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E. Zorzetto, A. D. Bragg, and G. Katul Phys. Rev. Fluids **3**, 094604 — Published 10 September 2018 DOI: 10.1103/PhysRevFluids.3.094604

Extremes, intermittency and time directionality of atmospheric turbulence at the cross-over from production to inertial scales

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Abstract

The effects of mechanical generation of turbulent kinetic energy and buoyancy forces on the 1 statistics of air temperature and velocity increments are experimentally investigated at the cross 2 over from production to inertial range scales. The ratio of an approximated mechanical to buoy-3 ant production (or destruction) of turbulent kinetic energy can be used to form a dimensionless 4 stability parameter ζ that classifies the state of the atmosphere as common in many atmospheric 5 surface layer studies. We assess how ζ affects the scale-wise evolution of the probability of ex-6 treme air temperature excursions, their asymmetry and time directionality. The analysis makes 7 use of high frequency turbulent velocity and air temperature time series measurements collected 8 at z=5 m above a grass surface at very large frictional Reynolds numbers $Re_* = u_* z/\nu > 1 \times 10^5$ 9 $(u_*$ is the friction velocity and ν is the kinematic viscosity of air). A multi-time measure of the 10 disbalance between forward and backward phase-space trajectories is employed to investigate the 11 time-directional properties of the scalar (temperature) field. Using conventional higher-order struc-12 ture functions, we find that temperature exhibits larger intermittency and wider multifractality 13 when compared to the longitudinal velocity component, consistent with laboratory studies and 14 simulations conducted at lower Re_* . We find that the magnitude of ζ , rather than the sign of the 15 heat flux, impacts the distribution of scalar increments at separation scales well within the inertial 16 subrange. Conversely, the direction of the heat flux fingerprints the observed time-directionality 17 properties of the scalar field in the first two decades of inertial sub-range scales. These combined 18 findings demonstrate that external boundary conditions, and in particular the magnitude and sign 19 of the sensible heat flux, have a significant impact on temperature advection-diffusion dynamics 20 within the inertial range. 21

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22 I. INTRODUCTION

Turbulence in fluids is prototypical of spatially extended nonlinear dissipative systems 23 characterized by large fluctuations that are active over wide ranging scales [1]. The dynam-24 ics of a substance or scalar advected by a turbulent flow (often termed 'scalar turbulence' 25 [2]) is by no means an exception to this description. Scalar turbulence shares many phe-26 nomenological parallels with the much studied turbulent velocity fluctuations, especially in 27 the inertial subrange. However, scalar turbulence also exhibits distinctive large- and fine-28 scaled temporal patterns (e.g. ramp-cliff) that are usually weak or all together absent from 29 their component-wise turbulent velocity counterparts [2–4]. This finding is particularly true 30 in the atmospheric surface layer (ASL) [5, 6], a layer within the atmospheric boundary layer 31 (ABL) that is sufficiently far above roughness elements but not too far from the ground 32 to be directly impacted by the Coriolis force. In the ASL, the frictional Reynolds number 33 $Re_* = u_* z / \nu$ can readily exceed 10⁵, where z is the distance above the ground surface, u_* is 34 the friction velocity related to the kinematic turbulent stress, and ν is the kinematic viscos-35 ity of air. A direct consequence of this large Re_* is a wide separation between scales over 36 which turbulent kinetic energy (k) is produced and dissipated. In the absence of thermal 37 stratification, k is produced at scales commensurate with z; however, the action of fluid 38 viscosity responsible for the dissipation of k occurs at scales commensurate to or smaller 39 than the Kolmogorov microscale $\eta_K = (\nu^3/\langle \epsilon \rangle)^{1/4}$, where $\langle \epsilon \rangle$ is the mean turbulent kinetic 40 energy dissipation rate that is proportional to u_*^3/z for a neutrally stratified ASL [6]. These 41 estimates of $\langle \epsilon \rangle$ and η_K result in $z/\eta_K \sim Re_*^{3/4} > 5000$ in the ASL, which is rarely achieved 42 in direct numerical simulations or laboratory studies. Embedded in this wide ranging scale 43 separation is the inertial subrange [7], where self similar scaling of velocity and air temper-44 ature structure functions is expected to hold for eddy sizes much larger than η_K but much 45 smaller than z. Integral scales or scales comparable to z are directly influenced by boundary 46 conditions imposed on the flow including surface heating (or cooling) in the ASL, whereas 47 small scales (e.g. η_K) may attain universality and local isotropy after a large number of 48 cascading steps away from the energy injection scales. 49

⁵⁰ Much attention has been historically dedicated to the inertial subrange and the subse-⁵¹ quent cross-over to the viscous or molecular regimes precisely because of the possible uni-⁵² versal character of turbulence at such fine scales [4, 8–12]. However, it is now accepted that

some coupling between small and large scales exists, especially for passive scalars [2, 4, 13], 53 that act to enhance intermittency buildup across scales and distort any universal behavior 54 by injecting the effects of the boundary conditions (or the k generation mechanism). Along 55 similar lines of inquiry, it has been conjectured that the presence of coherent ramp-cliff 56 patterns in concentration (or temperature) time series are responsible, to some degree, for 57 this coupling [4]. Ramp-cliff structures are characterized by local intense scalar gradients 58 separated by large quiescent regions. The presence of ramp-cliff structures in scalar time 59 series has been shown to break locality of eddy interactions and determine some departures 60 from small scale isotropy. 61

Sweep-ejection dynamics connected to the presence of ramps are likely to play a major 62 role in observed extreme value statistics, as shown e.g., for Lagrangian velocity sequences in 63 plant canopy turbulence [14]. Moreover, ramps are asymmetric and produce non-zero odd 64 ordered structure functions, sharing striking resemblance with flight-crash events recently 65 reported for the turbulent kinetic energy of Lagrangian particles [15]. Even though ramps 66 have been extensively observed experimentally [3], studied as surface renewal processes [13], 67 and from a Lagrangian perspective [2, 16], a unified picture describing their effects on inertial 68 scales statistics remains lacking and motivates the work here. 69

Our main objective is to investigate two questions about scalar turbulence at scales span-70 ning production to inertial subranges: How do ramp-cliff patterns modify (i) the probability 71 of extreme scalar concentration or air temperature excursions and its corollary intermit-72 tency buildup, and (ii) symmetry and time reversibility of scalar turbulence. These two 73 questions are explored for differing turbulent energy injection mechanisms (mechanical and 74 buoyancy forces) in the ASL. Here we focus on the production-to-inertial scales instead of 75 the usual inertial to viscous ranges for the following reasons. First, any cross-scale coupling 76 with ramp-cliff patterns is likely to be sensed at large scales commensurate with the ramp 77 durations. Second, these scales are deemed most relevant when constructing sub-grid scale 78 models for improving Large Eddy Simulations [17–20]. Third, these scales encode much 79 of the scalar variance that is needed when deriving phenomenological theories for the bulk 80 flow properties based on the spectral shapes of the turbulent velocity and air temperature 81 [21–25], especially for the ASL. 82

