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Author(s)	Yano, Junya; Aoki, Tatsuki; Nakamura, Kazuo; Yamada, Kazuo; Sakai, Shin-ichi
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## Life Cycle Assessment of Hydrogenated Biodiesel Production from Waste Cooking Oil Using the Catalytic Cracking and Hydrogenation Method

## Author names and affiliations

Junya YANO\*, Tatsuki AOKI\*, Kazuo NAKAMURA\*\*, Kazuo YAMADA\*\*\*, Shin-ichi SAKAI \*

\*Environment Preservation Research Center, Kyoto University, Yoshida-Honmachi, Sakyo-ku, Kyoto 606-8501, Japan \*\*Advanced Scientific Technology & Management, 134 Chudoji Minamimachi, Shimogyo-ku, Kyoto 606-8813, Japan \*\*\* Kyoto City Environmental Policy Bureau, 384 Ichinofunairicho, Nakagyo-ku, Kyoto 604-0924, Japan

## **Corresponding author**

Junya YANO Environment Preservation Research Center, Kyoto University, Yoshida-Honmachi, Sakyo-ku, Kyoto 606-8501, Japan E-mail: yano@eprc.kyoto-u.ac.jp Phone: +81-75-753-7709

## Highlights

- LCA with uncertainty analysis was conducted for BDF production from waste cooking oil.
- HBD production scenario reduced total environmental impacts by 50–71% compared with incineration.
- FAME-type BDF provides limited future benefit compared with fossil-derived diesel.
- A shift from FAME-type BDF to HBD would more effectively reduce total environmental impacts.

#### 1 Abstract

 $\mathbf{2}$ There is a worldwide trend towards stricter control of diesel exhaust emissions, however presently, there are technical impediments to the use of FAME (fatty acid methyl esters)-type 3 biodiesel fuel (BDF). Although hydrogenated biodiesel (HBD) is anticipated as a new diesel 4 fuel, the environmental performance of HBD and its utilization system have not been  $\mathbf{5}$ adequately clarified. Especially when waste cooking oil is used as feedstock, not only biofuel 6 7production but also the treatment of waste cooking oil is an important function for society. A 8 life cycle assessment (LCA), including uncertainty analysis, was conducted to determine the environmental benefits (global warming, fossil fuel consumption, urban air pollution, and 9 acidification) of HBD produced from waste cooking oil via catalytic cracking and 10hydrogenation, compared with fossil-derived diesel fuel or FAME-type BDF. Combined 11 12functional unit including "treatment of waste cooking oil" and "running diesel vehicle for 13household waste collection" was established in the context of Kyoto city, Japan. The 14calculation utilized characterization, damage, and integration factors identified by LIME2, which was based on an endpoint modeling method. The results show that if diesel vehicles 15that comply with the new Japanese long-term emissions gas standard are commonly used in 1617the future, the benefit of FAME-type BDF will be relatively limited. Furthermore, the scenario that introduced HBD was most effective in reducing total environmental impact, 18 19meaning that a shift from FAME-type BDF to HBD would be more beneficial.

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## 21 Keywords

# 22 Waste-to-fuel, Biodiesel fuel (BDF), Hydrogenated biodiesel (HBD), Waste cooking oil,

- 23 Catalytic cracking method, Life cycle assessment (LCA)
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#### 27 **1. Introduction**

28To develop a low-carbon society, it is important to promote the production of biofuels such as biodiesel fuel (BDF). Biofuels are produced worldwide: Biofuel consumption in road 29transport accounted for 1.3 Mboe/day (million barrels of oil equivalent per day) as of 2011, 30 and is expected to increase to 4.1 Mboe/day in 2035, an increase from 3% of road transport 31fuel demand in 2011 to 8% in 2035. Of this, biodiesel consumption accounted for 0.4 3233 Mboe/day and is estimated to be 1.1 Mboe in 2035 (IEA, 2013). The fuels that are currently 34under development utilize non-food feedstock, including waste (Naik et al., 2010; Sims et al., 2010; Takamizawa et al., 2013). Such fuels are thought to be more environmentally desirable, 35because biofuels derived from food crops such as soybeans are associated with a number of 36 problems: competition with food agriculture for land and water use, and widely varying 37 38 assessments of net greenhouse gas (GHG) reductions once land-use change is taken into 39 account (Fargione et al., 2008; Searchinger et al., 2008).

40 In Kyoto city, Japan, waste cooking oil has been collected from households since 1998, and used to produce BDF since 2004. The BDF production facility operated by Kyoto city has 41 a capacity of 5 kL/day (1,500 kL/yr), and is the largest facility managed by a local 4243government in Japan. The alkali catalysis method is commonly used to convert waste cooking oil to BDF, which consists of fatty acid methyl esters (FAME) (Meher, 2006; Salvo, 2012). 44As of fiscal year (FY) 2012, approximately 1,300 kL of FAME-type BDF has been produced 45annually from waste cooking oil (approximately 196 kL from households and 1,110 kL from 46businesses). Considering that the generation of waste cooking oil from households was 47estimated to be approximately 1,140 kL within Kyoto city, this represents a collection rate of 4817% waste cooking oil from households for BDF production. The produced BDF has been 49used as fuel for city buses (B20) and household waste collection vehicles (B100) within 50Kyoto city. 51

From the standpoint of air pollution, there has been an increasing focus globally on the 5253control of gas emissions from diesel vehicles (EC, 2007). As shown in Table 1, Japan has also 54established and enforced gas emission standards that regulate pollutants such as  $NO_x$  and particulate matter (PM) emissions in a number of stages (DELPHI, 2014). For instance, in the 5556case of  $NO_x$  emissions, the standard value for vehicles of gross weight more than 3.5 tons is 3.38 g-NO<sub>x</sub>/kWh for the new short-term emissions gas standard (2002-2004), 2.00 57g-NO<sub>x</sub>/kWh for the new long-term standard (2005–2008), and 0.700 g-NO<sub>x</sub>/kWh for the post 5859new long-term standard (2009 onward). However, FAME-type BDF is occasionally problematic when used in diesel vehicles (Fukuda et al., 2008; WFCC, 2013). In particular, 60 some technical problems have arisen in terms of the suitability of BDF for new-model diesel 61 vehicles equipped with diesel particulate filters and NO<sub>x</sub> reduction devices after 62implementation of the new long-term emissions standard. 63

