CO₂ emission reduction for Japanese petrochemicals

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Abstract

Energy efficiency in the Japanese industry is one of the highest in the world. As a consequence, reduction of CO₂ emissions is considered to be difficult and costly. However little attention has been paid as of yet to changes related to so-called non-energy use of fossil fuels. The analysis in this paper suggests that a large number of options exist for emission reduction in the Japanese petrochemical industry. This includes the introduction of biomass feedstocks, the introduction of new catalytic production processes, and changes in waste handling. The use of bioplastics and the use of CO₂ feedstocks seem costly options for GHG emission reduction that should not be applied on the short term. Japanese GHG emissions can be reduced by 7.7% if the optimal set of emission mitigation options is applied. About 60 Mt emission reduction (4.9%) can be achieved by changes on the supply side, another 35 Mt emission reduction (2.8%) can be achieved by changes in waste management. While changes in waste management can be implemented before 2010, biomass introduction on the supply side will probably require a longer lead-time. About half of the emission reduction is cost–effective, but will require further technology development. The other half can be achieved at a cost level of 10,000 yen/t CO₂ (80 US$/t CO₂). The latter part is based on proven technology that is currently not cost–effective. © 2002 Elsevier Science Ltd. All rights reserved.

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1. Introduction

At the third conference of the parties (COP3) of the United Nations Framework Convention on Climate Change (UNFCCC) in Kyoto the Japanese government has committed itself to a 6% reduction of greenhouse gas (GHG) emissions in the period 2008–2012, compared to the emissions in 1990 [1]. The ratification of the Kyoto protocol is uncertain, but even more significant emission reduction targets can be expected on the longer term, if climate change proves to be a real threat. Ninety percent of the Japanese GHG emissions are carbon dioxide (CO₂) emissions [2], the bulk of which is related to the use of fossil fuels. Approximately, half of the CO₂ emissions are emitted by industry. As a consequence, any significant emission reduction implies also emission reduction in industry. However the reduction of industrial CO₂ emissions is a difficult task. The Japanese industrial energy efficiency is reputedly one of the highest in the world, see, for example, [3]. This fact has often been quoted as a main problem why any significant emission reduction cannot be achieved on the short or medium term [4]. However energy efficiency is not the only strategy to achieve CO₂ emission reduction. For example significant quantities of fossil fuel are used as feedstocks, a non-energy application. The petrochemical industry produces plastics, solvents, resins and other synthetic organic materials from fossil fuel feedstocks such as naphtha and liquefied petroleum gas (LPG). An overview of energy and material flows is provided in Fig. 1. The industry is largely oil based. Gas, coal and biomass feedstocks are of secondary importance. Fossil fuel feedstocks are converted into intermediates such as ethylene. These intermediates are further processed into products such as plastics. Plastics constitute 60% of the product mix. Plastics are applied in many sectors and different products.

Fig. 1 shows that the petrochemical system is a complex system. Many types of flows must be considered...
For proper analysis of energy and materials use within the system. Also many feedback loops exist. On the supply side return naphtha is fed back from aromatics processing plants to the refineries, 5.1 Mt per year. Energy-by-products from steamcracking amount to 4.5 Mt output. The output of energy by-products represents 30% of the system input of feedstocks (in physical terms). On the demand side, feedbacks exist because waste materials are recycled and used for energy recovery in incineration plants. As a consequence of these feedback loops, a broad system perspective is required for proper assessment of the impact of CO₂ emission mitigation strategies.

In energy terms, the feedstock use represents approximately 60% of the total energy use in the petrochemical industry (based on [5]). Most of the feedstock carbon is ultimately converted into CO₂, e.g. if plastic waste is incinerated. As a consequence a reduction of non-energy use related CO₂ emissions. This could be a feasible strategy to reduce the emissions despite the high industrial energy efficiency. In fact, because of the large fraction of feedstocks in the total fossil energy use, reduction of emissions related to feedstocks is potentially of higher importance than a reduction of energy use. Moreover a reduction of feedstock use can simultaneously reduce other environmental problems. For example materials efficiency can reduce feedstock consumption and can simultaneously reduce waste volumes and the related environmental problems [6]. However these secondary benefits are beyond the scope of this paper. This results in an underestimation of benefits.

The Japanese energy efficiency is reputedly high, while no efforts have been done yet to reduce non-energy use related CO₂ emissions. As a consequence the latter options look more promising. A previous assessment study found an energy efficiency potential of about 6 Mt CO₂ emission reduction at a tax level of 30,000 yen/t CO₂, about 250 US$/t CO₂ [7]. This is a very high tax, indicating that the actual potential is limited. Also analysis of energy efficiency options such as combined heat and power generation (CHP) requires detailed insight into the heat balance of individual petrochemical sites, beyond the scope of this study. Because energy efficiency is not considered the emission reduction potentials in this study represent a lower estimate.

The following three questions will be answered in this paper:

—Which options exist to reduce GHG emissions in the life cycle of petrochemicals in Japan in relation to non-energy use?
—What is the potential contribution of these options in terms of GHG emission reduction?
—What is the cost-effectiveness of these options?
2. Methodological issues

2.1. System boundaries

Changes in one part of the petrochemical life cycle may have consequences for the CO₂ reduction potentials in other parts of the life cycle. For example, the introduction of biomass feedstocks will reduce the CO₂-benefits of recycling. As a consequence an integrated life cycle approach is required for proper assessment, considering all options simultaneously.

Widespread co-production in the petrochemical industry complicates life cycle assessment (LCA). For example, the emissions from naphtha steam cracking must be allocated to a broad array of products. Different allocation schemes can result in different conclusions regarding the optimal policies. In this analysis, the “equivalent output” approach has been applied. An example is shown in Fig. 2. In both cases shown, the system boundaries have been chosen in such a way that the total output of the naphtha cracker equals the total output of the methanol to olefins (MTO) process plus the output of energy carriers and materials from the refinery. As a consequence, both systems supply the same service ‘equivalent output’ and can be compared.

The spatial dimension of environmental impacts is generally neglected in LCA studies. However, location is very important in the environmental policy reality. For example GHG policy targets have been formulated on the country level. The life cycle of a material may extend over several countries. The issue is of special relevance in relation to CO₂ accounting for petrochemicals because the bulk of the carbon is contained in the materials (so-called feedstock carbon, see Table 1). This carbon is only emitted as CO₂ in case the products are incinerated (which may be in a different country than where the production took place). Basically a global perspective is required for proper assessment, see [8]. However such an analysis is beyond the scope of this study. Instead, the full life cycle impacts are considered.

