IMPROVING INPUTS FOR ORGANIC FARMING

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Table of Contents

1. Executive summary

The use of recycled fertilizer from urban waste could aid in closing the nutrient cycle between urban and rural areas, while also lowering the energy consumption of mineral fertilizer production and the reliance of the organic sector on manures from conventional farming. However, first the recycled fertilizers need to be evaluated in respect to their yield effect, nitrogen (N) efficiency, resulting nutrient budgets, environmental impact, especially N losses and accumulation of potentially toxic elements (PTEs), and their effect on soil fertility in the short and long term. In order to do so, data from a long-term field trail, which investigates fertilization of stored human urine, compost from household waste and sewage sludge in comparison to mineral fertilization, different cattle manures (deep litter, manure and slurry) and unfertilized treatments, and model predictions of the soil-plant-atmosphere model DAISY were evaluated. Human urine performed similar to the mineral N fertilization for yield, N efficiency, and nutrient budget, while sewage sludge and compost were more similar to the animal manures with lower yields, N efficiencies and higher nutrient imbalances, especially P and S surpluses. Contrastingly to human urine, compost and sewage sludge hold a risk of PTE accumulation in the soil. Yet, the effect on plant uptake and soil fertility seemed to be neglectable. Between 34% and 55% of N supply were lost in the fertilized treatments. Nitrate leaching was the main loss pathway, but human urine had relative high losses due to ammonia volatilization. Within the compost and straw rich manure treatments, the surplus of applied N, resulted in increase in soil N storage, which is accompanied with an increase in soil carbon. Especially compost showed high potentials for increasing soil organic matter. The study showed that each fertilizer has advantages and disadvantages, and thus they should be utilized in mixtures according to their strength.

2 Introduction

Due to 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 aid to close the nutrient cycle between urban and rural areas and lower the need for non-renewable fertilizers such as rock phosphate and mineral N fertilizers, whose production is a main 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). However, recent developments, such as the farm to fork strategy, which aims at 25% organic land in the EU by 2030, will increase nutrient scarcity in the future. This brought new attention towards the topic of urban waste fertilizers in organic farming (Løes et al. 2017; Möller et al. 2018; Milestad et al. 2020). Recycling urban wastes as fertilizers for organic farming could enable further growth of the organic sector or help substitute unwanted inputs, often referred as "contentious" such as rock phosphate or conventional manures in organic farming.

Considerable amounts of nutrients can be recycled from urban waste streams. Zoboli et al. (2016), for example, estimated that recycling urban waste materials could substitute P imports by 70%. The biggest sources 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 composts and sewage sludge lead to higher soil C contents (Peltre et al. 2015; Peltre et al. 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 efficiency and control soil borne pathogens (Litterick et al. 2004; Obriot et al. 2016; Vermeiren et al. 2021).

Despite the large potential of recycling fertilizers from urban waste, the current use is limited. The reasons for that are manifold. In many areas there is a lack of infrastructure to collect waste materials for recycling (Ott and Rechberger 2012). Additionally, several urban wastes are not permitted for use in organic farming, due to concerns of contaminations with, among others, potentially toxic elements (PTE) (Løes et al. 2017) and thus farmers are reluctant to use them (Oelofse et al. submitted). Yet, the reputation of recycled fertilizers is worse than they probably actually are. There have been many technical improvements that lowered the contamination load. 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). PTE, especially Cadmium (Cd), pose a risk to human health (Åkesson et al. 2014) and can be considered as a key factor in the evaluation of recycled fertilizers. The concern about PTE also prevents conventional farmers from using recycled fertilizers, so that they use mineral fertilizers instead. Availability and homogeneity can be a problem for recycled fertilizers. Due to their organic character, nutrient content and release can vary.

Recycled fertilizers are most often multi-nutrient fertilizers, whose 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 between nutrients (Möller 2018). Many recycled fertilizers such as composts have a lower nitrogen (N) to phosphor (P) ratio than plant offtake, which can result in an oversupply of P with its negative environmental impacts (Zikeli et al. 2017).

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.

This makes the synchronization of nutrient supply and N demand more complicated and in general results in a lower fertilization effect than mineral fertilization, especially for N (Pang and Letey 2000; Schröder 2014). However, in the literature various results for the N effectiveness of recycled fertilizers can be found (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 available in the long term (Frossard et al. 2016; Schnug and Haneklaus 2016).

An inefficient use of recycled fertilizer N can lead to higher N emissions to the environment in the form of nitrate leaching, nitrous oxide emissions and volatile ammonium losses (Yoshida et al. 2016; Bruun et al. 2016; Gómez-Muñoz et al. 2017). The magnitude of theses N losses are subjected to many influencing factors, such as soil type, weather and climate, application time and soil incorporation of the fertilizers, crop demand and composition of the organic matter (Gerke et al. 1999; Cabrera et al.2005; Cameron et al. 2012; Bruun et al. 2016). Measuring all the different pathways, that N can be lost from the system, in a field trial is difficult and expensive. 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 are able to predict the fate of N after fertilization (Bruun et al. 2006; Yoshida et al. 2016; Bruun et al. 2016). Daisy is an example of such a model (Hansen et al. 2012). DAISY 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 divers crop rotations after application of recycled fertilizers (Yoshida et al. 2016; Bruun et al. 2016).

Yet, to be able to check 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, PTE, soil nutrients and yield effects, take a long time to develop and reach their whole potential (Pang and Letey 2000; Macholdt et al. 2021). The CRUCIAL experiment is a field trial was designed to investigate the long-term effect of recycled fertilizers on soil fertility and the risk of accumulation of contaminations (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 the use in agriculture. But if recycled fertilizers should substitute part of the need of mineral fertilizers in conventional farming, fulfil the nutrient demand and substitute unwanted inputs, such as conventional manures, in organic farming in the future, they need to be tested for their agronomic potential and environmental impact. Thus, this study aims to compare different 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 using data from the CRUCIAL trail and Daisy simulations. The following research questions will be addressed: a) Is the DAISY model an adequate tool to simulate long term use of recycled fertilizers? b) Are recycled fertilizers able to supply N to the crop efficiently and in adequate amounts? c) Does the use of recycled fertilizers result in nutrient imbalances in form of surpluses of P and K? d) Does the use of recycled fertilizer lead to soil pollution with PTE? e) Does the use of recycled fertilizers lead to an increase in N losses in form of ammonia volatilization, N_2O emissions and nitrogen leaching? and f) How does fertilization with recycled fertilizers effect soil organic matter?

3 Methods and Material

3.1 Field experiment

3.1.1 Experimental Design

For this study, data was 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 run by the university of Copenhagen in Denmark (55_400N, 12_180E). The experiment was established in 2002, but after initial soil investigation was remodeled in fall 2002. Since 2003, the trial has been run in the same way. The trial is set up in a random block design with 3 blocks a 11 plots (each 33 m by 27 m). The eleven different treatments are composed of eight different organic fertilizer applications (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) treatment) and three control treatments (Table 1). The control treatments were a mineral fertilizer treatment and two unfertilized controls of which one received green manure as undersown clover grass (*Trifolium* sp. and *Lolium perenne*) most autumns (Table 2). All fertilizers were applied in the amount of the recommended plant-available N (depending on the crop around 100 kg available N ha^{-1} (Table 1) based on mineral N fertilizer replacement as stated by the Danish Fertilizer Regulations (Anon 2013). The accelerated treatments (CMA, CHA and SA) received approximately three times the amount of the normal treatments. The aim of the accelerated treatments is to simulate a longer term effect of application of those fertilizers, and to investigate their effect on soil. The main properties of the fertilizers are listed in Table 1. The fertilizers were applied in general prior to sowing and incorporated into the soil by ploughing, with the exception of NPK, HU and CS, which were applied in spring at the start of growing season.

Table 1: Fertilizer treatments applied and their main properties in the CRUCIAL trial since 2002. Values show the mean over all application years.

Treatment		N_{total} applied (kg N ha^{-1} year^{-1})	DM %	N % DM	NH_4 % DM	C % DM	C/N
CH	Composted household waste	388	66.2	1.95	0.04	23.7	12.1
CHA	CH accelerated	1164	66.2	1.95	0.04	23.7	12.1
S	Sludge	200	25.7	4.75	0.89	31.7	6.67
SA	S accelerated	577	25.0	4.56	0.84	31.0	6.79
HU	Human urine	150	0.45	48.5	39.4	NA	
DL	Deep litter cattle manure	308	34.1	1.84	0.04	34.3	18.7
CMA	Cattle manure accelerated	401	22.0	2.13	0.52	45.8	21.4
CS	Cattle slurry	114	7.25	2.77	1.23	43.6	15.8
NPK	NPK fertilizer	108	100	20.7	9.10		
U	Unfertilized						
GM	Green manure						

The crop rotation was dominated by spring cereals (barley, oats and wheat), but also some winter crops were cultivated throughout the years (Table 2). Additionally, all plots were split in 2008, 2009 and 2010 into an organic and a conventional treatment. For a detailed description of the field experiment consult Magid (2006) and Gómez-Muñoz et al. (2017).

Table 2: Crop sequence of the Crucial trial. A mixture of white cover and ryegrass was undersown as a cover crop in spring.

Year	Crop	Cover crops	Year	Crop	Cover crops
2002	Wheat	all plots	2012	Oats	GM plots
2003	Oats	all plots	2013	Spring barley	GM plots
2004	Spring barley (silage)	none	2014	Spring wheat	GM plots
2005	Spring rape	none	2015	Oats	GM plots
2006	Spring wheat	all plots	2016	Winter wheat	none
2007	Oats	all plots	2017	Spring barley	GM plots
2008	Spring barley	none	2018	Spring wheat	GM plots
2009	Spring barley (silage)	none	2019	Spring barley	GM plots
2010	Ryegrass (silage)	none	2020	Spring wheat	GM plots
2011	Winter wheat	GM plots			

3.1.2 Measurements

Several measurements were taken in the field experiment (Supplementary Table 1). Agronomic yield of grain and straw, or total biomass in case of silage crops, were measured each year. Nitrogen and C content of the harvested material were also determined from 2002 to 2015. The dry matter nitrogen and carbon content of the fertilizers were also measured in the same years, while contaminations with PTE and NH_4 were only assessed from 2002 to 2008. Soil measurements were taken regularly. Soil texture was analyzed in 2008 and 2011 (only six plots). Soil C and N were analyzed in 2001, 2002, 2006, 2007, 2009, 2011, 2013, 2019 (only few plots) and 2020. Mineral soil nitrogen was measured in 2002, 2003 and twice in 2004. PTE levels in the soil were investigated in 2001, 2006, 2009, 2011, and 2013.

