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Stable isotopic evidence for the excess leaching of unprocessed atmospheric nitrate from forested catchments under high nitrogen saturation

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Table S1. Comparison of dry and wet deposition rates of atmospheric nitrate (kg N ha^{-1} yr^{-1}) among several studies and with references.

Site	Site type	Date	$D_{atm\text{-}bulk}$	$D_{atm\text{-}wet}$	$D_{atm\text{-}dry}$	$D_{atm\text{-}total}$	Reference
FK (Japan)	forest	2009-2018	4.8	4.2	5.6	9.7	this study
MY (Japan)	forest	2009-2018	3.4	3.1	2.6	5.6	this study
KJ (Japan)	forest	2012-2014	6.4	-	-	-	Nakagawa et al., 2018
IJ (Japan)	forest	2012-2014	-	5.9	0.3	6.2	Nakagawa et al., 2018
Fellow (USA)	forest	2010	-	-	-	3.3	Rose et al., 2015
Uryu (Japan)	forest	2008-2011	-	2.1	0.5	2.6	Tsunogai et al., 2014
Qingyuan (China)	forest	2014-2017	7.0	-	-	-	Huang et al., 2020
DPRN (USA)	forest	2016-2017	1.5	-	-	-	Bostic et al., 2021
Xishan (China)	forest	2010-2011	-	7.3	10.1	17.4	Shen et al., 2013
GEF (Japan)	5 forest sites	2005	7.0	-	-	-	Tobari et al., 2010
Chinese forest	31 forest sites	1980-2009	5.3	-	-	-	Fang et al., 2011
Europe forest	194 forest sites	1985-1995	-	5.5	-	-	Dise et al., 2009
DBW (China)	city	2006-2008	-	-	6.7	-	Shen et al., 2009
QZ (China)	city	2006-2008	-	-	7.7	-	Shen et al., 2009
CASTNET (USA)	8 mix sites	2004	-	3.1	1.9	5.0	Elliott et al., 2009
Japan	28 mix sites	2010-2017	-	3.9	2.3	6.2	ARNSR
USA	continent	1978-1994	-	1.6	1.5	3.2	Holland et al., 2005
Western Europe	continent	1978-1994	-	2.6	2.9	5.5	Holland et al., 2005

ARNSR (Acid Rain National Survey Report of Japan 2010-2017; https://tenbou.nies.go.jp/envgis_explain/acid_rain/content.html)

-No data

Table S2. The annual amount of precipitation, amount of precipitation, average temperature, flux of stream water (F_{stream}), and M_{atm}/D_{atm} ratio in the FK1, FK2, and MY, along with those in the catchments studied in past studies.

	Precipitation mm	Temperature °C	F_{stream} mm	M_{atm}/D_{atm} %
FK1	1777	15.9	902	14.1
FK2	1777	15.9	902	6.6
MY	3981	10.8	3266	1.3
KJ	2500	13.0	1276	9.4
IJ1	3310	13.0	2057	6.5
IJ2	3310	13.0	2057	2.6
Fernow1	1450	9.3	567.1	3.6
Fernow2	1450	9.3	449	6.3
Fernow3	1450	9.3	577.7	10.3
Uryu	1170	5.6	500	0.7
Qingyuan	709	4.5	309	5.8

Table S3. Stream nitrate concentrations, $\delta^{15}N$, $\delta^{18}O$, $\Delta^{17}O$ of stream nitrate, and $\delta^{18}O$, δD of stream water at FK1 catchments.

