Supplementary

Energy crops in regional biogas systems: an integrative spatial LCA to assess the influence of crop mix and location on cultivation GHG emissions

S 1.1 Overview of RELCA model

Figure S1. Provides the overview of the different steps involved in the RELCA "REgional Life Cycle inventory approach". A region is defined as one scale lower than a country. RELCA integrates regionally distributed biomass inventory with regionally distributed biobased conversion plant data. It is an attributional life cycle accounting approach, which is retrospective and complies with the ISO LCA standards [1], as well as GHG accounting method of the Intergovernmental Panel on Climate Change IPCC [2].

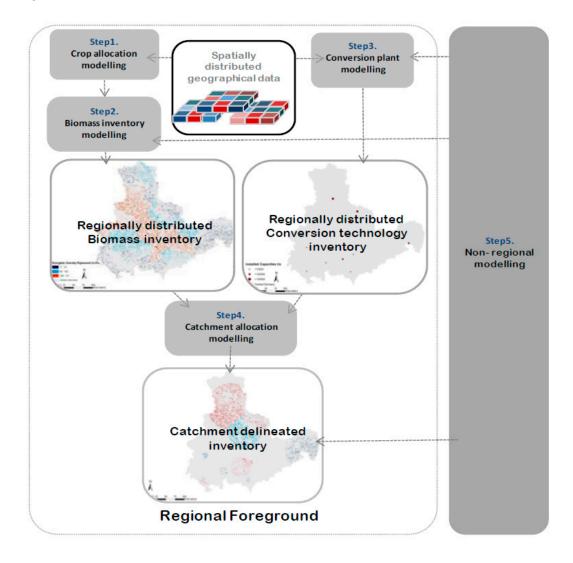


Figure S1. the overview of the different modelling steps associated with RELCA (Adapted from [3,4]).

RELCA has been used to determine the regional distribution of GHG emissions from the foreground activities, as well as GHG emissions from non- regional activities (indirect burdens). The latter refers to the associated activities producing these flows and is assumed to be outside of the region, along with their associated environmental burdens (i.e., released anywhere else but the region of focus and are therefore not considered with a spatial orientation). RELCA combines conventional

geographical modelling and catchment delineation to assess the potential environmental implications of bioenergy configurations (i.e. bioenergy plants and their biomass catchments) within a region. For further detailed description, please refer to [3,4]. For this modelling exercise, we changed the order of the RELCA approach, starting with conversion modelling first, due to modelling the digestate.

S1.2 Spatial indicators relationship to size (installed capacity)

Configurations of feedstock refer to the combination of all crops being supplied within a biogas catchment, in the case presented in this paper, MS, Cer and GS. The land areal demands (LADs) of biogas catchments calculated here for the CG region were for direct land use only, i.e., related to the crops directly feed into the biogas plants.

For LADs there is no clear relationship to size of the particular biogas plant (i.e., installed capacity), Figure S2a. For emission intensity (EI) of the biogas catchments, a weak logarithmic relationship with installed capacity was observed Figure S2b.

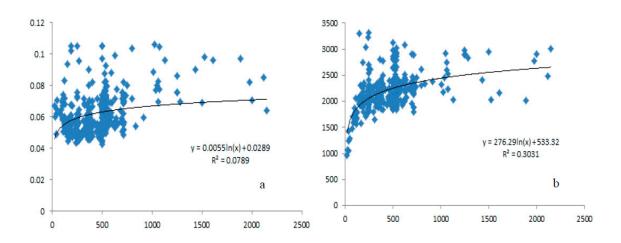


Figure S2. a) Installed capacity, x axis (kW) against LAD values, y axis (haMWhel⁻¹); b) Installed capacity, x axis (kW) against EI values, y axis (CO_{2eq}ha⁻¹). Graphs exclude two outlier values, for which installed capacity were greater than 2,500 kW (n = 423).

S1.3 Crop Allocation Modelling (CRAM)

The crop allocation modelling or CRAM approach of Wochele et al. [5] was implemented to determine the potential regional distribution of the different crop types; maize silage (MS), grass silage (GS) and cereal grains (Cer), which were used in regional biogas production for 2010/2011. In order to implement the CRAM approach, two land cover data sets, the Corine land cover [6] and Atkis® [7] (a digital topographic map), were organised into land use parcels or grid cells of 6.25 hectares (250 × 250 m) using the Fishnet function of ESRI ArcGIS 10.1® [8] and harmonised. For each 6.25 hectare grid cell, important regional geographical variables (e.g., climate, soil types, agricultural suitability) were also overlaid. CRAM then uses the geographical attributes of the gridded land use layer (e.g., soil type, slope) with regional cropping statistics for the year of focus [9], to produce a regionally distributed energy crop layer. The CRAM simulated distribution of crops and yields can be seen in Figure S3 A-C.

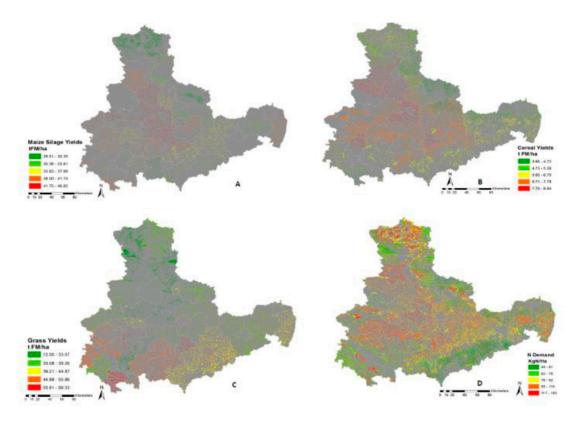


Figure S3. Simulated CRAM crop distributions and yields for the CG region for: A) Maissilage, B) Cereals, C) Grasslands, as well as D) the estimation of Nitrogen fertilizer demand for all biogas crops for the year of modelling (Section S1.6). Maps produced using Arc GIS® software by ESRI (adapted from: [10]).

S1.4 Generation of model biogas plants and biogas clusters

The biogas conversion step is outside the scope of this paper, for greater detail on the life cycle inventory (LCI) development of the biogas plants and the use of biogas clusters please refer to [10]. For this paper what is relevant is the net electricity production of a biogas plant and the resulting land area demand (i.e., catchment) to produce the associated amount of net electricity (main article section 2.6). List of the regional clusters identified by [1], as well as the number of affiliated biogas plants are outlined in Table S1.

Classes	Cluster 1	Size kWel	Feedstock ²	Number ³
SLdom	1	< 150	Mans	27
SLdom	2	150-500	Mans	126
ECdom	3	150-500	EC	19
SLdom	4	501-1000	Mans	65
ECdom	5	501-1000	EC	19
ECdom	6	> 1000	EC	10
SLdom	7	150-500	Na	76
SLdom	8	501-1000	Na	69
ECdom	9	> 1000	Na	14

Table S1. Overview of biogas cluster, class and number of biogas plants per cluster for the CG region (adapted from [10]).

