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DUST FROM RURAL AUSTRALIA AS A POTENTIAL SOURCE OF PERSISTENT ORGANIC POLLUTANTS TO THE SOUTHERN HEMISPHERE

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ABSTRACT

The DustWatch program has monitored dust events and collected dust samples from across Australia since 1990 to monitor the extent and severity of wind erosion. A retrospective analysis of persistent organic pollutants (POPs) in 10 samples from the Buronga DustWatch Site was undertaken to understand contaminants in dust at this site, and their potential trends and sources over time. Although this study represents a small number of analysed samples, results suggest that dust may be an important vector for POPs in Australia. All families of POPs investigated were found in the samples, varying in both concentration and through time. Further investigation regarding the impact of dust containing POPs on receptors downwind is required and other samples in the DustWatch dataset (about 4,300 samples) should also be assessed for their potential to inform long-term trends of these chemicals in the environment and background concentrations at contaminated sites.

Keywords: Long-range atmospheric transport; background concentrations; PFAS; OCP; PCB; PBDE

INTRODUCTION

Long Range Atmospheric Transport (LRAT) across oceans and continents is a key characteristic of persistent organic pollutants (POPs) and occurs as a result of several well-known processes (Bidleman 1999). It moves POPs from pollution sources to the remotest areas of the globe, including the poles (Hung et al. 2010; 2016; Kallenborn et al. 2013) and remote mountain ranges (Arellano et al. 2011; Lavin and Hageman 2013), and can act as a vector for pollution that occurs in the deepest parts of the ocean (Jamieson et al. 2017). Atmospheric movement of chemicals from source to sink can occur via a number of processes. This includes dust LRAT (Nguyen et al. 2019) and global distillation, whereby POPs evaporate in tropical and temperate regions and precipitate in cold climates such as the Arctic and Antarctic (Bidleman 1999). Grasshoppering/global chromatography, whereby POPs undergo cycles of deposition and re-emission as they are transported from sources to sinks, can also play a role. The deposition of POPs in deserts and snow-pack as a result of these processes has the potential to create an important secondary source of these pollutants to people

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and environments, which are then exposed as a result of re-emission during dust events or snow-melt (Iwata et al. 1993; Bidleman 1999).

The Stockholm Convention's Global Monitoring Plan is an important tool for measuring global POPs transport (Kallenborn et al. 2013). Atmospheric transport of POPs is monitored as part of this plan, with gaseous phases contributing the majority of contaminant burden to the Earth's ecosystems (e.g. Iwata et al. 1993). There are only limited numbers of monitoring locations for atmospheric POPs in the Southern Hemisphere (Kallenborn et al. 2013). The DustWatch program has monitored dust events and collected dust samples from across Australia since the 1990s, with the aim of monitoring extent and severity of wind erosion (Leys et al. 2008). The longest-running station, called Buronga and located near Mildura, takes physical samples using a high-volume air sampler, as described in Love et al. (2019). These dust samples can potentially act as an historical record of atmospherically transported POPs. The Buronga site sits within the semi-arid south-eastern dust pathway of Australia, which is known to transport significant amounts of dust both within Australia and long distance to New Zealand and Antarctica (Lim et al. 2011; Marx et al. 2005; Love et al. 2019; Nguyen et al. 2019). This study presents a preliminary retrospective analysis of 10 dust samples collected between 1990 and 2018 from Buronga and aims to identify contaminants in dust at this site, and their potential trends and sources over time.

DustWatch Australia collects spatial and temporal patterns of dust activity across Australia through the use of multiple data collection methods, including Bureau of Meteorology (BoM) observations, citizen-science and an instrumented dust network, along with remote sensing and modelling approaches (Leys et al. 2008; Baddock et al. 2015). Dust samples collected by DustWatch at the Buronga long-term monitoring station, 6 km east of Mildura, were selected for analysis as part of this research (Figure 1). Mildura is a region of high agricultural productivity (Australian Bureau of Statistics 2017) and dust emission (O'Loingsigh et al. 2017). Dust from Mildura is therefore a potentially important source of legacy POPs, such as organochlorine pesticides, in the southern hemisphere (Marx et al. 2005).

Mean maximum summer temperatures in Mildura (BoM station 076031) are 37.1°C and mean winter minimum temperatures, 13.7°C (Australian Government Bureau of Meteorology 2018). Annual rainfall is 290 mm, with a maximum of 657 mm and minimum as low as 122 mm. The prevailing wind direction is from the south-west region, which is mainly used for dryland agriculture and irrigated horticulture. Strong dust-entraining winds in the Mildura area are predominately associated with frontal systems, which result in a shift in prevailing wind directions from north through west then southerly (Love et al. 2019). Sixty-six per cent of dust events come from the south-west quarter (Love et al. 2019). Dust collected in Mildura can come from distant sources, including the Lake Eyre rangeland region to the north-west, pastoral rangelands to the north, cropping lands to the
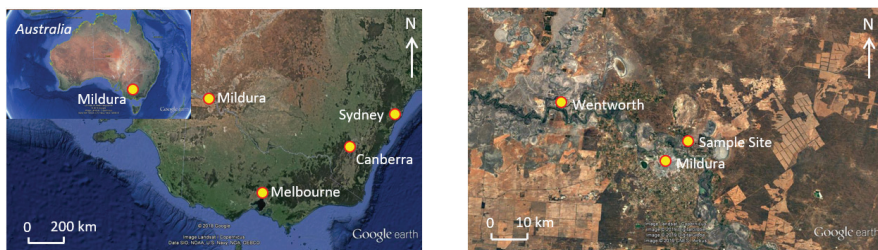

