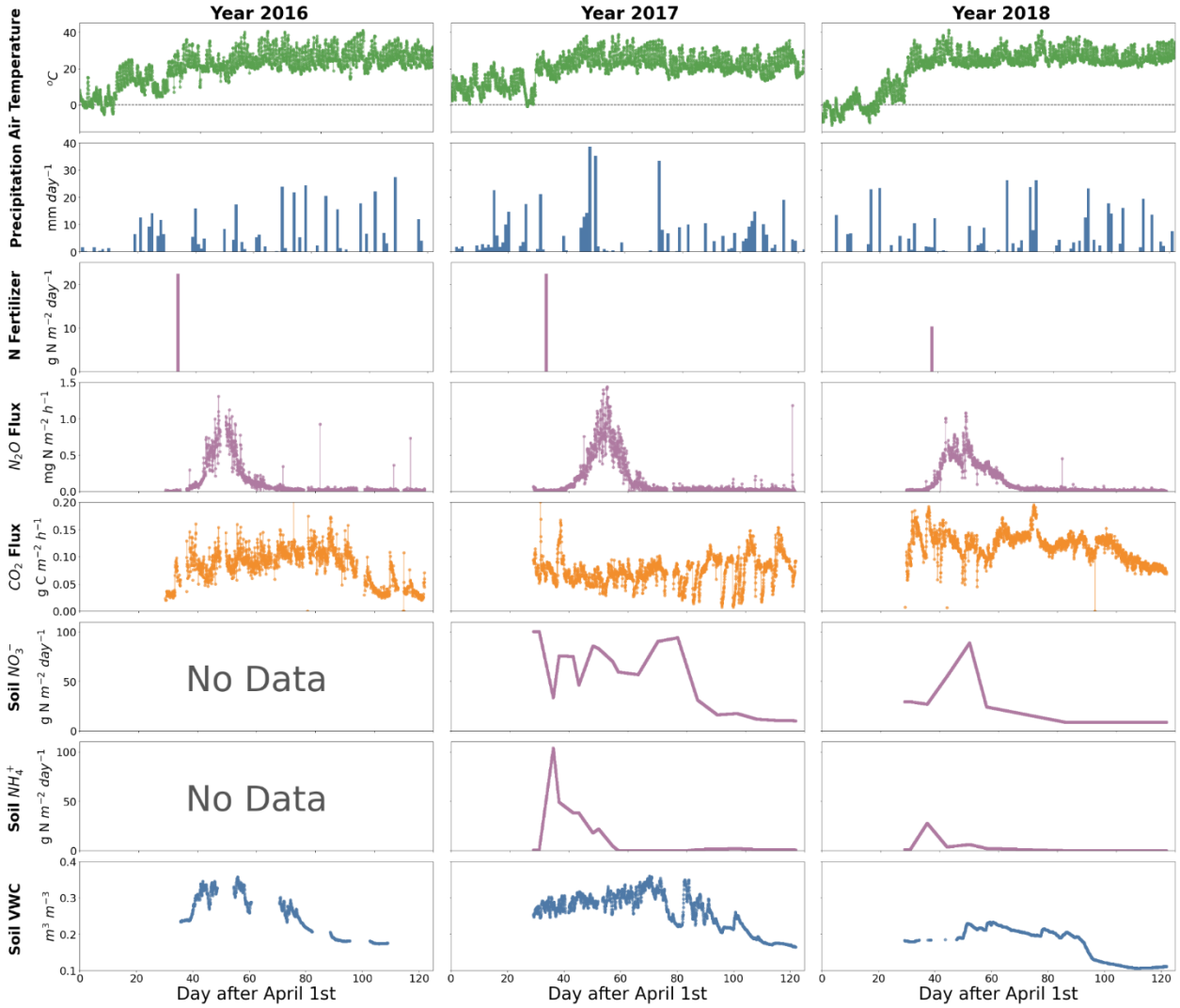


Figure S1: Time series example of observation data collected from mesocosm chamber 1. The precipitation, N fertilizer, Soil NO_3^- and NH_4^+ data are in daily time scale, while other data are in hourly time scale. Temperature presents in green; water related variables (precipitation and soil VWC) are in blue; N related variables (N fertilizer, N_2O flux, Soil NO_3^- and NH_4^+) are in purple; and CO_2 is in orange. Anomaly points will be down-weighted by daily averaging method with quality check in later processes, which is mentioned in section 2.2.2 last paragraph.

