Reduced COHN volatile speciation and solubility in basaltic melt: Terrestrial volcanism, atmospheric evolution and deep C cycling

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Partitioning of reduced C-O-H-N volatiles between magmas, atmospheres, solid mantles and core-forming metal influences early climatic conditions and deep volatile cycles on the Earth, Moon, and Mars. There is a poor consensus on the identity and concentrations of reduced volatiles in natural magmas, as well as their dependencies on T, P, and fO_2. We determine the speciation and solubility of C-O-H-N volatiles in terrestrial and martian basalts under C-saturated, H_2O-poor conditions at IW-3 to IW+1.4 in experiments at 1.2 GPa and 1400° C. Speciation was determined by vibrational spectroscopy and C and H concentrations by SIMS. Depending on fO_2 and melt H-contents, dissolved species can include OH^-, CO_3^{2-}, H_2, CH_4, and both Fe-carbonyl-like and C-O-H-N-species. CH_4 is relevant only with >300 ppm dissolved H and fO_2<IW. At fO_2 relevant to a post-core formation terrestrial magma ocean (MO), <5 ppm C is soluble in silicate, making a MO both easily C-saturated and a poor host for C. For C, we measure $D_{carbide/melt}$ of $\sim 10^4$, suggesting that C strongly partitions into any metal present. Surprisingly, at these reduced, high-P conditions all C in melt is contained in a complex molecule containing both N-H and C=O bonds, possibly an amide. The solubility of this species relative to overlying atmospheres remains poorly characterized. CO-bearing species similar to Fe-carbonyls are dissolved at intermediate fO_2 relevant to volcanic degassing on Mars but probably not a terrestrial magma ocean or the more reduced regions of the Moon.