Supplemental material for ACP manuscript "Deposition of dinitrogen pentoxide, N_2O_5, to the snowpack at high latitudes"

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The eddy covariance (EC) method is a more direct method to measure fluxes than the aerodynamic gradient method. However, measurements using the EC method require high-frequency (at least 10 Hz) observations of both air motion and the species interacting with the surface. Because our chemical instruments and inlet have a response time lower than 1 second, we cannot use the EC method to measure N_2O_5 deposition fluxes. However, the meteorological observations were sufficient for measurement of sensible heat fluxes by both the aerodynamic gradient method and the EC method, allowing validation of the gradient method for that property. Using the generalized stability correction factors, atmospheric assumptions, and meteorological data for N_2O_5 flux and applying the same constraints to an aerodynamic method for sensible heat flux we introduce the same level of uncertainty. Using an independent measurement from one sonic anemometer to determine the sensible heat flux by the eddy covariance method, we validate our aerodynamic flux method by correlating the two sensible heat fluxes. Similarly, we have used the sonic anemometer measurements and the EC method to calculate the friction velocity and boundary layer parameters.

S.1 Boundary layer characteristics

Because the aerodynamic method is applicable under very restricted stability conditions, it is necessary to ensure that little change in flux occurs across the observations heights. In addition, the two measurement heights need to remain in the same atmospheric layer. These conditions are achieved if the highest measurement point remains below 10% of the boundary layer height (Oke 1987; Monteith and Unsworth 1990), therefore constraining the measurements in the "constant flux layer". Stull (1988) estimates the constant flux layer to be the lowest 5% of the boundary layer boundary . In addition to different percentages of the constant flux layer, there are several different parameterizations to define the boundary layer height (Zilitinkevich and Baklanov, 2002) we have chosen the most applicable to our data, but note the difficulty in estimating a boundary layer height under stable Arctic conditions (Anderson and Neff, 2008). Because the highest measurement height is 2.38 meters, this criterion requires boundary layer heights of more than 24 meters (at 10% estimate of the constant flux layer). The gradient boundary layer height, H_g, is given by (Arya, 2001)

$$H_g = 1.2 u_* (f N)^{-0.5} \quad . \tag{S.1}$$

In this equation, u_* is the friction velocity, f is the Coriolis parameter, and N is the Brunt-Väisälä frequency, which is given by

$$N^2 = \frac{g}{\theta}\left[\frac{\Delta\theta}{\Delta z}\right] . \tag{S.2}$$

In Equation (S.2), g is the gravitational constant, θ is the potential temperature, and z is the measurement heights. The friction velocity can be calculated using the gradient method by

$$u_* = k \frac{\Delta U}{\left[\ln\left(z_2 / z_1\right)\right]} \left(\Phi_m\right)^{-1} . \tag{S.3}$$

In Equation (S.3), Φ_m, is the stability correction factor for momentum and is unity, less than unity or greater than unity under neutral, unstable or stable conditions respectively. The variable *U,* is the wind speed, *k* the von Karman constant, which has the value 0.4, and the z_i values are the measurement heights. The gradient boundary layer height using Eq. (S.1) ranges from 15 meters to 150 meters for the three nights under study. This calculation ensures that most of the time the sampling remains within the constant flux layer. The average gradient boundary layer height during the observation periods used to measure fluxes was 45 meters.

Reasonable agreement was found between values of the gradient boundary layer height and the EC calculations, which use a direct measurement of turbulent components of the wind; to compute the friction velocity and the Obukhov length (*L*) to determine the boundary layer height. The Obukhov length is a buoyancy length scale of a height of turbulence near the surface (Arya, 2001). The calculation of the friction velocity, u_*, using EC observations is given by Stull (1988)

$$u_* = \left((u'w')^2 + (v'w')^2\right)^{\left(1/4\right)}. \qquad \text{(S.4)}$$

Where the primed variables are the turbulent components and the mean, is represented by a bar over the variable. For example, the vertical component of the wind, *w*, has its fluctuations and means defined by

$$w' = w - \overline{w}. \qquad \text{(S.5)}$$

All properties (*e.g.* vertical wind, *w*) are measured at 10 Hz, and the turbulent quantities are calculated based on the streamline coordinates rotation and mean subtraction by half-hour block with the corresponding de-trending of the signals. Similar separations of fluctuations exist for horizontal wind components, u and v, and temperature, T. The Obukhov length, L, is given by

$$L = -\frac{u_*^3 \overline{\theta}}{kg(\overline{w'\theta'})}. \quad \text{(S.6)}$$

The product $\overline{w'\theta'}$ is the covariance of the vertical wind velocity, w, and the potential temperature, θ. In Equation (S.6), the friction velocity is calculated from high frequency data using Eq. (S.4). The eddy covariance derived stable boundary layer height, H, is (Arya, 2001)

$$H = d(u_* L / f)^{0.5}. \quad \text{(S.7)}$$

In Equation (S.7), d is a constant value of 0.3.The average boundary layer height is 41 meters by the eddy covariance method for the flux measurement periods, in close agreement with the gradient boundary layer height. Therefore, we find that the average boundary layer height from these two methods is 43 meters.

