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Rainfall feedback via persistent effects on bioaerosols

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Abstract

Consistent temporal differences between ice nucleus concentrations after and before a heavy fall of rain have been found in four areas of Australia. Closely similar differences were found between rainfall quantity or frequency at 106 sites in south-eastern and 61 sites in south-western Australia that had > 92 years of daily rainfall records. The differences suggest an impulsive increase in ice nuclei or in rain on the day following heavy rain that decreased exponentially with time and was often still detectable after 20 days. The similarity of ice nucleus concentrations, bacterial populations, bioaerosols and rainfall responses to heavy rain strongly corroborate the involvement of biological ice nuclei in a rainfall feedback process. Cumulative differences of after-before rainfall amount or frequency for each rainfall event were next combined to form a historical record of the feedback process for each site. Comparison of cumulative totals pre-1960 and post-1960 showed differences bearing apparent relations to upwind coal-fired power stations, growth of metropolitan areas and increased areas of cultivation of wheat. These observations suggested that fungal spores or other bioaerosols as well as ice-nucleating bacteria were involved in the feedback. The overall conclusion is that interactions between micro-organisms, bioaerosols and rainfall have impacts over longer time spans and are stronger than have been previously described.

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. Positive short-term feedback effects on rainfall have been predicted and attributed to changes in surface albedo and Bowen ratio 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). Rainfall could also have short term feedback effects due to influence on atmospheric

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concentrations of particles involved in rain formation, on cloud condensation nuclei (CCN) and ice nuclei (IN) in particular. For example, the concentration of airborne biological particles and IN in a forest eco-system increased by an order of magnitude during rain and up to one day thereafter in periods of extended leaf wetness (Huffman et al., 2013). The IN increases were associated with increases in atmospheric concentrations of various micro-organisms including bacteria and fungi. Many of these were also giant CCN (GCCN, defined as CCN > 2µm in diameter) capable of broadening the size distribution of cloud drops and thus assisting in the formation of rain by the coalescence process. The authors speculated that the airborne bioparticles ("bioaerosols") could trigger subsequent rainfall, leading to the conclusion that bioaerosols, IN and rainfall might be more tightly coupled than previously assumed. Whatever the subsequent impact of these particles on rainfall, the significant result of these and similar studies is that rainfall enhances concentrations of airborne biological particles with physical properties that can influence atmospheric processes. There are no known phenomena by which mineral or inert CCN, GCCN or IN could be enhanced in this way.

As further support for this process, it has been demonstrated that population increases from ten-fold to thousand-fold of the ice nucleation active bacterium *Pseudomonas syringae*, on leaves of a snap bean crop, occurred after intense rain for a period of up to three days (Hirano et al., 1996). Some of these bacteria can become airborne due to impact of raindrops and other processes involved in aerosolization. Growth of vegetation that harbors ice nucleation active microorganisms such as *P. syringae* is stimulated by rain and would provide an increased habitat prolonging the initial stimulation. *P. syringae* cells are preferentially lifted into the atmosphere during the warmest part of sunny days when leaves are dry and wind speeds $> 1\,\mathrm{m\,s^{-1}}$ (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.

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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 $-15\,^{\circ}C$ 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. The first indication that IN concentrations might have a longer relation to rainfall than a few days was found in 1956 (Bigg, 1958) when a spectacular but fluctuating increase lasting at least 25 days followed an intense rainfall of 135 mm in less than 24 h.

Another approach to revealing feedbacks on rainfall involved an analysis of decade-long records of daily rainfalls from many sites. This analysis 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 (Soubeyrand et al., 2014).

These observations evoke questions about the underlying processes and if local atmospheric concentrations of CCN, GCCN and IN are involved. There are numerous ways in which CCN, GCCN and IN of biological origin may affect rainfall (Möhler et al., 2007). Typical concentrations of CCN active at 0.3 % in maritime situations are on the order of 50 cm^{-3} and in continental situations, 300 cm^{-3} (Twomey and Wojciechowski, 1969) whereas IN and GCCN are usually at least four orders of magnitude fewer. Because of this disproportion, changes in IN and GCCN concentration due to rain are likely to have much more influence on subsequent rain.

Increased IN concentrations in clouds colder than $0\,^{\circ}C$ 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 $> 24\,\mu m$ at temperatures between -3 and $-8\,^{\circ}C$ (Hallett and Mossop, 1974; Crawford et al., 2012). Enhancement of atmospheric IN in this temperature range could result from emissions

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of various biological ice nucleators such as *P. syringae,* while enhanced GCCN concentrations could provide the relatively large cloud drops. Increased GCCN concentrations also do not necessarily lead to more rain, although increasing its frequency. In shallow clouds, the formation of drizzle can reduce their water content, potentially decreasing the amount of rain. The importance of these different factors is most likely specific to different sites and seasons, therefore it is important to identify regions and periods of time where rainfall occurs in positive or negative feedback cycles in order to elucidate the details of the underlying processes.