2.2. GHG accounting

Apart from CO₂, five other types of greenhouse gases are accounted for in the Kyoto protocol. The most important ones (apart from CO₂) are methane (CH₄) and nitrous oxide (N₂O). Methane emissions in the life cycle of petrochemicals may occur in oil and gas production and transportation. Nitrous oxide emissions occur in the production of adipic acid. However both types of emissions can be reduced comparatively easily, the real policy problem is the long-term reduction of CO₂ emissions. For this reason, the following analysis focuses on petrochemical CO₂ emissions. In the case of biomass-based production, N₂O emissions from fertiliser use have been accounted for, because the reduction of these emissions is more problematic [9].

The CO₂ accounting for petrochemicals in this analysis follows the Intergovernmental Panel on Climate Change (IPCC) guidelines [10] in the sense that carbon storage is accounted for long life products, and emissions are accounted for incineration of these products. However the IPCC guidelines use the category non-energy use in the energy statistics as a basis to estimate carbon flows, while the current study is based on actual materials flows. The difference can be substantial: the current Japanese national GHG account overestimates carbon storage by 23 Mt (an overestimation of 20%, 1.9% of the Japanese CO₂ emissions) [11]. The reason is partial energy use of fossil fuels that are categorized as non-energy use, and the neglect of emissions during use of short-life products such as solvents and detergents. The accounting in this paper is considered to be more accurate than the IPCC approach [11]. Because of the accounting differences, results of life cycle analysis studies do no necessarily correspond with the results in
Table 2
Cost assumptions for resources per unit of energy (excluding taxes) [12]

<table>
<thead>
<tr>
<th>Commodity</th>
<th>Price (yen/GJ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Naphtha</td>
<td>350</td>
</tr>
<tr>
<td>Gas oil</td>
<td>300</td>
</tr>
<tr>
<td>Gasoline</td>
<td>350</td>
</tr>
<tr>
<td>Heavy oil</td>
<td>275</td>
</tr>
<tr>
<td>Ethane</td>
<td>400</td>
</tr>
<tr>
<td>LPG</td>
<td>400</td>
</tr>
<tr>
<td>Natural gas</td>
<td>300</td>
</tr>
<tr>
<td>Wood chips (SE Asia)</td>
<td>200</td>
</tr>
<tr>
<td>Wood chips (Japan)</td>
<td>400</td>
</tr>
<tr>
<td>Electricity</td>
<td>1250</td>
</tr>
<tr>
<td>Process heat</td>
<td>500</td>
</tr>
</tbody>
</table>

In case the current national CO2 accounting method is followed, it is recommended to adjust the national accounting scheme.

2.3. Cost accounting

The cost–effectiveness of emission mitigation strategies depends on the resource prices and the capital costs. In this type of capital-intensive operations, labour costs are generally of secondary importance. A 12% discount rate was applied as an approximation of the rate of return of industrial investment projects. A life span of 25 years has been assumed for all capital investments. Based on these assumptions, investment costs have been annualised using an annuity factor of 0.13. Operation and maintenance costs have been set at 4% of the investment costs. Regarding physical process inputs, prices for energy and feedstocks are listed in Table 2, based on [12].

Investment costs were taken from the Western European MATTER model database [13] with a 10% increase because of the higher investment costs in Japan compared to Western Europe (land costs and materials costs are higher). Operations and maintenance cost were also taken from the MATTER database, given the similar labour costs in both regions [14]. Prices for current production processes were also taken from MARKAL MATTER database. For a number of petrochemicals the current market price has been used as a reference value for calculation of cost–effectiveness (Table 3, based on [15]). Usually market prices are higher than production costs, so this results in an overestimation of the cost–effectiveness. However given the fierce competition, the gap between production costs and market prices is small.

2.4. Technical and physical constraints

The potential for emission reduction of a specific option is a function of the emission reduction per unit of activity and the potential activity. The potential activity depends on the market volume and the substitution potential. Not all markets can be substituted completely, e.g. because of technical constraints. For example, the quality of re-extruded plastics is lower than for virgin material. Plastic waste injection in blast furnaces is limited by the technical minimum coke requirements and the chlorine content. Another type of constraint is the resource availability. For example, biomass production is limited by the land availability and the yield per hectare. Liquefied Petroleum Gas (LPG) availability is limited by the structure of fossil fuel supply and the composition of the natural gas resources. Estimates for resource availability are given in Table 4, based on market studies and resource availability data. A more detailed discussion of biomass supply potentials is provided in Section 3.1.1. Based on the constraints per option and based on the CO2 impact per option, it is possible to calculate the potential contribution of an option to GHG emission reduction in Japan.

3. Characterisation of GHG emission reduction strategies

In order to reduce GHG emissions related to non-energy use in this particular sector, a limited number of options exist [6]:

- Substitute petrochemical oil feedstocks;
- Substitute synthetic organic materials;
- Increase the efficiency of synthetic organic materials use;
- Recycle waste materials;
- Energy recovery from waste materials.

Materials substitution (e.g. use of steel and wood

Table 3
Market price of chemicals used for the assessment

<table>
<thead>
<tr>
<th>Commodity</th>
<th>Price (yen/t)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ethylene</td>
<td>60,000</td>
</tr>
<tr>
<td>Propylene</td>
<td>50,000</td>
</tr>
<tr>
<td>Butadiene</td>
<td>45,000</td>
</tr>
<tr>
<td>BTX</td>
<td>30,000</td>
</tr>
<tr>
<td>Phenol</td>
<td>90,000</td>
</tr>
<tr>
<td>Butanol</td>
<td>110,000</td>
</tr>
</tbody>
</table>

Table 4
Resource availability limitations, 2010–2020

<table>
<thead>
<tr>
<th>Type</th>
<th>Quantity (Mt/yr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Biomass</td>
<td>5–50</td>
</tr>
<tr>
<td>Plastic waste</td>
<td>10</td>
</tr>
<tr>
<td>Propane/butane</td>
<td>10</td>
</tr>
<tr>
<td>Ethane</td>
<td>5</td>
</tr>
</tbody>
</table>
instead of plastics) and increased efficiency of materials use will not be discussed in more detail in this paper. Especially the assessment of materials efficiency potentials requires detailed insight into the materials/product combinations and the industrial design potentials to reduce materials consumption. Such an analysis is beyond the scope of this study. However economic model simulations suggest a materials efficiency potential of up to 30% [6,16]. The topic is beyond the scope of this study. As a consequence the emission reduction potential is underestimated.

