Response to Referee 1

We thank the referee for the helpful comments. Point-by-point responses are included below. Briefly we clarify the main findings of the ms and include some additional sensitivity calculations.

General comments

Comment

> The manuscript deals with the interesting and atmospheric relevant topic of the dispersion and processing of cooking and road traffic generated aerosol particles in urban street canyons. The authors address this topic using the building-resolving computational fluid dynamics model PALM, which also include the sectional aerosol dynamics module SALSA. The model was setup for an hypothetical and simplified street canyon with road traffic emissions or cooking generated aerosol emissions from the surrounding buildings. The authors address how the type, location and aerosol dynamics of the emissions influence the concentration in the street canyon.
>
> Apart from a few typos and minor grammatical errors I think the manuscript is generally well written. I agree with reviewer 2 that it could benefit from some restructuring. Also consider if you need all 16 figures. After careful revision I think the manuscript has the potential to be publishing as an atmospheric relevant "technical note" in atmospheric chemistry and physics.

Response: Yes, we agree that there were too many figures. We have moved two figures from Appendix E to Supplementary Material. New figures and tables have also been moved to Supplementary Material.

Comment

> What I mainly miss with the manuscript is a more careful motivation to the choice of the simplified (idealized) street canyon, the primary particle emission size distributions from the different emission sources, the meteorological conditions and the location of the cooking emissions, especially since the model is compared against real observations of wind velocity profiles from a wind-tunnel study (Figure 4), and observations from a specific street canyon in Cambridge (Figure 5).

Response: Yes, this is a good point. In any numerical study of urban pollutant dispersion, the representativeness of the geometry, meteorological conditions and source specification is always an issue. The choices made in this manuscript are generic ones of wide applicability:

> The street canyon is recognised as the basic geometric unit of the built environment (Oke 1988). A unit aspect ratio is a canonical choice because it mimics the effect of deep urban canopies, where relatively poor ventilation and strong pollutant trapping occur, though of course the precise flow details differ. The simplicity of the street canyon geometry makes it especially suitable for investigating the effects of physical processes such as chemistry (e.g. Zhong et al. 2015).

The assumption of constant density and a wind perpendicular to the canyon axis is another standard choice that is of great relevance to urban air quality. Ventilation improves for unstable conditions and is largely unchanged for stable stratification (Duan & Ngan 2019; see also our reply to Referee 2). With respect to the wind direction, a perpendicular external wind leads to the occurrence of a canyon vortex and strong pollutant trapping.

The source locations and emission spectra are also intended to be generic choices. Just as with vehicular emissions, in which moving discrete sources are represented by a line or area source, the near-ground, isolated and column cooking sources are meant to be plausible idealisations. A systematic derivation lies far beyond the scope of this study: indeed, we are unaware of a comparable analysis for vehicular emissions. The emission spectra are taken from well-known studies.

The generic nature of these choices is now mentioned in the Introduction (l. 45):

After reviewing the methodology (Sect. 2), results are presented for several idealised but generic emission scenarios, e.g. traffic, deep frying and cooking emissions (Sect. 3).

We also argue in the Discussion that these choices do not affect one of the key findings of this study, which is that the nature of the aerosol dynamics is largely determined by the ratio of the coagulation and deposition timescales to the dynamical timescale. Of course, fine details of the aerosol dynamics necessarily depend on the choices described above; however, the relative importance of coagulation and deposition is largely insensitive to most of these choices. In the case of cooking emissions, similar behaviour is obtained so long as the coagulation timescale (τ_{coag}) remains long compared to the deposition (τ_{depo}) and dynamical timescales. The dynamical timescale, which may be taken to be the mean canyon circulation timescale (T_c) or the mean tracer age (MTA), characterises the time required for a pollutant to escape from the canyon or the amount of time elapsed since the pollutant was released. This argument is closely related to that of Harrison 2018, who showed that the dynamics of gas phase pollutants depends on the ratio of the chemical timescale to the dynamical timescale (residence time). Roughly speaking, the extent of chemical transformation within the canopy depends on the timescale over which pollutants are allowed to react. In the present case, the relevant dynamical timescale for coagulation is the total time elapsed since a particle was emitted. Small changes in τ_{depo} or τ_{coag}, which inevitably accompany modifications to the configuration, should not have a significant effect on the aerosol dynamics.

