Geochemical and microbiological fingerprinting of airborne dust that fell in Canberra, Australia in October 2002

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[1] During the night of October 22-23, 2002, a large amount of airborne dust fell with rain over Canberra, located some 200 km from the coast at an average altitude of 650m. It is estimated that during that night about 6g.m^{-2} of eolian dust fell. We have conducted a vast number of analyses to "fingerprint" some of the dust and used the following techniques: grain size analysis, SEM imagery, major, trace and rare earth elemental, plus Sr and Nd isotopic analyses, organic compound analyses with respective compound-specific isotope analyses, pollen extraction to identify the vegetation sources, molecular cloning of 16S

rRNA genes in order to identify dust bacterial composition. In addition we investigated the meteorological conditions that led to the dust being transported, with some reaching Canberra, and MODIS satellite imageries were also examined. Grain sizes of the mineral dust show a bimodal distribution typical of proximal dust rather than what is found over oceans and the bimodal aspect of size distribution confirms wet deposition by rain droplets. The inorganic geochemistry points to a source along the Darling River in NW New South Wales, a region that is characteristically semi-arid and, both the organic chemistry and palynoflora of the dust confirm the location of this source area. Meteorological reconstructions of the event again clearly identify the area near Bourke-Cobar as being the source of the dust. DNA analyses show that most obtained 16S rRNA sequences belong mainly to three groups: *Proteobacteria* (25%), *Bacteriodetes* (23%), and *gram-positive bacteria* (23%).

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1. Introduction

[2] The contribution of dust plumes from Africa to the Atlantic Ocean, the Caribbean and the Amazon Basin, is now well documented [*Duce et al.*, 1980, *Savoie et al.*, 1980, *Talbot et al.*, 1988, *Swap et al.*, 1992, *Prospero*, 1999, *Stuut et al.*, 2005], especially since the availability of satellite imagery. Estimates of the amount of dust transported from Africa identify that huge amounts of sediment are deflated from African soils and lake basins. Similarly, the contribution of aeolian dust down wind of the huge Chinese 'dust bowl' [*Garrison et al.*, 2003] has been a major concern for countries such as Korea and Japan, especially with respect to human health; one major concern being the effect of dust causing respiratory problems to humans [*Griffin et al.*, 2001b, *Griffin and Kellogg*, 2004]. More recently, alarming discoveries on the microbial content of African dust reaching the Caribbean region raised concerns not only for human health but have been shown to affect the 'health' of coral reefs with several bleachings of corals registered after African dust reached the western shores of the Atlantic Ocean [*Shinn et al.*, 2000]. Already, *Griffin et al.* [2003] and *Kellogg et al.* [2004 and references therein] have identified up to 120 bacteria in African dust, some of which are of direct concern to human health. In addition, high concentrations of particular metals, such as mercury found in Florida, are now considered to have originated from African dust [*Landing et al.*, 1995]. All these phenomena associated with aeolian dust deserve much consideration in Australia as too few investigations have been carried out on dust composition, especially with respect to its organic composition and microbiology, plus metals. Much Australian dust is special as it partially originates from (hyper)saline lake floors, i.e. containing microbial life that is adapted to saline conditions. We will investigate the effects of Australian dust on oceanic life, including corals and the seafloor microbiology. The study presented herewith is the first attempt at using as many techniques as possible to 'fingerprint' a sample of airborne dust, much of which became available after a spectacular dust fall associated with some rain in October 2002 in Canberra, the capital of Australia.

2. The Canberra Dust Event of October 22-23, 2002

[3] The year 2002 proved to be a year of extensive drought in Australia and, in particular, for the vast ~1.10^6 km^2 Murray Darling Basin, due to a significant rainfall deficiency combined with above maximum temperatures that led to a reduction in vegetation cover [*McTainsh et al.*, 2005]. It is no surprise therefore that on October 22 that year a frontal system

accompanied by vast quantities of dust moved through South Australia and, over the following 36 hours, it transgressed eastern Australia. The dust associated with this front spanned over 2,400 km from Mackay [21° 09'S 149° 11'E] in Queensland in the north down to the Victorian-New South Wales border [37°30'S, 150°S] in the south (Figure 1). The front eventually continued over the Tasman Sea where a dust plume was tracked by satellite imageries on October 24, 2002 [see *Chan et al.,* 2005: 340] that also clearly show dust over the southern part of the Great Barrier Reef.

[4] *McTainsh et al.* [2005] described the meteorological data for this spectacular event and concluded that the "dust storm was 2,400 km long, up to 400 km across and 1.5 to 2.5 km in height". These authors also estimated that the dust load was 3.35 – 4.84 Mt and concluded by saying that this event represented the largest dust storm reported for Australia. During the night of October 22-23, 2002, it rained in Canberra which was located under the path of the dust plume. The rain that fell yielded a large amount of brown mud that covered many surfaces in the Canberra area, including cars parked outside overnight. Atmospheric conditions that led to the entrainment of dust, and its transport via the atmospheric front in eastern Australia are discussed in *McTainsh et al.* [2005] and *Chan et al.* [2005], but are further developed here, especially with respect to its deposition through the rain in the Canberra region.

[5] Canberra is located at an average altitude of 650m on the eastern edge of the Murray Darling Basin [which is drained by 2 of the Australian major rivers, the River Murray and the Darling River] (Figure 1). The latter river drains a large portion of the northern and western parts of the Basin which is characterised by being semi-arid to arid, and with rains principally falling in the Summer.

[6] This paper aims at identifying the nature of the dust that fell in Canberra; to 'fingerprint' it chemically with respect to its inorganic and organic components. In addition, DNA was extracted from Polymerase Chain Reaction (PCR) from a sub-sample of the dust, and its palynoflora was also studied. The overall aim was to identify the source of the dust and to determine characteristics of airborne dust that may one day be used to identify aeolian components in lacustrine and deep-sea cores in the Australian region.

FIGURE 1 ABOUT HERE

3. Material and methods

3.1. Collecting the dust

[7] By chance, one of us (PDD) cleaned a series of almost horizontal windows the weekend prior to the Canberra dust fall that occurred on the night of October 22-23, 2002. These windows are next to a house in the leafy suburb of Aranda which is located near the centre of the city, away from any industrial area, north of the Black Mountain Reserve. The morning after the event, a thick, dried paste of brown mud covered the windows and the material was carefully raked using clean, unused razor blades. This material was transferred into clean glass scintillation vials and subsequently stored in a small refrigerator at 4° C in the micropalaeontological laboratory in the Department of Earth and Marine Sciences [=DEMS, now Research School of Earth Sciences = RSES] at the Australian National University [ANU].

[8] Several sub-samples of the dust material were selected for a variety of analyses, including one that was sent to the Max Planck Institute of Marine Microbiology some 2 years after its collection. Detailed chemical analyses of another sub-sample, including major- and trace-elements and rare earths, together with the organic compounds were performed. Additional work on other sub-samples included observation of the material by scanning electron microscopy, grain size analysis and pollen extraction, and is also reported in this paper.

[9] By weighing another sub-sample, it is estimated that 6g.m^{-2} of dry weight fell (rained) on the windows of the Aranda home; this amount is equivalent to the estimate for the Australian continent over one entire year [see *Prospero et al.*, 2002] !!

3.2. Grain size analysis of the dust sample

[10] Grain sizes of the minimally dispersed dust were measured with a Coulter laser particle sizer (LS200) at the Research Center Ocean Margins, Bremen, Germany, resulting in particle-size distributions from 0.04 – 2000 µm. Particle sizes are presented as percentage volume per size class for 92 classes.

