# Anaerobic digestion with partial ozonation minimises greenhouse gas emission from sludge treatment and disposal

Water Science & Technology **Vol 53 No 3 pp 255–263** 

**H. Yasui*, R. Matsuhashi**, T. Noike*** and H. Harada******

*R&D Division, Kurita Water Industries, 1-1 Gochoyama, Nogi, Tochigi, Japan, 329-0105 (E-mail: *hidenari.yasui@kurita.co.jp*)

**Institute of Environmental Studies, Graduate School of Frontier Sciences, The University of Tokyo, 7-3-1, Hongo, Bunkyo-ku, Tokyo, Japan, 113-8656 (E-mail: *matu@globalenv.t.u-tokyo.ac.jp*)

***Advanced Research Institute for the Sciences and Humanities, Nihon University, 4-2-1 Kudan-kita, Chiyoda-ku, Tokyo, Japan, 102-0073 (E-mail: *noike@arish.nihon-u.ac.jp*)

****Department of Civil and Environmental Engineering, Nagaoka University of Technology, 1603-1, Kamitomioka, Nagaoka, Niigata, Japan, 940-2188 (E-mail: *ecohara@vos.nagaokaut.ac.jp*)

**Abstract** A novel anaerobic digestion process combined with partial ozonation on digested sludge was demonstrated for improving sludge digestion and biogas recovery by full-scale testing for 2 years and its performance was compared with a simultaneously operated conventional anaerobic digestion process. The novel process requires two essential modifications, which are ozonation for enhancing the biological degradability of sludge organics and concentrating of solids in the digester through a solid/liquid separation for extension of SRT. These modifications resulted in high VSS degradation efficiency of ca. 88%, as much as 1.3 times of methane production and more than 70% reduction in dewatered sludge cake production. Based on the performance, its energy demands and contribution for minimisation of greenhouse gas emission was evaluated throughout an entire study of sludge treatment and disposal schemes in a municipality for 130,000 p.e. The analysis indicated that the novel process with power generation from biogas would lead to minimal greenhouse gas emission because the extra energy production from the scheme was expected to cover all of the energy demand for the plant operation, and the remarkable reduction in dewatered sludge cake volumes makes it possible to reduce $N_2O$ discharge and consumption of fossil fuel in the subsequent sludge incineration processes.
**Keywords** High-efficiency anaerobic digestion; Kyoto protocol; sludge minimisation

## Introduction

Minimisation of greenhouse gas (GHG) emission as proposed in the COP3 (Third Conference of Parties to the UN Framework Convention on Climate Change) led to the implementation of comprehensive regulations to reduce the production of wastes in Japan. In the new regulations, in effect since 2001, the priority is placed on developing new ways to reduce the amount of waste on-site and to recycle biomass as much as possible. Large quantities of municipal sludge produced from treatment plants contribute to the waste stream and pose significant challenges for appropriate treatment, disposal or re-use. Anaerobic sludge digestion for on-site reduction of sludge quantities and biogas recovery is one of the important treatment options that fits into the framework of new regulations and also to meet Kyoto protocol. Although a significant amount of municipal sludge is anaerobically digested, conventional solid reduction efficiencies remain at only 40–60% in the current situation. It is therefore desirable to study and develop modified schemes of anaerobic digestion to improve the digestion efficiency and reduce sludge production on-site.

