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# DECREASE IN AMBIENT AIR LEAD CONCENTRATIONS IN VARNA, BULGARIA, ASSOCIATED WITH THE INTRODUCTION OF UNLEADED GASOLINE

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**Abstract:** An examination of ambient air lead monitoring data was used to demonstrate success of banning the import and use of leaded gasoline in Bulgaria. From 1996–2007 air lead levels in Varna, the third largest city, decreased up to 63-fold.

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## INTRODUCTION

In the 21st century, levels of lead in the air in Eastern Europe were on a steady rise until the early 1990s, with the main source being the combustion of leaded gasoline [5, 9]. While by the end of 1996, many industrialized countries had completely phased out the use of leaded gasoline [18], for many countries of Central and Eastern Europe it was in 1993 that environmental action programmes began to emerge to phase-out gasoline lead content [11]. Lead is a recognized neurotoxin, which even at low exposure levels can impair the neurological and intellectual development of children [10, 11, 14]. Effects in adults include cancer, heart and neurological diseases [1, 2, and 15]. According to the European Union Law the use of leaded gasoline should be entirely eliminated by 01/01/2005 [4]. Bulgaria adopted such legislation in 1999 and agreed to discontinue the production, import and use of the leaded gasoline by the end of 2003 (http://www.unep.org/pcfv/PDF/Bulgarian-national-programme-for-lead.pdf).

Varna is the third largest city in Bulgaria with a population of about 350,000. Car traffic is the main source of airborne Pb in Varna [3]. Thus, monitoring data from Varna was thought to be representative for the determination of the programme effectiveness.

The objectives were to analyze data from 2 air-monitoring stations in Varna, Bulgaria, for the years preceding and following the law-enforced removal of lead additives to fuel in Bulgaria (2003) to determine the efficacy of the implemented regulation.

## MATERIALS AND METHODS

The study utilized air monitoring that collected data from 1996–2007 from 2 monitoring stations: One was situated in the downtown area (station “Cherno more”), and the other – in a remote suburban county (station “Vladislavovo”). Sampling units operated at the 2 locations for 10 consecutive days each month. Each air sample was collected on a perchlor vinyl filter for 8 hours a day (08:00–16:00). The results were averaged as arithmetic means for monthly and yearly values. Active sampling methods and standardized analytical methods were used.

Atomic Absorption Spectrophotometry (AAS) was used to determine the content of lead in air in the range from 0.1–20 μg/cm$^3$ in the analytical sample. The Bulgarian

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annual average Permissible Exposure Level (PEL = 0.5 µg/m$^3$ in 2005, lowered from 1.0 µg/m$^3$ in 1998) is lower than the US National Air Quality Standard quarterly average of 1.5 µg/m$^3$ issued in 1978 and revised in 2008 to 0.15 µg/m$^3$.

Since the main source of airborne lead in industrial cities, such as Varna, is leaded gasoline combustion, we used traffic data from the Regional Inspectorate for Public Health Protection and Control, to asses traffic differences among stations. The Inspectorate provided data on traffic intensity since 1999 for the 2 sampling stations. The data consisted of vehicles counts between 07:00–08:00, 09:00–10:00; and between 16:00–17:00 pm in 2 consecutive days during the most heavy traffic months in the year, namely July and/or August.

The data were statistically analyzed by years and sampling stations using analysis of variance (ANOVA).

## RESULTS

In 1996, the ambient average air Pb concentration in the downtown station was 0.192 µg/m$^3$ and gradually decreased to 0.023 µg/m$^3$ in 2003, an 8-fold drop and a statistically significant decrease ($p<0.001$) (Fig. 1). There was a similar trend in the station "Vladislavovo". At the beginning of the study the concentrations were 0.125 µg/m$^3$ and decreased to 0.002 µg/m$^3$ in 2003 ($p<0.02$), a 63-fold drop. In 2004–2007, the Pb concentrations at both stations were below the detection limit of the method.

