Korea J. Appl. Microbil Bioeng.
Vol. 8, No. 1, 61~73 (1980)

Studies on the Night Soil Treatment

Youki Ose, Ph. D.
Tetsuya Ishikawa, M. Pharm.
Takahiko Sato, Ph. D.

Gifu Pharmaceutical University Lab. of Environmental Hygiene
5-6-1, Mitahora-higashi Gifu City, Gifu
JAPAN

1. Introduction

In Japan, night soil had been used as fertilizer long time. It was a precious resources, but it was contaminated with epidemic bacteria and parasites, so caused diseases very often.

In Table I, the state for night soil treatment in Japan is shown. The population using the flash toilet was 37% in 1977. Flash toilet is connected to sewage system or to domestic treatment plant. The population use the storage type toilet was 63% and night soil stored in this type were treated by any plant. Only 2.6% of collected night soil was used as fertilizer, and the others were treated by several methods.

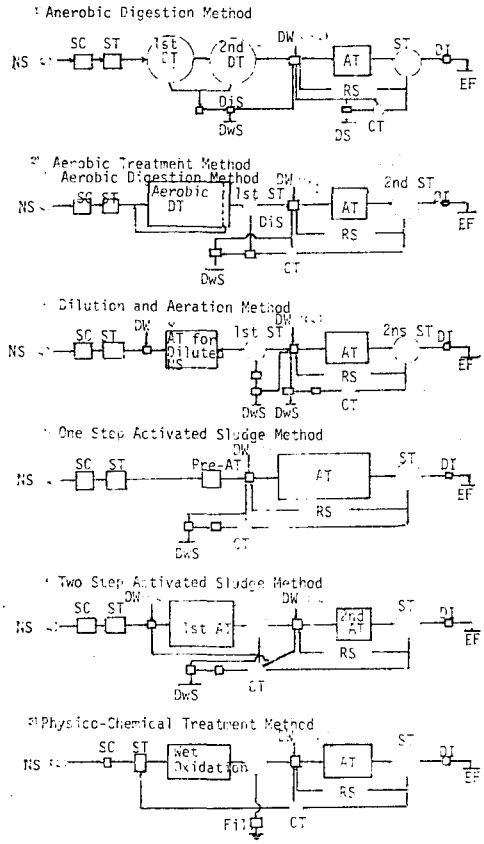
Since 1950s, night soil has not been used as fertilizer year by year, because chemical fertilizer had been produced and hygenic aspects. So the damping into sea was carried out.

Many govermental policy for treatment of night soil has been improved with the advance of techniques for treatment. In 1958, "Cleaning Act" was established. By this law, night soil treatment plant started to be built. In this year, Ministry of Health and Welfare published the standards for the construction and management of night soil treatment plant.

In 1970, "Cleaning Act" was revised and became to "Waste Treatment and Cleaning Act", and in

Table I · Yearly State of the Night Soil Treatment in Japan

	1963	1968	1973	1976	1977
Total Population in Treatment Plan (×1000 people)	58533	76080	106664	112589	113904
(%)	60.7	78.5	98.2		
Flash Toilet (×1000 people)	9836	17976	14787	39427	43169
Sewage System	5496	7980	13451	20387	21963
Domestic Treatment	4340	9999	28238	19040	21206
Don't Use the Flash Toilet	48697	58104	78426	73162	70735
	kl/D %	kl/D %	kl/D %	kl/D %	kl/D %
Total Volume of the Night Soil	64102 100	83963 100	108689 100	108680 100	107765 100
Collected Night Soil	53138 82.9	75047 89.4	85610 78.8	92836 85.4	92394 85.7
Fall into Sewage System	5281 8.2	5529 6.6	5307 4.9	7170 6.6	6973 85.7
Night Soil Treatment Plant	15137 23.6	47604 56.7	62901 57.9	69211 63.7	70060 65.0
Used as Fertilizer	6935 10.8	2444 2.9	4427 4.1	3540 3.2	2838 2.6
Fall into Sea	13122 20.5	14167 16.9	12975 11.9	12915 11.9	12523 11.6
The Others	12663 19.8	5303 6.0	—	—	—
Treated by Each House	10964 17.1	8916 10.6	23079 21.2	15849 14.6	15375 14.3



NS : Night Soil
St : Storge Tank
SC : Screen
DW : Dilution Water
AT : Aeration Tank
DT : Digestion Tank
RS : Return Sludge
CT : Concentration Tank
DiS : Digested Sludge
DwS : Dewatered Sludge
ST : Sedimentaiton Tank
DI : Disinfection
EF : Effluent

Fig. 1. Flow Sheet of Night Soil Treatment Plant

1977 Ministry of Health and Welfare published the new standards for the construction and management of night soil treatment plant.

Fig. 1 shows the standards for the construction of night soil treatment plant[1]. The standard methods consist from three types anaerobic digestion treatment method, aerobic treatment method and phisco-chemical treatment method.

Anaerobic system requires 30~60 days for digestion and the scale of tank require the large volume. Moreover, it must be heated to keep the temperature. So, this method is expensive to establish the plant.

As physico-chemical method, only wet oxidation treatment method is autorized.

Aerobic treatment method contains five types, that is, aerobic digestion method, aeration method for diluted night soil, one step activated sludge method, two step activated sludge method. In this method the degree of dilution is twenty fold or ten fold diluted.

Generally, treatment ability of activated sludge became lower in cold area, so the underground construction or heating installation for the plants are required to keep the temperature.

Aerobic digestion method can keep its treatment ability in cold area and not require the heating energy.