Table 1

64 65

These problems, which include the mixing of fuel with engine oil and the poor 66 performance of NO<sub>x</sub> reduction devices, have become major impediments to BDF use. 67 Therefore, there are some challenges involved in producing new diesel fuels. HVO 68 (hydrotreating of vegetable oils) has been developed and commercially supplied to mainly EU 69 70regions. HVO is also known as renewable diesel or HDRD (hydrogenation derived renewable 71diesel) in the USA, and as HBD (hydrogenated biodiesel) in the Far East including Japan (Neste Oil, 2014). HVO consists mainly of paraffins and is free of aromatics, oxygen, and 7273sulfur. HVO generally shows higher cetane index and higher oxidation stabilities compared to FAME-type BDF (Bezergianni et al., 2013). HVO can be applied not only to vegetable oil but 7475also to animal fats. Therefore, the HVO production method is expected to contribute to the 76expanding feedstock of BDF and resulting increases in fuel supplies. Neste Oil is the world's largest producer of HVO, which it supplies under the brand name "NExBTL." Its production 77capacity is approximately 2 million ton/yr from four facilities. HVO is made by 78hydrotreatment of vegetable oils and animal fats, but additionally, waste and residues such as 7980 waste animal fat accounted for over half of feedstocks (Neste Oil, 2013). The OPTIBIO project operated for 3.5 years, between autumn 2007 and December 2010, to demonstrate the 81 use of NExBTL for city buses in Helsinki. The project confirmed that HVO can replace 82fossil-derived diesel without any modifications to the vehicles or refueling system (Nylund et 83 al., 2011). The Worldwide Fuel Charter (WWFC) now evaluates HVO as being highly suited 84 as a blendstock for diesel fuel (WWFC, 2013). 85

86 HVO has been also developed in Japan, where it is often called HBD. Attention has been given to the production method, namely catalytic cracking and hydrogenation (Tani et al., 87 2011a, 2011b), and a three-year demonstration project for this new method, involving Kyoto 88 city, ASTEM, and other companies, was operational between April 2012 and March 2015 89 90 (ASTEM, 2013; Kakuta, 2014; Takasuga, 2014). Unlike the general methods, the catalytic 91cracking process promotes decarbonization, which has the notable benefit of reducing the consumption of energy and H<sub>2</sub>. Hydrogenation after cracking requires normal pressure and 9293 temperature conditions (1.0 MPaG at 150°C) and less H<sub>2</sub>, whereas direct hydrogenation requires high pressure and temperature (4-6 MPaG at 300-350°C) and consumes 10 times the 9495 amount of H<sub>2</sub>. Therefore, hydrogenation after cracking is suitable for small- and mid-sized 96 production facilities that utilize regional feedstocks such as waste cooking oil. The produced HBD has similar characteristics to those of diesel fuel, including calorific content and boiling 97 point. The characteristics of some fuels, including HBD produced in the demonstration 98 project at Kyoto city (ASTEM, 2013), are summarized in the electronic supplementary 99100 material.

101 A life cycle assessment (LCA) was performed to evaluate the effectiveness of BDF use in 102reducing negative environmental impacts. Liang et al. (2013) quantitatively showed that 103 feedstocks had different environmental performances by comparing seven feedstocks including waste cooking oil. Dufour et al. (2012) used LCA to evaluate six environmental 104 impacts for four types of free fatty acid-rich wastes (used cooking oil, animal fats, sewage 105sludges), and concluded that biodiesel fuel from used cooking oil potentially achieved the 106 107 most favorable environmental performance. On the other hand, when waste cooking oil was used for BDF production, the treatment (BDF production) method was also an important 108factor in environmental performance (Morais, 2010; Varanda, 2011). The environmental 109110 performance of HBD and its utilization system has not been adequately clarified. Garraín et al. (2014) showed that HBD blend diesel (13% blend) from soybean oil could reduce fossil fuel 111 112consumption by 2% and GHG emissions by 9% at well-to-tank stage compared with FAME 113blend diesel. Neste Oil (2014) estimated that the GHG reduction effects of NExBTL compared with fossil-derived diesel were 47%, 49%, and 91% for palm oil, rapeseed oil, and 114animal fat feedstocks, respectively; and, by conducting exhaust gas emission tests, showed 115that emissions of PM were reduced by approximately 30-40%. Evaluating exhaust gas 116117emissions such as NO<sub>x</sub> and PM from HBD production and its utilization system by means of a 118 life cycle approach is also necessary to determine environmental performance compared with 119the FAME-type BDF that is generally used. Arvidsson et al. (2011) conducted an LCA of 120 HBD production from rapeseed oil, palm oil, and Jatropha considering four environmental 121impacts: fossil fuel consumption, global warming, acidification, and eutrophication. The 122functional unit of the analysis focused on fuel supply (1 kWh of energy output). However, 123biofuel production and the treatment of waste cooking oil are important functions for society, 124especially when waste cooking oil is used as feedstock.

The purpose of this study was to clarify the effects of HBD on reducing environmental impacts compared with fossil-derived diesel fuel, or FAME-type BDF. An LCA including characterization, damage assessment, integration assessment, and uncertainty analysis was conducted to evaluate a number of environmental impacts including global warming and air pollution.

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#### 131 **2. Materials and methods**

#### 132 **2.1. Functional unit and system boundary**

Table 2 shows the characteristics of the waste cooking oil considered in this study. Combined functional unit including "treatment of waste cooking oil" and "running diesel vehicle for household waste collection" was established. The first functional unit was assumed to be the treatment of 1,142 kL/yr and 1,108 kL/yr of waste cooking oil from households and businesses, respectively. The second functional unit assumed that 41.1 TJ of diesel fuel (fossil-derived diesel fuel, FAME-type BDF, and HBD) was consumed by diesel
vehicles used to collect household waste within Kyoto city. Both amounts reflected the actual
situation in Kyoto city as of FY2012. Only waste cooking oil was considered as feedstock,
although animal fats could be also used to produce HBD.

The system boundary included the collection of waste cooking oil, treatment or recycling (BDF production), and fossil-derived diesel fuel and/or BDF consumption of a diesel-powered collection vehicle. With regard to fossil fuel consumption, the system boundary considered the stages from raw material extraction to final use (combustion), and associated environmental impacts were allocated to the process that consumed the fossil fuel.

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#### Table 2

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## 149 **2.2. Environmental impacts**

LIME1, a Japanese life-cycle impact assessment method, was developed for the first term (1998–2003) of a national LCA project, whereas LIME2 was developed for the second term (2003–2006) (JLCA, 2012). LIME2 was based on an endpoint modeling method, and considered environmental conditions, such as weather and population density, in Japan such as weather and population density. Therefore, LIME2 is mainly applied in Japan (European Commission, 2010).