In this study, ambient air lead concentrations were below the limits for Bulgaria and the US, and were steadily decreasing at both stations throughout the study period.

Although the decrease in Pb concentrations was similar at both stations, the difference in the values between the 2 stations is statistically significant for each year ($p<0.05$), probably due to different traffic intensity. At sampling station "Cherno more" the traffic intensity for the studied period varied from 1,410–1,634 vehicles per hour. 95% of all vehicles were cars, 0.5% motorbikes, 2.8% trucks and 1.4% buses (the rest, 0.3% of vehicles were trolleys). At station "Vladislavovo" the traffic intensity varied from 502 to 635 vehicles per hour, of which 84.4% cars, 0.7% motorbikes, 2.6% trucks and 10.8% buses (and 1.5% trolleys).

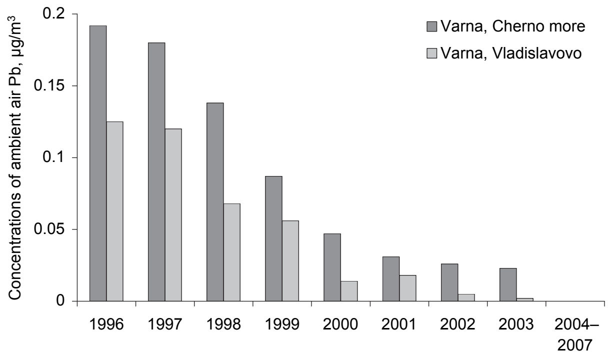


**Figure 1.** Average annual concentrations of ambient air Pb, µg/m$^3$, 1996–2007.

## DISCUSSION

The statistically significant decrease of ambient air Pb concentrations in Varna from 1996–2007 suggests that the implementation of law was sufficiently strictly-enforced to be effective. By the end of September 2003, several small gasoline stations were still selling minimal quantities of imported lead-containing gasoline. This explains the minimal but still present Pb content in air in 2003, while after 2004 Pb was not detected above the lower limit of detection (LLOD) of AAS.

As seen from the results for the traffic intensity for both stations, the number of vehicles at the downtown station "Cherno more" is 2.6–2.8 times higher than at the suburban station "Vladislavovo". In both stations, the highest % of vehicles are cars and as the traffic intensity is higher at station "Cherno more", so is the comparative Pb aerosols concentrations. Trucks and buses, being also of higher count numbers at station "Vladislavovo", use naphtha fuel and therefore they do not contribute to lead aerosols.

The 2 air monitoring stations were closed in 2008, and in 2009 there is only one station in Varna that monitors for airborne lead, located outside of the office of the Regional Inspectorate of Environment and Waters. Although permanent phase-out of leaded gasoline resulted in a dramatic decrease in airborne lead emissions in Europe [6, 16], lead continues to be emitted into the air from aircraft engines, non-exhaust emissions, such as brake wear [5], re-suspended dust containing lead emitted during the leaded gasoline era [8], and possibly diesel and gasoline combustion due to the lead content of crude oil [12]. The observed trend of higher air lead concentrations in the city centre than suburbs, and continuous decrease over the years has been also observed in other European studies. Although many other European countries reported an earlier and steeper decrease in annual average lead concentrations, the ones we report in Varna are lower than those in many other industrialized cities in Europe [7, 12, 13].

Other exposures to lead, such as that from occupations and lead-based paint, need to be investigated in Bulgaria. A recent report of lead levels in new enamel paints reported levels as high as 59,400 ppm, found in Belarus [17], suggesting that lead may also be present in new paints in Bulgaria. In view of the lack of a safe threshold for lead [19], further investigations are needed to determine the presence of other lead exposure sources in Bulgaria.

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## Disclaimer

The findings and conclusions in this report are those of the authors and do not necessarily represent the official position of the Centers for Disease Control and Prevention or of the Agency for Toxic Substances and Disease Registry.

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