3.2 Daisy model description

DAISY is a one-dimensional mechanistic model, which simulates water, nitrogen, carbon, and pesticides in the bioactive zone near the soil (Hansen et al. 1991; Abrahamsen and Hansen 2000; Hansen et al. 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 nitrogen model (simulating nitrification, denitrification and transport of ammonium and nitrate), and a soil organic matter (SOM) model (simulating mineralization of carbon and nitrogen). 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/.

3.3 Model Setup

We used DAISY Model 5.67 for the simulations. The weather data was taken from Taastrup weather station (Svane and Petersen 2021) and the daily values for precipitation, global radiation and temperature from 1991 till 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. The calibration of the organic fertilizers decomposition and N mineralization was based on standard calibration from built-in data within the model (CS, CMA, DL) or based on previous research. 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 treatment was treated as a mineral fertilizer, due to the high mineral N and low dry matter (DM) and C content of the fertilizer. The fertilizer calibrations were checked by comparing simulated and measured values for soil C and total N. If there were discrepancies, parameters for turn-over rate and C/N of the different AOM pools were adjusted to achieve a better fit. The built-in crop modules were used and calibrated to fit the measured yield data by the parameters for maximum photosynthesis efficiency and N efficiency. 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 comparing observed with predicted values to the 1:1 slope as suggested by Piñeiro et al. (2008) and by comparing the measures for root mean squared error (RMSE), and mean absolute percentage error (MAPE), where the measure 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.

3.4 Nutrient budget, nitrogen and carbon balance and nitrogen efficiency

The nutrient budgets for the measured data were calculated as the difference between fertilizer nutrient input and crop output. For N, P, K and the PTEs Cd, Cu, and Zn a simple budget was calculated, where only fertilizers and BNF are considered as inputs and crop offtakes as outputs. For the crop contents measured data or standard values from the literature are used (Grytsyuk et al. 2006; Bachinger and KTBL 2015; Weissengruber et al. 2018). For the nutrient and PTE contents of the fertilizers, measurements done within the CRUCIAL trial are used, and if there were no measurements for the year available, the mean was used instead. In addition to the simple budgets based on the field trial data, a more comprehensive balance for N and C was given by the DAISY output and investigated. Since there is a focus on the loss pathways of nitrogen, the different passages that N can be lost from the system (N leaching, volatilizing, denitrification, surface runoff and N_2O emissions) were investigated with the model output data. Since DAISY estimates only the total N loss by denitrification but does not differentiate between N_2 and N_2O, the SimDen model by Vinther and Hansen (2004) was used to estimate the ratio between N_2 and N_2O (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. The resulting ratios for each treatment can be seen in Supplementary Table 2 and vary between 0.18 and 0.20.

In order to compare the different treatments on their agronomic value, the mineral fertilizer equivalent (MFE) was calculated, where data availability allowed it. The MFE shows the relation of the apparent nitrogen use efficiency of an organic fertilizer compared to mineral N fertilization and is calculated as the following:

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

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

Since measures of N uptake where not available for the whole trial period, agronomic efficiency was used as an alternative measure, which is based on DM yield of the harvested product (grain yield or whole crop yield for silage crops) rather than N uptake and calculated similarly as the following:

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

Results for agronomic efficiency and MFE from the years 2004, 2009 and 2010 were discarded, since the unfertilized treatment resulted in higher yields than the mineral fertilized one. In order to explain the remaining variation between calculated agronomic efficiencies throughout the years, the influence of duration of application, amount of applied N, fertilizer treatment, and cultivated crop was tested in a linear mixed model.

3.5 Statistical analysis

All statistical analyses were done in R (R Core Team 2018). In order to detect significant differences, linear mixed 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 bottle up approach. Data visualisation the results were plotted using the *ggplot2* package.

4 Results

4.1 Calibration of the DAISY model

The DAISY model simulations were first checked for their hydraulic parameter description, and the permeability of the adjacent ground water soil layer was adjusted for the soil type JB5 as described in the DAISY manual (Styczen et al., 2004).

Afterwards, the fit of the fertilizer calibrations were evaluated by comparing the predicted and observed values for organic soil carbon (Figure 1) and total soil nitrogen content (Figure 2). The ovrall fit for soil C was slightly more precise with an MAPE of 10.4% and an IA of 0.94, while soil total N was showed a fit of MAPE=12.4% and IA=0.81 (Supplementary Figure 1). However, the fit differed depending on fertilization treatment. For soil organic C the mean absolute percentage error (MAPE) was below 15% for all treatments except the accelerated compost treatment, which had an MAPE of 25%. The index of Agreement (IA) ranged from 0.18 to 0.93 for all treatments, but was lowest for the GM, HU and NPK treatments (0.18, 0.29, 0.37). Therefore, original calibrations regarding turnover rates and initial fractions of the different organic matter pools in the model (AOM pools) were used. The simulated total amount of N also fitted well to the measured values. The MAPE was below 15% for all treatments besides CHA and ranged between 7%-22%. The IA ranged from 0.24 (CS treatment) to 0.82 (CMA treatment). There seem to be a general trend that with increasing time, the total N in the soil gets underestimated by the model especially for the CH, CHA, S, SA and NPK treatment.

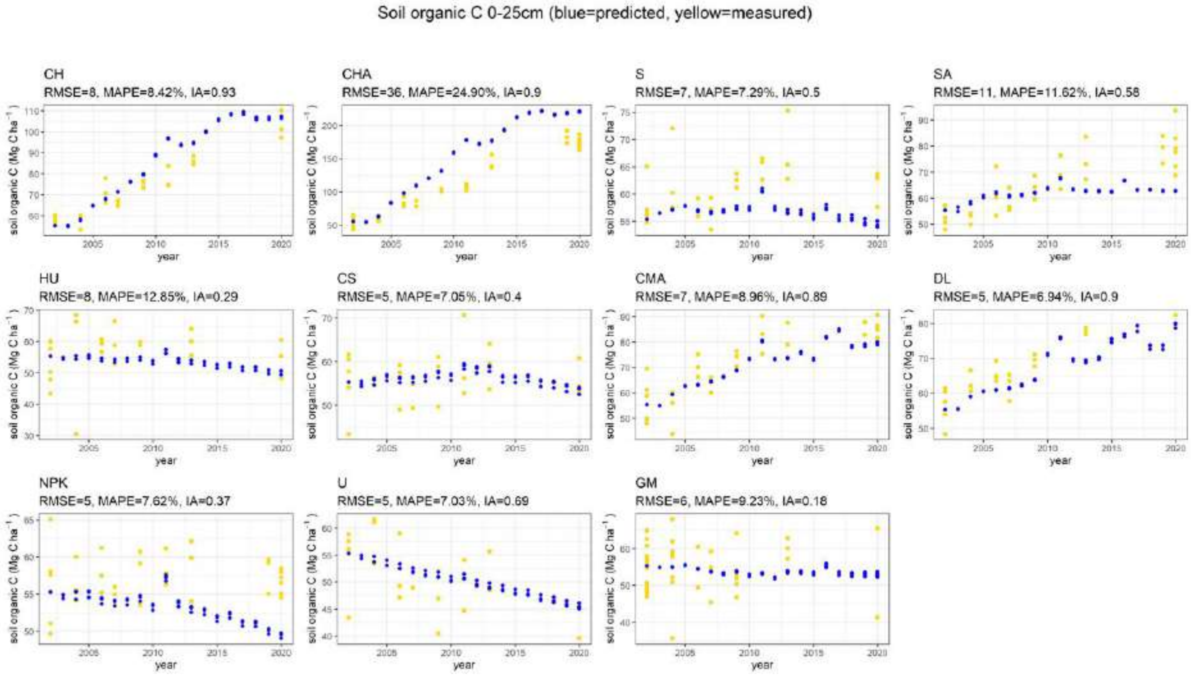

Figure 1: Fit of the predicted model values (blue) and measured field data (yellow) for soil organic matter in the layer of 0-25 cm. Estimates for the fit are given for each treatment in the form of RMSE, MAPE, and index of agreement (IA).

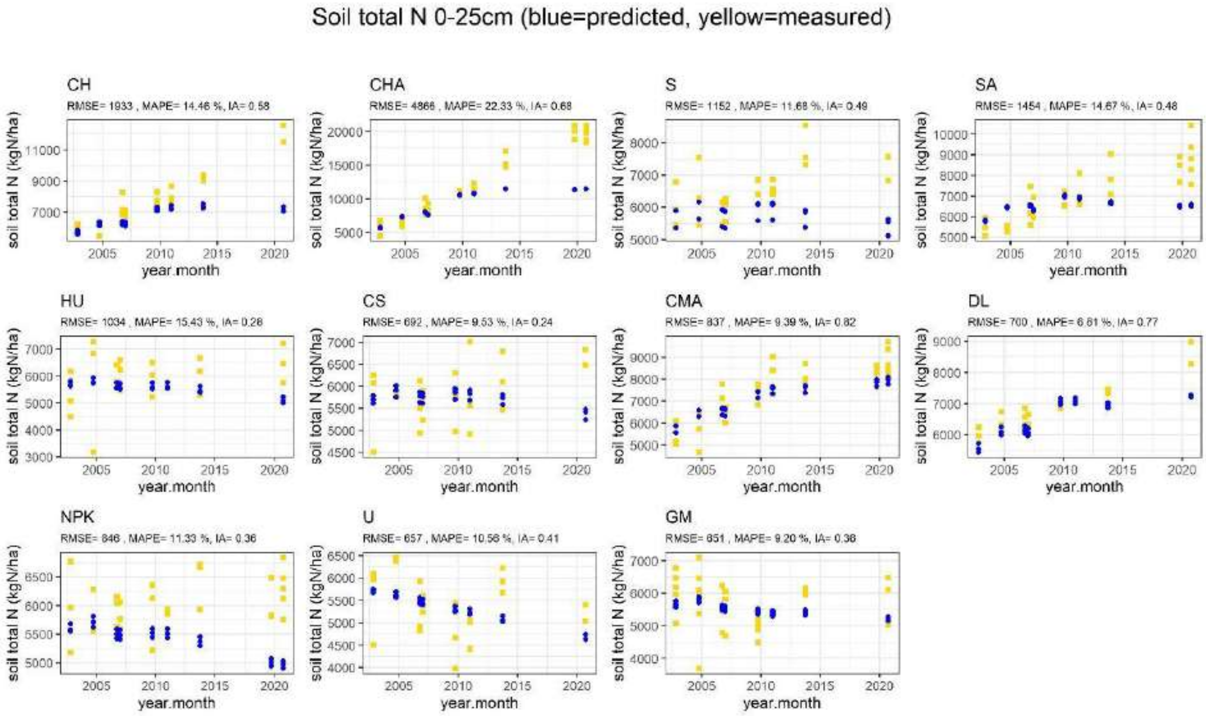

Figure 2: Fit of the predicted model values (blue) and measured field data (yellow) for soil total nitrogen in the layer of 0-25 cm. Estimates for the fit are given for each treatment in the form of RMSE, MAPE, and index of agreement (IA).

