New Technology for Wastewater Control

-Cutting-edge of Wastewater Treatment Technology-

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1. MBR for municipal wastewater treatment

The membrane bioreactor (MBR) is a wastewater treatment technology that offers many advantages. In Japan, although MBRs have long been used for industrial wastewater treatment or for reuse of wastewater in large buildings and so on, the introduction of MBRs in sewerage systems has lagged behind compared with other water related fields. However, the first MBR for municipal wastewater treatment in Japan started operation in March 2005, and this accelerated the introduction of MBRs in Japanese sewerage systems. Five MBR plants for municipal wastewater treatment are in operation at present. In addition, there are some 10 MBR plants currently in the design or planning stage. The number of MBRs for municipal wastewater is expected to increase in the years ahead. Some fruits of research work of JSWA about MBR are introduced to below.

Risk reduction of the water environment

It is known that a high degree of virus removal could be achieved by MBR.[1] According to our previous research, it was found that Colipharges and *Norovirus* of approximately one-tenth membrane pore size were almost completely removed by a MF membrane. It was also found that most of the virus existed in the activated sludge, indicating that they attached themselves to the activated sludge. [2] It is suggested that the virus captured in the gel layer that forms on the surface of the membrane also contribute to virus removal. [3] Therefore it is considered that virus removal is influenced by the chemical cleaning of the membrane.

The behavior of *Norovirus* captured in the MBR activated sludge and the influence of the chemical cleaning of the membrane was investigated using a pilot scale MBR which treats 48 m^3/d of actual municipal wastewater. A hollow fiber MF membrane unit with 0.4μm pore size was immersed in the oxic tank and operated at a permeate flux of 0.8 m^3/m^2/d.

The pilot plant was operated with a HRT of 6 hrs in Run-1, in which the oxic tank HRT was 3 hrs. During the experiment however, in order to evaluate the effect of the HRT on virus removal, the total HRT was then extended to 18 hrs in which the oxic tank HRT was 6 hrs in Run-2 and 18 hrs in Run-3 respectively.

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The number of *Norovirus* in the activated sludge decreased with prolonged oxic tank HRT, and the number of G I type in Run –2 was about 1/1000 that of Run-1 as shown in Table 1.

Table 1. *Norovirus* measurement results for different aerobic tank HRT

	Run−1		Run−2		Run−3	
Oxic tank tank HRT(hrs)	3		6		18	
Genome type	G I	G II	G I	G II	G I	G II
Influent	4.20E+07	3.50E+08	2.50E+07	1.90E+08	2.50E+07	1.90E+08
Aerobic tank sludge	1.10E+08	6.20E+07	1.50E+06	N.D.	2.70E+05	N.D.
Effluent	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.

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This indicates that the aerobic decomposition of the virus, which was adsorbed in the activated sludge and retained in the oxic tank, will progress with the elapse of time.

The number of *Norovirus* was measured at 30, 60, and 120 minutes after the in-line chemical membrane cleaning using a 0.3% NaOCl solution. No *Norovirus* were detected in the MBR effluent after the completion of chemical cleaning procedure. This may suggest that the role of gel layer of the membrane surface on virus removal is less important than the adsorption effect of the activated sludge.

Application to BNR process

Figure 1 shows a new biological nutrient removal (BNR) process for large facilities with membrane separation.[4] This process combines the step feed multi-stage nitrification-denitrification process, which is a popular BNR process in Japan, and membrane separation. In a single-stage MBR, in order to obtain nitrogen removal efficiency of more than 80%, circulation ratio of some 400% (R=4) is necessary.