Figure 1. Location of sampling site within (a) Australia and south-eastern Australia; and (b) the Mildura area within which the Buronga sample site is located.

south-west and local agricultural production areas, including those under irrigation, surrounding the town (O'Loingsigh et al. 2017). There was an average of eight dust events per year when the dust concentration exceeded 100 μg/m^3 during this study period.

METHODS

Sample collection

Dust samples were collected by use of a high-volume air sampler (HiVol) installed at Buronga, New South Wales (34.17°S, 142.20°W). The advantage of a HiVol over real-time concentration monitors (e.g. DustTrak™) is that the total range of particles in the air are collected and that a physical sample is available for subsequent laboratory analyses. The HiVol draws air through a Whatman Glass micro-fibre filter (EPM 2000, 254 x 203 mm) at a rate of approximately 42 m^3 per hour. Dust sampling has occurred in this region for 29 years (1990 to present) through collaboration of various state government agencies and universities. The HiVol runs continuously 24 hours a day, seven days a week, except when filter papers are changed, which takes about five minutes. Total sample duration for each filter paper has changed through time, from weekly durations in the early 1990s to daily (24 hrs) since the 2000s. All sample times and weights are recorded allowing the resultant dust concentration (Dc) of each filter paper to be calculated as micro-grams per cubic metre (μg/m^3) using the formula (Equation 1):

$$Dc = Md/Ft \qquad (1)$$

where Dc is dust concentration (in μg/m^3); Md, net weight of dust on filter paper (in μg); and Ft, total flow of air through filter paper (in m^3).

Filter papers were desiccated and pre-weighed in the laboratory, sealed in plastic bags, and stored in sealed bags within the laboratory until ready for deployment. A batch of 30 to 60 filters was then transported to Buronga, where they remained in their bags within an airconditioned office ready for subsequent use. A filter paper was removed from its bag, placed in the sampling cradle and placed on the HiVol sampler during each sampling phase. Post sampling, each filter paper was returned to its original bag and stored in the airconditioned office until a batch was returned to the laboratory with no preservation for weighing. Handling of filters throughout this process involved the use of non-powdered latex gloves. Post weighing, filters in their original individual bags were boxed and stored at room temperature (i.e. in an air-conditioned room) within a university rolling stack with no preservation and were only removed for sub-sampling.

Sample selection

Ten dust samples were chosen for analysis. The filters were selected across the 28-year record between 1990 and 2018, based on representation across the 28-year record, dust storm seasonality and filter paper mass. This initial search was performed via desktop analysis of the database, followed by sample retrieval and visual confirmation.

Temporal representation across the 28-year record roughly aligns with the episodic nature of wind erosion. Big dust storm years produce events of larger sediment masses, resulting in the selected filters spread unevenly across the record. Seasonality was considered an important factor in filter paper selection. Heavier (dust laden) filter papers (0.2 g to 0.3 g) are generally associated with large dust storms and larger dust storms frequently occur between December and February in Mildura (Ekström et al. 2004). Eight of the 10 filters were from months between December and February and two from between September 2009 and 2011 (Figure 2). Some of the biggest dust storms in the area occur in September and October, but they are infrequent and associated with long-running droughts.
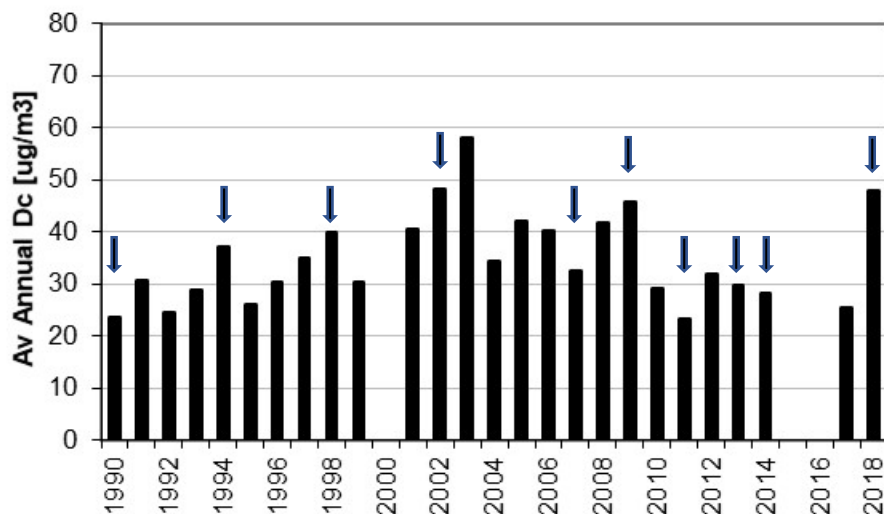
Figure 2. Average annual dust concentration (Dc) at Buronga. Arrows mark years in which samples presented in this paper were collected.

