1. Major Concerns
   a) In the introduction part, comparison between different existing techniques is not enough, need some improvement to highly the significance of your current study.

2. Minor Concerns
   1) line 35, "since they do not require direct access to the source". Argument is not strong since other popular techniques also has this feature.
   2) line 62, "turbulent channel flow". Is it more suitable to state here that we focus on atmospheric lower boundary layer that can be represented by channel flow. Otherwise, people may be confused by the ABL and channel flow.
   3) line 65, "The LES represents perfect field conditions, without interference of confounding factors that may degrade the performance of source estimation methods under actual field conditions." The argument is weak, one of the reason you prefer virtual experiment via LES is it can serve as a benchmark that can be controlled ideally.
   4) line 71, "his". A typo, should be "this"?
   5) equation 1. Please indicates explicitly what is x, y, and z direction since people from different area may prefer different definition of y and z. Also, the formula has no explicit dependence on x. Please change the variance either as a function of x, or in other explicit way.
   6) line 104. More precise, "averaged plume centerline" instead of just "plume centerline".
   7) line 189, "Onto the sampling angle, the instantaneous wind direction measured at each sampled point was added." What does this exactly mean?
   8) line 191, "delta y". Better use another symbol to avoid confusion with grid resolution.
   9) line 199, "The concentration measurements were taken perpendicular to the mean wind over the whole width of the domain". The data is reliable when they are close to boundary since you impose periodic boundary condition laterally. Artificial effects may be introduced for plume close to the boundary.
   10) line 205, "The same time delay was applied between each consecutive transect in an individual set". Not clear for how these procedures preformed, better with some illustrations.
   11) Figure 1 (c). dy symbol should be changed, refer to comments 8).
   12) line 220, "To separate these two processes". Not clear and how is it related to the definition you introduced following.
   13) line 245, "Despite the discrepancy in the mean wind, very good agreement is found between the wind speed variations (Fig. 2 b) and covariances (Fig. 2 c). Very good agreement is found for the triplet correlations as well (Fig. 2 d)). Any interpretation why mean value deviation is large, while the one for higher order statistics is small. This is a little bit contrary to typical cases we encountered in everyone's study, where the mean value deviation is small, but higher order statistics do not agree very well.
   14) line 250, "Following the good agreement of the higher order velocity statistics, we expect that the mixing of the plume in the crosswind directions is well represented in the LES". This is indirect derivation, any direct quantity you can get to reach the same conclusion.
   15) line 255, "roughness velocity", Friction velocity?
   16) line 255, "delta". You define the turn over time based on boundary layer height, which is the effect of viscosity diffusion. Then why not the advection time scale?
   17) Figure 3. The agreement between your result and experiment one is because the position you select is close to the source, therefore the diffusion is dominant. What if you shift your virtual measurement point further downstream, where the advection effect is significant after the boundary layer of the scalar is fully developed. Also the scalar boundary layer height is different from fluid boundary layer height. Beside, please define m3 and m4 at somewhere.
   18) line 270, "To give the 270 reader an intuitive understanding of the spatial characteristics of the turbulence". Why turbulence, should that be the plume structure? Also, should that be the spatial distribution of the plume.
   19) Figure 4. Last subfigure has discontinuity for the plume concentration. Better to make it continuous by adjusting the color level range to avoid any confusion.
20) Figure 5. Any good reason that you put 100 instantaneous results here? People typically use mean + variance to get the same idea you want to show.

21) line 284, "an average". Please specify here whether it is spatial average or temporal average?

22) line 284, "over a large number of time steps". A large time window?

23) Figure 6. It is the isoline, right? Please specify it. The domain height is not the same for each case, better be the same for comparison.

24) line 296, "the width of the Gaussian plume and the highest LES plume only start to diverge far from the source". Why far field? Any reason?

25) line 302, "but are much smaller than the Briggs Gaussian plume coefficients". Why?

26) line 319, "Instead of emitting the tracer as a point source, we use a Gaussian distribution to pre-disperse the source". Suggest to conduct a comparative study to see how sensitive of the source strength estimation if you change the Gaussian kernel size, also, how about use HyperGaussian.

27) Figure 9. Please specify the line symbol in words in figure caption. Same for other figures if it is applicable.

28) Line 360. Reminder the reader where you get the deviation of mixing.

29) line 375, "estimation error up to". Please specify where is the corresponding figure.

30) line 380. Perhaps one reason is that your domain width is not large enough and the plume reach the boundary.

31) line 470, "different stability condition". One of the helpful references is the "Xiao, S., Peng, C., & Yang, D. (2021). Large-eddy simulation of bubble plume in stratified crossflow. Physical Review Fluids, 6(4), 044613," Where the plume structure is no longer to be a Gaussian shape vertically when the plume is developed underwater (corresponding to stable boundary layer in ABL).