Verification of the Effect on Risk Due to Reduction of Benzene Discharge

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Abstract

The time course of ambient benzene level and benzene discharge was investigated. Data obtained by continuous monitoring and monthly monitoring showed a decreasing trend of ambient benzene level. The rate of decrease was around 15-30 % per two years from FY 1997 to FY 1999. The discharge data of benzene reported by several organizations were collected and arranged. The total amount of benzene discharged decreased by 25% from 1997 to 1999. Risk reduction due to the regulation of benzene content in gasoline as predicted in our previous report was verified by the obtained data.

Key Words: benzene, nitrogen oxide, health risk, exposure, automobile, population risk

1 Introduction

Benzene is an aromatic volatile organic compound characterized by the U.S. Environmental Protection Agency (U.S. EPA) as a "known" human carcinogen via all routes of exposure based upon convincing human evidence, as well as supporting evidence from animal studies (U.S. EPA, 1998). In Japan, the Air Pollution Control Law was revised in 1996 and the

Environmental Quality Standard level for benzene was established at 3 ug/m^3 in 1997. The government enforced the regulation, that the permissible upper limit of benzene content in gasoline should be decreased to 1 vol. % from January 2000.

In our previous study (Kajihara et al., 2000a; Kajihara et al., 1999; Kajihara and Ishizuka, 1998), we evaluated the aggregate population cancer risk due to ambient benzene for the entire Japanese population, using data of ambient NOx levels measured at air-pollution-monitoring stations nationwide, and the regression equation between the levels of benzene and NOx. The population-weighted exposure levels for the roadside population and the general-environment population were calculated to be 6.7 ug/m^3 and 3.2 ug/m^3, respectively, at the 1997 benzene level, and 84 % of the entire population was exposed to a lifetime cancer risk level of 1 x 10^{-5} or greater. The annual number of cancer deaths was estimated to be 29.6 cases. Due to the regulation that established the upper limit of benzene content in gasoline at 1 vol. %, the total emission of benzene was predicted to be reduced by 27% as compared to the emission in 1993. The annual number of cancer deaths was expected to be reduced by 8.8 cases through the regulation of benzene content in gasoline. In this report, the time courses of benzene level, discharge and risk were investigated using monitoring data and reported discharge data .

2 Time course of benzene level

We have used the data of nitrogen oxide (NO_x), considered to be a surrogate material for benzene, because the use of NOx data would allow greater spatial resolution in the assessment of benzene exposure if NO_x and benzene levels were highly correlated. The relationship between ambient NOx and benzene levels was investigated by monitoring one location. The ambient benzene and NOx levels were monitored from May 1997 to October 2000 at the Institute of Environmental Science and Technology located on the campus of Yokohama National University (YNU) in Yokohama City. The apparatus and the details of the method for monitoring VOC including benzene and NOx are the same as those of a previous report (Kajihara et. al., 2000a).

The time course of change in benzene and NOx levels over a three and a half year period is shown in Figure 1. In some periods, plots for NOx and/or benzene are missing because the pollutants' levels could not be measured due to trouble with the experimental apparatus. The levels of the two pollutants showed similar seasonal trends: the levels in winter were higher than those in summer. However, it was difficult to clarify the decreasing trend because the seasonal change was too large and there were too many missing data. In our previous report (Kajihara et. al., 2000a), the regression equation (1) below was obtained by linear regression analysis between the daily average data of NOx and benzene levels measured in the period from May 1997 to Oct. 1998.

$$\boldsymbol{B} = 0.067\, \boldsymbol{N} + 0.90 \qquad (R^2 = 0.74) \qquad (1)$$

Here, ***B*** represents the concentration of benzene in ug/m^3 and ***N*** represents the concentration of NOx in ppb. Benzene and NOx levels were strongly correlated.

