N$_2$O dynamics in the western Arctic Ocean during the summer of 2017

Jang-Mu Heo$^1$, Seong-Su Kim$^1$, Sung-Ho Kang$^2$, Eun Jin Yang$^2$, Ki-Tae Park$^2$, Jin Young Jung$^2$, Kyung-Ho Cho$^2$, Ju-Hyoung Kim$^3$, Alison M. Macdonald$^4$, Joo-Eun Yoon$^1$, Hyo-Ryeon Kim$^1$, Sang-Min Eom$^1$, Jae-Hyun Lim$^5$ & Il-Nam Kim$^{1,2}$

The western Arctic Ocean (WAO) has experienced increased heat transport into the region, sea-ice reduction, and changes to the WAO nitrous oxide (N$_2$O) cycles from greenhouse gases. We investigated WAO N$_2$O dynamics through an intensive and precise N$_2$O survey during the open-water season of summer 2017. The effects of physical processes (i.e., solubility and advection) were dominant in both the surface (0–50 m) and deep layers (200–2200 m) of the northern Chukchi Sea with an under-saturation of N$_2$O. By contrast, both the surface layer (0–50 m) of the southern Chukchi Sea and the intermediate (50–200 m) layer of the northern Chukchi Sea were significantly influenced by biogeochemically derived N$_2$O production (i.e., through nitrification), with N$_2$O over-saturation. During summer 2017, the southern region acted as a source of atmospheric N$_2$O (mean: +2.3 ± 2.7 μmol N$_2$O m$^{-2}$ day$^{-1}$), whereas the northern region acted as a sink (mean –1.3 ± 1.5 μmol N$_2$O m$^{-2}$ day$^{-1}$). If Arctic environmental changes continue to accelerate and consequently drive the productivity of the Arctic Ocean, the WAO may become a N$_2$O “hot spot”, and therefore, a key region requiring continued observations to both understand N$_2$O dynamics and possibly predict their future changes.

Arctic air temperatures have dramatically increased over the past two decades. This strong warming is frequently referred to as “Arctic Amplification” and is one of the main results of rapidly increasing concentrations of greenhouse gases in the atmosphere$^{1-4}$. Consequently, the extent of annual sea ice has rapidly decreased since the 1980s$^{5-7}$, implying that we may face a sea-ice-free Arctic summer in the near future$^8$. The western Arctic Ocean (WAO) has also experienced rapid environmental changes, such as increased heat transport and sea-ice reduction$^{9,10}$.

The WAO is geographically composed of the Chukchi, East Siberian, and Beaufort Seas, the Canadian Arctic Archipelago, and the Canada Basin (Fig. 1a). During the summer season, latitudinal differences in both physical and biogeochemical features have been clearly determined from the Bering Strait to the Chukchi Borderland$^{11-13}$. The southern region (i.e., the area extending from the Bering Strait to the Chukchi Shelf) is relatively warm, saline, and eutrophic due to the presence of Pacific waters that enter the WAO, bringing heat and nutrients with them. As a result, this region is one of the most productive stretches of ocean in the world$^{14,15}$. By contrast, the northern region, extending from the Chukchi Borderland to the Canada Basin, is mainly affected by freshwater originating from sea ice melt and rivers, and is therefore relatively cold, fresh, and oligotrophic. It is also important to note that latitudinal variations in the distribution of bacterial communities within the WAO can be caused by both physical and biogeochemical factors$^{12}$. These environmental variations are therefore extremely relevant to nitrous oxide (N$_2$O) dynamics.

N$_2$O production through marine nitrogen cycle processes (i.e., nitrification and denitrification) is directly linked to climate change, contributing to both the greenhouse effect and ozone depletion$^{16,17}$. Although it has also been reported that a substantial portion of the N$_2$O budget may be effluxed from the global ocean (4.4 ± 0.7 Tg N year$^{-1}$), N$_2$O data are still limited, particularly over much of the Arctic Ocean$^{18}$. That said, several N$_2$O investigations in the WAO have previously been conducted and have provided important information regarding N$_2$O dynamics$^{13,19-25}$. In particular, high N$_2$O concentrations were observed in the Chukchi Sea shelf region, whereas most of the under-saturated N$_2$O was found in the high-latitude regions of the WAO. However,
prior to the study presented here, no intensive investigation synthesizing information on the dynamics of WAO N₂O (i.e., distributions of the concentration and flux and their controlling environmental factors) in the water column (from the surface to the bottom) had been conducted.

