

## Article

# Using Food Waste in Organic Fertilizer: Modelling Biogenic Carbon Sequestration with Associated Nutrient and Micropollutant Loads

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**Abstract:** What are the effects, measured as flows of biogenic carbon, plant nutrients, and pollutants, of moving organic waste up the waste hierarchy? We present a case study of Denmark, where most of the organic fraction of household waste (OFHW) is incinerated, with ongoing efforts to increase bio-waste recycling. In this study, one-third of the OFHW produced in North Zealand, Denmark, is diverted away from incineration, according to the Danish Waste Resource Plan 2013–2018. Co-digestion of OFHW, and digestate application on agricultural soil, utilizes biogenic carbon, first for energy conversion, and the remainder for long-term soil sequestration, with additional benefits for plant nutrient composition by increasing the N:P ratio in the digestate. We show a dynamic model of the biogenic carbon flows in a mix of OFHW co-digested with livestock manure and sewage sludge, addressing the contribution of OFHW to long-term carbon sequestration compared to other agricultural residues and bio-wastes over a time span of 100 years. In addition, we trace the associated annual nutrient and cadmium loads to the topsoil. At constant annual input rates and management practices, a diversion of 33% of OFHW would result in an increased organic carbon build-up of approximately 4% over the current amounts applied. The addition of OFHW, moreover, beneficially adjusts the N:P ratio of the digestate mix upwards, albeit without reaching an ideally high ratio by that measure alone. Cd loads from OFHW remain well below regulatory limits.

**Keywords:** dynamic material flow analysis; bio-waste; food waste; biogas; carbon sequestration; nutrient cycling

## 1. Introduction

The challenge of a circular economy is the effective and sustained closing of multiple material cycles in a production system. The local recycling and valorization of the organic fraction of household waste (OFHW) as a fertilizer product holds a potential to contribute to climate change mitigation by long-term soil carbon sequestration and energy recovery, as well as to increased nutrient cycling in the agri-food system [1]. This issue has generally been tackled from a waste management perspective, as in [2–4], or been focused on specific biotechnological or agricultural aspects such as anaerobic digestion or soil processes [5–9]. A synthesis, in a simple yet practicable model to trace key mass flows associated with the production of organic fertilizer from OFHW—(biogenic) carbon, plant nutrients, pollutants—and to give an indication of the viability and potential benefits of the end product, remains an open and pertinent issue in the opinion of the authors.

This article uses a region of Denmark for an illustrative case study, but aims to present more generally applicable results. For many years, a usual means of waste disposal in Denmark has been incineration, which was ongoing at a steady rate of approx. 28–30% of waste incinerated in the period

2013–2017 and only recovers the energy content in the waste thus disposed of [10]. Due to its high water content and low heating value, however, OFHW is not particularly valuable for incineration with energy recovery in a waste-to-energy (WtE) plant [11,12]. While a number of technologies exist to recover phosphorus, as an essential plant nutrient, from ash, incineration is left with the disadvantage of emitting carbon stored in the waste to the atmosphere as CO<sub>2</sub>. Furthermore, the recovery of phosphorus from incineration residues is energy intensive, resulting in additional CO<sub>2</sub> emissions, and is costly [13], so that nutrients may end up being lost in incineration ash.

The phosphorus content of OFHW is relatively low, while the nitrogen content is high. This makes OFHW a suitable co-substrate for anaerobic digestion, especially in manure-based biogas plants, with a potential for a significant increase in bioenergy production as well as avoiding emissions from CO<sub>2</sub>-intensive nitrogen fertilizer production [14]. Anaerobic digestion allows for moving OFHW up the waste hierarchy from incineration and energy recovery [2]: The easily degradable fraction of biogenic carbon is utilized for energy production, and nutrients are returned to soil as well as the recalcitrant, slowly degradable biogenic carbon fraction, thus constituting a degree of material recycling. Energy recovery from and soil sequestration of biogenic carbon have been shown to be the main drivers of the overall environmental benefits of anaerobically digesting OFHW [15], particularly in light of forecasted negative soil C fluxes over the next 100 years [16]. The additional positive effects in support of soil biological activity and nutrient cycling have been highlighted, for example, by [6].

We present a case study of the local production of organic-waste-derived fertilizer obtained by diverting OFHW in the present catchment area of a WtE plant in North Zealand (Denmark) to co-digestion at eight sludge-based and five manure-based biogas plants [17]. At present, the application of OFHW to agricultural soils plays a negligible role in Denmark [14,18]. In the case study area, 132 kilotons (dry matter (DM)) of OFHW are currently co-incinerated with municipal waste, and 24.7 kilotons (DM) of livestock manure, as well as 35.2 kilotons (DM) anaerobically digested sludge, are spread on agricultural land. We examine the long-term consequences of an alternative scenario in which 33% of organic household waste are recycled to produce biogas and fertilizer, as was laid out in the Danish Waste Resource Plan 2013–2018 [19]. The livestock manure produced is anaerobically digested in this scenario, so that the OFHW diverted to anaerobic digesters is co-digested with either sewage sludge or manure.

To quantify the contribution of OFHW to soil organic carbon in this scenario, we examine a dynamic model of the biogenic carbon stocks and flows in the system over a 100-year time horizon using the STELLA modelling software (version 1.9; <https://iseesystems.com>). The model quantifies only biogenic carbon stocks and flows and as such excludes indirect emissions, e.g., from energy consumption or transport. We model carbon flow and stock dynamics in the waste management system, comprising organic household waste generation, production, consumption, storage processes, anaerobic digestion, and spreading on soil. Soil processes are modelled using an assumption of first order degradation kinetics, based on literature data for wastes and manures representative of the case study area [7,8,19,20].

The dynamic biogenic carbon model is accompanied by a quantification of the annual nitrogen and phosphorus loads via the organic fertilizers applied to soil, in order to determine to which extent the digestate-derived fertilizers can meet plant demand for N and P at an ideal N:P ratio [5], as well as regulatory demands related to the nutrient content of organic fertilizers [21]. Because only organic materials meeting a minimum nutrient content are permitted for use as organic fertilizers, and a sufficiently high N:P ratio is necessary for effective fertilizing use, a suitable mix of input materials is needed to obtain a suitable product. The cadmium load to soil was quantified, in addition to carbon and nutrients, due to its diet-related health impacts. Cd is present in relatively high concentrations in mineral P fertilizer, but also in sewage sludge, manure, and food waste, albeit in lower concentrations [13,14], and is of regulatory relevance [22].