1). Clusters (CL) 1–6 had a subset with adequate data to generate model biogas plants, whereas CL7– 9 had no data available with regards to dominant feedstock and therefore, model plants were developed for these plants based on an analysis of mean feedstocks associated with the same installed electrical capacity category i.e., CL7 is the average of CL2 and CL3, CL8 is the average of CL4 and CL5, and CL9 was assumed to be the same as CL6. 2) These are the predominant feedstock associated with this cluster (i.e. contributes a greater weight to the feedstock mix). Mans = Animal manures (a mixture of slurry (9%DM) and Manure (25%)). EC = Energy crops: Maize silage; Cereal grains: Rye, Barley, Triticale; N/A = not applicable because they had no feedstock data associated with them. 3) The number of data points or biogas plants associated with each cluster

The quantity of the feedstock (F_i) required to support 1kW installed capacity, was determined for each of the different clusters *j* and are outlined in Table S2. To determine the full amount of feedstock required for a biogas plant associated with a particular cluster *j*, each F_i (Table S2) was then multiplied by the installed capacity (IC_{Bp}) of the individual biogas plant (Equation 1). From this the net electricity of the individual biogas plants could be estimated summing the total methane production of each of the feedstocks based on their methane potentials (CH_{4,i}) and multiplying it by; the energetic content of methane (9.97kWh), the assumed efficiency of the cluster (ŋ_{el}) and accounting for the assumed system losses of 10% (Equation2, Table S3).

$$F_{i,Bp} = (IC_{Bp} \times F_{i,j}) \tag{1}$$

$NE_{BP}(kWhel) = [(F_i \times CH_{4i}) \dots + (F_{i+n,j} \times CH_{4i+n,j})] \times 9.97 \times \eta_{el} \times 0.9$ $\tag{2}$

Table S2. Feedstock (Fi) mixtures in tonnes were calculated for 1 kW installed electricity capacity based on the operating conditions determined for that cluster (t kW⁻¹ installed) (adapted from [1]).

			SLdom ³				ECda	om ³	
Feedstock1	CL1	CL2	CL4	C7	C8	CL3	CL5	CL6	CL9
AS^2	70.71	53.77	43.11	47.41	40.69	7.52	4.02	4.37	4.37
AM^2	2.34	2.10	1.49	2.01	2.26	1.48	2.83	-	-
MS	5.48	6.01	6.34	6.32	7.24	16.15	15.10	15.16	15.16
Cer	1,46	0.82	1.33	0.72	0.99	0.29	0.54	0.61	0.61
GS	1.08	1.84	1.91	1.72	1.77	0.71	0.77	0.25	0.25
Totals	81.07	62.69	54.19	58.18	52.96	26.15	23.25	20.39	20.39
Electrical efficiency (%) ŋel,j	33	38	41	39	41	39	41	43	43

1). Feedstock (Fi): As= Animal slurry, AM = animal manure, MS= Maize silage, Cer= Cereals: Rye, Barley, Triticale, GS= grass leys (intensive grassland on arable land and pastures (extensive grasslands). 2). SLdom= Slurry dominant clusters, ECdom = energy crop dominant clusters. 3). CL 6 and CL9 were assumed to be the same

Table S3. Feedstock parameters and conversion factors assumed for associated feedstock, according to [11–15] (adapted from [10]).

Feedstock (Fi)	AS	AM	MS	Cer	GS
DM%	9–10 *	22-24	31	86	31
ODM ¹ (% DM)	80	85	95	97	90
(bi) Biogas (Nl / kg ODM)	380	450	650	730	600
(CH4,i) Methane potential ² (%)	55	55	52	52	52
(ρi) Biogas Density kg/m ³	1.28	1.28	1.32	1.32	1.32
	1	Nutrients ³			
Total N g/Kg	2.62-4.35	3.8-4.09	4.5	16.5	8.01
NH_N4 g/Kg	1.31-2.17	1.07-1.13	0.48	3.04	1.15
P g/Kg	0.65	1.45	0.79	3.76	1.10
K g/Kg	4.42	5.39	3.98	5.00	5.4
Carbon (%DM) ⁴	36	35	45	45	45

1). Organic Dry matter as a percentage of DM content. 2). Methane potential as a percentage volume of biogas produced. 3). Total N and plant available N (NH_N4) range provided from modelling calculations. 4). Factors derived from CANDY database [15]. * %DM for animal slurries and animal manures estimated as part of calculations relating to slurry credits see [1]. With regards to nutrient balances we assume minimum or no difference between ensiled and fresh feedstocks.

It was also assumed that all plants were operating under ideal conditions (i.e., no breakdowns, readily available feedstock). The regional distribution of the biogas clusters showed that they were broadly spread out across the region, with no significant spatial clustering. However, it must be noted that a greater number of biogas plants belonging to the EC_{dom} clusters were located in the northern part of the CG region in the Federal state of Saxony-Anhalt (Figure S4).

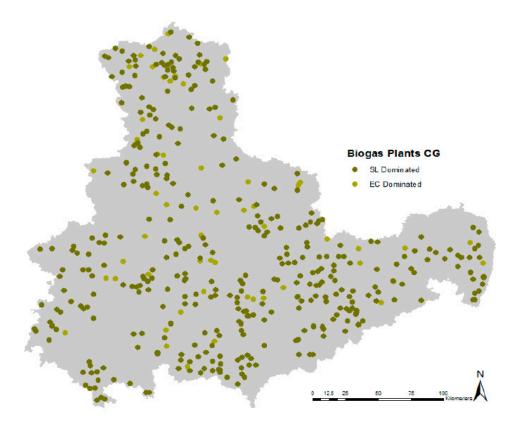


Figure S4. Distribution of biogas plants in CG region, grouped into two classes, slurry dominant (SLdom) and Energy crop dominant (ECdom). (Source: DBFZ, adapted from [10])

S1.5 Regional yields and land use for biogas

Land dedicated to biogas (focus on MS cultivation)

While the data for 2010 was modelled, the best available data to cross check the results was 2011. According to [16], the amount of land devoted to maize for biogas production in Saxony in the year 2011 was approx. 21% of the total cultivated Maize areas (approx. 16,000 ha). For 2010 the modeled results estimated approx. 26% of the land area devoted to maize being used directly by biogas plants (16,463 ha). Therefore, our results are somewhat in the range of what was reported for this Federal state.

