Summary of Risk Assessment Report
Series 7: Phthalate Ester DEHP

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Compiled by:
New Energy and Industrial Technology Development Organization (NEDO)
and
Research Center for Chemical Risk Management (CRM),
National Institute of Advanced Industrial Science and Technology (AIST)

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1. Introduction

Di(2-ethylhexyl)phthalate (DEHP) is used mainly as a plasticizer for polyvinyl chloride (PVC) resin, and the shipment of DEHP in Japan in 2001 exceeds 200,000 tons. Soft PVC products containing DEHP have extensively been used around us, including sheets/films, wire/cable, agricultural films, wallpaper, building materials, hoses/gaskets, footwear, medical devices, etc.

DEHP is a low volatile and hydrophobic substance with the vapor pressure of 3.04×10^{-5} Pa and the octanol/water partition coefficient (log *Kow*) of 7.60. DEHP, having bioconcentration factor in fish of 600-fold or somewhat more at the maximum, is not a highly accumulative substance. Although this substance is considered to be readily biodegradable at the routine inspection according to the Law Concerning the Evaluation of Chemical Substances and Regulation of their Manufacturing, etc., its estimated degradation half-life in the environment is relatively long (estimate in this study report, 1 day in the atmosphere; 15 days in surface water; 200 days in soils and 3,400 days in bottom sediments).

As shown above, soft PVC products have been widely used by the general population and DEHP is hydrophobic substance with a relatively long degradation half-life time in the environment. Therefore, DEHP has been detected in various environmental media and foodstuffs.

Various adverse effects, including an endocrine disrupting effect, of DEHP, one of the substances which were subjected to hazard assessment, have been evaluated at the Chemical Substance Council of Ministry of Economy, Trade and Industry. As a result, it has been indicated that “since its effects on reproductive/developmental toxicity has been observed in the previous findings irrespective of the presence or absence of endocrine disrupting effect, a human health risk should be evaluated based on the assessment of hazard and exposure to examine the concept of appropriate risk management.”

As for the environmental risk of DEHP it has also been considered at the initial assessment of environmental risk by Ministry of the Environment that “DEHP is a candidate to be assessed for risk in detail in freshwater environment and that it is necessary to collect information for seawater environment.”

In addition, hazard and risk assessments of DEHP have been conducted by the US National Toxicology Program (NTP) and Agency for Toxic Substances and Disease Registry

(ATSDR) of US-EPA, and in EU and Canada.

Thus hazard and risk have been assessed in Japan and other countries, the use of soft PVC containing DEHP for some applications has been restricted in Japan, and various voluntary actions have already been conducted by the industry. As for the appropriate risk management based on the risk assessment of DEHP, however, it is necessary to conduct evaluation and examination by collecting more information and analyzing the exposure in detail. Under these circumstances, detailed risk assessment of DEHP for humans and organisms in the environment was conducted as shown below.

(1) In addition to the existing reports about hazard and risk assessment, related references were investigated and analyzed comprehensively to determine the adverse effects on human health and organisms in the environment. Then the endpoint in assessing the risk for human health and the environment was selected, and the no observed adverse effect level (*NOAEL*) and the no observed effect concentration (*NOEC*) were determined.

(2) Based on the environmental monitoring data, the distributions of human intake and environmental exposure level of DEHP were estimated. By comparing these distributions with the above *NOAEL* and *NOEC*, the risks of DEHP to human health and to organisms in the environment were assessed.

(3) Since the transport of DEHP from its emission sources to humans or organisms in the environment could not be grasped quantitatively from the environmental monitoring data, the environmental emission of DEHP from the business firm and from the soft PVC products in use was estimated. The transport of DEHP from the environmental emission source to humans and organisms in the environment was estimated quantitatively with mathematical models, and the cost-effectiveness of the measures for reduction of DEHP emission was also evaluated.

2. Emission of DEHP into the Environment and Its Quantity

DEHP has been used as a plasticizer for soft PVC in a large amount. Soft PVC has wide-ranging uses and many of the products have a considerable long life of use time. Therefore, it is considered that emission into the environment occurs in various stages of a series of lifecycle, including manufacture of DEHP, manufacture of soft PVC and processing it to various products, use of products and disposal of products. Accordingly it becomes necessary to estimate the emission at various life stages.

The amount of DEHP emission from business firms into the environment at manufacture of

DEHP and manufacturing/processing of soft PVC or other products containing DEHP was obtained from the investigation data of PRTR system in fiscal year of 2001.

Since the use categories of product encompasses a wide-range and the useful life of each product is different, the emission from soft PVC products in use into the environment was estimated as the emission into the environment at the use of products based on the shipment of DEHP for each use category. In addition, the emission of DEHP into the environment for each form of disposal, such as recycling, incineration and landfill after disposal of products, was also estimated.

2.1 Emission from business firms into the environment

392,359 kg of DEHP was emitted into the environment from the business firms which are required to notify the emission by PRTR (Pollutant Release and Transfer Registers) regulations. (The firms included those which deal with the chemical substances subjected to the PRTR control and are expected to emit those substances into the environment, having more than 21 employees, operating business (businesses) belonging to any of 23 types of businesses defined in government decree and having the business firms handling with more than 5 tons/year of chemical substances subjected to the control.) 99.8% of the DEHP from those business firms was emitted into the atmosphere. Hereafter the emission of this kind is called “emission subjected to notification.”

The “emission not subjected to notification” included in the PRTR survey is a sum of the emission from the operation site which is exempted from the notification by PRTR regulations, operation sites and the emission from home. The quantity of the emission is 1,180,200 kg/year in total. Of the un-notified emission, 98.8% is the emission from the operation sites classified as lower than the bottom cut level operation sites but engaging in the businesses subjected to control, and the remainder consists of the emission from the operation sites not engaged in the businesses subjected to control and the emission from home. Since most of the un-notified emission is the emission lower than the bottom cut level from the sites operating the businesses subjected to control, the form of emission is mostly considered the emission into the atmosphere, as in the case of the operation sites to be notified.