Selection of filter papers based on mass was critical to ensure sufficient sample for analysis. All selected filters had a similar mass, around 0.2 g to 0.3 g. Half of each filter paper was used for chemical analysis with the rest stored for further studies. The half filter paper was then cut into two quarters. One quarter was used for per- and poly-fluoroalkyl substance (PFAS) analysis and the other for analyses of the other 13 chemical groups (See next section, Chemical analyses).

Chemical analyses

A brief qualitative assessment of land-use surrounding Mildura and chemicals potentially associated with this land-use was undertaken as the basis for selecting chemicals for analysis within dust samples. Families of POPs associated with agricultural land-uses and listed on the Stockholm Convention were chosen, including the organochlorine pesticides aldrin, dieldrin, endrin, chlordane, endosulfan, Dacthal, hexachlorocyclohexane (HCH) and heptachlor, as well as dichlorodiphenyltrichloroethane (DDT) and its derivatives tetrachlorodiphenylethane (DDD) and dichlorodiphenyldichloroethane (DDE). Eight (PCB11, PCB28, PCB52, PCB101, PCB118, PCB153, PCB138 and PCB180) polychlorinated biphenyls (PCBs) were also selected from the industrial group of chemicals, due to their ubiquitous nature. Six poly-brominated diphenyl ethers [PBDE47, PBDE100, PBDE99, PBDE154, PBDE153, PBDE183 (PBDEs)] and 32 PFASs were also analysed as a means of assessing the potential influence of domestic chemicals used in Mildura on the surrounding environment (Table 1). It was expected that dust collected at Buronga would have multiple source areas including local and distant dusts. The chemicals selected for analysis were chosen based on potential local sources, but this does not imply that detected chemicals were locally derived.

Chemical analysis was undertaken at the Queensland Alliance for Environmental Health Sciences within the University of Queensland. Filters were stored in a laboratory desiccator overnight before being weighed at room temperature (25°C). Polychlorinated biphenyls, PBDEs and pesticides

were analysed on one quarter of the filter following methods described in He et al. (2017) and Wang X et al. (2017). Briefly, the filter was spiked with a solution containing multiple ^{13}C-labelled PCB congeners, ^{13}C-labelled pesticides and ^{13}C-PDBEs at different levels as internal standards for quantification purposes. Subsequently, these samples were extracted with an Accelerated Solvent Extractor (ASE, Thermo Scientific™ Dionex™ ASE™ 350) using a mixture of *n*-hexane and acetone (1:1, v:v) in 33 mL stainless steel vessels. The ASE conditions were set with pressure at 1500 psi; temperature at 100°C; static cycle time at 10 min; flush volume at 60%; purge time at 120 s; and number of cycles at 2.

Extracts were then concentrated to 1 mL by a gentle stream of purified nitrogen. The concentrated extracts were then cleaned up using a chromatographic column (from bottom to top) containing 4 g of neutral alumina, 2 g of neutral silica gel and 2 g of sodium sulfate. A mixture of *n*-hexane and DCM (42 mL; 1:1, v:v) was used to elute the samples from the column. Eluants were carefully blown down by a gentle stream of purified nitrogen to near dryness and reconstituted with 500 pg of ^{13}C$_{12}$-PCB 141 (in 50 µL of iso-octane). The latter was employed as the recovery/instrument standard for estimating the recoveries of the spiked internal standards and monitoring the performance of the analytical instrument. Samples were analysed using a TRACE GC Ultra coupled to a TSQ Quantum XLS triple quadrupole mass spectrometer equipped with a TriPlus Autosampler.

The other quarter of the filter was used for the analysis of PFAS. The filters were spiked with a solution containing multiple ^{13}C-labelled PFAS (Wang X et al. 2017) and placed into a Falcon tube. Five mL of acetone was added to each tube and the samples were extracted by shaking for 30 min and sonicating for 15 min. Samples were then centrifuged for 10 min at 3000 rpm (1847 x g). The supernatant was subsequently transferred to a new Falcon tube. The extraction process was repeated and the supernatants combined. Extracts were then evaporated and reconstituted into 1 mL of 99:1 (v:v) MeOH/NH$_4$OH. The extracts were then passed through an Envi-Carb SPE cartridge into a 2 mL LC vial. Another 500 µL of 99:1 (v:v) MeOH/NH$_4$OH was added onto the cartridge and the eluant was collected into the same LC vial. Envi-Carb SPE cartridge clean-up removes impurities from the samples by retaining them on graphitised carbon. These impurities/interferences/co-extracts have a potential to interfere with the later LC-MS/MS analysis. Examples of these impurities include aromatic organic compounds which are strongly associated with the graphitised carbon. In the meantime, any Π electrons in PFCAs (Perfluorinated carboxylic acids) are strongly associated with the highly electronegative fluorine atoms and therefore would not interact effectively with the sorbent (Powley et al. 2005). The analytes (PFAS), in a purified version, are therefore eluted and collected under the cartridge. The purified samples were analysed by liquid chromatography (Shimadzu, Nexera 2 UHPLC system, Kyoto, Japan) coupled with a tandem mass spectrometer equipped with an IonDrive source (SCIEX QTRAP® 6500+, Ontario, Canada) (Bräunig et al. 2017).