Crops	Yields ¹ tFMha ⁻¹	10 years average 2003–2012	Hectares cropped in 2010 (ha)
MS	26.5-46.8 (35.2)	39.03	206,578
Cer		5.21	
Rye	4.09-8.13 (4.86)	5.75	125,676
Barley	3.01-8.64 (7.1)	5.75	380,853
Tricale	3.85-7.71 (4.86)	5.39	55,618
		Grass Silage ²	
Grass Leys	2.63–14.73 (8.25)	_	59,576
Pasture ³	1.8–10.7 (6.82)	-	494,715

Table S4. Overview of the regional yields and total hectares cultivated in 2010 relevant for biogas production Values unless otherwise stated are in tonnes fresh matter per hectare (ha). Data source [10,17,18].

1). Average in brackets and italics. 2). Grass is presented her in tDM ha⁻¹. 3). This is the combination of the grassland statistical categories "Wiesen and Weiden"

According to [19], the total amount of arable land dedicated to *circa* 140 biogas plants was approx. 27,000ha, of which approx. 11,900 ha were devoted to maize silage. For Thuringia O'Keeffe et al.1 modelled 118 biogas plants (i.e., due to data availability etc.), with a total land area of 20,541 ha and a total MS demand of 11,853 ha. Again, for this Federal state, the modelled results fall close to what was reported.

Data for cross checking the land demands for biogas production in Saxony- Anhalt (SA) was limited, according to [20], in 2011 *circa*. 275 biogas plants were in operation in SA, with approx. 55,000 ha of arable land devoted to MS. RELCA modelling for 2010, based on 150 biogas plants (54% of the plants determined by [20]), estimated that 29,883 ha MS were used for biogas production, approx. 54% of the 55,000 ha outlined by [20].

However, there are potentially many discrepancies with these estimates and without better data; it is difficult to know how uncertain our values are.

S1.6 Management modelling and assumptions (adapted from[1])

Rates of digestate application for the different crops were based in accordance with the various national and regional agricultural authorities [12,21–24]. The rate of digestate applied, for each constituent grid cell of a biogas catchment, had to be adjusted for the different crops based on the potential amount of nutrients being supplied from the digestate of the associated biogas plant. Three major modelling constraints were also implemented (see S1.6.3). It was assumed that the digestate was applied using a trailing hose [25]. The fertiliser characteristics for the digestate are outline below and the resulting difference between digestate applied and NPK demand (nitrogen, phosphorus and potassium), was then used to estimate the chemical fertilizer required for each constituent crop grid cell of the biogas catchment.

S 1.6.1 N management flows

To estimate the amount of nitrogen fertiliser applied (N_{applied}) per grid cell, the "N-Basis-Sollwert" method was used (Equation 3). Best farming practices were assumed for all energy crops [12,22,26], with the recommended N rate dependent on yields. The various modelling constraints are outlined in Table S5 and Table S6, for arable land and grassland respectively.

$$N_{applied} = Nrate_{Rec} - N_{min} \pm Add_{adj}$$
(3)

Crops	High yields t ha ⁻¹ a ⁻¹	NrateRec Kg N ha ⁻¹ a ⁻¹	Av. yields tha ⁻¹ a ⁻¹	NrateRec Kg N ha ⁻¹ a ⁻¹	Low yields tha ⁻¹ a ⁻¹	NrateRec Kg N ha ⁻¹ a ⁻¹
Maize silage	>50	210	35-50	190	<35	170
Rye	>7	140	5-7	120	<5	110
Barley	>8	120	5.5-8	140	<5.5	150
Triticale	>7	120	5-7	140	<5	150

Table S5. Modelling constraints for nitrogen fertiliser (N) rates for arable energy crops, based on yields [12]. All values are on an annual basis. Constraints derived from regional reports [12,22,26].

Table S6. Modelling constraints for estimating Nitrogen fertiliser rates (Nrate_{Rec}) for grasslands (kgN ha⁻¹ a⁻¹) using different soil, climate combinations. Constraints derived from regional reports [12,22,26].

Rainfall ¹	Light soils ²	Medium soils	Heavy soils
Low	120	123	-
Medium	120	125	127
High	122	123	130

1). Rainfall was categorised as follows: low < 650mm/a, medium from 650–750mm/a and high > 750mm/a. 2). Soils were classified based on clay content as follows: Light soils < 12%, Medium soils12–25%, Heavy soils > 25%

The average mineralized nitrogen in the soil (*N*_{min}), estimated for each federal state was derived from various regional reports and datasets (Table S7) [12,26–28].

Soil classes ¹	Saxony	Saxony-Anhalt	Thuringia
Sandy	32	33	0
Loamy sand	39	44	38
Sandy Loam	48	48	41
Loam	57	47	44
Loamy clay	58	56	45

Table S7. Assumptions for the N_{min} content of soils based on regional reports.

1 = Sandy soils < 5% clay; Loamy sand 5–12% clay; Sandy Loam 17–25% clay; Loam 25–35%; Loamy clay > 35% clay.

Additionally, if the Akazahl value of a grid cell was less than 40, then the nitrogen fertiliser rate required (*NrateRec*) were adjusted (*Addadj*) by subtracting 10 kg N ha⁻¹.

S1.6.2 Non-N Management flows

Table S8. Crop management practices (excluding N demand) assumed for the CG region, foreground regional flows (All units are kg ha⁻¹ a⁻¹, unless otherwise stated) (Adapted from [10]).

Management	Maize silage	Grass silage ²	Cereals Rye	Barley	Triticale	
Sowing rate ³	0.038	0.003	0.087	0.19	0.19	
		Fertil	isers ⁴			
D Domond	19.65-34.73	1.34-43.95	14.32-22.79	19.39-30.24	13.48-26.22	
P Demand	(26.16)	(20.4)	(14.11)	(24.84)	(17.39)	
KD	105-186	17.3–251	20.45-32.55	27.7-43.2	52-102	
K Demand	(140)	(110)	(24.44)	(35.48)	(74.79)	
		Crop pro	otection ⁵			
Herbicide	2.13	0.68	0.3	0.3	0.3	
Insecticide	0.012					
Fungicide			0.84	0.45	0.05	
Growth					0.05	
Regulator					0.05	

Data relating the upstream production flows are outlined in [10].
 Crass silage relates to two broad categories of grass lands: grass leys (intensive grassland on arable land) and pastures, herbicide was assumed to be only applied to leys.
 Sowing rates are in t ha₁ seeds. For grassland this refers to leys only.
 Fertilisers – Nutrient applied P= phosphorus provided by P2O₅ in fertiliser; K= potassium provided by K2O in fertiliser. P

& K rates/demand were estimated based on assumed take off from yield [21,29–31]. The rates of P and K modelled (mean in brackets) are provided here. CaO = assumed took 1.785 kg of CaCO₃ to neutralise the same area as 1kg of CaO [32] (used to convert to Eco Invent units). For all a blanket amount was assume of 3 t ha₋₁ for arable land and 2t ha₋₁ for grassland [24]. 5). Data on crop protection products and recommended dosages was gathered for the region. Once a final list of plant protection products was identified, the active ingredients of

the crop protection products were determined [33]. The active ingredients associated with a fungicide, herbicide and pesticide products were then cross checked with the national survey data of Roßberg et al. [34].

