Temporal Variations In Airborne Heavy Metals In Daegu, South Korea

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Abstract: Heavy metals released from various sources in urban environments are a public concern. In this study, heavy metal concentrations were measured in ambient air in Daegu, South Korea from 2016 to 2018. To explore the possible influences of temporal factors on the heavy metal distributions, we used data on the concentrations of airborne metals provided by the Daegu Metropolitan Institute of Health & Environment. The airborne concentrations of lead, cadmium, and arsenic tended to decrease over the study period. Lead and cadmium showed systematic seasonal variations in their airborne concentrations. The airborne concentration of arsenic tended to increase significantly in spring and fall compared with summer, whereas those of chromium and manganese did not show significant seasonal fluctuations over the study period. Our findings suggest that the airborne concentrations of some metals are strongly influenced by temporal factors.

Index Terms: Airborne concentration, Heavy metals, Temporal variation, Urban air

1. INTRODUCTION
As a consequence of industrialization and urbanization, people living in large cities can be seriously affected by environmental pollutants, including heavy metals. Since various types and levels of heavy metals accumulate in urban environments, the presence of heavy metals in the environment has caused great concern [1]. Heavy metals are widely distributed environmental pollutants that do not degrade in nature and thus have a lasting impact on ecosystems and human health [2], [3]. In humans, toxic metals such as lead (Pb), cadmium (Cd), chromium (Cr), manganese (Mn), and arsenic (As) have long half-lives and can accumulate over time [4]. Therefore, chronic exposure to metals can have various adverse effects on human health, even at low exposure levels [5], [6]. In urban areas, large amounts of heavy metals are released into the atmosphere from several sources of air pollution [7]. Daegu is one of the largest metropolises in the Republic of Korea, with a population of approximately 2.5 million [8]. Like many other cities, Daegu is facing an air pollution problem due to pollutants emitted by industries and vehicles, and the related public health issues are a major concern. Therefore, the Daegu Metropolitan Institute of Health & the Environment (DMIHE) monitors the atmospheric concentrations of many heavy metals, including Pb, Cd, Cr, Mn, and As. With changes in the levels and sources of metal emissions in Daegu, emissions into the environment change annually or seasonally. Therefore, in this study, the levels of toxic metals (Pb, Cd, Cr, Mn, and As) in the urban outdoor environment and the annual and seasonal variations in these metal concentrations in ambient air in Daegu were determined.

2 MATERIALS AND METHODS

2.1 Study Area
We measured heavy metal concentrations in ambient air in Daegu, which is located in southern Korea, from 2016 to 2018. Sampling sites were located in the middle of crowded areas (Daemyung region) in Daegu.

2.2 Instrumental Analysis of Metals
The concentrations of heavy metals in ambient air were based on the metal concentrations in airborne particulate matter provided by the DMIHE. The monthly mean Pb, Cd, Cr, Mn, and As concentrations are stored in a DMIHE database, which is available from the Daegu Air Quality Information portal (http://air.daegu.go.kr). The basic sampling and measurement procedures used for these metal species followed protocols established by the Ministry of Environment in the Republic of Korea (KMOE) and have been described elsewhere [9]. In brief, samples were collected on Whatman PM2000 glass fiber filters and pretreated with 35 mL of a HNO₃:H₂O₂ (6:1, v/v) mixture, followed by repeated heating, filtering, and resolubilization. Then, the metal concentrations were analyzed by atomic absorption spectrometry (polarized atomic absorption spectrophotometer, Z-8100 model, Hitachi, Japan). The detection limits of all metals were 0.25 ng/m³ (Cd) or 1–5 ng/m³ (Pb, Cr, Mn, and As). The basic quality assurance and control procedures for the metal measurements follow protocols established by the KMOE.

2.3 Statistical Methods
We used mean values to describe the distributions of airborne metal levels according to year or season. Trend tests of the mean airborne metal concentrations during the four seasons were conducted using linear regression analysis. All statistical analyses were conducted using SAS 9.4 (SAS Institute, Cary, NC, USA).

3 RESULTS AND DISCUSSION
The arithmetic mean concentrations (in ng/m$^3$) of all metals over the study period were 19.60 for Pb, 0.74 for Cd, 2.74 for Cr, 19.19 for Mn, and 2.60 for As. The heavy metal monitoring data obtained during the study period are shown in Fig. 1.

The mean concentrations of the elements in 2016 decreased in the order of Pb > Mn > As > Cr > Cd. However, in 2018, the concentrations were in the order of Mn > Pb > Cr > As > Cd. Similar observations in the ambient air of Coimbatore, a metropolis in India, have been reported [10]. The order of the airborne concentrations of heavy metals in Coimbatore was Pb > Cr > Cd, similar to our observations in Daegu. Pb and Mn were also the dominant metals in the urban atmosphere of the largest cities in the Slovak Republic [11]. The concentrations of most metals, except Cr and Mn, tended to decrease throughout the study period. The concentration of Cr increased slightly throughout the study period, and that of airborne Mn was higher in 2018 compared with 2017. The presence of varying concentrations of different heavy metals over the study period in the ambient air of Daegu could reflect changing patterns in vehicular transportation and industrial emissions [12]. To evaluate the significance of temporal factors, the seasonal variability in the metal concentrations could be subdivided into three different classes: (1) Pb and Cd, (2) Cr and Mn, and (3) As (Fig. 2). The Pb and Cd concentrations tended to be lowest during summer and were increased significantly during spring and fall, with the highest concentrations occurring during winter (p for trend <0.05). The seasonal differences in ambient Pb and Cd levels may be due to variations in the sources and levels of Pb and Cd in the environment. However, the Cr and Mn concentrations did not show significant seasonal fluctuations, and the airborne Cr level was relatively consistent, irrespective of temporal factors. These results suggest that important factors other than season influence the Cr concentration. Finally, the As concentration was significantly higher during spring and fall than summer (p<0.05).

The seasonal variations in each heavy metal concentration observed in this study are consistent with those in urban Beijing, China [13]. There, the Pb and Cd concentrations were highest during winter, followed by fall, and lowest in summer, similar to our results. Epidemiological studies have shown significant associations between the concentrations of air pollutants, such as Pb, Cd, and As, and adverse health impacts [14]. Air pollution contributes to eye irritation, asthma, and bronchitis, which invariably reduce health-related quality of life. Therefore, the health-related quality of life of residents could be improved by implementing integrated strategies for air pollution management in cities via regulation of the emission sources causing the temporal variations.

4 CONCLUSION

Environmental pollution is a growing social concern in Korea, and many regulatory measures have been implemented to reduce the emission of air pollutants. However, air pollution in Korean cities is still a serious problem, and large proportions of the populations living in large cities are currently exposed to toxic levels of air pollutants. In this study, we analyzed the heavy metal concentrations in Daegu, Korea during 2016–2018. The individual airborne metal concentrations in Daegu showed different temporal fluctuation patterns. A trend analysis of the annual and seasonal concentrations of the metals revealed specific patterns, resulting in classification of the metals into three groups. The information on air pollution patterns in Daegu provided by this study has implications for both scientific research and policy. The study findings will help...
researchers understand the complex temporal fluctuations in air pollutants and determine the associated risk factors for public health. This study will also be a valuable reference for the local government in planning and implementing relevant policies to improve air quality.

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