Quality control

All filters were handled using standard and replicated filter handling protocols.

Three glass fibre filters were used as blank samples to evaluate contamination during sampling and analysis. These blank samples were placed on active samplers as per other filters but for 10 minutes rather than 24 or 72 hours and without pump operation. Method detection limits were then calculated as follows (Equation 2):

$$MDL = \mu_c + 3s \qquad (2)$$

where μ_c is the mean concentration of analytes in the three blank samples and s is their standard deviation.

RESULTS AND DISCUSSION

The average Dc at Buronga changed from year to year depending on climate and land management practices (Love et al. 2019), with the sample years used in this study shown by the arrows in Figure 2. Each filter selected had a dust event Dc of >200 μg/m³, which is about six times greater than the annual average Dc of 35 μg/m³. The dust samples used were from very large dust events whose Dc magnitude was in the top three per cent of all events measured. The use of large events was driven by the need to have >1 g of sample to analyse. The results of all chemical analyses are presented in Table 1 and Figure 3, with each chemical group analysed discussed in further detail within the following sections.

Results are presented in relation to human health investigation levels (HILs) for soils in residential areas in which vegetables are grown (i.e. HIL-A) presented in Australia's National Environment Protection (Assessment of Site Contamination) Measures 1999 (the ASC NEPM). Such comparison is made, noting that the soil-based HILs are relevant for oral and not respiratory exposure routes (National Environment Protection Council 2013) and that detailed consideration of the human health impacts of chemicals in dust is outside the scope of our research.

Organochlorine pesticides

Organochlorine pesticides (OCPs) are chlorinated carbon chemicals whose use as insecticides spread along with agriculture after World War Two (Rosner and Markowitz 2013). The most widely distributed insecticide in the world, an OCP called DDT, was only banned in Australia in 1987 despite substantial knowledge of its bioaccumulation and toxicity by the 1940s and 1950s (Australian Government Department of Environment and Energy 2017; Rosner and Markowitz 2013). This chemical and its breakdown products, DDE and DDD, were found throughout the dust samples analysed for this study, with the exception of that collected in 1994. While DDT concentrations were higher earlier in the sampling period, reaching 68 ng/g in 1998, DDE was highest in 1998 at 239 ng/g and DDD in 2018 at 22 ng/g. Concentrations of DDT, DDE and DDD were comparatively low in terms of risks to human health, with the sum of DDT, DDE and DDD concentrations being considered acceptable in soils from which vegetables are grown and eaten by humans at concentrations of 240 ng/g or less (National Environment Protection Council 2013).

Both DDT, DDD, as well as DDE are known to undergo LRAT from tropical countries (including Central America) where DDT is still used. All three – DDT, DDD and DDE – have been detected
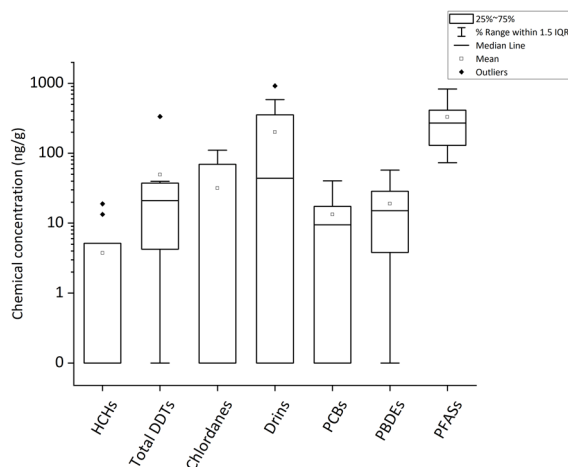

Figure 3. Results for all chemicals analysed from Buronga samples collected between 1990 and 2018. IQR indicates interquartile range.

Table 1. Results for all chemicals analysed from Buronga samples collected between 1990 and 2018. NQ indicates analyte was not quantifiable due to the lack of response for the corresponding internal standard. All concentrations are in ng/g.

