Atmos. Chem. Phys., 15, 2313–2326, 2015
www.atmos-chem-phys.net/15/2313/2015/
doi:10.5194/acp-15-2313-2015

Atmospheric Chemistry and Physics
Open Access

Persistent after-effects of heavy rain on concentrations of ice nuclei and rainfall suggest a biological cause

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Received: 18 September 2014 – Published in Atmos. Chem. Phys. Discuss.: 9 October 2014
Revised: 13 January 2015 – Accepted: 3 February 2015 – Published: 3 March 2015

Abstract. Rainfall is one of the most important aspects of climate, but the extent to which atmospheric ice nuclei (IN) influence its formation, quantity, frequency, and location is not clear. Microorganisms and other biological particles are released following rainfall and have been shown to serve as efficient IN, in turn impacting cloud and precipitation formation. Here we investigated potential long-term effects of IN on rainfall frequency and quantity. Differences in IN concentrations and rainfall after and before days of large rainfall accumulation (i.e., key days) were calculated for measurements made over the past century in southeastern and southwestern Australia. Cumulative differences in IN concentrations and daily rainfall quantity and frequency as a function of days from a key day demonstrated statistically significant increasing logarithmic trends ($R^2 > 0.97$). Based on observations that cumulative effects of rainfall persisted for about 20 days, we calculated cumulative differences for the entire sequence of key days at each site to create a historical record of how the differences changed with time. Comparison of pre-1960 and post-1960 sequences most commonly showed smaller rainfall totals in the post-1960 sequences, particularly in regions downwind from coal-fired power stations. This led us to explore the hypothesis that the increased leaf surface populations of IN-active bacteria due to rain led to a sustained but slowly diminishing increase in atmospheric concentrations of IN that could potentially initiate or augment rainfall. This hypothesis is supported by previous research showing that leaf surface populations of the ice-nucleating bacterium *Pseudomonas syringae* increased by orders of magnitude after heavy rain and that microorganisms become airborne during and after rain in a forest ecosystem. At the sites studied in this work, aerosols that could have initiated rain from sources unrelated to previous rainfall events (such as power stations) would automatically have reduced the influences on rainfall of those whose concentrations were related to previous rain, thereby leading to inhibition of feedback. The analytical methods described here provide means to map and delimit regions where rainfall feedback mediated by microorganisms is suspected to occur or has occurred historically, thereby providing rational means to establish experimental set-ups for verification.

1 Introduction

Unraveling the basis of land–atmosphere interactions with feedbacks on rainfall (Pielke et al., 2007; Morris et al., 2014) is increasingly important in the context of climate change. Rain influences a wide range of the earth's surface characteristics including soil moisture, plant proliferation, etc. These characteristics could in turn promote or reduce subsequent rainfall, thereby leading to positive or negative feedbacks, respectively. Positive short-term feedback effects on rainfall have been predicted and attributed to changes in surface albedo and Bowen ratio (the ratio of sensible to latent heat flux) by soil moisture (Eltahir, 1998). However, global scale observational analysis of the coupling between soil moisture and precipitation found no evidence for feedback due to soil moisture (Taylor et al., 2012). Rosenfeld et al. (2001) described a potential feedback effect involving suppression of rainfall by desert dusts. The suppression could lead to drier conditions, favoring further dust storms. Sufficiently intense, prolonged and frequent dust storms required to make such a feedback important do not occur in the regions that will be considered in the present study. If sustained changes in atmospheric concentrations of the particles involved in rain formation follow a fall of rain, then a potential feedback will result. The particles that would be implicated in rainfall feedback are cloud condensation nuclei (CCN), giant cloud condensation nuclei (GCCN, i.e., CCN > 2 µm diameter) and ice nuclei (IN). Möhler et al. (2007) have discussed the ways in which all these particles may affect rainfall.

Measurements of IN concentration in a forest ecosystem showed an increase of these concentrations by an order of magnitude during rain and up to one day thereafter in periods of extended leaf wetness (Huffman et al., 2013; Prenni et al., 2013). Many bacterial and fungal species were involved. The authors speculated that subsequent rainfall could be triggered, leading to the conclusion that airborne microbiological particles, IN and rainfall might be more tightly coupled than previously assumed. Hirano et al. (1996) had earlier noted that populations of the strongly ice nucleation-active bacteria *Pseudomonas syringae* on a bean crop increased by 10-fold to 1000-fold following intense rain. That very large surface increase suggests the possibility of a prolonged increase in airborne IN. Although full details of aerosolization are unknown, *P. syringae* cells have been found to be preferentially lifted into the atmosphere during the warmest part of sunny days when leaves are dry and wind speeds > 1 m s^{-1} (Lindemann et al., 1982; Lindemann and Upper, 1985). Under suitable meteorological conditions, a proportion of the rainfall-enhanced populations would become airborne, leading to an intermittent increase in cloud-active particles relative to those present before the rain. A possible longer-term influence on bacterial populations is growth of vegetation that harbors ice nucleation active microorganisms such as *P. syringae.* This would provide an increased habitat supporting an increased bacterial population and thereby increased numbers of bacterial cells transferred to the atmosphere.

Typical concentrations of CCN active at 0.3 % supersaturation in maritime situations are on the order of 50 cm^{-3} and about 300 cm^{-3} in continental situations (Twomey and Wojciechowski, 1969), whereas IN and GCCN are usually at least four orders of magnitude fewer. If IN and GCCN have a significant influence on rainfall, the order of magnitude changes in their concentration following rain in a forest ecosystem, shown by Huffman et al. (2013), will have a much greater potential influence on subsequent rain than the much smaller proportional changes in CCN concentration.

Increased IN concentrations in clouds colder than 0 °C do not necessarily always lead to increased rainfall. If too many ice crystals form, they may not grow large enough for the resulting raindrops to survive the journey to the ground. Rapid multiplication of ice crystals can occur in clouds that contain droplets of a diameter > 24 µm at temperatures between −3 and −8 °C, (Hallett and Mossop, 1974; Crawford et al., 2012). Enhancement of atmospheric IN in this temperature range could result from emissions of various biological IN such as *P. syringae,* while enhanced GCCN concentrations could provide the relatively large cloud drops needed for multiplication to occur. Increased GCCN concentrations also do not necessarily lead to more rain, even in situations where frequency is increased. In shallow clouds, the formation of drizzle can reduce their water content, potentially decreasing the amount of rain.

For this study, an essential difference between mineral dust aerosols and microbiological aerosols is the relationship between their concentrations and rainfall. Scavenging by raindrops temporarily reduces the concentrations of mineral dust aerosols while rain increases the concentration of microbiological aerosols, and possibly for much longer periods.

The first indication that IN concentrations might have a longer relation to rainfall than a few days was found in 1956 when Bigg (1958) noted that median daily average IN concentrations increased from 0.4 L^{-1} at −20 °C from 11 November to 21 December to 40 L^{-1} on 22 December following a fall of 135 mm of rain in less than 24 h. Median IN concentrations during the following 30 days were 1.75 L^{-1}. Similarly, during a campaign of daily measurements of IN concentrations at 24 sites covering the whole eastern half of Australia, Bigg and Miles (1964) found that mean concentrations of IN on a day with rain increased logarithmically with the mean amount of rain per gauge. It ranged from an overall mean of about 100 IN m^{-3} active at −15 °C for < 1 mm of rain to 1000 IN m^{-3} for 15 mm of rain. Thus, even very light rainfalls led to increased atmospheric IN concentrations on average.

If IN concentrations increase following heavy rain and if IN are important in initiating rainfall, then increased rainfall might follow heavy rain. This speculation led us to study century-long records of daily rainfalls from many sites. Our previous work (Soubeyrand et al., 2014) established that in a band along western and southern coasts of Australia there were statistically significant increases in the amount of rain that occurred in the 20 days following a heavy rainfall relative to the 20 days before it and significant decreases in a limited region in the north of Australia.

The overall aim of the present work is to examine in more detail changes in both IN concentrations and the patterns of rainfall following rainfall. Here we deploy data from past IN measurements and more rainfall sites than in the previous work (Soubeyrand et al., 2014) and we appraise the special conditions that might have influenced rainfall at the various sites. In light of the complex interactions that can occur between aerosols and cloud processes and their consequences on rainfall, we will first characterize the dynamics of IN concentrations over time following heavy rain. We will then show that in large areas of southeastern and southwestern Australia, cumulative differences in the amount of rainfall after vs. before heavy rain usually follow a similar pattern that is well correlated with the dynamics of IN concentrations. Anomalies in the patterns of rainfall feedback will then be examined to determine whether they can be related to land use or other factors.