The results quantify the extent to which atmospheric carbon stored in OFHW is conserved after anaerobic digestion, and the carbon in the resulting organic-waste-derived fertilizer is sequestered

in agricultural soil, while monitoring the associated nutrient and cadmium inputs and the nutrient compositions obtained to ensure a suitable organic fertilizer product. The dynamic model itself can furthermore inform Life Cycle Assessments with regard to the net process carbon footprint of local bio-waste management systems when calculating the potential for climate change mitigation of organic-waste-derived fertilizers.

## 2. Materials and Methods

### 2.1. System Description

The case study examines an area in North Zealand (Nordsjælland), Denmark, the part of the Danish island of Zealand north of Copenhagen and delimited by the Isefjord to the west, Kattegat to the north, and Øresund to the east. The area comprises the catchment area of the Vestforbrænding waste-to-energy plant west of Copenhagen and 68,608 ha. of cropland [8]. In this area, 70 kilotons of OFHW, 20.6 kilotons of manure, and 41.5 kilotons of sewage sludge (by dry matter (DM)) are produced annually.

In the reference system, OFHW is incinerated, raw manure is spread on farmland, and sewage sludge is anaerobically digested, with the resulting digestate likewise applied to farmland. In the alternative scenario studied in this article, manure is also anaerobically digested and one-third (33%) of OFHW diverted away from incineration to be co-digested with either manure or sewage sludge, after which the digestate is again applied on farmland. This scenario would, moreover, necessitate the establishment of three new manure-based biogas plants to supplement the current eight sludge-based biogas plant in the case study area [17].

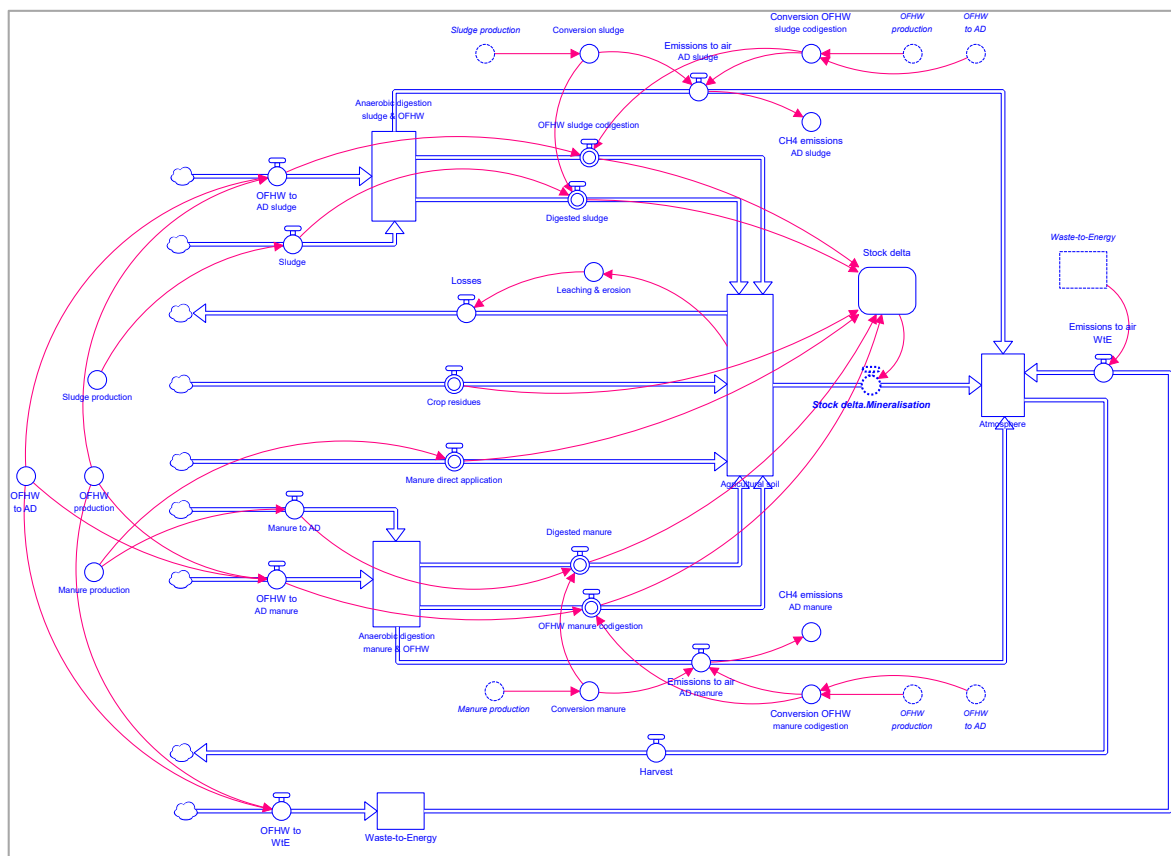
#### System Boundary

The geographical system boundary comprises 32 municipalities in North Zealand comprising the catchment area of the Vestforbrænding waste incineration plant in Greater Copenhagen. In process terms, the system boundary is delineated by the management (and re-use, where applicable) of the wastes and manure generated within the geographical system boundary, so that the generation phase of these materials is not included in the system [17].

### 2.2. Model Description

The system was modelled in the STELLA Professional software (version 1.9; iseesystems.com). Figure 1 shows a diagram of the model generated in STELLA, with the different types of the model's elements represented as follows:

- Processes indicate functional relationships [15] between stocks and represent substance flows inside the model. They are represented as arrows with a valve;
- Parameters, or converters in the terminology of the STELLA software, define processes (which in turn determine flows) or other converters;
- Stocks are represented as rectangles and are time-dependent. The time scale of the model was chosen as 1 year. None of the stocks contains C at year 0;
- A red arrow, or connector, represents a direct influence of one model component on another.



**Figure 1.** Diagram of the biogenic carbon flows as modelled in the STELLA software. OFHW: organic fraction of household waste; WtE: waste-to-energy (incineration); AD: anaerobic digestion.

