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Tritium in Australian precipitation: A 50 year record

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SUMMARY

Tritium in precipitation has been measured in Australia over the past 50 years, as an essential research tool in hydro-climate studies, and to contribute to the Global Network for Isotopes in Precipitation (GNIP). Tritium, a component of the water molecule (HTO), is the only true age tracer for waters. The elevated level of tritium in the environment as a result of last century's atmospheric thermonuclear testing delivers the benefit of tracing groundwater systems over a 100 year timeframe. The concentration of tritium in Australian precipitation reached a maximum of 160 Tritium Units (TU) in 1963, during one of the most intense periods of nuclear weapons testing. From 1963 to present we observe a rapid drop in the concentration of tritium, more than expected from natural decay, and this can be attributed to the wash out of tritium into the oceans and groundwater. Spikes in the tritium level are superimposed over this general trend; the first around 1969, with levels ranging from 39.4 to 84.4 TU was due to French atmospheric weapon testing, and again in 1990, levels peaked between 6.6 and 12.9 TU, which is attributed to tritium leaking from underground tests in the French Pacific. Since 1990 the levels of tritium have declined globally and regionally. Currently the levels of tritium in Australia are stabilising to around 2–3 TU increasing with latitude across the continent, suggesting that today the tritium in precipitation is predominantly natural. The spatial distribution of tritium is presented and found to be dominated by the annual stratosphere–troposphere exchange in combination with latitude and continental effects. A precipitation amount effect is also observed for inland sites.

1. Introduction

Tritium (^{3}H) is the radioisotope of hydrogen with a half-life of 12.32 years (Lucas and Unterweger, 2000). Incorporated directly into the water molecule in the global hydrological system, tritium is the only direct tracer for groundwater dating (Cartwright and Morgenstern, 2012; Morgenstern and Daughney, 2012). Tritium is naturally produced in the atmosphere by cosmic ray spallation of nitrogen. It is also produced anthropogenically by a variety of nuclear activities (CNSC, 2009; Akata et al., 2011). By far the most significant release of tritium into the atmosphere was from atmospheric thermonuclear explosions which introduced tritium directly into the stratosphere, and resulted in an increase across the worldwide distribution (Begemann and Libby, 1957; Martell, 1963). Once in the stratosphere, tritium is rapidly oxidised into tritiated water (HTO) and transferred into the troposphere by stratosphere-to-troposphere transport in spring, where it is rapidly removed by precipitation (Morishima et al., 1985; Ehhalt et al., 2002) and then transported in the environment through the hydrological cycle.

This bomb-tritium input is an effective and useful environmental tracer for studying hydrogeological systems. For several years tritium concentrations in groundwater have been widely applied as a semi-quantitative tool for the identification of modern recharge from the post-1960s thermonuclear era (e.g. Allison and Hughes, 1975; Scanlon et al., 2002; Huang and Pang, 2010; Yangui et al., 2012). Lumped parameter modelling of tritium transport has also been used to quantitatively calculate the mean transit time and mean of the residence time distribution of water through an aquifer (Zuber, 1986; Zuber and Maloszewski, 2001). More recently tritium is increasingly being applied to calibrate and validate three-dimensional groundwater flow and solute transport models of aquifer systems at the regional scale (Orban et al., 2010; Zuber et al., 2011). With a levelling off of bomb-tritium levels in precipitation to natural levels, unique groundwater ages are now being obtained from tritium measurements (Morgenstern et al., 2010; Cartwright and Morgenstern, 2012; Morgenstern and Daughney, 2012).

Tritium concentrations in the atmosphere are known to vary across latitudes and between seasons (Hauglustaine and Ehhalt,

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2002). When atmospheric tritium concentrations are uniform, a continental effect is evident whereby tritium in precipitation at continental sites is higher relative to coastal sites due to (i) the dilution of atmospheric tritium with low concentrations in oceanic water (Momoshima et al., 1991; Vreca et al., 2006), and (ii) a larger proportion of water vapour inland being 're-evaporated', originating from terrestrial evapotranspiration, which generally has higher tritium concentrations than marine vapour. Harries and Calf (1980) measured tritium in Australian ocean waters in the late 1970s and found values ranging from 1 TU in the Southern Ocean south of Tasmania to 3 TU in the Tasman Sea off Sydney. Dilution and mixing of higher concentrations of tritium in rainfall within a water depth of 75 m (Bainbridge, 1963; Kakiuchi et al., 1999) and the efficient removal of tritiated water from the atmosphere by vapour exchange over the ocean (Eriksson, 1965) led to these low levels. Although no measurements have been recorded for the Australian region, the background levels of tritium in water vapour evaporated from the ocean would be slightly lower than these due to isotopic fractionation during evaporation, although fractionation of ~16% (Gat et al., 2001) is close to the measured uncertainty in tritium.

Tritium in precipitation is dominated by a seasonal cycle where a north to south mixing of air masses from the stratosphere to the troposphere occurs in mid-latitudes with a maximum in spring (Gat et al., 2001; Yasunari and Yamazaki, 2009). The time histories of tritium fallout, seasonal variation patterns and the continental effect in Northern Hemisphere stations are comprehensive and well documented in comparison to the Southern Hemisphere.

This study presents an extensive tritium data set, extending back to 1962, collected from a network of stations throughout Australia. The aim is to construct a more detailed picture of fallout distribution and identify the influence of thermonuclear weapons testing in the Southern Hemisphere. The tritium composition in monthly rainfall will also be used to examine temporal and spatial variability across the continent and ascertain how these phenomena have changed as the tropospheric tritium levels have gradually decreased; thereby providing tritium reference data for hydrology studies in Australia.

2. Methods

2.1. Sampling stations

Since 1960 monthly precipitation samples have been collected for ^{3}H analysis from various meteorological stations throughout Australia under the International Atomic Energy Agency (IAEA)/World Meteorological Organisation (WMO) program now known as the Global Network of Isotopes in Precipitation (GNIP), and independently by the Australian Nuclear Science and Technology Organisation (ANSTO), previously known as the Australian Atomic Energy Commission (AAEC). This paper presents the tritium dataset collected by the AAEC/ANSTO since 1970 (Calf et al., 1976a, 1977; Calf and Stokes, 1979, 1981, 1983, 1985, 1987). Data prior to this time was obtained from the GNIP database (IAEA/WMO, 2006). The locations of the sampling stations which have operated throughout Australia are shown in Fig. 1.

Site locations and the time-span of monthly sample collections at each meteorological station are listed in Table 1. The tabulated data is restricted to the collection stations with a relatively complete record.

In Australia, the most extensive datasets with the longest continuous sampling period(s), spanning 1962 until present, are for two meteorological stations: Brisbane and Darwin. An additional four stations, Adelaide, Alice Springs, Melbourne and Perth, also commenced operation in the early 1960s under the IAEA/WMO program. In 1970 the AAEC independently established a network of 16 meteorological sampling stations, duplicating the existing six IAEA/WMO sites and adding an additional 10 locations in NSW, Queensland and Tasmania. In 1979, Cape Grim (Tasmania), which is Australia's baseline station, was added to the IAEA/WMO network. Sampling was discontinued in 1983 at Bundaberg, Charleville, Claredale, Hobart, Ryde and Toowoomba, and the AAEC took over responsibility for the IAEA/WMO network in Australia; duplicate sampling ceased at this time. At the end of 1991 the network was further downsized to four locations; Brisbane, Cape Grim, Darwin and Melbourne; in 1999 collection in Melbourne ceased. Since 2005 monthly precipitation samples have been collected from the Alice Springs, Brisbane, Cape Grim, Darwin, Melbourne and Perth stations for tritium analysis. In 2006 nine new precipitation sampling stations were established by ANSTO, primarily for $\delta^{2}H$ and $\delta^{18}O$ analyses, at Adelaide, Charleville, Cobar, Kyeemagh, Meekatharra, Mildura, Mount Isa, Wagga Wagga and Woomera. Because analytical capacity for tritium is limited, only a few samples from these sites have been analysed, and the remainder will be archived.

For overlapping collection periods, the r^2 value between the GNIP and ANSTO datasets for each site was greater than or equal to 0.81. However the agreement between the two datasets for Perth was poorer; with a r^2 value of 0.6, which was influenced by six samples (two months in each year) between 1970 and 1972. The six samples either had a large concentration for the ANSTO data and a lower concentration for the GNIP data, or vice versa. As these are historical data, insufficient information precludes a reason for the discrepancy.

2.2. Sampling method

Precipitation was sampled on a calendar month basis at locations outlined in Table 1. Historical records suggest that both the IAEA/WMO and the AAEC programs used similar sampling methods, consisting of a composite sampler comprised of a funnel in a large glass or steel collector. No technique to prevent evaporation of the sample appears to have been used in these samplers up to 2005. In 2005–2006 the precipitation isotope sampling network in Australia was revised with daily rain gauge samples composited manually from a standard rain gauge into a HDPE bottle over each month. This reduces potential evaporative losses compared with the previous technique.

2.3. Analytical technique

Tritium activity in a composite of the total precipitation at a station during each month has been determined at the low level tritium laboratory, at AAEC/ANSTO since 1971. Currently tritium is measured on a 9.00 g ± 0.05 g distilled and electrolytically enriched sample, using a sample to Perkin Elmer Ultimagold uLLT scintillant ratio of 0.69:1 by volume. The samples are counted for between 1000 and 2000 min on a Perkin Elmer 1220 Quantulus scintillation counter, to give a detection limit of 0.13–0.21 TU (2011 samples); NIST reference materials are used to calibrate the counter.

