Acknowledgements

This work was supported in part by the Natural Sciences and Engineering Research Council of Canada, Photonics Research Ontario, the Fundacion Ramon Areces, the Spanish CICyT project and the European Community Project. We are grateful to I. Sokolov and N. Coombs for their technical assistance with the AFM and SEM images (Figs 1 and 2).

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Deep convective clouds with sustained supercooled liquid water down to −37.5 °C

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In cirrus[1] and orographic wave clouds[2], highly supercooled water has been observed in small quantities (less than 0.15 g m^{-3}). This high degree of supercooling was attributed to the small droplet size and the lack of ice nuclei at the heights of these clouds[1,2]. For deep convective clouds, which have much larger droplets near their tops and which take in aerosols from near the ground, no such measurements have hitherto been reported. However, satellite data suggest that highly supercooled water (down to −38 °C) frequently occurs in vigorous continental convective storms[3]. Here we report *in situ* measurements in deep convective clouds from an aircraft, showing that most of the condensed water remains liquid down to −37.5 °C. The droplets reach a median volume diameter of 17 μm and amount to 1.8 g m^{-3}, one order of magnitude more than previously reported[2]. At slightly colder temperatures only ice was found, suggesting homogeneous freezing. Because of the poor knowledge of mixed-phase cloud processes[4], the simulation of clouds using numerical models is difficult at present. Our observations will help to understand these cloud processes, such as rainfall, hail, and cloud electrification, together with their implications for the climate system.

Cloud droplets do not readily freeze at 0 °C, but often remain liquid at colder temperatures in a "supercooled" state. Freezing of the cloud droplets can then be triggered by ice nuclei, or the droplets can freeze in an homogeneous fashion. The lowest temperatures at which pure water droplets can exist in a supercooled state for times longer than a fraction of a second before homogeneously freezing depends on the drop size. According to both theory[5] and laboratory experiments[6], a 10-μm cloud droplet freezes homogeneously near −39 °C. The coldest previously reported *in situ* measurements of supercooled liquid water content (SLWC) at temperatures below −32 °C, in excess of the sensitivity of the measuring instruments (0.02 g m^{-3} for hot-wire probes[7]), was 0.14 g m^{-3} at −36 °C in orographic lenticular wave clouds[2]. An SLWC of 0.06 g m^{-3} at the base of cirrus clouds, between −35° and −36 °C, was reported by Sassen[1], who stated that "in comparison with earlier reported aircraft measurements, the detection of such highly supercooled water is unique".

Both reports[1,2] suggested that the dearth of ice nuclei derived from the Earth's surface in the upper troposphere prevented heterogeneous nucleation, allowing for the observed homogeneous nucleation at such cold temperatures in altocumulus and cirrus clouds[8]. No previous reports are available for observations of similar highly supercooled water and homogeneous freezing in convective clouds with roots near the surface. In view of the reported findings to the contrary here, we can only speculate about the reasons they have not been reported previously. Perhaps low priority was given to such measurements, because it was felt that clouds taking in air rich in ice nuclei from the boundary layer would glaciate long before reaching the point of homogeneous freezing[9]. Or perhaps such measurements were not made because of the safety problems involved in penetrating vigorous cumulonimbus towers at the −30 to −40 °C isotherm levels in storms that typically contain hail and frequent lightning.

The first indications available to us that highly supercooled water might exist in convective clouds were obtained by remote sensing from satellites over Thailand, using the technique reported in ref. 3. The inference of supercooled water at temperatures below −30 °C prompted us to fly with the Thai King Air cloud-physics aircraft to measure the cloud microstructure in Thailand clouds. Penetrating feeders of cumulonimbus clouds, an SLWC of 2.4 g m^{-3} was measured at the operational ceiling of the aircraft (9,300 m above sea level, a.s.l.) at a temperature of −31.6 °C (ref. 10). The cloud-base temperature was +13 °C at 2,800 m a.s.l., and the SLWC was measured by the King hot-wire instrument.

Analysis of the satellite data using the methodology of Rosenfeld and Lensky[3] suggested that supercooled water occasionally occurred at temperatures approaching −40 °C in cumulonimbus clouds over the western USA. An opportunity to validate these satellite inferences came when Weather Modification Inc. gave us access to its Lear jet cloud-physics aircraft in the period 9–14 August 1999 for measurements in Texas clouds. The aircraft cloud-physics instrumentation included the following: (1) a hot-wire cloud liquid water probe, model LWC-100 (Droplet Measurement Technologies). The measurement efficiency of the sensor depends on the drop sizes. For the observed distribution, it underestimated the SLWC by not more than 10%. (2) An air temperature probe, model 102AU1AP (Rosemount). (3) A forward scattering spectrometer probe (FSSP) for droplet sizes in the range 0.5–47 μm, model FSSP-100 (Particle Measuring Systems Inc). (4) An optical array particle imaging probe, for the range 25—800 μm, model OAP-2D2-C (Particle Measuring Systems, Inc.).