The model only describes biogenic carbon (C) flows in the system, i.e., the carbon imported to the system via sewage sludge, manure, and the organic fraction of household waste (OFHW). These materials are eventually applied to soil, where the development of soil C stocks and mineralization of the added C amounts are modelled over a 100-year timeframe. Processes, stocks, and flows are modelled in kg C and calculated in intervals of 1 year. Annual N, P, and Cd loads to soil are included in the model, while fate modelling in top soil was excluded for these elements.

### 2.2.1. Processes and Parameters

The model comprises 17 processes in total, which are further defined by 16 parameters. These are described in sub-sections below, grouped according to their functional roles and relationships, as illustrated by the diagram in Figure 1, with their definitions given in Tables A1 and A2 in Appendix A. Stocks are described in Section 2.2.2.

#### OFHW, Manure, and Sludge Production

In the reference scenario, 100%, or 70 kilotons DM, corresponding to 35.2 kilotons C, calculated from [16] and [17] of OFHW, are routed to WtE annually. Thirty-three percent of OFHW (23 kilotons DM, or 11.7 kilotons C) are co-digested annually with either sewage sludge or manure in the alternative scenario. OFHW is mainly made up of kitchen or food waste [4,23]. The manure to be managed in the system amounts to 24.7 kilotons DM, or 10.5 kilotons C [11] annually, of which 20.6 kilotons DM (8.7 kilotons C) are produced within sufficient proximity of anaerobic digestion (AD) plants to be co-digested with OFHW. Sludge production is 41.5 kilotons DM (20 kilotons C) per year, to be co-digested with OFHW as well. These values are set as constants throughout the timespan examined,

assuming no change in, e.g., demographics and agricultural practices, which would be beyond the scope of this study.

#### Application on Fields, Crop Uptake and Soil C Mineralization, Harvest, Crop Residues

Because a part of the degradable organic matter in the respective feedstocks is already converted to methane during anaerobic digestion (for the calculation, see Appendix A, Table A2), the ratio of degradable to recalcitrant C is lower in digested than in undigested manure, sludge, or OFHW. This can result in a larger fraction of the applied C to be stabilized in soil, with approximately twice as much C stabilized long-term for digested compared to undigested material, in the case of manure [7].

Parameters such as temperature, precipitation, and the C:N ratio impact the degradability of an organic material applied to land, with anaerobic digestion decreasing the C:N ratio and increasing degradability [24,25]. Due to the dependence of C:N ratio on other chemical feedstock characteristics [24], and factors such as seasonal variation, we used literature values for the purpose of this study. The values used describe C mineralization dynamics in agricultural soils in temperate, north-western European climates, representative of the Danish situation and the predominantly loamy soils [26] on Zealand. Table 1 gives an overview of the sources and values used in this study and the respective amounts of C applied in the model. For all materials applied in the case study, turnover rates,  $k$ , for fast- and slow-turnover C pools, applicable for conditions such as those examined, were available. The humification coefficient,  $h$ , indicates the estimated fraction of recalcitrant—virtually non-degradable—organic matter remaining in soil in the long term, i.e., several decades or longer [7–9], and might even be viewed as a rough proxy for C soil sequestration.

**Table 1.** Amounts applied (in tons C) and turnover rates,  $k$  (fast- and slow-turnover C pools  $k_1$  and  $k_2$ ), in the scenario with 33% diversion of OFHW to anaerobic digestion and land application. The humification coefficient,  $h$  (in % of total C applied), denotes the recalcitrant fraction of the applied C subject to humification and remaining in the soil.

	C Applied [kg C]	$k_1$ [yr <sup>-1</sup> ]	$t_{1/2, 1}$ [yr]	$k_2$ [yr <sup>-1</sup> ]	$t_{1/2, 2}$ [yr]	$h$ [%]	Reference
Digested OFHW	2,075,500	32.85 42% <sup>a</sup>	0.021	10.95 10% <sup>a</sup>	0.063	48%	[27]
Cattle manure	1,358,280	83.95 9% <sup>a</sup>	0.008	2.37 52% <sup>a</sup>	0.29	39%	[8]
Digested cattle manure	3,792,250	229.95 3% <sup>a</sup>	0.003	2.52 74% <sup>a</sup>	0.28	23%	[8]
Pig slurry	405,720	48.29 29% <sup>a</sup>	0.014	3.18 31% <sup>a</sup>	0.22	40%	[8]
Digested pig slurry	1,132,750	20.88 38% <sup>a</sup>	0.033	0.33 7% <sup>a</sup>	2.11	55%	[8]
Digested sludge <sup>b</sup>	8,005,000	32.85 42% <sup>a</sup>	0.021	10.95 10% <sup>a</sup>	0.063	48%	[27]
Crop residues	26,550,000	72.09 5% <sup>a</sup>	0.010	0.99 28% <sup>a</sup>	0.70	67%	[8]

<sup>a</sup> Fractional amount of C in the rapid ( $k_1$ ) and slow ( $k_2$ ) turnover pools. <sup>b</sup> Values for digested OFHW used as an approximation.

The cumulative amount of the mineralization of C added to soils in the form of digested OFHW, manure, digested manure, digested sewage sludge, and crop residues left on or returned to fields is assumed to follow first-order kinetics (see [11,18]), as in Equation (1):

$$C_m = \sum_{t=0}^{t=n} C_{input} [1 - \exp(-k * t)] \quad (1)$$

where  $C_m$  is the amount of C mineralized from time 0 to  $t$ ,  $C_{input}$  is the size of the C pool from the input material at time 0, and  $k$  is the turnover rate of the C pool, with  $C_m$  for each input year and the amounts summed over the entire time frame examined.

If values for a two-pool model were available, the equation was adapted after [6], with the pool of added degradable C divided into a fast-turnover and a slow-turnover pool (Equation (2)):

$$C_m = \sum_{t=0}^{t=n} C_{input, k1} [1 - \exp(-k_1 * t)] + \sum_{t=0}^{t=n} C_{input, k2} [1 - \exp(-k_2 * t)] \quad (2)$$

where  $k_1$  and  $k_2$  denote the fast and slow turnover rates and  $C_{input, k1}$  and  $C_{input, k2}$  are the sizes of the corresponding C pools.

