

**OXYGEN ISOTOPIC CONTAMINATION DURING HF-HCl DISSOLUTION OF BULK CARBONACEOUS CHONDRITES; Jérôme Halbout, François Robert and Marc Javoy, Laboratoire de Géochimie des Isotopes Stables, IPGP & Université Paris VII, 75251 PARIS CEDEX 05, FRANCE.**

Carbonaceous chondrite acid-resistant residues (ARs) have been shown to display isotopic compositions of the light elements far outside the terrestrial range. From the magnitude of the variations observed for hydrogen and carbon, it has been argued that the kerogen which is the main constituent of these ARs could contain traces of presolar compounds. Oxygen however was found to have a close-to-terrestrial isotopic composition in carbonaceous chondrite ARs [1]. Since the solar system has an oxygen isotopic signature different from that of interstellar clouds, this shows that the proportion of hypothetical non-solar oxygen cannot exceed a few parts in ten thousand in these ARs [2]. However, preparation of ARs from bulk meteorites causes an isotopic contamination of the samples by the solvents. Particularly sensitive to this contamination are the isotopic compositions of hydrogen and oxygen, as shown in [3] and [1], respectively.

Robert *et al.* [4] reported a comprehensive study of the hydrogen isotopic contamination in ARs from CI Orgueil. They determined the proportion of hydrogen imputable to contamination at different pyrolysis temperatures, and found that it reached a maximum of 14.2% in the room-temperature to 200°C pyrolysis step. They could calculate the isotopic composition of the D-rich meteoritic end-member:  $\delta D = +1360$  permil/SMOW. But their experimental results also showed that this end-member itself resulted possibly from the mixing of two indigenous hydrogen components, one of which having a  $\delta D$  up to 6700 permil.

We report here a similar study of the oxygen isotopic contamination in Orgueil ARs. Four sets of H<sub>2</sub>O, HCl-H<sub>2</sub>O and HF-HCl-H<sub>2</sub>O solutions were prepared, three of which were labeled in <sup>18</sup>O. The isotopic composition of each set of solutions was determined by CO<sub>2</sub> equilibration [5];  $\delta^{18}O$  values ranged from -4.4 to +125.5, and  $\delta^{17}O$  from -2.3 to +30.7. Three acid residues were prepared with each set of solutions so that all ARs were subjected to a known isotopic contamination throughout their preparation. The dissolution protocol was the same as in [4] for all ARs, but the duration of each acidification step was slightly extended for the ARs labeled "2" compared to those labeled "1". Residues were never allowed to dry out during their preparation, until they were dried and stored in an argon atmosphere prior to oxygen extraction. Thus, none of the residues was ever exposed to air for more than a few minutes. Oxygen was extracted by overnight reaction with BrF<sub>5</sub> at 750°C after a five hours preliminary outgassing [1]. One of the three ARs prepared with each set of solutions was outgassed at 750°C, and the two others at 430°C. This differential pyrolysis permits to determine how much contamination has *not* been removed at two different temperatures, chosen after the temperature of pyrolysis of indigenous components characterized by their hydrogen isotopic signature [6].

Fig.1 shows the relation between the isotopic composition of each AR and that of the solutions used throughout its preparation. ARs prepared following the same protocol with different labeled sets of solutions and outgassed identically define correlation lines that attest of the consistency of the experimental results. From the slopes of these correlation lines, it is possible to deduce that AR430,2 (residue that was exposed the longer time to the solvents, and outgassed at 430°C only) recorded a maximum contamination representing 32% of the oxygen extracted, in perfect agreement with our previous estimation [1].

