

Health Effects due to the Reduction of Benzene Emission in Japan

Hideo Kajihara ¹, Akihiro Fushimi ²

¹ Graduate School of Science and Technology, Niigata University, 8050, Ikarashi 2nocho, Niigata, 950-2181, JAPAN

kajihara@gs.niigata-u.ac.jp

² Institute of Environmental Science and Technology, Yokohama National University, 79-7 Tokiwadai, Hodogaya-ku, Yokohama, Kanagawa, 240-8501, JAPAN

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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 shown to be adequate.

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 for all routes of exposure based upon convincing human evidence, as well as supporting evidence from animal studies [1]. In Japan, the Air Pollution Control Law was revised in 1996 and the Environmental Quality Standard level for benzene was established at 3 $\mu\text{g}/\text{m}^3$ in 1997. The government enforced the regulation, that the permissible upper limit for benzene concentration in gasoline should be decreased to 1 vol. %, from January 2000.

In our previous study [2 - 4], we evaluated the aggregate population cancer risk due to ambient benzene for the entire Japanese population, using data of ambient NO_x levels measured at air-pollution-monitoring stations nationwide, and the regression equation between the levels

of benzene and NO_x. The population-weighted exposure levels for the roadside population and the general-environment population were calculated to be 6.7 ug/m³ and 3.2 ug/m³, respectively, at the 1997 benzene level, and 84 percent 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 prediction of the time course of benzene level, discharge and risk were investigated by using the monitoring data and reported discharge data .

2 Time course of benzene level

We have used the data of nitrogen oxide (NO_x) as a surrogate material for benzene, because the use of NO_x would allow for greater spatial resolution in the assessment of benzene exposure if NO_x and benzene levels were highly correlated. The relationship between ambient NO_x and benzene levels was investigated by monitoring one location. The ambient benzene and NO_x 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 NO_x are the same as those of previous report[2].

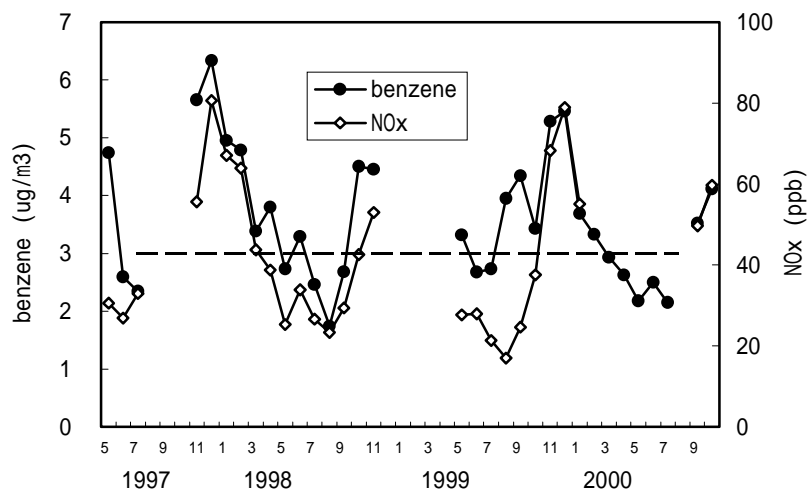


Fig.1 Seasonal trends of the levels of benzene and NO_x monitored at Yokohama. Dotted line represents the Environmental Quality Standard level for ambient air.

The time course of change in benzene and NOx levels over a three and a half year period is shown in Figure 1. In the period where plots for NOx and/or benzene are missing, the pollutants' levels could not be measured due to trouble with 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

