Atmospheric Environment 111 (2015) 195–201

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Quantification of atmospheric lead emissions from 70 years of leaded petrol consumption in Australia

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HIGHLIGHTS

- A complete compilation of leaded petrol sales figures for Australia.
- Lead emissions were calculated from leaded petrol concentrations and sales figures.
- Nearly one quarter million tonnes of lead released from petrol across Australia.
- Peak emissions of lead from leaded petrol occurred during the 1970s.
- Elevated ambient lead levels in cities were the result of leaded petrol.

ARTICLE INFO

Article history:
Received 9 December 2014
Received in revised form
8 February 2015
Accepted 7 April 2015
Available online 8 April 2015

Keywords:
Air quality
Australia
Emission inventory
Leaded petrol

ABSTRACT

Lead is a persistent pollutant and the subject of many environmental studies, yet, in Australia, the extent of atmospheric lead emissions from the use of leaded petrol is unquantified. This paper details the first comprehensive account of leaded petrol sales and its lead concentrations over the 70 years of use in Australia. The resulting atmospheric lead emissions are calculated to provide the most complete understanding of the volume of lead released to the Australian continent from the consumption of leaded petrol. Atmospheric emissions of lead to the entire Australian continent from leaded petrol are calculated to total 240,510 tonnes over seven decades of use, peaking at 7869 tonnes in 1974. Total emissions for individual states and territories range from 1745 to 67,893 tonnes, with New South Wales responsible for the largest emissions. The effect of regulations on allowable concentrations of tetraethyl-lead additives are observed in the reduction of lead emissions in New South Wales and Victoria. The consequences to human health and the environment of leaded petrol consumption in Australia's populous cities are examined against historical air quality data and blood lead levels.

1. Introduction

The history of leaded petrol use has been described in detail (Nriagu, 1990; Needleman, 1998; Oudijk, 2010), from the discovery of lead (as tetramethyl- or tetraethyl-lead) as an antiknock agent, associated public health concerns, to the prohibition of its use in petrol. The use of leaded petrol resulted in the emission of large quantities of lead that are still present in the ambient environment, which may continue to cause concerns for health (Mielke et al., 2011). Although the United States was responsible for 80% of all leaded petrol sold globally prior to 1970 (Nriagu, 1990), Australia was a substantial consumer of lead petrol products. The use of leaded petrol in Australia over a 70 year period, from 1932 (Cook and Gale, 2005) to 2002 (Fuel Standard (Petrol) Determination, 2001), was a major contributor to atmospheric lead levels (Australian State of the Environment Committee, 2001; Department of the Environment and Heritage, 2004). Leaded petrol emissions accounted for up to 90% of atmospheric lead (Department of the Environment and Heritage, 2004) (where no other major lead industry was present) as 75% of lead in petrol was emitted from engine combustion (Australian Bureau of Statistics (ABS), 1997; U.S. EPA, 1986) (with a further 2% emitted to the atmosphere from fuel tank evaporation (Australian Bureau of Statistics (ABS), 1997).

As Australia's population grew, from 6.5 million in 1932, to 10 million in 1959, to just shy of 20 million in 2002 (Australian Bureau of Statistics (ABS), 2008), so did the demand for leaded petrol in Australia (Donnelly, 1981), resulting in elevated ambient lead

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http://dx.doi.org/10.1016/j.atmosenv.2015.04.012
1352-2310/

levels, particularly in metropolitan areas (Australian State of the Environment Committee, 2001; Abeeb et al., 2003). The adverse health effects of environmental lead have been thoroughly documented (Bellinger, 2008; Needleman, 2004; Rossi, 2008) along with the correlations of petrol-derived ambient lead levels and childhood blood lead levels in America (Annest et al., 1983; Lovei, 1998; Schwartz et al., 1985). With the introduction of unleaded petrol in Australia in 1985 (Australian Institute of Petroleum (AIP), 1986) and the subsequent phase out of leaded petrol by 2002 (Fuel Standard (Petrol) Determination, 2001), ambient lead levels have fallen in metropolitan urban centres to levels less than 10% of Australia's current guideline for lead in ambient air (0.5 $\mu g/m^3$) (Abeeb et al., 2003; National Environment Protection Council (NEPC), 1998).

Although leaded petrol has not been used in Australia in over a decade, environmental lead contamination persists. Understanding the history and extent of lead pollution is essential for evaluating long term environmental and human impacts (Cook and Gale, 2005). Studies quantifying emissions of petrol-derived lead in Australia are only available for limited years (Farrington and Australian Environment Council, 1988; Farrington et al., 1981) or locations (Abeeb et al., 2003; O'Connor et al., 1990; Gulson et al., 1983). The total volume of lead emitted into Australia's atmosphere from petrol consumption has never been determined despite multiple studies (Birch et al., 2011; Cooney et al., 1989; Cowie et al., 1997; Olszowy et al., 1995; Roberts et al., 1983) of the environmental and human health consequences. This study provides the first comprehensive compilation of Australian leaded petrol data, enabling calculation of the associated annual lead emissions. This new data allows consideration of the historical impact and future implications of leaded petrol consumption in Australia.

