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Effluent limitations through best practicable control technology: evaluation of the semiconductor and electronic component manufacturing industry

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ABSTRACT

The effluent limitations of industrial wastewaters are based on uniform regulatory criteria for effluent discharge facilities in Korea. However, an individual effluent limitation for each effluent discharge facility is being applied for regulation of industrial wastewater in US and EU. This study is conducted to propose effluent limitations for the semiconductor and electronic component (SEC) manufacturing industry on the basis of the best practicable control technology (BPT) currently available reflecting on the wastewater characteristics. BPTs were evaluated using conventional pollutants including biological oxygen demand (BOD), chemical oxygen demand (COD), suspended solids (SS), total nitrogen (TN), and total phosphorus (TP). We found BPT for semiconductor manufacturer in Korea was coagulation and sedimentation → biological treatment → activated carbon filter → and sand filter. Based on this BPT, effluent limitations for the SEC manufacturing industry in Korea were BOD_5 with 1.81 mg/L, COD_{Mn} with 1.22 mg/L, SS with 0.55 mg/L, TN with 13.98 mg/L, and TP with 0.05 mg/L, respectively. All of these values were substantially lower than current effluent limitations. This result indicates that effluent limitations need to be separated for industrial sectors in Korea.

Keywords: Effluent limitations; Best practicable control technology currently available (BPT); Semiconductor and electronic component (SEC) manufacturing industry; National pollutants discharge elimination systems (NPDES)

1. Introduction

The characteristics of industrial wastewaters vary due to the differences in manufacturing processes, raw materials used in productions, the abilities of environmental management, and related wastewater treatment methods. Treatment results may be substantially different depending on manufacturing facilities even in the same industrial sector. The effluent limitations have been derived and implemented differently according to the industrial sectors in the US and in Europe [1,2].

In the US, Environmental Protection Agency classified the industrial sectors into 500 subcategories

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over 56 categories, and suggested guidelines for separate effluent limitations for those categories to manage and reduce industrial water pollutants efficiently [1,3]. Long-term treated water quality data were statistically analyzed to establish technology-based effluent limitation (TBEL). In estimating effluent limitations, variability factors are reflected on 95% (daily maximum concentration) or 99% (monthly average) to consider the variation width. After introducing best practicable control technology (BPT) on the basis of TBEL in 1977, water quality was substantially improved. In 1972, US Government reported that only 30–40% of the assessed waters met water quality goals. This percentage increased into 60–70% in 2002.

In Europe, according to the Integrated Pollution Prevention and Control (IPPC) [2,4], industrial sectors are classified into 6 categories: energy, metal production and processing, mineral industry including ceramics and glass manufacturing, chemical industry, waste management, and others. Best Available Technique (BAT) is determined based on the industrial sector, and the most appropriate Emission Limit Values for individual manufacturing facilities are approved [4]. The BAT concept was integrated into the Industrial Emissions Directive, one of the most important directives to reduce pollution [5]. Dijkmans [6] successfully applied BAT for vehicle refinishing sector and manure processing sector. More recently, Berzosa et al. [7] proposed a new method to assign responsibilities of greenhouse gas emission to consumers as well as producers, and BAT played an important role in their approach.

Effluent limitations have been implemented to control pollutants discharged from industrial facilities as a regulatory measure for accomplishing environmental standards in Korea. Water Quality Conservation Act of Korea designated 42 pollutants including organic matters, suspended solids (SS), metals, and volatile organic compounds (VOCs), and classified 4 regions according to the regional water quality levels [8]. However, aforementioned approaches considering industrial sectors in the US and Europe are not constitutionally reflected in Korea, and uniform effluent limitations and same pollutants are being applied to all industrial sectors. Recently, Korean experts in water quality and water resources management have pointed out that the application of uniform effluent limitations is not appropriate to efficiently respond rapidly changing industrial environment [9–12]. Korean Government has also strived to improve related institutions including the application of BAT or BPT currently available [12].

