

# Gravitational separation in polar firn. Response to H CRAIG.

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Schwander

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- J. Schwander, J.-M. Barnola, C. Andrie, M. Leuenberger, A. Ludin, D. Raynaud, B. Stauffer, J. Geophys. Res. 98, 2831 (1993).
- H. Craig, Y. Horibe, T. Sowers, *Science* 242, 1675 (1988); carrier diffusion [H. Craig, *ibid.* 159, 93 (1968)] is another process that may be responsible for fractionation effects.
- T. Sowers, M. Bender, D. Raynaud, J. Geophys. Res. 94, 5137 (1989).
- 5. Note however, that the elemental enrichment order deduced by Sowers *et al.* from their data [(4), p. 5147] is that air gases are lost in the order Ar, O<sub>2</sub>, N<sub>2</sub>, which is stated to be that predicted from molecular diameters, following (3). This is an error: their data actually show that the order of gas loss in their samples is O<sub>2</sub>, Ar, N<sub>2</sub>, as predicted and observed (3).
- H. Craig, *Eos* 69, 1211 (1988); \_\_\_\_\_, D. Burtner, R. C. Wiens, *ibid.* 70, 1151 (1989).

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Response: Craig discusses the  $O_2/N_2$  ratios shown by Schwander *et al.* (1), but it was not stated in that paper that these ratios illustrated gravitational fractionation. Furthermore, in our *Science* article (2), we discussed exclusively the ice record of greenhouse gases (especially  $CO_2$  and  $CH_4$ ); our reference to Schwander *et al.* (1) concerned the  $CO_2$  and  $CH_4$  results of that paper.

Gases will be gravitationally fractionated only if the ratio of the diffusivity (D) to the convection velocity (w) is larger than the firn thickness; indeed they must then be fractionated. This condition is satisfied, at least for the Greenland site of Summit (1). and so gases in the firn are in fact enriched in heavier elements and isotopes because of gravitation. This argument was first made by Craig et al. [note 14 in (3)], who pointed out that the assumption was first made that "gases ... continuously readjust to the time-independent equilibrium state . . . is approximately true." The dominance of the diffusivity is also clearly indicated by the <sup>85</sup>Kr, CO<sub>2</sub>, and CH<sub>4</sub> data measured in the firn at Summit (1). These results could not be explained if the diffusive equilibrium were not closely fulfilled; effusion at Summit obviously plays a minor role, but additional effects on gas fractionation are not ruled out.

Craig predicts that firn air enrichments in <sup>18</sup>O will differ by up to 0.15 per mil depending on whether fractionation in the firn results from effusion or gravitational fractionation. Analytical precision allows one to look at such a difference. Craig *et al.* (3), for example, noted that the maximum  $\delta^{15}$ N and  $\delta^{18}$ O enrichments for the D57 ice core are predicted exactly from the firn depths and temperatures for gravitational equilibrium. We note that the  $\delta^{18}$ O values would need to be shifted by 0.1 per mil to be explained by effusion, when the experimental accuracy is 0.04 per mil.

On the basis of such observations, Craig et al. wrote (3, p. 1678)

Thus, 60 years after publication of the canonical theorem of Gibbs, a fluid system at the earth's surface—atmospheric gases in the firn layers of polar ice caps—has provided experimental verification [italics ours] of this effect . . .

namely, gravitational fractionation.

Finally, Craig's <sup>84</sup>Kr/<sup>36</sup>Ar measurements appear to confirm the role of gravitational fractionation.

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- Geophys. Res. 98D, 2831 (1993). □ 2. D. Raynaud, J. Jouzel, J. M. Barnola, J. Chappel laz, R. J. Delmas, C. Lorius, *Science* 259, 926 (1993); *ibid.* 260, 1411 (1993).
- (1995), *ibid.* 200, 1411 (1995). 3. H. Craig, Y. Horibe, T. Sowers, *ibid.* 242, 1675 (1988).
- 4. We thank M. Bender for useful discussions.

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### **Response:** Gravitational Separation in Polar Firn

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