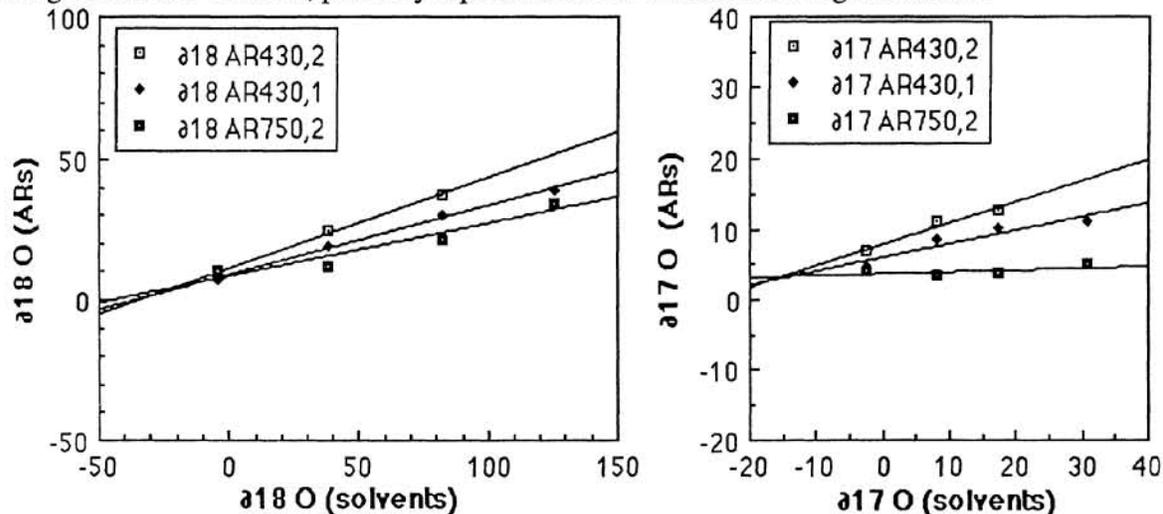
In each of the plots in fig.1, the correlation lines intersect at the same point. Both of these points lie out of the first bissector of each of the plots. One can show mathematically that this implies that the AR oxygen affected by the contamination is not a pure phase: two distinct meteoritic oxygen carriers must be invoked. The unicity of the intersection point in each plot shows that these carriers released their oxygen in the same mixing ratio in all the cases encountered in this study. This is also attested by the perfect correlation between the values of the slopes and of the zero-intercepts of the correlation lines in fig.1. Since the mixing ratio between the meteoritic oxygen carriers cannot be revealed by these experiments, only the oxygen isotope compositions of the bulk of the ARs (contamination excluded) and of one of the pure end-members can be deduced simply: ( $\delta^{18}O$ ,  $\delta^{17}O$ ) values are (3.7, 3.3) for the bulk Orgueil ARs, and (-23.6, -15.4) for the end-member oxygen carrier. The latter composition could represent that of a refractory phase in the Orgueil meteorite.

## OXYGEN ISOTOPES IN ORGUEIL ACID RESIDUES

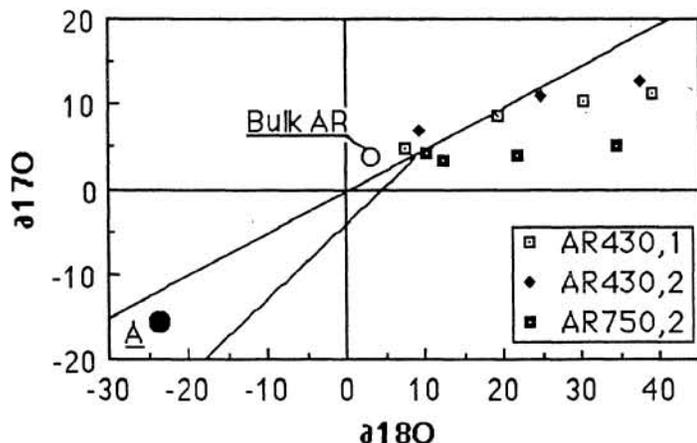
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Fig.2 features the results (experimental and calculated) of this study in the three-isotope plot. Each symbol represents the different contaminations of residues having undergone the same outgassing and the same dissolution schedule. They do not define straight lines, contrary to simple mixing processes. This shows that the contamination involves also isotopic fractionation processes.

As in the case of the hydrogen study [4], the isotopic composition obtained for the bulk ARs is characteristic of residues prepared following this protocol. Changing the dissolution protocol might change the unknown mixing ratio and give a hold on the isotopic composition of the second indigenous end-member, probably representative of the insoluble organic matter.



**Figure 1:** Oxygen isotopic composition of Orgueil acid resistant residues (ARs) against the composition of the solvents used throughout their preparation.



**Figure 2:** Isotopic composition of Orgueil acid residues prepared with labeled reagents. The white dot "Bulk AR" represents the composition calculated when the contamination by the solvents is subtracted. The black dot "A" corresponds to the composition calculated for one pure meteoritic component, in a model where the "bulk AR" results from the mixing of two end-members.

Acknowledgements to CNRS (ATP "Physico-chimie des molécules interstellaires") and INSU (ATP "Planétologie").  
References: [1] Halbout J., Robert F. and Javoy M. (1986) *Geochim. Cosmochim. Acta* **50**, 1599-1609. [2] Halbout J., Robert F. and Javoy M. (1988) *Geochim. Cosmochim. Acta* **52**, (in press). [3] Kerridge J.F., Shipp R. and Chang S. (1985) *Lunar Planet. Sci.* **XVI**, 432-433. [4] Robert F., Halbout J. and Javoy M. (1987) *Meteoritics* **22**, (in press). [5] Epstein S. (1980) *Lunar Planet. Sci.* **XI**, 259-261. [6] Robert F. and Epstein S. (1982) *Geochim. Cosmochim. Acta* **46**, 81-95.