The DAISY simulation was able to predict the DM grain yield, whole crop DM yield, and grain N content to an certain extent (Figure 3). The estimates for model fit varied considerable with MAPE ranging ranged from 17% to 35% MAPE and from and an IA ranging from 0.72 to 0.93. The crop models were all adjusted to fit the measured values better. The crop models were all adjusted to fit the measured values better. Specifically, the *FM* parameter, which describes the maximum assimilation rate, and the parameter for the maximum NH_4^+ and NO_3^- uptake per unit of root length (*MxNH4Up* / *MxNO3Up*). There were differences between how well the individual crop models were able to predict the measured variable. The best fit was reached by the spring barley model, while the spring rape seed 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 measures can be found in the Supplementary Table 3.

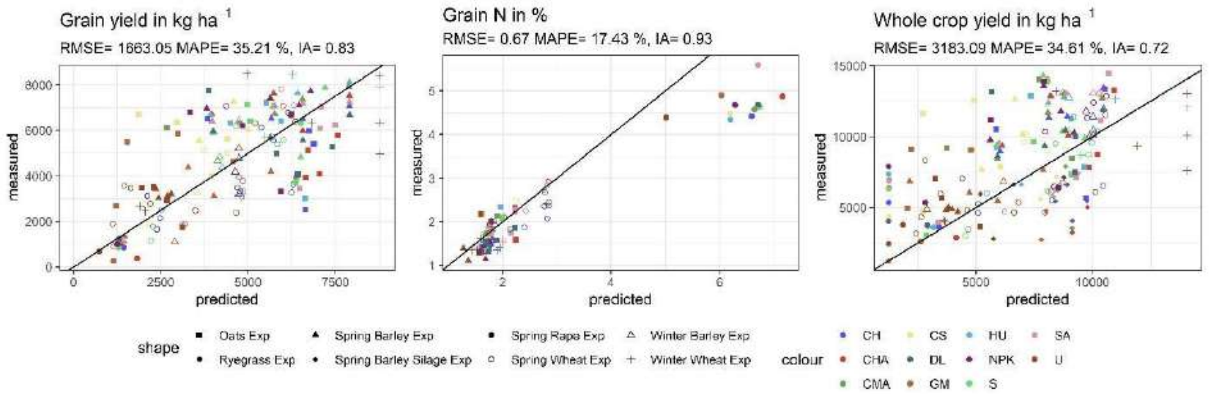

Figure 3: Fit of the predicted model values (x-axis) to the measured data (y-axis) for grain yield (kg DM ha^{-1}), grain nitrogen content (in %) and whole crop yield (kg DM ha^{-1}). Estimates for the fit are given in the form of RMSE, MAPE, and index of agreement (IA). Dots represent a value pair of measured and predicted data, shape the crop species and color the fertilization treatment.

4.2 Yield effect

The different fertilizer regimes resulted in different DM 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 treatments (Figure 3). In order to be able to compare the different years and crops with each other, we used the relative yield in relation to the year average over all treatments. The results of the linear mixed model analysis revealed that not only the fertilization, but also the years of application duration as well as the cropped culture and their interaction with the fertilization had significant influences on the DM yield (Supplementary Table 4). Overall, the highest yields were found for mineral fertilization (NPK), HU, and the accelerated treatments (CHA, SA, CMA), which had a high excess of N (Figure 4). The recycled bulky fertilizers (CH & S) showed lower yields, comparable to that of cattle manures. The unfertilized treatments (U and GM) yielded the lowest yield with only 60% and 71% of the year's average respectively. Due to the interactions of the treatment factor with the other factors, the influence of application duration and cultivated crop were tested on subsets for each fertilizer scheme separately. The application duration only significantly influenced a few treatments. The yield increased with increasing duration for CHA, SA, and CMA while it decreased for the U and GM treatment. The cultivated crops only showed different yield responses for the CHA, SA, HU, NPK, GM and U treatments (Supplementary Figure 2). CHA and SA treatment yielded higher for the winter-sown crops, which was the opposite for the GM treatment.

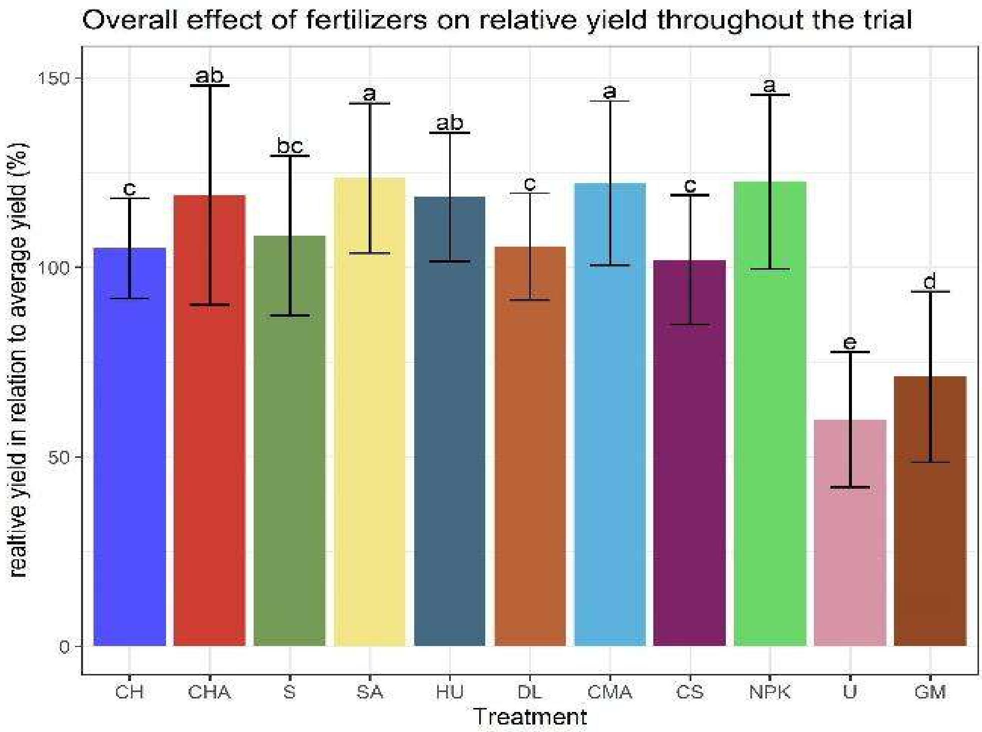

Figure 4: Overall yield effect of the different fertilizer treatments on the relative yield.

The difference between the measured N concentration of the grains were less pronounced than the yield effect, although there were some significant differences (Supplementary Table 4). The CHA and SA treatment showed slightly higher values, while the unfertilized treatment resulted in the lowest grain N concentration (Supplementary Figure 3). The grown crop had a huge influence due to phenological differences. The 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 much higher N concentration, was cropped in the beginning of the trial, while toward the end, mostly spring cereals were grown. The interactions between treatment and the other two factors were not significant.

4.3 Mineral fertilizer equivalent and agronomical N use efficiency

With the aim to assess the N efficiency, the mineral fertilizer equivalent (MFE), which is based on the N yield, was calculated. However, the availability of measured N yields throughout the trial is limited (no measurements after 2014). Therefore, the agronomic efficiency, which is based on the available dry matter yield, was used for a closer investigation over the whole period. Even though measurement periods and measurements that MFE and agronomic efficiency rely on are different, the values and the pattern of the treatment effect are similar (Table 3). The mineral fertilization resulted in the highest values (100% by definition), followed by CS and HU, the fertilizers with the lowest organic matter fraction and the lowest C/N ratio (Table 1). For the more carbon rich fertilizers (CH, S, DL, CMA), S shows significantly higher values than the others, while the accelerated treatments (especially CHA) show lower values. The one noticeable difference between MFE and agronomic efficiency is that S and SA show higher values for MFE than for agronomic efficiency.

In order to assess effects on application duration, amount of applied N, cultivated crop and their interactions, the agronomic efficiency was analyzed in a linear mixed model (Model: Agronomic efficiency ~ Treatment*Years of application*Applied N amounts *Crop; Supplementary Table 5). All factors investigated were significant. The crop factor resulted in only few differences with oats having the highest agronomic efficiency (55%, a) followed by winter barley (49%, ab) and finally winter wheat, spring barley and spring oilseed rape (45-41%, b). Since the interactions between treatment and the duration and amount of application were also significant, we analyzed these factors for each treatment separately (Model: Agronomic efficiency ~ Years of application*Applied N amounts +Crop; Supplementary Table 6). The amount of N applied showed significant effects for all treatments besides CHA, while the duration of application showed only significant influences for CHA, S, HU, and CS. Yet, for the CHA, HU and CS treatment, there were significant interactions between the applied N amount and the duration of the application, which hinders the interpretation of the factors on their own. Thus the only treatment, which shows a pure duration effect is S, where the agronomic efficiency decreased with longer application duration. Yet, data provided in Supplementary Figure 4, indicated for most organic fertilizers an increase of efficiency during the first 5 years, while CH decreases.

Table 3: Means for MFE (mineral fertilizer equivalent in %) and agronomic efficiency (in %) as affected by Treatment.