S1.6.3 Constraints for digestate application

The first constraint was to keep the applied total organic nitrogen below the legally specified limit of 170kg N per ha per year. The second constraint relates to the composition of the digestate and the most appropriate quantity which can be applied in order to avoid excessive over application of P and K (see below). The third constraint was that the amount of digestate applied across all crops should not exceed the amount available to be spread (i.e., how much digestate available).

If initial rates of digestate resulted in a greater application of N or a surplus of P, K (above the specified limit) or exceeded the amount of digestate available, then it was assumed that a lower volume of digestate was applied. The reduction in application rates continued uniformly across the catchment until all constraints were satisfied across all biogas catchments.

		0							
	CL1	CL2	CL3	CL4	CL5	CL6	C7	C8	CL9
Dry matter	6.9	7.03	8.39	7.15	9.24	8.31	7.09	7.27	8.31
Total	3.48-3.52	3.57-3.66	5.36-5.41	3.82-3.90	6.12-6.19	5.9	3.58-3.73	3.82–3.97	5.87-5.91
Nitrogen ^{1,2}	0110 0102	0.07 0.00	0.00 0.11	0.02 0.00	0.12 0.12	017		0102 0000	0.07 0.01
Ammonical									
Nitrogen	1.70–1.78	1.70 - 1.75	1.31 - 1.34	1.71–1.76	1.42 - 1.45	1.2	1.65 - 1.74	1.67–1.76	1.18-1.20
(NH4N)									
P^3	1.97	1.97	2.59	2.09	2.79	2.79	2.11	2.11	2.77
K ³	5.45	5.39	3.09	5.34	3.13	2.35	5.33	5.28	2.31

Table S9. Fertiliser characteristics of digestate associated with each cluster –output from modelling kg m⁻³. Ranges relate to range of biogas plant size associated with a particular cluster. Values unless otherwise stated are in kg m⁻³.

 Values presented are the interquartile ranges. 2). Median spreading rates ranged from 15–25m³ per ha across each of the 9 clusters. 3). P = phosphorus provided by P₂O₅ in fertiliser; K= potassium provided by K₂O in fertiliser

S1.6.4 Phosphorus constraints

Based on regional reports and a series of sensitivity test, the overshot of P was capped at 40 kgP ha⁻¹, thus, also limiting the K application rates.

Table S10. Recommendation for Phosphorus and Potassium application (or overshoot) rates. Derived
from Regional reports [12,22,35,36].

Fertiliser Recommendations ² kg Pha ⁻¹			Fertiliser Recommendations ³ kg Kha ⁻¹			
Soil index	Definition ¹	Arable land	Grasslands	Arable land	Grasslands	
А	Very low	+35 to +50	+27 to +40	+55 to +100	+55 to +100	
В	Low	+10 to +25	+20 to +10	+20 to +50	+20 to +50	
С	Medium	0 to 2*	0	0	0	
D	High	-2to -8	-5 to -10	-20 to -80	-20 to -80	
Е	Very high/Excess	0	0	0	0	

1). Refers to nutrient content of soil refer to P and K. 2). Fertiliser can be added or subtracted from P offtake. The range depends on the CAL-Phosphate found in the soil (mg P per 100 g soil) and soil classification. 3). Fertiliser can be added or subtracted from K offtake. The range depends on the CAL-Potassium found in the soil (mg K per 100 g soil) and soil classification.

S1.7 Nitrogen sourced emissions to air

Nitrous oxide emissions (N₂O) from the cultivation of each associated crop were calculated for each constituent grid cell within the biogas catchment, according to the German national guidelines outlined in [14]. This required estimating emissions using a Tier 2 approach, shown in Equation 4, adapted to calculate the relevant regional flows modelled here.

$$N_2ON = \sum [(E_{Napplied} \times EF_{1Brocks}) + (E_{Nresidues} \times EF_{1IPCC}) + (E_{NNH3Fert} \times EF_{NH3}) + (E_{N_NOfert} \times EF_{NO})]$$

$$(4)$$

S1.7.1 Emission factor- EF1Brocks

Using the Geographical variables outlined in Brocks *et al.* [37] - Table S10, the distribution of the nitrous oxide emission factors EF (kg N₂O per kg N fertilizer applied) could be simulated using MATLAB 2017b [38] based scripts. The resulting CG distribution of emission factors associated with the simulated distribution of energy crops can be seen in Figure S5.

Table S11. Emissions factor EFBrocks [37].

Geo-climate categories	Values ¹
1. Redoximorphic soils ²	1.02
2. Well-aerated & Warm-Dry ³	1.21
3. Well aerated Warm-Wet ⁴	1.64
4. Well-aerated & Cold⁵	4.29

Emission factors are for (%) of chemical nitrogen fertiliser applied. 2). Redoximporhic soils found in the Soil map of Germany [39] (Soil No: 7,8,10,11,12,9,22,23, 24, 28, 43, 47, and 48). 3).. Areas which have ≤100 days of frost and < 600 mm of precipitation. 4). Areas which have <100 days of frost and > 600 mm of precipitation 5). Areas which have ≥ 100 days of frost per year. 4)

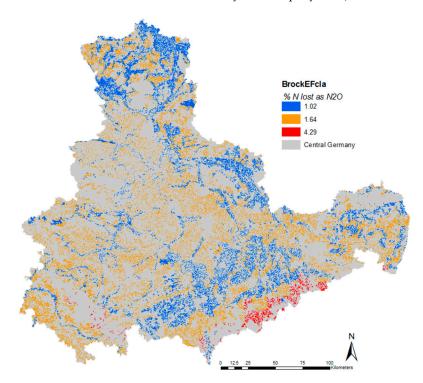


Figure S5. Distribution of Brocks emission factors (EF) for the CGregion.

The Brocks EF was then used in combination with N_{applied} (Section S1.6) to estimate the potential distribution of emissions associated with producing energy crops for biogas systems in CG. For the CG region Brocks EF1, EF 3, and EF4 were determined.

S1.7.2 Estimation of Crop residue N (EnResidues)

The emissions relating to crop residues was estimated using IPCC equation 11.6 [40] (adapted).

$$E_{N_{residues}} = \sum \left(Crop_{DM} \times Frac_{Renew} \times RAG \times NAG \times (1 - Frac_{Renov}) + (NBG \times RG) \right)$$
(5)

Table S12. Assumptions relating to the different variables in Equation 5 for estimating Enresidues. The values used were taken from [14].