In early days anaerobic digestion method was used, but anaerobic digestion method is changing to aerobic treatment, because of the odorous pollu-

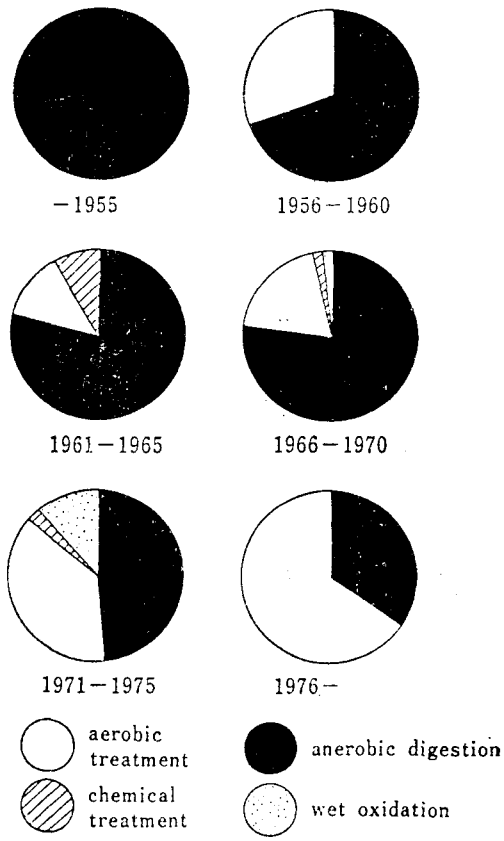


Fig. 2. The Percentage of Treatment Plant constructed in Each Period

tion and the high cost. Fig. 2 shows the percentage of treatment plant constructed in each period. Nowadays the most of the new plants are aerobic method. [2]

2. Components of night soil

The collected night soil contains not only night soil, but also paper, femal's menstral uses, and birth control uses and the other falling materials.

The night soil shows the different composition according to the meals, age and health condition. The duration and temperature of storage affect the components, too.

Table Ⅱ shows the standard composition of collected night soil[1].

Table Ⅱ. Standard Composition of Collected Night Soil

pH	7～9
BOD	13500mg/*l*
COD	7000mg/*l*
SS	21000mg/*l*
Total dried residue	30000mg/*l*
Total nitrogen	5000mg/*l*
Chloride (Cl^-)	5500mg/*l*
Phosphate	1000mg/*l*
Bacteria	10^8～10^9/m*l*
Coliform group bacteria	10^6～10^7/m*l*

Chlorine ion showed 5, 500ppm, and many researchers[3]~[6] indicated the same value, but recently it was found that the concentration of chlorine ion was reduced to 3, 500ppm[7] and the concentration of the other components was also reduced. This is caused by the mixing of excess sludge gathered from the domestic treatment plants[8].

In 1977, the total volume of treated night soil by treatment plant in Japan was 70, 060k*l*/d, and the domestic treatment plant sludge treated in night soil treatment plant was 10, 900k*l*/d. The ratio was about 7 : 1.

Nitrogen and phosphorus are fertilizer's components and these caused the eutrophication in lake and sea, and the scientific techniques for the removal of these substances are the most important subjects. Aerobic digestion can effectively remove the nitrogen. [9]~[11]

In night soil, many kind of bacteria have been found[12]. Almost of them are Enterobacteria, and epidemic bacteria canlive in them, too. [13]~[15] In 1962～63, we identified the *vibrio parahaemolyticus* from the collected night soil[16]. This bacteria cause the food poisoning. The seasonal pattern of isolation percentage from night soil and the number of patients in Japan had the close relation. After our publication, some researcher found these phenomenon, too. [14][18][19] The other researcher found *Salmonella* in the night soil. [14][2]

The components of BOD change day by day and the BOD value fluctuates. As shown in Fig. 3[21], particle fraction changed to soluble fraction, and the soluble BOD was removed by bacterial action.

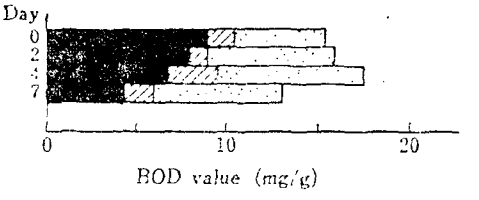


Fig. 3 Changes of BOD Values in Centrifuged Fractions

Each column in Fig. 2-a is indicated as follows:
: 120×*g* precipitate
: 67075×*g* precipitate
: 67075×*g* supernatant
} 1200×*g* supernatant.

BOD was mainly consisted from lower fatty acids. [21]~[26] The most of them is acetic acid, and the others are propionic, butyric and valelic acids. These lower fatty acids were utilized by bacteria and the BOD value was reduced.

The BOD values of contents in storage toilests were different, but the collected and mixed night soil showed about 13, 500ppm of BOD on the average. This is the basic value calculation for construction of the plant.

3. Treatment ability in the process of aerobic digestion

Undiluted night soil is falled into the digestion

tank and aerated for thirteen days. By the data of standard construction, the aeration ratio is $3m^3/m^3$ /d.[1] The temperature in the tank can be kept over 30°C, without heating even in tge cold season.

3.1 Removal of BOD

Fig. 4 shows the change of BOD at each parts of this treatment plant.[27] BOD was reduced in the first tank and the removal percentage is about 83% as mean value.

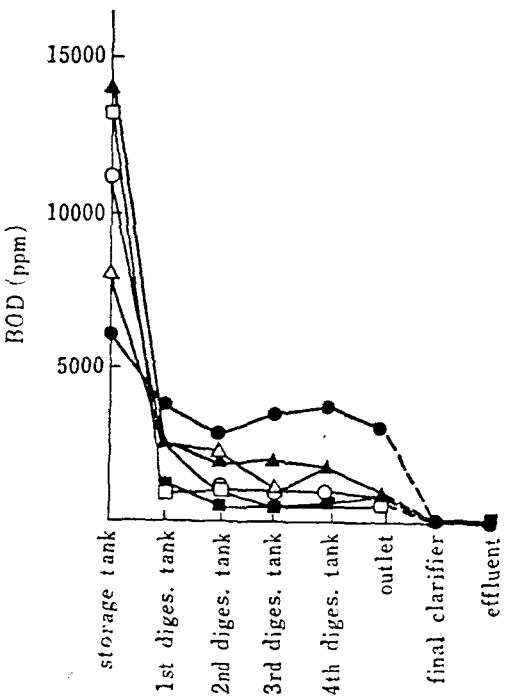


Fig. 4. Chango of BOD at Each Treatment Process of the Plant for Undiluted Night Soil

—○— : 13. Jan. —●— : 24. Jan.
—△— : 6. Feb. —▲— : 18. Feb.
—□— : 5. Aug. —■— : 2. Sep.

