

Nitrous oxide (N2O) in Macquarie Harbour, Tasmania

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Abstract. Fjord-like estuaries are hotspots of biogeochemical cycling due to their steep physicochemical gradients. The spatiotemporal distribution of nitrous oxide (N2O) within many of these systems is poorly described, especially in the Southern Hemisphere. The goals of this study are to describe the spatiotemporal distribution of N2O within a Southern Hemisphere fjord-like estuary, the main environmental drivers of this distribution, the air-sea flux of N₂O, and the main drivers of N₂O production. Sampling surveys were undertaken in Macquarie Harbour, Tasmania, to capture N2O concentrations and water column physicochemical profiles in winter (July 2022), spring (October 2022), summer (February 2023), and autumn (April 2023). N2O samples were collected from middle water depths in the ocean (5 m), minor river (1 m) endmembers, the major river (10 m) endmember at 2 m from the bottom, and at five depths through the water column at four stations within the main harbour body.

Results indicate that N₂O was consistently supersaturated (reaching 170% saturation) below the system's freshwater lens where oxygen concentrations are often hypoxic but infrequently anoxic. In the surface lens, levels of N₂O saturation vary with estimated river flow and with proximity to the system's main freshwater endmember. The linear relationship between apparent oxygen utilisation and ΔN_2O saturation indicates that nitrification is the process generating N₂O in the system. When river flow was high (July and October 2022), surface water N₂O was undersaturated (as low as 70%) throughout most of the harbour.

When river flow was low (February and April 2023) N_2O was observed to be supersaturated at most stations. Calculated air–sea fluxes of N_2O indicated that the system is generally a source of N_2O to the atmosphere under weak river flow conditions and a sink during strong river flow conditions. The

diapycnal flux was a minor contributor to surface water N_2O concentrations, and sub-halocline N_2O is intercepted by the riverine surface lens and transported out of the system to the ocean during strong river flow conditions. In a changing climate, western Tasmania is expected to receive higher winter rainfall and lower summer rainfall, which may augment the source and sink dynamics of this system by enhancing the summer and autumn efflux of N_2O to the atmosphere.

This study is the first to report observations of N_2O distribution, generation processes, and estimated diapycnal and surface N_2O fluxes from this system.

1 Introduction

Despite the fact that fjords and fjord-like estuaries represent only a small portion of the coastal area worldwide, they are responsible for sequestering 11 % of the global organic carbon (C) burial along terrestrial margins (Smith et al., 2015; Bianchi et al., 2018, 2020). These systems are significant sources of greenhouse gases (GHGs) to the atmosphere (Wilson et al., 2020; Rosentreter et al., 2023; Bange et al., 2024). Many are heavily stratified with strong water column physicochemical gradients (Acuña-González et al., 2006; Inall and Gillibrand, 2010; Hartstein et al., 2019; Salamena et al., 2021, 2022; Maxey et al., 2022). These gradients can be influenced by mesoscale climate drivers like the North Atlantic Oscillation (NAO) and Southern Annular Mode (SAM; see Austin and Inall, 2002; Gillibrand et al., 2005; Maxey et al., 2022) and local-scale drivers like freshwater input and marine intrusions (Inall and Gillibrand, 2010; Hartstein et al., 2019; Maxey et al., 2020; Salamena et al., 2022).

Nitrous oxide (N_2O) is a potent greenhouse gas (GHG), the increased presence of which in the atmosphere is primarily driven by emissions from agricultural soils (Tian et al., 2020, 2023). With a global warming potential nearly 300 times that of CO₂. N₂O is a key focus of climate studies (Myhre et al., 2013; Etminan et al., 2016; Eyring et al., 2021; Forster et al., 2021). Biological N₂O production occurs through the microbially mediated processes of ammonia oxidation, nitrite (NO_2^-) reduction, and nitrate (NO_3^-) reduction (Kuypers et al., 2018). In marine systems, N₂O production is influenced by environmental conditions such as dissolved oxygen (DO) availability, inorganic nitrogen (N) availability, light availability, temperature (e.g. Raes et al., 2016), pH (e.g. Breider et al., 2019), and microbial community composition (e.g. Wu et al., 2020). Many coastal systems are experiencing a reduction in DO availability (Limburg et al., 2020; Testa et al., 2023) and an increased presence of N2O as a consequence (Laffoley and Baxter, 2019; Ji et al., 2020; Wilson et al., 2020; Wan et al., 2022; Orif et al., 2023; Resplandy et al., 2024).

Estuarine systems often have disproportionately high biological productivity relative to other marine systems (Walinsky et al., 2009; Gilbert et al., 2010; Bianchi et al., 2018, 2020). This also applies to N_2O dynamics, with approx. 33 % of marine N₂O emissions coming from estuaries (Bange et al., 1996; Seitzinger et al., 2000; Murray et al., 2015; Reading, 2022; Rosentreter et al., 2023). Estuaries can act as net sinks (Maher et al., 2016; Wells et al., 2018) or sources (De Bie, 2002; Zhang et al., 2010; Sánchez-Rodríguez et al., 2022) of N₂O depending on physical drivers of air-sea fluxes including waterbody and atmospheric concentration gradients, current velocities, depth, and wind speed (Wells et al., 2018; Bange et al., 2019). Other factors include land use modification (Reading et al., 2020; J. Chen et al., 2022) and the presence of microplastics (C. Chen et al., 2022). Despite the advancements made thus far, our understanding of marine N₂O distribution and atmospheric emissions is poorly constrained and needs improvement (Bange et al., 2019, 2024), especially in Southern Hemisphere fjord-like systems (Yevenes et al., 2017). Much of the current uncertainty lies with a lack of in situ data describing seasonal N2O dynamics being available to constrain global emission models (Bange et al., 2019).

The purpose of this study was (1) to investigate the distribution and seasonal variability in N_2O concentrations and emissions in a Southern Hemisphere fjord-like estuary and (2) to decipher the major physical and biological drivers of these emissions.

2 Methods

2.1 Study area

Macquarie Harbour is a Southern Hemisphere fjord-like estuary located on the western coast of Tasmania, Australia (Fig. 1). The harbour is oriented NW-SE and is approximately 33 km long, 9 km wide, and has a surface area of 276 km². The mouth of the harbour is constricted by a shallow (4-8 m) and long (14 km) sill known as "Hells Gates". Hells Gates muffles tidal forcing, resulting in harbour water levels primarily determined by the river flow and wind set-up (Hartstein et al., 2019). The morphology of this system results in sharp gradients of DO, salinity, and temperature that are seasonally dependent (Cresswell et al., 1989; Hartstein et al., 2019; Maxey et al., 2022). In surface waters, dissolved oxygen (DO) concentrations are nearly always in equilibrium with the air but decrease sharply through the halocline (~ 8 to 15 m). There is almost no DO produced below the halocline (8 to 12 m deep) due to high chromophoric dissolved organic matter (CDOM) levels limiting primary production at the surface (Maxey et al., 2017, 2020). Sub-halocline layers ($\sim 15 \text{ m}$ to a few metres from the bottom) are observed to have DO concentrations below 62.5 µM more than 50 % of the time (see Maxey et al., 2022). Near the seabed, episodic marine intrusions (deep-water renewal) refresh the supply of DO near the mouth of the system but refresh the upper reaches of the harbour less frequently (see Andrewartha and Wild-Allen, 2017; Hartstein et al., 2019; Maxey et al., 2022). This process is driven by low atmospheric pressure, sustained NW winds, and low catchment rainfall, which itself is influenced by the Southern Annular Mode (SAM) (Hartstein et al., 2019; Maxey et al., 2022). In the harbour's upper reaches, DO concentrations fall below 31 µM nearly a third of the time (Maxey et al., 2022). Hydrodynamic and oxygen tracer numerical model simulations of the harbour by Andrewartha and Wild-Allen (2017) estimate that 50% of the harbour's basin waters are replaced every 65 d during low-river-flow conditions and approximately 110 d during normal flow conditions.

