Brown Coal reduces Slurry Emissions

Basic Investigations of the Reduction of Emissions from Slurry through Treatment with Fine-Grain Brown Coal

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Fine-grain brown coal from the Lausitz region has been tested in the laboratory at dosages of 3 and 5 mass-% as an alternative to swimming layers of slurry and straw. Three series of trials were carried out using slurry with dry matter contents (DM) common in practice: cattle slurry with DM = 5%, 7%, and 8.9% as well as pig slurry having a DM = 5.2% and 7.6%

Together with floating slurry particles, fine-grain brown coal forms dense swimming layers, which cause a drastic reduction in odorant emission. In cattle slurry stored under undisturbed conditions, the reduction amounted to approximately 70% and, after homogenizing, to 90% to 98%. In pig slurry, a 92% to 98% reduction was achieved, both before and after homogenizing.

The addition of brown coal reduces the pH-value of the slurry by about 0.2 to 0.3 units and NH_3 release by up to 30%.

The treatment of pig slurry with fine-grain brown coal improves the conditions for effective solid-liquid separation into an easily conveyable liquid and a well-transportable solid with high contents of nutrients and humin.

Investigations regarding the use of the large humin content of this brown coal for the long-term formation of humus remain to be carried out. In addition, questions concerning process engineering and the economic efficiency of slurry treatment with brown coal still need to be answered.

Keywords
Cattle and pig slurry, emissions, odour, brown coal, solid-liquid separation

Introduction

Animal husbandry causes some significant emission problems. From the stalls and from the storage containers as well as during the spreading of liquid and solid manure, gases and odours are released. Odours often lead to disputes between animal producers and neighbours who are annoyed. Lawsuits are no rarity in such cases.

For this reason and because the slurry must be stored for at least six months (at some locations up to 10 months), different container covers have been introduced into practice in recent years [1, 2]. These constitute a compromise between the emission-reducing effect and service life on the one hand and the expenses on the other hand. Solid covers, such as massive coverings or tent roofs, are highly efficient, but expensive. Swimming covers, such as plastic film, also reduce emissions considerably. In addition, they are cheaper and may very well reach a service life of up to ten years [3].

Natural swimming covers, which form on top of stored slurry if stem-like feed residues are present (cattle) or if litter is specially used for this purpose (pigs), are recognized as an emission-reducing measure by the environmental authorities [4]. Chopped straw spread on top of the stored slurry and mixed in also leads to a dense swimming layer. However, its effect on the release of nitrous oxide, carbon dioxide, and methane has not yet been clarified definitively [5, 6]. During spreading, the stem particles also cause blockage problems in the distribution organs, especially if drag hose- and injection techniques are employed. Therefore, a swimming layer without stem particles would be very advantageous.

Initial tests of fine-grain brown coal from the early Tertiary carried out by Lausitzer Braunkohle AG showed that odour release from cattle slurry can be reduced significantly directly after mixing because the adsorption of odorants to the brown coal begins immediately. The swimming layer which forms from light slurry particles and the coal particles results in an additional, durable reduction in odour emission.

This method, which is still in the initial stage of development, differs considerably from the "slurry treatment through brown coal" technique developed and tested by the Rheinbraun company, which has so far not been able to establish itself due to economic reasons. In this technique, 15 mass-% of mine-moist raw brown coal was mixed in with the goal of treating the products of solid-liquid separation further in order to guarantee their transportability and marketability [7, 8].

The objective of slurry treatment with only 5 mass-% of fine-grain brown coal from the Lausitz region mainly consists in a strong reduction of odour emission. In addition, one may assume that, on the one hand, the humin content of the brown coal can make a contribution towards soil amelioration in the form of stable humus, while on the other hand the nutrients contained in the slurry are bound to the brown coal, which provides long-term and environmentally friendly availability. This contribution reports on investigations at the laboratory scale which were mainly aimed at the reduction of odour and gas release and at the quantification of sedimentation behaviour during the storage of cattle and pig slurry.

Material and Methods

Since the original feed substances and the digestion processes of the animals are different, cattle and pig slurry exhibit differentiated physical behaviour even though their dry matter content is the same. In trial 1, cattle slurry was therefore tested with a DM = 7% and 5%, but with the same brown coal dose of 3%. In trial 2, the original DM content amounted to 8.9% with 3% and 5% of added brown