High-altitude measurements in the tops of vigorous growing convective elements of cumulonimbus clouds were done with the Lear jet on 11 and 13 August 1999. The visibly most-vigorous new convective elements were measured as they grew through the measurement flight level. In addition, extensive measurements were made at all levels down to the cloud base, in order to document the vertical microphysical evolution of the cloud. Cloud base on both days was near 3,500 m a.s.l. at a temperature of 10 °C.

On the flight of 11 August (20:28–23:42 GMT) just to the west of Lubbock, Texas (34° N, 102° W), an SLWC of 0.6 g m^{-3} was observed at −35.9 °C, 0.9 g m^{-3} at −35.6 °C, and 1.5 g m^{-3} at −34.4 °C. Larger values of SLWC, up to 2.4 g m^{-3}, were recorded at warmer cloud temperatures. Direct measurements of the updraught velocity were not available. However, a rate of climb of 6 m s^{-1} was sufficient to keep up with the rate of growth of the tops of some of the clouds containing highly supercooled water. The residence time of the water was measured by repeated penetrations in the same cloud, which was narrow and clearly isolated. In three passes spaced at 3.5-min intervals, the temperatures and maximum cloud water contents were 1.2 g m^{-3} at −32.9 °C, 1.5 g m^{-3} at −32.7 °C, and 0.4 g m^{-3} at −35.2 °C. This means that the large amounts of highly supercooled cloud water were not a transient feature, but rather were long lasting, with freezing time of about 7 min.

On the flight of 13 August (20:26–23:09 GMT), to the north of Midland, Texas (33°N 102°W), effort was focused on documentation of the transition from water clouds to ice clouds in vigorous convective elements. Extensive documentation of the clouds from

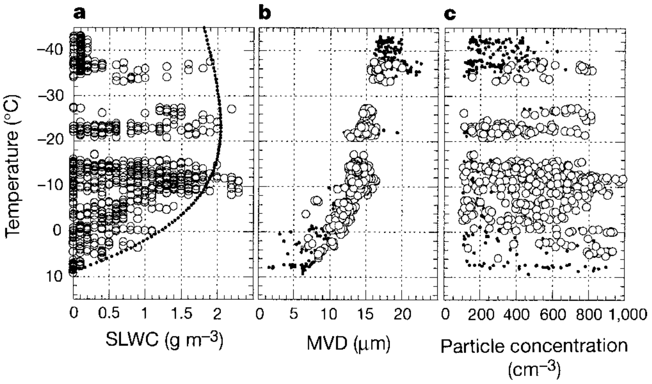

Figure 1 Cloud microstructure and composition as a function of temperature. Data were obtained on 13 August 1999. Each point represents one second of measurements, or 150–200 m of cloud path. **a**, The supercooled liquid water content (SLWC). Note the abrupt decrease of SLWC at −38 °C, indicating the point of homogeneous freezing. The dotted curve marks half the adiabatic value of the water content, as a reference for the rate of consumption of cloud water by freezing and other processes. **b**, The median volume diameter (MVD; as measured by the FSSP instrument) of the cloud particles for all points containing more than 100 particles $\mathrm{cm^{-3}}$. The open circles denote cloud segments with liquid water contents greater than $0.3\,\mathrm{g\,m^{-3}}$, as measured by the hot-wire instrument. **c**, As **b**, but for the FSSP-measured concentration of cloud particles.

their bases was done as on the 11 August. The vertical evolution of cloud microstructure was found to be similar for both days. We therefore show in Fig. 1 only data from 13 August. As illustrated in Fig. 1a, maximum SLWC values of nearly half the adiabatic water content was measured throughout the cloud depth, up to the −37.5 °C isotherm. The aircraft windscreen was instantly covered with rime ice during readings of high SLWC despite the windscreen heater, showing that there really was ample supercooled water at these very low temperatures. An abrupt vanishing of the SLWC was recorded at colder temperatures in the same clouds. The remaining reading of up to $0.2\,\mathrm{g\,m^{-3}}$ in obviously fully glaciated clouds (that is, at temperatures < −40 °C, where theory dictates that droplets greater than 1 μm in size must freeze homogeneously[5]) is attributed to the cooling effect of the frozen cloud drops on the hot wire (D. Baumgardner, personal communication). The lack of significant freezing at higher temperatures, and the sudden freezing at −38 °C, indicates that homogeneous freezing is the main glaciating mechanism in the measured convective clouds. This observation has a number of implications.

