

Available online at www.sciencedirect.com



JOURNAL OF ENVIRONMENTAL SCIENCES <u>ISSN 1001-0742</u> CN 11-2629/X www.jesc.ac.cn

Journal of Environmental Sciences 19(2007) 709-713

# Impacts of converting from leaded to unleaded gasoline on ambient lead concentrations in Jakarta metropolitan area

Akira KONDO<sup>1</sup>, Esrom HAMONANGAN<sup>2,\*</sup>, Satoshi SODA<sup>1</sup>, Akikazu KAGA<sup>1</sup>, Yoshio INOUE<sup>1</sup>, Masaharu EGUCHI<sup>3</sup>, Yuta YASAKA<sup>3</sup>

Graduate School of Engineering, Osaka University, Yamada-oka 2-1, Suita, Osaka 565-0871, Japan. E-mail: kondo@see.eng.osaka-u.ac.jp
Environmental Management Center, Komplek PUSPIPTEK, JI. Raya, Puspiptek, Serpong, Bantan 15314, Indonesia
Research Center for Environmental Preservation, Osaka University, Yamada-oka 2-1, Suita, Osaka 565-0871, Japan

Received 28 June 2006; revised 28 September 2006; accepted 14 October 2006

#### Abstract

Total suspended particulate mater (TSP) concentrations were monitored for one year from July 2000 and for one year from April 2003 in Jakarta City. Thirteen elemental TSP components, aluminum (Al), sodium (Na), iron (Fe), lead (Pb), potassium (K), zinc (Zn), titanium (Ti), manganese (Mn), bromine (Br), copper (Cu), chromium (Cr), nickel (Ni), and vanadium (V) were analyzed by a sequential X-ray fluorescence spectrometer. Al, Na, Fe, K, and Pb were major components at most of the sampling locations in 2000. However, only Pb in 2003 dramatically decreased to one tenth. The phase-out of leaded gasoline began on July 1, 2001 in Jakarta City and lead content in gasoline decreased to one tenth, too. The decrease in Pb concentration was a result of the phase-out of leaded gasoline, as lead emissions mainly are exhaust gas from vehicles.

Key words: TSP concentration; lead concentration; unleaded gasoline

# Introduction

Jakarta is situated on the northern coast of Java Island around the mouth of the Ciliwung River. It covers an area of approximately 665 km<sup>2</sup> along the coastline. The topography is very flat, with an average elevation of 7 m above sea level. The two main seasons, the dry season (April to September) and the wet season (October to March), have an average temperature of 27°C, with high relative humidity, from 50% to 100% (EMC, 1994).

The rapid growth of motor vehicle use, industry, and population from 1950 to the present has strongly affected the region's environmental quality. A disparity between the number of motor vehicles and available road length has created a traffic jam problem in every section of the city, and has caused air quality deterioration. Moreover, old vehicles are still operated and worsen the air quality problem.

Air quality in Jakarta City has been monitored under several projects led by: (1) Environmental Management Center of Indonesia (EMC, 1994); (2) Japan International Cooperation Agency (JICA) and Environmental Impact Management Agency of Indonesia (EIMA) (JICA, 1995); (3) Bapedal and East Java Pollution Control Implementation Project (PCI project) (Cohen *et al.*, 1997); (4) Kantor Pengkajian Perkotaan dan Lingkungan DKI-Jakarta (KP- PL, 1997); and, (5) Gippsland Center for Environmental Science, Monash University (Zou and Hooper, 1997). JICA (1995) have operated automatic ambient air monitoring instruments at 4 points in Jakarta City since 1993. However, calibration and maintenance of the automatic instruments has not been sufficient because of insufficient operational budgets and a lack of air pollution control experts. Thus, the continuous monitoring could not be maintained. As part of the present study, we, in cooperation with EMC, have measured air quality at 20 points in the Jakarta City area for one year, beginning in July 2000. Measured and numerically simulated results of the distribution of gaseous air pollutants, sulfur dioxide (SO<sub>2</sub>), nitrogen dioxide  $(NO_2)$ , and nitrogen oxides  $(NO_x)$  have been previously reported (Hamonangan et al., 2002, 2003). Almost the identical monitoring configuration was initiated in 2003 through a JICA phase II program, again in cooperation with EMC.