coal. In trial 3, pig slurry having a DM = 5.2% and 7.6% with a brown coal dose of 3% and 5% was investigated (**table 1**).
Thus the practical DM contents were taken into account as well as the more pronounced sedimentation behaviour of pig slurry. In addition, an attempt was made to reduce the percentage of brown coal even further than originally planned.
Fine-grain brown coal is strongly hydrophobic. For this reason, special care had to be taken to mix the coal particles in completely. In trials 1 and 2, each batch was given a mixing and reaction time of 20 min. The mixing equipment consisted of a funnel-shaped container and a simple agitating body with a cross vane (R = 300 min^{-1}).
For the production of 50 l batches of pig slurry and brown coal (trial 3), more efficient mixing equipment was used (**figure 1**).
The slurry and the fine-grain brown coal were mixed by an agitator with different directions of rotation and by pumping. At different agitator and pump parameters, mixing duration amounted to 10 min (**table 2**).
As shown above, the three samples with the same initial DM content were treated identically. Stirring alone requires too much time for mixing and involves the danger of coal particles not being completely absorbed by the slurry. Intensive mixing through heavy turbulence or the mechanical load which occurs in a screw pump allows the goal to be met.
The uncovered laboratory containers for the storage of the slurry-coal mix were put up in an air-conditioned room (**figure 2**). The chosen temperature was 20°C in order to achieve high release rates of odorants and gases.

The measured experimental values were:
- pH-value and slurry temperature
- odorant and gas emissions
- sedimentation behaviour
- chemical substances of content

For the time of gas concentration measurement, the containers are covered (**figure 3**). Outdoor air with an air flow rate of 1,500 l/h is led over the slurry surface. The escaping air, which is contaminated with odorants and gases, is filled into bags or sucked in by the multi-gas monitor (Brüel & Kjær, model 1302) for gas analyses. The odorant concentrations are determined by the olfactometer TO 7.
As shown in Figure 2, the measuring scale at the plexiglass containers enables the position of the different temporally variable slurry layers, such as the sinking, liquid, and swimming layers, to be read.

Table 1: Investigated variants (kind of slurry, DM content, addition of brown coal)

	Control			Variant 1			Variant 2		
	DM_{orig} %	BC %	DM_{mix} %	DM_{orig} %	BC %	DM_{mixh} %	DM_{orig} %	BC %	DM_{mix} %
Trial 1 **Cattle slurry**	7.0	0	7.0	7.0	3	10.3	5.0	3	7.5
Trial 2 **Cattle slurry**	8.9	0	8.9	8.9	3	11.4	8.9	5	12.7
Trial 3 **Pig slurry**	5.2 7.6	0 0	5.2 7.6	5.2 7.6	3 3	7.2 10.0	5.2 7.6	5 5	8.7 11.3

Table 2: Mixing parameters for the production of the slurry-coal mixtures

Initial DM content %	Added brown coal %	Agitator rpm min^{-1}	Duration left min	Duration right min	Pump rpm min^{-1}	Pump Duration min
5.2	5	1,700	6	4	290	10
	3	1,700	6	4	290	10
	0	1,700	6	4	290	10
7.6	5	1,700	6		290	8
		1,700		2	290	
		2,043		2	340	2
7.6	3	1,700	6		290	8
		1,700		2	290	
		2,043		2	340	2
7.6	0	1,700	6		290	8
		1,700		2	290	
		2,043		2	340	2

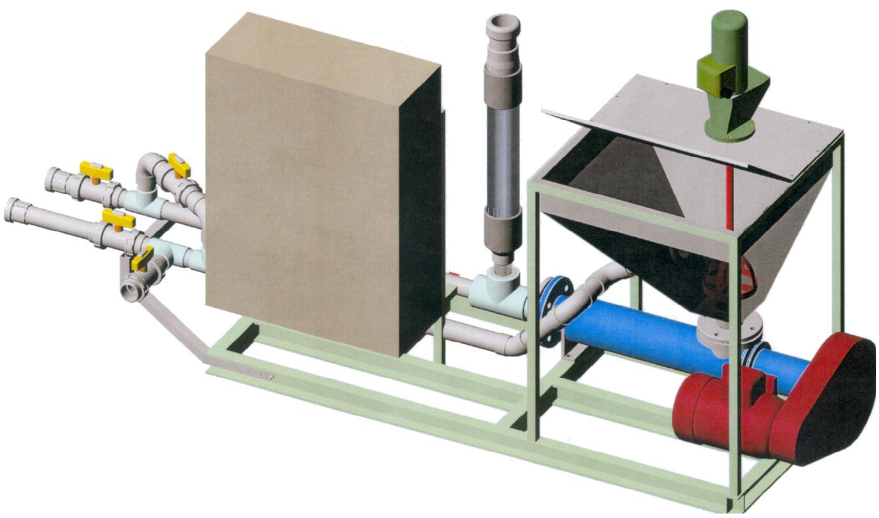


Figure 1: Mixing equipment with agitator and rotary screw pump

Results and Discussion

Substances of Content

The dry matter contents of the control samples were in the typical range for cattle and pig farms (**table 3**).
They changed according to the mass relations of slurry and fine-grain brown coal. The total nitrogen (N_{tot}) and ammonium nitrogen (NH_4-N) contents were generally within the range of the standard values [9].
In trial 2, the P_{tot}- and Mg_{tot}-content was within the standard range, whereas the K_{tot} value was 45% below the standard value. In trial 3, the K_{tot}- and Mg_{tot}-contents corresponded to those found under practical conditions, while the P_{tot} value was 50% lower. These deviations are meaningless for the investigations carried out here because these chemical components do not change when carbon is added. In all cases, brown coal slightly lowers the pH-value, which may contribute to the reduction of NH_3 release.

