Biofiltration of exhaust air from animal houses: removal efficiencies and practical experiences

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SUMMARY
Two wood-chip biofilters (capacity and surface area for biofilter #1: 75.000 m^3/hour from poultry manure dryer, 68 m^2; biofilter #2: 100,000 m^3/hour from fattening pig house, 188 m^2; media depth: 25 cm) were monitored during 6 - 10 months. Average ammonia (NH_3) and odour removal efficiencies were 42 - 62%. NH_3 outlet concentrations appeared rather constant and independent of fluctuations in inlet concentrations, so higher inlet concentrations led to higher calculated removal efficiencies. Part of the inlet NH_3 was converted to nitrous oxide (N_2O). The average N_2O production was equal for both biofilters (0.5 g N_2O/m^2/hour) although NH_3 loading rates were very different (30 and 2.4 g NH_3/hour). Average air loading rates were 657 and 341 m^3/m^2/hour, which equal air residence times of 1.4 s and 2.6 s; average pressure drops were 287 and 22 Pa. It is concluded that biofilters have potential for emission reduction at animal houses, but it is recommended that they are professionally designed, operated, and monitored. Especially high pressure drop (clogging/fouling) and homogenous moistening of the biobed need attention.

Keywords: biofilter, biofiltration, NH_3, odor, N_2O, pigs, poultry

INTRODUCTION
Intensive livestock production is connected with a number of environmental effects which include ammonia (NH_3) and odour emissions from animal houses. For mechanically ventilated animal houses, one of the available mitigation techniques is end-of-pipe treatment of the ventilation air. In several European countries (like the Netherlands and Germany) packed-bed air scrubbers (both acid scrubbers and biotrickling filters) are applied on a large scale for this purpose. However, odour removal efficiencies is relatively low for scrubber systems, that were developed mainly for ammonia removal (Melse & Ogink 2002; Melse *et al.* 2012a). Another possible end-of-pipe mitigation technique is the use of a biofilter (or biobed) (Chen & Hoff 2009; Arends *et al.* 2008; Dumont *et al.* 2014a; Chen *et al.* 2009; Nicolai *et al.* 2006).

In contrast to most scrubbers, a biofilter has an organic-based packing material or medium (*e.g.* a mixture of materials such as compost, wood bark, wood chips, peat, perlite, and organic fibres) that is intermittently wetted. Biofilters are used in many industrial sectors (food industry, paint and lithographic industry, waste water treatment etc.), but full-scale applications on farms are scarce. In a biofilter, water is distributed on top of the packing material usually by spraying nozzles. Contaminated air is introduced in a pressure plenum underneath the bed and flows upwards (counter-current) through the bed, resulting in intensive contact between air and moist packing material enhancing mass transfer of pollutants from gas to liquid phase. The air that leaves the biofilter is usually water saturated; any excess water might be discharged from the pressure plenum.

Ammonia removal takes place by nitrifying bacteria that grow on the moist packing material. As in a biotrickling filter, the ammonia dissolves in the water phase and is converted to nitrite (or nitrous acid) and subsequently to nitrate (or nitric acid) by a bacterial

process called "nitrification". If anaerobic zones exist in the biofilter, also denitrification can take place, meaning that part of the nitrite and/or nitrate is converted to nitrogen gas (N_2). In a biofilter nitrous oxide (N_2O) might be produced as by-product from both nitrification and denitrification. N_2O is a strong greenhouse gas with a Global Warming Potential (GWP) of 298, which means that 1 kg of N_2O has the same impact as 298 kg of CO_2 on a time horizon of 100 years (IPCC 2007).

Due to stricter odour emission standards, there is a renewed interest in application of biofilters in livestock production in the Netherlands. Therefore research was initiated for one-year performance trials at two locations, a poultry and a pig farm. The aim of the research was to determine the performance of the biofilter with regard to removal of ammonia and odour, and to assess the possible generation of greenhouse gases, i.e. methane (CH_4) and N_2O.[1].