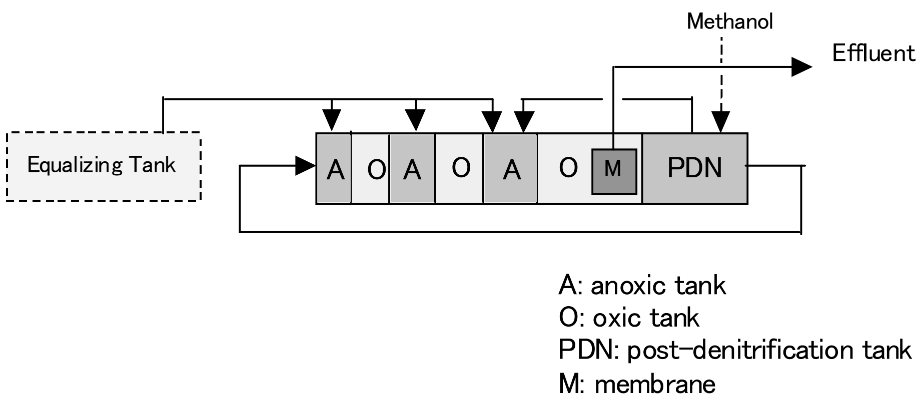

Figure 1. Flow of the BNR process with membrane

However, in the membrane BNR process shown in Figure 1, a T-N removal efficiency of about 80% is acquired at a circulation ratio of 200%. The experiment results showed that stable nitrification and denitrification could be achieved even in low water temperature period. Good and stable biological phosphorus removal can be achieved if the first anoxic tank acts as an anaerobic tank. This is enabled by adding methanol to the post-denitrification tank and eliminating NOx-N in the returned mixed liquor.

Perspective of the future development of MBR

The MBR will not remain just one of many wastewater treatment technologies, but is expected to become a core technology that will be used for various types of wastewater management.

In Japan, MBR is expected to be increasingly used for municipal wastewater treatment. Although the MBR has been used mainly for small scale plants so far, it will likely be applied to larger plants in the future. To enable the wider use of MBR for various purposes, the following issues are considered to be essential.

1) Optimization of the design method.
2) Optimization of the maintenance method, especially fouling control and chemical cleaning of membrane.
3) Reduction of membrane cost and prolongation of membrane life.
4) Reduction of energy consumption, especially air supply for membrane cleaning.

JSWA will continue technology development of MBR in cooperation with universities and private enterprises and will conduct the second technical evaluation of MBR based on the data obtained from the five operating actual MBR plants in the near future.

2. Removal of EDCs by ozonation[5]

Since the natural estrogens 17 β-estradiol (E2) and estron (E1), and the synthetic estrogen 17 α-ethynyl estradiol (EE2) have strong endocrine disrupting effects and the tendency to persist in effluent from wastewater treatment plants, effective measures are needed to remove them from wastewater.

In our research, to gain an understanding of the characteristics of estrogen decomposition by ozonation, experiments were conducted using effluent from an actual wastewater treatment plant. In this experiment, estrogen was added to effluent at a concentration of 200 ng/l and 20ng/l before the ozonation experiments.

The results showed 90% or more of estrogen concentration and estrogenic activity of E2, E1 and EE2 to be removed at an ozone dose of 1 mg/l as seen in Figure 2. At an ozone dose of 3 mg/l, the estrogen concentration and estrogenic activity of E2, E1 and EE2 in the treated water fell below the detection limit.