LIME2 comprises 19 category endpoints that are connected by 15 environmental impact 156categories (JLCA, 2012). These environmental impact categories are assessed by integration, 157following characterization and damage assessment. Characterization is the first step, in which 158potential environmental impacts are assessed for each impact category. It is possible to 159compare or integrate the impacts of two or more environmentally damaging substances on the 160 specific impact category. Damage assessment is the second step in assessing the amount of 161damage that can occur for each object of protection. In LIME, four items were defined as 162objects of protection: human health, social assets, biodiversity, and primary production. The 163 164Disability Adjusted Life Year (DALY), which is used internationally for health statistics, was 165defined as the damage index for human health. An economic index in Japan, that can comprehensively measure the impact on various components (agricultural products, forests, 166167marine products, and resources), was defined as the damage index for social assets. EINES, which was originally based on the methodology for assessing extinction risk in the field of 168169 conservation ecology, was defined as the damage index for biodiversity. Net primary 170production (NPP), which is widely used as an index of ecosystem richness in the fields of biology and landscape architecture, was defined as the damage index for primary production. 171172Integration is the final step, in which the results of the four objects of protection are converted into a single index by means of weighting factors. In LIME2, conjoint, analysis was adopted 173174for weighting the area of protection.

The considered environmental impacts in this study were fossil fuel consumption, global warming, urban area air pollution, and acidification. The calculation utilized characterization, damage, and integration factors identified by LIME2. These factors were retrieved from the website of the Life Cycle Assessment Society of Japan (JLCA). The impact categories and their objects are listed in Table 3. With regards to global warming, CO<sub>2</sub> emissions derived from biomass are regarded as carbon-neutral, and were therefore excluded from the calculation. Table 4 shows a list of impact categories and corresponding damage factors.

Table 3

Table 4

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### 186 **2.3.** Scenario setting

187The scenarios in this study are listed in Table 5. As a base scenario, all waste cooking oil 188from households and businesses was assumed to be collected with mixed waste and then incinerated, while diesel fuel was used to operate household waste collection vehicles. With 189 regard to a baseline scenario, two sub-scenarios were considered, based on the diesel vehicle 190191types covered by the Japanese emission gas standard: one where diesel vehicles complied 192 with the new short-term emissions gas standard (scenario S1-short), and the other where 193diesel vehicles complied with the new long-term emissions gas standard (scenario S1-long). S1-short is the scenario that reflects the general, current situation in Japan, while S1-long 194reflects the future exhaust gas control situation. 195

196For the BDF utilization scenarios, 196 kL/yr of waste cooking oil from households (17% as collection efficiency) and 1,108 kL/yr from businesses were collected separately, from 197 which BDF was produced by the alkali catalysis method (scenario S2-short), or the catalytic 198cracking and hydrogenation method (scenario S3-long). The remaining 947 kL/yr from 199households was incinerated with mixed waste. Because of the above-mentioned technical 200201problems for diesel particulate filters required for the long-term emissions gas standard, it was assumed that FAME-type BDF was used only for diesel vehicles that complied with the new 202short-term emissions gas standard (S2-short). It could be said that S2-short reflects the current 203204situation in Kyoto city; on the other hand, HBD could be used for diesel vehicles that comply with both the new short-term and the new long-term emission gas standards. However, HBD 205206was used for diesel vehicles that only complied with the new long-term emissions gas 207standard in S3-long, in order to evaluate the more desirable HBD utilization system.

Fig. 1 shows the system flow of each scenario, and each process setting is explained in section 2.4.

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211

### Table 5

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#### 213

## Fig. 1

214 **2.4.** Unit processes and data collection

Important parameters used in this study are listed in Table 6, and each process is briefly introduced below. The later sections also discuss the uncertainties that were considered for some parameters.

218

219 **2.4.1.** Collection

A grid city model (Ishikawa, 1996) was used for calculating the annual collection distance and diesel fuel consumption. Direct emissions of CO<sub>2</sub> and CH<sub>4</sub>, N<sub>2</sub>O, SO<sub>x</sub>, NO<sub>x</sub>, PM<sub>2.5</sub>, and PM<sub>10</sub> from household waste collection vehicles were then estimated.

In S1-short and S1-long, all waste cooking oil from households was assumed to be collected twice a week with mixed waste, and the weight of waste cooking oil accounted for 0.50% of household waste. Therefore, the estimated amount of diesel fuel consumption was allocated based on weight. In S2-short and S3-long, 947 kL/yr of waste cooking oil, which was failed to be collected separately, was also assumed to be collected with mixed waste.

In S2-short and S3-long, FAME-type and HBD could both be used to power household waste collection vehicles. However, this benefit was not included in the "collection" process, but rather in the "running household waste collection vehicle" process, as will be explained later. Only fossil-derived diesel fuel was consumed in this process.

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#### 233 **2.4.2.** Incineration with energy recovery

In this process, it was assumed that waste cooking oil and waste glycerin, which was co-generated by the FAME-type BDF production process using an alkali-catalyzed method (S2-short), were incinerated with household waste in the incineration facility within Kyoto city. Direct emissions of CH<sub>4</sub>, N<sub>2</sub>O, SO<sub>x</sub>, NO<sub>x</sub>, PM<sub>2.5</sub>, and PM<sub>10</sub> from waste combustion were calculated. CO<sub>2</sub> emissions from fossil-derived carbon were also counted because waste glycerin contained unreacted methanol. It was assumed that there was no residue, as nearly all the waste cooking oil was combustible.

Electricity was considered as energy consumption and calculated using an empirical formula based on household waste composition (NIES, 2008). At the same time, an electricity production facility with an efficiency of 15% was assumed to be associated with the incineration facility. Both consumption and substituted electricity refer to commercial electricity from utility companies. The weighted average emission factors for 10 companies in Japan were used in this analysis.

