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# Prediction of the Transport and Fate of Polycyclic Aromatic Hydrocarbons (PAHs) in Masan Bay, South Korea

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A numerical simulation of the polycyclic aromatic hydrocarbons (PAHs) in Masan Bay, Korea, was conducted using a program written in STELLA to estimate the transport of organic chemicals in the coastal environment. A sensitivity analysis of dissolved PAHs and PAHs in suspended particulate matter (SPM) indicated that PAH levels were significantly influenced by the settling velocity, the adsorption and desorption rates. PAHs in phytoplankton were sensitive to all coefficient. The mass balance model indicated that the standing stocks of PAHs in water, in SPM, and in phytoplankton were $2.01\times10^{4}$ g, $1.15\times10^{3}$ g, and $7.5\times10^{-1}$ g, respectively. Flux were characterized by a high rate of desorption, followed by descending rates of adsorption, settling, and flow to the open sea. The scenario predicted a "safe" concentration level of dissolved PAHs of less than 30 ng/L if the input loads from rivers, the atmosphere, or both rivers and the atmosphere were reduced to 50%, 50%, or 30% of the current levels, respectively.

**Keywords**: PAHs, Mass–balance model, Masan Bay, Fate, Simulation

## INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are pollutants that are found extensively in marine ecosystems throughout the world. These hydrocarbons comprise at least two 5–6 carbon aromatic rings (Eisler, 1987). PAHs are insoluble persistent environmental pollutants that are generated by the incomplete combustion of fossil fuels. PAHs are introduced mainly from human activities rather than by natural processes (Badger, 1962).

The National Institute of Environmental Research (Korea) classifies PAHs as persistent bio–accumulative and toxic substances (PBTs) and the United Nations Economic Commission for Europe (UNECE) classifies PAHs as persistent organic pollutants (POPs), a group that comprises 16 substances: aldrin, chlordane, dieldrin, endrin, heptachlor, hexachlorobenzene, mirex, toxaphene, dichloro diphenyl trichloroethane (DDT), polychlorinated biphenyls (PCBs), dioxins, furans, chlordecone, PAHs, hexabromobiphenyl, and Hexachlorocyclohexane (HCH) (UNECE, 1998). POPs are characterized by toxicity, persistence, bioaccumulation, and long–range environmental transport.

Studies in many countries have examined PAHs levels in rivers, estuaries, and marine environments that were polluted by urbanization and industrialization activities. For example, studies have been conducted on the Mississippi River in the US (Mitra and Bianchi, 2003), the Elbe River in Germany (Gotz *et al*., 1998), the Seine River in France (Fernandes *et al*., 1997), the Jiulong River in China (Maskaoui *et al*., 2002), and the Daliao River watershed in China (Guo *et al*., 2007). Lee *et al*. (2005) conducted a study that focused on the river that feeds Masan Bay in South Korea. Studies of coastal seawater have been conducted in England and Wales (Law *et al*., 1997), on the Baltic Sea (Witt, 1995), on the Aegean Sea (Maldonada *et al*., 1999) and in Daya Bay (Zhoua and Maskaoui, 2003) and Xiamen Bay (Zhou *et al*., 2000) in China. The inflow of pollutants into the Mokpo coastal region (Moon *et al*., 2007) and the Young–il Bay area in Korea have also been investigated (Moon *et al*., 2001). Thus, the temporal and spatial distributions of PAHs have been extensively examined. However, many of these studies emphasized the distribution of PAHs and did not assess the risks associated with the presence of PAHs, nor did they address the fate of the PAH pollutants. The aim of the present study was to develop a predictive model for the PAHs distribution in Masan Bay.

PAHs cause both acute and chronic toxicity at relatively low concentrations, and the environmental pollution caused by PAHs can damage humans through the food web. Thus, removal of PAH pollution has become a major concern (Juhasz and Naidu, 2000). To address the health issues posed by the accumulation of POPs and PBTs pollutants, we have conducted a numerical simulation of the PAH levels in Masan Bay using a computational model written in STELLA to predict the transport properties and the ultimate fate of organic chemicals in the coastal environment (Kim *et al*., 2004).