The standard error of measurement for samples analysed prior to 1990 has been reported as ±0.5 TU. As the tritium levels have steadily decreased since then and the mean tritium concentration in precipitation is now ~2 TU in the southern hemisphere there has been a need to achieve a higher level of sensitivity. The combined standard uncertainty (1 σ) in the measurements is typically reported at ~6% of the tritium concentration which corresponds to ~0.15 TU. The improvement in the quantification limit and the uncertainty has been achieved by gradually optimising the methodology and sampling systems described in Calf et al. (1976b). Improvements in counting statistics have been achieved by

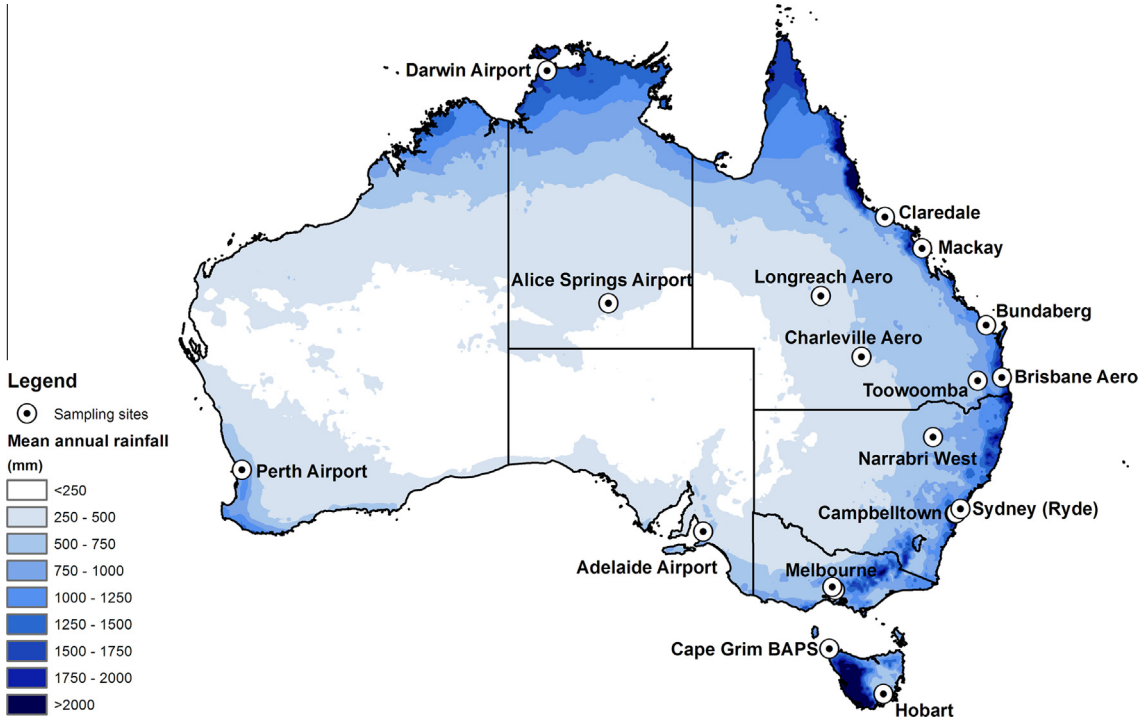

Fig. 1. Map of Australia depicting the network of Australian precipitation sampling stations and mean annual rainfall. The isotopes and data recorded at each site include tritium, deuterium and oxygen-18 content and the amount of precipitation in monthly precipitation samples.

Table 1
Tritium in precipitation sample summary including the meteorological station name, the World Meteorological Organization (WMO)/IAEA station number (IAEA/WMO, 2006), the Bureau of Meteorology (BOM) station number, latitude and longitude given in decimal degrees, altitude of station in metres above mean sea level, number of samples and the period of record for each Australian site.

Station name	WMO/IAEA number	BOM number	Latitude (°N)	Longitude (°E)	Altitude (mAHD)	n[c]	Period of record
Adelaide Airport	946750	023034	−34.9524	138.5204	2	329 (98)	1962–1991, 2005–2007
Alice Springs Airport	943260	015590	−23.7951	133.8890	546	239 (56)	1964–1991, 2005–2012
Brisbane Aero	945760	040223/	−27.4178	153.1142		413 (138)	1962–1982, July 1994–2012
		040842	−27.3917	153.1292	5		
Brisbane RFC	–	040214	−27.48	153.03	38	123 (65)	1983–June 1994
Bundaberg Post Office	–	039015	−24.8667	152.3467	14	141	1970–1982
Campbelltown Swimming Centre	–	068081	−34.0833	150.8167	75	168	1970–1984
Campbelltown (Kentlyn)[a]	947650	068160	−34.0542	150.8772	115	70	1985–1991
Cape Grim BAPS[b]	949540	091245	−40.6828	144.6900	94	338 (78)	1979–2012
Charleville Aero	945100	044021	−26.4139	146.2558	302	133	1970–1982
Claredale	–	033001	−19.5775	147.4075	11	129	1970–1982
Darwin Airport	941200	014015	−12.4239	130.8925	30	362 (149)	1963–2012
Hobart (Ellersie Road)	–	094029	−42.8897	147.3278	51	155	1970–1982
Longreach Aero	943460	036031	−23.4372	144.2769	192	168	1970–1991
Mackay M.O	943670	033119	−21.1172	149.2169	30	226	1970–1991
Melbourne Regional Office	948680	086071	−37.8075	144.9700	31	432 (228)	1960–1999
Melbourne Airport[d]	948660	086282	−37.6655	144.8321	113	68	2005–2012
Narrabri West Post Office	–	053030	−30.3401	149.7552	212	135	1970–1987
Perth Airport	946080	009021	−31.9275	115.9764	15	335 (88)	1963–1991, 2006–2012
Ryde[e] (Concord Golf Club)	–	066013	−33.85	151.10	15	151	1970–1982
Toowoomba	–	041103	−27.5836	151.9317	691	151	1970–1982

[a] Data presented for Campbelltown during 1970–1984 were samples collected from the Campbelltown Swimming Centre station, when the station closed on 31 December 1984 sampling re-commenced on 1January 1985 at Campbelltown (Kentlyn), situated 6.5 km away. Missing rainfall data from this dataset was taken from the nearest BOM station at Minto, Aderney St (068220), 3.7 km away.

[b] Missing rainfall data taken from BOM station 091011, Cape Grim (Woolnorth), 2.5 km away. Samples at this station were collected by ERNI or Eigenbrodt samplers which operate when rainfall intensity exceeds three drops per hour.

[c] Duplicate samples collected by GNIP and AAEC/ANSTO from which r^2 was calculated is in brackets.

[d] In Melbourne sampling resumed in 2005 at an alternate station situated 19.9 km from the initial site.

[e] The exact location of sampling at Ryde is unknown. The closest continuous rainfall record to the Ryde locality is Concord Golf Club, 4.3 km south. Rainfall amount and location data are for this station.

increasing the electrolytic concentration from ~29 to ~52-fold. Chemical recovery has also been improved through the use of programmable power supplies to more closely regulate the current density applied during electrolysis, and improved quality control in sample handling. The inclusion of three background samples (rather than one) in each LSC protocol, as well as counting all

samples/standards/backgrounds for up to 2000 min each, has greatly improved the statistics used to calculate the quantification limit and uncertainty. Additionally, a reduced background count rate has been achieved through the exclusive use of Quantulus scintillation counters with both passive (lead) & active (guard PMT) shielding of cosmic rays.

2.4. Treatment of data

Data presented for the mean annual tritium concentration has been weighted by the amount of precipitation for a given station and calculated using the method outlined in IAEA (1992). As per Taylor and Fletcher (1988) the mean annual (July–June) tritium values are calculated to ensure the annual seasonal tritium peak (spring–summer) is captured in a single average and not split between two years. Tritium concentrations are expressed in tritium units as used in the GNIP database, where 1 TU equals 0.11919 ± 0.00021 Bq kg^{-1} of water (Gröning and Rozanski, 2003).

3. Results and discussion

3.1. Sources and trends of tritium in the atmosphere

Australian observations of the average weighted annual values for tritium concentrations in precipitation at studied locations dating back to 1963 are presented in Fig. 2, and are a measure of the total tritium deposited at each station. Examination of the data highlights the changes in tritium levels that occurred in the Southern Hemisphere since 1962 due to atmospheric nuclear weapons testing and the gradual decrease in the levels of tritium in precipitation following their ban. The data clearly records three periods of elevated tritium levels. For the interpretation of the data, the study period was divided into three phases based on the different testing regimes.

