Supplementary information for

Low-pressure storms drive nitrous oxide emissions in the Southern Ocean

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Supplementary Text S1: Calculation of Air-Sea Gas Exchange

The air-sea flux of N_2O depends on the disequilibrium between ocean and atmosphere:

$$\Delta pN_2O = pN_2O_{sw} - pN_2O_{atm} \tag{S1}$$

where ΔpN_2O is the air-sea partial pressure disequilibrium in natm, pN_2O_{sw} is the partial pressure of N_2O in the surface seawater, and pN_2O_{atm} is the partial pressure of N_2O in the atmosphere at the air-sea interface.

We calculated the atmospheric partial pressure of N_2O at the air-sea interface from its atmospheric mixing ratio, the barometric pressure at sea level, and the vapor pressure of water at the air-sea interface. Because one mole of N_2O occupies 0.7% less volume than an ideal gas, we also included a fugacity factor[1] to account for non-ideal behavior:

$$pN_2O_{atm} = XN_2O_{atm} \cdot f \cdot (P - rh \cdot vp_{sw}) \tag{S2}$$

where pN_2O_{atm} is the partial pressure of N_2O in the atmosphere, *f* is a fugacity factor to correct for the non-ideal behavior of N_2O, *P* is the barometric pressure, *rh* is the relative humidity at the air-sea interface, and vp_{sw} is the vapor pressure of seawater. We assumed that the relative humidity at the air-sea interface was 100%, i.e., *rh* = 1.

Using an updated version of an existing gas exchange toolbox[2], we calculated air-sea N_2O flux as the average of two different parameterizations of air-sea gas exchange:

$$F = F_{W14} + F_{L13} \tag{S3}$$

where F denotes the air-sea flux of N_2O associated with a given BGC Argo float profile. F_{W14} and F_{L13} denote the air-sea flux of N_2O in μmol m^{-2} day^{-1} calculated according to ref[3] and ref[4], respectively. The first of these parameterizations of air-sea gas exchange[3] has a single, quadratic dependence on wind speed:

$$F_{W14} = (1 - f_{ice}) \cdot 0.251 \cdot (U_{10})^2 \cdot \left(\frac{Sc}{660}\right)^{-0.5} \cdot k_H \cdot \Delta pN_2O \tag{S4}$$

where f_{ice} is the fractional ice cover, 0.251 is a scaling factor[3], U_{10} is the wind speed at 10 m above the sea surface in m s^{-1}, Sc is the dimensionless water-side Schmidt number of N_2O, k_H is the Henry's Law constant in mol m^{-3} atm^{-1}, and ΔpN_2O is the air-sea partial pressure disequilibrium in atm.

The second gas exchange parameterization[4] has a quasi-linear dependence of diffusion across the air-sea interface but a roughly cubic dependence for bubble-mediated exchange. The total flux is the sum of the diffusive and bubble-mediated components:

$$F_{L13} = -(1 - f_{ice}) \cdot (F_D + F_P + F_C) \tag{S5}$$

where F_{L13} denotes the air-sea flux of N_2O in μmol m^{-2} day^{-1}, F_D is the diffusive component of gas exchange, F_P is the component due to large bubbles that partially dissolve, and F_C is the component due to small bubbles that completely dissolve. Note that the sign of the flux must be reversed so that a positive value corresponds to flux out of the water. These three components are modeled as:

$$F_D = k_s \cdot G_{eq,1atm} \cdot (p_{corr} - \sigma) \tag{S6}$$

$$F_P = k_b \cdot G_{eq,1atm} \cdot \big((1 + \Delta p)p_{corr} - \sigma\big) \tag{S7}$$

$$F_C = XN_2O_{atm} \cdot k_c \tag{S8}$$

where k_s, k_b, and k_c are the piston velocities for the diffusive, large bubble, and small bubble components of gas exchange, respectively. The term Δp denotes the supersaturation maintained by large bubbles that partially dissolve. $G_{eq,1atm}$ is the concentration of N_2O in equilibrium with the atmosphere at 1 atm:

$$G_{eq,1atm} = XN_2O_{atm} \cdot f \cdot (1 - rh \cdot vp_{sw}) \tag{S9}$$

p_{corr} is a correction to the local barometric pressure at sea level:

$$p_{corr} = \left(\frac{P - rh \cdot vp_{sw}}{1 - rh \cdot vp_{sw}}\right) \tag{S10}$$

and σ is the ratio of the seawater and atmospheric partial pressures of N_2O, referenced to 1 atm:

$$\sigma = \frac{pN_2O_{sw}}{XN_2O_{atm} \cdot f \cdot (1 - rh \cdot vp_{sw})} \tag{S11}$$

The wind speed-dependent components of the flux are:

$$k_s = \frac{U_{10} \cdot \sqrt{C_d}}{r_{wt} + r_{at} \cdot \alpha} \tag{S12}$$

$$k_b = 1.98 \times 10^6 \left(U_{10}\sqrt{C_d \, {}^{\rho_a}/_{\rho_w}}\right)^{2.76} \left(\frac{Sc}{660}\right)^{-2/3} \tag{S13}$$

$$k_c = 5.56 \left(U_{10}\sqrt{C_d \, {}^{\rho_a}/_{\rho_w}}\right)^{3.86} \tag{S14}$$

$$\Delta p = 1.5244 \left(U_{10}\sqrt{C_d \, {}^{\rho_a}/_{\rho_w}}\right)^{1.06} \tag{S15}$$

where C_d is the wind speed-based drag coefficient, r_{wt} is the water-side resistance to transfer, r_{at} is the air-side resistance to transfer, and α is the dimensionless solubility of N_2O in seawater, following ref[5]; ρ_a and ρ_w are the densities of air and water, respectively.

These parameterizations[3,4] represent high-end and low-end estimates of N_2O flux respectively, because N_2O is highly soluble (Ostwald solubility of ~1)[1] and thus the nonlinear, bubble-mediated component is likely to be small (Supplementary Fig. 13). We used the ERA5 reanalysis barometric pressure at sea level, sea ice cover, and u- and v-components of the wind speed[6] to calculate air-sea flux.

The total integrated N_2O flux from the Southern Ocean was calculated by summing the total flux from each zone and month:

$$F_t = \sum_{i=1}^{5} \sum_{j=1}^{12} F_{i,j} \tag{S16}$$

where $F_{i,j}$ is the integrated flux in zone *i* and month *j*, in units of $\mu mol\ zone^{-1}\ month^{-1}$. $F_{i,j}$ is calculated by weighting the mean flux in a given zone and month by the area of that zone and number of days in that month:

$$F_{i,j} = \frac{1}{n}\left(\sum_{k=1}^{n} F_k\right) \cdot A_{zone} \cdot D_{month} \tag{S17}$$

where F_k denotes individual fluxes in zone *i* and month *j* in units of $\mu mol\ m^{-2}\ day^{-1}$, *n* is the number of profiles occurring in that zone and month, A_{zone} is the area of that zone in m^2, and D_{month} is the number of days in that month. The areas of each zone were based on previously published values corresponding to climatological mean locations of fronts[18]. Similarly, area- and time-integrated fluxes associated with individual float profiles were calculated from:

$$F_{integrated} = \frac{1}{n}(F) \cdot A_{zone} \cdot D_{month} \tag{S18}$$

where *F* is the flux associated with an individual float profile.

Supplementary references

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