

Table S1. Details of the observational data used for model comparison.

Sample location	Time period of sample collection	Time averaging	Nature of samples	Total number of samples	Analytical method	$\Delta^{17}\text{O}(\text{‰})$ range	Reference
Alert, Canada (82°N, 62°W)	April 2005 – July 2006	Monthly	Aerosol	100 ^a	Bacterial denitrification	24.6 – 32.9	(Morin et al., 2008)
Summit, Greenland (73°N, 39°W)	July 2003 – January 2006	Monthly	Snowpit	17 ^a	AgNO ₃ pyrolysis	23.5 – 32.6	(Kunasek et al., 2008)
Princeton, NJ, USA (40°N, 75°W)	November 2002- December 2003	Monthly	Event-based precipitation	66 ^a	Bacterial denitrification	22.4 – 27.1	(Kaiser et al., 2007)
La Jolla, CA, USA (33°N, 117°W)	March 1997 – February 1998	Monthly	Aerosol	41 ^a	AgNO ₃ pyrolysis	22.2 – 30.5	(Michalski et al., 2003)
Bermuda (32°N, 65°W)	January and August 2001	Monthly	Event-based precipitation	4 ^a	Bacterial denitrification	23.0 – 26.8	(Hastings et al., 2003)
COCA cruise (21-29°N, 16-26°W)	May – June 2003	Daily	Aerosol	16	AgNO ₃ pyrolysis	22.1 – 27.8	(B. Alexander, unpublished data)
Atlantic cruise (28°S-52°N, 21°W-12°E)	April – May 2007	Daily	Aerosol	22	Bacterial denitrification	23.9 – 31.0	(Morin et al., 2009)
PNF, Ecuador (4°S, 79°W)	October 2004 – April 2006	Multi-annual	Fog water	30 ^b	AgNO ₃ pyrolysis	17.7	(Brothers et al., 2008)
Yungay, Chile (24°S, 70°W)	2003 - 2004 and 2006	Multi-annual	Passive deposition (wet + dry)	3	AgNO ₃ pyrolysis	23.0	(Ewing et al., 2007)
DDU, Antarctica (66°S, 140°E)	January – December 2001	Monthly	Aerosol	30 ^a	Bacterial denitrification	23.5 – 38.9	(Savarino et al., 2007)
South Pole (90°S, 0°E)	December 2003 – December 2004	Monthly	Aerosol	15 ^a	AgNO ₃ pyrolysis	24.8 – 38.1	(McCabe et al., 2007)

^aConcentration-weighted average $\Delta^{17}\text{O}(\text{nitrate})$ over all samples collected during each month of the year

^bConcentration-weighted average $\Delta^{17}\text{O}(\text{nitrate})$ over all samples collected during the course of 3 years

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