Review of "The Impact of Multi-Scale Turbulence Structures within the Urban Canopy of a Basin City on Haze Pollution Processes" by Liu et al.

This paper presents an analysis of the relationship between turbulence statistics and PM2.5 concentrations. The authors identify a typical pattern of haze pollution and attribute this pattern to the evolution of turbulent eddies. However, the paper lacks theoretical rigor and contains basic errors. The primary issue of the paper is that the partitioning between sub-mesoscale motions and turbulence motions is artificial and the explanation of haze pollution pattern using this partitioning is subjective. The authors claimed the emergence of sub-mesoscales even during daytime hours, but no solid evidence is given to substantiate this claim. Accordingly, the reviewer cannot recommend publication in ACP. Below some comments are listed that may help improve the manuscript.

Major comments:

1. The paper is motivated by the unique features of urban boundary layer in basins. So, how the observed haze pollution pattern differs from non-urban areas?
2. The authors refer to intermittency as the variation of turbulence intensity, which differs from the traditional definition of intermittency in fluid mechanism. To this reviewer, this is too phenomenological to start with, as quiescent turbulence can be simply related to the decrease in the energy production, instead of sub-mesoscale motions.
3. The partition of sub-mesoscale motions and turbulence motions is artificial and lack theoretical basis. This might be true in some specific situations, e.g., the gradient Richardson number exceeds 0.25, as waves can survive and turbulence is not influenced by the wall. However, in neutral and unstable boundary layers, waves are dynamically unstable and will definitely break into turbulence. Given that large scale turbulence is anisotropic in neutral and unstable boundary layers, the scales beyond the plateau in energy spectrum could be still turbulence, see relevant works by Marusic.
4. As the observations are collected within the canopy layer, the observed fluid motions must be local, the authors have to prove that there are sub-mesoscale motions in UCL and they are important.
5. The observed haze pollution pattern can be simply explained as: (1) The decrease of PM2.5 from 00:00 to 08:00 is due to the decrease in the emission according to Fig. 2b; (2) The increase of PM2.5 from 08:00 to 14:00 is due to the increase in the emission according to Fig. 2b; (3) The decrease of PM2.5 from 14:00 to 18:00 is due to the strengthening turbulence by unstable stratification according to Fig. 2f, 2i; (4) The increase of PM2.5 from 18:00 to 00:00 is due to the suppressed turbulence by stable stratification according to Fig. 2f, 2i.
6. Fig. 4a should move to method section.
7. The review does not see the necessity of the discussion on the time scale of turbulence in Sect. 3.2. For one thing, 15-min scales on 20 Jan also contributes significantly to TKE, but why PM2.5 is less changed?
8. "Acevedo et al. (2014) found that TKE increased exponentially with the time scale in the sub-mesoscale motions range. Fig. 3 showed that when the eddy scale exceeded a certain threshold (e.g., 15 min), the TKE spectral value increased with the timescale. That suggested the presence of sub-mesoscale motions in this situation." Why does this follow logically? At

the first place, the increase of TKE spectral value with timescale does not necessarily indicate the presence of sub-mesoscales. Secondly, the reviewer does not see the increase of TKE spectral value in Fig. 3.

9. Consequently, the discussion on \delta KE in Page 13 is of less interests. The authors should plot spontaneous velocities to see whether sub-mesoscale motions emerge.
10. Page 14 Lines 2-5: do the authors mean that sub-mesoscales are more efficient in mixing than turbulence by this sentence?
11. Page 14 Lines 5-11: If valid, why LIST increases on 21 Jan, but PM2.5 decreases?
12. Page 15 Line 13: the figure can be shown in the supplementary.
13. Page 16 Line 22: gust is due to the boundary-layer-scale turbulent eddies, not sub-mesoscale motions.
14. The results are not well organized.
15. Page 18 Lines 15-19: The plateau in the pre-multiplied u spectra may be due to attached eddies.
16. Fig. 6a2-d2: Pre-multiplied uw spectrum obeys a -4/3 scaling at small scales, which is not evident in Fig. 6a2-d2. Can the authors give a reason for this?

Minor comments:

1. Page 3 Line 4: UCL is urban canopy layer.
2. Page 12 Line 5: ‘eddy eddies’ should be revised.