, Mg, S) and potentially toxic elements (PTE) in g ha^{-1} (Cu, Cd, Zn). Shown are the means (dots) and the standard errors (lines). The letters show significant differences among the fertilization treatments (alpha = 0.05). For the fertilization abbreviations consult Table 1

N is very high (157%). For all fertilizations, leaching was the main N loss pathway (Fig. 3, Supplementary Table S10), followed by N_2 losses from denitrification, N_2O emissions from nitrification and denitrification, and lastly surface losses, which are mainly NH_3 volatilization from fertilizer application. Yet, the high N surpluses of the fertilizer treatments with high organic matter contents (CH, CHA, SA, DL, and CMA) also resulted in high soil N accumulation and therefore increased soil organic N concentrations. In contrast, the fertilizers with less added organic matter and the unfertilized controls (NPK, HU, U, GM, and CS) resulted in a net mineralization of the soil organic N. Comparing the accelerated fertilizations to the non-accelerated, it can also be observed that the proportion of N lost to the applied N increases with increasing application rates (Table 3).

Carbon flows in the cropping system

A closer look at the C balance as predicted by the *Daisy* model showed that the main inflows are the net

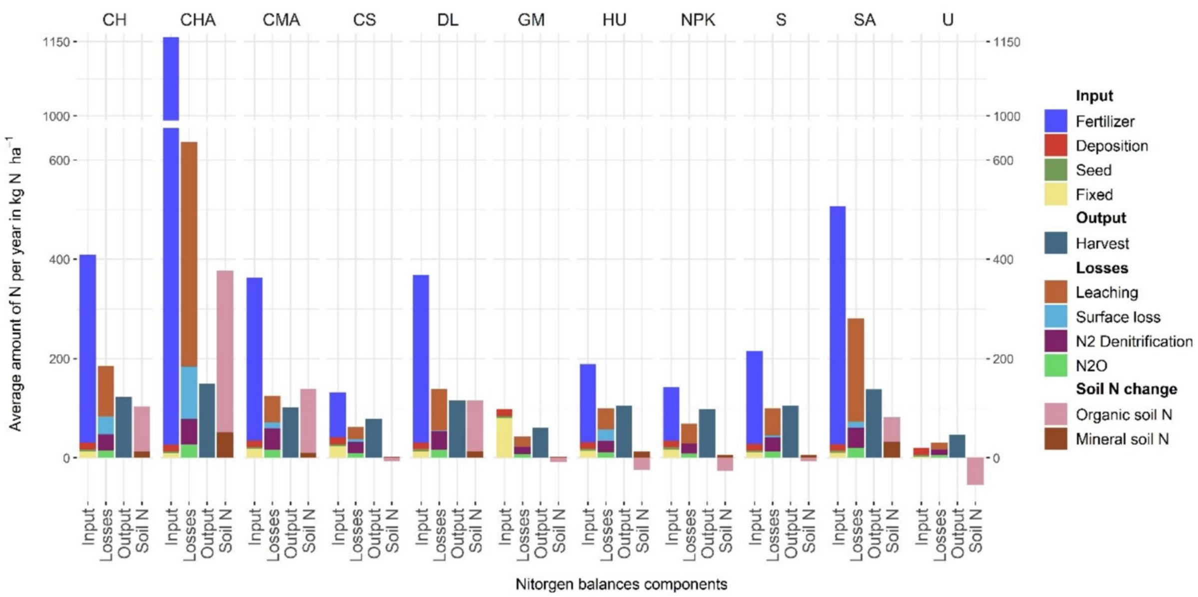

Fig. 3 Nitrogen balances in the soil layer from 0–200 cm for all fertilization schemes divided by input (fertilizer, deposition, seeds, biologically fixated nitrogen (fixed)), output (harvested grain and crop residue), nitrogen losses (through leaching, surface loss (run off and mostly volatilization), N_2 from denitrification, N_2O from nitrification and denitrification) and change in soil N (of organic or mineral nitrogen storage; positive values mean an increase) as simulated by the *Daisy* model. Bars represent the fertilizer treatment annual average. For the treatment abbreviations consult Table 1