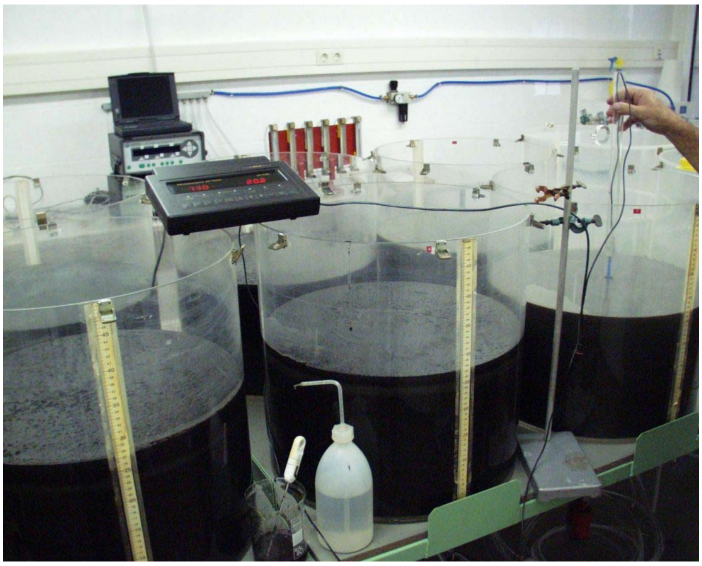


Figure 2: Laboratory container filled with 50 l of slurry-coal mixture

Figure 3: Covered laboratory containers during gas concentration measurement and the filling of the air bags for olfactometric analyses

Odorant Emissions

For sampling and the measurement of o-dorant concentrations, different conditions of the stored slurry were chosen.

In all trials, air samples were taken uniformly 20 hours after the trial had been set up and at the end of each individual experiment. The total duration of the trials differed in the individual series and amounted to 21 days (trial 1), 11 days (trial 2), and 13 days (trial 3).

However, the more significant distinctive criterion was sampling before or after three-minute homogenizing. In practice, one tries whenever possible not to destroy the swimming layer which has formed on the slurry surface in order to use its emission-reducing effect. Once in a while, however, homogenizing is necessary in order to avoid solid sediments on the bottom of the container or in order to guarantee even distribution of plant nutrients during spreading on fields. The results show clear trends (**table 4**).

On the one hand, odour release from pig slurry <u>without</u> brown coal (control) is higher than odour release from cattle slurry, both in an undisturbed condition and after stirring.

On the other hand, odour concentration over slurry increases with storage duration, especially if one compares trials 1 and 3. In practice, long-term storage can rather be expected to lead to a decrease, especially since the slurry temperatures in autumn/winter range below 20°C.

The outstanding effect is the reduction of odour emission through slurry treatment with fine-grain brown coal. Before homogenizing, this effect amounts to at least 70% in cattle slurry with 3% BC. After homogenizing, the respective values are 90 to 98%.

In pig slurry, brown coal is even more efficient with regard to odour reduction, as clearly shown in **Figure 4**.

Before and after homogenizing, the reduction effect amounts to 92% to 99% and 92% to 98% respectively.

The question of whether an increase in the amount of brown coal added from 3% to 5% provides further effects is of economic interest. Odour release is reduced further, but only by an additional 0.5% to 6%, which is insignificant given reduction rates of more than 90%.

pH-Value

The addition of brown coal causes the pH-values of the slurry to diminish. This decrease is slightly more pronounced in cattle slurry than in pig slurry. However, the reduction of the pH-value is not significant. It amounts to 0.2 to 0.3 units (table 3).

The gradation in the order control, slurry + 3% BC, and slurry + 5% BC remained the same in trials 1 and 2 (**figure 5**). This provides a partial explanation for ammonia release, which is graded in the same manner.

