Supporting Information (SI)

Table 1. Carbonate contents and isotopic composition of the aerosol samples collected at four different sites: Mount Soledad (MSD); Chemistry Building at Univ. of California San Diego (BCH); Scripps Institute of Oceanography Research Pier (SIO), La Jolla, California and White Mountain Research Station (WMRS), California, USA. Here sample names ending in A denotes coarse fraction (> 1μm) and B denotes fine fraction (< 1.0μm). Collection date is expressed as dd-mm-yy. Standard deviations in the table are 1σ SD for replicate mass spectrometric analysis. Overall reproducibility for acid digestion, flourination and gas chromatography with laboratory standards (n = 5) is ± 0.3‰ for ^{17}O, ^{18}O and ± 0.1‰ for $\Delta^{17}O$. Here $\delta\,^{i}O' = 10^3 \ln(1+\delta\,^{i}O/10^3)$ with $^{i}O = {}^{17}O$, ^{18}O and $\Delta^{17}O = \delta\,^{17}O' - 0.524\,\delta\,^{18}O'$

Sample	CO_3^{-2} ($\mu g/cm^{-3}$)	$\delta\,^{17}O'$ (‰)	$\delta\,^{18}O'$ (‰)	$\Delta^{17}O$ (‰)
MSD160107A	3.92	16.57 ± 0 .04	24.95 ± 0.05	3.49
MSD160107B	1.49	18.14 ± 0.03	30.66 ± 0.02	2.07
BCH151106A	3.40	12.79 ± 0.05	18.78 ±0.02	2.95
BCH151106B	2.46	16.18 ± 0.07	25.28± 0.03	2.93
SIO270407A	2.37	17.43 ± 0.06	31.62 ± 0.04	0.86
SIO270407B	1.20	8.16 ± 0.05	14.29 ±0.02	0.67
SIO250507A	6.47	15.85 ± 0.03	24.19 ± 0.03	3.16
SIO250507B	1.03	10.02 ± 0.05	17.41 ± 0.04	0.90
SIO260607A	2.82	16.53 ±0.06	24.15 ± 0.03	3.87
SIO260607B	1.08	7.41 ±0.02	12.92 ± 0.04	0.64
SIO160207B	4.38	14.58 ± 0.05	25.90 ± 0.02	1.01
SIO200407B	2.06	12.50 ± 0.06	23.18 ± 0.03	0.36
SIO040507A	2.11	13.64 ± 0.04	22.36 ± 0.03	1.92
SIO230307A	1.83	9.94 ± 0.03	17.53 ± 0.02	0.75
WMRS080807A	3.55	12.89 ± 0.04	23.01± 0.02	0.83
WMRS080807B	2.93	13.44 ±0.05	24.59± 0.03	0.55

Table 2. Carbonate contents and isotopic composition of various soil samples. Here $\delta\,^{i}O' = 10^3 \ln(1+\delta\,^{i}O/10^3)$ with $^{i}O = {}^{17}O$, ^{18}O and $\Delta^{17}O^{*} = \delta\,^{17}O' - 0.524\,\delta\,^{18}O'$

Sample	CO_3^{-2} (%)	$\delta\,^{17}O'$ (‰)	$\delta\,^{18}O'$ (‰)	$\Delta^{17}O^{*}$ (‰)
Arizona Test Dust	2.98	15.10 ± 0.03	29.05 ± 0.02	-0.12
Owen Lake Dust	3.04	18.15 ± 0.03	34.98 ± 0.03	-0.18
Black Rock Desert Dust Nevada	4.21	17.97 ± 0.04	34.65 ± 0.02	-0.18
YaDan GanSu China	4.42	16.38 + 0.02	31.71 ± 0.01	-0.23
Grand Canyon Red Soil	0.03	20.97 ± 0.03	40.24 ± 0.02	-0.11
Commercial cement sample	3.06	14.38 ± 0.04	27.67 ± 0.03	-0.11

Table 3. Enrichment in CO_2 produced after acid digestion of $CaCO_3$ (3.2 ± 0.02 mg) that has been reacted with O_3 in the presence of thin water film.

Sample	Reaction time (min)	H_2O (μmole)	O_3 (μ mole)	$\delta\,^{17}O'$ (‰)	$\delta\,^{18}O'$ (‰)	$\Delta^{17}O^{*}$ (‰)
*R3P3-C1	0.0	55.5	0	17.82 ± 0.15	34.12 ± 0.08	-0.05
*R3P58	30	55.5	27	20.86 ± 0.03	36.49 ± 0.02	1.73
*R3P61	60	55.5	27	22.28 ± 0.04	37.90 ± 0.03	2.42
*R3P60	1320	55.5	27	23.36 ± 0.1	39.15 ± 0.03	2.84
**R6P33	30	55.5	0	6.76 ± 0.02	13.03 ± 0.02	-0.06
**R3P42	30	55.5	25	9.84 ± 0.02	16.49 ± 0.03	1.19
**R3P47	30	55.5	50	12.21 ± 0.05	19.48 ± 0.03	2.00

O_3 concentration varied by ± 2 μmoles

*$CaCO_3$ powder derived from oolite (R. Clayton's lab).