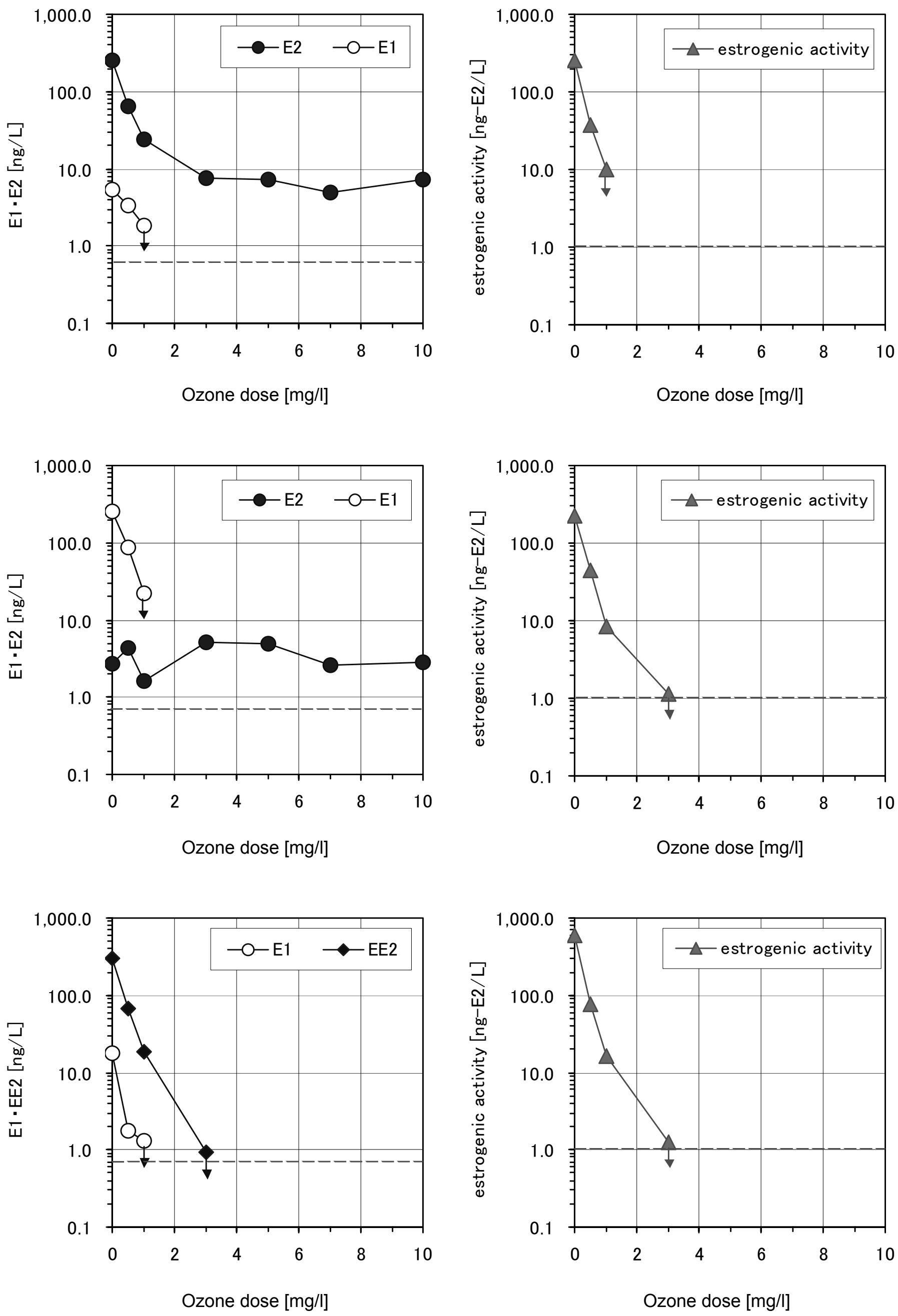

Figure 2. Changes of estrogen concentration and estrogenic activity with ozone dose.
Initial estrogen concentration: 200ng/l, contact time: 15min.
(Top row:E2 added, middle row:E1 added, bottom row:EE2 added)

The removal rate was not influenced by type of estrogen. No generation of byproducts with estrogenic activity was observed. It is concluded that estrogen in secondary treated wastewater can be almost entirely removed at the practical ozone dose rate applied for the purpose of disinfection, which is up to about 5 mg/l.

3. New biological nitrogen removal method –annamox process-

The biological nitrification-denitrification process is widely used for nitrogen removal from municipal wastewater. The annamox reaction is a biological nitrogen removal reaction that completely differs from usual nitrification-denitrification as expressed by the following chemical reaction.

$$1.0NH_4^{+}+1.32NO_2^{-}+0.066HCO_3^{-}+0.13H^{+}\rightarrow 1.02N_2+0.26NO_3^{-}+0.066CH_2O_{0.5}N_{0.15}+2.03H_2O$$

Under the anaerobic conditions, ammoniac nitrogen and nitrite react producing nitrogen gas.

The bacteria which participate in the reaction are called annamox bacteria and they are tinged with peculiar red color.

There are following merits in the annamox reaction as compared with the conventional nitrification-denitrification reaction.

1) Since about the half of ammoniac nitrogen in wastewater has to be converted into nitrite, the required oxygen amount is theoretically about 50% of that of the conventional nitrification-denitrification process.
2) Since the biological reaction is an autotrophic reaction, carbon source for denitirfication is not necessary.
3) The excess sludge production is small by the same reason.

On the other hand, there are following subjects remaining to be solved.