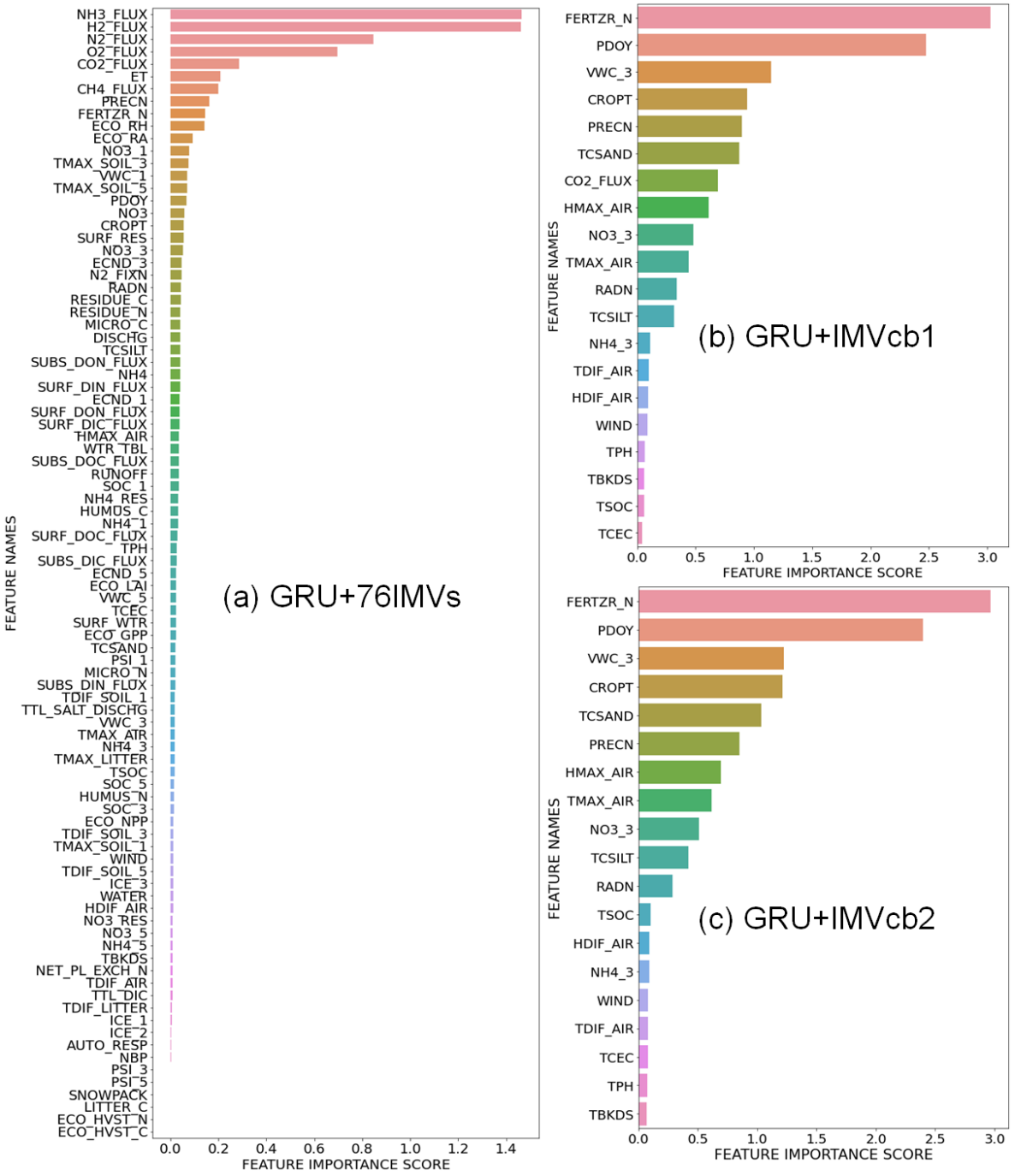
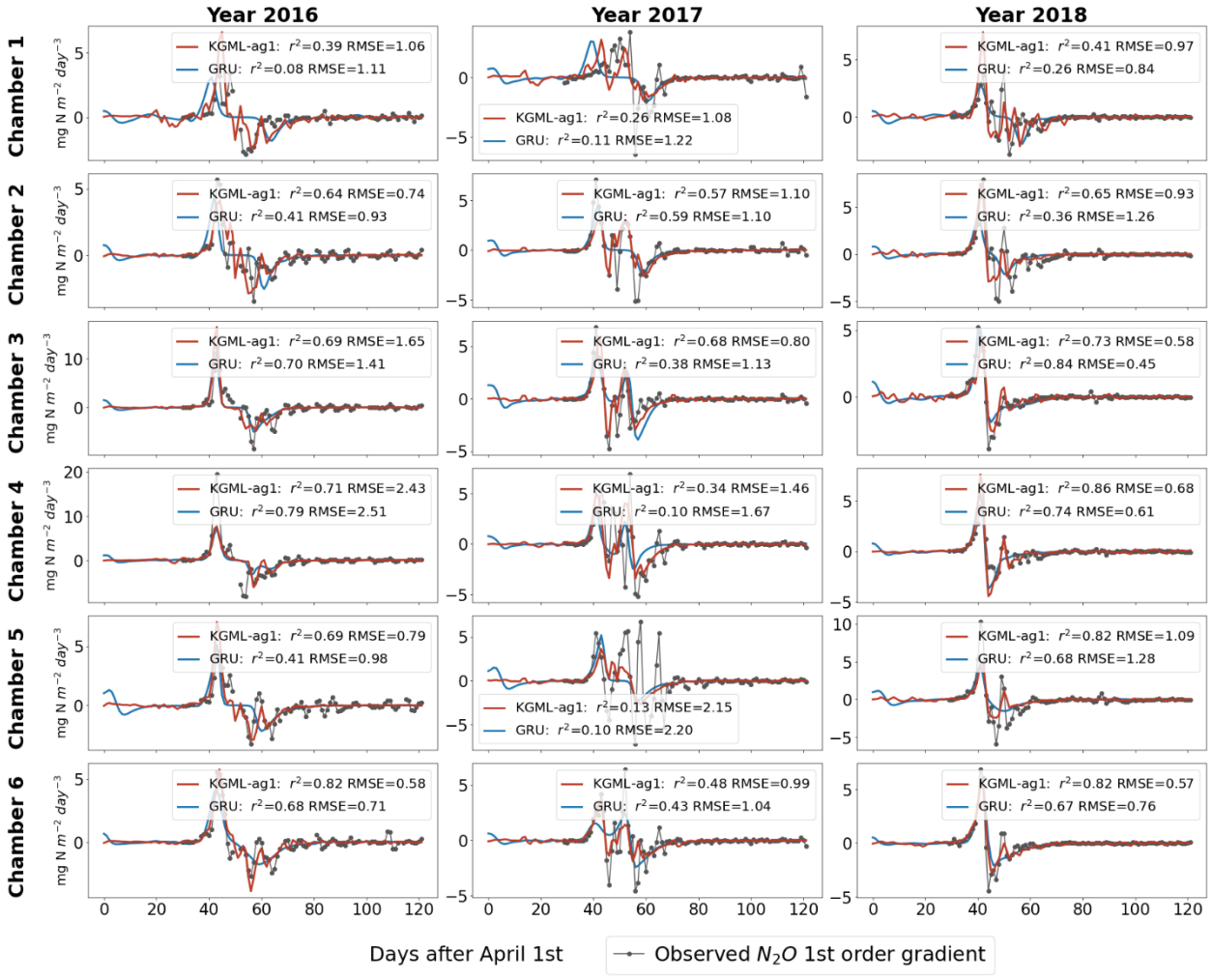


Figure S2: Feature importance test for intermediate variables (IMVs) with GRU models. To be noticed the VWC, NO_3^-, and NH_4^+ from third layer soil, which are presented in the main text are abbreviated here as VWC_3, NO_3_3, and NH_4_3 to be distinguished from the same variables from 1st and 5th layers. Details of the variables can be found in Table S1.



15 **Figure S3: N_2O flux 1st order gradient time series comparisons between non-pretrained GRU model and KGML-ag1. The black-dot line represents the observation, while blue represents GRU and red represents KGML-ag1.**