S.2 Comparison of heat flux measured by the eddy covariance method and the aerodynamic gradient method.

To validate the use of the aerodynamic gradient method, we compared sensible heat flux measured by the aerodynamic gradient method to the direct eddy covariance method. The eddy covariance derived sensible heat flux, Q_{EC}, is (Arya , 2001)

$$Q_{EC} = \rho c_p \overline{w'T'}. \quad \text{(S.8)}$$

In Equation (S.8), ρ is the air density, c_p is the heat capacity.

The aerodynamic gradient method can also be used to calculate the sensible heat flux, Q_{aero}, using an equation similar to Eq. (2) in the main paper, (Arya, 2001)

$$Q_{aero} = -k^2 \rho c_p \frac{\Delta \bar{u} \Delta \bar{T}}{[\ln(z_2/z_1)]^2} (\Phi_M \Phi_h)^{-1} \qquad \text{(S.9)}$$

Note that the same general stability flux correction factors are used in Eq. (S.9) and Eq. (2) in the main text. The use of these correction factors is critical in arriving at a good correlation between the two methods shown below, and thus it is important that we consistently use these validated factors in the calculation of N_2O_5 deposition velocity in the main text.

Figure S.1 shows the heat fluxes measured by the two different methods, which are in good agreement with an $r^2 = 0.76$, and importantly, the slope is within standard error of unity. The heat flux ranged from +8 to -70 W/m^2, with the average heat flux of -5.4 W/m^2. At the larger negative heat flux values, the atmosphere becomes increasingly stable, requiring large stability correction factor. The effect of these large corrections is seen in the high scatter at negative heat flux values.

An alternative method, suggested by a referee, to calculate the flux of N_2O_5 uses the sonic anemometer data to calculate the effective tracer diffusivity and multiples that by the observed chemical gradient of N_2O_5. Use of that method results in a deposition velocity of 0.49 +/- 0.58 cm/s. The resulting deposition velocity is 0.1 cm/s (17%) lower, but well within the variability/error of the gradient method, indicating that the two methods are equivalent.

S.3 Estimation of height of N_2O_5 snowpack deposition

In a stable boundary layer, the flux is a function of height, with a maximal value at the surface that decreases to zero at the top of the boundary layer. The shape of this flux profile is not known N_2O_5, but by extension of similarity theory, we expect that the shape of the deposition flux profile is similar to that of the momentum flux profile, $\tau(z)$. Micrometeorological observations of the stable boundary layer typically found in the Arctic indicate that the momentum flux profile is described by a power law, (Arya, 2001)

$$\tau(z) = \rho u_*^2 (1 - z/H)^{\alpha_1} \quad . \qquad \text{(S.10)}$$

The exponent value, α_1, can change depending upon the development of the stable boundary layer (Arya, 2001), but is often assumed to be 1.75 (Lenschow et al., 1988; Smedman 1991). If we assume the same power law dependence for the flux profile of N_2O_5, we find that the vertical dependence of the flux is

$$F_{N_2O_5}(z) = \left(F_{N_2O_5}\right)_{surf} (1 - z/H)^{\alpha_1} \quad . \qquad \text{(S.11)}$$

In this equation, the surface flux, which we have observed, is indicated by the "surf" subscript, and the flux is a function of height, z. We define the effective height for N_2O_5 deposition, z_{eff}, by the height of the box profile (constant flux with height through some effective height, z_{eff} with the same integral as the power law flux profile, Eq. (S.11). The result is

$$z_{eff} = \frac{H}{1 + \alpha_1} \, . \qquad \text{(S.12)}$$

We use the average boundary layer height, $H = 43$ meters, and $\alpha_1 = 1.75$, to find that $z_{\mathrm{eff}} = 15$ meters.

S.4 Estimation of required fetch

The placement of a flux measurement tower must allow for adequate fetch distance of a minimum of 100 meters per meter of the measurement height of the tower (Businger et al., 1971). The fetch distance is said to be farther in the case of stable atmospheric conditions and more than 100 times the maximum height of the measurement tower (Horst and Weil, 1994).