In order to investigate the time course of the relationship between NOx and benzene levels, the regression equations obtained from the data measured in fiscal years (FYs) 1997, 1998, 1999, are shown below.

$$\boldsymbol{B} = 0.064\, \boldsymbol{N} + 1.04 \qquad (R^2 = 0.79, \text{FY 1997}) \qquad (2)$$

$$\boldsymbol{B} = 0.067\, \boldsymbol{N} + 0.81 \qquad (R^2 = 0.65, \text{FY 1998}) \qquad (3)$$

$$\boldsymbol{B} = 0.039\, \boldsymbol{N} + 2.31 \qquad (R^2 = 0.40, \text{FY 1999}) \qquad (4)$$

Here, Eq. (2) corresponds to FY 1997, Eq. (3) corresponds to FY 1998, and Eq. (3) corresponds to FY 1999. The data that were used to derive each regression equation did not include the missing data due to apparatus trouble shown in Fig.1.

The coefficient of determination, R^2, which represents the degree of correlation was smaller for Eq. (4) than for the others. This seemed to be caused by the accidental and temporary increase in benzene level when the wind was easterly in August and September 1999 (Kajihara et. al. 2000b). This accidental and temporary phenomenon seemed to be caused by the existence of an emission source of benzene on the east side of the monitoring point in these two months. The large-scale petrochemical complex located about 10 km east of the monitoring point might be related to it. By excluding the data measured in August and September 1999, the regression equation showed strong correlation.

$$\boldsymbol{B} = 0.050\, \boldsymbol{N} + 1.39 \qquad (R^2 = 0.70, \text{FY1999 without Aug. and Sep.}) \qquad (5)$$

The slope of the regression equation was found to decrease by 13-14%, from 0.064 of Eq.(2) or 0.067 of Eq. (3) to 0.050 of Eq. (5). This decreasing rate of the slope, 13-14% per two years, was roughly regarded as the time course of benzene level in Yokohama. The intercepts of the regression lines, which correspond to the background level of benzene, seemed to be constant at around 1 ug/m^3 from 1997 to 1999. Regression lines (2),(3) and (5) are shown in Fig.2.

Benzene levels have been monitored from 1997 by the Japan Environment Agency and municipalities at a frequency of about once a month or less. The annual average benzene levels at nine monitoring points, where benzene levels have been measured continuously every month from FY 1997 to FY 1999, were 3.3ug/m^3 in FY 1997 (JEA, 1998), 3.0ug/m^3 in FY 1998 (JEA 1999), and 1.9ug/m^3 in FY1999 (JEA, 2000). On the other hand, the annual average NOx levels at the same monitoring points were 38 ppb in FY 1997, 36 ppb in FY 1998, and 33 ppb in FY 1999. The ratios of benzene level to NOx level monitored at the same points were 0.089 in FY 1997, 0.083 in FY 1998, and 0.057 in FY 1999.

The reduction ratio of benzene level to NOx level over a two-year period was 36%. Ambient benzene level data obtained by continuous monitoring in Yokohama and those obtained by nationwide monthly monitoring by JEA and municipalities (JEA 2000) showed a decrease in the ambient benzene level compared to the NOx level in Japan . The rate of decrease of ambient benzene level in Japan seems to be about 18-36% from 1997 to 1999.

3. Time course of benzene discharge

The government enforced a regulation that the permissible upper limit of benzene content in gasoline should be decreased to 1 vol. % from the level of 5 vol. % from January 2000. The discharge data of benzene were reported by various organizations, such as governmental standing committees or guild associations. who however, often quoted each other's values, making identification of the original value and who estimated the value difficult. In our previous work (Kajihara et. al., 2000a) , we collected data of the time course of benzene emission reported by three organizations, Petroleum Association of Japan (PAJ), Petroleum Council and Central Environment Council.

In this report, as newly released discharge data of benzene were available from some guild associations, the time course of total benzene discharge in recent years could be re-estimated. The Petroleum Association of Japan has released the time course of benzene content in gasoline and the discharge data from gasoline vehicles and petroleum institutes. The Japan Chemical Industry Association has released discharge data from the chemical industry. The Iron and Steel Institute of Japan and the Japan Gas Association have released discharge data from coke furnace. Japan Paper Association has released discharge data of benzene as incineration by-products.