Table 3 The annual average nitrogen balance is shown as annual averages of total nitrogen input, losses, output, and changes in soil nitrogen storage change (positive values indicate an increase) as simulated by the *Daisy* model

Fertilization	Input kg ha^{-1}	Losses kg ha^{-1}	Output kg ha^{-1}	Soil N storage change kg ha^{-1}
CH	409	185 [45%]	123 [30%]	101 [25%]
CHA	1158	636 [55%]	149 [13%]	375 [32%]
S	215	100 [47%]	106 [49%]	6 [3%]
SA	507	281 [55%]	138 [27%]	84 [17%]
HU	189	99 [52%]	106 [56%]	− 15 [− 8%]
DL	368	139 [38%]	115 [31%]	114 [31%]
CMA	363	125 [34%]	101 [28%]	139 [38%]
CS	132	62 [47%]	78 [60%]	− 5 [− 4%]
NPK	143	69 [48%]	98 [68%]	− 22 [− 16%]
U	20	31 [157%]	47 [238%]	− 54 [− 275%]
GM	98	43 [44%]	61 [62%]	− 7 [− 7%]

Values are presented in absolute terms and as percentages of the annual average total nitrogen inputs (in parentheses). For the fertilization abbreviations consult Table 1. (input: fertilizer, deposition, seeds, fixated nitrogen; losses: leaching, volatilization, surface loss, N_2 from denitrification, N_2O from nitrification and denitrification; output: harvested products; soil N storage change: organic & mineral soil N storage change)

photosynthesis and fertilizer inputs for the organic fertilizers (Fig. 4). The net photosynthesis was similar for all fertilizations apart from a lower value for the unfertilized control. The third largest input was the bioincorporated C of residues, which consisted mostly of roots, fallen dead leaves and stubble. This

