

SUPPLEMENTARY INFORMATION

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Globally important nitrous oxide emissions from croplands induced by freeze-thaw cycles

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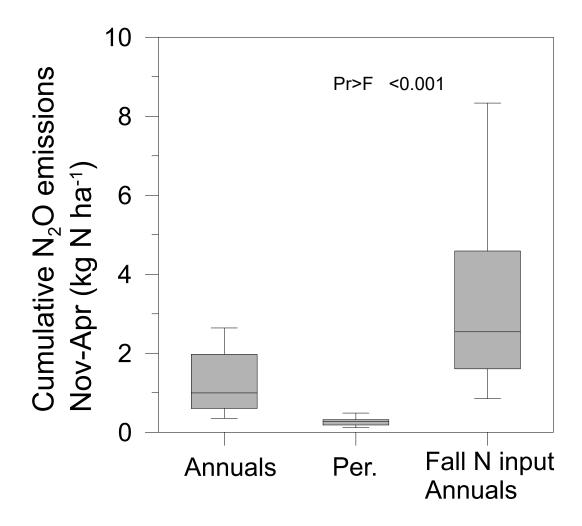


Figure S1 Box plot of cumulative non-growing season N_2O emissions (Nov-Apr) for the CA1 and CA2 sites separated by management (bar indicates 5^{th} and 95^{th} percentiles): annual and perennial crops, and annual crops receiving fall manure or inorganic nitrogen fertilizer. The p-value for the F test indicates that the cropping system accounts for a significant portion of the variability of the dependent variable: N_2O emissions. N=44 for annual system, N=7 for perennial system, N=9 for fall nitrogen application to annuals.

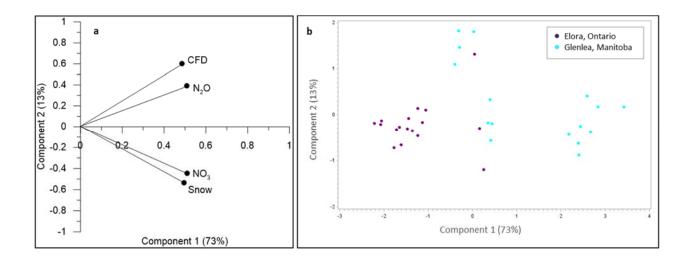


Figure S2 Relationship between cumulative freezing degree days (CFD), cumulative N_2O emissions (Nov-Apr, kg N ha⁻¹), soil NO₃- concentration, and snow depth data based on **a**, factor analysis and **b**, principal component analysis with data partitioned by site based. All data from the CA1 and CA2 sites.

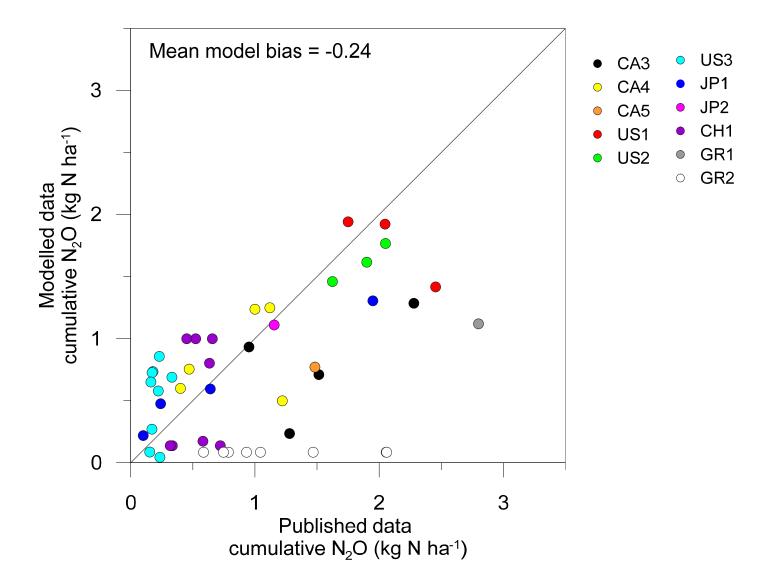


Figure S3 | Validation of the exponential-to-plateau model for predictions of cumulative N_2O emissions (Nov-Apr) compared to that of previously published data from Canada (CA3, CA4 and CA5), USA (US1 and US2), Japan (JP1 and JP2), China (CH1) and Germany (GR1 and GR2). Line indicates 1:1 slope. See Table S4 for description of sites.

