Atmospheric Environment 127 (2016) 107–117

Contents lists available at ScienceDirect

Atmospheric Environment

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

Assessing the impact of atmospheric stability on locally and remotely sourced aerosols at Richmond, Australia, using Radon-222

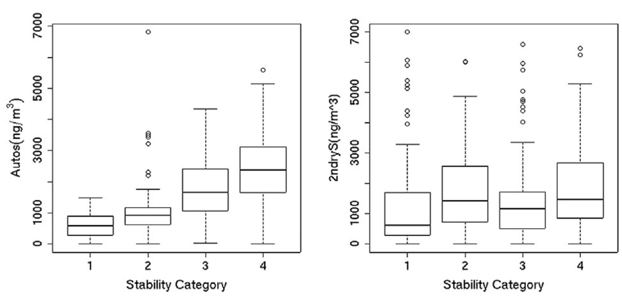
Jagoda Crawford*, Scott Chambers, David Cohen, Alastair Williams, Alan Griffiths, Eduard Stelcer

Australian Nuclear Science and Technology Organisation, Locked Bag 2001 Kirrawee DC, New South Wales, 2232, Australia

HIGHLIGHTS

- Dominant source fingerprints identified for $PM_{2.5}$ at Richmond, Australia.
- A radon-based stability classification scheme modified for 24-h PM data.
- Local sources more strongly affected by stability than secondary sources.
- Local source concentrations, 4 to 12 times higher under stable conditions.
- Pasquill–Gifford stability typing underestimated concentrations by factor of 2.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 19 August 2015
Received in revised form
9 December 2015
Accepted 10 December 2015
Available online 13 December 2015

Keywords:
$PM_{2.5}$
Radon
Atmospheric stability
Local aerosols
Secondary aerosols

ABSTRACT

A flexible radon-based scheme for the classification of nocturnal stability regimes was used for the interpretation of daily-integrated $PM_{2.5}$ aerosol observations collected at Richmond, Australia, between 2007 and 2011. Source fingerprint concentrations for the dominant locally and remotely sourced aerosols were analysed by nocturnal radon stability category to characterise the influences of day-to-day changes in daily integrated atmospheric mixing. The fingerprints analysed included: smoke, vehicle exhaust, secondary sulfate and aged industrial sulfur. The largest and most consistent stability influences were observed on the locally sourced pollutants. Based on a 5-year composite, daily integrated concentrations of smoke were almost a factor of 7 higher when nocturnal conditions were classed as “stable” than when they were “near neutral”. For vehicle emissions a factor of 4 was seen. However, when the winter months were considered in isolation, it was found that these factors increased to 11.5 (smoke) and 5.5 (vehicle emissions) for daily average concentrations. The changes in concentration of the remotely sourced pollutants with atmospheric stability were comparatively small and less consistent, probably as a result of the nocturnal inversion frequently isolating near-surface observations from non-local sources at night. A similar classification was performed using the commonly-adopted Pasquill–Gifford (PG) stability typing technique based on meteorological parameters. While concentrations of fingerprints associated with locally-sourced pollutants were also shown to be positively correlated with atmospheric stability using the PG classification, this technique was found to underestimate peak pollutant concentrations under stable atmospheric conditions by almost a factor of 2.

* Corresponding author.
E-mail address: Jagoda.Crawford@ansto.gov.au (J. Crawford).

http://dx.doi.org/10.1016/j.atmosenv.2015.12.034
1352-2310/

1. Introduction

It is well established that natural and anthropogenic aerosols can affect human health (e.g. Moloi et al., 2002; Russell and Brunekreef, 2009; Dockery, 2009; Lu et al., 2015). As a result regulatory guidelines have been imposed in many urban centres for various anthropogenic emissions (e.g. VES, 2012), as well as maximum concentrations to which the general public may be exposed on a daily or annual basis (USEPA, 2007; NSW, 2011). Consequently, with the increasing global population, and related industrial and power needs, there are growing needs to characterise the range of pollution events to which the public might be exposed, with particular emphasis on extreme or exceedance events from routine releases.

As in many large cities, in Sydney during winter domestic heating emissions combine with exhaust from peak-hour traffic in the shallow morning inversion layer, resulting in "brown haze" and pollutant levels that can exceed threshold guidelines (e.g. Duc et al., 2013; Hinkley et al., 2008; Gupta et al., 2007; Corbyn, 2004; Leighton and Spark, 1997; Liu et al., 1996). In summer photochemical pollution events are more common; their severity linked to prevailing winds and cloudiness (Hart et al., 2006; Leslie and Speer, 2004). Mitigation strategies therefore require knowledge of the source types leading to high pollution events.

When the chemical composition of atmospheric particulate matter (PM) samples is determined a range of known source types, or "fingerprints", can be identified using multivariate methods, such as Principle Component Analysis (PCA; Jollife, 1986), Positive Matrix Factorisation (PMF; Paatero and Tapper, 1994) and UNMIX (Henry, 2002). Results from "fingerprint" analyses can thus be used to help apportion PM measurements to various emission sources in the vicinity (Cohen et al., 2014; Crawford et al., 2013). In addition to their emission rates, PM concentrations within the atmospheric boundary layer (ABL) are also affected by chemical transformations, dispersion and deposition, and finally by the characteristics of the atmospheric volume into which they mix (e.g. Veleva et al., 2010; Perrino et al., 2001, 2008; Avino et al., 2003, 2015).