The main source of freshwater to the harbour is located on its southeast end (the Gordon River) and drains a nearly pristine catchment (including the Franklin River) of approximately 5682 km² (Macquarie Harbour Dissolved Oxygen Working Group, 2014; Fig. 1). The Gordon River discharges an estimated 180 000 t organic carbon (OC) per year into the estuary (Maxey et al., 2020, 2022). It should be noted that this area receives the some of the highest rainfall (more than 2500 mm yr⁻¹) volume in Australia (Dey et al., 2019). The King River, located on the harbour's northern end, is the second-largest contributor of fresh water to the estuary and drains a catchment area of 802 km². Unlike the Gordon River, the King River has a history of receiving treated mining (e.g. copper) effluent and transporting this to the harbour (Carpenter et al., 1991; Teasdale et al., 2003).



Figure 1. Macquarie Harbour, Tasmania. Water sampling stations are shown with red circles. The Kennaook/Cape Grim Air Pollution monitoring station is shown as a green star (see inset map). Cape Sorell Weather Station shown as an orange star. The Gordon Above Denison Stream Gauge is shown as a red star (see inset map). Aquaculture lease boundaries are shown as hollow rectangles. Lease locations are sourced from Land Information Systems Tasmania (LISTmap – https://maps.thelist.tas.gov.au/ last access: 1 November 2024). Station names reflect general harbour locations, where KR1 indicates King River 1; C10 and C08 indicate Central Harbour 10 and 08, respectively; WH2 indicates World Heritage Area 2; and GR1 indicates Gordon River station 1. Coordinates are displayed in GDA_1994_MGA_Zone_55 format. Bathymetry through the system is shown as a dashed line. Note that this track excludes stations KR4 and KR1.

2.2 Experimental design

Nitrous oxide distribution was assessed by collecting water samples across seven stations, including the harbour's endmembers (the mouths of the Gordon and King rivers and the harbour mouth at Hells Gates Inlet; see Fig. 1 and Table 1) and stations along the longitudinal axis of the harbour where the deepest basins are located (named KR1, C10, C08, and WH2). Samples collected at endmember stations were collected from a single depth as these stations are shallow. Samples in the harbour body were collected at five depths from the surface (2 m) to approx. 1 m from the seabed. Collection campaigns were conducted in July 2022, October 2022, February 2023, and April 2023. At each station and depth, three replicate vials (n = 3) were collected for the determination of N₂O concentration.

2.3 Field sampling

At each station, water quality sonde profiles were collected from the surface to the seabed at 1 m intervals using a YSI EXO-1 equipped with optical DO (accuracy from 0 to $625 \,\mu\text{M} \pm 3 \,\mu\text{M}$ or 1 % of the reading, whichever is greater; precision is 0.03 μ M), salinity (accuracy ± 0.1 % or 1 % of the reading, whichever is greater; precision is 0.01), temperature (accuracy is ± 0.15 °C, and precision is 0.01 °C), and depth sensors. Sonde calibration was checked and corrected (when needed) for each sampling period.

Water samples were collected at various depths (see Table 1) using a 5 L Niskin bottle sampler. Water sample parameters include dissolved total ammoniacal N (NH₃+ NH_4^+) (TAN), NO₃, and N₂O. N₂O samples were collected in triplicate immediately after retrieval of the Niskin bottle by transferring water from the bottle through silicone tubing into a 20 mL borosilicate vial. Sample water was added to the vial by placing the tubing at the bottom and allowing the vial to overflow several volumes before sealing with a butyl rubber stopper and aluminium crimp. After ensuring the sample vial was free of bubbles, 50 µL of saturated mercury chloride (HgCl₂) solution was injected into the sample to arrest biological activity. All N2O samples were shipped to GE-OMAR in Kiel, Germany, for analysis. Samples were measured in July and August 2023 within 4 to 12 months after sampling and were not affected by the storage time (Wilson et al., 2018).

Water collected for dissolved inorganic N was filtered immediately using 0.45 μ m polyethersulfone syringe filters (Whatman Puradisc). Samples were stored in a chilled dark container until being transported to Analytical Services Tasmania in Hobart, Australia, for analysis. Dissolved total TAN and NO₃⁻ were analysed using a Lachat Flow Injection Analyser. TAN and NO₃⁻ analyses used methods based on APHA standard methods (2005) 4500-NH₃ H (reporting limit 0.005 mg L⁻¹) and 4500-NO₃⁻ L⁻¹ (reporting limit 0.002 mg L⁻¹).

2.4 Analysis of rainfall and river loading estimation

Rainfall and river discharge were analysed using methods presented in Maxey et al. (2022), where rainfall and stream gauge data were collected from the Gordon River catchment, the Strathgordon Rainfall Gauge Station and the Gordon Above Denison (GAD) Stream Gauge (Fig. 1). The rainfall and flow metrics computed include the average daily rainfall over a 20 d period prior to sampling; total accumulated rainfall 20, 10, 5, and 3 d prior to sampling; estimated Gordon River flow into the estuary; and measured flow at the GAD Stream Gauge.

Gordon River flow was estimated by scaling daily rainfall to the size of the catchment and assuming a rainfall and runoff coefficient of 0.70 adopted from a neighbouring catchment with similar land cover, geology, and slope (Willis, 2008). Additional streamflow from Gordon River dam releases was estimated by subtracting scaled rainfall contributions to river flow measured at the GAD Stream Gauge. This flow was added to the estimated runoff entering the harbour. Rainfall and flow data were provided by the Australian Bureau of Meteorology (BOM). NO_3^- and TAN loading were estimated my multiplying the measured concentration of each parameter at station GR1 (see Fig. 1 and Table 1) by the estimated Gordon River flow.

2.5 Analysis of water column N₂O concentrations, air–sea flux, and diapycnal flux

2.5.1 Determination of N₂O concentrations

Water samples were analysed for N_2O using the static headspace equilibration method followed by gas chromatographic separation (HP Agilent 5890) and detection with an electron capture detector (ECD) as described in Bange et al., (2019), Bastian (2017), and Kallert (2017). The concentration of N_2O in the samples was calculated with the following equation (Eq. 1; see Bange, 2006):

$$C_{\rm obs} = \frac{x' P V_{\rm hs}}{RT P V_{\rm wp}} + X' \beta P.$$
(1)

 C_{obs} is the concentration (nmol L⁻¹) of N₂O in the water sample; x' is the measured dry mole fraction of N₂O in the sample vial's headspace; P is the ambient pressure set to 1 atm; V_{hs} and V_{wp} are the volumes of the headspace in the vial and water in the vial, respectively; R is the gas constant; T is the temperature during equilibrium; and β is the solubility of N₂O (Weiss and Price, 1980). The mean relative error in the concentration values obtained was 2.4 % (±0.16).

2.5.2 Estimation of N₂O air-sea fluxes and N₂O saturations

 N_2O air-sea fluxes (F in μ mol m⁻² d⁻¹) were estimated using equations from Zhang et al. (2010) and Bange et al. (2019) (Eq. 2):

$$F = K \times (C_{\rm obs} - C_{\rm eq}). \tag{2}$$

 C_{obs} is the measured concentration $(nmol L^{-1})$ of N₂O in the water sample, while C_{eq} is the air-equilibrated seawater N₂O concentration calculated for in situ temperature and salinity using the solubility data of Weiss and Price (1980). *K* is the gas transfer velocity, which in the absence of direct measurements can be expressed as a function of the wind speed and the Schmidt Number (*Sc*). For this study we sourced daily average wind speed from the Cape Sorrel Weather Station at the northern end of Macquarie Harbour (http://www.bom.gov.au/climate/data/index.shtml, last access: 1 October 2024, station ID 097000; see Fig. 1 for the station location). *K* was estimated using relationships

Station	Station depth (m) (MSL)	Dissolved oxygen salinity temperature	N ₂ O	$\begin{array}{c} \text{TAN} (\text{NH}_3 + \\ \text{NH}_4^+) \end{array}$	NO ₃
HG3 352484, 5325594	8	Every metre	5 m	5 m	5 m
KR4 365018, 5327550	3	1 m	1 m	1 m	1 m
KR1 361316, 5325972	36	Every metre	2, 12, 20, 30, 35 m	2, 12, 20, 30, 35 m	2, 12, 20, 30, 35 m
C10 363708, 5320464	44	Every metre	2, 12, 20, 30, 42 m	2, 12, 20, 30, 42 m	2, 12, 20, 30, 42 m
C08 365489, 5317238	47	Every metre	2, 15, 25, 35, 45 m	2, 15, 25, 35, 45 m	2, 15, 25, 35, 45 m
WH2 370218, 5309894	32	Every metre	2, 12, 20, 25, 30 m	2, 12, 20, 25, 30 m	2, 12, 20, 25, 30 m
GR1 377784, 5300603	12	Every metre	10 m	10 m	10 m