MATERIALS AND METHODS

Description of biofilter sites

The performance of two biofilters was monitored for a period between 6 and 10 months. One of the biofilters (#1) was located at a laying hen house (30,000 places) where a manure drying unit was operated. In the unit poultry litter and droppings were collected on a permeable cloth and part (about 1/3) of the warm exhaust air of the house was forced through the cloth with fans, resulting in rapid drying of the litter. Next this air was led through the biofilter. The remaining 2/3 of the exhaust air was released untreated. The other biofilter (#2) was used for treatment of exhaust air of a fattening pig house (1,320 places). In Fig. 1 a schematic of the biofilter design is given.

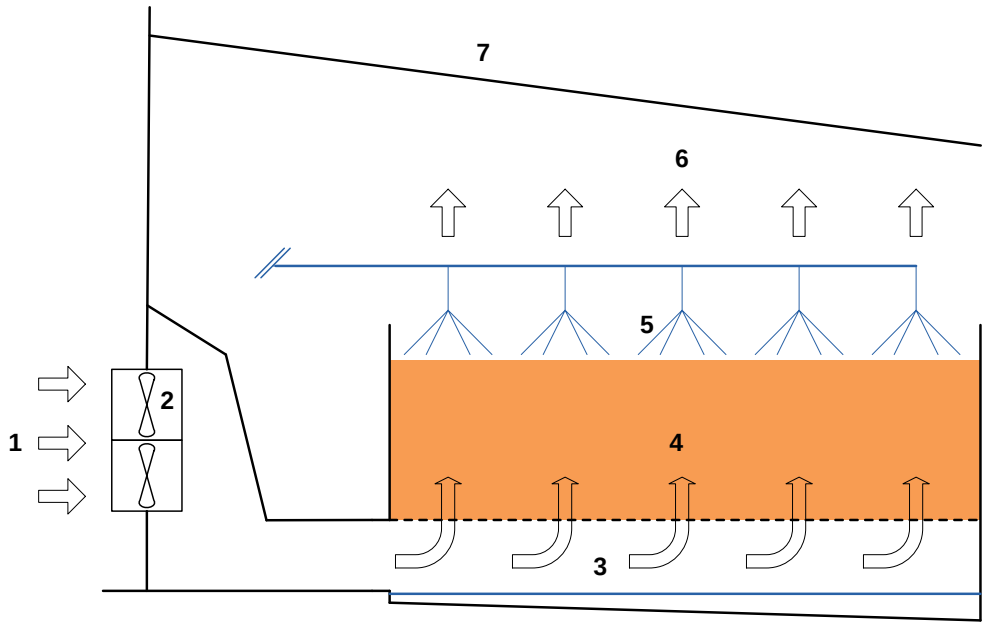

Figure 1. Schematic of biofilter (cross-section); 1: inlet air; 2: fans; 3: pressure plenum; 4: layer of wood chips; 5: sprinklers, 6: outlet air; 7: roof.

Both biobeds consisted of a 25 cm thick layer of organic material, mainly containing of wood chips (size: 2 - 3 cm). For biofilter #1, between the wood chips also finer particles were present; for biofilter #2, the finer fraction was sieved out prior to filling the biofilter. The bulk density of the material was about 600 kg/m^3 at a dry matter content of about 30%. The biobed

[1] This study has been reported in Dutch as Melse & Hol 2012 and Melse *et al.* 2014.

was humidified by spray nozzles placed on the surface of the bed. The spraying system was controlled by a timer and usually operated 3 - 4 minutes per hour. If necessary, the spray time settings were adjusted by the operator after weekly visual inspection of the biobed conditions. Above the biofilter was an inclined roof to diminish weather influences (rainfall and blazing sun light) in order to promote stable humidity conditions in the bed. Underneath the bed was a pressure plenum (height: 50 cm) where the air was introduced. Every two months, both biofilter sites were visited and measurements were carried out. In Table 1, the main characteristics of the biofilters are listed.