The Emission of DEHP from business firms into the atmosphere can be classified roughly into the emission from two processes, the manufacturing process of DEHP and the manufacturing/processing of soft PVC and other DEHP-containing products. According to an interim report of the panel for risk assessment and management of phthalate esters, the emission from the manufacturing process of DEHP into the atmosphere is very small.

2.2 Emission from PVC products in use and after disposal into the environment

From the mean useful life of DEHP application (general films/sheets, agricultural PVC, leather, industrial materials, wire/cable, hose/gasket, building materials, wallpapers, footwear, coating materials/pigments/adhesives), the lifespan function of DEHP for each application was introduced. Based on this function, the secular change in the stock amount and the amount of DEHP disposed was estimated from 1952 to 2001. In addition, the emission coefficient of DEHP from PVC products in use into the atmosphere was estimated based on the thickness of PVC resin used for each application of DEHP and the ratio of use indoors and outdoors. Then by multiplying the stock amount by the coefficient, the secular change in the DEHP emission from PVC products in use into the atmosphere was obtained.

For the emission of DEHP into water system, the emission into the environment due to dissolution of DEHP from PVC products in use and leaching from the final disposal site after disposal were estimated. The emission of DEHP from PVC products used outdoors was estimated from the stock amount of DEHP and the emission coefficient. The emission from PVC products used indoors and final disposal sites was estimated by multiplying the use of water and the amount of leaching water by the monitoring concentration of DEHP.

2.3 Emission of DEHP into the atmosphere

Table 1 summarizes the emission of DEHP into the atmosphere at the manufacture of DEHP and soft PVC products, as well as the emission of DEHP at the use of products. The emission in the Kanto region is larger than that in other regions as seen from Table 1. In the Kanto region, the emission from the un-notified business firms accounts for more than a half of the whole emission.

Table 1　Emission of DEHP into the atmosphere for each region (2001) [ton/year]

Region	Emission subjected to notification	Emission not subjected to notification	Emission resulting from the products in use	Total
Hokkaido	0	15	54	69
Tohoku	16	37	54	108
Kanto	151	439	208	798
Hokuriku	0	46	19	65
Chubu	77	83	26	186
Tokai	21	189	64	274
Kinki	70	269	84	423
Chugoku	24	39	39	103
Shikoku	21	18	47	86
Kyushu	11	43	161	215
Okinawa	0	2	5	6
Total	392	1,180	762	2,334

2.4　Emission of DEHP into water system

Table 2 summarizes the emission of DEHP from the PVC products in use and the emission of DEHP from final disposal sites. All the emitted DEHP does not reach the water system for public use, and DEHP passing through the sewage-treatment plants is treated at a removal rate of 97%. As for the amount of DEHP finally reaching the water system for public use, the contribution of PVC products used outdoors is the largest, accounting for 90% or more of the whole.

Table 2　Emission of DEHP into water system [ton/year]

		Emission	Amount reaching the water system for public use
Resulting from the products in use	Used outdoor	979 - 2,284	866 - 2,067
	Used indoor	165	53
Industrial waste disposal plant		0.4	0.4

3. Estimation of Environmental Fate

The environmental fate of DEHP emitted into the atmosphere and water system from business firms and the soft PVC products in use cannot be clarified from the existing surveillance of environmental monitoring. Therefore, the fate of DEHP in the general environment was estimated using the compartment models such as atmosphere, soil, surface water and plants.

3.1 Fate of DEHP emitted into the atmosphere

The fate of DEHP in the atmosphere was estimated using the general environmental condition in the Kanto region and the following points were clarified.

(1) 60 to 70% of DEHP emitted into the atmosphere was adsorbed to the airborne particles. In the atmospheric environment on local authority basis, advection largely contributes to its disappearance. A part of DEHP in the atmosphere transfers on the soil by deposition, and it was estimated that about 80% of the whole deposition was attributable to wet deposition of the adsorbed state of airborne particles.

(2) Almost the whole amount of DEHP deposited to soils is adsorbed to soil particles and disappears mainly by degradation, and a part of the remainder is transported to water environment due to soil erosion. Contribution of leaching, runoff, resuspension and volatilization was estimated to be low.

(3) Most of DEHP in the aerial part (leaves, stems and fruits) of plants result from deposition and absorption from the atmosphere, and contribution of uptake from the root is small. Therefore, it was estimated that DEHP in the soil hardly contributed to the concentration of DEHP in the aerial part of plants.

(4) Almost whole amount of DEHP transferred into livestock comes through feeds (plants), and it was estimated that contribution of direct intake from the atmosphere and soil was low.

(5) DEHP emitted into the atmosphere was considered to transfer to the aerial part of plants by absorption and deposition. It was also considered that a part of DEHP transfers into livestock through feed crop and is finally taken by humans through agricultural crops and livestock products.

3.2 Fate of DEHP emitted into water system

The fate of DEHP was estimated assuming a virtual river and the following points were clarified.

(1) As for the DEHP loaded into the riveve

phase.

(2) In the aqueous phase, DEHP is transported to the outside of the system mainly by advection. A part of the DEHP disappears from the aqueous phase by degradation and in association with sedimentation of suspended particles to the bottom sediment. It was estimated that the contribution of volatilization and diffusion to the bottom sediment phase is low.

(3) From the bottom sediment phase, DEHP mainly disappears by degradation and resuspension, and it was estimated that the contribution of diffusion into the aqueous phase is low.

(4) DEHP advected from rivers to the nearshore waters is diluted, mixed and bioconcentrated in fish and shellfish. The bioconcentration factor is considered about 600 L/kg.

