LCA OF WASTE MANAGEMENT SYSTEMS

Comparison of different treatment options for palm oil production waste on a life cycle basis

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Abstract

Background, aim, and scope Globally, 45 million metric tonnes of palm oil has been produced in 2009. The production of 1 t crude palm oil requires 5 t of fresh fruit bunches (FFB). On average, processing of 1 t FFB in palm oil mills generates 230 kg empty fruit bunches (EFB) and 650 kg palm oil mill effluent (POME) as residues. These residues cause considerable environmental burdens, particularly greenhouse gas emissions. In order to reduce those emissions, four waste management options are compared in the present study using 1,000 kg of FFB as functional unit. Methods A detailed life cycle model has been used to calculate the environmental impacts of POME and EFB treatment. The options under investigation are: (1) dumping EFB and storing POME and ponds, (2) returning EFB to the plantation and POME as before, (3) using EFB and POME for co-composting and returning the produced compost to the plantation, (4) generating biogas from POME and thereafter as in (3). The CML 2001 method included in the GABI 4.3 software package has been used for the impact calculations. Sensitivity analysis has been carried out in order to estimate the influence of good and poor management practice on the environmental performance.

Results and discussion The main contributor to the GWP is methane from POME and EFB dumping. The GWP of palm oil mill waste treatment can be reduced from 245 kg CO_{2eq} per ton FFB to up to 5 kg CO_{2eq} per ton FFB due to reduced methane emissions and nutrient recycling. Co-

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composting of POME and EFB leads to considerable nutrient recovery, in addition to GWP reduction. Thus, the composting process reduces not only environmental burdens; it also leads to net environmental benefit regarding most environmental impact categories, e.g., acidification potential, eutrophication potential, ozone layer depletion potential, etc. due to the avoided emissions from inorganic fertilizer production. The recovery of nutrients in EFB can be achieved by solely returning it to the plantation, but only the combined treatment of EFB and POME allows nutrient recovery from POME while methane emissions from pond systems are avoided simultaneously. The fermentation of POME to produce biogas reduces environmental burdens when operating under best practice conditions. However, fugitive biogas emissions of more than 2% reverse that beneficial effect.

Conclusion and recommendation A life-cycle-based comparison of conventional and advanced treatment systems for EFB and POME can support decision makers regarding waste treatment options and provide information on technology risks involved. The results of this study may be used as basic calculation data for clean development mechanism for palm oil mills. LCA is shown to be a powerful tool to estimate and compare environmental impacts of different options. Unfortunately, it is rarely used in the palm oil industry in order to improve or optimize palm oil production systems. This study has shown that nutrient recovery from POME and EFB offers considerable environmental and economic benefits to palm oil production systems. However, using EFB for energy production, as it is discussed and realized by some palm oil mills, prohibits environmental beneficial POME utilization. Best waste management practice reduces emissions at palm oil mills and consequently the carbon footprint of palm oil products. Co-composting of EFB and POME, with or without prefermentation of POME in a biogas plant, is a profitable way to use the nutrients from both POME and EFB.

Keywords Compost · Empty fruit bunches (EFB) · Greenhouse gas (GHG) · Life cycle assessment (LCA) · Methane · Nutrient recycling · Palm oil mill effluent (POME)

1 Introduction

Indonesia has recently become the world's largest palm oil producer. It has produced approximately 21 million tonnes in 2009 (USDA 2010). Increasing global demand for palm oilpartly driven by subsidies for biodiesel-is mainly covered by increasing production in Indonesia. During the period from 2000 to 2009, the mature palm area in Indonesia has grown at an average annual rate of 10% or 250,000 ha, while palm oil production has increased by 17.4% or 1.25 million tons per annum (USDA 2009). This has put Indonesia among the top three largest emitters of GHG in the world due to land use change and deforestation (PEACE 2007). Therefore, sustainable management of palm oil plantations is paramount not only for Indonesia. Sustainability of palm oil production systems is determined by land use or land use change, plantation management, and oil mill processing. One of the most significant factors of sustainability is the nutrient management on palm oil plantations (Chan 2005). Whereas several publications are available about palm oil and palm oil residues as renewable energy source (Tamunaidu and Bhatia 2007; ICTSD 2008; Wicke et al. 2008; Joelianingsih et al. 2008; Sumathi et al. 2008; Chew and Bhatia 2008; Tan et al. 2009), some about life cycle assessment (LCA) of land use and plantation management (Hansen 2007; Schmidt 2007; Schmidt et al. 2009; Rettenmaier et al. 2007; Yusoff and Hansen 2007), only a few are known about LCA of palm oil mill waste and waste water treatment process and utilization (Subramaniam et al. 2008; Yusoff and Hansen 2007).

