

# PULP MILL BIO-SLUDGE - SLUDGE PROPERTIES AND RECYCLING

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SUMMARY: Sustainable management of bio-sludge from the pulp and paper industry and the sustainable supply of protein to producers of animal fodder is desirable. Using larvae's of black soldier flies for reduction of bio-sludge and production of protein for animal fodder is one possible recycling method that could meet these needs. The purpose with this paper is to assess the attractiveness of this solution compared to recycling other useful substrates in the sludge from a circular economy perspective. Furthermore, the suitability of the sludge as feed for the larvae's from a protein to fat quota perspective and the suitability of the larvae's as raw material for animal fodder from a contamination perspective are assessed. The chemical content of bio-sludge from ten Swedish mills have been analysed. The main result is that recycling of bio-sludge for its protein content is for seven of the mills the most economically attractive option. Bio-sludge can be a feed suitable for larvae's of black soldier flies and the larvae's suitable for animal fodder although the heavy metals could in a worst case scenario exceed the limit values for animal fodder.

#### **1. INTRODUCTION**

The pulp and paper industry generates large amounts of bio-sludge at waste water treatment plants. It is wet and has no obvious economic value, so present industrial focus is not on recycling methods but on disposal. Common disposal methods are incineration and composting (Sivard et al., 2013 Figure 10). In Sweden and Norway about half of this sludge is incinerated and the other half is composted. In Finland almost all biosludges is incinerated whereas landfilling of the sludge is common in Chile where half the sludge is landfilled and the other half is incinerated (Sivard et al., 2013 Figure 10a-d). Biomass waste in general could be recycled for its energy content or as a resource for chemical products or solid materials. New solutions should apply to circular economy thinking.

Norgren et. al. (2015) performed a literature study and compiled a list of 23 novel methods for recycling bio-sludge. They were assessed for their technical maturity and degree of circularity (circular economy-thinking). They concluded that the production of single cell protein (SCP) and a lead absorber, from these three perspectives, were the two more suitable methods. An alternative to the SCP method could be larvae's of black soldier flies (BSF) which could constitute a source of protein for animal feed besides degradation of organic waste (Diener et al., 2011, pp. 357, 358).

The focus of this paper is to assess the potential of bio-sludge as feed for BSF larvae from the perspective of suitability and economic drivers. This paper aims to set the circular economy framework for recycling of bio-sludge from the pulp and paper industry. Sludge from ten Swedish mills were studied and their content of useful substrates and detrimental content like heavy metals were analysed.

#### **2. METHOD**

Ten Swedish mills representing different combinations of raw material, type of process and bleaching. Fresh bio-sludge samples were taken on one single occasion between January and April 2016 and sent to Mid Sweden University, Sundsvall for homogenizing. Four to eight litres of samples were taken from the mills and homogenized by the use of a trowel for three minutes. Larger pieces were divided by hand. Forty litres were taken from one of the mills and homogenized for 15 minutes by the use of a drilling machine and a concrete mixing paddle (Figure 1). The samples were stored in a refrigerator directly after homogenizing for a maximum of five days before sending to the laboratories.



Figure 1 Tools for homogenizing



Figure 2 Containers for sludge samples

The samples for metal analysis were sent in 50 ml plastic tubes (Figure 2) to Ragn-Sells AB laboratory in Högbytorp, Bro, Sweden. The samples for the other analysis were sent in 2.2 litre plastic buckets to Eurofins laboratory in Lidköping, Sweden.

The content of substrates and metals have been analysed by Eurofins Environment Testing Sweden AB, Eurofins Food & Feed Testing Sweden, MoRe Research Örnsköldsvik AB and Ragn-Sells laboratory in Bro. In total 10 parameters were analysed. The parameters and the corresponding standard methods used are listed in Table 1.

