Response to Referee 2

We thank the referee for the interesting and helpful comments. Point-by-point responses are included below. For convenience, we first summarise our responses to the three major points and situate them with a broader scientific context.

1. *The time scale of mean circulation, i.e. residence time in the street canyon, of 380 s is very long in comparison to other published studies on street canyons.*

   Pollutant dispersion and aerosol dynamics are strongly influenced by the key dynamical timescales. As is generally the case in fluid dynamics, different timescales may be defined. Our choice of the mean canyon circulation timescale, T_c, is motivated by the finding that pollutant dispersion from relatively deep urban canopies (in the skimming flow regime) occurs on this timescale (Lo & Ngan 2017). Since T_c approximates the e-folding timescale of the mean concentration and the mean age of air within a unit-aspect-ratio street canyon (Lo & Ngan 2016), it is a reasonable choice for this study of aerosol dynamics. One of our key findings is that coagulation within a representative urban canopy depends on the age of air or mean tracer age.

   The timescales quoted by the referee correspond to a different definition. The dilution timescale (Ketzel and Berkowicz 2004) characterizes the rate at which the volume of a plume changes (or equivalently the turbulent diffusion of a pollutant). Applying the version for an urban canopy (Nikolova et al. 2014), yields a much shorter timescale that agrees with previous studies.

2. *The formation of a stable vortex holds for neutral conditions, but it needs to be tested what consequences unstable conditions with thermal convection have on the concentration distribution in the street canyon.*

   It is certainly true that the occurrence of a central canyon vortex, which strongly influences the mean circulation and dynamical timescales, could be affected by unstable stratification. However, a canyon vortex does persist for a bottom-heated street canyon. This was first demonstrated in the two-dimensional Reynolds-Averaged Navier-Stokes calculations of Kim and Baik (2001). In a recent study, we have confirmed this using three-dimensional large-eddy simulation (Figure 1). The vortex persists over a wide range of bulk Richardson numbers, $-0.4 \leq Rb \leq 0.4$. From field measurements taken inside a real urban street canyon (Nakamura and Oke 1988), this range covers stable to moderately unstable conditions. Therefore, the assumption of neutral flow should generalize to representative urban conditions.

3. *Another aspect to consider is that when the wind is parallel with the street, the recirculating structure within the cavity disappears completely, and the concentration field becomes very different.*

   Yes, the flow structure and aerosol concentrations are very different for an external wind parallel to the canyon axis. However, this does not affect our main finding, which is that the aerosol dynamics within the urban canopy depend on the ratio of the aerosol processes to the relevant dynamical timescales. As explained more fully in our response to Referee 1, the results described in the ms correspond to a regime in which the coagulation timescale is long relative to the dynamical timescale while the deposition timescale is of the same order of magnitude; therefore, both processes bear the imprint of the mean circulation, as may be seen in the relative difference fields. This claim has been verified by considering a parallel external wind. Although the structure of the passive scalar field changes from θ=0° to θ=90° (Figs. 2a,c), it does not follow that the qualitative effect of the aerosol dynamic processes also changes. At θ=90°, the number concentration field continues to reflect the structure of the mean circulation (Figs. 2b,d). This can be attributed to the ratios, $\tau_{\mathrm{coag}}/T_{\mathrm{c}}$ and $\tau_{\mathrm{depo}}/T_{\mathrm{c}}$, remaining qualitatively unchanged for θ=90°. While the absence of strong vertical motions increases the mean circulation timescale, the coagulation timescale continues to be much longer than the dynamical timescale and the deposition timescale continues to be slightly shorter (Table 1).

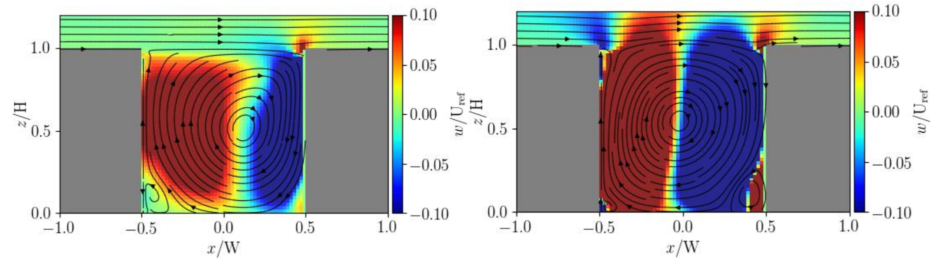

Figure 1: Spanwise-averaged vertical streamlines for a bottom-heated unit-aspect-ratio street canyon and an external wind perpendicular to the canyon axis (Wang et al. 2021). (a) Neutral flow (Rb=0); (b) Unstable flow (Rb=-0.39).