3.3. Observations of grain particles of the dust under the SEM

[11] A small amount of dust material was transferred onto non-conductive double-sided sticky tape, then coated with Au-Pd in an small Ar chamber, and then observed under a Cambridge scanning electron microscope [SEM] at the Electron Microscope Unit at the Research School of Biological Sciences at ANU.

3.4. Inorganic geochemical analyses of the dust

[12] Major elements Na, Mg, Al, Si, P, S, K, Ca, Ti, Mn, Fe were determined by XRF on a Philipps (PANalytical) PW2400 X-ray spectrometer in the former DEMS [now RSES]at ANU. Lithium borate discs were prepared by fusion of 0.27g of dried sample powder and 1.72g of "12-22" eutectic lithium metaborate-lithium tetraborate. The major elements were calibrated against 28 international standard rock powders. Lithium borate discs were then dissolved in 2% HNO_3 and, after dilution, concentrations of the trace element Sc, V, Y, Cr, Mn, Co, Ni, Cu, Zn, Ga, Rb, Sr, Y, Zr, Nb, Mo, Cd, Sn, Cs, Ba, La, Ce, Pr, Nd, Sm, Gd, Dy, Er, Yb, Lu, Hf, ^{207}Pb, ^{208}Pb, Th and U were measured by ICP-MS at RSES at ANU. The samples were calibrated against NIST 612 blue glass as an external standard material, using CaO or SiO_2 as an internal standard component. The external standard was re-analysed every 10-15 samples and interpolated to correct for drift.

3.5. Extraction of the organic compounds from the dust sample

[13] A sub-sample of the Canberra dust has been extracted with a mixture of methanol, dichloromethane and phosphate buffer at pH 7.4 (2:1:0.8; v/v) [*Sturt et al.*, 2004]. After sonication for 5–10 min, further dichloromethane and buffer were added to the mixture to achieve a final methanol/dichloromethane/buffer ratio of 1:1:0.8. The phases were separated and the extraction repeated three more times. The total lipid extract (TLE) was reacted with BSTFA in pyridine to obtain trimethylsilyl (TMS) derivatives. The derivatized TLE was analyzed on a Thermoquest Trace GC-MS (Supelco SLB-5 ms column, 1 ml/min constant He carrier gas flow, temperature program: 60°C-10°C/min-150°C-4°C/min-320°C with 40 min holding time). Compound identification is based on published mass spectra and retention

times. Compound-specific stable carbon isotope ($\delta^{13}C$) analyses were conducted on a Finnigan MAT 252 GC-irm-MS at the University of Bremen, using the same capillary column and GC conditions as described above. Reported $\delta^{13}C$ values were corrected for the introduction of additional C during the preparation of TMS derivatives.

3.6. Molecular analysis of the dust sample

[14] The microbial communities within the dust sample were investigated by cloning of 16S rRNA genes. DNA was extracted from the dust sub-sample (ca. 300-500 mg) following the procedure established in *Abed and Garcia-Pichel* [2001]. For cloning, the 16S rRNA genes were amplified by PCR using the universal primers GM3F and GM4R (*Muyzer et al.,* 1995). The amplification was performed at an annealing temperature of 42 °C. The PCR products were purified using the QIA quick PCR purification kit (Qiagen, Hilden, Germany) and were cloned using the TOPO TA Cloning Kit (Invitrogen, Karlsruhe, Germany) according to the manufacturer`s instructions. The obtained clones were screened for the presence of inserts and the positive clones were then sequenced.

[15] The obtained sequences of the clones were analysed and compared using the ARB program [*Ludwig et al.,* 1998]. The alignment was corrected manually. Phylogenetic trees were calculated by maximum parsimony based on long 16S rRNA sequences (more than 1300 bp). The obtained sequences were then inserted into the reconstructed tree by applying parsimony criteria, without allowing changes in the overall topology.

[16] For determining the number of operational taxonomic units (OTUs), similarity matrices among the sequences were calculated with the ARB program. One OTU was defined for sequences which have more than 97% similarity. Rarefaction curves were calculated using the freeware program aRarefactWin (available at http://www.uga.edu/~strata/software/Software.html). The coverage of the clone libraries, species richness, species evenness and Shannon-Weaver index of diversity were calculated as previously described in [*Good*, 1953; *Atlas and Bartha*, 1998; *Singleton et al*., 2001].

3.7. Pollen extraction from the dust

[17] The sample was prepared in the palynological laboratory at Monash University and was initially treated with warm 10% Na-pyrophosphate ($Na_4P_2O_7$) and sieved over a 210 and a 7

μm mesh. Organic material retained in the 7 μm mesh was isolated from the remaining inorganic fraction using heavy liquid separation with sodium polytungstate ($Na_46[H_2W_{12}O_40].H_2O$; s.g. 2.0, 20 min at 2000 rpm). The isolated material was then treated with acetolysis (9 parts $(CH_3CO)_2O$: 1 part H_2SO_4, 10 min). The remaining residues were dehydrated with ethanol (C_2H_5OH). Slides were mounted with glycerol ($C_3H_5(OH)_3$) and sealed with paraffin.

3.8. Satellite imagery of the October 2002 dust event

[18] To detect and map the presence of the plume over the Australian continent, it was decided that a Normalised Difference (D) dust-enhancing algorithm developed by Miller (2003) for the Moderate Resolution Imaging Spectroradiometer (MODIS) sensor should be used. Furthermore, a simple satellite imagery time-series of the plume was created by combining day and night-time thermal infrared imagery of the MODIS and Advanced Very High Resolution Spectrometer (AVHRR).

Dust Enhancement Algorithm

[19] The Miller-algorithm was developed at the Naval Research Laboratory (NRL) in Monterey, California, for United States military desert operations. The MODIS sensor is found on both of NASA's sun-synchronous Terra and Aqua satellite platforms orbiting the earth at 705 km altitude. MODIS has a cross-track scan width of 2330 km and records over 36 channels, 20 reflected solar channels ranging from 0.4 μm to 1.6 μm and 16 thermal emissive channels ranging from 3.6 μm to 14.4 μm. All channels are available at 1 km spatial resolution, with reflective channels 1 to 7 also available in their original format of 250 m (1 and 2), and 500 m (3 to 7). The Miller algorithm is primarily based on the 11 μm and 12 μm Thermal Infrared (TIR) channels, namely 31 and 32, at 1 km resolution and spectral bands 1, 3, 4 and 26.

[20] Two variants were developed; one to enhance the presence of dust over water (D*wat*) and one to enhance the presence of dust over land (D*land*). Of the two variants, only the latter was investigated in the context of the goals of this particular study. As described in Miller [2003], D*land* was developed using both Reflectance (R) and Brightness Temperature (T) channels of

the MODIS sensor. Thermal emissive values need to be converted to Brightness Temperature using an inverse Planck function (equation 1):

$$T = \left(\frac{hc}{k\lambda}\right)\frac{1}{\ln\left(2hc^2\lambda^{-5}L^{-1}+1\right)} \quad \textbf{[1]}$$

where L is the radiance (Watts/m^2/steradian/m), h is the Planck's constant (joule second), c is the speed of light in vacuum (m/s), k is the Boltzmann gas constant (joules/Kelvin), λ = band or detector centre wavelength (m), and T is the temperature (Kelvin). The Miller land algorithm is then applied as follows:

$$\mathbf{D}land = L1 + L3 - L4 + (1.0 - L2) \quad \textbf{[2]}$$

[21] The terms in equations 2 and 3 are defined in Table 1 with numbers in brackets identifying the different MODIS channels.