4. Risk of Human Health

4.1 DEHP intake

Using the concentrations of DEHP in indoor air and outdoor air measured by the Tokyo Metropolitan Government in 2000 and the dietary DEHP concentration measured by Japan Food Research Laboratories in 1998 and 2001, the DEHP intake was estimated by Monte Carlo simulation for age groups over 1 year old. Table 3 shows the intake by male general population estimated using the dietary DEHP concentration in 1998.

Table 3 Estimated DEHP intake for age groups (males)

Age group (Year old)	DEHP intake (µg/kg/day)			
	Mean	5 percentile	50 percentile	95 percentile
Whole	6.7	0.86	4.1	21.3
1	21.7	2.6	13.0	68.2
5	13.6	1.7	8.2	42.2
10	10.0	1.3	6.2	30.5
13 - 15	7.1	1.0	4.5	21.6
16 - 19	5.9	0.81	3.7	18.0
20 - 29	5.3	0.75	3.4	16.6
30 - 39	5.6	0.78	3.5	17.2
40 - 49	5.6	0.82	3.5	17.3
50 - 59	6.2	0.92	4.0	18.6
60 - 69	6.1	0.86	3.8	17.8

As shown in Table 3, the DEHP intake in childhood is considerably higher than that in adulthood. In addition, intake through foods largely contributes to the intake, but inhalation of indoor and outdoor air hardly contributes. A part of this DEHP intake was considered to be attributable to the transfer from PVC gloves to food at the time when the measures for suppressing emission by operation sites were ongoing.

The mean DEHP intake in one year-old children estimated using the dietary concentration in 2001 was 6.1 μg/kg/day (range of 5 to 95 percentiles: 1.1 to 17.5 μg/kg/day) for males and 5.7 μg/kg/day (range of 5 to 95 percentiles: 0.8 to 15.9 μg/kg/day) for females. The intake through food largely contributes to the intake, but inhalation of indoor and outdoor air hardly contributes. In addition, the mean DEHP intake in all age groups was 1.9 μg/kg/day (range of 5 to 95 percentiles: 0.4 to 5.4 μg/kg/day) for males and 1.8 μg/kg/day (range of 5 to 95 percentiles: 0.4 to 5.0 μg/kg/day) for females.

Based on the monitoring data, the DEHP intake in the infants aged less than 1 year through breast milk, artificial milk and baby foods was also estimated. Since infants use concomitantly milks (breast milk and artificial milk) and baby foods, the total intake was also estimated. Since it was estimated that the DEHP concentration in artificial milk was higher than that in the breast milk, artificial milks were assumed as milks. Table 4 shows the results in baby boys.

Table 4 Estimated DEHP intake through milks and baby foods (baby boys)

Age of infants	DEHP intake (μg/kg/day)			
	Mean	5 percentile	50 percentile	95 percentile
At birth	13	0.96	6.4	44
30 days	9.0	0.67	4.5	31
1 to less than 2 months	7.8	0.58	3.9	27
2 to less than 3 months	6.4	0.47	3.2	22
3 to less than 4 months	8.3	1.4	5.5	23
4 to less than 5 months	7.6	1.3	5.0	22
5 to less than 6 months	7.2	1.4	5.0	20
11 to less than 12 months	11	2.0	7.5	30

4.2 Estimation of the main exposure route to general population in the Keihin area

The DEHP intake through agricultural crops and livestock products was estimated in the Keihin area, the largest consuming region, considering the spatial distribution of the concentration of DEHP in the atmosphere calculated with an atmospheric model AIST-ADMER and data on the production and shipment of agricultural crops and livestock products. In addition, the intake through aquatic products was estimated using the monitoring data of the DEHP concentration in sea, rivers and lakes waters and bioconcentration factors. In order to evaluate the effects of regional variation of concentration, production and shipment on the results of estimation, Monte Carlo simulation was conducted. As a result, the mean DEHP intake through domestic agricultural crops in males in Metropolitan of Tokyo was estimated as 0.49 μg/kg/day (range of 5 to 95 percentiles: 0.064 to 1.5 μg/kg/day), and that through domestic livestock products (dairy products, beef, pork and poultry) was estimated as 1.0 μg/kg/day (range of 5 to 95 percentiles: 0.05 to 3.5 μg/kg/day). The mean DEHP intake through aquatic products was estimated to be 0.14 μg/kg/day (range of 5 to 95 percentiles: 7.5×10^{-4} to 0.39 μg/kg/day).

From these results, it was estimated that a part of DEHP emitted into the atmosphere transfers into agricultural crops and livestock. It was also estimated that the general population in the Keihin area ingests DEHP mainly through domestic livestock products collected nationwide and ingests DEHP from domestic agricultural crops shipped to the Keihin area and imported livestock products (Fig. 1). For each source of emission, contribution of emission from the business firms not subjected to notification in the PRTR system into the atmosphere was estimated to be great.