The concentration of N₂O in the atmosphere has been steadily increasing since pre-industrial times, and because the points in time at which the N₂O concentration of water parcels were in equilibrium with the atmosphere prior to being ventilated are all different, it is necessary to calculate equilibrium N₂O concentration to accurately estimate the amount of N₂O production. Although there have been a few studies attempting to accurately calculate the N₂O concentration of the water column, the N₂O concentration of the water column in other studies has been calculated from the contemporary atmospheric N₂O concentration. Hence, in this study, the equilibrium N₂O concentration was calculated using a tracer gas.

Based on an intensive and precise N₂O survey of the WAO water column during the open-water season of summer 2017, we (1) present spatial distributions of N₂O concentrations and fluxes, (2) identify physical and/or biogeochemical factors controlling the distributions, (3) determine whether the WAO is a source or sink for atmospheric N₂O, and (4) speculate regarding future changes in the WAO N₂O flux in response to rapid Arctic climate changes.

Materials and methods

Sampling and measurement of basic physical/biogeochemical parameters. In August 2017, the icebreaker R/V Araon collected physical and biogeochemical samples from 30 WAO stations from the Bering Strait to the Chukchi Borderland (Fig. 1a). At each sampling location, the vertical profiles of the potential temperature (θ), salinity (S), and dissolved oxygen (DO) were measured using a conductivity-temperature-depth instrument (CTD; SBE 911 plus, Sea-Bird Electronics, Inc., USA). The CTD temperature and conductivity-salinity diagram with N² information (blue to red gradient color bar) were used to determine the N₂O concentration of the water column. Vertical distributions of (b) potential temperature (°C), and (c) salinity (psu) along a latitudinal transect from the Bering Strait to the Chukchi Borderland (black solid line shown in a). (d) Potential temperature–salinity diagram with N² information (blue to red gradient color bar); vertical distributions of (ε) dissolved oxygen (μmol L⁻¹) and (f) dissolved inorganic nitrogen (DIN; μmol L⁻¹). The inset in (f) shows the DIN inventory (g N m⁻²) between the surface and 10 m at each station (red to blue gradient color bar). Note that this figure was generated using MATLAB program (ver. R2019b and www.mathworks.com).

Dissolved N₂O measurements using a cavity ring-down spectrometer. For ease and convenience of gas extraction, we used the headspace method to extract dissolved N₂O gas from the samples (see Supplementary Text S2). Subsamples were obtained by transferring 40 mL of water samples from 120-mL glass bottles into a 100-mL glass gas-tight syringe, followed by the addition of 40 mL of high-purity N₂O-free air. The gas-tight syringe was shaken using an action shaker for 10 min to achieve equilibrium of gases between the sample and headspace phases (Supplementary Fig. S1). This equilibrium gas was injected into a Cavity Ring-Down Spectrometer (CRDS), which is a laser-based technique that uses the optical absorbance characteristics of the gas. CRDS has recently been widely and frequently used to measure greenhouse gases in various marine environments. Herein, we used a commercially available CRDS (Model G2308, Picarro Inc., USA) for N₂O measurements (Supplementary Fig. S1). As N₂O concentration obtained by the CRDS is the concentration in the headspace, a calculation is required to determine the concentration of dissolved N₂O in the seawater sample (Eq. 1):
\[ N_2O_{\text{conc.}} = \left( \beta \cdot x \cdot P \cdot V_w + \frac{x \cdot P}{R \cdot T} \cdot V_{hs} \right) / V_w, \]  

(1)

where \( \beta \) is the Bunsen solubility (nmol L\(^{-1}\) atm\(^{-1}\)) determined from the relationship between seawater \( \theta \) and \( S \)\(^{34}\); \( x \) is the dry gas mole fraction (ppb) measured in the headspace; \( P \) is the atmospheric pressure (atm); \( V_w \) is the volume of the water sample (mL); \( V_{hs} \) is the volume of the headspace phase (mL); \( R \) is the gas constant (0.082057 L atm K\(^{-1}\) mol\(^{-1}\)); and \( T \) is the equilibration temperature in Kelvin (K)\(^{35}\).