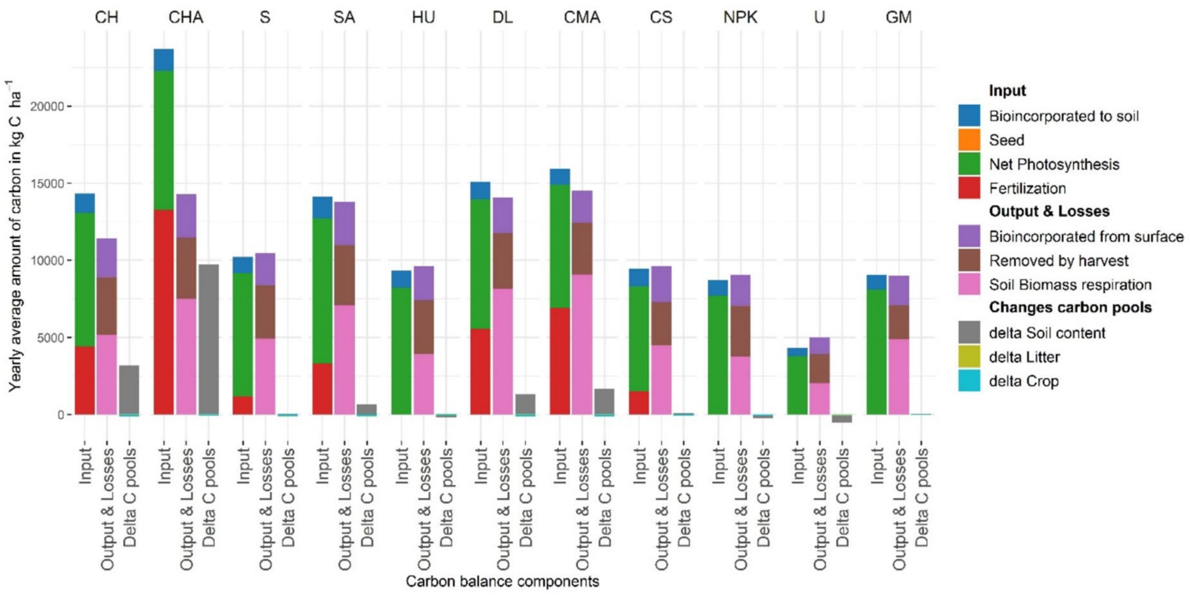

Fig. 4 Carbon balances for all fertilization schemes divided by input (bioincorporated to soil, seed, net photosynthesis, fertilization), and output (bioincorporated from surface, removed by harvest, soil biomass respiration) as simulated by the *Daisy* model. Bars represent the annual average for each fertilization treatment. For the treatment abbreviations consult Table 1

input was mirrored by the loss of surface C through bioincorporation, the smallest losses of C. It was assumed that half of the losses of surface C were transformed into soil C while the other half was respired by the soil microbes. The other main C outflows were microbial respiration and C removed by harvest. For the organic fertilizers the respiration was especially large, even larger than the harvest export.

During the trial period, the measured data as well as the model output indicated that the compost fertilizations resulted in the highest increase in soil C, followed by the other bulky organic fertilizers CMA, DL, SA, and S in descending order (Table 4). The other fertilization schemes (NPK, HU, CS) and GM either did not or only caused a minor change in soil C, whereas the unfertilized control led to a decrease in soil C. Additionally, it is noteworthy that there is considerable variance within the measured values. In relation to the supplied amounts of C, the highest soil C sequestration measured as an increase of soil organic C content per kg supplied C_{org} was achieved with DL (0.75 kg C_{org} per kg supplied C_{org}) followed by compost application (0.47–0.52 kg C_{org} per kg supplied C_{org}), when compared to the NPK fertilizer treatment, respectively. A lower specific C_{org} accumulation was found with sewage sludge (0.20 kg C_{org} per kg supplied C_{org}) and CS (0.15 kg C_{org} per kg supplied C_{org}). The accelerated fertilizations showed distinct lower values.

The *Daisy* model output showed a huge increase in soil C from the CH and CHA fertilization, mainly driven by the pure addition of organic matter (AOM, Table 4) which remained inert (> 90% of soil C in the form of AOM) and was not transformed into the other pools, soil organic matter and soil microbial biomass. The other organic fertilizer transformed a higher proportion of the added organic matter to soil organic matter (SOM, Table 4) and soil microbial biomass (SMB, Table 4). Changes in SMB were less in absolute values, yet they were especially increased in the bulky cattle manures fertilizations (DL and CMA), CHA, and SA, while only the unfertilized control resulted in a decrease. The loss of total soil C (e.g., HU, U, NPK) could be explained by a mineralization of the existing soil organic matter (decreasing SOM, Table 4).

Table 4 Mean change in soil carbon as measured and predicted by the model output for each fertilization as sum over the whole trial period divided by total soil carbon (Soil C), soil organic matter (SOM), soil microbial biomass (SMB) and added organic matter (AOM)

Fertilization	Measured data			Model output			
	Soil C	SE		Soil C	SOM	SMB	AOM
	kg C ha^{-1}	kg C ha^{-1}		kg C ha^{-1}	kg C ha^{-1}	kg C ha^{-1}	kg C ha^{-1}
CH	47,478	3834	b	56,500	1847	310	54,344
CHA	119,448	4602	a	174,883	11,071	999	162,813
S	6078	560	de	743	746	37	−40
SA	23,688	2361	cd	11,485	8422	480	2583
HU	-642	1025	e	−2107	−2819	50	661
DL	25,038	6965	c	23,196	20,330	866	1999
CMA	29,898	3552	bc	29,990	25,718	1198	3074
CS	798	915	e	1176	510	120	545
NPK	1278	1931	e	−2841	−3357	15	501
U	−11,802	5155	e	−8343	−8177	−158	−8
GM	−2082	1983	e	706	132	123	451