We have added a new table (Table 5) that summarises these timescales for the different emission scenarios.

Table 5. Dynamical and aerosol timescales for different emission scenarios.

Source location	T_c (s)	MTA (s)	Emission scenario	τ_{depo} (s)	τ_{coag} (s)
TR	382	584	TR	150	1.1×10^7
NG	382	652	NG-D	150	1.8×10^5
			NG-B	150	1.2×10^6
CO	382	599	CO-D	150	2.2×10^5
			CO-B	150	1.8×10^6

In all cases, τ_{depo}/MTA <1 and τ_{coag}/MTA >>1. We also discuss how the ratio of these timescales may change for different configurations (Sec. 6, l.372):

> **With other emission scenarios or flows, quantitative differences are unavoidable, but qualitative differences in the aerosol dynamics are not expected in most cases. For cooking emissions, the coagulation timescale is much longer than the relevant dynamical timescale (Table 5), which implies that coagulation will continue to be controlled by the ageing of fluid parcels or the mean tracer age. The dynamical timescales change with the wind direction (Supplementary Material, Table S-1), but the coagulation timescale, τ_{coag}, remains much longer. For stratified flow, the MTA will decrease for unstable stratification and increase for stable stratification but the effect should be relatively small (see Duan and Ngan (2019) for building array results). The situation is more complicated for deposition insofar as τ_{depo} is not much less than the relevant dynamical timescale, i.e., the canyon circulation timescale T_c. Qualitatively different behaviour is expected only for a much smaller T_c, such as may occur for unstable flow or a street canyon with lateral openings. In this case, deposition will be less spatially localised and will no longer proceed to completion. For cooking emissions, the relative contribution of deposition would therefore decrease compared to the cases examined in this paper, for which τ_{depo}/T_c < 1.**

It is likely that the coagulation timescale will remain long in most cases. The deposition timescale may not be short relative to the dynamical timescale for certain cases, e.g. convectively unstable flows or a finite street canyon, but this should serve to reinforce the importance of coagulation for cooking emissions.

Comment

No very much specific information is given about how the aerosol dynamics is represented in the model in the current study. Only the primary particle emissions are described, with some details. Especially I miss information about how the condensation of different vapors were treated in the model. E.g. what properties were assigned to the semi-volatile condensable vapors HNO3, NH3 and SVOCs.and how do you calculate their volatility with respect to the aerosol particle phase? For HNO3, NH3 is should depend on the aerosol liquid water content and acidity

Response: The treatment of condensation is described in Appendix A. Since chemical transformations are not considered in this study, we focus on the equilibrium saturation ratio and saturation vapour mole concentration. Other properties, such as the volatility with respect to the aerosol particle phase, are therefore excluded.

Specific comments

Abstract, I miss one sentence which motivate why this study is important from an atmospheric chemistry and physics perspective.

Response: A new sentence summarising the points made in response to the general comments has been added to end of the Abstract:

It is argued that the qualitative nature of the aerosol dynamics within urban canopies is determined by the ratio of the aerosol timescales to the relevant dynamical timescale (e.g. the mean age of air).

L24-26, "Deposition is usually the only aerosol process included in urban CFD models as it is the most important for traffic emissions within street canyons (Kumar et al., 2011)." The reference to this statement is a bit old, is this statement still true? Also consider to replace "as it is the most important" with as it is often assumed to be the most important loss process of ultrafine particles. If you do not consider other process you cannot judge their importance. Hear you also only refer to loss processes and not formation processes such as atmospheric new particle formation which can be a major source of ultrafine particles also in urban environments.

Response: The reference is quite old, but the statement is still true. Karl et al 2016 also find that the dry deposition is the most important aerosol process in urban environments (with a relative difference in particle number of ~15%). Furthermore, more recent numerical studies, e.g. Kim et al 2019, only include deposition. We have modified the text and added a reference (l. 25).

[...] as it is often taken to be the most important loss process of ultrafine particles emitted by [...]

