

Isotopic studies of megacryometeors in western India

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Four unusually large ice-stones (megacryometeors) weighing several kilograms fell in western India during October–November 2010. Samples were retrieved from three fall events. To study the origin and formation of these megacryometeors, $\delta^{18}\text{O}$, δD , chemical composition and γ -activity were measured. The range of $\delta^{18}\text{O}$ – δD is similar to the local meteoric water, suggesting origin from local atmospheric water vapour. The relative humidity at ~4 km altitude was higher during a few days before and after the fall. No γ -activity from cosmogenic radionuclides such as ^{26}Al could be detected in the inherent dust. These studies indicate terrestrial origin of these megacryometeors.

Keywords: Atmospheric moisture, cosmogenic radionuclides, ice stones, megacryometeors, stable isotopes.

LARGE icy conglomerates, occasionally falling from a clear sky even when there are no clouds or precipitation, have recently been termed as megacryometeors¹. They are easily distinguished from hailstones by their unusually massive (~1 kg to few hundred kilograms) size. In the past, such ice stones have also been referred to as ice-meteors² and hydro-meteors³. Although their formation is not clearly understood, they are considered to have an origin different from large hailstones. Megacryometeors may share textural, structural and compositional similarities with hailstones that are usually formed due to updraft of warm air and downdraft of cold air inside the cumulonimbus clouds during thunderstorm in which supercooled droplets crystallize, grow in size and eventually fall. In contrast, megacryometeors form suddenly even during non-cloudy, clear sky when there is no thunderstorm activity^{1,4}. The mechanism that generates hails and hailstorms is well understood in the context of weather scenario, meteorological parameters, heat and radiation budget, etc.^{5–7}. However, the formation mechanism of megacryometeors is poorly understood due to unpredictability of location and time of fall and logistic difficulties of spontaneous sample collection and proper preser-

vation. Therefore, only samples of opportunity have been studied so far^{8–11}.

Several causal mechanisms hypothesized for the formation and fall of megacryometeors are: (i) aircraft icing; (ii) waste water released from aircraft lavatories; (iii) leakage from aircraft water tanks; (iv) condensation trails (contrails) of jet planes and (v) extraterrestrial (e.g. cometary) origin. It has, however, not been possible to identify and associate any specific factor for a particular fall.

Many meteorites, asteroids and comets contain large amounts of water and there is some evidence, though highly controversial, that a lot of small icy comets fall on the earth^{12–15}. Keeping this possibility in mind and to ascertain their origin, it is necessary to study the megacryometeors by various techniques.

A list of various events of ice-stone falls in different parts of the world, including megacryometeors, has been compiled by Martinez-Frias *et al.*⁹, and Martinez-Frias and Huertas¹⁶. It is alarming that megacryometeor fall frequency has increased since 1950 and that 46 fall events have been recorded between 2001 and 2006 alone¹⁶. However, it is not clear if the increase in fall frequency signifies any systematic change in the atmospheric component of the hydrological cycle or better public awareness. Another noteworthy point is the higher frequency of larger and massive ice-stones. The earliest recorded fall at Córdoba, Spain weighed ~2 kg and subsequently the heaviest fall reported till 2005 was ~18 kg. However, recent reports show ice-stones weighing ~50 kg at Campinas, Brazil, ~200 kg at Itapira, Brazil in 1997 and ~400 kg at Toledo, Spain in 2004 (ref. 16).

Results of isotopic and chemical studies of ice and melt water samples from three of the four megacryometeors that fell in quick succession in western India during October–November 2010 are reported in this article.

Fall locations: geography and weather

The four megacryometeor fall events were: (i) Haripura village, Jhunjhunu district, Rajasthan on 1 October 2010; (ii) Titoli village, Ratnagiri district, Maharashtra on

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7 October 2010; (iii) Borana village, Surendranagar district, Gujarat on 8 October 2010 and (iv) Sultana Ka Bas village, Jhunjhunu district on 11 November 2010. Locations of these events are shown in Figure 1 and a summary of important weather features around these locations is given in Table 1.

The Jhunjhunu district in which two events occurred is located in the extreme northeastern part of the Thar desert in western India¹⁷. The arid climate of this region is characterized by very hot summers and very cold winters with poor rainfall during southwest monsoon period (June–September). It is a drought-prone district with mean annual rainfall of 486 mm, based on 1971–2006 data. The long-term average of rainfall in October (11 mm) and November (5 mm) in Jhunjhunu district is negligibly small. No rainfall was reported for this district on both days of the fall events.

The Surendranagar district, located in the central-western part of Gujarat, is also arid. It is a drought-prone area with average annual rainfall of 490 mm. The long-term monthly average rainfall for the month of October is 10.8 mm at the nearest meteorological observation station, Ahmedabad (<http://www.imd.gov.in/section/climate/ahmedabad2.htm>). No rainfall was reported for this district on the day of the fall event.

The Ratnagiri district is located in Maharashtra on the west coast of India. The climate of this region is warm and humid with long-term average annual rainfall ranging from 7446 mm in the hilly areas to 2155 mm in the coastal region. The long-term average monthly rainfall in

October ranges from 80 to 234 mm. On the day of the fall event a small amount (~2.1 mm) of rainfall has been reported for this district.

It is noteworthy that Jhunjhunu and Surendranagar districts are located below international aviation corridor through which numerous international flights operate everyday.

Since all the four megacryometeor fell during post-monsoon period when no hailstorm activity was recorded, the fall of any large icy-conglomeration during this period is an extremely unusual phenomenon. Out of the four megacryometeor fall events, samples of ice or melt water could be retrieved only from three events as described below.

Sampling procedure

The samples studied were retrieved and preserved by some alert villagers soon after the fall events. The samples were subsequently collected by the Public Health Engineering Department (PHED), Jhunjhunu, Government of Rajasthan or by the Physical Research Laboratory (PRL), Ahmedabad within a couple of days of each fall event.

The megacryometeor sample of the fall event on 1 October 2010 in Haripura village was originally collected in the form of ice and was preserved in an airtight container. As there was no refrigeration facility available in the village, the ice melted during storage and the melt water was retrieved by the PHED laboratory. The exact dimension or weight of the megacryometeor could not be ascertained. However, a mini-crater (depth 17.5 cm × width 20 cm × breadth 30 cm) created by the impact was identifiable by conspicuous absence of grass, destroyed due to impact (Figure 2). From the dimension of the mini-crater, and as described by onlookers, the megacryometeor could have been ~30 kg in weight.

Unfortunately, the sample of the second event of megacryometeor fall on 7 October 2010 in Titoli village could not be retrieved due to logistic difficulties. This sample could possibly have given information regarding ice stones from humid area in the coastal part of Maharashtra, since other samples collected in this study came from arid areas of Rajasthan and Gujarat.

