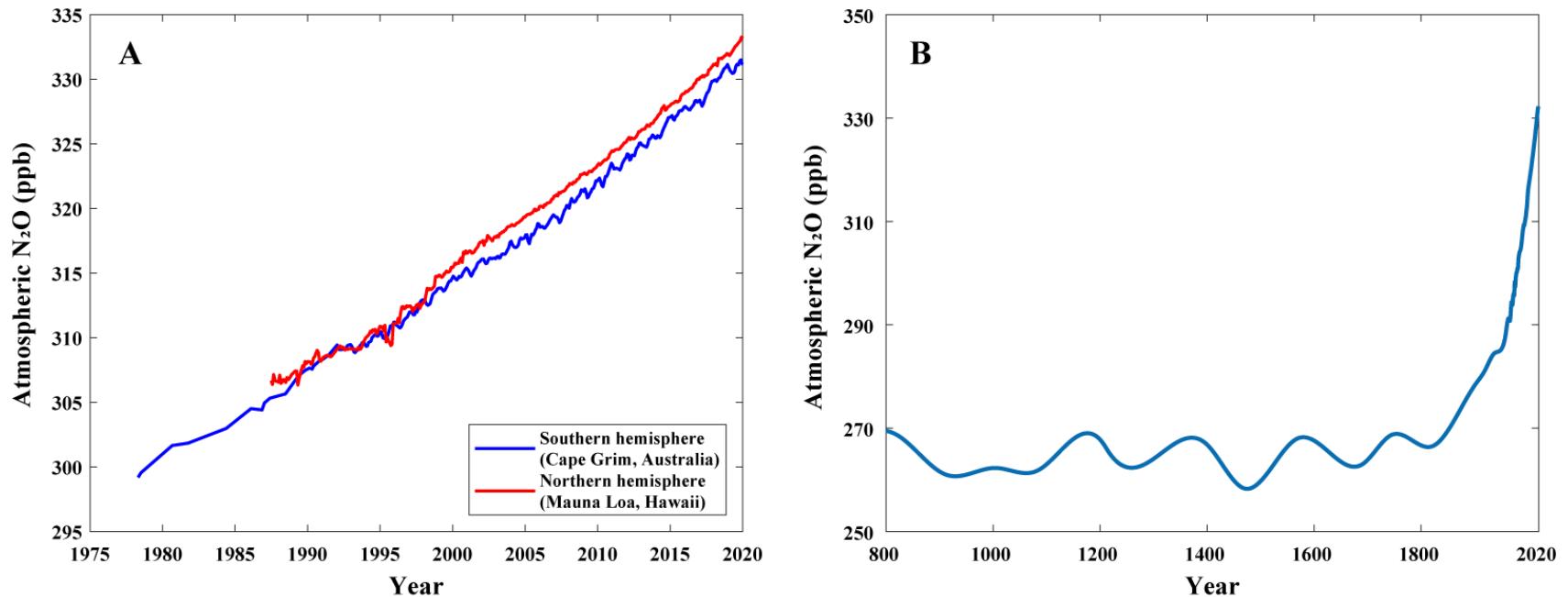
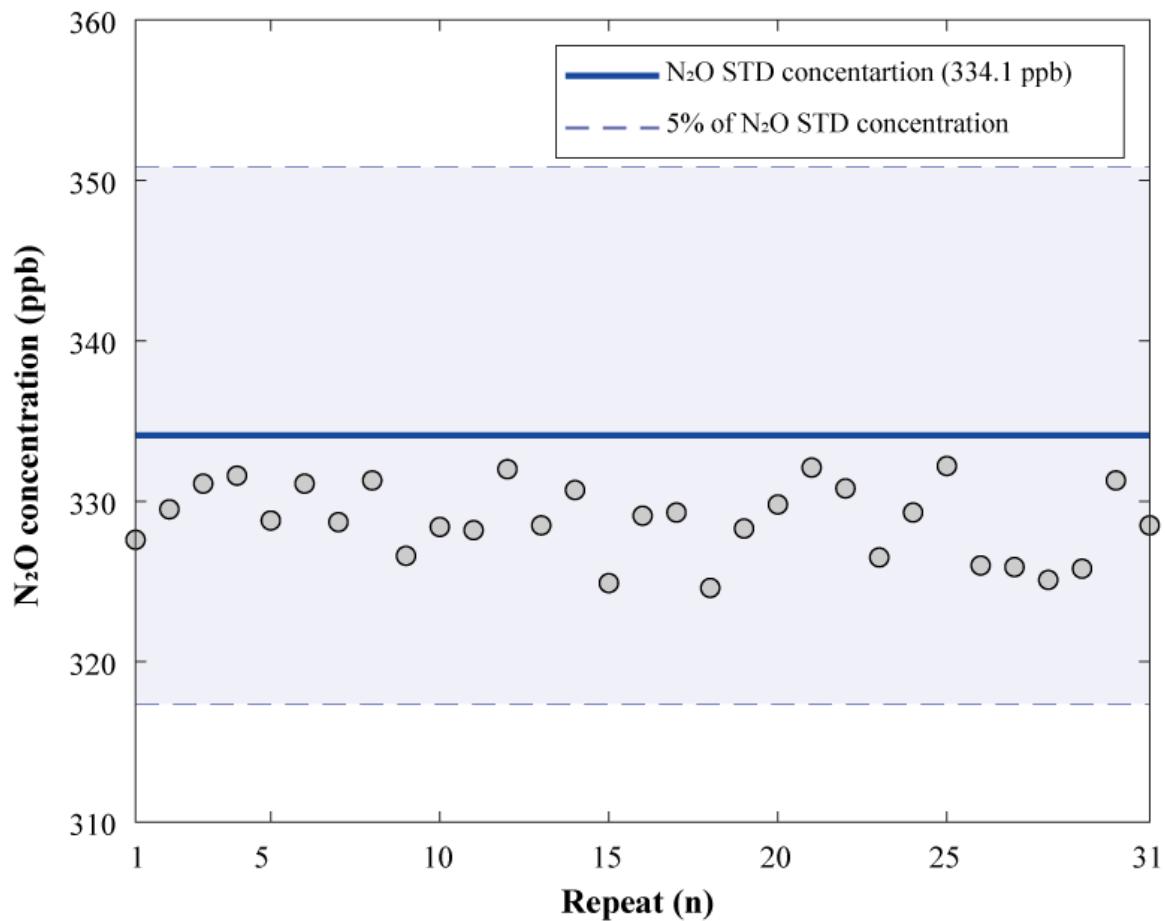