2 Data sources

2.1 IN data

Data on IN concentrations come from measurements made between 1956 and 1995, the records of which have been held by the first author. These are the only long-term measurements of IN that we are aware of; no other such data are available. Results from four sequences will be presented. The details of their coordinates, start and finish dates, elevations and measurement method used are shown in Table 1.

There is a huge disparity in the information obtained from laboratory instruments and what actually occurs in natural clouds concerning cloud formation processes and lifetimes. As a result, no laboratory instrument can provide an accurate measure of IN concentration in natural clouds. What they can do, however, is to provide consistent relative measurements that allow temporal changes in concentration to be assessed. In this light, absolute concentrations (which cannot in any case be directly related to those in a natural cloud) are irrelevant. In each of the four sequences of data analyzed here, the method of measurement was consistent within the sequence and reflects the temporal changes measured and their relationship to rainfall. The air intakes in each series were approximately 1.5 m a.g.l. It could be argued that the temperatures that were used in the four sequences of measurements were well below the warmest temperature at which mineral dusts can nucleate ice. However, measurements made in and out of dust storms in 1957 with a cloud chamber at 34° S, 146° E (Bigg, unpublished) showed that IN concentrations only began to increase above pre-dust levels at temperatures below −22 °C. There may be other sites where mineral dusts will be important at warmer temperatures, but the November measurements made at the site discussed in the following paragraph showed their concentrations to be very low.

The first data sequence (Bigg, 1958) consisted of an 82 day sequence of measurements at the single site shown in Table 1, located in a coastal forest of melaleuca trees near the north end of Bribie Island (26.6° S, 153.2° E). It was selected because it was free from anthropogenic influences in the upwind direction. The measurements were taken to test a hypothesis by Bowen (1956) that rainfall was influenced by the arrival at the surface of dust from meteor showers on specific dates in November to January. The instrument used was similar to that described by Bigg (1957) except that the cloud volume was 10 L instead of 1 L. Up to 10 individual measurements were made at approximately 2 h intervals between 6 a.m. and 9 p.m. and were combined to form a daily average. During the first month, IN concentrations were so low that it was necessary to operate at a temperature of −20 °C to obtain a significant number of ice crystals.

Bigg and Miles (1964) described a large-scale attempt to detect specific sources of IN at the sites shown in Table 1, Group 1. It was expected that dusts in the semi-arid interior would lead to much higher concentrations there than in high rainfall areas. Instead it was the high rainfall areas that had the highest mean concentrations. Schnell and Vali (1976) were the first to point out that the highest concentrations of IN were in regions where biogenic sources were most active.

"Millipore" cellulose ester filters (HA white, plain, nominal pore size 0.45 µm) sampling 30 000 L each day were exposed in parallel with similar filters continuously sampling air for the daily total of 300 L used for IN measurements. The dust content of the high-volume filters was assessed by microscopy and reflectivity and showed no consistent relationship to IN active at −15 °C. The method for detection of IN concentrations was described by Bigg et al. (1963). Stevenson (1968) developed a method that allowed a good control of humidity and better contact of the filters with a temperature controlled plate. That method was used in all subsequent work although results were not significantly different from the earlier technique. Four filters were first floated on a viscous oil to seal the pores of the filter and to bring any buried particles to the surface. They were then placed on an initially warm thick metal plate in the processing chamber that could have its temperature lowered to −15 °C. At 2 mm above the filters was a second metal plate thinly covered with ice and initially held at −15 °C. The bottom plate was then cooled slowly and once the temperature equilibrium was established, the temperature of the upper plate was raised sufficiently to create a small supersaturation on the filters. The resulting condensation was visible after ice crystals had grown, a circular ring cleared of condensation around each crystal being distinguishable from the condensation-covered area. The measurement was primarily of condensation freezing and probably failed to measure contact nucleation. The presence of hygroscopic particles near a potential IN on a filter reduced the relative humidity below water saturation and could reduce the probability of formation of an ice crystal. The reason for limiting samples to 300 L was to limit losses from this cause.

At the two remaining groups of sites listed in Table 1, continuous sampling was also used to collect 300 L air samples, with duplicates used to detect changes due to storage and blanks to ensure the number of IN on unexposed filters was low enough to be ignored. As before, the filters were sent in sealed containers by post to the processing laboratory and were processed within 6 weeks of collection. No significant changes in IN after storage for up to a year were found.

From 1987 to 1989 measurements of IN concentrations were made at the eight well-spaced sites of Group 2 in Table 1. These were taken in conjunction with a cloud seeding experiment in Victoria aimed at increasing the water for a new reservoir (Long, 1995). The aim of the IN measurements

Table 1. Atmospheric ice nucleus measurement sites.

Single site (Sect. 4.1).[a] From 50–100 10 L samples per day were collected and tested for IN activity in a cloud chamber at −20 °C.									
Latitude ° S	Longitude ° E	Height[b]	Start[c]	End					
26.6° S	153.2° E	5	11/11/1956	30/1/1957					
Group 1: Bigg and Miles, 1964 (Sect. 4.2). Samples of 300 L were tested at −15 °C with the filter method.									
Latitude ° S	Longitude ° E	Elevation	Start	End	Latitude ° S	Longitude ° E	Elevation	Start	End
16.3	133.4	212	5/9/1962	17/5/1964	30.3	153.1	6	2/9/1962	17/5/1964
16.9	145.8	4	7/9/1962	17/5/1964	31.1	150.8	410	3/9/1962	31/12/1965
19.3	146.8	7	3/9/1962	17/5/1964	31.5	159.1	6	8/9/1962	17/5/1964
20.7	140.5	190	11/91962	17/5/1964	33.7	150.1	1150	14/9/1962	31/12/1965
21.2	149.2	7	5/9/1962	17/5/1964	33.8	150.0	120	1/5/1962	31/12/1965
23.8	133.9	546	6/9/1962	17/5/1964	34.2	142.1	52	7/9/1962	17/5/1964
23.9	151.2	19	18/9/1962	17/5/1964	34.8	149.6	750	11/9/1962	31/12/1965
26.4	146.3	307	4/9/1962	17/5/1964	35.2	147.5	223	11/9/1962	17/5/1964
27.4	153.1	5	5/9/1962	17/5/1964	37.7	140.8	66	8/9/1962	17/5/1964
27.6	135.4	119	13/9/1962	17/5/1964	38.1	147.1	8	12/9/1962	17/5/1964
29.7	151.7	1060	13/9/1962	31/12/1965	41.0	145.7	20	11/9/1962	17/5/1964
Group 2: Melbourne Water (Sect. 4.3). Samples of 300 L were tested at −15 °C with the filter method.									
Latitude ° S	Longitude ° E	Elevation	Start	End	Latitude ° S	Longitude ° E	Elevation	Start	End
37.3	146.1	340	12/03/1987	30/04/1989	37.6	146.7	440	12/03/1987	30/04/1989
37.6	145.5	174	12/03/1987	30/04/1989	37.8	145.6	230	12/03/1987	30/04/1989
37.6	146.3	1207	12/03/1987	30/04/1989	38.0	145.4	500	12/03/1987	30/04/1989
37.6	146.1	1158	12/03/1987	30/04/1989	38.0	146.4	470	12/03/1987	30/04/1989
Group 3: Tasmania (Sect. 4.3)[d]. Samples of 300 L were tested at −15 °C with the filter method.									
Latitude ° S	Longitude ° E	Elevation	Start	End	Latitude ° S	Longitude ° E	Elevation	Start	End
42.28	146.28	667	20/06/1992	23/12/1994	41.43	146.11	590	1/06/1992	31/12/1994
42.13	146.75	576	1/06/1992	25/12/1994	41.63	145.6	380	1/06/1992	31/12/1994
41.98	146.72	1052	1/06/1992	31/12/1994					

[a] Rain fell on 10 of the 42 days between 11 November and 21 December, totalling 60 mm, and on 11 of the 42 days from 23 December to 30 January, totalling 203 mm.
[b] Above-ground sampling heights and elevations are indicated in meters.
[c] Starting and end dates of sampling campaigns are indicated as day/month/year.
[d] The coordinates and elevations here are for the nearest rainfall measuring sites. The IN sites differ only slightly from these.

was to test the hypothesis that there were persistent effects on IN of seeding with silver iodide. Four of the sites were in forested areas (mainly eucalyptus) and the others were in open woodland or pasture. The seeding agents used were silver iodide or dry ice (frozen carbon dioxide) dispensed from aircraft in clouds. Using days with seeding as key days and a similar method to those of Sect. 3, the longer (five years) and more numerous and closely spaced series of rainfall measurements showed that there were persistent after-effects on IN of seeding (Bigg, 1995). The cause has not yet been found. Area rainfall exceeded the 10 mm threshold on 107 of the 750 days covered by the IN measurements and only 6 of those days corresponded to seeding events. None of those days had unusually high IN concentrations so any influence of AgI on the analysis would have been slight.