Benzene discharges before regulation and in 1995, 1997 and 1999 were estimated and are shown in Table 1 and Fig.3. Though the date of the discharge data before regulation could not be accurately identified, it seemed to be around 1993 or 1994. The total discharge of benzene, which was about 22,000 tons before regulation, was estimated to slightly decrease to about 20,000 tons in FY 1997, and to 15,000 tons in FY 1999. The decreasing ratio from 1997 to 1999 was about 25%. The decreasing ratio of ambient benzene level was not so much different from the decreasing rate of benzene discharge, as shown in Fig.3.

4. Relationship between population and ambient benzene level

The population distribution exposed to ambient benzene was evaluated using NOx data measured at monitoring stations nationwide and the regression equation (1) between the levels of benzene and NOx for FY 1997. The method for correlating the population to exposure levels was the same as previous work (Kajihara et. al., 2000a). Benzene levels were assumed to decrease by 30 % as already mentioned. Histograms of the populations exposed to each ambient benzene level in FY 1997 and in FY 1999 are shown in Fig. 4.

The benzene level in which the largest population was included changed from 2-3 ug/m^3 to 1-2 ug/m^3. The ratio of the population those was exposed to benzene levels greater than the EQS level for ambient benzene of 3 ug/m^3 decreased from 54% to 21%. However, in the case of the roadside population, the ratio of the population exposed to levels greater than the EQS level of slightly decreased from 98% to 89%.

4. Estimation of risk

The excess lifetime cancer risk of the population living in each region was calculated as the product of the personal exposure level and the unit risk for benzene. The following equation was led as the relationship of personal exposure level and ambient level for benzene when the ambient benzene level was within 0-20 ug/m^3 in Japan (Ono, 1998).

$$B_{personal} = 22.86\, B_{ambient} / (4.265 + B_{ambient}) \qquad (6)$$

Here, $B_{personal}$ represents the personal exposure level of benzene in ug/m^3, and $B_{ambient}$ represents the outdoor benzene level where the man live in ug/m^3. The equation (6) was used for estimating the personal level in each region.

The unit risk for benzene was suggested to be in the range of 2.2×10^{-6} (m^3/ug) - 7.8×10^{-6} (m^3/ug) by the US EPA(1998). We used 5.0×10^{-6} (m^3/ug) as the unit risk, which is the average value of the suggested range. Histograms of the populations exposed to each personal benzene level or benzene induced excess lifetime cancer risk in FY 1997 and in FY 1999 are shown in Fig. 5.

The aggregate population risk of the total population in Japan was estimated using Eq. 7, which is the sum of the annual population risk for each region, obtained by multiplying the annual individual risk by the population for each region.

$$R = (\Sigma U B_i P_i) / L \qquad (7)$$

Here, R is the aggregate population excess cancer risk caused by one year's exposure to benzene. U is the unit risk for benzene, B_i is the personal exposure level in region i, P_i is the population in region i and L is the average lifetime, set as 70 years. The aggregate cancer risk, R, was estimated to be 86 cases for benzene level in 1997 and 69 cases for the level in 1999.

The risk reduction was estimated to be 17 cases.

5 Summary and Conclusions

The time courses of ambient benzene level and benzene discharge were investigated. The rate of decrease of the ambient benzene level was around 15-30 % per two years from FY 1997 to FY 1999. The discharge data of benzene were found to be decreased by 25% from 1997 to 1999. The change of the ambient benzene level was well explained by the change of the discharge data. Risk reduction due to the reduction of benzene discharge was estimated to be 17 cases per year.

6 Acknowledgments

This work was supported by CREST (Core Research for Evolutional Science and Technology) of Japan Science and Technology Corporation (JST).

