Synopsis

Compositional Heterogeneity of Hailstones: Atmospheric Conditions and Possible Environmental Implications

Hail is precipitation in the form of balls or irregular chunks of ice, which are usually produced by convective clouds, in association with multicell, supercell, and cold fronts. Hail begins as tiny ice pellets that collide with water droplets. As the attached droplets freeze, the pellets become larger. An icy conglomeration is called a hailstone when it reaches a diameter of around 5 mm or more (1). Broadly, hailstones are the result of the updrafts and down drafts which take place inside the cumulonimbus clouds of a thunderstorm, where supercooled water droplets exist. Continued deposits of supercooled water cause the ice crystals to grow into hailstones that generally have passed through several stages of accretion, from the first stage (graupel), to small hail, to hailstones. The more times a hailstone is tossed up and down through the cloud, the larger the hailstone will be. Hailstones can reach a speed of 90 mph (140 km hr^{-1}) as they fall to the ground!

From 8 to 17 January 2000, numerous unusually big hailstones—weighing from around 300 g to more that 3 kg—fell in different parts of Spain under unusual atmospheric conditions (2, 3) producing damages in cars and industrial storage. Documented references regarding the fall of large blocks of ice go back to the first half of the 19th century; e.g. 1829 in Córdoba, Spain: 2 kg; 1851 in New Hampshire: 1 kg. The fall of a large hailstone, which measured 26 x 14 x 12 cm and weighed 2.04 kg, is cited, in Germany, in 1936 (4). More recently, some authors (5–8) give many other cases of large hailstones; for instance a large block of ice of almost 2 kg which fell in Kazakhistan, and one of almost one kg which fell at Strassbourg. Probably, the best-documented fall of an ice chunk was April 2, 1973 in Manchester, England. The block weighed 2 kg and consisted of 51 layers of ice. Its origin was not determined (9). For many years the largest hailstone officially reported in the United States was one that fell at Potter, Nebraska, on 6 July 1928. It had a circumference of 43 cm and weighed 680 g. This record was surpassed on 3 September 1970 at Coffeyville, Kansas, USA. The giant hailstone measured 18 cm accross,

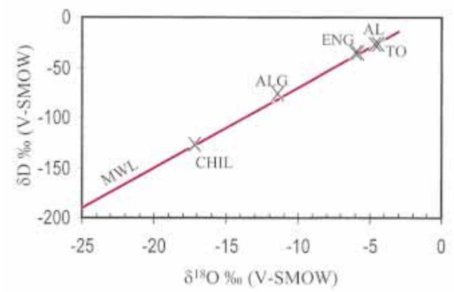
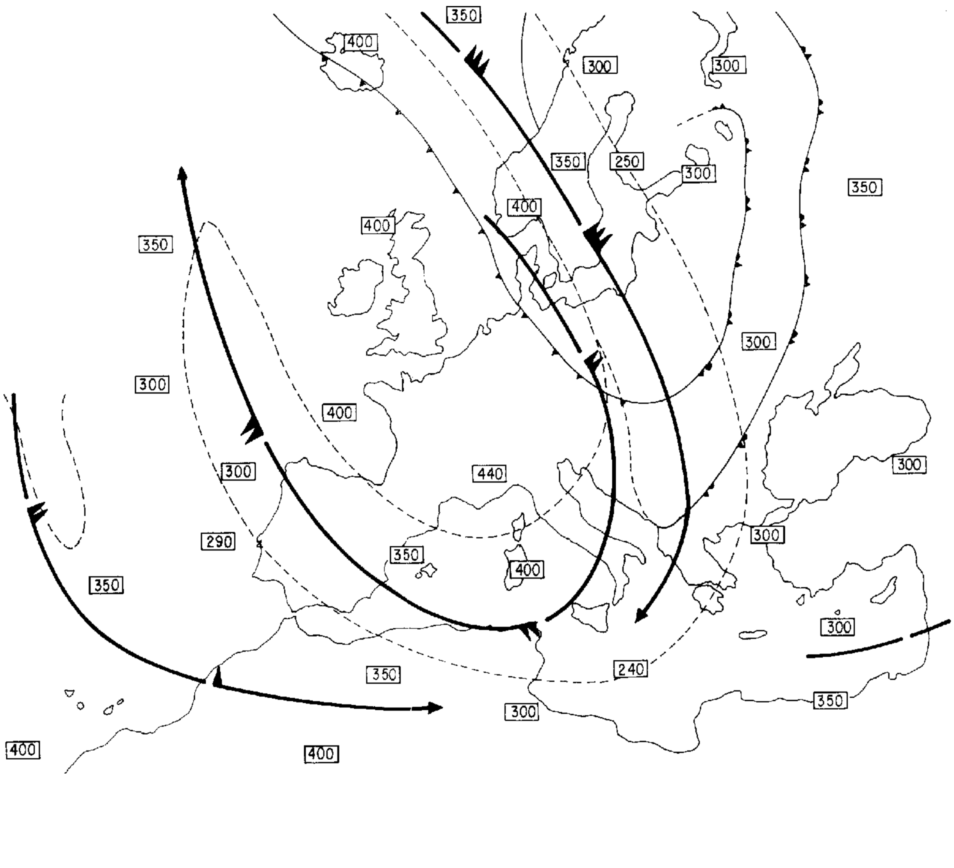