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#### 248 2.4.3. FAME-type BDF production by alkali catalysis method

249Waste cooking oil and methanol were used for BDF production by an alkali-catalyzed 250method. Electricity and paraffin oil were considered as energy consumption. Inventory data were obtained from the BDF production facility in Kyoto city. It should be noted that 251fossil-derived carbon in the produced FAME-type BDF accounted for approximately 7.0% of 252the total carbon content because methanol, which was produced from natural gas, was used 253for FAME-type BDF. Co-generated waste glycerin contained KOH as a catalyst, unreacted 254255methanol, and waste cooking oil. Waste glycerin was assumed to be treated during incineration with the energy recovery process mentioned previously. The characteristics of 256waste glycerin are presented in the electronic supplementary material. 257

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### 259 **2.4.4. HBD production by catalytic cracking and hydrogenation method**

260The HBD production process can be described as follows: Firstly, waste cooking oil was 261degraded at around 400–500°C, and the organic acids contained in the decomposed oil were decomposed for conversion into hydrocarbons at the reactor. Offgas consisting of CO<sub>2</sub>, CO, 262H<sub>2</sub>, CH<sub>4</sub>, and other hydrocarbons, was also produced at this stage. Secondly, the produced 263hydrocarbons were separated into high-, intermediate- (biodiesel), and low-boiling-point oils 264265using two condensers. Thirdly, a minor amount of acid contained in the biodiesel was removed, and the biodiesel was refined for long-term stability. Finally, the refined biodiesel 266267was hydrogenated using H<sub>2</sub> at 150–250°C in order to improve stability for oxidation and heat. This study refers to hydrogenated diesel fuel as HBD. In total, 90.7% of the input waste 268cooking oil on an energy basis could be recovered in the form of HBD, other oils, and offgas. 269270The HBD showed an energy yield of 52.8%, which was lower than that of FAME-type BDF.

Inventory data were obtained from a demonstration project by ASTEM. Electricity was considered as energy consumption. Offgas, and high- and low-boiling-point oils, which were co-generated in this process, were combusted for heating supply. Of these types, only the low-boiling-point oil could be exported from the facility as surplus energy after heating. Therefore, it was assumed that naphtha was substituted by surplus low-boiling-point oil on a heating value basis.

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#### 278 **2.4.5. Diesel fuel production**

The production of diesel fuel as commercially used in Japan was assumed in this process. Inventory data provided by JLCA (JLCA, 2012) were used to calculate  $CO_2$ ,  $SO_2$ ,  $NO_2$ , and PM<sub>10</sub> emissions. All PM emissions were regarded as PM<sub>10</sub> in this process because of a lack of data.

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#### 284 **2.4.6. Running household waste collection vehicle**

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Direct emissions of  $CO_2$ ,  $SO_x$ ,  $NO_x$ ,  $PM_{2.5}$ , and  $PM_{10}$  were calculated in this process.  $CO_2$ 

286and SO<sub>x</sub> emissions were calculated using the elemental composition of the fuels. The other 287emission factors (EFs) were assumed to differ according to the types of diesel vehicles covered by the Japanese emission gas standards: the new short-term emissions gas standard 288(S1-short, S2-short), and the new long-term emissions gas standard (scenario S1-long, 289S3-long). The type of fuel also affected the EFs. Therefore, the results of emission testing 290from the vehicles currently used for household waste collection by Kyoto city, which 291292 considered the type of fuel, were adopted as EFs (Kyoto city) after converting the data from 293g/kWh to g/L. The proportions of  $PM_{10}$  and  $PM_{2.5}$  were based on a previous study by Motoshita (2008), since these data were absent from the emission testing in Kyoto city. 294However, it was assumed that there was no difference between EFs in HBD and those in 295diesel fuel, because there were no actual data for HBD. The demonstration project by ASTEM 296297plans to include a fleet running test.

298 299

#### Table 6

#### **300 3. Results and Discussion**

#### 301 3.1. Characterization

302The estimated characterization results are shown in Fig. 2. With regard to global warming and fossil fuel consumption, there are no differences between S1-short and S1-long. In every 303 scenario, CO<sub>2</sub> emission was the dominant contributor to global warming compared with other 304emissions, CH<sub>4</sub>, and N<sub>2</sub>O. The running household waste collection vehicle process 305contributed to increases in GHG emissions in S1 and S3-long, because the process consumed 306 fossil-derived diesel fuel. GHG emissions from the incineration process in S2-short exceeded 307 those in S1 and S3-long, because the production of FATE-type BDF in S2-short incinerated 308 waste glycerin containing carbon from fossil-derived methanol. Nevertheless, net total GHG 309 emissions from S2-short showed a negative value (-150 t-CO<sub>2</sub> eq/yr), indicating that the use of 310 waste cooking oil for FAME-type BDF could help reduce net GHG emissions. On the 311312contrary, in S1 and S3-long, net total GHG emissions were estimated to be 1561 t-CO<sub>2</sub> eq/yr 313and 547 t-CO<sub>2</sub> eq/yr respectively. GHG reduction effects from energy recovery in incineration and substituted naphtha were not sufficiently large to cancel out the GHG emissions, which 314315were mainly derived from the running of household waste collection vehicles. GHG emissions in S3-long were reduced by 65% compared with S1, which used fossil-derived 316 317diesel. On the other hand, RED (European Commission, 2009: Directive 2009/28/EC) introduced GHG reduction effects of 47% and 65% by HVO from rapeseed oil and sunflower, 318respectively. NExBTL from palm oil, and rapeseed oil reduced GHG emissions by 47% and 31949%, respectively, compared with fossil-derived diesel (Neste Oil, 2014). It was implied that 320 321HBD production from waste cooking oil and its utilization system in this study could achieve 322equivalent GHG reduction, despite the different system boundaries employed by the two

323 studies.

324Fossil fuel consumption in S2-short and S3-long could be reduced by 120% and 58%, as 325326

compared with S1. The FAME-type BDF production process consumed more fossil fuels than HBD, because the latter could use by-products such as offgas for heating supply. Surplus low-boiling-point oil also contributed to reductions in the net consumption of fossil fuel. 327

Urban area air pollution and acidification showed similar results, which indicated that the 328 329 type of diesel vehicle covered by the Japanese emission gas standard had the largest impact on 330urban area air pollution. Although  $NO_x$  emission was the dominant contributor to urban area air pollution and acidification in every scenario, the emission amounts differed by the type of 331diesel vehicle. In S1-short and S2-short, NO<sub>x</sub> emissions from household waste collection 332333 vehicles that complied with the new short-term emissions gas standard were dominant. On the 334contrary, it was apparent that a shift to vehicle types that complied with the new long-term 335emissions gas standard could dramatically reduce NO<sub>x</sub> emissions. As net results, urban area air pollution and acidification in S1-long were decreased by 90% and 78% respectively, 336compared with S1-short. Energy recovery during incineration also contributed to reducing 337emissions of air pollutants especially SO<sub>x</sub>, which was one factor that made S1-short and 338339 S1-long superior to S2-short and S3-long, respectively, in terms of urban area air pollution and acidification. In S3-long, reductions of 76% in urban area air pollution and 75% in 340 acidification were achieved compared with S1-short. 341