To validate the CRDS-based \( N_2O \) measurements, the measurement accuracy was examined by repeatedly measuring an \( N_2O \) standard gas, which was certified as 334.1 ppb by the Korea Research Institute of Standards and Science, before and after the sample measurement with an interval of 20 samples. The measurements of standard gas were well reproduced within a deviation of approximately 3% (Supplementary Fig. S2). In addition, we repeatedly measured the reference water (RW) of known concentration \( (N_2O_{\text{RW}} = 7.74 \text{ nmol L}^{-1}) \) obtained by equilibrating the ambient air \( (N_2O_{\text{air}} = 337.3 \text{ ppb}) \) with seawater \( (T = 20.5^\circ C \text{ and } S = 33.93 \text{ psu}) \) for 24 h in the laboratory\(^{36}\). The \( N_2O_{\text{RW}} \) was estimated from the \( T \) and \( S \) of the equilibrated water\(^{34}\). The analytical precision was approximately 4% (Supplementary Table S2).

Because we collected single water samples to measure the dissolved \( N_2O \) concentrations during the 2017 summer survey, we conducted measurements of duplicate samples collected at different times and in different environments (Supplementary Fig. S3). The measurement discrepancy between duplicate samples was no greater than 3% (Supplementary Tables S3 and S4).

**Estimations of excess \( N_2O \) and biogeochemical tracers (AOU and \( N^\circ )$.** The excess \( N_2O \) (\( \Delta N_2O \)), which is the amount of biogeochemically derived \( N_2O \), was estimated as the difference between the equilibrium \( N_2O \) (expressed as \( [N_2O]_{eq} = N_2O_{air} \beta \cdot P \)), where \( N_2O_{air} \) is the atmospheric \( N_2O \) level) and measured \( N_2O \) concentration \( (N_2O_{measured})^{30,37,38} \) and is expressed as follows:

\[ \Delta N_2O \text{ (nmol L}^{-1} \text{)} = N_2O_{measured} - N_2O_{eq}. \]  

(2)

To accurately estimate \( \Delta N_2O \), we need to estimate the time at which a water parcel has its last contact with the atmosphere (i.e., ventilation age) because \( N_2O_{air} \) varies temporally\(^{39}\). Although this concept has not been applied in many studies, a few studies have estimated \( \Delta N_2O \) by analyzing isotopes\(^{20,25} \) or estimating the convection rate\(^{22}\). However, in these previous studies, there was also a limitation in the uncertainty that the conversion rate. In addition, a negative linear relationship between \( \Delta N_2O \) and \( AOU \) indicates that nitrification \( \text{NH}_4^+ \rightarrow \text{NO}_2^- \rightarrow \text{NO}_3^- \) is the main pathway of \( \Delta N_2O \) production\(^{29,44} \). In addition, \( \Delta N_2O \) has been widely used as an indicator of excess nitrogen (e.g., a nitrogen fixation) or deficit (e.g., denitrification; \( \text{NO}_3^- \rightarrow \text{NO}_2^- \rightarrow \text{N}_2/O_2 \)) relative to phosphorus\(^{40}\). In this study, \( \Delta N_2O \) was calculated as \( \text{AOU} = -16 \times [\text{PO}_4^{3-}] \), where 16 is the Redfield ratio of N to P. A negative linear relationship between \( \Delta N_2O \) and \( \text{AOU} \) indicates that \( \Delta N_2O \) is mainly produced through denitrification\(^{13,46,50}\).

**Results and discussion**

**Summer oceanographic conditions.** During the summer open-water season, it is apparent that the physical and biogeochemical properties of the WAO surface waters show a latitudinal gradient from the Bering Strait to the Chukchi Borderland\(^{12}\). The southern Chukchi (SC) (i.e., Bering Strait to Chukchi Shelf), which is mainly influenced by relatively warm and nutrient-enriched Pacific waters, is a highly biologically productive marine environment displaying a generally high chlorophyll-\( a \) biomass\(^{12,55} \) (Fig. 1a). By contrast, the northern Chukchi (NC) (i.e., the Chukchi Borderland and Canada Basin) is primarily driven by freshwater inputs from melting sea ice and rivers, and is characterized as cold, fresh, and oligotrophic, displaying a generally low chlorophyll-\( a \) biomass (Fig. 1a)\(^{52-54}\).

