Supporting Information for "Intrinsic constraints on asymmetric turbulent transport of scalars within the constant-flux layer of the lower atmosphere"

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Introduction

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In this document, the inference of a and b parameters in ϕ_{ww} is first presented. As shown in Fig. S1, the choice of parameters $a = 1$ and $b = 4$ recover the ϕ_{ww} measurements even when the scalar fluxes are not constant with z. This agreement is conditioned on using local fluxes of heat and momentum in the $-\zeta$ calculations and u_* normalization.

The document also includes two sections discussing the sufficient conditions for scalar fluxes to be z-independent (Text S1) and how to extend the framework to other scalars (Text S2, and Fig. S2- S4).

Lastly, Fig. S5 shows the variations of ΔS_o and γ with ζ for θ at the dryland site. It is clear that both variables become functions of ζ when measured fluxes among the four levels have to be within 10% of each other, which can be described by the same fitted functions for θ at the lake site.

Text S1: The sufficient conditions for scalar fluxes to be z-independent

The 'sufficient conditions' for $h = 1$ include 1) f is only a function of ζ, and 2) $\kappa_v \zeta(\partial f/\partial\zeta) = 1/g_1 - 1/g_2 = 0$. To explore further, we write that $\overline{w'\theta'} = (\overline{w'\theta'})_s + (\overline{w'\theta'})_{ns}(z)$ where $(\overline{w'\theta'})_{ns}$ is a function of z introduced by the vertical transport term. For $\overline{w'\theta'}$ to be z-independent and equal to the surface flux, $(\overline{w'\theta'})_{ns}$ must be identically zero at any z. Note that $(\overline{w'\theta'})_{ns}$ is zero when $z = 0$ by definition. Substituting $\overline{w'\theta'} = (\overline{w'\theta'})_s + (\overline{w'\theta'})_{ns}(z)$ into equation 12 of the main paper yields

$$\kappa_v \zeta f \frac{\partial h_{ns}}{\partial \zeta} + \left(\kappa_v \zeta \frac{\partial f}{\partial \zeta} + \frac{1}{g_2}\right)(1 + h_{ns}) = \frac{1}{g_1}, \tag{1}$$

where $h_{ns} = (\overline{w'\theta'})_{ns}/(\overline{w'\theta'})_s$ and thus $h = 1 + h_{ns}$. With $1/g_1 = 1/g_2$ and $\zeta\partial f/\partial\zeta = 0$, the above equation reduces to

$$\kappa_v \zeta f \frac{\partial h_{ns}}{\partial \zeta} + \frac{1}{g_2} h_{ns} = 0. \tag{2}$$

Given that f and g_2 are only functions of ζ, h_{ns} must be only a function of ζ. Furthermore, the fact that h_{ns} is zero when $z = 0$ (i.e., when $\zeta = 0$) requires h_{ns} to be identically 0 at any ζ.

Text S2: Extension to other scalars

The analysis can be extended to scalars other than θ such as water vapor q and CO_2 concentration. As alluded to in the main text, the buoyancy term involves the covariance $\overline{\theta' s'}$ instead of $\overline{\theta'\theta'}$ for s other than θ. Following the same derivation, the necessary conditions for scalar flux to be z-independent remain: 1) g_1, g_2, and $f = \overline{w'w's'}/(u_*\overline{w's'})$ are only functions of ζ, and 2) $\kappa_v\zeta(\partial f/\partial\zeta) = 1/g_1 - 1/g_2$. However, the stability correction function for the relaxation time scale g_1 now involves $R = \theta_*\overline{\theta' s'}/(s_*\overline{\theta'\theta'})$, where $s_* = -\overline{w's'}(z=0)/u_*$, as follows

$$g_1 = \left[\phi_{ww}^2\phi_s - \alpha\zeta\phi_{\theta\theta}^2 R\right]^{-1}, \tag{3}$$

where $\phi_s = \kappa_v z/s_*(\partial\overline{s}/\partial z)$. R can be further simplified to $R = R_{\theta s}(R_{w\theta}/R_{ws})$ where $R_{\theta s} = \overline{\theta' s'}/(\sigma_\theta\sigma_s)$, $R_{ws} = \overline{w's'}/(\sigma_w\sigma_s)$, $R_{w\theta} = \overline{w'\theta'}/(\sigma_w\sigma_\theta)$. It is clear that R needs to be only a function of ζ for g_1 to be only a function of ζ. For water vapor q, the variation of R with respect to ζ has been studied previously by *Katul et al.* [2016] using the lake data and the scatter is non-trivial under near-neutral conditions. However, it is pointed out that what truly matters for g_1 is $-\zeta R$ (see equation 3), whose variation with $-\zeta$ when the constant-flux assumption is satisfied at the 5% level is shown in Fig. S2. The scatter is significantly reduced when compared to the results in *Katul et al.* [2016] and $-\zeta R$ can be viewed as only a function of ζ. This further results in that f is only a function of ζ, as shall be confirmed later in Fig. S4.

