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Tracing changes in atmospheric sources of lead contamination using lead isotopic compositions in Australian red wine

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HIGHLIGHTS

- Red wine was investigated as a proxy for atmospheric lead conditions in Australia.
- Elevated lead concentrations in wine corresponded to peak lead emissions from petrol.
- Lead isotopic compositions confirmed the contribution of leaded petrol to wine.
- Strontium isotopes were used to measure provenance for future wine security.

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ABSTRACT

Air quality data detailing changes to atmospheric composition from Australia's leaded petrol consumption is spatially and temporally limited. In order to address this data gap, wine was investigated as a potential proxy for atmospheric lead conditions. Wine spanning sixty years was collected from two wine regions proximal to the South Australian capital city, Adelaide, and analysed for lead concentration and lead and strontium isotopic composition for source apportionment. Maximum wine lead concentrations (328 μg/L) occur prior to the lead-in-air monitoring in South Australia in the later 1970s. Wine lead concentrations mirror available lead-in-air measurements and show a declining trend reflecting parallel reductions in leaded petrol emissions. Lead from petrol dominated the lead in wine ($^{206}Pb/^{207}Pb$: 1.086; $^{208}Pb/^{207}Pb$: 2.360) until the introduction of unleaded petrol, which resulted in a shift in the wine lead isotopic composition closer to vineyard soil ($^{206}Pb/^{207}Pb$: 1.137; $^{208}Pb/^{207}Pb$: 2.421). Current mining activities or vinification processes appear to have no impact with recent wine samples containing less than 4 μg/L of lead. This study demonstrates wine can be used to chronicle changes in environmental lead emissions and is an effective proxy for atmospherically sourced depositions of lead in the absence of air quality data.

1. Introduction

Australia's lead mining history dates back to 1841 in South Australia (SA) (Drew, 2011) but was sporadic and ceased by the early 20th century as the deposits were generally of low tonnage. Mining operations in the region south of Adelaide, SA, including the ANGAS zinc-lead-silver mine, have seen a recent resurgence. Multiple smelting operations commenced with early mining, but it is the lead smelters in Port Pirie, SA, continuous since 1889, that have released the largest volumes of lead in SA (NPI, 2015b). In addition to emissions from mining and smelting activities, approximately 22,000 tonnes of lead were released from the combustion of leaded petrol in SA (Kristensen, 2015). Lead emissions from petrol consumption reached peak levels in the 1970s and declined from 1981 following regulation of the concentration of lead in fuel followed by the introduction of unleaded petrol in 1985 (Kristensen, 2015). Despite well-established prior knowledge of the adverse effects of lead toxicity (Needleman et al., 1979), monitoring and analysis of lead emissions into the Australian environment was limited. Outside of Port Pirie, availability of the lead-in-air data is limited to the state capital, Adelaide, and is only publicly available between 1982 and 2001 (Australian State of the Environment Committee, 2001).

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Lead emissions in Australia have inevitably altered the continent's atmospheric lead levels over the last 150 years, as has been the case elsewhere in the southern hemisphere (Bollhöfer and Rosman, 2000). Given the deficiency of lead-in-air measurements, trends in long term air quality have not been captured, and monitoring commenced after peak emissions from leaded fuels. Although current mining operations in SA are small, there is a long history of mining and smelting. With the established link between lead-in-air concentrations and blood lead levels (Annest et al., 1983), particularly from leaded petrol and mining and smelting operations (Thomas et al., 1999; Hilts, 2003; Taylor et al., 2014), a complete analysis of lead-in-air would assist in evaluating the long term health effects from lead emissions in Australia (see Taylor et al., 2016).

Environmental proxies have demonstrable potential forproviding substitute data for assessing historic anthropogenic lead emissions to Australia's atmosphere. Wine is a rarely used proxy but its seasonal production and bottling has obvious potential for re-tracing emissions and depositions over the recent industrial past. Lead in wine is sourced in part by plant uptake from soil lead but also from anthropogenic sources (Almeida and Vasconcelos, 2003). Temporal analysis of European wines has shown that the pattern of lead concentration in wines follows the consumption of leaded petrol in Europe (Lobiński et al., 1994; Rosman et al., 1998; Médina et al., 2000; Mihaljevič et al., 2006).

Lead isotopic analysis of wines from Bordeaux, France showed that lead in the wines changed over time to reflect the dominant source of atmospheric lead pollution in southern France (Médina et al., 2000). Other European studies have found that lead isotopic compositions in wine may not always reflect those of leaded petrol, but reflect the isotopic signature of local, dominant metallurgical industries (Larcher et al., 2003; Mihaljevič et al., 2006). These studies confirm atmospheric deposition as being the dominant contributor to the lead content and isotopic composition of wine. Some studies have shown that contamination from tin-lead foil capsules in the presence of corrosion and cork disintegration can dominate the source of lead in wine (Gulson et al., 1992). Other studies have attributed the lead in wine to machinery or additives used during the vinification process where environmental contamination is a minor source of lead (Almeida and Vasconcelos, 2003; Stockley et al., 2003).