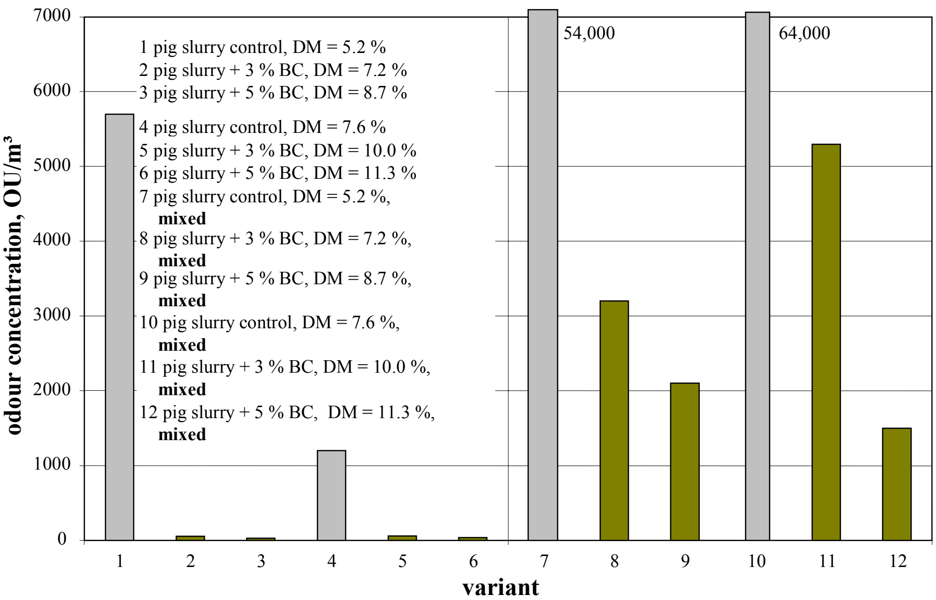


Figure 4: Odour concentration over pig slurry after 13 days of storage

Table 3: Substances of content and pH values on the mixing day

	DM %[1)]	N_{tot} g/kg DM	NH_4 - N g/kg DM	NH_4 - N / N_{tot}	P_{tot} g/kg DM	K_{tot} g/kg DM	Mg_{tot} g/kg DM	pH
Trial 1, Cattle slurry								
Control	7.0	49.2	25.2	0.513				7.45
+ 3 % BC	10.3	33.4	16.9	0.507				6.98
+ 3 % BC	7.5	34.0	16.5	0.484				7.10
Trial 2, Cattle slurry								
Control	8.9	50.8	22.3	0.439	8.4	29.1	6.7	6.93
+ 3 % BC	11.4	41.2	16.8	0.408	6.4	23.0	6.1	6.71
+ 5 % BC	12.7	33.1	14.9	0.450	5.8	19.8	5.7	6.64
Trial 3, Pig slurry								
Control	5.2	75.3	44.4	0.590	30.0	32.4	9.7	6.86
+ 3 % BC	7.2	52.9	27.8	0.518	19.8	19.5	7.3	6.80
+ 5 % BC	8.7	46.2	24.2	0.524	15.4	16.3	6.4	6.65
Control	7.6	63.2	29.0	0.459	28.3	23.4	8.9	6.76
+ 3 % BC	10.0	48.1	23.1	0.480	21.3	17.4	7.4	6.66
+ 5 % BC	11.3	41.6	16.8	0.404	18.4	14.2	7.0	6.55

1) in relation to the original substance

Table 4: Odorant concentrations over slurry without and with the addition of brown coal

	Timer after mixing	before stirring OU/m³			after stirring OU/m³		
Trial 1		**Control**	**+ 3 % BC**	**+ 3 % BC**	**Control**	**+ 3 % BC**	**+ 3 % BC**
Cattle slurry	20 h	100	28	27	140[1)]	85	120
	21 d	210	1,100	180	19,000	470	450
Trial 2		**Control**	**+ 3 % BC**	**+ 5 % BC**	**Control**	**+ 3 % BC**	**+ 5 % BK**
Cattle slurry	20 h	1,200	250	80	30,000	500	190
	11 d	540	160	76	2,000	200	190
Trial 3		**Control**	**+ 3 % BC**	**+ 5 % BC**	**Control**	**+ 3 % BC**	**+ 5 % BK**
Pig slurry DM_1	20 h	450	34	32	43,000	790	590
	13 d	5,700	57	30	54,000	3,200	2,100
DM_2	20 h	450	36	25	48,000	1,300	1,100
	13 d	1,200	60	38	64,000	5,300	1,500

1) covered with a 1 mm layer of brown coal after stirring

Gas Emissions

Of the environmentally and climatically relevant gases, ammonia (NH_3) and methane (CH_4) were detected. Nitrous oxide (N_2O), however, was not found.

The single daily measurements exhibited typical courses (**figure 6**).

Values of outdoor air concentration are followed by 12 values of concentration over the different samples, of which the first two are not used for mean value formation and evaluation. The NH_3 values show clear gradation according to the DM content and brown coal dosage.

This trend continues over the entire duration of the trials. On all measuring days, NH_3 concentration exhibits clear differentiation according to BC addition (**figure 7**).