The samples of ice and melt water of the third fall event of 8 October 2010 in Borana village were retrieved from the occupant of the house on which this megacryometeor fell, breaking the tiled roof. The ice pieces were preserved in two airtight plastic containers in a domestic refrigerator, but due to power failure only one piece of ice (with some melt water) survived in one container, whereas all the ice melted in the other and only melt water could be obtained. From the size of the broken tiles (approximately 110 cm × 65 cm) due to impact of megacryometeor (Figure 3a), it is estimated that the ice

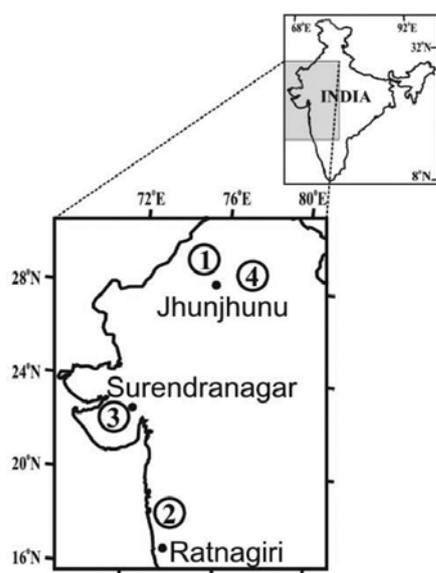


Figure 1. Location of the four megacryometeor fall events in western India – (1) Haripura village, Jhunjhunu district, Rajasthan on 1 October 2010; (2) Titoli village, Ratnagiri district, Maharashtra on 7 October 2010; (3) Borana village, Surendranagar district, Gujarat on 8 October 2010 and (4) Sultana Ka Bas village, Jhunjhunu district on 11 November 2010.

Table 1. Geographical and weather information about megacryometeor fall locations*

District/state/geographical area	Jhunjhunu, Rajasthan, northwestern India	Surendranagar, Gujarat, western India	Ratnagiri, Maharashtra, coastal western India	
Date of fall (dd/mm/yyyy)	1/10/2010 and 11/11/2010	8/10/2010	7/10/2010	
Latitude (°N)	28.1	22.47	17.13	
Longitude (°E)	75.33	71.47	73.31	
Long-term average ground-level weather parameters				
Principal rainy months	June to September	June to September	June to September	
Average annual rainfall (mm)	485.6	490	7446 (Hilly areas) 2155 (Coastal)	
Average monthly rainfall (mm) in the month of fall	11 in October 5 in November	<10.8	234 (Hilly areas) 80 (Coastal areas)	
Mean number of rainy days in the month of fall	2 (October) 2 (November)	2	11	
Mean number of days with hail in the month of fall	0	0	0	
Mean daily minimum temperature (°C) in the month of fall	17	20.9	23.6	
Mean relative humidity (RH) (%) in the month of fall	44.3 (October) 42 (November)	33.5	68	
Ground-level weather parameters on the day of fall (3 hourly intervals over 24 h)				
	Jhunjhunu 1/10/2010	Ratnagiri 7/10/2010	Surendranagar 8/10/2010	Jhunjhunu 11/11/2010
Total rainfall (mm)	0	2.1	0	0
Average temperature (°C)	31.8	27	31.6	22.8
Average RH (%)	43	83	32	45
Average cloud coverage (%)	0.8	10	2.8	19
Average pressure (mb)	1008.4	1006.1	1004.5	1012
Average wind (miles/h)	3.8	4.8	7.3	4.8

*The information in this table is compiled from various sources listed below.

1. <http://www.worldweatheronline.com/weather-averages/India/> (accessed on 26 July 2011).
2. <http://www.imd.gov.in/section/climate/ahmedabad2.htm> (accessed on 26 July 2011).
3. <http://www.imd.gov.in/section/hydro/distrainfall/webbrain/rajasthan/jhunjhunu.txt> (accessed on 26 July 2011).
4. <http://www.esrl.noaa.gov/psd/> (accessed on 2 September 2011).
5. CGWB¹⁷.

stone could have weighed ~20 kg. Another large ice piece was reported to have fallen in an adjoining farm, but no sample or evidence could be found.

The ice sample from the fourth event of 11 November 2010 in Sultana Ka Bas village was timely collected and preserved in refrigerated condition (Figure 3b) by PHED scientists. All these samples were transported to PRL in thermos flask where ice and melt water were separated before storage in a deep freeze at -5°C until their isotopic and chemical analyses could be carried out.

Isotopic and chemical analyses

The isotopic composition of water in its various phases is expressed in δ per mil (‰) units with reference to international standard [$\delta^{18}\text{O}$ or $\delta\text{D} = (R_{\text{sample}}/R_{\text{std}} - 1) \times 1000$]. R denotes the abundance ratio of heavy to light isotope ($R = {}^{18}\text{O}/{}^{16}\text{O}$ or ${}^2\text{H}/\text{H}$) in the sample or standard. The melt water samples obtained from each megacryometeor were analysed for $\delta^{18}\text{O}$ and δD using standard equilibration method^{18,19}. Briefly, liquid water samples were equi-

librated with CO_2 and H_2 . The equilibrated CO_2 and H_2 gases were analysed using an isotope ratio mass spectrometer (IRMS) to measure ${}^{18}\text{O}/{}^{16}\text{O}$ and D/H ratios to compute $\delta^{18}\text{O}$ and δD respectively. The reproducibility of measurements was better than 0.1‰ for $\delta^{18}\text{O}$ and 1‰ for δD . Isotopic analyses were done at PRL using the analytical facility developed under the National Programme on Isotope Fingerprinting of Waters of India (IWIN)²⁰⁻²⁵. A detailed description of the analytical procedures can be found in Maurya *et al.*²⁶.

The chemical analyses (electrical conductivity (EC), total dissolved solids (TDS), pH, chloride) were carried out at PHED laboratory for samples from Haripura and Sultana Ka Bas, and at PRL for samples from Borana.

Examination of ice stones indicated that they were not homogeneous and were made of visually different types of fragments, some well compacted into transparent ice and others fluffy, snow-like material. The ice contained significant amount of inherent dust. The surface of ice stones was washed with double-distilled water to remove any adhering dust from the environment and while

handling. The clean interior melt water samples containing inherent dust were analysed using a gamma ray spectrometer, covering the energy range 0.1 to 2 MeV. In this range many gamma rays, emitted by cosmogenic radio-



Figure 2. A mini-crater (depth 17.5 cm \times width 20 cm \times breadth 30 cm) formed by impact of a megacryometeor which fell at Haripura village in Jhunjhunu district, Rajasthan on 1 October 2010 is identifiable by conspicuous absence of grass destroyed due to impact.