The overall aim of this 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 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 subsequent 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 south-eastern and south-western 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 Methods

2.1 Data manipulation

IN concentrations fluctuate according to meteorological conditions such as mixed atmospheric depth, wind strength and rain. Rainfall is even more variable, so that to detect any consistent temporal changes longer than a few days that might be related to a heavy fall of rain, it is necessary to use very long data sequences and to combine

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each of the individual events. This is known as the superposed epoch method that has often been used to detect periodicities in geophysical and meteorological time series (e.g. Singh, 2006).

We defined as “key days” the days on which rainfall exceeded a certain threshold. The raw data were daily sequences of IN concentrations, rainfall intensities, or days with any rain. These were transformed into time series of differences of those values on a given number of days after a key day to those on the same number of days before it. Averages over the whole of the data sequence under examination were then formed. Fluctuations with time still occurred, particularly in the case of rainfall intensity data. Formation of cumulative differences (CD) as a function of days from a key day averaged over the whole data sequence under examination reduced those fluctuations to a level where any consistent trends that occurred were revealed. Soubeyrand et al. (2014) have described the method in detail for rainfall, including the determination of the statistical significance of any average trends with time that are found and problems resulting from changes of those trends on a long time scale.

At sites having highly seasonal rainfall, part of the CD in rainfall after-before a key day can arise if key days are not symmetrically distributed about the rainfall maximum. In some cases this artefact is important. To assess it, the mean rainfall for each day of the year was calculated for each site. The analysis was conducted using actual key days, but the rainfalls in the 20 days preceding and following a key day were the mean rainfalls of the entire series for the corresponding calendar dates. In this way, CD due to the seasonal effect alone was revealed. It increased approximately exponentially with time from a key day and was nearly always positive. This suggests that key days predominantly occurred before the annual maximum in rainfall. By subtracting the new series from the original, bias from this cause was eliminated. Many south-western sites had short wet seasons and the seasonal bias was as high as 30 % of the original series at 20 days from a key day.

2.2 Sources of the data

Data on IN concentrations come from records held by the first author. Results from four sequences will be presented. The first (Bigg, 1958) consisted of three months of daily average measurements at −20 °C using a 10 L mixing cloud chamber at a single site, 26.6° S, 153.2° E. The remaining three all used continuous sampling to collect daily 300 L air samples on filters. The IN on each filter were detected by lowering the temperature to −15 °C, raising the humidity to water saturation and counting the ice crystals that resulted. Multiple sites spread over wide areas for periods of 18 months to three years were used. The filter method (Bigg et al., 1963) does not detect contact nucleation and so tends to underestimate concentrations. The presence of large hygroscopic particles near a potential IN on a filter reduces the relative humidity below water saturation and their growth can result in immersion of the nucleus, again leading to underestimates. However, in unpolluted sites away from the sea the method gives reliable relative concentrations and can be operated with minimal attention and cost at multiple sites for long periods.

Rainfall records used are daily totals listed by the Australian Bureau of Meteorology. These are now readily available online from www.bom.gov.au/climate.

3 Persistent effects of rainfall on IN concentrations in the atmosphere

3.1 Measurements of IN with a 10 L mixing cloud chamber at −20 °C at a single site, 26.6° S, 153.2° E, (Bigg, 1958)

From early November to 21 December 1956 only a few light falls of rain occurred but on the night of 21 December and morning of 22 December 135 mm of rain fell in a period of about 12 h. A spectacular increase in IN concentrations followed, slowly dying away though fluctuating through January. There were no further large falls of rain during this period. Using the method described in Sect. 2.1, the CD in IN concentration after-

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before the key day, 22 December, as a function of the number of days from the key day are shown in Fig. 1.

3.2 Measurements of daily average IN at −15 °C at 24 sites over eastern Australia for periods of 18 months to 3 years

The experiment was described by Bigg and Miles (1964). IN concentrations at −15 °C at 1.5 m above the ground surface were estimated by the membrane filter method. Figure 2a shows the location of the sites which operated for periods of 18 months to three 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. For this analysis, key days had rainfalls ≥ 25 mm. Figure 2b shows the mean CD in IN concentrations after-before a key day as a function of time from the key day. A logarithmic curve fits the data well ($R^2 = 0.89$) except for days 4 to 6.

3.3 Measurements of daily average IN concentrations at −15 °C at 8 sites for three years

From 1987 to 1990 measurements of IN concentrations were made at eight well-spaced sites in the area bounded by latitudes 37.6–38° S, and longitudes 144.9° −147.5° E. These were in conjunction with a cloud seeding experiment in Victoria, (Bigg, 1995; Long, 1995). Four were in forested (eucalyptus) sites and the remainder were in open woodland or pasture. The seeding agents used were silver iodide or dry ice (frozen carbon dioxide) dispensed from aircraft in clouds. Seeding was carried out on only 11 % of the occasions in which mean rainfall in the area exceeded 10 mm due to unsuitability of the clouds for seeding, randomization of seeding, or the time at which rain fell and more of these occasions used CO_2 than AgI. There is some evidence that AgI deposited on the ground enhances bacterial ice nucleation over a much longer period than three weeks (Bigg, 2015) and therefore might have contributed to

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the changes seen in Fig. 3a. Key days in the IN analysis had rainfalls ≥ 10 mm as there were an insufficient number of key days if heavier rainfalls were used. The cumulative after-before differences revealed recurrent peaks on a logarithmic trend (Fig. 3a)

3.4 Measurements of daily average IN concentrations at −15 °C at 5 sites for three years

IN measurements in conjunction with a dry ice cloud seeding experiment in Tasmania (Searle, 1993) were made for three years at −15 °C using the filter method. The five remote forested sites were in an area bounded by latitudes 41.4–42.4° S and longitudes 145.5–146.8° E. Key days had rainfalls ≥ 10 mm as for the previous case. The cumulative after-before differences are presented in Fig. 3b and again showed a logarithmic trend (R^2 = 0.97) extending to at least 20 days.