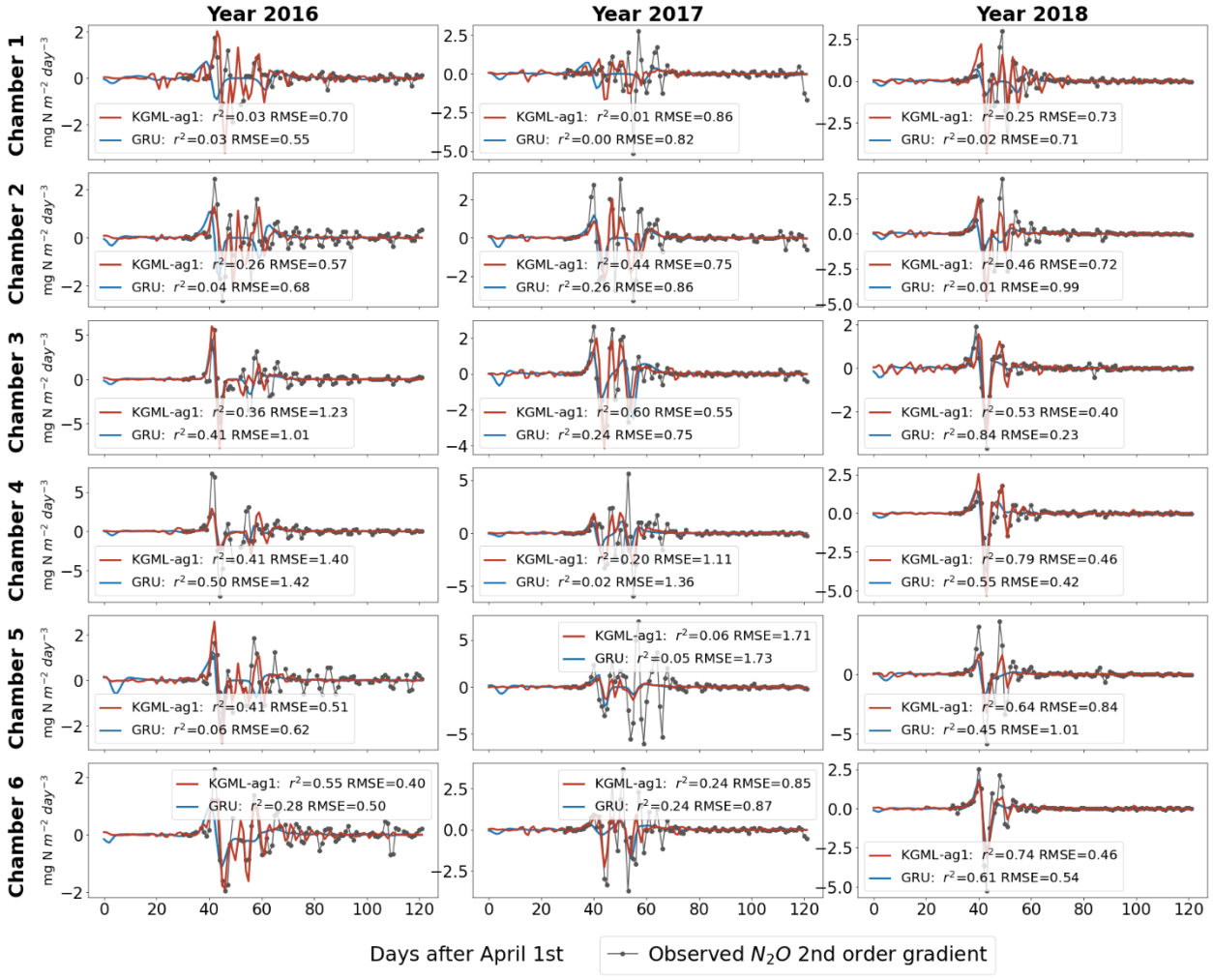


Figure S4: N_2O flux 2nd order gradient time series comparisons between non-pretrained GRU model and KGML-ag1. The black-dot line represents the observation, while blue represents GRU and red represents KGML-ag1.

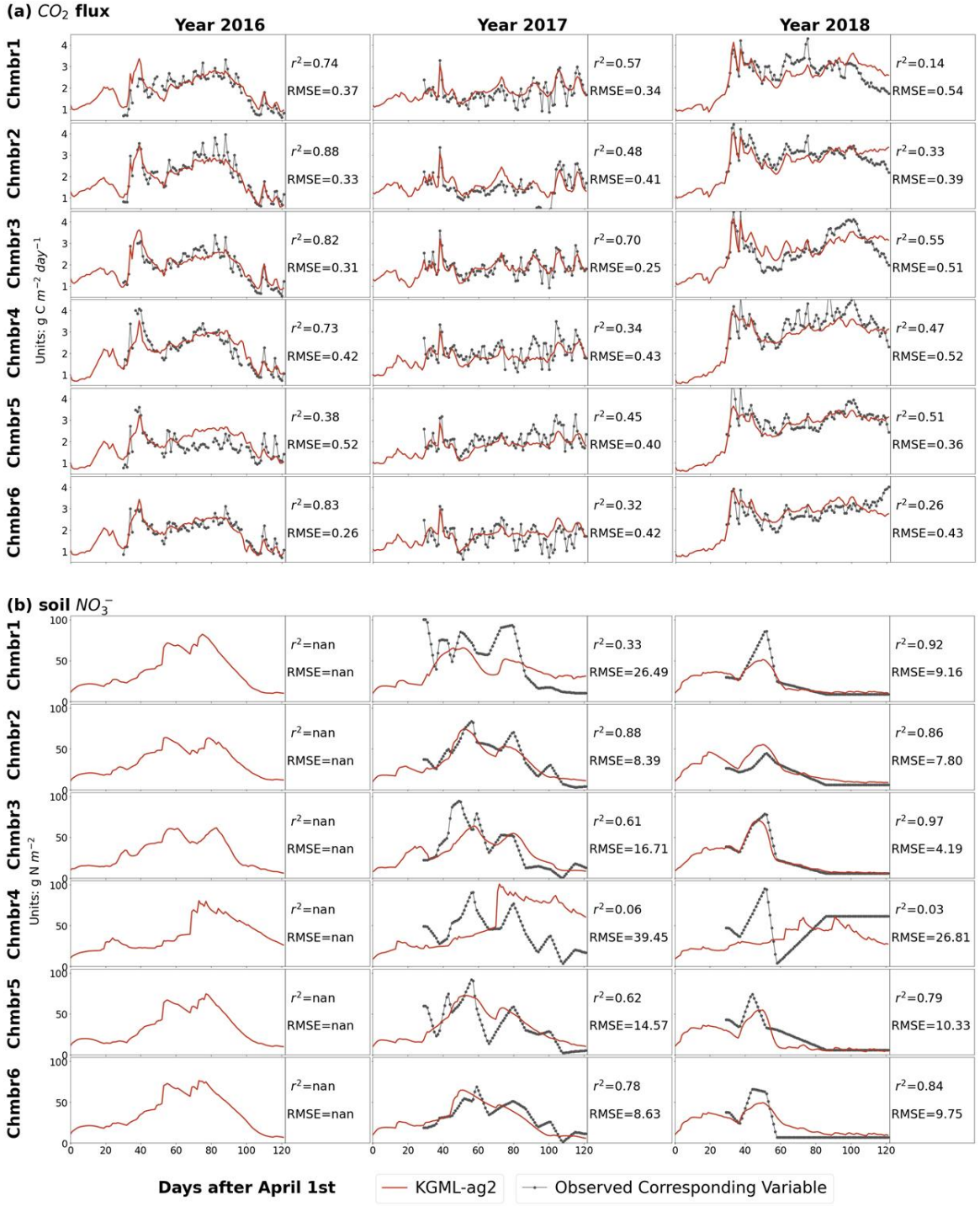


Figure S5: IMVs prediction from KGML-ag2. The black-dot line represents observations and the red line represents the results from KGML-ag2. Chmb is the abbreviation for chamber. r^2 and RMSE are calculated and present in each year and chamber.