Given the paucity of air quality data in Australia, this study evaluates whether historic wine samples can be used to provide reliable surrogate information. In order to determine whether age-dated wine can be used as a proxy for atmospheric emissions, lead isotopic composition of wines and local vineyard soil were measured to apportion atmospheric lead depositions to their anthropogenic origins. Wine-derived lead concentration and isotopic data was also evaluated against leaded petrol emissions data for SA (Kristensen, 2015) and the isotopic compositions of available archive air filters from South Australia Environment Protection Authority (SAEPA). Strontium isotopes have also been employed to provenance wine, sometimes in conjunction with lead isotopes (Balcaen et al., 2010). In contrast to industrial emissions of lead, there are no known anthropogenic sources of strontium in wine, indicating the potential to use strontium isotopes to establish the influence of natural soil on wine composition, including that of lead.

2. Methods and materials

2.1. Field methods

Wine samples were collected from wineries established in the viticulture regions of McLaren Vale and Langhorne Creek in SA (Fig. 1) spanning the years 1963–2012 (n = 60; Supplementary Table S1). To limit potential confounding factors, wine samples were all of a red variety (Shiraz or Cabernet Sauvignon), produced from known vineyards within the same wine region, and were free from blending with wine from other regions. Soil samples (n = 12; Supplementary Table S2) were collected from vineyards that provided wine samples for the study. Surface soils (0–2 cm) were collected to characterise the effect of atmospheric depositions and sub-surface samples (below 20 cm dependant on bedrock depth) were taken to characterise background soil concentrations. Soil samples were sieved to <2 mm to remove rocks and course particulates.

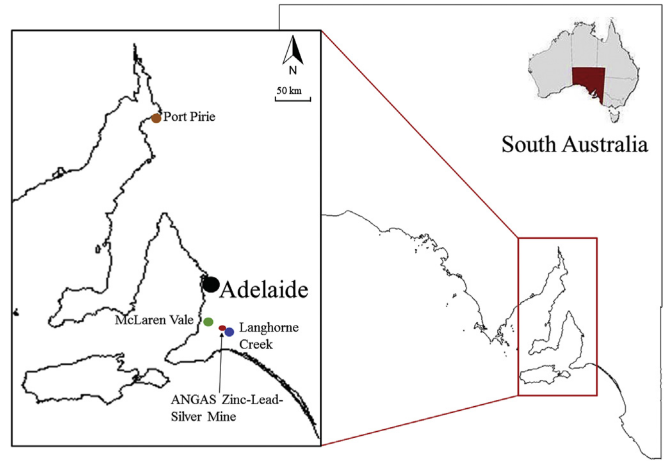

Fig. 1. Locations of McLaren Vale and Langhorne Creek wine regions in relation to the main sources of lead emissions in Adelaide (petrol) and Port Pirie (lead smelter) (map drawn by authors).

Air filter papers were collected from SAEPA who had sampled in central Adelaide city (Parkside: April 1997–June 2003). The city air filters provide a measure of aerosol composition during the later years of leaded petrol emissions, while those from an area in north Adelaide (Osborne: January 2002–December 2004), represent local industrial emission sources (Fig. 1). No filter papers were available prior to 1997. Given the fact that sampled wine regions are located either side of ANGAS zinc-lead-silver mine (Fig. 1), ore samples were sourced directly from the mine in 2012 to characterise this contemporary source.

2.2. Laboratory methods

Wine, soil and air filter samples were subject to acid extractable digestion and measurement at the National Measurement Institute, North Ryde, Sydney. Wine samples (5 mL) were digested on a hot block with 16 M HNO_3 (3 mL) after evaporating the alcohol. In order to test the homogeneity of a wine bottle, six separate 5 mL samples were taken from one bottle and analysed, returning a lead concentration of 1.10 μg/L (SD 0.13; n = 6). The homogeneity in sampling wine for lead was evaluated by replicate analysis (n = 10) of multiple bottles of the same wine from a single vineyard in the same year (2011), which returned a lead concentration of 1.13 μg/L (SD 0.11). Soil samples (1 g) and air filter samples were digested in concentrated HCl (3 mL) and HNO_3 (3 mL). Lead concentrations and isotopic compositions were determined on a Q-ICP-MS (PerkinElmer ELAN DRCII). Matrix spikes of wine samples returned recoveries of 82–84% for wine samples and a correction factor applied. Replicate analysis returned RSDs <4.7% for wine samples and <5.0% for soil samples. Soil sample matrix recoveries were 85–90% with recoveries of internal reference materials AGAL-10 (Hawkesbury River Sediment) 100% and AGAL-12 (biosoil) 99%.