FK1	NO_3^- μM	$\delta^{15}N(NO_3^-)$ /10^3	$\delta^{18}O(NO_3^-)$ /10^3	$\Delta^{17}O(NO_3^-)$ /10^3	$\delta^{18}O(H_2O)$ /10^3	$\delta D(H_2O)$ /10^3
2019/11/1	121.3	-0.70	3.94	2.25	-6.63	-44.47
2019/11/29	114.1	-0.49	5.13	2.17	-7.12	-45.36
2019/12/25	112.2	-0.89	5.13	2.36	-7.34	-46.39
2020/1/31	105.1	-0.49	6.34	2.32	-7.36	-46.01
2020/3/2	107.9	-0.76	6.43	2.47	-7.36	-45.45
2020/4/2	102.6	-0.25	5.49	2.22	-7.42	-45.53
2020/7/1	111.6	-0.23	5.36	2.63	-7.63	-47.22
2020/7/31	111.6	-0.27	6.78	2.60	-7.82	-48.27
2020/9/1	113.8	0.51	6.67	2.35	-7.79	-48.24
2020/10/1	109.8	0.08	7.26	2.44	-7.70	-48.10
2020/11/4	107.8	-0.34	6.48	2.73	-7.65	-47.28
2020/12/1	103.4	0.35	7.61	2.61	-7.68	-47.41
2021/1/15	102.3	-0.68	6.70	3.06	-7.86	-47.66
2021/2/2	115.6	-0.37	6.11	3.01	-7.64	-45.07
2021/3/1	102.9	0.97	8.47	2.46	-7.89	-47.92
2021/3/31	97.5	0.31	6.75	2.61	-7.78	-47.26
2021/4/27	100.2	0.90	7.51	2.70	-7.87	-48.17
2021/6/1	103.8	0.49	5.75	2.56	-7.78	-47.29
2021/7/1	107.8	1.36	6.65	2.87	-7.93	-48.25
2021/8/2	118.8	1.46	4.52	2.02	-7.94	-48.61
2021/9/1	118.6	1.16	6.31	2.53	-7.97	-48.62
2021/10/1	114.5	0.51	6.43	2.57	-7.96	-48.84
2021/11/1	115.1	1.04	8.36	3.27	-7.93	-48.42
2021/12/1	113.3	-0.17	6.41	3.15	-7.95	-47.43
2021/12/24	105.5	0.59	6.96	2.99	-7.89	-48.22

Table S4. Stream nitrate concentrations, $\delta^{15}N$, $\delta^{18}O$, $\Delta^{17}O$ of stream nitrate, and $\delta^{18}O$, δD of stream water of the total FK catchments (FK1 +FK2).

FK1+FK2	NO_3^- μM	$\delta^{15}N(NO_3^-)$ /10^3	$\delta^{18}O(NO_3^-)$ /10^3	$\Delta^{17}O(NO_3^-)$ /10^3	$\delta^{18}O(H_2O)$ /10^3	$\delta D(H_2O)$ /10^3
2019/11/1	106.7	0.90	-0.32	1.71	-7.39	-47.39
2019/11/29	92.8	1.24	2.56	1.98	-7.10	-45.44
2019/12/25	96.8	0.26	2.21	1.90	-7.46	-47.48
2020/1/31	98.1	0.79	1.09	1.45	-7.44	-47.55
2020/3/2	97.0	0.72	0.34	1.66	-6.90	-44.37
2020/4/2	92.0	-1.16	-0.67	1.17	-7.38	-45.78
2020/7/1	96.9	-0.56	1.00	1.93	-7.62	-48.07
2020/7/31	73.9	1.33	3.63	1.91	-7.90	-50.03
2020/9/1	82.7	4.51	2.57	1.44	-7.76	-48.44
2020/10/1	85.2	1.97	2.05	1.71	-7.79	-48.70
2020/11/4	89.3	0.74	1.55	1.85	-7.68	-47.78
2020/12/1	80.3	-0.11	1.41	1.96	-7.67	-47.57
2021/1/15	97.5	0.37	3.49	2.11	-7.91	-47.29
2021/2/2	142.6	0.88	2.54	2.06	-7.51	-43.21
2021/3/1	92.2	0.94	2.56	2.06	-7.83	-47.11
2021/3/31	88.9	1.32	1.37	1.57	-7.68	-45.94
2021/4/27	80.8	0.82	1.43	1.26	-7.78	-47.24
2021/6/1	88.7	0.38	2.10	1.27	-7.83	-47.95
2021/7/1	90.4	1.54	1.08	1.47	-8.03	-49.59
2021/8/2	132.5	0.82	-0.41	0.80	-7.89	-49.31
2021/9/1	84.3	0.97	3.54	2.01	-8.17	-50.46
2021/10/1	83.7	1.63	2.28	1.65	-8.13	-49.77
2021/11/1	86.9	1.34	2.15	1.34	-7.97	-48.50
2021/12/1	112.9	0.44	2.66	2.37	-8.10	-48.39
2021/12/24	82.6	0.78	2.55	1.86	-7.92	-48.21

Table S5. Stream nitrate concentrations, $\delta^{15}N$, $\delta^{18}O$, $\Delta^{17}O$ of stream nitrate, and $\delta^{18}O$, δD of stream water at FK2 catchment only.