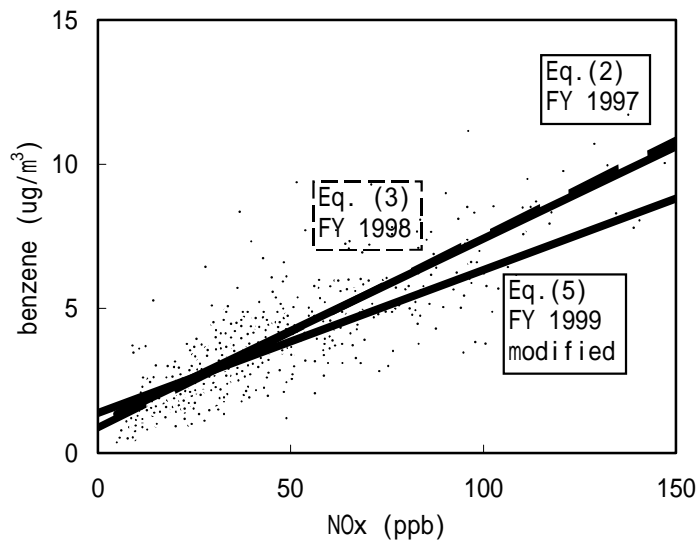


Fig.2 Regression lines between NOx and benzene levels for the data for FY 1997, FY 1998 and FY 1999. The dotted line representing FY 1998 almost overlapped with the line for FY 1997.

too large and there were too many missing data. In our previous report[2], 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.

$$B = 0.067 N + 0.90 \quad (R^2 = 0.74) \quad (1)$$

Here, B represents the concentration of benzene in $\mu\text{g}/\text{m}^3$ 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.

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

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

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

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 strength of correlation for Eq. (4) was smaller than others. This seemed to be caused by the accidental and temporary increase in benzene level only happened when wind was easterly in August and September 1999 [5]. This accidental and temporary phenomenon seemed to be caused by the existence of an emission source of benzene on the east side of monitoring point in these two months. Though this accidental and temporary benzene emission source could not be identified, the fact that there is 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.

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

The slope of the regression equation was found to decrease 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 intercept 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),(5) are shown in Fig.2.

Benzene levels have been monitored from 1997 by the Japan Environment Agency and municipalities at frequency of about once a month or less. At 46 monitoring points, benzene levels have been measured continuously every month from FY 1997 to FY 1999. The average benzene levels monitored monthly at the 46 points were 3.6 ug/m^3 in FY 1997, 3.5 ug/m^3 in FY 1998, and 2.4 ug/m^3 in FY1999 [4]. The reduction ratio for two years was 33%. Ambient benzene level data obtained by continuous monitoring in Yokohama and those obtained by nationwide monthly monitoring by JEA and municipalities[6] both showed a decrease in the ambient benzene level in Japan. The rate of decrease seems to be about 18-33% from 1997 to 1999.

3. Time course of benzene discharge

The government enforced a regulation that the permissible upper limit for benzene concentration 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. However, the reported discharge values were often quoted each other, then identification of the original value and who estimated the value was difficult. In our previous work[2], we collected the time course of benzene emission reported by three organizations, Petroleum Association of Japan (PAJ), Petroleum Council[7] and Central Environment Council[8].

TABLE 1. Estimation of Change in Benzene

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, Incineration 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

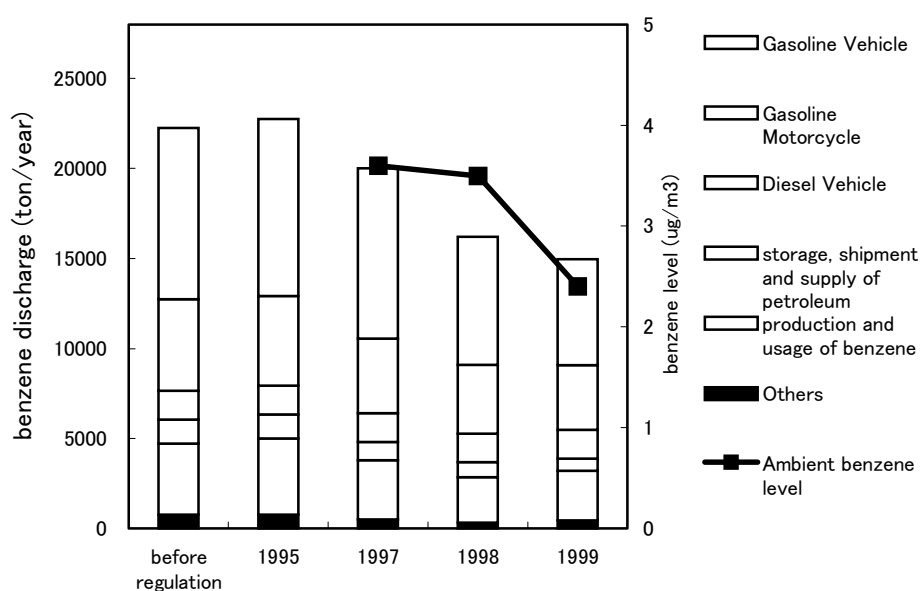


Fig. 3 Comparison of time course of benzene discharge and ambient benzene level in Japan in the 1990s.