doi: 10.2166/wst.2006.104

Recent study on an anaerobic digestion process with partial ozonation on digested sludge is one of such efforts to achieve large biogas conversion efficiencies (Weemaes *et al.*, 2000; Goel *et al.*, 2003a–d; Yasui *et al.*, 2003, 2005a). Based on the results of the laboratory-scale experiments in which 80% of biogas conversion efficiency was achieved using anaerobic digestion with ozonation, we proceeded to a full-scale testing to verify the process performance with respect to solid degradation and biogas recoveries. In the study LCA methodology was highlighted to conduct a comparative analysis regarding GHG emission for the conventional and the high efficiency anaerobic digestion process with partial ozonation (Matsuhashi *et al.*, 1997; Yasui *et al.*, 2005b). This article will focus on the outline of full-scale tests and its LCA whether it will result in reduction of GHG emission in the total sludge treatment system. Since the impact of the process on sludge treatment system is highly site specific, pertinent site-specific data are essential for precise LCA. In this regard, a city having 130,000 inhabitants was chosen to collect comprehensive field data in order to present future alternatives for sophisticated sludge treatment and disposal systems, by formulating existing sludge treatment and disposal facilities of the anaerobic digestion plant, two incineration plants and two landfill leachate treatment plants.

## Materials and methods

### Full-scale tests

The full-scale test was carried out at a municipal wastewater treatment centre having secondary treatment capacity of 7,800 $m^3$/day (BOD removal only). The daily production of primary sludge and secondary excess sludge at the treatment centre was 1,035 kg-DS (874 kg-VS) and 759 kg-DS (649 kg-VS), respectively. The treatment centre has two mesophilic anaerobic digestion tanks, each of 1,125 $m^3$ capacity, with a mechanical mixer. The HRT of each anaerobic digester on the basis of the flow rate of municipal sludge fed was set at 45 days. Based on historical operational data, the efficiency of TS and TVS reduction in the conventional digestion process was estimated to be 54 and 62%, respectively. Approximately 10,000 t/year of dewatered sludge cake with water content of about 80% was produced from the plant. During the test period, one of the digestion tanks was modified to a new process scheme (test reactor, Figure 1), while the other digester was operated according to the conventional digestion scheme (control reactor). The municipal sludge was divided and introduced in equal amounts to the reactors during the full-scale testing. To complete the test process scheme, an ozoniser (average ozone input: 24 kg-$O_3$/day) to decompose sludge with ozone and a centrifugal device to withdraw inorganic-rich digested sludge with maintaining high VSS concentration in the tank were installed in the flow lines of the test reactor.

For sludge ozonation, a fraction of the digested sludge was fed to the ozoniser and returned to the anaerobic digestion tank after removing residual ozone and oxygen in the liquid. A large fraction of the ozonated-digested sludge is then biologically converted to biogas in the anaerobic digestion tank. The flow rate of sludge fed to the ozonation reactor varied in the range of 22–44 $m^3$/day. Implementing a centrifuge for sludge returning in the

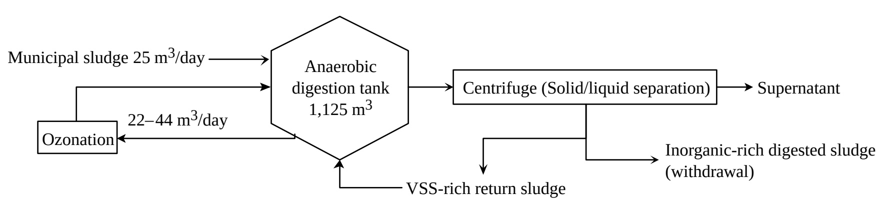


**Figure 1** Process scheme of the test reactor – the high-concentrated anaerobic digestion with partial ozonation of digested sludge

test reactor, retention time for the degradation of VSS compounds was extended. The centrifugal device was operated with and without addition of polymer coagulants depending on whether the thickened sludge is recycled to the anaerobic digester or discharged for disposal. When thickened sludge was discharged for disposal, the centrifugal device was operated without addition of polymer coagulants. Inorganic solids having higher density were concentrated by the centrifugal force into the thickened sludge, leading to withdrawal of sludge with low TVS/TS ratio (inorganic-rich solids). The decantant having lower density (VSS-rich solids) was returned back to the anaerobic digestion tank. When the thickened sludge was used for recycling as return sludge, the centrifugal device was operated with the addition of polymer coagulants. The supernatant was discharged at the head of the primary settling tank in the wastewater treatment plant.



