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# Characterisation of the major dust storm that traversed over eastern Australia in September 2009; a multidisciplinary approach



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# ABSTRACT

In September 2009, a major dust storm passed over the southeast coast of Australia and covered the large city of Sydney that is located on the eastern coast of Australia. It was referred to as the 'Red Dawn' event. Along its course across the state of New South Wales, the dust plume passed over Canberra, the capital of Australia located some 300 km from Sydney. In this study we identified the sources of the dust and tracked the progression of the dust storm using satellite imagery. We also examined the meteorological conditions that led to the formation of the dust plume. We also investigated the microbial and lipid composition, grain-size distribution, pollen content and geochemical composition of several samples of the dust that fell in Canberra (called 'Canberra dust'), with particular attention paid to the ratio of some rare earth elements as well as strontium and neodymium isotopes. This was done to identify a geochemical and palynological 'fingerprint' of this dust to determine the source of the material. Collectively, the meteorological and fingerprinting analyses identified the principal location of dust entrainment as between the large playa Lakes Gairdner and Torrens in arid South Australia, some 1600 km away from Canberra. We also determined through Sr isotope analyses and other elements measured in the dust that fell in Canberra and the township of Eden, located south of Sydney, that the dust changed composition slightly over a few hours, being the consequence of local dust entrainment along the path of the dust plume across South Australia and New South Wales. In addition, we present a scenario which explains dust transport away from the direction of the major dust plume. Those investigations carried out on the Canberra dust are compared with a previously documented event when dust was also sampled in Canberra in 2002. The origin and composition of the 2002 dust plume was clearly different from that which occurred in 2009, as demonstrated by the different chemical (organic and inorganic) composition, grain-size characteristics and palynological fingerprints, as well as microbial composition.

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# 1. Introduction

Airborne dust is frequently generated in arid and semi-arid regions, but can travel huge distances over landmasses and also reaching oceans. With an estimated budget of 2000 million tonnes

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(Shao et al., 2011), dust is becoming more prevalent and is causing grave concerns under the scenario of climatic change as well as the alteration of the landscape by human-induced practices, such as land clearance, massive fires and the imposition of vast numbers of grazing animals, (Griffin and Kellogg, 2001, 2004). It has even been suggested that the planet may be returning to conditions of a 'dusty world' similar to those experienced during the last glacial period (Kohfeld and Harrison, 2001). Dust has been shown to have possible, direct detrimental effects on human health, such as asthma in the Caribbean (Gyan et al., 2005) and Valley Fever in California (Schneider et al., 1997), or indirectly as a carrier of disease

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to coral reefs (Shinn and Griffin, 2000; Griffin et al., 2001; Griffin and Kellogg, 2001, 2004; Kellogg and Griffin, 2006), or contributing to high mercury concentrations in lakes in Florida after travelling from West Africa (Holmes, 2003). In Australia, an extensive, decade-long drought during the first decade of the new millennium culminated in several major dust storms travelling as far as New Zealand (Leys et al., 2011, and references therein) and New Caledonia, at times passing over the Great Barrier Reef offshore north east Australia.

These impacts of dust on the atmosphere, biosphere, oceans and human health have brought about targeted efforts by the scientific community to trace dust back to its origins to better understand the relationship between the sources of this dust and the impacted sinks. Many disciplines have contributed to these efforts, such as remote sensing (e.g., Miller, 2003; El-Askary et al., 2006; Baddock et al., 2009; Ginoux et al., 2012), modelling (e.g., Draxler et al., 1994; Draxler and Hess, 1998; Koven and Fung, 2008) and trace element analysis (e.g., Kamber, 2009; De Deckker et al., 2010; Marx et al., 2014) for example. Few studies however have combined all of these disciplines, or innovated by trying less conventional ways of tracing dust storms (e.g., pollen, microbes). De Deckker et al. (2008) conducted one of the first geochemical and microbiological fingerprinting studies on dust collected in Canberra, Australia from a large dust storm that occurred in October 2002. Lim et al. (2011) conducted a microbiological study of Australian dust that contained a microflora sampled both on dust-free



Fig. 1. Map of eastern Australia showing the location of many sites mentioned in the text. The brown shading delineates the regions of high altitude.

days and dusty days. More recently, Munday et al. (2013) examined the microorganisms found in samples from a dry saline lake floor at Lake Gnarpurt in western Victoria, Australia (Fig. 1), and compared them with aerosols collected in air filters at different heights above the lake floor. Several studies by Evans et al. (2006, 2008, 2009) report on the microbial composition of rain samples obtained in rainwater tanks located in the city of Newcastle, some 150 km north of Sydney.

To better understand the role of dust in the environment, both at regional and global scale, it is therefore critical to compile the characteristics, composition and impacts of dust plumes; from their origins and trajectories, to their impact on the atmospheric, terrestrial and aquatic environments, as well as human health. A crucial element of this process is to understand the characteristics of sources and this begins with the identification of where these sources are located. Remote sensing can show what geomorphological features in source regions contribute to large dust plumes, but by the time such plumes reach monitored or impacted sinks downwind, it is no longer clear where the dust originated from.

On 23 September, 2009, Sydney woke up to what is now widely referred to as the "Red Dawn" dust storm. The city was blanketed by a thick layer of dust that had been picked up approximately 1000-1500 km to the west of Sydney the previous day and had made the journey to the east coast of Australia overnight. Although this dust storm event received worldwide media coverage, it was part of a longer series of dust entrainment and deposition events for the Australian continent that lasted approximately one week from 21-28 September, 2009. A special combination of events is required to provide the conditions suitable for large-scale dust entrainment events. The year 2009 came at the end of one of the worst droughts of the Australian historical record, the so-called Millennium Drought (Van Dijk et al., 2013). This left highly erodible inland regions with little or no protective vegetation cover, making them vulnerable to strong frontal winds. These strong winds, with sufficient surface friction velocities and unstable atmospheric conditions, can be generated by the intense front-trough systems that are characteristic of the austral spring in south-central Australia (Smith et al., 1995; Sturman and Tapper, 2006). These systems also provide a mechanism by which dust can be transported towards the southeast, the northeast and even to the northwest in the anticyclonic flow behind the trough-front system.

In this study, we combine a unique dust storm forecasting routine with remote sensing and meteorological analyses to establish the chronology of this large dust storm. We then present the analyses from the fields of geochemistry, sedimentology, biochemistry, microbiology and palynology on Red Dawn dust samples collected in Canberra and coastal New South Wales. This is done to provide the scientific community with a benchmark study on multi-disciplinary dust fingerprinting and source identification. Some of these approaches are well known and published widely while some, such as organic chemistry or microbiology, have rarely or never been used before to fingerprint dust.

# 2. Methods

#### 2.1. Prediction of dust storm events

Due to the time-sensitive nature of sampling and the cost of air/ dust filter analysis – especially for its microbial content - it is vital to develop a reliable dust event prediction routine to prepare equipment and maximise the chances of collecting dust during the sampling runs. To that end, a number of datasets were combined to (a) predict the occurrence of dust events, and (b) determine in advance the trajectories of the dust plumes associated with these events. To identify the onset of dust storms, we used the Navy Aerosol Analysis and Prediction System (NAAPS)<sup>3</sup> model, developed by the Navy Research Laboratory (NRL) in Monterey, California. The meteorological fields for the model are derived from the Navy Operational Global Atmospheric Prediction System (NOGAPS) (Hogan and Rosmond, 1991; Hogan and Brody, 1993). The dust prediction model takes into account wind speed and entrainment threshold wind velocity, surface moisture and particle size. The NAAPS model dust source layer is however limited and is restricted to the dustier regions of the Lake Eyre Basin as defined by long-term Total Ozone Mapping Spectrometer (TOMS) imagery. We therefore derived our knowledge on sources from detailed satellite mapping studies by Bullard et al. (2008) and O'Loingsigh (2009).