3.5 Conclusions concerning the effects of rainfall on IN concentrations

In each of the four separate experiments, IN concentrations were on average enhanced following a heavy fall of rain and the enhancement diminished exponentially with time over a period of about three weeks. These coherent results across numerous sites based on different methods of measuring IN concentrations strongly support that these results represent a real effect. Hence, this prolonged enhanced IN concentration is therefore a potential cause of the differences in rainfall after-before a key day with heavy rainfall found by Soubeyrand et al. (2014). In the next section, extensions to the data set of Soubeyrand and co-workers will be used to investigate the relationship further.

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4 Changes in cumulative differences (CD) in rainfall after-before a heavy fall of rain

4.1 Calculation procedures

The CD in rainfall after-before a key day can be called a feedback factor, denoted by F. Two possible variables in the determination of F are rainfall quantity (F_Q) and rainfall frequency (F_ν) (occasions with > 0 mm of rain). Large random fluctuations in F_Q can result from extreme daily rainfalls. F_ν reduces the fluctuations in the series because after-before differences are capped to 20 instead of being unlimited. The two measures are not necessarily equivalent. Rainfall frequency will contain drizzle events that contribute very little to total rain, and therefore it is partly an indication of initiation of rain, while augmentation may be more important for rainfall amount. F_Q or F_ν for each individual event can be combined to form cumulative historical series F_{hQ} or $F_{h\nu}$ that test the stability of feedback with time.

4.2 Areas chosen for detailed analysis

A total of 106 sites in south-eastern Australia (mainly in Victoria) and 61 in the south western part of Western Australia were selected for analysis. In superposition analyses it is important to have many key days in order to reduce random fluctuations. However, the sensitivity for detecting a repeated signal is reduced if more than one key day occurs within the 41 day sequences used here. A compromise was to select only the 300 days of highest rainfall as key days. A lower limit of 20 mm for a key day was imposed so that at some low rainfall sites the number of cases was less than 300.

4.3 Results

We have seen that there was an approximately logarithmic increase in cumulative after-before key day differences in atmospheric IN concentrations up to at least 20 days after rain. Using the same type of analysis for F_ν, logarithmic increases were again

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almost universal. Figure 4a (south-eastern group) shows the mean trend of F_ν with time after a key day for the 22 % of sites that followed the logarithmic trend most closely ($R^2 = 0.99$). For a further 22 % of sites $R^2 = 0.97$. Such close fits are certainly highly significant. Similar mean logarithmic trends ($R^2 = 0.97$) were found for 60 % of sites in the south-western group of sites, shown in Fig. 4b. At many of the remaining sites, the logarithmic trend terminated before 20 days.

The regularity of the logarithmic increase of F_ν is entirely consistent with the corresponding results for atmospheric IN concentrations seen in Figs. 1 and 3b and to a lesser extent those in Figs. 2b and 3a. The extent of this agreement suggests that IN are an important factor in what amounts to a rainfall feedback effect.

An unexpected effect was found at 10 % of the sites (mainly near-coastal sites) in the south-east and at 18 % of the sites in the south-west. Means of F_ν at these sites are shown in Fig. 5a (south-east), b (south-west uncorrected), and c (b corrected for the seasonal artefact). The oscillatory nature could arise from a series of pulses at intervals of 5 to 7 days superimposed on the logarithmic curves of Fig. 4a. Figure 3a also showed three pulses in IN concentrations following key days, though less regular than those of Fig. 5.

4.4 The ratio of pre-1960 to post-1960 historical cumulative time series $F_{h\nu}$ and F_{hQ}

In preliminary analyses, (Soubeyrand et al., 2014) marked shifts in the trend of F_{hQ} and $F_{h\nu}$ at numerous locations were noted 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. Therefore, data sets were divided in half at this date. A similar series for rainfall quantity will be called F_{hQ}. Ratios of pre-1960 to post-1960 $F_{h\nu}$ for each south-eastern site are plotted in Fig. 6. Circles show the location of each site, a red star shows the location of a large complex of power stations fuelled by brown coal and the associated coal mining activities. The first very large power station (Hazelwood) was built between 1964 and 1971 and has been a notorious polluter. According to the National Pollution Inventory (www.npi.gov.au) of 2005–2006, this power station emitted 2.9×10^6 kg of PM_{10} particulates 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 but, while their individual particulate emissions have been less, their combined SO_2 output has been considerably greater. Melbourne's population grew from about 900 000 in 1900 to 1.5 million in 1956 and 4 million in 2010.