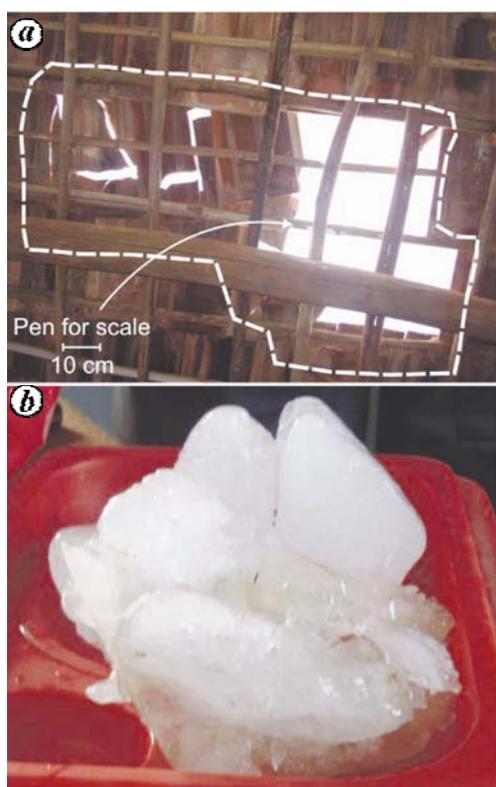


Figure 3. *a*, A portion (approximately 110 cm \times 65 cm) of the tiled roof broken into pieces due to impact of megacryometeor which fell in Borana village, Surendranagar district on 8 October 2010. *b*, Ice sample of the megacryometeor which fell at Sultana Ka Bas village in Jhunjhunu district, Rajasthan on 11 November 2010.

isotopes in meteorites or comets, during their exposure in space, are expected. Prominent among these radioisotopes is ^{26}Al (mean life \sim 1 million years), expected to be >0.1 dpm/g of cosmic dust. The water (487 ml) together with the dust was first counted for a period of about a week (8618 min) in a Marinelli beaker, using high-purity germanium gamma-ray detector, located in a 10 cm thick lead shield²⁷. No signal was detected and therefore, to increase the counting efficiency, aluminum was chemically separated and counted for ^{26}Al activity. For this purpose, after adding known amount of stable aluminum carrier, the dust was dissolved in hydrofluoric acid and digested with nitric acid, perchloric acid and hydrochloric acid. Aluminum hydroxide was precipitated, dried and counted on the hyperpure germanium gamma ray spectrometer.

Results and discussion

The measured values of $\delta^{18}\text{O}$, δD and d -excess ($=\delta\text{D} - 8 \times \delta^{18}\text{O}$) of ice and melt water samples are given in Table 2. The $\delta^{18}\text{O}$ – δD regression of ice and melt water samples defines a line [$\delta\text{D} = (7.7 \pm 0.8) \times \delta^{18}\text{O} + (5.8 \pm 1.7)$; $n = 8$ and $r^2 = 0.94$; Figure 4] which is referred to as local megacryometeor regression line (LMRL). The slope and intercept of LMRL is similar to that of local meteoric water line (LMWL) [$\delta\text{D} = (7.7 \pm 0.2) \times \delta^{18}\text{O} + (7.2 \pm 0.7)$; $n = 120$; $r^2 = 0.96$; Figure 4 (inset)] obtained for precipitation at Ahmedabad (23.03°N; 72.55°E), situated in western India, near one of the fall locations (Surendranagar)²³. The range of isotopic composition ($\delta^{18}\text{O}$: -1.22‰ to -3.53‰ ; δD : -2.8‰ to -21.5‰) of ice and melt water is well within the range of precipitation ($\delta^{18}\text{O}$: 3.7‰ to -13.2‰ and δD : 30‰ to -92‰) at Ahmedabad²³ which is taken as representative of western India. As the slope, intercept and isotopic range of LMRL are similar to representative regional precipitation, it seems that the studied megacryometeors may have been derived from the local tropospheric vapour. It may however be noted that

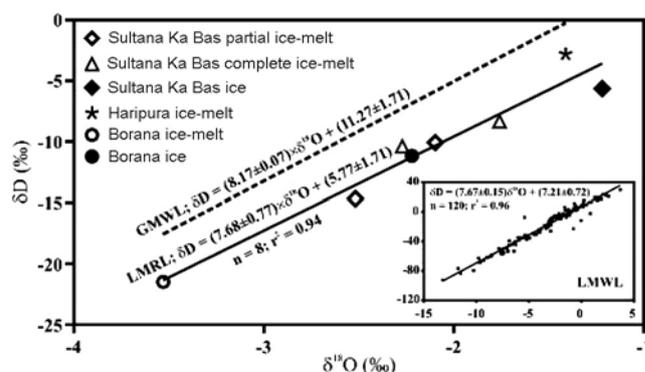
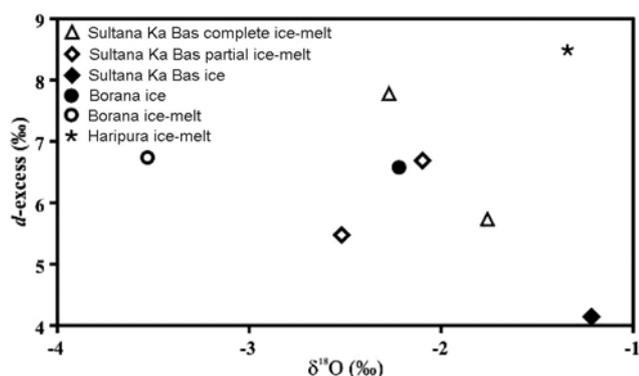


Figure 4. The $\delta^{18}\text{O}$ – δD regression line for ice and melt water samples of the studied megacryometeors local megacryometeor regression line (LMRL) in western India along with the global meteoric water line (GMWL)²⁸ and local meteoric water line (LMWL) for Ahmedabad (inset).

Table 2. $\delta^{18}\text{O}$, δD and d -excess of ice and ice-melt samples of megacryometeors from western India

Location	Sample type	$\delta^{18}\text{O}$ (‰)	δD (‰)	d -excess (‰)
Haripura, Jhunjhunu	Complete ice-melt	-1.41	-2.8	8.5
Sultana Ka Bas, Jhunjhunu	Complete ice-melt-1	-2.27	-10.4	7.8
Sultana Ka Bas	Complete ice-melt-2	-1.76	-8.3	5.7
Sultana Ka Bas	Partial ice-melt-1	-2.52	-14.7	5.5
Sultana Ka Bas	Partial ice-melt-2	-2.10	-10.1	6.7
Sultana Ka Bas	Ice	-1.22	-5.7	4.1
Borana, Surendranagar	Ice	-2.22	-11.2	6.6
Borana	Ice-melt	-3.53	-21.5	6.7
Ahmedabad	Amount weighted average rainfall (2005–08)	-3.9	-23	7.9

**Figure 5.** The d -excess– $\delta^{18}\text{O}$ plot of ice and melt water of various megacryometeors. The melt water from Haripura with comparatively high d -excess and enriched $\delta^{18}\text{O}$ is particularly distinct in the ensemble of samples.

intercepts of both LMRL ($5.8 \pm 1.7\text{‰}$) and LMWL ($7.2 \pm 0.7\text{‰}$) are lower than that for Global Meteoric Water Line (GMWL; $11.27 \pm 0.65\text{‰}$)²⁸. While the lower slope of LMWL is indicative of evaporation from falling raindrops in this region²⁹, the lower slope of LMRL may or may not be indicative of evaporation/sublimation during fall of the studied megacryometeors, as the same could also arise from the vapour mass with low d -excess values.