	CropDM	Fracrenew	Fracmow	RAG	NAG	NBG	RG
Maize Silage	0.28	1	1	1/1	0.0038	0.007	0.44
Rye	0.86	1	1	1/0.9	0.005	0.011	0.42
Barley	0.86	1	1	1/0.7	0.005	0.0014	0.37
Triticale	0.86	1	1	1/0.9	0.005	0.008	0.42
Grass leys	0.2	0.4	0.33	1/0.5	0.0048	0.012	1.2
Grass pastures	0.2	0.1	0.5	1/0.5	0.005	0.012	0.8

Crop DM= Crop dry matter kgkg ⁻¹. *Fracnew*= Duration of cropped system (x_{renew}, i). *Fracmow*=frequency of harvesting (x_{mow}, i). For annual crops x_{mow}, i = 1. For the exceptions mentioned x_{mow}, i = 0.33. *RAG*= Ratio of

above ground crop residues to yield. *NAG*= Nitrogen content of the above-ground crop residues (x_N, above, i). *Frac_{Remov}*= Fraction of total above ground crop biomass that is removed from the field as a crop product (i.e.

yield of crop harvested to biomass left on field)

S1.7.2 Estimation of Indirect N2O (ENNH3Fert, EN_NOFert,)

It was assumed that digestate was applied using a trailing hose [25]. The loss of NH₃ due to spreading was estimated according to the German national guidelines outlined by [14]. The indirect nitrous oxide emissions resulting from ammonia volatilisation, due to digestate application (Table S14) and chemical fertiliser (Table S15) application were estimated.

Table S13. Factors for estimating the ammonia emissions from spreading of digestate.

	Reference ¹	% Loss
Arable land ²	0.5	22
Grassland ³	0.6	30

1). Reference refers to the "reference situation" for losses of NH₃_N, from which losses for other spreading situations can be estimated. Value in kgNH₃ loss per kg TAN N applied (Total ammonical nitrogen). 2). It was assumed that the digestate was incorporated within 24hours. 3). It was assumed that the digestate was incorporated of > 0.3m

Table S14. Mineral fertilisers NH₃ emissions factors as a function of spring temperature (^oC) taken from₁₄ used to estimate ENNH3Fert for each grid cell (Adapted from [3]).

Fertiliser type	EF
Calcium ammonium nitrate	$0.0008 + 0.0001.ts^{1}$
Anhydrous ammonium ²	0.0127+0.0012.ts
Urea	0.1067+0.0035.ts
Ammonium sulphate ³	0.0107+0.0006.ts
Ammonium nitrate ³	0.0080+0.0001.ts

Spring temperatures (ts) for the months March, April, May, which were found to be in the range of 4.6–8.5 °C.
 Assumed to be similar to Urea ammonium nitrate.
 The statistics referred to an N mixture which was assumed to be 50:50 Ammonium sulphate: Ammonium nitrate (supplementary material, A3).

Nitric oxide emissions of NO were estimated per grid cell using the EF of 0.012 kg NO_N kg⁻¹N applied outlined by Stehfest and Bouwman₄₁ and according to [14]. The global warming potential (GWP) characterisation factors used were according to the IPCC [41] recommendations for a 100year period. These were: GWP of 1 kg CO_{2eq} for CO₂, N₂O was assumed to have a GWP of 265 kg CO_{2eq} and CH₄ was assumed to have a GWP of 28 kg CO_{2eq}.

S1.8 Contribution analysis – non nitrogen cultivation activities

MachineOpsEmis (Direct and Indirect) and other auxiliaries (non-N)

Field operations (e.g. ploughing, harvesting) contributed between 2–11% of the total cultivation emissions for the different crops. Field operations for cereals were found to be significantly higher per hectare. This is because, cereals were distributed on slightly heavier soils (i.e. greater clay content), resulting in a higher diesel demand, which resulted in higher emissions. The category other refers to all other auxiliaries used in the production of the crops (i.e., P, K fertilisers and crop protection products). The combination of these other inputs ranged from 2–12% of the total cultivation emissions. Again Cer had a significantly slightly higher percentage contribution from this category, but in absolute values it was lower than MS.

S1.9 Soil N2O emissions - comparison with the literature

Validating spatially simulated emissions is currently not possible, as the large volume of experimental data to support such simulations simply does not exist. However, if we look at other simulations for parts of the region (Saxony), we can identify that our simulations fall within a similar range.Haas et al [42], determined a range of N₂O emissions for all arable and grassland soils, from less than 1 kg N₂O_N ha⁻¹ to 14.8 kg N₂O_N ha⁻¹, with an average 3.02 kg N₂O_N ha⁻¹. Furthermore, Butterbach-Bahl et al. [43] in their study of Saxony also determined a much more extensive range of emissions from agricultural soils, ranging from 0.5–26.0 kg N₂O_N ha⁻¹. In our approach we determined a range from 0.73 to 8.87 kg N₂O_N ha⁻¹, with a mean value of 2.59 kg N₂O_N ha⁻¹for grassland and arable land for the federal state of Saxony. In our study, the highest emissions were found in the southern part of Saxony, as in this area conditions were found that supported the highest emission factors [37] (S1.7).

In addition to the simulated values, if we select only the German sites from the literature collection of Stehfest and Bowman [44], used to calculate the IPCC emission factor, the ranges of direct N₂O_N emissions coming from mineral soils and across all nitrogen rates, was found to be $0.37-9.73 \text{ kg N}_2\text{O}_N \text{ ha}^{-1}$ for Mais silage (n = 17). For cereals (mainly wheat and barley), the emission range was from $0.41-14.88 \text{ kg N}_2\text{O}_N \text{ ha}^{-1}$ (n = 49) and for Grass silage 1.5–11.2 kg N₂O_N ha⁻¹ (n = 4). Therefore, our values fell in line with those found in the literature.

S1.10 Spatial assessments of biogas catchments

S1.10.1 Catchments of SLdom Clusters

CL2 had the largest number of biogas plants associated with it (n = 127), therefore the biogas plants were scattered across the CG region, but tended to be located in areas with significantly lower EIs, in the south and north. However, approx. one third of the plants were found in the central areas with significantly higher EIs. This combined with the higher proportion of MS in its feedstock mix resulted in CL2 to have significantly higher EIs than CL1. This is because CL1 plants were predominantly (not all) located in areas, south east and north with low EI intensities, hence the significantly lower EI when compared to the other clusters. However, the biggest difference between these clusters relates to the land areal demand, which relates to the crop mixtures being supplied to the biogas plants and their associated yields, which are also influenced by location. The yields of energy crops supplied to the plants in CL1 were found to be comparable to CL2 and to be higher than the median yield for MS (> 2–3%) and Cer (> 2%). Grass silage yields for CL1 was much lower than the median (< 60%). However, as no major differences were observed in relation to the median yields

found in the catchments, the next biggest difference, between the clusters relates to the share of cereals in the mix and its associated areal demands. For CL1, Cer contributed 60% of the total land area demand of the cluster, unlike CL2, for which Cer contributed 36%, CL2 also had the lower share. What this means is that for CL2, the catchment areas required to meet the feedstock demand of the biogas plants were smaller and in turn the overall number of summed emissions was less, thus, translating into a favourable emission profile, per energetic output. Similar trends were also seen across CL8 and CL4, with CL8 having a similar regional distribution to CL2 and CL4 similar to CL1.