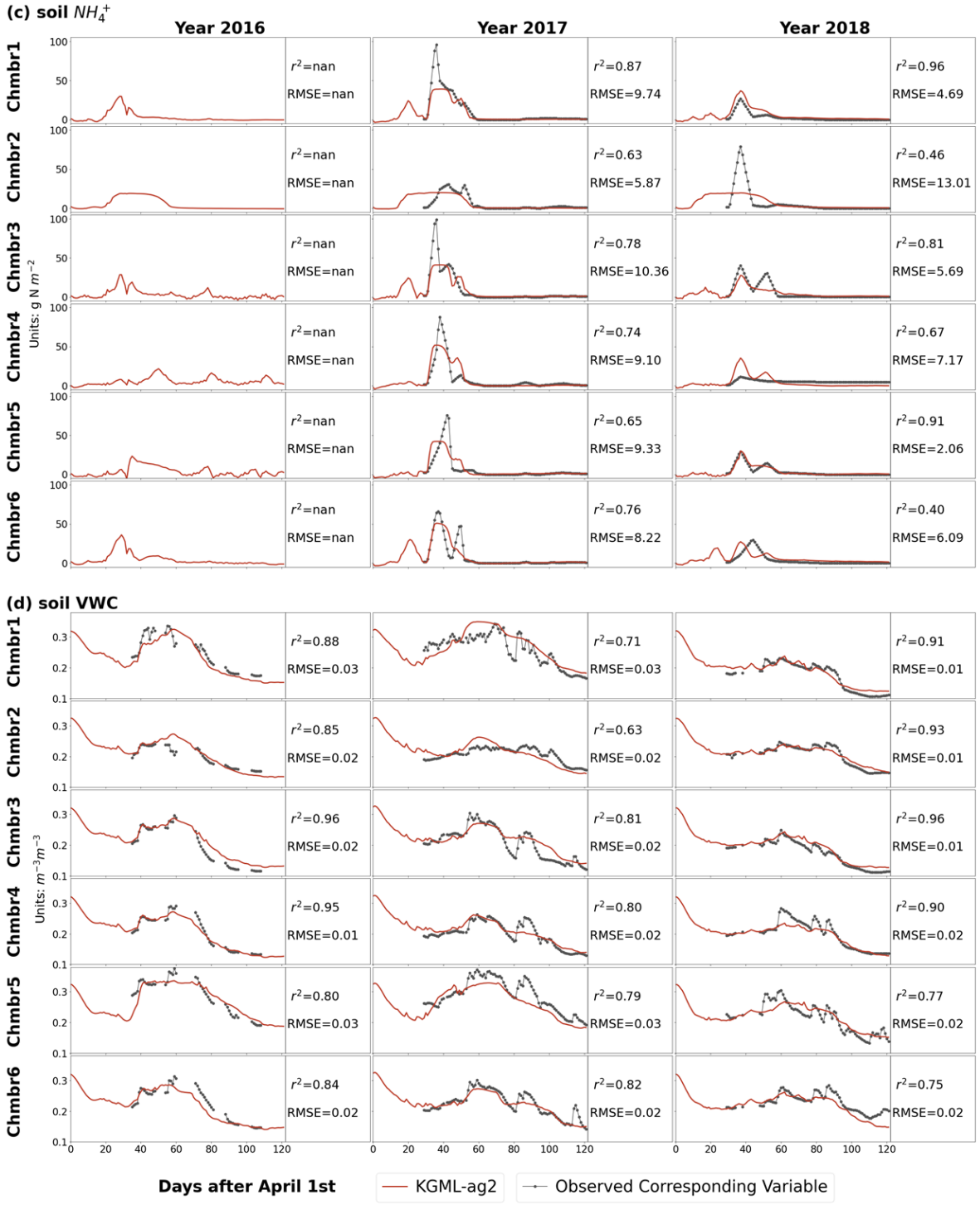
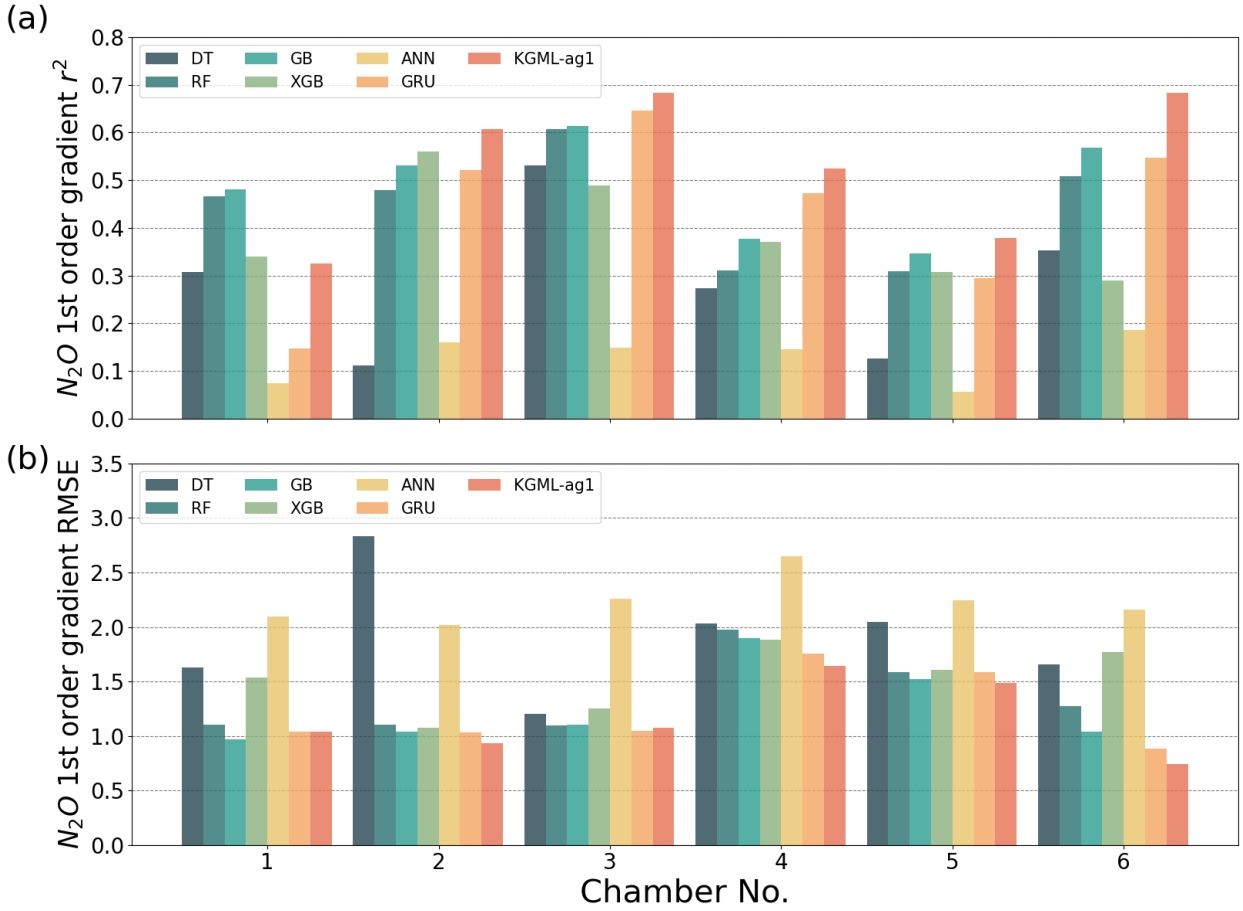


Figure S5 contd.: IMVs prediction from KGML-ag2. The black-dot line represents observations and the red line represents the results from KGML-ag2. Chmb is the abbreviation for chamber. r^2 and RMSE are calculated and present in each year and chamber.