By combining this detailed source knowledge and the NAPPS model prediction of dust storm start times, we used the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) Model to forecast the trajectories of these dust storms up to three days in advance. If any of the forecast dust plumes passed over our Melbourne or Canberra sampling sites, we organised to have all microbiological and geochemical samplers operational for a 24-h period corresponding to the predicted passage of the dust plume at those sites.

We found this multi-step prediction routine to be quite effective at predicting dust events and we were able to predict all moderate to severe dust storms in eastern Australia for the period 2007–2009, including the Red Dawn dust storm.

#### 2.2. Satellite imagery

We used satellite imagery to track the development of dust storms and to identify the location of their sources. On the day of the predicted events, we looked for plumes in near-real time imagery from the Moderate Resolution Imaging Spectroradiometer (MODIS) sensor on board NASA's Terra and Agua satellites. Day-time images (overpasses at approx. 10:00 and 14:00 AEST) were processed to produce True Colour Composite images (from MODIS reflective channels) and monochromatic Brightness Temperature Difference (BTD) images between MODIS bands  $32(12 \mu m)$  and 31(11 µm) (Miller, 2003). Night-time images (overpasses at approx. 23:00 and 02:00 AEST) were also processed using the latter technique only. In a study on Lake Eyre Basin dust sources for the period 2004–2007, O'Loingsigh (2009) conducted a visual analysis of >2000 true-colour MODIS images. This familiarity with the eastern half of the Australian continent on dusty and non-dusty days, enabled us to clearly identify plumes by their colour and linearity in the daytime colour imagery. We found that night-time thermal BTD images were more difficult to interpret. The cold air of a front is relatively easy to discern over the warn skin of the earth, highlighting the position of the front from space. Dust entrained by that front takes the shape of the front and it therefore becomes difficult to differentiate cold air from dust. Once the front has passed over a source however, westerly winds continue to feed dust into the front, producing plumes that seem to 'chase' the front. These chasing plumes are not a feature of cold air, but of dust sources, and as such were found to be useful as a tell-tale sign that the front itself was dusty.

# 2.3. Sample collection

We conducted several types of analyses on dust collected in Canberra and also in Eden, New South Wales (NSW) on the east coast of Australia (Fig. 1). Table 1 summarises the collection sites and the analysis we carried out.

<sup>&</sup>lt;sup>3</sup> http://www.nrlmry.navy.mil/aerosol\_web/Docs/globaer\_model.html

Samples for microbiological analysis were characterised using both culture and DNA-based approaches. The samples were collected using pre-sterilised filter housings containing 47 mm diameter cellulose nitrate filters with a pore size of  $0.22 \,\mu m$  (Nalgene, Rochester, NY, USA, Cat# 145-0020). The equipment was installed at Mount Stromlo close to the location of the Australia National University's astronomical observatory at an isolated site away from most anthropological activities, approximately 12 km west south west of the Canberra city business centre, and 15 km from Aranda (Table 1). For further details on the equipment refer to Lim et al. (2011). Each filter was attached to an AirChek XR5000 hand held vacuum pump (SKC inc, Eighty Four, PA) pre-calibrated to a flow rate of 2.3 L/min. Samples were collected over two different periods on 22-23 September 2009 - the first was from 12:00 to 17:00 AEST, when visibility was lowest and before any rain had fallen. The second set of filters was deployed at 17:00 AEST and left out overnight, during the rain event. Two filters were used during each collection period - one for culture analysis and one for direct DNA extraction. To collect rain, a sterilised 18 cm polyethylene funnel was attached to the mouth of a sterile 500 ml Schott bottle, mounted 2 m above the ground and left out overnight. Once collected, all samples were transported back to the laboratory and stored at 4 °C for a maximum of 24 h prior to further processing in the case of culture-based samples, or stored at -80 °C for DNA based analysis.

Sedimentological, pollen and lipids analyses were conducted on rained-out dust collected in a series of collection bowls deployed at regular 2–3 hourly intervals from 19:00 AEST on 22 September onwards in Aranda (Table 1) a suburb close to the Canberra central business district (Fig. 2). Once deployed, bowls were left untouched until all were collected at the same time in the morning of 23 September when rain had stopped. The 04:00 AEST bowl (not shown) was the last to capture sediment. By 06:00 AEST the rainfall samples were clear.

After the event, most of the water was decanted from the bowls, and these were then left to rapidly dry indoors before the dust component could be stored prior to analysis. For pollen and lipid extraction, sediment from the bowl that was placed at 23:00 AEST was used. For grainsize analysis, sediment from the bowl placed at 19:00 AEST was analysed.

Inorganic geochemical analyses were carried out on samples from the 19:00 AEST on 22 September and the 04:00 AEST the following day as well as site further downwind in the coastal town of Eden, NSW. This was done to provide insights into the changing composition of dust plumes (if any) as they pass over a fixed sampling site.

Our investigations benefitted from inland reference point data (Figs. 4 and 5) based on previous studies and a series of sampling seasons carried out in various parts of Australia during which we collected samples adjacent to large salt lakes and dunes, believed to be the source of airborne material. These inland sampling sites had been carefully identified using satellite imagery on which dust-enhancement algorithms were applied (Miller, 2003) for all major dust storms of the 2000s. Thus, we visited the Strzelecki Desert which includes lakes Frome, Blanche and Callabonna as part

of a chain of lakes that fed into Lake Eyre during the last interglacial (see Cohen et al., 2011) and where a predominance of Pleistocene and Pliocene non-marine surficial sediments abound. We also visited the Eyre Peninsula, with its southern end consisting of marine/non-marine Quaternary and Tertiary sediments, and further north, south, and west of the Flinders Ranges which consist of Archean up to Mesozoic lithologies. These samples were added to the survey of the Murray Darling Basin rivers carried out by Gingele and De Deckker (2005) and Gingele et al. (2007) and further elaborated by De Deckker et al. (2010) and De Deckker and Norman (2010).

# 2.4. Microbiology: sample preparation

The microbial communities within the dust storm samples were characterised using both culture and DNA-based approaches.

Three different culture media were utilised for the cultivation of organisms – R2A (Difco Cat #218263), Trypticase Soy Broth (Difco Cat #211768) and Marine Broth (Difco Cat #279110). Liquid media were supplemented with 1.5% (w/v) agar to make Trypticase Soy Agar (TSA) and Marine Agar (MA). In a laminar flow hood, filters collected at each sampling time were removed from the housing, divided using a sterile scalpel, and placed on each of the three culture media. Rainwater samples were serially diluted in sterile water and 100  $\mu$ L of the dilution was spread on the agar plate. Plates were then incubated at room temperature in the dark for 72 h. After culturing, colonies were counted and expressed as colony-forming units per mL (CFU/mL) for rainwater, and colony-forming units per cubic metre of air (CFU/m<sup>3</sup>) for air samples.

After incubation, all plates were photographed and colonies representative of all colony types were selected for purification and identification. Selected colonies were picked onto a clean agar plate of the same type using a sterile pipette tip. After 48 h of growth, cultures were inspected for purity and transferred to 2-ml, 96-well (round deep well) plates (Axygen Scientific, Inc., Union City, CA). The plates were covered with breathable sealing film and incubated at 28 °C with shaking at 175 rpm for 48 h.

DNA was extracted from cultures using the protocol described by Stephenson et al. (2009). DNA was extracted from filters using a MO BIO PowerWater Kit (Carlsbad, CA) as per the manufacturer's instructions. The full 16S rRNA gene was amplified using the Ec16SrRNA1538Fwd and Ec16SrRNA1538Rev primers and the Polymerase Chain Reaction (PCR) parameters described by Grasby et al. (2003). The PCR products from the cultures were purified using a Wizard Purification Kit (Promega, Alexandria, NSW, Australia) according to the manufacturer's instructions and sequenced as described below.