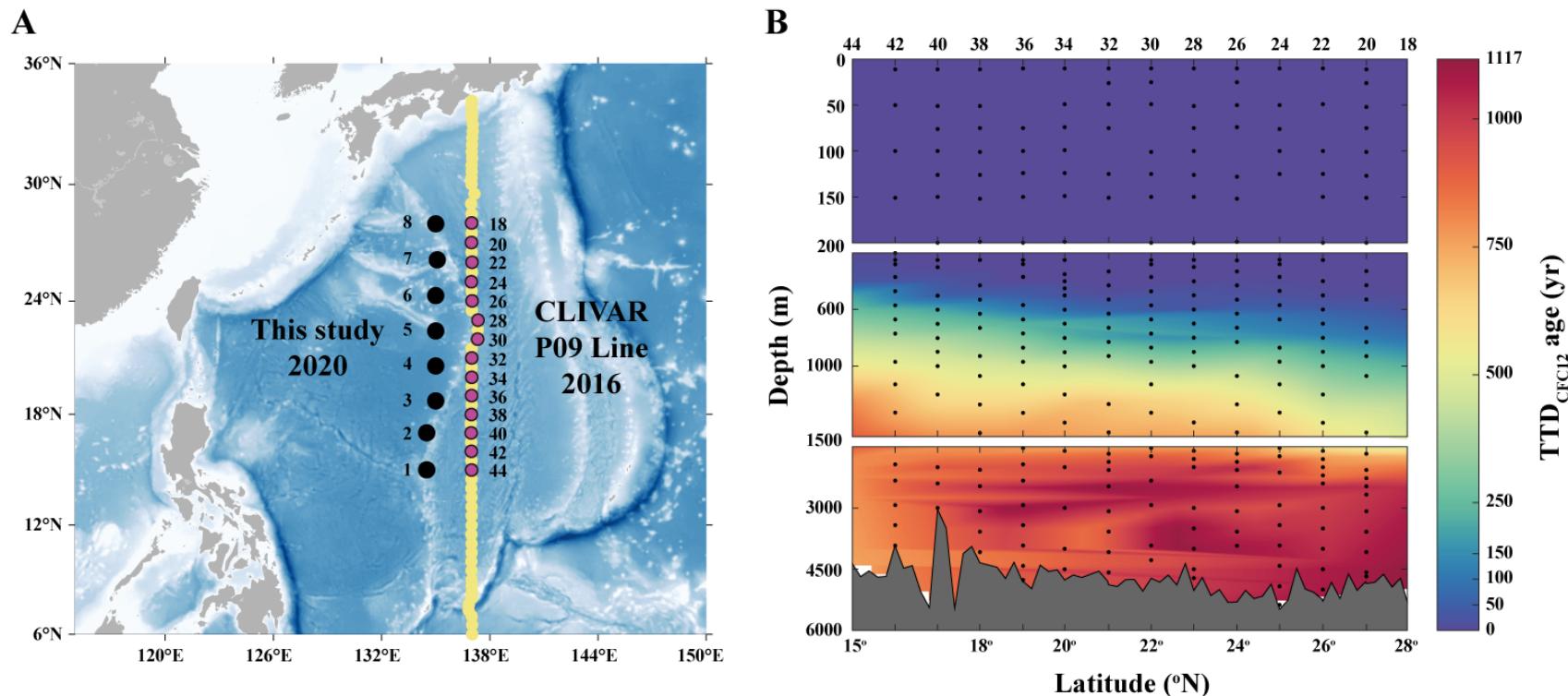
## Supporting Information



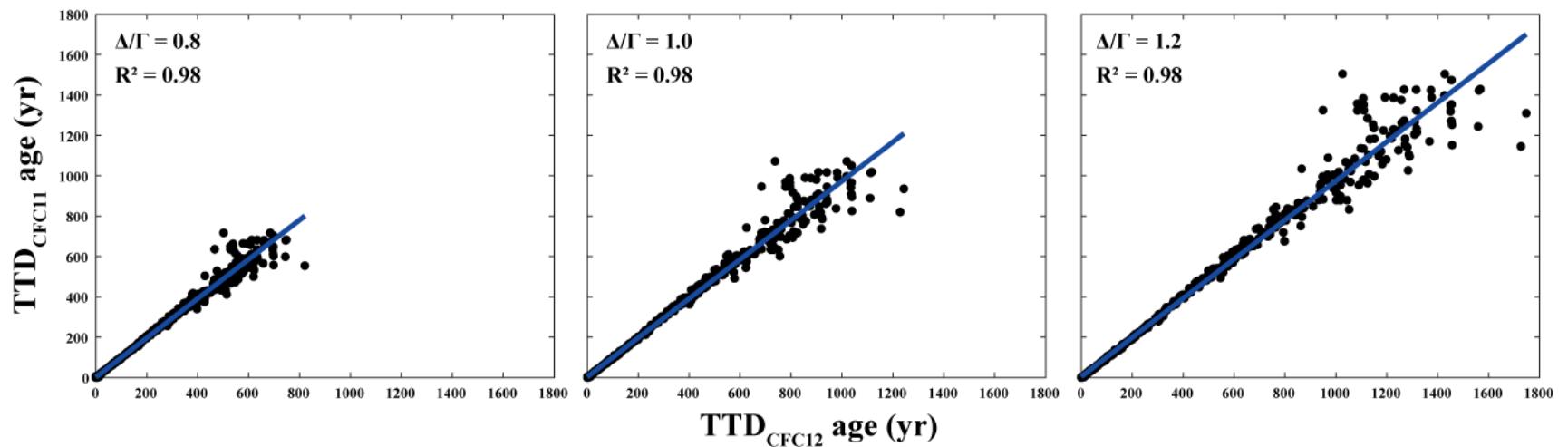
**Figure S1.** (A) Comparison of atmospheric N<sub>2</sub>O concentrations in the Northern and Southern Hemispheres. From 1987 to 2020, the mean difference in N<sub>2</sub>O concentration between the two locations was  $1.13 \pm 0.63$  ppb. (B) The line chart