B102-P001 **Room: Poster Session Hall** **Time: May 22**

Sulfur isotope analysis in 3.0Ga Pongola Supergroup, South Africa

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The composition of Earth's early atmosphere is constrained by sulfur isotope ratios of mineral samples in various dated rocks. Sulfur mass-independent-fractionation (MIF) is observed only in geological records older than ˜2.0 Ga, but not in younger strata (e.g., Faquhar and Wing 2003). The MIF is a unique isotope fractionation which is caused by photochemical reactions of sulfurous gas under low oxygen level. Hence, pre-2.0-Ga atmosphere is thought to have been oxygen-poor. However, recent investigations reported that large signature of MIF would have been absent between 3.0 and 2.8 Ga (Ohomato et al., 2006; Ono et al., 2006a; Kasting and Ono 2006). This implies the possibility that the oxygen content of the atmosphere once increased, and then got back to the reduced condition again in this period (Ono et al., 2006a).

However, if the MIF signal is small (lower than 0.3 permil D33S), it is difficult to distinguish MIF from MDF (Mass-Dependent-Fractionation), because MDF also creates such small variations of D33S values (Farquhar et al., 2003; Ono et al., 2006b). In order to distinguish the small MIF and MDF, we performed high-precision multiple sulfur isotope analysis including 36S. Moreover, the samples analyzed by previous studies are limited mostly in shale samples. Other various types of lithofacies may potentially have larger MIF signals. Therefore, we analyzed other types of rocks including stromatolitic carbonate, sandstone, and conglomerate in different depositional settings from Pongola Supergroup (2985-2837Ma), South Africa.

The D33S values of pyrites are from -0.21 to 0.30 permil, which are similar to those reported by previous studies. This means that small or no MIF (within the range of +/-0.5 permil D33S) is a widespread character in this period than previously thought. Furthermore, as a result of investigating D33S/D36S relationships, it was found that 4 samples can be explained only by MDF processes, but at least 3 samples, which included in 2 clastic rocks of Mozaan Group and 1 carbonate rock of Nsuze Group, show characteristics of MIF produced by photolysis of SO2 at 193nm (D33S/D36S = -1; Farquhar et al., 2001), despite of their small D33S values. This indicates that similar photochemical processes were operated in the 2985-2837Ma atmosphere like other Archean ages. The observed small D33S values regardless of different depositional settings probably imply that the oxygen content of the atmosphere was in a transition from 10-5 to 10-2 PAL. This is consistent with a hypothesis that the increase of oxygen level caused drop of methane concentration, triggering the so-called Pongola glaciation (Young, 1998; Ono et al., 2006b).