The PCR products from the filters were used to construct clone libraries using the pGEM-T easy vector system (Promega) in *Escherichia coli* JM109 cells as recommended by the manufacturer (Promega). Positive clones were PCR amplified using the SP6/T7 primer set, and PCR products were purified as previously described, and sequenced as described below.

All sequencing was performed by the Australian Genome Research Facility (AGRF, St. Lucia, Queensland, Australia). Sequence

Table 1

List of sampling sites and activities performed on the samples.

Site name	Site location	Sampling type	Analysis performed		
Mt Stromlo, Canberra	35°19′08″S, 148°59′46″E, 676 m asl	Active pumps and rain collection	Microbiology		
Aranda, Canberra	35°15′29″S, 149°05′06″E, 678 m asl	Passive rain collection bowls	Lipids, Sedimentology, Geochemistry, Pollen		
Eden, NSW <sup>1</sup>	37°03′49″S, 149°54′16″E, 10 m asl	Dry deposition	Geochemistry		

<sup>1</sup> Courtesy of Dr. Ann Felton.



**Fig. 2.** Photograph showing the passive rain collection samplers in Aranda, Canberra, showing the time when the bowls were deployed. Once deployed, bowls were left untouched until all were collected at approximately 08:00 AEST on 23 September, by which time the plume had cleared Canberra. The 04:00 AEST bowls (not show here) were the last to collect dust, with the 06:00 AEST bowls only holding clear water.

identification was performed using the Genbank BLAST algorithm (http://www.ncbi.nlm.nih.gov/BLAST/). Sequences were aligned using ClustalW and phylogenetic trees were created in MEGA4 (Tamura et al., 2007).

These methods are different from those presented in De Deckker et al. (2008) but we tentatively compare the 2002 and 2009 dust storm samples collected in Canberra wherever possible.

# 2.5. Lipids

One of the samples that fell with the rain in the suburb of Aranda in Canberra at 23:00 AEST on the night of 22 September, 2009 was dried at room temperature and an aliquot of 243 mg was extracted twice by 10 min ultrasonication in 4 ml dichloromethane: methanol (9:1). The extracts were combined, the solvent removed in a stream of purified nitrogen gas and the resulting total lipid extract (TLE) derivatised using BSTFA (20  $\mu$ l, 60 °C, 60 min) to obtain trimethylsilyl (TMS) derivatives. The derivatised TLE was analysed on an Agilent 6890 gas chromatograph (GC) linked to a Micromass AutoSpec Premier double sector mass spectrometer (MS). Samples were introduced into the GC using a PTV injector in splitless mode at 40 °C (held for 0.1 min) and ramped to 300 °C at a rate of 260 °C/min. All samples were injected in hexane to avoid deterioration of chromatic signals by FeCl2 build-up in the MS ion source through use of halogenated solvents (Brocks and Hope, 2013). The GC oven was fitted with a DB-5MS capillary column (60 m × 0.25 mm i.d., 0.25 µm film thickness) and heated from 40 °C (held for 4 min) to 315 °C at 4 °C/min with a total run time of 120 min using He as carrier gas. The AutoSpec was run in full-scan mode at 1000× resolution with a scan duration of 0.7 s plus 0.2 s interscan delay over a mass range of 55–600 Da. The MS source was operated at 260 °C in El-mode at 70 eV ionisation energy and with 8000 V acceleration voltage.

Oleanolic, betulinic and ursolic acid were identified using authentic standards (Sigma), and saturated, unsaturated and methylated fatty acids using the BAME standard mixture (Sigma). Other compounds were identified based on published mass spectra and GC retention times. Two  $\mu$ g of 18-methyleicosanoic acid methyl ester were added to the TLE as internal standard. Concentrations cited in the text are semi-quantitative and were estimated using integrated signal areas of base ions and are not corrected for differences in MS response.



Fig. 3. Synoptic charts taken from the Australian Bureau of Meteorology showing the progression of meteorological features seen before, during and after the Red Dawn Dust event. (a) 21 September 2009 16:00 AEST; (b) 22 September 2009 16:00 AEST; (c) 22 September 2009 22:00 AEST; (d) 23 September 2009 22:00 AEST.



**Fig. 4.** Map showing the location of the regolith samples collected in the Murray Darling Basin, plus the Onkaparinga River near Adelaide (blue dots). As well as from the Eyre Peninsula and west of the Flinders Ranges, plus in the vicinity of Broken Hill (red dots). The blue dots refer to samples discussed in Gingele and De Deckker (2005) and De Deckker et al. (2010) and De Deckker and Norman (2010) and numbers are those referred to in the geochemical section and satellite image (Fig. 17). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The dust samples collected in Canberra during the October 2002 dust storm and used for lipids analysis were also derived from wet deposition. The analytical methods are similar and we compare the two events where possible.



**Fig. 5.** Enlarged version of the map shown in Fig. 4 showing the location of the sampling sites on the Eyre Peninsula and above as well as those in the vicinity of Broken Hill in northwestern NSW. The numbering are those of the samples which are discussed in the text.

# 2.6. Sedimentology

Grain-size distribution of the minimally dispersed Canberra dust sample, which was collected in the bowl and later on dried, were analysed with a Beckmann Coulter laser particle sizer (LS13320) at the Royal Netherlands Institute for Sea Research (NIOZ), resulting in particle-size distributions from 0.4 to  $2000 \,\mu$ m. Particle sizes are presented as percentage volume per size class for 92 classes. The analysis of the 2002 Canberra dust was done in the exact same way with the only difference being the Beckmann Coulter laser particle sizer series number (LS200 instead of the LS13320 used in this study). Results were therefore directly comparable.

# 2.7. Geochemistry

The two Canberra (from separate bowls) and one Eden dust samples were analysed for Sr and Nd isotopic compositions as well as Sr, Rb, Nd, Sm element concentrations measured by thermal ionisation mass spectrometry using a ThermoFinnigan Triton TI multicollector mass spectrometer operated in static mode at the Research School of Earth Sciences at the Australian National University. Following the digestion of the sample in HF-HCl, the analyte elements were purified by conventional cation chromatography. Sr isotopic compositions were measured on single Ta filaments after loading in H<sub>3</sub>PO<sub>4</sub>. Nd was run on Ta (evaporation)-Re (ionisation) double filaments after loading in HNO3 and dilute H<sub>3</sub>PO<sub>4</sub>. Reference values for <sup>87</sup>Sr/<sup>86</sup>Sr and <sup>143</sup>Nd/<sup>144</sup>Nd measured on the Triton mass spectrometer during this study are  $0.710224 \pm 0.000011$  (2SD, n = 6) for the NIST SRM-987 Sr standard, and  $0.512135 \pm 0.000010$  (2SD, n = 4) for the Ames nNd-1 standard, respectively. Sr, Rb, Nd and Sm concentrations were measured on the same samples by isotope dilution after addition of mixed Sr-Rb and Nd-Sm enriched isotope spikes prior to digestion. Trace element concentrations were also determined by solution aspiration ICP-MS on separate splits of the samples following digestion with HF-HNO<sub>3</sub>. Prior to these analyses, the solutions were passed through a small column containing inert Elchrome® resin to remove organics. Instrument drift was corrected for by normalisation to Be, In, Re and Bi element spikes that were added to each solution as internal standards. Additional information on procedures and errors is available in the work of Gingele and De Deckker (2005).

Procedures for analysis of the trace and rare earth elements were the same as those applied in De Deckker et al. (2008).