3.1.1. Phase 1

Prior to above ground thermonuclear testing between 1954 and 1962, the mean background tritium level in Australian precipitation due to cosmogenic origin was estimated to be 3.1 ± 1.0 TU (Allison and Hughes, 1977). This value was obtained from vintage wine samples prior to and including 1954, from unirrigated vineyards in the Barossa Valley or McLaren Vale districts of South Australia. The standard deviation, however, should be used with caution as the sample size was not specified. Since 1962 all Australian precipitation contained elevated tritium levels (Fig. 2a). Increased levels of anthropogenic tritium were injected into the stratosphere from the mid-1950s, but particularly during 1961–1962, which is classified as one of the most intense periods of above ground thermonuclear weapons testing. In total 541 atmospheric tests were conducted resulting in a total explosive yield of 440 Mt, of which 182 Mt was fission yield. Of the total fission yield, 158 Mt was radioactive debris which was carried into the troposphere and stratosphere, and over time dispersed globally. Up to 1600 MCi (59.2×10^{18} Bq) of tritium was released into the atmosphere before 1963 (Klement, 1964). During this time most atmospheric testing occurred in the Northern Hemisphere with a total fission yield of 140 Mt (TNT equivalent) of energy released mostly into the stratosphere compared to a fission yield of 18 Mt released into the Southern Hemisphere; taking into account testing that was conducted at or near the equator injecting debris into both hemispheres. From the limited atmospheric testing which occurred in the Southern Hemisphere (Fig. 3), 12

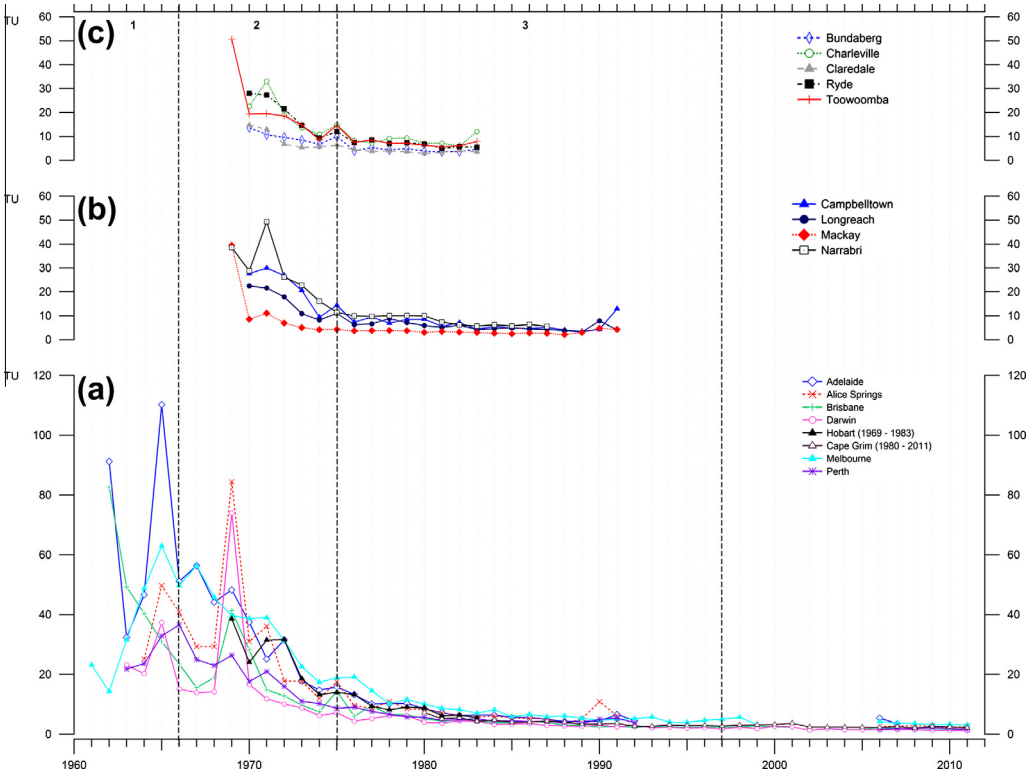

Fig. 2. Time series of weighted mean annual (July–June) tritium concentration in precipitation at different stations throughout Australia. The date and data point are plotted on 1 Jan of each year. Stations are grouped based on the period of record (a) 50 year record, (b) 21 year record and (c) 12 year record. The numbers within the dotted lines represent the different phases of nuclear weapons testing. Phase 1 is the period of atmospheric weapon tests mainly in the Northern Hemisphere; Phase 2 (1966–1974) represents the period of French atmospheric testing on the Mururoa and Fangataufa atolls in French Polynesia; and Phase 3 (1975–1996) represents the period of underground testing in French Polynesia.

detonations were performed in Australian territory and involved devices in the kiloton range (kt) from the United Kingdom tests between 1952 and 1957. These tests were conducted off the Monte Bello Islands in Western Australia, Emu Field and Maralinga Range in South Australia (Table 2a). Testing with higher yields was performed by the UK at Malden Island and Christmas Island in the Pacific. At the USA test site on Christmas Island located in the Northern Hemisphere, very near to the equator (Table 2b), 24 airdrop tests over 78 days were conducted in 1962 with a total yield of 23.3 Mt. Due to the predominantly northern location of atmospheric testing, increased levels in the Southern Hemisphere were 10 to 100 times smaller compared to the Northern Hemisphere (Clark and Fritz, 1997). Of the six stations monitored, maximum tritium levels ranged between 36.6 and 110.2 TU, the highest yearly mean levels in Australia were observed in Adelaide and reached 110.2 TU in 1965, peaking after the moratorium in 1963. This first observed tritium concentration peak in meteoric water, 'tritium pulse', we attribute to be collectively due to the 1962 high yield USA testing on Christmas Island and the global dispersal of the 1962 atmospheric high yield tests in the Northern Hemisphere. This injected debris into the stratosphere which was transported to the Southern Hemisphere through slow inter-hemisphere exchange, taking up to 2 years (Begemann and Libby, 1957; Schell et al., 1970).

Regionally, other measurements of Southern Hemisphere precipitation also showed reduced bomb-peak levels. Kaitoke, New Zealand (GNIP station 934170, 41.1°S, 175.17°E), an island country, represents the most complete GNIP tritium record in the Southern Hemisphere. Kaitoke has a more muted response to the nuclear testing than many Australian sites with the highest recorded annual weighted mean (July–June) tritium content in precipitation of only 38.5 TU between 1964 and 1965, two years after the testing peak of 1962. This reduced pulse is partly due to the location of atmospheric nuclear testing but is mainly explained by the bulk of precipitation at the coastal station being diluted considerably by the ocean and not subject to a strong continental influence as is the case for Australia.

3.1.2. Phase 2

After the moratorium, France continued atmospheric testing until 1974 (Table 2c). Between 1966 and 1974, the French army conducted 41 atmospheric nuclear tests at the Fangautaufa and Mururoa atolls in French Polynesia with a total yield of 3.74 Mt and 6.43 Mt at each site respectively. Of the 41 tests the greatest total explosive yield was in 1968 when a total of 4595 kt was released from July to September (Fig. 4). It included a 2600 kt thermonuclear balloon which was detonated on the 24/08/1968, 520 m above Fangataufa lagoon and was the largest single test conducted over the eight year period. The top of the cloud head reached an altitude of 24 km injecting debris into the stratosphere; a hiatus in 1969 followed. A consequence of higher yield tests is the introduction of a greater proportion of debris into the stratosphere; which takes up to two years for the dispersion and deposition of the material (Eriksson, 1965). This large release of radioactive material into the atmosphere is reflected in the Australian values and corresponds well with the second period of elevated tritium levels between 1966 and 1974 (Fig. 2). Tritium levels reached peak values in 1969 in the range of 39.4 to 84.4 TU. The yearly mean tritium levels in 1969 at Darwin and Alice Springs were 74.0 and 84.4 TU, respectively; these levels are double those of the preceding year. Elevated levels at the Brisbane (41.4 TU), Mackay (39.4 TU) and Toowoomba (50.6 TU) monitoring stations were also observed in that year.

To examine the possible impacted areas from the atmospheric tests, forward trajectories were calculated from the Fangataufa test site (22°14′S, 138°45′W) for each hour of the day on 24/08/1968, at a number of starting heights. These forward trajectories were generated using the PC version of HYSPLIT v4.0 (HYbrid Single-Particle Lagrangian Integrated Trajectory; Draxler and Rolph, 2003). The meteorological data used from the calculations were the gbl files available at ftp://arlftp.arlhq.noaa.gov/archives/reanalysis, which are of 2.5 × 2.5° resolution.