Fig. 5[28] shows the contrubution of soluble, fine and coarse particle's fraction in night soil and treated water to the BOD. BOD value in the soluble fraction vanished and that in fine particle's fraction did, too. The fraction of coarse particle decreased, but did not vanished. In the first tank,

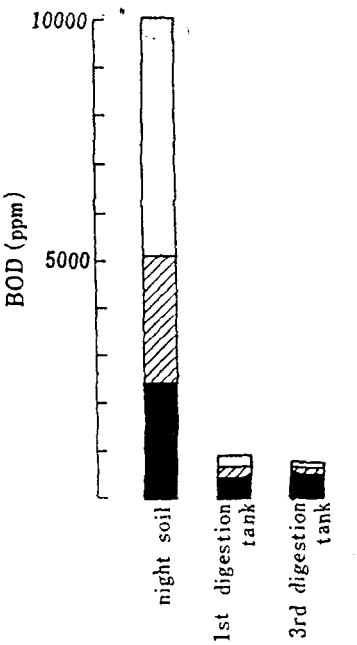


Fig. 5. Contribution of Night Soil and Treated Water Fraction to the BOD

■ : fraction of coarse particles.
▨ : fraction of fine particles.
□ : soluble fraction.

coarse and fine particle's fractions changed to soluble substances and were removed by bacteria.

Fig. 6[28] shows the elution diagrams of night soil and treated water on Sephadex G-25. These samples were supernatant of centrifuged at 12,000 $\times g$ *for 15 min.*

By aerobic treatment, the high peak in 120~180m*l* de creased, and this high molecular substances might change to low molecular substances. The high peak in 230m*l* decreased, too, and the peak in 360~400m*l* vanished completely. But the peak in 240~280m*l* remained unchanged and this phenomenon suggested that the fraction had non-degradability substances.

Fig. 7[28] shows the change of volatile fatty acid's concentration at each treatment process. Volatile fatty acids are main components of BOD and the concentraiton of the acids in storage tank is equivalent to about 5,000ppm of BOD.

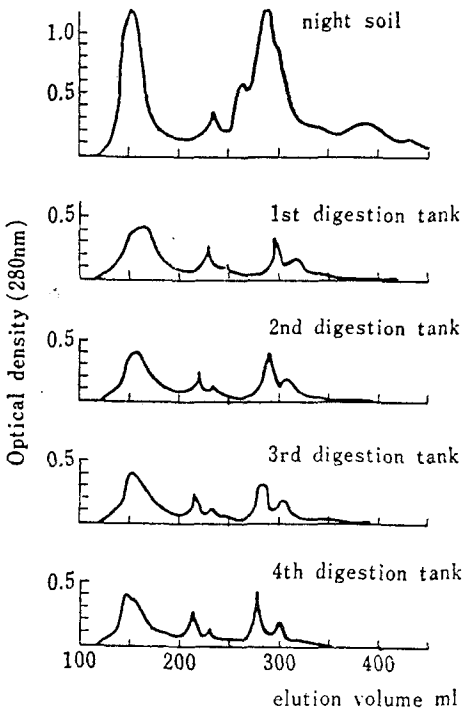


Fig. 6. Elution Diagram of Night Soil and Treated Water on Sephadex G 25

Solvent : 0.02% NaN_3.
Bed dimentions : 80×2 cm.
Sample : supernatant centrifuged at 1200×*g*.
Flow rate : 0.5 ml/min.

3. 2 Biological phase

In the first and second tank, only bacteria were found and protozoa connot be found, but protozoa can be found in the third and forth tank as shown in Table Ⅲ.[11]

Some researchers[10][29] and the authors[30] isolated and identified the bacteria in night soil treatment tank of the actual or experimental plant. But as shown in Table Ⅳ, the species are not same.

In the undiluted night soil digestion tank, only bacteria were found But in the activated sludge of ten fold diluted night soil treatment plant[31] and in activated sludge treated with anerobic digestion solution, protozoa were found.

In general, the identified bacteria bacteria in

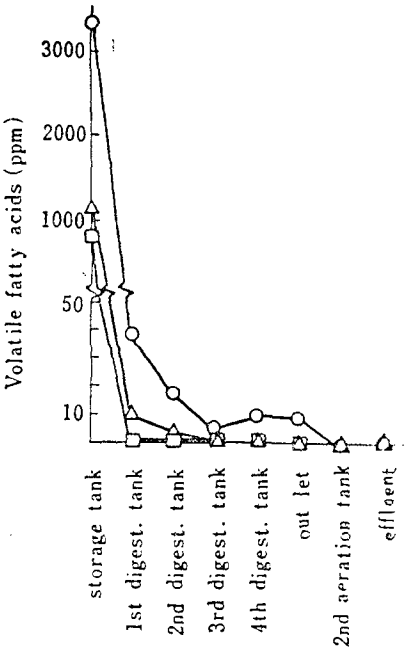


Fig. 7 Change of Volatile Fatty Acids Concentration at Each Treatment Process

—○— : acetic acid.
—△— : propionic acid.
—□— : butyric acid.

activated sludge were not same because the conditions of activated sludge were different, and the most suitable species in the tank will grow as the dominant bacteria. Like in activated sludge, bacteria phase in each aerobic digestion plant was not same.

The authors identified the three strains from the tank of undiluted night soil treatment plant,[30] and the removal pattern of mixed volatile fattyacids by those bacteria were tested.