Table 1. Sampling stations showing coordinates, parameters, and sampling depth for each parameter (in m).

in Nightingale et al. (2000), Raymond and Cole (2001), and Wanninkhof (2014). Fluxes at Macquarie Harbour's endmember stations used K values that account for additional forcings like bottom shear (see Raymond and Cole, 2001; Zappa et al., 2003; Abril and Borges, 2004; Beaulieu et al., 2012; Rosentreter et al., 2021). Deeper stations in the harbour's main body (i.e. KR1, C10, C08, WH2) have surface layers that are separated from the seabed by more than 10 m. Wind-based K_{600} estimators were used to estimate air-sea flux in those locations (see Nightingale et al., 2000; Raymond and Cole, 2001; Wanninkhof, 2014). Atmospheric N₂O for this estimation was sourced from monthly mean baseline greenhouse gas mole fractions measured at the Kennaook/Cape Grim Baseline Air Pollution Station, located in northwestern Tasmania. This station measures atmospheric N₂O using a gas chromatograph (GC) equipped with an ECD (https://www.csiro.au/en/research/ natural-environment/atmosphere/latest-greenhouse-gas-data last access: 1 November 2024). N₂O saturation (in %) was computed as N₂O saturation = $100 \times (C_{obs}/C_{eq})$.

2.5.3 Estimation of diapycnal N₂O flux

N₂O diapycnal fluxes (F_{dia} ; Eq. 3) from basin waters (sample depths of 20 or 25 m) to the harbour's surface lens (sample depths of 2 m) were estimated as follows:

$$F_{\rm dia} = K\rho \frac{\rm d[N_2O]}{\rm dz},\tag{3}$$

where z is depth. Diapycnal diffusivity (K_{ρ} ; Eq. 4) is computed with the local buoyancy frequency (N²), Γ is set to 0.2 (Osborn, 1980), and ε is the dissipation rate of turbulent kinetic energy assumed to be on the upper end of values for the mixing zone of stratified systems 1×10^{-5} (Arneborg et al., 2004; Mickett et al., 2004; Fer, 2006).

$$K_{\rho} = \Gamma \frac{\varepsilon}{N^2} \tag{4}$$

2.6 Data analysis

The relationships between N_2O saturation and water quality parameters such as DO concentration, salinity, temperature, nitrate, and ammonium concentrations are determined using Pearson correlation. Differences in mean N2O saturation between season, depth and each sampling station were tested using a two-way ANOVA. Differences between rainfall and river flow metrics between seasons were tested using one-way ANOVA, and pair-wise testing using Bonferroni's correction was undertaken where significant differences between seasons were detected. The relationship between rainfall and river flow metrics from the Gordon River and surface water N₂O saturation and N₂O air-sea flux at each station was analysed using the Pearson correlation. The standard deviation (SD) of the mean air-sea flux and diapycnal flux was computed from error propagated from replicate observations of N₂O wind speed, N₂O concentration, and density (where appropriate) using methods from Ku (1966). Contour plots were made with Plotly Chart Studio (Plotly Technologies Inc.) with the title "Collaborative data science" (Montréal, QC, Canada, 2015; https://plot.ly, last access: 1 November 2024).

3 Results

3.1 Rainfall and river loading

The 20 d rainfall accumulation ranged from a low of 117 mm in July 2022 to a high of 139 mm in April 2023 (see Fig. 2a). Average (\pm se) daily rainfall was similar across all months and ranged from 5.12 (\pm 2.57) mm in July 2022 to 5.79 (\pm 3.03) mm in October 2022 (see Fig. 2b) with no seasonal differences detected (p = 0.4326).

Estimated flow at the Gordon River mouth and GAD stream gauge was greater in July and October 2022 than February and April 2023 (Fig. 2c). Significant seasonal differences in flow measured at the GAD stream gauge were detected ($p = 5.5 \times 10^{-7}$), with greatest flow in July and October 2022 and decreasing flow over February and April 2023. July flows at the GAD stream gauge were observed to be 107.6 (±15.9) m³ s⁻¹ and in April 2023 were observed to be 30.5 (±2.2) m³ s⁻¹ (Fig. 2d).

Estimated NO₃⁻ and TAN loading varied with NO₃⁻ loads of 1.69 td^{-1} observed in July 2022, which then dipped to 0.31 td⁻¹ in October 2022 and then increased again to 1.77 and 2.77 td⁻¹ in February and April 2023 (Fig. 2e). TAN loading mirrored this pattern, with peaks occurring in October 2022 and February 2023 and lows occurring in July 2022 and April 2023. N₂O loading from the Gordon River was observed to be 0.015 td⁻¹ in July 2022, 0.012 td⁻¹ in October 2022, 0.015 td⁻¹ in February 2023, and 0.016 td⁻¹ in April 2023 (Fig. 2f).

3.2 Water column physicochemical profiles

DO profiles at the stations located within the main body of the harbour show a well-oxygenated surface layer that rapidly attenuates with depth (Fig. 3a) through the halocline (Fig. 3b). There is a prominent riverine surface lens in the main harbour extending to depths of up to 8 m depending on sampling period and location within the estuary. Salinity in the surface waters was lower in July and October 2022 (6 to 13) than February and April 2023 (greater than 20). Below the halocline, salinity ranged from approx. 28 to 32.

The DO gradient between the surface and sub-halocline waters was steeper in October relative to July 2022, with October 2022 DO concentrations approaching single digits (3.1 μ M) at station WH2, which is the nearest station to the Gordon River mouth (see Fig. 1). In general, the sub-halocline concentrations of DO were lower with proximity to the Gordon River mouth. The temperature of the freshwater surface layer ranged from about 9 to 19 °C but showed little variation below the halocline where temperature ranged between 13 and 16 °C (Fig. 3c).

Nitrate concentrations in the surface water lens tended to be lower than those observed at sub-halocline depths (Fig. 4a). The greatest NO_3^- concentrations were observed 2 m above the seabed at station WH2 in July and October 2022 and at mid-basin depths at stations C10 and C08 during those same periods, with concentrations reaching 1.77 µmol. TAN concentrations were often observed below detection limits (0.3 µmol) but were greatest in the surface lens or within the halocline itself when detectable (Fig. 4b). TAN concentrations at WH2 tended to be found at higher levels through the water column relative to other stations (down to about 20 m) reaching 1.53 µmol at 15 m in October.

3.3 N₂O distribution

At each harbour station, depth and season (and their interaction) significantly impacted N₂O saturation (two-way ANOVA, $\alpha = 0.05$, degree of freedom (d.f.) = 59). At 2 m, N₂O saturation was observed to be below 100% at all stations in July 2022 (Figs. 5 and 6) and at stations KR1, C10, and C08 in October 2022. In February and April 2023, N₂O saturation in the harbour was above 100% through the water column except in KR1 surface waters. The maximum N₂O

concentrations were observed in the sub-halocline. Among the sub-halocline observations, the maximum N_2O concentrations (reaching over 170%) were observed at the base of the Hells Gates sill at station C10 in October 2022.

All endmember N₂O concentrations were undersaturated in July 2022. In October, stations KR1 and HG3 were observed to be approx. 100% saturated, but N₂O at station GR1 was undersaturated. In February and April 2023, N₂O concentrations were supersaturated at all endmember stations. There were statistically significant linear correlations between N₂O saturation and salinity (r = 0.494; $p = 5.5 \times$ 10^{-7} , n = 92), temperature (r = 0.391; $p = 1.2 \times 10^{-4}$; d.f. = 90), DO concentration (r = -0.563; $p = 5.2 \times 10^{-9}$; d.f. = 90), and nitrate concentration (r = 0.559; $p = 6.9 \times 10^{-9}$; d.f. = 90) in the harbour stations (Fig. 7). The correlation between N₂O saturation and the TAN concentration, however, was not statistically significant (r = 0.174; p = 0.31; d.f. = 34).