Measurements at Group 3 sites in Table 1 were made in conjunction with a cloud seeding experiment in Tasmania using dry ice as the seeding agent to determine whether the naturally occurring IN concentrations affected apparent seeding success.

2.2 Rainfall sites

The sites and periods of records for rainfall data are given in Table 2. The rainfall records used were daily totals (mm) listed by the Australian Bureau of Meteorology and are readily available online from http://www.bom.gov.au/climate. These sites represent a much denser network of rainfall sites than those used in a previous study in which we elaborated the method of calculation to reveal rainfall feedback patterns (Soubeyrand et al., 2014). In that study we showed that in the southeastern and southwestern corners of Australia at most sites there was significantly more rain (or rain occurrences) in the 20 days following a day with rain above a certain threshold than in the preceding 20 days, suggesting a feedback effect. We also showed that this effect was usually greater before 1960 than after it.

The technique to elucidate rainfall feedback depends on the identification of the days in a series of data with a threshold amount of rainfall. Selection of threshold rainfalls depends on a balance between obtaining enough cases in the total series to reduce random variations sufficiently and not so many that more than one key day often occurs in a 41 day

Table 2. Southeast group of rainfall sites.

Site	Latitude ° S	Longitude ° E	Start	End	Elevation (m)	Rain threshold (mm)	R^2 for logarithmic curve[a]	% CD^b_{FH}	Site	Latitude ° S	Longitude ° E	Start	End	Elevation (m)	Rain threshold (mm)	R^2 for logarithmic curve[a]	% CD^b_{FH}
25 518	36.18	140.57	1910	2010	64	20.0	0.74	7.2	84 016	37.57	149.92	1864	2010	15	40.0	0.84	4.6
26 005	38.06	140.69	1868	2006	40	25.0	**0.94**	10.3	84 025	37.85	147.77	1905	2010	11	30.5	0.46	2.9
26 007	36.71	140.96	1889	2010	103	21.6	0.00	5.6	84 030	37.69	148.46	1883	2010	41	40.0	0.76	11.1
26 014	37.61	140.58	1892	2009	105	25.7	**0.97**	13.6	85 020	38.04	147.18	1884	2009	3	30.0	0.76	5.0
69 018	35.91	150.15	1895	2010	17	62.2	0.00	2.2	85 023	38.14	145.86	1899	2010	135	33.8	0.70	10.8
69 029	37.36	149.70	1909	2010	35	40.0	**0.91**	7.2	85 033	38.42	147.09	1906	2010	15	25.4	**0.97**	13.8
69 107	36.74	149.71	1901	2010	125	41.9	0.63	1.0	85 040	38.69	146.15	1906	2010	145	31.0	**0.94**	10.5
70 009	36.80	149.20	1879	2010	785	30.0	0.84	13.7	85 042	38.02	145.89	1899	2010	250	35.4	**0.92**	12.6
70 027	37.05	148.99	1889	2010	750	35.0	0.42	3.9	85 045	38.44	145.82	1900	2010	205	36.6	0.78	9.2
70 057	35.82	149.63	1898	2009	730	44.0	**0.90**	12.7	85 049	38.48	145.93	1891	2010	52	30.5	0.80	9.7
70 067	36.51	149.28	1894	2010	1075	36.0	0.88	8.4	85 050	37.80	147.46	1901	2010	35	30.0	0.78	5.3
71 000	36.00	148.77	1887	2010	1015	30.0	0.54	4.8	85 059	38.20	146.26	1897	2010	150	33.0	0.10	6.
74 034	35.99	146.36	1890	2010	143	27.4	0.01	4.1	85 063	38.61	146.32	1903	2010	235	35.0	**0.97**	15.4
74 236	36.02	146.76	1893	2010	158	25.0	**0.94**	12.7	85 073	38.32	147.18	1898	2010	28	26.7	0.43	7.4
77 008	35.92	142.85	1890	2010	100	20.1	**0.97**	19.6	85 084	38.66	146.33	1905	2010	20	33.3	0.27	7.4
77 036	35.83	141.94	1901	2009	79	20.0	0.00	5.1	85 085	38.22	146.17	1902	2010	43	30.0	0.77	5.7
78 038	36.27	142.22	1900	2010	115	20.3	**0.97**	16.4	85 093	38.17	145.95	1888	2010	143	35.1	0.48	6.7
78 041	36.25	143.18	1887	2010	134	25.0	**0.96**	26.3	85 096	39.13	146.42	1872	2010	95	30.0	0.27	3.2
78 078	36.37	141.24	1883	2010	142	21.6	0.66	10.5	86 066	37.75	145.34	1885	2010	101	31.8	0.33	5.8
79 008	36.94	141.37	1903	2010	155	21.3	**0.97**	18.1	86 070	37.64	145.55	1892	2010	174	38.4	0.86	8.7
79 011	37.00	141.30	1890	2009	162	22.8	0.64	11.1	86 071	37.81	144.97	1855	2010	31	30.0	0.36	10.7
79 035	36.62	142.47	1883	2010	145	22.0	0.87	16.0	86 073	37.55	144.88	1902	2009	270	27.7	0.72	7.5
79 039	37.09	143.34	1897	2010	260	27.2	0.79	14.6	86 085	37.99	145.34	1866	2009	120	33.5	0.82	11.6
79 040	36.62	143.26	1880	2010	240	27.2	0.8	12.7	86 096	37.72	145.01	1910	2010	94	25.7	**0.99**	11.3
79 046	37.09	142.43	1890	2010	440	31.0	**0.94**	15.7	86 121	37.75	145.68	1878	2007	170	43.4	0.35	5.1
80 015	36.16	144.76	1881	2010	96	24.1	0.82	9.8	87 000	37.88	144.26	1892	2010	247	25.7	**0.94**	12.3
80 039	36.19	144.06	1886	2010	100	23.0	0.87	13.5	87 006	37.60	144.23	1875	2010	509	30.5	0.70	8.7
81 000	37.09	143.47	1884	2010	242	28.7	**0.90**	14.4	87 009	38.05	144.15	1898	2010	106	25.0	**0.94**	9.5
81 007	36.46	145.66	1903	2010	130	26.0	0.89	12.4	87 023	38.16	144.40	1897	2009	4	25.0	**0.98**	15.5
81 008	36.54	144.78	1899	2010	144	26.4	0.87	9.0	87 054	38.27	144.66	1896	2010	15	25.0	0.70	12.6
81 047	36.77	143.83	1888	2008	210	26.2	0.01	3.2	88 001	37.19	145.71	1877	2010	221	30.0	0.21	6.2
81 051	36.16	145.88	1889	2010	122	26.7	0.70	4.7	88 002	36.89	145.23	1900	2010	152	27.9	0.89	16.5
82 002	36.55	145.97	1882	2006	170	30.8	0.32	4.0	88 007	37.03	145.85	1902	2010	297	30.0	0.79	13.3
82 006	36.22	146.28	1908	2009	142	30.0	0.8	5.8	88 020	37.34	144.16	1869	2010	612	36.8	0.76	12.4
82 009	36.61	146.54	1910	2010	238	34.6	**0.91**	9.0	88 023	37.23	145.91	1887	2010	230	30.0	0.29	9.2
82 011	36.20	147.9	1891	2006	314	31.2	0.75	8.7	88 029	36.96	144.69	1858	2010	294	30.6	0.89	13.9
82 016	36.75	145.57	1883	2010	181	31.0	0.76	6.0	88 031	36.75	146.42	1903	2010	230	44.5	0.5	5.0
82 029	36.46	146.43	1902	2010	158	30.0	0.75	5.8	88 044	37.51	145.75	1904	2009	420	39.6	0.72	9.3
82 047	36.19	147.36	1887	2009	220	36.1	0.84	10.3	88 048	37.11	144.06	1897	2010	217	25.0	0.52	10.5
82 057	36.30	146.73	1899	2010	392	30.0	0.86	9.6	88 060	37.45	145.21	1884	2010	488	40.1	0.83	7.5
83 019	37.05	146.09	1901	2010	316	30.0	0.16	2.0	88 067	37.22	145.42	1885	2010	193	30.0	0.60	8.8
83 025	37.10	147.60	1879	2009	685	30.0	0.44	3.7	89 002	37.51	143.79	1908	2010	435	24.1	**0.95**	18.4
83 031	36.75	146.42	1903	2010	240	44.5	0.50	5.0	89 003	37.25	141.84	1884	2010	180	23.6	0.84	10.0
83 073	36.72	146.82	1910	2010	1350	70.0	**0.99**	11.2	89 025	37.69	143.37	1897	2010	295	24.8	0.71	11.8
84 003	37.71	147.83	1882	2010	22	40.6	0.65	5.8	89 034	37.55	142.74	1902	2010	250	22.6	**0.92**	20.1
84 005	37.50	148.17	1883	2010	140	38.1	0.82	5.8	90 005	38.20	143.64	1903	2010	120	25.0	0.35	8.8