Table 1*: Terms for Over-Land Dust Enhancement [Miller, 2003]*

Term	Expression	Normalization Bounds
L1	T(32) – T(31)	-2 to 2 K
L2	T(31)	(31)[a] to T_{max}(31)
L3	2R(1) – R(3) – R(4) –L2	-1.5 to 0.25
L4	R(26) > 0.05 ?, else 1	(n/a)

[a]See equation 4

$$\mathbf{T_{dyn}}(31) = T_{max}(31) - 21 \textbf{ if } (T_{max}(31) < 301 \text{ K}), \textbf{ otherwise } (T_{max}(31) - 273)/4 + 273 \quad \textbf{[3]}$$

[22] The algorithm combines the principles that, "*1) elevated dust produces a depressed brightness temperature against the hot skin temperature of the land background, 2) this cool layer of dust can be differentiated from water clouds having the same radiometric*

temperature based on its coloration properties, and 3) mineral dust often produces a positive (and of opposite sign to cirrus) 12–11 mm difference" [Miller, 2003 p. 12-3]. For a full description of the Miller algorithm terms and parameterisation values refer to Miller (2003)

Satellite Imagery Time Series

[23] To create a high temporal resolution time-series of the progress of the plume, Advanced Very High Resolution Radiometer, (AVHRR) imagery was acquired to *fill the gaps* between MODIS overpasses. NOAA-AVHRR sensors have very similar geometric properties to MODIS but only record in 5 or 6 channels (depending on the age of the NOAA platform), 3 reflective bands and 2 or 3 thermal bands. Both the 1-km High Resolution Picture Transmission (HRPT) or Local Area Coverage (LAC) and 5 km Global Area Coverage, (GAC) imagery formats were acquired to increase to overall coverage time-series of the plume to approximately 12 day- and night-scenes in a 24-hour period.

[24] As solar reflective channels in both sensors are not operational at night, it was decided that the visual time series would be created using thermal infrared bands (operational during both daytime and night-time hours) common to both sensors. The imagery was therefore processed to derive the Brightness Temperature Difference (BTD – seen as L1 term in Miller algorithm) between the 11 μm and 12 μm channels present in both sensors (bands 31 and 32 in MODIS, 5 and 6 in AVHRR). As seen above, the depressed brightness temperature difference produced by dust in the imagery is vulnerable to water clouds with the same radiometric temperature if the *colouration properties* of dust in daytime solar reflective channels are not used. This limitation will be kept in mind when referring to the results of the visual time series.

Image Processing - MODIS

[25] MODIS Level 1B (calibrate radiance) 1-km resolution scenes matching the event were acquired from NASA's Earth Observing System (EOS) Data Gateway. NOAA-AVHRR HRPT/LAC and GAC scenes were acquired from the National Environment Satellite, Data, and Information Service (NESDIS) online archives. Both datasets were processed using Research Systems, Inc. (RSI) Environment for Visualizing Images (ENVI) software. The MODIS was geo-referenced using a Lambert Conformal Conic projection on the WGS-84 mapping datum while the AVHRR imagery was imagery was corrected for solar zenith angles and calibrated for Top of Atmosphere (ToA) percentage reflectance in Channels 1, 2 and 3,

and for Brightness Temperature in Kelvin for Channels 4 and 5 before being geo-referenced using a Lambert Conformal Conic projection on the WGS-84 mapping datum.

[26] The dust-enhancement stage of the satellite imagery analysis was carried by applying the Miller algorithm to the MODIS scenes. The day-night time-series was then created from the BTD of the 11 µm and 12 µm channels in both sensors. To simplify the visualisation of the BTD-derived time-series, the resultant images were scaled as for L1 in the Miller algorithm, whereby the [-2, 2] scaling was retained, but contrary to processing for daytime Miller D*land*, where normalisation between [0, 1] is required, the scenes for this study were displayed on a blue-red colour class scale matching the [-2, 2] histogram stretch.

3.9. Trajectory analysis and meteorology

[27] The NOAA HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Transport and Dispersion Model [*Draxler and Rolph*, 2003] available on the web at (http://www.arl.noaa.gov/ready/hysplit4.html) was used to develop trajectories for the airmass that was associated with the dust plume, part of which travelled over Canberra. The model uses the final run (FNL) archive of the National Centers for Environmental Prediction (NCEP) Global Data Assimilation System (GDAS) model output. Other relevant meteorological data were also derived from the FNL archived data set.

4. Results

4.1. Observation of dust particles and other material under the SEM

[28] A set of 6 scanning electron microphotographs showing the content of the dust that was entrained with the rain droplets during the night of 22/23 of October 2003 are presented in Figure 2. The grains clearly vary in size, and several of them consist of agglomerates of much smaller particles, many of which are angular to sub-angular. Over 50% of the mineral grains consist of quartz, a very common mineral in arid/desertic environments in Australia. This is confirmed by the colour of the sediment that is ‘light reddish brown’ [5YR 5/6 on the Munsell

soil colour chart], again a characteristic of much of the Australian interior. Up to 200 µm long organic fibres are clearly visible in most photographs (Figures 2A, B, D E) and are thought to originate from disaggregated faecal pellets of grazing animals. Charcoal particles are also present (Figure 2F). Despite thorough search of the stubs under the SEM, no pollen grains could be seen despite their richness recognised in another sub-sample that was specifically treated for pollen extraction. Another attempt was made to identify minute (eg. a few µm long) cellular particles typical of microbes, but none could be seen.

FIGURE 2 ABOUT HERE

4. 2. Grain-size analysis

[29] The grain-size analysis shows a relatively well sorted bimodal distribution with a dominant mode around ~26 µm and secondary mode around ~150 µm.

Classically, the grain-size distribution of a wind-blown deposit is unimodal, and very well sorted [e.g. *Krumbein and Pettijohn*, 1938]. However, in the case of the Canberra dust event, deposition of the dust resulted from both wet- and dry deposition, which explains the fine tail of the distribution. Due to the presentation of the grain-size distribution in volume percent, the clay is somewhat underestimated as its contribution to the total volume of the sample is relatively small, and therefore this size fraction appears as a tail rather than an individual peak. In addition, the clays in grain-size distributions analysed with a laser particle sizer are often underestimated due to the particle shape of clays [e.g. *Konert and Vandenberghe*, 1997].

[30] On the coarse end of the distribution, there is a small shoulder, which was also observed in an aeolian dust sample collected from the atmosphere over the Atlantic Ocean offshore Cameroon (Figure 3). This shoulder can be explained due to the shape of the mica minerals and other platy particles that are abundant in this sample (see Figure 2). Due to the shape of these particles they show the similar aerodynamic behaviour and will have more or less the same settling velocities as smaller spherical particles like quartz and feldspar minerals. The laser particle sizer, however, describes these platy particles on an equivalent-volume basis, and therefore their “true size” is larger than their “aerodynamic size”. Would the grain-size analyses have been carried out using an estimation of the size

based on settling of the particles, this additional shoulder would most likely not have been observed.