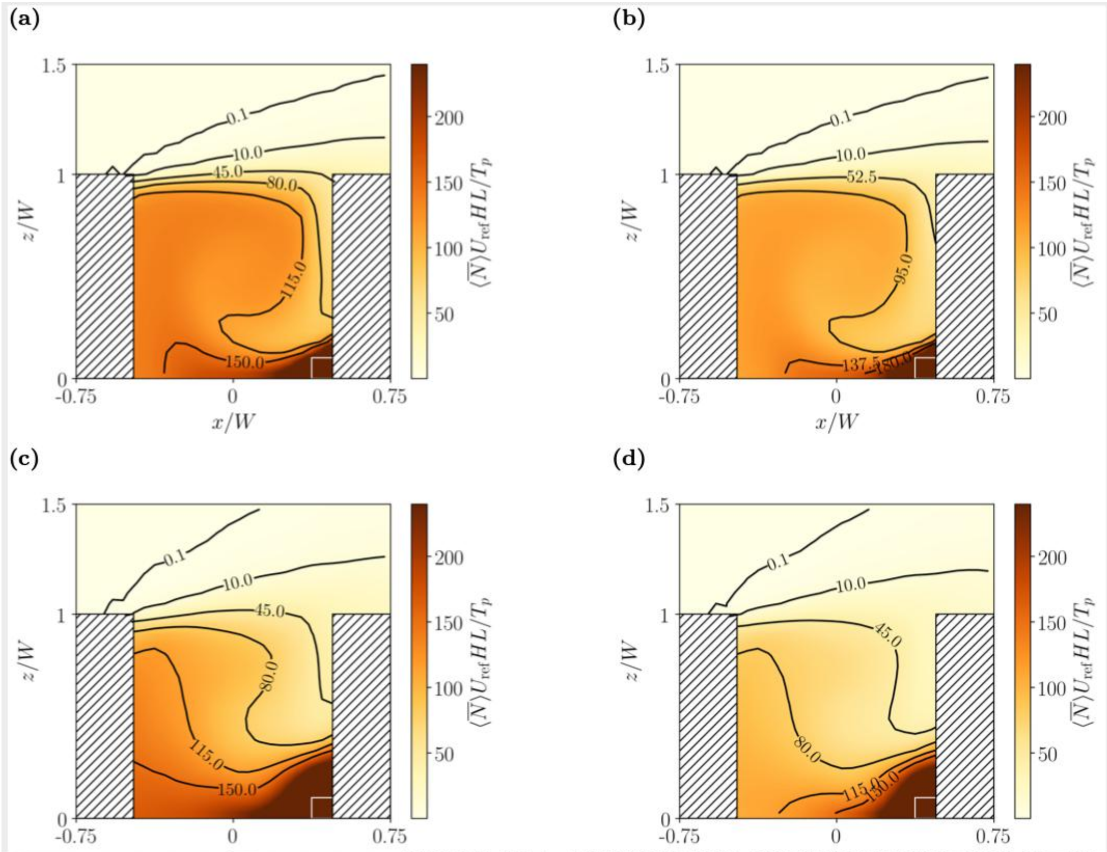

Figure 2: Comparison of results for different wind directions with respect to the axis of unit-aspect-ratio street canyon: (top) perpendicular (θ=0°); (bottom) parallel (θ=90°). (a,c) without microphysical processes; (b,d) with microphysical processes.

θ	τ_{coag}/T_c	τ_{depo}/T_c
0°	3141	0.4
90°	1529	0.5

Table 1: Comparison of timescales for θ=0° and 90°.

General Comment

Comment

This technical note deals with the spatial distribution of particles emitted from road traffic and from cooking sources in different vertical levels inside a street canyon. The PALM model coupled with the sectional aerosol dynamics model SALSA is used to determine the relevance of aerosol processes in a similar configuration as in Kurppa et al. (2019). From the analysis of time scales it is concluded that deposition mainly affects particles in the air close to the canyon surfaces, while the relevance of coagulation is related to the mean tracer age. Compared to Kurppa et al.