## 2.8. Pollen

A sample was derived from the passive rain collector deployed at 23:00 AEST in Aranda, Canberra, and prepared in the palynological laboratory at the University of Göttingen, Germany. The sample was initially treated with warm 10% Na-pyrophosphate  $(Na_4P_2O_7)$  and sieved over a 200 µm and a 7 µm mesh. Material retained below the 7 µm mesh was given a 10% HCl treatment followed by acetolysis (9 parts [CH<sub>3</sub>CO]<sub>2</sub>O: 1 part H<sub>2</sub>SO<sub>4</sub>, 10 min). Organic material was isolated from the remaining inorganic fraction using heavy liquid separation with sodium polytungstate (Na<sub>6</sub>[H<sub>2</sub>W<sub>12</sub>O<sub>40</sub>].H<sub>2</sub>O; specific gravity 2.0, 20 min at 2000 rpm). The isolated material was dehydrated with ethanol (C<sub>2</sub>H<sub>5</sub>OH). Slides were mounted with glycerol  $(C_3H_5(OH)_3)$  and sealed with paraffin. The pollen analysed from the 2002 Canberra dust event (see De Deckker et al., 2008) were extracted using basically the same method outlined above, but analysis was performed in a different laboratory.

# 3. Results and discussion

# 3.1. Meteorology and remote sensing

Leys et al. (2011) identify the enormity of the Red Dawn event over the coast of eastern Australia, from Sydney to Townsville (thus extending over 3000 km), displacing an estimated 2.54 million tonnes of material.

On 21 September, a low pressure cell over Adelaide, South Australia (Fig. 1) anchored a pressure trough reaching into the dry centre of Australia, where most of the continent's dust sources are known to be located (Bullard et al., 2008). This engendered some considerable entrainment of the Lake Eyre Basin, the Strzelecki Desert of South Australia, the Simpson Desert and parts of western NSW. Overnight, between September 21 and 22, the dust travelled over central NSW, heading for southern NSW and the southeast coast of Victoria.

On the morning of September 22, the morning (Terra) MODIS imagery also showed renewed (post-frontal) entrainment in Central Australia, highlighting active sources across seven degrees of latitude ( $\sim$ 770 km) and 10 degrees of longitude ( $\sim$ 1000 km). The cold front that had been located in the Great Australian Bight the previous day had moved over southern SA. By noon on 22 September, when our microbiology pumps were deployed in an already dusty Canberra, dust from central South Australia had reached well into NSW and Queensland (QLD) and the town of Broken Hill in NSW recorded near-total darkness at 16:00 AEST with visibility down to 10–20 m under street light conditions at its worst.

Overnight between September 22 and 23, this South Australian dust became embedded within the frontal system and spread across most of NSW and the southern half of Queensland, with what appears to be increased dust being pulled into the cyclonic vortex of the low pressure cell sweeping over country NSW and towards Canberra (Fig. 6).

Heavy rainfall over Canberra that night brought down some of the dust in the rain. The lifting and instability associated with cold front systems (Sturman and Tapper, 2006) can provide the rainfall mechanism to deliver the dust back to the surface. There is already ample evidence that mineralised particles can act as nuclei for rain droplets (Twohy and Anderson, 2008; Twohy et al., 2009), and more importantly K-felspars minerals (Atkinson et al., 2013). It is unclear however what role the Red Dawn dust played in the Canberra rainfall that night, but in a review of the October 2002



**Fig. 6.** Image showing the MODIS Terra overpass at 22:05 AEST BT12-11 Refer to Fig. 3 for the relevant synoptic chart for the Australian continent at 22:00 AEST on the same day.

event in Canberra where dust was also washed out by rain, De Deckker et al. (2008) suggest that dust may have influenced rainfall during the dust storm as the dust plume traversed through the area where dew point was reached.

On the morning of September 23, the dust storm had reached Sydney (Fig. 7) and this caused numerous disturbances. Visibility was dramatically reduced causing traffic chaos and the airport was also closed. There were numerous reports of respiratory complaints with many admissions to hospitals as reported by Ramachandran (2009).

The dust storm front cleared Sydney by the afternoon of September 23 and reached Brisbane at approximately 16:00 AEST that afternoon. The plume then became fully embedded in the fronttrough system and reached from the cyclonic vortex of the low pressure cell moving towards New Zealand over the Tasman Sea all the way north to northern Queensland.

#### 3.2. Microbiology

Identification of the cultured organisms and the clone library revealed that the composition of the microbiota obtained from the two aerosol and one rain sample was similar (data not shown), so for subsequent analysis, all samples were pooled.

The analyses of the cultured results indicate that there were similar bacterial loads in the air during the day and overnight  $(8 \times 10^2 \text{ and } 5 \times 10^2 \text{ CFU/m}^3, \text{ respectively})$  with up to  $10^4 \text{ CFU/}$ mL present in the rainwater. Fifty-three representative colonies were selected from the different samples and represented 3 phyla and 7 bacterial families (Fig. 8).

A total of 235 sequences were obtained from the clone library of the dust/rain samples (Fig. 8). Representatives of nine phyla and 27

QLD Brisbane NSM Sydney 1elbourne

Fig. 7. MODIS Terra overpass at 09:00 AEST on September 23, 2009 (left clearly showing the dust plume.

bacterial families were identified, with 63 sequences (26.8% of the total) showing the greatest similarity to uncultured organisms in the BLAST database. However, many could be assigned to a phylum, and sometimes family, based on phylogenetic tree clustering (data not shown). Actinobacteria was the most common phylum (36.2% of isolates), with the family Geodermatophilaceae representing 41 of the 73 identifiable Actinobacteria. Of the clones identifiable to a species level, Geodermatophilus obscurus was the most common. The number of Firmicutes identified was low, in contrast to the cultured data, in which Firmicutes were most numerous.

There was minimal overlap in taxa identified between the culture- and DNA-based results. Bacillaceae comprised 40% (21/53) of the cultured isolates, but only 0.4% of the clone library (one clone). The only other bacterial family and genus which appeared in both datasets was Oxalobacteraceae and Massilia, respectively, which represented 0.9% (2/235) of the clone library and 7.5% (4/ 53) of the cultured isolates. This lack of overlap in data between the two methods supports previous studies which show that only a very small proportion of bacterial species can be cultivated in a laboratory environment (Amann et al., 1995).

The majority of the taxa cultured here are widespread soil organisms. Interestingly, several of the less frequently recovered isolates have been previously recovered from clinical settings. One cultured isolate was identified as Erwinia persicina (Enterobacteriaceae family, Proteobacteria phylum - indicated as 'others' in Fig. 8) has been isolated from an elderly patient with multiple health problems in Texas, USA, in 1998 (O'Hara et al., 1998). Arthrobacter oxydans (Micrococcaceae) has also been isolated from clinical specimens several times in a variety of locations (Mages et al., 2008; Wauters et al., 2000), but the pathogenicity of the species is yet to be determined. An isolate of Bacillus licheniformis (Bacillaceae) was also identified, which is known to be a cosmopolitan opportunistic human pathogen (Blue et al., 1995; Santini et al., 1995).

Comparison of the clone library generated here with the one constructed from samples collected from the last major dust storm to affect the Australian east coast in 2002 is difficult, as relatively few sequences were analysed and a rarefaction of only 20% was reported (De Deckker et al., 2008). However, some similarities and differences are clear; 36% of sequences in our dataset are assigned to the phylum Actinobacteria, while none were found in the 2002 dust storm. Other major differences were found in the number of Bacteroidetes and Firmicutes. Each phylum accounted for approximately one quarter of sequences in 2002, but only 7% and 0.4%, respectively, in the 2009 event. Aside from phyla with less than 10% abundance, the only noticeable similarity is the representation of Proteobacteria ( $\sim 25\%$ ) found in each set of samples.