To investigate the hydrographic conditions during the summer of 2017 in the WAO, we analyzed the vertical distributions of \( \theta, S, \text{DO, and DIN} \) along a latitudinal transect from the Bering Strait to the Chukchi Borderland (Fig. 1a–e,f) and used a \( \theta-S \) diagram to examine the composition of water masses (Fig. 1d). The distributions of \( B, S, \text{and DIN} \) in the surface waters (< 50 m depth) suggest generally warmer, more saline, and DIN-richer waters in the SC (mean \( \theta_{50-50} \) of 5.14 °C, \( S_{50-50} \) of 32.33 psu, and \( \text{DIN}_{50-50} \) of 5.31 μmol L\(^{-1}\)) compared to the NC (mean \( \theta_{50-50} \) of 1.00 °C, \( S_{50-50} \) of 29.65 psu, and \( \text{DIN}_{50-50} \) of 1.20 μmol L\(^{-1}\)) (Fig. 1b,c,f). In addition, the DIN\(_{10-10} \) inventory is higher in the SC (mean of 0.61 g m\(^{-2}\)) than in the NC (mean of 0.01 mg m\(^{-2}\)) (Fig. 1f), indicating a greater potential for driving a higher primary production in the SC, as found by Grebmeier et al.\(^{55}\).
The intermediate depths (50–200 m) in the NC are completely occupied with cold (< 0 °C), saline (31.5–33.6 psu), DO minimum (~ 170 μmol L⁻¹), and nutrient-rich (DIN > 10 μmol L⁻¹) waters. These water characteristics are associated with Pacific Winter Water (PWW)⁵⁷,⁶⁰,⁶¹, which forms in the Bering Sea during winter. The PWW is identified by a signature minimum N* on the θ-S diagram (Fig. 1d).

Below the PWW, the maximum θ (~ 1.25 °C) and high salinity (~ 34.89 psu) water observed between 200 and 1000 m in the deep NC (Fig. 1b,c) is typically called Atlantic Water (AW)⁵⁶,⁶⁰. In contrast to PWW, AW is associated with a relative maximum N* (Fig. 1d). The densest waters (θ < 0 °C and S = ~ 34.95 psu), with a relatively uniform θ/S, are distributed from below ~1000 m to the bottom and are defined as Arctic Bottom Water (ABW)⁶²,⁶³. ABW is associated with a maximum N* signature along the transect (Fig. 1d).

In summary, during the summer of 2017, the study area consisted of five water masses: PSW, FW, PWW, AW, and ABW, recognizable in both the vertical and horizontal directions. Further details on the physicochemical characteristics of these water masses are provided in Supplementary Table S6. To discuss N₂O dynamics, the water column was divided into three layers based on the vertical distribution of the water masses in the study area: surface (0–50 m), intermediate (50–200 m), and deep (200–2200 m) areas (Fig. 2).

**N₂O dynamics: distribution and controlling factors.** Within the surface layer, the vertical distribution of SC N₂O concentrations shows a pattern of increasing concentration with depth (i.e., surface, ~ 11.1 nmol L⁻¹; up to 50 m, ~ 19.4 nmol L⁻¹), whereas those in the NC are uniformly distributed with values of approximately 16.6 nmol L⁻¹. The mean N₂O concentrations of the SC and NC are estimated as 14.7 ± 2.1 and 15.9 ± 0.8 nmol L⁻¹, respectively; however, the mean N₂O saturation is higher in the NC (113% ± 10%, over-saturation) than in the SC (95% ± 5%, under-saturation).

Toyoda et al.²⁵ published N₂O measurements taken in 2014 and 2015. In the 2014 data vertical N₂O concentrations in the SC were observed to increase with depth (up to 23 nmol L⁻¹), while those in the 2015 data were homogeneous (~ 15 nmol kg⁻¹) throughout the water column. Toyoda et al. surmised that the annual variations in N₂O distribution may be a function of whether or not stratification occurs within the water column. Based on our results, we suggest that waters over-saturated with N₂O in the SC bottom water could migrate towards the surface as a result of vigorous vertical mixing.

There is a subsurface N₂O maximum (~ 22 nmol L⁻¹) in the intermediate layer with a mean N₂O concentration and saturation of 17.2 ± 1.5 nmol L⁻¹ and 107% ± 10%, respectively. PWW, showing the lowest DO (~ 170 μmol L⁻¹), nutrient-rich (DIN > 10 μmol L⁻¹), and relatively minimum N* (~ 19.91 μmol L⁻¹) features, fully occupies the intermediate layer (Fig. 1). Consequently, the highest N₂O concentration (22.0 nmol L⁻¹) and saturation (138%) are found in the intermediate layer. These results are consistent with those of previous studies¹³,¹⁹,²¹,²⁴ where subsurface N₂O maxima ranged from 18.1 to 24.6 nmol L⁻¹ and were recorded at depths of between 100 and 200 m.