Table 1. *Main characteristics of the two biofilters*

Location	Source of air	Maximum ventilation rate (m^3/h)	Surface area (m^2)	Wood chip bulk volume (m^3)	Minimum EBRT[3] (s)
#1	poultry manure dryer for 30,000 laying hens	75,000	68	17	0.8
#2	1,320 fattening pigs, partly slatted floors	100,000	188	47	1.7[4]

[1] Empty Bed Residence Time, calculated as the wood chip bulk volume (m^3) divided by the maximum ventilation rate (m^3/s).

Gas measurements

The ammonia measurements were carried out in duplicate with an impinger method. A small amount of sample air is continuously drawn at a fixed flow rate controlled by a critical orifice (1 l/min) through a pair of acid containing impingers (nitric acid, 0.03 - 0.2 M; 0.5 l each), connected in series. NH_3 is trapped by the acid and accumulates in the bottles during 24 hours. The sampling flow rate and nitric acid concentration are chosen in such a way that the second impinger will not contain more than 5% of the amount of NH_3 trapped in the first impinger to be sure no NH_3 breaks through. All sampling lines were made of Teflon to prevent adsorption of NH_3. Finally, the NH_3 concentration of the air is calculated from the nitrogen content of the acid solution in the bottles, which is determined spectrophotometrically (NNI 1998), the air sampling flow rate and the weight of the impingers before and after the measurement.

For measurement of odour and greenhouse gases (CH_4 and N_2O), air samples were collected in initially evacuated 50 L Nalophane (PET) bags using a lung sampler. Each bag was placed in an airtight container and the inlet of the bag was connected to the air sampling port; next the bag was filled by creating an underpressure in the surrounding airtight container by means of a pump. In this way there is no contact between the air sample and the pump. The sampling time was 2 hours (between 10:00 and 12:00) for the odour sample and 24 hours for the CH_4 and N_2O measurement; the air sampling flow rate was controlled by a critical orifice. Fluctuations in the composition of the air are thus time. The sampling system is equipped with a heating system to prevent condensation in the bag or in the Teflon lines. For odour, an additional filter (pore diameter: 1 - 2 μm) was placed at the inlet of the sampling line to prevent intake of dust that otherwise might contaminate the olfactometer. The CH_4 and N_2O concentration in the bags were determined in duplicate with a gas chromatograph (Carbo Erba Instruments, GC 8000 Top; column: Haysep; detector: ECD/HWD). Odour concentrations were determined in compliance with the European olfactometric standard EN13725 (CEN, 2003) and expressed in European Odour Units per m^3 air (OU_E/m^3); the sensitivity of the odour panel is based on the 20 - 80 ppb n-butanol range.

During the 24-hour measurements, the airflow rate through the biofilters was measured continuously with measuring fans; also the pressure drop over the biobed was measured. Furthermore, the temperature and relative humidity (RH) of the inlet and outlet air of the

biofilter was monitored. The differences between inlet and outlet concentrations were analysed using a one-tailed Student's t-test.

Packing material and water measurements

Samples were taken of the packing material (wood chips) at different spots and depths; these samples were mixed and then analysed for content of dry matter (DM), ash, total-N, NH_3/NH_4^+-N, NO_2^--N, and NO_3^--N. Prior to measurement of pH and EC (electric conductivity), water was added to the sample in a 4 : 1 mass ratio, the mixture was stirred for a moment, left for half an hour and stirred again. The same analyses were done on the percolate water that was collected in the pressure plenum. Furthermore, for biofilter #2 the water use of the sprinkling system and the percolate water production was measured.