S1.10.2 Catchments of ECdom Clusters

The majority of CL5 plants were located in the central part of the region where the EIs were found to be significantly higher. Unlike CL9, where most (not all) of the biogas plants and their catchments were located in the significantly lower emitting southern and northern areas of the CG region. This is the reason why CL5 has the higher EI range in comparison to CL9. In the catchments of CL5, MS yields are approx. 10% higher than the median, the GS yields 6.5% higher than the median, with Cer yields close to the median. In contrast, for CL9, MS and GS yields were found to be lower than the median (7% and 18% respectively). This was the reason for CL9 to have the relatively higher LADs. However, although CL9 had the higher LADs, the relatively lower EIs counteracted the high LAD values leading to the better than expected performance of CL9.

S1.11 Profiles of locations – Results

Locations and catchment class2	SLRed	SLBlue	ECRed	ECBlue
Plants3	11	25	6	5
1 141100		Geo-Climate Variables	Ũ	C C
Mean AZ value	32.0	42.0	45.0	42.0
Clay content (%)	28.3	21.1	21.1	15.0
Total annual	20.5	21.1	21.1	15.0
precipitation (mma-1)	1189	815	799	827
Mean No. Frost days	94	88	88	89
		Yields (ta ⁻¹)		
MS	32.05	27.63	30.25	27.63
Cer	6.06	5.37	5.44	5.31
GS	49.33	32.20	31.8	34.7
		N Demand kgNha ⁻¹		
MS	102.00	123	119.2	120.0
Cer	78.00	84.5	47.7	70.2
GS	64.50	78	36.0	61.0
		% N Demand met with ChemN		
MS	32%	43%	67%	67%
Cer	55%	59%	66%	68%
GS	12%	27%	12%	39%
60	1270	Emission factors4	12/0	0,00
Brocks EF 1	5%	30%	16%	38%
Brocks EF 3	51%	70%	84%	62%
Brocks EF 4	44%	0%	0%	0%
DIOCKS EF 4	44 %		0%	0%
	0104	Emissions (KgCO2eq ha ⁻¹)	2050	1(07
mean Soil N2O	2104	1688	2059	1697
mean Fieldops	182	184	161	159
mean FertN emis	114	198	312	301
(upstream)				
		% Catchment areas5		
MS	55%	52%	86.0%	85.6%
Cer	35%	37%	11.7%	12.5%
GS	11%	11%	2.2%	1.8%
		EI Crops in catchments6		
MS	4204	2822	2795	2247
Cer	2404	1663	1066	835
GS	1993	1629	1490	1135
		Catchments indicators		
EI (kgCO2eq ha-1)	2,775	2,189	2643	2239
LAD (ha MWh-1)	0.058	0.071	0.0964	0.1046
GHG				
(KgCO2eqkWhel-1)	0.169	0.147	0.235	0.225
		Related clusters7		
1		7%		0%
2		17%		3%
3		16%		16%
4		0%		0%
5		11%		0%
6		10%		0%
6 7		10% 1%		0% 1%
8		0%		4%
9		0%		0%

Table S15. Profile for the poorest performing Bcats of the SLdom and ECdom clusters (90th percentile1) with regards to geographical and regional location factors.

1. For SLdom 90th percentile GHGculti emissions > 0.1411 kg $CO_{2eq}kWel^{-1}$, For EC_{dom}, GHGculti emissions > 0.2255 kg $CO_{2eq}kWel^{-1}$. 2. Locations identifiable based on significantly high CultiEmis denoted as "red" or significantly low CultiEmis, denoted as blue, biogas catchments ordered into their main type of feedstock class SLdom or ECdom 3. No. of biogas plants found in these locations. 4. See section S1.7. 5. The percentage catchment occupied by the various crop categories. 6. The median EI for the different crops found in these locations – medianed across all biogas catchments (kgCO_{2eq}ha⁻¹). 7. The percentage of plants associated with each biogas cluster found with these GHGculti categories

Locations and catchment class ²	SLRed	SLBlue	ECRed
Plants ³	11	12	6
	Geo-Clima	te Variables	
Mean AZ value	60.5	43.0	64.0
Clay content (%)	15.0	17.4	15.0
Total annual precipitation			
(mma-1)	918	1026	875
Mean No. Frost days	88	90	87
-	Yield	s (ta-1)	
MS	42.95	37.84	39.53
Cer	7.78	6.58	6.04
GS	47.08	41.27	38.0
	N Demar	nd kgNha-1	
MS	142.00	142	138.2
Cer	102.00	92	49.8
GS	84.50	71	76.6
	% N Demand 1	net with ChemN	
MS	51%	51%	71%
Cer	66%	62%	49%
GS	33%	21%	50%
	Emissio	n factors4	
Brocks EF 1	66%	86%	68%
Brocks EF 3	34%	14%	32%
Brocks EF 4	0%	0%	0%
	Emissions (K	(gCO2eq ha-1)	
mean Soil N2O	1816	1715	1746
mean Fieldops	169	176	168
mean FertN emis	288	191	377
(upstream)	200	191	577
	% Catchr	nent areas5	
MS	52%	53%	85.2%
Cer	36%	36%	12.6%
GS	12%	11%	2.2%
	EI Crops in	catchments6	
MS	2609	2500	2569
Cer	1596	1505	864
GS	1469	1407	1524
	Catchment	ts indicators	
EI ($CO_{2eq} kW_{el}$ -1)	2,082	1,935	2535
LAI (ha MWh ⁻¹)	0.045	0.050	0.0711
GHG (CO _{2eq} kW _{el⁻¹})	0.0993	0.0968	0.178
/		clusters7	
1	7%	0%	
2	17%	3%	
3			16%
4	0%	0%	
5			11%
6			10%
7	1%	1%	
8	0%	4%	
9			0%

Table S16. Profile for the best performing SL_{dom} and EC_{dom} clusters (10th percentile₁) with regards to geographical and regional location factors.

