

Regarding the Result of 1998
Air Environmental Surveillance
on Exogenous Endocrine
Disrupting Chemicals

Air Pollution Control Division, Air Quality
Bureau, Environment Agency

1. Summary of surveillance

(1) Objectives of Surveillance

Exogenous endocrine disrupting chemicals (hormone disrupting chemicals) are chemical substances that disrupt the hormonal functions of living organisms, and thus are pointed out as being likely to impair the reproductive functions and cause malignant tumors. Although many questions remain unsolved scientifically, there is a growing national concern over the likelihood that when taken into the body even in an extremely small amount, they affect the body and continue to have effects over future generations.

Concerning this problem, the Environment Agency set forth its basic position, specific future approaches and scientific knowledge in the SPEED '98 (Strategic Programs on Environmental Endocrine Disruptors) in May 1998. In accordance with these programs, a surveillance was started to assess the nationwide pollution status with particular emphasis on 67 substances suspected of having an endocrine disrupting effect.

This surveillance was conducted nationwide with regard to the air environment after the standardization of sampling method and sample analyzing method for benzo(a)pyrene and phthalate diesters that are highly likely to be detected in the ambient air. The surveillance was also conducted for future reference on substances that were not listed in the SPEED '98 but were measurable using the same measuring method.

(2) Summary of surveillance

Benzo(a)pyrene and phthalate diesters were measured in accordance with "Manual for Hazardous Air Pollutants Measuring Method" and "Report on 1995 Chemical Substances Analyzing Method Development Research" respectively.

As for phthalate diesters, since products using them are found in various places, an experiment was conducted in advance to review measures to reduce the blank value, and the surveillance was carried out under strict supervision in all the handling stages including the collection, transportation and analysis of samples, in order to reduce their contamination as much as possible.

In this surveillance, individual local public bodies were placed in charge of collecting samples, and all the samples collected were analyzed by the Japan Environmental Sanitation Center.

2.1 Sampling sites

Samples were collected for the surveillance of general environmental conditions at nationwide 178 sites in three districts (industrial, residential and suburban districts) of 59 prefectures and cabinet-order designated cities, and for the surveillance of environmental conditions in roadside areas and their hinterland at a total of 20 sites in 10 districts.

2.2 Sampling time

For benzo(a)pyrene and phthalate diesters, samples were collected during the period from October to December 1998.

Since there was a need to remove as many factors leading to contamination as possible during sampling for the measurement of phthalate diesters, rainy days were avoided for collecting samples. When it was raining at the start of sampling, the sampling was suspended, but when it started to rain during sampling, the sampling was continued.

2.3 Sampling methods

Benzo(a)pyrene, etc.

High volume air samplers or low volume air samplers were used to collect dust in the air on filters for consecutive 24 hours. Filters used were made of quartz fiber or glass fiber.

Phthalate diesters

Clean glass-fiber filters and activated-carbon filters washed with acetone were attached to special filter holders, transported in sealed condition to sampling sites, and used to collect samples at a flow rate of 7-8 per minute for consecutive 24 hours. The greatest possible care was used to collect samples to prevent contamination from surroundings.

2.4 Sample analyzing methods

Benzo(a)pyrene, etc.

The filters were regulated to a constant weight at 20 and 50% relative humidity and measured before and after sampling. At this stage dust concentration was obtained from the weight increase measured.

The filters were cut off into the adequate number of pieces; they were subjected to ultrasonic extraction with dichloromethane, and the extracted solution was dissolved in acetonitrile, and analyzed by a high performance liquid chromatograph (HPLC).

Phthalate diesters

The samples were prepared for analysis as promptly as possible after they were collected, and were analyzed under standardized conditions as much as possible. The filters removed from their holders were subjected to ultrasonic extraction with dichloromethane; required quantities were dispersed, condensed and then analyzed by a gas chromatograph mass spectrometer (GC/MS).

2.5 Accuracy control

Benzo(a)pyrene, etc.

Filters were obtained and analyzed from the same lot from which local public bodies used filters for sampling as disguised blank samples. A travel blank test was conducted at 18 sites that accounted for approximately 10% of total samples collected, and the value obtained were compared with the disguised blank value to see if there is any contamination during transportation.

A duplicate measurement was also made at 18 sites that accounted for approximately 10% of total samples collected (at the same sites where the travel blank test was conducted). The objective of this duplicate measurement was to evaluate the accuracy of all work processes by collecting and analyzing two samples under the same conditions. The evaluation criteria was set at 30% variation rate or less.

As for the sensitivity variation of the equipment used, the reference solution having a concentration level that lies in the middle of the measurement lines was measured once for approximately every ten samples to make sure that the sensitivity variation of the substance to be measured stayed within $\pm 20\%$ as compared with the sensitivity measured at the time when the measurement lines were drawn.

If the travel blank value was large, and the contamination of the sample was suspected, or if the dual measurement produced significantly different results, the reliability of measurement value was considered questionable; thus, the sample was remeasured in accordance with the procedure for dealing with abnormal values specified in "Manual for Hazardous Air Pollutants Measuring Method."

Phthalate diesters

The filter washed with acetone and kept in sealed condition for a disguised blank test was prepared and analyzed as in the case of the sample. As in the case of benzo(a)pyrene, a travel blank test and a dual measurement were conducted at 18 sites that accounted for approximately 10% of all samples taken. As for the sensitivity variation of the equipment used, the reference solution having a concentration level that lies in the middle of the measurement lines was measured before and after the sample was measured to make sure that the relative sensitivity variation between the substance to be measured and the reference substance stayed within $\pm 20\%$ as compared with the relative sensitivity measured at the time when the measurement lines were drawn. If an abnormal value was obtained, the same procedure described above for benzo(a)pyrene was followed.