** AR grade calcite powder obtained from Solvey.

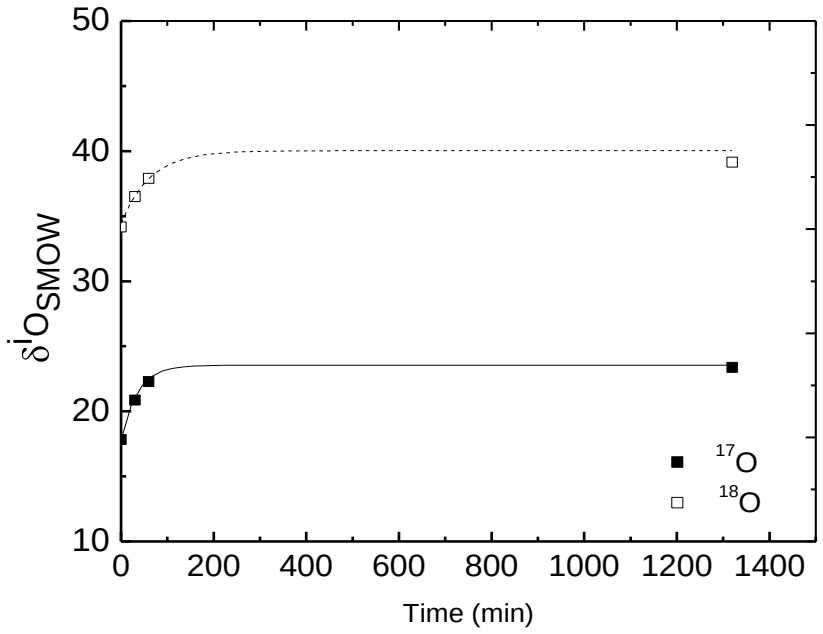

Fig. S1a. Kinetics of oxygen isotope enrichment in laboratory carbonates ($^{i}O = ^{17}O$ and ^{18}O). $CaCO_3$ (3.2 $\pm$ 0.02 mg) in the presence of ozone (27 $\pm$ 2 μmole with mean $\delta^{17}O' =$ 53 $\pm$ 2 and $\delta^{18}O' = 50 \pm 2$ ‰) and Millipore water (55.5 $\pm$ 0.5 μmoles).

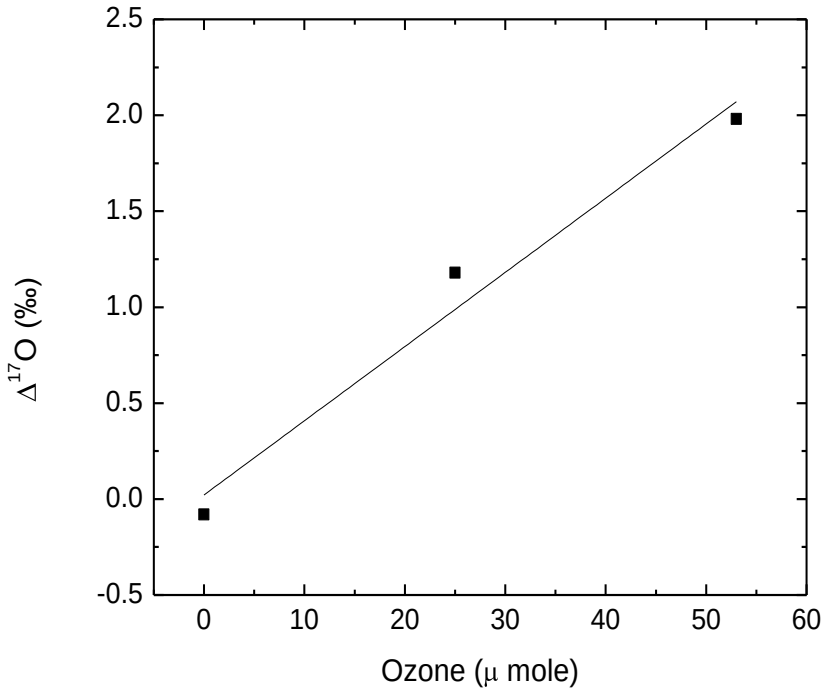

Fig. S1b. Anomalous oxygen isotopic enrichment in CO_2 produced from calcium carbonate after reaction with varying concentration of ozone for 30 minutes under following experimental conditions. $CaCO_3$ (3.2 $\pm$ 0.02 mg), ozone (0, 25 $\pm$ 2, 50 $\pm$ 2 μmole with mean $\delta^{17}O' = 53 \pm 2$ and $\delta^{18}O' = 50 \pm 2$ ‰) and Milli pore water (55.5 $\pm$ 0.5 μmoles).