RESULTS AND DISCUSSION

Operational parameters

For biofilter #1, the average air loading rate was 657 m^3/m^2/hour (n=4; sd=187), which equals an EBRT of 1.4 s, and resulted in an average pressure drop of 287 Pa (n=5; sd=40). For practical application at animal houses this might be problematic as noramlly the ventilation system will not to be able to yield this kind of pressure. For biofilter #2, the average air loading rate was about two times as low, *viz.* 341 m^3/m^2/hour (n=6; sd=216), which equals an EBRT of 2.6 s, and resulted in an average pressure drop of only 22 Pa (n=5; sd=16). Although the pressure drop is expected to be proportional to the square air flow rate, the higher pressure drop at biofilter #1 will also be caused by accumulation of dust (air from poultry, and especially from a manure dryer, has a much higher dust concentration than air from a pig house) and sieving out the finer fraction from the wood chips. The average inlet temperature was 15 - 22˚C for biofilter #1 and 23 - 25˚C for #2; the outlet air was 1 - 5˚C lower as a result of adiabatic cooling. The average RH of the inlet air was > 85% for biofilter #1 and > 60% for #2; the RH of the outlet air was always > 95%. Despite the high relative humidity of the outlet air, frequently dry spots were observed at biofilter #2 which might have facilitated short-circuiting of the air and decreased removal efficiency. Increased air flow at dry spots might result in even further dry-out of these spots.

Gas measurements

Table 2 and 3 show that the average ammonia and odour removal was 47% and 62% for biofilter #1, and 42% and 52% for biofilter #2. Furthermore, the tables show that the ammonia and odour inlet concentrations for biofilter #1 were higher than for biofilter #2. This can be explained by the fact that the manure drying will results in evaporation of ammonia and odour compounds. Also the removal efficiencies were higher for biofilter #1. The average ammonia loading rate was 30 and 2.4 g NH_3/hour for biofilter #1 and #2, respectively.

For the last three measurements at biofilter #1, operating conditions may have decreased the ammonia removal efficiency. On 01-Nov-10 and 13-Dec-10 the pH was relatively low which might have resulted in a low nitrification rate. Prior to the measurement on 10-Feb-11, the humidification system had been switched off for a week. Possibly this resulted in short-circuiting and local accumulations of nitrogen compounds and subsequent stripping of ammonia from the water phase, resulting in a negative removal efficiency. If this last measurement is left out, the average ammonia removal efficiency would increase to 74% (sd=17; $P < 0.005$).

The other ammonia measurements in Table 2 and 3 show that the highest ammonia inlet concentrations resulted in the highest removal efficiencies. No relation was found between ammonia loading rate and removal efficiency. Instead, it seems that the outlet ammonia

concentration was not influenced by the inlet concentration, which means that a high inlet concentration would result in a high measured removal efficiency, and vice versa. For ammonia removal in a biotrickling filter, which is a similar process, the same phenomenon was found by Melse *et al.* (2012b). These authors showed that the ammonia concentration in the outlet air was in equilibrium with the ammonium concentration in the liquid phase, and could be estimated by taking into ammonium concentration, temperature and pH. The amount of ammonium present in the water phase might function as a buffer and remain constant, independent of fluctuating inlet concentrations. In comparison with Melse *et al.* (2012b), the air outlet temperature of the biofilter was 5 - 10˚C higher. It can be calculated that a temperature increase of 5˚C will result in an increase of the ammonia concentration of about 25%. Furthermore, the pH of the percolate water (Table 7 and 9) was relatively high, whereas for a biotrickling filter a pH range of 6.5 - 7.5 is considered normal (Melse & Ogink 2005); a pH increase of one unit will result in a tenfold equilibrium ammonia concentration. These two parameters could be the reason that for a relatively high ammonia outlet concentration was found for the biofilters, which resulted in lower ammonia removal efficiency than for the reported biotrickling filters.

Although the odour loading rate for biofilter #1 was three times as high as for #2, (1165 vs. 358 OU_E/m^2/hour), the removal efficiency was higher. As for ammonia, also for odour removal the higher inlet concentrations may have played a role.