The production of 1 tonne crude palm oil requires 5 t of fresh fruit bunches (FFB), and its processing palm oil mills generates on average 1,150 kg empty fruit bunches (EFB) and 3,250 kg palm oil mill effluent (POME). In addition, mesocarp fiber and shells are present/produced, but those are used to generate heat and power on site. The treatment and utilization of EFB and POME are substantial parts of the palm oil production system. Both have high contents of nutrient and organic matter and are suitable as fertilizer and soil conditioner for crop production. Fresh EFB can be used as mulch in order to recycle nutrients and organic matter, but it has some disadvantages like temporary immobilization of nutrients, hampering fruit harvesting; harboring of rats and snakes; risk of fire; breeding site for rhinoceros beetles (Oryctes rhinoceros), one of the most serious oil palm pests; difficulties to distribute during rainy season; and high weight and volume in

relation to nutrient content (Sunitha and Varghese 1999). Land application of EFB and/or compost in plantation has an effect on several soil physical and chemical properties: increase of soil organic matter content, infiltration, aeration, soil fauna microactivity and cation exchange capacity, improvement of soil structure and soil water retention, root density and growth, soil temperature fluctuation, and support of weed suppression. These effects result in higher yields compared to untreated plantations, between 2% and 75% with a mean of 13% and 23%, respectively (Goh et al. 1994; Gurmit et al. 1990; cited in Chee and Chiu 1999). Some palm oil mills return EFB as mulch material in plantation; but still a considerable proportion dumps the EFB as waste material. Leakage of nutrients and heavy metals from EFB dumping sites has severe environmental effects, such as eutrophication and an increase of toxicity in the soil. However, no confident data about environment burdens caused by EFB dumping sites are reported (Reijnders and Huijbregts 2008).

Background Land application of POME needs high investment and maintenance cost if it is done in a sustainable way. Sustainability is given when the application rate is limited by the nutrient demand of the oil palm (including losses by surface runoff, leaching, and evaporation), and an accumulation of heavy metals by using the same application area over a long time period can be avoided. A widespread system in palm oil mills for POME is still the treatment in pond systems with land application or discharge of the outlet. But the high methane emissions from pond systems, up to 8 m³/t fresh fruit bunches (FFB; Table 1), are a serious problem for the atmosphere, and discharge of POME to surface water is not only a pollution of the environment but also a high monetary loss as nutrients are wasted (Schuchardt et al. 2006). Covering the ponds to collect the biogas seems to be only a short-term solution because the well-known problem of pond maintenance (desludging) cannot be solved for a longer period of time. The only practical way to capture the biogas is a fermenter system. The biogas system with a fixed-bed fermenter is a very effective and flexible one for the highly polluted wastewater and changing conditions in palm oil mills (Wulfert et al. 2002). The biogas production in a fermenter system is lower compared to anaerobic ponds because of different retention times (see Table 1). Wulfert et al. (2002) found for POME a CH₄ yield of 0.251 kg/kg COD-degraded, same as the IPCC value (IPCC 2006b).

2 Methods and assumptions

2.1 Technology

For the composting process, an open windrow composting system (composting time between 6 and 10 weeks, 20 times

Table 1 Biogas from POME infixed-bed fermenter and anaero-bic pond (Wulfert et al. 2002;Schuchardt et al. 2001)