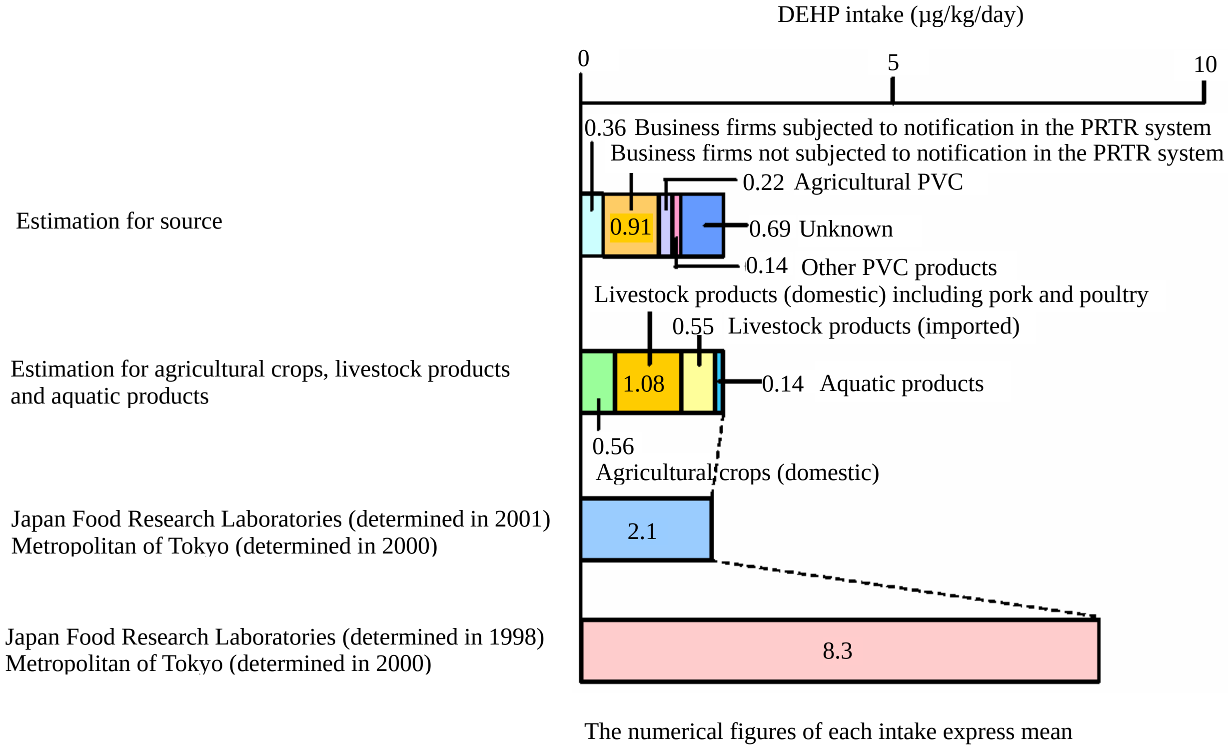

Fig. 1 Summary of estimated DEHP intake in the general population in the Keihin area

4.3 Hazard assessment and dose-response evaluation

DEHP and its main metabolites (mono(2-ethylhexyl) phthalate and 2-ethylhexanol) show no genotoxicity in most studies. Also, since hepatocellular carcinoma seen in rats and mice seemed to be specific to rodents considering from its mechanism of action, carcinogenic potential of DEHP in humans is considered to be low. Therefore, carcinogenesis was not employed as an endpoint of health risk in humans.

Testicular toxicity and reproductive toxicity were employed as the noncarcinogenic adverse effect. In marmosets, a primate, since testicular toxicity is not observed at high doses, it is slightly questionable to employ it as an endpoint in humans. But considering that testicular toxicity has been employed at the establishment of the temporary tolerable daily intake (*TDI*) by Ministry of Health, Labour and Welfare and employed as an endpoint in the initial environmental risk assessment by the Ministry of the Environment, NTP assessment document, the provisional version of EU assessment document and ATSDR assessment, it was decided to employ testicular toxicity as a tentative endpoint at the present point of time in this assessment document. From the above-mentioned, the *NOAEL* (3.7 mg/kg/day) in the study of Poon et al. reporting the lowest *NOAEL* for testicular toxicity was used for risk assessment.

In the developmental/reproductive toxicity study, adverse effects of DEHP have been observed. In the provisional version of EU assessment document, the results of developmental toxicity study by Arcadi et al. were employed. However, since there is uncertainty in doses, it was decided not to employ them in this assessment, and the *NOAEL* (14 mg/kg/day) for reproductive effects observed in the study of Lamb et al, was used for risk assessment.

As a minimum margin (*Margin*) required to decide to have no concern for the risk of testicular toxicity, a product 30 of 3 explaining the interspecies difference in the sensitivity between rats and humans and 10 explaining individual difference was considered to be valid. The interspecies difference in sensitivity, 3, is a product of the interspecies difference in toxicokinetics (1 was employed as the value on the safe side) and the interspecies difference in toxicodynamics (3 was employed by rounding the default value (2.5)). As the individual difference in sensitivity, 10, generally used as a default value, was employed.

As the minimum margin for evaluating the risk for reproductive toxicity, a product 100 of 10 explaining the interspecies difference between mice and humans and 10 explaining individual difference was considered to be valid.

For the interspecies difference in sensitivity, the transfer of DEHP and its metabolites into fetuses is unknown and reproductive toxicity is not specific to rodents. Accordingly, 10 generally used as a default value (a product of 4 explaining toxicokinetics and 2.5 explaining toxicodynamics) was employed, and for the individual difference in sensitivity, 10 generally used as a default value was employed.

4.4 Risk evaluation

As shown in Fig. 2, the risk (*Risk*) for testicular toxicity and reproductive toxicity was calculated as the probability that the intake in humans (*Intake*) can exceed the value obtained by dividing the *NOAEL* in experimental animals by the minimum margin (*Margin*) at risk evaluation considering individual difference and interspecies difference ($Prob(Intake \geqq NOAEL/Margin)$). However, this probability does not show the increment of the incidence of adverse effects. It is expected that the increment of the incidence of adverse effects is very small in comparison with this excessive probability.