depicts the history of interpolated atmospheric N<sub>2</sub>O levels. The sources of the dataset are given in section 2.2.



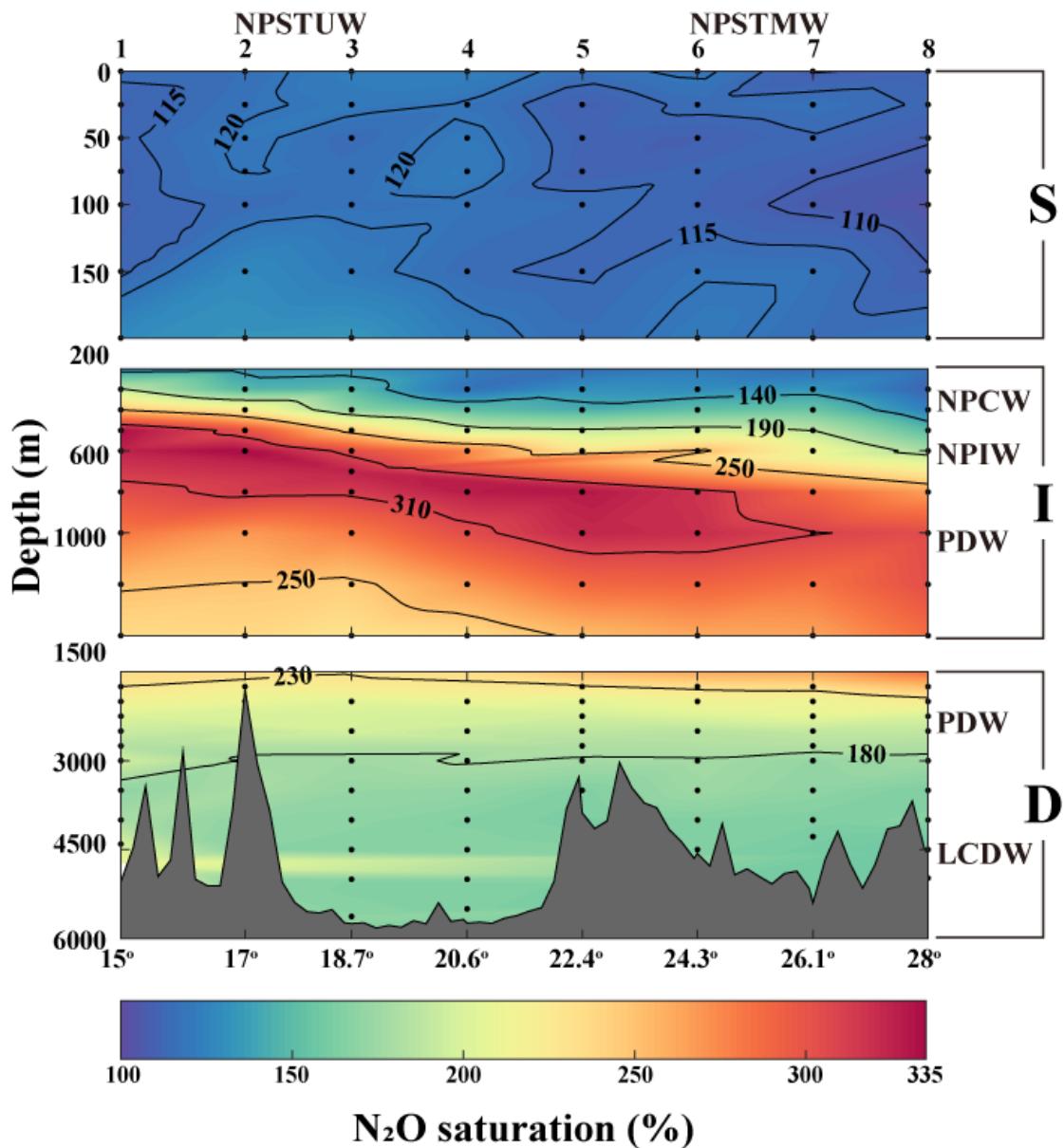
**Figure S2.** N<sub>2</sub>O standard gas measurements (gray circles). Blue solid line represents a certified value of standard gas as 334.1 ppb. Dotted lines represent ±5 % levels of the certified value.