The length of time for the trajectories to pass over Australia varied with the starting altitude of the forward trajectory at Fangataufa. Fig. 5 shows 22 day forward trajectories starting at 2 km above ground level at Fangataufa. After 22 days the air masses had passed over Australia and were at altitudes from 0.5 km to 10 km above ground level. Higher levels of tritium in precipitation were also recorded at Alice Springs and Adelaide for September 1968. It took 36 days to reach Australia when a starting height of 5 km above ground level at Fangataufa was used, whereas for a starting height of 24 km above ground level the air masses were still at 24 km above ground level after 6 months of simulation. However it is well known that trajectory accuracy decreases with the distance travelled (Stohl, 1998) and these simulations can only serve as an indication. It is interesting to note that air masses ini-

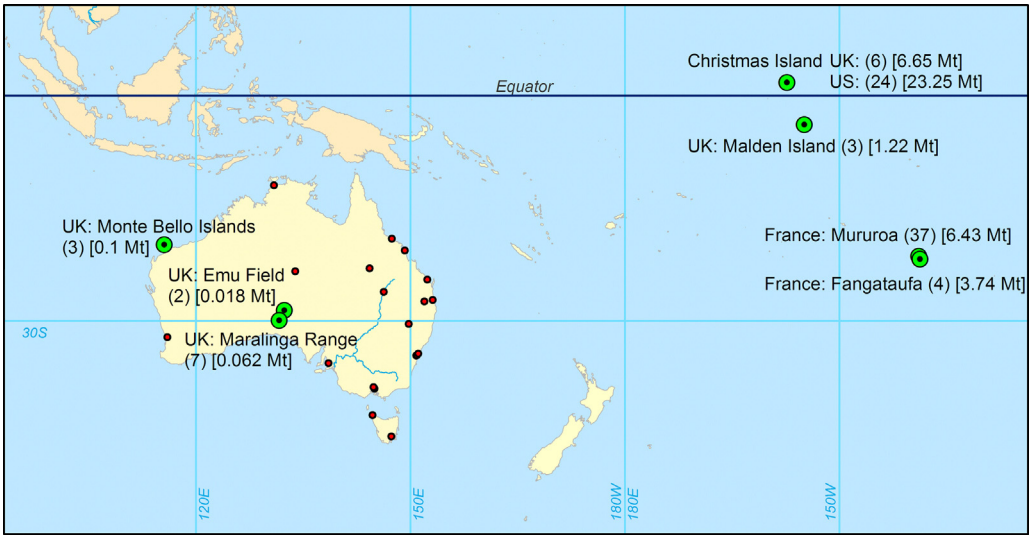

Fig. 3. Map of Australiasia showing the atmospheric nuclear test site locations in the Southern Hemisphere and test sites close to the equator. Key: Country conducting tests: Site name (number of tests) [Yield (Mt)]. The precipitation sampling sites are shown as small dots.

Table 2
Summary of the atmospheric nuclear tests conducted in the Southern Hemisphere by (a) the United Kingdom, (b) the United States of America and (c) France. Date and yield of individual tests, the total number of tests and total explosive yields measured in units of mega tonnes (Mt) of trinitrotoluene (TNT) equivalent are provided. (Table has been adapted from Warner and Kirchmann, 2000.)

Test sites	Date: yield (Mt)		Number of tests	Total yield (Mt)
(a)				
Australia				
Emu Field, South Australia	14/10/53: 0.01		2	0.018
	26/10/53: 0.008			
Monte Bello Islands, Western Australia	03/10/52: 0.025		3	0.100
	16/05/56: 0.015			
	19/06/56: 0.06			
Maralinga Range, South Australia	27/09/56: 0.015	14/09/57: 0.001	7	0.062
	04/10/56: 0.0015	25/09/57: 0.006		
	11/10/56: 0.003	09/10/57: 0.025		
	22/10/56: 0.01			
			Subtotal	0.18
Pacific Ocean				
Christmas Island	08/11/57: 1.8	11/09/58: 0.8	6	6.649
	28/04/58: 3	23/09/58: 0.025		
	22/08/58: 0.024			
	02/09/58: 1			
Malden Island	15/05/57: 0.3		3	1.22
	31/05/57: 0.72			
	19/06/57:0.2			
			Subtotal	7.87
(b)				
Pacific Ocean				
Christmas Island	25/04/62: 0.19	08/06/62: 0.782	24	23.25
	27/04/62: 0.41	09/06/62: 0.21		
	02/05/62: 1.09	10/06/62: 3		
	04/05/62: 0.67	12/06/62: 1.2		
	08/05/62:0.1	15/06/62: 0.8		
	09/05/62: 0.1	17/06/62: 0.052		
	11/05/62: 0.05	19/06/62: 2.2		
	12/05/62: 0.5	22/06/62: 0.0815		
	14/05/62: 0.097	27/06/62: 7.65		
	19/05/62: 0.073	30/06/62: 1.27		
	25/05/62: 2.6	10/07/62: 1		
	27/05/62: 0.043	11/07/62: 3.88		
			Subtotal	23.25
(c)				
French Polynesia				
Mururoa	02/07/66: 0.028	04/07/71: 0.009	37	6.433
	18/07/66: 0.05	08/08/71: 0.004		
	11/09/66: 0.11	14/08/71: 0.955		
	04/10/66:0.205	25/06/72: 0.5 kt		
	05/06/67: 0.014	30/06/72: 0.004		
	27/06/67: 0.12	27/07/72: 0.006		
	02/07/67: 0.022	21/07/73: 0.011		
	07/07/68: 0.115	28/07/73: 0.05 kt		
	15/07/68: 0.45	18/08/73: 0.004		
	03/08/68: 0.15	24/08/73: 0.2 kt		
	08/09/68: 1.28	28/08/73: 0.006		
	15/05/70: 0.013	16/06/74: 0.004		
	22/05/70: 0.224	07/07/74: 0.15		
	24/06/70: 0.012	17/07/74: 0.004		
	03/07/70: 0.914	25/07/74: 0.008		
	27/07/70: 0.05 kt	15/08/74: 0.096		
	06/08/70: 0.594	24/08/74:0.014		
	05/06/71: 0.034	14/09/74: 0.332		
	12/06/71: 0.44			
Fangataufa	24/09/66: 0.125	30/05/70: 0.945	4	3.742
	24/08/68: 2.6	03/08/70: 0.072		
			Subtotal	10.17

tially travelled in a westerly direction and then spun around to an easterly direction. Air masses starting at higher altitudes travelled further west than depicted in Fig. 5 before continuing east.

Similarly monitoring of New Zealand, Fiji, Tonga, Samoa and Tahiti showed that the total beta activity in air and rain was elevated for the whole period from 1966 to 1975 (Atkinson et al., 1984). Although fission products from the tests were expected to circle the globe in an eastward direction, intense monitoring of the Pacific region indicated that radioactive material had also been swept west to the central South Pacific suggesting a strong linkage between the concentration of fission products and French atmospheric tests from 1966 to 1974 (Atkinson et al., 1984).

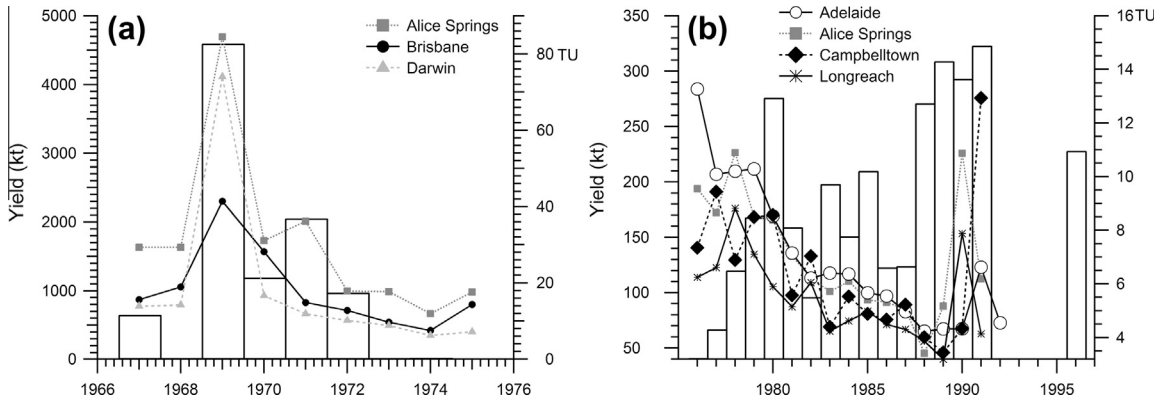

Fig. 4. July–June annual yields(bars) from (a) the French atmospheric nuclear tests conducted between 1966–1974 at Mururoa and Fangataufa Atolls (the annual explosive yield was particularly high in 1968 due to a short period of intense testing) and (b) from underground testing (data obtained from Warner and Kirchmann, 2000). Overlayed is the precipitation weighted mean annual tritium (July–June) from selected stations.

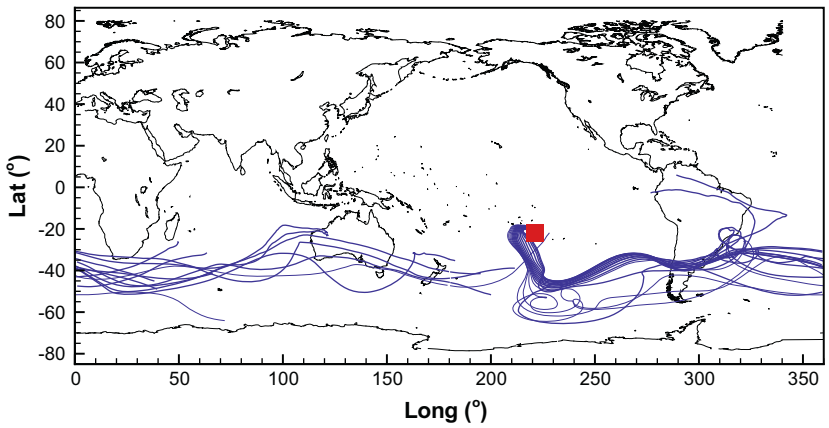

Fig. 5. Hourly forward trajectories produced by the HYSPLIT model. Trajectories start 2 km above the ground from the Fangataufa test site on 24/08/1968.