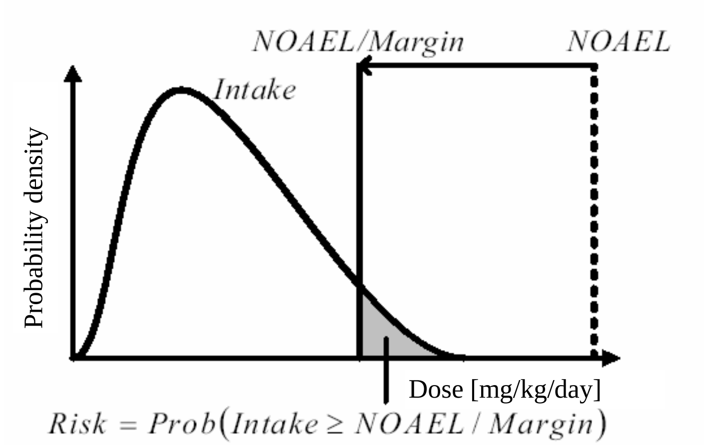

Fig. 2 Definition of the index of human health risk

Testicular toxicity

Table 5 shows the results of calculation of the risk ($Risk_{testis}$) for testicular toxicity induced by DEHP ingested by inhalation of air and through meals based on the concentrations of DEHP in indoor and outdoor air measured by the Tokyo Metropolitan Government in 2000 and the dietary DEHP concentration measured by Japan Food Research Laboratories in 1998. In one-year old baby of the highest intake, the $Risk_{testis}$ was less than 1%, and it is considered that the *Margin* of 30 has been almost assured between $NOAEL_{testis}$ and the intake.

Table 5 Results of calculation of the risk for testicular toxicity

Age group (Years old)	$Risk_{testis}$[%]	Age group (Years old)	$Risk_{testis}$[%]
1	0.98	13-15	0.03
2	0.63	16-19	<0.01
3	0.44	20's	<0.01
4	0.31	30's	<0.01
5	0.26	40's	<0.01
6	0.15	50's	0.01
10	0.07	60's	<0.01

It is controversial whether it is proper or not to use the estimated short-term intake in the babies aged less than 1 year. Nevertheless, the risk for testicular toxicity in baby boys aged less than 1 year was calculated as the probability that the DEHP intake at concomitant use of milks and baby foods may exceed the value obtained by dividing $NOAEL_{testis}$ by $Margin_{testis}$ Table 6 shows the calculation results. It is considered that the risk for testicular toxicity in infants is not at a level of concern.

Table 6 Calculation results of the risk for testicular toxicity in baby boys resulting from powdered milk and baby foods

Age	$Risk_{testis}$[%]	Age	$Risk_{testis}$[%]
At birth	0.51	3 to less than 4 months	0.08
30 days	0.23	4 to less than 5 months	0.06
1 to less than 2 months	0.14	5 to less than 6 months	0.02
2 to less than 3 months	0.08	11 to less than 12 months	0.09

As shown above, it is considered that the risk for testicular toxicity is not at the level of concern in any age group aged above 1 year and infants aged less than 1 year. The intake based on the survey in 2001 by Japan Food Research Laboratories is about 1/3 of that in 1998, and it is considered that the risk in any age group aged above 1 year is also not at a level of concern.

Reproductive toxicity

The subjects to be exposed were males and females aged 16 to less than 60 years. Table 7 shows the results of calculation based on the concentration of DEHP in indoor and outdoor air measured by the Tokyo Metropolitan Government in 2000 and the dietary DEHP concentrations determined by Japan Food Research Laboratories in 1998. In males and females in all age groups, the calculated risk for reproductive toxicity ($Risk_{repro}$) is below 0.01%, and it is considered that the *Margin* of 100 is assured between $NOAEL_{repro}$ and the intake.

Table 7 Results of calculation of the risk for reproductive toxicity

Age group (year old)	$Risk_{testis}$[%]	
	Males	Females
16 - 19	0.01	<0.01
20’s	<0.01	<0.01
30’s	<0.01	<0.01
40’s	<0.01	<0.01
50’s	0.01	<0.01

4.5 Cost-effectiveness of the emission-reduction measure

The cost of exhaust gas treatment measures and the effects of those measures on atmospheric emission reduction were estimated for the business firms subjected to notification and those not subjected to notification in the PRTR system, which were estimated to contribute greatly to the DEHP intake.

When the emission-reduction measures are taken for the business firms dealing with DEHP, it is necessary to use the exhaust gas treatment facility of the collection method different from that of volatile organic chemicals. Because DEHP exists mainly as fume and mist in the air in those business firms.

For the business firms subjected to notification reporting to emit DEHP of over 1 ton a year into the atmosphere in the survey of PRTR system in fiscal year 2001, it was assumed as follows:
HEAF (roll glass felt mode) is introduced into the business firms emitting over 1 ton to less than 10 tons of DEHP into the atmosphere in a year, and
A pipe filter facility is introduced into the business firms emitting over 10 tons of DEHP in a year.
30 HEAFs and 15 pipe filter facilities are required, respectively, and the cost required to reduce 1 ton of the emission into the atmosphere was estimated as 2.14 million yen (the collection rate at usual operation was assumed as 90%). In association with this emission reduction, it was estimated that the DEHP intake in the general population in the Keihin area might be reduced slightly (0.2 to 0.4 µg/kg/day).

If HEAF is introduced as a treatment facility into 500 business firms of plastic manufacturers accounting for 3/4 of the business firms not subjected to notification, the cost requiring reduction of 1 ton of emission per business firm was estimated as 2.98 million yen, and the DEHP intake in the general population in the Keihin area was estimated to reduce by 0.7 to 0.9 µg/kg/day. However, many of the business firms not subjected to notification have small scale of operation, and there is a possibility that introduction of facilities as a voluntary reduction measure may become a large burden to business firms.

4.6 Summary of human health risk assessment

In this assessment document, the risk of DEHP for human health in Japan was assessed based on the available data reported previously and current scientific findings. As shown in each case, an assumption was given to complement insufficient or deficient data in estimating the intake from the monitoring data and in estimating the main exposure route of DEHP from the emission source to humans by mathematical modeling. The validity of these assumptions will be verified in the future surveys and investigations. The items awaiting future surveys and investigations are listed below.