The main precipitation-bearing winds in the vicinity of the power stations are from the southwest (Long,1995) and pre-$F_{h\nu}$/post-$F_{h\nu}$ decreased downwind from the power complex after the first large station was in operation. There was also an increase in $F_{h\nu}$ in and downwind from the metropolitan area after 1960. The yellow and red areas in the west and extending towards the northeast coincide with the pattern of the main areas cultivating wheat. A major change in the type of wheat grown followed the loss of UK markets after 1960, with hard grain white varieties being introduced for exports to Asia (www.abs.gov.au). This was coupled to a policy to cultivate only varieties that were resistant to rust diseases and especially after the devastating stem rust epidemics of wheat in New South Wales in the 1970's (Park, 2008). These events might be pertinent to the changes in feedback trends in light of the fact that urediospores of rusts are emitted into the atmosphere during disease epidemics and are highly ice nucleation active (Morris et al., 2013).

Contours of pre-1960 $F_{h\nu}$/post-1960 $F_{h\nu}$ were constructed by interpolation between pairs of neighboring sites. Sites are not closely spaced in many areas of the south-western group, so errors in the contours are certain to be present. However, Fig. 7 suggests a similar depressing effect on pre-1960 $F_{h\nu}$/post-1960 $F_{h\nu}$ downwind from the major coal-fired Muja power plant to that downwind from the power stations in the south-eastern group. The predominant rain-bearing winds were from the west. Muja (M in Fig. 7) opened in 1966 and has rivalled Hazelwood in the south-eastern group as one of the largest particulate emitters in Australia. Perth's population was 100 000 in 1911, 400 000 in 1961 and 1.5 million in 2010 and appears to have been associated

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with a downwind increase in pre-1960 $F_{h\nu}$/post-1960 $F_{h\nu}$ similar to that downwind from Melbourne. However, the industrial complex of Kwinana is also upwind from Perth. Its power station originally used oil, changed to coal in 1973 and to natural gas in the mid-1980s. Its potential influence on rainfall feedback is therefore difficult to assess.

4.5 Magnitude of the after-effects of heavy rain

For each site, the rainfall quantity and frequency were summed separately for the 20 days before and the 20 days after each key day. The difference was then expressed as a percentage of that in the 20 days before a key day and averaged over each entire series. Figure 8 shows that the feedback was generally greater in the west than in the east, and in both cases was greater for rainfall frequency than rainfall quantity. An important question is how feedback is influenced by key day rainfall. If it remains appreciable for key days of lower rainfall than those used, Fig. 8 could be a serious underestimate. In the south-western group of sites, 24 had $F_Q > 10\,\%$ and $F_\nu > 16\,\%$ offering the best chance of detecting the relationship. Combining all 24 sites, Fig. 9a shows peaks at about 28 and 41 mm in F_Q, falling steeply for key days with < 20 mm rainfall. Separating the sites into two groups with annual rainfalls < 400 mm (11 sites) and > 750 mm (5 sites) showed that the 28 mm peak was due to the dry sites and the 40 mm peak to the wet ones. F_ν varied less with key day rainfall and also showed small maxima at about 28 and 40 mm. Extrapolation suggests that F_Q would become negligible at arid sites for key days below about 15 or 20 mm for wetter sites, while F_ν might continue to remain near 15 % for much lower key day rainfalls.

5 Discussion

Here we have described changes in IN concentrations that lasted for weeks following rain. The changes fluctuated and this might explain why they have not previously been noted. To validate and characterize the trend in these post-rain changes in IN concentration, long-term continuous measurements of cumulative concentration differences after-before the rain event are required. Access to such data allowed us to reveal the underlying logarithmic trend of these changes in concentration. The consistency of the trends in both IN and rainfall strongly supports that the phenomenon we report here is real. We present arguments that the main cause for these trends is a sudden increase in airborne IN concentrations on the day after rain that then, on average, decays exponentially with time. Only biological particles, that could multiply as a consequence of rainfall, could be candidates for such IN. Previous research has shown that biological IN contribute to the increase in atmospheric IN observed soon after a rainfall (Huffman et al., 2013). IN active bacteria such as *P. syringae*, whose population size on leaves is known to rapidly increase after rainfalls > 20 mm (Hirano et al., 1996), could be one of the biological IN that contribute to the phenomenon observed here. After the rain has ceased, the conditions necessary for emigration of that bacterium from leaf to air ensure that airborne concentrations will fluctuate greatly from day to day. In the absence of a further stimulus, an exponential decrease in the inflated population seems probable and corresponds to observations of population dynamics of this bacterium in the field (Hirano and Upper, 2000).