Treatment effect on MFE (%)									
Treatment	Estimate	Std	r	Min	Max	Q25	Q50	Q75	groups
CH	19.9	7.8	27	9.5	38.9	13.5	17.6	24.4	cd
CHA	10.4	6.6	27	-14.5	18.2	9.3	11.4	15.2	d
S	70.1	32	27	7.8	135.8	45.4	70.6	94.1	b
SA	38.2	15.4	27	5.4	73.7	28.1	38.9	46.9	c
HU	81.1	24.5	26	37.3	132.6	64.9	74.9	94.6	ab
DL	37	36.3	27	3.5	196.4	17.6	26.1	43.5	c
CMA	37	16.9	24	11.5	72	27.1	34.4	46.8	c
CS	92.7	57.5	26	17	282.7	54.2	82.9	107.2	a
NPK	100	0	27	100	100	100	100	100	a
Treatment effect on agronomic efficiency (%)									
Treatment	Estimate	Std	r	Min	Max	Q25	Q50	Q75	groups
CH	24.6	10.7	45	5.2	48.4	15.6	25.4	31.7	d
CHA	10.2	5.6	45	-4.2	26.3	6.9	10.3	12.7	e
S	54.8	31.7	45	-19.5	146	35.6	54.2	69.9	c
SA	22.4	11.1	45	2.5	66.3	14.3	21.6	28.1	de
HU	70.4	27.9	45	26.1	143.9	48.5	68.2	85.6	b
DL	32.5	12.5	45	8.5	68.2	23.6	31.6	37	d
CMA	30.9	13.9	42	7.6	73.8	22.3	28.7	39.1	d
CS	78.4	47.8	45	-9.9	234.5	49.6	74.8	95	b
NPK	100	29.4	45	-19.1	207.6	91.7	101	106.5	a

Model: MFE/AE (%) ~ treatment + years of application + amount of applied N + cropped culture + years of application : treatment + amount of applied N : treatment + cropped culture : treatment + block

4.4 Nutrient budgets

Due to their organic nature, all of the organic fertilizers are multi-nutrient fertilizer. In order to investigate which fertilization scheme resulted in the most balanced nutrient management, the nutrient budgets based on the CRUCIAL field trial data were compiled. The accelerated treatments (CHA / SA

/ CMA) were excluded, since an application of approximately three times the realistic values resulted in high budget surpluses for all nutrients (e.g.; approx. 1000 / 500 / 250 kg N ha-1 year-1 respectively). In general, fertilization with a more carbon 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 (Figure 5). The HU treatment performed very similarly to the NPK treatment. The unfertilized controls (U and GM) resulted in deficits for all nutrients. The pattern for the PTEs is similar, the organic fertilizers show surpluses while the other slight deficits. CH had the highest surplus for all PTEs, with the largest one for Cu.

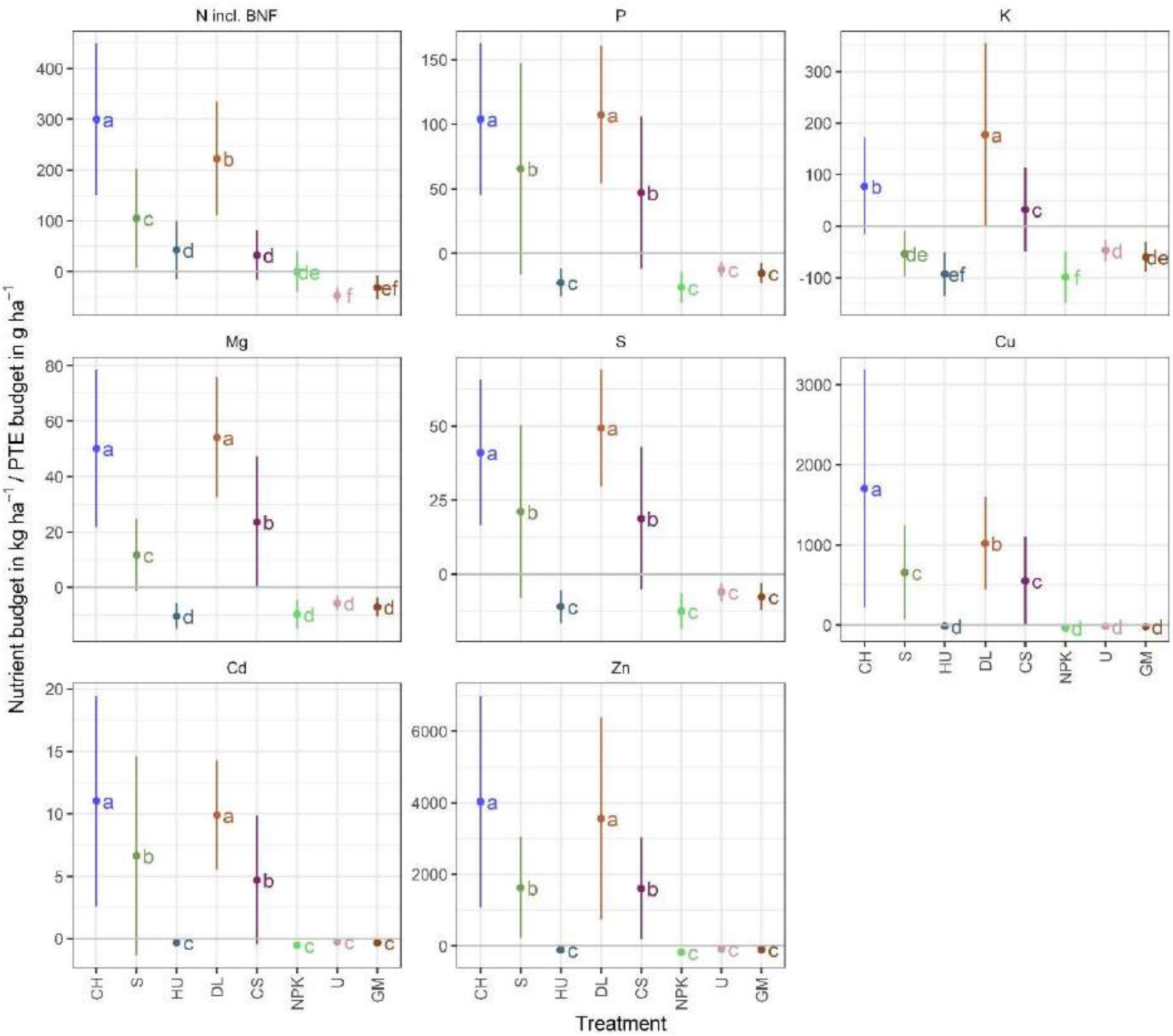

Figure 5: Input-output budgets for the main nutrients in kg ha-1 (for N including biological nitrogen fixation (BNF), P, K, Mg, S) and potentially toxic elements (PTE) in g ha-1 (Cu, Cd, Zn). Shown are the means (dots) and the standard deviation (lines). The letters show significant differences between the treatments (α=0.05).

4.5 Nitrogen balance

Concerning the high N surpluses as described in section 4.4, 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 0-200 cm shows that the highest absolute N losses were assessed for the organic treatments (Figure 6, Supplementary Table 7), which increase 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} $year^{-1}$ was found for NPK and HU. The values of the proportion of N losses from the total N application (Table 4) show a small range (34%-55%), with the exception of the U treatment where the proportion of lost N is very high (157%). For all treatments, leaching was the main N loss pathway (Figure 6, Supplementary Table 7), followed by N_2 losses from denitrification, N_2O emissions from nitrification and denitrification, and lastly surface losses, which are mainly volatilization from fertilizer application. Yet, the high surpluses of N of the organic fertilizer treatments resulted also in high soil N accumulation. Accordingly to the modelling output, especially soil organic N increased in the CH, CHA, SA, DL, and CMA treatments. Contrastingly, the less organic fertilizers NPK, HU, U, GM, and CS resulted in a net mineralization of the organic soil N. Comparing the accelerated treatments 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 4).

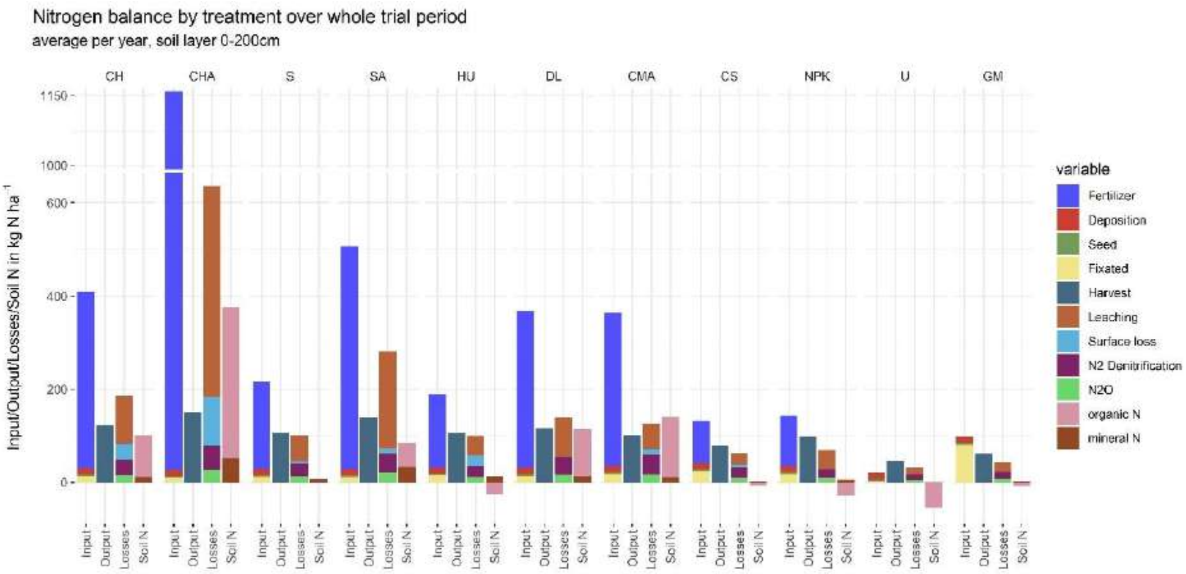

Figure 6: Nitrogen balances for all fertilization schemes divided by input (fertilizer, deposition, seeds, fixated nitrogen), output (harvested product), nitrogen losses (through leaching, surface loss (mostly volatilization), N_2 from denitrification, N_2O from nitrification and denitrification) and change in soil N (of organic or mineral nitrogen storage; positive values mean an increase). Bars represent the treatment yearly average.