For this reason, several researchers in Korea have proposed technologies or techniques similar to those of US or EU establishing effluent limitations based on wastewater treatment technology. Kim et al. [13] analyzed foreign BAT evaluation systems, and suggested BAT evaluation procedures including selection of candidate technology, economical acceptability, and evaluation of technology parameters. Kim et al. [14] proposed the procedure establishing effluent limitations based on wastewater treatment technology for individual wastewater discharging facilities of pulp, paper, and paper board manufacturing industry. This procedure includes the evaluating procedure of wastewater treatment technology applicable in Korea. More recently, Kim et al. [9] suggested technology-based effluent limitations on the basis of the BPT for petroleum refining industry. However, these efforts are yet fragmental and inadequate to persuade policy-makers for establishing separate effluent limitations reflecting on the wastewater characteristics of individual industrial sectors. Thus, this study was conducted to propose new effluent limitations based on the wastewater characteristics of industrial sectors using BPT in Korea. Semiconductor and electronic component (SEC) manufacturing industry was selected for TBEL estimation in this study.

2. Materials and methods

2.1. Semiconductor and electronic component industry in Korea

The SEC manufacturing industry is one of the most important in all industrial sectors in Korea. Korea is the largest SEC manufacturer in the world. The total number of SEC manufacturing facilities was 957 in 2010 in Korea [15]. This industry is the biggest discharger of wastewaters among 82 industrial sectors in Korea. The percentage of SEC manufacturing facilities is only 1.98%, but the amount of discharged wastewater per day is 740,086/d, 20.4% of total amount of wastewater discharged to water body (Table 1). The manufacturing processes in SEC manufacturing industry include washing, surface cleansing, and etching. These processes requires extremely high precision, and generates various conventional and hazardous wastes [16]. Large amount of water used with organic solvents during these processes, and then discharged to water bodies as the wastewater containing conventional water pollutants, and toxic materials such as metals and VOCs. In the US, HF and toxic organic solvents used as etchant or cleansing agent as well as the conventional water pollutants such as pH, oil and grease, and BOD_5 are being regulated. In addition, metals including Sn, Cr, Cu, Ni, and Pb, and VOCs including toluene, methylene chloride, and

Table 1
Characteristics of wastewater generated and discharged from SEC manufacturing industry in Korea [15]

	Numbers of facility	Amount of wastewater generate in facilities (m^3/d)	Amount of wastewater discharge to water body (m^3/d)
Total	48,266	5,229,147	3,446,857
Semiconductor and electronic components industry	957	740,086	702,209
Ratio(%) via total	1.98	14.2	20.4

dichlorobromomethane are also being regulated for SEC manufacturing industry [17].

However, as uniform effluent limitations are being applied for all industrial sectors in Korea, long-term data are not yet managed and collected to evaluate the applicability of effluent limitations for metals and VOCs in SEC industry. Accordingly, only conventional water pollutants were considered for proposing BPT and TBEL in this study.

2.2. Applicable wastewater treatment technology

In the US, pH control → lime and chlorinate calcium (removed fluorine) → coagulation with polymer → filtration and sorption were proposed as BPT for cathode ray tube industry, one of the subcategories of SEC industry [17].

In Korea, coagulation and sedimentation process was suggested using calcium salts to remove fluoride in discharged wastewaters caused by HF used during manufacturing processes of SEC industry [18]. Reverse osmosis membrane method was also suggested for recycling treated wastewater [19]. In order to treat wastewater from SEC industry, multiprocesses for wastewater treatment including physical and chemical treatment, reverse osmosis, activated carbon filter (or sand filter) is additionally required as well as traditional biological treatment [20].

2.3. General steps for implementing effluent limitation

General steps to establish individual effluent limitations for discharging facilities can be consisted of three stages: (1) data survey, (2) technology assessment, (3) examination of current limitation[9]. In the data survey stage, the data on types and concentrations of pollutants, types and levels of applied treatment technologies are collected. Water quality data collected from national watershed monitoring networks near the discharge facilities are investigated, and then potentially dischargeable pollutants are figured out by the notified data on the amount of pollutants from the facilities. Additional site investigation and water quality analysis can be possible.