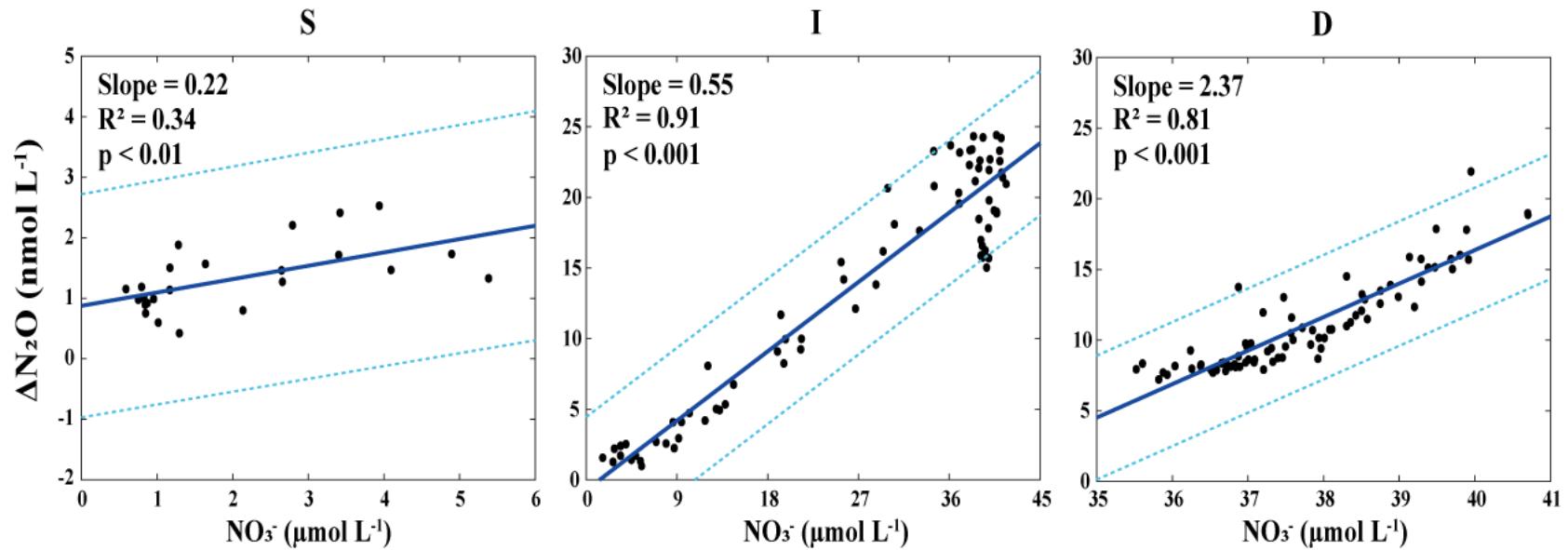
**Figure S3.** The map of the study area, CLIVAR P09 cruise (2016), and the vertical distribution of TTD<sub>CFC12</sub> age during the CLIVAR P09 cruise. **(A)** Black dots represent stations where sampling for this study was carried out, and yellow/purple dots indicate the sampling stations of the P09 cruise. The CFCs data set was obtained from CLIVAR stations colored in purple. **(B)** The vertical distribution of TTD<sub>CFC12</sub> age is colored with blue to red gradients. Black dots represent sampling depth, and the upper part of the *x*-axis represents station numbers of the P09 cruise.



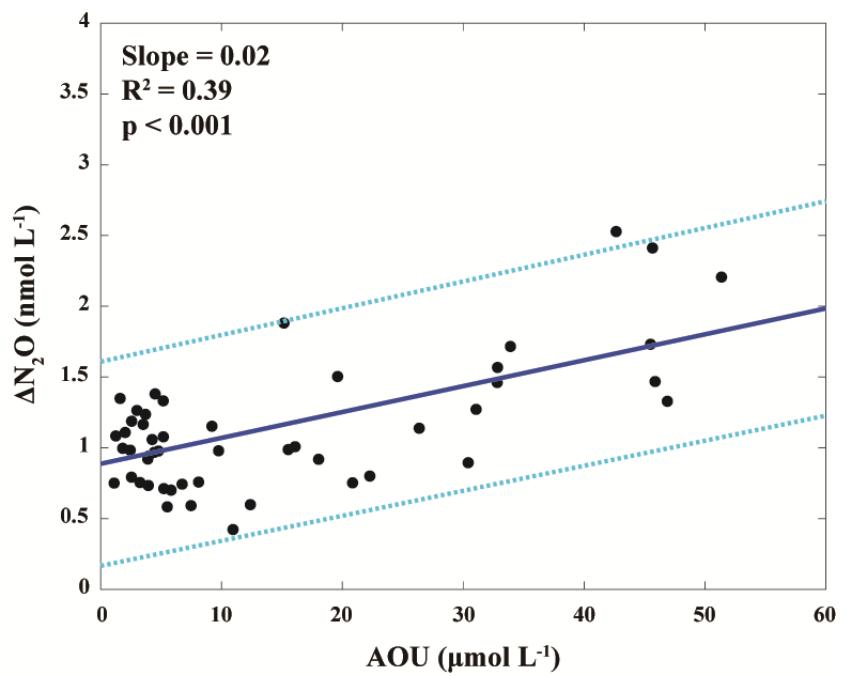
**Figure S4.** The correlations between  $\text{TTD}_{\text{CFC11}}$  and  $\text{TTD}_{\text{CFC12}}$  ages along three  $\Delta/\Gamma$  ratios (0.8, 1.0, and 1.2). The solid lines represent the 1:1 regression line.