In addition to gaseous air pollutants, total suspended particulate mater (TSP, aerodynamic diameter  $<10 \mu$ m) is also a significant air pollutant. The serious public health effects of lead, from petrol combustion and lead processing, are widely recognized. The USA, Japan, Sweden, Canada, parts of Europe, and Australia have completely eliminated lead in petrol (Thomas and Kwong, 2001). In most European Union countries the permissible amount of lead in petrol is highly limited. In Africa, Latin America, and most of Asia, however, there are few restrictions on

<sup>\*</sup>Corresponding author. E-mail: kondo@see.eng.osaka-u.ac.jp.

lead in petrol and the lead concentration level in African cities is ten times higher than that of the typical European cities.

In Jakarta, a lead phase-out program was inaugurated on 1 July 2001, replacing leaded gasoline with unleaded. Our monitoring was performed before and after the phaseout of leaded gasoline. The elemental components of TSP, as well as TSP concentrations in 2000 and in 2003 are reported in the present paper. The changes in lead concentrations due to the phase-out of leaded gasoline will be discussed as well.

# **1** Experiments and methods

#### 1.1 TSP sampling

### 1.1.1 Sampling locations

TSP concentrations were measured during two periods, from December 2000 to November 2001 and from April 2003 to March 2004. TSP samplers were set at 20 locations in Jakarta City during the first measurement period, as shown in Fig.1. TSP samplers during the second measurement period were set at 8 locations (1, 2, 4, 7, 8, 15, 17, and 20) in Fig.1.

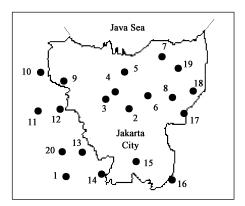


Fig. 1 Sampling points for TSP in Jakarta City area.

#### 1.1.2 Sampling instrument

An easily operational apparatus developed by the authors was used to collect TSP from ambient air (Fig.2). The instrument is inexpensive, and consists of a filter holder and a diaphragm pump, equipped with a filter made of poly tetra fluoro ethylene (PTFE) with a 25-mm diameter and 0.8 µm pore size, run at a flow rate 1.2 L/min by 1.5 V alkali battery. The flow rate was calibrated by a Shibata instrument standard orifice flow meter. The sampling period was two weeks and sampling was performed once every month from December 2000 to December 2001. Alkali batteries were exchanged every week to maintain the flow rate. However, pressure losses occurred often as sampling time passed. A low volume air sampler, with a flow rate of 6 L/min was used during the period from April 2003 to March 2004. The sampling period was one week. The TSP concentration was determined from the average flow rate and the weight of the filter before and after sampling.

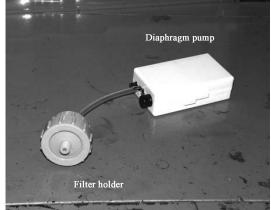


Fig. 2 TSP Sampling apparatus.

#### 1.1.3 Analysis of elemental components in TSP

Thirteen elemental TSP components, aluminum (Al), sodium (Na), iron (Fe), lead (Pb), potassium (K), zinc (Zn), titanium (Ti), manganese (Mn), bromine (Br), copper (Cu), chromium (Cr), nickel (Ni), and vanadium (V) were measured using a sequential X-ray fluorescence spectrometer (XRF-1700, Shimadzu). This method was widely used to analyze the heavy metals in TSP (Andrew et al., 2000; Tanner and Parkhurst, 2000; Alonso et al., 1997). To accurately evaluate the concentration of elemental components, a calibration standard has to be prepared in the same manner as the TSP sampling. The concentrations of the 13 elemental components were strongly different. Two kinds of calibration standards were prepared to identify the concentration of elemental components. SRM1648 (US National Institute of Standards and Technology standard reference materials) was diluted with ethanol 99%, pasted uniformly on the surface of the PTFE filter and dried at room temperature for at least 1 d. After that, the calibration standard was determined based on the relationships between the intensity energy of X-ray fluorescence and the quantity of an elemental component. Some elemental components contained only a small quantity in the filters sampled. The calibration standard for a small quantity was prepared by mixing SRM 1648 with ascorbic acid. The correlation coefficients of the 13 elemental components were over 0.95, with the exception of nickel (not shown here).