#### LCA methodology

For LCA study, a city having 130,000 habitants in Niigata prefecture, Japan was chosen where a large amount of biogas utilisation was expected. The flow-scheme of municipal sludge treatment is illustrated in Figure 2. About 7.7 t-TS/day of primary sludge and 3.7 t-TS/day of excess sludge are produced by the conventional activated sludge process without nutrient removal. The mixed sludge is digested in a mesophilic anaerobic digestion process having a hydraulic retention time of 52 days. The process produces 4,500 $Nm^3$/day of biogas with 66% of VSS degradation. Most biogas is purified to reach 96% of methane gas concentration, using water scrubber to remove $CO_2$ gas. The methane gas is then forwarded to a local gas company and distributed to citizens as an alternative to natural gas. In this way, 3% of total natural gas consumption by citizens is saved consistently, which contributes in reducing about 1,000 t of GHG emission every year. The daily production of dewatered sludge cake is about 21.6 t, having a water content of 76% and VSS/SS ratio of 0.58.

Approximately one-third of the dewatered sludge cake is incinerated along with municipal solid waste at a municipal incineration facility adjoining the wastewater treatment plant. Owing to the high calorific value of the solid waste, supplemental fuel is not used for the incineration. The incinerated ash is landfilled at a municipal disposal site located 15 km away from the plant. Inorganic compounds in the leachate are further removed at the site by coagulation and N/DN treatment. The rest of the dewatered sludge cake is hauled to a private incineration plant located 16 km away from the city and incinerated together with municipal dewatered sludge cake collected from other 27 wastewater treatment plants in the prefecture (63 t-cake/day in total). Heavy oil and rubber taken from waste tyres are used as alternative fuel for the incineration. The operation of the incinerator is shut down once every two weeks for cleaning and maintenance. Accordingly, a significant amount of fuel is consumed to re-heat the incinerator after maintenance. Every year 430 t of incinerated ash is recycled to the cement industry as a raw material for cement and 1,900 t is landfilled

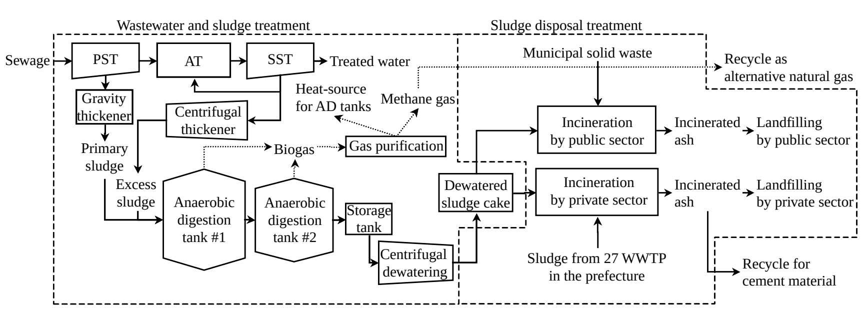


**Figure 2** The current flow-scheme of sludge treatments for the city

in the plant. The inorganic compounds in the leachate are removed by a similar manner to that mentioned the above.

The basic sludge treatment and disposal process flow scheme consists of conventional anaerobic digestion, dewatering, incineration and landfilling. In the alternative formulation, the high-concentrated anaerobic digestion with partial ozonation of digested sludge was used in place of conventional anaerobic digestion. In each alternative, two scenarios were formulated based on the type of beneficial use of biogas, i.e. the scenarios of (1) internal recycle: power generation by gas engine to supply electricity for the plant facilities and (2) external recycle: distribution as alternative natural gas to local sectors. In all scenarios, it was assumed that ozonation equipment for the anaerobic digestion plant would be operated at night-time only in order to utilise night electricity which has lower GHG emissions. This is because the power stations of the local electric power company are operated with a high share of nuclear power generation and there is less consumption of fossil fuels in this time period. Total GHG emission in the sludge treatment system of the city was given based on the models with input–output LCA procedure to analyse energy and material-flow matrix (Matsuhashi *et al.,* 1997; Yoshida *et al.*, 2002). The energy and material-flow matrix and its algorithm to seek the solution for the optimum process scheme are shown in Figures 3 and 4, respectively.