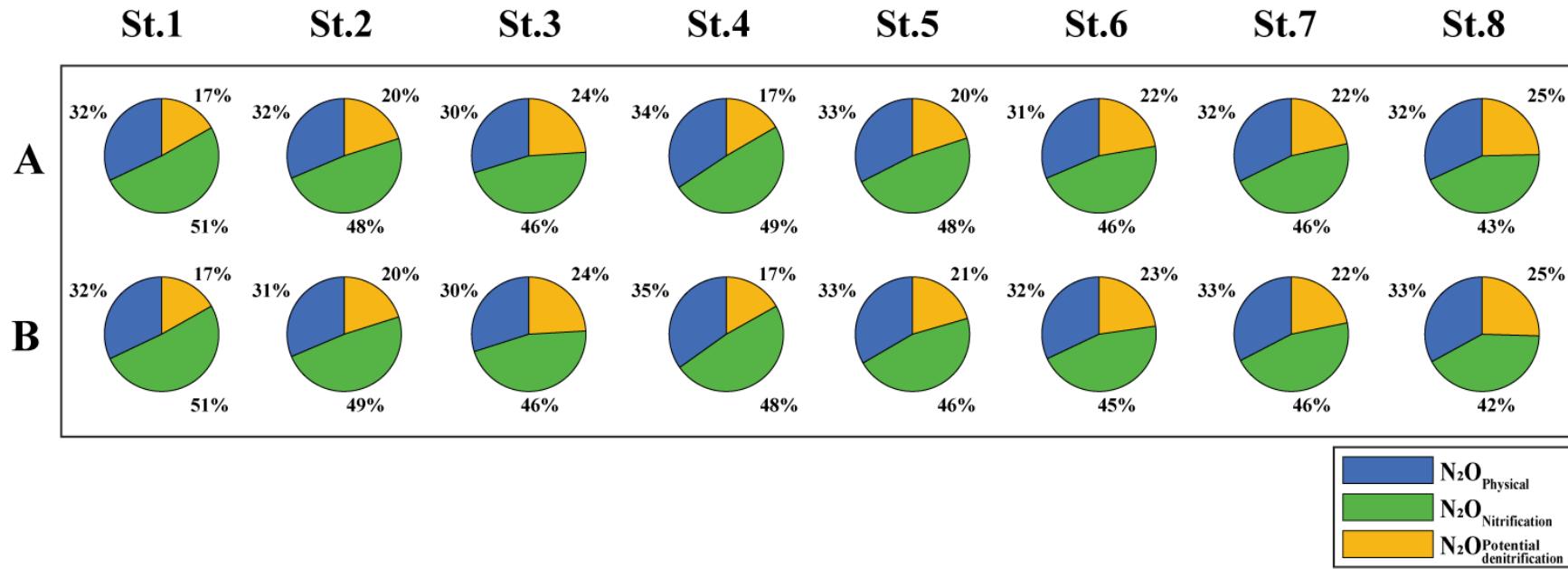
**Figure S5.** Vertical profiling of  $\text{N}_2\text{O}$  saturation in different layers: surface (0–200 m), intermediate (200–1500 m), and deep layers (1500–5774 m) of the STWNPO during this investigation. The black solid lines represent the contour lines and the abbreviations of water masses indicate the distribution of water masses. The top and bottom of the  $x$ -axis represent the station numbers and latitudes, respectively.