Table 4: Average yearly nitrogen input, losses, output, and soil nitrogen storage change (positive values mean an increase) and the proportion of losses, output and soil N change in relation to the total input. (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 change: organic & mineral soil N storage change)

Treatment	Input	Losses	Output	Soil change	% Losses	% soil change	% Output
	kg ha^{-1}	kg ha^{-1}	kg ha^{-1}	kg ha^{-1}	% input	% input	% input
CH	409	185	123	101	45	25	30
CHA	1158	636	149	375	55	32	13
S	215	100	106	6	47	3	49

SA	507	281	138	84	55	17	27
HU	189	99	106	-15	52	-8	56
DL	368	139	115	114	38	31	31
CMA	363	125	101	139	34	38	28
CS	132	62	78	-5	47	-4	60
NPK	143	69	98	-22	48	-16	68
U	20	31	47	-54	157	-275	238
GM	98	43	61	-7	44	-7	62

4.6 Carbon balance

One of the many advantages of organic fertilizers is the potential to increase soil carbon. A closer look at the carbon balance (Figure 7) shows that the main flows are inputs of C are the net photosynthesis and fertilizer for the organic fertilizers. The net photosynthesis is similar for all treatments with an exception of a lower value for the U treatment. The third largest inflow was the bioincorporated C of residues, which is in absolute values immensely lower than the other two. The main C outflows were soil microbial biomass respiration and the carbon removed by harvest. For the organic fertilizers the biomass respiration was especially large, even larger than the harvest export. The third outflow was the loss of C on the surface through bioincorporation, however most of it can be found as a direct inflow in the category “bioincorporated to soil”. The change in soil C stock is equivalent to the difference between out- and inflows of C in the system.

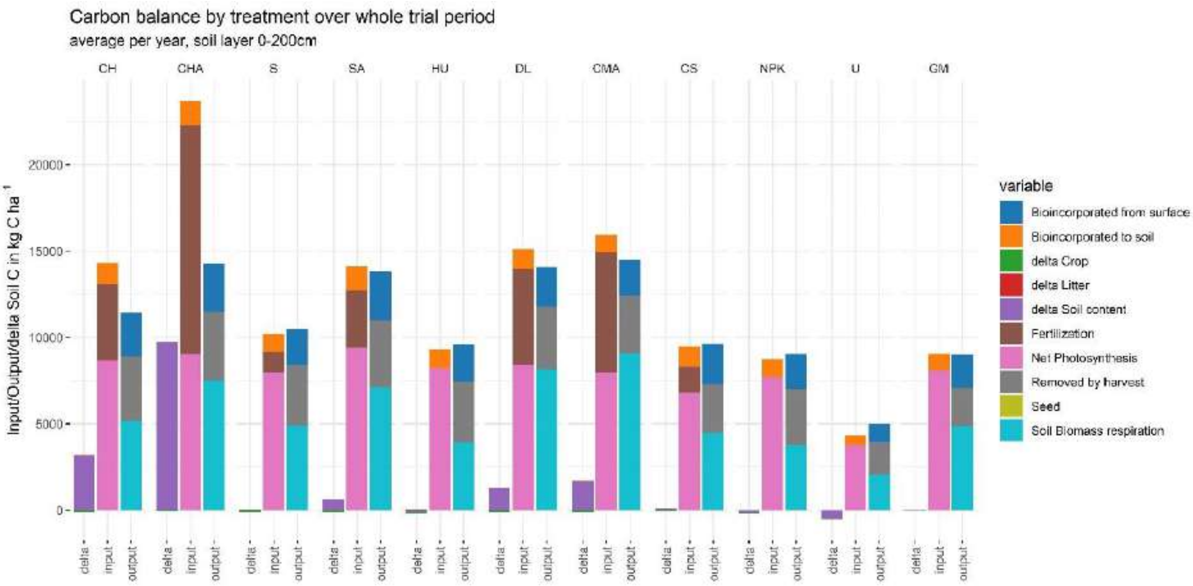

Figure 7: Carbon balances for all fertilization schemes divided by change in system C storage (delta soil, delta litter aka residual on soil surface, delta crop); input (bioincorporated to soil, seed, net photosynthesis, fertilization), and output (bioincorporated from surface, removed by harvest, soil biomass respiration). Bars represent the treatment yearly average.

During the trial period, the measured data as well as the model output indicates that the compost treatments resulted in the highest increase of soil C, followed by the other bulky organic fertilizers in descending order CMA, DL, SA, and S (Table 5). The other fertilization schemes (NPK, HU, CS) and

GM treatment did almost not result in a change of soil C, while the unfertilized treatment resulted in an apparent decrease in soil C. It is also noticeable that there is a huge variance within the measurements.

A closer look at the change in soil C is made possible by the DAISY model output. It shows that the huge increase of soil C of the CH and CHA treatment, mainly driven by the pure addition of organic matter (AOM, Table 5), while the other organic fertilizer transformed a higher proportion of the added organic matter to soil organic matter. The loss of total soil C (e.g., HU, U, NPK) can be explained by a mineralization of the existing soil organic matter. Changes in soil microbial biomass were less in absolute values, yet they were especially increased by the bulky cattle manures (DL and CMA), CHA, and SA., while only the U treatment resulted in a decrease.

Table 5: Mean change in soil carbon as measured and predicted by the model output for each treatment 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). A positive value implies an increase over the trial period and vice versa. For the measured data, the standard deviation (std) and statistical differences (as indicated by letters) are given.

Treatment	Measured data			Model output			
	Soil C	std		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	47478	6641	b	56500	1847	310	54344
CHA	119448	7972	a	174883	11071	999	162813
S	6078	3345	de	743	746	37	-40
SA	23688	8928	cd	11485	8422	480	2583
HU	-642	6152	e	-2107	-2819	50	661
DL	25038	1776	c	23196	20330	866	1999
CMA	29898	971	bc	29990	25718	1198	3074
CS	798	4089	e	1176	510	120	545
NPK	1278	1585	e	-2841	-3357	15	501
U	-11802	3434	e	-8343	-8177	-158	-8
GM	-2082	12064	e	706	132	123	451

5 Discussion

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

The DAISY model has proven to be an adequate tool to simulate long term use of recycled fertilizers. The model's prediction of grain DM yield, grain N and whole crop yield was similar precise as in other studies. Yin et al. (2017) found IAs ranging from 0.4 to 0.7 for most models within a model comparison. They also observed, that major crops that are modelled frequently like winter wheat or spring barley predicted the observed more precise than less frequently modelled crops like in our case spring oilseed rape. 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; Bruun et al. 2016). The results of these calibrations are then extrapolated quite much in time in the simulations of the field experiments and that will obviously lead to some uncertainties. Incubation experiments are also 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, organic fertilizers can have very different mineralization dynamics depending on the composition, maturity and bulking material used (Bruun et al. 2006). The crucial experiments could potentially have been used to recalibrate the parameters for these materials but we estimate this would have resulted in very unreliable estimates as well given the uncertainty in many other areas such as SOM decomposition, crop residue production and degradation of crop residues. The overestimation of the prediction for the GM treatment is, with high probability, due to cover crop establishment problems under field conditions. These cannot be simulated by the model. In conclusion, the calibration of the DIASY model was successful, however the limitations of model predictions need to be considered while interpreting the result.

5.2 High yields of recycled fertilizers are coupled with low agronomic efficiency and nutrient and PTE surpluses

The aim of this study was to assess the suitability of recycled fertilizers as substitutes of mineral fertilizers in conventional farming and of animal manures from conventional provenience in organic farming systems in the future. Overall, results indicated that human urine has a similar performance as mineral fertilization. In terms of yield, MFE, nutrient and PTE budgets, it did not differ from mineral fertilization. The only difference can be seen for the agronomic efficiency, where human urine showed lower values. The N in human urine consists of mainly urea and ammonia N (Table 1, Gómez-Muñoz et al. 2017; Martin et al. 2020). Urea needs to be mineralized first and is therefore less rapidly available and will be used later in the growing season. This might lead to a high N yield but lower dry matter yield, and thus a lower agronomic efficiency compared to mineral fertilizer equivalent.

In addition, large ratio of N to the other nutrients and PTEs leads to negative budgets for all nutrients and PTEs besides N. Thus, HU must be combined, on a long term perspective, with other nutrient sources low in N, but high in other nutrients (e. g. composts) in order to achieve a balanced systems. The main drawback is the very low availability of human urine from source separated waste water collection. Additionally, stored human urine, as used in the trial, has a high volume per kg N, which makes transportation and application costly. Stripping the nutrients from the raw product could be a relevant measure to make handling easier for farmers, yet energy consumption, which can be depending on technology extensive, and additional costs for farmers need to considered (Martin et al. 2020).

The other recycled fertilizers, compost household wastes and sewage sludge, were comparable with the cattle manures. 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 differs highly. This is reflected in the resulting agronomic efficiency differences and is most likely due to different proportions of ammonia from total N, C/N ratio and turn-over rate of N and C of the fertilizers (Gutser et al. 2005; Gómez-Muñoz et al. 2017). The high efficiency of CS can be attributed to the high proportion of N as ammonium (44%). Further, CS has the essential advantage towards the other fertilizers, that it 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 compost, deep litter and sewage sludge have lower contents of easily available N compounds like ammonia. The rather high efficiency of CMA, considering the high application rate, is due to 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 towards a fast mineralization and lower immobilization rate. CH and DL 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 regards 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 has 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 caused also high nutrient surpluses, especially for N, P, and, K. Organic fertilizers are always 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). In comparison to animal manures, the recycled fertilizer shows lower contents of K and Mg, which might make an additional K source necessary. If organic fertilizers from urban wastes should substitute mineral fertilization or animal manures, there is always the need for an additional source of N (e.g. BNF) to balance out the imbalances between nutrients.

The accelerated treatments (CHA / SA / CMA) yielded even similar results to the NPK treatment 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 maximal N application rates (Oenema et al. 2011), quite apart from the low use at high costs for the farmers. However, 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 accumulation in the long term (Magid 2006).

Few 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, a regular application results in excess, which can be seen for the organic fertilizers used in the trial. CH shows an especially high surplus of Cu, due to high application rates, while S results in Zn surpluses. The same phenomena 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 concentration of Cu and Zn were elevated in the accelerated compost and sewage sludge treatments compared to the unfertilized control in the CRUCIAL trial after 10 years of application, which represents more than 100 years of simulated normal applications. In spite of these high amounts of PTE inflows, the measured values in the accelerated treatments were still below half of the suggested threshold values by Tóth et al. (2016). Furthermore, such an accelerated approach does not take into consideration the potential leaching of PTE´s 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 accelerated sewage sludge treatment. In peas shoots they found elevated Zn uptake the treatments with urban waste compared to the unfertilized treatment. Both were far below the EU threshold and considered negligible. The increased Zn concentration could even be regarded as a beneficial side-effect due to the fact that 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. Further, soil properties especially pH, structure, and OM play a dominant role in the availability of PTEs and the crop uptake (Hooda et al. 1997; Sungur et al. 2014).

