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THE HYDROGEN AND DEUTERIUM CONCENTRATIONS IN CHONDRITES.

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The hydrogen isotopic ratio in the protosolar nebula has been estimated on the basis of [1] i) spectroscopic data obtained inside and outside the solar system, ii) $^3\text{He}/^4\text{He}$ and $^4\text{He}/\text{H}$ ratios measured in the solar wind, on the surface of the moon and in the carbonaceous chondrites, using the assumption that the sun has totally converted the deuterium into ^3He .

As regards the terrestrial planets, evolution of the primitive atmospheres has no doubt altered the initial D/H ratio through mechanisms occurring, during the earth degassing. On the moon it has not been possible to detect any primordial hydrogen as the nuclear phenomena blank out the (hypothetical) primordial deuterium of lunar rocks. BOATO [2] has measured the D/H ratio in carbonaceous chondrites. He was not able to show exactly the influence of terrestrial pollution on extraterrestrial rocks and no interpretation has been put forward to explain the large isotopic variations in carbonaceous chondrites.

As things now stand, three problems remain unsolved : i) the influence of terrestrial contamination on water and deuterium concentrations extracted from chondrites, must be estimated ii) some of the results on carbonaceous chondrites show a deuterium enrichment by a factor 10 when compared with the average isotopic ratio of the estimated primordial hydrogen ($1.9 \pm 1.1 \cdot 10^{-5}$). One must suggest therefore, a mechanism that can explain this enrichment within the framework of modern theories of the formation of the solar system, or suppose large isotopic heterogeneities iii) new measurements on ordinary chondrites are needed in order to ascertain whether this isotopic enrichment is an overall problem of telluric planets or whether it only affects a few individual carbonaceous chondrites.

I - Experimental methods.

The water is extracted from the rock in a two step process : first at 180°C during 4 hours (H_2O^-) and then up to 1200°C for 1.5 hours (H_2O^+). The first extracted water is called H_2O^- and probably represents mainly the adsorbed terrestrial water. The water is reduced to H_2 and the amount and isotopic ratio of gas are measured with a double collecting mass spectrometer. The isotope analyses are reported in δ notation as the permil deviation of the sample D/H ratio to the sample SMOW (Standard Mean Ocean Water) :

$$\left[\left(\frac{(D/H)_s}{(D/H)_{SMOW}} \right) - 1 \right] 10^3 = \delta D \text{‰/SMOW}$$

The overall reproducibility of the $\delta D \text{ H}_2\text{O}^+$ is estimated at $\pm 2 \text{‰}$. The relative uncertainty on the water concentration is equal to 2 %; this error increases because of weighing errors for very small samples.

II - Results.

Water and isotopic concentration of H_2O^+ are reported in fig.1; it shows a correlation between the water, the deuterium concentrations and the petrologic types of chondrites.

Figure 1.

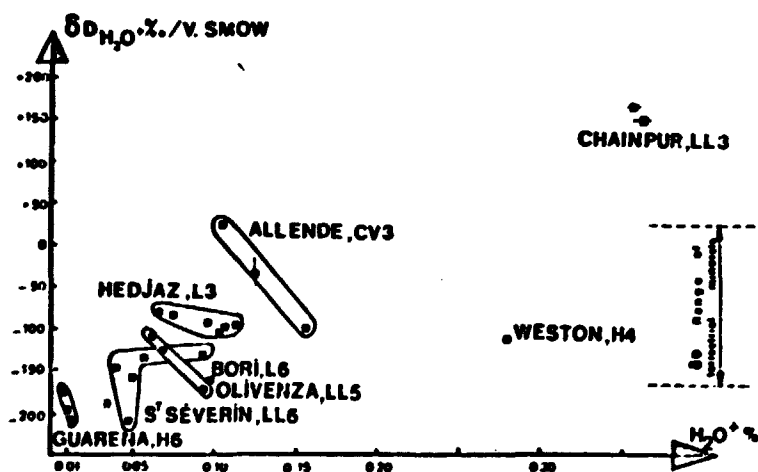
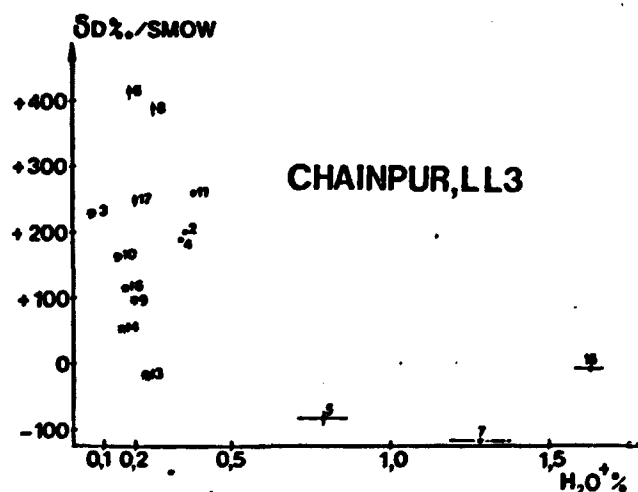
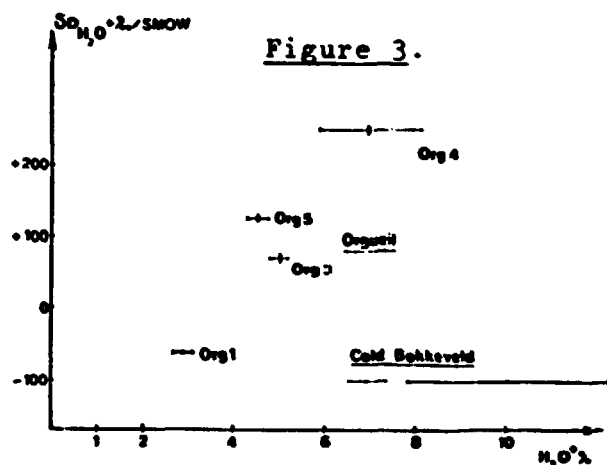


Figure 2.