With increasing storage duration, the amount of reduction grows as well. This is caused by the swimming layer of slurry

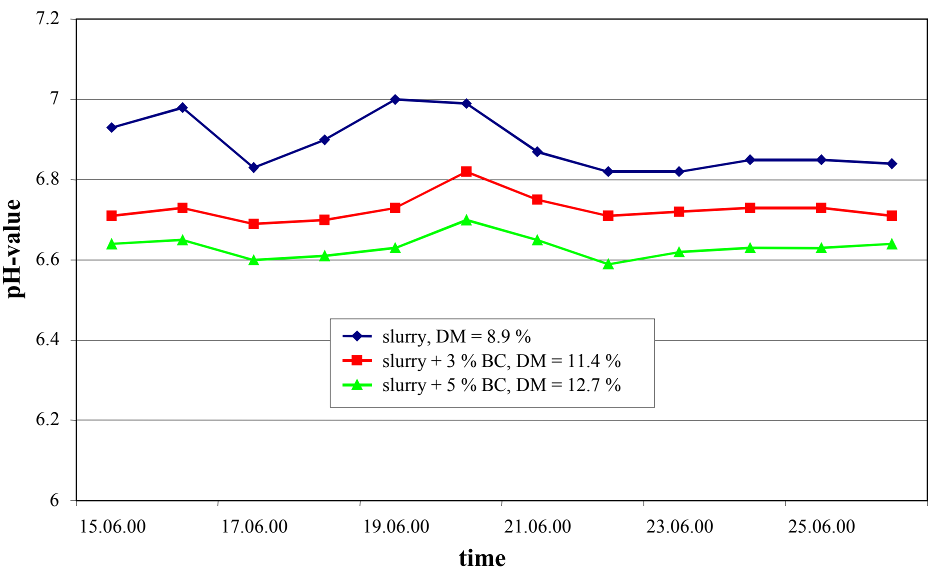


Figure 5: Course of the pH-values over the storage time; trial 2, cattle slurry

and brown coal particles impeding NH_3 release.
For the above-mentioned storage duration, the mean reduction of NH_3 release amounted to 17% (3% BC) and 28% (5% BC) (**figure 8**).
Methane emission was also reduced through the addition of brown coal. On average, reduction amounted to 17% (3% BC) and 29% (5% BC) (figure 8). These differences are statistically secured.
When pig slurry was treated with fine-grain brown coal, the reduction rates for NH_3 and CH_4 release were less obvious. In the lower DM range, they were in the order of 30% for ammonia. The samples with a higher DM content, however, did not exhibit any statistically secured differences. The results for methane were similar.
In the literature, ammonia reduction rates of 70% (cattle slurry) and 30% (pig slurry) due to a natural swimming layer are mentioned. If chopped straw was used, these values reach 80% (cattle and pig slurry) [10]. Artificial swimming layers with fine-grain brown coal cause emissions from cattle slurry to diminish by up to 30%. For pig slurry, the result is not clear. This shows that the release of odorants and gases do not correlate.

Sedimentation behaviour
Cattle slurry contains more colloidal particles than pig slurry. For this reason, cattle slurry is more viscous than pig slurry. In a quiescent condition (e.g. during storage), the suspended and colloidal particles behave differently despite the same dry matter content so that the slurry also sediments differently.
If the layer courses of the slurry samples, which are measured daily, are connected to form a series, layer diagrams for the duration of storage are gained.
Thus, it can be determined that the cattle slurry used for trial 2 both with and without the addition of brown coal separates into layers only to an insignificant extent (**figure 9**).
On the original slurry, a thin swimming layer interspersed with gas bubbles formed. As of the fifth day, a liquid layer developed below (figure 9a). Both together account for only 6% of the total volume. 94% is virtually homogeneous slurry.
In general, slurry with a 3% addition of brown coal shows the same behaviour (figure 9b). The brown coal particles make the swimming layer denser and the sedimented slurry more compact. The vertical movement of the solid particles is largely inhibited. Gases can diffuse through the