Fig 8-1. shows the result by *E. coli communior*. After acetic acid was vanished, propionicacid was removed, but butylic acid remained.

Fig 8-2, shows the result by *Bacillus megaterium*. Acetic acid could be removed but propionic and butylic acids could not.

Fig. 8-3 shows the result by *Aerobacter aeragenes*. This strain was isolated as acetic acid-utilizing-bacteria by Z. Inoue.[34] In night soil, acetic acid can be found so high concentration that he tried to isolate the bacteria by the medium, which had the acetic acid as the only carbon source. This

Table Ⅲ. Biologic Examination of Digestion Tank

	3 rd Diges. tank	4t h Diges. tank	2n d aeration tank	Final clarifier
Bacteria sp.	++++	++++	++++	++++
Zoogloea sp.	++++	++++	++++	++++
Flagellatae sp.	++++	++++	++++	++++
Vorticella sp.	++++	++++	++++	++++
Opercularia sp.	++++	++++	++++	++++
Monas sp.	++++	++++	++++	++++
Roteria sp.	++++	++++	++++	++++
Nematoda sp.	++++	++++	++++	++++
Oscillatoria sp.	++++	++++	++++	++++
Diatomeae sp.	++++	++++	++++	++++
Chlorella sp.	++++	++++	++++	++++
Scenedesmus sp.	++++	++++	++++	++++
Hormidium sp.	++++	++++	++++	++++
Stiaeoclonium sp.	++++	++++	++++	++++

Table Ⅳ Bacterial phase in Aerobic Digestion Tank

Identified Specis	Isolated from	Researcher	Literature
Bacillus cerus var maycodis			
Clostridium Tertium			
Escherichies coli Ⅰ *α*			
Escherichias coli Ⅱ *α*	in laboratiory	Kodama	10
Slaphylococcus intreus			
Streptococcus faecalis			
Bacillus subtilis			
Achronobacter pestilleu			
Bacillus magaterium	plant	Ishikawa et al	30
E. coli communior			
Stoplyecoccus epidemid			

Table Ⅴ. Dissimilation Rates of Organic Acids by Bacteria

Name of bacteria	Dissimilation rates (%)						
	Folmic acid	Acetic acid	Propionic acid	Butyric acid	Valeric acid	Pyruvic acid	Oxalic acid
E. coli	90.4	41.1	49.0	−	−	58.1	−
Bacillus megaterium	+	20.3	−	−	9.1	13.1	+
Propionic acid Utilized bacteria	−	50.0	62.9	38.2	35.8	36.9	−

+ : oxygen uptake was detected but could not be determined.
− : oxygen uptake was not detected.

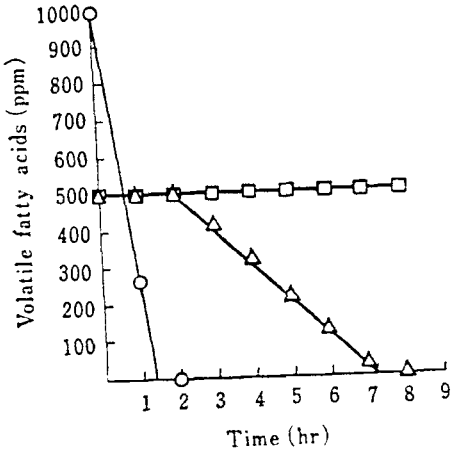


Fig. 8-1. Removal of Volatile Fatty Acids by *E. coli* var. *communior*

—○— : acetic acid.
—△— : propionic acid.
—□— : butyric acid.

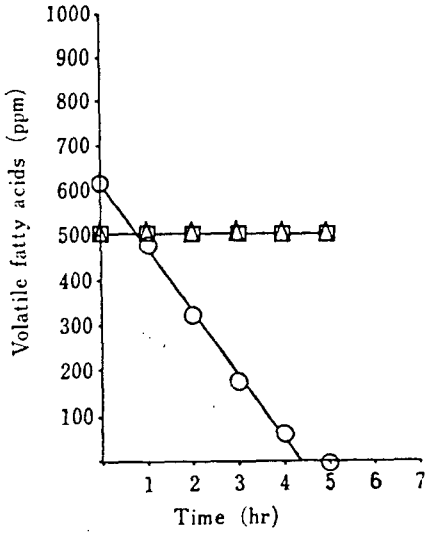


Fig 8-3 Removal of Volatile Fatty Acids by *Aerobacter aerogenes*

—○— : acetic acid.
—△— : propionic acid,
—□— : butyric acid.

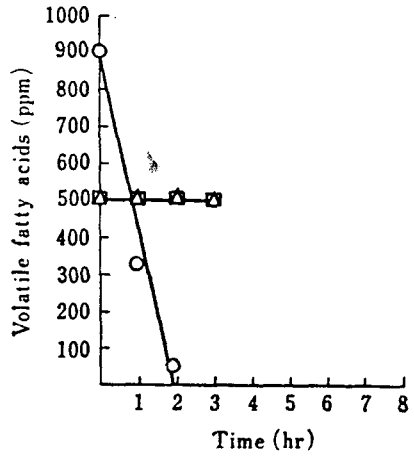


Fig. 8-2. Removal of Volatile Fatty Acids by *Bacillus megaterium*

—○— : acetic acid.
—△— : propionic acid.
—□— : butyric acid.

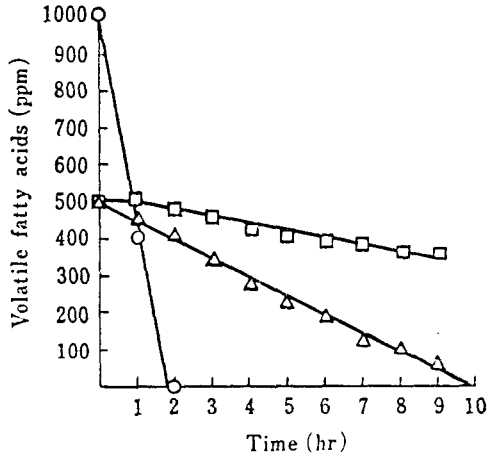


Fig. 8-4 Removal of Volatile Fatty Acids by Propionic Acid Utilized Bacteria

—○— : acetic acid.
—△— : propionic acid.
—□— : butyric acid.

bacteria could remove only acetic acid, and not the other acids.