5.3 Yield and efficiency dependent on application duration and amount of applied N

Long-term field experiments allow us to investigate trends of e.g. 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 depletion). However, they can also represent effects apart from the main treatment effects like changes in management or cultivars (Reckling et al. 2021). Treatments 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 (Macholt 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 (Petersen et al. 2010). Mulvaney et al. (2009) also claimed that mineral N fertilization depletes the soil N pool and thus in the long term results in future higher needs of N fertilization or lower yields. 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.

Regarding the efficiency of N usage, the picture is less clear. Here, the amount of applied N plays also a major role. Higher N application rates reduced the efficiency due to increased potential N losses and lower yield increases with increasing N application, which is also highly supported by the literature (Raun and Johnson 1999; Fageria and Baligar 2005; Omara et al. 2019). Further, there were significant interactions of applied N amount and duration of the treatment. 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 treatment application must be interpreted with caution, since they might be skewed due to the different application rates. Further, the effect does not seem to be linear, but rather result in a plateau function. The change in efficiency occurred mainly in the short term (first 5 years, Supplementary Figure 4). 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 effects of unmineralized materials and accumulation of mineralized N after crop uptake that is not leached out over winter (Pang and Letey 2000). For compost we see a decline in efficiency in the first 5 years, which could be due to high N immobilization. Fertilizers with a relative excess of C (high C/N ratios) 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 increase mineralization potential and the efficiency does not decrease further. In the long term (more than 5 years) all organic fertilizers seems to reached be an equilibrium and no further changes in efficiency occur. The changes observed from 2014 onwards can be distributed 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 cattle slurry, human urine and sewage sludge, while a decrease for compost. These distinctions between the organic fertilizers can be explained by the C/N ratio and available ammonium (Gómez-Muñoz et al. 2017).

5.4 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 of 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 and sewage sludge, 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 manures also stimulate soil microbial activity as seen by the increased biorespiration. This is in line with findings of Peltre et al. (2017), who also point out that compost and cattle manures are higher in lignin and have a higher stability 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 the treatments without organic fertilizers. 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 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 by recycled fertilizers, especially compost, needs to be evaluated towards the potential negative effects environmental impacts due to N_2O and NH_3 emissions. Further, the potential of soil C sequestration can vary depending on management, climate factors and type of material (Berti et al. 2016).

The change in organic soil N followed the same pattern as the changes of soil C. For HU and NPK we see a net decrease of total soil N, while it increases for the animal manures and recycled fertilizers. With the increased soil N, the mineralization potential increased as well. This enhances on the one hand the 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 (Fageria and Baligar 2005). For all treatments besides NPK, CS and HU, the losses made up the biggest proportion of the total N inputs, 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 treatments. 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 N in the other fertilizers needs to be mineralized first, which is depending among others on soil temperature and mainly occurs in summer. Since mostly summer cereals are cultivated in the trail, 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 lead 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).

The greatest loss of nitrogen 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 an 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). Yet, this would only be a possibility for non-bulky fertilizers (NPK, CS, HU, or digestates). For most treatments, surface losses, which were mostly ammonia volatilization during fertilizer application, was 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 taken into account 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 due to their greenhouse gas characteristics.

6 Conclusion

The results of the study suggest that recycled fertilizers are able to substitute mineral fertilization in conventional farming and organic fertilizers from conventional origin in organic farming to a certain extent. All recycled fertilizer treatments investigated in the study are suitable nutrient sources but show different advantages and disadvantages.

Fertilization with human urine resulted in high yield levels and efficient N use comparable to mineral fertilization. The loads of PTEs are also neglectable. Yet, management practices needs to be optimized to mitigate the relative high climate relevant N losses through ammonia volatilization. Further, human urine is mainly a pure nitrogen fertilizer and needs to be paired with other nutrient sources in order to supply crops sufficiently with the other major plant nutrients P, K, S, and Mg. An optimal paring partner could be compost or sewage sludge. Contrastingly to human urine, both fertilizers enhance the soil fertility through increase of soil organic matter and N mineralization potential. In addition, they show a surplus of especially P and S, in relation to available N, thus making an additional N source necessarily to avoid nutrient imbalances. Both of them show lower yields and lower N efficiency, due to lower plant N availability. Which leads to the main obstacle in using sewage sludge and especially compost as N fertilizers, which is the synchronization of N supply and N plant demand. This is also the main factor for the increased N losses through nitrate leaching. Further research on how management practices can enhance the N efficiency of recycled fertilizer, rich in organic matter, is therefore urgently needed. Additionally, different treatments of urban wastes, like anaerobic digestion of household waste, could aid in enhancing the N efficiency.

The risk of accumulation of PTEs due to high loads from recycled fertilizers, still exists for compost and sewage sludge, but is similar to the risk associated with animal manures. Thus a substitution of animal manures with recycled fertilizers would not increase the PTE accumulation risk. Further, it seems not to pose a threat to soil fertility or human health, at least in regions with a positive climatic water balance or drained soils.

Concluding it can be said, that all fertilizers have their strength and weaknesses and thus there is no one-fits-all solution. Depending on the individual needs of the farm, a mixture of different fertilizers coupled with optimized management practices could be the optimal solution.

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8 Supplementary

8.1 Supplementary Table

Supplementary Table 1: Measurement schema for the Crucial trial.

	Harvest data		Fertilizers				Soil data					
Year	yield	N	DM	C & N	NO3 & NH4	PTE	Texture	Bulk density	C_{org}	N	N_{min}	PTE
2001									x	x		x

2002	x	x	x	x	x				x	x	x	
2003	x	x	x	x	x	x					x	
2004	x	x	x	x	x	x					2x	
2005	x	x	x	x	x	x						
2006	x	x	x	x	x	x			x	x		x
2007	x	x	x	x	x	x			x	x		
2008	x	x	x	x	x	x	x					
2009	x	x	x	x					x	x		x
2010	x	x	x	x								
2011	x	x	x	x			x*	x*	x	x		x
2012	x	x	x	x								
2013	x	x	x	x					x	x		x
2014	x	x	x	x								
2015	x	x	x	x								
2016	x											
2017	x											
2018	x											
2019	x								x*	x*		
2020	x		x						x	x		

*only few plots

Supplementary Table 2: Ratio of N_2O losses to total denitrification (N_2O / (N_20+N_2)). Treatment abbreviations: CH= compost from household waste, CHA= accelerated CH, CMA= cattle manure accelerated, CS= cattle slurry, DL= deep litter from cattle, HU= human urine, NPK= mineral nitrogen fertilization, S=sewage sludge, SA= S accelerated, U= unfertilized, GM= green manure

Treatment	N_2O / (N_20+N_2) ratio
CH	0.186
CHA	0.183
CMA	0.186
CS	0.191
DL	0.187
HU	0.198
NPK	0.200
S	0.188
SA	0.185
U	0.200
GM	0.200

Supplementary Table 3: Model fit evaluation parameter (root mean squared error (RMSE), and mean absolute percentage error (MAPE) and index of agreement (IA)) for different measurements for all observed and simulated values and separated by treatment and crop.

Treat.	Measurement	Model fit RMSE	MAPE	IA
	Grain DM yield	4653.71	51.25	0.59
	Grain N %	0.73	17.75	0.92
	Grain N yield	41.69	52.11	0.6
	Whole crop DM yield	3417.01	89.56	0.62
	Whole crop N %	81.3	4924.59	0.01
All	Whole crop N yield	88.49	97.14	0.4
	Grain DM yield	1884.5	42.73	0.76
	Grain N yield	60.21	82.09	0.13
	Soil Carbon	8281.43	8.42	0.93
CH	Soil N	1855.25	15.65	0.6
	Grain DM yield	1628.46	52.67	0.83
	Grain N yield	58.61	106.17	0.45
	Soil Carbon	35546.59	24.9	0.9
CHA	Soil N	5253.33	21.92	0.67
	Grain DM yield	1370.59	28.85	0.84
	Grain N yield	28.6	32.91	0.42
	Soil Carbon	6611.6	7.29	0.5
S	Soil N	1157.42	11.22	0.43
	Grain DM yield	1413.08	26.07	0.86
	Grain N yield	41.88	36.6	0.48
	Soil Carbon	10850.44	11.62	0.58
SA	Soil N	1340.66	13.44	0.48
	Grain DM yield	1592.01	25.61	0.82
	Grain N yield	33.71	34.53	0.23
	Soil Carbon	8346.4	12.85	0.29
HU	Soil N	820.56	11.51	0.33

Treat.	Measurement	Model fit RMSE	MAPE	IA
	Grain DM yield	1949.28	30.74	0.68
	Grain N yield	59.41	73.09	0.04
	Soil Carbon	5297.44	6.94	0.9
DL	Soil N	483.48	4.84	0.83
	Grain DM yield	1491.83	25.42	0.84
	Grain N yield	33.28	36.91	0.22
	Soil Carbon	7351.7	8.96	0.89
CMA	Soil N	763.03	10.12	0.85
	Grain DM yield	1955.48	26.64	0.67
	Grain N yield	35.37	30.97	0.19
	Soil Carbon	5020.08	7.05	0.4
CS	Soil N	753.78	9.13	0.38
	Grain DM yield	1970.74	29.84	0.73
	Grain N yield	36.43	38.68	0.37
	Soil Carbon	5453.96	7.62	0.37
NPK	Soil N	754.86	10.23	0.31
	Grain DM yield	935.78	29.81	0.68
	Grain N yield	8.52	19.63	0.64
	Soil Carbon	4750.03	7.03	0.69
U	Soil N	623.34	9.54	0.53
	Grain DM yield	1787.08	70.67	0.63
	Grain N yield	51.06	197.95	0.08
	Soil Carbon	6372.59	9.23	0.18
GM	Soil N	671.6	7.36	0.56

Crop	Measurement	Model fit RMSE	MAPE	IA
	Grain DM yield	2212.81	40.56	0.54
	Grain N %	0.29	15.13	0.68
Oats	Grain N yield	44.84	47.79	0.45
	Grain DM yield	2088.83	60.5	0.72
Spring barley	Grain N %	0.61	22.23	0.53
	Grain N yield	22.83	19.91	0.8
	Grain DM yield	492.75	63.32	0.43
Spring rape	Grain N %	1.93	41.46	0.22
	Grain N yield	46.56	128.28	0.3
	Grain DM yield	1397.91	35.52	0.82
Spring wheat	Grain N %	0.29	10.76	0.87
	Grain N yield	42.81	51.58	0.65
	Grain DM yield	1096.61	37.83	0.78
Winter barley	Grain N %	NA	NA	NA
	Grain N yield	NA	NA	NA
	Grain DM yield	1689.2	23.59	0.88
Winter wheat	Grain N %	0.29	16.19	0.49
	Grain N yield	43.32	34.41	0.75
	Grain DM yield	3442.68	78.72	0.5
Spring barley silage	Grain N %	0.56	26.81	0.45
	Grain N yield	44.86	59.81	0.49
	Grain DM yield	4195.84	64	0.45
	Grain N %	1.03	99.17	0.28
Ryegrass	Grain N yield	47.46	58.49	0.48

Abbreviations: DM yield =dry matter kg ha^{-1}, N%=nitrogen content in percentage, N yield=nitrogen yield in kg N ha^{-1}; CH=compost from household wastes, CHA= accelerated CH, S=sewage sludge, SA= S accelerated, HU=human urine, DL=deep litter, CMA=cattle manure accelerated, CS=cattle slurry, NPK=mineral fertilization, U=unfertilized, GM=green manure

Supplementary Table 4: Results of the statistical analysis of influences on relative dry matter yield (% in relation to year average yield) and N content of the grains (%).