Chemical	BU1990	BU1994	BU1998	BU2002	BU2007	BU2009	BU2011	BU2013	BU2014	BU2018
α-HCH	<0.00225	<0.00225	<0.00225	<0.00225	<0.00225	NQ	<0.00225	4.92	<0.00225	NQ
β-HCH	<0.00300	NQ	<0.00300	NQ	NQ	<0.00300	NQ	<0.00300	NQ	<0.00300
γ-HCH	NQ	5.14	NQ	<0.00300	<0.00300	<0.00300	16.6	8.43	NQ	<0.00300
δ-HCH	NQ	<0.00950	NQ	<0.00950	<0.00950	<0.00950	2.32	<0.00950	NQ	<0.00950
Total HCHs	-	**5.14**	-	-	-	-	**18.9**	**13.3**	-	NQ
trans-chlordane	94.9	111	41.6	66.1	<2.19	NQ	<2.19	<2.19	<2.19	<2.19
cis-chlordane	<0.00275	<0.00275	<0.00275	3.49	<0.00275	NQ	<0.00275	<0.00275	<0.00275	<0.00275
Total Chlordanes	**94.9**	**111**	**41.6**	**69.6**	-	-	-	-	-	-
op-DDE	<0.000275	<0.000275	13.5	<0.000275	<0.000275	NQ	<0.000275	<0.000275	0.491	<0.000275
pp-DDE	<0.882	<0.882	239	<0.882	<0.882	NQ	<0.882	13.0	7.08	<0.882
op-DDD	10.4	<0.00205	8.28	4.24	<0.00205	<0.00205	7.07	<0.00205	8.59	21.6
pp-DDD	3.69	<0.00173	<0.00173	<0.00173	9.73	<0.00173	<0.00173	7.35	<0.00173	<0.00173
op-DDT	<0.00275	<0.00275	6.69	<0.00275	5.16	<0.00275	<0.00275	<0.00275	<0.00275	<0.00275
pp-DDT	25.5	<0.00240	67.6	<0.00240	9.73	3.99	<0.00240	<0.00240	21.3	<0.00240
Total DDTs	**39.7**	-	**335**	**4.24**	**24.6**	**3.99**	**7.07**	**20.4**	**37.4**	**21.6**
aldrin	<0.213	<0.213	<0.213	<0.213	<0.213	<0.213	<0.213	<0.213	<0.213	<0.213
dieldrin	<2.11	<2.11	63.4	583	354	919	48.1	<2.11	<2.11	39.5
endrin	0.452	<0.00375	<0.00375	<0.00375	<0.00375	<0.00375	0.262	<0.00375	<0.00375	<0.00375
Total Drins	**0.452**	-	**63.4**	**583**	**354**	**919**	**48.4**	-	-	**39.5**
PCB11	<0.712	32.8	10.0	28.8	8.87	NQ	16.3	15.3	<0.712	<0.712
PCB28	<0.331	4.21	<0.331	10.5	<0.331	NQ	<0.331	<0.331	<0.331	<0.331
PCB52	<0.0644	<0.0644	<0.0644	<0.0644	<0.0644	NQ	1.12	1.00	<0.0644	3.32

Chemical	BU1990	BU1994	BU1998	BU2002	BU2007	BU2009	BU2011	BU2013	BU2014	BU2018
PCB101	<0.401	<0.401	<0.401	<0.401	NQ	<0.401	<0.401	<0.401	<0.401	<0.401
PCB118	<0.000633	<0.000633	<0.000633	<0.000633	<0.000633	NQ	<0.000633	<0.000633	<0.000633	<0.000633
PCB153	<0.00224	<0.00224	<0.00224	<0.00224	NQ	NQ	NQ	<0.00224	<0.00224	<0.00224
PCB138	<0.0616	<0.0616	<0.0616	1.06	<0.0616	<0.0616	<0.0616	<0.0616	<0.0616	<0.0616
PCB180	<0.000750	<0.000750	<0.000750	<0.000750	<0.000750	NQ	<0.000750	<0.000750	<0.000750	<0.000750
Total PCBs	-	**37.0**	**10.0**	**40.4**	**8.87**	-	**17.4**	**16.3**	-	**3.32**
PBDE47	<1.12	<1.12	<1.12	<1.12	<1.12	NQ	<1.12	<1.12	<1.12	<1.12
PBDE100	<0.00250	0.900	<0.00250	3.61	8.22	<0.00250	<0.00250	<0.00250	8.75	8.29
PBDE99	14.9	14.0	40.6	<0.629	25.6	15.3	<0.629	<0.629	18.8	<0.629
PBDE154	<0.00275	<0.00275	<0.00275	0.215	5.18	NQ	<0.00275	<0.00275	1.01	<0.00275
PBDE153	6.54	<0.00575	<0.00575	<0.00575	18.5	NQ	<0.00575	<0.00575	<0.00575	<0.00575
PBDE183	<0.356	<0.356	<0.356	<0.356	NQ	NQ	<0.356	<0.356	<0.356	<0.356
Total PBDEs	**21.5**	**14.9**	**40.6**	**3.82**	**57.5**	**15.3**	-	-	**28.5**	**8.29**
perfluorobutanoic (PFBA)	<0.517	<0.517	<0.517	6.38	235	93.2	366	619	189	12.7
perfluoropentanoic (PFPeA)	<0.197	4.10	<0.197	<0.197	<0.197	<0.197	<0.197	<0.197	<0.197	<0.197
perfluorohexanoic (PFHxA)	<0.966	<0.966	<0.966	<0.966	<0.966	<0.966	<0.966	<0.966	<0.966	17.7
perfluoroheptanoic (PFHpA)	6.04	12.2	4.76	<0.232	5.04	<0.232	<0.232	<0.232	<0.232	20.7
perfluorooctanoic (PFOA)	18.3	69.9	NQ	<0.779	22.2	NQ	NQ	NQ	NQ	691
perfluorononanoic (PFNA)	1.12	<0.0694	<0.0694	0.102	6.29	<0.0694	<0.0694	<0.0694	<0.0694	3.95
perfluorodecanoic (PFDA)	<0.224	<0.224	<0.224	<0.224	13.0	<0.224	<0.224	<0.224	<0.224	7.21
perfluoroundecanoic (PFUnDA)	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400
perfluorododecanoic (PFDoDA)	<0.100	<0.100	<0.100	<0.100	10.5	<0.100	<0.100	2.22	<0.100	1.94
perfluorotridecanoic (PFTrDA)	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400
perfluorotetradecanoic (PFTeDA)	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400