To achieve the study objectives, high frequency measurements of the three velocity components and air temperature fluctuations in the ASL are used to explore flow statistics at

the transition from production to inertial scales. In particular, the focus is on the first two 85 decades dominated by approximate inertial subrange effects, where the transition from the 86 large eddies to the universal equilibrium or inertial range occurs. The statistical properties 87 of temperature increments within this range of scales is examined with the goal of addressing 88 to what extent the tail properties (and thus the probability of extreme events) at fine scales 89 still carry signatures from the production ranges and in particular of large coherent struc-90 tures such as ramp-cliffs. The experiments here span several atmospheric stability regimes 91 that dictate to what degree turbulent kinetic energy is mechanically or buoyantly generated 92 (or dissipated) depending on surface heating (or cooling) and on the turbulent shear stress 93 near the ground [26]. However, due to the large Reynolds number encountered in the ASL, 94 the stable stratification is not sufficiently severe to allow for a transition to non-turbulent 95 regimes. Therefore, the turbulence can be studied as three dimensional and fully developed. 96 The manuscript is organized as follows: In section II, the budget for turbulent kinetic 97 energy forced by a mean velocity gradient and buoyancy is reviewed so as to define the key 98 variables and dimensionless quantities pertinent to ASL flows. Then, the statistical tools 99 used to characterize intermittency and time directionality of the scalar field are introduced. 100 Section III presents the experimental setup, data processing, and compares the outcome of 101 this experiment with predictions from traditional turbulence theory in the inertial subrange. 102 The results obtained investigating extreme values and time directional properties for velocity 103 and temperature are then presented in section IV. In section V the main conclusions are fea-104 tured. The appendix shows that distortions of the inertial range due to stable stratification 105 are not relevant for the range of scales studied here. 106

107 II. THEORY

¹⁰⁸ A. Overview of ASL similarity at large- and small-scales

The turbulent kinetic energy budget for a stationary and planar homogeneous flow in the absence of subsidence is given by

$$\frac{\partial k}{\partial t_0} = 0 = -\overline{u'w'}\frac{dU}{dz} + \beta_o g\overline{w'T'} + P_D + T_T - \epsilon, \qquad (1)$$

where $k = (\overline{u'^2 + v'^2 + w'^2})/2$ is the turbulent kinetic energy, u', v', and w' are the turbulent velocity components along the mean wind (or x), lateral (or y), and vertical (or z) directions,

respectively, t_0 is time, and the five terms on the right-hand side of Eq. (1) are mechanical 113 production, buoyant production (or destruction), pressure transport, turbulent transport of 114 k, and viscous dissipation of k, respectively, β_o is the thermal expansion coefficient for gases 115 $(\beta_o = 1/T, T \text{ is air temperature here}), g \text{ is the gravitational acceleration}, -\overline{u'w'} = u_*^2$ is the 116 turbulent kinematic shear stress near the surface, and $\overline{w'T'}$ is the kinematic sensible heat 117 flux from (or to) the surface. When $\overline{w'T'} > 0$, buoyancy is responsible for the generation 118 of k and the ASL is classified as unstable. When $\overline{w'T'} < 0$, the ASL is classified as stable 119 and buoyancy acts to diminish the mechanical production of k. The relative significance of 120 the mechanical production with respect to the buoyancy generation (or destruction) may be 121 expressed as 122

$$-\overline{u'w'}\frac{dU}{dz} + \beta_o g\overline{w'T'} = \frac{u_*^3}{\kappa z} \left[\phi_m(\zeta) + \frac{\kappa z \beta_o g\overline{w'T'}}{u_*^3}\right] = \frac{u_*^3}{\kappa z} \left[\phi_m(\zeta) - \zeta\right],\tag{2}$$

123 where

$$\frac{dU}{dz} = \frac{u_*}{\kappa z} \phi_m(\zeta), \quad \zeta = \frac{z}{L}, \quad L = -\frac{u_*^3}{\kappa g \beta_o \overline{w'T'}},\tag{3}$$

and $\phi_m(\zeta)$ is known as a stability correction function reflecting the effects of thermal stratifi-124 cation on the mean velocity gradient ($\phi_m(0) = 1$ recovers the von Karman-Prandtl log-law), 125 $\kappa \approx 0.4$ is the von Karman constant, and L is known as the Obukhov length as described by 126 the Monin and Obukhov similarity theory [26]. The physical interpretation of L is that it is 127 the height at which mechanical production balances the buoyant production or destruction 128 when $\phi_m(\zeta)$ does not deviate appreciably from unity. For a neutrally stratified ASL flow, 129 $|L| \to \infty$ and $|\zeta| \to 0$. The sign of L reflects the direction of the heat flux, with nega-130 tive values of L corresponding to upward heat fluxes (unstable atmospheric conditions) and 131 positive values of L corresponding to downward heat flux (stable atmosphere). 132

Several bulk flow statistics in the ASL can be reasonably described by the aforementioned 133 Monin-Obukhov similarity theory, including the mean air temperature gradient dT/dz and 134 the air temperature variance $\overline{T'^2}$, both varying with ζ when normalized by a temperature 135 scale $T_* = -\overline{w'T'}/u_*$. However, the statistics of some large-scale features within the tem-136 perature time series traces, such as the statistics of ramp-cliff patterns, do not scale with 137 z. For starters, the ramp characteristic dimension is generally larger than z and their du-138 ration exceeds the mean vorticity time scale $(\kappa z \phi_m(\zeta)^{-1}) u_*^{-1}$. Ramps have been observed 139 within canopies, near the canopy atmosphere interface, and other types of flows as reviewed 140 elsewhere [4, 13]. While z/L may not be the proper scaling parameter for ramps, it does 141

indirectly impact many of their features in air temperature time traces sampled within the
ASL. For example, in stably stratified ASL flows, the temperature ramps appear 'inverted'
when compared to their near-neutral counterparts. The amplitudes and durations of ramps
can increase with increasing instability due to weaker shearing and intense buoyant updrafts
[27, 28].

At small scales associated with the inertial subrange, the velocity and temperature secondorder structure functions are commonly described by the Kolmogorov 1941 (K41) theory [7] given as

$$S_u^2(r) = \overline{[\Delta u(r)]^2} = 4C_{o,u}(\langle \epsilon \rangle r)^{2/3}, \qquad (4)$$

150

$$S_w^2(r) = \overline{[\Delta w(r)]^2} = 4C_{o,w}(\langle \epsilon \rangle r)^{2/3}, \tag{5}$$

151

$$S_T^2(r) = \overline{[\Delta T(r)]^2} = 4C_{o,T} \langle \epsilon_T \rangle \langle \epsilon \rangle^{-1/3} r^{2/3}, \tag{6}$$

where $\Delta u(r) = u(x+r) - u(x)$, $\Delta w(r) = w(x+r) - w(x)$, and $\Delta T(r) = T(x+r) - T(x)$ are the velocity and temperature increments at separation distance (or scale) r, $\langle \epsilon \rangle$ and $\langle \epsilon_T \rangle$ are the k and temperature variance dissipation rates respectively, $C_{o,u}$ and $C_{o,w}$ are the Kolmogorov constants for the longitudinal and vertical velocity components, and $C_{o,T}$ is the Kolmogorov-Obukhov-Corrsin (KOC) constant. These scaling laws, obtained under the assumptions of similarity and local isotropy, appear to hold reasonably in the ASL for scales smaller than z/2 [29]. Moreover, the normalized third order structure functions

$$S(r) = \frac{S_u^3}{\left(S_u^2\right)^{3/2}} = \frac{\langle \Delta u(r)^3 \rangle}{\langle \Delta u(r)^2 \rangle^{3/2}} \tag{7}$$

159 and

$$F(r) = \frac{S_{TTu}^3}{S_T^2 \left[S_u^2\right]^{1/2}} = \frac{\langle \Delta u(r)\Delta T(r)^2 \rangle}{\langle \Delta T(r)^2 \rangle \langle \Delta u(r)^2 \rangle^{1/2}}$$
(8)

must be constant to recover K41 predictions for S_u^2 and S_T^2 in the inertial range [30].