		Fermenter	Anaerobic pond
POME	m³/t FFB	0.65	0.65
CODdiss	kg/m ³ POME	25	25
CODsus	kg/m ³ POME	25	25
Hydr. retention time	Day	2	75
CODdiss-removal	-	0.90	0.90
CODsus-removal	-	0.15	0.60
CODdiss-removal	kg/m ³ POME	22.50	22.5
CODsus-removal	kg/m ³ POME	3.75	15.0
Sum COD-removal	kg/m ³ POME	26.3	37.5
Bacteria mass	_	0.05	0.05
	kg/m ³ POME	1.31	1.88
CODeff	kg/m ³ POME	24.9	35.6
	kg/t FFB	16.2	23.2
Spec. Biogas yield	m³/kg COD	0.55	0.55
Spec. CH ₄ yield	m³ CH ₄ /kg COD	0.35	0.35
	kg CH ₄ /kg COD	0.25	0.25
CH ₄ content	%	64	64
CH ₄ yield	m ³ /m ³ POME	8.73	12.47
Biogas yield	m ³ /m ³ POME	13.72	19.59
CH ₄ yield	m ³ /t FFB	5.67	8.10
Biogas yield	m³/t FFB	8.92	12.74

turning) is used as described by Schuchardt et al. (2002b). The composting process includes several activities such as: (a) the chopping of the EFB using a cutting mill; (b) forming of longitudinal heaps; (c) turning of the heaps using a self-driving windrow turning machine; (d) watering of the heaps using wastewater (POME) in order to balance the high water evaporation; and (e) finishing step. The total POME will be added to the chopped EFB during composting in open windrows, which are turned regularly by a turning machine. The electricity needed for operating the shredding machine is supplied by the biomass-powered heat and power plant. The temperature in windrows rise up to 70°C; at this stage of the composting process, more than 3.5 m³ POME per ton of EFB can be evaporated within 6 to 8 weeks. The produced compost can be either used directly on the plantation or it can be sold after finishing (screening), depending on the market demand. For this study, it is assumed that compost is used on the plantation.

2.2 Goal and scope

The goal of the paper is to evaluate environmental impacts of treating EFB and POME in palm oil mills. Conventional treatment systems for EFB (dumping or mulching in plantation) and POME (pond systems with discharging to precipitation) will be compared to advanced treatment systems (co-composting of EFB and POME in open windrow systems, with or without anaerobic pretreatment of POME in a

fermenter) as described by Schuchardt et al. (1998, 2002a, b, 2008) and Wulfert et al. (2002). Furthermore, the replacement of mineral fertilizer in oil palm plantation by EFB-POME compost will be included in the life cycle assessment.

2.3 Methodology

A life cycle approach has been used to calculate the environmental impacts from palm oil mill residue treatment systems outlined in Fig. 1. The ISO 14040/44 methodology for LCA has been used for these purposes (ISO 2006a, b). The aim of the study is to estimate the environmental impacts and particularly GHG emissions caused by EFB and POME in different waste treatment system.

Four waste management options are investigated:

- 1. Dumping of EFB and treating POME in ponds
- 2. Returning EFB to the plantation as mulch and treating POME in ponds
- 3. Using EFB and POME for co-composting
- 4. Using POME to produce biogas before co-composting

Assumptions used for this study:

- Compost is returned to the palm oil plantation according to the fertilizer demand.
- Leachate from composting area is returned to plantation or to compost.
- Infrastructure and capital goods are excluded.

Fig. 1 Palm oil production system including waste treatment and power generation (figures are expressed per functional unit, i.e., 1 t FFB)



- The palm oil residues are considered as burden free apart from transport emissions.
- The composting plant is located close to the palm oil mill, where electricity is produced on site from biomass (fibers and shells).
- No land use change occurs.

The residues derived from treating 1 tonne FFB are subjected to this analysis. Hence, 1-tonne FFB is chosen as functional unit, and system expansion is used to credit the system for substituting inorganic fertilizer such as potassium chloride, ammonium nitrate, magnesium sulfate, and triple phosphate when compost or EFB is returned to the plantation. For option 4, we assume that biogas generated from POME replaces diesel, which is used as starting fuel in the power plant. Alternatively, the biogas could replace fibers and shells in the power plant; however, those arrive burden free at the power plant apart from transport emissions and consequently the result for option 4 would be almost the same as for option 3 in that case.

The composition of POME and EFB is shown in Table 2. Only the emissions of fossil-derived carbon dioxide have been included in the calculations of the total GHG emissions: biogenic carbon dioxide has not been included in the waste as it is derived from the biomass that has been present in the waste and which had adsorbed that amount of carbon during the growth phase. Methane emissions from the biogenic carbon sources have been considered according to the IPCC suggestions (IPCC 2007).