Figure 1. δ18O (V-SMOW) vs. δD (V-SMOW) values of selected hailstones which fell in different areas of Spain. Note the sample(s) match with the Meteoric Water Line (Craig's Line). CHIL: Chilches; ALG: Algemesí; ENG: Enguera; AL: Alcudia; TO: Tocina.

Figure 2. Maps of Europe obtained from the World Area Forecast Center, London, UK, displaying the significant wind shear and tropopause heights (in feet x 10^2) which correspond to January 17th 2000. Note the low values of the tropopause height in some areas (< 250 x 102 feet). Wind speed is in knots. Each triangle indicates 50 knots and each associated line 10 knots.



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about 44 cm in circumference, and weighed more than 750 grams (10).

CHARACTERIZATION AND ATMOSPHERIC SCENARIO

Textural and Compositional Features

The turbulent updrafts and downdrafts within the cloud send the hailstones up and down several times, where they gather layer upon layer of ice. The number of layers in a hailstone reveals the number of up-down journeys it has made before reaching the earth. Thus, most hailstones acquire onion skin layers from traveling up and down in a storm. However, recent research (11) has shown that there is not one simple process of hail formation. Hailstones may actually form in several ways. Some hailstones can grow while balanced in an updraft and have little layering. Stones may also form around raindrops that are carried high into the storm and freeze. Finally, some hailstones form around ice crystals (11).

Hailstones that fell in Spain display variable fabric properties which include zones of "massive ice", large isolated cavities, mm-sized oriented air bubbles, and ice layering. The thickness of the layers range from less than 1 mm to more than 1 cm. Also, tiny solid particles can be found randomly disseminated in the interior of some blocks. First chemical and isotopic analyses (2) evidenced compositional heterogeneity with large densities of ions—up to 5 times larger than normal meteoric waters—and corresponding to solutions of halite, calcite, anhydrite and quartz or feldspar aerosols. New hydrochemical analyses, by the combination of capillary electrophoresis, molecular absorption spectrometry (UV-Vis) and ICP-AES, indicate that the blocks of ice are formed from waters of variable mineralization (between 106 and 858 μS cm^{-1}), with very low values of SiO_2 (< 0.7 ppm), and the presence of NH_4 (0.21 to 0.78 ppm) in all samples. $\delta^{18}O$ and δD (V-SMOW) of the samples fall within the Meteoric Water Line (12). The distribution of the samples on Craig's line (Fig. 1) suggests either a variation in condensation temperature and/or different residual fractions of water vapor (Rayleigh processes) (13). The most positive values are typical of rainwater in Spain. Isotopic mapping of δD values in the hailstones display: *i*) significant general variations from –24.4‰ to –126.4‰, and b) specific variations of up to 25 δD within some individual blocks.

THEORETICAL MODELING

In order to try to explain the physical parameters, which could rule the formation of the hailstones, we have modelled a plausible scenario for the events, based on the well-known theory of nucleation (14). The free energy for nucleation in homogeneous media is:

$$G = -(S-1) * \mu(T) * n + \gamma(T) * n^{2/3} \quad \text{Eq. 1}$$

where μ and γ are the chemical potential of equilibrium and the surface free energy for a given temperature, respectively, and S and n are the supersaturation of the vapor and the number of molecules in the aggregate. The critical nuclei is given, taking $dG / dn = 0$, by