FK2	NO_3^- μM	$\delta^{15}N(NO_3^-)$ /10^3	$\delta^{18}O(NO_3^-)$ /10^3	$\Delta^{17}O(NO_3^-)$ /10^3	$\delta^{18}O(H_2O)$ /10^3	$\delta D(H_2O)$ /10^3
2019/11/1	103.5	1.30	-1.41	1.57	-7.58	-48.13
2019/11/29	88.2	1.73	1.83	1.93	-7.10	-45.46
2019/12/25	93.4	0.57	1.44	1.78	-7.50	-47.77
2020/1/31	96.6	1.10	-0.15	1.24	-7.46	-47.92
2020/3/2	94.6	1.08	-1.18	1.45	-6.78	-44.10
2020/4/2	89.8	-1.39	-2.20	0.92	-7.37	-45.84
2020/7/1	93.7	-0.64	-0.14	1.75	-7.62	-48.29
2020/7/31	65.7	1.92	2.46	1.66	-7.93	-50.68
2020/9/1	76.0	5.82	1.23	1.14	-7.75	-48.50
2020/10/1	79.9	2.53	0.49	1.49	-7.81	-48.88
2020/11/4	85.2	1.04	0.19	1.60	-7.69	-47.92
2020/12/1	75.2	-0.25	-0.44	1.76	-7.67	-47.62
2021/1/15	96.5	0.61	2.75	1.89	-7.93	-47.21
2021/2/2	148.5	1.09	1.93	1.90	-7.48	-42.89
2021/3/1	89.8	0.94	1.08	1.96	-7.82	-46.91
2021/3/31	87.1	1.57	0.05	1.31	-7.66	-45.62
2021/4/27	76.6	0.79	-0.31	0.85	-7.75	-46.97
2021/6/1	85.4	0.36	1.13	0.93	-7.84	-48.13
2021/7/1	86.7	1.59	-0.43	1.09	-8.06	-49.95
2021/8/2	135.4	0.70	-1.35	0.57	-7.88	-49.44
2021/9/1	76.8	0.90	2.61	1.83	-8.24	-51.08
2021/10/1	77.0	1.99	0.93	1.36	-8.18	-50.07
2021/11/1	80.7	1.44	0.22	0.74	-7.98	-48.53
2021/12/1	112.8	0.57	1.84	2.20	-8.13	-48.61
2021/12/24	77.7	0.84	1.24	1.53	-7.93	-48.21

Table S6. Stream nitrate concentrations, $\delta^{15}N$, $\delta^{18}O$, $\Delta^{17}O$ of stream nitrate, and $\delta^{18}O$, δD of stream water at MY catchments.

MY	NO_3^- μM	$\delta^{15}N(NO_3^-)$ /10^3	$\delta^{18}O(NO_3^-)$ /10^3	$\Delta^{17}O(NO_3^-)$ /10^3	$\delta^{18}O(H_2O)$ /10^3	$\delta D(H_2O)$ /10^3
2019/10/29	9.1	1.60	-3.78	0.39	-8.24	-54.96
2019/11/27	5.7	0.31	-4.37	0.44	-8.18	-53.89
2019/12/23	11.4	2.28	-1.15	0.95	-8.18	-53.04
2020/1/27	15.3	0.47	-0.80	0.38	-8.02	-52.31
2020/2/25	7.4	-0.24	-1.98	0.27	-8.15	-52.89
2020/3/26	4.9	0.45	-2.49	0.42	-8.40	-53.32
2020/6/29	11.2	0.52	1.65	0.99	-7.96	-50.27
2020/8/25	8.4	-0.02	-4.67	0.18	-8.62	-54.61
2020/9/28	5.9	2.42	-1.11	0.58	-8.77	-56.59
2020/10/27	5.3	0.55	-4.48	0.37	-8.54	-54.41
2020/11/25	4.9	0.41	-4.82	0.18	-8.47	-54.23
2020/12/21	6.3	-0.77	-0.64	0.53	-8.58	-54.76
2021/2/1	13.1	1.95	-1.37	0.92	-8.64	-53.39
2021/3/1	8.6	0.77	-1.57	0.95	-8.73	-53.63
2021/3/29	8.0	1.54	-1.66	0.92	-8.37	-51.02
2021/4/27	3.5	0.82	-3.25	0.40	-8.45	-52.51
2021/6/1	5.8	1.23	-0.87	0.52	-8.10	-49.53
2021/6/30	6.2	-0.68	-5.61	0.38	-8.49	-52.89
2021/7/30	5.9	-0.25	-5.14	0.29	-8.27	-51.26
2021/8/30	5.2	0.68	-4.59	0.50	-8.62	-55.08
2021/9/29	4.5	-0.33	-5.00	0.29	-8.63	-54.98
2021/10/28	4.9	-0.46	-5.01	0.19	-8.55	-54.21
2021/12/1	7.7	0.09	-2.71	0.71	-8.64	-53.94
2021/12/22	5.0	-0.29	-4.89	0.74	-8.69	-54.88

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