30 **Figure S6: The comparisons of N_2O 1st order gradient prediction accuracy r^2 (a) and (b) RMSE, between four tree-based ML models (DT, RF, GB and XGB), two deep learning models (ANN and GRU) and KGML-ag1 model in 6 chambers.**

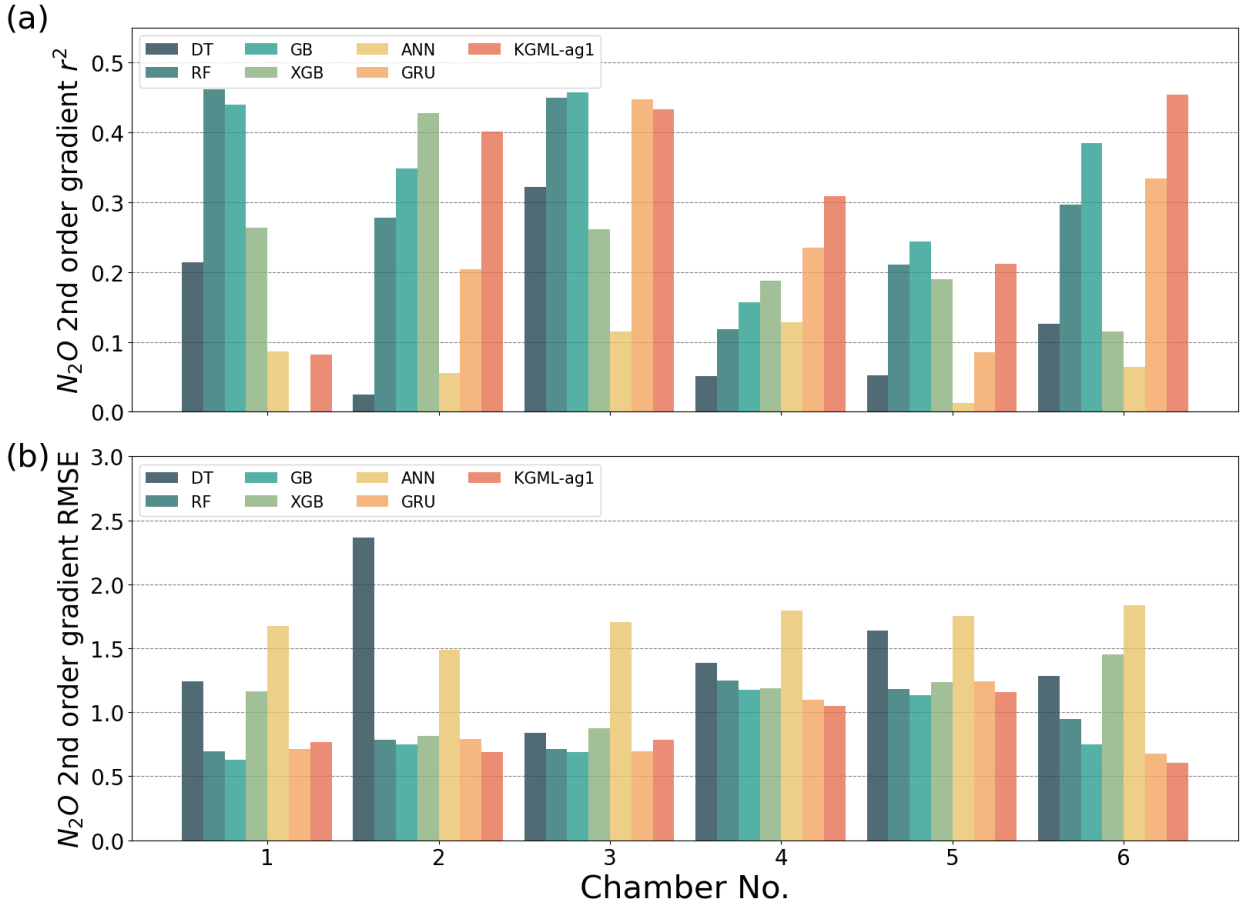


Figure S7: The comparisons of N_2O 2nd order gradient prediction accuracy r^2 (a) and (b) RMSE, between four tree-based ML models (DT, RF, GB and XGB), two deep learning models (ANN and GRU) and KGML-ag1 model in 6 chambers.