3.4 N₂O air-sea and diapycnal fluxes

Atmospheric N₂O mole fractions measured at Kinnaook / Cape Grim Air Pollution Station (see Fig. 1) were observed to increase from 334.7 ppb in July 2022 to 335.9 ppb in February 2023. The April 2023 atmospheric N₂O mole fraction was slightly lower than that observed in February 2023 at 335.6 ppb. Average (\pm SD) wind speeds were observed to be 6.6 (\pm 3.7) m s⁻¹ in July, 5.6 (\pm 2.5) m s⁻¹ in October, 6.3 (\pm 3.4) m s⁻¹ in February, and 6.4 (\pm 4.0) m s⁻¹ in April.

Estimated N₂O air–sea flux in the main harbour stations (KR1, C10, C08, WH2) ranged from $-12.88 (\pm 6.00) \mu mol N_2O m^{-2} d^{-1}$ at C10 in July 2022 (negative sign indicates absorption of N₂O into the surface waters from the atmosphere) to 7.31 (± 3.43) $\mu mol N_2O m^{-2} d^{-1}$ at the same station in February 2023 (using the "high" K₆₀₀ estimator from Raymond and Cole, 2001; see Table 2)

Station KR1 was always observed to be a site of atmospheric N₂O uptake and was every non-endmember station in July 2022. Near the head of the system, station WH2 was observed to be a net source of N₂O to the atmosphere from October 2022 to April 2023, and stations C10 and C08 (positioned above the deepest basins) were net sources in February 2023 and April 2023.

Estimated diapycnal fluxes (\pm SD) using local buoyancy frequencies showed a consistent upwards movement of N₂O from the sub-halocline to surface layers, with the smallest fluxes observed in July 2022 ($49 \pm 2.3 \text{ mol N}_2\text{O m}^{-2} \text{d}^{-1}$ at C08), and the largest fluxes observed in October 2022 (up to 1308 nmol N₂O m⁻² d⁻¹ at WH2) and February 2023 (up to 1200 $\pm 47.3 \text{ nmol N}_2\text{O m}^{-2} \text{d}^{-1}$ at C10); see Table 3. Patterns in the size of the diapycnal flux generally reflected the patterns of N₂O % saturation, with the largest fluxes occurring in October 2022 during the periods of greatest N₂O %



Figure 2. Rainfall and estimated Gordon River loading estimates for each sampling event: (a) accumulated rainfall (mm) 10, 5, and 3 d prior to each sampling event; (b) average (mean) daily rainfall over a 20 d period prior to each sampling event; (c) estimated Gordon River flow into the harbour in millions of cubic metres per day; (d) daily mean flow ($m^3 s^{-1}$) over the 20 d prior to sampling (\pm standard error) at the Gordon Above Denison Stream Gauge; (e) estimated nitrate and ammonium loads entering the harbour from the Gordon River; and (f) estimated N₂O load (t d⁻¹) entering the harbour from the Gordon River.

saturation. Overall, the magnitude of the estimated diapycnal fluxes was smaller than estimated air–sea fluxes.

4 Discussion

Our study is the first to report on N₂O distribution and airsea flux from an Australasian fjord-like estuary. We set out to investigate how N₂O concentrations varied along horizontal and depth gradients, how N₂O concentrations and estimated surface water emissions vary seasonally, how N₂O concentrations vary with freshwater inputs, and whether the relationship between apparent oxygen utilization (AOU) and Δ N₂O could help clarify the primary mechanism for N₂O generation in this system. We used surface water observations, local wind speed (from Cape Sorell weather station), and atmospheric N₂O mole fractions (from Kennaook/Cape Grim; Fig. 1) to estimate N₂O air–sea flux (based on Zhang et al., 2010, and Bange et al., 2019) and found that Macquarie Harbour functions as both a site of atmospheric uptake and emission of N₂O. Most harbour stations were estimated to be removing atmospheric N₂O in July and October 2022 (when river flow was greater) and emitting N₂O into the atmosphere in February and April 2023 (during low-river-flow periods; see Fig. 8 and Table 2). Pearson correlations show that when freshwater flow is high, N₂O air–sea flux is negative (indicating uptake from the atmosphere), and when freshwater flow is low, N₂O air–sea flux is positive (Table 2). Our observations highlight that freshwater flow is a key driver of N₂O emissions in this estuary. In addition, Gordon River flow is heavily influenced by hydroelectric dam



Figure 3. Dissolved oxygen (μ M) (**a**), salinity (**b**), and temperature (°C) (**c**) profiles (referencing height from seabed) collected at stations KR1, C10, C08, and WH2 in July 2022 (red dots), October 2022 (yellow dots), February 2023 (blue dots), and April 2023 (purple dots). Measurements were made every 1 m.

release (up to $\sim 28\%$ of the flow in July 2023). Rainfall in the catchment area may offset the effects of dam release, but our observations did not capture this as rainfall itself was not significantly correlated with N₂O concentrations or air–sea flux.

The river endmember concentrations of N_2O were often observed to be undersaturated, as observed in the South

Platte River basin, USA (McMahon and Dennehy, 1999); Neuse River estuary, USA (Stow et al., 2005); headwater streams in Ontario, Canada (Baulch et al., 2011); and Upper Mara River basin, Kenya (Mwanake et al., 2019). Our observations of river endmember N_2O concentrations were similar to the lower end of the concentrations reported in McMahon and Dennehy (1999) (approx. 80 % saturation) but not as low



Figure 4. Nitrate NO_3^- (a) and TAN (b) concentrations with depth (referencing height from seabed) collected at stations KR1, C10, C08, and WH2 in July 2022 (red dots), October 2022 (yellow dots), February 2023 (blue dots), and April 2023 (purple dots). Data presented as having a concentration of 0.0 are below the detection limits of the analyte.

as those reported Jackson Creek, Ontario, Canada, in Baulch et al. (2011), where some observations reached < 20% saturation. N2O undersaturation in those systems was attributed to complete denitrification (use of N2O as a terminal electron acceptor by denitrifiers) in streams with high DOC loads, low DO, and low NO_3^- concentrations. It should also be noted that up to 28% of the estimated Gordon River flow was found to be associated with flow through the Gordon Above Denison Stream Gauge (a proxy for hydroelectric dam and reservoir release to the Gordon River). Boreal reservoirs have been shown to be net sinks of atmospheric N₂O (Hendzel et al., 2005), which was attributed to increased N₂O demand to drive complete denitrification. There is good reason to believe that N₂O may be scavenged in the Gordon and King rivers as well because they do often have high DOC concentrations, high water column DO demand (Maxey et al., 2020), and low DO concentrations near the stream bed (Maxey et al., 2022).

Below the estuary's predominately freshwater surface lens, the fjord-like morphology drives suboxic conditions like those observed in the sub-halocline waters at station WH2 in October 2022 (see Fig. 3; Hartstein et al., 2019; Maxey et al., 2020, 2022). While these conditions do not always persist, DO concentrations below $31 \,\mu\text{M}$ have been observed to occur more than 30% of the time up the estuary, specifically at station WH2 (Maxey et al., 2022). In the low-DO sub-halocline layers of the harbour we observed the maximum N₂O concentrations (Figs. 5 and 6). Sub-halocline N₂O saturation was observed to generally range from approx. 110% to 170%, with the highest values observed within the deeper basins near the foot of the sill (stations C10 and C08; Fig. 6).

In the harbour's sub-halocline layer there is not enough light to support photosynthesis (Hartstein et al., 2019; Maxey et al., 2017, 2020, 2022), and thus the main source of oxygen is advection from marine intrusions. N₂O producing microbes have been observed to populate this layer of the harbour (see Da Silva et al., 2022), and our observations of supersaturated N₂O in these layers show that those microbes are active. Linear relationships between AOU and



Figure 5. Mean (\pm standard error) N₂O % saturation observed at each sampling station with depth and across seasons. Note that a dashed red line indicating 100 % at the time of sampling has been placed in each panel for reference.