Crop uptake of CO<sub>2</sub>-C is determined by the parameters quantifying productivity, the amount of edible harvest of a given year, and crop residues left on or returned to fields, with crop residues forming the bulk of the annual biogenic C addition to soil.

#### Leaching and Erosion

Losses of C from added organic materials through leaching and erosion were estimated based on [28], representative of the Danish situation, yet at a low value of 1% annually of organic C added. Because these losses correlate directly with the amount of humified, stable organic C in the soil [24], leaching and erosion increases with increasing soil C sequestration.

#### Biogas Plants and Waste-to-Energy Plants

In the model, the losses of biogenic C to air from AD plants are determined by the parameters “Conversion sludge”, “Conversion OFHW sludge codigestion”, “Conversion manure”, and “Conversion OFHW manure codigestion” for sludge- and manure-based biogas plants, respectively, whereas in the case of WtE these losses are a fixed fraction of the (static) OFHW input. These “conversion” parameters for the biogas plants define the conversion of the biogenic C in the various feedstocks to biogas (Table A2 in Appendix A), based on the feedstocks’ CH<sub>4</sub> yields; these yields, usually reported in m<sup>3</sup>/t DM input, were further converted to kg C to fit with the model’s mass balance.

Losses of C through air emissions from the WtE facility and the manure-based and sludge-based biogas plants are based on the values reported by [17]. The carbon loss to air from biogas plants (Appendix A, Table A2) was calculated based on Equation (3):

$$C_{air, AD} = EF_{CH_4-C} * V_{CH_4} * \frac{M_C}{M_{CH_4}} + \frac{0.65}{0.35} * V_{biogas} * \frac{M_C}{M_{CO_2}} \quad (3)$$

where the emission factor,  $EF$ , for methane losses from the biogas plant is 1.3% for sludge-based biogas plants and 4.2% for manure-based biogas plants [18].  $EF$  values were corrected for the ratio of CH<sub>4</sub> to CO<sub>2</sub> in biogas, here assumed at 0.35/0.65.  $M_C/M_{CH_4}$  and  $M_C/M_{CO_2}$ , denote the C content in CH<sub>4</sub> and CO<sub>2</sub>, respectively.

#### 2.2.2. Stocks

Stocks are defined by the processes and parameters that determine a stock’s in- and outflows. As such, they consist of simple additions and subtractions, while the “mineralization” process (Equations (1) and (2)), for example, determines soil accumulation of biogenic C. The Anaerobic Digestion and Waste-to-Energy stocks do not accumulate C, P, or Cd over one year; their purpose is in linking processes, routing flows, and delivering output flows to be acted upon further by the processes for final disposal or treatment. Table A3 in Appendix A gives the definition of the stocks used in the model.

#### 2.3. Nitrogen, Phosphorus, and Cadmium Loads

Apart from carbon sequestration, the content of plant nutrients, as well as of heavy metals, is crucial in ensuring the viability of using organic-waste-derived fertilizers on agricultural land.



Therefore, the annual loads of N, P, and Cd to soil resulting from the application of digested sludge, raw manure, digested manure, and digested OFHW in the case study were quantified in addition to the dynamic model for biogenic C. Table 2 summarizes the nitrogen, phosphorus, and cadmium contents and annual amounts applied via the fertilizer materials. Table 3 gives an overview of applicable regulatory limits for heavy metal contents in organic fertilizers.

**Table 2.** Nitrogen (total), phosphorus (total), and cadmium contents, on a dry matter (DM) basis, in the materials applied to land, as well as in the undigested feedstocks not applied to land in the case study.

	N	P	N:P	Cd	Reference
	(kg/kg DM)	(kg/kg DM)		(mg/kg DM)	
Sludge *	0.012	0.008	1.5	0.87	[14,17]
Digested sludge	0.03	0.023	1.5	0.88	[14,17]
Manure	0.05	0.013	3.9	0.29	[14,17]
Digested manure	0.07	0.02	3.5	0.43	[14,17,28]
OFHW *	0.024	0.0028	8.4	0.02	[17,23]
Digested OFHW	0.032	0.0039	8.3	0.037	[17,23,29]

\* Undigested feedstocks not applied on land in the case study.

**Table 3.** Limits for heavy-metal content applicable to the relevant organic fertilizer materials in the Danish and EU context.

	Cd	Hg	Pb	Ni	Cr	Zn	Cu	Reference
Sludge, conventional agriculture [mg/kg DM]	0.8	0.8	120	30	100	4000	1000	[22]
Sludge, conventional agriculture [mg/kg P]	100	200	10,000	2500	-	-	-	[22]
Composted or anaerobically digested organic household waste, organic agriculture [mg/kg DM]	0.7	0.4	45	25	70	200	70	[30]
Organic fertilizer containing carbon and nutrients [mg/kg DM]	1.5	1	120	50	2 (Cr VI)	800	300	[21]