The d -excess– $\delta^{18}\text{O}$ plot can often provide information such as the origin of vapour from low humidity region³⁰, recycling of vapour³¹, solid condensation under supersaturated environment^{32–34} and secondary evaporation from falling precipitation^{18,35–40}. Some of these aspects may also have relevance to megacryometeors, although such studies have not been made so far. Considerable secondary evaporation from falling precipitation results in decreased d -excess and correspondingly increased $\delta^{18}\text{O}$, and consequent linear trend between the two parameters for an ensemble of samples. This, however, is not observed in case of the megacryometeor samples (Figure 5) in spite of the fact that the d -excess values of the two ice samples are lower (4.1 and 6.6‰; Table 2) than the amount-weighted average d -excess (7.9‰) of rainfall at Ahmedabad. That the observed low d -excess of ice is not due to partial melting during storage (Figure 5) is sup-

ported by the fact that in case of megacryometeor at Borana the d -excess of corresponding melt water is similar to ice; and in case of Sultana Ka Bas only slightly higher than ice (Figure 5). If the low d -excess of ice were due to partial melting, the corresponding melt water should have considerably higher d -excess which is not the case.

The absence of any distinct trend between d -excess and $\delta^{18}\text{O}$ can be ascribed to the possibility that pristine isotopic composition of the megacryometeor (at altitude of formation), in each event, was slightly different. This is also evident from the fact that isotopic composition of melt water of Haripura megacryometeor is distinctly different from other samples in the ensemble, suggesting that the ice from which it melted (Figure 5) must have had considerably different isotopic composition. However, it is also to be noted that the two most enriched ^{18}O values of megacryometeors are observed at Haripura (-1.41‰) and Sultana Ka Bas (-1.22‰), which are located in a warmer and more arid region compared to Borana, where the most depleted ^{18}O value (-3.53‰) is observed. Though difficult to speculate due to limited data availability, it may well be due to different isotopic composition of atmospheric moisture in the two regions around the time of fall of the megacryometeor.

There is only one other comprehensive stable isotope study of megacryometeors which has been reported from Spain⁹. The $\delta^{18}\text{O}$ – δD regressions indicate (Figure 6) subtle differences in isotopic character of megacryometeors from western India and Spain. The major difference between the two sets is that all samples from Spain (average d -excess $\sim 15\text{‰}$) fall above the GMWL, whereas samples from western India (average d -excess $\sim 6.5\text{‰}$) fall below the GMWL. In the case of Spain, both slope and intercept of LMRL are higher than LMWL⁴¹, whereas for western India, both slope and intercept of LMRL and LMWL are comparable. Possible reasons for the observed high d -excess of megacryometeors from Spain could be: (i) kinetic fractionation during their condensation under supersaturated environment³² and/or (ii) significant contribution of land-derived recycled vapour in the megacryometeor mass. The isotopic composition of megacryometeors from western India is largely in agreement

Table 3. D/H ratio and δD in various types of samples from terrestrial and extraterrestrial environments

Sample	D/H ($\times 10^{-6}$)	δD (‰)*	Reference
Solar nebula	20	-870	64, 65
Protosolar nebula hydrogen	26 \pm 10 to 30 \pm 4	-833 \pm 64 to -807 \pm 26	66, 67
Bulk Earth	149 \pm 3	-43 \pm 19	68
Earth's atmosphere	155	0	18, 45
Earth's meteoric water	-	10 to -250	18, 45
Megacryometeors (western India)	155.3 to 152.4	-2.8 to -21.5	Present study
Megacryometeors (Spain)	151.8 to 136	-25.7 to -127.1	9
Comet Halley	316 \pm 34	1028 \pm 218	69-71
Comet Hyakutake	290 \pm 10	861 \pm 64	72
Comet Hele-Bop	330 \pm 80	1118 \pm 513	47
Comet 103P/Hartley2	161 \pm 24	34 \pm 150	48
LL3 Meteorite			
Interstellar water (clays)	730 \pm 120	3686 \pm 77	46, 47
Interstellar water (pyroxene chondrules)	479 \pm 8	2075 \pm 51	
Water out-gassed from carbonaceous meteorites	130 to 280	-165 to 798	73-75
Organic macromolecules extracted from carbonaceous chondrites	320 to 1200	1054 to 6700	75-80
Vienna standard mean ocean water (VSMOW)	155.76 \pm 0.05	0	81

*Some of the cited publications have mentioned hydrogen isotopic composition either only in D/H or δD units. For the sake of convenience, values in both units are presented using the formula: $\delta D = [(D/H)_{\text{sample}}/(D/H)_{\text{VSMOW}} - 1] \times 10^3$. The value of $(D/H)_{\text{VSMOW}} = 155.76 \times 10^{-6}$.

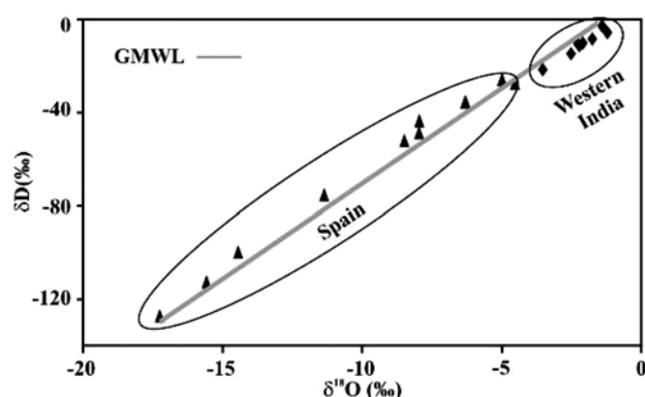


Figure 6. Comparison of $\delta^{18}\text{O}$ and δD values of megacryometeors from western India and from Spain with reference to GMWL²⁸.

with that of the local precipitation, hence these do not suggest any special condensation mechanism or recycled source.

In the past, there has been some speculation about extraterrestrial origin of megacryometeors⁴². There have also been news reports of extraterrestrial life forms found in the ice meteorites (<http://www.24-7pressrelease.com/press-release/ice-meteorite-found-with-extraterrestrial-lifeforms-182667.php>). However, according to Beech⁴³, and Beech and Nikolova⁴⁴ extraterrestrial origin of megacryometeors is highly improbable due to almost impossible survival under sublimation in space and catastrophic destruction during journey through the Earth's atmosphere.

In this context, deuterium content of megacryometeors could also be a diagnostic tool to verify its terrestrial vis-à-vis extraterrestrial origin. Compared to water in the Earth's atmosphere and hydrosphere ($\delta D = \sim +10\text{‰}$ to -250‰)^{18,45}, extraterrestrial objects such as water out-

gassed from carbonaceous meteorites ($\delta D = \sim -165\text{‰}$ to 798‰) and ice in comets ($\delta D = \sim 860\text{‰}$ to 1100‰) have large deuterium enrichment^{46,47}. These measurements refer to comets that had their origin from Oort cloud. However, recently a Jupiter family comet 103P/Heartley2 with origin in the Kuiper belt was reported⁴⁸ to have δD value of 34 ± 150 , which overlaps with the terrestrial source values. A compilation of hydrogen isotopic compositions reported for various types of samples from terrestrial and extraterrestrial environments is given in Table 3. The δD range of megacryometeors, both from western India and Spain, is considerably lower compared to the reported ranges for comets (Halley, Hyakutake and Hele-Bop) or interstellar water (in clays and pyroxene chondrules in LL3 meteorites), except for the comet 103P/Heartley2. Therefore, based on deuterium isotopic composition, the possibility of extraterrestrial origin of the studied megacryometeors cannot be ruled out.