We estimated the required fetch distance for a 2.38 meter measurement height using the relationship between height and surface roughness as proposed in Horst and Weil (1994) as Z_m/Z_0. This ratio is the normalized measurement height, where Z_m is the maximum measurement height in meters and Z_0 is the roughness length in meters. The roughness length (Z_0) indicates the roughness of the surface and effect on neutral stability (Stull, 1988).The ratio of Z_m/Z_o , using 2.38 m and 0.005 roughness length for snow from Stull (1988) is 476. Using these values and method described in Horst and Weil (1994), we would need 1,000 meters of required fetch to calculate a flux under the most stable atmospheric conditions. To avoid reaching our fetch estimated distance under the most stable conditions, with the stability characterized by a Richardson number of 0.25, we use a conservative stability range of -0.1 to 0.12 for the Richardson number.

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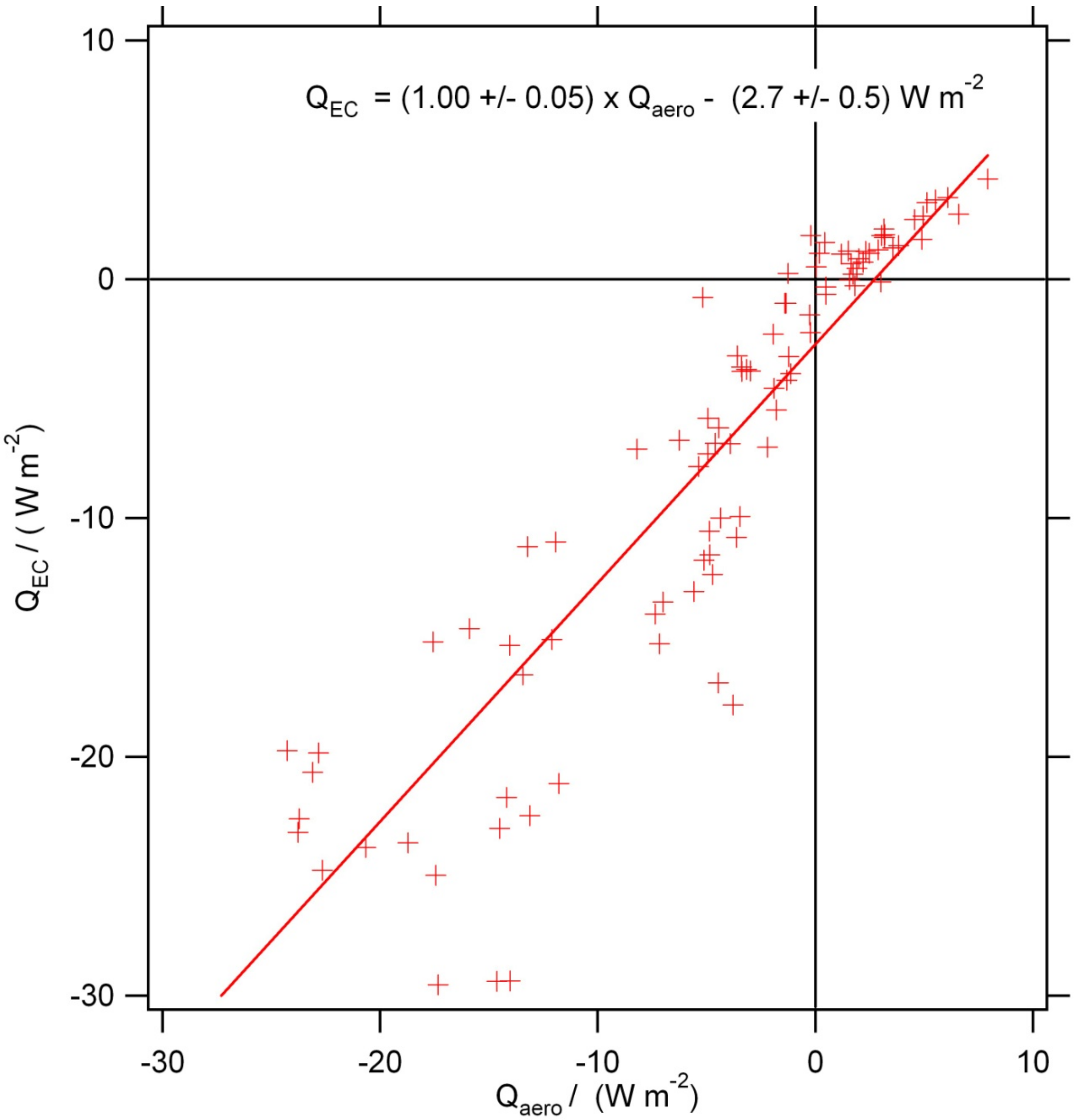

Figure S.1: Sensible heat flux by the eddy covariance method, Q_{EC}, versus sensible heat flux by the aerodynamic gradient method, Q_{aero}. The errors are reported as +/- 1 standard deviation.