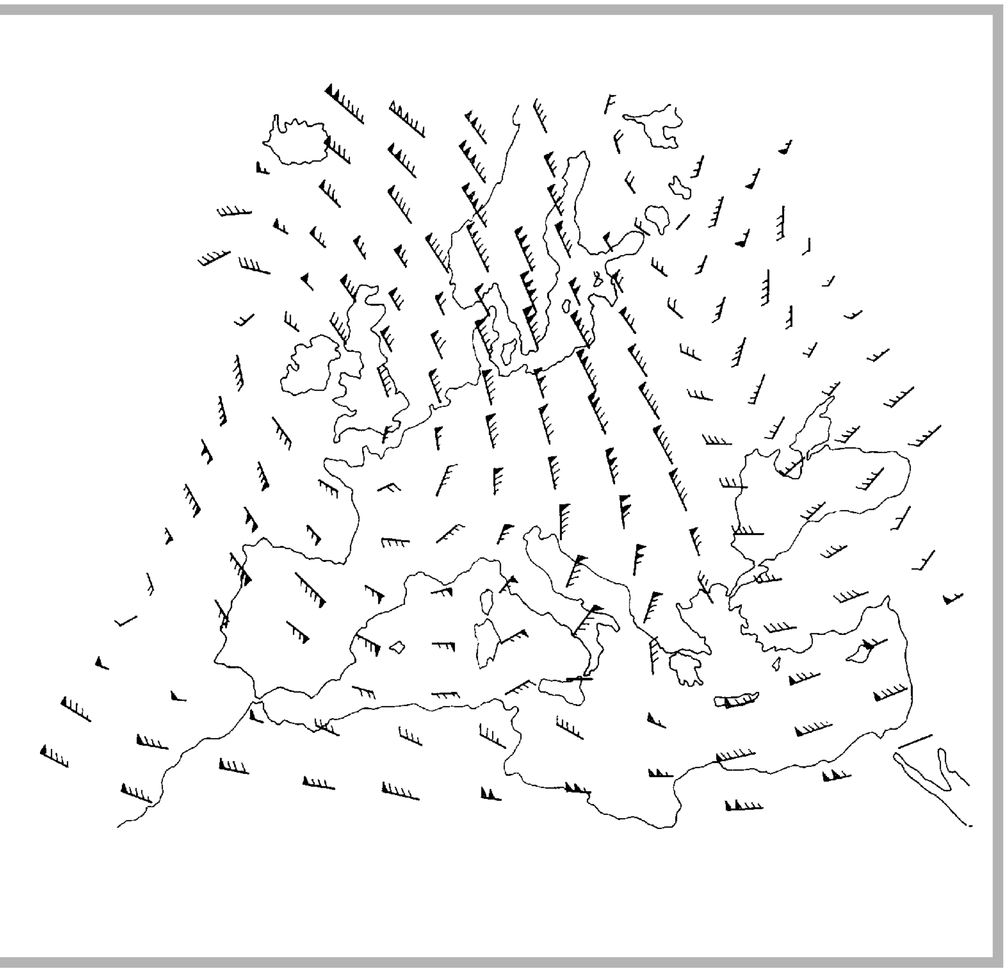
$$n_c = \left(\frac{2\gamma}{3\mu * (S-1)}\right)^{1/3} \quad \text{Eq. 2}$$

Therefore, when $S >> 1$ many nuclei are formed close to each other because the value G_c (critical free energy) is accessible by T fluctuations. The distance between the nuclei is small, and they cannot grow too much, because they are all competing for vapour molecules. When $S < 1$ there is no condensation. For $S \approx 1$ the value of n_c tends to infinity. However, the critical energy of nucleation is extremely high and condensation cannot take place due to temperature fluctuations alone.

Nevertheless, if an external *perturbation* (see below) is produced within the vapor volume (i.e. extra cooling, injection of ion concentration—heterogeneous nucleation—irruption of a sound wave, etc.), then nuclei will form at large distances from each other. The particle condensed will be of ice if the temperature is well below zero, and they can grow large at the expense of surrounding molecules, as the nuclei are scattered. Theoretical estimations show that the radius R of the ice chunks is

$$R \approx \frac{\rho_v}{\rho_W}\left(1 + \frac{V_T}{V}\right)h \quad \textit{Eq.3}$$

where ρ_v, ρ_w, V_T, V and h are the gas and water densities, the thermal and falling velocities and the height of the gas volume. This gives $R = 6$ cm, 4 cm and 2 cm for $S \sim 1$ at $T = -5$, –10 and –20ºC, and $h \sim 1$ km. It is to be noticed that the growing process is controlled by the thermal velocity of the water molecules. In the above calculations, the latent heat of the hailstone is taken away by the carrier air molecules. Also, the nucleation at S = >1 is consistent with the large number of ions in the specimens detected by the chemical analyses. Other alternative possibility could be that ice crystallites from the frozen stratosphere, where they are known to exist in clouds, enter a region of larger humidity (from around 7 or 8 km down) and starts growing.