## MATERIALS AND METHODS

### Study area

A large harbor in the southern part of Korea, the Masan Bay is a semi–enclosed embayment surrounded

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by two cities of Changwon and Jinhae (Fig. 1). And Masan Bay receives a wide range of industrial and sewage discharges and wastes from shipping activity. And this area is one of the important area in Korea since this area have long history of contamination (MOMAF, 2002). Due to the increase in the input loads of domestic and industrial wastewater as well as industrial and sewage discharges and wastes generated by the rapidly growing large population attracted to the development of the Masan Free Export Zone and the formation of Changwon Machine Industrial Complex in the 1970s, the average COD concentration of Masan Bay from 1999 to 2004 showed the quality of water which is involved in level Ⅲ of Water Quality Criteria in Sea Area. During the summer season, the Masan Bay Area suffers from degraded water quality that causes oxygen deficiency and red tides at the sea bottom (MOMAF, 2005). To improve the marine environment, the Special Masan Bay Management Plan was established, and the Total Pollution Loads Management System (TPLMS) has been launched and in operation.

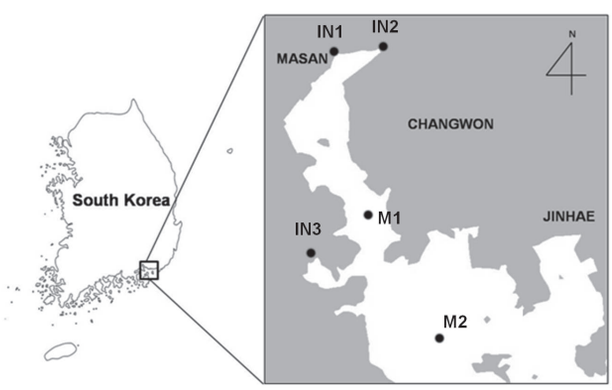


**Fig. 1.** Map showing the sampling station (M1, M2) and discharge from river and WWTP (IN1, IN2, and IN3).

## Model composition

Figure 2 presents a model written using STELLA. PAHs are mainly transported to the marine environment via two routes: 1) atmospheric deposition or 2) rivers. Other input sources are urban runoff from domestic or industrial wastewater and spillage of petroleum of petroleum products by ships (Boehm and Farrington, 1984). For simplicity, only the loading flow of PAHs from the rivers and the atmosphere were included in this model.

The PAHs flowing into the Masan Bay area are subject to several chemical and physical processes. This model examined the tidal outflow of dissolved PAHs into the open sea, photolysis of dissolved PAHs, adsorption onto and desorption from suspended particulate matter (SPM), and the settling of sediment. State variables consisted of dissolved PAHs, PAHs in SPM, and PAHs in phytoplankton. Table 1 presents the equations applied in this model.

## Estimates for the input data

The input data applied in this study are listed in Table 5. The volume, area, and cross–sectional area in our model were derived from the input data in the sea water flow field model by You (2009) and You *et al.* (2009). The grid length of x and y was each 500 m and volume, area, and cross–sectional area were calculated from depth data of Masan Bay using grid depth data divided into 5 levels at 4 m intervals on the vertical water column. The input loads considered in this model were the dissolved PAHs from the river input loads, the PAHs