This possibility of extraterrestrial origin of the studied megacryometeors from western India is however ruled out based on the absence of γ -activity due to any of the γ -emitting cosmogenic radionuclides expected in meteorites and interplanetary dust. Of nearly a dozen gamma-emitting radionuclides detected in meteorites, ²⁶Al should be detectable (>0.1 dpm/g) in the amount of dust present (0.45 g) if the inherent dust in any of the three megacryometeors was exposed to cosmic rays in the interplanetary space. Counting rate in the ²⁶Al channels (1808 KeV) for chemically separated and precipitated aluminum was found to be 7.96 ± 3.6 cpd (counts per day), whereas the background counts rate was 7.66 ± 2.5 cpd. This signal was indistinguishable (within statistical error), indicating absence of ²⁶Al activity in the embedded particulate matter in the studied megacryometeors. Thus there is no evidence of exposure of the dust, embedded in the mega-

cryometeor, in the interplanetary space. Microscopic examination indicated that the dust was mostly sand expected from the local desert and did not show presence of any organic threads, filaments or any other indication of life-form in the melt water.

Earlier studies, based on atmospheric soundings from National Ocean and Atmospheric Administration (NOAA) coupled with NCEP/NCAR reanalysis data, indicated that formation of megacryometeors may be associated with certain atmospheric attributes such as undulations of the tropopause, meandering of polar front, ozone anomalies and strong wind turbulence^{10,16,49}. To study if such perturbations could be identified in relation to the fall events from western India, we show in Figure 7 relative humidity and temperature at three pressure levels (1000, 850 and 600 mb) during a few days before and after the fall events. The data are obtained from NCEP reanalysis⁵⁰ from NOAA (<http://www.esrl.noaa.gov/psd/data>). A conspicuous increase in the relative humidity is noticed at all the fall locations, from nearly zero to ~40% (Haripura), >20% (Borana) and ~30% (Sultana Ka Bas) at 600 mb level (~4 km altitude) during a few days before and after the events (Figure 7a–c). Similar variation in relative humidity at 800 and 1000 mb pressure levels does not exist but, in general, an increasing trend is noticeable after the fall events. Although no conspicuous change in temperature is observable at any altitude, the temperature at 600 mb level was 0°C at Borana (Figure 7e) and very close to 0°C at other two locations (Figure 7d and f) on the fall days. These observations suggest that the studied megacryometeors could have been formed at an altitude greater than ~4 km (600 mb level) with subzero temperatures.

From the average vertical profile of atmospheric temperature at Ahmedabad during post-monsoon period^{22,23}, the sub-zero temperature is normally observed above ~5 km. The possibility of megacryometeor formation at altitude much greater than 5 km (hence at much lower temperature than 0°C) can be ruled out in view of the fact that at temperatures much below 0°C, the kinetic effects of vapour deposition under supersaturated condition become effective, resulting in higher *d*-excess in the ice³², which is not observed in case of the megacryometeors studied here (Figure 5). Thus, it seems reasonable to hypothesize that the studied megacryometeors from western India condensed within an altitude range of 4–6 km.

It is noteworthy that, apart from natural perturbations in large-scale synoptic meteorology, aircraft condensation trails (contrails) are associated with increased relative humidity and decreased air temperature on small spatial scales. The aircraft engines emit water vapour, CO₂, NO_x, hydrocarbons, CO, sulphur gases, and soot and metal particles as by-products of high-temperature combustion of jet fuel. Emission of water vapour and resultant increase in relative humidity is responsible for contrail formation with possible contribution of sulphur-bearing gases in

providing condensation nuclei. Persistent contrails often evolve and spread into extensive cirrus cloud cover, which is indistinguishable from naturally occurring clouds. Contrails persist when the ambient atmosphere is ice-supersaturated. Without ice supersaturation, contrail ice crystals evaporate on timescales of seconds to minutes⁴⁹. Persistent contrails are more significant in terms of local climate because they have a more substantial effect on the radiative balance. Persistent liner contrails are estimated to cover, on average, about 0.1% of the Earth's surface⁵¹. The highest percentage of contrail cover occurs in regions with the highest air traffic⁵². In this context, it may be noted that the region of megacryometeor fall is one of the busiest air corridors in India. Thus, aircraft-mediated formation of megacryometeors is possible. However, the possibility that the studied megacryometeors could have fallen from aircraft as frozen waste flushed from toilets is unlikely because no traces of blue disinfectants normally used in airplanes were present in the samples.

The chemical properties of the melt water samples from three fall events are given in Table 4. A comparison of these with the reported values for rainwater in different regions (urban, forested, coastal, arid, etc.) of India^{29,53–57}

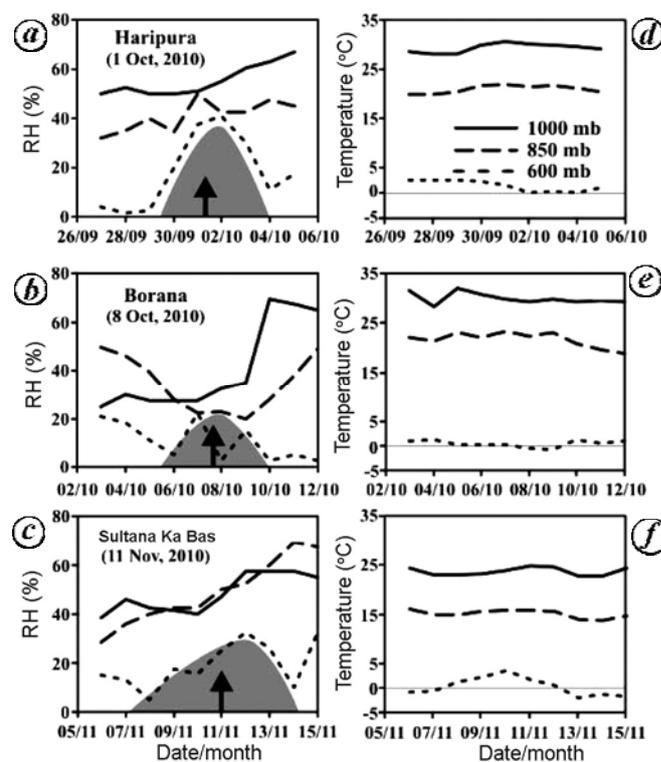


Figure 7 a–f. Temporal variation in relative humidity (RH) and temperature at three pressure levels during a few days before and after the day of the megacryometeor fall event, marked by an arrow. A conspicuous increase in the relative humidity at 600 mb (~4 km) is observed at all locations. The plot is based on NCEP reanalysis data provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA, from their website <http://www.esrl.noaa.gov/psd/>.

