



## Supplement of

## Deposition, recycling, and archival of nitrate stable isotopes between the air–snow interface: comparison between Dronning Maud Land and Dome C, Antarctica

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Figure S1: (a) Shown are e-folding depths as a function of wavelength at two locations at Dronning Maud Land (DML; Pit A and B). Times in UTC indicate the median time of the experiment. The vertical solid lines mark the 315-325 nm range, where photolysis rate coefficients of NO<sub>3</sub><sup>-</sup> have their maximum (e.g. Fig. 5b in Frey et al. (2009). (b) Frontal view of a snow pit side wall showing precored horizontal holes in the top 27 cm of surface snowpack, where the fibre optics probe was inserted horizontally to measure down-welling irradiance. (c) Snow surface with a metal frame on top to keep the fibre optics probe at a 45° angle while it is pushed gradually into the snowpack to measure down-welling irradiance as a function of snow depth. The black cloth covers the spectrometer and a field laptop.



Figure S2: Modelled  $J(^{14/15}NO_3^{-})$  profiles in snow: base case at Dome C (dashed lines) and profiles scaled with various e-folding depths; shown as an example is the  $J(^{14/15}NO_3^{-})$  profile at a solar zenith angle (SZA) of 70° and total column ozone (TCO) of 300 DU typical for January at 75 °S on the East Antarctic Plateau. EDF: e-folding depth.



Figure S3: Dated snow pit profiles of nitrate, sodium, and non sea salt (nss)-sulphate mass concentration and  $\delta^{15}$ N-NO<sub>3</sub><sup>-</sup> at Dronning Maud Land (DML).



Figure S4: Nitrate mass concentration and  $\delta^{15}$ N-NO<sub>3</sub><sup>-</sup> composition in new deposition (diamond dust and hoar frost) and skin layer at Dome C during austral summer in 2007.





Figure S5: Seasonal wind rose at Kohnen Station. Data source: Utrecht University automatic weather Station at DML05/Kohnen (AWS9; https://www.projects.science.uu.nl/iceclimate/aws/files\_oper/oper\_20632TS10). Wind roses were made using the openair package in R following Carslaw (2014);Carslaw and Ropkins (2012).



Figure S6: Daily aerosol and skin layer ion mass concentrations at Dronning Maud Land (DML) for the month of January 2017. The shaded area of the skin layer mass concentrations reflects the spatial variability within 2500 m<sup>2</sup> area at the site.



Figure S7: Box and whisker plot comparing the atmospheric and terrestrial  $\delta^{15}$ N-NO<sub>3</sub><sup>-</sup> source signature to Dronning Maud Land (DML) atmospheric, skin layer, and snow pit  $\delta^{15}$ N-NO<sub>3</sub><sup>-</sup> signatures and Dome C snow pit  $\delta^{15}$ N-NO<sub>3</sub><sup>-</sup> signatures. Dome C data source: Frey et al. (2009).



Figure S8: Seasonal distribution of snowfall at Dronning Maud Land (DML), which has a bimodal distribution with the greatest accumulation in austral autumn and early austral summer. Data source: ERA Interim data, with monthly averages derived from 1980 to 2018 (https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era-interim).

Table S1: Nitrate blank mass concentrations for exposure blank, procedural blank and laboratory blank and detection limits. n: number of blanks.

Snow pit	n	NO <sub>3</sub> <sup>-</sup> mass concentration (ng g <sup>-1</sup> )	±
Instrumental blank	12	0.7	0.2
Exposure blank (Corning tubes)	12	6	3.0
Limit of detection (LOD)	7	3	
Surface snow	n	NO <sub>3</sub> <sup>-</sup> mass concentration (ng g <sup>-1</sup> )	±
Instrumental blank	15	5	1.2
Exposure blank (Whirl-pak® bags)	5	4	1.0
Limit of detection (LOD)	10	1	
Aerosol	n	NO <sub>3</sub> <sup>-</sup> mass concentration (ng g <sup>-1</sup> )	±
Laboratory blank	3	3	2.0
Procedure blank	4	280	78.9
Exposure blank	2	330	49.2
Limit of detection (LOD)	3	3	

Table S2: Atmospheric nitrate mass concentration and dry deposition fluxes over the ISOL-ICE campaign at Kohnen Station, Dronning Maud Land (DML). No data: n.d.