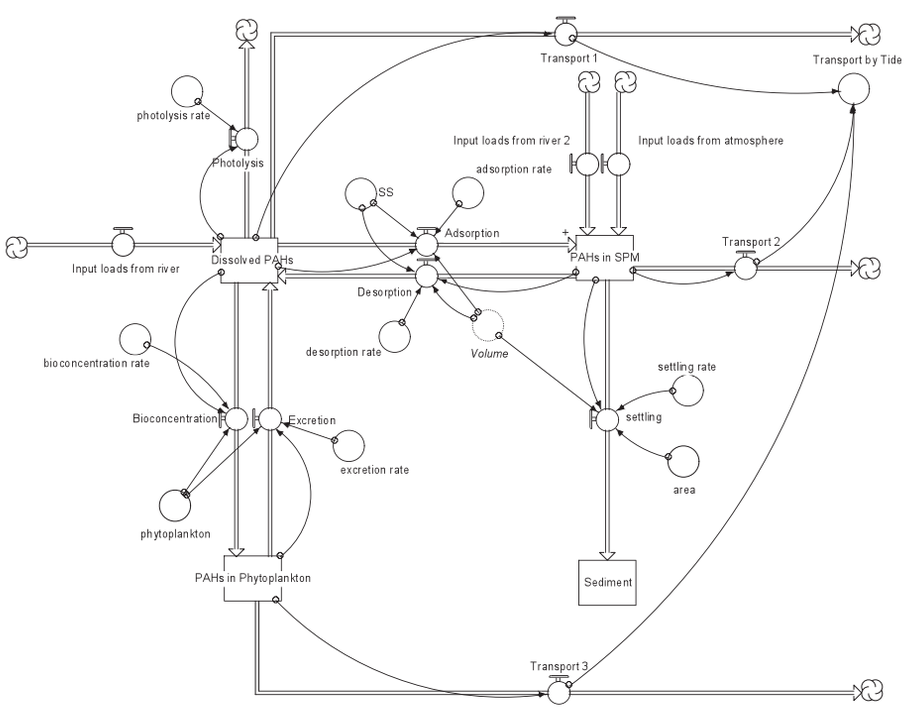


**Fig. 2.** The conceptual diagram of PAH fate in the model.

**Table 1.** Equations used to construct the model

Dissolved PAHs(t)=Dissolved PAHs(t–dt)
+ (Input loads from river – Adsorption + Desorption
– Photolysis – Bioconcentration of Phytoplankton
+ Excretion from Phytoplankton – Transport1)*dt

PAHs in SPM(t) = PAHs in SPM(t–dt)
+(Input loads from river + Input loads from atmosphere
+ Adsorption – Desorption – Settling – Transport2)*dt

PAHs in Phytoplankton(t) = PAHs in Phytoplankton(t–dt)
+(Bioconcentration of Phytoplankton
–Excretion from Phytoplankton–Transport3)*dt

Photolysis = Photolysis * Dissolved PAHs
Adsorption = Adsorption rate * Dissolved PAHs * SS
Desorption = Desorption rate * PAHs in SPM * SS
Bioconcentration = Bioconcentration rate * Dissolved PAHs * Phytolankton
Excretion = Excretion rate * PAHs in phytoplankton * Phytoplankton
Settling = Sinking velocity * PAHs in SPM
Transport1 = Transport rate by tide * Dissolved PAHs
Transport2 = Transport rate by tide * PAHs in SPM
Transport3 = Transport rate by tide * PAHs in Phytoplankton

Input loads from river
Input loads from atmosphere
Transport by tide
Phytoplankton = Standing stock of phytoplankton
SPM = Standing stock of Suspended Particulate Matter
INIT Dissolved PAHs = Initial concentration of Dissolved PAHs
INIT PAHs In SPM = Initial concentration of PAHs in SPM
INIT PAHs In Phytoplankton = Initial concentration of PAHs in Phytoplankton

**Table 2.** Concentration of PAH compounds discharge from the river and WWTP into Masan Bay

| PAHs | Dissolved PAHs (ng/L) | | | PAHs in SPM ($\mu$g/kg dry wt.) | | |
|---|---|---|---|---|---|---|
| | IN1 | IN2 | IN3(Duckdong WWTP) | IN1 | IN2 | IN3(Duckdong WWTP) |
| Nap | 16.52 | 54.74 | 13.34 | 2327.56 | 1653.32 | 775.35 |
| AcPy | 0.87 | 1.28 | 0.43 | 10.21 | 10.91 | 6.63 |
| AcP | 0.58 | 3.36 | 1.10 | 33.53 | 40.70 | 21.98 |
| Flu | 1.89 | 24.59 | 4.33 | 176.56 | 213.49 | 99.96 |
| PhA | 7.06 | 36.21 | 13.05 | 917.59 | 1672.74 | 820.47 |
| Ant | 0.25 | 1.06 | 0.45 | 10.14 | 13.86 | 10.42 |
| FluA | 1.89 | 0.23 | 2.65 | 498.06 | 2302.02 | 1082.16 |
| Pyr | 2.19 | 5.79 | 3.59 | 413.86 | 1869.42 | 833.29 |
| BaA | ND | ND | ND | ND | ND | ND |
| Chr | ND | ND | ND | 131.19 | 495.88 | 117.60 |
| BbF | 0.23 | 0.46 | ND | 1673.24 | 621.01 | 183.54 |
| BkF | ND | 0.24 | ND | ND | ND | ND |
| BaP | 0.09 | 0.29 | 0.21 | ND | ND | 32.26 |
| InP | ND | ND | ND | ND | ND | 3.94 |
| DbA | ND | ND | ND | ND | ND | 20.25 |
| BghiP | ND | ND | ND | ND | ND | 13.90 |
| Total PAHs | 31.71 | 128.25 | 39.14 | 6191.94 | 8893.37 | 4021.74 |