At the technology assessment stage, effluent limitations need to be determined whether technology based (TBEL) or water quality based (WQBEL) on the basis of wastewater characteristics evaluated in the first stage. TBEL-based method is characterized as its thorough analysis of the industry types including treatment facilities, as well as effluent characteristics [13]. Long-term water quality data are commonly used for deriving limitation values with BPT level through statistical analysis such as regressional analysis [21] and 95 percentile approach [9,22]. The methodology for establishing WQBEL contains the principle of risk assessment. In WQBELs, 3 methods have been widely used. Method 1 uses water quality monitoring data. Ambient water quality criteria for the protection of human health and aquatic organisms are used for establishing effluent limitations in method 2. Supposing steady state and complex mixed condition, method 3 uses concentration of pollutants and water quantity discharged from facilities, in lower and upper streams [23]. In accordance with the anti-backsliding principle, established limitations cannot be exceeded the current limitations, and more strict limitation among TBEL and WQBEL is adopted, in principle.

Institutional legitimacy and technical feasibility for established limitations are re-examined at the final examination of current criteria stage. Effluent limitations are determined and notified to the competent authority, Regional Environmental Office of Korea (REOK). After that, final effluent permission reflecting on the final effluent limitations is issued by REOK through examining the presented limitations. More details on these steps can be found elsewhere [21,25].

2.4. Establishment of TBEL in Korea

Effluent limitations based on the BPT were derived. Effluents from 19 big SEC facilities were analyzed. Even though various heavy metals and VOCs were detected in the effluents, this study focused on conventional water pollutants because long-term monitoring data on other toxic materials are not yet enough to derive any effluent limitations.

The first step to achieve this goal was to collect water quality data from the facilities currently being operated properly, and then TBEL were estimated in the BPT level based on statistical analysis. On the basis of the data collected, concentration values corresponding to 95% (X_{95}) and 99% (X_{99}) in the lognormal distribution were considered as the limitation values for the monthly average, and daily maximum, respectively. Those values are applied in accordance with the characteristics of discharge facilities in the US, while the representative values of treated water quality are estimated by the average values measured twice per 30 min in water quality investigations without applying average water quality concepts in Korea.

In this study, the daily maximum value (X_{99}) was applied for estimating TBEL considering the stability of the treated water quality. Variability factors (VF) were also applied to reflect the variability of treated water quality on establishing effluent limitations [24].

2.5. Analysis of wastewater characteristics

Grab samples from 19 SEC manufacturing facilities were collected using polyethylene bags for 14 metals, 5 conventional water pollutants and 1,4-dioxane and phenols, and glass bottles with Teflon caps for VOCs analyses. Those materials selected in this study are classified and presented in Table 2. Inductively coupled plasma-mass spectrometry (ICP-MS) was used for analyzing metal contents in treated wastewaters, and gas chromatography (GC) with purge and trap was used for VOCs analyses. Various conditions and parameters in ICP-MS and GC are summarized in Table 3.

2.6. Evaluation of effluent limitation based on wastewater treatment technology

Among those materials mentioned above, conventional water pollutants including biological oxygen demand (BOD), chemical oxygen demand (COD), SS, total nitrogen (TN), and total phosphorus (TP) were selected for establishing TBELs based on BPT. Average concentrations and variability factor of conventional pollutants from 19 SEC manufacturing facilities were estimated. The treatment technology which represented the best removal efficiency for 5 conventional pollutants was selected as the BPT for SEC industrial facilities in Korea. Finally, TBELs were estimated as multiplying average concentrations of BPT applied facilities with variability factor.

$$\text{ELs} = \text{average concentrations of BPT} \times \text{VF}$$

3. Results and discussions

3.1. Wastewater characteristics

Not only conventional water pollutants which were used for BPT estimation, but also various pollutants including metals and VOCs were found in the effluents discharged from SEC facilities (Table 4). Concentrations of metals and VOCs were similar to those reported in previous studies [25,26]. These materials are not yet being regulated for the same industrial sector in Korea. We previously mentioned that long-term monitoring data on other toxic materials are not yet enough to derive any effluent limitations, and those materials were not included to determine BPT in this study. This is another reason that other materials were not considered.