Sample name	Start time	End time	*Corrected volume (m <sup>3</sup> )	NO3 <sup>-</sup> mass concentration (ng m <sup>-3</sup> )	NO3 <sup>-</sup> dry deposition flux (pg m <sup>-2</sup> s <sup>-1</sup> )
Kohnen16-17_N_01	03/01/2017 09:33	04/01/2017 08:07	1651	12	99
Kohnen16-17_N_02	04/01/2017 08:28	05/01/2017 08:33	1815	10	84
Kohnen16-17_N_03	05/01/2017 09:14	06/01/2017 07:09	1613	17	137
Kohnen16-17_N_04	06/01/2017 07:29	07/01/2017 06:34	1719	19	152
Kohnen16-17_N_05	07/01/2017 06:55	08/01/2017 07:37	1872	7	54
Kohnen16-17_N_06	08/01/2017 07:56	09/01/2017 06:24	1661	8	66
Kohnen16-17_N_07	09/01/2017 06:40	10/01/2017 06:24	1722	8	66
Kohnen16-17_N_08	10/01/2017 06:40	11/01/2017 06:15	1890	6	45
Kohnen16-17_N_09	11/01/2017 06:34	12/01/2017 06:22	1784	9	72
Kohnen16-17_N_10	12/01/2017 06:39	13/01/2017 06:24	1724	13	106
Kohnen16-17_N_11	13/01/2017 06:44	14/01/2017 06:26	1736	10	79
Kohnen16-17_N_12	14/01/2017 06:47	15/01/2017 08:26	1811	8	67
Kohnen16-17_N_13	15/01/2017 08:45	16/01/2017 06:25	1574	14	112
Kohnen16-17_N_14	16/01/2017 06:44	17/01/2017 06:24	1754	8	63
Kohnen16-17_N_15	17/01/2017 06:42	18/01/2017 06:17	1717	4	31
Kohnen16-17_N_16	18/01/2017 06:34	19/01/2017 06:23	1392	5	38
Kohnen16-17_N_17	19/01/2017 06:43	20/01/2017 06:23	1686	4	35
Kohnen16-17_N_18	20/01/2017 06:47	21/01/2017 05:59	1742	5	36
Kohnen16-17_N_19	21/01/2017 06:15	21/01/2017 09:36	256	n.d.	n.d.
Kohnen16-17_N_20	21/01/2017 10:00	21/01/2017 13:34	286	n.d.	n.d.
Kohnen16-17_N_21	21/01/2017 14:02	21/01/2017 17:44	267	n.d.	n.d.
Kohnen_16-17_N_22	21/01/2017 18:05	21/01/2017 21:36	257	n.d.	n.d.
Kohnen_16-17_N_23	21/01/2017 22:00	22/01/2017 01:35	256	n.d.	n.d.
Kohnen_16-17_N_24	22/01/2017 02:00	22/01/2017 05:37	257	n.d.	n.d.
Kohnen_16-17_N_25	22/01/2017 06:00	22/01/2017 09:34	256	n.d.	n.d.
Kohnen_16-17_N_26	22/01/2017 10:00	22/01/2017 13:36	265	n.d.	n.d.
Kohnen_16-17_N_27	22/01/2017 14:00	22/01/2017 17:38	268	n.d.	n.d.
Kohnen_16-17_N_28	22/01/2017 18:00	22/01/2017 21:43	271	n.d.	n.d.