Line 35-36, "There are strong reasons for expecting the dispersion of traffic-generated and cooking-generated aerosols to differ qualitatively." Consider to reformulate this sentence.

Response: The text has been rephrased as follows (l. 35): "The dispersion of cooking- and traffic-generated aerosols differ in two key respects."

L37, "diameter of O(10 nm)" What do you mean with O?

Response: O stands for "order of". This is the so-called Big O notation which is commonly used in numerical modelling. The sentence in question notes that cooking emissions contain a higher proportion of particles with a diameter of around 10 nm.

> L44 "The effects on the aerosol dynamic processes are highlighted" Do you mean the effects of the aerosol dynamic processes are highlighted ?

Response: No, we did indeed mean to refer to the effects of the emission scenarios *on* the aerosol dynamic processes. The analysis in Sec. 3.3, for example, focuses on how the effects of coagulation and deposition depend on the emission scenario. While the effects *of* the aerosol dynamic processes are obviously related, the emphasis is somewhat different. Nevertheless, the sentence has been deleted for brevity.

> L56-57, "the inclusion of transient dynamics allows for nonlinear aerosol processes to be represented more accurately (see Sec. 5.2)." What do you mean with this statement?

Response: Our intention here was to draw a distinction between steady (RANS) and unsteady (LES) calculations. Even if one is interested only in the time average, the neglect of temporal fluctuations is problematical when there are nonlinear terms because the time average of products of the fluctuations does not vanish. The same argument lies behind all turbulence models. In the present case, the modelling of nonlinear aerosol processes such as coagulation will be less accurate with steady RANS. This is now explained on l.56:

> **With a steady model, temporal fluctuations are neglected, thereby necessitating a turbulence parameterisation for the aerosol dynamics.**

> L60-61, "Nucleation, which is computationally expensive to simulate, is not considered in this work." In which way do you mean that nucleation is computationally expensive to simulate? Usually nucleation is parameterized as a rate only depending on e.g. the H2SO4 concentration, or H2SO4 and NH3. The concentrations of these vapors you anyway have to calculate in the model for the condensation growth.

Response: Nucleation is computationally expensive because it occurs on a short timescale. According to Rönkkö et al., the nucleation timescale in the exhaust of a vehicle is $\tau_{\text{nucl}} \sim 0.7$s. To resolve the process accurately, a smaller timestep (and a finer grid) is required; Ketzel and Berkowicz noted that it's simpler to represent nucleation by modifying the representation of the source. Nonetheless, the computational cost is not necessarily prohibitive: Kurppa et al. (2019) noted that the computational cost of nucleation is comparable to that of deposition. We have therefore modified the wording (l.60):

> **Nucleation, which is most relevant in the immediate vicinity of the source (Rönkkö et al., 2007) and can be treated by modifying the emissions (Ketzel and Berkowicz, 2004), is not considered in this work.**

Thank you for bringing this point to our attention.

> L71-72, "semi-volatile (NVOCs) and non-volatile organics (SVOCs)". It should be semivolatile (SVOCs) and non-volatile organics (NVOCs)

Response: Fixed.

> L73, "however, chemical transformations are excluded." What exactly do you mean with this statement? Did you not consider any gas-phase chemistry at all? If this is the case, please state this clearly.

Response: Yes, we simply meant to say that all gas phase chemical reactions are excluded. This is now stated unambiguously in the revised text (l. 72): "[...] gas phase chemical reactions are excluded."

> L83, "The flow is driven by an external pressure gradient, dp/dx = -0.0006 Pa m−1." I cannot judge if this is a reasonable value. Can you add some information about typical values and a reference?

Response: This value has been used in many previous studies (e.g. Duan et al 2019). Using this value, the streamwise velocity at z/H = 2.5 is U~3 m/s. This is now explained on l.83:

> **[...] This value has been widely used in previous CFD studies (e.g. Duan et al., 2019); it yields a streamwise velocity U $\sim$ 3ms^{-1} 90 at z/H = 2.5. [...]**

> L109-110, "The emission factor for the number of particles emitted by a vehicle per unit distance travelled is 3.0×10^{14} km−1 veh−1 (Fujitani et al., 2020)" This, cannot always be a fixed value. At least replace "is" with e.g. "was estimated to be".