3.2. Determination of BPT for SEC manufacturing industry

Wastewater treatment technologies currently being used in 19 SEC manufacturing facilities selected in this study were summarized in Table 5. Wastewaters discharged from other industrial facilities were firstly collected from all unit manufacturing processes, and

Table 2
Analysis of toxic water pollutants in treated wastewater from SEC manufacturing facilities

Items		Pollutants
Conventional water pollutants		BOD, COD, SS, TN, TP
Toxic water pollutants	Heavy metals	Cu, Cr, Cr^{+6}, Mn, Cd, Fe, Ni, Zn, As, Se, Sn, Ba, Pb, CN
	VOCs	TCE, PCE, 1,1-dichloroethene, dichloromethane, chloroform, 1,1,1-trichloroethane, carbon tetrachloride, benzene, bromodichloromethane, toluene, ethylbenzene, dibromochloromethane, m,p,o-xylene, bromoform, 1,2-dichloroethane, 1,1-dichloroethylene,
	Others	1,4-dioxane, phenols

Table 3
C/MS operating conditions for analysis of VOCs

Item	Condition
P&T	Velocity xpt purge and trap, Teledyne Tekmar
Trap	Tekmar trap K(vocarb 3000), Supelco
Pre perge	30 seconds
Purge	11 minutes at 40℃
Dry purge	40℃
Desorbe	4 minutes at 250℃
Bake	6 minutes at 250℃
GC/MS	7890A/5975C, Agilent Technologies
Inlet temperature	220℃
Split ratio	20:1
Column	DB-624(Agilent J&W GC Columns), 30 m × 0.320 mm ID × 1.80 μm
Column temperature	5 minutes at 35℃
	20 minutes at 195℃ (8℃/min)
	4min at 245℃(25℃/min)
	DB-624 (Agilent J&W GC Columns), 30 m × 0.320 mm ID × 1.80 μm
Column flow	1.4 mm/min
Carrier gas	H_2 (99.999%, flow rate 60 psi)

Table 4
Concentration of heavy metals and VOCs in treated wastewater from SEC manufacturing facilities

Non-conventional water pollutants (heavy metals and VOCs)	Concentration (mg/L)
Copper	0.008–0.804
Manganese	0.629
Iron	0.390
Nickel	0.245
Tin	0.032
Barium	0.11
Chloroform	0.002
Dichloromethane	0.09
Toluene	0.001–0.96
Ethylenebenzene	0.001
Xylene	0.001
Phenols	0.007–0.021

then treated together (i.e. Pulp, paper and paper board manufacturing facilities [14] and petroleum refining facilities [9]. In SEC manufacturing facilities, however, wastewaters generated in unit manufacturing processes were firstly treated individually, and collected and treated together at the final treatment stage. Basic treatment methods usually being applied in SEC manufacturing facilities include biological treatment using activated sludge, physicochemical treatments such as coagulation and sedimentation. Various individual treatment processes such as pH control, 1, 2 coagulation and sedimentation, sand or activated carbon filter, H_2O_2 oxidation, chelate are additionally applied according to the differences in concentrations and types of pollutants, and permit standards of individual discharging facilities.

In order to establish TBEL, the average concentrations of conventional water pollutants from 19 selected SEC manufacturing facilities were analyzed, and the results are summarized in Table 6. The average values were 31.2 mg/L for BOD_5, 23.1 mg/L for COD_{Mn}, 5.5 mg/L for SS, 16.4 for TN, and 0.7 for TP, respectively. Even though those facilities are manufacturing same products, they are discharging various wastewaters according to the differences in unit manufacturing processes. Thus, the concentrations of conventional pollutants were substantially different according to individual manufacturing facilities as different treatment technologies were applied for individual facilities on the basis of the wastewater characteristics from those facilities.

3.3. Establishment of TBEL on the basis of BPT

TBELs for conventional water pollutants in wastewaters discharged from SEC manufacturing facilities were estimated using BPT suggested in previous sections in this study, and the results are summarized in Table 7. After the final treatment, the average concentrations of conventional pollutants in wastewaters discharged from 19 facilities were as follows: BOD_5 was 31.2 mg/L, COD_{Mn} was 23.10 mg/L, SS was 5.2 mg/L, SS was 5.2 mg/L, TN was 14.3 mg/L, and TP was 0.6 mg/L, respectively. Then, we estimated the variability factors for the conventional pollutants based on the data on water quality analysis. The VF of BOD_5 was 1.24, COD_{Mn} was 1.72, SS was 1.37, TN was 2.44, and TP was 2.63. Thus, effluent limitations for BOD_5 can be estimated as follows:

$$\text{Average concentration of BPT applied facilities} \times \text{VF} = 1.46 \times 1.24 = 1.81$$

Other parameters were calculated same way. As a result, COD was 1.22, SS was 0.55, TN was 13.98, and TP was 0.05. Those values were substantially lower than current effluent limitations for individual pollutants. The relatively lower removal efficiency of TN was observed as no specific nitrogen removal processes were installed.