in SPM from the river input loads and from the atmosphere. We used the actual values of dissolved PAHs from the river, and the PAH concentrations in SPM are listed in Table 2. Input loads consisting of dissolved PAHs were calculated from the flow rates and dissolved PAH concentrations measured in August 2008. Input loads of total PAHs among the SPM were calculated from the river flow rate and the total PAH concentration in SPM (see Table 3).

Dissolved PAHs and suspended PAHs from rivers were defined in this study using field measurements. The phytoplankton standing stock used in the model was calculated from the concentration of chlorophyll–a and on the quantities measured in the summer field survey

**Table 3.** River flow rate from rivers and WWTP, and PAHs loading flux (g/day) of dissolved and SPM (Suspended Particular Matter) in water collected from river and WWTP into Masan Bay

| Stations | River flow rate $m^3$ | Dissolved PAHs | PAHs in SPM |
|---|---|---|---|
| | | Loading flux (g/day) | |
| IN1 | 22075.06 | 0.70 | 0.68 |
| IN2 | 8031.19 | 1.03 | 0.35 |
| IN3 | 607869.2 | 12.05 | 16.00 |
| Total | 337975.4 | 13.78 | 17.03 |

of Kim *et al*. (1994). We calculated the total PAH in the input load from the atmosphere using the expression 153 $\mu$g/m$^2$/year to determine sediment per year per unit area, derived by Lee *et al*. (2005) from the sediment particulate density measured in atmospheric bulk samples. The sedimentation velocity was verified using an ecosystem model described in a study of Jinhae Bay by Hong *et al*. (2007).

## RESULTS AND DISCUSSION

### Application of the model and sensitivity analysis

Figure 3 shows the concentration of PAHs calculated to examine the applicability of the model. The calculated concentrations of dissolved PAHs, PAHs in SPM and PAHs in phytoplankton stabilized within a relatively short period of time. The calculated concentration of dissolved PAHs was 38.8 ng/L and the observed concentration was 41.1 ng/L, with a relative error of −5.89%. The observed concentration of PAHs in this paper represented the average concentration at 2 sampling stations (M1, M2) in the study area (see Fig. 1). These values were the observed concentration in the surface water and bottom water at each sampling station, measured one time in August 2008. These concentration values are listed in Table 4.