A positive value implies an increase over the trial period and vice versa. For the measured data, the fertilization treatment means and standard errors (SE) are given. Letters indicate significant differences between treatment means ($p < 0.05$). For the treatment abbreviations consult Table 1

Discussion

Short-term fertilizer calibrations might overestimate turnover rates in the long term

The *Daisy* model has proven to be a useful tool to simulate long-term effects of the use of recycled fertilizers. The model's prediction of grain dry matter yield, grain N and whole crop yield was equally precise as in other studies. Yin et al. (2017) found IAs ranging from 0.4 to 0.7 for most models in a model comparison. They also observed that major crops that are modelled frequently like winter wheat or spring barley predicted the observed yields more precisely than less frequently modelled crops. This is because more effort has been put into calibrating and validating commonly used crop modules. The predicted values of soil C and N differed slightly from the measured values, especially in the long term. Yet, there was also a huge variation in the measurements. The fertilizer calibrations for sewage sludge and household waste compost were taken from rather short-term incubation experiments (190 or 120–500 days respectively; Bruun et al. 2006, 2016). The results of these calibrations are then extrapolated considerably in time in the simulations of the field experiments and that will obviously lead to some uncertainties. Incubation experiments are an abstraction of field conditions. For example, Kan et al. (2021) found that the sieved soil used in incubation experiments increases mineralization rates temporarily due to a destruction of structure. Further, these organic fertilizers can have very different mineralization dynamics depending on the composition, maturity and bulking material used (Bruun et al. 2006). This could be an explanation for the detected uncertainties in the model prediction and highlights the variability of organic fertilizers. The mineralization rates of N and C are the parameter that causes the most uncertainties in the simulation of organic fertilizers in the *Daisy* model and can be used for sensitivity analysis. Bruun et al. (2006) showed in a sensitivity analysis that changing the mineralization pattern by varying the C/N ratio of the AOM1 pool to a reasonable extent resulted in 0–25% change in N leaching while it did not influence N_2O emissions nor NH_3 volatilization. The CRUCIAL experiments could potentially have been used to recalibrate the parameters for these materials, but attempts resulted in very unreliable estimates as well, given the lack of continuous mineral soil N measurements and the uncertainty in many other areas such as soil organic matter decomposition, crop residue production (below ground and stubble) and degradation of crop residues and yearly variability of the material. The overestimation of the prediction for the GM fertilization is, with high probability, due to cover crop establishment problems, especially sowing issues, under field conditions which cannot be covered by model predictions. Further, models have been shown to be more uncertain in simulating intercropped cultures like grass clover leys due to issues with handling the interspecies competition (Jensen et al. 1999). These cannot be simulated by the model. In conclusion, the calibration of the *Daisy* model was successful; however, the limitations of model predictions need to be considered while interpreting the result. Further validation of the model's performance for determining N leaching and emission fluxes from long-term organic fertilizer application is needed.

Moderate yields of recycled fertilizers are coupled with low relative agronomic efficiency and nutrient and PTE surpluses

The aim of this study was to assess the suitability of recycled fertilizers as substitutes for mineral fertilizers in conventional farming and for animal manures from conventional sources in organic farming systems. Overall, results indicated that HU has a similar performance to NPK. In terms of yield, MFE, nutrient and PTE budgets, it did not differ from mineral fertilization. The only difference was seen for the relative agronomic efficiency, where HU showed lower values. The N in stored human urine consists mainly of ammonia and some urea and organic N (Table 1; Gómez-Muñoz et al. 2017; Martin et al. 2020). Therefore, there were high ammonia volatilization losses due to the liquid form of human urine. Furthermore, stored human urine is very prone to ammonia losses because urea hydrolysis increases pH. In addition, the large ratio of N to the other nutrients and PTEs leads to negative budgets for all nutrients and PTEs except N. Thus, HU must be combined, in a long-term perspective, with other nutrient sources that are low in N but high in other nutrients (e. g., composts) to achieve a balanced system. The main drawback is the very low availability of human urine from source separated wastewater collection. Additionally, stored human urine, as used in the trial, has a high volume per kg N, which makes transportation and application costly. Extraction of nutrients from the raw product could be a relevant measure to make handling easier for farmers, yet energy consumption, which can be extensive depending on technology, and additional costs for farmers need to be considered (Martin et al. 2020).