Over land, the atmospheric mixing depth changes daily, typically reaching maximum values early in the afternoon and minimum values prior to sunrise (Stull, 1988). As vertical dispersion is shallow when the atmosphere is thermally stably stratified at night, stable conditions have been linked to pollution exceedance episodes (e.g. Grange et al., 2013; Ji et al., 2012; Desideri et al., 2006; Essa et al., 2006; Avino et al., 2003). Numerous measures of atmospheric stability (or degree of vertical mixing) have been devised and applied with varying degrees of efficacy (USEPA, 2007). Knowledge regarding the seasonality in frequency of stable conditions, and the range of concentrations expected on the most stable days, is crucial for decision makers when considering the need for emission mitigation strategies. Furthermore, the efficacy of mitigation strategies is best evaluated after the state of the atmosphere has been classified by stability regime, since the fraction of time for which the different atmospheric conditions dominate can vary from year to year.

Chambers et al. (2015a,b) developed an atmospheric stability classification scheme based on hourly surface measurements of the terrestrially-ubiquitous noble gas Radon-222 (radon), and compared it to the commonly used Pasquill–Gifford (PG) stability typing method based on routinely available meteorological observations (USEPA, 2007). They found that the radon-based scheme generally performed better than the PG scheme; in particular, the most stable category of the PG scheme was less selective of the strongly stable nights than the radon-based scheme. To apply either of these stability classification schemes directly to particulate measurements, however, is non-trivial, as $PM_{2.5}$ measurements for source fingerprint identification are generally available only on a 24hr-integrated midnight-to-midnight basis (e.g. Begum et al., 2005; Zhou et al., 2009), whereas the stability classification schemes are generally best suited to daily-resolved (hourly) measurements.

In this study, the impact of the atmospheric stability on the concentrations of dominant source fingerprints (separated into locally and remotely sourced aerosols) is analysed using $PM_{2.5}$ measurements at Richmond (Sydney), Australia. The radon-based stability classification scheme of Chambers at al., 2015a,b is adapted for application to the 24-h integrated aerosol data, and mean concentrations of each source type are then evaluated by stability category in order to characterise the effect of changing atmospheric stability. Finally, the results of this analysis are compared against a similar analysis performed using the PG stability classification scheme.

2. Study site and methods

2.1. Site and study domain

Richmond (33.618°S, 150.748°E; 24 m above sea level) is ~50 km northwest of Sydney (5–10 km east of the Blue Mountains, and 50–55 km west of the coast). In the immediate vicinity, topography is relatively flat. Apart from Sydney, other potentially significant sources of primary and secondary $PM_{2.5}$ include 8 coal-fired power stations that serve the greater Sydney region (Fig. 1).

Most measurements, including hourly climatological and air quality observations (courtesy of New South Wales Office of Environment and Heritage, NSW OEH), hourly radon observations and daily-integrated $PM_{2.5}$ sampling, were conducted at the University of Western Sydney, Richmond Campus. Traffic density was observed on the Richmond bridge, 4.5 km to the northwest.

Results presented pertain to the 5-year period Jan-2007 to Dec-

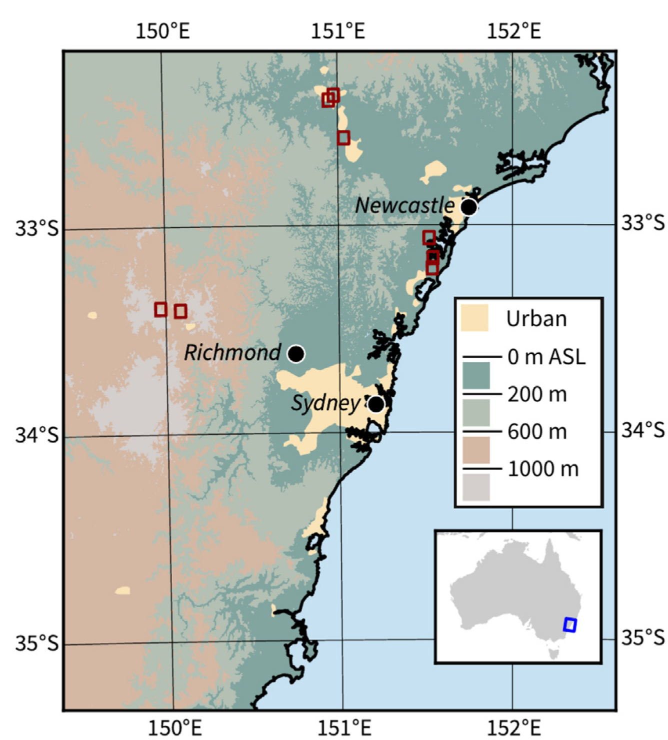


Fig. 1. Location of measurement site (Richmond, NSW), with respect to nearby urban centres (Sydney, Newcastle) and eight power stations (open boxes).