Sample name	Start time	End time	*Corrected volume (m <sup>3</sup> )	NO3 <sup>-</sup> mass concentration (ng m <sup>-3</sup> )	NO3 <sup>-</sup> dry deposition flux (pg m <sup>-2</sup> s <sup>-1</sup> )
Kohnen_16-17_N_29	22/01/2017 22:02	23/01/2017 01:43	268	n.d.	n.d.
Kohnen_16-17_N_30	23/01/2017 02:05	23/01/2017 05:49	270	n.d.	n.d.
Kohnen_16-17_N_31	23/01/2017 06:05	24/01/2017 06:22	1764	5	45
Kohnen_16-17_N_32	24/01/2017 06:39	25/01/2017 07:56	1821	6	50
Kohnen_16-17_N_33	25/01/2017 08:33	26/01/2017 06:07	1570	8	29
Kohnen_16-17_N_34	26/01/2017 06:36	27/01/2017 06:07	1718	1	7
Kohnen_16-17_N_35	27/01/2017 06:36	28/01/2017 06:07	1682	1	4

\*STP: standard temperature and pressure.

Table S3: e-folding depths calculated from irradiance profiles in the surface snowpack at two locations (pit A and B; same locations as snow pit A and B respectively) at Dronning Maud Land (DML). Reported are mean and standard deviation of e-folding depth in the 315-325 nm range, where the photolysis rate coefficients of NO<sub>3</sub><sup>-</sup> have their maximum.

Pit	Sampled depth	Time of experiment	Method	e-folding depth	
	range (cm)	(UTC)		(± 1 σ, cm)	
Pit A	0-22	27/01/2017 11:30	Horizontal	$4.0\pm1.3$	
Pit A	0-12	27/01/2017 16:00	Horizontal	$1.9\pm0.5$	
Pit B	0-29	28/01/2017 11:46	Horizontal	$0.9\pm0.3$	
Pit B	0-29	28/01/2017 12:00	Horizontal	$1.3 \pm 0.1$	
Pit B	0-41	28/01/2017 12:48	°45	$2.7\pm0.1$	
Pit B	0-21	28/01/2017 17:00	°45	$1.1 \pm 0.4$	