## 1.2 Lead content in gasoline

#### 1.2.1 Methods

In Indonesia, gasoline is produced by the government oil company called Pertamina. This company produces three types of gasoline, Super TT (gasoline with little lead content), Premix (gasoline with lead content of 0.15 g/L) and Premium (gasoline with 0.15 g/L lead content and a different octane number than Premix). The Super TT and Premium gasoline samples were received in May, 2002, after the phase-out of leaded gasoline, and the lead content in each gasoline was measured by D3237-97 Standard (test method for lead in gasoline using atomic absorption spectroscopy).

# 1.2.2 Lead content in gasoline before and after the phase-out

The amount of gasoline consumption and the lead content of gasoline in 2000 are shown in Table 1. The measured lead content of gasoline in 2002 is also shown in Table 1. Approximately 80% of the gasoline consumed in Jakarta is Premium. After the phase-out of leaded gasoline, the lead content of Premix and Premium decreased by about one tenth.

Table 1 Gasoline type and lead content in Jakarta City

Туре	Fuel consumption <sup>a</sup> (10 <sup>3</sup> L/a)	Lead content before July, 2001 <sup>b</sup> (g/L)	Lead content after July, 2001 (g/L)
Super TT	20680	0.005	0.012
Premix	71880	0.15	0.016
Premium	445049	0.15	0.016

<sup>a</sup>Pertamine UPPDN DKI-Jakarta (2000); <sup>b</sup>Surat keputusan dirjen MI-GAS, department ESDM (1997).

# 2 Measurement results

#### 2.1 TSP concentrations

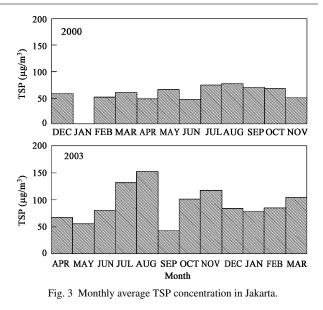
Table 2 shows the average TSP concentration and the flow rate measured in several air quality studies. The PCI project measured only  $PM_{2.5}$  (particulate mater, aerodynamic diameter <2.5 µm) concentrations, which was the lowest concentration. The TSP concentration measured by our study in 2000 was low. The flow rate at the start and the end of the sampling often changed, as described in Section 1.2.2. To calculate TSP concentration, the average flow rate (the medium value of the flow rates at the start and the end) was used. The flow rate could be underestimated if the flow rate drops quickly because of pressure loss.

The monthly average TSP concentrations in Jakarta City from December 2000 to November 2001 and April 2003 to March 2004 are shown in Fig.3. Indonesia established a national draft guideline for TSP (90  $\mu$ g/m<sup>3</sup> (annual mean) and 230  $\mu$ g/m<sup>3</sup> (24 h average)), following the World Health Organization (WHO) guidelines. Cohen *et al.* (1997) reported that the concentration during the dry season in Jakarta was higher than that in the rainy season. Seasonal changes in TSP concentrations were not as apparent in the

Table 2 Average TSP concentration measured by several studies

Reference	Average TSP concentration ( $\mu$ g/m <sup>3</sup> )	Flow rate (L/min)
a	181-500	1200 (high volume sampler)
b	265	1200 (high volume sampler)
с	42	25 (low volume sampler)
d	206	1200 (high volume sampler)
e	>260	1200 (high volume sampler)
f	119	10-20 (andersen sampler)
g	60	1.2
ĥ	94	6 (low volume sampler)

(a) World Bank, 1994; (b) EMC, 1994; (c) Cohen *et al.*, 1997; (d) JICA and EIMA, 1995; (e) KPPL, 1997; (f) Zou and Hooper, 1997; (g) This study, 2000; (h) This study, 2003.



present study, and were relatively higher than other months in July and August, partially coinciding with Cohen *et al.* (1997).