1) Since the growth rate of annamox bacteria is low, enrichment of bacterial culture is not easy in starting-up of the plant.
2) Applicable conditions are limited to wastewater with relatively high ammonia concentration, low C/N ratio and high water temperature.
3) The stable conversion of half of the ammoniac nitrogen in the wastewater into nitrite is essential as the preceding procedure.

It is supposed that the annamox process is suitable to treat sidestreams from anaerobic digester or from deodorization equipment of sludge drying process. Especially the reduction of nitrogen load of sidestreams from sludge treatment process will enable good and stable performance of biological nitrogen removal process. JSWA is carrying a research on the utilization of the annamox process including a joint research with private companies.

References

[1] Ottoson,J., Hansen,A., Bjorlenius,B., et al, “Removal of Viruses, Parasitic Protozoa and Microbial Indicators in Conventional and Membrane Processes in a Wastewter Pilot Plant”
Water Research 40 (2006) 1449-1457

[2] Oota,S., Murakami, T.,Takemura, K. and Noto, K. “Evaluation of MBR Effluent Characteristics for Reuse Purposes” Water Science & Technology Vol. 51 No6-7 pp441-446, 2005

[3] Ueda,T.,Horan,N.j.,”Fate of Indigenous Bacteriophage in a Membrane Bioreactor”
Water Research 34 (2000) 2151-2159

[4] Oota,S., Murakami, T., and Uriu, M. “Advanced BNR Process Combined with Membrane Separation”, Proceeding of IWA Asia-Pacific Regional Conference “Aspire 2005” (CD-ROM), July 2005

[5] Hashimoto,T., Takahashi, k., and Murakami, T. “Characteristics of estrogen decomposition by ozonation”, Water Science & Technology Vol. 54 No.10 pp87-93, 2006

2006.1.24

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Introduction of JSWA

- Public organization established by the government and local municipalities
- Purpose: Support local municipalities in sewage works
- Activities

Planning/Design/Construction of WTPs
Technical assistance
Training of local municipality staff
Research & Development

2

JSWA Mouka research center

3

Contents of the presentation

1. MBR for municipal wastewater treatment
2. Removal of EDCs by ozonation
3. New biological nitrogen removal method

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MBR for municipal wastewater treatment

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Outputs of JSWA Researches on MBR

- "Technical evaluation of MBR "(2003)
  - Process configuration
  - Operation & maintenance
  - Performance
  - Conditions of MBR application
  - Costs
  - etc.
- "The temporary design guideline "(2003)
- Five MBRs are in operation (2006)

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MBRs in operation or under construction

Name of the plant	Total design capacity (m^3/d)	Capacity for installation (m^3/d)	Membrane type	Start of operation
Fukusaki	12,500	2,100	Flat sheet	April, 2005
Kobuhara	240	240	Flat sheet	July, 2005
Yusuhara	720	360	Flat sheet	December, 2005
Okutsu	580	580	Hollow fiber	April, 2006
Daito	2,000	1,000	Flat sheet	September,2006
Kaietsu	230	230	Hollow fiber	April, 2007
Zyosai	1,375	1,375	Hollow fiber	March, 2008
Heta	3,200	2,140	Flat sheet	March, 2008
Ooda	8,600	1,075	Undecided	March, 2009

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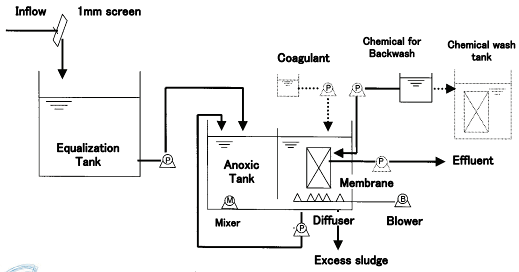
Basic Flow of MBR



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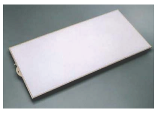
Membrane modules (FS)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FS Membrane module for large-scale application

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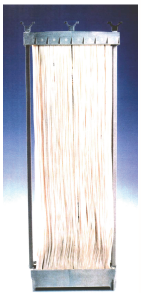
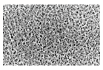
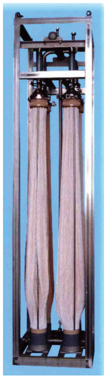
HF Membrane module