The culture based results in this study are similar to those of another dust/rain event recorded in the Canberra region earlier in 2009 (Lim et al., 2011) in that the primary taxon found is Bacillus, however the next most abundant taxon in the Lim et al rain samples was Pseudomonas, which was not found at all in the 53 colonies examined in our study. Vastly different analytical techniques make further comparison difficult, however it is clear that there are culture-induced biases in both cases, when viewed against the clone libraries above.

Our sampling techniques were not appropriate for the analysis of fungal spores, but a recent study by Hallegraeff et al. (2014), who analysed samples of the Red Dawn dust storm collected at sea between Brisbane and Sydney (Fig. 1), found high concentrations of Aspergillus sydowii fungal spores, similar to those found to be responsible for outbreaks of sea fan disease in the Caribbean (Griffin et al., 2001; Griffin and Kellogg, 2001). This is an important finding as the Red Dawn dust plume passed over the World Heritage Great Barrier Reef.

Although the approach discussed above was not applied to source samples, it nevertheless informs on dust microbial





Fig. 8. Composition of the Canberra dust storm microbiota collected at Mt Stromlo on 22/23 September 2009 (all three samples are pooled in this diagram). The number of clones and cultured isolates of the different bacterial phyla is indicated. The legend designates the patterns that reflect the number of cultured isolates, and the different families represented amongst the different phyla from the clone library.

composition. As for geochemical and pollen libraries, microbial profile libraries will help future work into the microbiological fingerprinting of dust. These need to be developed and increased over time.

# 3.3. Lipids

The distribution of solvent extractable lipids in the sample of dust that fell into the passive rainfall sampler deployed at 23:00 AEST in Canberra overnight (Fig. 9) shows relatively abundant *n*-alkanes in the carbon number range  $C_{20}$  to  $C_{34}$  with a maximum at  $C_{29}$  and a pronounced odd-over-even predominance (carbon preference index CPI = 20) (Fig. 9A). Much less abundant are *n*-alkanes with 16–20 carbon atoms and a local maximum at  $C_{17}$  and no visible even or odd predominance. Also present in traces, and eluting directly after  $n-C_{17}$  and  $n-C_{18}$  respectively, are the acyclic isoprenoid hydrocarbons pristane and phytane (not shown). The only other identified acyclic isoprenoids in the dust extract are  $\alpha$ -tocopherol (vitamin E) and tocopherol acetate (Fig. 9D).

Some of the most abundant compounds in the dust are saturated fatty acids (FA) with a pronounced even-over-odd carbon number distribution in the range  $C_8$  to  $C_{30}$  (Fig. 9B). An exception is nonanoic (pelargonic) acid (C9:0) that is more abundant than its even-numbered homologues C8:0 to C14:0. The two most abundant alkanoic acids are palmitic (C16:0) and stearic (C18:0) acid, with a secondary maximum at C26:0. Monounsaturated (C14:1, C16:1, C18:1, C20:1) and diunsaturated acids (linoleic acid, C18:2<sup>9:12</sup>) were also identified. Conspicuous is the presence of a series of glycerol esters of FAs C12:0, C14:0, C16:0, C18:0 and C18:1 that may represent partial degradation products of phospholipids (Fig. 9D).

*n*-Alcohols with 8–31 carbon atoms were also identified in the Canberra dust in similar abundances to *n*-alkanes but with a strong even-over-odd predominance and a maximum at  $C_{26}$  (Fig. 9C). Similar to the FA distribution, nonanol with 9 carbon atoms is significantly more abundant than its close even-numbered homologues. In contrast, sterol concentrations and diversity are relatively low in the dust extract. Identified were cholesterol and

the common phytosterols sitosterol, brassicasterol, stigmasterol and campesterol (in order of abundance) as well as the more unusual  $5\alpha$ -stigmastanol. Diagnostic faeces indicators such as  $5\beta$ -stigmastanol, epi- $5\beta$ -stigmastanol, coprostanol, cholestanol and epi-cholestanol were not detected.

The most abundant compounds in the dust are the pentacyclic triterpenoids oleanolic acid (160  $\mu$ g/g), betulinic acid (29  $\mu$ g/g) and ursolic acid (570  $\mu$ g/g). Detected in lower concentrations were lupeol,  $\alpha$ - and  $\beta$ -amyrin, and several putative terpenoids with molecular ions from m/z 484 to 598 that remain unidentified (Fig. 9D). The most prominent unidentified signals in the mass chromatogram are five compounds of a homologous series characterised by the base ion m/z 143 and molecular masses at 158, 244, 330, 416 and 502 (the last value only tentatively identified). These masses are consistent with a series of oligoesters or oligoethers.

The short-chain  $(C_{10}-C_{18})$  *n*-alcohols and saturated and unsaturated FAs detected in the dust are abundant in many plants and other organisms and not specific for any particular source. Unusual, however, is the high concentration of pelargonic acid (C9:0) in comparison to even-numbered FAs in the range C8:0-C14:0 (Fig. 9B). Although odd-carbon numbered FAs are commonly scarce in natural samples, pelargonic acid may have been generated by atmospheric oxidation of oleic acid (C18:1<sup>9</sup>) (Rogge et al., 1993) or decomposition of oleic acid in cooking oil (Schauer et al., 2002). However, in the Canberra dust, these processes are not likely a major source of C9:0 as other typical atmospheric oxidation products or cooking oil residues were beneath detection limits. Alternatively, pelargonic acid may have been introduced as a pesticide. C9:0 is abundantly applied as a herbicide on crop plants, as a weed killer in urban areas (Dayan et al., 2009) and blossom thinner for fruit trees in orchards (Fallahi, 1997). Such an anthropogenic origin is, for instance, a likely source of elevated concentrations of pelargonic acid in surface soils of crop fields in San Joaquin Valley in California (Table 2 in Rogge et al., 2007). Notably, C9:0 concentrations are much lower in fugitive dust from open lot dairies and cattle feedlots in the same area where C9:0 is probably not applied (Table 2 in Rogge et al., 2006). Although natural pelargonic acid is rare in the biosphere, it may occur in relatively high



**Fig. 9.** The distribution of volatile lipids in the solvent extract of the Canberra 2009 dust sample. The upper panel gives relative concentrations of (A) *n*-alkanes, (B) straight-chain saturated fatty acids, and (C) straight-chain saturated alcohols. Absolute concentrations were estimated using integrated signal areas of base ions and are not corrected for differences in MS response. (D) Total ion chromatogram annotated with compound identifications.  $\bullet = n$ -alkanes,  $\bigcirc =$  saturated *n*-alcohols and  $\vee =$  straight-chain saturated fatty acids. Ca:b indicates fatty acids with a, carbon atoms and b, double bonds. Std. = internal standard. Major unidentified compounds are characterised by their base ion (b) and molecular ion (M).

# Table 2

Comparison of organic compounds identified in the 2 dust samples collected in Canberra during the dust storms of 2003 and 2009, and interpretation of their origin.