Table 2. *Performance of biofilter #1: NH_3 and odour*

Date	NH_3-in (ppm)	NH_3-out (ppm)	R.E.[1] (%)	Odour-in (OU_E/m^3)	Odour-out (OU_E/m^3)	R.E.[1] (%)
23-Aug-10	22	3.5	84	7,899	2,415	69
27-Sept-10	46	3.4	93	6,393	517	92
01-Nov-10	52	21	60	4,261	1,995	53
13-Dec-10	180	74	59	7,326	3,983	46
10-Feb-11	31	49	-60	6,029	3,023	50
Average:	*66*	*30*	*47*[2]	*6,381*	*2,386*	*62*[3]
	(sd=65)	*(sd=31)*	*(sd=62)*	*(sd=1398)*	*(sd=1285)*	*(sd=19)*

[1] R.E. = removal efficiency; [2] $P < 0.1$; [3] $P < 0.001$.

Table 3. *Performance of biofilter #2: NH_3 and odour*

Date	NH_3-in (ppm)	NH_3-out (ppm)	R.E.[1] (%)	Odour-in (OU_E/m^3)	Odour-out (OU_E/m^3)	R.E.[1] (%)
14-May-13	16	4.4	72	2452	801	67
08-Jul-13	6.7	3.0	55	2640	1420	46
19-Aug-13	7.2	5.8	19	1074	502	53
15-Oct-13	7.1	6.3	10	3022	2704	11
09-Dec-13	7.3	6.2	16	5452	1850	66
25- Mar-14	18	3.5	81	8032	2308	71
Average:	*10*	*4.9*	*42*[2]	*3779*	*1597*	*52*[3]
	(sd=5.1)	*(sd=1.4)*	*(sd=31)*	*(sd=2523)*	*(sd=855)*	*(sd=26)*

[1] R.E. = removal efficiency; [2] $P < 0.025$; [3] $P < 0.005$.

In Table 4 and 5, the inlet and outlet concentrations of CH_4 and N_2O are given for both biofilters. For CH_4, no significant difference was found between inlet and outlet concentrations, although concentrations were much higher at biofilter #2. This can be explained by emissions from the anaerobic digestion of the pig manure that is stored underneath the pens. It is not clear why the inlet concentrations for the last three measurements at biofilter #2 were much higher than earlier.

For N_2O, however, a significant increase was found: for both biofilters the outlet was about two times as high as the inlet concentration. Some production of N_2O is normally found at biofilters or biotrickling filter treating ammonia containing air (Trimborn *et al.* 2003;

Trimborn 2006; Dumont *et al.* 2014b; Maia *et al.* 2012; Yang *et al.* 2014a, 2014b; Melse & Mosquera 2014), although reported ranges vary. Tables 4 and 5 show that the production of N_2O-N, calculated as a fraction of the inlet NH_3-N, was 2.0% and 21% for biofilter #1 and #2, respectively. However, the N_2O production rate expressed per m^2 of was similar, in both cases 0.5 g N_2O/m^2/hour. Apparently the N_2O production rate per m^2 is not directly related to the NH_3 loading rate, which largely differ between the biofilters. Possibly the production of N_2O can be reduced by reducing the dry matter content of the wood chips by decreasing the amount of spray water (Yang *et al.* 2014a; Maia *et al.* 2012); also the pH value may influence the N_2O production rate (Yang *et al.* 2014b).

Table 4. *Performance of biofilter #1: CH_4 and N_2O*

Date	CH_4-in (ppm)	CH_4-out (ppm)	N_2O-in (ppm)	N_2O-out (ppm)	N_2O-N production (% of NH_3-N in)
23-Aug-10	2.7	2.5	0.33	0.88	5.1
27-Sept-10	3.6	2.8	0.45	0.87	1.8
01-Nov-10	3.4	3.1	0.39	0.58	0.7
13-Dec-10	3.3	3.4	0.62	1.62	1.1
10-Feb-11	3.4	3.5	0.46	0.63	1.1
Average:	*3.2*[1] *(sd=0.32)*	*3.1*[1] *(sd=0.42)*	*0.45*[2] *(sd=0.11)*	*0.92*[2] *(sd=0.42)*	*2.0 (sd=1.8)*

[1] $P > 0.15$; [2] $P < 0.025$.

