

When did the Great Oxidation Event begin?

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A defining hallmark of Earth's habitability is its richly oxygenated surface. Our planet's transition from an anoxic to a permanently oxygenated world, known as the Great Oxidation Event (GOE), indelibly transformed Earth's biosphere, eventually enabling the origin of more complex modes of life. Yet, considerable uncertainty remains regarding the timing and tempo of oxygenation during the GOE, particularly its onset.

The GOE has come to be defined by the disappearance of mass-independent fractionations of sulfur isotopes (S-MIF; non-zero $\Delta^{33}\text{S}$ values) from the sedimentary record. An unintended consequence of this framework is that the end of the GOE has received far more attention than its beginning. Here, we shift the focus to the often-overlooked start of the GOE. With the *first* O_2 -induced perturbation of S-MIF production and/or preservation as the defining criterion, when does the GOE commence?

Using a compilation of existing S-MIF data, we find significant evidence for changes to the global sulfur cycle beginning at ~ 2.5 Ga. The earliest recorded loss of S-MIF appears in multiple cratons shortly after this time. These S-MIF disruptions closely follow the strongest case to date for pre-GOE O_2 , recorded in the Mount McRae Shale from Western Australia (2.495 ± 0.014 Ga). In fact, a closer examination of this "whiff" of O_2 reveals that S-MIF attenuation occurs within the Mount McRae Shale itself. High molybdenum and rhenium abundances—the seminal evidence for enhanced oxidative weathering during the "whiff" interval—are invariably associated with muted $\Delta^{33}\text{S}$ values. This suggests that the ~ 2.5 Ga whiff of oxygen may mark the initiation of the GOE, as defined by S-MIF. Corroborating this, a compilation of molybdenum abundances in shales identifies ~ 2.5 Ga as the foremost statistical change point, suggesting a fundamental shift in the nature of oxidative weathering in the wake of the whiff. These findings indicate that the well-studied ~ 2.5 Ga pulse of O_2 may be more significant than once thought, marking the beginning of Earth's formal transition to a more oxic world.