Properties	Method	Uncertainty	Laboratory	Accredited	
DS (%)	SS-EN 12880:2000	10 %	В	Y	
Lignin (% DS)	Tappi T 222	2.5 %	С		
Fat (g/100g)	2009/152/EU mod.	10 %	А	Y	
N <sub>tot</sub> (mg/kg)	SS-EN 13342	10 %	А	Y	
Crude protein (%)	Calculated from N (Nx6.25)	20 %	В	Y	
P (mg/kg DS)	NMKL No 161 1998 mod./	15 %	В		
	ICP-AES				
K (mg/kg DS)	ICP-MS. Standard SS		D		
	028150				
Hg (mg/kg DS)	SS-EN 16277:2012	30 %	В		
Cd (mg/kg DS)	ICP-MS. Standard SS		D		
	028150				
Pb (mg/kg DS)	ICP-MS. Standard SS		D		
	028150				

Table 1 Standard methods for analysis

A=Eurofins Food & Feed Testing Sweden, B=Eurofins Environment Testing Sweden, C=MoRe Reseach Örnsköldsvik AB, Sweden, D=Ragn-Sells AB. DS = Dry substance,  $N_{tot} = Total$  nitrogen

To screen for economic drivers a rough estimate of potential economic values of different possible circular economy routes were assessed. Four bio-chemicals in the bio-sludge were assessed together with the value if used as a biofuel (thus destroying the potential as biochemical raw material). A 100 % extraction rate has been assumed for each substrate. The results shall be seen as a theoretical maximum.

For assessing the suitability of the bio-sludge as a feed for BSF-larvae a comparison with other tested feeds has been made by using data found in the literature. In addition, the impact of heavy metals on the suitability of prepupae as animal feed has been assessed by a comparison with current limit values for animal feed. All metals in the sludge are assumed to end up in the prepupae.

# **3. RESULTS**

### 3.1. Sludge properties

In Table 2 the complete results of the chemical analysis are displayed. In figure 3 the content of nutrients and useful substrates are displayed as normalized values, setting the highest measured value to 100. Table 3 compare found data with data found by Norgren et. al. (2015). For mill J, lignin, phosphorus and mercury are not analyzed.

Table 2 Results of chemical analysis.

Properties	Mill A	Mill	Mill	Mill D	Mill	Mill	Mill	Mill	Mill	Mill
-		В	С		Е	F	G	Η	Ι	J
DS (%)	18	19	10	20	30	16	21	19	26	11
N <sub>Tot</sub>	13,000	16,000	7,900	11,000	7,800	2,900	6,900	8,300	5,500	9500
(mg/kg)										
Crude	6.1	8.3	4.3	4.5	4.4	1.5	3.8	4.2	3.2	5,1
protein										
(%)	~ <b>-</b>		• •	~ <b>-</b>		0.0	1.0			~ -
Fat (%)	0.7	0.8	0.3	0.5	3.3	0.8	1.8	1.4	1.3	0,7
Lignin	38	20	32	17	40	23	40	37	30	N.D.
(% DS)										
P (mg/kg	8,600	19,000	15,000	21,000	3,000	1,700	6,500	8,800	4,800	N.D.
DS)										
Hg	0.042	0.032	0.07	0.14	0.024	0.034	0.022	0.022	0.022.	N.D.
(mg/kg										
DS)	1.2	0 1	1.0	0.2	2.5	1.2	1 /	1.2	0.0	27
Ca (mg/kg	1.3	8.1	1.8	0.5	2.5	1.3	1.4	1.3	0.8	2,7
(IIIg/kg DS)										
K	2 700	3 900	3 300	2 200	670	590	1 000	1 700	245	4 600
(mg/kg	2,700	5,900	5,500	2,200	070	590	1,000	1,700	210	1,000
DS)										
Pb	5.4	4.9	90	15	2.9	11	7.7	6.6	8.1	3,2
(mg/kg										
DS)										
100										



Figure 3 Sludge content of resources. Standardized values.

Properties	Results from literatu (converted)	This study		
	Min	Max	Min	Max
Crude protein (%)	3.6	13	1.5	8.3
Fat (%)	6.3	6.3	0.2	3.3
Lignin (% DS)	9	13	17	40

Table 3 Consistency of the results and literature data from Norgren et. al. (2015)

# 3.2 Economic drivers for circular economy solutions

The potential economic value of the substrates derived from the sludge has been calculated. The input data used for this are displayed in Table 3.