Table 5
Wastewater treatment processes on each SEC manufacturing facility

Items	Wastewater treatment processes
1	Coagulation and sedimentation
2	Coagulation and sedimentation + activated carbon filter
3	Coagulation and sedimentation + activated sludge
4	H_2O_2 oxidation + activated sludge
5	1st coagulation and sedimentation + 2nd coagulation and sedimentation + biological treatment
6	Coagulation and sedimentation + chelate + activated sludge + activated carbon filter
7	Coagulation and sedimentation + biological treatment + activated carbon filter + sand filter

Fig. 1 represents the concentration distributions of conventional pollutants in treated wastewater discharged from 13th facility, which was selected BPT applied. The number of samples for each category is different as SEC manufacturing facilities excluded those results from samples with extremely high values. This rule was also applied for previous studies on BPT for different industrial sectors in Korea [9,14]. Concentrations of those pollutants in treated wastewaters were below current effluent limitations, and maintained stable conditions. The concentrations of organic materials in SEC manufacturing wastewaters were generally lower than those discharged from the facilities in other industrial sectors.

The wastewater treatment processes of 13th facility, which represented the best water treatment quality among those 19 facilities, were selected as BPT in this study. The wastewater treatment processes of the facility were as follows: coagulation and sedimentation → biological treatment (contact oxidation) → activated carbon filter → sand filter (Fig. 2). Compared with the treatment processes in BPT for the petroleum refining industry determined by Kim et al. [9], those in SEC manufacturing industry are more complicated. This result indicates that the numbers and kinds of pollutants in SEC wastewaters are more various than those in the wastewaters from petroleum refining facilities.

Table 6
Average concentrations of conventional water pollutants in treated wastewater discharged from 19 individual SEC manufacturing facilities

	Average concentration (mg/L)				
Facility	BOD_5	COD_{Mn}	SS	Total nitrogen	Total phosphorus
1	65.1	65.8	–	25.1	2.70
2	–	8.2	3.0	20.4	0.16
3	–	4.3	5.3	29.7	1.07
4	3.9	6.6	1.6	17.4	0.45
5	2.9	4.8	1.5	18.9	0.03
6	41.7	78.4	19.3	31.5	0.23
7	11.4	4.3	1.6	22.2	3.70
8	186.3	76.6	38.4	48.9	1.19
9	–	37.7	5.2	11.7	0.29
10	2.2	4.6	1.4	4.3	0.10
11	–	5.6	0.8	9.0	0.04
12	–	4.2	1.5	17.6	–
13	1.46	0.7	0.4	5.7	0.02
14	5.6	10.1	4.4	5.2	0.40
15	1.2	8.1	2.0	17.3	–
16	42.7	22.2	–	2.4	0.20
17	63.0	87.3	4.0	6.8	0.20
18	5.6	6.7	2.0	12.7	0.80
19	3.2	2.0	0.4	4.4	–
Average	31.2	23.1	5.5	16.4	0.7
Min	–	0.7	0.4	2.4	0.02
Max	186.3	87.3	38.4	48.9	3.7

Table 7
Case study of TBELs for SEC manufacturing industry

Best practicable control technology currently available in category	Items	Limits (mg/L)				
		BOD_5	COD_{Mn}	SS	TN	TP
pH Neutralization ↓	Total average concentration in category	31.20	23.10	5.20	16.40	0.60
Coagulation and sedimentation ↓	Variability factor(VF)	1.24	1.72	1.37	2.44	2.63
Biological treatment ↓	Lower average concentration in category (“A” facility)	1.46	0.71	0.40	5.73	0.02
Activated carbon filter ↓	Technical-based effluent limitations (TBELs)	1.81	1.22	0.55	13.98	0.05
Sand filter	Current effluent limitations	30	40	30	30	4