[a] Correlation of CD_F with a logarithmic curve. Those > 0.9 are in bold type.
[b] Percent difference between CD_{FH} for the whole length of each record and the mean rainfall (for the whole series of each site) in the 20 days before key days.

period. (The latter reduces the sensitivity of the test for differences between the 20 days after a key day and the 20 days preceding it). Therefore, we targeted obtaining 300 cases per series, with a minimum threshold set to 20 mm. At a few low rainfall sites the number of cases fell below 300. Soubeyrand et al. (2014) made calculations of the confidence levels of the after-effects of heavy rain in the two areas chosen. Because the present study involves more than 160 sites, individual confidence levels have not been calculated. Accumulated totals of differences in 20-day rainfalls following and preceding days with rain equal to or greater than the threshold are shown in Tables 2 and 3 as a percentage of mean 20-day rainfalls preceding such days. High confidence levels can be expected to accompany high totals.

Selection of sites within the two large areas chosen was based on the length of record (a minimum of 97 years), the amount of missing data (maximum 5 %) and separation of sites: > 20 km except for a few close pairs used to test consistency of records. Tables 2 and 3 give site positions, start and finish dates, site elevation and some details of apparent feedback that will be described in Sect. 5.

3 Basic method for calculating after-effects of heavy rain

Rainfall occurs at widely varying intervals and with a large variation in amount, dictated mainly by meteorology. IN concentrations are also highly variable, although always > 0 at the usual temperatures of measurement. Meteorological factors such as wind strength and depth of atmospheric mixing as well as specific sources of IN are some of the causes of changing IN concentrations.

Soubeyrand et al. (2014) developed a mathematical formulation of a superposition technique that is an effective way to detect changes with time resulting from a particular event occurring at irregular intervals. To aid understanding of this technique, a simple example of the basic procedure is presented below. The procedure for detecting any after-effects of heavy rain on the quantity of rain, the frequency of occurrence of rainfalls (i.e., any rainfall > 0 mm), or on IN concentrations is exactly the same.

Here we consider occasions where > 25 mm of rain fell in a 24 hour period, called a “key day”. We want to compare the quantity of rain following and preceding all such events as a function of time from the event. To search for after-effects in the 20 days following a key day, the following steps can be used:

1. The sequence of 41 days, extending from 20 days before the first key day (day −20) to 20 days after it (day 20), is placed in a column. For the next such event, the sequence of 41 days (extending, as before, from 20 days before the second key day to 20 days after it) is placed in a column so that its key day is aligned with that in the first column. This is repeated for all key days.

2. The mean of each day, starting from 20 days previous to and 20 days after the key day, is then calculated for the entire key day series.

3. The mean for day −1 is subtracted from that of day 1. This is repeated for each pair of days. This will create a sequence of differences D_1, D_2,… D_{20} after–before a key day.

4. To reveal a trend, cumulative differences (CD) of the difference sequence are calculated as $\mathrm{CD}_1 = D_1$, $\mathrm{CD}_2 = \mathrm{CD}_1 + D_2$, $\mathrm{CD}_3 = \mathrm{CD}_1 + \mathrm{CD}_2 + D_3$… and so on.

Suppose instead that rainfall quantity in the whole 20 days preceding a key day is summed and subtracted from that in the 20 days following that key day. Forming a cumulative sum of these differences over the whole series of key days provides a historical record of changes in CD with time.

If a day with rain above the key day threshold occurs in the 20 day sequences before or after a key day, it is included. Excluding such cases reduces the ability to detect a consistent response to key day effects, at the expense of smoothing that response.

The steps 1–4 listed above can be expressed as:

$$\mathrm{CD}_j = \sum_{j'=1}^{j} \left\{ \left(\frac{1}{n} \sum_{k \in \text{key days}} d_{k+j'} \right) - \left(\frac{1}{n} \sum_{k \in \text{key days}} d_{k-j'} \right) \right\}, \qquad (1)$$

where d_k is rainfall quantity or occasion at key day k, $d_{k+j'}$ is rainfall quantity or occasion j' days after key day k, $d_{k-j'}$ is rainfall quantity or occasion j' days before key day k, and n is number of key days in the rainfall series under consideration.

To calculate cumulative differences as a function of time from a key day, CD_Q will be used where quantity of rain is the variable and CD_F where frequency of rain (number of occasions with rain > 0 mm) is the variable. For calculating historical cumulative differences, the corresponding symbols CD_QH and CD_FH will be used. The dimensions of CD_Q

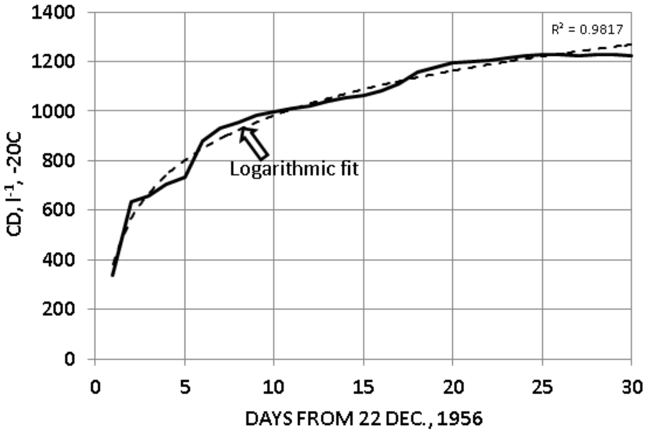

Figure 1. CD in IN concentrations at 26.6° S, 153.2° E following a day with 135 mm of rain.

and CD_QH are mm, while CD_F and CD_FH are dimensionless. Soubeyrand et al. (2014) have provided a package for making the calculations automatically, including estimates of statistical significance (http://cran.r-project.org/web/packages/FeedbackTS/index.html).

There is a potential difficulty with the calculations of CD_Q and CD_F. If key days are not distributed evenly about the annual maximum frequency of occurrence or maximum quantity of rain, a bias in CD of purely meteorological origin results. In the relatively short rainy season north of about 28° S in Western Australia this produced significant errors, as large as 30 % in the tropics. To counter this effect, the mean rainfall for each day of the year is calculated for the whole length of records. Day of the year for each key day is listed and calculation of $\mathrm{CD'}_\mathrm{Q}$ and $\mathrm{CD'}_\mathrm{F}$ identical to those above is carried out using 41 day sequences from the mean daily rainfall, centered on the day-of-year number of each key day. The corrections $\mathrm{CD}_\mathrm{Q} - \mathrm{CD'}_\mathrm{Q}$ and $\mathrm{CD}_\mathrm{F} - \mathrm{CD'}_\mathrm{F}$ should remove the artifact.

4 Persistent effects of rainfall on atmospheric IN concentrations

4.1 The single site measurements described in Sect. 2.1 and Table 1

Median concentrations from 11 November to 20 December were 0.4 L^{-1} and from 22 December to January were 1.75 L^{-1}. On the night of 21 December 1956, 135 mm of rain fell and IN concentrations averaged 40 L^{-1} on the following day. Rain fell on 10 of the 42 days before 21 December, totalling only 60 mm, and on 11 of the 42 days after it, totalling 203 mm. From these, the day with by far the greatest rainfall was chosen as the sole key day.

Using the method described in Sect. 3, the cumulative differences (CD) in IN concentration are shown in Fig. 1.

Table 3. Southwest group of rainfall sites.