Table 4. Chemical properties of melt water samples of three megacryometeors from western India

Parameter	Haripura	Borana	Sultana Ka Bas
pH	6.2	6.5	6.3
TDS (ppm)	120	243	110
EC ($\mu\text{s}/\text{cm}$)	233	486	223
Chloride (mg/l)	30	66	20

reveals that pH of megacryometeor samples (6.2–6.5) is well within the reported pH range (4.8–7.2) for rainwater, but the chloride concentration and EC of megacryometeor samples (chloride: 20–66 mg/l; EC: 223–486 $\mu\text{s cm}^{-1}$) are higher than the reported range (chloride: around 1–8.4 mg/l; EC: around 25–100 $\mu\text{s cm}^{-1}$) for rainwater. These higher values of chloride and EC possibly indicate higher salt content in the atmospheric air in the region of ice formation/accumulation. This may possibly be indicative of the local origin in the arid region above western India. Among the three stations from where megacryometeor samples were collected, Borana has much higher values of EC and chloride concentration compared to the other two stations. This may be possibly due to the proximity of Borana village to a low-lying corridor, linking the Gulf of Cambay and the Gulf of Kutchchh, where surface soil (and also the shallow groundwater) is known to have higher salinity due to evaporative enrichment of salts in the stagnant water, draining from adjoining higher elevation areas. Also, the proximity to the Arabian Sea coast may be another reason for higher salt content in the local atmosphere and any precipitation therefrom.

Our observation that all the sampled megacryometeors have higher EC and chloride concentration than local rainwater may also indicate the role of hygroscopic soluble inorganic atmospheric salts in reducing the degree of supersaturation required for condensation of atmospheric water vapour to occur on inorganic aerosols^{58–60}. In addition to this direct effect, small amounts of inorganic salt can also affect the ability of the organic particles to act as cloud condensation nuclei and resultant condensation of water vapour on organic aerosols^{61–63}.

A simple calculation shows that a small volume ($5-8 \times 10^3 \text{ m}^3$) of atmospheric air above 850 mb, even in this dry region, contains sufficient moisture ($5 \text{ g}/\text{m}^3$ or $\sim 0.5\%$) for the formation of a 25–40 kg megacryometeor. But, if the efficiency of vapour to ice condensation is low, larger air mass is required. However, an aircraft moving through an atmospheric disturbance encounters enough air and may provide enough condensation surfaces on its body and wings, at some sheltered locations, for a megacryometeor to form.

Summary and conclusion

Results of a multi-proxy study of three megacryometeors that fell in quick succession in western India, during

October–November 2010, are reported. Cosmogenic radioactivity, specifically ^{26}Al , characteristic of extraterrestrial matter, was not detected in the ice and melt water samples. The chemical parameters of these samples are within the normal range of variation in rainwater in the region, except for chloride and EC. The oxygen and hydrogen isotopic composition of the ice and melt water samples reveals that the range of observed variation is also well within the range of variation for local precipitation. The slope and intercept of $\delta^{18}\text{O}-\delta\text{D}$ regression line of ice and melt water are also comparable to those of the LMWL. These observations indicate that water molecules in the megacryometeors should be of atmospheric origin in western India. The three fall locations lie below one of the busiest air corridors in western India. This coupled with the observed increase in relative humidity at above 4 km elevation, during a few days before and after the fall events, suggests that perturbations in atmospheric parameters and/or mediation of aircraft may be responsible for their formation in the present cases.

- Martinez-Frias, J. and Travis, D., Megacryometeors: fall of atmospheric ice blocks from ancient to modern times. In *Environmental Catastrophes and Recovery in the Holocene*, Brunel University, West London, UK, 2002.
- Meaden, G. T., The giant ice meteor mystery. *J. Meteorol.*, 1977, **2**, 137–141.
- Corliss, W. R., Ice falls or hydrometeors. In *Tornados, Dark Days, Anomalous Precipitation and Related Weather Phenomena: A Catalog of Geophysical Anomalies*, Glen Arm, MD, USA, 1983, pp. 40–44.
- Brink, K., Travis, D. and Martinez-Frías, J., Upper tropospheric conditions associated with recent clear-sky ice falls. In 57th Annual Meeting, The Wisconsin Geographical Society, UW-Eau Claire, USA, 19–20 September 2003.
- Matrosov, S. Y., Reinking, R. F., Kropfli, R. A. and Bartram, B. W., Estimation of ice hydrometeor types and shapes from radar polarization measurements. *J. Atmos. Ocean.–Technol.*, 1996, **13**, 85–96.
- Matrosov, S. Y., Heymsfield, A. J. and Wang, Z., Dual-frequency radar ratio of nonspherical atmospheric hydrometeors. *Geophys. Res. Lett.*, 2005, **32**, L13816.
- Walko, R. L., Cotton, W. R., Feingold, G. and Stevens, B., Efficient computation of vapor and heat diffusion between hydrometeors in a numerical model. *Atmos. Res.*, 2000, **53**, 171–183.
- Borah, R. R. and Saikia, B. J., Spectroscopy of megacryometeor. In Goldschmidt Conference, Special Edition of *Geochimica et Cosmochimica Acta*, 2010, **74**(11), Suppl. 1, p. A104.
- Martinez-Frias, J. *et al.*, Oxygen and hydrogen isotopic signatures of large atmospheric ice conglomerations. *J. Atmos. Chem.*, 2005, **52**, 185–202.
- Orellana, F. A. *et al.*, Monitoring the fall of large atmospheric ice conglomerations: a multianalytical approach to the study of the Mejrada del Campo megacryometeor. *J. Environ. Monit.*, 2008, **10**, 570–574.
- Rull, F., Delgado, A. and Martínez-Frías, J., Micro-Raman spectroscopic study of extremely large atmospheric ice conglomerations (megacryometeors). *Philos. Trans. R. Soc. London, Ser. A*, 2010, **368**, 3145–3152.
- Frank, L. A., Sigwarth, J. B. and Craven, J. D., On the influx of small comets into the Earth's upper atmosphere I. Observations. *Geophys. Res. Lett.*, 1986, **13**, 303–306.