For compilation of energy and material data for sludge treatment and disposal in the city, additional data for LCA analysis, e.g. energy and materials consumed in the anaerobic digestion plant, the incineration plants and landfilling leachate treatment plants, were collected by conducting individual plant surveys and extracting information from annual operation reports and technical articles. The experimental data as described above were used to estimate the production of dewatered sludge cake. The amount of dewatered sludge cake, energy and material data formed the basis for evaluating GHG emission in the subsequent treatment/disposal processes (Ministry of Land, 2000; JSA, 2001; Yasui *et al.*, 2005b). The minimal $LCCO_2$ (minimal GHG emission) was identified by quasi-Newton method equipped in Microsoft Excel® solver.

## Results and discussion

### Full-scale experiment

The trend of SS concentration and TVS/TS ratio in the reactors are as shown in Figure 5. In the control reactor SS concentration and TVS/TS ratio remained almost constant at 15–16 and 0.70–0.72 g/L, respectively. During the start-up phase of the test reactor, no sludge was withdrawn. This led to an increase in TS concentration in the anaerobic digestion tank to 55 g/L. TVS/TS ratio of the digested sludge kept on decreasing consistently owing to improvement of organic solids degradation in the phase. To prevent further

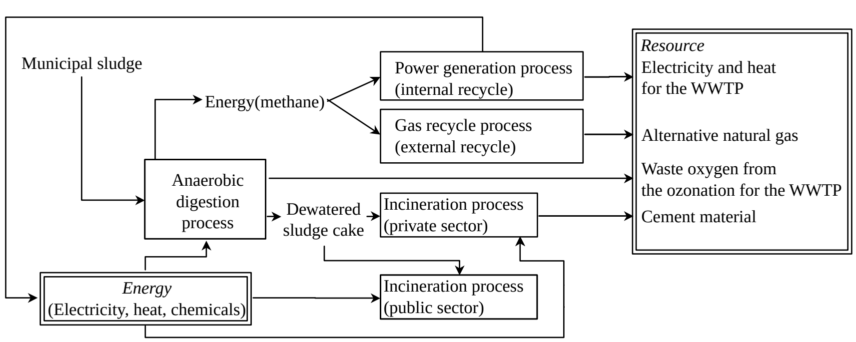


**Figure 3** The energy and material-flow matrix of the city for municipal sludge treatments and disposal processes

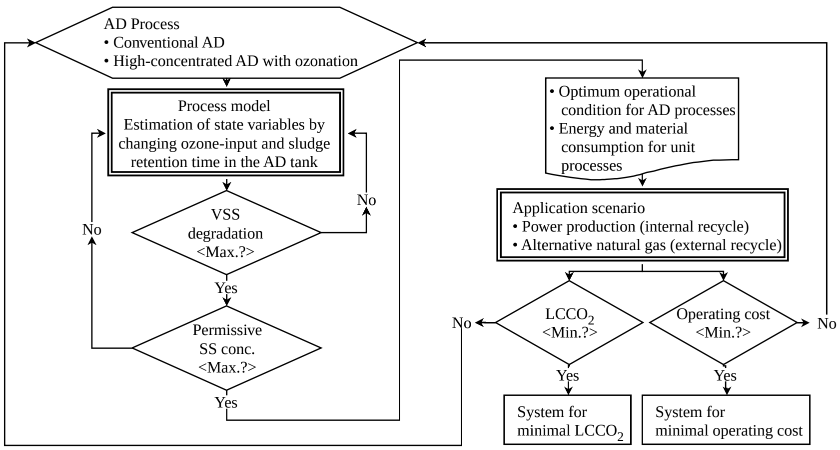


**Figure 4** The algorithm seeking the optimum process scheme

increase and to maintain TS concentration at about 60 g/L, intermittent sludge withdrawal was conducted in the test reactor from June 2003 as keeping TVS/TS ratio at about 0.6. Corresponding to the start of withdrawal, the TVS/TS ratio ceased decreasing and remained constant after August 2003.