 ΔN_2O (slope = 0.0154; r = 0.596; $p = 2.4 \times 10^{-23}$; Fig. 7c) and NO₃⁻ and N₂O saturation (r = 0.559; $p = 6.9 \times 10^{-9}$; Fig. 7d) indicate that N₂O production likely occurs primarily through the ammonia oxidation (nitrification) pathway (Yoshinari, 1976; Walter et al., 2004; Brase et al., 2017). Our observations are on the lower end of reported N₂O yield per mole O₂ consumed (see Suntharalingam and Sarmiento, 2000; Brase et al., 2017), which may be an artefact of mixing and loss dynamics such as basin water DO recharges from marine intrusions and loss to aerobic respiration and the atmosphere. This suggests that some portion of sub-halocline pelagic oxygen demand in the harbour can be attributed to nitrifying microbes (albeit at a much lower rate compared to aerobic respiration). Ji et al. (2020) also observed similar relationships in the Saanich Inlet, a seasonally anoxic fjord-like

estuary in British Columbia, but in that system anoxic conditions are more persistent (Bourbonnais et al., 2013; Manning et al., 2010) compared to Macquarie Harbour (Maxey et al., 2022). Deep-water renewal and marine intrusions have been hypothesised to stimulate N₂O production in the Saanich Inlet (Capelle et al., 2018; Michiles et al., 2019; Ji et al., 2020) and Baltic Sea (Walter et al., 2006) and may also be stimulating it in Macquarie Harbour as well. In the Baltic Sea, Walter et al. (2006) and Myllykangas et al. (2017) observed enhanced N₂O production in areas receiving significant marine intrusions. Positive correlations between AOU and Δ N₂O observed in western Baltic Sea waters (Walter et al., 2006), along with mean (11 years from 2006–2017) seasonal variations in DO and N₂O observed through the water column at the Boknis Eck Time Series Station (Eckernförde



Figure 6. Contour plots of mean N_2O % saturation (left column) and mean DO concentration in units of µmol (right column) observed at stations HG3, C10, C08, WH2, and GR1 from July 2022 to April 2023. Shaded red areas on the DO plots indicate low oxygen concentrations (< 93 µmol). Relative positions of the stations are shown in the top-left plot. The *y* axis displays depth in metres relative to mean sea level.

Bay, southwestern Baltic Sea), indicate a tight coupling between DO supply and N_2O production (presumably by nitrification) and consumption (presumably by denitrification) pathways in that area (Ma et al., 2019). The reintroduction of marine water on the upstream side of a dam in the Nakong River, South Korea, was found to affect bottom water trapping (stagnation), DO conditions, N process rates, processspecific gene abundances, and subsequently the fate of N in that system (Huang et al., 2024). Marine intrusions primarily refresh the DO supply adjacent to the sill in Macquarie Harbour (near station C10). As we observed a positive correlation between AOU and ΔN_2O marine intrusions offer a



Figure 7. Correlation between N₂O percentage saturation observed across the harbour and (a) salinity, (b) temperature, (c) Total Ammoniacal Nitrogen (TAN) concentration, and (d) nitrate concentration. The correlation between AOU (μ M) and Δ N₂O (nM) is shown in (e). The relationship between N₂O % saturation and DO % saturation is shown in (f). Pearson correlation coefficients (*r*) and their associated *p* values are shown in each panel.

possible explanation for the higher sub-halocline N_2O concentrations observed in this part of the harbour (see Fig. 7e).

One other possible pathway of water column N_2O production might be through denitrification as DO concentrations at WH2 in October 2022 approached single digits (3.1 µM). This station has the highest basin residence time compared the others used in this study. Low oxygen concentrations may also likely be found under the harbour's fish farms due to the aerobic respiration of farm debris (Maxey et al., 2020). Though whether denitrification functions as a production process or a loss process will depend upon the drivers of DO concentration (i.e. respiration rates, physical mixing, etc.) and may differ depending on the location of the basins in this system. It is likely the main driver of undersaturated N₂O concentrations in the Gordon River.

We conceptualise that during periods of high river flow, the surface water lens thickens and transports water undersaturated with N_2O quickly across the harbour surface and out of Hells Gates inlet. N2O from the continuously oversaturated sub-halocline water is entrained in the surface lens (diapycnal flux) and transported laterally and out of the system in its dissolved form. During periods of low river flow, the surface lens is thinner and residence times longer (Andrewartha and Wild-Allen, 2017; Maxey et al., 2022). We suspect that N2O from the oversaturated sub-halocline water then diffuses through the surface layer and is emitted into the atmosphere in its gaseous form (Fig. 9). Our estimates of diapycnal flux indicate that the mass transport from sub-halocline waters is smaller ($\sim 2 \times$ smaller) than the airsea flux, supporting this idea. This conceptual model suggests that the harbour surface lens functions to capture both gaseous N₂O from the atmosphere and dissolved N₂O generated in the sub-halocline layer and transport it to the ocean in its dissolved form during high-flow periods (Fig. 9).

This study focuses on characterising N_2O dynamics at end-members and at stations through the harbour's longi-

Table 2. Estimated air–sea N₂O flux (mean µmol N₂O m⁻² d⁻¹ ± SD) of the main harbour stations using calculations presented in Bange et al. (2019) and Zhang et al. (2020) and a range of k_{600} parameterisations from Wanninkhof (2014; W_{2014}), Raymond and Cole (2001; RC_{Low}, RC_{Mid}, and RC_{High}), and Nightingale et al. (2000; N_{2000}). Positive values indicate the flux of N₂O from the harbour water to the atmosphere. Negative values indicate flux of N₂O from the atmosphere into the harbour water. Estimated Gordon River flow and Mean (20 d) Gordon Above Denison (GAD) Stream Gauge data are also shown for each month in addition to the Pearson correlation and associated *p* values between flow metrics, rainfall, and air–sea flux (and surface water percentage saturation).