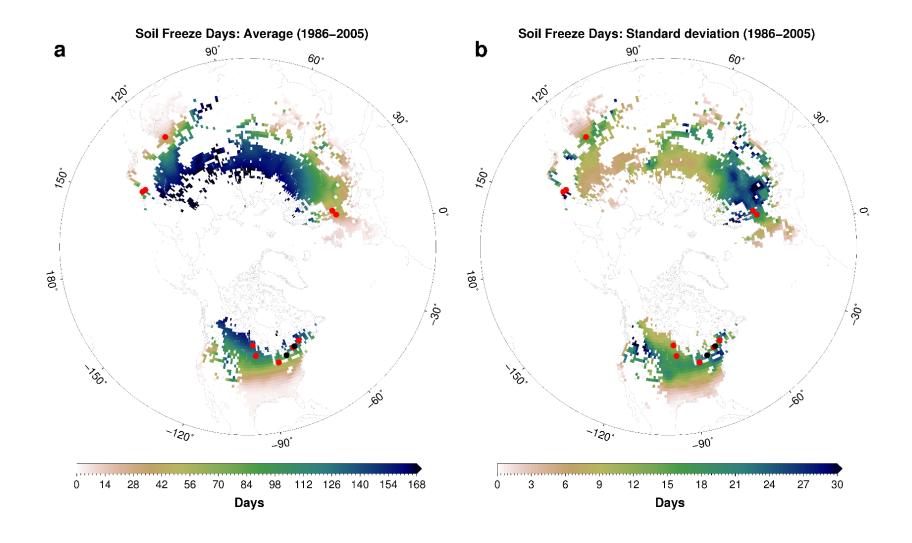


Figure S4 Global distribution of **a**, average and **b**, standard deviation of number of days with soil freezing derived from three re-analysis models (ERA-Interim, MERRA-Land and GLDAS) for the 1986-2005 period in cropland areas of the Northern Hemisphere identified using Terra and Aqua MODIS data. Black dots indicate study sites and red dots validation sites.

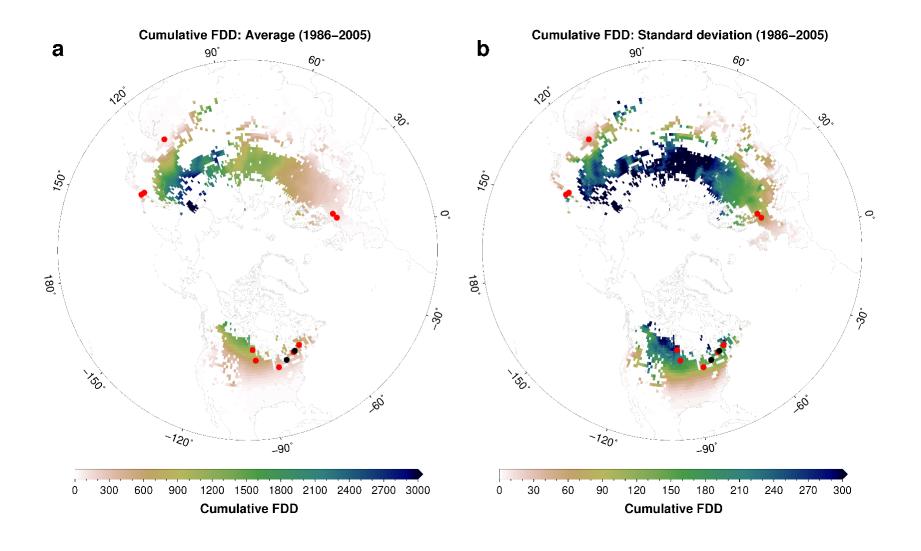


Figure S5 | Global distribution of **a**, average and **b**, standard deviation of cumulative freezing degree-days (FDD) over Nov-Apr calculated using soil temperature from three re-analysis models (ERA-Interim, MERRA-Land and GLDAS) for the 1986-2005 period in cropland areas of the Northern Hemisphere identified using Terra and Aqua MODIS data. Black dots indicate study sites and red dots validation sites.