¹⁶¹ However, relevant deviations from K41 scaling have been reported for higher order struc-¹⁶² ture functions, especially for the scalar fluctuations. These deviations arise as (i) Eqs. (4) -¹⁶³ (6) do not account for intermittency related to spatial variability of the actual ϵ and ϵ_T , and ¹⁶⁴ (ii) the hypothesis of local isotropy might not hold for scalars due to non-local interactions ¹⁶⁵ across scales [31]. A signature of the latter is the large structure skewness for temperature ¹⁶⁶ determined by ramp structures [4, 29]. Many models, starting from Kolmogorov's log-normal ¹⁶⁷ dissipation rate refinement [32], seek to relax some of the restrictive assumptions of K41 so as to explain the anomalous scaling observed in higher order moments. For scalars, these
 corrections are commonly expressed as

$$S_T^n = C_n \left(\epsilon r\right)^{n/3} \left(r/L_I\right)^{\zeta_n' - n/3} \tag{9}$$

where the exponent ζ'_n implies a scaling different from K41 that depends on the moment order *n*. The presence of an integral time scale L_I suggests an explicit dependence on large scale eddy motion within the inertial subrange. One estimate of L_I may be derived from the integral length scale of the flow given by

$$L_I = U \cdot I_w = U \cdot \int_0^\infty \rho_w(\tau_0) d\tau_0, \tag{10}$$

where $\rho_w(\tau_0)$ is the vertical velocity autocorrelation function and τ_0 is the time lag. Here, I_w is presumed to be the most restrictive scale given that w' is the flow variable most impacted by the presence of the boundary.

The statistics of air temperature increments across scales (τ_0/I_w) for different ζ conditions are explored with a lens on two primary features: buildup of heavy tails and destruction of asymmetry originating from ramp-cliff structures at the cross-over from $\tau_0/I_w > 1$ to $\tau_0/I_w \approx 0.1$. Because changes in ζ do result in changes in I_w , the time (or space) lags are presented in dimensionless form as $\tau = \tau_0/I_w$, so that the increments of a flow variable Δs , with $\Delta s = \Delta u, \Delta w, \Delta T$ at a given dimensionless scale τ , can be expressed as $\Delta s(\tau) =$ $s(t + \tau) - s(t)$, where $t = t_0/I_w$.

¹⁸⁴ B. Probabilistic description of intermittency across scales

The intermittent behavior of ASL turbulent flows has been documented by several experiments [33, 34], and a number of models have been proposed to capture the effects of intermittency on the flow statistics in the inertial range of scales (e.g., lognormal, bi- and multi-fractals - beta model, log-stable, She-Leveque vortex filaments, etc). Common to all these models is the hypothesis of local isotropy and the accounting for uneven distribution of eddy activity in the space/time domain, which explains the anomalous scaling of higher order even structure functions.

Here, a statistical description of scalar increments is used to fingerprint large-scale signatures across scales τ for different ζ . If such fingerprints exist, the dissipation rates ϵ and ϵ_T ¹⁹⁴ need not be sufficient to describe all aspects of the inertial range statistics. The one-time ¹⁹⁵ probability density function (pdf) of the increments $\Delta s(\tau)$ of the flow variable s = u, w, T¹⁹⁶ at a given dimensionless scale τ , can be expressed as [35]

$$p(\Delta s) = \frac{N}{q_o(\Delta s)} \exp \int_0^{\Delta s} \frac{r_o(\Delta s')}{q_o(\Delta s')} d\Delta s'.$$
 (11)

This expression is exact when Δs are realizations of a stationary stochastic process S(t)197 under the condition $p(\Delta s) \to 0$ as $\Delta s \to \infty$. Here $q_o(\Delta s) = \langle \dot{S}^2 | \Delta s \rangle / \langle \dot{S}^2 \rangle$ and $r_o(\Delta s) =$ 198 $\langle \ddot{S} | \Delta s \rangle / \langle \dot{S}^2 \rangle$ are the normalized averages of the first and second order conditional derivatives 199 of the process S(t), and N is a normalization constant. Eq. (11) generalizes previous results 200 obtained by Sinai and Yakhot [36] and Ching [37] for the pdf of temperature fluctuations 201 and their increments, where the term $r_o(\Delta s)$ was linear $(r_o(\Delta s) = -\Delta s)$. Eq. (11), while 202 derived for a twice-differentiable process, can be interpreted as the steady-state solution of a 203 Fokker Planck equation with $p(\Delta s)$ vanishing at infinite boundaries, with drift and diffusion 204 coefficient equal to r_0 and q_0 respectively [38, 39]. 205

Although Eq. (11) can be directly computed from an observed time series, the estimation 206 of the conditional derivatives in $q_o(\Delta s)$ and $r_o(\Delta s)$ becomes inevitably uncertain as Δs 207 approaches the tails of the pdf. However, a number of parametric distributions commonly 208 used in statistical mechanics arise as particular cases of Eq. (11) when $r_o(\Delta s) = -\Delta s$, 209 such as Gaussian (q_o constant), power-laws ($q_o(\Delta s) \sim \Delta s^2$) and stretched exponentials 210 $(q_o(\Delta s) \sim \Delta s^a, 0 < a < 2)$. To facilitate estimation and comparisons with data, two different 211 parametric models for the tails of Eq. (11) are here adopted: a Stretched Exponential 212 (SE) and a q-Gaussian distribution (QG). The first arises from multiplicative processes of 213 normal-distributed random variates [40], while the second maximizes a generalized measure 214 of information entropy proposed by Tsallis [41–43]. While QG does not have a clear physical 215 basis in the context of turbulent flows [44], it has been widely used in the analysis of 216 turbulence simulations and data [13, 45-47]. We employ these two models to infer tail 217 behavior as well as to test the independence of our findings from the particular parametric 218 distribution used to characterize $p(\Delta s)$. The QG and SE pdfs are given as 219

$$p_{QG}(\Delta s) = N(q) \cdot \left(1 + (q-1)\frac{\Delta s^2}{2\psi^2}\right)^{\frac{1}{1-q}},$$
(12)

220

$$p_{SE}(\Delta s) = \frac{\eta}{\lambda} \left(\frac{\Delta s}{\lambda}\right)^{\eta-1} \cdot \exp\left(\frac{\Delta s}{\lambda}\right)^{\eta}.$$
(13)

Both pdf models have two degrees of freedom corresponding to a scale (ψ, λ) and shape (η, q) parameter. We adopt the (symmetric) QG model and the SE fitted separately to right and left tails of $p(\Delta T)$.

224 **C.**

C. Probabilistic description of asymmetry and irreversibility across scales

The presence of ramp-cliff structures has been conjectured to result in non-local interac-225 tions of different size eddies within the inertial subrange [4]. This non-locality affects both 226 even and odd moments of higher order. A statistical framework to investigate the effects of 227 ramps on the asymmetric nature of velocity and scalar increments for different atmospheric 228 stability classes is now discussed. Sharp edges associated with cliffs might directly inject 229 scalar variance at much smaller scales and thus alter the magnitude and sign of odd order 230 moments within the inertial range (depending on z/L). The presence of asymmetry has been 231 investigated based on odd-ordered structure functions [4] or multipoint correlators [48]. In 232 particular, a simple measure for the persistence of asymmetry at small scales is the skewness 233 of the scalar increments $S_T^3 = \langle \Delta T(\tau)^3 \rangle / \langle \Delta T(\tau)^2 \rangle^{3/2}$. The structure skewness of air tem-234 perature has been found to scale as $Re_{\lambda} = \sigma_u \lambda / \nu$ (where λ is the Taylor microscale and σ_u 235 is the root mean square of the longitudinal velocity fluctuations) and thus for a boundary 236 layer $S_3^T \sim Re_*^{1/2}$. However, for large values of Re_{λ} experimental evidence suggests that S_3^T 237 tends to plateau and become independent of Re_{λ} [4, 31]. 238

A further signature of ramp-cliff structures is that increments $\Delta T(\tau)$ may exhibit a time 239 directional (or 'irreversible') behavior. Time reversibility implies that the trajectories of 240 a stationary process Θ_t exhibit the same statistical properties when considered forward 241 or backward in time. In particular, for a reversible time series the n-points joint pdf of 242 $(\Theta_1, \Theta_2, ..., \Theta_n)$ is equal to the joint pdf of the reversed sequence $(\Theta_n, \Theta_{n-1}, ..., \Theta_1)$ for every 243 n. While testing this general definition of reversibility would require perfect knowledge 244 of the phase space trajectories, a weaker definition is the so called lag-reversibility. This 245 condition only requires the two-points pdfs to be equal: $f_{\Theta_t,\Theta_{t+\tau}}(\Theta_1,\Theta_2) = f_{\Theta_{t+\tau},\Theta_t}(\Theta_2,\Theta_1).$ 246 While this definition is less general, it still provides a necessary condition for testing time 247 reversibility. Moreover, it is consistent with the traditional descriptions of turbulence that 248 are primarily based on two-point statistics. Lag reversibility implies that [49] 249

$$R_{\tau} = \rho_c(\Theta_t^2, \Theta_{t+\tau}) - \rho_c(\Theta_t, \Theta_{t+\tau}^2) = 0.$$
(14)

where ρ_c denotes a correlation coefficient between two variables. This condition can be directly tested across different τ and ζ using a conventional correlation analysis.