(1) Monitoring survey for estimation of intake and exposure route

· Survey of the dietary DEHP concentration at a frequency at which the annual average intake can be estimated

· Survey of the concentrations of DEHP in indoor air, outdoor air and individual food group at a frequency at which the validity of modeling can be evaluated

(2) Study on reproductive toxicity

· Study on the difference in the mechanism of onset of reproductive toxicity between rodents and primates

(3) Study on the environmental emission source and the emission

· Study on the elaboration of the lifetime function and the emission coefficient for each soft PVC product

5. Environmental Risk

Screening-level environmental risk assessments of DEHP for aquatic organisms in Japan were conducted using estimated statistical values based on surface water and sediment monitoring data and effect threshold values based on a large aquatic toxicity database. Individual level effects such as survival, reproduction, growth and development on aquatic organisms were selected as the endpoint for this assessment. Margin of exposure (*MOE*), which is defined as the ratio of the exposure estimate divided by the effect threshold, was used to characterize the aquatic risks of DEHP for the exposure through water and bottom sediment. Then the necessity of risk management/measures was evaluated taking the uncertainty into consideration. Fig. 3 shows the flow of the environmental risk assessment in this evaluation document.

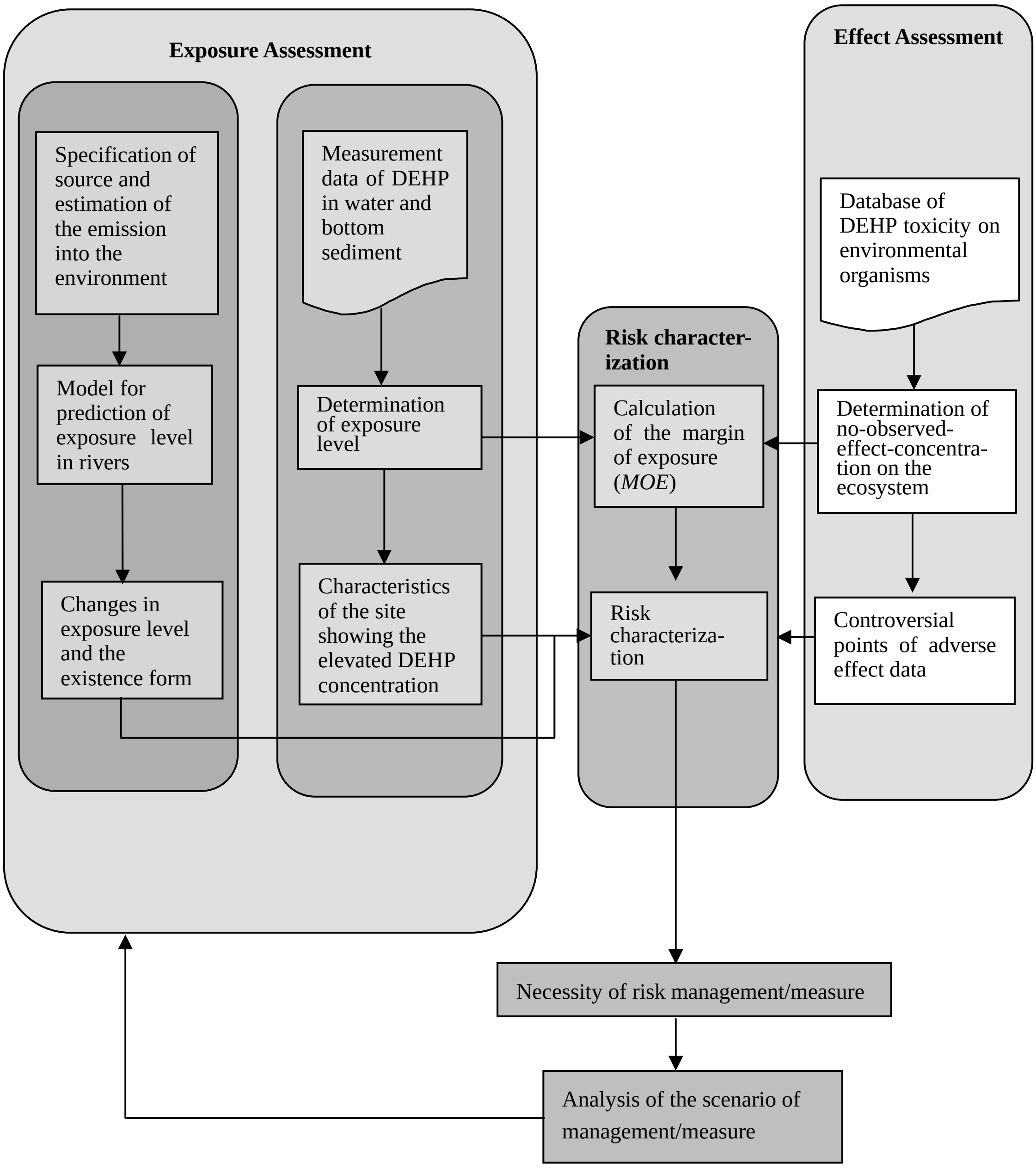

Fig. 3 Flow of environmental risk assessment of DEHP

5.1 Exposure level and the point showing high concentrations

Tables 8 and 9 show the mean concentration and 95 percentile for each water system (rivers, lakes and sea areas) and for each fiscal year determined by conducting statistical analysis of the monitoring data of DEHP in water and bottom sediments provided by Ministry of the Environment, Ministry of Land, Infrastructure and Transportation and Local Government Units. In the statistical analysis of monitoring data, the reliability of each data was not evaluated. Instead we took a stance to handle all the available data in the same manner. Risk was evaluated based on the value of 95 percentile covering the majority of surface waters for public use on the basis of the evaluation of the exposure in general environment.

As for the sites of the elevated DEHP concentrations, the characteristics and source of the point of determination were discussed. As a result, the sites where DEHP is detected at the elevated concentrations were commonly the one into which untreated waste water resulting from human activities is considered to flow, and there were many sites where BOD levels were high.

