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Chemical characterisation and source identification of atmospheric aerosols in the Snowy Mountains, south-eastern Australia

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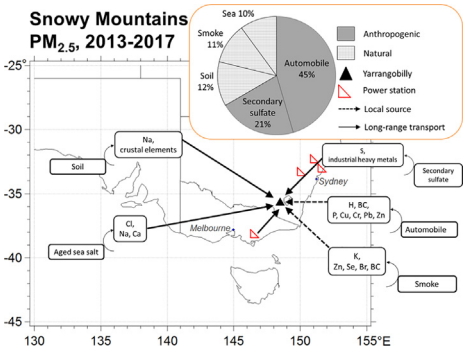
HIGHLIGHTS

- First PM2.5 dataset (2013–2017) for the Snowy Mountains alpine region, SE Australia
- Sources of PM2.5: automobile, secondary sulfate, smoke, soil and aged sea salt
- Long-range transport of Na and aeolian dust impacts this remote inland site.
- Drought and El Niño conditions enhanced smoke and soil aerosol loadings.

GRAPHICAL ABSTRACT



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ABSTRACT

Characterisation of atmospheric aerosols is of major importance for: climate, the hydrological cycle, human health and policymaking, biogeochemical and palaeo-climatological studies. In this study, the chemical composition and source apportionment of $PM_{2.5}$ (particulate matter with aerodynamic diameters less than 2.5 μm) at Yarrangobilly, in the Snowy Mountains, SE Australia are examined and quantified. A new aerosol monitoring network was deployed in June 2013 and aerosol samples collected during the period July 2013 to July 2017 were analysed for 22 trace elements and black carbon by ion beam analysis techniques. Positive matrix factorisation and back trajectory analysis and trajectory clustering methods were employed for source apportionment and to isolate source areas and air mass travel pathways, respectively. This study identified the mean atmospheric $PM_{2.5}$ mass concentration for the study period was (3.3 ± 2.5) μg m^{-3}. It is shown that automobile (44.9 ± 0.8)%, secondary sulfate (21.4 ± 0.9)%, smoke (12.3 ± 0.6)%, soil (11.3 ± 0.5)% and aged sea salt (10.1 ± 0.4)% were the five $PM_{2.5}$ source types, each with its own distinctive trends. The automobile and smoke sources were ascribed to a significant local influence from the road network and bushfire and hazard reduction burns, respectively. Long-range transport are the dominant sources for secondary sulfate from coal-fired power stations, windblown soil from the inland saline regions of the Lake Eyre and Murray-Darling Basins, and aged sea salt from the Southern Ocean to the remote alpine study site. The impact of recent climate change was recognised, as elevated smoke and windblown soil events correlated with drought and El Niño periods. Finally, the overall

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implications including potential aerosol derived proxies for interpreting palaeo-archives are discussed. To our knowledge, this is the first long-term detailed temporal and spatial characterisation of $PM_{2.5}$ aerosols for the region and provides a crucial dataset for a range of multidisciplinary research.

1. Introduction

The main aerosol sources which influence the atmosphere of the Australian continent include windblown soil-dust, sea-salt, biomass burning, and biogenic secondary organic aerosols from volatile organic aerosols (Rotstayn et al., 2009). Aerosols are solid and/or liquid particles suspended in air (Pöschl, 2005). These particles may be emitted directly into the atmosphere from i) natural sources, such as: biogenic volatile organic compounds emitted from vegetation, sea-salt, soil-dust, volcanoes, biomass burning; or ii) anthropogenic sources, such as: coal-burning, metal smelting, vehicle emissions (Colbeck and Lazaridis, 2014). Once in the atmosphere, the aerosols are transported downwind of their source area, during which time chemical reactions can occur, resulting in the formation of secondary aerosols from precursor gasses such as SO_2 and NO_x (e.g. Squizzato et al., 2013; Suni et al., 2008). Thus aerosols contain chemically distinct concentrations of trace metals, characteristic of their supply source, air mass history and geographical proximity to coastal, desert, rural, industrial and polar emission sources.

Aerosols suspended in the atmosphere are known to impact the climate system. Aerosol particles have a direct radiative forcing effect on the Earth's surface and atmosphere, because they can absorb or scatter incoming solar radiation. When solar radiation is reflected back to space, a smaller amount of solar energy reaches the ground and this has a cooling effect on the regional and global climate, whereas some aerosol particles can absorb solar radiation and this warms the atmospheric layer. Aerosols also have indirect radiative forcing effects on the climate through modifying the formation and microphysical cloud properties, leading to a cooler climate. Acting as condensation nuclei, soluble aerosols can increase the condensation moisture in clouds by increasing the droplet number concentration and therefore cloud albedo, or the reflection of solar radiation to space. Aerosols also decrease the droplet size in clouds, suppressing precipitation (and affecting the hydrological cycle), which may also lead to an increase in cloudiness and reflection of solar radiation. These direct and indirect aerosol effects on the climate are determined by aerosol particle size, structure and chemical composition (Pöschl, 2005).