A second test for reversibility of scalar trajectories is here performed based on the Kullback-Leibner measure, a form of relative entropy that determines the average distance between the entire pdf of forward and backward trajectories [39, 50, 51]. Again, the analysis here is restricted to the inspection of lag-reversibility (n = 2) across scales τ . In such a restricted form, this measure reduces to

$$\langle Z_{\tau} \rangle = \int_{\Omega_{\Theta}} \int_{\Omega_{\Theta_{\tau}'}} p(\Theta_{\tau}'|\Theta) p(\Theta) \log \frac{p(\Theta_{\tau}'|\Theta)}{p(-\Theta_{\tau}'|\Theta)} d\Theta_{\tau}' d\Theta, \tag{15}$$

where $\Theta'_{\tau} = \Delta \Theta(\tau)/\tau$, and the domains of integration Ω_{Θ} and $\Omega_{\Theta'_{\tau}}$ correspond to the populations of the random variables Θ and Θ'_{τ} respectively. Eq. (15) determines, at each dimensionless scale τ , the average distance between the probability of the transition $\Delta \Theta(\tau)$ and its inverse, at every given level Θ .

A statistical mechanics interpretation of Eq. (15) would imply that for a system in non-equilibrium steady state, the *Fluctuation Theorem* must hold so that

$$\log \frac{p(-Z_{\tau})}{p(Z_{\tau})} = -Z_{\tau} \tag{16}$$

²⁶³ for the variable Z_{τ} computed at some level Θ

$$Z_{\tau}(\Theta) = \log \frac{p(\Theta_{\tau}'|\Theta)}{p(-\Theta_{\tau}'|\Theta)}.$$
(17)

Note here the usage of conditional probabilities instead of their unconditional forms employed in recent flight-crash studies of Lagrangian fluid particles [15] that also made use of Fluctuation Theorem and time-reversibility. Eq. (15) has been shown to have general validity [51] independent of the underlying dynamics or statistical-mechanics interpretations, when considering conditional statistics.

269 III. DATA AND METHODS

The three velocity components and air temperature measurements were sampled at 56 Hz using an ultra-sonic anemometer positioned at z = 5.2 m above a grass-covered surface at the Blackwood Division of the Duke Forest, near Durham, North Carolina, USA. The anemometer samples the air velocity in three non-orthogonal directions by transmitting ²⁷⁴ sonic waves in opposite directions and measuring their travel times along a fixed 0.15 m ²⁷⁵ path length. Temperature fluctuations are then computed from measured fluctuations in ²⁷⁶ the speed of sound assuming air is an ideal gas. The non-orthogonal sonic anemometer ²⁷⁷ design used here has proven to be the most effective at reducing flow distortions induced by ²⁷⁸ the presence of the instrument.

The experiment resulted in 123 time series records (henceforth termed 'runs') each having 279 a duration of 19.5 minutes (65536 data points at 56Hz), covering a range of different atmo-280 spheric stability conditions [29]. Of these, only 34 runs passed a stationarity test and were 281 included in the analysis (see Table I for a summary of the properties of the flow for these 282 runs). The assumption of stationarity is necessary so as to (i) decompose the flow variables 283 into a mean and fluctuating part, (ii) adopt Eqs. (11) and (15) so as to describe intermit-284 tency and time irreversibility respectively, and (iii) compute the integral scales needed in 285 delineating the transition from production to inertial. To test the dataset for stationarity, 286 we employ the second order structure functions of velocity components (u, w) and air tem-287 perature T. Runs were included only if the slope of $S_s^2 = \langle [s(t+\tau) - s(t)]^2 \rangle$ for time delays 288 larger than about 9 minutes (30000 sample points) was smaller than a fixed value (0.01). 289 For the 34 runs meeting this strict stationarity criterion, second order structure functions 290 for w and T are featured in Fig. 1. As expected, structure functions exhibit an approximate 291 2/3 scaling at fine scales and transition to a constant value as the autocorrelation weakens 292 at large separation distances. 293

The presence of a stable stratification is known to produce distortions on the spectral properties of turbulence at scales commensurate with (and larger than) the Dougherty-Ozmidov length scale [52]. We investigated this issue (see the Appendix for more details) finding that stable stratification effects are only relevant at scales larger than the integral scale I_w considered here and not in the inertial range.

As earlier noted, the most restrictive (i.e. smallest) integral time scale is I_w associated with the vertical velocity w due to ground effects. We assume that this time scale characterizes the transition from production to inertial ranges for all three flow variables u, w, T. Eq (10) is here evaluated by integrating $\rho_w(\tau)$ up to the first zero crossing so as to avoid the effects of low frequency oscillations. Figure 1 illustrates the integral time scales of wand T as a function of ζ , where the aforementioned integral time scales are normalized by the mean vorticity time scale $dU/dz = \phi_m(\zeta)u_*(\kappa_v z)^{-1}$. It is clear that such normalized I_w is approximately constant across stability regimes and suggests I_w to be proportional to the duration of vortices most efficient at transporting momentum to the ground for all ζ . Conversely, the temperature integral time scale is much longer than I_w for near-neutral conditions and only approaches I_w for strongly unstable conditions.

A known limitation of sonic anemometry is the presence of distortions at high frequencies 310 due to instrument path-averaging. For this reason, the smallest time scale considered in the 311 analysis is $0.05 \cdot I_w$, which corresponds to a minimum travel path of 30 cm (or twice the 312 sonic an emometer path length). Taylor's frozen turbulence hypothesis [53] $(r = -\overline{U}t)$ was 313 employed to convert values of τ to separation distances r within the inertial subrange even 314 though the turbulent intensity σ_u/U is not small as shown in Table I. For this reason, we 315 adopt the dimensionless lag τ for analysis and presentation. The τ can be interpreted as 316 temporal or spatial noting that distortions due to the use of Taylor's hypothesis impact 317 similarly the numerator and denominator. 318

For every run, ζ was computed using Eq. (3) and then employed to classify the ASL stability condition. Most of the runs in the dataset are unstable with a wide range of $|\zeta|$, while only 4 runs are characterized by $\zeta > 0$. To ensure a balanced statistical design, two stability classes are selected with the same number of runs (8) in each class: strongly unstable $(|\zeta| > 0.5)$ and near neutral runs $(|\zeta| < 0.072)$. A summary of the bulk flow properties for these runs are featured in Table (I).

In the analysis, each flow variable s (s = u, w, T) is normalized to zero-mean and unitvariance (labeled as s_n). Then, at scale τ , a time series of $\Delta s(\tau) = s_n(t + \tau) - s_n(t)$ is constructed and again normalized to have unit variance.