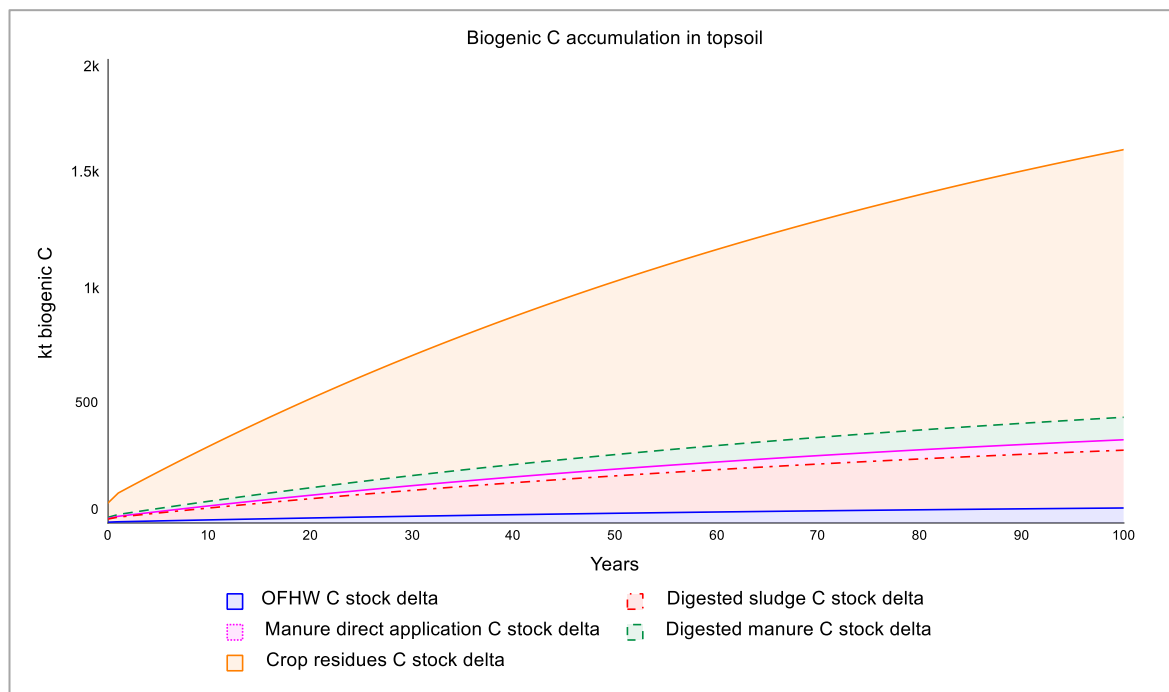
### 3. Results and Discussion

Figure 2 shows the net accumulation of biogenic C in the agricultural soils under the alternative management scenario outlined in Section 2.1 for 100 annually repeated management cycles.

Following the scenario with 33% of OFHW diverted from incineration, carbon from digested OFHW approximately makes up an additional 4% over the sum of biogenic C from sludge, manure, and crop residues sequestered over a 100-year time frame. An increase of this amount, to a theoretical upper limit of 100% of OFHW in the case study area diverted from incineration and used as fertilizer, this increase in biogenic C sequestration would amount to 12% over the amount already sequestered by the other input materials.

Thomsen et al. [7] indicate that, over a timespan of one year or longer, the decomposition and mineralization of biomass added to soil differs little between fresh and digested plant biomass and manure. Due to the predominance of crop production in the case study area, and a concomitantly low livestock production [25], crop residues (mainly straw) form the most important fraction of the biogenic carbon accumulation in soil. While the absolute amounts of digested OFHW are small compared to the total amount of biogenic C stored in soil through the other materials already applied to land (Figure 2), it is worth noting that the recalcitrant fraction in digested OFHW is comparable to that of the other materials in this study (see Table 1). A similar fraction, per amount of OFHW applied, remains in the

soil in the long term compared to the other materials, with the large amounts (in terms of biogenic C) of crop residues remaining on or returned to fields each year, and their comparatively low degradability, seemingly reducing the relative impact of OFHW application.



**Figure 2.** Net accumulation of biogenic carbon in soil in the study area over 100 years (in kilotons biogenic C), under constant annual application rates (see Table 1) and management cycles. The figure is shown as a stacked chart showing the contribution of each input to the total soil biogenic carbon sequestered in the system. The contribution of a hypothetical 33% of OFHW anaerobically digested and applied to land, in the scenario studied, is shown in blue (solid line).

The recalcitrant fractions (between 23% and 67%) of the materials applied remain in soil and decompose over decades or centuries [7], while the half-life times of the degradable fractions are considerably shorter than one year.

Methane emissions from biogas plants are a significant tradeoff of diverting OFHW away from incineration and to anaerobic digestion and subsequent fertilizer use, as the global warming potential of these methane emissions offset a part of the emission savings through soil C sequestration. Tracing, however, the amounts of biogenic C in OFHW that end up as greenhouse gas (GHG) emissions, diverting OFHW away from incineration in our scenario, still results in a net GHG emission reduction: A single one-year management cycle with diversion of 33% of OFHW from incineration results in a net saving of about 29% of greenhouse gas emissions (originating from the biogenic C in OFHW only) in relation to incineration only (Table 4), or an emission of 90.74 kt CO<sub>2</sub>-eq. (33% of OFHW applied to land as fertilizer) as compared to 128.4 kt CO<sub>2</sub>-eq. (all OFHW incinerated). Raising the amount of OFHW digested, instead of incinerated, to 100% would reduce the biogenic C from OFHW emitted to the atmosphere to 14.43 kt CO<sub>2</sub>-eq., a reduction of 89% compared to the opposite option (incineration only).



**Table 4.** Potential net savings of greenhouse gas emissions (originating from the biogenic C content in OFHW only), single one-year management cycle. Emissions count CO<sub>2</sub> emissions from incineration, biogas combustion, and fugitive losses of CH<sub>4</sub>, as well as emissions from soil over 100 years.

OFHW Diverted from Incineration	kt CO <sub>2</sub> -eq. Emitted from OFHW[kt CO <sub>2</sub> -eq.]	CO <sub>2</sub> -eq. Net Emission Saving[%]
0%	128.34	0%
33% (23.4 kt DM)	90.74	29%
100% (70 kt DM)	14.43	89%

#### *Nitrogen, Phosphorus, and Cadmium Loads*

The phosphorus demand of the crops produced in the case study area is approximately 1400 t, or 20 kg P/ha. The materials applied, without the addition of OFHW, meet approximately 42% of the annual P plant demand of 20.2 kg P/ha in the area, which would show a minor increase (to 45% of P demand) with 100% of OFHW used on land.

The nitrogen and phosphorus demand of the crops in the case study area results in an ideal N:P ratio of at least 6.5 [14] to avoid the legal limit for N input (170 kg N/ha) being reached before the limit on P input (30 kg P/ha; [31]), necessitating the purchase of additional N fertilizer. The addition of OFHW, with an N:P ratio of 8.3 (Table 2), as an organic fertilizer could be expected to have the potential of adjusting the N:P ratio upwards due to its comparatively low P content. The N:P ratio of the materials applied, however, cannot be greatly influenced solely by increasing the amount of OFHW applied, with the N:P ratio ranging from 2 (0% of OFHW used as fertilizer) to 2.9 (100% of OFHW as fertilizer). An additional option for obtaining an N:P ratio favorable for crops can be the gradual phasing out of the use of digested sludge on land. This is increasingly the case in several European countries due to concerns over heavy metals and organic pollutants in sludge, combined with the possibility of recovering nutrients from ash [32,33]—which is, however, a costly process in energetic and monetary terms [13,34]. In such a case, increasing the amount of OFHW to be digested to an upper limit of 100%, while removing sludge from digestion, can yield a fertilizer product with an N:P ratio as high as 6.03—while, however, decreasing soil C sequestration by approximately 15%.