3.1. Feedstock substitution

Three alternatives exist for oil feedstocks that may reduce GHG emissions:

- Biomass feedstocks (e.g. wood, kitchen waste);
- CO₂ feedstocks;
- New fossil fuel feedstocks.

3.1.1. Biomass feedstocks

Biomass (consisting of plant and animal derived feedstocks) is the only renewable carbon source. As a consequence it is CO₂ neutral: carbon storage during growth equals carbon emissions during use and waste incineration. Introduction of biomass as oil substitute reduces CO₂ emissions. Currently biomass feedstocks constitute the basis for the so-called oleochemistry: a significant part of the surfactants and detergents are derived from natural oils such as palm oil. Ethylene production from ethanol (a biomass based product) has been used on a commercial scale in the past, but has lost its relevance because of the high production costs compared to production based on fossil fuels [17]. Acetic acid is still produced from wood and from ethanol, but mainly for food purposes. These are only some examples where biomass is used. However, biomass represents currently less than 5% of the feedstock used for synthetic organic products worldwide [18].

The problem regarding biomass strategies, in the case of Japan, is the high population density, which limits the possibilities to produce large quantities of biomass for non-food purposes. Japanese primary energy use amounts to 23 EJ per year, its biomass surplus (both primary and secondary sources) amounted to 2 EJ in 1990 [19]. However the bulk of this biomass consists of wood and forestry residues from the rugged Japanese mountain areas, a resource with high extraction costs. Estimates for future global biomass surplus availability range from 50 to 450 EJ in 2050 [20]. Biomass in other world regions may become available at 100–200 Y/GJ (free on board fob). In case of large-scale biomass plantations, even intercontinental trade of solid biomass or biomass derived liquid energy carriers may become attractive [21].

As a consequence only import strategies deserve special attention for large-scale biomass introduction in Japan. The import of liquids seems the most attractive strategy. The energy content of biomass liquids is generally lower than the energy content of solid biomass. As a consequence transportation of liquids results in lower environmental impacts and lower transportation costs.

Biomass can enter the system at three levels. These three levels are discussed separately:

- Feedstocks for existing basic chemicals from steam cracking, e.g. for ethylene production;
- Feedstocks for intermediate chemicals, e.g. for butanol production;
- Biopolymers that substitute polymers such as polyethylene.

3.1.1.1. Substitution of basic chemicals. Three types of liquid biomass feedstocks have been considered as substitutes for existing basic chemicals:

- Ethanol;
- Methanol;
- HTU oil (Hydro Thermal Upgrading).

The selection is not exhaustive. For example pyrolysis oils have also received a lot of attention during the last decade, see e.g. [22]. However the quality of the liquid product is insufficient and product upgrading proves to be costly. As a consequence the emphasis in development has shifted. Fischer–Tropsch (FT) technology, based on gasification is another alternative [23]. However development is largely driven by technology development in the transportation sector, which represents the bulk of the oil market. Fuel cells are widely considered the car engines of the future. It is difficult to reform FT diesel into a suitable input for fuel cells. As a consequence methanol is preferred.

3.1.1.1.1. Ethanol production and ethanol dehydrogenation

Ethanol production for alcoholic beverages is a well-established industry. Some ethanol is used as transportation fuel (e.g. sugar cane based ethanol in Brazil and corn based ethanol in the United States). This production is based on sugars and starch. However recent R&D has resulted in new feedstock options. The main advances relate to the fermentation of cellulose and hemicellulose, where formerly only free sugars and starch could be fermented. As a consequence of these developments, it will be possible to ferment wood and grass (so-called lignocellulose crops). The advantage is the much higher yield (or lower cost) of these biomass types compared to sugar
and starch crops. The production of ethanol from cellulose is an established technology, the production of ethanol from hemicellulose is not yet proven on a commercial scale, but it is forecast to become available in a couple of years.

The ethanol production from lignocellulose can be split into [24]:

1. Pre-treatment to mobilise the biomass constituents;
2. Hydrolysis to convert biomass constituents into free sugars;
3. Fermentation of the sugars to produce ethanol;
4. Distillation of the water/ethanol mixture.

Pre-treatment consists of a combination of chipping, milling and grinding and subsequent chemical treatment. Hydrolysis can be split into chemical hydrolysis, using sulphuric acid, and enzymatic hydrolysis. The enzymatic hydrolysis is expected to yield considerably more ethanol than the acid hydrolysis, but requires further development. The main fossil energy consumption in ethanol production is currently related to the distillation process. There is ample room for a reduction of energy requirements based on introduction of molecular sieves and vacuum distillation. The ethanol can be dehydrated to ethylene. This process has been applied in the past but has been abandoned because of the high cost of ethanol feedstocks [17]. Ethanol may become much cheaper in the future because of cheaper lignocellulose feedstocks can be used and processing costs are reduced.

One Japanese company is currently selling a gasoline substitute containing about 50% ethanol [25]. The fuel production costs amount to 79 yen/litre, compared to about 30–40 yen for a litre of gasoline. However this product is currently not taxed (contrary to gasoline). This example shows that ethanol can be competitive, even in the current market conditions. If ethanol is attractive in the gasoline market, it may also become attractive as a petrochemical feedstock, allowing a gradual switch to sustainable petrochemicals.

3.1.1.1.2. Methanol production and methanol to olefins

The methanol production process is based on gasification of carbon containing feedstocks and subsequent methanol synthesis [26]. Current methanol production is largely based on gas feedstocks because gas poses the cheapest alternative. The so-called Batelle reactor design for methanol production from biomass [27] is currently being tested on a pilot plant scale in the United States.

Methanol to olefins (MTO) produces ethylene and propylene via catalytic dehydration of methanol. Experiments show that the product mix depends on the catalyst and the reaction temperature [18]. The feasibility of MTO has been proven on the pilot plant scale [28], but so far no commercial plants have been built because of unfavourable process economics.