The generality of the work here to other scalars is demonstrated using the eddy-covariance data earlier described. To ensure that significant amount of data segments that satisfy the constant-flux assumption remain, additional filtering for $s = q$ is required

at the dryland site. Close inspection reveals that the LE at 2 m at the dryland site is significantly lower than those at the other three levels even if the criterion is increased to 20%. In addition, the CO_2 fluxes do not satisfy the constant-flux layer assumption at the two sites due to intermittent respiration bursts. As a result, only the water vapor flux at the lake site is considered.

Fig. S3 compares the modeled and measured f for q at the lake site and shows that the ICEM performs reasonably. Fig. S4 examines the variations of ΔS_o and γ with ζ for q at the lake site. It can be seen that as the criteria for 'constant' flux become more stringent, the scatter is reduced. When the fluxes among the four levels have to be within 5% of each other, ΔS_o and γ are only functions of ζ, which can be described by the same fitted functions for θ, highlighting the universal character of ΔS_o and γ when the constant-flux condition is imposed.

References

Katul, G. G., D. Li, H. Liu, and S. Assouline (2016), Deviations from unity of the ratio of the turbulent schmidt to prandtl numbers in stratified atmospheric flows over water surfaces, *Phys. Rev. Fluids*, *1*(3), 034,401.

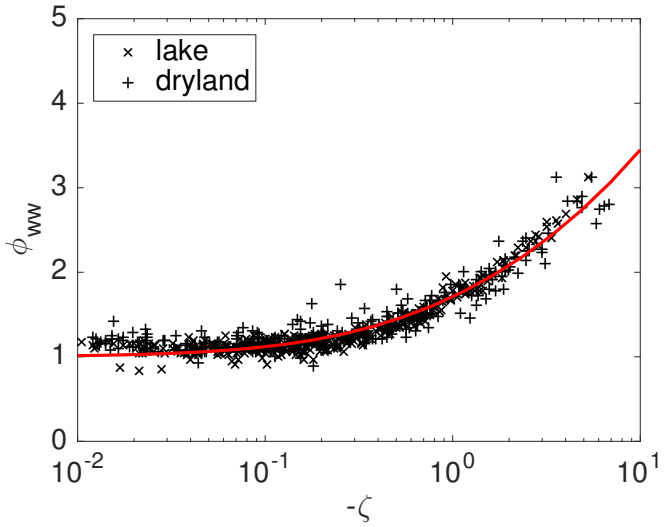

Figure S1. The variation of ϕ_{ww} with $-\zeta$ at the lake and dryland sites. The fitted red line is $\phi_{ww} = (1 - 4\zeta)^{1/3}$.

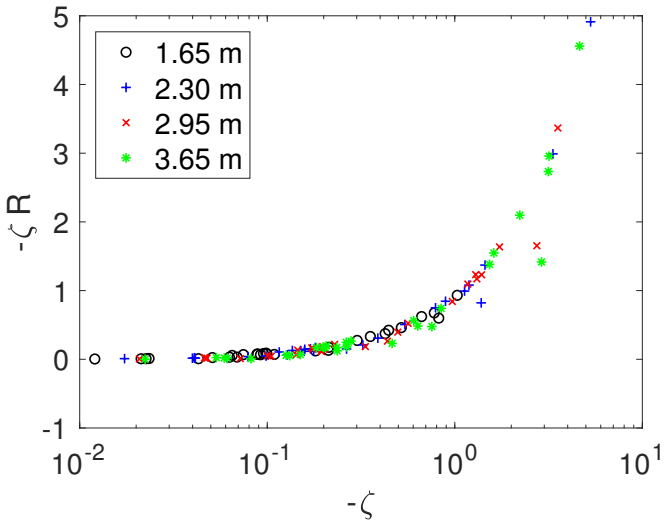

Figure S2. The variation of $-\zeta R$ with $-\zeta$ at the lake site. Here R is caculated for q.