Fig. 8-4 shows the result by propinic acid-utilizingbacteria. This bacteria was isolated as the same method as acetic acid-utilizing-bacterium by the authors.[30] This bacteria could remove these three acids, but the removal rates were different.

The dissilulation rates of organic acids by bacteria are shown in Table V.[30] Mckinney[35], Placak[36] and Sato,[37] one of the authors, reported the dissimulation rate of acetic acid by activated sludge as 40~97%.

The removal rates of volatile fatty acids in the first tank were mesued. The removal rate for acetic acid was 19.6mg/MLSS/hr, and this rate by the activated sludge in sewage treatment plant was

Table Ⅵ Percentage Oxidation and Utilization of Amino Acibs by Bacteria

Amino Acids	Utilization and Percentage Oxidation	
	B. megaterium	*E. coli var. communior*
L-Alanine	75.2%	64.7%
L-Valine	+	—
L-Leucine	+	—
L-Methionine	—	—
L-Cystine	—	—
L-Aspartic acid	+	+
L-Glutamic acid	+	+
L-Lysine HCl	—	+

+ : O_2-uptake was detected but Percentage Oxidation could not be calculated.
— : O_2-uptake was not detected.

35.9mg/MLSS/hr. In the undiluted night soil treatment plant, the plant was running with good removal of BOD in spite of the removal rates of fatty acids were low, and the concentrations of fa tty acids were high. This mechanism was supposed to be owed to the high number of bacteria. (38)

InTable Ⅵ, the rates of the amino acids oxidation by the isolated are shown. (30)

Some bacteria can ozidize the some amino acid, but the other amino acid cannot utilized.

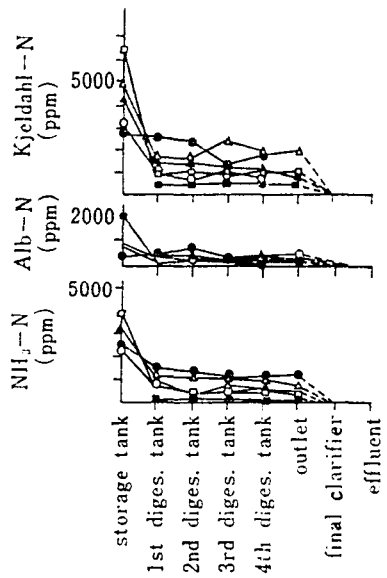


Fig. 9. Change of Nitrogen in Digestion Tank

—○— : 13. Jan. —●— : 24. Jan.
—△— : 6. Feb. —▲— : 18. Feb.
—□— : 5. Aug. —■— : 2. Sep.

3.3 Nitrogen

As shown in Fig. 9, nitrogen was well removed in the first tank. Most of the removed nitrogen was due to NH_3-N, and albuminoid-N. (11)

The contribution of soluble, fine and coarse particle's fractions in night soil and treated water to the nitrogen aee shown in Fig. 10. In soluble and fine particle's fractions, like BOD, nitrogen was

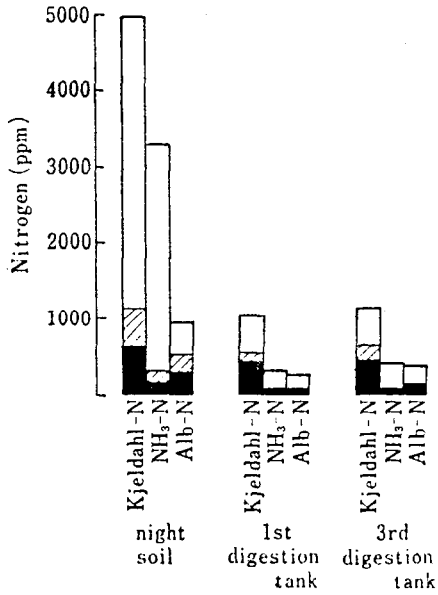


Fig 10. Contribution of Night Soil and Treated Water Fraction to the Nitrogen

■ : fraction of coarse particles.
▨ : fraction of fine particles.
□ : soluble fraction.

removed, but was not reduced in the coarse particle's fraction. The removal rates of Kjeldahl-N, albuminoid-N and NH_3-N were 88%, 92%, and 92%, respectively.[28] Nowadays, denitrification is very important subject from the point of prevention of the eutrophication, and this removal results are very useful.

In aerobic digestion tank, pH was in the range of 7.5~8.9 and the temperature was over 30°C, so ammonia stripping will occur.[11] Murata detected 3,000 ppm of NH_3-N in air duct connected to aeration tank.[39]

In the aerobic digested solution, NO_2-N was produced as 50ppm in winter and its concentration reached up to 350ppm in summer[11]. This NO_2-formation was affected by the loas of nitrogen and by the aeration time, but BOD-oxidazing-bacteria an and denitrifying bacteria were not affected by the formed NO_2.[40]

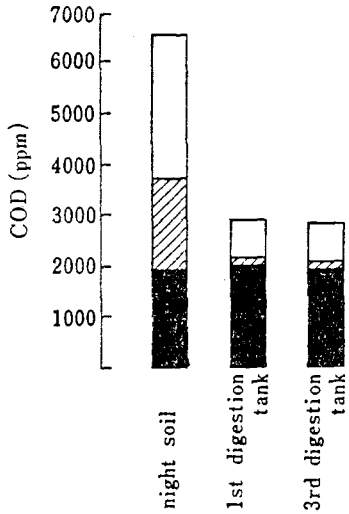


Fig. 11. Contribution of Night Soil and Treated Water Fraction to the COD

: fraction of coarse particles.
: fraction of fine particles.
: soluble fraction.