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 discharge data from gasoline vehicle and petroleum institute. 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 have released discharge data of benzene as incineration by-products.

Benzene discharges before regulation and in 1995, 1997, 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 seems to be nearby 1993 or 1994. The total discharge of benzene, which was about 22000 tons before regulation, was estimated to slightly decrease to about 20000 tons in FY 1997, and largely decrease to 15000 tons in FY 1999. The decreasing ratio from 1997 to 1999 was about 25%. The decreasing rate of ambient benzene level was not so much different from the decreasing rate of benzene discharge as shown in Fig.3.

4. Reduction of risk

Population risk 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 [2]. Benzene levels were assumed to decrease by 30 % as already mentioned. Histograms of the populations exposed to each 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 $\mu\text{g}/\text{m}^3$ to 1-2 $\mu\text{g}/\text{m}^3$. The ratio of the population those was exposed to benzene levels greater than 3 $\mu\text{g}/\text{m}^3$ decreased from 54% to 21%. However, in the case of the roadside population, the ratio of the population exposed to levels greater than 3 $\mu\text{g}/\text{m}^3$ slightly decreased from 98% to 89%.

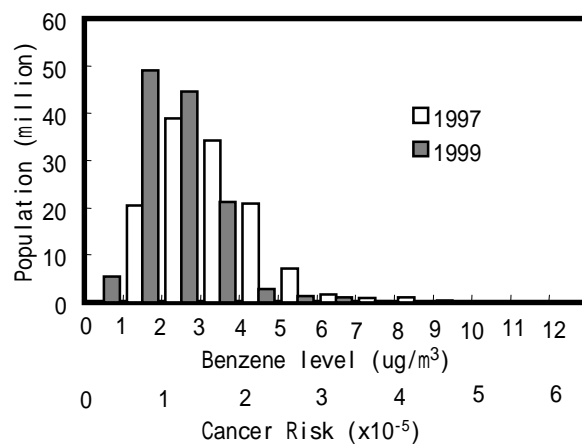


Fig. 4 The change of histograms for the benzene level and cancer risk due to benzene to which the Japanese population was exposed.

4 Summary and Conclusions

The time course of ambient benzene level and benzene discharge was investigated. The rate of decrease of ambient benzene level was around 15-30 % per two years from FY 1997 to FY 1999. The discharge data of benzene was found to be 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 shown to be adequate.

5 Acknowledgments

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References

1. U.S. EPA (1998): Carcinogenic Effects of Benzene: An Update. Washington DC: Office of Research and Development, EPA/600/P-97/001F.
2. Kajihara H et al (2000): Population risk assessment of ambient benzene and evaluation of benzene regulation in gasoline in Japan, *Environmental Engineering and Policy*, 2: 1-9.
3. Kajihara H et al (1998): Evaluation of human health risk due to benzene exposure in Japan, *Proceedings of the 1st International Workshop on Risk Management of Chemicals*, Yokohama, pp59-66.
4. Kajihara H et al (1999): Exposure assessment of benzene from vehicles in Japan, *Proceedings of the 2nd International Workshop on Risk Management of Chemicals*, Yokohama, pp. 62-70.
5. Kajihara H (2000)et al: Continuous monitoring of benzene in urban atmosphere ~ effect of benzene regulation in gasoline ~, *The Abstracts for 9th Symposium on Environmental Chemistry*, pp500-501.
6. Japan Environment Agency (2000): *The Result of Hazardous Air Pollutants Monitoring in fiscal 1999*, (in Japanese).
7. The expert committee for the quality of the petroleum products (1996): *The report for the desirable quality hereafter of petroleum products*. Tokyo: Petroleum Council (in Japanese)
8. The expert committee for automobile exhaust gases (1996): *The midterm report on the countermeasure for reduction of automobile exhaust gases*. Tokyo: Central Environment Council (in Japanese).