During the steady state, the degree of sludge digestion was summarised based on the mass balance using (1) amount of municipal sludge fed to the reactor, (2) change in SS concentration in the reactor and (3) amount of solid withdrawn during the operation. The results of this analysis are as shown in Figure 6. As the sum of points (2) and (3) is equal to the undigested sludge mass, the slope of the graph indicates the fraction of undigested solid in the municipal sludge. A clear linear relationship can be seen in the control reactor between the amount of fed municipal sludge and of undigested solid. The line slope of 0.46 (i.e. digestion efficiency = 54%) was consistent with the historical data of

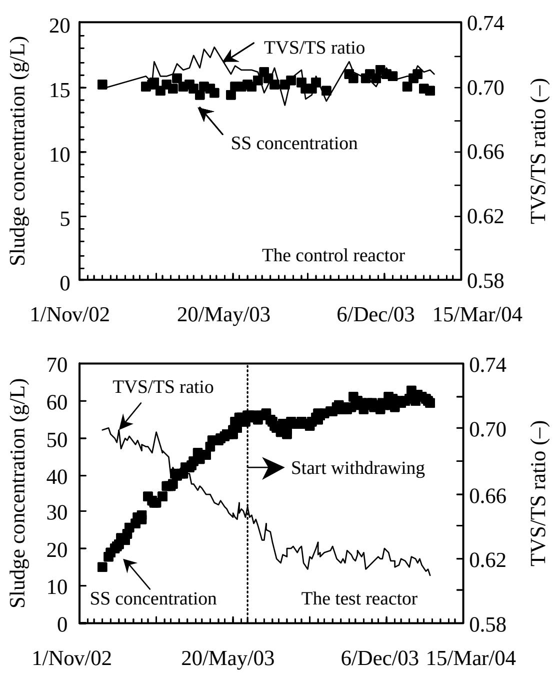


**Figure 5** The trend of SS concentration and TVS/TS ratio (top: the control reactor, bottom: the test reactor)

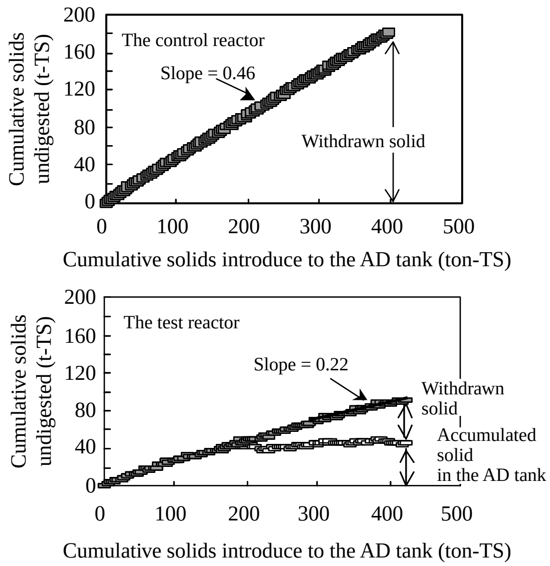


**Figure 6** The fraction of undigested sludge (top: the control reactor, bottom: the test reactor)

digestion at the sludge treatment plant. The test reactor showed significantly lower undigested solid mass. During the steady-state period, only 22% of TS was withdrawn from the test reactor that was less than half of that withdrawn from the control reactor.