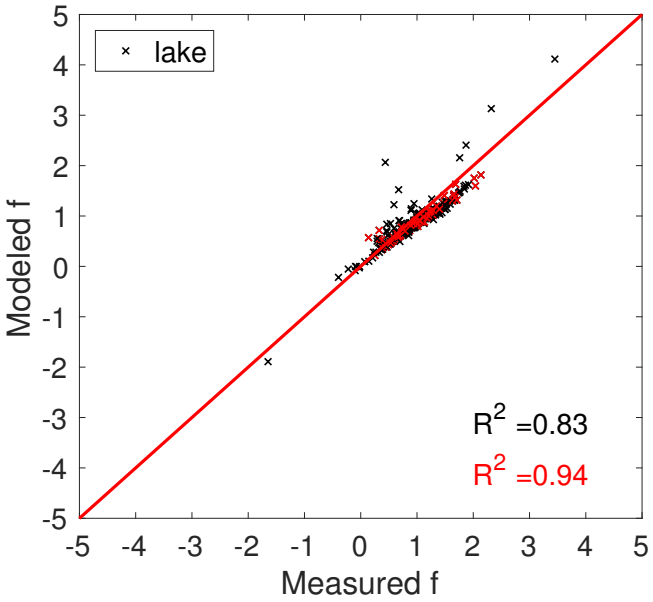

Figure S3. The comparison between measured and modeled f for q at the lake site. The black markers represent cases when no control is imposed on turbulent flux variations with z among the four levels, and the red markers represent cases when fluxes among the four levels have to be within 5% of each other.

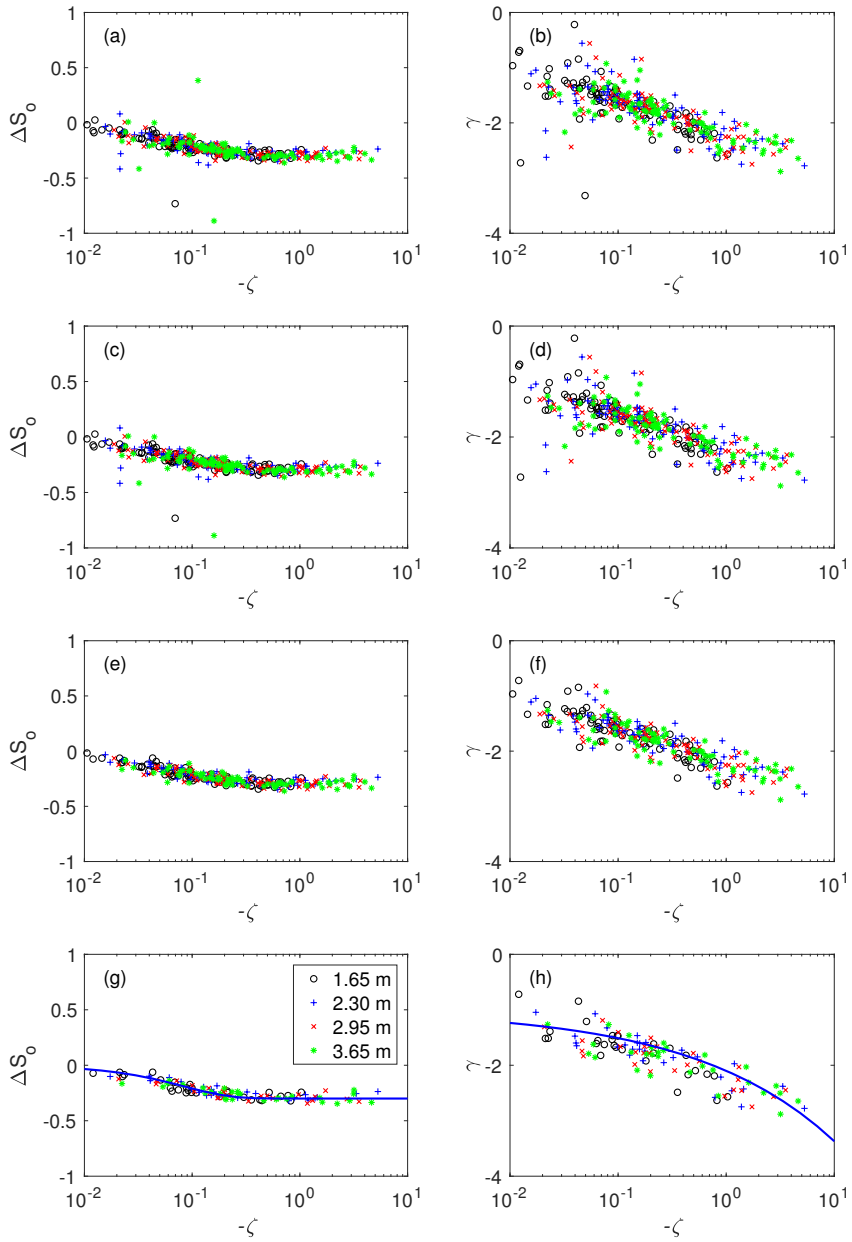

Figure S4. The ΔS_o (left) and γ (right) for q at the lake site when (a, b) no control is imposed on flux variations among the four levels, (c, d) fluxes among the four levels have to be within 50% of each other, (e, f) fluxes among the four levels have to be within 10% of each other, and (g, h) fluxes among the four levels have to be within 5% of each other. The fitted blue lines in (g, h) are $\Delta S_o = 0.3\left[e^{12\zeta} - 1\right]$, $\gamma = -1.1\,(-\zeta)^{1/3} - 1$.