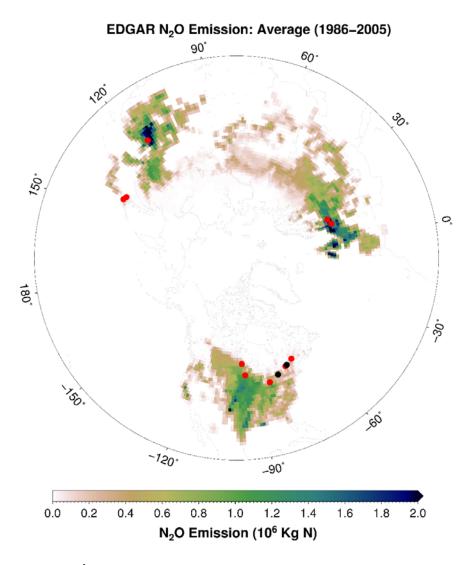


Figure S6 Mean global distribution of annual direct N_2O emissions retrieved from the EDGAR database (http://edgar.jrc.ec.europa.eu, 2016) for areas with frozen cropland from 1986-2005. Black dots indicate study sites and red dots validation sites. Note that the original EDGAR information at 0.1 degree resolution was summed over 1 x 1 degrees for comparison to Figure 3.

Table S1 | Description of the two experimental sites

	Elora, Ontario (CA1)	Glenlea, Manitoba (CA2)
Study coordinates	43°38′N 80°25′W, 376 m amsl	49°38′N 97°9′W, 235 m amsl
Number of years (Study years)	14 (2000-2007; 2009-2014)	9 (2006-2014)
Number of plot-years ^z	56 (28) ^y	36 (16) ^y
Number of flux values		
Total daily	13682 (4429) ^y	8590 (1756) ^y
Total half-hourly	90574 (31208) ^y	58196 (10910) ^y
Mean annual temperature x (°C)	6.7	3.0
Mean total annual precipitation (mm)	946	521
Median snow depth and range (cm)	9 (2 to 21)	20 (5-29)
Mean soil nitrate content and range (mg kg ⁻¹) Oct-Nov	5.3 (1.6 to 18.2)	17.1 (2.3 to 35.6)
Median N_2O emissions and range (kg N $ha^{\text{-}1}\ yr^{\text{-}1})$ Nov-Apr	0.69 (0.29 to 2.5)	2.1 (0.94 to 2.8)
Mean proportion of annual $\ensuremath{N_2} O$ emissions and range (%) Nov-Apr	53 (19 to 93)	29 (7 to 52)

²four large plots were monitored in each year yielding four time-series per year

Yvalue in brackets refers to datasets on annual crops without fall nitrogen application for which the Nov-Apr period was used in this study

^{*30-}yr normal, Environment Canada 2010

Table S2 | Overview of field experiments conducted at the two sites where N2O flux was measured using a micrometeorological method and corresponding data sources.

Site name	Year	Crops	Management treatments	Publication
Elora, Ontario (CA1)	2000-2005	Corn-Soybean-Winter wheat-Corn- Soybean-Corn	Conventional practices vs. best management practices	¹ Wagner-Riddle et al. (2007)
,	2005-2007	Corn-Soybean-Corn	Conventional tillage vs. no-till	² Congreves et al. (2016)
	2009-2011	Corn-Soybean-Winter barley	Conventional tillage vs. no-till; crop residue removed vs. crop residue returned	^{2,3} Partially in Nemeth et al. (2014); Congreves et al. (2016)
	2011-2014	Perennial grass-legume mixture and Corn monoculture	Manure injection vs. broadcast application (perennial); fall vs. spring manure application (corn)	⁴ Abalos et al. (2016)
Glenlea, Manitoba (CA2)	2006-2008	Corn-Faba-Spring wheat	Intensive tillage vs. reduced tillage	⁵Glenn et al. (2012)
(==,	2008-2010	Perennial grass-legume mixture and Spring wheat-Rapeseed	NA	⁶ Maas et al. (2013)
	2010-2012	Spring wheat-Corn	Late fall vs. pre-plant spring anhydrous ammonia application	⁷ Tenuta et al. (2016)
	2012-2014	Corn-Soybean-Spring wheat	NA	Unpublished