For illustration purposes, Fig. 2 shows sequences of fluctuations u', w', T' extracted from runs in unstable and stable atmospheric regimes. In the first case, temperature fluctuations clearly exhibit ramp-cliff structures occurring with time scales larger than I_w . In the stable/near neutral case, large scale scalar structure are still present even though their structure is qualitatively different from the unstable case, and may include inverted ramp structures as in Fig. 2(B) when $\overline{w'T'} < 0$.

To test the effects of these coherent structures on inertial subrange statistics, and in particular to isolate the effect of temperature ramps on intermittency and asymmetry, synthetic time series are used and are constructed as follows. First, a phase-randomization of the original temperature records [54] is performed by preserving the amplitudes of the Fourier

coefficients while destroying coherent patterns encoded in the phase angle. A synthetic saw-338 tooth time series is then superimposed on the time series obtained by phase-randomization. 339 Here a coefficient α measures the relative weight of the ramps with respect to the phase-340 randomized sequence. This combination yields realizations of a renewal process (see Fig. 341 2(C) for a representative example with $\alpha = 0.5$) that is unconnected with Navier-Stokes 342 scalar turbulence, but mimics sweep-ejection dynamics [13]. Synthetic ramps are here gen-343 erated with exponentially distributed durations and with a mean duration set to a multiple 344 of the integral time scale $(2 \cdot I_w \text{ in Figure 2(C)})$. The resulting time series is again normalized 345 to have zero mean and unit variance. 346

TABLE I: Bulk flow properties for the runs in our dataset.

The table reports the atmospheric stability parameter ζ , the Obukhov length $L_{[m]}$, the sensible heat flux $H = \rho C_p \overline{w'T'}$ [Wm^{-2}] (where ρ is the mean air density and C_p is the specific heat capacity of dry air at constant pressure), the mean air temperature $T_{[\circ C]}$ and mean velocity $U_{[m/s]}$, and the integral time scale for $w_{[s]}$, the turbulent intensity σ_u/U , the temperature standard deviation $\sigma_T_{[\circ C]}$, and vertical velocity standard deviation $\sigma_w_{[m/s]}$.

Run	ζ	L	H	T	U	I_w	σ_u/U	u^*	σ_T	σ_w
1	-11.56	-0.4	93.2	33.9	2.1	2.62	0.44	0.08	0.48	0.40
2	-1.31	-4.0	121.6	26.9	1.0	7.58	0.72	0.17	0.54	0.30
3	-0.89	-5.8	73.1	27.8	0.5	6.62	0.91	0.16	0.37	0.30
4	-0.81	-6.4	79.9	32.7	0.7	5.75	1.05	0.17	0.61	0.29
5	-0.80	-6.5	138.1	27.4	0.8	8.18	0.48	0.21	0.57	0.31
6	-0.67	-7.7	149.8	31.4	0.9	11.64	1.04	0.23	0.63	0.38
7	-0.59	-8.8	118.1	34.8	1.5	3.43	0.71	0.22	0.58	0.34
8	-0.52	-10.0	85.4	32.5	2.1	1.74	0.37	0.21	0.44	0.37
9	-0.45	-11.5	78.6	31.7	1.1	7.44	0.61	0.21	0.43	0.30
10	-0.44	-11.7	110.7	31.9	1.2	5.89	0.65	0.24	0.49	0.37
11	-0.44	-11.8	39.4	34.4	1.3	3.19	0.45	0.17	0.32	0.29

Run	ζ	L	H	T	U	I_w	σ_u/U	u^*	σ_T	σ_w
12	-0.40	-13.0	36.6	34.1	1.7	2.30	0.39	0.17	0.37	0.28
13	-0.37	-14.0	65.1	25.2	1.6	2.91	0.39	0.21	0.35	0.27
14	-0.33	-15.6	48.0	28.9	1.4	2.58	0.41	0.20	0.27	0.30
15	-0.33	-15.8	4.8	33.4	1.6	1.59	0.35	0.09	0.09	0.23
16	-0.29	-18.2	115.2	32.1	2.7	2.16	0.37	0.28	0.44	0.47
17	-0.28	-18.5	136.2	29.2	0.9	6.88	1.11	0.30	0.56	0.37
18	-0.27	-19.1	108.6	30.5	1.7	3.56	0.62	0.28	0.54	0.34
19	-0.17	-29.7	70.5	29.5	2.6	2.22	0.29	0.28	0.36	0.42
20	-0.15	-33.8	63.2	32.9	2.2	2.97	0.39	0.28	0.36	0.40
21	-0.14	-37.9	30.9	34.2	1.6	4.17	0.49	0.23	0.34	0.32
22	-0.12	-44.4	118.6	31.0	2.6	3.78	0.42	0.38	0.49	0.42
23	-0.09	-56.5	26.7	33.9	1.9	3.39	0.31	0.25	0.15	0.31
24	-0.08	-61.7	49.7	31.7	2.0	3.50	0.41	0.31	0.27	0.39
25	-0.08	-65.1	17.6	34.0	2.2	3.22	0.29	0.23	0.13	0.31
26	-0.07	-72.5	28.8	31.5	1.8	2.71	0.41	0.28	0.29	0.30
27	-0.04	-126.2	45.1	31.0	4.3	1.21	0.33	0.39	0.35	0.71
28	-0.03	-171.8	3.9	31.3	1.7	3.18	0.39	0.19	0.15	0.30
29	-0.02	-261.4	46.1	31.2	3.8	1.37	0.39	0.50	0.23	0.72
30	-0.02	-304.3	47.1	29.4	5.0	0.84	0.31	0.53	0.21	0.80
31	0.002	2397.4	-0.4	31.2	1.9	1.94	0.44	0.22	0.69	0.32
32	0.01	525.5	-1.3	32.9	0.9	3.00	0.51	0.19	0.18	0.23
33	0.05	93.8	-20.7	29.8	2.6	1.52	0.30	0.27	0.23	0.39
34	0.07	71.4	-14.2	30.4	1.9	2.18	0.37	0.22	0.25	0.28

347 IV. RESULTS

The main questions to be addressed here require determination of (i) the probability of extreme scalar concentration excursions and concomitant intermittency, and (ii) scalar asymmetry and time irreversibility across scales. Here, tools introduced in sections II B and II C are used to investigate how these two features vary from production to inertial scales for temperature traces, and to compare this behavior with the observed velocity components. Comparison of these quantities for runs recorded in different atmospheric stability conditions allows to test whether significant coupling across scales exists, and to what extent velocity and temperature statistics are universal at the smallest scale examined here.

A. Probabilistic description of intermittency across scales

We first investigate the intermittent behavior of both scalar and velocity components 357 by assessing to what extent the scaling of even-order structure functions departs from K41 358 predictions. Inspection of scaling exponents ζ'_n in Eq. (9) for u, w, T confirms that K41 pre-359 dictions significantly overestimate scaling exponents for structure functions of order higher 360 than 2, as shown in figure 3(A). The scaling exponents obtained for the scalar T show 361 reasonable agreement with previous experimental results (Fig. 3(B)), with values systemat-362 ically lower than predicted by the Kraichnan model in the limiting case of time-uncorrelated 363 velocity field [55]. The values of ζ'_n averaged over the set of runs observed during the ex-364 periment are lower for the scalar, especially when compared to the longitudinal velocity 365 components. From this analysis, intermittency effects appear stronger for the scalar than 366 for the longitudinal velocity. 367

The empirical pdfs of velocity and air temperature increments ($\Delta s = \Delta u, \Delta w, \Delta T$) for runs in the near-neutral ($|\zeta| < 0.072$) and strongly unstable ($\zeta < -0.5$) classes (Fig. 4) show clear transitions from a quasi-Gaussian regime at large lags ($\tau = 2$ in figure) to distributions with sharper peaks and longer tails at scales well within the inertial subrange ($\tau = 0.05$). This behavior has been documented for a wide range of turbulent flows [56] and is associated with the build up of intermittency [32] due to self-amplification inertial dynamics [57].