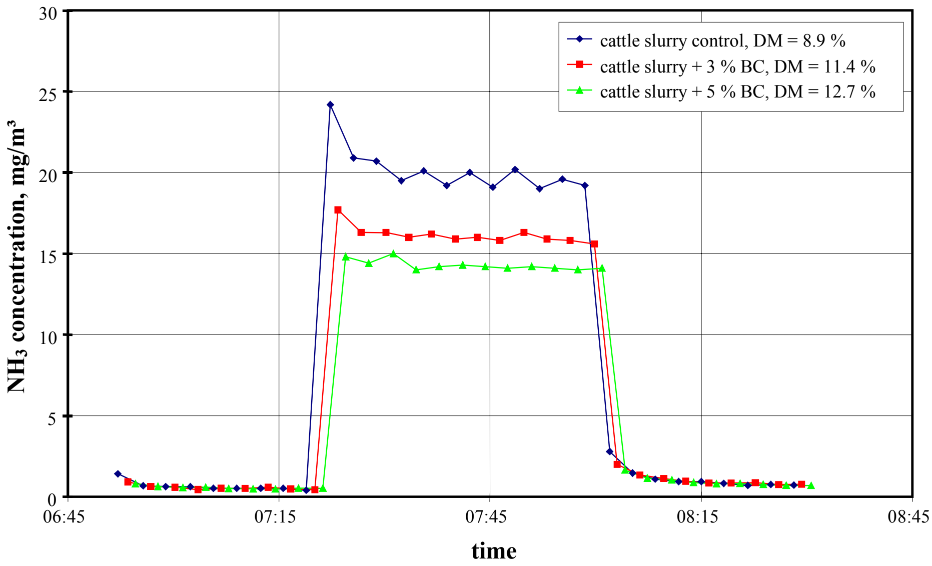


Figure 6: Course of the ammonia concentration over cattle slurry (single measurement, trial 2, 7th day)

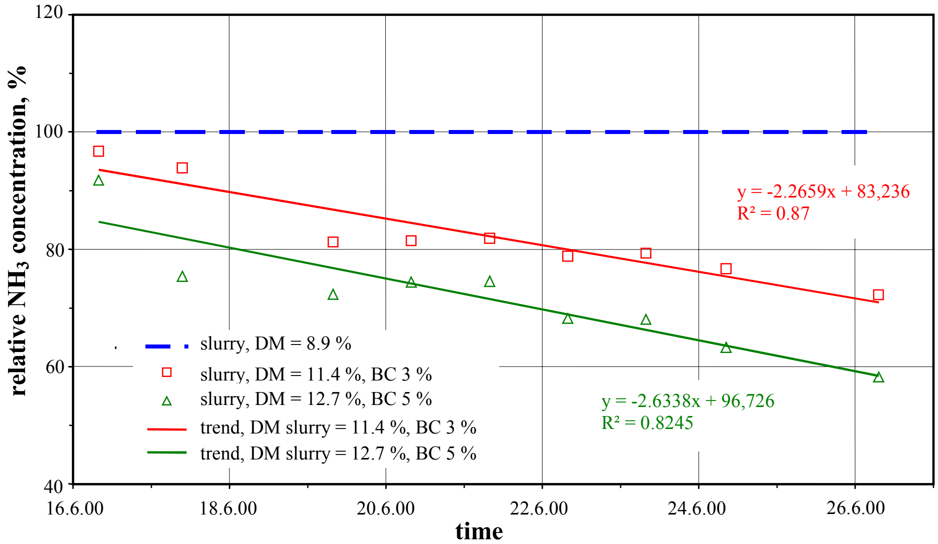


Figure 7: Relative ammonia concentration over cattle slurry as a function of storage time