3.1.1.1.3. HTU oil production and HTU oil pyrolysis

HTU is an acronym for HydroThermal Upgrading. Biomass (wet chips or a slurry) is treated with water in a mixed reactor at temperatures of 300–350°C at pressures of 120 to 180 bar for 5–10 min. This type of conversion process is called hydrothermal liquefaction. The oxygen content of the biomass is reduced from 40 to about 10–15% (w/w) by the selective formation of CO2. Under these conditions, an organic liquid (or “biocrude”) is formed, which resembles crude oil. The biocrude can be upgraded to a naphtha-like product by removal of the remaining oxygen (by catalytic hydrodeoxygenation, using hydrogen). The upgraded product can be used as a feedstock for the production of chemicals based on the existing steamcracking technology [29,30]. The HTU process can take different feedstocks, and could be an attractive option for feedstocks with a high water content such as kitchen waste or waste water treatment sludge, since no drying is required.

A pilot plant for HTU oil production was opened in The Netherlands in 1999. However this plant is currently not operating because Shell, one of the funding parties, has withdrawn from the project. However, the process developers claim they can produce an oil product at a price that is competitive with current crude oil prices [30]. Previous attempts in the in the United States in the 70s and 80s to develop a similar process have failed because of problems with the biomass injection system and because of insufficient understanding of the reaction mechanisms and the process dynamics. The HTU process and its predecessors are based on the use of water as a solvent. Other organic solvents may be more promising (solvolysis) because they can reach similar temperatures in a liquid state at atmospheric pressures, resulting in significantly reduced process equipment costs [31]. More research is required before this process will become operational.

3.1.1.2. Substitution of intermediates. A large number of intermediate chemicals can be produced from biomass. Important technologies are fermentation (for alcohols) and flash pyrolysis (for aromatics and olefins). Three examples are discussed in more detail: butadiene, butanol and phenol. These options have been selected because they seem promising [32].

3.1.1.2.1. Butadiene from wood flash pyrolysis

Butadiene is an important intermediate in the production of different types of synthetic rubber. It is currently a by-product from steam cracking and it is derived from refineries. However it could also be produced from wood via flash pyrolysis. Earlier analyses suggest this would be a cost-effective strategy.

3.1.1.2.2. Butanol and acetone from fermentation

Butanol and acetone can be co-produced by fermen-
tation of glucose, e.g. from corn. The classical fermentation route involves large scale anaerobic batch fermentation followed by distillation. The yields amount to 23% butanol, 9% acetone and 2% ethanol on dry weight basis.

3.1.1.2. Biopol

Bioplastics consist of a mixture of polyhydroxybutyrate (PHB) and polyhydroxyvalerate (PHV), produced by micro-organisms. PHB is produced when glucose is used as substrate, PHV is produced when valerate is used as substrate. One ton of biopol requires three to four tons of glucose fed to the bacteria [35]. Certain bacteria strains produce such high concentrations of PHB/PHV that they can be extruded to plastic without prior purification. The properties of Biopol are very close to PP properties, with the notable difference that it is biodegradable. Glucose can be produced from starch (e.g. from rice or potatoes). The GHG emission equals 0.75 t CO₂ eq/t biopol. The costs of this material are rather high: approximately 1 million yen per ton (five to ten times the price of oil based plastics). Assuming a PP substi-

3.1.2. CO₂ feedstocks

Carbon dioxide from flue gases and from natural gas production, poses an alternative carbon source. Especially in Japan, extensive research efforts have focused on this strategy. Different products are being investigated, only one pathway is discussed in more detail. The synthesis of methanol via CO₂ hydrogenation is considered one of the most promising processes for the fixation and utilisation of CO₂ [36]. In combination with the MTO process (see above), it is possible to produce petrochemicals via this route.

The production of the hydrogen for the hydrogenation process requires significant amounts of energy. Hydrogen production accounts for 93% of the total energy required, CO₂ separation and liquefaction accounts for 6%. Total energy requirement is 28 GJ of electricity per ton of methanol. The source of the energy for hydrogen production is crucial for the resulting CO₂ balance. In case of a CO₂ free electricity source, the net balance is a reduction of GHG emissions by 1.38 t/t methanol. However in case of an average emission for electricity production of 0.1 CO₂/t, the net balance is an emission of 1.42 t CO₂/t.

The emission for methanol production from natural gas (the current production process) is approx. 0.22 t CO₂/t (feedstock carbon storage is accounted for in both cases). Obviously the use of CO₂ feedstock will be a costly strategy for GHG emission reduction, because CO₂-free electricity is a costly energy source. Given this conclusion, this option is not analysed in more detail in this paper.

3.1.3. New fossil fuel feedstocks

The petrochemical industry in the United States and Canada is largely based on natural gas derived ethane. A gas pipeline projects is currently evaluated that will bring 15–20 Billion Cubic Metres (BCM) Russian gas to Japan [37]. The Sachalin gas is connected with oil reserves. Such gas often contains significant quantities of ethane, propane and butane that could be used for petrochemical production. Assuming a 5% content of these components (based in North Sea gas composition data), the gas would contain 1–1.5 Mt of resources that could be used for petrochemicals. Additional gas derived feedstocks could be bought from gas production sites elsewhere (e.g. from Canada). Pro-
3.1.3.1. Oxidative coupling of natural gas. Natural gas can be used for production of C2 hydrocarbons. Rising oil prices in the 70s and 80s resulted in intense research concerning oxidative coupling of methane, in order to substitute costly oil derived feedstocks. In the center of attention is the development of catalysts that favor the reaction of CH4 and O2 to ethylene, based on the so-called oxidative coupling reaction mechanism [38]. Other oxygen containing compounds such as CO and CO2 are undesirable by-products. The reaction takes place at atmospheric pressures and temperatures of 750–900°C. At 20–30% methane conversion, selectivities to C2 of 70–80% are currently achieved (39% selectivity towards C2H4 if the consecutive cracking of product ethane is included). Considerable amounts of steam are generated as by-product. Research is aimed at increasing both conversion and selectivity. There are currently no plans for commercialization.

3.1.3.2. Ethane and LPG feedstocks for steamcrackers.

Ethane can be cracked in steamcracking installations to yield a mix of petrochemicals. The technology is well known and widely available. Steamcracking of propane and butane (the main LPG constituents) is also widely applied. It is just a matter of costs whether these options are attractive at this moment. The CO2-impact of such a switch away from naphtha is not clear at first sight because the product mix differs considerably.