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TABLE 1. Estimation of Change in Benzene Discharge

Fiscal year	before regulation	1995	1997	1998	1999
Benzene in Gasoline (vol. %) [a]	2.3	2.2	1.4	1.1	under 1%
Vehicle					
Gasoline Vehicle [a]	9496	9840	9450	7100	5896
Gasoline Motorcycle [c,e]	5096	4978	4133	3816	3605
Diesel Vehicle [d]	1600	1600	1600	1600	1600
Discharge from storage, shipment and supply processes of petroleum[a]	1333	1337	1019	849	671
Discharge from production and usage processes of benzene [c] [b]	3960	4251	3287	2504	2740
Others(Coke furnace, Incinaration byproducts) [f] [g]	760	760	504	329	459
Total Discharge	22245	22766	19993	16198	14971

a)Japan Petroleum Association, b)Japan Chemical Industry Association, c) Petroleum Council, d) Central Environment Council, e) Estimation of this work, f) The Iron and Steel Institute of Japan, g) Japan Paper Association

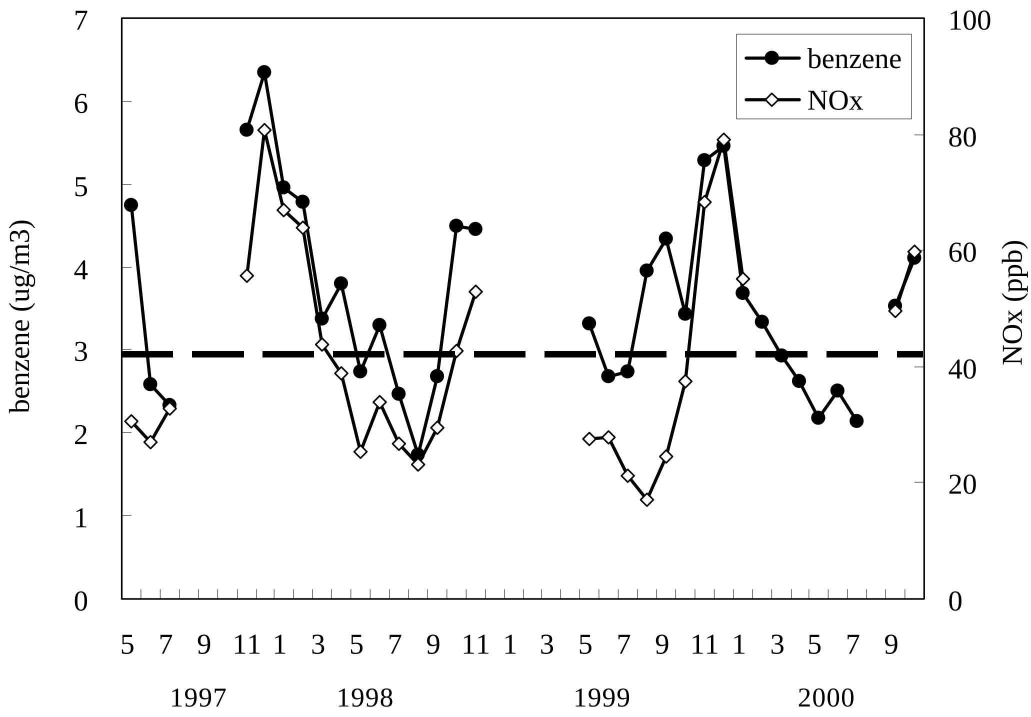

Fig.1 Seasonal trends of the levels of benzene and NOx monitored at Yokohama. Dotted line represents the Environmental Quality Standard level for ambient air.