(2019) the novelty of the present work appears to be the consideration of particles emission spectra from kitchen exhaust ducts, which have a higher fraction of small particles than emission spectra from traffic.

The presentation of the Methodology part should be better organized, in particular with a separate section for the emission scenarios. The emission scenarios need to be in one place early in the method section because all the result sections are referring to the scenario abbreviations. The validation section is confusing.

Response: The emission scenarios are now defined in a separate subsection, Sec. 2.2.3, that precedes the results. We have confirmed that the scenario abbreviations are not referred to prior to this section.

Changes to the validation section are described under 'Specific Comments'.

Comment

My main concern is that the time scale of mean circulation, i.e. residence time in the street canyon, of 380 s is very long in comparison to other published studies on street canyons. For example, Nikolova et al. (2014) report CFD simulations of aerosol particles for a real street canyon in Antwerp having unit aspect ratio and a dilution time scale of 110 s for low wind. Ketzel and Berkowicz (2004) give dilution time scales within a range of 45−120 s. The long recirculation cycle does not seem realistic even at low winds, hence leading to an overestimation of the contribution of coagulation to the reduction of mean particle number concentrations compared to the case with no aerosol processes.

Response: The dilution timescale (Ketzel and Berkowicz 2004) differs from the mean circulation timescale calculated in the ms. In the version adopted by Nikolova et al. (2014), $T_{\mathrm{di}}=10H/u_{roof}$, where u_{roof} is the RMS streamwise velocity at the roof level. By contrast, we use $T_c= 2(\frac{W}{Urms}+\frac{U}{Wrms})$, where u_{rms} and w_{rms} are the RMS streamwise and vertical velocity over the entire canyon. Since it is defined using velocity data at the roof level, T_{di} is a local timescale: the contributions of velocity fluctuations inside the canyon, and the influence of the topography, are essentially ignored. While T_{di} may be more suitable for other applications, T_c is a natural choice for a study focused on aerosol dynamics within the canyon. As noted above, there is a close connection between Tc and the mean age of air. Nevertheless, we emphasize that the differences are a consequence of the definitions. Using the definition of Nikolova et al. (2014), we calculate $T_{\mathrm{di}} \sim 60$ s, which falls within the range quoted by the referee. There is no reason to believe that the values of T_c, which are consistent with previous studies (e.g. Lo and Ngan 2017), are indicative of an unrealistic or unphysical flow regime.

Comment

The formation of a stable vortex holds for neutral conditions, but it needs to be tested what consequences unstable conditions with thermal convection have on the concentration distribution in the street canyon. Another aspect to consider is that when the wind is parallel with the street, the recirculating structure within the cavity disappears completely, and the concentration field becomes very different. The authors should state such important limitations of the presented CFD simulations early in the text. Therefore, I suggest emphasizing that an idealized configuration of the street canyon was chosen, for the purpose of the study of particle emissions from different pollutant sources inside the street canyon.

Response: As explained above, the central canyon vortex persists under unstable stratification (Fig. 1). Furthermore, the effects of aerosol dynamic processes are qualitatively similar when the external wind is parallel rather than perpendicular to the canyon axis (Fig. 2). The relative importance of deposition or coagulation will change appreciably under two conditions. First, the residence time or mean age decreases dramatically so that $\tau_{\mathrm{coag}}/T_C \lesssim 1$ or $\tau_{\mathrm{depo}}/T_C \gtrsim 1$. Second, there is very intense local turbulence so that deposition or coagulation rates may be significantly larger within certain regions or structures. Given the persistence of the central canyon vortex for unstable stratification and the relatively insensitivity of the ratios to the wind direction (Table 1), we conclude that these conditions are not satisfied for coagulation, which is the dominant process for cooking emissions. With a much shorter dynamical timescale, so that $\tau_{\mathrm{depo}}/T_C \gtrsim 1$, one expects the relative importance of coagulation to increase.

These issues are discussed in Section 6. See also our response to Referee 1.

Specific comments

1. Section 2.2.1: Provide more details on the configuration of the street canyon and mention which aspects are different to the street canyon simulated in the work of Kurppa et al. (2019).