In the analysis of the site-specific exposure concentrations, Tamagawa River was taken as an example. The main source of DEHP loading to Tamagawa River was specified, and the emission loads from the sources were estimated. As a result, it was shown that the contribution of DEHP eluted from the products for outdoor use by contacting with rainwater was the largest, which accounted for about 78% of the whole emission load to Tamagawa River. Taking the results of the emission estimation as the input data, the DEHP concentration in water of Tamagawa River was predicted using an aquatic system model called AIST-SHANEL. As a result, the sites and season in which the DEHP concentration becomes to be relatively high in Tamagawa River were confirmed visually, though it is hard to quantitatively discuss the prediction accuracy of the model. In addition, this modeling results showed the usefulness of an aquatic model such as AIST-SHANEL to analyse exposure concentrations of contaminants and to support communicating the assessment results among stakeholders.

Table 8 Results of estimation of the DEHP concentration in water of various water systems

Water system	Year of determination	Number of sample	Geometric mean [μg/L]	Geometric standard deviation	95 percentile [μg/L]
Rivers	1998	1,742	0.17	4.7	2.12
	1999	2,025	0.13	4.5	1.55
	2000	1,472	0.09	7.4	2.55
	2001	1,594	0.08	7.9	2.31
	2002	1,476	0.08	7.9	2.28
Lakes	1998	141	0.13	5.6	2.22
	1999	116	0.04	5.6	0.66
	2000	57	0.15	2.5	0.68
	2001	79	0.05	6.2	1.07
	2002	83	0.02	10.1	1.09
Sea area	1998	209	0.20	4.2	2.11
	1999	235	0.09	4.4	1.03
	2000	229	0.04	8.6	1.55
	2001	213	0.03	8.2	0.80
	2002	237	0.01	12.6	0.52

Table 9 Results of estimation of the DEHP concentration in the bottom sediment of various water systems

Water system	Year of determination	Number of sample	Geometric mean [μg/kg-dry]	Geometric standard deviation	95 percentile [μg/kg-dry]
Rivers	1998	197	184	8.9	6,660
	1999	173	331	7.3	8,730
	2000	95	259	7.8	7,660
	2001	175	177	11.4	9,720
	2002	115	42	18.5	5,060
Lakes	1998	10	542	6.6	12,000
	1999	11	259	4.8	3,420
	2000	28	109	3.5	840
	2001	35	159	2.7	790
	2002	11	94	7.6	2,650
Sea area	1998	29	151	4.6	1,510
	1999	31	135	6.4	2,860
	2000	29	225	4.1	2,250
	2001	43	89	5.4	1,400
	2002	38	78	5.1	1,130

5.2 Adverse effects on organisms in the environment

The adverse effects of DEHP on organisms in the environment were investigated and examined comprehensively to determine the *NOEC* used for risk characterization. The result of the effect assessment is summarized in Table 10.

Since DEHP has very low water solubility and easily forms colloidal dispersions in water, it causes problems in preparation of test water, maintenance of exposure concentration and interpretation of test results. Ecological effects of DEHP have been examined in many studies, but there are very few studies in which clear concentration-effect relationship was obtained. The effect concentration or *NOEC* in many studies are expressed as “above the highest test concentration,” and there are very few studies presenting the specific value of effect concentration.

As for the exposure via water, *MOE* was calculated from the data for aquatic invertebrates ($NOEC_{invert}$ = 77 µg/L) by Rhodes et al. reporting the lowest *NOEC* among the studies on ecological effects conducted by relatively reliable methods as the $NOEC_{water}$. The result of this study is considered not the intrinsic toxicity but the physical effects captured in the films formed on the surface of test water. At this time, however, it was decided to employ this data in risk assessment, because it is difficult to clearly differentiate the physical effects from the intrinsic toxicity and the physical effects is a type of adverse effects on aquatic organisms attributable to the characteristics of DEHP.

As for the exposure via sediment, very limited toxicity data are available at this time. Since DEHP is easily adsorbed to particles and to accumulate in the bottom sediment, and some benthic organisms directly ingesting bottom sediment, the exposure through bottom sediment is important for such species. *NOECs* from sediment toxicity studies by Call et al.for aquatic invertebrates and by Solyom et al. for amphibian were selected for use in risk characterization. Both tests are judged to be relatively reliable for use in risk assessment. The lower *NOEC* was the data showing no effect in amphibia at levels above 1,000 mg/kg-dry reported by Solyom et al. In this evaluation document, a tentative value of NOECsed = 1,000 mg/kg-dry was set to calculate *MOE* for descriptive purposes..

Table 10 Summary of adverse effects of DEHP on organisms in the environment

Organism group	Exposure route	*NOEC* used for risk assessment	Findings/remarks
Fish	Water	No adverse effect at the level attainable in the water environment	· There is no highly reliable data showing some effect in the range of colloidal dispersions. · In many studies using solubilizing agents, no effect was observed at the concentrations two orders of magnitude greater than the water solubility. Those levels are also considered as the value difficult to attain in the actual environment.
	Feed	Not subjected to evaluation	
Invertebrates	Water	$NOEC_{invert}$: 77 µg/L (Rhodes et al., 1995)	· Data obtained by the studies in 1970's, showing the effects at the concentration near the solubility in water or lower, is less reliable and has been rejected in the review in many authorities. · In the studies using the solubilizing agents appropriately, no effect has been observed at the concentrations two orders of magnitude higher than the water solubility. · The effects observed in the range of concentrations at which DEHP exists in a condition of stable colloid dispersion are not considered to be caused by the intrinsic toxicity. They are possibly considered the physical effects captured in the formed surface films on test water or the undissolved substance.
	Bottom sediment	$NOEC_{sed.invert}$: 3,000 mg/kg/dry (Call et al., 2001)	· The toxicity study of the exposure through bottom sediment is still developing, and there is no established method. · The results of toxicity of bottom sediment vary, and the interpretation is very difficult. · Bottom sediment is the final sink of DEHP in the water environment and detected frequently in the environment. · Benthic organisms are easily exposed to DEHP existing in the bottom sediment.
Algae	Water	No adverse effect at the level attainable in the water environment	· There is no highly reliable data showing effects at the concentrations lower than the water solubility. · In many studies using solubilizing agents, no effect was observed at the concentrations two orders of magnitude higher than the water solubility. The level is considered as the value difficult to assume in the actual environment.
Amphibia	Bottom sediment	$NOEC_{sed.amphib}$: 1,000 mg/kg-dry (Solyom et al., 2001)	· Since the method and conditions of study have not been established, it is difficult to interpret the results. · In the recent toxicity study on incubation of frog eggs, no effect bas been observed even at doses above 1,000 mg/kg-dry.
Terrestrial organisms	-	Not subjected to evaluation	· There is no highly reliable data examined on the effects on terrestrial organisms (including birds). · There is no report showing effects at the level that can attain in the environment.