 For SLdom 10th percentile GHGculti emissions < 0.1 kg CO_{2eq} kW_{el⁻¹}, For ECdom, GHGCulti emissions < 0.18 kg CO_{2eq} kW_{el⁻¹}.
 Locations identifiable based on significantly high CultiEmis denoted as "red" or significantly low CultiEmis, denoted as blue, biogas catchments ordered into their main type of feedstock class SLdom or ECdom. 3. No. of biogas plants found in these locations 4. See section S1.7. 5. The percentage catchment occupied by the various crop categories. 6. The median EI for the different crops found in these locations (kg CO_{2eq} kW_{el}⁻¹). 7. The percentage of plants associated with each biogas cluster found with these GHGculti categories

	SI	Ldom	Ec	dom
	Median	Range	Median	Range
GHGculti (Kg CO_{2eq} $kW_{el^{-1}}$)	0.1213	0.087-0.204	0.203	0.164–0.269
EI (KgCO _{2eq} ha ⁻¹)	2138	972–3157	2615	2000-3125
LAD (ha MWel ⁻¹)	0.0566	0.0427-0.0829	0.0811	0.643-0.106

Table S17. Overview of median and range of GHG_{culti} and spatial indicators EI and LAD, aggregated to biogas catchment class level SL_{dom} and EC_{dom}.

S1.12. Sensitivity analysis

Naturally, one of the major uncertainties in this study relates to the emissions from soils, particularly soil N₂O and we have shown that the modelling results here are within the ranges found within the literature (S.1.10). That being said it still remains an uncertainty for many assessments of biobased systems, due to the lack of empirical data.

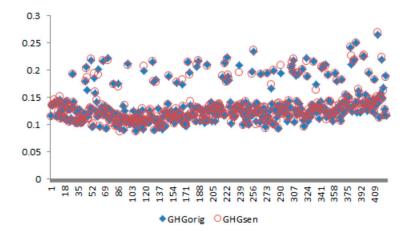


Figure S6. Comparison of GHG_{culti} for base simulation of the regional crop distribution and the second distribution simulation for all biogas plants (xaxis, n=425), y axis is kg CO_{2eq} kW_{el⁻¹}.

Another uncertainty relates to the simulated regional distribution of the crops. Therefore, in order to test the sensitivity of results to this we have ran the CRAM model with a second and different regional distribution of the crops. The mean error in the GHG_{culti} between the two distributions was found to be 0.0003 kg CO_{2eq}kWhel-1, for EI it was 6.60 kg CO_{2eq} ha-1 and for LAD, it was -0.0001 ha MWhel-1. Overall the results changed slightly, but not significantly (Figure S5).

Additionally, it is difficult to compare the approaches and results presented in this paper with the literature, as it is the first (at time of writing); to attempt a more integrative assessment of biogas systems for an entire region. In spite of these uncertainties, we have identified many important points of considerations for a more sustainable future regional biogas supply.

References

 General guide for life cycle assessments-detailed guidance document for life cycle assessment (LCA). ILCD handbook-International Reference Life Cycle Data System, European Union, 2010; http://bookshop.europa.eu/en/international-reference-life-cycle-data-system-ilcd-handbookpbLBNA24708/

- 2. *Guidelines for National Greenhouse Gas Inventories.In: Agriculture, forestry and other land use, vol.* 4;IPCC Intergovernmental Panel of Climate Change,2006; https://www.ipcc-nggip.iges.or.jp/public/
- 3. O'Keeffe, S.; Majer, S.; Drache, C.; Franko, U.; Thrän, D; Modelling biodiesel production within a regional context A comparison with RED Benchmark. *Renewable Energy* **2017**, *108*, 355–370.
- 4. O'Keeffe, S.; Wochele-Marx, S.; Thrän, D.; RELCA: a REgional Life Cycle inventory for Assessing bioenergy systems within a region. *Energy, Sustainability and Society.* **2016**, *6*, 1–19.
- Wochele, S.; Priess, J.; Thrän, D.; O'Keeffe, S. Crop allocation model "CRAM" an approach for dealing with biomass supply from arable land as part of a life cycle inventory. In *EU BC&E Proceedings* 2014; Hoffmann, C., Baxter, D., Maniatis, K., Grassi, A., Helm, P., Ed. ETA-Florence Renewable Energies, Florence, 2014: p.p; 36–40.
- 6. CORINE Land Cover; CLC2006; Federal Environment Agency, DLR-DFD, 2009.
- 7. ATKIS (Authorative Topographic-Cartographic Information System) -DLM Daten, GeoBasis-DE/BKG; BKG, Bundesamt für Katographie und Geodäsie, 2010.
- 8. ESRI, ArcGIS[®] and ArcMap[™] are the intellectual property of Esri (Environmental Systems Research Institute) and are used herein under license. Copyright © Esri. All rights reserved. www.esri.com."
- German Federal Statistical Office, Wiesbaden. Bodenfläche nach Art der tatsächlichen Nutzung, 2010;https://www.destatis.de/DE/Publikationen/Thematisch/LandForstwirtschaft/Flaechennutzung/Bode nflaechennutzungPDF_2030510.pdf?__blob=publicationFile.
- 10. O'Keeffe, S.; Franko, U.; Oehmichen, K.; Daniel-Gromke, J.; Thrän, D. Give them Credit- the GHG performance of regional biogas systems. *GCB Bioenergy*. **2019**, *11*, 791–808.
- Biogas-Messprogramm II 61 Biogasanlagen im Vergleich; FNR, Fachagentur Nachwachsende Rohstoffe e.V., 2009. http://www.fnr-server.de/ftp/pdf/literatur/pdf_385-messprogramm_ii.html
- 12. Düngung in Thüringen 2007 nach "Guter fachlicher Praxis,,; TLL, Thüringer Ministerium für Landwirtschaft, Forsten, Umwelt und Naturschutz. Landwirtschaft und Landschaftspflege in Thüringen, 2007; http://www.tll.de/www/daten/pflanzenproduktion/duengung/dung0108.pdf
- 13. Basisdaten für die Umsetzung der Düngerverordnung. Stand 2013; LfL, Bayerische Landesananstalt für landwirtschaft, 2013. http://www.lfl.bayern.de/iab/duengung/031245/
- 14. Calculations of emission from German Agriculture -National Emission Inventory Report (NIR) 2011 for 2009 -Methods and data; Special Issue (Sonderheft) 342. Thünen-Institue, 2011.
- Franko, U., Modelling approaches of soil organic matter turnover within the CANDY system. In *Evaluation* of Soil Organic Matter Models: Using Existing Long-Term Datasets, Powlson, D. S.; Smith, P.; Smith, J. U., Eds. Springer Berlin Heidelberg: Berlin, Heidelberg, 1996; pp 247–254.
- 16. *Energiepflanzenanbau in Sachsen. Daten und Fackten;* LfULG, Landesamt für Umwelt Landwirtschaft und Geologie,2015; www.smul.sachsen.de/lfulg.
- 17. *Landwirtschaftliche Bodennutzung und pflanzliche Erzeugung*; German Federal Statistical Office, Wiesbaden, 2010.
- 18. Land- und Forstwirtschaft, Fischerei, Bodenfläche nach Art der tatsächlichen Nutzung. German Federal Statistical Office, Wiesbaden.
- 19. Standpunkt, Maisanbau für die Biogaserzeugung in Thüringen; TLL, Thüringer Landesanstalt für landwirtschaft,2011; http://www.tll.de/www/daten/publikationen/standpunkte/st_biom.pdf
- Energiepflanzen f
 ür Biogasanlagen. Sachsen-Anhalt. FNR, Fachagentur Nachwachsende Rohstoffe e.V., 2012;https://mediathek.fnr.de/media/downloadable/files/samples/f/n/fnr_brosch_energiepflanzen_sa_web.pdf

