

Supplementary Material:

SML collection:

SML-CA samples were collected off a small boat (whaler) with an outboard motor in Patricia Bay, Canada. It was sunny and cool (about 10 °C), with winds of about 3.5-4.0 m/s. The sea-surface microlayer was extracted with a glass plate (8×10 inches) submerged in an orientation perpendicular to the sea surface and withdrawn at a rate of about 6 cm/s. The microlayer samples were scraped off the glass plate with a neoprene squeegee and into 100-mL HDPE bottles. Fewer than 10 dips were required to fill each bottle.

SML-EqPOS samples were collected at the stations #5 (0°N 115°W; February 3, 2012, T_{air}: 24.7 °C, Average wind speed: 1.1 m/s) and #10 (0°N 140°W; February 13, 2012, T_{air}: 23.6 °C, Average wind speed: 4.4 m/s) along the eastern equatorial Pacific Ocean during the Equatorial Pacific Ocean and Stratospheric/Tropospheric Atmosphere Study (EqPOS) cruise by R/V Hakuho Maru. The sampling operations were conducted proximity 500 m off the research vessel during daytime using a PMMA rotating drum SML sampler operated at 10 rpm rotating speed. Collected SML samples were then transferred to HDPE bottles for storage and kept frozen at -18 °C until analysis. Estimated sampled SML thickness was 33 and 41 μm for those collected at the stations #5 and #10, respectively.

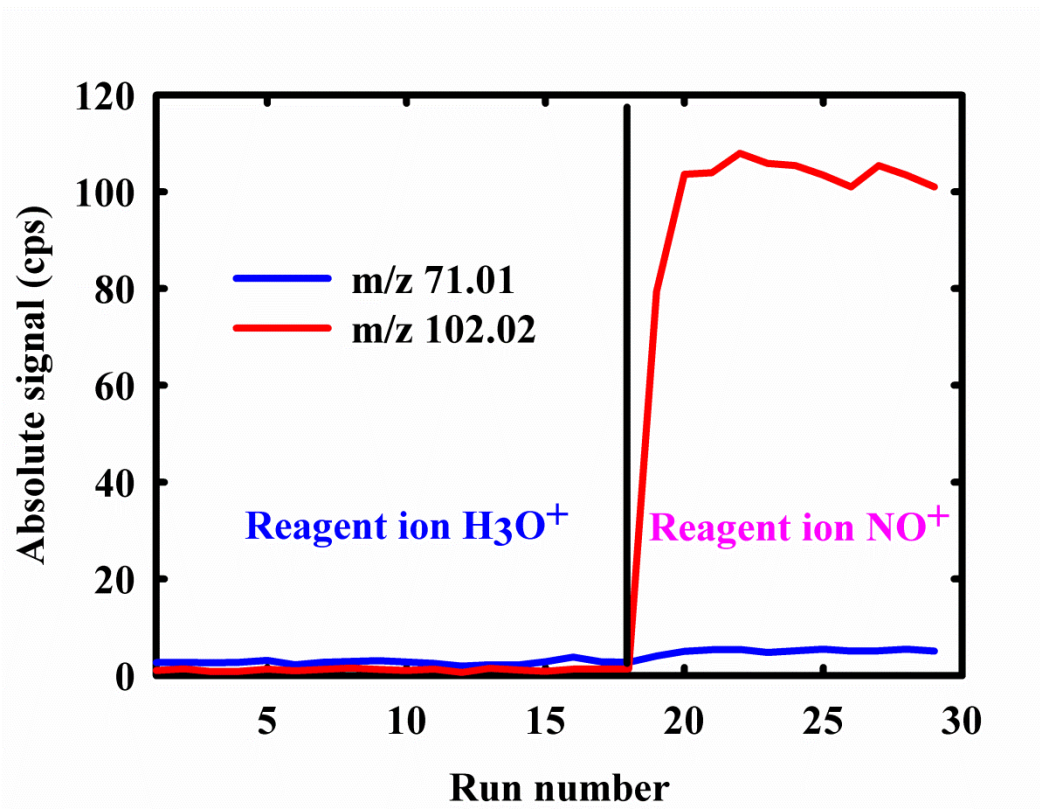


Figure S1. PTR-TOF-MS mass spectrum of the signals at m/z 71.01 and 102.02 from methyl glyoxal recorded with H₃O⁺ and NO⁺ as the reagent ions.