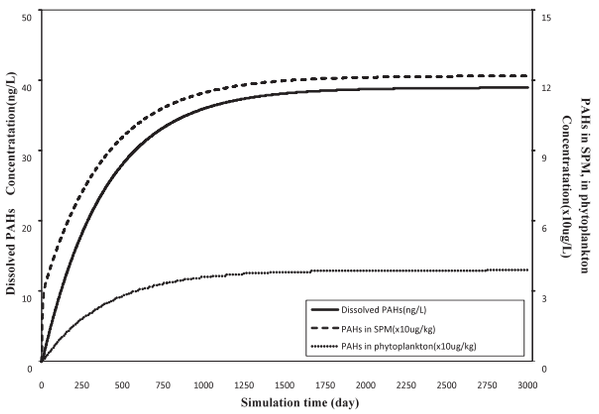


**Fig. 3.** Simulated PAH concentrations using the model.

**Table 4.** Concentration (ng/L) of individual PAH compounds in the surface and bottem seawater of Masan Bay

| PAHs | Surface seawater | | Bottom seawater | |
|---|---|---|---|---|
| | M1 | M2 | M1 | M2 |
| Nap | 13.87 | 13.60 | 32.15 | 34.82 |
| AcPy | 0.63 | 0.52 | 0.53 | 0.68 |
| AcP | 0.96 | 0.99 | 0.99 | 1.05 |
| Flu | 2.89 | 2.99 | 2.83 | 3.36 |
| PhA | 8.13 | 10.13 | 9.96 | 10.62 |
| Ant | 0.29 | 0.35 | 0.27 | 0.30 |
| FluA | 1.15 | 1.58 | 1.20 | 1.29 |
| Pyr | 1.24 | 1.93 | 1.63 | 1.59 |
| BaA | ND | ND | ND | ND |
| Chr | ND | ND | ND | ND |
| BbF | ND | ND | ND | ND |
| BkF | ND | ND | ND | ND |
| BaP | 0.05 | ND | 0.20 | 0.14 |
| InP | ND | ND | ND | ND |
| DbA | ND | ND | ND | ND |
| BghiP | ND | ND | ND | ND |
| Total PAHs | 29.21 | 32.08 | 49.76 | 53.86 |

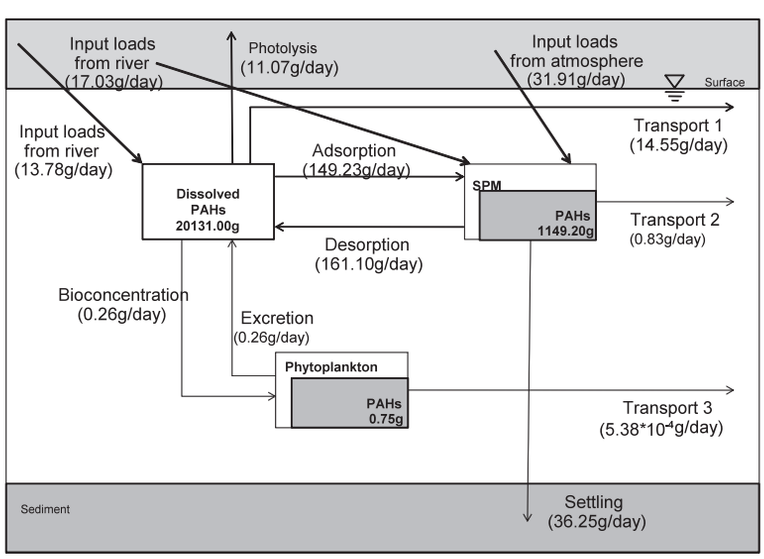


**Fig. 4.** The mass balance of PAHs in Masan Bay calculated using the model.

The sensitivity of the model, defined using the calibrated model, coefficient, was analyzed by calculating the predicted concentrations as a function of the state variable's which were varied by a factor of at most 2 or at least 1/2. Table 6 lists the results of the parameter sensitivity analysis as a function of the state variables. The results suggest that larger resulting values correspond to a greater influence of the state variables on the calculated concentration. Dissolved PAHs concentrations showed changes of more than 30% as the adsorption rate, desorption rate and settling rate were increased or decreased. The concentration of dissolved PAHs was greatly affected by the photolysis rate.

Similarly, the concentration of PAHs in SPM was also influenced by change in the adsorption rate, desorption rate and settling rate. In the case of PAHs in phytoplankton, the concentration changed by more than 10% with an increase or decrease in almost all rates, showing that this concentration is sensitive to all parameters. The sensitivity analysis indicated that greater values of the parameters, produce a the larger influence on the concentrations of the state variables. Further studies are needed to identify the important influencing factors.