Are IN solely responsible for the feedback? IN concentrations mainly influence the quantity of rain falling from deep supercooled clouds while GCCN can initiate rain through the coalescence process in clouds warmer than 0 °C. Drizzle may fall from shallow clouds if their concentrations are sufficient, without adding appreciably to the quantity of rain. Differences in the response of F_Q and F_v to enhancements of IN and GCCN following rain might therefore result. Huffman et al. (2013) showed the wide range of bioaerosols released to the air by rain. Those whose leaf-surface populations are enhanced by the rain, e.g. fungi, could potentially contribute to feedback through their GCCN properties if they become airborne, even if they are inactive as IN. Many types of bacteria that do not have appreciable ice nucleating properties can act as GCCN because of their hydrophilic surfaces that prevent desiccation and their relatively large sizes. Fungal spores that germinate during rain will also later emit GCCN

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spores. A more delayed influence of rain on GCCN may come from pollen released by plants because of rain. There is therefore a strong probability that both biological IN and GCCN are involved in the rainfall feedback process. The different responses of F_Q and F_v to key day rainfalls seen in Figs. 8 and 9 suggest such a dual influence.

The repeated maxima observed in Figs. 5a, c and 3a may also require an explanation other than the enhancement of populations of IN active bacteria such as *P. syringae*. 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 and suggests a possible mechanism for the pulsations in F. Because the spores are large and wettable they should also act as 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 could speculate that the maxima of F_v in Fig. 5 might have arisen in this way from fungi having that property.

5.1 The influence of coal-based power stations on $F_{h\nu}$

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 simultaneously emitted SO_2. This means that an enhancement of biological GCCN concentrations due to rain will represent only a very small proportion of those 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. CCN and IN enhancements due to the emissions can also alter the potential for rain. CCN concentrations will be very large but IN enhancements are unknown. Extensive cloud microphysical measurements in and near affected areas would be needed to determine whether GCCN ratios are the main cause of the $F_{h\nu}$ decrease downwind from power stations.

5.2 The influence of metropolitan areas and associated industries on $F_{h\nu}$

Measurements of CN (condensation nuclei – 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 south-westerly 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 and the Muja Power station. 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 last 30 years their sources will have diminished as a result of clean air policies. In Melbourne, an aluminum smelter at 38.4° S, 44.19° E that after 1969 emitted about 40 Mt of SO_2 per year, would have created similar very high CCN concentrations downwind. The most obvious difference between aerosol production by cities and power stations is the much larger number of GCCN produced by the latter but this does not explain the opposing effects on $F_{h\nu}$ in the two situations. The metropolitan areas both in the south-eastern and south-western 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 micro-organisms to those in the downwind areas might be responsible for the downwind increase.

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5.3 Sites with anomalous F

A number of sites had feedbacks that differed greatly from those nearby and had properties that suggested the cause. The highest F_Q was at Mt. Buffalo Chalet, at 1350 m, located close to the base of a 1720 m rocky peak. Convective activity induced by uplift leads to more clouds and more reaching sub-zero temperatures than over surrounding plains. 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 high F_Q. Other high altitude sites situated on bare flat plateaus did not have unusually high F_Q. Another site with much higher F_Q and F_v than those of the nearest sites was beside a large reservoir at 400 m altitude in a deep valley almost surrounded 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 F_v was accompanied by a negative F_Q. After 1960, both F_Q and F_v 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 F 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 suggesting the difference was due to local influences rather than a climatic shift. This suggests a possible reason for the anomalies that will need to be tested experimentally before being accepted. Powdery mildew is a common fungal disease of grapes and 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 F_v and reducing F_Q, than post-1960. Increased effectiveness of anti-fungal spraying could also have contributed to the changes.

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Relatively high F values were associated with wheat belt areas in both the south-eastern and south-western sites. Grain crops might enhance concentrations of ice nucleating bacteria and fungi as several of the various bacterial pathogens attacking the aerial parts of wheat, including various *P. syringae* pathovars 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).

One further observation that is puzzling is that three lighthouse sites exposed to the prevailing rain-bearing winds (Capes Leeuwin and Naturaliste in the south-west corner of Western Australia and Northumberland in the south-eastern group) with very little land upwind showed higher than average values of both F_v and F_Q. Active bacterial IN have been found in sea water (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.

6 Conclusions

The first important result from this work is that we show that concentrations of airborne IN not only increase on a day with rain but persist for up to about 20 days, even though their rate of production declines exponentially with time. Another important result is that rainfall quantity and frequency following a heavy fall of rain are increased relative to the preceding days for a similar length of time and with similar exponential decreases with time. This constitutes rainfall feedback. Although the rain derived from feedback is small compared to total rainfall, it is evidently 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. A major factor in the feedback is strongly suggested by 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. Fungal spores could also contribute to feedback as either

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IN or GCCN. Overall, our results corroborate important links between microorganisms and rainfall that are stronger and persist for longer than previously described.

Apparent effects on rainfall feedback of power stations are almost certainly related to particulate emissions. The provision of a more suitable habitat for ice nucleating microorganisms than natural vegetation may account for the relatively large feedbacks associated with metropolitan and wheat-growing areas. The processes we describe here open exciting questions where direct microphysical and microbiological observations would add invaluable data toward eludicating specific mechanisms involved.