Response: Yes, the recommended text is now used.

> L115-116, "The emission factors for the number of particles emitted per unit time by a kitchen of unit volume are 3.75×10^{10}m−3 s−1 and 4.31×10^{9}m−3 s−1, for deep frying and boiling, respectively." Replace "are" with e.g. "were estimated to be".

Response: Yes, "were estimated to be" is now used.

> Figure 3. The selected traffic emission spectrum from Janhäll et al., 2004 is relatively old. Is this still representative for the more modern car fleet today? I imagine that the fraction of nucleation mode particles may have gone up while the soot mode may have decreased with more modern cars? But, I may be wrong. Can you find any more recent references to at least compare

with? Quite a lot of your conclusions are based on the selected size distributions of traffic, deep-frying and boiling emission size distributions.

Response: Yes, this is a good point. We have checked some more recent references. The number distribution measured by Schneider et al. 2015 is broadly consistent with the spectrum of Janhäll et al., 2004, as may be seen in Figure 1.
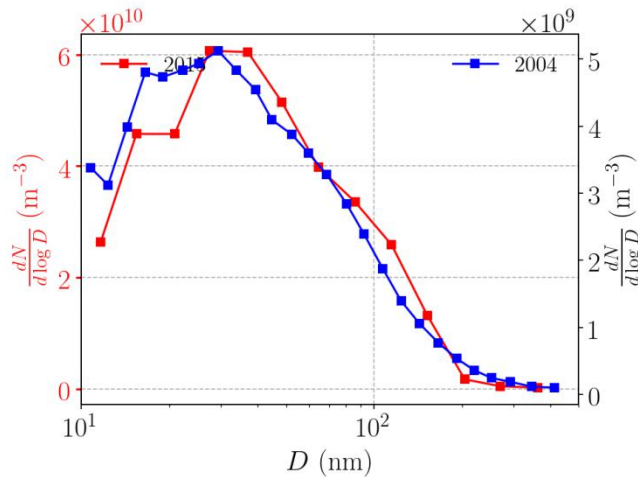

Figure 1: Comparison of size spectra measured within street canyons. Data are taken from Schneider et al. 2015 (red) and Janhäll et al., 2004 (blue).

Indeed, the mean particle diameters (i.e. 47.5 nm and 47.9 nm) agree well.

Line 148-149, "Following K19, the coupled PALM-SALSA model is validated against evening measurements of the aerosol number concentration within a real street canyon in Cambridge, UK (Kumar et al., 2008). Can you really evaluate your model results against these observations? How similar are the Cambridge street canyon compared to your idealized street canyon. How does the meteorological conditions during the measurements agree with the neutral conditions with the temperature fixed at 300 K?

Response: Yes, we believe that it is fair to compare our model results to the observations. The street canyon geometry in our model (W=H=12 m, L = 167 m) is essentially identical to that of the real one (W=11.75m, H=11.6 m, L = 167 m). The emissions along the street canyon and background concentrations are identical to those in Kurppa et al. 2019. The main difference is that our domain is much smaller as we exclude the buildings surrounding the street canyon where the measurements were taken.

For consistency with the evening measurements, the temperature was fixed at 274 K for the validation only; the value of 300 K was used for the results proper. This is now explained on l. 149:

For consistency with the evening measurements, the temperature is fixed at 274 K.

L151, "only are considered." Change to are only considered.

Response: We believe that the original wording, "Using the traffic data in K19, emissions from the street canyon only are considered", more accurately conveys the intended meaning, which is that emissions from neighbouring streets are excluded.

Figure 5, I miss a describing text and reference to Fig. 5 in the manuscript

Response: Yes, Fig. 5 is now referred to on l. 151:

[...] Vertical profiles of the aerosol number concentration are compared in Fig. 5. [...]

L169-170 "Deep frying (NG-D) and boiling (NG-B) yield identical concentrations in the absence of aerosol dynamic processes. Please explain why this is the case. E.g. Deep frying (NG-D) and boiling (NG-B) yield identical concentrations in the absence of aerosol dynamic processes because the location of the emission sources are identical.