Properties	Value	Range	References
Economic value of phosphorus (€/ton)	1,961		(Rosenqvist, 2016, p. 18)
Economic value nitrogen (€/ton)	1,089		(Rosenqvist, 2016, p. 18)
Economic value potassium (€/ton)	763		(Rosenqvist, 2016, p. 18)
Reported market value of protein	534	305-763	(Graham, 2016. Personal
(€/ton wet)			Comunication)
Market value of fat (€/ton)	381	327-436	(Fahlström, 2016. Personal
			Comunication)
Density fat (kg/l)	<1		Fat floats on water and is
			therefore lighter than water.
Calorimetric heating value of sludge	20.0	18-22.5	(Norgren et al., 2015, p. 4)
(MJ/kg DS, ash free)			
Ash content of sludge (% DS)	27.0		(Norgren et al., 2015, p. 4)
Calorimetric heating value of recycled	22.0	20-24	(Strömberg and Herstad
wood (MJ/kg DS, ash free)			Svärd, 2012, p. 68)
Dry substance of recycled wood (%)	76.7	-	(Strömberg and Herstad
		15.8/+12.2	Svärd, 2012, p. 68)
Ash content of recycled wood (% DS)	5.8	-2.6/+10.1	(Strömberg and Herstad
			Svärd, 2012, p. 68), (Sveriges
			Riksbank, 2016)
Evaporation energy water (MJ/kg)	2.43		
Lower heating value of recycled wood	4.3		Calculated from the data
(MWh/ton) (Calculated)			above
Market value recycled wood (€/MWh)	11.0		(Harrysson, 2016 Table 1)
Market value lignin (EUR/ton)	225	200-250	(Backlund and Nordström,
			2014 Table 9), (Sveriges
			Riksbank, 2016)
Market value activated carbon (€/ton)	1,946	560-3,300	(Backlund and Nordström,
			2014 Table 10)
Rate of extraction (%)	100		

Table 4 Input data for economic calculations

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In Figure 4 the theoretical maximum economic value of six substrates potentially derived from the sludge is displayed together with the uncertainty. The uncertainty is influenced by the method used

for laboratory analysis and the range of economic value. These are accounted for in Table 1, column "Uncertainty" and in Table 3, column "Range". For the protein, lignin and fat, it is the economic value that represents the greatest uncertainty but for the rest of the parameters it is the uncertainty of the analysis that is displayed. In addition to the substrates in the figure, the total carbon was analyzed but only for one mill (J), and is therefore not displayed in the Figure. Of the dry substance the total carbon was 46 % which corresponds to 900 EUR/ton DS if it could be converted to activated carbon. Negative values for sludge as biofuel are obtained for two of the mills (C, J).

Please note the significant differences among the mills but also among the different parameters. Also note that there may be a major impact through the combination of the following five aspects, (I) uncertainty of the laboratory analysis, (II) the price range, (III) the quantity of these resources that actually can be extracted, (IV) the ash content and (V) the dry substance. An example of the current cost for composting of the sludge is 44  $\notin$ /ton wet sludge (Öhman and Fougner, 2014, p. vii) which corresponds to 147 - 400 EUR ton DS.



Figure 4 Potential economic value as driver for circular economy solutions (EUR/ ton DS of sludge)

# 3.3 Comparison with other BSF feed and animal feed regulation

Oonincx et al. (2015) tested four different feeds derived from food waste for breeding BSF larvae. They analyzed the content of crude protein, phosphorous and total fatty acids (TFA) on a dry matter basis (2015, p. 5), the results are displayed in Figure 5. Their conclusion were that (2015 Figure 1) a high protein and high fat (HPHF) diet is better for the larvae than a low protein, low fat diet. Therefore HPHF is used for comparison with bio-sludge. Please note that TFA is not directly comparable to fat according to 2009/152/EU mod. The TFA method probably results in a lower value then what the EU-method would for the same sample (Åkerlind, 2009, p. 3). For mill I the phosphorus bar is not visible because of the very low value.



Figure 5 Suitability of sludge as BSF feed

The dry substance of prepupae of BSF contains about 43-44 % of protein (Diener et al., 2011, p. 358), (Čičková et al., 2015, p. 74). This has been used for calculating the amount of prepupae from the produced amount of protein. The worst case scenario for the concentration of Hg, Cd and Pb in the prepupae have been calculated by dividing the total Hg, Cd and Pb content in the sludge with the amount of prepupae. The results are compared to the limit values of the European Union for animal feed (Nordin and Wejdemar, 2014, p. 6), (European Commission, 2011, p. 9). The results are shown in Figure 6.