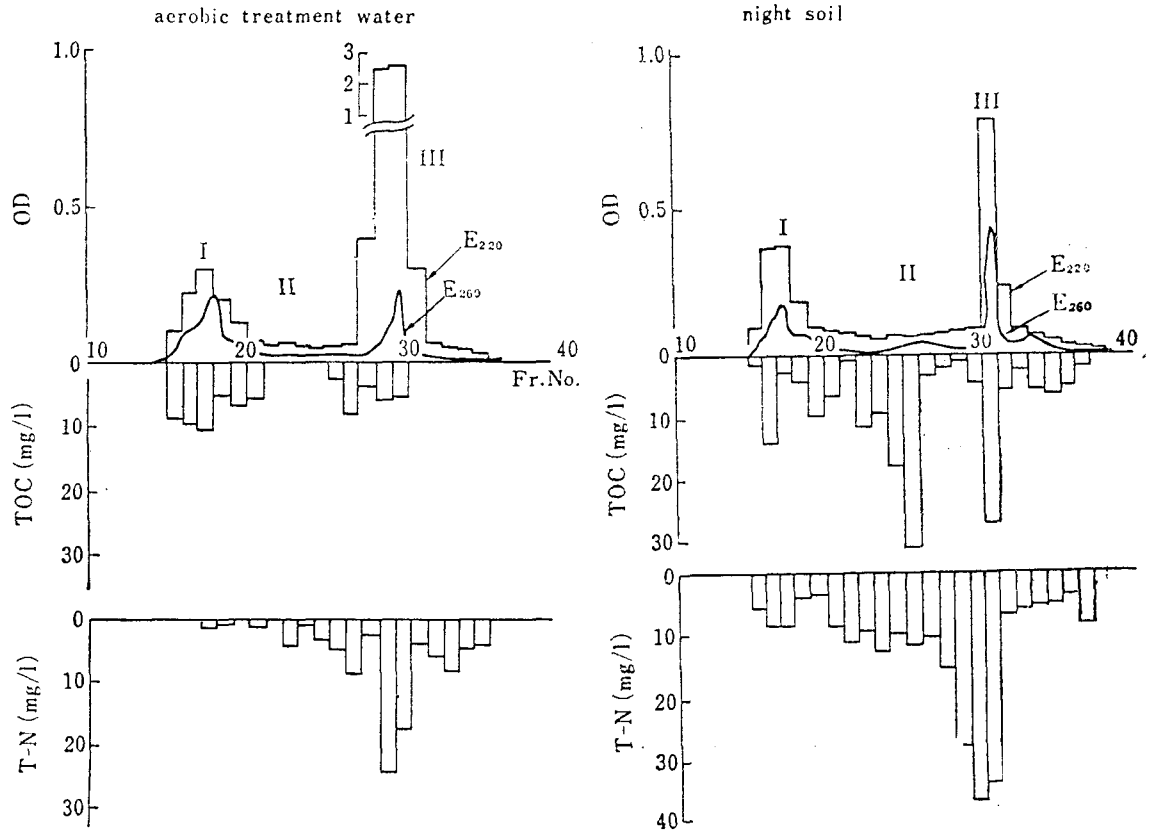


Fig. 12 Elution Diagram of Night Soil and Aerobic Treated Water on Sephadex

3.4 Removal of COD substances

In night soil, BOD value is 1.5~2 fold of COD value. By the biological tre atment, BOD substances are easily removed and COD substances are not so well removed. After the biological treatment, BOD value is 1/3 value is 1/3 of COD value[9].

As shown in Fig. 11, COD substances in soluble and fine particle's fraction was removed but those in coarse particle's fraction was not. [11]

Chigusa[40] reported the elution diagram of night soil and aerobic treated effluent by Sephadex as shown in Fig. 12.

Fraction I was colored and the COD value of this part took half values to total COD. By aerobic treatment, fraction II was well removed, and Fraction III increased in correspondence with NO_2 and NO_3 formation. 1ppm of NO_2-N was equivalent to 11.7ppm of COD. [41] Thus, the COD components in the treated solution mainly owed to the color substances and NO_2.

4. Residual substances in effluents

The authors identified the residual substances in the effluent after the aerobic digestion by GC-MS. [42]

Fig. 13 shows reconstructed ion chromatogram or total ion monitor chromatogram by GC-MS of ether extracts of night soil, effluent from undiluted night soil treatment plant, the effluent from anaerobic digestion treatment plant and the activated tank of anaerobic night soil treatment plant.

Table VII shows the detected compounds by GC-MS. In collected night soil, indole, methylindole and oxindole, that is the odorous substances, were

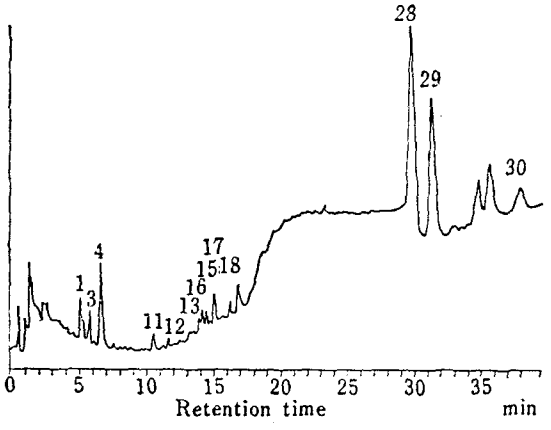


Fig. 13-1 Reconstructed Ion Chromatogram by GC-MS of Ether Extracts from Night Soil

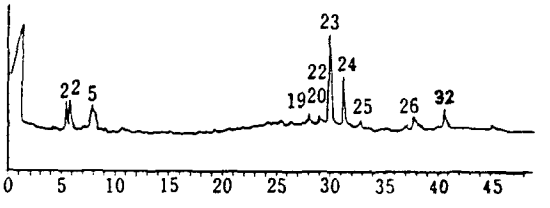


Fig. 13-2 Total Ion Monitor Chromatogram by GC-MS of Ether Extracts from Effluent of Undiluted Night Soil Treatment Plant