Next, the degradation of VSS compounds was evaluated in the same manner as mentioned above. The un-decomposed fraction of VSS compounds in the municipal sludge was only 12% at the test reactor and 37% at the control reactor. The biogas production in both reactors was consistent with the quantity of TVS degradation. The biogas production at the test reactor was 1.3 times higher than that at the control reactor. According to the measurement of gas production rate and gas analysis, 0.54–0.57 $Nm^3$ of methane gas was produced through the degradation of 1 kg of degraded TVS compounds. The biogas was composed of 59–61% methane, 38–40% $CO_2$ and 1–2% miscellaneous gas, which did not show significant variation between the test and control reactors. Since only little ozone was dosed to the sludge at the ozonation stage, it is considered that the ozone treatment is not influential in the loss of the COD fraction for the methane source (Goel *et al.*, 2003a).

The production of dewatered sludge cake for the test and the control reactor is compared in Figure 7. The amount of dewatered sludge cake from the test reactor was significantly less than that of the control reactor. In the steady-state operation, the cake produced from the test reactor was 0.46 t/day, which was less than 30% of that produced from the control reactor. Owing to the lower TVS/TS ratio of sludge in the test reactor, the water content of dewatered sludge cake was correspondingly reduced from 80% to less than 70%, as shown in Figure 8. This decrease in water content contributed a substantial reduction in volume of the dewatered sludge cake required for hauling and further disposal. Furthermore, the evaluation study for incineration of the dewatered sludge cake suggested that the energy value of the VSS compounds was enough to vaporise the water in the cake. This means that the application of anaerobic digestion process with ozonation makes it possible to minimise the consumption of supplement fuel for the subsequent incineration process. In addition, since the digestion minimises the amount of nitrogenous compounds in the sludge, the potential $N_2O$ production at the incineration stage can also be reduced. As $N_2O$ on the greenhouse effect is 310 times higher than $CO_2$, the reduction of the fuel and sludge quantity to be incinerated may contribute in suppressing GHG emission.

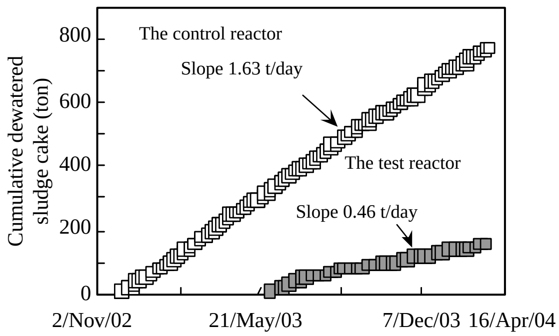


**Figure 7** The production of dewatered sludge cake

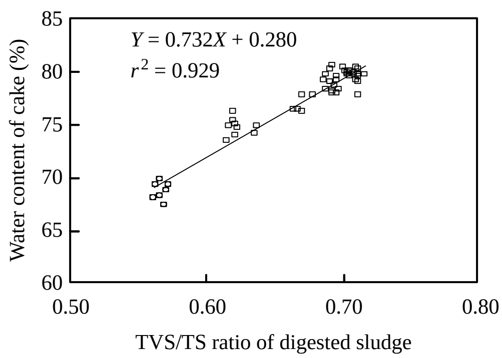


**Figure 8** Reduction of cake volume by lowering TVS/TS ratio of sludge

#### Evaluation of $LCCO_2$ for sludge treatment and disposal

Based on the operational data and process model developed by Yasui *et al.* (2005a), the expected dewatered sludge cake production at the city was simulated as a function of daily ozone-input, as shown in Figure 9. It appears that the sludge production reduces significantly as the amount of ozone-input is increased whereas the decrease in sludge reduction is not a linear function. This is because addition of higher ozone-input reduces the biological sludge retention time in the AD tank since more sludge mass is withdrawn for the ozonation (Yasui *et al.*, 2005a). This too greater sludge recirculation through ozonation also results in the decrease of TVS digestion efficiency for both the primary sludge and excess sludge. Consequently, the minimum achievable amount of dewatered sludge cake is expected to be about 3,100 t/year under the given operating conditions at the wastewater treatment plant. The use of a gas engine is expected to cover all of the electricity and heat demand for the AD process if the daily ozone-input is below 360 $kgO_3$/day (data not shown).