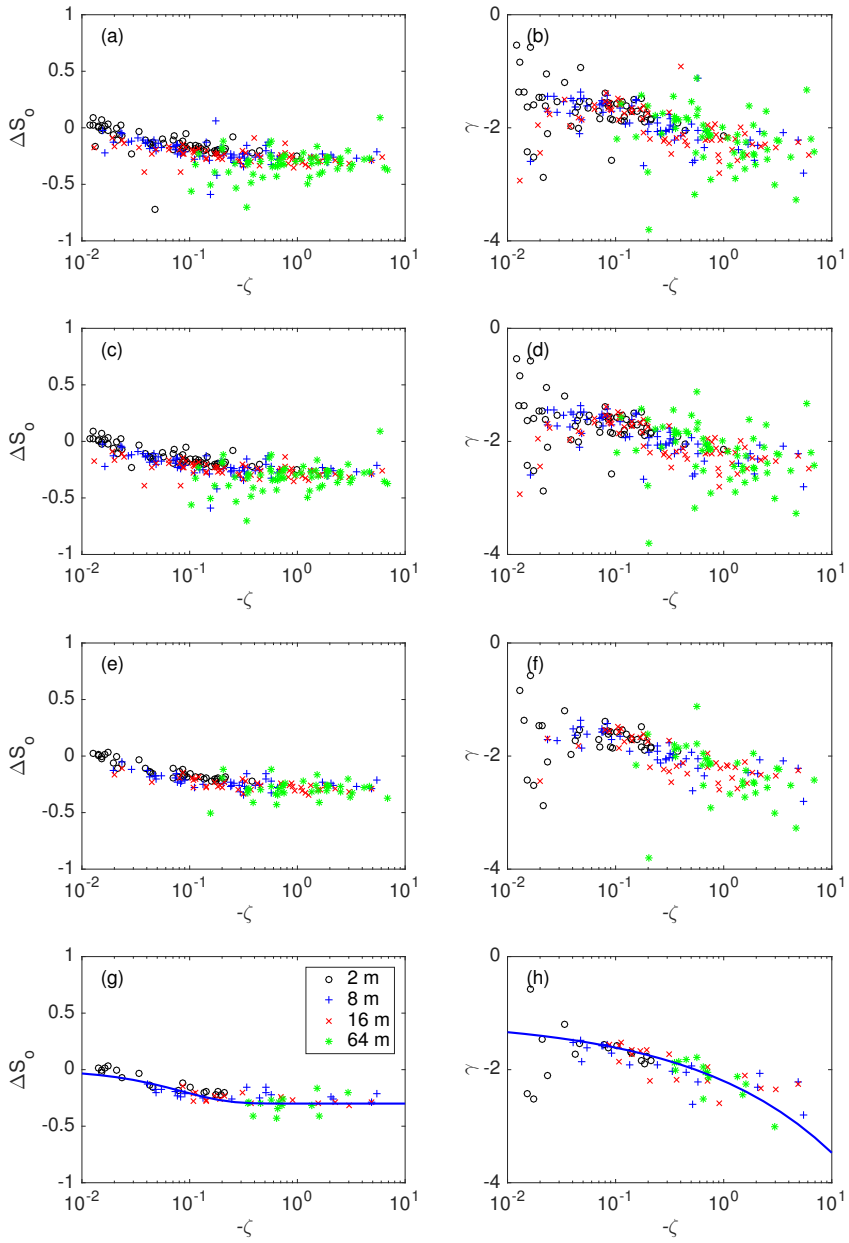

Figure S5. The ΔS_o (left) and γ (right) for θ at the dryland site when (a, b) no control is imposed on flux variations among the four levels, (c, d) fluxes among the four levels have to be within 100% of each other, (e, f) fluxes among the four levels have to be within 20% of each other, and (g, h) fluxes among the four levels have to be within 10% of each other. The fitted blue lines in (g, h) are $\Delta S_o = 0.3\left[e^{12\zeta} - 1\right]$, $\gamma = -1.1\,(-\zeta)^{1/3} - 1$.