3.1.3. Phase 3

The third period of elevated tritium levels were recorded in Australian precipitation between 1975 and 1996, coinciding with the era of underground testing. Between 1990 and 1991 the mean tritium concentration in precipitation at four sampling stations peaked between 6.6 and 12.9 TU, which is two to six times the expected background level (Fig. 2). In 1990 tritium concentrations increased from 5.2 to 10.9 TU at Alice Springs and 3.2 to 7.9 TU at Longreach. Likewise in 1991 tritium levels were elevated from 4.3 to 12.9 TU at Campbelltown and 4.3 to 6.6 TU in Adelaide (Fig. 4b). This indicates a local source of technogenic tritium. One third of the total energy released from all underground tests occurred between 1988 and 1990 (Fig. 4) and it is believed that the dominant contributor to tritium in the atmosphere was from the 137 underground tests that were conducted at the Mururoa and Fangataufa Atolls. If underground tests are well contained there should theoretically be no release, however all underground tests resulted in the release of tritium to the atmosphere (Atkinson et al., 1984; Schoengold et al., 1996; Happell et al., 2004). Although there is no GNIP tritium data available for the French Polynesian stations to corroborate this, in 1983 Atkinson et al. (1984) measured the levels of tritium in the interstitial air of the surface terrain to be 2500 times higher than expected from atmospheric fallout due to venting from underground cavities. Pitman et al. (1995) also identified visible damage to the structural integrity of the atoll which led to the release of volatile radioisotope material to the biosphere and surrounding oceans.

Since underground testing largely ceased in the early 1990s, tritium levels in precipitation have declined with a fluctuating quasi-exponential decay rate, and are approaching pre-bomb levels. Due to the rapid uptake of tritiated water into oceans and groundwater, this decay rate is faster than that of natural radioactive decay rate. It is not clear if the decline in tritium in Australian precipitation has ceased, however it appears to be stabilising at levels in the 2–3 TU range.

3.2. Seasonal trends

The tritium concentrations in monthly precipitation samples during the period of nuclear test activity exhibit pronounced seasonal variation. Monthly tritium values for Brisbane (Fig. 6) are generally representative of the pattern observed at all Australian stations. Within the annual cycle, a clear maximum is observed in early spring between August and September and extends into summer, with the minimum concentration occurring in March/April. The monthly long term trend over the Australian continent (Fig. 7a) exhibits the same seasonal influence. This is explained by the stratosphere-to-troposphere exchange which is the main mechanism that transports tritium enriched water vapour to the troposphere annually during the spring, temporarily elevating the tritium levels, followed by an autumn minimum (Rozanski et al., 1991; Alley, 1993; Yasunari and Yamazaki, 2009). In the Southern Hemisphere this enhanced mass flux is located around 35°S, in the region near the subtropical jet, where tropopause folds occur preferentially (Koch et al., 2006; Skerlak et al., 2013). The springtime tritium peak extending into January is due to a maximum evapotranspiration flux in summer (Gat et al., 2001). During the period of thermonuclear testing the annual range was large. As

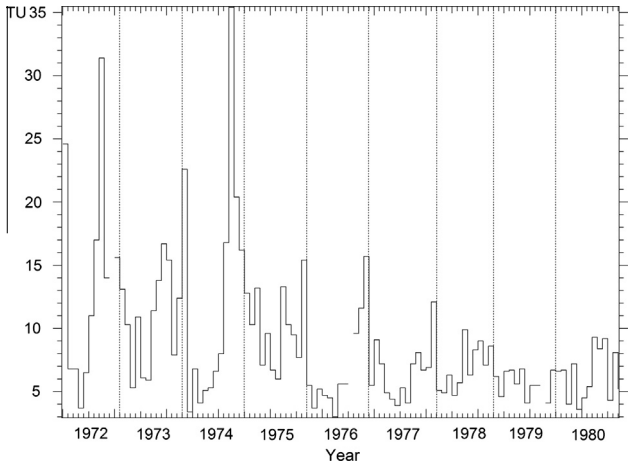

Fig. 6. Time series of monthly tritium concentration in precipitation observed in Brisbane between 1972 and 1980, highlighting the seasonal cycle which becomes less pronounced as tritium levels gradually decline.

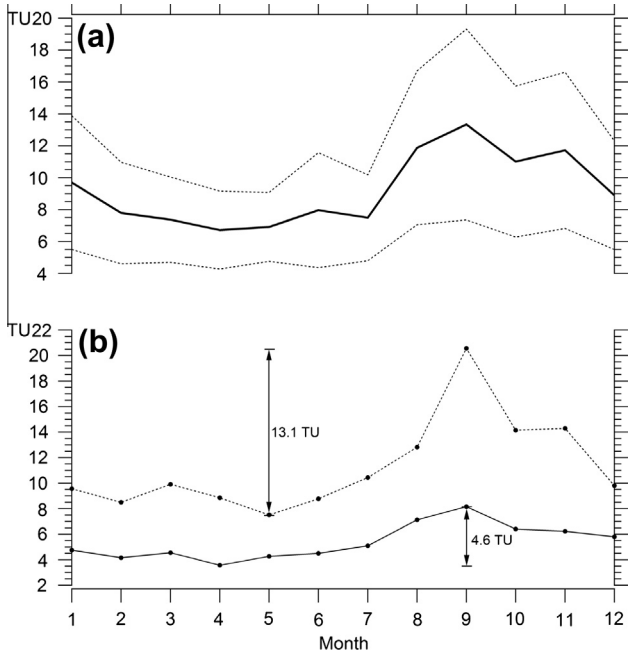

Fig. 7. (a) Annual tritium cycle in precipitation; seasonal pattern derived based on monthly data averaged over the entire period of collection from all stations. Solid line is the mean concentration; bounding lines represent the standard deviation. (b) Tritium concentrations of monthly precipitation samples at Alice Springs (dotted line) and Brisbane (solid line) exhibit variation with the expected seasonal influence. However the spring peak is clear in Alice Springs and the difference between the maximum to minimum mean monthly tritium concentration in precipitation is almost three times that at the Brisbane station which shows a smoother trend. This is explained by atmospheric air being diluted by marine moisture which is depleted in tritium and continental enrichment as moisture moves inland.

tritium levels return to pre-1952 levels, the annual cycle with a spring maximum is still observed but is less pronounced; essentially natural concentrations in autumn–winter now range from 1 to 3 TU with spring values being up to twice these.

3.3. Ocean–continental trend

The proximity of a sampling station to the coast has an effect on the tritium concentration. This is known as a continental effect, where the mean yearly tritium concentrations are higher in the interior of the continent compared to coastal sites. At coastal sites the lower atmosphere is directly influenced by the ocean. Water vapour over the ocean is depleted in tritium due to the transfer of HTO by vapour exchange into the ocean. Therefore precipitation at coastal sampling sites is lower in tritium, as a large fraction of the precipitation is diluted by molecular exchange of tritium between the rain droplet and ocean water vapour. This molecular exchange mechanism, as described by Bolin (1958), is generally less inland as the fraction of ocean water vapour within the atmospheric water decreases, resulting in an increasing tritium concentration in precipitation from the coast landward. However this is dependent on meteorological factors including the moisture source region, i.e. inland or coastal, and prevailing weather systems (Crawford et al., 2013).

Seasonal variations of tritium in precipitation at coastal sites are less pronounced compared to inland stations, as a large fraction of the precipitation is from water vapour of oceanic origin, having low tritium content. This correlation is discernible when two sites at similar latitudes are compared. It can be seen in Fig. 7b that the difference between the minimum and maximum tritium concentrations at Alice Springs (7.5–20.6 TU) is greater than that observed at Brisbane (3.6–8.2 TU). Also the peak concentration at Alice Springs is 2.5 times that at Brisbane, confirming a lower tritium content at coastal sites, as well as a reduced seasonal variation pattern, evidenced by a flatter amplitude.

Tritium enrichment at continental sites is also due to Bolin's molecular exchange mechanism. However instead of rain droplets exchanging with depleted marine water vapour, they are enriched by re-evaporated continental water which has a higher tritium concentration than the cloud water. In addition to a reduced supply of oceanic vapour, re-evaporated water from the continent is enriched in tritium due to continued seasonal stratospheric input as well as evapotranspiration, which returns tritium enriched water vapour to the lower atmosphere. This inland gradient is greatest during spring when the tritium transfer between the stratosphere and troposphere is a maximum and extends to summer when re-evaporation of high HTO from the continents is greatest.

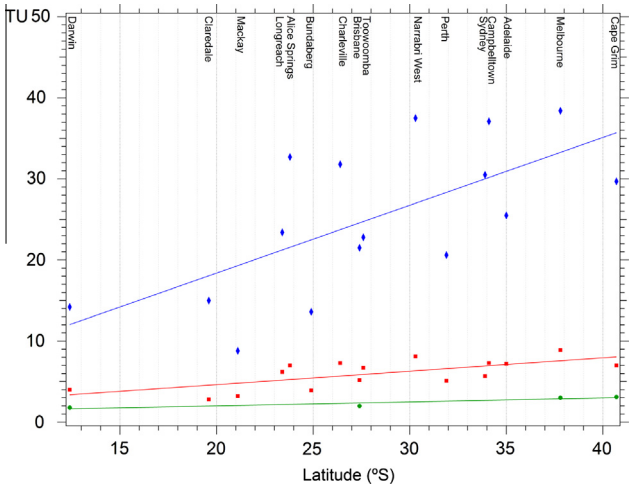

Fig. 8. Correlation between annual mean tritium concentration and latitude for: 1970 (closed diamond); 1980 (closed square); and 1999 (closed circle) precipitation. The solid lines represent the slope for each dataset. The linear regression equation and square of the correlation coefficient is: $y = 0.84x + 1.6$, $r^2 = 0.4$ (1970); $y = 0.17x + 1.3$, $r^2 = 0.5$ (1980); and $y = 0.05x + 1.0$, $r^2 = 0.9$ (1999). An increasing latitudinal effect is also evident with higher tritium values trending south of the continent.