As shown in Fig. 2, the compost production system has been credited for inorganic fertilizer replacement and for the production of diesel, providing the same energy as the biogas produced from POME. The credits have been implemented by subtracting the emissions from the life cycle of fertilizer production and diesel production, respectively. Although the nutrients in EFB and compost, especially nitrogen, are not immediately available, they become available within the lifetime of the plantation and are going to substitute inorganic fertilizer. In the case of EFB dumping, all nutrients are released to the environment during the lifetime of the plantation.

3 Results

3.1 Life cycle impact assessment

The first set of results show the environmental impacts of the four palm oil mill residue treatment options. The LCA modeling has been carried out in GaBi 4.3 (PE Europe 2003), using the CML 2001 method for estimating the environmental impacts (Guinée 2002). The environmental impacts considered in this study are shown in Table 3. Updated IPCC characterization factors for GHG are used to calculate the GWP. The GWP is emphasized in Table 4 and discussed in detail below.

3.2 Comparison of different residue treatment options

The results are based on life cycle emissions obtained from LCA databases. Additionally, N_2O emissions are calculated according to IPCC suggestions, and methane emissions from POME are measured (Wulfert et al. 2002; Schuchardt et al. 2008). It is assumed that all nutrients from dumping sites are released to the environment. Furthermore, we have not considered the enrichment of heavy metals in soils at the dumping site, which would lead to even higher toxicity impacts for option 1.

Table 2 Content of nutrients and heavy metals in EFB, POME, andcompost (Schuchardt et al. 2002a, b, Schuchardt et al. 2008, ownanalyses)

		EFB	POME ^a	Compost ¹
Mass	t/t FFB	0.23	0.65	_
Dry matter	kg/t	350	41	500
Organ. DM	kg/t DM	937	-	562
pН	-	5.5-5.7	4-4.5	7.7
Bulk density	t/m³	0.35	1	0.5
CODtot	kg/m³	-	50	-
Carbon	kg/t DM	432	370	351
C/N ratio	-	54	20	15
Nitrogen	kg/t DM	8.0	18.3	26.3
Phosphorus	kg/t DM	0.97	4.39	4.56
Potassium	kg/t DM	24.0	55.4	79.6
Calcium	kg/t DM	1.80	10.73	10.1
Magnesium	kg/t DM	1.80	15.12	12.8
Plumbum	g/t DM	1.80	7.1	7.8
Cadmium	g/t DM	< 0.3	< 0.3	< 0.3
Chrom	g/t DM	49.9	54.0	29.7
Copper	g/t DM	14.0	30.6	38
Nickel	g/t DM	30.5	1.34	10
Mercury	g/t DM	< 0.5	< 0.5	< 0.5
Zinc	g/t DM	37.9	80.2	154

^a The content of heavy metals in the POME is from analysis of cake from a three-phase decanter

^b The nutrient contents are calculated based on the EFB and POME data and a dry matter degradation of 60% during 10 weeks of composting time. The values differ between 11% and 45% from the analyzed data

At dumping sites, 20% aerobic and 80% anaerobic conditions occur (IPCC 2006a), and the biodegradable carbon is equally converted to carbon dioxide and methane. We have assumed 40% anaerobic conditions for EFB dumping in this study in order not to avoid overestimating

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the beneficial effects of the residue treatment. Methane emissions from EFB dumping are calculated, assuming 49% biodegradable carbon in EFB as measured in Schuchardt et al. (1998). Moreover, it is assumed that 0.5% of the nitrogen in EFB is converted to nitrous oxide at EFB dumping sites. Returning untreated EFB to the plantation does not just recycle nutrients; extensive application reduces anaerobic conditions. As a consequence, the formation of methane is reduced significantly compared to EFB dumping.

The comparison of all environmental impacts for the four treatment options is shown in Table 4. All environmental impacts decrease from option 1 to option 4, e.g., GWP from 245 to 5.1 kg CO_{2eq} , HTP from 9E–3 to -3E-1 kg DCB_{eq} , and AP from 1E–3 kg to -3E-3 SO_{2eq} . These values depend on the fertilizer production systems as system expansion is used for this study. They would be higher for fertilizer production systems with lower environmental impacts as those from the database. Negative values reveal not just reduced environmental impacts but also a net environmental benefit. The main difference between options 2 and 3 is avoidance of methane formation in ponds and nitrogen recovery from POME in option 3, leading to considerable reduction of GWP and EP compared to option 1 and option 2.