First, heterogeneous nucleation by ice nuclei was incapable of freezing much of the cloud water. However, it not likely that this was because of a dearth of ice nuclei. The air ingested into the cloud base had substantial amounts of aerosols of unknown composition. The FSSP measured average concentrations of >0.5-μm particles of 0.5 particles $\mathrm{cm^{-3}}$. Most insoluble aerosols of that size become ice nuclei below −25 °C. A possible explanation might be the very small collision efficiencies between the <200-μm ice crystals that were nucleated heterogeneously and the <40-μm cloud droplets (ref. 4).

Second, most of the water remained in the form of large concentrations of small cloud droplets (Fig. 1b), with FSSP-measured median volume diameter (MVD) increasing with height, reaching 17 μm at the −37.5 °C isotherm, just below the height of homogeneous freezing (see Fig. 1c).

Third, although the FSSP cannot resolve ice from water, the fact that the SLWC was half the adiabatic water content implies the existence of liquid water droplets of at least the MVD size of 17 μm. Such droplets freeze homogeneously near −38 °C (ref. 5).

Fourth, the nearly constant water content (of half the adiabatic value; see Fig. 1b) and the small MVD of the cloud droplets suggest that only a small fraction of the cloud water was converted into precipitation. That is confirmed by the optical array particle imaging probe measurements, showing that ice particles of greater than ~50 μm exceed a concentration of $\sim 10\,\mathrm{l^{-1}}$ in the high-SLWC clouds only at temperatures below −30 °C.

Fifth, the maintenance of the MVD through the glaciation at −38 °C indicates that the cloud droplets freeze into small ice particles, presumably in the form of frozen droplets. This ice leaves the cloud through the cumulonimbus anvil, and is not likely to contribute to the precipitation. Therefore, the observation of apparent homogeneous freezing in cumulonimbus clouds indicates very poor precipitation efficiency.

Sixth, the deep layer of high SLWC provides large growth potential for the small concentration of ice precipitation particles that are initiated in the lower parts of the updraft. This means that such highly supercooled and persistent SLWC represents favourable conditions for the growth of large hail.

Last, given the empirical necessity for liquid water for appreciable electrification in laboratory experiments[11], the extension of supercooled droplets to greater heights above the 0 °C isotherm might explain observations[12] that deep continental clouds have the strongest electrification of all cumulonimbus types.

The accuracy of the instrument reading is critical in the context of these findings. A calibration check was made to the instruments before and after the reported flights, and they were found to have been properly calibrated. The temperature readings were validated against the radiosonde balloon, which was launched from Midland at 00 GMT on 14 August. The sounding was representative, because it was within 100 km and 1 h of the time of the aircraft measurements. The comparison of the aircraft-measured temperature (T_a) with the sounding temperature (T_s) at the point where $1.8\,\mathrm{g\,m^{-3}}$ of SLWC was measured shows the following. In cloud, at a static pressure of 265 hPa, $T_a = -37.5\,^{\circ}\mathrm{C}$; T_s at the same pressure was −38.1 °C. Out of cloud in level flight, $T_a = -37.5\,^{\circ}\mathrm{C}$ at a static pressure of 260 hPa, whereas T_s was −39.3 °C at the same pressure. Direct checks of the temperature probe on the ground provided: $T_a = +0.4\,^{\circ}\mathrm{C}$ at 0 °C, and $T_a = -9.7\,^{\circ}\mathrm{C}$ at −10 °C. These comparisons suggest that our measurements provide a conservative estimate of the temperature at which homogeneous freezing took place. It might in fact have occurred at a slightly lower temperature.

We note that the Lear jet was available for only 6 days, and on only 2 days were attempts made to document the highly supercooled portions of the clouds, although potentially suitable clouds occurred also on other days of that week. The fact that the highly supercooled water was found in both the two attempts strengthens the evidence from satellite indications that the reported observations are not rare occurrences.

We report here that, in some clouds, most of the condensed water remains liquid until the point of homogeneous freezing; we consider that this will necessitate a significant revision of cloud models simulating rain, hail, and cloud electrification. Incorporation of these changes in global circulation models will probably produce substantial changes in our understanding of the way that aerosols, clouds and precipitation are affecting the global climate. For example, aerosols that serve as small cloud condensation nuclei—which make the clouds more continental (that is, with smaller droplets)—might reduce their precipitation efficiency and thus inhibit rainfall in polluted areas. Reduced precipitation would mean more water vapour flux to the upper troposphere and lower stratosphere, and reduced net latent heat release, which in fact would change both the radiative and heating forcing of the climate system.

Note added in proof: Observations conducted by the authors in the last week of January and the first week of February 2000 at Mendoza, Argentina, documented five more cases of supercooled cloud water: up to $4\,\mathrm{g\,m^{-3}}$ of supercooled cloud water was found at −38 °C, vanishing abruptly at lower temperatures. ☐

Received 3 September 1999; accepted 17 April 2000.