The bulk of the pdf of temperature increments at any given scale can also be characterized by the coefficients of Eq. (11). Results show some differences between runs with differing $|\zeta|$ (Fig. 5). Namely, for runs in the strongly unstable class, q_0 exhibits a more pronounced peak around the origin and is characterized by larger asymmetry at the cross-over scale $\tau = 1$ compared to their near-neutral counterparts (Fig. 5(A)). Moreover, the results here confirm that a choice of linear $r_0(\Delta T)$ and quadratic $q_0(\Delta T)$ appear reasonable for ASL flows. In the case of an unstable ASL, the term $r_0(\Delta T)$ remains linear, while inspection of $q_0(\Delta T)$ ³⁸¹ suggests that a dependence on *s* with an exponent smaller than 2 might be more appropriate, ³⁸² corresponding to stretched exponential tails for $p(\Delta T)$ for small lags τ in unstable ASL flows. ³⁸³ Comparison with the same data after run-by-run spectral phase randomization [54] shows ³⁸⁴ that the latter exhibits almost Gaussian behavior, confirming that the emergence of long ³⁸⁵ tails at inertial scales is primarily a consequence of non linear structures in the original time ³⁸⁶ series.

The variation of the tail parameters η and q with decreasing scale τ (Fig. 6) provides a robust measure of how the distributional tails of $p(\Delta T)$ evolve at the onset of the inertial range. For temperature differences, the rates of change across scales of both η and q appear to be dependent on the magnitude of the stability parameter ζ . Consequently, while at large scales - where the pdf closely resembles a Gaussian - neither η nor q exhibit a significant dependence on ζ , for scales well within the inertial subrange stability is clearly impacting the tail behavior of ΔT (Fig. 7).

This evidence suggests that the observed intermittency is not only internal (i.e., not only 394 due to variability in the instantaneous dissipation rate [9]) but is also directly impacted by 395 the larger scale eddy motion that sense boundary conditions. In particular, when buoyancy 396 generation is significant, the heat flux $\overline{w'T'}$ is connected with the sweep and sudden ejection 397 of air parcels, corresponding with the sharp edges of the temperature ramps [3, 13]. The 398 resulting sawtooth behavior could be responsible for the injection of scalar variance at small 399 scales (instead of a gradual cascade), acting in particular on the negative tail of the ΔT pdf, 400 as evident from Fig. 5(A). On the other hand, the buildup of non-Gaussian statistics for 401 velocity increments is not as impacted by the stability regime, and therefore the dominant 402 effects are in this case primarily an effect of internal intermittency. 403

404 B. Probabilistic description of asymmetry across scales

To compare the data sets used here with laboratory studies, we first test the validity of Obukhov's constant skewness hypothesis, which would require the third order structure function of the longitudinal velocity component being constant within the inertial range. Figure 8 reports the values of the third order structure functions (Eqs. (7) and (8)) evaluated at the onset of the inertial subrange as delineated by the w time series. Both are approximately constant for scales smaller than I_w . While comparison with experiments shows good agreement for $S(\tau) \simeq -0.25$, $F(\tau)$ is systematically smaller than its anticipated value [29] (-0.4) for all ζ .

For the scalar T, The presence of a finite third order temperature structure function signifies that local isotropy is not fully attained in the range of scales explored here. The temperature skewness S_T^3 exhibits a plateau for scales smaller than I_w (Fig. 9(A)) similar to previous measurements reported in grid turbulence forced by a mean temperature gradient [58]. Moreover, S_T^3 levels off to positive values for $\zeta > 0$, while it becomes negative for $\zeta < 0$. This finding is consistent with the presence of ramp-like structures when $\zeta > 0$ (mildly stable conditions) that are inverted when compared to their unstable counterparts.

The findings here confirm that at the cross-over from production to inertial, imprints of 420 ramp structures persists well into the inertial subrange. The consequence of these imprints on 421 time-reversibility is now considered for temperature sequences. The irreversibility analysis 422 detects strong irreversibility at large scales that slowly decreases at the onset of the inertial 423 range (Fig. 9). This finding is consistent with the idea that atmospheric stability determines 424 a preferential direction for the large-scale scalar structures, which becomes progressively 425 weaker at scales smaller than $\tau = 1$. Here, the sign of the heat flux has a primary effect 426 on the orientation of the ramps, as captured by R_{τ} . Furthermore, phase randomization 427 is shown to destroy much of this time irreversibility (Fig. 9(B)) while the addition of 428 synthetic ramps, either with positive or negative orientation, produces values of R_{τ} that 429 closely resemble observations of stable and unstable ASL respectively. These synthetic 430 experiments also recover the sign of the third order moment S_T^3 (Fig. 9(A)) but not its 431 magnitude at smaller scales. As one would expect, a sawtooth time series does not fully 432 reproduce inertial scale scalar dynamics, even though it does clearly capture the qualitative 433 effect of boundary conditions on scalar ramp-cliffs. 434

Additional insight can be obtained by the relative entropy measure defined in Eq. (15), 435 which was here evaluated by integrating the relative entropy over the joint frequency distri-436 bution of normalized temperature fluctuations and their increments at each scale τ . We used 437 a coarse binning for estimating the joint pdf $p(T'(\tau), T)$ and assumed [51] that only finite 438 probability ratios contribute to $\langle Z_{\tau} \rangle$. To check the consistency of this approach, calculations 439 of Eq. (15) were repeated using a phase space reconstruction technique based on embedding 440 sequences $(T_t, T_{t+\tau})$ with delay time τ and embedding dimension 2, which confirmed the 441 validity of this approach (results not shown). 442

The averaged relative entropy $\langle Z_{\tau} \rangle$, while insensitive to the ramp orientation, at every given level T quantifies the imbalance between forward and backward probability fluxes of temperature trajectories (Fig. 10(A)). Again, irreversibility of scalar records increases with the lag τ and here tend to plate at larger scales ($\tau > 1$).

⁴⁴⁷ Phase-randomized time series, by comparison, exhibit smaller values of $\langle Z_{\tau} \rangle$ in the inertial ⁴⁴⁸ range. As one would expect, the excess is thus likely a direct result of the presence of scalar ⁴⁴⁹ ramps. The presence of asymmetric patterns in temperature time traces further suggests that ⁴⁵⁰ in the inertial range scalar turbulence is more time-irreversible than velocity, as confirmed ⁴⁵¹ by the larger values of $\langle Z_{\tau} \rangle$ at inertial scales (Fig. 10(B)).

Time-irreversibility of phase space trajectories was further investigated by testing if a 452 significant difference exists between the probability distribution $p(T'_{\tau}|T)$ and $p(-T'_{\tau}|T)$. To 453 this end, a Kolmogorov-Smirnov (KS) test was performed at the significance level 0.05. At 454 every scale τ , results were averaged over different values of T and across runs within the same 455 stability class. The results from the KS test confirm the picture obtained from the relative 456 entropy measure $\langle Z_{\tau} \rangle$: The pdf of forward and backward temperature diverge significantly 457 as the scale τ increases as shown in figure 10, panels (C) and (D). While this test does not 458 capture the sign of the ramps, the behavior of near neutral runs exhibit some difference 459 from the case of relevant heat flux: near neutral runs appear on average more reversible 460 than unstable runs at the same dimensionless scale τ . 461

462 V. DISCUSSION AND CONCLUSIONS

In this work, statistical measures for the frequency of extreme fluctuations and the timedirectional behavior of observed time series were applied to scalar turbulence in the ASL. It was demonstrated that i) the extreme value properties of the scalar markedly depend on the external forcing, and ii) scalar dynamics is characterized by time-irreversible behavior at the scales of injection of scalar variance in the turbulent flow. This time-irreversibility propagates down to the smaller scales of the flow examined here, thus carrying fingerprint of the energy injection mechanism.