Response: When aerosol dynamical processes are excluded, the aerosol evolution for NG-D and NG-B is completely passive. The normalised concentrations (eq. 1) are identical because differences arising from the emission strengths are eliminated. This is now explained on l. 169:

Deep frying (NG-D) and boiling (NG-B) yield identical normalised concentrations in the absence of aerosol dynamic processes.

L175-176 "One possible explanation for this discrepancy is that the emission spectra differ: the mean particle size is larger in the current study, i.e. 47.9 nm rather than 32.7 nm." This again makes me wonder about how representative the selected traffic emission spectrum is.

Response: As discussed above (see Figure 1), the traffic emission spectrum of Janhäll et al., 2004 does not appear to be inconsistent with more recent measurements. Nevertheless, we agree that representativeness of the emission spectrum is an important issue for this (or any other study of aerosols in the outdoor environment).

To investigate this issue, we have carried out sensitivity calculations for emission scenario NG-B:
- Displacement to large scales (LD): the particle diameter of each size bin is doubled
- Displacement to small scales (SD): the particle diameter of each size bin is halved

Vertical profiles of the mean number concentration are similar for LD and SD (Figure 2). Moreover, the relative difference fields show a similar spatial structure (Figure 3). We therefore conclude that the representativeness of the emission spectrum for boiling emissions should not be a serious issue. Similar behaviour may be expected for deep frying.

These results are now referred to in the Discussion:

Although the results inevitably depend on the emission spectrum — mean concentrations for boiling and deep frying differ by ∼ 30% for near-ground emissions and ∼ 15% for column emissions (Table 2) — there is no evidence for strong sensitivity. Test calculations in which the emission spectrum for NG-B is scaled by a factor of 2 or 0.5 show limited sensitivity. For example, the vertical profiles show a nearly identical shape with mean concentrations differing by less than 5% with respect to the default emission spectrum (Supplementary Material, Fig. S-4) [here Fig. 2]. Furthermore, the spatial structure of the relative difference fields is almost identical (Supplementary Material, Fig. S-5) [here Fig. 3].
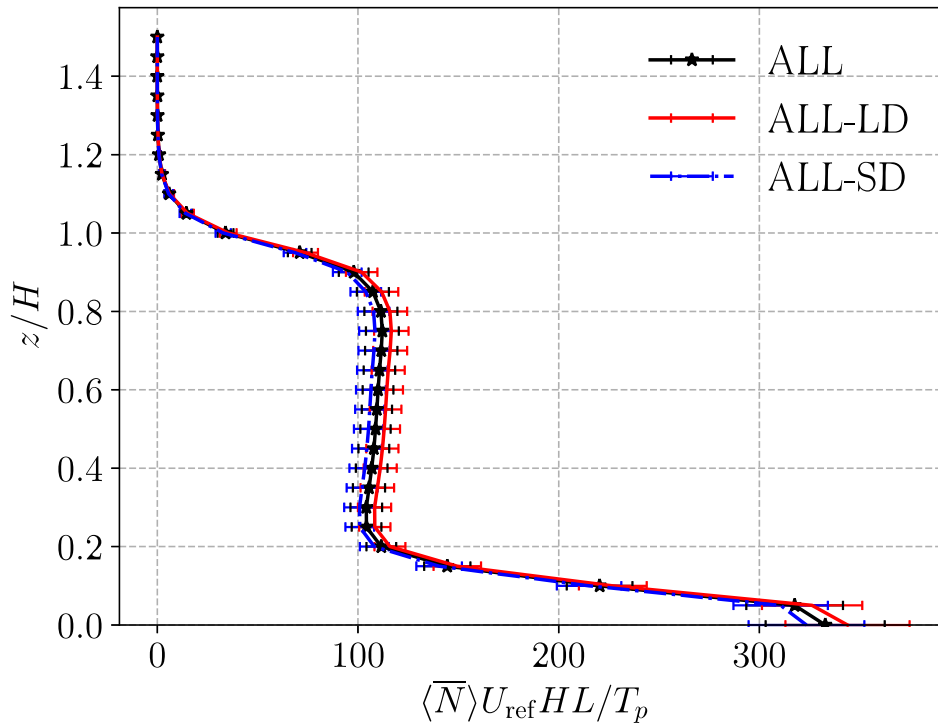