Site	Latitude ° S	Longitude ° E	Start Start	End	Elevation (m)	Rain threshold (mm)	R^2 for logarithmic curve[a]	% CD^{b}_{FH}	Site	Latitude ° S	Longitude ° E	Start	End	Elevation (m)	Rain threshold (mm)	R^2 for logarithmic curve[a]	% CD^{b}_{FH}
8061	30.28	116.63	1911	2010	340	20.0	**0.93**	**19.8**	10 052	31.45	117.22	1910	2010	300	20.0	0.87	19.8
8066	30.70	117.06	1907	2010	310	20.0	0.78	20.7	10 092	31.48	118.28	1904	2010	315	20.0	0.86	19.9
8084	30.26	116.21	1908	2008	320	20.0	**0.94**	**20.5**	10 097	30.66	117.26	1910	2010	375	20.0	0.87	16.5
8086	29.15	115.24	1900	2010	120	23.9	0.03	8.3	10 121	31.64	117.49	1911	2010	250	20.0	0.88	22.4
8096	28.15	114.57	1903	2010	200	23.0	**0.91**	**26.0**	10 123	31.99	118.18	1904	2010	280	20.1	0.86	28.5
8104	28.15	114.67	1910	2010	280	21.8	0.85	20.8	10 124	31.51	118.18	1904	2010	280	20.1	0.68	12.6
8137	30.89	116.72	1907	2005	283	20.6	0.77	21.0	10 126	31.12	117.79	1910	2010	290	20.0	0.82	19.9
8238	30.10	116.23	1908	2010	350	20.0	0.71	20.6	10 133	30.83	117.32	1909	2010	320	20.0	0.88	24.1
9001	32.13	116.00	1903	2010	55	36.8	0.66	13.4	10 149	31.34	117.82	1902	2010	320	20.0	0.85	18.0
9513	33.21	115.85	1909	2010	30	37.6	0.73	14.3	10 502	34.26	118.22	1907	2010	210	20.1	0.17	13.2
9518	34.37	115.14	1903	2010	13	31.2	0.89	10.8	10 504	34.08	117.04	1911	2008	300	20.1	**0.96**	**15.4**
9519	33.54	115.02	1907	2010	109	29.7	**0.96**	**12.6**	10 510	33.16	116.80	1911	2010	280	22.6	**0.93**	**16.5**
9530	34.38	116.41	1903	2010	140	26.2	0.75	13.0	10 524	32.37	117.01	1907	2010	250	22.6	0.85	17.1
9531	34.96	117.36	1911	2010	20	30.7	0.54	9.4	10 528	33.25	117.66	1903	2009	310	20.0	0.83	19.9
9534	33.57	115.82	1907	2010	63	36.3	0.88	11.5	10 536	32.33	117.87	1910	2010	295	20.3	**0.96**	**17.8**
9551	35.02	117.76	1907	2010	10	30.0	**0.96**	**9.5**	10 541	33.86	118.82	1907	2010	280	20.0	**0.97**	**16.2**
9557	33.95	120.13	1907	2010	15	23.1	0.71	10.5	10 546	33.31	117.74	1910	2010	270	20.1	0.84	15.3
9559	34.91	117.99	1911	2009	50	25.7	**0.90**	**12.1**	10 547	33.52	116.80	1910	2010	230	22.4	**0.94**	**18.0**
9561	34.49	117.63	1907	2010	262	21.8	0.88	10.5	10 561	33.03	117.39	1911	2010	315	20.3	0.86	12.1
9564	34.94	117.92	1907	2010	12	27.0	0.59	6.3	10 586	32.01	118.23	1910	2010	270	20.1	**0.92**	**20.9**
9581	34.62	117.64	1907	2010	300	24.1	0.62	8.5	10 628	32.01	117.40	1911	2010	250	20.0	0.82	19.7
9591	34.64	117.38	1907	2009	230	24.1	0.84	11.9	10 633	33.58	120.05	1907	2010	232	20.1	0.73	15.0
9594	34.43	119.36	1907	2009	60	27.2	0.88	11.5	10 634	32.31	116.77	1911	2010	260	23.1	0.83	14.9
9616	34.09	116.66	1903	2010	280	25.4	0.83	11.6	10 635	33.79	116.91	1909	2010	260	21.6	0.86	17.6
9619	34.15	116.20	1903	2010	240	31.0	**0.94**	**9.2**	10 636	33.16	117.67	1907	2008	340	20.1	0.87	15.1
10 002	31.37	117.90	1911	2010	270	20.0	**0.96**	**20.3**	10 654	32.78	117.50	1911	2010	350	20.3	**0.91**	**20.5**
10 032	31.00	117.40	1907	2010	300	20.0	0.87	24.7	10 658	32.83	116.99	1905	2009	300	23.8	0.45	13.1
10 039	31.01	117.20	1907	2010	295	20.0	0.57	14.8	10 668	32.47	118.15	1910	2010	280	20.0	**0.94**	**21.6**
10 040	31.61	117.88	1910	2010	251	20.0	0.88	26.5	11 009	32.67	123.58	1907	2007	160	20.0	0.77	25.4
10 042	31.19	117.03	1904	2010	273	20.1	**0.92**	**15.4**	12 011	30.99	119.11	1911	2010	362	20.0	0.85	13.5
10 045	31.01	117.13	1907	2010	290	20.0	0.58	13.0	12 033	33.23	121.74	1906	2009	210	20.0	0.77	14.1

[a] Correlation of CD_F with a logarithmic curve. Those > 0.9 are in bold type.
[b] Percent difference between CD_{FH} for the whole length of each record and the mean rainfall (for the whole series of each site) in the 20 days before key days.

The remarkable feature of this curve is its close approximation to a logarithmic form. Note that the log curve has flattened out soon after 20 days (between 20 and 25 days). This appeared to be the case in all subsequent measurements, therefore 20 days from a key day was subsequently taken as the standard length of all CD series for both IN and rain.

4.2 Group 1 measurements of IN, Sect. 2.1, Table 1 and Fig. 2a

Figure 2a shows the location of the sites at which daily mean IN were measured for periods of 18 months to 3 years. Sites marked with a circle were Meteorological Offices located at aerodromes. Ground cover in the immediate vicinity was mostly mown grass, ranging from sparse at inland sites to thick at coastal sites. The remaining sites (triangles) were rural homesteads surrounded by pastures or open woodlands. With this lengthy series of measurements including many sites where large daily rainfalls were relatively frequent, threshold rainfalls for key days were set at 25 mm.

Figure 2b shows the relationship between CD and the number of days after a key day. A logarithmic curve again fit the data well ($R^2 = 0.89$) except for days 4 to 6.

4.3 Groups 2 and 3 IN measurements, Sect. 2.1, Table 1

Key days in the IN analysis in the series for both groups 2 and 3 had to be set at rainfalls ≥ 10 mm as there was an insufficient number of key days if heavier rainfalls were used. The cumulative after–before differences in the case of Group 2 revealed recurrent peaks on a logarithmic trend (Fig. 3, left). The Group 3 measurements showed a smaller effect but a better fit to a logarithmic curve.

We conclude from the four cases listed here that the cumulative response of IN concentrations to a heavy fall of rain is approximately logarithmic, implying an impulsive increase on day 1, decreasing exponentially with time to about day 20.

5 Persistent increases in rainfall following days with rainfall above a threshold

As ice crystals can initiate precipitation from clouds, it is relevant to consider whether the increased concentrations of atmospheric IN for up to 20 days following heavy rain as described above led to corresponding increases in the quantity of rain or the number of days with rain > 0 mm. If this is the case, it would be consistent with a feedback process between rainfall (that generates IN) and IN (that generate rainfall). Here we present CD in rainfall for a range of sites. We focus on CD_F, which is a more regular measure of after-effects than CD_Q and has a generally greater percentage increase following rain than CD_Q.

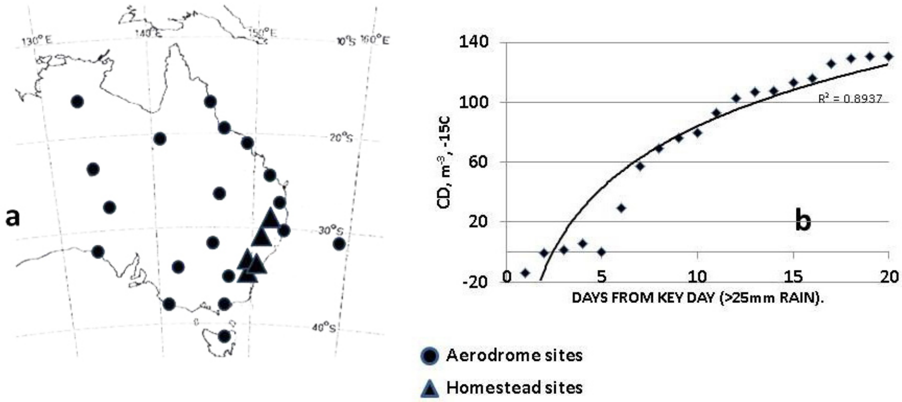

Figure 2. **(a)** IN measurement sites, **(b)** CD in IN as a function of days after key days with ≥ 25 mm of rain.