The deep layer is mainly composed of AW (generally distributed between 200 and 1000 m with a mean SF₆-based ventilation age of 24.3 ± 3.9 years) and ABW (generally, below ~ 1000 m with an SF₆-based ventilation age of 46.6 ± 14.5 years) (Supplementary Fig. S6). Under these conditions of great age and relative stability, N₂O concentrations should show little variation (Supplementary Table S7). The N₂O concentrations were constant at 13.9 ± 1.0 nmol L⁻¹ in the AW zone, and showed a slightly decreasing trend in the ABW zone (bottom, 12.9 ± 0.8 nmol L⁻¹). The saturation values in both zones are mostly less than 100% (i.e., under-saturated conditions).

The distribution of SC N₂O exhibited different patterns from the distribution of NC N₂O within the surface layer. These results are indicators of the effect of the physical solubility, which is mainly determined by T and S³⁴, and is dominant in the NC (cold and fresh) compared to the SC (warm and saline)²³. In addition to the physical solubility, Randall et al.⁶⁴ reported that the N₂O of sea-ice meltwater was greatly under-saturated, and several studies¹³,¹⁹,²¹,²⁴ have suggested that the under-saturated N₂O in the NC surface water may be related to the dilution of melting sea ice.

The over-saturated SC, which is known to be a high biological productivity region⁵¹, is presumed to have biogeochemically derived N₂O (i.e., ΔN₂O) production that also contributes to the concentration. In addition, the ΔN₂O production of each water parcel was precisely calculated, resulting from the SF₆-derived ventilation date. To identify potential ΔN₂O production sources in the SC, we evaluated both the negative linear relationship between ΔN₂O and N* and the positive linear relationship between ΔN₂O and DIN. The relationship between ΔN₂O and AOU was not considered. This approach was taken because the SC is a shallow shelf region where the entire water column is kept in relatively close contact with the atmosphere. Plots of ΔN₂O versus DIN show significantly positive correlations (R² = 0.50, p < 0.05) (Fig. 2), suggesting that nitrification is likely to serve as the main sources of ΔN₂O in the SC.⁵⁶ Interestingly, plots of ΔN₂O versus N* show weak negative correlations (R² = 0.10, p < 0.05) and scattered distributions. It has been suggested by both Hirota et al.²⁰ and Toyoda et al.²⁵ that ΔN₂O production in the SC can be attributed to sedimentary denitrification resulting from the coupling of the inverse correlation between ΔN₂O and N* and the stable isotope composition of N₂O. Here, the somewhat ‘scattered’ relationship observed between ΔN₂O and N* may be due to the efflux of N₂O produced by sedimentary
denitrification. Regrettably this hypothesis remains un-tested due to the absence of robust sedimentary data, leaving scope for future work.

Similar results have been reported in earlier studies. According to Zhang et al.\textsuperscript{13}, the N\textsubscript{2}O concentration in the SC increased with depth, and both the ΔN\textsubscript{2}O-AOU and ΔN\textsubscript{2}O-N\textsuperscript{*} relations were significant, suggesting that vigorous ΔN\textsubscript{2}O production is generated through sedimentary nitrification and denitrification. Wu et al.\textsuperscript{21} also observed high N\textsubscript{2}O concentrations corresponding simultaneously to the oxygen minimum and high concentrations of NH\textsubscript{4}+ over the Chukchi Sea continental shelf, and suggested that N\textsubscript{2}O production is derived from sedimentary nitrification, denitrification, and nitrification in the water column. Fenwick et al.\textsuperscript{19} also suggested that the significant relationship between ΔN\textsubscript{2}O and N\textsuperscript{*} represents the primary source of denitrification and that the significant scatter found in this relationship is due to the influence of other nitrogen cycling processes on ΔN\textsubscript{2}O production, albeit an insignificant relationship between ΔN\textsubscript{2}O and AOU. The results of these studies support our findings.