Fig. 2

342343

#### **3.2.** Damage assessment 344

Fig. 3 shows the estimated damage for the four endpoints (in all endpoints, lower values 345indicate less damage). NO<sub>x</sub> emissions from household waste collection vehicles affected 346 human health. NOx emissions accounted for 42% and 67% of overall damage in the S1-short 347and S2-short scenarios, compared with 18% and 34% in S1-long and S3-long. PM2.5 emission 348from incineration also affected human health in all scenarios. SO<sub>x</sub> emissions showed negative 349350value as a result of the substitution effect of the electricity production in incineration with energy recovery process, although their effect was small. In S2-short, CO<sub>2</sub> emission also 351352showed negative value because this scenario consumed no fossil-derived diesel to operate the 353household waste collection vehicles. These results imply that, as a treatment for waste cooking oil, a shift from incineration to BDF production contributes to protecting human 354355health, mainly as a result of avoiding the emission of  $PM_{2.5}$ .

Crude oil extraction for diesel production, and subsequent emission of NO<sub>x</sub> by operating 356household waste collection vehicles, were the dominant sources of damage to social assets. In 357 S2-short, NO<sub>x</sub> emissions accounted for 92% of the total impact on social assets. The result 358indicated that shifting from incineration of waste cooking oil as in S1-short to FAME-type 359

BDF production as in S2-short had more benefits than shifting from diesel vehicles compliant with the new short-term gas standard (such as S1-short) to those compliant with the new long-term gas standard (such as S1-long).

Biodiversity was affected only by coal consumption. Electricity produced via incineration with energy recovery process was the dominant factor even if the electricity consumed in the other processes was considered.

There were two major factors in primary production: one was the coal reduction effect achieved by electricity substitution at incineration with energy recovery process, and the other was  $NO_x$  emission associated with operating household waste collection vehicles. Because S1-long could benefit from both these factors, the scenario showed the least impact on biodiversity.

The results are also summarized in Table 7. Comparing the four scenarios, S3-long showed the best environmental performance for human health and social assets; S1-short and S1-long were the best for biodiversity. S1-long also showed the best performance for net primary production. It is important to evaluate these four endpoints in combination, which is discussed in the next section.

> Fig. 3 Table 7

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## 378

## **379 3.3. Integration**

The estimated integration results from each scenario are shown in Fig. 4. Compared with S1-short, S1-long could reduce urban area air pollution and acidification, whereas S2-short could reduce global warming and fossil fuel consumption. In terms of the net results, the use of FAME-type BDFs in S2-short could reduce environmental impacts by 42% compared with S1-short, while S1-long also reduced environmental impact by 42%.

These results imply that if diesel vehicles compliant with the new long-term emissions gas 385386 standard are commonly used in the future, as was considered in S1-long, the benefit of using 387FAME-type BDF (S2-short) will apparently be relatively low. This is because FAME-type BDF cannot be used for vehicles that comply with the long-term emissions gas standard. 388 389 However, if HBD is produced using the catalytic cracking and hydrogenation method, environmental impacts could be reduced by 71% and 50% compared with those in S1-short 390 391and S1-long, respectively. Therefore, it was concluded that a shift from FAME-type BDFs to 392HBD in the future would be effective in reducing environmental impacts, including not only global warming but also fossil fuel consumption, urban air pollution, and acidification. 393

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#### **397 3.4. Uncertainty analysis**

Some parameters include uncertainties, which must therefore be considered in the LCA. Clavreul et al. (2012) reviewed and categorized uncertainties in LCAs for waste management systems into three uncertainties using the framework introduced by Huijbregts et al. (1998), one of which is parameter uncertainty. In this study, uncertainty analysis was conducted for the four parameters in Table 8, which strongly influenced the results.

Electricity was the largest energy source, followed by diesel fuel. The range was taken as the minimum to maximum EFs of electricity consumption during 5 years (FY2008–2012). The default value was accordingly the maximum value in 5 years.

The efficiency of energy recovery is one of the fundamental factors for determining the 406environmental performance of incineration processes (Gentil et al., 2010). The limitations 407408 imposed by technical issues or treatment capacity mean that an incineration facility might not 409 include an electricity production facility. In Japan, approximately 310 of 1,100 incineration facilities include electricity production facilities, and electricity production efficiency was 41011.7% as of FY2011 (MOE, 2013a). The Japanese Ministry of the Environment (MOE, 411 2013b) aims to achieve 21% average electricity production efficiency for incineration 412facilities that will be constructed during FY2013-2017. Therefore, 0% (no electricity 413production facility) and 20% were considered as minimum and maximum values. 414

The EF of NO<sub>x</sub> derived from exhaust gas from household waste collection vehicles 415depends not only on fuel type but various factors: carrying capacity, running speed, etc. The 416actual EF of HBD produced by the catalytic cracking and hydrogenation method will be 417clarified via the proposed fleet running test during the demonstration project by ASTEM. 418Therefore, to establish the tendency of uncertainty derived from the EF, the minimum and 419maximum EFs of NO<sub>x</sub> were determined by comparing the average value reported in some 420previous studies (JPEC, 2005; Koyano et al., 2009). Due to lack of data, it was assumed that 421the EF of HBD equaled that of diesel fuel. 422

The EFs of  $PM_{2.5}$  and  $NO_x$  at an incineration facility depend on the flu gas treatment system installed at the facility (Møller et al., 2011; Shiota et al., 2011). The EF range of  $PM_{2.5}$ was determined using the range of four facilities (Shiota et al., 2011). The EF of  $NO_x$  was not included in uncertainty analysis because  $NO_x$  emissions during incineration had smaller impacts compared with other emissions.

428

### Table 8

429

The results are shown in Fig. 5 and Fig. 6. With regard to the EF of electricity consumption,  $CO_2$  emission factor affected global warming, human health, and integration, whereas the other EFs (NO<sub>x</sub> and SO<sub>x</sub>) had smaller effect. The EFs had significant impact on the ranking between S1-long and S2-short. Electricity production efficiency had a large effect on all impact categories except acidification, and on all endpoints and the integration result. It had larger impact on the results for S1-short and S1-long than in the other scenarios because there was greater use of incineration. It was also indicated that the inclusion of an electricity production facility contributed to reducing environmental impacts in all scenarios. In the absence of an electricity production facility, the integration results in each scenario increased by 30%, 53%, 28%, and 45%, respectively.