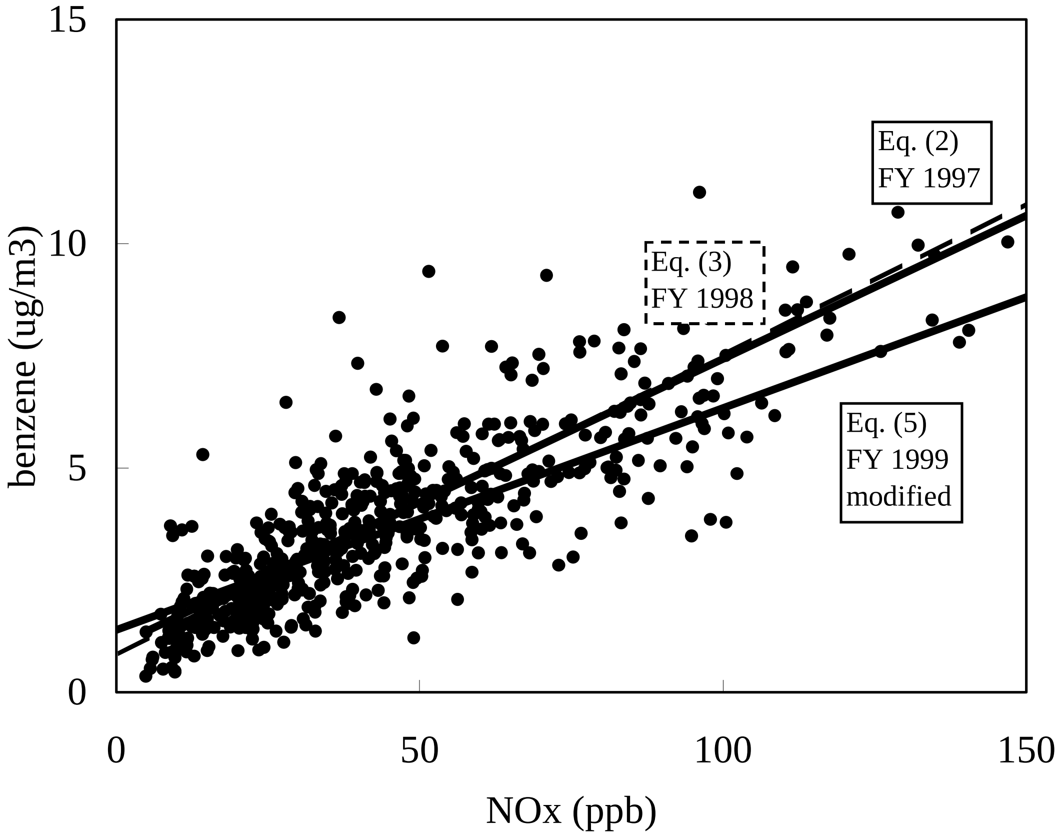

Fig.2 Regression lines between NOx and benzene levels for the data for FY 1997, FY 1998 and FY 1999. The dotted line representing FY 1998 almost overlapped with the line for FY 1997.

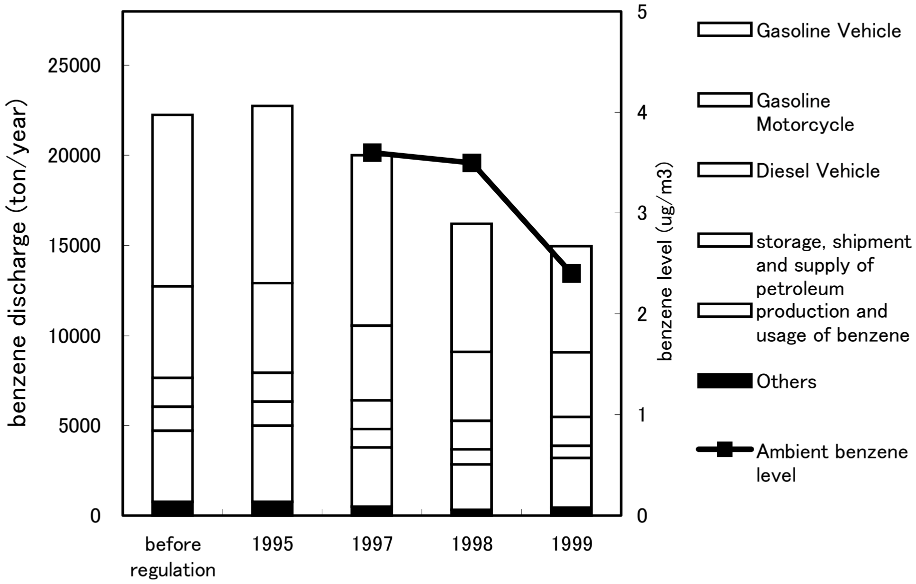

Fig. 3 Comparison of time course of benzene discharge and ambient benzene level in Japan in the 1990s.