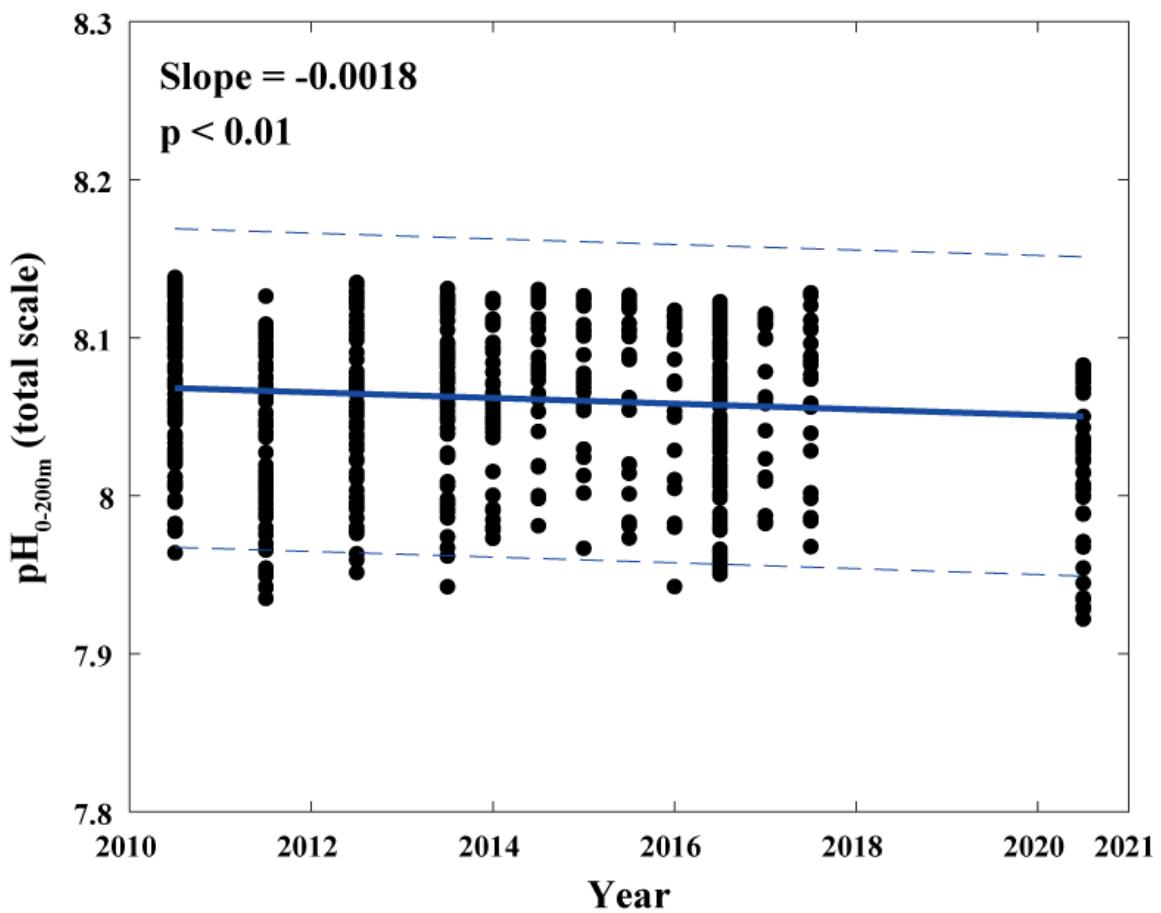
**Figure S6.** The correlations of  $\Delta\text{N}_2\text{O}$  with  $\text{NO}_3^-$  in each layer (surface, intermediate, and deep) of the STWNPO. The blue and cyan lines represent the linear regression and the 95% prediction interval respectively.



**Figure S7.** Correlations of  $\Delta\text{N}_2\text{O}$  with AOU at the surface layer (0–200 m). The blue and cyan lines represents the linear regression and the 95% prediction interval respectively.



**Figure S8.** The estimated mean fractions of  $\text{N}_2\text{O}$  production derived from physical ( $\text{N}_2\text{O}_{\text{eq}}$ ) and biogeochemical ( $\text{N}_2\text{O}_{\text{Nitrification}}$  and  $\text{N}_2\text{O}_{\text{denitrification}}$ ) processes in the intermediate layer of the STWNPO during this study.  $\text{N}_2\text{O}_{\text{Nitrification}}$  was estimated using the slope of  $\Delta\text{N}_2\text{O}/\text{AOU}$  (**A**) and  $\Delta\text{N}_2\text{O}/\text{NO}_3^-$  (**B**). Given the error of  $r_{\Delta\text{N}_2\text{O}:\text{NO}_3^-}$ , the error for estimating  $\text{N}_2\text{O}_{\text{Nitrification}}$  was  $\pm 1.8\%$ .



**Figure S9.** The temporal pH trend in the surface layer (0–200 m) of the STWNPO. The historic data were obtained from the stations indicated on the CLIVAR P09 transect line (refer to purple dots in Supplementary Fig. S3). The decreasing rate of surface pH was  $-0.0018 \text{ yr}^{-1}$  ( $p < 0.01$ ).

**Table S1.** The  $\Delta/\Gamma$  ratio in various environments.

Region	$\Delta/\Gamma$ Ratio	Tracer	References
Global	1	CFC12	Waugh et al. (2006)
North Atlantic (Subpolar)	1	CFCs, tritium, helium	Waugh et al. (2004)
Arctic, Nordic Seas, North Atlantic	0.8		
North Pacific, Southern Ocean	1.2	CFCs, SF6	He et al. (2018)
elsewhere	1		
North pacific (directly ventilated)	<1.0		
North pacific (ventilated by physical mixing)	>1.0	CFC11, SF6	Sonnerup et al. (2013)
Nordic Seas	0.5~1.5 (probable 1.0)	CFC11, CFC12	Olsen et al. (2010)
Northern South China Sea, Western North Pacific	0.8	CFC12, SF6	Wang et al. (2021)
Indian Ocean	1	CFC11, CFC12	Alvarez et al. (2009)
Global	1	CFC12, SF6	Shao et al. (2016)
South Pacific	0.6	CFC11, CFC12, SF6	Kim et al. (2013)

**Table S2.** The ranges and averages of excess N<sub>2</sub>O differences from the TTD<sub>CFC12</sub> ages of three  $\Delta/\Gamma$  ratios (i.e., 0.8, 1.0, and 1.2 [ $\Delta N_2O_{0.8}$ ,  $\Delta N_2O_{1.0}$ , and  $\Delta N_2O_{1.2}$ ]) and unmodified excess N<sub>2</sub>O ( $\Delta N_2O_{\text{Unmodified}}$ ).