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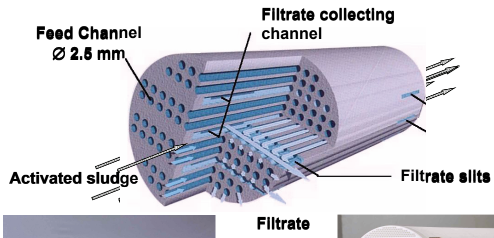
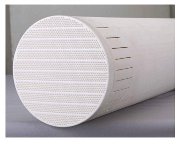
Ceramic monolith membrane module



Purposes of MBR application

- Small footprint
- Advanced treatment (C,N,P)
- Reuse of effluent
- Retrofitting
- Grade up of treatment
- Avoid chrolination in disinfection

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Fukusaki Plant

The first MBR for municipal wastewater in Japan

Started operation from the end of March, 2005
Capacity 2,100m³/d (Planned Capacity 12,500m³/d)

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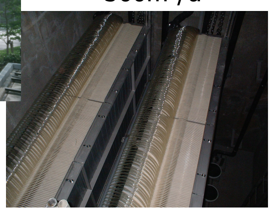
Yusuhara Plant

360m^3/d



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Okutsu Plant

580m^3/d

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Kaietsu Plant

230m^3/d

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Risk reduction of the water environment by MBR

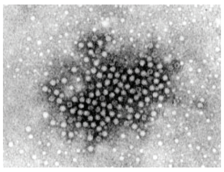
Norovirus

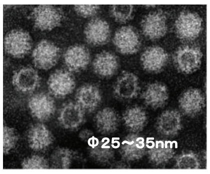
Main cause of food poisoning

Outbreak in this winter

Very small in the size

Tolerant toward chlorine disinfection





From HP of Yokohama-city and Kyoto-city

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Pilot plant

Capacity (m^3/d)	48
MLSS (g/L)	8–10
Membrane module	HF
Material	PVDF
Pore size (μm)	0.4
Flux (L/m^2/hr)	33
TMP (kPa)	<30

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Experimental condition

	Total HRT (hrs)	Oxic HRT (hrs)
Run-1	6	3
Run-2	18	6
Run-3	18	18

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Results of *Norovirus* mesurement

	Run-1		Run-2		Run-3	
Oxic tank tank HRT(hrs)	3		6		18	
Genome type	GⅠ	GⅡ	GⅠ	GⅡ	GⅠ	GⅡ
Influent	4.20E+07	3.50E+08	2.50E+07	1.90E+08	2.50E+07	1.90E+08
Aerobic tank sludge	1.10E+08	6.20E+07	1.50E+06	N.D.	2.70E+05	N.D.
Effluent	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.

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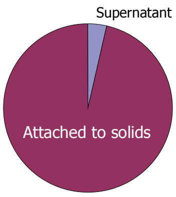
Location of Norovirus



Attached to flocks → Rejection by membrane

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Influence of in-line chemical cleaning

Elapsed time after chemical cleaning	*Norovirus* type	
	GⅠ	GⅡ
30min	N.D.	N.D.
60min	N.D.	N.D.
120min	N.D.	N.D.

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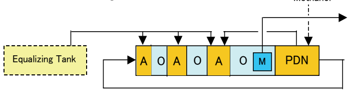
Summery of *Norovirus* removal

- High removal efficiency by MF MBR
- Adsorption of virus to sludge flocks is the reason of removal
- Gel layer on the membrane surface is not important for virus removal

Application of MBR to BNR process

Application of Membrane to Large-scale BNR Process

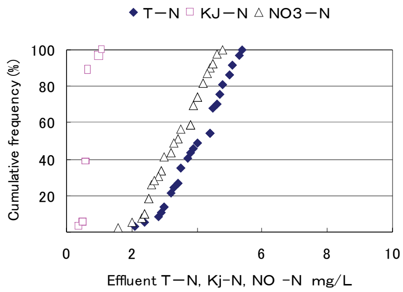
HRT 10hrs, T-Neff≦5mg/l



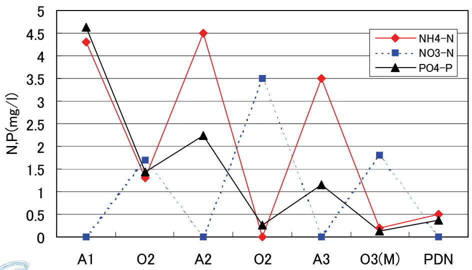
A: anoxic tank
O: oxic tank
PDN: post-denitrification tank
M: membrane