	Df	Sum Sq	Mean Sq	F value	Pr(>F)
Effect on relative dry matter yield (% in relation to year average yield)					
Treatment	10	347331	34733	116.3	8.14E-133
Years of application	1	0.1	0.1	0.0004	0.983
Crop	7	2	0.3	0.0009	1.00
Block	3	1483	494	1.7	0.175
Treatment:Years of application	10	20118	2012	6.7	5.64E-10
Treatment:Crop	70	87594	1251	4.2	2.75E-22
Residuals	594	177361	299		
Effect on grain N content (%)					
Treatment	10	5.40	0.54	7.21	5.91E-10
Years of application	1	52.10	52.10	695.37	3.03E-73
Crop	4	232.77	58.19	776.64	4.18E-137
Block	3	0.19	0.06	0.82	0.48
Treatment:Years of application	10	0.69	0.07	0.92	0.51
Treatment:Crop	40	4.07	0.10	1.36	0.08
Residuals	243	18.21	0.07		

Model: relative yield (%) / N grain content (%)~ treatment + application duration (years) + cropped culture + application duration : treatment+ cropped culture : treatment + block

Supplementary Table 5: Results of the ANOVA on influences on agronomic efficiency. Bold font represents significance (α=0.05).

Results of the ANOVA on effects on agronomic efficiency

	Df	Sum Sq	Mean Sq	F value	Pr(>F)
Years of application	**1**	**6344.9**	**6344.9**	**15.0**	**1.30E-04**
Applied N amount	**1**	**194569.6**	**194569.6**	**459.9**	**2.54E-64**
Treatment	**8**	**143710.2**	**17963.8**	**42.5**	**2.66E-46**
Crop	**5**	**6353.3**	**1270.7**	**3.0**	**1.15E-02**
Block	3	825.1	275.0	0.7	5.83E-01
Treatment : Years of application	**8**	**11538.6**	**1442.3**	**3.4**	**8.85E-04**
Treatment : Crop	**40**	**25154.5**	**628.9**	**1.5**	**3.44E-02**
Treatment : Applied N amount	**8**	**45964.1**	**5745.5**	**13.6**	**5.29E-17**
Residuals	327	138342.0	423.1		

Model: Agronomic efficiency (%) ~ treatment + years of application + amount of applied N + cropped culture + years of application : treatment + amount of applied N : treatment + cropped culture : treatment + block

Supplementary Table 6: : Results of the ANOVA on influences on the agronomic efficiency for each fertilization scheme separately in a submodel. Bold font represents significance (α=0.05).

Treat.	factor	Df	Sum Sq	Mean Sq	F value	Pr(>F)	
CH	Years of application	1	147	147	2.1	1.53E-01	n.s.
CH	Applied N amount	1	1361	1361	19.8	7.96E-05	***
CH	Crop	5	1010	202	2.9	2.52E-02	*
CH	Years of application:Applied N amount	1	0	0	0.0	9.58E-01	n.s.
CH	Residuals	36	2475	69			
CHA	Years of application	1	203	203	18.8	1.10E-04	***
CHA	Applied N amount	1	5	5	0.4	5.08E-01	n.s.
CHA	Crop	5	635	127	11.8	8.49E-07	***
CHA	Years of application:Applied N amount	1	141	141	13.1	8.89E-04	***
CHA	Residuals	36	387	11			
S	Years of application	1	3911	3911	8.0	7.60E-03	*
S	Applied N amount	1	15631	15631	32.0	2.02E-06	***
S	Crop	5	6881	1376	2.8	3.03E-02	*
S	Years of application:Applied N amount	1	230	230	0.5	4.97E-01	n.s.
S	Residuals	36	17602	489			
SA	Years of application	1	212	212	3.9	5.61E-02	.
SA	Applied N amount	1	1303	1303	24.0	2.07E-05	***
SA	Crop	5	1832	366	6.7	1.59E-04	***
SA	Years of application:Applied N amount	1	164	164	3.0	9.11E-02	.
SA	Residuals	36	1957	54			
HU	Years of application	1	5260	5260	23.9	2.14E-05	***
HU	Applied N amount	1	14696	14696	66.6	1.04E-09	***
HU	Crop	5	4074	815	3.7	8.42E-03	*
HU	Years of application:Applied N amount	1	2315	2315	10.5	2.57E-03	*
HU	Residuals	36	7938	220			
DL	Years of application	1	19	19	0.2	6.83E-01	n.s.
DL	Applied N amount	1	1369	1369	12.5	1.15E-03	*
DL	Crop	5	1482	296	2.7	3.56E-02	*
DL	Years of application:Applied N amount	1	41	41	0.4	5.43E-01	n.s.
DL	Residuals	36	3946	110			
CMA	Years of application	1	262	262	1.9	1.75E-01	n.s.
CMA	Applied N amount	1	1214	1214	8.9	5.34E-03	*
CMA	Crop	5	1400	280	2.1	9.69E-02	.
CMA	Years of application:Applied N amount	1	547	547	4.0	5.35E-02	.
CMA	Residuals	33	4505	137			
CS	Years of application	1	9520	9520	8.3	6.76E-03	*
CS	Applied N amount	1	22788	22788	19.8	8.03E-05	***
CS	Crop	5	9109	1822	1.6	1.90E-01	n.s.
CS	Years of application:Applied N amount	1	17499	17499	15.2	4.07E-04	***
CS	Residuals	36	41486	1152			

Model: agronomic efficiency ~ Years of application*Applied N amounts +Crop for each fertilizer treatment separately

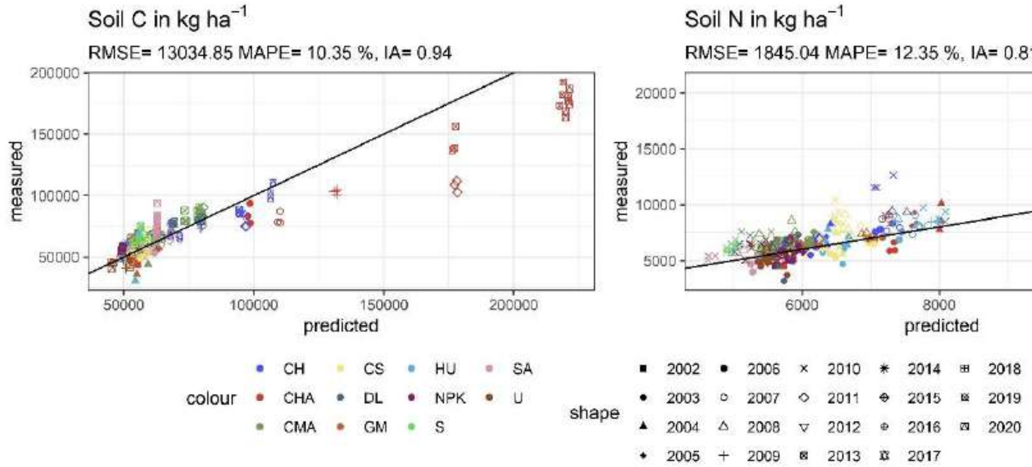
Supplementary Table 7: Nitrogen balances for all fertilization schemes divided by input, output, nitrogen losses and change in soil N (positive values mean an increase).Given are the proportion of the total input and means in kg N ha^{-1} year^{-1}.

Treat.	Type	Sum	Sub-type	% Input	Mean
CH	Input	409	Deposition	3.3	14
			Seed	1.0	4
			Fixated	3.1	13
			Fertilizer	92.6	378
	Loss	185	Leaching	25.0	102
			Surface loss	8.6	35
			N2O	3.5	14
			N2 Denitri.	8.1	33
	Out	123	Harvest	30.0	123
	Soil N	101	organic N	22.0	90
			mineral N	2.8	11
CHA	Input	1158	Deposition	1.2	14
			Seed	0.3	4
			Fixated	0.7	9
			Fertilizer	97.7	1132
	Loss	636	Leaching	39.0	452
			Surface loss	9.2	106
			N2O	2.3	26
			N2 Denitri.	4.5	52
	Out	149	Harvest	12.9	149
	Soil N	375	organic N	28.0	324
			mineral N	4.4	51
S	Input	215	Deposition	6.4	14
			Seed	1.9	4
			Fixated	4.8	10
			Fertilizer	87.0	187
	Loss	100	Leaching	25.5	55
			Surface loss	2.2	5
			N2O	5.7	12
			N2 Denitri.	13.2	28
	Out	106	Harvest	49.1	106
	Soil N	6	organic N	-0.4	-1
			mineral N	3.0	7
SA	Input	507	Deposition	2.7	14
			Seed	0.8	4
			Fixated	1.9	10
			Fertilizer	94.6	479
	Loss	281	Leaching	41.2	209
			Surface loss	2.4	12
			N2O	4.0	20
			N2 Denitri.	8.0	40
	Out	138	Harvest	27.2	138
	Soil N	84	organic N	10.2	52
			mineral N	6.4	32