The Chainpur meteorite has been divided into several mineralogical fractions and the results are reported in fig. 2. The data reported as a star corroborate the interpretation we previously made on the case of others chondrites [3]: the chondrite's matrix is water rich (sample nb.13) and deuterium poor as compared with chondrules (sample nb.3 is a single chondrule). The mineralogical fraction of density < 2.8 , extracted from the matrix, is represented by samples 5, 7 and 15. The grain size of sample 15 is lower than 10μ and water has been extracted together with a large amount of C-rich gaseous components. Samples 2 and 4 are duplicates; they approximately represent the whole rock. Samples 6 and 8 (duplicates) are anomalously enriched in deuterium but the deuterium rich mineral(s) has not been identified; they came from a $> 3.3 \text{ g cm}^{-3}$ mainly matrix fraction and do not represent more than a few percent of the whole rock.

The results of Orgueil are reported in fig. 3; the correlation shows that, as the sulphate content increases, the water and deuterium contents decrease.



III - Discussion.

1) Terrestrial contamination estimate.

Most of chondrites studied have δD 's similar to terrestrial rocks or atmospheric water vapor. However, all terrestrial processes suffered by terrestrial rocks give δD 's substantially lower than 0‰. The adsorption of terrestrial water vapor is a likely process which could blank out the extraterrestrial δD 's in the case of low δD meteorites. Experimental results seem to rule out such a contamination of H_2O^+ .

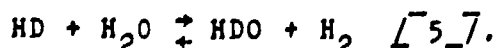
The H_2O^+ fractions of Chainpur have δD 's very far from any terrestrial rocks although they correspond to unshocked meteorite and low water contents. The $\delta D_{H_2O^-}$ of Chainpur varies from -150 to -50‰ and corresponds to terrestrial contamination. There is no correlation between $\delta D_{H_2O^-}$ and $\delta D_{H_2O^+}$ which varies from -130 to +420‰. For ordinary chondrites, the range of $\delta D_{H_2O^-}$ is never larger than that observed in Chainpur; hence it is likely that H_2O^+ must not be affected by terrestrial contamination. There is no relationship between the δD 's of the precipitations at the sites of the meteorite descent, and $\delta D_{H_2O^+}$.

2) Possible deuterium variation models.

At the time of condensation and accretion of the meteorite parent bodies, hydrogen in the form of H_2 or/and H_2O is bound into the rocks. The D/H ratio established during this period is called "primary". Secondary, processes such as the addition of protons by the solar wind, in situ deuterium growth by the galactic cosmic rays induced by spallation, or water loss during thermal metamorphism, can wipe out the primary isotopic variations.

None of these processes seems either to have taken place or to have affected the primordial isotopic ratio of the chondrites in such a large range as that of the measured δD 's: i) Some of them (Chainpur, Orgueil) have not been metamorphosed and this is apparent in the very large heterogeneities existing in these meteorites. ii) The exposure ages can only affect the $\delta D_{H_2O^+}$ by few permil [4] - iii) The gas rich Weston which is the more likely to have been modified, can be corrected by only 40‰ by taking into account the solar wind protons implanted with He.

On the contrary, isotopic exchange between water and hydrogen in the nebula can enrich water in deuterium very much by the exchange:



For $(D/H)_{H_2} = 1.9 \cdot 10^{-5}$, the temperature of the last equilibration between the nebula² and the water of Quareña is 225° K and 185° K in the case of the mean δD of Chainpur. These "isotopic temperatures" do not disagree with ice appearance temperatures, calculated on the basis of thermodynamic considerations [6]. These low temperatures may mean that, constituents condensed at a very low temperature, were incorporated into the rocks at the moment of the accretion. In this assumption, the increase of water concentration in chondrites, would be caused mainly, by the relative increase in compounds condensed at very low temperatures.

However, for Chainpur, high δD 's seem to be associated with high temperature minerals and low δD 's with low temperature minerals. This, would indicate at least for Chainpur an alternative explanation: a diffusion model with preferential loss of light hydrogen. In such a degassing model the isotopic fractionations would be much more important than these occurring in the nebula between gases or in the parent body between minerals and water.

[1] - D. YORK. Comments on Astrophys. Gordon - Breach Sc. Pub. Ltd, vol. 7, n° 1, p. 1-11 (1977)

[2] - G. BOATO. Geochim-Cosmochim. Acta, vol. 6, p. 209-220 (1954)

[3] - F. ROBERT, L. MERLIVAT, M. JAVOY. Meteoritics, vol. 12, n° 3, p. 349-354 (1977)

[4] - L. MERLIVAT, M. LELU, G. NIEF, E. ROTH, Proc. Lunar. Sci. Conf, 7th. p. 649-658 (1976)

[5] - J. GEISS, H. REEVES, Astron. Astrophys., 18, p. 126-132 (1972)

[6] - L. GROSSMAN, J.W. LARIMER, Reviews of Geophys. and space Physics, Vol. 12, n° 1, p. 71-101 (1974)