Biomarkers for	2002 Canberra dust	2009 Canberra dust
General biomass	Abundant Low molecular weight saturated and unsaturated fatty acids; Animals: cholesterol Plants: sitosterol	Abundant Low molecular weighty saturated and unsaturated fatty acids; fatty acid glycerol esters; Animals: cholesterol Plants: sitosterol, brassicasterol, stigmasterol, campesterol, stigmastanol
Biomass burning	Abundant Abundant PAH <sup>®</sup> and N-bearing compounds; retene and dihydroabietic acid as indicators of conifer burning	None
Gymnosperms	Abundant Retene, dihydroabietic acid and unknown terpenoid	None
Angiosperms	Very low Low concentrations of two unidentified triterpenoids; Conspicuous absence of cuticular waxes	Abundant $\alpha$ - and $\beta$ -amyrins, lupeol; very high concentrations of pentacyclic triterpenoids ( <i>Eucalyptus</i> ?) Very abundant cuticular waxes including <i>n</i> -alkanes, alcohols and fatty acids
Fossil fuel	None	Present Diesel oil combustion residues
Agricultural activity	None	Present Animal feed supplement (tocopherol acetate); possible herbicide (pelargonic acid)
Faecal matter	None	None
Dry lake beds/salt lakes	None	None

\* PAH = polycyclic aromatic hydrocarbons.

concentrations in the bark of *Eucalyptus* (Domingues et al., 2011), a common genus of trees in the dust source area. Such a biogenic contribution to the Canberra dust is supported by the presence of pelargonic alcohol (1-nonanol, C9) that also occurs in excess abundance (Fig. 9C) but has no common anthropogenic source. Thus, in the Canberra dust, pelargonic acid is most likely derived from debris of *Eucalyptus* tissue with a possible contribution from pesticides.

Also abundant in the Canberra dust are *n*-alkanes, *n*-alkanoic acids and *n*-alcohols with 20–33 carbon atoms and pronounced carbon number preferences (Fig. 9A–C). These distributions are typical of epicuticular wax coatings of terrestrial plants and may have been introduced by direct abrasion of leave surfaces or through plant debris in the dust source (Rogge et al., 2006). In contrast to *n*-alkanes with 21 and more carbon atoms, *n*-C<sub>16</sub> to *n*-C<sub>20</sub> display no carbon number predominance (Fig. 9A). This distribution, together with the presence of acyclic isoprenoids pristane and phytane, is typical for diesel and diesel combustion residues emitted from trucks and farming equipment (Brocks et al., 2003). Diesel residues may either have contaminated the soil in the dust source area or, more likely, were introduced by airborne soot and aerosols.

Tocopherol acetate is another component in the dust with a likely anthropogenic origin (Fig. 9D). It is a stable derivative of vitamin E ( $\alpha$ -tocopherol) that is added to cattle feed as a dietary supplement. Its presence indicates that some dust may have come from animal feedlots (Rogge et al., 2006). In contrast, vitamin E itself, a natural component of plant matter, is oxidation-sensitive and was only detected in traces.

The most abundant sterol in the Canberra dust is cholesterol (Fig. 9D). Cholesterol occurs in all animals, but smaller concentrations can also be found in some plants and fungi. In urban environments, meat cooking activity can be a major source of airborne cholesterol (Rogge et al., 1991). However, as the dust never travelled through extensive urban areas, a direct source from soil fauna is more likely. The four phytosterols detected in the dust, sitosterol, campesterol, stigmasterol and brassicasterol are generally the most common sterols in terrestrial vegetation (Rogge et al., 2006) and not specific for any particular source. Also detected in minor concentrations was  $5\alpha$ -stigmastanol. This sterol is directly found in some plants (lida et al., 1981) but more commonly in faecal matter and soil where it forms as a bacterial reduction product of C<sub>29</sub>-phytosterols (Evershed et al., 1997; Rogge et al., 2006). However, in the Canberra dust sample, the major faecal derivatives of phytosterols,  $5\beta$ -stigmastanol and epi- $5\beta$ -stigmastanol (Rogge et al., 2006), were below detection limits. Thus, the  $5\alpha$ -stigmastanol was most likely produced by the degradation of sitosterol in soil. The very low concentration or absence of 5β-stigmastanol, epi-5β-stigmastanol and other common faecal indicators such as coprostanol, cholestanol and 5<sub>β</sub>-campestanol is notable. Although kangaroo, rabbit, cow and sheep droppings are abundant in the Australian landscape and easily disintegrate into fine particles, they evidently did not contribute detectable faecal biomarkers to the 2009 dust.

All of the identified terpenoids in the dust (oleanolic, betulinic and ursolic acids, lupeol,  $\alpha$ -amyrin and  $\beta$ -amyrin) are abundant in angiosperms but not in conifers (Otto et al., 2005). Oleanolic, betulinic and ursolic acids are structurally closely related and constitute the most abundant molecules in the dust, adding up to a total of ~750 g per ton, or an estimated  $1.9 \times 10^6$  kg for the entire dust storm (using Leys et al., 2011 figures). Although the terpenoids are widespread among angiosperms, a likely source in the Canberra dust are trees of the genus *Eucalyptus* that may contain high concentrations of oleanolic, betulinic and ursolic acids (e.g., Siddiqui et al., 2000; e.g., Domingues et al., 2011). The three acids are particularly concentrated in the surface layer of bark. For example up to 56 g/kg ursolic acid occur in the dried bark of branches of *Eucalyptus globulus*. However, the acids are also present in lower concentrations in cuticular wax, e.g., up to 2 g/kg wet-weight ursolic acids in adult leaves (Domingues et al., 2010). *Eucalyptus* leave and bark extracts also yield  $\beta$ -amyrin and sitosterol, albeit in much lower concentrations, consistent with the lower abundance of these compounds in the dust (Fig. 9D).

Table 2 compares the molecular composition of dust that fell in Canberra in October 2002 (De Deckker et al., 2008) and September 2009. The differences are significant and point to disparate sources of dust. Satellite images of the 2002 dust storm show that it travelled through plumes of smoke of vegetation fires (refer to Fig. 12A-C in De Deckker et al., 2008). Consequently, the dust that later fell in Canberra contained several molecular markers of biomass burning. By contrast, biomarkers diagnostic for smoke or fire were not detected in the 2009 dust. Moreover, identified plant biomarkers in 2002 were derived from conifers while angiosperm biomarkers were insignificant. Particularly notable in 2002 was the absence of plant waxes that are commonly ubiquitous and abundant in soils. Conversely, the 2009 dust lacked evidence of conifers but was strongly dominated by typical angiosperm biomarkers including highly abundant triterpenoids and plant waxes. Furthermore, the 2002 dust showed little evidence of anthropogenic activity while the 2009 dust contained residues of diesel fuel, animalfeed supplement and possibly weed killer. Surprisingly, neither the dust of 2002 or 2009 contained detectable levels of faecal matter. Also absent were biomarkers diagnostic for cyanobacteria, algae and halophilic Archaea that would indicate a dust source from dry lake beds and salt lakes.

## 3.4. Sedimentology

The grain-size analysis shows a well-sorted relatively unimodal distribution with a dominant mode around ~40  $\mu$ m and a small shoulder at ~103  $\mu$ m (Fig. 10). This well-sorted character is typical for wind-blown deposits that are the result of dry deposition (e.g., Krumbein and Pettijohn, 1938) although due to the nature of grain-size analysis with a laser particle sizer, the coarse-grained shoulder may be more prominent as a result of platy minerals, which show the same aerodynamic behaviour as finer-grained spherical minerals (Stuut et al., 2005; De Deckker et al., 2008).

The particle-size distribution of the 2009 Canberra dust sample compares very well with dust samples also collected in Canberra



**Fig. 10.** Plot of the size distribution of the Canberra dust sample collected in 2009 (orange line) compared with the one collected in October 2002 (red line), and duststorm samples collected off the Sahara Desert over the Atlantic Ocean in 1998 (M41-D24, Stuut et al., 2005), 2005 (M65-D5), and 2012 (M89-D10). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

during the October 2002 dust storm (De Deckker et al., 2008). The particle-size distributions are generally relatively narrow, typical for wind-blown sediments, which are well-sorted. These distributions were characterised by a fine tail, possibly as a result of wet deposition, a dominant mode between 20 and 90  $\mu$ m and often a small shoulder between 100 and 200  $\mu$ m. Microscope observations of these samples showed that the dominant mode consists of quartz and feldspar particles, and the coarse shoulder of platy minerals like micas. The platy shape of these coarser minerals determines their aerodynamic behaviour (De Deckker et al., 2008). The fine-grained tail may however be an artefact of the laser, which tends to underestimate the clay fraction in natural samples due to the particle shape of clays (Konert and Vandenberghe, 1997).