In Australia, this continental effect is prevalent when inland and coastal stations at the same latitude are compared (e.g. Campbelltown: Sydney, Toowoomba: Brisbane and Alice Springs: Longreach) with inland stations having higher mean values (Fig. 8) particularly during the thermonuclear era.

3.4. Latitudinal trend

The deposition of tritium through precipitation at all stations was also compared latitudinally across the Australian continent (Fig. 8). Typically, the concentration of tritium in precipitation

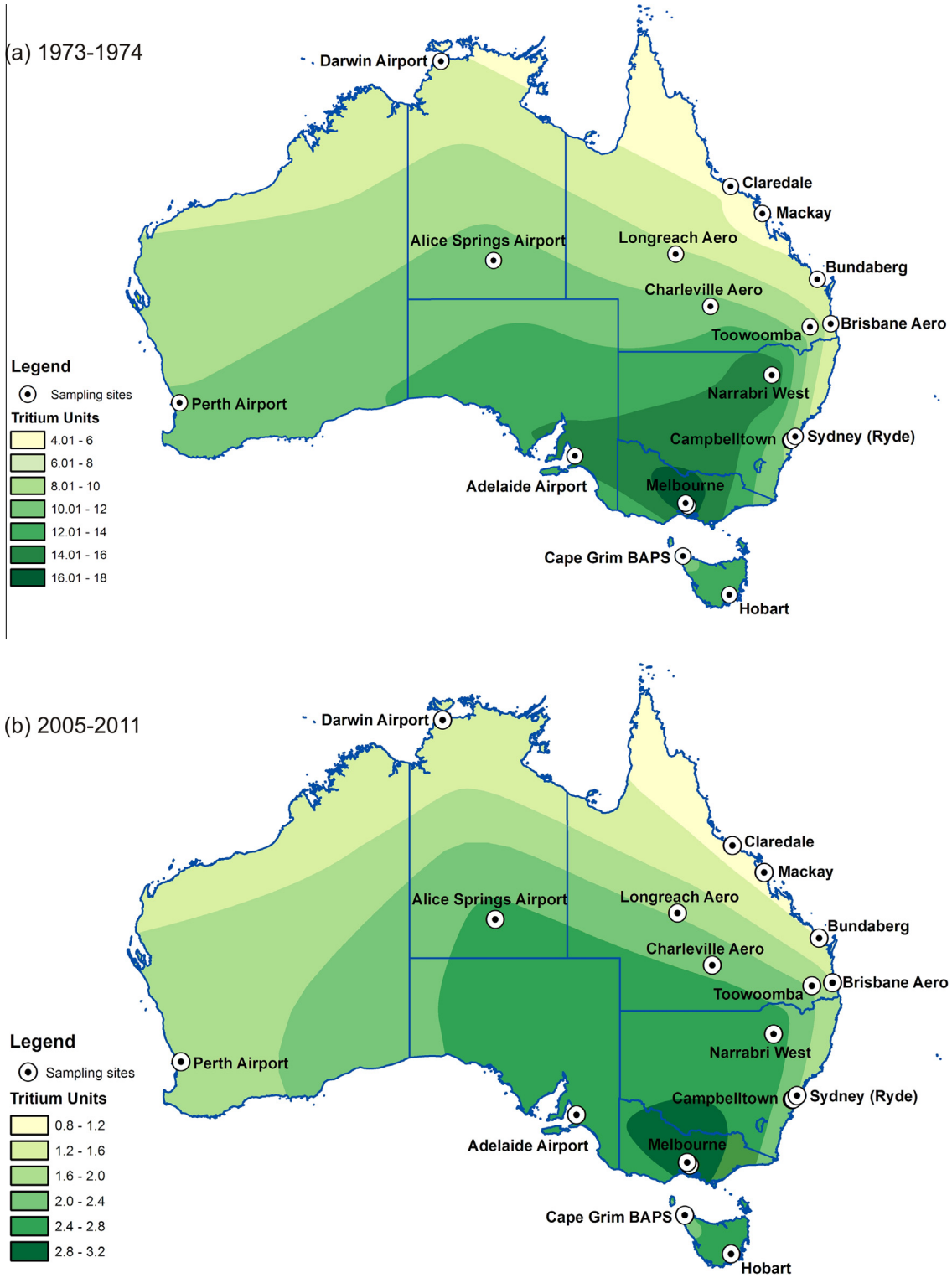

Fig. 9. (a) Rainfall weighted tritium in Australian precipitation for the period July 1973–June 1974, incorporating the Queensland floods of January 1974 and (b) rainfall weighted tritium in Australian precipitation for the period July 2005–June 2011. This map provides an indication of the average input tritium for modern groundwater recharge.

increases with latitude, with high values in the polar regions (Rozanski et al., 1991). Higher maximum mean annual tritium values are observed in South-East Australia both during the nuclear testing period (Fig. 9a) and the present (Fig. 9b). This latitudinal tritium gradient over Australia; which shows a southward-increasing trend, is a direct consequence of the seasonal stratosphere-to-troposphere HTO transfer (Section 3.2) which occurs in the region of subtropical jet streams at approximately 35–40°S (Koch et al., 2006; Skerlak et al., 2013) and is additionally confirmed by seasonal fluctuations of tritium concentrations in precipitation as shown in Fig. 7. Our data also reveal that the slope of the tritium concentration as a function of latitude has decreased with the gradual cessation of anthropogenic tritium being injected into the stratosphere.

3.5. Altitude effect

Higher concentrations of tritium in precipitation as altitude increases (altitude effect) may be due to two mechanisms. Firstly, the upper troposphere is enriched in tritium moisture; at higher altitudes clouds penetrate the higher layers of the troposphere incorporating enriched tritium moisture. Secondly, the tritium concentration in the precipitation at lower tropospheric layers may decrease due to isotopic exchange between liquid water and atmospheric moisture depleted in tritium in the low tropospheric layers (Jouzel et al., 1977).

Taylor (1966) recognised that an ideal locality to study the altitude effect on tritium concentrations in precipitation would consist of an inland mountainous belt which receives oceanic rainwaters, such as the Southern Alps of New Zealand. However, elevations in Australia are relatively low, with an average elevation of only 330 m; hence a clear altitude effect would not be expected. Despite this, the possibility of an altitude effect was examined. The weighted annual mean tritium concentrations were compared with altitude for three periods; 1970, 1980 and 2008 and whilst there is a slight positive correlation ($r = 0.25$, $r = 0.35$ and $r = 0.36$ respectively) it can be explained by the low tritium values at the lower latitude coastal sites, as the majority of Australian stations are located on the coast. Continental and latitude effects outweigh any possible altitude effect in this data.

3.6. Precipitation amount effect

Fractionation of hydrogen isotopes during condensation and evaporation processes has been studied in detail for deuterium (Craig and Gordon, 1965); however tritium fractionates even more strongly than deuterium due to its greater mass difference ($\alpha = 0.9$ and $\alpha_k = 1.1$, Herczeg and Imboden, 1988). This fractionation may result in an inverse correlation between tritium in rainfall and precipitation amount, also referred to as an 'amount effect', as HTO will condense preferentially, due to its lower saturation vapour pressure than H_2O, (Craig and Lal, 1961) and become concentrated in early rainout and diluted in more complete rainout. However, unlike deuterium, tritium has anthropogenic and cosmogenic atmospheric sources that may affect this relationship.

Table 3
Correlation between normalised ^{3}H (monthly ^{3}H/12 month ^{3}H) and monthly precipitation amount. Correlations significant at the 99%ile confidence interval are italicized.

Normalised ^{3}H	Pearsons r	n	t	Sig	Slope	Slope error
Adelaide	−0.03	326	0.62	0.54	−0.00049	0.00079
Alice Springs	−0.28	235	4.43	*0.00*	−0.00228	0.00051
Brisbane	−0.18	526	4.24	*0.00*	−0.00088	0.00021
Bundaberg	−0.15	141	1.78	0.08	−0.00049	0.00028
Campbelltown	−0.20	239	3.21	*0.00*	−0.00123	0.00038
Cape Grim	−0.04	336	0.66	0.51	−0.00027	0.00041
Charleville	−0.31	128	3.62	*0.00*	−0.00273	0.00075
Claredale	−0.29	129	3.40	*0.00*	−0.00077	0.00023
Darwin	−0.03	358	0.64	0.52	−0.00005	0.00008
Hobart	−0.01	166	0.07	0.95	−0.00007	0.00105
Longreach	−0.35	168	4.85	*0.00*	−0.00299	0.00062
Mackay	−0.21	234	3.35	*0.00*	−0.00046	0.00014
Melbourne	0.04	480	0.90	0.37	0.00027	0.00070
Narrabri	−0.23	144	2.77	*0.01*	−0.00147	0.00055
Perth	−0.23	332	4.38	*0.00*	−0.00133	0.00030
Ryde	−0.40	151	5.27	*0.00*	−0.00225	0.00043
Toowoomba	−0.12	161	1.52	0.13	−0.00072	0.00047

A precipitation amount effect is observed in monthly tritium data collected at some Australian sites. To determine the extent of the amount effect at each site we normalised each month's tritium value to the 12 month average and determined the correlation with monthly rainfall (Table 3). A medium inverse correlation (significant at a 99% confidence interval) was found for 10 out of the 17 locations. Fig. 10 gives typical examples of the distribution of normalised ^{3}H with respect to rainfall for a correlated (Alice Springs) and a non-correlated (Cape Grim) dataset. The slope, or amount effect, was greatest for the most inland sites: Alice Springs, Charleville and Longreach. In contrast, Adelaide, Melbourne, Cape Grim and Hobart, along the southern coastline, and Darwin, in the north, showed no correlation and Toowoomba and Bundaberg in Eastern Australia did not exhibit a significant correlation. For deuterium this effect is stronger in lower latitudes; this is also seen to some extent for the Australian tritium data.