Station	<i>K</i> ₆₀₀	Jul 2022 $\mu mol N_2O$ $m^{-2} d^{-1}$	$\begin{array}{c} {\rm Oct} \ 2022 \\ \mu {\rm mol} \ N_2 {\rm O} \\ {\rm m}^{-2} \ {\rm d}^{-1} \end{array}$	Feb 2023 $\mu mol N_2O$ $m^{-2} d^{-1}$	Apr 2023 μ mol N ₂ O $m^{-2} d^{-1}$	Gordon flow vs. surface flux	GAD flow vs. surface flux	GAD flow vs. % N ₂ O sat.	Rainfall vs. surface flux
KR1	RC _{High} :	-11.07 ± 5.17	-04.01 ± 1.77	-03.30 ± 1.54	-03.17 ± 1.66	r = -0.8316	r = -0.8624	r = -0.8726	r = 0.5577
	RC _{Mid} :	-08.43 ± 4.42 -04.69 ± 3.17	-03.19 ± 1.39 -01.93 ± 1.27	-02.33 ± 1.34 -01.46 ± 0.99	-02.44 ± 1.41 -01.38 ± 0.99	$p = 7.3 \times 10$	$p = 5.1 \times 10$	$p = 2.1 \times 10$	$p \equiv 0.000$
	N2000:	-0.85 ± 0.31	-0.30 ± 0.08	-0.25 ± 0.09	-0.24 ± 0.11				
	W_{2014} :	-0.78 ± 0.25	-0.27 ± 0.05	-0.23 ± 0.07	-0.22 ± 0.09				
C10	RC _{High} :	-12.88 ± 6.00	-01.21 ± 0.53	07.31 ± 3.43	02.60 ± 1.36	r = -0.8298	r = -0.9091	r = -0.8795	r = 0.2751
	RC _{Mid} :	-09.83 ± 5.14	-00.96 ± 0.48	05.65 ± 2.98	02.00 ± 1.16	$p = 8.4 \times 10^{-4}$	$p = 4.2 \times 10^{-5}$	$p = 1.6 \times 10^{-4}$	p = 0.387
	RC _{Low} :	-05.46 ± 3.68	-00.58 ± 0.38	03.22 ± 2.19	01.13 ± 0.81				
	N_{2000} :	-0.99 ± 0.36	-0.09 ± 0.02	0.67 ± 0.23	0.20 ± 0.09				
	W_{2014} :	-0.91 ± 0.29	-0.08 ± 0.02	0.61 ± 0.18	0.18 ± 0.07				
C08	RC _{High} :	-03.50 ± 1.63	-01.69 ± 0.74	04.08 ± 1.91	04.57 ± 2.40	r = -0.8547	r = -0.8804	r = -0.8447	r = 0.1846
	RC _{Mid} :	-02.67 ± 1.40	-01.34 ± 0.67	03.15 ± 1.66	03.52 ± 2.03	$p = 3.97 \times 10^{-4}$	$p = 1.6 \times 10^{-4}$	$p = 5.4 \times 10^{-4}$	p = 0.566
	RCLow:	-01.49 ± 1.00	-0.81 ± 0.53	01.80 ± 1.22	01.98 ± 1.43				
	N_{2000} :	-0.27 ± 0.10	-0.12 ± 0.03	0.31 ± 0.11	0.35 ± 0.15				
	W_{2014} :	-0.25 ± 0.08	-0.11 ± 0.02	0.29 ± 0.08	0.32 ± 0.13				
WH2	RC _{High} :	-10.88 ± 5.06	02.63 ± 1.15	02.40 ± 1.13	03.50 ± 1.84	r = -0.8071	r = -0.8269	r = -0.8077	<i>r</i> = 0.6316
	RC _{Mid} :	-08.30 ± 4.34	02.09 ± 1.04	01.85 ± 0.98	02.69 ± 1.56	$p = 1.51 \times 10^{-3}$	$p = 9.1 \times 10^{-4}$	$p = 1.5 \times 10^{-3}$	p = 0.028
	RCLow:	-04.61 ± 3.11	01.26 ± 0.83	01.06 ± 0.72	01.52 ± 1.09				
	N_{2000} :	-0.84 ± 0.30	0.19 ± 0.05	0.19 ± 0.06	0.27 ± 0.12				
	W_{2014} :	-0.77 ± 0.24	0.17 ± 0.03	0.17 ± 0.05	0.25 ± 0.10				
Gordon (m ³ s ⁻¹)	River flow)	383.6±38.9	360.3 ± 54.1	342.6 ± 74.6	324.3 ± 26.6	-	-	-	-
GAD flo $(m^3 s^{-1})$)	107.6 ± 15.9	73.7±12.1	38.8 ± 5.1	30.5 ± 2.2	_	-	_	-

Table 3. Estimated diapycnal N₂O flux (nmol N₂O m⁻² d⁻¹ ± SD) calculated form local buoyancy frequencies from 20 to 2 m within the main harbour stations. Positive values indicate the flux of N₂O from the basin water (20 m) to the surface lens (2 m).

Station	$\begin{array}{c} July \ 2022 \\ nmol \ N_2 O \ m^{-2} \ d^{-1} \end{array}$	$\begin{array}{c} \text{October 2022} \\ \text{nmol}N_2\text{O}\text{m}^{-2}\text{d}^{-1} \end{array}$	February 2023 nmol N_2O m ⁻² d ⁻¹	$\begin{array}{c} \text{April 2023} \\ \text{nmol N}_2\text{O}\text{m}^{-2}\text{d}^{-1} \end{array}$
KR1	80 ± 3.5	282 ± 17.7	992 ± 12.9	395 ± 8.6
C10	140 ± 4.5	1200 ± 47.3	1040 ± 65.3	454 ± 16.2
C08	49 ± 2.3	782 ± 12.1	778 ± 37.4	348 ± 18.6
WH2	117 ± 4.0	125 ± 2.8	$1,308\pm67.8$	240 ± 18.0

tudinal axis. Other areas of the harbour, most prominently the shallow embayments around the parameter of the system and the areas occupied by fin fish farms, were not included here. Fin fish aquaculture can increase water column DO demand near the pens in this system (Maxey et al., 2020) and introduces particulate organic material to the water. Whether this manifests in altered N cycling dynamics (especially DO-sensitive processes like nitrification and denitrification) would be system specific and has never been described in this system. High particle loads have been shown to induce denitrification in normoxic waters (e.g. Wan et al., 2023; Frey et al., 2020; Codispoti et al., 2005; Nevison et al., 2003; Usui et al., 2001; Robinson et al., 1998), and thus an N_2O sink might be present even under farms even in more oxygenated basins. Future studies should investigate the impacts of fin fish aquaculture on DO and N_2O cycling.

One source of uncertainty in our approach is in using literature-derived estimators for air–sea and diapycnal flux estimations. We also used literature-derived k_{600} estimates from Nightingale et al. (2000), Raymond and Cole (2001), and Wanninkhof (2014) to compute N₂O air– sea flux. Literature-derived estimators of K_{600} and eddy diffusivity are commonly used when direct measurements are unavailable (Tang et al., 2024; Li et al., 2023; Murray et al.,



Figure 8. Mean air-sea flux $(\mu M m^{-2} d^{-1})$ vs. (a) GAD flow $(m^3 d^{-1})$, (b) estimated Gordon River flow $(m^3 d^{-1})$, (c) daily rainfall (mm) (20 d mean), and (d) percentage of estimated Gordon River flow. These variables are accounted for by the GAD Stream Gauge (proxy for hydroelectric dam release) Error bars indicate ± 1 standard error.

2020), but to reduce uncertainty these are ideally measured in situ. Likewise, we presented diapycnal flux estimates using turbulent eddy diffusivities from Fer (2006) that were not measured in Macquarie Harbour.

Previous work in Australian estuaries with pristine catchments (like Macquarie Harbour) has shown that many tend to function as a sink for atmospheric N₂O (Maher et al., 2016; Wells et al., 2018). Our study adds the caveat that water column and atmospheric exchange may also depend on factors controlling river flow in deeper stratified systems. Despite the advancements made to date, many of the deeper estuaries in Chile, Australia, and New Zealand are lacking descriptions of N2O exchange between the water column and atmosphere (e.g. Bathurst Harbour, Tasmania; fjords of the South Island, New Zealand; estuaries on Stewart Island, New Zealand). Given that these systems have relatively pristine catchments, they offer an opportunity to better understand natural fjordlike estuarine responses to the climate drivers of N2O dynamics. Mesoscale climate oscillations (i.e. the SAM and NAO) have been shown to affect rainfall, river flow, and DO concentrations in this and other fjord-like estuaries (Maxey et al., 2022; Austin and Inall, 2002). In western Tasmania, SAM in its positive phase results in increased orographic rainfall and a greater propensity for higher river flow, possibly tilting the source and sink balance to net N₂O uptake during these periods.

Climate change predictions for Tasmania's western Coast (which includes the Macquarie Harbour catchment) indicate that the region will experience a more extreme precipitation regime with increased winter precipitation and decreased summer precipitation (Grose et al., 2010; Bennett et al., 2010). If these future predictions result in more extreme seasonality in Gordon River flow, then the harbour may respond in kind with a larger variation in N2O air-sea flux, i.e. greater N₂O atmospheric uptake in winter and greater N₂O emission in summer. However, given that the river flow is somewhat regulated by the hydroelectric dam, our study suggests that flow regulation has the potential to augment harbour N₂O emissions. Releasing water during extreme low-rainfall periods might allow N₂O slowly accumulating in sub-halocline waters to be released in the exported surface lens. Fjords and fjord-like estuaries are defined by their strong stratification and sensitivity to freshwater inputs. With climate change, rainfall patterns are expected to become more extreme and thus alter the river flow and subsequently N2O source sink dynamics in these systems on a global scale. In systems that are expected to experience increasingly drier conditions, they may shift from net sinks of N2O to sources and further perpetuate the accumulation of N_2O in the atmosphere.