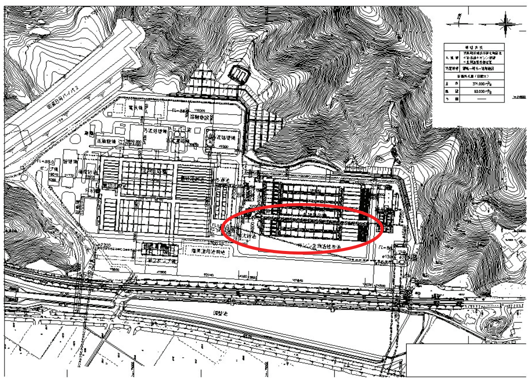
Pilot plant

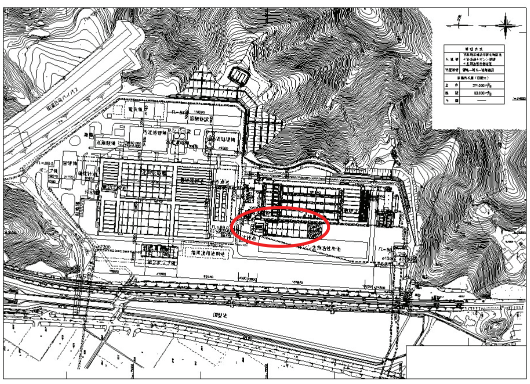
Nitrogen removal



Profiles in each tank

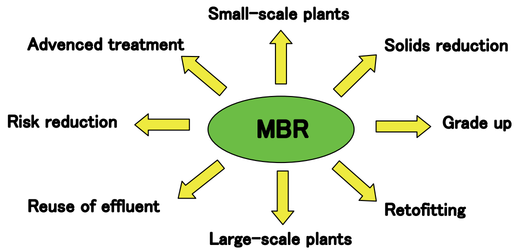






Perspective of future development of MBR

MBR as a core technology



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- Opimization of the design method
- Optimization of the maintenance methods, especially fouling control and chemical cleaning of membrane
- Reduction of membrane cost and prolongation of membrane life
- Reduction of energy consumption, especially supply air for membrane cleaning

are essential.

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Removal of EDCs by ozonation

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Purpose of the study

- To reveal the efficiency and characteristics of estrogen decomposition by ozonation in treated wastewater.

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Ozonation experiment

Object substances	17β-estradiol (E2) Estrone (E1) 17α-ethynl estradiol (EE2)
Initial concentration	20, 200ng/L
Ozone dose	0.5, 1, 3, 5, 7, 10mg/L
Contact time	15 minutes (fixed)

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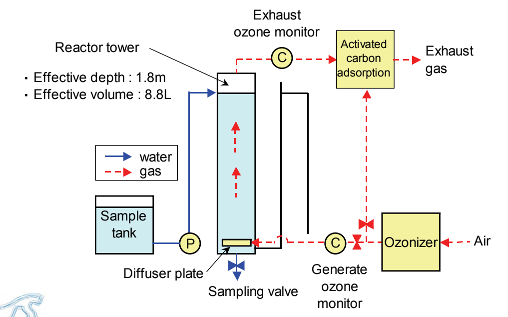
Experimental devices



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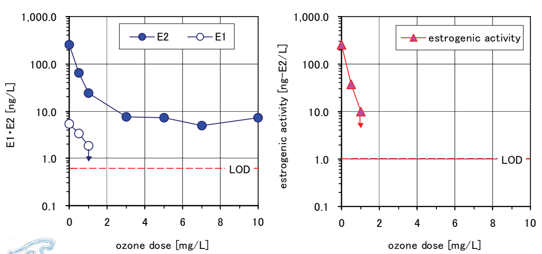
Changes of estrogen and estrogenic activity with ozone dose (1)



