

(Non-?) Traditional Hg stable isotope geochemistry in the early 1920's

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In 1919 Sir Francis Aston discovered the multiple stable isotopes of mercury (Hg) using his mass spectrograph. Only one year later Johannes N Brønsted and György de Hevesy used high precision density measurements to prove that liquid Hg distillation into a vacuum fractionates Hg isotopes as a function of isotope mass. In more detailed studies during the 1920's Brønsted and colleagues managed to produce Hg vapors and residual liquids that were heavily fractionated by up to -84‰ and +74‰ on the $\delta^{202}\text{Hg}$ scale. They derived the kinetic isotope fractionation law, and used Rayleigh equations and diagrams to estimate Hg isotope separation factors. They also explored the natural variations in Hg isotope abundances, by converting cinnabar (HgS) into liquid Hg and measuring its density. By sheer coincidence they did not find any density variations for nine global cinnabar deposits, within their measurement uncertainty of 0.4 - 1.2 ‰ on the $\delta^{202}\text{Hg}$ scale. Today we know that the $\delta^{202}\text{Hg}$ of cinnabar may vary by up to 5 ‰ across a single cinnabar deposit.

In 2006 Jean Carignan was interested in the potential isotopic fractionation of volatile metals under natural and industrial conditions. He proposed to examine the distillation of liquid Hg in a vacuum, without knowing that nearly one century before him scientists had looked at the issue. Using MC-ICPMS, we made nearly identical observations as Brønsted and co-workers: a kinetic isotope fractionation factor of 1.0067 for the $^{202}\text{Hg}/^{198}\text{Hg}$ pair (Estrade *et al.*, 2009). We also performed Hg isotope analysis of Hg vapor that is in equilibrium with liquid Hg. Here we observed deviations from theoretical mass dependency for the two odd Hg isotopes, ^{199}Hg and ^{201}Hg , in what has become the first experimental evidence for nuclear volume fractionation of Hg isotopes. This contribution will look back on both the early and more recent measurements.

[1] Estrade, N., Carignan, J., Sonke, J.E., Donard, O.F.X., 2009. Mercury isotope fractionation during liquid-vapor evaporation experiments. *Geochimica et Cosmochimica Acta*, 73: 2693-2711.

Satellite and aircraft views of relationships between particles, cloud water, and rain water

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A poorly characterized process in warm clouds is the conversion of cloud water to rain water especially with regard to the rate by which this complex process occurs. Using a satellite remote sensing data set, this conversion process is examined in a global sense over oceans to identify regional differences and relationships with relevant environmental parameters [1]. We show that a faster conversion process coincides with conditions of reduced atmospheric stability, higher low-level wind speeds, and low aerosol index values. Aircraft measurements are used in a stratocumulus cloud environment to provide more views on relationships between environmental factors influencing water in clouds [2].

[1] Sorooshian, Wang, Feingold & L'Ecuyer (2013), A satellite perspective on cloud water to rain water conversion rates and relationships with aerosol types and atmospheric stability. *Geophys. Res. Atmos.*, 118, doi: 10.1002/jgrd.50523

[2] Sorooshian, Wang, Coggon, Jonsson, Ervens (2013), Observations of sharp oxalate reductions in stratocumulus clouds at variable altitudes: organic acid and metal measurements during the 2011 E-PEACE campaign. *Environ. Sci. Technol.*, doi: 10.1021/es4012383.