The annual average TSP concentrations at the 20 sampling locations during the two measuring periods are shown in Fig.4.

TSP concentrations in 2000 did not exceed the national draft guideline for TSP, although they may be underestimated. TSP concentrations in 2003 exceeded the national draft guideline for TSP at sampling points 7 (Tanjung Priuk), 8 (Pulogadung), 15 (Cijantung), 17 (Duren Sawit), and 20 (Pamulang). The high concentrations of TSP at these five locations were caused by heavy traffic. The TSP emissions from vehicles, including re-suspended and exhaust gas, contributed 60% of all emissions in Jakarta City (Hamonangan *et al.*, 2002). Sampling location 1 (EMC), in

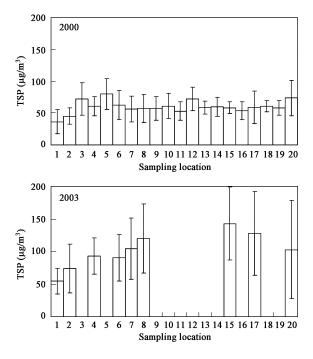


Fig. 4 Annual average TSP concentration in Jakarta.

712

a rural area, showed the lowest TSP concentration because of the lower traffic volume.

#### 2.2 Elemental component concentrations

The average percentages of 13 elemental components in 2000 and of 8 components (Al, Na, Fe, K, Pb, Zn, Ti and Cu) in 2003 are shown in Fig.5. Al, Na, Fe, and K were the major components, representing  $10^0$  order of percentage for TSP at most of the sampling locations. Pb was also a major component in 2000. Generally the soil contains a high concentration of Al, K, and Fe and sea spray from the Java Sea includes Na.

Pb is usually not contained in natural sources, and vehicle exhaust gases and lead processing are major contributors of Pb. These results suggest that rapid urbanization and motorization were detrimental to air quality in Jakarta City before 2000. Tetra alkyl lead was added as an antiknock agent in motor vehicles to raise the gasoline octane number. However, lead is detrimental to human health, impacting a child's nervous system (Parkins, 1974) and causing decreased intelligence quotient in children. Moreover, lead contributes to adult heart disease. Therefore, the Government of Indonesia started a phase-out program of leaded gasoline. The implementation of the phase-out was postponed until 2000 because of an economic crisis. Pb decreased to below  $10^{-1}$  order of percentage in 2003. The rate of Pb decrease at all locations was almost the same, and the Al, Na, Fe and K concentrations did not change from 2000. The rate of lead decrease in gasoline and lead concentration in atmosphere due to the phase-out was approximately 90%.

The phase-out of leaded gasoline in Japan began in 1975. The decrease in atmospheric lead concentrations after two years was reported to be approximately 50% (Ministry of Environment in Japan, 1978). The results of the present study suggest that the phase-out of leaded gasoline in Jakarta City was effective and widespread. The Pb concentration at sampling location 1 (EMC) in 2003 was the highest, although the concentration of 0.89  $\mu$ g/m<sup>3</sup> in 2000 decreased to 0.13  $\mu$ g/m<sup>3</sup> in 2003. These concentrations do not reflect the effects of reduced traffic volume, which is supported by the low TS concentration. In addition, lead processing operations are expected to contribute to the concentration. Countermeasures for emission sources other than gasoline are also required to decrease Pb concentrations.

# **3** Conclusions

TSP concentrations were monitored for one year from July 2000 at 20 locations and for one year from April 2003 at 9 locations in Jakarta City. Thirteen elemental TSP components were analyzed. Lead content in gasoline decreased from 0.15–0.016 g/L after the phase-out of leaded gasoline began on July 1, 2001 in Jakarta City. The TSP concentration did not exceed the national guidelines