**Table 5.** The parameters used in the model

| Definition | Unit | Value |
|---|---|---|
| Volume[a] | $m^3$ | 516,875,000 |
| Average depth[a] | m | 6.26 |
| Area[a] | $m^2$ | 76,120,281 |
| Cross sectional area[a] | $m^2$ | 67,500 |
| Dissolved PAHs | | |
| Input loads from river | g/day | 13.78 |
| PAHs in SPM | | |
| Input loads from river | g/day | 17.03 |
| Input loads from atmosphere[b] | g/day | 31.91 |
| Standing stock of phytoplankton[c] | $mg/m^3$ | 37.10 |
| Sinking velocity of SPM[d] | m/day | $2.14\times10^{-1}$ |
| Photolysis rate | 1/day | $5.5\times10^{-3}$ |
| Bioconcentration of phytoplankton[e] | L/kg | $1.0\times10^{3}$ |
| Uptake rate of phytoplankton[f] | L(kg day) | $3.8\times10^{2}$ |
| Excretion rate of phytoplankton | 1/day | $3.8\times10^{-1}$ |
| SPM partition coefficient[g] | L/kg | $3.17\times10^{3}$ |
| Adsorption rate | L/(kg day) | $4.1\times10^{2}$ |
| Desorption rate | 1/day | $1.4\times10^{-1}$ |

[a] Calculated based on water depth data.
[b] Lee *et al.* (2005).
[c] Kim *et al.* (1994).
[d] Hong *et al.* (2007).
[e] Law *et al.* (1997) .
[f] Calculated by multiplication of the BCF and excretion rate for phytoplankton.
[g] Calculated by multiplication of the partition coefficient and desorption rate for organic carbon.

**Mass balance**

Figure 4 shows the mass balance of PAHs in Masan Bay, calculated from the results of the model simulation. The standing stock of PAHs in Masan Bay was $2.01\times10^{4}$g in the water column, $1.15\times10^{3}$g in SPM and $7.5\times10^{-1}$g in phytoplankton.

The input loads indicated that $1.4\times10^{1}$g/day of dissolved PAHs flowed from the rivers into Masan Bay, $1.1\times10^{1}$g/day was introduced by SPM from the rivers, and $3.2\times10^{1}$g/day of SPM was introduced from the atmosphere.

The flux of dissolved PAHs was dominated by, a highest value of $1.49\times10^{2}$g/day dissolution of PAHs adsorbed onto SPM. Effluent to the open sea proceeded at a rate of $1.5\times10^{1}$g/day, photolysis proceeded at a rate of $1.1\times10^{1}$g/day, and bioconcentration in the phytoplankton proceeded at $2.6\times10^{-1}$g/day.

Flux of PAHs in the form of SPM included, desorption to form dissolved PAHs at a high rate of $1.6\times10^{2}$g/day. The next highest rate of $3.6\times10^{1}$g/day occurred via sedimentation, and the effluent to the open sea is proceeded at a rate of $8.3\times10^{-1}$g/day.

The flux of PAHs in phytoplankton was characterized by excretion to yield dissolved PAHs at a rate of $2.6\times10^{-1}$g/day PAHs, and $5.4\times10^{-4}$g/day PAHs flowed out to the open sea.

The highest rate of loss of $1.46\times10^{1}$g/day PAHs corresponded to flow to the open sea in the form of dissolved effluent. The second largest loss rate of $8.3\times10^{-1}$g/day

**Table 6.** The sensitivity analysis of PAHs concentration by change of coefficients value

| Item | The changes of PAHs (%) | | |
|---|---|---|---|
| | in water | in SPM | in phytoplankton |
| Photolysis coefficient × 0.5 | 11.471 | 8.638 | 10.667 |
| Photolysis coefficient × 2 | −17.107 | −12.882 | −17.333 |
| Bioconcentration rate × 0.5 | 0.000 | 0.000 | 50.667 |
| Bioconcentration rate × 2 | −0.001 | −0.001 | 98.667 |
| Excretion rate × 0.5 | −0.001 | −0.001 | 98.667 |
| Excretion rate × 2 | 0.000 | 0.000 | −50.667 |
| Adsorption rate × 0.5 | 34.903 | −24.492 | 34.667 |
| Adsorption rate × 2 | −34.264 | 23.738 | −34.667 |
| Desorption rate × 0.5 | −34.969 | 24.223 | −36.000 |
| Desorption rate × 2 | 40.162 | −28.075 | 38.667 |
| Sinking velocity × 0.5 | 38.914 | 42.316 | 38.667 |
| Sinking velocity × 2 | −34.422 | −37.376 | −34.667 |

PAHs corresponded to SPM adsorption, and a rate of $5.38\times10^{-4}$g/day PAHs flowed out after having been accumulated in the bodies of phytoplankton.