The low temperatures (i.e., high gas solubility) characteristic of PWW may be a potential cause of the N\textsubscript{2}O maximum observed within the intermediate layer. If, however, N\textsubscript{2}O concentrations were high solely due to the water parcel’s high solute capacity, we should expect dissolved DO concentrations to be similarly elevated. However, the DO concentrations measured within the intermediate layer were low and the AOU was high. SF\textsubscript{6} is likewise affected by solubility as other gases. Based on the assumption that dissolved SF\textsubscript{6} concentrations in any given water parcel will be in equilibrium with the adjacent atmosphere prior to being ventilated, one can determine the ventilation date and the precise equilibrium N\textsubscript{2}O concentrations of the water parcels corresponding to that ventilation date through reference to the SF\textsubscript{6} concentrations. Compared to the equilibrium N\textsubscript{2}O levels at

Figure 2. Vertical distribution of N\textsubscript{2}O measured in the WAO water column during the summer of 2017 with saturation levels (N\textsubscript{2}O\textsubscript{sat} (%) = (N\textsubscript{2}O\textsubscript{measured}/N\textsubscript{2}O\textsubscript{eq}) × 100; blue to red gradient color bar). Based on the vertical composition of water masses in the study area (surface, Pacific summer water and fresh water; intermediate, Pacific Winter Water; and deep areas, Atlantic Water and Arctic Bottom Water), we represent the vertical N\textsubscript{2}O distribution by dividing into three layers (i.e., surface, 0–50 m; intermediate, 50–200 m; and deep layer, 200–2200 m). To show contrasting values associated with saturation levels (%), the surface layer is shown separately for SC and NC. To investigate the potential sources of biogeochemically derived N\textsubscript{2}O (ΔN\textsubscript{2}O) production, the figures on the right-hand side show the correlations of ΔN\textsubscript{2}O with DIN and N\textsuperscript{*} in the surface layer of the SC (green color boxes), and with AOU and N\textsuperscript{*} in the intermediate layer (orange color boxes), including statistical information (R\textsuperscript{2} and p values). The inset shows the correlation between ΔN\textsubscript{2}O and DIN in the intermediate layer. Note that this figure was generated using MATLAB program (ver. R2019b and www.mathworks.com).
the time of water mass formation (i.e., ventilation date), the N₂O concentrations observed in PWW were clearly indicative of over-saturation.

The highest N₂O concentration and saturation in the intermediate layer suggest that here N₂O production is significant. The relationships between ΔN₂O and AOU have positive (R² = 0.21, p < 0.05) correlations. In addition, a positive linear relationship between ΔN₂O and DIN is well represented (R² = 0.29, p < 0.05). The results suggest that nitrification may contribute to ΔN₂O production in the intermediate layer. Meanwhile, the relationships observed between ΔN₂O and N⁺ were weak (R² = 0.13, p < 0.05) potentially as a result of interaction between the bottom water and sediments on the shelf slope.

In addition, the lateral input of shelf waters (i.e., PSW) might contribute to the N₂O concentrations of the intermediate layer18. Zhang et al.13, Wu et al.21, and Toyoda et al.25 have all suggested that the subsurface N₂O maximum may be attributable not only to its local production within the water column (e.g., nitrification), but also to its northward transportation from the SC. Given that PSW input increases with latitude16, lateral transport of N₂O may be a significant factor in determining the characteristics of surface/intermediate layers within the NC.

According to Zhan et al.22 and Fenwick et al.19, in the deep layer of the Arctic Ocean, both the decreasing N₂O and oxygen with depth and the estimated NO₃⁻ regeneration rate (2.3 × 10⁻⁵ μmol L⁻¹ a⁻¹) indicate that nitrification may be insignificant for N₂O accumulation. Denitrification may also be insignificant for N₂O accumulation because of the relatively high oxygen concentration. It has been suggested that the N₂O levels observed in the deep layer samples may have occurred because the water was last ventilated during pre-industrial times. This hypothesis is based on the estimated convection rate. Offering their own interpretation of the isotopic data, Toyoda et al.25 suggested that the N₂O concentrations observed in the deep layer were derived from a mixture of water ventilated under pre-industrial atmospheric conditions and N₂O produced by nitrification occurring within the water column.

Here, based on the SF₆-based ventilation age, ventilation dates of the deep water masses (i.e., AW and ABW) were determined to be from circa 1955.9 to 1995.1. The N₂O concentrations of the deep layer were undersaturated, compared to equilibrium values in atmospheric N₂O of the ventilation dates. These uniformly undersaturated N₂O concentrations and the relatively homogeneous hydrographic properties suggest that deep N₂O concentrations are mainly determined by physical mixing processes such as advection and formation, rather than the involvement of biogeochemical processes (i.e., nitrification and denitrification).