Table S3 | Characterizing the relationship between cumulative freezing degree-days (CFD) and N₂O emissions for the Nov-Apr period using three different models.

	Exponential-to-plateau (1)	Linear regression (2)	Multiple linear regression (3)
p value	<0.0001	<0.0001	<0.0001
N	44	44	33
Correlation	pseudo-R ² 0.74	Adj-R ² 0.56	Adj-R ² 0.60
AIC	74	-57	-45
BIC	80	-55	-43
SSE	12	11	7.0
MSE	0.29	0.26	0.23
RMSE (kg N ha ⁻¹ yr ⁻¹)	0.52	0.51	0.46
CV	42%	41%	37%
MAPE	50%	33%	27%
Model bias (kg N ha ⁻¹ yr ⁻¹)	-2.1	2.1	0.72

 $⁽¹⁾ y = 1.98(1 - e^{(-0.00724x)})$

(3) $y = 0.00133x_1 + 0.0298x_2 + 0.668$ where $y = \text{cumulative } N_2O \text{ emissions } (\text{Nov} - \text{Apr}), x \text{ (and } x_1) = \text{CFD accumulated from Nov} - \text{Apr}, \text{ and } x_2 = 0.00133x_1 + 0.00298x_2 + 0.668$ average soil NO_3^- concentration during the previous fall.

⁽²⁾ y = 0.002x + 0.747

Table S4 \mid Listing of frequently cited studies linking freeze-thaw cycles and N₂O emissions, and reason for exclusion from our validation dataset.

Site	Soil Type	Crop	Temperature (°C) ^y	Reference	Reason for exclusion
Wisconsin, USA	Silt Loam	NA	-2 to 8 (2)	⁸ Goodroad and Keeney (1984)	Low frequency N₂O measurements
Wisconsin, USA	Kiddler Loam	Corn	-1 to 4 (1)	⁹ Cates and Keeney (1987)	Low frequency N ₂ O measurements (during winter)
Delhi, Ontario, Canada	Sandy Loam	Corn	-5 to 13 (15)*	¹⁰ Burton and Beauchamp (1994)	Not possible to calculate FDD, Unknown N ₂ O sampling frequency
NA	Loam	Rapeseed	-16 to 8	¹¹ Teepe et al. (2001)	Lab study
St- Lambert, Chapais, Quebec, Canada	Loam; Sandy Loam	NA, Barley	-4.4 to 17.5 (15)	¹² van Bochove et al. (2001)	Low frequency N₂O measurements (6)
Elora, Ontario, Canada	Silt Loam	Soybean	-1 to 15 (5)*	¹³ Drewitt and Warland (2007)	Measurement of belowground N ₂ O concentration, not N ₂ O emissions
Eastern Finland	Dystric Regesol, Silty Sand	Fescue Lawn	-10 to 10 (5)*,w	¹⁴ Maljanen et al. (2007)	Not cropland
Northeast Bavaria,	Haplic Podsol	Spruce Forest	-2 to 15 (5) ^x	¹⁵ Goldberg et al. (2010)	Not cropland
Germany Lower Saxony,	Silty Loam	Winter Wheat	-4 to 3	¹⁶ Röver et al. (1998)	Not possible to calculate FDD
Germany Elora, Ontario, Canada	Silt Loam	Winter Wheat, Corn	0 (5) ^v	¹⁷ Wagner-Riddle et al. (2008)	Included in the database as part of Wagner-Riddle et al. 2007
Ellerslie, Alberta, Canada	Eluviated Black Chernozemic, Malmo Loam	Barley	-1 to 10	¹⁸ Nyborg et al. (1997)	N ₂ O emissions measured during 31-33 days
Bergen, Norway	NA	Beech Forest	−7 to ~25	¹⁹ Skogland et al. (1988)	Lab study, N₂O not measured, Not cropland
Ronhave, Denmark	Sandy Loam	Spring Barley	-20 to 10	²⁰ Christensen and Christensen (1991)	Lab study
Various Locations	Typic Claicaquoll; Aquic Hapludoll; Typic Hapludoll	Corn, Soybean, Prairie	-20 to 25	²¹ DeLuca et al. (1992)	Lab study, N₂O not measured
Alaska, USA	Taiga soils; Tundra soils	Forest, Tundra	-5 to 5 ^u	²² Schimel and Clein (1996)	Lab study, N₂O not measured
Clinton & Delhi, Ontario, Canada	Grey Brown Luvisols: Sandy Loam; Sand	NA NA	NA	²³ Ryan et al. (2000)	N₂O not measured
Giessen, Germany	NA	Grassland	-20 to 20	²⁴ Müller et al. (2002)	Not cropland
Uppsala, Sweden	Typic Eutrochrept: Post-Glacial Silt Loam	Various	–5 to 5	²⁵ Herrmann and Witter (2002)	Lab study, N₂O not measured