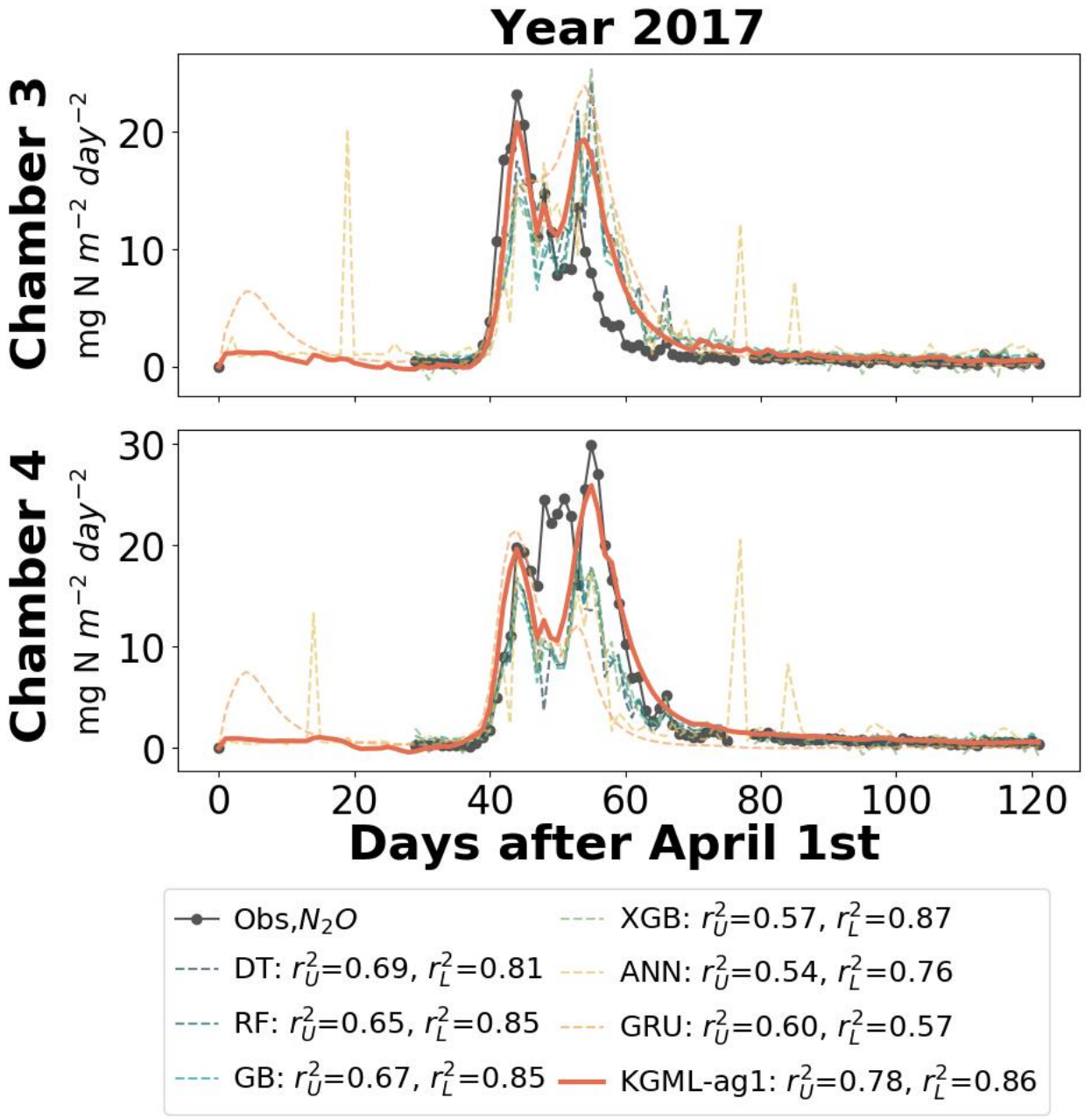


Figure S8: N_2O flux time series comparisons between KGML-ag1 predictions (red solid line), pure ML models (other colored dashed line) and observations (black-dot line) from cross-validation on two representative panels of chamber 3 and 4 in 2016. The r^2 value was calculated between observations and model simulations. r^2_U represents the r^2 value from upper panel (chamber 3) and r^2_L represents the r^2 value from lower panel (chamber 4).

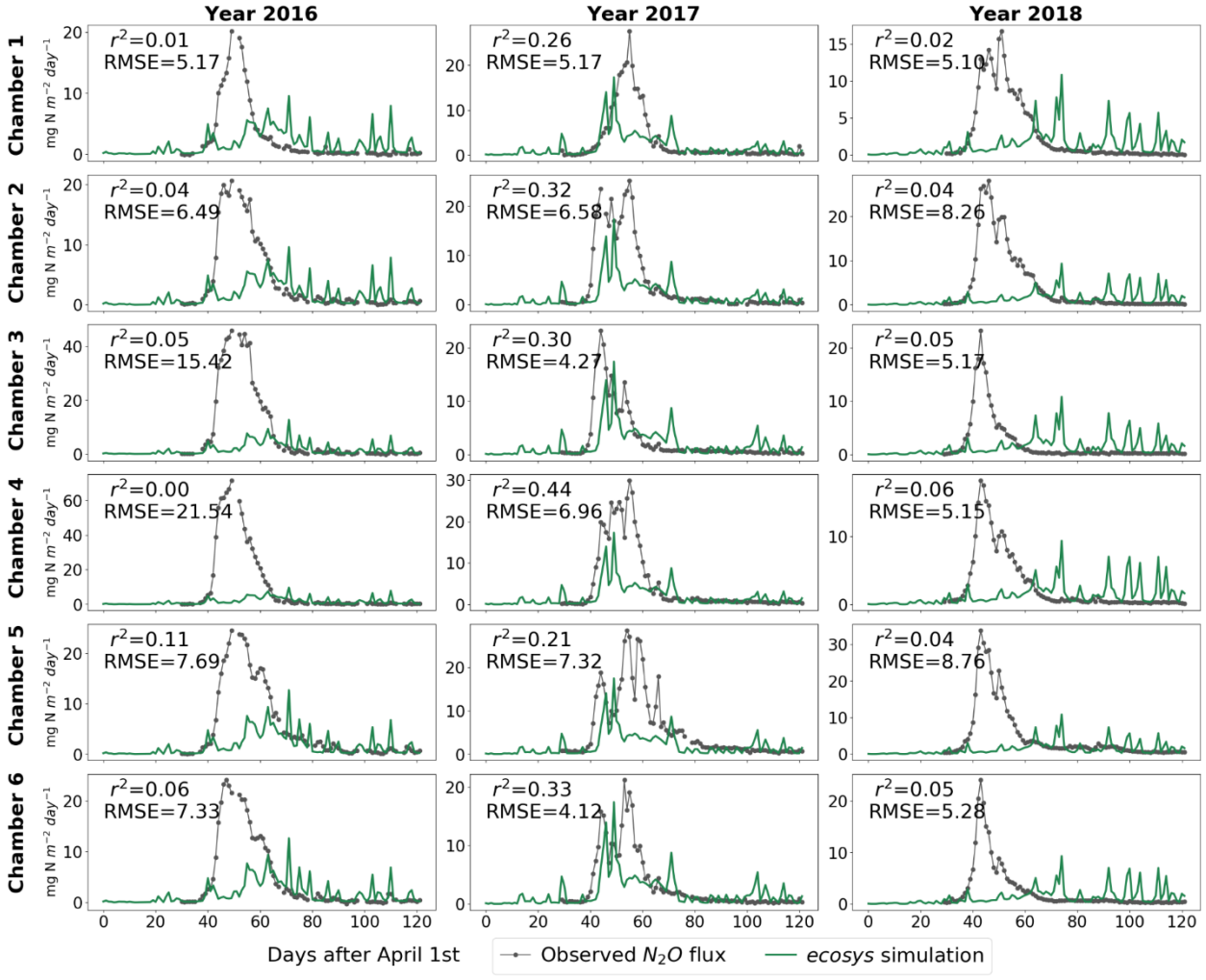


Figure S9: N_2O flux time series comparisons between *ecosys* simulations (green line) and observations (black-dot line).