**Estimation of N₂O flux: WAO source or sink during the summer of 2017?** To determine whether the WAO was a net source or sink for atmospheric N₂O during the summer of 2017, we used the air-sea gas exchange equation to estimate the N₂O flux as follows:

\[
N_2O_{flux} = k_w \cdot \left( [N_2O]_{measured} - [N_2O]_{eq} \right),
\]

where \([N_2O]_{measured}\) is the surface-water N₂O concentration, and \(k_w\) is the gas transfer velocity (cm h⁻¹). In previous studies19,21,22, three model approaches were used to estimate \(k_w\) in the WAO, i.e., those developed by Wanninkhof87, Wanninkhof and McGillis88, and Nightingale et al.89. It should be noted that we used the \(k_w\) of Wanninkhof89 instead of that of Wanninkhof87 to more accurately reflect \(k_w\) in Eq. (3) (refer to Supplementary Text S1). Fenwick et al.19 and Zhan et al.22 used the weighted mean wind data (60 days prior to sampling) to avoid an overestimation of the instantaneous wind data in the process of calculating \(k_w\). However, we used the mean wind data during sampling to provide results as observation-based snapshots. The mean differences in the estimated N₂O flux from the three models are 0.3 μmol N₂O m⁻² day⁻¹ in the SC and 0.2 μmol N₂O m⁻² day⁻¹ in the NC (Supplementary Table S8). To facilitate the presentation of our results, we employed the mean value of the N₂O flux averaged from the three models.

A map illustrating the spatial distribution of the summer WAO N₂O fluxes (Fig. 3a) indicates that the SC (Sts. 1–9) is exclusively occupied by positive (+: sea → air) N₂O fluxes ranging from 0.1 to 8.6 μmol N₂O m⁻² day⁻¹, whereas the NC (Sts. 10–30) is dominated by negative (−: air → sea) N₂O fluxes ranging from −4.3 μmol N₂O m⁻² day⁻¹ to zero. Interestingly, a contrasting distribution of the N₂O fluxes between the SC (+) and NC (−) is apparent along the FZ, similar to that suggested for the physical and biogeochemical properties determined by Lee et al.23 (see their Fig. 1).

Our results agree well with those of several previous studies. The positive N₂O fluxes were estimated by Hirota et al.20, although their research area was limited to the south of the SC, and the investigation was conducted in few locations. Wu et al.21 and Zhan et al.22 likewise estimated positive N₂O fluxes in the SC and negative N₂O fluxes in the NC, although the flux values were calculated using only one model. In addition, Fenwick et al.19 estimated relatively lower fluxes, suggesting that these results may be due to either of the different calculation approaches (e.g., weighted mean wind data over 60 days prior to sampling), varying oceanographic conditions (e.g., dilution by melting of sea ice with low N₂O concentration) or both. In addition, the locations are intensive near the coast. Zhan et al.24 also used the weighted mean wind data for three different models, and the SC was identified as a source of atmospheric N₂O, but the NC was not identified as either a source or a sink. Despite the similar surface N₂O concentrations (~16.5 nmol L⁻¹) with our dataset, these different results may be due to different calculation approaches (i.e., different air-sea exchange models and mean wind data). Toyoda et al.25 estimated negative and positive N₂O fluxes in the SC, respectively, in 2014 and 2015. However, the fluxes investigated during late summer were more positive than during that early autumn in each of two years. They suggested that the SC can act as both source and sink depending on the season.

Until this study, there has not been a clear estimation of the controlling factors of N₂O fluxes in the WAO. To investigate the factors controlling the WAO N₂O fluxes of summer 2017 for the first time, we examined correlations between the N₂O flux and physical and biogeochemical parameters, such as the sea surface temperature.
Take together, these results suggest that during the summer of 2017, the SC acted as a source (mean of +2.3 ± 2.7 μmol N2O m−2 day−1) and the NC served as a sink (mean of −1.3 ± 1.5 μmol N2O m−2 day−1) for atmospheric N2O. The summer W AO N2O fluxes were significantly influenced by physical variables associated with the solubility (i.e., SST and SSS) and biogeochemically derived N2O production, implying that the distribution of the W AO N2O flux is typically strongly susceptible to environmental changes.