We have considered whether this amount effect is actually due to the timing of the stratosphere–troposphere exchange with

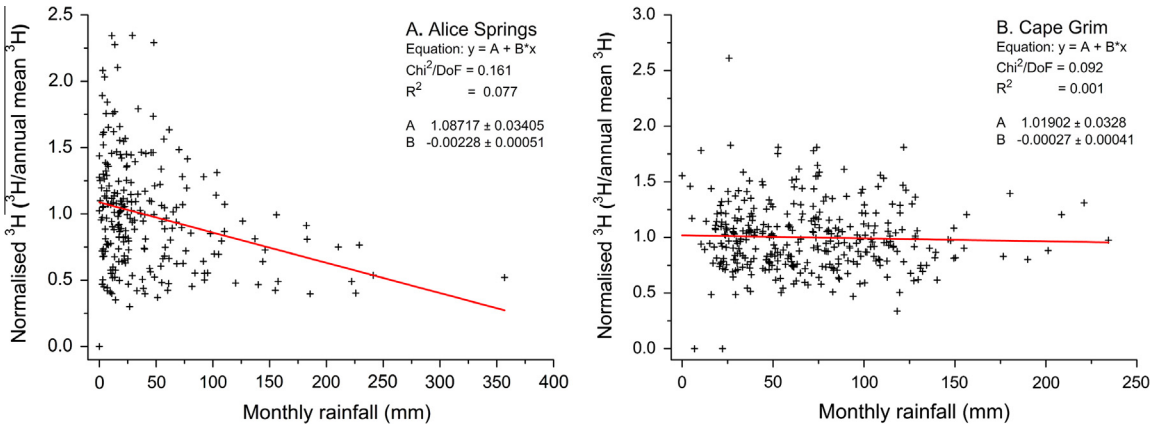

Fig. 10. Normalised ^{3}H vs rainfall and linear regressions lines for (A) Alice Springs and (B) Cape Grim.

respect to the seasonal rainfall. Adelaide, Hobart, Cape Grim and Perth have winter maximum rainfall. In these cases the spring–summer tritium peak would coincide with reduced rainfall and an inverse correlation would be expected. This is only observed for Perth. In the case of Melbourne where there is little seasonality in rainfall with a slight spring maximum a positive correlation may be expected. This is not observed. In the lower latitudes where the stratosphere–troposphere exchange has less influence the coastal sites all have strong summer maximum rainfall and the inland sites have a weaker summer maximum. This would mean that mid-range rainfall amounts would coincide with the spring leak resulting in little if any correlation. However, these are the areas in which the inverse correlation is seen. The major exception is Darwin where the rainfall is monsoonal and the dry season months commonly have little or no rain at all.

3.7. Modern tritium input to Australian groundwater

The usefulness of tritium in the age dating of groundwater depends on the availability of a precipitation input function. In many parts of Australia groundwater recharge is episodic and may only occur during major flooding events. During the period of record reported in this study a major flood event occurred in January 1974 in SE Queensland in association with cyclone 'Wanda' which potentially contributed to groundwater recharge over a widespread area of Queensland and northern NSW. Five of the eight sites in the region experienced the maximum monthly rainfall for the 50 year sampling period and with very high rainfall seen in the remaining three, as well as Alice Springs and Darwin. The high rainfall amount resulted in low tritium concentrations in Queensland for the month ranging from 3.4 TU in Brisbane to 5.7 TU in Charleville, with a continental effect still being observed. The rainfall weighted average tritium from July 1973 to June 1974 is mapped in Fig. 9a providing an indication of the spatial variation in tritium concentration.

The pre-bomb pulse tritium level of 3.1 ± 1.0 TU for the Adelaide region (Allison and Hughes, 1977); Section 3.1.1) has been used as a background level for groundwater dating throughout Australia. This dataset shows that Adelaide and Melbourne have higher rainfall tritium levels than the rest of Australia indicating that the true tritium background would have been lower at most sites. Fig. 9b illustrates the likely variability of tritium in modern precipitation using data from July 2005 to June 2011. In order to interpolate, weighted average tritium for the period 1977–1982 from sites no longer in operation was scaled down to present values based on the strong linear relationship between the two periods for the 5 sites still in operation ($r^2 = 0.987$).

When comparing variations to the distribution of tritium in Australian precipitation over thirty years (Fig. 9) it is clearly evident that the patterns of spatial distribution remain essentially unchanged; with higher maximum mean annual tritium values persisting in South-East Australia. This distribution is a direct consequence of the stratosphere-to-troposphere exchange mechanism which occurs around 35–40°S in the Southern Hemisphere (Sections 3.2 and 3.4).

4. Conclusions

The five decade dataset has provided an observational foundation for understanding the spatial and temporal deposition of anthropogenic tritium in precipitation across the Australian continent; mainly seasonally from stratosphere-to-troposphere exchange and limited local testing in the Southern Hemisphere. On the temporal scale we related three periods of elevated levels to different eras of atmospheric and underground testing. We clearly showed that stratosphere-to-troposphere exchange is the dominant driver for the timing of the seasonal peak in Australia and for the spatial gradient of tritium levels in precipitation across the continent. A continental effect was observed and was shown to be influenced by seasonality and prevailing weather systems. A correlation with precipitation amount was also observed for the inland sites. The first spatial distribution map for variations of natural tritium levels across the Australian continent is presented in Fig. 9b and combined with the temporal variation (Fig. 2) has obvious implications as a critical input for accurate groundwater dating studies in Australia. Further improvements in the spatial resolution of the tritium distribution would benefit groundwater studies and could be achieved by collecting and analysing annual composite samples at a wider range of locations.

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References

Akata, N., Kakiuchi, H., Shima, N., Iyogi, T., Momoshima, N., Hisamatsu, S., 2011. Tritium concentrations in the atmospheric environment at Rokkasho, Japan before the final testing of the spent nuclear fuel reprocessing plant. J. Environ. Radioact. 102, 837–842.

Alley, W.M., 1993. Regional Ground Water Quality. Van Nostrand Reinhold, New York, NY, 635 p.

Allison, G.B., Hughes, M.W., 1975. The use of environmental tritium to estimate recharge to a South-Australian aquifer. J. Hydrol. 26, 245–254.

Allison, G.B., Hughes, M.W., 1977. The history of tritium fallout in southern Australia as inferred from rainfall and wine samples. Earth Planet. Sci. Lett. 36, 334–340.

Atkinson, H.R., et al., 1984. Report of a New Zealand, Australian, and Papua New Guinea scientific mission to Moruroa Atoll, October–November 1983. New Zealand Ministry of Foreign Affairs, Wellington.

Bainbridge, A.E., 1963. Tritium in surface waters of the north pacific. In: Nuclear Geophysics. National Academy of Sciences, National Research Council, Washington DC, pp. 129–137.

Begemann, F., Libby, W.F., 1957. Continental water balance, ground water inventory and storage times, surface ocean mixing rates and worldwide water circulation patterns from cosmic ray and bomb tritium. Geochim. Cosmochim. Acta 12 (4), 277–296.

Bolin, B., 1958. On the use of tritium as a tracer for water in nature. In: Proceedings of the Second United Nations Conference on the Peaceful Uses of Atomic Energy, Geneva, Switzerland, pp. 336–343.

Calf, G.E., Stokes, R.C., 1979. AAEC Tritium List No. 3, 1977–1978. AAEC/E473.

Calf, G.E., Stokes, R.C., 1981. AEC Tritium List No. 4, 1979–1980. AAEC/E519.

Calf, G.E., Stokes, R.C., 1983. AAEC Tritium List No. 5, 1981–1982. AAEC/E569.

Calf, G.E., Stokes, R.C., 1985. AAEC Tritium List No. 6, 1983–1984. AAEC/E610.

Calf, G.E., Stokes, R.C., 1987. AAEC Tritium List No. 1, 1985–1986. AAEC/E661.

Calf, G.E., Seatonberry, B.W., Smith, L.W., 1976a. The Measurement of Natural Levels of Tritium in Water. AAEC/E373.

Calf, G.E., Seatonberry, B.W., Smith, L.W., Stokes, R.C., 1976b. AAEC Tritium List No. 1, 1970–1974. AAEC/E377.

Calf, G.E., Smith, L.W., Stokes, R.C., 1977. AAEC Tritium List No. 2, 1975–1976. AAEC/E414.