The other recycled fertilizers, composted household wastes and sewage sludge, were comparable with the cattle manures in terms of yield effect, MFE, relative agronomic efficiency, and nutrient budgets. The normal application rates of CH and S yielded similar to the cattle manures with the slight trend of S having higher yields. However, the amount of total N needed to achieve similar yields varied greatly. This is reflected in the resulting relative agronomic efficiency differences and is most likely due to different proportions of ammonia from total N, C/N ratio and mineralization rate of N and C of the fertilizers (Gutser et al. 2005; Gómez-Muñoz et al. 2017). The high MFE and relative agronomic efficiency of CS can be attributed to the high proportion of N as ammonium (44%). Furthermore, CS similar to NPK and HU, has the essential advantage over the other bulky fertilizers (S, CH, CM, DL) in that they can be applied while the crop is growing. This helps synchronize the N supply with the N plant demand (Pang and Letey 2000). The other organic fertilizers investigated in this study, are rather solid bulky materials, which need to be incorporated into the soil before crop cultivation. This is one reason for the lower efficiency. Additionally, fertilizers like CH, DL and S have lower contents of easily available N compounds like ammonia. The rather high efficiency of CMA, considering the high application rate, is due to the relative high amount of ammonium (24%), while the high efficiency of S is more likely a result of a moderate amount of ammonia coupled with a low C/N ratio, which also hints at a faster and higher net N mineralization. The CH and DL fertilizations show low amounts of ammonium since it volatilizes and immobilizes during the maturing process or storage before field application (Eklind and Kirchmann 2000; Sommer 2001). With regard to a more efficient N use and preserving more nutrients from household waste, anaerobic digestion instead of composting might be a more suitable option. Anaerobic digestion preserves more N from the substrate and thus has a higher efficiency (comparable to slurry or solid manure storage). Moreover, digestates also have the advantage of possible application during the vegetation period (Möller and Müller 2012; Benke et al. 2017; Möller 2018).

The rather low N efficiency of organic fertilizers and the resulting high application rates also caused high nutrient surpluses, especially for N, P, and K. Organic fertilizers are multi-nutrient fertilizers, however the stoichiometry of nutrients does not always match the plant demand. In general, they contain too little N in comparison to the other nutrients, which results in a surplus of the other nutrients if they are used to fulfill the N demand of the crops (Zikeli et al. 2017; Reimer et al. 2020). Compared to animal manures, recycled fertilizer like compost, human urine, and sewage sludge, show lower contents of K and Mg, which might make an additional K source necessary. If organic fertilizers from urban wastes like compost or sewage sludge are to substitute mineral fertilization, there is always the need for an additional source of N to offset the nutrient imbalances and to specifically avoid high P surpluses (Zikeli et al. 2017; Reimer et al. 2024). The additional N could come from BNF through legume or from N rich fertilizers like HU.

The accelerated fertilizations (CHA/SA/CMA) had similar crop yields compared to the NPK fertilization due to the high excess of applied N. Yet, the high application rates resulted in enormous nutrient surpluses and an inefficient use of N. In practice, these application rates would not be possible in most European countries due to legislative limits on nutrient budgets (N and P) or maximum N application rates (Sutton 2011), quite apart from the low use at high costs for the farmers. Nevertheless, the intent of these treatments was never to illustrate farming practice but simulate a longer period of application to investigate the risk of soil PTE accumulations in the long term (Magid 2006).