FIGURE 3 ABOUT HERE

4.3. Major, trace and rare-earth elemental composition of the Canberra dust and Sr and Nd isotopic ratios

Background information on the Murray Darling Basin

[31] *Gingele et al.* [2005] carried out an extensive geochemical sampling of the major rivers in the Murray Darling Basin [MDB]. The principle behind this project was that by sampling fluvial sediments, one would obtain an overview of the nature of the sediments in the entire catchment, knowing that each river transports material that is weathered in each sub-catchment of the basin, and that once they are joined, the fluvial sediments contain a combination of material from catchments upstream and that various proportions of supply at the junction of 2 rivers could be estimated downstream by comparing a selected group of elements and their isotopes such as Sr and Nd [for further details, refer to *Gingele et al.*, 2005]. In all cases, those authors only used the clay (<2µm) fraction for all their analyses and they were able to discern trace and rare earth elemental characteristics that relate to local source rocks. In particular, as various tributaries of the Murray and Darling Rivers originate in geological formations of different age and composition, significant differences in the respective mineralogical, chemical and isotopic signatures occur between the two river systems. Of note is that the Darling River system is characterised by abundant smectite, low Ba, K, Rb, Cs, Sn, and high Hf and Zr concentrations, low relatively constant $^{87}Sr/^{86}Sr$ (0.708 – 0.717) and wide range of eNd(0) values (1.4 to –6.2). In contrast, the Murray River system shows high illite values, high Ba, K, Rb, Cs and Sn concentrations, low Hf and Zr concentrations, a wide range of $^{87}Sr/^{86}Sr$ (0.726 – 0.775) and relatively constant eNd(0) values (–8.9 to –10.3) [*Gingele et al.*, 2005]. This crucial data set has been used here to identify the possible source(s) of the airborne dust that 'rained down' in Canberra in October 2002. The geochemical data for this Canberra dust are available in Appendix 1.1.

FIGURE 4 ABOUT HERE

Comparison of all elements and some isotopes from the Canberra dust and Murray Darling Basin fluvial sediments

[32] Principal component analysis was performed using the CANOCO 4.0 program [**CANO**nical **CO**mmunity ordination by correspondence analysis; see *ter Braak*, 1986; *ter Braak and Smilauer*, 1998] to determine the close similarity of the Canberra dust against the fluvial samples from 32 sites in the MDB [sample GVC4 from the Onkaparinga River was excluded here]. In total, 47 parameters were compared [major, traces and rare earth as well as Sr and Nd isotopic ratios]. Several sites plot in close vicinity as shown for the 2 principal axes [see Figure 5], and all appear to be originating from the Darling River in the

Some elemental ratios

[33] Bi-plots used for comparing 2 elements from the MDB and Canberra dust data set were created with the aim of identifying similarities between the Canberra dust and some of the 32 sites from the MDB. Only a few elements would show some similarity between the Canberra dust and several of the sites in the MDB. Nevertheless, we attempted to compare elements such as Ba versus K, Hf versus Sr, Ti versus Zr and Rb versus Cs, and none of the first 3 ratios, for example, showed any similarity between the Canberra Dust and any of the sites in the MDB. Only the last comparison of the 2 elements Rb and Cs shows some obvious coincidence as displayed in Figure 6 with several sites in the Darling catchment and one from the Murray [the Macquarie River] which, in fact, is in close vicinity to several of the tributaries of the River Darling.

FIGURE 5 and 6 ABOUT HERE

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Insoluble lanthanides

[34] We have also attempted to normalise [by dividing the value for each element for any MDB site by the value for the corresponding element in the Canberra dust] all the elements for all the fluvial samples in the MBD, including the one from the Onkaparinga River [GVC-4], and only the insoluble lanthanides [Pr, Sm, Gd and Dy] demonstrated a level of

closeness with the Canberra dust. It is no surprise that insoluble REEs are proving to be those that can be used for fingerprinting the origin of the Canberra dust. Those are presented in Figure 7. Three other elements (Y, La and Ce) when normalised to Canberra dust also show closeness to the Canberra dust for site MD-9 in the MDB, but Nd did not; however, the combination of Nd and Sr isotopes provide a better way of fingerprinting the fluvial samples [see section below] for comparison with the Canberra dust.

FIGURE 7 ABOUT HERE

Sr and Nd isotopic ratios

[35] *Gingele et al.* [2005] have already successfully used the isotopic ratios of Sr [$^{87}Sr/^{86}Sr$] and Nd [$^{143}Nd/^{144}Nd$] to fingerprint the fluvial sediments in the Murray Darling Basin [same locations shown in figure 1] and have also applied this method to identify the origin of riverine clays extracted in a deep-sea core located offshore the mouth of the River Murray [*Gingele et al.*, 2007]. We have used the same approach with the analysis of the Canberra dust concerning the Sr and Nd isotopes and once again the plot shown in figure 8 identifies that the samples plot close to the cluster of several of the tributaries of the Darling River, but even closer to 2 samples from Murray tributaries [MD-1: Macquarie River and MD-9: Bogan River] display close affinity with the Canberra dust. Again, those 2 latter samples are in close vicinity of the Darling. We also benefited from the analysis of 3 samples of aeolian dust collected from Central and Western Australia and these are very distinct from the Canberra dust and other samples from the MDB. This is easily explained by the much older geologies for those 3 additional dust samples [A, B, C; see figure 1].

FIGURE 8 ABOUT HERE

4. 4. Organic composition of the dust sample

a. GC-amenable compounds

493 [36] All identified lipid compounds in the Canberra dust sample are listed in Table 2 and
494 indicated in figure 9. The most abundant identified compound in the GC-MS trace is a
495 methyl-polyaromatic hydrocarbon (PAH) of with molecular weight of 242 *da* and a $\delta^{13}C$
496 value of -27.1‰ VPDB. Short-chain C_{12} – C_{18} *n*-alkanoic acids are present in moderate
497 amounts as saturated and mono-unsaturated homologues with even-carbon-number
498 predominance. They range in $\delta^{13}C$ values from -25.6 to -29.1‰ VPDB. *n*-C_{18}-alkanol is
499 present in small amounts, with a $\delta^{13}C$ value of -28.8‰ VPDB. Several N-bearing compounds
500 have tentatively been identified, e.g., *n*-$C_{22:1}$ nitrile and *n*-$C_{18:1}$ amide. Retene and
501 dihydroabietic acid ($\delta^{13}C$ = -30.3‰ VPDB) are present in small amounts, as are cholesterol
502 and β-sitosterol. Two tentatively identified triterpenoid compounds with unconstrained
503 functional groups are present in trace amounts. Several phthalate compounds are present in
504 the sample. No long-chain (>C_{20}) *n*-alkanes, *n*-alkanols and *n*-alkanoic acids were detected.
505 Markers for contributions from fossil fuel and petroleum, such as extended hopanes, are
506 absent. Also, water-soluble compounds, such as sugars, are not present in the investigated
507 dust extract likely due to the employed extraction scheme.
508
509
510 **TABLE 2 ABOUT HERE**
511 **FIGURE 9 ABOUT HERE**
512
513
514 *b. Intact polar membrane lipids*
515 [37] Due to their relatively short turnover time [cf. *White et al.*, 1979], intact membrane lipids
516 are considered to be representative of fresh biomass present in environmental samples [*Sturt*
517 *et al.*, 2004; *Biddle et al.*, 2006; *Lipp et al.*, 2008]. To date, no analysis of this compound
518 class has been reported. The applied protocol [*Sturt et al.*, 2004] is unselective and detects
519 molecular signals from all three domains of life, Archaea, Bacteria, and Eukarya. The
520 distribution of intact polar lipids registered in the Canberra dust is extremely simple (Fig. 10)
521 and consistent with predominant contribution of plant debris to the fresh biomass transported
522 with the dust. The two main compounds are phosphatidylcholine (PC) and
523 phosphatidylethanolamine (PE). Their combined concentration in the dust sample amounts to
524 about 430 ppm. Monounsaturated C_{16} and C_{18} are the most abundant acyl moieties in both PE
525 and PC, which is consistent with the distribution of free fatty acids (Fig. 9, Table 2).