13. Frank, L. A., Sigwarth, J. B. and Craven, J. D., Reply to comment on the influx of small comets into the Earth's upper atmosphere I. Observations. *Geophys. Res. Lett.*, 1986, **13**, 979–980.
14. Frank, L. A., Sigwarth, J. B. and Craven, J. D., On the influx of small comets into the Earth's upper atmosphere II. Interpretation. *Geophys. Res. Lett.*, 1986, **13**, 307–310.
15. Wasson, J. T. and Kyte, F. T., Comment on the letter on the influx of small comets into the Earth's atmosphere II: Interpretation. *Geophys. Res. Lett.*, 1987, **14**, 779–780.
16. Martinez-Frias, J. and Huertas, A. D., Megacryometeors: distribution on Earth and current research. *Ambio*, 2006, **35**, 314–316.
17. CGWB, Groundwater Brouchure, Jhunjhunu District, Rajasthan, W.R. Central Ground Water Board, Jaipur, Ministry of Water Resources, Government of India, 2008, p. 28.
18. Clark, I. D. and Fritz, P., *Environmental Isotopes in Hydrogeology*, Lewis, Boca Raton, FL, USA, 1997, p. 328.
19. Epstein, S. and Mayeda, T. K., Variations of the $^{18}\text{O}/^{16}\text{O}$ ratio in natural waters. *Geochim. Cosmochim. Acta*, 1953, **4**, 213.
20. Maurya, A. S., Shah, M., Deshpande, R. D., Bhardwaj, R. M., Prasad, A. and Gupta, S. K., Hydrograph separation and precipitation source identification using stable water isotopes and conductivity: River Ganga at Himalayan foothills. *Hydrol. Process.*, 2011, **25**, 1521–1530.
21. Gupta, S. K. and Deshpande, R. D., The need and potential applications of a network for monitoring of isotopes in waters of India. *Curr. Sci.*, 2005, **88**, 107–118.
22. Deshpande, R. D. and Gupta, S. K., National Programme on Isotope Fingerprinting of Waters of India (IWIN). In Glimpses of Geosciences Research in India, the Indian Report to IUGS, Indian National Science Academy (eds Singhvi, A. K., Bhattacharya, A. and Guha, S.), INSA, New Delhi, 2008, pp. 10–16.
23. Deshpande, R. D., Maurya, A. S., Kumar, B., Sarkar, A. and Gupta, S. K., Rain–vapor interaction and vapor source identification using stable isotopes from semi-arid Western India. *J. Geophys. Res.*, 2010, **115**, D23311.
24. Deshpande, R. D. and Gupta, S. K., Oxygen and hydrogen isotopes in hydrological cycle: new data from IWIN National Programme. *Proc. Indian Natl. Sci. Acad.*, 2012, **78**, 321–331.
25. Deshpande, R. D., Maurya, A. S., Kumar, B., Sarkar, A. and Gupta, S. K., Kinetic fractionation of water isotopes during liquid condensation under super-saturated condition. *Geochim. Cosmochim. Acta*, 2013, **100**, 60–72.
26. Maurya, A. S., Shah, M., Deshpande, R. D. and Gupta, S. K., Protocol for $\delta^{18}\text{O}$ and δD analyses of water sample using delta V plus IRMS in CF mode with gas bench II for IWIN National Programme at PRL, Ahmedabad. In 11th ISMAS Triennial Conference of Indian Society for Mass Spectrometry, ISMAS, Hyderabad, 2009, pp. 314–317.
27. Bhandari, N., Murty, S. V. S., Suthar, K. M., Shukla, A. D., Ballabh, G. M., Sisodia, M. S. and Vaya, V. K., The orbit and exposure history of the Piplia Kalan eucite. *Meteor. Planet. Sci.*, 1998, **33**, 455–461.
28. Rozanski, K., Araguas-Araguas, L. and Gonfiantini, R., Isotopic patterns in modern global precipitation. In *Climate Change in Continental Isotopic Records*, American Geophysical Union Monograph, 1993, pp. 1–36.
29. Deshpande, R. D., Groundwater in and around Cambay Basin, Gujarat: some geochemical and isotopic investigations. Ph D thesis, M.S. University of Baroda, Vadodara, 2006, p. 158.
30. Gat, J. R. and Carmi, I., Evolution of the isotopic composition of atmospheric waters in the Mediterranean Sea area. *J. Geophys. Res.*, 1970, **75**, 3039.
31. Gat, J. R., Oxygen and hydrogen isotopes in the hydrological cycle. *Annu. Rev. Earth Planet. Sci.*, 1996, **24**, 225–262.
32. Jouzel, J. and Merlivat, L., Deuterium and oxygen-18 in precipitation: modelling of the isotopic effects during snow formation. *J. Geophys. Res.*, 1984, **89**, 11749.
33. Jouzel, J., Merlivat, L. and Lorius, C., Deuterium excess in an East Antarctic ice core suggests higher relative humidity at the oceanic surface during the last glacial maximum. *Nature*, 1982, **299**, 688–691.
34. Souchez, R., Jouzel, J., Lorrain, R., Sleewaegen, S., Stiévenard, M. and Verbeke, V., A kinetic isotope effect during ice formation by water freezing. *Geophys. Res. Lett.*, 2000, **27**, 1923–1926.
35. Cappa, C. D., Smith, J. D., Drisdell, W. S., Saykally, R. J. and Cohen, R. C., Interpreting the H/D isotope fractionation of liquid water during evaporation without condensation. *J. Phys. Chem. C*, 2007, **111**, 7011–7020.
36. Craig, H., Gordon, L. I. and Horibe, Y., Isotopic exchange effects in the evaporation of water. *J. Geophys. Res.*, 1963, **68**, 5079–5087.
37. Matsui, E., Salati, E., Ribeiro, M., Reis, C. M., Tancredi, A. and Gat, J. R., Precipitation in the Central Amazon Basin: the isotopic composition of rain and atmospheric moisture at Belem and Manaus. *Acta Amazonica*, 1983, **13**, 307.
38. Roesli, H. P., Joss, J. and Schüepp, M., Possible influence of evaporation below cloud base on rain enhancement. *J. Appl. Meteorol.*, 1974, **13**, 783–787.
39. Rosenfeld, D. and Mintz, Y., Evaporation of rain falling from convective clouds as derived from radar measurements. *J. Appl. Meteorol.*, 1988, **27**, 209–215.
40. Stewart, M. K., Stable isotope fractionation due to evaporation and isotopic exchange of falling waterdrops: applications to atmospheric processes and evaporation of lakes. *J. Geophys. Res.*, 1975, **80**, 1133–1146.
41. IAEA/GNIP (International Atomic Energy Agency/Global Network for Isotopes in Precipitation), Water isotope system for data analysis, visualization, and electronic retrieval (WISER); <http://nds121.iaea.org/wiser/>
42. Saul, J., Ice meteorites: is it prudent to ignore anecdotal reports. *Meteorite Q.*, 2006, **12**, 20–21.
43. Beech, M., The problem of ice meteorites. *Meteorite Q.*, 2006, **12**, 17–19.
44. Beech, M. and Nikolova, S., The endurance lifetime of ice fragments in cometary streams. *Planet Space Sci.*, 2001, **29**, 23–29.
45. Kendall, C. and McDonnell, J. J. (eds), *Isotope Tracers in Catchment Hydrology*, Elsevier, Amsterdam, 1998, p. 839.
46. Deloule, E. and Robert, F., Interstellar water in meteorites? *Geochim. Cosmochim. Acta*, 1995, **59**, 4695–4706.
47. Deloule, E., Robert, F. and Doukhan, J. C., Interstellar hydroxyl in meteoritic chondrules: implications for the origin of water in the inner solar system. *Geochim. Cosmochim. Acta*, 1998, **62**, 3367–3378.
48. Hartogh, P. *et al.*, Ocean-like water in the Jupiter-family comet 103P/Hartley2. *Nature*, 2011, **478**, 218–220.
49. Screen, J. A. and MacKenzie, A. R., Aircraft condensation trails and cirrus. *Weather*, 2004, **59**, 116–121.
50. Kalnay, E. *et al.*, The NCEP/NCAP 40-year reanalysis project. *Bull. Am. Meteorol. Soc.*, 1996, **77**, 437–470.
51. Sausen, R., Gierens, K., Ponater, M. and Schumann, U., A diagnostic study of the global distribution of contrails. Part I: present day climate. *Theor. Appl. Climatol.*, 1998, **61**, 127–141.
52. Mannstein, H., Meyer, R. and Wendling, P., Operational detection of contrails from NOAA-AVHRR data. *Int. J. Remote Sensing*, 1999, **20**, 1641–1660.
53. Das, R., Das, S. N. and Misra, V. N., Chemical composition of rainwater and dustfall at Bhubaneswar in the east coast of India. *Atmos. Environ.*, 2005, **39**, 5908–5916.
54. Khemani, L. T., Momin, G. A., Rao, P. S. P., Safai, P. D., Singh, G. and Kapoor, R. K., Spread of acid rain over India. *Atmos. Environ.*, 1989, **23**, 757–762.
55. Momin, G. A. *et al.*, Study of chemical composition of rainwater at an urban (Pune) and a rural (Sinhagad) location in India. *J. Geophys. Res. D*, 2005, **110**, D08302.