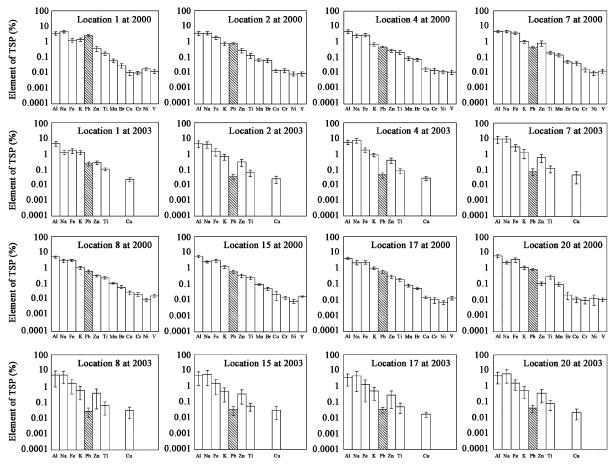


Fig. 5 Weight percentage of elemental components in TSP.

No. 6

at any locations in 2000, but a few locations exceeded the guidelines in 2003. Al, Na, Fe, K, and Pb were major components which were represented in TSP at  $10^0$  order of percentage at most of the sampling locations in 2000. However, only Pb decreased in  $10^{-1}$  order of percentage for TSP in 2003. Before 2005, the phase-out of leaded gasoline was implemented only in several cities. The effectiveness of the policy in decreasing Pb concentrations will soon require its implementation throughout Indonesia.

#### Acknowledgements

The authors greatly appreciate the financial support of the Nissan Science Foundation for part of this study. Gratitude is also extended to EMC staff, and to the JICA Decentralized Environmental Management System Project.

# References

- Alonso C D, Martins M H R B, Romano J *et al.*, 1997. Sao Paulo aerosol characterization study[J]. Journal of the Air & Waste Management Association, 47(12): 1297–1300.
- Andrews E, Saxena P, Musarra S *et al.*, 2000. Concentration and composition of atmospheric aerosols from the 1995 SEAVS experiment and a review of the closure between chemical and gravimetric measurement[J]. Journal of the Air & Waste Management Association, 50(5): 648–664.
- Cohen D, Gras J, Garton D, 1997. Study of fine atmospheric particles and gases in the Jakarta region[M]. Indonesia: Bapedal.
- EMC (Environmental Management Center), 1994. Annual report on air quality monitoring and studies[M]. Indonesia:

Bapedal.

- Hamonangan E, Kondo A, Kaga A *et al.*, 2002. Simulation and monitoring of sulfur dioxide and nitrogen oxide in the Jakarta metropolitan area[J]. Asian Journal of Energy & Environment, 3(3/4): 159–183.
- Hamonangan E, Kondo A, Kaga A *et al.*, 2003. Retrieval of emission loads from measured nitrogen oxide concentrations in Jakarta City[J]. Clean Air and Environmental Quality, 37(2): 32–36.
- JICA (Japan International Cooperation Agency), EIMA (Environmental Impact Management Agency of Indonesia), 1995. The study of integrated air quality management for Jakarta metropolitan area[M]. Indonesia: Bapedal.
- KPPL (Kantor Oengkajian Perkotaan Dan Lingkungan DKI-Jakarta), 1997. Laporan Hasil Pemantauan Udara Jakarta City[Z]. Indonesia (in Indonesian).
- Ministry of Environment in Japan, 1978. Environmental White Paper[N]. Ministry of Environment (in Indonesian).
- Parkins C H, 1974. Air pollution[M]. McGraw Hill. 354-355.
- Tanner R L, Parkhurst W J, 2000. Chemical composition of fine particles in the Tennessee Valley Region[J]. Journal of the Air & Waste Management Association, 50(8): 1299–1307.
- Thomas V, Kwong A, 2001. Ethanol as a lead replacement: phasing out leaded gasoline in Africa[J]. Energy Policy, 29(13): 1133–1143.
- World Bank, 1994. Urban air quality management strategy in Asia Jakarta report[R]. World Bank Technical Report 379.
- World Health Organization/United Nations Environmental Programme (WHO/UNEP), 1992. Urban air pollution in mega cities of the world[M]. Blackwell Oxford UK: WHO-UNEP.
- Zou L Y, Hooper M A, 1997. Size-resolved airborne particles and their morphology in Central Jakarta[J]. Journal of Atmospheric Environment, 31(8): 1167–1172.