Station	$\Delta N_2O_{1.0}-\Delta N_2O_{0.8}$	$\Delta N_2O_{1.0}-\Delta N_2O_{1.2}$	$\Delta N_2O_{1.0}-\Delta N_2O_{\text{Unmodified}}$
	(nmol L <sup>-1</sup> )		
1	-0.17 – 0.13 (0.06 ± 0.05)	-0.13 – 0.19 (0.05 ± 0.05)	0 – 2.43 (1.43 ± 1.11)
2	-0.17 – 0.28 (0.06 ± 0.07)	-0.24 – 0.17 (0.08 ± 0.08)	-0.26 – 2.39 (1.35 ± 1.11)
3	-0.18 – 0.26 (0.06 ± 0.07)	-0.40 – 0.14 (0.12 ± 0.12)	-0.34 – 2.40 (1.27 ± 1.10)
4	-0.17 – 0.18 (0.05 ± 0.06)	-0.39 – 0.07 (0.08 ± 0.10)	-0.30 – 2.47 (1.26 ± 1.15)
5	-0.18 – 0.37 (0.08 ± 0.09)	-0.40 – 0.33 (0.12 ± 0.15)	-0.30 – 2.46 (1.25 ± 1.15)
6	-0.08 – 0.31 (0.09 ± 0.09)	-0.39 – 0.10 (0.08 ± 0.11)	-0.26 – 2.37 (1.23 ± 1.12)
7	-0.08 – 0.23 (0.07 ± 0.07)	-0.24 – 0.19 (0.07 ± 0.08)	-0.28 – 2.50 (1.26 ± 1.19)
8	-0.19 – 0.20 (0.07 ± 0.08)	-0.40 – 0.06 (0.12 ± 0.15)	-0.26 – 2.47 (1.18 ± 1.15)
Mean	0.07 ± 0.07	0.09 ± 0.11	1.28 ± 1.12

**Table S3.** The water masses identified during this study. The values in parentheses are the values observed in this study.  $\theta$ , S, and  $\sigma_\theta$  refer to potential temperature, salinity, and potential density anomaly, respectively.

Water Mass	$\theta$ (°C)	S (psu)	$\sigma_\theta$ (kg m <sup>-3</sup> )	Layer	Characteristics	References
NPSTMW	16 – 20	34.6 – 34.8	25.0 – 25.6	Surface	Relatively high DO (~205.71 μmol L <sup>-1</sup> )	Hanawa and Talley (2001) Oka (2009) Rainville et al. (2014)
NPSTUW	19 – 25	34.6 – 35.4	23.0 – 25.0	Surface	Relatively low DO (~195.11 μmol L <sup>-1</sup> )	Suga et al. (2000) O'Connor et al. (2002) Behrens et al. (2018)
NPCW	10 – 22	34.2 – 35.2	25.2 – 26.4	Intermediate	Total alkalinity Minimum	Emery (2001) Behrens et al. (2018)
NPIW	5 – 12	34.0 – 34.3	26.6 – 27.4	Intermediate	Salinity Minimum (~34.15 psu)	Talley (1993) Behrens et al. (2018)
PDW	1.4 – 6.4	33.4 – 34.7	27.6 – 27.8	Deep	Oxygen Minimum (~55 μmol L <sup>-1</sup> ), Nutrient Maximum (NO <sub>3</sub> <sup>-</sup> : ~41.6 μmol L <sup>-1</sup> ), N* Minimum (-7.3 μmol L <sup>-1</sup> )	Amakawa et al. (2009) Talley (2011) Behrens et al. (2018)
LCDW	0.98 – 1.2	34.6 – 37.4	27.7 – 28.2	Deep	Low Silicate, High Salinity (~34.68 psu)	Emery (2001) Talley (2011)

**Table S4.** The slopes of  $\Delta\text{N}_2\text{O}/\text{AOU}$  and  $\Delta\text{N}_2\text{O}/\text{NO}_3^-$  in various ocean environments.

Region	Slope $_{\Delta\text{N}_2\text{O}/\text{AOU}}$ (mol N <sub>2</sub> O/mol O <sub>2</sub> )	Slope $_{\Delta\text{N}_2\text{O}/\text{NO}_3^-}$ (mol N <sub>2</sub> O/mol NO <sub>3</sub> <sup>-</sup> )	Depth (m)	References
Western Pacific, Indian Ocean	0.125 + 0.00993T	—	0 – 5900	Butler et al. (1989)
Tropical Atlantic	0.121 (central) 0.106 (eastern)	—	< 300	Forster et al. (2009)
Eastern Tropical South Pacific (O <sub>2</sub> > 50 μM)	0.13	—	< 350	Kock et al. (2016)
Eastern Tropical Pacific, Arabian Sea	0.2 – 0.3	—	σ <sub>θ</sub> = 26.9	
South Atlantic, South Indian	0.01 – 0.05	—	σ <sub>θ</sub> = 26.9	
Indian Ocean (0–5°N)	>0.22	—	200 – 300	Nevison et al. (2003)
Western Pacific (0–10°N)	>0.22	—	300	
Atlantic (20°N)	>0.14	—	300	
	0.0473 (subtropical)	0.2497	< 1000	
North Atlantic	0.0785 (tropical) 0.0942 (tropical)	0.4848 0.7379	< 500 > 500	Walter et al. (2006)
Western Arctic Ocean	0.039	0.22	75 – 150	Zhang et al. (2015)
South Pacific Subtropical Gyre	0.176 (central) 0.142 (eastern)	0.403 (AOU/NO <sub>3</sub> <sup>-</sup> ) 0.188 (AOU/NO <sub>3</sub> <sup>-</sup> )	200 – 600	Charpentier et al. (2007)
Gulf of Mexico	0.048, 0.096	—	0 – 32	Walker et al. (2010)
East China Sea, South Yellow Sea	0.0015 – 0.12	0.021 – 1.16	0 – 800	Chen et al. (2021)
	0.02	0.22	0 – 200	
Western North Pacific	0.09	0.55	200 – 1500	This study
	0.16	2.37	1500 – 5774	