Response: The street-canyon configuration is described in in Sec 2.2.1. The dimensions of the domain and topography are specified, as well as the grid spacing, boundary conditions, mean-flow forcing and stability. Details on the numerical schemes may be found in Sec. 2.1.1.

Differences with respect to the configuration of Kurppa et al. (2019) are now clearly stated (l.86):

> The configuration described above differs in several respects from K19. First, an idealised street canyon is used in place of realistic topography within a neighbourhood-scale domain. Second, there is uniform grid spacing rather than stretched grid. Third, the computational lid height of 5H is decreased from 13Havg, where Havg is the mean building height.

The most important difference is the substitution of a unit-aspect-ratio street canyon for realistic topography.

2. Section 2.2.2 has to be divided in a section on configuration of SALSA in this study and a section on emission scenarios.

Response: As mentioned above, a separate section on emission scenarios has been created.

3. More details on the coupling of SALSA with PALM need to be provided. For example, are the particle emissions entering into the SALSA model or first into the PALM model? Deposition of particles: only to street surface or also and wall surfaces; in which distance from the surface are particles affected by deposition? Condensation of which gases?

Response:

i. Aerosol emissions are handled by SALSA rather than PALM. Hence a particle is subjected first to aerosol processes before being transported and advected by PALM. This is now explained on l. 100:

    Pollutants are emitted from uniform area sources. Since aerosol emissions are handled by SALSA rather than PALM, pollutants are subjected to aerosol dynamic processes before being transported and advected by PALM.

ii. Deposition occurs occur on all street and wall surfaces. Figure 13 shows that the deposition velocity is maximized in their immediate vicinity (i.e. within the first grid box). In fact, the deposition velocity vanishes away from the first grid box; therefore, only gravitational settling occurs at these points. This is now explained on l. 62:

    Briefly, deposition removes particles near surfaces; the deposition velocity is non-zero within the first grid box at a surface, e.g. from $z = 0$ to $z = \Delta z$ (cf. Fig. 13a); away from the surface, only gravitational settling occurs.

iii. Condensation of H2SO4, HNO3, NH3, semi-volatile (NVOCs) and non-volatile organics (SVOCs) is included. This is now mentioned explicitly on l.71:

    Gaseous components, namely H2SO4, HNO3, NH3, semivolatile (SVOCs) and non-volatile organics (NVOCs), may condense onto particles [...]

4. Section 2.3: the presentation of the validation is unclear. Several references to figures are missing. Maybe first mention what kind of validations were performed, then describe each test in a separate paragraph.

Response:
Yes, the presentation could have been clearer. Following the referee's suggestion, several changes have been made:

i. A new paragraph summarizing the different types of validation tests has been added to beginning of the section (l. 155).

Several validation tests have been performed. First, the mean velocity statistics are validated against measurements of flow over parallel unit-aspect-ratio streets canyons (Brown et al., 2001). Second, passive scalar statistics are also validated (Pavageau and Schatzmann, 1999). Finally, the performance of the coupled PALM-SALSA model is compared to previous studies (Kumar et al., 2008; Kurppa et al., 2019).

ii. Figure references have been added where necessary.
iii. The PALM and PALM-SALSA tests are described in separate paragraphs.

5. P. 7 lines 148 - 153: explain the difference of the simplified computational domain. used in the validation and the computational domain in K19. Is the simplified computational domain intended to mimic the real street canyon in Cambridge? I think Figure 5 belongs to this validation, but it is not referenced here.

Response: Yes, the simplified computational domain is intended to mimic the real street canyon in Cambridge. The text has been modified to make this clearer (l. 149):

For simplicity, the computational domain is focused on this street canyon: no other buildings are included. In particular, a single street canyon of 167 m × 12 m × 12 m is centred inside a domain of 167 m × 60 m × 60 m.

An explicit reference to Figure 5 is now included (l. 151): "Vertical profiles of the aerosol number concentration are compared in Fig. 5."

6. P. 13 line 2: Figure 8 shows only boiling. Where is the figure panel for isolated kitchens, deep frying? Figure parts fig. 8a and 8d are not referenced in the text.

Response: We have added results for isolated kitchens and deep-frying (I-D-z0) (now Fig. S-1 of the Supplementary Material). The corresponding values have been added to Table 3.

Table 3. As in Table 2, but for deep-frying and boiling emissions from isolated kitchens.