(3) Substances subjected to surveillance

The substances surveyed in this surveillance include benzo(a)pyrene and phthalate diesters that are suspected of having an endocrine disrupting effect in the SPEED '98, considered to be discharged into the air, and for which a measuring method is established.

The surveillance was also conducted for future reference on substances not listed in the SPEED '98 but measurable at the same time. Substance numbers

assigned in the SPEED '98 are provided below just for reference.

Substances surveyed		
	Substances	SPEED'98
Benzo(a)pyrene	Benzo(a)pyrene	43
	Benzo(k) fluoranthene*	-
	Benzo(ghi) perylene*1	-
Phthalate diesters	Diethyl phthalate	42
	Di-n-propyl phthalate	65
	Di-n-butyl phthalate	40
	Di-n-pentyl phthalate	63
	Dihexyl phthalate	64
	Butyl benzyl phthalate	39
	Di-(2-ethylhexyl) adipate	45
	Dicyclohexyl phthalate	41
	Di-(2-ethylhexyl) phthalate	38
Di-iso-nonyl phthalate	-	

*1 Since benzo(k) fluoranthene, benzo(ghi) perylene and di-iso-nonyl phthalate are measurable at the same time, they were analyzed together with the substances listed in the SPEED '98.

*Benzo(a)pyrene is found in emissions from automobiles, cigarette smoke, and other exhaust gases produced by combustion, and it has been pointed out since a long time ago that it is a carcinogenic substance. It is also listed as a substance suspected of being an endocrine disrupting chemical (hormone disrupting chemical). As one of the 22 "high-priority harmful substances" against which countermeasures are required to be taken immediately, it has been regularly monitored since 1999 under the Air Pollution Control Law.

*Phthalate diesters are added mainly as a plasticizer for plastic products, and is found everywhere around us since plastic is used almost on all products we have

at home. It is listed as a substance suspected of being an endocrine disrupting chemical (hormone disrupting chemical).

(4) Summarized results of surveillance

Results of benzo(a)pyrene measurement

With regard to the measurement results for the general environment and roadside areas, minimum and maximum values calculated from the measurement for each district are shown in Tables 1-1 and 1-2.

Benzo(a)pyrene was detected within the following ranges.

- Industrial district: 0.056 ~ 2.4ng/m³
- Residential district: 0.048 ~ 1.9ng/m³
- Suburban district: (0.021) ~ 2.4ng/m³
- Roadside area: 0.14 ~ 1.4ng/m³
- Hinterland: 0.11 ~ 1.1ng/m³

These values were within the ranges of values obtained from the surveys conducted so far by the Environment Agency.

Similar results were produced for benzo(k) fluoranthene and benzo(ghi) perylene that were analyzed during this surveillance for future reference. A value larger than the minimum limit value of detection but smaller than the minimum limit value of determination is shown in ().

Measurement results for each site are shown in Tables 1-3 and 1-4.

Phthalate diesters measurement result

The measurement results showing minimum and maximum values calculated for each district are given in Table 2-1.

Phthalate diesters, di-n-butyl phthalate and di-(2-ethylhexyl) phthalate were found almost in half of the samples. However, since blank control is difficult for these three substances as compared with others, the minimum limit value of detection was set to a higher value. Di-(2-ethylhexyl) adipate was detected in about 70% of the samples.

Di-n-propyl phthalate, di-n-pentyl phthalate and dicyclohexyl phthalate were not practically detected, while dihexyl phthalate was not found at all. Di-n-propyl phthalate, di-n-pentyl phthalate and dihexyl phthalate are not produced in our country, and it can be said that these results reflected the domestic production and consumption amounts of the chemicals.

Diethyl phthalate, di-n-butyl phthalate, di-(2-ethylhexyl) adipate and di-(2-ethylhexyl) phthalate were detected within the following ranges.

Unit:ng/ m³

	Volume ranges of substances detected (total)	Industrial district	Residential district	Suburban district
Diethyl phthalate	<1.7 ~ 18	<1.7 ~ 18	<1.7 ~ 8.7	<1.7 ~ 8.2
Di-n-butyl phthalate	<20 ~ 160	<20 ~ 100	<20 ~ 140	<20 ~ 160
Di-(2-ethylhexyl) adipate	<0.58 ~ 21	<0.58 ~ 8.7	<0.58 ~ 21	<0.58 ~ 5.9
Di-(2-ethylhexyl) phthalate	<33 ~ 360	<33 ~ 170	<33 ~ 320	<33 ~ 360

Note: Values smaller than the minimum limit value of detection are indicated as < minimum limit value of detection.

The values for diethyl phthalate and di-(2-ethylhexyl) adipate were about the same, while the values of di-n-butyl phthalate were about ten times larger than them, and the values of di-(2-ethylhexyl) phthalate were approximately 13 to 20 times larger than them.

As compared with the values of di-n-butyl phthalate, di-(2-ethylhexyl) phthalate and di-(2-ethylhexyl) adipate obtained from the surveys conducted so far by the Environment Agency, the values obtained during this surveillance were within the ranges of the past values.

Di-iso-nonyl phthalate surveyed in this surveillance for future reference was not practically detected.

The measurement results for each site are shown in Table 2-2.

2. Steps to be taken in the future

This surveillance was conducted under the SPEED '98 (Strategic Programs on Environmental Endocrine Disruptors) to assess the nationwide pollution status with emphasis on substances that were selected from the 67 chemicals suspected of having an endocrine effect as those that are highly likely to be discharged into the ambient air. Of the chemicals suspected of having an endocrine disrupting effect, those whose concentration in the air can be measured will be surveyed one after another in order to assess their concentration levels in the environment, and thereby to extend our knowledge.