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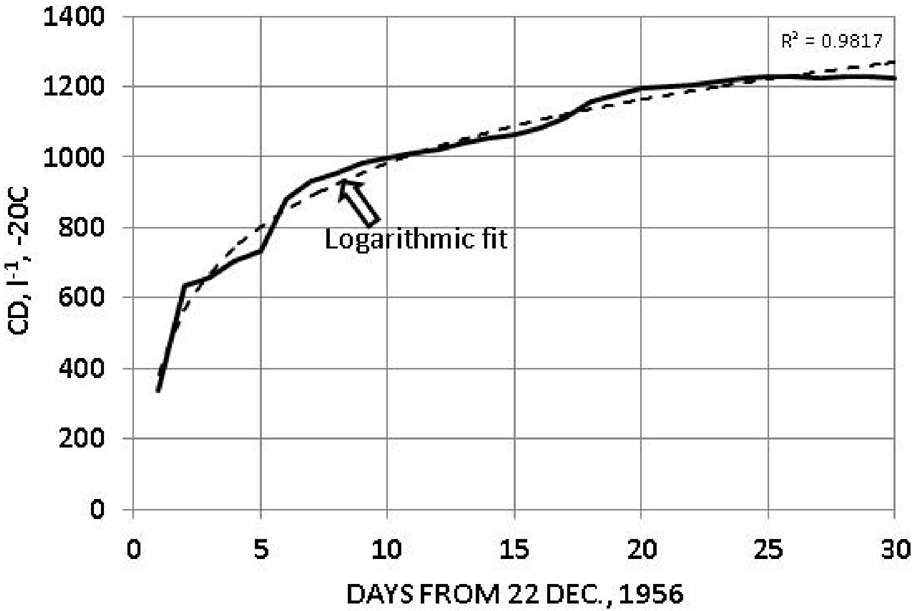

Figure 1. Cumulative differences per liter at −20 °C in mean daily IN concentrations between days d_i and d_{-i} from d_0, 22 December, a day with 135 mm of rain, where $-30 < i < 30$. A logarithmic fit to the data is shown.

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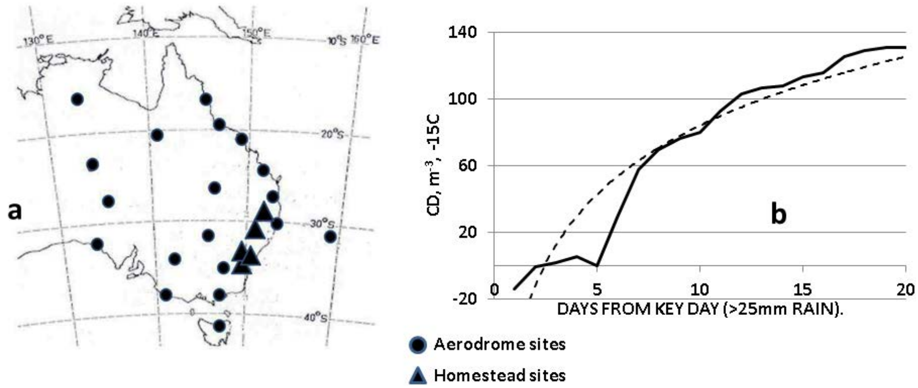

Figure 2. (a) IN measurement sites. **(b)** Cumulative differences, CD, in IN after-before key days having > 25 mm of rain in 24 h. A fitted logarithmic trend is shown, $R^2 = 0.89$. The daily mean concentration for days −20 to −1 from a key day was 15.3 m^{-3}.

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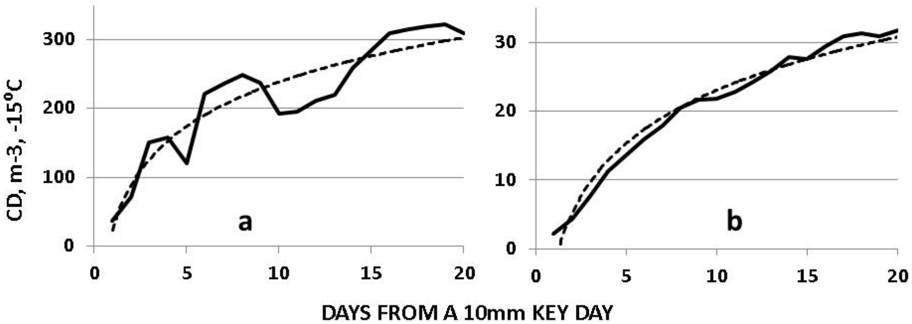

Figure 3. (a) Cumulative after-before key day differences in IN concentrations in Victoria as function of time from a key day, 1987–1990, **(b)** in Tasmania, 1992–1994.