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ATMOSPHERIC CONDITIONS AND POSSIBLE ENVIRONMENTAL IMPLICATIONS

One of the main aims of the climatic research is the analysis of anthropogenic environmental influence on natural processes. Within this scope, the study of precipitation (rain, hail, snow, etc.), as the most important cleansing mechanism of the atmosphere, is the particular interest. It has been suggested (15) that the formation of precipitation in the temperate climate of Europe is closely related to the existence of mixed-phase clouds (i.e. clouds containing both supercooled cloud droplets and ice crystals). Nevertheless, a great uncertainty still exists regarding the mechanisms, which are responsible for the formation of the initial nuclei.

Thus, a first significant point is to find out what could be the cause of the atmospheric *perturbation* that led to the ice nucleation. Could it be caused by extra ionization following a depression of ozone in the area? A lower ozone concentration implies more intense UV radiation entering the atmosphere, and additional ionization. As previously defined, the hailstones fell from January 8 to 17. The examination of ozone distribution maps from NASA shows that, on January 5, a thin jet of ozone depression passed through all the areas in Spain where the falls of hailstone occurred. It is commonly posited that global warming and ozone depressions are linked. Despite the fact that the greenhouse effect leads to an increase in the GMST (Global Means Surface Temperature), in the stratosphere, it actually leads to cooling. As Malham of NOAA's Geophysical Fluid Dynamics Laboratory indicates, cooling of the stratosphere is one of the most certain results of the greenhouse gas build-up (16). In addition, it is important to note that a significant relationship exists between water vapor and ozone that can involve serious environmental consequences. Water vapor in the atmosphere is the key trace gas controlling weather and climate, and plays a central role in atmospheric chemistry, influencing the heterogeneous chemical reactions that destroy stratospheric ozone. An increase in water vapor could thus lead to greater ozone loss (17).

A second critical atmospheric feature, which contributes to the formation of large hailstones, is the existence of significant changes in wind velocity with increasing height (shear). This shear has to be such that small hail fired out of the top of the storm eventually falls into the strong inflow at the base of the storm to be swept round again, perhaps several times. With each passage through the storm, the size of the chunk increases. A good example can be found in the High Plains of the United States. This usually occurs when an elevated mixed layer of dry air is advected over the region from the higher Mexican plateaus under mid-level south-westerly flow conditions (18). When this elevated mixed layer, with its high temperature lapse rates, overlays a relatively shallow surface layer of moist air advected northwest from the Gulf of Mexico, very unstable atmospheric conditions develop that result in strong thunderstorm updrafts capable of producing giant hail (greater than 7 cm). A similar mechanism probably took place in Spain. Figure 2 shows a significant wind shear that affected the Iberian Peninsula, and other areas of Europe, at 17 January 2000, when some of the falls of large hailstones occurred.

Different works have tackled the nature and principal features of the tropopause. This dynamic level divides the upper troposphere from the lowermost stratosphere exhibiting unusual singularities (i.e. undulations), which condition many atmospheric interchange mechanisms affecting the formation and development of hydrometeors (19). Tropospheric undulations are displayed as a downward extension of the tropopause, due to descent at and above the tropopause. Besides the coalescence of the singular atmospheric features described above, two events were identified in Spain in which the typical "near-surface moist(er) layer", capped by a subsidence inversion, became replaced by a much deeper, nearly neutral, moist layer which extended to a lowered tropopause. In fact, during these events the tropopause was observed to sink from about 10–11 km to as low as 7 km over the northern Spanish coast (La Coruña) on 13–14 January. These two events took place from the 8 to the 11, and from the 13 to the 16. In both cases, the moist air mass moved from west to east and covered the whole of Spain and surrounding coastal areas.

During the 38-year period of 1955–1993, there was a large increase in the number of large (> 4 inches) hail reports (20). In accordance with the scientific priorities for Global Atmospheric Chemistry Research in Europe, the new directions in environmental strategies require the study of the atmosphere as a whole, taking into account the interactions and modifications to different scales. Some environmental problems linked with the study of soils, toxic snow, glaciers and permafrost (21–23) demonstrate that climate change can be manifest in different ways giving rise to different types of impacts. We suggest paying more attention to the fall of these unusually large hailstones, which could be indicating that changes are taking place in the atmosphere, probably involving complex mechanisms in which aerosols, ozone depression, water vapor, atmospheric extra-cooling, tropopause sinking and wind shear conditions are interacting.

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