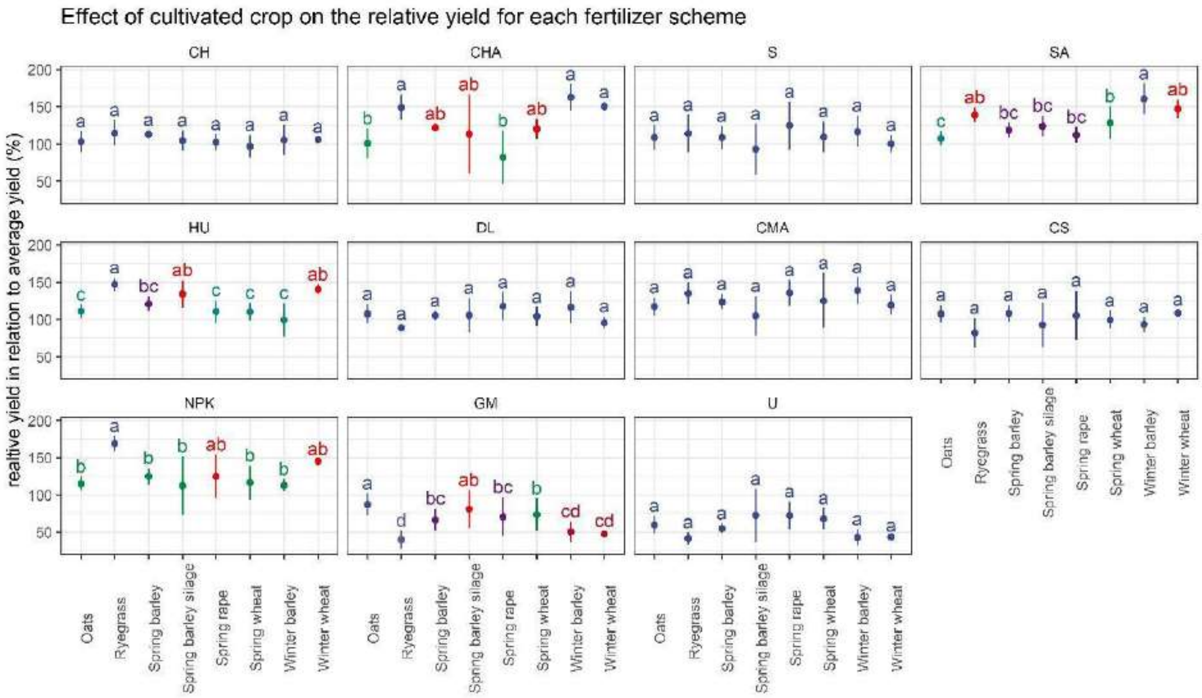
Treat.	Type	Sum	Sub-type	% Input	Mean
HU	Input	189	Deposition	7.2	14
			Seed	2.1	4
			Fixated	7.2	14
			Fertilizer	83.5	158
	Loss	99	Leaching	22.2	42
			Surface loss	11.9	23
			N2O	5.7	11
			N2 Denitri.	12.5	24
	Out	106	Harvest	55.8	106
	Soil N	-15	organic N	-14.1	-27
			mineral N	6.4	12
DL	Input	368	Deposition	3.7	14
			Seed	1.1	4
			Fixated	3.6	13
			Fertilizer	91.6	337
	Loss	139	Leaching	22.8	84
			Surface loss	0.3	1
			N2O	4.2	16
			N2 Denitri.	10.3	38
	Out	115	Harvest	31.3	115
	Soil N	114	organic N	27.7	102
			mineral N	3.3	12
CMA	Input	363	Deposition	3.8	14
			Seed	1.1	4
			Fixated	4.7	17
			Fertilizer	90.4	328
	Loss	125	Leaching	14.8	54
			Surface loss	3.3	12
			N2O	4.4	16
			N2 Denitri.	12.0	44
	Out	101	Harvest	28.0	101
	Soil N	139	organic N	35.7	129
			mineral N	2.8	10
CS	Input	132	Deposition	10.4	14
			Seed	3.0	4
			Fixated	18.0	24
			Fertilizer	68.6	90
	Loss	62	Leaching	18.1	24
			Surface loss	4.6	6
			N2O	6.8	9
			N2 Denitri.	17.1	23
	Out	78	Harvest	59.5	78
	Soil N	-5	organic N	-5.3	-7
			mineral N	1.1	1

Treat.	Type	Sum	Sub-type	% Input	Mean
NPK	Input	143	Deposition	9.6	14
			Seed	2.8	4
			Fixated	11.9	17
			Fertilizer	75.8	108
	Loss	69	Leaching	28.2	40
			Surface loss	0.0	0
			N2O	5.9	8
			N2 Denitri.	13.8	20
	Out	98	Harvest	68.4	98
	Soil N	-22	organic N	-19.9	-29
			mineral N	4.3	6
U	Input	20	Deposition	69.8	14
			Seed	19.5	4
			Fixated	10.7	2
			Fertilizer	0.0	0
	Loss	31	Leaching	70.7	14
			Surface loss	0.2	0
			N2O	25.0	5
			N2 Denitri.	61.4	12
	Out	47	Harvest	237.6	47
	Soil N	-54	organic N	-277.6	-54
			mineral N	2.5	0
GM	Input	98	Deposition	13.9	14
			Seed	4.4	4
			Fixated	81.7	80
			Fertilizer	0.0	0
	Loss	43	Leaching	20.9	21
			Surface loss	0.0	0
			N2O	7.6	7
			N2 Denitri.	15.1	15
	Out	61	Harvest	62.0	61
	Soil N	-7	organic N	-8.8	-9
			mineral N	2.0	2

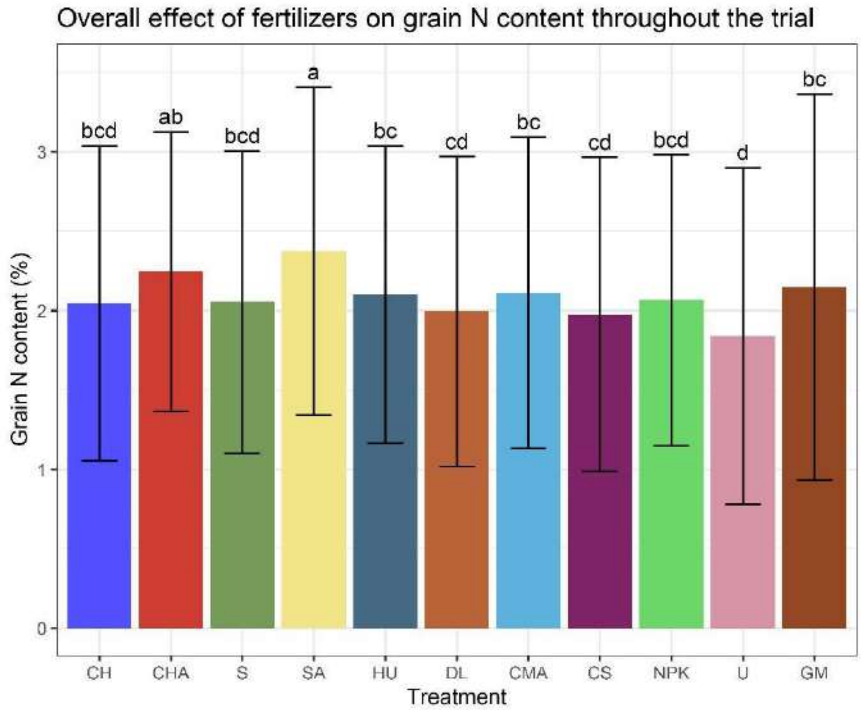
8.2 Supplementary Figures



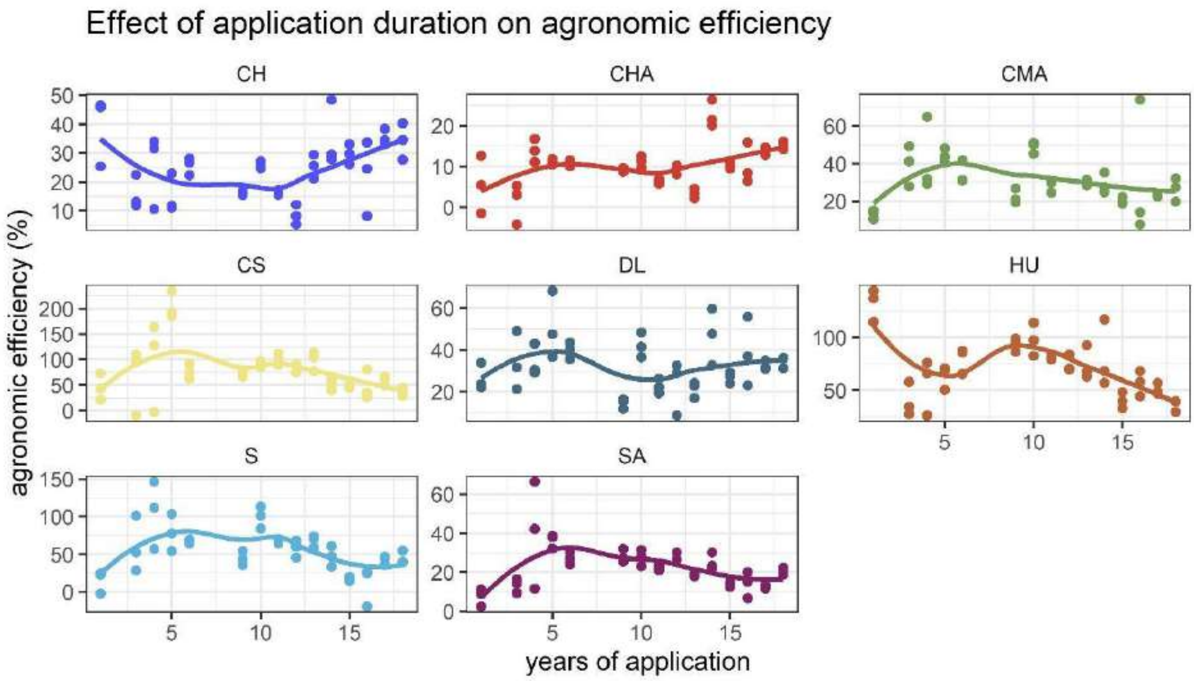
Supplementary Figure 1: Fit of the predicted model values (x-axis) to the measured data (y-axis) for soil carbon (C; in kg ha^{-1}) and soil total nitrogen (N; in kg ha^{-1}). Estimates for the overall fit are given in the form of RMSE, MAPE, and index of agreement (IA). Dots represent a value pair of measured and predicted data, shape the year of measurement and color the fertilization treatment.



Supplementary Figure 2: Effect of the cultivated crop on the relative DM yield in relation to the year's average yield (in %) for each fertilizer scheme separately. Letters indicate significant differences, dots represent the mean and bars the standard deviation.



Supplementary Figure 3: Influence of the fertilization treatment on the grain N content (in %) over all years and crops. Letters indicate significant differences, bars the means and error bars the standard deviation.



Supplementary Figure 4: Agronomic efficiency throughout the duration of the experiment. Dots represent the agronomic efficiency for each repetition of the treatment; lines represent the trend as predicted by the LOESS method.