The EF of exhaust gas from household waste collection vehicles compliant with the new long-term standard had a bigger impact on the results than vehicles compliant with the new short-term standard. This was because the range (3.32–10.1 g/L) was wider in S1-long and S3-long than those in S1-short (16.2–17.3 g/L) and S2-short (16.2–18.3 g/L).

The EFs of  $PM_{2.5}$  in incineration facilities tended to give wider ranges for the integration results in all scenarios compared with the other three parameters used in the uncertainty analysis. This was because the EF had a large effect on human health. It should be noted that the EF might include sufficient uncertainty to reverse the ranking between the S1-long and S2-short scenarios.

Electricity production efficiency and EF of  $PM_{2.5}$  at incineration facility had larger impacts than the other parameters on result of integration. Therefore, the treatment of waste cooking oil is the significant factor in determining the results. The results of uncertainty analysis indicated that ranking between S1-long and S2-short might be reversed. However, it could be also said that S1-short, which reflects the current situation in Japan, was the worst scenario, and that S3-long was the desired scenario.

This study focused on four parameters for uncertainty analysis. However, there remain further uncertainties, as follows:

- Actual EFs of exhaust gas from household waste collection vehicles using HBD are
   currently unknown. Exhaust gas sampling and analysis will therefore be required during
   the proposed fleet running test.
- The exhaust gas emission standard has been getting stricter than that in the new long-term 461 standard in Japan. Therefore, when those diesel vehicles currently in use are exchanged 462463 for vehicles using HBD that meet stricter performance criteria than the new long-term standard, further reduction of the environmental impacts will be possible. On the other 464hand, blending FAME-type BDF with fossil-derived diesel fuel such as B5 is one solution 465466to extend the future use of diesel vehicles that comply with the new short-term standard. However, in such case, the environmental impacts associated with the exhaust gas 467emissions will not be reduced. 468
- 469 Due to salting during cooking, waste cooking oil might contain higher Cl content than
   470 other feedstock such as unused vegetable oil and animal fats. Cl contents in both waste

471 cooking oil and in cracking oil before hydrogenation are less than 10 ppm. Because the
472 Cl content could contribute to acidification, more detailed Cl behavior will also need to
473 be considered in future studies.

Fig. 5

474

475

476

### 477 **4.** Conclusions

There is a worldwide trend towards stricter control of exhaust emissions from diesel 478vehicles. Certain technical issues have become major impediments to the use of FAME-type 479BDF in diesel vehicles that comply with the new long-term emissions standard. The purpose 480 of this study was to clarify the reduction effects of HBD produced by the catalytic cracking 481 482and hydrogenation method on environmental impacts, as compared with fossil-derived diesel 483fuel or FAME-type BDF. An LCA including uncertainty analysis was conducted to evaluate four environmental impacts: global warming, fossil fuel consumption, urban area air pollution, 484and acidification. Both the operation of diesel vehicles and the treatment of waste cooking oil 485were also considered as functional units. 486

- 487 Our conclusions are as follows:
- If diesel vehicles that comply with the new long-term emissions gas standard are
   commonly used in the future, the benefit of using FAME-type BDF will be relatively
   modest.
- The results including uncertainty analysis indicate that S1-short, which reflects the
   current situation in Japan, was the least optimal scenario, and that S3-long, which
   introduced HBD, was most effective in terms of reducing environmental impact.
- Therefore, a shift from FAME-type BDF to HBD in the future would be more effective in
   reducing total environmental impacts comprising not only global warming but also fossil
   fuel consumption, urban air pollution, and acidification.
- 497

## 498 Abbreviations list

- 499 BDF : biodiesel fuel
- 500 DALY : disability-adjusted life year
- 501 DAP : deposition-oriented acidification potential
- 502 EINES : expected increase in number of extinct species
- 503 FAME : fatty acid methyl esters
- 504 FY : fiscal year
- 505 GHG : greenhouse gas
- 506 GWP : global warming potential
- 507 HBD : hydrogenated biodiesel

- 508 HDRD : hydrogenation derived renewable diesel
- 509 HVO : hydrotreating of vegetable oils
- 510 LCA : life cycle assessment
- 511 NPP : net primary productivity
- 512 PM : particulate matter
- 513 UAF : urban air pollution characterization factor
- 514

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Short torm	Long torm	New	New	Post new
Short-term	Long-term	short-term	long-term	long-term
1994	1997	2003	2005	2009
13 mode	13 mode	13 mode	JC08	JC08
6.8	4.50	3.38	2.0	0.7
0.96	0.25	0.18	0.027	0.010
9.20	7.40	2.22	2.22	2.22
3.80	2.90	0.87	0.17	0.17
	Short-term 1994 13 mode 6.8 0.96 9.20 3.80	Short-term       Long-term         1994       1997         13 mode       13 mode         6.8       4.50         0.96       0.25         9.20       7.40         3.80       2.90	Short-term         Long-term         New short-term           1994         1997         2003           13 mode         13 mode         13 mode           6.8         4.50         3.38           0.96         0.25         0.18           9.20         7.40         2.22           3.80         2.90         0.87	$\begin{array}{ccc} & & & & & & & & & & & & \\ & & & & & & $

Table 1 Exhaust gas emission standards for heavy commercial vehicles in Japan

The heavy commercial vehicle category has a gross vehicle weight > 3.5 tons (> 2.5 tons before 2005).

Implementation dates refer to new vehicle models.

## Table 2 Characteristics of waste cooking oil

Density	0.92	ton/kL
Moisture content	0.2	wt%
Lower heating value	36.6	MJ/kg
Elemental composition		
С	78.1	wt%
Н	11.5	wt%
0	9.8	wt%
S	2.2	ppm

## Table 3 Impact categories, their objects, and units of characterization factors

Impact category	Object	Unit of
		characterization factor
Fossil fuel consumption	Crude oil, Coal, Natural gas	Consumption energy [MJ]
Global warming	CO <sub>2</sub> , CH <sub>4</sub> , N <sub>2</sub> O	GWP [kg-CO <sub>2</sub> eq]
Urban area air pollution	SO <sub>x</sub> , NO <sub>x</sub> , PM <sub>2.5</sub> , PM <sub>10</sub>	UAF [kg-SO <sub>2</sub> eq]
Acidification	$SO_2$ , $NO_x$	DAP [kg-SO <sub>2</sub> eq]

GWP: global warming potential, UAF: urban air pollution characterization factor, DAP: deposition-oriented acidification potential.