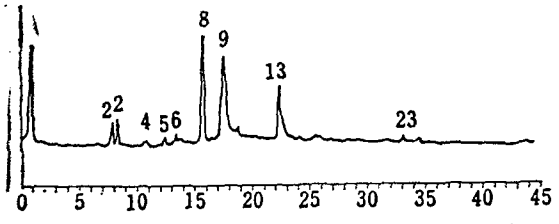


Fig. 13-3 Total Ion Monitor Chromatogram by GC-MS of Ether Extracts from Digested Night Soil

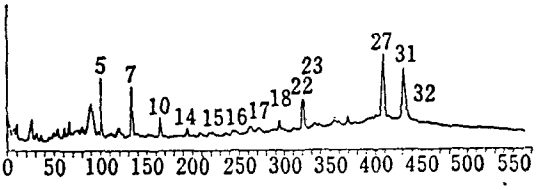


Fig. 13-4 Total Ion Monitor Chromatogram by GC-MS of Ether Extracts of Effluent from Aeration Tank of Anaerobic Night Soil Treatment Plant

Table Ⅶ Detected Compounds in Collected Night Soil and in the Effluents from the Treatment Plants

Peak No.	Compounds	M. W.	Collected Night soil	Effluent[a]		
				1	2	3
1	Phenol	94	++			
2	Ethyl hexenate	142		++	++	
3	Dichlorobenzene	146, 148	++			
4	Cresol	138	++		+	
5	Undecane	156		++	+	++
6	Cyclohexane carboxylic acid	128			+	
7	Dodecane	170				++
8	Ethyl phenylacetate	164		+	+++	
9	Phenylacetic acid	136			+++	
10	Tridecane	184				+
11	Indole	117	+			
12	Methylindole	131	+			
13	Oxindole	133	+		++	
14	Tetradecane	198				+
15	Pentadecane	212				+
16	Hexadecane	226	+			+
17	Heptadecane	240	+			+
18	Octadecane	254	+			+
19	Di-i-butyl phthalate	278		+		
20	i-Butyl-*n*-butyl phthalate	278		+		
21	Nonadecane	268	+			
22	Methyl palmitate	270		+		++
23	Dibutyl phthalate	278		+++	+	++
24	Ethyl palmitate	284			++	
25	Butylhexyl phthalate	306		+		
26	Diheptyl phthalate	362		+		
27	Dioctyl adipate	370				+++
28	Coprostanol	388	++++			
29	Cholesterol	386	++++			
30	Sitosterol	414	+++			
31	Dioctyl phthalate	390		++		+

a) Effluent 1 : effluent from undiluted night soil treatment plant.
Effluent 2 : effluent from digested night soil treatment plant.
Effluent 3 : effluent from aeration tank of anaerobic night of anaerobic night soil treatment plant.

detected. The oxindole was identified as the first report by the authores. Coprostanol, that is index substance of night soil, cholesterol and sitostanol were found. Some metabolites of amino acids, chemicals used in toilet, aliphatic carbon hydrates and some environmental pollutants were also found. After the treatment, indole and methyindole vanished, but dichlorbenzene remained.

5. Chloroform formation in the effluents by chlorination

Chloroform has the carcinogenic toxicity. In 1974, Symons[43] found chloroform in all the tested drinking water of eighty American cities and the control of chloroform in drinking water became

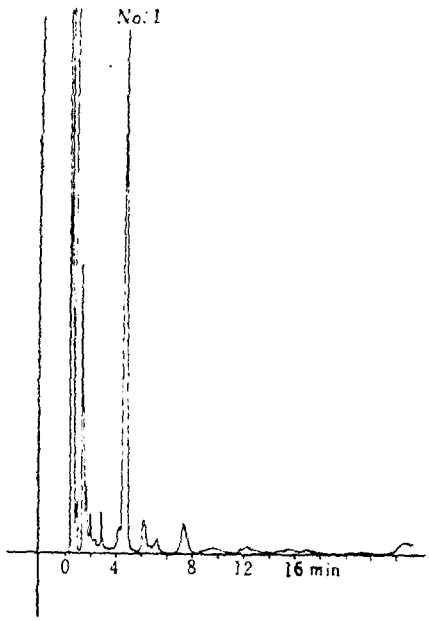


Fig 14-1 Gas Chromatogram of Chlorinated Effluent of Night Soil Treatment Plant
No1 is chloroform

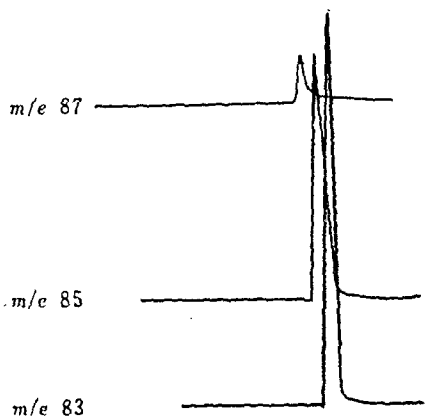


Fig 14-2 MID (GC-MC) Chromatogram of Chloroform

very important. It was found that the one of the chloroform-forming-substances in water was humic substance.[44]~[46] In the effluent discharged from the night soil treatment plant, the existance of this substance was supposed. And the authors[47] detected the chloroform as shown in Fig. 14. Chlorform was identified by MID chromatogram.

6. Conclusion

In Japan, the scientific technology for the night soil treatment is developing and many plants are running. In Korea, the most difficult points to use a biological treatment method is the atmospheric condition, that is, the low temperature in winter. Undiluted method is suitable for cold area because this method can keep the temperature over 30°C without heating even in winter. In Japan, the researchers for this method are increasing, and the papers are publishing. In this paper, the authors review this method and introduse the biological reaction in the treatment plant.