5.3 Characterization of environmental risk

The risk for aquatic organisms was evaluated by determining the value obtained by dividing the *NOEC* value by the environmental concentration, i.e., the margin of exposure (*MOE*) and by taking the uncertainty into consideration.

As for the criteria of *MOE* in evaluating the environmental risk, a value of 10, i.e., uncertainty associated with extrapolation from laboratory to outdoors for both water and bottom sediment, was considered to be valid taking the previous findings and the weight of evidences about the adverse effects of DEHP into consideration.

The results of calculation of *MOE* in water are shown in Table 11. Here, the geometrical mean and the value of 95 percentile derived by statistical analysis of the monitoring data are shown. In addition, the *MOE* for the maximum value of measured data is shown as a reference value. As a result, more than 99% of the estimated environmental concentration in the general water system showed the *MOE* greater than 10.

Since DEHP easily adsorbs to particles in water and the bottom sediment, the proportion of DEHP existing as a dissolved state considered contributory to toxicity (bioavailable fraction) is expected to be lower than the actual reported value. Therefore, when the *MOE* is determined using the bioavailable fraction of DEHP concentration as the exposure concentration, the *MOE* value could be higher.

In addition, the presence of dissolved organic substances and surface-active agents in the actual environment can promote the solubility of DEHP in the environmental water and can increase the proportion of DEHP existing in the dissolved state. This phenomenon reduces the possibility of onset of physical effects of colloid particles observed in laboratories on aquatic organisms.

The effects of coexisting substances playing a role of solubilizing agent that exists in the natural environment on the toxicity of DEHP are unknown. However, no effect has been observed at the highest test concentration in many previous toxicity studies using solvents or dispersing agents, and the level is about two orders of magnitude higher than the maximum detection level detected in the actual water system. From the above mentioned, there is a extremely low possibility that DEHP may pose adverse effects on aquatic organisms at the present level of detection, even if DEHP in the actual environment exists in the dissolved state. Therefore, the possibility that aquatic organisms may suffer adverse effects at the present contamination level of DEHP in the water quality of the general water system in Japan is considered to be very low, and the risk is considered not to be at the level of concern.

Table 11 Result of calculation of the *MOE* in water

	Fiscal year 1998			Fiscal year 1999			Fiscal year 2000			Fiscal year 2001			Fiscal year 2002		
	Rivers	Lakes	Sea water	Rivers	Lakes	Sea water	Rivers	Lakes	Sea water	Rivers	Lakes	Sea water	Rivers	Lakes	Sea water
GM[1)]	456	591	380	602	2,081	416	653	461	1,400	856	2,026	2,655	700	3,667	2,655
95%[2)]	36	35	37	51	109	57	28	99	44	30	67	82	31	61	82
MAX[3)]	4.1	18.8	7.7	1.3	32	18	1.8	77	5.5	3.7	11	8.6	1.8	15	7.7

1) Geometric mean, 2) 95 percentile, 3) Maximum value (Measured value)

The results of calculation of *MOE* in bottom sediment are shown in Table 12. The values of *MOE* in the benthic organisms through bottom sediment are above 10 at all but one points in the general water system. From these findings, it is considered to be little concern that the benthic organisms may suffer from adverse effects at the present contamination level of DEHP in the bottom sediment found in the general water system in Japan, and the risk is considered not to be at the level of concern.

Table 12 Results of calculation of *MOE* in bottom sediment

	Fiscal year 1998			Fiscal year 1999			Fiscal year 2000			Fiscal year 2001			Fiscal year 2002		
	Rivers	Lakes	Sea water	Rivers	Lakes	Sea water	Rivers	Lakes	Sea water	Rivers	Lakes	Sea water	Rivers	Lakes	Sea water
GM[1)]	5,438	1,846	6,628	3,020	3,861	7,832	3,867	9,179	4,442	5,659	6,294	11,266	23,906	10,622	12,761
95%[2)]	150	83	662	115	292	350	131	1,190	444	103	1,266	714	198	377	885
MAX[3)]	4.8	250	278	43	208	152	77	909	400	23	526	588	36	345	417

1) Geometric mean, 2) 95 percentile, 3) Maximum value (Measured value)

Judging from the results of risk assessment described above and the DEHP contamination level observed in the general water system in Japan at present, it is considered unnecessary to take immediate measure for risk management of environmental effects. This is a conclusion derived after examining sufficiently the existing available data. In this evaluation, however, the conditions assumed from the standpoint of safety for the descriptive purposes due to lacking data and uncertainty are also included. Therefore, it is necessary to investigate and examine the following items further for verification of such the assumption and for more reliable environmental risk assessment:

· Sophistication of the method of estimation of the emission from the DEHP-containing products used outdoors into the water system

· Elucidation of the mechanism of occurrence of the effects on aquatic organisms in the colloidal dispersion system

· Development of the reliable study on ecological effects on the benthic organisms

· Accumulation of the data for adverse effects of the degradation products of DEHP on organisms in the environment

· Regular monitoring at the point where DEHP was detected at high concentrations and an investigation to elucidate its sources