3.1.3.3. New production routes, based on catalytic conversion. Probably the most promising development in petrochemical feedstock use is based on the development of new catalytic conversion routes [39]. These strategies aim for the substitution of the production routes via the basic chemicals ethylene and propylene with direct conversion of alkanes (ethane, propane and butane) to petrochemicals such as vinylchloride monomer (VCM), acrylonitrile (AN) and butadiene. Because of the reduction of the number of process operations, the energy savings and the cost savings are considerable. Cost savings up to 30% are reported. All three production routes are currently tested at a pilot plant scale, commercialisation is expected within the next decade.

3.1.3.4. Use of other oil derived feedstocks. Basically all light oil products such as gasoline or diesel oil can be cracked. In reality, their price prevents such applications. However, a trend exists towards heavier feedstocks than naphtha. This trend is driven by the decreasing hydrogen fraction in the crude oil mix and the increasing demand for gasoil in the total product mix. As a consequence, the price gap between light hydrocarbon products and heavy hydrocarbon products is increasing. This is an incentive to switch to the cracking of heavy hydrocarbons. Gasoil cracking is already widely applied, for example in Western Europe. A recent trend is so-called deep catalytic cracking (DCC) using vacuum gasoil and other heavy oil products. The key to this technology is a new type of catalyst, the process plant is similar to the widely fluid used catalytic cracking (FCC) process in existing refineries. The technology has been developed in China and is applied on a commercial scale [39].

3.2. Materials recycling and energy recovery

Materials recycling and energy recovery are treated as one category because the difference is often not clear in the case of petrochemical products. The current practice is energy recovery in waste incinerators, but the efficiency of these installations is low. Japanese waste management can be divided into household waste management (50 Mt/year) and business waste management (agriculture, industry and services, 400 Mt/year). The quantity of household waste was virtually constant during the last decade [40]. The household waste is collected by the municipalities. Eighty percent is incinerated, the remainder is landfilled (8%), recycled (5%) or treated otherwise. The waste collection schemes differ per municipality. Some municipalities collect mixed waste, others collect combustible waste and incombustible waste separately. However the definition of incombustible waste varies. In some municipalities, plastics are part of the incombustible waste, while in other municipalities it is part of the combustible waste. This difference can be explained by different incineration facility characteristics (suitable for combustion of waste with high energy content or not suited).

In 1999 Japan had a total of 1872 waste incinerators with a total capacity of 37 Mt waste per year. Seventy three percent of this capacity (442 incinerators) is of the full continuous type with an average size of 42 kt per year. This is still one order of magnitude below the size of European waste incinerators which is in the range of 200–1000 kt per year. Given the economies of scale, investment costs are 2–4 times higher for 40 kt incinerators in comparison with 200–500 kt incinerators (based on [41]). A sharply increased demand for new incinerators is forecast as new tighter regulations for dioxins go into effect in December 2002. Japan had in the mid 90s the highest dioxin emissions of all industrialized countries. 80% of these emissions originate from waste incinerators [42]. The government has been giving subsidies to incineration plants with a capacity of 350 kt per year and more. As a result interest in large waste incineration facilities is growing. Energy is currently only recovered at part of these installations, and the own energy consumption is considerable and increasing because of new environmental regulations. As a conse-
quencing the average net electricity efficiency is below 5%.

Recycling of materials requires generally less energy than materials production from primary resources. Moreover, the feedstock energy can be saved. That is why the Japanese government is aiming for recycling. A new collection program for PET bottles has been launched recently. Nowadays about 30% of all PET bottles is collected for recycling. Recycling capacity lags behind collection, the bulk is downcycled into fibres [43]. Starting in 2002, the use of coloured bottles is forbidden. This will facilitate the recycling of PET bottles. Approximately 60 kt of polystyrene foam were recycled in 1998 [44]. There is some activity by product manufacturers such as the collection of used bumpers by car dealers, see for example [45], and disassembly of electronic equipment by the electronic equipment manufacturers, e.g. [46]. However the quantities are still limited, less than 100 kt is collected and recycled. Starting in 2001, recycling of household equipment is mandatory. This system is funded by a charge for disposed equipment. In contrast for passenger cars a system is being established where the buyers of new cars pay for the recycling. No numerical recycling targets have been set for plastics.

A specific characteristic of the Japanese waste collection is the use of small plastic bags (10–40 litres), compared to the large waste bins (100–200 litres) that are used in other industrialised countries. Different waste categories are collected on different days of the week. This practice can be attributed to the comparatively small dwellings and the lack of adequate waste storage space in the residential areas, and it increases the waste management costs. Another special feature of the Japanese waste management system is that the costs for collection are largely borne by the municipalities (no unit pricing system). There is a levy on waste disposal bags, but this levy is comparatively low (about 300 yen/t compared to waste treatment costs of 50,000 yen/t). As a consequence there is no incentive for source reduction.

Plastic waste recycling can be split into three categories:

- Back-to-polymer recycling (BTP)
- Back-to-monomer recycling (BTM)
- Back-to-feedstock recycling (BTF)

Four technologies have been selected for further analysis, covering all three categories (re-extrusion, pyrolysis, injection into blast furnaces and hydrogenation). A number of other options exist such as solvent separation, hydrogen production from waste plastics, production of polyols etc. However, they are not considered in more detail because that their commercialisation seems further away.

Also three types of energy recovery options will be discussed:

- Incineration in cement kilns;
- Gasification;
- Grate firing.

3.2.1. Re-extrusion (BTP)

Re-extrusion technology is the most widely applied recycling technology. The plastic is ground and extruded. The material output quality depends, to a large extent, on the waste input quality. Additives can improve the product quality. If mixed plastics are used as inputs, the resulting material will only be suitable for a limited number of applications. For example the bulk of Polyethylene Terephthalate (PET) is downcycled from bottles into fibres. Recycling into bottles is limited due to regulations. However, smart technologies make it possible to use recycled materials even within this sector (e.g. multi layer PET bottles). In other sectors the use of recycled materials is less problematic (e.g. for pallets, crates or carrier bags). The environmental benefits of re-extrusion depend on the specific recycling scheme [47].