Some of the substances in organic fertilizers are considered to be potentially harmful to the environment like PTEs (Cu, Cd and Zn). Since the plant demand for PTEs is low or absent, regular application results in excess, which can be seen for the organic fertilizers used in the trial. The CH fertilization shows an especially high surplus of Cu, due to high application rates, while S results in Zn surpluses. The same phenomenon was also observed by Weissengruber et al. (2018) and Möller et al. (2018). The question however remains if these surpluses are harmful. López-Rayo et al. (2016) found that only the soil concentrations of Cu and Zn were elevated in the CHA and SA fertilizations compared to the unfertilized control in the CRUCIAL trial after 10 years of application, which represents more than 100 years of simulated normal applications. Despite these high amounts of PTE inflows, the measured soil concentrations in the accelerated fertilizations were still below half of the suggested threshold values by Tóth et al. (2016). This might be due to PTE leaching to lower soil layers and especially due to the high buffer capacity of the soil. Furthermore, such an accelerated approach does not take into consideration the potential leaching or immobilization of PTEs over time. In addition, López-Rayo et al. (2016) investigated the crop uptake of heavy metals and found only elevated Cd uptake by oat grains in the SA fertilization. In peas shoots they found elevated Zn uptake in the treatments with urban waste fertilization compared to the unfertilized control. Both were far below the EU threshold and considered negligible. The increased Zn concentration could even be regarded as a beneficial side-effect since Zn is an essential element in human nutrition (López-Rayo et al. 2016). This leads to the assumption that—within a certain range–even high surpluses of PTEs do not result in negative consequences to human health and the soil, mainly due to the high buffer function of the soil and the ability of the plants to take up the nutrients they need. Further, soil properties especially pH, structure, and OM play a dominant role in the phytoavailability of PTEs and the crop uptake (Hooda et al. 1997; Sungur et al. 2014).

Yield and efficiency dependent on experiment duration and amount of applied N

Long-term field experiments allow us to investigate trends of such as yield over time. These trends are the result of the long-term effects of the treatments and can be positive (e.g., due to increased soil fertility) or negative (e.g., due to soil nutrient depletion). However, they can also represent effects apart from the main treatment effects like changes in management or cultivars (Reckling et al. 2021). Fertilizations with a high addition of organic matter (CHA, SA, CMA) seem to increase yields in the long term, most likely due to increased soil fertility (Macholdt et al. 2021). By contrast, omitting fertilization (U and GM) decreases the yield, most likely due to soil N depletion and decrease in soil organic matter as observed in this study as well as by Petersen et al. (2010).

Although we observed a depletion of the soil N pool for NPK and HU in this study, an effect on yields over time was not observed. This indicates that fertilizer N and the remaining soil N pool were sufficient for the crop N demand or could be an indicator of crop N uptake from other sources not considered in the *Daisy* model such as non-symbiotic N fixation (Yan et al. 2020).

Regarding the efficiency of N usage, the picture is less clear. In this context, the amount of applied N also plays a major role. At high N application rates there is a reduced yield effect per additional unit N applied. This is known as the reduction of the marginal N efficiency at high N applications due to increased N losses (Raun and Johnson 1999; Fageria and Baligar 2005; Omara et al. 2019). Furthermore, there were significant interactions between the amount of applied N and years of the fertilizations. These interactions are mostly due to a change in management after 2014. The amount of applied N abruptly decreases for CH and CHA and increases for CS, CMA, HU, S and SA in 2015. Afterwards, it stayed almost constant between 2015 and 2020. Thus, the effect of duration of the experiment must be interpreted with caution since they might be skewed due to the different application rates. Additionally, the effect does not seem to be linear, but rather to reach a plateau. The change in efficiency occurred mainly in the short term (first five years, Supplementary Fig. S7). The efficiency for most organic fertilizers, except CH, increased. It takes two or more years until organic fertilizers reveal their full potential due to carry over and residual effects of unmineralized materials and accumulation of mineralized N after crop uptake that is not leached out over winter (Pang and Letey 2000). For CH and CHA, there was a decline in efficiency in the first five years, which could be due to high N immobilization. Fertilizers with a relative excess of C (high C/N ratios) like CH might immobilize N in the soil since they have a wider C/N ratio than the surrounding soil (Möller 2018). However, this effect seems to apply only to the short term. After a while, the immobilization seems to be balanced out by an increased mineralization potential and the efficiency does not decrease further. In the long term (more than five years) all organic fertilizers seem to reach an equilibrium between immobilization caused by new additions and mineralization caused by older additions and no further changes in efficiency occur. The changes observed from 2014 onwards can be attributed to the abrupt above-mentioned change of N application rates. Gómez-Muñoz et al. (2017) found an increase of efficiency by comparing the MFE in the first and tenth year of application for CS, HU, S and SA, while a decrease was observed for CH and CHA. These distinctions between the organic fertilizers can be explained by the C/N ratio and available ammonium (Gómez-Muñoz et al. 2017). A wide C/N ratio lowers the MFE of organic fertilizers while higher amounts of ammonia N increases the MFE (Gutser et al. 2005; Webb et al. 2013).