Figure 6 Theoretical maximum content of metals in prepupae

## 4. DISCUSSION

#### 4.1. Bio-sludge properties

In Figure 3 the varying concentration of the useful substrates are displayed. For all the parameters the spread of the results are significant. The parameter with the smallest gap between maximum and minimum is lignin, which has a max/min-quota of 2. The most variable parameter is phosphorus which has a quota of 12.

A comparison with literature data (Table 3) found by Norgren et. al. (2015) show that the protein content of the mills in this study is consistent with previous study. On the other hand the carbohydrate and lignin content are higher but fat content is lower.

#### 4.2. Economic drivers for circular economy solutions

Figure 4 shows that for seven of the mills it is the protein that is the most economically valuable substrate, five of these seven have a significant margin to the second most valuable substrate. But for four out of the nine mills the lignin may be as economically valuable as the protein. This is mainly caused by the uncertainty of the market value of protein. But please note that all the five parameters of uncertainty mentioned in section 3.2 contribute to this.

The economic drivers of the other substrates are significantly lower than protein and lignin except for mills B and D where phosphorus may replace lignin as the second most economically valuable resource. Furthermore the potential value of the activated carbon for mill J was four times higher than for the protein. However, according to Norgren et.al. (2015 Tables 2, 3) active char production by pyrolysis was considered as a non-mature technology. If technologies for production of activated carbon from waste water sludge mature, it may be a viable option.

#### 4.3 Sludge suitablility as BSF feed

In Figure 5 the bio-sludge's protein and fat content are compared to the content in a HPHF feed. For none of the mills do both protein and fat content meet or exceed the levels of the HPHF feed. If we on the other hand look at the protein/fat-quota we see that bio-sludge from mills E, G, H and I are closest to the HPHF feed and therefore may be suitable as BSF feed.

#### 4.4 Prepupae suitability as animal feed

In a worst case scenario, all the metals from the sludge end up in the prepupae that would be used as animal feed and as a result the limit values for animal feed are exceeded (Figure 6). Cadmium and lead exceed their limit values for seven and five of the ten mills respectively, in contrast to mercury which only exceeds its limit value for two of the mills.

#### **5. CONCLUSIONS**

There are considerable differences in concentration of useful substrates in bio-sludge from the mills in this study. Mills that are developing and implementing methods to recycle resources from biosludge should consider these differences and the potential benefits of economy of scale, efficiency, etc. by cooperating with other mills.

The economic driver often sets protein as the most valuable substrate but lignin can for some of the mills be more valuable. On the other hand depending on the technology applied the rate of extraction of recyclables can change and as a result, also the most preferable recycling option. The market value of the product also has the potential to change the most valuable recycling option. In the longer term production of activated carbon may be an even more valuable option than protein.

Assessing the suitability of bio-sludge as BSF feed by comparing the protein to fat quota shows that bio-sludges from mill E, G, I and H are more similar to a proven suitable HPHF feed than the other sludges compared. The fate of the metals Hg, Cd and Pb in the production process might be a problem and the limit values for animal feed may in a worst case scenario be exceeded.

#### **6. FURTHER WORK**

Since the rate of extraction has been shown to be an important parameter that can change the order of the most preferable recycling options it should be assessed for the top alternatives, protein and lignin. In addition the production of activated carbon has shown to be potentially very valuable and should be further studied. Furthermore, improved dewatering should be included in the assessment because it could change the order of the most economic valuable resources.

The sludges from mills E, G, I and H have been pointed out as potential suitable feeds for BSF larvae but verifying experiments at bench scale with larvae have not yet been performed. Such trials to verify larvae growth and protein extraction rate should be performed. In addition the optimal quota of protein and fat, the impact of potassium additive should be studied. Furthermore, to minimize the risk of high levels of heavy metals in animal feed the fate of these heavy metals should also be studied.

Understanding why bio-sludge quality differs among the mills is important. This should be examined because it may reveal methods to alter the upstream process so the recycling of biosludge could be further improved. For instance we see in (Figure 3) that two mills (J, E) of the ten have the highest concentration in four (total nitrogen, protein, fat, lignin) of the six measured parameters. What is the explanation for that? The mills in this study constitute different combinations of raw material (tree species), type of process, chemicals used and configuration of water treatment plant etc. The impact on sludge quality of these parameters should be further studied. It is reasonable to believe that sludge quality over timer can vary significantly and this should be further studied.

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