Scheyern, Munich, Germany	Dystric Eutrochrept: Silt Loam	NA	-12 to 4	²⁶ Sehy et al. (2004)	Lab study
NA	Mollic Gleysol: Silt Loam	Potatoes	-20 to 5	²⁷ Mørkved et al. (2006)	Lab study
Tsukui District, Kanagawa, & Cübu Region, Japan	Various	Grassland, Oak Forest, Arable field, Pine forest	-13 to 4	²⁸ Yanai et al. (2007)	Lab study
La Pocatière, Québec, Canada	Orthic Humic Gleysol	Barley, Red Clover	-12 to 4	²⁹ van Bochove et al. (2000b)	Lab study
Bavarian tertiary hills, Germany	Dystric Eutrochrept	Grassland	-20 to 10	³⁰ Sharma et al. (2006)	Lab study, Not cropland
New Hampshire, USA	Sandy Loam	American Beech, Sugar Maple, Yellow Birch	NA	³¹ Tierney et al. (2001)	N₂O not measured
Germany, Sweden, and Finland	NA	NA	0 to 30 ^t	³² Holtan-Hartwig et al. (2002)	Lab study
Ariss, Ontario, Canada	Silt Loam	Corn	-5 to 15 (5)	³³ Wagner-Riddle et al. (2010)	Included in the database as part of Wagner-Riddle et al. 2007
Scheyern, Munich, Germany	Typic Udifluvent, Dystric Eutrochrept	Corn	NA	³⁴ Sehy et al. (2003)	Not possible to calculate FDD
Shanxi province, China	Mottlic Hapli- Ustic Argosols	Winter Wheat, Corn	-5 to 30*	³⁵ Liu et al. (2012)	Not possible to calculate FDD
Potsdam, Germany	Sandy Loam	Rape, Rye, Triticale, Hemp, Poplar, Willow	NA	³⁶ Kavdir et al. (2008)	Not possible to calculate FDD
Montana, USA	Amsterdam Silt Loam	Wheat, Pea, Grass- Alfalfa	NA	³⁷ Dusenbury et al. (2008)	Not possible to calculate FDD
Munich, Germany	NA	Potato, Winter Wheat, Corn	–18 to 20 ^x	³⁸ Ruser et al. (2001)	Not possible to calculate FDD

 $^{^{\}rm z}$ depth (cm below surface) of soil sample collection shown in brackets $^{\rm y}$ depth (cm below surface) of temperature measurement shown in brackets

^{*} estimated from figure

bare soils (-10 to 0°C); snow-covered soils (0 to 10°C)

average temperature from Jan.-Feb.

stored at -20°C

 $^{^{\}rm t}$ soils subjected to different temperature cycles ranging between 0 and 30°C

 Table S5| Overview of field experiments used for model validation.