**Table S5.** The slopes of  $\Delta\text{N}_2\text{O}/\text{AOU}$  and  $\Delta\text{N}_2\text{O}/\text{N}^*$  of the intermediate layer, NPIW, and PDW in the deep layer

	$\Delta\text{N}_2\text{O}/\text{AOU}$		$\Delta\text{N}_2\text{O}/\text{N}^*$	
	Slope	p value	Slope	p value
<b>Intermediate layer</b>	0.09	< 0.001	-2.32	< 0.001
<b>NPIW</b>	0.11	< 0.001	-1.35	< 0.05
<b>PDW in deep layer</b>	0.16	< 0.001	-0.84	< 0.001

**Table S6.** Observed wind speeds at the height of 29 m ( $U_{29}$ ) and estimated wind speeds at the height of 10 m ( $U_{10}$ ) during this investigation.

Station	Year	Month	Date	Time	Observed $U_{29}$ ( $\text{m s}^{-1}$ )	Estimated $U_{10}$ ( $\text{m s}^{-1}$ )
1	2020	5	27	13:13 – 13:43	6.72	6.15
2	2020	5	30	15:28 – 15:58	5.51	5.04
3	2020	5	31	08:53 – 09:23	4.74	4.34
4	2020	6	1	00:43 – 01:13	5.38	4.92
5	2020	6	2	00:53 – 01:23	3.85	3.52
6	2020	6	2	19:10 – 19:40	6.94	6.36
7	2020	6	3	11:45 – 12:15	7.27	6.65
8	2020	6	4	08:44 – 09:14	13.59	12.44

**Table S7.** Estimated N<sub>2</sub>O fluxes using three  $k_w$  models during this investigation of the STWNPO. W&M<sub>1999</sub>, N<sub>2000</sub>, and W<sub>2014</sub> refer to the kw models of Wanninkhof and McGillis (1999), Nightingale et al. (2000), and Wanninkhof (2014), respectively.

Station	N <sub>2</sub> O Flux ( $\mu\text{mol m}^{-2} \text{d}^{-1}$ )				
	W&M <sub>1999</sub>	N <sub>2000</sub>	W <sub>2014</sub>	Mean	STD
1	1.3	2.0	1.9	1.8	0.4
2	0.8	1.5	1.4	1.2	0.4
3	0.7	1.7	1.5	1.3	0.5
4	1.2	2.3	2.1	1.8	0.6
5	0.3	0.8	0.7	0.6	0.3
6	2.1	3.0	2.9	2.7	0.5
7	0.8	1.0	1.0	0.9	0.2
8	7.1	4.8	5.1	5.6	1.3
<b>Mean</b>			2.0	0.3	

**Table S8.** N<sub>2</sub>O fluxes of various regions.

Region	N <sub>2</sub> O <sub>Flux</sub> (μmol m <sup>-2</sup> d <sup>-1</sup> )	References
<b>Pacific</b>		
East China Sea	6.9 – 7.0	Zhang et al. (2019)
Northeastern coast of Australia	-0.2 ± 0.1	Reading et al. (2021)
Yellow Sea	0.2 – 7.7	Chen et al. (2021)
Okhotsk Sea	~2.5	Zhan et al. (2021)
Northeast subarctic Pacific	~12.5 12.7 – 30.7	Fenwick and Tortell (2018) Farías et al. (2009)
Eastern tropical South Pacific	27 – 1825 23 – 108	Arevalo-Martínez et al. (2015) Ji et al. (2019)
Subtropical North Pacific (Hawaii)	< 0.4 ± 0.2 0.04 – 0.13	Dore et al. (1998) Butler et al. (1989)
Western North Pacific (subtropical)	0.7 – 0.9 ~0.5 0.3 – 7.1	Breider et al. (2015) Yoshikawa et al. (2016) This Study
Western North Pacific (subarctic)	1.3 – 2.4 ~6.3	Breider et al. (2015) Yoshikawa et al. (2016)
<b>Indian</b>		
Indian Shelf	-1.2 – 3243	Naqvi et al. (2006)
<b>Atlantic</b>		
Nordic Sea	-4.4 ± 0.4	Zhan et al. (2016)
Southern Iberian Atlantic Basin	3.6 ± 13.7	Sierra et al. (2020)
Eastern subtropical South Atlantic	52.7 ± 43.2 ~46	Morgan et al. (2019) Frame et al. (2014)
Gulf of Mexico	3.2 – 5.2	Walker et al. (2010)
<b>Black Sea</b>		
Northwest shelf	1.6 – 4.4	Amouroux et al. (2002)
Deep basin	3.1 – 5.2	
<b>Arctic</b>		
Southern Chukchi Sea	2.3 ± 2.7	Heo et al. (2021)
Northern Chukchi Sea	-1.3 ± 1.5	
<b>Antarctic</b>		
Antarctic peninsula	-3.65 ± 0.95	Zhan et al. (2015)

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