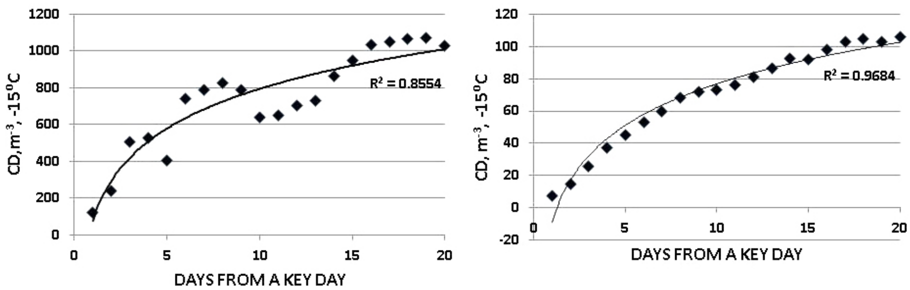

Figure 3. CD in IN as a function of days after a key day for **(left)** Group 2, Table 1 sites, **(right)** Group 3, Table 1 sites.

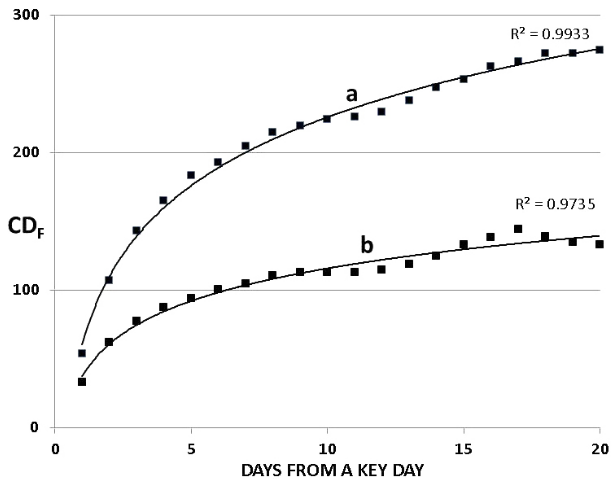

Figure 4. Mean CD in rain frequency (CD_F) (number of occasions of rain > 0 mm) **(a)** for 22 % of southeastern sites, **(b)** for 60 % of southwestern sites.

5.1 CD_F for sites listed in Tables 2 and 3

We have seen that there was an approximately logarithmic increase in cumulative after–before key day differences (CD) in atmospheric IN concentrations up to at least 20 days after rain. Using the same type of analysis for CD_F, approximately logarithmic increases were again almost universal. For 34 % of the sites in the southeastern group and 69 % of the sites in the southwestern group the correlation R^2 between CD_F and a logarithmic curve was ≥ 0.85 within 20 days from a key day. Figure 4a shows the very close fit ($R^2 = 0.99$) to a logarithmic curve of the mean CD_F curve for those 34 % of southeastern sites. Figure 4b shows the mean CD_F curve for those 69 % of southwestern sites. The correlation with a logarithmic curve was $R^2 = 0.97$. The improved correlations obtained by averaging many cases implies that most curves contained small random deviations from a logarithmic form. The comparable logarithmic increases in CD_F following heavy rain events to those in IN suggests a common cause.

At some sites CD_F had oscillations superimposed on a logarithmic curve. At 10 % of the southeastern sites and 18 % of the southwestern sites the oscillations had consistent periods of 5 to 7 days, suggesting that they were not random variations. Average CD_F curves for the sites are shown in Fig. 5a and c (Fig. 5c is Fig. 5b corrected for the artifact described in Sect. 3). At a larger number of sites, the oscillations had smaller or less regular periods, making it less certain that they were not simply random fluctuations. It should be noted that CD of IN in Fig. 3 (left) also had three oscillations superimposed on a logarithmic curve, though not as regular or large as those of Fig. 5.

At most of the remaining sites (mainly in low rainfall areas), CD_F was closely logarithmic for at least 10 days from a key day but at some point between 10 and 20 days, an accelerating downward trend began.

5.2 CD_{FH} for the southeastern group of sites listed in Table 2

Soubeyrand et al. (2014) found marked shifts in the trend of CD_{QH} and CD_{FH} at numerous locations at ca.1960. This date corresponds to the beginning of a period of industrialization and changes in demography in Australia leading to, among other things, important changes in land use. The rainfall records for each site in the groups to be used here were therefore divided to have equal numbers of key days in two groups, the division typically being about 1960. The positions of all sites are shown by open circles on the map in Fig. 6. In order to look at the possibility that industrial or population centers influence persistent after-effects of rain, the locations of large power stations are marked. The first of a complex of power stations labeled H (Hazelwood) was built between 1964 and 1971 and had a capacity of 1600 MW. It was fueled by brown coal and has been a notorious polluter. According to the National Pollution Inventory (http://www.npi.gov.au) of 2005–2006 this power station emitted 2.9×10^6 kg of PM_10 particulates (particles > 10 µm) in that year. It also emitted 1.2×10^5 kg of SO_2 and many other chemical compounds. Other large electricity generating plants have subsequently been built in the same area and, while their individual particulate emissions have been less, their combined SO_2 output has been considerably greater. A 150 MW power station at Anglesea, marked A in Fig. 6, opened in 1969 and also produced large amounts of both SO_2 and particulates. The growth of a city certainly changes the nature of the land on which rain falls and may possibly also alter the effects of the rain. A large area surrounding M on Fig. 6 is occupied by the city of Melbourne. Its population grew from about 900 000 in 1900 to 1.5 million in 1956 and to 4 million in 2010.

CD_{FH} for the entire period of records has been listed for each site in Table 2. Pre- and post-ca.1960 CD_{FH} was then calculated separately for each site. R is defined as CD_{FH} (pre-1960)/CD_{FH} (post-1960). Contours of R were then prepared, using interpolation between nearby sites to locate specific contours. In some areas there is a large spacing between sites due to the absence of long or complete rainfall records. Errors in drawing contours may therefore occur. To test this, after preparing the contours of Fig. 6, in regions where sites were closely spaced some were omitted and contours were redrawn. Some shifting of the contours resulted but did not substantially alter the overall pattern. The most conspicuous feature of Fig. 6 is the area with high R to the northeast of the power station complex. It implies a reduction in CD_{FH} after about 1960. Less prominent increases in R occurred west of Melbourne and in a band near its northeastern corner extending to the northeast. In contrast, a substantial area surrounding Melbourne, the adjacent southeast coastline and a narrow extension northeast from Melbourne, R decreased in the later period, implying an increase in CD_{FH}. This was also the case for an area along the northern edge of Fig. 6 between longitudes 141 and 146° E.

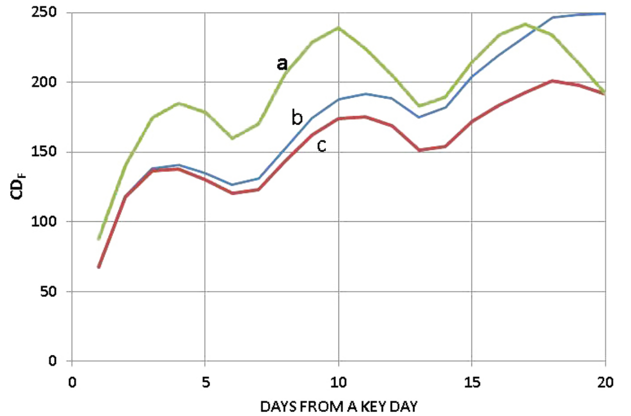

Figure 5. Mean CD_F for **(a)** 10 % of southeastern sites and **(c)** 18 % of southwestern sites showing regular oscillations with time after a key day. **(c)** is **(b)** corrected for the artifact described in Sect. 3.

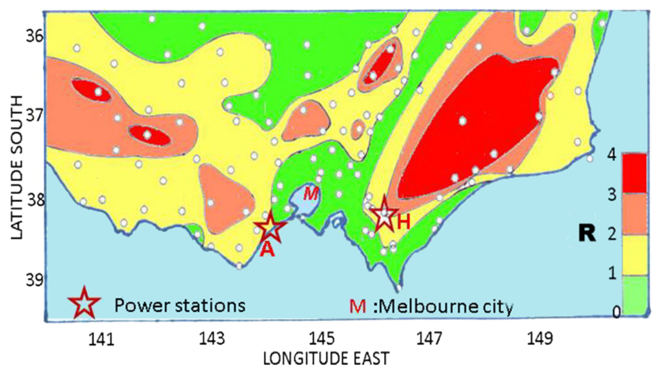

Figure 6. Contours of R (ratio of pre-1960 to post-1960 CD_{FH}) for the sites in Table 2. Hazelwood (H) and Anglesea (A) power stations are marked with a star, the city of Melbourne with M.