(Initial estrogen concentration : E2, 200ng/L)

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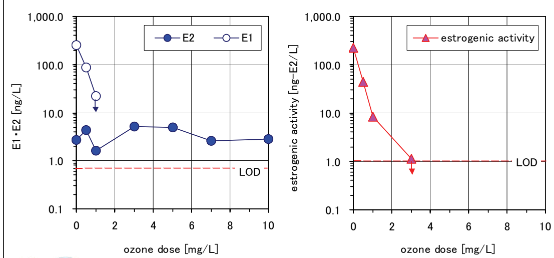
Changes of estrogen and estrogenic activity with ozone dose (2)



(Initial estrogen concentration : E1, 200ng/L)

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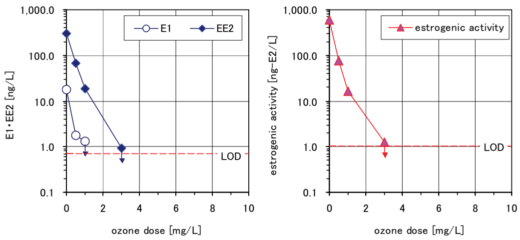
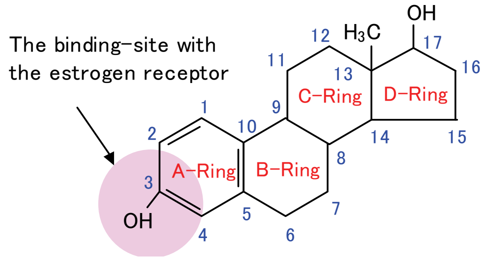
Changes of estrogen and estrogenic activity with ozone dose (3)



(Initial estrogen concentration : EE2, 200ng/L)

Chemical structure of E2



Initial concentration and kinetic constant

Kind of estrogen	Estrogen concentration		Estrogenic activity	
	Initial concentration P_0[ng/L]	Kinetic constant k[min^{-1}]	Initial concentration P_0[ng/L]	Kinetic constant k[min^{-1}]
E2	252	0.205	261	0.286
E1	253	0.171	226	0.236
EE2	299	0.191	589	0.250
E2	36.4	0.121	21.3	0.190
E1	48.4	0.130	37.4	0.156
EE2	18.1	0.115	18.0	0.112

Conclusions

- The removal efficiencies of estrogens and estrogenic activity were greater than 90% for an ozone dose of about 1 mg/L.
- Estrogens could be removed almost completely at an ozone dose of 5mg/L or less, which is a practical ozone dose level for disinfection.

Conclusions (continued)

- No byproduct with estrogenic activity was produced during the decomposition process of estrogen by ozonation.
- The kinetic constant *k* was not much influenced by the type of estrogen.

New biological nitrogen removal method -annamox process-

annamox (anaerobic ammonium oxidation)

$$NH_4 + 1.32NO_2^- + 0.066HCO_3^- + 0.13H^+ \rightarrow$$
$$1.02N_2 + 0.26NO_3^- + 0.066CH_2O_{0.5}N_{0.15} + 2.03H_2O$$

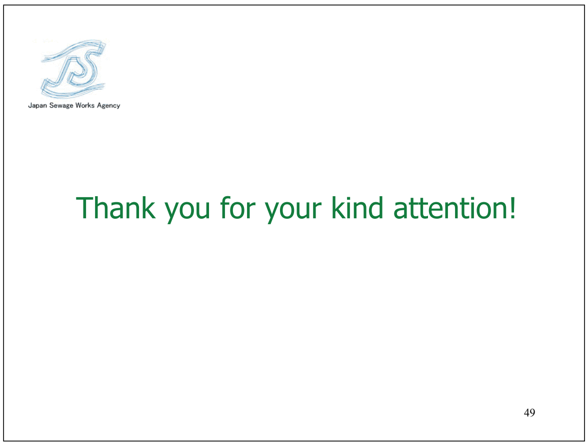
- Reduced oxygen requirement
- External carbon source is not necessary
- Small excess sludge production

Possible application of annamox process

Reduction of N load of sidestream from;

- Anaerobic digester
- Deodorization equipment of sludge drying or composting process

A pilot-scale experiment starts soon.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Thank you for your kind attention!
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