Chemical	BU1990	BU1994	BU1998	BU2002	BU2007	BU2009	BU2011	BU2013	BU2014	BU2018
perfluorohexadecanoic (PFHxDA)	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400
perfluorooctadecanoic (PFODA)	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00	<1.00
perfluorobutanesulphonate (PFBS)	3.20	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	3.75
perfluoropentanesulphonate (PFPeS)	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100
perfluorohexanesulphonate (PFHxS)	4.23	5.42	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	4.63
perfluoroheptanesulphonate (PFHpS)	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400
perfluorooctanesulfonate (PFOS)	33.6	112	19.8	10.8	18.1	1.90	2.64	2.04	<0.100	37.7
perfluordodecanesulphonate (PFDoDS)	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400
8:2 FTS	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100
6:2 FTS	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400
4:2 FTS	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100
PFECHS	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100	<0.100
FOSA	<0.400	<0.400	<0.400	<0.400	11.3	<0.400	<0.400	<0.400	<0.400	<0.400
NEtFOSAA	<1.60	<1.60	<1.60	<1.60	<1.60	<1.60	<1.60	<1.60	<1.60	<1.60
NMeFOSAA	<1.60	<1.60	<1.60	<1.60	<1.60	<1.60	<1.60	<1.60	<1.60	<1.60
NMeFOSA	<40.0	<40.0	<40.0	<40.0	<40.0	<40.0	<40.0	<40.0	<40.0	<40.0
NEtFOSA	<4.00	<4.00	<4.00	<4.00	<4.00	<4.00	<4.00	<4.00	<4.00	<4.00
NMe FOSE	37.0	88.1	64.2	36.6	19.0	7.75	9.68	7.88	9.58	27.2
Net FOSE	97.9	120	41.0	19.3	<0.400	<0.400	<0.400	<0.400	<0.400	<0.400
Total PFASs	**201**	**412**	**130**	**73.2**	**340**	**103**	**378**	**631**	**198**	**828**

in the Antarctic (Kallenborn et al. 2013), northern USA and Canada (Bidleman 1999). The ratio of p,p-DDE to p,p-DDT is an indicator of recently used (≤1) or weathered (>3) DDT (e.g. Aigner et al. 1998; Kallenbourn et al. 2013; Iwata et al. 1993). Low DDE/DDT ratios may suggest active use of the chemical, as still occurs in tropical regions outside of Australia for malaria control (Kallenborn et al. 2013), but perhaps also a greater residency of DDT due to low levels of microbial activity and chemical metabolism in the, leading to limited transformation of DDT to DDE and/or DDD (Bidleman 1999). Stronger affinity of DDT than DDE to soils or dust also causes low DDE/DDT in some instances. Higher DDE/DDT ratios highlight the transformation from DDT and therefore represent weathered DDT from secondary diffusive sources. DDE and DDT only co-occurred in 1998. where the ratio of 4 indicated a secondary source of DDT or metabolic breakdown in Mildura, and in 2014, where the ratio of 0.3 may indicate a primary source and/or slow breakdown of DDT in the environment. Given DDT has been banned in Australia since 1987, a long-range source of DDT from outside Australia may be most likely for the 2014 sample. This is consistent with reporting of <1 DDE/DDT in Antarctica (noting concentrations of DDE were 0.2 to 0.42 pg/m^3) (Kallenborn et al. 2013).

Of the organochlorines, HCHs have one of the highest LRAT capacities and atmospheric concentrations (Beyer et al. 2000; Iwata et al. 1993). They were the most abundant OCP found in an Antarctica study with α-HCH occurring at concentrations of 0.2 to 0.46 pg/m^3. Only γ-HCH was found in three samples collected from Buronga in 1994, 2011 and 2013, with concentrations of 5 to 13 ng/g. No HIL is outlined for HCH in the ASC NEPM. Historically α-HCH predominates in the environment (Walker et al. 1999). The ratio of α/γ HCH is also used to indicate local sources of HCH versus LRAT, with α-HCH occurring in the environment as a result of the photoisomerisation of γ-HCH (Walker et al. 1999). γ-HCH is also the principle ingredient of lindane and α-HCH of technical-HCH, with differing proportions of HCH isomers also indicating use of different pesticides (e.g. Baek et al. 2011; Walker et al. 1999). The results from Buronga are considered to indicate a local source for HCH and / or lindane, with partitioning of α-HCH into the air from dust / soil and away from the site potentially driving the very low α/γ HCH ratio. Microbial degradation of HCH may also be a factor (Bidleman et al. 2015).