Possible reasons for the patterns of Fig. 6 will be considered in the Discussion, Sect. 6. First we will consider whether similar patterns emerge round a power station and population center for the sites given in Table 3.

5.3 CD_{FH} for the southwestern group of sites listed in Table 3

Open circles in Fig. 7 show the locations of all sites used. A red star shows the position of a 970 MW power station (Muja) commissioned in 1966. It has been a major emitter of particulate matter. P shows the location of Perth, the only major city in the area. Its population was 100 000 in 1911, 400 000 in 1961 and 1.5 million in 2010.

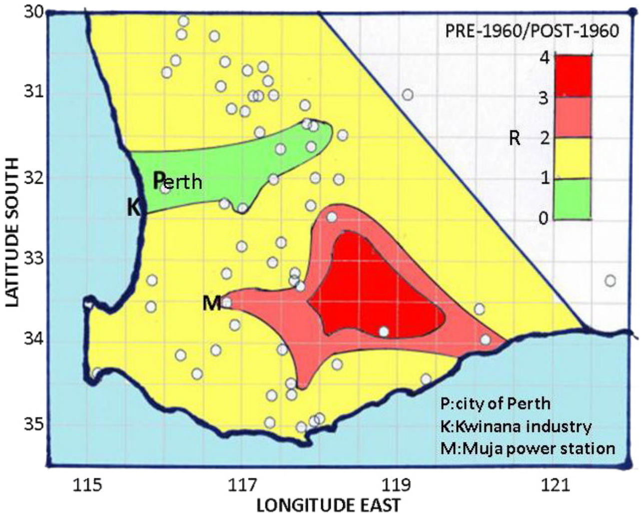

Figure 7. Contours of R (ratio of pre-1960 to post-1960 CD_{FH}) for the sites in Table 3.

CD_{FH} for each site, for the entire period of records, are shown in Table 3. R and its contours were calculated in exactly the same way as in Sect. 5.2. Contour errors in the eastern sector will obviously be large because of the sparse sites. To indicate this probable unreliability, a portion of the diagram has been left unshaded. The values of R at the two sites within the excluded zone were consistent with the extrapolations made from the more closely spaced sites. Rain-bearing winds in the whole area were predominantly from the western quadrant. A general slight increase in R over most of the area occurred after 1960, consistent with lower CD_{FH} post-1960. The exceptions were a slight decrease in R downwind from Perth, resembling that downwind from Melbourne, though not as widespread. The largest increases in R occurred downwind from the Muja power station. That also is consistent with the increases noted from power stations seen in Fig. 6.

5.4 Magnitude of the after-effects of heavy rain

CD_Q and CD_F were calculated separately for the entire period of records at each site. These were compared with the mean 20-day totals of rain quantity or frequency of occurrence of rain preceding key days to give an estimate of the magnitude of the apparent feedback effects. Figure 8 shows that changes in both quantity and frequency of rain were generally greater in the west than in the east and in both cases were greater in frequency than in quantity. Rainfall on key days and the associated 20 following days usually constitutes < 30 % of the total rainfall at low rainfall sites and as little as 10 % at high rainfall sites. The changes may be less substantial than they appear in Fig. 8 as a result, but more substantial if rainfalls below the chosen thresholds add to the effect.

5.5 How the choice of key day threshold influences CD_Q and CD_F

Following a rainfall, if there are comparable effects of key days with lower thresholds than those used here, Fig. 8 could be an underestimate of changes due to rain. In the southwestern group of sites, 24 had overall $CD_Q > 10\,\%$ and $CD_F > 16\,\%$ offering the best chance of detecting the relationship. The differences D_Q in rainfall quantity or D_F in rainfall frequency between totals in the 20 days following and preceding each key day were calculated separately for each key day, then grouped for all sites according to 315 blocks of key day rain in ascending order of key day rain. There was no consistent trend for key day thresholds from 20 to 70 mm. The variations within that range were on the order of only one standard deviation in D_Q or D_F in the groups of 315 and therefore probably not significant. The possibility remains that the effects of rain on subsequent rain are larger than demonstrated here, but it remains unproven.

6 Discussion

CD for IN and rain both have logarithmic changes with time from a key day lasting for comparable periods. This similarity would be very unlikely without a mechanistic relationship between IN and rain, i.e., that the IN were one of the main causes for the initiation or stimulation of rain. Hirano et al. (1996) and Huffman et al. (2013) found IN concentrations to increase after heavy rain in a period shorter than a day. If those added IN were directly implicated in the processes that led to additional rain, then no delays between changing IN concentrations and rain quantity or frequency would result if general meteorological conditions were favorable to rain. It could be argued that the close relationship between the logarithmic forms of CD for IN and CD for rain could be due to an unknown influence that affected both equally; however, we are not aware of a process that could have such an influence. A causal relationship between IN and rain is the simplest explanation that we can propose. Reasons why such a relationship should exist will now be discussed.

6.1 Biological influences on both IN and rainfall

Hirano et al. (1996) showed that leaf populations of IN-active bacteria increased from 10-fold to 1000-fold following intense rain. Huffman et al. (2013) also showed a large increase of airborne IN in response to rain in a period shorter than one day. The assumption made here is that the number of epiphytic IN bacteria that become airborne is, on average, proportional to the epiphytic population size. After rain has ceased, the conditions necessary for emigration of the enlarged epiphytic population of bacteria from leaf to air ensure that airborne concentrations will actually fluctuate greatly from day to day. It is for this reason that to detect prolonged trends in airborne concentrations following

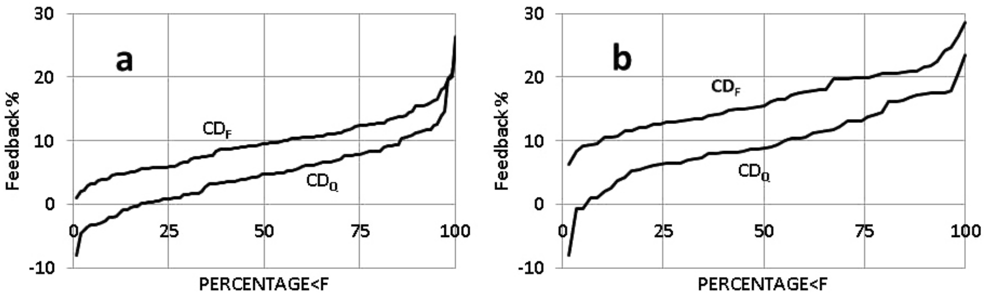

Figure 8. Comparison of CD_Q and CD_F with mean 20-day **(a)** rainfall quantity and **(b)** frequency of occurrence, preceding key days.

heavy rain it is necessary to use averaged concentrations synchronized to days with rain. Based on those assumptions, the logarithmic form of CD for IN could be due to an average slow exponential decay of the enlarged bacterial population resulting from, for example, migration or death. It is generally accepted that IN are involved in one of several processes by which rain is initiated. The similar responses of IN and rainfall to days of heavy rain demonstrated in Sects. 4 and 5 are therefore consistent with a feedback effect in which rain increases atmospheric IN concentrations, which can then initiate subsequent rain.

Not only the bacterium *P. syringae* may be involved in after-effects of rain. Huffman et al. (2013) found that airborne concentrations of many species of bacteria and fungi also increased strongly during and briefly following rain. Overall, this is an expected effect of free moisture on microorganisms, as water is a limiting factor for proliferation of microorganisms on plant surfaces. While many of these microorganisms were IN, others were relatively inactive as IN but because of their size and wettability most would have been active as GCCN. The extent to which rain would lead to sustained increases in GCCN depends on whether their epiphytic population densities were enhanced and whether subsequent transfer to the atmosphere was proportional to those densities. A contribution of GCCN to prolonged rain-induced changes to subsequent rainfall is a possibility. One indication that it may be real is provided by the oscillatory changes in CD_F shown in Fig. 5a and c and in CD for IN in Fig. 3 (left). The time delay between rain-induced germination of spores and subsequent spore release would provide periodic increases in GCCN. The discovery that urediospores of rust fungi are capable of acting as efficient IN (Morris et al., 2013) revealed an abundant non-bacterial agent that could influence precipitation as both IN and GCCN. Emission of fungal spores by rain and their deposition elsewhere is usually responsible for the spread of many plant diseases. This is the case in particular for the rusts of grains, diseases that have been widespread and of major importance in Australia since the large-scale cultivation of grains (Park, 2008). *Puccinia* species attacking grain crops produce spores capable of re-infecting the host plant. Potentially this could lead to recurrent maxima in spore release following an infestation triggered by rain. We speculate that the maxima of CD_F in Fig. 5 might have arisen in this way from fungi having that property. Pollen formation and release trigged by rain could potentially provide even further delayed after-effects of rain if the pollen served as GCCN.