It is well known that the pdfs of scalar increments develop heavier tails with decreasing scales in the inertial range when compared to their velocity counterparts. The analysis here shows that within the first two decades of the inertial subrange, this buildup of tails also

carries the signature of turbulent kinetic energy generation. The direct injection of scalar 473 variance from large scales seem to hinder any universal description of ΔT statistics within 474 this range of scales. Instead, the pdf of $\Delta T(r)$ for ASL flows appear to be conditional on 475 the value of ζ at scale r. This finding reinforces previous experimental results [59] obtained 476 for a different type of flow (turbulent wake). In this case, the scalar injection mechanism 477 was shown to impact higher order scaling exponents of the temperature structure functions. 478 This dependence on atmospheric stability regime for $p(\Delta T)$ further suggests that the 479 topology of large eddies, and in particular the presence of ramp-cliff scalar structures, may be 480 responsible for the scale-wise evolution of intermittency and the persistent time directionality 481 at fine scales. This intermittency excess observed in the transition from production to 482 inertial scales is consistent with self-amplification dynamics taking place that further excite 483 the excess of scalar variance injected by the ramps. 484

However, while measures of intermittency appear to be dependent on the absolute value 485 of ζ , i.e., on the relative magnitude of shear and buoyancy production terms (regardless on 486 the sign of the heat flux), the analysis of asymmetry and time reversibility clearly sense 487 the sign of the heat flux H more than the magnitude of ζ itself. This effect is arguably 488 a product of the preferential orientation that the external temperature gradient imposes 489 on the scalar ramp-cliffs, as explained by sweep-ejection dynamics. This hypothesis was 490 here further tested by comparisons with synthetic time series that mimic ramp-cliff patterns 491 observed in the scalar time series. The analysis confirmed that much of the observed time 492 irreversibility, as well as its dependence on the sign of H, are recovered by these surrogate 493 time series (Fig. 9). 494

The analysis of time directional properties showed that time-irreversible behavior for the 495 scalar is stronger at the large scales of the flow where boundary conditions, and in particular 496 the sign of H, determine the orientation and structure of the eddies. At finer scales, time 497 irreversibility as quantified by both $\langle Z_{\tau} \rangle$ and R_{τ} progressively decreases as advection destroys 498 the preferential eddy orientation imposed by boundary conditions. Note that this behavior 499 is not captured by a simple measure of skewness such as S_T^3 (Fig. 9(A)), which is small at 500 large scales and plateaus in the inertial range consistent with previous experiments [4] and 501 numerical simulations [60], thus showing that local isotropy is not fully attained at the finer 502 scales examined here. 503

⁵⁰⁴ Turbulent flows exist in a state far from thermodynamic equilibrium, with the flow statis-

tics exhibiting irreversibility. This irreversibility is typically described in terms of fluxes of 505 energy or asymmetries in the pdfs of the fluid velocity increments [61]. Similar methods 506 could be used to describe irreversibility in the scalar field, e.g. using S_T^3 , and this would 507 imply that the irreversibility of the scalar field is stronger at smaller scales than it is at 508 larger scales. However, in this paper we have used alternative measures to quantify the 509 irreversibility, namely $\langle Z_{\tau} \rangle$ and R_{τ} . These quantities paint a different picture, namely that 510 it is the largest scales, not the smallest (inertial) scales in the scalar field that exhibit 511 the strongest irreversibility. A potential cause for these differing behaviors is that whereas 512 fluxes and quantities such as S_T^3 are multi-point, single-time quantities, $\langle Z_\tau \rangle$ and R_τ are 513 single-point, multi-time quantities. Thus, these two ways of describing irreversibility pro-514 vide different perspectives about the nature of irreversibility in turbulence, which involves 515 fields that evolve in both space and time. This difference in perspectives is a topic for future 516 inquiry. 517

Collectively, the results presented in this paper suggest the following picture for ASL 518 turbulence at the cross-over from production to inertial. Increasing instability in the ASL 519 leads to increases in the mean turbulent kinetic energy dissipation rate (as evidenced by Eq. 520 (1)) and its spatial autocorrelation function and pdf. The consequences of this increased 521 dissipation with increased instability has different outcomes for velocity and scalar turbu-522 lence. For velocity, refinements to K41 appear sufficient to explain the observed scaling in 523 the inertial subrange. For scalar turbulence, the picture appears more complicated. Inter-524 mittency buildup with decreasing (inertial) scales is more rapid when compared to their 525 velocity counterparts, and the signature of the temperature variance injection mechanism 526 persists at even the finer scales explored here. 527

Turbulence and scalar turbulence are characterized by a constant flux of energy and 528 scalar variance from the scales of production down to dissipation. While early theories 529 hypothesized a cascade only depending on these quantities, experimental evidence to date 530 supports a more complicated picture. The multi-time information encoded in $\langle Z_{\tau} \rangle$ reveal 531 that time-reversibility is not constant across scales, as do the fluxes of information entropy. 532 Probability fluxes forward and backward in time are not balanced in general for air tem-533 perature increments, especially at the cross-over from production to inertial. Furthermore, 534 these fluxes carry the signature of the external boundary conditions (i.e. H) and show that 535 dissipation rates themselves are not independent of the large-scale dynamics. Although a 536

formal analogy between Eq. (15) and the thermodynamics of microscopic non-equilibrium steady state systems exists, we stress that in the present application turbulent fluctuations are macroscopic and are the result of non-linear and non-local interactions.

540 Appendix: Stable stratification and distortions of the inertial subrange

In general, stable stratification limits the onset and extent of the inertial subrange given its damping effect in the vertical direction [52]. Here, we show that the scales for which these effects are relevant occur at scales larger than the inertial range examined here. The Ozmidov length scale [62] (originally suggested by Dougherty [63] in 1961), is defined as the scale above which buoyancy forces significantly distort the spectrum of turbulence.

This length scale, sometimes labeled as the Dougherty-Ozmidov scale, can be expressed as

$$L_0 = \sqrt{\frac{\epsilon}{N^3}},\tag{A.1}$$

where ϵ is, as before, the mean turbulent kinetic energy dissipation rate and N is the Brunt Väisälä frequency, defined as

$$N = \sqrt{\frac{g}{T} \frac{dT}{dz}}.$$
 (A.2)

In the study used here, no information was provided about the actual mean potential temperature gradient dT/dz. However, an approximated estimate of L_0 for the runs collected in case of stable atmospheric stratification may be conducted. Note that only 4 runs follow this stability class as runs not meeting strict stationarity requirements were excluded from the analysis (and they were mainly collected in unstable atmospheric conditions). The mean dT/dz was computed using Monin- Obukhov similarity theory as

$$\frac{dT}{dz} = -\left(\frac{T^*}{K_v z}\right)\phi_T\left(\frac{z}{L}\right) \tag{A.3}$$

where $k_v = 0.41$ is the von Karman constant, z = 5.1 m is the distance from the ground, $T^* = \frac{\langle w'T' \rangle}{u^*}$, and for mildly stable stratification

$$\phi_T = \phi_m = 1 + 4.7 \left(\frac{z}{L}\right). \tag{A.4}$$

⁵⁵⁸ The mean turbulent kinetic energy dissipation rate was computed as

$$\epsilon = \frac{u^{*3}}{k_v z} \left(\phi_m - \frac{z}{L} \right) \tag{A.5}$$

Figure 11(A) shows that the quantity

$$I_s = \frac{I_w u^* \phi_m}{k_v z} = constant \simeq 0.4 \tag{A.6}$$

is almost constant across runs and exhibits a value slightly lower than the expected 0.4.