Chlordane, banned from use and importation to Australia in 1997, had concentrations of 42 and 111 ng/g between 1990 and 2002 in this study. Such concentrations are comparatively high in relation to the HIL-A outlined in the ASC NEPM, with 50 ng/g of chlordane considered the acceptable limit for soils in which vegetables are grown for human consumption (National Environment Protection Council 2013). All occurrences but two were in the form of *trans*-chlordane. Heptachlor was only detected once at 5 ng/g in 1990 and *cis*-chlordane was 3 ng/g in 2002. Technical chlordane, the name for the insecticide product, contains nearly equal proportions of *trans*- and *cis*-chlordane and occurred in similar concentrations within the atmosphere prior to being banned (Bidleman et al. 2004). The predominance of *trans*-chlordane in this study may be due to the out-gassing of *cis*-chlordane from soils from which the dust was sourced, from the dust itself, or from local sources. In an Arctic study, *trans*-chlordane declined to a greater degree than *cis*-chlordane, particularly in the summer (Bidleman et al. 2002). In studies from non-polar regions, *trans*-chlordane was more predominant (Tombesi et al. 2014; Chakraborty et al. 2010).

Aldrin, endrin and dieldrin, known to be among the most toxic of the organochlorine insecticides since the early 1950s, were banned by the Australian Government in 1992, 1987 and 1988 respectively (Australian Government Department of Environment and Energy 2017). Dieldrin is one of the most persistent of all pesticides and is the principal metabolite of aldrin (Jorgenson 2001). Aldrin was below detection limits in all samples. Endrin was detected in the 1990 dust sample at 0.5 ng/g. This concentration is below the 10 ng/g HIL-A guideline for endrin outlined

in the ASC NEPM. Concentrations of dieldrin were as high as 919 ng/g in 2009, a high dust year, but were below detection limits in 1990, 1994, 2013 and 2014 (Table 1). The HIL-A for the sum of aldrin and dieldrin is 6 ng/g. These results suggest that either aldrin is no longer being applied to soils in the Mildura area, and has been completely metabolised to dieldrin, and/or that dieldrin has potentially been transported to Mildura as a result of LRAT.

Polybrominated biphenyls

Polychlorinated biphenyls (PCBs) are a specific class of 209 organochlorine chemicals used for their dielectric properties, including as coolants and lubricants in transformers and capacitors, until their use was banned in the mid-1970s (Rosner and Markowitz 2013). They are notable for being particularly toxic to humans and other animals (Rosner and Markowitz 2013) and persistent in the environment (Iwata et al. 1993). Many were sold under the trade name Arochlor. Commonly detected gaseous PCBs that have higher vapour pressures such as PCB118, PCB153 and PCB180 were not detected, while those with lower-vapour pressures including PCB11, PCB28, PCB52 and PCB138 were. The lower congener numbered PCB11 and PCB28 were observed in multiple samples and occurred in the highest concentrations, with PCB11 reaching 33 ng/g in 1994 and PCB28 as high as 11 ng/g in 2002. Higher congener numbered PCB52 was observed in three samples and at a concentration of 3.3 ng/g (2018 dust sample). The polychlorinated biphenyl PCB138 was observed in the 2002 dust sample at a concentration of 1.0 ng/g. The concentration of PCBs in seven of the 12 detections of PCBs in samples analysed for this research were higher than typically occurs in soils within rural areas across the globe, where one study found PCB concentrations in rural soils were generally between 0.1 and 7 ng/g (Ruiz-Fernández et al. 2014). They were also above the HIL-A outlined in the ASC NEPM, with a total concentration of all PCBs of 1 ng/g acceptable. One difference between soils and dust is that dust is enriched with soil nutrients and organic matter due to the lighter weight of clay and organic particles (Leys 1999; Leys and McTainsh 1994). Any preferential binding of PCBs to either the clay or organic fraction would therefore most likely result in a higher concentration in dust compared to soils. Outlier PCB concentrations as high as 958 ng/g have been measured in rural soils, with authors citing LRAT as the source of PCBs in this case (Ruiz-Fernández et al. 2014). Higher concentrations of lower congener numbered PCBs are more common in the lower latitudes. The frequent occurrence of low numbered PCBs in this study potentially indicates LRAT (Iwata et al. 1993).