- 21. Hinweise zur umweltgerechten Düngung von Körner-, Silo- und Energiemais; LLG, Landesanstalt für Landwirtschaft, Forsten und Gartenbau, Sachsen Anhalt, 2011; http://www.llg.sachsen-anhalt.de/fileadmin/Bibliothek/Politik_und_Verwaltung/MLU/LLFG/Dokumente/11_mais_dueng.pdf
- 22. Grundlagen der Düngebedarfsermittlung für eine gute fachliche Praxis beim Düngen; LLG, Landesanstalt für Landwirtschaft, Forsten und Gartenbau, Sachsen-Anhalt, 2001; https://llg.sachsen-anhalt.de/fileadmin/Bibliothek/Politik_und_Verwaltung/MLU/LLFG/Dokumente/GL_Duengebedarfserm ittlung.pdf
- 23. *Bestimmung des Kalkbedarfs von Acker- und Grünlandböden;* VDLUFA, Verband Deutscher Landwirtschaftlicher Untersuchungs- und Forschungsanstalten, 2000.
- 24. *Leistungs-Kostenrechnung Pflanzenbau;* KTBL-Kuratorium für Technik und Bauwesen in der Landwirtschaft e. V., 2012; http://daten.ktbl.de/dslkrpflanze/postHv.html#Auswahl
- 25. *Betriebsplanung Landwirtschaft 2010/11 22nd ed.*; KTBL-Kuratorium für Technik und Bauwesen in der Landwirtschaft e. V., ed., Darmstadt, 2010.
- 26. *Düngebedarfsermittlung*. BEFU, Sächsisches Landesamt für Umwelt, Landwirtschaft und Geologie, 2013; http://www.landwirtschaft.sachsen.de/landwirtschaft/1868.html
- 27. Nmin-Vergleichswerte Frühjahr 2010; LLFG, Landesanstalt für Landwirtschaft, Forsten und Gartenbau, Sachsen-Anhalt,2010;http://www.llfg.sachsenanhalt.de/fileadmin/Bibliothek/Politik_und_Verwaltung/MLU/LLFG/Dokumente/Nmin2010_2.pdf
- 28. *Aktueller Rat zur Nmin- und Smin-Situation Thüringer Böden im Frühjahr 2010*. TLL, Thüringer Ministerium für Landwirtschaft, Forsten, Umwelt und Naturschutz, 2010.
- 29. Leitlinie zur effizienten und umweltverträglichen Erzeugung von Silomais zur Fütterung und Nutzung als Gärsubstrat in Biogasanlagen; TLL, Thüringer Ministerium für Landwirtschaft, Forsten, Umwelt und Naturschutz, 2007; http://www.tll.de/ainfo/pdf/sima0807.pdf
- 30. *Leitlinie zur effizienten und umweltverträglichen Erzeugung von Winterroggen*. TLL, Thüringer Ministerium für Landwirtschaft, Forsten, Umwelt und Naturschutz, 2009; http://www.tll.de/ainfo/pdf/ll_wro.pdf
- 31. *Leitlinie zur effizienten und umweltverträglichen Erzeugung von Wintergerste*. TLL, Thüringer Ministerium für Landwirtschaft, Forsten, Umwelt und Naturschutz, 2015; http://www.tll.de/ainfo/pdf/ll_wg.pdf
- 32. *Kalkdünger;* BayWa, 2014; http://www.baywa.de/pflanzenbau_obst/ackerbau_gruenland/duengemittel/kalkduenger/
- 33. *Online database on plant protection products.* BVL, Bundesamt für Verbraucherschutz und Lebesmittlesicherheit, 2013; https://portal.bvl.bund.de/psm/jsp/
- 34. Roßberg D.; Erhebungen zur Anwendung von Pflanzenschutzmitteln in der Praxis im Jahr 2011. *Journal für Kulturpflanzen*. **2013**, 65, 141–151.
- 35. *Phosphordüngung nach Bodenuntersuchung und Pflanzenbedarf;* VDLUFA, Verband Deutscher Landwirtschaftlicher Untersuchungs- und Forschungsanstalten, 1997.
- 36. *Kalium-Düngung nach Bodenuntersuchung und Pflanzenbedarf Richtwerte für die Gehaltsklasse C;* VDLUFA, Verband Deutscher Landwirtschaftlicher Untersuchungs- und Forschungsanstalten, 1998.
- 37. Brocks, S.; Jungkunst, H. F.; Bareth G. A regionally disaggregated inventory of nitrous oxide emissions from agricultural soils in Germany-A GIS based empirical approach. *ERDKUNDE* **2014**, 125–144.
- 38. *MATLAB and Statistics Toolbox, Release 2017b;* MATLAB, The MathWorks, Inc., Natick, Massachusetts, United States.
- 39. Soil Map of Germany 1:1.000.000; BÜK 1000, Federal Institute for Geosciences and Natural Resources, 2007.

- 40. IPCC; Guidelines for National Greenhouse Gas Inventories, prepared by the National Greenhouse Gas Inventories Programme, edited by: Eggleston, H.S, Buendia, L., Miwa, K., Ngara, T. and Tanabe, K., Volume 4, Chapter 11, N2O emissions from managed soils, and CO2 emissions from lime and urea application, IGES, Hayama, Japan, **2006**.
- 41. IPCC, Climate Change 2014: Synthesis Report . Contribution of Working Groups I, II and III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Core Writing Team, R.K. Pachauri and L.A. Meyer (eds.)]. IPCC, Geneva, Switzerland, 151 pp. **2014**.
- 42. Haas, E.; Klatt, S.; Fröhlich, A.; Kraft, P.; Werner, C.; Kiese, R.; Grote, R.; Breuer, L.; Butterbach-Bahl, K. Landscape DNDC: a process model for simulation of biosphere–atmosphere–hydrosphere exchange processes at site and regional scale. *Landscape Ecol* **2013**, *28*, 615–636.
- Butterbach-Bahl, K.; Kesik, M.; Miehle, P.; Papen, H.; Li, C. Quantifying the regional source strength of Ntrace gases across agricultural and forest ecosystems with process based models. *Plant and Soil* 2004, 260, 311-329.
- Stehfest, E.; Bouwman, L. N2O and NO emission from agricultural fields and soils under natural vegetation: summarizing available measurement data and modeling of global annual emissions. *Nutr Cycl Agroecosyst* 2006, 74, 207–228.