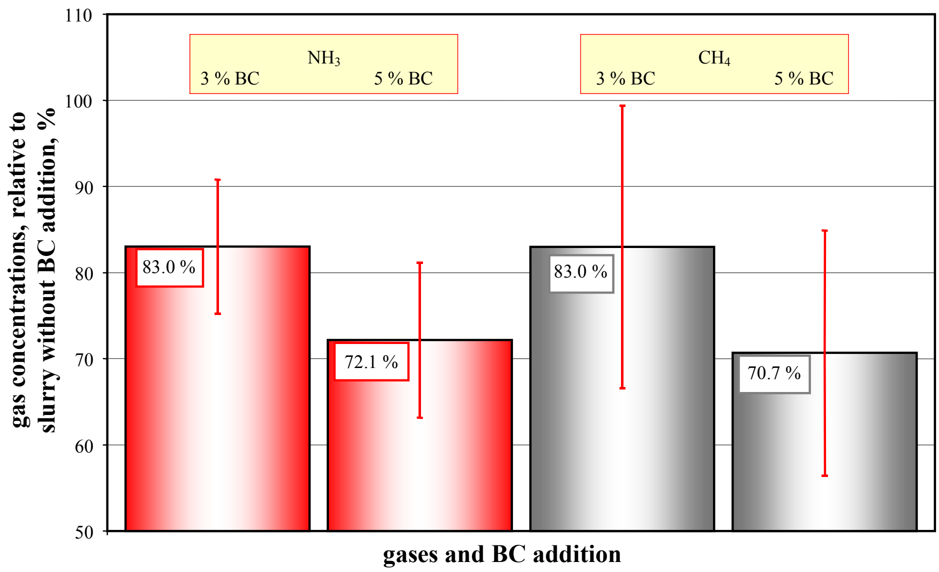
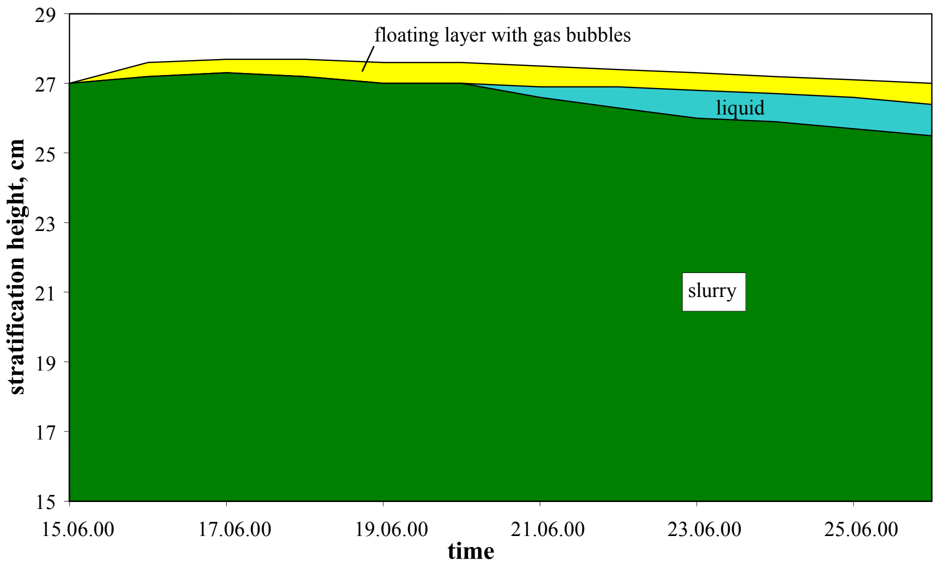
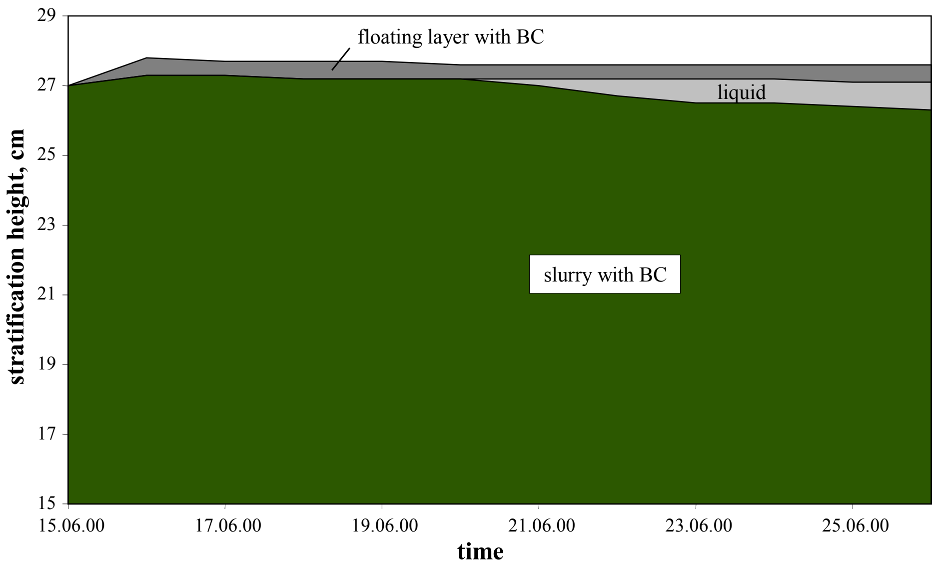


Figure 8: Relative change of the average NH_3 and CH_4 concentrations over cattle slurry with brown coal additives of 3% and 5% (slurry without brown coal = 100%)



a) cattle slurry, control, DM = 8.9 %



b) cattle slurry + 3 % BC, DM = 11.4 %

Figure 9: Course of the sedimentation of cattle slurry without and with brown coal

layers. Odours, however, are kept from emitting by the swimming layer.
In the examined original pig slurry, separation into a foam-/swimming layer began immediately (**figure 10a**). After five days, the liquid became more extensive. After 14 days, the foam-/swimming layer and the liquid accounted for 5% and 47% of the volume respectively. The remaining volume (48%) was taken up by homogeneous slurry.
The pig slurry treated with 3% brown coal immediately formed a compact swimming layer of light slurry particles and coal particles. Only after four days did a dark-coloured liquid develop with homogeneous slurry below. After 14 days, the percentage of the swimming layer, the liquid, and the slurry amounted to 14%, 42%, and 44% respectively.
For a short time, the swimming layer was 6 cm thick, which corresponds to 20% of the total height. After 14 days, it still measured 3.7 cm. The swimming layer largely prevents odour release in a quiescent condition and after homogenizing because it quickly forms again due to the slurry and coal particles floating on the surface.