The separation of the digestates into liquid and solid fractions would dramatically increase the N:P ratio of the liquid digestate fraction, with about 35% of digestate N and only 4% of digestate P in the liquid fraction [14,35]. Applying the liquid fraction in the case study area, however, would necessitate transporting the P-rich solid fraction outside the system boundaries, thus—because about 92% of carbon are in the solid digestate fraction [35]—offsetting a considerable share of the potential for soil C sequestration in the study area as well. Conversely, the transport and targeted application of separate N- and P-rich fractions where needed can well be an effective future strategy for digestate management, as suggested by [36], as well as [14].

The abovementioned regulations on nitrogen and phosphorus input, meanwhile, are unlikely to be affected by the addition of digested OFHW. Even an increase of 0% to 100% of OFHW used as fertilizer would only increase P input from 8.4 to 9.1 kg P/ha and N input from 17.5 to 22.5 kg N/ha. With P, moreover, as the price driver of mineral fertilizer, at about 1.6 EUR/kg P (as compared to about 0.3 EUR/t N; [13]), the full replacement of mineral fertilizer with OFHW-based organic fertilizer (with its high N:P ratio, i.e., low P content) is quite unrealistic, at least in the area examined in this study.

With regard to heavy metals in sludge- or organic-waste-derived fertilizers, Danish legislation sets a limit (Table 3) of 0.8 mg Cd/kg DM for sludge applied to conventional agricultural land (or 100 mg/kg P). The limit is 0.7 mg Cd/kg DM for digested organic household waste, which may be applied to organically farmed agricultural land under certain preconditions [30]. The EU Fertilizer Regulation [21] sets a limit of 0.7 mg Cd/kg DM for organic fertilizers containing both organic carbon and nutrients [21]. Because the cadmium content of digested OFHW is relatively low (see Table 2), cadmium, as a micropollutant, does not appear as an obstacle to using OFHW for fertilizer production. As [5] point out, and Table 2 shows, it is only the micropollutant contamination in sludge that can

impede its suitability for agricultural use; a phasing out of sludge application on land, as mentioned above in the context of adjusting N:P ratios, would, in this case, also considerably decrease the Cd load on agricultural soil [33].

#### 4. Conclusions

At present, the application of fertilizer derived from the organic fraction of household waste, or OFHW, still plays a minor role in the management of organic waste in Denmark. This study aimed to highlight the potentials in moving OFHW up the waste hierarchy, in terms of soil biogenic carbon sequestration and nutrient flows, by applying a large portion of the organic fraction of household waste, or OFHW, on agricultural soil after anaerobic digestion, instead of incinerating this fraction with municipal solid waste. In doing so, the easily degradable fraction of organic carbon is utilized for energy production, while the considerable recalcitrant fraction is stored in soils instead of being emitted to the atmosphere during incineration. Likewise, the nutrient content that would otherwise potentially be lost in incineration ashes is returned to the soil.

The diversion of one-third of OFHW to anaerobic digestion and land application would increase soil organic C build-up, compared to the reference system (incineration of all OFHW), by approximately 4%. Diverting 100% of OFHW to anaerobic digestion would increase the additional soil organic C build-up from OFHW to 12% over the reference system. Fugitive emissions of methane from biogas plants are a sizeable tradeoff to soil C sequestration through digestate as fertilizer; these emissions do not outweigh the net process greenhouse gas savings from soil sequestration. Compared to incineration of all OFHW, a single, one-year management cycle diverting one-third of OFHW from incineration to soil would result in a net saving of 29% of CO<sub>2</sub>-eq. emissions originating from the biogenic carbon in the organic fertilizer.

In addition to soil C sequestration, the suitability of the materials applied to soil for meeting plant nitrogen and phosphorus demand was quantified, as were the associated cadmium loads, to avoid the risk of adverse health effects. With regard to nutrients, the N:P ratio of OFHW does lift the N:P ratio of the input mix of OFHW, sludge, manure, and crop residues towards an ideally high 6.5, albeit without reaching this value; liquid/solid separation and targeted application of the liquid (high N:P) or solid (low N:P) fraction could be indicated as an additional management step. Another option to reach an ideally high N:P ratio—over 6 in this case study—could be the phasing out of using sewage sludge on land, with potential recovery of nutrients from mono-incineration ash. Both these options do, however, have their drawbacks in terms of transport (especially solid-liquid separation), monetary, or energetic (especially ash treatment) cost. Additionally, the cadmium loads associated with using digested OFHW on agricultural soil are a crucial potential hindrance. These inputs, meanwhile, were shown not to be an obstacle to adding digested OFHW to soil as a fertilizer in our case study—although a reduction in the amount of sludge applied to land would lead to reduced cadmium loads as well.

While this study presents a model using average, or typical, literature data appropriate to the area examined to present a workable model, it could be expanded on and refined in several ways in the future. The carbon/nitrogen, or C:N, ratio of materials anaerobically digested and applied to land influence their degradability, which can have an obvious effect on soil carbon sequestration. Closely linked to C:N ratios and degradability are variations in climatic and seasonal variations, with different crops' nitrogen demands dependent on growth phase, growing season, and crop type.

While the application of OFHW-based fertilizer, therefore, cannot be regarded as a cure-all, it does contribute to beneficial outcomes regarding carbon sequestration and nutrient composition, while avoiding negative impacts from cadmium contamination.