Table 5. *Performance of biofilter #2: CH_4 and N_2O*

Date	CH_4-in (ppm)	CH_4-out (ppm)	N_2O-in (ppm)	N_2O-out (ppm)	N_2O-N production (% of NH_3-N in)
14-May-13	15	8.8	1.4	1.8	4.6
08-Jul-13	2.6	2.1	0.46	0.53	18
19-Aug-13	7.6	8.9	0.55	0.87	10
15-Oct-13	50	113	0.50	2.5	69
09-Dec-13	58	48	1.2	2.2	21
25- Mar-14	51	51	1.0	1.5	6.1
Average:	*31*[1] *(sd=25)*	*38*[1] *(sd=41)*	*0.84*[2] *(sd=0.39)*	*1.6*[2] *(sd=0.65)*	*21 (sd=24)*

[1] $P > 0.25$; [2] $P < 0.05$.

Packing material and water measurements

In Table 6 and 7 the analysis results of packing material ('wood chips') and percolate water are given for biofilter #1. Both tables show that the amount of ammonium and nitrite in the biofilter increased over time. Hardly any nitrite was found which indicates that full nitrification took place. As mentioned before, possibly the accumulation of ammonium eventually resulted in stripping of ammonia what could explain the negative ammonia removal efficiency for the last measurement. As mentioned before, the pH of measurement 3 and 4 is quite low and might have resulted in a lower removal efficiency, as a pH below 6 is considered as hampering the nitrification process.

Table 6. *Analysis of packing material for biofilter #1*[1]

Component	23-Aug-10	27-Sep-10	01-Nov-10	13-Dec-10	10-Feb-11
NH_4-N (g/kg)	0.32	0.027	0.60	0.83	1.77
NO_2-N (g/kg)	< 0.010	< 0.010	< 0.010	< 0.010	< 0.010
NO_3-N (g/kg)	0.33	< 0.010	0.60	0.70	1.86
Total-N (g/kg)	3.08	2.53	3.81	4.24	6.47
DM (g/kg)	289	275	269	256	281
Ash (g/kg)	24	30	25	26	31
pH (-)	7.0	6.2	5.2	5.5	6.9

[1] The presented values are the averages of 3 mixed samples.

Table 7. *Analysis of percolate water for biofilter #1*[1]

Component	23-Aug-10	27-Sep-10	01-Nov-10	13-Dec-10	10-Feb-11
NH_4-N (g/kg)	0.009	0.034	0.027	0.151	n.a.
NO_2-N (g/kg)	< 0.010	< 0.010	< 0.010	0.017	n.a.
NO_3-N (g/kg)	0.011	0.023	0.017	0.071	n.a.
Total-N (g/kg)	0.015	0.060	0.056	0.249	n.a.
DM (g/kg)	< 0.2	0.2	< 0.2	0.26	n.a.
Ash (g/kg)	< 0.2	< 0.2	< 0.2	0.15	n.a.
pH (-)	7.5	7.4	8.3	7.8	n.a.
EC (mS/cm)	0.21	0.57	0.35	1.6	n.a.

[1] Some rain water might have entered the pressure plenum and have diluted the percolate samples, as the pressure chamber appeared not to water tight.

In Table 8 and 9 the analysis results of packing material and percolate water are given for biofilter #2. Table 9 shows that the ammonium concentration (and EC) were high at the first two measurements; this might have been caused by low water spraying and discharge amounts during or before this period. However, in contrast to biofilter #1, no accumulation of nitrogen compounds in the packing material (Table 8) took place over time, probably because the NH_3 loading rate was much lower than for biofilter #1. Furthermore, Table 10 shows the amount of spraying water (water use) and discharge water. On average 65% of the spraying water was discharged as percolate, thus 35% was evaporated.