## 3.5. Palynology

The analysed sample weighed 0.34 g and only a total of 563 pollen grains were counted. The estimated amount of pollen was 534,252 grains per gram of sediment. Only two *Sporormiella*-type (often associated with animal faecal matter) spores and 107 charcoal particles were seen in the slide during the pollen count, giving a charcoal/pollen ratio of 0.19. The dryland pollen sum totals 540, accounting therefore for 96% of the total pollen sum. Fig. 11B provides a representative display of the major groups recognised. The woody-herbaceous taxa Asteraceae/Tubuliflorae and Amaranthaceae/Chenopodiaceae comprise 62% of the dryland pollen, while pollen from trees and shrubs account for 19% of dryland pollen sum. The latter group is dominated by *Callitris*, which accounts for 59% of the woody taxa. This implies an arid region with sparse trees and shrubs.

Consultation of the South East Australian Pollen database (D'Costa and Kershaw, 1997), shows that the relatively high *Callitris* values (a native conifer), combined with the high Asteraceae/Tubuliflorae and Amaranthaceae/Chenopodiaceae numbers

would point to the area just to the north of Port Augusta in South Australia. This area is south of the Lakes Torrens and Gairdner area which is characterised by a low rainfall and excessive evaporation. The palynological data support the meteorological and satellite observations and geochemical findings.

Comparison of the data for the two Canberra dust events is presented in Fig. 11. It is clear that the much higher abundance of Asteraceae Tubuliflorae and Amaranthaceae/Chenopodiaceae in the 2009 events compared to the lower values for *Eucalyptus* points to much more arid conditions on the ground north of Port Augusta. Of interest also is that the presence of the (introduced to Australia) Pinus pollen in the 2002 dust sample relates to the dust storm having picked up, during the event, ubiquitous pine pollen produced in the Canberra region. At that time, pine plantations did abound around Canberra, but were later completely destroyed from the massive fires on January 2003. Those forests have not been replanted to a large extent and this explains why no *Pinus* pollen was recovered from the second dust event. In addition, the inclusion of the pine pollen in the first spectrum clearly indicates that additional material (clay particles, organic compounds, microbes etc.) can be entrained as the dust moves across the landscape. Similarly, in the 2002 dust markers for biomass burning (retene and dihydroabietic acid, indicative of gymnosperm burning) were recognised (see De Deckker et al., 2008), and this is explained by the fact that the dust plume passed over some smoke plumes resulting from several bush fires that were active in NSW during the dust storm. Similarly, the presence of diesel oil combustion residues, animal feed supplement and possible herbicides (Table 2) in the 2009 Canberra dust sample points to the same phenomenon, viz. additional material and components can be entrained during the movement of the fronts and dust plumes, even if the source region of the dust is devoid of such substances. The other important observation that was not made in De Deckker et al. (2008) was that, although the major front that carried the dust across



Fig. 11. Comparison of the pollen spectra for October 2002 (A) and September 2009 (B). Also shown is the suggested sites of origins (\*) for the detected pollen.



**Fig. 12.** Graphic illustration showing the location of the sampling sites in the Murray Darling Basin which were used to identify the source of the dust that fell in Canberra in October 2002. The red arrow points to the movement of a large part of the dust perpendicular to the principal direction of the fronts that entrained and carried the aeolian material. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

southeast Australia travelled in a southwest to northeast direction, the dust that fell in Canberra originated from the northwest part of NSW, having travelled along part of the front at right angles. This is entirely the result of the interplay of northwest to southeast flow ahead of the front in the lowest part of the troposphere, and the southwest flow behind the front, graphically illustrated in Fig. 12.

## 3.6. Inorganic geochemistry

The plot of Rb (ppm) versus Nd (ppm) clearly shows (Fig. 13A) that the two Canberra dust samples were poorer in the concentration of these two elements than the one collected at the coastal town of Eden further along the dust path than Canberra (Fig. 1). Surprisingly, the passive rain bowl deployed at 19:00 AEST on 22 September accumulated less material than the bowl deployed 9 h later at 04:00 AEST. The difference in elemental composition between the samples can be interpreted as a progressive spatial/ temporal evolution of the dust composition as the storm traversed over eastern Australia (Fig. 12A). The Sm/Nd versus Sr (ppm) plot (Fig. 12B) shows greater compositional differences between all three samples compared to the <sup>143</sup>Nd/<sup>144</sup>Nd and <sup>87</sup>Sr/<sup>86</sup>Sr isotope ratios, effectively highlighting the variability between the three samples (Fig. 13C and refer also to Table 3).

Results of the ICP analyses for major, trace elements and Rare Earth Elements (REE) of samples from the 49 inland localities were compared against the two samples of dust collected in Canberra. Of importance is the comparison of some of the REE as described previously by De Deckker et al. (2008) for the Murray Darling Basin samples and the dust fall in Canberra in 2002. In particular, the following REE normalised to the 2009 Canberra dust (Figs. 13 and 14): Samarium, Praseodymium and Gadolinium.

First, the Sm concentration in the two Canberra samples and the one Eden sample were compared with the surface samples from South Australia and near Broken Hill (Fig. 14), and 7 other sampling sites (Fig. 5). We found that sites 12, 18, 20, 21, 27, 32 and 34 had almost the same amount of Sm.

When the same samples were compared for Pr and Gd (Fig. 14) however, only three (12, 28 and 31) of the seven selected surface samples, plus one south from those latter sites (site 42a for Lake Giles, Fig. 15) produced values similar to the Canberra and Eden dust samples.

By combining these findings, we were able to pinpoint sources in the vicinity of lakes Torrens and Gairdner (Fig. 1) as the primary source of a large proportion of the dust that traversed towards the east coast of Australia in September 2009. This supports similar findings based on the pollen spectrum of the Canberra dust as discussed above.

The comparison of the <sup>87</sup>Sr/<sup>86</sup>Sr and <sup>143</sup>Nd/<sup>144</sup>Nd values obtained from our field campaigns to the isotopic analyses of the 2002 and 2009 dust collected in Canberra clearly identifies different sources for these two events. The 2002 Canberra dust originated from the Murray sub-basin (Fig. 17), where Bourke and Cobar are located, whereas the 2009 Canberra and Eden dust samples originate from regions between the Murray sub-basin and central and western South Australia (Fig. 16). It is evident that the Lake Eyre Basin and the Darling sub-basin did not supply the Canberra and Eden dusts during the 2002 and 2009 events. Nevertheless, we ought to remark that the dust which reached Brisbane (Fig. 1) to the north may have come in part from elsewhere such as indicated in the DustWatch report prepared by Leys et al. (2009).

Large dust storms along the east coast of Australia are often assumed to have originated in the Lake Eyre Basin (McGowan et al., 2008). The Lake Eyre Basin did indeed provide dust to the Red Dawn event, but the satellite imagery and above analysis also highlight many regions outside that basin. A number of studies exist on the geochemistry of the 2009 Red Dawn dust storm based



**Fig. 13.** Series of 3 plots for comparison between the 2 Canberra dust samples (collected at 7 pm on 22 September and 4am on 23 September, 2009) with the Eden sample collected on September 23. (a) Nd (ppm) versus Rb (ppm); (b) Sm(ppm)/Nd(ppm) versus Sr (ppm); and (c) <sup>143</sup>Nd/<sup>144</sup>Nd versus <sup>87</sup>Sr/<sup>86</sup>Sr.

#### Table 3

Showing the results of geochemical analyses of the 3 dust samples, two of which were collected in Canberra (Aranda) on the night of September 22–23, 2009 and the other that was collected on 23 September, 2009 at Eden, some 200 km SE of Canberra.