3.2.2. Pyrolysis of mixed plastic waste (BTM)

Pyrolysis is the process where mixed plastic waste is heated in an oxygen-free atmosphere. At a temperature of several hundred degrees Celsius, the hydrocarbons decompose to yield a mixture of solid, liquid, and gaseous products. Product composition depends on temperature and pressure. The higher the temperature, the more gaseous products are formed. A pyrolysis option that is currently being developed in Japan is the conversion of waste plastics in existing coke ovens in the iron and steel industry [48]. First the waste is sorted manually, followed by separation of PVC based on differences in density. The resulting plastic waste is processed into an RDF type pellet (Refuse Derived Fuel). This pellet is used as feedstock for the coke ovens. The coke oven product mix consists of 40% cokes oven gas, 20% cokes and 40% oil. The energy efficiency of a coke oven is approx. 82%, if the gas use for firing is accounted for. The advantage compared to direct injection of waste plastic into blast furnaces (see below) is higher value products and reduced interference with the sensitive blast furnace operation.

3.2.3. Injection into blast furnaces

Plastic waste is injected in blast furnaces to serve as substitute for coke and coal. Mixed plastic waste injection into blast furnaces is currently applied at commercial scale in Germany [49,50]. Also in Japan this technology is applied at NKK's Keihin plant on a scale of 30 kt annually [51]. Before plastic waste is added the chlorine content must be reduced in case PVC is present in the plastic mix, because chlorine can affect the blast furnace iron product quality. Packaging waste is virtually PVC waste free, but demolition waste and shredder
waste may contain significant amounts of chlorine. PWMI and NKK are currently testing a rotary kiln for chlorine removal [52]. Chlorine removal costs 20,000–30,000 yen/t plastic waste. Coal and coke are replaced on a thermal par basis. In regulatory terms this process is considered a recycling process because part of the energy use in the blast furnace is accounted for by the chemical reduction of iron ore. However in practice the CO2 impacts of, for example, combustion in cement kilns is the same. The different categorization will make a difference from a policy point of view because recycling is considered to be superior to incineration.

3.2.4. Hydrogenation VEBA process (BTF)

Mixed plastic waste can be treated with hydrogen to produce a naphtha-like product and a hydrogenation residue that can be used in coke production. A commercial scale plant is operated in Germany. The technology can be characterised as a thermal hydrocracking/hydrogenation process. The reactions take place in a liquid phase reactor and a gas phase reactor at temperatures of 400–450°C and a pressure of up to 250 bar. At present the main problem is the feeding of plastics into the reactor. Data for hydrogen consumption are still uncertain, but they would seem to be significantly higher compared to what might be expected on the basis of the plastics chemical structure [53]. At this moment there is no interest in this technology in Japan. However given the lack of indigenous oil resources and the relative scarcity of oil in comparison to coal, this option may become attractive on the longer term.

3.2.5. Plastic waste incineration in cement kilns

The production of cement clinker offers multiple possibilities for the utilization of alternative raw material and fuel due to high temperature technical processing and the basic chemical environment which binds the chlorine. Old tires, shredder scraps, municipal solid waste (MSW) and plastic waste are widely used. Up to 50% of the fuel energy from MSW could be realized without a deterioration of the clinker quality and maintaining the energy efficiency [54]. Plastic waste is already applied in Japan, for example in the Tokuyama cement kiln [52]. Current research is aiming for pre-treatment chlorine removal from PVC containing plastic waste.

3.2.6. Gasification

Gasification technology receives a lot of attention because of high energy conversion efficiencies and low emissions. A plant for treatment of 165 kt waste per year has been built in Aomori in 1999 [52]. This plant is based on Ebara’s Gasification and Ash-melting combustion technology. This plant treats shredder dust and municipal sewage sludge. Together with Ube industries, Ebara is working on higher efficiency systems for production of ammonia from (plastic) waste, based on two-staged pressurized gasification [55]. However given the relative ease of transportation of nitrogen fertilizers (the main ammonia product) and given the abundant availability of natural gas for ammonia production in other world regions, this option seems less attractive. As a consequence only gasification and subsequent electricity production is analysed in more detail. Gasification units for energy recovery have been built in Germany, Sweden and in Italy [56]. Thermoselect, one prominent gasification technology, has been licensed by Mitsubishi.

Given the fact that the complexity of these new processes is much higher than the complexity of conventional waste incineration systems (requiring more process steps), the energy efficiency is in many cases not higher than for current state-of-the-art waste incineration systems. For example, an optimised Thermoselect process has an electric efficiency of only 20.4%, similar to current waste incinerators [6]. However lower costs, reduced toxic emissions and less residues can make such new technologies an attractive alternative.

3.2.7. Grate firing

Grate firing in combination with a steam boiler working under moderate steam conditions is the most commonly used technique to recover energy from municipal solid waste (MSW). Due to the characteristics of MSW (low energy content) and due to the corrosive nature of the off-gases, energy recovery with a high efficiency is hard to achieve. In case of electricity production, the gross efficiency is comparatively low because of the unfavourable steam cycle conditions. Moreover, the internal energy consumption is comparatively high (for cleaning of flue gasses, etc.). These two factors result in a net efficiency that is much lower than the usual electrical efficiencies in power production. Current grate firing systems achieve a net efficiency of 20–22%, compared to 55–60% for new gas fired combined cycles.

A combination with a gas turbine for steam reheating can increase the efficiency of the system. Three plants are already in operation in the cities of Takasaki, Sakai and Kitakyushu. The net power generation efficiency (for the waste heat part) is increased to approx. 25%. However, the efficiency of the system depends on the size of the gas turbine. In case of a 200 kt/yr incinerator with a 15 MW gas turbine for reheating and reburning (38% of the total heat input is gas). The efficiency of the total system is 29% (compared to 26.8% for two stand-alone systems). In case of a 40 MW gas turbine (61% of the total heat input is gas), the efficiency of the total system is 36.5% (compared to 34.9% for two stand-alone systems) [57]. In conclusion, some efficiency improvements are possible in waste incinerators, but the efficiencies remain low compared to other electricity generation schemes. As a consequence it makes sense to use other waste treatment systems (see below).
4. Static life cycle analysis of the CO₂ impacts of emission reduction strategies

4.1. Production side options

The GHG balance for the four ethylene production options is shown in Fig. 3. The three improvement options that are based on wood include N₂O emissions from fertiliser use. A transportation distance of 5000 km has been assumed for the liquids, and a transportation energy of 0.2 MJ/t.km. In the case of ethanol production, the lignin residue has been credited at a value of 30 kg CO₂/GJ (approximately half the value for natural gas, in order to account for the low efficiency of heat recovery from lignin because of the high water content).