Air quality is the most visible effect of aerosols in the environment, affecting visibility, aviation and road traffic safety as well as human health. Fine particulate matter with an aerodynamic diameter smaller than 2.5 μg ($PM_{2.5}$) can foster acute and chronic diseases (Pope III et al., 2009), hence to reduce air pollution, policy interventions, targeting concentration limits and air quality guidelines, are established based on monitoring data (WHO, 2006). Atmospheric deposition of aerosols is also of specific interest in ecosystem biogeochemistry (Mahowald, 2011). Aerosols provide trace metals that are essential for productivity in terrestrial (Chadwick et al., 1999) and marine ecosystems (Jickells, 1995). Alternatively they can have a negative impact, for example acid rain, due to atmospheric sulfate or nitrate deposition, by enhancing the leaching of nutrients from land ecosystems (Likens, 2010) or results in ocean acidification in coastal regions (Doney et al., 2007). Depositing aerosols are also known to alter the physical and chemical properties of soils and sediments (Simonson, 1995; Hesse and McTainsh, 2003; Rutlidge et al., 2014). In addition the deposition of trace metals from aerosols in natural archives is important in palaeoclimate studies (Sigl et al., 2015). The concentration of aerosols deposited in soils, sediments, peat, speleothems and glaciers have been used as palaeo proxy indicators of climate and environmental changes (Rea, 1994; Kohfeld and Harrison, 2001; Frisia et al., 2005; Frappier, 2006; Marx et al., 2011; Allan et al., 2015; Ridley et al., 2015). Thus an understanding of the composition, source and transport of atmospheric aerosols are highly relevant for studies in various disciplines including climate processes, human health and policies, terrestrial and marine biogeochemical cycling and palaeo-climatology.

Our study site is located in Kosciuszko National Park, a high elevation-alpine site in mainland Australia (1059 m above sea level). The location of the study site is a sensitive monitor for past hydroclimate and environmental change as it is a region prone to ash input from fire activity and wind-blown dust deposition, as it is located within the south-eastern pathway of dust transport from active emission sources in central Australia (Shao et al., 2011). The site is also strategically unique in Australia, being a remote inland site; it is conducive to study the impact of long-range atmospheric transport from regional and natural emission sources.

The impetus for the current study is to define the chemical characteristics and input sources and gain a better understanding of transport processes of atmospheric particulate matter to karst in the Snowy Mountains alpine region, Yarrangobilly, SE Australia. This study was conducted as part of a wider project to reconstruct past environmental change from cave deposits (speleothems) in order to better understand past variability in climate, fire history and environmental change in the Snowy Mountains region. Trace elements are one of the commonly used proxies for providing information on the hydro-climate regime surrounding the depositional conditions in sediments and carbonate rock archives. As atmospheric aerosols are a direct source of trace elements, constraining the sources of trace elements may be highly relevant for the reconstruction of past environmental change from speleothems. Although the atmosphere may supply a significant source of elements to the soil and hence drip-waters, less attention has been placed on quantifying the atmospheric input from various sources above the cave. Dredge et al., 2013 highlighted that aerosols brought into the cave by air currents are a potential source of elements for speleothem deposition, however neither the source of aerosols nor the atmospheric processes associated with them were investigated. Hence there is a current knowledge gap in our understanding of the holistic role of atmospheric input, particularly aerosols, as a potential source of elements that have been transported in the cave via infiltrating drip-water and incorporated into speleothem calcite.

This paper presents the first high resolution four-year $PM_{2.5}$ dataset for the region. Ion beam analysis (Cohen et al., 1996) was used to identify the elemental composition of the $PM_{2.5}$ samples, following which positive matrix factorisation (PMF; Paatero and Tapper, 1994) was applied to identify the contributing sources. Additionally, long-range source contribution regions to this remote inland site were identified using backward air mass trajectory calculations. To compliment the study, results were also compared to one of the global "baseline" atmospheric measurement sites at Cape Grim in Tasmania Australia, a remote coastal site that receives airflow predominately from the Southern Ocean and therefore with little impact from regional anthropogenic influences. The current data set is important for achieving a better understanding of the composition of atmospheric aerosols from natural and anthropogenic origin and from local and long-range transported sources in the Snowy Mountains. In essence, this work will provide a benchmark to understand the local and regional dynamics of aerosol sources, providing the most extensive aerosol baseline dataset for application in other studies regionally.