Summary and Conclusions

In search of alternative coverings for slurry containers for the reduction of odour and noxious gas release, the treatment of cattle and pig slurry with fine-grain brown coal from the Lausitz region was tested in three series of laboratory trials.
Determined dosage amounted to 3 and 5 mass-%. The DM contents of the slurry corresponded to values common in practice.
The addition of brown coal reduces the pH-value of the slurry, but not so far that this reduction would result in greater advantages for the reduction of ammonia emission, which on average amounted to 30%. Together with floating slurry particles, fine-grain brown coal forms dense swimming layers, which lead to a drastic reduction in odour release. In cattle slurry stored under undisturbed conditions, odour emission was reduced by 70% and, after homogenizing, by 90% to 98%. In pig slurry, odour reduction amounted to 92% to 98% both before and after homogenizing.
This is an excellent result, which would need to be confirmed in a large-scale trial.
If pig slurry, whose tendency to form layers is strong at a DM content of 4% common in practice and still significant at a 6% to 8% DM content, is treated with fine-grain brown coal, this opens up the technical possibility of effective solid-liquid separation into an easily conveyable liquid and a compostable, well-transportable solid with high nutrient and humin content. Studies on its long-term effect with regard to humus formation remain to be carried out.
Technical realization on a farm must include the development and testing of a highly efficient mixing system for slurry and fine-grain brown coal as well as the further process-technological development and economic efficiency of slurry treatment with brown coal.

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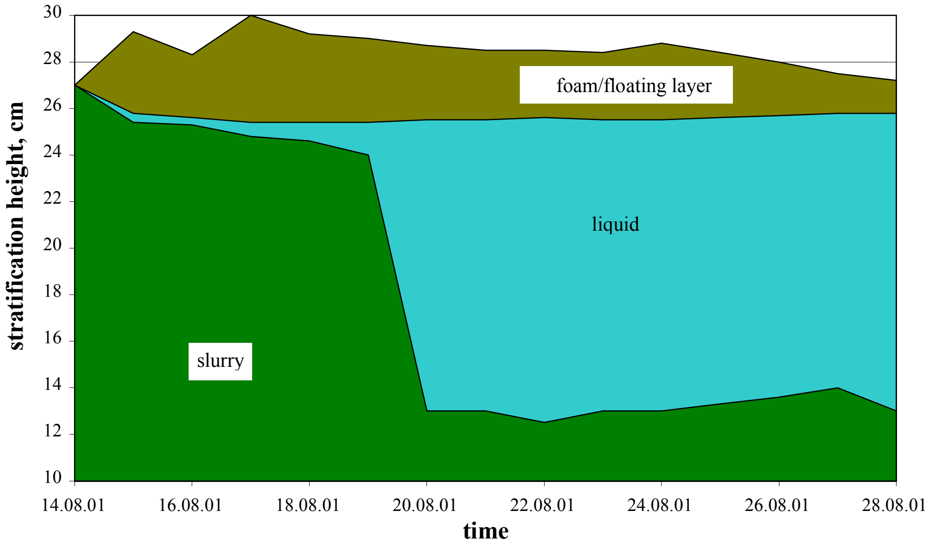
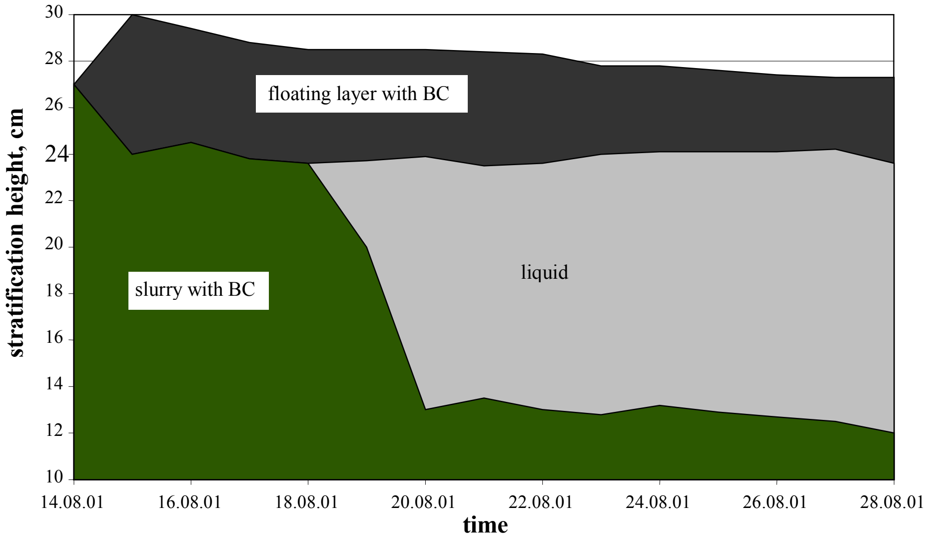


a) pig slurry, control, DM = 7.6 %



b) pig slurry + 3 % BC, DM = 10.0 %

Figure 10: Course of the sedimentation of pig slurry without and with brown coal

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