Cartwright, I., Morgenstern, U., 2012. Constraining groundwater recharge and the rate of geochemical processes using tritium and major ion geochemistry: ovens catchment, southeast Australia. J. Hydrol. 475, 137–149.

Clark, I.D., Fritz, P., 1997. Environmental Isotopes in Hydrogeology. Lewis, New York, USA, 328 p.

CNSC, 2009. Investigation of the Environmental Fate of Tritium in the Atmosphere – Part of the Tritium Studies Project-, INFO0792. Canadian Nuclear Safety Commission, Canada.

Craig, H., Gordon, L.I., 1965. Deuterium and oxygen 18 variations in the ocean and the marine atmosphere. In: Tongiorgi, E. (Ed.), Stable isotopes in oceanographic studies and paleotemperatures. Lab di Geologia Nucleare, Pisa, pp. 9–130.

Craig, H., Lal, D., 1961. The production rate of natural tritium. Tellus 13 (1), 85–105.

Crawford, J., Hughes, C.E., Parkes, S.D., 2013. Is the isotopic composition of event based precipitation driven by moisture source or synoptic scale weather in the Sydney Basin, Australia? J. Hydrol. 507, 213–226.
Draxler, R.R., Rolph, G.D., 2003. Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT), model. <http://www.arl.noaa.gov/ready/hysplit4.html>.
Ehhalt, D.H., Rohrer, F., Schauffler, S., Pollock, W., 2002. Tritiated water vapor in the stratosphere: vertical profiles and residence time. J. Geophys. Res. 107 (D24), 4757. http://dx.doi.org/10.1029/2001JD001343.
Eriksson, E., 1965. An account of the major pulses of tritium and their effects in the atmosphere. Tellus 17, 118–130.
Gat, J.R., Mook, W.G., Meijer H.A.J., 2001. Atmospheric Water. In: Mook, W.G. (Ed). Environmental Isotopes in the Hydrological Cycle: Principles and Applications. IHP-V Technical Documents in Hydrology No. 39, vol. 2. UNESCO, Paris.
Gröning, M., Rozanski, K., 2003. Uncertainty assessment of environmental tritium measurements in water. Accred. Qual. Assur. 8, 359–366.
Happell, J.D., Östlund, H.G., Mason, A.S., 2004. A history of atmospheric tritium gas (HT) 1950–2002. Tellus 56B, 183–193.
Harries, J.R., Calf, G.E., 1980. Tritium measurements in the Tasman Sea and the Southern Ocean. Aust. J. Mar. Freshwater Res. 31, 737–745.
Hauglustaine, D.A., Ehhalt, D.H., 2002. A three-dimensional model of molecular hydrogen in the troposphere. J. Geophys. Res. 107, 4330. http://dx.doi.org/10.1029/2001JD001156.
Herczeg, A.L., Imboden, D.M., 1988. Tritium hydrologic studies in four closed-basin lakes in the Great Basin, USA. Limnol. Oceanogr. 33 (2), 157–173.
Huang, T.M., Pang, Z.H., 2010. Changes in groundwater induced by water diversion in the Lower Tarim river, Xinjiang Uygur, NW China: evidence from environmental isotopes and water chemistry. J. Hydrol. 387, 188–201.
IAEA, 1992. Statistical treatment of data on environmental isotopes in precipitation. Technical Report Series 331, IAEA Vienna, 781 pp.
IAEA/WMO, 2006. Global Network of Isotopes in Precipitation. The GNIP Database. <http://www.iaea.org/water>.
Jouzel, J., Merlivat, L., Pourchet, M., 1977. Deuterium, tritium, and activity in a snow core taken on the summit of Mont Blanc (French Alps). Determination of the accumulation rate. J. Glaciol. 18 (80), 465–470.
Kakiuchi, H., Momoshima, N., Okai, T., Maeda, Y., 1999. Tritium concentration in ocean. J. Radioanal. Nucl. Chem. 239, 523–526.
Klement, A.W., Jr., 1964. Radioactive fallout from Nuclear weapon tests. AECS Symposium Series. Division of Technical Information Extension, Oak Ridge, Tennessee, USA. Proceedings of the Second Conference, Germantown, Maryland.
Koch, P., Weirnli, H., Davies, H.C., 2006. An event-based jet-stream climatology and typology. Int. J. Climatol. 26, 283–301.
Lucas, L.L., Unterweger, M.P., 2000. Comprehensive review and critical evaluation of the half-life of tritium. J. Res. Natl. Inst. Stand. Technol. 105 (4), 541–549.
Martell, E.A., 1963. On the inventory of artificial tritium and its occurrence in atmospheric methane. J. Geophys. Res. 68, 3759–3769.
Momoshima, N., Okai, T., Kaji, T., Takashima, Y., 1991. Distribution and transformation of various chemical forms of tritium in the environment. Radiochim. Acta 54, 129–132.
Morgenstern, U., Daughney, C.J., 2012. Groundwater age for identification of impacts of land-use intensification and natural hydrochemical evolution on groundwater quality – the National Groundwater Monitoring Programme of New Zealand. J. Hydrol. 456–457, 79–93.
Morgenstern, U., Stewart, M.K., Stenger, R., 2010. Dating of streamwater using tritium in a post nuclear bomb pulse world: continuous variation of mean transit time with streamflow. Hydrol. Earth Syst. Sci. 14, 2289–2301.
Morishima, H., Kawai, H., Koga, T., Niwa, T., 1985. The trends of global tritium precipitations. J. Rad. Res. 26, 283–312.
Orban, P., Brouyere, S., Batlle-Aguilar, J., Couturier, J., Goderniaux, P., Leroy, M., Maloszewski, P., Dassargues, A., 2010. Regional transport modelling for nitrate trend assessment and forecasting in a chalk aquifer. J. Contam. Hydrol. 118, 79–93.
Pitman, M., collaborators, 1995. The impact of nuclear testing at Mururoa and Fangataufa. In: A paper proposed for the South Pacific Environment Ministers Meeting, August. Brisbane.
Rozanski, K., Gonfiantini, R., Araguas-Araguas, L., 1991. Tritium in the global atmosphere: distribution patterns and recent trends. J. Phys. G: Nucl. Part. Phys. 17, S523–S536.
Scanlon, B.R., Healy, R.W., Cook, P.G., 2002. Choosing appropriate techniques for quantifying groundwater recharge. Hydrogeol. J. 10, 18–39.
Schell, W.R., Sauzay, G., Payne, B.R., 1970. Tritium injection and concentration distribution in the atmosphere. J. Geophys. Res. 75 (12), 2251–2266. http://dx.doi.org/10.1029/JC075i012p02251.
Schoengold, C.R., DeMarre, M.E., Kirckwood, E.M., 1996. Radiological effluents release from U.S. continental tests 1961 through 1992, United States Department of Energy Report NV-317 (Rev. 1).
Skerlak, B., Sprenger, M., Wernli, H., 2013. A global climatology of stratosphere–troposphere exchange using the ERA-interim dataset from 1979 to 2011. Atmos. Chem. Phys. Discuss. 13, 11537–11595.
Stohl, A., 1998. Computation, accuracy and applications of trajectories—a review and bibliography. Atmos. Environ. 32 (6), 947–966.
Taylor, C.B., 1966. Tritium in Southern Hemisphere precipitation 1953–1964. Tellus 18 (1), 105–131.
Taylor, C.B., Fletcher, E.H., 1988. Assessment of data for tritium in Southern Hemisphere precipitation, 1960–1983. Institute of Nuclear Sciences, DSIR, New Zealand, Report, No.INS-R-379, 20p.
Vreca, P., Bronic, I.K., Horvatincic, N., Baresic, J., 2006. Isotopic characteristics of precipitation in Slovenia and Croatia: comparison of continental and maritime stations. J. Hydrol. 330, 457–469.
Warner, F., Kirchmann, R.J.C., 2000. Nuclear test explosions: environmental and human impacts Scientific Committee on Problems on the Environment (SCOPE). John Wiley and Sons Ltd, West Sussex, England, p. 275.
Yangui, H., Abidi, I., Zouari, K., Rozanski, K., 2012. Deciphering groundwater flow between the Complex Terminal and Plio-Quaternary aquifers in Chott Gharsa plain (southwestern Tunisia) using isotopic and chemical tools. Hydrol. Sci. J. 57 (5), 967–984.
Yasunari, T.J., Yamazaki, K., 2009. Impacts of Asian dust storm associated with the stratosphere-to-troposphere transport in the spring of 2001 and 2001 on dust and tritium variations in Mount Wrangell ice core, Alaska. Atmos. Environ. 43, 2582–2590.
Zuber, A., 1986. Mathematical models for the interpretation of environmental radioisotopes in groundwater systems. In: Fritz, P., Fontes, J.-C.h. (Eds.), Handbook of Environmental Isotope Geochemistry. Elsevier, Amsterdam, pp. 1–59.
Zuber, A., Maloszewski, P., 2001. Lumped-parameter models. In: Mook, W.G. (Ed.), Environmental Isotopes in the Hydrological Cycle: Principles and applications, vol. 6: Modelling. Tech. Doc. Hydrol. 39, UNESCO/IAEA, Vienna, pp 5–35.
Zuber, A., Kania, J., Rozanski, K., Purtschert, R., 2011. On some methodological problems in the use of environmental tracers to estimate hydrogeologic parameters and to calibrate flow and transport models. Hydrogeol. J. 19, 53–69.