For PM2.5 and PM10, characterization factor was not available in LIME2.

	Human health	Social assets	Biodiversity	Primary
Impact category				production
	[DALY]	[Yen]	[EINES]	[NPP]
Fossil fuel consumption		✓	1	1
Global warming	$\checkmark$	1		
Urban area air pollution	1			
Acidification		1		1

## Table 4 Impact categories and corresponding objects of protection

DALY: disability-adjusted life year, Yen: Japanese yen, EINES: expected increase in number of extinct species, NPP: net primary

productivity

## Table 5 Scenario setting

		S1-short	S1-long	S2-short	S3-long
Functional unit 1: Tr	eatment of waste cooking	oil (kL/yr)			
Waste cooking oil	Incineration	1,142	1,142	947	947
from households	FAME-type BDF or	_	_	196	196
	HBD production				
Waste cooking oil	Incineration	1,108	1,108	—	—
from businesses	FAME-type BDF or	_	—	1,108	1,108
	HBD production				
Functional unit 2: Running diesel vehicle for household waste collection (kL/yr)					
Diesel vehicle compl	ied with:	short	long	short	long
Fossil-derived fuel co	onsumption	1,157	1,157	_	502
FAME-type BDF cor	sumption	_	_	1,252	_
HBD consumption		_	_	_	652

short: the new short-term emission gas standard

long: the new long-term emission gas standard

## Table 6 Important parameters for unit processes

Process and parameters		Values	Units	Specific features	References
Collection					
Diesel consumption	Collected as mixed	0.109	L/kL of WCO	Allocated basing on weight	Estimated
	waste				
	Source separation	11.4	L/kL of WCO	From households	Estimated
	Source separation	9.07	L/kL of WCO	From businesses	
Incineration with energy recovery					
Electricity consumption		158	kWh/ton of WCO		NIES, 2008; and calculation
Electricity production efficiency		15.0	%	Produced by steam turbine	Assumed
Emission factor	$CH_4$	0.120	kg/ton of carbon		Yasuda, 1997
	N <sub>2</sub> O	0.565	kg/ton of WCO		MOE, 2009
	SO <sub>x</sub>	150	g/ton of WCO	All sulfur (S) content in waste cooking oil was	Calculation
				assumed to be emitted in this process	
	NO <sub>x</sub>	500	g/ton of WCO		Hirai et al., 2001
	PM <sub>2.5</sub>	798	g/ton of WCO		Shiota, 2011; and calculation
	$PM_{10}$	108	g/ton of WCO		Shiota, 2011; and calculation
	$CO_2$	100	kg/ton of waste		Calculation
			glycerin		
FAME-type BDF production by alkali catalysi.	s method				
Waste cooking oil consumption		1.04	L/L of BDF		Data obtained from Kyoto city
Methanol consumption		0.131	kg/L of BDF		Data obtained from Kyoto city
KOH consumption		7.60	g/L of BDF		Data obtained from Kyoto city

	Paraffin consumption		0.0274	L/L of BDF		Data obtained from Kyoto city
	Electricity consumption		0.184	L/L of BDF		Data obtained from Kyoto city
	Waste glycerin generation		0.396	L/L of BDF		Data obtained from Kyoto city
HBD	production by catalytic cracking and hydrog	genation method				
	Wastewater generation		0.0400	L/L of WCO		Data by demonstration test
	Electricity consumption		0.240	kWh/L of WCO		Data by demonstration test
	H <sub>2</sub> consumption		0.0926	m <sup>3</sup> /L of WCO		Data by demonstration test
	N <sub>2</sub> consumption		0.125	kg/L of WCO	Used as carrier gas	Data by demonstration test
	Heat energy consumption		9.07	MJ/L of WCO	Supplied by high- and low-boiling oils, and offgas	Data by demonstration test
	Products and produced energy	HBD	0.50	L/L of WCO		Data by demonstration test
		High-boiling-point oil	0.04	L/L of WCO		Data by demonstration test
		Low-boiling-point oil	0.16	L/L of WCO		Data by demonstration test
		Offgas	0.13	m <sup>3</sup> /L of WCO		Data by demonstration test
	Emission factor	CH <sub>4</sub>	0.00590	kg/m <sup>3</sup> of wastewater		MOE et al., 2012
Diese	el fuel production					
	Electricity consumption		0.0075	kWh/L		JLCA, 2013
	Emission factor	CO <sub>2</sub>	0.091	g/L		JLCA, 2013
		SO <sub>x</sub>	0.087	g/L		JLCA, 2013
		NO <sub>x</sub>	0.071	g/L		JLCA, 2013
		PM	9.0×10 <sup>-5</sup>	g/L	All PM was regarded as PM <sub>10</sub>	JLCA, 2013
Runn	ing household waste collection vehicle					
	Emission factor	CO <sub>2</sub>	2.62	kg/L		MOE, 2009
	(diesel fuel, new short-term st.)	SO <sub>X</sub>	1.16	g/L	All sulfur (S) content in diesel fuel was assumed to be	Calculation
					emitted in this process	
		NO <sub>X</sub>	18.8	g/L		Emission testing, calculation

	PM <sub>2.5</sub>	2.85	mg/L		Emission testing; Motoshita,
					2008; and calculation
	PM <sub>10</sub>	1.29	mg/L		Emission testing; Motoshita,
					2008; and calculation
Emission factor	CO <sub>2</sub>	2.62	kg/L		MOE, 2009
(diesel fuel, new long-term st.)	SO <sub>X</sub>	1.16	g/L	All sulfur (S) content in diesel fuel was assumed to be	Calculation
				emitted in this process	
	NO <sub>X</sub>	4.29	g/L		Emission testing, calculation
	PM <sub>2.5</sub>	1.08	mg/L		Emission testing; Motoshita,
					2008; and calculation
	PM <sub>10</sub>	4.91	mg/L		Emission testing; Motoshita,
					2008; and calculation
Emission factor	CO <sub>2</sub>	0.133	kg/L	Carbon derived from methanol was considered	Calculation
(FAME-type BDF, new short-term st.)					
	SO <sub>X</sub>	0.00405	g/L	All sulfur (S) content in waste cooking oil was	Calculation
				assumed to be emitted in this process	
	NO <sub>X</sub>	17.3	g/L		Emission testing, calculation
	PM <sub>2.5</sub>	5.92	mg/L		Emission testing; Motoshita,
					2008; and calculation
	$PM_{10}$	26.8	mg/L		Emission testing; Motoshita,
					2008; and calculation
Emission factor	SO <sub>X</sub>	0.00405	g/L	All sulfur (S) content in waste cooking oil was	Calculation
				assumed to be emitted in this process	
(HBD, new long-term st.)	NO <sub>X</sub>	4.29	g/L	Assumed to be equal to the factor in fossil-derived	Calculation
				diesel case	