56. Rao, P. S. P., Momin, G. A., Safai, P. D., Pillai, A. G. and Khe-
mani, L. T., Rainwater and throughfall chemistry in the Silent
Valley forest in South India. *Atmos. Environ.*, 1995, **29**, 2025–
2029.
57. Satsangi, G. S., Lakhani, A., Khare, P., Singh, S. P., Kumari, K.
M. and Srivastava, S. S., Composition of rain water at a semi-arid
rural site in India. *Atmos. Environ.*, 1998, **32**, 3783–3793.
58. Kulmala, M., Korhonen, P., Vesala, T., Hansson, H.-C., Noone, K.
and Svenningsson, B., The effect of hygroscopicity on cloud drop-
let formation. *Tellus B*, 1996, **48**, 347–360.
59. Rudich, Y., Khersonsky, O. and Rosenfeld, D., Treating clouds
with a grain of salt. *Geophys. Res. Lett.*, 2002, **29**, 17-1–17-4.
60. Drofa, A. S., Ivanov, V. N., Rosenfeld, D. and Shilin, A. G.,
Studying an effect of salt powder seeding used for precipitation
enhancement from convective clouds. *Atmos. Chem. Phys.*, 2010,
10, 8011–8023.
61. Pierce, J. R. and Adams, P. J., Global evaluation of CCN forma-
tion by direct emission of sea salt and growth of ultrafine sea salt.
J. Geophys. Res. D, 2006, **111**, D06203.
62. Bilde, M. and Svenningsson, B., CCN activation of slightly solu-
ble organics: the importance of small amounts of inorganic salt
and particle phase. *Tellus B*, 2004, **56**, 128–134.
63. Svenningsson, B. *et al.*, Hygroscopic growth and critical super-
saturations for mixed aerosol particles of inorganic and organic
compounds of atmospheric relevance. *Atmos. Chem. Phys. Dis-
cuss.*, 2005, **5**, 2833–2877.
64. Gautier, D. and Owen, T., Cosmological implications of elemental
and isotopic abundances in atmospheres of giants planets. *Nature*,
1983, **304**, 691–694.
65. Geiss, J. and Reeves, H., Cosmic and solar system abundances
of deuterium and helium-3. *Astron. Astrophys.*, 1972, **18**, 126–
132.
66. Geiss, J. and Reeves, H., Deuterium in the early solar system.
Astron. Astrophys., 1981, **93**, 189–199.
67. Geiss, J., Primordial abundance of hydrogen and helium isotopes.
In *Origin and Evolution of the Elements* (ed. Prantzos, N.), Cam-
bridge University Press, 1993, pp. 89–106.
68. Lecuyer, C., Gillet, P. and Robert, F., The hydrogen isotope com-
position of seawater and the global water cycle. *Chem. Geol.*,
1998, **145**, 249–261.
69. Balsiger, H., Altwegg, K. and Geiss, J., D/H and 180/160 ratio in
the hydronium ion and in neutral water from in situ ion measure-
ments in comet Halley. *J. Geophys. Res. A*, 1995, **100**, 5827–
5834.
70. Eberhardt, P., Reber, M., Krankowsky, D. and Hodges, R. R., The
D/H and $^{18}\text{O}/^{16}\text{O}$ ratios in water from comet P/Halley. *Astron.
Astrophys.*, 1995, **302**, 301–316.
71. Wyckoff, S., Comets: clues to the early history of the solar sys-
tem. *Earth Sci. Rev.*, 1991, **30**, 125–174.
72. Bokelée-Morvan, D. *et al.*, Deuterated water in comet C/1996 B2
(Hyakutake) and its implications for the origin of comets. *Icarus*,
1998, **133**, 147–162.
73. Boato, G., The isotopic composition of hydrogen and carbon in the
carbonaceous chondrites. *Geochim. Cosmochim. Acta*, 1954, **6**,
209–220.
74. Robert, F., Merlivat, L. and Javoy, M., Deuterium concentration in
the early solar system: a hydrogen and oxygen study. *Nature*,
1979, **282**, 785–789.
75. Kerridge, J. F., Carbon, hydrogen and nitrogen in carbonaceous
meteorites: abundances and isotopic compositions in bulk sam-
ples. *Geochim. Cosmochim. Acta*, 1985, **49**, 1707–1714.
76. Anders, E., Hayatsu, R. and Studier, M. H., Organic compounds in
meteorites. *Science*, 1973, **182**, 781–790.
77. Robert, F. and Epstein, S., The concentration and isotopic compo-
sition of hydrogen, carbon, and nitrogen in carbonaceous meteor-
ites. *Geochim. Cosmochim. Acta*, 1982, **46**, 81–95.
78. Yang, J. and Epstein, S., Interstellar organic matter in meteorites.
Geochim. Cosmochim. Acta, 1983, **47**, 2199–2216.
79. Robert, F., Javoy, M., Halbout, J., Dimon, B. and Merlivat, L.,
Hydrogen isotope abundances in the solar system. Part I: unequi-
librated chondrites. *Geochim. Cosmochim. Acta*, 1987, **51**, 1787–
1805.
80. Halbout, J., Robert, F. and Javoy, M., Hydrogen and oxygen iso-
tope compositions in kerogens from the Orgueil meteorite: clues
to solar origin. *Geochim. Cosmochim. Acta*, 1990, **54**, 1453–1462.
81. Hageman, R., Nief, G. and Roth, E., Absolute isotopic scale for
deuterium analysis of natural waters. Absolute D/H ratio for
SMOW. *Tellus*, 1970, **22**, 712–715.

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