FIGURE 10 ABOUT HERE

4.5. Microbiology

[38] DNA was successfully extracted and a total of 48 clones were sequenced and phylogenetically analysed to obtain a measure of bacterial diversity. The calculated rarefaction curve showed that the clone library was far from saturation with a homologous coverage of ca. 19%. The diversity seems to be high, as most of the randomly picked clones were different from each other. The obtained sequences were distributed among 39 operational taxonomic unit OTUs and belonged mainly to the groups, *Proteobacteria* (12 clones), *Bacteriodetes* (11 clones), and *Firmcutes* (11 clones). The remaining fell into the groups *Planctomycetes* (5 clones), *Acidobacteria* (6 clones) and *Cyanobacteria* (1 clone). The single obtained cyanobacterial sequence is related to a cyanobacterium that has been associated with corals, some of which are healthy, but also those that suffer from the blank-band disease, as well as dead corals [*Shinn et al.,* 2000, *Shinn,* 2000]. Many of the affiliates to cultured species originate from soils or desert crusts, usually first isolated at large distances from Australia.

FIGURE 11 ABOUT HERE

4.6. Pollen content of the dust sample

[39] The material extracted from the Canberra dust sub-sample was very rich in pollen [see Table 3]; one transect under high magnification was enough to reach a high pollen sum. The dryland pollen spectrum is made up of an almost equal number of tree and shrub, and herbaceous and woody herbaceous pollen. Tree and shrub pollen are dominated by *Callitris* and *Eucalyptus*-type pollen with a minor component of *Acacia* pollen, while Amaranthaceae/Chenopodiaceae, Poaceae, Asteraceae, Tubuliflorae and Leguminosae make up the bulk of the herbaceous and woody herbaceous pollen (see Appendix 2). In addition, several aquatic taxa are present making up about 5.1% of the total pollen count. Also notable is the presence of fungal spores of the *Sporormiella* type; spores of this type are associated with ascomycete fungi found on the dung of herbivores and are common on the dung of large

herbivores (Davis and Shafer, 2006). Charcoal particles are abundant, with a value of 39.3 when expressed as percentage of the total pollen sum. Of interest also is the presence of *Pinus* pollen which is an introduced genus to Australia but which would not normally be associated with a pollen spectrum of this composition. It is noteworthy that in the Canberra region there were a substantial number of pine forests [*Pinus radiata*] which existed at the time of the dust event, but which have almost all been destroyed by the devastating fires of January 2003. It is, therefore, of no surprise to find pine pollen as a producer of large amounts of pollen that can travel long distances due to its 2 globular sacs.

FIGURE 12 ABOUT HERE

4.7. Satellite imageries

Dust enhancement

[40] Figure 13a (A) shows the plume as it was presented by *McTainsh et al.* [2005] in a true-colour SeaWiFS image at 09:00 Eastern Standard Time (EST) on the 23rd October 2002. Scenes B and C are colour-optimised Red-Green-Blue (B) and Miller-algorithm (C) variants of the same plume as seen by MODIS at 13:25 EST. In both scenes the plume is clearly differentiable from the land, sea and cloud pixels. The smoke plumes referred to in the McTainsh et al. (2005) image (A) are also clearly visible in the B and C scenes.

[41] However, the MODIS sensor had recorded several more scenes before and between the 09:00 and 13:25 overpasses. Figure 13b (A) shows a night-time MODIS scene of the southern Lake Eyre Basin region at 23:50 EST on the 22nd October 2002. The BTD depression associated to the passage of the cold front is clearly visible over NSW and southwest Queensland while dust plumes can be seen rising from the Lake Callabonna region, the Lake Frome islands and the southern Strzelecki Desert between Lake Frome and the township of Broken Hill. Figure 13b (B and C) shows the plume at 10:50 EST on the 23rd October 2002 from a MODIS overpass centred on the Lake Eyre Basin. B is a colour variant of the Miller-algorithm while C is the monochromatic variant of the same scene. The colour variant (B) shows how cloud cover (white) is mostly concentrated to the south of the Australian continent while the dust plumes are clearly identifiable in red tones over the darker background of the desert floor. With knowledge of the position of cloud cover from Scene B, the monochromatic variant (C) of the Miller-processed imagery was well suited to identifying linear dust plumes in white and their sources (arrows) against the dark (black) background of the desert floor.

This imagery revealed further NSW sources in the Darling River region between Menindee and Wilcannia and pastoral land between Hay and Griffith. Some linear plume-like features are also visible at the back of the main plume over the NSW-QLD border at longitude 148^{o}E although these may be truncated plumes not connected to the ground. Plume-like features were also seen from western regions of the Northern Territory right through central Queensland, highlighting regions such as the Davenport Ranges northeast floodplain, the Dulcie Ranges northeast floodplain, along with the Ranken, Georgina, McKinlay, Diamantina, Mayne, Vergemont, Thompson, Barcoo, Paterson and Culgoa river floodplains.

[42] Figure 13c is a selection of the time-series and is derived from MODIS and AVHRR-GAC scenes. This sequence shows early evidence of a BTD depression over NSW and SA (**A** – MODIS) at 23:50 EST on the 22nd, (see Scene A of Fig. 13b) with dust plumes rising from the southern Strzelecki Desert between Lake Frome and the town of Broken Hill; **B**(MODIS) shows the plume further into NSW at 01:20 EST on the 23rd: **C** (AVHRR-GAC) shows that the fully visible plume primarily over NSW and parts of southern Queensland at 9:30 EST on the 23rd; **D** (AVHRR-GAC) shows the plume *signature* at its largest (in the series) at 12:20 EST on the 23rd; **E** and **F** (MODIS) show the plume saddling the southern Queensland coast at 22:50 EST on the 23rd and 00:25 EST on the 24th, respectively; **G** (MODIS) shows that the plume to have increased in length and is extended well over the Great Barrier Reef and into the Tasman Sea by 09:50 EST on the 24th.

FIGURES 13 a, b, c ABOUT HERE

4.8. Trajectory analysis of the plume that brought dust to Canberra on October, 22/23, 2002

[42] *McTainsh et al.,* [2005] have shown previously that this dust storm was associated with a rapidly moving cold front and was approximately 2,400 km long (aligned northwest-southeast along the frontal axis), 400 km wide, and constrained below 1.5-2.5 km above ground level (AGL) underneath a subsidence inversion. Figure 14 shows the cold front-trough system as it passed through Canberra at 04 EST on October 23.

FIGURE 14 ABOUT HERE

[43] Figure 15 is a composite diagram showing the meteorological conditions associated with the dust transport to Canberra. Figure 15(a) shows forward atmospheric trajectories originating from the lower boundary layer (200 m AGL) at 31 deg. S and 145 deg. E (very close to the town of Bourke). The plots were produced at 12 hourly intervals from 00 UTC (10 EST) on 20 October using the NOAA HYSPLIT Transport and Dispersion Model [*Draxler and Rolph*, 2003]. The trajectory plots show that early in the period transport from the Bourke region was generally towards the north under a relatively light southerly wind regime. The near-surface meteorology plots for Bourke [Figure 15(b)] show that this southerly flow continued until 18 UTC on 21 October (04 EST on 22 October). From 00 UTC (10 EST) on 22 October until the passage of the front at about 18 UTC northerly winds dominated. This northerly flow was associated with high temperatures and very low humidities. No rainfall was associated with the frontal passage through Bourke, assisting with dust entrainment. The trajectory plot for 00 UTC (10 EST) on 22 October (Figure 15(a)) shows clearly that air originating in the near-surface layer at Bourke was advected directly over Canberra (35.18° S, 148.11° E) at a height of close to 1000 m AGL some 18 hours later. This equates to an average atmospheric transport speed of ~40 km hr over the 700 km distance. Precipitation in Canberra associated with the pre-frontal cloud band that began at or before 18 UTC (Figure 15(c)) produced the dust washout over Canberra. The archived upper air sounding for Canberra (Figure 15(d)) confirms that the cloud base was at about 550 hpa (~3,500 m AGL) and that the atmosphere was saturated and precipitating above that level.