Site name	Year	Management treatments	Crop	Soil texture/type	Flux method	Frequency of measurements	MAT ^z (°C)	MAP ^y (mm)	CFD ^x	Publication
Ontario, Canada (CA3)	2012- 2014	Raw dairy manure vs. digestate dairy manure vs. inorganic fertilizer	Corn	Clay, sandy loam	Chambers	Weekly, bi- weekly, monthly	6.6	920	77.8	³⁹ Schwager et al. 2016; Unpublished
Québec, Canada (CA4)	2009- 2013	With vs. without pig slurry	Barley	Sandy loam and silty clay	Chambers	Weekly, bi- weekly	4.4	1231	85	⁴⁰ Chantigny et al. 2016
Ontario, Canada (CA5)	1997	NA	Corn	Clay loam	Flux- gradient	30 min	6	943	68	⁴¹ Pattey et al. 2007
Minnesota, USA (US1)	2005- 2008	Tillage (chisel/moldboard vs. strip), rotation (2 vs. 4-yr) and fertilizers (no fertilizer vs. fertilizer)	Corn- soybeans	Loam, silty clay loam, clay loam	Chambers	Weekly, bi- weekly	5.78	645	412.5	⁴² Johnson et al. 2010; Unpublished
Minnesota, USA (US2)	2009- 2012	No tillage	Corn- soybeans	Loam, silty clay loam, clay loam	Chambers	Weekly, bi- weekly, monthly	5.78	672	242.1	⁴³ Johnson and Barbour, 2016
Michigan, USA (US3)	2010- 2013	Ambient snow cover vs. no-snow cover vs. double-snow cover	Winter wheat-corn-soybeans	Loam	Chambers	Daily	9.9	1027	44	⁴⁴ Ruan and Robertson, 2016
Hokkaido, Japan (JP1)	2008- 2009	Removal of snow vs. acceleration of snow cover melting vs. untreated control	NA	Volcanic ash-derived andosol	Chambers	2-3-day intervals, weekly	8.8, 6.0 ^w	1022, 941 ^w	62.9	⁴⁵ Yanai et al. 2011
Hokkaido, Japan (JP2)	2004- 2005	NA	Corn	Histosol	Chambers	Weekly, monthly	7.9	1365	113.5	⁴⁶ Katayanagi and Hatano, 2012
Lower Saxony, Germany (GR1)	1995- 1996	NA	Oil seed rape	Loam	Chambers	Weekly	Unknown	Unknown	115	⁴⁷ Teepe et al. 2000

Brandenburg, Germany (GR2)	1999- 2007	Nitrogen fertilizer applications	Rape, rye, triticale, hemp, poplar, willow	Loamy sand	Chambers	4 times a week	9.9	590	6	⁴⁸ Hellebrand et al. 2008
Shandong Province, China (CH1)	2011- 2012	Fertilizers (high vs. balanced vs. controlled release vs. no fertilizer), tillage (rotary vs. deep plowing vs. no tillage) and irrigation (flood vs. decreased)	Winter wheat- corn	Sandy-loam	Chambers	Daily, weekly	12.5	543	50.6	⁴⁹ Shi et al. 2013

^zmean annual temperature

^ymean annual precipitation

 $^{^{}x}$ cumulative freezing degree-days (soil temperature at 5 cm depth < 0°C) for November to April

wfor two study sites

Table S6 Comparison of cumulative N_2O emission averages (and standard deviations) for Nov-Apr obtained from experiments and predicted by using the exponential to plateau-model and the average freezing-degree day (CFD) estimates derived from ERA-Interim, MERRA-Land, and GLDAS for each site.

