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N, C and Ar isotopes of fluid inclusions from 3.5 Ga hydrothermal system: Constraints on early mantle and atmosphere

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Nitrogen isotopic record of atmosphere and mantle reflects history of atmosphere-crust-mantle interaction. However, there is no consensus on N istope ratio of paleoatomosphere [1, 2]. In addition, N isotopic evolution of ancient-mantle is not well documented.

Here, we report N, C and Ar isotope ratios of fluid inclusions in agate associated with silica dikes from 3.5 Ga North Pole area in the Pilbara craton, Western Australia. Because the silica dikes are suggested to be remnant of seafloor hydrothermal systems at 3.5 Ga [3], it is possible that fluid inclusions contain air and mantle components at that time. Raman microspectrometry identified H_2O, CO_2, N_2, CH_4 in the inclusions. Crushing experiments showed that $\delta^{15}N$ values, $N_2/^{36}Ar$ ratios, $^{40}Ar/^{36}Ar$ ratios and $\delta^{13}C$ values for the inclusions are –5.9 ~ +0.8‰, 22000 ~ 99100, 1010 ~ 3810 and –3.9 ~ +0.5‰, respectively. Correlation between the $\delta^{15}N$ values and $N_2/^{36}Ar$ ratios suggests that N and ^{36}Ar in the inclusions can be explained by mixing of two components (i.e. contemporary upper mantle with $\delta^{15}N \leq -5.9‰$ and $N_2/^{36}Ar \geq 99100$ and air with $\delta^{15}N \geq +0.8‰$ and $N_2/^{36}Ar \leq 22000$). On the other hand, correlation between the $\delta^{15}N$ values and $^{40}Ar/^{36}Ar$ ratios suggests that fluids with $\delta^{15}N \geq -2‰$ have various ratios of $^{40}Ar/^{36}Ar$ of 1340 ~ 3810 while those with $\delta^{15}N \leq -2‰$ have raletively constant ratios of $^{40}Ar/^{36}Ar$ of 1010~1180. Definitive explanation for the variation of $^{40}Ar/^{36}Ar$ ratios are not yet given, because the variation can be produced by many factor such as mixing of varoius components (i.e., upper mantle, air and components with excess Ar) and in situ radioactive production of ^{40}Ar. However, measurement of K content of agate powder after crushing suggests that K in agate lattice can not make the observed variation of $^{40}Ar/^{36}Ar$ ratios.

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Prebiotic methane abundance in the hydrogen/CO_2-rich atmosphere

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Decreased solar luminosity and multiple lines of geologic evidence in favor of a "liquid" ocean on the ancient Earth set a puzzle known as the "Faint Young Sun" paradox. For several decades, elevated atmospheric CO_2 levels were considered to be the most self-consistent solution for the warm early Archean/Hadean climate [1]. However, to offset a ~25% decreased solar luminosity (at ~3.5 Gyr ago) and keep the mean global surface temperature at ~288K, CO_2 should have been at a steady-state concentration of about 0.3 bars. At such high levels, CO_2 would condense in the Earth's polar regions (as it does on Mars today) and no longer could be considered as the only "stabilizer" of the early Archean/Hadean climate. Climate simulations [2] show that 100-1000 ppm of methane would be sufficient to maintain warm climate under decreased solar luminosity without invoking huge CO_2 levels.

The major argument against methane in the prebiotic atmosphere is the lack of substantial methane volcanic flux even in the early Earth history because of the early mantle differentiation.

However, previous calculations assumed a high ("diffusion-limited") rate of hydrogen loss to space. If atmosphere was anoxic, hydrogen should have been lost at a much (~100 times) slower rate [3]. In hydrogen-rich atmospheres CH_4 molecules could have been effectively "recycled" after initial photolysis. Additionally, high CO_2 levels (if present) would partially shield CH_4 molecules from UV photolysis allowing further CH_4 build up. Here we demonstrate that 100-1000 ppm could be maintained with a much smaller methane flux in the hydrogen/CO_2-rich atmospheres.

We conclude that methane should have been abundant even in the prebiotic environment and most likely was responsible for the lack of irreversible glaciations early in Earth history

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