Organic fertilizers increase soil carbon and nitrogen at the cost of higher nitrogen losses

One of the major advantages of organic fertilizers is the increase in soil fertility through increased soil C and N. Compost showed the highest potential for enhancing soil C and N storage, followed by the straw-rich organic manures (like CMA and DL), and sewage sludge(S and SA), while the materials low in organic matter resulted in a decrease. Thus, organic fertilizers, especially compost, show the potential for mitigating climate change through soil C sequestration (Tully and McAskill 2020). Compost and manure also stimulate soil microbial activity as seen by the increased biorespiration. This is in line with findings of Peltre et al. (2017), who also pointed out that compost and cattle manures are higher in lignin and have a higher proportion of stable C than sewage sludge. In addition, straw was removed from the field in the investigated trial, which reduced the plant residue contribution to increasing soil C. This effect is especially noticeable in treatments without organic fertilization. An increase of soil C is related to other benefits like soil porosity, aggregate stability, and tilth (Grosbellet et al. 2011; Annabi et al. 2011; Peltre et al. 2015), as well as the long-term availability of fertilizer P, especially important in soils with low P contents as found in organic farming (Vermeiren et al. 2021). However, the C sequestration potential of recycled fertilizers, especially compost, needs to be evaluated considering the potential negative environmental impacts due to N_2O and NH_3 emissions. Finally, the potential of soil C sequestration can vary depending on management, climate factors, type of material, and application rate (Hua et al. 2014; Berti et al. 2016). The effect of application rate can be observed in the accelerated treatments where soil C sequestration efficiency decreased with fertilizer C input. In the literature either a linear or logarithmic relation between C input and soil C increase is reported (Hua et al. 2014). A linear relation would result in a constant C sequestration factor independent of the application rate, while other studies including the presented results indicate that at high C input rates the soil C sequestration efficiency declines which is hypothesized to be due to soil C saturation (Zhang et al. 2012; Guo et al. 2014).

The change in organic soil N followed the same pattern as the changes in soil C. For HU and NPK there was a net decrease of total soil N, while it increased for the animal manures and recycled fertilizers. With the increased soil N, the mineralization potential increased as well. This enhances on the one hand soil fertility and crop uptake, but on the other hand, it also leads to higher potential for N losses. Losses of N from the cropping system through leaching, volatilization, surface run-off and denitrification are the main reasons for an inefficient use of fertilizer N. For all fertilizations except NPK, CS and HU, most of the total N inputs were lost from the system, which fits with a global N recovery rate below 50% (Fageria and Baligar 2005). The lower N loss rate could be due to a better synchronization of crop demand and N supply in these fertilizations. The N in mineral N fertilizers, HU and CS has a higher plant availability and can thus be taken up right away after application. The organic N in the other fertilizers needs to be mineralized first, which depends among others on soil temperature and mainly occurs in summer. Since mostly summer cereals are cultivated in the trial, the N uptake in late summer and autumn is limited. This can result in high N losses during that time. Additionally, high amounts of applied N led to the highest N losses in absolute values. Optimizing the amount of applied N is one of the major tools to reduce N losses (Fageria and Baligar 2005; Kühling et al. 2021). This experiment also highlights the shortcomings of using reference values of N availability for organic fertilizers without considering additional factors such as years of continuous application, or C/N ratio of the material.