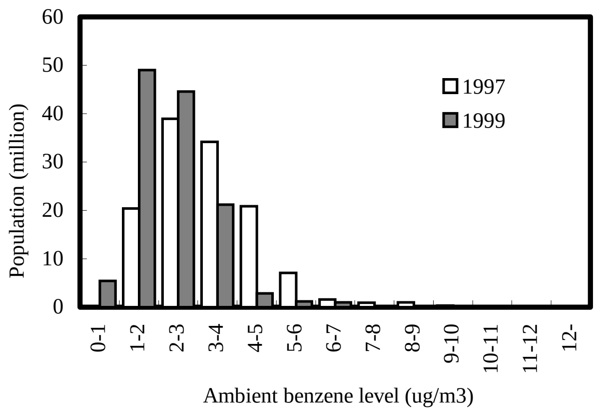

Fig. 4 The change of histograms representing the relationship between the ambient benzene level and the Japanese population living there.

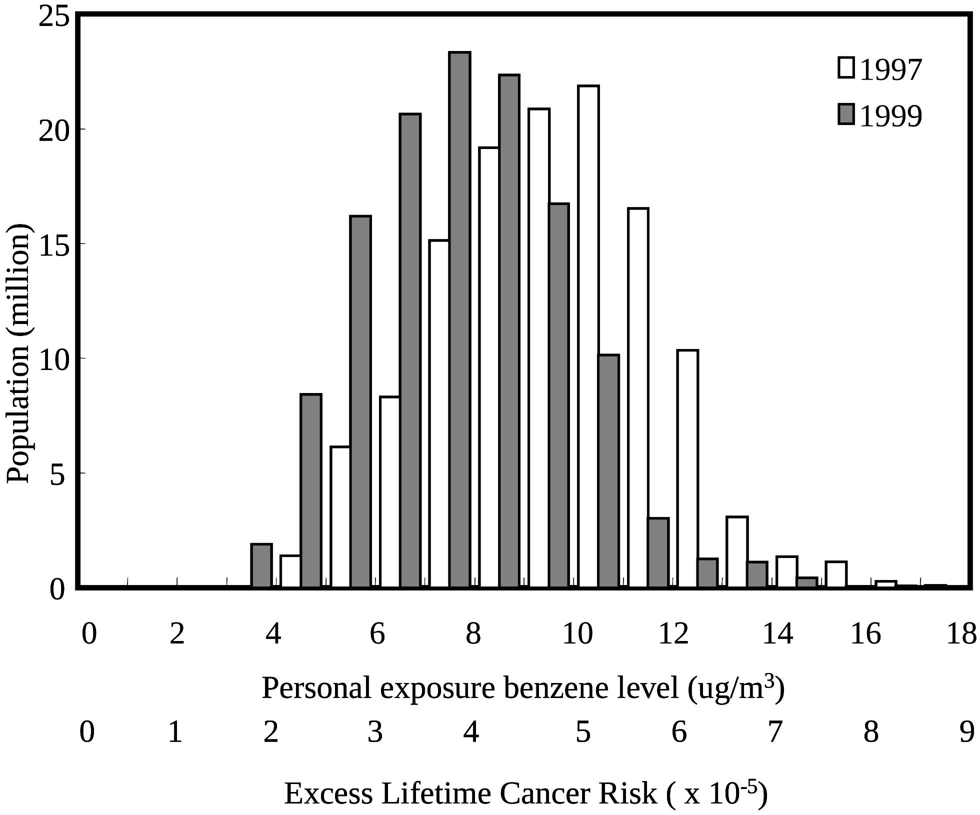

Fig.5 Histogram of personal exposure level and excess lifetime cancer risk of benzene to which the Japanese population was exposed in