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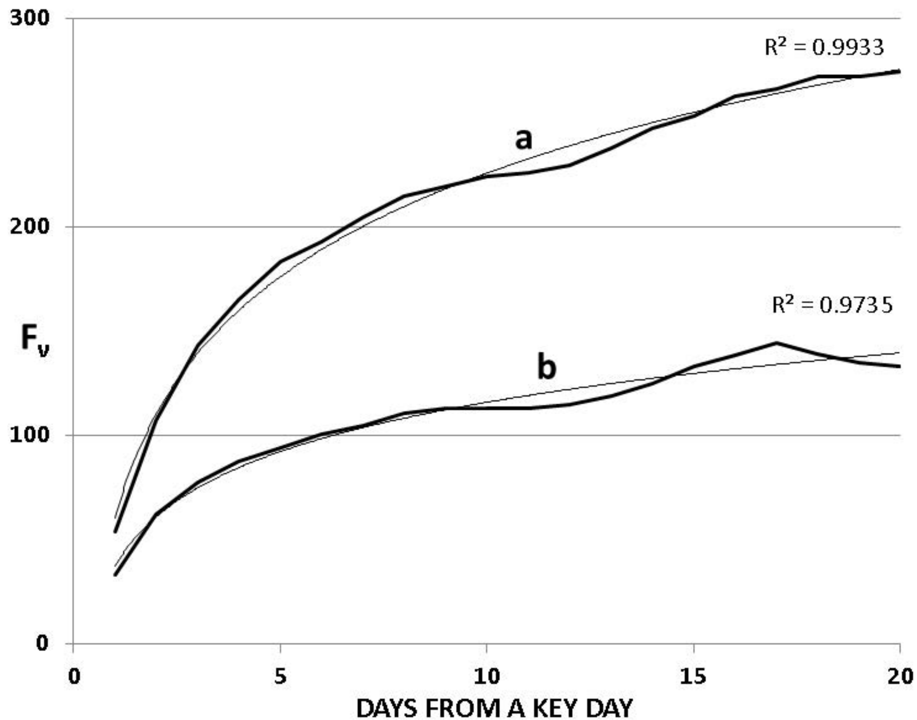

Figure 4. Mean trend in F_v following key days at 22 % of Victorian sites **(a)** and at 60 % of western Australian sites **(b)**. Fitted logarithmic curves are shown.

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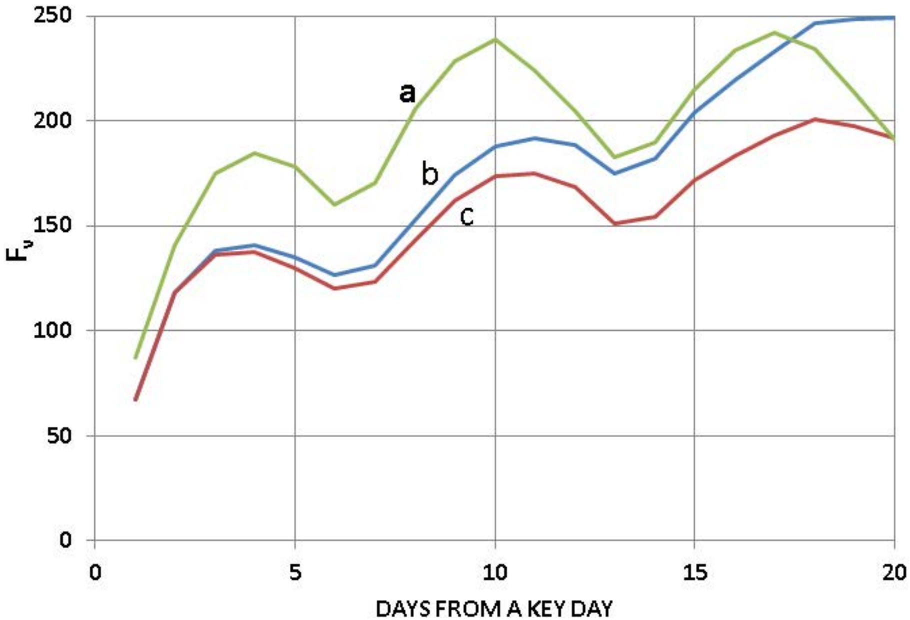

Figure 5. a: Mean trend in F_v following key days at 10 % of Victorian sites, b: at 18 % of Western Australian sites, c: b corrected for the seasonal effect described in Sect. 2.1.

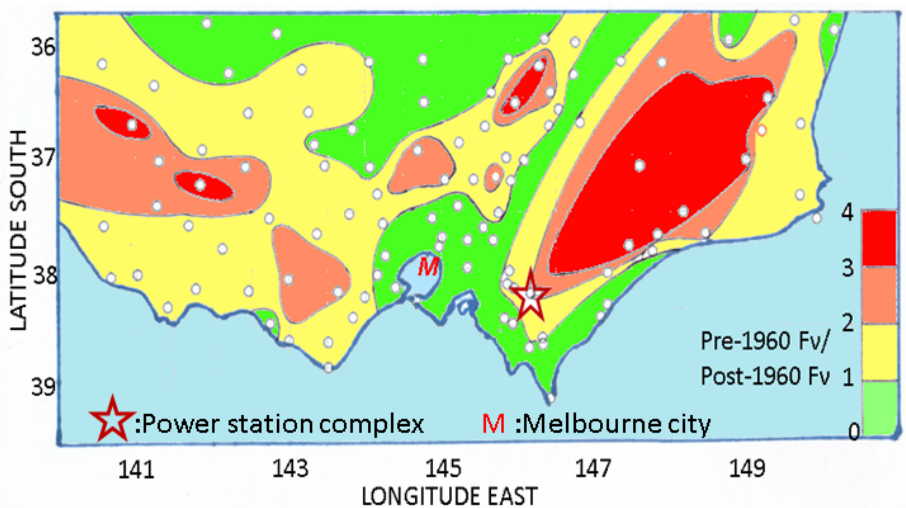

Figure 6. The ratios of pre-1960 to post-1960 cumulative historical series $F_{h\nu}$.