45

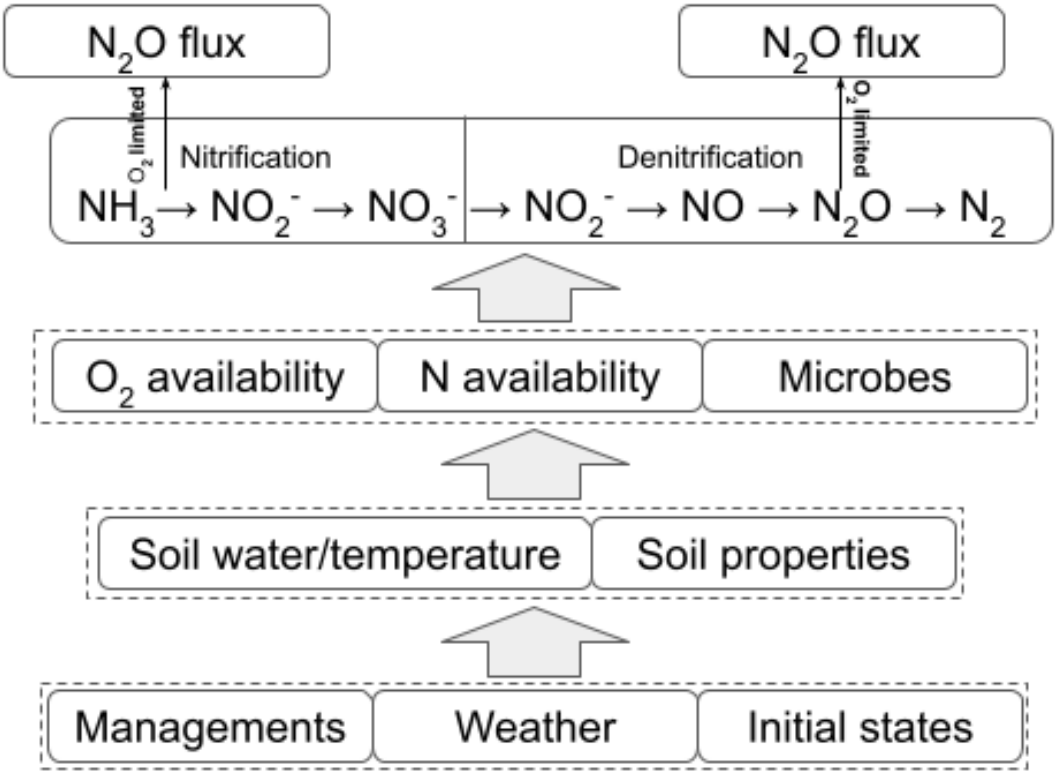


Figure S10: The simplified schema of N_2O flux related variables and processes.

Table S1: Variable short abbreviation, category (IMV represents intermediate variable, W represents weather forcing, FN represents the N fertilizer rate, SCP represents soil/crop property and target variable), description and units.

No.	Abbreviation	Variable categary	Descriptions	Units
1	RESIDUE_C	IMV	Total residue C on soil surface and in soil profile	g C m^{-2}
2	HUMUS_C	IMV	Total particulate + non-particulate C in soil profile	g C m^{-2}
3	LITTER_C	IMV	C in above + below-ground litterfall	g C m^{-2}
4	CO2_FLUX	IMV	CO_2 flux at the soil surface	g C m^{-2} day^{-1}
5	O2_FLUX	IMV	O_2 flux at the soil surface	g O_2 m^{-2} day^{-1}
6	AUTO_RESP	IMV	Below-ground autotrophic (root) respiration	g C m^{-2} day^{-1}
7	MICRO_C	IMV	Microbial C in all residue and humus complexes	g C m^{-2}
8	SURF_RES	IMV	Residue C on soil surface and in soil profile	g C m^{-2}
9	CH4_FLUX	IMV	CH_4 flux at the soil surface	g C m^{-2} day^{-1}
10	SURF_DOC_FLUX	IMV	Flux of organic C across all external surface boundaries in runoff and sediment	g C m^{-2} day^{-1}
11	SUBS_DOC_FLUX	IMV	Flux of organic C across all external subsurface boundaries in water dischage	g C m^{-2} day^{-1}
12	SURF_DIC_FLUX	IMV	Flux of inorganic C across all external surface boundaries in runoff and sediment	g C m^{-2} day^{-1}
13	SUBS_DIC_FLUX	IMV	Flux of inorganic C across all external subsurface boundaries in water dischage	g C m^{-2} day^{-1}
14	NBP	IMV	Net biome productivity	g C m^{-2} day^{-1}
15	SOC_1	IMV	Residue + humus C in soil layer 1, 5cm depth	g C m^{-2}
16	SOC_3	IMV	Residue + humus C in soil layer 3, 15cm depth	g C m^{-2}
17	SOC_5	IMV	Residue + humus C in soil layer 5, 28cm depth	g C m^{-2}
18	H2_FLUX	IMV	H_2 flux at the soil surface	g H_2 m^{-2} day^{-1}
19	ECO_HVST_C	IMV	C removed in harvest	g C m^{-2}
20	ECO_LAI	IMV	Leaf area index	m^2 m^{-2}
21	ECO_GPP	IMV	Gross primary productivity	g C m^{-2} day^{-1}
22	ECO_RA	IMV	Autotrophic respiration	g C m^{-2} day^{-1}
23	ECO_NPP	IMV	Net primary productivity	g C m^{-2} day^{-1}
24	ECO_RH	IMV	Heterotrophic respiration	g C m^{-2} day^{-1}
25	TTL_DIC	IMV	Total stocks of dissolved inorganic C	g C m^{-2}
26	ET	IMV	Evapotranspiration rate	mm day^{-1}
27	RUNOFF	IMV	Overland surface flow	mm day^{-1}
28	WATER	IMV	The total amount of water in the rooting zone of the soil profile	mm day^{-1}
29	DISCHG	IMV	Water discharge flux through all subsurface boundaries	mm
30	SNOWPACK	IMV	The equivalent water content of snow + ice + water in the snowpack	mm
31	VWC_1	IMV	The volumetric water content in soil layer 1, 5cm depth	m^3 m^{-3}
32	VWC_3	IMV	The volumetric water content in soil layer 3, 15cm depth	m^3 m^{-3}
33	VWC_5	IMV	The volumetric water content in soil layer 5, 28cm depth	m^3 m^{-3}