The greatest loss of N took place through leaching. This is in line with the literature (Gerke et al. 1999; Basso and Ritchie 2005; Fumagalli et al. 2013). Yet, comparing studies directly is always difficult, since the amount of leaching is highly dependent on climate, weather, quality and quantity of added fertilizer, N surplus, soil hydraulic properties and management practices (Gerke et al. 1999; Beaudoin et al. 2005; Basso and Ritchie 2005; Blicher-Mathiesen et al. 2014). This explains the variation in the literature as well as in model predictions. A split application could potentially decrease leaching emissions in climates with a water surplus in the vegetational period since it matches the N supply better with crop demand and mitigates leaching losses (Meisinger and Delgado 2002; Sene et al. 2019). However, this would only be a possibility for non-bulky fertilizers (NPK, CS, HU, or anaerobic digestates). For most fertilizations, surface losses, which were mostly ammonia volatilization during fertilizer application, were the second largest N loss pathway. Fertilizers with high amounts of ammonia N are especially prone to volatilization losses, such as HU. For organic fertilizers which mature before application (CH, CMA), these volatile N losses often happen already before application and are therefore not included in this study. However, these pre-application losses should be considered for a holistic comparison between fertilizer sources and their treatment before field application (Bernstad and la Cour Jansen 2011; Benke et al. 2017). In absolute values the losses due to N_2O emissions from nitrification and denitrification were smaller, yet they can have a high impact on climate change.

Conclusion

Recycled fertilizers present a viable alternative to mineral fertilizers in conventional farming and animal manures in organic farming. However, their use requires careful management to optimize benefits and mitigate drawbacks. Human urine shows promise due to its high yield levels and efficient N use, but its application must be managed to reduce ammonia volatilization. The largest barrier to using stored human urine in agriculture is the lack of knowledge and infrastructure for collection and transportation. Compost and sewage sludge are more available and enhance soil fertility and organic matter but face challenges with N availability and synchronization with plant demand, leading to potential nutrient imbalances and increased N losses. The risk of PTE accumulation from recycled fertilizers exists but is comparable to that from animal manures. This suggests that using primary sources like compost and sewage sludge may be preferable to products from energy-intensive nutrient recycling technologies, which often come at the cost of added organic matter. Nevertheless, further research is needed to improve the N efficiency of recycled fertilizers, particularly those rich in organic matter, and to explore alternative treatments for urban wastes. All fertilizers have their strengths and weaknesses and thus there is no one-fits-all solution. Depending on the individual needs of the farm, a mixture of different recycled fertilizers coupled with optimized management practices could be the optimal solution.

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Author contributions All authors contributed to the study conception and design. Data collection from the field trial was performed by mainly the Section for Plant and Soil Sciences of University of Copenhagen namely J.M. and M.R. Material preparation, the modeling exercise and analysis were performed by M.R. with support of S.B.. The first draft of the manuscript was written by M.R. and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Availability of data and material The data and the parameterization files from the DAISY model exercise are available online under the following link (https://osf.io/pfqm9/?view_only=3269928951e04ec7b4b33c8a87c1f3bf). All necessary data from the CRUCIAL trail is available in the supplementary. Due to ongoing research the full data set can only be obtained by contacting Jakob Magid (jma@plen.ku.dk) or Marie Reimer (reimer.mari@gmail.com). The R code that was used is available under the following link (https://osf.io/pfqm9/?view_only=3269928951e04ec7b4b33c8a87c1f3bf).

Code availability The R code that was used is available under the following link (https://osf.io/pfqm9/?view_only=3269928951e04ec7b4b33c8a87c1f3bf).

Declarations

Conflict of interest The authors have no competing interests to declare that are relevant to the content of this article.

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