Literature

1) Ministry of Health and Welfare of Japan "Standards for the constraction of night soil treatment plant". 1977
2) Odor Control Association of Japan "Odor Control for night soil". Tokyo, 1977, p. 13
3) I. Horasawa: J. Japan Water Works Assoc. **211**, 35 (1952)
4) A. Hoda, J. Japan Water Works Assoc. **260**, 41 (1956)
5) T. Miyanohara: J. Water and Waste (Japan). 8, 642 (1966)
6) Y. Ose, M. Kiriyama, S. Watanabe, S. Tani and T. Hirota: EISEI KAGAKU. **18**, 393 (1972)
7) T. Terada, T. Takahashi: SEIKATSU TO KANKYO. **20**, (6), 92 (1977)
8) S. Ohono: J. Water and Waste (Japan). **21**, 3 (1979)
9) N. Muto: Japan. J. Hyg. **13**, 287 (1958)
10) T. Kodama, N. Muto, K. Yagome, S. Ohno, Y. Shigeta and H. Matsumoto: Kanagawa-ken Eisei Kenkyu-sho Hokoku. **9**, 111 (1959)
11) Y. Ose, T. Ishikawa, T. Sato, Y. Ikuine, M. Kohno, A. Yamada, S. Watanabe and S. Tani: EISEI KAGAKU. **19**, 189 (1973)
12) E. Hiraoka, H. Kishi, H. Ohmori, S. Kawanishi, T. Yamayoshi and T. Ono: Japan

J. Pub. Health. **20**, 467 (1973)
13) R. Takano: "History of Toilet". Koseikaku, Tokyo, p. 142 (1943)
14) S. Akabane, Y. Asakawa, S. Nakatsugawa, M. Murakami and M. Noguchi: Japan J. Pub. Health. **20**, 473 (1973)
15) A. Honda, Z. Inoue, T. Hotta, M. Kobayashi and R. Sasaki: Water Purif. Liauid Wast Treatment (Japan). **5**, (5), 37 (1964)
16) Y. Ose and T. Ikeda: J. Food Hyg. (Japan). **5**, 206 (1964)
17) Y. Ose, T. Ikeda, Y. Furuyama and I. Takagi: J. Food Hyg. (Japan). **6**, 513 (1965)
18) T. Kato, H. Ohara, S. Yamai, K. Hobo et al: Kanagawa-ken Eisei Kenkyu-sho Hokoku. **17**, 57 (1967)
19) T. Nakamura: Modan Media (Japan). **15**, (4), 201 (1969)
20) S. Akiyama: Media Circle (Japan). **14**, (8), 399 (1969)
21) Y. Ose, K. Nakamuro and M. Sugiura: EISEI KAGAKU. **19**, 264 (1973)
22) S. Hashimoto: HAKKO KAGAKU KAISHI. **37**, 425 (1959)
23) A. Honda, T. Nakadoi, Z. Inoue and H. Watanabe: J. Japan Water Works Assoc. **301**, 67 (1959)
24) I. Endo: J. Japan Water Assoc. **337**, 85 (1962)
25) Y. Tezuka: J. Water and Waste (Japan). **6**, 297 (1964)
26) Y. Ose, R. Funasaka and T. Sato: EISEI KAGAKU. **19**, 239 (1973)
27) S. Ohno, I. Takahashi, T. Sakurai et al: Water Purif. Liquid Wast Treatment (Japan). **6**, 71, 21 (1965)
28) T. Ishikawa, Y. Ose and T. Sato: EISEI KAGAKU. **19**, 277 (1973)
29) S. Hashimoto and I. Matsumaru: HAKKO KAGAKU KAISHI. **38**, 409, 520 (1960)
30) T. Ishikawa, J. Fukuyama, Y. Ose and T. Sato: Ann. Proc. Gifu Coll. Pharm. **26**, 8 (1978)
31) N. Saruta, S. Ishinishi and T. Inoue: J. Water and Waste (Japan). 4, 691, 825 (1962)
32) I. Morishita: Water Purif. Liquid wast Treatment (Japan). **3**, (7), 23 (1962)
33) T. Ishikawa, Y. Ose, T. Sato and Z. Inoue, EISEI KAGAKU. **24**, 226 (1978)
34) A. Inoue, A. Honda and T. Ishikawa: Water Purif. Liquid Wast Treatment. **15**, 223 (1974)
35) Mckinney, R. E. Jeris: J. S. Sewage and Ind. Waste. **27**, 728 (1955)
36) Placak, O. R., Ruchhoft C. C.: Sewage Works J. **19**, 423 (1974)
37) T. Sato: Water Purif. Liquid Wast Treatment Treatment (Japan). **6**, (6), 19 (1965)
38) T. Ishikawa, Y. Ose and T. Sato: Water Res. **13**, 681 (1969)
39) K. Murata, Y. Yamada and H. Tsubono: Water Purif. Liquid Wast Treatment (Japan). **18**, 651 (1977)
40) K. Chigusa, H. Takemoto: PPM. (11), **25** (1978)
41) T. Ishikawa, Y. Ose and T. Sato: EISEI KAGAKU. **25**, 19 (1979)
42) T. Ishikawa, Y. Ose and T. Sato: "Proceedigs of 5th Symposium of Enironmental Pollutants and their Toxicology". p. 23 (1978)
43) T. M. Symons, T. A. Beller, J. K. Carswell et al: JAWWA. **67**, 634 (1975)
44) J. J. Rook: Environ. Sci. Technol. **11**, 478 (1977)
45) K. Kajino: J. Japan Water Works Assoc. **514**, 17 (1977)
46) B. Tomita, N. Hamamura and Y. Ose: Water Purif. Liquid Wast Treatment (Japan). **20**, 153 (1978)
47) T. Ishikawa, Y. Ose, T. Sato and B. Tomita: EISEI KAGAKU. **24**, 235 (1978)