Polybrominated diphenyl ethers

Polybrominated diphenyl ethers (PBDE) are flame retardants added to plasticisers, hydraulic fluids and coatings for electronic devices to inhibit flammability. Australia banned the import and manufacture of both octaBDE (PBDE congener numbers 194-205) and pentaBDE (PBDE congener numbers 82-127) in 2007 (Australian Government 2020). Congener PBDE99 was found in six of the 10 dust samples collected at concentrations from 14.0 ng/g to 40.6 ng/g. Congener PBDE153 had the next highest measured concentration at 18.5 ng/g in 2007 and PBDE100 was measured in four of the 10 samples (max 9 ng/g in 2014). Congeners PBDE47 and PBDE183 were below detection in all samples. The maximum concentration of the sum of PBDEs permissible under the ASC NEPM's HIL-A guideline is 1 ng/g. The concentrations of PBDEs in Buronga dust are similar to levels reported in rural Denmark where concentrations of 0.03 to 40 ng/g were measured in soils (Ruiz-Fernández et al. 2014). They are, however, far lower than concentrations reported in contaminated sites where concentrations can reach more than 10,000 ng/g (Ruiz-Fernández et al. 2014). It is considered likely that PBDEs in Buronga dust are the result of processes such as LRAT or historic contamination in nearby towns such as Mildura.

Per- and poly-fluoroalkyl substances

Chemicals PFOS, PFOA and their precursors are water repellent and heat resistant surfactants listed on the Stockholm Convention and are part of a group of more than 3,000 PFAS chemicals (Swedish Chemicals Agency 2015). They have been used since the 1950s, with PFOS and PFOA degradation end-points for other PFAS as well as being used in their own right (e.g. Wang Z et al. 2017). They typically contaminate indoor dust at concentrations of 1,000s of ng/g and are also known to influence surrounding outdoor environments (Wild et al. 2015). Fire stations, airports and landfills are also key environmental point sources of PFAS (Heads of EPAs Australia and New Zealand 2018).

The PFAS chemicals PFBA, PFOA and PFOS dominated the Buronga dust samples from 2002. Net FOSE (perfluorooctane sulfonamido ethanol) (C8) was found in all samples collected between 1990 and 2002 before falling below detection limits. From 2007, PFBA occurred in every sample and at concentrations of up to 619 ng/g. With the exception of PFOS in 2014, PFOS and NMe FOSE were present in every sample at concentrations of 1.9 to 112 ng/g and 7.7 to 88 ng/g respectively. A greater variety of PFAS were found in 2018 than in any other year, with 11 PFAS chemicals found at concentrations of up to 690.9 ng/g (PFOA).

The PFAS levels in this study were well below current human health screening levels for residences with gardens or accessible soil in Australia, which are 9 µg/g for PFOS and PFHxS and 100 µg/g for PFOA (Heads of EPAs Australia and New Zealand 2018). A municipal sewage treatment plant, landfill and fire-fighting training ground are to the south-west of Buronga and may be the source of PFAS for the dust samples presented herein.

IMPLICATIONS

Although this study represents a small number of analysed samples, results suggest that dust may be an important vector for POPs in Australia. All families of POPs investigated were found in the samples, varying in both concentration and through time. This poses interesting questions surrounding source, persistence and analysis.

A key driver of chemical precipitation and persistence in the environment are cold climatic conditions (Hung et al. 2016; 2010; Kallenborn et al. 2013). While some of the POPs discussed in this research are likely to be products of local chemical use (e.g. HCH), the appearance of others implies re-entrainment from LRAT (e.g. PCB138). The most likely Australian sources of dust passing through Mildura are, however, subject to extreme heat and desiccation. Further research is required to understand whether soil at Mildura is a primary source of the chemicals investigated or whether soil and dust act as secondary sources of chemicals transported to the site from elsewhere via mechanisms such as LRAT or global distillation.

Whilst concentrations of POPs in the gaseous phase are a more significant contributor to global contamination loads than POPs entrained to dust (Iwata et al. 1993), greater understanding of the role that dust may play as a vector is required. That significant volumes of dust are transported around the world as part of the global dust cycle is undisputable, with hemispheric transport pathways documented from multiple continents and moving millions of tonnes globally per annum (Ginoux et al. 2012; Shao et al. 2011). Australian dust is frequently transported from the Mildura area to Australia's east coast (Cohen et al. 2011), New Zealand (Marx et al. 2005) and the Southern Ocean (Gabric et al. 2010). Long-range atmospheric transport from unknown sources delivers POPs to New Zealand (Lavin and Hageman 2013), the Southern Ocean and Tasman Sea (Bhattachan and D'Odorico 2015) and potentially the Mariana and Kermadec trenches (Jamieson et al. 2017) and

Mexico (Ruiz-Fernández et al. 2014). This research highlights the potential importance of Australian dust sources to the transport of POPs globally.

This research indicates that dust may contribute important quantities of POPs to soils in Australia and receptors further abroad. While research suggests such dust can be an important vector of POPs exposure to humans in some instances (e.g. Whitehead et al. 2015), research in this area is limited and outside the scope of this paper. Further investigation regarding the impact of dust containing POPs on receptors down-wind is required and other samples in the DustWatch dataset (about 4300 samples) should also be assessed for their potential to inform long-term trends of these chemicals in the environment and background concentrations at contaminated sites.

ACKNOWLEDGEMENTS

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