A number of implicit assumptions determine the outcome of this analysis to a large extent. First of all it is important to define the system boundaries in the comparison: should emissions abroad be accounted. Generally in LCA, no difference is made between national and foreign emissions. However given the national character of the GHG policy targets, the difference is relevant. In the case of ethanol, the lignin by-product use abroad has been credited. This may be valid from a life cycle point of view, but it is not valid in case the impact on the Japanese CO₂ balance is accounted. The value above the y-axis is a better estimate for the impact on Japanese emissions. However in the case of MTO, the credits for LPG arise because the propylene yield of MTO is higher than for naphtha cracking. As a consequence, the propylene recovery from refinery FCC off gases is reduced, and these gases can be used for heating purposes within Japan, so the balance of positive and negative impacts should be considered if the impact on the Japanese CO₂ balance is accounted for. The system boundaries determine in this case which is the better option.

Next, the resource requirements for the three wood based options differ. The ethanol option requires an input of 7.9 t of wood, the MTO option requires an input of 11.8 t wood, and the HTU option requires an input of 9.6 t wood. From a resource efficiency point of view ethanol seems the best option.

Looking at the CO₂ impacts, HTU seems the most attractive option, followed by MTO and by ethanol. The emission reduction is in all cases substantial compared to the naphtha case: up to 80% lower emissions. However one must account for the fact that the products contain carbon. The total carbon content of ethylene, propylene and butadiene of each option is equivalent to 5.5 tons of CO₂. This reduces the relative emission reduction (in the production stage, off course the benefits will occur in the waste incineration stage because the carbon in waste from biomass origin does not need to be accounted for).

The CO₂ balance of the natural gas based alternatives and the oil-based alternatives for basic chemicals production have been analysed in a similar way the biomass alternatives have been analysed. The result is that oxidative coupling can reduce emissions by 0.7 t CO₂/t ethylene, the other options have marginal effects for the CO₂ balance.

Fig. 4 shows the CO₂ savings that can be achieved in case fossil fuel based intermediate chemicals production is substituted with biomass based production. It has been assumed that all energy is generated from biomass in the biomass case (neutral CO₂ balance). The figure shows that the main saving relates to feedstocks. It is imposs-
4.2. Materials recycling and energy recovery

The balance for different types of recycling and energy recovery options is shown in Fig. 5. The figures refer to the case of polyethylene and polypropylene. Each process is characterised by two bars. The left bar indicates the impact of individual inputs and outputs, the right bar “net” indicates the net CO\textsubscript{2} impact (compared to disposal). The bars suggest that re-extrusion is the most attractive option (net impact compared to incineration without energy recovery a reduction of 3.5–4 t CO\textsubscript{2}/t waste), followed by cement kilns and blast furnaces (3.5 t CO\textsubscript{2}/t waste) hydrogenation (0.4 t CO\textsubscript{2}/t waste) and pyrolysis. Gasification and grate incineration are the least attractive options from a CO\textsubscript{2} point of view.

Re-extrusion is limited by the waste quality. Estimates for Western Europe case suggest that approx. 15% of all plastic waste can be treated this way. Still the comparison suggests that an emission reduction of 3.5 t/t is feasible in case a switch occurs from incineration without energy recovery to plastic waste use in blast furnaces or in cement kilns.

5. Analysis of the cost–effectiveness of emission reduction strategies

The functional unit for comparison of ethylene production have been discussed in Section 2. For production of chemical intermediates, the functional is one ton of product. For waste, the functional unit is one ton of waste treated.

Table 5 shows the costs per functional unit for the improvement option, for the current production/treatment process (the reference) and the CO\textsubscript{2} reduction impact. In the last column, the costs per ton of CO\textsubscript{2} reduction have been calculated using this information. A number of conclusions can be drawn from Table 5. The improvement options reduce in many cases the CO\textsubscript{2} emission significantly. In a number of cases, they reduce simultaneously the production costs considerably. The reason these technologies are not yet widely applied is that they have not yet been proven on a commercial scale.

Especially the catalytic production of petrochemical intermediates, biomass based production of intermediates and the cracking of heavy feedstocks deserve special attention as cost–effective technologies. However a switch to heavier oil feedstocks has no benefits from a CO\textsubscript{2} emission point of view, while intermediates production (especially based on the biomass options) has CO\textsubscript{2} benefits. The use of biomass feedstocks for production of basic chemicals is attractive from a CO\textsubscript{2} reduction point of view, but the costs are comparatively high. The HTU route has the lowest cost, below 10,000 yen/t CO\textsubscript{2}. This cost level is not completely hypothetical. However the technical feasibility of this process is not yet proven on a commercial scale. In conclusion biomass feedstocks may become attractive in Japan if ambitious GHG emission reductions are aimed for, but an active development is required. Moreover significant cost savings and CO\textsubscript{2} emission reductions are possible in plastic waste handling. The latter type of options is currently
### Table 5
Calculated production costs and CO₂ emission reduction costs