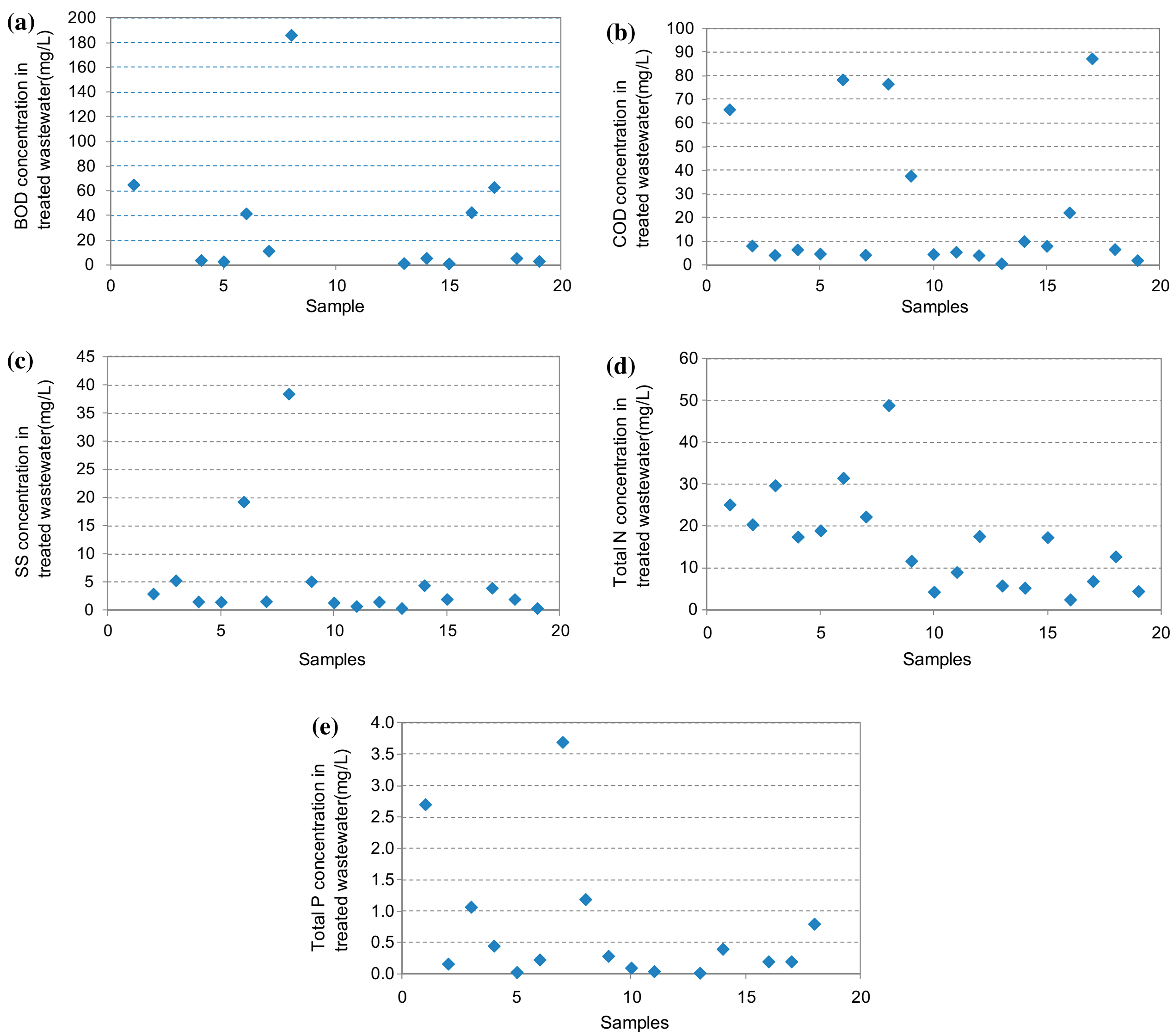

Fig. 1. Concentrations of conventional pollutants in treated wastewater from 13th facility: (a) BOD_5, (b) COD_{Mn}, (c) SS, (d) Total N, and (e) Total P.