Figure 2: Vertical profiles of the mean number concentration for emission scenario NG-B and all aerosol processes. The vertical profiles correspond to the default emission spectrum (ALL), displacement to large scales (ALL-LD) and displacement to small scales (ALL-SD).
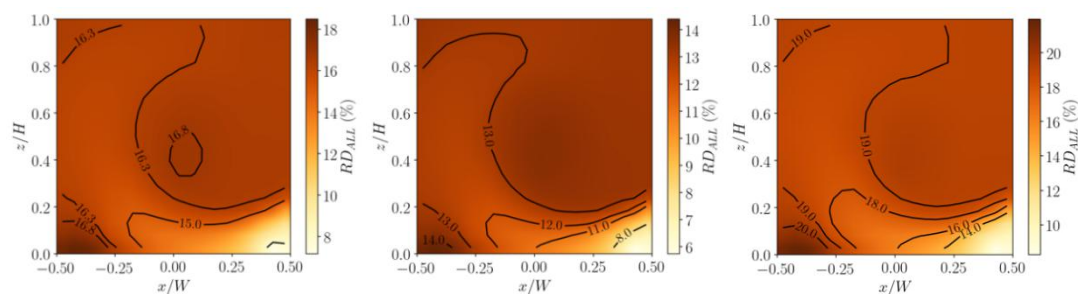
Figure 3: Relative difference fields for NG-B: (left) default emission spectrum; (middle) displacement to large scales, LD; (right) displacement to small scales, SD.

L176-177 "This is significant because smaller particles may have a larger deposition velocity" When you refer to small particles I think you mean submircon particles < 1000 nm in diameter. In this, case are not small particles (e.g. ultrafine particles) always having greater deposition velocities than larger >100 nm diameter particles?

Response: Actually, by smaller particles we mean particles whose mean diameter is less than 100 nm. For the deposition parameterisation of SALSA, the deposition velocity increases monotonically for $D < 100$ nm (Kurppa et al. 2019, Figure 1). Clarification has been added to l.176: "[...] because smaller particles (with a diameter less than 100 nm; K19) may have [...]". For urban surfaces, the deposition velocity is usually (but not necessarily) smaller.

L216, "Condensation has a negligible effect ..." Does this not also depend on the model assumptions/limitations? Also evaporation of semi-volatile species from the fresh exhaust particles could potentially have large influence on the particle number size distribution, especially at the selected high temperature of 300 K. Some recent studies claim that particles can grow very rapidly by nitric acid an ammonia condensation, see e.g:

Wang, M., Kong, W., Marten, R. et al. Rapid growth of new atmospheric particles by nitric acid and ammonia condensation. Nature 581, 184–189 (2020). https://doi.org/10.1038/s41586-020-2270-4.
Could the importance of such claimed rapid growth phenomenon be studied and verified or dismissed using PALM-SALSA?

Response: Yes, it's conceivable that condensation could have a greater effect under certain conditions, but given that its primary effect is to increase particle volume, one may expect the effect on the number concentration to be weak in most cases. Nevertheless, the wording has been qualified (l. 216):

> **Condensation has a negligible effect on the aerosol number concentration, which is consistent with the notion that it primarily serves to increase the volume of particles**

We agree that, under the right conditions, evaporation could have a noticeable effect on the particle number size distribution. But given that numerical studies of aerosols

in the urban environment usually include deposition only, we follow Kurppa et al. (2019) and consider deposition, condensation and nucleation only. Inclusion of evaporation would require a very small timestep.

In theory, PALM-SALSA could be used to investigate the occurrence of the rapid growth phenomenon described in the Nature study; however, the computational cost could be quite high as PALM-SALSA is designed for large scales (e.g. neighbourhoods rather than reaction chambers) and longer timescales. Furthermore, a large number of size bins may be needed to resolve the increase in the particle size.

L275 "O" What do you mean?

Response: As explained in the comment to l. 37, O refers to the Big O notation.

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