3.2.1 Environmental impacts of the advanced waste treatment options

The advanced palm oil mill waste treatment options (option 3 and option 4) are investigated in detail concerning their GWP. Figure 2 shows the GWP of composting and composting with previous biogas generation from POME, assuming that 100% biogas is recovered.

Emissions from the composting process are calculated based on the assumptions that 1% of the total carbon input



Fig. 2 GWP of composting and composting with previous biogas generation from POME

Environmental impact	Abbreviation	Unit
Abiotic resource depletion potential	ADP	kg Sb-equiv.
Acidification potential	AP	kg SO ₂ -equiv
Eutrophication potential	EP	kg phosphate-equiv.
Freshwater aquatic ecotoxicity potential	FAETP	kg DCB-equiv.
Global warming potential (100 years)	GWP	kg CO ₂ -equiv.
Human toxicity potential	HTP	kg DCB-equiv.
Ozone layer depletion potential	OLDP	kg R11-equiv.
Photochem. ozone creation potential	POCP	kg ethene-equiv.
Terrestric ecotoxicity potential	TETP	kg DCB-equiv.

Table 3 Environmental impactsaccording to the CML method

is converted to methane and 0.5% of the total nitrogen input to nitrous oxide (IPCC 2006b). In addition, ancillary material and emissions from diesel consumption of compost turning machines are considered. The composting plant is the main contributor to GWP due to the formation of methane and nitrous oxide. The contribution of ancillary materials and transport of EFB from the mill to the composting plant is almost negligible. Although the composting process causes emissions of approximately 16 kg CO_{2eq} alone, the avoided burden from nutrient recycling due to the application of compost on the plantation results in a total GWP of 7.4 kg CO_{2eq} , and with biogas recovery it is further reduced to 5.1 kg CO_{2eq} . Obviously, returning nutrients to the plantation has a tremendous effect on GWP but also across all environmental impact categories.

The difference between option 3 and option 4 is relatively small for all environmental impacts apart from GWP because nutrients in POME are recovered in both options. Despite the potential benefit of biogas generation from POME, there is also a certain risk involved. The calculations above are based on the assumption that all generated biogas is recovered. This is a best-case assumption as usually certain biogas losses occur due to leakages from the plant itself and from storage in open basins or ponds after fermentation. In order to estimate the risk concerning this matter, a sensitivity analysis is carried out and the result is shown in Fig. 3.

Table 4 Environmental impacts per tonne FFB

	Option 1	Option 2	Option 3	Option 4
ADP	1.2E-03	-2.9E-02	-1.6E-01	-1.6E-01
AP	1.3E-03	-1.1E-02	-3.1E-02	-3.1E-02
EP	1.4E+00	1.1E+00	-3.0E-03	-3.0E-03
FAETP	4.6E-04	-4.3E-03	-4.6E-02	-4.7E-02
GWP	2.5E+02	1.3E+02	7.3E+00	5.1E+00
HTP	9.3E-03	-7.6E-02	-3.3E-01	-3.3E-01
OLPD	3.6E-10	-1.5E-07	-3.0E-07	-3.0E-07
POCP	5.9E-02	3.1E-02	6.0E-04	5.5E-05
TETP	1.3E-04	-1.5E-03	-1.4E-02	-1.4E-02

The GWP reduction diminishes rapidly if the biogas is not completely recovered. When approximately 2% methane is lost, the GWP equals that of composting without biogas generation. Generally, the loss from anaerobic biogas production plants is between 0.1% and 8.6%, depending on the degradation rate in the fermenter, the hydraulic retention time, and the storage conditions of the effluent (FNR 2009). This result reveals that the management practice has a significant effect on the environmental performance of biogas plants. Poor management can reverse the reduction of emission, particularly when the total GHG reduction potential is low as in the case when comparing option 3 with option 4.

In principle, the same applies to the operation of composting plants, and therefore the best and worst operation conditions for composting are calculated based on IPCC default values. The value for nitrous oxide formation varies between 0.5% and 5%, while the estimated methane emissions range from 0.5% to a few percent (IPCC 2006b); we have assumed up to 5% for the worst-case calculation. The GWP of the composting plant increases to 79 kg CO_{2eq} when it is poorly managed compared to 16.0 kg CO_{2eq} when managed well. The total GWP increases from 7.4 to 72.7 kg CO_{2eq}. However, the GWP of treatment option 3 is considerably lower compared to the GWP of option 2 (GWP 128 kg CO_{2ea}) and option 1 (GWP 245 kg CO_{2ea})—even if the plant is poorly managed. The composting process recovers not just nutrient from EFB and POME, it also dries POME simultaneously, which is its main advantage.