FIGURE 15 ABOUT HERE

5. Discussion

5.1. Grain sizes recovered from the Canberra dust

[44] The dominant mode of the Canberra dust sample compares very well with the two aeolian dust samples that were collected offshore Africa, and which are estimated to be transported over several 100's km, which is comparable to the source (Bourke) to sink (Canberra) distance of the Canberra dust sample. In addition, the dominant mode compares

very well with similar dust studies carried out on proximal dust collected relatively close to the source area on land [e.g., *McTainsh et al.*, 1997].

[45] Since the size of the dust particles is not only related to horizontal distance but also to vertical transport through the atmosphere, it cannot be expressed in absolute wind speeds [*Stuut et al.*, 2005]. In addition, the dust particles that were deposited during the Canberra dust event resulted not only from dry deposition but also from wet deposition by rain droplets that formed just above the dust.

5.2. Source of the Canberra dust based on its inorganic composition

[46] It has been fortuitous that *Gingele et al.* (2005) carried out a geochemical survey of the Murray Darling Basin, although on a grand scale with only 32 sampling stations for an area covering some 1.10^6 km^2**.** Nevertheless, the concept these authors used was that fluvial clays, if suitably chosen, can represent an 'overview' of the clays that will have originated within the catchment of a river. We consider that there may be some additional aeolian component in rivers, but this amount today is minimal compared to what is being produced from the surrounding weathered lithologies. The fortunate aspect also is that the River Murray and the Darling River are draining very different lithologies and also of different ages [for further details, refer to *Gingele et al.*, 2005]. These differences are noticeable with a selection of particular elements and ratios of some of them. For example, the insoluble lanthanides, the ratio of Cs to Rb and Sr and Nd isotopic ratios, all clearly point to a source in the vicinity of Bourke to Cobar in western NSW in the proximity of the Darling River and some of its tributaries [see Figure 1]. This is further confirmed by pooling all the elements [major, trace and rare earths] together with the isotopic ratios of Sr and Nd, in conducting a correspondence analysis of all the data against the Canberra dust, and this further confirms the location of the dust source, in this particular case, close to Bourke [see figure 5].

5.3. Sources of organic compounds in the Canberra dust

a. GC-amenable compounds

[47] The short-chain (C_{12} – C_{18}) *n*-alkanoic acids and alkanols are non-specific biogenic markers forming basic units for all plant fats, oils and lipids [*Simoneit and Mazurek*, 1982].

691 The high abundance of the *n*-alkenoic acids, e.g., *n*-$C_{16:1}$ and *n*-$C_{18:1}$, points to a recent
692 biosynthetic origin, as these compounds are rapidly oxidized in the atmosphere [*Kawamura*
693 *and Gagosian,* 1987]. Their $\delta^{13}C$ values indicate a predominant C_3 plant origin. The absence
694 of long-chain, i.e., $>C_{20}$, *n*-alkanes, *n*-alkanols and *n*-alkanoic acids in the sample imply a
695 negligible contribution from plant waxes [*Eglinton and Hamilton,* 1967], which are normally
696 found in high abundance in aerosols from grasslands or forest areas with leaf-bearing trees
697 [*Schefuß et al.,* 2003]. Most polyaromatic hydrocarbons [=PAHs] are common constituents of
698 combustion emissions and are not source specific. The sole occurrence of the abundant alkyl
699 PAH in this sample, however, points to biomass instead of fossil fuel burning. Alkyl PAHs
700 were detected in relatively high concentrations in pine wood smoke [*Simoneit and Elias,*
701 2000]. The absence of other fossil fuel markers, i.e., extended hopanes and steranes, supports
702 this conclusion. The $\delta^{13}C$ value of the alkylated PAH suggests a C_3 plant source. The
703 occurrence of retene, proposed as atmospheric tracer for conifer burning [*Rahmdahl,* 1983],
704 further points to conifer wood as a source of the thermal combustion products. This
705 suggestion is additionally supported by the occurrence of dehydroabietic acid, a molecular
706 marker for softwood burning [*Simoneit,* 1999]. In that respect also the relative high
707 abundances of N-bearing compounds in the sample are of interest, as their occurrence in
708 aerosols has been inferred to indicate burning of nitrogenous biomass (*Bin Abas et al.*, 2004).
709 Also β-sitosterol, although present in all vegetation types and thus not source-specific, is often
710 used as an indicator for direct emission from plants during biomass burning processes
711 [*Simoneit et al.,* 2004]. The two triterpenoids in the sample are likely degradation products of
712 major triterpenoids found in angiosperms [*Simoneit,* 2002], whereas cholesterol either
713 indicates an input from microbial sources or from cooking activity in urban areas [e.g., Rogge
714 et al., 1991]. The abundant phthalates in the sample might indicate emissions from burning of
715 urban refuse, as they are major constituents of plastics. We can, however, not rule out a
716 potential contamination from the sample container.
717 [48] Based on these inferences on lipid sources, the compounds have been summed up to a
718 source apportionment based on quantifications of their total ion currents (TIC). These are: (a)
719 general vegetation, (b) angiosperm vegetation, (c) biomass burning, subdivided into conifer
720 fuel, where a distinction was possible, and (d) urban burning and cooking or contamination
721 (Figure 16).
722 [49] The inference of biomass through the identification of specific compounds is of no
723 surprise as examination of satellite images of the dust plume showed that it travelled through

several smoke plumes, as at the time of the 'dust event', SE Australia was affected by several bush fires.

FIGURE 16 ABOUT HERE

[50] Compound 1 [Table 2, general vegetation in figure 16] has a fragmentation pattern typical of resins from softwood and, therefore, would fall into the category of conifer-derived markers. The same applies to the absence of plant waxes indicative of leaf-bearing vegetation. Conifer needles do not contain many wax compounds. *Callitris*, being a gymnosperm, belongs to this category. In addition, markers for plant combustion such as retene and dihydroabietic acid (plus the unidentified resin product), point to the presence of gymnosperms. No lipid trace of cyanobacteria such as n-heptadecane or 2-methyl-hopanoids were detected.

[51] Compounds diagnostic of grass waxes play only a minor role in the extractable organic matter associated with the dust consistent with negligible input of grass-derived biomass. Surprisingly, pollen suggest a sizeable contribution of particles originating from grasslands to the dust. This may indicate that (a) transport mechanism of grass pollen and wax compounds differ and/or (b) the wax signal in the extractable organic matter is rapidly overwhelmed by compounds from other sources, incl. plant combustion, anthropogenic processes, microbial matter sourced from desert crusts, and plant debris not associated with waxes.

b. Intact membrane lipids

[52] Notably, the concentration of intact membrane lipids is with 440 ppm rather high, suggesting a significant contribution of fresh biomass to the dust. Although the two major lipids are known constituents in bacteria, the simplicity of the distribution and the high predominance of PC are consistent with a predominant origin of these membrane lipids from plants and fungi. PC is the major membrane lipid in eukaryotic cells and only present in a relatively small fraction of bacteria (cf. Sohlenkamp et al., 2003). PE is widely distributed in all domains of life but its presence in the dust sample is probably reflective of fresh cellular debris from higher land plants in which it is a major compound type (Lerch and Stegemann, 1966). Compounds typical for bacterial membrane constituents, e.g., phosphatidylglycerol or glycolipids, both commonly found in cyanobacteria (cf. Hölzl and Dörmann, 2007), are not detected under the analytical conditions applied. Intact membrane lipids of Archaea and Bacteria, possibly derived from soils and crusts, are likely to be present in the dust but

presumably at concentration levels that are orders of magnitude lower than those of plant derived analogues.