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## Appendix A

**Table A1.** Definition of the processes used in the STELLA model.

Process	Definition	Unit	Description	Reference/Remarks
Manure production	24,700,000	t DM	Amount of manure (dry mass (DM)) produced in the case study area	[8]
OFHW_production	70,000,000	t DM	Amount of manure (DM) produced in the case study area	[8]
Sludge_production	41,500,000	t DM	Amount of manure (DM) produced in the case study area	[8]
Crop_residues	26,550,000	kg C/yr	C content in crop residues returned to/left on fields	[8]
Manure_direct_application	$\text{Manure\_production} \times 0.42 \times 0.17$	kg C/yr	Fraction of manure applied directly to land	[8]
OFHW_manure_codigestion	$\text{OFHW\_to\_AD\_manure} - \text{Conversion\_OFHW\_manure\_codigestion}$	kg C/yr	Amount of digestate produced by manure-based AD	[8]
Losses	Leaching_and_erosion	kg C/yr	Organic C lost through leaching and erosion	
Mineralization	$\begin{aligned} & \text{"Co-digested\_sludge\_OFHW"} \times (1 - h_{\text{OFHW}}) \times (1 - \text{EXP}(-k_{\text{AD\_OFHW}} \times t)) + \\ & \text{"Co-digested\_manure\_OFHW"} \times (1 - h_{\text{OFHW}}) \times (1 - \text{EXP}(-k_{\text{AD\_OFHW}} \times t)) + \\ & \text{Digested\_sludge} \times (1 - h_{\text{slu\_dig}}) \times (1 - \text{EXP}(-k_{\text{AD\_sludge}} \times t)) + \text{Digested\_manure} \\ & \times 0.77 \times (1 - h_{\text{cattle\_dig}}) \times (1 - \text{EXP}(-k_{\text{AD\_manure}} \times t)) + \text{Digested\_manure} \\ & \times 0.23 \times (1 - h_{\text{pig\_dig}}) \times (1 - \text{EXP}(-k_{\text{AD\_manure}} \times t)) + \\ & \text{Manure\_direct\_application} \times 0.77 \times (1 - h_{\text{cattle}}) \times (1 - \text{EXP}(-k_{\text{manure}} \times t)) + \\ & \text{Manure\_direct\_application} \times 0.23 \times (1 - h_{\text{pig}}) \times (1 - \text{EXP}(-k_{\text{manure}} \times t)) + \text{Crop\_residues} \times \\ & (1 - h_{\text{crop}}) \times (1 - \text{EXP}(-k_{\text{crop\_residues}} \times t)) \end{aligned}$	kg C/yr	Mineralization of the degradable fraction biogenic C in 77% cattle, 23% pig manure [8]	See also Equations (1) and 2; values for $k$ , see Table 1 $h_{\text{slu\_dig}}$ : recalcitrant C fraction $h$ in sludge $h_{\text{cattle\_dig}}, h_{\text{pig\_dig}}$ : recalcitrant C fraction $h$ in digested cattle/pig manure $h_{\text{cattle}}, h_{\text{pig}}$ : recalcitrant C fraction $h$ in cattle/pig manure $h_{\text{crop}}$ : recalcitrant C fraction $h$ in crop resid. $h_{\text{OFHW}}$ : recalcitrant C fraction $h$ in OFHW
Manure_to_AD	$\text{Manure\_production} \times 0.8$	kg C/yr	80% of manure sent to biogas plants	
OFHW_to_AD_manure	$\text{OFHW\_production} \times (1/3) \times 0.22$	kg C/yr	22% of the 33% of OFHW production diverted from WtE are routed to manure-based biogas plants	[8]
Digested_manure	$\text{Manure\_to\_AD} - \text{Conversion\_manure}$	kg C/yr	Amount of digested manure produced by manure-based AD	[8]
Emissions_to_air_AD_manure	$\text{Conversion\_OFHW\_manure\_codigestion} + \text{Conversion\_manure}$	kg C/yr	Air emissions of C ( $\text{CH}_4$ losses and $\text{CO}_2$ ) of manure-based AD	[8]
OFHW_to_AD_sludge	$\text{OFHW\_production} \times (1/3) \times 0.78$	kg C/yr	78% of the 33% of OFHW production diverted from WtE are routed to sludge-based biogas plants	[8]
Sludge	Sludge_production	kg C/yr	C content in annual production of sewage sludge	[8]
Digested_sludge	$\text{Sludge} - \text{Conversion\_sludge}$	kg C/yr	Amount of digestate produced by sludge-based AD	[8]
Emissions_to_air_AD_sludge	$\text{Conversion\_sludge} + \text{Conversion\_OFHW\_sludge\_codigestion}$	kg C/yr	Air emissions of C ( $\text{CH}_4$ losses and $\text{CO}_2$ ) of sludge-based AD	[8]
OFHW_sludge_codigestion	$\text{OFHW\_to\_AD\_sludge} - \text{Conversion\_OFHW\_sludge\_codigestion}$	kg C/yr	Amount of digested sludge produced by sludge-based AD	[8]
Emissions_to_air_WtE	Waste-to-Energy	kg C/yr	Emissions to air from WtE (incineration)	[8]
Harvest	45,000,000	kg C/yr	C content in harvested crops	[8]
OFHW_to_WtE	$\text{OFHW\_production} \times (2/3)$	kg C/yr	Fraction of OFHW directed to WtE (2/3)	[8]