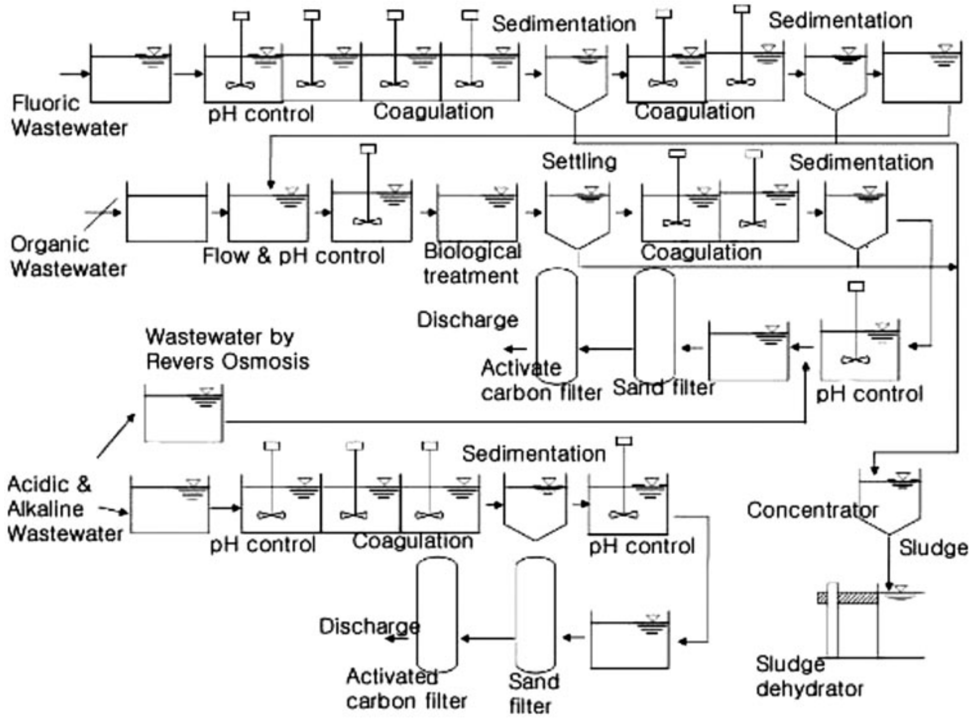

Fig. 2. Diagram of wastewater treatment processes as BPT currently available of semiconductor and electronic components industry.

Sometimes BAT is used as TBEL. However, BAT evaluation method considers a series of elements including environmental aspects (i.e. atmosphere and waste materials), description of used techniques, benefit assessment such as total amount of energy consumption, secondary effects, and legal applicability. Thus, economical aspects were brought up by the reference facilities where the techniques have already been applied [27]. Accordingly, BAT is not appropriate as estimation of effluent limitations in this study were only based on the current treatment technology and treatment levels, and other evaluation factors were not considered.

4. Conclusions

We derived BPT-based effluent limitations for SEC manufacturing industry, and proposed the effluent limitations on the basis of BPT derived in this study. The BPT for SEC was coagulation and sedimentation → biological treatment (contact oxidation) → activated carbon filter → sand filter. The proposed TBELs for SEC industry were 1.8 mg/L for BOD_5, 1.2 mg/L for COD_{Mn}, 0.6 mg/L for SS, 14.0 mg/L for TN, and 0.05 mg/L for TP, respectively.

It is not appropriate to apply the BPT suggested in this study for all other SEC manufacturing facilities in Korea as the kinds and concentrations of pollutants in wastewaters vary according to the individual discharging facilities, and the components of treatment plants. Operating conditions are designed for the treatment of specific wastewater for each facility. In order for TBEL to be implemented, the institutional complement applying different effluent limitations for various industrial sectors reflecting on the wastewater characteristics and treatment technology levels. In addition, various metals and VOCs were detected from discharged wastewaters of SEC manufacturing facilities even though they are not yet being regulated in Korea. The amounts and kind of these metals and VOCs in Korea were similar to those in the US which are currently being regulated properly.

If the application systems of uniform effluent limitations can be properly revised, and BPT can be applied to the individual discharging facilities, we observed that we can remove pollutants from SEC facilities more efficiently than the effluent limitations currently being used in Korea.

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