5.4. Source of the Canberra dust based on its palynoflora

[53] The potential pollen source area around Cobar in western New South Wales [see figure 1] is envisaged as it contains the following main vegetation types.:Open *Eucalyptus* woodland with a lower stratum of *Acacia* and *Callitris*; open mixed *Eucalyptus* and *Callitris* woodland with grasses; open low *Acacia* woodland with a mixed lower stratum and smaller areas of open mixed *Eucalyptus* and *Callitris* forest and of *Eucalyptus* shrubland with a mixed lower stratum. The presence of *Pinus* pollen is really significant and is considered to point to some additional pollen being taken by the dust when it was over the Canberra region where extensive pine forests occurred until recently. (Note that *Pinus* is a prolific pollen producer and its pollen can travel long distances).

5.5. Microbiology of the Canberra dust

[54] The obtained clones in our gene library contained almost full 16S rRNA genes, thus reflecting only living bacterial populations. A comparison with culture-based techniques, showed that the diversity in the dust sample has been strongly underestimated. The dust study of *Griffin et al.* [2001a] reported 5 species in one dust event, and 10 species in a second dust event (of which 4 were identical to the first samples). In a second study, *Griffin et al.* [2003] reported 19 bacterial isolates from various dust samples. In our DNA-based approach, the clones related to cultivable strains formed only a fraction of the total number of clones while the rest could not be related to known strains.

[55] The number of recovered clones does not represent the full diversity, the rarefaction curve indicated only ca. 20% coverage, thus more sequencing is needed. In spite of that, the Canberra dust appears rich in *novel micro-organisms* because more than 50% of sequences had a 8-12% sequence divergence to their closest relative. The very high diversity can be attributed to the large area from which the dust originates, including a wide diversity of habitats. It is, however, unlikely that the diversity in dust reflects the diversity from the original habitats. The transport by dust will have selected microorganisms resistant to desiccation, oxygen, spore and radiation, and select for attached cells. Many of the obtained

clones were related to clones from soil or soil crust origin (Fig. 11). It is understandable, therefore, that microorganisms associated with soils and soil crusts are strongly represented in dust. In addition, a strongly-represented group are the *Actinobacter,* which harbour a large number of spore-forming pathogens. Some of these originate from extreme environments (eg. hypersaline, thermal and arid Antarctic soils) [*Stackebrandt and Schumann,* 2006]**.** On the other hand, no known pathogens were identified and this is not surprising as they are rare in most environments. It is possible, nevertheless, that the rhizosphere associates (Fig. 1 tree, e.g. clones D5, 6, 9, 36, 48 and 54) have pathogenic properties for plants [*Coombs and Franco*, 2003]**.**

[56] Finally, from the clones with the least known relations, one is considered to be a cyanobacterium associated with Black Band disease in corals [*Shinn et al*., 2000, *Shinn,* 2000]. The less than 95% related clones are related to strains from North America, Asia, Africa and Antarctica, and therefore could be considered to be cosmopolitan in distribution. We might, therefore, conclude (although cautiously at this stage) that Australia is not isolated from the rest of the world, from a microbial point of view at least!

5.6. Meteorological conditions associated with the dust event

[57] The multi-proxy evidence of an origin near Bourke, New South Wales, for the dust deposited on Canberra overnight on 22-23 October, is unequivocally supported by the available meteorological and satellite remote sensing data. The cold front of 22-23 October was relatively intense and was associated with a parent low pressure system well to the south of the continent, and a trough extending well into the tropics (Figure 14). Such intense, fast-moving and geographically extensive front-trough systems are characteristic of the Australian continent in spring, when strong thermal contrasts across the land mass support intense frontogenesis well away from parent depressions in the Southern Ocean [*Sturman and Tapper,* 2005]. Many of the continent's most intense dust storms are associated with such features [*McTainsh et al.,* 2005].

[58] It is worth noting here that while the frontal progression and apparent movement of the body of the plume is towards the north-east (refer Figure 13), air motion ahead of and (at higher levels in the atmosphere) just behind the front is at right angles to the movement of the front (see *Sturman and Tapper,* 2005 for a detailed explanation of frontal structure in the Australian region). It is this motion that is moving dust in this instance toward the south-east, from the north-west of New South Wales towards Canberra.

826
827
828 **5.7. Satellite imageries of use to determine conditions of the dust plume**
829
830 [59] For this particular event, the application of the dust-enhancement algorithm developed by
831 *Miller* [2003] proved quite useful at identifying dust plumes as early as the night of October
832 22nd 2003. Although *McTainsh et al.* [2005] report the first signs of dust on the ground in
833 northern Victoria and western NSW in the evening of the 22nd, the first available imagery
834 with any visible signs of plumes was a 20:35 EST AVHRR-GAC scene (not shown here). At
835 that time northern Victoria and western NSW were under cloud cover associated to the front
836 and the imagery could only identify a brightness temperature difference depression north of
837 the cloud cover, most probably related to changes in atmospheric water-vapour conditions as
838 cold southerly air passes over the region. Although it is unclear if the *front-like feature* visible
839 in Figure 13b/c (A) and 13c (B) is related to dust, cold air or both, dust plumes were clearly
840 identified feeding into the back of it whenever there was no cloud to obstruct visibility. As the
841 event progressed it separated from the cloud line in the southern regions of the Australian
842 continent and could then be tracked quite easily in both fully Miller-processed and the more
843 limited BTD-processed imagery. In daytime imagery, while the plume was still somewhat
844 covered by cloud, the colour variant of the Miller-algorithm proved very useful at
845 differentiating between dust in red tones while leaving clouds in white. However, for plume
846 and source-identification purposes, the monochromatic variant of the Miller-algorithm was
847 found to be more useful as the white linearity of the plumes was easily identifiable against the
848 much darkened terrestrial background of the desert floor. The time series based on the BTD of
849 the 11 μm and 12 μm thermal infrared channels of both the MODIS and AVHRR sensors was
850 only useful after the position of the plume and cloud had been determined from the Miller-
851 processed imagery and because the event was quite large. Based on the limitations associated
852 to this technique described above, its use is only recommended for events for which the
853 presence of a plume has been established. It is unknown how this approach would work for
854 smaller events or deflation under more extensive cloud cover.
855
856
857 # 6. Conclusions
858

[60] We have successfully ‘fingerprinted’ the dust that fell over Canberra overnight in October 2002. The dust’s main provenance is along the semi-arid lands surrounding the Darling River, most likely in the vicinity of Bourke and Cobar. This is supported by a combination of trace element and REEs analyses and also Sr and Nd isotopes when compared with the database obtained by *Gingele et al.* [2005] for the Murray Darling Basin. The dust we studied here did not come from outside the Murray Darling Basin. The dust also consisted of a large spectrum of organic compounds, some of which confirm the nature of the vegetation such as is found along the Darling River. Evidence for biomass burning, mostly of softwood, is also detected and could have been taken by the dust plume on its journey to eastern Australia when it travelled though smoke produced by bush fires that proliferated in October 2002 as a result of long-lasting drought conditions. Contamination of the dust plume while in transit before falling over Canberra is further shown by the presence of *Pinus* pollen, an exotic taxon that is absent or extremely rare in the semi-arid interior of Australia. This indicates that the dust plume collected addition ‘material’ apart from at its source, and this is confirmed by the occurrence of plasticizer compounds and lipids ascribed to urban burning processes which point on a rapid overprint of aerosol lipid signatures in the urban sampling environment.