Sample	<sup>87</sup> Sr/ <sup>86</sup> Sr	2SE	Sr ppm	Rb ppm	±	<sup>143</sup> Nd/ <sup>144</sup> Nd	2SE	Nd ppm	±	Sm ppm	±	εNd
Canberra 7 pm Canberra 4 am	0.716882 0.7162	0.000011 0.000008	116.62 152.93	84.24 91.66	0.85 1.09	0.512276 0.512285	0.00001 0.000008	26.937 27.653	0.001 0.001	5.514952 5.690306	0.001113 0.000896	-7.06 -6.89
Eden	0.719213	0.000009	120.26	98.4	1.12	0.512257	0.000006	28.271	0.002	5.790998	0.00035	-7.44

<sup>143</sup>Nd/<sup>144</sup>Nd values were corrected to a value of 0.511860 for the La Jolla reference standard and ε-Nd is calculated relative to a bulk earth value of 0.512638.

on air samples collected in Sydney that were compared broadly and solely to ground samples collected near the centre of the Lake Eyre Basin. For example, Rahdi et al. (2010) and Box et al. (2010) collected a series of samples during the Red Dawn dust event and also three days later during a second event. Both studies used Ion Beam Analysis (IBA) to examine major element ratios (e.g., Fe/ Al and Fe/Si) and compared the results to reference samples collected in central regions of the Lake Eyre Basin, putting forward this region as the source of dust that passed over Sydney. A direct comparison between their geochemical results and ours was not possible as their studies contained few elements and none in common with our study. Interestingly, our own analyses (Sr and Nd isotopes and some REE) on the Canberra dust suggests that for Canberra at least, the sources of the dust were located south of the Lake Eyre Basin (Lake Torrens and Lake Frome), the Murray-Darling Basin and western NSW. This is also supported by the satellite imagery (Fig. 17) and especially the pollen spectra.

The finding that two samples of the same dust plume taken at different times but at the same location and in the same way (i.e., the two Canberra samples) could have different geochemical signatures is interesting. This highlights dust compositional change along the same dust path (dust path passing over Canberra). Additionally, it is interesting to find that dust caught downwind can produce a third signature (Eden), or, for collection sites latitudinally close to Canberra (Sydney), be attributed to sources hundreds of kilometres from the sources we identified (i.e., Rahdi and Box et al., 2010 studies). These differences may highlight the variability in the dust composition within a dust plume, which has



**Fig. 14.** Samarium (Sm) concentrations for regolith samples from South Australia normalised against that of the Canberra 2009 dust sample that fell at 4 am. Samples labeled 4 am and 7 am were collected in Canberra.



**Fig. 15.** Praseodymium (Pr) and Gadolinium (Gd) concentrations for regolith samples from South Australia normalised against the Canberra dust sample that fell at 4 am. Samples 12 and 21 provide the closest match.

ramifications for any research concerned with the geochemical fingerprinting of downwind dust based on single samples. The differences of many hundreds of kilometres between the suggested sources of the Red Dawn dust for the Canberra-Sydney region (based on various methods of analysis) suggests either that (a) material from sources across a wide area meets and mixes well before reaching the coast, or that (b) results are dependent on the analytical method(s) used to fingerprint the dust, or (c) both.

Researchers aiming to identify the origin and impact of aeolian (as opposed to fluvial) dust in the oceans from deep-sea cores for example, where terrigenous material is mixed with marine pelagic sediments, are unlikely to be able to single out any one location precisely and entirely. Sr and Nd isotope analysis, as well as sediment grain sizes, can however help distinguish between fluvial and aeolian origins. For example, Gingele et al. (2007) were able to distinguish between aeolian versus fluvial contributions based on Nd isotopes analysed in a core from offshore the mouth of the Murray River near Kangaroo Island in southern Australia.



**Fig. 16.** A comparison of the <sup>87</sup>Sr/<sup>86</sup>Sr and<sup>143</sup>Nd/<sup>144</sup>Nd isotopic compositions of dust collected from the 2002 and 2009 events, as well as the 2009 Eden dust, compared to regolith samples from potential Australian dust source regions. The envelopes are hand drawn and no statistical analysis was used.



**Fig. 17.** Satellite image showing the location of some of the sites that have comparable Sm, Gd and Pr values to the 2009 dust samples. In particular, sites 20 and 28 represent opposite ends of Lake Gairdner and are located more than 100 km apart, and the northern end of Lake Torrens (site 12). The extensively folded Flinders Ranges are visible in dark brown between Lakes Torrens and Frome.

## 4. Conclusions

In this study we examine in great detail the progress and composition of one of Australia's largest dust storms in recorded history, the Red Dawn dust storm. We combined a dust-storm forecasting routine with remote sensing and meteorology to provide a detailed chronological account of this event and used analyses from the fields of geochemistry, sedimentology, biochemistry, microbiology and palynology to fingerprint some of its dust. Wherever possible we compared this analysis to that done for a similar event in 2002. The salient findings of this study are:

1. The Red Dawn was an austral spring dust event, similar in many respects to previously documented dust events for eastern Australia (e.g., De Deckker et al., 2008). Dust entrainment was

associated with the strong northwest winds and unstable atmosphere ahead of a trough-front system passing over a highly erodible landscape, while the dust trajectory was controlled by the northwest flow ahead of the front and the southwest flow behind the trough-front system.

- 2. The lipid compositions of the 2002 and 2009 dust storms differ significantly, indicating that they had origins and/or followed paths with contrasting vegetation and land use patterns. The stark organic geochemical difference between the two dust events opens the possibility of a lipid reference library that may allow distinguishing between dust from different Australian landscapes and vegetation zones. There are hundreds of additional metabolomic markers that were not scrutinised in the present study. Thus, inclusion of a wider variety of source specific markers and their compound specific C-, H- and N-isotopic compositions may allow different soil types and vegetation zones at a relatively fine scale.
- 3. Geochemically, we show that it is possible to identify the source of dust entrainment if sufficient regolith samples from potential dust sources are available. Sr and Nd isotopic compositions as well as the relative and absolute abundances of REE can be used with some confidence to pinpoint a source. Elemental fingerprinting is however probably most effective when combined with several other fingerprinting methods, as shown in this study, and certainly involving more than just a few baseline elements found everywhere in the Australian landscape (e.g., Fe, Al, Ti).
- 4. The microbiology of dust can vary considerably between dust storms and, depending on the techniques employed, different organisms can be identified. It is assumed also that the microbiota could change during the passage of a dust plume as additional organisms can be subsequently entrained, and viability may vary with local conditions. At this stage, we cannot ascertain – based on the samples we analysed – that it is possible to fingerprint dust from its microbiological composition, and further research is therefore recommended.
- 5. Grain-size analysis of dust samples can also help determine if some wet deposition did occur (i.e., if a tail of fine-grained particles is found), thus implying some washing out of the atmosphere by rain.
- 6. Pollen spectra are a very useful tool for fingerprinting the source of the dust, but we found that additional pollen can be entrained along the pathway of the dust plume, making the identification of the initial source of the dust more problematic.
- 7. Finally, Australia is too large a continent to collect inland source reference samples at random for any of the fields of science presented in this study. As such, it is important to know exactly where active dust sources are located during the few months or years (i.e., <5 years) leading up to events. This can be done using remote sensing as shown in this study.

We have demonstrated here the complexity of dust plumes as they travel over the landscape. We conclude from this study that a multi-disciplinary approach is advantageous over using a single approach, but that it is unlikely that any single source of the dust can be definitively identified, and instead analyses will only ever highlight locations that partially contributed material to an already well formed and mixed passing dust plume. Only an increase in offshore and inland reference sampling will help refine these fingerprinting techniques in the future.

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