2. Methods and approach

Sales figures for leaded petrol from 1949 to 2002 were compiled from the Australian Bureau of Statistics (ABS) and Australian Institute of Petroleum (AIP) monthly bulletins and yearly reports. Sales figures are not available prior to 1949, instead ABS petrol import figures of petrol are used as surrogates because imports constituted the dominant proportion of petrol used in Australia until 1954 (Petroleum Information Bureau (Australia), 1960a). Sales and import figures for the years 1933–1957 are reported for the financial year (July 1st to June 30th) rather than the calendar year. Limited sales figures are also included in government reports (Berry et al., 1993; Cosgrove, June 2003; Trewin, 1997), but they do not always align with official sales figures reported by the AIP due to differing regional boundaries (Supplementary Fig. S1). Concentrations of tetraethyl-lead added to petrol have been reported by Associated Octel (Associated Octel, 1964; Associated Octel, 1968; Associated Octel, 1969; Associated Octel, 1975; Associated Octel, 1993; Octel, 1979), who was responsible for leaded petrol in Australia. Petrol regulations and actual average lead in petrol concentrations have been reported emission inventories and related studies (Nriagu, 1990; Farrington and Australian Environment Council, 1988; Farrington et al., 1981; O'Connor et al., 1990; Roberts et al., 1983; Trewin, 1997; Australian Institute of Petroleum, 1997; Australian Institute of Petroleum, 1998; Donovan, 1996).

Annual atmospheric lead emissions are calculated from leaded petrol sales data and the concentration of lead in each litre of petrol (data in Supplementary Tables S1 and S2) and the percentage of lead emitted from engine exhausts (Australian Bureau of Statistics (ABS), 1997; U.S. EPA, 1986; European Environmental Agency, 2013). Where the actual petrol lead concentration average is not known, the regulated maximum value of lead additives is used. Emissions from vehicles are calculated as follows:

$$E_{Pb/y} = Pb_{g/L} \times S_{L/y} \times 0.75$$

where $E_{Pb/y}$ is lead emissions from petrol per year, $Pb_{g/L}$ is the grams of lead per litre of leaded petrol, $S_{L/y}$ are the yearly sales of leaded petrol and 0.75 is the emission factor of lead emitted from petrol combustion (Department of the Environment and Heritage, 2004; Australian Bureau of Statistics (ABS), 1997; Brunekreef, 1984). Using this calculation method the correlation coefficient is 0.986 (Pearson correlation, $p < 0.00001$) between the calculated emissions of South Australia (SA) presented in this paper and those previously reported (Abeeb et al., 2003) ($n = 12$; largest available dataset), indicating the annual lead emission values calculated herein are robust and consistent with other studies.

3. Results and discussion

3.1. Leaded petrol sales

Sales figures (Supplementary Table S1) for total leaded petrol sales in Australia include both standard grade and super grade leaded petrol sales. Premium or “super” grade leaded petrol was introduced in June 1955 (Petroleum Information Bureau (Australia) and October, 1960b). Although sales figures are for states and territories, where available, sales figures for capital cities are provided. In New South Wales (NSW), Victoria (VIC), SA and Western Australia (WA), the sales figures for the capitals Sydney, Melbourne, Adelaide and Perth, are more than half the respective State annual total sales in 1976 (Donnelly, 1981; Farrington et al., 1981) and in 1985 (Australian Institute of Petroleum (AIP), 1986; Farrington and Australian Environment Council, 1988), with the exception of Sydney. For both these years, Queensland (QLD) capital, Brisbane, accounted for less than 50% QLD leaded petrol sales (Donnelly, 1981; Australian Institute of Petroleum (AIP), 1986; Farrington and Australian Environment Council, 1988; Farrington et al., 1981).

3.2. Lead petrol regulations

Initially, with the primary goal of achieving better fuel quality by increasing octane ratings, concentrations of lead in petrol were not regulated. However, it was not possible to raise the octane ratings indefinitely through the addition of greater quantities of tetraethyl-lead (Petroleum Information Bureau (Australia), 1959), with octane ratings plateauing at a lead concentration of 0.84 g/L (Michalski and Unzelman, 1980). As awareness that lead in petrol was a major environmental and health problem, the National Health and Medical Research Council recommended that yearly atmospheric lead emissions from petrol should not be permitted to rise above 1973 levels (National Energy Advisory Committee, 1978). Exhaust gases from leaded petrol combustion had a lead concentration of 75–80 mg/m^3 based on 0.84 g/L petrol lead concentration, which was four times the then statutory limit for stationary lead sources discharged at height (Australian Environment Council, 1977) (instead of ground level as in the case of leaded petrol consumption).

In Australia, regulatory power is vested with state and territory governments to introduce legislation relating to motor vehicles (Faiz et al., 1996; Environment Australia, 2000). As such, legislation for the reduction of lead concentration in petrol was introduced by a number of states, but not nationwide and not simultaneously. Changes in petrol lead concentration across the states and territories are summarised in Fig. 1. While these changes detail the maximum allowable lead concentrations, the actual lead