[61] The most surprising finding was the vast array of microbiobial taxa recognised in the sample. Many of the clones relate to organisms that have been recognised in arid regions such as in the western interior of North America, and also from as far away as Antarctica. Our findings do not imply that such arid locations are the provenance of the taxa found in the dust that originated along the Darling River, but that extreme environments on the planet do share many biological characteristics.

[62] Extensive examination and processing of MODIS satellite images demonstrate that the dust plume increased its load during its trajectory towards the Tasman Sea with some of the dust travelling as far as New Zealand and also delivering vast amount of dust to the Great Barrier Reef. This is particularly significant in light with the findings of *Shinn et al.* [2000], *Griffin et al.* [2001a] and *Garrison et al.* [2003] that microbes and fungi transported in dust plumes that originated in Africa do play a role in the demise of coral reefs in the Carribean.

[63] We will now apply many of the techniques listed in this study to fingerprint as many sites as possible in Australia that are potential sources of airborne dust. This is basic survey-type work, but it is not a trivial task. Australia is a vast continent, with diverse geologies, climatic conditions and associated vegetations. To know the composition of its regolith and the microbiota it nurtures and the organic imprints it leaves in this country of contrasting

conditions will help us better determine the impact of airborne dust on the health of our environment and eventually the oceans surrounding Australia where some of the dust eventually falls.

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Figure captions

Figure 1. Maps showing the location of the Murray Darling Basin in Australia and all the major tributaries of the 2 major Rivers Darling and Murray. The location of three dust samples from central and northwestern Australia that are compared against the dust sample originating from Canberra discussed in this paper are also shown.

Figure 2. Selection of scanning electron microphotographs of some of the particles that made the dust that fell over Canberra during the night of 22/23 of October 2002. The grey background consists of the double-sided sticky tape. Note the presence of long (>200 µm) fibres (A, B, D and E) and other organic remains (A, E) and charcoal (F). Note that photo B is an enlargement of image A to show the agglomeration of small particles on larger ones. Image E confirms the bimodality of the particles discussed in section 4.2.

Figure 3. Grain-size distribution of the Canberra dust, compared with the distributions of dust collected from the atmosphere above the Atlantic Ocean offshore Cameroon [*Stuut et al.*, 2005] and offshore Senegal [*Stuut*, unpublished results].

Figure 4. Map showing the major rivers in the Murray Darling Basin [MDB] and the location [red dots] of the fluvial samples studied by *Gingele et al.* [2005] for which geochemical characterisitics are compared against the Canberra dust that fell during the night of 22/23 of October 2002. Note the presence of an additional sample taken from the Onkaparinga River south of Adelaide and that is located outside the MDB catchment.

Figure 5. Diagram showing the close similarity of several samples [inside blue circle] from the Murray Darling Basin with the Canberra dust [blue dot] using the CANOCO 4.0 program [**CANO**nical **CO**mmunity ordination by correspondence analysis]. All elements [major, traces and rare earth as well as Sr and Nd isotopes] were used for comparison between all samples. Numbers refer to the location of the fluvial samples located on the map in Figure 5 and should be preceded with the MDB prefix. The Onkaparinga sample has been omitted from this figure.

Figure 6. Elemental bi-plot to show that the Rb versus Cs can be used to identify the similarity of the Canberra dust sample with several fluvial sites along in the Darling catchment, including one from Macquarie River [MDB-1]with is closely located to several of the Darling tributaries. Note that all other sites in the Murray catchment plot far away from the Canberra dust values.

Figure 7 shows all the composition of the Murray Darling Basin and Onkaparinga River samples with respect to the insoluble Lanthanides rare earth elements [Pr, Sm, Gd and Dy] that have all been normalised against the Canberra dust. Numbers with a MDB prefix represent samples originating from different parts of the Basin. For relevant location, refer to Figure 4. Arrows indicate 4 locations [MDB 2, 8, 10 and 11] for which normalised values for the 4 REEs are very similar to those of the Canberra dust.

Figure 8. Plot of $^{87}Sr/^{86}Sr$ versus $^{143}Nd/^{144}Nd$ isotopic ratios for the Canberra dust compared with the values obtained by *Gingele et al.* [2005] for fluvial samples from Darling River tributaries (red circles) and River Murray tributaries (empty black circles). Three additional aeolian samples from Central and Western Australia are shown [for their respective locations, refer to figure 1].

Figure 9. Total ion chromatogram of lipid compounds identified in the total lipid extract of Canberra dust, numbers refer to compounds listed in Table 2.

Figure 10. Positive ion base-peak chromatograms (upper panel) and density map (lower panel) from HPLC-MS analysis according to the protocol by *Sturt et al.* (2004) of intact polar membrane lipids in sample from Canberra dust event. Abbreviations: PE = phosphatidylethanolamine, PC = phosphatidylcholine, I.S. = internal standard: 1-O-hexadecyl-2-acetoyl-*sn*-glycero-3-phosphocholine.

Figure 11. 16S rRNA-based phylogenetic reconstruction based on maximum-parsimony methods, showing the affiliation of sequences obtained from the Canberra dust sample. The dust clone sequences were inserted into the reconstructed consensus tree by applying parsimony criteria without allowing changes in the overall tree topology. The tree was simplified for clarity by omitting all sequences between clusters. The bar indicates 10% sequence divergence.

Figure 12. Pie diagram to show the percentage distribution of the pollen in the Canberra dust sub-sample. Note that the largest percentage relates to woody taxa with almost equal amounts of herbaceous taxa and woody herbaceous taxa.

Figure 13a. (A) SeaWiFS satellite image 09:00 hours local eastern standard time (EST) on 23 October 2002 (taken from *McTainsh et al.,* 2005), showing the main dust plume passing over eastern Australia (note the smoke plumes from bushfires in south east Queensland and central east NSW, which indicate wind directions at the time of the event), (B): 13:25 hours EST – MODIS colour-optimised Red-Green-Blue image and (C) processed with the Miller Dust Enhancement Algorithm.

Figure 13b. (A) Monochromatic 11 μm and 12 μm channels BTD scene at 23:50 EST on the night of October 22nd 2002 showing plumes rising from the Lake Frome Basin. (B) full-colour and (C) monochromatic variants of the Miller algorithm showing the plume at 10:50 EST on October 23rd 2002, with plumes still rising from sources in SA, NSW, QLD and the NT. LF = Lake Frome, LC = Lake Callabonna region. Arrows indicate recognised dust tracks.

Figure 13c. MODIS-AVHRR time series based on the brightness temperature difference (BTD) of the 11 μm and 12 μm thermal infrared channels of both sensors.

Figure 14. Synoptic chart showing the cold front-trough system passing through Canberra at 04 EST on October 23 (18 UTC, October 22).

Figure 15. Composite diagram of the meteorological conditions associated with dust transport to Canberra, 22-23 October, 2002. (a) Forward atmospheric trajectories originating at 200 m AGL above 31° S, 145° E (12 hourly intervals from 00 UTC on October 20 until 12 UTC on October 22). Trajectories are each of 48 hours duration. (b) Meteorological data for 31° S, 145° E (near Bourke, New South Wales) for the period 06 UTC October 22 until 06 UTC October 23. (c) Meteorological data for Canberra, Australian Capital Territory (35.18° S, 148.11° E) for the period 00 UTC October 22 until 00 UTC October 24. (d) Upper air sounding data for Canberra, 18 UTC October 22. See the text for details of data sources.

Figure 16. Source apportionment of GC-amenable lipid compounds of Canberra dust sample.