<table>
<thead>
<tr>
<th>Option</th>
<th>Option cost (yen/unit activity)</th>
<th>Reference (yen/unit activity)</th>
<th>CO₂ reduction (t CO₂/unit activity)</th>
<th>Emission reduction cost [yen/t CO₂]</th>
</tr>
</thead>
<tbody>
<tr>
<td>MTO</td>
<td>195,000</td>
<td>60,000</td>
<td>7.3</td>
<td>18,500</td>
</tr>
<tr>
<td>Ethanol dehydrogenation</td>
<td>130,000</td>
<td>60,000</td>
<td>4.0</td>
<td>17,500</td>
</tr>
<tr>
<td>HTU oil cracking</td>
<td>86,000</td>
<td>60,000</td>
<td>8.0</td>
<td>3250</td>
</tr>
<tr>
<td>Butadiene flash pyrolysis</td>
<td>85,000</td>
<td>45,000</td>
<td>4.2</td>
<td>9500</td>
</tr>
<tr>
<td>Phenol flash pyrolysis</td>
<td>62,000</td>
<td>90,000</td>
<td>3.6</td>
<td>−7800</td>
</tr>
<tr>
<td>Butanol fermentationa</td>
<td>26,000</td>
<td>110,000</td>
<td>3.4</td>
<td>−25,000</td>
</tr>
<tr>
<td>VCM catalytic production</td>
<td>40,000</td>
<td>45,000</td>
<td>0.5</td>
<td>−10,000</td>
</tr>
<tr>
<td>AN catalytic production</td>
<td>70,000</td>
<td>100,000</td>
<td>0.5</td>
<td>−60,000</td>
</tr>
<tr>
<td>Butadiene catalytic production</td>
<td>40,000</td>
<td>45,000</td>
<td>0.5</td>
<td>−10,000</td>
</tr>
<tr>
<td>Oxidative coupling</td>
<td>105,000</td>
<td>60,000</td>
<td>0.7</td>
<td>28,000</td>
</tr>
<tr>
<td>Ethane cracking</td>
<td>99,500</td>
<td>60,000</td>
<td>0</td>
<td>−</td>
</tr>
<tr>
<td>LPG cracking</td>
<td>87,000</td>
<td>60,000</td>
<td>0</td>
<td>−</td>
</tr>
<tr>
<td>Gas oil cracking</td>
<td>53,000</td>
<td>60,000</td>
<td>0</td>
<td>−</td>
</tr>
<tr>
<td>DCC</td>
<td>44,500</td>
<td>60,000</td>
<td>0</td>
<td>−</td>
</tr>
<tr>
<td>Waste incineration with energy recovery</td>
<td>10,000</td>
<td>5000</td>
<td>0.9</td>
<td>5500</td>
</tr>
<tr>
<td>Waste pyrolysis</td>
<td>1000</td>
<td>5000</td>
<td>1.1</td>
<td>−3600</td>
</tr>
<tr>
<td>Waste gasification</td>
<td>2500</td>
<td>5000</td>
<td>0.9</td>
<td>−2750</td>
</tr>
<tr>
<td>Waste hydrogenation</td>
<td>0</td>
<td>5000</td>
<td>2.7</td>
<td>−1800</td>
</tr>
<tr>
<td>Waste blast furnace</td>
<td>2100</td>
<td>5000</td>
<td>3.3</td>
<td>−900</td>
</tr>
<tr>
<td>Waste cement kiln</td>
<td>−4000</td>
<td>5000</td>
<td>3.8</td>
<td>−2400</td>
</tr>
<tr>
<td>Waste re-extrusion</td>
<td>−40,000</td>
<td>5000</td>
<td>4.2</td>
<td>−10,000</td>
</tr>
</tbody>
</table>

a Acetone by-product (0.39 t/t butanol) valued at 50,000 yen/t.

actively pursued, and the technical feasibility has been proven abroad.

The potentials for emission reduction are a function of the emission reduction per unit of activity and the activity. The activity is constrained by resource availability and market volumes. The resulting emission reduction potentials are shown in Table 6. The figures for emission reduction cannot be added because options compete for limited quantities of resources (e.g. cement kilns and blast furnaces compete for plastic waste). However the figures suggest that approx. 60 Mt emission reduction is possible on the supply side, while 35 Mt emission reduction can be achieved by improvements in waste management. The total amounts to a national emission reduction of 7.7%. The last column indicates the timing, based on the life of existing capital equipment and the current state of development of different technologies. An emission reduction of approx. 20 Mt seems possible within the time frame of the Kyoto protocol, another 35 Mt until 2020, and another 40 Mt beyond 2020. Such a scenario would result in a sustainable biomass based petrochemical industry. However the availability of sufficient biomass is a key issue for the achievement of this goal.

Optimal waste treatment may imply increased waste transportation. Coke ovens, blast furnaces and cement kilns are not evenly spread over the country. The regional imbalance of waste generation and waste treatment facilities is indicated in Table 7. Model calculations suggest that transportation increases by 0.2–0.5 billion t.km in case a CO₂ tax is introduced (+0.1 to +0.2% on a national level [6]). The results indicate significant waste transportation between rural and urban areas and between Kinki and Chubu, Chubu and Kanto, and Kanto and Tohoku (Fig. 6). These flows are caused by the imbalance of local waste generation and local waste treatment facilities. The impacts of such transportation can be substantial on a local scale, especially in the neighbourhood of large waste treatment facilities.

### 6. Conclusions

Energy efficiency in the Japanese industry is one of the highest in the world. As a consequence, reduction of CO₂ emissions is thought to be difficult and costly. However little attention has been paid as of yet to changes related to so-called non-energy use of fossil fuels. The analysis in this paper suggests that a large number of options exist for emission reduction in the Japanese petrochemical industry. This includes the introduction of biomass feedstock, the introduction of catalytic production processes, and change in waste handling. The use of bio-plastics and the use of CO₂ feedstock seem costly options for GHG emission reduction that should not be applied on the short term.

Japanese GHG emissions can be reduced by 7.7% if the optimal set of emission mitigation options is applied. About 60 Mt emission reduction (4.9%) is achieved by changes on the supply side, another 35 Mt emission...
reduction (2.8%) is achieved by changes in waste management. He changes in waste management can be implemented before 2010, biomass introduction on the supply side will probably require a longer lead time. About half of the emission reduction is cost–effective, but will require further development of new technology. The other half can be achieved at a cost level of 10,000 yen/t CO₂, in combination with technological development. The main policy uncertainties exist with regard to the availability of biomass and the feasibility of the technologies, especially on the petrochemical supply side. About half of the emission reduction potential is uncertain as of yet, the other half has been proven on a commercial scale in other countries.

The analysis in this paper is static in the sense that a number of interactions are not considered. For example the price of biomass may increase in the future if significant global CO₂ reductions are aimed for, because electricity producers want to use the same resource. Other analyses suggest the demand for cement may be reduced significantly if CO₂ policies are introduced. Structural changes in the steel industry may affect the potential to use plastic waste [58]. Such a change would reduce the potential for waste plastic incineration.
cement kilns. One level of fossil fuel prices has been applied in this study, prices may change as the demand structure or the political situation changes. Dynamic systems engineering studies can help to analyse such effects, see for example [6,59]. Finally the impacts of emission reduction policies on the competitive position of the Japanese petrochemical industry have not been analysed in detail, but they can be substantial [8]. Given the sensitivity of the profitability of this sector for energy prices, this is an issue that should be analysed on more detail before policies are formulated.

References