	Cumulative N ₂ O emissions (kg N ha ⁻¹)				
	Based on experimental N ₂ O and	Based on the exponential-to-			
	CFD data	plateau model and estimated CFD			
Canada (Ontario, CA1)	0.853 (0.54)	1.82 (0.19)			
Canada (Manitoba, CA2)	1.92 (0.61)	1.95 (0.01)			
Canada (Ontario, CA3)	1.44 (0.33)	1.88 (0.07)			
Canada (Quebec, CA4)	0.84 (0.34)	1.88 (0.07)			
Canada (Ontario, CA5)	1.48 (n/a)	1.88 (0.08)			
USA (Minnesota, US1	2.28 (0.03)	1.93 (0.67)			
and US2)	,	, ,			
USA (Michigan, US3)	0.21 (0.05)	1.50 (0.37)			
Japan (Hokkaido, JP1 and JP2)	0.82 (0.80)	1.32 (0.91)			
Germany (Lower	2.80 (n/a)	0.82 (0.48)			
Saxony, GR1)					
Germany (Brandenburg,	1.21 (0.55)	1.32 (0.80)			
GR2)					
China (Shandong, CH1)	0.53 (0.14)	1.32 (0.80)			
Mean prediction bias		0.29			
Mean absolute error		55%			

n/a, standard deviations not applicable because sample size =1

Table S7 | Cumulative freezing degree-days calculated from soil temperature at 5 cm $<0^{\circ}$ and cumulative N₂O emissions over Nov-Apr for the main study sites (CA1 and CA2, see Table S1) and validations sites (see Table S5).

(300 10010 33).	Cumulative Freezing	Cumulative N ₂ O emissions
Identifier	Degree-days (<0 C)	(Nov-Apr)
(Table S1 and S5)	30 7- (7	Kg N/ha
		<u> </u>
CA1	179.3	1.157
CA1	33.7	0.420
CA1	370.4	2.524
CA1	97.2	0.826
CA1	149.0	1.005
CA1	88.5	0.991
CA1	146.7	1.272
CA1	77.8	0.388
CA1	39.1	0.572
CA1	59.1	0.579
CA1	64.9	0.650
CA1	58.4	0.856
CA1	52.7	0.603
CA1	62.2	0.950
CA1	67.7	1.429
CA1	71.7	0.544
CA1	39.6	1.973
CA1	38.7	0.581
CA1	15.1	0.811
CA1	20.9	0.420
CA1	9.5	0.639
CA1	58.8	0.746
CA1	107.6	1.293
CA1	36.5	0.292
CA1	18.8	0.352
CA1	101.7	0.382
CA1	43.0	0.338
CA1	114.3	0.722
CA2	444.7	2.293
CA2	538.1	2.645
CA2	533.3	2.224
CA2	579.2	2.072
CA2	490.8	1.574
CA2	479.8	1.170
CA2	462.7	1.280
CA2	473.2	0.939

CA2	706.0	2.083
CA2	794.1	2.507
CA2	751.9	2.641
CA2	852.8	2.765
CA2	550.1	1.727
CA2	504.5	2.456
CA2	778.2	1.277
CA2	627.4	1.045
CA3	61.3	1.514
CA3	144.6	2.279
CA3	17.5	1.279
CA3	87.8	0.952
CA4	40.0	1.220
CA4	66.2	0.470
CA4	49.8	0.400
CA4	137.6	1.120
CA4	135.2	1.000
CA5	68.1	1.483
US1	442.7	4.326
US1	173.5	2.454
US1	488.8	2.048
US1	544.9	1.750
US2	233.9	1.900
US2	184.5	1.624
US2	307.8	2.051
US3	78.3	0.230
US3	63.9	0.179
US3	59.0	0.332
US3	20.2	0.170
US3	6.1	0.152
US3	3.1	0.235
US3	63.1	0.171
US3	54.9	0.162
US3	47.6	0.222
JP1	16.1	0.100
JP1	37.9	0.240
JP1	49.2	0.640
JP1	148.4	1.950
JP2	113.6	1.155
CH1	96.9	0.523
CH1	96.9	0.450
CH1	96.9	0.656
CH1	71.7	0.633

CH1	9.9	0.335
CH1	9.9	0.317
CH1	9.9	0.721
CH1	12.6	0.581
GR1	115.0	2.800
GR2	6.0	2.056
GR2	6.0	0.932
GR2	6.0	2.060
GR2	6.0	0.786
GR2	6.0	1.469
GR2	6.0	0.747
GR2	6.0	1.044
GR2	6.0	0.586

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