GHG emission in the sludge treatment and disposal is summarised in Figure 10. A typical bathtub-curve was obtained for GHG emission depending on the amount of ozone-input. In the operation range of lower ozone-input (left-hand side), the amount of GHG emission became closer to that for the conventional AD process. The major contribution here is the production of $N_2O$ derived from the large quantity of dewatered sludge cake to be incinerated. Since more $N_2O$ is generated from nitrogenous compounds in sludge organics at lower incineration temperatures and it has 310 times higher GHG effects than $CO_2$, careful analysis is needed for a sludge incineration process to evaluate GHG emission. On the other hand, in the case of higher ozone-input (right-handside), the GHG emission was also elevated due to more consumption of electricity for

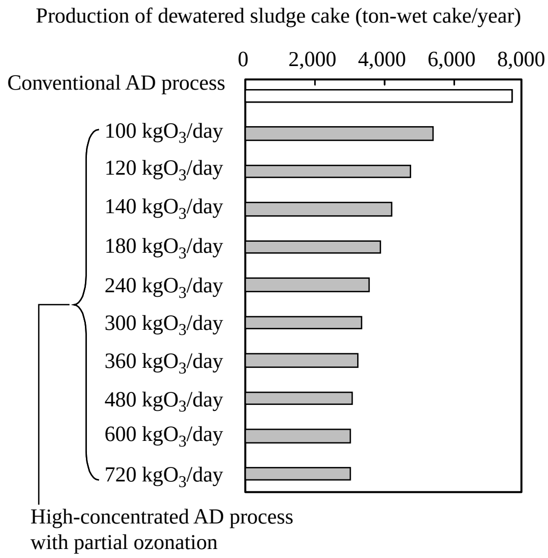


**Figure 9** Expected dewatered sludge cake production

ozonation. The amount of GHG seemed to be least in the ozone-input range of 240–300 kg-$O_3$/day. In addition, the amount of waste oxygen as a by-product from the ozonation is calculated to be 2,200–2,700 kg-$O_2$/day. This amount is expected to cover more than 60% of oxygenation demand in the aeration of the wastewater treatment plant (data not shown). The reduction of GHG corresponds to the saving in crude oil consumption of 600 kL/year. Accordingly, the application of the process can contribute to the reduction of 4.6 L-crude oil/p.e. per year for the city, which is 60% of the government's goal on the basis of population equivalent.

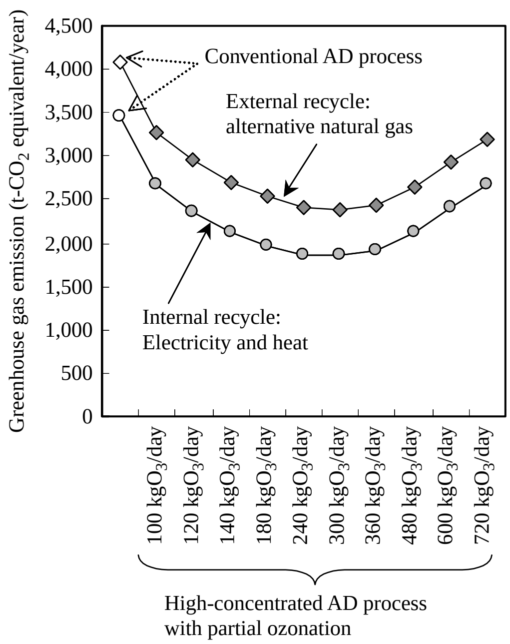




**Figure 10** The estimation of greenhouse gas emission in sludge treatment and disposal in the city

Among the options for biogas utilisation, the analysis suggests that internal recycle (for source of electricity by gas engine) would be a proper option for the city and a further 500 t-$CO_2$ equivalent/year can be reduced from the current process scheme (external recycle for alternative natural gas). This result can be explained by considering the fact that the fundamental unit of $CO_2$ emission equivalent to the local electric power company serving the area is higher than that of the gas company. Since the fundamental unit of $CO_2$ equivalent (kg-$CO_2$/kWh) of an electric power company strongly depends on its composition of power source (e.g. nuclear, water, LNG, coal), the method of biogas utilisation should be based on the local energy and material-flow matrix.