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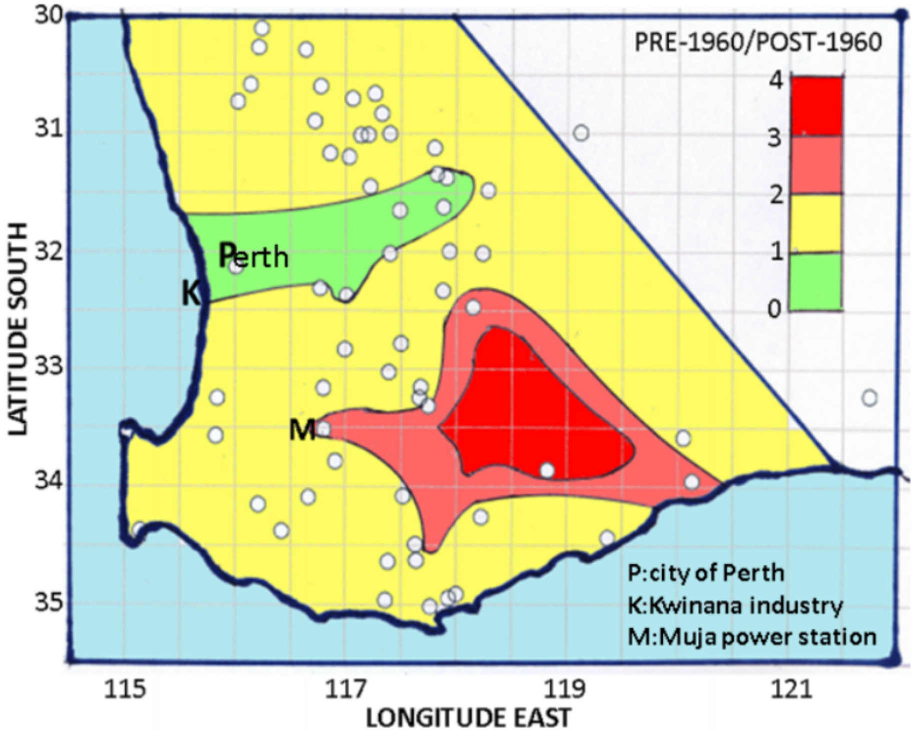

Figure 7. Contours of pre-1960 $F_{h\nu}$/post-1960 $F_{h\nu}$ in the Western Australia group of sites.

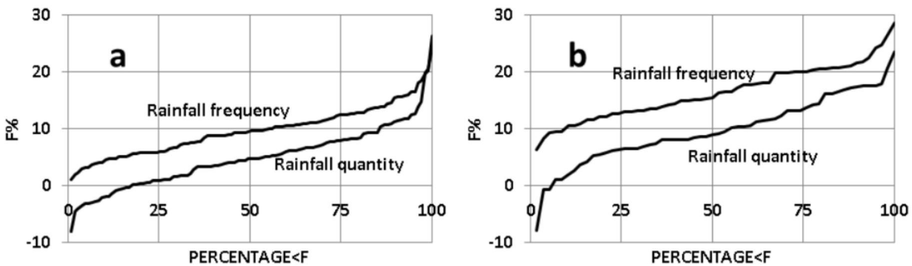

Figure 8. (a) Rainfall feedback F_Q and F_v for the south-eastern group of sites, **(b)** for the south-western group of sites, all years being considered.

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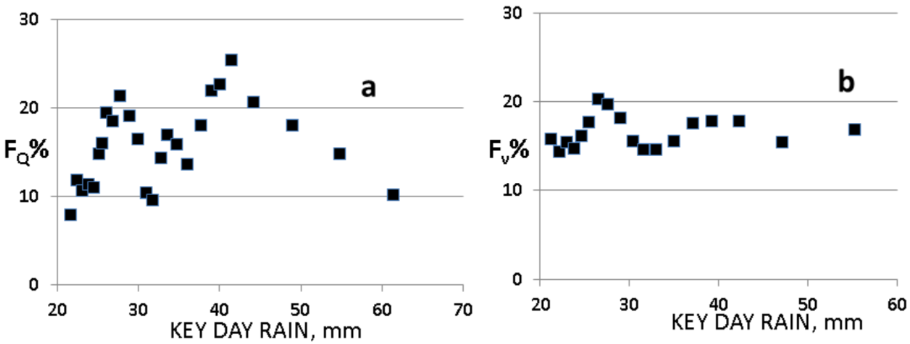

Figure 9. Rainfall feedback F_Q **(a)** and F_v **(b)** as a function of key day rainfall for 24 sites in the south-west group for which overall $F_Q > 10\,\%$ and $F_v > 16\,\%$.

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