Time step (weeks)	Accumulation repartition (fraction)	<sup>1</sup> Atmospheric NO <sub>3</sub> <sup>-</sup> mass concentration (ng m <sup>-3</sup> )	Atmospheric boundary layer height (m)	<sup>2</sup> Air temperature (K)	<sup>2</sup> Air pressure (mbar)	Ground level ozone (ppbv)	D <sup>17</sup> O of ozone (‰)	Exported fraction (fraction)	Stratospheric flux repartition (fraction)	Stratospheric δ <sup>15</sup> N-NO <sub>3</sub> <sup>-</sup> (‰)	Stratospheric D <sup>17</sup> O (‰)	Long distance repartition (fraction)	Long distance δ <sup>15</sup> N- NO <sub>3</sub> <sup>-</sup> (‰)	Long distance D <sup>17</sup> O (‰)	<sup>3</sup> Ozone column (Dobson units)	Mean solar zenith angle (°)
0	0.019	8.93	50	247.9	681.3	32.9	25.2	0.2	0.067	19	42	0.019	0	30	306	112.9
1	0.019	8.87	50	251.6	684.5	34.0	25.2	0.2	0.067	19	42	0.019	0	30	298	112.6
2	0.019	9.40	50	247.4	677.4	34.5	25.2	0.2	0.067	19	42	0.019	0	30	294	112.0
3	0.019	10.40	50	242.9	676.5	35.1	25.2	0.2	0.067	19	42	0.019	0	30	284	111.0
4	0.019	11.17	50	249.0	676.1	34.9	25.2	0.2	0.067	19	42	0.019	0	30	269	109.8
5	0.019	11.15	50	250.6	679.9	35.0	25.2	0.2	0.067	19	42	0.019	0	30	256	108.2
6	0.019	10.29	50	250.9	684.1	34.0	25.2	0.2	0.067	19	42	0.019	0	30	243	106.5
7	0.019	9.59	50	249.8	684.2	34.3	25.2	0.2	0.067	19	42	0.019	0	30	221	104.5
8	0.019	9.25	50	250.6	680.2	32.1	25.2	0.2	0.067	19	42	0.019	0	30	211	102.3
9	0.019	11.07	50	249.6	681.2	33.8	25.2	0.2	0.067	19	42	0.019	0	30	189	100.0
10	0.019	13.99	50	247.1	677.7	33.8	25.2	0.2	0.067	19	42	0.019	0	30	166	97.5
11	0.019	18.93	50	251.0	681.8	33.0	25.2	0.2	0.067	19	42	0.019	0	30	146	94.9
12	0.019	24.77	50	239.8	664.2	32.2	25.2	0.2	0.050	19	42	0.019	0	30	137	92.3
13	0.019	30.61	50	222.3	674.0	31.7	25.2	0.2	0.033	19	42	0.019	0	30	131	89.6
14	0.019	34.82	50	239.2	684.2	30.7	25.2	0.2	0.017	19	42	0.019	0	30	136	86.9
15	0.019	38.25	50	242.9	676.9	30.4	25.2	0.2	0.000	19	42	0.019	0	30	146	84.3
16	0.019	39.25	50	238.9	676.4	29.2	25.2	0.2	0.000	19	42	0.019	0	30	158	81.7
17	0.019	39.77	50	245.1	661.5	28.2	25.2	0.2	0.000	19	42	0.019	0	30	180	79.2
18	0.019	39.77	50	245.3	665.9	27.9	25.2	0.2	0.000	19	42	0.019	0	30	199	76.8
19	0.019	42.20	50	232.2	666.1	29.0	25.2	0.2	0.000	19	42	0.019	0	30	213	74.6
20	0.019	43.95	50	233.4	672.5	32.2	25.2	0.2	0.000	19	42	0.019	0	30	229	72.6
21	0.019	46.44	50	228.5	681.6	35.5	25.2	0.2	0.000	19	42	0.019	0	30	243	70.9
22	0.019	47.82	50	238.3	666.9	32.2	25.2	0.2	0.000	19	42	0.019	0	30	257	69.5
23	0.019	47.40	50	228.0	649.1	33.1	25.2	0.2	0.000	19	42	0.019	0	30	272	68.3
24	0.019	44.66	50	246.8	677.8	31.4	25.2	0.2	0.000	19	42	0.019	0	30	281	67.5
25	0.019	41.45	50	247.5	678.1	32.4	25.2	0.2	0.000	19	42	0.019	0	30	284	67.1
26	0.019	37.43	50	241.4	666.8	26.1	25.2	0.2	0.000	19	42	0.019	0	30	281	67.1