From Table 8 it can be calculated that the amount of water in the biobed, that might buffer fluctuations in the inlet NH_3 concentration, is about 100 kg/m^2. If we assume, for example, that the inlet NH_3 concentration would double from 10 ppm (7 mg/m^3) to 20 ppm (14 mg/m^3) during one hour, at an airflow of 341 m^3/m^2/hour, the loading rate would increase with 2.0 g NH_3-N/m^2/hour. If this amount dissolves in the water buffer this would mean an increase of 2.0 mg NH_4^+-N/l in the liquid phase. Table 9 shows that the actual ammonium concentrations in the percolate are 100 to 5,000 times higher than that, so it is expected that this buffering will actually take place.

Table 8. *Analysis of packing material for biofilter #2*[1]

Component	14-May-13	08-Jul-13	19-Aug-13	15-Oct-13	09-Dec-13	25-Mar-14
NH_4-N (g/kg)	0.03	2.88	2.24	2.50	2.100	0.45
NO_2-N (g/kg)	<0.010	0.195	0.094	<0.010	<0.010	<0.010
NO_3-N (g/kg)	0.042	0.255	1.185	0.179	0.421	0.345
Total-N (g/kg)	1.58	6.49	5.92	3.75	5.14	3.36
DM (g/kg)	285	405	389	287	289	242
Ash (g/kg)	7.4	11.6	13.8	9.5	9.6	8.8
pH (-)	6.35	6.7	7.0	6.7	7.6	6.9
EC (mS/cm)	n.a.	1.4	3.8	2.3	3.5	1.0

[1] The presented values are the averages of 4 mixed samples.

Table 9 *Analysis of percolate water for biofilter #2*

Component	14-May-13	08-Jul-13	19-Aug-13	15-Oct-13	09-Dec-13	25-Mar-14
NH_4-N (g/kg)	8.69	9.58	0.33	1.72	1.94	2.33
NO_2-N (g/kg)	0.116	0.468	< 0.010	0.25	< 0.010	0.024
NO_3-N (g/kg)	4.85	4.53	< 0.010	0.51	0.89	1.25
Total-N (g/kg)	13.5	17.3	0.38	2.68	2.95	3.92
DM (g/kg)	46.6	n.a.	1.5	6.6	7.2	9.9
Ash (g/kg)	2.7	n.a.	0.7	1.0	0.9	1.0
pH (-)	8.3	7.6	7.3	8.2	8.7	8.0
EC (mS/cm)	58	70	3.3	14	15	19

Table 10 *Water use and percolate water production for #2*

Date	Spraying water use ($l/m^2/day$)	Percolate production ($l/m^2/day$)	Percolate production (% of water use)
14-May-13	21	n.a.	n.a.
08-Jul-13	32	12	39
19-Aug-13	35	n.a.	n.a.
15-Oct-13	20	17	87
09-Dec-13	n.a.	n.a.	70
25- Mar-14	21	15	n.a.
Average:	*23*	*15*	*65*

CONCLUSION

The two biofilters that were monitored were able to achieve a removal of ammonia and odour compounds in a range between 42 - 62%. It seems that outlet ammonia concentration is rather constant and not influenced by fluctuating air inlet concentrations. This means that higher ammonia inlet concentrations will lead to higher removal efficiencies, and vice versa. Furthermore, it was found that part of the NH_3-N was converted to N_2O-N. The average N_2O production per m^2 of biobed seems to be equal for both biofilters, although ammonia loading rates were very different.

It is concluded that biofilters have potential for emission reduction of ammonia and odour at animal houses, but it is recommended that they are professionally designed, operated, and monitored. Especially high pressure drop (clogging/fouling) and homogenous moistening of the biobed need attention. Furthermore, packing material lifetime needs to be determined as removal efficiencies might deteriorate over time.

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