34	SURF_WTR	IMV	Near surface volumetric water content	$m^3\ m^{-3}$
35	ICE_1	IMV	The volumetric ice content in soil layer 1, 5cm depth	$m^3\ m^{-3}$
36	ICE_2	IMV	The volumetric ice content in soil layer 3, 15cm depth	$m^3\ m^{-3}$
37	ICE_3	IMV	The volumetric icecontent in soil layer 5, 28cm depth	$m^3\ m^{-3}$
38	PSI_1	IMV	The matric water potential in soil layer 1, 5cm depth	Mpa
39	PSI_3	IMV	The matric water potential in soil layer 3, 15cm depth	Mpa
40	PSI_5	IMV	The matric water potential in soil layer 5, 28cm depth	Mpa
41	WTR_TBL	IMV	Depth of the water table from the surface	m
42	RESIDUE_N	IMV	Total residue N on soil surface and in soil profile	$g\ N\ m^{-2}$
43	HUMUS_N	IMV	Total particulate + non-particulate N in soil profile	$g\ N\ m^{-2}$
44	FERTZR_N	FN	N fertilizer applied	$g\ N\ m^{-2}$
45	NET_PL_EXCH_N	IMV	Net N exchange between soil and plants	$g\ N\ m^{-2}\ day^{-1}$
46	NH4	IMV	Total $NH_4^+ + NH_3$ in the soil profile	$g\ N\ m^{-2}$
47	NO3	IMV	Total NO_3^- in soil profile	$g\ N\ m^{-2}$
48	SURF_DON_FLUX	IMV	Flux of organic N across all external surface boundaries in runoff and sediment	$g\ N\ m^{-2}\ day^{-1}$
49	SUBS_DON_FLUX	IMV	Flux of organic N across all external subsurface boundaries in water dischage	$g\ N\ m^{-2}\ day^{-1}$
50	SURF_DIN_FLUX	IMV	Flux of inorganic N across all external surface boundaries in runoff and sediment	$g\ N\ m^{-2}\ day^{-1}$
51	SUBS_DIN_FLUX	IMV	Flux of inorganic N across all external subsurface boundaries in water dischage	$g\ N\ m^{-2}\ day^{-1}$
52	N2O_FLUX	Target variable	N_2O flux at the soil surface	$g\ N\ m^{-2}\ day^{-1}$
53	NH3_FLUX	IMV	NH_3 flux at soil and plant surfaces	$g\ N\ m^{-2}\ day^{-1}$
54	N2_FIXN	IMV	Aerobic + anaerobic non-symbiotic N_2 fixation + symbiotic N_2 fixation	$g\ N\ m^{-2}\ day^{-1}$
55	MICRO_N	IMV	Total microbial N in all residue and humus complexes	$g\ N\ m^{-2}$
56	NH4_1	IMV	Total $NH_4^+ + NH_3$ concentration in soil layer 1, 5cm depth	$g\ N\ m^{-2}$
57	NH4_3	IMV	Total $NH_4^+ + NH_3$ concentration in soil layer 3, 15cm depth	$g\ N\ m^{-2}$
58	NH4_5	IMV	Total $NH_4^+ + NH_3$ concentration in soil layer 5, 28cm depth	$g\ N\ m^{-2}$
59	NO3_1	IMV	Total $NO_3^- + NO_2^-$ concentration in soil layer 1, 5cm depth	$g\ N\ m^{-2}$
60	NO3_3	IMV	Total $NO_3^- + NO_2^-$ concentration in soil layer 3, 15cm depth	$g\ N\ m^{-2}$
61	NO3_5	IMV	Total $NO_3^- + NO_2^-$ concentration in soil layer 5, 28cm depth	$g\ N\ m^{-2}$
62	NH4_RES	IMV	Residue $NH_4^+ + NH_3$ on soil surface and in soil profile	$g\ N\ m^{-2}$
63	NO3_RES	IMV	Residue $NO_3^- + NO_2^-$ on soil surface and in soil profile	$g\ N\ m^{-2}$
64	ECO_HVST_N	IMV	N removed in harvest	$g\ N\ m^{-2}\ day^{-1}$
65	N2_FLUX	IMV	N_2 flux at the soil surface	$g\ N\ m^{-2}\ day^{-1}$
66	RADN	W	Solar Radiation	$W\ m^{-2}$
67	TMAX_AIR	W	Max air temperature	^oC
68	TDIF_AIR	W	Difference between max and min air temperature	^oC
69	HMAX_AIR	W	Max humidity	fraction

70	HDIF_AIR	W	Difference between max and min humidity	fraction
71	WIND	W	Wind speed	$m\ s^{-1}$
72	PRECN	W	Precipitation	$mm\ day^{-1}$
73	TMAX_SOIL_1	IMV	The maximum temperature in soil layer 1, 5cm depth	^oC
74	TDIF_SOIL_1	IMV	The difference between max and min temperature temperature in soil layer 1 , 5cm depth	^oC
75	TMAX_SOIL_3	IMV	The maximum temperature in soil layer 3, 15cm depth	^oC
76	TDIF_SOIL_3	IMV	The difference between max and min temperature temperature in soil layer 3, 15cm depth	^oC
77	TMAX_SOIL_5	IMV	The maximum temperature in soil layer 5, 28cm depth	^oC
78	TDIF_SOIL_5	IMV	The difference between max and min temperature temperature in soil layer 5, 28cm depth	^oC
79	TMAX_LITTER	IMV	The maximum temperature in litter	^oC
80	TDIF_LITTER	IMV	The difference between max and min temperature temperature in litter	^oC
81	ECND_1	IMV	Electrical conductivity in soil layer 1, 5cm depth	$dS\ m^{-1}$
82	ECND_3	IMV	Electrical conductivity in soil layer 3, 15cm depth	$dS\ m^{-1}$
83	ECND_5	IMV	Electrical conductivity in soil layer 5, 28cm depth	$dS\ m^{-1}$
84	TTL_SALT_DISCHG	IMV	Total salt discharge through water through all subsurface boundaries	$g\ Mg^{-1}\ day^{-1}$
85	PDOY	SCP	Plant day of the year	day
86	CROPT	SCP	Crop type, 1 for corn and 0 for soybean	unitless
87	TBKDS	SCP	Depth weighted averaged bulk density in soil profile	$Mg\ m^{-3}$
88	TCSAND	SCP	Depth weighted averaged sand content in soil profile	$g\ kg^{-1}$
89	TCSILT	SCP	Depth weighted averaged silt content in soil profile	$g\ kg^{-1}$
90	TPH	SCP	Depth weighted averagedpH in soil profile	unitless
91	TCEC	SCP	Depth weighted averaged cmol+ kg-1 in soil profile	$cmol^{-1}\ kg^{-1}$
92	TSOC	SCP	Depth weighted averaged soil organic carbon in soil profile	$g\ C\ kg^{-1}$

Table S2: N_2O prediction accuracy comparisons between LSTM and GRU models on synthetic data, with different combinations of IMVs (+9 or +58IMVs) and different sliding sliding window settings during training (e.g. 2y1y represent window size is 2 years and the window move 1 year after 1 iteration). Training Efficiency is also compared between LSTM and GRU models for the first two experiments, with changing the training counties = 3, 10, 30, 70, validation counties = 1, 2, 5, 10, and batch size (county numbers input in each iteration) = 1, 5, 5, 5, 5.

	N_2O prediction accuracy		Training effeciency			
Experiment settings	Test r^2	Test RMSE	Train=3, val=1, batch =1	Train=10, val=2, batch =5	Train=30, val=5, batch =5	Train=70, val=10, batch =5
LSTM+9IMVs+1y1y	0.74	1.32	3.8s	3.3s	9.2s	22s
GRU+9IMVs+1y1y	0.81	1.08	3.5s	2.7s	7.2s	17s
LSTM+58IMVs+1y1y	0.91	0.6				
GRU+58IMVs+1y1y	0.92	0.59				
LSTM+58IMVs+2y2y	0.86	0.76				
GRU+58IMVs+2y2y	0.9	0.66				
LSTM+58IMVs+2y1y	0.89	0.67				
GRU+58IMVs+2y1y	0.91	0.6				