Time step (weeks)	Accumulation repartition (fraction)	<sup>1</sup> Atmospheric NO <sub>3</sub> <sup>-</sup> mass concentration (ng m <sup>-3</sup> )	Atmospheric boundary layer height (m)	<sup>2</sup> Air temperature (K)	<sup>2</sup> Air pressure (mbar)	Ground level ozone (ppbv)	D <sup>17</sup> O of ozone (‰)	Exported fraction (fraction)	Stratospheric flux repartition (fraction)	Stratospheric δ <sup>15</sup> N-NO <sub>3</sub> <sup>-</sup> (‰)	Stratospheric D <sup>17</sup> O (‰)	Long distance repartition (fraction)	Long distance δ <sup>15</sup> N- NO <sub>3</sub> <sup>-</sup> (‰)	Long distance D <sup>17</sup> O (‰)	<sup>3</sup> Ozone column (Dobson units)	Mean solar zenith angle (°)
27	0.019	34.15	50	241.0	673.8	29.1	25.2	0.2	0.000	19	42	0.019	0	30	275	68.1
28	0.019	31.44	50	244.4	680.3	26.0	25.2	0.2	0.000	19	42	0.019	0	30	276	69.1
29	0.019	30.86	50	241.6	669.0	17.7	25.2	0.2	0.000	19	42	0.019	0	30	273	70.5
30	0.019	30.18	50	245.9	673.4	22.7	25.2	0.2	0.000	19	42	0.019	0	30	269	72.2
31	0.019	30.25	50	243.3	677.7	25.4	25.2	0.2	0.000	19	42	0.019	0	30	267	74.1
32	0.019	29.37	50	244.9	677.6	21.0	25.2	0.2	0.000	19	42	0.019	0	30	259	76.3
33	0.019	27.66	50	242.3	677.1	21.6	25.2	0.2	0.000	19	42	0.019	0	30	260	78.7
34	0.019	24.76	50	240.6	678.1	20.9	25.2	0.2	0.000	19	42	0.019	0	30	259	81.2
35	0.019	21.71	50	240.5	677.2	20.4	25.2	0.2	0.000	19	42	0.019	0	30	257	83.8
36	0.019	18.77	50	241.0	676.4	19.4	25.2	0.2	0.000	19	42	0.019	0	30	253	86.5
37	0.019	16.45	50	239.2	680.1	20.2	25.2	0.2	0.000	19	42	0.019	0	30	252	89.2
38	0.019	14.60	50	241.0	677.7	21.5	25.2	0.2	0.000	19	42	0.019	0	30	253	91.9
39	0.019	13.59	50	240.8	672.3	21.1	25.2	0.2	0.000	19	42	0.019	0	30	254	94.6
40	0.019	12.46	50	237.9	679.4	22.5	25.2	0.2	0.000	19	42	0.019	0	30	253	97.2
41	0.019	11.47	50	242.2	680.8	24.4	25.2	0.2	0.000	19	42	0.019	0	30	258	99.7
42	0.019	9.81	50	230.1	677.9	25.2	25.2	0.2	0.000	19	42	0.019	0	30	267	102.1
43	0.019	8.11	50	234.1	662.8	25.7	25.2	0.2	0.000	19	42	0.019	0	30	279	104.3
44	0.019	6.51	50	241.7	673.2	27.5	25.2	0.2	0.000	19	42	0.019	0	30	281	106.3
45	0.019	5.77	50	242.7	673.5	29.0	25.2	0.2	0.000	19	42	0.019	0	30	290	108.1
46	0.019	5.70	50	240.1	676.0	29.6	25.2	0.2	0.000	19	42	0.019	0	30	288	109.7
47	0.019	6.84	50	237.0	674.7	30.8	25.2	0.2	0.000	19	42	0.019	0	30	291	111.0
48	0.019	8.03	50	238.1	671.2	31.2	25.2	0.2	0.000	19	42	0.019	0	30	297	111.9
49	0.019	9.11	50	235.1	664.7	32.1	25.2	0.2	0.017	19	42	0.019	0	30	301	112.6
50	0.019	9.53	50	236.3	667.1	32.6	25.2	0.2	0.033	19	42	0.019	0	30	303	112.9
51	0.019	9.26	50	240.7	674.4	33.6	25.2	0.2	0.050	19	42	0.019	0	30	304	112.9

<sup>1</sup>Weller and Wagenbach (2007)

<sup>2</sup>Utrecht University automatic weather Station DML05/Kohnen (AWS9; <u>https://www.projects.science.uu.nl/iceclimate/aws/files\_oper/oper\_20632</u>) <sup>3</sup>NIWA Bodeker version 3.3 at the location of the snow pit averaged from 2000-2016 (<u>http://www.bodekerscientific.com/data/total-column-ozone</u>) Table S5: Average photolysis rate coefficients of nitrate in snow above the East Antarctic Plateau observed at a SZA of 68° compared to model estimates from TUV-snow. DML: Dronning Maud Land.

Location	TCO (DU)	$2\pi J(NO_{3}) (s^{-1})$	Period of observations
<sup>1</sup> DML	306	0.98 x10 <sup>-8</sup>	1-14 January 2017
<sup>2</sup> Dome C	287	2.52 x10 <sup>-8</sup>	1-14 January 2012
<sup>3</sup> TUV-snow	275	2.71 x10 <sup>-8</sup>	
<sup>3</sup> TUV-snow	300	2.57 x10 <sup>-8</sup>	
<sup>3</sup> TUV-snow	325	2.44 x10 <sup>-8</sup>	

<sup>1</sup>This study

<sup>2</sup>Kukui et al. (2013)

<sup>3</sup>Erbland et al. (2015)

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