A multitude of environmental changes that occur in the W AO may directly and indirectly influence the distribution of W AO N2O fluxes (see Fig. 3a). Among the changes observed, the increasing inflow of the Pacific waters10,72−73 and the rapidly declining sea-ice extent9,11,74,75 have received substantial attention to date. Based on these two phenomena, we can speculate that the distribution of W AO N2O fluxes revealed in this study will change in the future (Fig. 4). Lewis et al.76 suggested that the increased phytoplankton biomass sustained by an influx of new nutrients, in addition to sea-ice reduction, has driven the Arctic Ocean (e.g., Chukchi Sea) to be increasingly more productive. The increased biomass would lead to intense nitrification and potentially benthic denitrification, resulting in increased N2O production within the water column. The increasing inflow of warm and nutrient-enriched Pacific waters into the W AO would likely extend the productive SC region northward, leading to an enlarged WAO role as an N2O source (positive, sea → air), whereas a rapid loss of the sea ice extent may ultimately lead to a sea-ice-free NC with a northward shift, resulting in a diminished role as an N2O sink (negative, air → sea). Should this potential scenario come to pass, we would expect the W AO to become an oceanic N2O “hot spot” source region, and we therefore suggest that this positive feedback scenario should be considered in an effort to improve the future trajectory of W AO changes.

Summary and conclusions
We investigated the distributions of the N2O concentration and flux, their controlling factors, and the role of the W AO as a source or sink for atmospheric N2O during the summer of 2017. In the surface layer (0–50 m, consisting of PSW + FW), the mean N2O concentration of the SC and NC is estimated to be 14.7 ± 2.1 and 15.9 ± 0.8 nmol L−1, respectively. However, the mean N2O saturation was higher in the SC (113% ± 10%, over-saturation) than in the NC (95% ± 5%, under-saturation). This result indicates that the effect of the physical solubility is dominant in the NC (cold and fresh) compared to the SC (warm and saline), and that the over-saturated SC is likely to gain additional biogeochemically derived N2O (i.e., ΔN2O) production through nitrification. The intermediate layer (50–200 m, occupied by PWW) exhibits a subsurface N2O maximum (≥ 20 nmol L−1) with a linear relationship between ΔN2O and AOU (positive). In the deep layer (200–2200 m, consisting of AW and ABW), deep N2O concentrations are mainly determined by the physical mixing processes, such as advection and formation. The estimated mean N2O flux across the air–sea interface was +2.3 ± 2.7 μmol N2O m−2 day−1 in the SC region (i.e., source) and −1.3 ± 1.5 μmol N2O m−2 day−1 in the NC region (i.e., sink), respectively, showing a contrasting distribution of N2O flux.

As our study was based on a single investigation, it is impossible for us to represent the entire 2017 calendar year, or even the entire summer of 2017. We are not, however, alone in suffering from this ‘limited data’ impediment. The Arctic Ocean is an extreme environment, acquiring year-round data is very difficult and extremely
costly. Consequently, our results are a mere snapshot of what is undoubtedly a much bigger picture. We intend to propose a direction for future work based on our experience of undertaking this study.

If Arctic changes are accelerated and consequently drive the Arctic Ocean in a more productive manner, the WAO may become an oceanic N\textsubscript{2}O "hot spot" source region. Given that these processes are relevant to global climate change, additional observations of the time series and more open-water seasons are required to support this scenario. Therefore, attention should be paid to future N\textsubscript{2}O dynamics in the WAO.

Data availability
Hydrographic data are available in Korea Arctic Ocean-data System (http://kaos.kopri.re.kr/cmm/main/mainPage.do). Atmospheric N\textsubscript{2}O data are available in ESRL (https://www.esrl.noaa.gov). The SF\textsubscript{6} data collected from the 2015 CLIVAR ARC01 cruise are available in CCHDO (https://cchdo.ucsd.edu/). The N\textsubscript{2}O flux and wind data are presented in the Supporting Information.

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Author contributions
S.S.K. conducted the nitrous oxide sampling and measurements. J.M.H., S.S.K., and I.N.K. developed the concept and design of the manuscript. A.M.M. contributed to the physical aspects. J.M.H., S.S.K., and I.N.K. wrote the manuscript. All authors discussed the results and commented on the manuscript.

Competing interests
The authors declare no competing interests.

Additional information
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