## Conclusions

A high-concentrated anaerobic digestion with partial ozonation of digested sludge was tested at a full-scale anaerobic digester. The process was successfully verified with improved biogas conversion efficiency of over 80% from the organics in municipal sludge. Owing to enhanced methane recovery and less production of dewatered sludge cake with lower water content, the process is expected to be a suitable option to maximise the possibility of biogas utilisation and contributes to minimising greenhouse gas production in the subsequent incineration stage. The LCA procedure can identify the optimum system configuration of sludge treatment, disposal and biogas utilisation that strongly depends on local energy and material flow.

## References

Goel, R., Tokutomi, T. and Yasui, H. (2003a). Anaerobic digestion of excess activated sludge with ozone pre-treatment. *Wat. Sci. Tech.*, **47**(12), 207–214.

Goel, R., Yasui, H. and Shibayama, C. (2003b). High-performance closed loop anaerobic digestion using pre/post sludge ozonation. *Wat. Sci. Tech.*, **47**(12), 261–268.

Goel, R., Tokutomi, T., Yasui, H. and Noike, T. (2003c). Optimal process configuration for anaerobic digestion with ozonation. *Wat. Sci. Tech.*, **48**(4), 85–96.

Goel, R., Komatsu, K., Yasui, H. and Harada, T. (2003d). Process performance and change in sludge characteristics during anaerobic digestion of sewage sludge with ozonation. *Wat. Sci. Tech.*, **49**(10), 105–114.

Japan Sewage Association (2001), Sewage statistical report (in Japanese).

Matsuhashi, R., Sudoh, O., Nakane, K., Yasui, H., Nakayama, S. and Ishitani, H. (1997). Life cycle assessments of sewage treatment technologies. In *Proc. IAWQ International Conference on Sludge Management Part 1*, Czestochowa, Poland, pp. 227–234.

Ministry of Land, Infrastructure and Transportation (2000). Report on reduction of greenhouse gas emission from municipal sludge incinerators. *Technical Memorandum of Public Works Research Institute*, No. 3731, (in Japanese).

Yasui, H., Goel, R., Fukase, T., Matsuhashi, R., Wakayama, M., Sakai, Y. and Noike, T. (2003). Bioleader®, a novel activated sludge process minimizing excess sludge production. In *Proc. IWA Leading Edge Conference*, Noordwijk/Amsterdam, The Netherlands.

Yasui, H., Komatsu, K., Goel, R., Li, Y.Y. and Noike, T. (2005a). Full-scale application of anaerobic digestion process with partial ozonation of digested sludge. *Wat. Sci. Tech.*, **52**(1–2), 245–252.

Yasui, H., Komatsu, K., Goel, R., Sato, A., Matsuhashi, R., Ohashi, A. and Harada, H. (2005b). Minimization of greenhouse gas emission by application of anaerobic digestion process with biogas utilization. *Wat. Sci. Tech.*, **52**(1–2), 545–552.

Yoshida, Y., Ishitani, H., Matsuhashi, R., Kudoh, Y., Okuma, H., Morita, K., Koike, A. and Kobayashi, O. (2002). Reliability of LCA considering the uncertainties of energy consumptions in input-output analyses. *Applied Energy*, **73**, 71–82.

Weemaes, M., Grootaerd, H., Simoens, F. and Verstraete, W. (2000). Anaerobic digestion of ozonized biosolids. *Wat. Res.*, **8**, 2330–2336.