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	PM <sub>2.5</sub>	1.08 mg/L	Assumed to be equal to the factor in fossil-derived	Assumed
			diesel case	
	PM <sub>10</sub>	4.91 mg/L	Assumed to be equal to the factor in fossil-derived	Assumed
			diesel case	
Common parameter				
Emission factor for electricity	CO <sub>2</sub>	0.570 g/kWh	Weighted average value of 10 commercial utility	Calculation
consumption			companies as of FY2012.	
	SO <sub>x</sub>	0.195 g/kWh	Weighted average value of 10 commercial utility	Calculation
			companies as of FY2012.	
	NO <sub>x</sub>	0.225 g/kWh	Weighted average value of 10 commercial utility	Calculation
			companies as of FY2012.	
	PM <sub>10</sub>	0.0117 g/kWh	Weighted average value of 10 commercial utility	Calculation
			companies as of FY2012.	

WCO: waste cooking oil.

		S1-short	S1-long	S2-short	S3-long
Human health	[DALY]	1.10	0.60	0.69	0.32
Social assets	[Million Yen]	5.08	3.64	2.02	1.43
Biodiversity	$[\times 10^{-10}$ EINES]	-27.4	-27.4	-9.87	-8.40
Primary Production	$[\times 10^3 \text{ NPP}]$	2.49	-1.51	4.21	0.48

## Table 7 Summary of damage assessment

	Min.	Default	Max.
A) EFs: electricity consumption (g/kWh)			
$CO_2$	0.412	0.570	0.570
SO <sub>x</sub>	0.138	0.195	0.195
NO <sub>x</sub>	0.170	0.225	0.225
B) Electricity production efficiency at incinerat	ion facility		
	0%	15%	20%
C) EF of NO <sub>x</sub> : exhaust gas from household was	te collection	n vehicle (g/I	L)
Diesel, the new short-term st.	16.2	18.8	-
Diesel, the new long-term st.	3.32	4.29	10.1
FAME-type BDF, the new short-term st.	16.2	17.3	-
HBD, the new long-term st.	3.32	4.29	10.1
D) EF of PM <sub>2.5</sub> : incineration facility (mg/ton of	WCO)		
	37.2	798	1999

## Table 8 Parameters considered in uncertainty analysis

EF: emission factor. WCO: waste cooking oil.



### Fig. 1 System flow of each scenario

White boxes: processes, black boxes: products and energy, dotted white boxes: excluded processes.

#### **TTT**Collection

Diesel fuel production

- BDF (HBD) production
- Runnning household waste collection vehicle (BDF)
- Surplus low-boiling point oil supply



## (a) Global warming



(c) Urban area air pollution

## Fig. 2 Results of characterization

- $\square$ Incineration
- BDF (FAME) prodcution
- Runnning household waste collection vehicle (diesel fuel)
- Energy recovery (incineration)

-D-Total



(b) Fossil fuel consumption



### (d) Acidification







(c) Biodiversity



## Fig. 3 Results of damage assessment

Point sources: emissions from facilities, non-point sources: emissions from diesel vehicles.



## Fig. 4 Integrated environmental impact of each scenario

WCO: waste cooking oil, Short-term st.: new short-term emissions gas standard, Long-term st.: new long-term emissions gas standard.



Fig. 5 Results of uncertainty analysis (Characterization) Uncertainties are shown as a range (Min.-Max.).

#### A) EFs: electricity consumption

- Electricity production efficiency at incineration facility
- C) EF of NO<sub>x</sub>: exhaust gas from household waste collection vehicle
- D) EF of PM<sub>2.5</sub>: incineration facility



**Fig. 6 Results of uncertainty analysis (Damage assessment and integration)** Uncertainties are shown as a range (Min.–Max.).

### <Electronic supplementary material>

Life Cycle Assessment of Hydrogenated Biodiesel Production from Waste Cooking Oil Using the Catalytic Cracking and Hydrogenation Method Waste Management, http://www.journals.elsevier.com/waste-management/

Junya YANO, Tatsuki AOKI, Kazuo NAKAMURA, Kazuo YAMADA, Shin-ichi SAKAI

Environment Preservation Research Center, Kyoto University, Yoshida-Honmachi, Sakyo-ku, Kyoto 606-8501, Japan E-mail: yano@eprc.kyoto-u.ac.jp

Table 1 Characteristics of Tossil-derived diesel fuel and BDFs					
	Fossil-derived diesel fuel	FAME	HBD	HVO	
	JIS2	Produced at Kyoto city	Produced in the demonstration project at Kyoto city* (ASTEM, 2013)	NExBTL (Neste Oil, 2014)	
Density at 15°C (kg/L)	0.823	0.884	0.844	0.77-0.79	
Kinetic viscosity at 40°C (-)	4.3	4.59	3.2	2.0-4.0	
Flash point (°C)	-	135	50	> 61	
Cetane index	56.3	52.5	51.7	> 70.0	
90% distillation point (°C)	329	-	360	< 320 (95% distillation)	
Oxidation stability (Rancimat method)	>48	5.9	> 36	< 25	
Pour point (°C)	-	-2.5	-13.1	-	
Clogging point (°C)	-	-5	-4	-	
Lower heating value	44.1	37.2	42.6	44.1	
(MJ/kg)					
C (wt%)	86.0	77.1	85.9	-	
H (wt%)	12.5	11.9	12.6	-	
O (wt%)	< 0.5	10.8	< 0.5	-	

## Table 1 Characteristics of fossil-derived diesel fuel and BDFs

\*Produced by the catalytic cracking and hydrogenation method

Table 2 Characteristics of waste glycerin					
Moisture content	22.0	wt%			
Glycerin	47.3	wt%			
Methanol	5.2	wt%			
Potassium (K)	3.2	wt%			
Oily fraction	22.4	wt%			
Elemental composition					
Biomass-based carbon (C)	93.0	wt%			
Fossil-derived carbon (C)	7.0	wt%			

## References

ASTEM, 2013. Report on R&D towards the utilization of 2nd generation biodiesel suitable for vehicles. Kyoto, Japan (in Japanese).

Neste Oil, 2014. Hydrotreated vegetable oil (HVO) – premium renewable biofuel for diesel engines.