The estimated values of the dimensionless Ozmidov number $L_0/(I_w u^* \phi_m)$ are reported in Figure 11(B). L_0 decreases with increasing stability ζ as the effect of buoyancy is felt by eddies of sizes progressively smaller. However, the values of the Ozmidov scale are consistently larger than the integral scale of the flow I_w for the 4 stable runs here. Hence, ignoring distortions caused by stable stratification on inertial subrange scales for the aforementioned 4 runs may be deemed plausible.

567 ACKNOWLEDGMENTS

E.Z. acknowledges support from the Division of Earth and Ocean Sciences, the Nicholas School of the Environment at Duke University, and from the National Aeronautics and Space Administration (NASA Earth and Space Science Fellowhip 17-EARTH17F-270). G.K. acknowledges support from the National Science Foundation (NSF-EAR-1344703, NSF-AGS-1644382, and NSF-DGE-1068871) and from the Department of Energy (DE-SC0011461). We wish to thank Marco Marani, Brad Murray and Amilcare Porporato for useful discussions, and two anonymous reviewers whose comments improved the quality of the manuscript.

575 **DISCLAIMER**

576 The authors declare no conflict of interest.

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FIG. 1. In the upper panels, the normalized second order structure functions for vertical velocity (A) and temperature (B) are shown for runs that are weakly unstable (blue dashed lines), strongly unstable (red lines), and stable (black dash-dot lines). Black lines indicate the value 1 and the 2/3 power law for reference; vertical dashed lines correspond to the dimensionless scales $\tau = 0.05$ (smallest scale not impacted by instrument path length), $\tau = 1$ (integral scale of the flow), and $\tau = 5$ (typical scale larger than I_w , while small enough not to be impacted by statistical convergence issues in structure functions calculations). Lower panels illustrate (C) the integral scales of the flow for s = T (circles) and s = w (crosses) as a function of the stability parameter $|\zeta|$, and (D) their ratio I_T to I_w again as a function of $|\zeta|$, where stable runs ($\zeta > 0$) are indicated by black squares.



FIG. 2. Sequences of velocity and temperature fluctuations extracted from a strongly unstable run (run 8, $\zeta = -0.52$, $I_w = 1.74s$, column A) and a stable/near neutral one (run 34, $\zeta = 0.07$, $I_w = 2.18s$ column B). The presence of ramps and inverted-ramp like structures respectively is marked by dashed vertical lines. Column (C) illustrates a phase-randomized sequence obtained from run 34 (top), a series of synthetic ramps with durations exponentially distributed with mean $2I_w$ (middle) and the surrogate time series obtained merging the above sawtooth pattern with the phase randomized time series (bottom), where the relative weight of the ramps α was set equal to 0.5.



FIG. 3. (A) Average values of the scaling exponents for longitudinal velocity u (triangles), vertical velocity w (squares), and temperature T (circles). Black continuous line and dashed line show respectively the K41 and the She-leveque predictions for the longitudinal velocity structure functions. Exponents are computed from scales ranging between $\tau = 0.05$ and $\tau = 0.2$. (B) Scaling exponents for temperature only; Mean and standard deviation values are computed over all the runs and are indicated by circles and vertical bars, respectively. Data from Mydlarsky and Warhaft (1990) [58] (squares), Antonia et al. (1984) [64] (triangles), Meneveau et al. (1990) [65] (*) and Ruiz et al. (1996) (diamonds) [66] are shown for comparison. The KOC scaling (black line) and results from the Kraichnan model (1994) [55] (dashed line) as reported in [4] are also presented for reference.



FIG. 4. Normalized probability density functions observed for increments of longitudinal velocity (A), vertical velocity (B) and air temperature (C) at large scales ($\tau = 2$, top panels) and small scales ($\tau = 0.05$, lower panels). The figure includes data from runs in the strongly unstable class ($\zeta < -0.5$, shown in red), and near neutral class ($|\zeta| < 0.072$, blue). Black lines show the standard Gaussian distribution for reference.



FIG. 5. Functions $q_0(\Delta T)$ and $r_0(\Delta T)$ estimated from the conditional derivatives of the original temperature time series, for the two classes of strongly unstable (red lines) and near neutral runs (blue dashed lines). The same quantities are reported for phase-randomized surrogate time series for comparison (grey circles). Results are shown for the central body of the pdf (within 3σ from the mean) for illustration purposes. Top panels (A,B) are computed for a lag equal to the integral time scale of the flow $\tau = 1$, while the bottom panels (C,D) correspond to the smaller time lag $\tau = 0.1$. Black lines $q_0 = 1$ and $r_0 = -\Delta T$ correspond to the standard Gaussian distribution.



FIG. 6. Evolution across scales τ of the q-Gaussian tail parameter q (A), and of the stretched exponential shape parameter η obtained from separate fit to the left (B) and right (C) tails of the distribution of temperature increments. Data from two stability classes are included: strongly unstable ($\zeta < -0.5$, red cirles) and near neutral conditions ($|\zeta| < 0.072$, blue triangles). Black lines and shaded areas indicate average values and standard deviations respectively computed over the entire dataset.



FIG. 7. Tail parameters of the pdf of temperature increments across stability conditions ζ . Results include the q-Gaussian tail parameter q (column A) and the stretched exponential shape parameter η , obtained from fitting the left (column B) and right tail (column C) of the distribution $p(\Delta T)$. Values of q and η are reported for large scales ($\tau = 5$, upper panels) and small scales ($\tau = 0.05$, lower panels). Triangles denote strongly unstable runs ($\zeta < -0.5$), squares denote stable runs ($\zeta > 0$) and circles refer to slightly unstable runs ($-0.5 < \zeta < 0$).



FIG. 8. Normalized third order structure functions $S(\tau)$ and $F(\tau)$ at the crossover from inertial to production scales. Vertical dashed line indicates the integral time scales, horizontal lines show the constant values 0.25 (A) and 0.4 (B). Results are shown for near neutral runs ($|\zeta| < 0.072$, blue dashed lines), strongly unstable runs ($|\zeta| > 0.5$, red lines), and runs with intermediate values of $|\zeta|$ (black dash-dot lines).



FIG. 9. Measures of asymmetry S_T^3 (A) and time irreversibility R_{τ} (B) computed for temperature increments for scales varying from $\tau = 0.05$ to $\tau = 5$. The plots include stable runs (black dashed lines), weakly unstable runs (blue dash-dot lines) and strongly unstable runs (red lines). For reference, the same quantities are computed for phase-randomized time series (cyan), and sythetic time series with sawtooth positive (blue) and inverted ramps (black). Shaded regions correspond to the 1 σ -confidence intervals over 34 realizations of the surrogate time series. Relative weight and mean duration of the synthetic ramps were set to $\alpha = 0.4$ and $2I_w$ respectively.



FIG. 10. (A) Mean and standard deviation over 34 time series of $\langle Z_{\tau} \rangle$ computed for scales varying from $\tau = 0.05$ to $\tau = 20$. Values of $\langle Z_{\tau} \rangle$ are shown for original temperature records (red), and surrogate time series obtained by phase-randomization (green). For comparison, the same analysis is reported for fractional brownian motion with Hurst exponent H = 1/3 (blue). (B) A comparison of $\langle Z_{\tau} \rangle$ for temperature (red), longitudinal velocity (yellow) and vertical velocity (green). The lower panel shows the Kolmogorov-Smirnov test average rejection rate (C) and average P-value (D) computed for all the temperature time series (cyan for mean value and 1σ confidence interval), and for different stability classes: strongly unstable runs ($\zeta < -0.5$, red), near-neutral runs ($|\zeta| < 0.072$, blue) and intermediate values ($0.072 < |\zeta| < 0.5$, black). KS test was performed at the 0.05 significance level, corresponding to the horizontal line in (D). The vertical dashed line marks the integral time scale I_w .



FIG. 11. (A) Quantity *Is* and its expected value 0.4 (black horizontal line) for the 4 stable runs in the dataset. (B) Normalized Ozmidov length for the same runs.