	NOAD	AD	difference
I-B-0.05	219.3 ± 7.2	200.0 ± 5.5	-9%
I-B-0.50	289.9 ± 10.8	276.2 ± 8.7	-5%
I-B-0.95	242.5 ± 13.8	231.4 ± 11.8	-5%
I-D-0.05	219.3 ± 7.2	181.3 ± 3.0	-17%
I-D-0.50	289.9 ± 10.8	264.4 ± 4.7	-9%
I-D-0.95	242.5 ± 13.8	232.4 ± 4.3	-4%

Figs. 8a and 8d are now referred to explicitly (l. 201).

> **Although trapping of particles within the vortex at the bottom leeward corner is less evident as the source height is increased from $z_0/H = 0.05$ to $z_0/H = 0.95$ (Figs. 8a-c),[...]. The vertical profiles (Fig. 8d) show [...]**

7. P. 14 lines 219 - 222: Time scale analysis for a street canyon in Cambridge by Kumar et al. (2008) reveals that dry deposition to road surface is much faster than deposition to wall surfaces. Can such a differentiation be made in this study as well?

Response: Yes, from the deposition velocity for NG-D (Figure 13), the deposition timescale may be estimated as Δ/v_d, where Δ is the grid spacing. This shows that the deposition timescale for road and wall surfaces is ~65 s and ~110 s, respectively. In agreement with Kumar et al. 2008, deposition to the road surface is faster.

8. P.17 lines 254 – 256: The "plume-like structure" for case CO-D cannot be inferred from Figure 11. Should this refer back to Figure 7? It should be better indicated in the plot, how the plume like structure from column kitchen emissions develops.

Response: The "plume-like structure" refers to the tongue of low concentrations with RDcoag < 22% emanating from the column source. The colour bar and contour interval have been adjusted to show the plume-like structures in Figs. 11c,d more clearly. The text has also been modified to aid interpretation (l. 256):

> **however, the column source covers a larger area and a plume-like structure (i.e. the tongue of low RD values between the canyon centre and the windward wall) develops away from it.**

9. Section 3.4: it is not immediately clear where in the street canyon the aerosol number distribution were taken. If it is the canyon average distribution, then the standard deviations should be included in Figure 12. Where in the street canyon should measurements be done to be most sensitive to emissions of each of the different source types?

Response: The aerosol number distributions shown in Fig. 12 represent canyon averages. The caption has been updated to make this clear. Error bars have been added to depict the temporal standard error. The temporal standard error is chosen (rather than, for example, the spatial standard deviation) because it characterizes the error in the estimator rather than the spatial distribution. A new paragraph describing the statistical errors in the size distributions has been added (l. 270):

> **The uncertainty in the estimate of the time-averaged size distributions is indicated with the (temporal) standard error. Errors are much smaller for the deep-frying cases, NG-D and CO-D. A plausible explanation is that temporal intermittency is greater for cases in which deposition plays a more important role, namely TR, NG-B and CO-B, because deposition only occurs**

near surfaces and is maximised inside the corner vortices. Coagulation, by contrast, occurs everywhere.

The determination of appropriate measurement locations is an interesting question. We have not determined these locations precisely; however, approximate locations can be estimated from the relative difference fields of Sec. 3.3.

10. Section 5.1: the description of the background particles needs to be improved. How is the background aerosol mixed into the street canyon - is the spatial distribution the same as for the emissions or is the background entering from the boundaries? Did the simulations take into account heterogeneous coagulation between the emitted particles and the background particles, or were they assumed to be of the same population?

Response: The background concentrations for the entire domain are fixed; hence, there is no spatial distribution. Yes, heterogeneous coagulation between the emitted particles and the background particles is included: to a large extent, the results described in Sec. 5.1 concern implications of this phenomenon.

To avoid confusion, the uniform nature of the background concentrations is now noted (l. 331):

> **Spatially uniform, constant background concentrations are prescribed over the entire computational domain.**

The inclusion of heterogeneous coagulation is also noted (l. 332, l. 336):

> **Note that the background is allowed to interact with the emissions through heterogeneous coagulation.[...] On account of aerosol processes involving the background only or the background and emission [...].**

We believe that the description of the background particles should now be clear.

Technical Corrections

P. 7 lines 139: correct "ine Appendix B".

Fixed.

P. 14 line 229: deep frying?

Fixed.

P. 26 line 431: delete "happens".

Fixed.

References

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