**Scenario analysis**

A scenario analysis was developed and implemented to bring Masan Bay to "safe" concentration level of dissolved PAHs, that is, below 30 ng/L, the limit established by US water quality guidelines. The scenario analysis consisted of three cases for reducing the input loads by 10%: (1) Reduce the input loads from the rivers by 10%, (2) reduce the input loads from the atmosphere by 10%, and (3) reduce the overall input loads by 10% (see Table 7).

The result of the scenario analysis indicated that in case I, the concentration of dissolved PAHs was 28.8 ng/L if the river input loads were reduced to 50%.

In case Ⅱ, the concentration of dissolved PAHs was 29.4 ng/L if the atmosphere input loads were reduced to 50%. In case Ⅲ, the concentration of dissolved PAHs was 27.2 ng/L if all entire input loads were reduced to 30%. In short, the concentration of dissolved PAHs in the Masan Bay satisfied the "safe" concentration (30 ng/L) if input loads from the rivers, the atmosphere, and both rivers and atmosphere were reduced by 50%, 50% or 30%, respectively (see Fig. 5).

**Table 7.** Composition of Scenario in model

| | Scenario |
|---|---|
| Case I | reduced 10% rate loads from river input |
| Case Ⅱ | reduced 10% rate loads from atmosphere input |
| Case Ⅲ | reduced 10% rate loads from entire input |

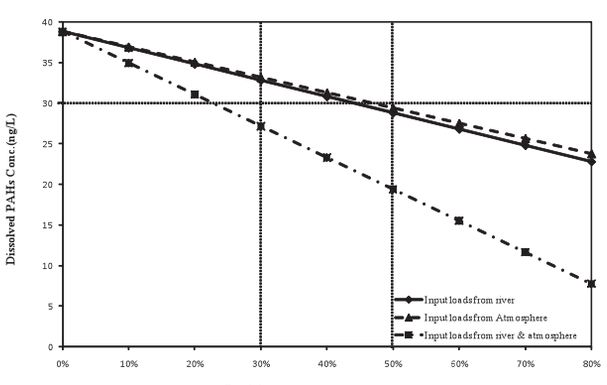


**Fig. 5.** The results of scenario analysis for each case.

## CONCLUSIONS

This study was performed to estimate the fate of PAHs in Masan Bay using a predictive computational model written In the STELLA computer language. The results of a numerical simulation of the PAHs levels in Masan Bay indicated that PAHs in the seawater were predicted with a relative error of −5.89%. The dissolved PAHs in the water layer were greatly affected by the settling velocity, the adsorption rate and the desorption rate. The adsorption rate, desorption rate and settling velocity were also the most important coefficients for PAHs in SPM. PAHs in phytoplankton were found to be sensitive to all parameters.

The standing stocks of PAHs were $2.01\times10^{4}$g in dissolved PAHs, $1.15\times10^{3}$g in SPM, and $7.5\times10^{-1}$g in phytoplankton. That is, the amount of standing stock in Masan Bay was on the order of the dissolved PAHs, PAHs in SPM and PAHs in phytoplankton. Desorption from SPM into dissolved PAHs was the transition path that with the largest loss rate. Outflow into the open sea from Masan Bay occurred in the form of dissolved effluent, which showed the highest loss rate of $1.46\times10^{1}$g/day.

In this study, we implemented a scenario developed to achieve "safe" concentration of dissolved PAHs in Masan Bay. The conditions were adjusted to achieve PAHs levels below 30 ng/L, the cut–off level established as safe by US water quality guidelines. A "safe" concentration of dissolved PAHs was achieved if the input load from the rivers of the atmosphere was reduced by 50%, or if the input loads from both the atmosphere and rivers were simultaneously reduced by 30%.

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