**Table A2.** Definitions of the parameters used in the STELLA model.

Parameter	Definition	Unit	Description	Reference/Remarks
Conversion_OFHW_sludge_codigestion	$(0.5 \times \text{OFHW\_production} \times (1/3) \times 0.78) \times (0.71 \times 12/16) + (35/65) \times (0.5 \times \text{OFHW\_production} \times (1/3) \times 0.78) \times (1.96 \times 12/44)$	kg C/yr	Biogas conversion of OFHW codigested with sludge	OFHW codigested with sludge; CH <sub>4</sub> yield: 0.5m <sup>3</sup> /t DM (calculated from [8]) CH <sub>4</sub> : 0.71 kg/m <sup>3</sup> CO <sub>2</sub> : 1.96 kg/m <sup>3</sup> Biogas: 65% CH <sub>4</sub> , 35% CO <sub>2</sub>
Conversion_OFHW_manure_codigestion	$(0.5 \times \text{OFHW\_production} \times (1/3) \times 0.22) \times (0.71 \times 12/16) + (35/65) \times (0.5 \times \text{OFHW\_production} \times (1/3) \times 0.22) \times (1.96 \times 12/44)$	kg C/yr	Biogas conversion of OFHW codigested with manure	OFHW codigested with manure; CH <sub>4</sub> yield 0.5m <sup>3</sup> /t DM (calculated from [8])
Conversion_sludge	$0.35 \times \text{Sludge\_production} \times (0.71 \times 12/16) + (35/65) \times (0.35 \times \text{Sludge\_production}) \times (1.96 \times 12/44)$	kg C/yr	Biogas conversion of anaerobically digested sludge	CH <sub>4</sub> yield 0.35m <sup>3</sup> /t DM
Conversion_manure	$((0.77 \times 0.175 \times \text{Manure\_production}) + (0.23 \times 0.205 \times \text{Manure\_production})) \times (0.71 \times 12/16) + (35/65) \times ((0.77 \times 0.175 \times \text{Manure\_production}) + (0.23 \times 0.205 \times \text{Manure\_production})) \times (1.96 \times 12/44)$	kg C/yr	Biogas conversion of anaerobically digested manure	CH <sub>4</sub> yield, cattle manure: ca. 175 m <sup>3</sup> /t DM; pig manure: ca. 205 m <sup>3</sup> /t DM [8]
CH4_emissions_AD_sludge	$\text{Emissions\_to\_air\_AD\_sludge} \times 0.013 \times 0.65 \times (16/12)$	kg CH <sub>4</sub> /yr	CH <sub>4</sub> emissions from sludge-based AD plants; 1,3% of produced methane	Emission factor fo 1.3% (vol/vol) based on [18]; assumed CH <sub>4</sub> content 65%
CH4_emissions_AD_manure	$\text{Emissions\_to\_air\_AD\_manure} \times 0.042 \times 0.65 \times 16/12$	kg CH <sub>4</sub> /yr	CH <sub>4</sub> emissions from manure-based AD plants; 4,2% of the produced methane	Emission factor of 4.2% (vol/vol) based on [18]; assumed CH <sub>4</sub> content 65%
k_AD_manure	$0.77 \times ((3/77 \times k1\_cattle\_dig) + (74/77 \times k2\_cattle\_dig)) + 0.23 \times ((38/45 \times k1\_pig\_dig) + (7/45 \times k2\_pig\_dig))$	1/yr	Degradation coefficient for digested manure	Fractions denote the relative amounts of fast- or slow-degrading C in the degradable C pool (see also Table 1): k1—fast turnover fraction, k2—slow turnover fraction). 77% cattle, 23% pig manure [8]
k_manure	$0.77 \times ((9/61 \times k1\_cattle) + (52/61 \times k2\_cattle)) + 0.23 \times ((29/60 \times k1\_pig) + (31/60 \times k2\_pig))$	1/yr	Degradation coefficient for manure applied directly on land	Fractions denote the relative amounts of fast- or slow-degrading C in the degradable C pool (see also Table 1): k1—fast turnover fraction, k2—slow turnover fraction). 77% cattle, 23% pig manure [8]
k_AD_sludge	$(42/52 \times k1\_slu\_dig) + (10/52 \times k2\_slu\_dig)$	1/yr	Degradation coefficient for digested sludge	Fractions denote the relative amounts of fast- or slow-degrading C in the degradable C pool (see also Table 1): k1—fast turnover fraction, k2—slow turnover fraction). 77% cattle, 23% pig manure [8]
k_crop_residues	$(5/33 \times k1\_crop) + (28/33 \times k2\_crop)$	1/yr	Degradation coefficient for crop residues left on or returned to fields	Fractions denote the relative amounts of fast- or slow-degrading C in the degradable C pool (see also Table 1): k1—fast turnover fraction, k2—slow turnover fraction). 77% cattle, 23% pig manure [8]
k_AD_OFHW	$(42/52 \times k1\_OFHW) + (10/52 \times k2\_OFHW)$	1/yr	Degradation coefficient for digested OFHW	Fractions denote the relative amounts of fast- or slow-degrading C in the degradable C pool (see also Table 1): k1—fast turnover fraction, k2—slow turnover fraction). 77% cattle, 23% pig manure [8]
Leaching_and_erosion	Agricultural_soil $\times$ 0.01	kg C/yr		Assumption
Manure_production	24,700,000	t DM	Amount of manure (dry mass, DM) produced in the case study area	[8]
OFHW_production	70,000,000	t DM	Amount of manure (DM) produced in the case study area	[8]
Sludge_production	41,500,000	t DM	Amount of manure (DM) produced in the case study area	[8]

**Table A3.** Definition of stocks used in the STELLA model.

Stock	Definition	Unit	Decription
Agricultural_soil	$\text{Agricultural\_soil}(t - dt) + (\text{Digested\_sludge} + \text{Digested\_manure} + \text{Crop\_residues} + \text{Manure\_direct\_application} + \text{"Co-digested\_manure\_OFHW"} + \text{"Co-digested\_sludge\_OFHW"} - \text{Losses} - \text{Mineralization}) \times dt$	kg C	C turnover/accumulation of agricultural soil
Anaerobic_digestion_manure_and_OFHW	$\text{Anaerobic\_digestion\_manure\_and\_OFHW}(t - dt) + (\text{Manure\_to\_AD} + \text{OFHW\_to\_AD\_manure} - \text{Digested\_manure} - \text{Emissions\_to\_air\_AD\_manure} - \text{"Co-digested\_manure\_OFHW"}) \times dt$	kg C	C turnover of manure-based AD plants
Anaerobic_digestion_sludge_and_OFHW	$\text{Anaerobic\_digestion\_sludge\_and\_OFHW}(t - dt) + (\text{OFHW\_to\_AD\_sludge} + \text{Sludge} - \text{Digested\_sludge} - \text{Emissions\_to\_air\_AD\_sludge} - \text{"Co-digested\_sludge\_OFHW"}) \times dt$	kg C	C turnover of sludge-based AD plants
Waste-to-Energy	$\text{"Waste-to-Energy"}(t - dt) + (\text{OFHW\_to\_WtE} - \text{Emissions\_to\_air\_WtE}) \times dt$	kg C	C turnover of WtE plant
Atmosphere	$\text{Atmosphere}(t - dt) + (\text{Emissions\_to\_air\_WtE} + \text{Emissions\_to\_air\_AD\_manure} + \text{Emissions\_to\_air\_AD\_sludge} + \text{Mineralization} - \text{Harvest}) \times dt$	kg C	C turnover/accumulation in the atmosphere caused by the processes in the case study

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