Approximately nine million hectares is used for palm oil plantations in Indonesia and Malaysia, representing 87% of global CPO production in 2009 (USDA 2009, 2010). Assuming an average yield of 20 t FFB per hectare and a GWP of 128 CO_{2eq} per tonne FFB (option 2), then the GWP reduction potential can be calculated according to:

GWP reduction potential = yield FFB/hectare \times area

$\times \Delta GWP/tFFB$

This rough estimate reveals that the GWP reduction potential is enormous, more than 21 million t CO_{2eq} , if

Fig. 3 GWP of option 4 depending on fugitive emissions



POME treatment in ponds and EFB returning is considered as state of the art. This amount is in a similar range as the annual GHG emissions of Slovenia in 2007.

4 Discussion and conclusions

Treatment of 1 tonne FFB in palm oil mills generates 230 kg EFB and 650 kg POME. Hence, waste management influences significantly the environmental performance of palm oil mills, e.g., if EFB is dumped and POME is stored in ponds, this "treatment" causes per tonne FFB a GWP of 245 kg CO_{2eg}, EP of 1.4 kg PO_{4eg}, and ADP of 1E-3 kg Sbeq compared to 7.4 kg CO2eq, EP of -0.003 kg PO_{4ea}, and ADP of -1.6E-1 kg Sb_{ea} when both are converted to compost and returned to the plantation. The environmental performance can be improved even further if POME is used to generate biogas before it is transferred to the composting plant. However, this just applies if the biogas plant is operated properly. Fugitive biogas emissions larger than 2% would have adverse impacts. The effect of temporary carbon storage due to compost application is not taken into account in this study, although that would further reduce the GWP for option 3 and option 4.

The main advantage of the composting process is that it allows the treatment of POME and EFB simultaneously and enables recovery of nutrient from both. Approximately 68% K, 43% Mg, 22% N, and 14% P are recovered when figures from Rettenmaier et al. (2007) are used as reference. Environmental benefits from co-composting derive from avoided methane emissions from open POME ponds and returning of nutrients to the plantation. At a global scale, the GWP reduction potential could be in the range of 20–25 million t CO_{2eq} . Hence, the proposed co-composting process might be eligible for CDM projects.

Returning EFB or compost to the plantation has several positive impacts on soil physical and chemical properties. That is hardly considered in LCA studies, although soil property improvement leads to higher yield of palm oil and avoids soil depletion according to Goh and Gurmit (Goh et al. 1994; Gurmit et al. 1990; cit. in Chee and Chiu 1999). Moreover, approximately 78 kg carbon per tonne FFB is returned to the plantation through compost application. Hence, converting EFB and POME to compost and returning it to the plantation are environmentally beneficial; it saves resources and leads to more sustainable palm oil production systems.

The demand for palm oil will increase due to increasing population (Corley 2009) and that will most likely cause land use change. If the demand for palm oil increases, enhancing the yield per hectare is an appropriate possibility to reduce the need for land use change. However, if EFB is used for energy production, as it is discussed and realized by some palm oil mills (Husain et al. 2003; Arrieta et al. 2007; Shuit et al. 2009), a sustainable POME utilization is not possible anymore.

5 Outlook

In addition to the benefits identified in this study, the application of compost on palm oil plantations has more to offer: temporary storage of soil carbon, improving soil quality, and protection from soil erosion just to name a few. Currently, those aspects are hardly part of the life cycle impact assessment. The development of regional-specific LCIA methods is hampered by the lack of regional and sitespecific data. To collect those data particularly for Indonesia is of mutual interest of palm oil plantation owners, palm oil mill operators, and the scientific community in order to develop more sustainable palm oil production systems. Acknowledgement The authors acknowledge Dr. Tjahono Herawan (Indonesian Oil Palm Research Institute, IOPRI, Medan, Indonesia) and Dipl.-Ing. Klaus Wulfert (UTEC, Bremen, Germany) for valuable data and discussion.

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