Drizzle may fall from shallow or warm clouds if GCCN concentrations are sufficient, without adding appreciably to the quantity of rain. CD_F may therefore be more influenced by enhancements of GCCN than of IN following rain, while CD_Q may be more influenced by IN that create ice crystals in deep supercooled clouds. Anticyclonic conditions often follow rain, and shallow warm clouds will be more likely to be present than deep supercooled clouds. This may account for the observed tendency shown in Fig. 8 for CD_Q to show a smaller proportional response to rain than CD_F.

6.2 The influence of coal-based power stations on CD_{FH}

The large numbers of PM_10 particulates emitted by the Hazelwood (Victoria) and Muja (Western Australia) power stations will rapidly become coated with the oxidation products of SO_2 that is simultaneously emitted. This means that an enhancement of biological GCCN concentrations due to rain will represent only a small proportion of GCCN always present downwind from the station, unlike the situation in a clean environment. Consequently, the influence of biological GCCN on initiating subsequent rainfall will be considerably reduced and R will have increased after the power stations were installed. CCN and IN enhancements due to the emissions can also alter the potential for rain. CCN concentrations will be very large and will tend to decrease the probability of rain formation but the presence of large concentrations of GCCN could reverse the effect. The observed changes in R suggest the latter to be the case. IN enhancements (if any) are unknown. Extensive cloud microphysical measurements in and near affected areas would be needed in order to specify the dominant cause of the CD_{FH} decrease downwind from power stations.

6.3 The influence of metropolitan areas and associated industries on CD_{FH}

Measurements of condensation nuclei (CN; particles of unspecified sizes and properties) concentrations from an aircraft as a function of altitude in an arc of radius 167 km centered on Perth airport in a southwesterly air stream revealed three broad regions downwind where concentrations greatly exceeded those in the background (Bigg and Turvey, 1978). These regions were downwind from sources in the metropolitan area, Kwinana (K in Fig. 7) and the Muja Power station (M). A very narrow plume of high concentration was also found close to the town of Bunbury on the coast south of Perth. At Kwinana the most important sources at the time of the measurements were an oil refinery distilling sulfur-rich Middle East oils and a nearby ammonia factory. Many of these anthropogenic CN would have become CCN at 167 km from the source due to deposition of sulfate and reaction with ammonia. However, in the past 30 years their sources have diminished as a result of clean air policies and this could have contributed in more recent years to the observed decline in R downwind from Perth or Melbourne. The metropolitan areas in both the southeastern and southwestern groups are large oases, well watered compared to their surroundings and having much imported flora. Observations are needed to determine whether a difference in the populations or properties of associated microorganisms to those in the downwind areas might also contribute to the downwind decrease in R.

6.4 The influence of irrigation areas on historical changes in R

Figure 6 shows a substantial area along the northern border of the diagram where R increased with time. Most of the sites lay along the Murray River or its tributaries and irrigated crops and pastures were numerous. It could be speculated that the expansion and intensification of irrigation led to more favorable habitats for IN-active bacteria, increasing CD_{FH}. If that is the case, the same hypothesis as suggested above for the metropolitan areas might apply.

6.5 Sites with anomalous CD_Q and CD_F

Several sites had CD_Q or CD_F that differed greatly from those nearby. Further research will be necessary to determine the cause of the anomalies but some speculative hypotheses may be useful in directing future research. The highest CD_Q was at Mt. Buffalo Chalet (site 83 073, Table 2). Its elevation is 1350 m and it is located close to the base of a 1720 m rocky peak. Convective activity induced by uplift over a sunlit mountain leads to more clouds with more reaching subzero temperatures than over surrounding plains (Cotton et al., 2011). IN will therefore more often be involved in initiating precipitation in the vicinity of an isolated peak and this could be the reason for the unusually high CD_Q. Other high-altitude sites such as 70 067 and 71 000 (Table 2) situated on bare flat plateaus where there was no local uplift to enhance cloud formation did not have unusually high CD_Q. Another site with much higher CD_Q and CD_F than those of the nearest sites was beside a large reservoir at 400 m altitude in a deep valley surrounded on three sides by forested mountains rising to 800 m. The presence of a large body of water nearby and the shelter from strong winds provided by the surrounding mountains would probably have prolonged conditions favorable for enhanced biological IN populations following heavy rain.

An unusual combination was at a site where one of the highest pre-1960 values of CD_F was accompanied by a negative CD_Q. After 1960, both CD_Q and CD_F were positive but very low. The site was in a large vineyard planted in 1889 that progressively in the 20th century became part of a much larger grape-growing area. The very low values of both measures of CD post-1960 also differed from those in the surrounding area. Mean rainfall frequency before key days was 32 % greater after 1960 than before it, while a nearby site showed only an 8 % difference. The difference was evidently due to local influences rather than a climatic shift. One speculative cause is spores of powdery mildew, a common fungal disease of grapes. Unlike most diseases it is favored by dry weather. Pre-1960, powdery mildew spores might therefore have more often provided GCCN populations sufficient to initiate drizzle, increasing CD_F and reducing CD_Q, than post-1960. Increased use and effectiveness of fungicidal spraying could also have contributed to the changes.

Relatively high pre-1960 CD values were associated with wheat belt areas in both the southeastern and southwestern sites. Grain crops might enhance concentrations of IN bacteria and fungi as several of the various bacterial pathogens attacking the aerial parts of the plant, including various *P. syringae* pathovars of wheat and *Xanthomonas campestris* pv. translucens, are ice nucleation active (Kim et al., 1987; Mittelstadt and Rudolph, 1998), as are urediospores of cereal rusts (Morris et al, 2013). It might be productive in future research to try to relate changes in CD with time to changes in varieties of grain or pathogen control measures.

One further observation that is puzzling is that three lighthouse sites exposed to the prevailing rain-bearing winds (Capes Leeuwin and Naturaliste in the southwest corner of Western Australia and Northumberland in the south-eastern group) had very little land upwind, yet showed higher than average values of both CD_F and CD_Q. Active bacterial IN have been found in seawater (Fall and Schnell, 1985) and these could be released to the atmosphere by the bursting of rain-induced bubbles in seawater. It is not obvious why this should lead to any effects after the rain has ceased unless they colonize land vegetation.

7 Conclusions

The results presented here represent an analysis of one of the very few sets of data on long-term measurements of IN. This led us to show that not only do concentrations of airborne IN increase on the day after rain but these increases persist for up to about 20 days, even though their rate of production declines exponentially with time. By comparing the patterns of the increase of IN after rainfall to those of the quantity and frequency of rainfall following a heavy fall of rain, we showed a similarity of these patterns: rainfalls are increased relative to the preceding days for a similar length of time and with similar exponential decreases with time as the changes in IN concentrations. The patterns of increase in rainfall are robust in that they are founded on century-long data series. The simplest hypothesis to explain the similarity is feedback: rain creates more airborne biological IN that then initiate more rain. Although the rain derived from feedback is small compared to total rainfall, it may be an underestimate because the analysis does not lend itself to dealing with the effects of more frequent but lighter falls of rain than those considered here. The effectiveness of bacteria such as *P. syringae* in nucleating ice, combined with their known responses to rain and the possibility that biological IN contribute to rapid ice crystal multiplication in some clouds, supports the notion that they are major factors involved in the feedback. Fungal spores could also contribute to feedback as either IN or GCCN. Overall, our results corroborate important links between microorganisms and rainfall that are stronger and persist for longer periods than previously described.

Our results also support the idea that particulate emissions from power stations have apparently reduced the feedback due to biologically derived rain-influenced particles. If the GCCN content of the particulate emissions increases the probability of precipitation, the influence of biological IN in increasing precipitation will be proportionally reduced. Furthermore, our results suggest that metropolitan areas, wheat-growing areas and irrigated regions provide more suitable habitats for ice nucleating microorganisms than natural vegetation, as inferred from the relatively large feedbacks associated with these areas compared to natural areas. Overall, the processes we describe here open exciting questions where direct microphysical and microbiological observations would add invaluable data toward elucidating specific mechanisms involved. The analytical methods used in this work could be deployed for historical rainfall data on other continents to identify a sufficient number of sites with comparable behaviors to test hypotheses about the effect of environmental context on rainfall feedback patterns.

Acknowledgements. Funding for the publication of this research was provided by a donation from the Epicurus Fund at DonorsTrust to Montana State University.

Edited by: A. Huffman

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