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Acknowledgements

We thank P. Sweeney, of Weather Modification, Inc., for providing the WMI Lear jet at no charge; we also thank R. Tilbury and S. Gordon for piloting the aircraft through the vigorous Texas clouds, and K. Bosch for technical support.

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Past temperature and δ^{18}O of surface ocean waters inferred from foraminiferal Mg/Ca ratios

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Determining the past record of temperature and salinity of ocean surface waters is essential for understanding past changes in climate, such as those which occur across glacial–interglacial transitions. As a useful proxy, the oxygen isotope composition (δ^{18}O) of calcite from planktonic foraminifera has been shown to reflect both surface temperature and seawater δ^{18}O, itself an indicator of global ice volume and salinity[1,2]. In addition, magnesium/calcium (Mg/Ca) ratios in foraminiferal calcite show a temperature dependence[3–5] due to the partitioning of Mg during calcification. Here we demonstrate, in a field-based calibration experiment, that the variation of Mg/Ca ratios with temperature is similar for eight species of planktonic foraminifera (when accounting for Mg dissolution effects). Using a multi-species record from the Last Glacial Maximum in the North Atlantic Ocean we found that past temperatures reconstructed from Mg/Ca ratios followed the two other palaeotemperature proxies: faunal abundance[6,7] and alkenone saturation[8]. Moreover, combining Mg/Ca and δ^{18}O data from the same faunal assemblage, we show that reconstructed surface water δ^{18}O from all foraminiferal species record the same glacial–interglacial change—representing changing hydrography and global ice volume. This reinforces the potential of this combined technique in probing past ocean–climate interactions.

Mg/Ca ratios have recently been used to construct a low-resolution, deep water temperature record for the Cenozoic era[9]. But there has been no validation of the method from detailed field-based calibrations for multiple species and comparison with other temperature proxies. High-resolution records obtained from planktonic foraminifera are needed to examine the response of the ocean surface to glacial–interglacial change and to provide surface ocean temperature estimates for boundary conditions of global models. Early studies of planktonic foraminifera gave conflicting evidence of a temperature effect on Mg partitioning (see, for example, ref. 10), but more recent work has yielded correlations between Mg concentrations of *Neogloboquadrina pachyderma* and annual sea surface temperature (SST) in modern core tops[3] and temperature-dependent Mg uptake by *Golobigerinoides sacculifer*, *Globigerina bulloides* and *Orbulina universa* in culture[4,5] (there is one single benthic Mg calibration based on a water depth transect[11]), together with some initial applications of the method[12,13]. Calibrations under culture are very valuable, but a prerequisite for palaeoceanographic applications is a demonstration that the temperature sensitivity of foraminiferal material is retained from their chamber formation through the development of gametogenic calcite—which thickens the shell at about the time of gamete release—and crust, the effects of dissolution, to residence at the sea bed. Core-top sediment

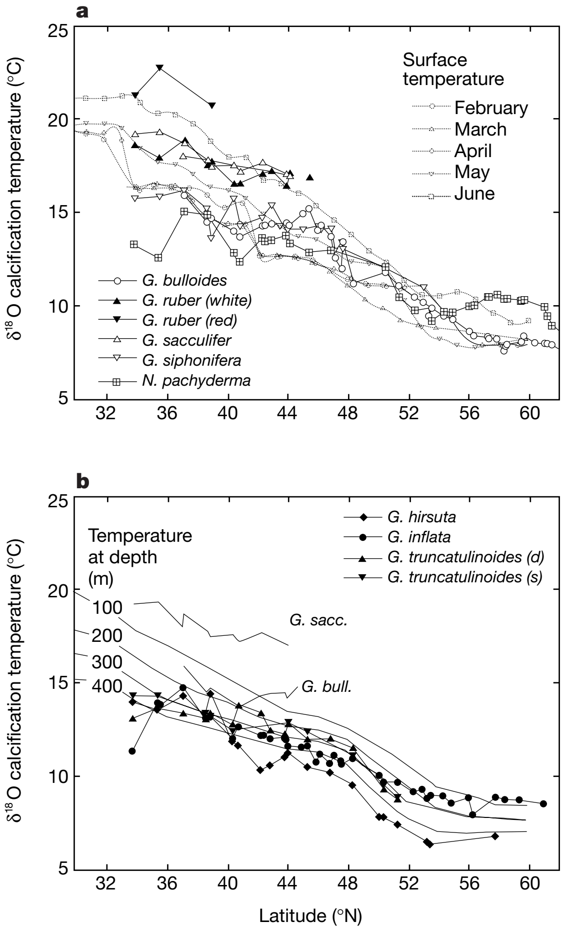

Figure 1 Foraminiferal δ^{18}O data versus latitude of core top compared with temperature data from an atlas[15]. **a**, Monthly surface temperatures. **b**, Annual surface temperatures at different water depths.