It is well established that fjord and fjord-like estuaries are important sites of C burial (Smith et al., 2015; Bianchi et al., 2018, 2020). This study supports the idea that they can also be important sites of atmospheric N_2O removal and transport. Macquarie Harbour air–sea flux estimates are similar in magnitude to observations made in other stratified estuar-



Figure 9. Conceptual model of Macquarie Harbour's N_2O dynamics. Panel (a) depicts the capture of N_2O generated in the sub-halocline during strong river flow conditions. Here N_2O is exported from the harbour in its dissolved form via undersaturated surface flows from the harbour to the ocean. Panel (b) depicts the efflux of N_2O from the harbour surface during low-flow conditions. Note that during these conditions the surface flows are weak and generally supersaturated with N_2O , permitting its escape in gaseous form to the atmosphere.

ies and enclosed seas such as the Reloncaví Estuary, Chile (Yevenes et al., 2017), and Eckernförde Bay, Germany (Ma et al., 2019) (Table A1). Macquarie Harbour, however, was observed to have lower fluxes of N_2O into the atmosphere than other river-dominated (but not fjord-like) estuaries (Elbe River, Germany; Schulz et al., 2023) including those on the Australian mainland's east coast (Wells et al., 2018).

5 Conclusions

In summary, river flow, and specifically river flow driven by hydroelectric dam release, significantly affects both surface water N₂O concentrations and air–sea flux in Macquarie Harbour. Importantly, when river flow is low, most of the harbour emits N₂O to the atmosphere. When river flow is high, most of the harbour removes N₂O from the atmosphere, intercepts the diapycnal flux, and laterally exports this N₂O to the ocean in its dissolved form. N₂O is continually supersaturated below the halocline, and the relationship between AOU and Δ N₂O and N₂O saturation and NO₃⁻ concentration indicates that the main N_2O generation process is likely nitrification. Climate change is predicted to result in wetter winters and drier summers for the western coast of Tasmania, which may result in augmented N_2O air–sea fluxes. This work represents the first descriptions of N_2O spatiotemporal distribution, estimated air–sea flux, estimated diapycnal flux, and N_2O production pathways in this system.

Appendix A

Table A1. N_2O fluxes and observed ranges of mean (\pm SD) N_2O concentration and saturation from both fjord-like and river-dominated estuaries around the globe and estuaries in Australia.

Location	System type	Measurement depth range	Mean air- sea N ₂ O flux $(\mu Mol N_2 O m^{-2} d^{-1})$		Mean N ₂ O concentration) (saturation) nM N ₂ O (%)	$\begin{array}{ccc} Min & and & max \\ N_2O & concentration \\ (saturation) & nM \\ N_2O \ (\%) \end{array}$	Reference
Macquarie Har- bour, western Tasma- nia, Australia	Fjord- like estuary	2 to 45 m	-09.83 ± 0.67 to 05.65 ± 1.22	-10.82 to 7.73	$11.7 \pm 1.6 \\ (121.8 \pm 17.8)$	7.87 to 17.12 (81 to 174)	This study
Reloncaví Es- tuary, Chile	Fjord- like estuary	0 to 5 m	0.86 ± 2.28	-1.58 to 5.60	$\begin{array}{c} 11.8 \pm 1.70 \\ (111 \pm 18.3) \end{array}$	8.34 to 14.5 (80 to 140)	Yevenes et al. (2017)
Reloncaví Es- tuary, Chile	Fjord- like estuary	10 to 200 m	-	-	$\begin{array}{c} 14.5 \pm 1.73 \\ (145 \pm 17.7) \end{array}$	10.5 to 17.0 (11 to 170)	Yevenes et al. (2017)
Chiloé Interior Sea, Chile	Fjord- like estuary	0 to 200 m	1.08 ± 1.41	-0.18 to 3.19	$\begin{array}{c} 12.6 \pm 2.36 \\ (121 \pm 17.5) \end{array}$	8.81 to 21.1 (87 to 160)	Yevenes et al. (2017)
Europa Sound, Magallanes Re- gion, Chile	Fjord- like estuary	1 to 10 m	-15.22 to -0.81	-	11.9 ± 5.7 to 12.7 ± 1.0	-	Farías et al. (2018)
Concepción Channel, Mag- allanes Region, Chile	Fjord- like estuary	1 to 150 m	0.69 to 7.70	-	13.6 ± 1.1 to 17.0 ± 0.02	-	Farías et al. (2018)
Sarmiento Channel, Mag- allanes Region, Chile	Fjord- like estuary	1 to 10 m	2.07 to 12.53	-	13.1 ± 0.1 to 16.5 ± 0.3	-	Farías et al. (2018)
Estero Peel, Magallanes Re- gion, Chile	Fjord- like estuary	1 to 10 m	0.11 to 2.01	-	13.1 ± 0.2 to 13.5 ± 0.5	-	Farías et al. (2018)
Estero Calvo, Magallanes Re- gion, Chile	Fjord- like estuary	1 to 10 m	0.04	-	13.9 ± 0.8	-	Farías et al. (2018)
Estero Amalia, Magallanes Re- gion, Chile	Fjord- like estuary	1 to 100 m	-0.08	-	14.2±1.7	_	Farías et al. (2018)
Estero las Montañas, Magallanes Region, Chile	Fjord- like estuary	1 to 10 m	-2.95	-	9.69 ± 1.6	_	Farías et al. (2018)
Smyth Chan- nel, Magallanes Region, Chile	Fjord- like estuary	1 to 300 m	1.07 to 11.2	-	14.3 ± 0.4 to 16.0 ± 0.5	-	Farías et al. (2018)
Última Esper- anza Sound, Magallanes Region, Chile	Fjord- like estuary	1 to 10 m	-3.7 to 10.4	-	12.1 ± 1.1 to 13.7 ± 0.07	-	Farías et al. (2018)
Almirante Montt Gulf, Magallanes Region, Chile	Fjord- like estuary	1 to 150 m	15.6	-	21.0±5.7	-	Farías et al. (2018)
Kirke Channel, Magallanes Re- gion, Chile	Fjord- like estuary	1 to 10 m	0.12 to 8.19	-	13.3 ± 0.1 to 15.4 ± 0.4	-	Farías et al. (2018)
Union Channel, Magallanes Re- gion, Chile	Fjord- like estuary	1 to 10 m	22.1	-	16.7 ± 0.8	-	Farías et al. (2018)

Table A1. Continued.

Location	System type	Measurement depth range	Mean air- sea N_2O flux (μ Mol N_2O m ⁻² d ⁻	Min and max air-sea N ₂ O flux ¹) (μ Mol N ₂ O m ⁻² d ⁻	Mean N ₂ O concentration ¹) (saturation) nM N ₂ O (%)	$\begin{array}{ccc} Min & and & max \\ N_2O & concentration \\ (saturation) & nM \\ N_2O \ (\%) \end{array}$	Reference
Union Sound, Magallanes Re- gion, Chile	Fjord- like estuary	1 to 10 m	2.86	-	14.8 ± 0.8	-	Farías et al. (2018)
Western Strait of Magellan, Magallanes Region, Chile	Fjord- like Estuary	1 to 10 m	143	-	15.71	-	Farías et al. (2018)
Eastern Strait of Magellan, Magallanes Region, Chile	Fjord- like estuary	1 m	36.3	-	16.4	_	Farías et al. (2018)
San Gregorio Cape, Magallanes Re- gion, Chile	Fjord- like estuary	1 m	24.8	-	12.07	_	Farías et al. (2018)
Central Otway Sound, Magal- lanes Region, Chile	Fjord- like estuary	1 m	35.5	-	11.4	-	Farías et al. (2018)
Northern Magdalena Channel, Mag- allanes Region, Chile	Fjord- like estuary	1 m	-0.22	-	11.4	_	Farías et al. (2018)
Chasco Sound, Magallanes Re- gion, Chile	Fjord- like estuary	1 m	6.81	_	16.01	_	Farías et al. (2018)
Western Cock- burn Channel, Magallanes Region, Chile	Fjord- like estuary	1 m	6.18	-	14.47	_	Farías et al. (2018)
Saanich Inlet, British Columbia, Canada	Fjord- like estuary	10 to 200 m	2.3 ± 2.5 to 3.9 ± 2.9	-	14.7	< 0.5 to 37.4	Capelle et al. (2018)
Saanich Inlet, British Columbia, Canada	Fjord- like estuary	Surface to 110 m	11.3 to 20.4	-	-	-	Cohen (1978)
Elbe River es- tuary, Germany	Well- mixed river- dominated estuary	1.2 m d	_	26.0 ± 23.5 to 100.7 \pm 101.2	-	(161 ± 53.6) to (243 ± 141.6)	Schulz et al. (2023)
Eckernförde Bay, Boknis Eck Time Se- ries Station, Baltic Sea, Germany	Enclosed sea	1 to 25 m	3.5±12.4	-19.0 to 105.7	(111±30)	(56 to 314)	Ma et al. (2019)
Eckernförde Bay, Boknis Eck Time Se- ries Station, Baltic Sea, Germany	Enclosed sea	1 to 25 m	-	-	10 to 17	-	Walter et al. (2006)

Table A1. Continued.

Location	System Measureme type depth range	ent Mean air- e sea N ₂ O flux (μMol N ₂ O m ⁻² d ⁻	- Min and max a air-sea N ₂ O flux $^{-1}$) (μ Mol N ₂ O m ⁻² d ⁻²	Mean N ₂ O concentration 1) (saturation) nM N ₂ O (%)	$\begin{array}{c} \mbox{Min and max} \\ \mbox{N}_2 O & \mbox{concentration} \\ \mbox{(saturation) nM} \\ \mbox{N}_2 O \end{tabular} \end{array}$	Reference
Baltic Sea, Germany	Enclosed 110 m sea	5–11	-	14 to 1523	-	Rönner (1983)
Gotland Basin, Baltic Sea, Ger- many	Enclosed 90 m sea	-	-	13	0 to 126 (0 to 450)	Brettar and Rhein- heimer (1991)
Northwestern Shelf, Black Sea	Enclosed – sea	1.6 to 4.4	_	6.5 to 8	_	Amouroux et al. (2002)
Northwestern Black Sea Shelf, Black Sea	Enclosed 70 m sea	3.1 to 5.2	-	7.5 to 10.2	_	Amouroux et al. (2002)
Cariaco Basin, Venezuela	Coastal Surface basin 400 m	to –	-	4.4 to 5.5	_	Hashimoto et al. (1983)
Guadalquivir Estuary, Gulf of Cádiz, Spain	River- 2 m dominated estuary	18.7±33.6	-	20.6 ± 24.3	_	Sierra et al. (2020)
Guadalquivir Estuary, Gulf of Cádiz, Spain	River- 2 m dominated estuary	0.3±0.5	_	6.7±0.4	_	Sierra et al. (2020)
Guadalquivir Estuary, Gulf of Cádiz, Spain	River- 2 m dominated estuary	0.9±21.6	_	7.3±15.4	_	Sierra et al. (2020)
Noosa River estuary, eastern Aus- tralia	River- 0.5 to 9.6 n dominated estuary	n -14.24 ± 14.02	-57.72to22.20	6.99 ± 0.43 (97 ± 2.2)	5.92 to 7.95 (90 to 103)	Wells et al. (2018)
Mooloolah River estuary, eastern Aus- tralia	River- 0.5 to 6.8 n dominated estuary	n -7.33±7.25	-48.76to16.31	$\begin{array}{c} 6.74 \pm 0.64 \\ (97 \pm 3.8) \end{array}$	5.19 to 7.71 (82 to 112)	Wells et al. (2018)
Maroochy River estuary, eastern Aus- tralia	River- 0.5 to 8.2 n dominated estuary	n 51.33±55.3	-34.94 to 179.64	$\begin{array}{c} 8.4 \pm 1.50 \\ (113 \pm 16.7) \end{array}$	6.07 to 12.93 (92 to 163)	Wells et al. (2018)
Pine River estuary, eastern Australia	River- 0.5 to 10.1 dominated estuary	m 17.10±39.44	-33.22 to 145.50	$7.1 \pm 0.76 \\ (102 \pm 6.24)$	6.05 to 8.57 (93 to 117)	Wells et al. (2018)
Brisbane River estuary, eastern Aus- tralia	River- 0.5 to 23.9 dominated estuary	m 209.54±143.59	15.42 to 662.62	9.8±1.36 (133±9.9)	6.75 to 12.75 (105 to 158)	Wells et al. (2018)
Middle reach, Brisbane River estuary, eastern Australia	River- Surface dominated estuary	14.5±1.19	5.4 ± 0.34 to 25.2 ± 1.87	-	13.1 to 17.9 (160 to 250)	Sturm et al. (2017)
Lower reach, Brisbane River estuary, eastern Australia	River- Surface dominated estuary	6.±0.51	3.7 ± 0.85 to 9.1 ± 1.19	-	9.2 to 12.7 (125 to 410)	Sturm et al. (2017)
Oxley Creek, eastern Aus- tralia	River- 2.1 to 13.1 dominated estuary	m 210.59 ± 60.23	91.54 <i>to</i> 280.16	$ \frac{11.7 \pm 1.34}{(156 \pm 19.7)} $	9.65 to 14.89 (139 to 199.7)	Wells et al. (2018)

Table A1. Continued.

Location	System Measurement type depth range	$\begin{array}{llllllllllllllllllllllllllllllllllll$	Min and max air-sea N ₂ O flux) (μ Mol N ₂ O m ⁻² d ⁻¹	Mean N ₂ O concentration) (saturation) nM N ₂ O (%)	$\begin{array}{c} \mbox{Min and max} \\ N_2O & \mbox{concentration} \\ (saturation) \ nM \\ N_2O \ (\%) \end{array}$	Reference
Nerang River estuary, eastern Aus- tralia	River- 0.5 to 6.8 m dominated estuary	-0.62 ± 20.87	-67.98 to 45.92	$\begin{array}{c} 6.73 \pm 0.43 \\ (100 \pm 4.3) \end{array}$	5.99 to 7.79 (88 to 109)	Wells et al. (2018)
Logan River estuary, eastern Aus- tralia	– 0.5 to 14.4 m	110.00 ± 153.55	-54.48 to 796.00	9.3 ± 2.36 (127 ± 27.5)	5.54 to 14.8 (81 to 191)	Wells et al. (2018)
Albert River estuary, eastern Australia	– 1.1 to 15.7 m	90.05 ± 73.32	-9.50 to 264.25	$10.10 \pm 2.24 \\ (131 \pm 29.8)$	7.32 to 15.1 (98 to 205)	Wells et al. (2018)
Darwin Creek, Australia	Mangrove $\sim 1 \mathrm{m}$ creek	-0.12	-	6.3 (98.9)	6.0 to 6.8 (95 to 104)	Maher et al. (2016)
Hinchinbrook Creek, Australia	Mangrove $\sim 1 \mathrm{m}$ creek	-3.43	-	6.1 (83.3)	5.6 to 6.8 (75 to 91)	Maher et al. (2016)
Melbourne Creek, Australia	Mangrove $\sim 1 \mathrm{m}$ creek	-1.33	-	7.9 (96.6)	6.9 to 9.1 (86 to 115)	Maher et al. (2016)
Morton Bay Creek, Australia	Mangrove $\sim 1 \mathrm{m}$ creek	-3.19	-	5.1 (77.4)	3.4 to 6.6 (50 to 105)	Maher et al. (2016)
Seventeen Seventy Creek, Australia	Mangrove $\sim 1 \mathrm{m}$ creek	-1.75	_	7.7 (94.3)	7.1 to 8.9 (88 to 106)	Maher et al. (2016)
Brisbane River, Australia		-	_	(285)	(135 to 435)	Musenze et al. (2014)
Coffs Creek, Australia		-	-	(219±37)	(53 to 386)	Reading et al. (2017)
Coffs Creek, Australia		-	-	(266.5±128)	(86 to 678)	Reading et al. (2020)
Boambee Creek, Australia		_	-	(197.1±75)	(87 to 329)	Reading et al. (2020)
Bonville Creek, Australia		-	-	(183.7±65)	(78 to 310)	Reading et al. (2020)
Pine Creek, Australia		-	_	(194.1±65)	(79 to 382)	Reading et al. (2020)
Yarra River, Australia	Salt – wedge estuary	_	_	(135.9±31)	_	Tait et al. (2017)

Data availability. We have used some of the data available in the MEMENTO database. The MEMENTO database is administered by the Kiel Data Management Team at the GEOMAR Helmholtz Centre for Ocean Research Kiel. The database is accessible through the MEMENTO web page: https://memento.geomar.de (Bange et al., 2009).

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